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An extended study on historical mercury accumulation in lake sediment of Shanghai: The contribution of socioeconomic driver

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ABSTRACT

Rapid industrialization and urbanization has caused large emission and potential contamination of mercury (Hg) in urban environment. However, little is known about the impact of socioeconomic factor on Hg accumulation in sediment. In the present study, historical record of anthropogenic Hg deposition of Shanghai was reconstructed by using three sediment cores from three park lakes (C1: Luxun Park; C2: Fuxing island Park; C3: Xinjiangwan Park). Meanwhile, the influence of socioeconomic factor to Hg emissions and sedimentary record was calculated based on an extended STIRPAT (stochastic impacts by regression on population, affluence and technology) model. The profiles of Hg levels and fluxes in the three sediment cores showed that Shanghai has recently undergone urbanization. The anthropogenic Hg fluxes exhibited fluctuant increases from ~1900 to present and accelerated after the establishment of the People's Republic of China in 1949 and the implementation of reform and opening up policy in 1978. The mean flux ratios of Hg in post-2000 were 2.2, 12, and 2.7 in the C1, C2 and C3 cores, respectively. The extended STIRPAT model was constructed based on strong positive relationships between socioeconomic factors and Hg fluxes, revealing that the proportion of coal consumption, the urbanization rate, and the proportion of heavy industry were the three most important driving factors for Hg accumulations in urban sediment of Shanghai.

1. Introduction

With the rapid urbanization and industrialization, numerous human activities such as municipal, industrial, traffic, and commercial operations, load various hazardous pollutants and cause adverse health effects in residents worldwide (Li et al., 2012b; Pacyna and Pacyna, 2001; Shao et al., 2013; Zhu et al., 2011). For examples, urban agglomeration in China has caused negative impacts on ecological system (Qiu et al., 2015). Chronic diseases such as cardiovascular and metabolic diseases were more found in urban than in rural residents (Zhu et al., 2011). As a well-known priority pollutant, mercury (Hg), has been widely concerned in recent years due to its high volatility, toxicity, bioaccumulation, and long-range transportation in the global range (USEPA, 1997). Hg can be released during the high-temperature production of industrial goods, the combustion of fossil fuels, and the incineration of municipal and industrial wastes, subsequently entering the atmosphere and depositing into sediment (Pacyna and Pacyna, 2001; Wong et al., 2006).

To clarify the spatial and temporal trends of toxic pollutants and to explore the influences of intensive anthropogenic activities in urban areas, lake sediment from city parks has generally been used as a reliable field archive (Chillrud et al., 1999; Li et al., 2013; Ma et al., 2016). Lake sediment cores are often taken to reconstruct their pollution histories and potential sources. Several studies indicated that the levels and fluxes of Hg in sediment cores were closely associated with fossil fuel consumption, land use change, urbanization rate, economic level, as well as environmental policy in studied areas (Bookman et al., 2008; Engstrom et al., 2007; Kang et al., 2016; Li et al., 2013; Muir et al., 2009; Outridge et al., 2011). Moreover, the amounts of Hg releasing from various sources have been well calculated (Bookman et al., 2008; Li et al., 2013; Outridge

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et al., 2011), however, the driving forces of socioeconomic factors (e.g., gross domestic product, population, and policy) on its emissions and/or accumulations were seldom examined (Liang et al., 2013).

Socioeconomic factors are considered as the primary drivers of environmental stress (Fodha and Zaghdoud, 2010; Liang et al., 2013). A variety of methods, including decomposition analysis (DA), time series analysis, causal analysis, correlation analysis, and gray relative analysis, have been employed to analyze the influence of these factors on the emission of pollutants (Dietz et al., 2007; Fodha and Zaghdoud, 2010; Lin et al., 2007; Liu et al., 2012). The DA, including index DA and structural DA, was the most used (Liang et al., 2013). A model of index DA, the stochastic impacts by regression on population, affluence and technology (STIRPAT) model, was developed to determine the contribution of anthropogenic factors causing environmental pressure (Dietz et al., 2007; Fan et al., 2006; York et al., 2003). However, no implication of the STIRPAT model was found in assessing the impact of socioeconomic factor on Hg accumulation in sediment.

In our previous study, the distribution of Hg in a sediment core from the Luxun Park of Shanghai were examined, but no quantitative analysis for the contribution of socioeconomic factors was determined (Yang et al., 2015). Herein, employing STIRPAT model, we conducted an extended study on the pollution of Hg in sediment cores of Shanghai, with objectives to (i) further determine the temporal variation, (ii) reconstruct the pollution history, and (iii) evaluate the impact of relevant socioeconomic factor.

2. Materials and methods

2.1. Study area and sampling

Shanghai is one of the most developed cities of China, which has more than 150 years history of urbanization and industrialization. During the past several decades, Shanghai has undergone urban land expansion, population explosion, economic growth, industrial output transformation, and urban infrastructure improvement, gradually evolving from a small fishing village into the largest economic center and industrial base in China. So far, Shanghai has become a hyper-urbanization and international city with an urbanization rate of ~90%. Hongkou District and Yangpu District, located at the central urban areas in Shanghai, are two of the earliest and the most concentrated districts of industry. Therefore, this study chose the Hongkou District and Yangpu District as studied areas to explore the pollution history of anthropogenic Hg during the urbanization of Shanghai.

Three sediment cores were collected from closed lakes in parks at the Hongkou District (C1) and the Yangpu District (C2 and C3), where few in-situ anthropogenic perturbations (e.g., dredging activity) occurred (Fig. 1). For the three studied lakes, no obvious inflows and outflows existed with surface areas of 3.4, 0.4, and 2.1 ha, respectively. The mean depth are 2.0, 1.8, and 2.5 m and the watershed/lake area ratios are 7.41, 9.48, and 8.76, respectively. The sampling campaign was conducted in September 2012. Using a gravity corer with a 6-cm internal diameter core barrel (XDB0205, Beijing Xindibiao Lab Instrument Co. Ltd., China), the sediment cores were taken from the middle of the lakes with lengths of 34, 19, and 17 cm for C1, C2, and C3, respectively. After collection, the samples were transported to laboratory immediately, sliced at 2-cm interval for C1 and 1-cm interval for C2 and C3, and kept at −20 °C until analysis.

2.2. Hg analysis and age dating

Sediments were freeze-dried, homogenized, and ground to pass a 200-mesh nylon sieve. Hg was analyzed by a DMA-80 direct Hg analyzer (Milestone, Shelton, CT), which involves the serial processes of thermal composition, catalytic reduction, amalgamation, desorption, and atomic absorption spectroscopy (Oh et al., 2010). Meanwhile, two standard reference materials (SRMs; soil ESS1 and ESS3) and a procedural blank were analyzed for every 10 samples. By comparing with the certified values in soil ESS1 (16 ± 3 μg/kg) and ESS3 (112 ± 12 μg/kg), the recoveries of Hg in 10 replicates ranged from 87% to 99% with the precision < 20%. In addition, the relative standard deviation for replicate samples was less than 15%. The detection limit of Hg in sediment was 0.0005 ng. Hg levels in all blank samples were below 0.02 μg/kg dry weight (dw). All Hg concentrations were corrected by subtracting the blank value.

Three sediment cores were aged by using the ^{210}Pb and/or ^{137}Cs method, which was described in details elsewhere (Yang et al., 2015). Briefly, the activities of ^{226}Ra, ^{210}Pb, and ^{137}Cs in sediment were measured by using a multichannel gamma spectrometer with a low background intrinsic germanium detector (ORTEC HPGe GWL series). Then those data were input in a constant rate of supply (CRS) model to estimate the sedimentation rate and age (Appleby, 2001). To improve the accuracy of sedimentation age determined by ^{210}Pb method, the ^{137}Cs stratigraphy was also analyzed for calibration, based on four peaks of ^{137}Cs activities in an ideal sediment core that were related to nuclear explosion events happened in 1954, 1963, 1974, and 1986, respectively (Grosbois et al., 2012).

2.3. Modeling methodology

To estimate environmental impact (I), described by the equation IPAT (Impact = Population × Affluence × Technology), the STIRPAT model was employed; the equation is shown as the following stochastic version (York et al., 2003).

$$I = a \times P^b \times A^c \times T^d \times e \quad (1)$$

Where I represents the deposition of Hg in sediment core per unit area in each year (t); b, c, and d are the elasticity of population (P), affluence (A), and technology (T), respectively − these parameters can be acquired by regression fitting with the least squares method. a is a constant scaling the model and e is an error term representing all variables not included in the model.

To fully analyze the impact of the driving factors on Hg deposition, the decomposed determinants (P_1, P_2, A_1, A_2, T_1, and T_2) were further introduced according to the above model. The extended STIRPAT model was listed as follows.

$$I = a \times P_1^{b_1} \times P_2^{b_2} \times A_1^{c_1} \times A_2^{c_2} \times T_1^{d_1} \times T_2^{d_2} \times e \quad (2)$$

Where P_1 and P_2 refer to the total population (10^4 person) and the percentage of urban population (urbanization rate, %), respectively; A_1 and A_2 are the per capita gross domestic production (GDP; Yuan per capita) and the proportion of heavy industry in industrial output (%), respectively; T_1 and T_2 are the energy consumption per unit GDP (t/Yuan) and the proportion of coal consumption in primary energy structure (%), respectively; the elasticity coefficients b_1, b_2, c_1, c_2, d_1, and d_2 are interpreted as the variation of Hg deposition for 1% change in P_1, P_2, A_1, A_2, T_1, and T_2, respectively.

2.4. Data sources

Two categories of data were required to support the extended STIRPAT model, including Hg fluxes in three sediment cores and six urbanization parameters of different years. The total population, population in urban areas, GDP, output value of heavy industry,