

Reviewer #2:

This manuscript presents the implementation of a new ternary H₂SO₄-H₂O-NH₃ parameterization, into the PMCAMx-UF model. The authors explore the ability of the model to reproduce observed number concentration during May 2008, when the intensive observation period of EUCAARI project took place. Apart from the testing of the new parameterization, sensitivity tests using the scaled Napari parameterization and sensitivity to the radiation scheme and natural emissions were performed. The topic and overall approach fits with GMD; therefore, I am in favor of accepting this work for publication in GMD after the authors have addressed the issues summarized below.

We thank the reviewer for his /her encouraging comments. Below you can find our point-by-point responses to all the issues raised.

*While several sensitivity tests are done, the paper lacks a proper statistics for each test. Figure 2 shows the results coming from ACDC-RADM-DE sensitivity study, however no information is given for the other studies presented in Table 1. It would be nice to see some numbers (*r*, over/under estimation factor, bias), to endorse the statement “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.”*

This issue was also raised by Prof. Pierce in his review. Table R1 below shows an example statistics for the results presented in Fig. 2 of the original manuscript. We have calculated similar statistics for all simulations and will add them as supplementary information and summarize them in the revised manuscript.

	Mean Observed (cm ⁻³)	Mean Predicted (cm ⁻³)	NMB (%)	NME (%)	Percent within a factor of 2
Aspvreten					
N ₁₀	2205	7424	237	243	33
N ₅₀	1405	1273	-9	47	65
N ₁₀₀	582	327	-44	51	57
Cabauw					
N ₁₀	7702	12245	59	73	69
N ₅₀	4757	3298	-31	37	81
N ₁₀₀	1925	1041	-46	50	50
Hyytiälä					
N ₁₀	2658	5573	110	127	48
N ₅₀	1119	1076	-4	61	57
N ₁₀₀	461	238	-48	57	43
Ispra					
N ₁₀	7791	13239	70	93	62
N ₅₀	4038	3035	-25	41	71
N ₁₀₀	1725	1035	-40	49	56
Mace Head					
N ₁₀	3204	11617	263	268	30
N ₅₀	1825	1889	4	41	74
N ₁₀₀	952	490	-49	54	35
Melpitz					
N ₁₀	9621	20693	115	143	46
N ₅₀	4358	3135	-28	37	81
N ₁₀₀	1736	778	-55	56	40
Vaviihill					
N ₁₀	3579	11309	216	224	24
N ₅₀	1898	2007	6	36	84
N ₁₀₀	785	551	-30	41	60
Overall					
N ₁₀	5090	11540	127	145	44
N ₅₀	2648	2171	-18	41	72
N ₁₀₀	1115	613	-45	51	49

Table R1. The statistics of the agreement between the ACDC-RADM-DE and the in-situ observations.

L82: Should be “Matsui et al., 2011, 2013”, not “Matsui et al., 2011a, 2013c”.

Thank you for pointing this out, we will modify this in a revised version of the manuscript.

L88-90: Matsui et al., 2013 study, already mentioned by the authors, have also assessed the ability of WRF-Chem to reproduce the vertical profile of observed Aitken particles for South Asia.

Thank you for pointing this out, we will modify the manuscript accordingly.

L133-134: Should be “Yu et al., 2006”, not “Yu et al., 2006a”.

Thank you for pointing this out, we will correct this in the revised manuscript.

L210-213: I assume that if the H₂SO₄, NH₃, RH, temperature and condensation sink are not falling into the mentioned range, the Vehkamaki et al., 2002, parameterization is applied. Is that right?

The Vehkamaki et al. (2002) parameterization is called as long as the H₂SO₄ concentration is greater than 10⁴ cm⁻³. As mentioned in section 2.1, the ternary and binary pathways are operating simultaneously. The ternary pathway is called only if the vapor concentrations of both H₂SO₄ and NH₃ are above the lower limit of the lookup table. At lower concentrations the formation rate is practically zero, as the boundaries of the lookup table are chosen so that they should cover the atmospherically relevant range. When the ternary pathway is called, the H₂SO₄, NH₃, RH, temperature and condensation sink are limited to the bounds of the ACDC lookup table if any of the parameters fall outside of the boundaries. Although we did not count the frequency of any exceedances above or below the lookup table bounds, we are confident that these exceedances are few since the bounds are so large compared to atmospherically relevant conditions. Furthermore, even if e.g. the vapor concentrations exceed the upper limits, the rate is likely already converged to a plateau, making it safe to use the values at the limits. We will clarify this in the revised manuscript.

L330-336: The authors show the scatter plots of predicted PNC using ACDC-RADMDE simulation vs observed PNC in several size ranges. Yet, at line 303 they state that ACDC-TUV-DE is the baseline simulation. Do they have any particular reason not to present the results coming from the default simulation? As can be seen in Table 1 the differences between the ACDC-RADM-DE and ACDC-TUV-DE simulations are minors. Furthermore, they use the ACDC-TUV-DE simulation results for the following plots. A little bit confusing.

The reason for presenting ACDC-RADM-DE in Fig. 2 of the original manuscript was to show a direct comparison with Fig. 3 of Foutoukis et al. (2012) with the only difference being the different NPF scheme. To avoid confusion, we will show results from ACDC-TUV-DE in Fig. 2 of the revised manuscript.

L373-379: Could you give an explanation why N₄ concentration increases in the upper boundary layer for ACDC-TUV-DE simulation? May you could present the particle formation rates for the ACDC-TUV-DE and Napari-TUV-DE simulation. Also, could you give an overestimation factor?

This is most like due to the simultaneous drop in temperature and concentrations of larger particles (i.e. the coagulation sink for the newly-formed particles), which enhance both the particle formation rate and their survival. In fact, the temperature dependence is one of the key differences between the new ACDC-based NPF representation and the previous semi-empirical approaches: while the latter do not usually have an explicit temperature dependence of the formation rates, the former does. The ACDC-TUV-DE simulations over-predict N₄ with a factor of 8-10 below 2 km and 4-9 above 2 km (see also responses to Prof. Pierce). We will revise the manuscript accordingly to clarify these issues.

L384-389: An index of agreement will sustain “the scaled Napari NPF scheme agrees reasonably well with the observations throughout the atmospheric column” and “reasonably well” statements.

We agree that these are subjective statements that need to be backed up by quantitative data. As discussed above, we will add two supplementary tables (showing the NMB, NME and correlation coefficients for all the simulations vs. in-situ and aircraft data, respectively) and summarize these statistics in the text. We have also calculated indexes of agreement for all our data (surface level and aircraft). For the Napari-TUV-DE simulations these values range from about 0.2 to 0.7 for all the size ranges throughout the atmospheric column, while for the ACDC-TUV-DE shows a large difference between the in-situ (0.3-0.6 for all sizes) and aircraft (0.02-0.7 for all sizes, with the poorest agreement for the smallest particles) data sets. We hope the inclusion of more statistical metrics will now do the job and will also go through the revised manuscript to remove all unnecessary subjective statements.

L433_435: The following sentence for a more scientifically sound expression should be rephrased: “We believe this is the first time that reasonable particle concentrations have been produced in a large-scale atmospheric a

Good point. We will modify the revised manuscript accordingly.

The authors should be more restrictive in using “reasonably well”, “are somewhat overpredicted by the ACDC-based NPF scheme”, “very promising” statements due to the fact that the lack of statistics throughout the paper does not sustain their claims.

Besides adding the aforementioned more quantitative analysis to support these statements, we will limit the usage of the phrases in question in the revised manuscript.