A random walk model to simulate the atmospheric dispersion of radionuclide

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Abstract. To investigate the atmospheric dispersion of radionuclide in large-medium scale, a numerical simulation method based on random walk model for radionuclide atmospheric dispersion was established in the paper. The route of radionuclide migration and concentration distribution of radionuclide can be calculated out by using the method with the real-time or historical meteorological fields. In the simulation, a plume of radionuclide is treated as a lot of particles independent of each other. The particles move randomly by the fluctuations of turbulence, and disperse, so as to enlarge the volume of the plume and dilute the concentration of radionuclide. The dispersion of the plume over time is described by the variance of the particles. Through statistical analysis, the relationships between variance of the particles and radionuclide dispersion characteristics can be derived. The main mechanisms considered in the physical model are: (1) advection of radionuclide by mean air motion, (2) mixing of radionuclide by atmospheric turbulence, (3) dry and wet deposition, (4) disintegration. A code named RADES was developed according the method. And then, the European Tracer Experiment (ETEX) in 1994 is simulated by the RADES and FLEXPART codes, the simulation results of the concentration distribution of tracer are in good agreement with the experimental data.

1. Introduction
Radionuclide can be released into the atmosphere in the form of gases or particles after nuclear accident. The spatial distribution of radionuclide radiation doses and the change of time have important significance to formulate and implement protective measures to reduce public radiation hazards. The diffusion behaviour of radionuclide, as a special air pollutant, mainly depends on the average wind speed and turbulence of atmosphere. Wind field leads to space displacement of radionuclide in the atmosphere, and radionuclide is mixed with the ambient air by turbulent motion. Most existing atmospheric diffusion models are established based on gradient transport theory, turbulence statistics theory and similarity theory, such as analytical solution derived from turbulence statistic - Gaussian model. The model has clear physical concept, which is more suitable for calculation of medium and small scale scope in steady state flow field. It has high computation efficiency and spatial resolution, which is still one of the most popular models at present. For example, Gaussian plume model is adopted for AERMOD model [1], ADMS model [2] and HotSpot program [3], etc. to simulate the radionuclide transport diffusion. However, because Gaussian model is based on stable and uniform conditions, it is determined that it only applies to radionuclide diffusion simulation of local scope generally [4, 5].
Atmospheric turbulence has a high degree of randomness. Therefore, the diffusion behavior of air pollutants in the atmosphere can be simulated by a random walk method. Namely, random motion of a large number of particles is tracked to simulate the diffusion process of pollutants in the air [6-8]. The random walk method for simulating atmospheric diffusion process of air pollutants reflects the random nature of turbulent diffusion, which not only can simulate the atmospheric diffusion of pollutant in homogeneous turbulence field [9-11], but also can be applied to simulate pollutant diffusion problem under non-uniform, unsteady and strong shear complex flow field condition [12, 13]. Currently, some scholars have tried to apply the random walk method to radionuclide atmospheric diffusion simulation [14, 15]. In China, Zheng, Yan Zheng, etc. use the random walk method to simulate the radiation of nuclear accident radioactive plume in small scale [16]. Establishment of medium and large scale radionuclide atmospheric diffusion calculation model and verification research on the reliability of the model calculation results has not been reported at home. In the paper, a calculation model suitable for radionuclide diffusion in large and medium scope is established based on the random walk method. Radionuclide atmospheric diffusion simulation program RADES is developed. The program is used for simulating and calculating RADES (ETEX) process. The calculation results are compared with measured results, and the correctness and reliability of the program are verified and analyzed.

2. Calculation model

In the random walk method, each radionuclide particle is regarded as an independent identifying particle. The trajectory of the particle is calculated by releasing a large number of particles. The particles are applied for describing the migration and diffusion of radionuclide in the atmosphere. Particles are transported in the flow field according to average wind. Meanwhile, a series of random displacements are used for simulating turbulent diffusion. Two functions of advection and turbulent diffusion are described. Finally, the distribution of radionuclide is estimated through overall distribution of the particles in space and time. The particle motion trajectory can be written into the following form by integral particle motion equation:

\[ X(t + \Delta t) = X(t) + V' \left( X(t), t \right) \Delta t + V'' \left( X(t), t \right) \Delta t \]

(1)

Wherein \( X \) refers to the three-dimensional coordinate component (x, y, z) of the particle; \( V' \) is the average wind velocity component (\( u', v', w' \)); \( V'' \) velocity component (\( u'', v'', w'' \)) of the turbulent pulsation; \( t \) is the time series; \( \Delta t \) is step for time. The pulsation speed of each time step is assumed by assuming that the motion obeys Markov (n + 1 moment is only associated with the n time), namely [18]:

\[
\begin{align*}
    u'(t + \Delta t) &= u'(t) + R_u \left(1 - R_u^2\right)^{1/2} \sigma_u \xi \\
    v'(t + \Delta t) &= v'(t) + R_v \left(1 - R_v^2\right)^{1/2} \sigma_v \xi \\
    w'(t + \Delta t) &= w'(t) + R_w \left(1 - R_w^2\right)^{1/2} \sigma_w \xi
\end{align*}
\]

(2)

In the two formulas of turbulent velocity components, the second item on the right represents the random part in the velocity fluctuation. \( \xi \) is the random number in line with the normal distribution (the mean value is 0 and the standard deviation is \( \sigma \)). \( \sigma_u = (\overline{u'^2})^{1/2} \), \( \sigma_v = (\overline{v'^2})^{1/2} \) and \( \sigma_w = (\overline{w'^2})^{1/2} \) are standard deviations of the pulsation quantity \( V'' \); \( R_i(\Delta t) = \exp(-\Delta t/T_{L_i}) \) Lagrange autocorrelation function; \( T_{L_i} \) is the Lagrangian time scale, where \( i \) represents three direction components of \( u, v, \) and \( w \).

It is obvious that the implementation of the random walk diffusion simulation has the key of determining the boundary layer turbulence diffusion parameters \( \sigma_i \) and \( T_{L_i} \). These parameters can be obtained by analyzing the real-time meteorological observation results. The calculation method is
closely related to the stability of the atmosphere [19, 20]. Concrete calculation method is shown as follows:

1) The atmospheric stability is unstable.

The standard deviation of turbulence velocity is shown as follows:

\[
\sigma_u = \sigma_v = u^* \times \left(12 + \frac{H}{2L}\right)^{1/3}
\]  (3)

\[
\sigma_u = \begin{cases}
\omega^* \times 0.96 \left(3 \frac{z}{H} - \frac{L}{H}\right)^{1/3} & \frac{z}{H} < 0.03 \\
\omega^* \times \min \left[0.96 \left(3 \frac{z}{H} - \frac{L}{H}\right)^{1/3}, 0.763 \left(\frac{z}{H}\right)^{0.175}\right] & 0.03 \leq \frac{z}{H} < 0.4 \\
\omega^* \times 0.722 \left(1 - \frac{z}{H}\right)^{0.207} & 0.4 \leq \frac{z}{H} < 0.96 \\
\omega^* \times 0.37 & 0.96 \leq \frac{z}{H} < 1.0
\end{cases}
\]  (4)

Lagrange time scale:

\[
T_L = T_L = 0.15 \times \frac{H}{\sigma_u}
\]  (5)

\[
T_L = \begin{cases}
0.1 \times \frac{z}{\sigma_u} \times \frac{1}{0.55 + 0.38 \times \frac{z - z_0}{L}} & \frac{z}{H} < 0.1, \frac{z - z_0}{L} < 1 \\
0.59 \times \frac{z}{\sigma_u} & \frac{z}{H} < 0.1, \frac{z - z_0}{L} \geq 1 \\
0.15 \times \frac{H}{\sigma_u} \left[1 - \exp \left(-\frac{5z}{H}\right)\right] & \frac{z}{H} \geq 0.1
\end{cases}
\]  (6)

2) The atmospheric stability is stable.

The standard deviation of turbulence velocity is shown as follows:

\[
\sigma_u = \sigma_v = u^* \times 1.3 \times \left(1 - \frac{z}{H}\right)
\]  (7)

\[
\sigma_u = u^* \times 2 \times \left(1 - \frac{z}{H}\right)
\]  (8)

Lagrange time scale:
\[ T_{Lu} = 0.15 \times \frac{H}{\sigma_u} \times \left( \frac{z}{H} \right)^{0.5} \]  
(9)

\[ T_{Lv} = 0.07 \times \frac{H}{\sigma_v} \times \left( \frac{z}{H} \right)^{0.5} \]  
(10)

\[ T_{Lw} = 0.10 \times \frac{H}{\sigma_w} \times \left( \frac{z}{H} \right)^{0.5} \]  
(11)

3) The atmospheric stability is neutral.

The standard deviation of turbulence velocity is shown as follows:

\[ \sigma_u = 2.3 \times u^* \]  
(12)

\[ \sigma_v = 2.0 \times u^* \]  
(13)

\[ \sigma_w = u^* \times 1.3 \times \exp \left( \frac{-2z}{H} \right) \]  
(14)

Lagrange time scale:

\[ T_{Lu} = T_{Lv} = T_{Lw} = \frac{0.5z}{\sigma_u} \times \frac{1}{1 + 15z/H} \]  
(15)

In the above formula, \( H \) is the boundary layer height, \( L \) is the length of Moring, \( \omega^* \) is the convective velocity, \( z_0 \) is the surface roughness, and \( u^* \) is the friction velocity. The motion trajectory of the particles can be completely determined by the number of turbulent diffusion parameters \( \sigma_i \) and \( T_{Li} \) under the above atmospheric stability conditions.

In addition, radionuclide particles suffer from the influence of decay, precipitation and other removal effects in the migration process of radionuclide particles, and the particle weight \( \omega \) will be changed as follows:

\[ \omega = \exp(-\Lambda t) \exp(-\beta t) \]  
(16)

Wherein, \( \Lambda \) refers to precipitation scavenging coefficient, and \( \beta \) refers to a decay constant.

If total radionuclide release is \( Q \), and total simulated particles are \( N \), then the total weight \( \sum_{j=1}^{n} \omega_j \) of the particles in each recording grid is calculated at the time of \( t \), and the concentration distribution of radionuclide at \( t \) moment is obtained:
$C_i = \frac{Q \sum_{j=1}^{n} \omega_j}{N \Delta V}$

(17)

Wherein, $i$ represents the record grid number, $n$ is the number of particles in a corresponding grid, $\omega_j$ is the weight of the $j$th particle in the grid, and $\Delta V$ represents the grid volume. In practical application, the time integral concentration is generally used instead of the instantaneous concentration in order to reduce the fluctuation of the calculation results:

$C_i = \frac{Q \sum_{j=1}^{N} \omega_j T_{i,j}}{N \Delta V}$

(18)

Wherein, $T_{i,j}$ the residence time of the $j$th particle in $i$th recording grid.

The above method is used for developing radionuclide atmospheric diffusion simulation program RADES. The procedure flow is shown in figure 1.

### 3. Calculation results and analysis

INSAG under IAEA cooperated with WMO for two large-scale particle tracer experiments [21, 22] in order to establish a set of calibration database which can be used as a planetary boundary layer atmospheric diffusion model. They are called RADES (ETEX). In the paper, two ETEX experiments are simulated and calculated in order to evaluate the reliability of RADES program. The tracer particles concentration at integral point time in each monitoring station between 0 to 90 hours after particle release is recorded. The nearest distance of the monitoring site and the release point is about 200km (Alencon site). The farthest distance is about 2000 km (Cervena site). Table 1 shows the release conditions of the two tracer experiments.

**Figure 1.** The flow chart of RADES code
Table 1. Summary of operating conditions of ETEX

| Release features          | The first tracer experiment | The second tracer experiment |
|---------------------------|-----------------------------|-----------------------------|
| Particle release position | Monterfil(48°03'30"N, 2°00'30"W) | Monterfil(48°03'30"N, 2°00'30"W) |
| Tracer particle           | PMCH                        | PMCP                        |
| Release starting time /UTC| 1994.10.23 16:00             | 1994.11.14 15:00             |
| Release ending time /UTC  | 1994.10.24 03:50             | 1994.11.15 02:45             |
| Total release /kg         | 340                         | 490                         |
| PMCH velocity/g·s⁻¹       | 7.98                        | 11.58                       |
| Meteorological condition  | No wind                     | Rain, heavy wind            |
| Wind direction            | West wind                   | West wind                   |

The concentration and measured results of the tracer particles in several typical sites are compared with the measured results as shown in fig.1 and 2. The results obtained by using the FLEXPART program are also presented in the figure. The FLEXPART is a model of atmospheric diffusion developed by Norwegian Atmospheric Research Institute, which has been widely recognized and applied in the field of atmospheric diffusion calculation [23]. It can be seen from the figure that the calculated results given by RADES and FLEXPART are consistent with the measured results in the aspect of radionuclide concentration and concentration change trend.

Figure 2. The tracer concentration at stations for tracer sampling during the first ETEX
Figure 3. The tracer concentration at stations for tracer sampling during the second ETEX

In the radionuclide diffusion event, radionuclide maximal concentration value $C_M$ and transit time $T_P$ are two important monitoring parameters in various monitoring sites. The maximum concentration and transit time are are compared with the measured results for analysis in order to further analyze the reliability of the calculated results as shown in fig.4 and 5, wherein,

$$D_{CM} = C_M^M - C_M^C$$ (19)

$$D_{TP} = T_P^M - T_P^C$$ (20)

Wherein, $C_M^M$ and $C_M^C$ are measured values and calculated values of the peak concentration of the tracer particles respectively. $T_P^M$ and $T_P^C$ are respectively the measured value and calculated value of the transit time. The y coordinate in the figure represents monitoring site number, figure (a) shows the comparison of the calculated result with the measured value of 49 monitoring sites in the first ETEX experiment. Figure (b) shows the comparison of the calculated results with measured values of 25 monitoring sites in the second ETEX experiments.
Figure 4. A comparison of maximum trace concentration in stations for tracer sampling. (a) for the first ETEX and (b) for the second ETEX.

Figure 5. A comparison of trace pass-time in stations for tracer sampling. (a) for the first ETEX and (b) for the second ETEX.

The average distance normal between the calculated value and the measured value is defined as follows in order to measure the consistency between the program calculated results and the measured result more intuitively:

$$
\| \alpha \| = \left[ \left( x_M^1 - x_M^C \right)^2 + \left( x_M^2 - x_M^C \right)^2 + \cdots + \left( x_M^n - x_M^C \right)^2 \right]^{1/2} / k
$$

Wherein, \( x_M^i (i = 1, 2, \cdots, k) \) is the monitoring value of the tracer particle peak concentration on the \( i \)th measurement site. \( x_M^C (i = 1, 2, \cdots, k) \) is the calculated value of the peak concentration on the \( i \) measurement site. \( k \) is the total number of monitoring sites. The average distance norm can be used to represent the overall fitness of the calculated results and the measured results. The average distance normal is smaller, the calculated results are more consistent with the measured results. Table 2 shows the average distance norm of the peak value concentration and transit time calculated time corresponding to the measured results. The average distance norms of RADES and FLEXPART given in the table are compared. It is obvious that the average distance normal of the RADES calculated result is smaller in both the first and the second ETEX experiments. It is obvious that the calculated result given by RADES is more consistent with the experiment result, thereby verifying that the calculation model established in the paper based on random walk method is correct and effective in radionuclide atmospheric diffusion. In addition, the calculated result of the second ETEX experiment given by two programs are greatly
different from the experiment data compared with the calculated result in the first ETEX experiment. It is obvious that the error of two calculation program calculated result and measured value is always larger than the calculated error under calm weather conditions in case of heavy wind and rain as well as other extreme weather conditions.

Table 2. The mean radial distance norm given by RADES and FLEXPART codes

|                  | Peak value concentration | Transit time |
|------------------|--------------------------|--------------|
|                  | ETEX1 | ETEX2 | ETEX1 | ETEX2 |
| RADES            | 0.14  | 0.69  | 2.24  | 4.05  |
| FLEXPART         | 0.55  | 0.97  | 2.27  | 4.94  |

4. Conclusion

(1) In the radionuclide atmospheric diffusion numerical simulation method established based on the random walk method, real-time or historical meteorological data can be utilized for calculating the radionuclide diffusion problem in medium and large scale scope.

(2) The self-developed radionuclide atmospheric diffusion simulation program RADES is utilized for simulating the RADES process. The change condition of monitoring site concentration with time, site peak value concentration, tracer particle transit time and other parameters are given through calculation. The results show that the calculated results of RADES given at simulation of sunny and wind-free weather conditions have small deviation from the measured results. The calculated results given at simulation of extreme weather conditions-heavy wind and rain have great deviation from the measured results.

(3) Radionuclide atmospheric diffusion process is very complex, which is not only affected by the atmospheric wind field, but also is related to the source nature, diffusion area characteristics of landform and physiognomy and climate humidity in different areas. Next, these influence factors will be added in existing calculation model in order to improve the reliability of the calculated results.

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