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Optimizing critical source control of five priority-regulatory trace elements from industrial wastewater in China: Implications for health management☆

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ABSTRACT

Anthropogenic emissions of toxic trace elements (TEs) have caused worldwide concern due to their adverse effects on human health and ecosystems. Based on a stochastic simulation of factors' probability distribution, we established a bottom-up model to estimate the amounts of five priority-regulatory TEs released to aquatic environments from industrial processes in China. Total TE emissions in China in 2010 were estimated at approximately 2.27 t of Hg, 310.09 t of As, 318.17 t of Pb, 79.72 t of Cd, and 1040.32 t of Cr. Raw chemicals, smelting, and mining were the leading sources of TE emissions. There are apparent regional differences in TE pollution. TE emissions are much higher in eastern and central China than in the western provinces and are higher in the south than in the north. This spatial distribution was characterized in detail by allocating the emissions to 10 km × 10 km grid cells. Furthermore, the risk control for the overall emission grid was optimized according to each cell's emission and risk rank. The results show that to control 80% of TE emissions from major sources, the number of top-priority control cells would be between 200 and 400, and less than 10% of the total population would be positively affected. Based on TE risk rankings, decreasing the population weighted risk would increase the number of controlled cells by a factor of 0.3–0.5, but the affected population would increase by a factor of 0.8–1.5. In this case, the adverse effects on people's health would be reduced significantly. Finally, an optimized strategy to control TE emissions is proposed in terms of a cost-benefit trade-off. The estimates in this paper can be used to help establish a regional TE inventory and cyclic simulation, and it can also play supporting roles in minimizing TE health risks and maximizing resilience.

1. Introduction

Trace elements (TEs) (Antoniadis et al., 2017; Shaheen et al., 2013) such as mercury (Hg), cadmium (Cd), arsenic (As), lead (Pb), and chromium (Cr) are elements that are present naturally in the environment but can cause serious damage to ecosystems and human health in cases of environmental pollution (Futsaeter and Wilson, 2013; Babula et al., 2008). Certain TEs can be easily converted to more-persistent and toxic compounds after entering the environment, at which point they can cause global pollution and health problems via bioaccumulation and dissemination through the food chain (Lin et al., 2012; Chandra Sekhar et al., 2004). For example, Cd, As, and Pb can accumulate in soil and enter the human body through the consumption of rice planted in such soil (Williams et al., 2009; Zhu et al., 2008), and As is particularly harmful to children (Yamaguchi et al., 2014; Norton et al., 2010).

Human activity is the main factor that affects the production of TEs in the terrestrial chemical cycle (Pirrone et al., 2010; Rauch and

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Pacyna, 2009). Given present levels of economic development, when the exploitation of TE resources by human activities—such as chemical production, dyeing and printing, mining, burning of fossil fuels, metal smelting, and irrigation with wastewater—accumulates to a certain level, TE pollutants are emitted in excessive amounts (Vitousek et al., 1997; Sen and Peucker-Ehrenbrink, 2012), which results in frequent TE pollution accidents worldwide. The main TE sinks are the atmosphere, soil, and water (Nriagu and Pacyna, 1988; Nriagu, 1979). China is an important center of TE emissions (Lin et al., 2012; Shetty et al., 2008; Pacyna et al., 2010) and has experienced a high frequency of TE pollution accidents due to anthropogenic emissions (Cheng et al., 2015). There were approximately 30 major TE pollution incidents during the 2009–2011 period alone, including the 2009 Hunan Liuyang Cd incident and the 2012 Guangxi Cd spill; limit-violating Pb levels in the blood of children in Fengxiang, Shaanxi; Jiyuan, Henan; and Qingyuan, Guangdong; Pb pollution in Neijiang, Sichuan; and As pollution in Linyi, Shandong (China Youth Daily Website, 2012; Chinawater net website, 2015). To further restrict increasing TE emissions, the State Council of the People's Republic of China ratified China's "Twelfth Five-Year Plan (FYP) for the Comprehensive Prevention and Control of Trace Element Pollution" in 2011 (The Central People's Government of the People's Republic of China, 2011). In this FYP, 14 provinces including Inner Mongolia, Jiangsu, Zhejiang, and Jiangxi were identified as the critical control provinces. Five industries including the non-ferrous trace element mining and smelting industries and the lead-acid battery manufacturing industry were identified as the critical industries to control. Hg, Cd, Cr, Pb, and As were listed as precedent-controlled toxic TEs.

Many studies have contributed to a global TE inventory (Futsaeter and Wilson, 2013). Nriagu et al (Nriagu and Pacyna, 1988; Nriagu, 1979, 1989). first calculated global emissions of TEs, and Pacyna and Pirrone also provided a global TE emission inventory. Regional inventories were then developed for Europe (Thevenot et al., 2007; Mayes et al., 2010), China (Cheng et al., 2015; Tian et al., 2010), South Korea (Pudasainee et al., 2014), and Japan (Nansai et al., 2012). These early studies focused on the effects of anthropogenic sources on the distribution of global TE emissions (Nriagu and Pacyna, 1988; Pacyna et al., 2010; Cheng et al., 2015; Nansai et al., 2012; Tian et al., 2012a,b; Tian et al., 2014a,b; 2015). Subsequently, studies have gradually expanded to natural sources (Pirrone et al., 2010; Shetty et al., 2008). Anthropogenic emissions are much greater than natural emissions, particularly emissions of Cd and Pb. Anthropogenic Cd and Pb emissions accounted for more than 10% in 1975 (Nriagu, 1979). Several studies have resulted in an inventory of anthropogenic atmospheric TE emissions including emissions from fossil fuels, metal smelting, and solid waste incineration (Tian et al., 2010, 2012a, 2012b; Wu et al., 2010; Tian et al., 2014a,b). TEs in aquatic environment cannot be ignored (Ip et al., 2007; Sun et al., 2011), yet there are very few studies of the sources of wastewater TE emissions, particularly studies involving refined gridding (Lin et al., 2012; Liu et al., 2016; AMAP/UNEP, 2013). Existing research on Hg includes the use of an emission factor model of the probabilistic distribution of Hg concentrations from various industrial sectors and the volume of wastewater discharged. Activity levels and emission factors are most important to the model. These emission factors vary greatly spatially and in terms of industrial levels. Previous studies often used a top-down model to perform an accounting, followed by a decomposition. They lacked localized data, particularly factors applied to industries in different provinces.

By establishing an emission factor model for 388 × 39 suburban sub-industries, we performed a bottom-up estimate of the aqueous emissions of the five precedent-controlled TEs by industrial sources in China. Sub-industries have rarely been considered in previous inventories (Wu et al., 2015). In contrast to previous provincial and regional emission models, the emission factors in the present method are based on a probabilistic distribution on a prefectural level. Based on these results, we, in the present study, combined a gridding of population with a gridding of TE emissions to evaluate environmental risk posed by TEs, and we conducted a cost-benefit trade-off optimization for establishing controls in the critical grids, i.e., those where the risks are greatest. This type of grid-based population-weighted TE risk assessment has rarely been performed before. TEs are bound to suspended sediment particles during transport and pose risks to environmental quality in downstream areas (Žagar et al., 2006; Li and Zhang, 2010a,b). Generally, local generation and transport from upstream are the two main ways through which emitted TEs affect water quality and related risks. In this paper, we focus on risks posed by local emissions. The risks posed by downstream transport were beyond the scope of this study. The relevant calculations can be found in the second section of this paper. The provided benefits are based on a definition of the control level of TE risk. The results of simulations of major industrial emitters and their spatial variations are explored and discussed in Section 3. Based on the risk ranking of the grid characterization and the cost-benefit trade-offs, suggestions for optimal nationwide TE control in the selected critical grid cells are proposed in Section 3. Generally, the original contribution of this research to the current knowledge body can be stated as follows. Firstly, it applied new data on activity level and more reliable data on localized emission factors for 388 × 39 suburban sub-industries in China, which have never been used in prior work. The application of good-quality and refined data would greatly improve the accuracy of emission estimate and risk assessment on aquatic trace elements. Secondly, it provides 10-km resolution emissions and identifies spatial hotspots of heavy metal risk in China, which provides important implications for targeted heavy metal pollution control and health intervention. Thirdly, open publication of our results makes it possible to further explore the aquatic heavy metal related health damage and its determinants in the future research.

2. Methodology, data sources, and key assumptions

2.1. Estimation of TE emissions

A bottom-up approach was adopted to evaluate the aqueous emissions of the five precedent-controlled toxic TEs (Hg, As, Pb, Cd, and Cr) from industrial sources in China. Hg emissions were calculated by combining the activity level of various prefectural industries with the best available TE-specific emission factors (EFs). The activity level represents the gross outputs of the various prefectural industrial sectors. The EF method relies on large amounts of data, and more data leads to more-reliable results (Fang et al., 2013). Thus, based on data from 388 cities in the China Pollution Source Census dataset, we developed an integrated EF model to calculate the annual emissions in China. The bottom-up estimates of emissions were developed using the following equations:

$$E_T = \sum_i \sum_j \left(A_{i,j}(t) \cdot EF_{i,j,e}(t)\right) \quad and$$
$$EF_{i,j,e}(t) = COI_{i,j,e}(t) \cdot \left(1 - PF_{i,j,e}(t)\right),$$

where E_T is the total emissions of TEs, A is the annual activity level, EF is the TE-specific EFs for the industrial sectors, COI is the fraction of TEs released from the sources of industrial processes, PF represents the fraction of TEs removed by pollution control devices installed on industrial emitters, t is the calendar year, i is the city, j is