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Estimation of the local and long-range contributions to particulate matter levels using continuous measurements in a single urban background site

Marianna Diamantopoulou [a], Ksakousti Skyllakou [a, b], Spyros N. Pandis [a, b, c, *]

[a] Department of Chemical Engineering, University of Patras, Patras, Greece
[b] Institute of Chemical Engineering Sciences, Foundation for Research and Technology Hellas (FORTH/ICE-HT), Patras, Greece
[c] Department of Chemical Engineering, Carnegie Mellon University, Pittsburgh, PA 15213, USA

HIGHLIGHTS

- Simple observation-based method to estimate local pollutant contributions.
- Requires continuous measurements in urban background site.
- Estimated local component includes nearby sources in some cases.

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ABSTRACT

The Particulate Matter Source Apportionment Technology (PSAT) algorithm is used together with PMCAMx, a regional chemical transport model, to develop a simple observation-based method (OBM) for the estimation of local and regional contributions of sources of primary and secondary pollutants in urban areas. We test the hypothesis that the minimum of the diurnal average concentration profile of the pollutant is a good estimate of the average contribution of long range transport levels. We use PMCAMx to generate "pseudo-observations" for four different European cities (Paris, London, Milan, and Dusseldorf) and PSAT to estimate the corresponding "true" local and regional contributions. The predictions of the proposed OBM are compared to the "true" values for different definitions of the source area.

During winter, the estimates by the OBM for the local contributions to the concentrations of total $PM_{2.5}$, primary pollutants, and sulfate are within 25% of the "true" contributions of the urban area sources. For secondary organic aerosol the OBM overestimates the importance of the local sources and it actually estimates the contributions of sources within 200 km from the receptor.

During summer for primary pollutants and cities with low nearby emissions (ratio of emissions in an area extending 100 km from the city over local emissions lower than 10) the OBM estimates correspond to the city emissions within 25% or so. For cities with relatively high nearby emissions the OBM estimates correspond to emissions within 100 km from the receptor. For secondary $PM_{2.5}$ components like sulfate and secondary organic aerosol the OBM's estimates correspond to sources within 200 km from the receptor. Finally, for total $PM_{2.5}$ the OBM provides approximately the contribution of city emissions during the winter and the contribution of sources within 100 km from the receptor during the summer.

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1. Introduction

Particulate matter has been linked to a series of health problems including damages to respiratory and cardiovascular systems and even premature death (Dockery et al., 1993; Pope et al., 2001). These particles also contribute to climate change by scattering and absorbing sunlight and changing the optical properties and lifetime of clouds (Schwartz, 1996).

Pollutants can travel from one region to another, thus the pollution levels in an urban area are due to emissions inside the city (local emissions) but also to emissions originating from places far away (long range-transport). In order to improve air quality in

* Corresponding author. Department of Chemical Engineering, University of Patras, Patras, Greece.
E-mail address: spyros@andrew.cmu.edu (S.N. Pandis).

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urban areas we need to quantify the local and regional source contributions. Three-dimensional chemical transport models (CTMs) are well suited to help quantify these source-receptor relationships since they simulate all the necessary processes that impact air pollution concentrations in the domain (Lawrence et al., 2007; Ying and Kleeman, 2009; Wagstrom and Pandis, 2011a).

A number of approaches have been proposed and used for the estimation of local and regional pollution in different areas. Modeling efforts with CTMs often use the so called 'zero-out analysis' or 'brute-force method' (Park et al., 2003; Knipping et al., 2006; Chin et al., 2007; Koo et al., 2009). This computationally expensive method requires several simulations with the corresponding CTM, zeroing out the emissions of each specific source area, or type. The direct decoupled method (DDM) (Dunker, 1981) calculates the sensitivity of concentrations to the change in emissions. This is done by calculating the local partial derivatives of the concentration of interest to the corresponding emission rates (Dunker et al., 2002). Ying and Kleeman (2006) developed the source-oriented external mixture (SOEM) model which separates the species into different categories based on their source and tracks them separately throughout the simulation. SOEM relates each secondary species to its precursor. For example the concentration of particulate sulfate is traced back to SO_2 sources. SOEM is an accurate but also computationally expensive approach.

A computationally efficient apportionment algorithm, the Particulate Source Apportionment Technology (PSAT), was developed by Wagstrom et al. (2008). The algorithm runs in parallel with the CTM and does not perform the most computational demanding calculations, those for the simulation of atmospheric chemistry (gas, aqueous, or particulate). It takes advantage of the fact that the probability of reaction of a pollutant at a given place and time is independent of its source. PSAT was used for the first time in the Eastern United States (Wagstrom and Pandis, 2011b) to explain source-receptor relationships for fine PM. Skyllakou et al. (2014) extended PSAT to simulate semivolatile organics with the volatility basis set (VBS) and used it for the first time over Europe focusing on local and regional contributions to PM in Paris.

Field measurements have been also used to estimate the role of transported pollution for a given receptor. Beekmann et al. (2015) used measurements in Paris and in surrounding areas to estimate the fraction of the transported PM. They reported that more than 70% of $PM_{2.5}$ was transported into Paris from areas far away from it. Freutel et al. (2013) and Crippa et al. (2013) also stressed the importance of regional transport into Paris for summer and winter respectively. During the REPARTEE experiment Harrison et al. (2012) found that regional processes dominate the concentrations of secondary organic aerosol in London. Stedman and Derwent (2008) used a site-specific PM source apportionment model in order to investigate the relationship between PM concentrations and primary PM emissions for 100 monitoring stations distributed throughout the United Kingdom. They found that the regional contribution to $PM_{2.5}$ in London was 75% of the total. Marcazzan et al. (2001) characterized the composition and the origin of $PM_{2.5}$ in Milan by using 24-h measurements of particulate matter in different areas in the city. Similar sulfate temporal behavior as well as mass concentrations were observed at all monitoring sites, indicating the existence of a dominant background regional component.

All previous observation-based studies have relied on measurements from multiple monitoring sites for the estimation of the local and regional components of PM species. New measurement approaches (monitors for continuous $PM_{2.5}$ and PM_{10} mass concentration, aerosol chemical speciation, black carbon, etc.) allow continuous characterization of PM mass concentration and composition (Fuzzi et al., 2015). In this study we demonstrate that continuous measurements from a single site can be used to estimate the regional contribution at least to a zeroth approximation.

We propose a new simple observation-based method (OBM) to estimate the contribution of local and regional sources of PM in urban centers. To test this method, PMCAMx is used together with the extended PSAT (Skyllakou et al., 2014) to provide both concentration "pseudo-observations" used as input for the OBM and also the true source contributions for evaluation of the OBM predictions.

2. Model description

2.1. The PMCAMx chemical transport model

PMCAMx is a regional three-dimensional CTM (Fountoukis et al., 2011) that simulates horizontal and vertical advection and dispersion, wet and dry deposition, gas, aqueous and aerosol-phase chemistry. The gas-phase chemical mechanism used in this application is based on the SAPRC99 mechanism (Environ, 2004) and includes 211 reactions of 56 gases and 18 free radicals. The mechanism employs one lumped monoterpene and sesquiterpene species, two olefins, two aromatics, and 5 lumped alkanes. PMCAMx has been evaluated in the US (Karydis et al., 2007; Murphy and Pandis, 2009), Mexico City (Tsimpidi et al., 2010; Karydis et al., 2010) and Europe (Fountoukis et al., 2011).

2.2. Particle source apportionment technology algorithm

The Particulate Matter Source Apportionment Technology (PSAT) was developed by Wagstrom et al. (2008). It has the ability to track different categories of sources, such as different source types, source areas, or combination of these. The basic idea behind PSAT is that the probability of reaction, deposition or transport of molecules of a pollutant at a given place and time does not depend on its sources. PSAT is computationally efficient because it does not track each specific species separately inside the CTM. It uses the apportionment of the upwind grid cell to apportion species after the transport calculations and of the same cell for chemistry and removal calculations. PSAT and the corresponding CTM (PMCAMx) run in parallel and PSAT does not interfere with the CTM's calculations. PMCAMx supplies PSAT with the fluxes and mass ratios. Afterwords, PSAT calculates the apportionment for each source separately. The apportionment calculations for the secondary species are based on the apportionment of their precursors. In this work we use the extended PSAT algorithm with the implementation of the VBS approach described by Skyllakou et al. (2014). PSAT has the ability to track all the components of organic aerosol (primary and secondary OA from semi-volatile, intermediate volatility, and volatile organic compounds).

2.3. Model application

In this study PMCAMx is applied over Europe. The model domain covers a region of 5400 × 5832 km^2. A horizontal grid resolution of 36 km × 36 km is used together with 10 vertical layers that extend up to 6 km. Two periods are simulated: one during winter from January 10 to February 9, 2010 and one during summer: July, 1–30 2009. In this study we focus on four European cities (Fig. 1): Dusseldorf, London, Milan, and Paris with populations of 0.6, 8.2, 3, and 12 million respectively. The metropolitan area of these four cities is 220, 8,400, 1,980, and 17,180 km^2 respectively. These cities were selected because they cover a wide range of populations, areas, climatic conditions and pollution levels. Each urban area is described inside PSAT by an appropriate number of computational cells. The number of the computational cells used

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