COMPARISON OF THREE SECONDARY ORGANIC AEROSOL ALGORITHMS IMPLEMENTED IN CMAQ

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1.0 INTRODUCTION

CMAQ versions currently in use contain three different secondary organic aerosol (SOA) algorithms. For simplicity, they are referred to here as Pandis', Odum's, and Schell's algorithms. In this paper, we show major implementation details of the three SOA algorithms in CMAQ, and present our analysis of the impacts of the SOA algorithms on organic aerosol modeling results.

2.0 THE THREE CMAQ SOA ALGORITHMS

General steps in the calculation of SOA formation in CMAQ are partly documented by Binkowski and Roselle (2003), Binkowski and Shankar (1995), USEPA (1999), and are analyzed in detail by Jiang and Roth (2003). Major differences in the three CMAQ SOA algorithms are the methods that they use to calculate mass formation rates of anthropogenic and biogenic SOA.

In Pandis' algorithm (Pandis et al., 1992; Bowman et al., 1995), constant aerosol yields for six pseudo SOA precursor species are used to convert mass production rates of the pseudo SOA precursor species in gas phase to the mass production rates of anthropogenic and biogenic SOA.

In Odum's algorithm (Odum et al., 1996), anthropogenic and biogenic aerosol yields are calculated as functions of total organic aerosol mass concentrations. The aerosol yields are then used to convert mass production rates of four pseudo SOA precursor species in gas phase to the mass production rates of anthropogenic and biogenic SOA.

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In Schell's algorithm (Schell et al., 2001), a system of nonlinear equations is constructed for gas/aerosol partitioning of 10 condensable species. The Clausius-Clapeyron equation is used to convert vapor pressures of the condensable species under a reference temperature of 298.0 K to the values under a current temperature T. The system of equations is solved by a numerical solver based on Newton's method (Press et al., 1992). The solution of the system of equations is treated as the new SOA formed during a time step, and is used to calculate the mass production rates of anthropogenic and biogenic SOA.

3.0 MODEL SET-UP

3.1 The Model

CMAQ version 4.1 was used as a base model for this study. The structure of the aerosol module AERO2 was modified so that each SOA algorithm was implemented as a replaceable sub–module of the aerosol module (Jiang and Roth, 2002). Three CMAQ executables were built by changing the SOA algorithm only and keeping the science and code for all other aerosol processes the same. Since Schell's algorithm was released as a part of the AERO3 module in CMAQ version 4.2, it was extracted from AERO3 and reorganized as a sub–module in AERO2.

3.2 Modeling Domains and Period

The nested horizontal grid domains focus on the Canadian Lower Fraser Valley and cover part of the Pacific Northwest in the US and southwest British Columbia in Canada. The outer domain contains 25×44 grid cells with 15km resolution. The inner domain contains 33×30 grid cells with 5km resolution. Model simulations were conducted for the period of 31 July to 7 August 1993 during the Pacific '93 field study (BC Environment, 1994).

Details of the modeling domains and model input data are available in Roth et al. (2003).

4.0 IMPACT OF THE SOA ALGORITHMS ON ORGANIC AEROSOL MODELING RESULTS

The three CMAQ executables were run on the outer domain for this part of the study.

4.1 Impact on SOA Spatial distribution

Time–averaged spatial distributions of SOA mass concentrations in the modeling domain generated by the three SOA algorithms are significantly different, both qualitatively and quantitatively. Among the three SOA algorithms, Schell's and Odum's algorithms show the strongest and the weakest capability in producing SOA, respectively. For anthropogenic SOA, the maximum time–averaged concentrations in the modeling domain generated by the Schell, Pandis, and Odum algorithms are 3.87, 0.31, and 0.004 µg m^{-3}, respectively. Corresponding values for biogenic and total SOA are 6.19 and 10.06 µg m^{-3} for Schell, 2.71 and 2.78 for Pandis, and 0.04 and 0.05 for Odum's algorithm. The maximum total SOA concentration due to Schell is 210 times the value due to Odum.

Correlation analysis of the time–averaged SOA concentrations in all grid cells shows that Schell's algorithm correlates reasonably well with Pandis' algorithm, while there are only limited correlations between either Schell's or Pandis' algorithm and Odum's algorithm. The slope of the Schell vs. Pandis SOA concentration trend–line is 3.36, which indicates a distinctively stronger capability of Schell's algorithm in SOA generation. Very high slope values, 178.5 and 61.4 respectively, of the Schell vs. Odum and Pandis vs. Odum trend–lines are caused by extremely low SOA concentrations from Odum's algorithm.

4.2 Impact on SOA Temporal Variation

Domain–averaged temporal variations of SOA concentrations generated by the three SOA algorithms are compared. The comparison shows that Schell's algorithm gives the highest concentrations and consistent temporal variation patterns for anthropogenic, biogenic, and total SOA. For anthropogenic SOA, Pandis' algorithm gives distinctively lower concentrations and different temporal patterns than Schell's algorithm. For biogenic and total SOA, the results of Pandis' algorithm are closer to those of Schell's algorithm, in terms of both concentration levels and temporal patterns. All the concentrations generated by Odum's algorithm are so low that their temporal variation curves almost overlap with the horizontal axis of the temporal variation graphs. As an example, Fig. 1 shows the comparison of the modeled total SOA concentrations.

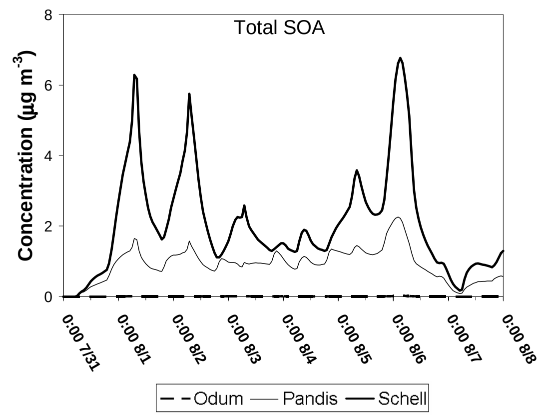

Fig. 1 Temporal variations of total SOA concentrations generated by the SOA algorithms.

Correlation analysis of hourly domain–averaged total SOA concentrations shows similar levels of linear correlations among the three SOA algorithms. As with the grid cell–based correlations discussed in Section 4.1, very high slope values of 109.82 and 338.67 of the Pandis vs. Odum and Schell vs. Odum SOA concentration trend–lines are caused by extremely low SOA concentrations generated by Odum's algorithm. A more reasonable but still high slope value of 2.77 for the Schell vs. Pandis trend–line reveals a stronger SOA–generating capability from Schell's algorithm than from Pandis' algorithm.

4.3 Impact on SOA/Fine Particle Mass Ratios

All three SOA algorithms give somewhat similar time-averaged spatial distribution patterns of SOA/fine particle mass ratios throughout the domain. However, values of the ratios vary significantly among the three SOA algorithms. The ratios reach as high as 79.69% by Schell's algorithm and as low as 0.003% by Odum's algorithm at specific domain locations. On a domain- and time-average basis, the ratios are 30.50%, 18.10%, and 0.11% by Schell, Pandis, and Odum's algorithms, respectively.

5.0 IMPACT OF THE SOA ALGORITHMS ON ORGANIC AEROSOL MODELING PERFORMANCE

For this part of the study, the three CMAQ executables were run on the nested outer and inner domains, while the analysis is focused on the inner domain. Inner domain organic mass concentrations generated by the three SOA algorithms are compared with the ambient organic aerosol measurement data at Pitt Meadows (PIME3) and Chilliwack (CHIL3) stations.

5.1 Comparison of Hourly Modeled Organic Aerosol Concentrations with 24-hour Average Measurements

As an example, Fig. 2 shows the comparison at the PIME3 site. At both sites, Schell's algorithm generates the highest values of organic aerosol concentrations among the three SOA algorithms. The measured 24–hour average concentrations are mostly within the modeled concentration ranges by Schell's algorithm for each of the seven 24–hour periods, with a few exceptions. Modeled organic aerosol diurnal patterns by Pandis' algorithm generally match well with those by Schell's algorithm. However, the Pandis algorithm's results are consistently lower than the results of Schell's algorithm. The Pandis algorithm's hourly results are also mostly lower than the observed 24–hour average numbers. Odum's algorithm significantly and consistently underestimates organic aerosol concentrations. Even the highest hourly modeling results by Odum's algorithm are significantly lower than the observed 24–hour averages for all the time periods in both sites.

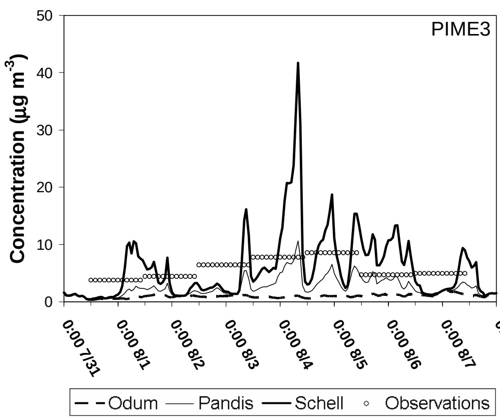

Fig. 2 Comparison of hourly modeled organic aerosol concentrations with measured 24-hour averages.

5.2 Comparison of 24-hour Average Modeled and Measured Organic Aerosol Concentrations

When 24–hour average modeled and measured organic aerosol concentrations are compared, the performance of Schell's algorithm varies depending on the date and location. However, on an episode-average basis, Schell's algorithm results match strikingly well with the measurement data at both sites. Both Pandis' and Odum's algorithms consistently under–predict the 24–hour average organic aerosol concentrations for all the time periods at both sites. However, the performance of Pandis' algorithm is consistently better than that of Odum's algorithm. Fig. 3 shows an example comparison at PIME3.

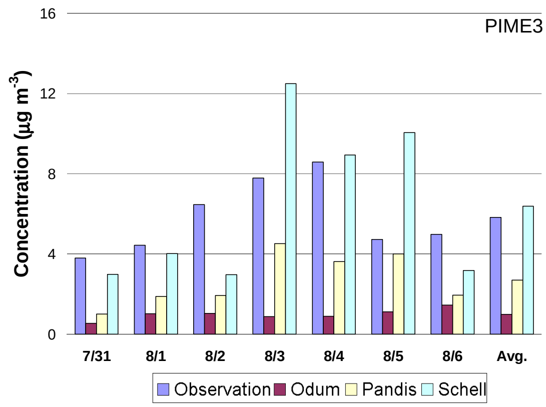

Fig. 3 Comparison of 24-hour average modeled and measured organic aerosol concentrations.

6.0 CONCLUSIONS AND DISCUSSIONS

The three SOA algorithms are substantially different in their science, SOA–generation capabilities, and organic aerosol modeling performance. Among the algorithms, Schell's algorithm is the most sophisticated in science, the strongest in SOA–generation capabilities, and the best in average performance. Odum's algorithm is the weakest in SOA-generation and in model performance, and is not conceptually suitable for use in current CMAQ. Despite its simplicity in science, Pandis' SOA–generation capability and performance is between those of Schell's and Odum's algorithms. Approximately, the SOA-generating capability of Schell's algorithm is three times that of Pandis' algorithm, and the capabilities of Schell's and Pandis' algorithms are orders of magnitude higher than that of Odum's algorithm. While various SOA concentration correlations among the three algorithms are noticeable, both Schell's and Pandis' algorithms correlate poorly with Odum's algorithm in SOA spatial distributions.

This version of Schell's algorithm does not include the concentration of an individual aerosol phase SOA species in the total concentration of the species in the ambient air. This should lead to overestimation of SOA concentrations. The fact that Schell's algorithm did not show overall overestimation as expected implies that other factors in the modeling system may have caused compensating underestimation of organic aerosol concentrations. After the completion of this study, a newer version of CMAQ was released in September 2003. According to the release note, Shell's algorithm has been modified to correct the problem discussed here.

The poor performance of Odum's algorithm was mainly caused by using the overall aerosol yields calculated by Odum's equation. This problem could readily be resolved by adapting an equation, such as the one in Jiang (2003), for the calculation of instantaneous aerosol yields under pre-existing organic aerosol concentration levels.

7.0 ACKNOWLEDGMENTS

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