Geosci. Model Dev. Discuss., 8, C1815–C1821, 2015
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Geoscientific Model Development Discussions
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8, C1815–C1821, 2015

Interactive Comment

Interactive comment on "Evaluation of the Community Multiscale Air Quality (CMAQ) model v5.0 against size-resolved measurements of inorganic particle composition across sites in North America" *by* C. G. Nolte et al.

C. G. Nolte et al.

nolte.chris@epa.gov

Received and published: 29 August 2015

Anonymous Referee #2

We thank the referee for the constructive comments on our manuscript. The referee's comments are noted in italics below, followed by our responses.

General comments:

The current paper compared the simulated size distribution of inorganic aerosol components with the observation in USA and Canada. The evaluation of simulated size

distribution is very important for evaluating model performance in simulating impacts of air pollutants on human health and environment. However, long-term and multi-station monitoring of such data is hard to obtain since the MOUDI observation usually needs money and labor. However, the current paper has overcome the limitation by collecting data from the several sets of field campaign across USA and Canada from different years and compared them to one long-term simulation data. The target of the manuscript is well within the scope of the current journal and the manuscript is well written and organized. Therefore, the current manuscript will be accepted after the authors reply to the referee's minor questions and comments, either general or specific, and revise the manuscript, accordingly.

One thing I would like to confirm is about the treatment of hygroscopic growth effects on both measured and simulated aerosol size distribution. My question is at what relative humidity aerosol size distribution was supposed to be measured by the MOUDI system, ambient, room, or even drier than both. Also, is the relative humidity consistent with that used for the derivation of simulated size distribution of chemical components? If the both humidity values are not consistent with each other, it is natural that the measured and simulated size distributions are inconsistent even though the simulation of aerosol size distribution is perfect. Please clarify the humidity measured and used for the calculation.

Because the MOUDI samples are collected at ambient conditions, aerosol water affects particle sizes, and hence on which stages the particles are collected. Aerosol water for all three modes is computed using ISORROPIA II (Fountoukis and Nenes, 2007), which uses Zdanovskii-Stokes-Robinson parameterizations. Aerosol water is included in calculating aerosol modal parameters (i.e., D_g and σ_g), and it is these "wet" diameters and standard deviations that are used in computing the size-composition distributions in the manuscript.

We have modified the text in Section 2.3 to specify that the "wet" modal parameters as output by the CMAQ AERODIAM file are used in the calculations.

GMDD

8, C1815–C1821, 2015

Throughout the manuscript, the authors discussed overprediction or underprediction of the peak diameter but it was not clear if the discrepancy is significant or minor. Are there any statistical measures for predictability of peak (or width) of the modeled size distribution?

Whether the discrepancy between observed and modeled size distributions is significant or minor is a good question, but one whose answer depends on what property of the aerosol is of interest for a particular application. Here we focus on $PM_{2.5}$ mass, and devote an entire section of the manuscript (Section 4) to the related question of the difference between estimating $PM_{2.5}$ using the modeled size distribution versus summing the Aitken and accumulation modes, as typically done with CMAQ. As we acknowledge in the conclusion, this evaluation of the modeled size distributions has focused on mass and only on inorganic species. Studies using predictions of carbonaceous size distributions, such as health impacts in urban areas, or studies of the radiative and optical properties of aerosols, might be more sensitive to errors in modeled size distributions.

Specific comments:

[1] p. 3866, ln. 4: "which includes updates to" updates from which version? 5.0.0, or previous?

The text has been revised to remove the ambiguity; see reply to comment [2] below.

[2] p. 3866, ln. 13: "AERO6" needs reference.

There is no single published reference for AERO6. However, to address the referee's comment, as well as a similar comment by Referee 1, we have revised the text to more clearly describe the version of the CMAQ aerosol model used in the present study:

> The CMAQ model configuration was the same for all simulations, with the only differences being in the year-specific emission and meteorological input data. Aerosols in CMAQ are represented using three lognormal modes—Aitken, accumulation, and coarse (Binkowski and Roselle, 2003).

GMDD
8, C1815–C1821, 2015

Inorganic species in the Aitken and accumulation modes are assumed to be in thermodynamic equilibrium with the gas phase, while gas-particle partitioning between the gas phase and the coarse mode is treated dynamically (Kelly et al., 2010). The secondary organic aerosol formulation in CMAQ has been described by Carlton et al. (2010). The simulations in this study used CMAQ version 5.0.1 with the AERO6 aerosol module, which includes speciation of trace metals (Reff et al., 2009; Appel et al., 2013) and source-specific ratios of organic mass to organic carbon (Simon and Bhave, 2012), and incorporates version II of the ISORROPIA thermodynamic equilibrium module (Fountoukis and Nenes, 2007).

[3] p. 3869, Eq. (2): "Dgv". If Dgv represents geometric volume mean diameter as noted in ln. 21 of p. 3869, $\sqrt{\rho_j}$ is better to be excluded from Eq. (2). If $\sqrt{\rho_j}$ is included in Eq. (2), Dgv could be Dgva, e.g. aerodynamic diameter of Dgv, or something like that.

We have modified Eq. (2) to remove the $\sqrt{\rho_j}$ and rewritten Eq. (3) so that it is clear that the aerodynamic diameter D_{pa} is the independent variable.

[4] p. 3873, ln. 21-24: "The PILS data . . . which partially accounts for the scatter in Fig. 5." It is a little bit ambiguous statement. If MOUDI gives accurate average of PILS and the durations of averaging are common for MOUDI and the simulation, the substantial variation of PILS does not account for the scatter in Fig. 5. Or, did you intend to mention the artifact of MOUDI, namely, chemical reactions occurring on the filter during the long duration, whereas PILS gives more accurate values? Please be specific.

The text has been modified to read: "The PILS data further demonstrate that aerosol concentrations varied substantially on much shorter timescales than could be captured by the integrated MOUDI measurements, which are subject to volatilization losses during sampling, particularly during summer (Lee et al., 2008a)."

GMDD
8, C1815–C1821, 2015

[5] p. 3874, ln. 17-25: How does the model treat K^+ emission from biomass burning? Judging from Eq. (6), 0.0176 ACORS could be the one but is it so? ACORS might include anthropogenic and biomass burning and so the factor 0.0176 for biomass burning K^+ may vary depending on the relative abundance of anthropogenic versus biomass burning particles in each grid cell. Also ACORS is the coarse mode particles but combustion generated K^+ may exist mostly in the fine mode. Does the model consider K^+ emission in the fine mode, which is just missing in Eq. (6)?

In AERO6, emissions of K^+ (and other trace metals) in the fine modes are treated explicitly in the model as described in Appel et al. (2013), based on the $PM_{2.5}$ inventory compiled by Reff et al. (2009). The referee is correct that coarse anthropogenic and biomass burning emissions are assigned to the lumped model species ACORS. ACORS is speciated chemically only in computing droplet pH within aqueous chemistry and prior to calling ISORROPIA to compute coarse mode condensation/evaporation. Since biomass burning is predominantly in the submicron mode, its contribution to ACORS is relatively small. We have modified the text to clarify: In AERO6, accumulation mode emissions from sea spray are chemically speciated into Na^+, Cl^-, SO_4^{2-}, Mg^{2+}, Ca^{2+}, and K^+ components, but coarse mode sea spray cations are lumped into a single species, ASEACAT, for computational efficiency during transport. Similarly, anthropogenic coarse primary emissions are lumped into ACORS, and coarse windblown soil dust is modeled as ASOIL. Concentrations of individual chemical components in the coarse mode are computed from ASEACAT, ASOIL, and ACORS: [equations unchanged]

[6] p. 3875, ln. 17: "the total mass of particles with aerodynamic diameters less than 2.5 μm" Please be a little more specific, e.g. "the total dry mass of particles with ambient aerodynamic diameters less than 2.5 μm". ("50% cut-off diameter is 2.5 μm " may not be needed here, though.)

The referee has a valid point that filters are equilibrated at a relatively low RH prior to being weighed, so that some aerosol water collected under high humidity is evapo-

GMDD
8, C1815–C1821, 2015

rated. However, at least for the U.S. Federal Reference Method for measuring $PM_{2.5}$, particulate water can remain bound to particles even after equilibration, so it is also not correct to say the standard is based only on dry mass. Instead we have modified the text as follows: "In the US, air quality regulations for particulate matter are based on the total mass of particles (after equilibration to room temperature and low humidity) with aerodynamic diameters less than 2.5 μm (Frank, 2006)."

[7] p. 3876, ln. 4: "summer" and "winter" Which months? Please specify.

In general in this paper, "summer" is June, July, and August (JJA); "winter" is December, January, and February (DJF). In this paragraph we are talking about the single year 2002, so "winter" refers to January, February, and December of that year. The text has been revised to clarify this: "The mass-weighted fractions of the accumulation mode and coarse mode in the PM2.5 size range averaged over the summer (June–August 2002) and winter (January, February, and December 2002) months are shown in Fig. 8."

[8] p. 3878, ln. 6-7: "implementation of a new gravitational settling scheme". It appears a gravitational settling scheme in a previous version has been updated to a new one. In this case, "implementation of a gravitational settling scheme" would be better.

The text has been modified as suggested.

[9] Table 3: "Dgv" Is it aerodynamic diameter as defined in Eq. (2) or geometric diameter?

It is the geometric volume mean diameter of the mode, consistent with how the term is defined in the text. We have revised Eq. (2) to remove the ambiguity regarding aerodynamic diameter.

[10] Figure 1 (for readers who are not familiar with American geography): The colored circles were hard to be identified. Can those be replaced by numbers or can numbers be added to the colored circles? Also, showing acronyms of sites in the right column of Figure 1 is helpful since only acronyms were used in the main text.

GMDD

8, C1815–C1821, 2015

We have modified Figure 1 to replace the site names with the acronyms used in the main text and have changed the symbols to make them larger and more distinguishable. The version of the plot in the discussion paper is from an .eps file, but the resolution seems degraded from the .pdf, which we are providing for the revised manuscript. We are also referencing Table 1 in the caption.

Interactive comment on Geosci. Model Dev. Discuss., 8, 3861, 2015.