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Trends in atmospheric particles and their light extinction performance between 1980 and 2015 in Beijing, China

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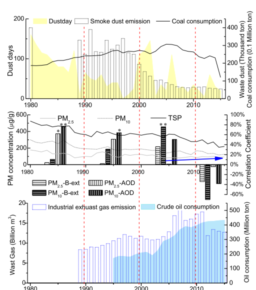
HIGHLIGHTS

- The decreased PM loading and increased $PM_{2.5}$ proportion were detected in Beijing for 1980-2015.
- PM_{10} might contribute to more horizontal and vertical light extinction than $PM_{2.5}$.
- The inverse U-shaped Kuznets Curve of PM_{10} exists, with the turning point occurring in 2000.

GRAPHICAL ABSTRACT



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ABSTRACT

This study explored the interdecadal variations and their horizontal and vertical light extinction performances of atmospheric particulate matter with aerodynamic diameters ≤2.5 μm ($PM_{2.5}$), particulate matter with aerodynamic diameters ≤ 10 μm (PM_{10}), and total suspended particulates (TSPs) in Beijing from 1980 to 2015, using data available from historical publications. Prominent declines of $PM_{2.5}$, PM_{10}, and TSPs were detected with long-term linear trends of −6.7, −4.3, and −1.9 μg m^{-3} yr^{-1}, respectively. Generally, on the annual scale during the studied period, it was found that $PM_{2.5}$ displayed negative correlation ($R^2 = 0.38$, $p < 0.01$) with visibility and positive correlation ($R^2 = 0.41$, $p < 0.01$) with aerosol optical depth (AOD). Comparably, PM_{10} exhibited robust negative correlation ($R^2 = 0.61$, $p < 0.01$) with visibility and positive correlation ($R^2 = 0.82$, $p < 0.01$) with AOD. The complicated interdecadal variations and light extinction performances of $PM_{2.5}$ were found, suggesting the changes on particle composition and vertical distribution of $PM_{2.5}$ in the atmosphere.

1. Introduction

Located within a region affected by East Asian dust, and as a developing megacity, Beijing experiences severe atmospheric particulate matter (PM) pollution derived from both natural and

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anthropogenic activities (Sun et al., 2004; Choobari et al., 2014). Documented regular outbreaks of atmospheric PM in Beijing during spring can be traced back hundreds of years (Zhang et al., 2006). Moreover, published data indicate that anthropogenic aerosol pollution has become exacerbated since 1975 (Beijing Western Suburb Environmental Investigation Team, 1977), following the implementation of several major economic reforms and adjustments made to the energy structure in Beijing. Quantification of the historical variation of atmospheric PM loading will provide valuable reference information for policy makers regarding public health, and illustrate the need for environmental protection measures in a megacity undergoing rapid economic and energy transformation.

One major obstacle to the investigation of historical PM variation in Beijing is the absence of a consistent long-term observational dataset. Although monitoring of atmospheric PM has been conducted by the Beijing Environmental Protection Bureau (BJEPB) since 1978, the monitoring targets have changed from as-deposited particles during 1979–2010, to total suspended particulates (TSPs) during 1991–2003, PM with aerodynamic diameters $\leq 10\ \mu m$ (PM_{10}) during 1997–2014, and PM with aerodynamic diameters $\leq 2.5\ \mu m$ ($PM_{2.5}$) from 2012 to the present. Thus, historical PM research in Beijing is dependent on reviewing disparate scattered observations, regressions from representative aerosol indices, or simulations by chemical transport models and climate models. Observations of $PM_{2.5}$ and PM_{10} loadings during 2004–2012, reported by Liu et al. (2015), have offered valuable insight regarding annual PM loadings on the decadal scale. Moreover, Lv et al. (2016) extended a time series of $PM_{2.5}$ observations to 12 years based on a review of the literature. However, to the best of our knowledge, similar research on multidecadal time scales has not been conducted.

Observations of visibility and aerosol optical depth (AOD) are used widely to estimate atmospheric PM concentrations in long-term research (Horvath, 1995; You et al., 2015; Founda et al., 2016). Routine observation of visibility has been conducted in Beijing since 1951 (Wang et al., 2015), whereas ground-based observation of AOD commenced in 2001 (You et al., 2015). These two indices were mainly used to invert historical $PM_{2.5}$ concentrations. For example, based on regression with visibility, Han et al. (2016) reported that urbanization was the major cause of $PM_{2.5}$ overloading in Beijing during 1973–2013. Based on AOD regression, Zeng et al. (2015) estimated that the average total mortality in the central Beijing area attributable to $PM_{2.5}$ was approximately 5100 individuals per year during 2001–2012. Unfortunately, the regression models used in the above studies were validated using daily or hourly $PM_{2.5}$ observational data acquired in recent years, which could have introduced considerable uncertainty with regard to estimations of historical PM concentrations. First, both PM_{10} and $PM_{2.5}$ are major causes of variation in visibility and AOD (You et al., 2015; Founda et al., 2016). The models were based on the assumption that $PM_{2.5}$ was the major component of PM_{10}, as found in recent conditions (You et al., 2015; Zeng et al., 2015; Han et al., 2016). However, the ratio of $PM_{2.5}$ to PM_{10} has historical volatility, e.g., it was 0.39 in 1988 (Ding et al., 1989) but 0.58 in 2011 (Gao et al., 2014); therefore, systematic examination of this relationship is required. Second, the models were based on the assumption that the correlations of $PM_{2.5}$ with both visibility and AOD have been constant over time, as has also been assumed in similar studies conducted in both Europe and North America (Hoff and Christopher, 2009). However, changes associated with the dramatic socioeconomic transition and the adjustment of the energy structure in China have resulted in changes of aerosol composition, e.g., the crustal mineral proportion of $PM_{2.5}$ has declined from 34.0% in 1989 (Chen et al., 1994) to 14.3% in 2014 (Li et al., 2017); therefore, the validity of this assumption must be ascertained.

Based on a comprehensive literature revision, the current study explored historical observations of $PM_{2.5}$, PM_{10}, and TSPs in Beijing for the period 1980–2015. The research had three primary objectives: (1) to quantify the historical variation and trend of PM pollution in Beijing, (2) to validate the relationship between PM concentrations and the optical index (as visibility and AOD), and (3) to understand how historical socioeconomic variations have shaped PM pollution and its associated light extinction behavior in Beijing.

2. Materials and methods

2.1. PM database

This study assumed that the PM observational data, collected by different instruments and at various sampling sites in Beijing, accurately represented the PM loadings at the sampling time. Overall, 113 sets of $PM_{2.5}$, 78 sets of PM_{10}, and 81 sets of TSPs data were collated from 117 studies (Table S1) on ground-based PM observations relevant to the studied period. To guarantee internal consistency of the datasets and to implement basic quality control, the following data selection principles were adopted: (1) sampling sites had to be located in urban areas of Beijing, far from specific points of pollution (e.g., coal-fired power stations); (2) the sampling period had to cover more than six months of any year. However, because of the lack of data during some periods (such as 1980–1990), some short-term observations were also included in this study.

Detailed sampling information (e.g., site location, instrument, and sampling frequency and period) of the PM observations is summarized in Table S1 and the sampling site locations are mapped in Fig. S1. Generally, most PM samples were collected in open areas, at heights of 1.5–30.0 m, on quartz fiber filters using high-, medium-, or low-volume air samplers with sampling fluxes in the range 1–1130 L min^{-1}.

2.2. Uncertainties associated with PM sampling grading method

In addition to instrument- and site-related uncertainties regarding the PM observations, another possible source of systematic error could be different size-grading methods adopted during sampling. For example, mass concentrations of PM_2 (i.e., particles with aerodynamic diameters $\leq 2\ \mu m$) and PM_7, PM_8, or PM_{11} (i.e., particles with aerodynamic diameters of ≤ 7, ≤ 8, or $\leq 11\ \mu m$) in certain historical studies (see Table S1) were respectively adopted as values of $PM_{2.5}$ and PM_{10} in this study, which would certainly cause errors during comparative analyses. According to Ding et al. (1989), the mass concentration differences between PM_7 and PM_{11} are within 12% for aerosols, whereas those between $PM_{2.1}$ and $PM_{3.3}$ are within 23%. In this study, the exact concentration differences between particles of different sizes were unclear and lacked calibration.

2.3. Construction and validation of PM datasets

Cross-year observational data was considered to represent the aerosol condition of the year with the longest observed time, and the mean value of some multiyear PM observational data was used as the annual PM concentration of each observed year. All the PM data were ranked chronologically and standardized for each year; data with standardized values beyond ± 3 were rejected as outliers. Finally, the mean value of all observed PM data in each year was calculated and defined as the annual PM concentration of that year, generating the PM time series (defined as PM-lit) used in this study.