

Impacts of anthropogenic source from the nuclear fuel reprocessing plants on global atmospheric iodine-129 cycle: A model analysis

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ABSTRACT

The long-lived radioactive iodine (^{129}I) is a useful geochemical tracer of radioactivity in the atmospheric environment. Although the behavior of ^{129}I in the atmosphere is still not fully known because of lack of continuous monitoring, we recently observed clear seasonal trends in air concentration and deposition of ^{129}I in Japan. Using these data, we developed a global atmospheric ^{129}I transport model to reveal key processes for the global atmospheric ^{129}I cycle. The physical and chemical processes of advection, turbulent diffusion, dry and wet deposition, atmospheric photolysis, gas–particle conversion in the atmosphere, anthropogenic sources of ^{129}I discharged from operating nuclear fuel reprocessing plants, and natural sources of ^{129}I volatilized from ocean and land were included into the model. The model generally reproduced the observed seasonal change in monthly air concentration and deposition of ^{129}I in Japan, and the global distribution of ^{129}I concentration in rain as presented in past literature. Numerical experiments changing the intensity of anthropogenic and natural sources were conducted to quantify the impact of anthropogenic sources on the global ^{129}I cycle. The results indicated that the atmospheric ^{129}I from the anthropogenic sources was readily deposited in winter and can be accumulated mainly in the northern part of Eurasia. In contrast, the atmospheric ^{129}I from the natural sources dominated the deposition in summer. These results suggested that the re-emission process of ^{129}I from the Earth's surface may be important as a secondary impact of ^{129}I in the global-scaled environment. Furthermore, although wet deposition dominated the total deposition in the Northern hemisphere, dry deposition regionally and seasonally contributed to the total deposition over arctic and northern part of Eurasia in winter, suggesting that the dry deposition may play a key role in the seasonal change of ^{129}I deposition in the high latitudes of the Northern hemisphere.

1. Introduction

Iodine-129 (^{129}I), which has a long half-life of 1.57×10^7 years, has been known as a useful geochemical tracer both in marine environments and in the atmosphere (Fehn et al., 2003; Moran et al., 1995; Nimz et al., 1998; Michel et al., 2012). ^{129}I is produced naturally via the interaction of atmospheric xenon with cosmic rays or the spontaneous fission of uranium-238 and uranium-235 in the lithosphere (Eissenbud and Gesell, 1997). The natural inventory of ^{129}I has been estimated to be approximately 1.5 TBq (Rao and Fehn, 1999). Meanwhile, ^{129}I has also been known as a major radionuclide byproduct of human nuclear activities since the 1940s. Due to its long half-life and continuous release from ongoing nuclear energy production, ^{129}I is perpetually

accumulating in the environment.

Anthropogenic ^{129}I has been released into the environment from nuclear weapons testing, nuclear accidents, and activities at nuclear fuel reprocessing plants and is therefore distributed unevenly across the globe. Nuclear weapons testing, mainly conducted in the 1960s, dispersed ^{129}I at 0.3–1.0 TBq into the atmosphere (Raisbeck et al., 1995; Wagner et al., 1996) with additional inputs of 40 GBq and 8 GBq during the Chernobyl accident (Gallagher et al., 2005) and the Fukushima Daiichi Nuclear Power Plant disaster (Hou et al., 2013), respectively. The prior and ongoing release from nuclear fuel reprocessing plants is the most important modern contributor to the environmental inventory of ^{129}I . The total amount of ^{129}I released from plants at Sellafield (UK) and La Hague (France) represents more than 90% of the global inventory

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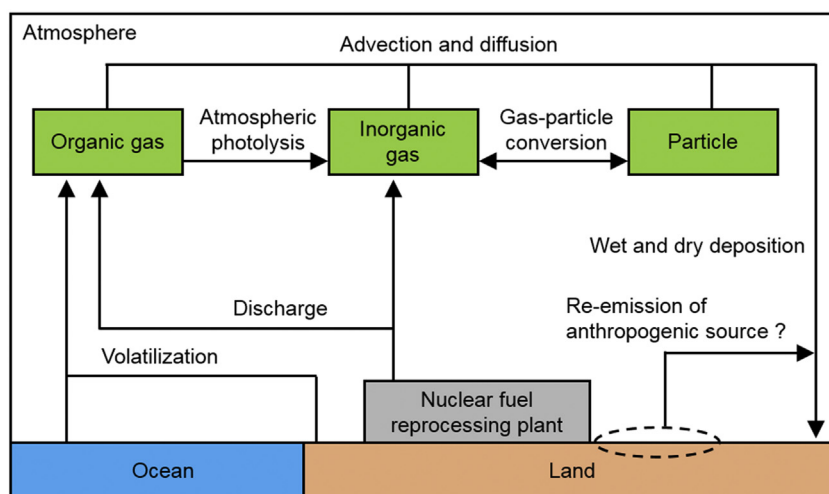


Fig. 1. Schematic illustration of known physical and chemical processes for the global atmospheric ^{129}I cycle. The arrow of the re-emission of anthropogenic sources, shown near the nuclear fuel reprocessing plant is a process newly suggested in the present study.

(Aldahan et al., 2007). Michel et al. (2012) showed that the total atmospheric and marine releases as of 2005 were 0.9 TBq and 9.0 TBq from the Sellafield plant, and 0.45 TBq and 21.8 TBq from the La Hague plant, respectively. The latest annual releases of ^{129}I to the atmosphere in 2015 were reported as 11 GBq/y from the Sellafield plant (Sellafield Ltd., 2016) and 5.8 GBq/y from the La Hague plant (AREVA, 2014, 2016). Reithmeier et al. (2010) estimated that the annual atmospheric release is approximately 10 GBq/y from another currently operating nuclear fuel reprocessing plant at Mayak in Russia. In addition, as a result of environmental releases from these plants, secondary emission of ^{129}I from oceans and land has also become an important source of atmospheric ^{129}I (Persson et al., 2007; Keogh et al., 2010).

The source of airborne ^{129}I has been a topic of interest for the study of global ^{129}I cycling in the atmosphere (Fig. 1), and most relevant studies have been conducted with direct measurements. In Europe, Krupp and Aumann (1999) reported that ^{129}I concentrations in Germany were controlled by volatilization over the North Atlantic Ocean. Gómez-Guzmán et al. (2012) suggested that air concentrations of ^{129}I in southern Spain were mainly affected by meridional winds. Zhang et al. (2016) reported that ^{129}I measured in atmospheric aerosols in Denmark could be attributed to secondary emissions of ^{129}I that had been discharged into the ocean, rather than direct gaseous release from the La Hague and Sellafield facilities. Although measurements are spatially and temporally limited compared to those made in Europe, the ^{129}I cycle has also been investigated in North America and Asia. Toyama et al. (2012) showed that atmospheric ^{129}I measured at several sites in Japan was mainly attributable to volatilization from open oceans and/or released from nuclear fuel reprocessing plants in Japan and Europe. However, the literature is sparse in terms of investigation of the seasonal quantitative contributions of those sources.

To investigate the behavior of atmospheric ^{129}I , numerical models have also been utilized. Reithmeier et al. (2010) simulated the climatological deposition pattern of ^{129}I using a box model, in which advection due to zonal wind, turbulent diffusion, dry and wet deposition, and anthropogenic and natural emissions were included. The regional transport pathway of airborne ^{129}I has been calculated based on trajectory analysis with the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) model (Gómez-Guzmán et al., 2012b, 2017). However, these models do not include chemical forms of iodine. The behavior of atmospheric iodine is also affected by its chemical form, such as elemental iodine, organic and inorganic iodide compounds, and iodide compounds adsorbed onto particles. The concentration varies with location, season, and climate as a result of the processes of advection and diffusion, atmospheric deposition, and atmospheric

chemistry (Hou et al., 2009). The organic gas of ^{129}I is transported in the atmosphere, and then, the inorganic gas is produced from the organic gas by atmospheric iodine photolysis. The inorganic iodine species is known to exist as particulate form (Saiz-Lopez et al., 2012; Jabbar et al., 2013).

Lately, we reported the long-term monthly continuous measurement of gaseous and particulate ^{129}I concentrations and deposition from 2006 to 2015 at Rokkasho in northern Japan (Hasegawa et al., 2017). Both gaseous and particulate ^{129}I concentrations showed an interesting seasonal variation—high and low during the winter and summer, respectively. This tendency is opposite from that observed in Canada and the United States, that is, maximum and minimum ^{129}I concentration in rain in summer and winter, respectively (Herod et al., 2015). Although the result can be related to the transport of ^{129}I from the European nuclear fuel reprocessing plants to the United States (Moran et al., 1999), major processes and emission sources determining global atmospheric ^{129}I cycle are still unclear.

In this study, we aimed to reveal the impact of anthropogenic emission sources on the global atmospheric ^{129}I cycle using our atmospheric dispersion model. We develop an atmospheric ^{129}I transport model that includes physical and chemical processes of ^{129}I with anthropogenic and natural emissions. We evaluate the calculation result by comparing with our monthly continuous ^{129}I data in Japan and literature data of ^{129}I concentration in rain in the Northern Hemisphere. On the basis of sensitivity analyses with changing modeled anthropogenic and natural source intensity, we investigate the role played by these factors in the global atmospheric ^{129}I cycle.

2. Model description

The newly developed atmospheric ^{129}I transport model is designed as an offline tracer model. It requires meteorological variables to calculate the transport of ^{129}I in the atmosphere, which are simulated by a meteorological model.

2.1. Meteorological model

To obtain variables that drive the atmospheric iodine-129 transport model, we use the Advanced Research Weather Research and Forecasting (WRF version 3.6.1) model, which is a non-hydrostatic, fully compressible model for mesoscale meteorological predictions developed by the National Center for Atmospheric Research (Skamarock et al., 2008). The WRF has been extended with a global version (Richardson et al., 2008; Zhang et al., 2012), allowing for global

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