

Original Research

Ecological Risks and Spatial Distributions of Heavy Metals in Beijing Atmospheric Dust

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Abstract

To investigate ecological risks and spatial distributions of heavy metals in atmospheric particles in the Beijing urban area, atmospheric dust was collected from 62 sites and 12 heavy metals in their <63 μm fractions were measured. Results indicate that V, Cr, Mn, Co, Ni, and Ba in the dust are basically unpolluted by human activities; whereas Cu, Zn, As, Cd, Sb, and Pb are moderately to heavily polluted. These metals (not including Ba) in most dust show a high ecological risk (RI = 809 ± 585, ranging 291-4,848), predominantly contributed by Cd (71%) and Sb (14%). Relatively higher risks mainly occur in the eastern and northwestern urban areas. The high risks in the east are caused mainly by Cd pollution associated with coal-burning and industrial and traffic activities, while those in the northwest are caused mainly by Sb pollution associated with point pollution sources and Cd pollution from traffic activity. This is significant for controlling atmospheric heavy metal pollution over the Beijing urban area and investigating atmospheric metal pollution in other cities.

Keywords: atmospheric particulate pollution, heavy metal, geo-accumulation index, ecological risk, spatial distribution, source

Introduction

Owing to the rapid development of industrialization and urbanization over the past several decades in China, atmospheric particulate pollution has become more and more serious in large cities, posing great risks to urban ecosystems and human health [1-3]. As an important component in urban ecosystems, atmospheric particles not only pose risks to biological components such as human beings, animals, and plants, etc., but also influence

the quality of water, soil, and sediment after being deposited onto the earth's surface [4-5]. Generally, risks of the atmospheric particulate pollution to ecosystems and human beings are largely caused by toxic pollutants adhering to particles. Among pollutants, heavy metals, which are hard to metabolize in a living body and do harm to circulatory and central and peripheral nervous systems [6-7], constitute one of the most important types in various environmental media.

Beijing, located northwest of the North China Plain, is one of the most seriously polluted cities for atmospheric particles in China [8-10]. It is not only the nation's capital, but also is one of the most densely populated cities in China,

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so investigating heavy metal pollution in atmospheric particles in the city is significant for protecting the urban ecosystem and human health and improving China's image. In fact, in recent years the heavy metal pollution of atmospheric particles in Beijing has aroused great attention, and a lot of valuable work has been done to investigate their concentrations, pollution levels, sources, temporal variations, controlling measures, and so on [8, 10-15]. However, until now few studies have examined the detailed spatial distributions and ecological risks of the heavy metals in atmospheric particles based on high-spatial-resolution sampling over the Beijing urban area due to the relatively heavy workload of the traditional filter membrane sampling method.

Under calm or weak wind conditions, atmospheric particles can gradually be deposited onto the ground and other object surfaces. At locations seldom disturbed by wind, rain, and human activities, the settled particles can accumulate as what is called atmospheric dust (fall). Generally, the atmospheric dust can maintain most physicochemical characteristics of atmospheric particles and thus are often used to investigate regional atmospheric particulate pollution, for it is relatively easier to be collected [7, 15-17]. In this study atmospheric dust samples were collected from 62 sites in the Beijing urban area, and 12 heavy metals were detected in their <63 μm fractions. The objects of this study are to:

- 1) Assess pollution levels and ecological risks of these heavy metals in atmospheric particles in the Beijing urban area.
- 2) Reveal spatial distributions of the risks.
- 3) Identity areas and metals with the highest risks and determine their main polluting sources.

The study is significant for not only establishing reasonable measures for air-quality management in the Beijing urban area, but also understanding atmospheric particulate pollution in other cities in China.

Experimental

Sampling and Analysis

Atmospheric dust samples were collected from 62 sub-areas over the Beijing main urban area in spring 2015 (Fig. 1). To obtain dust samples with certain regional representativeness, relatively large residential quarters that are relatively far away from obviously polluting sources were usually chosen for dust sampling. Generally, dust was collected mainly from uncleaned windowsills, flat roofs, and surfaces of pipeline installed on buildings that are seldom disturbed by wind, rain, and human activities and usually more than five meters above the ground. At each sampling site dust was collected from several sub-sites using a small brush or a dry-type vacuum cleaner and these sub-samples were mixed as one at last [15].

After being taken back to the laboratory, <63 μm fraction of each dust was isolated by passing a 250 mesh (~63 μm) sieve and only this fraction was used

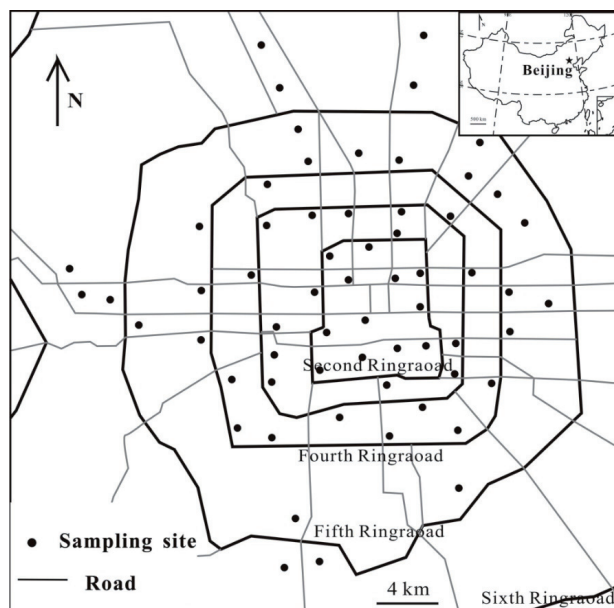


Fig. 1. Sixty-two sampling sites for atmospheric dust in the Beijing urban area.

for heavy metal analysis [15, 18]. After being fully dried in an oven, about 0.125 g dust was weighed and hot-digested with concentrated HNO_3 , HClO_4 , HF , and HCl in a Teflon beaker on a hot plate. Concentrations of Co, Ni, Cu, As, Cd, Sb, and Pb in dust were determined by an Agilent 7700x inductively coupled plasma mass spectrometry (ICP-MS) and V, Cr, Mn, Zn, and Ba by a Leeman Labs Profile inductively coupled plasma atomic emission spectrometry (ICP-AES). Procedural blanks, standard reference material (GBW07309), and duplicates for every ten-sample were used to control the quality of these measurements. The determined concentrations of these elements in the blanks were <1% of the average corresponding element values. The detected concentrations of these elements in the reference material were about $100 \pm 8\%$ of the standard values.

Geo-Accumulation Index

Geo-accumulation index (I_{geo}) of heavy metals is calculated by [19]:

$$I_{\text{geo}} = \log_2[X_i/(1.5X_{\text{ib}})] \quad (1)$$

...where X_i represents the concentration of metal i in atmospheric dust and X_{ib} is that in the local (Beijing) natural soil [20-21].

According to Müller [19], the I_{geo} indexes can be divided into seven classes as follows: 1) $I_{\text{geo}} \leq 0$, practically unpolluted; 2) $0 < I_{\text{geo}} \leq 1$, slightly polluted; 3) $1 < I_{\text{geo}} \leq 2$, moderately polluted; 4) $2 < I_{\text{geo}} \leq 3$, moderately to heavily polluted; 5) $3 < I_{\text{geo}} \leq 4$, heavily polluted; 6) $4 < I_{\text{geo}} \leq 5$, heavily to extremely polluted; and 7) $I_{\text{geo}} > 5$, extremely polluted.

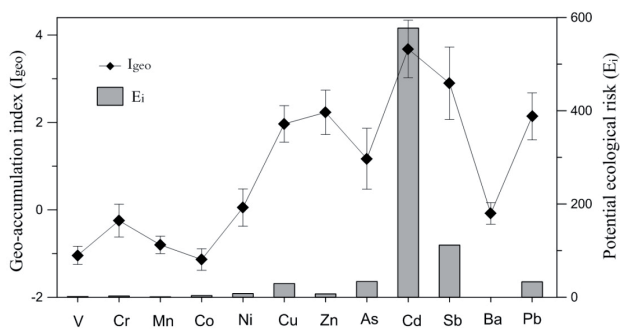


Fig. 2. Geo-accumulation indexes (I_{geo}) and potential ecological risks (E_i) (not including Ba) of heavy metals in the $<63 \mu\text{m}$ fractions of the 62 atmospheric dust.

Potential Ecological Risk Assessment

Potential ecological risks are assessed by the following equations [22]:

$$E_i = T_i \times \frac{C_s^i}{C_b^i} \tag{2}$$

$$RI = \sum E_i \tag{3}$$

...where E_i , the potential ecological risk of metal i in the dust; T_i ($\text{Cd} = 30, \text{As} = \text{Sb} = 10, \text{Co} = \text{Ni} = \text{Cu} = \text{Pb} = 5, \text{V} = \text{Cr} = 2, \text{Mn} = \text{Zn} = 1$) [15, 22-24], the toxic-response

factor of metal i ; C_s^i , the concentration of metal i in dust; C_b^i , the background value of metal i in the Beijing natural soil [20-21]; and RI , the potential ecological risk of all the measured metals in the dust.

The ecological risk levels can be divided into five classes [22, 25]: $E_i < 40, RI < 150$, low ecological risk; $40 \leq E_i < 80, 150 \leq RI < 300$, moderate ecological risk; $80 \leq E_i < 160, 300 \leq RI < 600$, considerable ecological risk; $160 \leq E_i < 320, 600 \leq RI < 1,200$, high ecological risk; and $E_i \geq 320, RI \geq 1,200$, very high ecological risk.

Results and Discussion

Levels of Heavy Metals in Atmospheric Dust

Concentrations of the 12 heavy metals in the $<63 \mu\text{m}$ fractions of the 62 atmospheric dusts from the Beijing urban area are shown in Table 1. The mean concentrations of V, Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Sb, Ba, and Pb in the dust are 0.7, 1.3, 0.9, 0.7, 1.6, 5.9, 7.0, 3.4, 19.2, 11.2, 1.4, and 6.6 times that of the local (Beijing) natural soil, respectively [20-21]. The pollution levels of these 12 metals decrease in the following order according to their I_{geo} values in Fig. 2: $\text{Cd} = \text{heavily polluted} > \text{Sb} > \text{Zn} > \text{Pb} = \text{moderately to heavily polluted} > \text{Cu} > \text{As} = \text{moderately polluted} > \text{Ni} = \text{slightly polluted} > \text{Ba} > \text{Cr} > \text{Mn} > \text{V} > \text{Co} = \text{practically unpolluted}$.

Among the twelve heavy metals, the mean concentrations of V, Cr, Mn, Co, Ni, and Ba are basically

Table 1. Statistical results for concentrations of heavy metals in the $<63 \mu\text{m}$ fractions of the 62 atmospheric dusts from the Beijing urban area. In addition, mean concentrations of these metals in the Beijing natural soil and in dust from other typical cities in China.

Metal	Min mg/kg	Max mg/kg	Mean mg/kg	Stdev	Soil mg/kg	Changchun mg/kg	Xi'an mg/kg	Wuhan mg/kg	Shanghai mg/kg	Changsha mg/kg	Guangzhou mg/kg	Hongkong mg/kg
V	36.0	73.0	57.9	7.9	79.2		84.8		59.6			25.5
Cr	45.0	179.0	86.0	24.8	68.1	96		130	242	403.5	176.2	324
Mn	373.0	796.0	607.1	79.4	705.0	692	581		904		539.8	639
Co	6.3	16.9	10.6	1.8	15.6		19.4	14	12.1			10.2
Ni	24.0	109.0	45.2	16.1	29.0		56.7	33			41.4	
Cu	84.0	356.0	138.4	49.6	23.6	68.4	102.7	138	141	126	192.4	534
Zn	382.0	2,156.0	722.7	335.5	102.6	465.4	798	835	699	1,542	1,777.2	4,024
As	9.0	180.0	23.9	24.3	7.1	23.3	28.5				20.1	
Cd	0.9	18.8	2.29	2.3	0.119	0.624		1.6	0.9	2.5	2.14	1.8
Sb	4.0	110.1	12.3	14.9	1.1						14.8	
Ba	535.0	1,054.0	752.3	131.1	531		916					
Pb	71.0	396.0	167.9	68.2	25.4	93.6	266.3	281	148	348	387.5	240
Data Source	This study	This study	This study	This study	a	[26]	[27]	[28]	[29]	[30]	[31]	[29]

^a Data of V, Cr, Mn, Co, Ni, Cu, Zn, Sb, Ba, and Pb in the local (Beijing) natural soil are from CNEMC [20], and As and Cd from Chen et al. [21].

close to that of their corresponding values in the Beijing natural soil [20-21] (Table 1), respectively. And their I_{geo} values are generally below or close to zero (Fig. 2), indicating no or slight pollution of these six metals in the atmospheric dust. This fact suggests that these six metals in the Beijing atmospheric dust are derived predominantly from natural sources and not or only slightly affected by human activities.

In contrast, the mean concentrations of the other six metals of Cu, Zn, As, Cd, Sb, and Pb in the atmospheric dust are often obviously (3.4-19.2 times) higher than that of their corresponding values in the Beijing natural soil [20-21] (Table 1). Besides, the I_{geo} values of Cu, Zn, As, Cd, Sb, and Pb are relatively higher, which are 2.0, 2.2, 1.2, 3.7, 2.9, and 2.1, respectively (Fig. 2), indicating moderate to heavy pollution levels of these six metals. This fact suggests that these six heavy metals in the atmospheric dust are often significantly affected by human activities and derived mainly from anthropogenic sources.

Compared with dust from other typical cities in China (Table 1), the mean concentrations of most heavy metals in the Beijing dust are higher than that from Changchun [26] and roughly close to that from Xi'an [27], Wuhan [28], and Shanghai [29], but obviously lower than those from Changsha [30], Guangzhou [31], and Hong Kong [29]. This comparison suggests a relatively lower pollution level of heavy metals in the dust from Beijing among typically large cities in China, despite human activities being more intensive and economies more developed in Beijing compared with most other cities in China. The relatively low pollution levels of most heavy metals in the Beijing dust may be due to that as the capital of China the Beijing government has taken many effective measures in recent years to control environmental pollution, such as shutting down or relocating heavily polluting units to suburban areas or surrounding satellite cities, improving energy-consuming structure, etc.

Ecological Risks of Heavy Metals in Atmospheric Dust

From the potential ecological risk assessment results in Fig. 2, it can be seen that the mean E_i values of these 11 metals (not including Ba) in atmospheric dust decrease in the order of $Cd = 577 > Sb = 112 > 40 > As > Pb > Cu > Ni > Zn > Co > Cr > V > Mn$. Among the 11 heavy metals, Cd is of the highest E_i values with an average of 577, suggesting a very high ecological risk level. The second highest E_i is Sb, which averages 112, suggesting a considerable ecological risk level. The E_i values of these two metals, which are strikingly higher than that of the others, account for 85% of the total ecological risk of the 11 heavy metals in atmospheric dust. The mean E_i values of As, Pb, and Cu are 29-34, suggesting a low ecological risk level of these three metals. These three metals contribute 12% of the total ecological risk. For the other six metals (V, Cr, Mn, Co, Ni, and Zn), their mean E_i values are all below 10, indicating negligible ecological risks of these metals in the dust.

Compared with the pollution levels of these metals (Fig. 2), it can be found that Cd and Sb, which are of high pollution levels, show high ecological risks, while V, Cr, Mn, Co, and Ni, which are of low pollution levels, show negligible ecological risks. However, Cu, Zn, and Pb are also of relatively high pollution levels, but they show low ecological risks, which is caused by obviously lower toxic-response factors of these three metals compared with Cd and Sb. This fact suggests the importance of ecological risk assessment for finding out the most threatening elements in environmental media.

The total ecological risks (RIs) of these 11 metals

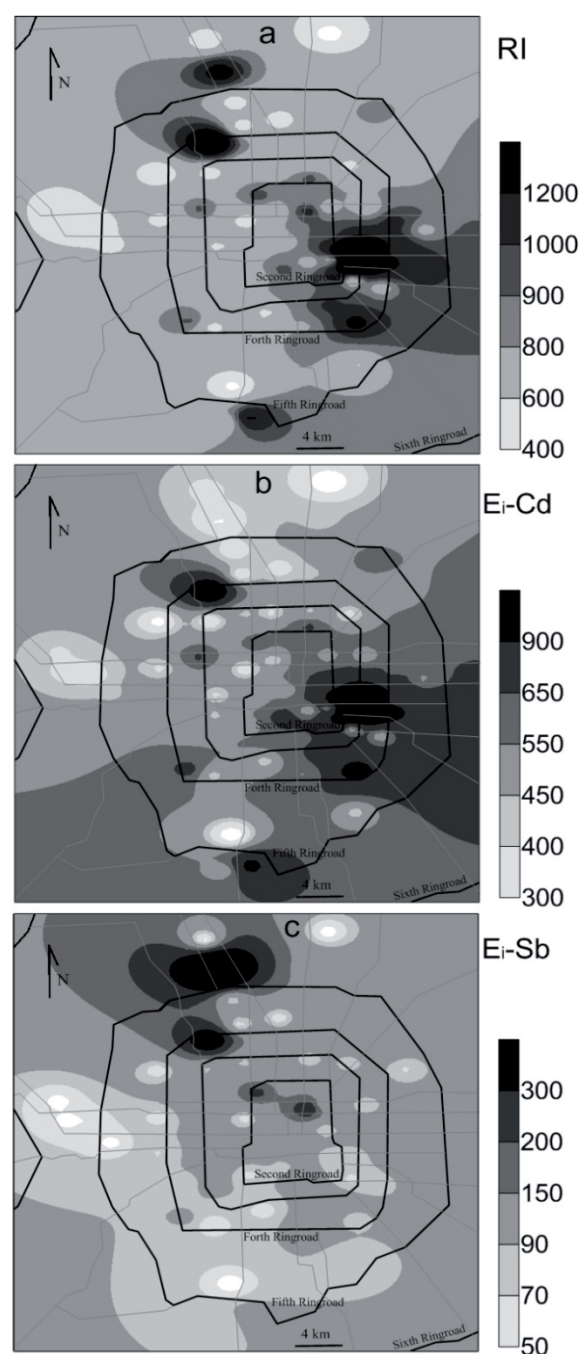


Fig. 3. Spatial distributions of potential ecological risks of heavy metals in atmospheric dust.

Table 2. Pearson's correlation matrix for concentrations of heavy metals in atmospheric dust.

	V	Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Sb	Ba	Pb
V		0	0	0	0.009	0.500	0.119	0.804	0.879	0.102	0.923	0.260
Cr	0.467**		0	0.106	0.025	0.015	0.500	0.752	0.438	0.886	0.291	0.215
Mn	0.699**	0.457**		0.001	0.054	0.785	0.206	0.987	0.478	0.539	0.967	0.845
Co	0.599**	0.207	0.419**		0.509	0.366	0.031	0.035	0.140	0.111	0.371	0.050
Ni	0.327**	0.284*	0.246	0.086		0.010	0.671	0.018	0.849	0.863	0.467	0.593
Cu	0.087	0.306*	0.035	0.117	0.325**		0.068	0.577	0.516	0.837	0.588	0.001
Zn	0.200	0.087	0.163	0.274*	0.055	0.233		0.831	0.923	0.856	0.028	0.095
As	0.033	0.041	-0.002	0.271*	0.303*	0.073	0.028		0.586	0.831	0.970	0.674
Cd	0.020	0.100	0.092	0.190	-0.025	0.084	0.013	0.071		0.648	0.945	0.123
Sb	0.210	0.019	0.080	0.204	0.022	0.027	0.024	0.028	0.059		0.231	0.001
Ba	0.013	-0.136	-0.005	0.116	-0.094	-0.070	0.280*	-0.005	0.009	-0.154		0.495
Pb	0.145	0.160	0.025	0.250	0.069	0.420**	0.214	0.055	0.198	0.414**	-0.088	

The left lower part is correlation coefficient; the right upper part is significant level.

** $P < 0.01$ (two-tailed); * $P < 0.05$ (two-tailed)

(not including Ba) are between 291 and 4,848, with an average of 809 ± 585 , suggesting a high ecological risk level of these heavy metals in atmospheric dust from the Beijing urban area. However, it should be noted that the above ecological risk results do not include other toxic elements (such as Hg), for they were not measured in this study. If considering these unmeasured toxic elements, the ecological risk of heavy metals in Beijing atmospheric dust must be higher. Even so, the above results are enough to suggest that the heavy metal pollution in the atmospheric dust poses great risks to the urban ecosystem in Beijing, and thus should be controlled as soon as possible – especially for Cd and Sb.

Spatial Distributions of Ecological Risks of Heavy Metals in Atmospheric Dust

Spatial distributions of the RIs of the 11 heavy metals in the atmospheric dust and that of Cd and Sb, which are of the highest E_i values, are shown in Fig. 3, which shows that relatively lower RIs of these heavy metals occur mainly in the western and northeastern urban areas in Beijing, while relatively higher RIs mainly occur in the eastern and the northwestern urban areas (Fig. 3a). The spatial distribution pattern is approximately similar to that of Cd (Fig. 3b), which is due to the dominant (71%) contribution of Cd to the RI.

Correlation analysis results suggest that Cd has no correlation with all the other metals (Table 2), implying that Cd pollution in atmospheric dust is likely affected by multiple polluting sources. Previous studies involving source apportionment of heavy metals in atmospheric particles from the Beijing urban area suggest different major sources of Cd, which may be due to the atmospheric particles being collected from different areas and during

different periods [8, 10, 12-13]. However, all of these studies indicate that the Cd pollution in atmospheric particles is usually associated with coal-burning, traffic, and industrial emissions. Based on the distribution of human activities (pollution emissions) in different Beijing urban areas, it can be further found that the highest RIs of Cd at the east second, third, and fourth ring roads are likely a consequence of relatively more coal-burning emissions [32] and traffic activities; the relatively higher RIs of Cd in the other eastern urban areas are likely associated with relatively more industrial activities [33] and coal-burning emissions [34]; and the relatively higher RI of Cd in the northwest Fourth Ring Road is probably due to relatively more traffic activities in this area. This information is significant for controlling heavy metal pollution in atmospheric particles in the Beijing urban area.

Among these 11 metals, the E_i of Sb, which contributes 14% of total ecological risk, is the second highest. From Fig. 3c) we can find that relatively lower RIs of Sb occur mainly in the western and southern urban areas, while relatively higher risks mainly occur in the northwestern urban area. Our previous study suggests that the enrichment of Sb in atmospheric dust is mainly associated with traffic-related emissions over the entire Beijing urban area [15]. Gao et al. [13] evaluates sources of toxic elements in total suspended particulates from the Beijing urban area and also finds the major contribution of vehicle emissions to Sb. However, the extremely higher RIs of Sb at the outside of the north Fourth Ring Road (Fig. 3c) may be associated with other point pollution sources such as waste incineration emissions, as suggested by Zhang et al. [8], for the traffic activities are moderate in this area. With respect to the other elements of V, Cr, Mn, Co, Ni, Cu, Zn, As, and Pb, they all show

low ecological risks and contribute no more than 15% of the total ecological risks, so their spatial patterns are not discussed in this study.

Conclusions

This study presents heavy metal results in the <63 μm fraction of atmospheric dust collected from 62 sites over the Beijing urban area. The results of 12 representative heavy metals suggest that V, Cr, Mn, Co, Ni, and Ba in the atmospheric dust show unpolluted or slightly polluted levels, indicating natural sources; whereas Cu, Zn, As, Cd, Sb, and Pb show moderately to heavily polluted levels, implying anthropogenic origins. The total ecological risks of these metals (not including Ba) in the atmospheric dust average 809 ± 585 , ranging from 291 to 4,848, suggesting high ecological risks of most dust samples. The risk levels of these metals in the dust decrease in the order of $577 = \text{Cd} = \text{very high ecological risk} > 112 = \text{Sb} = \text{considerable ecological risk} > \text{low ecological risk} = \text{As} > \text{Pb} > \text{Cu} > \text{Ni} > \text{Zn} > \text{Co} > \text{Cr} > \text{V} > \text{Mn}$. Relatively higher risks mainly occur in the eastern and northwestern urban areas. The high risks in the east are mainly caused by Cd pollution associated with coal-burning and industrial and traffic activities, while those in the northwest are mainly caused by Sb pollution associated with point pollution sources and Cd pollution with traffic activities. These results suggest that the heavy metal pollution in atmospheric particles poses great risks to the urban ecosystem of Beijing, and thus it should be controlled as soon as possible, especially for Cd in the eastern area.

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