APPLICATION OF AN EMISSIONS SOURCE APPORTIONMENT METHOD FOR PRIMARY PM COMPONENTS IN A REGIONAL AIR QUALITY MODEL

Mehrez Samaali*, Mourad Sassi, Véronique Bouchet
Air Quality Modeling Application Section, Canadian Meteorological Centre, Environment Canada, 2121 Trans-Canada highway, Dorval, QC, H9P 1J3, Canada.

Michael Moran
Air Quality Research Division, Environment Canada, 4905 Dufferin Street, Toronto, ON, M3H 5T4, Canada.

1. INTRODUCTION

Particulate matter of aerodynamic diameter lower than 2.5um (PM2.5) are pollutants of concern for human health. Therefore, control strategies are applied to reduce emissions that contribute to their presence and formation in the atmosphere (Kleeman and Cass., 2001). These controls strategies use 3D regional Eulerian air quality (AQ) models as tools to assess the spatial and temporal distributions of ambient PM2.5 levels in response to changes in emissions from various sources. As models don't generally track information about the origin of the precursors that led to its formation, source apportionment methods are required to quantify the contribution of individual sources to the total PM pollution. In the last decade, many studies on sources apportionment methods ranging from relatively simple approaches focusing on primary sources (e.g. Bhave et al., 2004; Lane et al., 2007) to complex ones handling secondary sources (e.g. Kleeman et al., 2001; Yarwood et al., 2004; Held et al., 2004) have been published. There are two main categories of source apportionment methods that are generally used in the context of an Eulerian 3D AQ model (Yarwood et al., 2004): (1) *sensitivity analysis methods*, and (2) *reactive tracer methods*. *Sensitivity methods* consist of measuring the model response to an input perturbation (for instance, evaluating the change of nitrate concentration due to a change in nitrogen dioxide emissions of a given source). *Reactive tracers methods* (also called *tagged species* methods) are more complex and consist of introducing extra species in the model to tag and track pollutants from specific sources. Both methods have been successfully implemented and tested in American AQ models such as CMAQ and CAMx. Although current Canadian AQ models (i.e. AURAMS and CHRONOS) have been used to perform scenario analyses in support of policy decisions, they do not include a sophisticated source apportionment capacity based on tagged species methods.

The objective of this paper is to present the implementation, the verification and the application of a tagged species method in a Canadian regional AQ model. As this method has been proven to be very complex particularly for secondary sources (e.g. Kleeman et al., 2001; Yarwood et al., 2004), the first step was to apply it for primary sources of organic and elemental carbons. Potential applications of the method to track the contribution of transport sources of primary organic (POC) and elemental carbons (EC) in major Canadian cities will also be discussed.

2. AQ MODEL DESCRIPTION AND SETTINGS

In this study, AURAMS (**A U**nified **R**egional **A**ir **Q**uality **M**odeling **S**ystem), Environment Canada's current 3D regional AQ model, was used to implement and verify the tagged species method. The model is used as a comprehensive tool for air quality research and regulatory applications to support policy makers in AQ management decisions in Canada. The model includes a full gas and particle phase chemistry description and simulates ozone and PM in the troposphere. Removal processes include wet and dry deposition of gas and particles as well as rain, fog and cloud processing. The model is driven off-line using GEM (Gong et al., 2006), Environment Canada's weather forecast model. Oxidant and aqueous-phase chemical processes are based on updated versions of the Acid Deposition and Oxidant Model (ADOM) mechanisms (Venkatram et al., 1988), while aerosol dynamics are

**Corresponding author:* Mehrez Samaali, Air Quality Modeling Application Section, Canadian Meteorological Centre, Environment Canada, 2121 Trans-Canada highway, Dorval, QC, H9P 1J3, Canada; e-mail: mehrez.samaali@ec.gc.ca.

described using the chemically speciated and size-resolved aerosol module CAM (Gong et al., 2002). Overall, the model resolves 157 species distributed in 49 gaseous species and 9 aerosols species (8 particulate and particle-bound water species) in 12 size bins each. The PM species include POC, EC, crustal material, sulfate, nitrate, ammonium, secondary organic carbon and sea salt. Only POC and EC are the focus of this study from a tagging perspective.

The model vertical height extends up to 30 km (30 levels) with relatively thin layers near the surface and thicker layers in the upper atmosphere. In the simulations presented here, the model uses a horizontal resolution of 42 km and covers the entire North American domain. Since the boundary conditions for AURAMS are currently defined using the no-gradient method, Eastern and Western boundaries extend well over ocean water to minimize potential errors of this method. Global fields generated with GEM were used to derive interpolated meteorological fields for the simulation domain. On-road, off-road, minor point and others emission sources were considered as area sources and released at the model surface level, while major points sources are allowed to inject emissions at the appropriate model vertical layer. While anthropogenic emissions fluxes are provided to the model with an hourly time step, biogenic emissions are calculated online using BEIS3v0.9.

The simulation was performed for the 2002 summer period (June-July-August) starting on May 25th 2002. The 25th-31st May period was included in the analysis in order to diagnose initial tagged species concentrations during the model's spin-up period.

3. SOURCE APPORTIONMENT METHOD DESCRIPTION

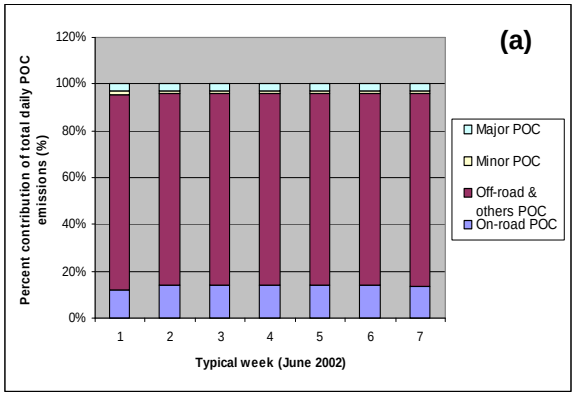
Eight PM tagged species (or reactive tracers) were introduced in AURAMS in order to tag and track the contribution of four main sources of POC and EC across the model domain : "on-road", "off-road and others", "minor point" and "major point" point sources. "On-road" sources include vehicles, tucks and motorcycles; "off-road and others" sources include construction, agriculture and domestic engines, marine, rail and air transport, coal, oil and natural gas combustion, etc.; "minor point" sources represent facilities with a stack height lower than 35m, and "major point" sources gather facilities with a stack height higher than 35 m (e.g. power plants, refineries, etc.).

With the tagging approach, the contribution of these sources can be obtained at each time step and grid point. The method was applied to the PM chemically and size-resolved representation used in AURAMS. As a consequence, the total number of species was increased from 108 to 204 in the aerosol module. As for the original POC and EC species, all dynamical, physical and chemical processes such as transport (3D advection) and vertical diffusion, removal as well as chemical emission speciation were solved for the tracers.

4. EMISSIONS INVENTORY

The primary emissions data are derived from the 2002 U.S. EPA's National Emission Inventory (NEI) Version 3 (U.S. EPA, 2007a), the 2002 Canadian inventory released by Environment Canada, and the 1999 Mexico's National Emissions Inventory (MNEI) released by the U.S. EPA (U.S. EPA, 2007b). The data were processed by SMOKE to produce temporal and spatial emissions rates for the four sources for all of 2002 and over the entire North American domain. It was assumed that the Mexican emissions did not grow over the 1999-2002 period.

Fig. 1 shows the relative contributions of the sources of interest ("on-road", "off-road and others", "minor" and "major" points) to the total emissions of POC and EC for a typical week in June 2002. "Off-road and others" sources are the largest sources (more than 80%) of POC (Fig. 1a), whereas on-road sources are the most important emitter (more than 55%) of EC (Fig. 1b). Minor and major point sources are the lowest contributors (less than 2% and 12 %, respectively) for both components. Of interest is also the small variation between Sunday, Monday, Saturday (days 1, 2 and 7, respectively) and the other week days which is due to the temporal allocation of emissions giving a relatively higher weight to these days than the other week days for both POC and EC (Fig. 1). Emissions from other months (i.e. July and August) are not presented here since they are practically identical to those for June.



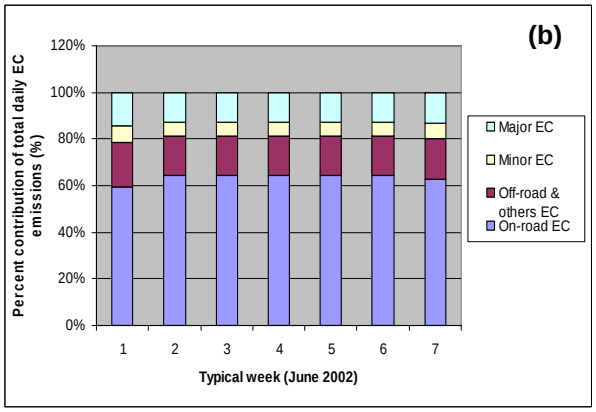


Fig. 1. Relative contribution of the tagged sources in total POC (a) and EC (b) emissions for a typical week in June 2002.

5. MODELING RESULTS

Time series of average concentrations and average relative contributions of the tagged species to the total POC and EC concentrations are displayed in Fig. 2 and Fig. 3, respectively. All results correspond to daily values averaged over all grid points and the first five vertical layers above the surface. Since there are no initial and boundary conditions used for POC and EC or the tracers, the tracer concentrations can be summed up and compared against total POC and EC concentrations for the entire simulation period. On average, the tracer concentrations sums (''Total-recalc'' curve in Fig. 2) agree well with total POC and EC reference (no tagging) concentrations (''Total'' curve in Fig. 2) over the whole domain: average absolute error are 2.2 10^{-4} ug/m^3 and 2.4 10^{-4} ug/m^3 and relative errors are 0.37 and 1.48 %, in figures 2a and 2b, respectively. These results are comparable to those outlined by Lane et al. (2007) who applied a sensitivity analysis method for primary organic aerosol apportionment using PMCAMx. Lane et al., (2007) attribute the small errors obtained with their method to numerical issues in the model and particularly to errors in the 3D vertical and horizontal transport, diffusion and removal processes calculations.

Over the entire simulation period, POC and EC are highest (0.077 ug/m^3 and 0.026 ug/m^3, respectively) on July 17th, 2002 and average levels are equal to 0.06 ug/m^3 and 0.02 ug/m^3, respectively. The daily variations of concentrations (Fig. 2) can be attributed to meteorology (wind magnitude and direction, temperature, clouds) and/or to the daily emissions levels which change from weekdays to week-ends. Overall, the contributions of each source to POC and EC concentrations agree reasonably well with the corresponding input emissions contributions (Fig. 1 *versus* Fig. 3).

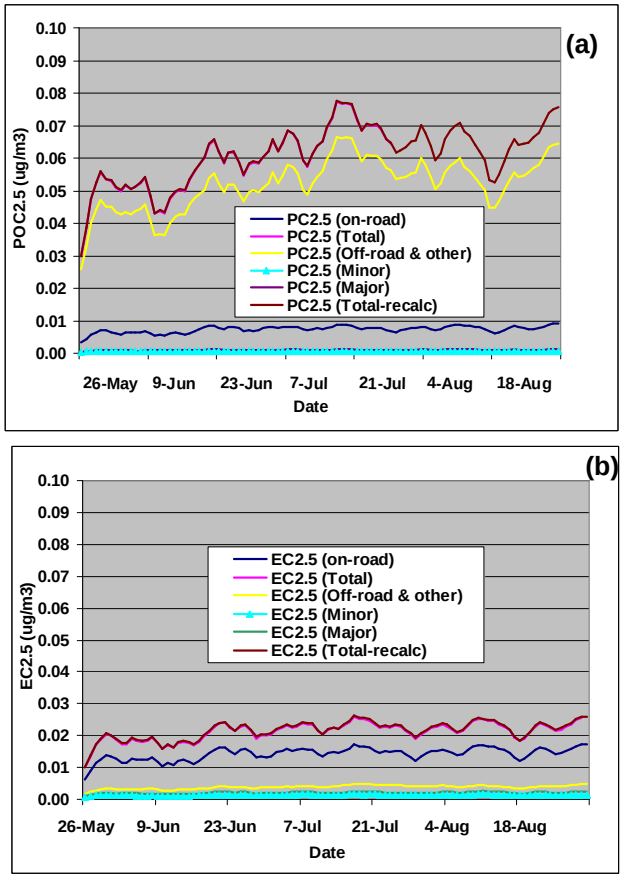
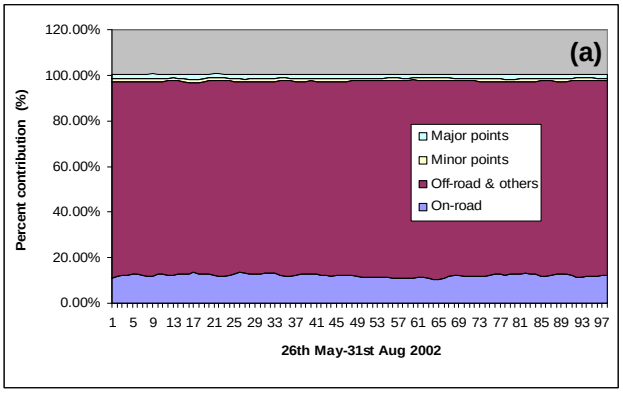


Fig. 2. Domain-wide mean daily PM2.5 POC (a) and PM2.5 EC (b) concentrations for 25th May-31st August 2002



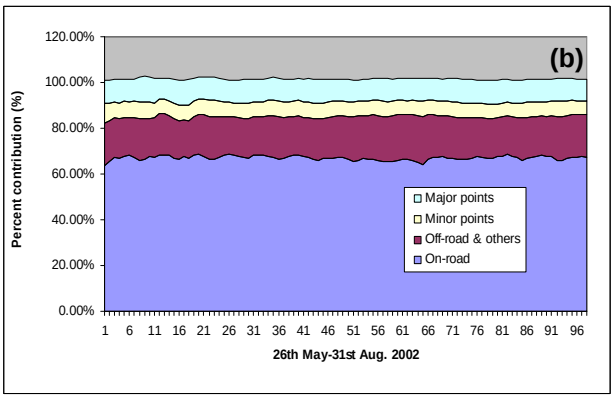


Fig. 3. Domain-wide mean daily contributions of the tagged species to total POC and EC concentrations for 26th May-31st August 2002.

Although the method was only applied to primary pollutants, it adds a new capability to AURAMS for other useful air quality applications at the regional scale. One potential application is to investigate the contribution of major tagged sources (i.e. "on-road" and "off-road and other" sources) in the largest cities in Canada. Fig. 4 and Fig. 5 show the contribution of these two sources in Montreal, Toronto, Ottawa, Vancouver, and Calgary. The contribution of on-road sources to POC and EC concentrations are highest in Montreal (more than 20% and 85%, respectively) when compared to the other Canadian cities. Ottawa and Toronto exhibit comparable contribution levels whereas Calgary is the city where on-road sources contribute the least. Vancouver is an intermediate case between Ottawa and Calgary (Fig. 4). The situation is reversed for "off-road and others" sources (Fig. 5). Calgary has the most important contribution of this source to POC and EC concentrations (more than 25% and more than 90%, respectively) because of the surrounding petroleum activities and off-road machines. Montreal shows the lowest contribution levels for both species. Toronto shows a similar contribution to that of Ottawa whereas Vancouver shows a slightly higher contribution (2-3% difference) compared to these two cities.

Overall, as for the entire domain, there is usually a correspondence between the relative fractions of emission and ambient concentration of each source for all of the cities. However, Calgary and Ottawa are particular cases with differences up to 6%. The transport of pollutants from surrounding cities with relatively high emissions levels of POC and EC could be an explanation of these differences. This hypothesis needs more investigation to better understand the source apportionment results.

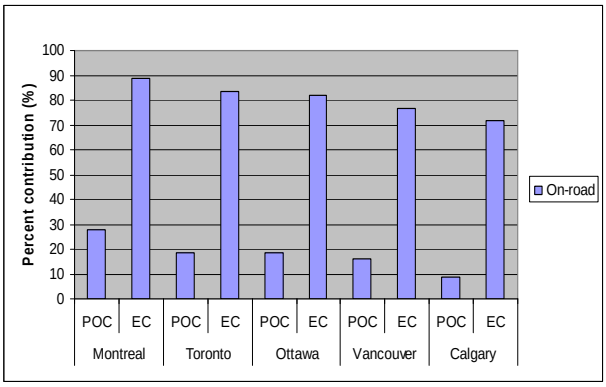


Fig. 4. Average contribution of on-road sources to total POC and EC concentrations in main Canadian cities for the entire simulation period.

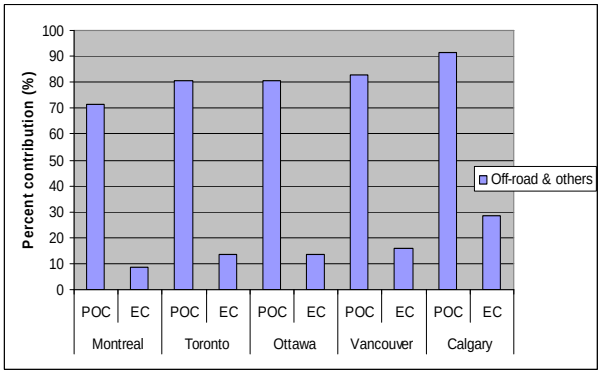


Fig. 5. Average contribution of off-road and other sources to total POC and EC concentrations in main Canadian cities for the entire simulation period.

Note that these results could also be made using zero-out methods as done by Lane et al., 2007, but the tagged species method is more efficient since it handle the individual sources with the whole model PM species in a unique tool (e.g. Yarwood et al., 2004; Kleeman and Cass, 2001) without having to run as many simulations as the zero-out approach. However, one disadvantage of the tagging method is the increase in computing time of a single simulation (by 62% in the study presented here) due to the increase in the number of model species. This is a consideration of concern particularly for future long-term (annual) simulations planned in this project. An optimization of all model routines is underway to try to keep the increase in computing resources to a minimum.

6. CONCLUSION AND FUTURE WORK

A source apportionment method for primary carbonaceous sources was implemented in AURAMS and a simulation over North America for

a summer period (June-July-August 2002) was carried out. The results were verified by comparing tagged species contributions to the total POC and EC concentrations and emissions. A good agreement was found between POC and EC source contributions and the respective input emissions as expected from since only primary species are considered here. The method was applied to apportion the contribution of a particular source (transportation) to POC and EC concentrations for major cities (Montreal, Toronto, Ottawa, Vancouver and Calgary) in Canada. With this new capability, AURAMS will be a useful tool to investigate the contribution of potential POC and EC transportation sources in Canada over annual simulations.

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