An estimation of the radioactive ^{35}S emitted into the atmospheric from the Fukushima Daiichi Nuclear Power Plant by using a numerical simulation global transport

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We present a numerical study carried out with the SPRINTARS model modified to account for the radioactive decay of ^{35}S compounds emitted from the Fukushima Daiichi nuclear power plant station after the hydrogen and vapor blast. The transport dynamics of the released material reproduced previous field observations. Four different emission scenarios were compared to the measurements of atmospheric ^{35}S in sulfate collected in La Jolla, Tsukuba, Kashiwa and Yokohama. Linear regressions of the relation between emitted and transported material that reached the sampling sites were used to estimate the amount of ^{35}S atoms and the amount of neutrons released in to the atmosphere. We estimate that a lower limit of 1.9×10^{16} ^{35}S atoms sec^{-1} were released after the events in March and this flux dropped to $4–39 \times 10^{14}$ ^{35}S atoms sec^{-1} at the end of the month. Based on this calculations we estimated a lower limit of 5.2×10^{21} slow neutrons m^{-2} sec^{-1} were emitted from the nuclear fuel rods to the sea water injected in the reactors after the events in March.

Keywords: Fukushima Daiichi nuclear power plant, radioactive isotopes, sulfate, global transport model, SPRINTARS

INTRODUCTION

The massive earthquake that affected the northeast coast of Japan on March 11th released a powerful Tsunami which hit the shores of Fukushima prefecture. The power shortage occasioned by this natural disaster produced the meltdown in the reaction vessels of units 1, 2 and 3 of the Fukushima Daiichi nuclear power plant produced a hydrogen fueled vapor blasts that lasted for a period of several days until March 15. Consequently, several radioactive species were released into the boundary layer, an event of such magnitude that has not been seen since the Chernobyl disaster and the extent, significance and consequences of such tragedy will remain a topic of research and monitoring for many years. The material released into the boundary layer includes a series of radioactive materials including radio nuclei of Iodine-131 (^{131}I), cesium-137 (^{137}Cs) (Chino et al., 2011; Yoshida and Takahashi, 2012; Yoshida and Kanda, 2012) and among its minor constituents sulfur-35 isotope (^{35}S).

^{35}S is a radionuclide with a half life of 87 days produced in the atmosphere by cosmic ray spallation of Argon-40 (Lal and Peters, 1967). Once produced, ^{35}S is rapidly oxidized to ^{35}SO$_2$ (~1 sec.) and by further oxidation this radionuclide containing molecule forms chemically stable sulfates (^{35}SO$_4^=$). Since all chemical properties of stable and radio isotopes are similar to the most abundant one (^{32}S, in this case), the aerosol formation process followed by wet and dry removal from the atmosphere is the same for all isotopic species. Due to its short half life, most of ^{35}SO$_2$ and ^{35}SO$_4^=$ produced in the stratosphere does not readily reach the troposphere before decaying to ^{35}Cl except during stratosphere-troposphere air exchange. The study of radionuclides produced by nuclear detonations over sea water has shown the presence of ^{35}S produced by slow neutron capture of ^{35}Cl via ^{35}Cl(n,p)^{35}S reaction (Dyrssen and Nyman, 1955). ^{35}S is also produced by slow neutrons via ^{34}S(n,γ)^{35}S. However, this process is 200 times slower than ^{35}Cl(n,p)^{35}S (Love and Sam, 1962). ^{35}S production from both pathways are likely to have taken place during the injection of sea water within the reaction vessels of units 1, 2 and 3.

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Priyadarshi *et al.* (2011) measured ^{35}S activity in sulfate collected at Scripps Institution of Oceanography (SIO), La Jolla California during March–April 2011. They observed a distinct increase in ^{35}S activity (1500 atoms m^{-3}) from the natural background (450 atoms m^{-3}) on 28 March 2011. Based on measurements and mathematical modeling, they explained that the observed enhancement in $^{35}SO_4^=$ was due to the long range transport of radioactive $^{35}SO_4^=$ produced within the nuclear reactor in Fukushima nuclear power plant. ^{35}S activity measured at La Jolla peaked after the hydrogen blasts at the Fukushima nuclear power plant. Their results complemented with a moving box model study estimated a concentration of 2 $\times 10^5$ molecules m^{-3} of ^{35}S in the marine boundary layer over Fukushima. An additional report by the same group (Priyadarshi *et al.*, in prep.) presented the fluctuation of radioactive species $^{35}SO_2$ and $^{35}SO_4^=$ monitored at the Tokyo Institute of Technology (TITECH), Atmosphere and Ocean Institute (AORI) and National Institute of Environmental Studies (NIES) in the Kanto area of Japan. At these monitoring sites, a maximum of 11778, 61402 and 5019 atoms m^{-3} were respectively measured during March 25–April 7.

A numerical simulation that models the global transport of atmospheric particles after the major emission of radioactive materials has been reported by Takemura *et al.* (2011). Their results replicate the transport and dissipation patterns reported by numerous measurements and simulations carried out throughout the planet (See Takemura *et al.* (2011) and references there in). Here we present the results of using a modify version of the code employed by Takemura *et al.* (2011) where radioactive decay of $^{35}SO_4^=$ is added to the simulation. Our results are compared to the timing and amount of ^{35}S material detected in California and Tokyo. Using field measurements as constraining conditions, we estimate the bulk amount of ^{35}S material released from the nuclear power plant.

MODEL DESCRIPTION

The model employed for this numerical study is a spectral radiation-transport model for aerosol species (SPRINTARS) developed by Takemura and co-workers (Takemura *et al.*, 2000, 2002, 2005). This code has been developed to calculate physical processes involved in the transport of particles including emission, molecular and turbulent diffusion, advection by inter-grid air motion, while deposited species are conformed by gravity, wet and dry deposition. The dynamic core of SPRINTARS is based on the atmospheric component of a global climate model MIROC (Watanabe *et al.*, 2010) and therefore is capable of calculating global atmospheric state, such as air motion, temperature, moisture, clouds and precipita-
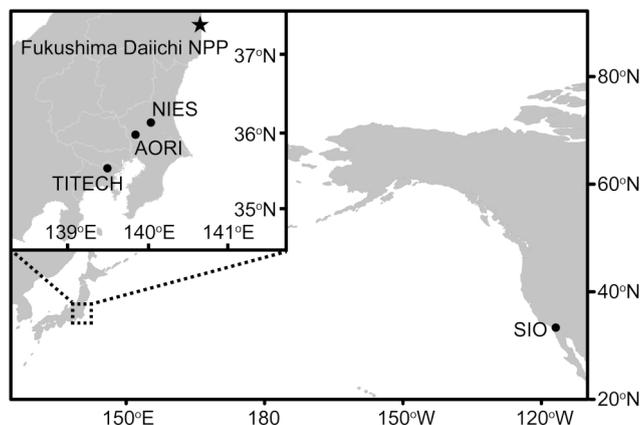

Fig. 1. Geographical position of Fukushima Daiichi nuclear power plant (37.4°N, 141.8°E) and $^{35}SO_4^=$ sampling sites TITECH (35.3°N, 139.3°E), AORI (35.9°N, 140.0°E), NIES (36.1°N, 140.1°E) and SIO (32.8°N, 117.3°W).

tion. The model grid consists of 20 vertical levels where the lowermost 4 are located below 1 km altitude while the horizontal resolution is approximately 0.56° by 0.56° in latitude and longitude (T213 spectral truncation in the dynamical core). In order to achieve higher accuracy, the atmospheric state generated by the dynamic core was assimilated to a 6-hourly data based on dynamic observations.

The release of material was approximated in the model by injecting particles in the lowest layer at the grid point where the nuclear power plant is located (Fig. 1). The emission of particles was kept constant from 1200 UTC, March 14 when measured gamma radiation was above 2 mSv/h at the plant (TEPCO, 2011), until the end of the simulation on April 10. This assumption is taken on the bases that the ^{35}S emissions are not known and the measured amounts of gamma radiation are a proxy of the released material, therefore from a model point of view the simulation represents the ratio in concentration between the emission domain and any given point at considerable distance. A more detailed presentation of the emission scenario is presented in Takemura *et al.* (2011). In this experiment the authors considered the emission to be composed of ^{137}Cs and other radioactive materials. In our study, emitted material is composed of sulfate aerosols since ^{35}S formed in the reactor is rapidly converted to $^{35}SO_4^=$, since the generated ^{35}S atom is highly unstable and is likely to be oxidized to $H_2^{35}SO_4$ (Giulianelli and Willard, 1974). The relation between mode radii (μm) and relative humidity is a deciding factor in the simulation of dry, wet and gravitational settling. In this study, the radii values for a 0–99% calculated relative humidity range was 0.0695–0.231 μm. For a detailed description and formulation of the computations of aerosols and their proper-
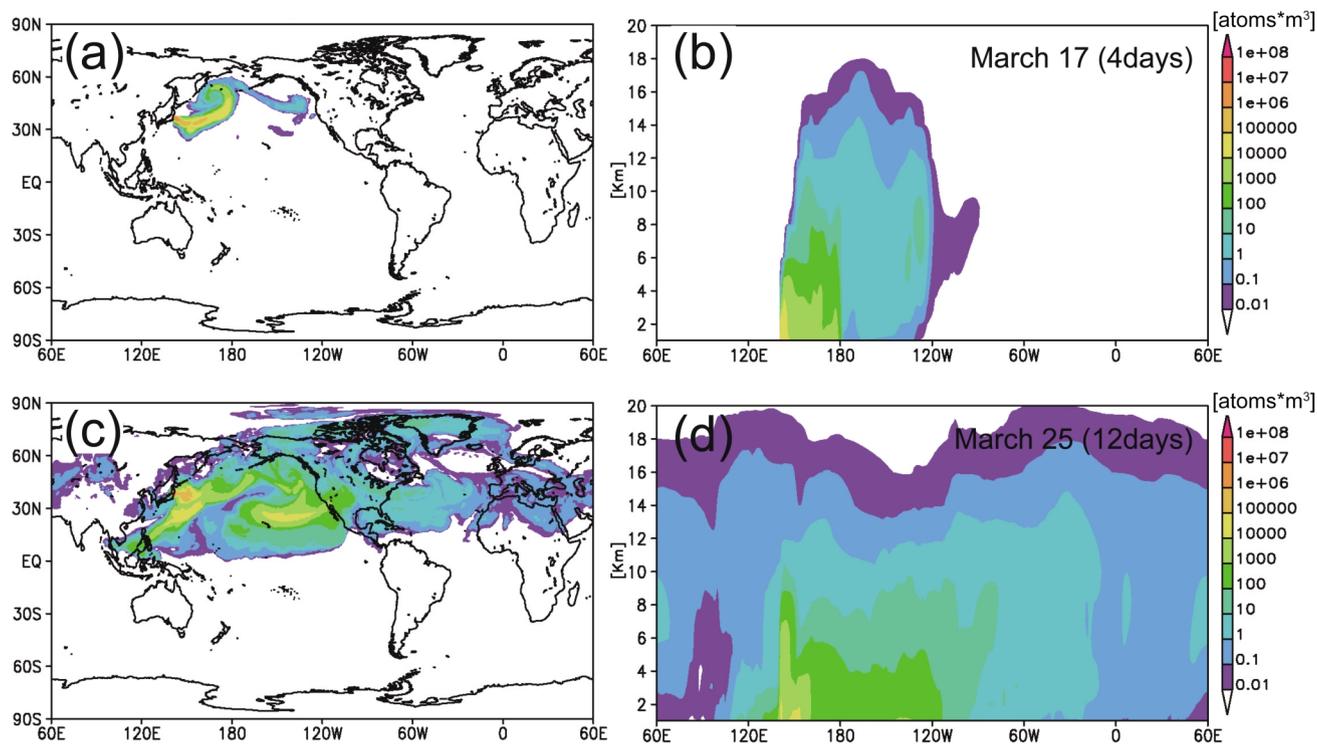
Fig. 2. Simulated near surface concentration of $^{35}SO_4^=$ material emitted continuously (1.43 × 10^{16} emission scenario) presented for the entire globe for March 17 (a) and March 25 (c). Panel b and d present latitudinal mean concentration in the northern hemisphere on the longitudinal vs. altitude plot. The chromatic scale represents atoms m^{-3}.

ties, see Takemura *et al.* (2000, 2002, 2005). The code has been modified to take into account the radioactive decay (half life 87 days) of emitted $^{35}SO_4^=$. In this model the mass balance of the emitted material consist of wet, dry, gravitational deposition and radioactive decay (81%, 11%, 0.02% and 0.7%, respectively).

The emission scenario for this study was set to 1.43 × 10^{16} estimated from the numerical values presented by Priyadarshi *et al.* (2011) and expanded to 3 different emission scenarios (1.43 × 10^{14}, 1.43 × 10^{15}, and 1.43 × 10^{17} atoms s^{-1}). Once the linearity of the relation between released material and the material that reached SIO, NIES, AORI and TITECH was confirmed, the number of atoms of ^{35}S emitted after the considered events (hydrogen and vapor blasts) were estimated.

RESULTS AND DISCUSSION

Figure 2 shows the simulated distribution of ^{35}S concentration emitted from Fukushima Daiichi nuclear power plant. The numerical data presented in this figure correspond to the scenario where 1.43 × 10^{16} atoms s^{-1} were released into the lowermost layer of the simulation grid. Panel a and b of Fig. 2 shows the initial wave of $^{35}SO_4^=$ species being transported eastwards. The simulation shows that the initial wave (concentrations above 10^{-13} atoms m^{-3}) reached 120°W latitude on March 17. The simulation presented in this report shows a transport time of 4 days since the initial injection of radioactive material. Panel c and d of Fig. 2 shows that the concentration of the transported $^{35}SO_4^=$ species arriving at La Jolla exceeded 1000 m^{-3} on March 25, 12 days after the initial blast. This maximum was also measured at La Jolla in a sample retrieved during March 24–28 (Priyadarshi *et al.*, 2011). After this 12 days maximum, the emitted $^{35}SO_4^=$ species is dispersed within 16 km altitude layer of the northern hemisphere.

Figure 3 presents simulated temporal variations of $^{35}SO_4^=$ compounds concentrations at 4 different sites within Japan (TITECH, NIES and AORI) and US (SIO). Since the emission flux of 1.43 × 10^{16} atoms s^{-1} was kept constant, large peaks sampled within Tokyo area and La Jolla suggest specific wind patterns of transported material with high concentration levels of $^{35}SO_4^=$ from the emission source. In order to validate the inferred conclusions, our calculations have been compared to field measurements carried out at TITECH, NIES, AORI and SIO and reported by Priyadarshi *et al.* (2011, in preparation). The simulation presents peaks with concentrations of $^{35}SO_4^=$ material of above 10000 atoms m^{-3} in Tokyo area

Site	Observed term	Measured $^{35}SO_4^=$ (atoms m^{-3})	^{35}S emission flux ($\times 10^{14}$ atoms s^{-1})	Neutron flux ($\times 10^{20}$ m^{-2}s^{-1})
SIO	24–28 March	1050*	183	52
AORI	23–25 March	17074	91	26
AORI	28–30 March	13893	4	1
AORI	30 March–1 April	61402	39	11
NIES	29 March–1 April	18049	4	1
TITECH	28–30 March	11778	7	2

*This value is corrected for the background levels of ^{35}S measured between March 2 and 9.
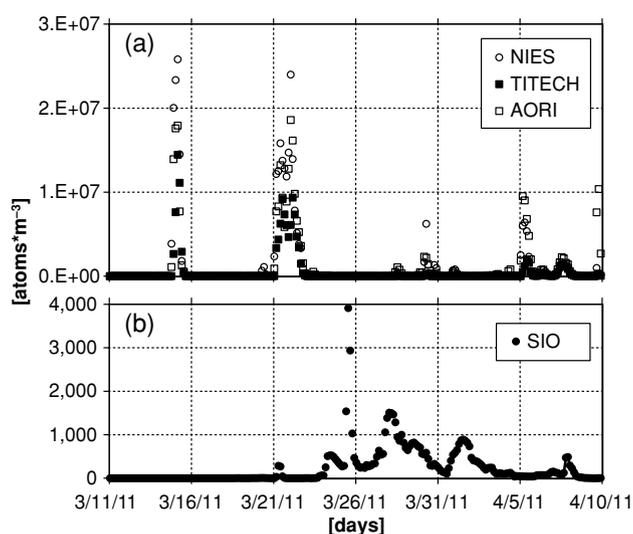

Fig. 3. Simulated temporal variation of $^{35}SO_4^=$ compounds concentrations at 1.43×10^{16} emission scenario for NIES, TITECH and AORI Japan (a) and SIO, US (b).

during the periods of March 14–15, March 20–24, March 28–April 1, April 3–8 and April 9 (Fig. 3a), and peaks of above 1000 atoms m^{-3} measured at SIO during March 25 and March 27–28 period (Fig. 3b). These modeled peaks replicate the timing of appearance with high degree of accuracy with the measurements of Priyadarshi *et al.* (2011 and in prep.) carried out in Japan (March 23–25, March 28–April 4) and US (March 24–28). The time evolution of the released material from the power plant shows that the measured peaks represent events where the emitted material was transported by southeastward winds in the case of Tokyo area and the North Pacific Jet stream for the case of SIO.

From the 4 calculated case scenarios, a linear regression of the relation between emitted flux and transported material concentration at each site was constructed and the measured amounts of ^{35}S atoms were employed to calculate the specific emitted flux of ^{35}S material from the nuclear reactors. Calculated fluxes were then employed to estimate the amount of slow neutrons emitted from the nuclear fuel rods that activated the sea water within the reactors. The methodology of this conversion was the same as the one employed by Priyadarshi *et al.* (2011). Table 1 presents a summary of observed $^{35}SO_4^=$, calculated ^{35}S emission flux and released neutron amount. Since the background levels of ^{35}S atoms produced by cosmic rays spallation have some fluctuation, data sets with large concentrations of ^{35}S species (above 10000 atoms m^{-3}) were employed for the case of Tokyo but background corrected values were employed for the measurements at SIO. Backtrace analysis presented by Priyadarshi *et al.* (2011) of ^{35}S species at SIO between March 24–28 shows that it took 10 days of transport since its, based on this analysis we estimate that 183×10^{14} atoms s^{-1} were emitted from the nuclear power plant between March 14 and 18 (Table 1). The measurements at AORI between March 23 and March 25 were used to estimate an emission of 91×10^{14} atoms s^{-1}. Joint measurements at AORI, NIES and TITECH between March 28 and April 1 estimate an emission of $4–39 \times 10^{14}$ atoms s^{-1} that were transported within a day from the source. Based on these results, the emission simulated between March 28 and April 1 represents approximately 8% of the initial emissions simulated between March 14–18. Chino *et al.* (2011) study on the time variation of released main radioactive nuclei ^{131}I and ^{137}Cs estimates a reduction in the order of 10–100 from the emission on March 14 until sometime between March 28 and April 1. If this initial amount of material is taken as representative of the occurred events we estimate that 52×10^{20} slow neutrons m^{-2} sec^{-1} were released from the fuel rods into the sea water present within the nuclear reactor. Priyadarshi *et al.* (2011) have estimated a release amount of 4×10^{11} slow neutrons m^{-2}. The large difference with our estimation comes from the intrinsic limit of the box model study by Priyadarshi

et al. (2011), which takes into account deposition and decay during the trans-Atlantic transport but is limited to estimate the actual amount of ^{35}S atoms present at inside the reactor. By considering different scenarios our model directly estimates the amount of material released from the reactor core. The estimated initial ^{35}S material and number of neutron represent a lower limit of the amount of radiation emitted from the nuclear reactors at Fukushima Daiichi nuclear power plant. These values can be used as a proxy to the total amount of radiation emitted since the melt down.

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