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Research Article

Understanding Variability of Haze in Eastern China

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Abstract

In recent years, there are heavy aerosol pollutions in eastern China. For example, in the Northern China Plain (NCP) and the capital city of Beijing, the concentrations of $PM_{2.5}$ (particle matter <2.5 µm in diameter) often reached to the levels of more than 200-400 µg/m³. In contract, the World Health Organization (WHO) identifies a safe level of air quality as containing 10-25 µg/m³. In addition to the high levels of aerosol pollutions, there is a largely temporal variability of the concentrations. This study discusses the major factors, which control the short-term variability (days to weeks) of the aerosol pollutions in eastern China. The results suggest that the meteorological parameters, including wind direction, wind speed, and vertical diffusion in the planetary boundary layer (PBL) have important effects on the short-term variability. The secondary formation of aerosols, especially during heavy haze periods, has also important contribution to the large variability. Because the secondary aerosols (SA) have a large portion in the composition of the total aerosols during the haze period, the formation of SA amplifies the magnitude of the variation, causing significant increase in the concentrations of PM_{2.5} during the heavy haze periods in eastern China.

Keywords: Air pollution; Variability; Meteorological factors; Planetary boundary layer

Introduction

China is one of the most rapidly developing countries in the world, but in the meantime it is suffering from severe air pollution due to heavy industrial/metropolitan emissions. This extremely high loading of aerosols has strong effects on climate and environment, e.g., the effects on solar radiation (cooling surface and slowing down the global warming), cloud formation, ozone photochemical activity, and visibility [1-5]. The heavy haze pollution also causes severe human's health problems, such lung cancer, asthma and other respiratory illnesses [6,7].

Unlike other regions in industrial countries (such as Europe and the US), the haze pollution (aerosol pollution) in China widely displaced in a very large area. The haze pollution is not only covered over large cities, but also over farmland areas. As a result, the haze in China has not only important impact on human's health, but also on the ecosystem in eastern China. The effect of regional haze pollution on the yields of rice and wheat in China. Their result shows that reduction of solar irradiance due to aerosol pollution can cause ~2% reduction in total rice production and ~6% reduction in total wheat production in eastern China [8].

In addition to the large spatial dispersion of aerosol pollution, there is also significant temporal variation of aerosol pollution during a short period (days to weeks) in eastern China [9-13]. The focus of this study is to better understand the causes of this short-term temporal variability of aerosol pollution, which has very important implication in haze pollution control in the region.

Materials and Methods

In this study, a long-term measurement of PM2.5 concentrations (from 2012 to 2016) is used to investigate the variability of aerosol pollution, and a regional chemical/transport WRF-Chem model (Weather Research and Forecasting Chemical model) is used to analyze the regional transport and the short-term variability.

Measured data

The sampling site of PM2.5 (the particle matter with a diameter <2.5 μ m) concentrations is located in the southeastern part of downtown Xian, China (34°13'N, 108°52'E). The surrounding area represents a mixture of industrial, commercial, residential, and traffic environments. It is worth emphasizing that a battery manufacturer (Ni related battery) and an automobile plant (welding of metals) were located within less than 7 km from the sampling site. 24-hour integrated daily PM2.5 samples were collected. The particles were collected on pre-fired (780°C, 3 hours) 47 mm Whatman quartz microfibre filters (QM/A , Whatman Inc., U.K.) with the mini-volume air samplers (Airmetrics, Eugene, OR, USA) that operated at a flow rate of 5 L min⁻¹.

WRF-Chem model

The model used in this study is a regional chemical/transport model (WRF-Chem). There are two major parts of the model, namely, a dynamical model (WRF) and a chemical model (Chem). The Weather Research and Forecasting (WRF) Model is a mesoscale numerical weather prediction system. The WRF model is a fully compressible and non-hydrostatic Euler model. Thirty-five vertical levels are used in a stretched vertical grid with spacing ranging from 50 m near the surface, to 500 m at 2.5 km and 1 km above 14 km. The detailed information regarding the parameters used in the WRF model, such as the PBL scheme, the land surface scheme, the microphysics scheme, and the cumulus cloud scheme can be found at the WRF website.

In addition to dynamical calculations, a chemical model is fully

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coupled with the dynamical model (WRF-Chem). The version of the model used in the present study includes an online calculation of dynamical inputs (winds, temperature, boundary layer, clouds, etc.); transport (advection, convection, and diffusion); dry deposition; gas phase chemistry, radiation, and photolysis rates. The chemical mechanism includes 158 reactions among 36 species. The aerosol modules used in the study are described as the aerosol module developed by EPA CMAQ.

Results and Discussion

Measure aerosol variability

The measured daily mean surface concentrations of $PM_{2.5}$ in a large city in China (Xi'an, with a population of 8 millions) during a 4-year period (from 2013 to 2016) (Figure 1).

The measured result shows three important feathers. First, the concentrations of $PM_{2.5}$ were very high, with a mean concentration of ~170 µg/m³, indicating that heavy haze events often occurred in this region. Second, there was a strong long-term variability. The winterpeak values of the concentrations reduced from 600-800 µg/m³ in 2013 to ~400 µg/m³ in 2016. This long-term change was mainly associated with the emission control. Third, there were a strong mid-term variation and a short-term variability. For example, there was a strong seasonal variation (mid-term), with higher concentrations during winter than during summer. There was also a very strong daily variation (short-term). The short-term variability can lead to the change of $PM_{2.5}$ concentrations in a magnitude of 200-300 µg/m³ in days.

The major controlling factors for causing this short-term variability can be expressed by the aerosol mass conservation equation:

$$\partial [x] / \partial t = \partial [x] / \partial t]_{F} + \partial [x] / \partial t]_{T} + \partial [x] / \partial t]_{V} + \partial [x] / \partial t]_{C} + \partial [x] / \partial t]_{D}$$

Where, [X] represents the PM_{2.5} mass concentration (µg/m³). ∂ [X]/ ∂ t]_E represents the change of concentration due to surface emission; ∂ [X]/ ∂ t]_T due to advection or horizontal transport; ∂ [X]/ ∂ t]_V represents vertical diffusion in the PBL height; ∂ [X]/ ∂ t]_C represents chemical reactions; and ∂ [X]/ ∂ t]_D represents dry and wet deposition.

For short-term variability, the emission term can be ignored, if we don't consider an aggressive activity for emission (e.g., for the large emission reduction during the Beijing Olympic game period). If we consider a dry weather, without precipitation, the deposition term can also be ignored. With the above considerations, the equation governed short-term variability can be simplified by the following equation:

$$\partial [x] / \partial t = \partial [x] / \partial t]_T + \partial [x] / \partial t]_V + \partial [x] / \partial t]_C$$

This equation shows that major controlling factors for causing short-term of $PM_{2.5}$ variability are (1) advection (regional transport), (2) vertical diffusion, and (3) chemical reaction (secondary aerosol formation).

Effect of advection and weather condition

In some regions of eastern China, the short-term variability is strongly controlled by advection and weather condition. For example, in the NCP region and the city of Beijing, the weather condition and wind directions are important factors in controlling the short-term variability (Figure 2).

In the south of NCP and Beijing, there is a large polluted area, existing heavy emissions of air pollutants. In contrast, in the north of NCP and Beijing, there is a large remote area (mountains and grass



Figure 1: The measured daily mean surface concentrations of PM₂₅ in Xi'an, China from 2013 to 2016. The green-dash line shows a long-term trend, and red line shows the mean value.



lands), with small emissions of air pollutants. As a result, under the south wind condition, the aerosol concentrations are generally high (right panel of Figure 2). In contrast, under the north wind condition, the aerosol concentrations are generally low (middle panel of Figure 2). In addition to the effect of the spatial distribution of emissions, the mountains in this region also play important roles. The mountains in the north of NCP act as walls to block the horizontal transport. As a result, under south wind condition, the aerosol particles are accumulated at the foothill of the mountains (Figure 2) [14].

Effect of vertical diffusion in PBL

During the heavy haze period, the wind speed is small. The surface concentrations of $PM_{2.5}$ are highly correlated with vertical mixing and the PBL height. The *in situ* measurements show that during the haze days, the PBL heights and vertical diffusion are significantly reduced due to the decrease of solar radiation, which reduces the thermo turbulence [15].

During a short period (10 days), there is a strong short-term variability of $PM_{2.5}$ in Beijing. For example, on 24^{th} , Oct. 2013, the concentration of $PM_{2.5}$ was less than 50 µg/m³. In contrast, on 27^{7h} , Oct. 2013, the concentration of $PM_{2.5}$ rapidly increased to 160 µg/m³. This short-term variability is anti-correlated with both wind speed and the PBL height (Figure 3).

On 27^{7h} of Oct. 2013, the wind speed was less than 1 m/s, and the vertical diffusion in the PBL played important roles. The shallow PBL height (~0.5 km) strongly depressed the vertical transport from the surface to the free troposphere. As a result, the surface concentration of $PM_{2.5}$ reached to a very high value. The surface concentration of $PM_{2.5}$ was strongly anti-correlated with the PBL height (right panel of Figure 3). Moreover, the anti-correlation was non-linearly correlated. For example, when the PBL was less than 1 km, the surface concentration of $PM_{2.5}$ was very sensitive with the PBL height. In this case, a small reduction of PBL height can cause a large increase in the surface

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concentration of PM_{25} (Figure 3).

Effect of chemical reactions

According to the measurements of chemical composition, there are high secondary aerosols in eastern China. The secondary aerosols (sulfate, nitrate, ammonium, secondary organic carbon) contribute to a large portion of total aerosols. Sulfate, nitrate, and ammonium aerosols contribute to about 40% and 33% in Beijing and in Shanghai, respectively (Figure 4) [14].

The formation of secondary aerosols involves complicated chemical processes, including gas-phase chemical reactions and multi-phase chemical reactions (heterogeneous chemical reactions). However, during the heavy haze periods, neither the gas photochemistry nor the aqueous chemistry can fully explain the formation of high secondary aerosols. For example, the estimated sulfate formation is less than measured values under the heavy haze condition in eastern China. There is a large missing source for the formation of sulfate particles during heavy haze periods in eastern China (right panel of Figure 4), and there is additional heterogeneous reaction occurred during the heavy haze conditions; i.e.,

 $SO_2(g) + 2NO_2(g) + 2H_2O(aq) \rightarrow 2H^+(aq) + SO_4^{2-}(aq) + 2HONO(g)$

By adding this reaction, the underestimation of the calculated sulfate concentrations is largely reduced. However, there is still a large uncertainty remained for the underestimation of secondary aerosols during the heavy haze periods in eastern China. Because secondary



Figure 3: The short-term variability of PM_{25} , wind speed, and PBL heights from 21 to 31, October 2013 (left panels) measured in Beijing. The anti-correlation between PBL height and PM_{25} concentrations is shown in right panel.



sulfate production during heavy haze period in China.

aerosols have a large portion in the chemical composition of the total aerosols during haze periods, and the formation of SA amplifies the magnitude of the short-term variation of $PM_{2.5}$ concentrations, and future study is needed to better understand this problem [15,16].

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Conclusion

China is one of the most rapidly developing countries in the world, but in the meantime it is suffering from severe air pollution due to heavy industrial/metropolitan emissions. Unlike other regions in industrial countries (such as Europe and the US), the haze pollution in China is widely displaced in a very large area, and has significant short-term variation in the concentration of aerosol particles in eastern China. This study discusses the major factors, which control the shortterm variability (days to weeks) of the aerosol pollutions in eastern China. The results suggest that there are 3 major factors in controlling the short-term variability, including (1) horizontal transport (wind direction, wind speed, and weather system), (2) vertical diffusion in the planetary boundary layer), and (3) the secondary formation of aerosols, especially during the heavy haze period. The detailed effects of these 3 major factors on the short-term variability are discussed in this study. Because the secondary aerosols have a large portion in the chemical composition of the total aerosols during the haze period, the formation of secondary aerosols amplifies the magnitude of the variation, causing significant increase in the concentrations of PM25. However, the current calculations often underestimate this effect, especially under heavy haze conditions in eastern China. Further study is needed for better understanding the formation of secondary aerosols.

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