Evaluation of the Effect of Horizontal Diffusion on the Long-Range Atmospheric Transport Simulation with Chernobyl Data

HIROHIKO ISHIKAWA*

Department of Environmental Safety Research, Japan Atomic Energy Research Institute, Japan

(Manuscript received 22 August 1994, in final form 14 November 1994)

ABSTRACT

The effect of horizontal diffusion on the long-range transport simulation is examined with a Lagrangian particle transport model. The transport of radioactivity released from Chernobyl is simulated by the model with different values of horizontal diffusivity. The computed concentrations are statistically compared with measured concentration. The best simulation is found when the magnitude of the horizontal diffusivity is between 3.3 \times 10^4 and 1.0 \times 10^5 m^2 s^{-1}. The performance of empirical formulas of horizontal diffusion, in which mean-square displacement \sigma_2 is specified as a function of time, is also examined.

A part of measured concentrations, which are relatively low concentrations, cannot be explained by transport and diffusion only. It is shown that these measured concentrations can be explained by resuspension of deposited radioactivity.

1. Introduction

In this paper, the effect of horizontal eddy diffusion on the long-range transport simulation is examined with a Lagrangian particle transport model. In the long-range transport simulation, the horizontal resolution of the meteorological field ranges from several tens to a hundred kilometers. The advective wind fields are usually given at discrete times, typically 6- or 12-h steps. Therefore, various local circulations fall in subgrid-scale phenomena. These circulations should be treated as eddy diffusion in the transport calculation.

The Chernobyl nuclear accident promoted the development of long-range transport models for radioactive materials. ApSimon et al. (1987) and Lange et al. (1988) applied their dispersion models to the Chernobyl accident. Developments of new long-range transport models or applications of existing air quality models to radioactivity started in many institutes after the accident (Albergel et al. 1988; Hass et al. 1990; Piedelievre et al. 1990, etc.). Japan Atomic Energy Research Institute also extended the capability of the real-time dose evaluation system (Imai et al. 1985) to simulate the long-range transport of radioactivity on a regional or hemispheric scale (Ishikawa and Chino 1991; Ishikawa 1994a).

In simulating the long-range transport of Chernobyl radioactivity, Ishikawa and Chino (1991) found that, although the high concentrations were simulated relatively well, the low concentrations were underestimated at many sites by their model. They also suggested that the performance of their model was subject to the strength of horizontal diffusion. Thus, it is necessary to examine the sensitivity of the long-range transport simulation to the magnitude of horizontal diffusivity used in the transport model.

In this paper, the transport of Cs-137 from Chernobyl over Europe is computed with different parameterization of horizontal eddy diffusion and the results are compared with the measurement data of REM Data Bank (Raes et al. 1990; Raes et al. 1989; De Cort et al. 1990).

2. Dispersion model

A Lagrangian particle dispersion model is employed in this study because it is totally free from the "numerical diffusion." In the model, a tracer plume is represented by a mass of particles. Each particle is generated at source region in small time steps. These particles are transported by a mean three-dimensional wind field. Three-dimensional random motion is added to the movement of each particle to represent the diffusion due to subgrid-scale eddy diffusion.

The model is formulated in the transformed map coordinates,

\[ x = P_x(\lambda, \phi), \quad y = P_y(\lambda, \phi), \]

where \( P_x \) and \( P_y \) are a pair of conformal map projections and \( \lambda \) and \( \phi \) are longitude and latitude, respec-
tively. The Lambert conformal conic projection is used in this study. In the vertical, a terrain-following height coordinate,

\[ z^* = \frac{z - z_e}{z_i - z_e} = \frac{z - z_e}{h}, \]

is used, where \( z_e(x, y) \) is the terrain height and \( z_i \) is a reference height, which is assumed as constant.

The movement of each particle is computed by

\[ x_{i+\Delta t} = x_i + m(u\Delta t + \delta x), \]
\[ y_{i+\Delta t} = y_i + m(v\Delta t + \delta y), \]
\[ z^*_{i+\Delta t} = z^*_i + w^*\Delta t + \delta z^*. \]

Here, \( x, y, \) and \( z^* \) represent the position of a particle in each direction. The \( m(x, y) \) is the map scale factor, which is the ratio of the transformed distance to the true distance. The \( u, v, \) and \( w^* \) are the advective wind components in the \( x, y, \) and \( z^* \) directions.

The \( \delta x, \delta y, \) and \( \delta z^* \) are the submovements of a particle in each direction, which represent the effect of transport due to subgrid-scale eddies. The horizontal components are computed by

\[ \delta x = (24K_{\text{hor}}\Delta t)^{0.5} R_i^{0.5}, \]
\[ \delta y = (24K_{\text{hor}}\Delta t)^{0.5} R_j^{0.5}, \]

where \( K_{\text{hor}} \) is the horizontal diffusivity, \( \Delta t \) (120 s) the time step, and \( R_i^{0.5}, R_j^{0.5} \) a random number between \(-0.5\) and \(0.5\). If the particles are placed at the same position at \( t = 0 \) and \( K_{\text{hor}} \) is homogeneous, then the position of particles at \( t > 0 \) are subject to the Gaussian distribution, in which the standard deviation \( \sigma_x \) is \( (2K_{\text{hor}}\Delta t)^{0.5} \).

To be precise, the “horizontal” submovement accompanies some \( z^* \) components where the constant \( z^* \) surface is not horizontal. However, this small component is ignored.

The vertical submovement is also computed by a random walk of the particles but with a different scheme. To consider the steep vertical gradient of vertical diffusivity \( K_z \), the following scheme developed by Diehl et al. (1982) is used:

\[ \delta z^* = \pm h \left\{ \left[ 2\Delta t(K_0 + K_z) + (\Delta tK_1)^2 \right]^{0.5} + \Delta tK_1 \right\}. \]

In this scheme, subgrid distribution of vertical diffusivity is linearly approximated between adjacent vertical grids as \( K_z(z) = K_0 + K_z(z) \). The sign, plus or minus, is randomly chosen at each time step.

Each particle carries a certain amount of radioactivity. The amount of the radioactivity carried by the \( n \)th particle \( \alpha_n \) changes according to

\[ \frac{d\alpha_n}{dt} = -kv_n\alpha_n - \Delta\alpha_n - \lambda_{\text{decay}}\alpha_n. \]

The term \(-kv_n\alpha_n\) represents the loss due to dry deposition. Each particle in the bottom layer gives a portion of its radioactivity to the underlying surface ac-

cording to this term. Here, \( v_n \) is the deposition velocity and \( k \) the weight, which depends on the height of particle \( z^*_p \), as \( k = 2[1 - (z^*_p / \Delta z_{\text{bot}})] / \Delta z_{\text{bot}}, \) where \( \Delta z_{\text{bot}} \) is the depth of the bottom layer. The term \(-\Delta\alpha_n\) is the loss due to the precipitation scavenging, where \( \Delta \) is the scavenging coefficient. The last term is the radioactive decay, where \( \lambda_{\text{decay}} \) is the decay constant of the nuclide.

The air concentration is computed with reference to the Eulerian grid. The air concentration at a certain unit cell \( C_{i,j,k} \) is computed by summing up the contribution of each particle to the cell and is expressed by

\[ C_{i,j,k} = \frac{1}{V_{i,j,k}} \sum b_{i,j,k}^n \alpha_n, \]

where \( V_{i,j,k} = m^{-2}h\Delta x\Delta y\Delta z^* \) is the volume of an Eulerian cell. Suffixes \( i, j, \) and \( k \) represent positions in the \( x, y, \) and \( z^* \) directions, respectively. The \( b_{i,j,k}^n \) is the contribution of the \( n \)th particle to the \( i, j, k \)th cell.

This is defined as the overlap of a Lagrangian cell around a particle over an Eulerian cell as illustrated in two dimensions in Fig. 1. This scheme is employed only to smooth the irregular fluctuation of the concentration at low-concentration areas, which is caused by the discrete nature of the Lagrangian particle model.

The output of the model is the air concentration averaged over a certain period, typically 6 h, rather than an instantaneous concentration.

The deposition is computed by summing up the first and the second terms of Eq. (2) at each horizontal location \((i, j)\).

3. Design of simulation

a. Computational domain, meteorology, and source terms

The computational domain is a 4508 km \( \times \) 4508 km area on the map coordinates. The area is divided into 49 \( \times \) 49 computational grids with 92-km intervals. The vertical depth of the computational

![Fig. 1. Computation of the concentration at Eulerian grid.](image-url)
domain is 6000 m; $z_i = 6000$ m is assumed. The vertical mesh number is 20, and the mesh width varies from 100 (bottom layer) to 500 m (top layer).

The advective wind fields are constructed using meteorological data from ECMWF (European Centre for Medium-Range Weather Forecasts), which were provided during the author’s participation in the Atmospheric Transport Model Evaluation Study (ATMES) jointly coordinated by the International Atomic Energy Agency (IAEA), World Meteorological Organization (WMO), and Commission of the European Communities (CEC) (Klug et al. 1992). The data were originally given in latitude–longitude grid in 1.125° resolution. In the vertical, the data were given at standard pressure levels up to 500 hPa or at sigma levels of the objective analysis model. The data at standard pressure levels are used in this study. The data cover 1200 UTC 25 April–1800 UTC 10 May with 6-h time steps. A mass-consistent wind model (Ishikawa 1994b) is used to fit the data to the present coordinates and

![Fig. 2. Comparison of the horizontal diffusivity and the corresponding horizontal growth of a “puff.”](image)

<table>
<thead>
<tr>
<th>Table 1. Numerical experiments conducted in this study.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experiments</td>
</tr>
<tr>
<td>---------------</td>
</tr>
<tr>
<td>A</td>
</tr>
<tr>
<td>B</td>
</tr>
<tr>
<td>C</td>
</tr>
<tr>
<td>D</td>
</tr>
<tr>
<td>E</td>
</tr>
<tr>
<td>F</td>
</tr>
<tr>
<td>G</td>
</tr>
<tr>
<td>H</td>
</tr>
<tr>
<td>I</td>
</tr>
</tbody>
</table>

![Fig. 3. Computational grids where measurements data are available.](image)
FIG. 4. Distribution of surface air concentration (experiment A).
grid system. The wind data in longitude–latitude coordinates are first fitted horizontally to the model grid at each pressure level. Then, vertical interpolation is used to obtain a value at each vertical grid point. Finally, the wind field is adjusted by the variational method to satisfy the mass conservation of compressible atmosphere. The wind field is constructed every 6-h time step. In the transport calculation, the wind field at intermediate time steps is computed by linear interpolation of 6-h fields.

The rainfall intensity used in the calculation of wet deposition is computed from the 6-h accumulated precipitation as analyzed by Scheele and Verver (1990).

The source terms specified in the ATMES (Klug et al. 1992) are used in this study. As for the initial shape of the plume, particles are randomly distributed between 0.1z_c and 1.9z_c, where z_c is the height of the initial plume mass center specified in the ATMES. The number of particles is 8000 per day.

b. Diffusion coefficients

Horizontal dispersion is caused by several processes. It is caused by horizontal inhomogeneity of the mean horizontal wind. Subgrid-scale fluctuations of horizontal wind also diffuse the material. Besides these two processes, the vertical diffusion and the vertical shear of horizontal wind cooperatively work to disperse atmospheric contaminants horizontally. These processes are not independent. If the contaminants are dispersed widely through one of these processes, there will be a greater chance that the contaminants will be further dispersed by other processes.

Contrary to the vertical diffusion, which can be determined with available meteorological data (e.g., Louis 1979), it is difficult to relate horizontal diffusion to available data in the long-range transport simulation. The effect of diffusion on long-range transport was examined by Brost et al. (1988) with the Cross-Appalachian Tracer Experiment (CAPTEX) data. Using the Prather scheme (Prather 1986), which is largely free from numerical diffusion, they concluded that \( K_{\text{hor}} = 3.3 \times 10^4 \text{ m}^2 \text{ s}^{-1} \) is acceptable but that \( K_{\text{hor}} = 3.3 \times 10^5 \text{ m}^2 \text{ s}^{-1} \) is too large.

From the Lagrangian point of view, various empirical formulas of horizontal spread of a puff such as Heffter (1965), Hage et al. (1967), and Gifford (1982) have been proposed. Although Gifford’s formula was derived based on theoretical considerations, it is referred to as “empirical” because the parameters in the formula were determined by fitting the formula to existing dispersion data. Later, Gifford (1984) gave a theoretical discussion on the values of the parameters.

In these formulas, the horizontal spread of a puff is expressed as a function of time after the release of the puff. These formulas seem to be more realistic than using a constant diffusion coefficient because the scale of turbulence, which acts on a plume, increases with time. Thus, it is worth it to examine how these formulas fit the Chernobyl situation, which is rather complicated in meteorology and source terms.

The mean-square displacement \( \sigma_y \) in these empirical formulas is related to the horizontal diffusivity by the following relation:

\[
K_{\text{hor}} = \frac{1}{2} \frac{d \sigma_y^2}{dt}.
\]

Conversely, if constant diffusivity is assumed, the spread of a puff \( \sigma_y \) is computed by the relation. The \( K_{\text{hor}} \) and \( \sigma_y \) for various formulas are compared in Fig. 2. In this study, the model runs with different parameterization of the horizontal diffusion as listed in Table 1. In experiments G, H, and I, \( K_{\text{hor}}(t) \) is computed with this relation for each particle. The \( t \) is taken from the lapse of time of each particle after its release.

The vertical diffusion coefficient is also important, because it promotes the horizontal spread of a plume when vertical shear of horizontal wind exists. In this study, however, a simple parameterization by Ishikawa and Chino (1991),
is used. Both an upper and a lower limit, 50 and 0.1 m$^2$ s$^{-1}$, respectively, are imposed on the values computed by this formula. The typical value in the present study is 50 m$^2$ s$^{-1}$ in the mixing layer (0 $< z^* <$ 1000 m). This results in fairly rapid vertical transport in the mixing layer, so that the horizontal spread of the plume due to vertical wind shear is sufficiently incorporated into the calculation. Of course, it is desirable to use a more sophisticated parameterization. However, when the meteorological fields are given at standard pressure levels with a 6-h interval, it is not easy to deduce the fine structure of the planetary boundary layer. From an applicational point of view, the simple parameterization of the present work seems to be practical.

c. Deposition parameters

The dry deposition velocity and the scavenging coefficient used in this model are 0.001 m s$^{-1}$ and 5.5 $\times$ 10$^{-5}$J$^{0.8}$ s$^{-1}$, where J (mm h$^{-1}$) is the rainfall intensity. These values are quoted from ApSimon et al. (1985). Jylhä (1991) estimated that the scavenging coefficients for Cs-137 and I-131 released at Chernobyl were (3.4 $\pm$ 0.9) $\times$ 10$^{-5}$J$^{0.59\pm0.08}$ s$^{-1}$ and (7 $\pm$ 5) $\times$ 10$^{-5}$J$^{0.69\pm0.12}$ s$^{-1}$, respectively, over southern Finland. These scavenging coefficients are close to those in ApSimon et al. (1985). Thus, it is decided to use the value.

4. Results and discussion

a. Measurement

Prior to the presentation of the computed results, the measurement data used in this section are briefly described. In the REM Data Bank, the surface air concentration for Cs-137, Cs-134, and/or I-131 are given at 85 locations. These data cover 70 grids of the simulation model as shown in Fig. 3. The sites, which will be referred to in this paper, are also shown in Fig. 3. The data vary in their sampling conditions such as in the time of filter change and the sampling duration. It is not obvious whether the time is reported in universal or local time. In the statistical comparison described in section 4c, daily values are reconstructed from the original data to mask these uncertainties.

The accumulated surface deposition data for cesium are also given from the REM Data Bank (De Cort et al. 1990). These data cover 330 grids of the simulation model.

b. General features of the computed plume

The evolution of the concentration near the surface in experiment A (the control run, $K_{hor} = 0$) is shown
in Fig. 4. The painted and hatched areas represent regions where the concentration exceeds 1.00 and 0.01 Bq m\(^{-3}\), respectively. The overall feature of the movement of the radioactive plume is similar to those in former studies (e.g., Ishikawa and Chino, 1991). It should be noted that the portion of the plume that covered central and western Europe, where most of the monitoring data existed, consisted of the radioactivity released from the afternoon of 26 April to the morning of 27 April. The radioactivity released during the afternoon of 27 April–3 May was transported to the east or south of Chernobyl, which covered eastern Europe, Greece, and Turkey. The radioactivity released on 4 and 5 May was first transported to the southwest of Chernobyl and was subsequently transported northward, which covered eastern and northern Europe.

In Fig. 5, the surface air concentration patterns of Cs-137 on 2 May computed with different values of horizontal diffusivity are compared. The distribution pattern of experiment B (\(K_{hor} = 1.0 \times 10^{4} \text{ m}^{2} \text{ s}^{-1}\)) is similar to that of experiment A (Fig. 4f), so that it is not presented. It can be seen that the area where concentration exceeds 0.01 Bq m\(^{-3}\) increases if the greater horizontal diffusivity is used. In experiment F, the hatched area covers most of the sites where measured data were available.

The difference of air concentration due to the difference of horizontal diffusivity is also seen in the temporal change at individual sites. In Figs. 6–8, the surface air concentration of experiments B, C, E, and F are compared with the measurement for Berlin, Ispra, and Paris. In these figures, the thin lines represent measured concentration and the thick lines represent computed concentration. The squares in the figures will be explained in a later section. At Berlin (Fig. 6), experiment B does not simulate the initial arrival of real plume. The computed arrival time is approximately 1 day later than observed, and the computed concentration has a gap between 1 and 3 May. In experiment E, the computed result is improved but it still has the same features as experiment B. In experiment E, the gap is filled up but the temporal change is not as sharp as that of the measurement. Experiment F fails to trace the variation of measured concentration. At Ispra (Fig. 7), measured concentration shows a neat temporal variation. Experiments B and C trace this variation well, but the arrival time is delayed in experiment B. Experiment E also traces the variation well, but the second simulated peak is not as sharp as the measured one. Experiment F fails to reproduce the variation. Paris (Fig. 8) is close to the western periphery of the plume where experiments E and F reproduce the measurement well.

c. Statistical performance

Before computing statistical quantities, it is informative to see the cumulative concentration distribu-
Fig. 8. Same as Fig. 6 but for Paris.

tions. The cumulative concentration distributions of computed results with different values of horizontal diffusivity are compared with that of measurement in Fig. 9. Each population of data are sorted according to the magnitude of concentration. Then, the concentrations corresponding to each percentile is plotted in Fig. 9.

The thick line shows the cumulative distribution of measured concentration. Although the measured data come from irregularly distributed sites (Fig. 3), the cumulative distribution shows a rather smooth curve. A characteristic feature is that there is a "shoulder" between the 5th and 20th percentile. This suggests that different mechanisms work below and above the 20th percentile.

The thin lines show the cumulative distribution of concentrations computed with different values of horizontal diffusivity. Generally speaking, every experiment underestimates the measured concentration because the curves of experiments are under that of measurement above the 50th percentile. This might be caused partly by the uncertainty of the source term. Experiment F ($K_{hor} = 3.3 \times 10^3 \text{ m}^2 \text{ s}^{-1}$) shows apparent underestimation of high concentrations and overestimation of low concentrations. This is attributed to excess horizontal dispersion. The curve for experiment E ($K_{hor} = 1.0 \times 10^2 \text{ m}^2 \text{ s}^{-1}$) follows well that for measurement between the 20th and 50th percentile but it shows underestimation above that. This suggests that the computed results are still dispersive. The curve for experiments D ($K_{hor} = 5.0 \times 10^3 \text{ m}^2 \text{ s}^{-1}$) and C ($K_{hor} = 3.3 \times 10^4 \text{ m}^2 \text{ s}^{-1}$) follows that for measurement in a wide range, although there is a general feature of underestimation. As for experiments B ($K_{hor} = 1.0 \times 10^4 \text{ m}^2 \text{ s}^{-1}$) and A ($K_{hor} = 0$), it is obvious that the model did not sufficiently compute the low concentrations.

The scatter diagrams of computed versus measured concentrations are compared in Fig. 10. The data are plotted in a logarithmic scale because both computed and measured data range in several orders. The values, which are less than $1.0 \times 10^{-5} \text{ Bq m}^{-3}$, are assumed as $1.0 \times 10^{-5} \text{ Bq m}^{-3}$. The solid lines are the regression lines.

The correlation coefficient

$$R = \frac{\sum(C_i - \bar{C})(O_i - \bar{O})}{\left[\sum(C_i - \bar{C})^2\sum(O_i - \bar{O})^2\right]^{0.5}}$$

and the variance

$$S_d^2 = \frac{1}{N - 1} \sum[(C_i - \bar{C}) - (O_i - \bar{O})]^2$$

are computed for the populations of computed data $C_i$ and measured data $O_i$. The variance, rather than the mean-square error, is used as the measure of dif-
ference because there is an uncertainty in the mean value of computed data, which is attributed to the uncertainty of the source term. The correlation is computed for both the actual concentration and the logarithm of concentration. The Spearman correlation, which is a correlation of the ranks, rather than the values, of each population (e.g., Weber et al. 1982), is also computed. The results are listed in Table 2 together with the slope of the first-order regression line.

Among experiments A–F, the slope of regression line decreases with increasing horizontal diffusivity. The slope should be unity if the real plume is correctly simulated. If the slope is more than unity, it implies that the dispersion is weaker than the reality. If the slope is less than unity, it implies that the simulated plume is too dispersive. In this sense, the best simulation will be found in experiments D and E.

The correlation is the highest in experiment E, then next highest in experiments D and C, respectively. The correlation of logarithm of concentration is the highest in experiment C, then next highest in experiments D and E, respectively. The variance is minimal in experiments C, D, and E. The Spearman correlation is the highest in experiment D and then next highest in experiments E and C, respectively. Although the difference of each statistical quantity through experiments A–F is too small to derive definite conclusion, it can be said that experiments C, D, and E generally show higher statistical performance than the others.

As for experiments G, H, and I, in which the horizontal diffusion is computed from empirical formulas of $\sigma_y$, the correlation of log concentration and Spearman correlation are comparable to those for experiments C, D, and E, although the correlation $R_{\text{linear}}$ and the variance are not as good as experiments C, D, and E. Slopes are near unity especially for experiments G and H.

It is a little curious that the performance of experiment G is not as good as that of experiment D. As seen in Fig. 2, the horizontal diffusivity of experiment G reaches its saturated value ($5.0 \times 10^4 \text{ m}^2 \text{ s}^{-1}$) approximately 12 h after the release of a puff. Therefore, the difference in experiments D and G must be attributed to the behavior of the growth of a plume during the first 12 h after the release. Suppose that the continuous plume consists of many subsequent puffs. It is obvious that the use of Gifford’s $\sigma_y$ is more realistic for the growth of the individual puff than the use of constant horizontal diffusivity. However, although the growth of an individual puff may be subject to the Gifford’s $\sigma_y$, subsequent puffs may be advected in different directions because of the “temporal” fluctuation of the mean wind. This effect is not fully reflected in the present simulation since the mean wind is specified in 6-h steps and the temporal fluctuation between the 6 h is reflected only by linear temporal interpolation. Qualitatively speaking, the larger horizontal submovements of particles in experiment D can be considered to fill up this effect.

d. Consideration of resuspension

As seen in the scatter diagrams in Fig. 10, there are still many data points on the abscissa even in experiments C, D, and E. These data points imply that the measured concentrations are not explained by the passage of radioactive plume. The surface air concentration at some sites computed in experiment D ($K_{\text{hor}}$
The air concentration near the surface due to resuspension $C_{\text{resus}}$ is conventionally estimated by

$$C_{\text{resus}} = f_{\text{resus}}C_{\text{dep}},$$

where $C_{\text{dep}}$ (Bq m$^{-2}$) is the surface deposition and $f_{\text{resus}}$ the resuspension factor. Resuspension is a very complicated phenomenon, which depends on various factors such as wind speed, surface wetness, particle size, etc. Actually, the reported values of the resuspension factor vary in a wide range between $1.0 \times 10^{-4}$ and $1.0 \times 10^{-10}$ m$^{-1}$ (Nicholson 1988). However, Garland and Pattenden (1991) stated that $f_{\text{resus}}$ is of the order
$10^{-6}$–$10^{-4}$ m$^{-1}$ soon after deposition based on the data from the field experiment.

The squares in Figs. 6, 7, 8, and 11 represent the air concentration near the surface due to the resuspension of surface deposition computed by Eq. (3) with $f_{\text{resus}} = 10^{-6}$ m$^{-1}$. The model-computed deposition is used for $C_{\text{dep}}$. The model-computed deposition accumulated until the end of calculation agreed to the measured deposition data from the REM Data Bank within a factor of 2, except for Ispra, where the measured deposition was considerably greater than the computed deposition. It is seen in the figures that the computed concentrations due to resuspension have magnitude sufficient to explain the measured concentrations, which were not explained by the plume. Considering the complexity of the actual resuspension, it can be concluded that resuspension explains the measured concentration after the passage of radioactive plume at these sites.

The effect of resuspension was also examined statistically. In this, $C_{\text{resus}}$ was computed for various magnitudes of $f_{\text{resus}}$ ($10^{-8}$, $10^{-7}$, $10^{-6}$, and $10^{-5}$ m$^{-1}$) and it was added to the air concentration due to radioactive plume. Then, the values are statistically compared with measured data. This comparison was performed for experiments A, B, C, D, and G but not for the other experiments because the slopes are less than unity.

**Table 2. Statistical performance of each experiment.**

<table>
<thead>
<tr>
<th>Case</th>
<th>$R_{\text{linear}}$</th>
<th>$R_{\text{log}}$</th>
<th>$\rho$</th>
<th>Variance</th>
<th>$S$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.460</td>
<td>0.739</td>
<td>0.664</td>
<td>1.62</td>
<td>1.27</td>
</tr>
<tr>
<td>B</td>
<td>0.562</td>
<td>0.768</td>
<td>0.744</td>
<td>1.28</td>
<td>1.25</td>
</tr>
<tr>
<td>C</td>
<td>0.617</td>
<td>0.786</td>
<td>0.771</td>
<td>1.13</td>
<td>1.18</td>
</tr>
<tr>
<td>D</td>
<td>0.630</td>
<td>0.778</td>
<td>0.780</td>
<td>1.12</td>
<td>1.10</td>
</tr>
<tr>
<td>E</td>
<td>0.658</td>
<td>0.771</td>
<td>0.773</td>
<td>1.11</td>
<td>0.926</td>
</tr>
<tr>
<td>F</td>
<td>0.573</td>
<td>0.744</td>
<td>0.705</td>
<td>1.34</td>
<td>0.696</td>
</tr>
<tr>
<td>G</td>
<td>0.599</td>
<td>0.769</td>
<td>0.771</td>
<td>1.16</td>
<td>1.06</td>
</tr>
<tr>
<td>H</td>
<td>0.557</td>
<td>0.778</td>
<td>0.781</td>
<td>1.23</td>
<td>0.992</td>
</tr>
<tr>
<td>I</td>
<td>0.554</td>
<td>0.761</td>
<td>0.779</td>
<td>1.24</td>
<td>0.828</td>
</tr>
</tbody>
</table>

$R_{\text{linear}}$—correlation of real concentration, $R_{\text{log}}$—correlation of logarithm of concentration, $\rho$—Spearman correlation, $S$—slope of the first-order regression for log concentration.

**Table 3. The statistical values when the resuspension is considered.**

The values in parentheses are the addition or reduction from the values listed in Table 2.

<table>
<thead>
<tr>
<th>Experiments</th>
<th>$R_{\text{log}}$</th>
<th>$S$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.716 (-0.023)</td>
<td>1.09 (-0.18)</td>
</tr>
<tr>
<td>B</td>
<td>0.764 (-0.006)</td>
<td>1.09 (-0.16)</td>
</tr>
<tr>
<td>C</td>
<td>0.792 (+0.006)</td>
<td>1.01 (-0.17)</td>
</tr>
<tr>
<td>D</td>
<td>0.789 (+0.011)</td>
<td>0.99 (-0.11)</td>
</tr>
<tr>
<td>G</td>
<td>0.780 (+0.011)</td>
<td>0.95 (-0.11)</td>
</tr>
</tbody>
</table>

**Fig. 11.** Comparisons of computed concentrations with measurements.
without the consideration of the resuspension in experiments E, F, H, and I, so that no improvement is expected by the addition of concentration due to resuspension.

Among the statistical quantities in Table 2, the correlation of log concentration and the slope of the regression line were sensitive to the consideration of resuspension. The highest statistical performance was obtained when \( f_{\text{resus}} = 10^{-7} \) m\(^{-1}\) for each experiment, contrary to the good agreements in Fig. 11 with \( f_{\text{resus}} = 10^{-6} \) m\(^{-1}\). The statistical values with \( f_{\text{resus}} = 10^{-7} \) m\(^{-1}\) are listed in Table 3. The values in parentheses are the differences of the scores from the values in Table 2. The slope is almost unity for each experiment. The correlation increases for experiments C, D, and G but it decreases for experiments B. The highest increase of log correlation is in experiment D and G. Figure 12 shows scatter diagrams of experiments C, D, and G with the consideration of resuspension. Only a few points remain on the abscissa, and the computed data fit the measurements well. The better statistical performance with \( f_{\text{resus}} = 10^{-7} \) m\(^{-1}\) may partly relate to the fact that air concentrations due to the plume are generally underestimated as mentioned in the previous section. Since the higher values in the measured concentrations, which are due to the plume, are generally underestimated, the better correlation is computed when the lower values, which reflect the contribution from resuspension, are similarly underestimated.

5. Conclusions and remarks

The effect of horizontal diffusion on the long-range transport calculation was evaluated with a Lagrangian particle dispersion model against the Chernobyl data. If a constant horizontal diffusion coefficient was used, the values between \( 3.3 \times 10^4 \) and \( 1.0 \times 10^5 \) m\(^2\) s\(^{-1}\) showed good results. It was also shown that \( K_{\text{hor}} = 1.0 \times 10^4 \) m\(^2\) s\(^{-1}\) was too small and \( K_{\text{hor}} = 3.3 \times 10^5 \) m\(^2\) s\(^{-1}\) was too large. Thus, it is concluded that the appropriate value of diffusion coefficient is between \( 3.3 \times 10^4 \) and \( 1.0 \times 10^5 \) m\(^2\) s\(^{-1}\).

The performance of time-dependent horizontal diffusion, which was derived from empirical formulas for horizontal displacement \( \sigma_y \), was also examined. The results were comparable to the experiments with constant horizontal diffusivity ranging in \( 3.3 \times 10^4 \leq K_{\text{hor}} \leq 1.0 \times 10^5 \) m\(^2\) s\(^{-1}\).

The resuspension of deposited radioactivity was considered to explain the measured concentrations, which were not simulated by transport and diffusion only. When the value of the resuspension factor was \( 10^{-6} \) m\(^{-1}\), the measured concentrations, which cannot be explained by the passage of radioactive plume, were well explained by the contribution of resuspension at several sites. From the statistical point of view, the best correlation was found when the resuspension factor was \( 10^{-7} \) m\(^{-1}\). With this resuspension factor, the correlation of the logarithm of concentrations increased for experiments C, D, and G. The best performance was achieved for experiment C \( (K_{\text{hor}} = 3.3 \times 10^4 \) m\(^2\) s\(^{-1}\)\), and the improvement was the largest for experiments D and G.

It must be noted that the generality of these conclusions is restricted to some extent due to the lack of measurement data near the release site. The majority of measured data were taken after 1 May, so that the nature of plume in the first 4 or 5 days were not fully incorporated in the evaluation. For example, although experiment E \( (K_{\text{hor}} = 1.0 \times 10^4) \) showed good statistical performance, the surface concentration distribution of experiment E seemed to be too dispersive in the early days. Experiments with empirical formulas of \( \sigma_y \) might show better performance if the data in the early stages of dispersion were included.

Acknowledgments. The meteorological data, measurement data, and the source terms were provided during the author's participation in the ATMES (Atmospheric Transport Model Evaluation Study), which...
was jointly coordinated by IAEA, WMO, and CEC. The author would like to express his thanks to these organizations for providing those data. Thanks are extended to Mr. O. Togawa of Japan Atomic Energy Research Institute and Prof. Y. Mitsuta of Disaster Prevention Research Institute of Kyoto University for their helpful comments on this study.

REFERENCES


