Development and application of a computationally efficient particulate matter apportionment algorithm in a three-dimensional chemical transport model

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An on-line and an off-line version of a computationally efficient particulate matter source apportionment algorithm have been developed and compared using the three-dimensional chemical transport model PMCAMx. Both versions of the algorithm use source specific-species that track the contributions of source locations or source classes. The two versions showed a good agreement with each other and with more accurate, computationally demanding methods. The off-line algorithm (Particulate Source Apportionment Technology, PSAT) is simpler to implement, has a lower computational cost and is suitable for a range of source apportionment studies. As a first application, PSAT was used to investigate the contribution of power plant SO\textsubscript{2} emissions to particulate sulfate concentrations in the Eastern United States. The impact of the transport of SO\textsubscript{2} emissions from the Chicago, IL area and the impact of these emissions on particulate sulfate concentration in surrounding areas were also studied as a second application. The implementation of PSAT for the SO\textsubscript{2}/particulate sulfate system only resulted in a 1\% increase in computation time over the base simulation. The algorithm provides a computationally efficient platform for the study of pollutant transport and source contributions on regional scales.

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1. Introduction

High concentrations of particulate matter (PM) result in human health problems (Dockery et al., 1993; Schwartz, 1999; Kunzli et al., 2000; Klemm et al., 2000), visibility deterioration (Ying et al., 2004), damage-sensitive ecosystems and play a key role in the energy balance of our planet (Seinfeld and Pandis, 1998). In order to effectively regulate emissions of PM and its precursors, it is necessary to understand the contributions of source classes (i.e., source types or locations) to PM concentrations in different locations (EPA, 2005). Chemical transport models (CTMs) provide a useful platform for the study of source contributions to both secondary and primary pollutant concentrations because they explicitly simulate the corresponding atmospheric processes: emissions, transport, removal, chemistry, and aerosol physics.

There have been several applications of PM source apportionment schemes to CTMs. The simplest approach is known as the brute-force method where impacts of sources are quantified by perturbing emission sources one-by-one, and calculating the changes in pollutant...
emissions resulting from the perturbation. This method is computationally inefficient because it requires running the model separately for each parameter and for each perturbation of interest. The brute-force approach quantifies the direct and indirect effects of an emissions reduction, that is, what would happen to the pollutant of interest and all other pollutants if the emission rate of that source was changed to zero (or to another value). This information is useful for the final design of emission control strategies. For non-linear systems, this information, is different than the current contribution of this source to the pollutant concentrations in the given receptor area (Seinfeld and Pandis, 1998; Blanchard, 1999). The direct decoupled method (DDM) can be used to study the local sensitivity of pollutant concentrations to changes in different input parameters, including emissions (Dunker, 1981). DDM is relatively computationally efficient (Hakami et al., 2003) but only provides local derivatives of contributions which may or may not be applicable for large (e.g., above 50%) perturbations (Dunker et al., 2002). McHenry et al. (1992) developed the Tagged Species Engineering Model (TSEM) to examine source–receptor relationships for particulate sulfate using output data from the Regional Acid Deposition Model (RADM). TSEM tracks five chemical species—all related to SO\(_2\) and particulate sulfate concentrations. Lane et al. (2007) developed a source-resolved model to track source contributions of primary PM which involves splitting the emission inventory into \(n\) inventories, one for each source, and the simulation is repeated \(n\) times, once for each source. This simple technique cannot be applied to secondary species as chemical reaction rates are often non-linear with respect to species concentrations.

The most accurate approach for tracking source contributions is the source-oriented external mixture (SOEM) method described by Ying and Kleeman (2006). SOEM divides each pollutant of interest into different source-specific species and tracks these source specific-species separately through the model. SOEM has the ability to accurately track source contributions to pollutant concentrations since its only assumption is that all PM species can be traced back to a specific precursor gas species or primary PM component. This method is computationally demanding since it requires the solution of an increasing number of stiff, coupled differential equations for each time step in the gas and aerosol chemistry modules and also, more importantly, an increased number of species in the aerosol calculations depending on the number of sources to be tracked since a separate set of species must be used for each source. Also adding to the computational demand, SOEM uses dynamic calculations and an externally mixed aerosol, a more thorough approach than the equilibrium approach used in this work but also much more computationally demanding. Another important factor that strongly impacts the required computation time is the number of aerosol size bins in the size distribution. To track contributions of ten source classes to primary species concentrations, SOEM required 97 CPU s simulation cell\(^{-1}\) simulation h\(^{-1}\) on a 466 MHz processor (Kleeman and Cass, 2001).

Despite the above progress an accurate and computationally efficient general technique for studying source contributions to PM in 3-D CTMs is still lacking. In this work, we describe and test two versions of an apportionment algorithm, the On-line Particulate Source Apportionment (OPSA) algorithm and the off-line Particulate Source Apportionment Technology (PSAT) algorithm. Preliminary applications of the algorithm to quantify contributions of different sulfur emission sources and regions to SO\(_2\) and particulate sulfate concentrations and the sulfate size distribution at various receptor areas are also presented.

2. Model description

The model used in these studies is PMCAMx, a regional three-dimensional (3-D) Eulerian CTM. PMCAMx was applied to a 3492 × 3240 km\(^2\) domain in the Eastern United States for the period from 12–19 July 2001 using a 36 × 36 km\(^2\) grid resolution and 14 vertical layers extending up to 6 km. For gas-phase chemistry, the Carbon Bond Mechanism version 4 (CB-IV) (Gery et al., 1989), which simulates 34 gas and 12 radical species, was utilized and the system of reaction rate equations was solved using the chemical mechanism compiler (CMC) of Environ (2004). CMC solves the differential equations describing gas-phase chemistry using a hybrid approach: the equations of species with short lifetimes (i.e., radicals) are replaced by algebraic equations using the pseudo-steady state approximation, the equations of the species with intermediate lifetimes are solved using a second-order implicit Runge–Kutta method and finally the equations for the slow-reacting species are solved explicitly (Environ, 2004). For the aerosol calculations, 10 logarithmically spaced size bins were used (ranging from 40 nm to 40 \(\mu\)m) to represent the size distribution. The thermodynamic bulk equilibrium approach and the internal mixture approximation are used for the aerosol calculations (Capaldo et al., 2000). Nucleation and coagulation were not included in the model, because they have a negligible impact on the PM mass concentrations and mass size distributions simulated here. The meteorological inputs were created using MM5 (Grell et al., 1995). The emissions inventory used was the Base E inventory from the MRPO (LADCO, 2003). Gaydos et al. (2007) and Karydis et al. (2007) have evaluated PMCAMx performance for this period against observed values. Gaydos et al. (2007) found fractional errors smaller than 50% for sulfate, ammonium and organic carbon and Karydis et al. (2007) reported good agreement between predicted and observed temporal PM concentration profiles in all four seasons with the a fractional error of less than 50% and fractional bias of less than 30% for the July 2001 time period used in this work.

3. Source apportionment algorithm description

Current CTMs mix all emissions from different sources together, resulting in the loss of information about source contributions to pollutant concentrations. Source apportionment requires splitting the pollutants from different sources or locations and calculating pollutant
concentrations from each source class while maintaining realistic computation times.

In CTMs like PMCAMx, simulation of atmospheric transport consumes approximately 10% of the computation time while gas-phase chemistry and aerosol dynamics consume the remaining 90% of the time. Since the advection and turbulent dispersion equations are a system of non-coupled, non-numerically stiff differential equations, the computational requirements for their solution scale approximately linearly with the number of species (Colella and Woodward, 1984). On the other hand, the computation requirements for the dynamic aerosol calculations (e.g., condensation/evaporation) increase super-linearly with the number of species because the corresponding differential equations are coupled and numerically stiff. As a result, maintaining reasonable computational demands requires minimal increases in the number of equations in the aerosol calculations.

The source apportionment task for PM is simplified by linking all secondary PM components directly to a specific precursor gas species (e.g., particulate sulfate to SO2, nitrate to NO3, secondary organic aerosol to the corresponding VOC precursors), for this reason, the proposed algorithm does not account for indirect effects such as an increase in nitrate concentrations that can result from sulfate reductions. Algorithms that study the sensitivities, rather than contributions, such as DDM can be used to determine these indirect impacts. The proposed algorithm is based on the fact that all molecules of a particular species have the same probability of reacting at a given location and time regardless of their source (e.g., SO2 emitted by power plants reacts identically to SO2 emitted by oil-refining facilities at a given point and time). PMCAMx, like many CTMs, utilizes operator splitting and we are able to exploit the separation of the different processes by developing a source apportionment algorithm that treats apportionment separately for each process. The algorithms proposed here require splitting of the emissions from the gas-phase chemistry in the operator splitting scheme.

3.1. On-line particulate source apportionment (OPSA) algorithm

The OPSA algorithm treats each source specific species separately during the emissions, transport, wet and dry deposition calculations but combines them into one species before the gas-phase chemistry and aerosol calculations. In order to track these separate sources, the emissions from the sources of interest must be separated out in the emissions processing step before they are input into the model. This will often require reprocessing of the input emissions. The flow of information in the algorithm in PMCAMx is shown in Fig. 1 using SO2 and sulfate as an example.

3.2. Treatment of gas-phase reactions

The treatment of gas-phase reactions is the same as in the Ozone Apportionment Technology (OSAT) described by Dunker et al. (2002) and has been modified for the reactions that are relevant to PM formation. In order to explain the treatment of gas-phase reactions let us first consider the case of gas A reacting to form gaseous species B

\[
A(g) + \text{oxidant} \rightarrow B(g) + \text{other products}
\]

Since the apportionment of the precursor gas does not change as a result of gas-phase chemistry, the concentration of species A from any source after the gas-phase chemistry time step would be given by

\[
c_{A,i}(t + \Delta t) = c_{A,i}(t) + \left[ c_A(t + \Delta t) - c_A(t) \right] \frac{c_{A,i}(t)}{c_A(t)} \tag{1}
\]

where \(c_{A,i}(t + \Delta t)\) and \(c_{A,i}(t)\) are the concentrations of species A from a given source at the end and the beginning, respectively, of the gas-phase chemistry time step and \(c_A(t + \Delta t)\) and \(c_A(t)\) are the corresponding total concentrations of species A.

Molecules of A from all sources have the same probability of reacting; therefore, in a time step \(\Delta t\) the concentration of B produced from A from source \(i\) would be proportional to the total B produced during the time step, \(c_B^i(t + \Delta t) - c_B^i(t)\), and the fraction of A coming from source \(i\), \(c_{A,i}(t)/c_A(t)\), in the beginning of the time step. As a result of this, the concentration of species B from any source after the gas-phase chemistry time step would be given by

\[
c_{B,i}(t + \Delta t) = c_{B,i}(t) + \left[ c_B^i(t + \Delta t) - c_B^i(t) \right] \frac{c_{A,i}(t)}{c_A(t)} \tag{2}
\]

where \(c_{B,i}(t + \Delta t)\) and \(c_{B,i}(t)\) are the concentrations of species B from a given source at the end and the beginning, respectively, of the gas-phase chemistry time step; \(c_B^i(t + \Delta t)\) and \(c_B^i(t)\) are the corresponding total concentrations of species B from all sources. One assumption inherent in this approach is that each product can be
linked to one of the precursor gases. Eqs. (1) and (2) are applied to the apportionment of $H_2SO_4(g)$ from $SO_2$, $HNO_3(g)$ from $NO_3$, and different condensable vapors from the corresponding VOC precursors. Use of the above approach also requires splitting of the addition of emissions from the gas-phase chemistry calculations.

### 3.3. Treatment of irreversible partitioning

The apportionment of condensing non-volatile PM material (e.g., sulfate and non-volatile SOA components) after the partitioning calculations is similar to that described above for gas-phase species after gas-phase chemistry calculations. So, for the general condensation of a non-volatile gaseous species

$$ B(g) \rightarrow C(p) $$

where $C$ is the particulate form of $B$ (e.g., sulfate and sulfuric acid). The apportionment of the precursor gas does not change due to the transfer of $B$ to the particulate phase. The concentration of the remaining precursor gas species $B$ from source $i$ is given by

$$ c_{B,i}(t + \Delta t) = c_{B,i}(t) + \left[ c_B(t + \Delta t) - c_B(t) \right] \frac{c_{B,i}(t)}{c_B(t)} \quad (3) $$

Similarly to the case of gas-phase chemistry, the concentration of the secondary particulate species ($C$) is then based on the apportionment of the precursor gas species and is given by

$$ c_{C,i}(t + \Delta t) = c_{C,i}(t) + \left[ c_C(t + \Delta t) - c_C(t) \right] \frac{c_{B,i}(t)}{c_B(t)} \quad (4) $$

where $c_{C,i}(t+\Delta t)$ and $c_{C,i}(t)$ are the concentrations of species $C$ from source $i$ at the end and the beginning, respectively, of the gas-phase chemistry time step; $c_C(t+\Delta t)$ and $c_C(t)$ are the corresponding total concentrations of species $C$ from all sources.

### 3.4. Treatment of reversible partitioning

A different approach is used for apportioning semi-volatile species (e.g., semi-volatile secondary organic aerosol, ammonium nitrate, ammonium chloride) after partitioning calculations than for non-volatile species (e.g., sulfate) due to the simultaneous condensation and evaporation. Since this version of PMCAMx uses a thermodynamic equilibrium assumption for the partitioning calculations, only the net amount transferred between the gas and particulate phase is known. We assume that because the characteristic time for equilibrium is much less than the time step in the model, the apportionment in the two phases will be equal. This is equivalent to assuming that molecules of a compound from a given source will have the time to distribute themselves proportionally between the two phases. The error introduced by this assumption will be examined in a subsequent section. For a semi-volatile gas (B) condensing to form secondary aerosol (C)

$$ B(g) \rightarrow C(p) $$

the fractions of B and C from source $i$ would be equal to each another and also equal to the apportionment of the sum of B and C before the partitioning calculations. The concentration of B and C attributed to source $i$ after partitioning calculations are then given by

$$ c_{B,i}(t + \Delta t) = c_B(t + \Delta t) \frac{c_{B,i}(t) + c_{C,i}(t)}{c_B(t) + c_C(t)} \quad (5) $$

### 3.5. Treatment of common products of two precursors

Occasionally there is the case where two precursors will produce the same oxidation product. A generalized example of this is

$$ A(g) + \text{oxidant} \rightarrow C(g) + \text{other products} $$

$$ B(g) + \text{oxidant} \rightarrow C(g) + \text{other products} $$

The apportionment of C can then be determined by weighting the apportionment of A and B by the corresponding reaction rates. The appropriate weighting factors would be

$$ z_A = \frac{r_{A \rightarrow C}}{r_{A \rightarrow C} + r_{B \rightarrow C}} \quad z_B = \frac{r_{B \rightarrow C}}{r_{A \rightarrow C} + r_{B \rightarrow C}} \quad (6) $$

where $r_{A \rightarrow C}$ and $r_{B \rightarrow C}$ are the rates for the corresponding reactions. The apportionment of C would then be based on the weighted combination of the apportionment of A and B:

$$ c_{C,i}(t + \Delta t) = c_{C,i}(t) + \left[ c_C(t + \Delta t) - c_C(t) \right] \frac{z_A c_{A,i}(t)}{c_A(t)} + z_B c_{B,i}(t) $$

$$ \frac{c_{C,i}(t)}{c_C(t)} \quad (7) $$

We must also consider the case of two precursors reacting with one another to form a single product of interest, such as the generalized example:

$$ A(g) + B(g) \rightarrow A'(g) + B'(g) $$

Examples of this type include the oxidation of organic vapors by NO3 forming another organic compound and nitric acid. This case is treated similarly to that of one precursor reacting to form one product where each product is matched with a precursor.

### 3.6. Off-line particulate source apportionment technology (PSAT) algorithm

The off-line PSAT algorithm uses the same equations as OPSA but simplifies the treatment of the apportionment
During the transport and removal calculations. A diagram outlining the information flow in PSAT is shown in Fig. 2 for the SO2/sulfate example. PSAT, unlike OPSA, uses the apportionment of the upwind grid cell to apportion species after transport calculations instead of tracking the source specific species separately. The apportionment of the newly transported \( A \) in a cell is based on the apportionment of \( A \) in the upwind grid cell and the fluxes of chemical species between cells as in OSAT (Dunker et al., 2002):

\[
c_{A_i}(t + \Delta t) = c_{A_i}(t) + \Delta c_{in} \times \frac{c_{A_i}(t)}{c_A^i(t)} - \Delta c_{out} \times \frac{c_{A_i}(t)}{c_A^i(t)}
\]

where \( \Delta c_{in} \) is the concentrations transported into the grid cell, \( \Delta c_{out} \) is the concentration transported out of the grid cell, \( c_{A_i}(t) \) is the concentration of species \( A \) from source \( i \) in the upwind grid cell and \( c_A^i(t) \) is the total concentration of species \( A \) from all sources in the upwind grid cell. Since operator splitting in PMCAMx treats advection in the east-west and north-south directions separately, there will be a separate upwind grid cell in each of the two directions. The resulting algorithm runs alongside PMCAMx with the PMCAMx modules sending the flux values for transport and the removal rates to the apportionment algorithm where the actual source tracking takes place.

4. Evaluation of OPSA

As an initial implementation and test both algorithms were applied to the case of SO2 reacting and condensing to form particulate sulfate focusing on two source classes, power plants and all remaining sources. Fig. 3 shows the spatial variation of the SO2 emitted from power plants and emitted from all other sources. The domain average contribution to emissions from power plants was 77%. Large amounts of SO2 are emitted from coal-burning power plants along the Ohio River Valley and north-eastern coast. There are a considerable number of non-power plants SO2 sources in south-eastern Texas and the West Virginia panhandle. PMCAMx with the OPSA algorithm is used to calculate the source contributions to SO2
and particulate sulfate. The results were tested for mass balance against the base case. The predicted total $\text{SO}_2$ (sum of $\text{SO}_2^{\text{Power Plants}}$ and $\text{SO}_2^{\text{Other}}$) on a cell-by-cell basis agreed with the base case to within 0.1 ppb and the particulate sulfate agreed within 0.05 $\mu g m^{-3}$. For $\text{SO}_2$, 99.9% of the OPSA predicted values were within 10% of the base case, 99.6% were within 5%, and 86% were within 1%. For particulate sulfate, all of the OPSA predicted values were within 10% of the base case, 99.97% were within 5%, and 80% were within 1%. The domain-wide error was 0.3% for $\text{SO}_2$ and 0.5% for particulate sulfate. The slight differences are likely caused by errors through numerical diffusion.

The error introduced by the assumption regarding the partitioning of semi-volatile compounds was tested by comparing the apportionment of SOA given by OPSA with that of a simulation tracking the source specific species separately throughout the model (similar to but not the same as SOEM because the aerosols are still internally mixed), a more accurate but computationally expensive approach. Both methods in this comparison use the bulk equilibrium assumption for partitioning calculations. For the purpose of this test, four organic precursors and four corresponding SOA species were used. Four cases were tested and for each case all the species were given identical physical properties (respectively): molecular weights of 150, 150, 150 and 180 g mol$^{-1}$; saturation concentrations of 0.023, 0.674, 0.007 and 0.008 $\mu g m^{-3}$ (at 281 K); and enthalpies of vaporization of 156, 156, 0 and 0 kJ mol$^{-1}$. While this will not give concentrations that are equivalent to what we would expect in the atmosphere, it will allow us to test the ability of our assumption to represent the partitioning of semi-volatile species. The actual SOA calculations in PMCAMx use four SOA classes with different physical properties. The SOA fraction is the fraction of the total SOA that each SOA species represents. The comparison of the SOA fraction predicted by the two approaches on a cell-by-cell basis is shown in Fig. 4 for test case #1. A 99.5% of the cells had OPSA SOA fractions within 10% of the value predicted by tracking the species separately, 99.1% were within 5% and 97% were within 1%. The root-mean-square error of the predicted SOA fraction is below 0.0002. The remaining three test cases showed even better agreement than the case above. These small differences suggest that our assumption (see Eq. (5)) about the partitioning of the semi-volatile aerosol components introduces small errors in a range of cases.

The implementation of OPSA to the case of the two $\text{SO}_2$ and particulate sulfate sources resulted in a 2% increase in CPU requirements while the use of PSAT resulted in an increase of less than 1%. The base model requires approximately 0.006 CPU s simulation h$^{-1}$ cell$^{-1}$ at 1.2 GHz. Tracking 10 sources using OSPA requires approximately 0.012 CPU s simulation cell$^{-1}$ simulation h$^{-1}$, while an existing algorithm like SOEM that uses practically no simplifying assumptions requires more than 97 CPU s simulation cell$^{-1}$ simulation h$^{-1}$ on a 466 MHz processor to track the same 10 source categories (Kleeman and Cass, 2001). It is also important to note that the underlying model with SOEM uses few simplifying assumptions compared to PMCAMx with PSAT (e.g., simulates mass transfer instead of assuming equilibrium).

5. Evaluation of PSAT

Since the piecewise parabolic method (PPM) (Gery et al., 1989) advection scheme with monotonic adjustments used in PMCAMx is a second-order scheme and the PSAT apportionment algorithm is linear, it is necessary to test its ability to adequately represent apportionment changes due to advection. This testing can be performed by comparing the results of PSAT against that of the more

![Fig. 4. Comparison of predicted SOA fractions from OPSA and a simulation tracking each SOA source-related species separately for four different SOA sources. There are approximately 35,000 data points in the above comparison.](image)
accurate OPSA. For this test we once more use the $\text{SO}_2/$ sulfate system studying power plants and all remaining sources.

Fig. 5 shows the comparison between the apportionment result from OPSA and PSAT for $\text{SO}_2$ and particulate sulfate on a cell-by-cell basis. For the $\text{SO}_2$ 99.8% of the PSAT predicted values fell within 10% of the values predicted by OPSA, 98% fell within 5%, and 75% fell within 1%. For particulate sulfate 99.4% of the PSAT predicted values fell within 10% of the value predicted by OPSA, 99.4% fell within the 5%, and 76% fell within 1%. The domain-wide errors for $\text{SO}_2$ and particulate sulfate fractions are 0.16% and 0.15%, respectively, and the biases are 0.0012 and 0.0011, respectively. These relatively small differences throughout the domain suggest that the errors introduced in PSAT by the non-linearity of the advection algorithm are negligible in most cases. These small errors are a good trade-off for the gain in computation time and simplicity of implementation (outside PMCAMx). PSAT is used for further applications in this paper.

6. Results

The concentrations of $\text{SO}_2$ and particulate sulfate attributed to power plants and other sources are shown in Fig. 6. The model predicts high concentrations of $\text{SO}_2$ from power plants along the Ohio River Valley and the north-eastern coast (Fig. 6a). There are also areas of high $\text{SO}_2$ concentrations from non-power plant sources (e.g., chemical manufacturing and oil refining) in the West Virginia panhandle, east Texas and Chicago (Fig. 6b). Areas with high $\text{SO}_2$ concentrations from power plants also have high particulate sulfate concentrations from the same
sources. There are also areas where power plants contributed substantially to particulate sulfate levels during the test period (e.g., Wisconsin, Kansas, Nebraska, Oklahoma and Alabama) even though there are few power plants in the immediate vicinity (Fig. 6c). This can be attributed to the long-range transport of pollutants into these areas from areas with larger numbers of power plants.

Fig. 7 shows the fraction of SO2 and PM2.5 sulfate that were attributed to SO2 from power plant emissions for this test period. A large fraction of SO2 in Maine, New Hampshire and Vermont is attributed to power plants (up to 95%) as a result of the prevailing winds from the Ohio River Valley and New England coastal areas. A small fraction (as low as 10%) of the SO2 in much of Texas is attributed to power plants, with SO2 resulting instead from the refineries and chemical industries along the Texas coast. Lower fractional contributions of power plant emissions to SO2 are also seen in the West Virginia panhandle, the South Carolina coast and eastern Tennessee/western Virginia due to extensive chemical processing in these areas. The source apportionment for particulate sulfate shows that SO2 emitted from power plants can contribute significantly to particulate sulfate in areas far removed from large power plants and the effects of power plant emissions on particulate sulfate are more distributed than the effects on SO2 (Fig. 7). While SO2 emitted from power plants has little impact on the SO2 concentrations in areas such as Texas and the West Virginia panhandle, these emissions still contribute significantly to the particulate sulfate concentrations in these areas. The overall average contribution of power plant emissions to ambient concentrations of SO2 and particulate sulfate are 76% and 77%, respectively, similar to their emissions but, on the other hand, their spatial pattern is quite different.

As an initial application of the ability of PSAT to track the contributions from source regions, the impact of SO2 emissions from the Chicago, IL–Gary, IN area (the region shown in Fig. 8) was studied for the period from 12–19 July 2001. The average concentration of SO2 originally from this source region is shown in Fig. 9a. The Chicago SO2 emissions for this period influence the neighboring areas (within a radius of approximately 100 km) but the particulate sulfate produced from these emissions can be transported over a much wider area (Fig. 9b). While winds transport SO2 into the surrounding areas, it also reacts to form particulate sulfate which can then be further transported (it has a relatively long lifetime) having a wider impact in the domain significantly impacting areas as far as 500 km from the source region.

**Fig. 6.** Average concentrations of SO2 and PM2.5 sulfate contributed by power plants and other sources: (a) SO2 from power plants (ppb), (b) SO2 from remaining sources (ppb), (c) PM2.5 sulfate from power plants (µg m⁻³) and (d) PM2.5 sulfate from remaining sources (µg m⁻³).
Fig. 10 shows the size-resolved apportionment for particulate sulfate in Pittsburgh, PA and Houston, TX for this 12–19 July 2001-time period. In Pittsburgh, approximately 65–80% of sulfate in all size bins is the result of power plant emissions. In Houston, the apportionment is more varied across the size distribution as a result of the large number of non-power plant sources (mainly refineries) in the vicinity of the city and the more distant power plants. The higher contributions of power plant sources to the larger size bins are explained by the transport of PM into the region from regions where power plants have a greater impact. These larger particles have on average been in the atmosphere for a longer time. This illustrates an interesting link between the size distribution of the particulate sulfate from each source and the location of the source.

7. Conclusion

Both OPSA and PSAT are computationally efficient and predict the same total concentrations as the original model. PSAT was found to be more computationally efficient and simpler to implement. PSAT provides a convenient platform for the study of pollutant transport...
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