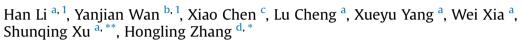
# Chemosphere 199 (2018) 702-708

Contents lists available at ScienceDirect

# Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

# A multiregional survey of nickel in outdoor air particulate matter in China: Implication for human exposure



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# HIGHLIGHTS

# G R A P H I C A L A B S T R A C T

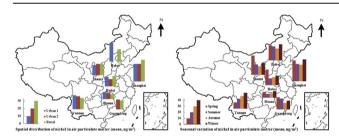
- We analyzed the Ni levels of 662 outdoor air samples from 7 provinces in China.
- The concentrations of Ni in air were in the range of 2.1–80.9 ng/m<sup>3</sup>.
- In most areas, the Ni concentrations were higher in winter and spring.
- Estimated daily intake doses of Ni through inhalation were calculated.
- This is a multiregional survey of Ni in outdoor air particulate matter in China.

# ARTICLE INFO

Article history: Received 16 October 2017 Received in revised form 16 January 2018 Accepted 22 January 2018

Handling Editor: Dr. R Ebinghaus

*Keywords:* Nickel Particulate matter Human exposure Outdoor air



# ABSTRACT

Nickel is a widespread environmental contaminant, and it is toxic to humans in certain forms at high doses. Despite this, nationwide data on nickel in outdoor air particulate matter and human exposure to nickel through inhalation in China are limited. In the present study, 662 outdoor air samples from seven representative provinces such as Shanghai, Hubei, Hunan, Hebei, Guangdong, Yunnan, and Shanxi were collected between March 2013 and February 2014 and analyzed by inductively coupled plasma mass spectrometry. The concentrations of nickel in the air were in the range of 2.1–80.9 ng/m<sup>3</sup> (geometric mean: 14.4 ng/m<sup>3</sup>). In most areas, the concentrations of nickel were higher in winter and spring than those measured in summer and autumn. The daily intake (median) of nickel through inhalation of air particulate matter was estimated. Although the nickel concentrations in some air samples were high, inhalation of the air particulate matter accounted for a minor part of the total nickel intake; however, the adverse effects of human exposure to nickel through inhalation and its potential sources require more attention, particularly in Shanghai. This is a multiregional survey of nickel in outdoor air particulate matter in China.

# 1. Introduction

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https://doi.org/10.1016/j.chemosphere.2018.01.114 0045-6535/© 2018 Elsevier Ltd. All rights reserved. Nickel (Ni) is a metallic element, and it exists in the earth's crust. In the 20th century, nickel exposure significantly increased owing to the wide range of applications of nickel (Michalak et al., 2012).



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Nickel and its compounds are used heavily in the manufacturing of stainless steel, nickel-cadmium batteries, jewelry, and medical prostheses (Cempel and Nikel, 2006), which result in increased levels of human exposure to nickel through environmental contamination (Merian, 1984). A study reported that nickel could be transported to a long-range through fine aerosol (Cong et al., 2010). Environmental exposure to nickel through anthropogenic origin occurs from traffic (Buekers et al., 2015), industrial activities (Huang et al., 2012) (stainless steel industry, nickel plating, alloy manufacturing, *etc.*), and the emissions from metal mining, smelting, and refining operations (Cempel and Nikel, 2006).

In rodents, the inhaled nickel substances are toxic to the lung, inducing adverse effects such as fibrosis, lung inflammation, proteinosis, and lung tumors (Dunnick et al., 1995; Oller et al., 2008). Nickel was also reported to be associated with disorders of the skin and kidney and of the cardiovascular and respiratory systems (Shi, 1994; Denkhaus and Salnikow, 2002; Grimsrud et al., 2002). On the basis of data from nickel workers and laboratory animals, most nickel compounds have been classified as human carcinogens (IARC, 2012). Oxides and the soluble forms of nickel were predominant in ambient air (Buekers et al., 2015). Because nickel is a human genotoxic carcinogen, the European Union (EU) recently set a target value for its concentration in ambient air as 20 ng/m<sup>3</sup> (Communities, 2001).

It is of considerable interest to assess human exposure to nickel for public health. Thus far, nickel has been detected in ambient air (Santos and Fernandez-Olmo, 2016), water (May et al., 2001; Alam et al., 2008; Birke et al., 2010), soil and dust (Wang et al., 2010), and so on. The inhalation of nickel is a minor source in terms of total daily intake, but it is of concern because of its adverse effects (Munoz and Costa, 2012; Buekers et al., 2015). Previously, nickel was shown to have a strong association with increased mortality, especially in the cold season (Huang et al., 2012). In particular, nickel in particulate matter was considered as a very important component for the association between particulate matter and lung cancer based on recent European multicohort studies (Raaschou-Nielsen et al., 2016). However, the profile of atmospheric nickel exposure to the general population in many regions of China is unclear (Duan and Tan, 2013), although the atmospheric nickel concentration in polluted mining areas (Zhang et al., 2009), industrial areas (Wang et al., 2013; Lau et al., 2014), and several other cities had been reported in China before (Piao et al., 2007; Wang et al., 2013). It is important to figure out whether the general population in any region are at high risk of exposure to nickel through inhalation.

In this paper, a multiregional survey of nickel concentrations was conducted with 662 outdoor air samples from 19 districts of seven provinces in China. Human exposure to nickel through inhalation of air particulate matter was calculated on the basis of the concentrations measured in the air samples.

## 2. Materials and methods

#### 2.1. Reagents

Nitric acid (68%) and hydrogen peroxide (30%) were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd (Beijing, China). A standard for Ni (IV-ICPMS-71A) was purchased from Inorganic Ventures (Christiansburg, VA, USA). Milli-Q water (Barnstead International, Dubuque, IA, USA) was used for analysis.

# 2.2. Sample collection

From March 2013 to February 2014, 662 outdoor air particulate matter samples were collected from two (urban area in big city and rural area in Hebei, urban area in small city and rural area in

Guangdong) or three sites (urban areas in both big and small cities and rural area) in seven provinces in China, including Shanghai city [Jing'an district (n = 48), Yangpu district (n = 48), Jiading district (n = 45)]; Hubei province [Qiaokou district: Wuhan city (n = 20), Huangpi district: Wuhan city (n = 20), Xiaonan district: Xiaogan city (n = 20)]; Hunan province [Tianxin district: Changsha city (n = 20), Liuyang and Changsha city (n = 20), Mount Hengshan: Hengyang city (n = 41)]: Hebei province [Yuhua district: Shijiazhuang city (n = 30), Guangyang district: Langfang city (n = 30)]; Guangdong province [Boluo county: Huizhou city (n = 30), Luoding county: Yunfu city (n = 52)]; Yunnan province (Panlong district: Kunming city (n = 40), Longyang district: baoshan city (n = 45), Xiangyun county: Bai autonomous prefecture, Dali city (n = 60)]; and Shanxi province [Beilin district: Xi'an city (n = 33), Xingping and Xianyang city (n = 30), Xinji town: Nanzheng county, Hanzhong city (n = 30)]. In total, there are three sampling sites each in Shanghai, Hubei, Hunan, Yunnan, and Shanxi, while only two sampling sites in Hebei and Guangdong, respectively, as representative sites for the corresponding geographical regions. A tiered sampling approach was adopted for the collection of air samples. Sampling was conducted simultaneously between Wednesday and Thursday at different sampling sites in each province. The sampling frequency was twice to four times every month for different provinces.

The Laoying 2030 Intelligent Air/TSP comprehensive sampler (Qingdao Laoshan Institute of Applied Technology, Qingdao, China) was placed 1.6 m above ground level in the sampling sites of urban and rural residential areas with no major industrial activities within 5 km around. The air samples were collected on 90-mm Whatman quartz fiber filters (Whatman International Ltd., Maidstone, UK) for 24 h. The outdoor air particulate matter was collected and then kept individually in a polystyrene box (SF-90BOX; Beijing Safelab Ltd, Beijing, China). Information on the sample number, sampling date, the actual air volume of the sample, the weight of the filter before and after sampling was recorded at the same room temperature and relative humidity. Subsequently, the samples were transported at 4 °C to the laboratory for analysis.

# 2.3. Sample preparation and instrumental analysis

The extraction procedure is provided elsewhere (Chakraborty and Gupta, 2010). Briefly, the filter was subjected to acid digestion with 15 mL of 68% HNO<sub>3</sub> and 4.5 mL of 30% H<sub>2</sub>O<sub>2</sub>. The residual solution was diluted to 50 mL with 1% HNO<sub>3</sub> and filtered (Whatman no. 1,  $\emptyset = 0.45 \mu$ m; Whatman International Ltd., Maidstone, UK) for subsequent analysis with inductively coupled plasma mass spectrometry (ICP-MS) with Agilent 7700 (Agilent Technologies, Santa Clara, CA, USA).

Blanks, duplicates, and matrix spikes were performed for each batch of 25 samples as per the procedure. The analytical accuracy was also assessed using certified reference material (AK-QC208-1; Beijing Century Audiocodes Biological Technology Co. Ltd., Beijing, China). The limit of detection for Ni was 0.58 ng/m<sup>3</sup>. The recoveries of Ni were 86%–102%.

#### 2.4. Calculation of daily exposure doses

Estimated daily intake (EDI; ng/kg/day) doses of nickel through inhalation were calculated as shown in equation (1) (USEPA, 2011).

$$EDI = (C \times AIR \times OEF)/BW$$
(1)

where, C is the nickel concentration in air samples  $(ng/m^3)$ , AIR  $(m^3/day)$ , and BW is the body weight (kg) as described previously

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