Prediction of the long-term atmospheric concentration of Cs-137 released by the Fukushima accident

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Abstract

In the present paper, we explain how to predict the long-term atmospheric concentration of Cs-137 released by the Fukushima accident in the 200 km vicinity of FD1NPP. The method is a modification of our study in 1997 (Hatano and Hatano). In the present study, we have two new results. First, we use the fractional difference calculus in order to obtain the long-term formula of the Cs-137 concentration. Second, we include the double-exponential term to reproduce the rapid decrease of the aerosol concentration at the very early stage after the emission. The formula is compared with the post-accident data in Tsukuba, it agrees with the data very well.

The concentration of radionuclides in the air is important because it imposes an inhalation risk, that will result in lung cancer. The radiation risk due to inner exposure is generally much larger than those from external exposure, precise estimates and predictions of the aerosol concentration is of social concern. We have been proposed a model for the long-term (such as a decade) atmospheric activity concentration based on the Chernobyl data. In the following, we derive the mean concentration of radioactive aerosols $\langle C\rangle$ with the consideration of the decrease at an early stage after the emission. The halfway of the derivation is similar to Hatano and Hatano (1997), hence we briefly explain the derivation of the formula of $\langle C\rangle$.

Let us start with the following assumption on the concentration C:

$$C(x,y,z,t) \simeq C(x,y,t) \times const., \tag{1}$$

where t is the elapsed day after the accident and the origin of the coordinate is set to the hypocenter. Gavrilov *et al.* measured the height-dependence of the atmospheric concentration of radionuclides and found that the annual average of the concentration is rather stationary with regard to the height from the ground. The purpose of the present model is the long-term behavior, we adopt their findings. The concentration is usually measured at a fixed height (1.5m), hence we model the concentration $C(x,y,t)$ at that height.

Setting the velocity components $v_1(t)$ and $v_2(t)$ for x- and y directions, respectively, where differentiation with regard to $x = x_1, y = x_2$ is written as $\frac{\partial}{\partial x_i}$, we have

$$\frac{\partial C(x,y,t)}{\partial t} + v_i(t)\frac{\partial C(x,y,t)}{\partial x_i} + \lambda_{env}(t)C(x,y,t) + \lambda_{dec}C(x,y,t) = 0. \tag{2}$$

The initial condition is $C(x,y,0) = \delta(x)\delta(y)$, that means an instant emission at $t = 0$ at the origin, the boundary condition is $C(\pm\infty, \pm\infty, t) = 0$.

Here λ_{dec} is the the sum of all the first-order decrease of a specific radionuclide in the atmosphere. The wind velocity v is not the actually measured ones, but is the effective velocity. The radionuclide may have been deposited on the ground but after a strong wind, it is lifted into the air (resuspended state). The average time resting on the ground t_{gnd} and that being uplifted in the air t_{air} makes v_i the effective convection velocity such as $v_i(t) = V_i\frac{t_{air}}{t_{gnd}+t_{air}}$, where V_i is the measured wind velocity. The term $\frac{t_{air}}{t_{gnd}+t_{air}}$ is much smaller than unity (typicaly, by the order of 4 to 6 depending on the elapsed time after emission). We assume that v_i is depend only on t, the wind field is postulated homogeneous in the area we are considering. Since the concentration C is the result of the resuspension-deposition cycle of nuclides, the removal processes, that remove nuclides from the cycle, are considered in both λ_{dec} and $\lambda_{env}(t)$. For example, the radioactive decay rate of a specific nuclide λ_{rad} can be included in λ_{rad}. Also, the soil-infiltration rate λ_{inf} and the runoff rate λ_{runoff} can be considered

as $\lambda_{dec} = \lambda_{rad} + \lambda_{inf} + \lambda_{runoff} + ...$ In this way, all the removal processes can be considered by just changing the value of λ_{dec} as far as they are treated as the first-order kinetics.

On the other hand, in the present study, we newly include a time-dependent removal mechanisms ($\lambda_{env}(t)$). This is the difference from our previous work. For example, the vegetation uptake rate is time dependent. Specifically, the rate of vegetation uptake is observed to decrease as the inverse of time: $\frac{A}{t}$, where A is a constant, determining the magnitude of the uptake, it is determined through the fitting of the data. We slightly modify the equation to

$$\frac{A}{1+t}, \tag{3}$$

in order to eliminate the divergence to the infinity at $t \to 0$. Since the model is intended to adopt a long-term data, the difference of 1 day (from t days to $t+1$ days) should be negligible.

We also add a new term to $\lambda_{env}(t)$ in order to reproduce the rapid decrease right after the emissions. The decrease rate itself depends on time as $Be^{-\beta t}$, where B and β are positive constants. In this case, the term $Be^{-\beta t}$ rapidly decrease to 0, when the initial unknown factors, that make the Cs-137 concentration high, become negligible. In this way, the time-dependent environmental-decay term yields

$$\lambda_{env}(t) = \frac{A}{1+t} + Be^{-\beta t}. \tag{4}$$

Incidentally, Hirose et al. measured the deposition of radionuclides in the city of Tsukuba and analyzed the data with the record of rainfalls. They showed that the 90% of the total deposition of Cs-137 in the area has come down with the rainfall. They also showed that the scavenging ratio (the washout ratio) is organized by the rainfall rate (mm/hour). It suggests that the air will be cleaned by every rainfall, hence the activity concentration in the air may depend on the time. We assume that these discrete events can be averaged out and thereby the mean concentration $\langle C \rangle$ can be obtained.

In the following, we solve Eq. (2). We delete the term $\lambda_{env}(t)C(x,y,t)$ and $\lambda_{dec}C(x,y,t)$ of Eq. (2) by introducing a new function $f(x,y,t)$

$$C(x,y,t) = \exp\left[-\lambda_{dec}t - \int_0^t \lambda_{env}(t')dt'\right] f(x,y,t) \tag{5}$$

As the result, Eq. (2) is written as

$$\frac{\partial f}{\partial t} + \frac{\partial (v_i(t) f)}{\partial x_i} = 0, \tag{6}$$

because v_i depends only on t.

We introduce the auto-correlation function in terms of the wind advection $v_i(t)$ during resuspension as

$$\langle v_i(t) v_i(s) \rangle \sim |t-s|^{-\gamma}. \tag{7}$$

The value of γ is set to $2/3$. We assume here that v_i is the probability variables, the bracket means the ensemble average. Hereafter, we do not fix the value of γ and treat γ as a variable.

Since we are interested in the mean value (*i.e.* the expected value) of the Cs-137 concentration, we decompose f into the ensemble average with regard to space $\langle f \rangle$ and the fluctuations $\tilde{f}$:

$$f(x,y,t) = \langle f(x,y,t) \rangle + \tilde{f}(x,y,t). \tag{8}$$

Equation (6) yields the following.

$$\frac{\partial \langle f \rangle}{\partial t} + \frac{\partial \tilde{f}}{\partial t} + v_i \frac{\partial \langle f \rangle}{\partial x_i} + \frac{\partial (v_i \tilde{f})}{\partial x_i} = 0. \tag{9}$$

The fluctuations $\tilde{f}$ and the convection velocity v_i should be zero when we take the ensemble average:

$$\langle \tilde{f} \rangle = 0, \qquad \langle v_i \rangle = 0. \tag{10}$$

Equation (9) yields Eq. (11) when we take account of the ensemble average of the both sides of Eq. (6).

$$\frac{\partial \tilde{f}}{\partial t} + v_i \frac{\partial \tilde{f}}{\partial x_i} = -v_i \frac{\partial \langle f \rangle}{\partial x_i} + \frac{\partial \langle v_i \tilde{f} \rangle}{\partial x_i} \tag{11}$$

This equation is the advection equation regarding $\tilde{f}$, with the source term on the right-hand side.

Let us define the Green's function G of the homogeneous form of Eq. (11):

$$\frac{\partial G(x,x_0;y,y_0;t)}{\partial t} + v_i(t)\frac{\partial G(x,x_0;y,y_0;t)}{\partial x_i} = 0, \tag{12}$$

Using the method of characteristics, we define the characteristic curve as $x = x_0 + \int_0^t v_x(t')dt'$, and $y = y_0 + \int_0^t v_y(t')dt'$. The Green's function is then obtained as

$$G(x,x_0;y,y_0;t) = \delta\left(x - x_0 - \int_0^t v_x(t')dt'\right)\delta\left(y - y_0 - \int_0^t v_y(t')dt'\right). \tag{13}$$

If the emissions are distributed in space, the solution to Eq. (11) is the convolution of the Green's function, then we have

$$\tilde{f}(x,y,t) = \int_0^t ds \int_{-\infty}^{\infty} dx_0 \int_{-\infty}^{\infty} dy_0 G(x,x_0;y,y_0;s)\left(-v_i(s)\frac{\partial\langle f(x_0,y_0,s)\rangle}{\partial x_i} + \frac{\partial\langle v_i(s)\tilde{f}(x_0,y_0,s)\rangle}{\partial x_i}\right). \tag{14}$$

We approximate the first term of the right-hand side of Eq. (14) as

$$\simeq -\int_0^t v_i(s)\frac{\partial\langle f(x_i,s)\rangle}{\partial x_i}ds. \tag{15}$$

Similarly, we approximate the second term of Eq. (14) as

$$\simeq \int_0^t \frac{\partial\left\langle v_i(s)\tilde{f}(x_i,s)\right\rangle}{\partial x_i}ds. \tag{16}$$

Since we need the term $\langle v_i\tilde{f}\rangle$, we multiply v_i to the both sides of Eq. (14), using Eq. (15) and Eq. (16), then take the ensemble average. It yields $\langle v_i\tilde{f}\rangle$ in the following form

$$\left\langle v_i(t)\tilde{f}(x,y,t)\right\rangle \simeq -\left\langle v_i(t)\int_0^t v_i(s)\frac{\partial\langle f(x_i,s)\rangle}{\partial x_i}ds\right\rangle + \left\langle v_i(t)\int_0^t \frac{\partial\langle v_i(s)\tilde{f}(x_i,s)\rangle}{\partial x_i}ds\right\rangle. \tag{17}$$

The second term on the right-hand side is zero, because $\langle v_i\rangle \equiv 0$. Namely,

$$\left\langle v_i(t)\int_0^t \frac{\partial\langle v_i(s)\tilde{f}(x_i,s)\rangle}{\partial x_i}ds\right\rangle = \langle v_i(t)\rangle\int_0^t \frac{\partial\langle v_i(s)\tilde{f}(x_i,s)\rangle}{\partial x_i}ds = 0. \tag{18}$$

Considering Eqs. (7) and (6), we have

$$\frac{\partial\langle f\rangle(x,y,t)}{\partial t} = \frac{\partial^2}{\partial x^2}\int_0^t |t-s|^{-\gamma}\langle f\rangle(x,y,s)ds + \frac{\partial^2}{\partial y^2}\int_0^t |t-s|^{-\gamma}\langle f\rangle(x,y,s)ds \tag{19}$$

This equation is the definition of the fractional derivatives. It may be rewritten as the following.

$$\frac{\partial\langle f\rangle(x,y,t)}{\partial t} = {}_0\mathcal{D}_t^{-(1-\gamma)}\frac{\partial^2}{\partial x^2}\langle f\rangle(x,y,t) + {}_0\mathcal{D}_t^{-(1-\gamma)}\frac{\partial^2}{\partial y^2}\langle f\rangle(x,y,t) \tag{20}$$

We adopt the operational calculus regarding the fractional differential term and the Laplacian. We have the solution at a fixed location as

$$\begin{aligned}\langle C(x)\rangle &\simeq A\exp\left[-\lambda_{dec}t\right]\exp\left[-\int_0^t \lambda_{env}(t')dt'\right] \\ &= A\exp\left[-\lambda_{dec}t\right]\exp\left[-a\ln(1+t)\exp(Be^{-\beta t})\exp(-\lambda' t)\right] \\ &= A\exp\left[-(\lambda_{dec}+\lambda')t + Be^{-\beta t}\right](1+t)^{-\alpha}.\end{aligned} \tag{21}$$

This equation is compared with the Cs-137 atmospheric concentration measured at Tsukuba, Ibaraki. We found the equation agrees the data very well.