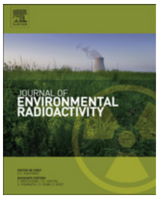
Journal of Environmental Radioactivity 99 (2008) 1644–1652

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad



Simulating mesoscale transport and diffusion of radioactive noble gases using the Lagrangian particle dispersion model

Cheol-Hee Kim[a,*], Chang-Keun Song[b], Sang-Hyun Lee[c], Sang-Keun Song[a]

[a] *Division of Earth Environmental System, Pusan National University, 30 Jangjeon-Dong, Geumjeong-Gu, Busan 609-735, Republic of Korea*
[b] *Global Environment Research Center, National Institute of Environmental Research, Incheon 404-170, Republic of Korea*
[c] *School of Earth and Environmental Sciences, Seoul National University, Seoul 151-747, Republic of Korea*

ARTICLE INFO

Article history:
Received 30 August 2007
Received in revised form 15 May 2008
Accepted 16 May 2008
Available online 30 June 2008

Keywords:
Lagrangian particle dispersion model (LPDM)
Radioactive noble gas (^{85}Kr exponent)
Mesoscale transport
JCO accident
Dilution factors

ABSTRACT

In order to simulate the impact of mesoscale wind fields and to assess potential capability of atmospheric Lagrangian particle dispersion model (LPDM) as an emergency response model for the decision supports, two different simulations of LPDM with the mesoscale prognostic model MM5 (Mesoscale Model ver. 5) were driven. The first simulation of radioactive noble gas (^{85}Kr exponent) emitted during JCO accident occurred from 30 September to 3 October 1999 at Tokai, Japan showed that the first arriving short pulse was found in Tsukuba located at 60 km away from the accidental area. However, the released radioactive noble gas was transported back to the origin site about 2 days later due to the mesoscale meteorological wind circulation, enhancing the levels of ^{85}Kr with the secondary peak in Tsukuba. The second simulation of atmospheric dilution factors (the ratio of concentration to the emission rate, χ/Q), during the underground nuclear test (UNT) performed by North Korea showed that high χ/Q moved to the eastward and extended toward southward in accordance with the mesoscale atmospheric circulations generated by mesoscale prognostic model MM5.

In comparison with the measurements, the simulated horizontal distribution patterns of ^{85}Kr during the JCO are well accord with that of observation in Tsukuba such as the existence of secondary peak which is associated with the mesoscale circulations. However, the simulated level of ^{85}Kr anomaly was found to be significantly lower than the observations, and some interpretations on these discrepancies were described. Applications of LPDM to two mesoscale emergency response dispersion cases suggest the potential capability of LPDM to be used as a decision support model provided accurate emission rate of accident in case of a large accident.

1. Introduction

Computer aided mathematical or pollution models are the important tools in the development of successful control strategies of air pollutants or hazardous materials. In the event of accidental release of harmful materials to the atmosphere, the decision-making process for the emergency response relies on the support of the numerical dispersion models via predicting the locations potentially affected by the passage of harmful clouds. However, many numerical or mathematical dispersion models have their own limitations for representing the real situation. It is, therefore, very important to minimize many sources of model uncertainties arising from various factors such as the source intensity, the dispersion model formulation and the meteorological input data.

The total error or uncertainty in the numerical dispersion models arises from three main components (AIChE/CCPS, 1996): uncertainties generated by input data errors in association with meteorological input data and emission strength, arisen from random variability of atmosphere caused by the stochastic nature of the atmosphere (Venkatram, 1988), and caused by the model formulation errors associated with assumptions of dispersion modeling and treatments of the pollutant transport.

Although an Eulerian approach for quantifying pollution transport and diffusion is used in simulating or predicting the time-varying deposition of harmful materials, it is well known that Lagrangian particle dispersion model (hereafter LPDM) gives a more complete view of the transport and diffusion processes of tracer released in a complex terrain area. Furthermore, the computational complexities associated with the simultaneous solution of many differential equations can be avoided by using the Lagrangian method, and, therefore, this method can get a relatively good result in assessing accidental releases from industrial complexes (Stohl, 1998; Peters et al., 1995; Song et al., 2003).

When a radioactive accident occurred at the JCO facilities in Tokai, Ibaraki, Japan on 30 September 1999, it was anticipated that

---

[*] Corresponding author. Tel.: +82 51 510 3687; fax: +82 51 515 1689.
E-mail address: chkim2@pusan.ac.kr (C.-H. Kim).

0265-931X/$ – see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jenvrad.2008.05.002

large amounts of the short-lived fission products were emitted (Igarashi et al., 1999, 2000a; Imanaka, 2000; Mitsugashira et al., 2000; STA, 1999). The situation of the accident was gradually unveiled as the dose assessments were carried out by employing radiation monitoring data measured with survey instruments and radiation monitors in and around the accident site. Total quantitative emissions of neutrons and gamma-rays during the JCO criticality accident were initially substantiated by Tanaka (2001), and the total number of fission reactions was identified to be 2.5×10^{18} on the basis of activity analysis of the fission products in the solution of the precipitation vessel, which was also turned out to be identical to that evaluated from the analysis of reactions in the stainless-steel net on the cooling tower at JCO Co., Ltd. (Tanaka, 2001). Endo et al. (2003) reported the intensity of fission neutron measured with a high-sensitive neutron collar during the JCO criticality accident, and presented the time evolution of neutron intensity for the appropriate dose assessment. While there were several answers to the question on the nuclear fission products during the JCO criticality accident, however, the possibility of transport of fission products to other regions and its simulation has not yet been quantitatively demonstrated. Igarashi et al. (2000a) estimated the effects of transport by applying a simple Gaussian box model to the analysis domain. They concluded that the transported radioactive noble gas, ^{85}Krypton (^{85}Kr exponent), would never be elevated by more than 1 mBq m^{-3} at city of Tsukuba. However, the actual three-dimensional behaviors of a plume transport cannot be described by a simple Gaussian model due to its limitation in describing temporal and spatial changes in wind speed/direction turbulent intensities over complex terrain. These limitations can be overcome with the use of the three-dimensional LPDM.

In North Korea, the Underground Nuclear Test (hereafter UNT) was known to be carried out on 9 October 2006, and the US and South Korean Geological Survey Web site recorded a light 3.9–4.2 magnitude earthquake on Richter Scale at 10:35 a.m., and designated UNT site somewhere located about 385 km northeast of Pyongyang, the North Korean capital. Although the scale of measured earthquake was about the same as small as moderate earthquake, it is highly critical to gather or sometimes predict to provide the accurate information on the distribution of radioactive concentrations for emergency response procedures in the event of a large accident such as UNT. Some studies demonstrate that the emergency response system based on meteorological and air quality models for the dispersion forecast is able to assist the decision-making process in cases involving nuclear accidents and oil spills (Galmarini et al., 2001; Saltbones et al., 1998; Annika et al., 2001).

This paper presents two simulations of three-dimensional transport and diffusion of a radioactive noble gas released during both the (1) JCO accident from 30 September to 3 October 1999 at Tokai in Japan and (2) atmospheric dilution factors (χ/Q) under the assumption of continuous emission of radioactive gas during UNT using the LPDM model. Here, atmospheric dilution factors (χ/Q, unit: s m^{-3}) mean the ratio of concentration (χ) to the emission rate (Q). The prognostic meteorological model, MM5, was employed in order to assess the impact of mesoscale meteorological three-dimensional wind fields, and discussion on the potential capabilities of LPDM in association with the mesoscale meteorological model was also described.

2. Modeling description

2.1. Lagrangian particle dispersion model (LPDM)

The LPDM model has been developed by Park (1994) and its performance has been tested using the data obtained from the SF$_6$ tracer experiment conducted in the central east coastline of South Korea (Park and Kim, 1998). For our convenience, we briefly describe the LPDM model.

The location of a conditioned particle (x_i) after a release time interval of Δt is computed in a Lagrangian manner (Smith, 1968; McNider, 1981; Arritt, 1985; Park, 1994) in such a way that,

$$x_i(t+\Delta t) = x_i(t) + \left(u_i(t) + u_i'(t)\right)\cdot\Delta t,$$

where u_i is the modeled grid-scale wind and u_i' is the turbulent fluctuation, which is computed by solving the Langevin diffusion equation using a Markov-chain formulation (Legg and Raupach, 1982)

$$u_i'(t+\Delta t) = R_{\mathrm{L},i}(\Delta t)\cdot u_i'(t) + u_i''(t),$$

where $R_{\mathrm{L},i}$ is the Lagrangian autocorrelation coefficient, and $u_i''(t)$ is a random turbulent component which is assumed to be independent of $u_i'(t)$. The Lagrangian autocorrelation coefficient $R_{\mathrm{L},i}(\Delta t)$ is assumed to be an exponential form (Gifford, 1982), i.e.,

$$R_{\mathrm{L},i}(\Delta t) = \exp\left[-\frac{\Delta t}{T_{\mathrm{L},i}}\right],$$

where $T_{\mathrm{L},i}$ is the Lagrangian integral time scale of the subgrid-scale turbulence. The Lagrangian integral time scale in the ith direction $T_{\mathrm{L},i}$ is related to the Eulerian integral time scale, $T_{\mathrm{E},i}$ which is directly derived from the turbulence parameterization of the meteorological model. Deposition is calculated using the resistance law as described by Park (1998b).

Using Taylor's hypothesis, the diffusion distance, σ_i of each particle in the ith direction after the Δt time interval, is estimated by (Park, 1998b)

$$\begin{aligned}\sigma_i(t+\Delta t) &= \sigma_i(t) + \sigma_{\mathrm{v},i}\Delta t, \quad \text{for } t \le 2T_{\mathrm{L},i},\\ \sigma_i^2(t+\Delta t) &= \sigma_i^2(t) + 2\sigma_{\mathrm{v},i}^2 T_{\mathrm{L},i}\Delta t, \quad \text{for } t \ge 2T_{\mathrm{L},i},\end{aligned}$$

where $\sigma_{\mathrm{v},i}$ is the standard deviation of wind speed in the ith direction and can be obtained from Park (1994) at each time step from the meteorological model. The full description of the model is given in Park (1998b). Estimates of ground-level concentration at any time are calculated using the kernel density estimator (Park, 1993, 1998a).

2.2. Calculation of dilution factors (χ/Q)

A kernel density estimator in Yamada and Bunker (1986) and Park (1994) is used to calculate elative pollutant concentrations. In this estimator, each particle represents a center of a puff, and the concentration distribution in the puff is assumed to be a Gaussian distribution where variances are determined as the time integration of the velocity variances encountered over the history of the puff. The concentration at a given time and space is determined as the sum of the concentrations of each puff (Park, 1998b; Song et al., 2003) i.e.,

$$\begin{aligned}\frac{\chi(x,y,z)}{Q} = {} & \frac{\Delta t}{(2\pi)^{3/2}}\sum_{k=1}^{N}\frac{1}{\sigma_{xk}\sigma_{yk}\sigma_{zk}}\exp\left(-\frac{(x_k-x)^2}{2\sigma_{xk}^2}\right)\\ & \times\exp\left(-\frac{(y_k-y)^2}{2\sigma_{yk}^2}\right)\left[\exp\left(-\frac{(z_k-z)^2}{2\sigma_{zk}^2}\right)\right.\\ & \left.+\exp\left(-\frac{(z_k+z)^2}{2\sigma_{zk}^2}\right)\right],\end{aligned}$$

where (x_k, y_k, z_k) is the location of kth particle, σ_{xk}, σ_{yk}, σ_{zk} are the standard deviations of a Gaussian distribution, Q is the emission rate, and N is the number of released particles. The width of puff, σ_i is obtained using the Taylor's hypothesis as estimated by Park (1998b).

2.3. Mesoscale meteorological model

The meteorological model used in this study is the prognostic Mesoscale Model version 5 (MM5v3, Pennsylvania State Univ./National Center for Atmospheric Research) to produce three-dimensional wind fields. The model domain is centered 36.5°N and 140.6°E, covering a large part of East Asia, including Japan, North and South Korea, and the eastern part of China (Fig. 1). The model consists of 81 × 81 horizontal grid points with the horizontal grid resolution of 30 km and 23 vertical layers (top level is 20 km) in a sigma coordinate. The MM5 has been initialized using the National Center for Environmental Program (NCEP) Reanalysis Database (2.5° resolution, 6 h interval), and the forecasted meteorological fields are converted to the terrain following coordinate to provide three-dimensional meteorological wind and turbulent fields to LPDM.

3. Application of LPDM

3.1. Case selection

A criticality accident occurred at the JCO facilities at Tokai, Ibaraki in Japan on 30 September 1999 (Fig. 1). According to reports by the Science and Technology Agency (STA, 1999), Mitsugashira et al. (2000) and Imanaka (2000), the estimated total number of fission events was in the range of about $1–4 \times 10^{18}$, resulting in the total ^{85}Kr emission of 1.48×10^7 Bq based on the fission yield of 2.5×10^{18} listed by the Nuclear Safety Commission (NSC, 1989).

Igarashi et al. (2000a) evaluated the influence of the JCO accident based on the observed meteorological variables at Japanese Meteorological Research Institute in Tsukuba which is situated 60 km southwest of Tokai (Fig. 1). They estimated ^{85}Kr of less than 1 mBq m^{-3} using the 'diffusion box' of 3600 m in length with a wind speed of 3 m s^{-1} and the horizontal and vertical diffusion coefficients of 100 and 1 m^2 s^{-1}, respectively, under the stable atmospheric condition and a weak wind speed (3 m s^{-1}).

North Korea announced that the UNT was carried out on 9 October 2006, yet North Korea's claim was put in doubt at first time. However, although the scale of measured earthquake was small, weak magnitude earthquake was recorded at South Korean Geological Institute. As other monitoring systems located in other countries including Russia and Japan had detected an explosion in North Korea early on October 9, it is believed to be an underground nuclear explosion, and was considered to be one of the most concerns threatening to peace not only for the Korean peninsula but for the East Asian region.

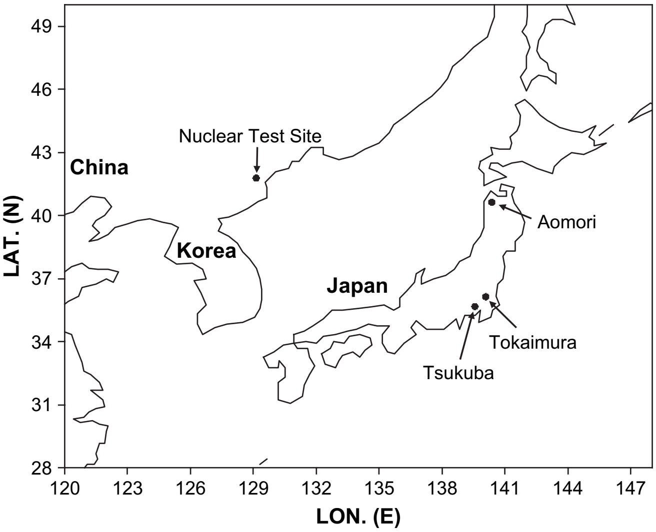


Fig. 1. Domain of three-dimensional Lagrangian particle dispersion modeling and the locations of JCO criticality accident and UNT.

3.2. Conditions for the numerical simulation

The Lagrangian particle release at the location of the accident is started at 10:30 LST on 30 September at the height of 10 m above the ground. The simulation has been conducted for 5 days after the first particle release. The Lagrangian particles are released at a rate of one particle per 1 s, and the duration of emission is assumed to be 20 h in total as reported by Mitsugashira et al. (2000) and Endo et al. (2003). As there were two periods in the progress of the JCO critical accident, the emission intensities are also divided into two periods: BURST and PLATEAU (Endo et al., 2003; Miyoshi et al., 2001; Tanaka, 2001). The BURST period is the first 25 min, in which the dose rate changed markedly in a short time period. The PLATEAU period is for the rest of hours after the BURST, during which the dose rate decreased gradually. During the BURST, the Lagrangian particles are released with 11% of total emission (1.48×10^7 Bq), and 89% was released during the PLATEAU. Therefore, total 1500 particles are released for the first 25 min (amount of radioactivity of a particle is 1085.3 Bq of ^{85}Kr), and thereafter 70,500 particles are released in total (amount of radioactivity of a particle is 186.8 Bq of ^{85}Kr). To calculate radioactive noble gas (^{85}Kr), a kernel density estimator (Yamada and Bunker, 1986; Park, 1994, 1998a) is used.

As there was no available information on the emission of the UNT, only dilution factors (χ/Q) based on the meteorological wind fields were simulated under the assumption that the radioactive noble gas was released continuously. We only used the gathered information announced to public such as the location of UNT and the local time that were estimated officially from the recorded earthquake in South Korea. Meteorological input data of MM5 were used, and other physical sink process such as dry and wet deposition was not taken into account in this study.

4. Results

4.1. Synoptic fields

The synoptic surface weather maps on both two cases are shown in Fig. 2. Weather maps on 1 and 3 October 1999 during the JCO accident were presented in Fig. 2a and b, and those on 10 and 12 October 2006 during the UNT were indicated in Fig. 2c and d.

On 1 October 1999, a high-pressure system located at the center of analysis domain yields a weak northeasterly over the accidental area. On October 3, a cold front in association with the low-pressure system located over the northern part of Japan is extended southeastward into the southern part of Japan with anti-clockwise airflows around the front. This system continuously moves northeastward followed by a surface high-pressure system located at eastern China. This low-pressure system results in a relatively strong wind over the accidental area on 3 October 1999 (Fig. 2a and b).

On 10 October 2006, strong high-pressure over Japan and a low-pressure system over the North Korea were located, and, southwesterly and westerly prevailed between the high- and low-pressure system with large horizontal pressure gradient around nuclear test site. On October 12, a new high-pressure system was located to the north of North Korea, and moved southeastward yielding southeasterly over Korean peninsula (Fig. 2c and d).

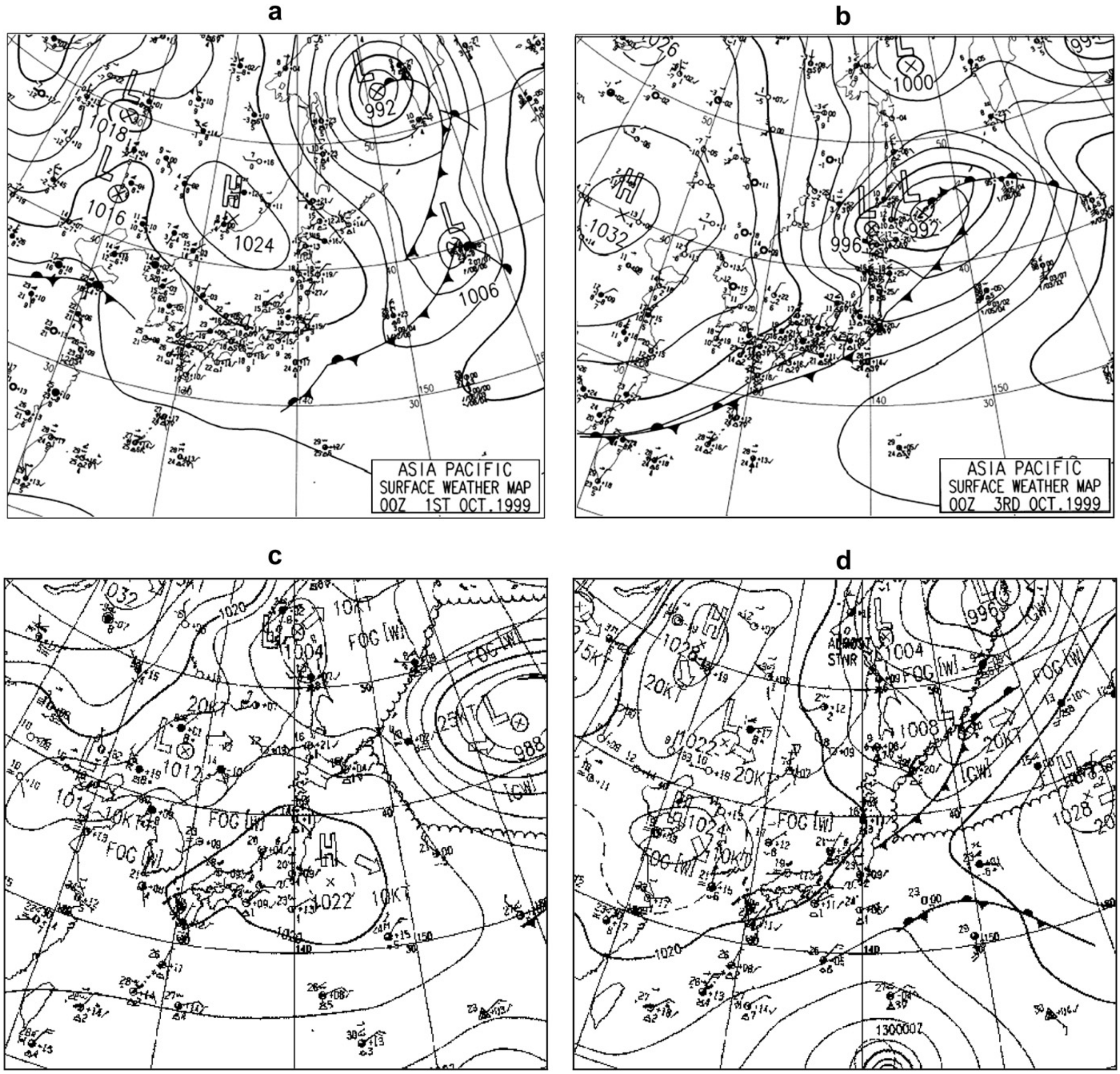


Fig. 2. Surface weather maps of (a) October 1 and (b) October 3 in 1999, and (c) October 10 and (d) October 12 in 2006.

4.2. *Simulated wind and concentration fields*

The diurnal variations of simulated wind fields by MM5 and concentration fields by LPDM with the kernel density estimator for both cases at 10 m above the ground level are shown in Fig. 3 and Fig. 4, respectively. The simulated wind fields around the JCO accidental area in Fig. 3 (left panel) prevail a weak westerly or northwesterly on September 30. The wind fields gradually change to the northerlies and north-easterlies on October 1 by veering, and then change to southerlies or south-westerlies over the accidental area on October 3 with a low-pressure center at around 145°E and 45°N (Fig. 3), implying that both pressure patterns and the features of wind fields compare favorably with those observed synoptic patterns in Fig. 2.

Fig. 3 (right panel) illustrates the horizontal distribution of ^{85}Kr at 10 m above the ground estimated by LPDM using the predicted wind and turbulent fields. On September 30 at 1200 LST (1.5 h after the initial particle being released), the simulated high concentration is mainly confined around the accidental area. Thereafter, it is continuously transported southeastward due to weak northwesterly flows on 0000 LST October 1 with the maximum value of more than 10 μBq m^{-3} (Fig. 3). The particles advected to southeastward produce relatively higher levels of ^{85}Kr at 138–141°E and 33–35°N over the eastern ocean of Japan on 1200 LST October 1 and 0000 LST October 2. As the time progresses, the released particles move to northeastward in association with the southwesterly in front of a cold front that is extending northeast to southwestwards along the eastern coastline of Japan at 1200 LST on 2 October (Fig. 3), resulting in relatively higher concentrations at the eastern coastal area of Japan. These high values of ^{85}Kr are widely extended to the eastern ocean of Japan after 0000 LST October 3.

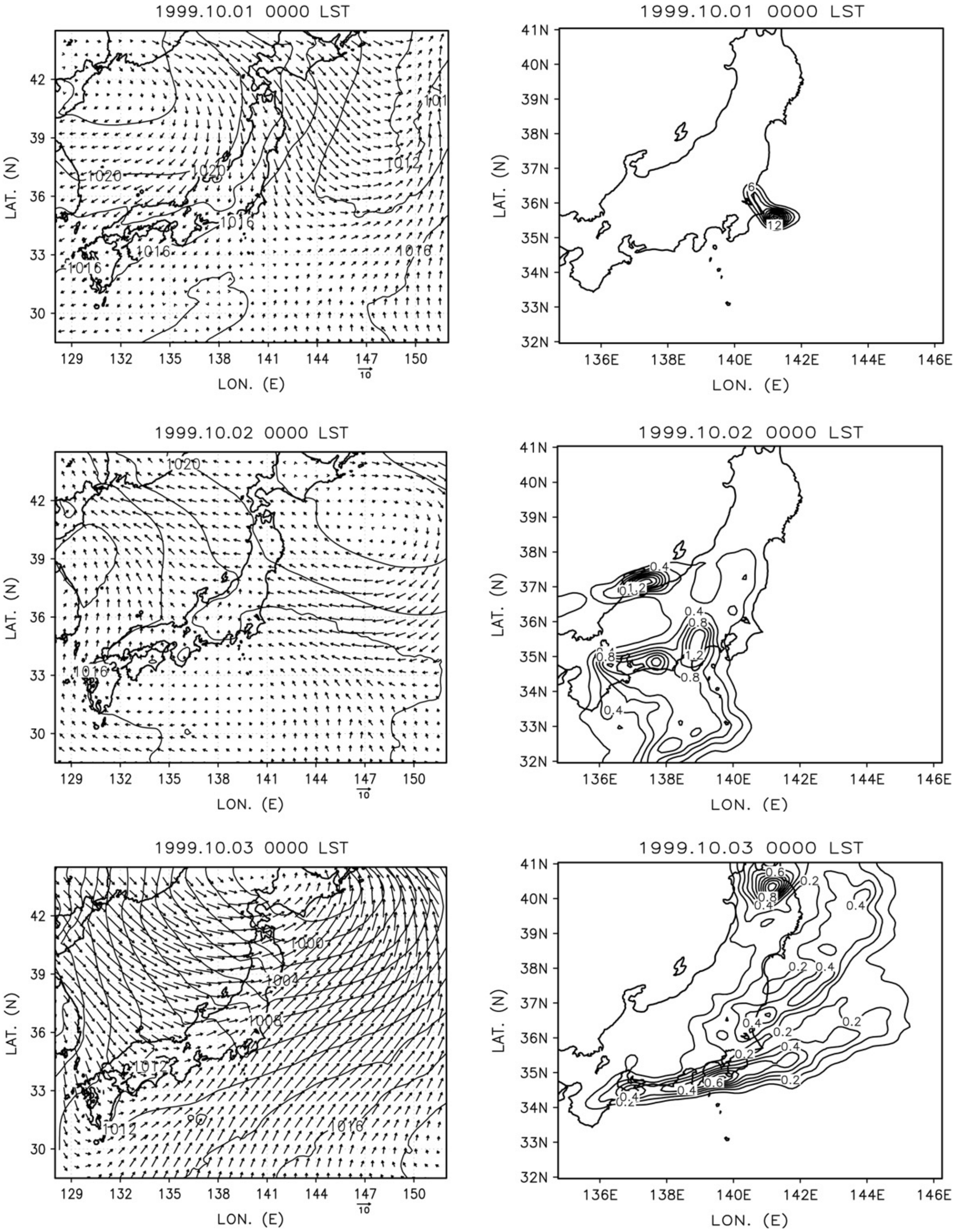


Fig. 3. Time variations of horizontal patterns of simulated surface wind and pressure fields (left panel), and ^{85}Kr (μBq m^{-3}) (right panel) at the ground level during the JCO accident from 1 to 3 October 1999.

Of particular interest during the simulation period is a fact that the secondary maximum values of ^{85}Kr are found to the southwestern area of the accidental site after 26 h from the accident, and then begin to move back to the northeast direction due to the shift of surface wind direction with time driven by the mesoscale meteorological features. This returned flow enhances the level of concentration around the accidental area.

Fig. 4 (right panel) shows the horizontal distribution of (χ/Q) at 10 m above the ground estimated by LPDM using the predicted wind and turbulent fields. Tracer particles are continuously released from the ground source located at UNT starting from 0900 LST on 10 October 2006. Due to the south-westerlies prevailing on 11 October 2006, the simulated high (χ/Q) is continuously transported northeastward around the UNT. In turn, as the wind direction changed to northwesterlies slowly on 11 October 2006, the simulated high (χ/Q) moved to southeastward with the maximum of more than order of 10^{-13} s m^{-3}. On 12 October 2006, relatively weak northlies of wind fields around downstream area of UNT

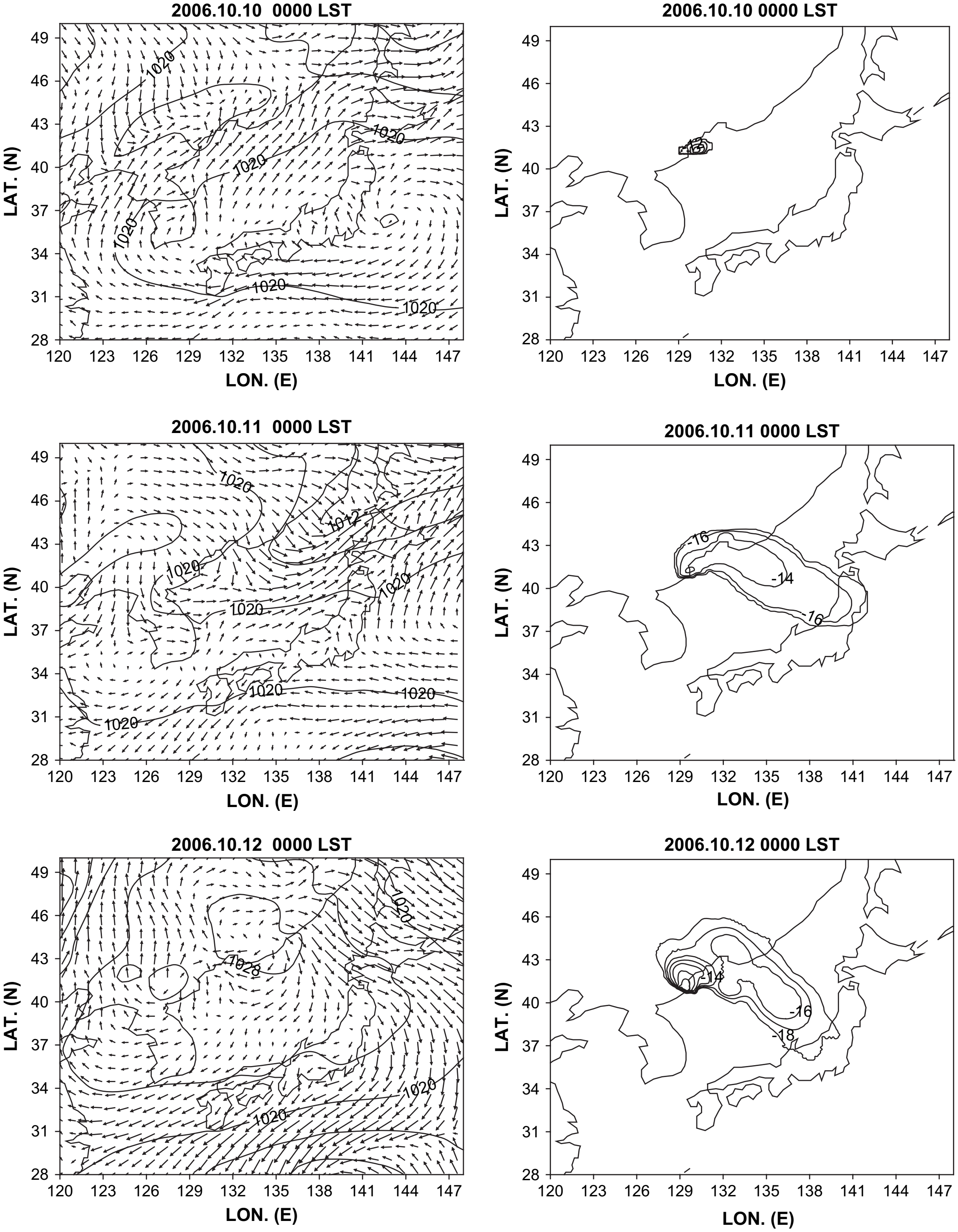


Fig. 4. Time variations of horizontal patterns of simulated surface wind and pressure fields (left panel), and dilution factors (χ/Q, unit: $\mathrm{s\,m^{-3}}$) at the ground level during the underground nuclear test (UNT) from 10 to 12 October 2006.

advected Lagrangian particles to southward producing second highest (χ/Q) zone with the (χ/Q) of $10^{-16}\,\mathrm{s\,m^{-3}}$, not extending further northeastward in accordance with, of course, mesoscale circulation (Fig. 4).

4.3. Comparison with observations

As there exist no available observations in association with UNT, we compared the simulated concentration against observation only for the case of JCO accident. In order to compare the simulated levels of ^{85}Kr with measurements for JCO accident, we have employed the daily samples of ^{85}Kr obtained at MRI (Meteorological Research Institute) in Tsukuba, which was previously described in detail by Igarashi et al. (2000a). Fig. 5a shows the measured ^{85}Kr at Tsukuba. Over the simulation period, the measured levels of ^{85}Kr were found between 1.33 and 1.38 $\mathrm{Bq\,m^{-3}}$ with relatively higher values for the period of B (1400 LST 30 September–1400 LST 1 October) that coincides with the JCO accident period. For the period of C, the measured concentration has decreased with the reported level of 1.36 Bq m^{-3}, thereafter it increased again up to 1.37 Bq m^{-3} (Fig. 5a).

The maximum level of ^{85}Kr in Tsukuba is simulated to be 6.7 μBq m^{-3} after 20 h from the JCO accident (Fig. 5b). Another significant increase in the level of ^{85}Kr has been simulated for the period of 1000 LST–2200 LST 2 October (Fig. 5b) as is the case in observations (Fig. 5a). The second maximum concentration is attributed to the returned particles in association with the local circulation for the period of 2–3 October as can be seen in Figs. 3 and 4. This temporal variation of ^{85}Kr indicates that LPDM provides more useful information on the behavior of the radioactive clouds than that from the simple Gaussian box model that does not allow us to examine the return flow effect. It is worthwhile to note that, despite the similar temporal variation of ^{85}Kr between the measured (Fig. 5a) and the simulated one (Fig. 5b), the rage of simulated concentration (order of μBq m^{-3}) is negligible compared with that of observation (order of Bq m^{-3}).

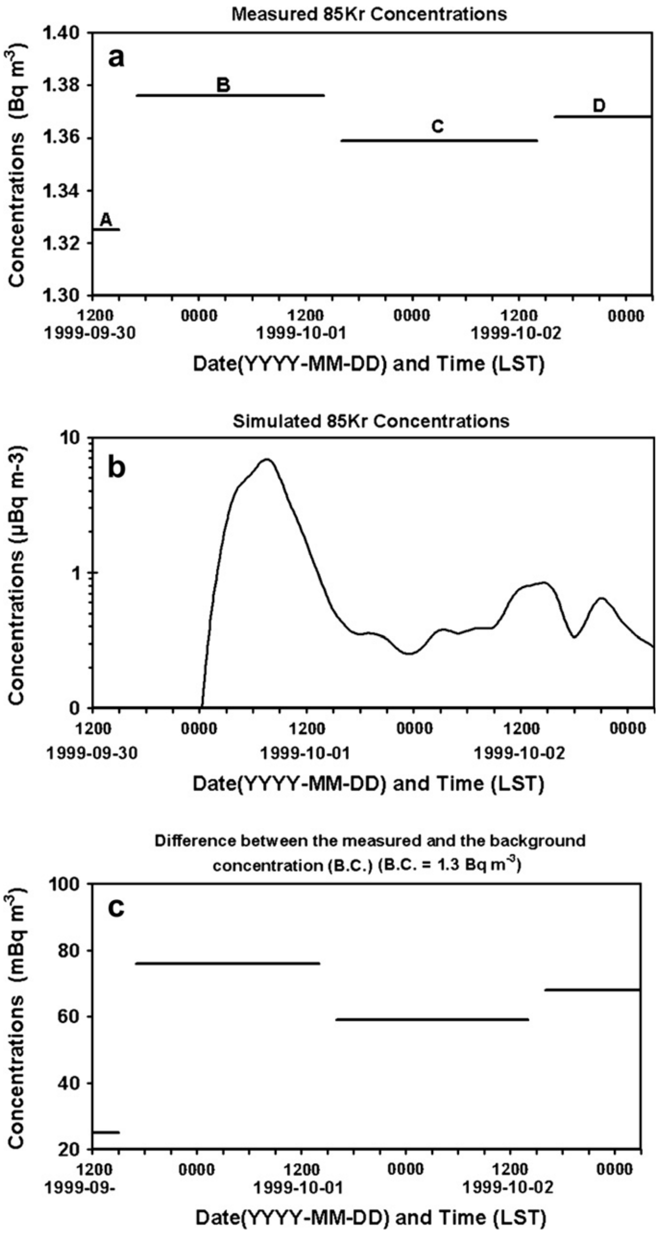


Fig. 5. Time variations of (a) observed levels of ^{85}Kr at Tsukuba located 60 km to the southeast direction from the accident place, (b) the simulated concentrations of ground level ^{85}Kr at Tsukuba, and (c) the difference between measured concentrations and background concentrations (1.3 Bq m^{-3}) of ground level of ^{85}Kr at Tsukuba. Here, (a)–(d) in the top panel represent daily averaged levels of ^{85}Kr.

The ^{85}Kr, mostly originated from nuclear fuel reprocessing plants, has spread out over the globe due to the long radioactive lifetime (half life is 10.8 years). The background ^{85}Kr at global level in the northern hemisphere is reported to be about 1.3 Bq m^{-3} and is still increasing with time world-widely (WMO, 1996; Igarashi et al., 2000b). Therefore, in this study, the background level (1.3 Bq m^{-3}) of ^{85}Kr needs to be subtracted from the observations to explore the actual effects of JCO criticality accident.

Fig. 5c shows the subtracted observation of ^{85}Kr, which is the measured values exceeding the background concentration (1.3 Bq m^{-3}). The range of observed exceedances or anomalies varies from 25 to 76 mBq m^{-3}, thereby producing the maximum difference of 51 mBq m^{-3} between A and B period. However, the simulated exceedances with the maximum of 6.71 μBq m^{-3} (Fig. 5b) are on average four orders of magnitude smaller than those observed at Tsukuba (Fig. 5c). Therefore, the previously estimated total number of fission events of 1–4 × 10^{18} yields the total emission ranging from 0.59 to 1.64 × 10^{7} Bq of ^{85}Kr during the criticality accident was not found to contribute at all to the observed temporal variation of the anomaly in Fig. 5c.

5. Discussion and possible interpretation on the discrepancies

In an attempt to explain the discrepancies and to simulate the observed actual anomaly of 51 mBq m^{-3} at Tsukuba during the JCO accident, we carried out additional bogus simulation based on the documented emission strength of ^{85}Kr from other major sources around Tsukuba. Igarashi et al. (2000a) have reported that the release rate of ^{85}Kr from the Tokai plant ranged from 0.22 to 1.39 GBq h^{-1} exponent (1.9 × 10^{4}–1.2 × 10^{5} GBq day^{-1} exponent) during the normal operation period in 1995 and 1996. Although the operation at Tokai plant stopped since 1997 and unfortunately there exits no quantitative information on Tokai plant since late-1990s is available, we carried out LPDM based on the maximum emission rate of 1.39 GBq under the same meteorological conditions used for the case of the JCO criticality accident. The result shows that, over the period of B (JCO accident period), the simulated maximum ^{85}Kr was found to be 10.3 Bq m^{-3} with relatively higher values for the period of D. This result is implying the sufficient emission strength for simulating the observations in Fig. 5c, making the assumption that the sources of ^{85}Kr with the strength of 1–2 GBq from around the Tsukuba can be one of the crucial influences to address the level of observations indicated in Fig. 5c.

One possible factor to explain the aforementioned discrepancies of simulation vs. observations of ^{85}Kr is a lack of emission sources located near JCO. There might also be other major local sources during the JCO accident such as small facilities around the JCO or outside of accidental area. Now taking into account of this surrounded emission gave rise to very large underestimations of the fission products in and around JCO. This means that under the assumption of no major local sources by which the Tsukuba observations can be biased, at least the influence of JCO accident does not show any detectable anomalies unless the emission is unrealistically assumed to be excessively higher.

The next possible factor is transport of the ^{85}Kr from higher concentration regions over the upstream region such as somewhere over Northern part of the Tsukuba at the time of accidental site. According to the report of Igarashi et al. (2000b), Tokai plant accounted for about 3% of the world-wide release of ^{85}Kr in mid-1990s, and a commercial reprocessing plant without any reducing measure in Aomori (in Fig. 1) is expected to be potentially accounted for up to 20%. Although Aomori plant was under construction at the time of JCO accident, the virtual emission strength corresponding to that of Aomori plant was simply considered in this study to get the approximate estimate of emission strength over Northern outside of Tsukuba. Under the crude assumption of the reported emission ratio of Aomori plant (20%) to Tokai plant (3%) as a potential emission strength, the virtual emission strength of 9.3 GBq ($=1.39 \times 20/3$ GBq) at Aomori plant was simply used. The simulated maximum ^{85}Kr was found to be 70 mBq m^{-3} over the period of B (JCO accident period), yielded very reasonable result in comparison with the observations in Fig. 5c. Therefore, this is suggesting that the possible emission strength of upstream region should be approximately on the order of several GBq, significantly higher values than estimated emission strength from JCO accident.

The next possible interpretation is in association with the levels of background concentrations' fluctuations such as annual or seasonal variations. Previous studies pointed out that the observed annual increase rate of background concentration is about 33 mBq m^{-3} based on the long-term measurements of ^{85}Kr. The seasonable variations also show higher fluctuations with the amplitude of approximately 200 mBq m^{-3} mainly due to the origin of air mass which is also closely related to the latitudinal trend of atmospheric ^{85}Kr (Levin and Hesshaimer, 1996). In summer maritime air mass formed at relatively lower latitudes with the lower levels of ^{85}Kr is brought to the inland, reducing the background concentrations, whereas the continental air mass formed at higher latitude is dominant, and as a result the background concentrations can be enhanced.

Likewise, the regional or global scale transport of ^{85}Kr into the analysis domain might explain the temporal variations of ^{85}Kr to some extent. The transport of higher background concentrations due to the bringing-in of continental air mass which is originating from the region of relatively higher latitude can increase the level of ^{85}Kr. This is also consistent with the meteorological features in Fig. 3. In Fig. 3, the northwestlies or northerlies was dominant over Japan from 1200 LST September 30 to 1200 LST October 1, implying the transport of background concentration with higher levels of ^{85}Kr.

6. Summary and conclusion

In an attempt to choose a dispersion model, it is important to consider the model formulation errors always arising from inadequate or incorrect representation of the atmospheric dispersion physics. In this study, we employed a three-dimensional atmospheric dispersion modeling system consisting of the Lagrangian particle dispersion model (LPDM) and prognostic mesoscale meteorological model MM5, and applied LPDM to two different simulations to assess the impact of the mesoscale meteorological conditions.

The first case of JCO criticality accident in Japan showed that the simulated anomaly of ^{85}Kr in Tsukuba located at 60 km away from the accidental area is found to be on the order of less than 10 μBq m^{-3} with the primarily peak of 6.7 μBq m^{-3}, and the LPDM simulated the secondary peak of 0.56 μBq m^{-3} after 2 days of the JCO accident transported by local or regional meteorological circulations. Thus it is shown that the LPDM quantitatively simulates the behaviors of released particles in accordance with the local circulations, suggesting the effectiveness of the simulation of hazardous materials in order to support the real time decision-making process in the case of emergencies. However, the range of reported total emissions from the JCO accident ranging from 0.59 to 1.64×10^7 Bq of ^{85}Kr shows significant deviations of simulated concentrations in comparison with the observations, indicating no detectable evidence from JCO accident to explain the variations of anomalies observed in Tsukuba. However, those discrepancies might be attributable to the transport of relatively higher ^{85}Kr background concentrations to Tsukuba by regional scale atmospheric circulations. More precise information on emission source and in detail the measurement of noble gas with regards to the diurnal and daily variations were warranted in order to better simulate and interpret the influence of accidental release such as JCO accident.

The second simulation of atmospheric dilution factors (χ/Q) during the UNT performed by North Korea showed that although the available observation does not exit to compare the model simulation, the time-dependent dilution factors were simulated well in accordance with the mesoscale meteorological features.

The information gained from the present study mainly serves the more practical purpose of testing the LPDM modeling for emergency response situations. However, apart from the purpose of this study, we also concluded that more confidence could be probably gained if the present LPDM dispersion model is verified by employing additional larger scale of experiments such as ETEX for an exhaustive evaluation of medium or large-scale atmospheric transport and dispersion processes. Such efforts should be made to evaluate the simulations above the surface as well as the ground-level simulation. This knowledge would give support to further model development or correction of the current dispersion model physics for the better emergency response model.

Acknowledgment

This study was financially supported by the Research Fund Program of Research Institute for Basic Sciences, Pusan National University, Korea, 2008, Project#. RIBS-PNU-2008-0035000.

References

AIChE/CCPS, 1996. Guidelines for Use of Vapor Cloud Dispersion Models. AIChE, New York. 271pp.

Annika, P., George, T., George, P., Konstantinos, N., Costas, D., Koutitas, C., 2001. The Poseidon operational tool for the prediction of floating pollutant transport. Marine Pollution Bulletin 43, 270–278.

Arritt, R.W., 1985. Numerical studies of thermally and mechanically forced circulations over complex terrain. Ph.D. dissertation, Department of Atmospheric Science, Colorado State University, 151pp.

Endo, A., Yamaguchi, Y., Sumita, K., 2003. Analysis of time evolution of neutron intensity measured with a High-sensitive neutron collar during the JCO criticality accident. Journal of Nuclear Science and Technology 40, 628–630.

Igarashi, Y., Aoyama, M., Miyao, T., Hirose, K., Komura, K., Yamamoto, M., 1999. Air concentration of radiocaesium in Tsukuba, Japan following the release from the Tokai waste treatment plant: comparisons of observations with predictions. Application of Radiative Isotopics 50, 1063–1073.

Igarashi, Y., Mivao, T., Aoyama, M., Hirose, K., Sartorius, H., Weiss, W., 2000a. Radioactive noble gases in surface air monitored at MRI, Tsukuba, before and after the JCO accident. Journal of Environmental Radioactivity 50, 107–118.

Igarashi, Y., Sartorius, H., Miyao, T., Weiss, W., Fushimi, K., Aoyama, M., Hirose, K., Inoue, H.Y., 2000b. ^{85}Kr and ^{133}Xe monitoring at MRI, Tsukuba and its importance. Journal of Environmental Radioactivity 48, 191–202.

Imanaka, T., 2000. Neutron dose and power released by the JCO critically accident in Tokaimura. Journal of Environmental Radioactivity 50, 15–20.

Galmarini, S., Bianconi, R., Bellasio, R., Graziani, G., 2001. Forecasting the consequences of accidental releases of radionuclides in the atmosphere from ensemble dispersion modeling. Journal of Environmental Radioactivity 57, 203–219.

Gifford, F.A., 1982. Horizontal diffusion in the atmosphere: a Lagrangian dynamic theory. Atmospheric Environment 16, 505–512.

Legg, B.J., Raupach, M., 1982. Markov-chain simulation of particle dispersion in inhomogeneous flows: the mean drift velocity induced by a gradient in Eulerian velocity induced by a gradient in Eulerian velocity variance. Boundary-Layer Meteorology 24, 3–13.

Levin, I., Hesshaimer, V., 1996. Refining of atmospheric transport model entries by globally observed passive tracer distributions of ^{85}Krypton and sulfur hexafluoride (SF6). Journal of Geophysical Research 101 (D11), 16745–16755.

McNider, R.T., 1981. Investigation of the impact of topographical circulations on the transport and dispersion of air pollutants. Ph.D. dissertation, Department of Environmental Science, University of Virginia, Charlottesville.

Mitsugashira, T., Hara, M., Nakanishi, T., Sekine, T., Seki, R., Kojima, S., 2000. Passive gamma-ray spectrometry for the determination of total fission events in the

JCO criticality accident'99 in Tokai. Journal of Environmental Radioactivity 50, 21–26.

Miyoshi, Y., Yamamoto, T., Nakajima, K., Yamane, Y., 2001. Study on the static and kinetic behavior during the JCO criticality accident. In: 2001 Annual Meeting of the American Nuclear Society, Milwaukee, WI, 17–21 June 2001. Transactions of the American Nuclear Society, vol. 84.

NSC (Nuclear Safety Commision), 1989. Nuclear Safety Comission Monthly Report, vol. 12(3). Nuclear Safety Commision, Japan, pp. 107–119 (in Japanese).

Park, S.-U., 1993. Air quality simulations using near surface meteorological observations. In: Workshop on the Regional Air Quality, Seoul, Korea, 12 August 1993, pp. 93–130.

Park, S.-U., 1994. Estimation of non-reactive pollutant concentration using the Lagrangian particle dispersion model. Journal of Korean Meteorological Society 30 (1), 1–20.

Park, S.-U., 1998a. Air quality simulation using surface meteorological observation data. Journal of Korean Meteorological Society 30 (3), 419–486.

Park, S.-U., 1998b. Effects of dry deposition on near-surface concentrations of SO_2 during medium-range transport. Journal of Applied Meteorology 37, 486–496.

Park, S.-U., Kim, S.-H., 1998. Estimation of dilution factors (X/Q) using the Lagrangian particle dispersion model. Korean Journal of Atmospheric Sciences 1 (1), 51–58.

Peters, L.K., Berkowitz, C.M., Carmichael, G.R., Easter, R.C., Fairweather, G., Ghan, S.J., Hales, J.M., Leung, L.R., Pennell, W.R., Potra, F.A., Saylor, R.D., Tsang, T.T., 1995. The current state and future direction of Eulerian models in simulating the tropospheric chemistry and transport of trace species – a review. Atmospheric Environment 29, 189–222.

Saltbones, J., Fodd, A., Bartnicki, J., 1998. Norwegian meteorological institute's real-time dispersion model snap (severe nuclear accident program): runs for ETEX and ATMES II Experiments with different meteorological input. Atmospheric Environment 32 (24), 4277–4283.

Smith, F.B., 1968. Conditioned particle motion in a homogeneous turbulent field. Atmospheric Environment 2, 491–508.

Song, C.-K., Kim, C.-H., Lee, S.-H., Park, S.-U., 2003. A 3-D Lagrangian particle dispersion model with photochemical reactions. Atmospheric Environment 37, 4607–4623.

STA (Science and Technology Agency), 1999. About the criticality accident of JCO Tokai works. In: Reference Material 1 at the 66th Meeting of the Nuclear Safety Commission on November 4, 1999. Science and Technology Agency, Japan (in Japanese).

Stohl, A., 1998. Computation, accuracy and applications of trajectory – a review and bibliography. Atmospheric Environment 32, 947–966.

Tanaka, S., 2001. Summary of the JCO criticality accident in Dokaimura and a dose assessment. Journal of Radiation Research 42 (Suppl.), S1–S9.

Venkatram, A., 1988. Structure of the PBL. In: Venkatram, A., Wyngaard, J.C. (Eds.), Lectures on Air Pollution Modeling. American Meteorological Society, Boston, MA, p. 402.

Yamada, T., Bunker, S., 1986. Development and application of a nested grid mesoscale model for transport and diffusion over complex terrain. In: AMS/APCA 5th Joint Conference on Air Pollution Meteorology, pp. 18–21.

WMO, 1996. World Meteorological Organisation. Environmental pollution monitoring and research programme report series no. 109. Report of an Expert Consultation on ^{85}Kr and ^{222}Rn: Measurements, Effects and Applications, Freiburg, Germany, 28–31 March 1995.