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Estimating representative background $PM_{2.5}$ concentration in heavily polluted areas using baseline separation technique and chemical mass balance model

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A B S T R A C T

The determination of background concentration of $PM_{2.5}$ is important to understand the contribution of local emission sources to total $PM_{2.5}$ concentration. The purpose of this study was to exam the performance of baseline separation techniques to estimate $PM_{2.5}$ background concentration. Five separation methods, which included recursive digital filters (Lyne-Hollick, one-parameter algorithm, and Boughton two-parameter algorithm), sliding interval and smoothed minima, were applied to one-year $PM_{2.5}$ time-series data in two heavily polluted cities, Tianjin and Jinan. To obtain the proper filter parameters and recession constants for the separation techniques, we conducted regression analysis at a background site during the emission reduction period enforced by the Government for the 2014 Asia-Pacific Economic Cooperation (APEC) meeting in Beijing. Background concentrations in Tianjin and Jinan were then estimated by applying the determined filter parameters and recession constants. The chemical mass balance (CMB) model was also applied to ascertain the effectiveness of the new approach. Our results showed that the contribution of background PM concentration to ambient pollution was at a comparable level to the contribution obtained from the previous study. The best performance was achieved using the Boughton two-parameter algorithm. The background concentrations were estimated at (27 ± 2) μg/m^3 for the whole year, (34 ± 4) μg/m^3 for the heating period (winter), (21 ± 2) μg/m^3 for the non-heating period (summer), and (25 ± 2) μg/m^3 for the sandstorm period in Tianjin. The corresponding values in Jinan were (30 ± 3) μg/m^3, (40 ± 4) μg/m^3, (24 ± 5) μg/m^3, and (26 ± 2) μg/m^3, respectively. The study revealed that these baseline separation techniques are valid for estimating levels of $PM_{2.5}$ air pollution, and that our proposed method has great potential for estimating the background level of other air pollutants.

1. Introduction

China's rapid industrialization and urbanization has caused particulate matter (PM) pollution to become the major environmental problem in large cities. Exposure to PM causes adverse health effects and it also limits visibility (Schichtel et al., 2001; Yadav et al., 2003). To develop effective pollution control policies and measures, we must improve our understanding of PM background concentration.

Background concentration has been defined as the concentration “that is not affected by local sources of pollution” (WHO, 1980; Menichini et al., 2007). Lenschow et al. (2001) assumed that PM concentration measured at a selected site corresponded to the contributions from regional and urban background emissions, as well as from the local nature sources. McKendry (2006) referred to background air pollution as concentrations arising from local natural emission sources in addition to natural and anthropogenic emissions transported into the selected airshed. Accurate background concentrations data represent a critical input for improving the urban airshed modeling results. While different definitions of PM background concentrations were proposed depending on the specific research needs, in the current paper, we define PM background concentration as the concentration that is indirectly affected by local emission sources that arise from

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human activities (McKendry, 2006; Menichini et al., 2007; Stein et al., 2007). Evaluating reliable estimate of background concentrations is crucial for the development of effective air quality management plans to control pollution.

The background concentration is not a fixed value. It varies on both a spatial and temporal scale, because it is influenced by the emissions of pollutants transported from other cities. Meteorological parameters, such as wind direction and speed, also affect the re-distribution of background concentration (Beelen et al., 2009; McNabola et al., 2011). A primary method for estimating background concentration is field observation at background sites, which are selected based on seven principles: five representative elements (representative of the region, atmosphere, scale, background, and ecology) and two stability elements (stability of the environment and region). The collected samples from background sites are assumed to be non-contaminated and thus can be used to directly represent the background concentration level. Many published studies have been carried out at background sites (Wang et al., 2004; Yao et al., 2012; Zhang et al., 2014). However, the data from the collected samples may not accurately reflect background signals, because the background sites may have already been polluted by local emission sources due to the rapid industrial development of cities, especially in heavily polluted areas. Thus, these so-called non-contaminated samples cannot be used to characterize the background concentration (Tchepel et al., 2010; Giostra et al., 2011). Another widely used method to determine the background concentration is the use of air-quality numerical simulation models, such as the Community Multiscale Air Quality Modeling System (CMAQ), Comprehensive Air Quality Model with Extensions (CAMx), Fifth-Generation Penn State/NCAR Mesoscale Model (MM5), Weather Research and Forecasting (WRF) model. However, this method is an exhaustive demanding method that requires detailed knowledge about the meteorology, land use, emission inventory, chemistry and specific computer capability.

Statistical methods, such as discriminant analysis, cluster analysis, baseline separation technique, robust local regression, have also been applied for background level estimation (Tchepel et al., 2010; Giostra et al., 2011; Gómez-Losada et al., 2015, 2016; McNabola et al., 2011; Langford et al., 2012; Pu et al., 2014). Such approaches use different statistical analyses to decompose the background component from the monitoring data (Giostra et al., 2011; Gómez-Losada et al., 2016). The statistical method of baseline separation technique has been used extensively in the field of hydrology to extract a baseflow signal from a time-series record of a steam flow. However, its application in atmospheric environment studies has been limited. The baseflow separation process is similar to the process of decomposing the background level from an air pollutant concentration dataset (McNabola et al., 2011). This method comes from the field of electrical engineering, where it is used to analyze and process signals. This signal-processing technique can be used to filter out high-frequency signals to obtain the low-frequency signals in an electronic circuit (Nejadhashemi et al., 2009; Li et al., 2013). Hydrologists assume that total steam flow comprises two components: delayed shallow surface flow, and baseflow from underground water (Smakhtin, 2001; Cherkauer and Ansari, 2005; Santhi et al., 2008; Aksoy et al., 2009). Surface flow is seen as a "quick-response" (high-frequency) signal, because of its quick response (i.e. produces spikes quickly) to rainfall, while baseflow is described as a "slow-response" (low-frequency) signal due to its relative stability in response to the instantaneous increase of rainfall. Similarly, the background concentration of an air pollutant is also assumed to be stable, and not subject to quick changes in the monitored concentration; it is therefore reasonable to treat the background concentration as a "slow-response" signal during the separation procedure. However, to the best of our knowledge, with the exception of one study (McNabola et al., 2011), the baseflow separation method has not been applied in the atmospheric field. In that study, different types of baseflow separation methods were applied to personal exposure time-series data to extract the background PM exposure level. The study concluded that it is valid and possible to separate the background component from air-pollution data using baseflow separation methods to improve the prediction of personal exposure to air pollutants. The study evaluated the background personal exposure on a selected day, and did not discuss long-term background concentration. In the present study, we tested the ability of the baseflow separation method to predict background $PM_{2.5}$ concentration by applying it to air-monitoring data collected at 12 observation sites in two heavily polluted Chinese cities in 2014. We used the chemical mass balance (CMB) model developed by the United States Environmental Protection Agency to ascertain the effectiveness of the baseline separation approach. The CMB model is one of several receptor models that use the chemical and physical characteristics of particles measured in ambient air (the receptor) and sources to quantify source contribution to ambient PM. We surmised that these results could indicate the contribution from local natural source, which could help us to indirectly determine the background level.

The objectives of this study were to (1) estimate the representative background concentrations of $PM_{2.5}$ in two heavily polluted Chinese cities using different baseline separation techniques borrowed from the hydrological field; and (2) ascertain the effectiveness of the proposed statistical method using the results from the CMB model. Our investigation is the first attempt to extract background component using a hydrological baseline separation method in polluted areas.

2. Materials and methods

2.1. Observation sites and air-quality data

Tianjin (39.1°N; 117.1°E) and Jinan (36.4°N; 117.0°E) are two significantly polluted cities located in the north-eastern region of China (Fig. 1). The annual average $PM_{2.5}$ concentration in 2014 was 83 μg/m^3 in Tianjin and 91 μg/m^3 in Jinan (TEPB, 2014; JEPB, 2014). These values are 2.4 and 2.6 times higher, respectively, than the Chinese air-quality standard of 35 μg/m^3. Tianjin is located in the northeast region of the North China Plain with the Bohai sea to the east and the Yanshan mountains to the north, covering a total area of approximately 11,946 km^2 with a population of about 15 million. Jinan borders Mount Tai to the south, and has a population of more than 7 million in an area of 8177 km^2. The annual average temperature and precipitation are 11–12 °C and 571 mm, respectively, in Tianjin, and 13–14 °C and 654 mm, respectively, in Jinan.

As shown in Fig. 1, 12 observation sites (Locations #1–6 in Tianjin and Locations #7–12 in Jinan) were selected. The $PM_{2.5}$ data (hourly average concentration) at these sites in 2014 were obtained from the air-pollution monitoring network operated by the China National Environmental Monitoring Centre (CNEMC). The daily mean concentrations at each site were calculated when 83% of the data were available (20 out of 24 h). The air-quality data are validated by the CNEMC before data analysis. One urban background site at Tianjin, located at Tuanbo swamp (Location #5), was selected as an example to demonstrate the baseline separation process. This site is upwind of the city, and no direct PM emission sources are located nearby. In addition, there are no obstructions or high buildings around this site, which guarantees the smooth interchange of air.

2.2. Baseline separation techniques

Five different separation methods (Lyne-Hollick, one-parameter algorithm, Boughton two-parameter algorithm, sliding interval, and smoothed minima) were used to estimate $PM_{2.5}$ background level. The separation was carried out using HydroOffice 2012 (Version 3) statistical program package and its module BFI+3.0 (Gregor, 2012). The module is designed to obtain baseflow separation curves from original time-series data by applying different separation filters.

(1) Recursive digital filters (RDFs)