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

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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 (^{133}Xe) 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 ^{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. These values are substantially larger than the entire ^{133}Xe inventory of FD-NPP of about 12.2 EBq derived from calculations of nuclear fuel burn-up. Complete release of the entire ^{133}Xe inventory of FD-NPP and 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 differences between our estimates.

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 ^{133}Xe, 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 ^{133}Xe and Stohl et al. (2012) of 12.4 EBq ^{133}Xe 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 ^{133}Xe inventory may be higher. Nevertheless, the ^{133}Xe 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 ^{133}Xe. 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 ^{133}Xe (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

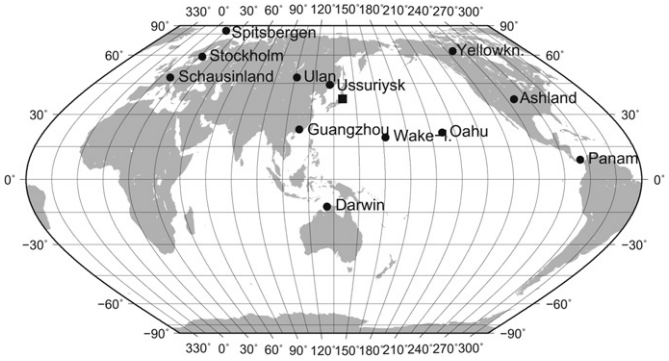

Fig. 1. Map showing the locations of stations used in this study. The location of FD-NPP is marked with a black rectangle. For lack of space, some station names are abbreviated: Ulan-Bator (Ulan), Wake-Island (Wake-I.), Panama (Panam), Yellowknife (Yellowkn).

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 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, $\Delta^{133}Xe_c$ (black crosses), increase until early April. After that, $\Delta^{133}Xe_c$ 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 $\Delta^{133}Xe_c$ 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 $\Delta^{133}Xe_c$ 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