Evaluation of Updated CMAQ Aerosol Treatments with a Focus on Ultrafine Particles

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Background

While regulatory modeling applications have traditionally focused on particulate mass, applications have expanded to include climate change and health impacts of ultrafine particles. As such, it is important to characterize not only aerosol mass but also the aerosol number size distribution. Here we describe the impacts of two updates to be included in the next major CMAQ release: an updated size distribution of particulate emissions and an updated binary nucleation parameterization.

Model Updates

PM Emissions: Currently $PM_{2.5}$ emissions are distributed into CMAQ's Aitken and accumulation modes according to species dependent modal mass fractions. These fractions and associated emissions size distribution parameters are based on historical measurements known to underestimate ultrafine particles. In an effort to improve upon these outdated measurements and better simulate aerosol size distributions, Elleman and Covert (2010) developed updated particulate emissions distributions based on a review of modern measurements from urban, power-plant, and marine source dominated regions at 4-15 km spatial scales. Here we implement their "urban" PM emissions distribution and modal mass fractions (Table 1).

Table 1. Updated (and original*) parameters for Aitken and accumulation mode particulate emissions. For each mode, D_g = geometric mean diameter by number, D_{gv} = geometric mean diameter by volume, and σ_g = geometric standard deviation.

Mode	% Mass	D_g (nm)	D_{gv} (nm)	σ_g
AKN	10 (EC/OC = 0.1%, other species = 0%)	25 (13)	60 (30)	1.7 (1.7)
ACC	90 (EC/OC = 99.9%, other species =100%)	120 (70)	280 (300)	1.7 (2.0)

*Original base case parameters are given in parentheses. The updated parameters are applied to all emitted, non-sea-salt species that have both Aitken and accumulation mode components.

Nucleation: CMAQ's current binary H_2SO_4-H_2O nucleation scheme of Kulmala et al. (1998) contains errors in the formulation that were corrected in the expanded nucleation parameterization of Vehkamaki et al. (2002). The new parameterization can lead to nucleation rates 1-4 orders of magnitude larger than Kulmala et al. (1998).

Simulations and Data Sources

Multiple model periods and domains were employed in an effort to assess the impacts of the proposed updates. Updates were added to CMAQ v.5.0.2+. Simulations were performed over CONUS at 12-km resolution for April-early June 2002, January 2011, and July 2011 for three cases: (1) a "Base" case, (2) a "PMEMIS" case with emissions updates, and (3) an "EMISNUC" case with emissions + nucleation updates. All model runs were preceded by at least 10 days of spinup. Number size distribution measurements from the Pittsburgh Air Quality Study (PAQS) provide the basis for our evaluation of CMAQ simulated number distributions (Stanier et al., 2004). CSN bulk PM mass concentrations were compared to modeled PM levels to estimate the change in mass concentration performance with the proposed updates.

Table 2. Average Pittsburgh number concentrations for the modeled period April 1- June 5, 2002

Obs or Simulation	Average number concentration (#/cm³)
Observed	22386
BASE	7480
PMEMIS	13300
EMISNUC	18100

+ with additional internal updates beyond the 5.0.2 release version

Modeled Impacts and Observations

Number distributions

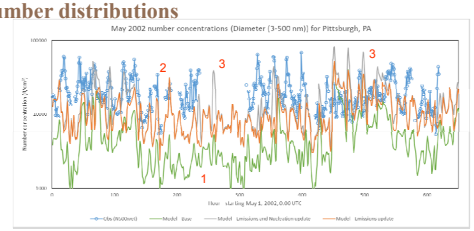

Figure 1. Modeled and observed hourly number concentrations between 3 and 500 nm at the PAQS measurement site for May 2002. [1] Base case modeled number concentrations are consistently and significantly lower than those observed. While the PMEMIS and EMISNUC simulations often lead to similar modeled number concentrations that are higher than the base case values [2], indicating the significant impacts of updated particulate emissions distributions on modeled number concentrations, there are also periods where the update to the binary nucleation parameterization leads to significant increases [3] in modeled hourly concentrations over the emissions updates alone.

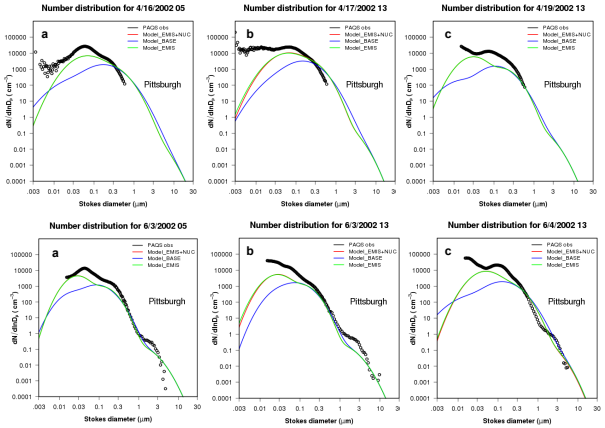

Figure 2. Modeled and observed number size distributions at the PAQS site for the base case, PMEMIS, and EMISNUC simulations (a) before, (b) during, and (c) after regional nucleation events for two periods. The differences between the PMEMIS and EMISNUC distributions here are minimal. The model updates lead to higher number concentrations shifted to lower sizes, closer to observed distributions. During periods with observed regional nucleation events, however, the observed concentrations are still significantly higher with peaks shifted to lower sizes, characteristics that are currently not well captured by the model.

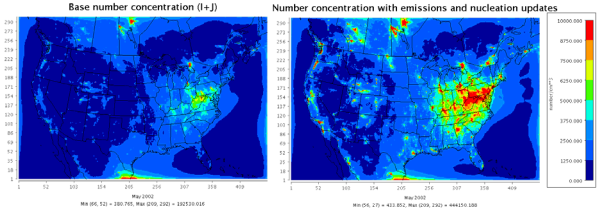

Figure 3. May 2002 average modeled number concentrations (Aitken + accumulation modes) for base case and EMISNUC simulations. The updates can lead to modeled number concentrations many times greater than the base, especially in regions with high SO_2 levels and PM emissions.

Mass concentrations

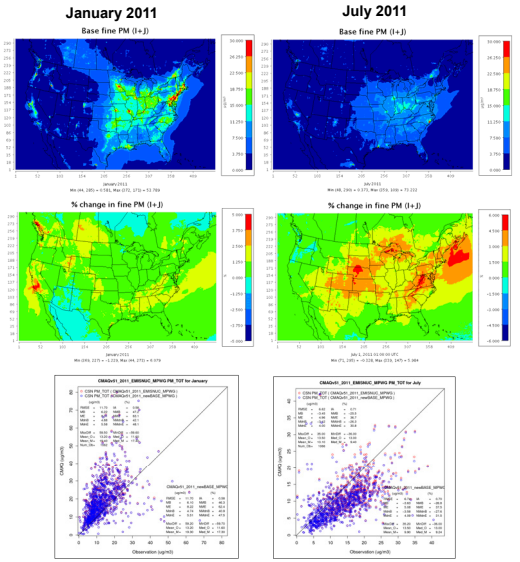

Figure 4. Average base case fine PM for January and July 20ll, the % change introduced with the EMISNUC updates, and the differences in modeled vs. observed PM concentrations for the two cases. As expected, overall mass concentration changes are small (< 5 %) and lead to small changes in model performance for PM mass. *Not shown: the nucleation parameterization update alone leads to only small changes in monthly average SO_4^{2-} concentrations (< 0.02 μg/m³)

Summary and Future Work

- CMAQ's binary nucleation scheme and particulate emissions parameters have been updated and will be released in CMAQ v.5.1.
- Number concentrations are significantly increased with the updates and better match the magnitude and size distribution of observed particles during Spring 2002 of the Pittsburgh Air Quality Study, though the modeled magnitude is typically lower and with a larger peak diameter than observed.
- Mass concentrations are not significantly impacted by the updates; though as we consider feedbacks between air quality and meteorology in the two-way model, impacts on optics and radiation may be more significant and/or varied.
- Future research will include assessing the impact of the updates on direct and indirect aerosol effects with WRF-CMAQ as well as investigating novel nucleation parameterizations that consider the impact of basic species (e.g., amines, NH_3) on nucleation rates, the growth of new particles due to organics, and the addition of a nucleation mode to better capture observed size distributions.

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References: **[1]** Elleman, R. and D. Covert (2010) Aerosol size distribution modeling with the Community Multiscale Air Quality modeling system in the Pacific Northwest: 3. Size distribution of particles emitted into a mesoscale model. J. Geophys. Res., v115, D03204. **[2]** Kulmala et al. (1998) Parameterizations for sulfuric acid/water nucleation rates. J. Geophys. Res., v103(D7). **[3]** Stanier, C. et al. (2004) Ambient Aerosol Size Distributions and Particle Number Concentrations Measured during the Pittsburgh Air Quality Study. Atmos. Env., v38, 3275-3284. **[4]** Vehkamaki, H. et al. (2002) An improved parameterization for sulfuric acid–water nucleation rates for tropospheric and stratospheric conditions. J. Geophys. Res., v107(22).

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