Analysis of Time Series of the Ambient Dose Rates

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Radioactive substances are diffused by meteorological effects and soil property or underground water. The movements affect the ambient dose rate slowly and with smoothing in the long term. We detect the phenomena at monitoring posts in Fukushima. The monitoring post measures the flux from radionuclides; therefore, we discuss relations between the flux and mass density. Time-decreasing of the flux is a function of convection-diffusion of carriers of the nuclides. At the same time, it is also a decay function of radionuclides. We discuss product of the two functions, and get an expression for the time series of radioactive contaminated grounds. By using the expressions, we research changes of typical contaminants in Fukushima. This indicates movements of radionuclides.

Keywords: Radioactive cesium, Re-diffusion, Radioactive substance, Ambient dose rate, Forest fire

1 INTRODUCTION

Main radioactive plumes were flown for northwest direction from Fukushima 1 nuclear power plant, when was at March 15, 2011 \cite{1}. Parts of the plumes are deposited on the ground by drizzle, and the radioactive substances are emitting gamma ray yet. Now, main radionuclides are Cesium-134 and 137 (\textsuperscript{134}Cs, \textsuperscript{137}Cs), which have 2 and 30 years of half-life periods \cite{2}.

Radioactive substances are re-diffused slowly after depositions by meteorological effects and properties of soils \cite{3}. The re-diffusion affects human exposure of gamma ray. Re-diffusion rate is small; therefore, to fix it, long-term and high precision measurements are required. Density of radioactive substance is defined by Becquerel per carrier weight \([\text{Bq/kg}]\). On the definition, measurement of the re-diffusion is correct, but sampling intervals, balancing of precision among many operations, are difficult to keep in long time (~5 years).

We develop a measurement method for re-diffusion, which is based on the ambient dose rate of scintillation detector. They can measure in high speed and keep stable equipment about one year. The dose rates can be got during several years under periodic maintenance.

2 DENSITY OF RADIOACTIVE SUBSTANCES AND EMISSION

Nuclear Regulation Authority in Japan (NRA) set Monitoring Posts (MP) for measuring ambient gamma ray dose rate from 2012 until 2014. Observations of each MP are published as a database \cite{4}. The time interval is per 10 min and the precision is 1 [nGy/h]. We believe they are the most reliable data in available environmental radiation data.

The dose rate is the flux, but is not density of mass; thus, the unit \([\text{Gy/h}]\) doesn’t correspond to \([\text{Bq/kg}]\) directly. However, if surrounding conditions are defined, a transformation between \([\text{Bq/kg}]\) to \([\text{Gy/h}]\) is enabled on a simulator \cite{5}. Where, a linear relation between \([\text{Bq/kg}]\) and \([\text{Gy/h}]\) is held. Then, under the restriction of one point, \([\text{Gy/h}]\) can be used instead of \([\text{Bq/kg}]\).

The unit for human exposure in radioactive ray is \([\text{Sv/h}]\). \([\text{Gy/h}]\) is converted to \([\text{Sv/h}]\) by using a coefficient, which is published by IAEA and other organizations \cite{6}. The NRA adopts the coefficient 1.0 \cite{4}. Hereafter, we use \([\text{Sv/h}]\) instead of \([\text{Gy/h}]\).

We define expressions for time series of the ambient dose rate. The expression transforms a vector into several parameters. If they are not fitting, but have physical property, the expression is signified. We can discuss behavior of radionuclides in environment based on the expression.

3 TIME SERIES EXPRESSION OF GAMMA RAY FLUX

Number of radioactive nucleus is decayed per unit time; it is expressed in \([\text{Bq}]\),

\[ M(t) = M(0) \exp(at), 0 \leq t, \quad (1) \]

Where \( M(0) \) is initial mass, and “\( t \)” is the elapsed-time from the origin time. \([\text{Bq}]\) is the number of decay nuclei per unit time, which is not the gravitational mass of nuclei. In discussions of radioactive matters, we regard as \([\text{Bq}]\) and the mass.

We define the origin time as 0:00, March 15, 2011, Japan Standard Time (JST). The “\( a \)” is the half-life constant; \( i.e., a = \text{Ln} (1/2)/\text{half-life} \text{ [time]} \). Those of \textsuperscript{134}Cs and \textsuperscript{137}Cs are \(-0.336_1, -0.0230_4\).
from 1990.

Radioactive cesium ions are moved very slowly in the soil. The ground has property radiation, which is about 50 [nSv/h] at east Japan. Snow and the melt-water cover emissions of gamma ray, and decrease the dose rates. We believe the period is from December 1 to April 30. Data in the period are processed as defects.

In equation (7) is, 

\[ \beta = \text{Ln} \left( E(t_{iL}, A, B) / \beta \right) \]  

By using the \( \beta \) and \( M_0 \), a trial curve can be drawn as first order approximation.

\section*{4 PROCESSING OF MEASUREMENTS}

We discuss expressions under actual observations. The conditions are;

(c1) \( M_0 \) and \( \text{obs} \left( t_0 \right) \) are unknown. We can measure dose rate when setting of equipment is completed. The time is the start time (\( t_s \)). In the case of NRA, the “\( t_0 \)” is after 400 [d].

(c2) The measurements include error, which must be decreased.

(c3) The measurements have defect data. There are snow days in east Japan. Snow and the melt-water cover emissions of gamma ray, and decrease the dose rates. We believe the period is from December 1 to April 30. Data in the period are processed as defects.

NRA sampling interval is per 10 min; to suppress noises, we average them per 1 day, and moreover adopt moving average method also. Using a half-window width (\( iw \)), we get,

\[ \text{obs} \left( j, iw \right) = \frac{1}{2iw+1} \sum_{i=0}^{iw} \text{obs} \left( j+iw \right) \]  

The two averages decrease the error until “0.0589/\sqrt{iw}”. We call it suppression rate. The “\( j \)” is discrete suffix that means [d].

It is important to determine \( \text{obs} \left( ts \right) \), because the precision affects the factor “\( F \)” in section 3, which transforms between observations and theory. We use \( iw = 15 \); suppression rate is 0.0152; thus, error is decreased under 2%.

\[ F \ E(\text{ts}, A, B, \beta) = F M_0 E_A r \exp(\alpha_A t_s) + E_B \exp(\alpha_B t_s) \]  

In equation (12), \( F \) and \( M_0 \) are not required at same time, the product, \( F = F M_0 \) is necessary. Information of \( \beta \) before “\( t_s \)”-time is not required. Therefore, we set \( ts = 0 \) in calculations for environmental effects. We get,

\[ \text{obs} \left( ts \right) = F M_0 E_A r \exp(\alpha_A t_s) + E_B \exp(\alpha_B t_s) \]  

Considering \( t = 0 \), \( M_0 \) is determined by observations \( \text{obs} \left( 0 \right) \). The prefix “\( \text{obs} \)” is used for observations. The number of argument of \( \text{obs} \left( t \right) \) is only one, “\( t \)” The physical appearance of \( \text{obs} \left( t \right) \) is the ambient dose rate in [Gy/h]. Where, there is no information on radionuclides. To connect \( \text{obs} \left( t \right) \) to \( E \left( t, A, B, \beta \right) \), a factor (“\( F \)” is introduced. The “\( F \)” is different at each observation point, whose function is to condense environmental fields into a scalar variable. It doesn’t depend on “\( t \)” but is a constant.

\[ \text{obs} \left( t \right) = F E \left( t, A, B, \beta \right). \]  

At \( t = 0 \), equation (7) is,

\[ E(0, A, B, \beta) = M_0 E_A r + E_B = \text{obs} \left( 0 \right) /F, \]

In an interval \([0, t_f] \), the “\( \beta \)” is determined. The \( t_f \) is last time of observations.

\[ \beta = \text{Ln} \left( E(t_{iL}, A, B) / \beta \right) / w / h_{iL}. \]  

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In equation (12), \( F \) and \( M_0 \) are not required at same time, the product, \( F = F M_0 \) is necessary. Information of \( \beta \) before “\( t_s \)”-time is not required. Therefore, we set \( ts = 0 \) in calculations for environmental effects. We get,
Where, \( \{E_A, r, a_A, ts, E_B, a_B\} \) is known, and \( F \) is calculated at “ts”. Then; we can calculate theoretical expectations (\( expE(t) \)) of the dose rate at any time after “ts.”

\[
expE(t) = -F[E_A r \exp(a_A t) + E_B \exp(a_B t)], \quad ts < t,
\]

(14)

Environmental effects are introduced as follows;

\[
expE(t, \beta, v) = -F[E_A r \exp(a_A t) + E_B \exp(a_B t)] \exp(\beta (t-ts)^2).
\]

(15)

The parameter “\( \beta \)” is determined in an interval \([0, N] \) and \( v = 1 \).

\[
\beta = \ln \frac{\text{obs}E(t_L)/\expE(t)}{h_L},
\]

(16)

Equation (18) doesn’t require the precision of equation (11); we use \( \nu = 1 \), suppression rate 0.9589 (~6%).

\[
R^2(\beta) = 1 - \sum \{H(j) - G(j, \beta)^2\}/\sum \{H(j) - \text{average}(H(j))^2\}.
\]

(19)

\[
dR^2(\beta)/d\beta = 0.
\]

(20)

Equation (20) is solved numerically, where iterative calculations are used. The initial guess of “\( \beta \)” is got from equation (16). Then;

\[
\expE(t, \beta_{\text{opt}}, 1) = -F[E_A r \exp(a_A t) + E_B \exp(a_B t)] \exp(\beta_{\text{opt}} (t-ts)).
\]

(21)

gives reasonable dose rates in almost observation points.

Under the \( \beta_{\text{opt}} \) is fixed, the “\( v \)” of equation (15) is optimized.

\[
G(j, \beta_{\text{opt}}, v) = \ln \{\expE(j, \beta_{\text{opt}}, v)\},
\]

(22)

\[
R^2(\beta_{\text{opt}}, v) = 1 - \{\sum \{H(j) - G(j, \beta_{\text{opt}}, v)^2\}/\sum \{H(j) - \text{average}(H(j))^2\}\},
\]

\[
dR^2(\beta_{\text{opt}}, v)/dv = 0.
\]

(23)

Since the effect of the optimization is small; moreover, equation (15) is an experimental representation; therefore, behind equation of motion is uncertain. We believe that the result should not recur to \( \beta \)-optimization again.

### 5 CHANGE OF GAUSSIAN CONTAMINATION

One dimensional diffusion equation of \( C(x,t) \): \( C \) is,

\[
dC/dt = D \frac{d^2C}{dx^2}, \quad 0<x<1, \ \ D = \text{diffusion const.}
\]

(24)

The general solution is,

\[
C(x,t) = \sum \lambda_n \text{Const}(\lambda_n) * \exp(\lambda_n t), \ \lambda/D = \rho < 0;
\]

\[
|\rho|^{0.5} = n \pi, \ \ n = \{1, 2, 3, \ldots\}.
\]

(25)

The “\( \text{Const}(\lambda_n) \)” is determined boundary and initial condition. Thus; time series of mass diffusion are linear combination of exponentials. Next, we consider the case of two dimension and adding convection. Two dimensional convection-diffusion equation of \( u(x,y,t) \): \( u \) is,

\[
\frac{\partial u}{\partial t} = D \left( \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} \right) - Cx \left( \frac{\partial u}{\partial x} \right) - Cy \left( \frac{\partial u}{\partial y} \right),
\]

\[
\{D, Cx, Cy\} = \text{const}, \ 0 < x < 1, \ 0 < y < 1.
\]

(26)

The differential expression is,

\[
\frac{\partial^2 u}{\partial x^2} = \{u(x+h,y,t) - 2u(x,y,t) + u(x-h,y,t)\}/h^2 + O(h^2),
\]

\[
\frac{\partial^2 u}{\partial y^2} = \{u(x,y+h,t) - 2u(x,y,t) + u(x,y-h,t)\}/h^2 + O(h^2),
\]

\[
\frac{\partial u}{\partial x} = \{u(x+h,y,t) - u(x-h,y,t)\}/2h + O(h^2), \ \ \frac{\partial u}{\partial y} = \{u(x,y+h,t) - u(x,y-h,t)\}/2h + O(h^2).
\]

(27)

The \( h \) is a small displacement. Thus, a forward expression is,

\[
u(x,y,t+dt) \sim D\{u(x+h,y,t) - 2u(x,y,t) + u(x-h,y,t)\}/h^2
\]

\[
+\{u(x,y+h,t) - u(x,y-h,t)\}/h^2 dt - Cx \{u(x+h,y,t) - u(x-h,y,t)\}/2h dt
\]

\[+Cy \{u(x,y+h,t) - u(x,y-h,t)\}/2h dt.
\]

(28)

We calculate equation (28) by using following specifications:

(s1) The mesh and constants are \( N \times N, \)

\[
\{D, Cx, Cy\} = \{1/1, 1/3N, 0\}, \ 0<x<1, \ 0<y<1, \ dh = 1/\text{N}, \ dt = 1/\text{N},
\]

where \( N = 101. \)

(s2) Initial conditions are,

\[
g(x,y) = \text{Norm} \exp[Alp \{(x-cx)^2 + (y-cy)^2\}],
\]

\[Alp = \ln(0.5)/\Delta^2, \ \ A = 3, \ \ (cx, cy) = (0.5, 0.5),\]

the Norm is determined by \( \int [g(x,y) dy] dx = 1 \).

(29)

The contamination shape is a Gaussian, which is a solution of the diffusion equation. The initial contamination is forwarded step wisely. The shape of actual contamination is more complex.

We believe the shape can be expanded by Gaussians, and adopt a Gaussian as first approximation. MPs locate near the Gaussian center, but is not just there. On a little distant point, we detect increased radiation during short period. Because, radionuclides approach the diffusion equation. The initial contamination is forwarded step wisely. The shape of actual contamination is more complex.

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center. The condition is set by considering actual MPs measurements.

This is a map for $xy$-plain, and natural logarithmic intensity of [Bq] is drawn by stepwise coloring. The map is a part of simulation plain whose size is 25 times larger than Figure. The x-axis is numbering per mesh, where one coordinate is $x = 1/100$. The numbering is same for y-axis. Hereafter, the map format is used. Logarithmic intensity of Gaussian is a quadratic function. The center of Gaussian is marked by a red circle.

It is clear diffusion and convection are found at the same time. Distribution seems to be a Gaussian, whose center is moved. A red round point is the center of contamination at $t = 0$. Because, homogeneous convection constant is set. [Bq]-values at the red and green rhombus points are stored. They are named #2 and #3. The time series changes are drawn in Figure 3.

The #2 and #3 points are neighborhood; however, time series are uneven, where the convection effect of equation (28) appeared. It is important information that curvature of the curves is greater than 1.0. If it is greater, radioactive contamination approaches the observation point. On regression of exponential functions, the curvatures of #2 and #3 are,

$$G(j) = 4.20 \times 10^{-2} \exp(-9.38 \times 10^{-1} j^{0.793}), R^2 = 0.998,$$

$$G(j) = 2.65 \times 10^{-2} \exp(-7.78 \times 10^{-4} j^{1.12}), R^2 = 0.998,$$

respectively. These digits are got in case of terminal condition (sp3). If you simulate until density of large steps, another $G(j)$ curve is got. General solution of the diffusion equation is known, whose time-depend-element is a combination of exponentials. The combination gives dent curves. The #3 curve is convex; i.e., the convection gives the curvature. The “$\nu$” of equation (15) is an index about moving of radioactive contamination. The result must be confirmed by observations.

6 ANALYSIS OF OBSERVATION DOSE RATE

The “$\nu$” is a small value around 1; therefore, high precision data are required. We search high dose rate points, which are listed in Table 1. Cumulative error in long-term expression is arisen by movement of contaminations. The movement would be caused by water. We focus on Ukedo and Ohta rivers, whose basins are through high contamination area. We select Ohgaki and Tetsuzan points. Two dams are near the points. We believe radioactive suspended solid is stacked there. Ogaki point is upstream, and Tetsuzan is downstream.

We hear forest fire at Date city, which happens at March 30, 2016. It is continued about 3 days, is extinguished at April 2. Re-diffusion by forest fire at Chernobyl in 2004 is reported \[10\]. The density of $^{137}$Cs+$^{90}$Sr is $10^{-5}$~$10^{-6}$ Bq/m$^3$. Researchers indicate dangerousness of Plutonium dust. The dose rate in Date-city was about 0.25 [$\mu$Sv/h]. This rate is small, and no information about Plutonium is known; however, we should report as the ambient dose rates at Date-city. We add the points, and get Table 2.

Calculated environmental indexes for Table 1 and 2 points are listed in Table 3.

6.1 Existence of point of $\nu>1$

We search points that are “$\exp[\beta(t-ts)], \nu>1$.” The typical example is in Figure 4.
Radioactive substances along river

Convection of radioactive substances along Ukedo river is found, and point #7 gives long half-life (HL), 31.3 [y]. Considering \( \nu = 1.0 \), no radioactive substance is moved in the soil. On the other hand; at #8 point, the HL is 10.2 [y], which is not so long. But, “\( \nu = 1.8 \)” shows much radioactive substances flow toward the detector. Same phenomena are found at #2 and #5 points. There is no river near the 2 points. We believe the increasing fact is a motion of radioactive substances in the soil. We interest in that #7 is upstream and #8 is downstream of dams as a circumstantial evidence. Dose rate curves for point #7 and #8 are in Figure 5 and 6.

Effects of forest fire

The forest fire at Date city continued about 3 days, and litter layers were burned. If the information is true, the soil structure would be changed. Under the rains, radioactive suspended solid will be flow out from the forest. It affects the ambient dose rate. However, dose rate of the forest is 0.25 [\( \mu \text{Sv/h} \)], which is very small compared with points in Table 1. The calculations are listed in Figure 7.

7 CONCLUSION

We propose long-term expressions for time series of the ambient dose rate. They are derived from decay expressions of radionuclides and an exponential part that is derived by simulations of the convection-diffusion equation. The expression is used about 5 years, and the determination coefficient is over 0.97.
parameters having environmental effects.

We apply the expressions to high contaminated area in Fukushima, and detect motion of radioactive substances as a parameter value. The motion is found strongly from upstream to downstream along rivers. It makes the environmental half-life period longer until 30 [y].

We also apply it to effects of after-disturbance of a forest fire; however, no effect is found. The location is contamination under 1/100 of the above mentioned districts; thus, we think the effect would be too small. At least, much radionuclide ash is not thrown out.

We calculate \( \nu \)-values at other MPs in Fukushima, where the values are “1.0.” Long-term re-diffusion of radio cesium is almost diffusion.

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