



Platinum group elements in the precipitation of the dry region of Xinjiang and factors affecting their deposition to land: the case of Changji City, China

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ABSTRACT

Platinum group elements and their compounds are a class of incident allergens and some platinum group element compounds also have carcinogenic effects. They accumulate in city environment as a result of emissions from catalysts used for vehicle exhausts. In this study, sixteen precipitation samples were collected on the north campus of Changji University located in the center of Changji. They were analyzed for palladium (Pd), platinum (Pt), and rhodium (Rh) by inductively coupled plasma–mass spectrometry. The average concentrations of Pd, Pt, and Rh were found to be 26.73 ng L⁻¹ (range: 3.18–84.25 ng L⁻¹), 1.71 ng L⁻¹ (range: below the detection limit to 6.38 ng L⁻¹), and 1.49 ng L⁻¹ (range: below the detection limit to 3.53 ng L⁻¹), respectively. Pd deposition was most pronounced for single precipitation events, reaching 35.47 ng m⁻² (range: 1.27–101.10 ng m⁻²), followed by Rh (max. 4.96 ng m⁻², range: 0–14.85 ng m⁻²) and Pt (max. 1.38 ng m⁻², range: 0–7.66 ng m⁻²). Both Pd and Pt were higher in winter than in other seasons in terms of their wet deposition amounts and their concentrations in precipitation, whereas Rh was lower in winter. Moreover, the results indicated that discharge from coal combustion in winter, the amount of precipitation, and the number of dry days before rainfall events all significantly affected the wet deposition amount and precipitation concentration of platinum group elements. Pd deposition flux was highest (reaching 5.47×10³ ng m⁻²) corresponding to 18 and 16 times the Rh and Pt fluxes, respectively. Finally, vehicle exhaust catalyst emissions from motor vehicles were not the only source of atmospheric platinum group metals in the city environment; in fact, combustion of coal in winter was found to be the dominant contributor of Pt and Pd in the atmosphere.

Keywords: Platinum group elements, city environment, precipitation, wet deposition, influence mechanism

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1. Introduction

Platinum group elements (PGEs) and their compounds are a class of high-incidence allergens and some PGE compounds also have carcinogenic effects (Zereini et al., 1998; Kalavrouziotis and Koukoulakis, 2009; Orecchio and Amorello, 2011; Zereini et al., 2012; Mihaljevic et al., 2013). Palladium (Pd), platinum (Pt) and rhodium (Rh) are widely used in vehicle exhaust catalysts (VEC), in order to control vehicle exhaust and improve the air quality. Pt and Pd are involved in the oxidation of hydrocarbons and CO, while Rh is used in the reduction of nitrogen oxides (NO_x). Indirectly, they also result in a decrease of environmental lead pollution because these converters require the use of unleaded gasoline. The catalytic converters were first used in North America since 1970s, then used in Europe (early 1990s) (Barbante et al., 2001), and widely used after 2000 in China. This indeed resulted in a significant decrease of various chemical species in urban air pollution. However, they continue to be released into the environment after exhaust gas purification for particulate matter (Charlesworth et al., 2011; Orecchio and Amorello, 2011; Zereini et al., 2012; Mihaljevic et al., 2013). There has been much relevant research on PGEs (Zereini et al., 1998; Kalavrouziotis and Koukoulakis, 2009; Bonanno, 2011; Charlesworth et al., 2011; Mathur et al., 2011; Orecchio and Amorello, 2011; Zereini et al., 2012; Mihaljevic et al., 2013) and also a number of related domestic reports (Wang et al., 2007; Liu and Wang, 2011; Zhu et al., 2011; Li and Gao, 2012; Wang and Li, 2012; Pan et al., 2013), most of which were focused on the urban road environment with

multi-media PGE temporal distribution factors, analysis of sources, analytical testing methods and other researches. PGE migration and transformation in the environment of arid regions have rarely been reported. Dry and wet deposition processes reduce the long-term accumulation of pollutants in the air, and they are important factors in maintaining a relatively stable atmospheric composition. Both dry and wet deposition of pollutants carry input to the surface and thus the terrestrial and aquatic ecosystems have been impacted (Grantz et al., 2003; Zhan et al., 2012). Due to a small amount of precipitation, wet deposition removes pollutants effectively, so the wet deposition of atmospheric pollutants has a profound significance (Grantz et al., 2003; Muezzinoglu and Cizmecioglu, 2006).

The city located in Xinjiang China which is in the center of Asia and Europe, and is known as the “Silk Road” of the new North Road. It has a typical continental arid climate, and the predominant wind direction is southwest. The annual precipitation is only about 200 mm. The heating period for the year is from October to April because of the cold winter, and coal has been the primary fuel used for heating. By the end of 2012, there were 140 000 vehicles in the city. As there is no large industry in the city, heating and vehicle emissions are the main sources of air pollution. Based on this information, in order to provide a reference for the in-depth study of urban environmental geochemical processes, we chose Changji in Xinjiang as a study area for the preliminary study of the wet deposition characteristics of PGEs in an arid urban atmosphere.

2. Materials and Methods

2.1. Sample collection

The selection of the sampling site was based on several factors, including ease of access, safety, minimizing potential for sample contamination, and representativeness. Sampling points were established on the Changji College North Campus. Based on the above criteria, the site chosen was Changji, which is located on the north slope of the Tianshan Mountains and the south margin of Junggar basin. It is at longitude 86°24'–87°37' and latitude 43°06'–45°20', which is in the center of Asia and Europe. Although the study area was in the downtown area of the city, it was more secluded and had less human interference than other areas in the city. The map of the study area is shown in Figure 1. The sampling period was one year (December 2011 through December 2012), and there were 87 precipitation events in total (40 rainy days, and 47 snowy days). Based on factors such as precipitation characteristics, six snowfall and ten rainfall events were selected for sampling (Table 1). The sampling container was polyethylene; it was washed with dilute nitric acid before precipitation to ensure that each sampling tool was free of pollution, and the container was secured while sampling. After the end of each precipitation, samples were frozen in polyethylene bottles at a temperature of -1°C . A total of sixteen samples were collected.

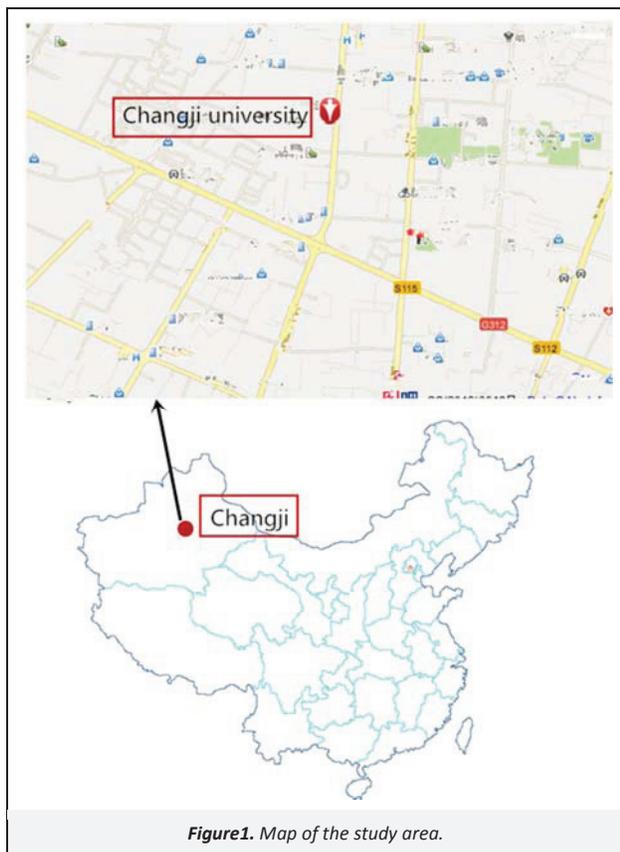


Figure 1. Map of the study area.

2.2. Sample analysis

Main instruments and reagents. The main instruments and reagents used were as follows: TPC-3000 (Yankee Environmental System, YES), a home-built cyclone impactor, an Agilent ICP-MS 7500Ce inductively coupled plasma mass spectrometer, and a Milli-Q water purification system. The internal standard stock solution was prepared using the Re standard and the tuning solution was a thallium mixed standard solution preparation

consisting of lithium, cobalt, yttrium, and cerium (Agilent, no. 5184-356).

Experimental methods. TPC-3000 (Yankee Environmental System, YES), and the home-built cyclone impactor were placed on a concrete platform with the sample intake at roughly 1 m above the surface. As soon as the precipitation sensor indicated the beginning of a precipitation event, the sampler was opened by an electric motor. The collection vessel was normally kept covered with a lid during periods of no precipitation. Samples were removed from the freezer and allowed to melt naturally at room temperature. Then the samples were analyzed directly after passing through the 0.45 μm membrane water. When we applied ICP-MS to determine the trace elements of Pt, Pd, and Rh in the samples, we found that Pt and Rh were interfered by small mass peaks. ^{108}Cd , ^{68}Zn , ^{40}Ar , ^{92}Mo , ^{16}O , ^{90}ZrO , ^{65}Cu , ^{40}Ar , ^{36}Ar , ^{69}Ga , ^{88}Sr , ^{17}O , and other interferences were generated on Pd, which could cause some difficulty when trace Pd is determined accurately. Therefore, in order to measure PGEs in samples accurately, Pt and Rh were determined using the normal mode while the ORS was used to measure Pd. ICP-MS conditions were optimized for the two selected modes before determination. The main operating parameters of the instrument were as follows: RF power 1 450 W, cooling gas 15.0 L min $^{-1}$, auxiliary gas 1.0 L min $^{-1}$, carrier gas 1.06 L min $^{-1}$. The isotopes of ^{103}Rh , ^{108}Pd , and ^{195}Pt were used for the analysis. The calibration standard solutions were prepared from a standard stock solution by dilution with 5% nitric acid. Detection limits of Rh, Pd, and Pt were 8.15 $\times 10^{-4}$, 7.14 $\times 10^{-3}$, and 6.71 $\times 10^{-3}$ ng g $^{-1}$, respectively. Their recoveries were more than 85%, and the relative standard deviation was less than 2.5%.

3. Results and Discussion

3.1. The PGE concentration in precipitation and influencing factors

PGE concentrations in precipitation. The average concentration in the precipitation of Pd was 26.73 (3.18–84.25) ng L $^{-1}$. In that range, the highest value occurred in the snowfall on January 17, 2012, and the lowest value occurred in the rainfall on June 3, 2012 and October 7, 2012. The average concentration of Pt was 1.71 (below detection limit, n.d.–6.38) ng L $^{-1}$, in which the highest value occurred in the snowfall on January 17, 2012 and Pt concentrations were below the detection limit in all rainfalls. The average concentration of Rh was 1.49 (n.d.–3.53 ng L $^{-1}$), in which the highest value appeared in the rainfall on April 29, 2012, and the lowest value appeared in the snowfall on December 31, 2011, February 6, 2012, and February 7, 2012, respectively. The Rh concentrations were all below the detection limit in these three snowfall events (Figure 2).

Seasonal variation of PGE concentrations. The seasonal variation trends of Pd and Pt concentrations are close to each other. They are higher in winter and lower in other seasons and they are higher in the snow and lower in the rain (Figure 2). The trend of Rh is the opposite of Pd and Pt, which is lower in winter and higher in other seasons, while it is lower in the snow and higher in the rain, except for sample 2-1 (Figure 2).

Influencing factors. Meteorological conditions are the main factors that influence the atmospheric pollution (Valiulis et al., 2002; Gunawardena et al., 2013). There were only long dry periods lasting 12 days before the snowfall in January 17, 2012, and a small amount of rainfall (only 1.2 mm). PGE accumulation reached a high level for a long dry period, and PGE concentrations reached a high level because of a small amount of precipitation. This may be the main reason for the highest concentration of Pd and Pt in the snow on January 17, 2012. There had been rainfalls on June 3, 2012 and October 7, 2012. Although the antecedent dry days were longer, the rainfalls were relatively larger, so they diluted the PGE concentrations. These are the main reasons why the concentration of Pd reached its lowest level in precipitation on June 3, 2012 and

October 7, 2012. According to these data, the rain characteristic is the main factor affecting the PGE concentration in precipitation. During the samplings, the predominant wind direction was southwest, and the direction was variable. The wind speeds were generally weak. Therefore, the present study did not address the effect of wind speed and direction. However, these factors could also be effective.

Several studies have identified automobile VEC emissions as the main source of PGEs in urban environments (Kalavrouziotis and Koukoulakis, 2009; Wang and Li, 2012; Zereini et al., 2012). During the sampling period, the traffic of Changji City was stable, which means that the PGE emissions were not highly variable. In addition, PGEs are relatively stable under all meteorological conditions, and the concentration changes in precipitation should tend to be consistent during similar weather conditions. However, this study

shows that Pd, Pt, and Rh concentrations are not completely consistent in precipitation, especially their seasonal variations. Although the seasonal changes of Pd and Pt are similar, their variation trend is exactly the opposite of Rh. In recent years, automobile VEC types changed from Pt type to Pd–Rh. Pd was the most common in motor vehicle emissions, followed by Rh while Pt is the least common (Wang et al., 2007; Bonanno, 2011; Charlesworth et al., 2011; Liu and Wang, 2011; Wang and Li, 2012). Pd/Rh is 3.67 (2.51–5.56) when automobile VEC emission is selected as the sole source in a road environment (Liu and Wang, 2011). In this study, the value of Pd/Rh is 17.94, which is far greater than the former. This shows that the urban atmospheric environment has other major sources of pollution, which are the main source of PGEs. In addition to exhausting car VEC emissions, there are other major source of Pd and Pt, and they may have seasonal determinants.

Table 1. Precipitation characteristics during sampling events

Map	Precipitation Data	Precipitation Form	Precipitation Amount ^a /mm	Time Length of Precipitation Accumulation	Dry Period Length Before Precipitation ^b /d
1	12.31.2011	Snow	0.6	3.5	7
2	01.17.2012	Snow	1.2	1.5	12
3	01.18.2012	Snow	1.2	4	1
4	02.01.2012	Snow	0.1	6	8
5	02.06.2012	Snow	0.8	Unknown	4
6	02.07.2012	Snow	1.1	Unknown	1
7	04.29.2012	Rain	2	3	6
8	05.19.2012	Rain	0.2	1	8
9	06.03.2012	Rain	4.2	5	13
10	06.18.2012	Rain	3.1	2	3
11	06.22.2012	Rain	2.4	6	5
12	09.15.2012	Rain	2	4	5
13	09.22.2012	Rain	14.7	13	5
14	09.30.2012	Rain	2.3	5	7
15	10.07.2012	Rain	14.2	11	4
16	10.19.2012	Sleet	5.9	8	11

^a Provided by Xinjiang Meteorological Bureau

^b Monitoring results of this research

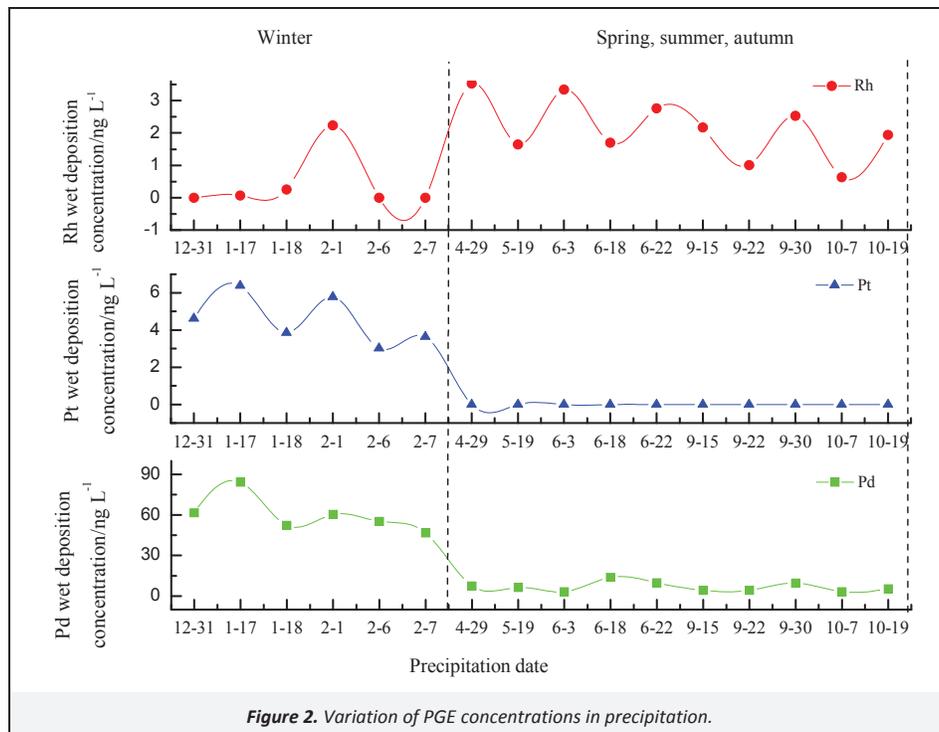


Figure 2. Variation of PGE concentrations in precipitation.

The source of Rh is stable and it mainly comes from automobile VEC emissions. In addition, the higher precipitation frequency and the shorter dry periods before rainfall result in smaller Rh accumulation in the atmosphere in winter. The Rh accumulation in the atmosphere is higher in winter, because the precipitation frequency is lower and the days of dry periods are generally longer before rainfall. These are the main reasons for the lower Rh concentration in wet deposition in winter, while it is higher in other seasons.

3.2. Variation of PGE wet deposition amounts and influencing factors

PGE wet deposition amounts. PGE wet deposition was calculated according to the following formula (Yang et al., 2009):

$$F_i = C_i \times 10^3 \times P \times 10^3 \quad (1)$$

where F_i indicates the amount of wet deposition for the element i (ng m^{-2}); C_i indicates the average concentration of element i in the precipitation (ng L^{-1}); and P is the amount of precipitation (mm).

The average deposition of Pd reached the highest level of 35.47 (1.27–101.10) ng m^{-2} in all precipitation events, followed by Rh and Pt with average fluxes of 4.96 (0–14.85) and 1.38 (0–7.66) ng m^{-2} , respectively. The highest wet deposition flux of Pd and Pt appeared in the snow on January 17, 2012. It is consistent with the concentration distribution in precipitation. The lowest amount of wet deposition of Pd appeared in the rainfall on May 19, 2012. The highest value of Rh appeared in the rainfall on September 22, 2012, June 3, 2012, and the lowest values were in the snowfalls on December 31, 2011, February 6, 2012, and February 7, 2012.

Seasonal variations of PGE wet deposition fluxes. The seasonal variations of deposition fluxes and PGE concentrations in precipitation were consistent. Pd and Pt appeared higher in the winter

and lower in other seasons; Rh appeared lower in the winter and higher in other seasons. In terms of seasonal variation of PGE concentrations in precipitation, seasonal changes in the Rh deposition fluxes are more obvious while the seasonal changes of Pd and Pt fluxes tend to be indistinct (Figure 3).

Influencing factors. When the pollutant concentrations in precipitation are certain, the smaller precipitation will remove fewer pollutants in the atmosphere. Pd concentration in precipitation was low on May 19, 2012. That was because the precipitation was small and removed little Pd in the atmosphere. That might be the main reason that the Pd wet deposition amount was the minimum. Pt concentrations in precipitation were generally below the detection limit in most of the seasons except for winter (Figure 3).

When the pollutant concentrations in precipitation are certain, the larger precipitation will remove more pollutants from the atmosphere. Rh concentration was low on September 22, 2012, but the rainfall reached 14.70 mm and was the largest rainfall during the study period. Rh concentration was high on June 3, 2012 and the precipitation was relatively high. These might be the main reasons that the Rh wet deposition flux was the highest on September 22, 2012 and June 3, 2012. Rh wet deposition flux was the lowest on December 31, 2011, February 6, 2012, and February 7, 2012, respectively. The small precipitation in winter and the low concentration of Rh in snow might be the main reason (Figure 3).

The Pd and Pt deposition fluxes were high in winter, while they were low in other seasons. The Rh deposition was low in the winter, while it was high in other seasons. The seasonal changes of wet deposition fluxes of PGE have suggested that Pd and Pt may have other important sources in winter in addition to VEC emissions. The precipitation was low in winter, while it was relatively high in other seasons. The concentration of Rh was low in winter, while the concentrations of Pd and Pt were high. The seasonal changes of the Rh deposition were more obvious, while the seasonal variations of Pd and Pt were not (Figure 3).

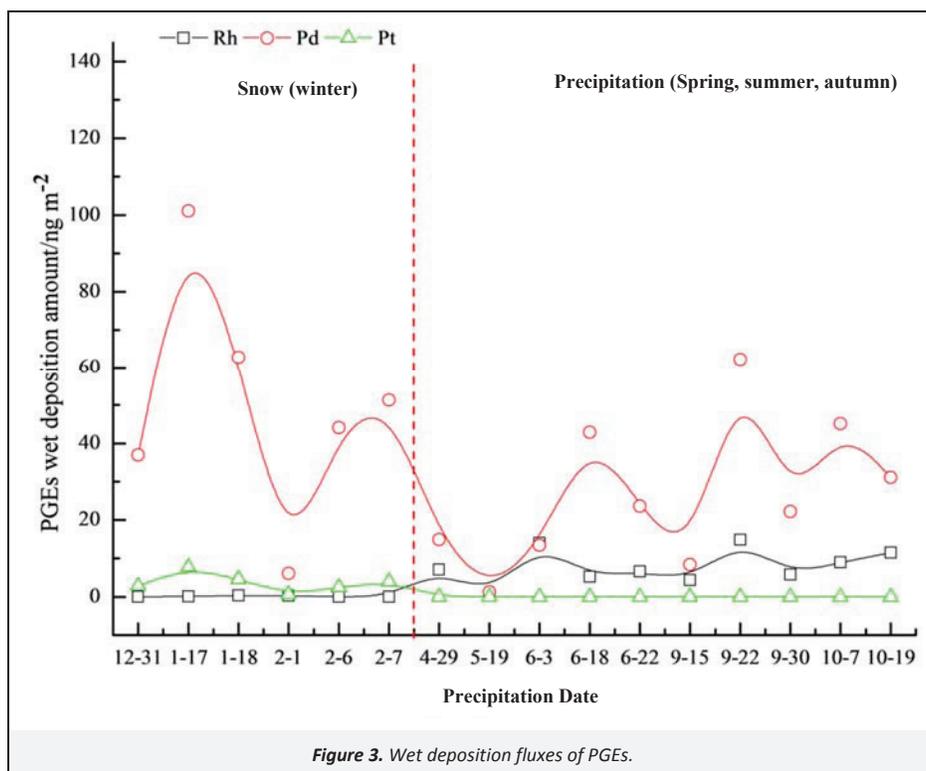


Figure 3. Wet deposition fluxes of PGEs.

3.3. PGE wet deposition fluxes

The annual average precipitation in Changji City is 204.6 mm. The calculation of deposition fluxes according to Equation (1) shows that the annual flux of Pd is maximum, at $5.47 \times 10^3 \text{ ng m}^{-2}$; next is Pt, for which the annual deposition flux is $3.50 \times 10^2 \text{ ng m}^{-2}$; the third is Rh, which reaches $3.05 \times 10^2 \text{ ng m}^{-2}$. The deposition flux of Pd is much greater than Rh and Pt, and the deposition flux of Pt is greater than Rh. The study of the emissions of VEC indicates that PGEs differ from the current car VEC types, which is mainly type Pd–Rh (Wang et al., 2007; Bonanno, 2011; Charlesworth et al., 2011; Liu and Wang, 2011; Wang and Li, 2012).

3.4. PGE sources in the city atmosphere

Previous research studies have confirmed the enrichment of PGEs in coal, which is mainly for Pd and Pt (Dai et al., 2003; Wang et al., 2008). Although different regional distribution of coal resources would cause a different content of trace element, several studies have shown that Pd and Pt were enriched in coal (Dai et al., 2003; Yang et al., 2007; Wang et al., 2008; Xia et al., 2009). Among them, the study by Yang et al. (2009) on the coal mining area of Guizhou Puan showed that the content of Pd was higher than the average mass fraction of Pd in China's largest PGE deposit (metal super large Cu Ni sulfide deposit).

Related research on Urumqi has shown that in addition to the emission of vehicle VEC, there may also be other major sources of Pt (Liu and Wang, 2011). The concentrations of PGE wet deposition in Changji City showed that Pd and Pt concentrations were high in winter while low in other seasons, and deposition flux of Pt was greater than Rh. This indicates that Pt and Pd have other sources in the atmosphere of Changji, and the coal used for residential heating in winter may be the main reason that the concentration of Pd and Pt was high in winter and the annual deposition flux was large. The emissions from VEC were not the only source of atmospheric PGEs in Xinjiang City, coal burning in the winter is one of the main sources, and it has a larger contribution to Pt and Pd concentrations in the atmosphere.

Most of the previous studies concluded that PGEs were mainly from VEC emissions (Kalavrouziotis and Koukoulakis, 2009; Li and Gao, 2012; Wang and Li, 2012; Zereini et al., 2012). That is because many research areas are in a warm climate, without heating or multiple forms of heating in winter, and there is a relatively short period for coal combustion in winter. Xinjiang is cold in the winter (there is a heating period of more than six months), and coal is always the main fuel used for heating. This may be the main reason why the present research is not consistent with the findings of previous research.

It is obvious that the source of pollutants, seasonal changes, precipitation amount, and the dry period before precipitation are the key factors that affect the PGE concentrations in wet deposition.

4. Conclusions

PGE wet deposition amount and concentrations in rainfall indicated that Pd and Pt were high in winter while low in other seasons; Rh is low in winter while high in other seasons. Pd in deposition settling flux was much larger than Rh, while Pt is much larger than Rh. PGE wet deposition fluxes and concentration in precipitation varied with the seasonal changes of sources, and they increased with the amount of precipitation and the dry period length before precipitation.

VEC emissions are not the only main source of PGEs in the city environment, and residential coal combustion in winter makes a larger contribution to Pd and Pt in the atmosphere.

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