Overall Comments

J. Pierce (Referee)

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Review of "Impact of model grid spacing on regional and urban-scale air quality predictions of organic aerosol" by Stroud et al.

This paper investigates how model grid size can affect the prediction of organic aerosol. The model uses state-of-the-art representations of primary and secondary organic aerosols. The effect of instantaneous dilution due to coarse resolution was found to have large effects on both primary and secondary organic aerosols.

This paper is well written and of interest to the ACP readership. I recommend it be published in ACP once some minor comments have been addressed.

K. Wesson (Referee)

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The authors examine the effects of grid size on modeled concentrations of primary and secondary organic aerosols. They use the Canadian model AURAMS and describe the updated science incorporated in the model. The paper compares measurements of organic aerosols to modeled concentrations using 42, 15 and 2.5 km grid spacing. The authors conclude that high-resolution grid spacing is important, particularly for capturing the location and magnitude of directly emitted primary organic aerosol emissions in an urban area. They also suggest that the finer grid resolution improves the model's ability to predict secondary organic aerosol concentrations by better representing the location of VOC emissions and better representing the chemical environment in which secondary organic aerosols are produced.

This paper is well written and of interest to the ACP readership. I believe it is a great addition to the current literature on the effects of grid spacing on modeled predictions. I recommend it be published in ACP once minor comments have been addressed.

Specific Comments from Reviewer #1 (Dr. Jeff Pierce)

Page 30352, Line 15: "IVOC emissions, which are not accounted for in the standard national emissions inventories, were calculated by multiplying the POA mass emissions by a factor of 15 1.5, which is calculated assuming that the POA emissions have the volatility basis set (VBS) in Robinson et al. (2007)." Did you mean to say "assuming that the POA emissions have the volatility distribution as in Robinson et al. (2007) within the volatility basis set (VBS). Also, it might be good to say that you are using the VBS and say what volatility bins you are using before this.

Page 30352, Line 16: I find the following confusing, "Emissions in the C* =10ˆ6 μgm−3 and C* =10ˆ5 μgm−3 volatility bins and half of the mass emissions in the C* = 10ˆ4 μgm−3 volatility bin are used to derive the factor of 1.5. It is assumed that the remaining half of the mass emissions in the C* =10ˆ4 μgm−3 bin is included in the POA emissions inventory."

Points (1) and (2). The authors have clarified the section on IVOC emissions and it now reads as follows:

"IVOC emissions are not accounted for in the standard national emission inventories. IVOC mass emissions were calculated by multiplying the POA mass emissions in the emission inventories by a factor of 1.5. In the emissions processing, it is assuming that the model low volatility organic emissions (IVOCs+POA) have the same volatility distribution as in Robinson *et al.* (2007). The factor of 1.5 is derived from the division of the mass in Robinson et al.'s higher volatility bins ($C^*=10^6$ $\mu g\ m^{-3}$ bin, $C^*=10^5$ $\mu g\ m^{-3}$ bin and half the mass in the $C^*=10^4$ $\mu g\ m^{-3}$ bin) by the mass in Robinson et al.'s lower volatility bins (half the mass in the $C^*=10^4$ $\mu g\ m^{-3}$ bin and all the POA mass emission in lower volatility bins.". The model uses the one reaction step, 2-product formulation (Odum et al., 2006), and not the full volatility basis set, as explained further on in the model methodology section.

Page 30356, line 25: "Table 1 lists the...". This should be mentioned during the STN comparison too, right?

Point (3). The authors have clarified the sentence and it now reads as follows:

"Table 1 also lists the evaluation statistics for the comparison with the IMPROVE sites."

Page 30357, line 1: Were the ICARTT2004 and BAQS-MET2007 comparison results published? Is this the Gong et al., 2010b citation thats shown a few sentences later?

Point (4). We have moved the Gong et al (2010b) citation 3 lines earlier after the initial reference to this study.

Page 30357, line 29: Has more effort be put into getting Canadian Emissions correct than US emissions?

Point (5). Both countries emission inventories are processed with the same computer program, SMOKE, so the processing steps are similar. However, the US has detailed emission data at the county level compared to Canada's provincial level. For mobile versions, the U.S. has more road types than Canada and each road type can have different chemical speciation profiles and diurnal profiles. We have identified improvements needed and are working, for example, on the spatial allocation of mobile emissions. For major point sources, speciated VOC emissions are available in Canada for each source, while in the US the VOC speciation is assigned indirectly, through Source Classification codes.

Table 2 and Page 30358, line 22: Can you expand Table 2 to give the statistics for the 42 km and 2.5 km data? This will help quantify some of the points you are making.

Point (6).We have expanded Table 2 to include the 42km grid statistics but expanding the table for 2.5km grid statistics requires a significant amount of additional processing time since the high resolution model runs several times slower than real clock time. We have added the following lines to the text, “The 42-km grid spaced simulations show degradation in model performance for almost all the statistical measures at all three sites. The mean bias worsens by ~1μg m^{-3} in going from the 15-km to the lower resolution 42-km grid spacing for all three sites. The correlation coefficient, R, also decreases from 0.65 to 0.54, from 0.72 to 0.68 and from 0.60 to 0.53 for Harrow, Bear Creek and Windsor, respectively.”

Page 30360, line 9: How do you define “maximum grid-cell PM2.5” is this the highest PM2.5 found anywhere in the domain at any time, the highest PM2.5 found anywhere in the domain once averaged over all times, or something else?

Point (7). The authors have clarified the sentence by adding the word “averaged”:

“However, the higher resolution model simulation predicts considerably higher maximum averaged grid-cell PM2.5 OA mass concentration”.

Figures 5-7: Would be nice to have a 3rd panel that is the ratio of the concentrations at 2.5 km resolution to 15 km resolution.

Point (8). The figures do not have the same number of grid points due to the difference in grid spacing so a ratio calculation was not performed.

Specific Comments from Reviewer #2 (Dr. Karen Wesson)

Page 30353, Lines 11-13: Readers might find it useful to have definitions of ΔHvap, αi and Ki.

Point (1). We have provided a short description of the variables:

Δ*H*vap is the enthalpy of vaporization
α1, α2 are the reaction product stoichiometric coefficients
K1, K2 are the gas-to-particle partitioning coefficients

Table 1: It would be interesting and strengthen the paper to see how the statistics vary with respect to the 42 km grid. Do you see the same results (i.e. under-estimation at STN and over-estimation at IMPROVE sites?) Perhaps you see very similar results and little or no improvement going from 42 to 15km? This would, at least, be interesting to note. Perhaps one must model at a finer scale (e.g. 2.5 km) before seeing improvements in model performance, particularly in an area with a significant concentration gradient.

Point (2). We have done the 42km grid statistics for the STN and IMPROVE sites and the results are now summarized in Table 1. We have added the following text, "Table 1 also lists the model statistics for the 42km grid spaced simulation compared to the STN data. There is degradation in model performance with an increase in model bias, a decrease in correlation slope and a decrease in the regression correlation coefficient, in going to the lower resolution. ".

For the IMPROVE comparison, the text now reads as follows, "The mean ± standard deviation of the IMPROVE 24-h measurements for all eastern sites for the BAQS-Met period was 3.4±2.5 μg m^{-3}. For this comparison the model marginally over-predicts, with a mean bias of +0.12 μg m^{-3} and a RMSE of 2.7 μg m^{-3}. The slope and y-intercept of the best-fit line in Fig. 2 are 0.45 and 2.0 μg m^{-3}, respectively. There was only a small degradation in model performance in going to the 42-km grid spaced simulation relative to the 15-km grid, compared to the IMPROVE data. This likely reflects the 42-km model grid spacing being sufficient to capture spatial and temporal variations for the IMPROVE data, which are from rural sites located in national parks."

Table 2: It would be good to show in the table how the 15km statistics compare to those of the other two grid sizes (i.e. 42 and 2.5 km).

Point (3). We have expanded the table to include the 42km grid statistics (see changes and comments to Reviewer#1 above) but expanding the table for 2.5km grid statistics requires a significant amount of additional processing time since the high resolution model runs several times slower than real clock time.

Page 30358, Lines 3-22: I wasn't really sure how the modeling of the aircraft OA measurements fit into the paper. It didn't really add anything to the spatial resolution discussion and I'm not sure how the model evaluation results support or refute any conclusions about improvements in the model science. Perhaps this should be omitted or tied in better to the conclusions.

Page 30362, Lines 21-23: Again, the model evaluation results for the aircraft data are simply listed but not directly tied into the paper conclusions.

Point (4) and (5). We have added a sentence to the conclusion to better tie the aircraft observations with the rural ground measurements.

"High-resolution model-measurement comparisons with aircraft OA data (2.5 km grid spacing with extractions along flight tracks) yielded a MB of +1.3 μgm^{-3} and a correlation coefficient of 0.51. The majority of the aircraft flight time was during the day and spent characterizing aged air masses with regional-scale spatial variations. As such the model OA biases were the same sign to those calculated with data from the rural IMPROVE sites, although the positive bias is considerably higher with the aircraft data set.