Land Use-related Chemical Composition of Street Sediments in Beijing

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Abstract

Background. More than 10 million people are currently living in Beijing. This city faces severe anthropogenic air pollution caused by an intense vehicle increase (11% per year in China), coal combusting power plants, heavy industry, huge numbers of household and restaurant cookers, and domestic heating stoves. Additionally, each year dust storms are carrying particulate matter from the deserts of Gobi and Takla Makan towards Beijing, especially in spring. Other geogenic sources of particulate matter which contribute to the air pollution are bare soils, coal heaps and construction sites occurring in and around Beijing. Streets function as receptor surfaces for atmospheric dusts. Thus, street sediments consist of particles of different chemical compositions from many different sources, such as traffic, road side soils and industry.

Methods. Distributions and concentrations of various chemical elements in street sediments were investigated along a rural-urban transect in Beijing, China. Chemical elements were determined with X-ray fluorescence analysis. Factor analysis was used to extract most important element sources contributing to particulate pollution along a main arterial route of the Chinese capital.

Results and Discussion. The statistical evaluation of the data by factor analysis identifies three main anthropogenic sources responsible for the contamination of Beijing street sediments. The first source is a steel factory in the western part of Beijing. From this source, Mn, Fe, and Ti were emitted into the atmosphere through chimneys and by wind from coal heaps used as the primary energy source for the factory. The second source is a combination of traffic, domestic heating and some small factories in the center of Beijing discharging Cu, Pb, Zn and Sn. Calcium and Cr characterize a third anthropogenic element source of construction materials such as concrete and mortar. Beside the anthropogenic contamination, some elements like Y, Zr, Nb, Ce, and Rb are mainly derived from natural soils and from the deserts. This is supported by mineral phase analysis, which showed a clear imprint of material in road dusts coming from the West-China deserts.

Conclusions. Our results clearly show that the chemical composition of urban road dusts can be used to identify distinct sources responsible for their contamination. The study demonstrates that the chemistry of road dusts is an important monitor to assess the contamination in the urban environment. Chemical composition of street sediments in Beijing comprises the information of different sources of atmospheric particles.

Recommendations and Outlook. This study is only a small contribution to the understanding of substance fluxes related to Beijing's dust. More effort is required to assess Beijing's dust fluxes, since the dust harms the living quality of the inhabitants. Especially the measurable superimposing of long scale transported dust from dry regions with the anthropogenic polluted urban dust makes investigations of Beijing's dust scientifically valuable.

Keywords: Atmospheric pollution sources; Beijing; factor analysis; geogenic dust; street sediments, chemical composition; particulate matter; urban dust; West-China deserts; X-ray diffractometry (XRD); X-ray-fluorescence (XRF)

Introduction

Beijing, the Chinese capital, faces severe dust pollution due to several sources, such as dust import from western deserts, industry and power plants, domestic combustion processes, and traffic. During the last decades, the urban population has grown to more than 10 million inhabitants. The economical growth of China has also left its footprints in Beijing. New industrial zones and domestic areas were developed and those induced a growing traffic amount. Drought climate during almost the whole year, strong monsoons in wintertime carrying dusts from the Gobi desert, heavy traffic and high population density, lots of construction sites and bare grounds make Beijing a 'dust' city.

Dusts in urban areas are contaminated by heavy metals from industry, traffic, and domestic heating. Dust can be emitted directly to the atmosphere through combustion processes or through erosion, respectively corrosion and abrasion of surfaces followed by wind transport. Dust deposits and accumulates on surfaces such as soils or streets, where the dust forms street sediments. In the case of street sediments, it is obvious that traffic will be a main source of contamination. Heavy metals such as Cu, Sn and Pb typically characterize pollution by traffic. The emission of Pb decreased in the last years due to the introduction of the automobile catalyst. The catalyst is not fully introduced yet in China, but leaded petrol is no longer available in Beijing. However, even unleaded petrol contains Ph and there are other sources of Pb in traffic. Such sources are tires, brakes, lubricants, antifreeze, metallic and car paint particles, road surfaces and substances against ice forming on street surfaces (Grüer 1982, Liermann and Stegen 1985, Muschak 1990, Gih et al. 1990, Hurtig 1990, Unger and Prinz 1992, Bartz 1994, Böhel et al. 1999).

Traffic is one of the major contributors to dust pollution (Foner 1992, Heinrichs 1993, Andrade et al. 1994, Heinrichs and Brumsack 1997, Kleeman et al. 1998, Pio et al. 1998). Thus, traffic, together with other dust sources, contributes to urban dust pollution that is endangering human health (Eickmann 1993, Lahmann 1996, Jendritzki et al. 1999). Pearce and Crowards (1996) focused on the relationship between particulate matter and human health. Their study suggests that as many as 12,000 deaths in the UK might be attributed to total atmospheric particle concentrations, or about 7,000 deaths if only PM_{10} (particulate matter smaller than 10pm in diameter) is considered. The amount of dust deposition and the deposition of heavy metals severely increases towards the surface in the vicinity of streets. An investigation in Karlsruhe, Germany, showed that the deposition of Zn and Pb in 0.1 m height is 2-5 fold the amount of the deposition in 1.5 m height (Norra 1997). These results highlight the need to investigate the near ground deposition of particulate matter in regard to the impact on playing children, plants, and animals. Elements show specific solubility in different dust and street sediment fractions (Tanner and Wong 1999, Li et al. 2001, Wang et al. 2002, Striebel and Gruber 1997). Atmospheric dust deposits and can become part of street sediments. An investigation of dusts and street sediments from Karlsruhe shows a higher mobility of Zn in atmospheric dusts sampled in 1.5 m height compared to street sediments, whereas Pb is more mobile in street sediments (Heiser et al. 1999a and 1999b). The mitigation of environmental problems which are linked to urban dusts requires the knowledge about harmful substance fluxes in urban areas. Such knowledge is necessary for the management of harmful substance fluxes as a part of the sustainable development of urban areas, since atmospheric particulate matter is one pathway for harmful elements to pollute resources such as soils, groundwater, plants, and the human health (Stiiben and Norra 2000).

Studies on urban atmospheric particulate matter and on street sediments have been carried out in some Chinese cities, such as Shanghai (Shu et al. 2001), Wuhan (Waldman et al. 1991), Beijing, Shengyang, Lanzhou, Taiyuan (all in Ning et al. 1996), Nanjing (Wang et al. 2002), Hong Kong (Tanner and Wong 2000, Li et al. 2001), and Taipei (Mao and Chen 1996). However, there is a lack of knowledge on the impact of anthropogenic activities on chemical urban dust composition for Beijing with respect to the loads of trace elements. Street surfaces are acceptors for urban dust deposition that integrate over the whole assembly of urban pollution sources but will be dominated by traffic pollution. Furthermore, these surfaces themselves, with their sediments, are sources of dust due to wind erosion. Therefore, street sediment samples were taken along a rural-urban transect of Beijing to investigate the pollution of street sediments and to study whether pollution of other sources then traffic can be detected.

1 Material and Methods

1.1 **Sample locations and collection**

Road dusts were collected in the major traffic roads of Shijingshan Road, Fucheng Road, Wenjin Street, Jingshan Street, Wusi Street and Chaoyangmen Street along a west/east transverse through Beijing. At the western end of the profile in Shijingshan district, the steel factory 'Capital Steel Group Company' is located covering an area of about 10 square kilometers. This plant was built in *1950* and is one of the biggest steel-producing companies in China. Coal is used as the main energy supplier for the steel factory. The sampling section runs across the center of the city, that is an old town of about 1000 years history. Some small old style factories and metal workshops were located here. The eastern end of the section is a suburb residence area of lower population and traffic density compared to other places of Beijing.

In spring 2001, 31 samples of road dusts were taken in the middle of the mentioned roads with an average sample distance of about 1000 m (Fig. 1). Sampling locations were changed to parallel streets when the roads were recently rebuilt or revealed a new asphalt surface. About 200 g of material was collected with a brush from comparable areas (approx. 100 m^2) at each location.

1.2 **Chemical and mineralogical analyses**

The samples were air dried and sieved. The fraction smaller than 0.125 mm was ground in an achate mill. The concentrations of Ti, Ca, K, Cr, Ni, Ga and V in the samples were measured by wavelength-dispersive X-ray-fluorescence (WDXRF). Energy-dispersive X-ray fluorescence (EDXRF) was used to analyze the concentrations of Mn, Fe, Cu, Zn, Pb, As, Rb, Sr, Y, Zr, Nb, Sn, Ba, La and Ce. These analyses were carried out with a Tracor Xray Spectrace *5000* (EDXRF) and a Siemens SRS 303 AS (WDXRF) at the Institute of Mineralogy and Geochemistry, Universität Karlsruhe, Germany. Calculation of detection limits is based on counting statistics of corresponding elements provided by Kramar (1997). WDXRF provides lower detection limits for light elements than EDXRF, whereas EDXRF has lower detection limits for most of heavy metals (Kramar 1997) and the advantage of simultaneous analysis. The measurements are frequently checked at the Institute of Mineralogy and Geochemistry against international standard materials such as GXR2, Soil5, Soil7, MRG1, MAG and PCC1 (Geostandard Newsletter 1989, Norra 1997, Norra 2001, Norra et al. 2001). On the average, the recovery is better than $\pm 10\%$. Results of XRF analyses are checked against other analytical methods like Instrumental Neutron Activating Analysis, Inductive Coupled Plasma Mass Spectrometry or Atomic Absorption Analyses by several other authors (Kramar 1993, Bergfeldt 1995, Castro *1995,* Manz 1995). Results of XRF analyses do not show any significant deviations. Problems of recovery can arise in cases of As in the presence of high concentrations of Pb or, vice versa, due to interference of specific fluorescence energy levels. This interference is demonstrated with GXR2 (Table 1), in which the concentration of Pb is more than 25-fold the concentration of As (Table 1). However, such interferences are corrected using a partial least square algorithm which allows one to correct up to an intensity ratio of approximately 1:10 (Kramar 1982, Kramar

personal communication) as in the case of Soil5 (reference values - As: 94 mg/kg, Pb: 130 mg/kg; analyzed by Norra 2001 (n=27) – As: 86.9 mg/kg, Pb: 145 mg/kg) and Soil7 (reference values $-$ As: 13.4 mg/kg, 60 mg/kg; analyzed by Norra 2001 (n=40) -As: 14.3 mg/kg, Pb: 60 mg/kg). Element concentrations analyzed in samples were above the detection limits for all elements (see Table 1). The methods are described in detail, inter alia, by Kramar et al. (1982), Kramar (1993, 1997 and 1999), Hahn-Weinheimer et al. (1995), Lindon et al. (1999) and Weber Dieffenbach (2000). The main mineral phases were analyzed by X-ray diffractometry (XRD) at the Institute of Mineralogy and Geochemistry, Universität Karlsruhe, Germany that runs a Siemens Kristalloflex DS00. The method of X-ray diffractometry is sufficiently described in numerous publications such as by Allmann (2003).

1.3 Statistics

The investigation of the chemical composition of road dusts deals with a complex system of contamination from various sources, which are very difficult to identify and to study separately (Bityukova et al. 2000). In this case, factor analyses (Davis 1986, Reyment and Jöreskog 1993, Stoyan et al. 1997, StatSoft 1984-1995) represent an excellent method to investigate the relationships of variables in order to extract the most important factors responsible for the element distribution in road dusts. Factor analysis reduces the number of variables and detects structures in the relationships between variables. Factor analysis combines most correlating variables to factors. The whole set of analyzed elements was used in the factor analyses to identify the different sources of contamination, as well as the relationships and spatial distributions of elements in urban road dust. The factors were extracted by the maximum likelihood method and were varimax rotated, i.e. the extracted factors are rotated in such a way that the variance of the factor loadings becomes a maximum (Bahrenberg et al. 1992). Factor loadings and factor scores were calculated. Factor loadings represent the correlation between variables and factors. The contribution of the single factors to the total variance can be calculated. The factor with highest correlations between the variables explains most of the total variance. Factor scores represent the values of factors at individual cases (sampling sites). The factor analysis was processed with the computer program STATISTICA.

2 Results

Mineralogical analyses showed that the main minerals of the road dusts in Beijing are quartz, feldspar, calcite and dolomite. On one hand, the mineral sources are the deserts of the western parts of China, especially in the case of quartz and feldspar. Three or four sand- and dust-storms occur every year carrying material from the deserts to Beijing consisting of quartz and feldspar. On the other hand, construction materials of street surfaces and buildings are typical urban sources for mineral dusts. Abrasion processes due to mechanical stress (wind, traffic, construction) releases minerals such as calcite and dolomite (Blume *1993,* Norra 1997).

Table 2 displays the results of the chemical analysis in road dust samples and a summary of statistical parameters (minimum, maximum, median, mean, standard deviation, coefficient of variation). No extremely high contamination of street sediments with toxic elements was detected. Frequency distributions of concentrations do not show any intense skewness since differences between mean and median are relatively low for most elements. Elements are ordered according to corresponding atomic mass. Some elements show characteristic profiles of concentrations along the transect (Fig. 2). These profiles are obviously linked to three main different land use types. A steel works is responsible for high Fe concentrations in the western parts of Beijing (Fig. 2a). Diffuse urban pollution due to traffic, heating systems, industry, etc. causes higher concentrations of Cu in the city's center (Fig. 2b). Both sources, steel plant and diffuse pollution, emit Zn and cause a bimodal distribution (Fig. 2c). Furthermore, areas of lower pollution are situated between the steel works and Beijing City, as well as to the east of Beijing City.

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Fig, 2a: Distribution of Fe concentrations along the transect

Fig. 2b: Distribution of Cu concentrations along the transect

Table 3: Factor loadings (loadings > 0.5 marked bold)

Factor analysis was used to group the 22 elements analyzed in this investigation. Table 3 shows the loadings of the factor analyses. 5 factors were extracted. These five factors account for 68.8% of the total variance. The factors are

determined by specific element combinations. This is shown by the factor loadings > 0.5 which are printed bold. Factor scores were calculated for each sample site (Table 4).

3 Discussion

Land-use influences on the chemical composition of street sediments are obvious, as shown in Fig. 2. In the case of Beijing, main influences are the steel plant in the west and the diffuse pollution of many different sources such as traffic, domestic and industrial combustion processes and the general waste disposal along the roads. The latter source correlates with the population density and leads to a higher pollution in the city center. The impact of industrial point pollution sources on urban dusts and soils was also proved for other industrial units, such as lead smelters and ferrous metallurgy factories (Tyutyunnik and Gorlitskii 1998, Hrsak et al. 2000, Norra et al. 2001). Factor analysis has been carried out for a more comprehensive understanding of the different pollution sources. Table 3 lists the loadings of five factors.

Factor 1 is dominated by positive correlations with elements which are emitted by the steel factory. Those elements are Fe, Ti, Mn, and V. These elements are characteristic for the production of steel and alloys. Additionally, V can be assigned to pollution from coal combustion that is part in the steel production process. K and Rb show significant negative correlations with factor 1. The authors assume that this result reflects opposing processes of impoverishment and enrichment of element concentrations in the street sediments. When elements originated in the steel plant are enriched in the street sediments, other elements have to be relatively impoverished; in the case of Beijing Rb and K. These both elements are typical tracers for the clay fraction, which is an indication that especially elements of geogenic origin seem to be impoverished in this factor. Factor 2 comprises elements of geogenic origin, and the comparison of factor 1 and 2 shows that most of the elements of factor 2 show low or no correlations with factor 1. As expected, the scores of factor 1 (see Table 4) are high near the steel plant and decrease towards the city (Fig. 3a), as do the Fe concentrations (see Fig. 2a).

Factor 2 is correlated to elements of geogenic origin such as Y, Zr, Nb, Ce, La, Rb, and Ti. Also Ni is slightly correlated to factor 2, even if the factor loading does not reach the chosen trigger value of 0.5. These elements are a typical natural element association. Sources of these elements can be local soils. Wind can swirl up particles of those soils that subsequently accumulate on the street surfaces. The scores of this factor generally decrease from west to east, thus indicating a further source (Fig. 3b): The main wind direction is

west and, on this basis, it can be supposed that long-range, transported dust deposits when the friction for the air currents increases at the urban border where the urban boundary layer (Kuttler 1993, Helbig *1999)* lifts up. This longrange, transported dust originates from dry areas west of Beijing and from deserts (Gobi, Takla Makan) in the western parts of China (Weixi 2002). The decreasing scores reveal the decrease of the geogenic dust content from west to east. Most of this dust portion seems to be deposited with the beginning of the urbanized area characterized by its relatively high surface friction. Decreasing amounts of geogenic dust are left over to be deposited with increasing distance from the western city boundary. Along the rural-urban transect, geogenic dust will be filtered out due to the urban friction and the increasing sealing of the urban surface prevents the up-swirling of local soil material. Thus, the impact of this dust source decreases from west to east.

Cu, Zn, Ba, Pb, and Ni are elements of factor 3 showing loadings larger than 0.5. This factor can be interpreted as a diffuse pollution source. One important contribution to this factor will be automobile emissions. However, pollution by other sources, such as domestic and industrial combustion processes, street markets, construction activities, handling of bulk goods, corrosion, mechanical abrasion by traffic and wind, spilled liquids on the roads, and waste, contribute to factor 3 as well. Specific sources of Cu, Zn, Pb, and Ni are alloys, asphalt and concrete surfaces, tires, brakes, automobile bodies, paints, ashes from combustion of coal, and many other technogenic materials (Merian 1984, Muschak 1990, Nriagu and Pacyna 1988). Ba sources in urban areas are paint, rubber, plastic, paper, ceramic, bricks, etc. (Biichel et al. 1999). The factor scores show a very serrated course along the transect with slightly higher values in the city center and with some extremely high values at specific sites (Fig. 3c). This course underlines the erratic impact of sources other than traffic, since, in the case of traffic as a single pollution source, the pollution should constantly increase with the general traffic volume as demonstrated in factor 4.

Factor 4 is dominated by high loadings for Cu, Sn, and Sr. Cu and Sn are typically used in brakes. Muschak (1990) analyzed the abrasion of Cu from automobile brakes between 83.8 and 5026.1 g/(km-a) along different types of roads (residential way to motorway). A high density of cross roads and the intensive use of the brakes will lead to additional emissions. Another typical source of Cu are electronic devices, in which alloys of Cu and Sn are also used (Swertka 2002). Furthermore, Sn and Cu alloys are used in bearings. Other Sn containing technogenic materials are tinplate, solder, PVC, and paints (Merian 1984). Strontium is technogenically used for pigments against corrosion and in alloy with AI and Fe (Biichel et al. 1999). The course of the factor scores along the transect shows increasing values towards the city center. This course represents the increasing traffic volume with the increasing intensity of urban activity (Fig. 3d). An unexpected feature of this factor is the higher correlation of Pb with factor 3 than with factor 4. This can be explained with a strong impact of local pollution, as was analyzed at sample site 5 where the Pb concentration is 176 mg/kg and contributes to the scores of factor 3 (see Fig. 3c). Furthermore, leaded petrol is no longer in use in Beijing. Consequently, traffic emission of Pb is reduced and other sources have become relatively more important.

Calcium and Cr determine factor 5 (see Table 3). Factor 5 represents the occurrence of calcareous construction waste, such as particles of concrete, mortar, lime and limestone. The occurrence of Ca is especially evident along roads (Blume 1993, Norra 1997). The co-occurrence of Cr can also be attributed to concrete and mortar. Higher Cr concentrations are found in basic materials of concrete production such as limestone, clay, calcareous marl, ash, slag, and coal (Cavelier & Foussereau 1995, Verein Deutscher Zementwerke 1999). Fig. 3e illustrates the similar courses of factor 5 scores, with Ca and Cr demonstrating erratically higher and lower concentrations along the transect.

Fig. 3c: Scores of factor 3

4 Conclusion

The chemical composition (22 elements) of street sediments of some of Beijing major roads is influenced by different, superimposing particle sources. Factor analysis reduces the 22 variables to 5 factors that can be explained as the determining sources of sediments on Beijing's roads:

- 1. steel factory emissions
- 2. geogenic background, represented by dust from western dry areas and bare soils of the city
- 3. diffuse pollution from many different anthropogenic sources
- 4. traffic emissions
- 5. particles of construction materials

Four different courses of factor scores were observed along the rural-urban transect:

- 1. The steel factory only pollutes the sediments locally at the westem end of the transect.
- 2. Scores of factor 2 representing the geogenic background and the long range transported dust from dry regions decrease slowly from west to east along the transect.
- 3. Traffic emissions (factor 4), and partly emissions from diffuse pollution sources (factor 3), show the highest impact in the urban center.
- 4. Scores of factor 5 together with concentrations of Cr and Ca representing particles from construction materials are erratically distributed along the transect.

Factor analysis was not sufficient to distinguish between the single pollution sources in the case of diffuse pollution due to the relatively small number of samples. These pollution sources not only contaminate the street sediments, but also pollute the urban aerosol, soils and plants. This study can only be a small contribution to the understanding of substance fluxes related to Beijing's dust. More effort is required to assess Beijing's dust fluxes, since the dust harms the living quality of the inhabitants. Especially the measurable superimposing of longscale, transported dust from dry regions with the anthropogenic polluted urban dust makes investigations of Beijing's dust scientifically valuable. Those investigations contribute to a more comprehensive understanding of complex dust distribution and mixing processes in urban areas.

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Mapping of Trace Metals in Urban Soils **The Example of Mühlburg/Karlsruhe, Germany**

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Abstract. Spatial distribution maps depicting the concentrations of antimony, lead, tin, copper and zinc, and the presence of land-use units were generated for Mühlburg, a district of the City of Karlsruhe, Germany. The influence of the spatial land-use structure on the distributions of the element concentrations is statistically evaluated and discussed. The variography for Mühlburg shows an average range of 200-400 m for the spatial correlations of Sb, Pb, Sn and Zn. The variograms of Pb and Zn are characterised by hole effects at 300 m distances, i.e. the result of repeated stronger spatial correlations for certain distances between the sample sites. Most probably, this is an effect of the typical urban structure of streets, buildings, green spaces, and industry. Kriging method was used for the interpolation of Sb, Pb, Sn and Zn concentrations. Only Cu does not show a spatial correlation. In this case, the interpolation was carried out with a smoothed triangulation routine. Pollution plumes of point sources such as lead works, a bell foundry and a coal-fired thermal power station superimpose the more diffuse pollution from traffic, household heating processes, waste material disposal, etc. The trace element concentrations in soils of housing areas increase with the age of the developed area. Industrial areas show the highest level of pollution, followed by housing areas developed before 1920, traffic areas, allotments, housing areas developed between 1920 and 1980, parks and sports areas, cemetery and housing areas developed after 1980.

It is demonstrated that spatial distribution maps of element concentrations indicate potential emission sources of harmful substances, even if the emission itself or the direct surrounding soil have not been analysed. The analytical tools presented enable town planners to discern areas of higher soil pollution. Detailed investigations can be focussed on these areas to evaluate the possibilities of soil usage and transfer. These methods enable one to manage urban soil in an adequate manner. For these reasons, the methods demonstrated support an urban environmental impact assessment and are a part of a sustainable urban soil management.