Journal of Zhejiang University SCIENCE A
ISSN 1673-565X (Print); ISSN 1862-1775 (Online)
www.zju.edu.cn/jzus; www.springerlink.com
E-mail: jzus@zju.edu.cn

Levels and statistical analysis of aerosol phase PAHs over Qingdao alongshore

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Received Jan. 20, 2007; revision accepted Apr. 25, 2007

Abstract: Atmospheric concentrations of polycyclic aromatic hydrocarbons (PAHs) were measured during various seasons at six different cities/locations in Qingdao alongshore. The annual average PAHs concentration ranged from 16 ng/m^3 (at a clean compared site) to 308 ng/m^3 (in an industry site). The average total particulate PAHs concentration was 74.5 ng/m^3 with a higher concentration of particulate PAHs in winter. Based on a year-round dataset, the sources of PAHs in the air of Qingdao were drawn by principal factor analysis and correlation analysis. The results indicated that vehicle emissions and oil burning were the main source of PAHs in Qingdao alongshore.

Key words: Polycyclic aromatic hydrocarbons (PAHs), Atmosphere aerosol, Factor analysis, Correlation analysis
doi:10.1631/jzus.2007.A1164 **Document code:** A **CLC number:** X2

INTRODUCTION

Polycyclic aromatic hydrocarbons (PAHs) are a group of organic compounds with two or more fused aromatic rings. Some PAHs such as benzo [a] pyrene have shown mutagenic and carcinogenic properties (Ravindra *et al*., 2001). PAHs mainly come from incomplete combustion processes of carbon-containing substances, e.g. vegetation fires and vehicle exhausts (Isidorov, 1990). They are mostly contained in atmospheric aerosols in minute amounts, due to their hydrophobicity and affinity for small particles. On the other hand, PAHs are usually concentrated in locations of high human activity, e.g. near major highways, in major cities and in some industrial soils (Henner *et al*., 1997; Bryselbout *et al*., 2000).

The PAHs were identified to be one of the major toxic air pollutants in urban environments (Tan *et al*., 2006). The majority of the population in cities in China reside along the eastern coast and in central plains. The outflow of air pollution from these highly populated regions has drawn attention in recent years (Wang *et al*., 2003). Vehicle exhaust, coal combustion both for power generation and for space heating, as well as wood combustion were identified to be the major PAH sources in the cities (Vestreng and Klein, 2002).

Earlier studies were focused on how to analyze PAHs and how they were distributed in the city atmosphere (Halsall *et al*., 1994). Recently, more attention has been attracted to identify the source of PAHs in air particles by statistical analysis (Ravindra *et al*., 2006; Luo *et al*., 2006), molecular identification (Fu *et al*., 1997) and stable carbon isotope (Okuda *et al*., 2002). By the stable carbon isotope approach, the pollution source is determined via measurement of δ^{13}C in an organic compound using an isotope mass spectrograph. By statistical analysis, organic pollutants in the aerosol from different sources are statically compared to find marked components for analysis of the pollution sources. It is becoming an important tool in the search for the PAHs sources because its original data are almost not affected by the decomposition of the pollution sources.

In this paper, the aerosol phase PAHs concentrations were monitored from six different areas in the city of Qingdao (China), for a year. Further the potential sources were also identified using multivariate statistical method.

EXPERIMENTAL DETAILS

Sampling sites

In this paper, we focus on the estimates of atmospheric deposition of PAHs from one year of sampling at 6 sites around Qingdao (Fig.1). To represent the industrial signal, sampling station was sited at Red-Star Chemical Plant. The Nanjing Road Qingdao Fishery Department site was chosen to represent the commercial area signal. The Shazikou station was located to represent clean area. The North Sailing Construction Engineering Department site is located in a residential area. The Sifang Hotel site and Jiayuguan site were chosen to represent dense traffic and culture area signal, respectively. The samples were collected at different auto air monitoring station in spring, autumn, winter. One hundred and eighty-nine samples were collected from the 6 stations in the Qingdao alongshore. The number of samples was 41, 25, 34, 35, 15, 35 for Red-Star Chemical Plant, North Sailing Construction Engineering Department, Nanjing Road Qingdao Fishery department, Sifang Hotel, Jiayuguan, Shazikou, respectively.

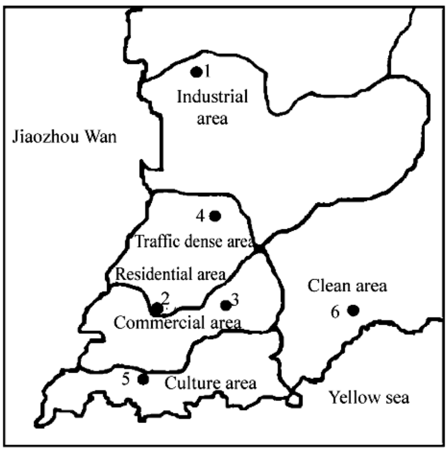

Fig.1 Map of Qingdao showing the air monitoring stations
1: Red-Star Chemical Plant; 2: North Sailing Construction Engineering Department; 3: Nanjing Road Qingdao Fishery Department; 4: Sifang Hotel; 5: Jiayuguan; 6: Shazikou

PAH analysis

1. Instrument and reagents

The HPLC equipment consisted of a HPLC pump and an LC-240 florescent detector (Perkin Elmer). Automatic atmosphere monitoring station was from API (USA). RAM-1020 β-radial dirt detector was obtained from Anderson (USA).

Reagent and materials. The PAHs standard mixed solution (100 mg/L) was purchased from Supleco, composed of acenaphthene (Acp), fluorene (Flu), phenanthrene (PA), anthracene (Ant), fluoranthene (FluT), pyrene (Pyr), benzo [a] anthracene (BaA), chrysene (Chr), benzo [b] fluoranthene (BbF), benzo [k] fluoranthene (BkF), benzo [a] pyrene (BaP), dibenzo [a,h] anthracene (DahA), benzo [ghi] perylene (BghiP), indeno [1,2,3-cd] pyrene (Ind); deionized water was from Mili-Q pure water system.

Glass fiber filter belt was 30 mm×20 m, which was pretreated as follows: Soak the glass fiber filter belt in the scarce nitric acid for a week before use, then put it into the Mili-Q water for a week, dry up naturally. Put it into the desiccator to further dry up until the fiber filter belt is uniform in weight for use.

2. Sample pretreatment

The atmosphere aerosol samples were collected from the automatic atmosphere sampling station. Minute particles from the atmosphere were gathered by the glass fiber filter belt. Put the macula on the glass fiber filter belt into glass tube after slicing into small pieces, add in certain amount of dichloromethane, ultrasonically extract three times, 15 min each time. Filter the extraction into K-D condenser after filtering with the 0.45 μm membrane filter. Condense the solution into about 0.5 ml at 65~70 °C water bath, then dry by passage of nitrogen gas. Use methanol to dissolve the residuals and set the volume at 0.50 ml for testing. Filter samples using the 0.45 μm needle filter before HPLC analysis.

3. PAHs HPLC analysis

Twenty μl injector loop, 100×4.6 mm ID PE CHROMSEP (3 μm stationary phase). Methanol-water was selected as the mobile phase. Elution with 15% methanol-100% water (v/v) was isocratic for 7 min, then linear gradient elution for 5 min, then the mobile phase ratio changes to 5/95 in 17 min. Under the conditions in this experiment, the 13 PAHs checked out can all reach ideal separation. PAHs have the following detection limits (pg): Acp 21.38, PA

16.34, Ant 0.90, FluT 16.68, Pyr 3.34, BaA 6.88, Chr 8.57, BbF 6.53, BkF 1.4, BaP 2.74, DahA 8.65, BghiP 13.73, Ind 55.19. The Relative Standard Deviation (RSD) for the above mentioned 13 PAHs recovery rate all fall in the scope of 6.0%~17.0% except for PA at 25%.

RESULTS AND DISCUSSIONS

Evaluation of site-specific PAHs concentrations in the Qingdao alongshore

The results of analysis on PAHs at different stations in the atmosphere at different seasons are shown in Table 1. The highest annual average concentration of PAHs was found at Red-Star Chemical Plant (industrial area, 24.4 ng/m^3 in spring, 54.0 ng/m^3 in autumn and 308 ng/m^3 in winter), followed with lower concentrations by Nanjing Road Qingdao Fishery Department site (commercial area, 32.6 ng/m^3 in spring, 71.8 ng/m^3 in autumn and 114 ng/m^3 in winter), Sifang Hotel (traffic dense area, 26.8 ng/m^3 in spring, 73.4 ng/m^3 in autumn and 101 ng/m^3 in winter), Jiayuguan (cultural area, 56.8 ng/m^3 in autumn and 103 ng/m^3 in winter), North Sailing Construction Engineering Department (residential area, 47.2 ng/m^3 in spring, 68.2 ng/m^3 in autumn) and Shazikou (comparatively clean area, 16.0 ng/m^3 in spring, 30.3 ng/m^3 in autumn and 64.7 ng/m^3 in winter). The total concentration of 13 PAHs at the six sites averaged 29.4 ng/m^3 in spring, 70.9 ng/m^3 in autumn, 138.1 ng/m^3 in winter.

This high concentration of PAHs at Red-Star Chemical Plant seems to be related with the activities of the industry in the vicinity of this sampling site. The sampling site in Sifang Hotel is located downtown near a main road with heavy traffic. At the observation point, the common source of PAHs is most likely the vehicular emission. The total average concentration of PAHs reported at Shazikou is comparatively lower than that found at the other sites. This observation can be justified, as the sampling site is primarily a rural one with negligibly small influence of industrial activities and/or emissions by vehicles, although some nearby agricultural activity can contribute to the increased background levels of PAHs.

The average total concentration of particulate PAHs reported in Qingdao alongshore air was 74.5 ng/m^3. FluT (17%), Pyr (13%), DahA (12%), Chr (11%), BbF (11%), BaA (9%), BaP (8%) and PA (7%)

Table 1 Concentration of individual PAH compounds (ng/m^3) at the sampling sites in Qingdao alongshore during various seasons

Location/seasons	Acp	PA	Ant	FluT	Pyr	BaA	Chr	BbF	BkF	BaP	DahA	Ind
Sifang Hotel/spring	n.d.	2.04	n.d.	6.68	1.60	2.26	2.31	3.17	1.46	2.52	2.15	n.d.
Sifang Hotel/autumn	7.48	2.20	0.42	14.8	12.1	8.49	8.64	7.84	3.33	5.90	5.84	n.d.
Sifang Hotel/winter	n.d.	7.83	0.54	17.0	14.5	7.03	11.4	11.0	3.74	6.10	10.6	7.06
Red-Star Chemical Plant/spring	n.d.	n.d.	0.15	2.90	1.82	2.90	3.36	3.54	1.44	2.73	3.70	n.d.
Red-Star Chemical Plant/autumn	n.d.	3.60	0.90	8.20	6.49	5.54	7.45	6.02	2.23	4.44	7.04	n.d.
Red-Star Chemical Plant/winter	12.5	22.0	5.12	42.9	43.1	25.0	35.1	28.5	10.8	23.6	44.5	14.1
Nanjing Road Qingdao Fishery Department/spring	n.d.	2.74	n.d.	5.17	2.33	2.84	2.68	4.92	1.92	2.88	3.09	n.d.
Nanjing Road Qingdao Fishery Department/autumn	n.d.	1.25	1.55	11.5	9.99	7.21	8.73	8.30	3.50	6.63	8.69	n.d.
Nanjing Road Qingdao Fishery Department/winter	2.18	5.70	1.75	22.9	15.4	9.15	12.4	10.9	4.34	8.18	11.4	9.26
Jiayuguan/autumn	n.d.	2.99	0.89	10.0	9.01	4.56	6.38	6.25	2.31	4.16	6.08	n.d.
Jiayuguan/winter	3.65	5.87	1.05	14.0	13.1	9.07	12.6	9.90	3.79	7.70	10.8	6.03
North Sailing Construction Engineering Department/spring	n.d.	9.33	1.87	7.36	4.17	3.07	5.20	4.42	1.92	3.30	3.56	n.d.
North Sailing Construction Engineering Department/autumn	n.d.	1.06	1.34	14.6	7.03	6.99	7.91	8.15	3.42	7.04	9.51	n.d.
Shazikou/spring	0.16	n.d.	n.d.	n.d.	0.82	1.72	2.16	2.30	0.97	1.85	2.53	n.d.
Shazikou/autumn	n.d.	2.13	0.21	6.13	4.34	0.77	3.20	5.07	1.69	1.59	4.94	n.d.
Shazikou/winter	3.43	1.92	0.66	9.42	9.44	5.67	6.99	6.11	2.23	3.55	10.0	n.d.

n.d.: not detected

were found to be the most common PAH compounds with relatively high concentrations in the particulate fraction in Qingdao alongshore.

Seasonal variation in the concentration of PAHs

The concentrations of PAHs during different seasons at various sampling sites in Qingdao are also shown in Table 1. The total PAH concentration measured at industry site was almost 13 fold during winter in contrast to spring samples. The average total concentrations of PAHs at heavy traffic area and commercial area were found to be four fold during winter in contrast to spring samples. A possible reason for the high concentration of particulate PAHs during winter season seems to be related with the very low temperature during these periods, which made favorable conditions for the condensation/adsorption of these species on suspended particles present in air (Ravindra *et al*., 2006).

Source identification

The judgment on the sources of atmosphere pollutants alongshore is commonly based on statistical method. In this paper, factor analysis and correlation analysis were adopted. SPSS statistical analysis software package was used in analyzing the different types of PAHs in the atmosphere aerosol in each district of Qingdao. The potential sources of the PAHs and their related factors are discussed.

1. Factor analysis

Factor analysis is multivariate statistical analysis method to explore the correlations among multiple variables, to use a few representing factors to explain the complex information provided in many variables, via mathematical transformation. Table 2 provides the results of factor analysis of total concentrations of PAHs at all the sites studied in Qingdao.

Table 2 provides the results of factor analysis of total concentrations of PAHs at all the sites studied in Qingdao alongshore. The PCA results showed that two of the factors explain the main part of the data variance. Most loading of PAHs is higher than 0.9. Only in Sifang Hotel sampling position, there is negative load for Pyr, BaA, and a low load (0.166) for Chr. The high factor loading of Pyr, BaA, BaP, BbF has been suggested for gasoline powered vehicles (Guo *et al*., 2003). Diesel emission has high factor loading for Pyr (Fang *et al*., 2004). The high factor loading of Chr, BbF indicates stationary emission sources (Kulkarni and Venkataraman, 2000). The high level of BaP and Chr has been suggested for steel industry emissions. The PAHs compounds from coke ovens are released during coking operation as fugitive emission in steel industries. Based on these observations, the factor 1 of samples at the six sites indicates vehicular emission, oil burning and stationary emission sources.

For Red-Star Chemical Factory, the loading is greater than 0.991 for factor 1 on 8 PAHs, and is the highest among all 6 monitoring stations. The load on TSP is the highest for factor 1 on all regular factors of atmosphere aerosol, and there is no load on NO_x. This indicates that for this station the PAHs mainly come from the TSP emitted from the boiler burning coals. TSP is the main medium and carrier for PAHs. The density of TSP in this area is the controlling factor for the density of PAHs. At Red-Star Chemical Factory monitoring position, there are big enterprises like Qingdao Steel Co., Qingdao Alkali Factory and many coal burning boilers emitting not only air pollutants TSP and SO_2, etc., but also many types of PAHs. The same is true for Nanjing monitoring station, where around its vicinity there are all types of PAHs carried by smoke and dirt emitted from the huge coal burning boilers of Qingdao Thermoelectric Factory and Thermo-source Company going into the atmosphere. This is an important reason why the load of 8 types of PAHs is high at these positions.

For factor 2, there is loading on TSP, CO, and there is relatively higher loading on SO_2, NO_x. There is no loading on the 7 types of PAHs at Red-Star Chemical Plant, North Sailing Construction Department, Nanjing Road Qingdao Fishery Department and Shazikou. The characteristics of load for common factor 2 at Sifang Hotel monitoring position are significantly different than that in other positions. At Sifang Hotel, there is high loading on Pyr and BaA, and there is no loading on the other five types of PAHs. This indicates that the impact of other sources to determine the density of PAHs in atmosphere aerosol at Red-Star monitoring positions is trivial. However, at Sifang Hotel, the impact is obvious, which is shown by the high loading on PAHs, Pyr and BaA at 0.505, 0.807 and 0.892. The loading of factor 2 on NO_x is also very high, reaching 0.892. This indicates that all types of PAHs may be related to the

Table 2 Factor analysis of total PAH concentration data set of various sampling sites

Element	Red-Star Chemical Plant		North Sailing Construction Department		Nanjing Road Qingdao Fishery Department	
	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2
TSP (μg/m^3)	0.551	0.509	0.732	0.269	0.305	−0.126
SO_2 (μg/m^3)	0.155	0.760	0.447	0.421	0.276	0.737
NO_x (μg/m^3)	−0.390	0.658	0.156	0.885	0.224	0.818
CO (μg/m^3)	0.286	0.521	3.190E−02	0.785	0.686	9.215E−03
ΣPAH (ng/m^3)	0.995	−3.851E−02	0.958	4.893E−03	0.981	−2.536E−02
Pyr (ng/m^3)	0.993	−2.379E−02	0.904	−2.229E−02	0.865	−5.543E−02
BaA (ng/m^3)	0.993	−2.379E−02	0.985	−8.527E−02	0.986	−8.626E−02
Chr (ng/m^3)	0.993	−2.379E−02	0.971	−1.864E−02	0.989	−3.986E−02
BbF (ng/m^3)	0.993	−2.379E−02	0.960	−0.177	0.977	1.631E−03
BkF (ng/m^3)	0.993	−2.379E−02	0.982	−2.953E−02	0.978	−3.942E−02
BaP (ng/m^3)	0.993	−2.379E−02	0.969	−6.954E−02	0.964	−8.013E−02
DahA (ng/m^3)	0.995	−3.412E−02	0.937	−0.176	0.969	−4.500E−02
Eigenvalue	9.456	1.552	8.112	1.725	8.130	67.746
Cumulative variance (%)	72.739	84.678	67.596	81.973	67.746	78.172
Element	Sifang Hotel		Jiayuguan		Shazikou	
	Factor 1	Factor 2	Factor 1	Factor 2	Factor 1	Factor 2
TSP (μg/m^3)	0.587	0.505	0.694	0.661	0.324	1.080E−02
SO_2 (μg/m^3)	−0.203	0.807	0.551	0.741	−0.355	0.705
NO_x (μg/m^3)	−0.101	0.892	0.776	0.619	0.294	0.740
CO (μg/m^3)	0.166	5.924E−02	0.559	0.487	−5.706E−02	0.470
ΣPAH (ng/m^3)	0.587	0.505	0.935	−0.316	0.976	6.477E−02
Pyr (ng/m^3)	−0.203	0.807	0.876	−0.237	0.782	−1.737E−03
BaA (ng/m^3)	−0.101	0.892	0.943	−0.317	0.940	0.125
Chr (ng/m^3)	0.166	5.924E−02	0.937	−0.336	0.983	−5.148E−02
BbF (ng/m^3)	0.988	2.393E−02	0.985	−0.113	0.959	−8.984E−02
BkF (ng/m^3)	0.917	5.977E−02	0.977	−0.107	0.965	−1.535E−02
BaP (ng/m^3)	0.940	−0.171	0.964	−0.148	0.927	9.459E−02
DahA (ng/m^3)	0.992	2.807E−02	0.962	−0.151	0.962	−6.169E−02
Eigenvalue	7.716	1.780	8.889	2.045	7.369	1.309
Cumulative variance (%)	64.300	79.131	74.076	91.114	61.409	72.313

NO_x in other sources of pollutants. It can be inferred that, for Sifang Hotel position, the factors deciding the density of PAHs in the atmosphere aerosol include not only sources of coal burning, but also other sources, in particular, gas burning and waste air emissions. Sifang Hotel is located in an area with heavy traffic, the emissions from automobils is a factor that cannot be underestimated.

2. Correlation analysis

Correlation analysis is used to judge the degree of relations between variables based on the correlation coefficients between parameters. Minute particles in the atmosphere are the medium and carrier of PAHs, and their density is closely related to all type of PAHs. Hence, the correlations between all types of PAHs in the minute particles in the atmosphere are analyzed in this paper. At the same time, there is certain correlation between all types of PAHs and regular monitoring factors NO_x and SO_2, which are also analyzed in this paper so that the sources of PAHs and their distribution in the atmosphere aerosol can be better understood from multiple angles and levels.

The normal person correlation coefficients were adopted in this paper, which measure the degree of linear correlations between two variables. The signs of the correlation coefficients represent the direction of correlations, and the absolute values represent the degree of correlations. There are close correlations between the 7 types of PAHs. The correlation coefficients for all PAHs are above 0.9 except for Pyr at a few positions, indicating that their sources are similar. The correlations between the 7 types of PAHs and TSP are different at certain positions. The correlation coefficients are between 0.4~0.7 for positions such as Red-Star Chemical Plant, North Sailing Construction Engineering Department, Sifang Hotel and Jiayuguan. The correlation coefficients, except for Pyr, are relatively small, between 0.2~0.4, for positions such as Nanjing Road and Shazikou. This indicates that TSP is a major factor influencing the density of PAHs. The correlations between the 7 types of PAHs and SO_2 are relatively low, between 0.1~0.3 except for positions Sifang Hotel and Jiayuguan. The correlations between the 7 types of PAHs and NO_x are high (0.35~0.7) at positions of Red-Star and Jiayuguan. The correlation coefficients are all relatively low for other positions. This indicates that the main sources of PAHs come from oil burning and waste air emission from automobils.

CONCLUSION

The concentration of PAHs in the atmosphere aerosol at 6 sites in Qingdao alongshore was determined by HPLC with florescent detector. The annual average PAH concentration ranged from 16 to 308 ng/m^3 and their levels were found to be related with site-specific characteristics. In general, both the industrial signal and the downtown signal sampling sites showed relatively high level of PAHs. The sources of PAHs in the air of Qingdao were obtained by principal factor analysis (PCA) and correlation analysis. The main sources of PAHs come from waste air emission from the automobiles and oil burning.

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