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Improving Global Xenon-133 Background Concentration Fields
By Applying Nudging in FLEXPART-LCM

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Abstract

The atmospheric ^{133}Xe background, characterised by spatiotemporally variable concentrations predominantly driven by routine civilian emissions from medical isotope production facilities, constitutes the critical baseline upon which eventual signals from non-civilian sources are superimposed. This is central to the radionuclide monitoring under the Comprehensive Nuclear-Test-Ban Treaty, enabling the detection and interpretation of signals from nuclear explosions. Although atmospheric transport models are, in principle, capable of reproducing ^{133}Xe background concentration fields, uncertain and incomplete emission inventories substantially limit their fidelity.

To address this limitation, spatially and temporally adaptive nudging was implemented in FLEXPART-LCM. The experimental design systematically varied horizontal kernel scales to balance physical plausibility against statistical performance and included two persistence-oriented sensitivity configurations. The first increased horizontal and vertical kernel scales to quantify downwind propagation and validation skill at independent stations, while the second employed a synthetic tracer to examine whether the observed limited impact was primarily attributable to decay rather than to insufficient nudging.

Averaged across assimilation stations, nudging reduced bias by 77% and mean squared error by 88%, and increased the coefficient of determination from 0.627 to 0.834. These improvements are primarily diagnostic of the implementation and are consistent with expectations for direct observational relaxation. By contrast, at independent validation stations, the changes were at most marginal and, on average, slightly negative, indicative of spurious corrections in regions lacking dynamical connectivity to assimilation stations. Increasing the kernel scales in three dimensions did not materially alter this outcome and slightly worsened performance at validation stations. As anticipated, the synthetic tracer expanded the domain of influence by approximately 104%, suggesting more favourable conditions for longer-lived xenon isotopes such as $^{131\text{m}}\text{Xe}$.

Given the present station geometry, nudging thus functions mainly as a local correction mechanism. Its broader impact is constrained by kernel scale, flow coherence, and the short half-life of ^{133}Xe , which limits the range of influence of nudging increments. Consequently, improvements in background concentration fields remain largely confined to regions governed by persistent wind patterns. Extending predictive skill beyond assimilation stations will require an increased fraction of reliably quantifiable observations and a denser, more strategically distributed monitoring network with enhanced coverage near source regions to enable the dynamically consistent propagation of corrective information.

Kurzfassung

Das atmosphärische ^{133}Xe -Hintergrundfeld, geprägt durch räumlich und zeitlich variierende Konzentrationen, die überwiegend aus routinemäßigen zivilen Emissionen von Anlagen zur Produktion medizinischer Isotope stammen, bildet die zentrale Referenz, auf der sich mögliche Signale nicht-ziviler Quellen überlagern. Dies ist für das radio-nuklidbasierte Monitoring im Rahmen des Comprehensive Nuclear-Test-Ban Treaty von zentraler Bedeutung, da es die Detektion und Interpretation von Signalen nuklearer Explosionen ermöglicht. Obwohl atmosphärische Transportmodelle prinzipiell in der Lage sind, ^{133}Xe -Hintergrundkonzentrationen zu reproduzieren, wird ihre Aussagekraft durch unsichere und unvollständige Emissionsinventare deutlich eingeschränkt.

Um die Einschränkung zu adressieren, wurde in FLEXPART-LCM ein räumlich und zeitlich adaptives Nudging implementiert. Das Versuchsdesign variierte systematisch horizontale Kernelskalen, um physikalische Plausibilität gegen statistische Güte abzuwägen, und umfasste zwei auf Persistenz ausgerichtete Sensitivitätskonfigurationen. Die erste erhöhte die horizontalen und vertikalen Kernelskalen, um die Ausbreitung und die Validierungsgüte an unabhängigen Stationen zu quantifizieren. Die zweite setzte einen synthetischen Tracer ein, um zu prüfen, ob die beobachtete begrenzte Wirkung primär durch Zerfall und nicht durch unzureichendes Nudging bedingt ist.

Im Mittel über die Assimilationsstationen verringerte das Nudging den Bias um 77 %, die mittlere quadratische Abweichung um 88 % und erhöhte das Bestimmtheitsmaß von 0,627 auf 0,834. Diese Verbesserungen sind vor allem diagnostisch für die Implementierung und entsprechen den Erwartungen an eine direkte Beobachtungsrelaxation. An unabhängigen Validierungsstationen waren die Änderungen hingegen höchstens marginal und im Mittel leicht negativ, ein Hinweis auf physikalisch unbegründete Korrekturen in Regionen ohne dynamische Konnektivität zu den Assimilationsstationen. Eine Erhöhung der horizontalen und vertikalen Kernelskalen änderte dieses Ergebnis nicht wesentlich und verschlechterte die Ergebnisse an den Validierungsstationen leicht. Wie erwartet vergrößerte der synthetische Tracer den Wirkungsbereich um etwa 104% und weist damit auf günstigere Bedingungen für längerlebige Xenon-Isotope wie $^{131\text{m}}\text{Xe}$ hin.

Kurzfassung

Unter der aktuellen Stationsgeometrie fungiert Nudging somit vor allem als lokaler Korrekturmechanismus. Die breitere Wirksamkeit wird durch die Kernelskala, die Kohärenz der Strömung und die kurze Halbwertszeit von ^{133}Xe begrenzt, die die Reichweite der Nudging-Inkrementen einschränkt. Verbesserungen der Hintergrundfelder bleiben daher weitgehend auf Regionen mit persistierenden Windmustern beschränkt. Für eine belastbarere Vorhersagegüte über die Assimilationsstationen hinaus sind ein höherer Anteil zuverlässig quantifizierbarer Beobachtungen sowie ein dichteres, strategisch verteiltes Messnetz mit verbesserter Abdeckung in Quellregionen erforderlich, um die aus den Beobachtungen abgeleiteten Korrekturen dynamisch konsistent zu propagieren.

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1. Introduction

1.1. The Comprehensive Nuclear-Test-Ban Treaty

The Comprehensive Nuclear-Test-Ban Treaty (CTBT), adopted by the United Nations General Assembly in 1996, represents a landmark initiative aimed at eliminating nuclear testing worldwide. As of 2025, 187 states have signed the CTBT, with 178 having completed the ratification process (Comprehensive Nuclear-Test-Ban Treaty Organization, 2025b). However, the CTBT has yet to enter into force, as its implementation remains contingent upon ratification by the 44 states listed in Annex II, those possessing nuclear technology capabilities, after which the CTBT becomes legally binding under international law (Comprehensive Nuclear-Test-Ban Treaty Organization, 2022).

In anticipation of the eventual enforcement of the CTBT, the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) was established. The CTBTO pursues two primary objectives: First, it aims to promote recognition and acceptance of the CTBT. Second, it implements a robust verification regime, consisting of the International Monitoring System (IMS). The IMS is a global network of monitoring stations designed to detect nuclear tests anywhere in the world. It employs a multidisciplinary approach incorporating seismic, hydroacoustic, infrasound, and radio-nuclide monitoring. The regime aims to achieve a detection probability of 90% within approximately 14 days for a 1 kt TNT-equivalent atmospheric explosion, or for venting from an underground or underwater explosion, provided that releases exceed relevant detection thresholds (Schulze et al., 2000).

The introduction of the CTBT marked a significant milestone in nuclear non-proliferation. Since its adoption, the number of nuclear tests has declined sharply, from more than 2,000 in the preceding five decades to only ten thereafter (Arms Control Association, 2024). This dramatic reduction underscores the influence of the CTBT. Nevertheless, the Democratic People's Republic of Korea (DPRK) continues to pose a challenge by conducting nuclear tests in defiance of international norms, emphasising the need for universal adherence. Alongside India and Pakistan, the DPRK remains one of three Annex II states that have not signed the CTBT, highlighting the critical role of continued diplomatic engagement in achieving its universal acceptance (Comprehensive Nuclear-Test-Ban Treaty Organization, 2025b).

1. Introduction

1.2. Radioxenon for Verification Purposes

An indispensable component of the CTBT verification regime is the detection of radioactive noble gases, in particular radioxenon. Due to its chemically inert nature, radioxenon can migrate through subsurface pathways and escape into the atmosphere, even in cases where underground explosions are largely contained. This fugitive behaviour, combined with its low reactivity, makes it a highly informative tracer for atmospheric monitoring (Bourret et al., 2021; Kalinowski et al., 2010). Compared to particulate radionuclides, radioxenon is more likely to escape from underground cavities, offering a complementary and often more sensitive detection channel for identifying clandestine nuclear tests (Saey et al., 2006).

The diagnostic value of radioxenon lies in its isotopic composition. Ratios between $^{131\text{m}}\text{Xe}$, ^{133}Xe , $^{133\text{m}}\text{Xe}$, and ^{135}Xe provide temporal information that supports the distinction between civilian and non-civilian sources and may be used to estimate the time elapsed since a release (Kalinowski et al., 2010). These isotopes have distinct half-lives, 11.96 days, 5.24 days, 2.20 days, and 9.14 hours, respectively, and their relative abundances evolve predictably over time. For example, $^{133\text{m}}\text{Xe}$ decays to ^{133}Xe through isomeric transition, while ^{135}Xe undergoes beta decay (Galan et al., 2018). Isotopic ratio analysis is thus a powerful instrument for event attribution and compliance verification under the CTBT (Galan et al., 2018; Ringbom et al., 2021). These unique isotope ratios constitute release “signatures”—distinctive fingerprints of nuclear tests. By comparing them to known patterns from civilian facilities, the origin of a detection can often be identified with greater confidence. However, such attribution requires not only precise isotopic measurements but also reliable modelling of atmospheric transport and decay processes.

In practice, isotopic activity ratios are visualised in so-called four-isotope plots, which facilitate the classification of measured signatures. As illustrated in Figure 1.1, typical signatures of operational reactor emissions fall within the “reactor domain,” whereas signatures of nuclear tests lie within the “nuclear explosion domain,” separated by a line derived from fission product evolution models (Kalinowski and Pistner, 2006; Kalinowski et al., 2010; Baré et al., 2023). Over time, the decay of short-lived isotopes and mixing with aged air masses shift these ratios towards the reactor domain, even for initially explosion-like signatures. Observational campaigns show that emissions from medical isotope production facilities, despite the use of highly enriched uranium targets, predominantly exhibit reactor-like isotopic ratios. This is attributed to internal mixing of air streams within the facilities and to atmospheric dilution during transport (Baré et al., 2023). While this reduces the risk of false positives in the CTBT verification regime, it simultaneously raises concerns that genuine nuclear test signals may be masked by similar processes.

1.2. Radioxenon for Verification Purposes

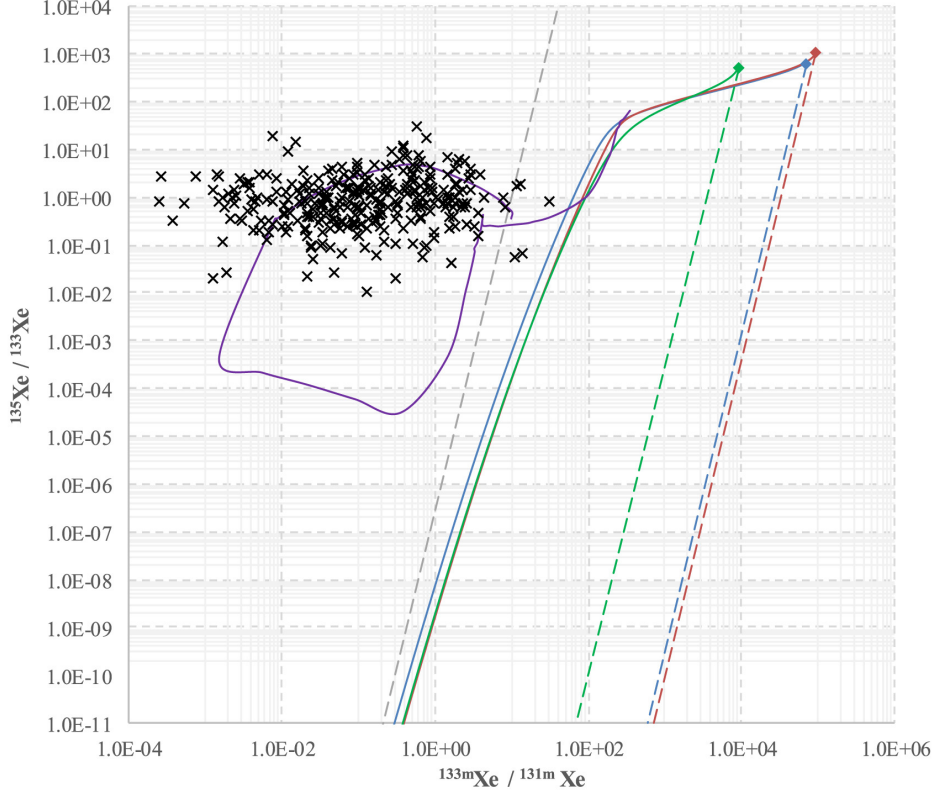


Figure 1.1.: Four-isotope plot comparing measured radioxenon signatures with theoretical signatures from nuclear tests and civilian sources, adapted from Baré et al. (2023). The most relevant elements are the grey dashed line, which separates the reactor domain (left) from the nuclear explosion domain (right), and the black crosses, which denote measured signatures. Additional elements are included for completeness but are not further interpreted here.

The capability of radioxenon detection was clearly demonstrated following the underground nuclear test conducted by the DPRK on 9 October 2006. Roughly two weeks after the event, elevated concentrations of ^{133}Xe were detected by the monitoring station in Yellowknife, Canada, consistent with a release from the DPRK test site (Comprehensive Nuclear-Test-Ban Treaty Organization, 2025a). The robust identification and attribution of such observations rely not only on isotopic signatures but also on a well-characterised background, ideally for all relevant radioxenon isotopes.

1. Introduction

1.3. Radioxenon Background Concentrations

In the context of radionuclide monitoring, the term “background” refers to atmospheric radioxenon originating exclusively from routine civilian operations, primarily from nuclear power plants, research reactors, and medical isotope production facilities. Potential nuclear explosion signals would be superimposed on this background. Accordingly, the background must be clearly distinguished from signals from nuclear tests to enable accurate compliance assessments under the CTBT verification regime (Baré et al., 2023). In practice, this is achieved only when the explosion signal exceeds background uncertainty by a sufficient margin.

Medical isotope production facilities are the predominant source, contributing up to 95% of global atmospheric radioxenon background concentrations at monitoring stations (Saey, 2009). Their emissions exhibit highly variable temporal patterns, including weekly cycles, irregular short-term spikes, and production-specific venting episodes, typically associated with operational procedures. This variability produces transient plumes that may simultaneously affect multiple stations, thereby complicating signal interpretation (Saey, 2009; Ringbom et al., 2021).

Although such emissions are of non-prohibited origin, their magnitude and variability pose a dual challenge, as they can under certain conditions mimic explosion-like signatures and may also obscure or dilute genuine signals from nuclear tests. Both effects reduce the discriminatory power of radioxenon monitoring and may influence the political interpretation of verification outcomes.

Reliable modelling of background concentrations is, therefore, indispensable. However, current emission inventories are subject to considerable uncertainty due to limited access to real-time stack measurements at source facilities. In the absence of such data, inventories often rely on operational assumptions, proxy estimates, or averaged emission rates, resulting in discrepancies between simulated and observed concentrations of several orders of magnitude (Ringbom et al., 2021). In addition, simplified treatment of radionuclide decay chains, for example, omitting $^{133\text{m}}\text{Xe}$ as a precursor to ^{133}Xe , can distort both spatial and temporal representations of the background.

Beyond uncertainties in source terms, errors in atmospheric transport modelling introduce additional biases stemming from limitations in meteorological forcing, unresolved sub-grid mixing, and numerical diffusion. To mitigate these shortcomings, data assimilation methods such as nudging are employed. Nudging adjusts simulated concentrations towards observations and thereby improves the accuracy and interpretability of background concentration fields. As they directly inform the CTBT verification regime, improving their representation is not merely a technical refinement but a prerequisite for credible and robust assessments.

2. Research Objectives

The reliable simulation of atmospheric ^{133}Xe background concentrations, particularly at radionuclide monitoring stations, is central to the timely and accurate identification of signals relevant to the Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification regime. Discrepancies between simulated and observed concentrations can erode confidence in atmospheric transport modelling, complicate the interpretation of observations, and potentially delay or mislead the attribution of nuclear signatures. Minimising these biases is, therefore, fundamental to strengthening the credibility of verification assessments conducted by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO).

The limited availability of temporally resolved and spatially comprehensive emission inventories, particularly for civilian sources such as medical isotope production facilities, remains an obstacle to accurate simulation. These gaps, compounded by uncertainties in atmospheric transport modelling and limited observational coverage, often lead to substantial mismatches between simulated and observed concentrations, in some cases by orders of magnitude. This persistent discrepancy underscores the need for correction techniques that are capable of systematically reducing model–observation differences and enhancing the reliability of simulation-based assessments.

Nudging presents a pragmatic and physically consistent approach to this challenge. By gradually steering simulated towards observed concentrations, nudging introduces corrections without disrupting the underlying dynamics of the model. This master’s thesis explores the potential of nudging to improve agreement with observations and investigates whether improvements at assimilation stations can be generalised to broader regions.

The overarching objective of this master’s thesis is defined as follows:

Improve global ^{133}Xe background concentration fields by applying nudging in FLEXPART-LCM.

To address this objective, the following sub-objectives are defined:

- **Implementation of nudging in FLEXPART-LCM.**
Integrate nudging for radionuclides, specifically ^{133}Xe , into FLEXPART-LCM, ensuring a stable and scientifically robust implementation.

2. Research Objectives

- **Specification of nudging parameters.**

Systematically explore horizontal kernel scales to identify an optimal configuration that minimises simulation–observation discrepancies while maintaining physical plausibility.

- **Evaluation at assimilation stations.**

Assess the performance of nudging at assimilation stations using standard metrics such as bias, mean squared error, and the coefficient of determination.

- **Evaluation at validation stations.**

Evaluate the generalisability of nudging by analysing its performance at validation stations excluded from assimilation, thereby assessing spatial transferability and robustness.

By addressing these sub-objectives, this master’s thesis aims to improve the representation of simulated atmospheric transport of ^{133}Xe and to provide a foundation for future advances in operational verification tools. The findings are expected to support the long-term objective of the CTBTO, which is to enhance modelling capabilities under realistic constraints.

3. Data

3.1. ^{133}Xe Emissions

The accurate representation of global ^{133}Xe emissions is a prerequisite for reliable atmospheric transport modelling and for the meaningful interpretation of radionuclide observations within the Comprehensive Nuclear-Test-Ban Treaty (CTBT) verification regime. For the reference year 2014, this master’s thesis draws upon emission inventories published by Kalinowski and Tatlisu (2020) and Kalinowski (2023), which provide temporally and spatially resolved estimates of civilian emissions worldwide. These sources constitute the dominant anthropogenic contributors to atmospheric ^{133}Xe (Achim et al., 2016).

The principal contributors considered in this master’s thesis fall into three categories:

- **Nuclear Power Plants**

Operating continuously worldwide, nuclear power plants emit ^{133}Xe as a by-product of routine fission during electricity generation. These releases exhibit irregular temporal patterns, with episodic peaks (Kalinowski, 2023). Although emissions are regulated and typically well controlled, their cumulative contribution, given the large number of facilities and operating hours, significantly affects global background concentrations.

- **Nuclear Research Reactors**

Primarily used for scientific experimentation and isotope production, research reactors exhibit diverse emission profiles, which are determined by reactor design and operational schedules. Neutron activation processes within these facilities release ^{133}Xe (Kalinowski, 2023).

- **Medical Isotope Production Facilities**

These facilities represent by far the most significant source of ^{133}Xe emissions, releasing substantial quantities during the production of radiopharmaceuticals such as ^{99}Mo , which decays to $^{99\text{m}}\text{Tc}$. The fission-based production pathways employed generate large volumes of fission gases, including ^{133}Xe (Kalinowski, 2023).

Other sources of ^{133}Xe exist but are excluded from this analysis due to their comparatively minor contributions or the lack of high-resolution inventories. These include:

3. Data

- **Spent Nuclear Fuel Reprocessing Facilities**

Emissions occur during the chemical separation of reusable nuclear material from spent fuel (Kalinowski, 2023).

- **Spallation Neutron Sources**

High-energy proton-induced reactions in target materials produce neutron activation products, including ^{133}Xe (Kalinowski, 2023).

Among the known sources, medical isotope production facilities are the dominant contributor to global emissions. As summarised in Table 3.1, they accounted for approximately 89.7% of the total emissions in 2014, corresponding to 7.89×10^{15} Bq. This overwhelming share underscores the necessity of accurately quantifying and incorporating their emissions into atmospheric transport models to minimise bias and ensure robust interpretation of radionuclide observations.

Type of Source	Number of Facilities	Temporal Resolution	Emissions (Bq/Year)
Nuclear Power Plants	173	Monthly	4.50×10^{14}
Nuclear Research Reactors	31	Annual	4.52×10^{14}
Medical Isotope Production Facilities	13	Annual	7.89×10^{15}

Table 3.1.: Total emissions by source type in 2014, including facility count and temporal resolution.

Notably, one of the thirteen medical isotope production facilities, located in Chalk River, Canada, provides emissions at daily resolution, offering enhanced temporal fidelity for atmospheric transport simulations.

Figure 3.1 illustrates the global distribution of known ^{133}Xe sources, highlighting regions of elevated source density. Prominent clusters include North America, particularly the Eastern United States, Western and Central Europe, and parts of South and East Asia such as India, South Korea, China, and Japan. In the Southern Hemisphere, sources are fewer, but major facilities operate in Argentina, South Africa, Australia, and Indonesia.

3.2. ^{133}Xe Observations

High-quality observations, indispensable for robust performance evaluation and impactful nudging, are provided by the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) through the radionuclide monitoring stations of the International Monitoring System (IMS) (Comprehensive Nuclear-Test-Ban Treaty Organization, 2022).

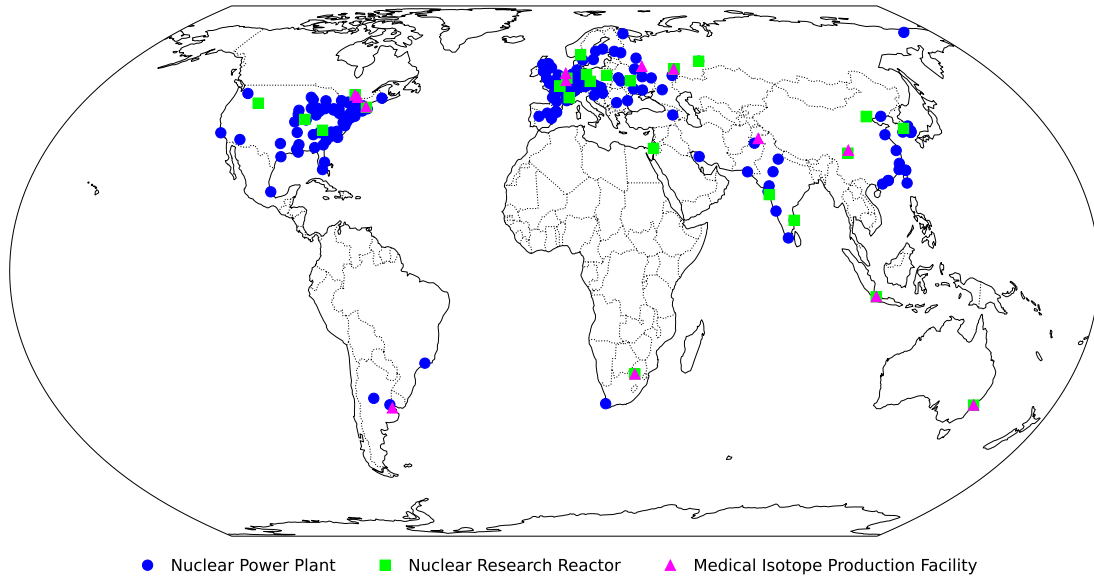


Figure 3.1.: Global distribution of ^{133}Xe emission sources in 2014.

3.2.1. Monitoring Systems

The IMS operates two principal noble gas monitoring systems:

- Swedish Automatic Unit for Noble Gas Acquisition (SAUNA)**
 Developed by the Swedish Defence Research Agency (FOI), SAUNA stations employ cryogenic sampling and beta–gamma coincidence detection to enable continuous, high-sensitivity monitoring of xenon isotopes under field conditions (Aldener et al., 2023).
- Système de Prélèvement Automatique en Ligne avec l'Analyse du Xénon (SPALAX)**
 Developed by the French Atomic Energy Commission (CEA), SPALAX stations extract and concentrate xenon from ambient air for in situ measurement using a fully automated process optimised for long-term field deployment (Fontaine et al., 2004).

3. Data

3.2.2. Observation Specifications

This master’s thesis utilises a comprehensive dataset of atmospheric ^{133}Xe observations provided by the CTBTO, covering the period from 1 January 2014 to 31 December 2020. To ensure consistency with emission inventories and to streamline the validation framework, the analysis is restricted to the reference year 2014. 20 monitoring stations were selected based on the availability of sufficient measurements, defined as valid ^{133}Xe observations reported from February through December 2014. January 2014 is excluded and used as a model spin-up period to stabilise initial conditions in the transport simulations.

SAUNA stations provide observations every 12 hours, whereas SPALAX stations report every 24 hours. Observed concentrations typically range from a few to several tens of millibecquerels per cubic metre (mBq m^{-3}) near emission sources and are accompanied by a minimum detectable concentration (MDC), which defines the threshold below which values are statistically uncertain. In remote regions or during periods of low emissions, the MDC is a critical reference for interpretation. Observations below the MDC are treated using the statistical substitution method described in Section 4.7.1.

3.2.3. Monitoring Stations

Table 3.2 lists the selected stations, both assimilation and validation stations, including the type of monitoring system, geographic coordinates, and the number of observations above and below the MDC. The stations span a wide latitudinal and longitudinal range, providing substantial global coverage for model evaluation. However, large regions, including the Atlantic, Africa, and the Middle East, lack adequate observational coverage, limiting representativeness in these areas. Figure 3.2 visualises the geographic distribution.

Country	Station Code	System	Latitude (°)	Longitude (°)	<MDC	>MDC
Australia	AUX09	SAUNA	−12.40	130.90	535	71
Brazil	BRX11	SAUNA	−23.00	316.60	622	9
Canada	CAX16	SPALAX	62.50	245.50	317	7
France	FRX27	SPALAX	−17.60	210.40	324	0
France	FRX30	SPALAX	−49.40	70.30	278	0
France	FRX31	SPALAX	5.20	307.30	22	0
Germany	DEX33	SPALAX	47.90	7.90	92	170
Japan	JPX38	SAUNA	36.30	139.10	286	154
Mexico	MXX44	SAUNA	28.00	245.90	564	6
Mongolia	MNX45	SPALAX	47.90	105.30	307	19
NZ	NZX46	SAUNA	−43.80	183.50	600	41
Norway	NOX49	SAUNA	78.20	15.40	413	77
Panama	PAX50	SPALAX	9.00	280.50	263	6
Sweden	SEX63	SAUNA	59.40	17.90	182	471
UK	GBX66	SAUNA	−7.30	72.40	582	12
UK	GBX68	SAUNA	−37.10	347.70	324	9
USA	USX74	SAUNA	37.20	260.20	467	73
USA	USX75	SAUNA	38.00	281.60	178	436
USA	USX77	SAUNA	19.30	166.60	630	3
USA	USX79	SAUNA	21.50	202.00	646	5

Table 3.2.: Selected monitoring stations, including monitoring system, geographic coordinates, and the number of ^{133}Xe observations above and below the MDC.

3.2.4. Validation Stations

Of the 20 stations listed in Table 3.2, five are designated as independent validation stations: FRX30, MNX45, PAX50, SEX63, and USX75. These stations are excluded from the nudging process and serve to evaluate spatial generalisation and robustness beyond regions directly influenced by assimilated observations.

The selection ensures a broad representation of climatic and geographic conditions. FRX30, located in the Southern Indian Ocean, samples a remote marine environment. MNX45 represents a high-latitude inland station in East Asia. PAX50 is situated in Central America, within tropical circulation regimes. SEX63 is a mid-latitude station in Northern Europe, and USX75 is characterised by persistently elevated ^{133}Xe concentrations.

3. Data

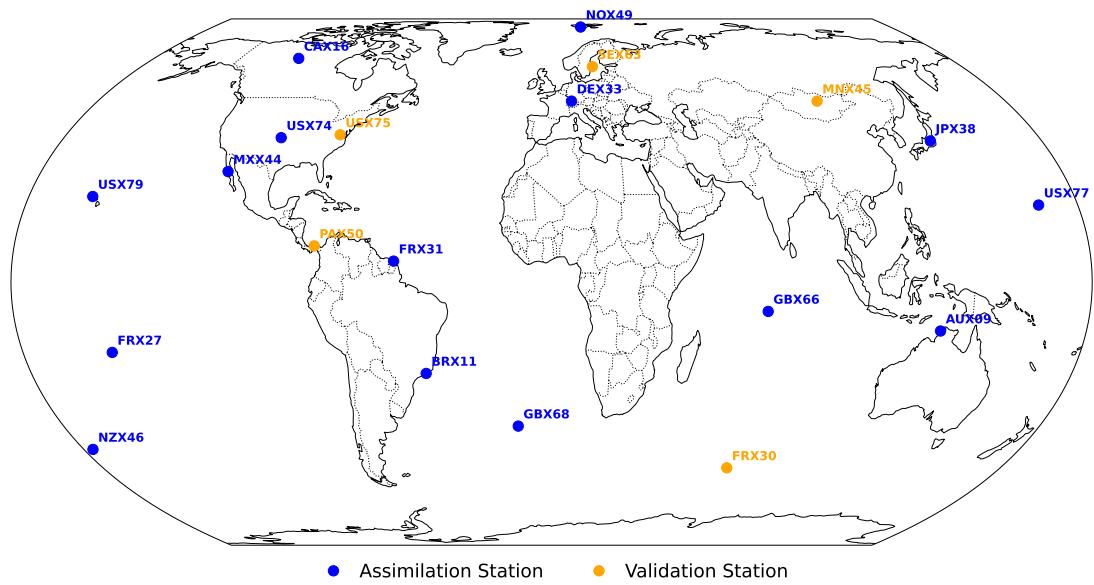


Figure 3.2.: Global distribution of the selected assimilation and validation stations.

4. Methodology

4.1. Introduction to FLEXPART-LCM

FLEXPART(FLEXible PARTicle Dispersion Model) is a state-of-the-art Lagrangian transport model designed to simulate the atmospheric dispersion and transport of trace gases and particles. The release of FLEXPART 11, which incorporates the Linear Chemistry Module (LCM), represents a significant advancement over previous iterations, introducing improvements in computational performance, transport accuracy, and modular flexibility (Bakels et al., 2024). These enhancements make the model well-suited for both research-driven and operational applications across a wide range of spatial and temporal scales.

Among the integrated features of FLEXPART-LCM, the global domain-filling mode and nudging are especially relevant to the objectives of this master’s thesis. While applicable in various modelling contexts, they are employed here to address specific challenges associated with sparse observational coverage and uncertainties in emission inventories. Originally developed within the FLEXPART-Chemistry Transport Model (FLEXPART-CTM) (Henne et al., 2018; Groot Zwaaftink et al., 2018), both approaches have been refined and optimised in FLEXPART-LCM. Their integration provides a robust framework for aligning simulated concentrations with observations in a physically consistent manner.

4.2. Command Configuration

The `COMMAND` file defines the control parameters governing the FLEXPART-LCM simulation of ^{133}Xe . The simulation covers the entire year of 2014, commencing with a one-month spin-up period from 1 to 31 January. This spin-up period allows the model to reach a dynamically balanced state and mitigates artefacts associated with the initialisation. It is excluded from all subsequent analysis.

The simulation is executed in a global domain-filling configuration (`MDOMAINFILL = 1`) and produces output at 12-hour intervals, with concentrations averaged over 1-hour windows. This temporal resolution offers a compromise between computational efficiency and the granularity required for comparison with observations. Since model output times do not always coincide with measurement timestamps, linear temporal interpolation is applied to synchronise simulated concentrations with the timing of available observations.

4. Methodology

To ensure consistent internal synchronisation across model components, the parameter `LSYNCTIME` is configured to 900 seconds (15 minutes). This interval governs the frequency at which subcomponents are synchronised. While shorter intervals improve temporal accuracy, they also increase computational demand.

Several additional configuration switches are activated to support the nudging functionality within FLEXPART-LCM. Although these features are broadly applicable in atmospheric transport modelling, they are specifically employed here to facilitate the implementation of nudging:

- `LCMOUTPUT = 1`
Enables the Linear Chemistry Module (LCM).
- `NUDGING = 1`
Activates the nudging functionality.
- `UNIT_INIT = 1`
Specifies the unit of the latitude profile used for initialisation. A value of 1 indicates units of Bq/m^3 . The latitude profile is described in detail in Section 4.6.

4.3. Release

The `RELEASE` file defines the initial configuration for particle injection. In this simulation, 50 million particles are released instantaneously at the beginning of the simulation period, uniformly distributed across the global domain. When operating in domain-filling mode, FLEXPART-LCM automatically extends the vertical release range to encompass the entire atmosphere (Stohl et al., 2010; European Centre for Medium-Range Weather Forecasts, 2021).

The choice of 50 million particles reflects a deliberate balance between computational efficiency and statistical robustness. This particle count ensures sufficient resolution of the tracer field in both space and time, reduces stochastic noise, and enables meaningful comparisons with observations.

4.4. Outgrid

The `OUTGRID` file defines the spatial resolution of the model output. A horizontal grid of $0.5^\circ \times 0.5^\circ$ is employed, matching the resolution of the ERA5 meteorological input data.

The model output is restricted to the lowest 100 metres above ground level, providing a representative approximation of surface-level air masses. This configuration ensures a sufficient number of particles for statistically reliable output, facilitating direct comparison with observed ^{133}Xe concentrations at monitoring stations.

4.5. Emissions

Hourly emissions of ^{133}Xe are defined in dedicated emission files, encompassing sources such as nuclear power plants, research reactors, and medical isotope production facilities. The emissions are provided in becquerels per second (Bq/s) and assigned to specific geographic source locations. This format enables a realistic representation of anthropogenic contributions to the atmospheric ^{133}Xe background.

The structure and formatting of these emission files follow the specifications outlined in Henne et al. (2018).

4.6. Initialisation

In FLEXPART-LCM, the global domain-filling mode enables particles to acquire mass from predefined emission sources, as specified in the emission files, provided they pass within a defined proximity of these sources. This mechanism ensures a spatially consistent and physically plausible mass distribution throughout the model domain, even in regions with sparse source coverage. It is particularly well suited for simulating background concentrations and long-range transport of ^{133}Xe , where direct emissions may be limited or geographically dispersed.

To initialise the background concentration field, a latitude-dependent profile is applied. Defined at a spatial resolution of 0.5 latitude, this profile assigns a uniform concentration of 0.1 mBq/m³ across all latitudinal bands. Although simplified, this approach offers a practical and computationally efficient approximation of the global ^{133}Xe background. The selected value of 0.1 mBq/m³ is justified by the relatively short half-life of ^{133}Xe (5.24 days), which ensures that any artificial influence from the initial field decays rapidly during the one-month spin-up period in January 2014. This makes the chosen value a scientifically sound and operationally robust assumption for model initialisation.

The corresponding file, `latitude_profile_Xe-133.txt`, is stored within the output directory and automatically accessed during model start-up to initialise the tracer field.

4.7. Observations

4.7.1. Treatment of Concentrations Below the Minimum Detectable Concentration

Every measurement from the International Monitoring System (IMS) includes a minimum detectable concentration (MDC), which marks the lowest concentration that can be distinguished confidently from background noise. With approximately 83% of observations falling below the MDC, the treatment of concentrations below the MDC constitutes a fundamental requirement for a reliable and unbiased evaluation. Neglecting or misrepresenting these observations may lead to significant distortions in the interpretation of background concentrations and compromise subsequent evaluation.

To address this challenge, a probabilistic estimation method is applied to infer expected values for concentrations below the MDC. This approach is grounded in empirical observations and statistically supported assumptions regarding the distribution of ^{133}Xe . For comparison, an alternative method, full-MDC substitution, is briefly discussed below.

Full-MDC Substitution

A conservative method occasionally used in operational contexts, as described by Kalinowski et al. (2010), is the full-MDC substitution approach. In this method, every concentration reported below the MDC is replaced by the full value of the MDC. It assumes that concentrations not accurately quantifiable are effectively at the detection threshold. While straightforward to implement and protective against underestimation, this approach systematically overestimates background concentrations, introducing an upward bias that can distort statistical evaluations.

Estimating Expected Concentrations Below the MDC

To obtain a more realistic representation of concentrations below the MDC, a probabilistic approach is employed. It assumes that concentrations follow an exponential distribution—a hypothesis supported by empirical evidence indicating that ^{133}Xe concentrations often exhibit exponential characteristics.

For each monitoring station s , the probability density function (PDF) of the exponential distribution is defined as:

$$f_s(C) = \lambda_s e^{-\lambda_s C}, \quad C \geq 0, \quad (4.1)$$

where C denotes the concentration, and λ_s is the rate parameter, calculated as the reciprocal of the sample mean μ_s of all observations above the MDC at station s :

$$\lambda_s = \frac{1}{\mu_s}. \quad (4.2)$$

Using this distribution, the expected concentration below the MDC for a given observation i at station s , denoted $C_{s,i}^{\text{below}}$, is computed by integrating the PDF over the interval $[0, \text{MDC}_{s,i}]$:

$$C_{s,i}^{\text{below}} = \int_0^{\text{MDC}_{s,i}} C \cdot f_s(C) dC, \quad (4.3)$$

which yields the following expression:

$$C_{s,i}^{\text{below}} = \left(1 - e^{-\lambda_s \cdot \text{MDC}_{s,i}}\right) \cdot \frac{1}{\lambda_s}. \quad (4.4)$$

This method provides an analytically derived, station-specific estimate for observations below the MDC that reflects the underlying statistical distribution of measured concentrations. By incorporating these values into further analyses, the approach avoids the systematic biases associated with full-MDC substitution while preserving the integrity of observations at low concentrations.

4.8. Nudging

The `NUDGING` file contains station-specific parameters that control the nudging scheme, namely the spatial kernel half-widths h_x , h_y , h_z , the temporal kernel half-width h_t , and the nudging relaxation timescale τ_n .

4.8.1. Nudging Formulation

Nudging is a data assimilation approach that incrementally adjusts simulated concentrations towards observations through continuous, small corrections. In this configuration, nudging operates on particle activity concentrations using ^{133}Xe observations. The procedure reduces systematic biases and improves agreement with observations while preserving the intrinsic transport dynamics of FLEXPART-LCM. The implementation follows the formulation of Henne et al. (2018) where each observation is associated with a four-dimensional kernel in space and time, and simulated concentrations are relaxed towards the observed state with weights that depend on spatial and temporal proximity.

Kernel Weights

Corrections are apportioned to individual particles using spatial and temporal kernel weights that quantify proximity between an observation and a particle.

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The spatial weight w_s for an observation–particle pair (i, j) is based on an Epanechnikov kernel and is defined by

$$w_s = 1 - r_{i,j}^2 \quad \text{for} \quad r_{i,j}^2 \leq 1, \quad (4.5)$$

with the normalised squared distance

$$r_{i,j}^2 = \left(\frac{x_j - x_i}{h_x} \right)^2 + \left(\frac{y_j - y_i}{h_y} \right)^2 + \left(\frac{z_j - z_i}{h_z} \right)^2. \quad (4.6)$$

Coordinates (x_j, y_j, z_j) refer to particle j and (x_i, y_i, z_i) refer to observation i . The kernel half-widths h_x , h_y , and h_z define the spatial domain of influence, and observations affect only particles for which $r_{i,j}^2 \leq 1$.

The temporal weight w_t employs a tricubic kernel given by

$$w_t = (1 - |r_t|^3)^3 \quad \text{for} \quad |r_t| \leq 1, \quad (4.7)$$

where the normalised temporal distance is

$$r_t = \frac{t_k - t_l}{h_t}. \quad (4.8)$$

Here t_k denotes the current model time and t_l denotes the timestamp of the closest valid observation. The temporal window of influence is controlled by h_t .

Particle Mass Adjustment

The incremental mass correction applied to particle j over a synchronisation interval Δt is

$$\Delta m_j = \frac{M_i - m_j}{\tau_n} \Delta t w_s w_t, \quad (4.9)$$

where M_i is the target mass implied by observation i , computed as $M_i = C_i \cdot M_{\text{air}}$. The observed concentration is C_i and M_{air} denotes the air mass associated with the model grid cell. The current particle mass is m_j . The relaxation timescale τ_n controls the rate at which the simulated mass approaches the observationally implied mass. Numerical stability requires $\Delta t < \tau_n$.

This formulation concentrates corrections on particles that are proximate to the observation in space and time and attenuates the correction smoothly with distance. The result is a physically plausible adjustment that preserves numerical stability and avoids abrupt perturbations of the transport field.

4.8.2. Practical Considerations

The choice of the kernel half-widths, henceforth referred to as kernel scale, and τ_n determines the balance between local fidelity and spatial generalisation. Larger kernel scales increase the spatial reach of observations but reduce localisation of corrections. Shorter relaxation times produce faster adjustment but risk introducing dynamical inconsistency. The parameters are, therefore, selected to optimise bias reduction at assimilation stations while maintaining coherent transport patterns.

4.8.3. Kernel Specifications

To implement nudging in FLEXPART-LCM, spatially and temporally adaptive kernels are employed, whose scales are chosen to reflect the local statistical character of the concentration field. The objective is to assign an influence domain to each observation that is commensurate with its representativeness, thereby enhancing assimilation fidelity while limiting the artificial spatial diffusion of observational information.

This formulation extends the methodology first proposed by Groot Zwaaftink et al. (2018) and later adopted by Vojta et al. (2022). The central premise is that the kernel scale should vary inversely with the observed variability. In regions where concentrations exhibit strong, small-scale variability, a compact kernel preserves salient gradients, while in regions characterised by smooth, low-variance backgrounds, a broader kernel improves representativeness without materially perturbing the large-scale field. Linking kernel scale to observed variability, therefore, yields an adaptive influence radius that respects the intrinsic spatial and temporal structure of the observations.

Conceptually, the procedure begins from a linear inverse scaling:

$$h_i = h_{\text{base}} \frac{\sigma_{\text{max}}}{\sigma_i}, \quad (4.10)$$

where σ_i denotes the standard deviation of concentrations at station i and σ_{max} denotes the maximum standard deviation across all stations. In practice, however, a direct inverse law produces extreme scales when a few stations exhibit anomalously large variability. To temper these extremes, the inverse relation is moderated by a logarithmic transform that compresses the dynamic range and yields a more balanced distribution of scales across stations. The implemented expressions are

$$h_{\text{x},i} = h_{\text{x},\text{base}} \lambda \ln \left(1 + \frac{\sigma_{\text{max}}}{\sigma_i} \right), \quad (4.11)$$

$$h_{\text{t},i} = h_{\text{t},\text{base}} 6 \ln \left(1 + \frac{\sigma_{\text{max}}}{\sigma_i} \right), \quad (4.12)$$

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where $h_{x,i}$ (with $h_{x,i} = h_{y,i}$) and $h_{t,i}$ denote the spatial and temporal kernel scales respectively for station i . The parameters $h_{x,\text{base}}$ and $h_{t,\text{base}}$ are baseline scales, and λ is a dimensionless spatial scaling factor that modulates the overall horizontal reach.

The baseline parameters are as follows:

- Base spatial kernel scale: $h_{x,\text{base}} = 0.5^\circ$
- Base temporal kernel scale: $h_{t,\text{base}} = 3600$ seconds
- Vertical kernel scale: $h_z = 500$ metres
- Nudging relaxation timescale: $\tau_n = 1800$ seconds

These choices reflect a deliberate compromise among representativeness, alignment with the model grid and meteorological input, and numerical stability. The horizontal baseline equals the OUTGRID and ERA5 resolution, and the temporal baseline matches the simulation averaging interval. The vertical scale focuses on nudging within the lower troposphere, where surface-level measurements are most informative. The relaxation timescale is configured to twice the internal synchronisation interval to achieve a substantial yet numerically stable adjustment rate consistent with the stability constraints of the model.

By construction, the adaptive kernel assigns broader scales to stations that exhibit low temporal variability and tighter scales to stations with pronounced temporal variability. The result is a nudging framework that prioritises local fidelity where necessary and enhances representativeness where appropriate, resulting in an assimilation that is both physically coherent and dynamically responsive.

4.8.4. Sensitivity of Nudging to the Spatial Scale Parameter

The sensitivity experiment quantifies how the spatial scaling factor λ affects assimilation behaviour. The factor is varied across three values (4, 6, and 8) to probe the dependence of assimilation outcomes on horizontal reach. Changes in λ alter the spatial kernel scales and thereby modulate the strength and geographic extent of observational influence.

The principal objective of these experiments is to identify the parametrisation that minimises systematic bias at assimilation stations while preserving physically plausible transport structures. The temporal scale remains fixed because the short radioactive half-life of ^{133}Xe constrains the effective temporal horizon of observational influence. Varying the temporal scale is, therefore, unlikely to materially alter results and would increase experimental complexity.

4.9. Species

The **SPECIES** file defines the physical and chemical properties of the tracer used in the simulation. For ^{133}Xe , a half-life of 5.24 days and a molar mass of 132.91 grams per mol (g/mol) are specified (International Atomic Energy Agency, 2025).

In addition to defining species-specific attributes, this file also establishes the linkage to the corresponding emissions. It specifies the path to the emission files, the file naming convention, and relevant metadata through the following parameters:

- **PEMIS_PATH="/flexpart/options/input/emissions/"**
Specifies the directory containing the emission files.
- **PEMIS_FILE="Em2014MMDD.nc"**
Specifies the filename pattern of daily emission files, with placeholders for month (MM) and day (DD).
- **PEMIS_NAME="Xe-133"**
Specifies the variable name within the emission files that corresponds to ^{133}Xe .
- **PEMIS_UNIT=0**
Specifies the emission unit type. A value of 0 denotes emissions per grid cell.
- **PEMIS_COEFF=1.0**
Specifies the conversion factor for transforming the input emissions into units of Bq/s.

This configuration enables the model to ingest spatially and temporally resolved emissions of ^{133}Xe . Emissions are read from daily NetCDF files based on the defined naming convention. The specified variable is extracted and scaled in accordance with the **PEMIS_UNIT** and **PEMIS_COEFF** parameters, ensuring consistent and correct integration into the simulation framework.

4.10. Statistical Parameters

To evaluate the accuracy and performance of the model under the influence of nudging, several statistical parameters are calculated. These metrics quantify the discrepancies between simulated concentrations C_i^{sim} and observed concentrations C_i^{obs} of ^{133}Xe .

4.10.1. Bias

Bias quantifies the average deviation of the simulation from the observations. It indicates whether the model systematically overestimates or underestimates concentrations. A positive bias implies overestimation, while a negative bias indicates underestimation. The bias is computed as:

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$$\text{Bias} = \frac{1}{n} \sum_{i=1}^n (C_i^{\text{sim}} - C_i^{\text{obs}}), \quad (4.13)$$

where n denotes the number of paired observations and model outputs.

4.10.2. Mean Squared Error

The mean squared error (MSE) measures the average of the squared differences between simulated and observed concentrations. As it penalises larger errors more severely, it is sensitive to outliers and provides a robust indicator of overall model accuracy:

$$\text{MSE} = \frac{1}{n} \sum_{i=1}^n (C_i^{\text{sim}} - C_i^{\text{obs}})^2. \quad (4.14)$$

4.10.3. Coefficient of Determination

The coefficient of determination (R^2) assesses how well the model captures the variability in observed concentrations. It represents the proportion of variance in the observations that is explained by the simulated concentrations. It is calculated as:

$$R^2 = 1 - \frac{\sum_{i=1}^n (C_i^{\text{sim}} - C_i^{\text{obs}})^2}{\sum_{i=1}^n (C_i^{\text{obs}} - \overline{C^{\text{obs}}})^2}, \quad (4.15)$$

where $\overline{C^{\text{obs}}}$ is the mean of the observed concentrations. An R^2 value of 1 indicates perfect agreement, while a value of 0 indicates that the model does not explain any of the observed variance.

These statistical parameters are standard in atmospheric model evaluation and are widely used to quantify model skill and identify potential biases or deficiencies in the simulation output (Van Leuven et al., 2023).

5. Results and Discussion

This chapter articulates and interprets the principal findings in direct relation to the overarching objective, namely the improvement of global ^{133}Xe background concentration fields through the targeted application of nudging within FLEXPART-LCM.

The results are organised according to the predefined sub-objectives. The first sub-objective, implementation of nudging for ^{133}Xe within FLEXPART-LCM, is summarised in Section 4. The remaining sections focus on the following:

- (1) Determination of optimal nudging parameters through sensitivity analyses of the spatial kernel scales.
- (2) Assessment of model performance at assimilation stations where nudging is actively applied.
- (3) Assessment of the generalisability of nudging at independent validation stations excluded from the assimilation process.

Throughout the chapter, performance is reported both for the complete observational record, which includes probabilistically estimated concentrations as described (hereafter "unconstrained record"), and for the subset restricted to observations above the minimum detectable concentration (MDC) (hereafter "constrained record"), thereby permitting an assessment that balances utilisation of the full record with emphasis on the most reliably quantifiable observations.

5.1. Baseline ^{133}Xe Background Concentration Field

To establish a reference for assessing the influence of nudging, Figure 5.1 depicts the global mean distribution of ^{133}Xe activity concentrations from the free-running simulation averaged over February to December 2014. This unconstrained field encapsulates the native background behaviour and constitutes the primary benchmark for evaluating nudged simulations. Figure 5.2 overlays known emission locations to clarify the spatial correspondence between source regions and simulated maxima. Pronounced concentration maxima occur downwind of emission clusters and are most prominent in the vicinity of medical isotope production facilities. Elevated means across the North Atlantic reflect the combined influence of densely clustered sources and prevailing westerly transport. Background concentrations in the Southern Hemisphere are more spatially isolated and can be attributed to individual facilities in Argentina, South Africa, and Australia.

5. Results and Discussion

The unconstrained field thus reveals persistent background concentrations along principal transport corridors, peak values proximate to source regions, and gradual attenuation with increasing distance from emissions—a spatial configuration that emphasises the potential for targeted observational constraints to reduce systematic model bias and improve the physical realism of simulated background concentration fields at hemispheric scales.

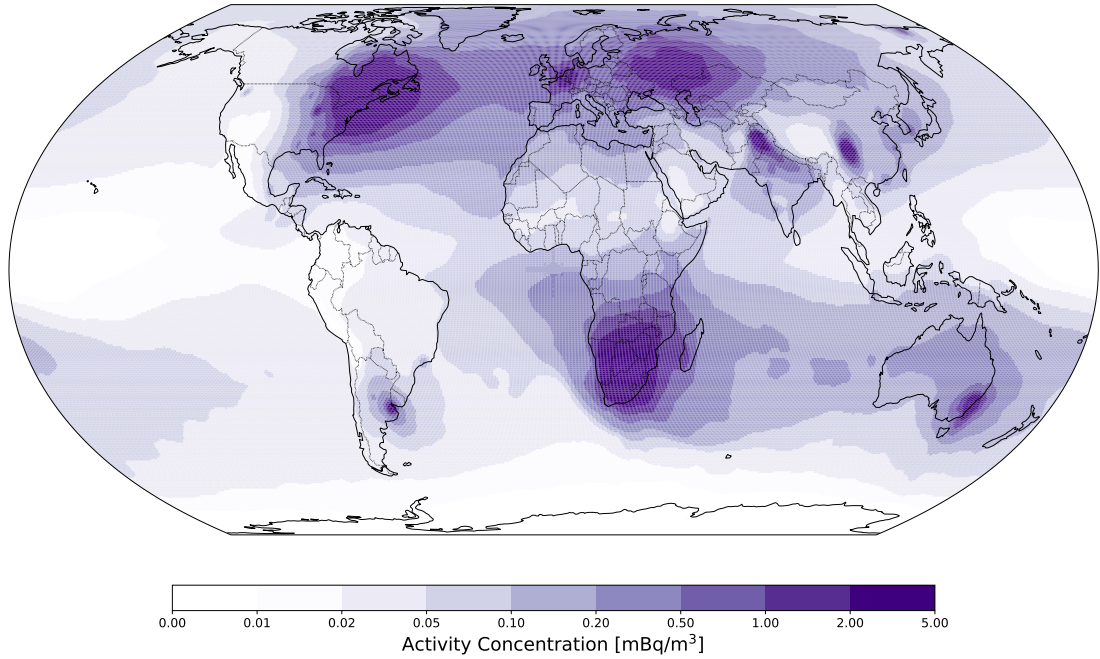


Figure 5.1.: Global mean ^{133}Xe activity concentration for February to December 2014 from the free-running simulation.

5.2. Selection of Nudging Parameters

The horizontal kernel scale represents a principal control in the nudging framework, as it prescribes the spatial extent over which a single assimilation station modifies the model state. Its tuning, therefore, directly shapes both the localisation of analysis increments and the degree to which sparse observations can inform regional and hemispheric background concentration fields. To characterise the sensitivity of model performance to the spatial scaling factor λ , that factor was varied across three representative values, $\lambda = 4, 6$, and 8 , thereby altering the decay rate of observational influence with distance.

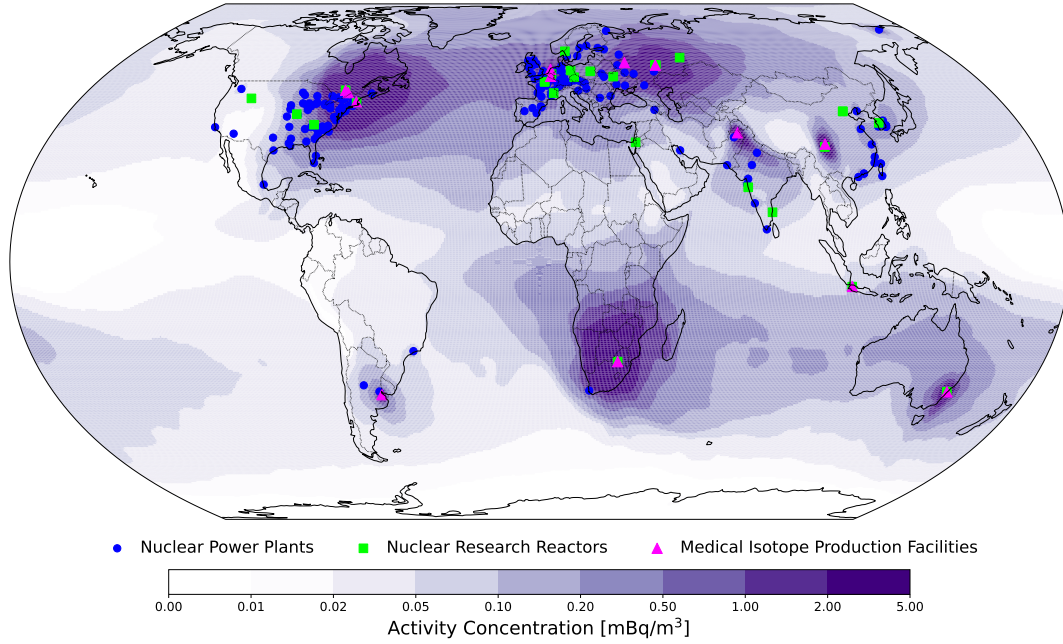


Figure 5.2.: Global mean ^{133}Xe activity concentration for February to December 2014 with emission sources overlaid.

Figure 5.3 presents the station-specific horizontal scales that result from these choices and illustrates the systematic expansion of influence with increasing λ , while simultaneously exposing persistent geographic gaps in direct observational reach, most notably across parts of Africa, the Middle East, and Central Asia, demonstrating that kernel tuning cannot substitute for improved observational coverage.

Figures 5.4 and 5.5 report the diagnostic statistics used to quantify performance, bias, mean squared error (MSE), and the coefficient of determination (R^2), with observational uncertainty propagated into the metrics. The comparison indicates that $\lambda = 8$ yields the lowest mean squared error for both records and the highest coefficient of determination for the unconstrained record. Quantitatively, for the unconstrained record, $\lambda = 8$ attains the lowest mean squared error (0.660) while maintaining a low bias (0.014) and a high coefficient of determination (0.808). This configuration outperforms the no-nudging baseline (Bias: 0.017, MSE: 3.892, R^2 : 0.628) and offers modest improvements over $\lambda = 4$ and $\lambda = 6$ in terms of error magnitude.

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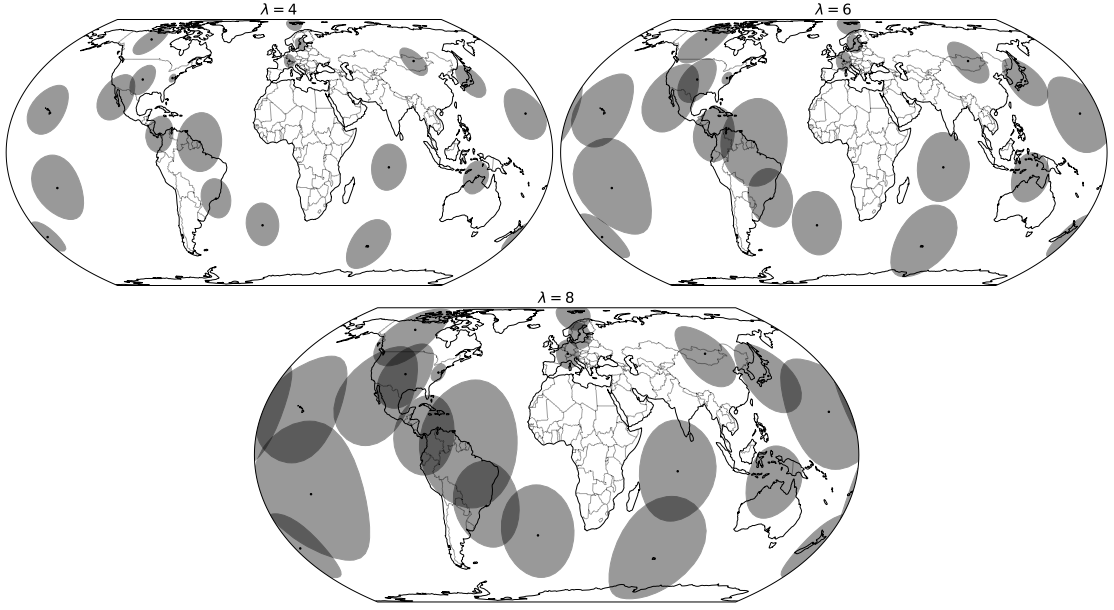


Figure 5.3.: Station-specific horizontal kernel scales for $\lambda = 4, 6$, and 8 .

For the constrained record, $\lambda = 8$ achieves the smallest bias (-0.011) and the lowest mean squared error (1.130). Although performance differences between the different kernel scales are generally modest, the trend is clear that larger kernels yield better results directly at assimilation stations. They cannot, however, be increased without bound, because overly large kernels diffuse observational influence into distant, physically unrelated regions and can distort concentrations far from the stations.

Physically, broader horizontal kernel scales are congruent with the transport and decay characteristics of ^{133}Xe . Radioactive decay, together with rapid atmospheric dispersion, limits the persistence of strictly local signals and thereby increases the relative utility of spatially distributed observational constraints. A larger observational footprint tends to improve representation of synoptic-scale features and enhance the effectiveness of a sparse network. All else equal, smaller kernels are preferable, as they better preserve sharp gradients and reduce the risk of introducing non-physical, long-range influence. In the present application, the choice of larger kernel scales is primarily a pragmatic response to the thin measurement network, which would otherwise leave substantial regions effectively unconstrained. The principal drawback of larger kernel scales is the smoothing of sharp local gradients, which can attenuate highly localised peaks. This trade-off must be balanced against the objective of improving large-scale background concentration fields.

5.2. Selection of Nudging Parameters

Accordingly, the choice of $\lambda = 8$ represents a pragmatic compromise that balances statistical improvement, physical consistency, and the targeted spatial scales, while recognising that further refinement would require either denser observational coverage or complementary assimilation methods capable of dynamically propagating observational information beyond local footprints.

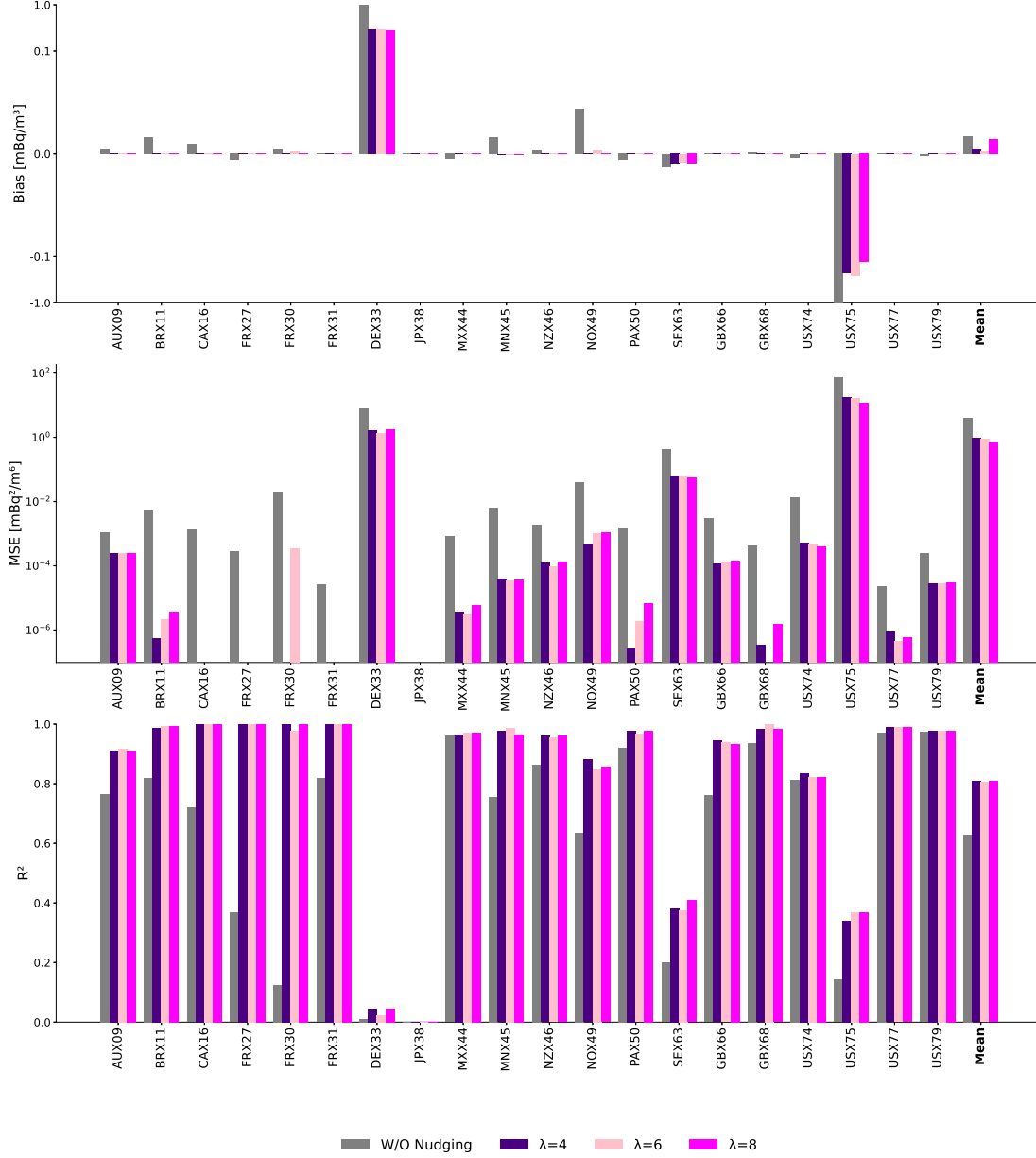


Figure 5.4.: Statistical performance of different kernel scales for the unconstrained record.

5. Results and Discussion

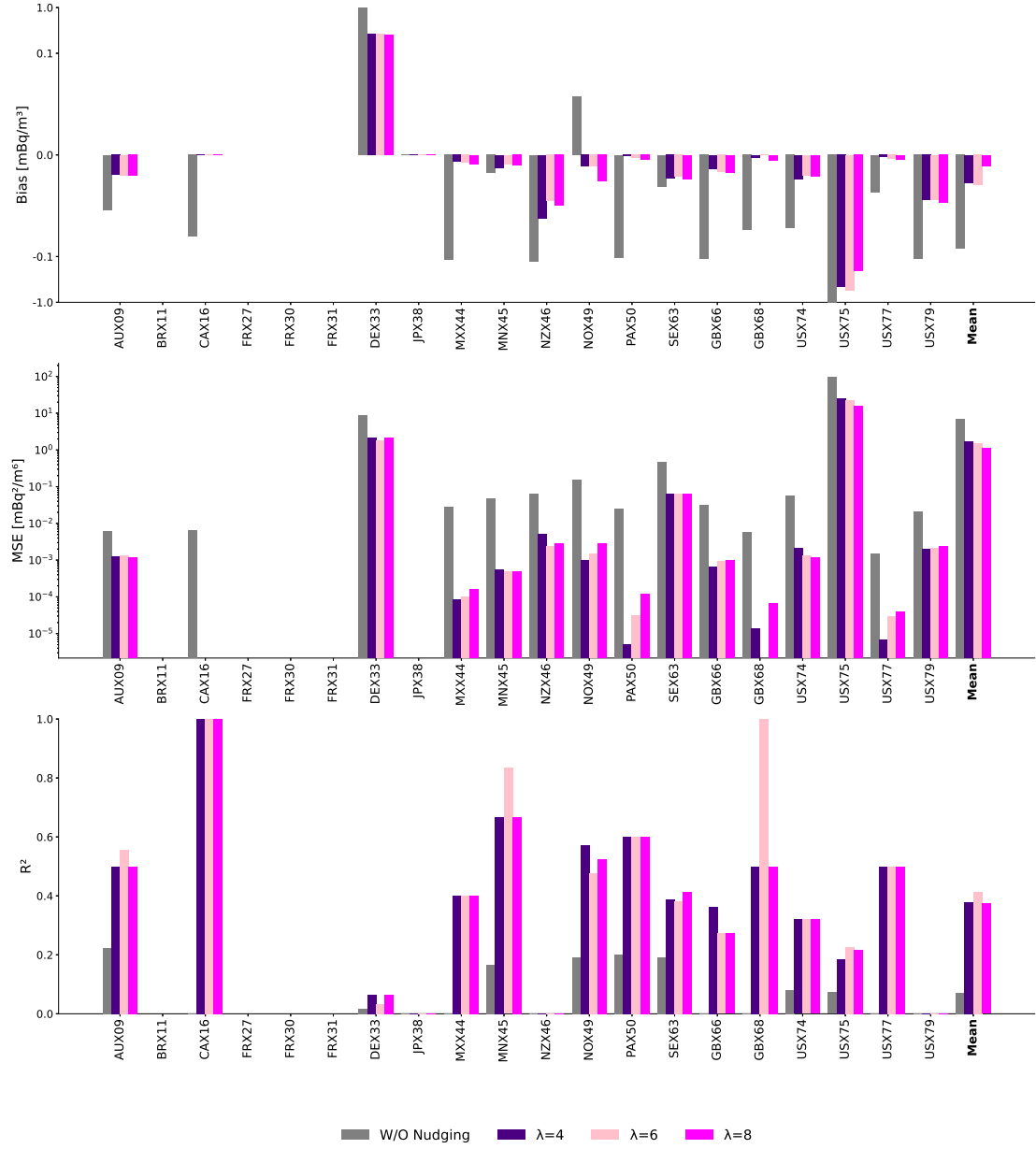


Figure 5.5.: Statistical performance of different kernel scales for the constrained record.

5.3. Impact of Nudging at Assimilation Stations

This section evaluates the impact of nudging at assimilation stations with the principal objective of quantifying improvements in model skill in the immediate vicinity of assimilated observations and thereby assessing both the technical fidelity of the implementation and the practical effectiveness of nudging within FLEXPART-LCM.

Figure 5.6, Figure 5.7, and Figure 5.8 present representative time series comparisons for three stations that typify differing transport regimes and source proximities, namely NOX49 in Norway, SEX63 in Sweden, and USX79 in the United States. The plots demonstrate that the nudged simulations align more closely with observed ^{133}Xe concentrations in both the timing and magnitude of peaks, thereby reducing phase offsets and amplitude discrepancies evident in the free-running model.

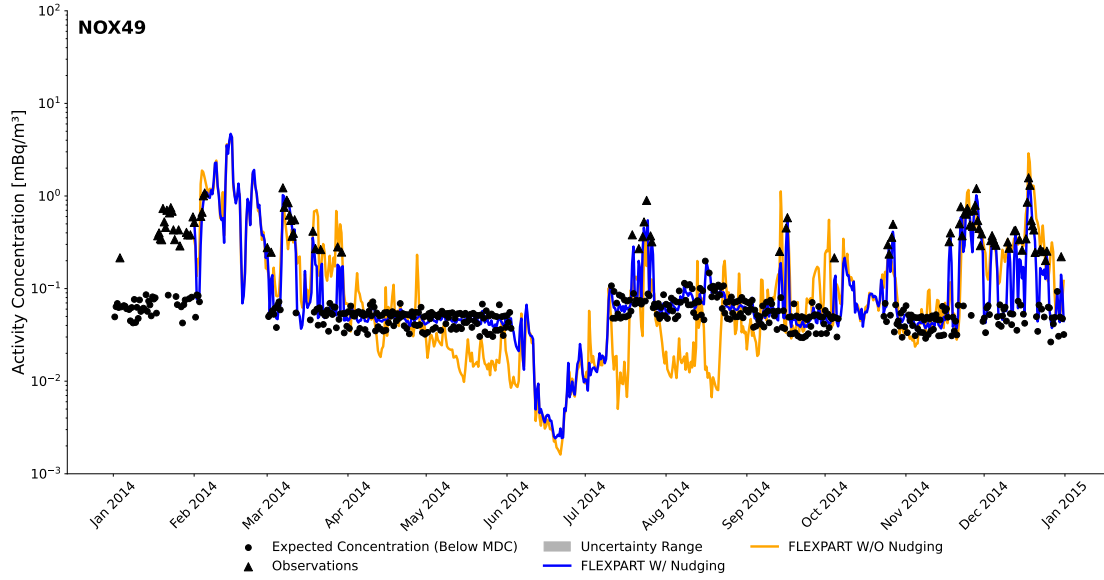


Figure 5.6.: Time series comparison of simulated and observed ^{133}Xe activity concentrations at station NOX49 (Norway).

A systematic assessment across all assimilation stations is provided in Figure 5.9, which displays station-wise diagnostics for bias, mean squared error, and coefficient of determination, and in Table 5.1, which summarises the results. The introduction of nudging reduces the mean bias by 77%, reduces the mean squared error by 88%, and increases the coefficient of determination from 0.627 to 0.834—improvements that demonstrate substantial reduction of both systematic and random errors within the assimilated domain.

5. Results and Discussion

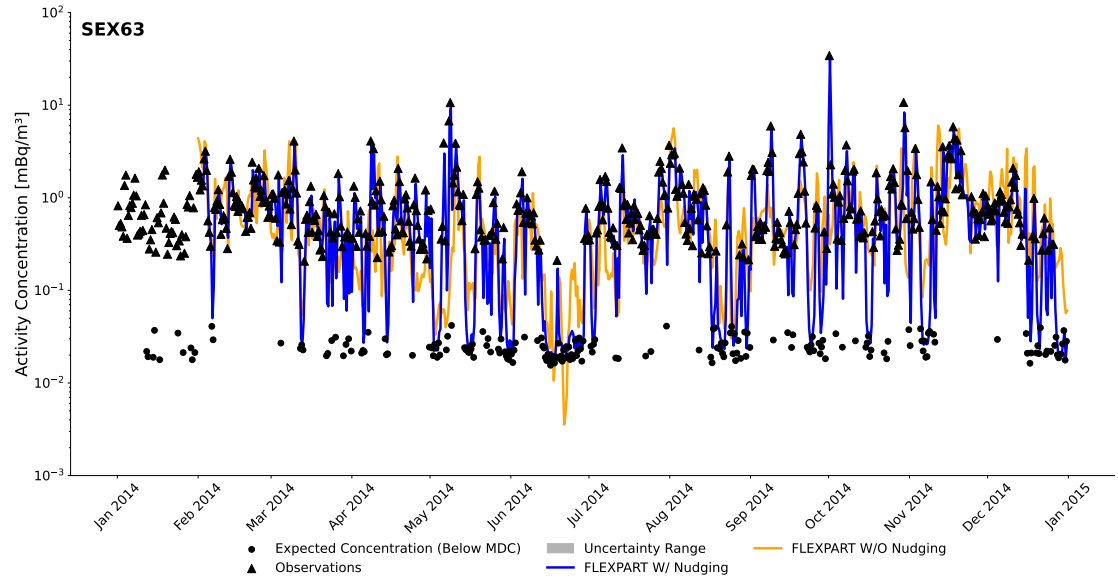


Figure 5.7.: Time series comparison of simulated and observed ^{133}Xe activity concentrations at station SEX63 (Sweden).

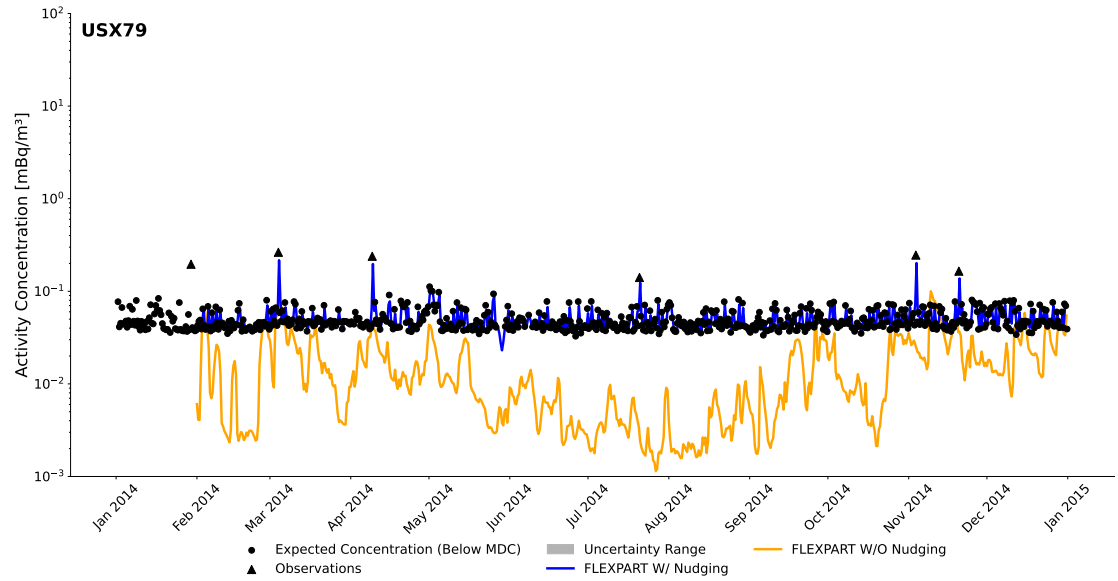


Figure 5.8.: Time series comparison of simulated and observed ^{133}Xe activity concentrations at station USX79 (USA).

5.3. Impact of Nudging at Assimilation Stations

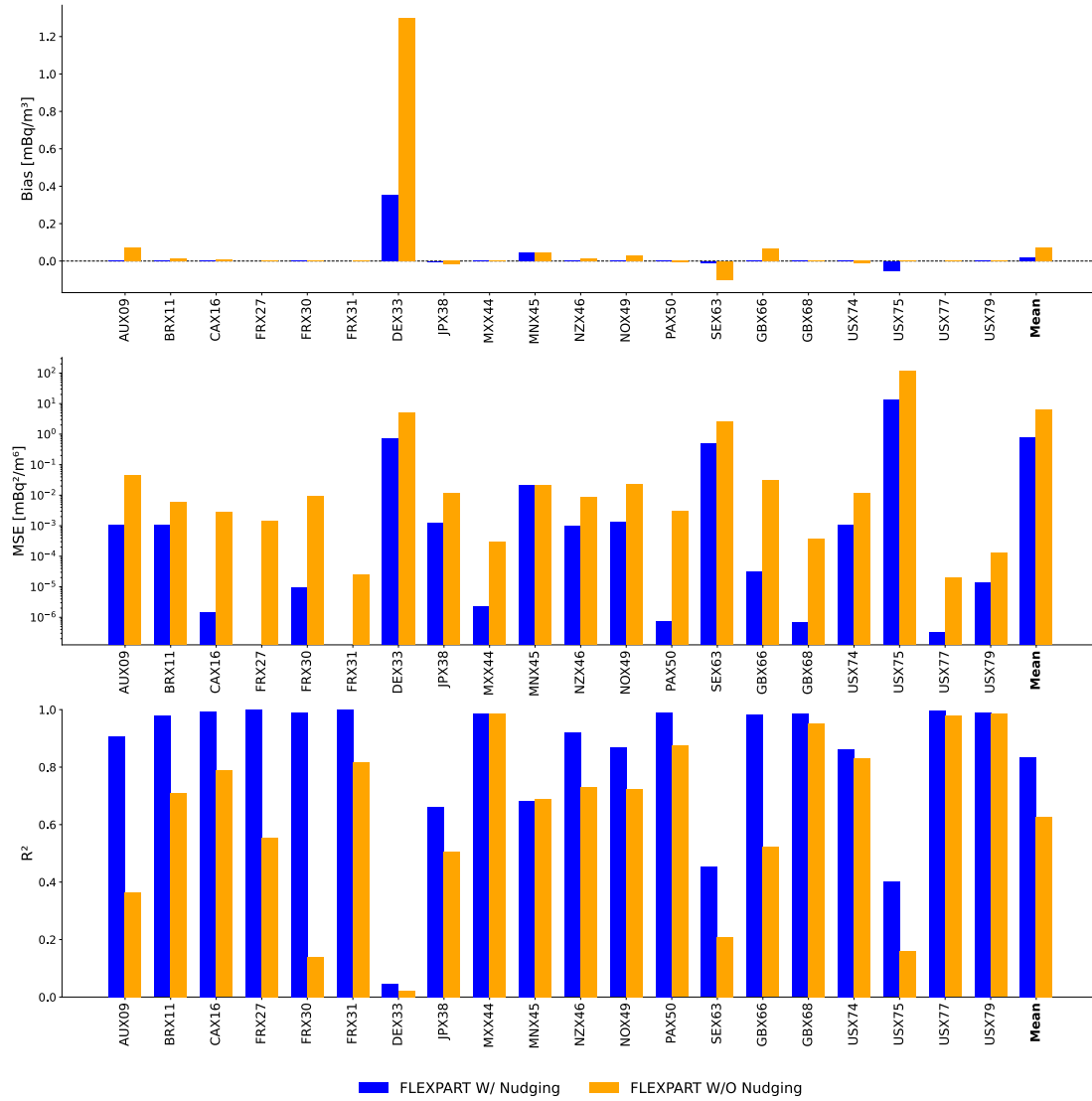


Figure 5.9.: Statistical performance at assimilation stations for the unconstrained record.

Table 5.1.: Mean statistical performance at assimilation stations for the unconstrained record.

Statistical Metric	Without Nudging	With Nudging
Bias (mBq/m³)	0.071	0.016
Mean Squared Error, MSE (mBq²/m⁶)	6.468	0.767
Coefficient of Determination, R^2	0.627	0.834

5. Results and Discussion

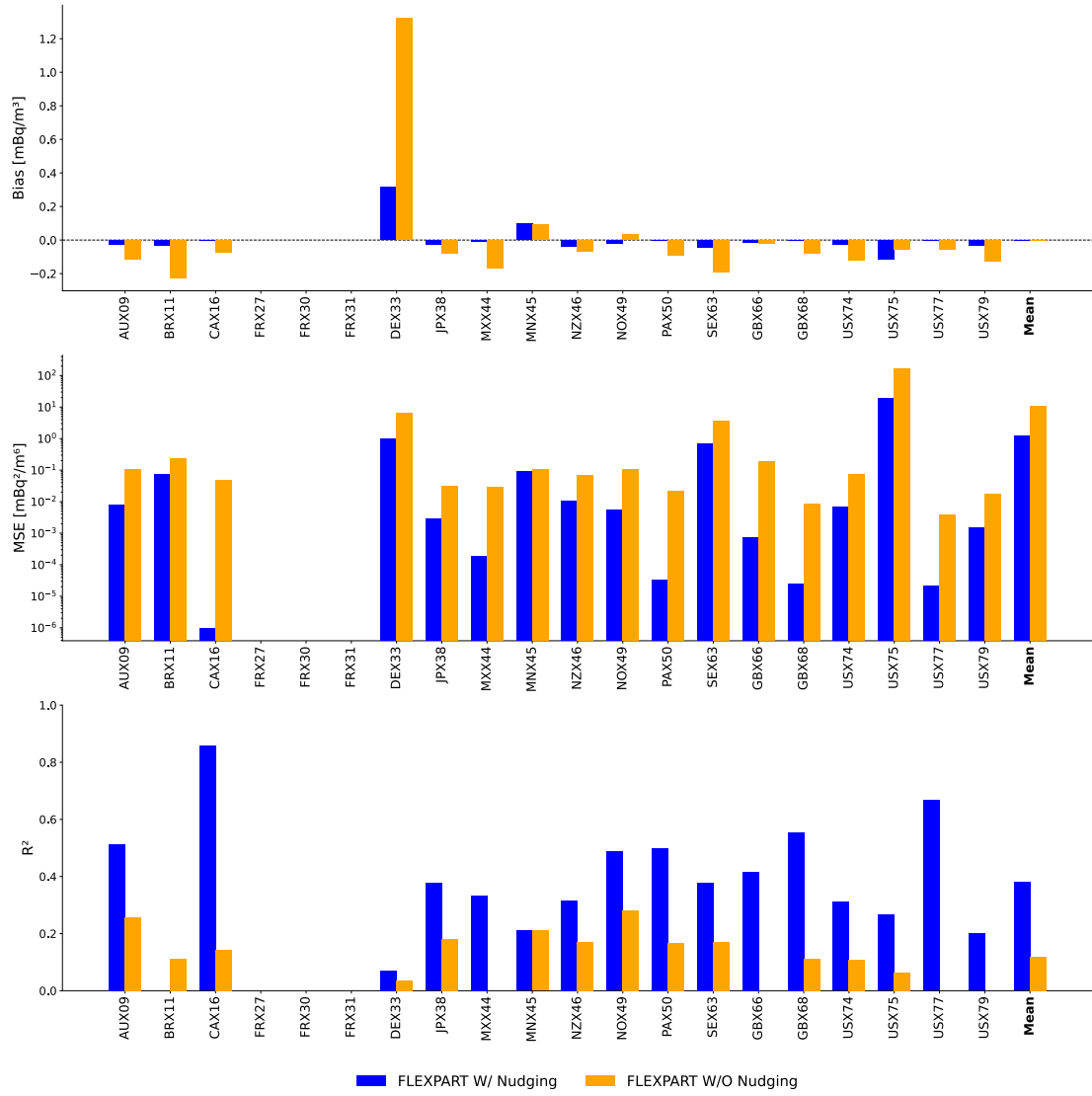


Figure 5.10.: Statistical performance at assimilation stations for the constrained record.

Table 5.2.: Mean statistical performance at assimilation stations for the constrained record.

Statistical Metric	Without Nudging	With Nudging
Bias (mBq/m ³)	-0.001	0.001
Mean Squared Error, MSE (mBq ² /m ⁶)	10.734	1.270
Coefficient of Determination, R^2	0.118	0.380

5.4. Impact of Nudging at Validation Stations

The robustness of the improvements is confirmed by restricting the evaluation to the constrained record. Figure 5.10, together with Table 5.2, present these results. Even under this stricter criterion, nudging reduces the mean squared error by 88% and increases the coefficient of determination from 0.118 to 0.380, thereby reinforcing the conclusion that the nudging configuration meaningfully improves model skill for high-confidence observations.

These results confirm that nudging substantially enhances the capacity of the model to reproduce observed ^{133}Xe background concentrations at assimilation stations and validate the selected nudging configuration with $\lambda = 8$ as effective for correcting local transport and source representation within regions covered by observations, while recognising that such improvements are inherently local and should not be extrapolated without caution to unassimilated regions where no direct observational constraint exists.

The subsequent section examines whether the improvements documented at assimilation stations generalise to independent validation stations withheld from the assimilation procedure, thereby evaluating the transferability of observational constraints beyond the directly constrained domain.

5.4. Impact of Nudging at Validation Stations

To evaluate the spatial generalisability of nudging beyond regions directly constrained by observations, a leave-one-out cross-validation experiment was performed. In each of five simulations, one validation station was withheld from assimilation while the other four validation stations, together with all assimilation stations, were assimilated. This ensured that each validation station was assessed without direct observational forcing and that validation statistics were obtained under conservative and statistically robust conditions appropriate for a globally sparse monitoring network.

Figure 5.11 summarises station-level diagnostics across the five validation stations. The numerical summaries are reported in Tables 5.3 and 5.4. Across the principal skill indicators, the application of nudging results in a small deterioration in performance at validation stations—a pattern that is modest when considering the unconstrained record but more pronounced when the analysis is restricted to the constrained record. For the unconstrained record, the coefficient of determination declines marginally and the mean squared error increases slightly, whereas for the constrained record, the coefficient of determination decreases more substantially and bias shifts further from zero.

5. Results and Discussion

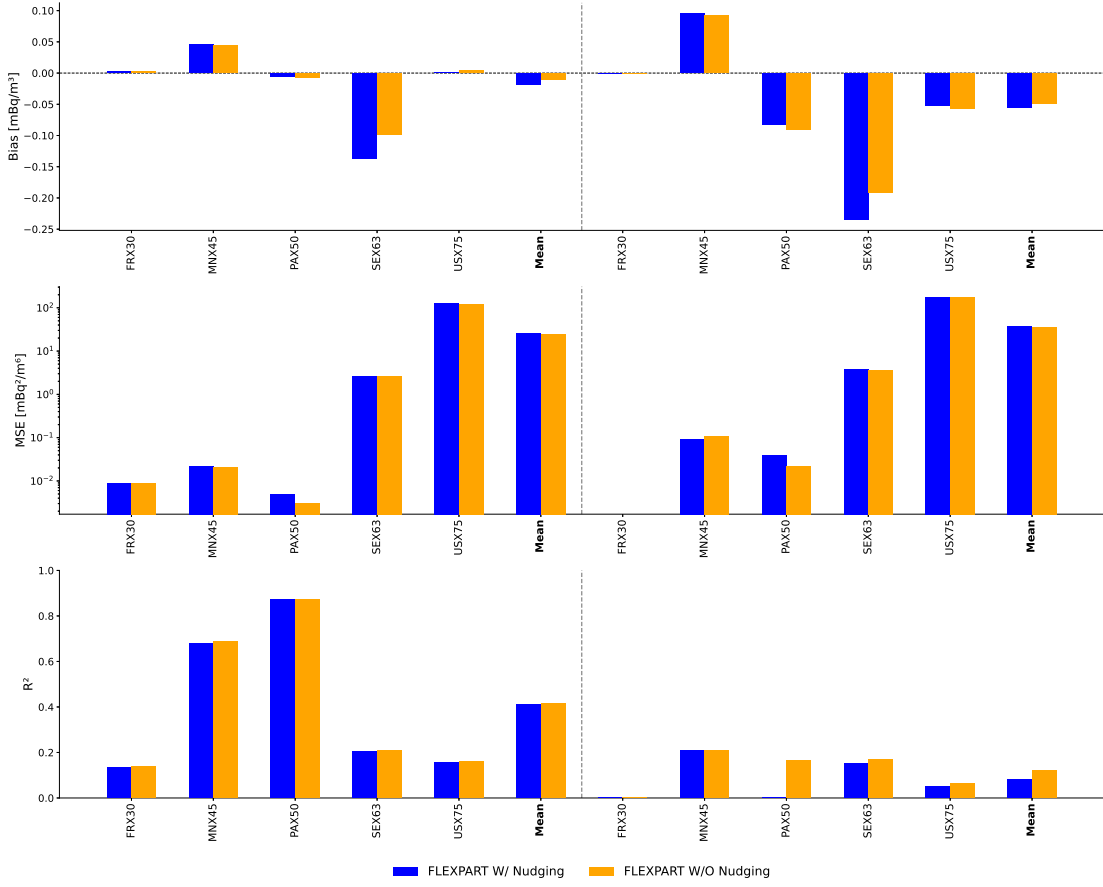


Figure 5.11.: Statistical performance at validation stations for the unconstrained record (left) and the constrained record (right).

Table 5.3.: Mean statistical performance at validation stations for the unconstrained record.

Statistical Metric	Without Nudging	With Nudging
Bias (mBq/m³)	-0.019	-0.011
Mean Squared Error, MSE (mBq²/m⁶)	24.829	25.648
Coefficient of Determination, R^2	0.415	0.409

5.5. Sensitivity Experiment: Increasing Vertical and Horizontal Influence

Table 5.4.: Mean statistical performance at validation stations for the constrained record.

Statistical Metric	Without Nudging	With Nudging
Bias (mBq/m ³)	-0.049	-0.055
Mean Squared Error, MSE (mBq ² /m ⁶)	35.048	36.278
Coefficient of Determination, R^2	0.122	0.083

5.5. Sensitivity Experiment: Increasing Vertical and Horizontal Influence

The preceding validation results reinforce the interpretation that nudging primarily acts as a local correction mechanism for short-lived tracers such as ¹³³Xe. Due to limited atmospheric memory, combined with rapid radioactive decay, the persistence of observationally induced adjustments is constrained, and improvements achieved near assimilation stations do not propagate in a sustained manner to independent stations.

This sensitivity experiment was designed to examine whether enlarging the horizontal and vertical reach of the nudging kernel could mitigate this limitation and extend skill to stations excluded from assimilation. Two controlled modifications to the standard configuration were introduced:

- The vertical kernel extent was increased from 500 to 1,000 metres to enable broader coupling across atmospheric layers.
- The spatial scaling factor was expanded from $\lambda = 8$ to $\lambda = 10$, aiming to enlarge the domain of influence for nudged observations.

Diagnostics at assimilation stations indicate incremental benefit from the expanded kernel scale. Relative to the standard configuration, the mean squared error averaged across assimilation stations decreased from 0.767 to 0.631, and the mean coefficient of determination rose from 0.834 to 0.835, consistent with the anticipated effect of broader kernels on local agreement with observations.

By contrast, the expanded kernel scale does not produce systematic improvements at independent validation stations and in some cases introduces modest deterioration. For example, at USX75, the bias increases from 0.001 to 0.037 and the mean squared error from 125.56 to 126.50, while the coefficient of determination changes only slightly from 0.154 to 0.165. When validation is restricted to the constrained record, mean squared error increases from 177.59 to 178.85, and the coefficient of determination shifts from 0.050 to 0.053. A similar pattern occurs at SEX63, where small adverse changes in bias and mean squared error of order 1–3% offset any marginal gains in the coefficient of determination. These responses confirm that enlarging the kernel footprint does not guarantee improved generalisation beyond the assimilated domain.

5. Results and Discussion

Two principal observations emerge from this experiment:

- (1) The spatial propagation of nudging remains limited even when the kernel is extended vertically and horizontally. Corrections are introduced locally and are rapidly diluted through mixing with surrounding non-nudged air masses as the flow advects them.
- (2) Broader kernels can introduce artefacts at remote stations when their footprints are not congruent with actual transport pathways. Beyond overcorrection and temporal mismatches, large kernels can also destroy real small-scale ^{133}Xe gradients that FLEXPART-LCM may already reproduce correctly without nudging, by smoothing physically meaningful contrasts. Avoiding such distortion would require smaller kernels. However, this simultaneously weakens the nudging impact to the point of limited utility under sparse observational coverage.

5.5.1. Limited Propagation of Nudging Influence

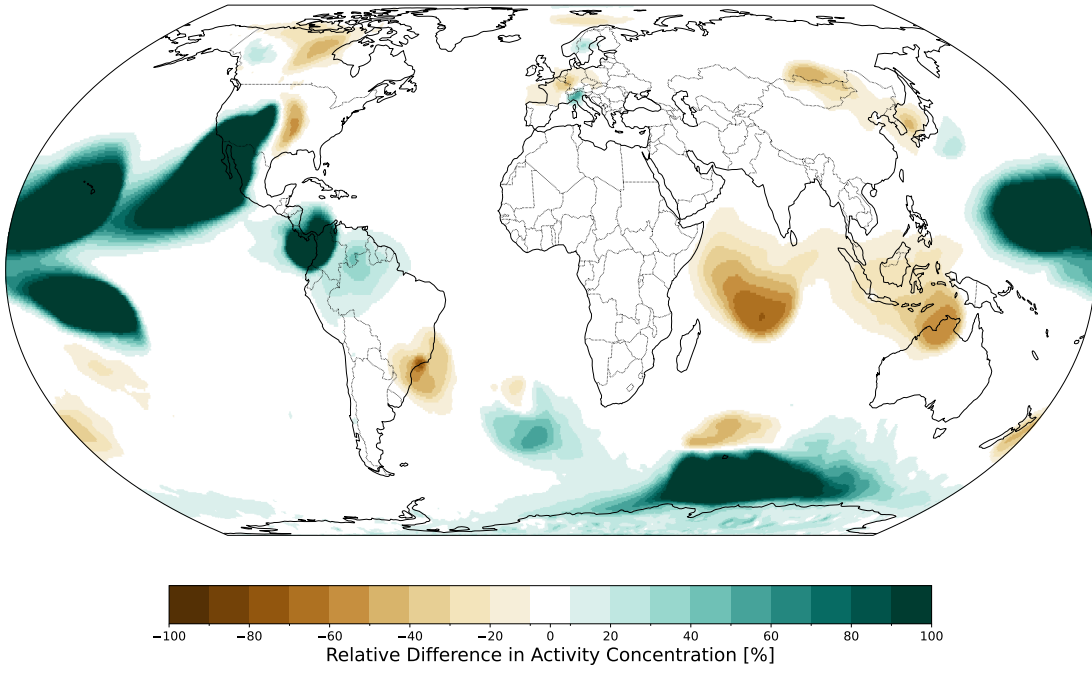
Although ^{133}Xe emissions are transported globally through atmospheric circulation, the influence of nudging remains spatially confined, as illustrated in Figure 5.12. This discrepancy arises as nudging operates within a finite influence radius defined by the kernel geometry. While it can correct concentrations near an assimilation station, these corrections are not inherently self-sustaining. As the tracer is advected downwind, it mixes with surrounding air masses uninfluenced by observational constraints. Over time and distance, the corrected signal is increasingly diluted, particularly in regions with sparse observational coverage. Moreover, varying wind patterns mean the nudging influence does not consistently follow a single trajectory and tends to average out over longer temporal scales.

This behaviour is illustrated in Figure 5.13. During the representative nine-day period from 1 to 9 July 2014, nudging-induced relative differences propagated coherently along dominant flow paths. However, when averaged over the entire year, these structures become diffuse and largely disappear—except in regions where persistent wind systems continuously channel the nudging effect in a preferred direction.

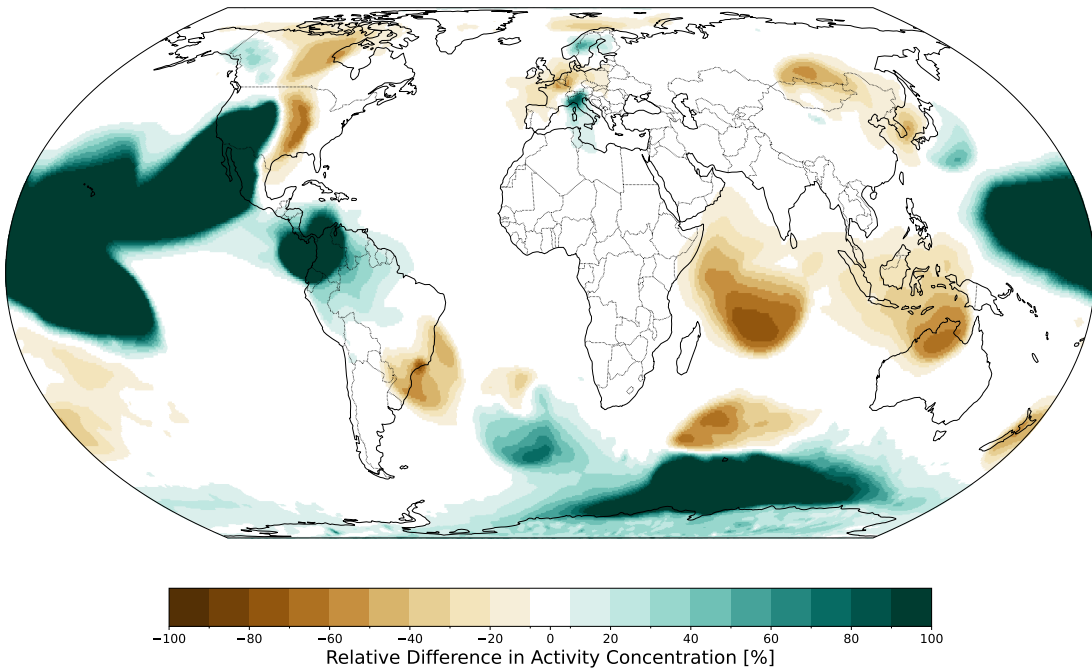
5.5.2. Degraded Performance at Validation Stations

The degraded performance at validation stations, USX75 in this case, may initially appear counterintuitive, given that the expanded kernel was intended to improve generalisation. Several mechanisms may account for this behaviour:

5.5. Sensitivity Experiment: Increasing Vertical and Horizontal Influence



(a)



(b)

Figure 5.12.: Mean relative difference in activity concentration for 2014 under a) the standard configuration and b) the expanded kernel configuration.

5. Results and Discussion

- **Gradient smoothing and signal dilution:** Wider kernels blur genuine atmospheric ^{133}Xe gradients, especially when the nudging strength is insufficiently tapered with distance. This homogenisation can bias concentrations and increase mean squared error at validation stations. The only robust mitigation is to reduce kernel size. However, smaller kernels undermine the effectiveness of nudging under a sparse measurement network, creating an inherent trade-off.
- **Overcorrection in dynamically decoupled regions:** Corrections are applied irrespective of meteorological connectivity. Adjustments upstream of USX75 may not represent the actual air mass influencing the station.
- **Incoherence with transport pathways:** Nudging assumes spatial proximity implies influence. In practice, stations separated by modest distances may lie on unrelated flow trajectories, so corrections can add noise rather than benefit.

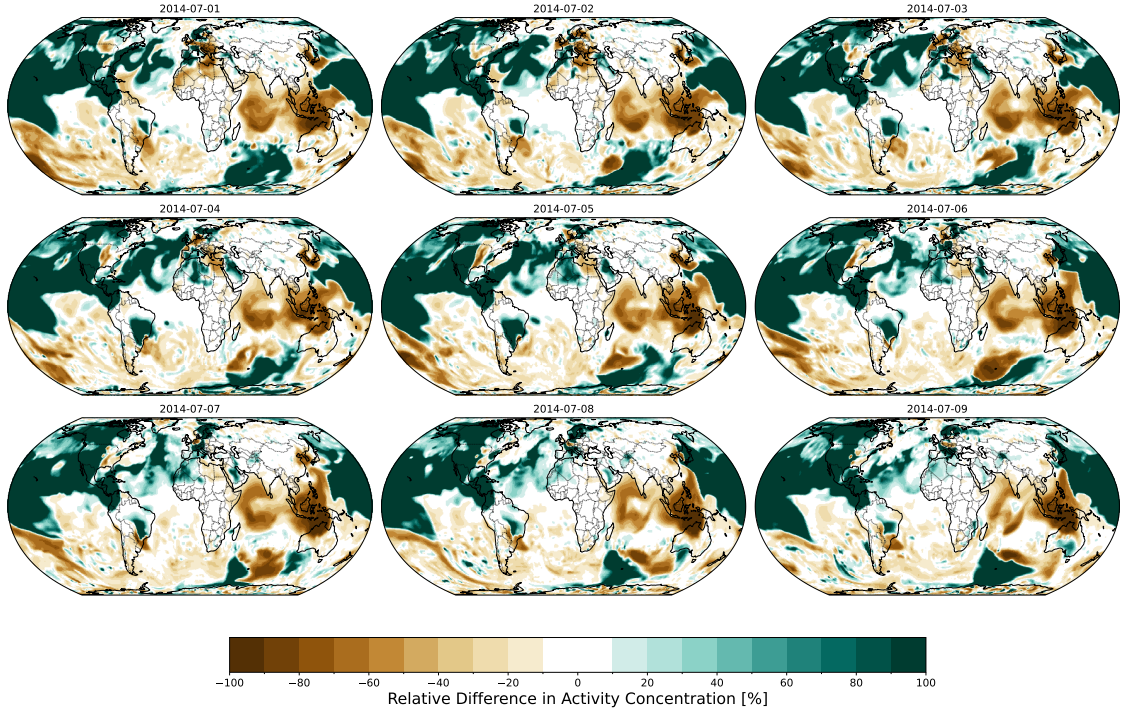


Figure 5.13.: Mean relative difference in activity concentration for 1–9 July 2014.

Taken together, these findings demonstrate that while broader kernels can yield incremental local improvements, they do not resolve the fundamental limitation imposed by tracer persistence and sparse station geometry. Nudging is intrinsically effective at reducing errors where observations are available, but its broader utility for ^{133}Xe is restricted by short lifetime, transport incoherence, and the lack of dense observational coverage. These results underscore that the apparent weakness of nudging at validation stations does not stem from a deficiency of the method itself, but from the physical characteristics of the tracer and the constraints of the monitoring system.

5.6. Sensitivity Experiment: Increasing the Half-Life of ^{133}Xe

This experiment isolates the role of radioactive persistence from the combined effects of station geometry and observational sampling. The model settings and nudging configuration were held constant, and only the tracer decay constant was modified by doubling its half-life. The objective is to demonstrate that the limited large-scale impact observed for ^{133}Xe arises from its short lifetime rather than from deficiencies in nudging. For this configuration, the influence increment, defined as analysis minus baseline, and the relative influence were computed. Binary masks based on a 5% relative influence threshold were applied to identify grid cells where nudging produces a material adjustment to concentrations. The resulting masks are shown in Figure 5.14.

At the 5% threshold, the domain influenced under the doubled half-life expands by approximately 104% relative to the standard configuration. The limited spatial overlap between the masks suggests that increased persistence extends the lateral reach and penetration depth of corrections, rather than merely amplifying the same locations. This pattern is consistent with a regime in which tracer persistence constrains the effective propagation of observational corrections for the given station geometry.

This experiment is a diagnostic construct and is not intended to improve realism for ^{133}Xe . Nevertheless, the observed expansion shows that nudging is intrinsically effective at propagating corrections when the tracer persists. The limited improvement in global background concentration fields for ^{133}Xe , therefore, reflects its short half-life, rather than an inherent weakness of nudging. By analogy, isotopes with longer half-lives, such as $^{131\text{m}}\text{Xe}$, would be expected to exhibit a larger effective area of influence under the same assimilation configuration.

5. Results and Discussion

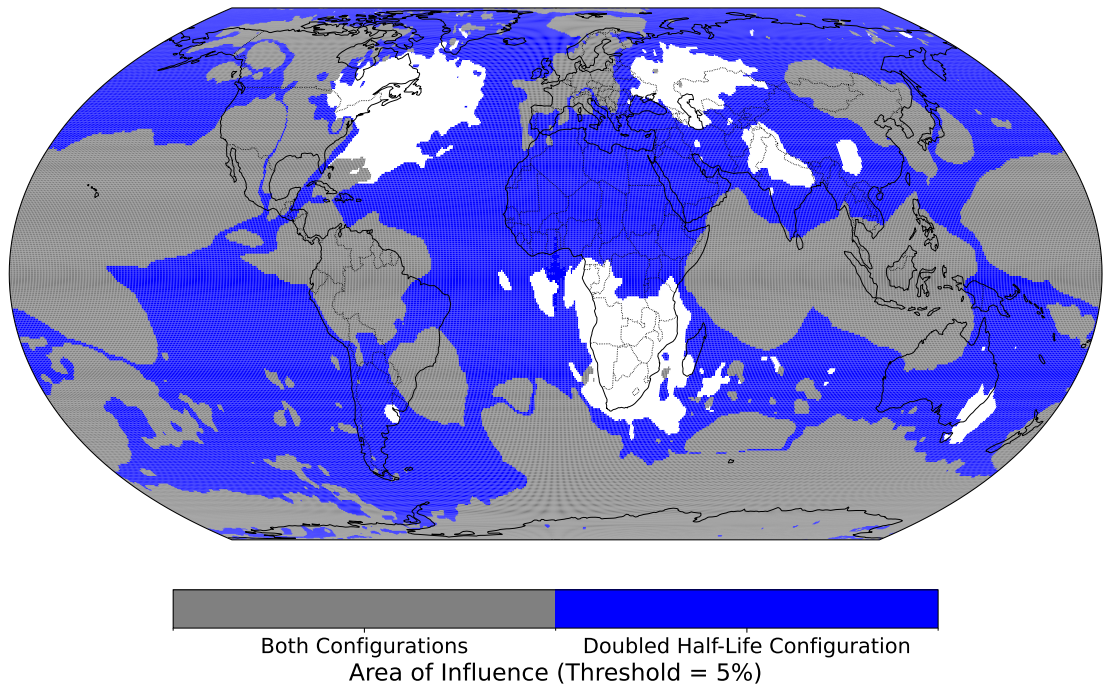


Figure 5.14.: Grid cells where the relative influence exceeds 5% for either configuration. Grey indicates overlap, blue indicates regions influenced only under the doubled half-life configuration. Cells below the threshold in both cases remain white.

6. Conclusion

The objective of this master’s thesis was to implement spatially and temporally adaptive nudging in FLEXPART-LCM and to systematically evaluate its capacity to improve global ^{133}Xe background concentration fields. A suite of sensitivity experiments was employed to delineate both the strengths and the mechanistic limits of kernel-based nudging for a short-lived radionuclide.

Nudging delivered robust local improvements. At assimilation stations, bias was reduced by 77% and mean squared error by 88%, and the coefficient of determination increased from 0.627 to 0.834. An intermediate horizontal kernel ($\lambda = 8$) offered the best balance of statistical skill and physical plausibility, improving performance at assimilation stations without evident non-physical artefacts, but with slight degradations at independent validation stations.

Any improvements, however, remained spatially confined. Under the present station geometry, nudging functions primarily as a station-level correction. The skill decays rapidly outside observational footprints, and independent validation stations show limited or no benefit. Enlarging horizontal and vertical kernel scales produced only marginal additional improvements at assimilation stations and did not reliably transfer skill to withheld locations. The results, therefore, underscore that kernel scales alone cannot substitute for observational representativeness or dynamical consistency.

The controlled persistence experiment, which doubled the half-life of the tracer, demonstrated that the radioactive lifetime strongly regulates the downstream propagation of observational corrections. At a 5% threshold, the influenced domain expanded by approximately 104%. This indicates that longer-lived xenon isotopes, such as $^{131\text{m}}\text{Xe}$, are intrinsically more amenable to broad-scale nudging under the same station configuration.

Five factors principally limit the broader applicability of nudging:

- The short half-life of ^{133}Xe constrains temporal persistence and shortens the effective spatial memory of the system.
- Emissions are spatially heterogeneous and clustered, while observational coverage is sparse, restricting the fraction of the domain that can be directly constrained.
- Excessive kernel scales risk smoothing physically meaningful gradients and misaligning corrections with transport pathways, yielding spurious adjustments.

6. Conclusion

- Detection limits, coupled with generally low and often well-contained civilian emissions, result in a paucity of reliably quantifiable observations, leaving limited potential for nudging to have a sustained impact.
- The remoteness of many observation stations means sampling often occurs late along the transport pathway. Thus, concentrations are frequently at or below the MDC, or only marginally above it. In the former case, nudging has essentially no leverage because the simulations and observations agree on low concentrations. In the latter, any correction decays rapidly below the background before reaching downstream stations.

In conclusion, nudging within FLEXPART-LCM is an effective instrument for reducing local errors in ^{133}Xe background concentrations at assimilation stations. Its capacity to generate transferable improvements at continental scales is, however, constrained by tracer persistence, station geometry, transport coherence, and the availability of high-quality observations. Addressing these constraints through a combination of (i) continued improvements to monitoring systems to lower detection limits, (ii) strategic densification of the network along recurrent transport corridors, (iii) leveraging the advantage of longer-lived species such as $^{131\text{m}}\text{Xe}$, and (iv) employing complementary assimilation methods offers a clear path to extend the demonstrated benefits. In this spirit, the CTBTO is encouraged to prioritise these pragmatic steps, thereby improving the probability of detecting weak releases and ensuring that non-detections are interpreted with appropriate caution in support of robust verification.

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A. Appendix: Nuclear Power Plants

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
Argentina	Atucha-1	-33.96	-59.20	3.41E+12
Argentina	Embalse	-32.23	-64.44	1.70E+12
Armenia	Armenian	40.18	44.14	1.70E+12
Belgium	Doel	51.32	4.25	6.83E+12
Belgium	Tihange	50.53	5.27	5.11E+12
Brazil	Angra	-23.00	-44.47	3.41E+12
Bulgaria	Kozloduy	43.74	23.77	3.29E+10
Canada	Bruce	44.32	-81.59	1.36E+13
Canada	Darlington	43.87	-78.71	6.83E+12
Canada	Pickering	43.81	-79.06	1.02E+13
Canada	Point Lepreau	45.06	-66.45	4.81E+12
China	Daya Bay	22.59	114.54	3.41E+12
China	Fuqing-1	25.44	119.44	1.70E+12
China	Hongyanhe	39.79	121.47	3.41E+12
China	Ling AO	22.69	114.55	6.82E+12
China	Ningde	27.04	120.28	3.41E+12
China	Qinshan	30.43	120.95	1.19E+13
China	Tianwan	34.68	119.45	3.41E+12
China	Yangjiang-1	21.70	112.26	1.70E+12
Czechia	Dukovany	49.08	16.14	5.97E+10
Czechia	Temelin	49.18	14.37	4.03E+12
Finland	Loviisa	60.37	26.34	1.05E+11
Finland	Olkiluoto	61.23	21.44	2.27E+12
France	Belleville	47.50	2.87	3.89E+11
France	Blayais	45.25	-0.69	2.03E+12
France	Bugey	45.79	5.27	9.85E+11
France	Cattenom	49.41	6.21	2.42E+12
France	Chinon	47.23	0.17	6.21E+12
France	Chooz	50.09	4.78	2.34E+11
France	Civaux	46.45	0.65	8.75E+11
France	Cruas	44.63	4.75	5.35E+11
France	Dampierre	47.73	2.51	8.75E+11
France	Fessenheim	47.90	7.56	7.49E+10

A. Appendix: Nuclear Power Plants

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
France	Flamanville	49.53	-1.88	2.54E+11
France	Golfech	44.10	0.84	1.62E+12
France	Gravelines	51.01	2.13	3.00E+12
France	Nogent	48.51	3.51	2.12E+11
France	Paluel	49.85	0.63	4.53E+07
France	Penly	49.97	1.21	5.65E+11
France	Saint-Alban	45.40	4.75	4.22E+11
France	Saint-Laurent	47.72	1.57	3.79E+12
France	Tricastin	44.32	4.73	2.42E+11
Germany	Brokdorf	53.85	9.34	2.40E+11
Germany	Emsland	52.47	7.31	2.34E+10
Germany	Grafenrheinfeld	49.98	10.18	5.70E+11
Germany	Grohnde	52.03	9.41	5.50E+12
Germany	Gundremmingen	48.51	10.40	4.60E+11
Germany	Isar-2	48.60	12.29	7.70E+07
Germany	Neckarwestheim-2	49.04	9.17	1.60E+08
Germany	Philippsburg-2	49.25	8.43	7.00E+08
Hungary	Paks	46.57	18.85	4.63E+12
India	Kakrapar	21.23	73.35	3.41E+12
India	Kalpakkam/Madras	12.55	80.17	3.41E+12
India	Kaiga	14.86	74.43	6.81E+12
India	Kudankulam-1	8.16	77.71	1.70E+12
India	Narora	28.15	78.40	3.41E+12
India	Rajasthan	24.87	75.61	1.02E+13
India	Tarapur	19.82	72.66	6.83E+12
Iran	Bushehr	28.82	50.88	1.70E+12
Mexico	Laguna Verde	19.72	-96.40	3.41E+12
Netherlands	Borssele	51.43	3.71	1.16E+11
Pakistan	Chasnupp	32.39	71.46	3.41E+12
Pakistan	Kanupp	24.84	66.78	1.70E+12
Romania	Cernavoda	44.32	28.05	3.41E+12
Russia	Balakovo	52.09	47.95	6.83E+12
Russia	Beloyarsk-3	56.84	61.32	1.70E+12
Russia	Bilibino	68.05	166.53	6.83E+12
Russia	Kalinin	57.90	35.06	6.81E+12
Russia	Kola	67.46	32.46	6.83E+12
Russia	Kursk	51.67	35.60	6.83E+12
Russia	Leningrad	59.84	29.04	6.83E+12
Russia	Novovoronezh	51.27	39.20	5.11E+12
Russia	Rostov	47.59	42.37	3.41E+12
Russia	Smolensk	54.16	33.24	5.11E+12

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
Slovakia	Bohunice	48.49	17.68	6.55E+10
Slovakia	Mochovce	48.26	18.45	1.68E+10
Slovenia	Krsko	45.93	15.51	7.64E+10
South Africa	Koeberg	-33.67	18.43	3.41E+12
South Korea	Kori	35.32	129.29	3.11E+12
South Korea	Shin-Kori/Saeul	35.33	129.31	3.11E+12
South Korea	Shin-Wolsong	35.72	129.47	1.59E+13
South Korea	Uljin/Hanul	37.09	129.38	3.21E+10
South Korea	Wolsong	35.71	129.47	1.60E+13
South Korea	Yonggwang/Hanbit	35.40	126.41	6.78E+11
Spain	Almaraz	39.80	-5.69	1.26E+12
Spain	Asco	41.20	0.56	1.44E+13
Spain	Cofrentes	39.21	-1.05	7.37E+11
Spain	Trillo-1	40.70	-2.62	3.38E+10
Spain	Vandelos-2	40.95	0.86	2.88E+08
Sweden	Forsmark	60.40	18.16	1.82E+12
Sweden	Oskarshamn	57.41	16.67	7.28E+11
Sweden	Ringhals	57.25	12.11	1.38E+11
Switzerland	Beznau	47.55	8.22	2.23E+12
Switzerland	Goesgen	47.36	7.96	3.70E+12
Switzerland	Leibstadt	47.60	8.18	1.16E+12
Switzerland	Muehleberg	46.96	7.26	2.90E+11
Taiwan	Chinshan	25.28	121.58	3.41E+12
Taiwan	Kuosheng	25.20	121.66	3.41E+12
Taiwan	Maanshan	21.95	120.75	3.41E+12
Ukraine	Khmelnitski	50.30	26.64	3.41E+12
Ukraine	Rovno	51.32	25.89	6.83E+12
Ukraine	South Ukraine	47.81	31.21	5.11E+12
Ukraine	Zaporozhye	47.51	34.58	1.02E+13
United Kingdom	Dungeness	50.91	0.96	3.41E+12
United Kingdom	Hartlepool	54.63	-1.18	4.00E+12
United Kingdom	Heysham	54.02	-2.91	8.00E+12
United Kingdom	Hinkley Point	51.20	-3.12	4.00E+09
United Kingdom	Hunterston	55.72	-4.89	3.41E+12
United Kingdom	Sizewell B	52.215	1.61	1.70E+12
United Kingdom	Torness	55.96	-2.40	3.41E+12
United Kingdom	Wylfa-1	53.41	-4.48	1.70E+12
USA	Arkansas One	35.31	-93.23	1.74E+09
USA	Beaver Valey	40.62	-80.43	7.59E+08
USA	Braidwood	41.24	-88.22	8.62E+09
USA	Browns Ferry	34.70	-87.11	9.14E+12

A. Appendix: Nuclear Power Plants

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
USA	Brunswick	33.95	-78.01	2.39E+09
USA	Byron	42.07	-89.28	3.23E+10
USA	Calvert Cliffs	38.43	-76.44	3.43E+11
USA	Callaway-1	38.76	-91.78	2.22E+09
USA	Catawba	35.05	-81.07	6.51E+10
USA	Clinton-1	40.17	-88.83	8.58E+09
USA	Columbia	46.47	-119.33	1.70E+12
USA	Comanche Peak	32.29	-97.78	1.94E+09
USA	Cooper	40.36	-95.64	4.54E+09
USA	Davis Besse-1	41.59	-83.08	2.55E+10
USA	Diablo Canyon	35.21	-120.85	1.08E+11
USA	Donald Cook	41.97	-86.56	9.34E+09
USA	Dresden	41.38	-88.26	5.99E+11
USA	Duane Arnold-1	42.10	-91.77	3.41E+11
USA	Enrico Fermi-2	41.96	-83.25	6.30E+11
USA	Farley	31.22	-85.11	5.43E+09
USA	Fitzpatrick	43.52	-76.39	1.61E+11
USA	Fort Calhoun-1	41.52	-96.07	2.78E+10
USA	Grand Gulf-1	32.00	-91.04	4.07E+12
USA	Hatch	31.93	-82.34	1.01E+11
USA	H.B. Robinson-2	34.40	-80.15	3.13E+09
USA	Hope Creek-1	39.46	-75.53	7.38E+10
USA	Indian Point	41.26	-73.95	1.26E+10
USA	Limerick	40.22	-75.58	1.41E+12
USA	Lasalle	41.24	-88.66	4.26E+13
USA	Mcguire	35.43	-80.94	1.25E+10
USA	Millstone	41.31	-72.16	2.82E+09
USA	Monticello	45.33	-93.84	6.57E+12
USA	Nine Mile Point	43.52	-76.40	1.08E+10
USA	North Anna	38.06	-77.78	1.01E+11
USA	Oconee	34.79	-82.89	1.54E+12
USA	Oyster Creek	39.81	-74.20	1.75E+11
USA	Palisades	42.32	-86.31	7.78E+10
USA	Paloverde	33.38	-112.86	4.11E+10
USA	Peach Bottom	39.75	-76.26	7.25E+10
USA	Perry-1	41.80	-81.14	1.97E+12
USA	Pilgrim-1	41.94	-70.57	3.07E+10
USA	Point Beach	44.28	-87.53	3.23E+10
USA	Prairie Island	44.62	-92.63	8.62E+07
USA	Quad Cities	41.72	-90.31	2.69E+11
USA	R.E. Ginna	43.27	-77.31	1.08E+11

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
USA	River Bend-1	30.75	-91.33	4.29E+10
USA	Salem	39.46	-75.53	8.41E+09
USA	Sequoyah	35.22	-85.09	4.60E+11
USA	Shearon Harris-1	35.63	-78.95	1.37E+10
USA	South Texas	28.79	-96.04	7.19E+10
USA	St. Lucie	27.34	-80.24	5.80E+11
USA	Susquehanna	41.08	-76.14	1.19E+10
USA	Surry	37.16	-76.69	7.60E+09
USA	Three Mile Island-1	40.15	-76.72	2.53E+09
USA	Turkey Point	25.43	-80.33	3.72E+10
USA	Vermont Yankee	42.77	-72.51	1.99E+09
USA	Virgil C. Summer-1	34.29	-81.31	5.48E+09
USA	Vogtle	33.14	-81.76	6.42E+09
USA	Waterford-3	29.99	-90.47	3.09E+12
USA	Watts Bar-1	35.60	-84.78	2.05E+09
USA	Wolf Creek	38.23	-95.68	9.66E+11
USA	Yankee Rowe	42.70	-72.90	2.32E+08

B. Appendix: Nuclear Research Reactors

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
Australia	OPAL	-34.05	150.97	4.94E+11
Belgium	BR-2	51.21	5.09	6.22E+10
Canada	NRU	46.05	-77.36	4.50E+14
China	CARR	39.74	116.14	1.28E+10
China	CEFR	39.74	116.03	1.39E+10
China	HFETR	29.73	103.55	4.67E+10
Czechia	LVR-15 Rež	50.17	14.36	9.33E+09
France	ILL High Flux Reactor	45.20	5.69	5.80E+10
France	OSIRIS	48.72	2.15	8.30E+09
France	ORPHEE	48.72	2.15	6.76E+09
Germany	BER-II	52.41	13.12	8.96E+09
India	DHRUVA	19.01	72.89	1.62E+11
India	FBTR	12.55	80.17	3.11E+10
Indonesia	RSG-GAS	-6.35	106.66	1.09E+10
Israel	IRR-2	31.00	35.14	5.57E+09
Norway	HBWR	59.12	11.40	1.74E+10
Poland	MARIA	52.12	21.34	1.48E+10
Russia	BOR-60	54.20	49.52	5.78E+10
Russia	IVV-2M	56.84	61.32	1.14E+10
Russia	MIR.M1	54.18	49.47	5.52E+10
Russia	RBT-10/2	54.20	49.52	3.86E+09
Russia	RBT-6	54.20	49.52	3.72E+09
Russia	SM-3	54.18	49.47	1.12E+11
Russia	VK-50	54.18	49.47	2.55E+11
South Africa	SAFARI-1	-25.80	27.95	2.74E+10
South Korea	HANARO	36.42	127.37	3.00E+10
Ukraine	WWR-M Kiev	50.38	30.53	5.55E+09
USA	ATR	43.58	-112.96	4.04E+11
USA	HFIR	35.91	-84.30	1.92E+11
USA	MITR-II	42.36	-71.09	7.46E+09
USA	MURR	38.93	-92.34	1.39E+10

C. Appendix: Medical Isotope Production Facilities

Country	Facility Name	Latitude (°)	Longitude (°)	Emissions (Bq/Year)
Argentina	CNEA	-34.82	-58.57	2.47E+11
Australia	ANSTO	-34.05	150.98	2.36E+12
Belgium	IRE	50.48	4.68	1.51E+12
Canada	CNL	46.05	-77.36	7.82E+15
Canada	Nordion	45.34	-75.92	4.11E+10
China	HFETR	30.51	104.03	1.00E+12
Indonesia	PT BaTek	-6.30	106.70	1.04E+10
Netherlands	Mallinckrodt	52.78	4.68	2.00E+09
Pakistan	PINSTECH PARR-1	33.65	73.26	1.00E+12
Russia	Karpov Institute	55.08	36.57	8.25E+11
Russia	NIIAR	54.19	49.48	5.50E+12
South Africa	NTP/NECSA	-25.80	27.95	6.3E+13
USA	LMI	42.55	-71.31	3.14E+07