Life Cycle Impact Assessment of Airborne Metal Pollution near Selected Iron and Steelmaking Industrial Areas in China

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ABSTRACT

Toxic metals in particulate matter pose a significant health risk to humans via inhalation and dermal exposure. Additionally, airborne pollution has negative impacts on terrestrial and aquatic quality as a result of atmospheric deposition. Iron and steelmaking industry is considered as a major contributor to airborne metal pollution. Given that China has been the largest steel producer and consumer since 1996, a detailed investigation of airborne metal pollution is required to assess the potential risks to both human health and ecosystem quality near iron and steelmaking areas in China. This study applied an environmental impact assessment approach to evaluate the freshwater ecotoxicity, terrestrial ecotoxicity, marine ecotoxicity and human toxicity caused by metal concentrations in PM_{1.1}, PM_{1.1-2.1} and PM_{2.1-9.0} fractions. Results showed that heavy metals Cu and Zn associated with steelmaking activities were largely responsible for aquatic and terrestrial ecotoxicity. This study also found that As and Pb contamination presented the largest fraction of the impacts on human toxicity. Findings presented in this study showed that more stringent control measures are required to improve the environmental performance of the iron and steelmaking industries in China.

Keywords: PM₁; PM_{2.5}; Human toxicity; Aquatic ecotoxicity; Terrestrial ecotoxicity.

INTRODUCTION

Air pollution is a by-product of the rapid economic growth with increasing global urbanization and industrialization. According to the World Health Organization, it was estimated that 91% of the global population lives in places where the air quality is below the threshold guidelines (WHO, 2018). People living under poor air quality conditions have increased potential to develop a range of pollution-related illnesses, such as heart disease, stroke, chronic obstructive pulmonary disease, lung cancer and acute respiratory infections (Cheng *et al.*, 2019).

Particulate matter (PM), as one of the common air pollutants, is a proxy indicator of air quality due to the strong association with public health impacts (WHO, 2018; Sarkodie *et al.*, 2019). Particles with diameters of 10 µm or less (PM₁₀) can penetrate deep inside human lungs, while PM_{2.5} particles have abilities to go through lung barriers and enter the blood system (WHO, 2018). It has been evidenced that 3% of cardiopulmonary and 5% of lung cancer deaths are associated with the global PM pollution (WHO, 2013).

One of the toxic components adhered to the PM particles is heavy metals, especially As and Pb, which present high toxicity at even trace concentrations (Jiang *et al.*, 2019). For example, clear clinic evidence of skin lesions, such as ulceration and cancers, was found for people who live at intensive high-As-containing coal burning areas (Liu *et al.*, 2002). Airborne Pb was also found to be responsible for the elevated Pb concentrations in children blood samples (Taylor *et al.*, 2019). Both contaminations have deleterious effects on children's neurological systems, behavioural and cognitive abilities (Calderon *et al.*, 2001; Wright *et al.*, 2006; Wasserman *et al.*, 2016).

Airborne particles incorporated with toxic metals can be deposited and further pollute surface waters, such as rivers, lakes and streams (Wesely and Hicks, 2000). According to the United States Environmental Protection Agency (U.S. EPA, 2016a, b), nearly half of rivers and streams (> 40%) as well as more than one-third of lakes in the U.S. were polluted and this fraction would be much larger in developing countries. Water pollution, such as heavy metal pollution, impairs the aquatic quality and poses a risk to plants and animals that depend on water to survive (Wilde *et al.*, 2006; Lavery *et al.*, 2009).

PM emissions also present a dominant contribution to terrestrial ecotoxicity (Wesely and Hicks, 2000). The airborne-metal particles can be carried to the ground as formations of

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raindrops, snowflakes or fog resulting in soil pollution. Toxic metals in soils are considered as a highly persistent pollutant due to their nondegradable property (Borggaard *et al.*, 2009). For example, the Pb contamination in soil can remain up to 700 years after deposition (Semlali *et al.*, 2004). Hence, the airborne metals also pose a significant risk to terrestrial health.

Given the high toxicity of airborne metal particles to humans and ecosystems, identifying air pollution sources is required for environmental management and remediation. Previous studies showed that the iron and steelmaking industries are important contributors to airborne metal emissions (WHO, 2007; Mohiuddin *et al.*, 2014; Mohiuddin *et al.*, 2016; Yang *et al.*, 2019). Hence, this study aims to apply environmental impact assessment method to identify the key toxic metals that pose risks to freshwater ecotoxicity, terrestrial ecotoxicity, marine ecotoxicity and human toxicity near iron and steelmaking areas with high population densities in China.

METHODS

Sampling Information

Five sampling sites, including four iron and steelmaking areas and a background site at Ningbo Nottingham University (UN) were selected in this study. The four steel plants were located in cities of Kunming (KM), Wuhan (WH), Nanjing (NJ) and Ningbo (NB), China (Fig. 1). Their background information, including population and meteorological data at each sampling site, are detailed in Table 1.

The steel plant at KM was built in 1939, which was the oldest facility among the sampled areas. There were three blast oxygen furnaces (BOFs) and one blast furnace (BF) at KM plant with iron and steel production of seven million tonnes per year. Steel plants located at WH and NJ were both built in 1958. The WH plant had an annual production of iron and steel up to 20 million tonnes with eight BFs and ten

BOFs facilities, while the NJ plant produced 9 million tonnes of iron and steel every year with five BFs, six BOFs and one electric arc furnace (EAF). The NB plant was established in 2003 equipped with two BFs and three BOFs with an annual iron and steel production of 4 million tonnes.

Sample Collection

Air filter samples at each sampling site were collected at local meteorological sites or on the roof of high buildings at less than 1 km from the steelmaking emission sources. A non-viable Andersen cascade impactor with 8 aluminium state plates (Model 20-800, Tisch Environmental) was applied to collect atmospheric particles at five sampling sites. The 50% cut off diameters (D_{50}) of the Andersen sampler stages were 9.0, 5.8, 4.7, 3.3, 2.1, 1.1, 0.65 and $< 0.43 \mu\text{m}$. The air flow was set up at 28.3 L min^{-1} for 24 hours during sampling with similar environmental and meteorological conditions. All samples were stored at 4°C until analysis.

Sample Analysis

Air filter samples were subjected to trace element analysis of As, Ba, Cd, Cr, Cu, Mn, Pb, Sb, V and Zn at the National Measurement Institute in Sydney using an inductively coupled plasma mass spectrometer (Agilent 7900).

Every sample batch ($n = 20$) included a filter blank and duplicate, blank spike, blank matrix and matrix spikes. Procedural blanks were below the Limit of Reporting of $< 0.01 \text{ mg kg}^{-1}$ for As, Ba, Cd, Cr, Cu, Mn, Pb, Sb, V and Zn. Recovery rates for As, Ba, Cd, Cr, Cu, Mn, Pb, Sb, V and Zn ranged between 95 and 106 % for all PM samples. The results on the concentration of each metal in the collected particle size ranges for the sampling sites are detailed by Yang *et al.* (2019).

Environmental Impact Assessment

Life Cycle Impact Assessment (LCIA) methods are used to establish a link between emissions produced during

Fig. 1. Sampling locations in this study.

Table 1. Background information at sampling cities Kunming (KM), Wuhan (WH), Nanjing (NJ) and Ningbo (NB). Population data was obtained from National Bureau of Statistics of China in 2017. The meteorological information during sampling period was obtained from China National Environmental Monitoring Centre.

Cities	Population (million)	Sampling date	PM _{2.5} ($\mu\text{g m}^{-3}$)	PM ₁₀ ($\mu\text{g m}^{-3}$)	Relatively humidity (%)	Temperature ($^{\circ}\text{C}$)	Wind speed (m s^{-1})	Precipitation (mm)
Min–max								
KM	5.6	2017/04/29–05/05	22–38	63–95	--	10–26	0.2–2.9	0
WH	8.5	2017/05/14–05/20	41–75	89–163	49–100	18–33	0.1–4.6	5.6
NJ	6.8	2017/05/28–06/04	25–72	77–118	21–86	19–34	0.2–6.8	0
NB	6.0	2017/06/13–06/19; 2017/06/29–07/01 ^a 2017/05/17–05/23 ^b	13–35	18–65	49–96	17–28	0.1–6.4	67 ^a 0 ^b

a. sampling period near iron and steelmaking area.

b. sampling period on UN campus.

--no available data.

manufacturing process and their potential environmental impacts. In this study, the OpenLCA 1.9.0 software was applied to assess environmental impacts caused by toxic metals in particulate matter collected near iron and steelmaking areas in China. PM metal concentrations at fractions PM_{2.1-9.0}, PM_{1.1-2.1}, and PM_{1.1} were input parameters as the End-of-Life modelling for 1 m³ of air as a reference flow property.

The ReCiPe Midpoint LCIA method was used for the present study to evaluate freshwater ecotoxicity, terrestrial ecotoxicity, marine ecotoxicity and human toxicity at high population densities. The fate and effects of metal emissions were expressed in kg 1,4-dichlorobenzene-equivalents (1,4-DB eq) in the midpoint assessment, which were calculated by the OpenLCA software.

RESULTS AND DISCUSSION

The PM particles collected in this study were subjected to mass concentration analysis to investigate the distribution of particle sizes emitted from iron and steelmaking plants in China. The study further applied the environmental impact assessment to determine the risks of PM particles presented across the four categories from surface freshwater, terrestrial and marine ecotoxicity to human health.

Mass Concentrations

The mass concentrations at fractions of PM_{1.1}, PM_{1.1-2.1} and PM_{2.1-9.0} are detailed in Fig. 2, and the public air quality data (PM_{2.5} and PM₁₀) derived from China National Environmental Monitoring Centre (CNEMC) are compiled in Table 1.

According to the CNEMC air quality data, the air quality index (AQI) at the sampling site KM was ‘Good’ (AQI = 51–100) with PM_{2.5} and PM₁₀ concentrations ranging from 22–38 $\mu\text{g m}^{-3}$ and 63–95 $\mu\text{g m}^{-3}$, respectively (Table 1). However, mass concentrations of PM_{1.1} (25–41 $\mu\text{g m}^{-3}$) and PM_{9.0} (83–145 $\mu\text{g m}^{-3}$) at KM measured in this study were higher than the CNEMC data. This was also found at NB site where the official AQI was ‘Excellent’ (AQI = 0–50) with the minimum values of PM_{2.5} and PM₁₀ at 13 and 18 $\mu\text{g m}^{-3}$, respectively, while the sampling results presented in this study

near the steel manufacturing area were 23 and 33 $\mu\text{g m}^{-3}$ for PM_{2.1} and PM_{9.0}, respectively. The elevated PM mass concentrations near iron and steel areas indicated the levels of impact of iron and steelmaking activities on local air pollution in China.

The results also showed that the mass concentration of the coarse PM fraction at 2.1–9.0 μm diameters at KM was significantly higher than other fractions (Fig. 2). This could be a result of the age of the steel plant at KM site, which has been operating for approximately 70 years and was equipped with limited particle control measures. The mass concentration of PM particles collected from WH and NJ had similar PM_{2.1-9.0} and PM_{1.1-2.1} fractions, but NJ had a slightly higher PM_{1.1} mass concentrations than WH. Steel plants at WH and NJ were both built in 1958 and have been upgraded several times during operation.

The three PM fractions collected at NB and UN sites had similar mass concentrations. Although NB site was located in an iron and steelmaking industrial area, PM samples were collected at upwind of emissions as the downwind was the East China Sea. The industrial steelmaking facility at the NB site was also of the most recent age with modern particle capture devices.

Environmental Impact Assessment

The environmental impact assessment categories investigated in this work using the measured trace metal concentrations for the three PM fractions are presented in Fig. 3. At the coarse fraction of PM_{2.1-9.0}, the sampling site of KM showed the highest ecotoxicity for freshwater, terrestrial, marine and human, followed by WH, NJ, NB and UN for the same size fraction. This may be because the steelwork at the KM site is the oldest and may be a reflection of the age of the facility. The KM steelwork plant is also located next to a busy main road. According to the previous studies by Barmpadimos *et al.* (2011) and Charron and Harrison (2005), the traffic emissions related to tyre and brake abrasion were also a significant contributor to the coarse PM particles. Hence, the long running operation of KM plant and the surrounded heavy traffic dominated the coarse PM pollution at KM.

Fig. 2. Average mass concentrations ($\mu\text{g m}^{-3}$) at fractions of $\text{PM}_{2.1-9.0}$, $\text{PM}_{1.1-2.1}$ and $\text{PM}_{1.1}$ across five sampling sites.

Fig. 3. Life cycle impact assessment of metal concentrations at fractions of $\text{PM}_{2.1-9.0}$, $\text{PM}_{1.1-2.1}$ and $\text{PM}_{1.1}$.

The same order of $\text{KM} > \text{WH} > \text{NJ} > \text{NB} > \text{UN}$ at $\text{PM}_{2.1-9.0}$ fractions was also found for the intermodal fraction of $\text{PM}_{1.1-2.1}$ for the human toxicity (Fig. 3). However, the maximum freshwater, marine and terrestrial ecotoxicity were found at the WH site where the steelwork plant was built in 1958 and had the largest annual iron and steel production of 20 million tonnes among the four investigated steelworks.

The fine $\text{PM}_{1.1}$ particles at WH, NJ, NB and UN had higher values in the categories of freshwater ecotoxicity, terrestrial ecotoxicity, marine ecotoxicity and human toxicity than their corresponding $\text{PM}_{1.1-2.1}$ and $\text{PM}_{2.1-9.0}$ fractions at each site. This is because small particles tend to be more polluted with heavy metals, due to the nucleation and coagulation mode of particle formation mechanisms, than the intermodal and especially the coarse fractions, where the mechanically

generated particles dominate (Samara and Voutsas, 2005; Hassanvand *et al.*, 2015). Furthermore, $\text{PM}_{1.1}$ particles can travel up to tens of kilometres in the atmosphere (WHO, 2006), posing a significant ecological risk as well as carcinogenic and mutagenic respiratory risks to humans over a large geographical area (Saeedi *et al.*, 2012; Zajusz-Zubek *et al.*, 2017).

Freshwater Ecotoxicity

The maximum freshwater ecotoxicity was found to be caused by $\text{PM}_{2.1-9.0}$ contamination at the KM steelmaking site (Fig. 4). The largest contributor to freshwater ecotoxicity at $\text{PM}_{2.1-9.0}$ was V with 55% contribution, followed by Cu with 26%, while the contribution of other elements to the freshwater ecotoxicity ranged from 0.05% to 6.6% (Fig. 4).

Previous studies showed that V contamination at coarse particles was used as an indicator of diesel combustion (Onat *et al.*, 2013; Wei *et al.*, 2014; Ogundele *et al.*, 2016). Hence, the traffic in addition to the local steelmaking activities were the dominant sources resulting in the freshwater ecotoxicity at the KM sampling site.

In addition to KM, the freshwater ecotoxicity at WH, NJ, NB and UN were largely contributed by Cu ranging from 45% to 77% followed by Zn ranging from 10% to 38% (Fig. 4). Cu and Zn contamination have been observed to play a significant negative role in the diversity and trophic structure of the biological communities, such as freshwater mussel and algae in the aquatic systems (Wilde *et al.*, 2006; Wang *et al.*, 2010; Wright *et al.*, 2018).

Terrestrial Ecotoxicity

The largest terrestrial ecotoxicity was found in the $PM_{1.1}$ particle size range at WH with $1.31E-09$ (kg 1,4-DB eq) (Fig. 5). The dominant contributor was Cu which was responsible for 72% of the terrestrial ecotoxicity at WH. Compared to $PM_{1.1}$ at WH, $PM_{1.1}$ at NJ posed a slightly lower terrestrial ecotoxicity dominated by Zn with 54% contribution (Fig. 5).

Higher Zn contribution for $PM_{1.1}$ at site NJ compared to WH may be related to the differences in the steelmaking facilities. Steelwork plants at NJ and WH both have blast furnaces (BF) and blast oxygen furnaces (BOF) for steel production, but NJ plant has additional electric arc furnace (EAF) for processing of melting scrap for recycling of steel (Yang *et al.*, 2019). It has been evidenced that EAF generates more Zn emissions than the BF-BOF route (8% vs. 19.4%) (Nyirenda, 1991; Jha, 2001).

Marine Ecotoxicity

Similar to terrestrial ecotoxicity, $PM_{1.1}$ at WH and NJ were found to contribute to higher marine ecotoxicity (Fig. 6). Cu in $PM_{1.1}$ at WH and NJ contributed 76% and 45% of the total terrestrial ecotoxicity. In addition to Cu, the Zn contamination in $PM_{1.1}$ was responsible for 19% and 46% of marine ecotoxicity at WH and NJ, respectively.

Both Cu and Zn are essential elements with functions in oxygen transport (e.g., Cu) (Kim *et al.*, 2008) and enzyme co-factors or metalloenzymes (e.g., Cu and Zn) (Soetan *et al.*, 2010). However, elevated Cu and Zn concentrations can result in marine pollution, damaging the quality of sediments (Miller *et al.*, 2000) and posing a risk to marine animals (Lavery *et al.*, 2009) and other organisms (Debelius *et al.*, 2009).

Human Toxicity

$PM_{1.1}$ had significantly higher human toxicity than the corresponding fractions of $PM_{1.1-2.1}$ and $PM_{2.1-9.0}$ at the five sampling sites (Fig. 7). PM emissions at the three size fractions in KM, WH and NJ posed higher health risks to humans than UN as a background site and NB site which has a flue gas recirculation system with 45% reduction of PM emissions during the steelmaking process (CEIA, 2017).

The human toxicity of PM emissions was dominated by Pb contamination, especially for the fine $PM_{1.1}$ particles. For example, the contribution of Pb on human toxicity of $PM_{1.1}$ size fraction for sites KM and NJ was up to 87% and 76%, respectively (Fig. 7). Airborne Pb emissions have cumulative toxic impacts on humans and adversely affect the neurological, hematological, gastrointestinal, cardiovascular and renal systems (U.S. EPA, 2017). As a result, around 0.6% of the global burden of disease is estimated to be associated with exposure to Pb (WHO, 2010b).

Fig. 4. Life cycle impact assessment of freshwater ecotoxicity based on metal concentrations at fractions of $PM_{2.1-9.0}$, $PM_{1.1-2.1}$ and $PM_{1.1}$.

Fig. 5. Life cycle impact assessment of terrestrial ecotoxicity based on metal concentrations at fractions of PM_{2.1-9.0}, PM_{1.1-2.1} and PM_{1.1}.

Fig. 6. Life cycle impact assessment of marine ecotoxicity based on metal concentrations at fractions of PM_{2.1-9.0}, PM_{1.1-2.1} and PM_{1.1}.

The contributions of As contamination to human toxicity were stable at the three size fractions which were 8%–19% at PM_{2.1-9.0}, 6%–26% at PM_{1.1-2.1} and 6%–26% at PM_{1.1} particles. Long-term exposure to airborne As emissions can increase the risks of developing lung cancer, especially at fine particles of PM_{1.1} which can penetrate deep into the alveolar regions of the human lungs (WHO, 2010a; Poh *et*

al., 2018).

In addition to As and Pb contamination, airborne Mn was responsible for 2%–11% of human toxicity at PM_{1.1}, but increased to 8%–18% at PM_{1.1-2.1} and reached maximum contribution of 23%–34% at PM_{2.1-9.0} size fractions. Unlike As and Pb, which are classified as carcinogenic to humans (Group 1) and possibly carcinogenic to humans (Group 2A),

Fig. 7. Life cycle impact assessment of human toxicity based on metal concentrations at fractions of PM_{2.1-9.0}, PM_{1.1-2.1} and PM_{1.1}.

respectively (WHO, 2010a, b), Mn is considered as a nutritional element under a safe limit (ATSDR, 2012). However, exceeding Mn concentrations can cause environmental implications with potential risks to human health (Elder *et al.*, 2006), especially for the children population. A number of studies documented the neurotoxic effects of Mn associated with learning disabilities and deficits in intellectual functions of children (Riojas-Rodriguez *et al.*, 2010).

CONCLUSION

This study aimed to investigate the contamination of PM particles near iron and steelmaking areas in China. PM samples at ranges of less than 1.1 μm , 1.1–2.1 μm and 2.1–9.0 μm in diameter were collected in this study. All the samples were subject to mass concentration and trace element analyses. LCIA method was used to assess the impact of trace element contamination at different PM fractions on human and ecological health.

The mass concentration results showed that PM samples collected from intensive iron and steelmaking areas were higher than those collected from background areas and the published PM_{2.5} and PM₁₀ data online at each city, confirming the industrial emission played a significant role to the local air quality. The LCIA results showed that PM metal emission had a much higher human toxicity than freshwater, terrestrial and marine ecotoxicity. Among the three PM fractions, PM_{1.1} particles posed significantly higher risks to both human and ecosystem health than the other two fractions. This study further revealed that the Cu and Zn contamination were the key contributors to freshwater, marine and terrestrial ecotoxicity, while Pb was the dominant contributor to human toxicity. Findings presented in this study provide a potential

guideline to help the steel mills make effective actions to control the key toxic metals present in the PM emissions, improving the local environmental quality and reducing the risks to public health.

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