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## Assessing the effect of long-range pollutant transportation on air quality in Seoul using the conditional potential source contribution function method



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#### HIGHLIGHTS

• The long-range transports of pollutants to Seoul were analyzed from 2001 to 2014.

• Long-range transport increase SO<sub>2</sub>, NO<sub>2</sub>, CO, PM<sub>10</sub> concentration by 14%, 9%, 6%, 21%.

• Transports from near Beijing and Harbin are becoming more significant recently.

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#### ABSTRACT

It is important to estimate the effects of the long-range transport of atmospheric pollutants for efficient and effective strategies to control air quality. In this study, the contributions of trans-boundary transport to the mean concentrations of SO<sub>2</sub>, NO<sub>2</sub>, CO, and PM<sub>10</sub> in Seoul, Korea from 2001 to 2014 were estimated based on the conditional potential source contribution function (CPSCF) method. Eastern China was found to be the major source of trans-boundary pollution in Seoul, but moderate sources were also located in northeastern China. The contribution of long-range transport from Japan was negligible. The spatial distributions of the potential source contribution function (PSCF) values of each pollutant showed reasonable consistency with their emission inventory and satellite products. The PSCF values of SO<sub>2</sub> and PM<sub>10</sub> from eastern China were higher than those of NO<sub>2</sub> and CO. The mean concentrations of SO<sub>2</sub>, NO<sub>2</sub>, CO, and PM<sub>10</sub> in Seoul for the period from 2001 to 2014 were 5.34, 37.0, and 619.1 ppb, and 57.4 4  $\mu$ g/m<sup>3</sup>, respectively. The contributions of long-range transport to the mean concentrations of SO<sub>2</sub>, NO<sub>2</sub>, CO, and  $PM_{10}$  in Seoul were 0.74, 3.4, and 39.0 ppb, and 12.1  $\mu$ g/m<sup>3</sup>, respectively, which are 14%, 9%, 6%, and 21% of the mean concentrations, respectively. The annual mean concentrations of SO<sub>2</sub> and NO<sub>2</sub> followed statistically significant increasing linear trends (0.5 and 1.6 ppb per decade, respectively), whereas the trends in the annual mean concentrations of CO and PM<sub>10</sub> were statistically insignificant. The trends in the ratio of the increased concentrations associated with long-range transport to the annual mean concentrations of the pollutants were statistically insignificant. However, the results indicate that the trans-boundary transport of SO<sub>2</sub>, NO<sub>2</sub>, CO, and PM<sub>10</sub> from eastern China consistently affected air quality in Seoul over the study period (2001–2014). Regionally, the effects of the long-range transport of pollutants from Beijing and Harbin-Changchun on air quality in Seoul have become more significant over this period.

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#### 1. Introduction

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http://dx.doi.org/10.1016/j.atmosenv.2016.11.017 1352-2310/© 2016 Elsevier Ltd. All rights reserved. Tropospheric aerosols and trace gases emitted from anthropogenic and natural sources affect both visibility and the radiative

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balance of the atmosphere (Myhre et al., 2013; Ramanathan et al., 2001; Watson, 2002). Short- and long-term exposure to air pollution can have adverse effects on human health, including both acute and chronic effects on a number of different systems and organs. Air pollution is thus linked with premature mortality and reduced life expectancy (Brauer et al., 2016; Kampa and Castanas, 2008; Pope et al., 2009). In particular, respiratory diseases are strongly associated with aerosols (Dockery et al., 1996; Pope et al., 2002), and asthma morbidity is exacerbated by air pollution (Weiss et al., 1993). Previous studies have estimated the relative risk of asthma from atmospheric pollutants including SO<sub>2</sub>, NO<sub>2</sub>, CO, and particulate matter of  $< 10 \,\mu\text{m}$  in diameter PM<sub>10</sub>; (Sunver et al., 1997; Thompson et al., 2001). Additionally, Huang et al. (2009) found that decrease in visibility by 8 km corresponds to 2.17%, 3.36%, and 3.02% increases in total, cardiovascular, and respiratory mortality, respectively, in Shanghai, which is associated with increases in SO<sub>2</sub>, NO<sub>2</sub>, and PM<sub>10</sub>. The Organization for Economic Co-Operation and Development (OECD) Environmental Outlook Baseline scenario projected that urban air quality will continue to deteriorate globally if no new policies are implemented (OECD, 2012). By 2050, outdoor air pollution (particulate matter and ground-level ozone) is projected to be the main cause of environmentally related deaths worldwide (OECD, 2012).

Air pollution has been reduced over recent decades in developed countries through the implementation of effective regulations and strategies (Guerreiro et al., 2014; USEPA, 2012). However, large amounts of air pollutants are emitted from East Asia, which are estimated to contribute 36%, 29%, and 36% of global emissions of SO<sub>2</sub>, NO<sub>x</sub>, and particulate matter of  $<2.5 \mu m$  (PM<sub>2.5</sub>), respectively (Cofala et al., 2012; Wang et al., 2014). Furthermore, emissions of NO<sub>x</sub> and non-methane volatile organic compounds (NMVOC) increased by 25% and 15%, respectively, from 2005 to 2010 due to inadequate control strategies in China (Wang et al., 2014). Meanwhile, emissions of these pollutants in other regions of East Asia decreased by between 13% and 17%, mainly because of stringent vehicle emission standards being implemented in South Korea and Japan (Wang et al., 2014). Furthermore, emissions of  $SO_2$  and  $PM_{2.5}$ in East Asia are estimated to have decreased by 15% and 12%, respectively, between 2005 and 2010 (Wang et al., 2014). Such spatial and temporal variability in emissions of air pollutants results in further complications in control strategies within East Asia because of the trans-boundary transportation of aerosols and trace gases (Fast et al., 2014; Lin et al., 2007; Schmidt et al., 2015; Tu et al., 2004; Zhao et al., 2007; Zien et al., 2014).

Previous studies have suggested various methods to estimate the contribution of long-range transported pollutants from source areas to downwind sites. Chemistry transport models (CTM) have improved our understanding of the processes associated with the long-range transport of emissions and their contribution to downwind areas (Fast et al., 2014; Jeong et al., 2011a; Schichtel et al., 2006). Methods of classifying air mass pathways based on backward trajectory calculations or wind direction have also been widely used; these methods are simple and use accurate air quality measurements at downwind sites (Fu et al., 2010; He et al., 2003; Kim, 2008; Kim et al., 2005, 2006a; Meng et al., 2007). Satellite measurements have provided valuable information on spatial variations in air quality and can be used to monitor long-range transport over regional to global scales (Kim et al., 2007; Kallos et al., 2007; Rajeev et al., 2000; Schmidt et al., 2015; van Donkelaar et al., 2008; Zhao et al., 2007; Zien et al., 2014). In addition, satellite measurements are important for the validation of, and data assimilation for, CTMs if we are to predict long-range transport and air quality (Adhikary et al., 2008; Bocquet et al., 2015; Elbern et al., 1997; Park et al., 2014; Saide et al., 2014). Hybrid receptor models that combine measurements and backward trajectory calculations have been widely used to find the potential source areas and preferred transport pathways of air pollutants (Ashbaugh et al., 1985; Bressi et al., 2014; Cheng et al., 1993; Heo et al., 2009; Hwang and Hopke, 2007; Polissar et al., 2001).

However, there are limitations to the methods described above. Simulations of the long-range transport of atmospheric pollution using CTMs have a large dependency on emission inventories (Ma and van Aardenne, 2004). In addition, CTMs typically have a large uncertainty in physical parameterizations, including turbulence closure and chemical mechanisms (Mallet and Sportisse, 2006). Satellite measurements typically represent total vertical column amounts of atmospheric pollution and can be affected by uncertainties in surface concentrations (Seo et al., 2015; Wang and Christopher, 2003; Xu et al., 2015).

Hybrid receptor models, such as the potential source contribution function (PSCF) and concentration weighted trajectory (CWT) models, also have uncertainties in measurements and backward trajectory calculations (Stohl, 1998). However, the uncertainties of in situ measurements are typically less significant for the analysis than other methods. Additionally, hybrid receptor models do not require information regarding chemical mechanisms or emission inventories. As the models use measurements from downwind sites, the target pollutants by this time have already been subjected to chemical processes during transport (Jeong et al., 2013). The conditional PSCF (CPSCF) method provides information on the quantitative significance of long-range transport from potential source areas by applying the traditional PSCF method (Jeong et al., 2011b. 2013). Jeong et al. (2011b) focused on the aerosols during one year and leong et al. (2013) analyzed the effects long-range transport of SO<sub>2</sub> in Seoul during the period from 2001 to 2010 using the CPSCF method, which were limited in terms of analysis period and species.

Seoul is one of the largest megacities in northeast Asia, with a population of more than 20 million (including neighboring areas). Air quality in Seoul is affected by both local emissions and the long-range transport of pollutants. Quantitative estimation of the contribution of long-range transport is essential if we are to develop effective strategies to control air quality in Seoul (Heo et al., 2009; Jeong et al., 2011b, 2013). In this study, the contributions of long-range transport to the annual mean concentrations of NO<sub>2</sub>, SO<sub>2</sub>, CO, and PM<sub>10</sub> in Seoul, South Korea (37°57′N, 126°98′E) were estimated between 2001 and 2014 using the CPSCF method. Furthermore, the spatial distributions of the calculated PSCF values are compared with satellite inversion products.

### 2. Data

The National Institute of Environmental Research (NIER) of the Korea Ministry of Environment (KME) measures SO<sub>2</sub>, NO<sub>2</sub>, CO, and PM<sub>10</sub> hourly over 40 sites in Seoul (Fig. 1), and data from 2001 to 2014 were used in this study. Hourly data from all 40 sites were averaged and used in the CPSCF analysis. SO2 samples were measured by ultraviolet (U.V.) fluorescence using sulfur dioxide analyzers (model 4108, Dasibi Environmental Corp., Glendale, CA; US Environmental Protection Agency reference method EQSA-1086-061) with a lower detection limit of 0.6 ppb.  $NO_2$  was measured by chemiluminescence using oxides of nitrogen analyzers (model 2108, Dasibi Environmental Corp.; US Environmental Protection Agency reference method RFNA-1192-089) with a lower detection limit of 2 ppb. CO mixing ratios were measured by a nondispersive method using carbon monoxide analyzers (model 3008, Dasibi Environmental Corp.; US Environmental Protection Agency reference method RFCA-0488-067) with a lower detection limit of 0.1 ppm. The mass concentrations of PM<sub>10</sub> were measured by  $\beta$ -ray

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