



Atmospheric emissions of Cu and Zn from coal combustion in China: Spatio-temporal distribution, human health effects, and short-term prediction[☆]



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ABSTRACT

China has become the largest coal consumer and important emitter of trace metals in the world. A multiple-year inventory of atmospheric copper (Cu) and zinc (Zn) emissions from coal combustion in 30 provinces of China and 4 economic sectors (power plant, industry sector, residential sector, and others) for the period of 1995–2014 has been calculated. The results indicated that the total emissions of Cu and Zn increased from 5137.70 t and 11484.16 t in 1995–7099.24 t and 14536.61 t in 2014, at an annual average growth rate of 1.90% and 1.33%, respectively. The industrial sector ranked as the leading source, followed by power plants, the residential use, and other sectors. The emissions of Cu and Zn were predominantly concentrated in the northern and eastern regions of China due to the enormous consumption of coal by the industrial and the power sectors. The emissions of Cu and Zn were closely associated with mortality and life expectancy (LE) on the basis of multiple regression analysis. Spatial econometric models suggested that Cu and Zn emissions displayed significantly positive relevance with mortality, while they exhibited negative correlation with LE. The influence of the Cu emission peaked in the north of China for both mortality and LE, while the impacts of the Zn emission on mortality and LE reached a maximum value in Xinjiang Province. The results of the grey prediction model suggested that the Cu emission would decrease to 5424.73 t, whereas the Zn emissions could reach 17402.13 t in 2020. Analysis of more specific data are imperative in order to estimate the emissions of both metals, to assess their human health effects, and then to adopt effective measures to prevent environmental pollution.

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1. Introduction

Atmospheric emissions of Cu and Zn have been a global concern due to their significant impacts on ecosystem and human health. Cu and Zn are integral micronutrients for plants and microorganism, both of which could act as activators of enzyme reactions or impose catalytic properties (Lefevre et al., 2014; Zhao et al., 2014). On the other hand, they can lead to toxic effects when they exceed plant or microorganism requirement (Bing et al., 2016). During the past

years, a rapid increasing of the Cu and Zn components in the aerosol particles in the urban atmosphere of China has been frequently reported (Cong et al., 2010). It was well documented that excessive exposure to Cu and Zn elements in the ambient air will cause chronic bronchitis, emphysema, peritonitis, asthma, and even lung cancer (Zhao et al., 2012). Given the toxicity of Cu and Zn, it is imperative to investigate and evaluate their emission characteristics in order to reduce their potential risk to public health.

Coal combustion is one of the major anthropogenic emission sources for trace elements such as Cu and Zn (Fytianos et al., 1998; Xu et al., 2004). There are two dominant pathways to release Cu and Zn to the environment during coal combustion. Most of Cu and Zn could be volatilized directly into the atmosphere via a high-temperature process. Also, they can enter the atmosphere through leaching of solid combustion by-products during their

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disposal (Llorens et al., 2001). Once Cu and Zn are released into the atmosphere, they tend to attach to fine particles and penetrate deeply into the respiratory tree (Uysal and Schapira, 2003). Besides, they could also transport over a long distance and eventually settle down, resulting in the adverse effects on the remote environment, such as mountain ecosystem and Polar regions (Han et al., 2015). Coal is also the most important energy resource in China as the result of its abundant reserves and relatively lower price, and it accounts for about 70% of the current primary energy consumption (Chen et al., 2013a). Moreover, the coal consumption in China keeps a steady growth during the past years and is predicted to decrease only slightly over the next decades (Luo et al., 2002). Thus, it is imperative to decipher the characteristics of the Cu and Zn emissions from coal combustion.

A growing body of studies centered on toxic trace elements and gaseous pollutants released from coal combustion and their effects on air pollution in China. Chen et al. (2013a,b) estimated F, As, Se, Hg, and Sb emissions from coal-fired power and heat generation, and found that emissions of these trace elements increased sharply from 2005 to 2009. Tian et al. (2011) reported the antimony (Sb) emission inventories from coal combustion in China, and suggested that total Sb emission released from coal combustion was much higher by four times in 2007 than that in 1980. In addition, particulate matter (PM), SO₂, and NO_x emissions in China have been estimated by some studies (Tian et al., 2013, 2014). To date, the report about atmospheric Cu and Zn emissions from coal combustion in China were relatively scarce. Moreover, the effects of Cu and Zn emitted from coal combustion on the human health are not known yet.

Air pollution has been considered to be closely linked to human health, including increased mortality and morbidity risk, increased rates of hospital admission visits, and exacerbation of chronic respiratory conditions. Kan and Chen, (2004) reported that total suspended particle (TSP), PM_{2.5} and black smoke in the ambient air appeared to display the consistent association with health outcomes. Dholakia et al. (2013) observed that the adoption of advanced control technologies to decrease PM_{2.5} were associated with the reduction of mortality. Chen et al. (2012) found that the NO₂ concentration was significantly related to the cardiovascular mortality in 17 cities of China. However, relatively few studies have examined that the comprehensive impacts of trace metal emissions on human health, especially from the spatial perspective. Although multiple regression model and Poisson generalized additive model could explain the relevance of pollutants and health outcomes, these methods neglect the autocorrelation in spatial level (Tang et al., 2017). Spatial econometric model could be used to accurately decipher the spatial correlation of pollutants and health risk. Therefore, many spatial econometric models were applied to investigate the health effects of the Cu and Zn emissions from coal combustion in the present study.

The main objectives of this work are: (1) to investigate the spatial and temporal variation of the Cu and Zn emissions during coal-fired process; (2) to assess the relationship between Cu and Zn emissions and human health from spatial perspective; and (3) to predict the Cu and Zn emissions in the near future in China. This work could arouse central and local government to pay more attention to trace metal pollution.

2. Materials and methods

2.1. Estimation of the Cu and Zn emissions

Herein, Cu and Zn emitted from coal combustion were calculated by integrating the provincial-level coal consumption data and relevant emission factors. The Cu and Zn emissions were classified

into four main economic sectors (power plants, industrial sector, residential sector, and other use sector). Formula (1) was adopted to calculate the Cu and Zn emissions:

$$E(t) = \sum_i \sum_j C_{ij}(t) A_{ij}(t) EF_{ij}(t) (1 - \eta_{PM}(t)) (1 - \eta_{FGD}(t)) \quad (1)$$

where E is the total atmospheric emissions of Cu and Zn from coal combustion; C is the mean content of Cu and Zn in coal consumed in a province; A is the coal consumption in each province; EF denotes the concentration of Cu and Zn in flue gas emitted from coal combustion facility; η_{PM} and η_{FGD} are the fraction of Cu and Zn eliminated by the existing dust collectors and flue gas desulfurization (FGD) devices, respectively; i denotes the region (province/autonomous region/municipality); t denotes the calendar year; and j is the emission source classified by economic sectors.

2.1.1. Mean content of Cu and Zn in coal

The concentrations of Cu and Zn in coal from different places varies substantially because of the difference of both coal-forming plants and coal-forming geological environments. The provincial Cu and Zn content of coal are summarized in Tables 1 and 2, respectively. In the present study, field test content data of Cu (831 samples) and Zn (942 samples) for different Chinese coal were collected from available literature. The corresponding data of Qinghai, Beijing, Tianjin, Shanghai, and Hainan are not available. Thus, the average Cu and Zn content in the surrounding Province were chosen to replace the missing values. Tibet Autonomous Region, Hong Kong and Macau Special Administrative Region, and Taiwan Province were not included in our study. Among of these 30 provinces, the lowest mean concentration of Cu is 1.5 µg/g in Ningxia Autonomous Region, whereas the highest concentration is 65.9 µg/g in Sichuan province. The lowest average concentration of Zn is 9.8 µg/g in Anhui Province, while the highest one is 88.1 µg/g in Guangxi Autonomous Region. No significant variation in Cu and Zn content in coal samples was reported among the different years. Therefore, it was assumed that the mean content of Cu and Zn in coal do not change during the period in our study.

2.1.2. Coal consumption

Coal combustion sources were classified into four sectors including power plants, industrial sector, residential sector, and other sector based on the operational patterns of combustion furnaces and their functions. Coal consumptions of different sectors in China from 1995 to 2014 were cited from Chinese Energy Statistical Yearbook of 1996–2015 (<http://www.stats.gov.cn/tjsj/ndsj/>). The coal consumption of Chongqing was included in Sichuan Province before 1997. Coal consumption by power plants and industrial sector increased dramatically since the beginning of the 21st century. Among the coal-consuming sectors, the power plants are the leading sector in coal growth, which increases by 16.7% annually, while the industrial sector (4.3%) and other sector (2.6%) have the moderate increases in coal use. The residential sector decreases slightly with the rate of 1.9% annually in coal consumption because coal using are replaced by the cleaner gaseous fuels and/or the electricity in many areas of China.

2.1.3. Emission factors and removal efficiencies of Cu and Zn

It is necessary to develop a detailed illustration of the methods in which coal is burnt in China because the emission rates of Cu and Zn greatly lie on operation conditions and combustion technologies. Four types of coal combustion facilities are pulverized-coal boilers, stoker fired boilers, fluidized-bed furnaces and coke furnaces, respectively. Pulverized-coal boilers were widely used in coal-fired power plants in the most regions of China, accounting for

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