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Source apportionments of ambient fine particulate matter in Israeli, Jordanian, and Palestinian cities☆

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ABSTRACT

This manuscript evaluates spatial and temporal variations of source contributions to ambient fine particulate matter ($PM_{2.5}$) in Israeli, Jordanian, and Palestinian cities. Twenty-four hour integrated $PM_{2.5}$ samples were collected every six days over a 1-year period (January to December 2007) in four cities in Israel (West Jerusalem, Eilat, Tel Aviv, and Haifa), four cities in Jordan (Amman, Aqaba, Rahma, and Zarka), and three cities in Palestine (Nablus, East Jerusalem, and Hebron). The $PM_{2.5}$ samples were analyzed for major chemical components, including organic carbon and elemental carbon, ions, and metals, and the results were used in a positive matrix factorization (PMF) model to estimate source contributions to $PM_{2.5}$ mass. Nine sources, including secondary sulfate, secondary nitrate, mobile, industrial lead sources, dust, construction dust, biomass burning, fuel oil combustion and sea salt, were identified across the sampling sites. Secondary sulfate was the dominant source, contributing 35% of the total $PM_{2.5}$ mass, and it showed relatively homogeneous temporal trends of daily source contribution in the study area. Mobile sources were found to be the second greatest contributor to $PM_{2.5}$ mass in the large metropolitan cities, such as Tel Aviv, Hebron, and West and East Jerusalem. Other sources (i.e. industrial lead sources, construction dust, and fuel oil combustion) were closely related to local emissions within individual cities. This study demonstrates how international cooperation can facilitate air pollution studies that address regional air pollution issues and the incremental differences across cities in a common airshed. It also provides a model to study air pollution in regions with limited air quality monitoring capacity that have persistent and emerging air quality problems, such as Africa, South Asia and Central America.

1. Introduction

Ambient $PM_{2.5}$ is a major concern to human health worldwide (Pope et al., 2002, 1995; Pope and Dockery, 2006). While many studies link ambient $PM_{2.5}$ exposure with premature mortality and morbidity, questions remain about which chemical species and sources of $PM_{2.5}$ are most damaging to human health. Numerous studies concerning source apportionment of $PM_{2.5}$ and its health effects have been conducted in order to support effective control strategies in urban environments of the USA, Europe and East Asian countries (Heo et al., 2014; Ostro et al., 2011; Peng et al., 2009; Sarnat et al., 2008). However, due to poor emission control and monitoring systems in many Middle Eastern countries, sources of $PM_{2.5}$ are poorly understood and improperly managed in these regions.

Urban air pollution exposure has been identified as a leading cause of disease in the Middle East (Lim et al., 2013). The energy production, industrial activity, transportation, and construction

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associated with the urbanization and industrialization of the Middle East are all potential sources of $PM_{2.5}$ (Tsiouri et al., 2014). Contributions from these factors, along with re-suspended soil from dust storms, have caused $PM_{2.5}$ exposure to become a public health concern in the region. However, estimating the risks of exposure to $PM_{2.5}$ chemical components and sources in order to develop appropriate air quality management strategies is quite difficult in this region due to the lack of information concerning $PM_{2.5}$ levels, spatiotemporal trends, transboundary impacts, and sources. Most previous studies of ambient particulate matter in this region have highlighted the impact of long-range transport of anthropogenic air pollution from Europe to the Middle East, as well as crustal aerosols from North Africa to the Middle East (Sarnat et al., 2010). Several other studies have also explored the removal of anthropogenic and natural sources of urban air pollution through wet and dry deposition processes (Al-Momani et al., 2008, 2002). A comprehensive evaluation of $PM_{2.5}$ in the Middle East is needed to develop meaningful and feasible solutions to reduce $PM_{2.5}$ exposure and its associated health impacts.

The Middle East Consortium for Aerosol Research Study (MECARS) was conducted to better understand the levels and spatiotemporal trends of $PM_{2.5}$ mass and its chemical components in Middle Eastern cities over a 1-year period from January to December 2007. The results found that $PM_{2.5}$ mass, as well as sulfate and crustal $PM_{2.5}$ components, were relatively homogeneous and showed strong site-to-site correlations among Israeli, Jordanian and Palestinian cities (Sarnat et al., 2010). The current analysis comprises a follow-up secondary analysis of the MECARS data and aims to identify source contributions to $PM_{2.5}$ in the region. Since many Middle Eastern cities exhibit similar broad trends in urban air pollution due to the influence of shared meteorology and lack of substantial geographical barriers to separate them, identifying the relative contribution of regional and local sources, as well as the size of areas affected by the sources, can provide key support for targeted, regulatory $PM_{2.5}$ control strategies. Ultimately, with this knowledge of specific $PM_{2.5}$ source contributions, inroads can be made towards the reduction of the public health burden of mortality and morbidity related to $PM_{2.5}$throughout the countries in this region.

2. Data and methods

Integrated 24-h $PM_{2.5}$ samples were collected concurrently at 11 sites in Israel, Jordan, and Palestine every six days from January to December 2007. The sampling sites were located in West Jerusalem, Eilat, Tel Aviv, and Haifa (Israel); Amman, Aqaba, Rahma, and Zarqa (Jordan); and Nablus, East Jerusalem, and Hebron (Palestine). The sites were located in populated areas (excluding the Rahma desert site) away from specific local emission sources. Abdeen et al. (2014) and Sarnat et al. (2010) describe the sampling locations and provide their geophysical information.

$PM_{2.5}$ samples were collected using a four-channel air sampler (URG-3000ABC), which was specially designed for this study and was fitted with two $PM_{2.5}$ cyclones operating at a 16.7 LPM flow rate. The sampler collected $PM_{2.5}$ onto two Teflon filters and one quartz fiber filter with a bypass system in the fourth channel that allowed both cyclones to operate at their designated flow rates and each channel to operate at 8.34 LPM. Flow rates were controlled with critical orifices and the samplers ran for 24 h, from midnight to midnight. Monthly field and lab blanks were analyzed for quality control and assurance and for blank corrections.

Teflon filter samples were used to collect and analyze gravimetric $PM_{2.5}$ mass, major ions (i.e. SO_4^{2-}, NO_3^-, and NH_4^+), and trace metals. Major ions and trace metals were determined by ion chromatography (Dionex) and X-ray fluorescence (XRF), respectively. Quartz filters were prebaked at 550 °C for a minimum of 16 h before use and were analyzed for elemental and organic carbon (ECOC) by thermal-optical analysis (Sunset Laboratory, Inc., Forest Grove, OR) using the ACE-Asia method (Schauer et al., 2003). Explicit details regarding filter sample handling and the chemical analysis procedures are provided by von Schneidemesser et al. (2010) and Sarnat et al. (2010).

Daily source contributions to $PM_{2.5}$ mass across the sampling sites were calculated by a positive matrix factorization (PMF) model, an advanced factor analysis technique based on a weighted least-squares fit and error estimates of the measured data (Paatero and Tapper, 1994). Detailed principles and applications of PMF have been previously described elsewhere in literature (Alolayan et al., 2013; Heo et al., 2013, 2009). The two-way factor analytical model, PMF2, was used in this study (Paatero, 2000).

Allocating uncertainty appropriately to each of measured data points is critical to PMF modeling because its application depends on estimated uncertainties. In this study, the procedure of Polissar et al. (1998) was applied to calculate observed data and its uncertainties as inputs of the PMF analysis. Values below method detection limits (MLDs) were replaced by half of the MDL, and their overall uncertainties were set at 5/6 of the MDL. Several missing data for each of chemical components were replaced by the geometric mean of the measured values as observed values, and the associated uncertainties were set at four times the geometric mean. The signal to noise (s/n) ratio for each of the chemical species was reviewed to determine which, if any, should be discarded or statistically weighted less in order to minimize distortion of the model fit (Heo et al., 2013). The s/n ratio was categorized to three different ranges applied in Heo et al. (2013), but no bad or weak species were discarded or statistically weighted less among the final input variables.

A total of 612 samples across the sampling sites were collected during the MECARS. Several extreme $PM_{2.5}$ events occurred, including dust storms on May 30, 2007 and October 9, 2007, and the Jewish holiday Lag B'Omer on May 6, 2007, which is celebrated in Israel with bonfires, leading to high regional concentrations of $PM_{2.5}$ and its biomass burning components. For the PMF analysis, these extreme events were excluded to avoid distortion of specific source contributions through abnormally high contributions of trace elements from these sources during the events. After excluding the extreme events, a total of 580 samples (53 at Aqaba, 56 at Amman, 53 at East Jerusalem, 54 at Hebron, 55 at Haifa, 43 at Eilat, 54 at Nablus, 49 at Rachma, 55 at Tel Aviv, 54 at West Jerusalem, and 54 at Zarqa) were used to perform the PMF model. Forty-six chemical species, including OC, EC, nitrate, sulfate, ammonium, Cl, Al, Si, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Ga, Se, Br, Rb, Sr, Yt, Zr, Nb, Mo, Pd, Ag, Cd, In, Sn, Sb, Ce, Sm, Eu, Tb, Hf, Ta, Wo, Ir, Au, Tl, Pb, and Ur, were used in the PMF model. PMF modeling usually requires information on at least 100 samples to produce reliable source profiles and contributions (US EPA, 2014). Each of the sampling sites collected only about 50 samples during the MECARS. Thus, the observed data points across sites were combined into one data set to generate the PMF model output and location-specific results of source contributions were then generated using the aggregated PMF in this study.

The PMF model was run multiple times with a variety of factor quantities, and different pseudorandom numbers used in the iterative fitting process were also examined to find the global optimal PMF solutions. The robust mode was used to reduce the effects of extreme values in the analysis, and the FPEAK parameter was applied to control rotational ambiguity. The measured $PM_{2.5}$ mass concentrations were included as input variables in PMF analysis in order to properly treat the mass closure issue; thus, the apportioned $PM_{2.5}$ concentrations from each source were calculated