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Identifying the impacts of climate on the regional transport of haze pollution and inter-cities correspondence within the Yangtze River Delta☆

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ABSTRACT

Regional haze pollution has become an important environmental issue in the Yangtze River Delta (YRD) region. Regional transport and inter-influence of $PM_{2.5}$ among cities occurs frequently as a result of the subtropical monsoon climate. Backward trajectory statistics indicated that a north wind prevailed from October to March, while a southeast wind predominated from May to September. The temporal relationships of carbon and nitrogen isotopes among cities were dependent on the prevailing wind direction. Regional $PM_{2.5}$ pollution was confirmed in the YRD region by means of significant correlations and similar cyclical characteristics of $PM_{2.5}$ among Lin'an, Ningbo, Nanjing and Shanghai. Granger causality tests of the time series of $PM_{2.5}$ values indicate that the regional transport of haze pollutants is governed by prevailing wind direction, as the $PM_{2.5}$ concentrations from upwind area cities generally influence that of the downwind cities. Furthermore, stronger correlation coefficients were identified according to monsoon pathways. To clarify the impacts of the monsoon climate, a vector autoregressive (VAR) model was introduced. Variance decomposition in the VAR model also indicated that the upwind area cities contributed significantly to $PM_{2.5}$ in the downwind area cities. Finally, we attempted to predict daily $PM_{2.5}$ concentrations in each city based on the VAR model using data from all cities and obtained fairly reasonable predictions. These indicate that statistical methods of the Granger causality test and VAR model have the potential to evaluate inter-influence and the relative contribution of $PM_{2.5}$ among cities, and to predict $PM_{2.5}$ concentrations as well.

1. Introduction

The enormous energy consumption associated with rapid economic development and urbanization in China has generated tremendous emissions of air pollutants. Most well-developed areas in China have suffered poor air quality in recent decades (Li et al., 2011; Lin et al., 2013; Wang et al., 2012; Zhu et al., 2010), especially with regards to severe haze pollution (An et al., 2015; Kang et al., 2013; Tang et al., 2016; Wang et al., 2015b), while the Yangtze River Delta (YRD), the Beijing-Tianjin-Hebei (BTH) region, the Pearl River Delta (PRD) region and the Sichuan Basin have been identified as 4 major haze areas experiencing significant declines in visibility (Zhang et al., 2012).

Due to the fast mobility of air mass, understanding the anthropogenic pollutants contribution from local and regional sources has become an essential research topic for designing effective control strategies for improving air quality. Several types of methods have been developed to estimate the contributions of different source regions to air pollutants, such as chemical transport model-based methods, back-trajectory based methods and receptor methods. Chemical transport model-based methods require detailed emission inventories, as well as topographic and meteorological conditions data. As a numerical simulation

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development, chemical transport models with emission inventories, such as CMAQ and NAQPMS, have become powerful tools for simulating pollutant dispersion in the atmosphere and evaluating the local/regional source contribution to pollutant concentrations (Lang et al., 2013; Streets et al., 2007; Wang et al., 2006; Wu et al., 2011). Back-trajectory based methods are based on backward air mass trajectories calculated by running HYSPLIT models combined with pollutant concentrations, such as backward trajectory cluster analysis, trajectory sector analysis (TSA) and the potential source contribution function (PSCF), which can be applied to the study of transport and regional sources of air pollutants (Li et al., 2012; Wang et al., 2015a). These methods are able to clearly demonstrate the geographic distributions of potential source regions, but they are not able to directly link the higher possibility of pollution at receptor regions to the high emissions from the potential sources. However, Jia et al. (2008) quantified the regional contribution of $PM_{2.5}$ solely from its "sawtooth cycles", while Sun et al. (2014) estimated the regional contribution of PM_1, NO_2 and CO based on the "steep evolution" of air pollutant concentrations. These techniques belong to receptor-oriented statistical methods, which do not need to track the transport process of air pollutants and do not rely on emission source conditions, topography and meteorological conditions. Consequently, receptor based statistical methods have become a popular method for quantifying the source sectors of suspended particles with chemical components (Song et al., 2006; Thurston et al., 2011), even though they are not able to identify the source locations of air pollutants.

Granger causality test is a popular method for identifying possible causal relationships between variables of a time series (Eichler, 2012), and can also be used to investigate causal relationships between different air pollutants (Hui, 2004; Yu and Liu, 2015). Vector autoregressive (VAR) models have been applied to air pollution research, such as for assessing the inter-dependence among air pollutants (Hsu, 1992; Hui, 2004; Kumar et al., 2009; Yu and Liu, 2015), correlations among air pollutants, meteorological data (wind speeds, temperatures, soil temperatures, dew points) and traffic data (Wang and Niu, 2009), and forecasting monthly temperature, humidity and cloud coverage (Shahin et al., 2014). Hence, to our knowledge, these methods have never been utilized to evaluate the inter-cities transport of particular pollutants or their relative source contribution in each city.

To characterize the properties of regional $PM_{2.5}$ pollutants, 3 sampling sites were established across 3 major cities within the YRD, i.e. Shanghai, Nanjing and Ningbo. The isotope ratios of the particle samples from these sites were analyzed and compared. For statistical analysis, the online $PM_{2.5}$ measurements from these sites were collected. For comparison purposes, similar data from Lin'an, a rural metrological monitoring station close to Hangzhou, was also collected to serve as a reference for the regional background. Granger causality tests of the time series data of the $PM_{2.5}$ and VAR models were introduced to investigate the inter-influence and relative source contributions of particle pollutants among cities.

2. Data and methods

2.1. Sample collection and isotope analysis

The daily $PM_{2.5}$ samples from the 3 major cities for 4 different seasons were collected simultaneously at the University of Nottingham Ningbo (Ningbo), Gulou Campus of Nanjing University (Nanjing) and Shanghai Academy of Environmental Sciences (Shanghai). For each site, about 20 samples for each season and a total of about 80 samples were collected from July 21, 2014 to April 28, 2015. The stable carbon and nitrogen isotopic composition of the samples were analyzed using a Thermo Scientific Delta V Advantage isotope ratio mass spectrometer with a Flash 2000 Organic Elemental Analyzer. Isotopic abundances were expressed relative to the international standard as follows:

$$\delta^{13}C(‰) = \left[(^{13}C/^{12}C)_{sample} / (^{13}C/^{12}C)_{standard} - 1 \right] \times 1000 \quad (1)$$

$$\delta^{15}N(‰) = \left[(^{15}N/^{14}N)_{sample} \Big/ (^{15}N/^{14}N)_{standard} - 1 \right] \times 1000 \quad (2)$$

The isotope ratios of particle in Ningbo, Nanjing and Shanghai were referred to as N_b, N_j and S_h, respectively.

2.2. $PM_{2.5}$ data

Fig. 1A presents the map of the YRD region with the relative locations of the observed sites of the 4 cities (Lin'an, Ningbo, Shanghai and Nanjing). The hourly average $PM_{2.5}$ concentrations from these cities were collected from the National Environment Protection website. $PM_{2.5}$ data were available between October 1, 2013 and September 30, 2015, except for Lin'an for which data were available from January 1, 2014. Daily and monthly average $PM_{2.5}$ concentrations were calculated based on hourly average data. For simplicity, the $PM_{2.5}$ data in Lin'an, Ningbo, Nanjing and Shanghai were referred to as LA, NB, NJ and SH, respectively.

2.3. Identification of prevailing winds

Since the regional transport and inter-influence of $PM_{2.5}$ is expected to change across monsoons, and the time series analysis is based on monsoons, backward trajectory statistics were performed to identify the seasonality of the regional prevailing winds. The 48-h backward air trajectories arriving at the City Government of Lin'an (30.240°N, 119.731°E), the University of Nottingham Ningbo (29.807°N, 121.569°E), Gulou Campus Of Nanjing University (32.060°N, 118.787°E) and Shanghai Academy of Environmental Sciences (31.176°N, 121,437°E) were calculated using an Hysplit-4 model from the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (http://ready.arl.noaa.gov/HYSPLIT.php), while meteorological data were obtained from the NCEP/NCAR reanalysis meteorological database. The model was run 4 times per day at UTC times of 0:00, 06:00, 12:00 and 18:00 (local times 08:00, 14:00, 20:00 and 02:00 of the next day, respectively), between 2012/10/01 and 2015/09/30, including 4380 backward trajectories at each observation site. The starting height was set at 500 m above ground level, which could diminish the effects of ground surface friction and represent winds in the lower boundary layer (Ara Begum et al., 2005). The trajectory clustering and probabilities of clusters were performed by the geographic information system (GIS) based software TrajStat (Wang et al., 2009). For each site, the cluster results and the proportion of clusters for each month were shown in Fig. S1 and Table S1, respectively.

2.4. Data analysis

To identify the impact of the prevailing wind on $PM_{2.5}$ transport and inter-influence among cities, and describe the $PM_{2.5}$ source contribution from each city, several sets of time series analyses were carried out. A Pearson correlation analysis was performed using SPSS software, while the Granger causality tests, cross correlation function analyses and VAR models were performed using R 3.2.3 (R Development Core Team).

The Granger causality test was first proposed in 1969 (Granger,