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Trans-boundary movement of mercury in the Northeast Asian region predicted by CAMQ-Hg from anthropogenic emissions distribution

Jin-Ho Sung[a], Debananda Roy[a], Joo-Sung Oh[a], Seung-Ki Back[a], Ha-Na Jang[a], Seong-Heon Kim[a], Yong-Chil Seo[a,*], Jeong-Hun Kim[b], Chong Bum Lee[c], Young-Ji Han[c]

[a] Dept. of Environmental Engineering, Yonsei University, Wonju 26493, Republic of Korea
[b] Division of Air Pollution Engineering, National Institute of Environmental Research, Incheon 22689, Republic of Korea
[c] Dept. of Environmental Science, College of Agriculture and Life Sciences, Kangwon National University, Chuncheon 24341, Republic of Korea

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ABSTRACT

The percentage contribution of trans-boundary mercury (Hg) from China at different locations in South Korea was estimated from Hg anthropogenic emission distributions using the Hg dispersion model, CMAQ-Hg. This investigation quantifies the trans-boundary Hg emissions as contribution ratios. In addition, the long-range transportation frequency is also calculated, to verify inflow cases from China. The seasonal distribution of the Hg contribution ratio was found to be highest in winter (40%), followed by fall (16%). Seasonal observations of Hg inflow frequencies were estimated as 40%, 25%, 21%, and 4% in winter, fall, summer, and spring, respectively, at the same location. Such results would be produced by the wind generally blowing from the west and northwest with a speed of 5.0 m/s and 4.5 m/s, respectively, during winter and fall, around the study area. This study made an effort to quantify the trans-boundary Hg transport and to plot Hg anthropogenic emissions distribution in the region.

1. Introduction

Global anthropogenic Hg emissions and their long-range transportation is the subject of growing concern today. Hg is recognized as one of the toxic pollutants, due to its neurotoxicity and long-range transportability, with a life time of 0.6–1.5 years in the atmosphere (Slemr et al., 1985; Pan and Carmichael, 2005). Atmospheric Hg exists in the forms of elemental mercury (Hg^0), oxidized mercury (Hg^{2+}) and particulate mercury (Hg_p) with different lifetimes. Hg^{2+} is more reactive and readily scavenged through wet and dry deposition. The Hg_p takes several days or weeks to deposit (Qin et al., 2016). According to the Hg emission inventory developed by Pacyna et al. (2003), more than 50% of the total global Hg emissions of 200 tons $year^{-1}$ comes from Asia (Pacyna et al., 2003). It was also reported that China is responsible for 30% of global anthropogenic Hg emissions (Pacyna et al., 2006). As per the Minamata convention, the major anthropogenic sources of Hg are coal-fired power plants, non-ferrous metal smelting facilities, cement clinker production facilities, and waste incineration facilities. The rebound Hg was estimated as Hg^0 speciation only. The existing air pollution control devices (APCDs) are able to remove some parts of Hg^{2+} and Hg_p from flue gases along with SOx, NOx, and PM (Pudasainee et al., 2017). The removal efficiency can be improved by using wet types of APCDs (Lee et al., 2006). As a result, flue gas is a richer source of Hg^0 in the atmosphere than of other species of Hg (Yang et al., 2007; Sjostrom et al., 2010). The Hg^0 acts as a trans-boundary element due to its chemically inert nature. The Hg^0 is not only dissipated but it can also be transported over long distances due to its high retention time in the atmosphere and Hg^{2+} is tends to deposit near the source. For this reason, it must be managed from a global perspective (Selin, 2009). Atmospheric Hg emitted from China could be a major source of Hg levels in South Korea due to transportation and dispersion. The Hg inflow pathway from different sources to a receptor has been the focus of various studies (Huang et al., 2016). Marumoto et al. (2015) verified the Hg movement pathway by conducting receptor modeling with the other air pollutants. Nguyen et al. (2010) and Choi et al. (2014) revealed that trans-boundary pollutants like Hg are transported from China to Korea via westerly winds, due to the physiographic features of Korea (Nguyen et al., 2010; Choi et al., 2014).

Trace amounts of Hg can be accumulated in the ecosystem due to inflows from the external environment and circumjacent sources (Marson et al., 2012). This atmospheric Hg is accumulated in water bodies due to precipitation (Seo et al., 2012). Sea foods and foods from fresh water (rivers and lakes) are very common in Korean meals. Consequently, mercury concentration in the blood of Korean people is

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* Corresponding author at: Dept. of Environmental Engineering, Yonsei University, Wonju 26493, Republic of Korea.
E-mail address: seoyc@yonsei.ac.kr (Y.-C. Seo).

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found to be higher than in other countries (Kim et al., 2014). According to Kim et al. (2010), Hg emissions from anthropogenic sources in Korea are estimated to average 12.8 tons in 2007 (Kim et al., 2010). The anthropogenic sources of Hg emissions are found to be higher in the suburbs of the big cities of South Korea (Kim et al., 2010; Pudasainee et al., 2014).

The atmospheric dispersion and transportation of trans-boundary mercury can be calculated using the CMAQ model (Byun and Schere, 2006). CMAQ is a three-dimensional photochemical model, based on the Eulerian dispersion model. The CMAQ model includes 36 chemicals, 93 chemical reactions, and 11 photochemical reactions, using CBN mechanisms. In addition, the four gas-phase reactions with four (O_3, Cl_2, H_2O_2 and OH) chemical species are considered in the CMAQ-Hg model (Bullock and Brehem, 2002). Due to these features, it can be used for modeling the atmospheric dispersion of ozone, dust, and air pollutants, because it uses a meteorological model based on a lattice system. Also, the modeling scale can be controlled, from city-scale to planetary-scale, (Bullock and Brehem, 2002). Very recently, the research groups of various nations have adopted this technique for atmospheric pollution research (Oh et al., 2008; Lee et al., 2011; Wang et al., 2015; Sharma et al., 2016; Wang et al., 2016; Bieser et al., 2017). Also, Baker and Bash (2012) used this model to study Hg dispersion in the atmosphere (Baker and Bash, 2012). Also, Wang et al. (2016) simulated atmospheric Hg level in China and the East Asia region using this model (Wang et al., 2016).

In this study, trans-boundary Hg was quantified by the output of the CMAQ-Hg model for the period of March 2008 to February 2009. This model calculates the Hg emissions from anthropogenic sources in Korea and China using a gridding system. Meteorological parameters are generally regarded as the important factors for the transport and distribution of pollutants (Dodla et al., 2013; Zhang et al., 2016). Meteorological data for the study area during the study period was collected from the Korea Meteorological Administration (KMA). The Mesoscale Model, version 5 (MM5) was applied to simulate the meteorological data for dispersion modeling. In this way, seasonal distributions of atmospheric Hg around Korea have been estimated as a contribution ratio. The seasonal contributions of Hg inflow from China were then assessed by deducting the background Hg concentrations from the actual Hg concentration in the study area. Seven locations were selected to estimate the trans-boundary mercury in South Korea. The monitoring stations are established at seven cities in South Korea: Seoul (A), Ganghwa (B), Chuncheon (C), Daejeon (D), Gwangju (E), Daehu (F) and Busan (G). These are shown in Fig. 1, and a detailed description of the study area is given in Table 1.

The exhaustive literature study reveals that quantification of trans-boundary Hg and its inflow from China are a new matter of concern today. This study gives the first insights into (1) the percentage contribution of trans-boundary Hg from China to South Korea, and (2) the long-range transportation frequency and inflow cases from China to Korea, using the CMAQ-Hg model.

2. Methods

2.1. Estimation of mercury emissions

Atmospheric Hg emissions can be subdivided into domestic and trans-boundary emissions. The dispersion of Hg from China was calculated by estimating the anthropogenic Hg emissions in Korea. Total Hg emissions could be calculated by Eq. (1) (Wu et al., 2016).

$$E_t = \sum (10^{-9} \times ef_1 \times A_{1,t}) \tag{1}$$

where, ‘E’ denotes Hg emissions, ‘ef’ represents the emission factor, ‘A’ stands for the activity, ‘l’ is the index of the emission sector, and subscript ‘t’ is time in years. The Hg emissions were calculated using the activities and emission factors listed in Table 2. Emission factors were taken from previous studies carried out in Korea during the same study period (Liu et al., 2016; Tartakovsky et al., 2016; KCA, 2008; KEPCO, 2008; KMOHW, 2008; KMOE, 2009a, 2009b, 2009c; PPS, 2008a, 2008b; KEEI, 2008; KPA, 2008; Kim et al., 2010). From these reports on emissions of Hg, a gridded Hg emissions distribution for Korea was drawn up. A gridded Hg emissions distribution for China was taken from a report published by the United Nations Environmental Program (UNEP, 2008).

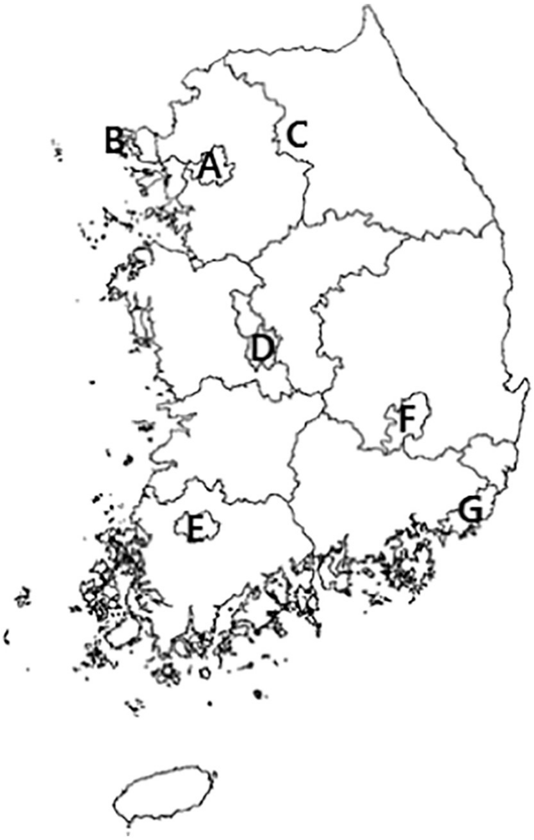

Fig. 1. Locations of Hg monitoring stations in South Korea.

Table 1
Details of locations in the study area.

Location	Latitude	Longitude	Location characteristics
Seoul (A)	37.579640	126.998894	The highest population and traffic density.
Ganghwa (B)	37.793088	126.415631	The nearest to China.
Chuncheon (C)	37.782912	127.609939	Identification of west wind effect.
Daejeon (D)	36.373430	127.443114	Located inland, with a continental climate. The nearest to the background station in South Korea.
Gwangju (E)	35.143436	126.911735	The nearest to an industrial complex. Identification of southwest wind effect.
Daegu (F)	35.849503	128.638530	Surrounded by mountains in all directions.
Busan (G)	35.175363	129.044796	The highest population and traffic density in the southern region. Identification of south wind effect.

2.2. Dispersion modeling

For this study, the Mesoscale Model, version 5 (MM5) and the CMAQ model (version 4.6) (Dodla et al., 2013; Bullock and Brehem,