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Radioactive gas diffusion simulation and inhaled effective dose evaluation during nuclear decommissioning

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ABSTRACT

During the decommissioning of the nuclear facilities, the radioactive gases in pressure vessels may leak due to the demolition operations. The decommissioning site has large space, slow air circulation, and many large nuclear facilities, which increase the difficulty of workers' inhalation exposure assessment. In order to dynamically evaluate the activity distribution of radionuclides and the committed effective dose from inhalation in nuclear decommissioning environment, an inhalation exposure assessment method based on the modified eddy-diffusion model and the inhaled dose conversion factor is proposed in this paper. The method takes into account the influence of building, facilities, exhaust ducts, etc. on the distribution of radioactive gases, and can evaluate the influence of radioactive gases diffusion on workers during the decommissioning of nuclear facilities.

1. Introduction

Decommissioning activities in nuclear facilities generate large amounts of radioactive gases. The radioactive gases mainly include radioactive inert gas, airborne radioactive iodine, antimony and other gases. In order to ensure the safety and reduce the internal exposure of radioactive gases to workers during the decommissioning, it is necessary to analyze and assess the distribution of radionuclides.

The nuclear decommissioning workflow is complex and numerous, and the air sampling assessment method cannot meet the assessment of radioactive gases. In order to dynamically evaluate the distribution of radionuclides and the committed effective dose from inhalation of workers, it is necessary to simulate the diffusion of radioactive gases by air modeling.

In the nuclear decommissioning scenario, the gas flow rate is low. The transport and dispersion of pollutants becomes weak under low wind conditions resulting in large ground level concentrations. Meanwhile, the nuclear decommissioning scene belongs to the indoor space, which has large space, good sealing and complex structure. The concentration and exposure of pollutants in the indoor environment are significantly higher than in ambient outdoor environment, and the indoor concentration can exceed the outdoor concentration by several orders of magnitude. In order to assess the chemical exposures in a lab, Keil et al. estimated air concentrations during distillation and extraction exercises using well mixed room model [1]. Considering the spatial variability in exposure intensity, Spencer et al. selected the near field-far field model to predict the solvent vapor concentrations in workplace [2]. Demou et al. evaluated the homogeneously mixed-one-box model, multi-zone model and eddy-diffusion model with data obtained from measurements in the indoor workplace and with field experiments using these products to simulate the same exposure conditions [3]. Drivas et al. developed an analytical indoor air dispersion model that considers the point-source dispersion with reflection from all walls and the general concentration decay in a rectangular room [4]. However, these methods do not take into account the obstacles of indoor facilities. At the same time, there are many leak sources and exhaust ducts in the nuclear decommissioning scene, which makes the simulation of the radioactive gas diffusion process more difficult.

In order to dynamically evaluate the distribution of radioactive gases in decommissioning environment, a modified eddy-diffusion model is proposed based on voxelization method and air modeling

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method. The model takes into account the influence of building, facilities, exhaust ducts, etc. on the distribution of radioactive gases, and can dynamically calculate the activity distribution of radionuclides. Taking into account the influence of radioactive gases diffusion on workers, an inhalation exposure assessment method is also proposed in this paper for estimating the committed effective dose intake via inhalation based on the modified eddy-diffusion model and the inhaled dose conversion factor.

2. Eddy-diffusion model

Based on the Fick's Law of diffusion, for a point source emitting at a steady state in an infinite space, the speed at which the emitted contaminants move through the space is directly proportional to the produce of the change in concentration with distance from the source and the molecular diffusivity [5]. In the indoor spaces, the eddy diffusion dramatically dominates molecular diffusion.

The eddy-diffusion model assumes that the transmission mechanism of indoor gas is turbulent diffusion. As shown in Fig. 1, for an elevated source, the region of this model is conveniently described by a sphere space. The standard solution to the concentration balance of a sphere of radius *r*, assumes infinite distance around source and takes the form of:

$$C_t(r) = \frac{G}{4\pi Dr}\left[1 - erf\left(\frac{r}{\sqrt{4Dt}}\right)\right] \tag{1}$$

where $C_t(r)$ is the concentration at any location *r* and any time *t* (mass/volume); *G* is the steady-state emission rate of source (mass/time); *erf* is error function; *D* is interpreted as an eddy diffusivity (area/time), which estimate by the mechanical energy balance:

$$D = \sqrt[3]{0.5 \cdot \alpha \cdot u_0 \cdot k^2 \cdot H^4} \tag{2}$$

where α is the air-exchange rate (s^{-1}); u_0 is the velocity at ventilation inlets (m/s); *k* is the von Karman's constant [6]; *H* is the height of room (m).

At steady-state conditions, the concentration is:

$$C_t(r) = \frac{G}{4\pi Dr} \tag{3}$$

As shown in Fig. 2, when the source is close to a surface such as a floor or wall, the region of this model is described by a hemispherical space. Neglecting deposition on the surface, the

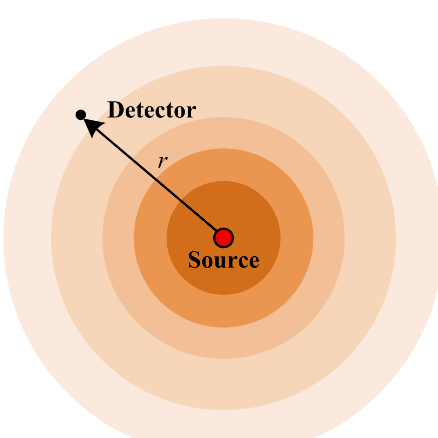

Fig. 1. The ideal eddy-diffusion model.

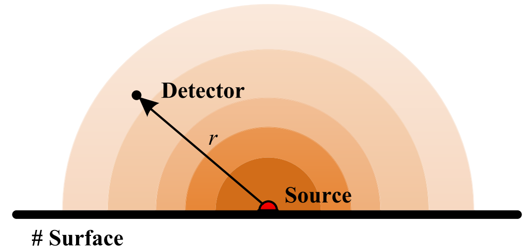

Fig. 2. The hemispherical diffusion model.

hemispherical diffusion model is used for the mass balance on a hemispherical space:

$$C_t(r) = \frac{G}{2\pi Dr}\left[1 - erf\left(\frac{r}{\sqrt{4Dt}}\right)\right] \tag{4}$$

The advantage of the eddy-diffusion model is that the concentration at different distances from the source can be calculated. The disadvantage is that it must be used with a small air velocity [7].

3. Modified eddy-diffusion model

In a radioactive factory building with limited indoor space, the ideal eddy-diffusion spherical model of radioactive gas will be affected by the scene and is not fully applicable. When the sources leak in the plant, the real concentration of the radioactive gases is larger than the concentration of ideal model due to the hindrance of the building and nuclear facilities. As shown in Fig. 3, in order to dynamically assess the concentration of indoor radioactive gases, the ideal eddy-diffusion model needs to be modified according to the plant space, nuclear facilities, exhaust pipes and other influencing factors. Based on the transmission mechanism of eddy diffusion, ignoring the surface deposition of radioactive gas on the floor, ceiling, walls and nuclear facilities, the revised eddy-diffusion model has the following two assumptions:

1) At any time, the total content of radionuclides in the gas diffusion boundary of the modified model is the same as the total content of radionuclides in the spherical boundary of the ideal eddy-diffusion model;
2) For the eddy-diffusion model with a fixed content of radionuclides in the gas diffusion boundary, if the volume inside the gas diffusion boundary changes, the concentration at each position of the modified diffusion model changes inversely with the volume change.

Based on the assumptions, for indoor space with multiple continuously leaking radioactive sources, the modified eddy-diffusion model of radioactive gas is:

$$C_{i,t}(x,y,z) = C_{i,t}(r) = \frac{V_{i,R}}{V_{i,t}} \cdot \frac{G_i}{4\pi Dr}\left[1 - erf\left(\frac{r}{\sqrt{4Dt}}\right)\right] \tag{5}$$

where *r* is the distance from source to position (*x, y, z*); at any time *t*, $C_{i,t}(x,y,z)$ is the concentration at position (*x, y, z*) of *i*-th source, and $C_{i,t}(r)$ is the concentration at location *r* (Bq/m^3); G_i is the emission rate of *i*-th source (Bq/min); $V_{i,t}$ is the volume of the puff model; $V_{i,R}$ is the volume of the ideal spherical space.

As shown in Fig. 4, in the infinite space, the uniform spherical cluster model is spherical in shape and has a volume of:

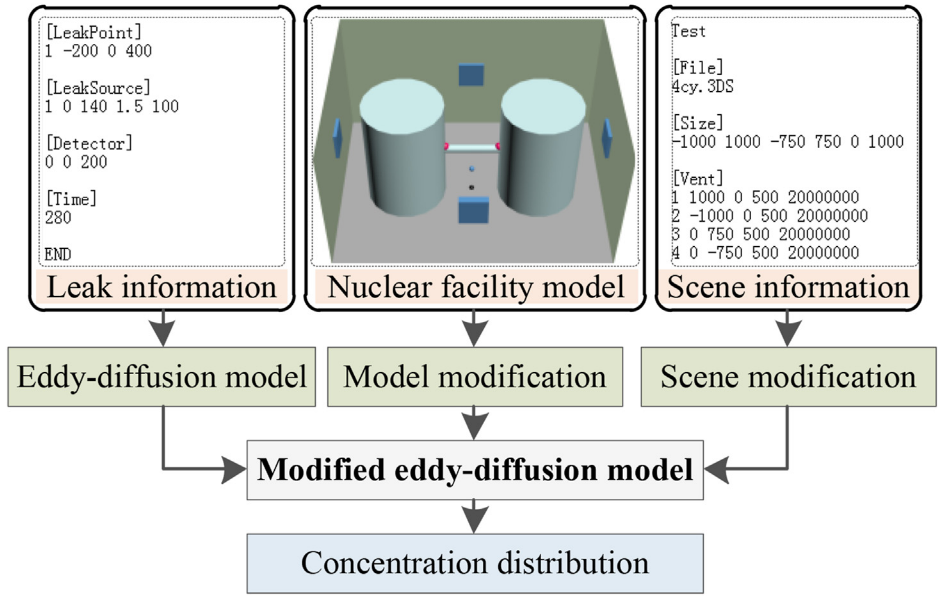

Fig. 3. Process for assessing the distribution of radioactive gas concentrations in nuclear decommissioning scenario.

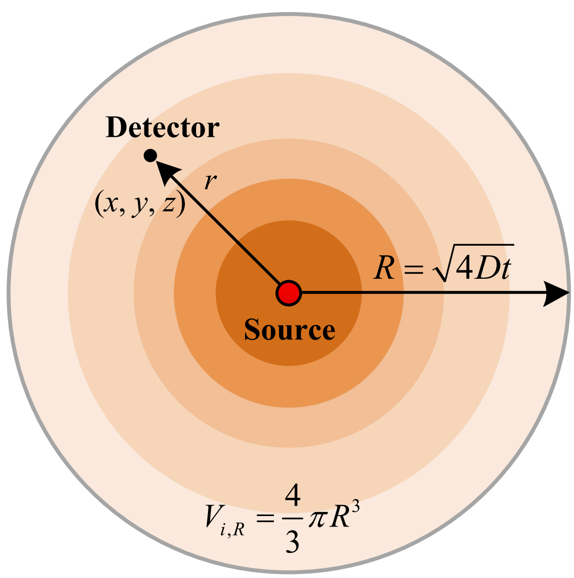

Fig. 4. The eddy-diffusion model in infinite space.

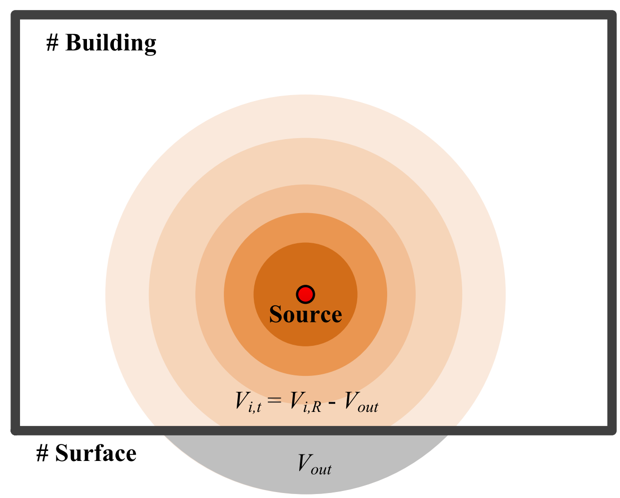

Fig. 5. The eddy-diffusion model is corrected by the building surface.

$$V_{i,R} = \frac{4}{3}\pi R^3 \tag{6}$$

where R is the radius of the outermost sphere of the model:

$$R = \sqrt{4Dt} \tag{7}$$

3.1. Obstruction of factory

In the process of radioactive gas diffusion, the indoor space of the decommissioning site can be of any shape, and the floor, ceiling and walls of the building will hinder the gas diffusion. As shown in Fig. 5, when the eddy-diffusion model is connected to the surfaces of the building, neglecting deposition on the surface, the corrected puff volume of the model is the volume of the ideal spherical space minus the volume of the spherical model outside the building surface:

$$V_{i,t} = V_{i,R} - V_{out} \tag{8}$$

where V_{out} is the volume of the spherical model outside the building surface.

For example, when the source is close to a flat surface, the region of this model is a hemispherical space ($V_{i,R} = 2V_{i,t}$), and the modified model can be described by the hemispherical diffusion model.

3.2. Obstruction of nuclear facilities

In the nuclear decommissioning scenario, the nuclear facilities are bulky, and occupy a large amount of space in the plant. In this paper, the CAD software is used to establish the nuclear decommissioning scenario. As shown in Fig. 6, the main facilities in the

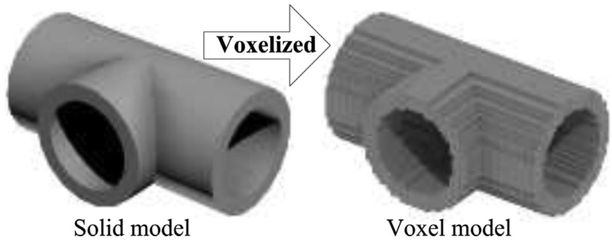

Fig. 6. Convert solid model to voxel model.

decommissioning scenario are converted into voxel models based on the voxelization method [8].

Due to the large volume of nuclear facilities in nuclear decommissioning scenarios, which will affect the diffusion of radioactive gases, it is necessary to correct the eddy-diffusion model. As shown in Fig. 7, the corrected puff volume of the model is the volume of the ideal spherical space minus the voxel volume of nuclear facilities within the spherical space:

$$V_{i,t} = V_{i,R} - V_{in} \tag{9}$$

where V_{in} is the voxel volume of nuclear facilities within the spherical space.

3.3. Influence of exhaust duct

The radioactive gas will be discharged to the outdoor through the exhaust duct, after being filtered and absorbed. When the diffusion model is connected to the exhaust duct, the radioactive gas is discharged to the outside, which means that the volume of the diffusion model becomes larger. As shown in Fig. 8, according to the position of the exhaust duct and the ventilation rate, the corrected puff volume is the volume of the ideal spherical space plus the volume of the radioactive gas discharged through the exhaust duct:

$$V_{i,t} = V_{i,R} + Q_k t_k \tag{10}$$

where Q_k is the ventilation rate of k-th exhaust duct (m^3/min); t_k is the contact time between the diffusion model and k-th exhaust duct.

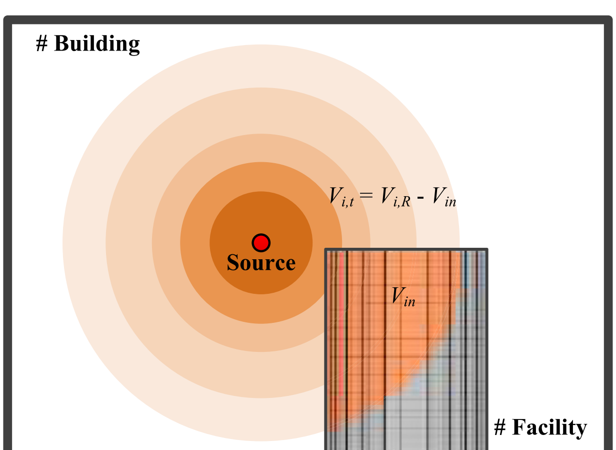

Fig. 7. The eddy-diffusion model is corrected by a nuclear facility.

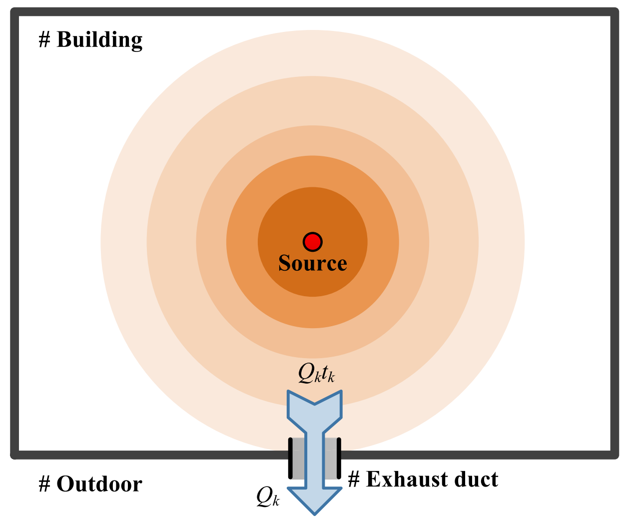

Fig. 8. The eddy-diffusion model is corrected based on the position and flow rate of exhaust duct.

3.4. Source stop leaking

The ideal eddy-diffusion model is suitable for continuous gas diffusion, and does not provide a method for calculating the indoor gas concentration after the source stops leaking. Therefore, the ideal model needs to be revised. As shown in Fig. 9, the source stops leaking at the time T. At this time, the radius of the outermost sphere of the model is R_T, the volume of the diffusion model is $V_{i,T}$, and the concentration at location r_T is $C_{i,T}(r_T)$. Based on Fick's Law of diffusion, radioactive gases continue to spread out. When the time t, the gases at location r_T spread to location r, which have:

$$\frac{r_T}{r} = \frac{R_T}{R} = \sqrt{\frac{T}{t}} \tag{11}$$

Since the total content of radionuclides in the gas diffusion boundary is constant, based on the assumption 2 of the modified model, the concentration at location r is:

$$C_{i,t}(r) = \frac{V_{i,T}}{V_{i,t}} C_{i,T}(r_T) = \frac{T}{t} \cdot \frac{V_{i,R}}{V_{i,t}} \cdot \frac{G_i}{4\pi Dr}\left[1 - erf\left(\frac{r}{\sqrt{4Dt}}\right)\right] \tag{12}$$

3.5. Impact of multiple sources

There are many leak sources in the nuclear decommissioning scenario, and the emission rates of different sources are different. As shown in Fig. 10, when multiple leak sources leak in the nuclear decommissioning scenario, the concentration of radioactive gases at a location (x, y, z) is the sum of the concentrations of all the leak sources at that location:

$$C_t(x,y,z) = \sum_i C_{i,t}(x,y,z) \tag{13}$$

where $C_t(x, y, z)$ is the concentration at position (x, y, z) and time t of all source (Bq/m^3).

4. Committed effective dose from inhalation

In order to dynamically estimate the internal exposure of worker when the radionuclides leak, an inhaled dose conversion

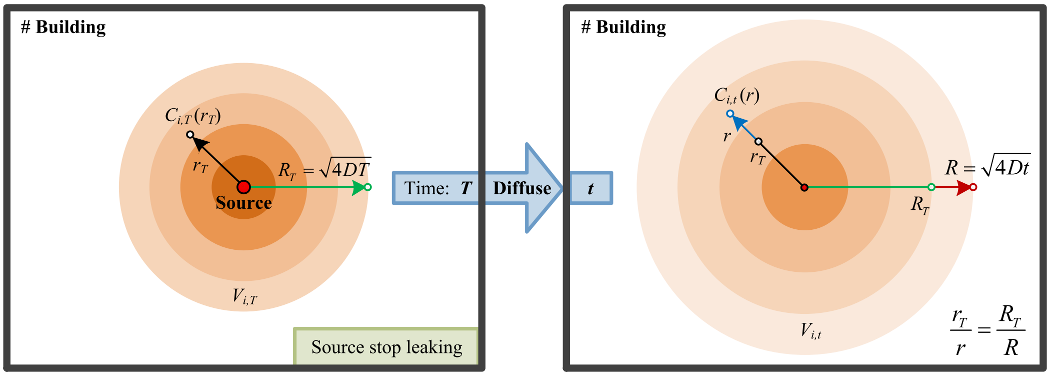

Fig. 9. The eddy-diffusion model is corrected when the source stop leaking.

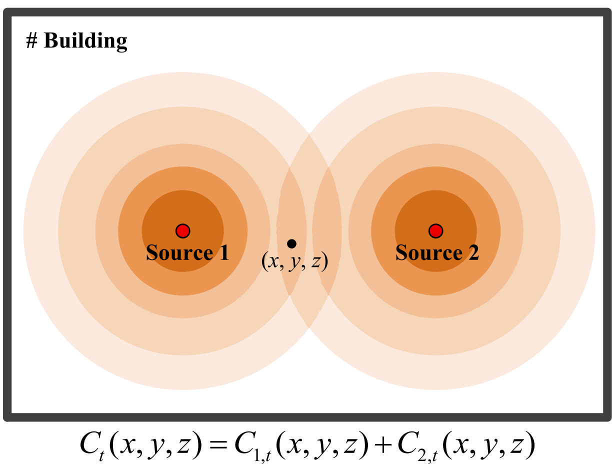

Fig. 10. The eddy-diffusion model is corrected based on the location of multiple sources.

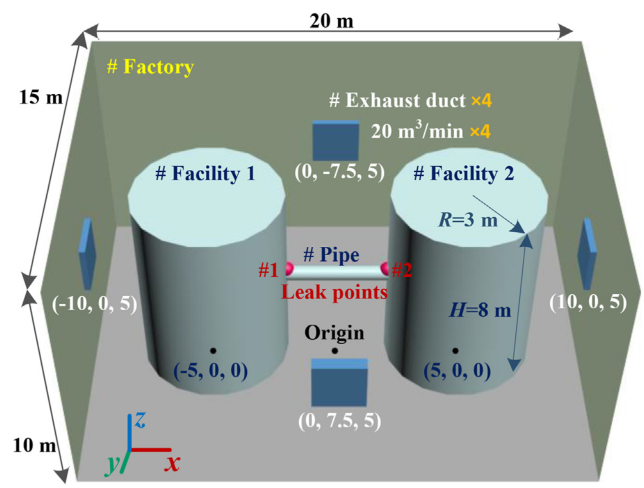

Fig. 12. A factory with two leaking cylindrical nuclear facilities.

factor method is proposed to calculate the committed effective dose due to inhalation.

Taking into account the influence of radioactive decay of radionuclides, the personal air sampling method is used to estimate the radionuclides intake via inhalation, which is calculated as follows [9]:

$$A_{j,inh} = D_j \cdot B = \int_0^{t_1} C_{t,j}(x,y,z) \cdot \frac{T_{j,1/2}}{0.693}\left(1 - e^{-\frac{0.693t}{T_{j,1/2}}}\right) dt \cdot B \tag{14}$$

where $A_{j,inh}$ is the time integral value of radionuclide j intake, Bq; D_j is the time integral concentration of radionuclide j in air, $(\mathrm{Bq}\cdot\mathrm{h})/\mathrm{m}^3$; $C_{t,j}(x, y, z)$ is the concentration of radionuclide j at position (x, y, z) and time t in room, Bq/m^3; $T_{j,1/2}$ is the half-life of radionuclide j, h; t_1 is the time of exposure to plume, h; B is the breathing rate of worker, m^3/h.

The committed effective dose intake via inhalation of radionuclide is:

$$E_{inh} = \sum_{j=1}^{n} E_{j,inh} = \sum_{j=1}^{n} A_{j,inh} e(g)_{j,inh} \tag{15}$$

where E_{inh} is the committed effective dose from inhalation, Sv; $E_{j,inh}$ is the committed effective dose of radionuclide j from inhalation, Sv; $e(g)_{j,inh}$ is the inhaled dose conversion factor of radionuclide j, Sv/Bq.

This method can select different inhaled dose conversion factor according to different categories of people, age, activity median aerodynamic diameter, and lung absorption category. When the specific parameter information is known, the estimated result of inhaled dose is more accurate.

As shown in Fig. 11, based on the modified eddy-diffusion model and the inhaled dose conversion factor, a code named as PASM

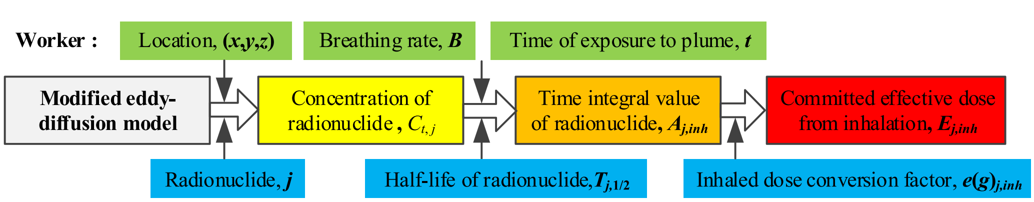

Fig. 11. The flow chart of PASM.

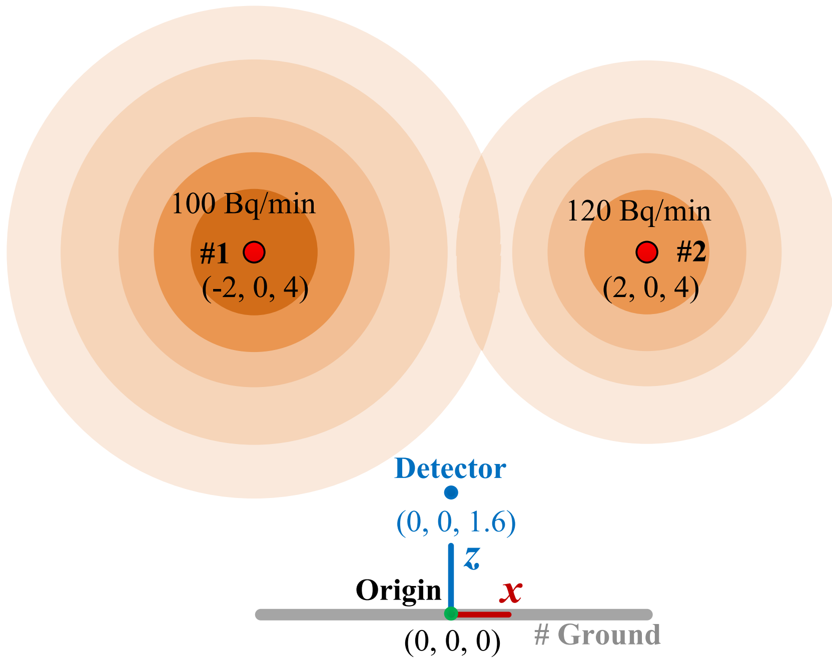

Fig. 13. Gas leakage scenario for ideal eddy-diffusion model.

implemented with C++ was developed for estimating the committed effective dose from inhalation. The PASM uses the radionuclide concentration calculated by the modified eddy-diffusion model as input data, and dynamically calculates the committed effective dose based on the worker's location and residence time.

5. Tests and results

As shown in Fig. 12, a nuclear fuel reprocessing plant filled with air was designed for radioactive gas diffusion, and the size unit used in the scenario is m. The situation is a 20 m × 15 m × 10 m factory with two same size cylindrical nuclear facilities. The radius of the cylindrical facility is 3 m, and height is 8 m. The origin of the factory is on ground in the middle of the room. Two nuclear facilities storing spent fuel are placed at (−5, 0, 0) and (5, 0, 0) respectively, and are connected by a leaking thin pipe located 4 m above the ground. The obstruction of the pipe on gas diffusion is ignored. The ^{244}Cm in the spent fuel has a long half-life and will spontaneously fission to produce ^{131}I. Due to the rupture of the pipeline, the radionuclide ^{131}I leaked from two connection points of pipe with a half-life of 8.04 days. The leak point 1 is located at (−2, 0, 4), and the leak point 2 is located at (2, 0, 4). The gas leakage at the point 1 occurred at a rate of 100 Bq/min. After the leakage at the point 1 during 100 min, the leakage at the point 2 began at a rate of 120 Bq/min. The leakage at the point 1 could stop at 140 min and that at the point 2 could stop at 180 min. The flow rate of each exhaust duct is 20 m^3/min, and there are four exhaust ducts, which are respectively

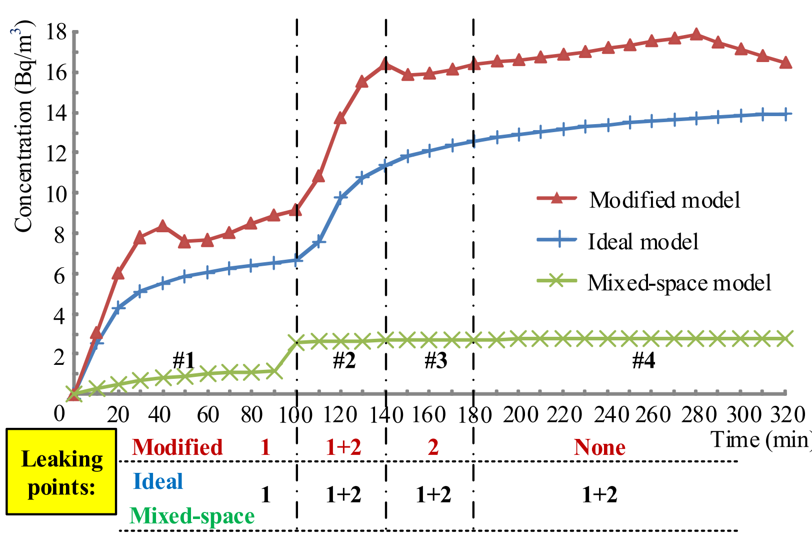

Fig. 14. Concentration of modified eddy-diffusion model, ideal eddy-diffusion model and mixed-space model.

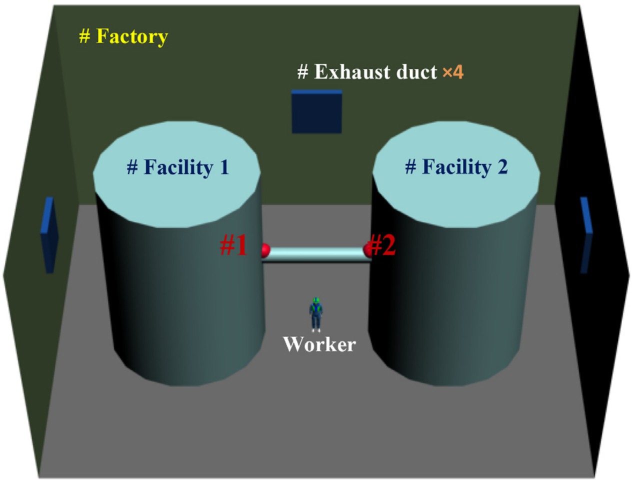

Fig. 15. A nuclear decommissioning leak scenario with a worker.

placed on the surrounding walls at (−10, 0, 5), (10, 0, 5), (0, 7.5, 5) and (0, −7.5, 5). Estimate by the mechanical energy balance, the diffusivity is 0.414 m^2/min.

5.1. Radiation gas diffusion

As shown in Fig. 13, since the applicability of the ideal eddy-diffusion model has been proven in many related papers, the ideal eddy-diffusion model and the mixed-space model under the same situations are used as control of modified eddy-diffusion model. In the comparison groups, the two leak points will not stop leakage.

As shown in Fig. 14, the calculation time of modified model, ideal model and mixed-space model at detector (0, 0, 1.6) is within 0.5 s, and the concentration at detector can be divided into four intervals in chronological order. Due to the limitations of the mixed-space model, in the four intervals, the gas concentration of the mixed-space model is much smaller than the ideal model and the modified model.

In interval 1, only point 1 leaked. For the ideal eddy-diffusion model, the concentration first increased dramatically and then tended to be stable. From the 10th minute onwards, the concentration of modified model was gradually higher than the ideal model because the gas diffusion was blocked by nuclear facilities and factory. From the 40th minute, the outermost boundary of the modified plume model was connected to the exhaust ducts, and the radioactive gas was discharged outside the factory building, causing the concentration to suddenly decrease and then slowly increase.

In interval 2, the point 2 also leaked. This leaded to a sharp increase in the concentration of the ideal model and the modified model.

In interval 3, the leakage at point 1 in the modified model stopped, which caused the concentration to drop suddenly, and the concentration started to increase slowly until the 150th minute.

In interval 4, the leakage at point 1 and 2 in the modified model stopped. Due to reflections from factory and nuclear facilities, the gas concentration will not decrease immediately. Until the 280th minute, the last smoke mass released by the point 2 spreads over the entire factory. As the exhaust ducts continues to pump out radioactive gas, the gas concentration of modified model begins to gradually decrease.

5.2. Committed effective dose intake via inhalation

As shown in Fig. 15, when the point 1 leaked, the worker was staying at the origin of the factory and performing light activity. The breathing height of the worker is 1.6 m, and the breathing rate is 1.5 m^3/h. The inhaled dose conversion factor of ^{131}I to worker is 1.1×10^{-8} Sv/Bq.

The change of the committed effective dose rate for worker inhalation calculated by PASM is shown in Fig. 16. When the time of exposure to plume is much shorter than the half-life of radionuclide, the committed effective dose rate calculated by PASM is positively correlated with the concentration of the radioactive gas. Therefore, the committed effective dose rate curve of PASM is consistent with the concentration curve of modified model in Fig. 14.

6. Discussion and conclusion

In order to dynamically evaluate the distribution of radioactive gases in decommissioned environment, a modified eddy-diffusion model is proposed in this paper based on the eddy-diffusion model. This modified model takes into account the influence of buildings, facilities, exhaust ducts, source stop leaking and multiple

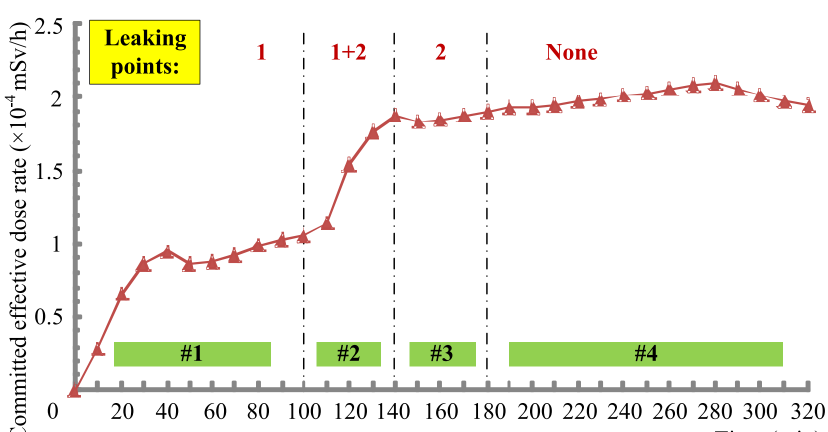

Fig. 16. Committed effective dose rate over time from inhalation.

sources on the gas diffusion, and can dynamically calculate the concentration of radioactive gases. Taking into account the influence of radioactive gases diffusion on workers, an inhalation exposure assessment method is proposed for estimating the committed effective dose intake via inhalation based on the modified eddy-diffusion model and the inhaled dose conversion factor.

In the test 1, the modified eddy-diffusion model is compared with the ideal eddy-diffusion model and mixed-space model. The results show that modified eddy-diffusion model can simulate the diffusion of radioactive gas in complex indoor leak scenarios, and the calculation time is short. In the test 2, PASM calculated the committed effective dose rate of the worker under the same leakage scenario. The novelty in this paper is summarized below:

1. A modified eddy-diffusion model is presented for evaluating the concentration of radioactive gases in complex indoor workplace.
2. Consider the impact of factory obstruction, facility blockage, duct exhaust, multiple sources and source stop leakage on diffusion.
3. An inhalation exposure calculation method is proposed for estimating the committed effective dose based on the inhaled dose conversion factor.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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