

Evidence of neutron leakage at the Fukushima nuclear plant from measurements of radioactive ^{35}S in California

Antra Priyadarshi, Gerardo Dominguez, and Mark H. Thiemens¹

9500 Gilman Drive, University of California, San Diego, La Jolla, CA 92093

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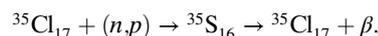
A recent earthquake and the subsequent tsunami have extensively damaged the Fukushima nuclear power plant, releasing harmful radiation into the environment. Despite the obvious implication for human health and the surrounding ecology, there are no quantitative estimates of the neutron flux leakage during the weeks following the earthquake. Here, using measurements of radioactive ^{35}S contained in sulfate aerosols and SO_2 gas at a coastal site in La Jolla, California, we show that nearly 4×10^{11} neutrons per m^2 leaked at the Fukushima nuclear power plant before March 20, 2011. A significantly higher $^{35}\text{SO}_4^{2-}$ activity as measured on March 28 is in accord with neutrons escaping the reactor core and being absorbed by the coolant seawater ^{35}Cl to produce ^{35}S by a (n, p) reaction. Once produced, ^{35}S oxidizes to $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ and was then transported to Southern California due to the presence of strong prevailing westerly winds at this time. Based on a moving box model, we show that the observed activity enhancement in $^{35}\text{SO}_4^{2-}$ is compatible with long-range transport of the radiation plume from Fukushima. Our model predicts that $^{35}\text{SO}_4^{2-}$, the concentration in the marine boundary layer at Fukushima, was approximately 2×10^5 atoms per m^3 , which is approximately 365 times above expected natural concentrations. These measurements and model calculations imply that approximately 0.7% of the total radioactive sulfate present at the marine boundary layer at Fukushima reached Southern California as a result of the trans-Pacific transport.

gas to particle conversion | radioactive sulfur-35

The Fukushima nuclear power plant, situated in northeastern Japan, is one of the largest nuclear power stations in the world, consisting of six boiling water reactors. A recent massive earthquake (9.0 magnitude) (<http://earthquake.usgs.gov/earthquakes/eqinthenews/2011/usc0001xgp/>) on March 11, 2011, followed by a deadly tsunami has extensively damaged the power plant by disabling the reactor cooling system, which led to nuclear radiation leaks and activation of a 20-km evacuation zone surrounding the plant (<http://www.iaea.org>). Three weeks subsequent to the earthquake, evidence surfaced of a partial nuclear meltdown in units 1, 2, and 3, visible explosions in units 1 and 3, and a possible uncovering of spent fuels pool associated with units 1, 3, and 4 (<http://www.iaea.org>). It is suspected that the explosions at unit 3 may have damaged the primary containment vessel. The events at units 1, 2, and 3 have been rated at level 7 (major release of radioactive material with widespread health and environmental effects requiring implementation of planned and extended countermeasures) on the International Nuclear Event Scale (<http://www.iaea.org>). The earthquake triggered the automatic shutdown of the three operating reactors—units 1, 2, and 3—and the control rods were completely inserted to terminate the nuclear fission reaction occurring within the fuel core. The details of a nuclear reactor design and operations are described by ref. 1. Residual heat produced at the reactor core due to the radioactive decay of fission products requires extended time periods to cool. Heat must be removed by cooling the system

to prevent the fuel rods from overheating and causing a core meltdown, which results in injection of neutrons and other fission products into the atmosphere. Spent fuel that has been removed from a nuclear reactor generates intense heat and is continuously cooled to provide protection from its radioactivity. As reported, maintaining sufficient cooling to remove the decay heat from the reactor was crucial; consequently, the spent fuel pool was the main challenge at the affected reactor site after the March 11 tsunami.

Because both on- and off-site power to the plant was disabled, seawater mixed with boric acid (to reduce the neutron flux in the core and thus slow down the nuclear reaction) was continuously pumped into the reactor vessels of units 1, 2, and 3 from March 13 to March 26, 2011 (<http://www.iaea.org>). On March 17, seawater was sprayed by helicopters and by concrete pumps on reactor unit 3 and spent fuel containment. The steam generated due to overheating was released through the relief valve into the atmosphere to avoid explosive hydrogen buildup within the reactor vessel. Approximately a few hundred tons of seawater were used as a coolant before switching to fresh water. A consequence is that salts and minerals present in seawater become radioactive by reaction with thermal neutrons emitted from the reactor. For example, nuclear detonation in seawater produces ^{35}S by slow neutron capture by ^{35}Cl via the (n, p) reaction (2). As described by ref. 3, mainly two isotopes, ^1H and ^{35}Cl , present in seawater absorb the slow neutrons. ^{35}Cl , which is highly abundant in seawater (0.55 mol/kg), absorb neutrons to produce radioactive ^{36}Cl and ^{32}P via $^{35}\text{Cl}[n,\gamma]^{36}\text{Cl}$ and $^{35}\text{Cl}[n,\alpha]^{32}\text{P}$ reaction, respectively. ^{35}Cl is also converted to radioactive ^{35}S :



$^{35}\text{Cl}[n,p]^{35}\text{S}$ has a substantial cross-section of $0.44 \pm 0.01\text{b}$ for thermal neutrons (4, 5). ^{35}S is also produced via $^{34}\text{S}(n,\gamma)^{35}\text{S}$, but approximately 200 times less than $^{35}\text{Cl}(n,p)^{35}\text{S}$ (2). The chemical state of ^{35}S produced after neutron capture is not known, but it is expected that ^{35}S is oxidized into $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ (6). We assumed that all ^{35}S produced in seawater is transferred to the atmosphere along with the water steam generated at the reactor core due to excessive heating.

Radioactive ^{35}S (β decay to ^{35}Cl , half-life approximately 87 d) is naturally produced in the atmosphere by cosmic ray spallation of ^{40}Ar (7). Once produced, ^{35}S rapidly oxidizes to $^{35}\text{SO}_2$ (approximately 1 s), which is removed from the atmosphere by wet and dry deposition. $^{35}\text{SO}_2$ may undergo gas and aqueous phase oxidation to produce $^{35}\text{SO}_4^{2-}$ aerosols, which are removed

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¹To whom correspondence should be addressed. E-mail: mthiemens@ucsd.edu.

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from the atmosphere. ^{35}S is the only radioactive isotope that simultaneously exists in both gas and aerosol phases and has a suitably short half-life to detect air motions, which renders it a sensitive tracer in understanding boundary layer chemistry and air mass transport on short time scales (8, 9). ^{35}S has recently been measured in sulfate aerosols in the Antarctic atmosphere to understand the boundary layer chemistry and stratospheric-tropospheric air mass exchange (10). ^{35}S measurements in sulfate collected from snow and water samples have been measured to determine the source and age of the ground water and also to understand the melting of glaciers and their contribution to lake and stream water (11).

The simultaneous gas and particle speciation and short half-life render ^{35}S a unique isotope to clock gas to particle transformation and transport. Any ^{35}S produced at the Fukushima nuclear plant as a result of earthquake-related damage and seawater-based cooling, combined with its speciation into both the particle and gas phases, could serve as a point source of artificially made ^{35}S and provides an excellent opportunity to understand gas-to-particle conversion and long-range transport over the Pacific.

Result and Discussion

We report ^{35}S measurements of atmospheric SO_2 and sulfate aerosols collected at Scripps Institution of Oceanography (SIO) Pier, La Jolla (32.8N, 117.3W, 10 m amsl) between March 9 and April 12, 2011. As shown in Fig. 1 (Table 1), $^{35}\text{SO}_4^{2-}$ activity typically varies between 180 and 475 atoms m^{-3} . A significant increase in $^{35}\text{SO}_4^{2-}$ activity (1,501 atoms m^{-3}) was observed on March 28, which is the highest activity ever recorded at this sampling site. Similarly, $^{35}\text{SO}_2$ concentrations increased to 120 atoms m^{-3} on March 28 from its background activity of 30–40 atoms m^{-3} .

We have continuously been measuring $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ at SIO since February 2009, and thus the background ^{35}S activity is well characterized at the sampling site before the Fukushima nuclear plant tragedy transpired. The annual average activity in $^{35}\text{SO}_4^{2-}$ and $^{35}\text{SO}_2$ is 458 ± 157 and 72 ± 61 atoms m^{-3} , respectively. Brothers et al. (9) reported a similar background activity at the Scripps Pier during 2008. At SIO, $^{35}\text{SO}_4^{2-}$ ($^{35}\text{SO}_2$) values fluctuate between 300 and 500 (100–150) atoms m^{-3} throughout the year except a few days when $^{35}\text{SO}_4^{2-}$ ($^{35}\text{SO}_2$) activity spikes to 950 (430) atoms m^{-3} , respectively. This sudden spike in activity

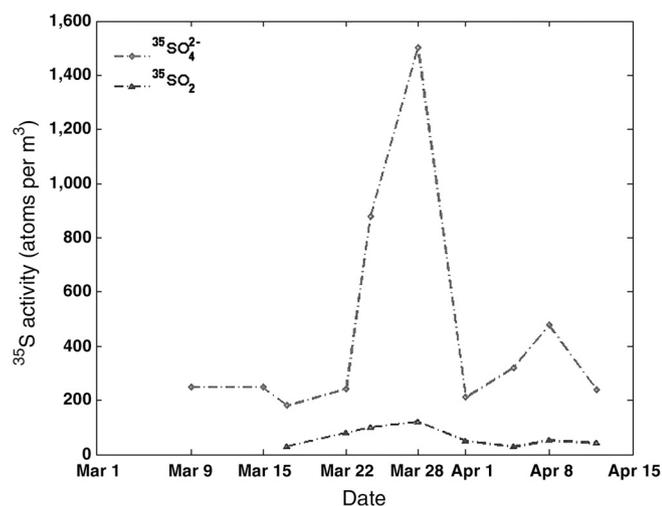


Fig. 1. ^{35}S activity measured in SO_2 gas and sulfate aerosol collected at SIO, La Jolla. A significant increase in $^{35}\text{SO}_4^{2-}$ concentration was observed on March 28, 2011, from the natural background $^{35}\text{SO}_4^{2-}$ activity. The higher activity observed at SIO is due to the presence of higher concentration of $^{35}\text{SO}_4^{2-}$ (365 times higher than the natural abundance) at Fukushima and the radiation plume being transported over the Pacific.

is due to stratospheric intrusion events (spring) and the Santa Ana wind events (easterly high pressure drive winds during late fall–winter) (that lead to the mixing of higher altitude air containing higher ^{35}S activity into the marine boundary layer (MBL). The average $^{35}\text{SO}_4^{2-}$ activity observed during spring is 523 ± 190 (Mar–Apr 2009) and 557 ± 156 (Mar–Apr 2010), whereas for $^{35}\text{SO}_2$ is 72 ± 30 atoms m^{-3} . The $^{35}\text{SO}_4^{2-}$ activity observed on March 28 is higher (>factor of 2) than the spring average, whereas $^{35}\text{SO}_2$ values lie within 2σ deviation. Even though the polar region has the highest ^{35}S production rate, the $^{35}\text{SO}_4^{2-}$ activity measured at a coastal site in Antarctica (Dumont D’Urville station) is significantly lower (900 atoms m^{-3}) than the activity observed on March 28.

Box Model Calculation

We analyzed two different possible natural scenarios that might cause $^{35}\text{SO}_4^{2-}$ concentrations to deviate from the background to the level observed on March 28.

Scenario 1 (Local Stratospheric Intrusion Event). During stratospheric intrusion, stratospheric air masses containing higher $^{35}\text{SO}_4^{2-}$ and $^{35}\text{SO}_2$ are mixed to the upper free troposphere and are subsequently downward transported to the lower altitude. A 4-Box model, as described by ref. 10, with the model parameters in Table S1 was used to determine the steady state concentration of $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ at Scripps (Table S2) and to calculate the volume fraction of stratospheric air mass mixing into the free troposphere during the stratospheric intrusion event. The model calculation (see SI Text) shows that nearly 40% of stratospheric air mass must mix into the free troposphere to increase the $^{35}\text{SO}_4^{2-}$ activity to 1,500 atoms m^{-3} as observed on March 28. This is highly unlikely because most of the stratospheric air mass (90%) returns back to the stratosphere within 6 h of cross-tropopausal exchange (12), and only a small fraction (1–10%) is entrained into the free troposphere (10, 12, 13). In addition, relative humidity and surface ozone concentrations do not exhibit any noticeable change at SIO during the sampling time period typically associated with stratospheric incursions. Consequently, it is regarded as unlikely that a stratospheric intrusion event occurring at Scripps or any region lying in the downwind direction from Fukushima caused the observed enhancement in $^{35}\text{SO}_4^{2-}$ and $^{35}\text{SO}_2$ on March 28.

Scenario 2 (Long-Range Transport of the Radiation Plume). Long-range transport of gases and aerosol from east Asia to Southern California is most intense during springtime (14, 15) because of strong midlatitude westerly winds (16). The Fukushima nuclear plant served as a point source of $^{35}\text{SO}_4^{2-}$ and $^{35}\text{SO}_2$ between March 13 and 26, and a radiation plume from Fukushima would be expected to reach Southern California (Scripps) approximately between March 20 and April 1 if the transport time is 6–7 d (17, 18). A 10-d back trajectory for air masses arriving at Scripps on March 28 using a HYSPLIT (19) model reveals that the air mass originated at Japan (Fig. 2) and the long-range transport occurred in the boundary layer with a mean transit altitude of 0.9 km. A moving box model, where a well-mixed box moves along the air mass back trajectory (Fig. 3), was developed to calculate $^{35}\text{SO}_4^{2-}$ and $^{35}\text{SO}_2$ concentrations at La Jolla due to long-range transport of the radiation plume from Fukushima. The transit time and corresponding transit altitude of the air mass were calculated from a HYSPLIT model for the last 10 d before it arrived at SIO (Table S3). On day 8, the box passes over the region near Fukushima and subsequently spends 4 d in the marine boundary layer before reaching La Jolla on March 28. As shown in Fig. 3, $^{35}\text{SO}_4^{2-}$ aerosol and gaseous $^{35}\text{SO}_2$ change their concentration through various processes such as dilution due to eddy diffusion, dry deposition over the ocean, oxidation, and the radioactive decay during long-range transport. The model parameters

Table 1. The measurements of ^{35}S activity contained in SO_2 (gas) and sulfate aerosol collected at Scripps, La Jolla, California, during March–April 2011

Sample	Start date	End date	Total air flow, m^3	Total disintegration per minute (measured)	^{35}S , atoms/ m^3	Total $[\text{SO}_4^{2-}]$, ng/m^3
$^{35}\text{SO}_4^{2-}$						
3-2-2011	3-9-2011		13,269	18.0 ± 0.4	246 ± 2	3.8
3-9-2011	3-15-2011		9,518	12.9 ± 0.3	246 ± 3	6.6
3-15-2011	3-17-2011		3,338	3.3 ± 0.2	180 ± 6	6.1
3-17-2011	3-22-2011		7,819	10.4 ± 0.3	241 ± 3	18.8
3-22-2011	3-24-2011		3,479	16.8 ± 0.3	876 ± 7	4.8
3-24-2011	3-28-2011		6,502	53.8 ± 0.4	1501 ± 5	3.9
3-28-2011	4-1-2011		6,394	7.4 ± 0.2	210 ± 3	6.5
4-1-2011	4-5-2011		6,511	11.5 ± 0.3	320 ± 4	10.6
4-5-2011	4-8-2011		4,802	12.6 ± 0.3	475 ± 5	9.3
4-8-2011	4-12-2011		6,437	8.4 ± 0.3	237 ± 4	5.5
$^{35}\text{SO}_2$						
3-15-2011	3-17-2011		3,338	1.2 ± 0.1	30 ± 6	1.3
3-17-2011	3-22-2011		7,819	3.3 ± 0.1	80 ± 3	1.4
3-22-2011	3-24-2011		3,479	1.7 ± 0.1	100 ± 6	0.9
3-24-2011	3-28-2011		6,502	4.3 ± 0.1	120 ± 3	0.8
3-28-2011	4-1-2011		6,394	1.8 ± 0.1	50 ± 3	1.9
4-1-2011	4-5-2011		6,511	1.0 ± 0.1	28 ± 2	1.0
4-5-2011	4-8-2011		4,802	1.4 ± 0.1	52 ± 3	1.0
4-8-2011	4-12-2011		6,437	1.4 ± 0.1	41 ± 3	1.2

are described in detail (10) (see *SI Text* and [Tables S1](#) and [S3](#)). On day 10, the initial concentration of $^{35}\text{SO}_4^{2-}$ ($^{35}\text{SO}_2$) in the box is taken to be 913 (391) atoms m^{-3} , the same as the average activity in the free troposphere ([Table S2](#)). The bidirectional air mass exchange from the box occurs depending on the dilution lifetime (τ_{dil}) of 4.9 d (17). The air mass flux into the box is a ratio of the background concentration of ^{35}S , i.e., $[\text{SO}_2(z)]_b$ and $[\text{SO}_4^{2-}(z)]_b$ present outside the box at that particular altitude (z) to the dilution lifetime. In the model, we assumed that ^{35}S was released mainly in the $^{35}\text{SO}_4^{2-}$ phase at the reactor core. The model sensitivity test ([Table S4](#)) shows that the model results are not sensitive to the dimension of the moving box but is sensitive to the dilution lifetime (τ_{dil}). Other parameters, such as oxidation lifetime (τ_{ox}) and the initial concentration of $[\text{SO}_2(z)]_b$ and $[\text{SO}_4^{2-}(z)]_b$ in the box had a very small effect on the model results.

The model predicts that a $[\text{SO}_4^{2-}]_b$ concentration of 2×10^5 atoms m^{-3} was present in the buffer layer at Fukushima ([Table S5](#) and [Fig. S1](#)). [Table S5](#) shows the effect on the concentration of $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ arriving at Scripps due to different proportions of $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ concentrations in the buffer layer (BuL) at Fukushima. Even considering the same concentration

of $^{35}\text{SO}_2$ and $^{35}\text{SO}_4^{2-}$ in the BuL at Fukushima, the model does not show any increase in $^{35}\text{SO}_2$ concentrations at La Jolla. This is because the air mass spent 55% of its transit time in the marine boundary layer where $^{35}\text{SO}_2$ is rapidly lost due to its oxidation to $^{35}\text{SO}_4^{2-}$ with dry deposition over the ocean (18). It is estimated that nearly 24% of $^{35}\text{SO}_2$ is oxidized to $^{35}\text{SO}_4^{2-}$ in the buffer layer. In the marine boundary layer, 66% of $^{35}\text{SO}_2$ is lost due to higher dry deposition over the ocean and oxidation to $^{35}\text{SO}_4^{2-}$, which is in agreement with ref. 20. Based on the model calculation and the measurements, it is determined that nearly 0.7% of the total $[\text{SO}_4^{2-}]$ (present at the marine boundary layer at Fukushima) has been transported to the Scripps Pier, La Jolla, California, in a trans-Pacific transport.

The model calculated $^{35}\text{SO}_4^{2-}$ concentration in the MBL (2×10^5 atoms m^{-3}) at Fukushima is higher than the stratospheric concentration ([Table S2](#)) and suggests an additional source of ^{35}S production due to the $^{35}\text{Cl}(n,p)^{35}\text{S}$ reaction. We calculated the total number of neutrons that leaked from the reactor core to account for our observations. We use leakage for sea water irradiated by the neutrons produced inside the core. Because the reactor core was melted, the neutron releasing outside is termed as leak. The reaction cross-section of $^{35}\text{Cl}(n,p)^{35}\text{S}$ was

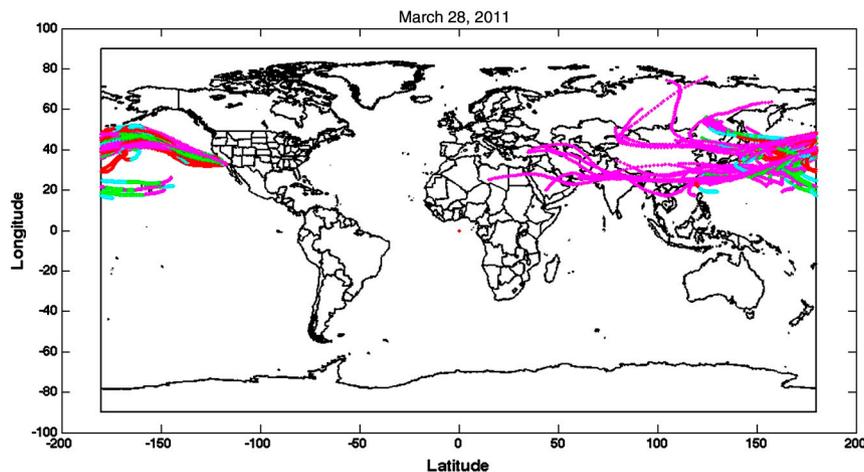


Fig. 2. Ten-day back trajectory obtained for the air mass reaching La Jolla, California, on March 28, 2011, from HYSPLIT model indicates that the air mass originated at Japan.

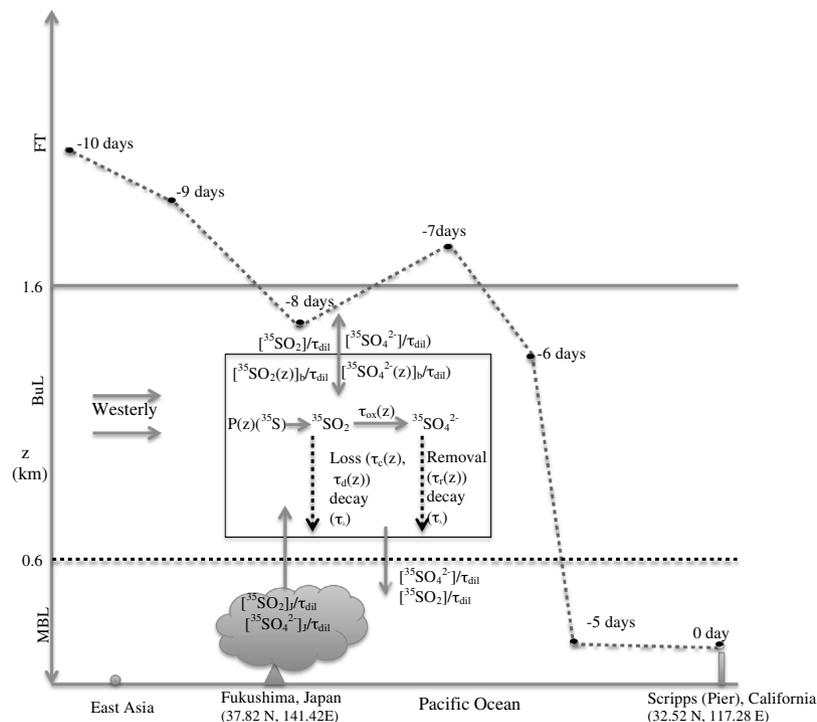


Fig. 3. Schematic description of a moving box model developed to calculate the concentration of $^{35}\text{SO}_4^{2-}$ and $^{35}\text{SO}_2$ at Scripps pier, La Jolla (California) due to the long-range transport of radiation plume from Fukushima. The atmosphere is divided into four layers: MBL, BuL, free troposphere (FT), and lower stratosphere (LS). τ_{dil} is the dilution lifetime of the radiation plume. τ_{ox} is the oxidation lifetime of $^{35}\text{SO}_2$, whereas τ_c and τ_d are the cloud scavenging and dry depositional lifetime of $^{35}\text{SO}_2$, respectively. τ_r is the removal lifetime of $^{35}\text{SO}_4^{2-}$ (see *SI Text*). The dotted line (red) represents the mean transit altitude at which air mass transport occurred; it was calculated from HYSPLIT model. The model parameters in the box were changed according to its transit altitude (Table S3).

taken from ref. 4. The attenuation length of neutrons in water at room temperature is 2.8 cm and increases at higher temperatures (21). Because of the high absorption cross-section of ^{35}Cl , seawater has more attenuation. The value of the attenuation length of the neutrons in seawater at temperatures higher than 1,000 °C is not known. For simplicity, the attenuation length was taken to be 2.8 cm. The concentration of $^{35}\text{SO}_4^{2-}$ at the source (reactor core) was assumed to be 10 times higher than the model-calculated $^{35}\text{SO}_4^{2-}$ concentration in the marine boundary layer. Considering all the possible reactions of neutrons with seawater (3), we estimate that a total of 4×10^{11} neutrons per m^2 were released before March 20 in which a fraction of 2×10^8 neutrons per m^2 reacted with ^{35}Cl to make ^{35}S .

Conclusion

We report the observation of radioactive ^{35}S produced by secondary reactions within the reactor core at the Fukushima power plant and estimate the total neutron leakage after the earthquake. The present work also provides a previously undescribed estimate of depositional and oxidation time scales of SO_2 and sulfate during trans-Pacific transport due to a singular strong and well-defined ^{35}S source. The sulfur data are unique because of the coexistence as gas and solid and adds previously undescribed insight into sulfur environmental transformational rates.

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