Bull. Environ. Contam. Toxicol. (2003) 70:422–429

DOI: 10.1007/s00128-003-0003-7

Bulletin of Environmental Contamination and Toxicology

Organic Pollution of Street Dust in the Handan City, China

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Received: 22 December 2001/Accepted: 21 October 2002

Organic pollution is still a serious problem for the cities in developing countries. Handan City is located in the Hebei Province, North China. It covers an area about 65 km^2 and has one million citizens. Street dusts are the important pollution source of aerosol. Coal is the only heating energy for the city in winter. The dusts contain a lot of small coal particles. Coal is main organic pollution source. It can contribute polycyclic aromatic hydrocarbons (PAHs) and pollute the city.

PAHs are a group of chemicals that are formed during the incomplete burning of coal, oil and gas, garbage or other organic substances. PAHs can be man-made or occur naturally. PAHs pollution of atmospheric particles has been studied by many scientists (Youngblood and Blumer, 1975; Simonich and Hites, 1994; Yunker and Macdonald, 1995; Liu et al., 2000; Cheng, et al., 1998). It has been proved that some PAHs are carcinogens according to the public health statement of Agency for Toxic Substances and Disease Registry in 1990 (ATSDR). Many PAHs have been considered as regulated compounds by the National Toxicology Program's (NTP), the Toxic Substance Control Act of 1976 (TSCA), the Federal Insecticide, Fungicide and Rodenticide Act of 1972 (FIFRA), Resource Conservation and Recovery Act of 1976 (RCRA) and the Comprehensive Environmental Response, Compensation, and Liability Act of 1980 (CERCLA). However, street dusts, the main sources of atmospheric particles, have not been noticed by so many people. The research of street dusts can reveal the source of PAH and give information of organic pollution in cities.

MATERIALS AND METHODS

In March 1999, six samples of street dusts were collected on the cemented street of Handan City by using a small plastic brush and a small aluminum shovel. The samples were stored in glass bottles. The samples were taken from south to north across the city center (Fig. 1).

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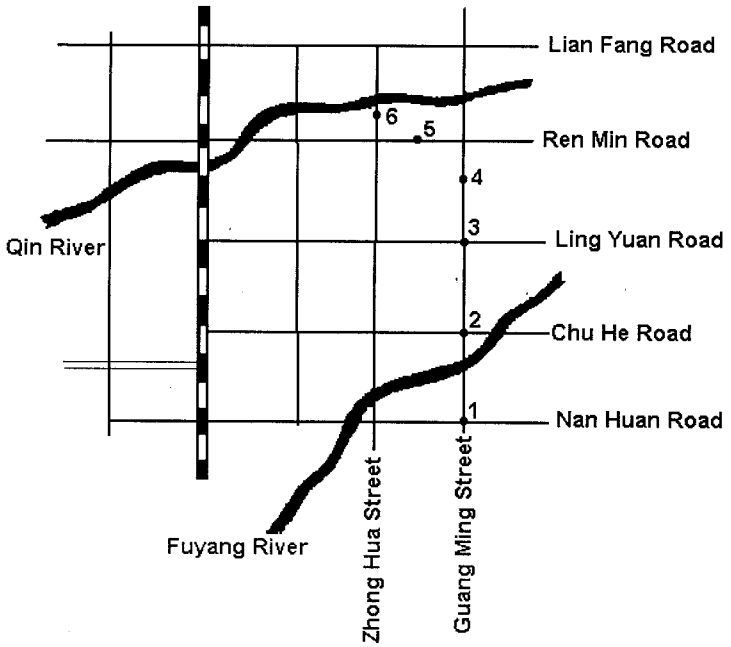

Figure 1. Sampling position in the Handan City

Geochemical analyses were carried out in the CNPC laboratory. For polycyclic aromatic hydrocarbon analysis, bulk samples were Soxhlet-extracted for 24 h using chloroform as solvent. Extract yields were determined gravimetrically after removal of the solvent. The extracts were separated into three fractions (hydrocarbon group separation) by column chromatography over pre-washed silica gel (70-230 mesh, 50×1 cm).

The gas chromatographic (GC) analyses were carried out on a HP-5890 gas chromatograph fitted with a silica capillary column (25m×0.2mm i.d.) coated with SE-54, temperature programmed from 60 to 320°C at 4°C/min.

GC/MS analyses were performed on a Finnigan-Mat SSQ70 GC-MS. A fused silica capillary column (25m×0.25mm i.d.) coated with SE-54 was used. The column temperature was programmed from 80 to 300 °C at 4 °C/min and was held at 300°C for 20 min. The ionizing voltage was 70eV and helium was used as carrier gas.

The organic carbon content (C_{org}) was measured using a Leco CR-12 carbon determinator. Carbonates were removed from the samples by prior treatment with concentrated hydrochloric acid.

Microscopic analyses were carried out in the laboratory of University Petroleum, Beijing. Organic petrographic composition were investigated on polished block samples using a Leitz MPV2 reflected light microscope equipped with a halogen lamp (oil 32/0.65, 548 nm, 3×3 μm, EMI9592 S-11).

RESULTS AND DISCUSSION

The results of organic geochemical analyses show in Table 1. Their contents are relative high.

Saturated hydrocarbon fraction was analyzed by gas chromatography. The compounds were identified and quantified (Table 2). The highest content reaches 1150 mg/kg in sample 2; the lowest content is 480 mg/kg in sample 3. The higher peak occurred in different parts of the GC traces (Fig. 2).

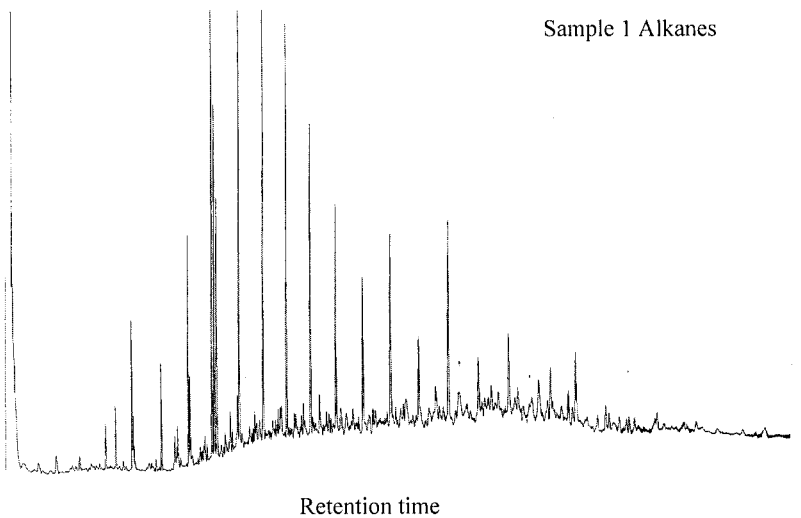

Figure 2. Selected GC trace of saturated hydrocarbon fractions in sample 1

Aromatic hydrocarbon fraction was further analyzed by GC and GC/MS. 41 aromatic compounds were identified and quantified (Table 3, Fig. 3). At least ten of them are carcinogens according to U.S. ATSDR public health statement of 1990. These compounds are anthracene, benzo(a)pyrene, chresene, fluoranthene, methylfluoranthene, benzo(b)fluoranthene, fluorene, phenanthrene, pyrene and naphthalene.

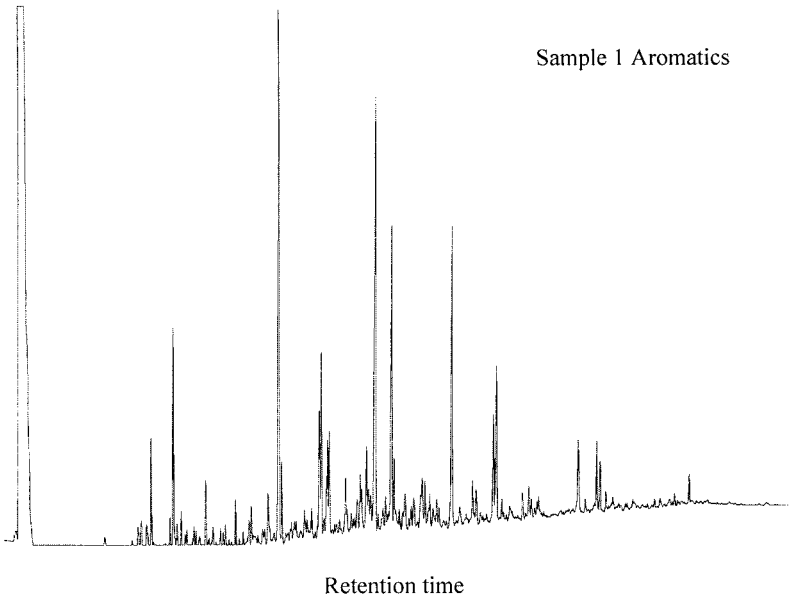

Figure 3. Selected GC trace of aromatic hydrocarbon fractions in sample 1

Table 1. Bulk organic parameters of the dust samples from the Handan City.

Analysis Number	Sample No.	TOC (%)	Extr (%)	Aro (%)	Alk (%)	Het (%)	Asph (%)
s2	number 1	5.66	0.44	15.33	31.56	32.44	20.67
s6	number 2	10.27	0.89	22.07	26.13	24.10	27.70
s5	number 3	4.76	0.73	14.47	25.50	25.86	34.18
s7	number 4	5.12	0.64	15.67	28.92	25.61	29.14
s3	number 5	4.78	0.45	13.53	29.95	24.64	31.88
s4	number 6	6.27	0.73	21.32	31.44	27.14	20.10
average		6.14	0.65	17.07	28.92	26.63	27.28

Abbreviations: TOC = total organic carbon; Extr = extract yield;
Aro = aromatics; Het = hetoros compound; Asph = asphalt.

Among these compounds benzo(a)pyrene belongs to strong carcinogenic PAH and has been studied by many people (Chen et al., 1999). Although the relative equivalency estimates for benzo(a)pyrene and the lifetime cancer risk assessments have been reported by many environmental scientists (Chen et al., 1999), few ambient standards have been set for the general public regarding exposure to PAH. The National Toxicology Program's (NTP), the toxic Substance Control Act of 1976 (TSCA), the Federal Insecticide, Fungicide and Rodenticide Act of 1972 (FIFRA), Resource Conservation and Recovery Act of 1976 (RCRA) and the Comprehensive Environmental Response, Compensation,

and Liability Act of 1980 (CERCLA) have suggested to regulate some individual PAH (Table 4). These compounds have reached very high value in the dust samples from the Handan City (Table 3). The highest benzo(a)pyrene content reaches 57 mg/kg in sample 2. Its average content is 16 mg/kg. The contents of benzo(b)fluoranthene are higher than the contents of benzo(a)pyrene. The highest content reaches 151 mg/kg in sample 2. Its average content is 42 mg/kg. The contents of fluoranthene and pyrene are even higher with average values of 125 and 79 mg/kg, respectively. The highest chrysene content reaches 140 mg/kg. The highest phenanthrene content reaches 350 mg/kg. The content of alkylated phenanthrenes reaches 130 mg/kg. The total identified PAHs reaches 2000 mg/kg in dusts, which can enter into air with wind and then enter into lungs of human and animal.

Table 2. Saturated hydrocarbons of the dust samples from the Handan City.

Analysis No.	S2 (mg/kg)	S6 (mg/kg)	S5 (mg/kg)	S7 (mg/kg)	S3 (mg/kg)	S4 (mg/kg)
Sample No.	Sample 1	Sample 2	Sample 3	number 4	Sample 5	Sample 6
C12	12.7	3.0		9.2	12.0	8.7
C13	11.7	3.0		5.4	6.5	6.8
C14	16.6	14.2	5.7	10.3	15.2	13.6
C15	39.1	60.8	25.2	45.2	66.3	40.7
C16	29.3	20.3	9.0	13.2	18.5	12.6
C17	61.6	40.5	17.9	15.5	14.1	14.5
C18	92.9	70.9	3.3	11.5	19.6	14.5
C19	120.3	81.1	39.9	32.1	25.0	16.5
C20	119.3	91.2	43.9	35	26.1	18.4
C21	118.3	111.5	65.9	51.6	38.0	20.4
C22	91.9	76.0	34.2	32.7	21.7	15.5
C23	62.6	65.9	33.4	25.7	19.6	17.5
C24	41.1	50.7	28.5	22.3	16.3	21.3
C25	50.8	76.0	65.9	45.4	25.0	31.0
C26	36.2	30.4	26.0	18.3	12.0	29.1
C27	43.0	60.8	43.9	33.1	22.8	35.9
C28	30.3	25.3	23.6	20.1	16.3	33.9
C29	40.1	35.5	27.7	22.8	19.6	33.0
C30	13.7	23.3	17.1	16.4	15.2	30.1
C31	10.8	20.3	20.3	16.5	13.0	24.2
C32	7.8	15.2	11.4	11.2	10.9	13.6
C33	6.8	14.2	9.0	8.7	9.8	10.7
C34		13.2	6.5	8.5	10.9	8.7
C35		11.1		5.4	8.7	5.8
pristane	23.5	15.2	6.5	3.3	5.4	7.8
phytane	68.4	48.6	21.2	17.5	14.1	13.6
average	1148.9	1078.4	585.9	536.9	482.6	498.5

Table 3. Aromatic hydrocarbons of the dust samples from the Handan City.

Analyse No.	S2 (mg/kg)	S6 (mg/kg)	S5 (mg/kg)	S7 (mg/kg)	S3 (mg/kg)	S4 (mg/kg)
Sample No.	Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6
naphthalene						8.4
2-MN						5.9
1-MN				0.2		0.7
2,6+2,7-DMN	0.3			0.4	0.6	2.0
1,3+1,7-DMN	1.4	2.8		1.5	2.0	5.5
1,6-DMN	1.9	3.1		1.8	0.9	4.2
2,3+1,4-DMN	2.3	2.8		2.2	1.2	3.0
1,5-DMN	8.0	14.2		8.2	3.7	15.5
1,2-DMN	0.3			0.2		1.0
MB	1.9	7.3		1.8	1.6	3.7
dibenzofuran	17.9	27.6		18.9	11.2	25.6
1,3,7-TMN	1.5	2.8		1.1	1.4	2.9
1,3,6-TMN	2.0	10.0		1.9	1.7	4.2
fruorene	4.4	16.8	2.5	4.5	1.1	7.4
MDBZ					2.7	
4-ethyl-1,6-DMN	3.0	6.9	2.6	3.2	3.3	5.9
DBT	4.7	29.9	5.4	4.3	2.1	4.4
phenanthrene	50.5	349.8	82.6	55.5	21.3	46.0
anthracene	5.2	30.7	8.1	5.7	2.9	5.1
3-MP	11.4	41.5	12.8	11.8	4.0	9.0
2-MP	13.1	49.4	15.4	13.5	4.5	11.4
9-MP	7.2	24.8	8.2	7.5	2.7	6.6
1-MP	6.8	22.8	7.9	6.3	3.4	7.4
2-PP	5.1	28.5	9.8	5.6	3.4	6.8
3,6-DMP	4.4		2.8	4.6	2.4	4.6
2,6-DMP	3.0		3.7	3.8	3.4	5.0
2,7-DMP	1.8	8.4	2.5	2.8	1.8	10.9
1,3+2,10+3,9+3,10-DMP	2.8	12.0	6.8	2.1	3.4	5.0
1,6+2,9-DMP	4.7	16.0	2.4	4.7	2.3	5.9
fluoranthene	48.0	417.3	148.9	54.0	31.0	48.7
pyrene	29.6	268.4	97.3	24.3	22.4	33.0
BNF	6.7	47.1	18.1	7.2	5.4	7.2
BNF	2.5		9.8	2.7	1.4	5.5
BNF	1.7	17.4	7.4	1.8	1.3	4.1

Table 3. Continued.

MF	2.5	8.9	7.3	3.5	2.6	9.1
MF	3.8	19.0	5.3	4.7	3.2	4.3
MF	3.7	11.3	6.7	3.7	5.7	3.9
triphenylene	10.3	108.7	41.2	12.3	9.9	13.3
chrysene	15.3	139.9	55.7	17.3	13.8	21.0
BZF	9.3	151.0	62.5	8.3	7.9	14.5
benz(e)pyrene	5.6	67.4	33.0	5.9	4.2	8.2
benz(a)pyrene	4.1	57.3	24.5	4.4	3.1	4.9
standard	28.4	86.7	46.0	32.4	26.1	66.7
total PAH	342.9	2048.4	722.5	358.4	233.0	436.0

Abbreviations: MB=methylbiphenyl; MN=methylnaphthalene;
DMN=dimethylnaphthalene; TMN=Trimethylnaphthalene;
MDF=methyldibenzofuran; DBT=dibenzothiophene;
MP=methylphenanthrene; phenylnaphthalene; DMP=dimethylphenanthrene;
BZN=benzo(b)naphtho(3,2-d)furan; MF=methylfluoranthene;
BZF=benz(b)fluoranthene.

Table 4. Carcinogenic PAHs.

Aro (ppm)	NTP	TSCA	FIFRA	RCRA	CERCLA
Naphthalene		Y	Y	Y	Y
fruorene	Y		Y		
Phenanthrene		Y			Y
3-Methylphenanthrene		Y			Y
Fluoranthene		Y		Y	Y
Pyrene		Y			Y
Chrysene		Y		Y	Y
Benz(b)fluoranthene	Y				Y
Benz(a)pyrene	Y	Y		Y	Y
total PAHs	Y	Y		Y	Y

Y=Regulated

The dominant PAH source may be from coal in the Handan City. The samples were taken on March 10, 1999. The heating boilers were still operating. Coal is only heating energy in the Handan City. Most coal particles are from coal transportation. These fine coal particles contain abundant PAHs. On the other hand, the incomplete burning of coal can also result in the formation of PAHs. The incomplete burning of gasoline of vehicles could be another source of PAHs.

In order to confirm the PAH source, the samples were built into polished blocks, which were further analyzed by use of microscope. Abundant coal particles can be clearly observed. The vitrinite reflectance (R_o) of the street samples was

determined on the polished samples. The R_o values vary from 0.75% to 2.5%. The reason is that the coal particles with different maturities were transported into the Handan City from different coal mines. The organic matter in the street dusts has similar maceral composition as in the heating coal. Vitrinite, inertinite and liptinite can be observed on the polished samples using microscope. The dominant maceral is vitrinite group.

Acknowledgments. This study was financially supported by the Deutscher Akademischer Austauschdienst (DAAD) and the Fundamental for Doctors.

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