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A time series analysis of multiple ambient pollutants to investigate the underlying air pollution dynamics and interactions

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HIGHLIGHTS

- DFA investigated the associations among aerosols and meteorological factors.
- The common trends of the yearlong and diurnal variations were identified.
- Common trends can reveal the secondary pollution including NOx processes.
- Interactions between aerosols were identified in different scenarios.

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ABSTRACT

Understanding the temporal dynamics and interactions of particulate matter (PM) concentration and composition is important for air quality control. This paper applied a dynamic factor analysis method (DFA) to reveal the underlying mechanisms of nonstationary variations in twelve ambient concentrations of aerosols and gaseous pollutants, and the associations with meteorological factors. This approach can consider the uncertainties and temporal dependences of time series data. The common trends of the yearlong and three selected diurnal variations were obtained to characterize the dominant processes occurring in general and specific scenarios in Taipei during 2009 (i.e., during Asian dust storm (ADS) events, rainfall, and under normal conditions). The results revealed the two distinct yearlong NOx transformation processes, and demonstrated that traffic emissions and photochemical reactions both critically influence diurnal variation, depending upon meteorological conditions. During an ADS event, transboundary transport and distinct weather conditions both influenced the temporal pattern of identified common trends. This study shows the DFA method can effectively extract meaningful latent processes of time series data and provide insights of the dominant associations and interactions in the complex air pollution processes.

1. Introduction

Exposure to particulate matter (PM), especially fine PM (i.e., PM 2.5, particulate matter particles with aerodynamic diameter $\leqslant 2.5\ \mu m$) can adversely affect human health (Dockery et al., 1993; Pope, 2000a,b; Pope et al., 2004). Recent studies have demonstrated that PM compositions are more closely associated with human health effects than size distributions are (Chuang et al., 2007; Bell et al., 2009; Patel et al., 2009). For example, sulfate and organic carbon (OC) in PM 2.5 were determined to be more closely associated with the reduction of heart rate variability than PM 2.5 itself is. PM composition can vary across space and time because of its relationships with other pollutants and meteorological factors, and can therefore result in the spatiotemporal variation of health effects attributed to PM 2.5 exposure (Bell et al., 2009). In general, PM concentrations consist of primary pollutants directly emitted from certain sources, and secondary pollutants, which are formed by chemical processes. Secondary pollutants account for approximately 35% of PM 10 mass in Taipei, because of strong photochemical activities (Chang and Lee, 2007a). In the United States, secondary aerosols also comprise a major portion of PM 2.5, and primarily include sulfates, nitrates, ammonium, and secondary organic aerosols (Fine et al., 2008). The formation of secondary aerosols is closely associated with *and* secondary processes involving both aerosol and gaseous pollutants (Seinfeld and Pandis, 2006; Chang and Lee, 2007a; Jimenez et al., 2009). In addition to the knowledge of the composition pattern of aerosols, for the purposes of air quality control, it is important to understand the underlying major dynamics of aerosols and their temporal patterns.

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The variations and interactions among aerosols and other air pollutants are constituted by a variety of complicated natural and anthropogenic mechanisms. Consequently, the multiple scales of temporal variation can appear in the time series of pollutant observations (Yu and Wang, 2013). Small time-scale variations and interactions can be closely associated with local emission and chemical reaction processes; therefore, this association can serve as a critical reference for local governmental agencies involved in air quality control. To understand the underlying processes that dominate changes in aerosol concentrations has been a major endeavor in air quality modeling for revealing major emission sources in particular areas under study (Cheng and Hopke, 1989; Chow and Watson, 2002; Srivastava and Jain, 2007). Receptor modeling by factor analysis, principle component analysis, and its variants (e.g., positive matrix factorization) has been widely used to identify the profiles of the common emission sources, thereby enabling the contributions of the identified sources in each sample to be determined (Hopke et al., 1976; Paatero and Tapper, 1994; Huang et al., 1999; Song et al., 2001; Hopke, 2003; Kim et al., 2004; Hammond et al., 2008). However, most of these methods are used to analyze the covariances among observed air quality levels, and neglect to consider the temporal correlations among the observations (Yu et al., 2013).

Dynamic factor analysis (DFA) is a dimension-reduction technique designed for multivariate time series data. Based on a structural time series model, DFA considers the uncertainty of multivariate observations and their latent smoothing functions (Zuur et al., 2003a). DFA can be used to determine common trends and explanatory variables in nonstationary, multivariate time series. Compared to other factor analysis methods, such as principle component analysis (PCA) or positive matrix factorization (PMF) analysis, DFA is designed to investigate multivariate time series data by explicitly considering the temporal autocorrelation among the factor scores as will be shown in the following section. The distinct feature allows DFA method to consider not only the associations across the variables of concern, but also the associations over time. DFA was originally developed for use in economics studies, but has recently been widely applied in environmental studies to analyze groundwater level variations (Ritter and Muñoz-Carpena, 2006), groundwater quality trends (Muñoz-Carpena et al., 2005; Kuo and Chang, 2010), topsoil water dynamics (Ritter et al., 2009), and ecological species (Zuur and Pierce, 2004; Kuo and Lin, 2010). Recent studies applied the DFA approach to investigate common temporal patterns of daily or weekly PM concentrations across monitoring stations (Yu and Lin, 2011; Kuo et al., 2011b; Yu et al., 2013), however, without analyzing the underlying interactions among aerosol and gaseous pollutants.

For the purposes to understand the air pollution dynamics, this study investigated the major long-term and diurnal latent contributors of ambient air pollutants, including aerosols and gaseous pollutants, for the entire 2009 and on days with certain PM 10 levels, respectively. The DFA method was used to identify the common trends of aerosols and their significant exogenous contributors (i.e., meteorological conditions). The analyzed dataset consisted of hourly measurements of ambient pollutants and meteorological factors in Taipei in 2009, including measurements of aerosols (e.g., sulfate, nitrates, and PM 2.5) and criteria pollutants (e.g., ozone, NO_2, and SO_2), as well as meteorological observations, such as temperature and relative humidity (RH).

2. Materials and method

2.1. Study area

The Hsin-Chuang supersite is located at the center of the Taipei metropolitan area, as shown in Fig. 1. The Taipei metropolitan area has a population of approximately seven million, and is situated in the second-largest basin in Taiwan (Chang and Lee, 2007c). The basin topography generally reduces convective circulation, and therefore elevates the concentration of ambient pollutants in the city. As shown in Fig. 1, the Hsin-Chuang supersite is located in a mixed residential and commercial area; however, it is surrounded by major industrial areas, including the WuKu industrial park and a petrochemical plant within 0.5–3 km. Chang and Lee (2008) identified three main contributors to air pollution in the Taipei area: traffic emissions, photochemical pollution, and transboundary pollution, in the analysis of air quality data from 1994–2003. Among these, transboundary pollution resulting from Asian dust storm (ADS) events was observed to substantially raise PM levels and change the distributional patterns of aerosols (e.g., elemental carbon (EC), sulfates, and nitrates) (Cheng et al., 2008; Chang et al., 2010b). Temporal variation in air pollution that occurs in Taipei is also closely associated with meteorological conditions; stagnant atmospheric circulation, for example, significantly increases the concentration of aerosols (Chuang et al., 2008; Chang et al., 2010a).

2.2. Data

Aerosol data has been collected hourly at the Hsin-Chuang supersite since March 2002, including sulfate, nitrate, EC, OC, polycyclic aromatic hydrocarbon (PAH), PM 10, and PM 2.5 measurements. In addition to aerosol observations, the Taiwan Environmental Protection Agency (TWEPA) has also regularly monitored the concentrations of criteria pollutants, including ozone, NO, NO_2, CO, SO_2, PM 2.5, and PM 10, and meteorological variables, including wind speed (WS), ambient pressure, temperature, and RH by using its island-wide monitoring network. The details of analytical methods and instruments for ambient air pollutant collections of the analysis are listed in Table S1. The TWEPA Hsin-Chuang station is located approximately 200 m from the Hsin-Chuang aerosol supersite. In this study, we analyzed data obtained hourly at both the supersite and the TWEPA station at Hsin-Chuang, including data related to ambient pollutants and meteorological observations. Because of the high similarity (i.e. correlation coefficients of 0.91 and 0.92 for PM10 and PM2.5, respectively) between the PM measures at the two stations, the supersite PM 10 and PM 2.5 records were used to represent the PM levels at Hsin-Chuang. In this study, hourly air quality and meteorological datasets for all of 2009 were investigated to reveal the major common underlying processes involved in year-long temporal variations in aerosols. The summary statistics of the hourly air pollutants and the meteorological data for the entire study period are listed in Table S2 (in Supplementary material), and the corresponding Pearson correlation coefficients among the datasets are listed in Table S3 (in Supplementary material). Because the diurnal pollution dynamics can highly depend upon various environmental conditions, this study uses PM10 as an indicator for the general condition of ambient environment. In this study, the air qualities with three scenarios are considered, i.e., PM10 at normal, extreme high, and extreme low levels. We obtained hourly data from 3 selected days with the median ($35.83\ \mu g\ m^{-3}$), maximal ($226.2\ \mu g\ m^{-3}$), and minimal ($7.40\ \mu g\ m^{-3}$) daily PM 10 levels in 2009, were further analyzed. The selected days corresponded to those in which normal conditions, an ADS event, and a rainfall event occurred: August 17 (Saturday), April 25 (Monday), and September 29 (Tuesday), respectively. The temporal variations in PM 10 levels for all of 2009, and for the three selected days, are shown in Fig. 2.

2.3. Dynamic factor analysis method

DFA has been used to reveal the underlying dominant contributors to changes in time series (Molenaar et al., 1992; Zuur et al., 2003a). This structural time series analysis method characterizes

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