

Uncertainty and source term reconstruction with environmental air samples

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ABSTRACT

Environmental air sampling is one of the principal monitoring technologies employed for the verification of the Comprehensive Nuclear-Test-Ban Treaty (CTBT). By combining the analysis of environmental samples with Atmospheric Transport and Dispersion Modelling (ATDM), and using a Bayesian source reconstruction algorithm, an estimate of the release location, duration, and quantity can be computed.

Bayesian source reconstruction uses an uncertainty distribution of the input parameters, or priors, in a statistical framework to produce posterior probability estimates of the event parameters. The quality of the event reconstruction directly depends on the accuracy of the prior uncertainty distribution. With many of the input parameters, the selection of the uncertainty distribution is not difficult. However, with environmental samples, there is one component of the uncertainty at the interface between sample measurements and the ATDM that has been overlooked. Typically, a much smaller volume or quantity of material is sampled from the much larger domain represented in the ATDM. By examining the response of a dense network of radionuclide detectors on the West Coast of Canada during the passage of the Fukushima debris plume, an initial estimate of this uncertainty was determined to be between 20% and 30% depending on sample integration time.

1. Introduction

The collection of environmental samples (either particulate aerosol collection or noble gas samples) in support of monitoring programs such as the CTBT, which bans the testing of nuclear weapons, poses several important questions when an observation of anthropogenic radionuclides occurs. Questions such as: Where was/is the source located? What was the nature of the activity that caused the release of material? How much radioactive material was released? What was the release or emission period? Are important to decision makers in formulating an effective response to any nuclear event. Source reconstruction (Meutter and Hoffman, 2020; Yee et al., 2014) using a stochastic framework, such as Bayesian inference, is a technique that can provide answers in the form of probabilistic estimates to these questions, and is now being applied to problems involving long range transport and sparse sensor networks. An important requirement of stochastic reconstructions is an accurate model of both the model parameters and the measurement system uncertainties.

One challenge with examining the uncertainties in these environmental samples is that it may not be fully clear how to apportion uncertainty to particular sources. Furthermore, some uncertainty sources

are difficult to assess (e.g. the radioactive material release mechanism, fractionation of material through atmospheric transport and dispersion, wet and dry deposition, preferential airflow through the sampling apparatus itself, etc.). Regardless of the uncertainty source, it is important to be clear about what uncertainty is being characterized (model or measurement system) and how it is used in the source reconstruction process so as to not account for it multiple times in a reconstruction algorithm.

Previous studies of sample collection uncertainty have examined the aerosol collection process, where evidence of inhomogeneity was present on the aerosol filter media (Gomez et al., 2014; Woods et al., 2013; Zhang et al., 2011)–(Gomez et al., 2014; Woods et al., 2013; Zhang et al., 2011). In studies of Fukushima debris, the authors found that ¹³⁷Cs had an underestimated uncertainty due to sample inhomogeneity of between 3% and 10.5% (Gomez et al., 2014). In these studies, sample inhomogeneity uncertainty has been viewed as caused by a combination of the particle size distribution along with inhomogeneous airflow through the sampler.

This physical process view however, misses an important element of uncertainty for source reconstruction that arises from the use of models in the interpretation of environmental data. Some aspects of

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reconstruction uncertainties related to modelling have already been examined (De Meutter et al., 2021; Dumont Le Brazidec et al., 2021), however no study has examined how representative the collected sample is compared to the model domain represented by the ATDM. ATDM models assume a level of homogeneity over a model domain that is much larger than the volume of the collected sample. Sample collection, both aerosol and ambient, involves acquiring or measuring a small volume of air that is assumed to represent a much larger volume. In practice, when using a sample measurement in an environmental study involving ATDM, it is assumed that the sample is representative of an entire grid box of the ATDM domain. This paper focuses on this interface between the ATDM and its environmental interpretation. While it is related to the sample collection process, the measurement/model interface has not yet been characterized.

Current high-resolution ATDM for meso- to continental- scale is typically done at 0.1° horizontal resolution (approximately 10 km) and the first level of the model is 500 m. If the averaging time or model output is hourly, the enclosed volume of the grid box at mid-latitude is approximately $5 \times 10^{10} \text{ m}^3$ before taking into account advection-diffusion processes during the sample collection period. For the sample collection process, a typical Canadian CTBT aerosol sampler draws around 20 000 m^3 of air each sample, while a current radioxenon CTBT samplers collect approximately 7 m^3 in 12 h (Ringbom et al., 2003) or 80 m^3 over a 24 h (Fontaine et al., 2004) period depending on the design (more recent systems collect 40 m^3 in an 8 h period (Le Petit et al., 2015)). From this very rough comparison alone, it is clear the assumption that the sample collected is not representative statistically to reflect the characteristics of the modelled volume, which is nine orders of magnitude larger than the sample. Since it is not practical to acquire a fully representative sample, there is an additional source of uncertainty due to the fractional collection of air from a much larger domain that needs characterizing. A full assessment of this uncertainty is particularly important for Bayesian and other stochastic inference algorithms.

In summary, it is the interface between the atmospheric model and the measurements system that has not been fully considered as part of an overall uncertainty budget. To explore this further, it is helpful to examine radioactive noble gas measurements performed at the time of the Fukushima reactor release. As noble gases do not have any particulate interactions, they provide an opportunity to characterize the uncertainty of small sample volumes and the use of ATDM in environmental interpretations of data.

1.1. Fukushima

The Fukushima reactor accident released between 9.8 PBq and 14.5 PBq of ^{137}Cs (Katata et al., 2015) and 11.4 EBq of ^{133}Xe in the first three days of the accident (Eslinger et al., 2014). The initial response in Canada involved modelling the release of radioactive debris using Lagrangian particle dispersion models developed at Environment and Climate Change Canada (ECCC), called *Modèle Lagrangien de Dispersion de Particules (MLDP)* (D'Amours et al., 2015). Lagrangian models use the release of a large number of particles, or more accurately parcels of air, to model the advection-diffusion process. During its transit the large plume travelled eastward from Japan to Canada as shown in Fig. 1. By the time of the plume's arrival at Vancouver Island, Canada, it was well mixed from its extended atmospheric transport and would be expected to be a semi-infinite cloud. However, even after travelling over 7000 km, there was a surprising amount of structure to the debris plume shown by an aerial survey conducted by Natural Resources Canada (NRCAN) (Sinclair et al., 2011).

At the time of the Fukushima reactor accident, the models used by ECCC for long-range transport were 1° by 1° with 3 h temporal resolution. The model output was generated between the surface and 500 m Above Ground Level (AGL) to capture planetary boundary effects. Thus, the overall volume of air in every grid box of the model domain was roughly $5 \times 10^{12} \text{ m}^3$ (cf. 20 000 m^3 particulate and 80 m^3 in the noble gas samples).

On March 20, 2011, NRCAN conducted an aerial survey along the western coast of Vancouver Island using an aircraft instrumented with specially configured NaI(Tl) detectors to assess and characterize the arrival of the Fukushima plume. The flight path and the corresponding dose rate measurements are shown in Fig. 2. The flight plan involved flying offshore at 250 m altitude along the coast for 350 km. The total survey time was 53 min. The counting or integration time of the samples was 20 s, which corresponds to a sample every 2.2 km along the flight path.

Examining the ^{133}Xe concentration in terms of the aircraft altitude along the flight path is plotted in Fig. 3. The change in altitude over the flight was relatively minor compared to the dimensions of the ATDM grid box. However, the magnitude of ^{133}Xe activity concentration change over the flight path could be quite large and was highly variable even over short distances. The aircraft spent the majority of its time in two grid boxes as shown in a 3D spatial domain in Fig. 4.

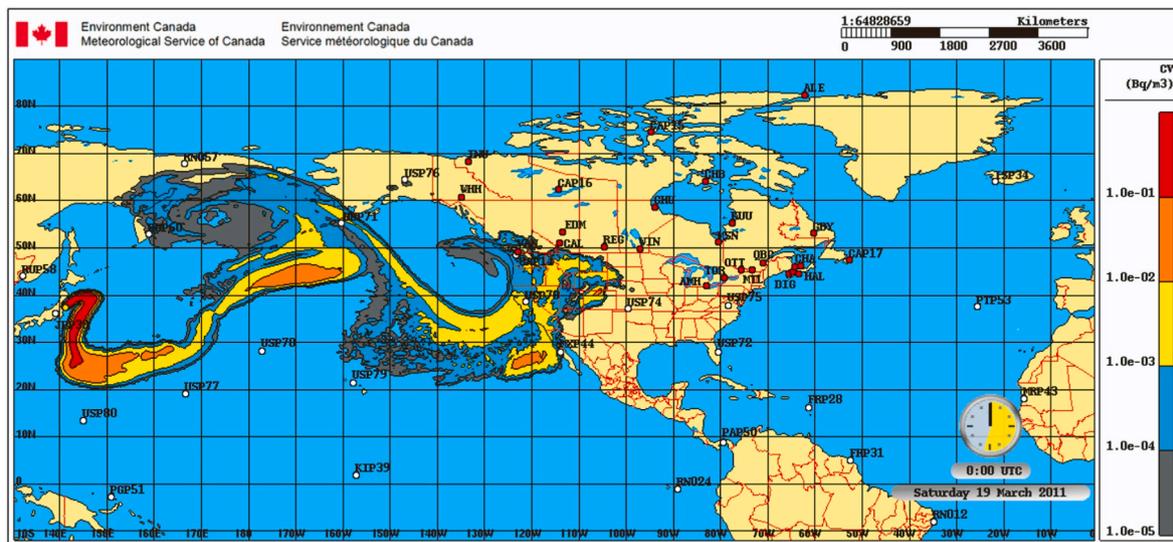


Fig. 1. A 3 h period showing the accident source term for ^{137}Cs propagated from the Fukushima reactor facility and the expected concentration at ground level. Although the model shows the behaviour of a different radioisotope, propagation of a ^{133}Xe source term would only change the absolute concentration predicted and not the relative differences shown. The labelled circles are the location of IMS aerosol samplers operated by the CTBTO.

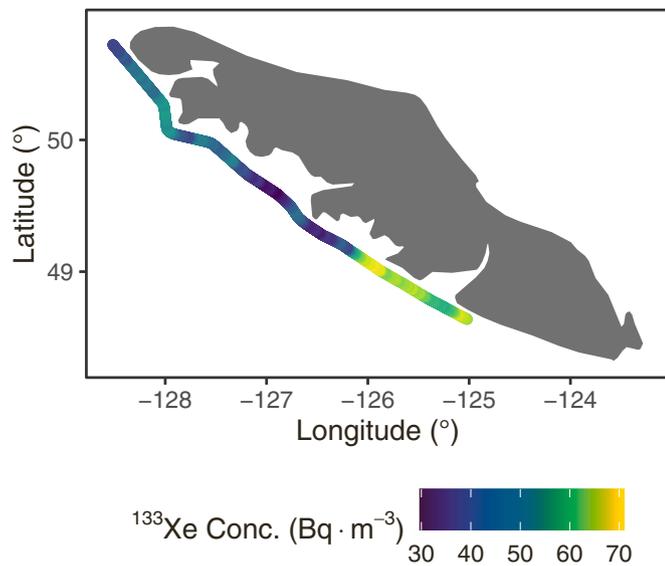


Fig. 2. Vancouver Island Survey of Fukushima Debris Plume. The dose data acquired by the NaI(Tl) system was converted to a concentration through the use of a Monte Carlo model.

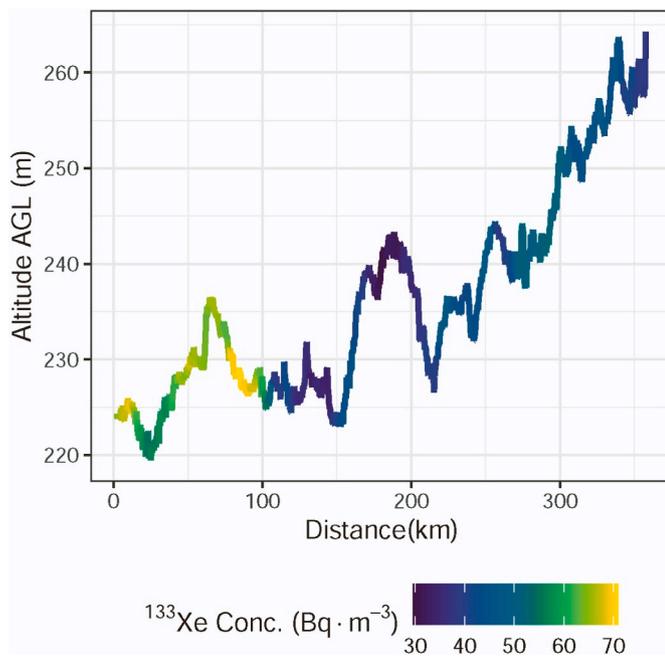


Fig. 3. ^{133}Xe Concentration and Aircraft Altitude AGL. The variation in aircraft altitude was relatively minimal during the flight and represents only an extremely small change in the domain of the associated dispersion model.

Conducting an aerial survey is essentially a high frequency sampling in an ATDM grid. The sampling showed the high degree of response variability that measurement systems may experience compared to what would normally be considered a constant activity concentration semi-infinite cloud. If the plume was homogeneous over the four grid cells (as shown in Fig. 4) of the 53 min survey flight, the mean concentration of ^{133}Xe would have been 48.2 Bq m^{-3} and the unbiased standard deviation, σ , would have been 11.2 Bq m^{-3} . However, if one looks at the activity concentration between 48° and 49° latitude and -126° and -127° longitude (which corresponds to the 80 km–120 km domain in Fig. 3), the activity concentration reported covers the full range of the entire flight from 30 Bq m^{-3} to 70 Bq m^{-3} all in the space of a single grid

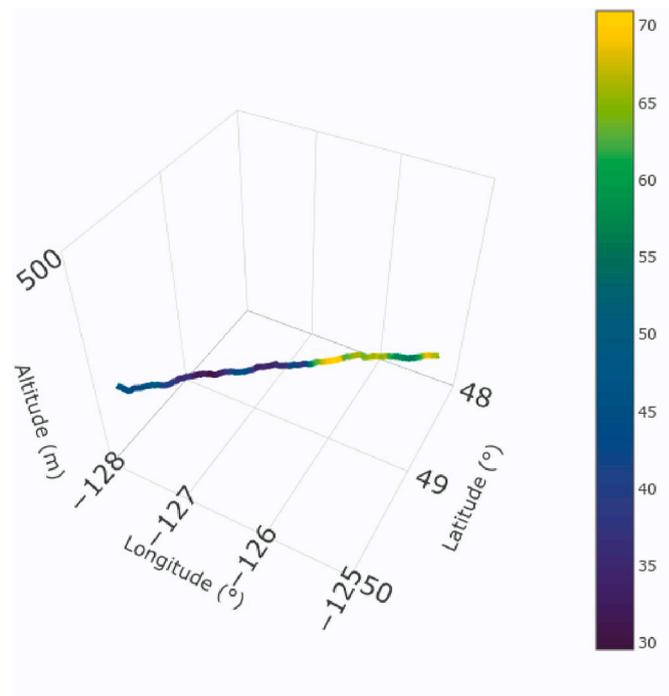


Fig. 4. ^{133}Xe Concentration (mBq m^{-3}) in 3D space. The flight shown in terms of the dispersion model domain output grid mesh (1° by 1° averaged over 500 m AGL). The estimation of xenon concentration was accurate to 200 m around the aircraft which is much thinner than the trace shown. The overall sample size is much smaller than the population.

cell. From this aerial survey there is clear evidence of a highly structured plume even after long distance transport and mixing. Furthermore there is initial evidence that a small sample collected from a grid cell has additional uncertainties beyond the plume homogeneity effects as shown by the aerial noble gas measurements.

2. Method

With the intra-cell variability established, it is possible to perform an initial quantification of the uncertainty using a network of ground-based detectors and examining data from the Fukushima accident. The goal is to develop an improved uncertainty budget, which will result in a more reliable and accurate inference of source parameters when used in reconstructive approaches, such as those used in Meutter and Hoffman (2020) and Dumont Le Brazidec et al. (Dumont Le Brazidec et al., 2021). Accurate knowledge of the source improves the understanding of an accident, including how it may evolve, which allows for responders to make better decisions during the response and recovery phases of an accident.

To derive a more accurate uncertainty budget, this study will use a dense network of low-energy but high-temporal resolution NaI(Tl) detectors located on Vancouver Island. By examining the response of the in-situ measurements from these detectors to the radioactive noble gas, ^{133}Xe ($\gamma = 80.9979 \text{ keV}$), it will be possible to estimate a sampling representativeness uncertainty. Furthermore, this first assessment of the inherent sampling uncertainty is free from any particulate/collector system effects such as particle size and preferential flow through an aerosol sampler.

For source reconstruction approaches, stationary ground sampler measurements are a typical input data source, so they are more appropriate than examining aerial survey data. Ground samplers often have longer sampling times, between 6 h and 24 h, so plume dynamics are much more important. Although source reconstruction with aerosol collector are the focus of this work, NaI(Tl) samplers are used as they

have a short integration time and there are multiple detectors present in the same geographic area, which is helpful to assess the variance.

On the Canadian West Coast, there is a relatively dense network of NaI(Tl) detectors that provided 15 min spectroscopic dose rate data during the transit of the Fukushima accident plume. The location of the Vancouver Island measurement sites is shown in Fig. 5. This region contains complex topography that will not fully be resolved by the ATDM, however the variability in response of the four different monitoring sites should give an indication of any modelling resolution related issues.

The network configuration was such that four detector sites were located very near one another in the same ATDM (0.1°) grid box: Sidney, Saanich, Metchosin and Victoria. If the Saanich location is designated as the origin, the distance from Saanich of all the other detectors is given in Table 1. The four sites previously mentioned would all be considered identical from a plume modelling viewpoint, as the distances between them would not be resolved by the dispersion model.

The dose data that was recorded at all detector locations is shown in Fig. 6. Qualitatively, all measurement locations, even the more distant sites, show similar features – same plume arrival and departure times, radioxenon dose amplitudes and shoulder profiles. In addition to showing the geospatial characteristics of the plume, the consistency of the dose traces at all locations shows that the detectors were operating with the correct gain and inter-detector biases were minimal during the transit of the plume. Furthermore, there is no evidence of topographical effects on the doses recorded at different locations, indicating that if these effects were present they were minimal in this study. The

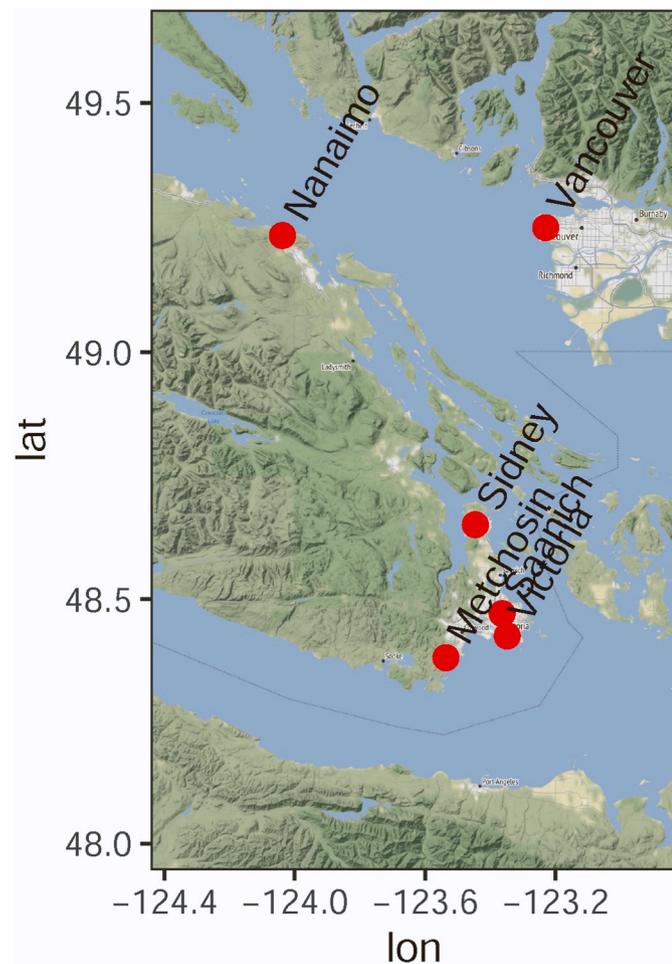


Fig. 5. The location of the NaI(Tl) detectors around Vancouver Island on the West Coast of Canada. The detectors are located on the Southern tip of Vancouver Island.

Table 1

Distances of the four closest detectors to the one located in Saanich. The measurement site at Saanich is the most centrally located of the four locations.

| Detector Location | Distance (km) |
|-------------------|---------------|
| Victoria | 5.2 |
| Sidney | 21.0 |
| Metchosin | 16.2 |
| Nanaimo | 98.3 |
| Vancouver | 87.3 |

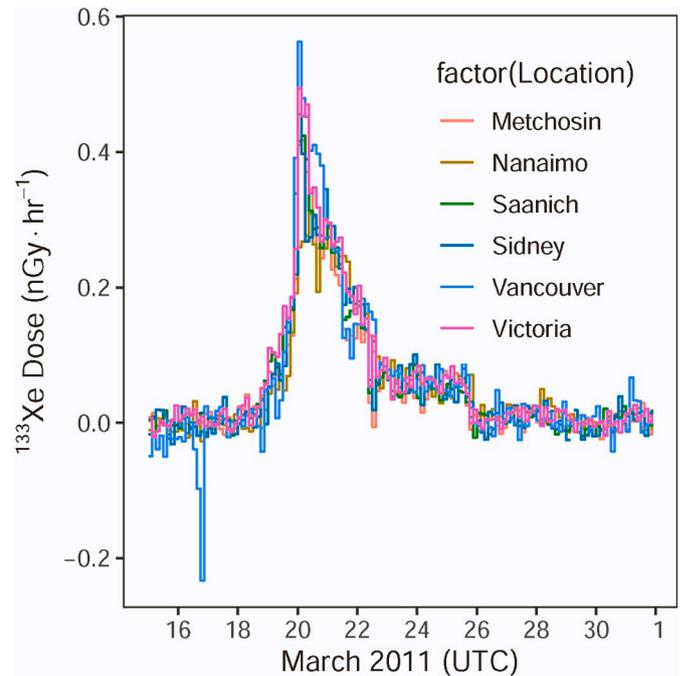


Fig. 6. Dose data from all stations near Vancouver Island during the entire passage of the noble gas plume from the Fukushima reactor. The 15 min integration time of the NaI(Tl) detectors was resampled into 3 h bins to align with the meteorological model output time.

qualitative behaviour of the detector response corresponds well with the ATDM models generated by ECCC (shown previously in Fig. 1) where the plume was relatively uniform in concentration over a broad domain.

Overall, the qualitative behaviour of the detector response combined with the broad-scale uniformity of the plume in its transit would suggest a relatively straight-forward application of the ATDM in reconstructing the source. However, the logarithmic scale typically used with ATDM obscures some of the inherent uncertainty at the interface between measurements and ATDM if the measurement data is examined in detail.

3. Results and discussion

To study the plume-model interface variance, the four station cluster was examined in detail. By imposing a threshold of 0.1 nGy h⁻¹ the low dose rate noise was removed. A reference station, Metchosin, was selected as it recorded the lowest doses, to see the relative behaviour between the different sites by setting its dose data to unity throughout the period of interest. The results are shown in the two panels of Fig. 7. This figure shows that there are periods near the beginning and end of the plume passage that are dramatically different on a relative basis and that the Saanich and Victoria stations can switch their relative dose ranking. This can be more easily seen in the bottom panel where the dose can approach twice the reference dose, however on a 24 h period the average dose was a factor of 1.5 greater for Victoria and 1.3 greater for

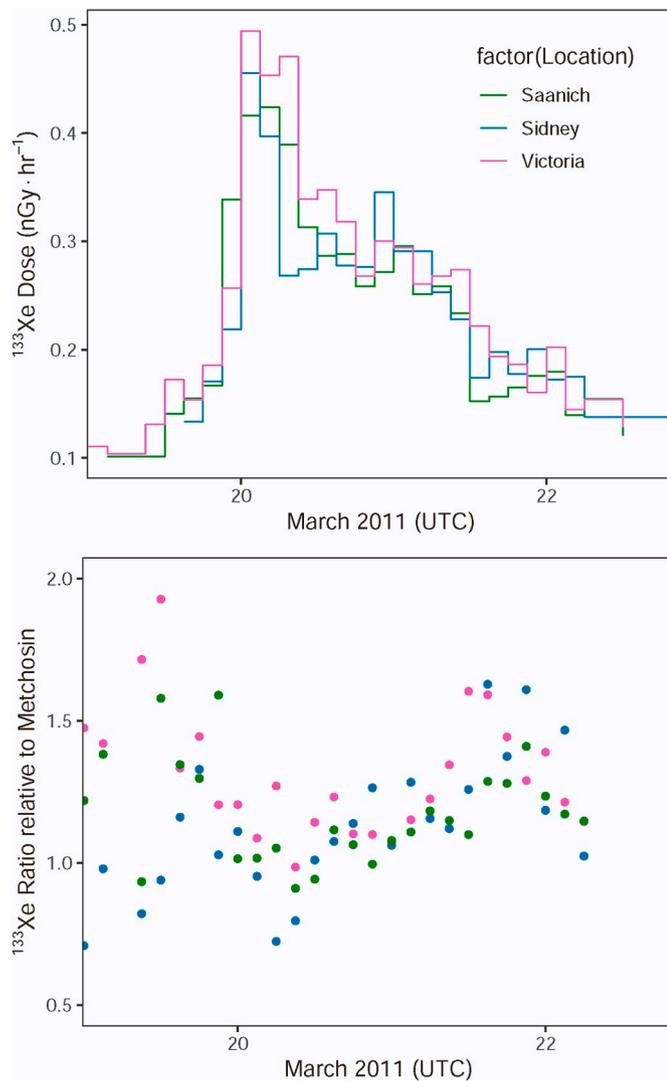


Fig. 7. Dose data from the three tightly clustered stations on Vancouver Island. The 15 min integration time of the NaI(Tl) detectors was resampled into 3 h bins to align with the meteorological model output time. The bottom panel shows the dose relative to the Metchosin site.

Saanich. On the tail end of the plume for the 24 h prior to March 22, the dose was a factor of 1.33 greater for Victoria and 1.2 greater for Saanich. In the middle of the observation period, or throughout March 20, the dose was only a factor of 1.11 greater for Victoria and 1.01 greater for Saanich. Overall the spread of the doses ranged from between 0.75 to almost a factor of 2 depending on the site and 3 h interval chosen.

The relative variance between the different locations during the passage of the plume would be difficult to replicate with an ATDM model. Since reproducing or predicting this behaviour is challenging, if not impossible, an empirical characterization was performed to estimate the variance of small samples being used in environmental interpretations. To examine the magnitude of variability during the plume passage period, the Interquartile Range (IQR) was calculated (Fig. 7 shows the source data for the 3 h integration time) as the relative doses were multi-modal in some cases. The results of this analysis for 15 min normal integration period of the detector and the manually integrated periods of 3 h, 6 h and 12 h were calculated by observation site. The latter calculations would reflect the performance of current and envisioned noble gas systems used by the CTBTO. Finally, the overall mean was calculated for each integration time. The results of this analysis are shown in Table 2.

Table 2

IQR of the observed dose ($>0.1 \text{ nGy h}^{-1}$) relative to Metchosin for several common sampler integration periods.

| Detector Location | Integration Time | | | |
|-------------------|------------------|-------------|-------------|-------------|
| | 15 min | 3 h | 6 h | 12 h |
| Victoria | 0.32 | 0.26 | 0.30 | 0.39 |
| Sidney | 0.35 | 0.25 | 0.18 | 0.14 |
| Saanich | 0.28 | 0.24 | 0.19 | 0.17 |
| Mean | 0.32 | 0.25 | 0.22 | 0.23 |

Interestingly, while the IQR diminishes as the integration period increases as expected for Sidney and Saanich, this is not true for the Victoria site. The overall dose from Fig. 6 shows that Victoria is the most dynamic of the monitoring sites, so the overall variability in the IQR as the integration time changes further illustrates the presence of an unaccounted variance. In terms of scale, the variability of 20%–30% fits in between those calculated by De Meutter et al. (De Meutter et al., 2021) (who found the model error to be up to a factor of 2) while examining reconstruction model error and the uncertainty caused by inhomogeneous deposition in an aerosol sampler of 5%–10% (Gomez et al., 2014).

4. Conclusion

Source reconstruction is a powerful tool to examine the properties of an unknown radioactive source using a network of detectors. However, the accuracy of any reconstruction that relies upon stochastic techniques such as Bayesian inference, depends on having an accurate assessment of uncertainty in the reconstruction process.

The use of ATDM with air sampling systems or in-situ measurement leads to an additional uncertainty element in reconstruction approaches as the measured value recorded at the sampler location is assumed to be uniform throughout the grid box of the model.

By examining the behaviour of a dense network of NaI(Tl) detectors during the Fukushima reactor accident, a first estimation of the uncertainty due to the representativeness of small sample volume collection compared to the environmental domain represented by ATDM was performed. For typical samplers where the sample collection time is between 6 h and 12 h an uncertainty of 23% was estimated from the Vancouver Island study based upon the mean IQR of the dose recorded by three samplers to an arbitrarily chosen reference sampler. Furthermore, the mean IQR did not change significantly between these two integration times and only increased slightly (by 2%) when a 3 h integration period was used. Further studies would be necessary, but there appears to be an intrinsic uncertainty for fractional air collection of approximately 25%.

Furthermore, the results of this examination raises some interesting questions regarding the quantification of uncertainty. For example, if ensemble models had been available for this case, how much additional uncertainty would their spread have predicted? It would also be an interesting study to examine the fractional air sampling uncertainty proportion with respect to the overall ensemble spread for various integrations periods. Secondly, the location of these samplers is in a region with complex topography and significant land-sea interactions. No immediate issues with the analysis could be identified from variability the detector response in this study. However, as the ATDM is not able to resolve the topographic features (1° resolution), this uncertainty assessment would be worth repeating at other locations with different topography, such as the prairies. If measurement data was available from different regions, further study to characterize this uncertainty would be worthwhile.

This first quantification is an important contribution to the total uncertainty budget for source reconstruction. Although it is smaller than the uncertainty from ATDM modelling, it can be very similar to activity concentration uncertainty for radionuclides near the limit of detection. An uncertainty of this magnitude is worth including in an overall

uncertainty budget when performing source reconstruction and merits consideration when using radionuclides in an environmental interpretation process. Furthermore, inclusion of this uncertainty in stochastic source reconstruction algorithms would improve the overall event assessment and characterization.

CRedit authorship contribution statement

Ian Hoffman: Conceptualization, Methodology, Software, Data Curation, Formal Analysis, Investigation, Visualization, Writing – Original Draft, Writing – Review & Editing. **Alain Malo:** Resources. **Kurt Ungar:** Writing – Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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