

The study by Gonzalez Carracedo et al. reports observations from the boreal forest that show that below 3 nm, the particle growth rate derived from ion observations is lower than that derived from total particle concentration measurements. This work confirms observations from a previous study (Gonser et al., 2014) and provides an explanation, through the combined analysis of real-atmosphere and laboratory measurements and the use of a process model. Because of its indirect impact on climate, NPF is a key atmospheric process whose study is crucial; the question addressed in this study is of particular interest since the possibility of generalizing observations made from ion measurements (often easier and more direct than neutral particle measurements) to the total particle population is critical. This is an otherwise well written paper, which I therefore recommend for publication in ACP. I suggest however that the comments listed below be addressed before final publication, with, in particular, a clarification in the description of the simulations that are at the heart of this study (Sects. 2.5 and 3.3).

Comment 1: P1, L29 : I think that a complementary reference to the more recent reviews by Kerminen et al. (2018) or Lee et al. (2019) could shed light on observations made during the last ~20 years, after the review by Kulmala et al. (2004) was published.

Comment 2: P2, L41-42: I would suggest referring to papers that describe the most commonly used methods for GR calculation at this point in the text; in addition to the “maximum” (Hirsikko et al., 2005) and appearance time (Lehtipalo et al., 2014) approaches which are more fully described later in the paper, the work of Dal Maso et al. (2005) could for example be mentioned.

Comment 3: P2, L44: In addition to Manninen et al. (2009), I would suggest referring here to the studies by Mirme and Mirme (2013) and Manninen et al. (2016) which are more dedicated to the description of the instrument and its functioning, and which are mentioned elsewhere in the text. Manninen et al. (2016) could also be added at L109.

Comment 4: P2, Sect. 2.1: It would be interesting to indicate / recall (here or later in the results Sect.) how many NPF events were considered in the analysis, for both Hyytiälä and CLOUD datasets.

Comment 5: P3, Table 1:

- If I am not mistaken, the names given to the simulations (according to the formulation used for J) are only used in Table 1, whereas their use in the text could help when describing successively the different simulations. I think that it should also be made clear that "simple" refers to the prescription of a diurnal profile of J, whereas "lehtipalo" indicates the use of parameterizations;
- Why is it indicated "not used" for the HOM dimers concentration while, in line with the formulation of the parameterizations of J (Eqs. 2 and 4), this variable is a priori used in the simulations (e.g.: L251-252)?
- Why is there no value for the total HOM concentration in the CLOUD experiments?
- In the line indicating the product of the precursor concentrations used in Eqs. 2 and 4, [HOM] should be replaced by $[HOM_{dim}]$, right?
- The meaning of $Q_{i.p.}$ and $N_{i.p.}$ should be explained.

I think that in general, in relation to Sect. 3.3, a clearer description of the simulations would be beneficial to the understanding of the results. However, Sect. 2.5 may come too early in the manuscript for such a detailed description, so one option would be to keep only the general description of the model in Sect. 2.5 and offer a more complete/detailed description of the simulations directly in Sect. 3.3 (and in this case, the last three columns of Table 1 could be removed and made into a table of their own in Sect. 3.3).

Comment 6: P3, L84-85: “we also find a parametrization for the ion-induced nucleation fraction based on the ion-pair production rate and the vapor concentrations at which ion-induced nucleation becomes less dominant”: the approach used to derive the parameterization is not clear to me, can the authors try to specify / clarify?

Comment 7: P4, L86: The equation number should be corrected (4 instead of 3).

Comment 8: P4, L94-96: “a good sensitivity towards low particle concentrations in the sub-10 nm range”, “the DMA-train can measure also sub-3 nm particle growth with an unprecedented sizing precision”: Is there a study in which the performance of the DMA-train has been compared with that of instruments that measure over a comparable size range, including in particular NAIS to also support L169-170 (“where however the total growth rate has generally higher uncertainties due to lower signal when compared to the DMA-train”)?

Comment 9: P5, Sect. 2.3:

- Since the mass spectrometry measurements were conducted at a different height than the other measurements (which may therefore raise questions about potentially different conditions with respect to forest canopy), I would suggest referring to Zha et al. (2018), who indicate that during daytime, i.e. the period of interest for NPF, HOM measurements (concentrations and composition distributions) are in fact similar below (i.e. near the ground) and above (i.e. at 35 m height) the canopy;
- I would suggest saying few words on the calibration factor used to convert CI-API-TOF signals into concentrations in molecules / cm³. At least, was the same coefficient applied to sulfuric acid and HOM signals?
- L125: “at similar instrument” → a similar instrument
- L126: “Lehtiplao” → Lehtipalo

Comment 10: P5, L136: “which cannot necessarily translated into a pure condensational growth rate”: the wording should be checked.

Comment 11: P6, L166-167: I take the opportunity of this first illustration of a result that is at the heart of this study to make a more general comment. For a given instrument and calculation method, GR_{Ions} appears to be indeed lower than GR_{Total} below 3 nm. I think it would be interesting to try to quantify (at least roughly) these differences (there is a factor of 2 indicated in the conclusions L338; it would be interesting to mention and discuss this earlier in the manuscript), especially to quickly discuss their magnitude/importance in relation to the uncertainties on the GR estimate related to the different calculation methods and/or the use of different instruments (e.g. Yli-Juuti et al., 2011). In other words, is the estimation error made on GR_{Total} considering GR_{Ion} of the same order, lower, higher, than the differences in the evaluation of GR_{Total} related to the use of different methods / instruments?

Comment 12: P6, L169: In order to avoid any confusion, I would clearly refer to NAIS: “when using the same instrument (i.e. NAIS)...”.

Comment 13: P7, Fig. 1.b: Similar to Fig. 1.a, I would suggest adding the lines delimiting the [1:2, 2:1] range to help in the visual evaluation of the differences between GR_{Ions} and GR_{Total}.

Comment 14: P7, L191: Since the ranges delimited by the boxes / error bars overlap in Fig. 2.a, I would suggest saying “The sub-3 nm ambient ion growth rates are on average clearly lower than the total growth rates...”.

Comment 15: P7, L192-193: “However, at larger sizes (3-8 nm), both the laboratory and ambient measurements show no significant differences between the apparent ion and total growth rates (Fig. 2b and 2d)”: I do not agree with this statement; the GRs obtained from ion measurements in CLOUD (Fig. 2.d) are on average higher than the total GR, with differences that appear to be of the same order as those found for the ambient measurements in the lower size range (Fig. 2.a).

Comment 16: P8, Fig. 2:

- Fig. 2.a-d: the meaning of the symbol should be defined explicitly (median, quartiles, range indicated by the error bars, signification of the diamonds);
- Fig. 2.e: In order to ease the reading of the figure, I would suggest keeping the same symbol for Hyytiälä and CLOUD for all instruments, and only use different colours to distinguish between the different instruments.

Comment 17: P10, L230-231: “with the latter becoming more and more significant at a later stage when also the neutral nucleation pathway dominant”: the wording should be checked.

Comment 18: P10, L245: “The results are presented in Figure we can clearly observe”: the wording should be checked.

Comment 19: P10, Sect. 3.3: Related to Comment 5, I would suggest clarifying the description of the simulations, as well as of some results. Specific points that I would in particular suggest to address are listed below:

- Does the “organic concentration” mentioned in L241 correspond to HOM_{tot} in Table 1? If so, why are the values given in the text and in Table 1 different (1×10^7 vs $2 \times 10^7 \text{ cm}^{-3}$)? More broadly, as done for sulphuric acid in L241, the concentrations chosen for the other precursors should at some point be discussed and justified;
- It think it would help to clearly recall at L240-242 that the mentioned species are used for GR calculation in the model (to avoid any confusion with J);
- L249: Eq. (4) was used as well, wasn't it?

I also strongly suggest to clarify the description of Fig. 5 (expanding / clarifying in particular the sentence that is currently in lines 250-252):

- indicating clearly that the measurements reported on Fig. 5.a are those from the CLOUD experiments which allowed to derive the parametrizations of J used in the simulation;
- specifying how the diurnal profile of the product of the concentration of measured precursors is obtained (average over NPF event days at the station?);
- indicating perhaps also clearly that the profile of the fraction of ion-induced nucleation represented in Fig 5.c is obtained by considering the idealized profile of the product of the concentration of precursors shown in the same figure.

Comment 20: P12, L282: udnerestimated → underestimated

Comment 21: P13, Fig. 6: the diurnal profile of sulfuric acid concentration shown in Figure 6.d appears to be different from that shown in Figure 4.d, and unless I am mistaken it is not indicated in the text that the sulfuric acid profile is different in the two simulations. Can the authors provide an explanation?

Comment 22: P14, Conclusions : This study is dedicated to the boreal forest but do we have any idea / can we anticipate the observations that could be made in other environments and in particular at high altitude, where the role of ions in nucleation is a priori more marked than at low altitude (Manninen et al., 2010; Sellegri et al., 2019)? Can we assume that we have less error in GR_{total} based on ion observations there, or is it difficult to anticipate without detailed knowledge of the precursors involved?

References :

Dal Maso, M., Kulmala M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P. P., and Lehtinen, K. E. J.: Formation and growth of freshatmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Environ. Res.*, 10, 323–336, 2005.

Kerminen, V.-M., Chen, X., Vakkari, V., Petäjä, T., Kulmala, M., and Bianchi, F.: Atmospheric new particle formation and growth: review of field observations, *Environ. Res. Lett.*, 13, 103003, <https://doi.org/10.1088/1748-9326/aadf3c>, 2018.

Lee, S.-H., Gordon, H., Yu, H., Lehtipalo, K., Haley, R., Li, Y., & Zhang, R. (2019). New particle formation in the atmosphere: From molecular clusters to global climate. *Journal of Geophysical Research: Atmospheres*, 124, 7098–7146. <https://doi.org/10.1029/2018JD029356>.

Sellegri, K., Rose, C., Marinoni, A., Lupi, A., Wiedensohler, A., Andrade, M., Bonasoni, P. and Laj, P. : New particle formation : a review of ground-based observations at Mountain research stations, *Atmosphere*, 10(9), 493, <https://doi.org/10.3390/atmos10090493>, 2019.

Zha, Q., Yan, C., Junninen, H., Riva, M., Sarnela, N., Aalto, J., Quéléver, L., Schallhart, S., Dada, L., Heikkinen, L., Peräkylä, O., Zou, J., Rose, C., Wang, Y., Mammarella, I., Katul, G., Vesala, T., Worsnop, D. R., Kulmala, M., Petäjä, T., Bianchi, F., and Ehn, M.: Vertical characterization of highly oxygenated molecules (HOMs) below and above a boreal forest canopy, *Atmos. Chem. Phys.*, 18, 17437–17450, <https://doi.org/10.5194/acp-18-17437-2018>, 2018.