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Distribution of platinum group elements in road dust in the Beijing metropolitan area, China

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Abstract

Dust samples collected from the Beijing metropolitan area (China) were evaluated to determine the distribution and the concentration of platinum group elements (PGEs). The dust particles that were smaller than 100 mesh size fraction (150 μm) were analyzed after aqua regia digestion. Concentrations of Pt, Rh, and Pd were found to be between 3.96 and 356.3 ng/g, 2.76 and 97.11 ng/g, and 0.1 and 124.9 ng/g, respectively, in the urban areas of Beijing, whereas for the background samples collected from the suburbs of Beijing, the concentrations of Pt, Pd, and Rh were very low and ranged from 0.1 to 0.9 ng/g, 0.5 to 1.4 ng/g, and 0.8 to 2.2 ng/g, respectively. The distributions of PGEs in road dust were an accurate reflection of the levels of pollution and were found to match with the local traffic conditions. A strong positive correlation was established among all the elements found in road dust. This suggests that emissions of abraded fragments from vehicle exhausts may be the source of the high concentration of Pt, Rh, and Pd in road dust along the main roads of Beijing.

Key words: platinum (Pt); palladium (Pd); rhodium (Rh); road dust; distribution; environmental pollution

Introduction

Autocatalytic converters containing Pt, Rh, and Pd have been successfully used for reducing emission of pollutants such as CO, hydrocarbons, and NO_x in exhaust by converting them to less toxic gases of CO_2, N_2, and H_2O (Palacios *et al*., 2000a). However, research on the distribution of platinum group elements (PGEs) has shown that the abrasion of catalytic converters causes an increase in the emission of microscopic particles containing low-solubility PGE (Moldovan *et al*., 2002; Bencs *et al*., 2003). Pt, Pd, and Rh that normally existed in the environment in trace amounts (in the g/kg range in road dust) are now ubiquitously distributed in urban areas (Gómez *et al*., 2002; Rumyana *et al*., 2003; Bruzzoniti *et al*., 2003; Whiteley and Murray, 2003; Niemelä *et al*., 2004). Therefore, traffic seems to be the main source of PGE contamination in urban areas.

Previously, PGEs were considered to have little effect on the biosphere. However, recent studies have shown that they may be toxic because they are found to accumulate in life forms including humans (Gómez *et al*., 2002; Moldovan *et al*., 2002), which is cause for concern. Moreover, it has been proved that some species of Pt, Rh, and Pd have high levels of biochemical activity, e.g. cytotoxicity and mutagenicity, and as they are emitted in the form of fine suspended particulate matter from converters, they may be inhaled and accumulated in the lungs of humans, leading to the risk of significant negative effects on human health after chronic exposure even at very low levels (Palacios *et al*., 2000b; Ravindra *et al*., 2004; EK Kristine *et al*., 2004).

In developed countries, the effect of PGEs emitted from catalytic converters on the environment and concern regarding the potential effects on human health have prompted more researchers to focus on the accumulation of PGEs in the environment including water, soil, plants, and dust (Rolf and Gerhard, 2001). Loading of autocatalytic converters containing PGEs has been made mandatory by law on newly manufactured vehicles for more than 20 years now in North America, Japan, and Europe (in USA in 1977; in Europe in 1980). Although many articles have been published by several research groups from different countries on the distribution of PGEs (e.g., Pt, Pd, and Rh) in the environment, to date, very few reports have been published from Asian countries and other developing countries.

Beijing, the capital of China, has a large population of more than 13.81 million. During the past decades, the number of vehicles has dramatically increased to more than 2 million. Loading of catalytic converters (Pt, Pd, and Rh) on gasoline-powered vehicles has been mandatory in Beijing since the late 1990s. Although the time that has elapsed since the installation of automobile catalytic converter has been made mandatory in Beijing is shorter than that in other developed cities, the monitoring of

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these metals and the study of their distribution in the environment are of prime importance not only for the present but also for the future.

The concentration of PGEs in the environment of Beijing was examined and evaluated. Dust samples were collected in 2002 and 2004. A procedure for the comprehensive and simultaneous determination of the distribution of Pt, Pd, and Rh in road dust and soils was developed using inductively coupled plasma mass spectrometry (ICP-MS). The limit of detection for Pt, Rh, and Pd was 0.2 ng/g, 0.2 ng/g, and 1.0 ng/g and the recoveries were 87.6%–104.7%, 93%–95%, and 98.2%–108.4%, respectively, which were satisfactory. The results show that the levels of PGEs in urban areas are obviously higher than those in background samples obtained from Wuling Mountain in Miyun District of Beijing and that the levels of PGEs in some areas showed an apparent increase in 2004 compared with those in 2002.

1 Experiment

1.1 Sample location and preparation

During the spring of 2002 and 2004, a total of 112 road dust samples between 0 and 2 cm below the surface were collected from urban areas with varying traffic conditions (Table 1 and Fig.1). Eleven background samples of 20 cm below the surface were collected with a 50×50 m^2 grid from the Wuling Mountain in the Miyun District, which is a suburb of Beijing (99 km far from the Beijing urban area), that is little influenced by the pollution of city transportation. Approximately three to four samples from different areas per sample location that totaled 200 g were collected, stored in plastic bottles, and labeled.

Table 1 Sampling locations, sample label and number, and traffic volume

Sample location	Labeled No. (n)[a]	Sampling year	Traffic volume[b] (vehicle/h)
The east second ring road	A1 (4)	2004	9660
The second south ring road	A2 (8)	2002	5835
	(6)	2004	
The west second ring road	A3 (8)	2002	11436
	(6)	2004	
The north second ring road	A4 (4)	2004	7206
The east third ring road	B1 (4)	2004	10794
The south third ring road	B2 (4)	2004	6954
The west third ring road	B3 (8)	2002	12798
	(6)	2004	
The north third ring road	B4 (8)	2002	12105
	(4)	2004	
The east fourth ring road	C1 (4)	2004	2130
The south fourth ring road	C2 (4)	2004	
The west fourth ring road	C3 (4)	2004	
The north fourth ring road	C4 (4)	2004	
The east fifth ring road	D1 (4)	2004	1314
The south fifth ring road	D2 (4)	2004	
The west fifth ring road	D3 (4)	2004	
The north fifth ring road	D4 (8)	2002	
	(6)	2004	
Miyun District	E (11)	2002	Little

[a] n is the number of samples collected from different areas in the same road; [b] Li and Tao (2000).

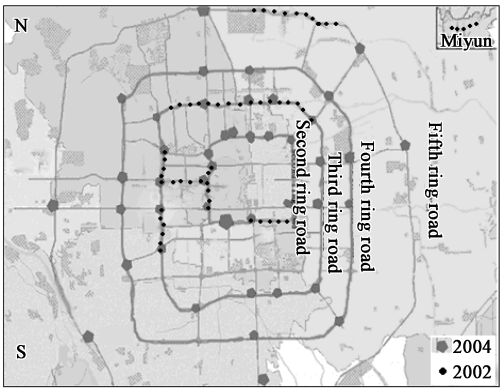

Fig. 1 Location map of sampling sites in Beijing metropolitan and suburb area in 2002 and 2004. The upright: Wuling Mountain in Miyun District is 99 km away from the center of Beijing.

All the samples were first air-dried for 24 h and then oven dried at 80°C for 4 h. The samples were crushed and homogenized for analysis (<100 mesh), using agate mortar and ball mill (QM-4H, Shanli Electronic Equipment Factory, Nanjing).

1.2 Instrumentation and reagents

Measurements were carried out using an ICP-MS spectrometer Agilent 7500c (Agilent Technologies Co. Ltd., USA). The optimum measurement conditions are shown in Table 2. Standard solutions of ^{7}Li, ^{59}Co, ^{89}Y, ^{140}Ce and ^{205}Tl (Agilent Co., USA) were diluted with 3% HNO_3 to 10 ng/ml to optimize ICP-MS performance. A total of 1 μg/ml solutions of ^{6}Li, ^{45}Se, ^{59}Co, ^{89}Y, ^{115}In, ^{159}Tb and ^{209}Bi (Agilent Co., USA) were also diluted using 3% HNO_3 to standardize the instrumental P/A factor.

A total of 100 ng/ml of Pt, Rh, Pd mixed standard solution was diluted using 3% HNO_3 to 0.005, 0.05, 0.5, 1, and 5 ng/ml to obtain the standard curve with r>0.999. Mixed standard solution of 1000 ng/ml of Pb, Zn, Cu, and Cd was diluted using 3% HNO_3 to 0.2, 1.0, 10, and 250 ng/ml to obtain the standard curve with r>0.999. The

Table 2 Measurement parameters of ICP-MS

ICP-MS parameter	
Sample introduction	
Temperature of spray chamber	2°C
Carrier gas	1.12 L/min
Blend gas	0 L/min
Plasma	
RF power forward	1400 W
RF power reflected	1 W
Tune	
CeO^+/Ce^+	<0.5%
Mass spectrometer	
Sampling cone	Nickel 1 mm orifice
Skimmer cone	Nickel 0.75 mm orifice
Measurement mode	Peak jumping (peak height)
Detected isotopes	^{103}Rh, ^{108}Pd, ^{195}Pt, ^{187}Re, ^{208}Pb, ^{66}Zn, ^{65}Cu, ^{111}Cd
Number of replicates	3
Internal standard	^{187}Re

Table 3 Average concentrations of Pt, Pd, and Rh in urban areas of Beijing*

	Year	Pt (ng/g)	Pd (ng/g)	Rh (ng/g)
A1	2004	169.19±43.5	31.94±30.22	43.29±19.85
A2	2002	81.97±49.34	41.20±14.49	20.42±10.22
	2004	143.35±11.48	18.55±18.88	26.4±6.22
A3	2002	110.84±46.04	44.04±14.23	24.82±7.90
	2004	165.96±35.58	18.38±4.82	29.51±4.98
A4	2004	204.19±133.79	59.37±56.06	53.46±38.01
B1	2004	100.43±26.48	15.59±8.00	21.89±2.89
B2	2004	87.11±24.92	26.7±15.14	21.02±7.37
B3	2002	72.39±49.89	30.84±13.11	14.58±8.26
	2004	100.88±17.76	19.02±14.81	22.81±6.73
B4	2002	78.21±59.41	29.96±11.01	12.44±4.59
	2004	75.64±15.94	19.58±9.74	20.94±5.22
C1	2004	67.07±46.96	17.71±8.65	11.22±2.70
C2	2004	88.55±12.99	37.46±24.87	30.48±16.24
C3	2004	30.32±24.88	3.05±2.00	6.25±2.76
C4	2004	30.95±11.00	11.19±6.28	10.14±5.67
D1	2004	15.5±1.24	1.4±1.84	12.62±9.90
D2	2002	18.28±6.56	10.69±2.41	5.06±1.13
	2004	6.54±0.85	4.77±3.07	2.77±0.01
D3	2004	6.16±3.11	2.72±2.32	3.34±0.54
D4	2002	36.67±15.54	48.71±37.14	8.74±2.74
	2004	7.91±1.08	4.15±3.94	3.23±0.67
E	2002	0.75±0.25	1.02±0.31	1.49±0.46

*Values are mean±*SD* (in each sampling region, 2–8 samples from different locations were collected, with a distance of more than 100 m separating one sampling location from the other).

contribution of interference to the mass of ^{108}Pd could be subtracted according to the following equation:

$$^{108}\text{Pd: } (108) \times 1 - (114) \times 0.03098 + (118) \times 7.2 \times 10^{-4} - (107) \times 4.573 + (109) \times 4.724$$

Therefore, the mass of ^{108}Pd had been calculated mathematically using Equation (1) and EPA 200.8. The corrections for the interference of HfO^+ to Pt were introduced as well.

The standard solutions of metal elements were purchased from Agilent (Agilent Technologies Co. Ltd., USA). Milli-Q water (18.2 MΩ/cm) was used for the experiment, the reagents and solvents used for digestion were of purest quality available, and 3% HNO_3 diluted by 70% HNO_3 was used in this experiment (BV-III level, The Institute of Chemical Reagent, Beijing, China).

1.3 Measurement of samples

Each sample was digested in aqua regia and analyzed using ICP-MS, with three replicates for each sample. Twelve milliliters of aqua regia ($HCl:HNO_3$=3:1) was added to a 150-ml conical beaker contained 2.5 g of sample. After being allowed to stand overnight, the sample was digested at 120°C for 3 h and then heated to near dryness at 200°C. Five milliliters of water was added to the residue and heated at 100°C for 10 min. The final solution was diluted to 100 ml. After 1:10 dilution, measurements were carried out immediately. The experimental conditions are listed in Table 2. All glassware used in the experiment was washed with hot aqua regia before use.

2 Results and discussion

2.1 Analytical results of PGEs

Although sample heterogeneity maybe one of the potential problems encountered during the analysis of Pt, Pd, and Rh concentrations, all the samples analyzed showed significant increase in the concentration of PGE compared with the background samples (E). The average concentrations of PGEs in the different sampling locations are shown in Table 3. The statistical parameters of analytical results are shown in Table 4.

2.2 PGEs abundance vs. traffic volume

The results in Table 3 and Fig.2 show that the sampling site with the highest traffic volume also had the highest concentrations of PGEs, which was in agreement with the results of other studies (Cicchella *et al*., 2003). As shown in Fig.2, the more the number of cars, the higher is the content of Pt, Pd, and Rh. In the urban areas of Beijing, the density of traffic is the highest in the second ring road and the third ring road, whereas the density of traffic in the fourth ring road comparatively less. Because the fifth ring road has not been completed, the stream of automobiles and the consequent density of traffic were less. Pt, Pd, and Rh show a similar general oblique distribution.

The results shown in Fig.2 also showed that there was a variation in PGE contamination levels. Pt was the major contaminant in every sampling location. The average concentration of Pt in urban areas was about 100 times higher than that in the background samples, which is similar to a previous report by Gómez *et al*. (2002). This result may reflect the earlier introduction of Pt-based catalysts in Beijing or may have resulted from the differences in

Table 4 Statistical analytical results of dust samples

	Pt (ng/g)		Pd (ng/g)		Rh (ng/g)	
	2002	2004	2002	2004	2002	2004
Samples	40	72	40	72	40	72
Mean±*SD*	15.89±8.22	97.16±69.24	34.21±16.42	19.92±21.04	22.74±17.06	79.66±52.42
Range	4.21–39.48	3.96–356.3	6.61–115.82	1.6–124.09	2.76–97.11	10.4–235.95
Median	13.72	90.52	32.58	13.73	20.31	69.04
Geometric mean	13.94	66.67	30.84	11.91	17.09	63.97
Detection limit	0.2		1		2	
Recovery	87.6%–104.7%		98.2%–108.4%		93%–95%	
Background value in Beijing area[a]						
Mean± *SD*	0.73±0.3		1.02±0.3		1.49±0.5	
Range	0.1–1.9		0.5–1.4		0.8–2.2	

[a]Collected from Wuling Mountain, Miyun District (99 km away from Beijing urban area), as reference background samples in 2002.

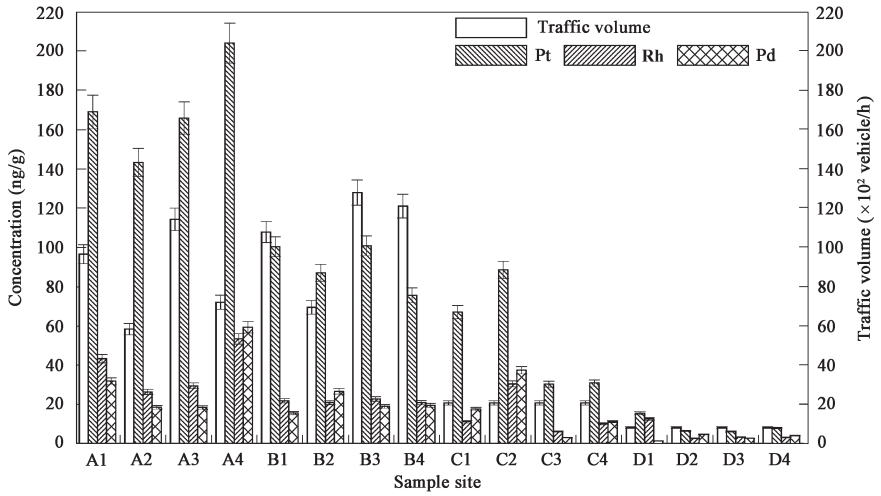

Fig. 2 Relationship between the distribution of Pt, Rh and Pd and traffic volume in Beijing urban areas in 2004.

PGEs loading during manufacture of vehicles, which may be indicative of Pt being the main catalytic component. The results indicate that concentrations of PGE is in the order Pt ≫ Rh and Pt ≫ Pd and that Rh and Pd had similar distributions.

2.3 Comparison of PGE concentrations between 2002 and 2004

The comparison of levels of PGE between 2002 and 2004 (Fig.3) showed that Pt and Rh concentrations in road dust had increased over the period of 2 years because of the rapid increase in the number of vehicles in Beijing. The results obtained from D2 and D4 areas are different from those obtained in other areas probably due to the fact that the D region (the fifth ring road) had not been completed until 2004.

2.4 Correlations between PGEs

If vehicular traffic is the main source of PGEs in the environment, the relationship among platinum group elements in urban areas will show positive correlations that could be stronger than those among platinum group elements in the suburbs.

Fig.4 is a series plot of correlations between Pt, Pd, and Rh including all the samples collected in 2004, in which Figs.4a1, b1, and c1 are the samples from urban areas and Figs.4a2, b2, and c2 are the samples from the Wuling Mountain in Miyun District, which is far from the modern city of Beijing and is economically backward and has a consequent lesser density of automobiles. Figs.4a1, b1, and c1 indicate the positive correlation of concentrations of Pt, Pd, and Rh, the concentrations of majority of urban road dusts fall within the same field and have similar ratio values, whereas no obvious ratio trend existed in farm land soils in the outskirts, as shown in Figs.4a2, b2, and c2. In urban samples, the relationship of Rh-Pt levels is slightly stronger than that of Rh-Pd and the correlation coefficients are 0.9396 and 0.9082, respectively. The correlation between Pd and Pt is the weakest, with R^2 = 0.8118. In samples of farm soils, all correlation coefficients were less than 0.6, with a range of 0.22 to 0.52. From the results shown in Fig.4, it can be concluded that anthropogenic inputs, especially vehicular traffic, in the downtown environment are the main source of the elevated concentrations of Pt, Pd, and Rh. Generally speaking, automobile manufactures do not publish the ratios of Pt, Pd, and Rh of the catalytic converters loaded to the vehicles that they manufacture; the catalytic converters are constantly improved to obtain optimized performances. These ratios vary for cars manufactured by the same or different manufacturers within the same country or from one country to another. Therefore, it could not be compared whether the ratios between PGEs in Figs.4a1, b1, and c1 agreed with those of catalytic converters. The results obtained from samples collected in 2002 showed a similar correlation.

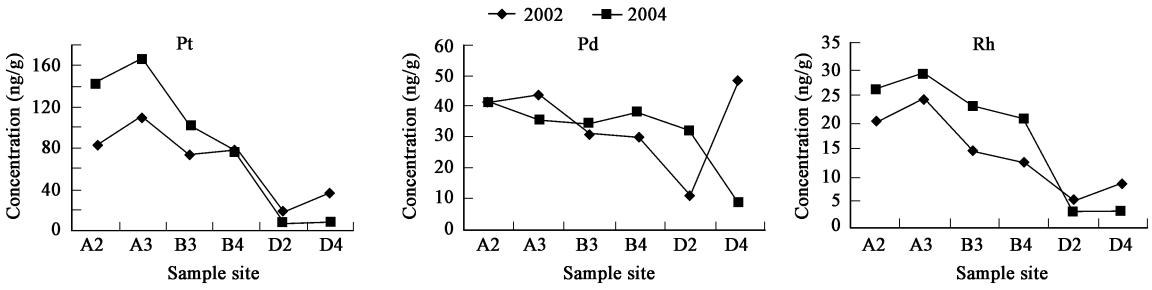

Fig. 3 Comparisons of Pt, Rh and Pd concentrations in samples collected from urban areas of Beijing in 2002 and 2004.

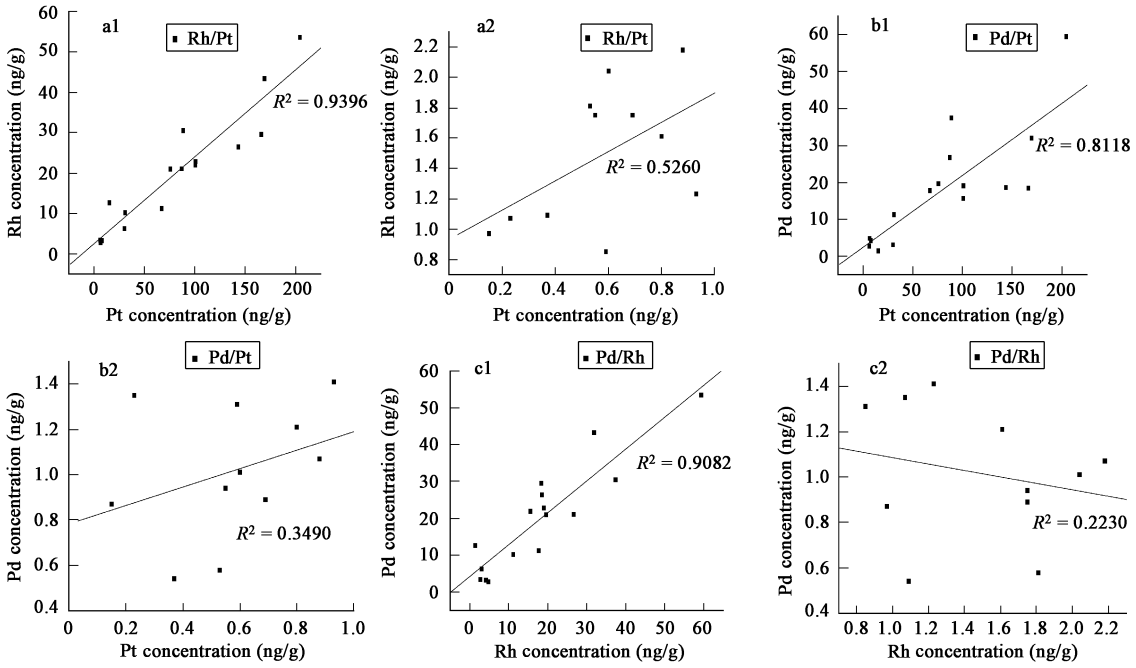

Fig. 4 Correlations between Pt, Pd and Rh collected from different areas in Beijing. a1, b1 and c1 were samples from urban area; a2, b2 and c2 were background reference samples from a mountainous area.

Table 5 Average contents (mean±*SD*) of Cu, Zn, Cd, and Pb in road dust collected from Beijing, China

Site	Cu (μg/g)	Zn (μg/g)	Cd (μg/g)	Pb (μg/g)
Second ring road	54.02±15.92	241.93±62.56	0.78±0.20	49.94±27.47
Third ring road	122.00±80.48	329.83±145.90	0.74±0.21	41.37±9.63
Fourth ring road	46.71±15.62	289.62±202.39	0.74±0.18	33.53±5.86
Fifth ring road	31.98±8.91	213.85±144.6	0.48±0.13	29.14±11.05

2.5 Correlations of PGEs with other metals

The advantage of using ICP-MS is that in addition to PGE, the concentrations of Pb, Cd, Cu, and Zn could be determined simultaneously, which have been used to evaluate the contribution of anthropogenic input to the urbanized environment. Table 5 shows the concentrations of Pb, Cd, Cu, and Zn in the selected samples that had high PGE levels in the A, B, C, and D regions. It can be seen that some of the elements such as Zn and Cd remain relatively constant in street dust and others such as Pb and Cu have the similar trend with the traffic volume. Table 6 shows the correlation coefficients of selected trace metals (Zn, Pb, Cd, Cu and Pt, Pd, Rh). Although some studies have shown a strong positive correlation between the PGEs and the above-mentioned metals (Cicchella *et al*., 2003), in our studies none of these metal elements showed mathematically strongly correlation with any of the PGEs except Pb. The average concentrations of Pt, Pd, Rh, and Pb in the road dust collected from every main road (the Second ring, Third ring, Fourth ring, and Fifth ring road) are shown in Fig.5. It is well known that Pb and Cu are often used to evaluate the pollution caused by emissions from motor vehicles, and levels of Zn and Cd influence of the corrosion of building materials. The presence of high levels of Pb was not an unexpected result in Beijing because lead additives had not been forbidden by law until recently and the total prohibition of the use of lead-added petrol is yet to be achieved. The strong correlation between PGE and Pb has further proved that vehicular traffic is the

Table 6 Correlation coefficients of distribution of Zn, Pb, Cd, Pt, Pd, and Rh in road dust of Beijing metropolitan area in 2004

	Pt	Pd	Rh	Zn	Pb	Cd	Cu
Pt	1						
Pd	0.95	1					
Rh	0.99	0.93	1				
Zn	0.02	0.09	0.02	1			
Pb	0.98	0.90	0.98	0.98	1		
Cd	0.62	0.82	0.61	0.23	0.23	1	
Cu	0.09	0.11	0.08	0.72	0.57	0.23	1

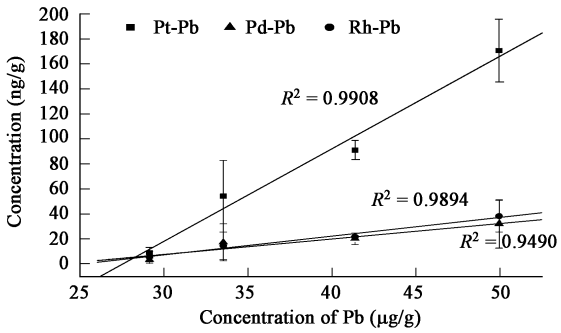

Fig. 5 Relationship between PGEs and average contents of Pb in dust among different ring roads in urban areas of Beijing in 2004 (Mean±*SD*).

main source of PGEs in the environment.

3 Conclusions

In conclusion, the distribution and concentration of the PGEs in the road dust in Beijing indicated that vehicular traffic were mainly responsible for pollution by PGEs, and automobile exhaust is the major source of elevated levels of PGE. Using ratios and associations with other metals, PGEs in road dusts can be attributed to catalytic converter attrition. The concentration level of PGEs appeared to be in the following order: Pt ≫ Rh and Pt ≫ Pd, and Rh and Pd have the similar distributions.

A direct relationship between PGEs concentrations and traffic volume could be established. There is a strong positive correlation in the group Pt-Pd-Rh in road dusts collected from urban areas of Beijing, although their release in the environment is from totally different types of cars, irrespective of whether the cars are equipped with a catalytic converter. The high Pt levels observed in dusts may reflect the earlier introduction of Pt-based catalysts in Beijing or may have resulted from the differences in PGE loading during manufacture of vehicles, which may be indicative of Pt being the main catalytic component.

Further research is required to better understand the behavior of PGEs emitted from autocatalysts in the urban environment of Beijing. PGE mobility, species, plant transfer coefficients, as well as bioaccumulation need to be studied further.

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