Atmospheric Environment 108 (2015) 140–150

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Sources and temporal variations of constrained PMF factors obtained from multiple-year receptor modeling of ambient $PM_{2.5}$ data from five speciation sites in Ontario, Canada

Uwayemi M. Sofowote [a,*], Yushan Su [a], Ewa Dabek-Zlotorzynska [b], Ankit K. Rastogi [c], Jeff Brook [d], Philip K. Hopke [e]

[a] Air Quality Monitoring and Assessment Unit, Environmental Monitoring and Reporting Branch Ontario Ministry of the Environment and Climate Change, 125 Resources Road, Etobicoke, Ontario M9P 3V6, Canada
[b] Analysis and Air Quality Section, Air Quality Research Division, Atmospheric Science and Technology Directorate, Science and Technology Branch, Environment Canada, 335 River Road, Ottawa, Ontario K1A 0H3, Canada
[c] Faculty of Medicine, University of Toronto, 1 King's College Circle, Toronto, Ontario M5S 1A8, Canada
[d] Process Research Section, Air Quality Research Division, Atmospheric Science and Technology Directorate, Science and Technology Branch, Environment Canada, 4905 Dufferin Street, Toronto, Ontario M3H 5T4, Canada
[e] Institute for a Sustainable Environment, Clarkson University, PO. Box 5708, Potsdam NY 13699-5708, USA

HIGHLIGHTS

- The coefficient of variation of imposed constraints was used to select the right post-PMF analyses.
- Post-PMF work used trajectory ensembles & surface winds conditional probabilities.
- Temporal trends of PMF factors showed time-based anthropogenic & natural influences.

ARTICLE INFO

Article history:
Received 24 September 2014
Received in revised form 21 February 2015
Accepted 23 February 2015
Available online 24 February 2015

Keywords:
$PM_{2.5}$
PMF
Receptor modeling
Speciation
Source apportionment

ABSTRACT

The Canadian National Air Pollution Surveillance (NAPS) network operates five fine particulate matter ($PM_{2.5}$) speciation sites in Ontario. Data from 2005 to 2010 from these sites were subjected to constrained positive matrix factorization (PMF) to obtain factors of particulate matter pollution. Eight factors were found to be common across the sites. These constrained factors were particulate sulfate and nitrate, aged combustion aerosols, biomass/wood combustion, non-ferrous metal smelting, crustal matter, steel works, and road salt. Other factors found at these sites were largely related to local sources in their respective neighbourhoods. Diagnostic results from constraining the PMF factors could be used to select the appropriate post-PMF analyses as they reveal the factors that were expected to have regional or local sources. Interpretation of the spatial trends of factors was guided by the extent of similarity of target ratios across the five sites as determined by their coefficient of variation (C.V.). On average, particulate sulfate had $PM_{2.5}$ mass contributions that ranged from 7% (Wallaceburg) to 36% (Simcoe) and nitrate species ranged from 7% (Simcoe) to 23% (Windsor). A clear low rural – high urban divide for particulate nitrate was also observed. These non-uniform impacts across the sites indicate that a single representative location is not suitable for studying $PM_{2.5}$ in Ontario. Temporal variations showed greater abundance of particulate sulfate in the summer and particulate nitrate in the winter respectively. Yearly median values of the sulfate factor that is largely energy-related have been declining for most sites since peaking in 2007. A trajectory ensemble model, simplified quantitative transport bias analysis (sQTBA) was used to identify potential source regions of the constrained sulfate and nitrate factors while radial plots of the conditional probability function (CPF) were used to explore the more locally-sourced factors. The US Midwest, east coast and Canada's west were dominant regional sources.

* Corresponding author.
E-mail address: Uwayemi.Sofowote@ontario.ca (U.M. Sofowote).

http://dx.doi.org/10.1016/j.atmosenv.2015.02.055

1. Introduction

In Canada, the National Air Pollution Surveillance (NAPS) program, a cooperative program of the federal, provincial, territorial, and some municipal governments, supports multiple air quality programs across the country, and has been in operation for over 40 years (Environment Canada, 2013a). This network is predominantly urban, and in addition to measuring criteria air pollutants (SO_2, NO_x, CO, O_3 and particulate matter (PM)); it also measures a wide variety of PM constituents and volatile organic compounds (VOCs) in the air at a subset of sites. In January 2003, the NAPS $PM_{2.5}$ chemical speciation program was initiated with the purpose of measuring significant $PM_{2.5}$ constituents that include ammonium nitrate, ammonium sulfate, trace elements, organic and elemental carbon (Dabek-Zlotorzynska et al., 2011). As of 2014, the program operates 12 active monitoring sites nationwide, with most of them located in major urban areas, and some in non-urban locations having $PM_{2.5}$ impacts from regional pollution sources.

In Ontario, 5 NAPS $PM_{2.5}$ sites in the speciation program (Fig. 1) are located along a corridor that extends from Windsor to Ottawa, covering roughly 700 km and spanning 3 degrees of latitude and 7 degrees of longitude in southern Ontario. These metropolitan areas account for about 56% of the province's population. The pollution data from these NAPS speciation sites can be used to obtain a better understanding of the variations and distributions of emissions sources impacting their general metropolitan locations within the province. Different exposures may arise across the province due to the variations in concentrations that may arise due to differences in proximity to emission sources, local meteorology, population demographics, and land usage patterns. Thus, there is a need to characterize the particulate matter pollution to which these different populations within the province are exposed. To this end, we applied constrained positive matrix factorization (PMF) of $PM_{2.5}$ speciation data over multiple years (2005–2010) as described by Sofowote et al. (2015). The current work follows from that study with the goal of using simple parameter values derived from the applied constraints to select the appropriate post-PMF analyses for spatial source apportionments. Post-PMF auxiliary receptor models include advanced trajectory ensemble models (TEMs) and conditional probability methods for surface wind data. TEMs aid in spatial assessment of PMF factors especially with regards to regional sources of pollution (Liu et al., 2003a). In this study, simplified quantitative transport bias analysis, sQTBA (Zhou et al., 2004; Brook et al., 2004), and conditional probability functions (CPF; Ashbaugh et al., 1985; Kim et al., 2003) were used.

Previously, Jeong et al. (2011) applied unconstrained PMF (in which no a priori information about sources was used) and potential source contribution function (PSCF) to data from five NAPS sites across Canada; including two in Ontario, They also performed a subsequent study including the site at Simcoe (Jeong et al., 2013). In this work, we have used sQTBA because it solves the natural transport potential function and unlike PSCF, it does not require a subjective definition of a 'polluted' criterion value. The natural transport potential function is infinitely large at the receptor location but approaches zero the more distant the trajectory segment is from the receptor. This approach yet again distinguishes it from PSCF since it does not assume that all segments of the trajectories have equal probability of reaching the receptor site. The potential for identification of spurious source regions situated around edges of the trajectory domain is thus intrinsically reduced. Finally, one of the goals of this study was to determine the sensitivity of receptor and trajectory ensemble models to the temporal and spatial variations in regional particulate matter pollution affecting southern Ontario over a six-year period that bracketed the 2008 global economic downturn.

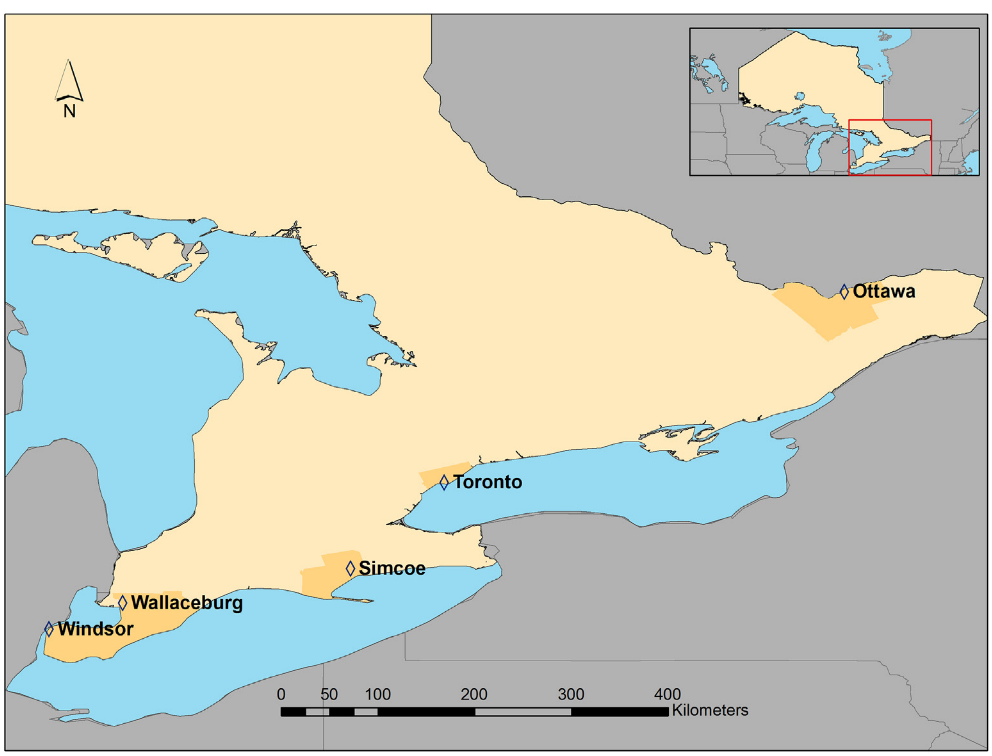

Fig. 1. Map showing locations of the 5 NAPS $PM_{2.5}$ speciation sites: Ottawa (OTT), Toronto (TOR), Simcoe (SMC), Wallaceburg (WBG) and Windsor (WND) and their census areas in Ontario.