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1 Title: Stochastic model for the fluctuations of the atmospheric concentration of radionuclides and its
2 application to uncertainty evaluation

3 Hiroyuki Ichige, Shun Fukuchi, Yuko Hatano
4 Graduate School of Systems and Information Engineering, University of Tsukuba

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Telephone number: +8190-1032-7643
Email: tanuki28213141@gmail.com

5 Abstract:
6 We propose a new model of the atmospheric concentration of a radionuclide with the inclusion of
7 fluctuations of the concentration. The model is a stochastic differential equation and we derive the
8 analytic solution of the equation. The solution agrees very well with the Chernobyl Cs-137 data.
9 The advantage of the model is that the uncertainty in radiation exposure risk, with regard to the
10 concentration fluctuations, can be quantitatively estimated. We show the range of fluctuations of ±σ,
11 ±2σ, ±3σ in the 10-year measurement of the atmospheric concentration in Chernobyl and confirmed
12 the validity of the model.

13 1. Introduction

14 In major nuclear power plant accidents, such as Chernobyl or Fukushima, a huge amount of
15 radionuclides have been released into the atmosphere. In such accidents, long-lived radionuclides,
16 cesium-137 and strontium-90, for example, pose a serious problem. Radionuclides carried in the
17 initial plume were deposited on the ground, and they keep imposing a risk to the public health for a
18 long period of time. In the Chernobyl case, it is believed that the resuspension-deposition cycle
19 contributes significantly to the airborne concentration of radionuclides (Klug et al., 1992; Ishikawa,
20 1995; Nicholson, 1998; Ould-Dada and Nasser, 1992). Since the resuspended nuclides make the
21 atmospheric concentration increase, it is considered one of the most important processes in the
22 long-term radiation risk assessment. In this accident, health effects on the humans, such as leukemia
23 and genetic abnormalities have been confirmed (IAEA, 2006; Arkhipov et al., 1994; Lazjukd et al.,

1997; Romanenko et al., 2008). Therefore precise predictions on the concentrations of radionuclides are necessary.

For that purpose, using the data of the Chernobyl accident, we have derived a simple formula on the mean atmospheric concentration (Hatano et al., 1997, 1998). In the formula, the atmospheric concentration of a specific radionuclide decreases as

$$C(t) = A\exp\left(-\lambda_{phys}t\right)t^{-\alpha}. \quad (1)$$

Here $C(t)$ is the concentration of a specific radionuclide measured at a fixed location, t is the number of days since the accident, λ_{phys} is the rate constant which includes all the first-order reactions (*e.g.* radioactive decay, adsorption rate on the soil, see Hatano and Hatano, 1997). A and α are constants that is determined by the fitting of the actual data. The power index $-\alpha$ in Eq. (1) is determined from the magnitude of the temporal autocorrelations of the wind velocity. In our previous studies (Hatano et al., 1997, 1998), we set α at $-\frac{4}{3}$. Equation (1) successfully reproduces the mean concentration of the Chernobyl data (Cs-137-134, Ce-144, Ru-106) over a decade. In present study, we allow more degrees of freedom on α, because the wind correlation may vary depending on sites. In this manner, we introduce more flexibility to the model and thereby make the model applicable in general cases. For the details of our model, see the references. We would stress that Eq. (1) is derived by averaging out all the fluctuations of microscopic processes; therefore it describes the mean behavior of the atmospheric concentration.

In the present study, we concentrate on how the data fluctuates from Eq. (1), and thereby estimate the maximum and the minimum concentrations of the atmospheric concentration. From the Chernobyl case, we have learned that the atmospheric concentration of radionuclides fluctuates very much, depending mainly on the meteorological conditions (the wind velocity, the humidity, rainfall, the amount of solar radiation, and the traffics). Estimating the magnitude of fluctuations would greatly contributes for radiation safety. For this purpose we proposed a model that can reproduce those fluctuations.

2. Model --- stochastic differential equation on the fluctuations

In this section, we propose a new model that can reproduce the fluctuations in the Cs-137 atmospheric concentration. The model is a stochastic differential equation, assuming that each deviation follows a stochastic process. We use here the Chernobyl data set. The measurement site is shown in Fig. 1 based on the report of the Japan Atomic Energy Research Institute (Ueno et al.,

2003). We choose 6 observation sites. Each site is assigned a number, e.g. "Point 20.0". The sites we choose are Point 6.0 (7 km to the southwest from the power plant), Point 8.0 (11 km, north), Point 11.0 (10 km, south), Point 13.0 (9 km, east), Point 21.0 (2 km, northwest), and Point 60.1 (3 km, northwest). These sites have longer observation period than others. As the radionuclide, we choose Cs-137, because Cs-137 was detectable over a decade because of their long half-life (~30years) and amount released was also very large. In Fig. 2, we show a good agreement between Eq. (1) and the data at Point 21.0 for a demonstrating purpose. Other graphs appear in Hatano et al., 1998.

Figure 3 shows the raw data of the atmospheric concentration of Cs-137 at Point 13.0 for 5000 days after the accident. The airborne concentration of Cs-137 fluctuates by various reasons: for example, wind transport from other places, ground surface disturbances such as rainfall, and detachment from surfaces such as trees or buildings. We assume that if we take a long enough period of time, these elementary processes achieve the equilibrium of fluctuations and can be treated as a stationary stochastic process. The repeated randomness is manifest as the resulting averaged behavior. We assume here that these fluctuations of the elementary processes are the Gaussian white noise. The Gaussian white noise is observed frequently in natural phenomena.

We model those fluctuations by means of the stochastic differential equation as follows. First, we define the residual, $X(t)$, between the data and Eq. (1), as

$$\begin{aligned} X(t) &= \ln\left(C(t)/\left(A\exp\left(-\lambda_{phys}t\right)t^{-\alpha}\right)\right) \\ &= \ln\left(C(t)\right) - \ln\left(A\exp\left(-\lambda_{phys}t\right)t^{-\alpha}\right). \end{aligned} \tag{2}$$

In Fig. 4 we see the behavior of $X(t)$ for Point 13.0. When we see $X(t)$ as a stochastic variable, we observe the following two things. One is that the negative values of $X(t)$ and the positive values of that appear almost at the same frequency throughout the measurement. Second, the displacement in $X(t)$ is almost *reversal*. Namely, if $X(t_i) > X(t_{i-1})$, then it is likely we have $X(t_{i+1}) < X(t_i)$ in the next time step, and *vice versa*. Therefore, the model may have the property of a correction force that neutralizes fluctuations:

$$E\left[\frac{dX(t)}{dt}\right] = -\gamma X\,, \tag{3}$$

or

$$E[dX(t)] = -\gamma X\,dt. \tag{4}$$

Here $E[\,]$ represents the expected value and $dX(t)$ is the displacement in X during small amount of time dt. We assume that the sum of $dX(t)$ and $\gamma X\,dt$ is the Gaussian white noise. That is,

$$dX(t) = -\gamma X\,dt + \sigma dW(t). \tag{5}$$

Here $dW(t)$ is the Gaussian white noise at time t, and γ is the parameter of the “reversal” force,

and σ is the parameter indicating the magnitude of the Gaussian white noise. The stochastic differential equation Eq. (5) is called as the Ornstein-Uhlenbeck process. If the Chernobyl data fluctuate as Eq. (5), we should recover the Gaussian white noise $\sigma dW(t)$ by substituting the actual data $dX + \gamma X dt$. In order to show that, we first estimate the values of γ and σ in the next section.

3. Results and discussion

3.1 Estimation of parameters γ and σ

In estimating the values of γ and σ, we demonstrate in Fig. 5 how the value of γ works on $X(t)$. The horizontal axis is $X(t)$ at Point 21.0 over 5000 days, and the vertical axis is its derivative $dX(t)/dt$. The straight line is the LSM (Least Squares Method) fit to the data. According to Eq. (3), the slope in Fig. 5 corresponds to the value $-\gamma$. As shown in this figure, we see a fairly good fit. The actual data have the tendency that an increase in X at a specific time results in a decrease in X at the next moment, and vice versa. However, the LSM fit in Fig. 5 is, in some part, not so good. This is due to the fact that the value of dt (time interval between observations) is sometimes very large in the actual data. In an extreme case, dt is about 30 days. Therefore, in the following, we calculate more accurate value of γ.

Let us start with Eq. (5). Solution of Eq. (5) is given by Susanne and Ove, 2008.

$$X(t) = \exp(-\gamma t)\left[X(t_0) + \sigma \int_0^t \exp(\gamma s)\, dW(s)\right], \tag{6}$$

where $X(t_0)$ is the initial value. Because $X(t)$ is the discrete in the actual data, we rewrite it as $X(t_0)$, $X(t_1)$, $X(t_2)$, $\ldots, X(t_i), \ldots, X(t_n)$. Due to the Markov property of Wiener process, it is possible to take the initial value at anywhere and Eq. (6) can also be expressed as follows:

$$\begin{aligned} &X(t_{i+1}) - \exp\big(-\gamma(t_{i+1} - t_i)\big) X(t_i) \\ &= \sigma \exp\big(-\gamma(t_{i+1} - t_i)\big) \int_0^{t_{i+1}-t_i} \exp(\gamma s)\, dW(s). \end{aligned} \tag{7}$$

If you replace t_i to 0 and $t_{i+1} - t_i$ to t, then Eq. (7) and Eq. (6) are equivalent. Hence, we define the value of the right-hand side of Eq. (7) as Y:

$$Y = \sigma \exp[-\gamma(t_{i+1} - t_i)] \sum_{j=0}^{K} \exp\big(\gamma s_j\big) \big[W\big(s_{j+1}\big) - W(s_j)\big]. \tag{8}$$

Here K is the number of discretization in s. From the definition of the Wiener process, $W\big(s_{j+1}\big) - W\big(s_j\big)$ is a random variable and follows the normal distribution with 0-mean and the variance $s_{j+1} - s_j$. Since the sum of the random variables, which follow the normal distribution, also follows the normal distribution, Y follows the normal distribution. The mean of the sum is

equal to 0, and the variance is follows from simple calculation:

$$V[Y] = \sigma^2 \exp(-2\gamma(t_{i+1} - t_i)) \times E\left[\sum_{j=0}^{K} \exp(2\gamma t_j)\left(W(s_{j+1}) - W(s_j)\right)^2\right]. \quad (9)$$

Here $V[\,]$ represents the variance and we use the relation $V[Y] = E[Y^2] - (E[Y])^2$ for calculation. Using $\left(W(s_{j+1}) - W(s_j)\right)^2 = s_{j+1} - s_j$, we obtained the following equation:

$$V[Y] = \sigma^2 \exp(-2\gamma(t_{i+1} - t_i)) \int_0^{t_{i+1}-t_i} \exp(2\gamma s) ds. \quad (10)$$

By solving this, Y, or the entire right side of Eq. (7) is a random variable that follows the normal distribution with 0-mean and with the variance $\sigma^2(1 - \exp(-2\gamma(t_{i+1} - t_i)))/2\gamma$. Note that variance solely depends on $(t_{i+1} - t_i)$. It means that, when the interval between observations is long, the variance of the fluctuations increases. On the contrary, when the interval is small, the variance of fluctuations decreases. In order to eliminate this dependence on the interval length, we normalize Eq. (7) by dividing the both sides with $1 - \exp\left(-2\gamma(t_{i+1} - t_i)\right)^{\frac{1}{2}}$ to obtain

$$\begin{aligned} &\frac{X(t_{i+1}) - \exp(-\gamma(t_{i+1} - t_i))X(t_i)}{\sqrt{1 - \exp(-2\gamma(t_{i+1} - t_i))}} \\ &= \frac{\sigma \exp(-\gamma(t_{i+1} - t_i)) \int_0^{t_{i+1}-t_i} \exp(2\gamma t)\, dW(s)}{\sqrt{1 - \exp(-2\gamma(t_{i+1} - t_i))}}. \end{aligned} \quad (11)$$

Now the entire right-hand side of Eq. (11) is a random variable that follows the normal distribution with 0-mean and the variance $\sigma^2/2\gamma$. In addition, with respect to the left-hand side, all parameters are known except for γ. Hence, γ is obtained by the following equation.

$$\sum_{i=1}^{n} \frac{X(t_{i+1}) - \exp(-\gamma(t_{i+1} - t_i))X(t_i)}{\sqrt{1 - \exp(-2\gamma(t_{i+1} - t_i))}} \mathrm{sgn} X(t_i) = 0. \quad (12)$$

Here sgn(x) is the signum function defined as follows.

$$sgn(x) = \begin{cases} -1 & (x < 0) \\ 0 & (x = 0) \\ 1 & (x > 1) \end{cases} \quad (13)$$

Then substituting thus-obtained γ to Eq. (11), we finally arrive at the equation for σ.

$$\sigma^2 = 2\gamma \sum_{i=1}^{n} \left(\frac{X(t_{i+1}) - \exp(-\gamma(t_{i+1} - t_i))X(t_i)}{\exp(-2\gamma(t_{i+1} - t_i))} \right)^2. \quad (14)$$

To summarize, we can calculate the parameters γ and σ with Eq. (12) and Eq. (14), using the actual data. We summarize the values of γ and σ of each observation site in Table1. These values

are used in the calculations in the next subsection.

3.2 Analytic Solution

In Section 3.1, we explained that, in order to show that $X(t)$ can be described by the Eq. (5), $dX + \gamma X dt$ should be the Gaussian white noise. This is equivalent to that the right side of Eq. (11) is the Gaussian white noise. We use the spectral analysis because the Gaussian white noise has the same power in its all frequencies.

The result is shown in Fig. 6. The horizontal axis represents the frequency and the vertical axis represents the intensity of the spectrum. Since observation times are unevenly spaced, we used the periodogram (Scargle, 1982; Schulz and Stattegger, 1997). The periodgram is one of the methods of detecting a periodic signal hidden in the noise in the case where the observation times are unevenly spaced. It is used instead of FFT in such cases. Except for a strong annual peak (and its subharmonics), the spectrum scales roughly as the white noise. Figure 6 is the spectrum of Point 21.0 and we confirmed that other five observation sites have the very similar white noise spectrum. In this way, our assumption used in Eq. (5) is justified for the present data set.

In Fig. 7, we plot the cumulative histogram of the right-hand side of Eq. (11) and found that it follows the normal distribution excellently. The curve showing along with the histogram represents the distribution function of the normal distribution. The values of the mean and the variance were obtained in Section 3.1. Figure 7 is the case for Point 21.0, and we found that the results of other five observation sites are almost identical with Fig. 7. As a result of these findings, it can be said that Eq. (5) is suitable for modeling the fluctuations.

Finally we arrive at the analytic solution of radioactive aerosols in the atmosphere. Using Eqs. (2), (5), and (7):

$$C(t) = A\exp\left(-\lambda_{phys}t\right) t^{-\alpha} \times \exp\left\{\exp[-\gamma(t-t_0)]\left[X(t_0) + \sigma\int_0^{t-t_0}\exp\left[\gamma(t-t_0)\right] dW(s)\right]\right\}. \quad (15)$$

By taking the logarithm on both sides of Eq. (11), we have

$$\ln C(t) = \ln\left[A\exp\left(-\lambda_{phys}t\right) t^{-\alpha}\right] + \exp[-\gamma(t-t_0)]\left[X(t_0) + \sigma\int_0^{t-t_0}\exp\left[\gamma(t-t_0)\right] dW(s)\right]. \quad (16)$$

The right-hand side of Eq. (16) is a random variable that follows the normal distribution with the mean $\hat{\mu}$ and the variance $\hat{\sigma}^2$ where

$$\hat{\mu} = \ln\left[A\exp\left(-\lambda_{phys}t\right)t^{-\alpha}\right] + X(t_0)\exp[-\gamma(t-t_0)]. \quad (17)$$

$$\hat{\sigma}^2 = \frac{\sigma^2}{2\gamma}(1 - \exp[-2\gamma(t - t_0)]) \tag{18}$$

Note that, for large t, $\hat{\mu}$ converges to the logarithm of Eq. (1). Thus we obtain the following equation describing the distribution of the fluctuation.

$$p\big(t_0, X(t_0), t, X(t)\big) = \frac{1}{\sqrt{2\pi\hat{\sigma}^2}} e^{-\frac{(X(t)-\hat{\mu})^2}{2\hat{\sigma}^2}}. \tag{19}$$

Here p is the transition probability density function; p is the probability where the residual $X(t)$ occurs when the initial residual is $X(t_0)$. To be precise, p describes the probability density of $X(t)$ under the initial condition:

$$\lim_{t \to t_0} p = \delta\big(X(t) - X(t_0)\big). \tag{20}$$

Note that $X(t)$ corresponds to $\ln C(t)$ (see Eg. (2)). The right-hand side of Eq. (19) is in the form of the normal distribution, $C(t)$ obeys the log-normal distribution.

In Fig. 8, we summarize the significance of the present study. Let us consider the circumstance in which we have to predict the future concentration. In usual case (upper half of Fig. 8), we calculate the average using the observed data that are available at the moment. The future concentration is estimated by extrapolation. However, the estimation depends on the extrapolation technique (upper half right). The ranges of fluctuations are sometimes assumed by the maximum and the minimum values.

In the present study, the fluctuation (the difference from one observation to another) gives information of the future concentration. The value of $\hat{\mu}$, that is asymptotically Eq. (1) as $t \gg 1$, gives the future concentration and $\hat{\sigma}$ the future standard deviation. As described in the previous subsections, Eqs. (12), (14) gives the value of γ and σ, with which we calculate $\hat{\mu}$ and $\hat{\sigma}^2$. In this way, the present method utilizes fluctuations for the behavior of the future concentration.

3.3 Comparison between the analytic solution and the Chernobyl data

Now we collect the results from previous sections and thereby make a comparison with the Chernobyl data. We use Eq. (1) as the mean concentration and $\hat{\sigma}$ as the standard deviation from the mean concentration. Figure 9 shows the comparison of the raw dataof Cs-137 at Point 6.0, 8.0, 11.0, 13.0, 21.0, and 60.1.

Many studies have reported that the concentration of particle in the atmosphere follows a log-normal distribution (e.g. Ashok et al., 1997; Eugene and Chan, 1997; Junya et al., 2002), and this is consistent with the result of the present study.

The thick solid lines indicate the mean concentration (Eq. (1)) and the dotted lines correspond to $\pm\ \hat{\sigma}$, $\pm\ 2\hat{\sigma}$, and $\pm\ 3\hat{\sigma}$. In the log-normal distribution, approximately 68.3% of the data should fall within the range of $\pm\ \hat{\sigma}$, 95.4% in $\pm\ 2\hat{\sigma}$, and 99.7% in $\pm\ 3\hat{\sigma}$. Table 2 gives the percentages for the present study. Our result is consistent with the theoretical percentages.

Finally, we mention the significance of our result from a practical aspect. Understanding the distribution of concentration helps us to make estimations of the risk of radiation exposure of workers. Consider the case as follows. In Fukushima, the working hours of workers are determined using the mean atmospheric concentration. In this case, if the concentration is increased temporarily by fluctuations, workers of some percentage might be exposed to excessive radiation. By using the proposed model, we can calculate a possible maximum risk of workers.

4. Conclusion

We have studied the fluctuations in the atmospheric concentration of Cs-137 in Chernobyl. We found the followings.

1. We proposed a new method to extract the characteristics of fluctuations. We define the fluctuations as the deviations from Eq. (1). Two parameters, γ (the magnitude of reverse effect) and σ (the magnitude of white noise), represent the characteristics. We showed the procedure of calculating γ and σ from the actual data.
2. We derived the analytic solution of the long-term concentration with the inclusion of random fluctuations with γ and σ.
3. The concentration of the Chernobyl data agrees excellently with the solution.
4. Using these results, we can estimate workers' radiation exposure in Fukushima with uncertainty, with which we know the probability that their exposure falls within the range of $\pm\sigma, \pm 2\sigma, \pm 3\sigma$.

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Notation

A	Constant that is proportional to the amount of nuclide that fell in observation point.
C	Radionuclide concentration in the atmosphere.
C_0	Radionuclide concentration that is observed at time of t.
$E[\,]$	Expected value.

p Transition probability density function of X.

t The number of days since accident.

t_0 Measurement date of the first post-accident.

t_i The i-th measurement day.

$V[\,]$ Variance.

W Wiener process.

x Random variable representing the value of X at time t.

x_0 X value at t_0

X Difference in log axis of the observed value and the average concentration.

Y Random variable.

α Constant representing the effect of advection and uptake of plant.

γ Model parameter indicating earliness of fluctuations converge.

$\hat{\mu}$ Mean of X.

λ_{phys} The rate constant which includes all the first-order reactions.

σ Model parameter indicating the magnitude of the fluctuation.

$\hat{\sigma}$ Variance of X.

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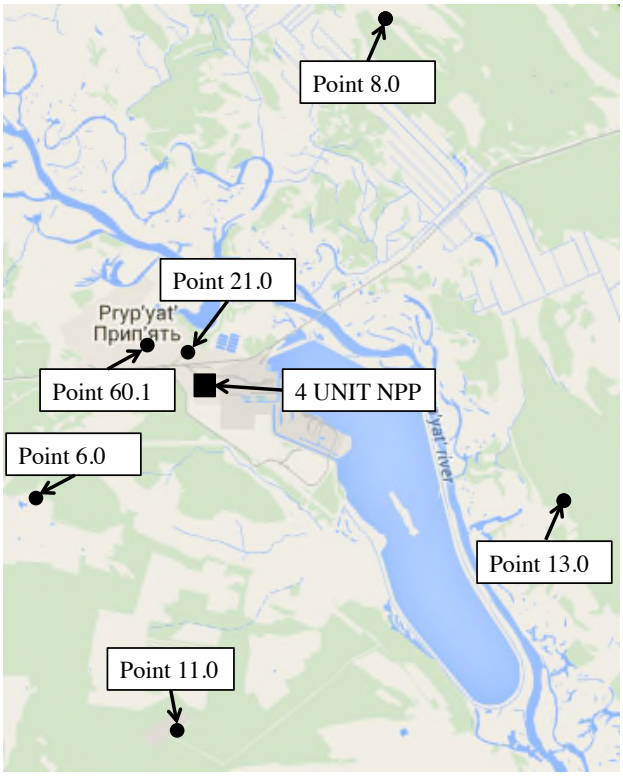

Fig. 1: The location of the Chernobyl power plant and the measurement site. “4 UNIT NPP” indicates the forth unit of the nuclear power plant, where the accident occurred. Annotations on the Google map.

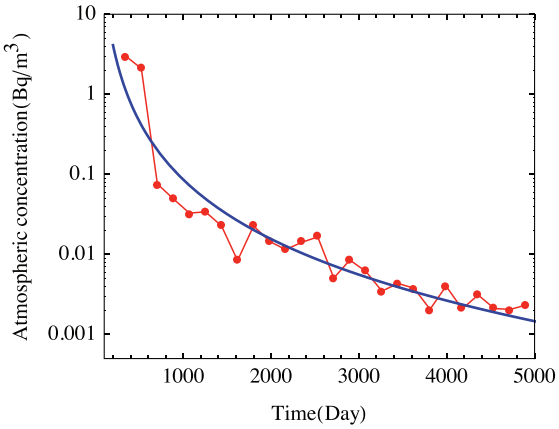

Fig. 2: Atmospheric concentration of 6-months average at Point 21.0.

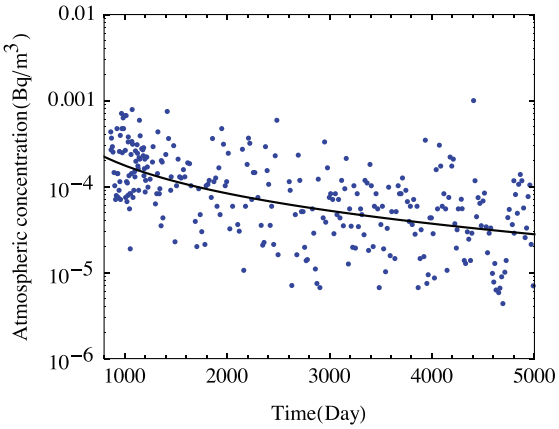

Fig. 3: Atmospheric concentration of Cs-137 at Point 13.0. Dots are the non-averaged original data and the soild line Eq.(1).

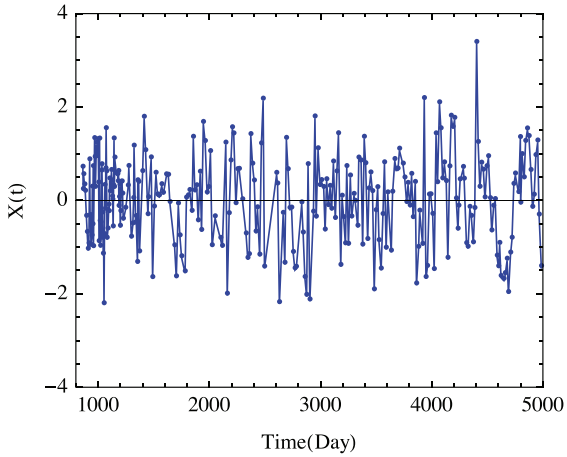

Fig. 4: Deviations X(t) defined as Eq. (2).

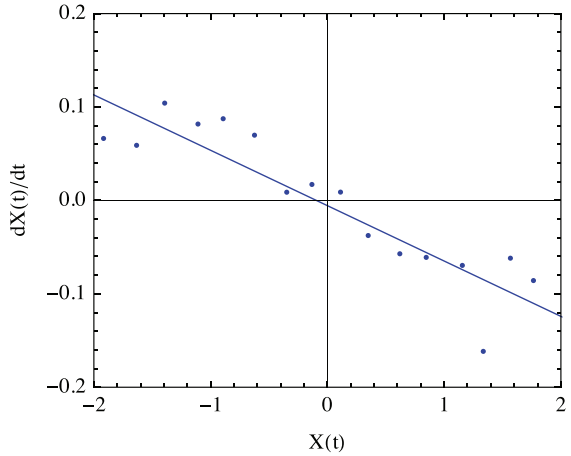

Fig. 5: Relation between X(t) and dX(t)/dt.

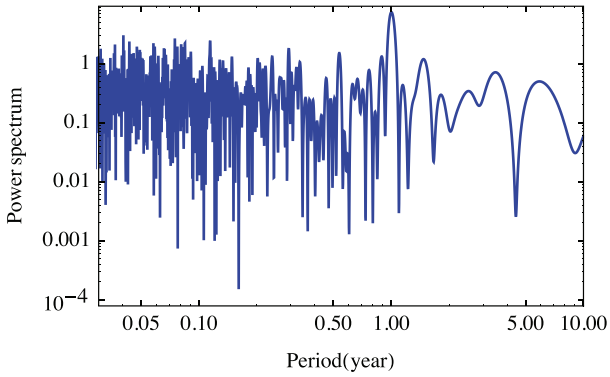

Fig. 6: Power spectrum of the Brownian part $dX + \gamma X dt$. The data of Point 21.0 are used. The spectrum shows the data are white noise.

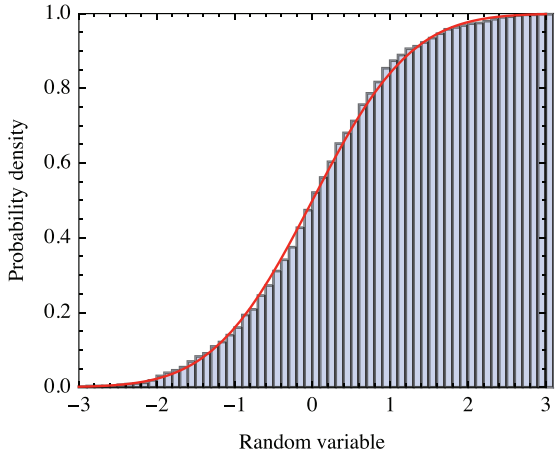

Fig. 7: Histogram of the $dX + \gamma X dt$. It follows the normal distribution. The data is Point 21.0.

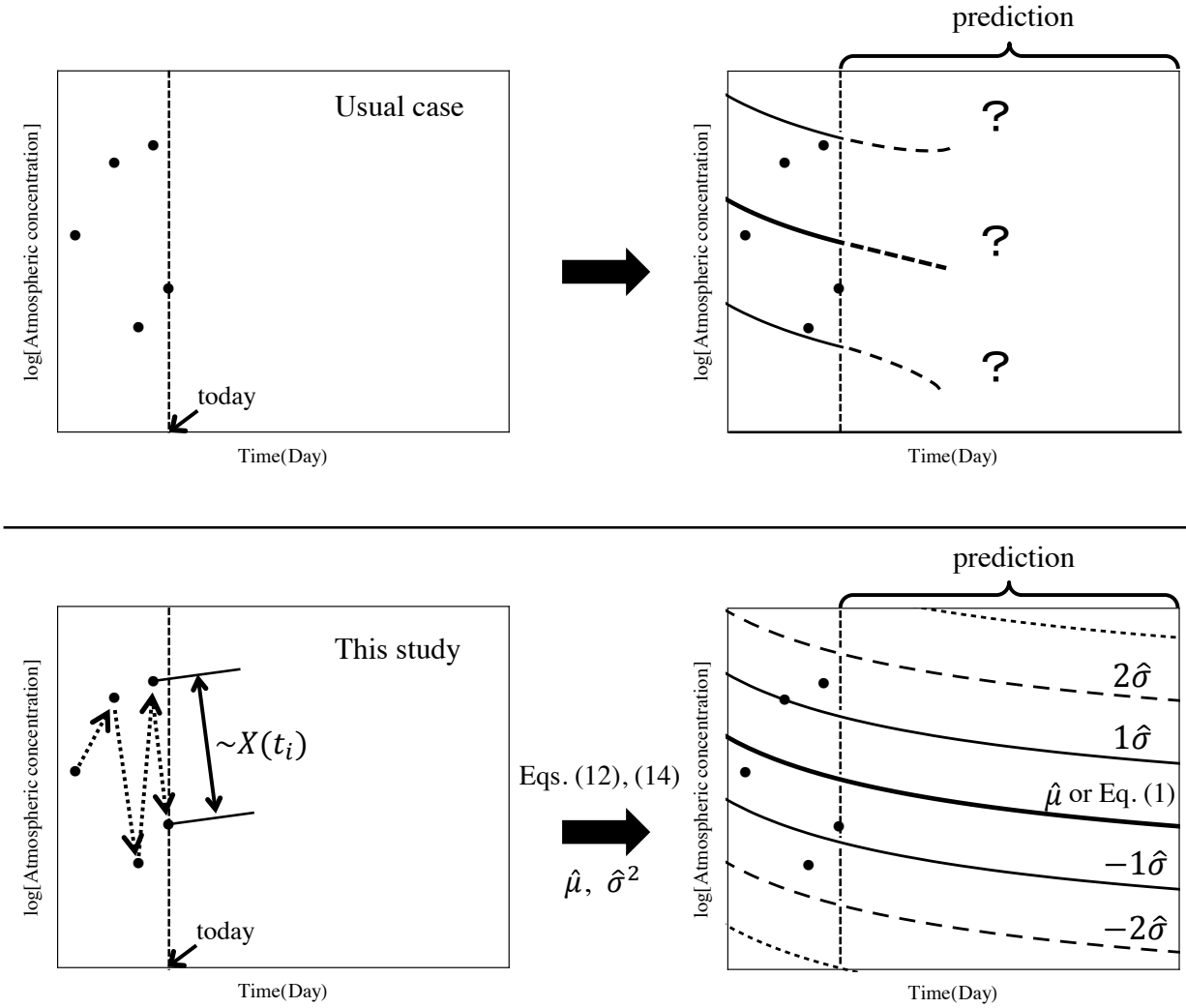

Fig. 8: Illustration of the significance of the present model. (upper half): usual case; (lower half): the present study.

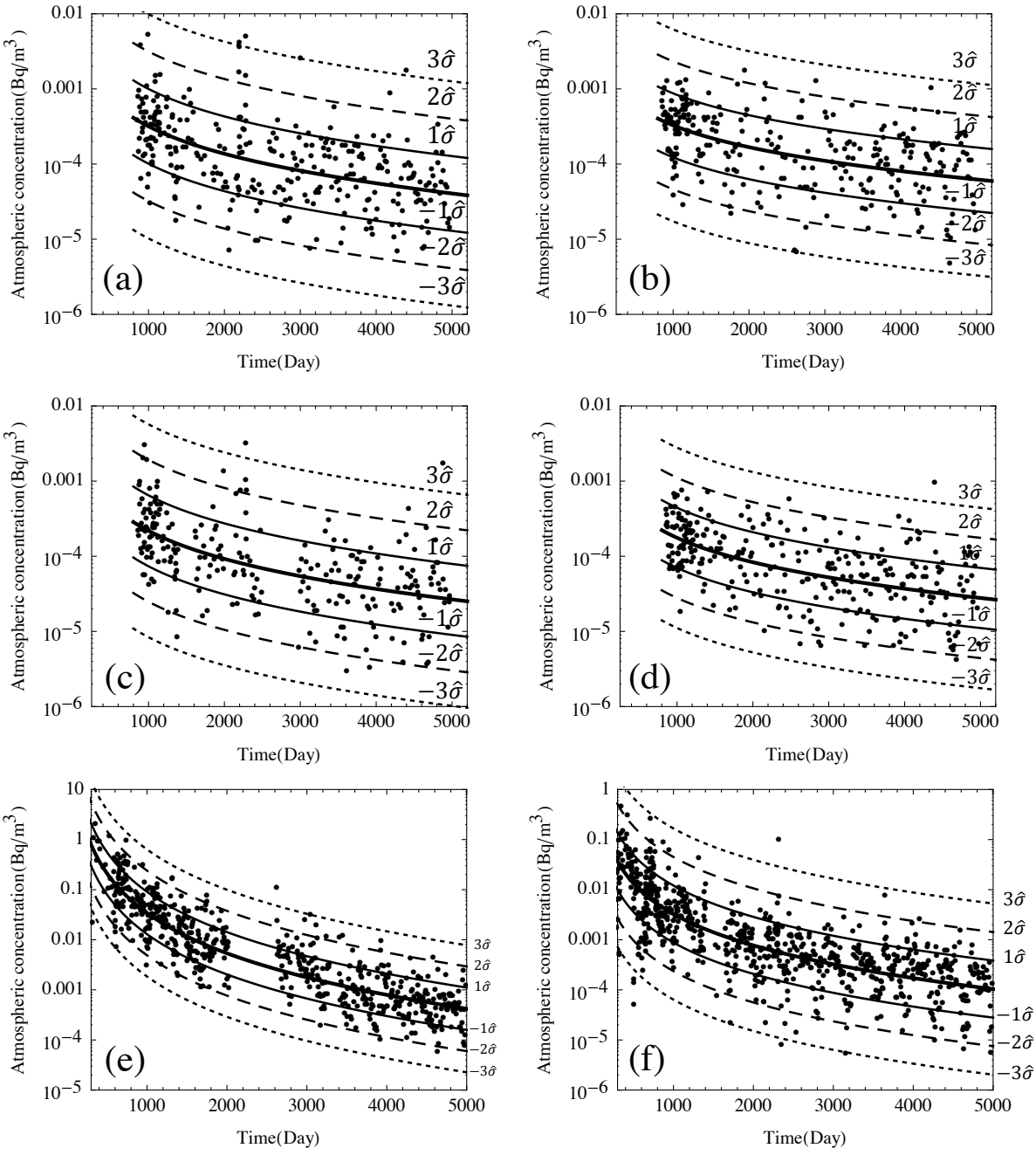

Fig. 9: Standard deviation of fluctuations added to the mean concentration. Curves indicate ±σ, ±2σ, ±3σ. (a) Point 6.0, (b) Point 8.0, (c) Point 13.0, (d) Point 11.0, (e) Point 21.0, (f) Point 60.1. In Point 21.0 and Point 60.1, observation sites are close to the reactor; the atmospheric concentration is higher than other cases, and earlier data are available (since 300 days after the accident). Even such cases, the fluctuations are within the given curves.

Table 1: The values of γ and σ in each observation site

Site number	A	α	γ	σ
Point 6.0	0.785	1.12	0.393	1.01
Point 8.0	0.118	0.846	0.314	0.779
Point 11.0	0.649	1.15	0.415	0.992
Point 13.0	0.173	0.988	0.371	0.794
Point 21.0[a]	3.07×10^{6}	2.63	0.280	0.791
Point 60.1[a]	2.84×10^{3}	1.97	0.353	1.04

[a] Since the sites of Point 21.0 and Point 60.1 are close to the reactor, values of A is very large compared to other sites.

Table, 2: The proportion of data within ± $\hat{\sigma}$, ± $2\hat{\sigma}$, and ± $3\hat{\sigma}$.

Code number	± $1\hat{\sigma}$ range	± $2\hat{\sigma}$ range	± $3\hat{\sigma}$ range
Point 6.0	0.724	0.941	0.986
Point 8.0	0.709	0.957	0.996
Point 11.0	0.720	0.947	0.992
Point 13.0	0.671	0.963	0.997
Point 21.0	0.691	0.950	0.997
Point 60.1	0.740	0.933	0.988
Average	0.709	0.949	0.993