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Global deposition and transport efficiencies of radioactive species with respect to modelling credibility after Fukushima (Japan, 2011)

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ABSTRACT

In this study we conduct a detailed comparison of the modelling response of the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident with global and local observations. We use five different model versions characterized by different horizontal and vertical resolutions of the same General Circulation Model (GCM). Transport efficiencies of ^{137}Cs across the world are presented as an indication of the expected radioactive impact. Activity concentrations were well represented showing lower Normalized Mean Biases (NMBs) when the better resolved versions of the GCM were used. About 95% of the results using the zoom configuration over Europe (zEur) remained within a factor of 10 from the observations. Close to Japan, the model reproduced well ^{137}Cs concentrations using the zoom version over Asia (zAsia) showing high correlations, while more than 64% of the modelling results were found within a factor of two from the observations and more than 92% within a factor of 10. Labile and refractory rare radionuclides calculated indirectly showed larger deviations, with about 60% of the simulated concentrations within a factor of 10 from the observations. We estimate that around 23% of the released ^{137}Cs remained into Japan, while 76% deposited in the oceans. Around 163 TBq deposited over North America, among which 95 TBq over USA, 40 TBq over Canada and 5 TBq over Greenland). About 14 TBq deposited over Europe (mostly in the European part of Russia, Sweden and Norway) and 47 TBq over Asia (mostly in the Asian part of Russia, Philippines and South Korea), while traces were observed over Africa, Oceania and Antarctica. Since the radioactive plume followed a northward direction before its arrival to USA and then to Europe, a significant amount of about 69 TBq deposited in the Arctic, as well. These patterns of deposition are fully consistent with the most recent reports for the accident.

1. Introduction

The accident at the nuclear complex of Fukushima on March 11th, 2011 resulted in a severe release of around 135 radionuclides (IRSN, 2012). It was the consequence of a high-magnitude earthquake (~9.0), which created several massive tsunamis that struck Japan almost 1 h later (Akahane et al., 2012) causing power loss, disruption of the cooling systems and finally hydrogen explosions. As a result, several radionuclides in gaseous or aerosol form were released at elevated levels and dispersed over long distances following the prevailing meteorological conditions of the following days. Air leakages from the FDNPP stopped around 42 d after (Stohl et al., 2012).

Since the accident, substantial attempts have been made by the radioecological community to address the exact spread (e.g. Christoudias and Lelieveld, 2013) and deposition of radioactive fallout (e.g. Evrard et al., 2012; Gudelis et al., 2012), as well as the health effects on humans and animals (Aliyu et al., 2015; Garnier-Laplace et al., 2011; Møller et al., 2013; Ten Hoeve and Jacobson, 2012; WHO, 2013). These efforts involved either measurements from all over the world (e.g. Masson et al., 2011; McMullin et al., 2012; Paatero et al., 2012; Pham et al., 2012) or employment of atmospheric dispersion modelling (Christoudias and Lelieveld,

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2013; Evangeliou et al., 2014; Kristiansen et al., 2012; Stohl et al., 2012).

Modelling has been proved to be a very powerful tool when assessing radioactive releases and conducting predictions of the final receptor points of fallout. The accuracy of the results affects scientific significance as transport and deposition of radioactive plumes are usually used for health assessments prior to epidemiological (when dealing with humans) or evolutionary ones (when dealing with non-human biota) (e.g. Evangeliou et al., 2013a). Nevertheless, the reliability of the modelling results can only be confirmed via validation with long-range observations prior to their use in health assessments. Uncertainties are attributed to (a) the source term of the release, (b) the meteorological package used (European Centre for Medium-Range Weather Forecasts, Global Forecast System etc.) and (c) the scavenging scheme (in-cloud, below-cloud scavenging etc.).

In this study we present a detailed comparison of the response of an Eulerian model with observations from the accident of Fukushima. For this reason, several different model versions were employed, which are characterized by differences in the horizontal and/or vertical resolution. Data from the global radionuclide network of the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) were used to validate our model using the aerosol-bound tracer ^{137}Cs and the noble gas ^{133}Xe (data were adopted from Christoudias and Lelieveld, 2013). Moreover, for a more localized validation over Europe and Japan, apart from CTBTO, measurements of ^{134}Cs, ^{137}Cs and ^{131}I from the Greek monitoring system were used, as well as random measurements of ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{129m}Te, ^{132}Te, ^{95}Nb, ^{90}Sr, ^{110m}Ag, ^{99}Mo, ^{241}Am, ^{238}Pu, $^{239-240}$Pu, and ^{241}Pu from several Japanese research groups. Furthermore, we estimate global lifetimes of ^{137}Cs after the accident and compare them with calculated ecological half-lives of ^{137}Cs with respect to radioecological notion. Transport efficiencies of ^{137}Cs to continents, oceanic and marine systems and several countries across the world are presented as an indication of the expected radioactive impact and they are associated with estimated dose–rates from deposition of ^{137}Cs.

2. Atmospheric transport modelling and setup

We used the LMDz-OR-INCA global chemistry-aerosol-climate model, which couples the LMDz (Laboratoire de Météorologie Dynamique) General Circulation Model (GCM) (Hourdin et al., 2006) and the INCA (INteraction with Chemistry and Aerosols) model (Hauglustaine et al., 2004). The interaction between the atmosphere and the land surface is ensured through the coupling of LMDz with the ORCHIDEE (ORganizing Carbon and Hydrology In Dynamic Ecosystems) dynamical vegetation model (Krinner et al., 2005). We used five different configurations (Fig. 1), that present a resolution of (i) 2.50° × 1.27° (144 grid-cells in longitude, 142 in latitude) over 19 hybrid vertical levels extending to the stratosphere (from now on 144 × 142 × 19), (ii) the same horizontal resolution (2.50° × 1.27°) over 39 vertical levels (referred to as 144 × 142 × 39), (iii) a very intensive (in terms of computing time) version of 1.29° × 0.94° (280 grid-cells in longitude, 192 in latitude) over 39 levels (from now on referred to as 280 × 192 × 39) and two zoom-versions centered over (iv) Europe (zEUR) and over (v) Japan (zASIA) in the configuration 144 × 142 over 19 and 39 vertical levels, respectively. To create the zoom-versions, the GCM offers the possibility to stretch the grid over specific regions keeping the same overall number of grid points. Hence, the model finally achieved a maximum resolution inside the zoom of 0.45° × 0.51°. A more detailed description and an extended evaluation of the GCM can be found in Hourdin et al. (2006). The large-scale advection of tracers is calculated based on a monotonic finite-volume second-order scheme (Hourdin and Armengaud, 1999). Deep convection is parameterized according to the scheme of Emanuel (1991). All the detailed schemes used in the module are presented in detail in Szopa et al. (2012).

The INCA module simulates the distribution of anthropogenic and natural aerosols and gasses. The aerosol module keeps track of both the number and the mass of aerosols using a modal approach to treat the size distribution, which is described by a superposition of five log-normal modes (Schulz, 2007), each with a fixed spread. To treat the optically relevant aerosol size diversity, particle modes exist for three ranges: sub-micronic (diameter < 1 μm) corresponding to the accumulation mode, micronic (diameter between 1 and 10 μm) corresponding to coarse particles, and super-micronic or super coarse particles (diameter > 10 μm). This treatment in modes is computationally much more efficient compared to a bin-scheme (Schulz et al., 1998). Furthermore, to account for the diversity in chemical composition, hygroscopicity, and mixing state, we distinguish between soluble and insoluble modes. In both submicron and micron size, soluble and insoluble aerosols are treated separately. Cesium-134, ^{136}Cs and ^{137}Cs were treated as soluble submicronic aerosols (e.g. Masson et al., 2013) within the model, while ^{133}Xe was treated as a gas, which means it is lost only due to physical decay ($t_{½}$ = 5.2 d). Iodine-131, ^{129m}Te, ^{132}Te, ^{95}Nb, ^{90}Sr, ^{110m}Ag, ^{99}Mo, ^{241}Am, ^{238}Pu, $^{239-240}$Pu, and ^{241}Pu were estimated using site-specific radioactivity ratios reported by Bossew (2013), Chaisan et al. (2013), Kinoshita et al. (2011), Lujaniene et al. (2012), Yamamoto (2013), Yamamoto et al. (2014a and 2014b), and Zheng et al. (2012).

The simulations lasted from January 1st, 2011 to December 31st, 2011. The model (LMDz-OR-INCA) runs in a nudged mode using the ERA Interim re-analysis data – 6 h wind fields, (ECMWF, 2011) with a relaxation time of 10 d for the regular grid (Hourdin and Issartel, 2000), whereas for the zoom version relaxing to 4.8 d in the center of the zoom and to 10 d outside. The scientific community still argues about the exact amounts of the releases. Uncertainties in model-predicted concentrations and depositions are directly related to uncertainties in source release rates. Terada et al. (2012) reported the total release of ^{137}Cs to be approximately 13 PBq (× 10^{15} Bq), based on an inverse estimation of the source term using Japanese stations only, whereas the IRSN reported releases of ^{137}Cs to be 20.6 PBq (IRSN, 2011). In the present study, we used the emission inventories for cesium isotopes and ^{133}Xe reported by Stohl et al. (2012) estimated by inverse modelling using the CTBTO network corresponding to a ^{134}Cs emission of 33 PBq, ^{136}Cs of 7.3 PBq, ^{137}Cs of 36.7 PBq and a ^{133}Xe emission of 15.3 EBq.

3. Aerosol lifetime of ^{137}Cs and effective half-life

Several definitions for atmospheric lifetime exist. In any domain of Earth's atmosphere the mass balance can be expressed as:

$$\frac{dB(t)}{dt} = S(t) - \frac{B(t)}{\tau(t)} \quad (1)$$

where $B(t)$ is the atmospheric burden, $S(t)$ is the release rate and $\tau(t)$ is the removal time over a given time-step. If one assumes equilibrium between release rate and deposition (steady state conditions), the mean steady state lifetime (τ_{ss}) will be:

$$\tau_{ss} = \frac{\overline{B}}{\overline{D}} \quad (2)$$

where $\overline{B}$ and $\overline{D}$ are the mean atmospheric burden and deposition over a given period (Croft et al., 2014). Finally, the instantaneous lifetime ($\tau_{instant}$) is used to characterize exponentially decreasing