Atmos. Chem. Phys. Discuss., 12, 12331–12356, 2012
www.atmos-chem-phys-discuss.net/12/12331/2012/
doi:10.5194/acpd-12-12331-2012

Atmospheric Chemistry and Physics Discussions

Atmospheric removal times of the aerosol-bound radionuclides ^{137}Cs and ^{131}I during the months after the Fukushima Dai-ichi nuclear power plant accident – a constraint for air quality and climate models

N. I. Kristiansen[1], **A. Stohl**[1], **and G. Wotawa**[2]

[1]Norwegian Institute for Air Research (NILU), Kjeller, Norway
[2]Central Institute for Meteorology and Geodynamics, Vienna, Austria

Received: 17 April 2012 – Accepted: 2 May 2012 – Published: 14 May 2012

Correspondence to: N. I. Kristiansen (nik@nilu.no)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Caesium-137 (^{137}Cs) and iodine-131 (^{131}I) are radionuclides of particular concern during nuclear accidents, because they are emitted in large amounts and are of significant health impact. ^{137}Cs and ^{131}I attach to the ambient accumulation-mode (AM) aerosols and share their fate as the aerosols are removed from the atmosphere by scavenging within clouds, precipitation and dry deposition. Here, we estimate their removal times from the atmosphere using a unique high-precision global measurement data set collected over several months after the accident at the Fukushima Dai-ichi nuclear power plant in March 2011. The noble gas xenon-133 (^{133}Xe), also released during the accident, served as a passive tracer of air mass transport for determining the removal times of ^{137}Cs and ^{131}I via the decrease in the measured ratios ^{137}Cs/^{133}Xe and ^{131}I/^{133}Xe over time. After correction for radioactive decay, the ^{137}Cs/^{133}Xe ratios reflect the removal of aerosols by wet and dry deposition, whereas the ^{131}I/^{133}Xe ratios are also influenced by aerosol production from gaseous ^{131}I. We find removal times for ^{137}Cs of 10.0–13.9 days and for ^{131}I of 17.1–24.2 days during April and May 2011. We discuss possible caveats (e.g. late emissions, resuspension) that can affect the results, and compare the ^{137}Cs removal times with observation-based and modeled aerosol lifetimes. Our ^{137}Cs removal time of 10.0–13.9 days should be representative of a "background" AM aerosol well mixed in the extratropical Northern Hemisphere troposphere. It is expected that the lifetime of this vertically mixed background aerosol is longer than the lifetime of AM aerosols originating from surface sources. However, the substantial difference to the mean lifetimes of AM aerosols obtained from aerosol models, typically in the range of 3–7 days, warrants further research on the cause of this discrepancy. Too short modeled AM aerosol lifetimes would have serious implications for air quality and climate model predictions.

ACPD
12, 12331–12356, 2012

1 Introduction

During nuclear accidents, radionuclides can be released into the atmosphere and transported over long distances. Caesium-137 (^{137}Cs) and iodine-131 (^{131}I) are the key radionuclides of greatest concern, because they are highly volatile and therefore quickly released into the environment, can be easily measured and constitute a significant risk to human health. While ^{131}I concentrations are important for the assessment of short-term exposure of the population, ^{137}Cs determines the long-term effect of a nuclear accident. Therefore, transport and removal of these species from the atmosphere are of major importance, as seen after the Chernobyl accident (NEA, 2002). These radionuclides attach mainly to the ambient accumulation-mode (AM) (~ 0.1–1 µm diameter) aerosols and share their fate during transport and removal from the atmosphere (Chamberlain, 1991). Thus, studies of these radionuclides are not only of interest per se, but can also be used to evaluate the behavior of AM aerosols which are detrimental for air quality and influence the global climate (Friedlander, 1977; Seinfeld and Pandis, 1998).

The radionuclide ^{137}Cs attaches mainly to the inorganic AM aerosol fraction, e.g. ammonium, sulfate and nitrate (Jost et al., 1986). For the emissions from the Fukushima Dai-ichi nuclear power plant accident in March 2011, there is direct evidence that the ^{137}Cs was attached to aerosols in the size range 0.1–2 µm diameter, identical to that of simultaneously measured sulfate aerosol, suggesting that ^{137}Cs primarily traced the fate of sulfate aerosol (Kaneyasu et al., 2012). Once attached, ^{137}Cs shares the fate of these aerosols, which grow by coagulation with other particles during transport (Jost et al., 1986) and are removed by wet and dry deposition. Thus, the removal rates of ^{137}Cs should be representative for the AM aerosols in general. If ^{137}Cs removal times can be determined from measurement data, this provides also a valuable constraint on the AM lifetime, for which otherwise few observational constraints exist. ^{131}I attaches to AM aerosols as well but, in contrast to ^{137}Cs, is released both as gas and in particulate form. The gaseous release fraction is typically as high as the particulate fraction. During

ACPD
12, 12331–12356, 2012

transport, there is an exchange between the gas and particle phases. Therefore, ^{131}I is less suitable for tracing the fate of non-volatile AM aerosols but can still impose upper limits on the AM aerosol lifetime and may also be considered as an indicator for the fate of semi-volatile aerosol species.

Reported aerosol lifetimes derived from ^{137}Cs and other radionuclides produced by cosmic rays, radon decay or nuclear bomb tests vary from 4 days to more than a month (Giorgi and Chameides, 1986), reflecting the different origin (e.g., surface or stratospheric) of radionuclide tracers. Aerosol residence times of $\leq$ 4 days in the lower troposphere and $\leq$ 12 days in the middle to upper troposphere may be seen as typical (Moore et al., 1973) but higher values of eight days for the lower troposphere have been reported as well (Papastefanou, 2006). Following the Chernobyl nuclear accident, the exponential decline of the ^{137}Cs concentrations indicated a residence time of 7 days (Cambray et al., 1987). Other observation-based methods not using radionuclide data suggest aerosol lifetimes from a few days to about one month in the troposphere (Williams et al., 2002; Paris et al., 2009; Schmale et al., 2011). Models give global average residence times of AM aerosol in the atmosphere on the order of 3–7 days (Chin et al., 1996; Feichter et al., 1996; Stier et al., 2005; Berglen et al., 2004; Liu et al., 2005; Bourgeois and Bey, 2011; Chung and Seinfeld, 2002; Koch and Hansen, 2005; Textor et al., 2006), which is rather shorter than the lifetimes obtained in most observation studies.

In this study we derive removal times for ^{137}Cs and ^{131}I in the atmosphere by using concentration measurements in a global network of background radionuclide monitoring stations operated by the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO; see Fig. 1) in Vienna. These measurements are unique, since the stations are globally nearly uniformly distributed, the data are globally inter-calibrated, and their high accuracy allows quantifying the radionuclide activity concentrations over several orders of magnitude. We used CTBTO measurements taken during three months following the accident at the Fukushima Dai-ichi nuclear power plant (FD-NPP) in March 2011 (Stohl et al., 2012), which released a pulse of

ACPD
12, 12331–12356, 2012

radionuclides to the atmosphere. The accident had been triggered by an earthquake on 11 March at 05:46 UTC and a related tsunami one hour later. While the earthquake led to an automatic emergency shutdown (scram) of the three running reactor blocks and the complete loss of off-site power, the tsunami caused the failure of the emergency cooling systems. Consequently, there was a rapid melt-down of the reactor cores and a massive injection of radionuclides into the atmosphere.

During the accident, the whole inventory of the noble gas ^{133}Xe was set free from the reactors. Xenon measurements are available from a subset (about 50 %) of the CTBTO stations measuring ^{137}Cs and ^{131}I. While both ^{137}Cs and ^{131}I attach to the ambient AM aerosol, the noble gas ^{133}Xe can be considered as a passive tracer of air mass transport. Thus, the aerosol removal times can be directly determined, to the authors' knowledge for the first time, by the decrease in the concentration ratios between two aerosols (^{137}Cs, ^{131}I) and a passive tracer (^{133}Xe) as a function of time. We compare the obtained removal times with observation-based and modeled aerosol lifetimes and discuss the implication of using the removal times as an estimate of AM aerosol lifetime.

2 Data and methods

We have used measurements of atmospheric activity concentrations of the noble gas ^{133}Xe and the aerosol-bound radionuclides ^{137}Cs and ^{131}I available from several stations operated by the CTBTO covering the whole Northern Hemisphere (Fig. 1). The stations are part of the International Monitoring System built up during the last 15 yr to measure signals (seismic, hydroacoustic, infrasound and radionuclides) from underground or atmospheric nuclear explosions. Measurements in the time period August 2010 to December 2011 were used for this study. The CTBTO stations are equipped with high-volume aerosol samplers. About 20 000 m^3 of air is blown through a filter, collecting particulate radionuclides with a collection period of 24 h. ^{137}Cs, ^{131}I and other aerosol-bound radionuclides are measured with high-resolution germanium detectors

ACPD
12, 12331–12356, 2012

(Schulze et al., 2000; Medici, 2001). The minimum detectable activity concentration (MDC) of ^{137}Cs varies for the different stations, but is on average about 1 µBq m^{-3}. A similar MDC is obtained for ^{131}I. CTBTO stations, however, only measure the aerosol-bound ^{131}I fraction, because the gaseous fraction is not trapped by the currently used filters. About 50 % of the CTBTO stations are also equipped with xenon detectors which measure four radioxenon isotopes. The isotope ^{133}Xe is the most prevalent and important one and therefore used here. The measurement accuracy for ^{133}Xe also varies for the different stations, but is on average about 0.1 mBq m^{-3}, and the collection period is typically 12 h.

Atmospheric background levels of ^{137}Cs, ^{131}I and ^{133}Xe were defined as the mean activity concentrations before the FD-NPP accident (August 2010 to 11 March 2011), and subtracted from all measurement values after the FD-NPP accident. The impact of this correction was negligible at most stations because of very low background values which rarely exceed the level of detection. The measured ^{133}Xe (half-life 5.25 days), ^{137}Cs (half-life of 30 yr) and ^{131}I (half-life 8.02 days) enhancements over the background were corrected for radioactive decay to the time of the earthquake. Figure 2 shows an example of the uncorrected and corrected time series data for the station Oahu. It is seen that the emission pulse of ^{137}Cs and ^{131}I is observable at this station until late May. The background of all three radionuclides is very low so the effect of the background subtraction is negligible for ^{137}Cs and ^{131}I while a small effect can be seen for ^{133}Xe in late May when the enhanced values are slightly lower than the uncorrected values.

The low background in combination with the high measurement sensitivity facilitates quantification of the radionuclides over a period of almost three months. This is long compared to the period of major emissions of about eight and four days for ^{137}Cs and ^{133}Xe, respectively (Stohl et al., 2012). Emissions of ^{131}I had a similar temporal behavior as ^{137}Cs (Chino et al., 2011), thus allowing us to consider all the radionuclide emissions as one single pulse. Although the ^{133}Xe emissions ceased before the ^{137}Cs and ^{131}I emissions, the highest emissions occurred during the same three days (Stohl et al., 2012; Chino et al., 2011), ensuring a high level of correlation between

ACPD
12, 12331–12356, 2012

the radionuclides. Thus, the removal of ^{137}Cs and ^{131}I can be gauged against an inert noble gas (^{133}Xe) tracer.

We use two different approaches to estimate the removal times of the aerosol-borne radionuclides. For both approaches the emissions of ^{137}Cs, ^{131}I and ^{133}Xe are treated as a single pulse, referenced to the time of the earthquake, allowing to determine the age (days) of the air mass containing the radionuclides when it reached each CTBTO station. Since all measurement samples use the same reference date, the removal time calculation does not depend on the assumed emission time, so the time of the earthquake was chosen for convenience.

The first approach uses a multi-box model to estimate the total atmospheric burden of ^{137}Cs, ^{131}I and ^{133}Xe. If we assume that the measured ^{137}Cs 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 atmospheric burden of ^{137}Cs follows from

$$[^{137}\mathrm{Cs}] = \sum_{i=1}^{N} A_i \times H_i \times {}^{137}\mathrm{Cs}_i \quad (1)$$

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 $^{137}\mathrm{Cs}_i$ the decay-corrected enhancement over the background at station i, averaged over a suitable time interval (here 4 days). Likewise, the calculation is done for the total atmospheric burdens of ^{133}Xe and ^{131}I. 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. The box model extended from 20° S to 90° N; latitudes of a few stations were shifted by a maximum of 3° latitude, to reduce clustering of stations at particular latitudes. The box model assumption that radionuclides are relatively well mixed within latitude bands is not fulfilled during the first weeks after the accident, so it was only used from 1 April. The results were not sensitive to variations of this date.

ACPD
12, 12331–12356, 2012

The second approach takes direct advantage of co-located measurements of ^{137}Cs, ^{131}I and ^{133}Xe without using the multi-box model. Only stations measuring all three radionuclides were used in this approach (Fig. 1). The 12-hourly ^{133}Xe data (after background subtraction and decay correction) were linearly interpolated to the sampling times (24 h) of the ^{137}Cs or ^{131}I data in order to calculate the ratio of the radionuclides. In addition to using all measurement data points individually, we also calculated the ratios of the mean activity concentrations for each day over all stations following

$$\omega(t) = \frac{\sum_{j=1}^{J} {}^{137}\mathrm{Cs}(t)}{\sum_{j=1}^{J} {}^{133}\mathrm{Xe}(t)} \quad (2)$$

where J is the number of stations performing simultaneous measurements at sample time t. This considerably reduces the scatter found for individually measured ratios and is more similar to the multi-box model approach.

The ratios ^{137}Cs/^{133}Xe and ^{131}I/^{133}Xe (and similarly, values of ω and the ratios for the atmospheric burdens) decrease with time as ^{137}Cs and ^{131}I are getting removed from the atmosphere, whereas ^{133}Xe is conserved. The time scale of the decrease (referred to as the removal time throughout this paper) is calculated based on a fitted trend line through the data, assuming that the data follow a model of exponential decay

$$\frac{{}^{137}\mathrm{Cs}(t)}{{}^{133}\mathrm{Xe}(t)} = \varepsilon \times \exp(-t/\tau) \quad (3)$$

where t is the sample time, τ is the removal time, and ε is the effective emission ratio at the time assumed for the emission pulse. Equally, the calculations were done using ^{131}I measurements. The fraction of variance in the ratios explained by the exponential model is given by the squared correlation coefficient R^2. Since all measurements are

ACPD
12, 12331–12356, 2012

referenced to the time of the earthquake, the actual temporal variability of the emissions is not considered, however it was accounted for in a sensitivity study included in Sect. 4.

3 Results

In the first approach, we estimate the total atmospheric burdens of ^{137}Cs, ^{131}I and ^{133}Xe, [^{137}Cs], [^{131}I] and [^{133}Xe], using the simple multi-box model. The suitability of the underlying assumption that the FD-NPP emissions of ^{133}Xe are relatively well mixed in latitude bands is confirmed by the small variation in [^{133}Xe] with time (Fig. 3a, blue line). The variability is particularly small after day 30 (10 April) when enough time had passed for a nearly complete mixing in the extratropics and before day 60 (10 May), after which measurement uncertainty and/or background subtraction are becoming more substantial due to the fact that radioactive decay has removed most of the FD-NPP emission pulse. This also demonstrates that ocean uptake of the slightly water-soluble ^{133}Xe is negligible on the time scale considered. The small overall decrease with time is more likely due to leakage of ^{133}Xe into the stratosphere and into the Southern Hemisphere, outside of the box model domain.

In contrast to [^{133}Xe], [^{137}Cs] decreases with time due to wet and dry deposition (Fig. 3a, green line). By fitting an exponential model to the change of [^{137}Cs] with time we find the time scale τ_b of ^{137}Cs removal from the atmosphere to be 9.3 days (95 % confidence interval (C.I.) 8.7–10.0 days). A more correct estimate of τ_b can be obtained from the ratio [^{137}Cs]/[^{133}Xe], as leakages to the stratosphere and Southern Hemisphere, which are not considered in the multi-box model, would affect estimates of [^{137}Cs] and [^{133}Xe] similarly. This gives a slightly longer τ_b of 10.0 days (C.I. 9.3–10.9) (Fig. 3a, black line). Likewise for ^{131}I, τ_b is found to be 15.3 days (C.I. 12.8–18.9) from the decrease of the [^{131}I]/[^{133}Xe] ratio (Fig 3b).

The other approach for estimating the removal time of ^{137}Cs and ^{131}I takes advantage of directly co-located measurements of the radionuclides. For each pair of simultaneous measurements, we calculated the ^{137}Cs/^{133}Xe and ^{131}I/^{133}Xe ratios over

ACPD
12, 12331–12356, 2012

time, decay-corrected and referenced to the time of the earthquake. The decrease of the $^{137}Cs/^{133}Xe$ and $^{131}I/^{133}Xe$ ratios vary for the different measurement stations and removal times range from 8.8 to 18.1 days for ^{137}Cs and 11.1 to 26.1 days for ^{131}I (Table 1). For ^{137}Cs, the shortest removal times (< 10 days) are found for the tropical sites (Wake Island and Oahu) which are affected by strong wet scavenging due to tropical precipitation. For Ulan-Bator, located at high altitude (~ 1300 m above sea level), the short removal time can probably be explained by the transport across high mountain chains and strong scavenging due to orographic precipitation. For the North American stations, the removal times for ^{137}Cs are longer and range from 13.1 to 18.1 days. The European stations give homogenous results with a removal time around 15 to 16 days. For Ussuriysk, a station mostly upwind but closest to FD-NPP, the removal time estimate has a large uncertainty range because variations in the $^{137}Cs/^{133}Xe$ emission ratio were occasionally transferred to the station directly without the damping effect of air mass mixing during long-range transport, which tends to eliminate short-time variations of the emission ratio. For ^{131}I the picture is not as clear because gas-to-particle exchange during transport is playing a role (see Sect. 4).

Combining the global set of measurements from all stations, the $^{137}Cs/^{133}Xe$ ratios decrease with a time scale τ_a = 13.9 days (C.I. 12.8–15.2 days) (Table 1, Fig. 3c, grey line). This removal time estimate has larger uncertainty than the multi-box model estimate due to more scatter in the individual data points, which probably also explains the somewhat longer removal time obtained. Applying an averaging of the activity concentrations for each day over all stations according to Eq. (2) (ω in Fig. 3c), we obtain a removal time τ_ω = 12.6 days (C.I. 11.8–13.6 days), a result closer to the box model estimate. For ^{131}I, the removal times from the decay of the $^{131}I/^{133}Xe$ ratios are τ_a = 17.1 days (C.I. 15.1–18.8 days), and τ_ω = 24.2 days (C.I. 20.5–29.7 days) (Table 1, Fig. 3d)

Overall, our different estimates for the removal times for ^{137}Cs are τ_b = 10.0, τ_a = 13.9 and τ_ω = 12.6 days, and for ^{131}I τ_b = 15.3, τ_a = 17.1 and τ_ω = 24.2 days (Table 1).

ACPD
12, 12331–12356, 2012

The three different estimates for each radionuclide agree approximately within the statistical uncertainty ranges.

4 Discussion

To assess the impact of some of the assumptions made in the analysis, we carried out two sensitivity tests for the method using the direct measurements of ^{137}Cs and ^{133}Xe. First, we used only measurement data after 1 April. This represents a relatively well-mixed case, as an emission pulse from East Asia typically is quite homogeneously mixed across the troposphere in the extratropical Northern Hemisphere, both zonally as well as vertically, after 25–30 days (see Fig. 2–4 in Stohl et al., 2002). In this well-mixed case the different temporal shape in the emissions of ^{137}Cs and ^{133}Xe gets less important. Figure 4a shows that τ_a = 14.3 days (C.I. 13.0–15.9 days) and τ_∞ = 12.9 days (C.I. 12.1–13.7 days) is obtained. These results are not significantly different from the removal times obtained when using the complete measurement data set.

For the second sensitivity test we used emission times and plume age calculations determined by the Lagrangian particle dispersion model FLEXPART (Stohl et al., 1998, 2005). Using the sensitivity of the modeled concentrations corresponding to each measurement sample to the emissions and time-varying source terms of ^{137}Cs (Stohl et al., 2012), we determined the modeled emission contributions to each measurement sample as a function of emission time. Only measurement data which the model clearly associated with the emissions from FD-NPP were used. Here, the effective emission time for a given measurement sample was considered to be the time with the highest emission contribution of ^{137}Cs. Before fitting the exponential model, time was counted relative to the effective emission time of every measurement sample. The resulting removal times (Fig. 4b) of τ_a = 13.5 days (C.I. 12.5–14.7) and τ_∞ = 11.9 days (C.I. 11.2–12.7 days) are again not significantly different from the results when using a single reference time for the emissions.

ACPD
12, 12331–12356, 2012

The longer removal time for ^{131}I than for ^{137}Cs is expected since ^{131}I has a large gaseous fraction, and the gas-particle conversion leads to the formation of new particulate iodine from gaseous iodine during the transport. Irrespective of the lifetime of gaseous ^{131}I, on-going particulate ^{131}I generation can only lead to longer removal times for aerosol-bound ^{131}I in our analysis. The relatively large difference in ^{131}I and ^{137}Cs lifetimes, however, suggests that the lifetime of gaseous ^{131}I must be longer than that of the AM aerosol carrying the particulate ^{131}I and ^{137}Cs. However, since we have no measurements of gaseous ^{131}I available, we cannot directly prove this. Nevertheless, the removal time derived from the ^{131}I data can only be considered as an upper estimate for the AM aerosol lifetime. Due to these complicating factors related to ^{131}I, most of the further discussion focuses on the results obtained for ^{137}Cs.

We can estimate the initial ^{137}Cs/^{133}Xe ratio at the reference time from the exponential model fitted to the data by extrapolating the fitted curve to time zero (the intercept value). From the box-model, the initial ^{137}Cs/^{133}Xe ratio is 7.2×10^{-5} and the method using the direct measurements gives an initial ratio of around 2×10^{-5}. These initial ratios are up to 2 order of magnitude lower than the ratio of the total releases of ^{137}Cs and ^{133}Xe found by Stohl et al. (2012) using an inversion technique (36.6×10^{15} Bq/15.3×10^{18} Bq ^{137}Cs/^{133}Xe, giving an emission ratio of 2.4×10^{-3}). Their total ^{137}Cs emissions have an estimated uncertainty of about 50 %, while ^{133}Xe emissions should be accurate to within 20 %. For ^{131}I we obtain an initial ^{131}I/^{133}Xe ratio of about 1×10^{-3} from our two methods. Compared to the ratio of the total release estimates of Chino et al. (2011) for ^{133}I and Stohl et al. (2012) for ^{133}Xe, of 1×10^{-2} (150×10^{15} Bq/15.3×10^{18} Bq ^{131}I/^{133}Xe), our estimate is roughly one order of magnitude lower.

The fact that our radionuclide ratios extrapolated to the time of the are so much lower than those reported for the emission ratios (even considering uncertainties in the emission ratios), suggests that this extrapolation is not valid. There are indeed indications in our data that the initial removal rates of ^{137}Cs and ^{131}I were higher than the removal rates encountered in the well-mixed situation afterwards. Figures 3c, d and, especially,

ACPD
12, 12331–12356, 2012

Fig. 4b show that most of the first few data points deviate strongly upward from the exponential model curve. Unfortunately, the initial phase of plume dispersion was not sampled by the CTBTO network and it is therefore not possible to derive removal rates or lifetimes for the first few days after emission. However, there are some potential reasons for higher initial removal rates, namely the fact that there was strong precipitation co-located with the plume during the period of the highest emissions leading to strong scavenging of the plume immediately after its emission (Stohl et al., 2012), and the fact that the initial plume was close to the ground, thus facilitating effective dry deposition. Furthermore, hot particles (particles that carry very high radioactivity, e.g., fragments of the nuclear fuel) were present in the FD-NPP plume (Paatero et al., 2012). Hot particles can be much larger than AM aerosols (Paatero et al., 2010) and, thus, deposit much more quickly, e.g., by gravitational settling. Thus, our derived AM aerosol lifetimes must be considered valid for a well-mixed background AM aerosol, whereas the lifetime of an aerosol emitted from the ground may be substantially shorter.

The major uncertainty factor with regard to our removal time estimate is the possibility of additional releases of radionuclides long after our assumed reference time. These additional releases can be either direct late emissions from FD-NPP or indirect releases by resuspension of deposited radionuclides. First we discuss the possibility that our results are influenced by resuspension. It has been seen that resuspension was important after the Chernobyl accident (Garland and Pomeroy, 1994), and monitoring data from Japan suggest resuspension occurred also there after the FD-NPP accident (Stohl et al., 2012). However, the data also show that the ^{137}Cs concentrations in Japan are lower by two to three orders of magnitude in between major plume passages, suggesting that resuspension was quite limited. Applying a ^{137}Cs resuspension rate $\gamma = 1 \times 10^{-9}\,s^{-1}$, typical for summer conditions in central Russia (Makhon'ko, 1979 as cited by Gavrilov et al., 1995), and accounting for the fact that only 20 % of the FD-NPP emissions were deposited over land (Stohl et al., 2012), we estimate that a fraction of 1.7×10^{-4} of the originally emitted ^{137}Cs could be resuspended in a 10-day period. For a 10-day removal time, we find that a fraction of 3.3×10^{-4} of the originally

ACPD
12, 12331–12356, 2012

emitted ^{137}Cs would still be suspended in the atmosphere after 80 days. Thus, even for the shortest removal times obtained in our study and at the very end of our study period, resuspension could account for only about half of the ^{137}Cs mass suspended in the atmosphere. For the moist climate of Japan, which received 90 % of the FD-NPP fall-out over land (Stohl et al., 2012), the resuspension rate is likely to be lower than the value used above. Even more importantly, long-range transport of resuspended ^{137}Cs to our monitoring stations is highly unlikely because of the larger particle sizes typical of resuspended material. In the surroundings of Chernobyl, ^{137}Cs activity size distributions with median diameters of 5–10 µm were measured for resuspended material (Garger et al., 1998). Such particles have a very short residence time in the atmosphere compared to AM aerosols and cannot be transported far from the source. Finally, there was also no indication of an elevated background level of ^{137}Cs after July, compared to the levels before the FD-NPP accident, at any of the stations. This implies that resuspension, which occurs over much longer time scales than aerosol deposition (Maxwell and Anspaugh, 2011) and should still be observable after July if it was important during the period of our study, did not impact the measurements used in this study.

Secondly, we discuss if direct late emissions affect our results. One study (Stohl et al., 2012) found ^{137}Cs emissions in late March to be about two orders of magnitude smaller than during the first week, and emissions were decreasing further in April, but there is a possibility that ^{137}Cs emissions had not ceased completely in April. We have carefully screened the measurement data (including also Japanese data as presented in Stohl et al., 2012) for any evidence for a late emission pulse from FD-NPP and found one possible event in late May seen at the station Ussuriysk that would be large enough to affect our results. We have excluded these data and ended the period of our study on 25 May to avoid impacts of this event on other stations. However, late emissions on the order of only 1 % of the maximum emission in the early phase could theoretically contribute to a large part to the measured activity concentrations in late May, and thus affect the estimated removal times. However, no other study has suggested strong late

ACPD
12, 12331–12356, 2012

emissions of ^{137}Cs. Regarding ^{131}I, there is also no evidence for late emissions from FD-NPP.

Lastly, we compare our estimated removal times of the aerosol-bound radionuclides to aerosol lifetimes reported in other studies. Observation-based estimates of aerosol lifetimes are sparse, range from less than 4 days to more than a month (Moore et al., 1973; Cambray et al., 1987; Papastefanou, 2006; Williams et al., 2002; Paris et al., 2009; Schmale et al., 2011), and are associated with substantial uncertainties. According to the existing aerosol models the average residence times of AM aerosol in the atmosphere are on the order of 3–7 days (Chin et al., 1996; Feichter et al., 1996; Stier et al., 2005; Berglen et al., 2004; Liu et al., 2005; Bourgeois and Bey, 2011; Chung and Seinfeld, 2002; Koch and Hansen, 2005; Textor et al., 2006). The aerosol lifetimes vary regionally and are generally longest in dry or cold regions (Koch et al., 1996). Modeled lifetimes longer than 10 days were obtained primarily for aerosols originating from the stratosphere (Koch et al., 1996).

Our removal times for ^{137}Cs and associated AM aerosol lifetimes range from 10 to 14 days and are compatible with the much larger range of aerosol lifetimes given in previous observation-based studies, but they are not consistent with the 3–7 days annual global averages obtained from the aerosol models. The difference cannot be explained by the fact that our study extended only over 80 days and covered only the Northern Hemisphere. The emissions were exposed to extratropical cyclones and experienced strong lifting in the North Pacific storm track (Stohl et al., 2012), a region where and during a time of the year when storm activity is considerably enhanced. This should have caused stronger-than-average rather than weaker-than-average wet scavenging of aerosols.

One possible explanation for the discrepancy between our estimated removal times and those reported for aerosol models is the different aerosol distribution. Our estimated removal times should be representative for AM aerosol which is already reasonably well distributed in the atmosphere (background aerosol), whereas aerosol models report lifetimes for aerosols which are either emitted at the surface, or formed mainly

ACPD
12, 12331–12356, 2012

in the boundary layer. Indeed, our comparison to the reported emission ratios for the FD-NPP accident suggests that aerosol removal rates must have been much larger (and, thus lifetimes shorter) shortly after the emission, compared to the later period. However, the comparison between our results and modelled aerosol lifetimes may still indicate that the lifetimes in the aerosol models are too short. This certainly deserves further clarification and investigation, for example by running the major aerosol models directly against the FD-NPP accident case, or at least specifically for a well-mixed aerosol that is more comparable to the FD-NPP accident case than the AM aerosol tracers for which lifetimes are normally reported.

5 Conclusions

The removal times of the aerosol-borne radionuclides ^{137}Cs and ^{131}I have been quantified by using a global set of measurements which recorded the activity concentrations following the release of both nuclides from the FD-NPP accident in March 2011. The radioactive noble gas ^{133}Xe also released during the accident served as a tracer of the atmospheric transport. The main findings from this study are summarized as follows:

- A removal time for ^{137}Cs of 10.0–13.9 days and for ^{131}I of 17.1–24.2 days was estimated from the ratios ^{137}Cs/^{133}Xe and ^{131}I/^{133}Xe, respectively. The removal times can serve as estimates of accumulation-mode (AM) aerosol lifetimes since the radionuclides attach to AM aerosols and trace their fate during transport and removal.
- The longer removal times for ^{131}I were affected by the gas-to-particle exchange that occurs during transport while no such effect influences ^{137}Cs which thus gives a better estimate of AM aerosol lifetimes. Lifetimes derived with ^{131}I must be considered as upper estimates.
- The removal rates must have been higher (and, thus, removal times shorter) during the initial phase of the plume transport that was not captured by our

ACPD
12, 12331–12356, 2012

measurements. This can be seen by too low aerosol/noble gas radionuclide ratios obtained when extrapolating the exponential fit back to the time of the accident, compared to reported emission ratios. The same is suggested by an upward deviation of the first few measurement data points from the exponential model fit. Therefore, the estimated removal times are valid only for an aerosol reasonably well mixed in the troposphere (a background aerosol) and not for fresh aerosol directly emitted from the ground.

- Our results are highly sensitive to possible late emissions of radionuclides. However, there is no evidence for such late emissions, neither in our data nor in the existing literature on the FD-NPP accident
- The effect of resuspension on the estimated removal times was likely negligible mainly due to the fact that resuspension is much smaller than the initial emission pulse and encompasses larger particles than AM aerosols.
- The estimated removal times are consistent with the large range of previous observation-based aerosol lifetimes, but they are about a factor of two higher than published model-based estimates on AM aerosol lifetimes in the atmosphere. The difference points towards a too quick removal of AM aerosol in models
- Our study should serve as encouragement for aerosol modelers to run their models against the FD-NPP accident case.

Appendix A

Statistical methods

From Eq. (3), the coefficients of the exponential model are ε (intercept) and $1/\tau$ (slope). The 95 % confidence interval (C.I.) for the removal time τ (Table 1) is obtained by taking the inverse of the upper and lower C.I. limits of the slope. This inverse-conversion

ACPD
12, 12331–12356, 2012

changes the distribution so that τ it is not longer normally distributed, thus τ is not the centre of the C.I. but can have a long tail towards longer τ (this is seen, e.g., at Ashland). The standard deviation for τ is not obtained due to the inverse-conversion. Therefore, only confidence intervals are reported throughout this paper, and not standard deviations.

Acknowledgement. We would like to thank all the scientists who produced the CTBTO measurement data. The research leading to these results has received partial funding from the European Union Seventh Framework Programme (FP7/2007-2013) under grant agreement no 282688 – ECLIPSE.

References

Berglen, T., Berntsen, T. K., Isaksen, I. S. A., and Sundet, J. K.: A global model of the coupled sulfur/oxidant chemistry in the troposphere: the sulfur cycle, J. Geophys. Res., 109, D19310, doi:10.1029/2003JD003948, 2004.

Bourgeois, Q. and Bey, I.: Pollution transport efficiency toward the Arctic: sensitivity to aerosol scavenging and source regions, J. Geophys. Res., 116, D08213, doi:10.1029/2010JD015096, 2011.

Cambray, R. S., Cawse, P. A., Garland, J. A., Gibson, J. A. B., Johnson, P., Lewis, G. N. J., Newton, D., Salmon, L. and Wade, B. O.: Observations on radioactivity from the Chernobyl accident, Nucl. Energ. 26, 77–101, 1987.

Chamberlain, A. C.: Radioactive Aerosols, Cambridge University Press, Cambridge, 1991.

Chin, M., Jacob, D. J., Gardner, G. M., Foreman-Fowler, M. S., Spiro, P. A., and Savoie, D. L.: A global three-dimensional model of tropospheric sulfate, J. Geophys. Res., 101, 18667–18690, 1996.

Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G., and Yamazawa, H.: Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from the Fukushima Dai-ichi nuclear power plant into the atmosphere, J. Nuc. Sci. Tech., 48, 1129–1134, 2011.

Chung, S. H. and Seinfeld, J. H.: Global distribution and climate forcing of carbonaceous aerosols, J. Geophys. Res., 107(D19), 4407, doi:10.1029/2001JD001397, 2002.

ACPD

12, 12331–12356, 2012

Feichter, J., Kjellstrom, E., Rodhe, H., Dentener, F., Lelieveld, J., and Roelofs, G.-J.: Simulation of the tropospheric sulphur cycle in a global climate model, Atmos. Environ., 30, 1693–1707, 1996.

Friedlander, S. K.: Smoke, Dust and Haze: Fundamentals of Aerosol Behavior, New York, Wiley-Interscience, 333 pp., 1977.

Garger, E. K., Kashpur, V., Paretzke, H. G., and Tschiersch, J.: Measurement of resuspended aerosol in the Chernobyl area. Part II. Size distribution of radioactive particles, Radiat. Environ. Biophys., 36, 275–283, 1998.

Garland, J. A. and Pomeroy, I. R.: Resuspension of fallout material following the Chernobyl accident, J. Aerosol Sci., 25, 793–806, 1994.

Gavrilov, V. P., Klepikova, N. V., Troyanova, N. I., and Rodean, H. C.: Stationary model for resuspension of radionuclides and assessment of ^{137}Cs concentration in the near-surface layer for the contaminated areas in the Bryansk region of Russia and Belarus, Atmos. Environ., 29, 2633–2650, 1995.

Giorgi, F. and Chameides, W. I.: Rainout lifetimes of highly soluble aerosols and gases as inferred from simulations with a general circulation model, J. Geophys. Res., 91, D13, 14367–14376, 1986.

Jost, D. T., Gäggeler, H. W., Baltensperger, U., Zinder, B., and Haller, P.: Chernobyl fallout in size-fractionated aerosol, Nature, 324, 22–23, 1986.

Kaneyasu, N., Ohashi, H., Suzuki, F., Okuda, T., and Ikemori, F.: Sulfate aerosol as a potential transport medium of radiocesium from the Fukushima nuclear accident, Environ. Sci. Technol., doi:10.1021/es204667h, in press, 2012.

Koch, D. and Hansen, J.: Distant origins of Arctic black carbon, a Goddard Institute for Space Studies ModelE experiment, J. Geophys. Res., 110, D04204, doi:10.1029/2004jd005296, 2005.

Koch, D. M., Jacob, D. J., and Graustein, W. C.: Vertical transport of tropospheric aerosols as indicated by ^{7}Be and ^{210}Pb in a chemical tracer model, J. Geophys. Res., 101(D13), 18651–18666, doi:10.1029/96JD01176, 1996.

Liu, X., Penner, J. E., and Herzog, M.: Global modeling of aerosol dynamics: model description, evaluation, and interactions between sulfate and nonsulfate aerosols, J. Geophys. Res., 110, D18206, doi:10.1029/2004JD005674, 2005.

Maxwell, R. and Anspaugh, L. R.: An improved model for prediction of resuspension, Health Phys., 101, 722–730, 2011.

ACPD

12, 12331–12356, 2012

Medici, F.: The IMS radionuclide network of the CTBT, Radiat. Phys. Chemi., 161, 689–690, 2001.
Moore, H. E., Poet, S. E., and Martell, E. A.: ^{222}Rn, ^{210}Pb, ^{210}Bi, and ^{210}Po profiles and aerosol residence times versus altitude, J. Geophys. Res., 78, 7065–7075, 1973.
Nuclear Energy Agency (NEA): Chernobyl: Assessment of Radiological and Health Impacts – 2002 Update of Chernobyl: Ten Years On, Organisation for Economic Co-Operation and Development (OECD) Publications, Paris, 155 pp., 2002.
Paatero, J., Hämeri, K., Jaakkola, T., Jantunen, M., Koivukovski, J., and Saxén, R.: Airborne and deposited radioactivity from the Chernobyl accident – a review of investigations in Finland, Boreal Env. Res. 15, 19–33, 2010.
Paatero, J., Vira, J., Siitari-Kauppi, M., Hatakka, J., Holmén, K., and Viisanen, Y.: Airborne fission products in the high Arctic after the Fukushima nuclear accident, J. Environ. Radioact., in press, 2012.
Papastefanou, C.: Residence time of tropospheric aerosols in association with radioactive nuclides, Appl. Radiat. Isot., 64, 93–100, doi:10.1016/j.apradiso.2005.07.006, 2006.
Paris, J.-D., Stohl, A., Nédélec, P., Arshinov, M. Yu., Panchenko, M. V., Shmargunov, V. P., Law, K. S., Belan, B. D., and Ciais, P.: Wildfire smoke in the Siberian Arctic in summer: source characterization and plume evolution from airborne measurements, Atmos. Chem. Phys., 9, 9315–9327, doi:10.5194/acp-9-9315-2009, 2009.
Schmale, J., Schneider, J., Ancellet, G., Quennehen, B., Stohl, A., Sodemann, H., Burkhart, J. F., Hamburger, T., Arnold, S. R., Schwarzenboeck, A., Borrmann, S., and Law, K. S.: Source identification and airborne chemical characterisation of aerosol pollution from long-range transport over Greenland during POLARCAT summer campaign 2008, Atmos. Chem. Phys., 11, 10097–10123, doi:10.5194/acp-11-10097-2011, 2011.
Schulze, J., Auer, M., and Werzi, R.: Low level radioactivity measurement in support of the CTBTO, Appl. Radiat. Isot., 53, 23–30, 2000.
Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, New York, John Wiley and Sons, 1998.
Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., and Petzold, A.: The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125–1156, doi:10.5194/acp-5-1125-2005, 2005.

ACPD
12, 12331–12356, 2012

Stohl, A., Hittenberger, M., and Wotawa, G.: Validation of the Lagrangian particle dispersion model FLEXPART against large scale tracer experiment data, Atmos. Environ., 32, 4245–4264, 1998.

Stohl, A., Eckhardt, S., Forster, C., James, P., and Spichtinger, N.: On the pathways and timescales of intercontinental air pollution transport, J. Geophys. Res., 107, 4684, doi:10.1029/2001JD001396, 2002.

Stohl, A., Forster, C., Frank, A., Seibert, P., and Wotawa, G.: Technical note: The Lagrangian particle dispersion model FLEXPART version 6.2, Atmos. Chem. Phys., 5, 2461–2474, doi:10.5194/acp-5-2461-2005, 2005.

Stohl, A., Seibert, P., Wotawa, G., Arnold, D., Burkhart, J. F., Eckhardt, S., Tapia, C., Vargas, A., and Yasunari, T. J.: Xenon-133 and caesium-137 releases into the atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the source term, atmospheric dispersion, and deposition, Atmos. Chem. Phys., 12, 2313–2343, doi:10.5194/acp-12-2313-2012, 2012.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, I., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 6, 1777–1813, doi:10.5194/acp-6-1777-2006, 2006.

Williams, J., de Reus, M., Krejci, R., Fischer, H., and Ström, J.: Application of the variability-size relationship to atmospheric aerosol studies: estimating aerosol lifetimes and ages, Atmos. Chem. Phys., 2, 133–145, doi:10.5194/acp-2-133-2002, 2002.

ACPD

12, 12331–12356, 2012

Table 1. Removal times (τ) for ^{137}Cs and ^{131}I estimated from the exponential decay of measured ^{137}Cs/^{133}Xe and ^{131}I/^{133}Xe ratios. τ_a when combining measurements from all 11 stations as in Fig. 3c, d (grey trend lines), τ_ω as calculated from the ratios ω (the mean activity concentrations for each day averaged over all stations using Eq. 2), as in Fig. 3c, d (black trend lines), and τ_b from the ratios obtained with the box model (Fig. 3a, b). The 95 % confidence intervals for the removal times are given (see Appendix A).

Station	τ (^{137}Cs)	95 % confidence interval	τ (^{131}I)	95 % confidence interval
Wake Island	8.8	[6.4,13.9]	15.2	[11.2,23.6]
Oahu	9.6	[7.7,12.9]	14.8	[12.0,19.1]
Ulan-Bator	8.8	[7.4,10.8]	11.2	[9.4,13.9]
Ussuriysk	14.1	[8.3,47.6]	11.1	[9.2,14.0]
Ashland	18.1	[13.6,27.2]	26.1	[17.5,51.2]
Charlottesville	15.7	[12.2,22.0]	14.5	[11.4,20.0]
Yellowknife	13.1	[11.7,14.9]	18.3	[15.5,22.5]
St. Johns	14.3	[11.0,20.8]	12.7	[10.2,16.8]
Schauinsland	16.2	[12.6,22.6]	11.2	[9.2,14.6]
Stockholm	15.3	[11.0,25.1]	24.7	[17.7,41.2]
Spitsbergen	15.1	[12.3,19.4]	20.4	[16.6,26.4]
τ_a	13.9	[12.8,15.2]	17.1	[15.7,18.8]
τ_ω	12.6	[11.8,13.6]	24.2	[20.5,29.7]
τ_b	10.0	[9.3,10.9]	15.3	[12.8,18.9]

ACPD
12, 12331–12356, 2012

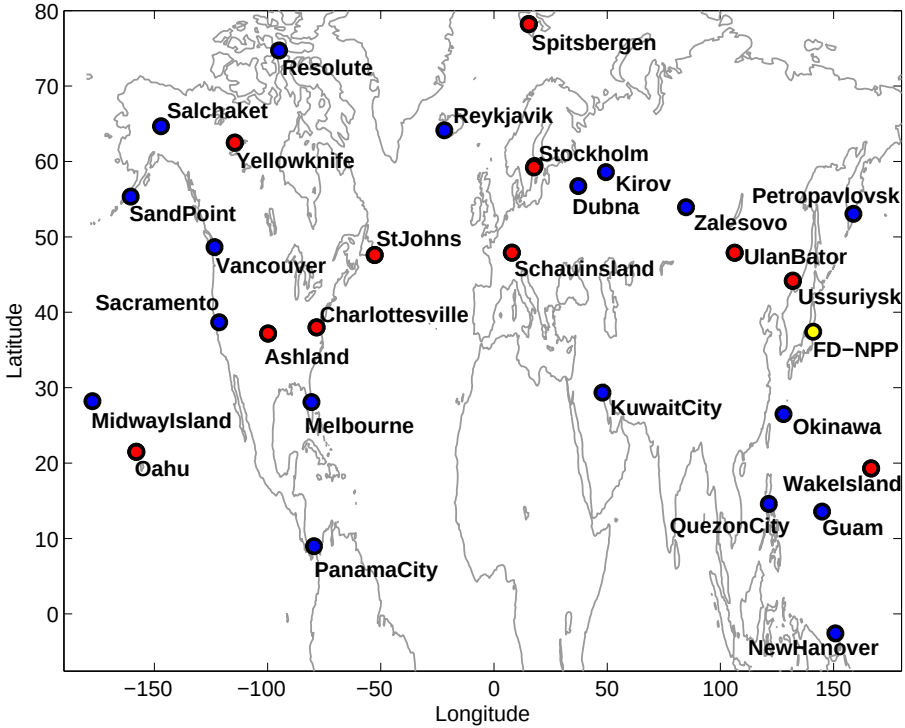

Fig. 1. Measurement station network. CTBTO stations measuring particulates only (^{137}Cs and ^{133}I; blue markers) and those simultaneously measuring particulates and noble gas (^{133}Xe; red markers). The position of FD-NPP is shown by a yellow marker.

ACPD
12, 12331–12356, 2012

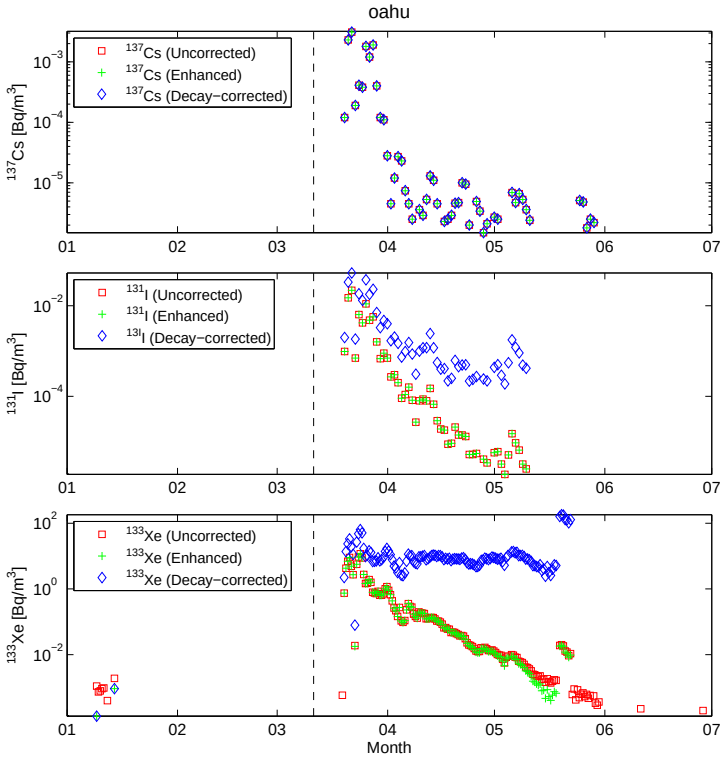

Fig. 2. Time-series of observed ^{137}Cs, ^{131}I and ^{133}Xe at the station Oahu. Shown are the observed activity concentrations (red squares), the observed concentrations with background subtracted (green plusses), and the decay-corrected enhancements over the background (blue diamonds). The time of the earthquake (11 March 2011) is indicated with the vertical dashed line.

ACPD
12, 12331–12356, 2012

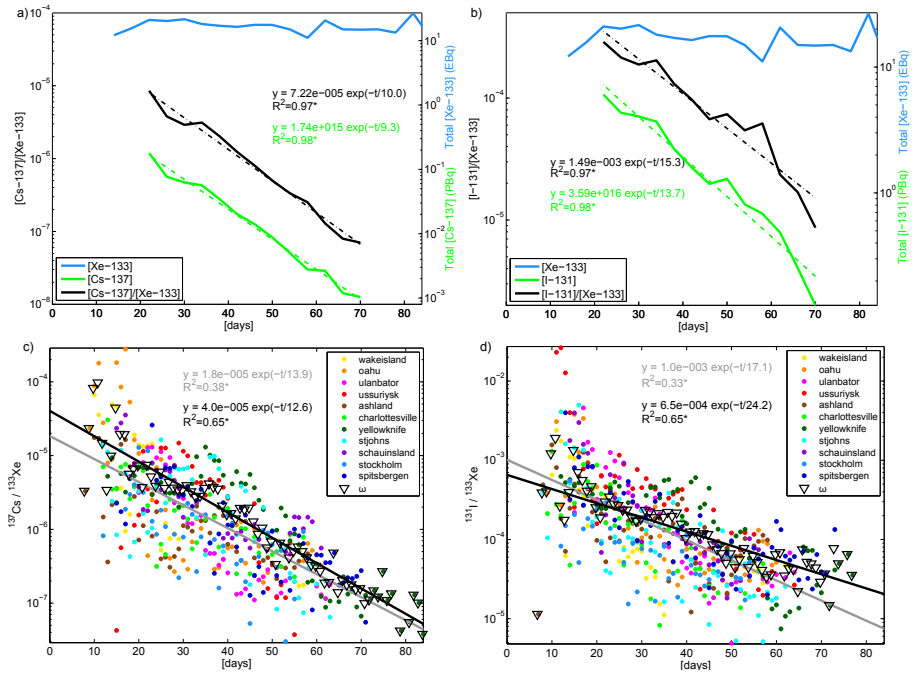

Fig. 3. ^{137}Cs and ^{131}I decrease with time. **(a)** Total atmospheric burden of [^{137}Cs], [^{133}Xe] and the [^{137}Cs]/[^{133}Xe] ratio over time, from a multi-box model. The fit of exponential decay models to the data (dotted lines and text insets) with removal time τ_b of 10.0 days, are also shown. The fraction of variance in the data explained by the exponential model is given by R^2. **(b)** Same as **(a)**, but for ^{131}I. **(c)** Measured ^{137}Cs/^{133}Xe ratios for 11 CTBTO-stations (colored marks) and the fit of an exponential decay model to the data (grey line) with removal time τ_a of 13.9 days. ω (black triangles) represent the ratio of the activity concentrations for each day averaged over all stations and the black line is the exponential fit through these ratios with removal time τ_ω of 12.6 days. **(d)** Same as **(c)**, but for the ^{131}I/^{133}Xe ratios. The time-axes on all four figures are in days after the time of the earth quake (11 March at 05:46 UTC).

ACPD
12, 12331–12356, 2012

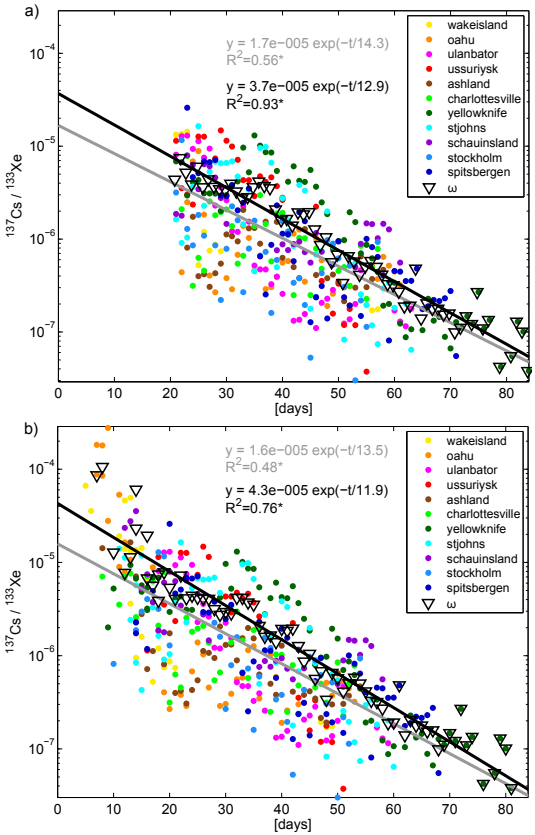

Fig. 4. Sensitivity tests for removal times τ of ^{137}Cs. Measured ^{137}Cs/^{133}Xe ratios and fit of exponential models to the data (solid lines). **(a)** Same as Fig. 3c, but using measurement data only after 1 April 2011, the exponential model yields estimates of τ_a = 14.3 and τ_ω = 12.9 days. **(b)** Using plume age calculations based on FLEXPART model output yields τ_a = 13.5 and τ_ω = 11.9 days. Notice that the age here is not referenced to the time of the earthquake but to the actual emission time most relevant for each sample.

ACPD
12, 12331–12356, 2012