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

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 estimate the total emission of the noble gas xenon-133 (^{133}Xe) using global atmospheric concentration measurements. We estimate the emissions using three different methods, one using a multi-box model, the other a dispersion model driven with two different meteorological input data sets. The three methods yield total ^{133}Xe releases of 16.7 EBq±1.9 EBq, 14.2±0.8 EBq and 19.0±3.4 EBq, respectively. These values are substantially larger than the entire ^{133}Xe inventory of FD-NPP of 12.2 EBq derived from calculations of nuclear fuel burn-up. Additional release of ^{133}Xe due to the decay of iodine-133 (^{133}I), which can add another 2 EBq to the ^{133}Xe 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 uncertainties.

Keywords: nuclear accident, Fukushima, Xenon-133

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 modeling. 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. This was also assumed by the Japanese authorities (*NERH*, 2011) who estimated a release of 12.2 EBq of ^{133}Xe, the most important radioactive noble gas with a half-life of 5.25 days. Surprisingly, using measured atmospheric concentrations at many stations in the Northern Hemisphere (NH) together with inverse modeling, *Stohl et al.* (2011) obtained a much higher release of 16.7 (13.4-20.0) EBq ^{133}Xe. In a revision of their discussion paper, more accurate decay corrections for the measurement data resulted in a slightly reduced estimate of 15.3 (12.2-18.3) EBq ^{133}Xe (*Stohl et al.*, 2012), 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 fuel burn-up estimates of *NERH* (2011) to 14.2 EBq ^{133}Xe. 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 modeling 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 this motivated us to use independent methods to calculate the total ^{133}Xe release.

In this study, we take advantage of the low minimum detectable activity concentration in ambient ^{133}Xe concentration measurements, which 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 days. 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.

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 hours, depending on the station. The isotope ^{133}Xe is measured with an accuracy of about 0.1 mBq m^{-3}.

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 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.

Figure 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 Δ^{133}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 Δ^{133}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, $\Delta^{133}\mathrm{Xe}_c$ (black crosses), increase until early April. After that, $\Delta^{133}\mathrm{Xe}_c$ 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 $\Delta^{133}\mathrm{Xe}_c$ after early April suggests that the FD-NPP ^{133}Xe emis-

sions 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 behavior 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 $\Delta^{133}\mathrm{Xe}_c$ 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 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 $\Delta^{133}\mathrm{Xe}_c$ 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 intercontinental pollution transport in the middle latitudes. After 25-30 days 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 $\Delta^{133}\mathrm{Xe}_c$ values in the NH and the arrival of $\Delta^{133}\mathrm{Xe}_c$ signals at Panama and Darwin.

Figure 3 shows $\Delta^{133}\mathrm{Xe}_c$ at the various stations averaged over four 20-day periods. 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 the inflation by the decay correction of pollution events from regular nuclear industry sources becomes important.

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 $\Delta^{133}\mathrm{Xe}_c$ 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

$$R = \sum_{i=1}^{N} A_i \times H_i \times \Delta^{133}\mathrm{Xe}_{c,i} \tag{1}$$

where N is the number of stations (latitude bands) used, A_i the area of latitude band i, H_i its tropospheric scale height, and $\Delta^{133}\mathrm{Xe}_{c,i}$ the decay-corrected enhancement over the background at station i, averaged over a suitable time interval. Using meteorological analysis data from the Global Forecast System (GFS) model of the National Centers for Environmental Prediction (NCEP), monthly mean tropospheric scale heights were obtained by dividing the air column density up to the last pressure level below the tropopause height with the surface density. Northern and southern boundaries of our latitude bands are located at mid-points between station latitudes and at 20^o S and 90^o N. The results are not sensitive to changes of the southern boundary, since the measured concentrations in the Tropics remain very low until the end of May (see Fig. 2).

Figure 4 shows values of R for 16 five-day intervals. 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 by the decay correction. After 15 June, release estimates become entirely unrealistic (not shown). Estimates using the central ten 5-day intervals show relatively little variability, suggesting that the method works best during that period. The overall negative trend during that period 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. This would mean that values in

late May underestimate the total atmospheric $\Delta^{133}\mathrm{Xe}_c$ 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 11-15 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

$$R = R_s \frac{\sum_{i=1}^{N} \Delta^{133}\mathrm{Xe}_{c,i}}{\sum_{i=1}^{N} {}^{133}\mathrm{Xe}_{s,i}} \tag{2}$$

where R_s is the total ^{133}Xe release used in the model simulation (16.7 EBq) and $^{133}\mathrm{Xe}_{s,i}$ 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 5-day intervals, 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 surprisingly 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 the observation-based multi-box model, and the comparison of dispersion model results driven with GFS and ECMWF meteorological data with observation data, we have obtained total ^{133}Xe releases from FD-NPP of 16.7±1.9 EBq, 14.2±0.8 EBq and 19.0±3.4 EBq, respectively. Error bounds are the standard deviations from independent estimates based on nine subsequent 5-day periods. These values compare well to the inverse-modeling estimate by *Stohl et al.* (2012) of 15.3±3.1 EBq but it is difficult to say which estimate is most reliable.

All three of our estimates are higher than the calculated ^{133}Xe inventory of FD-NPP of 12.2 EBq (*NERH*, 2011), suggesting that either the inventory estimate is too low, or an additional source of ^{133}Xe is required. Adding the ^{133}Xe produced from the decay of ^{133}I, increases the effective FD-NPP ^{133}Xe inventory to 14.2 EBq ^{133}Xe, which corresponds to the lowest of our estimates. Our other two estimates and the result of *Stohl et al.* (2012) are, however, larger.

The findings of our study corroborate that the whole ^{133}Xe inventory of FD-NPP must have been released into the atmosphere. Furthermore, all the additional ^{133}Xe produced by the decay of ^{133}I must have been released as well. It seems that so far this has not been taken into account in other release estimates, except our own paper by *Stohl et al.* (2012). Our results provide some indication that even more ^{133}Xe might have been released, as two of our estimates and the result of *Stohl et al.* (2012) are higher than what can be explained by the combined inventories of ^{133}Xe and ^{133}I. Given the uncertainties of and differences between our estimates, this is not a robust finding, however. Nevertheless, one may wish to explore the possibility that additional ^{133}Xe sources either at FD-NPP or at other nuclear power plants

in Japan where the earthquake has triggered emergency shutdown. Another possibility is that the ^{133}Xe inventory estimates for FD-NPP are too low.

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Figure 1: Figure 1: Map showing the locations of stations used in this study. The location of FD-NPP is marked with a black rectangle.

Figure 2: Figure 2: Time series of observed ^{133}Xe concentrations at Yellowknife (top), Ashland (middle) and Darwin (bottom). Shown are the observed concentrations of ^{133}Xe (red squares), the observed concentrations with background subtracted Δ^{133}Xe (blue plusses), and the decay-corrected enhancements over the background $\Delta^{133}\mathrm{Xe}_c$ (black crosses). Measurements below the detection limit have been set to a constant value of 0.1 mBq m^{-3} for displaying purposes.

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

Figure 4: Figure 4: Estimates of total Δ^{133}Xe emissions from FD-NPP corrected to the time of the earthquake using all measurement data averaged over five-day intervals (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.

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