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Impacts of C-uptake by plants on the spatial distribution of ^{14}C accumulated in vegetation around a nuclear facility—Application of a sophisticated land surface ^{14}C model to the Rokkasho reprocessing plant, Japan

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ABSTRACT

The impacts of carbon uptake by plants on the spatial distribution of radiocarbon (^{14}C) accumulated in vegetation around a nuclear facility were investigated by numerical simulations using a sophisticated land surface ^{14}C model (SOLVEG-II). In the simulation, SOLVEG-II was combined with a mesoscale meteorological model and an atmospheric dispersion model. The model combination was applied to simulate the transfer of $^{14}CO_2$ and to assess the radiological impact of ^{14}C accumulation in rice grains during test operations of the Rokkasho reprocessing plant (RRP), Japan, in 2007. The calculated ^{14}C-specific activities in rice grains agreed with the observed activities in paddy fields around the RRP within a factor of four. The annual effective dose delivered from ^{14}C in the rice grain was estimated to be less than 0.7 μSv, only 0.07% of the annual effective dose limit of 1 mSv for the public. Numerical experiments of hypothetical continuous atmospheric $^{14}CO_2$ release from the RRP showed that the ^{14}C-specific activities of rice plants at harvest differed from the annual mean activities in the air. The difference was attributed to seasonal variations in the atmospheric $^{14}CO_2$ concentration and the growth of the rice plant. Accumulation of ^{14}C in the rice plant significantly increased when $^{14}CO_2$ releases were limited during daytime hours, compared with the results observed during the nighttime. These results indicated that plant growth stages and diurnal photosynthesis should be considered in predictions of the ingestion dose of ^{14}C for long-term chronic releases and short-term diurnal releases of $^{14}CO_2$, respectively.

1. Introduction

The Rokkasho reprocessing plant (RRP) of the Japan Nuclear Fuel Limited (JNFL), located in the northern part of Japan, is a nuclear fuel reprocessing plant that handles a large amount of spent nuclear fuel. The JNFL has been undergoing test operations since 2006 using actual spent nuclear fuels (Aomori Prefecture, 2008; Akata et al., 2011, 2013) and is planning to start commercial operations at the RRP in the near future. Therefore, predictions of the radiological impacts on the public from operations at the RRP are highly essential (Akata et al., 2013; Tani et al., 2013).

Radiocarbon (^{14}C) is a key radionuclide when considering the radiological impacts of a reprocessing plant because the dose from ^{14}C comprises a large proportion of the total dose originating from radionuclides released during operations (UNSCEAR, 2000; Shinohara, 2004a, 2004b). A large fraction of ^{14}C released from a reprocessing plant into the atmosphere is transported as a gaseous form of $^{14}CO_2$ (Fontugne et al., 2004; Shinohara, 2004b; Koarashi et al., 2005) and deposits to land when radioactive plumes pass through. In this situation, the uptake of $^{14}CO_2$ by plants during photosynthesis (foliar $^{14}CO_2$ uptake) is a major ^{14}C transfer mechanism from the atmosphere to the land (Levin et al., 1988; McCartney et al., 1988; Otlet et al., 1983, 1990). In Japan, rice paddy fields are widely distributed and provide the Japanese staple diet of rice grains. Therefore, an accurate prediction of the accumulation of released ^{14}C in the rice grains cultivated around the RRP is important for determining the effects of the RRP on dose to the public (Aomori Prefecture, 2008).

In general, the dose due to the ingestion of ^{14}C that is routinely released from a nuclear facility has been estimated from the ^{14}C-specific activity in the air, which is based on atmospheric dispersion calculations. This approach assumes that equilibrium conditions

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exist between the ^{14}C-specific activity in the air and in the crop (diet) during the duration of ^{14}C discharge (USNRC, 1977; UNSCEAR, 2000; Shinohara, 2004a, 2004b; Aquilonius and Hallberg, 2005; Roussel-Debet et al., 2006). However, the equilibrium assumption is questionable if the organic ^{12}C accumulation in plant organs is considered to significantly depend on their growth stage (Otlet et al., 1990; Milton et al., 1995; Yamada et al., 2008; Koarashi et al., 2011; Tani et al., 2013). Certainly, ^{14}C-specific activities among the air and some plant organs such as leaves tend to attain equilibrium instantaneously (Milton et al., 1995; Koarashi et al., 2011; Aulagnier et al., 2012), because of the rapid turnover of organic ^{12}C (Koarashi et al., 2011). In contrast, the ^{14}C-specific activity of fruits and grains is determined by the $^{14}CO_2$ concentration in the air during a specific period when ^{12}C accumulation in these organs occurs (Otlet et al., 1990; Koarashi et al., 2008, 2011). For example, the ^{14}C-specific activity of rice grains is dominantly determined by the $^{14}CO_2$ concentration in the air during the period from heading to maturity (Koarashi et al., 2011). Clearly, the accumulation of C in the target organ at each growth stage should be accounted for predictions of the ^{14}C-specific activity of plants.

Furthermore, diurnal variations in the foliar $^{12}CO_2$ uptake affect the ^{14}C-specific activity of the plant. The ^{14}C-specific activity of vegetation is known to be affected by the $^{14}CO_2$ concentration in the air, particularly during daylight hours (Levin et al., 1988; Collins and Gravett, 1995; Milton et al., 1995). This diurnal pattern of ^{14}C accumulation is caused by the fact that atmospheric $^{14}CO_2$ is absorbed by foliage only during the stomatal opening and the photosynthesis that occur under light (Guenot and Belot, 1984; Ota et al., 2012; Aulagnier et al., 2013). Overall, the atmospheric dispersion of the released $^{14}CO_2$, the growth stage of the target vegetation, and diurnal variations in photosynthesis must be considered to estimate the specific activity of ^{14}C in vegetation affected by $^{14}CO_2$ releases from a nuclear facility.

However, current atmospheric dispersion models which are used in dose estimations do not include such land-surface $^{14}CO_2$ transfer mechanisms to calculate the ^{14}C-specific activity in vegetation. Until recently, the atmospheric dispersion of $^{14}CO_2$ around nuclear facilities has been studied by using Gaussian dispersion models (Levin et al., 1988; McCartney et al., 1988; Shinohara, 2004a, 2004b; Dias et al., 2009; Koarashi et al., 2008, 2011, 2015) or by a more sophisticated dynamic Lagrangian (particle tracing) model coupled with a meteorological model (Akata et al., 2013). However, the transfer of atmospheric $^{14}CO_2$ to the underlying vegetative ecosystem has not been included in these models. The lack of land-surface ^{14}C transfer predictions in these atmospheric dispersion models is mainly due to the difficulty in the ability to calculate $^{14}CO_2$ dynamics in a vegetated ecosystem. These dynamics involve physical, physiological, and microbiological processes such as the transport of $^{14}CO_2$ in the atmosphere and soil, foliar $^{14}CO_2$ uptake via stomata, root uptake of $^{14}CO_2$ in the soil, and accumulation of organic ^{14}C to each plant organ, all of which vary dynamically in time (Amiro et al., 1991; Evenden et al., 1998; Galeriu and Melintescu, 2014).

On the other hand, many land surface ^{14}C models that include these $^{14}CO_2$ transport processes have been developed (Aquilonius and Hallberg, 2005; Melintescu and Galeriu, 2005; Keum et al., 2008; Takahashi et al., 2011; Tani et al., 2011, 2013; Aulagnier et al., 2012, 2013; Le Dizès et al., 2012; Ota et al., 2012; Melintescu et al., 2013; Galeriu and Melintescu, 2014). For example, the POM^{14}C model (Aquilonius and Hallberg, 2005) predicts the annual ^{14}C transfer from the air to plants by using values of the $^{14}CO_2$ concentration in the air and plant photosynthesis that are averaged over the growing season. There are more sophisticated ^{14}C models, such as TOCATTA-χ (Aulagnier et al., 2013), PLANT-W (Melintescu and Galeriu, 2005; Melintescu et al., 2013), and SOLVEG-II (Ota et al., 2012), that calculate foliar $^{14}CO_2$ uptake over short timescales (e.g., <1 h) based on a realistic calculation of ^{12}C and ^{14}C transport in vegetated land surface. Coupling a sophisticated land surface ^{14}C model with a Lagrangian dispersion model should be effective for simulating the dynamically varying atmospheric dispersion and foliar uptake of the discharged $^{14}CO_2$.

The present study investigated the impacts of plant C uptake on the spatial distribution of ^{14}C accumulated in vegetation around a nuclear facility. For this purpose, we performed numerical simulations using a sophisticated land surface ^{14}C model (SOLVEG-II) coupled with a meteorological model (MM5) and a Lagrangian atmospheric dispersion model (GEARN). The coupled model was applied to the local area around the RRP to establish the dispersion of $^{14}CO_2$ and accumulation of ^{14}C in rice plant during test operation of the RRP in 2007, and to estimate dose delivered from the ^{14}C accumulated in the rice grain. Then, the impacts of plant growth stages and diurnal foliar CO_2 uptake on the spatial distribution of released ^{14}C accumulated in rice plants were investigated based on numerical experiments using different hypothetical $^{14}CO_2$ release patterns from the RRP.

2. Materials and methods

2.1. Models

We performed numerical simulations that used SOLVEG-II under the meteorological and $^{14}CO_2$ concentration conditions calculated by MM5 and GEARN, respectively. First, the meteorological and atmospheric $^{14}CO_2$ concentration fields around the RRP were simulated by MM5 and GEARN with observed $^{14}CO_2$ release rates from the RRP. Then, SOLVEG-II was applied to each horizontal grid of the domain using the outputs of the meteorological and $^{14}CO_2$ dispersion calculations to calculate the $^{14}CO_2$ transfer from the atmosphere to the underlying vegetative ecosystem.

The MM5 model is a non-hydrostatic fully compressible model for mesoscale meteorological predictions (Grell et al., 1994). This model predicts three-dimensional fields of wind, precipitation, and diffusion coefficients based on atmospheric dynamic equations at an appropriate spatial and temporal resolution by using nested domains.

The Lagrangian dispersion model GEARN calculates the advection, diffusion, and dry and wet deposition of airborne radioactivity onto the land by tracing the three-dimensional movement of many marker particles following time-dependent meteorological fields calculated by MM5 (Terada and Chino, 2008; Terada et al., 2013; Katata et al., 2015). Compared with simple Gaussian dispersion model, this kind of Lagrangian model has advantages in predicting plume dispersion under atmospheric conditions with vertical sheer or temporal change in wind field. The performance of the MM5-GEARN-coupled simulation to predict atmospheric transport has been validated for various release events, such as the European tracer experiment (ETEX) (Furuno et al., 2004), ^{137}Cs dispersion and deposition at Chernobyl (Terada and Chino, 2005, 2008) and following the Fukushima accident (Katata et al., 2015), and the dispersion of ^{85}Kr released from past test operations of the RRP (Terada et al., 2013). Model-prediction of the MM5-GEARN coupled simulation has also been validated for local scale (~10 km) dispersion and deposition of the Fukushima-derived ^{137}Cs over valley area near the plant (Katata et al., 2012).

The land surface model SOLVEG-II is a one-dimensional multilayer atmosphere-soil-vegetation model that consists of four main modules (atmosphere, vegetation, soil, and radiation) (Yamazawa, 2001; Nagai, 2002, 2003, 2005), and calculates the transport of heat, water, and $^{12}CO_2$ in a vegetative land. The model is driven by meteorological input data under the upper atmospheric boundary condition. Modeled processes for the exchanges of heat, water,