A new conceptual model for quantifying transboundary contribution of atmospheric pollutants in the East Asian Pacific rim region

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A B S T R A C T

Transboundary transport of air pollution is a serious environmental concern as pollutant affects both human health and the environment. Many numerical approaches have been utilized to quantify the amounts of pollutants transported to receptor regions, based on emission inventories from possible source regions. However, sparse temporal–spatial observational data and uncertainty in emission inventories might make the transboundary transport contribution difficult to estimate. This study presents a conceptual quantitative approach that uses transport pathway classification in combination with curve fitting models to simulate an air pollutant concentration baseline for pollution background concentrations. This approach is used to investigate the transboundary transport contribution of atmospheric pollutants to a metropolitan area in the East Asian Pacific rim region. Trajectory analysis categorized pollution sources for the study area into three regions: East Asia, Southeast Asia, and Taiwan cities. The occurrence frequency and transboundary contribution results suggest the predominant source region is the East Asian continent. This study also presents an application to evaluate heavy pollution cases for health concerns. This new baseline construction model provides a useful tool for the study of the contribution of transboundary pollution delivered to receptors, especially for areas deficient in emission inventories and regulatory monitoring data for harmful air pollutants.

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1. Introduction

The adverse impacts of air pollutants (e.g., particulate matter (PM), ozone, and persistent organic pollutants) on human health and the environment are well recognized (Brunekreef and Holgate, 2002; Krewski et al., 2009; WHO, 2003, 2006). Their effects are not limited to emission sources in the nearby area only, as atmospheric pollutants can be transported from their source regions to different continents (receptor regions), even on a global scale (Akimoto, 2003; Jaffe et al., 1999; Lelieveld et al., 2002; Wilkening et al., 2000; Zhang et al., 2008).

For PM pollution, the particle concentration, surface area, and composition (e.g., organic and inorganic compounds) are important factors that mediate the effects of PM on human health (Daigle et al., 2003; Lighty et al., 2000). Many studies indicate that PM pollution is associated with cardiovascular mortality and respiratory diseases (Kaiser, 2005; Krewski et al., 2009; Laden et al., 2006; Pope et al., 2002, 2004), and can even affect the central nervous system (Block and Calderón-Garcidueñas, 2009; Calderón-Garcidueñas et al., 2008; Genc

et al., 2012; Kim et al., 2010). Exposure to fine particles, defined by aerodynamic diameter < 2.5 μm (PM2.5), has been demonstrated to have a particularly high risk of daily mortality and a long-term risk of death (Kaiser, 2005; Krewski et al., 2009; Laden et al., 2000). These particles can also be transported over longer distances when compared with coarse particles (e.g., PM10) (Jeong et al., 2011; Lin et al., 2005; Nam et al., 2009). The toxicity of particulates also varies depending on their chemical composition. For instance, the adsorption of some carcinogenic polycyclic aromatic hydrocarbons (PAHs) by PM may increase the health risks of PM. Particle-bound PAHs may result in health effects with more severity than are observed with PM alone (Grimmer, 1983; IARC, 1984; WHO, 2006).

Modeling approaches have been widely used for the study of air pollutant transboundary transport patterns (Chin et al., 2007; Mauzerall et al., 2000; Wagstrom and Pandis, 2011; Wang et al., 2009; Zhang et al., 2008; Zhang et al., 2011) and for investigating the effects of transboundary transport on air quality in receptor regions (Huebert et al., 2003; Kajino et al., 2013; Lin et al., 2005; Lin et al., 2007; Naja and Akimoto, 2004; Primbs et al., 2007; Yang et al., 2007), particularly the East Asian region. These studies have provided valuable knowledge for understanding transport patterns and the atmospheric pollutant impacts on potential receptors. However, the accuracy of input

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parameters, model selection, and the transboundary contribution algorithms might increase either the burden of simulation work or the uncertainty of the output.

Over the past few decades, the rapid economic growth and urbanization of East Asia has led to unprecedented atmospheric pollution. Some studies have indicated that emission inventory uncertainty and sparse observation data in the East Asian continent make the understanding of the relationship between the sources and the receptors—and therefore the quantification of the transboundary transport delivered to the receptors—a significant challenge (Chen et al., 2013; He et al., 2011; Wang et al., 2011; Zheng et al., 2009). An alternative method could be useful for quantifying atmospheric pollution imported from neighboring countries for those areas lacking emission inventories or regulatory monitoring of harmful pollutants.

Taiwan is the primary receptor located in the western Pacific region, which is a critical area for studying the impacts of air pollution exported from the East Asian continent. The objectives of this study are therefore to determine the seasonal variation of atmospheric pollutants for a metropolitan area in southwestern Taiwan and to construct an alternative method for quantifying the transboundary transport contribution of particulate pollutants in a metropolitan area by using back trajectory analysis to select baseline database from two-year monitoring data sets of particulate matter (PM10 and PM2.5) and PAH data.

2. Methodology

2.1. Particulate matter data

The present study investigated the transboundary transport contribution of PM for the study area using hourly particulate matter (PM10 and PM2.5) concentration data from the Qianzhen station. This is the closest air quality monitoring station to the PAH sampling site (Fig. 1) in the Taiwan Air Quality Monitoring Network (TAQMN). The PM data quality control is based on the Taiwan EPA Data Quality Assurance Procedures (DQAP), which follow those prescribed by the U.S. Environmental Protection Agency, established in 1991. The DQAP covers regular system maintenance (e.g., daily zero and span checks, biweekly precision checks, and monthly instrument function checks) and data performance auditing (Taiwan EPA, 2014). In this study, a valid PM daily concentration was defined as the average of at least sixteen hours of effective data within a 24-hour collection to provide a better representative daily average. The data were collected for the study period of May

2007 to April 2009. Hourly meteorological data, including air temperature, rainfall, wind speed, and wind direction, were collected from the monitoring station of the Central Weather Bureau.

2.2. PAH data

Particulate PAH data were collected at the university campus (22°37′N, 120°15′E, Fig. 1) between May 2007 and April 2009 [these data sets were also used in (Lai et al., 2011, 2013)]. The data of sixteen PAHs designated as priority pollutants by the US Environmental Protection Agency (U.S. EPA, 1982), as well as BaP data, were selected for estimating the transboundary transport contribution. Detailed methods of sampling processes, equipment pre-treatment procedures, and sample extraction, preparation, and analysis procedures were published previously (Lai et al., 2011). The PAH data quality control was ensured by performing the measurements of field and laboratory blanks simultaneously. In addition, four perdeuterated PAHs (naphthalene-d_8, fluorene-d_{10}, fluoranthene-d_{10}, and perylene-d_{12}) were utilized as surrogates for the determination of the efficiency of the sample extraction and analysis. Two-year observation samples were analyzed separately, with mean recoveries of $57.1 \pm 15.2\%$ and $56.4 \pm 11.9\%$ for naphthalene-d_8, $69.8 \pm 14.0\%$ and $77.1 \pm 16.6\%$ for fluorene-d_{10}, $73.2 \pm 17.4\%$ and $87.2 \pm 25.5\%$ for fluoranthene-d_{10}, and $78.3 \pm 10.5\%$ and $92.0 \pm 19.3\%$ for perylene-d_{12} for the period of May 2007–April 2008 and May 2008–April 2009, respectively.

Sampling limitations resulted in only 52 valid data measurements in the two-year collection. Given the evaluation of the transboundary PAH transport contribution, a multiple linear regression analysis (MLR) was utilized to simulate the concentrations of PAHs for the study period. Based on the results of the statistical significance (see Table S1) and the ability to predict concentrations, the parameters of PM10, temperature, and relative humidity were selected for multiple regression analyses of the 16 PAHs and BaP. The equation of the multiple regression is:

$$y = \alpha + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 \tag{1}$$

where x is the independent variable (the values of three parameters: PM10, temperature, and relative humidity), y is the value of the dependent variable for values x of the independent variables, and α, β_1, β_2, and β_3 are regression coefficients. The comparison of the simulated and measured PAH concentrations (sixteen PAHs and BaP) is shown in Fig. S1. In general, the simulated PAH concentrations, for both the
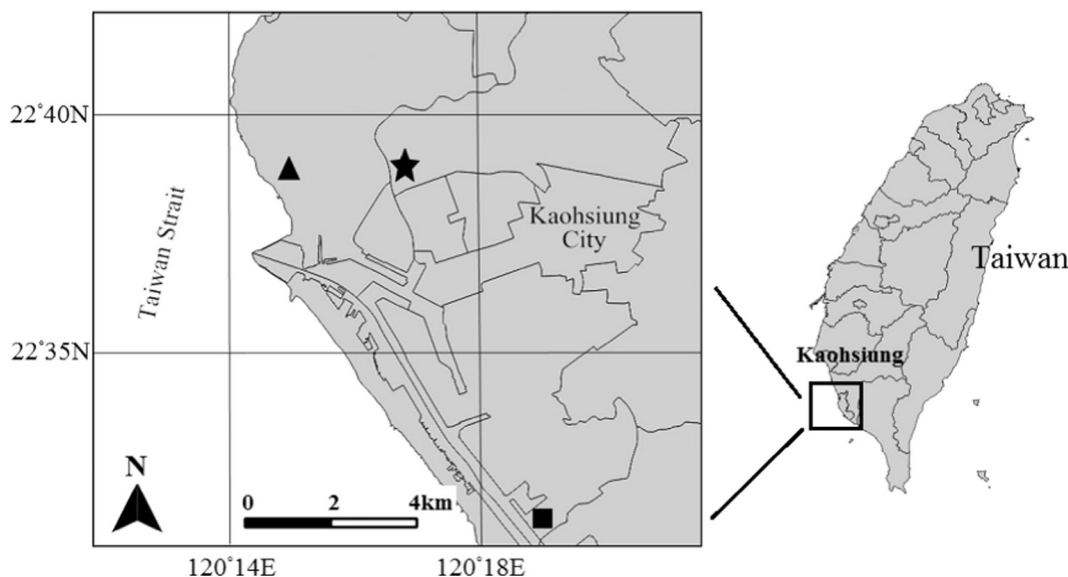
Fig. 1. Map of the sampling sites. The star symbol indicates the location of the PM10 and PM2.5 monitoring site, the triangle symbol represents the location of the PAH sampling site, and the square symbol represents the location of the meteorological data monitoring site.