Geosci. Model Dev. Discuss.,
https://doi.org/10.5194/gmd-2018-171-AC1, 2018

Geoscientific Model Development Discussions

Interactive comment on "Two new submodels for the Modular Earth Submodel System (MESSy): New Aerosol Nucleation (NAN) and small ions (IONS) version 1.0" *by* Sebastian Ehrhart et al.

Sebastian Ehrhart et al.

s.ehrhart@mpic.de

Received and published: 19 November 2018

AR: We thank Referee 1 for their time and for the useful suggestions and correction. Here we addressed all comments (reporting the original comments). The manuscript as been improved accordingly.

RC: P2 L4: Merikanto et al. (2007) was not the first. Napari et al. (2002), An improved model for ternary nucleation of sulfuric acid–ammonia–water, The Journal of Chemical Physics 116, 4221 was earlier (and there may be others earlier than this).

AR: We will include the suggested reference, also rephrasing the sentence in the revised version: "Napari et al. (2002) derived a parameterisation of the H_2SO_4-NH_3-H_2O system based on theoretical calculations and an improved parameterisation was developed by Merikanto et al. (2007)."

RC: P2 L32: what does EMAC stand for?

AR: EMAC stands for ECHAM/MESSy Atmospheric Chemistry. The definition was given in the abstract but we added again the definition to the mentioned position in the revised manuscript.

RC: P4 L2: Confusing. Does 214Bi go to 214Po first?

AR: The referee is correct that ^{214}Bi undergoes a β^- decay to ^{214}Po with a half life time of 20 min. ^{214}Po α decays almost immediately (half life time 160 μs). For this reason the aforementioned alpha decay is not explicitly treated by DRADON. The same is true for R5, the decay of ^{210}Pb to ^{206}Pb, which goes via ^{210}Bi and ^{210}Po to ^{206}Pb (both β^-) at life times much shorter than the 22 years for the first step. We also realised that the Proton number of Bi was incorrectly given as 81 instead of the correct 83. We removed proton numbers in the revised manuscript to improve readability. We also indicate now the charge of the β decay.

RC: R1-R5: Do all of the species here need to be advected in the model? Many of the species lifetimes are shorter than typical advection timescales.

AR: The reactions R1-R5 are part of the DRADON submodel, an already existing submodel in MESSy. Advection for each singular tracer can be (de)activated via namelist, and therefore user-dependent. Indeed, for such tracers, advection could be avoided as already done for many other species (e.g. O_3^P).

RC: P4 L17: What is IGRF? Citation for where these “first 3 coefficients” come from?

AR: IGRF is the International Geomagnetic Reference Field. We added this missing definition to the text and added the relevant citation.

RC: P4 L31. The discussion around Eq 2 largely discusses radius, and the variable is “r_um”, so I was surprised to see that it was defined as “diameter”. Is it actually diameter or is it radius? If it is diameter, it would be better to have the variable be “d” and the use “diameter” for the rest of the discussion.

AR: Eq 2 is parameterised for a radius in micro meter. For clarity, we use now $d_{\mu m}/2$ in Eq 2 and 3 and changed all text to diameter. We hope this avoids confusion.

RC: P5 L3-10: So is growth of small charged particles to larger sizes where they are then “large ions” a loss of small ions then?

AR: It is a loss of ions which can nucleate. An ion that nucleated is an intermediate size ion. For ions with a diameter of up to 10 nm the ion-ion recombination coefficient is still around 1.6e-6 cm^3 s^{-1}. Assuming absence of aerosol particles, or any other loss, and an ion pair production rate of 5 i.p. cm^{-3} s^{-1} at ground level (Figure 4 a) the steady state ion pair concentration would be around 1800 i.p. cm^{-3}. This would mean an ion life time of 350 s, roughly 6 minutes. Losses to aerosol particles will change this picture slightly but will also lower the overall lifetime of small ions. All factors included the lifetime of small ion clusters is very short while the time for an ion cluster to grow to a diameter larger than 10 nm is in most circumstances longer, a typical rule of thumb is 1 nm/h for a [H_2SO_4] = 1e7 cm^{-3}. The size of ions becomes important when the conductivity of air is calculated. For such a calculation a more detailed ion aerosol model is required that describes ions size resolved.

RC: P6 L12: “oni”

AR: We corrected it to "on".

RC: Table 1 and Section 3.2.1: If I’m correct, this evaluation of the placement NAN be- fore, within, or after GMXe has to do with operator splitting and master timesteps vs. internal GMXe timesteps. By taking NAN out of GMXe nucleation is called on the master timestep and then other aerosol microphysical processes are called in GMXe for the master timestep. When NAN is in GMXe, I’m guessing it can be called more frequently in some internal GMXe timestep. The balance between condensation and nucleation are quite sensitive to the timestep and order of operations, especially when the timestep is similar to or longer than the condensation sink timescale (and this could explain why the results in Figures 5 and 6 are sensitive to the placement of NAN for some cases by not for most). Am I correct about this? If yes, it would make sense to frame the motivation and discussion of Table 1 and Section 3.2.1 around errors due to timesteps and order of operations. Right now, the paper is fairly cryptic as to why the differences arise (“linearization to non-linear processes”, but if my hypothesis is correct, I think the explanation is straightforward.

AR: The referee is partially right. We added the following paragraph to the revised MSs, to motivate the analysis and Section 3.2.1: "Nucleation rates typical follow a power law with respect to vapour concentrations, see for example Kashchiev (1982) and Oxtoby and Kashchiev (1994). Therefore small changes in the vapour concentration, here H_2SO_4 and NH_3, can have a large influence on the nucleation rate. Condensation proceeds typically faster than nucleation, it is reasonable to place the nucleation after the condensation in a time step. Therefore, the original implementation of GMXe calculates nucleation after it calculates the amount of vapour that condensed on aerosol particles. There is no internal shorter time step in GMXe. However, condensation is not the only process affecting vapour concentrations, or particle concentration. Therefore aerosol particle concentrations are also sensitive to the placement of GMXe within MESSy’s

interface layer. Unfortunately, making microphysical processes available for as many submodels and potential users as possible is best achieved as a submodel, as MESSy has currently no unified interface definition for sub-submodels, i.e. a submodel of a submodel. Therefore, implementation of NAN and IONS as submodel was preferred as both models can be called independently of the choice of other submodels."

RC: P11 L18: “For two of the stations”. It’s 3 stations, right?

AR: The referee is correct, we show 3 stations. We corrected it and also added now Mace Head into the text.

RC: P12 L10: “Large uncertainties remain, mainly due to the incomplete nature of the im- plemented nucleation rate parameterizations.” This sounds like the authors are saying that if we just fixed our nucleation rate parameterizations, most of the model uncer- tainties in aerosol predictions would go away. However, simulating nucleation perfectly would only marginally improve simulations (e.g., Lee, L. A., Pringle, K. J., Redding- ton, C. L., Mann, G. W., Stier, P., Spracklen, D. V., Pierce, J. R., and Carslaw, K. S.: The magnitude and causes of uncertainty in global model simulations of cloud conden- sation nuclei, Atmos. Chem. Phys., 13, 8879-8914, doi:10.5194/acp-13-8879-2013, 2013.), Or am I misinterpreting what the authors are trying to say here?

AR: The uncertainties of the nucleation rates are due to the incomplete nature of the parameterisation. As the referee mentioned in their reply: ".. for the overall aerosol predictions many factors contribute to the overall uncertainty." We rephrase the sentence to avoid misinterpretation.

References

Kashchiev, D.: On the relation between nucleation work, nucleus size, and nucleation rate, The Journal of Chemical Physics, 76, 5098–5102, doi:10.1063/1.442808, 1982.

Merikanto, J., Napari, I., Vehkamäki, H., Anttila, T., and Kulmala, M.: New parameterization of sulfuric acid-ammonia-water ternary nucleation rates at tropospheric conditions, Journal of Geophysical Research: Atmospheres, 112, n/a–n/a, doi:10.1029/2006JD007977, http://dx.doi.org/10.1029/2006JD007977, d15207, 2007.

Napari, I., Noppel, M., Vehkamäki, H., and Kulmala, M.: Parametrization of ternary nucleation rates for H2SO4-NH3-H2O vapors, Journal of Geophysical Research: Atmospheres, 107, AAC 6–1–AAC 6–6, doi:10.1029/2002JD002132, http://dx.doi.org/10.1029/2002JD002132, 4381, 2002.

Oxtoby, D. W. and Kashchiev, D.: A general relation between the nucleation work and the size of the nucleus in multicomponent nucleation, The Journal of Chemical Physics, 100, 7665–7671, doi:10.1063/1.466859, 1994.

Interactive comment on Geosci. Model Dev. Discuss., https://doi.org/10.5194/gmd-2018-171, 2018.