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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ultrafine Particulate Matter Source Contributions across the Continental United States" *by* Melissa A. Venecek et al.

Anonymous Referee #3

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In spite of their evident importance, there are very few data on ambient ultrafine particles, especially their mass concentrations and sources, in the scientific literature. The present papers aims to bring new insight into this topic using model simulations and concentrating on selected episodes in a large number of cities. In my opinion, this kind of a study is very welcome and should eventually be published. Before accepting the paper for publication, there are several issues that need to be clarified and revised in the paper. My detailed comments in this regard are given below.

Major issues

It is not entirely clear to me why the authors selected air pollution episodes lasting a

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Discussion paper

few days as the sole basis for estimating PM0.1 mass concentrations in different cities? Would a few days be much too short time period to get reliable information on different sources, and would selection of photochemical pollution episodes bias the importance of some sources over the others?

Since PM0.1 mass is the combined result of primary particle emissions (and nucleation) into this size range, and subsequent accumulatoin of secondary meterial by these particls, the authors should explain in more detail how they determined PM0.1 mass concentration (and the related source contribution) from their model simulations and what are the related uncertainties. There are several issues related to this. First, how many size bins the used model has in the sub-0.1 um size range and how close to the 0.1 is the border between the two nearest size bins? Now the authors only mention the number of size bin over the whole particle size range from 10 nm to 10 um (page 7). Second, what is the actual particle diameter used in model simulations? Mass measurements rely usually on aerodynamics diameters (impactors), while number size distribution measurements in the ultrafine size range rely usually on electric mobility diameters. These two diameters may differ substantially (up to a factor 2) for ambient aerosol particles, and the diameter used in a model can be either one of these two or something else. This is an important issue because PM mass size distributions often have a steep gradient at around 0.1 um, making the PM0.1 mass concentration very sensitive to the diameter chosen to represent the size 0.1 um. Third, the authors state that they do not care about nucleation because it only affects the particle number concentration but not the PM mass concentration. This is not true. Think, for example, a situation where 2 sources dominate the ultrafine particle number concentration: nucleation and a combustion source that produces particles with a peak diameter slightly below 100 nm. When these particle age in the atmosphere for a while and accumulate secondary material from the gas phase, those originating from nucleation tend to remain in the sub-100 nm size range while a big part of combustion particle may grow past 100 nm. As a result, whether or not to include nucleation also affects PM0.1 um mass. This issue should, at the very least, mentioned in the manuscript.

It is unclear to me how authors keep track on the different sources contributing to the PM0.1 mass concentration. I understand that keeping track particle numbers from different sources is possible, but how this is done for PM mass as a big fraction of it is formed secondarily in the atmosphere?

The authors use ozone and OC/BC concentrations in PM2.5 to evaluate their model. This is fine. However, it is clear that this sort of model evaluation does not guarantee that the model works well for PM0.1. While I do understand there are too few PM0.1 measurements around for a proper model evaluation in this respect, I still think that the authors should be more honest to state this explicitly in the manuscript (a good performance for ozone does, by no means, guarantee that also PM0.1 is simulated well).

I do not understand the last particle of the discussion before the conclusions, including equations 2 and 3 and figure 8. The obtained results misses a proper methodological description, and the actual results does not seem very helpful in the context of this paper. I would recommend removing this part of the analysis altogether from this paper.

Minor and technical issues

Section 2.3. Did the authors use real-meteorological data when calculating biogenic emissions using the MEGAN model? This is important because biogenic emissions are very sensitive to ambient temperatures. Please explain in the text.

What is the “Actual Max” for in the caption of figure 3? The given numbers are extremely accurate (3-5 digits) and well out of the scale of the relevant concentrations in these two sub-figures.

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