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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling of organic aerosols over Europe (2002–2007) using a volatility basis set (VBS) framework with application of different assumptions regarding the formation of secondary organic aerosol" *by* R. Bergström et al.

Anonymous Referee #2

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This paper compares the predictions organic aerosols over Europe using four different treatments described previously in the literature. Since the parameters and assumptions used by the VBS framework varies, it is useful to examine the differences in the resulting SOA and total organic matter. The paper is well-written, utilizes a wide range of data to compare with model predictions, and presents material that is suitable to ACP. Nevertheless, there are a number issues that need to be addressed before it is suitable for publication

General Comments:

Most of the plots and statistics average the results over the five-year period (which are useful); however, some discussion on whether there is any yearly variation in the performance of the four treatments is warranted. There is some mention of "important seasonal variations" on page 5445, but without further discussion. In addition, the paper relies mostly on bias as the statistical metric, but it would be useful to include others, such as correlation coefficient when there is enough temporal data.

The amount of material used to describe the model, and some important processes are not discussed as indicated in my specific comments. The model description needs to be modified to address those processes.

The study focuses on using particulate matter data (OC, OM, EC) to evaluate the model. However, gas-phase measurements also provide useful information. Most importantly are known precursors for SOA, such as isoprene and other biogenic emissions. Table 3 indicates that the VBS treatment will depend on high or low NOx regimes, so getting NOx correct will be important as well. Where there no measurements at all of these quantities over the 5-year period? Very little is said regarding gas-phase precursors. Since SOA is often correlated with ozone in the summer time, an evaluation of predicted ozone would shed some additional light on the performance of the model. The authors discuss many of the uncertainties associated with primary particulate emissions – which is important, but neglect discussion on gas-phase chemistry.

Specific Comments:

Acronyms in general: There are too many acronyms used in the manuscript and at times it is difficult to follow the points in the text. The authors should reduce the number of acronyms. Some could easily be written out (e.g. PCM) so that the text would be more readable.

Page 5427, abstract: Most of the acronyms could be written out in the abstract, although I understand that some acronyms are necessary in the main text.

Page 5429, lines 5 – 11: The VBS framework has been widely used and evaluated by many regional modeling studies now. It would be useful to include some references here.

Page 5429, lines 17-20: Emissions are often blamed, fairly or unfairly, on the uncertainties in predictions of particulates. Meteorological factors, which are not mentioned anywhere in the manuscript, affect transport, mixing, secondary formation, dry deposition, and wet removal. If these factors are simulated well by a model, they will also contribute to uncertainties in particulates. Aerosol chemistry also depends on gas-phase chemistry, and SOA is often correlated with ozone formation. Uncertainties in photochemistry likely contribute to SOA as well.

Page 5430, lines 6-9: I agree that comparisons with carbon-14 are important to determine whether models represent fossil and modern sources of carbon. But how uncertain are these measurements? There have been some studies with co-located measurements that indicate significantly different results. There have also been some SOA modeling studies that have already compared those results with carbon-14 data (e.g. Hodzic et al, 20xx).

Page 5430, line 16: MSC-W is not defined.

Page 5430, line 22: A 50 km grid spacing is used, which is very coarse. As discussed in a few places later in the text, it is problematic to compare some of the point measurements with the grid-cell values – especially in urban areas. It is well known that SOA is often correlated with ozone, and ozone concentrations are usually too low near urban sources when a coarse grid spacing is used. So, SOA predictions in this study should be lower than observed at many stations, especially those in the vicinity of large variations in emission rates. If the SOA predictions were close to the observations, a higher grid spacing in the same model would likely produce positive biases which would be

opposite of the conclusions drawn in this study. Some additional discussion regarding resolution and in the implications are needed up front before the results are presented.

Page 5431, end of section 2: The authors need to describe how wet removal is included in the model, which is important for the long simulation periods performed in this study. Accurate predictions of organic aerosol, along with other aerosol species, will also require predictions of precipitation to be well represented. If wet removal is not included in the model for this study, an important pathway of the aerosol lifecycle is not included which affects how well the four organic aerosol treatments perform. Another factor that needs to be mentioned is how lateral boundary conditions are handled.

Page 5431, line 23: The day/night factors are mentioned, but do emissions have a smooth diurnal variation or is it a step function as the text implies. Please be more specific.

Page 5432, line 19: Am I correct to assume that the fire emissions are an 8-day average? Fires are usually more sporadic, and it seems that such a temporal variation will introduce uncertainties into the model simulations.

Page 5436, lines 15-19: The authors only present measurements from one AMS deployment. It would seem that there would be much more data available for the 2002-2007 period (Zhang et al. 2007). Since this paper has few measurements presented, it would be useful to include the comparison in this study. Not sure why it needs to be presented elsewhere.

Page 5440, line 1: Not just the PAA version can lead to overestimations, Shrivastava et al. (ACP, 2011) showed that PAP can also produce too much SOA. Recent laboratory and modeling studies (Vaden et al., PNAS, 2011) have shown that the VBS framework evaporates SOA far too quickly compared to observations. Some additional descriptions of the problems with VBS need to be discussed somewhere in this section. Although there are problems with VBS, there are few suitable alternative approaches that could be used for regional models.

Page 5443, end of section 6.3: This section discusses how OM:OC ratios vary, but fail to describe earlier on how oxygen is handled by the VBS treatments. Every VBS scheme arbitrarily assumes different numbers of oxygen atoms added per VBS bin.

Page 5445, lines 22-26: Please include a correlation coefficient for Figs. A1 and A2. There is quite a bit of scatter in the results.

Page 5447, line 9: It should be relatively straight-forward to check the site location when making a assessment of its proximity to local emission sources.

Page 5448, lines 1-5: While this plot is useful, it is difficult to see any differences among three of the four treatments. This is consistent with the averages over much of Sweden shown in Fig. 3. Why show this station versus another one where there might be larger differences among the four treatments? The reasons to show this site are not stated. Where is this site located? It would be useful to include it on one of the spatial distribution plots.

Page 5449, line 16: This is the first time I see how the boundary conditions for particulates are treated. This needs to be stated earlier in the model description section. How important will long-range transport from North America be in contributing time-varying boundary conditions for Europe? It would seem that coupling the regional model with a global model would provide more realistic variations in particulate matter from long-range transport. Of course, the regional model would then be subject to errors from the global model. But it would be preferred than using constant values over a 5-year period.

Page 5450, line 18: this is the first mention of representativeness of the measurements when comparing to the coarse model. More such discussion elsewhere is needed.

Pages 5451-5452: Much of this discussion regarding performance in summer versus winter on this page is confusing. The text goes back and forth between summer and winter. Why not talk about one season first before moving to the other season? Also,

the tables need to be referred to more frequently. Since the text moves back and forth, it is difficult following which table or parts of the table are being discussed.

Page 5452, line 7: The authors state that OCbb is severely underestimated, but in Table 5, the observations are 0.13-0.28, and the model results are 0.13-0.24. That does not look too low to me.

Page 5457, line 26: Long-range transport as well?

Page 5458, lines 11-12: This sentence could be deleted. It was just mentioned earlier as the last bullet on the previous page.

Figure 3, Should the lower-left panel be labeled “PAPA”? There are 2 panels labeled “PAP”.

Figure 4, Is it possible to have the same scale for all panels? It would enable the reader to more quickly determine the relative contributions of the sources. This would likely require a non-linear scale.

Figure 6, It is difficult to see whether there are any significant differences among PAP, PAPA, and PAA. I suggest having one panel with the observations and 4 lines with the total OC from each treatment. Then have pie charts showing the average components along with the bias and correlation coefficient. An arrow could be used to point to the period where biomass burning is significant. It is hard or next to impossible to see time variations in other components.

Figure 7, Same comment as Figure 6 applies here.

Table A1, The latitude and longitude of the stations are listed here. But it would also be useful to have a plot showing where the stations are.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 5425, 2012.