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Airborne observations of aerosol optical properties of mineral dust mixtures in the eastern Mediterranean

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Abstract

Atmospheric aerosol particles, from both natural and anthropogenic sources, interact with solar radiation and clouds, affecting the Earth's radiation budget and contributing to uncertainties in climate projections. Mineral dust, an abundant aerosol worldwide, originates from wind erosion of deserts, forms elevated layers, and travels long distances, mixing with anthropogenic aerosols, like black carbon and sulfate aerosols. This mixing may influence the optical and radiative properties of these aerosol layers. Studies on mineral dust-pollution mixtures have shown varying effects depending on the region and aerosol composition, highlighting the need for further investigation.

This thesis focuses on assessing the effects of mixing mineral dust and anthropogenic aerosols ("pollution") using techniques that measure the aerosol optical properties. Through laboratory experiments, model simulations, and aircraft in situ observations, this thesis (1) evaluates uncertainties in the particle scattering coefficient of mineral dust from a polar integrating nephelometer, and (2) establishes guidelines for its accurate determination depending on aerosol type and investigated particle size range. It (3) quantifies the impact of pollution on the intensive optical properties of mineral dust from the Arabian Peninsula and the Sahara desert, (4) examines how well mineral dust events can be identified in polluted conditions using intensive aerosol optical properties, and (5) assesses the change in the direct radiative effect efficiency of mineral dust layers for increasing pollution.

This thesis is based on two core instruments: an Aurora 4000 Polar Integrating Nephelometer (Aurora 4000 nephelometer, Ecotech - now Acoem), and a Tri-color Absorption Photometer (TAP, Brechtel), providing the particle scattering and absorption coefficients, and the angular scattering distribution, which are essential properties for better understanding the aerosol's direct radiative effect. To support the analysis, measurements of aerosol microphysical properties and optical simulations were also performed.

This thesis involved three primary experiments: a laboratory closure experiment, a simulated closure experiment, and observations from an aircraft field experiment in the eastern Mediterranean. The laboratory experiment measured laboratory-generated aerosols with extensive instrumentation. The simulated experiment used numerical simulations of instrumental responses to support the interpretation of laboratory findings. A comprehensive data set was collected aboard the research aircraft Falcon during the Absorbing aerosol layers in a changing climate: aging, lifetime and dynamics (A-LIFE) aircraft field experiment which aimed to investigate the properties of absorbing aerosol layers in the eastern Mediterranean, a region influenced by mineral dust from the Arabian Peninsula and the Sahara desert, and pollution.

Quantifying uncertainties of direct in situ measurements of the particle scattering coefficient

The largest uncertainty in particle scattering coefficient measurements from integrating nephelometers arises from the "angular correction", which primarily compensates for light scattering truncation at outermost angles (i.e., 0° and 180°).

Through laboratory and simulated closure experiments, this thesis evaluated different existing methods to derive the nephelometer angular correction for coarse, irregularly shaped particles such as mineral dust particles. The irregular particle shape has a minimal effect on nephelometer angular truncation, and assuming sphericity to calculate the angular correction for mineral dust particles results in only 2% underestimation of the scattering coefficient. However, the non-sphericity may have a greater influence on the input variables of the various methods for calculating the angular correction. The particle size from optical particle sizers is influenced by the non-spherical shape, leading to an average overestimation of the particle scattering coefficient of 6%, and up to 22% for mineral dust, but this error may be offset by an inaccurate refractive index. This finding has important implications for optical closure studies, where accounting for particle shape effects in size distribution retrievals could improve the accuracy of retrieved refractive indices.

The conclusion of this thesis is that there is generally no "best" angular correction, but depending on aerosol type and the investigated particle size range different angular corrections should be used. For mineral dust, the angular correction based on the scattering Ångström exponent should be used, because it is least affected by the particle shape. The associated uncertainty is less than 13% in this case.

Assessing effects of mixing Arabian and Saharan dust with pollution

Using airborne in situ observations from the A-LIFE field experiment in the eastern Mediterranean, this thesis provides insight into the effect of increasing pollution content in mineral dust layers.

This thesis shows that the intensive aerosol optical properties significantly change as the pollution content increases, while differences between Arabian and Saharan dust source regions are mostly not statistically significant. This finding highlights the need for caution when retrieving the properties of pure mineral dust, and the importance of comparing measurements with similar level of pollution.

Intensive aerosol optical properties, which are important for aerosol type identification, i.e., the Ångström exponents of scattering, absorption and single-scattering albedo, change from values associated with mineral dust towards values often associated with anthropogenic aerosols as the pollution content increases. This result indicates that the Ångström exponents should not be used as sole indicators to identify mineral dust events. The Ångström exponents can help quantify the pollution, once the presence of mineral dust has been established. The scattering Ångström exponents seem to be the most appropriate, as 72% of its variability is explained by the pollution contribution in terms of mass.

The single-scattering albedo and asymmetry parameter, which are key properties for understanding the aerosol direct radiative effect, decrease significantly from pure to polluted mineral

dust layers, e.g., at a wavelength of 525 nm, median values decrease from 0.96 to 0.89 and from 0.67 to 0.56, respectively. The changes in both parameters may have opposing effects on the shortwave direct radiative effect efficiency, potentially cancelling each other out. However, the change depends on the surface albedo, and therefore, the shortwave direct radiative effect efficiency can be significantly influenced by the presence of pollution.

In conclusion, this thesis reduces uncertainties in the direct in situ measurements of the scattering coefficient of mineral dust particles. It systematically investigates mixtures of mineral dust and pollution on the basis of comprehensive airborne observations, and highlights the importance of quantifying the pollution content for assessing the direct radiative effect of mineral dust mixtures.

Kurzzusammenfassung

Atmosphärische Aerosolpartikel aus natürlichen und anthropogenen Quellen interagieren mit der Sonnenstrahlung und Wolken, beeinflussen den Strahlungshaushalt der Erde und tragen zu Unsicherheiten von Klimavorhersagen bei. Mineralstaub, ein weltweit häufiges Aerosol, wird durch Winderosion in Wüsten emittiert und weite Strecken transportiert, auf denen es sich mit anthropogenen Aerosolen wie Ruß oder Sulfataerosol vermischt. Diese Vermischung kann die optischen und Strahlungseigenschaften dieser Aerosolschichten beeinflussen. Studien über Mischungen aus Mineralstaub und anthropogenen Aerosolen zeigen je nach Ort und Aerosolzusammensetzung sich widersprechende Ergebnisse, die weiteren Forschungsbedarf notwendig machen.

In dieser Dissertation wird der Einfluss von anthropogenen Aerosolpartikeln („Verschmutzung“) auf die Eigenschaften von Mineralstaubmischungen mithilfe von Messungen der optischen Aerosoleigenschaften untersucht. Anhand von Laborexperimenten, Modellsimulationen und in-situ Beobachtungen mit Flugzeugen werden in dieser Dissertation (1) Messunsicherheiten des Partikelstreukoeffizienten von Mineralstaub bei einem polaren integrierenden Nephelometer quantifiziert und (2) Korrektorempfehlungen für eine genauere Bestimmung des Partikelstreukoeffizienten in Abhängigkeit vom Aerosoltyp und dem untersuchten Partikelgrößenbereich abgeleitet. Weiters (3) wird der Einfluss von Verschmutzung auf die intensiven optischen Eigenschaften von Saharastaub und arabischem Staub quantifiziert. Zudem (4) wird untersucht, wie gut Mineralstaubmischungen anhand von intensiven optischen Aerosoleigenschaften detektiert werden können, und (5) es wird abgeschätzt, wie zunehmende Verschmutzung von Mineralstaubschichten deren Effizienz hinsichtlich der direkten Strahlungswirkung verändert.

Zwei Messinstrumente sind von zentraler Bedeutung in dieser Dissertation: ein Aurora 4000 Polar Integrating Nephelometer (Aurora 4000 Nephelometer, Ecotech - jetzt Acoem), das den Partikelstreukoeffizienten und die winkelabhängige Streufunktion detektiert, sowie ein Tri-color Absorption Photometer (TAP, Brechtel) zur Bestimmung des Absorptionskoeffizienten. Zudem wurden Messungen der mikrophysikalischen Partikeleigenschaften und optische Simulationen durchgeführt.

Im Rahmen dieser Arbeit wurden drei Arten von Experimenten durchgeführt: ein Laborexperiment bei dem verschiedene Partikeltypen mit einer umfassenden Instrumentierung im Detail untersucht wurden, ein Simulationsexperiment zur Interpretation der Laborergebnisse und ein Flugzeugexperiment im östlichen Mittelmeerraum. Dabei wurden umfangreiche Daten an Bord des DLR-Forschungsflugzeugs Falcon im Rahmen der „Absorbing aerosol layers in a changing climate: ageing, lifetime and dynamics“ (A-LIFE) Mission erhoben. Die A-LIFE Daten bildeten die Basis für die Untersuchung des Einflusses von Verschmutzung auf die Eigenschaften von Mineralstaubmischungen im Rahmen dieser Dissertation.

Quantifizierung der Unsicherheiten bei direkten in-situ-Messungen des Partikelstreuoeffizienten

Die größte Unsicherheit bei der Messung des Partikelstreuoeffizienten mit integrierenden Nephelometern entsteht dadurch, dass Nephelometer das gestreute Licht bei sehr kleinen und sehr großen Streuwinkeln (d. h. 0° und 180°) nicht vollständig erfassen. Daher muss eine sogenannte „Winkelkorrektur“ („angular correction“) auf die Daten angewendet werden.

Anhand von Labor- und Simulationsexperimenten wurden in dieser Dissertation verschiedene bestehende Methoden zur Ableitung der Nephelometer-Winkelkorrektur für grobe ($> 1 \mu\text{m}$) nicht-kugelförmige Partikel wie Mineralstaub untersucht. Die asphärische Partikelform hat nur minimale Auswirkungen auf den Wert der Nephelometer-Winkelkorrektur. Die Annahme von kugelförmigen Partikeln bei der Berechnung der Winkelkorrektur für Mineralstaubpartikel führt zu einer Unterschätzung des Streukoeffizienten von nur 2%. Die Nichtkugelförmigkeit kann jedoch einen größeren Einfluss auf Eingangsgrößen von verschiedenen Methoden zur Berechnung der Winkelkorrektur haben. Die von optischen Partikelspektrometern gemessene Partikelgröße wird durch die nicht-sphärische Form beeinflusst, was zu einer durchschnittlichen Überschätzung des Partikelstreuoeffizienten von 6% und bis zu 22% für Mineralstaub führt. Unter bestimmten Bedingungen wird dieser Fehler durch einen ungenauen Brechungsindex kompensiert. Dieses Ergebnis hat wichtige Auswirkungen auf optische Schließungsexperimente, bei denen die Berücksichtigung von Partikelformeffekten bei der Bestimmung der Größenverteilung die Genauigkeit der ermittelten Brechungsindizes verbessern könnte.

Der Vergleich verschiedener Winkelkorrekturen im Rahmen dieser Dissertation zeigt, dass es zwar im Allgemeinen keine „beste“ Winkelkorrektur gibt, aber je nach Aerosoltyp und Partikelgrößenbereich unterschiedliche Korrekturen verwendet werden sollten. Für Mineralstaubmessungen wird die auf dem Ångström-Exponenten der Streuung basierende Winkelkorrektur empfohlen, da sie am wenigsten von der Partikelform abhängig ist. Die damit verbundenen Unsicherheiten betragen bis zu 13%.

Bewertung der Auswirkungen der Vermischung von Mineralstaub aus der Sahara und dem arabischen Raum mit Verschmutzung

Anhand von flugzeug-getragenen in-situ Messungen im Rahmen des A-LIFE-Feldexperiments im östlichen Mittelmeerraum gibt diese Dissertation Einblick in die Auswirkung unterschiedlicher Verschmutzungsanteile in Mineralstaubschichten.

Diese Dissertation zeigt, dass sich alle intensiven optischen Eigenschaften von Aerosolpartikeln mit zunehmendem Verschmutzungsanteil signifikant verändern, während die Unterschiede zwischen verschiedenen Staubquellenregionen (Sahara und arabischer Raum) meist nicht statistisch signifikant sind. Dieses Ergebnis unterstreicht, dass bei der Ermittlung der Mineralstaubeigenschaften der Verschmutzungsanteil beachtet werden muss und nur Messungen mit ähnlichem Verschmutzungsanteil für Vergleiche herangezogen werden sollten.

Intensive optische Eigenschaften von Aerosolpartikeln, die für die Identifizierung des Aerosoltyps wichtig sind wie z.B. Ångström-Exponenten der Streuung, der Absorption und der Einfachstreuungsalbedo, ändern sich mit zunehmendem Verschmutzungsanteil von Werten, die mit Mineralstaub assoziiert werden, zu Werten, die häufig mit anthropogenen Aerosolen asso-

ziiert werden. Dieses Ergebnis zeigt, dass die Ångström-Exponenten nicht als alleinige Indikatoren zur Identifizierung von Mineralstaubeignissen verwendet werden sollten. Die Ångström-Exponenten können aber helfen, den Verschmutzungsanteil zu quantifizieren, sobald das Vorhandensein von Mineralstaub mit anderen Methoden festgestellt wurde. Der Ångström-Exponent der Streuung scheint am besten dafür geeignet zu sein, da sich 72% seiner Variabilität durch den Verschmutzungsanteil erklären lässt.

Die Einfachstreuungsalbedo und der Asymmetrieparameter, Schlüsseleigenschaften für das Verständnis der direkten Strahlungswirkung von Aerosolpartikeln, nehmen von nicht verschmutzten zu verschmutzten Mineralstaubschichten deutlich ab. Bei einer Wellenlänge von 525 nm sinken z.B. die Medianwerte der Einfachstreuungsalbedo von 0,96 auf 0,89 und die Medianwerte des Asymmetrieparameters von 0,67 auf 0,56. Diese Änderungen können die Effizienz der Strahlungswirkung im kurzwelligen Spektralbereich gegensätzlich beeinflussen und sich dadurch unter bestimmten Bedingungen auch aufheben. Die Veränderung hängt jedoch von der Oberflächenalbedo ab, sodass die Effizienz der direkten Strahlungswirkung im kurzwelligen Spektralbereich durch die Verschmutzung signifikant beeinflusst werden kann.

Zusammenfassend zeigt diese Dissertation, dass es keine „beste“ Winkelkorrektur für Nephelometerdaten gibt. Es werden aber Empfehlungen für die Verwendung der am besten geeigneten Winkelkorrektur abhängig vom Aerosoltyp und der Partikelform gegeben. Damit trägt diese Dissertation dazu bei, die Unsicherheiten bei der direkten in-situ Messung des Streukoeffizienten von Mineralstaubpartikeln zu reduzieren. Basierend auf umfassenden Flugzeugmessungen untersucht diese Arbeit Mischungen von Mineralstaub und Verschmutzung systematisch. Dabei wird gezeigt, dass die Quantifizierung des Verschmutzungsanteils für die Bewertung der direkten Strahlungswirkung von Mineralstaubmischungen von großer Bedeutung ist.

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“At first, the mountain is a mountain. Then, it is no longer a mountain. In the end, the mountain is again a mountain.” – Ancient Zen saying

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List of Publications

This section lists all publications related to this dissertation. The first two papers present the main results of the dissertation, while the other two include contributions from the dissertation. Finally, the list of the conference contributions from this dissertation is reported.

Publications constituting this dissertation results

Paper I (published)

Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections

Teri, M., Müller, T., Gasteiger, J., Valentini, S., Horvath, H., Vecchi, R., Bauer, P., Walser, A., and Weinzierl, B.

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Paper II (accepted)

Pollution affects Arabian and Saharan dust optical properties in the eastern Mediterranean

Teri, M., Gasteiger, J., Heimerl, K., Dollner, M., Schöberl, M., Seibert, P., Tipka, A., Müller, T., Aryasree, S., Kandler, K., and Weinzierl, B.

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Other publications containing contributions from this dissertation

3. **Characterization of aerosol over the eastern Mediterranean by polarization sensitive Raman lidar measurements during A-LIFE – aerosol type classification and type separation**, Groß, S., Freudenthaler, V., Haarig, M., Ansmann, A., Toledano, C., Mateos, D., Seibert, P., Mamouri, R.-E., Nisantzi, A., Gasteiger, J., Dollner, M., Tipka, A., Schöberl, M., **Teri, M.**, and Weinzierl, B., *Atmospheric Chemistry and Physics*, 25, 3191–3211, <https://doi.org/10.5194/acp-25-3191-2025>, 2025.
4. **Mineral dust mixtures in the eastern Mediterranean: overview and selected highlights of the A-LIFE aircraft field experiment**, Weinzierl, B., M. Dollner, M., Gasteiger, J., **Teri, M.**, Schöberl, M., Heimerl, K., Kupc, A., Tipka, A., Seibert, p., Huntrieser, H., Wagner, R., Kandler, K., Aryasree, S., Müller, T., Brilke, S., Fölker, N., Sauer, D., Reitebuch, O., Groß, S., Freudenthaler, V., Toledano, C., Haarig, M., Mamouri, R., Amiridis, V., Ansmann, A., and the A-LIFE Science Team, in preparation.

Conference contributions

1. **The A-LIFE field experiment in the Eastern Mediterranean -Overview and early results**, Weinzierl, B., Gasteiger, J., **Teri, M.**, Dollner, M., Seibert, P., Philipp, A., Schöberl, M., Heimerl, K., Huntrieser, H., Wagner, R., Müller, T., Ansmann, A., Mateos, D., Toledano, C., and the A-LIFE Science Team, EGU-Meeting (oral), Vienna, 2018.
2. **The A-LIFE field experiment in the Eastern Mediterranean -Overview and early results**, Weinzierl, B., Gasteiger, J., **Teri, M.**, Dollner, M., Seibert, P., Philipp, A., Schöberl, M., Heimerl, K., Huntrieser, H., Wagner, R., Müller, T., Ansmann, A., Mateos, D., Toledano, C., and the A-LIFE Science Team, Dust Workshop (oral), Tenerife, 2018.
3. **Airborne multi-wavelength scattering measurements in Saharan and Arabian dust layers**, **Teri, M.**, Gasteiger, J., Müller, T., Dollner, M., Seibert, P., Philipp, A., Schöberl, M., Chouza Keil, F., G., and Weinzierl, B., First A-LIFE Workshop (oral), Vienna, 2018.
4. **Airborne black carbon measurements during A-LIFE**, Heimerl, K., **Teri, M.**, Dollner, M., Gasteiger, J., Seibert, P., Philipp, A., and Weinzierl, B., First A-LIFE Workshop (oral), Vienna, 2018.
5. **The A-LIFE field experiment in the Eastern Mediterranean -Overview and early results**, Weinzierl, B., Gasteiger, J., **Teri, M.**, Dollner, M., Seibert, P., Philipp, A., Schöberl, M., Heimerl, K., Huntrieser, H., Wagner, R., Müller, T., Ansmann, A., Mateos, D., Toledano, C., and the A-LIFE Science Team, Dust Workshop (oral), Darmstadt, 2018.
6. **Airborne multi-wavelength polar scattering measurements in Saharan and Arabian dust layers during A-LIFE**, **Teri, M.**, Gasteiger, J., Müller, T., Valentini, S., Horvath, H., Schöberl, M., Dollner, M., Seibert, P., Philipp, A., Vecchi, R., and Weinzierl, B., Second A-LIFE Workshop (oral), Darmstadt, 2018.
7. **Black Carbon in Saharan and Arabian dust layers during A-LIFE**, Heimerl, K., **Teri, M.**, Dollner, M., Gasteiger, J., Schöberl, M., Seibert, P., Philipp, A., and Weinzierl, B., Second A-LIFE Workshop (oral), Darmstadt, 2018.
8. **Airborne multi-wavelength scattering measurements in Saharan and Arabian dust layers during A-LIFE**, **Teri, M.**, Gasteiger, J., Müller, T., Dollner, M., Seibert, P., Philipp, A., Chouza Keil, F. G., Schöberl, M., and Weinzierl, B., European Research Course on Atmospheres (poster), Grenoble, 2019.
9. **Black Carbon in Saharan and Arabian dust layers during A-LIFE**, Heimerl, K., **Teri, M.**, Dollner, M., Gasteiger, J., Schöberl, M., Seibert, P., Philipp, A., and Weinzierl, B., 12th International Conference on Carbonaceous Particles in the Atmosphere (oral), Vienna, 2019.

10. **Basics and Examples of Aerosol Optics**, Teri, M., Gasteiger, J., Schöberl, M., Dollner, M., Heimerl, K., Seibert, P., Philipp, A., and Weinzierl, B., VDSP Science day (oral), Vienna, 2019.
11. **Validation of the truncation error correction for the Aurora 4000 polar nephelometer**, Teri, M., Müller, T., Gasteiger, J., Valentini, S., Horvath, H., Vecchi, R., and Weinzierl, B., European Aerosol Conference 2020 (oral), Online, 2020.
12. **Evaluation of the angular correction for the Aurora 4000 polar nephelometer**, Teri, M., Müller, T., Gasteiger, J., Valentini, S., Horvath, H., Vecchi R., and Weinzierl, B., Third A-LIFE Workshop (oral), Online, 2021.
13. **Die Quellen von Aerosol im östlichen Mittelmeer während der A-LIFE-Feldmesskampagne 2017**, Seibert, P., Philipp, A., Gasteiger, J., Heimerl, K., Teri, M., Dollner, M., and Weinzierl, B., DACH Meteorologentagung (poster), Garmisch-Partenkirchen, 2019.
14. **Model-based closure experiments with optical particle counters for dust-like aerosols**, Gasteiger, J., Walser, A., Dollner, M., Teri, M., Weinzierl, B., EGU General Assemblies (oral), Online, 2021.
15. **Optical properties of the mixed aerosol layers measured during the A-LIFE field experiment in the Eastern Mediterranean**, Teri, M., Gasteiger, J., Heimerl, K., Dollner, M., Schöberl, M., Seibert, P., Tipka, A., Maherndl, N., Müller, T., Aryasree, S., Kandler, K., and Weinzierl, B., International Aerosol Conference 2022 (oral), Athens, 2022.
16. **The impact of mixing pollution and mineral dust from different sources on aerosol optical properties in the Eastern Mediterranean**, Teri, M., Gasteiger, J., Heimerl, K., Dollner, M., Schöberl, M., Seibert, P., Tipka, A., Müller, T., Aryasree, S., Kandler, K., and Weinzierl, B., Staubtag (oral), Leipzig, 2023.
17. **The impact of mixing pollution and mineral dust from different sources on aerosol optical properties in the Eastern Mediterranean**, Weinzierl, B., Dollner, M., Gasteiger, J., Teri, M., Schöberl, M., Heimerl, K., Kupc, A., Tipka, A., Seibert, P., Huntrieser, H., Wagner, R., Kandler, K., Sudharaj, A., Müller, T., Brilke, S., Fölker, N., Sauer, D., Reitebuch, O., Groß, S., Freudenthaler, V., Toledano, C., Haarig, M., Mamouri, R., Amiridis, V., Ansmann, A., and the A-LIFE Science Team, American Geophysical Union Fall Meeting 2023 (oral), San Francisco, 2023.
18. **From mineral dust mixtures to microplastic - insights from recent aircraft filed experiments and laboratory studies**, Weinzierl, B., Dollner, M., Schöberl, M., Teri, M., Gasteiger, J., Heimerl, K., Beres, N., Wieland, L., Held at University of Natural Resources and Life Sciences (oral), Vienna, 2024.

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Preamble

1. Motivation and background

Climate change is one of the most urgent challenges of our time, affecting both humans and ecosystems. Over the past decade, rising global surface temperature and ocean heat content, as well as decreasing glacier mass and sea ice extent, have continuously set new records (WMO, 2025b). Extreme weather events, including heavy rains, floods, heat waves, and droughts, have become more frequent (WMO, 2025a).

According to the 6th Intergovernmental Panel on Climate Change report (IPCC, 2023), the global surface temperature has increased by 1.1°C in 2011-2020 compared to pre-industrial levels (1850-1900). Climate change is caused primarily by anthropogenic greenhouse gases, which contribute to a warming effect ranging from 1.0°C to 2.0°C . Other human activities, including aerosol particle emissions, have been shown to have a cooling effect ranging from 0.0°C to 0.8°C , partly masking warming by greenhouse gases. However, aerosol particles can also contribute to warming the atmosphere (Stocker et al., 2013; IPCC, 2023).

The Earth’s climate is altered when the balance between incoming shortwave solar energy and outgoing longwave energy at the top of the atmosphere is perturbed (e.g., Seinfeld and Pandis, 2016). The net change in the Earth’s radiation budget in Wm^{-2} (radiative effect) and its change from the pre-industrial era to the present day (radiative forcing) caused by various natural and anthropogenic factors have been estimated (IPCC, 2023). The radiative effect and forcing by aerosol particles are still uncertain (Bender, 2020; IPCC, 2021), largely contributing to challenges in estimating future climate changes using global climate models (e.g., Stocker et al., 2013; Forster et al., 2021; Watson-Parris and Smith, 2022). These uncertainties result from an incomplete understanding of aerosol interactions with clouds, and limited measurements of the interactions of anthropogenic and natural aerosols with radiation (Li et al., 2022).

1.1 Aerosol basics

Aerosols are stable suspensions of solid or liquid particles in the air (e.g., Hinds, 2012). Aerosol particles are emitted into the atmosphere from natural sources, including deserts, volcanoes, sea spray and plants, as well as from anthropogenic activities, such as industrial processes, vehicle emissions and the combustion of fuels. Particles can be emitted directly or formed in the atmosphere by conversion of gas into particles, such as sulfates, nitrates or organics. Due to their relatively low atmospheric lifetime, spanning from a few hours to several weeks, aerosol particles have a non-homogeneous spatiotemporal distribution, with tropospheric concentration in the range of $10^2 - 10^5 \text{ cm}^{-3}$ in number and of $1 - 100 \mu\text{g m}^{-3}$ in mass (Pöschl, 2005). Once in the atmosphere, aerosol particles can grow, interact with other aerosol species, be transported thousands of kilometres, and deposit on the Earth’s surface or be incorporated

into cloud droplets. Aerosol particles are an important part of the Earth-atmosphere system, substantially influencing visibility (e.g., Singh et al., 2017), human health (Arfin et al., 2023) and climate (IPCC, 2023).

Aerosol particles influence the Earth’s radiation budget by scattering and/or absorbing the shortwave radiation (direct radiative effect) and modifying cloud properties (indirect radiative effect) (e.g., Charlson et al., 1992; Stocker et al., 2013; IPCC, 2023). The direct radiative effect uncertainties are due, on the one hand, to the non-homogeneous spatial and temporal variability of aerosol particles, on the other hand, to their diverse microphysical and chemical properties (Li et al., 2022). Composition, shape and size of aerosol particles influence their interaction with radiation (e.g., Bohren and Huffman, 2008; Seinfeld and Pandis, 2016). Some aerosol particle types, including sulfates, nitrates, and sea salt, are purely scattering aerosol particles. They reduce the amount of solar radiation reaching the Earth’s surface, contributing to a cooling effect (e.g., Li et al., 2022). Conversely, some aerosol types can also absorb the solar radiation, leading to a cooling effect at the surface while heating the air layer where the aerosol resides. Absorbing aerosol particles are mainly black carbon (BC) and organic carbon particles (Bond et al., 2013), originated from incomplete combustion of fossil fuels and biomass, and mineral dust particles emitted by wind via mobilization of soil particles in arid regions of the world (Kok et al., 2017). While black carbon is the most efficient atmospheric absorber (Bond et al., 2013), mineral dust is the most abundant aerosol in terms of mass (Choobari et al., 2014), and can also absorb the Earth’s outgoing long-wave radiation. Mineral dust particles can form elevated layers that can be transported thousands of kilometres (Weinzierl et al., 2017; Adebisi et al., 2023a), undergoing ageing and mixing processes that can modify the properties of these layers. Even the sign of the net direct radiative effect of mineral dust is uncertain (e.g., -0.48 to $+0.20$ Wm^{-2} , Kok et al., 2017). These uncertainties arise from variability in its microphysical and optical properties (Kok et al., 2017; Li et al., 2021; Adebisi et al., 2023b), and contribute largely to the overall uncertainty of mineral dust’s radiative effect (Kok et al., 2023).

Global climate models require more accurate knowledge of the aerosol optical properties to reduce the uncertainties in climate predictions (e.g., Laj et al., 2020; Watson-Parris and Smith, 2022; Kok et al., 2023). This dissertation investigates the optical properties of aerosol particles, particularly focusing on mineral dust and its mixtures with anthropogenic aerosols.

1.2 Atmospheric particle size distribution

An important parameter governing the particle-light interaction, but also their lifetime and contribution to cloud formation, is the particle size, ranging from a few nanometers to about $100\text{ }\mu\text{m}$ (e.g., Seinfeld and Pandis, 2016). The size distribution of atmospheric aerosol particles is characterized by four modes: nucleation, Aitken, accumulation, and coarse, which arise from various sources, formation and transformation processes (e.g., Whitby, 1978; Kulmala et al., 2004; Raes et al., 2000). A hypothetical particle number size distribution¹ illustrating these four modes is shown in Figure 1.1. The size ranges of the modes vary slightly in the literature. The definitions used in this thesis are given, for example, by Raes et al. (2000).

¹particle number concentration per logarithmic size interval, $dN/d(\log D)$, plotted against particle diameter.

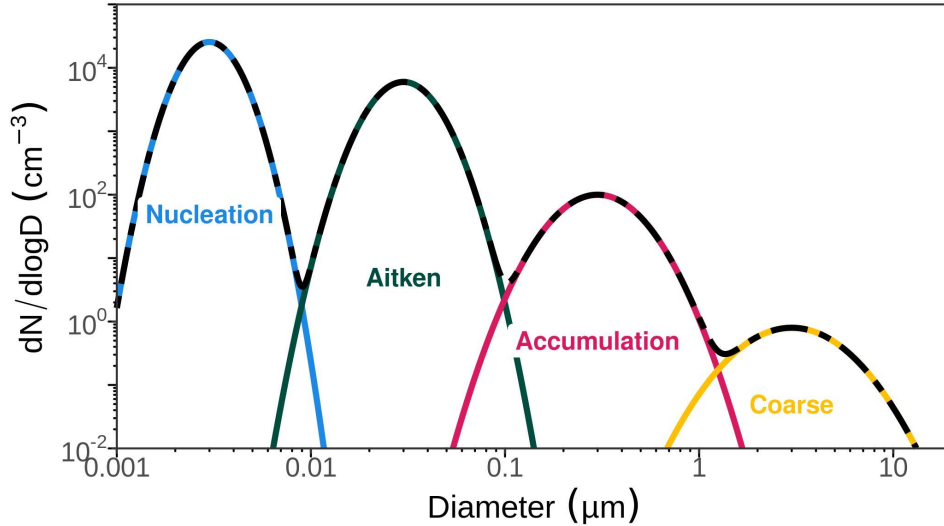


Figure 1.1: Hypothetical particle number size distribution illustrating the four atmospheric modes: nucleation, Aitken, accumulation, and coarse. The particle number concentration is chosen in the order of 10^3 cm^{-3} , which corresponds to a particle mass concentration of about $100 \mu\text{g m}^{-3}$, assuming a particle density of 2 g cm^{-3} .

The nucleation mode consists of secondary particles smaller than about 10 nm formed by gas-to-particle conversion (Kulmala et al., 2004). The Aitken mode includes primary particles emitted from combustion processes, along with secondary materials condensed on them, with sizes ranging between 10 and 100 nm. Particles in the accumulation mode have sizes from 100 nm to $1 \mu\text{m}$, and result from the coagulation of primary particles and the condensation of secondary materials on them.

The coarse mode contains particles larger than $1 \mu\text{m}$, mainly originating from mechanical processes. Mineral dust particles emitted by wind erosion belong to this mode, as also other natural aerosol particles, such as sea salt released by sea spray. Natural aerosol particles often have diameters larger than $1 \mu\text{m}$, though fine natural aerosol particles (diameter $< 1 \mu\text{m}$) can also result from processes such as gas-to-particle conversion (Hinds, 2012).

Anthropogenic aerosol particles are predominantly fine particles with diameter smaller than $1 \mu\text{m}$. Particles can transfer from the nucleation and Aitken mode to the accumulation mode through coagulation and growth processes (e.g., condensation of vapors). These mechanisms are less efficient for accumulation mode particles due to their larger sizes, limiting their growth towards the coarse mode. However, particles in the coarse mode can also mix with other species through exchange of mass with the gas phase (Raes et al., 2000).

Different regions, such as urban, rural or desert areas, can have different characteristic particle size distributions, with the four modes present in different relative proportions. However, the number size distribution is typically dominated by the nucleation and Aitken modes. Although high in number, particles in these modes are too small to contribute substantially in mass. Indeed, the accumulation and coarse modes dominate the volume and mass size distribution, and also the scattering and absorption properties of the aerosol population, which will be explained in details in the following section (Seinfeld and Pandis, 2016).

1.3 Particle-light interaction

When light encounters a particle, the electric charges within the particle are excited and begin to oscillate. This results in energy being re-radiated in all directions, a process known as scattering. Additionally, some of the energy may be transformed into thermal energy due to absorption (Seinfeld and Pandis, 2016).

The energy scattered by a particle is proportional to the intensity of the incident light, i.e., the energy flux crossing an area perpendicular to the light propagation. The proportionality coefficient is the particle scattering cross-section, C_{sca} , representing the effective area determining the scattering in units of m^2 . The particle absorption cross-section, C_{abs} , can be defined analogously. The sum of the scattering and absorption cross-sections is the particle extinction cross-section, C_{ext} , and represents the effective area determining the extinction, the light removed from the incident beam of light.

The particle scattering and absorption cross-sections depend primarily on (1) the wavelength of the incident radiation λ , (2) the particle diameter D , (3) the complex refractive index $n(\lambda) + ik(\lambda)$, and (4) the particle shape (e.g., Seinfeld and Pandis, 2016). The complex refractive index, which depends on λ and the particle composition, characterizes the interaction between electromagnetic radiation and the particle material relative to the surrounding medium. The real part, n , describes the propagation speed of radiation through a material and is primarily associated with scattering. The imaginary part, k , predominately describes absorption, being zero for non-absorbing materials (e.g., Bohren and Huffman, 2008; Petzold et al., 2013).

The scattering and absorption of light by a spherical particle is a classical problem in physics, which was formalized mathematically within the Mie Theory for homogeneous spherical particles (Mie, 1908; Bohren and Huffman, 2008; Horvath, 2009). Mie theory provides a complete formal solution of the Maxwell's electromagnetic equations for the scattering and absorption of infinite electromagnetic waves from homogeneous, isotropic, spherical particles. Thus, the scattering and absorption cross-sections and the angular distribution of scattering, i.e., the scattering phase function, for spherical particles of arbitrary size and refractive index can be calculated with Mie theory.

As an example, Figure 1.2 shows the scattering and absorption properties of aerosol particles at three wavelengths for the hypothetical particle number size distribution in Figure 1.1, assuming particles with a spherical shape and a refractive index of $1.53 + i0.01$. These plots indicate that the accumulation and coarse mode dominate the scattering and absorption of these hypothetical aerosols.

The scattering properties of particles in the accumulation mode show a more pronounced wavelength dependence than particles in the coarse mode. Indeed, particles smaller than the wavelength of the light scatter more efficiently at shorter wavelengths, as their behavior approaches the Rayleigh scattering regime (Rayleigh, 1899), where the scattering has a strong wavelength ($\sim \lambda^{-4}$) dependence. In contrast, particles with diameters comparable to or larger than the wavelength show a weaker wavelength dependence. This difference in wavelength dependence is responsible for many beautiful atmospheric phenomena, such as red sunsets, the blueness of the sky (which appears bluer in purer air), and the whiteness of clouds due to the larger size of water droplets (e.g., Seinfeld and Pandis, 2016).

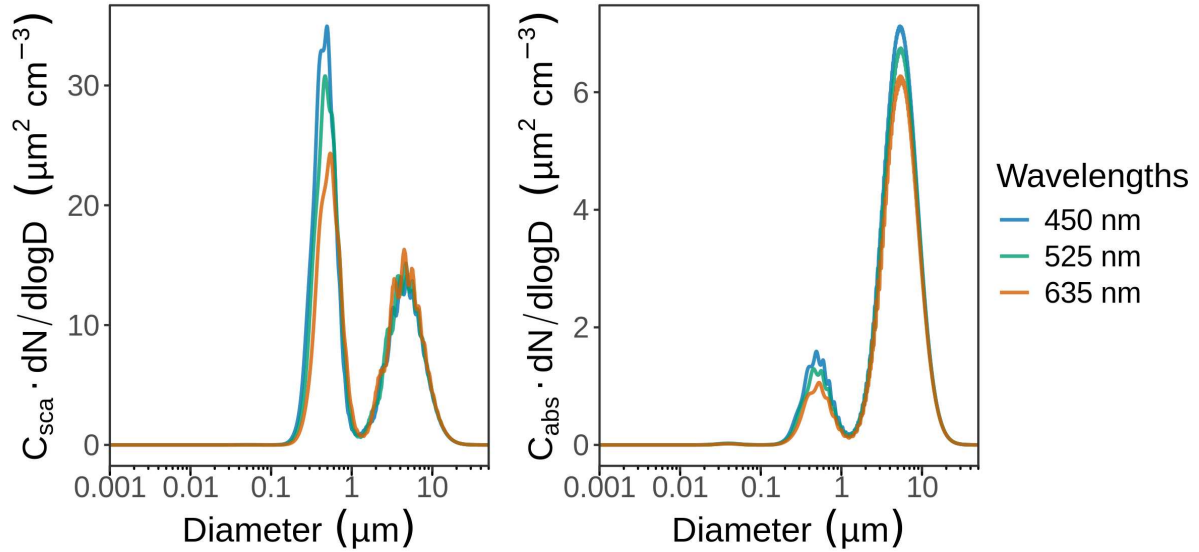


Figure 1.2: Scattering and absorption properties for a hypothetical particle size distribution as a function of particle diameter. The left plot shows the particle scattering cross-section, and the right plot shows the particle absorption cross-section, both multiplied by the hypothetical particle number size distribution illustrated in Figure 1.1. Simulations were performed using Miepython, a Python module developed by Scott Prahl. The particles were assumed to have a spherical shape with the particle size distribution of Figure 1.1 and with refractive index $1.53 + i0.01$. The colors indicate the wavelengths 450 nm (blue), 525 nm (green), 635 nm (red).

1.4 Optical properties of an ensemble of particles

The scattering and absorption properties of an ensemble of particles can be obtained from the single particle properties, if the scattering can be considered independent and no multiple scattering occur. These conditions are satisfied for many atmospheric conditions, when the distance between particles is sufficiently large and the atmosphere is optically thin. The particle scattering and absorption coefficients (σ_{sp} and σ_{ap}) represent the fraction of light that aerosol particles scatter out of or absorb from a parallel beam of light per unit length. These coefficients are calculated by summing C_{sca} and C_{abs} of all particles in an air parcel, and dividing by the parcel's volume.

The particle scattering and absorption coefficients at multiple wavelengths can be used to derive a set of intensive optical properties, which provide information on the aerosol type dominating the optical properties, and are essential for understanding and estimating its direct radiative effect. The scattering and absorption Ångström exponents (SAE, AAE) – representing the wavelength dependence of the scattering and absorption coefficients (σ_{sp} , σ_{ap}) – are linked to different aerosol characteristics: particle size and composition, respectively (Ångström, 1929; Schuster et al., 2006; Liu et al., 2018). Thus, the Ångström exponents have been used in literature to identify different aerosol types (e.g., Cappa et al., 2016), although the threshold values may vary depending on the specific site, measurements methods and wavelengths (Ealo et al., 2016). $SAE < 1$ indicates dominance of coarse particles, while larger SAE suggests fine particles. Pure BC particles have an AAE around 1, as BC absorbs across the solar spectrum (e.g., Bond,

2001). In contrast, mineral dust and organic carbon, which absorb more efficiently at shorter wavelengths, typically show higher AAE values (e.g., Valentini et al., 2020).

The single-scattering albedo (SSA), and the asymmetry parameter (g) are two key optical properties for estimating the aerosol direct radiative effect (Charlson et al., 1991; Haywood and Shine, 1995). The SSA represents the fraction of light extinction attributed to the scattering; it equals 1 for purely scattering aerosol particles, and it decreases if absorbing particles are present. The asymmetry parameter, g , provides information about the asymmetry of the angular distribution of the scattered light. For atmospheric aerosols, it is in the range 0.4 – 0.8, where larger values indicate more forward scattering due to larger particles (e.g., Horvath et al., 2018; Pandolfi et al., 2018). A more detailed description of these properties and their atmospheric values can be found in section 2.3.2 of Teri et al. (2024), whose accepted version is in the appendix of this dissertation.

As an example, the particle scattering and absorption coefficients are determined for the hypothetical atmospheric aerosol of Figure 1.1 by integrating the curves shown in Figure 1.2. The resulting values for σ_{sp} are 44.4, 42.0, and 38.1 Mm^{-1} at blue, green, and red wavelengths, respectively. For σ_{ap} , the values are 10.1, 9.4, and 8.6 Mm^{-1} at the same wavelengths. The corresponding values of SAE, AAE, at the blue and red wavelengths are 0.45, 0.46, and of SSA, and g at the green wavelength are 0.82, and 0.76, respectively. These values are quite realistic for an aerosol dominated by coarse particles ($\text{SAE} < 1$), with a tendency to forward scattering ($g = 0.76$), and where BC particles dominate the absorption (low AAE). However, the atmospheric aerosol optical properties may differ significantly from this hypothetical case. In fact, not only a hypothetical particle size distribution was used, but also the same refractive index ($1.53 + i0.01$) and shape (spherical) were assumed for all particles.

The refractive index of aerosol particles depends on their composition, it can vary with the wavelength of the incident radiation, and it can also exhibit considerable variability within the same aerosol type. The chosen real part of the refractive index, 1.53, is appropriate for efficient scattering materials and it aligns with values of sulfate, secondary organic, and mineral dust aerosols (e.g., Toon et al., 1976; Hess et al., 1998). The imaginary part, $i0.01$, is consistent with a weakly absorbing aerosol particles. As BC particles have a considerably higher refractive index (e.g., $1.95 + i0.79$ at 550 nm, Bond and Bergstrom, 2006), this hypothetical aerosol may be realistic for a mixture of BC and non-absorbing particles. In contrast, the imaginary refractive index of mineral dust particles is lower and depends on the wavelength, with a value in the range $i0.0006$ to $i0.0048$ at 520 nm according to Di Biagio et al. (2019). The mineral dust’s refractive index can vary based on the relative proportion of various minerals, including quartz, hematite, ilite, kaolinite, and calcite, which may be present as individual particles or as aggregates (Sokolik and Toon, 1999).

Another property that influences the aerosol optical properties is the particle shape. For instance, mineral dust particles are irregularly shaped (Kandler et al., 2009) and do scatter more in the forward direction and less in the backward direction compared to spherical particles (Horvath et al., 2018). Also, BC particles can have a non-spherical shape as they coagulate, forming aggregates with a chain-like structure (Bond et al., 2013). Although computational methods, such as the T-Matrix (Mishchenko and Travis, 1998) and the discrete dipole approx-

imation (Yurkin and Hoekstra, 2011), exist to account for particle shape in optical modeling, they tend to take orders of magnitude more computation time, compared to Mie theory.

The already complex characterization of the optical properties of individual particles is even more challenging considering the particle mixing state, i.e., how chemical components are distributed among particles. Aerosol particles in the atmosphere are subject to ageing and mixing processes, leading to two primary mixing states: external mixtures, where each particle arises from only one source, and internal mixtures, where different components are combined within the same particle (Seinfeld and Pandis, 2016). The particle mixing state can influence the aerosol optical properties. For instance, secondary material can condense on BC particle aggregates, forming a coating that enhances the absorption by BC particles (e.g., Liu et al., 2015). Accurately representing the complexity of internal and external mixtures in aerosol optical modelling poses important challenges (Fierce et al., 2017; Yao et al., 2022).

Accurate knowledge of the aerosol optical properties is crucial for assessing the influence of aerosol particles on the Earth’s climate. The aerosol direct radiative effect depends on the particle extinction coefficient, single-scattering albedo, and asymmetry parameter. Traditionally, these properties are obtained from remote sensing retrievals (e.g., Granados-Muñoz et al., 2019) or via optical modeling based on measured particle size distributions (e.g., Hermann et al., 2016; Vasilatou, 2021). However, these methods require assumptions on the particle type, shape, refractive index and mixing state. Alternatively, several in situ measurement techniques can provide the aerosol optical properties, in principle, without the need for these assumptions.

This dissertation deploys some of these techniques, specifically an integrating nephelometer and a filter-based absorption photometer, to investigate the aerosol optical properties of mineral dust, and mineral dust-pollution mixtures.

1.5 Airborne in situ observations of mineral dust

In the past years, a series of intensive field experiments, including airborne and ground-based measurements, have been conducted with the objective of investigating the properties of mineral dust particles through the tropospheric column and better characterizing the mineral dust’s radiative effect. These efforts mainly focused on the study of pure mineral dust layers from the Sahara desert near the source region and after transport. For example, the Saharan Dust Experiment (SHADE) was conducted over the eastern part of the Atlantic Ocean (Formenti et al., 2003), the Puerto Rico Dust Experiment (PRIDE) investigated the dust transport over the subtropical Atlantic Ocean, the African Monsoon Multidisciplinary Analysis (AMMA) studied the properties of mineral dust in Western Africa (Formenti et al., 2011), and the Fennec 2011 campaign was conducted over the Mauritania and Mali regions of the Sahara desert (Ryder et al., 2013). The Saharan Mineral Dust Experiment (SAMUM) was one of the most extensive study of Saharan dust properties. It was designed as a closure study, combining airborne and ground-based in situ and remote sensing measurements with model simulations. The first part of this campaign, known as SAMUM-1, focused on the properties of freshly-emitted pure mineral dust in the source region in Morocco. The second part SAMUM-2 investigated aged mineral dust transported into the Cape Verde region, and also its mixtures with marine and biomass-burning aerosols (Ansmann et al., 2011). The Saharan Aerosol Long-range Transport and Aerosol-Cloud-

Interaction Experiment (SALTRACE) investigated the properties of Saharan dust transported into the Caribbean (Weinzierl et al., 2017).

Within this effort, it has become evident that further studies on mixtures of mineral dust with other aerosol types are necessary. The Asian Pacific Regional Aerosol Characterization Experiment (ACE-ASIA) investigated the properties of mineral dust-pollution mixtures in Asia, and its results suggest that black carbon particles within elevated mineral dust layers might have a proportionately stronger impact on atmospheric heating than black carbon particles at lower altitude (Seinfeld et al., 2004). A few case studies using data collected during the SAMUM experiment found that the absorbing properties of mineral dust may be significantly affected by the mixing with anthropogenic aerosols. For example, in the polluted city of Casablanca (Morocco), the absorption Ångström exponent has a value of about 1 – characteristic for BC particles– despite the high mineral dust concentration (Petzold et al., 2009). The polluted plume over the city of Dakar (Senegal) was investigated during a Saharan dust outbreak, revealing a significant influence of the pollution on the particle size distribution and absorption coefficient of the aerosol mixture but not on the absorption Ångström exponent. The Chemistry-Aerosol Mediterranean Experiment (CHARMEX) was conducted with the aim to characterize Saharan dust plumes in the polluted western and central Mediterranean basin (Mallet et al., 2016). However, the analysis of the measured optical properties revealed a low variability of the absorption properties of mineral dust layers, suggesting that the Saharan dust optical properties were not significantly affected by mixing with pollution (Denjean et al., 2016). In contrast, measurements during the Dynamics-Aerosol-Chemistry-Clouds Interactions in West Africa (DACCIWA) experiment showed enhanced aerosol light absorption in mineral dust layers due to mixing with pollution (Denjean et al., 2020). In conclusion, mineral dust from various source regions may be affected differently by mixing with pollution, and thus, this influence needs to be further investigated in different areas and source regions.

This thesis is written in the framework of the A-LIFE (Absorbing aerosol layers in a changing climate: ageing, lifetime and dynamics) project, which has the overall aim to investigate the properties of absorbing aerosols (specifically mineral dust - black carbon mixtures) during their atmospheric lifetime in the eastern Mediterranean. This region is of unique interest because it is a hot spot for climate change and pollution. The eastern Mediterranean and Middle East regions are warming nearly twice the global average, increasing extreme weather events (Zittis et al., 2022). The eastern Mediterranean is an atmospheric crossroad, where pollution, including black carbon and sulfate aerosols, co-exist with mineral dust from the Arabian peninsula and the Saharan desert (Dayan et al., 2017).

During the A-LIFE field experiment, conducted in April 2017, a new data set was collected on board the research aircraft Falcon through the tropospheric column over the eastern Mediterranean. The A-LIFE overview paper, including a detailed description of the field experiment, the meteorological conditions influencing the measurements, and a classification of the aerosol types measured on board the aircraft Falcon, is in preparation (Weinzierl et al., in preparation, 2025). An investigation of the aerosol types affecting the optical properties measured with lidars and solar photometers during the A-LIFE field experiment is provided by Groß et al. (2025). The work of this thesis contributed to the two aforementioned publications, however, the core pub-

lications of this dissertation are Teri et al. (2022) and Teri et al. (2024), whose overall objective is presented in the following section.

1.6 Research questions and thesis structure

The overall objective of this dissertation is to improve the understanding of the optical and radiative properties of mineral dust and its mixtures with pollution. Two key instruments were utilized to achieve this goal: the Aurora 4000 Polar Integrating Nephelometer (Aurora 4000, Ecotech –now Acoem) and a Tri-color Absorption Photometer (TAP, Brechtel), which provide the particle scattering and absorption coefficients and information on the angular scattering distribution. While the measurement principle of these instruments minimizes the need for assumptions regarding aerosol type, refractive index, shape or mixing state, these properties can influence the measurement accuracy.

The data used in this dissertation were collected through three experiments. First, a laboratory closure experiment aimed to evaluate the quality of the scattering coefficient measurements from the Aurora 4000 nephelometer, particularly for mineral dust. Second, a simulated closure experiment aimed to interpret the laboratory findings and to extend them to different particle sizes, refractive indices, and shapes. Third, a comprehensive data set used was collected during the A-LIFE aircraft field experiment, which aimed to investigate the properties of absorbing aerosol layers in the eastern Mediterranean, a region influenced by mineral dust from two source regions and local and transported pollution.

These extensive experiments allowed for the quantification of nephelometer measurement uncertainties, particularly for irregularly shaped mineral dust-like aerosol (Paper I, Teri et al. (2022), published) and the investigation of the optical properties of mineral dust-pollution mixtures (Paper II, Teri et al. (2024), accepted for publication in ACP). The research questions of this dissertation are as follows:

1. How accurately can the Aurora 4000 nephelometer measure the particle scattering coefficient of coarse-mode irregularly shaped mineral dust particles?
2. What is the most appropriate method for determining the particle scattering coefficient using an integrating nephelometer, for different particle sizes, refractive indices, and shapes?
3. How does the pollution affect the optical properties of mineral dust layers, and does this effect vary depending on the mineral dust’s source region?
4. To what extent can measurements from the Aurora 4000 and TAP instruments be used to identify mineral dust layers in the presence of pollution?
5. What is the effect of pollution on the radiative properties of mineral dust layers?

This cumulative dissertation is structured as follows.

The methods used to address this thesis’s scientific questions are presented in chapter 2. This chapter includes descriptions of the measurement principles, operations, and limitations of the Aurora 4000 Polar Integrating Nephelometer (section 2.1) and the Tri-color Absorption

Photometer (TAP; section 2.2). Additionally, the algorithms for data quality control and processing are described in sections 2.1.4 and 2.2.4 for the Aurora 4000 and TAP, respectively. Other instruments and optical simulation tools utilized throughout this dissertation are summarized in section 2.3. Finally, the three main experiments of this thesis – the laboratory closure experiment, the simulated closure experiment, and the A-LIFE field experiment – are described in section 2.4.

The results of this cumulative dissertation are presented through two publications in chapters 3 (Paper I, Teri et al. (2022), published) and 4 (Paper II, Teri et al. (2024), accepted for publication in ACP). These chapters present the abstracts and essential information, while the complete manuscripts are provided in the appendixes. Paper I (Teri et al., 2022) presents findings from the laboratory closure experiment and the simulated closure experiment. It assesses the uncertainties associated with the Aurora 4000 nephelometer, with a focus on coarse-mode irregularly shaped particles. The paper improves our understanding of various methods to obtain the particle scattering coefficient by integrating nephelometer for various particle sizes, refractive index and shapes, therefore addressing research questions 1 and 2. Paper II (Teri et al., 2024) presents results from the A-LIFE field experiment, investigating the effect of increasing amount of pollution within mineral dust layers from two source regions on the intensive aerosol optical properties. It discusses the implications for the identification of mineral dust layers in the presence of pollution and the direct radiative effect of mineral dust layers. Consequently, this paper answers the research questions 3, 4, and 5.

A summary of this dissertation, including a detailed answer for each research question, is provided in chapter 5.

2. Methods

This dissertation utilized laboratory experiments, model simulations, and aircraft in situ observations. The following sections introduce the core instruments of this dissertation: the Aurora 4000 Polar Integrating Nephelometer (Aurora 4000, Ecotech–now Acoem) and the Tri-color Absorption Photometer (TAP, Brechtel). These instruments provide essential information for understanding the optical properties, and thus the radiative effects, of aerosol mixtures. Subsequently, additional instruments measuring the aerosol microphysical properties and optical simulation tools used in this thesis work are described. Last but not least, the three experiments of this dissertation are outlined in section 2.4: a laboratory closure experiment, a simulated closure experiment, and the A-LIFE field experiment.

2.1 Aurora 4000 Polar Integrating Nephelometer

The Aurora 4000 Polar Integrating Nephelometer is a commercially available instrument manufactured by Ecotech–now Acoem (Ecotech, 2011). It measures the light scattered by particles and air in a sample volume at three wavelengths: $\lambda = 450, 525, \text{ and } 635 \text{ nm}$ (blue, green, and red) and for up to 18 angular sectors spanning from α to 180° , where α can be any angle in the interval 0° to 90° . Thus, the Aurora 4000 nephelometer provides values proportional to the particle scattering coefficients $\sigma_{\text{sp},\lambda}^\alpha$, the fraction of light scattered by particles out of a light beam per unit length. $\sigma_{\text{sp},\lambda}^\alpha$ for the total ($\alpha = 0^\circ$) and the other angular sectors (α between 10° and 90°) are obtained subject to calibrations with particle-free gases and corrections for angular illumination and truncation.

The particle scattering coefficient is an extensive aerosol property, which scales with the concentration of particles contributing to light scattering, which typically is the largest contribution to light extinction. By measuring the particle scattering coefficient at different wavelengths and for different angular sectors, key climate-relevant intensive optical properties can be derived, such as the scattering Ångström exponent and the asymmetry parameter. The particle scattering coefficient can also be used to determine the single-scattering albedo if either the particle absorption or extinction coefficient is available.

Figure 2.1 shows the Aurora 4000 nephelometer, which is part of the University of Vienna’s equipment, and it is currently integrated into the "Aerosol, Clouds and Trace Gases Research Infrastructure (ACTRIS)"

National Facility at the rooftop of the Faculty of Physics building. This instrument was de-

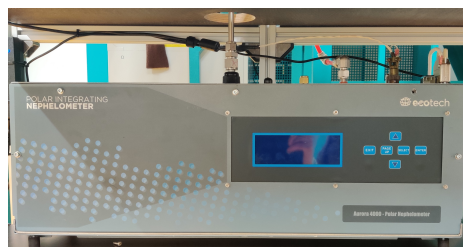


Figure 2.1: The Aurora 4000 nephelometer of the University of Vienna. Picture taken at the ACTRIS National Facility in the Faculty of Physics.

ployed for airborne measurements during the A-LIFE field experiment, and thus, it was specially designed and modified for airborne measurements, i.e., to operate with an external pump at low pressure and to fit into the aircraft rack. Figure 2.2 shows the flow plan of the Aurora 4000 nephelometer, highlighting the internal modifications performed by the company Acoem (in red) and the components added externally (in yellow) for the A-LIFE field experiment.

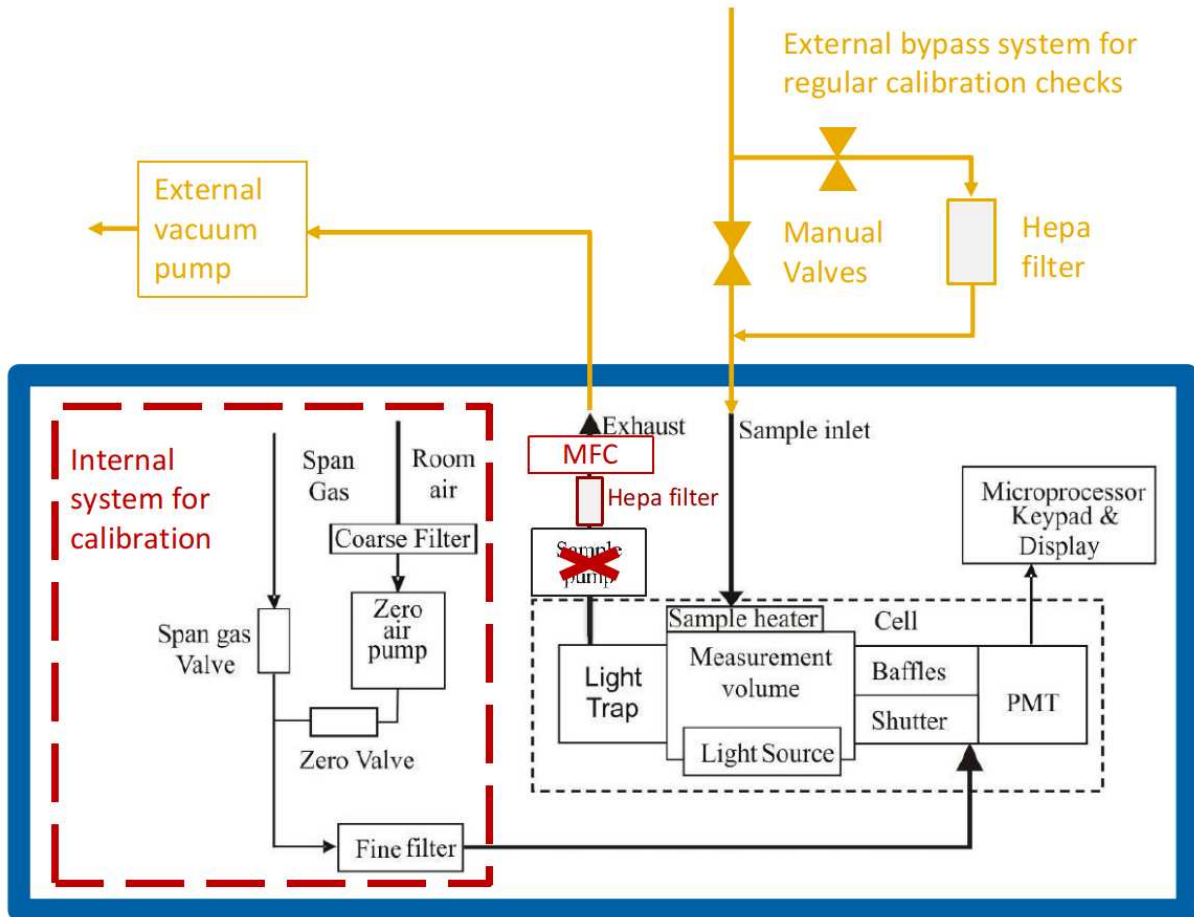


Figure 2.2: Flow plan of the Aurora 4000 nephelometer during A-LIFE. The internal flow plan is shown inside the blue box. Modifications made by Acoem are shown in red: the sample pump was removed, a Standard Mass Flow Controller (MFC KOFLOC, MODEL 3660) was added for flow regulation, a HEPA filter was introduced to protect the MFC from aerosol particles, and the instrument case was shortened to fit into the aircraft rack. The internal calibration system was left unchanged, but it was used only for a first calibration at the beginning of the field experiment. The instrument was integrated into the research aircraft by adding external components, as shown in yellow: a vacuum pump was used to allow operation at low pressure, and a bypass system was built to perform calibration checks regularly. (Figure modified from the Aurora 4000 nephelometer user manual (Ecotech, 2011) with permission from Acoem.)

The sample air is continuously drawn through a black anodized aluminium cell, which is internally coated with matte black paint, to minimize light scattering from internal walls, and pneumatically and optically sealed to prevent the entry of external light and air. The light source, located at the cell top, consists of light-emitting diodes covered with a glass diffuser, ensuring an evenly distributed illumination, known as Lambertian illumination. The scattered photons are detected by a photomultiplier tube placed at 90° to the light source. Periodically,

the light source is shaded by a glass with known transmittance (reference shutter) to check and compensate for variations of the light source intensity and light scattering from the internal walls. Different sensors provide information about temperature, pressure, and relative humidity. A detailed description of the Aurora 4000 nephelometer, its components, and operating procedures can be found in the user manual (Ecotech, 2011). The measurement principle of integrating nephelometers and polar integrating nephelometers is explained in the following section, and also in Teri et al. (2022).

2.1.1 Measurement principle

The Aurora 4000 nephelometer uses the measuring principle of integrating nephelometers. Integrating nephelometers, now widely used in monitoring stations, laboratory and field experiments (e.g., Pandolfi et al., 2018), were first introduced by Beuttell and Brewer (1949) for estimating the horizontal visibility during nocturnal military operations. An historical review of the designs and applications of integrating nephelometers can be found in Heintzenberg and Charlson (1996), while a description of the measurement principle is provided by Anderson et al. (1996).

The measurement principle of integrating nephelometers is based on their optical geometry. They perform a geometric integration of the light scattered by particles and air over nearly the full solid angle. The light detector is placed orthogonally to the light source, allowing it to collect light scattered from each volume element situated along the axis of the viewing cone, corresponding to different scattering angles.

Figure 2.3 shows the geometry of integrating nephelometers (left) and polar integrating nephelometers (right). The green area shows the measurement volume, i.e., the intersection between the volume illuminated by the light source (in yellow) and the conical volume seen by the light detector (in blue). θ is used to indicate the scattering angle, i.e., the angle between the direction of the incident beam and the one of the scattered light received by the light detector.

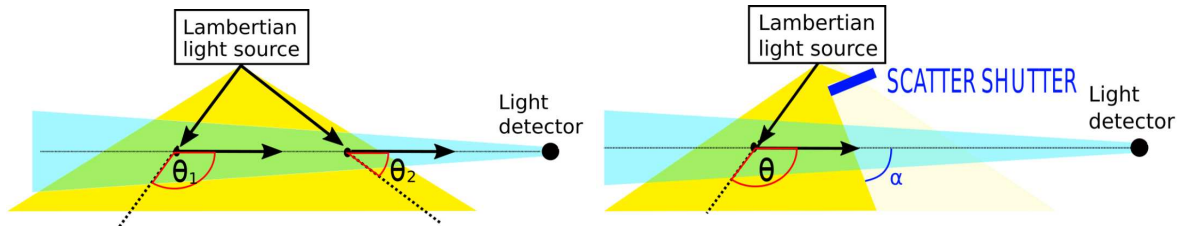


Figure 2.3: Scheme of the measurement principle of integrating nephelometers (left) and of polar integrating nephelometers (right). The volume illuminated by the Lambertian light source is represented by the yellow area, the conical volume seen by the light detector by the blue area, and the intersection between them, i.e., the measurement volume, by the green area. For a more detailed scheme, see Anderson et al. (1996).

The angular illumination function provided by a Lambertian light source follows the cosine emission law, i.e., the intensity of the emitted light is proportional to the cosine of the angle between the direction of the emitted light and the surface normal to the light surface. Consequently, the angular illumination function can be expressed as $\sin(\theta)$ for an ideal nephelometer. The signal measured by integrating nephelometers is proportional to the total scattering coefficient, ideally for the full range of the scattering angle, θ from 0° to 180° .

Polar integrating nephelometers are additionally equipped with a scatter shutter, which can be positioned in front of the light source and periodically shades different sectors of the light illumination, as shown in Figure 2.3.b. For a given angular position α of the scatter shutter, the measurement volume is reduced such that the geometrical integration is performed only for light scattered at angles larger than α , i.e., in the angular sector from α to 180° .

More detailed information on the measurement principle of the Aurora 4000 nephelometer can be found in section 2 and 3 of Teri et al. (2022), included in this thesis appendix.

2.1.2 Calibration

The particle scattering coefficient for each angular sector is obtained from the nephelometer signal by calibration with gases of known scattering coefficient and subtraction of the Rayleigh scattering coefficient of particle free air for the corresponding angular sector. The calibration of nephelometers is based on the assumption of a linear response of the measured signals to changes in the scattering coefficient. The calibration line parameters are determined by measurements of two gases with a high and a low scattering coefficient value, usually CO_2 and particle-free air. Routine measurements of the calibration gases are recommended (Anderson and Ogren, 1998; WMO/GAW, 2016), to provide immediate and highly accurate feedback on instrument performance, allow correction of calibration drift to improve accuracy, and build up a record of instrument noise and drift to quantify the measurement precision. Details about the Aurora 4000 nephelometer calibration can be found in the user manual (Ecotech, 2011).

2.1.3 Nephelometer angular truncation and illumination correction

Measurements of integrating nephelometers and polar integrating nephelometers are affected from angular non-idealities: the impossibility to measure the light scattered in the extreme forward and backward direction, the non-perfect Lambertian illumination and the non-perfectly sharp separation of the scatter shutter. While the angular sensitivity function of an ideal nephelometer can be expressed by a sine function, the angular sensitivity function of a real nephelometer considers its angular non idealities. The angular sensitivity function of the Aurora 4000 nephelometer was derived by Müller et al. (2012), and it is also provided in section 3.1 and Table 1¹ of Teri et al. (2022). This function was validated within this thesis using laboratory measurements of aerosols with known size and refractive index (see section 2.4.1).

The particle scattering coefficient in the complete angular range is substantially affected by these angular non-idealities. Thus, the Aurora 4000 measurements need to be corrected by applying a factor called "angular correction". Since the light scattered in the extreme forward direction increases with the size of particles (Heintzenberg and Charlson, 1996), and can also be affected by the aerosol type (Bond et al., 2009), and shape (Schladitz et al., 2009, 2011), calculating the angular correction is non-trivial.

A summary of different available methods to obtain the angular correction, along with their limitations, is available in Table 2 and Section 3 of Teri et al. (2022). These methods include the angular correction C_{SAE} , an empirical parametrization using as input variable the scattering Ångström exponent (Anderson and Ogren, 1998; Müller et al., 2011; Teri et al., 2022), the angular

¹Please see the Corrigendum for a misprint of one parameter in Table 1.

correction C_{polar} , an empirical parametrization using the polar measurements of the Aurora 4000 nephelometer Müller et al. (2012), and the angular correction C_{phase} (Heintzenberg et al., 2006), requiring knowledge of the phase function, which can be derived via optical simulations from the particle size distribution measured, for example, with an OPS ($C_{\text{phase,OPS}}$).

These methods are evaluated within this thesis through a laboratory closure experiment and a simulated closure experiment, and guidelines for calculating the most appropriate angular correction in various situations are presented (see Figure 8 of Teri et al. (2022)).

2.1.4 Data processing algorithm and quality control

Within the work for this thesis, an algorithm for processing data from the Aurora 4000 nephelometer was developed, and the code was written in the language R (R Core Team, 2024). The algorithm includes all the necessary steps for converting the raw data to the final form, including cleaning, determining calibration parameters, processing and plotting. The data processing involves the correction of the particle scattering coefficients for calibration drift, the conversion to standard temperature and pressure (273.15 K, 1013.25 hPa), the subtraction of Rayleigh scattering, and the angular truncation and illumination correction, as well as the determination of uncertainties. Additionally, the derivation of the relevant aerosol optical properties is also included in the algorithm. The algorithm was developed through the analysis of laboratory and field data, accompanied by a rigorous data quality control process. Further details can be found in the supplementary information of Teri et al. (2022) (Section S1.4) and Teri et al. (2024) (Section S2). Both supplementary information are included in the appendices of this thesis.

2.2 Tri-color Absorption Photometer (TAP)

The Tri-color Absorption Photometer (TAP) is a commercially available instrument developed by Brechtel (Brechtel, 2016). It measures the light transmission at three wavelength (blue 467 nm, green 528 nm, and red 652 nm) through a filter on which aerosol particles are meanwhile collected. Thus, it allows the determination of the particle absorption coefficient σ_{ap} , the fraction of light absorbed by particles per unit length.

The particle absorption coefficient is an important aerosol property because it quantifies the light absorption by the particles, which affects the Earth’s radiation budget, warming the atmosphere. When available at several wavelengths, it can be used to determine other useful intensive aerosol properties such as the absorption Ångström exponent and the single-scattering albedo, which can also have a spectral dependency.

Figure 2.4 show the A-HEAD rack, containing the TAP and Aurora 4000 instruments of the University of

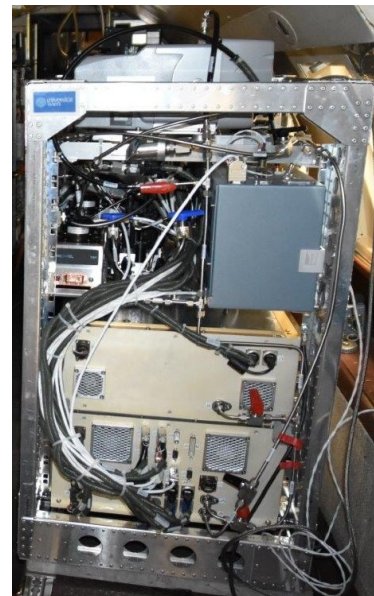


Figure 2.4: TAP and Aurora 4000 instruments during the A-LIFE field experiment. (Credit: B. Weinzierl, www.a-life.at).

Vienna, on board of the Falcon aircraft during the A-LIFE field experiment. The instruments were both placed behind the same external bypass system, and vacuum pump. In addition, the TAP was connected to a flow controller (MFM, Alicat scientific), keeping a constant volume flow through the instrument, despite the pressure change at different altitudes.

The TAP is a compact instrument designed to facilitate the replacement of the 47 mm glass-fibre filter. Unlike other absorption photometers, it employs ten solenoid valves to cycle through eight sample spots and two reference spots. O-rings are used to clamp the filter material, preventing leakage between spots. The sample air flows through one of the sample spots and then, once free of particles, through one of the two reference spots. Light-emitting diodes (LEDs) are used as a light source, and the light transmitted through both spots is measured simultaneously, monitoring and compensating in real-time light transmission changes not related to aerosol particles, including filter variations, flow rate changes, or light source intensity fluctuations. Further details about the TAP instrument and its components are available in the user manual (Brechtel, 2016). The following section provides a more detailed explanation of the measurement principle of absorption photometers.

2.2.1 Measurement principle

The Tri-color Absorption Photometer (TAP, Brechtel) determines the particle absorption coefficient using the same measurement principle as other filter-based absorption photometers, such as the Particle Soot Absorption Photometer (PSAP, Radiance Research) and the Aethalometer (AE33, Aerosol Magee Scientific). These instruments are usually preferred over in situ measurements such as photoacoustic and extinction minus scattering techniques because they are less expensive and easier to operate. A comprehensive overview of the absorption measurement techniques and their advantages and disadvantages can be found in Moosmüller et al. (2009).

Figure 2.5 shows a schematic of the measurement principle of filter-based absorption photometers. These instruments operate by collecting aerosol particles on a filter spot area, A , at a known volume flow rate, Q , and measuring the change in light transmission, $I(t)$, through the filter. For a given time interval, Δt , the light transmission change, $I(t)/I(t - \Delta t)$, is related to the attenuation coefficient of the particle-filter bulk, σ_{atn} , via a relationship in the form of the Beer-Lambert law:

$$\frac{I(t)}{I(t - \Delta t)} = e^{-\sigma_{atn}(t) \frac{Q\Delta t}{A}} \quad (2.1)$$

The attenuation coefficient of the particle-filter bulk is related to the particle absorption coefficient under a critical assumption: that the light absorption by aerosol particles in an air parcel is equivalent to the light absorption through a filter on which the aerosol particles of that air parcel are deposited. However, this assumption does not account for the optical properties of the filter material itself and for interactions between the optical properties of the particles and the filter, such as multiple

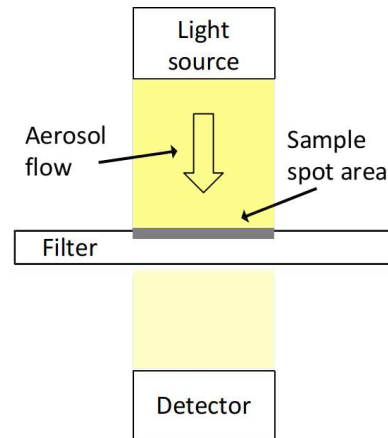


Figure 2.5: Scheme of the measurement principle of filter-based absorption photometers.

scattering. Consequently, an empirical filter loading correction factor, dependent on the light transmission change with respect to the particle-free filter, must be applied. Furthermore, the contribution of particle scattering to the change in light transmission must also be considered.

More details on the measurements principles of filter-based absorption photometers are available in instrument user manuals (e.g., Brechtel, 2016; Springston, 2018), and in several publications (e.g., Bond et al., 1999; Ogren, 2010; Müller et al., 2014; Davies et al., 2019).

2.2.2 Corrections for filter loading and particle scattering

A disadvantage of filter-based absorption photometers is the necessity of correcting the measurements to take into account the absorption enhancement resulting due to multiple scattering within the filter matrix and the reduction in light transmittance due to particle scattering. While both corrections depends on the properties of the filter medium, the absorption enhancement depends on the amount of particles on the filter (i.e., the filter loading) and the reduction in light transmittance due to particle scattering requires simultaneous additional scattering measurements, e.g., by a nephelometer.

Various empirical parametrizations for the filter loading and particle scattering corrections are available in the literature. A simple correction, assuming a linear response of the instrument to the particle scattering and absorption coefficients, was developed by Bond et al. (1999) and revised by Ogren (2010). This correction has a high uncertainty, which reaches up to 100 % for weakly absorbing aerosols (e.g., when the single-scattering albedo is larger than 0.97), as shown by Ogren et al. (2017). Furthermore, it was derived for the green wavelength (574 nm), and is not directly applicable to other wavelengths.

An alternative empirical correction method, initially proposed by Virkkula et al. (2005) and later revised by Virkkula (2010), is applicable to three wavelengths in the visible spectrum (467, 530, and 660 nm). This correction method introduces a logarithmic dependence of the filter loading correction factor as the transmission increases and also accounts for a dependency on the single-scattering albedo. The instrument response to the scattering coefficient is considered to be linear.

A more complex algorithm, which considers a two-stream radiative transfer model and experimental calibrations, was developed by Müller et al. (2014). However, the application of this method requires additionally the asymmetry parameter and a non-trivial code.

An evaluation of these three correction methods applied to the TAP data for ambient aerosols is available in Davies et al. (2019). Among them, the correction by Müller et al. (2014) emerged as the best performing method. The approach proposed by Virkkula et al. (2005) and Virkkula (2010) also provided significant improvements compared to the method by Bond et al. (1999) and Ogren (2010).

All these correction schemes were derived for the glass-fibre Pallflex E70-2075W filters, and their application is restricted to this specific filter material. However, Pall Corporation discontinued the production of the Pallflex E70-2075W filters. The only valid filter material is Azumi M371. Nevertheless, a complete characterization with relative correction is not available yet. Fortunately, throughout this thesis, the well-characterized Pallflex E70-2075W filters were used.

2.2.3 Calibrations

It is not possible to calibrate the filter-based absorption photometers using an absorbance standard. However, to obtain an accurate value of the particle absorption coefficient, it is necessary to determine the accurate values of the sample volume flow, of the filter spot area, and of the light transmitted through the filter material without particles. Thus, the volume flow must be calibrated using a bubble flow meter. The spot area must be carefully determined after the measurements. A baseline measurement range for the instrument must be established for each new filter. This is accomplished by measuring the light transmitted through all spots using particle-free air. Further details on this procedure, which is known as the white filter check, can be found in the TAP user manual (Brechtel, 2016).

2.2.4 Data quality control and processing

Within the work for this thesis, an algorithm for processing data from the TAP was developed, and the code was written in R (R Core Team, 2024). The algorithm was developed by analyzing laboratory and field data and includes all the necessary steps for converting the raw data to the final form, including data processing, quality controls, calibrations, filtering and plotting.

The data processing involves the calculation of the attenuation coefficient of the particle-filter bulk from the raw transmissions, as well as the application of the corrections for filter loading and particle scattering. Throughout this thesis, the two empirical corrections available, the Bond et al. (1999) and Ogren (2010) correction and the Virkkula et al. (2005) and Virkkula (2010) correction, were both used. Additionally, the derivation of the relevant aerosol optical properties is also included in the algorithm.

Data quality control includes inspections of the white filter check measurements and transmission throughout the measurement time series and comparing the derived particle absorption coefficients with other relevant measured quantities. The calibrations include determining the spot area and calibrating the sample volume flow. A limitation of filter-based methods for aircraft measurements is the sensitivity to variations in the inlet pressure (Petzold et al., 2013). Consequently, only measurements at constant altitudes are used.

Details on the TAP operation, calibrations, data quality control and processing throughout this thesis can be found in the supplementary information of Teri et al. (2022) (Section S1.1) and Teri et al. (2024) (Section S1). The supplementary information of both publications is included in the appendices of this thesis.

2.3 Additional measurement instruments and simulation tools

In addition to the Aurora 4000 and TAP instruments, other instruments and tools were used throughout this thesis, in laboratory and simulated experiments. The following sections briefly describe the working principles of these instruments and tools.

2.3.1 Cavity Attenuated Phase Shift Extinction (CAPS PM_{ext}) Monitor

A Cavity Attenuated Phase Shift Extinction (CAPS PM_{ext}) monitor developed by Aerodyne Research was deployed in laboratory experiments within this thesis for the measurement of the particle extinction coefficient at a wavelength of 530 nm.

This instrument utilizes two high-reflectivity mirrors to artificially extend the optical path through a cell of approximately 30 cm, producing optical paths spanning several kilometres. This feature makes the instrument sensitive to the particle extinction coefficient, even at low particle concentrations. A LED light source generates a square-modulated wave, which is detected as distorted at the opposite end of the sample cell. The phase shift of the distorted wave is a function of the fixed instrument properties and particle extinction coefficient.

Further information on the instrument and its measurement principle can be found in the CAPS PM_{ext} user manual (Sedlacek, 2016). Details on the operation and data analysis algorithm for the Aerodyne CAPS PM_{ext} within this thesis are provided in the supplementary information of Teri et al. (2022) (Section S1.3) attached in the appendix of this thesis.

2.3.2 Instruments for measuring aerosol microphysical properties

In laboratory experiments within this thesis, the aerosol microphysical properties were measured using two optical spectrometers: the Ultra High Sensitivity Aerosol Spectrometer (UHSAS) from Droplet Measurement Technologies (DMT) Incorporated and the Optical Particle Sizer (OPS Model 3330) from TSI Incorporated; and a Condensation Particle Counter (CPC Model 3772) from TSI Incorporated.

The DMT UHSAS measures the particle number size distribution in the diameter range from 60 nm to 1 μm , while the TSI OPS measures it from 0.3 μm up to 10 μm . Both instruments measure the light scattered by single particles when illuminated by a laser beam. Light pulses registered by the detector are counted and sorted into different classes according to their pulse height. Calibrations using monodisperse polystyrene latex (PSL) particles of different known sizes allow for the different pulse height classes to be related to size classes. The particle diameter determined in this way is called the optical equivalent diameter, which is the diameter of a sphere with the same refractive index of the calibration material.

The two instruments use different wavelengths, detector geometries and systems to detect and process the particle signals. The TSI OPS uses a vertically polarized laser beam with a wavelength of 660 nm and an elliptical mirror to collect the light scattered over solid angles from 30° to 120° and to focus it onto a photodiode. The DMT UHSAS uses a vertically polarized laser beam with a wavelength of 1054 nm and two detection systems collecting the light scattered over solid angles of 33° – 75.2° and 104.8° – 147°. The two detection systems utilize an avalanche photodiode and a PIN photodiode to detect the particles in the lower and upper size ranges, respectively. Additional information on these instruments can be found in the DMT UHSAS user manual (DMT, 2013) and in the TSI OPS user manual (TSI, 2013).

Within this thesis, the particle counting efficiency of both instruments was estimated using the TSI CPC 3772. The TSI CPC 3772 measures the number concentration of aerosol particles over a wide concentration range ($0 - 10^4 \text{ cm}^{-3}$). In the instrument, sample aerosol particles are enlarged by passing through a butanol saturator and a condenser, and subsequently, they are

optically detected. Since particles are grown, even tiny particles, down to 10 nm in diameter, can be detected. Further information on the TSI CPC 3772 and its measurements principle can be found in the user manual (TSI, 2007).

A detailed description of the data analysis algorithm for these three instruments within this thesis is provided in the supplementary information of Teri et al. (2022) (Section S1.5) attached in the appendix of this thesis.

2.3.3 Tools for optical simulations

The optical properties of aerosol particles can be obtained using modeling tools based on scattering theory, if the microphysical properties of the aerosol particles are known. Indeed, scattering theory provide solutions to the Maxwell equations for the interaction of electromagnetic radiation with particles. The necessary input parameters are the wavelength of the radiation and the shape, size and refractive index of the particles.

For spherical particles, an analytical solution of Maxwell equations can be derived using the classical Lorentz-Mie theory (Mie, 1908; Horvath, 2009), which is limited to the scattering of infinite electromagnetic plane waves by homogeneous, isotropic, spherical particles. For particles with a non-spherical shape, a solution can be derived using the T-Matrix (Mishchenko and Travis, 1998), a method efficient for randomly oriented rotationally symmetric particles. Other methods for calculating the optical properties of non-spherical particles include approximate numerical techniques, such as geometric optics (e.g., Yang et al., 2007), efficient for particles large compared to the incident wavelength, and the discrete dipole approximation (Yurkin and Hoekstra, 2011), often used for arbitrarily-shaped particles but computationally demanding.

Various optical modeling tools, based on the different methods, were employed in this thesis to conduct optical simulations. The optical simulation within this thesis serves mainly for support and comparison to the optical measurements. For instance, the aerosol optical properties were calculated using the measured size distribution and were then compared to the values obtained from the measurements using the Aurora 4000 and TAP instruments. The following sections provide a brief overview and references to the optical modeling tools used.

Miepython Miepython is a pure Python module developed by S. Prahl using Mie theory, and based on the algorithm described by Wiscombe (1980). It provides functions to calculate the extinction and scattering efficiency and the scattering at various angles for non-absorbing or partially absorbing spheres. The code can be found at <https://github.com/scottprahl/miepython>, while extensive documentation is available at <https://miepython.readthedocs.io>. This code has been used within this thesis to perform Mie calculations, especially to obtain the optical properties of single spherical particles.

Cops Cops (Collection of tools for Optical Particle Spectrometers) is a Mie code written in Python based on Bohren and Huffman (2008) and developed at the University of Vienna by A. Walser, and not publicly available yet. The code include a toolkit for theoretical scattering calculation of various Optical spectrometer. This code has been used within this thesis to calculate the scattering signals of the TSI OPS and DMT UHSAS for spherical particles.

ADDA ADDA is an open source C software package developed by Yurkin and Hoekstra (2011). It uses the discrete dipole approximation (DDA) to calculate the scattering and absorption of electromagnetic waves by particles of arbitrary shape and composition. The code is available at <https://github.com/adda-team/adda>. This code was used in collaboration with J. Gasteiger during this thesis work to simulate the scattering signals of the TSI OPS for irregularly shaped particles. Details and results on these calculations are available in the supplementary information of Teri et al. (2022) available in the appendix of this thesis (see, for example, sections S2, S3 and S4 as well as Figure S2 and Figure S3).

MOPSMAP The MOPSMAP package (Modeled Optical Properties of enSeMbles of Aerosol Particles) is a computationally fast optical modeling tool developed by Gasteiger and Wiegner (2018). It includes a wide data set of pre-calculated optical properties, a Fortran code to calculate the optical properties of aerosol ensembles, and a user-friendly web interface (<https://mopsmap.net/>). This thesis used this tool to calculate the optical properties of aerosol particles measured in laboratory experiments using the binned size data provided by the TSI OPS and DMT UHSAS as input. Details are reported in Teri et al. (2022) available in the appendices of this thesis (see section 4.2 and the supplement information section S1.8). Additionally, this tool was used to extend the measured optical properties to cover the full ambient size distribution. Details are available in the supplementary information of Teri et al. (2024) available in the appendix of this thesis (section S3).

2.4 Experiments

Three main experiments were conducted to address the research questions outlined in section 1.6: two closure experiments—a laboratory experiment and a simulated experiment—and the A-LIFE field experiment.

Closure experiments, which derive the same property using different techniques, aim to characterize specific aerosol properties and minimize the measurement uncertainties (Weinzierl et al., 2017). The laboratory and simulated closure experiments aimed to investigate and reduce measurement uncertainties associated with the Aurora 4000 nephelometer (research questions 1 and 2). The laboratory closure experiment focused on measurements of re-suspended mineral dust particles. The simulated closure experiment aimed to support the interpretation of the laboratory closure experiment results by clarifying irregular shape effects via optical simulations and extending the analysis to include different particle sizes, refractive indices, and shapes.

The A-LIFE field experiment provided a comprehensive data set of the aerosol properties in the eastern Mediterranean, including optical properties from the Aurora 4000 nephelometer and the TAP, as well as microphysical properties. This data set allowed for a detailed characterization of mineral dust layers and the impact of increasing pollution content on their properties (research questions 3-5).

2.4.1 Laboratory closure experiment

One of the aims of this thesis is to evaluate the quality of the Aurora 4000 nephelometer measurements (research question 1 and 2). This evaluation is crucial, as the instrument was used also during the A-LIFE field experiment to measure the scattering properties of mineral dust.

Accurately determining the particle scattering coefficient of mineral dust particles—which are predominantly in the coarse mode and irregularly shaped—is challenging for integrating nephelometers. To obtain accurate particle scattering coefficients across the complete angular range, an angular correction has to be applied to integrating nephelometer data. The angular correction compensates for the impossibility of measuring the light scattered by particles in the extreme forward and backward directions, as well as deviations from ideal Lambertian illumination (see also section 2.1.3). Since larger particles scatter more in the forward direction, the angular correction is highly sensitive to the particle size (e.g., Heintzenberg and Charlson, 1996). Additionally, the influence of the irregular shape is poorly understood. Thus, existing methods for calculating the angular correction require a re-evaluation, especially for irregularly shaped particles in the coarse mode (Schladitz et al., 2009, 2011; Quirantes et al., 2008; Sorribas et al., 2015).

To address this goal, a laboratory closure experiment was designed, prepared, and executed as part of this thesis. This experiment validated the angular sensitivity function of the Aurora 4000 nephelometer derived by Müller et al. (2012), and evaluated and compared different methods to calculate the angular correction with a focus on mineral dust aerosols.

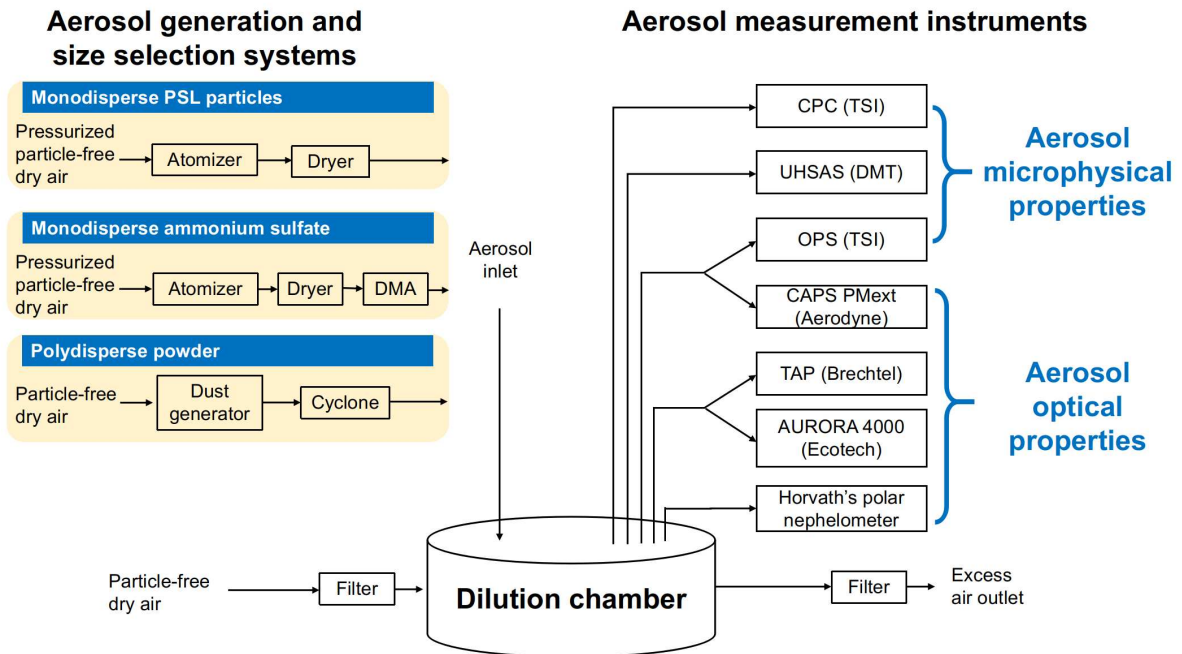


Figure 2.6: Scheme of the laboratory closure experiment set-up. Aerosols of known size and composition were generated and introduced into a dilution chamber. The upper left part of the figure shows aerosol generation and size selection systems for PSL particles, ammonium sulfate, and polydisperse powders (mineral dust, silica dust, and volcanic ash). The upper right shows instruments measuring aerosol microphysical properties (e.g., particle number concentration and size distribution) and optical properties (e.g., particle extinction, absorption, and scattering coefficients, and angular distribution of scattered light). (Adapted from Figure 1 of Teri et al. (2022).)

Figure 2.6 shows the set-up scheme of the laboratory closure experiment. Aerosol of known size and composition, including polystyrene latex (PSL) particles, ammonium sulfate (AS), and polydisperse powders (mineral dust, silica dust, and volcanic ash), were generated, injected into a dilution chamber, and measured by multiple instruments, simultaneously. The aerosol microphysical properties, including the particle number concentration and particle size distribution, were measured by the TSI CPC, the DMT UHSAS, and the TSI OPS (see section 2.3.2). The aerosol optical properties, including the particle scattering, absorption, and extinction coefficients, were measured by the Ecotech (now Acoem) Aurora 4000 nephelometer, the Brechtel TAP, and the Aerodyne CAPS PMext (see sections 2.1, 2.2, and 2.3.1, respectively). Additionally, the Horvath’s polar nephelometer, a custom-built polar nephelometer (Horvath, 2015; Horvath et al., 2018), measured the angular distribution of the light scattered by particles at a wavelength of 532 nm.

Results from this closure laboratory experiment allowed to compare different angular corrections (including C_{SAE} , C_{polar} , $C_{phase,OPS}$). The particle scattering coefficients measured with the Aurora 4000 nephelometer and corrected with the different angular corrections were compared with a reference value, which was obtained the difference between the extinction coefficient and the absorption coefficient, measured by the Aerodyne CAPS PMext and the Brechtel TAP, respectively (see Figure 3 of Teri et al. (2022)).

A detailed description of this laboratory closure experiment, its data analysis, and its results are available in Teri et al. (2022) (see sections 4.1, 4.2, and 4.3, and Figure 2 for a flow chart of the data analysis).

Validation of the Aurora 4000 angular sensitivity function

The closure laboratory experiment also allowed the validation of the Aurora 4000 angular sensitivity function. This function accounts for the instrument’s angular non-idealities (see also section 2.1.3), and is parametrized to account for deviations from a sine function (Müller et al., 2012). Although different angular corrections exist to derive the particle scattering coefficient from the Aurora 4000 nephelometer measurements, they all rely on the Aurora 4000 angular sensitivity function. Therefore, its validation is crucial to ensure the quality of the Aurora 4000 nephelometer measurements.

The microphysical properties measured during the laboratory experiment served as input for simulating the aerosol optical properties, which were then compared with the measurements. A comparison between simulated and measured extinction coefficients is provided in the supplementary information of Teri et al. (2022) (see section S2 and Figure S1). The excellent agreement demonstrates the consistency between the particle size distribution and the aerosol optical property measurements.

Figure 2.7 shows the comparison between the simulated and measured Aurora particle scattering coefficients. Measurements were performed during the laboratory closure experiment using monodisperse particles of polystyrene latex (PSL) with diameters ranging from 200 nm to 1.8 μm , and ammonium sulphate (AS) with diameters ranging from 200 nm to 850 nm. The measured particle coefficients are values from the Aurora 4000 nephelometer without angular correction. The simulated particle scattering coefficients were derived via optical simulations

based on measured microphysical properties, and the Aurora 4000 angular sensitivity function (Müller et al., 2012).

The linear regression analysis forced through the origin shows excellent agreement between simulations and measurements, within $3 \pm 2\%$ and coefficient of determination (R^2) greater than 0.99, indicating less than 1% variability. This result indicates that the parametrization of the angular sensitivity function proposed by Müller et al. (2012) is valid also for the Aurora 4000 nephelometer unit of the University of Vienna, and thus, corroborates the quality of the Aurora 4000 nephelometer measurements.

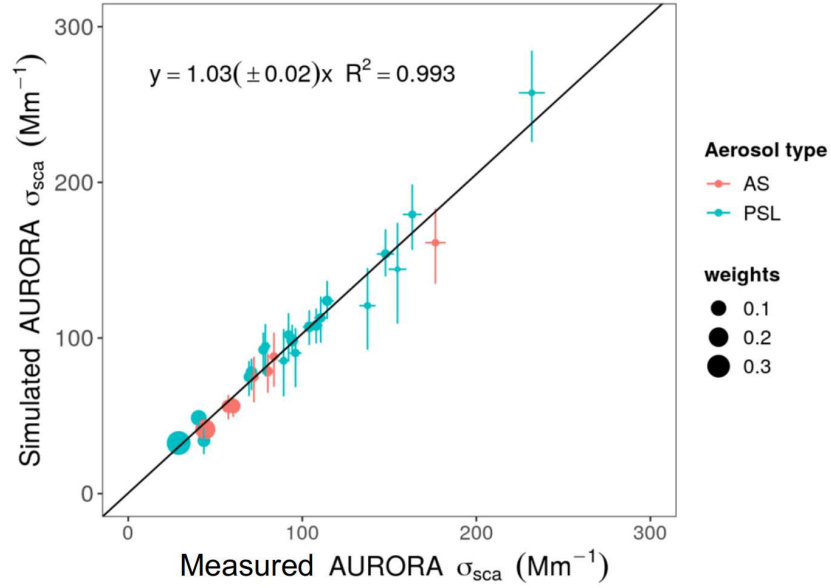


Figure 2.7: Comparison of simulated and directly measured Aurora particle scattering coefficients. The colour of the points indicates the aerosol type: blue for polystyrene latex (PSL) particles and red for ammonium sulfate (AS). The size of the dots indicates the weights (i.e., inverse of the simulation result variance) used for the linear regression analysis. Reported error bars are the instrument error of about 2 – 3% for the Aurora 4000 and the 2.5th and 97.5th percentiles for the simulations. Results from this experiment are published in Teri et al. (2022), but this specific plot was not included in the manuscript.

2.4.2 Simulated closure experiment

A simulated closure experiment was conducted to complement the laboratory closure experiment, aiming to improve understanding of the particle scattering coefficient derivation from the Aurora 4000 nephelometer (research questions 1 and 2). Within the laboratory experiment, different methods to calculate the angular correction—compensating for the nephelometer non idealities (see section 2.1.3)—were tested, focusing on coarse-mode irregularly shaped mineral dust particles (see section 2.4.1).

Two angular corrections performed relatively well for mineral dust laboratory measurements (with an average deviation of 7%): C_{SAE} , using as input parameter the scattering Ångström exponent, and $C_{phase,OPS}$, using the phase function derived via optical simulation from the particle size distribution measured with the TSI OPS. These results were surprising because both angular corrections assume the particle shape to be spherical. Previous studies by Schladitz et al. (2009, 2011) suggested that particle shape could be a major cause of observed discrepancies. Additionally, a simple approach was used to calculate the $C_{phase,OPS}$ angular correction: the refractive index of the calibration material ($1.59 + i0.00$) was assumed for both the particle size distribution retrieval and the optical calculations. The good performance of $C_{phase,OPS}$ suggested that using the refractive index of the calibration material might compensate for discrepancies related to the effect of irregular shape.

These unexpected findings raised questions regarding the effect of the irregular shape and the reliability of C_{SAE} and $C_{phase,OPS}$ for different particle sizes, refractive indices and shapes. The simulated closure experiment was designed to answer the following questions:

- (i) What is the effect of the irregular shape on the nephelometer angular truncation?
- (ii) What are the uncertainties of the angular correction based on the scattering Ångström exponent (C_{SAE}) for different particle sizes, refractive indices and shapes?
- (iii) What are the uncertainties of the angular correction derived from an optical particle spectrometer ($C_{phase,OPS}$) using different approaches for different particle sizes, refractive indices and shapes?

Figure 2.8 shows a simplified version of the flow chart for the overall simulated closure experiment (for more details, see Figure 4 of Teri et al. (2022)). This experiment compared distinct simulations of the angular correction: the "original" and the "derived" angular correction, performed for the same randomly selected aerosol parameters. The "original" angular correction was calculated using the Aurora 4000 angular sensitivity function (Müller et al., 2012). The "derived" angular correction was obtained based on instrumental responses and assumptions required when the aerosol is unknown (e.g., in a field experiment).

The effect of irregular shape on the nephelometer angular error (question i) was evaluated by comparing the "original" angular correction—calculated using the discrete dipole approximation (ADDA code from Yurkin and Hoekstra, 2011)—with the one calculated with Mie theory (see Figure 5 of Teri et al. (2022)).

The uncertainties of C_{SAE} (question ii) were evaluated by comparing the "original" angular correction—calculated for different particle sizes, refractive indices and shapes—with C_{SAE} derived

by simulating the SAE. Similarly, the uncertainties of $C_{\text{phase,OPS}}$ (question iii) were evaluated by comparing the "original" angular correction with the OPS-derived angular correction. The OPS-derived angular correction was obtained by simulating the OPS response and applying different approaches (e.g., assuming different refractive indices, see caption of Figure 2.9). Figure 2.9 shows a flow chart specific for the OPS-derived angular correction, including the individual steps for the simulations of the TSI OPS response.

Results of this simulated closure experiment allowed to support the interpretation of the laboratory closure experiment results for irregularly shaped particles (research question 1), and extend the evaluation of C_{SAE} and $C_{\text{phase,OPS}}$ to various particle sizes, refractive indices, and shapes (see Figures 5 and 6 of Teri et al. (2022)). Thus, results from the laboratory and simulated closure experiments, in combination with other literature results, allowed for the formulation of guidelines to establish the most appropriate angular correction in several situations answering the research question 2 (see Figure 8 of Teri et al. (2022)).

A detailed description of this simulated closure experiment, its data analysis, and its results are available in Teri et al. (2022) (see section 5 and S4 in the supplementary information).

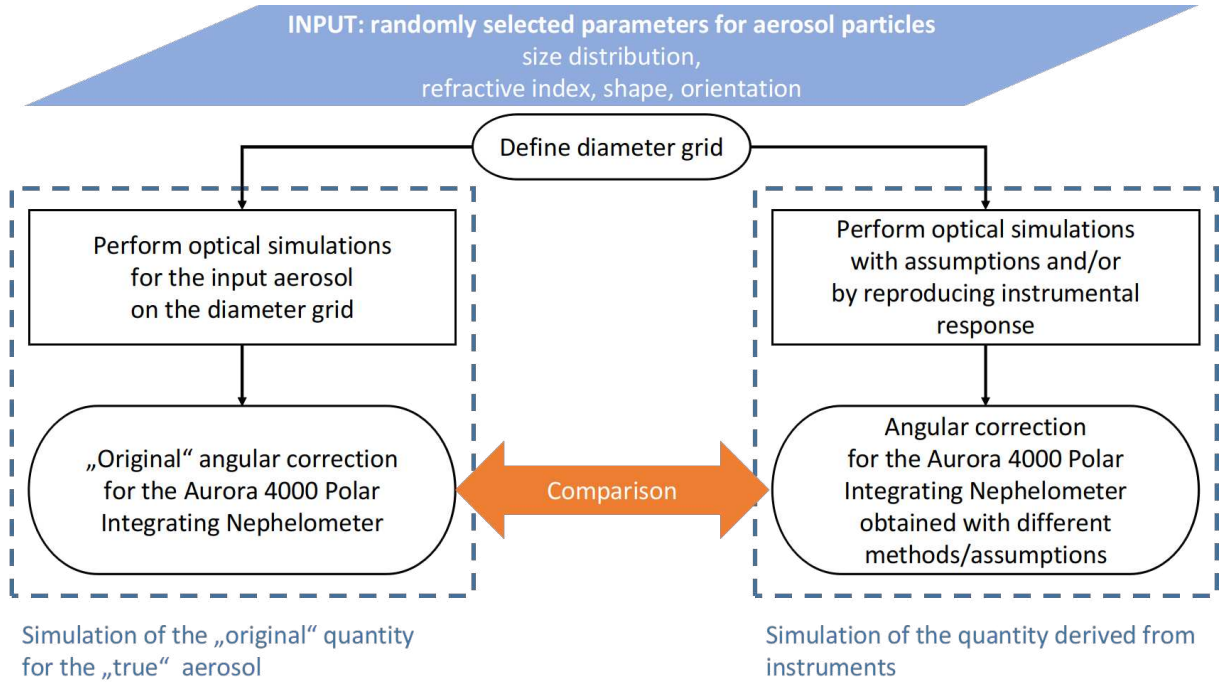


Figure 2.8: Simplified flow chart for the simulated closure experiment. The experiment compared values of the angular correction, obtained for the same "true" aerosol (using randomly selected input parameters, as indicated in the blue box at the top) and diameter grid, but different optical simulation procedures. The "original" angular correction—expected for the "true" aerosol—was simulated, as indicated on the left side of the flow chart. The "derived" angular correction was simulated considering specific instrumental responses and assumptions necessary when the measured aerosol is unknown (e.g., in a field experiment), as indicated on the right side of the flow chart. A more detailed flow chart including details for answering the question (iii) is reported in Figure 2.9. (For more details see Figure 4 of Teri et al. (2022)).

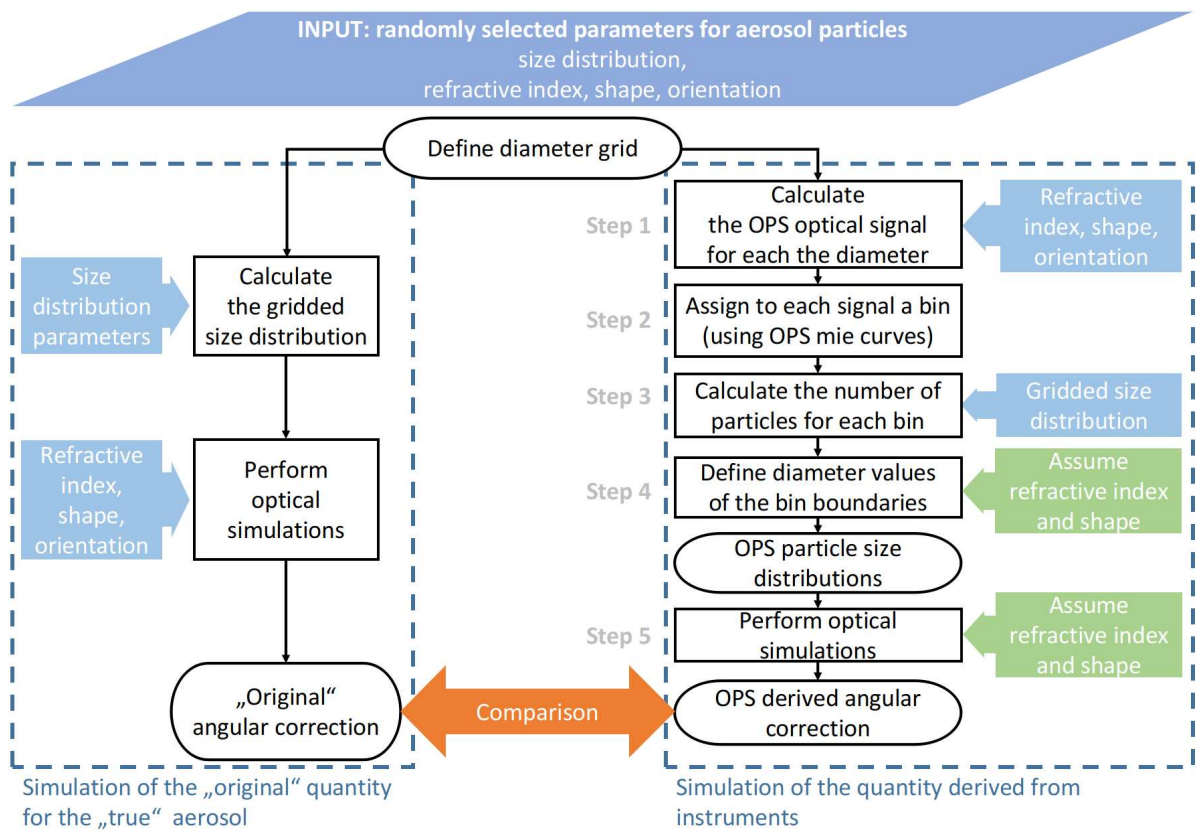


Figure 2.9: Flow chart for the OPS-derived angular correction (question iii) in the simulated closure experiment. The flow chart shows the methodology used to investigate the uncertainties in the Aurora 4000 nephelometer angular correction derived from OPS measurements. The left side details the simulation of the "original" angular correction, while the right side outlines the steps for simulating the TSI OPS response and deriving the angular correction. Blue boxes indicate the required input at the individual step.

Green boxes indicate the assumptions needed for calculating the OPS-derived angular correction. Three approaches using different assumptions were tested: (a) using the refractive index of the calibration material and spherical shape for both the size distribution retrieval (step 4) and the optical simulations (step 5); (b) using the refractive index of the calibration material and spherical shape for step 4, but the refractive index of the "true" aerosol and spherical shape for step 5; (c) using the refractive index of the "true" aerosol and spherical shape for both steps 4 and 5.

(For more details see Figure 4 of Teri et al. (2022)).

2.4.3 A-LIFE field experiment

The "Absorbing aerosol layers in a changing climate: ageing, lifetime and dynamics" (A-LIFE) project is a European Research Council (ERC)- funded project that aims to study the properties of light-absorbing aerosol mixtures, mainly mineral dust and black carbon mixtures.

During the A-LIFE field experiment in the spring of 2017, aerosol data were collected with sophisticated in situ and remote sensing instruments integrated into the German Aerospace Center's (DLR) research aircraft Falcon 20 E-5. Instrumentation included aerosol size distribution measurements between 5 nm and 930 μm , a wind lidar and meteorological sensors. The Aurora 4000 nephelometer and the TAP measured the interaction between particles and radiation, providing aerosol optical properties. The airborne measurements during A-LIFE were also complemented by ground-based in situ and remote sensing instrumentation supports the A-LIFE project. Further information on A-LIFE is available at <http://a-life.at>.

The aircraft base at the Paphos airport in Cyprus provided access to the eastern Mediterranean, a region of unique interest due to the convergence of different aerosol types. In particular, the region is affected by mineral dust layers from the two nearby deserts, the Arabian peninsula in the Middle East and the Saharan desert in Northern Africa and by anthropogenic pollution (e.g., Dayan et al., 2017). During the A-LIFE field experiment, several Arabian and Saharan dust outbreaks were measured in mixtures with local and transported anthropogenic pollution (Weinzierl et al., in preparation, 2025). Figure 2.10 shows the flight path of the 22 research flights performed by DLR research aircraft Falcon 20 E-5. As a background, a natural colour satellite image from April 27, 2017, was used to show the influence of the regional aerosol loading by mineral dust. This Figure was taken from Figure 1 of Teri et al. (2024).

In this thesis, measurements collected in the eastern Mediterranean during the A-LIFE field experiment were used, to investigate the effect of pollution within mineral dust layers on aerosol optical properties. For this, data of the aerosol optical properties measured during the A-LIFE field experiment with the Aurora 4000 nephelometer and the TAP instrument have been analyzed together with the measured microphysical properties. The aim of the data analysis was to investigate how different quantities of pollution affect the intensive aerosol optical properties of mineral dust layers, and whether these effect varies depending on the mineral dust's source region (research question 3). The details of this analysis and its results are reported in sections 2 and 3 of Teri et al. (2024), which is attached in the appendix of this thesis. The paper also discusses the implications of these analysis results for the identification of mineral dust layers using the Aurora 4000 nephelometer and TAP measurements (research question 4) and for the direct radiative effect of mineral dust layers (research question 5).

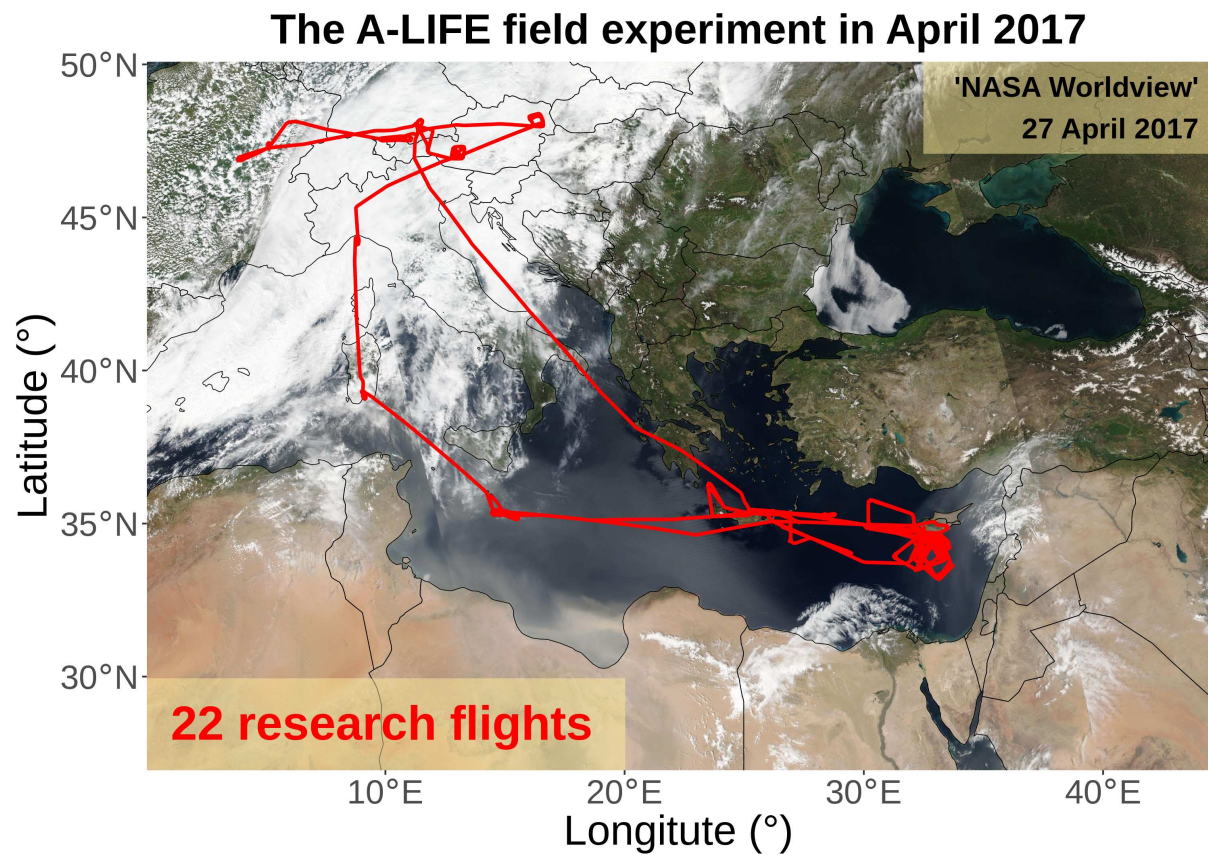


Figure 2.10: The A-LIFE field experiment in April 2017. The red line indicates the flight path of the 22 research flights performed during the A-LIFE field experiment onboard of the Deutsches Zentrum Luft- und Raumfahrt (DLR) research aircraft Falcon 20 E-5. The background is a natural color satellite image (26.9°N-50.1°N, 0.0°E-44.5°E) from 27 April 2017. (Source: "NASA-Worldview", <https://worldview.earthdata.nasa.gov/>). This figure was taken from Figure 1 of Teri et al. (2024).

Publications

3. Paper I: Teri et al. (2022)

Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections

Abstract

Aerosol particles in the atmosphere interact with solar radiation through scattering and absorption. Accurate aerosol optical properties are needed to reduce the uncertainties of climate predictions. The aerosol optical properties can be obtained via optical modeling based on the measured particle size distribution. This approach requires knowledge or assumptions on the particle refractive index and shape. Meanwhile, integrating nephelometry provides information on the aerosol scattering properties directly. However, their measurements are affected by angular non-idealities, and their data need to be corrected for angular truncation and illumination to provide the particle scattering coefficient. We performed an extensive closure study, including a laboratory and a simulated experiment, aiming to compare different nephelometer angular truncation and illumination corrections (further referred to as “angular corrections”). We focused on coarse-mode irregularly shaped aerosols, such as mineral dust, a worldwide abundant aerosol component. The angular correction of irregular particles is found to be only $\sim 2\%$ higher than the angular correction of volume-equivalent spheres. If the angular correction is calculated with Mie theory, the particle size distribution is needed. Our calculations show that if the particle size distribution is retrieved from optical particle spectrometer measurements and the irregular shape effect is not considered, the angular correction can be overestimated by about 5% and up to 22%. For mineral dust, the traditional angular correction based on the wavelength dependency of the scattering coefficient seems more accurate. We propose a guideline to establish the most appropriate angular correction depending on the aerosol type and the investigated size range.

Bibliographic Information

Teri, M., Müller, T., Gasteiger, J., Valentini, S., Horvath, H., Vecchi, R., Bauer, P., Walser, A., and Weinzierl, B.: Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections, *Atmos. Meas. Tech.*, 15, 3161–3187, <https://doi.org/10.5194/amt-15-3161-2022>, 2022.

Author's contribution

I designed and wrote this publication, incorporating contributions and comments from co-authors. All figures and tables, including those in the supplement were prepared by myself, except for Figures S2 and S3 which were provided by Dr.J.Gasteiger. This publication presents results from laboratory and simulated closure experiments that I conceptualized together with my supervisor and co-authors. I set-up and tested the laboratory experiment, operated the instruments (with the exception of "Horvath's polar nephelometer"), and performed the experiment together with co-authors. I developed the data analysis algorithm and conducted data quality assurance of most instruments (Aurora 4000 nephelometer, TAP, CAPS PM_{ext}, TSI OPS 3330, UHSAS, CPC). The optical simulations for the laboratory closure experiment analysis were performed by myself using MOPSMAP. For the simulated experiment, I carried out optical simulations for homogeneous spherical particles using the Python modules Miepython and Cops(see thesis section 2.3.3). Simulations for irregularly-shaped particles were conducted by Dr.J.Gasteiger using the code ADDA, though I was actively involved in the scientific discussions related to the irregularly-shaped particles. Scientific discussions with co-authors led to refinements in data analysis and interpretation. I wrote the manuscript, coordinated the internal and external review process, prepared the responses to the reviewers with support of my supervisor and co-authors, incorporated the reviewer and co-author comments, and finalized the manuscript for publication.

Relevance for this dissertation

Direct in situ measurements of aerosol scattering properties are crucial to constrain the aerosol direct radiative effect. Integrating nephelometer data is affected by substantial uncertainties due to the impossibility of detecting the light scattered in the near-forward and backward directions. This publication provides advancements in understanding existing corrections for irregularly-shaped particles, reducing their uncertainties up to 13% and discussing the implications for closure studies (research questions 1). Guidelines for an appropriate angular correction for different particle sizes, refractive index and shapes were provided (research question 2). Additionally, the laboratory experiment was essential in characterizing the Aurora 4000 nephelometer instrument and ensuring the robustness of the scattering data of the A-LIFE data set. The results of this study were considered in finalizing the data collected during the A-LIFE field experiment. Thus, this publication contributed substantially to this thesis and also laid the foundation for Chapter 4.

Metrics

Since its release as a preprint on 21 December 2021, this publication has received a total of 3,891 views, including 2,694 HTML views, 1,110 PDF downloads, and 87 XML views. Additionally, the supplementary material has been downloaded 314 times. Citation data shows eight citations according to Crossref and nine citations on Google Scholar. (Last updated: 8 April 2025).

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4. Paper II: Teri et al. (2025)

Pollution affects Arabian and Saharan dust optical properties in the eastern Mediterranean

Abstract

Uncertainties in the direct radiative effect of mineral dust result from the variability in its optical properties, but are also influenced by mixing with anthropogenic aerosols ("pollution"), e.g., black carbon or sulfates. This study investigates the effect of mixing pollution with mineral dust from different source regions on the intensive optical properties. The Ångström exponents of scattering and absorption, the single-scattering albedo, and asymmetry parameter are determined from in situ measurements during the A-LIFE aircraft field experiment over the eastern Mediterranean, where Arabian and Saharan dust mixed with pollution. Our results show that all intensive dust optical properties change significantly with increasing pollution content. Interestingly, the differences between the optical properties of Arabian and Saharan dust are negligible. We discuss the implications of these results for the identification of mineral dust events and for their direct radiative effects. The pollution masks the mineral dust signal, which calls for caution when using Ångström exponents to identify mineral dust events. Furthermore, asymmetry parameter and single-scattering albedo change from pure to polluted mineral dust layers (e.g., at 525 nm, median values decrease from 0.67 to 0.56 and from 0.96 to 0.89, respectively). These changes have opposing effects on the shortwave direct radiative effect efficiency (i.e., the direct radiative effect per unit of aerosol optical depth), potentially canceling out each other. Still, the effect can be significant depending on surface albedo. In conclusion, quantifying pollution content in mineral dust layers is essential for accurately assessing their local direct radiative effect.

Bibliographic Information

Teri, M., Gasteiger, J., Heimerl, K., Dollner, M., Schöberl, M., Seibert, P., Tipka, A., Müller, T., Aryasree, S., Kandler, K., and Weinzierl, B.: Pollution affects Arabian and Saharan dust optical properties in the eastern Mediterranean, EGU sphere [preprint], <https://doi.org/10.5194/egusphere-2024-701>, 2024.

Author's contribution

I designed and wrote this publication, incorporating contributions and comments from co-authors. I prepared all figures and tables, including those in the supplement, except Figures 4, 5, S4, S5, and S6, which were provided by Dr. S. Aryasree and modified by myself. This publication presents results from the A-LIFE field experiment, an intensive aircraft field campaign conceptualized and coordinated by my supervisor. I participated in the A-LIFE field experiment as a master's student, contributing to the preparation, calibration, and performance testing of the University of Vienna's Aurora 4000 nephelometer and TAP instrument for airborne measurements. I also assisted with the instrument integration into the DLR research aircraft Falcon, and participated in research flights. During my PhD, I developed the data analysis algorithm, conducted the data quality assurance and processing for the Aurora 4000 and TAP instruments, and derived intensive aerosol optical properties. I conceptualized this publication through discussions with my supervisor. I performed the data analysis, combining optical properties derived by myself with microphysical properties provided by co-authors. I presented the preliminary results, coordinated the scientific discussion among co-authors, and refined the data analysis and interpretation based on the scientific discussions. I wrote the manuscript with input and contributions from co-authors. I coordinated the internal editing and revision process as well as the review process, prepared the responses to reviewers with support from co-authors, and finalized the manuscript for publication.

Relevance for this dissertation

This paper constitutes the scientific core of this dissertation. In situ measurements of aerosol optical properties in the eastern Mediterranean are investigated to assess the effect of mixing pollution and mineral dust. Particularly, the paper assessed the effect of increasing pollution within mineral dust layers from different source regions on intensive aerosol optical properties (research question 3). These properties are crucial for the characterization of mineral dust layers, especially in the presence of pollution. Indeed, they can provide near-real-time information on the aerosol type (research question 4) and are crucial for understanding the direct radiative effect of the mixed aerosol layers (research question 5). Additionally, the effect of pollution on the aerosol intensive optical properties of mineral dust layers with different origins was investigated (research question 5).

Metrics

Since it was released as a preprint on 10 April 2024, this publication has received a total of 841 views, including 529 HTML views, 185 PDF downloads, and 127 XML views. Additionally, the supplementary material has been downloaded 95 times. The preprint has two citations on Google Scholar. (Last updated: 8 April 2025).

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Conclusions

5. Summary and conclusions

The high variability in mineral dust optical properties leads to large uncertainties in its direct radiative effect. Mixed aerosol layers composed of both mineral dust and anthropogenic aerosol can form. These mixed aerosol layers can exhibit optical properties that differ significantly from those of the pure aerosol layers. In situ measurements of the aerosol optical properties are required for different locations and mineral dust source regions to improve the understanding of the impact on climate of these mixed aerosol layers.

This cumulative dissertation had the overall objective to improve the understanding of optical and radiative properties of mineral dust and its mixtures with pollution using in situ measurements of the aerosol optical properties. This effort involved laboratory measurements, instrument characterizations and calibrations, model simulations, algorithm developments, and in situ aircraft observations. This thesis was conducted in the framework of the "Absorbing aerosol layers in a changing climate: aging, lifetime, and dynamics" (A-LIFE, 2017) project, which had the overall aim of characterizing absorbing aerosol layers in the eastern Mediterranean, where mineral dust from different source regions co-exist with anthropogenic aerosols.

Figure 5.1 provides a schematic overview of this dissertation, illustrating the instrumentation utilized, the experiments involved, the algorithms developed, and the scientific impact. The core instruments are an Aurora 4000 Polar Integrating Nephelometer (Aurora 4000, Ecotech—now Aecom) and a Tri-color Absorption Photometer (TAP, Brechtel), which provide multi-wavelength particle scattering and absorption coefficients. These instruments were deployed during the A-LIFE field experiments, and also in complementary laboratory experiments. Additional instruments and optical simulations tools utilized in this thesis are listed in figure 5.1 (for more details, see section 2.3). Three primary experiments were involved: (i) a laboratory closure experiment, (ii) a simulated closure experiment, and (iii) the A-LIFE field experiment. The laboratory and simulated experiments constitute a comprehensive closure study, evaluating the quality of particle scattering coefficients measured by integrating nephelometers with a focus on mineral dust particles. The laboratory experiment involved measurements of different laboratory-generated aerosols—including dispersed soil samples—in a dilution chamber with extensive instrumentation. The simulated closure experiment used numerical simulations of instrumental responses to support the interpretation of laboratory findings extending them to different particle sizes, refractive indices, and shapes. The A-LIFE field experiment involved aircraft in situ observations of several properties of mineral dust-pollution mixtures. The resulting data set constitutes the basis for assessing the effects of mixing mineral dust and pollution on their optical and radiative properties. Various algorithms were developed to process the data and control their quality for the Aurora 4000 nephelometer and TAP instrument, as well as the laboratory and simulated closure experiment analysis.

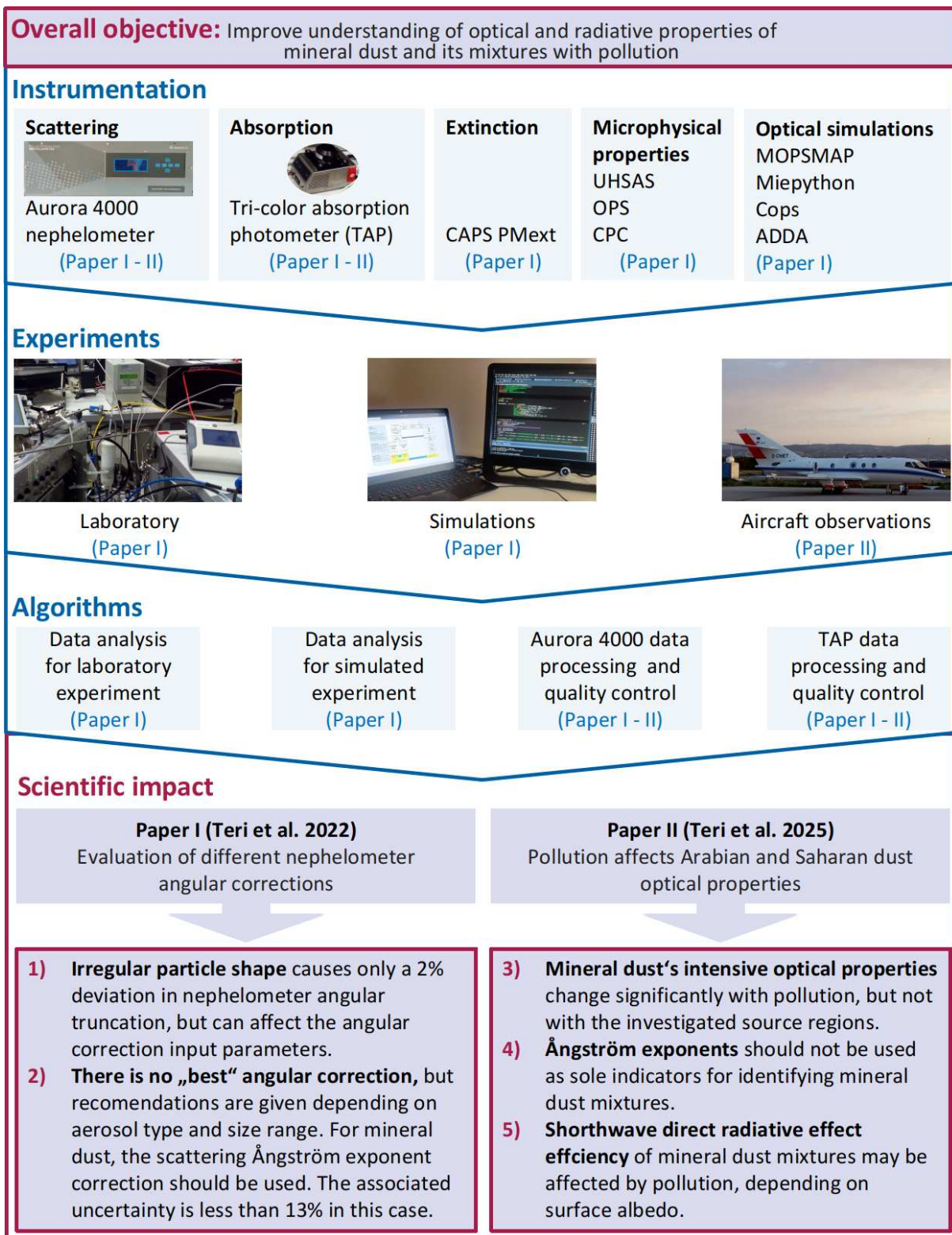


Figure 5.1: Thesis summary scheme: instrumentation, experiments, algorithms, and scientific impact. Instrumentation includes an Aurora 4000 Polar Integrating Nephelometer, a Tri-color Absorption Photometer, a Cavity Attenuated Phase Shift Extinction (CAPS PMext) Monitor, an Ultra High Sensitivity Aerosol Spectrometer (UHSAS), an Optical Particle Sizer (OPS), a Condensation Particle Counter (CPC), and simulation tools: Miepython, Cops (Collection of tools for Optical Particle Spectrometers), ADDA (based on discrete dipole approximation), and MOPSMAP (Modeled Optical Properties of enSeMbles of Aerosol Particles). See section 2.3.

The scientific results of this dissertation are presented in two publications:

- Teri et al. (2022) (paper I) provides an assessment of the Aurora 4000 nephelometer uncertainties with a focus on coarse-mode particles with irregular shapes, discusses the implications for closure studies of mineral dust, and concludes by giving guidelines for an appropriate angular correction.
- Teri et al. (2024) (paper II) provides evidence that increasing pollution within mineral dust layers affects the aerosol optical properties in the eastern Mediterranean, and provide a quantification of this change. It also notes that the optical properties of Saharan and Arabian dust are similar when comparing cases with equal pollution content. Additionally, the paper discusses the implications for the near-real-time identification of mineral dust events and the direct radiative effect of mineral dust-pollution mixtures.

Furthermore, the findings of this thesis contributed to two further publications (Groß et al., 2025; Weinzierl et al., in preparation, 2025). The following sections summarize the key findings by addressing the scientific questions of this dissertation.

1) How accurately can the Aurora 4000 nephelometer measure the particle scattering coefficient of coarse-mode irregularly shaped mineral dust particles?

The largest uncertainty in measuring the particle scattering coefficient using integrating nephelometers arises from the impossibility of measuring the light scattered in the forward and backward directions (i.e., angular truncation at angles close to 0° and 180°).

Through an extensive closure study, this thesis validates the Aurora 4000 nephelometer angular sensitivity function and evaluates the uncertainties of the angular correction in the cases of coarse-mode irregularly shaped mineral dust particles. Results from the laboratory closure experiments showed an excellent agreement within 3% between measured and simulated particle scattering coefficients for monodisperse polystyrene latex (PSL) and ammonium sulfate (AS) particles (see Figure 2.7). Additionally, the simulated closure experiment revealed that assuming a spherical shape to calculate the angular correction for coarse-mode irregular mineral dust particles leads to a small underestimation of only 2 % (see Figure 5 in Teri et al. (2022)). These findings demonstrate that the Aurora 4000 nephelometer provides accurate scattering coefficient measurements and that sphericity is a reasonable approximation to calculate the nephelometer angular correction.

While non-sphericity minimally affects nephelometer angular error, it has a much larger impact on the particle size distribution retrieved from optical particle sizers (OPSs). Simulations assuming spherical particles overestimates the measured extinction coefficients (see Figure S1 in Teri et al. (2022)). This overestimation seems to arise from size retrievals errors caused by particle non-sphericity (see Figure S2 in Teri et al. (2022)). As a result, the angular correction $C_{\text{phase,OPS}}$, calculated from particle phase function simulations based on OPS size distributions, leads to an overestimation of the particle scattering coefficient by an average of about 6% and up to 22% for mineral dust particles if sphericity and the correct refractive index are assumed (see Figure 7 last row, approach c in Teri et al. (2022)). However, using the calibration material's refractive index for the size distribution retrieval seems to compensates for non-sphericity

(see Figure 7 last row, approach a in Teri et al. (2022)), explaining the good performance of the angular correction $C_{\text{phase,OPS}}$ in the laboratory closure experiment (see Figure 3.h in Teri et al. (2022)). These findings have important implications for refractive index retrievals based on optical closure studies (e.g., Abo Riziq et al., 2007; Mack et al., 2010; Di Biagio et al., 2019). As discussed in section 6.2 of Teri et al. (2022), accounting for particle shape effects in size distribution retrievals may improve the accuracy of refractive indices derived from closure studies.

In conclusion, the particle scattering coefficients of coarse-mode irregularly shaped mineral dust particles can be accurately measured with the Aurora 4000 nephelometer as the effect of non-sphericity on the nephelometer angular error is minimal. For mineral dust measurements, the most appropriate angular correction is the Scattering Ångström exponent based correction C_{SAE} , which has a 13% uncertainty and uses a parametrization based on spherical particles (Table 3 of Teri et al. (2022)). This correction performed well for mineral dust particles also during the laboratory closure experiment (see Figure 3 of Teri et al. (2022)). These findings contribute to improving the reliability of nephelometer measurements and advancing the understanding of closure studies for mineral dust aerosols.

2) What is the most appropriate method for determining the particle scattering coefficient using an integrating nephelometer, for different particle sizes, refractive indices, and shapes?

Throughout extensive experiments designed and performed within this thesis, a better understanding of the impact of different particle sizes, refractive indices, and shapes on determining the particle scattering coefficients with integrating nephelometers was achieved. Different methods to calculate the angular correction, required to compensate for the nephelometer non-idealities were compared and evaluated. This thesis concluded that there is no generally "best" angular correction, but the angular correction should be selected depending on the aerosol type and the investigated size range. Figure 8 of Teri et al. (2022) provides guidelines to establish which angular correction is appropriate in several situations.

The angular correction C_{polar} , using the polar measurements of the particle scattering coefficient, is reliable for sub-micrometer particles with an uncertainty of 2% (Müller et al., 2012) while strongly underestimating the particle scattering coefficient for larger particle sizes, as shown in Figure 3.c and 3.d of Teri et al. (2022). Thus, the use of this correction is recommended if the aerosol inlet has a sub-micrometer cut-off or if sub-micrometer particles dominate the aerosol scattering.

The angular correction C_{SAE} , using the measured scattering Ångström exponent, resulted the most appropriate angular correction in most of the analyzed cases and may serve as a standard method, even if its uncertainty can be relatively high. Indeed, this method is the one most often used in literature (e.g., Pandolfi et al., 2018; Valenzuela et al., 2015; Costabile et al., 2013; Clarke et al., 2007). The uncertainty can be different depending on the investigated size range and the aerosol types. An uncertainty of 5% needs to be considered if the aerosol inlet has a sub-micrometer cut-off or if the aerosol scattering is dominated by sub-micrometer particles, as shown in (Bond et al., 2009) as well as in the simulations of (Teri et al., 2022, Figure 6,

fourth row, last column). An uncertainty of 13% has to be considered for coarse-mode weakly absorbing polydisperse aerosols of spherical and irregular shape, and thus especially for mineral dust particles (Teri et al., 2022, Figure 7, rows 3, 5, and 6, last column). An uncertainty of 33% must be considered if coarse-mode highly absorbing particles are present (Teri et al., 2022, Figure 7, row 4, last column). Although not very common, this situation can be found for example within fire smoke plumes (Schöberl, 2024).

The angular correction $C_{\text{phase,OPS}}$, using the particle phase function simulated based on the size distribution measured by two optical particle sizers, was found to be the most accurate angular correction in the case of coarse-mode weakly absorbing polydisperse aerosols of spherical shape with an uncertainty of 3% (Teri et al., 2022, Figure 7, row 3, column 3), and in the case of coarse-mode highly absorbing polydisperse aerosols of spherical shape with an uncertainty of 25% (Teri et al., 2022, Figure 7, row 3, column 3). However, this correction requires not only simultaneous size distribution measurements but also knowledge of the aerosol type, refractive index, and shape. For this reason, it is a rarely used method in the literature (e.g., by Di Biagio et al., 2019). The uncertainty can rise to $\sim 25\%$ for weakly absorbing spherical particles and to $\sim 60\%$ for highly absorbing spherical particles if the refractive index is assumed to be the one of the calibration material (Teri et al., 2022, Figure 6 and 7, rows 2,3 and 4, column 1 and 2).

Finally, due to the high accuracy of the nephelometer angular sensitivity function and the relatively high uncertainties introduced by the different angular corrections, reporting nephelometer data both with and without angular correction is recommended to allow its use in closure experiments without introducing additional uncertainties.

3) How does the pollution affect the optical properties of mineral dust layers, and does this effect vary depending on the mineral dust’s source region?

This thesis investigated mineral dust layers from two different source regions, the Sahara desert and the Arabian Peninsula. Both mineral dust types were investigated for different pollution contents. Several intensive optical properties, derived from airborne in situ measurements, were analyzed. Some, including the Ångström exponents of scattering and absorption and single-scattering albedo (SAE, AAE, SSAE), are commonly used for identifying aerosol types. Others, such as the asymmetry parameter and the single-scattering albedo (g and SSA), are essential for understanding the radiative effect of aerosols.

The analysis focused on 87 sequences classified as either Saharan dust (67) or Arabian dust (20) according to the A-LIFE aerosol classification scheme. It was investigated how each intensive aerosol optical property is influenced by increasing pollution content.

Our analysis revealed that all analyzed intensive aerosol optical properties, including the Ångström exponents (SAE, AAE, and SSAE), the asymmetry parameter (g) and the single-scattering albedo (SSA), changed significantly as the pollution content in the mineral dust layer increased. Specifically, higher pollution content leads to significantly higher SAE and SSAE and significantly lower AAE, g , and SSA. These findings have consequences on the identification and on the radiative effect of mineral dust layers, which were extensively discussed in Teri et al. (2024) and will be addressed in the scope of the answer to research questions 4 and 5, respectively.

Comparing mineral dust's intensive aerosol optical properties from the two investigated source regions for similar pollution content, the differences were mostly not statistically significant (see Figure 7 and Table 2 in the accepted version of Teri et al. (2024)). This result suggests that the mineral dust from the source regions observed during A-LIFE (Arabian Peninsula and Sahara desert) does not have a major intrinsic difference that lead to statistically significant variations in the measured intensive aerosol optical properties. This result was unexpected because differences in the optical properties of mineral dust can arise due to differences in the mineralogical compositions (Sokolik and Toon, 1999; Di Biagio et al., 2019), as well as from differences in how these particles mix with pollution (Kok et al., 2023; Denjean et al., 2016; Seinfeld et al., 2004). For a detailed discussion of the comparison between Arabian and Saharan dust properties, see section 4.1 in Teri et al. (2024).

In conclusion, the findings from this thesis emphasize the need for caution when retrieving the optical properties of pure mineral dust, as mineral dust layers often contain mixtures of other aerosol components. Specifically, when analyzing differences between mineral dust from various source regions, it is crucial to evaluate the pollution content and only compare measurements with similar levels of pollution.

4) To what extent can measurements from the Aurora 4000 and TAP instruments be used to identify mineral dust layers in the presence of pollution?

Three intensive aerosol optical properties, derived from the Aurora 4000 and TAP instruments, are important in the framework of aerosol types identification (e.g., Costabile et al., 2013; Cazorla et al., 2013; Cappa et al., 2016) and can be derived in near-real time: the Ångström exponents of scattering, absorption and single-scattering albedo (SAE, AAE, and SSAE). These properties represent the wavelength dependence of scattering, absorption and single-scattering albedo, respectively, and are sensitive to different aerosol characteristics. The SAE is primarily influenced by the particle size, the AAE by the particle composition, while the SSAE is associated with both (e.g., Schmeisser et al., 2017).

Results from this thesis show that pollution significantly affects the Ångström exponents of mineral dust layers (see Figure 7 in the accepted version of Teri et al. (2024)). Thus, the signal of mineral dust may be masked in mineral dust-pollution mixtures. The SAE, AAE, and SSAE values measured in mineral dust layers during the A-LIFE field experiment were used to compare the classification scheme proposed by Cappa et al. (2016) with the A-LIFE aerosol classification scheme (see Figure 8 in the accepted version of Teri et al. (2024)). Sequences classified as pure mineral dust (either Arabian or Saharan) by the A-LIFE aerosol classification scheme have values of the Ångström exponents which fall in the "Dust dominated region" accordingly to Cappa et al. (2016) and Collaud Coen et al. (2004) ($SAE < 0.5$, $AAE > 2$, and $SSAE < 0$). In contrast, polluted mineral dust sequences have values falling mainly in the "Mixed BC/BrC region" ($SAE > 1.5$, AAE in the range $1.5 - 2$) of the Cappa et al. (2016) classification. Moderately polluted mineral dust sequences show a larger variability in the Ångström exponent values. Thus, results from this thesis point out that the Ångström exponents should not be used as sole indicators to detect the presence of mineral dust because their values shift towards values often associated with anthropogenic aerosols as the pollution content increases within the mineral dust layer.

Additional independent methods are crucial to detect mineral dust in mixed aerosol layers. Once the presence of mineral dust has been established with independent methods, the Ångström exponents can be useful to estimate the amount of pollution mixed into the mineral dust layer. The best parameter among the three Ångström exponents for estimating the amount of pollution mixed into a mineral dust layer is the SAE because the pollution contribution in terms of mass explain 72 % of the variability against 43 % and 30 % for AAE and SSAE, respectively (see Table 2 in Teri et al. (2024)).

5) What is the effect of pollution on the radiative properties of mineral dust layers?

The asymmetry parameter g and the single-scattering albedo SSA are key intensive aerosol optical properties for understanding the aerosol direct radiative effect (e.g., Sherman et al., 2015). This thesis investigated values of g and SSA derived from airborne in situ observations during A-LIFE, both measured in the aircraft cabin and obtained by extending the measured values to the ambient size distribution.

Results from this thesis show that g and SSA decrease significantly as the pollution content increases within the mineral dust layer. For example at 525 nm, values of g and SSA decreases from pure to polluted mineral dust layers from 0.67(0.63 – 0.72) to 0.56(0.50 – 0.59) and from 0.96(0.94 – 0.98) to 0.89(0.84 – 0.94), respectively (see values in Table 1 of Teri et al. (2024)). This findings align with previous studies, with differences likely due to variations in the measured diameter size range and methods for identifying and quantifying the pollution.

The decrease in the single-scattering albedo (SSA) and the asymmetry parameter (g) suggests that the increase in pollution content may have two opposing effects on the shortwave direct radiative impact of the mineral dust layer. The decrease in SSA indicates increased particle absorption, which reduces the surface cooling effect (or enhances the atmospheric warming effect). In contrast, the decrease in g implies that less solar radiation is scattered towards the Earth’s surface, contributing to an increase in the cooling effect. This latter effect can be better understood by considering that an increase in pollution denotes not only the presence of more black carbon particles but also more fine mode anthropogenic aerosol particles.

To estimate how the direct radiative effect changes for increasing pollution content compared to a pure mineral dust layer with the same aerosol optical depth (AOD), the shortwave direct radiative effect efficiency (SW DREE) was calculated, using the values of g and SSA representative for the ambient size distribution and a simple formula developed by Charlson et al. (1991) and Haywood and Shine (1995). Although its absolute numbers are highly uncertain (e.g., Yu et al., 2006), the functional relationship between SW DREE, g and SSA remains valid (Sherman et al., 2015). Thus, important conclusions were drawn considering the change of SW DREE for increasing pollution within a mineral dust layer.

When assuming the globally-averaged surface albedo, the pollution content in mineral dust layers does not have a statistically significant effect on the shortwave direct radiative effect efficiency (SW DREE), likely due to a balance between the effects of increased absorption and decreased particle size. However, the SW DREE can be influenced by the pollution content in the mineral dust layer above surfaces with different albedo. High albedo surfaces (e.g., deserts

or snow) may experience reduced cooling (or increased warming of the atmosphere) with more pollution, while low albedo surfaces (as oceans) may see enhanced cooling, although this latter effect was not statistically significant in this data.

In conclusion, this thesis reduces the uncertainties of the direct in situ measurements of coarse-mode irregularly shaped mineral dust particles by improving the understanding of the nephelometer angular correction. The implications for closure studies were discussed, and guidelines for choosing the appropriate angular correction were given. Additionally, it systematically investigates the intensive optical properties of mineral dust-pollution mixtures on the basis of comprehensive airborne observations in the eastern Mediterranean. It provided evidence that mineral dust optical properties are affected by the pollution content, while the differences between the investigated mineral dust source regions are not statistically significant. It shows that the identification of mineral dust mixtures should not be solely based on Ångström exponents. Additionally, this thesis highlights the importance of quantifying the pollution content within mineral dust mixtures because it may affect the direct radiative effect efficiency of the mixed aerosol layers.

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Appendix

Enclosed papers

The papers of this thesis are enclosed in the following pages.



Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections

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Abstract. Aerosol particles in the atmosphere interact with solar radiation through scattering and absorption. Accurate aerosol optical properties are needed to reduce the uncertainties of climate predictions. The aerosol optical properties can be obtained via optical modeling based on the measured particle size distribution. This approach requires knowledge or assumptions on the particle refractive index and shape. Meanwhile, integrating nephelometry provides information on the aerosol scattering properties directly. However, their measurements are affected by angular non-idealities, and their data need to be corrected for angular truncation and illumination to provide the particle scattering coefficient. We performed an extensive closure study, including a laboratory and a simulated experiment, aiming to compare different nephelometer angular truncation and illumination corrections (further referred to as “angular corrections”). We focused on coarse-mode irregularly shaped aerosols, such as mineral dust, a worldwide abundant aerosol component. The angular correction of irregular particles is found to be only $\sim 2\%$ higher than the angular correction of volume-equivalent spheres. If the angular correction is calculated with Mie theory, the particle size distribution is needed. Our calculations show that if the particle size distribution is retrieved from optical particle spectrometer measurements and the irregular shape effect is not considered, the angular cor-

rection can be overestimated by about 5 % and up to 22 %. For mineral dust, the traditional angular correction based on the wavelength dependency of the scattering coefficient seems more accurate. We propose a guideline to establish the most appropriate angular correction depending on the aerosol type and the investigated size range.

1 Introduction

The atmosphere contains aerosol particles that scatter or absorb the solar radiation (direct effect) and act as cloud condensation nuclei (indirect effect), influencing the Earth’s energy budget and thus the climate system. The aerosol radiative forcing constitutes one of the largest uncertainties in climate predictions (IPCC, 2013). To reduce this uncertainty, climate models require more accurate information on the optical properties of aerosol particles. In particular, the particle scattering coefficient, the angular distribution of the scattered light, and the particle absorption coefficient are crucial quantities for calculating the aerosol direct radiative forcing. The traditional method to obtain the aerosol optical properties is to calculate them via optical modeling based on the particle size distribution measured with particle spectrometers (e.g.,

Hermann et al., 2016; Vasilatou, 2021), but different measurement techniques also exist to measure the aerosol optical properties directly. For example, integrating nephelometry, firstly introduced by Beuttell and Brewer (1949), is extensively used in monitoring stations, field campaigns, and laboratory experiments (e.g., Pandolfi et al., 2018; Cappa et al., 2016; Di Biagio et al., 2019). An integrating nephelometer performs a geometrical integration of the light scattered by gas molecules and aerosol particles in an enclosed volume over almost the complete angular range (Heintzenberg and Charlson, 1996; Anderson et al., 1996; Horvath, 1973). Therefore, it is possible to obtain values close to the total scattering coefficient without assuming the particle refractive index and shape.

Different measurement techniques and models could be used to independently derive the same parameters of an aerosol. This practice, called “closure experiment”, aims not only to characterize a specific parameter but also to minimize the measurement uncertainties (Weinzierl et al., 2017). In particular, the closure, between optical properties simulated from the measured size distribution and the optical properties measured directly, in principle allows constraining the refractive index of the investigated aerosol (e.g., Abo Rizeq et al., 2007; Mack et al., 2010; Kassianov et al., 2014).

The largest uncertainty of nephelometer measurements is the impossibility of measuring the light scattered by particles and air in the whole angular range, the angular truncation error (Heintzenberg and Charlson, 1996; Anderson et al., 1996), combined with the angular non-idealities of the light source (Anderson et al., 1996; Müller et al., 2009). Therefore, data from integrating nephelometers must be corrected to obtain the particle scattering coefficient. Since larger particles scatter more into the forward direction than smaller particles, the angular truncation error depends on size; the aerosol type can also affect the angular correction, and its determination is not straightforward. This correction is commonly called “truncation correction”, but the correct term is angular correction, since it takes into account both errors: the angular truncation and the non-idealities of the light source.

An empirical angular correction, exploiting the wavelength dependence of scattering, was developed by Anderson and Ogren (1998) for the nephelometer TSI model 3563. This angular correction was adapted for the Ecotech Aurora 3000 nephelometer by Müller et al. (2011a). The correlation between the scattering wavelength dependence and angular correction is due to the dependency of both quantities on the particle size. However, Bond et al. (2009) and Massoli et al. (2009) pointed out that this angular correction might be erroneous in some cases since the wavelength and angular dependence of scattering can be different for absorbing particles. Even if the uncertainty of this angular correction is only about 5 %, Bond et al. (2009) remarked that this uncertainty is almost as large as the angular truncation error itself in the sub-micrometer size range, where absorbing particles are relevant.

Another method of calculating the angular correction uses Mie theory and the angular sensitivity function, which takes into account the geometrical limitations of the nephelometer (Heintzenberg et al., 2006). This method requires knowledge of the particle size distribution and the refractive index, in addition to the nephelometer measurements. However, Heintzenberg et al. (2006) found larger uncertainties ($\sim 20\%$) associated with super-micrometer particles. In addition, Schladitz et al. (2009, 2011) showed that this approach also requires the knowledge of particle shape in the case of non-spherical particles, such as mineral dust aerosol. Mineral dust, raised from desert areas by the wind and transported around the globe, is the most abundant aerosol component in terms of dry mass (Choobari et al., 2014; Textor et al., 2006). A study based on the spheroids model (Quirantes et al., 2008) indicated that the effect of non-sphericity on the angular correction might be small. Meanwhile, Sorribas et al. (2015) estimated an uncertainty of about 13 % for the Anderson and Ogren (1998) angular correction in the case of coarse-mode desert dust aerosol and recommended the re-evaluation of this angular correction for coarse-mode aerosol.

This work aims to evaluate and compare different nephelometer angular truncation and illumination corrections with a focus on mineral dust aerosol. We used the Ecotech Aurora 4000 polar nephelometer, which measures the light scattering in 18 different angular sectors in addition to the total scattering coefficient. Müller et al. (2012) measured the angular illumination function of the Aurora 4000 polar nephelometer and provided its angular sensitivity function for all angular sectors. An alternative angular correction, exploiting the information on scattering for different angular sectors of the Aurora 4000 nephelometer, was developed by Müller et al. (2012). This angular correction, in addition to spherical particles, considers several particle shapes.

Very recently, when our data were already processed, a new angular correction was proposed by Qiu et al. (2021). The new angular correction exploits the wavelength dependency of the scattering coefficient, the hemispheric back-scattering fraction, and new technologies such as the random forest machine learning. The random forest machine learning was trained based on a data set representing regional anthropogenic aerosol in the North China Plain, including mainly sub-micrometer particles. The utilized data set limits the use of the new angular correction to sub-micrometer spherical particles. Since our work mainly focused on coarse-mode irregularly shaped particles, we did not consider this angular correction in our comparison.

We conducted an extensive closure study, including a laboratory and a simulated experiment, focusing on coarse-mode irregularly shaped particles (mineral-dust-like). Laboratory test aerosols were generated using polystyrene latex (PSL) particles, ammonium sulfate (AS), and soil dust samples. Their extinction, scattering, and absorption properties, as well as the particle number concentration and size distribution,

bution, were measured by seven instruments in parallel. To better interpret the results of the laboratory experiment, we performed a simulated closure experiment randomly selecting several size distributions, refractive indices, and shapes (e.g., irregularly shaped dust-like particles (Gasteiger et al., 2011)). We estimated the uncertainties in the angular correction of the Aurora 4000 polar nephelometer calculated using parallel particle size distribution measurements from a TSI OPS 3330. The TSI OPS 3330 is an optical particle spectrometer, a class of instrument widely used to measure the aerosol size distributions during aircraft field experiments (e.g., Weinzierl et al., 2017; Spanu et al., 2020; Brock et al., 2021).

In Sect. 2, we describe the principle of operation of the Aurora 4000 polar nephelometer. In Sect. 3, we review the methods considered for calculating the angular correction and their main limitations. In Sect. 4, we describe the laboratory closure experiment and the most important results. In Sect. 5, we describe the simulated closure experiment and its main outcomes. In Sect. 6, we discuss the results of both the laboratory and the simulated experiments and we compare them with the literature. In Sect. 7, we draw the main conclusions and we give recommendations for an appropriate angular correction.

2 Principle of operation of the Aurora 4000 polar nephelometer

The Aurora 4000 polar nephelometer is a three-wavelength integrating nephelometer. Integrating nephelometers measure the light scattered by particles and air in a volume. The light detector is placed orthogonally to a nearly Lambertian light source¹ such that the angular illumination function is similar to a $\sin(\theta)$, where θ is the scattering angle (Müller et al., 2009). Volume elements along the detector field of view contribute to the measured signal with light scattered at different θ . The measured signal is the integral over nearly all angles of the angular scattering function $\gamma(\Omega)$, the fraction of light scattered out of a parallel beam of light per unit length and per unit solid angle Ω with respect to the direction of the incident beam. In other words, integrating nephelometers measure a signal proportional to the scattering coefficient σ_s , the fraction of light scattered out of a parallel beam of light per unit length. A full description of the measurement principle of an integrating nephelometer is available in the appendix of Anderson et al. (1996).

The Aurora 4000 polar nephelometer uses as light sources three light-emitting diodes (LEDs) at different wavelengths: blue ($\lambda_B = 450$ nm), green ($\lambda_G = 525$ nm), and red ($\lambda_R = 635$ nm). It is marked as “polar” because it integrates $\gamma(\Omega)$ in several angular sectors. A shutter can block the angular illu-

mination, such that the detector measures only the light scattered between the shutter position α and 180° . The shutter of the Aurora 4000 can consecutively assume up to 18 angular positions ($0, 10, 15, \dots, 90^\circ$). For a given shutter position α and wavelength λ , the signal measured by the Aurora 4000 can be written as

$$S_\lambda^\alpha \propto 2\pi \int_{\theta=0^\circ}^{\theta=180^\circ} \gamma_\lambda(\theta) Z_\lambda^{\text{Aurora 4000}}(\alpha, \theta) d\theta, \quad (1)$$

where $Z_\lambda^{\text{Aurora 4000}}(\alpha, \theta)$ describes the angular sensitivity of the Aurora 4000 for shutter position α at scattering angle θ (see Eq. 5 in Sect. 3.1). The signal S_λ^α is proportional to the scattering coefficient $\sigma_{s,\lambda}^\alpha$ in the angular sector $\alpha - 180^\circ$. The two extreme positions $\alpha = 0^\circ$ and $\alpha = 90^\circ$ yield a signal proportional to the total scattering coefficient $\sigma_{s,\lambda}^{0^\circ}$ and hemispheric back-scattering coefficient $\sigma_{s,\lambda}^{90^\circ}$, respectively.

Calibration with particle-free gases of known Rayleigh scattering coefficient leads to the constant of calibration K_λ^α , which relates the measured signal S_λ^α to the particle scattering coefficient $\sigma_{sp,\lambda}^\alpha$ in the angular sector $\alpha - 180^\circ$:

$$\sigma_{sp,\lambda}^\alpha = K_\lambda^\alpha S_\lambda^\alpha - \sigma_{sR,\lambda}^\alpha, \quad (2)$$

where $\sigma_{sR,\lambda}^\alpha$ is the Rayleigh scattering coefficient of particle-free air in the angular sector $\alpha - 180^\circ$. It can be derived integrating in the angular sector $\alpha - 180^\circ$ the Rayleigh angular scattering function $\gamma_R(\theta) = \frac{\sigma_{sR,\lambda}^{0^\circ}}{4\pi} \frac{3}{4} (1 + \cos^2(\theta))$:

$$\sigma_{sR,\lambda}^\alpha = \sigma_{sR,\lambda}^{0^\circ} \left[\frac{1}{2} + \frac{1}{8} \cos^3(\alpha) + \frac{3}{8} \cos(\alpha) \right], \quad (3)$$

where $\sigma_{sR,\lambda}^{0^\circ}$ is the Rayleigh total scattering coefficient of air (e.g., Bucholtz, 1995) at the wavelength λ calculated at the temperature and pressure of the carrier gas. The second term is a value changing from 1 for $\alpha = 0^\circ$ to 0.5 for $\alpha = 90^\circ$.

3 Angular truncation and illumination correction

3.1 Angular sensitivity function

In an ideal nephelometer, the light source is perfectly Lambertian and the angular integration is complete from α to 180° . Thus, the angular sensitivity function can be written as

$$Z_\lambda^{\text{ideal}}(\alpha, \theta) = \begin{cases} 0 & 0^\circ < \theta < \alpha \\ \sin(\theta) & \alpha < \theta < 180^\circ. \end{cases} \quad (4)$$

Any real nephelometer is affected by non-idealities that need to be considered in the angular sensitivity function. First, the angular truncation error: the light scattered into the extreme forward ($\theta = 0^\circ$) and backward ($\theta = 180^\circ$) directions cannot be measured, and the detector field of view is restricted to

¹A light source is Lambertian if the emitted radiance is proportional to $\cos(\phi)$, where ϕ is the angle between the direction of the emitted light and the surface normal to the light surface.

angular detection limits (Anderson et al., 1996; Moosmüller and Arnott, 2003). Second, the light source is not perfectly Lambertian and the angular illumination function deviates a little from a sine function (Anderson et al., 1996; Müller et al., 2009). Third, the separation by the shutter is not perfectly sharp and the shadowing of the shutter has to be considered when its position is different from zero (Müller et al., 2011a). The angular sensitivity function of the Aurora 4000 polar nephelometer can be written as (Müller et al., 2012)

$$Z^{\text{Aurora 4000}}(\alpha, \theta) = \begin{cases} 0^\circ < \theta < \theta_1 & \\ \max \left\{ 0, \beta_1 \sin(\theta)^{\beta_2} \min \left[1, \left(\frac{\theta - \delta_1(\alpha)}{\delta_2(\alpha)} \right) \right] \right\} & \theta_1 < \theta < \theta_2 \\ \theta_2 < \theta < 180^\circ & \end{cases} \quad (5)$$

The angular limits of detection θ_1 and θ_2 are reported in the Aurora 4000 polar nephelometer user manual. β_1 and β_2 describe the deviation from the sine as measured by Müller et al. (2012) with the method described in Müller et al. (2009). $\delta_1(\alpha)$ and $\delta_2(\alpha)$ account for the shadowing of the shutter as a function of its position α and are provided by Müller et al. (2012). The parameters for the Aurora 4000 polar nephelometer are reported in Table 1 and are the same for the three wavelengths.

The ideal particle scattering coefficient $\sigma_{\text{sp}}^{\text{ideal}, \alpha}$ and the Aurora 4000 particle scattering coefficient $\sigma_{\text{sp}}^{\text{Aurora 4000}, \alpha}$ can be explicitly written as

$$\sigma_{\text{sp}, \lambda}^{\text{ideal}, \alpha} = 2\pi \int_{\theta=0^\circ}^{\theta=180^\circ} \gamma_{\text{p}, \lambda}(\theta) \sin(\theta) d\theta, \quad (6)$$

$$\sigma_{\text{sp}, \lambda}^{\text{Aurora 4000}, \alpha} = 2\pi C_{\text{R}, \alpha} \int_{\theta=0^\circ}^{\theta=180^\circ} \gamma_{\text{p}, \lambda}(\theta) \times Z^{\text{Aurora 4000}}(\alpha, \theta) d\theta, \quad (7)$$

where $C_{\text{R}, \alpha}$ takes into account the angular truncation and illumination correction implicitly performed through the particle-free gases calibration (Anderson and Ogren, 1998; Müller et al., 2011a). It can be calculated as

$$C_{\text{R}, \alpha} = \frac{\int_{\theta=0^\circ}^{\theta=180^\circ} \gamma_{\text{R}}(\theta) \sin(\theta) d\theta}{\int_{\theta=0^\circ}^{\theta=180^\circ} \gamma_{\text{R}}(\theta) Z^{\text{Aurora 4000}}(\alpha, \theta) d\theta}. \quad (8)$$

For the Aurora 4000 polar nephelometer, the Rayleigh angular truncation and illumination correction for $\alpha = 0^\circ$ is a non-negligible value:

$$C_{\text{R}, 0^\circ} = 1.06. \quad (9)$$

Taking into account both the angular truncation and the non-Lambertian illumination, the angular correction C_α for

any position of the shutter α can be defined, following the definition given by Anderson and Ogren (1998), as

$$C_\alpha = \frac{\sigma_{\text{sp}}^{\text{ideal}, \alpha}}{\sigma_{\text{sp}}^{\text{Aurora 4000}, \alpha}}. \quad (10)$$

In this work, we focus on the angular correction C_{0° , that is the angular correction to obtain the particle scattering coefficient $\sigma_{\text{sp}}^{0^\circ}$ from the particle scattering coefficient measured by the Aurora 4000 for shutter position $\alpha = 0^\circ$, $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$. Since larger particles scatter into the forward direction more than smaller particles, C_{0° is size-dependent and increases with increasing particle size. This effect was originally reported by Heintzenberg and Quenzel (1973). Different methods have been proposed in the literature to calculate the angular correction C_{0° . The angular correction can be calculated analytically using the definition (Eq. 10) if the angular distribution of scattering by particles, i.e., the particle phase function, is known. To highlight the importance of the particle phase function to apply this method, we refer to this angular correction as C_{phase} . Other methods of calculating the angular correction exploit the relationship between C_{0° and quantities measured by the nephelometer. We consider an angular correction based on the wavelength dependence of the particle scattering coefficient, the scattering Ångström exponent, C_{SAE} , and one based on the polar measurements of the particle scattering coefficient C_{polar} . In the following sections, we describe each method and its limitations by reviewing the related literature, as also summarized in Table 2. To the best of our knowledge, there is currently no literature comparing these corrections and describing which correction works best.

3.2 Angular correction using the phase function: C_{phase}

The angular correction can be calculated on the basis of the definition Eq. (10) if the particle phase function $P_{\text{p}}(\theta)$ is available. Considering the definition of particle phase function ($P_{\text{p}}(\theta) = 4\pi \gamma_{\text{p}}(\theta) / \sigma_{\text{sp}}$) and writing explicitly the ideal and Aurora 4000 particle scattering coefficient (see Eqs. 6–9) with $\alpha = 0^\circ$, the angular correction can be written as

$$C_{\text{phase}} = \frac{\int_{0^\circ}^{180^\circ} P_{\text{p}}(\theta) \sin(\theta) d\theta}{\int_{0^\circ}^{180^\circ} P_{\text{p}}(\theta) Z^{\text{Aurora 4000}}(0^\circ, \theta) d\theta} \frac{1}{C_{\text{R}, 0^\circ}}, \quad (11)$$

where the second term takes into account the angular truncation and illumination error for Rayleigh scattering. Knowledge about the particle phase function is required to calculate the first term. The particle phase function for a given wavelength can be calculated via Mie theory if the particles are spherical and homogeneous with a known size distribution and refractive index. This method was proposed, e.g., by Heintzenberg et al. (2006). However, Schladitz et al. (2009) found high discrepancies between Mie-calculated and measured scattering coefficients for desert dust events and

Table 1. Parameters of the Aurora 4000 angular sensitivity function given by Müller et al. (2012).

	θ_1	θ_2	β_1	β_2	$\delta_1(\alpha)$	$\delta_2(\alpha)$
Aurora 4000	9°	170°	1.00	1.109	$0.4156 \cdot \alpha + 0.0041 \cdot \alpha^2$	$1.1935 \cdot \alpha + 0.0082 \cdot \alpha^2$

Table 2. Different angular corrections for the particle scattering coefficient and their limitations.

Correction	Formula	Literature	Reported limitations
C_{phase}	$\frac{\int_{0^\circ}^{180^\circ} P_p(\theta) \sin(\theta) d\theta}{\int_{0^\circ}^{180^\circ} P_p(\theta) Z^{\text{Aurora 4000}}(0^\circ, \theta) d\theta} \frac{1}{C_{R,0^\circ}}$	Angular correction developed by Heintzenberg et al. (2006). The particle phase function $P_p(\theta)$ can be obtained via Mie simulations if the size distribution and refractive index are known. $Z^{\text{Aurora 3000}}(0^\circ, \theta)$ was measured by Müller et al. (2011a). $Z^{\text{Aurora 4000}}(0^\circ, \theta)$ was measured by Müller et al. (2012) (see Eq. 5 and parameters in Table 1).	Large uncertainties were found for super-micrometer particles (Heintzenberg et al., 2006). If the particle shape differs from spheres, this angular correction may be not applicable (Schladitz et al., 2009, 2011).
C_{SAE}	$a + b \cdot \text{SAE}^*$	Angular correction developed by Anderson and Ogren (1998) for the TSI nephelometer. The parameters for the Aurora 3000 are given by Müller et al. (2011a). The parameters for the Aurora 4000 are given in Table 3.	Large uncertainties are expected for coarse-mode aerosol (Anderson and Ogren, 1998); Sorribas et al. (2015) recommend this correction to be re-evaluated for coarse-mode aerosol. This correction can be erroneous for absorbing particles (Bond et al., 2009; Massoli et al., 2009).
C_{polar}	$\left(c + d \cdot \frac{\sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ} - \sigma_{\text{sp}}^{\text{Aurora 4000}, 20^\circ}}{\sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ}} \right)^{-1}$	Angular correction developed for the Aurora 4000 by Müller et al. (2012). The parameters for the Aurora 4000 are given in Eq. (14).	This angular correction underestimates the true scattering coefficient for large particles (Müller et al., 2012).

showed that the particle shape also needs to be considered in the case of non-spherical particles. As an alternative, the particle phase function can be measured directly using a non-integrating polar nephelometer, an instrument that uses a rotating arm to measure the angular scattering function with high angular resolution without performing the angular integration (e.g., Waldram, 1945; Horvath et al., 2018).

3.3 Angular correction using the scattering Ångström exponent: C_{SAE}

A method to calculate the angular correction C_{0° without additional measurements exploits the wavelength dependence of scattering expressed by the scattering Ångström exponent (SAE). If the particle scattering coefficient is measured at two wavelengths, λ_1 and λ_2 , the scattering Ångström exponent can be obtained as

$$\text{SAE}(\lambda_1/\lambda_2) = -\frac{\log(\sigma_{\text{sp},\lambda_1}/\sigma_{\text{sp},\lambda_2})}{\log(\lambda_1/\lambda_2)}. \quad (12)$$

Anderson and Ogren (1998) found a linear relationship between the SAE and C_{0° and proposed that the angular cor-

rection can be constrained using the SAE. The angular correction C_{SAE} exploiting the SAE is given by

$$C_{\text{SAE}} = a + b \cdot \text{SAE}^*, \quad (13)$$

where SAE^* is the scattering Ångström exponent calculated using uncorrected nephelometer measurement of σ_{sp} . Wavelength pairs are λ_B and λ_G for the angular correction at the blue wavelength (λ_B), λ_B and λ_R for the angular correction at the green wavelength (λ_G), and λ_G and λ_R for the angular correction at the red wavelength (λ_R). The parameters a and b are derived from Mie calculations of C_{0° for ranges of particle sizes and refractive indices. The parameters a and b for the TSI nephelometer model 3563 are given by Anderson and Ogren (1998) and for the Ecotech Aurora 3000 nephelometer by Müller et al. (2011a). To apply this method to the Ecotech Aurora 4000 polar nephelometer, we calculated the parameters for the angular correction C_{SAE} considering the angular sensitivity function measured by Müller et al. (2012). We used the same size distribution and refractive indices used by Anderson and Ogren (1998) and Müller et al. (2011a). The parameters are given in Table 3.

Table 3. Angular corrections for Aurora 4000 for the total scattering coefficient C_{0° as a function of the scattering Ångström exponent: $C_{0^\circ} = a + b \cdot \text{SAE}^*$. For correction of the scattering coefficients for the blue wavelength (λ_B), the scattering Ångström exponent calculated from uncorrected scattering coefficients of blue and green wavelengths (λ_B , λ_G) is used. At wavelength λ_G and λ_R , scattering Ångström exponents at the wavelength pairs (λ_B , λ_R) and (λ_G , λ_R) are used, respectively.

Wavelength		$\lambda_B = 450 \text{ nm}$		$\lambda_G = 525 \text{ nm}$		$\lambda_R = 635 \text{ nm}$	
Ångström exponents		$\text{SAE}^*(\lambda_B, \lambda_G)$		$\text{SAE}^*(\lambda_B, \lambda_R)$		$\text{SAE}^*(\lambda_G, \lambda_R)$	
Parameters		a	b	a	b	a	b
Aurora 4000	No size cut-off	1.361	−0.159	1.352	−0.151	1.334	−0.135
	Sub-micrometer size cut-off	1.137	−0.037	1.127	−0.036	1.105	−0.03

Calculations are limited to weakly absorbing particles with the imaginary part of the refractive index k in the range of 0.00–0.01. The real part of the refractive index n is in the range of 1.40–1.52. The size distributions are bimodal log-normal functions with a geometric standard deviation of 1.8 and a geometric volume median diameter in the range of 0.2–0.4 μm for the fine mode and 2.0–4.0 μm for the coarse mode. The fine-mode volume fraction varies between 0.1 and 0.9. Calculations are done considering either the complete size distribution (no size cut-off) or a sub-micrometer size cut-off, with an aerodynamic diameter smaller than 1 μm (0.77 μm for density $\rho = 1.7 \text{ g cm}^{-3}$). This size segregation was introduced by Anderson and Ogren (1998). They point out that the angular correction is better constrained by the SAE when only sub-micrometer particles are measured, while larger uncertainties $\sim 23\%$ are expected when coarse-mode particles are also measured.

Further limitations of this method were found by Bond et al. (2009) theoretically and by Massoli et al. (2009) experimentally. They point out that the uncertainty of the angular correction C_{SAE} increases for absorbing particles. Indeed, the wavelength dependence of the particle scattering coefficient is related to particle size only indirectly.

3.4 Angular correction using the polar measurements of the particle scattering coefficient: C_{polar}

A new angular correction based on the specific feature of the Aurora 4000 that integrates the angular scattering function $\gamma(\theta)$ at many different angular sectors ($\alpha - 170^\circ$) was developed by Müller et al. (2012).

In particular, the particle scattering coefficients $\sigma_{\text{sp}}^{\text{Aurora 4000}, \alpha}$ for $\alpha = 10^\circ$ and $\alpha = 20^\circ$ are used to obtain the polar factor

$$\left(\sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ} - \sigma_{\text{sp}}^{\text{Aurora 4000}, 20^\circ} \right) / \sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ}.$$

The angular correction C_{polar} is given by

$$C_{\text{polar}} = \left(1.043 - 0.7651 \frac{\sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ} - \sigma_{\text{sp}}^{\text{Aurora 4000}, 20^\circ}}{\sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ}} \right)^{-1}. \quad (14)$$

The parameters were derived from scattering theory using both Mie and discrete dipole approximation codes. Calculations were done for different shapes: spheres, cubes and nested cubes, and three different kinds of dust shapes. The size parameter ($x = \pi d_p / \lambda$) ranges from 0.1 to 28. The imaginary part of the refractive index k is in the range of 0.00–0.1. The real part of refractive index n is in the range of 1.53–1.55. Via model simulations, Müller et al. (2012) showed that the angular correction C_{polar} is more accurate than the angular correction C_{SAE} for volume median diameters below 1 μm and for a large range of refractive indices and particle shapes, and that both corrections underestimate the particle scattering coefficient for particles larger than 1 μm .

4 Laboratory closure experiment

4.1 Experimental methods

To test the different angular corrections for the Aurora 4000 polar nephelometer, we deploy several instruments to measure the optical and microphysical properties of laboratory-generated aerosol, both monodisperse and polydisperse. The experimental set-up is shown in Fig. 1. The Aurora 4000 polar nephelometer and a self-built polar nephelometer, hereafter referred to as Horvath's polar nephelometer (Horvath et al., 2018), were used to measure the particle scattering coefficients at $\lambda = 450$, 525, and 635 nm and the particle phase function at $\lambda = 532$ nm, respectively. Besides the particle scattering properties, the particle extinction coefficient at 530 nm was measured by a cavity attenuated phase shift monitor PMext (CAPS PMext, Aerodyne) and the particle absorption coefficient at 467, 528, and 652 nm by a tricolor absorption photometer (TAP, Brechtel). The particle number concentration was measured with a condensation particle counter (CPC 3772, TSI) and the particle size distribution with two optical particle spectrometers: the UHSAS (DMT) and the OPS 3330 (TSI), covering the nominal size range 0.06–1 and 0.3–10 μm , respectively. All instruments were connected to a stainless-steel chamber where the aerosol flow was mixed and diluted with particle-free dry air. Nearly monodisperse aerosols were generated using PSL particles of

a nominal diameter in the range of 0.1–1.8 μm or ammonium sulfate in combination with a Vienna-type differential mobility analyzer (DMA) (Steiner et al., 2010), obtaining particle size distributions with a diameter in the range of 0.2–0.85 μm . Polydisperse aerosols were generated by blowing dry air through a hermetic glass bottle containing the sample powder. The bottle cap had two holes for the air inlet and outlet. A metal tube was placed from the inlet almost to the bottle bottom so that the blown air flowed horizontally to the surface where the sample powder was deposited. An electrical vibrating device was mounted outside the bottle and could be activated to mobilize the powder better. The samples included synthetic non-absorbing silica dust and soil samples collected from desert areas such as sediments from a plain near Tagounite (Morocco), river sediments at Tagounite (Morocco), sand from the Sahara desert, dust from Mauritania, and soil dust from Zagora (Morocco). Moreover, one sample of volcanic ash from Eyjafjallajökull (Iceland) was included.

Measurements were repeated twice, with and without a cyclone for size-selective sampling, in order to achieve different size distributions. Although the nominal aerodynamic size cut-off of the cyclone was $\sim 1 \mu\text{m}$, we could not attain the nominal airflow through the cyclone and the achieved dimensional cut-off was most probably larger than $1 \mu\text{m}$. We measured volume median diameters in the range of 0.85–1.85 μm for measurements with the cyclone and in the range of 2.20–2.79 μm for measurements without the cyclone.

Before each measurement, the dilution chamber was filled in with particle-free dry air to remove any residual particles. Stable concentrations were required for at least 35 min due to the temporal resolution of Horvath's polar nephelometer. Moreover, concentrations corresponding to a particle extinction coefficient between 100 and 200 Mm^{-1} were preferred, since they are the CAPS PM_{ext} best condition of operation. At the beginning of each measurement, the flow was set to satisfy these requirements. However, these conditions were challenging to obtain, in particular for the mineral dust measurements. If an adjustment was required during one measurement, a manual bottle shaking was preferred rather than a flow adjustment to avoid changes in the size cut-off.

4.2 Data evaluation

The data evaluation of the laboratory experiment had the overall aim of investigating the uncertainty of the particle scattering coefficient measurements by testing different angular corrections. A summary of data evaluation from the raw data is provided in Fig. 2. The top line displays all instruments involved in the laboratory experiment. The descending arrows include all corrections performed to obtain the measured quantities of each instrument (indicated in yellow boxes). Black arrows indicate when data from an instrument are an input for the correction of a different instrument. The tested angular corrections are reported in red boxes. The angular corrections C_{SAE} , based on the scattering Ångström ex-

ponent, and C_{polar} , based on the polar measurements of the particle scattering coefficient, are obtained directly from the Aurora 4000 polar nephelometer measurements. The angular correction based on the phase function C_{phase} is calculated, on the one hand, considering the particle phase function measured by Horvath's polar nephelometer, from now on referred to as $C_{\text{phase,H}}$. On the other hand, this angular correction is calculated considering the particle phase function simulated using the size distribution measured by the two OPSs (DMT UHSAS and TSI OPS 3330), from now on referred to as $C_{\text{phase,OPS}}$. The four angular corrections C_{SAE} , C_{polar} , $C_{\text{phase,H}}$, and $C_{\text{phase,OPS}}$ were tested comparing the corrected Aurora 4000 particle scattering coefficient with the difference of the particle extinction coefficient and the particle absorption coefficient. In addition, the particle size distributions measured by the two OPSs were used to simulate the Aurora 4000 scattering coefficients and to obtain the simulated angular corrections $C_{\text{SAE,OPS}}$ and $C_{\text{polar,OPS}}$. The six angular corrections were compared to each other.

The most important considerations of the data analysis of each instrument are summarized below. Further details of the data analysis are provided in the Supplement. All data were converted to standard temperature and pressure (STP: $T_0 = 273.15 \text{ K}$, $P_0 = 1013.25 \text{ hPa}$). The Aurora 4000 polar nephelometer was calibrated with particle-free air and CO_2 , following the procedure recommended by Anderson and Ogren (1998). A set of calibrations were performed, and the combined uncertainty due to calibration and noise for 1 min data was found to be about 8 %, while for the data averaged along with longer sequences, it is smaller than 3 %. The higher uncertainty for the 1 min data was due to the temporal resolution of about 30 s obtained by measuring the scattering coefficient $\sigma_{\text{sp},\lambda}^\alpha$ for all the 18 available positions of the scatter shutter.

The particle extinction coefficient σ_{ep} at $\lambda = 530 \text{ nm}$ was measured by the CAPS PM_{ext} . Due to the high reliability and low uncertainties ($\pm 3 \%$ according to Petzold et al., 2013, and Onasch et al., 2015) of the CAPS PM_{ext} , this instrument was considered to be the reference. It is not affected by angular truncation error, since it measures the phase shift of a square modulated signal after being reflected several times between two highly reflective mirrors placed at the extremes of the measurement chamber. The CAPS PM_{ext} was calibrated and corrected for non-linear behavior considering measurements of the Aurora 4000 particle scattering coefficient for 0.203 μm PSL particles. For this correction, the Aurora 4000 particle scattering coefficient was corrected for angular truncation and illumination correction. The angular correction was calculated with Mie simulations considering a log-normal size distribution ($d_g = 203 \text{ nm}$, $\sigma_g = 1.026$, $m(\lambda = 525 \text{ nm}) = 1.582 + i0.00$ as reported in Devon and Rudin, 1987). Besides a larger uncertainty, this calibration may introduce some compensation for the inaccuracy of the Aurora 4000 angular sensitivity function $Z^{\text{Aurora 4000}}(0^\circ, \theta)$ and therefore some circularity in our analysis. However, the

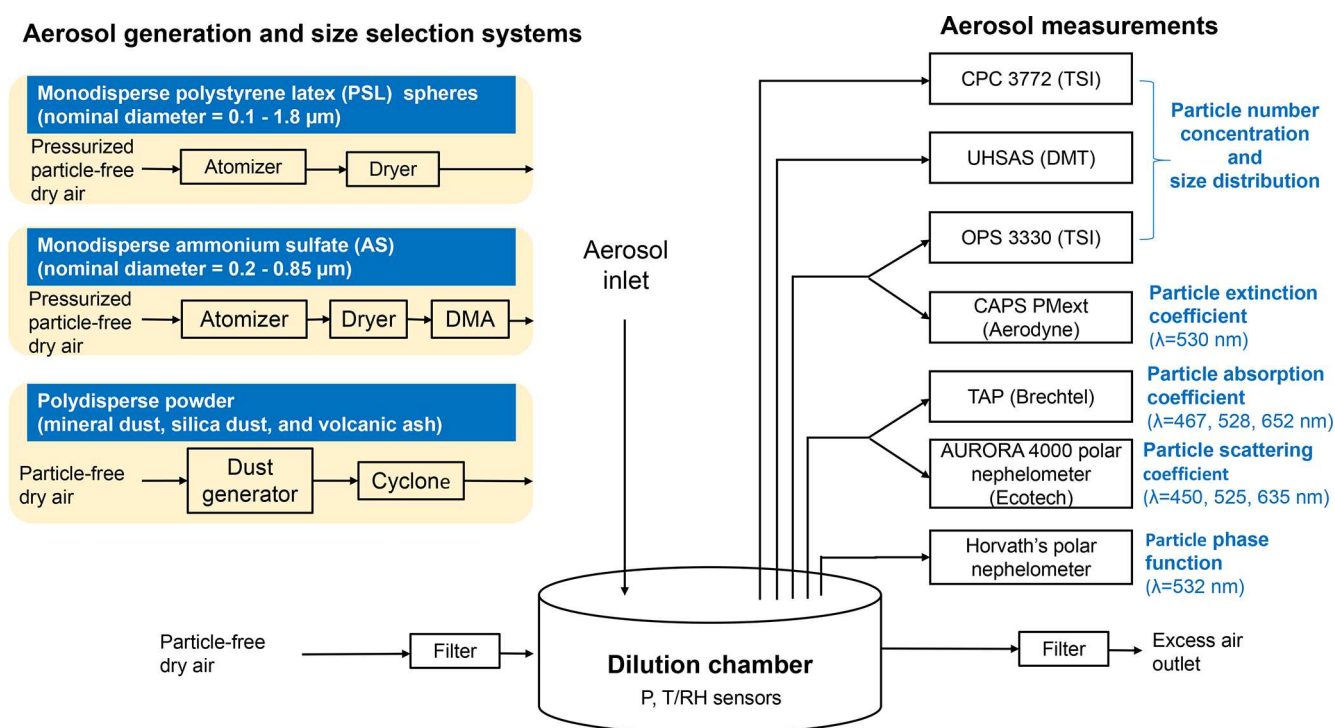


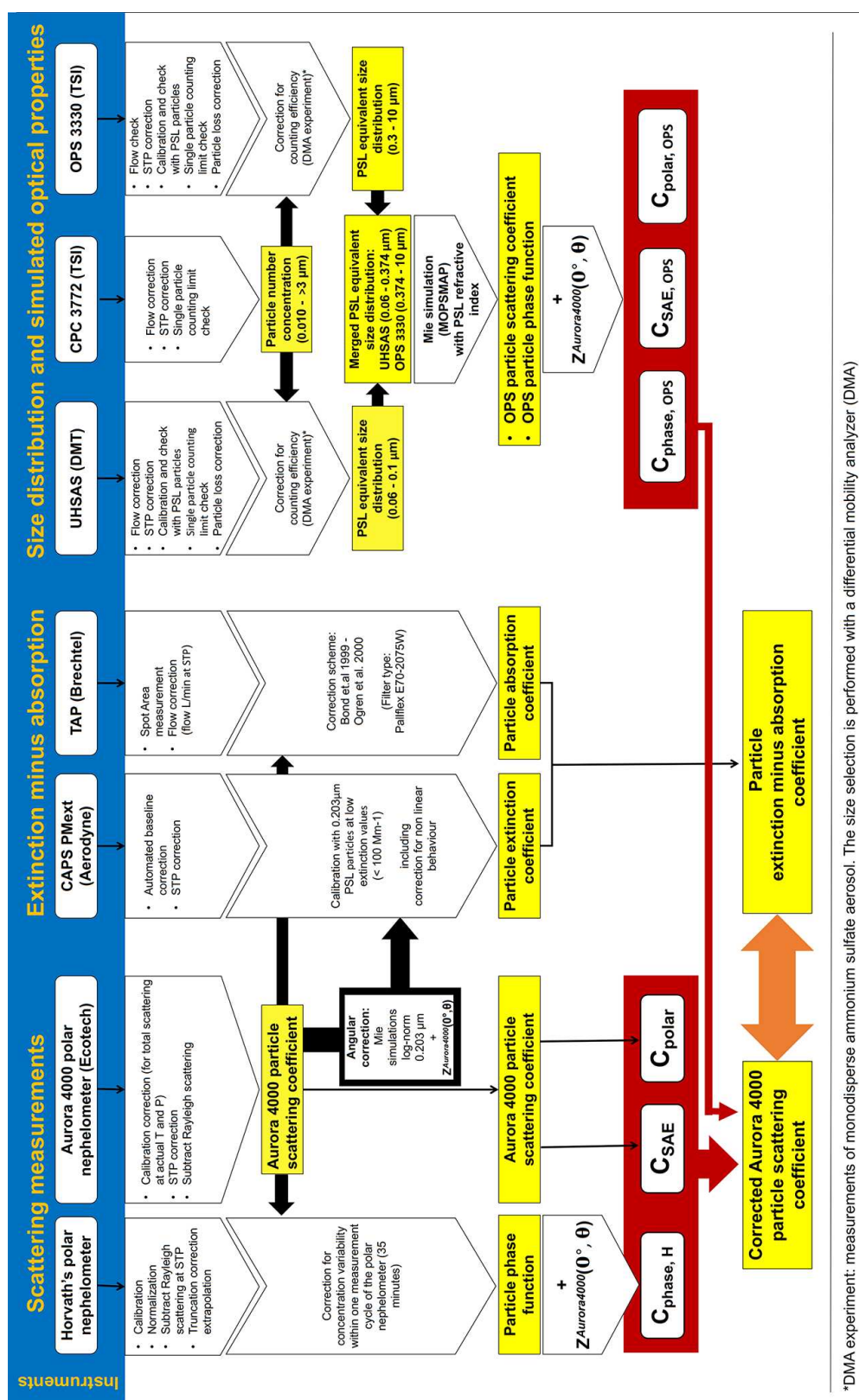
Figure 1. Scheme of the laboratory closure experiment set-up. Different aerosol types, such as monodisperse polystyrene latex (PSL) spheres, ammonium sulfate (AS), and polydisperse powder (mineral dust, silica dust, and volcanic ash), were generated using three different aerosol generation and size selection systems (in the upper left). The aerosol generation and size selection systems were connected via the aerosol inlet to a dilution chamber, where the aerosol flow is mixed with particle-free dry air. The dilution chamber was operated at a pressure slightly above ambient. Seven measurement instruments were connected to the dilution chamber to measure extinction, scattering, and absorption aerosol properties as well as particle number concentration and size distributions.

calculated Aurora 4000 angular correction for this size is very small (1 %).

Since mineral dust is expected to absorb in the green and blue wavelengths, the particle absorption coefficient was measured by the TAP at three different wavelengths: blue (467 nm), green (528 nm), and red (652 nm). The particle absorption coefficients were corrected using the angular correction developed by Bond et al. (1999) and refined by Ogren (2010). The single scattering albedo (SSA) was calculated from the particle extinction coefficient measured by the CAPS PM_{ext} at $\lambda = 530$ nm and the particle absorption coefficient measured by the TAP at $\lambda = 528$ nm and resulted in the range 0.95–0.99 for mineral dust measurements, in agreement with literature values (Ansmann et al., 2011; Schladitz et al., 2009; Müller et al., 2011b; Petzold et al., 2011). The related uncertainty for SSA in the range of 0.95–0.99 and the raw (i.e., uncorrected) absorption coefficient $> 3 \text{ Mm}^{-1}$ is estimated to increase from 35 % to 100 % as SSA increases (Ogren et al., 2017). Even considering the highest uncertainty (~ 100 %), this error hardly affects the difference between particle extinction coefficient and particle absorption coefficient because we measured only weakly absorbing aerosol with a low absorption coefficient. For all the measurements where some absorption is expected, the difference

between particle extinction coefficient and particle absorption coefficient is regarded as the reference value for light scattering because it is the best we can get for weakly absorbing irregular particles in the coarse mode. If the aerosols are expected to be non-absorbing as for PSL particles, AS, and synthetic silica, the particle absorption coefficient is set to zero and the particle extinction coefficient measured by the CAPS PM_{ext} is set as a reference value.

The Horvath's polar nephelometer was deployed to measure the particle phase function $P_p(\theta)$ at $\lambda = 532$ nm. Its detector is mounted on a rotating arm with an angular resolution of 5° , which can be moved between 5 and 175° , detecting the light scattered into its direction. A complete measurement cycle takes 35 min. Calibrations were performed with CO_2 and particle-free air. The extrapolation procedure described in Horvath (2015) is used in order to extend the phase function to 0 and 180° . The accuracy of the full integration of the phase function given by this procedure is 1 %. However, the particle phase function measured by Horvath's polar nephelometer has high uncertainty due to the low temporal resolution of the instrument and the highly variable concentration during mineral dust measurements. For compensation of the variable signal, Horvath's polar nephelometer signal at each measurement angle is divided by the 1 min average Au-



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ra 4000 particle scattering coefficient $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$ at the green wavelength. Since $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$ is proportional to the particle concentration and the aerosol type does not change, the value obtained at each angle is proportional to the particle angular scattering function $\gamma_p(\theta)$. The particle phase function $P_p(\theta)$ is obtained by normalization. In the data set collected during this experiment, there are a few cases where the extrapolation procedure performed to extend the particle phase function to 0° leads to a decreasing phase function from 10° to 0° . These results were considered unrealistic and excluded from further data analysis.

For each measurement, data were averaged along sequences with a constant number concentration or along the 35 min measurements of Horvath's polar nephelometer even if the number concentration is not constant. The result is not affected as long as the average is performed at the same time for all instrumentation. No significant variation in the scattering Ångström exponent $\text{SAE}(\lambda_B, \lambda_R)$ was observed during the selected sequences. We considered all measured quantities at the green wavelength. Although the instruments involved have slightly different wavelengths (up to 7 nm difference), no wavelength correction has been considered.

The particle size distribution was obtained by merging the particle size distribution measured by the UHSAS in the range of 0.06–0.374 μm and the OPS 3330 in the range of 0.374–10 μm . The counting efficiency of the two instruments was estimated by comparing their particle number concentration with the particle number concentration given by the CPC in monodisperse ammonium sulfate aerosols. The accuracy of the CPC number concentration reported by the manufacturer is 10 %. The calibration of the two OPSs was performed with PSL particles. The calibration accuracy was found to be $\pm 2.5\%$ for the UHSAS and $\pm 5\%$ for the OPS 3330, in agreement with the estimate given by the manufacturer. The particle number concentration was mainly below the single-particle counting limits reported by the manufacturers of the three instruments. The particle loss in the tubes from the dilution chamber to the different instruments was estimated to be similar for all instruments except for the UHSAS and the OPS 3330, using the Particle Loss Calculator software (PLC, von der Weiden et al., 2009). A size-dependent correction factor was estimated and applied to the UHSAS and the OPS 3330 measurements. Further details are reported in the Supplement.

The particle size distribution was obtained assuming the PSL refractive index, e.g., the PSL-equivalent nominal diameter values for the bin boundaries provided by the manufacturer. The PSL equivalent size distribution was used to calculate the OPS particle scattering coefficient and the OPS particle phase function via optical simulations. The optical simulations were performed with the program MOPSMAP (Gasteiger and Wiegner, 2018), using the option to give the binned size data directly as input. We considered spherical shapes only (Mie code) and the refractive index of PSL

particles was used, i.e., $m = 1.591 + i0.00$ at $\lambda = 450\text{ nm}$, $m = 1.582 + i0.00$ at $\lambda = 525\text{ nm}$, and $m = 1.574 + i0.00$ at $\lambda = 635\text{ nm}$ taken from Devon and Rudin (1987). This choice was done to be consistent with the particle size distribution used as input where the PSL particle refractive index was assumed. To estimate the uncertainty of the simulated optical properties, the simulations were repeated 1000 times modifying randomly the number concentration and the bin diameters of the input data. More detailed information is provided in the Supplement.

4.3 Results of the laboratory closure experiment

The four angular corrections C_{SAE} , C_{polar} , $C_{\text{phase,H}}$, and $C_{\text{phase,OPS}}$ were tested by applying each of them to the Aurora 4000 particle scattering coefficient at $\lambda_G = 525\text{ nm}$, called corrected $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$, and comparing them to the difference between the particle extinction coefficient and the particle absorption coefficient ($\sigma_{\text{ep}} - \sigma_{\text{ap}}$), where σ_{ep} and σ_{ap} were measured by a CAPS PM_{ext} at $\lambda = 530\text{ nm}$ and by a TAP at $\lambda = 528\text{ nm}$, respectively. $\sigma_{\text{ep}} - \sigma_{\text{ap}}$ represents the reference value for the particle scattering coefficient as discussed in Sect. 4.1. The ratios of corrected $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$ to $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ as a function of the PSL equivalent volume median diameter (PSL eq. VMD) are shown in Fig. 3. Each row of Fig. 3 (e.g., a–b, c–d, e–f, or g–h) shows results for a different angular correction. The results for monodisperse aerosols (PSL and AS) are shown in the panels on the left (Fig. 3a, c, e, g), and the results for polydisperse aerosols (mineral dust, silica dust, and volcanic ash) in the panels on the right (Fig. 3b, d, f, h). Results of a correlation analysis for each angular correction are reported in Table 4. The slope of linear regression through the origin and the coefficient of determination were calculated for each angular correction vs. $(\sigma_{\text{ep}} - \sigma_{\text{ap}})/\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$, which represents the reference angular correction. The slopes represent the average deviation from 1 of the ratio between the corrected $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$ and the reference particle scattering coefficient. Further details for each correction are described in the following sections.

4.3.1 Results for the angular correction C_{SAE}

The angular correction C_{SAE} (Eq. 13), using the scattering Ångström exponent, was calculated with the parameters in Table 3. For the monodisperse aerosol measurement, we used the parameters for sub-micrometer size cut-off in the case of nominal $d < 1\text{ }\mu\text{m}$ and the one for no size cut-off in the case of nominal $d > 1\text{ }\mu\text{m}$. For polydisperse aerosol measurements, we used the parameters for no size cut-off.

The $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$ corrected with the angular correction C_{SAE} agrees with $\sigma_{\text{ep}} - \sigma_{\text{ap}}$ within $\pm 15\%$ for most of the measurements (Fig. 3a, b). In particular, the agreement is excellent for polydisperse measurement with a volume median diameter smaller than 2 μm , with almost all values of the ratio between corrected $\sigma_{\text{sp}}^{\text{Aurora 4000}, 0^\circ}$ and $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ equal

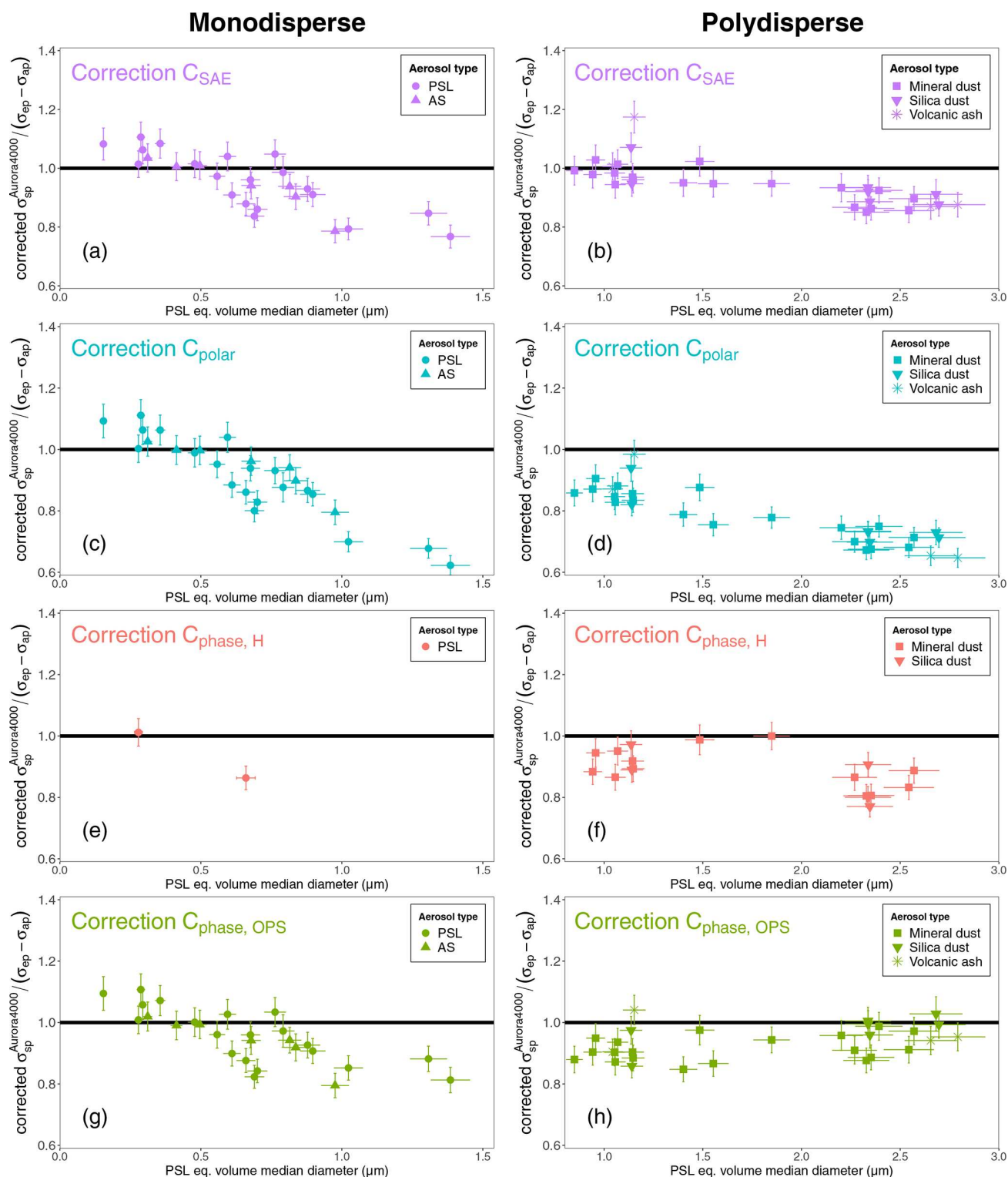


Figure 3. Result of the laboratory experiment for each angular correction. Ratio of the Aurora 4000 particle scattering coefficient corrected for angular truncation and illumination error ($\text{corrected } \sigma_{\text{sp}}^{\text{Aurora4000},0^\circ}$) and the difference between the particle extinction coefficient and the particle absorption coefficient ($\sigma_{\text{ep}} - \sigma_{\text{ap}}$) vs. the measured PSL equivalent volume median diameter. Each panel row shows the results for the $\sigma_{\text{sp}}^{\text{Aurora4000},0^\circ}$ corrected with a different angular correction: (a, b) for the angular correction C_{SAE} ; (c, d) for the angular correction C_{polar} ; (e, f) for the angular correction $C_{\text{phase,H}}$; (g, h) for the angular correction $C_{\text{phase,OPS}}$. Monodisperse aerosol measurements are reported in the panels on the left (a, c, e, g); polydisperse aerosol measurements are reported in the panels on the right (b, d, f, h). Error bars represent the 95 % confidence interval. Different symbols refer to different aerosol types.

Table 4. Results of the regression analysis ($C_{\text{columns}} = \text{slope}(\sigma_{\text{ep}} - \sigma_{\text{ap}})/\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$ and the coefficient of determination R^2) of each angular correction in the table columns C_{columns} and the ratio between the reference particle scattering coefficient vs. the Aurora 4000 particle scattering coefficient, $(\sigma_{\text{ep}} - \sigma_{\text{ap}})/\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$.

			C_{columns}			
			C_{SAE}	C_{polar}	$C_{\text{phase,H}}$	$C_{\text{phase,OPS}}$
$(\sigma_{\text{ep}} - \sigma_{\text{ap}})/\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$	Monodisperse	Slope	0.92 ± 0.02	0.87 ± 0.03	–	0.93 ± 0.02
		R^2	0.989	0.978	–	0.992
	Polydisperse	Slope	0.93 ± 0.01	0.77 ± 0.02	0.88 ± 0.02	0.93 ± 0.01
		R^2	0.995	0.987	0.994	0.997

to 1 within the expected uncertainties (Fig. 3b). For a larger volume median diameter, the angular correction C_{SAE} leads to an underestimation of $\sigma_{\text{ep}} - \sigma_{\text{ap}}$ by up to 15 %, showing a slightly size-dependent performance. For monodisperse aerosol larger than $1 \mu\text{m}$, the corrected $\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$ underestimates $\sigma_{\text{ep}} - \sigma_{\text{ap}}$ by about 20 % (Fig. 3b). Results from the linear regression analysis show that the angular correction C_{SAE} underestimates the reference angular correction on average by 8 % for the monodisperse aerosol measurements and by 7 % for the polydisperse aerosol measurements (Table 4).

4.3.2 Results for the angular correction C_{polar}

The angular correction C_{polar} (Eq. 14), using the polar measurements of the Aurora 4000 particle scattering coefficients, shows a performance that strongly depends on particle size for both monodisperse and polydisperse aerosol. The agreement between corrected $\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$ and $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ is within $\pm 10 \%$ for small sizes (PSL eq. $\text{VMD} < 600 \text{ nm}$). Conversely, for larger sizes the corrected $\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$ underestimates $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ by up to 40 % (Fig. 3c, d). Results from the linear regression analysis show that the angular correction C_{polar} underestimates the reference angular correction on average by 13 % for the monodisperse aerosol measurements and by 23 % for the polydisperse aerosol measurements (Table 4).

4.3.3 Results for the angular correction $C_{\text{phase,H}}$

The angular correction $C_{\text{phase,H}}$, using the phase function measured by Horvath's polar nephelometer, shows a performance that does not depend on particle size but generally underestimates $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$. The corrected $\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$ is in agreement with $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ for only a few measurements, while it underestimates $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ by 23 % in the worst case (Fig. 3e, f). Results from the linear regression analysis show that the angular correction $C_{\text{phase,H}}$ underestimates the reference angular correction on average by 12 % for the polydisperse aerosol measurements (Table 4).

4.3.4 Results for the angular correction $C_{\text{phase,OPS}}$

The angular correction $C_{\text{phase,OPS}}$, using the particle phase function simulated considering the UHSAS and OPS PSL equivalent size distributions, shows a performance that does not depend on particle size. The corrected $\sigma_{\text{sp}}^{\text{Aurora 4000, 0}^\circ}$ agrees within $\pm 20 \%$ with $(\sigma_{\text{ep}} - \sigma_{\text{ap}})$ for all sizes and aerosol types. In particular, the agreement for the large size ($\text{VMD} > 2 \mu\text{m}$) of the polydisperse aerosol measurement is within $\pm 10 \%$ (Fig. 3g, h). Results from the linear regression analysis show that the angular correction $C_{\text{phase,OPS}}$ underestimates the reference angular correction on average by 7 % for both the monodisperse and the polydisperse aerosol measurements (Table 4).

4.3.5 Results of the comparison among different angular corrections

To compare different angular corrections, we performed a correlation analysis among all the considered angular corrections. The slopes of linear regression through the origin and coefficients of determination obtained are reported in Table 5 for monodisperse aerosols and in Table 6 for polydisperse aerosols. In Tables 5 and 6, results highlighted in bold indicate an agreement within $\pm 3 \%$.

The angular corrections C_{SAE} and C_{polar} agree within $\pm 3 \%$ with the corresponding OPS-simulated angular corrections, called $C_{\text{SAE,OPS}}$ and $C_{\text{polar,OPS}}$ (see Fig. 2 and related text in Sect. 4.1), both for the monodisperse and polydisperse cases. The angular correction $C_{\text{phase,OPS}}$ agrees within $\pm 3 \%$ with C_{SAE} , $C_{\text{SAE,OPS}}$, and $C_{\text{polar,OPS}}$ for the monodisperse aerosol measurements and with C_{SAE} and $C_{\text{SAE,OPS}}$ for the polydisperse aerosol measurements.

The angular correction $C_{\text{phase,H}}$, obtained with the phase function measured with Horvath's polar nephelometer, agrees within 6 % with the angular correction $C_{\text{phase,OPS}}$, obtained from the OPS-simulated phase function for the polydisperse measurements.

In the case of the polydisperse aerosol measurements, the agreement within $\pm 3 \%$ between each angular correction and the corresponding OPS-simulated angular correction is sur-

Table 5. Monodisperse aerosol: PSL and AS. Results of the regression analysis ($C_{\text{rows}} = \text{slope} \cdot C_{\text{columns}}$ and coefficient of determination R^2) of each angular correction reported in the table rows C_{rows} vs. each angular correction reported in the table columns C_{columns} . Results within $\pm 3\%$ are highlighted in bold.

C_{columns}		C_{rows}				
		C_{SAE}	C_{polar}	$C_{\text{SAE,OPS}}$	$C_{\text{polar,OPS}}$	$C_{\text{phase,OPS}}$
C_{SAE}	slope	1	1.04 ± 0.01	0.995 ± 0.002	1.02 ± 0.01	0.996 ± 0.005
	R^2	1	0.996	0.99999	0.998	0.999
C_{polar}	slope	–	1	0.95 ± 0.01	0.978 ± 0.005	0.95 ± 0.02
	R^2	–	1	0.995	0.999	0.993
$C_{\text{SAE,OPS}}$	slope	–	–	1	1.03 ± 0.01	1.002 ± 0.004
	R^2	–	–	1	0.997	0.9996
$C_{\text{polar,OPS}}$	slope	–	–	–	1	0.97 ± 0.01
	R^2	–	–	–	1	0.995

Table 6. Polydisperse aerosol: mineral dust, silica dust, and volcanic ash. Results of the regression analysis ($C_{\text{rows}} = \text{slope} \cdot C_{\text{columns}}$ and coefficient of determination R^2) of each angular correction reported in the table rows C_{rows} vs. each angular correction reported in the table columns C_{columns} . Results within $\pm 3\%$ are highlighted in bold.

C_{col}		C_{row}					
		C_{SAE}	C_{polar}	$C_{\text{phase,H}}$	$C_{\text{SAE,OPS}}$	$C_{\text{polar,OPS}}$	$C_{\text{phase,OPS}}$
C_{SAE}	slope	1	1.22 ± 0.01	1.06 ± 0.01	0.973 ± 0.004	1.22 ± 0.01	0.99 ± 0.02
	R^2	1	0.997	0.997	0.999	0.997	0.993
C_{polar}	slope	–	1	0.88 ± 0.02	0.802 ± 0.004	1.004 ± 0.002	0.82 ± 0.02
	R^2	–	1	0.995	0.999	0.9998	0.983
$C_{\text{phase,H}}$	slope	–	–	1	0.91 ± 0.01	1.14 ± 0.02	0.94 ± 0.02
	R^2	–	–	1	0.997	0.994	0.994
$C_{\text{SAE,OPS}}$	slope	–	–	–	1	1.252 ± 0.007	1.03 ± 0.02
	R^2	–	–	–	1	0.999	0.989
$C_{\text{polar,OPS}}$	slope	–	–	–	–	1	0.82 ± 0.02
	R^2	–	–	–	–	1	0.984

prisingly good. First, the refractive index of the calibration material was assumed in the size distribution retrieval. Second, we did not consider any irregular shape effect, even if our polydisperse aerosols are irregularly shaped. Based on previous closure experiments (Schladitz et al., 2009, 2011), we did not expect to achieve good agreement using Mie theory for irregularly shaped particles. This result leads us to the following questions: what is the impact of particle shape on the nephelometer angular truncation and illumination error? What is the impact of different refractive indices and shapes on the angular correction $C_{\text{phase,OPS}}$ derived from the size distribution measured by an OPS? What is the effect of different possible approaches (e.g., assuming different refractive indices) to obtain the OPS size distribution and the simulated angular correction $C_{\text{phase,OPS}}$? To answer these questions and interpret the laboratory experiment results, we

performed an extensive modeling effort simulating the TSI OPS 3330 response for several particle types.

5 Simulated closure experiment

5.1 Simulation methods

We conducted a simulated closure experiment to answer the questions raised by the laboratory experiment result, in particular concerning the effect of particle shape on the determination of the particle scattering coefficient. We estimated the effect of the particle shape on the “original” angular correction C_{0° (see Eq. 10 with $\alpha = 0^\circ$) and on the two methods to calculate the angular correction that showed the best results for mineral dust aerosol during the laboratory experiment: the C_{SAE} and $C_{\text{phase,OPS}}$. The overall goal of these simula-

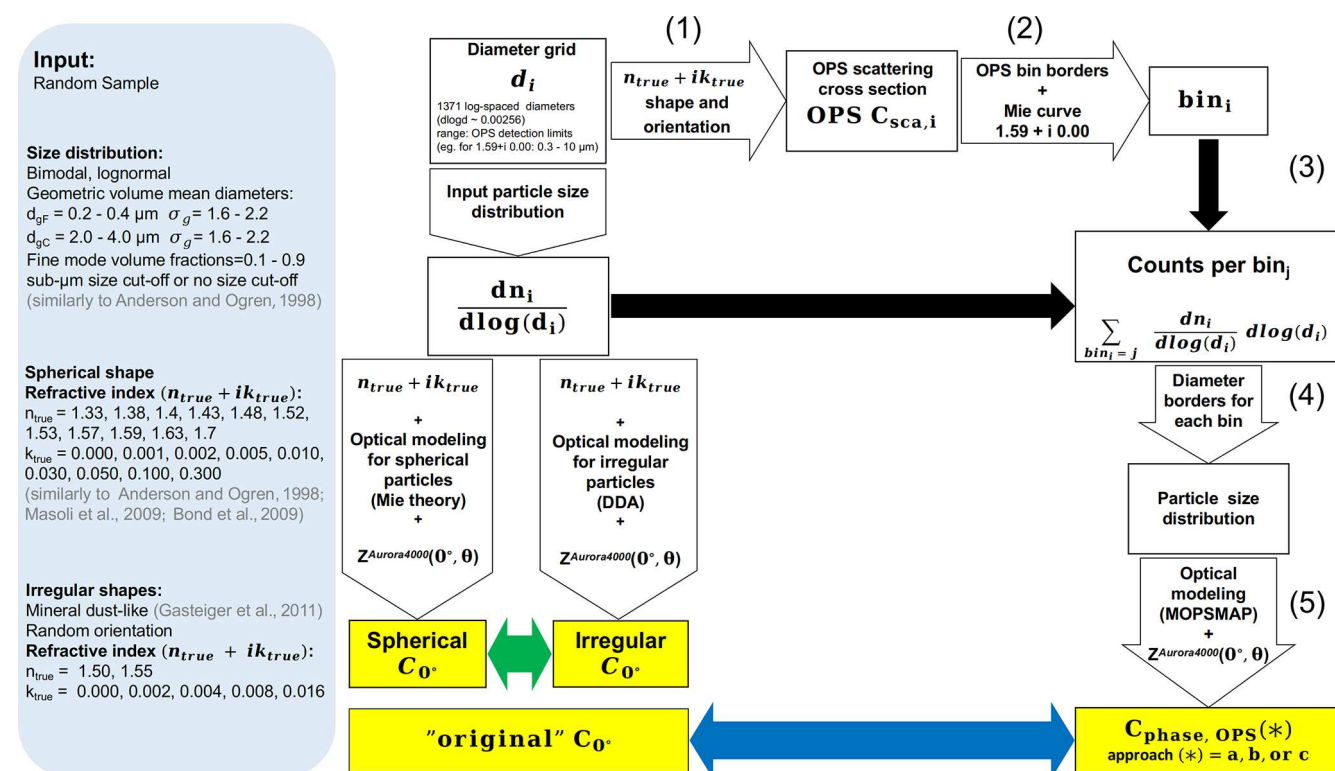


Figure 4. Flow chart for the simulated closure experiment. The input parameters are selected randomly among the values indicated in the left box. The “original” angular correction C_{0° is calculated directly on a given diameter grid for the input size distributions and refractive indices (left side of the flow chart), for homogeneous spherical particles with Mie calculations (spherical C_{0°), and for irregular particles with discrete dipole approximation (irregular C_{0°). To simulate the angular correction $C_{phase, OPS}$ corresponding to each original C_{0° considered, the steps 1–5 are performed. Different approaches to run the five steps are possible.

tions is to estimate the effect of the different approaches on the angular correction $C_{phase, OPS}$, e.g., assuming different refractive indices for the particle size distribution retrieval.

The flow chart for this simulated closure experiment is represented in Fig. 4. Several size distributions and refractive indices were selected randomly among values indicated in the left box of Fig. 4. The original angular correction C_{0° was calculated directly on a given diameter grid for the input particle size distributions, refractive indices, and shapes. For homogeneous spherical particles, we hereafter will call it spherical C_{0° and for irregular particles irregular C_{0° . The irregular C_{0° was compared with the spherical C_{0° obtained for volume-equivalent spheres with the same particle size distribution and refractive index (green arrow, Fig. 4). The response of the TSI OPS 3330 and the angular correction $C_{phase, OPS}$ for the selected particle size distributions and particle types were simulated (step 1–5 in Fig. 4). The angular corrections $C_{phase, OPS}$ obtained with different approaches were compared with the original angular correction C_{0° obtained for the same aerosol types (blue arrow, Fig. 4). We performed the same comparison for the angular correction C_{SAE} .

We selected 5000 random samples with particle size distributions and refractive indices similar to previous studies on the nephelometer angular correction (Anderson and Ogren, 1998; Müller et al., 2011a; Bond et al., 2009; Massoli et al., 2009). The particle size distributions were bimodal log-normal with the geometric volume mean diameter varying in the range of 0.2–0.4 μm for the fine mode and in the range of 2.0–4.0 μm for the coarse mode. The geometric standard deviation varied in the range of 1.6–2.2. Fine-mode volume fraction varied in the range of 0.1–0.9. Half of the simulations were performed with sub-micrometer size cut-off, half of them with no size cut-off. The real part of the refractive index varied in the range of 1.33–1.70 and the imaginary part in the range of 0.0–0.3. Calculations were performed for homogeneous spherical particles using the miepython module (Prah, 2020), developed by Scott Prah and based on Wiscombe (1980).

Additionally, we selected 1000 random samples with refractive indices similar to mineral dust aerosol (e.g., Di Biagio et al., 2019): the real part of the refractive index was chosen between 1.50 and 1.55, and the imaginary part was selected in the range of 0.000–0.016. The calculations for these samples were repeated twice for homogeneous spheri-

cal particles and for irregular shapes selected among mineral dust shapes from Gasteiger et al. (2011) and using the discrete dipole approximation code (ADDA, Yurkin and Hoekstra, 2011).

The selected values of the refractive index for each case are referred to hereafter in the text as well as in Fig. 4 as the “true” refractive index ($n_{\text{true}} + ik_{\text{true}}$).

Calculations were performed on a diameter grid with size range corresponding to the TSI OPS detection limits. The OPS diameter detection limits corresponding to each $n_{\text{true}} + ik_{\text{true}}$ were calculated via Mie theory considering the manufacturer-provided diameter detection limits of 0.3–10 μm for homogeneous spherical particles with a refractive index $1.59 + i0.00$. In the case of irregularly shaped particles, the ADDA calculations were limited up to a volume-equivalent diameter of 5.8 μm .

Although particles smaller than 0.3 μm are still relevant for the calculation of the angular correction, here we limited the calculations to the OPS size range because we would like to assess the impact of the assumptions of refractive index and shapes in calculating the angular correction from the size distribution measured by an OPS. Considering a size range of 0.06–10 μm and combining the size distribution of UHSAS and OPS 3330, we obtained similar results, but we did not report these results here.

For each randomly sampled set of particle size distribution, refractive index, and shape, the response of the TSI OPS 3330 was simulated (step 1–2–3, Fig. 4). An optical particle spectrometer, such as the TSI OPS 3330, measures the scattering cross section (OPS C_{sca}) by collecting the light scattered in the angular range 30–150° with a mirror. We simulated the OPS $C_{\text{sca},i}$ for each diameter of the grid d_i (step 1, Fig. 4), using a (custom) Mie code based on Bohren and Huffman (2008) for homogeneous spherical particles and the ADDA code from Yurkin and Hoekstra (2011) for the mineral-dust-like shapes from Gasteiger et al. (2011). In the latter, the orientations and shapes were selected randomly for each d_i . Each simulated OPS C_{sca} was sorted into the corresponding OPS bin (step 2, Fig. 4). In order to perform this step, we had to convert the manufacturer-provided diameter bin boundaries for the calibration material ($m = 1.59 + i0.00$) into scattering cross-section bin boundaries. The randomly sampled size distribution was considered only in the following step, to reduce the computational time (step 3, Fig. 4). As a result, we obtained the counts for each of the 16 bins of the TSI OPS 3330. Based on this simulated OPS histogram, we used three different approaches to calculate the particle size distribution (step 4, Fig. 4) and the angular correction $C_{\text{phase,OPS}}$ (step 5, Fig. 4).

Approach a: The particle size distribution was obtained (step 4, Fig. 4) with the manufacturer-provided PSL-equivalent nominal diameter values for the bin boundaries. The angular correction $C_{\text{phase,OPS}}$ was calculated (step 5, Fig. 4) via Mie theory with the refractive index

of the calibration material, i.e., the refractive index of PSL particles ($1.59 + i0.00$).

Approach b: The particle size distribution was obtained (step 4, Fig. 4) with the manufacturer-provided PSL-equivalent nominal diameter values for the bin boundaries. The angular correction $C_{\text{phase,OPS}}$ was calculated (step 5, Fig. 4) via Mie theory with the true refractive index, i.e., the randomly selected refractive index used as a input.

Approach c: The particle size distribution was obtained (step 4, Fig. 4) with diameter values of the bin boundaries calculated through an inversion procedure. As a refractive index, we assumed the true refractive, i.e., the randomly selected refractive index used as a input. The angular correction $C_{\text{phase,OPS}}$ was calculated (step 5, Fig. 4) via Mie theory with the true refractive index.

In addition, we calculated the angular correction C_{SAE} for the same size distributions and aerosol types. For the calculation of the correction C_{SAE} the parameters obtained with sub-micrometer size cut-off or with no size cut-off are used in the corresponding case.

Further details of the method used for this sensitivity study are described in the Supplement.

5.2 Results of the simulated closure experiment

5.2.1 Non-sphericity effect on the angular correction

To estimate the effect of the particle shape on the nephelometer angular correction, we compared the angular correction for randomly oriented mineral-dust-like irregular particles with the angular correction obtained for the volume-equivalent spheres with the same size distribution and refractive index (green arrow, Fig. 4). In the case of size distribution with sub-micrometer cut-off, the angular correction obtained with Mie simulation (i.e., for spherical particles) underestimates the angular correction for irregular particles by 1 % on average as shown in Fig. 5a. In the case of size distribution with no cut-off, the angular correction obtained with Mie simulation (e.g., for spherical particles) underestimates the angular correction on average by 2 % and up to 3.5 %, as shown in Fig. 5b. These results indicate that the effect of the non-spherical shape contributes to an uncertainty on the angular correction smaller than 3.5 %.

5.2.2 Results of the different approaches to calculate $C_{\text{phase,OPS}}$

The $C_{\text{phase,OPS}}$ calculated using the three different approaches (a, b, c; Sect. 5.1) were compared with the original angular correction C_{0° calculated on the input size distribution considering the true refractive index and shape. For input particles with a spherical shape, the comparison is done with

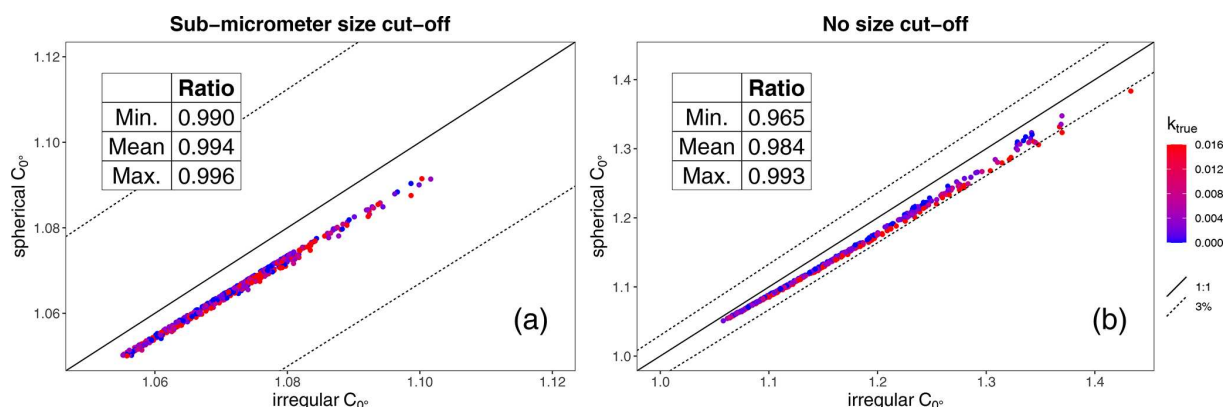


Figure 5. Comparison between the angular correction for irregular particles and for spherical particles. Calculations are performed for $\lambda = 525$ nm and on a diameter range of 0.06–5.8 μm . Panel (a) shows the comparison for particle size distributions with sub-micrometer size cut-off. Panel (b) shows the comparison for particle size distributions with no size cut-off. The color indicates the imaginary part of the refractive index used in the simulations k_{true} . The solid line is the 1 : 1 line. The dashed lines are the $\pm 3\%$ lines. The minimum, mean, and maximum value of the ratio between spherical C_0 and irregular C_0 is reported in the table.

the Mie-calculated spherical C_0 ; for input particles with irregular shape the comparison is done with the ADDA calculated irregular C_0 (blue arrow, Fig. 4). The results of this comparison are summarized in Fig. 6 for the size distribution with sub-micrometer size cut-off and in Fig. 7 for the size distribution with no size cut-off. In each column, the comparison for an angular correction $C_{\text{phase,OPS}}$ calculated with one of the three different approaches (see Sect. 5.1) or for the angular correction C_{SAE} is shown. The six panel rows of Figs. 6 and 7 report results for six aerosol categories:

1. non-absorbing spherical particles, with $n_{\text{true}} = 1.59$;
2. non-absorbing spherical particles, with $n_{\text{true}} = 1.33$ –1.70;
3. weakly absorbing spherical particles with $n_{\text{true}} = 1.33$ –1.70 and $k_{\text{true}} = 0.001$ –0.010;
4. absorbing spherical particles with $n_{\text{true}} = 1.33$ –1.70 and $k_{\text{true}} = 0.030$ –0.300;
5. weakly absorbing spherical particles with mineral-dust-like refractive index $n_{\text{true}} = 1.50$ –1.55 and $k_{\text{true}} = 0.000$ –0.016;
6. weakly absorbing irregular particles with mineral-dust-like shape and refractive index $n_{\text{true}} = 1.50$ –1.55 and $k_{\text{true}} = 0.000$ –0.016.

Several observations can be made looking at the results represented in Figs. 6 and 7. First, the panels in the first row of Figs. 6 and 7 show that the effect of the TSI OPS 3330 binning on the angular correction is smaller than 0.7 %. Indeed, $1.59 + i0.000$ is the refractive index used for the calibration of the TSI OPS 3330 and, in this case, the difference between C_0 and $C_{\text{phase,OPS}}$ is the result of the TSI OPS 3330 binning.

Second, if the particle size distribution is calculated with the calibration refractive index, the use of the true refractive index in the optical modeling of the angular correction $C_{\text{phase,OPS}}$ does not improve the accuracy significantly. Indeed, panels in the first and second columns (approaches a and b) look quite similar for all aerosol types, both with sub-micrometer and with no size cut-off.

Third, if the particle size distribution is calculated with the true refractive index, the agreement between $C_{\text{phase,OPS}}$ and the original C_0 significantly improves, for all cases with spherical particles. The agreement is within 3 %–4 % for all weakly absorbing aerosols with spherical particles for both with and with no size cut-off (rows 2 and 3, column 3 of Figs. 6 and 7).

Fourth, in the case of absorbing aerosol, the $C_{\text{phase,OPS}}$ (c) calculated with the refractive index correction can have large uncertainties. In the case of size distributions with sub-micrometer size cut-off, when $k_{\text{true}} = 0.1$ –0.3, $C_{\text{phase,OPS}}$ (c) overestimates C_0 by up to 21 % (row 4, column 3 of Fig. 6). The comparison plot shows unusual features as the retrieval procedure used for the refractive index correction failed. Indeed, the TSI OPS 3330 Mie curve for $k_{\text{true}} = 0.3$ is almost flat in the diameter range from 0.5 to 1.0 μm . The angular correction C_{SAE} is accurate within 4 % in this case. In the case of size distributions with no size cut-off, $C_{\text{phase,OPS}}$ (c) can have up to $\pm 25\%$ uncertainty, when $k_{\text{true}} = 0.1$ –0.3 (row 4, column 3 of Fig. 7).

Finally, an interesting result is found in the case of irregular particles when no size cut-off is considered (last panel row of Fig. 7). The angular correction $C_{\text{phase,OPS}}$ (a), obtained with the refractive index of the calibration material, agrees on average with the original C_0 within 0.3 %. However, the relationship is strongly dependent on the imaginary part of the refractive index. If $k_{\text{true}} = 0.000$, $C_{\text{phase,OPS}}$ (a) overestimates the original C_0 up to 8 %. If $k_{\text{true}} = 0.016$, $C_{\text{phase,OPS}}$

Sub-micrometer size cut-off

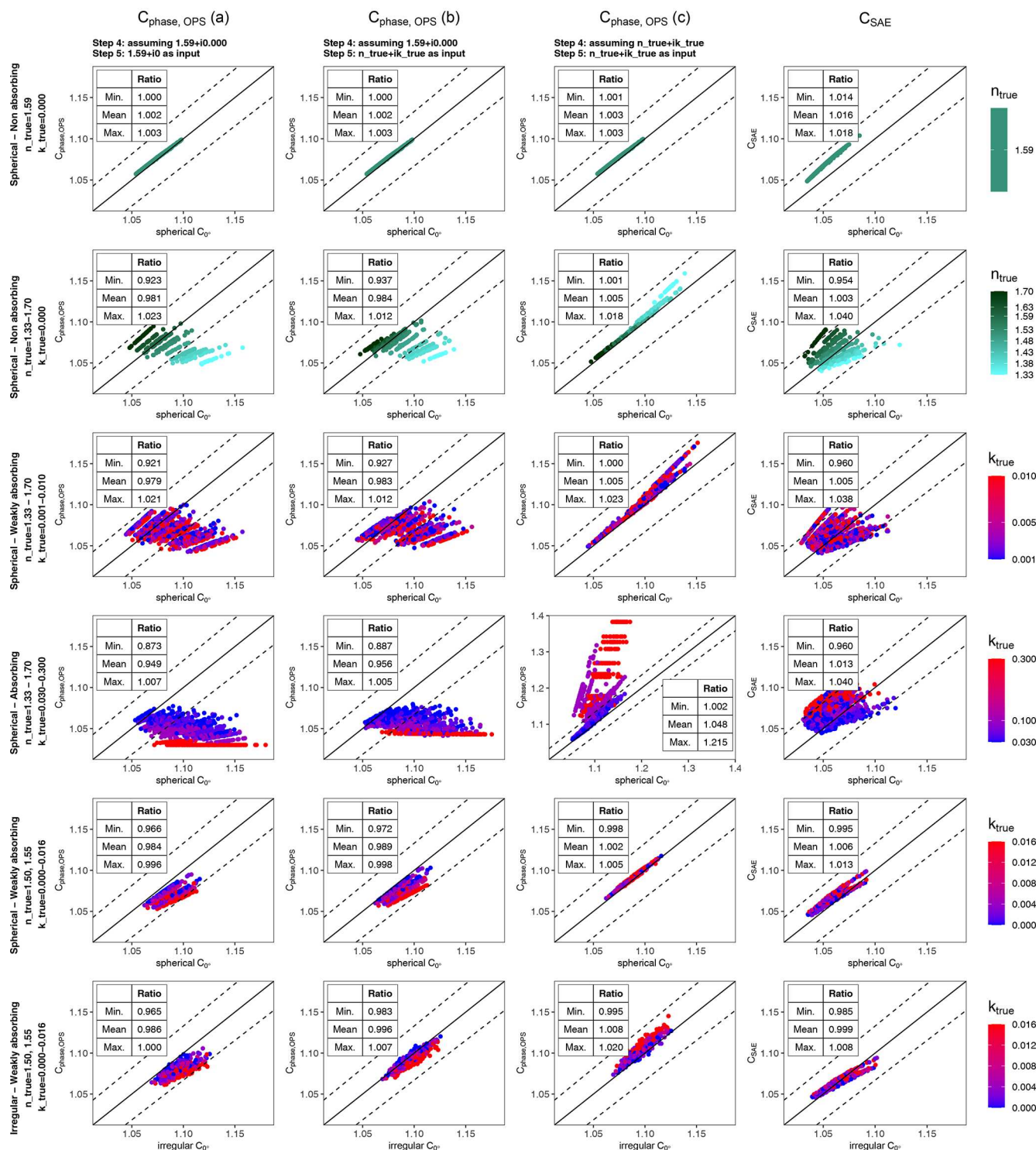


Figure 6. Simulated closure experiment to assess the limitations and uncertainties of the angular correction obtained with Mie theory from the size distribution measured by an OPS. The comparisons for the angular correction $C_{\text{phase, OPS}}$ calculated with the three approaches (a, b, c) are shown in the first three columns. The comparison for the angular correction C_{SAE} is reported in the last column. Different lines refer to different aerosol input. The color code indicates different values of the real part of the refractive index for the non absorbing cases (rows 1–2) and of the imaginary part of the refractive index for all the other cases (rows 3–6). The solid lines are the 1:1 lines. The dashed lines are the $\pm 3\%$ lines. The minimum, mean, and maximum value of the ratio between the quantity reported on the y axis and the quantity reported on the x axis is reported in the tables.

No size cut-off

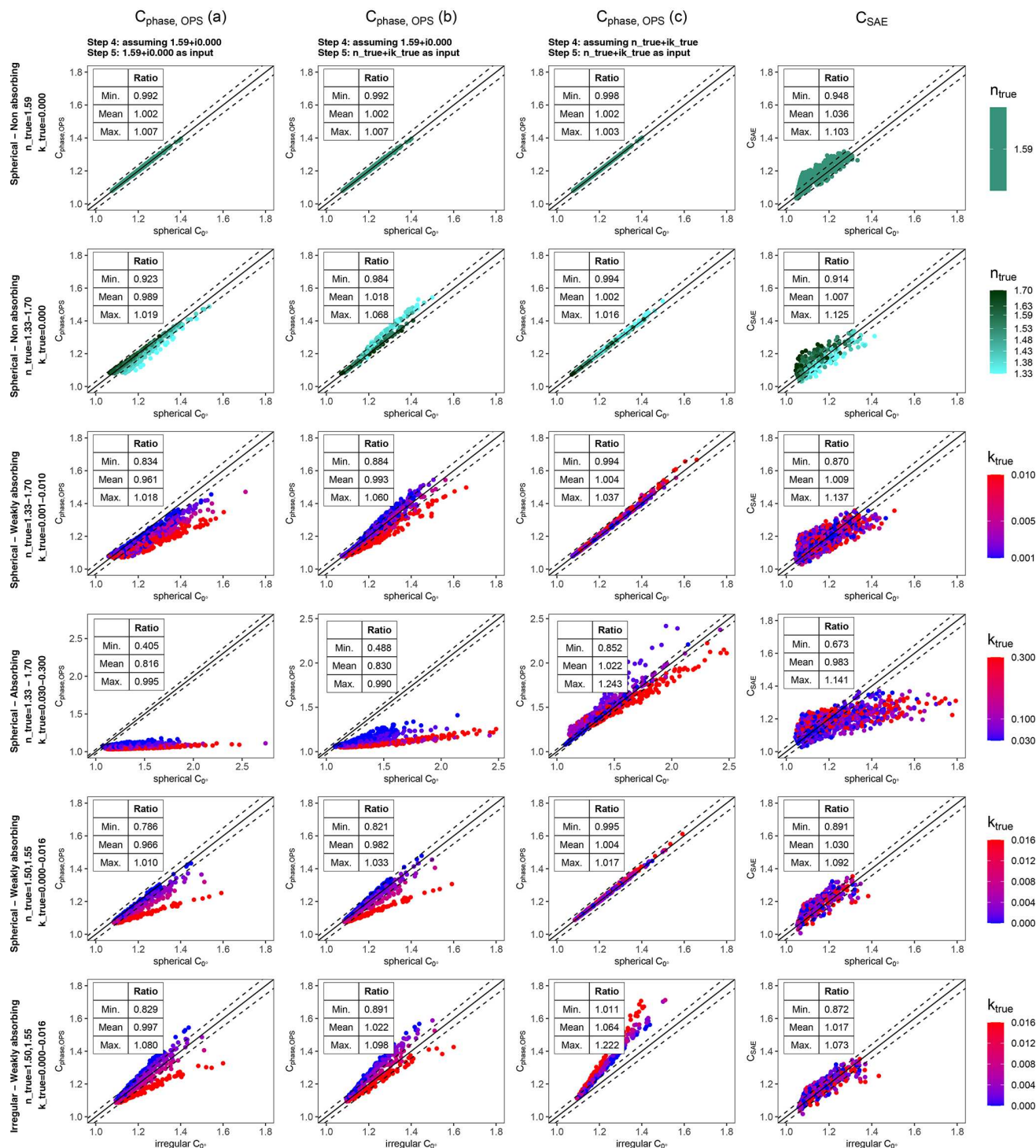


Figure 7. Simulated closure experiment to assess the limitations and uncertainties of the angular correction obtained with Mie theory from the size distribution measured by an OPS. The comparisons for the angular correction $C_{\text{phase, OPS}}$ calculated with the three approaches (a, b, c) are shown in the first three columns. The comparison for the angular correction C_{SAE} is reported in the last column for comparison. Different rows refer to different aerosol input. The color code indicates different values of the real part of the refractive index for the non absorbing cases (rows 1–2) and of the imaginary part of the refractive index for all the other cases (rows 3–6). The solid lines are the 1 : 1 lines. The dashed lines are the $\pm 3\%$ lines. The minimum, mean, and maximum value of the ratio between the quantity reported on the y axis and the quantity reported on the x axis is reported in the tables. Calculations are limited for computational purposes to particle diameter ranges of 0.3–10 and 0.3–5.8 μm for homogeneous spherical particles and for irregular particles.

(a) underestimates the original C_{0° up to 18 %. Surprisingly, when the true refractive index is taken into account, the angular correction $C_{\text{phase,OPS}}$ (c) systematically overestimates the original C_{0° on average by 5 % and up to 22 % in the worst case.

6 Discussion

6.1 Nephelometer angular corrections

Integrating nephelometer measurements require an angular correction to provide the particle scattering coefficient. Several methods to calculate the angular correction are available in the literature and are covered by our laboratory and modeling study. The applicability and uncertainty of these corrections depend on the particle type and the availability of other measurements.

The angular correction C_{polar} is a method developed by Müller et al. (2012). Nephelometer data corrected with the angular correction C_{polar} (Fig. 3c–d) are in agreement with the reference particle scattering coefficient only for particles with sizes smaller than 1 μm , while they strongly underestimate (up to $\sim 40\%$) the particle scattering coefficient for larger particle sizes. This observation is in agreement with the model calculation performed by Müller et al. (2012). Müller et al. (2012) also showed that this correction is not affected by the imaginary part of the refractive index and that it can reduce the uncertainty of the angular correction for sub-micrometer to about 2 %.

The angular correction C_{SAE} is the method most commonly used in the literature (e.g., Pandolfi et al., 2018; Valenzuela et al., 2015; Costabile et al., 2013; Clarke et al., 2007). Nephelometer data of polydisperse coarse-mode irregularly shaped aerosols corrected with the angular correction C_{SAE} (Fig. 3b) are in very good agreement with the reference particle scattering coefficient for small volume median diameters ($< 2\ \mu\text{m}$). For volume median diameters larger than 2 μm , the reference particle scattering coefficient is underestimated by up to 15 %. This performance is much better than the one achieved with the C_{polar} angular correction. This result is in agreement with the model calculations of weakly absorbing polydisperse aerosol of spherical and irregularly shaped particles (Fig. 7 last panel column, rows 3, 5, and 6), which show that the angular correction C_{SAE} has an overall uncertainty of about $\pm 13\%$. A possible explanation for the underestimation of the angular correction C_{SAE} for large particle sizes (volume median diameter $> 2\ \mu\text{m}$) is that the SAE is already around zero. Even if the SAE can reach slightly negative numbers, it is no longer linearly related to the particle size.

The overall higher uncertainty of the angular correction C_{SAE} for coarse-mode aerosol is already highlighted by Anderson and Ogren (1998). For this reason, they introduce the sub-micrometer size segregation and point out that very

accurate information, within a few percent, on the particle scattering coefficient of sub-micrometer polydisperse non-absorbing aerosol can be achieved, as confirmed experimentally by Massoli et al. (2009).

Nevertheless, the angular correction C_{SAE} is not very accurate in the case of absorbing aerosol. The SAE is only indirectly related to the particle size, and this relationship is altered for absorbing particles. Bond et al. (2009) showed that the angular correction C_{SAE} might have an error of about 5 % for sub-micrometer aerosol. Even if this error looks rather small, Bond et al. (2009) point out that the error is in the same order of magnitude as the angular correction for absorbing sub-micrometer particles. In addition, Massoli et al. (2009) showed this issue experimentally and pointed out that for sub-micrometer particles, the relationship between SAE and angular correction is also strongly dependent on the real part of the refractive index. Our calculations (last panel column of Fig. 6) confirm the finding of Bond et al. (2009) and Massoli et al. (2009). In addition, our calculations for particle size distributions with no size cut-off (Fig. 7) show that, in extreme cases, if coarse-mode highly absorbing particles are present, the uncertainty of the C_{SAE} correction may rise up to 33 %.

The angular correction $C_{\text{phase,OPS}}$ is a rarely used method in the literature (e.g., Di Biagio et al., 2019) as it requires simultaneous particle size distribution measurements and also knowledge about the refractive index and particle shape. For the mineral dust measurements, the performance of the angular correction $C_{\text{phase,OPS}}$ is surprisingly good (Fig. 3h) even though no refractive index or shape is considered in the size distribution retrieval. This result might lead one to the conclusion that there is no need to take into account the refractive index or shape of the measured aerosol for the calculation of the angular correction $C_{\text{phase,OPS}}$. The optical particle spectrometer is an optical cross-section selector rather than a particle sizer. The use of the refractive index of the calibration material might lead to compensations of the effects of refractive index and shapes on the measured particle size distribution (see Supplement in Sect. S3 and Fig. S3) and, thus, on the calculated angular correction. Results of our simulated closure experiment (Fig. 7, last row) suggest that the good performance of the angular correction $C_{\text{phase,OPS}}$ for mineral dust measurements (Fig. 3h) might be partly due to a compensation of the non-spherical shape effect by the small imaginary part of the refractive index. In the case of mineral dust measurements, the particle size distribution by an OPS, obtained with the manufacturer-provided PSL-equivalent nominal diameters of the bin boundaries, leads to an angular correction which is on average correct (within $\pm 0.3\%$). However, $C_{\text{phase,OPS}}$ has a strong dependence on the imaginary part of the refractive index; it overestimates the true angular correction by up to 8 % for non-absorbing particles and underestimates it by up to 17 % for absorbing particles (Fig. 7, last row, approach a).

The direct effect of particle shape on the angular correction itself (i.e., original C_{0°) is very small. Our calculations (Fig. 5) show that the angular correction for dust-like irregularly shaped particles is on average 2 % higher than the angular correction for volume-equivalent spheres. This result is in agreement with the one obtained by Quirantes et al. (2008), who found that angular corrections calculated for oblate particles and prolate spheroids are similar within 1 %–2 % to results obtained for equal-projected-area spheres. The observation that the near-forward scattering is quite insensitive to the particle shape (e.g., Mishchenko et al., 1995) further supports this finding.

If the true refractive index is assumed for the particle size distribution retrieval, the $C_{\text{phase,OPS}}$ is systematically overestimated on average by 5 %–6 % up to a maximum of 22 % (Fig. 7 last row, approach c). This result highlights that even if the direct effect of the particle shape on the angular correction looks rather small, the non-sphericity contributes to a higher uncertainty in the determination of the particle size distribution and thus in the angular correction $C_{\text{phase,OPS}}$.

In the case of irregularly shaped particles, the angular correction C_{SAE} looks more accurate, and it requires less effort than considering the non-sphericity for the retrieval of the size distribution. Our calculations show that the angular correction C_{SAE} is accurate on average within 2 % with an overall uncertainty of about ± 13 % (Fig. 7 last row, last column). In addition, the angular correction C_{SAE} shows a less strong dependence on the imaginary part of the refractive index than the angular correction $C_{\text{phase,OPS}}$ (a), calculated with the refractive index of the calibration material.

Results of the simulated closure experiment show that the angular correction $C_{\text{phase,OPS}}$ calculated with the approach (c), e.g., assuming the true refractive index for both the particle size retrieval and the optical calculations, is more accurate than the ones calculated for the other approaches for all cases except for irregular particles (Fig. 7). The overall uncertainties obtained are within a few percent for slightly absorbing particles, but larger uncertainties (about ± 25 %) are found for absorbing aerosol. Quite similar results were obtained for the other two approaches (a and b), where the angular correction $C_{\text{phase,OPS}}$ is calculated from the PSL equivalent particle size distribution and the optical simulations used the PSL refractive index for the approach (a) and the true refractive index for the approach (b). This result highlights that the angular correction is quite insensitive to the refractive index as also pointed out by Bond et al. (2009), who estimate about 2 % uncertainty due to the refractive index when calculating the angular correction with Mie theory. Our analysis points out that while the angular correction is quite insensitive to both the refractive index and the shape, it is critical to assume the correct refractive index and shape for the particle size distribution retrieval and in the calculation of the angular correction $C_{\text{phase,OPS}}$.

6.2 Implications for optical closure studies

Optical closure studies, where the aerosol optical properties measured directly are compared with the aerosol optical properties simulated using the measured size distribution and optics theory, allow not only the evaluating and minimizing of the measurement uncertainties but also constraining specific parameters. Iteratively comparing the measured particle scattering and absorption coefficient with the corresponding derived quantities, it is possible to retrieve the aerosol refractive index (e.g., Abo Rizeq et al., 2007; Mack et al., 2010).

If the instruments' uncertainties are comparatively high, it is difficult to achieve a perfect closure, and the refractive index retrieval might fail. For instance, Schladitz et al. (2009, 2011) found discrepancies in the range of 32 %–42 % between measured and Mie-simulated optical properties and could not determine an average refractive index in the case of high mineral dust particle concentrations. These studies attributed the discrepancies to a superposition of irregular shape effect on both the nephelometer angular correction and the sizing and counting uncertainty. While Schladitz et al. (2009, 2011) used a combination of a differential mobility particle sizer and an aerodynamic particle sizer, Di Biagio et al. (2019) could retrieve the refractive index of mineral dust within a chamber closure experiment using a combination of two OPSs. Even if they recognized the potential role of non-sphericity in affecting their results, they assumed the mineral dust to be spherical for the whole data treatment.

Our study suggests that while the particle shape affects the angular correction C_{0° only by 2 % (Fig. 5), the overestimation of particle size of mineral dust by optical particle spectrometers can be estimated to result on average in about 5 % up to 22 % overestimation of the angular correction (Fig. 7 last row, approach c). This result allows the decoupling of the effect of the coarse-mode irregular particles on the angular correction itself and on the measured size distribution. Our analysis suggests that the non-sphericity mainly affects the retrieval of the particle size distribution, and therefore a more accurate refractive index might be obtained by considering the shape in the particle size retrieval.

6.3 Limitations of the study

Measurement uncertainties of the measurement instruments Aurora 4000, CAPS PM_{ext} , and TAP might partly explain the observed deviations in the result of the laboratory experiment. An additional source of uncertainty is given by the parameters of the Aurora 4000 angular sensitivity function $Z^{\text{Aurora 4000}}(0^\circ, \theta)$ (see Eq. 5). Indeed, all angular corrections tested rely on the parameters and truncation angles of $Z^{\text{Aurora 4000}}(0^\circ, \theta)$ (Table 1).

Differences in the angular sensitivity function of the Aurora 4000 and the Aurora 3000 (Müller et al., 2011a) lead to a difference of 3 %–6 % in the angular correction C_{SAE} . The truncation angles of Aurora 4000 are 9 and 170° according

to the Aurora 4000 manual, while the truncation angles of Aurora 3000 are 10 and 171° (Müller et al., 2011a). To our knowledge, there is no experimental method to verify these angles. In addition, the parameters of the Aurora 4000 angular illumination function measured by Müller et al. (2012) are different from those of the Aurora 3000 angular illumination function published by Müller et al. (2011a). We cannot exclude that small differences may exist even within different units of the same model. The Aurora 4000 used in this experiment was the Aurora 4000 of the University of Vienna, while the angular sensitivity function was measured on the unit of the TROPOS Institute by Müller et al. (2012).

In our data analysis, we introduced some circularity when the CAPS PM_{ext} was calibrated considering the total particle scattering coefficient for measurements of PSL particles with a nominal diameter of 200 nm, as discussed in Sect. 4.2. The particle scattering coefficient was obtained from the value measured by the Aurora 4000 corrected for the angular truncation and illumination error. The angular correction was calculated with the method C_{phase} , where the particle phase function was obtained via Mie calculations assuming a log-normal particle size distribution and assuming the validity of $Z_{\text{Aurora 4000}}(0^\circ, \theta)$. This correction might contribute to compensations in the comparison between the reference particle scattering coefficient and the corrected Aurora 4000 particle scattering coefficient. Nevertheless, the angular correction increases with particle sizes and the parameters of the angular sensitivity function remain a source of uncertainty for coarse-mode particles.

A method to evaluate the accuracy of the angular sensitivity function could consist of considering the angular correction $C_{\text{phase,H}}$, obtained from the particle phase function measured by Horvath's polar nephelometer. Indeed, the latter does not require assumptions on the refractive index and shape of the measured aerosols. The particle scattering coefficient measured by Horvath's polar nephelometer might be used as a reference value for this purpose. Unfortunately, it was not possible to obtain the particle scattering coefficient from Horvath's polar nephelometer because of the very variable signal during dust measurements and the low temporal resolution (35 min) of the instrument. The angular correction $C_{\text{phase,H}}$ leads to an underestimation of the reference particle scattering coefficient of on average 12 % (Table 4). Even if we do not fully understand the reasons for this discrepancy, we cannot attribute it completely to the uncertainty of the angular sensitivity function. Most probably, the use of the 1 min Aurora 4000 particle scattering coefficient to cope with the very variable signal introduced a high uncertainty. The Horvath's polar nephelometer has been corrected for its angular truncation error considering an extrapolation procedure (Horvath, 2015). Even if the accuracy reported for this procedure is better than 1 %, we cannot exclude being worse for our measurements where the instability of the aerosol number concentration together with the use of coarse-mode irregular particles represents a major challenge.

7 Summary and conclusions

We performed an extensive closure study, including laboratory and modeling effort, to evaluate and compare different angular corrections for the Aurora 4000 polar nephelometer. We focused on their performance for coarse-mode irregularly shaped aerosol such as mineral dust, which is the most abundant aerosol worldwide in terms of dry mass (Choobari et al., 2014; Textor et al., 2006). In the laboratory experiment, we used soil samples collected in desert areas, synthetic silica, and a volcanic ash sample to generate polydisperse test aerosol. In addition, PSL particles and ammonium sulfate were used to generate monodisperse test aerosol. We tested and compared four angular corrections: C_{SAE} using the measured scattering Ångström exponent, C_{polar} using the polar measurements of the particle scattering coefficient, $C_{\text{phase,H}}$ using the particle phase function measured by Horvath's polar nephelometer, and $C_{\text{phase,OPS}}$ using the particle phase function simulated on the basis of the size distributions measured by the two optical particle sizers.

We compared the particle scattering coefficients corrected with each angular correction with the reference particle scattering coefficient obtained as the difference between the measured particle extinction and absorption coefficients. As a result, we found that the angular correction C_{polar} is reliable for sub-micrometer particles while strongly underestimating the true particle scattering coefficient for larger particle sizes. The two angular corrections that showed the best performance for mineral dust measurements are C_{SAE} and $C_{\text{phase,OPS}}$, as they both underestimate the reference angular correction on average by 7 %. The good performance of $C_{\text{phase,OPS}}$ for mineral dust aerosol looks surprising because a simple approach was used for its calculation. The refractive index of the calibration material was assumed for both the particle size distribution retrieval and the optical calculations. In addition, the non-spherical shape was not considered.

To interpret these results, we performed an extensive modeling effort, simulating a closure experiment for randomly selected particle size distributions, refractive indices, and shapes. First, we estimated the effect of the particle shape on the original angular correction C_{0° . We found that using the Mie theory to calculate the angular correction for mineral-dust-like irregular shapes leads to an underestimation of about 2 %. Second, we investigated the uncertainties of the two angular corrections C_{SAE} and $C_{\text{phase,OPS}}$ that showed the best performance for mineral dust measurements. The signal measured by a TSI OPS 3330 was simulated for the selected particle size distributions, refractive indices, and shapes.

The angular correction $C_{\text{phase,OPS}}$ was calculated with three different approaches: (a), (b), and (c) (see Fig. 4 and related text). Overall, the angular correction $C_{\text{phase,OPS}}$ (c), which considers the refractive index of the selected aerosol for both the particle size distribution retrieval and the optical

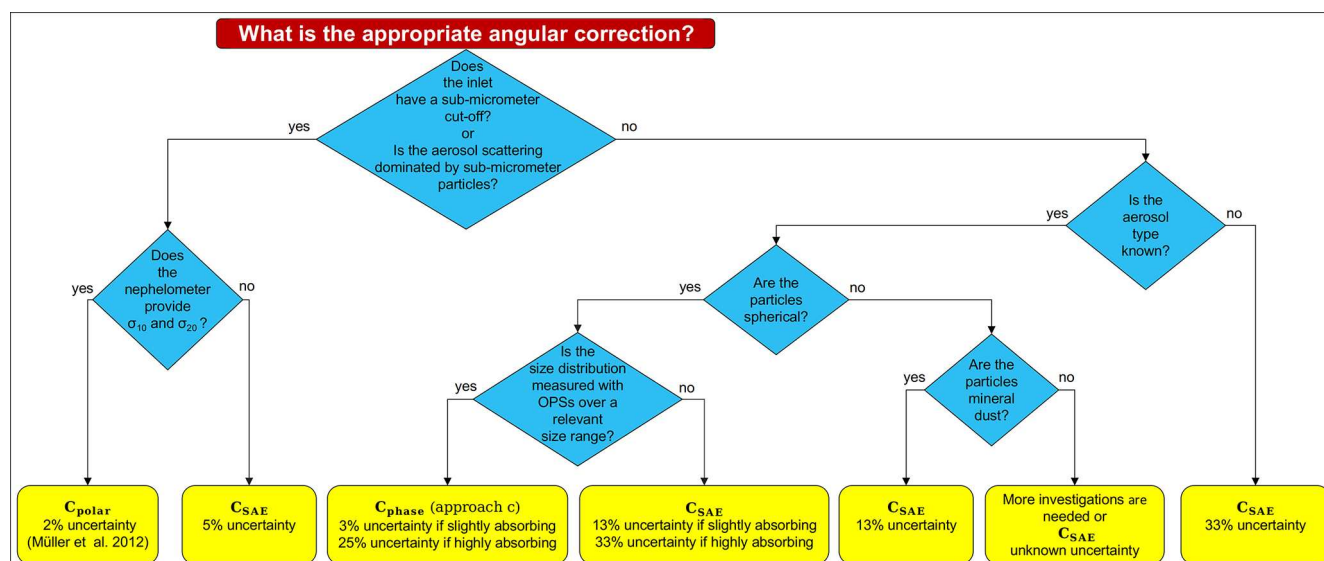


Figure 8. Flow chart to decide which angular correction is appropriate in different situations. The uncertainty related to the appropriate angular correction is identified on the basis of the results of the sensitivity study reported in Figs. 6 and 7.

calculations, leads to an improvement in the agreement between the derived angular correction and the original angular correction C_{0° . An exception is the case of irregular particles, where the angular correction $C_{\text{phase,OPS}}$ (c) overestimates the original angular correction C_{0° on average by 5 % and up to 22 %. Using the angular correction $C_{\text{phase,OPS}}$ (a) (i.e., the refractive index of the calibration material was assumed for both the particle size distribution retrieval and the optical calculations), the effect of the non-spherical shape seems to be almost compensated for by the effect of the imaginary part of the refractive index. In conclusion, through an extensive closure experiment and modeling effort, we could decouple the effect of the coarse-mode irregular particles on the angular correction itself and on the measured size distribution.

7.1 Recommendation for the use of an appropriate correction method

Based on the current knowledge and the present work, we conclude that there is no generally “best” method to calculate the angular correction. Rather the angular correction should be selected depending on the aerosol type and the investigated size range. The angular correction C_{SAE} is the most accurate in many of the analyzed cases and may serve as the “default” method, even if its uncertainty can be relatively large (up to 33 %). If higher accuracy of the particle scattering coefficient by a nephelometer is desired, the angular correction might be obtained via optical simulations from the measured aerosol size distribution $C_{\text{phase,OPS}}$. However, the retrieval of the particle size distribution from the OPS would need to consider the correct refractive index and the particle shape, even if the direct effect of the particle shape on the

nephelometer angular truncation and illumination error was found to be comparatively small.

We provide uncertainties for different approaches to calculate the angular correction from the measured particle size distribution, and we indicate the least uncertain approach for several situations. The reported uncertainties are calculated on the basis of the simulated closure experiment and do not consider the uncertainty of the angular sensitivity function. We recommend the procedure shown in the flow chart in Fig. 8 to establish the nephelometer data correction method.

If the measurements are performed with a sub-micrometer cut-off or if the aerosol scattering is dominated by sub-micrometer aerosols,

- and the nephelometer provides the scattering coefficient for various angular ranges (in particular $\sigma_{\text{sp}}^{\text{Aurora 4000}, 10^\circ}$ and $\sigma_{\text{sp}}^{\text{Aurora 4000}, 20^\circ}$), the angular correction C_{polar} can be used. The uncertainty for this method is 2 % (Müller et al., 2012).
- and the nephelometer does not provide the scattering coefficient for various angular ranges, the angular correction can be calculated with the C_{SAE} method, with 5 % uncertainty.

A size segregation into sub-micrometer and total aerosol is often motivated by technical issues (e.g., the inlet efficiency). However, a sub-micrometer cut-off may also be applied on purpose to better characterize the fine aerosol fraction (e.g., Cappa et al., 2016), in particular in mixtures where the fine- and coarse-mode aerosol are dominated by different aerosol types (e.g., in mineral dust and black carbon mixtures).

If the measurements are performed without a sub-micrometer cut-off or if the aerosol scattering is dominated by coarse-mode aerosols,

- the size distribution is not measured over a relevant size range, or the aerosol type is not known, the angular correction can be calculated with the C_{SAE} method. However, the uncertainty is up to 33 % in the case of large ($> 1 \mu\text{m}$) absorbing particles and 13 % for slightly absorbing particles.
- the aerosol type is known, the particles are spherical, and the size distribution is measured over a relevant size range, the best method is the $C_{\text{phase,OPS}}$ with the approach (c), considering the true refractive index via an inversion procedure to retrieve the measured size distribution. The uncertainty is 3 % in the case of slightly absorbing aerosols, while it is 25 % if the aerosols are highly absorbing.
- the aerosol type is known and the particles are mineral dust, the angular correction C_{SAE} can be used with an uncertainty of 13 %. To further reduce the uncertainty, the correction $C_{\text{phase,OPS}}$ could be calculated from an optically measured size distribution, but it would need to consider not only the refractive index but also the irregular shape.
- the aerosol type is known to be non-spherical but it is not mineral dust, more investigation is needed on the angular correction. However, C_{SAE} can still be used but with an unknown uncertainty.

Last but not least, nephelometer data should always be reported in both forms with and without angular correction. Indeed, the uncorrected nephelometer particle scattering coefficients might be used within a closure experiment to constrain the particle refractive index without introducing additional uncertainty in the nephelometer scattering coefficient by the angular correction method.

By improving the understanding of the nephelometer angular correction for irregularly shaped coarse particles – in particular mineral dust – this study reduces uncertainties in observations of aerosol scattering properties. Future studies with the aim of further reducing the uncertainty of scattering measurements should focus on environments with a high load of absorbing particles like urban sites or moderately absorbing mixtures such as mixtures of mineral dust and black carbon.

Appendix A: List of symbols

Symbol	parameter
σ_s	scattering coefficient
σ_{sp}	particle scattering coefficient
σ_{sR}	Rayleigh scattering coefficient
σ_{ep}	particle extinction coefficient
σ_{ap}	particle absorption coefficient
θ	scattering angle
γ	angular scattering function
Ω	solid angle with respect to the direction of the incident beam
λ	wavelength
λ_B	blue wavelength of the Aurora 4000 polar nephelometer = 450 nm
λ_G	green wavelength of the Aurora 4000 polar nephelometer = 525 nm
λ_R	red wavelength of the Aurora 4000 polar nephelometer = 635 nm
α	shutter position in degree
ρ	particle density
d	particle diameter
x	size parameter
n	real part of the refractive index
k	imaginary part of the refractive index
SAE	scattering Ångström exponent
SAE*	scattering Ångström exponent calculated using the uncorrected Aurora 4000 measurement of σ_{sp}
Z^{ideal}	angular sensitivity function for an ideal nephelometer
$Z^{Aurora\ 4000}$	angular sensitivity function of the Aurora 4000 polar nephelometer
$\theta_1, \theta_2, \beta_1, \beta_2, \delta_1, \delta_2$	parameters of the angular sensitivity function
S	signal measured by the Aurora 4000 polar nephelometer
$\sigma_{sp}^{Aurora\ 4000, \alpha}$	particle scattering coefficient measured by the Aurora 4000 polar nephelometer for shutter position α
OPS C_{sca}	scattering cross section of the optical particle spectrometer
$C_{R, \alpha}$	Rayleigh angular correction for shutter position α
C_α	angular correction for shutter position α
C_{0°	angular correction for total particle scattering coefficient ($\alpha = 0^\circ$)
C_{phase}	angular correction calculated using the particle phase function
$C_{phase, OPS}$	angular correction calculated using the Mie simulated particle phase function with the OPS particle size distribution
$C_{phase, H}$	angular correction calculated using the particle phase function measured by Horvath's polar nephelometer
C_{SAE}	angular correction calculated using the SAE
C_{polar}	angular correction calculated using the polar measurements of the particle scattering coefficient
$C_{SAE, OPS}$	angular correction calculated using the Mie simulated SAE with the OPS particle size distribution
$C_{polar, OPS}$	angular correction calculated using the Mie simulated polar measurements of σ_{sp} with the OPS particle size distribution

Data availability. Data of the laboratory study shown in this paper are available on request to bernadett.weinzierl@univie.ac.at and marilena.teri@univie.ac.at.

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Author contributions. MT, TM, and BW designed the study. MT, SV, and BW prepared the experiment. MT, SV, TM, and HH tested the experimental set-up and performed the measurements. MT analyzed the data with support from BW, TM, and JG. HH analyzed and provided Horvath's polar nephelometer data. MT performed the simulated closure experiment with contributions from AW for the Mie code reproducing the TSI OPS signal and JG for the ADDA

calculations. The interpretation of the results is the outcome of numerous discussions among MT, TM, JG, SV, HH, RV, PB, AW, and BW. MT wrote the paper with contributions from JG, PB, and BW. All co-authors read and commented on the paper.

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Supplement of

Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections

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S1 Data evaluation

In the following sections, details on the data evaluation of the laboratory experiment are reported. An overview of the data evaluation is given in Sec.4.2 and in Fig.2. of the main text.

S1.1 Particle scattering coefficient

- 5 The Aurora 4000 polar nephelometer measured the particle scattering coefficient $\sigma_{sp,\lambda}^{Aurora4000,\alpha}$ for 18 positions of the shutter: for $\alpha = 0^\circ$ and for α in the range $10 - 90^\circ$ with 5° resolution at three different wavelengths: blue ($\lambda_B = 450$ nm), green ($\lambda_G = 525$ nm), and red ($\lambda_R = 635$ nm). This setting led to a temporal resolution of $26 - 36$ s. The calibration was performed with particle-free air and CO_2 . In addition, several calibration checks were performed before, during, and after the experiment period, as recommended by Anderson and Ogren (1998). All data from the Aurora 4000 polar nephelometer were corrected
- 10 using the average of all calibration checks performed (e.g., for $\alpha = 0^\circ$ and $\lambda_G = 525$ nm, slope = 1.003 ± 0.008 , offset = $-0.4 \pm 0.2 \text{ Mm}^{-1}$) and converted to standard temperature and pressure (STP $T_0 = 273.15$ K $P_0 = 1013.25$ hPa). The error (90 % of confidence) of the total scattering coefficient due to calibration is within ± 3 %, in agreement with the uncertainty of 2.5 % given in the Aurora 4000 user manual. The Rayleigh scattering coefficients at STP were subtracted, and the Aurora 4000 particle scattering coefficients ($\sigma_{sp,\lambda}^{Aurora4000,\alpha}$), not corrected for angular truncation and illumination error, were obtained. The
- 15 combined uncertainty due to calibration and noise for 1 minute data was within ± 8 %, while for the average data along the selected sequences it was within ± 3 %.

S1.2 Particle phase function

- The Horvath's polar nephelometer, a self-built polar nephelometer by Horvath et al. (2018), measured the angular distribution of the light scattered by particles without angular integration, i.e., the particle phase function ($P(\theta)$ at $\lambda = 532$ nm). Its detector
- 20 was mounted on a rotating arm with an angular resolution of 5° , which was moved between 5 and 175° detecting the light scattered into its direction. A complete measurement cycle took 35 min. Calibrations were performed with CO_2 and particle-free air very frequently, every measurement day twice a day, to check the stability of the light source. The Rayleigh scattering coefficient values of the calibration gases are considered at standard temperature and pressure (STP $T_0 = 273.15$ K $P_0 = 1013.25$ hPa). The phase function was obtained by dividing the measured signal at every angle by the integral of all angles.
- 25 The Rayleigh scattering coefficient at STP was subtracted so that the phase function is for particles only. The extrapolation procedure described in (Horvath, 2015) was used to extend the particle phase function to $\theta = 0^\circ$ and $\theta = 180^\circ$. The accuracy of the full integration of the particle phase function given by this procedure is 1 %. Since one measurement cycle took 35 min, the shape of the measured particle phase function may be affected by changes in the aerosol concentration. During dust measurements, the aerosol concentration was very variable. Thus, the measured particle phase function was corrected using
- 30 the 1-minute average signal from the Aurora 4000 polar nephelometer. The Horvath's polar nephelometer signal subtracted by the Rayleigh scattering is divided by the Aurora 4000 particle scattering coefficient at 0° at $\lambda_G = 525$ nm, $\sigma_{sp,\lambda_G}^{Aurora4000,0^\circ}$. Since $\sigma_{sp,\lambda_G}^{Aurora4000,0^\circ}$ is proportional to the light scattered by particles, the value obtained at each angle is proportional to the particle angular scattering function $\gamma_p(\theta)$. The particle phase function $P_p(\theta)$ is obtained by normalization.
- For the data collected during this experiment, there are few cases where the extrapolation procedure performed to extend the
- 35 particle phase function to $\theta = 0^\circ$ leads to a decreasing particle phase function from $\theta = 10^\circ$ to $\theta = 0^\circ$. These results were considered unrealistic and these measurements were excluded in further data analysis.

S1.3 Particle extinction coefficient

- The particle extinction coefficient σ_{ep} at $\lambda = 530$ nm was measured by a Cavity Attenuated Phase Shift Spectroscopy extinction Monitor (CAPS PM_{ext} Monitor, Aerodyne). The CAPS PM_{ext} performed automatically a zero measurement every 5
- 40 minutes. The given particle extinction coefficient σ_{ep} was converted to standard temperature and pressure (STP $T_0 = 273.15$ K $P_0 = 1013.25$ hPa). The instrument was calibrated by comparing the particle extinction coefficient σ_{ep} measured for 203 nm PSL particles at low particle concentration ($< 100 \text{ Mm}^{-1}$) with the particle scattering coefficient σ_{sp} , corrected for angular truncation and illumination error using the angular correction C_{phase} . For this purpose, the particle phase function was

simulated considering a log-normal size distribution with geometric median diameter $d_g = 203$ nm and geometric standard deviation $\sigma_g = 1.026$. The refractive index was $1.582 + i0.000$ for $\lambda = 525$ nm (Devon and Rudin, 1987). Between 15 % and 30 % of the particles were bi-spheres as indicated by the size spectra from the Ultra-High Sensitivity Aerosol Spectrometer (UHSAS, DMT). Scattering by the bi-sphere was simulated using a bisphere code (Mishchenko and Mackowski, 1994). The miepython module developed by Scott Prahl and based on Wiscombe (1980) was used for the single spheres. The angular correction obtained was $C_{phase} = 1.011 \pm 0.002$. During the experiment, the CAPS PM_{ext} was accidentally evacuated. The evacuation may cause contamination of the mirrors, consequently an increase of the loss of signal, and thus a decrease of sensitivity. However, the loss of signal registered after the event is lower than the maximum value of 1000 Mm^{-1} recommended by the manufacturer. Therefore, measurement data after the event can still be used. All polydisperse aerosol measurements were performed before the event. Only a few PSL and ammonium sulfate monodisperse aerosol measurements were performed after the event. Two different calibrations are used: before the event slope = 1.02 ± 0.01 , after the event slope = 1.22 ± 0.02 . The calibration correction included a second degree term ($= 8 \cdot 10^{-5} \text{ Mm}$) to correct for non linear behavior of the CAPS PM_{ext} at high particle extinction coefficient ($> 200 \text{ Mm}^{-1}$). This term was obtained during a previous intercomparison experiment at the TROPOS institute by comparing the particle extinction coefficient measured with the particle scattering coefficient for 200 nm PSL particles in the range $0 - 2000 \text{ Mm}^{-1}$. The accuracy of the particle extinction coefficient measured by the CAPS PM_{ext} was ± 3 % according to Petzold et al. (2013) and Onasch et al. (2015).

50 S1.4 Particle absorption coefficient

The particle absorption coefficient σ_{ap} at wavelengths 467 nm, 528 nm, 652 nm was measured by a tri-color absorption photometer (TAP, Brechtel). We used 47 mm diameter glass-fiber Pallflex E70-2075W filters that are well characterized and have been used widely also with other absorption photometers such as the Particle Soot Absorption Photometer (PSAP, Radiance Research) (Bond et al., 1999; Müller et al., 2014; Virkkula, 2010; Virkkula et al., 2005; Ogren et al., 2017; Davies et al., 2019). The white filter check was performed following the manufacturer indications. The diameter of each of the 10 spots was measured with a caliber for three times. Average value of all measurements is $d = 7.5 \pm 0.2$ mm, with coefficient of variation 2 %, yielding to a spot area of $43.9 \pm 2.3 \text{ mm}^2$. The TAP sample flow was controlled downstream of the measurement cell by a mass flow meter (MFM, Alicat scientific) and measured by an internal flow meter. The sample flow was calibrated with a bubble flow meter (Gilibrator 2, Gilian) at different pressures. The obtained linear least square regression parameters (slope = 0.961 ± 0.004 , offset = $-0.06 \pm 0.01 \text{ Std L min}^{-1}$, $R^2 = 0.99997$) are used to correct the measured sample flow. The calculated actual volume flow rate was $1.99 \pm 0.02 \text{ L min}^{-1}$. Data were converted to standard temperature and pressure (STP $T_0 = 273.15 \text{ K}$ $P_0 = 1013.25 \text{ hPa}$). The 1-minute average absorption coefficients are corrected using the correction scheme developed by Bond et al. (1999); Ogren (2010), using the 1-minute average Aurora 4000 scattering coefficient non-corrected for angular truncation and illumination error. The filter transmission during all measurements was larger than 0.90, higher than the lower limit ($= 0.7$) of the Bond et al. (1999) correction scheme. The single scattering albedo SSA, calculated on the basis of particle extinction coefficient measured by the CAPS PM_{ext} at $\lambda = 530$ nm and absorption measured by the TAP at $\lambda = 528$ nm, for mineral dust measurements is in the range $0.95 - 0.99$, in agreement with literature values (Ansmann et al., 2011; Schladitz et al., 2009; Müller et al., 2011b; Petzold et al., 2011). Ogren et al. (2017) estimated the uncertainty associated with the Bond et al. (1999) algorithm as function of the SSA and the raw absorption coefficient for 30 min averaging absorption coefficient. The raw absorption coefficient $> 3 \text{ Mm}^{-1}$ and the single scattering albedo $SSA = 0.95 - 0.99$, during mineral dust measurements, lead to an uncertainty of the absorption coefficient increasing with SSA from 35 % to 100 %. Since we used weakly absorbing aerosol only, the high relative uncertainty of the particle absorption coefficient has only a very limited effect on the uncertainty of the difference between the particle extinction coefficient and the particle absorption coefficient, i.e., the value considered as a reference in this work.

85 S1.5 Particle size distribution

The particle size distribution was measured with an Ultra-High Sensitivity Aerosol Spectrometer (UHSAS, DMT) in the range 60 – 1000 nm and by an Optical Particle Sizer (OPS 3330, TSI) in the range $0.3 - 10 \mu\text{m}$. Moreover, the total particle number concentration in the size range from 10 nm to $> 3 \mu\text{m}$ was measured by a condensation particle counter (CPC 3772, TSI).

For each instrument, the particle number concentration was calculated from the 1-second count rate and the aerosol flow. The flows were calibrated using a bubble flow meter (Gilian 2, Gilibrator), leading to a volume flow of $53.3 \pm 0.2 \text{ cm}^3 \text{ min}^{-1}$ for the UHSAS, $1.00 \pm 0.05 \text{ L min}^{-1}$ for the OPS, $1.06 \pm 0.01 \text{ L min}^{-1}$ for the CPC. Concentrations were corrected to standard temperature and pressure (STP $T_0 = 273.15 \text{ K}$ $P_0 = 1013.25 \text{ hPa}$) and 1 min average data were produced to reduce noise.

The OPS and UHSAS counting efficiency was estimated comparing their total particle number concentration with the CPC particle number concentration for measurements of monodisperse ammonium sulfate aerosol, where the size was selected with the DMA. Nominal sizes chosen were 402, 501, 602, 702, 850 nm for the comparison with the OPS and 200, 301, 402, 501, 602, 702 nm for the one with the UHSAS. The maximum particle number concentration measured by the CPC was $250 \pm 25 \text{ cm}^{-3}$ for the comparison with the OPS and $1330 \pm 130 \text{ cm}^{-3}$ for the comparison with the UHSAS, well below the single-particle counting limit of 10^4 cm^{-3} reported by the CPC user manual. The error reported for the CPC particle number concentration is the accuracy of $\pm 10 \%$ given by the manufacturer. The least-square linear regression forced through the origin led to a slope of 1.059 ± 0.005 ($R^2 = 0.9999$) for the OPS and of 0.901 ± 0.004 ($R^2 = 0.9999$) for the UHSAS. The high R^2 indicates that coincidences were not occurring for UHSAS and OPS in the considered particle number concentration range.

For this comparison, the OPS and UHSAS particle number concentration of each size bin was corrected for particle loss considering the CPC and OPS or UHSAS particle transmission of the corresponding volume-equivalent diameter (for particle loss calculation see Sec.S1.6). The correction factor for the UHSAS to the CPC transmission varies with size in the range 1.02 – 1.09. The total UHSAS efficiency increased by 3 % after the correction was applied. The correction factor for the OPS to the CPC increases monotonically with particle size from 1.00 to 1.07. However, the correction factor is lower than 1.01 up to $3.5 \mu\text{m}$. Considering the particle loss correction, the OPS efficiency decreased by 0.1 %.

For all the measurements performed during the experiment, the OPS total particle number concentration was below the single-particle counting limit of 1000 cm^{-3} reported by the OPS user manual and the dead time correction was unnecessary. The UHSAS total particle number concentration exceeds the single-particle counting limit of 3600 cm^{-3} in four cases only, i.e., for PSL particles with nominal diameter 0.097, 0.9, 1.6, 1.8 μm . In these cases the UHSAS measures a large amount of very small residual particles that are formed from the PSL liquid when it is evaporated. Since those small particles have a very small contribution to the particle scattering coefficient, no correction was applied.

The calibration of UHSAS and OPS was performed with PSL particles before the experiment. The accuracy of the calibration was checked using the PSL particle measurements performed during the experiment. A log-normal fit was performed for each particle size distribution and the geometric mean diameter obtained was compared with the nominal diameter. Least-square linear regression led to a slope of 0.96 ± 0.01 and an offset of $18 \pm 10 \text{ nm}$ ($R^2 = 0.999$) for the OPS calibration, in agreement with the particle sizing uncertainty of $\pm 5 \%$ given by the manufacturer; a slope of 1.001 ± 0.004 and an offset of $0.4 \pm 1.4 \text{ nm}$ ($R^2 = 0.9999$) for the UHSAS calibration when using the linear map for the size binning of the UHSAS; a slope of 0.999 ± 0.002 and an offset of $0.9 \pm 1 \text{ nm}$ ($R^2 = 0.9999$) for the UHSAS calibration when using the logarithmic map, within the particle sizing uncertainty of $\pm 2.5 \%$ given by the manufacturer.

Comparison between the particle number concentration in overlapping bins (374 – 721 nm) led to an agreement within 1 – 5 % for PSL particles measurements (nominal diameter = 400, 508, 565, 600, 700 nm), within 2 – 15 % for AS measurements (nominal diameter = 400, 508, 565, 600, 700 nm), within 5 – 20 % for mineral dust measurements. PSL equivalent size distributions were obtained for all aerosol samples by combining the UHSAS particle size distribution in the range 0.06 – 0.374 μm with the OPS particle size distribution in the range 0.374 – 10 μm .

S1.6 Particle loss corrections

The particle loss in the tubes from the chamber to the different instruments was calculated with the Particle Loss Calculator software (PLC, von der Weiden et al. (2009)). Settings for the calculations were ambient temperature $T = 20 \text{ C}$ and ambient pressure $P = 1013 \text{ hPa}$. The aerosol density $\rho_0 = 1 \text{ g cm}^{-3}$ and shape factor $\chi_0 = 1$ were chosen to get the particle transmission as function of the aerodynamic equivalent diameter (d_{ae}). The particle transmission along the tubing system of all instruments was in the same range except for the UHSAS and the OPS. In particular, the maximum and minimum transmission (T_{max} and T_{min}) among the transmissions of all optical properties instruments (Aurora 4000, Horvath's polar nephelometer, CAPS PM_{ext} and TAP) differ with their midpoint transmission ($(T_{min} + T_{max})/2$) by less than ± 0.01 for $d_{ae} = 0.06 - 1.00 \mu\text{m}$ and less than ± 0.07 for $d_{ae} = 1.00 - 10.00 \mu\text{m}$. Considering the midpoint transmission and the transmission of the OPS and

the UHSAS, the particle loss correction factor was calculated as function of the particle diameter d_i as $(T_i^{mid}/T_i^{UHSAS}, T_i^{mid}/T_i^{OPS})$. The uncertainty of each correction factor is estimated considering the maximum and minimum transmission of optical properties instruments.

The aerodynamic equivalent diameter was converted to the volume-equivalent diameter d_{ve} of each aerosol type considering their density ρ_p and shape factor χ (PSL $\rho_p = 1.05 \text{ gcm}^{-3}$, $\chi = 1$; AS $\rho_p = 1.77 \text{ gcm}^{-3}$, $\chi = 1$; mineral dust $\rho_p = 2.6 \text{ gcm}^{-3}$, $\chi = 1.25$):

$$d_{ve} = d_{ae} \left(\frac{\rho_p}{\rho_0 \chi} \right)^{-1/2} \quad (1)$$

Moreover, an additional correction is considered for the large sizes of mineral dust to take into account the effect of the non spherical shape on the sizing by the OPS. The optical form factor, the ratio between the extinction coefficient of a particle and the extinction coefficient of a volume-equivalent sphere is proportional to χ^2 for size parameter larger than 10 ($d > 2.1 \mu\text{m}$ for $\lambda_{OPS} = 660 \text{ nm}$) (Pfeifer, 2014). We assume that the scattering cross section seen by the OPS is proportional to the particle extinction coefficient. Since for large size the particle extinction coefficient is proportional to the geometric cross section, the OPS diameter can be written:

$$d_{OPS} = d_{ve} \chi \quad (2)$$

Considering all aerosol types, the correction factor for the UHSAS varies within its detection size range ($0.06 - 1 \mu\text{m}$) from 1.021 ± 0.002 to 1.119 ± 0.006 . The correction factor for OPS in the size range $0.3 - 1 \mu\text{m}$ is negligible $1.000 \pm 0.001 - 0.998 \pm 0.005$; for the size range $1 - 3 \mu\text{m}$ it is within $0.999 \pm 0.003 - 0.98 \pm 0.04$, still a small variation but with a larger error; for larger size (until $10 \mu\text{m}$) the correction factor decreases until to 0.6 ± 0.2 for PSL particles, 0.4 ± 0.3 for AS and 0.5 ± 0.2 for mineral dust.

The particle number concentration of each size bin of OPS and UHSAS was corrected using the correction factor corresponding to the mid diameter of each bin was used. Simulations considering the maximum and minimum transmission yields to total particle scattering coefficient within $\pm 2.4 \%$ of the one obtained considering the midpoint transmission.

S1.7 Sequence selection and temporal averaging

All data were recorded with 1-second resolution, except for the UHSAS which was set to accumulate counts for 10 s, the Aurora 4000 polar nephelometer which has $\sim 30 \text{ s}$ resolution for the measurement of $\sigma_{sp,\lambda}^{Aurora4000,0^\circ}$, and the Horvath's polar nephelometer which takes 35 minutes to complete a cycle backward and forward from scattering angle 5° to 175° . 1-minute averaging was performed for all data in order to reduce the noise, except for the Horvath's polar nephelometer. The time series for all data was visually compared with the time series of the CPC particle number concentration to check the time synchronization. Moreover, the 1-minute average data were used for corrections where data from more instruments were required. For each measurement, sequences with constant particle number concentration and no change in the scattering Ångström exponent $SAE(\lambda_G, \lambda_R)$ were selected and all data were averaged along with the sequences. If a measurement with the Horvath's polar nephelometer was available, the data were averaged along the corresponding 35 minutes. Even if the particle number concentration is not constant, the result is not affected as long as the average is performed for the same time period for all instrumentation. No significant variation of the scattering Ångström exponent $SAE(\lambda_G, \lambda_R)$ were observed during the selected sequences.

S1.8 Optical simulations using measured particle size distribution

The measured particle size distributions were used to simulate via scattering theory the optical properties of the measured aerosols. In particular, the particle scattering coefficient, the particle extinction coefficients, and the particle phase function were simulated for each selected sequence considering the PSL equivalent size distribution obtained merging the UHSAS($0.060 - 0.374 \mu\text{m}$) and OPS($0.374 - 10 \mu\text{m}$). The optical simulations were performed with the program MOPSMAP (Gasteiger and Wiegner, 2018), using the option to give the binned size data directly as input. We considered spherical shapes only (Mie

code) and as refractive index we used the refractive index of PSL particles. $m = 1.591 + i0.00$ was used for $\lambda = 450$ nm, $m = 1.582 + i0.00$ for $\lambda = 525$ nm, $m = 1.574 + i0.000$ for $\lambda = 635$ nm (Devon and Rudin, 1987). This choice has been made to be consistent with the particle size distribution used as input where a PSL particle refractive index was used for the inversion. The simulated particle phase function was integrated considering the Aurora 4000 angular sensitivity function and the simulated Aurora 4000 scattering coefficients non corrected for angular truncation and illumination error were calculated. The uncertainty of the simulated values was estimated by repeating the simulations 1000 times with input binned size data randomly modified within their uncertainty. For modification of the particle number concentration a normal distribution with mean = 1 and standard deviation = 0.05 was assumed. This choice has been made considering the largest uncertainty over the particle number concentration of ± 10 % given by the CPC accuracy and introduced using the CPC to estimate the UHSAS and OPS efficiency. We assumed that the CPC manufacturer accuracy represents a 95 % confidence interval. For modification of the bin diameter a normal distribution with mean = 1 and standard deviation = 0.025 was assumed. This choice is consistent with the ± 2.5 % sizing uncertainty of UHSAS and ± 5 % sizing uncertainty of OPS reported by the manufacturers and confirmed by the calibration check with PSL particles. The 95 % confidence interval over simulated values was estimated considering the 2.5th and 97.5th percentiles.

S2 Measured particle extinction coefficient vs simulated particle extinction coefficient

To test the consistency between the particle size distribution measurements and the optical measurements, we compared the simulated particle extinction coefficient with the particle extinction coefficient measured by the CAPS PM_{ext} . Fig.S1 shows the comparison for all selected sequences. The linear regression forced through the origin weighted considering the inverse of the variance of the simulation results leads to an excellent agreement for PSL particles and ammonium sulfate (AS) measurements with slope = 0.94 ± 0.02 and coefficient of determination $R^2 = 0.987$, slightly lower than the 3 % accuracy of CAPS PM_{ext} .

For polydisperse measurement of mineral dust, silica and volcanic ash the weighted least squares forced through the origin leads to slope = 1.05 ± 0.03 and $R^2 = 0.991$ for cases where the measured volume median diameter (VMD) was smaller than $2 \mu\text{m}$ and to slope = 1.20 ± 0.02 and $R^2 = 0.995$ for cases with $VMD > 2 \mu\text{m}$.

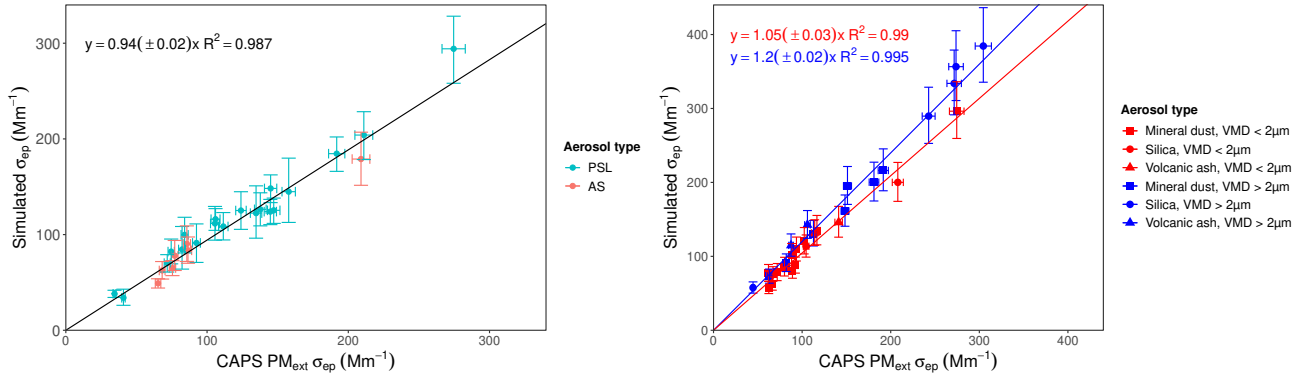


Figure S1. Comparison of the simulated particle extinction coefficient with the particle extinction coefficient measured by CAPS PM_{ext} . The left panel shows the result for PSL particles measurements (light blue) and ammonium sulfate (AS) measurements (orange). The right panel shows the result for mineral dust, silica and volcanic samples for volume median diameter smaller than $2 \mu\text{m}$ (red) and volume median diameter larger than $2 \mu\text{m}$ (blue). Reported error bars are the instrumental accuracy of 3 % for CAPS PM_{ext} and 2.5th and 97.5th percentiles for the simulations.

In order to explain these discrepancies for the case with large dust we performed model calculations with the discrete dipole approximation code ADDA (Yurkin and Hoekstra, 2011) for dust particles considering three different irregular shapes (C, D,

and F from Gasteiger et al. (2011)), different refractive indices ($1.50 + i0.00$ and $1.55 + i0.00$), a volume-equivalent diameter range upto about $5 \mu\text{m}$ and 18000 particle orientations.

Figure S2 shows in the left panel a flow chart of the calculations. The simulated OPS cross-sections ($\lambda = 660 \text{ nm}$) for the irregular mineral dust are inverted assuming a smoothed Mie curve for PSL spheres with refractive index $1.572 + i0.000$. The inverted diameters are combined with PSL particles refractive index $1.582 + i0.000$ to simulate the particle extinction coefficient at the wavelength of the CAPS PM_{ext} ($\lambda = 530 \text{ nm}$) which are then compared to the particle extinction coefficient of the irregularly shaped particle at this wavelength. The right panel of Fig.S2 shows the ratio between the particle extinction coefficient derived via the PSL-based OPS size inversion and the directly simulated particle extinction coefficient as function of diameter. According to the simulations, it can be expected that the derived particle extinction coefficient by coarse particles is overestimated by 10 – 40 %. This overestimation can explain the slope= 1.2 found for $\text{VMD} > 2 \mu\text{m}$ from our measurements. On the other hand, the overestimation for coarse particles might be compensated by an underestimation for fine particles, if they contribute enough to the mineral dust aerosol as in case of $\text{VMD} < 1 \mu\text{m}$.

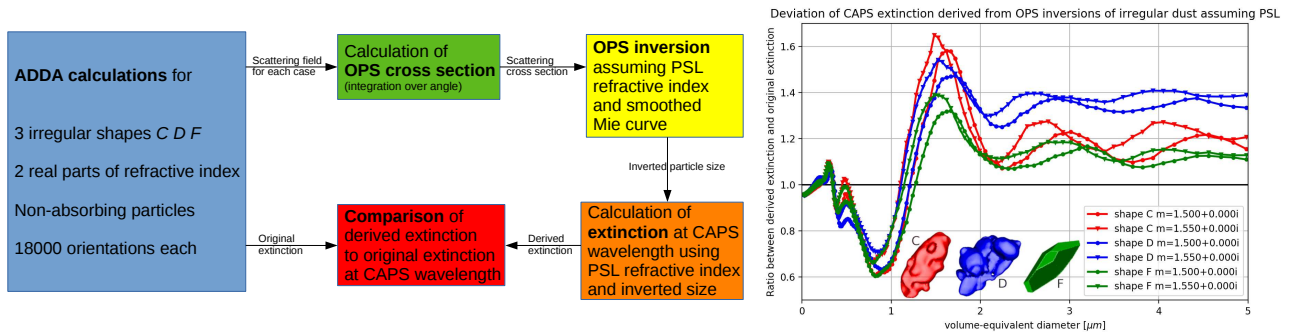


Figure S2. Simulations of the particle extinction coefficient discrepancies for irregularly shaped dust particles when PSL spheres are assumed for OPS size inversion. The left panel shows a flow chart of the simulations. The right panel illustrates the simulated ratio between the derived particle extinction coefficient and the original particle extinction coefficient of the irregularly shaped particle as function of the particle size at $\lambda = 530 \text{ nm}$ the CAPS PM_{ext} wavelength. The colors illustrate results for three different irregular shapes with non-absorbing refractive indices $1.50 + i0.00$ and $1.55 + i0.00$.

S3 Sensitivity of the TSI OPS 3330 to particle refractive index and shape

215 An optical particle sizer, such as the TSI OPS 3330, measures the particle scattering cross sections and sorts particles into
different size bins based on their detected scattering cross section. For the TSI OPS 3330, the manufacturer provides nominal
diameter values for the bin boundaries for the calibration material, i.e., PSL particles with refractive index $m = 1.59 + i0.00$
and spherical shape. The particle size distribution obtained with such a measurement is a PSL-equivalent size distribution. Any
220 deviation from the refractive index or shape of the calibration material will increase the uncertainty of the derived particle size
distribution.

Figure S3 shows the relationship between the TSI OPS 3330 scattering cross section and the particle volume-equivalent
diameter for the calibration material (in black) and for different refractive indices in the range of mineral dust particles, both in
the case of spherical particles and in the case of irregularly shaped particles.

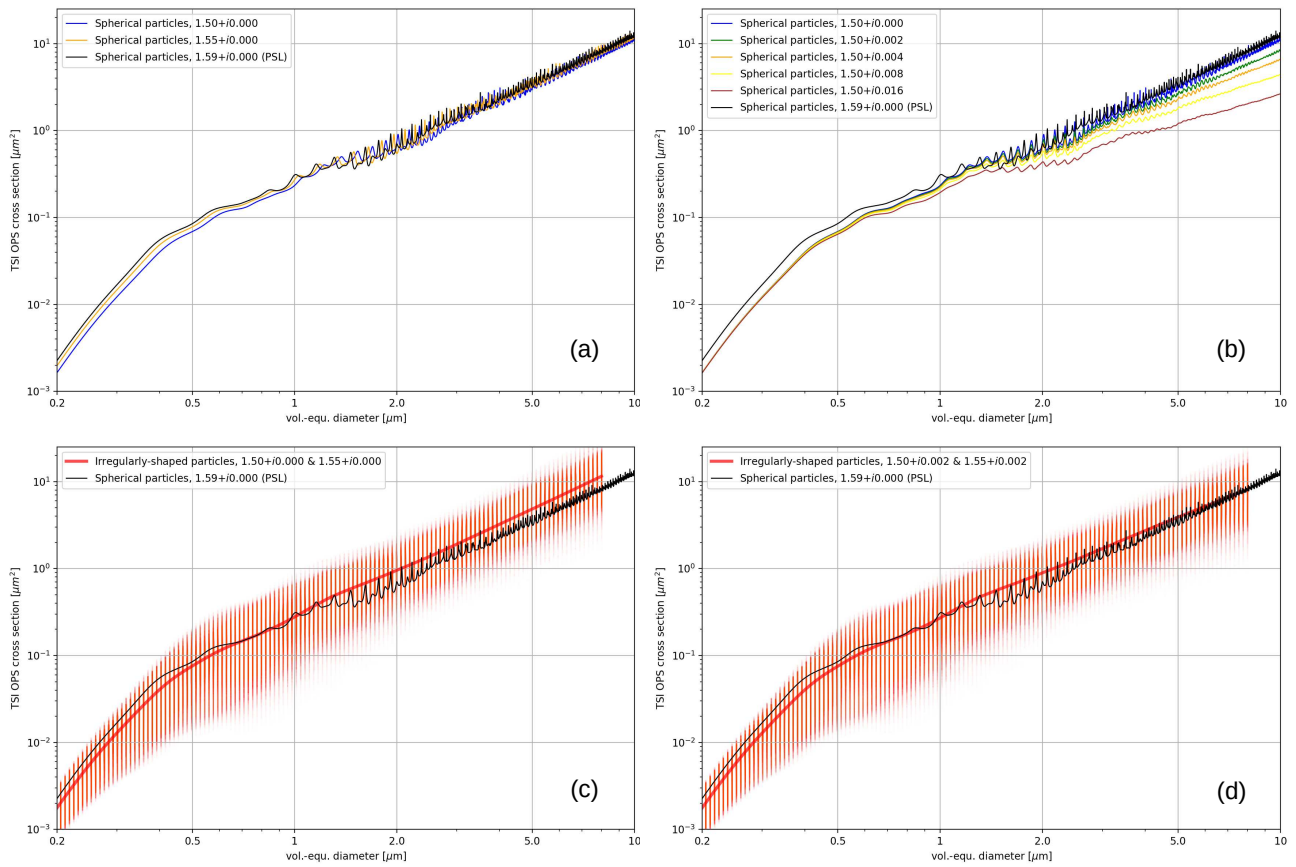


Figure S3. Scattering cross section modeled for the TSI OPS 3330. The black line in all four panels is the TSI OPS 3330 scattering cross section for PSL particles, i.e., the calibration material ($m = 1.59 + i0.00$ and spherical shape). In addition, each panel shows the TSI OPS 3330 scattering cross section for particles with different refractive indices or shapes relevant for mineral dust particles: (a) non-absorbing spherical particles with different real part of the refractive index ($n = 1.50, 1.55$); (b) spherical particles with different imaginary part of the refractive index ($k = 0.000, 0.002, 0.004, 0.008, 0.016$); (c) non-absorbing and (d) slightly absorbing irregularly shaped particles. Red dots correspond to 18000 orientations of six different mineral dust shapes (Gasteiger et al., 2011) and the red line is the average.

The TSI OPS 3330 scattering cross section of coarse-mode particles ($d > 1 \mu\text{m}$) is not very sensitive to the real part of the refractive index between $m = 1.50 + i0.00$ and $m = 1.59 + i0.00$ (Fig.S3a). Thus, the uncertainty of the PSL equivalent particle size distribution for non-absorbing spherical particles with mineral dust-like refractive index is negligible. The TSI OPS 3330 scattering cross section for coarse-mode particles is more sensitive to the imaginary part, and the PSL-equivalent diameters underestimate the diameter of absorbing particles (Fig.S3b). For example, a moderately absorbing spherical particle with imaginary part $k = 0.004$ and a diameter of about $5 \mu\text{m}$ would have the same scattering cross section as a PSL particle with about $4 \mu\text{m}$ diameter, and therefore, the OPS would size it with a PSL-equivalent diameter of about $4 \mu\text{m}$.

If particles have an irregular shape, the relationship between TSI OPS 3330 scattering cross section and particle volume-equivalent diameter is more complex as also the shape and the orientation of the particle plays a role. Figure S3c and d show the TSI OPS 3330 scattering cross section for 6 irregular shapes typical for mineral dust (Gasteiger et al., 2011) in 18000 orientations (red dots). The average is indicated by the red line. Coarse non-absorbing irregular particles have, on average, a slightly increased scattering cross section with respect to spherical particles with the same volume-equivalent diameter (Fig.S3c). Therefore, the particle size is slightly overestimated by a TSI OPS 3330 calibrated with PSL particles. In case of slightly absorbing particles (e.g., with imaginary part ~ 0.002) the TSI OPS 3330 calibrated with PSL has on average a better performance (Fig.S3d). Therefore, the effect of irregular shape might partly compensate the effect of the imaginary part of the refractive index.

Furthermore, the variability of the scattering cross section of irregularly shaped particles of fixed size might lead to additional effects on higher orders of the size, e.g., particle area or volume, so that the agreement of the average shown in Fig.S3d, does not necessarily result in good performance for higher orders using the same imaginary part. For any non linear function of the size separate investigations are needed.

S4 Details of the simulated closure experiment

This section describes the details of the simulated closure experiment. Selected particle size distributions, refractive indices, and shapes are used as input to simulate the TSI OPS 3330 response. The angular correction $C_{phase,OPS}$ is obtained from the OPS particle size distributions through optical simulations considering different approaches, e.g., assuming different refractive indices or shapes. The angular correction $C_{phase,OPS}$ can be compared with the "original" C_{0° , obtained simulating the Aurora 4000 polar nephelometer response for the corresponding particle size distributions, refractive indices, and shapes. The overview of the flow chart of the simulated closure experiment is given in Sec.5.1 and in Fig.4 of the main text.

We selected 5000 random samples with input particle size distributions and refractive indices similar to Anderson and Ogren (1998), Müller et al. (2011a) Bond et al. (2009) and Massoli et al. (2009). The particle size distributions were bimodal log-normal with geometric volume median diameter ranging from 0.2 to $0.4 \mu\text{m}$ for the fine mode and from 2.0 to $4.0 \mu\text{m}$ for the coarse-mode. The geometric standard deviations were selected for each mode in the range between 1.6 and 2.2 . The fine mode volume fraction was varied between 0.1 and 0.9 . For each case, either the complete particle size distribution (no size cut-off) or a particle size distribution with a nominal aerodynamic $1 \mu\text{m}$ cut off (sub-micrometer size cut off) was considered. The sub-micrometer size cut off was modeled as a hyperbolic tangent function with a 50% geometric diameter cut point at $0.85 \mu\text{m}$ and 10% and 90% transmission $0.05 \mu\text{m}$ above and below the cut point, similarly to Massoli et al. (2009). For homogeneous spherical particles, the refractive index was chosen among $1.33, 1.38, 1.4, 1.43, 1.48, 1.52, 1.53, 1.57, 1.59, 1.63, 1.70$ for the real part and among $0.00, 0.001, 0.002, 0.005, 0.01, 0.03, 0.05, 0.10, 0.30$ for the imaginary part.

Additionally, 1000 random samples were selected with refractive index similar to mineral dust particles: 1.5 , or 1.55 for the real part and among $0.00, 0.002, 0.004, 0.008, 0.016$ for the imaginary part. The calculations for these samples were repeated twice for homogeneous spherical particles and for irregular shapes selected among mineral dust shapes from Gasteiger et al. (2011).

Optical simulations for homogeneous spherical particles were performed with a Mie code (miepython module - <https://github.com/scottprahl/miepython>, developed by Scott Prahl and based on Wiscombe (1980)), and for irregularly shaped particles with the discrete dipole approximation (ADDA code - <https://github.com/adda-team/adda>, Yurkin and Hoekstra, 2011).

The original C_{0° was simulated for each selected refractive index, particle size distribution and shape considering the Aurora 4000 angular sensitivity function for $\alpha = 0^\circ$. The selected refractive indices are referred hereafter as the true refractive index, $n_{true} + ik_{true}$.

The original angular correction C_{0° is referred hereafter as the spherical C_{0° or irregular C_{0° for homogeneous spherical particles or for irregularly shaped particles respectively. For the calculation of the irregular C_{0° , we considered the average over the mineral dust-like irregular shapes (A-F) from Gasteiger et al. (2011), and 18000 particle orientations.

Calculations were performed on a diameter grid, selected with 1371 log-spaced diameters with resolution $d\log(d) \sim 0.00256$ in the size range, equivalent to the OPS diameter detection limits. The OPS diameter detection limits corresponding to each $n_{true} + ik_{true}$ were calculated via Mie theory considering the diameter detection limits of $0.3 - 10 \mu\text{m}$ for homogeneous spherical particles with refractive index $1.59 + i0.00$ given by the manufacturer. For the irregularly shaped particle calculations, the diameter grid upper size was restricted to the upper size available for the ADDA calculations: $4.9 \mu\text{m}$ for $\lambda_B = 450 \text{ nm}$, $5.8 \mu\text{m}$ for $\lambda_G = 525 \text{ nm}$, $7.0 \mu\text{m}$ for $\lambda_R = 635 \text{ nm}$.

The angular correction $C_{phase,OPS}$ corresponding to each original C_{0° was simulated with the following five steps (See steps 1-5 of the flow chart in Fig.4 of the main text):

Step 1: the signal measured by the TSI OPS 3330, the scattering cross section $C_{sca,i}$, was calculated for the selected true refractive index for each diameter of the grid d_i . In the case of homogeneous spherical particles, we calculated the OPS $C_{sca,i}$ using a (custom) Mie code based on Bohren and Huffman (2008), in which we considered the light polarization to calculate the intensity of the scattered light as a function of both scattering and azimuth angle. To obtain the OPS $C_{sca,i}$, we integrated the result over the spherical angle covered by the OPS detector. In case of irregular particles, we calculated the OPS $C_{sca,i}$ for each mineral dust-like shapes (A-F) from Gasteiger et al. (2011) using the ADDA code (Yurkin and Hoekstra, 2011). The orientations and shapes were selected randomly for each d_i . For different random samples, the results in terms of angular correction were affected less than 0.1% .

Step 2: the size selection executed by the TSI OPS 3330 was reproduced, i.e., each OPS $C_{sca,i}$ was assigned to a bin_i from 1 to 16. The C_{sca} values for the OPS bin boundaries were calculated from the PSL-equivalent nominal diameter bin boundaries provided by the manufacturer and the OPS Mie curve (i.e., the OPS C_{sca} as a function of diameter) for the refractive index of $1.59 + i0.00$, i.e., the refractive index of PSL particles.

Step 3: the selected size distribution $dn/d\log(d)$ was considered by calculating the counts for each $bin_{j=1,16}$ as

$$Counts_{j=1,16} = \sum_{bin_i=j} \frac{dn_i}{d\log(d_i)} d\log(d_i), \quad (3)$$

The introduction of the selected particle size distribution at this step helps to reduce the computational time.

Step 4: the diameter values of the bin boundaries were selected with different approaches to obtain the binned size data. Either the nominal diameter values provided by the manufacturer for the PSL particle refractive index ($1.59 + i0.00$) are used, or an inversion procedure is applied to obtain the bin boundaries for other refractive indices. In the latter case, we calculated the bin boundaries through a Mie curve inversion, considering a smoothed Mie curve of the true refractive index.

Step 5: the optical calculations to obtain the angular correction $C_{phase,OPS}$ were performed with the program MOPSMAP (Gasteiger and Wiegner, 2018). We used the MOPSMAP option to give the binned size data directly as input. Different approaches were applied to select the input refractive indices for the optical simulations.

Different approaches to run steps 4 and 5 are possible. We considered the following three possibilities:

Approach a: In step 4, the PSL-equivalent nominal diameter values for the OPS bin boundaries provided by the manufacturer were used to obtain the binned size data. In step 5, the angular correction $C_{phase,OPS}(a)$ for the randomly selected aerosol was calculated with Mie calculations. Input for the optical calculations were the binned size data obtained in step 4, the refractive index of PSL particles ($1.59 + i0.00$) and the green wavelength of the Aurora 4000 nephelometer ($\lambda_G = 525 \text{ nm}$).

Approach b: In step 4, the PSL-equivalent nominal diameter values for the OPS bin boundaries provided by the manufacturer were used to obtain the binned size data. In step 5, the angular correction $C_{phase,OPS}(b)$ was calculated for the randomly selected aerosol with Mie calculations assuming the true refractive index, i.e., the refractive index selected for corresponding random selected aerosol.

315 **Approach c:** In step 4, the diameter values of the bin boundaries were calculated by an inversion procedure assuming the true refractive index, i.e., the refractive index selected for each randomly selected aerosol. In step 5, the angular correction $C_{phase,OPS}(c)$ was calculated for the randomly selected aerosol with Mie calculations assuming the true refractive index.

None of the approaches considers the non-sphericity of the irregularly shaped particles in steps 4 or 5.

S5 Relevance of particles in the size range 0.06 – 0.3 μm

320 In order to calculate the angular correction, the particle size distribution in the diameter range 0.06 – 0.3 μm is relevant as well. We compared the original angular correction C_{0° calculated for the size range 0.06 – 10 μm and the TSI OPS 3330 diameter detection limits (Fig.S4). This comparison shows that considering only the OPS size range leads to an overestimation of the angular truncation and illumination error by 1 % up to 50 % for the calculations without size cut off and up to 10 % for the calculations with sub-micrometer size cut off, increasing with the imaginary part of the refractive index.

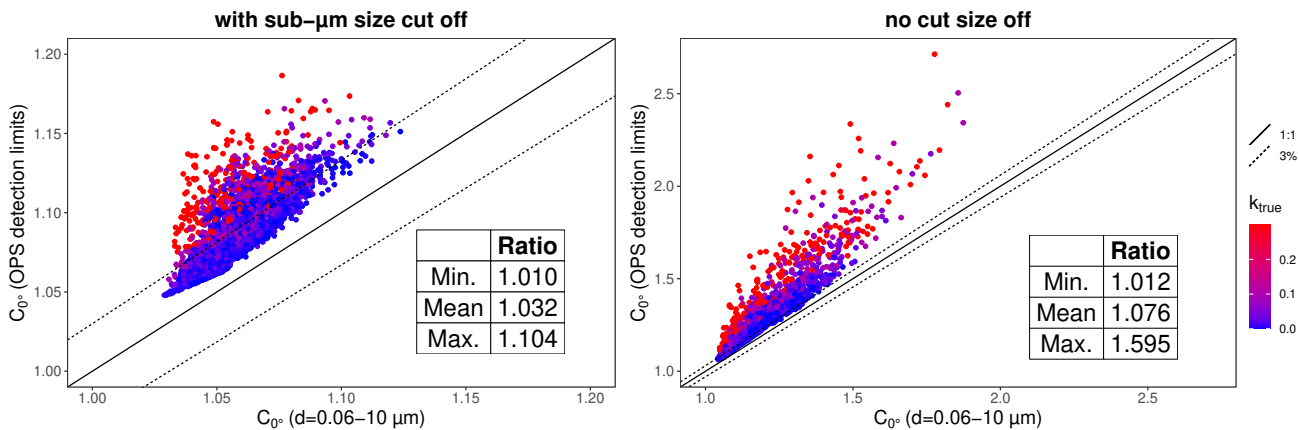


Figure S4. Comparison between the angular correction calculated considering the size range 0.06 – 10 μm and considering the OPS detection limits. Calculations are for the green wavelength ($\lambda = 525 \text{ nm}$). The left panel shows the comparison for size distributions without a cut off. The right panel shows the comparison for size distributions with a sub-micrometer cut off applied. Different colors refers to different values of the imaginary part of the refractive index. The solid line is the 1 : 1 line. The dashed lines are the $\pm 3\%$ lines.

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Corrigendum to **“Impact of particle size, refractive index, and shape on the determination of the particle scattering coefficient – an optical closure study evaluating different nephelometer angular truncation and illumination corrections” published in Atmos. Meas. Tech., 15, 3161–3187, 2022**

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During manuscript preparation, the following two things
were written incorrectly and as a result were reported inac-
curately in the published paper:

1. Table 1 reports the parameter of the Aurora 4000 angu-
lar sensitivity function measured by Müller et al. (2012).
The parameter +0.0082 in Table 1 should be −0.0082
because $\delta_2(\alpha)$ in Eq. (1) in Müller et al. (2012) is writ-
ten as

$$\delta_2(\alpha) = 1.1935 \cdot \alpha - 0.0082 \cdot \alpha^2.$$

We would like to point out that this typo in the paper
does not affect the results of the paper because in the
data analysis code the correct value of $\delta_2(\alpha)$ was used.

2. We stated that our tricolor absorption photometer (TAP,
Model 2901, Brechtel) determines the particle ab-
sorption coefficient for the wavelengths of 467, 528,
and 652 nm as reported in other publications (e.g.,
Davies et al., 2019) as well as on the Brechtel website
(https://www.brechtel.com, last access: 17 July 2023).

However, we recently discovered that the unit of the
University of Vienna has slightly different wavelengths,
which are 465, 520, and 640 nm. As the differences be-
tween the wavelengths are smaller than the full-width
half maximum (FWHM) reported in the user manual
(i.e., 25, 35, and 22 nm), this difference is negligible and
has no impact on the paper results.

The incorrect wavelengths were reported in Fig. 1 and
in Sects. 4.1, 4.2, 4.3, and S1.4.

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Physics, Chinese Academy of Sciences) for spotting the typo in Ta-
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Pollution affects Arabian and Saharan dust optical properties in the Eastern Mediterranean

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Abstract. Uncertainties in the direct radiative effect of mineral dust result from the variability in its optical properties, but are also influenced by mixing with anthropogenic aerosols ("pollution"), e.g., black carbon or sulfates. This study investigates the effect of mixing pollution with mineral dust from different source regions on the intensive optical properties. The Ångström exponents of scattering and absorption, the single scattering albedo, and asymmetry parameter are determined from in-situ measurements during the A-LIFE aircraft field experiment over the Eastern Mediterranean, where Arabian and Saharan dust mixed with pollution. Our results show that all intensive dust optical properties change significantly with increasing pollution content. Interestingly, the differences between the optical properties of Arabian and Saharan dust are negligible. We discuss the implications of these results for the identification of mineral dust events and for their direct radiative effects. The pollution masks the mineral dust signal, which calls for caution when using Ångström exponents to identify mineral dust events. Furthermore, asymmetry parameter and single scattering albedo change from pure to polluted mineral dust layers (e.g., at 525 nm, median values decrease from 0.67 to 0.56 and from 0.96 to 0.89, respectively). These changes have opposing effects on the short-wave direct radiative effect efficiency (i.e., the direct radiative effect per unit of aerosol optical depth), potentially canceling out each other. Still, the effect can be significant depending on surface albedo. In conclusion, quantifying pollution content in mineral dust layers is essential for accurately assessing their local direct radiative effect.

1 Introduction

Large amounts of mineral dust particles are emitted into the atmosphere by winds blowing over arid regions worldwide (Choo-bari et al., 2014). Once in the atmosphere, they can form layers at high altitudes and travel long distances (Weinzierl et al.,

2017; Adebisi et al., 2023). Mineral dust particles interact directly with solar and terrestrial radiation via scattering and absorption, as well as indirectly by altering the cloud properties. Consequently, mineral dust particles affect the Earth's energy budget and its climate system. The direct radiative effect of mineral dust is still uncertain due to significant uncertainties in its microphysical and optical properties (Kok et al., 2017; Li et al., 2021). This uncertainty contributes significantly to the overall uncertainty of the effective radiative effect of mineral dust (Kok et al., 2023).

The properties of mineral dust layers may be modified through aging and mixing processes (Riemer et al., 2019). For example, the mineral dust size distribution can change (e.g., Weinzierl et al., 2011) or the mineral dust layer can mix (e.g., Petzold et al., 2011; Denjean et al., 2016) with anthropogenic aerosol such as black carbon or fine particles (which we will refer to as "pollution" in this study). Especially, black carbon particles, emitted from fossil and biomass fuel combustion, exhibit a higher absorption efficiency and different spectral behavior than mineral dust particles (Bond et al., 2013; Caponi et al., 2017). Hence, black carbon particles within mineral dust layers can play a critical role in enhancing the mineral dust absorption properties, resulting in a relatively greater impact on atmospheric heating compared to low altitudes black carbon particles, since their residence time at higher altitudes is longer (Seinfeld et al., 2004).

The aerosol direct radiative effect depends on the aerosol optical properties, in particular the particle extinction coefficient σ_{ep} (the sum of the particle scattering and absorption coefficients), the asymmetry parameter g (i.e., the cosine-weighted average of the angular distribution of the scattered light, Andrews et al., 2006), and the single scattering albedo SSA (the fraction of the extinct light that is scattered). These aerosol optical properties are generally derived from remote sensing retrievals (e.g., Granados-Muñoz et al., 2019) or from in-situ measurements of the particle size distribution via optical modeling (e.g., Kok et al., 2017). These methods require assumptions about aerosol types, refractive indices, and mixing states.

Direct in-situ measurements of the particle scattering and absorption coefficients in the visible spectrum can be used to calculate intensive aerosol optical properties that allow a better understanding of the optical signature and the direct radiative effect of aerosol mixtures. These intensive aerosol optical properties include the wavelength dependence of the particle scattering and absorption coefficients and of the single scattering albedo, known as the scattering, absorption and single scattering albedo Ångström exponents (SAE, AAE, and SSAE), which provide near-real-time information on the aerosol types that impact the aerosol optical properties (e.g., Costabile et al., 2013; Cazorla et al., 2013; Cappa et al., 2016). Additionally, the asymmetry parameter (g) and the single scattering albedo (SSA) are crucial for determining the intrinsic short-wave direct radiative effect of the aerosol mixture (e.g., Sherman et al., 2015).

These intensive aerosol optical properties are sensitive to different aerosol characteristics. The SAE is primarily related to the particle size (Ångström, 1929), with $SAE < 1$ found for the predominance of coarse aerosol particles and $SAE > 2$ for the predominance of fine particles (e.g., Schuster et al., 2006; Gyawali et al., 2012). The AAE is mainly linked to the particle composition (e.g., Liu et al., 2018), with $AAE \sim 1$ for pure black carbon particles, since it absorbs efficiently through the complete solar spectrum (e.g., Bond, 2001). Higher AAE values correspond to absorbing aerosol particles whose absorption increases with decreasing wavelength, such as mineral dust and organic matter (called brown carbon) aerosols (e.g., Petzold et al., 2009; Sandradewi et al., 2008; Russell et al., 2010; Valentini et al., 2020). The single scattering albedo Ångström exponent (SSAE) is sensitive to both aerosol size and composition. Thus, it has been proposed as an additional parameter indicating the presence

of mineral dust particles (Collaud Coen et al., 2004). The asymmetry parameter g indicates the asymmetry of the angular distribution of scattered light, varying from 1 in the extreme case of light scattered exclusively in the forward direction to 0 for symmetric scattering. For atmospheric aerosol particles, g is typically in the range of about 0.4 to 0.8 and decreases with decreasing particle size (e.g., Horvath et al., 2018; Pandolfi et al., 2018). The SSA equals 1 if only purely scattering aerosols (e.g., sulfate) are present and decreases as more absorbing aerosols are present. Therefore, g considers the effect of particle sizes, and the SSA takes into account the effect of aerosol absorption on the direct radiative effect.

Several intensive field experiments, including airborne and ground-based measurements, were performed to study the properties of mineral dust. The main focus has been on the properties of pure mineral dust layers close to the source region and after transport (e.g., Formenti et al., 2003, 2011a; Ryder et al., 2013; Denjean et al., 2016; Weinzierl et al., 2017; Adebisi et al., 2023). The most comprehensive field experiment on the properties of Saharan dust was the Saharan Mineral Dust experiment (SAMUM). The first part of the field campaign (SAMUM-1, Heintzenberg, 2009) studied the properties of pure mineral dust in Morocco, whereas SAMUM-2 investigated far-transported, aged dust and as well as its mixture with marine, urban and biomass-burning aerosols, pointing out the importance of further studies on mixtures of aerosols (Ansmann et al., 2011).

Meanwhile, several studies investigated the feasibility of using the Ångström exponents (SAE, AAE, and SSAAE) to detect mineral dust events in near-real-time using ground based in-situ measurements (e.g., Collaud Coen et al., 2004; Valenzuela et al., 2015; Ealo et al., 2016). They found out that the presence of mixtures may mask the signal of mineral dust particles and only strong mineral dust events can be detected. Therefore, for detecting mineral dust events at specific places, the intensive aerosol optical properties need to be evaluated and calibrated with previous sensitivity tests (Ealo et al., 2016).

Studies on mineral dust-pollution mixtures are sparse and conducted mainly in Asia (e.g., Clarke et al., 2004; Seinfeld et al., 2004; Yang et al., 2009), but few field campaigns were also conducted in Western Africa (Petzold et al., 2011; Denjean et al., 2020), and in the western Mediterranean (Mallet et al., 2016; Denjean et al., 2016). Large variability in the obtained results from different studies indicate that internal (Clarke et al., 2004; Quinn et al., 2004) or external (Shi et al., 2003; Song et al., 2005) mixtures of black carbon and mineral dust may significantly change the optical properties of mineral dust layers. Additionally, different source regions may be affected differently by the mixing with pollution. Petzold et al. (2011) showed that the pollution plume from the city of Dakar had a significant influence on the particle size distribution and absorption properties of mineral dust particles, whereas the absorption Ångström exponent was still dominated by the mineral dust fraction. In Casablanca, a different scenario was observed with an AAE value of ~ 1 despite the high mineral dust concentrations Petzold et al. (2009). In Asia, Seinfeld et al. (2004) found that carbonaceous components in the mineral dust layers changed the SSA from 0.97 to 0.85 at 550 nm. Similarly, in West Africa, Denjean et al. (2020) measured enhanced aerosol light absorption in mineral dust layers due to mixing with pollution, leading to a decrease in SSA from 0.92 to 0.81. On the contrary, in the western Mediterranean, Denjean et al. (2016) observed moderately absorbing mineral dust layers with SSA values between 0.90 and 1.00 at 530 nm, indicating that the Saharan dust optical properties were not significantly affected by mixing with pollution. In proximity to Arabian and Saharan dust source regions, Kim et al. (2011) investigated aerosol optical properties based on AERONET observation, finding no systematic difference in the optical properties of pure Arabian and Saharan dust.

Nevertheless, their analysis suggests that the optical properties of mineral dust mixtures may be affected differently depending on the source regions.

In conclusion, mixtures of black carbon and mineral dust may significantly change the optical properties of mineral dust layers. These changes can have consequences on identifying mineral dust layers by using the Ångström exponents (SAE, AAE, and SSAAE) (e.g., Collaud Coen et al., 2004; Valenzuela et al., 2015; Ealo et al., 2016) and on the direct radiative effect by influencing g and SSA (e.g., Seinfeld et al., 2004; Denjean et al., 2020). However, optical properties of mineral dust from various source regions may be affected differently by mixing with pollution (Seinfeld et al., 2004; Denjean et al., 2016; Kok et al., 2023) and this influence needs to be further investigated.

In April 2017, the A-LIFE (Absorbing aerosol layers in a changing climate: aging, lifetime and dynamics, <https://www.a-life.at>) aircraft field experiment investigated the aerosol properties in the Eastern Mediterranean (Weinzierl et al., in preparation, 2024). The region is an atmospheric crossroad and a pollution hot spot (Zittis et al., 2022), where aerosols of different origins (natural vs anthropogenic) co-exist, including mineral dust from the two nearby deserts (the Arabian Peninsula and the Saharan desert) and local and transported pollution (e.g., Dayan et al., 2017). We focus on the intensive aerosol optical properties measured airborne during the A-LIFE field experiment.

The main goal of this paper is to analyze how the mixing of mineral dust and pollution impacts the intensive optical properties of mineral dust and whether these impacts vary depending on the source region of mineral dust. Specifically, we aim to investigate how mineral dust-pollution mixtures affect existing aerosol classification schemes and modify the direct radiative effect of the mineral dust. Thus, we investigate intensive aerosol optical properties that are relevant for the identification of aerosol types, i.e., the scattering, absorption, and single scattering albedo Ångström exponents (SAE, AAE, SSAAE). We also study intensive aerosol optical properties that are relevant for the understanding of the aerosol radiative effect, i.e., the asymmetry parameter and the single scattering albedo (g and SSA). In addition, we use a simple formula by Haywood and Shine (1995) to estimate the change in the short-wave direct radiative effect efficiency (SW DREE) at the top of the atmosphere due to increasing pollution content in the mineral dust layer.

110 2 Methodology

To investigate the influence of mixing mineral dust and pollution on the intensive aerosol optical properties, we analyzed data collected in spring 2017 on board the DLR Falcon 20 E-5 aircraft during the field experiment of the ERC-funded A-LIFE project (Absorbing aerosol layers in a changing climate: aging, lifetime and dynamics, <https://www.a-life.at>). The particle scattering and absorption coefficients measured directly in Arabian and Saharan dust layers were used to derive intensive aerosol optical properties, including the scattering, absorption and single scattering albedo Ångström exponents (SAE, AAE, and SSAAE), the single scattering albedo (SSA), the asymmetry parameter (g), and the short-wave direct radiative effect efficiency (SW DREE). Since data with different relative contribution of mineral dust and black carbon are available, we used the microphysical properties to quantify the pollution content within mineral dust layers. Thus, we investigated the relationships between each intensive aerosol optical property and the aerosol microphysical properties. Additionally, we investigated the differences in these relationships for two different source regions of mineral dust (namely, the Arabian Peninsula and the Saharan desert). The following sub-sections describe the A-LIFE aircraft field experiment, the instrumentation relevant for this study and how the data were analyzed.

2.1 The A-LIFE aircraft field experiment in the Eastern Mediterranean

The A-LIFE aircraft field experiment conducted ground-based and airborne measurements with the aim to investigate the properties of absorbing aerosol mixtures, with a focus on mineral dust-black carbon mixtures. The airborne measurements were carried out on board of the Deutsches Zentrum für Luft- und Raumfahrt (DLR) research aircraft Falcon 20-E5, and the aircraft was based at the Paphos airport in south-west of Cyprus (34.7°N, 32.5°E). This location allowed access to the Eastern Mediterranean region and to mineral dust layers originating from the two nearby deserts, the Arabian Peninsula in the Middle East and the Saharan desert in Northern Africa as well as to anthropogenic pollution (Weinzierl et al., in preparation, 2024).

Figure 1 shows the flight paths of the 22 research flights. Apart from the two test flights in Central Europe and the transfer flights from the DLR airport to the Paphos aircraft base in Cyprus, most of the flights were conducted over the Eastern Mediterranean, close to Cyprus, but also in the area of Crete and Malta. The background of Figure 1 is a natural color satellite image from April 27, 2017, showing the influence of the regional aerosol loading by mineral dust from the two nearby deserts. Mineral dust from the Arabian Peninsula impact the eastern parts of the Mediterranean basin, while mineral dust from the Sahara desert influences the central part of the Mediterranean basin.

During the field experiment, several outbreaks of Arabian and Saharan dust occurred and the elevated mineral dust layers were also mixed with local and transported anthropogenic pollution, allowing the investigation of mineral dust-pollution mixtures. A detailed description of the field experiment and the meteorological conditions influencing the measurements can be found in the A-LIFE overview paper (Weinzierl et al., in preparation, 2024).

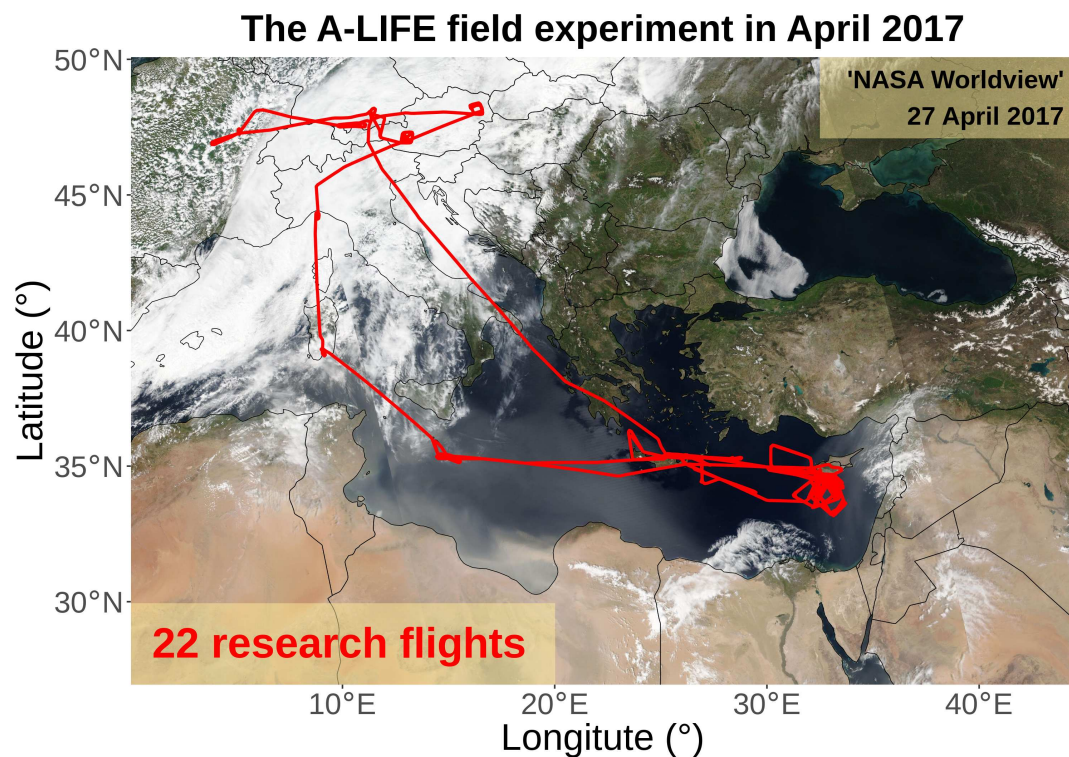


Figure 1. The A-LIFE field experiment in April 2017. The red line indicates the flight path of the 22 research flights performed during the A-LIFE field experiment onboard of the Deutsches Zentrum Luft- und Raumfahrt (DLR) research aircraft Falcon 20 E-5. The background is a natural color satellite image (26.9°N-50.1°N, 0.0°E-44.5°E) from 27 April 2017. (Source: "NASA-Worldview", <https://worldview.earthdata.nasa.gov/>, last accessed 16 January 2024.)

140 2.2 Instrumentation

For the A-LIFE field experiment, the DLR aircraft Falcon 20 E-5 was equipped with comprehensive in-situ aerosol instrumentation, a wind lidar, and impactors for subsequent chemical analysis of the filter samples. Most in-situ aerosol instruments were deployed in the Falcon aircraft cabin behind an aerosol inlet and transport system (in-cabin). The cut-off diameter of the Falcon aerosol sampling system ranges between 2.4 μm to 6.4 μm , as characterized in detail in Schöberl et al. (2024). A list of
145 the in-cabin and wing-mounted instruments is available in the supplemental material of (Schöberl et al., 2024).

Instrumentation relevant for this study includes an in-cabin polar integrating nephelometer (Aurora 4000 polar nephelometer, Ecotech, Australia) and an in-cabin Tricolor Absorption Photometer (TAP, Brechtel Manufacturing Inc., Hayward, USA), which measured the aerosol scattering and absorption coefficients at three wavelengths, respectively. An in-cabin Single Particle Soot Photometer (SP2, Droplet Measurement Technologies Inc. (DMT), Longmont, CO, USA) measured the mass and size
150 of individual refractory black carbon particles and provides the mass concentration of refractory black carbon.

The particle size distribution was determined from a combination of instruments with overlapping size ranges: The total number concentration of particles was measured with an in-cabin condensation particle counter (TSI CPC, model 3760a, TSI Inc.). Size resolved-particle number concentration was measured with an in-cabin optical particle counter (SkyOPC, model 1.129, GRIMM Aerosol Technik, Ainring, Germany), and two wing-mounted optical spectrometers, the Ultra-High Sensitivity
155 Aerosol Spectrometer-Airborne (UHSAS-A, DMT), and the Cloud and Aerosol Spectrometer (UNIVIE CAS), which is part of University of Vienna's second-generation Cloud, Aerosol, and Precipitation Spectrometer (UNIVIE CAPS, DMT).

In-situ aerosol samples were collected in the Falcon aircraft cabin on Transmission Electron Microscope (TEM) grids and boron substrates using a Micro Inertial Impactor (MINI), sampling particles with aerodynamic diameters above and below 1 μm (Kandler et al., 2007). The morphology and elemental composition of individual particles, with a projected area diameter
160 ranging from 0.5 to 3.0 μm , was analyzed using a Environmental Scanning Electron Microscope (FEI ESEM Quanta 400 FEG, Eindhoven, The Netherlands) coupled with an X-Max 150 energy-dispersive X-ray spectroscopy (EDX) detector (Oxford). For selected samples, the mixing state of particles with projected area diameter smaller than 0.5 μm was obtained using a Transmission Electron Microscope (CM20 TEM, Philips, Eindhoven, The Netherlands).

2.3 Data analysis

165 For consistent analysis of the A-LIFE data set, flight time series were divided into 262 sequences, corresponding to flying periods at constant altitude, in the same air mass, and out of clouds (Murphy et al., 2004; Dollner et al., 2024). All data were averaged along these sequences and converted to standard temperature and pressure conditions (STP, 273.15 K; 1013.25 hPa). Data analysis for each instrument is described in the following sections.

2.3.1 Directly measured aerosol optical properties

170 This work focuses on understanding the aerosol optical properties measured directly on board the Falcon research aircraft during A-LIFE by the Aurora 4000 polar nephelometer and the TAP, both installed in-cabin. Therefore, the measured aerosol

optical properties are the optical properties of the aerosol passing through the Falcon sampling system (Schöberl et al., 2024). Details on the operation and data analysis of the Aurora 4000 nephelometer and the TAP are reported in the supplementary information (Sections S1 and S2).

175 The Aurora 4000 polar nephelometer measured the in-cabin particle scattering coefficients ($\sigma_{\text{sp},\lambda}^{\text{Aurora 4000}, \alpha}$) of four angular sectors (from α to 180° , with $\alpha = 0^\circ, 20^\circ, 50^\circ, 90^\circ$) at three wavelengths λ : blue (450 nm), green (525 nm), and red (635 nm). Several calibrations with particle-free air and CO_2 were performed, and the data were corrected following Anderson and Ogren (1998). The total particle scattering coefficients ($\sigma_{\text{sp},\lambda}$) were obtained by correcting $\sigma_{\text{sp},\lambda}^{\text{Aurora 4000}, 0^\circ}$ for angular truncation and illumination as indicated in Teri et al. (2022). The overall uncertainty on $\sigma_{\text{sp},\lambda}$ was calculated to be within $\sim 13 - 30\%$ for
180 sequence averages considered in this work.

The TAP measured the in-cabin particle absorption coefficient ($\sigma_{\text{ap},\lambda}$) at three different wavelengths: blue (465 nm), green (520 nm), and red (640 nm). $\sigma_{\text{ap},\lambda}$ was corrected for filter loading and scattering effects with the Virkkula et al. (2005); Virkkula (2010) correction scheme for the widely used glass fiber Pallflex E70-2075W filters. According to Davies et al. (2019), we assumed an uncertainty on $\sigma_{\text{ap},\lambda}$ of 40% for all three wavelengths.

185 To ensure the accuracy of the data we performed calibrations and data quality controls as reported in the supplementary information (Section S2). In particular, a strong correlation ($R^2 > 0.92$ for all three wavelengths) was found between the particle absorption coefficient and the mass concentration of refractory black carbon (Figure S1 in the supplementary information). The slopes obtained from the linear regression analysis (19.4, 15.9, and $10 \text{ m}^2 \text{g}^{-1}$ for the blue, green, and red wavelength, respectively) are consistent with the literature values of the black carbon mass absorption coefficient (e.g., Ohata et al., 2021). The
190 slightly higher values obtained from our measurements and the observed wavelength dependence are consistent with the presence of the additional contribution of mineral dust absorption. This comparison indicates good quality of the aerosol absorption coefficient measurements.

2.3.2 Intensive aerosol optical properties calculated from direct measurements

Our investigation focused on the intensive aerosol optical properties derived from the Aurora 4000 polar nephelometer and TAP
195 measurements. In particular, we calculated intensive aerosol optical properties that have extensively been used in literature (e.g., Cappa et al., 2016) to distinguish between different aerosol types such as the scattering, absorption and single scattering albedo Ångström exponent (SAE, AAE, SSAE), and intensive aerosol optical properties that are important for understanding and estimating the aerosol direct radiative effect (e.g., Haywood and Shine, 1995; Sherman et al., 2015), such as the asymmetry parameter (g) and the single scattering albedo (SSA), as well as the short-wave direct radiative effect efficiency (SW DREE).
200 An overview of the optical properties used in this study is given in table A1 in appendix.

The scattering Ångström exponent (SAE), which represents the wavelength dependence of the particle scattering coefficient (Ångström, 1929), was calculated by using the total particle scattering coefficients $\sigma_{\text{sp},\lambda}$ measured by the Aurora 4000 polar nephelometer at blue and red wavelengths (i.e., $\lambda_B = 450 \text{ nm}$ and $\lambda_R = 635 \text{ nm}$), as:

$$\text{SAE}_{\lambda_B/\lambda_R} = -\frac{\log(\sigma_{\text{sp},\lambda_B}/\sigma_{\text{sp},\lambda_R})}{\log(\lambda_B/\lambda_R)} \quad (1)$$

205 The absorption Ångström exponent (AAE), which represent the wavelength dependence of the particle absorption coefficient, was calculated by using the particle absorption coefficients $\sigma_{ap,\lambda}$ measured by the TAP at blue and red wavelengths (i.e., $\lambda_B = 465\text{ nm}$ and $\lambda_R = 640\text{ nm}$), as:

$$AAE_{\lambda_B/\lambda_R} = -\frac{\log(\sigma_{ap,\lambda_B}/\sigma_{ap,\lambda_R})}{\log(\lambda_B/\lambda_R)} \quad (2)$$

210 The single scattering albedo Ångström exponent (SSAAE), which represent the wavelength dependence of the single scattering albedo, was calculated according to Moosmüller and Chakrabarty (2011) as:

$$SSAAE_{\lambda_B/\lambda_R} = SAE_{\lambda_B/\lambda_R} - EAE_{\lambda_B/\lambda_R} \quad (3)$$

where EAE is the extinction Ångström exponent calculated at blue and red wavelengths of the Aurora 4000 polar nephelometer (i.e., $\lambda_B = 450\text{ nm}$ and $\lambda_R = 635\text{ nm}$) by using a formula analogous to Eq. 1. The extinction coefficient $\sigma_{ep,\lambda}$ was calculated as the sum of $\sigma_{sp,\lambda}$ and $\sigma_{ap,\lambda}$ measured by the Aurora 4000 polar nephelometer and by the TAP, respectively.

215 We did not apply any correction for the small difference in the wavelengths of the two instruments, since the centroid wavelengths differ less than the full width at half maximum (FWHM) specified by the manufacturer, and we assumed the correction to be negligible.

The asymmetry parameter g , which is the cosine-weighted average of the angular distribution of scattered light (Andrews et al., 2006), was calculated at wavelength λ using the particle scattering coefficients measured by the Aurora 4000 polar nephelometer at different angular sectors ($\sigma_{sp,\lambda}^{\text{Aurora 4000}, \alpha}$, with $\alpha = 0^\circ, 20^\circ, 50^\circ, 90^\circ$) following the method developed by Müller et al. (2012):

$$g\lambda = \frac{\sum_n^{N-1} (\sigma_{sp,\lambda}^{\text{Aurora 4000}, \alpha_n} - \sigma_{sp,\lambda}^{\text{Aurora 4000}, \alpha_{n+1}}) \cos(\frac{\alpha_n + \alpha_{n+1}}{2})}{\sum_n^{N-1} (\sigma_{sp,\lambda}^{\text{Aurora 4000}, \alpha_n} - \sigma_{sp,\lambda}^{\text{Aurora 4000}, \alpha_{n+1}})} 1.7752 - 0.6599 \quad (4)$$

225 The first part of the function is analogous to the definition of the asymmetry parameter. The parameters were found by comparison with the result from scattering theory considering different aerosol types including different sizes and shapes (Müller et al., 2012).

The single scattering albedo (SSA), which is the fraction of the light extinction that is due to scattering, was calculated at wavelength λ using the total particle scattering coefficient measured by the Aurora 4000 polar nephelometer $\sigma_{sp,\lambda}$ and the particle absorption coefficient $\sigma_{ap,\lambda}$ measured by the TAP, as:

$$SSA_\lambda = \frac{\sigma_{sp,\lambda}}{\sigma_{sp,\lambda} + \sigma_{ap,\lambda}} \quad (5)$$

230 The asymmetry parameter (g) and the single scattering albedo (SSA) are key parameters for understanding the aerosol direct radiative effect. We used the g and SSA values measured during A-LIFE for a first estimation of the aerosol direct radiative effect. We used a simple formula derived by Charlson et al. (1991); Haywood and Shine (1995) for calculating the

short-wave aerosol direct radiative effect efficiency (SW DREE) at top-of-atmosphere for an optically thin, partially absorbing atmosphere.¹

235 The SW DREE is the short-wave direct radiative effect (SW DRE) per unit aerosol optical depth (AOD) and is to the first order independent of AOD:

$$\text{SW DREE} = \text{SW DRE}/\text{AOD} = -DS_0T_{\text{atm}}(1 - A_c)\beta\text{SSA}[(1 - R_s)^2 - (2R_s/\text{SSA}\beta)(1 - \text{SSA})] \quad (6)$$

where D is the fractional day length, S_0 is the solar constant, T_{atm} is the atmospheric transmission, A_c is the cloud fraction, R_s is the spectrally averaged surface albedo, SSA is the particle single scattering albedo, and β is the particle average upscatter
240 fraction, i.e. the fraction of the incident solar radiation that is scattered upward to space. β can be obtained from g by using the relationship $\beta = -0.2936g^3 + 0.2556g^2 - 0.4489g + 0.5053$ (Wiscombe and Grams, 1976).

For the non-aerosol-related parameters we used $D = 0.50$, $S_0 = 1370 \text{ Wm}^{-2}$, $T_{\text{atm}} = 0.76$, $A_c = 0.60$, and $R_s = 0.15$ as suggested by Haywood and Shine (1995). Since the values of g_λ and SSA_λ calculated via equations 4 and 5 are the values measured for the aerosol passing through the Falcon sampling system, they are representative for the in cabin NSDs. We
245 applied a correction factor to obtain the values of g and SSA representative for the ambient NSDs ($g_{\lambda,\text{ambient}}$ and $\text{SSA}_{\lambda,\text{ambient}}$). We used $g_{\lambda,\text{ambient}}$ and $\text{SSA}_{\lambda,\text{ambient}}$ at the green wavelength $\lambda = 525 \text{ nm}$ to calculate the SW DREE. A detailed description of the method used to extend the optical properties is in the supplementary information (Section S3).

Although the values obtained from this simplified formula have large uncertainties compared to more advanced radiative transfer calculations, as reported by Wendisch et al. (2001) and Yu et al. (2006), its functional relationship though the SW DREE
250 dependence on SSA and g remain valid. If all others parameters are kept constants, this equation can be used to compare the intrinsic radiative effect efficiency of aerosol at the TOA, as widely done in literature (e.g., Sheridan and Ogren, 1999; Virkkula et al., 2011; Sherman et al., 2015; Shen et al., 2018; Hu et al., 2023).

¹Other publications (e.g., Sherman et al., 2015) refer to this quantity as the short-wave direct radiative forcing efficiency (SW DRFE) adopting the same terminology used by the original authors (Charlson et al., 1991; Haywood and Shine, 1995). However, the term „Radiative Forcing“ is defined as the TOA net radiative flux change due to the change in a climate change driver relative to a pre-industrial reference period (e.g., the year 1750 in the IPCC report AR5). Thus, we used the term „Radiative Effect“, defined as the TOA net radiative flux change resulting from the presence of an atmospheric component.

2.3.3 Aerosol microphysical properties

In this work, we use the microphysical aerosol properties measured during A-LIFE on board of the Falcon research aircraft.

255 An overview of the microphysical properties used for the analysis of this study is given in the table A2 in the appendix.

The mass concentration of refractory black carbon (M_{rBC}) and the mixing state of rBC was measured by the SP2 installed in the cabin of the DLR Falcon research aircraft. The SP2 calibration was performed with fullerene soot as recommended by Schwarz et al. (2010) and the detection range for rBC particles was 70–980 nm mass equivalent diameter, assuming void-free spheres with a bulk density of 1.8 g/cm³ (Moteki et al., 2010) for rBC particles. Although the uncertainty of the calibration fit
260 was found to be about 10 %, we assumed an M_{rBC} uncertainty of 25 % as recommended by Schwarz et al. (2008). Additionally, rBC particles with a heavy coating were identified by comparing the delay time between the incandescent light signals and the scattering light signal.

The number concentration of coarse mode aerosol (N_{coarse}) as well as the coarse mode size distribution was measured in the nominal size range 0.5–50 μm by the wing-mounted UNIVIE CAS. Details of the calibration and data analysis for this
265 instrument are provided in Dollner et al. (2024) and Spanu et al. (2020).

For each A-LIFE flight sequence, the particle number size distribution representing the size range passing through the inlet system (in-cabin NSD) was derived from a combination of measurements with TSI CPC, UHSAS-A, and SkyOPC and parameterized with a combination of three log-normal distributions. The size distribution of the ambient aerosol (ambient NSD) additionally includes measurements from the UNIVIE CAS and covers the size range from 10 nm to 50 μm . For details
270 see Table A2 in the appendix.

Mass concentrations for various size ranges were derived from these two sets of NSDs by assuming particles with a spherical shape and density based on the modeled aerosol composition. In particular, we calculated the mass concentration of sub- μm particles ($M_{\text{sub-}\mu\text{m}}$), the mass concentration of super- μm particles passing through the inlet system ($M_{\text{super-}\mu\text{m in-cabin}}$), and the mass concentration of super- μm particles including the measurements of the wing-mounted CAS instrument ($M_{\text{super-}\mu\text{m ambient}}$).

275 The chemical composition of single particles was obtained from the offline analysis of in-situ samples collected during the A-LIFE research flights. Particles with projected area diameter ranging from 0.5 to 3 μm were analyzed using the environmental scanning electron microscope (ESEM), obtaining their morphology and elemental composition. Particles with projected area diameter smaller than 0.5 μm were analyzed using the transmission electron microscope (TEM), obtaining the mixing state of particles for a few selected samples. The composition and mixing state of aerosol particles during A-LIFE is discussed in
280 section 2.3.6. The composition of individual mineral dust particles with projected area diameter ranging from 0.5 to 3 μm was derived using the ESEM coupled with the energy-dispersive X-ray spectroscopy (EDX) detector, and the mineral dust optical properties for each sample were derived using an empirical approach (Di Biagio et al., 2019; Aryasree et al., 2024). Details and results of this analysis are provided in section S4 of the supplementary information.

2.3.4 A-LIFE aerosol classification

285 During the A-LIFE field experiment, all research flights followed a similar pattern: first, the Falcon aircraft climbed to high altitude to probe the structure of the atmosphere with a wind lidar, and then the aircraft descended to lower altitudes with a step profile, followed by further stepwise ascents and descents. With this approach, 262 measurement sequences at constant altitude outside of clouds were defined. For the investigation of the aerosol properties of different aerosol types, the 262 flight sequences were classified into four main aerosol types, each with three sub-types using the A-LIFE classification scheme (Weinzierl et al.,
290 in preparation, 2024). First, the A-LIFE classification scheme distinguishes measurement periods inside mineral dust layers from other aerosol layers based on the presence of coarse mode particles measured with UNIVIE CAS and on model results from Lagrangian dispersion model FLEXPART which was run in backward mode (Stohl et al., 1998; Seibert and Frank, 2004; Pisso et al., 2019) with emission data from the Copernicus Atmospheric Monitoring Service (CAMS; Morcrette et al., 2009; CAMS, 2019). Mineral dust layers are further classified as Saharan or Arabian dust depending on the origin of the majority of
295 the mineral dust mass modeled with FLEXPART. This classification results in 174 sequences with and 88 sequences without a major contribution of mineral dust. Sequences without major mineral dust contribution are separated into mixtures with enhanced and low coarse mode contribution based on observations of the UNIVIE CAS instrument. Additional distinctions to all four main types (Saharan dust, Arabian dust, mixture with coarse mode, and mixture with low/no coarse mode), are made to study the aerosol properties for different pollution contents. The mass concentration of refractory black carbon (M_{rBC}), as
300 measured by the SP2, is chosen as a proxy for pollution. The degree of pollution is established based on the ratio between the coarse mode aerosol number concentration (N_{coarse}), as measured by the UNIVIE CAS, and M_{rBC} . Thus, based on the pollution degree $N_{\text{coarse}}/M_{\text{rBC}}$, each sequence is assigned to a pollution sub-type (pure, moderately-polluted, polluted). The A-LIFE aerosol classification was validated by comparison with the single particle chemical composition obtained from the offline analysis of in-situ samples collected during the A-LIFE research flights. Details about the A-LIFE aerosol classification
305 can be found in Weinzierl et al. (in preparation, 2024).

2.3.5 Statistical analysis approach

For this study, the A-LIFE data-set was restricted to sequences in the Eastern Mediterranean with major mineral dust contribution according to the A-LIFE aerosol classification and with available intensive aerosol optical properties. Thus, we could investigate the intensive aerosol optical properties for a total of 87 sequences, among which 37, 40, and 10 sequences are clas-
310 sified as pure mineral dust, moderately-polluted mineral dust, and polluted mineral dust, respectively. The sequences classified as Saharan dust and Arabian dust are not evenly distributed among subgroups: 35(2) sequences are classified as pure Saharan (Arabian) dust and pure Arabian dust, 32(8) as moderately-polluted Saharan (Arabian) dust and moderately-polluted Arabian dust, and 10 as polluted Arabian dust, while no sequence classified as polluted Saharan dust was available. Due to the uneven distribution of available sequences among the three sub-groups from the A-LIFE classifications scheme, it was necessary to
315 analyze the change rate of each property with increasing pollution content not only for the three sub-types, but also with a continuum variable. For this continuum variable, we chose a mass ratio which is a dimensionless quantity. Since the mass

concentration of refractory black carbon (M_{rBC}) is a tracer for pollution, we used the ratio between M_{rBC} and the mass of super- μm aerosol particles as a measure of the pollution contribution within the mineral dust layer. We used the mass of super- μm aerosol particles that pass the aerosol inlet system ($M_{\text{super-}\mu\text{m in-cabin}}$) because the intensive aerosol optical properties were also measured inside the aircraft cabin. For the analysis of change rates of the intensive aerosol optical properties representative for the "full" ambient NSDs (i.e., $g_{\lambda,\text{ambient}}$, $\text{SSA}_{\lambda,\text{ambient}}$, and SW DREE, see section S3 in the supplementary information), we used the mass of super- μm aerosol particles in the diameter size range $1 - 50 \mu\text{m}$ ($M_{\text{super-}\mu\text{m ambient}}$).

Figure 2 shows the relationship between the pollution content in terms of mass and the A-LIFE aerosol classification scheme. Sequences classified as pure mineral dust have low values of $M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}}$ (see panel (a)) and $M_{\text{rBC}}/M_{\text{super-}\mu\text{m ambient}}$ (see panel (b)). In contrast, both ratios have higher values for sequences classified as moderately-polluted and polluted mineral dust. Thus, the pollution content increases as these ratios increase.

A higher pollution content also corresponds to a higher relative mass contribution of fine mode aerosols (sub- μm particles). Figure 3 shows the comparison between the mass of the sub- μm aerosols ($M_{\text{sub-}\mu\text{m}}$) and M_{rBC} in panel (a) and between their contribution relative to the mass of super- μm aerosol on a log-log scale in panel (b). $M_{\text{sub-}\mu\text{m}}$ is almost two orders of magnitude larger than M_{rBC} indicating the presence of a large number of small particles other than black carbon. These sub- μm particles can be other anthropogenic particles, such as sulfate, nitrates, or organics, as suggested by the relatively high correlation ($r = 0.59$) between the mass concentration of particles and M_{rBC} . However, a fraction of these sub- μm particles likely also includes mineral dust (see also left panel of Figure 5). The slope of the linear regression line for pure mineral dust is significantly higher than those for moderately-polluted and polluted mineral dust. The correlation coefficients for each sub-type are also higher: $r = 0.91$ for pure, 0.79 for moderately-polluted, and 0.97 for polluted mineral dust, compared to the grouped value. This difference can be due to the impact of sub- μm mineral dust particles, which influence the $M_{\text{sub-}\mu\text{m}}/M_{\text{rBC}}$ ratio more in the case of pure mineral dust than in the polluted case.

Nevertheless, the high correlation coefficient between the ratios $M_{\text{sub-}\mu\text{m}}/M_{\text{super-}\mu\text{m in-cabin}}$ and $M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}}$ ($r = 0.89$) indicates the equivalence of choosing one of these two ratios as a measure of the pollution contribution in terms of mass within the mineral dust layer. Due to the skewed distribution of the data, a log transformation is performed and $\log_{10}(M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}})$ is used to evaluate the change rate of aerosol properties for increasing pollution content within the mineral dust layer.

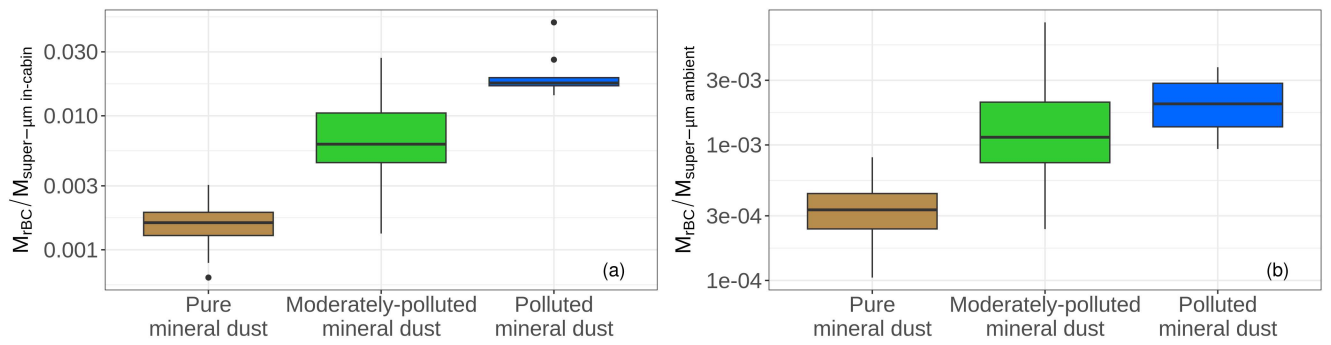


Figure 2. Box-plot of the pollution contribution in terms of mass for the three sub-types: pure dust (brown), moderately-polluted dust (green), and polluted dust (blue) according to the A-LIFE aerosol classification. Panel (a) shows the pollution contribution in terms of mass for aerosol particles passing the Falcon aerosol sampling system ($M_{rBC}/M_{super-\mu m \text{ in-cabin}}$). Panel (b) shows the pollution contribution in terms of mass for the ambient NSD. The plots show results for 87 sequences, among which 37, 40, and 10 are classified as pure mineral dust, moderately-polluted mineral dust, and polluted mineral dust, respectively. The boxes indicate the interquartile range (IQR $25^{th} - 75^{th}$), the horizontal lines the median, the whiskers the largest value that is not greater than $1.5 \cdot IQR$, and data points outside this range are shown as single dots.

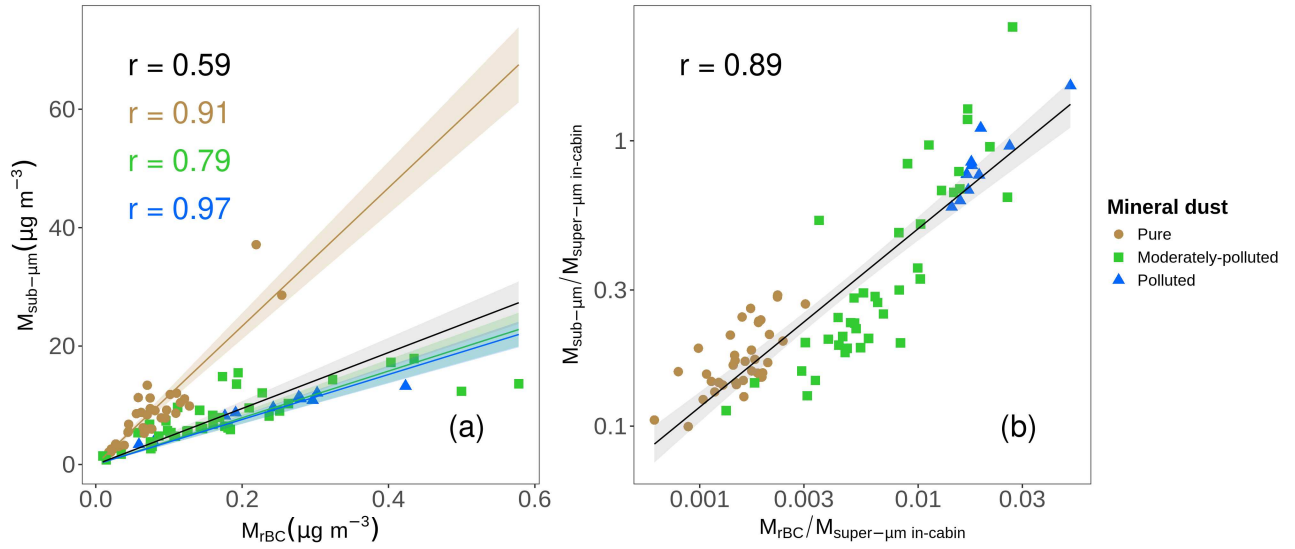


Figure 3. Mass of sub- μm particles and mass of refractory black carbon. Panel (a) shows the comparison between the mass of sub- μm particles ($M_{\text{sub-}\mu\text{m}}$) and the mass of refractory black carbon (M_{rBC}). Panel (b) shows the comparison on a log-log scale between the contribution of particle size in terms of mass ($M_{\text{sub-}\mu\text{m}}/M_{\text{super-}\mu\text{m in-cabin}}$) and the pollution contribution in terms of mass ($M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}}$), where $M_{\text{super-}\mu\text{m in-cabin}}$ is the mass of super- μm particles passing through the aerosol inlet of the DLR Falcon aircraft (Schöberl et al., 2024). The plots show results for 87 sequences, among which 37, 40, and 10 are classified as pure mineral dust, moderately-polluted mineral dust, and polluted mineral dust, respectively, following the A-LIFE aerosol classification as indicated by the color code. Pearson's correlation coefficients r and correlation lines are provided on each panel, in black considering all the points and following the color code when considering the pollution sub-types individually.

2.3.6 Composition and mixing state of aerosol particles during the A-LIFE field experiment

Not only the pollution content within a mineral dust layer can affect its optical properties, but also the mixing state of the aerosol (internal versus external mixture) might have an influence. For example, the absorption of BC particles is enhanced when a coating is present on them (e.g. Liu et al., 2015). In addition, the use of a simplified mixing state in model simulations can lead to significant uncertainties in the derived optical properties (Yao et al., 2022). Thus, the particle morphology and elemental composition were investigated using electron microscopy techniques.

Figure 4 shows examples of images of different aerosol particle types collected during the A-LIFE field experiment for three randomly-selected samples, collected in pure, moderately-polluted, and polluted mineral dust. The images were obtained with the transmission electron microscope (TEM) for particles with projected area diameter smaller than $0.5\ \mu\text{m}$ (on the left) and with the environmental scanning electron microscope (ESEM) for particles with projected area diameters ranging from 0.5 to $3\ \mu\text{m}$ (on the right). TEM images reveal individual and mixed particles, i.e., sulfate, soot and mineral dust particles, but also soot with a sulfate coating in core-shell structures, while ESEM images primarily show mineral dust particles.

Figure 5 shows a quantitative analysis of the different particle types and mixing states for the same three samples shown in Figure 4. The two graphs report values obtained for projected area diameter smaller than $0.5\ \mu\text{m}$ and in the range $0.5 - 3\ \mu\text{m}$, respectively. In the larger diameter range, mineral dust is the predominant particle type observed in all three samples. This result is consistent with observations from other samples collected during the A-LIFE field experiment. Additionally, the analysis revealed that between 2 and 10% of the collected mineral dust particles were found in internal mixtures with sulfate. Previous studies conducted in Saharan dust near-source and outflow regions (Usher et al., 2002; Kandler et al., 2011, 2020), demonstrated that internal mixtures of mineral dust with sulfate particles are frequently observed in polluted environments and they are often due to heterogeneous reactions. In the smaller diameter range, mineral dust particles mixed with soot, sulfate, or other components were less than 1%, and the particle composition varied notably across the three samples. The pure mineral dust sample contained over 80% of mineral dust particles, with minor amounts of soot and sulfate particles. In contrast, the polluted mineral dust sample had over 60% of soot particles, and around 3% in a core-shell structure of soot coated with sulfate. The moderately-polluted sample contained approximately 70% spherical sulfate particles, along with around 20% of soot particles coated with sulfate in a core-shell structure. The core-shell particle amounts observed in transmission electron microscope images align with SP2 results. Figure 6 shows the fraction of rBC particles with a heavy coating across the 87 sequences considered in this study. The coated fraction is less than $\sim 5\%$ in half of the sequences and below $\sim 20\%$ in all, making the three samples representative of rBC mixing states across all sequences.

We would like to emphasize that the particle morphology and mixing state information based on TEM and ESEM analysis in this study is limited to three samples. The impact of particle mixing state on the aerosol optical properties should be investigated in a future study with additional data on particle mixing state.

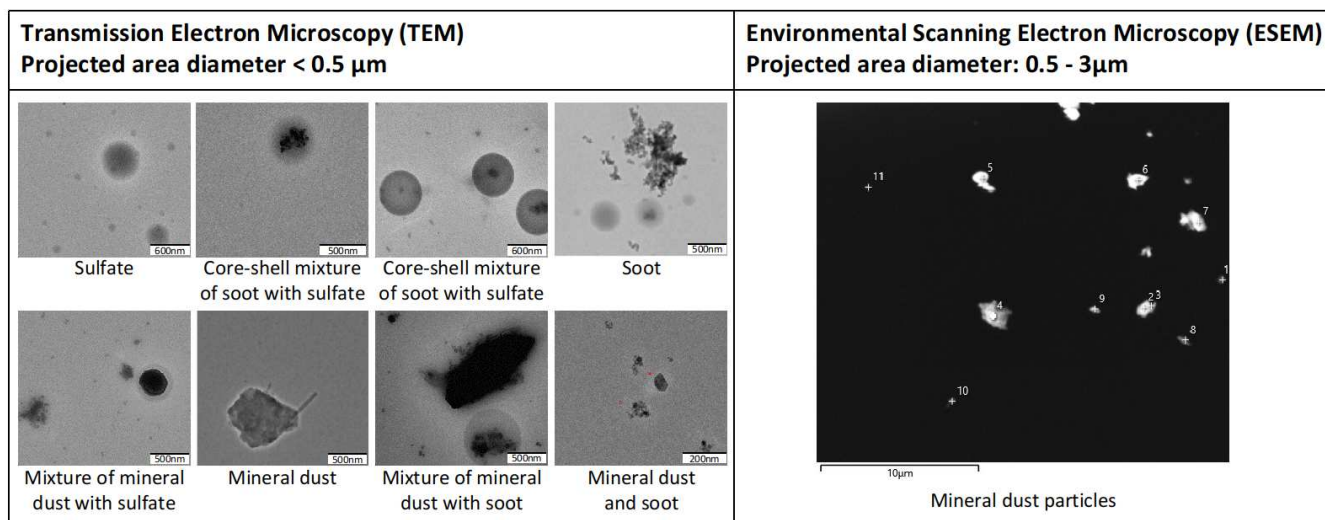


Figure 4. Examples of images of different aerosol particle types and their mixing state from three randomly-selected samples, collected during measurements in pure, moderately-polluted, and polluted mineral dust. The figure shows images of particles collected during the A-LIFE field experiment and observed with the transmission electron microscope for particles with projected area diameter smaller than 0.5 μm (on the left), and with the environmental scanning electron microscope for particles with projected area diameter in the range 0.5 – 3.0 μm (on the right).

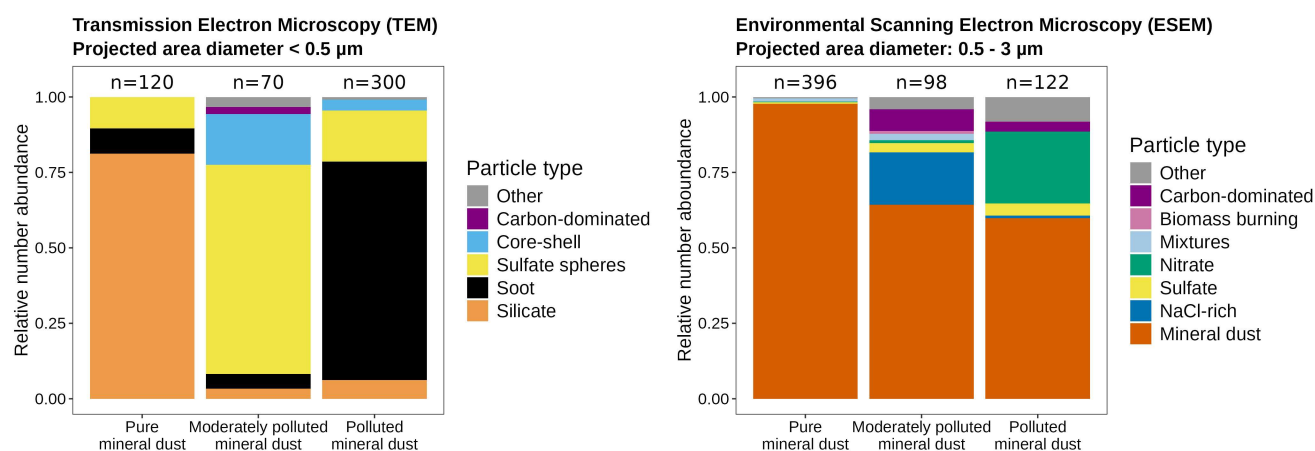


Figure 5. Chemical composition and mixing state of particles. The graphs show the relative number abundance of various aerosol particle types and their mixing state for the same three samples shown in Figure 4. The graph on the left shows the observations with the transmission electron microscope (TEM) for particles with a projected area diameter smaller than 0.5 μm , and the graph on the right shows the observations with the environmental scanning electron microscope (ESEM) for particles with a projected area diameter in the range 0.5 – 3 μm . The number of particles (n) analyzed for each sample is provided above the corresponding bar. Note that "Silicate" in the left panel is a subset of mineral dust in the right panel.

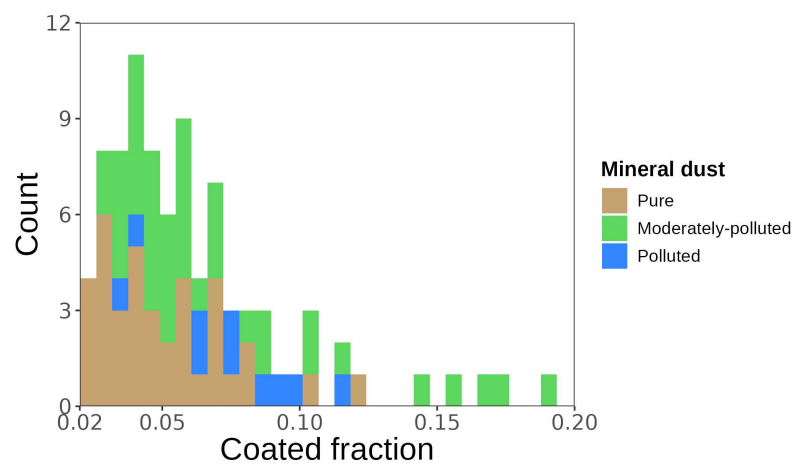


Figure 6. Coated fraction of rBC particles derived from SP2 measurements. The plot shows results for 87 sequences, among which 37, 40, and 10 are classified as pure, moderately-polluted, and polluted mineral dust, respectively, following the A-LIFE aerosol classification as indicated by the color code.

3 Results

To evaluate the impact of mixing pollution and mineral dust from different source regions on the intensive aerosol optical properties, we investigated the intensive aerosol optical properties of sequences measured in-cabin of the Falcon aircraft classified as pure, moderately-polluted, and polluted mineral dust. Table 1 shows the summary statistics for the intensive aerosol optical properties (SAE, AAE, SSAE, g , and SSA) for pure, moderately polluted, and polluted mineral dust sequences. The intensive aerosol optical properties of pure and polluted mineral dust show significantly different values, as confirmed by the analysis of variance ². However, moderately-polluted mineral dust presents a more complex picture. It exhibits significant differences from the other two groups in SAE, g at all wavelengths, and SSA at the red wavelength. Nevertheless, it shows no significant differences from polluted mineral dust in AAE and SSA at blue and green wavelengths, nor from pure mineral dust in SSAE.

In addition, we investigated the relationship between the intensive aerosol optical properties and $M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}}$, which expresses the pollution contribution in terms of mass in the mineral dust layer. Figure 7 shows each intensive aerosol optical property (SAE, AAE, SSAE, g , SSA, and SW DREE) as a function of the pollution contribution in terms of mass $M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}}$ on a log-scale. The relationships between the intensive aerosol optical properties and $\log_{10}(M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}})$ are close to linear. Least-square linear regression analysis was performed for all mineral dust sequences and for the mineral dust from the two source regions (the Arabian Peninsula and the Saharan desert) individually. Results for the linear regression analysis are reported in Table 2.

The Ångström exponents (SAE, AAE, and SSAE), which are intensive aerosol optical properties often used for identifying aerosol types, change significantly with increasing pollution contribution in the mineral dust layer. The relationships between each Ångström exponent and the pollution contribution are statistically significant, as indicated by the p -values well below the threshold of 0.05. For a unit increase of $\log_{10}(M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}})$, SAE increases by 1.33 ± 0.09 , AAE decreases by 2.0 ± 0.3 , and SSAE increases by $+0.08 \pm 0.01$. The determination coefficient R^2 values suggest that the pollution contribution accounts for 72% of the variability in SAE, while it explains 43%, and 30% of the variability in AAE, and SSAE, respectively.

The asymmetry parameter and single scattering albedo (g and SSA) also change significantly ($p < 0.001$) with increasing pollution contribution in the mineral dust layer. When $\log_{10}(M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}})$ increases by a unit, g_{λ_G} decreases by 0.08 ± 0.01 and SSA_{λ_G} decreases by 0.079 ± 0.006 . The pollution contribution is responsible for 49% and 67% of the variability in g and SSA, respectively. Median values of g and SSA measured at the wavelength λ_G change from 0.67 to 0.56 and from 0.96 to 0.89, respectively, from pure mineral dust to polluted mineral dust.

For the intensive aerosol optical properties representative for the ambient NSD (see Figure B1 and Table B1 in the appendix), the effect of increasing the pollution contribution is also significant, as indicated by the p -value < 0.001 . When $\log_{10}(M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}})$ increases by a unit, $g_{\lambda_G, \text{ambient}}$ decreases by 0.05 ± 0.01 and $\text{SSA}_{\lambda_G, \text{ambient}}$ decreases by 0.044 ± 0.007 . From pure mineral dust to polluted mineral dust, the median value of $\text{SSA}_{\lambda_G, \text{ambient}}$ change from 0.93 to 0.89; the median value of $g_{\lambda_G, \text{ambient}}$ change from 0.71 to 0.56.

²Statistical tests were conducted using R (R Core Team, 2021) and following the methodology outlined in Bevens (2023) to assess significant differences between group means (analysis of variance, ANOVA), and locate specific differences (Tukey's HSD, Honestly Significant Difference, post-hoc test).

These results suggest that also the direct radiative effect of mineral dust layers might be affected by the increase of pollution content. For the first estimation of this impact, the short-wavelength direct radiative effect efficiency (SW DREE) was calculated for our data. However, the result of the linear regression analysis between SW DREE and the pollution contribution indicates that the relationship is not statistically significant ($p > 0.05$). The SW DREE increases only slightly by $0.7 \pm 0.9 W m^{-2} AOD^{-1}$ for a unit increase of $\log_{10}(M_{rBC}/M_{super-\mu m \text{ ambient}})$. Thus, our data do not provide enough evidence to conclude that the SW DREE changes with increasing pollution content when considering the global average surface albedo.

The results for the mineral dust from the two source regions (the Arabian Peninsula and the Saharan desert) are similar. The slopes and intercepts obtained from the linear regression analysis are in agreement within the error bars for almost all intensive aerosol optical properties. Thus, the change rate of the intensive aerosol optical properties (SAE, AAE, SSAE, g , and the SW DREE) for increasing pollution contribution in terms of mass agree within error bars for the mineral dust from the two different source regions. Small differences are found for the SSA. Indeed, the intercept is 0.77 ± 0.02 and 0.71 ± 0.02 for Arabian and Saharan dust, respectively. For a unit increase of $\log_{10}(M_{rBC}/M_{super-\mu m \text{ in-cabin}})$, SSA decreases by 0.068 ± 0.009 for Arabian dust and by 0.087 ± 0.008 for Saharan dust.

Table 1. Summary statistics (median and four percentiles) for each aerosol type of the intensive aerosol optical properties measured inside the cabin of the Falcon research aircraft behind an aerosol sampling system during A-LIFE for 87 measurement sequences among which 37, 40, and 10 are classified as pure, moderately-polluted, and polluted mineral dust by the A-LIFE aerosol classification scheme. For g_{λ} data were only available for 66 sequences, among which 29, 29, and 8 are classified as pure, moderately-polluted, and polluted mineral dust, respectively.

		$SAE_{\lambda_B/\lambda_R}$	$AAE_{\lambda_B/\lambda_R}$	$SSAE_{\lambda_B/\lambda_R}$	g_{λ_B}	g_{λ_G}	g_{λ_R}	SSA_{λ_B}	SSA_{λ_G}	SSA_{λ_R}
Pure mineral dust	median	-0.02	3.50	-0.10	0.68	0.67	0.69	0.95	0.96	0.98
	3^{rd}	-0.58	0.82	-0.18	0.62	0.63	0.65	0.92	0.94	0.95
	25^{th}	-0.17	2.48	-0.11	0.66	0.65	0.67	0.95	0.95	0.98
	75^{th}	0.16	4.22	-0.07	0.69	0.69	0.70	0.96	0.97	0.99
	97^{th}	0.48	7.04	-0.01	0.74	0.72	0.75	0.98	0.98	1.00
Moderately-polluted mineral dust	median	0.58	1.92	-0.08	0.61	0.63	0.64	0.91	0.91	0.94
	3^{rd}	-0.14	0.68	-0.16	0.56	0.56	0.52	0.86	0.84	0.85
	25^{th}	0.30	1.52	-0.11	0.59	0.59	0.61	0.89	0.89	0.91
	75^{th}	0.97	2.36	-0.05	0.64	0.65	0.66	0.93	0.93	0.96
	97^{th}	2.18	2.79	0.11	0.67	0.70	0.69	0.95	0.95	0.97
Polluted mineral dust	median	1.73	1.70	0.01	0.56	0.56	0.54	0.89	0.89	0.88
	3^{rd}	1.29	1.48	-0.05	0.49	0.50	0.50	0.86	0.84	0.85
	25^{th}	1.57	1.65	-0.00	0.55	0.54	0.52	0.88	0.87	0.88
	75^{th}	1.89	1.75	0.02	0.57	0.57	0.56	0.90	0.89	0.90
	97^{th}	2.02	2.16	0.08	0.58	0.59	0.58	0.95	0.94	0.95

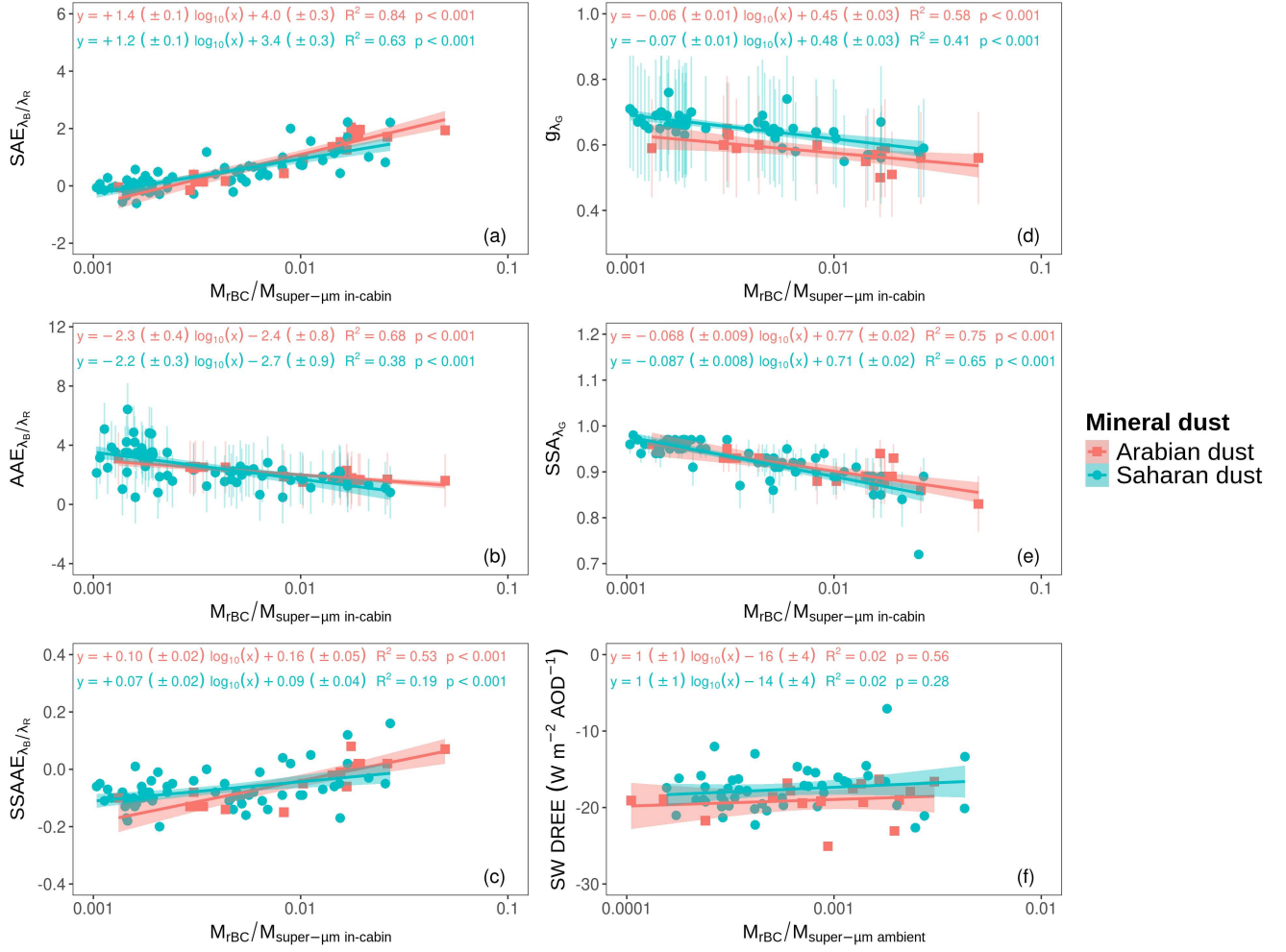


Figure 7. Intensive aerosol optical properties of mixtures of pollution and mineral dust from different source origins. Each panel shows a different intensive optical property as a function of the pollution contribution in terms of mass ($M_{rBC}/M_{\text{super-}\mu\text{m in-cabin}}$): **(a)** wavelength dependence of the scattering coefficient at $\lambda = 450$, and 635 nm ($SAE_{\lambda_B}/\lambda_R$), **(b)** wavelength dependence of the absorption coefficient at $\lambda = 465$, and 640 nm ($AAE_{\lambda_B}/\lambda_R$), **(c)** wavelength dependence of the single scattering albedo ($SSAE_{\lambda_B}/\lambda_R$), **(d)** asymmetry parameter (g) at $\lambda = 525 \text{ nm}$, **(e)** single scattering albedo (SSA) at $\lambda = 525 \text{ nm}$, and **(f)** short-wave direct radiative effect efficiency (SW DREE) calculated with a simplified formula (Haywood and Shine, 1995), the measured values of g and SSA at $\lambda = 525 \text{ nm}$ and global average parameters. g and SSA were extended to the ambient particle size distribution (see section S3 in the supplementary information). Thus, the SW DREE is plotted as function of $M_{rBC}/M_{\text{super-}\mu\text{m ambient}}$, where $M_{\text{super-}\mu\text{m ambient}}$ is the mass of the ambient super- μm particles.

The plots show results for 67(20) Saharan (Arabian) dust sequences. Results for g_λ and SW DREE are based on 49(17) Saharan (Arabian) dust sequences. The color code indicates the mineral dust type. Least squares regression results, including regression line and equation, determination coefficient and p -value, are shown in the corresponding mineral dust type color.

Table 2. Results of the linear regression analysis of each intensive aerosol optical property vs. the pollution contribution in terms of mass $\log_{10}(M_{\text{rBC}}/M_{\text{super-}\mu\text{m in-cabin}})$. These results were derived from 87 sequences classified as mineral dust (67 Saharan, 20 Arabian dust sequences). Result for g_{λ} and SW DREE are based on 66 sequences (49 Saharan, 17 Arabian dust sequences). The data used for the analysis are shown in Figure 7.

		$\text{SAE}_{\lambda_B/\lambda_R}$	$\text{AAE}_{\lambda_B/\lambda_R}$	$\text{SSAAE}_{\lambda_B/\lambda_R}$	g_{λ_G}	SSA_{λ_G}	SW DREE
All	slope	+1.33 ($\pm$ 0.09)	-2.0 ($\pm$ 0.3)	+0.08 ($\pm$ 0.01)	-0.08 ($\pm$ 0.01)	-0.079 ($\pm$ 0.006)	+0.7 ($\pm$ 0.9)
	intercept	+3.7($\pm$ 0.2)	-2.3 ($\pm$ 0.6)	+0.12($\pm$ 0.03)	+0.45 ($\pm$ 0.02)	+0.74 ($\pm$ 0.01)	-16 ($\pm$ 3)
	R ²	0.72	0.43	0.3	0.49	0.67	0.01
	significance	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p = 0.4$
Arabian dust	slope	+1.4 ($\pm$ 0.1)	-2.3 ($\pm$ 0.4)	+0.10 ($\pm$ 0.02)	-0.06 ($\pm$ 0.01)	-0.068 ($\pm$ 0.009)	+1 ($\pm$ 1)
	intercept	+4.0($\pm$ 0.3)	-2.4 ($\pm$ 0.8)	+0.16 ($\pm$ 0.05)	+0.45 ($\pm$ 0.03)	+0.77 ($\pm$ 0.02)	-16 ($\pm$ 4)
	R ²	0.84	0.68	0.53	0.58	0.75	0.02
	significance	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p = 0.56$
Saharan dust	slope	+1.2 ($\pm$ 0.1)	-2.2 ($\pm$ 0.3)	+0.07 ($\pm$ 0.02)	-0.07 ($\pm$ 0.01)	-0.087 ($\pm$ 0.008)	+1 ($\pm$ 1)
	intercept	+3.4($\pm$ 0.3)	-2.7 ($\pm$ 0.9)	+0.09($\pm$ 0.04)	+0.48 ($\pm$ 0.03)	+0.71 ($\pm$ 0.02)	-14 ($\pm$ 4)
	R ²	0.63	0.38	0.19	0.41	0.65	0.02
	significance	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p < 0.001$	$p = 0.28$

4 Discussion

4.1 Comparison of mineral dust's optical properties from Arabian and Saharan source regions

Differences in the optical properties of mineral dust particles from different source regions can arise due to differences in the mineralogical compositions (Sokolik and Toon, 1999; Di Biagio et al., 2019), but also due to differences in the mixing processes with pollution (Kok et al., 2023; Denjean et al., 2016; Seinfeld et al., 2004). We investigated the differences in mineral dust layers' intensive aerosol optical properties (SAE, AAE, SSAE, g , SSA, and SW DREE) from two different source regions (the Arabian Peninsula and the Saharan desert) in the Eastern Mediterranean. We found that the intensive aerosol optical properties change significantly with the increase of pollution content within the mineral dust layer. On the contrary, the differences between Arabian and Saharan dust are mostly not significant (see Figure 7) for all intensive aerosol optical properties except for the SSA, where minor differences are found. These results suggest that there is no major intrinsic difference in the mineral dust from Arabian and Saharan dust source regions that causes a significant difference in the intensive aerosol optical properties.

Our findings align with observations of other in-situ aerosol properties measured during the A-LIFE field experiment such as the single particle chemistry analysis and the particle volume size distributions, and are also consistent with previous research. For instance, Kim et al. (2011) analyzed the mineral dust optical properties using sun photometers at 14 AERONET sites. Their study found no systematic difference in the Ångström exponent, single scattering albedo and asymmetry parameter for pure mineral dust layers from the Arabian and Saharan source regions.

Single particle chemistry analysis of mineral dust particles reveals comparable features with only slight variations between Arabian and Saharan dust samples (see Figure S4 and S5 in the supplementary information). Both exhibit a high percentage ($\sim 60 - 62\%$) of silicate-like particles, while Saharan dust samples contain more clay-like minerals (Illite and Kaolinite), and Arabian dust samples show higher carbonate content. These observations align with previous Saharan dust studies (Kandler et al., 2009; Formenti et al., 2011b). Limited studies on Arabian dust (Attiya and Jones, 2020; Engelbrecht et al., 2009, 2016) reveal significant variability in mineralogy across the Arabian Peninsula. Thus, identifying specific characteristics of Arabian dust remains challenging, but studies consistently note higher carbonate and lower clay content compared to Saharan dust.

Our observations suggest that Saharan dust particles were slightly more absorbing than Arabian dust particles over the Eastern Mediterranean. The linear regression analysis intercepts for directly measured SSA were 0.77 ± 0.02 and 0.71 ± 0.02 for Arabian and Saharan dust, respectively (see Figure 7(f)). This outcome aligns with the small difference found in the amount of iron oxides and hydroxides obtained from single particle chemistry analysis. Saharan dust contains about 3% these compounds, whereas Arabian dust samples exhibit roughly half that amount. This difference results in minor variations in the imaginary part of the refractive index, as shown in Figure S6 (supplementary information). For instance, the imaginary part of the refractive index, at the green wavelength 520 nm, has a value of 0.006 for Saharan dust and of 0.005 for Arabian dust. Consequently, the calculated SSA values of mineral dust particles indicate a 4% increase for Arabian dust compared to Saharan dust. The observed imaginary refractive index values are higher than those of Saharan dust measured in other locations, such as over the Northwest Sahara or the Atlantic (Kandler et al., 2007, 2011; Aryasree et al., 2024).

On the contrary, differences between Arabian and Saharan dust were observed in the lidar ratio by several past studies (e.g., Schuster et al., 2012; Mamouri et al., 2016; Nisantzi et al., 2015), as well as during the A-LIFE field experiment (Groß et al., 2024). Higher values are reported for the lidar ratio of Saharan dust compared to the one of Arabian dust. The lidar ratio is an important intensive aerosol optical property in the framework of aerosol typing effort. It is obtained as the ratio of the extinction coefficient and the back-scattering coefficient at 180° measured by a lidar, and it is sensitive to the particle composition, particle size distribution, and particle shape. According to Groß et al. (2013), the lidar ratio decreases with increasing particle size and increases with increasing light absorption. In addition, the lidar ratio decreases with increasing real part of the refractive index (Gasteiger et al., 2011).

The reason for the differences in Arabian and Saharan dust's lidar ratio is unknown. Most of the authors attribute the lidar ratio difference to the different geochemical characteristics of the source region soil, with different illite concentrations for the two source regions, which influence the real part of the refractive index (e.g., Schuster et al., 2012; Filioglou et al., 2020). However, the lidar ratio can be influenced for various reasons. For example, Hu et al. (2020) pointed out that the lidar ratios can also be influenced by the presence of coarse-mode and giant ($d > 40\mu m$) particles, for example, when measured close to the source region. Nisantzi et al. (2015) acknowledges that the retrieval of the mineral dust lidar ratio may be affected by pollution if present in the boundary layer or mixed into the mineral dust layer.

Understanding the reason for the differences in the lidar ratios of Arabian and Saharan dust is out of the scope of this paper. However, we highlight that care is needed when retrieving the optical properties of pure mineral dust because they may contain mixtures of other components, such as black carbon or other small anthropogenic particles. In particular, when analyzing the differences between different dust types, evaluating the pollution content and comparing only measurements with similar pollution content is essential. While the pollution content has shown a significant effect on the mineral dust intensive optical properties, the mineral dust source region (the Arabian Peninsula or the Saharan desert) does not appear to significantly affect these properties.

4.2 Implications for the identification of aerosol types

The scattering, absorption and single scattering albedo Ångström exponents (SAE, AAE, and SSAE) are sensitive to different aerosol characteristics. Thus, different aerosol classification schemes deploying these properties exist in literature (e.g., Russell et al., 2010; Lee et al., 2012; Costabile et al., 2013; Cazorla et al., 2013; Cappa et al., 2016). Most of these studies agree that high SAE values suggest the presence of fine mode aerosols (e.g., BC, sulfates, or nitrates) and low SAE values indicate large aerosols (e.g., sea salt and mineral dust). Furthermore, they indicate that AAE values around 1 are associated with BC and higher AAE values suggest the presence of mineral dust and/or brown carbon. In addition, negative SSAE values have been associated with mineral dust outbreaks (e.g., Collaud Coen et al., 2004). However, different studies report slightly different threshold values for different aerosol types, likely due to differences in the particle size distribution measured at different sites and with different techniques and to different wavelength pairs used to calculate the intensive aerosol optical properties. Different classification schemes and the respective threshold values are reviewed in Schmeisser et al. (2017) and Valentini et al. (2020).

We compared our measurements with the classification scheme proposed by Cappa et al. (2016). Figure 8 shows the classification matrix (AAE vs. SAE) for the sequences classified as mineral dust by the A-LIFE aerosol classification scheme. In the upper panel, the points are color-coded by the SSAAE. In the lower panel, the color indicates the aerosol type according to the A-LIFE aerosol classification scheme.

For pure mineral dust sequences, our measurements align with observations of previous studies. Measured median values for pure mineral dust sequences are -0.02 , 3.50 , and -0.10 for SAE, AAE, and SSAAE, respectively (see Table 1). Most sequences classified as pure mineral dust fall in the "Dust dominated region" of the AAE vs. SAE plot according to the classification scheme proposed by Cappa et al. (2016) ($SAE < 0.5$, and $AAE > 2$). In addition, all pure mineral dust cases have SSAAE < 0 , consistently with values reported by Collaud Coen et al. (2004) and Kaskaoutis et al. (2021).

However, our results show that the Ångström exponents change significantly for increasing pollution content within the mineral dust layer (Figure 7 and Table 2). Median values for polluted mineral dust sequences are 1.73 , 1.70 , and 0.01 for SAE, AAE, and SSAAE, respectively (see Table 1). These values are more similar to values measured for anthropogenic aerosols (e.g., Lee et al., 2012, $SAE = 0.8 - 1.8$, $AAE = 1.0 - 1.5$). Indeed, polluted mineral dust sequences are mainly in the "Mixed BC/BrC region" of the AAE vs. SAE according to the Cappa et al. (2016) classification scheme (Figure 8). Moderately-polluted mineral dust sequences have SAE, AAE, and SSAAE values with a larger variability. Thus, these sequences fall in different regions according to the Cappa et al. (2016) classification scheme, including "Dust dominated", "Mixed Dust/BC/BrC", and "BC dominated" regions. Thus, mineral dust can be present for several combinations of the optical properties SAE, AAE, and SSAAE.

This result indicates that the Ångström exponents (SAE, AAE, and SSAAE) should not be used as sole indicators to detect the presence of mineral dust in case of mixing with pollution, because the optical properties shift towards values associated with anthropogenic aerosols as the presence of pollution increases within the mineral dust layer. A few authors already pointed out that the presence of pollution may mask the mineral dust signal (Esteve et al., 2012; Ealo et al., 2016; Pandolfi et al., 2018). Therefore, independent methods are needed to identify mineral dust events in mixtures with pollution.

Once the presence of mineral dust is already elucidated, the Ångström exponents (SAE, AAE, and SSAAE) can be useful parameters to estimate the amount of pollution mixed into the mineral dust layer. Indeed, a relationship between all the three Ångström exponents (SAE, AAE, and SSAAE) and the pollution contribution in terms of mass exists (Figure 7, Table 2). Therefore, the Ångström exponents (SAE, AAE, and SSAAE) can be used to select cases where non-dust aerosols have a minimal impact on the aerosol optical properties and study the properties of pure mineral dust. For example, Müller et al. (2011), Kim et al. (2011), Horvath et al. (2018), and Tian et al. (2018) used SAE and or AAE to select pure mineral dust cases and study other optical and microphysical properties. In addition, among the three Ångström exponents (SAE, AAE, and SSAAE), the pollution contribution in terms of mass seems to better account for the variability in SAE (72%), while for AAE and SSAAE, the pollution contribution in terms of mass explains 43% and 30% of the variability (see Table 2). This result suggests that SAE might be the most reliable intensive aerosol optical property for estimating the amount of pollution in a mineral dust layer. However, it is crucial to confirm the presence of mineral dust using additional, independent methods.

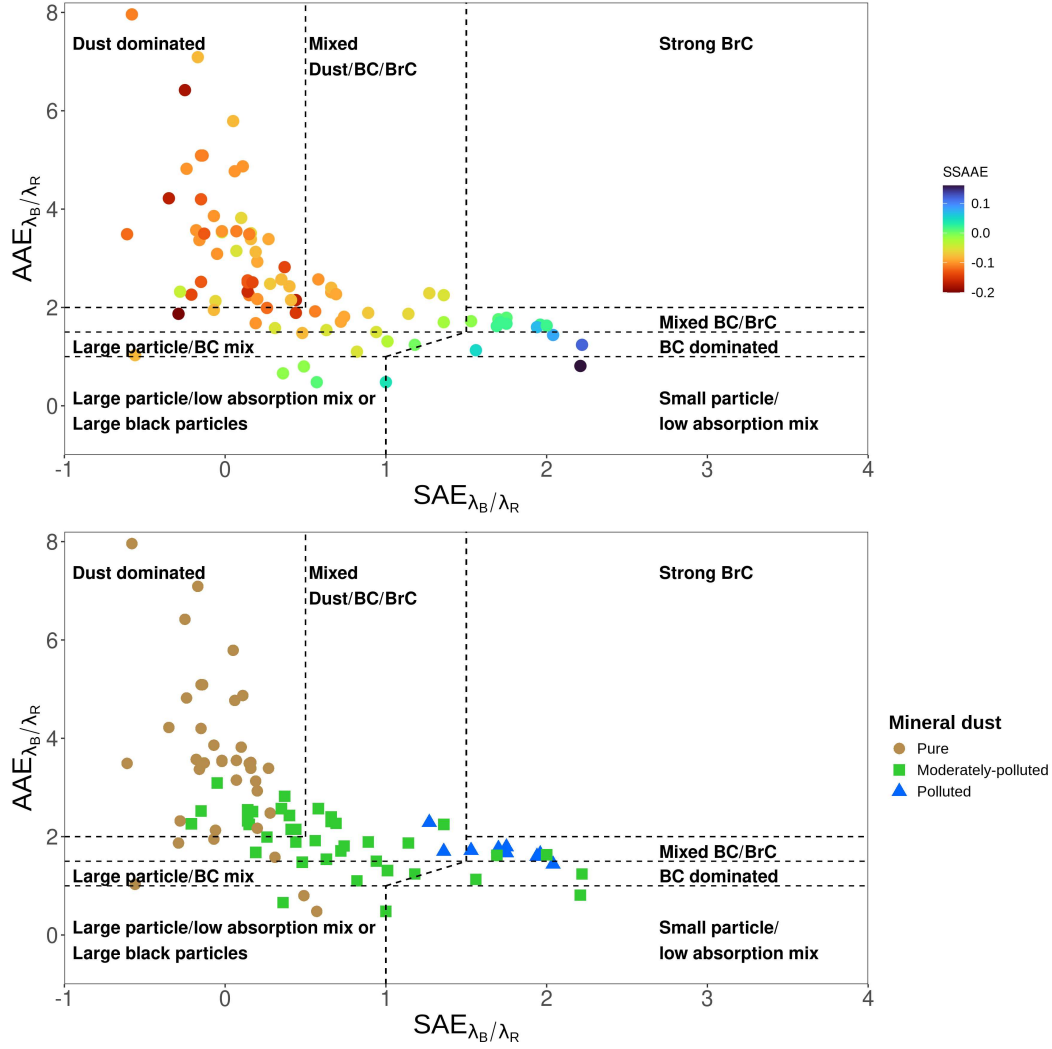


Figure 8. Aerosol classification schemes using intensive aerosol optical properties. Both panels show the absorption vs. scattering Ångström exponent matrix ($AAE_{\lambda_B/\lambda_R}$ vs. $SAE_{\lambda_B/\lambda_R}$) overlaid with the Cappa et al. (2016) aerosol classification scheme: aerosol types are reported in black, and the thresholds are indicated with dashed black lines. In the upper panel, the color code indicates the single scattering albedo Ångström exponent ($SSAAE_{\lambda_B/\lambda_R}$). In the lower panel, the colors and shapes indicate the aerosol types following the A-LIFE aerosol classification scheme.

4.3 Asymmetry parameter and single scattering albedo of pure mineral dust layers

The asymmetry parameter (g) and the single scattering albedo (SSA) are key intensive aerosol optical properties to understand the direct radiative effect of mineral dust. The large variability of these optical properties for mineral dust in the atmosphere may be due to differences in the mineralogical composition and mixture with other aerosol types. Still, it can also arise from differences in the measurement methods or retrieval technique and the measured particle diameter size range.

For pure mineral dust layers, the values of g and SSA measured during A-LIFE are mainly consistent with literature values measured during different field campaigns. We consider the values of g and SSA measured in the aircraft cabin representative of the in-cabin NSDs, as well as the values obtained extending these values to the ambient NSDs (10 nm - 50 μ m), which are thus representative of the ambient NSD.

Figures 9 shows the spectral values of g for sequences classified as pure mineral dust directly measured in-cabin of the Falcon aircraft and extended to the ambient NSD, as well as, values from other field campaigns. Values of g from our measurements are in the range of values from other measurement campaigns, and a better agreement is shown for values extended to the ambient NSD (Figure 9). The value of g at the green wavelength (525 nm) extended to the ambient size distribution (10 nm – 50 μ m) is in excellent agreement with the value ($g = 0.71 \pm 0.03$) measured directly with a polar nephelometer by Horvath et al. (2018) in Sierra Nevada (Spain). Values of g for pure mineral dust obtained from other aircraft measurements performed close to Saharan dust source regions (e.g., Formenti et al., 2011a; Ryder et al., 2018; Denjean et al., 2020) or after transport through the Mediterranean (Denjean et al., 2016) agree with our measurements values extended to the ambient NSD. These studies calculated the intensive optical properties through an inversion closure study combining the particle number size distribution in various size ranges, as indicated in Figures 9 and 10, with direct measurements of the optical properties and the Mie theory.

Figure 10 shows the spectral single scattering albedo (SSA) for pure mineral dust layers in this work in comparison with the results of other studies. Our measurements of SSA are consistent with the most values from other measurement campaigns using inversion closure study (e.g., Formenti et al., 2011a; Ryder et al., 2018; Denjean et al., 2016), but also from direct measurements (e.g., Schladitz et al., 2009; Müller et al., 2011). However, most mean values from other campaigns fall within the range between the values for our in-cabin and ambient SSA values. For example, the SSA at $\lambda = 525$ nm changes from a value measured in-cabin of 0.96 ± 0.02 to a value representative for the ambient NSD of 0.93 ± 0.03 (see Tables 1 and B1). Ryder et al. (2013) reported optical properties measured inside the aircraft cabin and mie-simulated from the number particle size distribution in the range 0.1 – 930 μ m, and show that the direct measurements overestimate the SSA by up to 0.11 due to inlet efficiencies. Ryder et al. (2018) showed comparable outcomes, suggesting that the variability in the observed SSA values may be also due to discrepancies in the measured diameter size range of different studies.

The values reported by Denjean et al. (2020) deviate significantly from our measurements but still are within the variability range. Their intensive aerosol optical properties were derived from an optical closure study combining size distributions and optical properties measured for an aerosol particle passing through a PM₅ isokinetic inlet. This difference might be due also to the unidentified presence of pollution mixed with the mineral dust.

Differences in the observed intensive aerosol optical properties (g and SSA) of pure mineral dust layers may be due to variations in the measured particle diameter size range, distinct measurement or retrieval techniques, and different methods used to classify pure mineral dust layers.

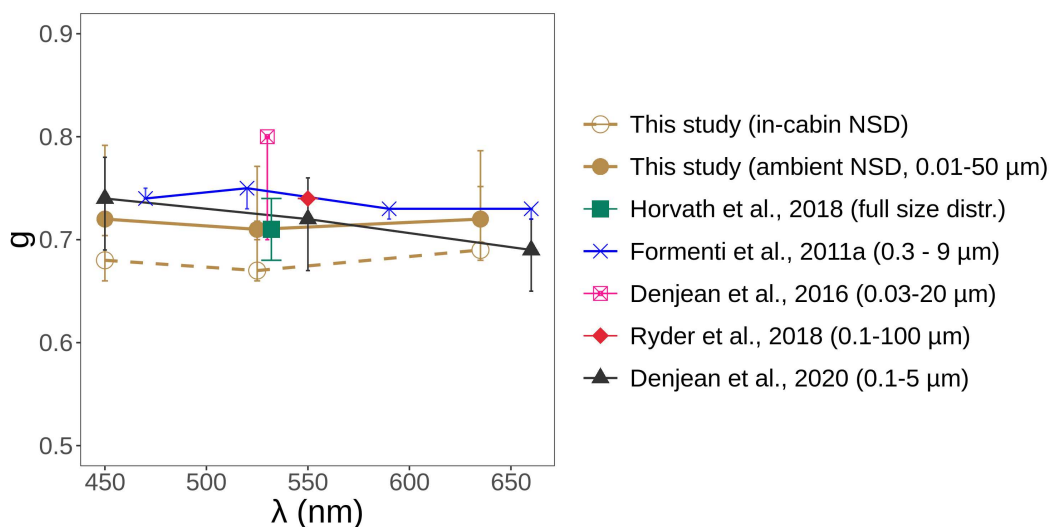


Figure 9. Spectral asymmetry parameter (g) for pure mineral dust layers. Data from this study are reported in brown. Empty circles indicate values measured in-cabin of the Falcon aircraft with the Aurora 4000 polar nephelometer, while full circles indicate values extended to the full-size distribution. Error bars indicate the variability obtained as the 3rd and 97th percentiles. For comparison, values issued from the AMMA (Formenti et al., 2011a), CHARMEX/ADRIED (Denjean et al., 2016), AER-D (Ryder et al., 2018), and DACCIWA (Denjean et al., 2020) field campaigns, as well as direct measurements performed with a polar nephelometer in Sierra Nevada (Spain) by Horvath et al. (2018) are shown. The measured diameter size range for each field experiment is given in the legend. Error bars correspond to the range of variability reported in the publications from which data are taken.

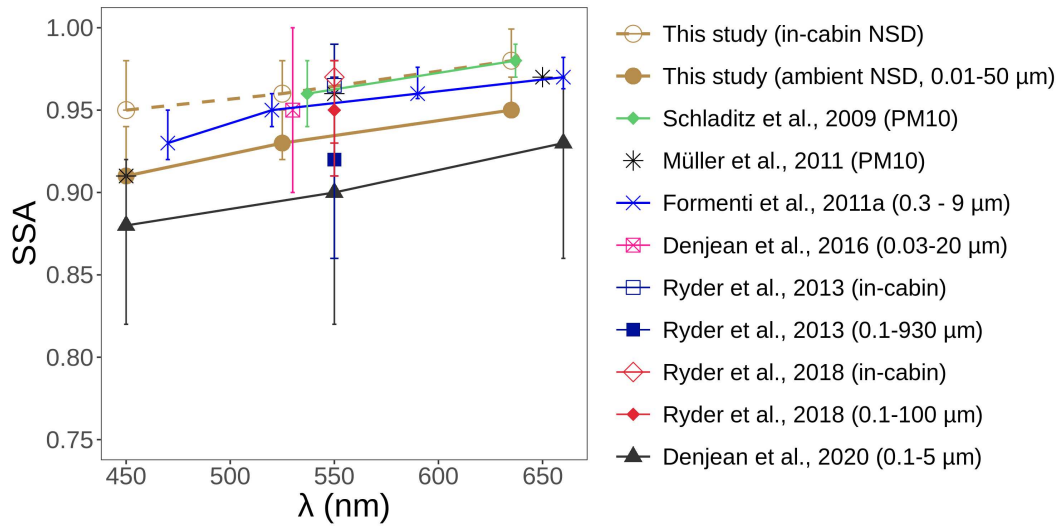


Figure 10. Spectral single scattering albedo (SSA) for pure mineral dust layers. Data from this study are reported in brown. Empty circles indicate values measured in-cabin of the falcon aircraft with the Aurora 4000 polar nephelometer and TAP, while full circles indicate values extended to the full-size distribution. Error bars indicate the variability obtained as the 3rd and 97th percentiles. For comparison, values issued from the SAMUM (Schladitz et al., 2009), SAMUM-2 (Müller et al., 2011), AMMA (Formenti et al., 2011a), CHARMEX/ADRIED (Denjean et al., 2016), Fennec 2011 (Ryder et al., 2013), AER-D (Ryder et al., 2018) and DACCIWA (Denjean et al., 2020) field campaigns are shown. The measured diameter size range for each field experiment is given in the legend. Error bars correspond to the range of variability reported in the publications from which data are taken.

4.4 Implications for the direct radiative effect

The asymmetry parameter (g) and the single scattering albedo (SSA) are two intensive aerosol optical properties essential for estimating the aerosol direct radiative effect. While the asymmetry parameter is affected by the particle size, the single
560 scattering albedo reflects the particle absorption's effect.

Our results show that both properties g and SSA decrease significantly for increasing pollution content within the mineral dust layer (Figure 7). g median values change from 0.67 to 0.56 for pure to polluted mineral dust at 525 nm, while SSA median values change from 0.96 to 0.89 (Table 1). Similar results were obtained, for example, by Seinfeld et al. (2004) in Asia and by Denjean et al. (2020) in West Africa. However, slightly different numbers can be due to the different diameter size ranges
565 measured and the different methods to identify the presence of pollution.

Since the changes in g and SSA are significant with increasing pollution within a mineral dust layer, the direct radiative effect efficiency may also be significantly affected. Indeed, a decrease in g indicates that less light is scattered in the forward direction, less energy reaches the Earth's surface, and thus the surface cooling effect may be enhanced compared to a pure mineral dust layer with the same aerosol optical depth (AOD). Conversely, a decrease in SSA indicates that more energy is
570 absorbed in the aerosol layer. Thus, the surface cooling effect may be reduced (or the atmospheric warming effect may be enhanced) compared to a pure mineral dust layer with the same AOD. These two opposing effects are due to the reduced particle size and the increased black carbon concentration associated with the increased pollution in the mineral dust layer. Indeed, the pollution includes not only only black carbon particles, but also fine mode anthropogenic particles (see Figure 3).

The simple formula, developed by Charlson et al. (1991) and Haywood and Shine (1995), can be used to evaluate the impact
575 of the mixing of pollution and mineral dust on the short-wave direct radiative effect efficiency (SW DREE) at the top of the atmosphere by investigating which of the two opposing effect (decrease in particle size vs. decrease in absorption) dominates in our measurements.

Several studies highlighted the limitation of this formula (e.g., Yu et al., 2006). First, the aerosol properties are assumed for a single wavelength considered representative of the visible range of the solar spectrum (i.e., 525 nm in our case). Second,
580 multiple scattering effects are neglected, and this formula is adequate only for thin atmospheres (Haywood and Shine, 1995). Third, the vertical profile of constituents is not considered. Fourth, the dependence on the solar zenith angle is not considered. Wendisch et al. (2001) showed that these simplifications caused large uncertainties, especially neglecting the multiple scattering. Hassan et al. (2015) compared this equation with the output of a more sophisticated model and found it erroneous for all-sky radiative effect but adequate for cloud-free conditions. Thus, despite the large uncertainties, the functional relationship
585 between the SW DREE, g and SSA remains valid. In conclusion, the Haywood and Shine (1995) formula can be used to compare the intrinsic forcing efficiency of different aerosols, as used for example by Sherman et al. (2015). Therefore, we focus not on the absolute number obtained with this formula but on its change rate for increasing pollution within the mineral dust layer. We believe that important conclusions might be drawn by considering the slopes of the relationship between the SW DREE and the pollution contribution in terms of mass in a mineral dust layer.

590 When considering a global average surface albedo, the pollution increase in a mineral dust layer does not seem to affect the short-wave direct radiative effect of the aerosol layer. Our results (Table 2) show that the SW DREE, calculated considering global average surface albedo and cloud fraction ($R_s = 0.15$ and $A_c = 0.60$), increases by $0.7(\pm 0.9) W m^{-2} AOD^{-1}$ per unit change of $\log_{10}(M_{rBC}/M_{super-\mu m \text{ ambient}})$. This change is not statistically significant given the wide confidence interval and the high p -value of 0.4. This result suggests that the direct radiative effect of a mineral dust layer with more pollution content does
 595 not change significantly compared to a pure mineral dust layer with the same AOD because the decreased size and increased absorption counteract each other.

However, the direct radiative effect of a mineral dust layer can strongly differ depending on the underlying surface albedo and also on the fraction of sky covered by clouds. The simple formula from Haywood and Shine (1995) (Eq. 6) allows to calculate the SW DREE taking into account variation of the surface albedo R_s and of the cloud fraction A_c . For the latter, only
 600 the case of clouds above the aerosol layer, i.e., no aerosol effect when the sky is covered by clouds, is considered.

We considered different surface albedo and cloud fraction values associated with different regions according to the Copernicus Climate Change Service (C3S), to assess the local change of the SW DREE due to the increase of pollution in a mineral dust layer. Figure 11 shows how the SW DREE changes for increasing pollution content into the mineral dust layer for different surface albedos ($R_s = 0.03, 0.15, 0.28, 0.68$ for ocean, global average, desert, and snow, respectively) in the case of cloud-free
 605 conditions $A_c = 0$. Figure 12 shows how the SW DREE changes for increasing pollution content into the mineral dust layer for different regions with a specific surface albedo and cloud fraction values.

When the mineral dust layer is above a surface with high surface albedo, as a desert or snow, the SW DREE increases for increasing pollution content. Thus, the cooling effect decreases (or the warming effect increases) for increasing pollution content compared to the case of a pure mineral dust layer. The results of the linear regression analysis between the SW DREE
 610 and the pollution contribution in terms of mass $\log_{10}(M_{rBC}/M_{super-\mu m \text{ ambient}})$ shows that the relationship between the two variables is statistically significant for cases with high surface albedo ($R_s \geq 0.23$), as indicated by the p -values below the threshold of 0.05.

When the mineral dust layer is above a surface with low surface albedo, as the ocean, the SW DREE decreases slightly for increasing pollution content. Thus, the cooling effect may increase for increasing pollution compared to the case of a pure
 615 mineral dust layer. However, the relationship is not statistically significant, as indicated by the p -values above the threshold of 0.05, and also by the wide confidence interval of the regression parameters. Thus, our data do not provide enough evidence to conclude that the SW DREE changes with increasing pollution content within the mineral dust layer. However, the relationship may become significant if more pollution is present in the mineral dust layer.

The presence of clouds does not modify the significance of the relationship, it changes the magnitude of the SW DREE
 620 and of the slope (Figure 12). Regions close to the Eastern Mediterranean that have high surface albedo are the Saharan desert and the Arabian Peninsula. These regions have a relatively low cloud fraction ($A_c = 0.21$), which leads to an increase by $+6(\pm 2) W m^{-2} AOD^{-1}$ of the SW DREE per unit increase of $\log_{10}(M_{rBC}/M_{super-\mu m \text{ ambient}})$. Turkey has several mountain chains which surface albedo present a strong annual cycle due to changes in vegetation and snow coverage. We considered in particular the Eastern part of Turkey of the Aras mountains. The annual average surface albedo of 0.23 and the cloud

fraction of 0.52 leads to absolute numbers of the SW DREE similar to the one of the Saharan desert and the Arabian Peninsula and slope $+3(\pm 1)Wm^{-2}AOD^{-1}$. During winter when the mountains are covered by snow the SW DREE increases by $+8(\pm 1)Wm^{-2}AOD^{-1}$ per unit increase of $\log_{10}(M_{rBC}/M_{super-\mu m \text{ ambient}})$. This slope is considerably smaller than the value of $+24(\pm 4)Wm^{-2}AOD^{-1}$ obtained without considering the effect of clouds (Figure 11). Regions with low surface albedo of interest for our measurements are the Mediterranean sea and the territory of Greece. The SW DREE in these regions changes by $-2(\pm 2)Wm^{-2}AOD^{-1}$ and $-1(\pm 1)Wm^{-2}AOD^{-1}$, respectively, per unit change of $\log_{10}(M_{rBC}/M_{super-\mu m \text{ ambient}})$.

These results suggest that for regions with a high surface albedo, quantifying the pollution within a mineral dust layer is very important to correctly estimate the local direct radiative effect of the mineral dust layer. Further studies are needed on the impact of mixing pollution and mineral dust, especially above surfaces with low surface albedo. In addition, the limitations and uncertainties of the SW DREE relationships do not allow further considerations. The generalization of these results requires calculations with a more sophisticated radiative transfer model, which is beyond the scope of this study..

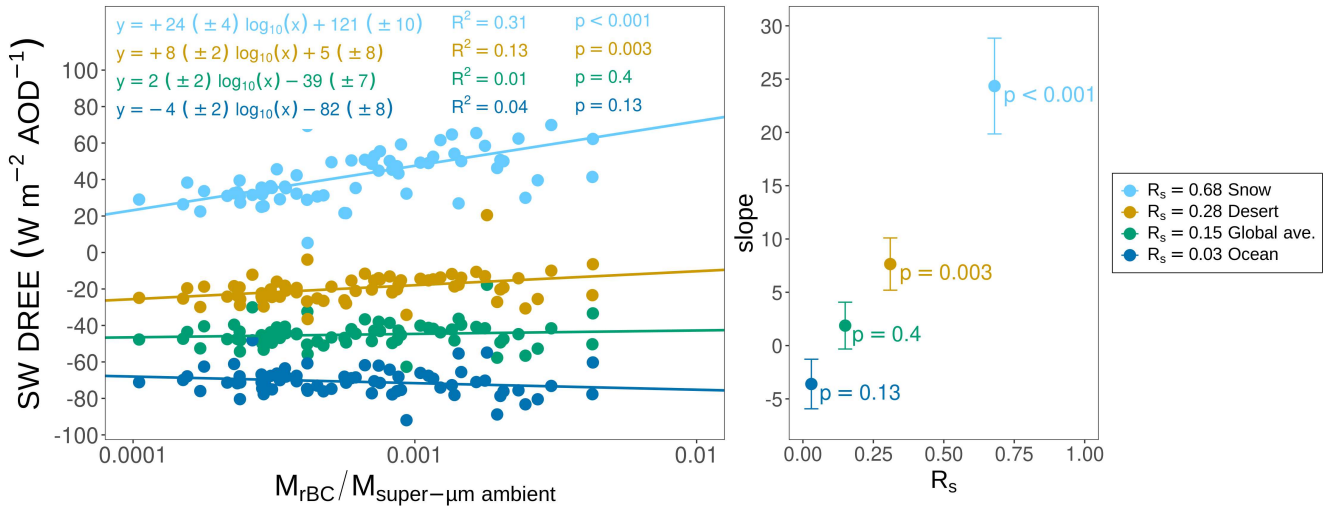


Figure 11. Change in the short-wave Direct Radiative Effect Efficiency (SW DREE) for increasing pollution in the mineral dust layer for different surface albedos in cloud-free conditions. The left panel shows the short-wave Direct Radiative effect Efficiency (SW DREE), calculated using a simplified formula (Haywood and Shine, 1995), the measured values of g and SSA at $\lambda = 525$ nm and global average parameters as in Sherman et al. (2015), as a function of the pollution contribution in terms of mass ($M_{rBC}/M_{super-\mu m \text{ ambient}}$). $M_{super-\mu m \text{ ambient}}$ is the mass of the ambient super- μm particles, since the values of g and SSA used to calculate the SW DREE were extended to the ambient particle size distribution (see section S3 in the supplementary information). Different colors indicate the SW DREE calculated for different surface albedos (R_s). The calculations are done for cloud-free conditions ($A_c = 0$). Respective least squares regression results, including regression line and equation, determination coefficient and p -value, are shown in the corresponding color. The right panel shows the slope obtained for each linear regression as a function of the surface albedo.

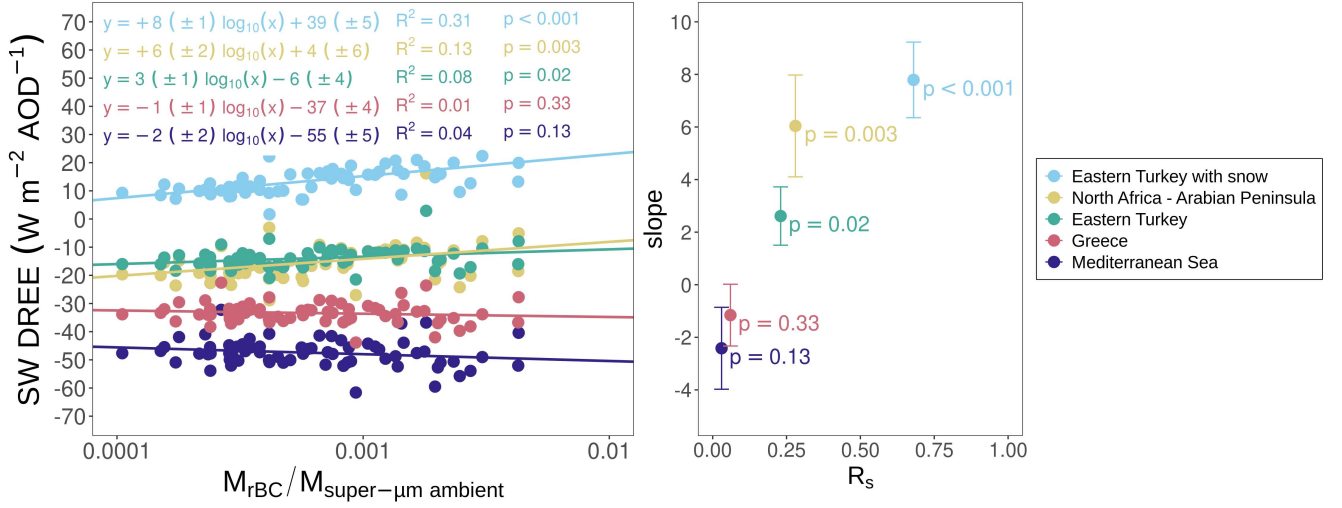


Figure 12. Change in the short-wave Direct Radiative Effect Efficiency (SW DREE) for increasing pollution in the mineral dust layer for different regions. The left panel shows the short-wave Direct Radiative Effect Efficiency (SW DREE), calculated using a simplified formula (Haywood and Shine, 1995), the measured values of g and SSA at $\lambda = 525$ nm and global average parameters as in Sherman et al. (2015), as a function of the pollution contribution in terms of mass ($M_{\text{rBC}}/M_{\text{super-}\mu\text{m ambient}}$). $M_{\text{super-}\mu\text{m ambient}}$ is the mass of the ambient super- μm particles, since the values of g and SSA used to calculate the SW DREE were extended to the ambient particle size distribution (see section S3 in the supplementary information). Different colors indicate the SW DREE calculated for different regions by considering different surface albedo (R_s) and cloud fraction (A_c) combinations. Annual average R_s and A_c were identified for each region using data from the Copernicus Climate Change Service (C3S): $R_s = 0.03$ and $A_c = 0.33$ for the Mediterranean sea, $R_s = 0.06$ and $A_c = 0.48$ for Greece, $R_s = 0.28$ and $A_c = 0.21$ for Saharan desert and Arabian Peninsula, $R_s = 0.23$ and $A_c = 0.52$ for Eastern Turkey and $R_s = 0.68$ and $A_c = 0.68$ for Eastern Turkey when covered by snow. Respective least squares regression results, including regression line and equation, determination coefficient and p -value, are shown in the corresponding color. The right panel shows the slope obtained for each linear regression as a function of the surface albedo.

5 Conclusions

During the A-LIFE aircraft field experiment in April 2017, we measured the properties of mineral dust and mineral dust-pollution mixtures in the Eastern Mediterranean. In this study, we analyzed the intensive aerosol optical properties directly measured during A-LIFE with an Aurora 4000 polar nephelometer and a Tricolor Absorption Photometer, such as the scattering, absorption and single scattering albedo Ångström exponent (SAE, AAE, and SSAAE), and the asymmetry parameter and the single scattering albedo (g and SSA). We investigated how these intensive aerosol optical properties change when mineral dust layers are mixed with anthropogenic aerosols/pollution in different quantities. In addition, we compared these results for mineral dust from the two source regions that affect the Eastern Mediterranean: the Arabian peninsula and the Saharan desert.

Our data show that all intensive aerosol optical properties (SAE, AAE, SSAAE, g , and SSA) change significantly with increasing pollution in the mineral dust layer, while the differences between the optical properties of mineral dust from the two different source regions are not significant. The values of the intensive aerosol optical properties of Saharan and Arabian dust are similar (within error bars) when comparing data with similar pollution content.

These findings have different implications (i) for the retrieval of mineral dust properties, (ii) for the identification of mineral dust episodes in the atmosphere, and (iii) for the direct radiative effect of mineral dust layers when mixed with pollution.

We highlight that care is needed when retrieving the optical properties of mineral dust because mineral dust layers often contain mixtures of other aerosol components. In particular, when analyzing the differences between mineral dust from different source regions, evaluating the pollution content and comparing only measurements with similar pollution content is essential.

The presence of pollution masks the mineral dust signal because the Ångström exponents (SAE, AAE, and SSAAE) are significantly affected by the presence of pollution. As a result, the identification of mineral dust cases using the Ångström exponents fails in case of mixtures with pollution particles. Once the presence of mineral dust has been established by independent methods, the Ångström exponents may be helpful in estimating the amount of pollution mixed into the mineral dust layer. Thus, the Ångström exponents can be used to select cases where non-dust aerosols have a minimal effect on the optical properties of mineral dust. Among the Ångström exponents (SAE, AAE, and SSAAE), SAE is the best parameter for estimating the amount of pollution mixed into a mineral dust layer.

Climate-relevant intensive aerosol optical properties (g and SSA) measured in pure mineral dust layers are in agreement with values reported by previous studies. However, differences in the values measured in the Falcon aircraft cabin and values extended to the ambient particle number size distribution point out that differences in the observed intensive aerosol optical properties in various studies may also be attributed to the discrepancies in the measured particle diameter size range.

The pollution in a mineral dust layer significantly impacts the climate-relevant intensive aerosol optical properties. The asymmetry parameter and the single scattering albedo (g and SSA) decrease significantly for increasing pollution content, (e.g., from pure mineral dust to polluted mineral dust median measured values decreased from 0.67 (0.63–0.72) to 0.56 (0.50–0.59) and from 0.96 (0.94–0.98) to 0.89 (0.84–0.94), respectively, at 525 nm). We would like to remind that an increase in pollution indicates not only an increase of black carbon particles, but also an increase of other fine mode anthropogenic aerosol particles, although we quantified the pollution using the refractory black carbon mass. Thus, the changes in g and SSA have two opposite

670 impacts on the short-wave direct radiative effect of the mineral dust layer. The decrease of g , due to a decrease in the particle size, leads to an increase in the cooling effect at the top of the atmosphere. The decrease of SSA, due to increased absorption, leads to a decrease in the cooling effect at the top of the atmosphere (or increase in the warming effect).

Based on a simple calculation of the short-wave direct radiative effect efficiency (SW DREE), these two effects are expected to counteract each other over a global average surface albedo. However, the SW DREE can have a significant change for
675 increasing pollution content if the calculation is done considering different surface albedos. For instance, if the mineral dust layer is above a surface with high surface albedo, such as desert or snow, the increase in pollution leads to a reduction of the cooling effect (or an increase of the warming effect). On the contrary, if the mineral dust layer is above a surface with low surface albedo, such as the ocean, an increase in pollution may result in an increase of the cooling effect. Thus, in this case, the effect of decreasing particle size is more significant than the effect of increased absorption. However, it is important to note
680 that this latter effect was not statistically significant in our data.

Further studies on the impact of mixing pollution and mineral dust are required to generalize these results. A more sophisticated radiative transfer model may be used to assess the impact of mixing pollution and mineral dust on the direct radiative effect efficiency. Additionally, particles in the mineral dust layer can be found in different mixing states. For example, mineral dust and rBC particles can be found in mixtures with sulfate, while internal mixtures between mineral dust and rBC seem
685 rare. Thus, future studies should also focus on investigating the impact of different mixing states of aerosol particles on the measured optical properties.

In conclusion, we would like to emphasize that an accurate quantification of the pollution content mixed in mineral dust layers is necessary because the presence of pollution has major consequences on the identification of mineral dust episodes and the retrieval of mineral dust optical properties and its direct radiative effect.

Table A1. Summary of optical aerosol properties used for the analysis of this study.

Symbol	Quantity	Size range	Method
$\sigma_{sp,\lambda}^{Aurora\ 4000,\alpha}$	Particle scattering coefficient	in-cabin*	Measured by the Ecotech Aurora 4000 polar nephelometer at three wavelengths ($\lambda = 450, 525, 635$ nm) and for four angular sectors ($\alpha = 180^\circ$, with $\alpha = 0, 20, 50, 90^\circ$).
$\sigma_{sp,\lambda}$	Total particle scattering coefficient	in-cabin*	Measured by the Ecotech Aurora 4000 polar nephelometer at three wavelengths ($\lambda = 450, 525, 635$ nm), for the angular sector $0 - 180^\circ$, and corrected for angular truncation and illumination as indicated in Teri et al. (2022).
$\sigma_{ap,\lambda}$	Particle absorption coefficient	in-cabin*	Measured by the Tricolor Absorption Photometer (TAP, Brechtel Manufacturing Inc., Hayward, USA) at three wavelengths ($\lambda = 465, 520, 640$ nm).
$SAE_{\lambda_B/\lambda_R}$	Scattering exponent	Ångström in-cabin*	$SAE_{\lambda_B/\lambda_R} = -\log(\sigma_{sp,\lambda_B}/\sigma_{sp,\lambda_R})/\log(\lambda_B/\lambda_R)$, with the blue and red wavelengths of the Aurora 4000 polar nephelometer ($\lambda_B = 450$ nm and $\lambda_R = 635$ nm).
$AAE_{\lambda_B/\lambda_R}$	Absorption exponent	Ångström in-cabin*	$AAE_{\lambda_B/\lambda_R} = -\log(\sigma_{ap,\lambda_B}/\sigma_{ap,\lambda_R})/\log(\lambda_B/\lambda_R)$, with the blue and red wavelengths of the TAP ($\lambda_B = 465$ nm and $\lambda_R = 640$ nm).
$SSAAE_{\lambda_B/\lambda_R}$	Single scattering albedo Ångström exponent	in-cabin*	$SSAAE_{\lambda_B/\lambda_R} = -\log(SSA_{\lambda_B}/SSA_{\lambda_R})/\log(\lambda_B/\lambda_R)$, where SSA is the single scattering albedo.
g_λ	Asymmetry parameter	in-cabin*	Calculated using the Müller et al. (2012) approximation using $\sigma_{sp,\lambda}^{Aurora\ 4000,\alpha}$.
SSA_λ	Single scattering albedo	in-cabin*	$SSA_\lambda = \sigma_{sp,\lambda}/(\sigma_{sp,\lambda} + \sigma_{ap,\lambda})$, where λ is the blue, green, or red wavelength of the Aurora 4000 polar nephelometer and of the TAP for $\sigma_{sp,\lambda}$ and $\sigma_{ap,\lambda}$, respectively.
$g_{\lambda,ambient}$	Asymmetry parameter of the ambient NSD	10 nm – 50 μ m	Obtained by extending to the ambient NSD the value of g_λ measured in the Falcon aircraft cabin (see section S3 in the supplementary information).
$SSA_{\lambda,ambient}$	Single scattering albedo of the ambient NSD	10 nm – 50 μ m	Obtained by extending to the ambient NSD the value of SSA measured in the Falcon aircraft cabin (see section S3 in the supplementary information).
SW DREE	Short-wave direct radiative effect efficiency	10 nm – 50 μ m	Calculated using a simplified formula derived by Haywood and Shine (1995): $SW\ DREE = SW\ DRE/AOD =$ $= -DS_0T_{atm}(1 - A_c)\beta SSA[(1 - R_s)^2 - (2R_s/SSA\beta)(1 - SSA)]$ where $D = 0.50$ is the fractional day length, $S_0 = 1370\ Wm^{-2}$ is the solar constant, $T_{atm} = 0.76$ is the atmospheric transmission, $A_c = 0.60$ is the cloud fraction, $R_s = 0.15$ is the spectrally averaged surface albedo, SSA is the single scattering albedo, and β is the average upscatter fraction, and can be obtained from g via a parametrization from (Wiscombe and Grams, 1976). SSA and g are the measured values at 525 nm of the ambient NSD.

*Size range of aerosol particles passing through the Falcon sampling system. The Falcon inlet and sampling system has a cut-off diameter (i.e., the diameter at which 50% of the particles pass the inlet and the sampling system) between 2.4 μ m and 6.4 μ m for flight altitudes between 2 and 8 km depending on the particle's density (see Table 2 and Figure 6 in Schöberl et al., 2024).

Table A2. Summary of microphysical aerosol properties used for the analysis of this study.

Symbol	Quantity	Size range	Method
M_{rBC}	Mass concentration of refractory black carbon	70 – 980 nm	Measured by a Single Particle Soot Photometer (SP2, DMT).
N_{coarse}	Number concentration of ambient coarse mode aerosol	0.5 – 50 μm	Measured by a optical particle counter, the wing-mounted Cloud and Aerosol Spectrometer (CAS) which is part of University of Vienna’s second-generation Cloud, Aerosol, and Precipitation spectrometer (UNIVIE CAPS, DMT)
Modeled aerosol mass concentration and composition	Modeled mass concentration of individual aerosol types along the flight path	100 nm – 10 μm	Obtained by combining source-receptor sensitivities calculated with the Lagrangian Particle dispersion model FLEXPART (Stohl et al., 1998; Seibert and Frank, 2004; Pisso et al., 2019) with emission data from the Copernicus Atmospheric Monitoring Service (CAMS; CAMS, 2019; Morcrette et al., 2009). For details, see section 2.3.4.
In-cabin NSD	Particle number size distribution representing the size range passing through the Falcon aerosol inlet and transport system	in-cabin*	Obtained by combining measurements of the in-cabin condensation particle counter (CPC, model TSI 3760a, TSI Inc.; in this study referred to as as TSI CPC), the wing-mounted active sampling Ultra-High Sensitivity Aerosol Spectrometer-Airborne (UHSAS-A, DMT), and the in-cabin optical particle counter (SkyOPC, model 1.129, GRIMM Aerosol Technik). Measured in-cabin NSDs were parameterized with a combination of three log-normal distributions. Refractive indices used for the derivation of geometrical particle size from OPC measurements are chosen according to the modeled aerosol composition.
Ambient NSD	Ambient particle number size distribution	10 nm – 50 μm	Obtained by combining measurements of the in-cabin condensation particle counter (TSI CPC), the wing-mounted UHSAS-A, the in-cabin SkyOPC, and the wing-mounted CAS component of the Vienna CAPS. Measured ambient NSDs were parameterized with a combination of three log-normal distributions. Refractive indices are chosen according to the modeled aerosol composition.
$M_{sub-\mu\text{m}}$	Mass concentration of sub- μm particles	100 nm – 1 μm	Calculated from the the in-cabin NSD, assuming particles with a spherical shape and density based on the modeled aerosol composition.
$M_{super-\mu\text{m in-cabin}}$	Mass concentration of super- μm particles passing through the Falcon aerosol inlet and transport system	$d > 1 \mu\text{m}$ in-cabin*	Calculated from the in-cabin NSD, assuming particles with a spherical shape and density based on the modeled aerosol composition.
$M_{super-\mu\text{m ambient}}$	Mass concentration of super- μm particles	1 – 50 μm	Calculated from the ambient NSD, assuming particles with a spherical shape and density based on the modeled aerosol composition.

*Size range of aerosol particles passing through the Falcon sampling system. The Falcon inlet and sampling system has a cut-off diameter (i.e., the diameter at which 50% of the particles pass the inlet and the sampling system) between 2.4 μm and 6.4 μm for flight altitudes between 2 and 8 km depending on the particle’s density (see Table 2 and Figure 6 in Schöberl et al., 2024).

Appendix B: Asymmetry parameter and single scattering albedo representative for the ambient NSD

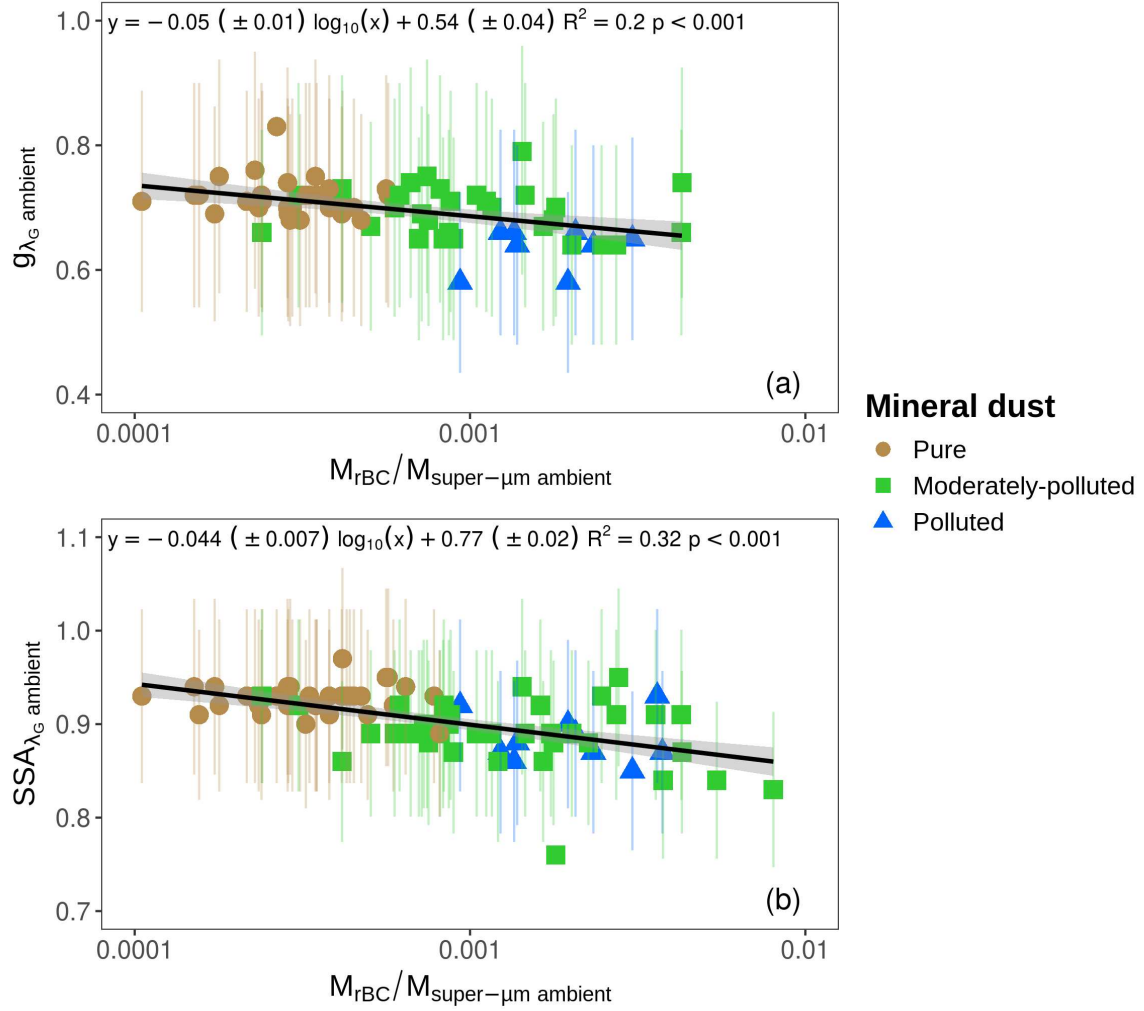


Figure B1. Intensive aerosol optical properties (g , and SSA) representative for the ambient NSD ($0.01 - 50 \mu m$). Each panel shows a different intensive optical property as a function of the pollution contribution in terms of mass ($M_{rBC}/M_{super-\mu m \text{ ambient}}$). Panel (a) shows the asymmetry parameter (g) at $\lambda = 525 \text{ nm}$, and panel (b) shows the single scattering albedo (SSA) at $\lambda = 525 \text{ nm}$. Least squares regression results, including regression line and equation, determination coefficient and p -value, are shown in black. The colors and shapes indicate the aerosol types following the A-LIFE aerosol classification scheme. The figure reports results for 87 sequences, among which 37, 40, and 10 are classified as pure, moderately-polluted, and polluted mineral dust, respectively. Results for g_{λ} were only available for 66 sequences, among which 29, 29, and 8 are classified as pure, moderately-polluted, and polluted mineral dust, respectively.

Table B1. Summary statistics for each aerosol type of $g_{\lambda,\text{ambient}}$ and $\text{SSA}_{\lambda,\text{ambient}}$ at three wavelengths ($\lambda_B = 450$ nm, $\lambda_G = 525$ nm, $\lambda_R = 635$ nm) representative for the ambient NSD ($0.01 - 50$ μm) measured out-cabin of the Falcon aircraft during A-LIFE for sequences classified as mineral dust by the A-LIFE aerosol classification scheme. The table reports results for 87 sequences, among which 37, 40, and 10 are classified as pure, moderately-polluted, and polluted mineral dust, respectively. Results for g_{λ} were only available for 66 sequences, among which 29, 29, and 8 are classified as pure, moderately-polluted, and polluted mineral dust, respectively.

		$g_{\lambda_B,\text{ambient}}$	$g_{\lambda_G,\text{ambient}}$	$g_{\lambda_R,\text{ambient}}$	$\text{SSA}_{\lambda_B,\text{ambient}}$	$\text{SSA}_{\lambda_G,\text{ambient}}$	$\text{SSA}_{\lambda_R,\text{ambient}}$
Pure mineral dust	median	0.72	0.71	0.72	0.91	0.93	0.95
	3^{rd}	0.67	0.68	0.69	0.89	0.90	0.92
	25^{th}	0.70	0.70	0.71	0.90	0.92	0.95
	75^{th}	0.74	0.72	0.76	0.92	0.93	0.96
	97^{th}	0.79	0.77	0.79	0.94	0.95	0.97
Moderately-polluted mineral dust	median	0.68	0.70	0.71	0.88	0.89	0.92
	3^{rd}	0.61	0.64	0.60	0.84	0.83	0.84
	25^{th}	0.66	0.66	0.68	0.87	0.88	0.90
	75^{th}	0.71	0.72	0.73	0.91	0.91	0.93
	97^{th}	0.74	0.76	0.75	0.93	0.94	0.96
Polluted mineral dust	median	0.56	0.56	0.54	0.89	0.89	0.88
	3^{rd}	0.49	0.50	0.50	0.86	0.84	0.85
	25^{th}	0.55	0.54	0.52	0.88	0.87	0.88
	75^{th}	0.57	0.57	0.56	0.90	0.89	0.90
	97^{th}	0.58	0.59	0.58	0.95	0.94	0.95

Data availability. The data used to obtain the results of this manuscript will be made publicly available in the University of Vienna data archive Phaidra (<https://doi.org/10.25365/phaidra.454>)

695 *Author contributions.* MT and BW conceptualized the study. MT performed the data analysis, and wrote the manuscript with the support of BW. BW coordinated the A-LIFE project. BW and MT performed the aerosol optical properties measurements during A-LIFE with the support of TM. MT performed the data quality assurance of the aerosol optical properties with the support of JG and BW. JG and MT calculated the simulated aerosol optical properties. BW, MD and KH performed the aerosol microphysical properties measurements during the A-LIFE field campaign. MD, MS, and JG calculated the combined particle number size distribution. KK and SA performed the single particle chemistry analysis of samples collected during A-LIFE. AT and PS performed the simulations with the Lagrangian particle dispersion model FLEXPART. All co-authors participated in the scientific discussion and reviewed the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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710 *Disclaimer.* The views expressed in this study are those of the authors and do not necessarily represent the views of the CTBTO Preparatory Commission

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S1 Data analysis and quality control of the Aurora 4000 polar nephelometer

The particle scattering coefficients were measured at three wavelengths ($\lambda_B = 450$ nm, $\lambda_G = 525$ nm, $\lambda_R = 635$ nm) using an Aurora 4000 polar nephelometer placed in the Falcon aircraft cabin behind the aerosol inlet system (in-cabin). The Aurora 4000 polar nephelometer has the unique feature of measuring the particle scattering coefficients for various angular sectors (ranging from the shutter position α to 180°), including the total particle scattering coefficient (measured when $\alpha = 0^\circ$).

During the A-LIFE field experiment, the Aurora 4000 polar nephelometer was mounted behind an isokinetic aerosol inlet (Schöberl et al., 2024) and measured the particle scattering coefficients for four angular sectors, with $\alpha = 0^\circ, 20^\circ, 50^\circ, 90^\circ$. This configuration enabled a temporal resolution of 6 – 15 s. A calibration with particle-free air and CO₂ was performed at the beginning of the measurement campaign and it was checked several times during and after the field experiment. The data obtained from the Aurora 4000 polar nephelometer was corrected using the average of all calibration checks, following the procedure recommended by Anderson and Ogren (1998). Additionally, multiple particle-free air measurements were conducted before and during the measurement flights to verify and correct for any potential background drift resulting from alteration in conditions during the measurements or of the carrier gas. The combined uncertainty due to instrumental noise, calibration and background drift uncertainties for 1-minute data is $\sim 3\%$. The Rayleigh scattering coefficients were subtracted to obtain the Aurora 4000 particle scattering coefficients ($\sigma_{sp,\lambda}^{Aurora\ 4000, \alpha}$). All the particle scattering coefficients were converted to standard temperature and pressure (273.15 K, 1013.25 hPa).

The total particle scattering coefficients ($\sigma_{sp,\lambda}$) were obtained by correcting $\sigma_{sp,\lambda}^{Aurora\ 4000, 0^\circ}$ for angular truncation and illumination as indicated in Teri et al. (2022), i.e., using the Anderson and Ogren (1998) method deploying the Scattering Ångström exponent (C_{SAE}) for cases with $SAE < 2$, and the Müller et al. (2012) correction C_{polar} for cases with $SAE > 2$. The assumed uncertainties for the angular correction are 13% and 2%, respectively. A median average was performed along selected flight sequences. For flight sequences data used in this work, the uncertainties of $\sigma_{sp,\lambda}$ including instrumental noise, calibration and background drift uncertainties, uncertainties of the correction to standard temperature and pressure, and the truncation error uncertainty, are within 13 – 30%.

S2 Data analysis and quality control of the Tri-color absorption photometer (TAP)

The particle absorption coefficient was measured using a tri-color absorption photometer (TAP, Brechtel) installed in the Falcon aircraft cabin behind an isokinetic aerosol inlet (Schöberl et al., 2024) inside the aircraft cabin. Measurements were taken at three different wavelengths ($\lambda_B = 465$ nm, $\lambda_G = 520$ nm, $\lambda_R = 640$ nm). The well characterized and widely used glass fiber Pallflex E70-2075W filters with a diameter of 47 mm were used (e.g., Virkkula, 2010; Davies et al., 2019). The filter was changed before each flight and the white filter check was performed according to the manufacturer indication. The light transmission through the filter decreases with the sampling time from about 1 to 0.85. Any deviation from a steady decrease in the filter transmission can be attributed to variation in inlet pressure, which modifies the filter optical properties during ascent and descent of the aircraft, as noted by Petzold et al. (2011). To avoid such artifact, we analyzed the TAP data exclusively for sequences at a constant altitude.

The spot area was determined to be 43.9 ± 2.3 mm² by measuring the diameter of each of the 10 spots with a caliber. The TAP sample flow was controlled by a mass flow meter (MFM, Alicat scientific) downstream of the measurement cell and measured by an internal flow meter. The sample flow was calibrated with a bubble flow meter (Gilibator 2, Gilian) by varying the pressure in the range 200 – 1000 hPa. The obtained linear least square regression parameters (Slope = 0.92, Offset = -0.03, $R^2 > 0.999$) were used to correct the measured sample flow. The calculated actual volume flow rate was 1.6 ± 0.2 L/min. Data were converted to standard temperature and pressure (273.15 K; 1013.25 hPa). Data sequences with the particle absorption coefficient below 0.6 Mm⁻¹ at least at one wavelength at ambient conditions were considered below the detection limit and excluded from the analysis. This decision follows the guidelines of the TAP user manual and Ogren et al. (2017).

The particle absorption coefficient was corrected for filter loading and scattering effects with the correction scheme developed by Virkkula et al. (2005) and modified by Virkkula (2010), using the particle scattering coefficient corrected for angular truncation and illumination error. This correction scheme was developed for the Particle Soot Absorption Photometer (PSAP, Radiance Research) and its performance was tested for the TAP by Davies et al. (2019). Their study calculated biases in the

particle absorption coefficient measured by the TAP for urban aerosol of 38 %, 37 %, and 27 % for the blue, green, and red wavelengths, respectively. We assumed an uncertainty of 40 % for all three wavelengths, considering that the wavelength of our TAP are slightly different from the TAP used by Davies et al. (2019).

We conducted additional data quality checks by comparing the particle absorption coefficient with the mass of refractory black carbon (M_{rBC}) measured by a Single Particle Soot Photometer (SP2). A correlation is expected between these two quantities since black carbon is the most important aerosol absorption component (e.g., Bond et al., 2013). We found a strong correlation with $R^2 > 0.92$ at all three wavelengths when excluding data points collected at pressure conditions lower than 400 hPa (see Figure S1). Data points collected at lower pressure appeared noisier and were excluded from our data. The high correlation between the particle absorption coefficient and the mass of refractory black carbon provided further evidence of the quality of the TAP data.

Additionally, we calculated the mass absorption coefficients (MAC_{corr}) from the correlation of the two quantities. We obtained values of $19.4 \text{ m}^2 \text{ g}^{-1}$, $15.9 \text{ m}^2 \text{ g}^{-1}$, and $10 \text{ m}^2 \text{ g}^{-1}$ for the wavelengths blue, green, and red respectively. These values are of the order of magnitude expected in literature for black carbon. For example, Ohata et al. (2021) reported values in the range of $10.8 - 15.1 \text{ m}^2 \text{ g}^{-1}$ for the MAC_{corr} measured in the Arctic at the green wavelength ($\lambda = 550 \text{ nm}$). Our values are slightly higher because the particle absorption coefficient measured by the TAP instrument includes the contribution of mineral dust, which is also expected to absorb in the blue and green wavelengths.

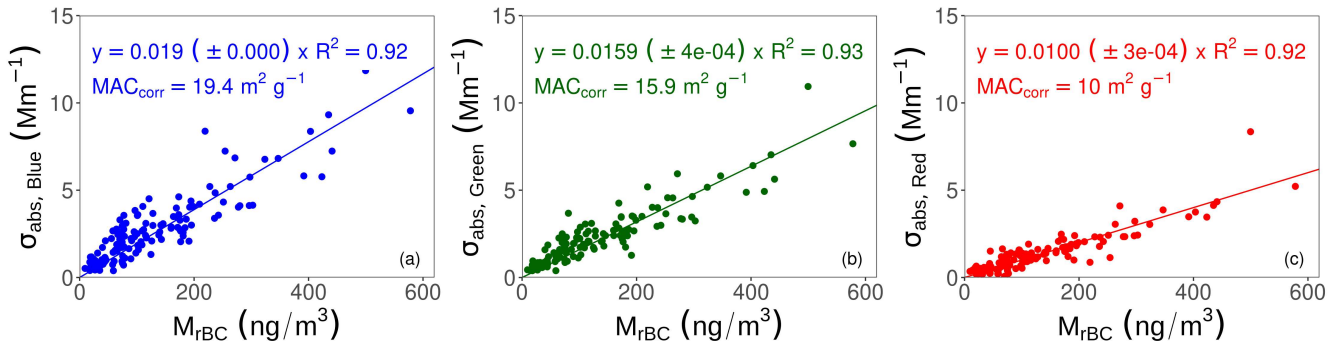


Figure S1. TAP data quality. The three panels (a,b, and c) show the comparison between the particle absorption coefficients ($\sigma_{\text{sap}, \lambda}$) at $\lambda = 465, 520$, and 640 nm , respectively, and the mass concentration of refractory black carbon (M_{rBC}). Results from linear regression analysis and corresponding mass absorption coefficient (MAC) values are reported on each figure.

S3 Extension of g_λ and SSA_λ to the ambient number size distribution

The asymmetry parameter (g) and the single scattering albedo (SSA) were calculated from the particle scattering and absorption coefficients measured in the aircraft cabin behind an aerosol sampling system (see section 2.3.2 of the main text). Thus, the obtained g and SSA values are representative of the measured aerosol particles in the size range passing through the Falcon aerosol inlet and transport system, and thus, for the in-cabin number size distributions (NSDs). We calculated the values of g and SSA representative of the ambient NSDs ($g_{\lambda, \text{ambient}}$ and $SSA_{\lambda, \text{ambient}}$) by multiplying the measured g and SSA values ($g_{\lambda, \text{in-cabin}}$ and $SSA_{\lambda, \text{in-cabin}}$) with a factor.

To calculate the correction factor to extend the measured optical properties to the ambient NSD, we performed optical simulations using the in-cabin and ambient NSDs as input of the MOPSMAP program (Gasteiger and Wiegner, 2018). The ambient NSDs were used in ambient relative humidity, while the in-cabin NSDs were used in dry conditions. The simulated optical properties were derived at three wavelengths ($\lambda_B = 450$ nm, $\lambda_G = 525$ nm, $\lambda_R = 635$ nm) considering the modeled aerosol composition (see section 2.3.5 in the main text). While mineral dust particles were assumed to be spheroids with an aspect ratio distribution according to Kandler et al. (2009), the other particle types were assumed to be spherical. The dust refractive index was taken from OPAC (Hess et al., 1998) in combination with a non-absorbing fraction of 0.5 (Gasteiger and Wiegner, 2018). Refractive indices from Li (1976) for sea salt, from Moise et al. (2015) for organic matter, and from Hess et al. (1998) for soot were considered, while a value of $1.53 + 0.00i$ was assumed for sulfate (e.g., Toon et al., 1976).

Figure S2 shows the comparison between the simulated and directly measured in-cabin intensive optical properties $g_{\lambda, \text{in-cabin}}$ and $SSA_{\lambda, \text{in-cabin}}$ at wavelengths 450, 525, and 635 nm. The directly measured intensive optical properties are reproduced by this approach within $\sim 16\%$ for $g_{\lambda, \text{in-cabin}}$ and within $\sim 10\%$ for $SSA_{\lambda, \text{in-cabin}}$. This comparison provides evidence that the approach used to extend the in-situ measured optical properties to ambient conditions is sufficiently accurate. A deeper discussion of the discrepancies between model simulations and measured optical properties is out of the scope of this paper.

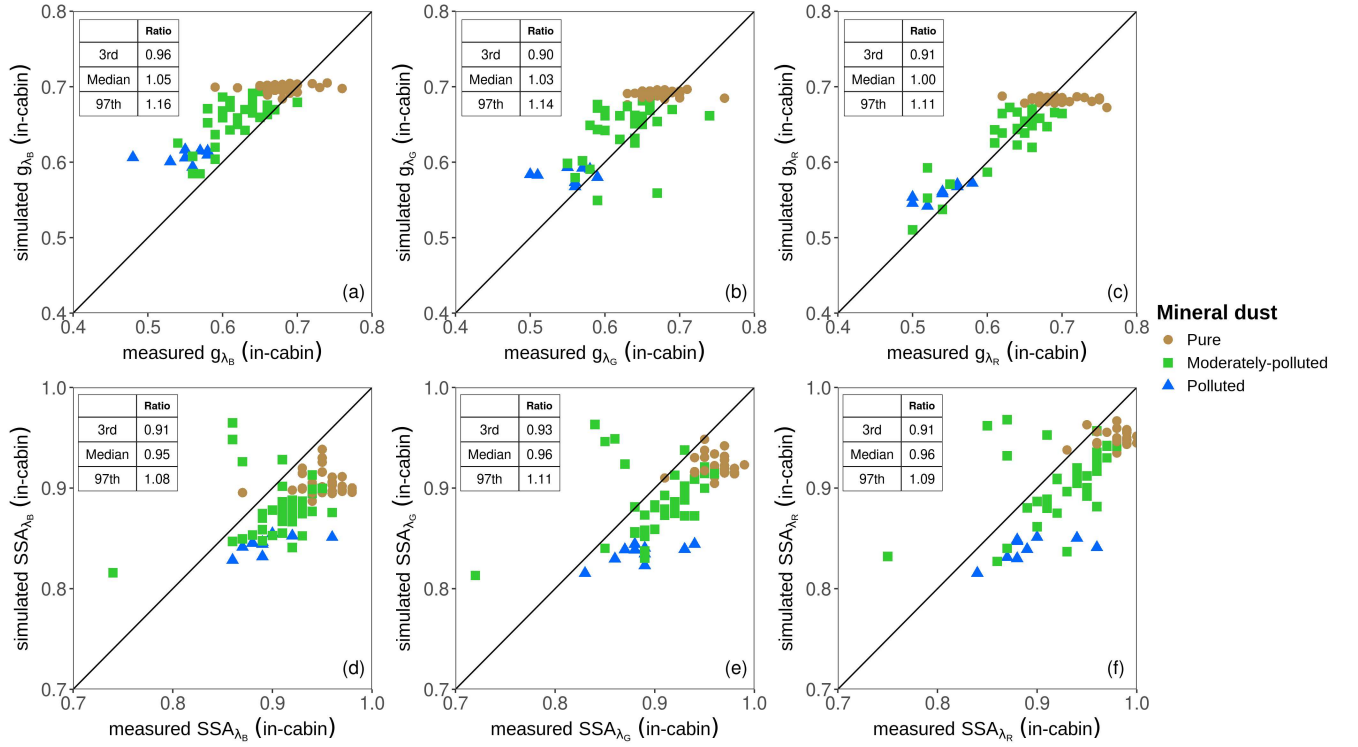


Figure S2. Comparison between the simulated intensive optical properties representative for the in-cabin NSD, and the intensive optical properties measured in the cabin of the Falcon research aircraft behind an inlet and a sampling system (in-cabin). Panels (a,b, and c) show the comparison for the simulated $g_{\lambda, \text{in-cabin}}$ and the measured $g_{\lambda, \text{in-cabin}}$ at wavelengths 450, 525, and 635 nm, respectively. Panels (d, e, and f) show the comparison for simulated $SSA_{\lambda, \text{in-cabin}}$ and the simulated $SSA_{\lambda, \text{in-cabin}}$ at wavelengths 450, 525, and 635 nm, respectively. The colors and shapes indicate the aerosol types following the A-LIFE aerosol classification scheme. The one to one line is shown in black. The median, 3rd, and 97th percentiles of the ratios between the simulated and measured values are reported in the upper left corner of the plots.

The correction factor to extend the intensive optical properties to the ambient NSD ($0.01 - 50 \mu\text{m}$) was calculated as the ratio of the simulated property for the ambient NSD and the simulated property for the in-cabin NSD:

$$85 \quad g_{\lambda,\text{ambient}} = g_{\lambda,\text{in-cabin}} \cdot \frac{\text{simulated } g_{\lambda,\text{ambient}}}{\text{simulated } g_{\lambda,\text{in-cabin}}} \quad (1)$$

$$SSA_{\lambda,\text{ambient}} = SSA_{\lambda,\text{in-cabin}} \cdot \frac{\text{simulated } SSA_{\lambda,\text{ambient}}}{\text{simulated } SSA_{\lambda,\text{in-cabin}}} \quad (2)$$

Figure S3 shows the comparisons between the simulated optical properties representative of the ambient NSDs and the simulated optical properties representative of the in-cabin NSDs. For each sequence, the deviation from the one-to-one line represents the correction factor applied to extend the intensive optical properties to the ambient NSD.

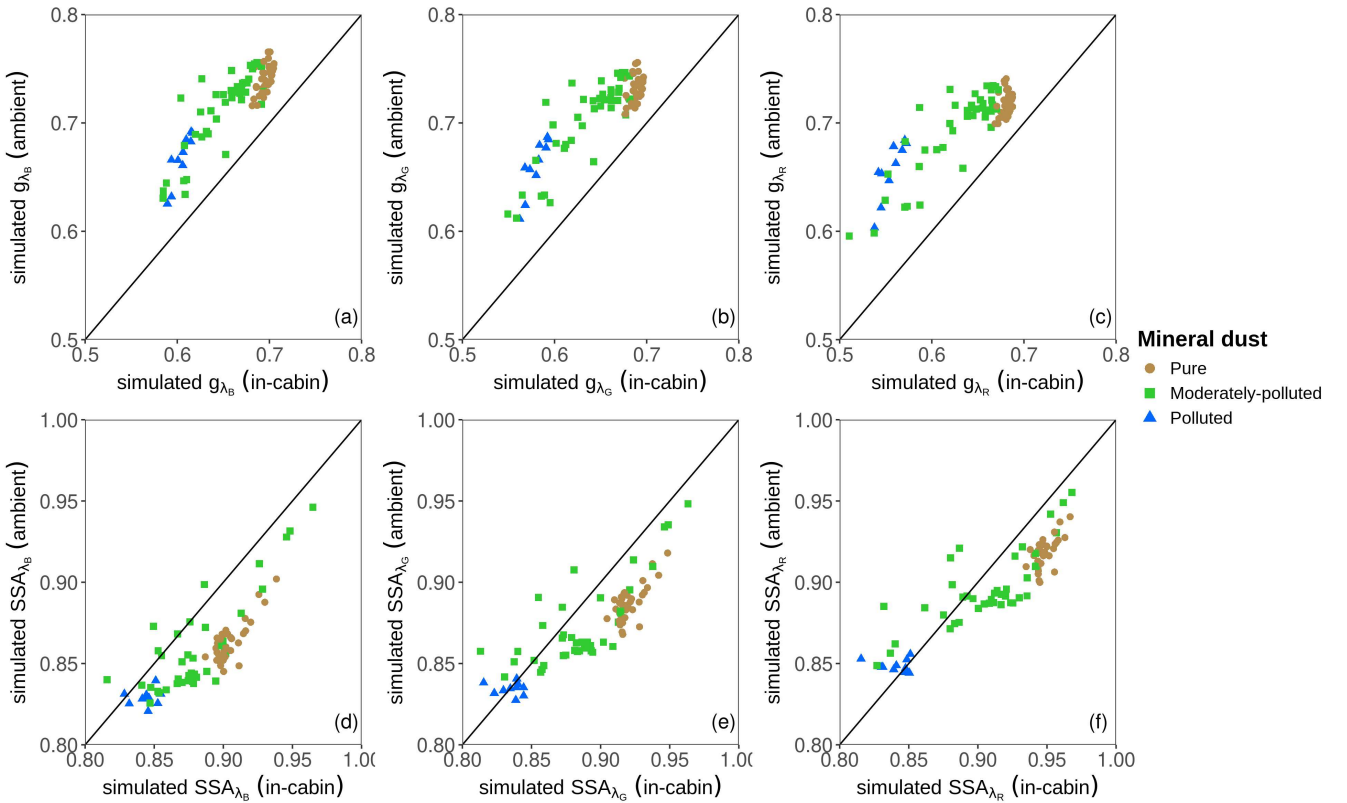


Figure S3. Simulated optical properties representative of the ambient NSD in comparison with the simulated optical properties representative of the in-cabin NSD. Panels (a,b, and c) show the comparison for the simulated $g_{\lambda,\text{ambient}}$ and the simulated $g_{\lambda,\text{in-cabin}}$ at wavelengths 450, 525, and 635 nm, respectively. Panels (d, e, and f) show the comparison for simulated $SSA_{\lambda,\text{ambient}}$ and the simulated $SSA_{\lambda,\text{in-cabin}}$ at wavelengths 450, 525, and 635 nm, respectively. The colors and shapes indicate the aerosol types following the A-LIFE aerosol classification scheme. The one to one line is shown in black.

90 S4 Single particle chemistry analysis of mineral dust particles collected during the A-LIFE field experiment

To support the interpretation of our results, we also used data from the single particle chemistry analysis of in-situ samples collected during the A-LIFE field experiment. A Micro Inertial Impactor (MINI) sampler collecting particles with diameters smaller than 1 μm and larger than 1 μm (Kandler et al., 2007) was placed in the Falcon aircraft cabin behind the aerosol inlet system (Schöberl et al., 2024). Particles were collected on Transmission Electron Microscope (TEM) grids and boron
95 substrates, with sampling time varying from three to ten minutes for each sample. Subsequently, the characteristics of single particles were studied under high vacuum conditions by an automated analysis in a Environmental Scanning Electron Microscope (FEI ESEM Quanta 400 FEG instrument, Eindhoven, The Netherlands) coupled with an X-Max 150 energy-dispersive X-ray spectroscopy (EDX) silicon drift X-ray detector (Oxford). Thus, the morphology and elemental composition of each particle (with projected area diameter from 0.5 to 3.0 μm) consisting of major elements F, Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti,
100 V, Cr, Mn, Fe, Zn and Pb is derived from EDX and given as normalized atomic percentages. The major dust components which are commonly found as ‘internal mixtures’ or aggregates were classified into dust particle-types by keeping each particles Al/Si ratios as the main constraint (Kandler et al., 2007). More details on the classification of the particle types used in this work are published and can be found as supplement data in (Panta et al., 2023).

The mineral dust optical properties for each sample are calculated using an empirical approach derived from Di Biagio et al.
105 (2019) and used by Kandler et al. (2020) and detailed in Aryasree et al. (2024) for particles from various mineral dust sources. The empirical relation links the total iron (Fe) present in mineral dust particles as oxides with the optical properties of mineral dust particles, such as the imaginary part of the refractive index (k) and the single scattering albedo (SSA).

S4.1 Mineral dust composition from single particle analysis

Data from the single particle chemistry analysis are used to support the interpretation of results of this work. Figure S4 shows
110 the percentage abundances of seven particle-type classes for Arabian and Saharan dust samples. The 7 particle-type classes were obtained from about 8000 particles measured under the scanning electron microscope. The analyzed samples were collected during flight sequences classified as pure and moderately-polluted Arabian and Saharan dust according to the A-LIFE aerosol classification (see section 2.3.4 in the main text). Additionally, Figure S5 shows different element ratios for particles in Arabian and Saharan dust sequences. Arabian and Saharan dust samples show a similar abundance with a higher percentage of Si-rich
115 particles or low Al/Si ratio particles named silicate-like particles hereafter. Mineral dust particles from both source regions have 60 – 62% of these silicate-like particles. Additionally, we observed a negligible variance in the Illite to Kaolinite (I/K) ratio, which is often used, in combination with ratios of other clay minerals-like particles, as a fingerprint to the active source regions Scheuven et al. (2013). The most significant difference observed in the mineralogy composition is a higher abundance of clay-like minerals (Illite and kaolinite) from the Sahara (21% against 8% for Arabian dust). While carbonate (e.g., Calcite,
120 Dolomite etc.) content is higher in Arabian dust (18%), a lower percentage is measured in Saharan dust (6%). This result is in agreement with several previous studies conducted in Saharan near-source regions, as well as in regions where mineral dust is transported, which characterize Saharan dust particles as having a higher clay mineral content (Kandler et al., 2009; Formenti et al., 2011). However, there are only limited studies in the Middle East and nearby Arabian dust sources to characterize the mineralogical characteristics of the Arabian dust (Attiya and Jones, 2020; Engelbrecht et al., 2009, 2016). All these studies
125 show high variability in mineral dust and soil mineralogy within the Arabian Peninsula, making it difficult to find an exact dust mineralogical characteristic. However, compared to Saharan dust, a high carbonate and low clay content is common in these studies. The percentage of iron oxides and hydroxides in Arabian dust is half that of the Saharan (3%, see Figure S4). To obtain a more accurate iron content description, we calculated iron’s total oxide percentage for each mineral dust particle using Table 3 values from Di Biagio et al. (2019) and the Fe mass of the particle. A summary of this method is available in Kandler et al.
130 (2020). For statistical certainty bootstrapping was done with 10,000 particles for the dust sequences. The result shows a $\sim 10\%$ increase in the total iron oxide weight in Saharan dust (median: 8.65 at a 95% confidence interval of 8.2 – 8.9) as compared to the Arabian dust median value of 7.8 (95% confidence interval of 7.6 – 8.1).

Since the mineral dust optical properties depend on the total iron content, this result indicates that the Saharan dust particles were more absorbing than the Arabian dust particles over the eastern Mediterranean. Indeed, the imaginary part of the refractive
135 index (k) has a value of 0.006 for Saharan dust and 0.005 for Arabian dust (see Figure S6). The SSA of mineral dust particles

showed a 4% increase for Arabian dust compared to Saharan dust. The imaginary refractive index values of mineral dust particles are higher than the characteristics of Saharan dust as seen from other locations over Northwest Sahara or the mineral dust outflow regions in the Atlantic (e.g., 0.001 – 0.004 at 520 nm Cape Verde) (Kandler et al., 2007, 2011; Haywood et al., 2008).

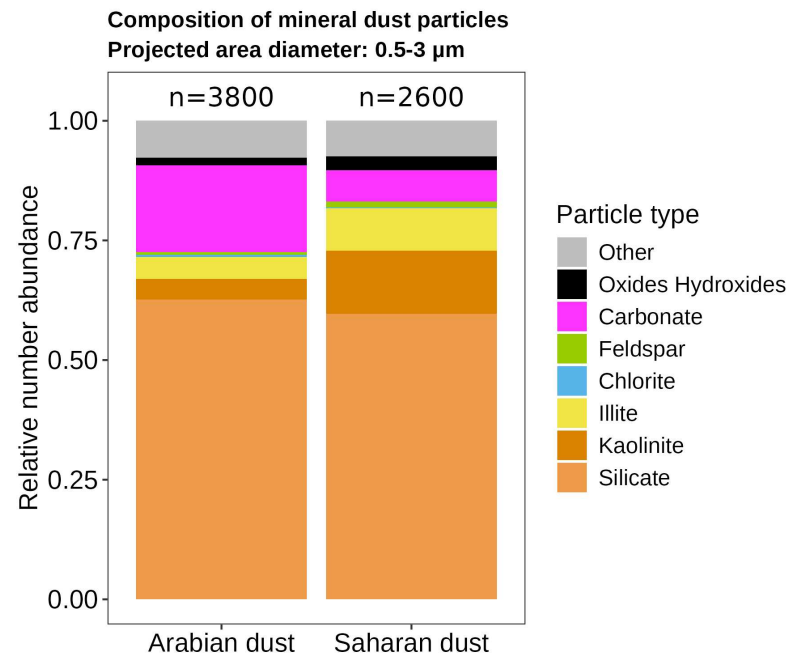


Figure S4. Composition of mineral dust particles with projected area diameter ranging from 0.5 to 3 μm . The graphs show the relative abundance of 7 dust particle types, as observed under the scanning electron microscope, for samples collected during sequences classified as Arabian and Saharan dust according to the A-LIFE aerosol classification. The pollution degree is not considered as only mineral dust particles are observable in this size range. The number of particles (n) analyzed is above the corresponding bar.

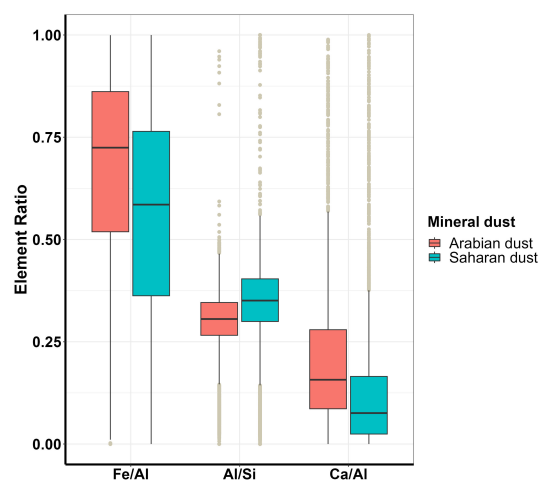


Figure S5. Element ratios (Fe/Al, Al/Si, Ca/Al) for Saharan and Arabian dust particles. The graph shows three elemental ratios obtained from about 8000 particles measured under the scanning electron microscope. The analyzed samples were collected during flight sequences classified as pure and moderately polluted Arabian dust and Saharan dust according to the A-LIFE aerosol classification scheme. The boxes represent the interquartile range (IQR 25th – 75th), the horizontal lines the median, the whiskers the largest value that is not greater than $1.5 \cdot IQR$, and data points outside this range are shown as individual dots.

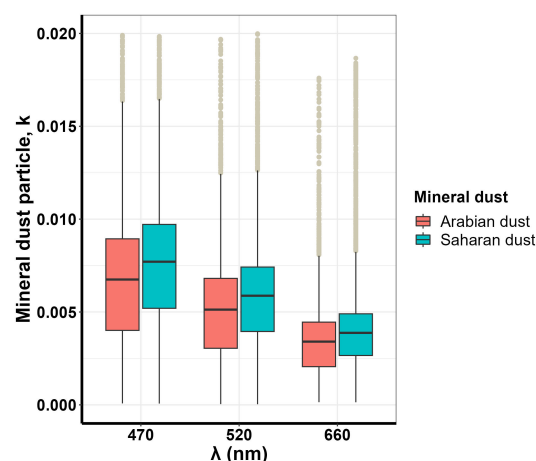


Figure S6. Spectral imaginary dust refractive index for Arabian and Saharan dust. The graph shows the median values of the imaginary part of the refractive index at three wavelengths obtained from single particle chemistry analysis. The analyzed samples were collected during flight sequences classified as pure and moderately polluted Arabian dust and Saharan dust according to the A-LIFE aerosol classification scheme. The boxes represent the interquartile range (IQR 25th – 75th), the horizontal lines the median, the whiskers the largest value that is not greater than $1.5 \cdot IQR$, and data points outside this range are shown as individual dots.

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