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ABSTRACT

The fast economic growth of China along the last two decades has created a strong impact on the environment. The occurrence of heavy haze pollution days is the most visible effect. Although many researchers have studied such problem, a high number of spatio-temporal limitations in the recent studies were identified. From our best knowledge the long trends of PM_{2.5} concentrations were not fully investigated in China, in particular the year-to-year trends and the seasonal and daily cycles. Therefore, in this work the PM_{2.5} concentrations collected from automatic monitors from five urban sites located in megacities with different climatic zones in China were analysed: Beijing (40°N), Chengdu (31°N), Guangzhou (23°N), Shanghai (31°N) and Shenyang (43°N). For an inter-comparison a meta-analysis was carried out.

An evaluation conducted since 1999 demonstrates that $PM_{2.5}$ concentrations have been reduced until 2008, period which match with the occurrence of the Olympic Games. However, a seasonal analysis highlight that such decrease occurs mostly during warmer seasons than cold seasons. During winter $PM_{2.5}$ concentrations are typically 1.3 to 2.7 higher than in summer. The average daily cycle shows that the lowest and highest $PM_{2.5}$ concentrations often occurs in the afternoon and evening hours respectively. Such daily variations are mostly driven by the daily variation of the boundary layer depth and emissions.

Although the PM_{2.5} levels have showing signs of improvement, even during the warming season the values are still too high in comparison with the annual environmental standards of China (35 μ g m⁻³). Moreover, during cold seasons the north regions have values twice higher than this limit. Thus, to fulfil these standards the governmental mitigation measures need to be strongly reinforced in order to optimize the daily living energy consumption, primarily in the north regions of China and during the winter periods.

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1. Introduction

In the recent decades China has been growing quickly. Today the country is not only the most populous country in the world but also one of the world's largest economies. Nevertheless, the fast economic development and high energy consumption, conjugated with stagnant atmospheric conditions and/or, in some cases, transport of regional emissions, has led to record heavy haze pollution days in many regions of the country. As result, in the last years a substantial proportion of Chinese people had their health

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affected. About 300,000 premature deaths were estimated per year due to outdoor air pollution in this country (Cohen et al., 2005; Zhang et al., 2008). Due to the severe magnitude of the problem the subject has become a major concern not only for citizens, which are requesting the implementation of efficient control policies, but also for the international community.

The main components of haze episodes are $PM_{2.5}$ concentrations, particulate matter with particle diameter smaller than 2.5 µm. Besides the impacts on climate (Maricp, 2013), this pollutant has important impacts on human health since it tends to penetrate into the gas exchange regions of the lung and blood streams causing permanent DNA mutations, heart attacks and premature deaths (Roy et al., 2012). A recent study involving 312,944 people from nine European countries revealed that for every increase of 5 µg m⁻³ in PM_{2.5} the lung cancer rate rose 18%



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(Raaschou-Nielsen et al., 2013). In China, although several studies reported acute (Chen et al., 2013) and chronic (Chu et al., 2015) human health problems, a review study concluded that the long-term health effects of $PM_{2.5}$ in the country remain unclear (Pui et al., 2014).

Due to the severe impacts of the problem several of the haze episodes recorded in China attracted a large attention from worldwide researchers (Huang et al., 2014; Zhao et al., 2011; Wang et al., 2005, 2013). Some studies report average daily values higher than 1000 μ g m⁻³ of PM_{2.5} (Wang et al., 2005) while the World Health Organization suggests a daily limit of 25 μ g m⁻³ and an annual limit of 10 μ g m⁻³ to mitigate its impacts on human health (WHO, 2005). Besides this limits, WHO defined three interim targets. The Chinese government adopted the mildest, the interim target 1 (IT1). In this case an annual limit of 35 μ g m⁻³ of PM_{2.5} and a daily limit of 75 μ g m⁻³ of PM_{2.5} was established. Nonetheless, these limits, defined to the IT1, corresponds to the highest mean concentrations reported in studies of long-term health effects, and may reflect higher but unknown historical concentrations that may have contributed to the observed health effects (WHO, 2005). According to the WHO (2005) in developed world the IT1 has been shown to be associated with significant mortality.

To study the problem, along the last two decades numerous observations of PM_{2.5} were conducted in different Chinese cities. In a literature review study, Peng et al. (2016) synthesized 23 studies of PM_{2.5} concentrations obtained between 1998 and 2011. Nevertheless, with few exceptions (e.g. (Peng et al., 2016; Zhang and Cao, 2015)) the studies conducted in recent years were mostly focused on a restricted number of cities, in particular Beijing and Shanghai (e.g. Chen et al., 2014; Tao et al., 2015; Zhao et al., 2016). Although some of them were focused on long term analysis (Chen et al., 2016), filter measurements have been mostly used to study the chemical composition of PM2.5 mass daily (Okuda et al., 2004; Zheng et al., 2005; Wang et al., 2005, 2006; Sun et al., 2004; Guo et al., 2010; Tao et al., 2013, 2014a; Li et al., 2013; Zhao et al., 2016) or weekly (He et al., 2001; Ye et al., 2003; Duan et al., 2006; Hagler et al., 2006; Chen et al., 2014; Yang et al., 2005). However, samples with duration from hours to days are not able to characterize the fast dynamics of atmosphere. Furthermore, some studies reveal that some gas and aerosol collector filters have important limitations since during the sampling collection significant loss of semi-volatile species might happen due to the evaporative processes (Dong et al., 2012).

To minimize the constraints observed, recently the filter measurement technique has been replaced by real-time measurement instruments able to characterize the fast dynamics of PM_{2.5} concentrations in the atmosphere (Zhao et al., 2009; Chang et al., 2013; Zhang and Cao, 2015; Wang et al., 2016; Lin et al., 2016). While some of the studies, based on this new technique, suggested a decrease of PM_{2.5} in China (Wang et al., 2016), a remotely sense study conducted by Peng et al. (2016) suggested that such risk was continuously expanding. According to Peng et al. (2016) the PM_{2.5} concentrations increased significantly from 1999 to 2011, especially in the central and eastern parts of China. The health risk in the central and eastern areas of the country was the highest and changes occurring faster along a south-north axis than along an east-west axis, and also faster along an east-south axis than along a west-north axis. Some authors suggested that some differences were attributed to the regional transport as well as the wet deposition by precipitation (Wang et al., 2016).

Several studies analysed significant seasonal and yearly differences of $PM_{2.5}$ mass characterization (e.g. Cao et al., 2007; Zhang and Cao, 2015; Xu et al., 2016). Still, for our best knowledge a long trend analysis based on field automatic measurements is missing. Furthermore, some author's highlight that the annual changes in PM_{2.5} pollution are yet unclear (Guo et al., 2014). Moreover, a full meta-analysis study is missing. Previous results were from studied on the PM_{2.5} concentrations dated back to about five years ago, situation of which might have changed due to the variations of emissions. Therefore, in order to clarify the trends of this pollutant, in this study automatic measurements of PM_{2.5} concentrations were analysed for five Chinese urban sites located in five different climatic zones. Based on these measurements a detailed characterization of PM_{2.5} focusing on the annual and seasonal daily cycle of PM_{2.5} concentrations was conducted and the potential causes were discussed. To perform an inter-comparison a meta-analysis was also conducted.

The paper is organized as follow. Section 2 presents the data collection and the methods applied to conduct the study. Results and discussion are presented in section 3 and the main conclusions are outlined in section 4.

2. Material and methods

To investigate the long trends of PM_{2.5} concentrations in China five urban megacities were selected. Section 2.1 presents the details about the sampling settings, while section 2.2 presents a description of the data collection process.

2.1. Sampling setting

Samples were collected in urban settings from five of the largest and most economically important megacities located in different climatic zones of Northern, Centre, Eastern and Southern China: Beijing (40°N), Chengdu (31°N), Guangzhou (23°N), Shanghai (31°N) and Shenyang (43°N). Fig. 1 shows the geographical location of such cities while Table 1 presents a short description of each one of them.

Beijing (BJ), the capital of China, is the second largest city in the country with 21.7 million of inhabitants in 2015 (BJSTATS, 2016). The city has a monsoon-humid continental climate. Summers are characterized by an increase of humidity due to the East Asian monsoon influence, while winters are cold, windy and dry due the effect of the Siberian anticyclone. Springs are generally dry with sandstorms blowing in from the Gobi Desert located approximately 400 km to the northwest. In Beijing the measurements for this study were conducted in the Chaoyang District, a mix of a residential and offices area located at approximately 8 km from the city centre (Tiananmen Square). Surrounding buildings range from 2 to 20 floors. At southeast is located a green park (1 km) while at west and east are located two main arterials, the 3rd Ring Road at 0.5 km and the 4th Ring Road at 2 km respectively.

Chengdu (CG) is the capital city of Sichuan province. This megacity is the major economic centre in Southwestern China with near 14.7 million of inhabitants in 2015 (CDSTATS, 2016). Chengdu is influenced by a humid subtropical climate. During winter the city is affected by the Siberian winds but the Qin Mountains, located in the north, help to shield from it. Even in non-rainfall periods Chengdu has one of the lowest annual sunshine totals nationally with monthly possible sunshine ranging from 16 to 38%. In Chengdu, the data for this study was collected in the Wuhou District, located at 3 km from the city centre. The place is a residential area surrounded by several banks and hotels. The main arterial is located 0.5 km south (2nd Ring Road - elevated road).

Guangzhou (GZ) is the capital city of Guangdong province. This megacity, located in South China, has 13.5 million inhabitants in 2015 (GZSTATS, 2016). The region is a developed industrial and commercial area located in the Pearl River Delta Region (PRDR) and mainly consist of floodplains. The area is a transitional zone of the East Asian monsoon system, with northeast winds in winter from

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