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Study on ^{14}C spatial distribution around Qinshan nuclear power plant in China

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Abstract To understand the spatial distribution of ^{14}C discharged from Qinshan nuclear power plant, where two HWRs and four PWRs are on commercial operation, simulation using Gaussian plume model was carried out. The predicted values were compared with the measured ^{14}C results of moss samples collected in 2010. A good fit was found between these two data sets, indicating that the spatial distribution of ^{14}C follows a Gaussian model. It can be concluded that HWRs are the dominant ^{14}C sources at Qinshan site and meteorological influences is the leading factor affecting the distribution of ^{14}C.

Keywords Qinshan NPP · ^{14}C · Gaussian plume model

Introduction

^{14}C, a long-lived radionuclide with half-life 5,730 years, is continuously produced in nuclear industry. It is well documented that ^{14}C in nuclear reactors is produced by neutron activation in the structural materials and component systems of the reactor [1]. According to existing data [2], the average release rate (average normalized release rates from 1990 to 1994) for the pressurized water reactor (PWR) and the heavy water reactor (HWR) was 0.22 and 1.6 TBq/GW_e, respectively. The produced ^{14}C may be released as $^{14}CO_2$ and ^{14}C hydrocarbons, depending on the type of reactor. For HWRs, the proportion of $^{14}CO_2$ in airborne releases is approximately 80 %, while for PWRs only 5–25 % of the released ^{14}C appears to be $^{14}CO_2$ [3].

The released ^{14}C, especially in the form of CO_2, is of particular interest because it can be mixed with natural carbon followed by the biological incorporation into biota, as carbon is the fabric of life. As a consequence, excess ^{14}C concentration may be found in surrounding plants. Several studies were performed to investigate the ^{14}C spatial distribution in atmosphere as well as in vegetation samples around nuclear power plant, by field investigation or model prediction [4–8].

In case of Qinshan NPP, where two HWRs (CANDU type) and four PWRs are employed, increased ^{14}C specific activities in biological samples were reported previously [9, 10]. Temporal variations of ^{14}C specific activity in terrestrial samples have been studied by analyzing a pine tree [9], and vegetation and food samples were also measured for radiation dose evaluation [10]. However, little study has been conducted on ^{14}C spatial distribution and the dispersion pattern of airborne effluents.

This study provides results of ^{14}C in moss samples collected at various distances from different wind directions as well as the predications from a Gaussian plume model. The objective of present research was to assess the ^{14}C spatial distribution in the vicinity of Qinshan NPP.

Experimental method and modeling approach

Experimental

Study site and sampling

The Qinshan site, located in Haiyan County, Zhejiang Province in the southeast of China, consists of three power

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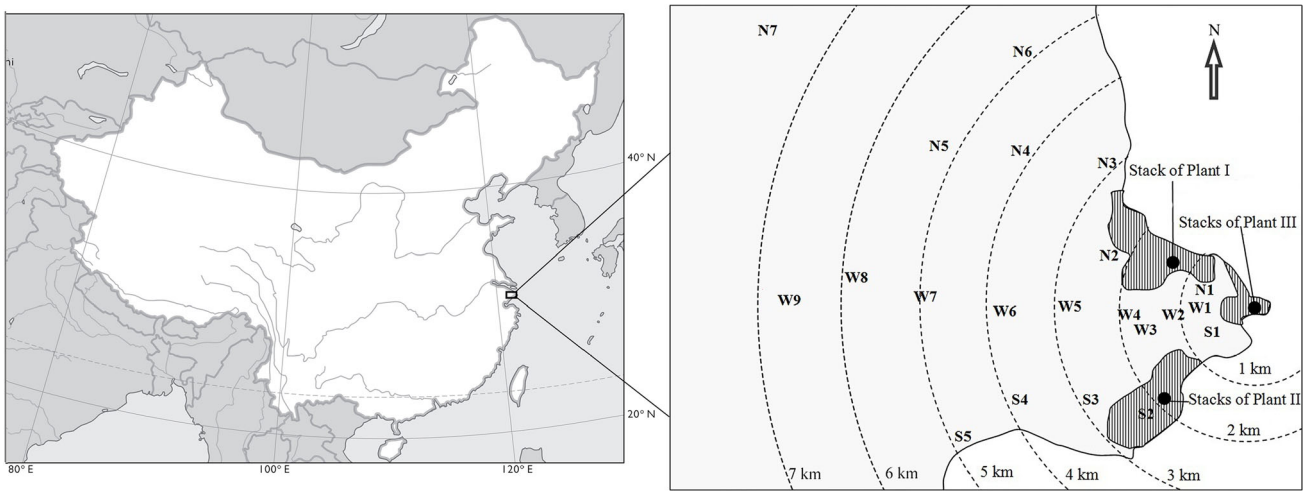

Fig. 1 Sites of collection of the moss samples from the Qinshan NPP

plants which were referred as Plant I, Plant II and Plant III (indicated in Fig. 1). Plant I, where the first PWR reactor (300 MW_e) in China was settled, started its operation in April 1994. Plant II comprises three PWR reactors (600 MW_e), and their operation began in April 2002, May 2004, and August 2010, respectively. Plant III uses two HWRs (CANDU type, 700 MW_e), which were put into operation in December 2002 and July 2003. The effluents are discharged from the stacks of each plant (stack heights are Plant I = 102 m; Plant II = 63 m; Plant III = 55 m), varying with time under normal operation.

The terrain in Qinshan site is basically flat, except for the hill named Qinshan (170 m high) at the west of Plant III's stacks. Meteorological information was obtained from three meteorological monitoring stations within the Qinshan site. The dominating wind direction in 2010 is from east and east-northeast.

As shown in Fig. 1, twenty-one moss samples were collected around the stacks of Plant III in December 2010, considering wind direction, distance to the stacks and availability for sampling. Four of the sampling spots (N_1, W_1, W_2 and S_1) were located on the Qinshan hill, while other sites were situated in the flat field. Moss was chosen as the environmental indicator for its ease of sampling, and distinct growing period which represents one year's ^{14}C assimilation [10]. Two reference moss samples were collected at Qiaosi in Hangzhou, 100 km away from Qinshan NPP.

Analytical procedure

After washing and vacuum drying (60 °C for 8 h), any foreign material was removed from the moss samples by hand. Then the samples were graphitized according to zinc reduction method [11]. Graphite samples were analyzed at Peking University AMS laboratory [12]. The results were determined according to the international convention, relative to the NIST oxalic acid standard (SRM-4990C) and corrected for isotopic fractionation [9].

Modeling approach

Atmospheric dispersion of ^{14}C

In the case of continuous releases, the Gaussian plume model was proved to be reliable to estimate long-term dispersion of radioactive effluents. In this study, the measured ^{14}C values in moss samples were compared to the values calculated using the following sector-averaged Gaussian plume model:

$$C(x,\theta_i,z) = \sqrt{\frac{1}{2\pi}}\frac{f_i \cdot Q}{x \cdot \Delta\theta} \cdot \sum_{S=A}^{F} \frac{f_s}{u_s \cdot \sigma_{z,s}} \cdot \left[\exp\left(-\frac{(z-h_e)^2}{2\sigma_{z,s}^2}\right) + xp\left(-\frac{(z+h_e)^2}{2\sigma_{z,s}^2}\right)\right]$$

In which $C(x, \theta_i, z)$ is the ^{14}C concentration in air (Bq/m^3) at a distance x (m) from the source, in sector θ_i at height above ground z (m); f_i is the frequency with which the wind blows into sector θ_i; Q is the release rate (Bq/s); $\Delta\theta$ is the sector width (radians); f_s is the frequency of stability class s; u_s is the average wind speed of stability class s (m/s); $\sigma_{z,s}$ is the vertical parameter for stability class s (m); and h_e is the effective release height (m).

For the consideration of reactor type and electricity capacity, the amount of ^{14}C discharged from Plant I was thought to be insignificant comparing to Plant II and Plant III [9]. Therefore, the influence of ^{14}C discharged from Plant I was not considered in this case. The ^{14}C concentrations were calculated separately for Plant II and Plant

III, and then added up before comparing with the measured activities.

Parameter determination

According to the discharge records, the monthly ^{14}C release rate (from January to December) in 2010 was used in calculation. For Plant II, the monthly ^{14}C release rate ranged from 1.36×10^2 to 3.94×10^3 Bq/s. The range for Plant III was from 2.87×10^3 to 2.21×10^4 Bq/s, which was nearly an order larger than that of Plant II. Since ^{14}C is not completely discharged in the form of CO_2 which can be absorbed by plants through photosynthesis, chemical forms of the ^{14}C effluents from plants are considered when calculating the source strength. In this case, CO_2 proportions

Table 1 Values of the parameters P and q for each stability case

Stability	Plant II		Plant III	
	P	q	P	q
A	0.337	0.844	0.376	0.809
B	0.294	0.822	0.376	0.809
C	0.247	0.783	0.376	0.809
D	0.295	0.704	0.240	0.770
E	0.136	0.719	0.216	0.731
F	0.227	0.622	0.216	0.731

Table 2 Results of ^{14}C specific activity in moss samples in the vicinity of Qinshan NPP

Sample ID	Specific activity (Bq/kg C)	Excess specific activity (Bq/kg C)
N1	250.7 ± 1.1	26.9 ± 1.3
N2	231.9 ± 0.8	7.1 ± 1.0
N3	225.1 ± 1.1	1.3 ± 1.3
N4	229.9 ± 1.0	6.1 ± 1.2
N5	228.6 ± 0.8	4.8 ± 1.0
N6	224.7 ± 0.8	1.1 ± 1.0
N7	223.0 ± 0.7	–
W1	264.3 ± 0.9	40.5 ± 1.1
W2	252.1 ± 1.1	28.3 ± 1.3
W3	237.3 ± 0.9	13.5 ± 1.1
W4	237.4 ± 0.9	13.6 ± 1.1
W5	230.4 ± 1.1	6.6 ± 1.3
W6	229.5 ± 0.8	5.7 ± 1.0
W7	229.1 ± 0.8	5.3 ± 1.0
W8	226.9 ± 1.4	3.1 ± 1.5
W9	225.1 ± 1.1	1.3 ± 1.3
S1	265.6 ± 1.3	41.8 ± 1.4
S2	233.3 ± 1.0	9.5 ± 1.2
S3	231.1 ± 1.1	7.3 ± 1.3
S4	231.8 ± 0.8	8.0 ± 1.0
S5	229.3 ± 1.0	5.5 ± 1.2
R[a]	223.8 ± 0.6	–

[a] Average ^{14}C specific activity of two reference samples

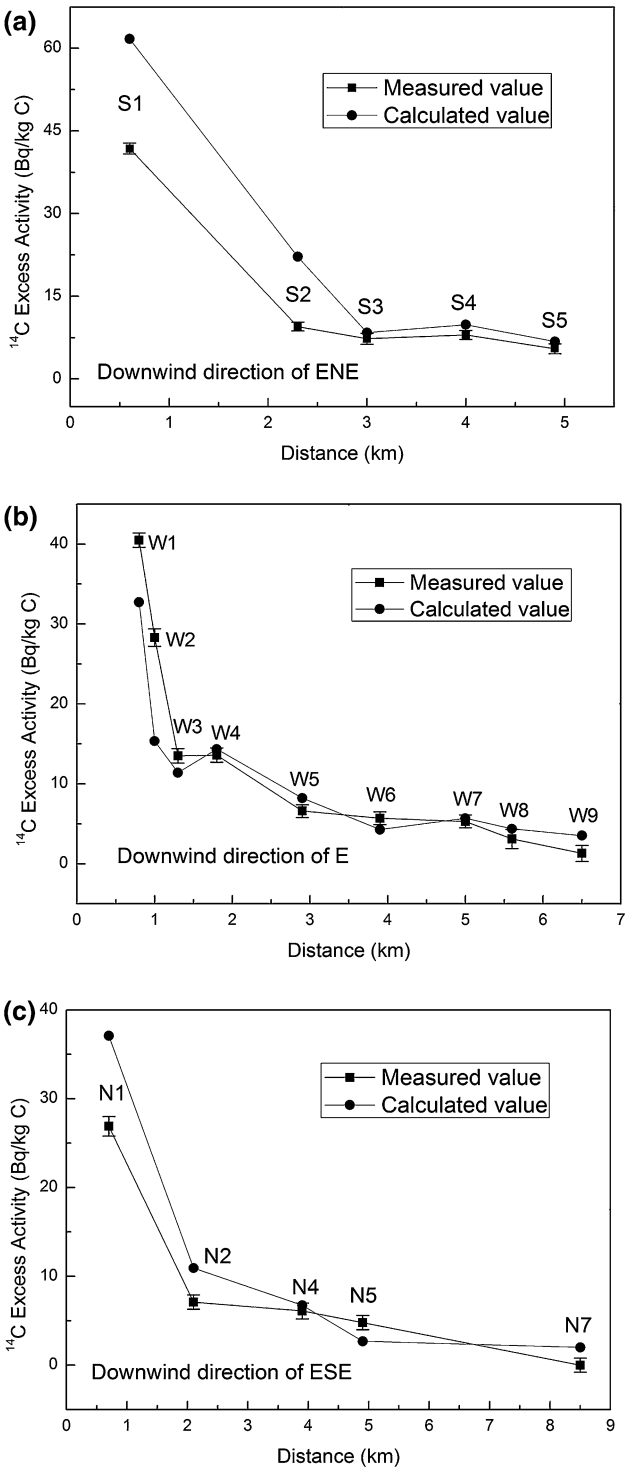

Fig. 2 Comparison between the measured excess ^{14}C and calculated activities of moss in different wind directions: (**a**) ENE; (**b**) E; (**c**) ESE

in airborne effluents of HWR and PWR used the values suggested by IAEA (80 % for HWR, 15 % for PWR) for lack of field data at Qinshan site [3].

Meteorological data were obtained from two monitoring stations (10 m above the ground) which were close to Plant II and Plant III, respectively. Monthly meteorological data that contained the frequency of the distribution of wind velocity categories per wind direction considering all the Pasquill stability classes was used in the statistics.

The vertical parameter for each stability measured by Hu et al. [13, 14] were chosen for calculation. The function of the vertical parameter is in the form of

$$\sigma_z = P \cdot x^q$$

Values for parameters were functions of stability, as listed in Table 1.

The plume rise calculation used Rupp's equation for momentum dominated plume is [15]:

$$\Delta h = \frac{1.5\ vd}{u}$$

Where Δh is the plume rise (m); v is effluent stack gas velocity (m/s); d is the inside stack diameter (m); u is the wind velocity (m/s). The effective height of the release h_e can be derived from the physical stack height plus the plume rise.

The dispersion of ^{14}C was calculated for each month in 2010, and the mean values averaged from January to December were used for comparison to the measured ^{14}C concentrations in moss.

Results and discussion

The results of the ^{14}C analysis in moss samples are shown in Table 2. The specific activity is expressed in Bq/kg C. The ^{14}C specific activity of moss samples ranges from 223.0 to 265.6 Bq/kg C with the maximum value 18.7 % above the reference value (223.8 Bq/kg C). An obvious decreasing trend can be found in the results of moss samples as the distance increased. Moss sample W_9, the farthest sample to Plant III's stack in the prevailing wind direction, has a value slightly above background. We can conclude that most of the ^{14}C discharged from Qinshan NPP was deposited in a radius of 6.5 km.

The annual average ^{14}C concentration of the sampling spot, calculated from Gaussian model, is expressed as Bq/m^3 originally. To compare with the measured values, the unit of the prediction is changed to Bq/kg C using the monitored CO_2 concentration (405 ppmv) of Zhejiang in 2010. Based on the assumption of specific activity equilibrium between moss and air, the predicated ^{14}C specific activity in moss equals to the calculated airborne ^{14}C concentration where the moss sample was collected.

Predictions for the ^{14}C excess concentrations in moss are compared with the observations in Fig. 2. Previous study demonstrates that the diffusion of ^{14}C discharged from the Qinshan NPP is mainly affected by the geographical environment and wind direction [10]. This study further discusses the two factors. As shown in Fig. 2, for most of the samples which were collected beyond the Qinshan hill, the model reproduces the measured values well in both magnitude and trend. But for samples collected on the hill (N1, W1, W2 and S1), difference in magnitude can be observed between the measured and calculated values. This is probably a consequence of the turbulence of local topography. In general, the predicted to measured value ratio is less than a factor of 2, which indicates a good fit of the model. Since the plume follows a Gaussian dispersion model, we can say that the meteorological condition is more important in affecting the plume dispersion than the topography in Qinshan site, whose influence is limited in the hill itself.

The contributions of the ^{14}C enrichment in moss samples from Plant II and Plant III are also compared in Table 2. According to modeling of gaseous release from Plant II's stack, the five sampling spots selected are the places where excess ^{14}C concentration can be expected due to the ^{14}C discharged from Plant II. As indicated in Table 3, calculated values from Plant III are significantly higher than those calculated from Plant II. This is supposed to be a consequence of different reactor types these plants employed. ^{14}C discharged from Plant II is an order less than that from Plant III and is mainly composed of hydrocarbons, which can't be incorporated by plants through photosynthesis. Therefore, Plant III is primarily accounted for the ^{14}C enrichment of the samples even though it is further away from these spots.

Table 3 Excess values calculated from Plant II and Plant III, and measured ^{14}C excess activities for samples downwind of the Plant II stacks

Sample	S3	S4	S5	W6	W7
Calculated values from Plant II (Bq/kg C)	1.3	1.6	0.2	1.0	0.6
Calculated values from Plant III (Bq/kg C)	7.1	8.2	6.7	5.1	3.2
Measured values (Bq/kg C)	7.3 ± 1.3	8.0 ± 1.0	5.5 ± 1.2	5.7 ± 1.0	5.3 ± 1.0

Conclusion

In this study, modeling methods was applied to study the spatial distribution of the ^{14}C discharged from Qinshan NPP. The enriched ^{14}C concentrations in moss samples mainly came from Plant III, whose influence was significantly higher than Plant II. The atmospheric dispersion of ^{14}C discharged from Plant III follows a Gaussian pattern, indicating meteorological condition is the leading factor in ^{14}C diffusion. In contrast, the disturbance of the elevated terrain (Qinshan hill) on the plume was relatively small and limited in the hill.

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