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Modelling the global atmospheric transport and deposition of radionuclides from the Fukushima Dai-ichi nuclear accident

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Abstract

We modeled the global atmospheric dispersion and deposition of radionuclides released from the Fukushima Dai-ichi nuclear power plant accident. The EMAC atmospheric chemistry – general circulation model was used, with circulation dynamics nudged towards ERA-Interim reanalysis data. We applied a resolution of approximately 0.5 degrees in latitude and longitude (T255). The model accounts for emissions and transport of the radioactive isotopes ^{131}I and ^{137}Cs , and removal processes through precipitation, particle sedimentation and dry deposition. In addition, we simulated the release of ^{133}Xe , a noble gas that can be regarded as a passive transport tracer of contaminated air. The source terms are based on Stohl et al. (2012) and Chino et al. (2011); especially the emission estimates of ^{131}I are associated with a high degree of uncertainty. The calculated concentrations have been compared to station observations by the Comprehensive Nuclear-Test-Ban Treaty Organisation (CTBTO). We calculated that about 80 % of the radioactivity from Fukushima which was released to the atmosphere deposited into the Pacific Ocean. In Japan a land area of 34 000 km² around the reactors, inhabited by nearly 10 million people, was contaminated by more than 40 kBq m⁻². We also estimated the inhalation and 50-yr dose by ^{137}Cs and ^{131}I to which the people in Japan have been exposed.

1 Introduction

On 11 March 2011 an earthquake occurred off the Pacific coast of Tōhoku, which triggered a powerful tsunami. The tsunami damaged the Fukushima Dai-ichi nuclear power plant complex, followed by a series of major accidents, giving rise to three INES (International Nuclear Event Scale) level 7 events. This caused the release of large amounts of radionuclides to the atmosphere (Butler, 2011; Stohl et al., 2012; Chino et al., 2011). In particular the isotopes of iodine and caesium adversely affect human health through the large-scale contamination of air, water, soils and agricultural

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products (Anspaugh et al., 1988). Although both radionuclides are released as gases, caesium has a low volatility and partitions into ambient aerosol particles, whereas iodine largely remains in the gas phase. Removal of these compounds from the atmosphere is governed by dry and wet deposition processes. Several initial studies of the atmospheric behaviour and budgets of radioactive species, notably ^{131}I and ^{137}Cs , were conducted using chemistry transport models. For example, the Austrian Weather Service (ZAMG) performed a daily forecast of the dispersion of radioactivity in the atmosphere. Morino et al. (2011) simulated the transport and deposition of the radionuclides over Japan using an off-line regional model.

The objective of the present study is to model the global atmospheric dispersion of radionuclides from the Fukushima accident and compute the deposition patterns using an atmospheric chemistry–general circulation model, initialized by estimated source terms. The emission strengths have been determined through inverse modelling by Stohl et al. (2012). The temporal emission profiles of the radioactive isotopes ^{133}Xe and ^{137}Cs were estimated at three reference release heights using the Lagrangian transport model FLEXPART (Stohl et al., 2005), driven by operational meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF) and the National Center for Environmental Prediction (NCEP) Global Forecast System (GFS) analyses, and constrained by a large number of available concentration and deposition data from Japan, North America and Europe.

Furthermore, we incorporated the preliminary estimates of release amounts of ^{131}I by Chino et al. (2011). A reverse estimation of the source term was conducted by coupling Japanese Environmental monitoring data with regional atmospheric dispersion simulations.

We briefly describe the model in Sect. 2, including the emissions of radionuclides and the representation of removal mechanisms (Sect. 2.1). The analysis of our results regarding the global deposition (Sect. 3.2) and a comparison of surface concentrations to station measurements (Sect. 3.1) are discussed in Sect. 3. Estimates of doses from

the inhalation and ground deposition of radionuclides are given in Sect. 3.3. A summary and conclusions are presented in Sect. 4.

2 Model simulation

5 The global ECHAM5/MESSy Atmospheric Chemistry (EMAC) general circulation model includes sub-models describing tropospheric and middle atmospheric processes and their interactions with oceans, land and vegetation, and trace species emissions of natural and anthropogenic origin (Jöckel et al., 2005). It uses the first version of the Modular Earth Submodel System (MESSy1) to link multi-institutional computer codes. The core atmospheric model is the 5th generation European Centre Hamburg
10 general circulation model (ECHAM5; Roeckner et al., 2006).

For the present study we applied EMAC version 1.9 (based on ECHAM5 version 5.3) at the T255L31 resolution, i.e. with a spherical spectral truncation of T255 (corresponding to a quadratic Gaussian grid of approximately 0.5 by 0.5 degrees in latitude and longitude) with 31 vertical hybrid pressure levels up to 10 hPa, a time step of two
15 minutes and three-hourly output. To test the effects of model resolution on the simulation we repeated the study on the reduced T106L31 resolution (corresponding to a quadratic Gaussian grid of about 1.1 by 1.1 degrees). Our simulation spans the period of 1 March–31 May 2011. The large-scale component of the model circulation dynamics was nudged by applying a Newtonian relaxation (Jeuken et al., 1996) towards the
20 European Centre for Medium-range Weather Forecasts (ECMWF) ERA-Interim reanalysis data (Simmons et al., 2007), produced at T255 resolution, to realistically represent the tropospheric meteorology of the simulated period.

The applied model setup comprises the submodels RAD4ALL for radiation and atmospheric heating processes, CLOUD for cloud formation and microphysical processes
25 including precipitation, and CONVECT for the vertical transports of trace species associated with convection. The DRYDEP (Kerkweg et al., 2006) and SCAV (Tost et al., 2006, 2007) submodels were used to simulate aerosol dry and wet deposition

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processes, respectively. The SEDI submodel was used to simulate particle sedimentation, of which the results will be presented below as part of the simulated dry deposition.

We focus on the radionuclides that were emitted as gases and partly attached to ambient aerosol particles: the semi-volatile isotopes of iodine ^{131}I (with a half-life of 8 days) and caesium ^{137}Cs (with a half-life of ~ 30 yr). We also modeled the emission of the isotope ^{133}Xe (with a half-life of 5.25 days). Since xenon is an unreactive noble gas and is not removed by deposition processes, it serves as a passive tracer of atmospheric transport. ^{137}Cs is modeled as a water-soluble aerosol with a standard lognormal distribution with mean radius $0.25\text{ }\mu\text{m}$ and a Henry's law coefficient equal to $1.0\text{ mol l}^{-1}\text{ atm}^{-1}$ and a density of 1000.0 kg m^{-3} . Due to the long decay lifetime of ^{137}Cs compared to the simulation period and the short timescales of the atmospheric removal processes considered, its radioactive decay is not taken into account in the simulation.

2.1 Emissions

We used the temporal emission patterns for the radioactive isotopes ^{133}Xe and ^{137}Cs for March and April 2011, estimated based on an inverse modelling method, applying the transport model FLEXPART, by Stohl et al. (2012). Tracers are emitted within three atmospheric layers over the Fukushima Dai-ichi power plant (which extend up to 50, 300 and 1000 m). Emission inventories are provided by Stohl et al. (2012) and used in our study between the 10th of March and the 20th of April 2011. The total amount of ^{137}Cs emitted is 36.7 PBq, and of ^{133}Xe 15.3 EBq (Stohl et al., 2012).

We also incorporated in our model preliminary emission estimates of the released amounts of ^{131}I (in total 150 PBq) and ^{137}Cs (in total 13 PBq) by Chino et al. (2011), based on the reverse estimation of the source term by coupling Japanese environmental monitoring data with regional atmospheric dispersion simulations. The System for Prediction of Environmental Emergency Dose Information (SPEEDI; Chino et al., 1993) network system, operated by the Japanese Ministry of Education, Culture, Sports,

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Science and Technology (MEXT, 2011), was used to calculate dose rates and atmospheric concentrations.

5 The emission inventory compiled by Chino et al. (2011) is limited by the use of Japanese station data only and a regional simulation domain. Also, the calculations
10 assumed constant radioactivity ratios for the different radionuclides based on iodine and caesium concentrations in rain, snow and vegetation in the area. These factors, and a number of other assumptions made to produce the emission inventory lead to a large uncertainty to the estimated release rates. Chino et al. (2011) mention that their emission estimate is associated with an error of at least a factor of five. This
15 large uncertainty should be taken into account for inter-comparisons between emission datasets and with model simulations, and is discussed in Sect. 3.2.

A comparison of the estimate of released ^{137}Cs by Stohl et al. (2012) and Chino et al. (2011) can be seen in the total model calculated atmospheric content (Fig. 1). It shows that the two emission estimates, both in magnitude and time profile, differ greatly. We
20 find that the spatial resolution of our model has little impact on the global atmospheric content of radionuclides.

3 Results

The comparison of our model results with station measurements is discussed in Sect. 3.1. The modeled global deposition of radionuclides is presented in Sect. 3.2.
20 The doses to the public from deposition and inhalation based on our results are calculated in Sect. 3.3.

3.1 Comparison with station measurements

The Comprehensive Nuclear-Test-Ban Treaty (CTBT), adopted by the United Nations General Assembly, foresees a ban of all nuclear explosions. For compliance,
25 a global International Monitoring System (IMS) of measurement technologies has been

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established by the CTBT Organization (CTBTO), with data delivered to the International Data Centre for of the CTBTO Preparatory commission in Vienna, Austria. The IMS comprises radionuclide measurement stations with continuous coverage, distributed around the globe. The subset of stations with available measurements used in our study can be seen in Figs. 2 and 3.

The stations operate high volume aerosol samplers using collection filters with a minimum detection capability (MDC) range typically between $1\text{--}10\text{ }\mu\text{Bq m}^{-3}$ for Cs. Since iodine largely travels in the gas phase, the filter measurements represent only a small fraction of airborne ^{131}I , thus associated with a relatively large measurement uncertainty. Only 20–50 % of ^{131}I is collected in the samples, since the IMS stations are using paper filters through which part of the iodine passes unattenuated (as reported by the CTBTO Virtual Data Exploitation Centre). Noble gas concentration measurement stations are also been set up worldwide, with ^{133}Xe being the most important and prevalent isotope. It is measured with typical MDC of about 0.2 mBq m^{-3} . The CTBTO station measurements of radionuclide concentrations can be directly compared to the concentrations simulated by our model.

There are some additional aspects that affect the CTBTO station measurements, which should be taken into account when comparing to the model simulation. The highest concentrations of Xe after the accident were outside the dynamic range of the noble gas system, so detections that are higher than 100 Bq m^{-3} are not expected to be accurate. It is also known that the Takasaki (JPX38) noble gas station in Japan was affected significantly by dead time in the first three weeks after the accident. The Japanese particulate station JPP38, also at Takasaki, was affected by radioactive leaks into the measurement room and power outages between 14 and 16 March 2011. Contamination at JPP38 lead to an overestimation of ^{137}Cs concentrations by about 1 mBq m^{-3} until August 2011. Similar contamination occurred at USP71 at Sand Point, Alaska, USA, leading to an overestimation in the range of $3\text{ }\mu\text{Bq m}^{-3}$ for ^{137}Cs . Therefore, for these locations we expect our model to underestimate the reported observations.

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The locations of the CTBTO measurement stations for radionuclide gases and particulates used for comparison, including station codes, can be seen in Fig. 3. In total, eight stations are available for noble gas observations and 37 for particulates. The availability of 3 months of measurements allows for the comparison over a wide range of meteorological conditions and radionuclide concentrations at each station location.

We focus on the concentrations of radionuclides in the lowest layer of our model and in the grid boxes where the stations are located, and compare with the measured concentrations at each station at the nearest model output time step to the station collection start time. The comparison for all ^{133}Xe stations is presented in Fig. 2 and selected stations for ^{137}Cs and ^{131}I in Figs. 3 and 4, respectively. The entire set of measurements and concomitant model results is presented in the Supplement. Our model does not take into account the resuspension of particles after they have been deposited, though we expect the effect to be small.

Overall, there is good coincidence of the time of first arrival of the radionuclide plume at the station and the time of first detection of radioactivity at a station, especially for stations in the Asia-Pacific and North American regions, taking into account coincidence timing uncertainties. As all the stations collect samples over a period of typically 12 h or more for each measurement performed, there is an associated uncertainty of the same order of magnitude over the time of first arrival of radionuclides from Fukushima at each measurement location. Furthermore, the measurement record is not continuous for all stations, resulting in cases where there is no observational coverage of the model prediction of the arrival of radionuclide plumes.

Good agreement is obtained for the inert noble gas ^{133}Xe (although only a limited number of stations is available). The agreement is not as good for ^{137}Cs and ^{131}I . The greater extent to which deposition processes are involved (dry deposition for ^{131}I , dry and wet deposition and sedimentation for ^{137}Cs), creates larger uncertainties in the source estimate from inversion and the modelling of atmospheric transport. It is notable that the results of the higher resolution T255 calculations are not systematically more consistent with the measurements than the lower resolution T106 results. The

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lower panels of Figs. 2, 3, and 4 present the root-mean-square-deviations (RMSDs) between the measurements and model results for both resolutions. We also computed the normalised RMSDs, and find that they are typically below 50 %, irrespective of the distance between the station and the emission source.

For the eight stations available with varying number of measurements, there is good agreement between the model results and the measurements for the noble gas ^{133}Xe , both for the higher and lower resolution (Fig. 2). Nevertheless, the modeled concentrations at JPX38 (Takasaki, Gunma, Japan) peak in March at values well outside the dynamic range of the measurement instruments. All measurements at the other stations are below 100 mBq m^{-3} , the reported upper range of the observation accuracy. The model not only represents the measurements well in the vicinity of the source, but also in North, Middle and South America (stations USX75, PAX50, FRX31), and even as far downwind as Germany (DEX33). Again, there are no indications that the higher resolution version of the model performs systematically better. Even though the ERA-Interim data, used to nudge the model to the actual meteorology, are produced at T255 resolution, the regridding of the data to the coarser T106 resolution seems to have little effect on the quality of the dynamics simulations.

For the comparison of the model simulation results to observations of the surface concentrations of ^{137}Cs (Fig. 3), reduced though nevertheless realistic agreement can be claimed, taking into account the inherent uncertainties based on the multitude and the complexity of the simulated removal processes (sedimentation, dry and wet deposition). In most cases there is close coincidence between the modeled and observed time of arrival of the first radioactive plume at the station and most model results fall within the factor of five range of the measurements (Fig. 3). However, our model shows the arrival of high concentrations of radioactive aerosols in the Asia-Pacific region (see for example USP80 at Upi, Guam, in the Supplement, and PHP52 in Tanay, the Philippines, in Fig. 3), which are not reflected in the station observations. There is clearly reduced agreement compared to the results for the noble gas ^{133}Xe , which we attribute

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to the additional uncertainties related to the simulation of removal processes, in particular by precipitation.

5 The model tends to generally underestimate the measurements of the atmospheric concentrations of ^{131}I based on the emission inventory by Chino et al. (2011). In the case of ^{131}I both the observations and emission estimates are associated with large uncertainty. The uncertainty in the estimated release rates is mainly caused by the accuracy of the dispersion calculations and the limited monitoring coverage in space and time. Chino et al. (2011) consider their results to have an error of at least a factor of five. Since ^{131}I is not removed by wet deposition and relatively slowly by dry deposition, 10 and our model has been shown to reproduce transport quite accurately, it appears that the discrepancies between measurements and model simulations to a large degree reflect inaccuracies in the applied emission strength and profile. Considering that the model typically underestimates ^{131}I concentrations, we expect that the actual emissions of ^{131}I were higher than those reported by Chino et al. (2011). Furthermore, since only 15 20–50 % of the ^{131}I is captured on the filters, the model calculations would be consistent with the observations by applying emissions of ^{131}I of at least a factor two to five higher than the 150 PBq of Chino et al. (2011).

3.2 Deposition of ^{137}Cs and ^{131}I

20 In this section we assess the atmospheric budget and deposition patterns of radioactivity by calculating the contributions of different removal processes, i.e. particle sedimentation, dry and wet deposition; the latter through large-scale and convective precipitation. Since the contribution of sedimentation is only small we present it as part of the dry deposition of ^{137}Cs (in the model ^{131}I is in the gas phase only). The global distribution of deposited ^{137}Cs radionuclides is shown in Fig. 5 for dry and wet deposition processes 25 separately (top and middle panel, respectively). Following the definition by the International Atomic Energy Agency (IAEA) (2005, 2009b), any area with activity $\geq 40 \text{ kBq m}^{-2}$ is considered to be contaminated (the most intense red scale). Formally, contamination means the presence of a radioactive substance on a surface in quantities in excess of

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$\geq 40 \text{ kBq m}^{-2}$ for beta and gamma emitters. Both, ^{137}Cs and ^{131}I are beta and gamma emitters. Since we integrate the deposition over the period after the accident until the end of May 2011, our results represent the accumulated contamination of these two radionuclides.

The prevailing winds during the emission period after 11 March transported most of the radionuclides in easterly direction, away from Japan and the continent. Precipitation events caused the deposition to occur predominantly over the ocean. Figure 5 illustrates that the dry deposition patterns are smoother than of wet deposition, i.e. less dependent on individual meteorological events. It also shows that dry deposition of ^{137}Cs is mostly confined to the Pacific Ocean region because this process only acts upon particles in the boundary layer. Only the radioactive particles that escape towards the free troposphere can be transported over much larger distances. Since exchange between the boundary layer and free troposphere is mostly dependent on convection events, the particulate contaminants are to a large extent removed by the associated precipitation. This is much less the case for ^{131}I , which has a low solubility, so that convection effectively redistributes this gas into the free troposphere where the wind speed is typically higher and transport distances larger.

Therefore, even though ^{131}I has a relatively short half-life, meteorological conditions associated with convection and vertical mixing over the Pacific Ocean promote its long-distance transport so that it contributes to radioactivity deposition worldwide (Fig. 5, bottom panel). Our model results suggest that the plumes that traversed the Pacific Ocean caused significant deposition of radioactivity over Continental North America, in particular Western USA, Western Canada and Eastern USA ($> 100 \text{ Bq m}^{-2}$). Our model results also show substantial deposition of radionuclides in regions southwest of Japan, e.g. around the Philippines (Fig. 6, upper panel).

Detailed budget analysis, based on the T255 resolution model results, shows that 29.3 PBq of the total of 36.7 PBq ^{137}Cs released by Fukushima was deposited to the ocean (equivalent to $\sim 80\%$) and the rest over land, mostly in Japan. Only a small fraction (less than 1%) was deposited within the Arctic circle (at latitudes higher than

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67° N). We estimate that of the total emitted amount of radioactivity, approximately 40 %, 60 %, and 75 % were deposited in regions at distances of 5°, 10° and 20° in latitude and longitude from the location of the source.

5 The regional deposition distribution of the sum of ^{137}Cs and ^{131}I at different scales is shown in Fig. 6. We estimate that the land area affected by the deposition of radioactivity (by both compounds) in excess of 40 kBq m^{-2} is approximately $34\,000 \text{ km}^2$. Using the population data from (CIESIN/CIAT/SEDAC, 2005) (referring to the year 2010), this part of Japan is inhabited by ~ 9.4 million people. The contaminated area includes, e.g. the city of Sendai (about 1 million inhabitants). The surface area that received a total
10 deposition greater than 10 kBq m^{-2} encompasses parts of the Tokyo metropolitan area, including Yokohama and Chiba, and approximately covers $60\,000 \text{ km}^2$, being inhabited by ~ 46 million people. We emphasize that this is based on the emission of $150 \text{ PBq } ^{131}\text{I}$ estimated by Chino et al. (2011), which might actually be a factor of five too low.

To test the potential of a larger source than $150 \text{ PBq } ^{131}\text{I}$, we performed a sensitivity
15 test by applying 5 times higher emissions, based on the emissions estimate uncertainty by Chino et al. (2011). Figure 7 shows the results, indicating significantly increased deposition of radioactivity worldwide (upper panel). The regional and national deposition patterns are illustrated by the middle and lower panel, respectively. If this were realistic, an area of $56\,000 \text{ km}^2$, inhabited by ~ 43 million people, would be contaminated
20 by more than 40 kBq m^{-2} , including the Tokyo metropolitan area. Although this large ^{131}I source is speculative, it seems more realistic than the low estimate of 150 PBq by Chino et al. (2011). Further study will be needed to resolve this important issue.

3.3 Doses from inhalation and ground deposition

25 We estimated the cumulative doses due to inhalation over the simulation period (11 March–31 May 2011) as well as the effective doses over 50 yr from ground contamination by applying conversion factors for ^{137}Cs and ^{131}I recommended by the International Atomic Energy Agency (International Atomic Energy Agency, IAEA, Appendix I). The

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contribution by noble gases, including ^{133}Xe , can be neglected as the dose ratios relative to ^{131}I are effectively zero.

The inhalation doses are converted from model calculated concentrations (Bq s m^{-3}) into Sv (Sievert), applying the factors 1.29×10^{-11} and 2.4×10^{-12} for ^{137}Cs and ^{131}I , respectively. The results are shown in Fig. 8. Note that these calculations do not comprise dosages to individuals from the ingestion of radionuclides, based on the assumption that food intervention levels prevent such exposure. The cumulative effective dose from inhalation in areas close to the Fukushima nuclear power plant is found to exceed 10 mSv for the nominal emissions (36.7 PBq ^{137}Cs and 150 PBq ^{131}I) and 20 mSv for the sensitivity test with five times higher ^{131}I emissions, i.e. the upper bound of the emissions uncertainty indicated by Chino et al. (2011). These estimates apply to adult members of the general public and not to workers who received higher doses in contaminated facilities.

Further, we calculated the effective dose to the public from exposure to ground contamination due to the deposition of ^{137}Cs and ^{131}I . The 50-yr ground deposition doses are derived from the conversion of deposited radioactivity in Bq m^{-2} into an effective dose in Sv, applying the factors 1.3×10^{-7} and 2.7×10^{-10} for ^{137}Cs and ^{131}I , respectively (International Atomic Energy Agency, IAEA). The cumulative effective dose from radioactive compounds remaining on the ground for a period of 50 yr is shown in the right panel of Fig. 8. The 50-yr effective dose is dominated by the effects of ^{137}Cs and is less sensitive to the large uncertainty of the deposited amounts of ^{131}I .

4 Conclusions

The release of radioactivity from the Fukushima Dai-ichi nuclear power plant accident and the atmospheric transport and deposition patterns between March and May 2011 were simulated using the EMAC atmospheric chemistry – general circulation model at different resolutions (T255 and T106). The atmospheric dynamics were nudged towards ERA-Interim reanalysis data of the ECMWF, available at T255 resolution.

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We initialized our model by applying emissions of the inert noble gas ^{133}Xe (15.3 EBq) and the semi-volatile gases ^{131}I (150 PBq) and ^{137}Cs (36.7 PBq). The latter attaches to ambient aerosol particles, whereas the iodine largely remains in the gas phase. The source terms are based on estimates from the literature, produced by inverse modelling and reverse estimation techniques. There are significant differences in the total amount and temporal release of ^{137}Cs between the two presently available estimates. Removal processes through precipitation, particle sedimentation, dry deposition and radioactive decay were taken into account.

Modeled surface concentrations were compared to station observations by the Comprehensive Nuclear-Test-Ban Treaty Organisation (CTBTO) global monitoring network for a selected number of stations. We obtained good agreement with measurements of ^{133}Xe , at both T255 and T106 resolution. The comparison for ^{137}Cs is less favorable, though not systematically biased based on the ^{137}Cs emission estimate of 36.7 PBq by Stohl et al. (2012). This is equivalent to somewhat less than half the ^{137}Cs source by Chernobyl (85 PBq, International Atomic Energy Agency, IAEA, 2006).

The model calculated deposition patterns show that most of the radioactivity which was released to the atmosphere ended in the Pacific Ocean (about 80 %). Although the prevailing wind direction during the accident was in easterly direction, some of the atmospheric radioactivity was transported toward the west and southwest, and deposited in Japan and to a lesser extent reached the Philippines. Based on the above mentioned emission estimates, we calculated that an area of Japan of 34 000 km² was contaminated by more than 40 kBq m⁻² of ^{137}Cs and ^{131}I , to which ~ 9.4 million people were exposed. Our model results indicate that a surface area of 60 000 km² received a total deposition greater than 10 kBq m⁻², a region inhabited by ~ 46 million people.

The source term of 150 PBq ^{131}I applied in our model, based on Chino et al. (2011), is associated with a systematic underestimate of simulated concentrations compared to the CTBTO measurements. Therefore, we performed an ^{131}I emissions sensitivity test, with a factor of five stronger source, i.e. the upper bound of the uncertainty range indicated by Chino et al. (2011). If this would be realistic, our model indicates

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that a relatively large and densely populated part of Japan of 56 000 km², including parts of the Tokyo metropolitan area has been contaminated by more than 40 kBq m⁻². Additional work will be needed to estimate the ¹³¹I source more accurately.

Finally, we estimated the cumulative doses to the public due to the inhalation of radionuclides over the simulated time period (11 March–31 May 2011) and the effective doses over a 50-yr period from ground contamination. The maximum inhalation dose around the Fukushima nuclear accident site is calculated to be > 10 mSv for the nominal emissions case and > 20 mSv for the sensitivity test to the emissions uncertainty quoted by Chino et al. (2011) (five times higher ¹³¹I emissions). These estimates only include ¹³⁷Cs and ¹³¹I, and neglect other isotopes of caesium and iodide and other radionuclides such as ¹³²Te. The maximum 50-yr dose due to ground contamination in the area around the Fukushima nuclear power plant is estimated to be > 125 mSv. This result is less sensitive to the high uncertainty of the ¹³¹I emissions since it is dominated by the deposition of ¹³⁷Cs.

Supplementary material related to this article is available online at:
**[http://www.atmos-chem-phys-discuss.net/12/24531/2012/
acpd-12-24531-2012-supplement.pdf](http://www.atmos-chem-phys-discuss.net/12/24531/2012/acpd-12-24531-2012-supplement.pdf)**

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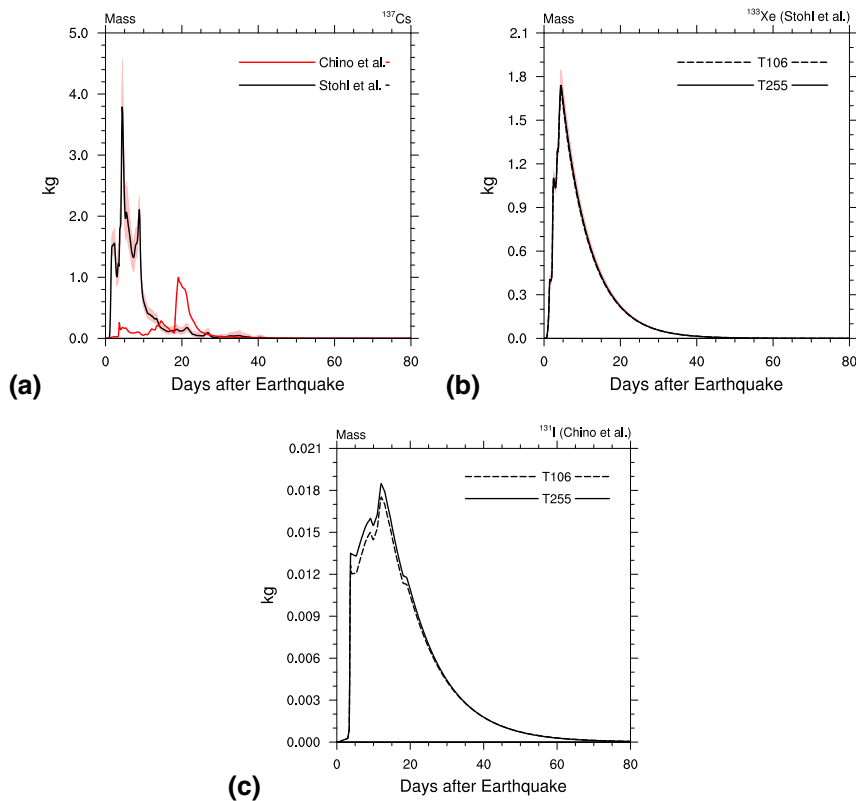


Fig. 1. Total atmospheric content (in kg) of ^{137}Cs (a) and ^{133}Xe (b) and ^{131}I (c) that originated from Fukushima Dai-ichi based on the emissions inventories by Stohl et al. (2012) and Chino et al. (2011). The red bands indicate the reported range of the emission source estimates, and the dashed lines correspond to the results of the T106 horizontal resolution simulation.

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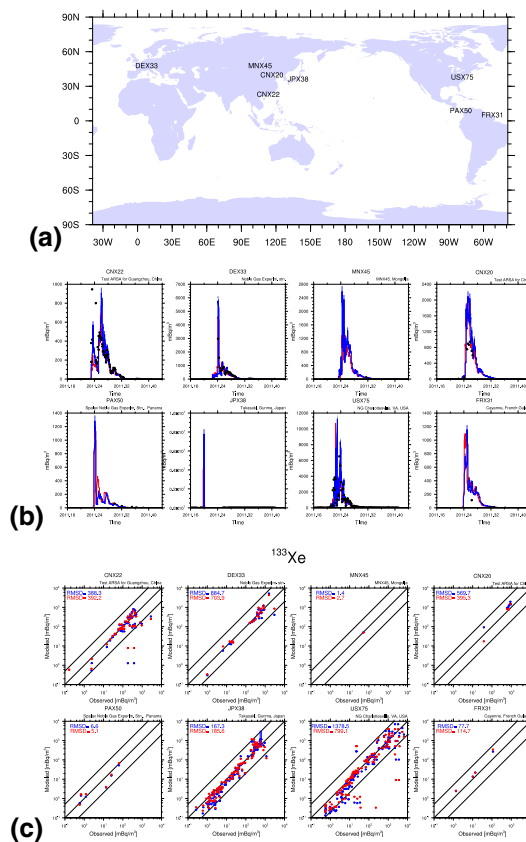


Fig. 2. (a) CTBTO measurement network station location and code for the noble gas ^{133}Xe for the subset of stations with measurements available used in our study. (b) Observed (black points) and modeled (solid lines) surface concentrations of ^{133}Xe at the eight stations. T255 horizontal resolution is shown in blue and T106 in red. The time axis represents the fraction (percentage) of the year 2011. (c) Modeled versus observed surface concentrations of ^{133}Xe at the eight stations. The diagonal lines represent the 1 : 1 ratio and the factor of 5 over- and underestimates.

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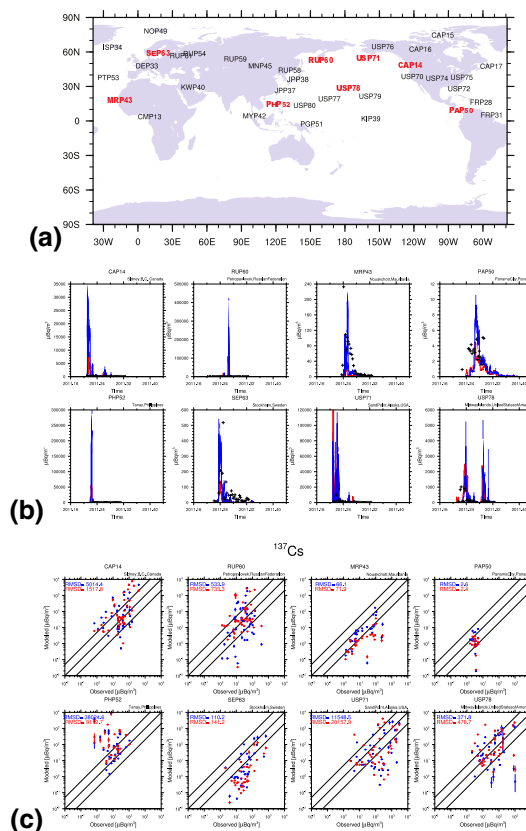


Fig. 3. (a) CTBTO measurement network station location and code for ^{137}Cs for the subset of stations with measurements available used in our study. Stations shown in our study are highlighted in red. (b) Observed (black crosses) and modeled (solid lines) surface concentrations of ^{137}Cs at the eight selected stations. T255 horizontal resolution is shown in blue and T106 in red. The time axis represents the fraction (percentage) of the year 2011. (c) Modeled versus observed surface concentrations of ^{137}Cs at the eight stations. The diagonal lines represent the 1 : 1 ratio and the factor of 5 over- and underestimates.

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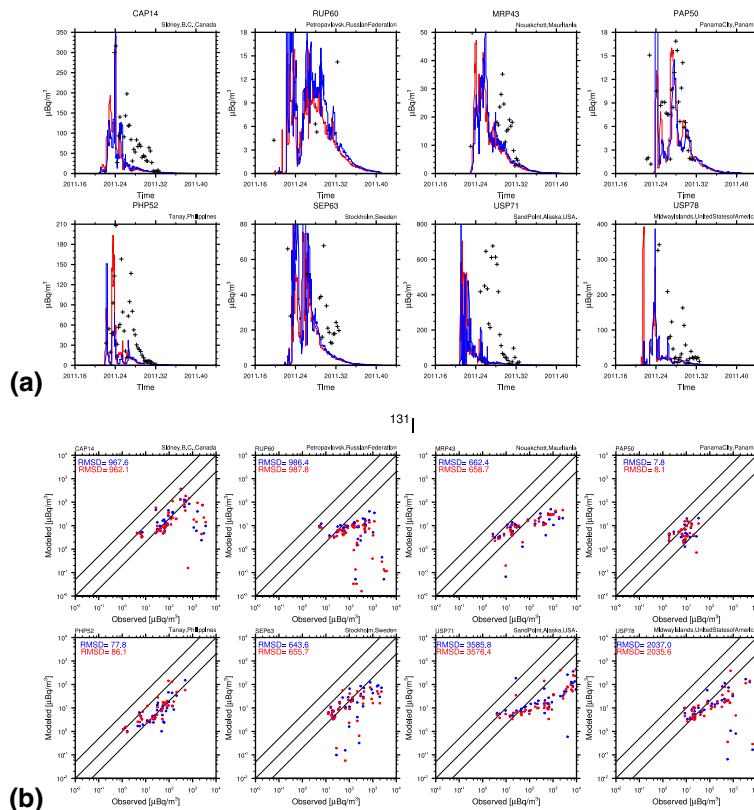


Fig. 4. (a) Observed (black points) and modeled (solid lines) surface concentrations of ^{131}I at eight selected stations. The station locations are the same as in Fig. 3. Due to the limited collection efficiency of the filter sampling of ^{131}I , the observations represent about 20–50 % of the ambient concentrations. T255 horizontal resolution is shown in blue and T106 in red. The time axis represents the fraction (percentage) of the year 2011. (b) Modeled versus observed surface concentrations of ^{131}I at the eight stations. The diagonal lines represent the 1 : 1 ratio and the factor of 5 over- and underestimates.

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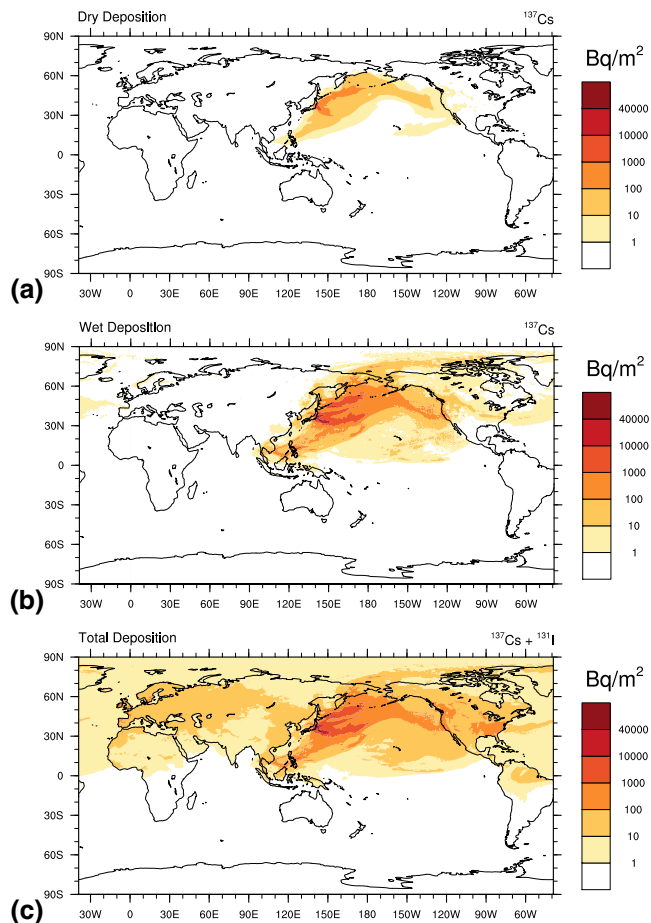


Fig. 5. Global distribution of dry (a) and wet (b) deposition (in Bq) of ^{137}Cs and total deposition (c) of ^{137}Cs and ^{131}I . Contours are on a logarithmic scale.

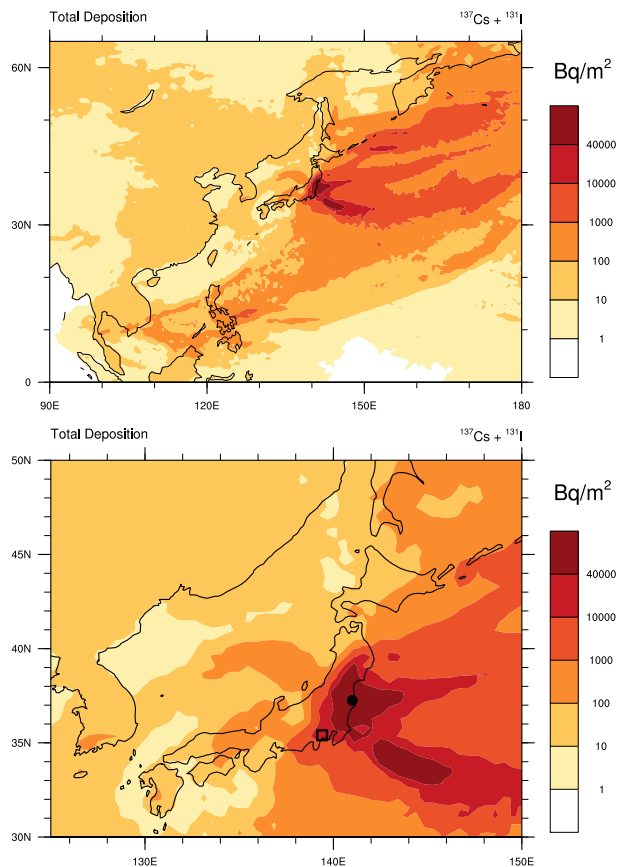


Fig. 6. Regional total deposition of ^{137}Cs and ^{131}I in Bq at different scales. The black point marks the location of the Fukushima nuclear power plant and the rectangle the Tokyo metropolitan area. Contours are on a logarithmic scale. The 40 kBq m^{-2} contour lines signify areas that received total deposition greater than the contamination activity level defined by International Atomic Energy Agency (IAEA) (2005, 2009b).

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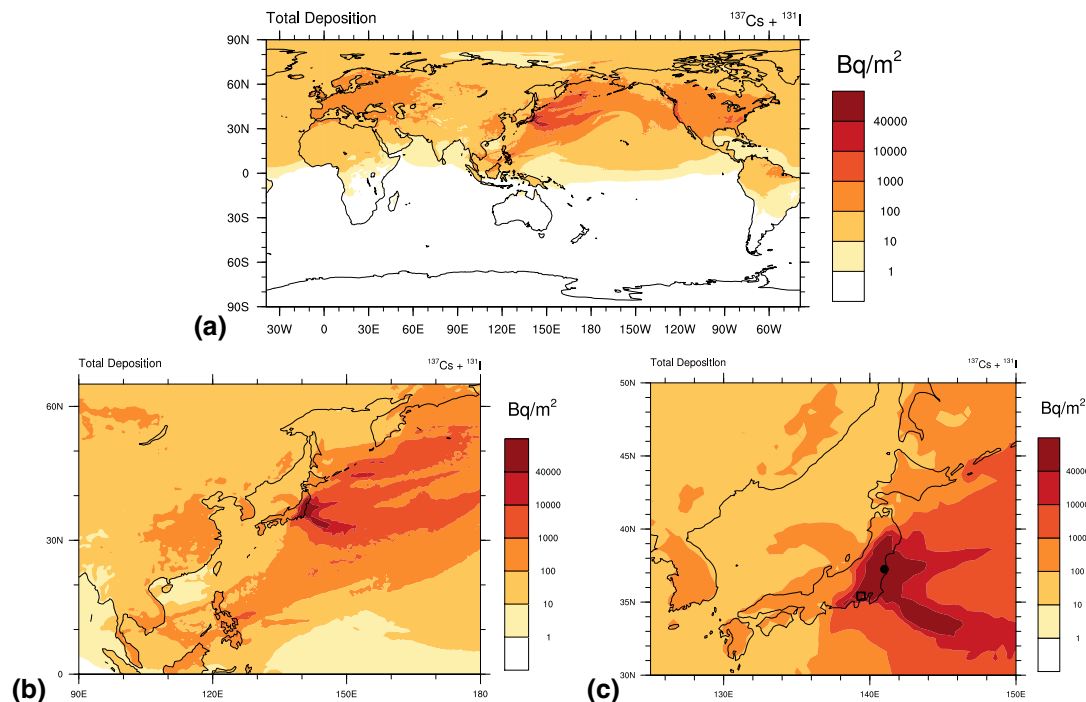


Fig. 7. Global (a), and regional (b Eastern Pacific; c Japan) combined total deposition of ^{137}Cs and ^{131}I in Bq , based on a sensitivity simulation with 5 times enhanced ^{131}I emissions (750 TBq). The black point marks the location of the Fukushima nuclear power plant and the rectangle the Tokyo metropolitan area. Contours are on a logarithmic scale. The 40 kBq m^{-2} contour lines signify areas that received total deposition greater than the contamination activity level defined by the International Atomic Energy Agency (IAEA) (2005, 2009b).

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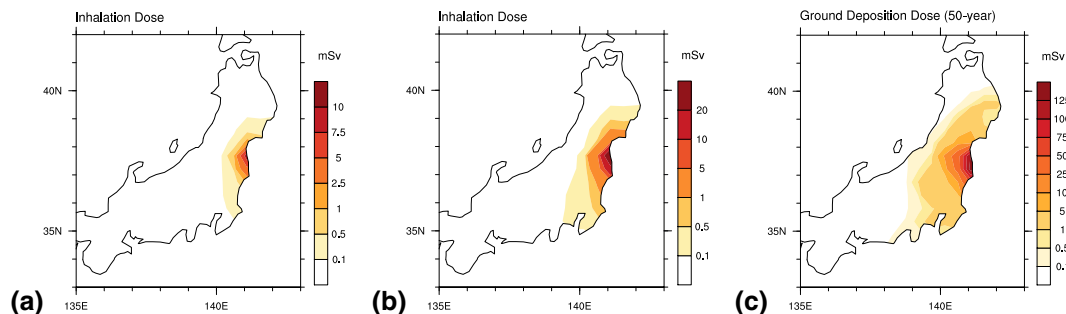


Fig. 8. Effective dose from the inhalation of airborne radionuclides for the nominal emission dataset (a); inhalation dose sensitivity test for the ^{131}I emission scaled by factor 5 (b); and 50-yr dose from the ground deposition of ^{131}I and ^{137}Cs (c) based on dose factors by the International Atomic Energy Agency (IAEA).

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