

## Ensemble Simulation of the Atmospheric Radionuclides Discharged by the Fukushima Nuclear Accident

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Enormous amounts of radionuclides were discharged into the atmosphere by a nuclear accident at the Fukushima Daiichi nuclear power plant (FDNPP) after the earthquake and tsunami on 11 March 2011. The radionuclides were dispersed from the power plant and deposited mainly over eastern Japan and the North Pacific Ocean. A lot of numerical simulations of the radionuclide dispersion and deposition had been attempted repeatedly since the nuclear accident. However, none of them were able to perfectly simulate the distribution of dose rates observed after the accident over eastern Japan. This was partly due to the error of the wind vectors and precipitations used in the numerical simulations. Unfortunately, their deterministic simulations could not deal with the probability distribution of the simulation errors.

Therefore, an ensemble simulation of the atmospheric radionuclides was performed using the ensemble Kalman filter (EnKF) data assimilation system coupled with the Japan Meteorological Agency (JMA) non-hydrostatic mesoscale model (NHM). The JMA-NHM has been used operationally for weather forecasts by JMA. Through this ensemble data assimilation, twenty members of the meteorological analysis over eastern Japan from 11 to 31 March 2011 were successfully obtained. Using this meteorological analysis, the radionuclide behavior in the atmosphere such as advection, convection, diffusion, dry deposition, and wet deposition was simulated. This ensemble simulation provided the multiple results of the radionuclide dispersion and distribution. Because the large (small) ensemble-spread of the multiple results indicates the low (high) accuracy of the numerical simulation, the probability distribution of the simulation errors is obtainable from the ensemble simulation. These statistics can provide information useful for the probabilistic prediction of atmospheric radionuclides.

Numerical simulations are able to collaborate with field observations in depicting the full picture of the radionuclide contamination in Fukushima.

## Imaging the reactivity and transport of <sup>99</sup>Tc through Fe-cement/rock barriers

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Geological disposal of radioactive waste requires a detailed understanding of the critical processes affecting geochemical transformations at laboratory and field scale. The interfaces between storage containers, repository and geological substrata can be viewed as a series of continuous barriers with varying porosity and chemical composition that governs flow and physical-chemical transformation of radionuclides during their long term transport. Using this approach, we have investigated the mobility of <sup>99</sup>TcO<sub>4</sub><sup>-</sup> transport through Fe-cement/sandstone engineered barriers using flow through experiment and gamma imaging of <sup>99m</sup>Tc isotope. A flow cell packed with nirex vault reference backfill material (NRVB) containing zero valent iron (ZVI) or magnetite and Sherwood sandstone in a barrier fashion was used. The transport of <sup>99m</sup>Tc in synthetic groundwater at pH 10 was imaged using a medical gamma camera at a flow rate of 5.7ml h<sup>-1</sup>. The Fe-bearing material was prepared to obtain a uniform distribution of 20% ZVI or 10% magnetite representative of steel materials that may create reactive hotspots in the disposal site. The results showed that the unmodified NRBV/sandstone systems have not capacity to retain <sup>99m</sup>Tc. In contrast, <sup>99m</sup>Tc was not transported through the NRBV containing ZVI and it was retained entirely by the modified barrier. The NRBV/sandstone system modified with magnetite showed retardation of <sup>99m</sup>Tc that was overcome over long periods of time, allowing the radionuclide to be eventually transported out of the flow cell. The results showed that <sup>99</sup>Tc transport is conservative through cement and sandstone. The implication is that this radionuclide will be transported to the outside natural environment surrounding the geological disposal. However, amendment with Fe-bearing materials, especially ZVI increases confinement of the radionuclide within the geological disposal reducing the risk of long term environmental contamination.