LATITUDINAL DISTRIBUTION PATTERNS OF POPs IN ATMOSPHERIC AEROSOLS IN THE EAST ASIAN MONSOON CLIMATE SYSTEM: IMPLICATIONS FOR ATMOSPHERIC LONG-RANGE TRANSPORT OF PERSISTENT ORGANIC POLLUTANTS - USING BERYLLIUM-7 AS A REFERENCE

Yang Y-L[1*], Zhu X-H[1], Geng C-Z[2], Li Y[3], Zhang G[4], Jiao X-C[1], Gai N[1], Rao Z[1], Li Y [2], Yin X-C[2]

[1] National Research Center of GeoAnalysis, Beijing 100037, China;
[2] College of Chemistry, Chemical and Environmental Engineering, Qingdao University, Qingdao 266071, China;
[3] Chinese Academy of Agriculture Sciences (CAGS), Beijing 100081, China;
[4] National Key Laboratory of Organic Geochemistry, Guangzhou Institute of Gochemistry, Chinese Academy of Sciences, Guangzhou 510640, China

Introduction

The long-range transportation processes of POPs can cause potential serious effects on the ecologic environment of remote areas which seem to be clean and has become one of the global concerns on the environment. The concepts of "cold condensation" and global fractionation" have been formulated in interpretation of the mechanisms of semi-volatile organic pollutants migrating to polar areas through evaporation and atmospheric transport[1,2,3]. However, only a small fraction of the inventory of POPs has reached the far north in the North Hemisphere. Soils have had a key role in 'protecting' the Arctic, by retarding re-cycling and retaining POPs, particularly in the major stores of temperate forests, grasslands and peat bogs[3]. The tracer should have no sources from land and its geographic distribution in troposphere is mainly governed by atmospheric circulation. Here we proposed a mechanism for supplementary interpretation of latitude distribution pattern of POPs in East Asian monsoon climate system (EAMCS) using cosmogenic nuclide berylium-7 (^{7}Be) as a reference**.** Berylium-7 can be an ideal tracer for atmospheric circulations and aerosols on a short-term time scale. Beryllium-7 (^{7}Be) is a natural radionuclide originated by interaction of cosmic rays with nitrogen and oxygen in the stratosphere and upper troposphere. The seasonal and longitudinal changes in ^{7}Be atmospheric production rate can be neglected. There is little report available on simultaneous study on ^{7}Be and persistent organic pollutants in the atmospheric aerosols[4]. Such a study could further explore ^{7}Be as the geochemical tracer and provide a different perspective on the global fate of persistent organic pollutants.

Materials and methods

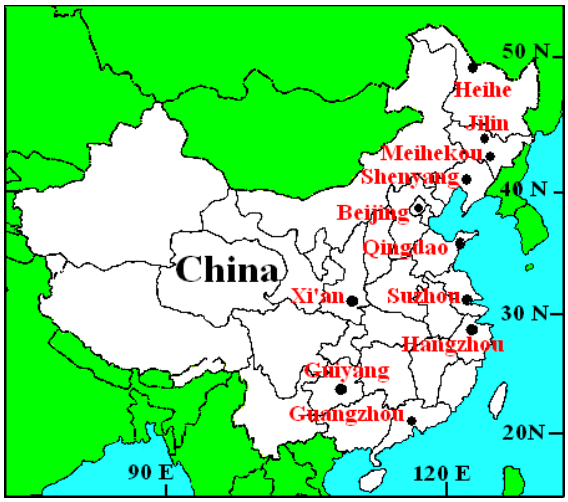

Fig. 1 Map of cities at defferent latitude in East Asian monsoon zone selected for this study

The sampling site for consecutive measurement of ^{7}Be in aerosols were in Xicheng District of Bejing (39°54′ N, 116°24′ E), Laoshan District of Qingdao (35°36′ N，119°30′ E; 75 m above sea level), and Tianhe District of Guangzhou (23°08.25′ N，113°19′ E; about 25 m above sea level). Near-surface atmospheric aerosol samples were collected at the frequency of 2–3 d per week using a large volume TSP (total suspended particles) sampler. The ^{7}Be activity was measured by its 477.16 keV (the branching ratio: 10.5 %) γ-ray using the high-resolution gamma-ray spectrometer measurement system (Canberra, US) with a BE5030 detector with a range for γ-ray energy of 3 keV ~ 3 MeV. The analyses of OCPs and PCBs in all samples and field blanks were carried out by a gas chromatograph (HP6890) equipped with a ^{63}Ni electric capture detector (ECD), using a fused silica capillary column (HP-5MS, 30 m×0.32 mm i.d., with film thickness of 0.25 μm).

Results and discussions

1. ^{7}Be concentrations in near-surface atmospheric aerosols

The seasonal distribution patterns of ^{7}Be concentrations in Beijing, and Qingdao were in the sequence of autumn > spring > winter > summer, significantly different from that observed in the low latitude city Guangzhou in South China which was in the sequence of spring > winter > summer > autumn. Our result is generally in accordance with that of Kulan et al.[5] and the fallout distribution of cosmogenic isotopes to the Earth's surface characterized by a mid-latitude peak.

It should be pointed out that our data did not cover the higher latitude (i.e. > 40 °N) and the Gaussian distribution pattern in Fig. 3 is only an approximation.

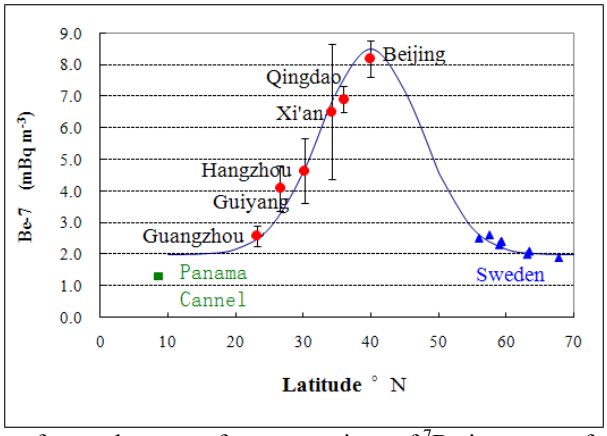

Fig. 3 Latitudinal distribution of annual means of concentrations of ^{7}Be in near-surface atmospheric aerosols in East Asia monsoon zone in China (solid circle) and comparison with some other areas in Sweden (solid triangle) and Panama Cannel (solid square) (ref. [5])

Fig. 4 presents the latitudinal distribution pattern of near-surface atmospheric ^{7}Be measured during October 19-22, 2009. This instantaneous latitudinal distribution of ^{7}Be showed that the distribution pattern still approximately followed a Gaussian curve, but compared with the annual average ^{7}Be distribution pattern, the maximum was significantly higher (15.2±0.9 mBq m^{-3}) and the peak was occuring at Suzhou (31.2 °N), i.e. the peak moving southward. With the help of the air mass back-trajectory, it can be seen that a cold air mass moved from Siberian region during the sampling period (Fig. 5). Averagely speaking, our result of the latitudinal distribution pattern of near-surface atmospheric ^{7}Be in the East Asian monsoon zone in China display a Gaussian distribution mode with the peak at Beijing (40 °N), but at a particular time, that distribution pattern can instantaneously move southward or northward depending on the season, and the value at the peak will also vary depending on the strength of the sinking air from the high elevation and the extents to which the air exchange between troposphere and stratosphere.

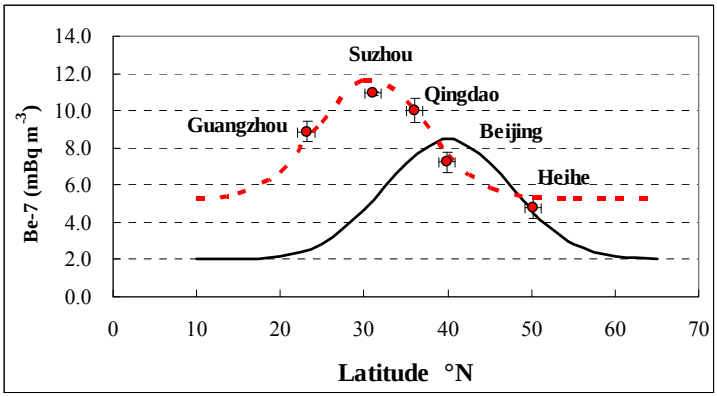

Fig. 4 Latitudinal distribution (the dotted curve of ^{7}Be in aerosols from different Chinese cities of East Asia Monsoon Zone during October 20-24, 2009 (the solid curve is the one shown in Fig. 3)

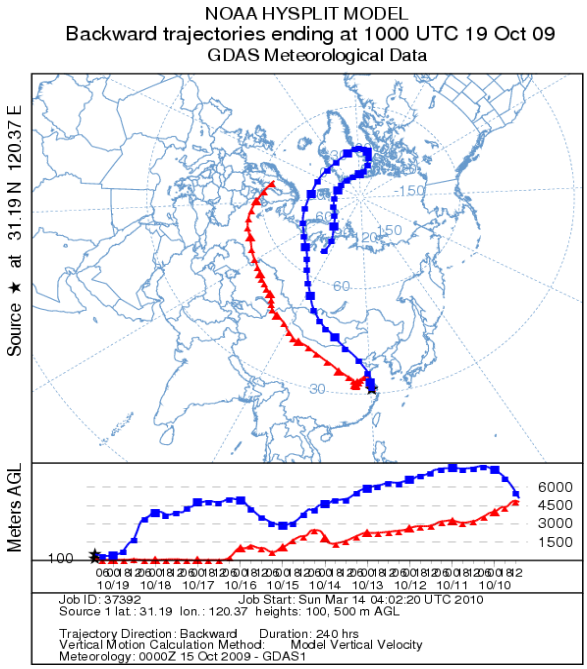

Fig. 5 Back-trajectories of air-masses at the sampling sites in Suzhou during the sampling period

2. Concentrations of POPs in air

Fig. 6(a) presents the concentrations of various groups of POPs studied in the same aerosol samples of ^{7}Be simultaneously collected at the five stations of different latitude on 19-24, October 2009. The three groups of POP compounds were all low at Qingdao that is located in the Shandong Peninsular north of the Yellow Sea. Influenced by the oceanic climate, air masses in Qingdao come from the ocean generally containing less aerosols, playing a dillution effect to the aerosols in continental air mass. Lammel et al. (2007)[6] investigated POPs in atmospheric aerosols in the Yellow Sea in summer and found that DDTs, HCB, HCHs, and PCBs concentrations were in the range of 16–180 pg m^{-3} (ref. 33). Our results for DDTs, HCHs, and PCBs in atmospheric aerosols at Qingdao during the sampling period were 10–200 pg m^{-3}, therefore the low concentrations of these POPs in aerosols at Qingdao could not be an occasional phenomenon. Passive air sampling were conducted in five cities (Qingdao, Beijing, Shenyang, Meihekou, Jilin) and the results are shown in Fig. 6(b). DDTs were the dominant compounds in these POPs at all stations. In contrast to the latitudinal distribution patterns in aerosols, the

latitudinal distribution patterns of these POPs in vapor phase showed a distinctive trend, i.e., the concentrations of the POPs in general decrease with increase of latitude.

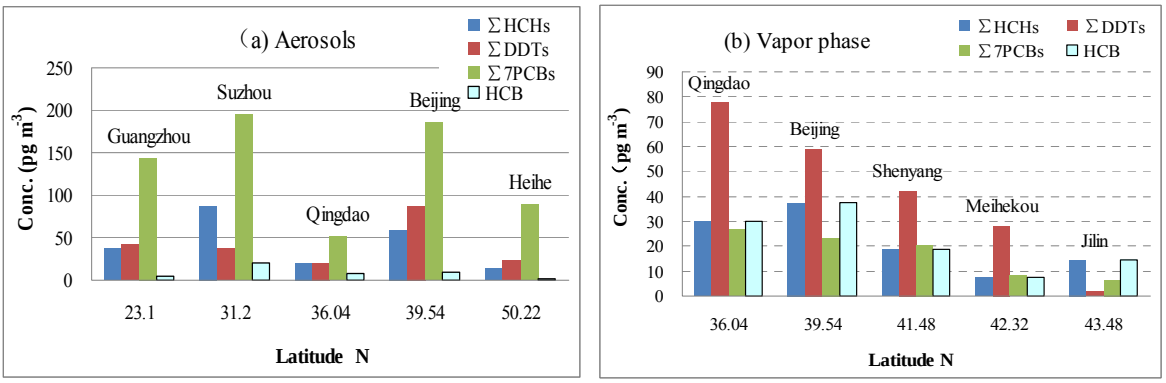

Fig. 6 Latitudinal distributions of concentrations of POP compounds in (a) aerosols and (b) passive air samplings from cities of different latitude in East Asia monsoon zone in autumn, 2009

3. Co-relationship between POPs and 7Be

ΣHCHs, ΣDDTs, and ΣPCBs showed good co-relationship with ^{7}Be (Fig. 8), suggesting that these POP compounds in atmospheric aerosols in Qingdao[7] may have same similar transport mode with ^{7}Be in these months, leading to higher consistency in their seasonal variation patterns. Similar correlations had been also found in Guangzhou during the period of July ~ December, 2007[4].

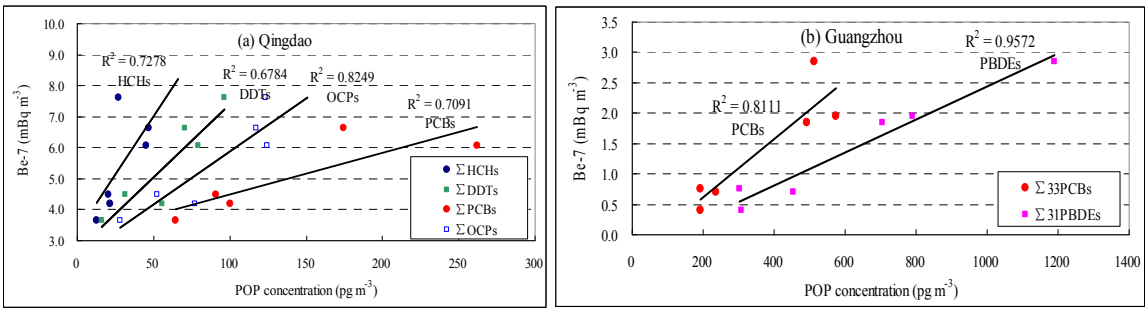

Fig. 8 Correlations between the monthly average concentrations of ^{7}Be with ∑POPs

Acknowledgements

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