

The total release of xenon-133 from the Fukushima Dai-ichi nuclear power plant accident

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ARTICLE INFO

Article history:

Received 12 April 2012

Received in revised form

25 May 2012

Accepted 4 June 2012

Available online

Keywords:

Nuclear accident

Fukushima

Xenon-133

ABSTRACT

The accident at the Fukushima Dai-ichi nuclear power plant (FD-NPP) on 11 March 2011 released large amounts of radioactivity into the atmosphere. We determine the total emission of the noble gas xenon-133 (¹³³Xe) using global atmospheric concentration measurements. For estimating the emissions, we used three different methods: (i) using a purely observation-based multi-box model, (ii) comparisons of dispersion model results driven with GFS meteorological data with the observation data, and (iii) such comparisons with the dispersion model driven by ECMWF data. From these three methods, we have obtained total ¹³³Xe releases from FD-NPP of (i) 16.7 ± 1.9 EBq, (ii) 14.2 ± 0.8 EBq, and (iii) 19.0 ± 3.4 EBq, respectively. These values are substantially larger than the entire ¹³³Xe inventory of FD-NPP of about 12.2 EBq derived from calculations of nuclear fuel burn-up. Complete release of the entire ¹³³Xe inventory of FD-NPP and additional release of ¹³³Xe due to the decay of iodine-133 (¹³³I), which can add another 2 EBq to the ¹³³Xe FD-NPP inventory, is required to explain the atmospheric observations. Two of our three methods indicate even higher emissions, but this may not be a robust finding given the differences between our estimates.

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1. Introduction

On 11 March 2011, an extraordinary magnitude 9.0 earthquake occurred about 130 km off the Pacific coast of Japan's main island Honshu, followed by a large tsunami (USGS, 2011). One of the consequences was a station blackout at the Fukushima Dai-ichi nuclear power plant (FD-NPP), which developed into a disaster leaving four of the six FD-NPP units heavily damaged. The result was a massive discharge of radionuclides. In the atmosphere, the radionuclides were transported throughout the Northern Hemisphere (Stohl et al., 2012) and could be detected at many stations (e.g. Bowyer et al., 2011).

The total amount of radioactivity released into the atmosphere is still uncertain. It can be estimated based on calculations of the radionuclide content of the nuclear reactors combined with accident simulations, or using ambient atmospheric monitoring data together with some sort of inverse modelling. Japanese authorities

used both approaches and provided estimates for many radionuclides (NERH, 2011).

Of all the radionuclide emissions, the radioactive noble gas releases can be quantified most accurately, since it is almost certain that the entire noble gas inventory of the heavily damaged reactor units 1–3 was set free into the atmosphere. For other radionuclides, only a small but highly uncertain fraction of the inventory was released into the environment. Complete noble gas release was also assumed by the Japanese authorities (NERH, 2011) who estimated a release of 12.2 EBq of ¹³³Xe, the most important radioactive noble gas with a half-life of 5.25 d. The inventory estimates of Bowyer et al. (2011) of 12 EBq ¹³³Xe and Stohl et al. (2012) of 12.4 EBq ¹³³Xe are nearly identical. While the excellent agreement may indicate that the inventory is known with high accuracy, the estimates are all based on similar methods, so the true uncertainty of the ¹³³Xe inventory may be higher. Nevertheless, the ¹³³Xe inventory should be known to within a few percent at most. However, using measured atmospheric concentrations at many stations in the Northern Hemisphere (NH) together with inverse modelling, Stohl et al. (2011) obtained a much higher release of 16.7 (13.4–20.0) EBq ¹³³Xe. In a revision of their discussion paper, more accurate decay corrections for the measurement data resulted in a reduced estimate of 15.3 (12.2–18.3) EBq ¹³³Xe (Stohl et al., 2012),

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but this is still a substantially higher value than the calculated ^{133}Xe inventory. This discrepancy has prompted a discussion with nuclear engineers whether such a high ^{133}Xe release is possible at all, given that the ^{133}Xe inventory is thought to be known with high accuracy (Di Giuli et al., 2011). A partial explanation was given by Seibert (2011): The decay of ^{133}I (half-life of 20.8 h), another radionuclide present in the reactor cores, into ^{133}Xe effectively adds about 16.5% to the ^{133}Xe inventory of FD-NPP. This would increase the estimates of NERH (2011) to an effective ^{133}Xe inventory of 14.2 EBq. Assuming that all the ^{133}Xe produced from ^{133}I decay is released into the atmosphere, this value is consistent, within error bounds, with the revised inverse modelling result of 15.3 (12.2–18.3) EBq ^{133}Xe by Stohl et al. (2012). However, based on the mean value, the discrepancy is not fully resolved and it is also uncertain whether all the ^{133}Xe produced from ^{133}I decay can be released as well.

Based on the above discussion, there is a need to better quantify the total release of ^{133}Xe into the atmosphere, and this motivated us to calculate the total ^{133}Xe release using methods that are independent of those used by Stohl et al. (2012). This is the purpose of the present study. Stohl et al. (2012) used measurement data from the first few weeks after the Fukushima accident with an inverse modelling approach based on a Lagrangian particle dispersion model to determine the ^{133}Xe emissions as a function of time. Here, we use a simpler approach that takes advantage of the low minimum detectable activity concentration in ambient ^{133}Xe concentration measurements of a global station network. This allowed quantification of the FD-NPP-related concentrations at all stations in the NH over a period of three months, despite the short half-life of ^{133}Xe of 5.25 d. Since the emissions become relatively well mixed in the atmosphere after a few weeks, we can use a very simple multi-box model to estimate the atmospheric ^{133}Xe inventory. With this simple approach we cannot determine the exact time of the emissions from FD-NPP, in contrast to Stohl et al. (2012), but we can estimate the total amount of ^{133}Xe released into the atmosphere with relatively high accuracy. In a second approach, we also use the ^{133}Xe emission source term of Stohl et al. (2012) to simulate the radionuclide dispersion over a period of three months using two different meteorological data sets, and then use the measurement data to re-scale the modelled total emissions of Stohl et al. (2012) to achieve a best fit with the measurement data.

2. Measurements of Xe-133

To verify compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT), a global international monitoring system is currently being built up, which includes measurements of several radioactive isotopes of the noble gas xenon (Wernsberger and Schlosser, 2004; Saey and de Geer, 2005). Currently, up to 25 stations are delivering noble gas data to the Preparatory Commission for the CTBT Organization (CTBTO). We have used data from all stations in the NH and Tropics with good data availability and without major influence from local sources, as shown in Fig. 1. The collection period of the xenon samples is 12 or 24 h, depending on the station. The isotope ^{133}Xe is measured with an accuracy of about 0.1 mBq m^{-3} . The measurement uncertainties are reported for every sample and are typically below 1% (partly below 0.1%) after the arrival of the FD-NPP plume and until about 20 April. At the end of May, when most of the ^{133}Xe activity released from FD-NPP had decayed, uncertainties are some 10–25%.

Even without the FD-NPP emissions, observed levels of ^{133}Xe in the atmosphere are highly variable due to small releases from medical isotope production facilities and nuclear power plants. The CTBTO network records ^{133}Xe “pollution episodes” regularly, especially at stations downwind of the known sources of

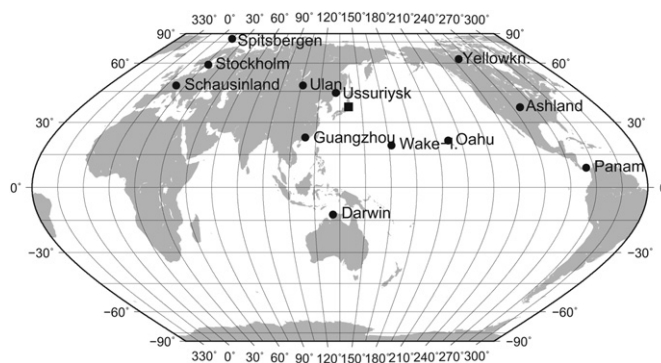


Fig. 1. Map showing the locations of stations used in this study. The location of FD-NPP is marked with a black rectangle. For lack of space, some station names are abbreviated: Ulan-Bator (Ulan), Wake-Island (Wake-I), Panama (Panam), Yellowknife (Yellowkn).

radioxenon (Wotawa et al., 2010). This known background is on the order of some mBq m^{-3} and was determined here by averaging all measured concentrations for each station for the period 1 January till 11 March 2011.

Fig. 2 shows three examples of the ^{133}Xe concentrations measured at Yellowknife, Ashland and Darwin. At Yellowknife (Fig. 2, top), the concentrations (red squares) reach a peak of some 2 Bq m^{-3} about two weeks after the Japanese earthquake and tsunami. After that peak, the measured concentration decline follows almost exactly the 5 d half-life exponential radioactive decay of ^{133}Xe (which would appear as a linear graph in the logarithmic plot). The measured values return to the detection limit as late as early June. The ^{133}Xe background at Yellowknife is very low and, thus, the enhancements over the background, denoted as $\Delta^{133}\text{Xe}$ in the following (blue plus signs), are nearly identical to the observed values. Only in late May and early June a small effect of the background subtraction can be seen, when $\Delta^{133}\text{Xe}$ values are slightly lower than ^{133}Xe values. Assuming that the ^{133}Xe enhancements over the background are entirely due to the emissions from FD-NPP, we can correct them for the radioactive decay since the time of the earthquake. The corrected values, $\Delta^{133}\text{Xe}_c$ (black crosses), increase until early April. After that, $\Delta^{133}\text{Xe}_c$ values show little variability but a slow decline by less than a factor of two until early June. Three points are remarkable: 1) The lack of variability in $\Delta^{133}\text{Xe}_c$ after early April suggests that the FD-NPP ^{133}Xe emissions were nearly uniformly mixed in the midlatitude troposphere. 2) The slow decline suggests a leak of ^{133}Xe from the midlatitudes into the Tropics and the Southern Hemisphere (SH) and possibly also into the stratosphere. It is also possible that vertical mixing in the troposphere was not complete in early April. 3) Substantial new ^{133}Xe emissions from FD-NPP in April or May can be ruled out, since, depending on the emission time, even emissions on the order of about 0.1–1% of the emissions that had occurred during the first week after the earthquake (Stohl et al., 2012), would be clearly detectable. This finding is relevant on the background of speculations about a possible recriticality in the damaged reactors.

At Ashland (Fig. 2, middle), the ^{133}Xe behaviour is similar to Yellowknife, but this site encounters more regional ^{133}Xe pollution events, which are inflated by the decay correction and add noise to the FD-NPP signal in late May and early June. Subtraction of the background helps to avoid a systematic increase of $\Delta^{133}\text{Xe}_c$ from late May.

At Darwin in the SH (Fig. 2, bottom), the signal from FD-NPP is relatively weak compared to the NH sites. Air masses containing

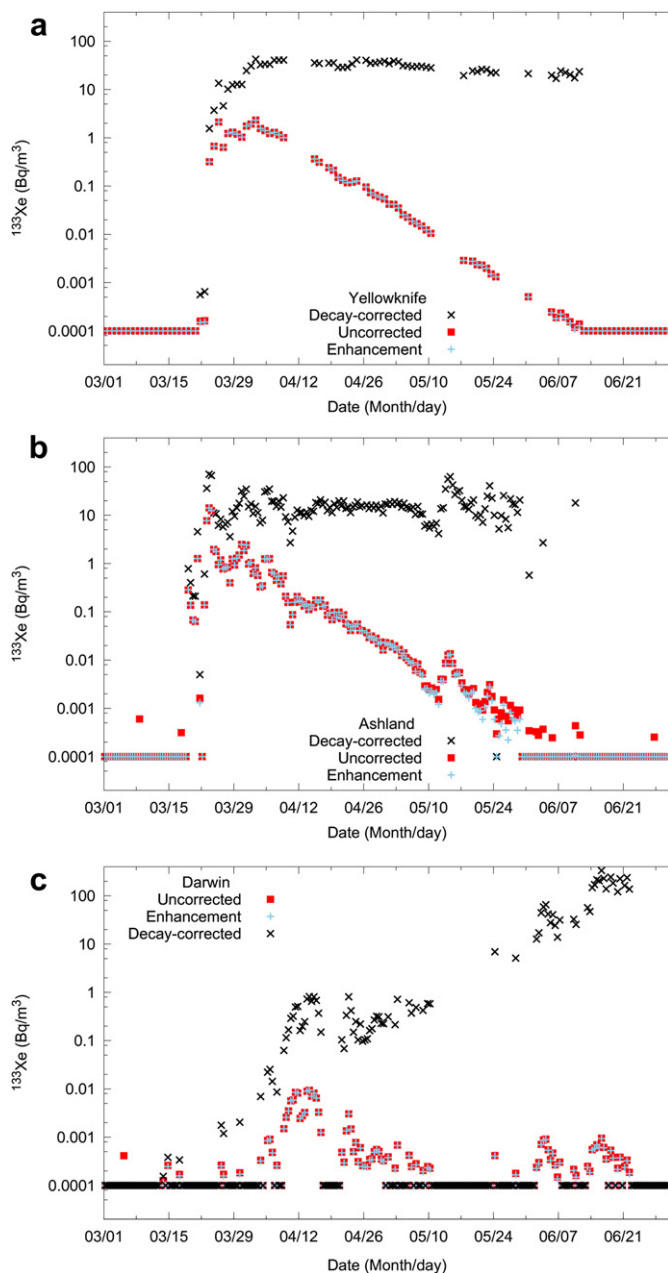


Fig. 2. Time series of observed ^{133}Xe concentrations at Yellowknife (top), Ashland (middle) and Darwin (bottom). Shown are the uncorrected observed concentrations of ^{133}Xe (red squares), the enhancements of the observed concentrations over the background (i.e., observations with background subtracted) $\Delta^{133}\text{Xe}$ (blue pluses), and the decay-corrected enhancements over the background $\Delta^{133}\text{Xe}_c$ (black crosses). Measurements below the detection limit have been set to a constant value of 0.1 mBq m^{-3} for displaying purposes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

FD-NPP emissions arrived in late March and early April but ^{133}Xe concentrations declined back towards the detection limit in May. The small rise in ^{133}Xe concentrations in June is likely caused partly by other sources than FD-NPP.

Summarizing these and the other observations, we find that the FD-NPP $\Delta^{133}\text{Xe}_c$ emission pulse is observable at all NH stations until at least late May. The small variability from about 10 April to 15 May at all stations suggests that the plume at that time was already well-mixed in the troposphere in broad zonal bands. These findings are consistent with current understanding of the time scales of

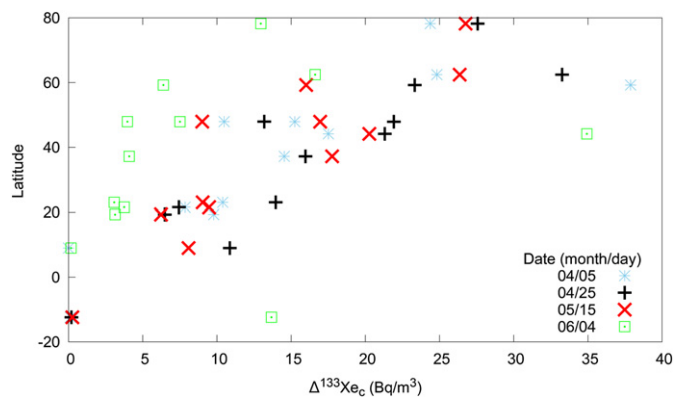


Fig. 3. Decay-corrected enhancements over the background, $\Delta^{133}\text{Xe}_c$ measured at the surface stations as a function of latitude and averaged over four intervals of 20 d (dates in figure legend refer to interval mid-points).

intercontinental pollution transport in the middle latitudes. After 25–30 d in the atmosphere, an emission pulse from East Asia is typically mixed quite homogeneously both zonally as well as vertically in the troposphere (see Figs. 2–4 in Stohl et al., 2002). Mixing into the Tropics and the SH results in a slow decrease of $\Delta^{133}\text{Xe}_c$ values in the NH and the arrival of $\Delta^{133}\text{Xe}_c$ signals at Panama and Darwin.

Fig. 3 shows $\Delta^{133}\text{Xe}_c$ at the various stations averaged over four periods of 20 d. The latitudinal variability is strongest for the first interval when the emitted ^{133}Xe was not yet well mixed in the troposphere, and for the last interval when measured concentrations return to their background levels and uncertainties in background levels are inflated by the decay correction.

3. A simple multi-box model

The findings of Section 2 suggest that it is possible to estimate the total ^{133}Xe release by inventorying the ^{133}Xe activity in the atmosphere using the CTBTO measurement data. If we assume that measured $\Delta^{133}\text{Xe}_c$ concentrations at the ground are representative for the depth of the tropospheric column and for the latitude band a certain station is located in, the total release R of ^{133}Xe from FD-NPP (decay-corrected to the time of the earthquake) follows from

$$R = \sum_{i=1}^N A_i \times H_i \times \Delta^{133}\text{Xe}_{c,i} \quad (1)$$

where N is the number of stations (latitude bands) used, A_i is the area of latitude band i , H_i is an appropriate tropospheric scale height, and $\Delta^{133}\text{Xe}_{c,i}$ is the decay-corrected enhancement over the background at station i , averaged over a suitable time interval. In meteorology, the atmospheric scale height is a measure of the effective “thickness” of an atmospheric layer (Glossary of Meteorology, American Meteorological Society, see <http://amsglossary.allenpress.com/glossary>). Since we assume that mixing of ^{133}Xe has occurred only within the troposphere, we obtain an effective tropospheric scale height by dividing the air column density up to the tropopause height with the surface density. For this calculation, we have used meteorological analysis data from the Global Forecast System (GFS) model of the National Centers for Environmental Prediction (NCEP) for integrating air density from the surface up to the last pressure level below the tropopause, and averaged 3-hourly scale heights to monthly values. Northern and southern boundaries of our latitude bands are located at mid-points between station latitudes and at 20° S and 90° N . The results are not sensitive to changes of the southern boundary, since the measured

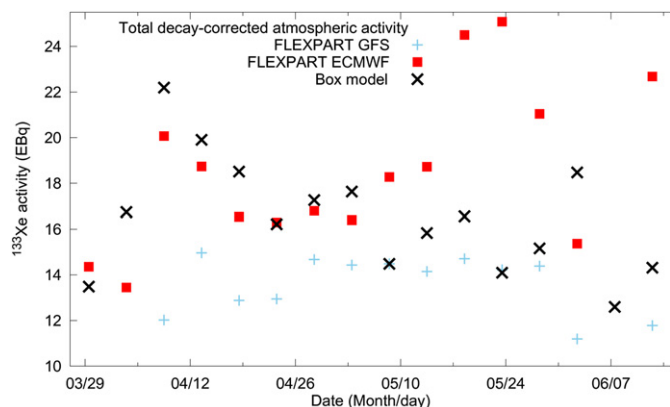


Fig. 4. Estimates of total $\Delta^{133}\text{Xe}$ emissions from FD-NPP corrected to the time of the earthquake using all measurement data averaged over intervals of 5 d (dates refer to interval mid-points) as a function of time. Calculations are based on a box model using tropospheric scale heights (black line), and comparisons with dispersion model simulations driven with ECMWF (red line) and GFS (blue line) meteorological data. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

concentrations in the Tropics remain very low until the end of May (see Fig. 2c and Fig. 3).

Fig. 4 shows values of R for 16 intervals of 5 d. Each value is a largely independent estimate of the FD-NPP emissions, however, with different quality. During the first three intervals, the plume is not yet well dispersed and measurements at all stations are still highly variable. The increase of the inventory estimate during that period is due to a large fraction of the emissions initially escaping detection by the measurement network. The last three estimates are also quite variable due to the inflation of noise (measurement errors, background subtraction) by the decay correction. After 15 June, release estimates become entirely unrealistic (not shown). Estimates using the central nine intervals of 5 d (from 11 April to 25 May) show relatively little variability, suggesting that the method works best during that period, which will later be used for deriving our best estimate of the FD-NPP ^{133}Xe release.

Using data only until 25 May is also justified when considering measurement uncertainty and background subtraction. Assuming that the reported measurement uncertainties for 12-hourly samples are random and uncorrelated, measurement errors at individual stations are below 1% for a 5 d period at the end of April and below about 5% for the last 5 d period (21–25 May). Resulting errors of R would be about a factor two smaller than these values (because 12 stations with different weights are used) and can be ignored compared to other errors. To give an upper limit for the error introduced by the background subtraction, we recalculated R values without any background subtraction at all. Differences between R values with and without background subtraction were less than 1% until early May and reached 25% for the last 5 d period (21–25 May). Thus, we conclude that differences in R found for individual 5 d intervals until 25 May cannot be due to measurement errors or background subtraction, except maybe for the last two or three 5 d periods. Instead, this variability must be due to the heterogeneity of the ^{133}Xe distribution and how it is sampled by the rather sparse measurement network.

The overall negative trend of R between 11 April and 25 May can be explained by leakage of ^{133}Xe to areas south of our southernmost station as well as mixing into the tropopause region and the stratosphere. Furthermore, xenon is slightly water soluble, which suggests slow uptake by the ocean may also be partly responsible for the decline. This would mean that values in late May underestimate the total atmospheric $\Delta^{133}\text{Xe}_c$ inventory. It is also possible

that ^{133}Xe is not yet well mixed vertically in early April and the first few estimates are too high but it is not certain that surface concentrations should be systematically higher than those aloft, given the initial lifting of the plume (Stohl et al., 2012) and the climatological transport characteristics of surface emissions from Asia (Stohl et al., 2002). For the 45 d period from 11 April to 25 May, we obtain an average $R = 16.7$ EBq with a standard deviation of 1.9 EBq.

4. Dispersion model calculations

We performed simulations of the ^{133}Xe dispersion from FD-NPP with the Lagrangian particle dispersion model FLEXPART (Stohl et al., 2005) using the detailed time-varying source term with a total emission of 16.7 EBq ^{133}Xe determined by Stohl et al. (2011). This source term contains ^{133}Xe releases primarily from 11 to 15 March, only minor emissions were identified after 16 March. Two alternative calculations using meteorological input data from ECMWF (European Centre for Medium-Range Weather Forecasts) and GFS were performed. Details of the simulations, extended here until 15 June 2011, are given in Stohl et al. (2011).

From the model results and the CTBTO measurement data, we can estimate R as

$$R = R_s \frac{\sum_{i=1}^N \Delta^{133}\text{Xe}_{c,i}}{\sum_{i=1}^N {}^{133}\text{Xe}_{s,i}} \quad (2)$$

where R_s is the total ^{133}Xe release used in the model simulation (16.7 EBq) and ${}^{133}\text{Xe}_{s,i}$ is the simulated decay-corrected ^{133}Xe concentration at station i . This means that we scale the original source term to best fit our observations, leaving its temporal shape unmodified. Compared to the simple box model, this approach, within the error bounds of the simulation, properly considers effects such as loss into the SH and the stratosphere.

The results, again averaged over intervals of 5 d, are shown in Fig. 4. Using model results based on ECMWF data, the release estimates until early May are slightly lower than but well correlated with those obtained from the purely observation-based multi-box model. From 18 May, the ECMWF estimates become highly variable. Using the GFS data, the release estimates are continuously lower than the multi-box model but they are remarkably stable from mid-April to end of May. If we again average over the period 11 April to 25 May, we obtain total releases of 14.2 ± 0.8 and 19.0 ± 3.4 EBq for the calculations using GFS and ECMWF meteorological data, respectively. The results are quite different, mainly due to stronger vertical lifting of the emissions in the ECMWF-driven simulation compared to the GFS-driven simulation. This was already seen during the first 1–2 weeks after the earthquake, with the GFS-driven simulation appearing more realistic (Stohl et al., 2012). Thus, the lower total ^{133}Xe release derived using the GFS data are likely to be a better estimate than the higher value derived using the ECMWF data.

5. Discussion and conclusions

Using (i) the observation-based multi-box model, (ii) comparisons of dispersion model results driven with GFS meteorological data with observation data, and (iii) such comparisons driven by ECMWF data, we have obtained total ^{133}Xe releases from FD-NPP of (i) 16.7 ± 1.9 EBq, (ii) 14.2 ± 0.8 EBq, and (iii) 19.0 ± 3.4 EBq, respectively. Error bounds are the standard deviations from independent estimates based on nine subsequent periods of 5 d. These values compare well to the inverse-modelling estimate by Stohl et al. (2012) of 15.3 ± 3.1 EBq.

It is interesting that both the inverse modelling of Stohl et al. (2012) and the simple method of this paper leads to lower emission estimates when the GFS meteorological data are used than with ECMWF data. Given the fact that in Stohl et al. (2012) the GFS-based results were in better agreement with observations, the lower estimate may be somewhat more credible. The simple box model leads to a medium value. We can take this as an indication that all three methods are reasonable, however, due to the different inherent sources of error of each method, which can be quantified only in parts, some caution is needed in the interpretation.

The conclusions drawn in Stohl et al. (2012), that the whole ^{133}Xe inventory of Fukushima Daiichi units 1–3 was released, and that in addition the whole ^{133}Xe that is produced from the decay of ^{133}I was released as well, is confirmed. The tendency towards release estimates which are even higher warrants further investigations into different directions, e.g. the uncertainties of the burn-up based calculated inventories, the possibility of releases from other sources, and in-depth studies of uncertainties related to radionuclide measurements and to atmospheric transport modelling, or, respectively, the meteorological assumptions behind the simple box model.

Acknowledgements

We would like to acknowledge the work of CTBTO in building up and maintaining its global radio-xenon measurement network. ECMWF and met. no granted access to ECMWF analysis data. The work was supported by the Norwegian Research Council in the framework of the SOGG-EA project.

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