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Spatial-temporal variation characteristics of air pollution in Henan of China: Localized emission inventory, WRF/Chem simulations and potential source contribution analysis

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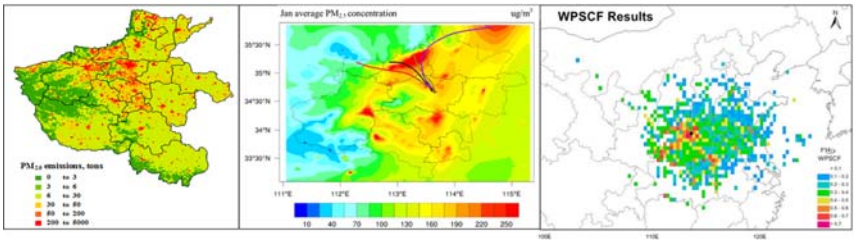
HIGHLIGHTS

- We develop an integrated emission inventory of multi-pollutants in Henan of China.
- We explore spatial-temporal variations of air pollution in Henan with WRF/Chem and back trajectories.
- We analyze causes of heavier pollution region and period with integrated methods.
- Heavier pollution in central Henan is mainly due to local intensive emissions.
- Emissions from surrounding provinces are also potential source of $PM_{2.5}$ in Henan.

GRAPHICAL ABSTRACT



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ABSTRACT

Henan is the most populous province and one of the most seriously polluted areas in China at present. In this study, we establish an integrated atmospheric emission inventory of primary air pollutants in Henan province for the target year of 2012. The inventory developed here accounts for detailed activity levels of 11 categories of primary anthropogenic emission sources, and determines the best available representation of emission factors. Further, we allocate the annual emissions into a high spatial resolution of 3 km × 3 km with ArcGIS methodology and surrogate indices, such as regional population distribution and gross domestic product (GDP). Our results show that the emissions of VOCs, SO_2, PM_{10}, $PM_{2.5}$, NO_X, NH_3, CO, BC and OC are about 1.15, 1.24, 1.29, 0.70, 1.93, 1.05, 7.92, 0.27 and 0.25 million tons, respectively. The majority of these pollutant emissions comes from the Central Plain Urban Agglomeration (CPUA) region, particularly Zhengzhou and Pingdingshan. By combining with the emission inventory with the WRF/Chem modeling and backward trajectory analysis, we investigate the temporal and spatial variability of air pollution in the province and explore the causes of higher pollutants concentrations in the region of CPUA during the heavily polluted period of January. The results demonstrate that intensive pollutants emissions and unfavorable meteorological conditions are the main causes of the heavy pollution. Besides, Weighted Potential Source Contribution Function (WPSCF) analysis indicates that local emissions remain the major contributor of

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$PM_{2.5}$ in Henan province, although emissions from the neighboring provinces (e.g. Shanxi, Shaanxi, Anhui, and Shandong) are also important contributors.

1. Introduction

Serious air pollution is believed to adversely affect human health and visibility and even can cause deaths (Valavanidis et al., 2008; Anderson et al., 2012; Xu et al., 2013). In recent years, severe regional air pollution occurs in eastern China, and significant levels are widespread across northern and central China (Rohde and Muller, 2015). Lelieveld et al. indicate that outdoor air pollution, mostly by $PM_{2.5}$, leads to a large premature death per year in China, predominantly in Beijing-Tianjin-Hebei, Yangtze river delta and Henan province (Lelieveld et al., 2015). Henan as the most populous province of China, has become one of the most seriously polluted areas in terms of ambient particulate matter concentrations in the middle east of China (van Donkelaar et al., 2010; Xu, 2012). With the center of provincial Capital city—Zhengzhou, the Central Plain Urban Agglomeration (CPUA) region has become the most populated and heavy-polluted regions in Henan province (Tian et al., 2011a; Zhang et al., 2012; Wang et al., 2012; Qiu et al., 2014). Ground-level observation data has indicated that the annual average number of haze days exceeded 100 in the region and the tendency was gradually worsening (Xu, 2012). Under suitable meteorological conditions, air pollutants can be transported to other regions from their origins of emissions. Wang et al. (2012) analyzed a heavy pollution episode over the southern Hebei area using modeling simulation, and their results indicate that pollutant transportation from Henan play a more significant role during heavy pollution periods (Wang et al., 2012). Furthermore, along with the rapid urbanization process and explosively growth of vehicles during the past decade, the type of air pollution in China has changed from the simple coal-smoke air pollution to complex air pollution, which is coupled with coal-smoke and vehicles exhaust (He et al., 2002; Molina and Molina, 2004; So et al., 2007; Tian et al., 2007). The emitted toxic pollutants in the atmosphere such as NO_X, SO_2, NH_3 and VOCs may react with O_3, hydroxyl radicals (•OH) and other reactive molecules to form secondary inorganic aerosol (SIA, including sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+)) and secondary organic aerosols (SOA), which can lead to much higher $PM_{2.5}$ concentration and the deterioration of atmospheric pollution (Liousse et al., 1996; Squizzato et al., 2013).

Emission inventory of air pollutants can not only help identify the regions with high emission intensity and the key sources with large contribution, but also can benefit for better understanding of the temporal and spatial variation characteristics of various air pollutants and guide air pollutants control management and forecast air quality.

The US, European and other developed countries have begun earlier to compile emission inventory of varied toxic air pollutants and have established a mature and complete emission inventory management system, like the US National Emission Inventory (NEI), the UK National Atmospheric Emission Inventory (NAEI), and the Australian National Pollutant Inventory (NPI). The emission inventory of Asia area has been established gradually during the past years, for example, Kato et al. evaluated the anthropogenic emissions of SO_2 and NO_X in Asia in 1992 (Kato and Akimoto, 1992), Streets et al. established an inventory of gaseous and primary aerosol emission in Asia (TRACE-P) in 2003 (Streets et al., 2003). Ohara et al. established an regional emission inventory of Asia (REAS) of 1980–2020, which contains the emissions of SO_2, NO_X, NH_3, BC, N_2O, OC, CO, CO_2, NMVOC and CH_4 (Ohara et al., 2007). Tsinghua university firstly published the Multi-resolution Emission Inventory for China (MEIC) in 2012, which provides all anthropogenic emissions of eight species, including SO_2, NO_X, CO, NMVOCs, NH_3, CO, PM_{10} and $PM_{2.5}$ in China for the year 1990–2012 and it has been widely used to deal with regional air pollution (He, 2012). Furthermore, a number of studies on air pollutants emissions in China have been reported in the past few years (Tian et al., 2002; H. Wang et al., 2009; Tian et al., 2011b; Zhao et al., 2012; Qiu et al., 2014), However, an integrated study on developing comprehensive atmospheric emission inventory of Henan province and investigating the temporal and spatial characteristic of regional air pollution is quite limited.

By coupling with emission inventory, 3-D air quality models have been increasingly applied to study the temporal and spatial pollution characteristics over China in the past few decades. Most of these studies are focused on the mainland China or the major polluted regions in China. Yet, few studies have been conducted to dedicatedly investigate the air pollution characteristics of Henan province. For instance, Wang et al. applied the MM5 and CMAQ modeling system to East Asia and Northeast China at 36- and 12-km horizontal grid resolutions, and the source contributions of major source regions and sectors to $PM_{2.5}$ concentrations in the three most polluted cities in southern Hebei are quantified by aiming at the understanding of the sources of the severe haze pollution in this region (Wang et al., 2014). Sun K. used WRF/Chem to study a severe haze episode that occurred over the Yangtze River Delta (YRD), in November 2013 (Sun et al., 2016). The online WRF/Chem model, coupled with urban canopy (UCM) and biogenic-emission models, was used to explore impacts of urban expansion on secondary organic aerosols (SOA) formation in the Pearl River Delta (PRD) (X.M. Wang et al., 2009). Zheng et al. used the Nested Air Quality Prediction Model System (NAQPMS) to assess the impact of regional transport on heavy fine particulate ($PM_{2.5}$) air pollution in Henan Province during a one-week regional haze (January 12th–19th, 2014) (Zheng et al., 2016). However, most of these studies focused on a single species or single period. The lack of urban-scale emission inventory was also one of the limitations of previous studies.

In this study, an integrated atmospheric emission inventory of primary air pollutants in Henan province for the target year of 2012 is firstly developed; then, WRF/Chem version 3.7.1 which can simulate the feedbacks of chemistry to meteorology is applied over Henan province and its heavily polluted CPUA region. Back trajectories cluster analysis is used to help explore the causes of heavier pollution in the region of CPUA and the period of January in winter. Finally, back trajectories combined with ambient measurements are used to conduct a Weighted Potential Source Contribution Function (WPSCF) analysis.

2. Methodology and data sources

2.1. Methodology

2.1.1. Methodology of calculating air pollutants emissions

We estimate the atmospheric emissions of 9 target pollutants (VOCs, SO_2, PM_{10}, $PM_{2.5}$, NO_X, NH_3, CO, BC and OC) from anthropogenic sources based on localized annual activities and source-specific emission factors in 2012. By referring to the compiled technical manuals of urban air pollutants emission inventory and taking into account of the characteristics of local emission sources in Henan, we classify the considered pollution sources into 11 major categories. i.e., power plants (PP), cement plants (CP), steel plants (SP), non-ferrous metal smelting (NFMS), other stationary combustion sources (OSCS), industrial processes sources (IPS), mobile sources (MS), anthropogenic ammonia emission sources (NH_3S), biomass burning sources (BBS), VOCs product-related sources (VOCsPS), and waste treatment sources (WTS). The specific emission source categories and the corresponding activity levels are listed in SI Table S1.