Road Traffic Related Influences upon $PM_{2.5}$ Load in Beijing, China

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Introduction

Air quality is an ongoing problem in Beijing due to rapid economic growths and increasing road traffic. A lot of emission reduction measures were performed to improve the air quality in Beijing during the Olympic Summer Games in 2008, which has cut down PM_{10} largely (see Figure 1).

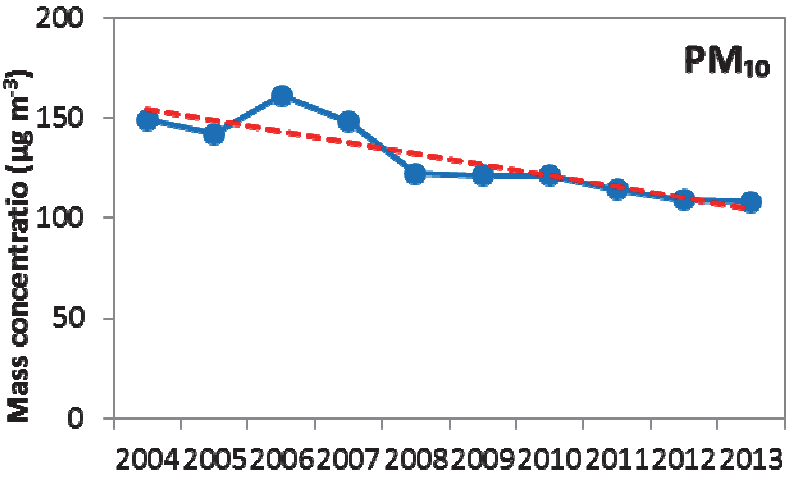

Figure 1: Annual variation of PM_{10} from 2004 till 2013 in Beijing (*Source: Beijing environmental state report 2004-2013*)

But high air pollution episodes have become much more frequent in Beijing during the last years (Wang et al., 2006; Zhang et al., 2014) and influence human health and visibility (Sun et al., 2006).

Since haze (reduced visibility to less than 10 km during wind speeds of less than 5 m s^{-1}) becomes increasingly conspicuous, some open questions are important. For instance: What is the origin of haze? How much is the contribution of anthropogenic emitted PM (especially by road traffic) to total PM exposure? To find answers to these questions the composition of $PM_{2.5}$ was discriminated and source attribution, particle characteristics and external impacts on the PM levels were investigated.

Two sequential High-Volume Samplers (HVS, Digitel DHA-80, Hegnau, Switzerland; 500 l min^{-1}) were used to collect $PM_{2.5}$ samples in Beijing automatically at CAS/IAP from 10 April till 08 June 2013:

- Quartz fibre filters (Ø 150 mm)
- Sampling time 24 hours (00:00 - 24:00); 4 hours during some haze episodes.

The meteorological data are from ZBAA (http://weather.uwyo.edu/upperair/sounding.html).

Chemical composition analyses

In order to discriminate the composition of $PM_{2.5}$ the inorganic elements (Na, Mg, Al, K, Ca, Fe, V, Cr, Mn, Co, Ni, Cu, As, Cd, Ba, Tl and Pb) and mass concentrations were analysed from sampler A by inductively coupled plasma mass spectrometry (ICP-MS) and filter weighing (gravimetric mass concentration determination), respectively. The inorganic water-soluble ions (Cl^-, SO_4^{2-}, NO_3^-, Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}), EC and OC as well as organic speciation of $PM_{2.5}$ were analysed from sampler B by ion chromatography (IC) and continuous flow analyzer, thermal/optical carbon analyser as well as gas chromatography - mass spectrometry (GC-MS), respectively.

Source apportionment

Positive matrix factorization (PMF3.0, EPA) was applied to determine factor profiles from measured composition of particles and analyses method uncertainties. Five different emission sources were determined using information about

- emission source markers and their relative importance,
- ratios between PM compounds and
- temporal variation of observed (input data) PM compound concentrations.

Also backward trajectory and cluster analysis with HYSPLIT4 were used to get information about emission source regions.

Further, enrichment factor analysis is applied to identify the sources of PM in Beijing.

Results

Three haze episodes were found: 18 - 25 April, 3 - 9 May and 1 - 8 June. The average $PM_{2.5}$ mass concentration was 89 µg m^{-3}, but during haze days 140 µg m^{-3} while only 45 µg m^{-3} during clear days.

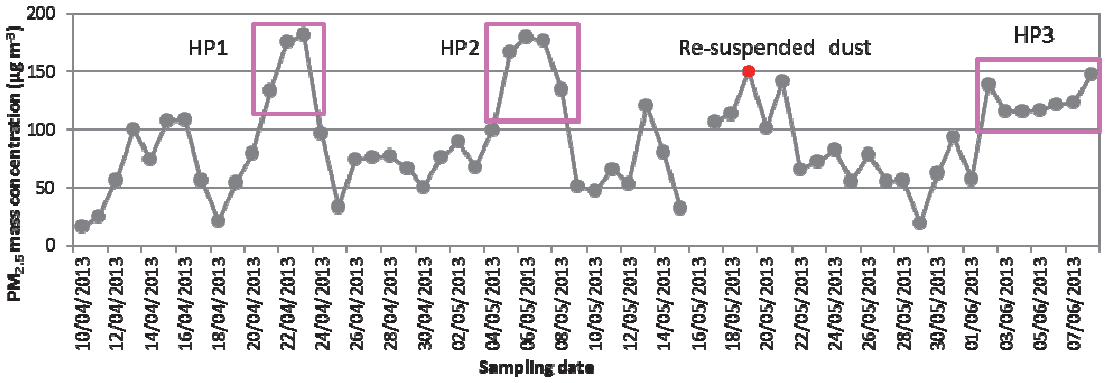

Figure 2: Temporal variation of $PM_{2.5}$ mass concentration from 10 April till 8 June 2013 in Beijing with indication of three haze episodes (HP1, HP2 and HP3) and a dust episode (re-suspended dust)

The $PM_{2.5}/PM_{10}$ ratios presented higher values during haze days (0.68) than during clear days (0.38) which indicate that fine particles and high transformation efficiency into secondary aerosol are the dominant factors for haze pollution.

PM characteristics

EC, OM (organic matter), Cl^-, SO_4^{2-}, NO_3^-, NH_4^+, K^+, Cu, Ni, Zn, As, Cd, Tl and Pb mass concentrations increased during haze days in comparison to clear days (see mass balance in Figure 3).

Especially secondary inorganic pollutant mass concentrations (NO_3^-, SO_4^{2-} and NH_4^+) were 6 times higher.

4 h sampling showed that average $PM_{2.5}$ mass concentration had the highest value during 16:00 – 20:00. NO_3^- mass percentage showed an obvious variation that had a high value during night (0:00 – 8:00) and a low value during day (12:00 – 20:00) while SO_4^{2-} mass percentage varied with $PM_{2.5}$ mass concentration basically and had the highest value between 12:00 and 20:00 (see Figure 4).

By comparing the inorganic element mass ratios of haze/clear and dust/clear days, Fe, Ca and Ba (crustal elements) were proposed to represent dust particles, and Zn, As and Pb were suggested to indicate haze particles.

The high amount of secondary inorganics indicates that major chemical species of $PM_{2.5}$ during haze episodes are originated from anthropogenic sources. NO_3^-, SO_4^{2-} and NH_4^+ are basically formed by gas-to-particle formation processes on the basis of chemical reactions of precursor gases. The mass ratio of NO_3^-/SO_4^{2-} during haze days (0.84) was higher than during 2001 - 2003 (0.80) (Wang et al., 2005) and 1999 - 2000 (0.58) (Yao et al., 2002) suggesting that vehicle exhaust emissions are a rational for haze days in Beijing.

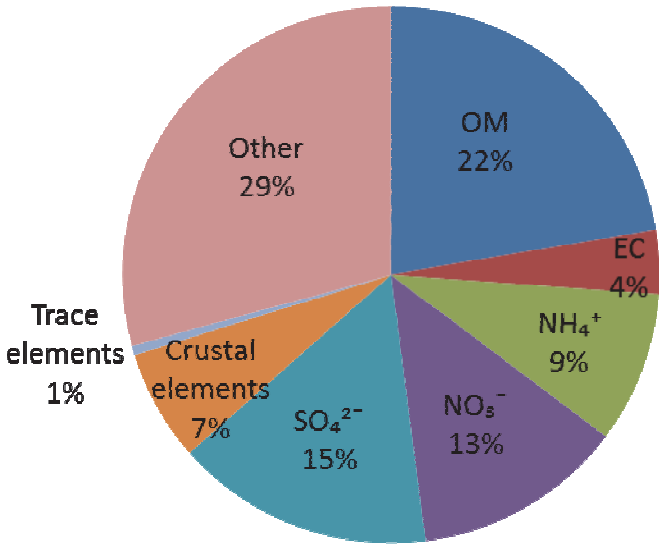

Figure 3: Mass balance of $PM_{2.5}$ compounds from 10 April till 8 June 2013 in Beijing

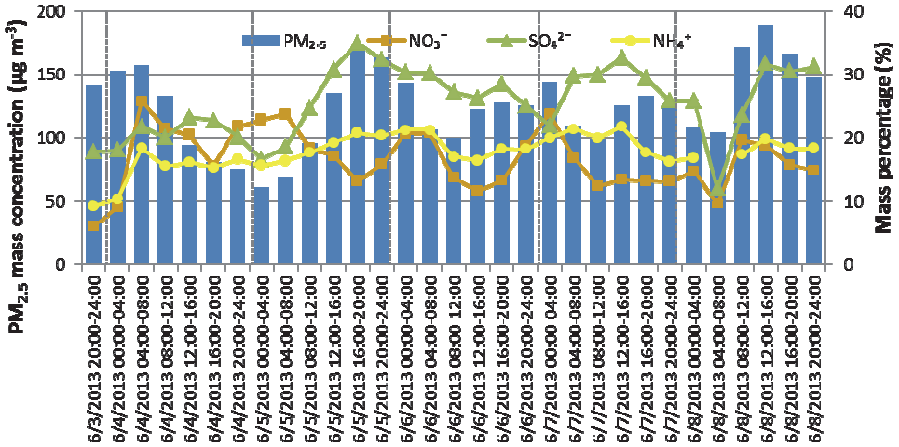

Figure 4: Secondary inorganic pollutant mass concentrations (NO_3^-, SO_4^{2-} and NH_4^+) of $PM_{2.5}$ during high-temporal sampling (4 hours) from 3 till 8 June 2013 in Beijing

PM emission sources

The result of PMF analyses are those (see Figure 5):

- The main sources of PM during this time period in Beijing are secondary inorganic ion formation, traffic, dust, fuel oil combustion and industry.
- The main difference between clear and haze days was, that dust and traffic emissions were found to be the main sources of PM during clear days while secondary inorganic ions formation was the dominant sources of PM during haze days.

Based on PAH diagnostic ratios, coal and liquid fossil fuel combustion were found to be the dominant sources of PAHs in PM in Beijing.

Some ratios of compounds as the Hopane and Homohopane index indicate vehicle exhaust emissions and coal combustion emissions and a high amount of these sources was detected.

It is found from enrichment factor analyses that those aerosols, which are originated from geogenic sources (found from Fe and Ba), are most likely re-suspended road dust and dust storm, and also from anthropogenic sources (found from Zn, As and Pb), such as vehicle exhaust, chemical industry, coal combustion and fertilizer application. Consequently, re-suspended dust, which is from previously deposited particles blown into the atmosphere by strong wind and vehicle driven turbulence, contributes to enhanced $PM_{2.5}$ load in Beijing (see Figure 2).

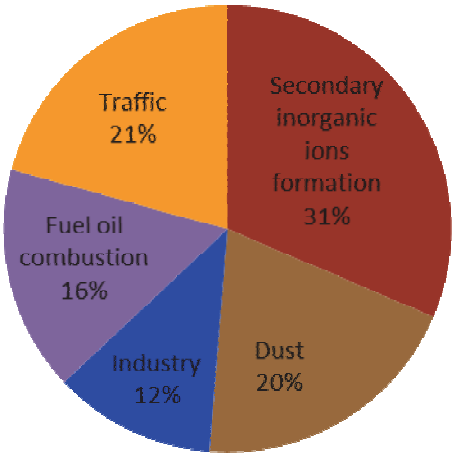

Figure 5: Result of source apportionment by PMF from 10 April till 8 June 2013 in Beijing

Finally, the backward trajectories from the South, calculated for 72 hours in 500 m altitude at the target point, were always found to be short and with high PM mass concentration (see Figure 6). Therefore, southerly flows were the main source of haze particles in Beijing. In this area industrial companies and big cities are in a distance of some 100 km.

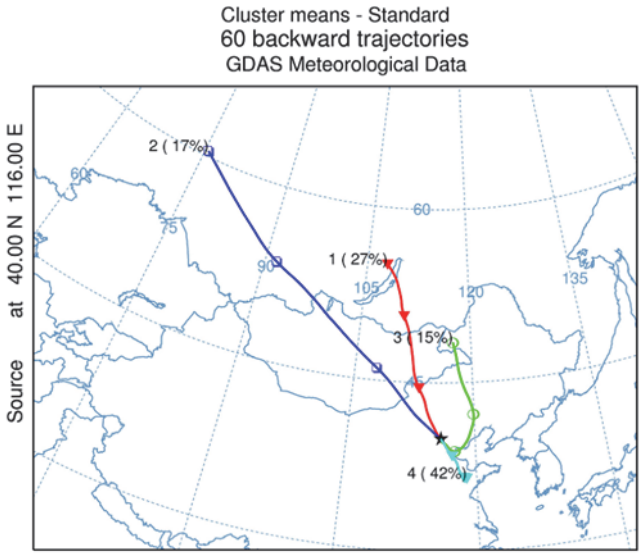

Figure 6: Cluster means of backward trajectories from 10 April till 8 June 2013 in Beijing

Conclusions and outlook

The $PM_{2.5}$ mass concentrations after the emission reduction measures during the Olympic Summer Games 2008 are still high.

Haze becomes more frequent because an increasing number of fine and anthropogenic particles is emitted.

The control of the emission of precursor gases of NO_3^-, SO_4^{2-} and NH_4^+ in the local and regional scale and thus of road traffic emissions is necessary for reducing haze.

Installation of cleaning equipment in industrial and mobile exhaust vents, improvement of road cleaning standards and reduction of construction dust also become important for improving air quality.

There is a need to do further research on formation mechanisms and sources of haze episodes during different seasons in order to improve the urban air quality.

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