

Interactive comment on “Implementation of state-of-the-art ternary new particle formation scheme to the regional chemical transport model PMCAMx-UF in Europe” by E. Baranizadeh et al.

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This paper documents the implementation of a new nucleation scheme, based on the ACDC model, into a chemical transport model, PMCAMx-UF over Europe. Also, sensitivities to the radiation scheme and natural emissions were performed. The paper is in review for GMD and has a focus on model development rather than science findings. I feel like the scope of the paper is appropriate for the journal, and the overall research approach is good. However, the paper makes claims that I feel are unsubstantiated, lacks quantitative evaluation in many places, and is unclear in some places. Thus, I feel that the paper needs some substantial revisions before publication.

Broad comments:

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1) Lack of quantification against measurements: Figure 2 shows comparison of a single simulation (ACDC-RADM-DE... not ACDC-TUV-DE, which seemed to be the default for the other figures). While a few numbers are given for the percentage of points that fall within a factor of 2 of measurements, there aren't other statistics for slope and bias (N10 seems to be biased 5-10x too high on average). Further, why aren't statistics given for the other simulations? It would be straightforward to quantitatively compare the simulations against the measurements in this way, and this would show us which assumptions moved the model closer to measurements.

Similarly, this can be done for the vertical profiles. N4 are more than 10x too high with the ACDC scheme throughout most (~70%) of the boundary layer, and N10 is more than a factor of 2 too high for half of the the boundary layer. Please give some summary statistics.

2) Unsubstantiated claims: The paper concludes with, "Overall, we consider our results very promising: a NPF scheme based on first-principles theory and no artificial scaling is shown to be a promising alternative to semi-empirical approaches in the description of particle formation in large scale atmospheric models." There is similar discussion earlier in the conclusions, "reasonable particle concentrations". Obviously, "very promising" and "reasonable" are subjective statements, and different people would view the 10x boundary-layer overprediction for N4 as not "very promising" or "reasonable". We can use Westervelt et al. (2014) for comparison, where they compare simulations using scaled Napari (1E-5 in this case) versus unscaled Napari (also "first principles", and in the unscaled case, without a tuning factor). Table 3 and Figure 2 in Westervelt shows that N10 is only 2x higher in the boundary layer for the unscaled Napari simulation relative to the scaled Napari simulation... similar to what was found in the current manuscript in review between ACDC and scaled Napari (1E-6 in the current manuscript). Thus, the overpredictions by the ACDC scheme relative to the scaled Napari in this current manuscript is not unlike comparing scaled and unscaled Napari. I would guess that the nucleation rates predicted by ACDC must be many orders of

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magnitude faster than scaled Napari in order to get N4 to be 10x too high given all of the microphysical dampening between nucleation rates and N4.

While I know that Napari cannot be right for the right reasons, it is very surprising that ACDC does so poorly here considering that it does ok relative to CLOUD measurements (is there a mistake in the lookup tables? are these formation rates per m^3 rather than per cm^3 ?). What are the mean nucleation rates predicted at some sites where nucleation rates have been well-characterized (e.g. Hyytiala)?

I agree that missing condensible material may be part of the issue here (adding more would likely lower N4 and increase N100). D'Andrea et al. (2013) lowered N10 by about 20-50% over Europe by adding 100 Tg yr⁻¹ globally of non-volatile SOA correlated with anthropogenic CO (see Figure 3). Thus, the N10 bias in the current manuscript may be somewhat fixed by missing condensible material. But regardless, please don't make unsubstantiated claims about the current setup since the ACDC rates seem to be similar to the unscaled Napari scheme that we love to hate.

Specific comments:

L62: Adams and Seinfeld (2002) did not use the Napari nucleation scheme.

L145: Why not use ACDC with no ammonia for binary nucleation?

L160-165 (as well as throughout): These lines come out of nowhere but seem important. Were natural sources not included in PMCAMx-UF before? I didn't figure this out until later in the paper. While there is a test simulation turning natural emissions on, there is never much discussion about this in the paper. Why isn't having natural emissions on the default setting in this paper (with a sensitivity simulation with them off). I realize you chose this because you didn't have natural emissions before, but shouldn't the simulations with natural emissions be expected to better simulate the size distribution? Please give attention to the text and discussion of natural emissions throughout to ensure that it is clear.

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L246-247 (and this entire paragraph): Why is TUV *not* called for cells with clouds thinner than optical depth 5? Don't you want to consider radiative transfer here? Earlier you discuss wanting to have the effects of simulated aerosols on photolysis rates... so wouldn't you want this for clear-sky or thin-cloud conditions? Confusing.

L319: "spatial concentration". Do you mean the relative spatial distribution of concentrations? Since you don't show figures for these other simulations, would it make sense to quantify this in some way, e.g. a correlation coefficient between simulations?

Figures: Why is the default simulation shown in figures sometimes ACDC-TUV-DE (most figures) and sometimes ACDC-RADM-DE (Figure 2)?

Figures 3 and 4: Why is the height axis log scale? Do we want to focus mostly on the boundary layer?

L377-379: "somewhat closer"? Within 1.5x for scaled Napari vs. outside of 10x for ACDC for most of the boundary layer?! Please be quantitative and avoid subjective judgement.

General grammar comment: Adverbs modifying an adjective do not need a hyphen (and should not have a hyphen), e.g. "vertically resolved", "chemically resolved", "newly formed", "hourly averaged". There is no ambiguity in the meaning with or without a hyphen. These should be removed.

On the other hand, it is extremely useful to hyphenate joint adjectives. For example, "Aitken-mode particles" should have a hyphen as they are particles in the "Aitken mode". There are not "mode particles" that are "Aitken". Same with "chemical-transport models". They are not "transport models" that are "chemical". How about "large scale atmospheric models"? What is a "scale atmospheric model", and what is so large about it? It is commonplace, unfortunately in my opinion, to omit hyphens when the writer thinks the meaning is unambiguous. However, if a writer never hyphenates joint adjectives, we get into trouble when the writer writes, "slow moving van". Is it a "moving

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van” that is going slow? Or is it a regular van that is “slow moving”? If a writer establishes that they will hyphenate whenever it is appropriate, we would know that “slow moving van” means a “moving van” that is going slow (else they would have written “slow-moving van”).

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