Second response to reviewers for "What chemical species are responsible for new particle formation and growth in the Netherlands? A hybrid positive matrix factorization (PMF) analysis using aerosol composition (ACSM) and size (SMPS)" by Nursanto, Farhan R; Meinen, R.; Holzinger, R.; Krol, Maarten C.; Liu, Xinya; Dusek, Uli; Fry, Juliane L. (Manuscript ID: EGUSPHERE-2023-554)

The last modification results in the following changes in figure numbering:

Before revision	After revision
Figure 1 and 2	Stay as Figure 1 and 2
	New figure entry as Figure 3.
	Figure 3. Relation between the size-driven factors (F4 and F3, linked to NPF and growth) with the composition-driven factors (F2 and F1, linked to the bulk atmospheric aerosol composition) illustrating aerosol formation progress.
Figure 3-6	Now Figure 4-7

## Responses to comments:

1) The authors interchange MO-OOA and LO-OOA for the F4 factor for NPF. Please check which one it is. Further, it would be good to explain, if it is LO-OOA, why it is less-oxidized, as it is typically thought that very low volatility compounds are needed NPF. As it is not straight-forward to say that LO-OOA means it is semi- or low-volatile, understand this can be complicated, but would be beneficial for the story to reduce confusion or provide motivation for future studies.

We thank you for the comments. We meant to refer to the F4 factor as LO-OOA and not MO-OOA, for all seasons. Confusion was produced because of a mistaken label in the Figure 6; we thank the reviewers for flagging this and have corrected it. We also noticed this error in the last paragraph of Sect. 3.2.2 mentioning that there are in total 1 LO-OOA and 2 MO-OOA factors instead of 2 LO-OOA and 1 MO-OOA factors. The corrected paragraph includes the line:

"The PMF analyses in this study resolved one POA factor (as HOA factor) and three SOA factors (two LO-OOA factors and one MO-OOA factor) across periods, ..."

We also added some text explaining our interpretation of what the reviewer notes as a surprising presence of LO-OOA rather than MO-OOA in the nucleation mode factor, in the first paragraph of section 3.3:

"The organic mass spectrum profile from each size-driven factors and their diurnal cycles in each period are shown in Fig. 6. Across seasons, LO-OOA is part of bulk composition related to nucleation-mode particles. The factors are assigned as LO-OOA due to their f<sub>44</sub>/f<sub>43</sub> values compared to other OOA factors (see the triangle plot in Fig. S6). The LO-OOA F4 profile resolved in this study is comparable to LO-OOA resolved in other aerosol mass spectrometry studies using CV (Zheng et al., 2020; Joo et al., 2021), although fragments with m/z > 50 are less prevalent. Several aerosol chamber experiments have reported that lower volatility and highly oxygenated organic molecules from biogenic and anthropogenic organic precursors play a dominant role in new particle formation and growth (Schobesberger et al., 2013; Ehn et al., 2014; Riccobono et al., 2014; Tröstl et al., 2016; Mohr et al., 2019; Pospisilova et al., 2020; Zhao et al., 2021). In this study, however, we surprisingly observe LO-OOA rather than MO-OOA associated with nucleation. This could imply that organic compounds with less oxygenation are more abundant and condense on freshly nucleated particles in this region, or that the ToF-ACSM delineation between LO-OOA and MO-OOA does not directly correspond to volatility in this case."

- 2) The explanation about F4 and F3 starting at line 429 is extremely appreciated. I almost wish it was sooner, as seeing Fig. 3, Fig. 5, and the description about the factors prior to the explanation is occurring. I understand this may impact the flow of the paper, but not discussing why F3 does not looking like F4 with nitrate will cause confusion until the reader gets to line 429.
- 3) Comment 2) may be rectified by incorporating Fig (2) from the responses. I really appreciate the updated figures, including the Fig (1) from responses that is now Fig (3) in manuscript. Inclusion of Fig (2) provides a great summary and cartoon of the results and hypothesis the authors are discussing. Introduction of this Figure potentially before Fig (3) in the manuscript and a guick "overview" or something along that line could be of use for the readers.

Thank you for this suggestion; we now include the schematic figure in the manuscript, to illustrate how F4 and F3 could be connected in different pathways of particle nucleation and growth. We added this new Fig. 3 with caption at the end of section 3.2.1:

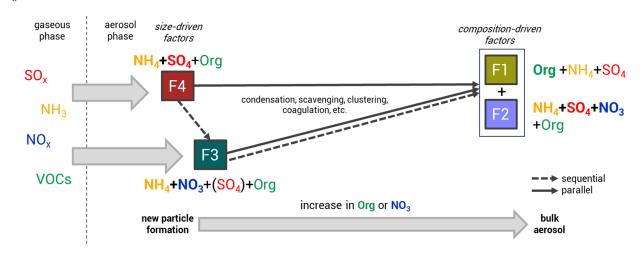


Figure 3. Potential relationships between the size-driven factors (F4 and F3, linked to NPF and growth) with the composition-driven factors (F2 and F1, linked to the bulk atmospheric aerosol composition), illustrating multiple possible aerosol growth pathways. From F4 (mainly ammonium sulfate) and F3 (mainly ammonium nitrate), particles can grow into F2 (OA and IA mixed) and/or F1 (OA-dominant), either sequentially (dashed), in parallel (solid), or combined. The particle formation and growth occur through condensation of gaseous precursors or particle coagulation. An increase in organics and NO<sub>3</sub> in the bulk composition is observed as particles progress along these pathways."

The accompanying text has been modified to refer to this figure at the end of Section 3.2.1.: "We can summarize that the NPF and growth follow the pathway starting from F4 and F3 into F2 and F1 (bulk aerosol composition), likely through processes such as condensation of gaseous precursors (SOx, NH3, NOx, and VOCs and their reaction products) or particle coagulation (see Fig. 3). We note that this does not imply that all aerosol growth proceeds sequentially through these four factors; a more detailed discussion of possible NPF and growth pathways is found below in Sect. 3.3.4"

And we have added reference to Fig. 3 in section 3.3.4.

## Additional minor changes:

We in the meantime have conducted our own detection limit measurements, so we have updated this in the text section 2.2.1.:

"The detection limits (measured similarly to Fröhlich et al., 2013) at 10-minute time resolution for this ToF-ACSM operating at Cabauw (a relatively polluted site in central Netherlands) are 0.38  $\mu$ g m<sup>-3</sup> for Org, 0.12  $\mu$ g m<sup>-3</sup> for NH<sub>4</sub>, 0.07  $\mu$ g m<sup>-3</sup> for NO<sub>3</sub>, 0.11  $\mu$ g m<sup>-3</sup> for SO<sub>4</sub>, and 0.09  $\mu$ g m<sup>-3</sup> for CI."

We have also updated the acknowledgements section and made other small edits throughout the manuscript; we have made no changes to the supplemental information but will re-upload the file.