PM2.5 SOURCE APPORTIONMENT COMPARISON OF CMAQ AND CAMX ESTIMATES

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1. INTRODUCTION

It is useful to understand what types of sources or regions are contributing to chemically speciated PM2.5 estimated by photochemical grid models. Understanding the contribution from particular sources to specific geographic receptor locations helps regulators develop effective emissions control strategies. Source apportionment is an alternative approach to zero-out modeling and has the advantage of being much more efficient with computational resources. For instance, to estimate the contribution from 20 source regions a total of 20 individual zero-out simulations would be needed compared to a single source apportionment simulation. The incremental run-time associated with the additional source region tracking is far less than performing numerous iterative simulations.

The Particle and Precursor Tagging Methodology (PPTM) has been implemented in CMAQ v4.6 and tracks contribution to mercury and PM sulfate, nitrate, ammonium, secondary organic aerosols, and inert species (ICF International 2007a, ICF International 2007b). PPTM estimates contributions from emissions source groups, emissions source regions, initial conditions, and boundary conditions to PM2.5 by adding duplicate model species for each contributing source. These duplicate model species (tags) have the same properties and experience the same atmospheric processes as the bulk chemical species. The tagged species are calculated using the regular model solver for processes like dry deposition and advection as bulk species. Non-linear processes like gas and aqueous phase chemistry are solved for bulk species and then apportioned to the tagged species.

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Particulate matter source apportionment technology (PSAT) has been implemented into the most recent version of the CAMx model (v4.5) and is publicly available (ENVIRON, 2008; Wagstrom et al, 2008). PSAT estimates the contribution from specific emissions source groups, emissions source regions, initial conditions, and boundary conditions to PM2.5 using reactive tracers. The tracer species are estimated with source apportionment algorithms rather than by the host model routines. PSAT tracks contribution to mercury and PM sulfate, nitrate, ammonium, secondary organic aerosol, and inert species. Non-linear processes like gas and aqueous phase chemistry are solved for bulk species and then apportioned to the tagged species.

The Milwaukee metropolitan area was chosen for an exercise comparing absolute model predictions and source contribution estimates using CAMx and CMAQ particulate source apportionment. The particulate source apportionment estimates from CAMx and CMAQ are compared for 11 specific geographic tags, all other sources that were not tagged, and boundary conditions at monitors located in Milwaukee and Waukesha counties in Wisconsin. The 11 tags constitute the emissions from all source sectors in entire counties or groups of counties. Individual apportionment contributions from each source region and absolute model predictions for PM2.5 species are compared between the CAMx and CMAQ particulate source apportionment applications.

2. METHODS

Each model was applied to a modeling domain (Figure 1) covering the Midwest United States with 12 km sized grid cells over 4 months in 2002: January, April, July, and October.

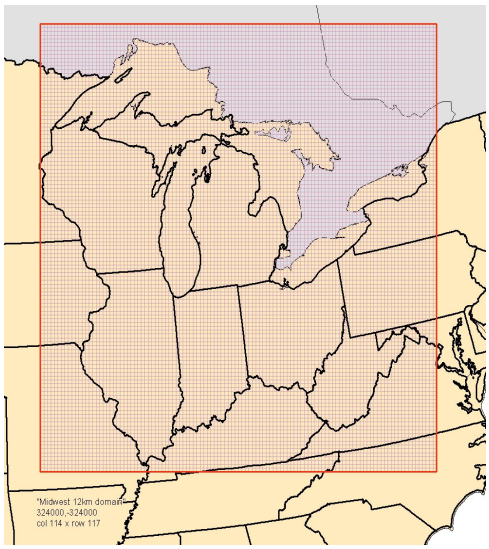

Figure 1. Modeling domain (red box)

Contributions are tracked for 11 county groups in the Milwaukee-Chicago area, initial conditions, boundary conditions, and all other emissions sources in the modeling domain that are not explicitly tracked as part of the 11 county groups (Figure 2). The SMOKE emissions model was run separately for each source region to ensure that each geographic region tag only contains emissions for that specific area.

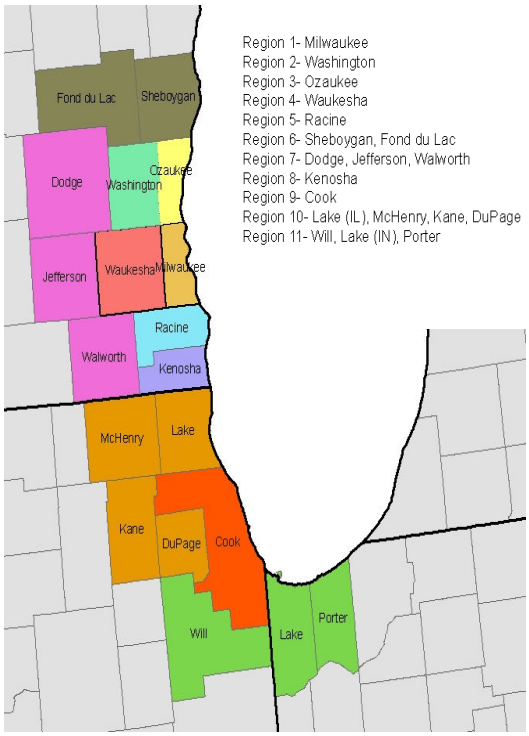

Figure 2. Source regions tracked for contribution

Fire emissions were not tracked as part of any county or county group tag and are included in the all other sources in the modeling domain tag. Emissions are based on a 2002 emission inventory for all source sectors (Strum et al, 2008).

Particulate source apportionment tracks contributions to particulate species from pre-cursor emissions. Emissions of nitrogen oxides are tracked through all intermediate nitrogen species to particulate nitrate ion. Ammonia emissions are tracked to particulate ammonium ion. Even though ammonium nitrate is chemically coupled, the apportionment schemes do not attempt to determine which species is limiting the formation, but directly attributes precursor gases to specific particulate ions (Figure 3). PM2.5 contribution is defined for the purposes of this evaluation as the sum of PM2.5 sulfate ion, nitrate ion, ammonium ion, organic carbon mass, and elemental carbon. The soil component is not included due to an emissions processing error that led to large systematic over-estimates of soil emissions.

$NOX \rightarrow NO3^-$
$SOX \rightarrow SO4^=$
$NH3 \rightarrow NH4^+$
$POC \rightarrow POC$
$PEC \rightarrow PEC$
$SOIL \rightarrow SOIL$

Figure 3. Emissions pre-cursor species (left) tracked to PM2.5 species (right)

Operational model performance is assessed for each model to help discern whether differences in model predicted source contribution may be related to differences in absolute model prediction. This comparison to observations at the receptor locations of interest also helps establish confidence in the estimated contribution. Metrics used to describe model performance include mean bias, gross error, fractional bias, and fractional error (Boylan et al., 2006). The bias and error metrics describe performance in terms of measured concentration units and the fractional metrics describe performance as a percentage. The best possible performance is when the metrics approach 0. The fractional metrics are bounded by 200%, which is considered very poor performance.

3. RESULTS & DISCUSSION

3.1 Operational Evaluation

Model performance at a Milwaukee county and Waukesha county Speciation Trends Network monitor are fairly similar for both models (see Figure 4). The largest differences in prediction are with the nitrate ion. CMAQ tends to predict higher concentrations of ammonium nitrate at this location. Both models show the best agreement with observations for PM2.5 sulfate ion.

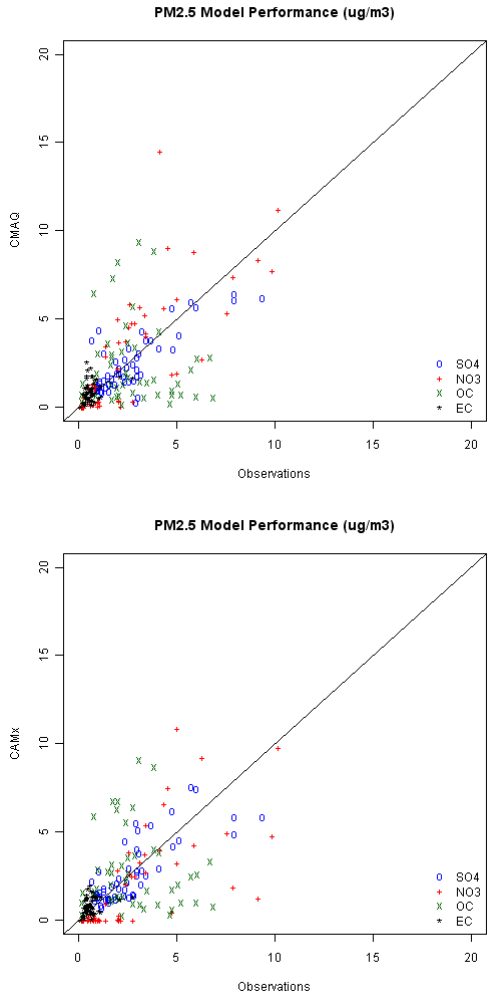

Figure 4. CMAQ (top) and CAMx (bottom) model performance at 2 monitor locations

CMAQ and CAMx predictions of PM2.5 sulfate ion, nitrate ion, organic carbon, and elemental carbon are shown in Figure 5. Both models tend to predict very similar concentrations of sulfate, organic carbon, and elemental carbon. The CMAQ model tends to predict more PM2.5 nitrate ion.

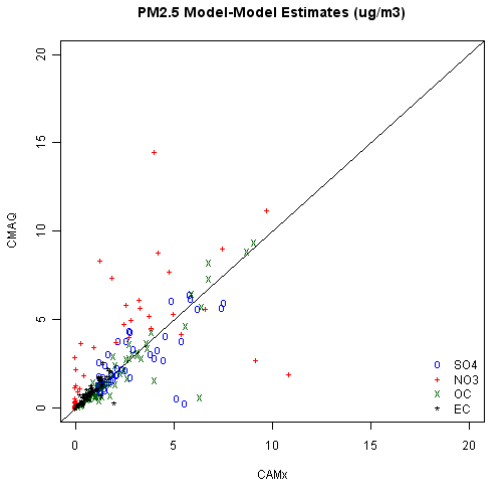

Figure 5. CMAQ and CAMx estimates of speciated PM2.5

The predictions at these monitor locations are fairly similar for each model, particularly for organic and elemental carbon. Table 1 shows bias and error metrics for each model at the Milwaukee and Waukesha county monitor locations. Both models have similar error estimates for each of the key PM2.5 species at these receptors.

Table 1. Model performance metrics at Milwaukee and Waukesha STN monitors

		CAMx			
specie	N	Bias	Error	Fr Bias	Fr Error
$SO4^{-}$	132	0.04	1.05	-1	34
$NO3^{=}$	132	-0.71	1.47	-98	115
OC	124	-0.93	1.97	-34	85
EC	132	0.05	0.30	0	49

		CMAQ			
specie	N	Bias	Error	Fr Bias	Fr Error
$SO4^{-}$	132	0.02	1.07	-7	35
$NO3^{=}$	132	0.37	1.33	-21	71
OC	124	-0.79	2.00	-28	84
EC	132	0.17	0.36	12	53

3.2 Source Estimate Comparison

The contribution from each of the source regions is estimated at receptor locations in Milwaukee and Waukesha counties. The model estimates are the average of the top 10% of modeled days and the average of all days. The source contribution is averaged

over all receptor locations in Milwaukee and Waukesha counties for each day before multiple days are aggregated. Figure 6 shows the estimated contribution over all days and the top 10% of days for each source region.

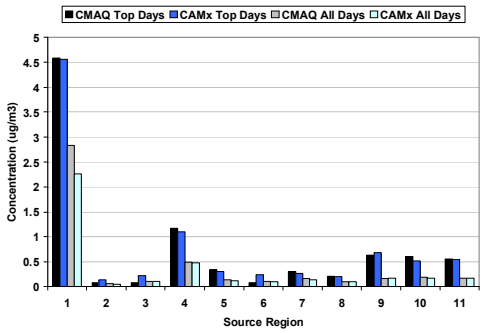

Figure 6. Contribution estimation of PM2.5 at Milwaukee county receptor

The CAMx and CMAQ estimated contribution from each source region to the Milwaukee/Waukesha area is consistent. Even though the models often predict different absolute concentrations for the PM2.5 species, the contribution estimates are fairly similar when aggregated over all days or over the top 10% of modeled days.

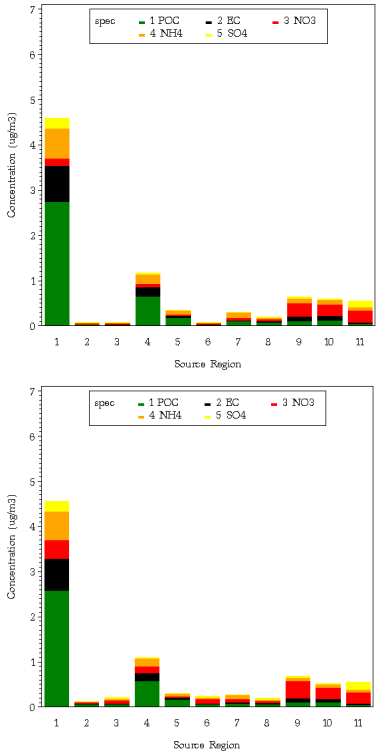

Figure 7. PM2.5 contribution by chemical species estimated by CMAQ (top) and CAMx (bottom)

The total PM2.5 contribution can also be examined at each receptor location by chemical composition. Figure 7 shows the average of the top 10% of modeled days to Milwaukee/Waukesha by chemical specie. This type of evaluation suggests that the contribution from more distance source regions tends to be from secondarily formed species like nitrate and sulfate and local contribution tends to be dominated by primarily emitted species.

The spatial patterns of contribution from the source regions tracked in each model are also consistent. Figure 8 shows the 4 month average PM2.5 contribution from each model for source regions Milwaukee county, Waukesha county, and Cook county. The estimated pattern and extent of high contribution are very similar between models.

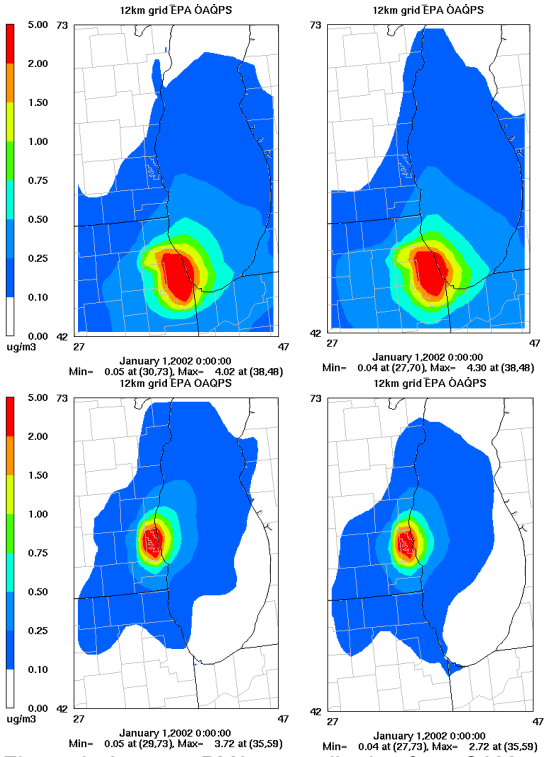

Figure 8. Average PM2.5 contribution from CAMx (right) and CMAQ (left) on bottom Milwaukee county (bottom) and Cook county (top).

The CMAQ and CAMx estimated source contributions are shown in Figure 9. These plots show all receptor locations in the Lake Michigan region over the entire modeling period. The source contributions include all source 11 county group source regions, all

other non-tagged emissions, and boundary conditions.

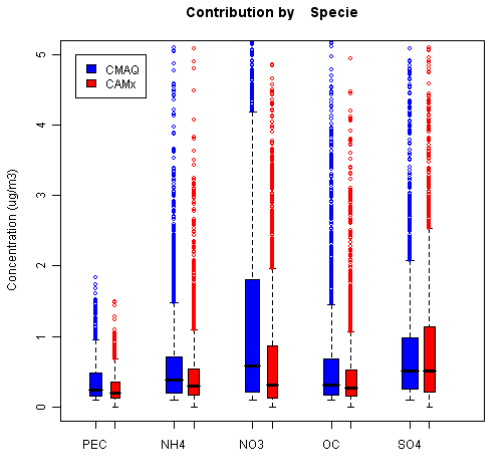

Figure 9. Chemically speciated PM2.5 contributions

In general, CMAQ tends to estimate more ammonium nitrate than CAMx and as a result higher contributions of nitrate and ammonium are seen at receptors across the modeling domain. Table 2 contains the coefficient of determination (r^2) and coefficient of variation (CV) of CMAQ and CAMx source contribution estimates by specie over all receptors in the Milwaukee region and days modeled (N=20,111).

Table 2. Source contribution relationships between CAMx and CMAQ estimates

Specie	r^2	CV
$SO4^=$	0.82	128
$NO3^-$	0.59	218
$NH4^+$	0.78	132
EC	0.89	91
OC	0.93	97

*N = 20,111

CMAQ and CAMx show the best agreement for species dominated by primary emissions like elemental carbon and primary organic carbon. The poorest agreement is for PM2.5 nitrate ion, which is likely due to differences in model formulation for nitrate aerosol formation and exacerbated by differences in advection and deposition as it tends to be secondarily formed specie that is more likely to transport in from nearby source regions.

Figure 10 shows the r^2 value comparing the relationship between CMAQ and CAMx source contribution estimates to the Milwaukee/Waukesha monitors from each of the 13 source regions. Washington County (region=2) contribution to these receptor locations ranges between 0.6 and 0.8. For most source regions, nitrate has the weakest relationship between models. Boundary condition estimates are fairly similar between models (N=1,547; r^2=0.66; CV=54).

Boundary condition contribution and contribution from all non-tagged emission in the modeling domain have a similar relationship between modeling systems as the tagged local county group (tags 1-11) at the Milwaukee/Waukesha receptors.

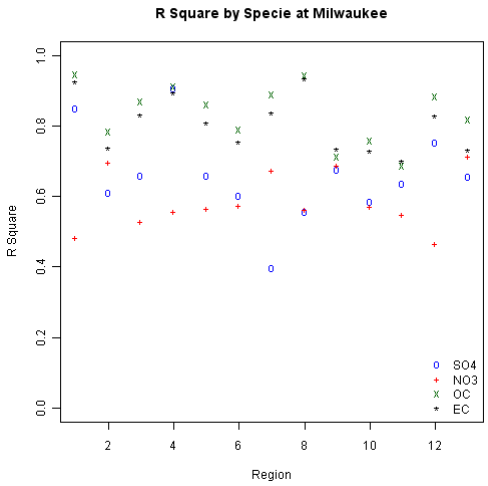

Figure 10. R-Square estimates for each specie by region to Milwaukee/Waukesha receptors (1-11 county tags; 12=non-tagged; 13=boundary conditions).

The primary source contributions have the strongest relationship between models, especially from the closest source regions (counties). Relationships between model estimates even for primary species weaken as the distance between source and receptor increases. This is likely due to differences in model formulation for advection and deposition processes.

The unpaired domain maximum 24-hr averaged initial condition contribution for each of the PM2.5 species over all grid cells in the modeling domain is shown in Figure 11 for the first 7 days of each quarter.

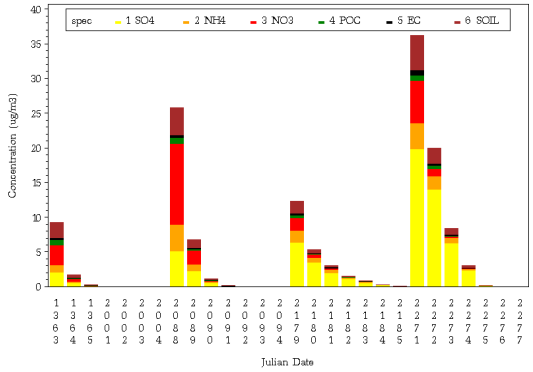

Figure 11. Domain maximum 24-hr average initial condition contribution for first seven days of each quarter

Total 24-hr averaged PM2.5 concentrations from initial conditions reduce below 1.0 ug/m^3 after 4 simulation days in each quarter. This suggests that 4 to 5 simulation days would be necessary to remove the influence of start-up conditions on model estimates of PM2.5.

4. CONCLUSION

CAMx and CMAQ are fairly consistent with absolute model estimates of PM2.5 species, with the exception of nitrate ion, of which CMAQ tends to predict higher concentrations. The modeling systems also tend to have strong relationships in predicted source contributions. Again, nitrate ion source contribution between models has the weakest relationship of the species examined in this study. Source contribution to the Milwaukee/Waukesha area tends to be largely local on the highest 10% of modeled days. The largest contributing species on the top 10% of modeled days include primarily emitted species including primary organic carbon and elemental carbon. Contributions from source areas furthest away from the Milwaukee/Waukesha area tend to be dominated by secondarily formed species such as nitrate ion and sulfate ion. Spatial peak contributions and extent of influence appear to be very similar between modeling systems. Despite differences in model formulations, the source contributions estimated by each modeling system compare well with each other. This increases the confidence that each is appropriately implemented and suitable for the estimation of source contribution.

5. REFERENCES

Aiyyer, A, Cohan, D., Russell, A., Stockwell, W., Tanrikulu, S., Vizuete, W., Wilczak, J., 2007. Final Report: Third Peer Review of the CMAQ Model. p. 23.

Boylan, J. W., Odman, M. T., Wilkinson, J. G., Russell, A. G.: Integrated assessment modeling of atmospheric pollutants in the southern Appalachian Mountains: Part II. fine particulate matter and visibility. Journal of Air & Waste Management Association. 56; 12-22: 2006.

Byun, D.W., and Schere, K.L., 2006. Review of the Governing Equations, Computational Algorithms, and Other Components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, J. Applied Mechanics Reviews, 59 (2), 51-77.

Chang, J.S., Brost, I.S., Isaksen, A., Madronich, S., Middleton, P., Stockwell, W.R., Walcek, C.J. 1987. A Three-Dimensional Eulerian Acid Deposition Model: Physical Concepts and Formulation. J Geophys. Res., 92, 14, 681-14,700.

Community Modeling & Analysis System (CMAS) – Reports from the CMAQ Review Process can be found at: http://www.cmascenter.org/r_and_d/cmaq_review_process.cfm?temp_id=99999 .

Dennis, R.L., Byun, D.W., Novak, J.H., Galluppi, K.J., Coats, C.J., and Vouk, M.A., 1996. The next generation of integrated air quality modeling: EPA's Models-3, Atmospheric Environment, 30, 1925-1938.

ENVIRON International Corporation. 2008. User's Guide Comprehensive Air Quality Model with Extensions (CAMx4) Version 4.50. ENVIRON International Corporation, Novato, California. www.camx.com

ICF International, 2007a. Implementation of Sulfur and Nitrogen Tagging in the Community Multiscale Air Quality (CMAQ) Model: Technical Description and Users' Guide. January 15, 2007.

ICF International, 2007b. Implementation of Ozone and Particle Precursor Tagging Methodologies in the Community Multiscale Air Quality (CMAQ) Model: Technical Description and User's Guide. November 20, 2007.

Nenes , A., Pandis, S.N., Pilinis C.: ISORROPIA: A new thermodynamic equilibrium model for multiphase multicomponent inorganic aerosols. Aquat.Geoch. 4; 123-152: 1998.

Strum, M., Houyoux, M., Mason, R. Technical Support Document: Preparation of Emissions Inventories For the 2002-based Platform, Version 3, Criteria Air Pollutants. January 2008. Accessed August 15, 2008. http://www.epa.gov/scram001/reports/Emissions%20TSD%20Vol1_02-28-08.pdf

U.S. Environmental Protection Agency, Byun, D.W., and Ching, J.K.S., Eds, 1999. Science algorithms of EPA Models-3 Community Multiscale Air Quality (CMAQ modeling system, EPA/600/R-99/030, Office of Research and Development).

U.S. Environmental Protection Agency, 2005. Technical Support Document for the Final Clean Air Interstate Rule: Air Quality Modeling; Office of Air Quality Planning and Standards, RTP, NC, March 2005 (CAIR Docket OAR-2005-0053-2149)

U.S. Environmental Protection Agency, 2006. Technical Support Document for the Final PM National Ambient Air Quality Standards Rule: Office of Air Quality Planning and Standards, Research Triangle Park, NC

U.S. Environmental Protection Agency, 2007. Regulatory Impact Analysis of the Proposed Revisions to the National Ambient Air Quality Standards for Ground-Level Ozone, July 2007 (EPA document number 442/R-07-008,).

Wagstrom, K. M., Pandis, S. N., Yarwood, G., Wilson, G., Morris, R. Development and application of a computationally efficientparticulate matter apportionment algorithm in a three-dimensional chemical transport model. Atmospheric Environment 42 (2008) 5650– 5659