Editor decision: Publish subject to minor revisions (review by editor):

We thank the Editor for reviewing the manuscript and for her comments to improve the quality of the manuscript. A point-by-point response to each comment is included below. Editor's comments are noted in bold. Changes in the manuscript are noted between quotation marks, underlined, and are referred to the corresponding line in the new revised version of the manuscript.

Dear Authors,

Thank you for the revised manuscript which has clearly improved thanks to reviewer's excellent suggestions.

As a final note, I would encourage the authors to reconsider incorporating some of the reviewer's suggestions better into the manuscript, particularly:

1. Reply on the comment concerning Figure 8a. I understand this comment differently, as a request to plot the points in the figure in the order from the smallest f44 towards the highest f44 (instead of ordering them with time), such that the highest values would be clearly visible and not covered by blue points. So please check this once more.

Sorry for misunderstanding the reviewer's comment, we have now considered the suggestion in the correct way, and we have updated Figure 8a accordingly. Also, the marker of the filtered data (small black dots in the previous version of the manuscript) has been removed for better visualization of the datapoints' colors.

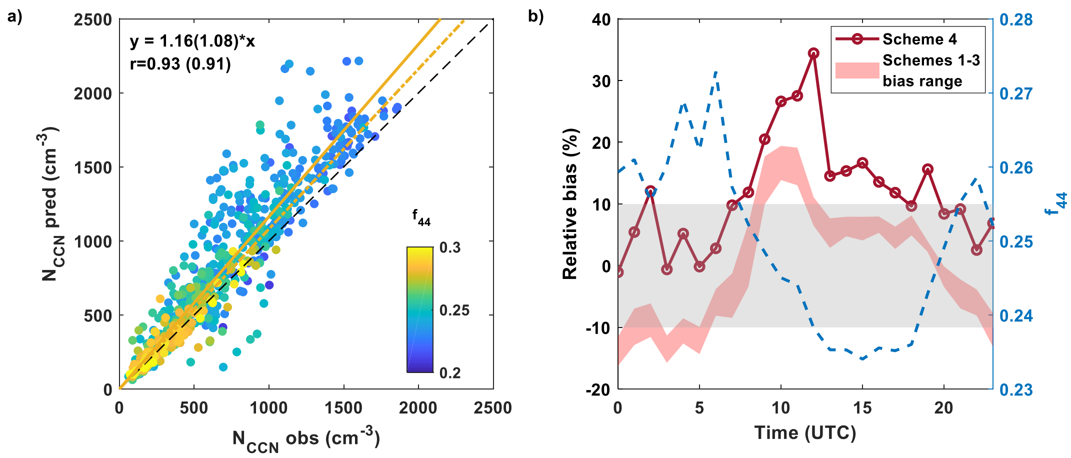

Figure 8. a) Scatter plot of predicted CCN concentrations (N_{CCN} pred) as a function of observed CCN concentrations (N_{CCN} obs) using the OA scheme 4. Datapoints are colored-coded by the corresponding f_{44} value. The black dash line represents the 1:1 line. The linear equation and Pearson correlation coefficient (r) are also included for all data and for filtered data in parenthesis (datapoints with f_{44}>0.25). The yellow solid and dash lines represent the linear regression of all and filtered data, respectively. b) Median diurnal evolution of the relative bias at SS=0.4% of the OA scheme 4 (left Y axis) and f_{44} (right Y axis). The grey shaded area represents the ±10% relative bias. The red shaded area represents the relative bias range for the other OA schemes shown in Figure 6b.

2. Adding possibly discussion of the reviewer's concerns (particularly the points raised by reviewer #2), such as the impact of external/internal aerosol mixtures on the comparison results. If this is not possible to evaluate quantitatively, could it still be addressed as an uncertainty/limitation of the study?

Following editor's suggestion, we have revised the whole manuscript in order to remark (in the results and conclusion sections) the limitation of the study to evaluate the impact of the aerosol mixing state on the prediction models' performance. However, we want to remark that the discussion of the results in the manuscript does not quantitatively address this issue since it is not possible with the measurements available in this study.

In lines 450-454: "Moreover, both methods assume internally mixed particles to estimate the overall κ, which is an important limitation in the case of externally mixed particles (Wang et al., 2010; Ren et al., 2018; Kulkarni et al., 2023). In this study, we would expect that during midday hours at SNS the aerosol population would be more externally mixed due to the influence of NPF events and vertical transport of particles; however, with the instrumentation and methods used here we cannot give conclusive information about aerosol mixing state and its impact on the different scheme's performance.".

In lines 604-605: "However, the analytical methods used in this study have limitations and do not consider aerosol mixing state information. For that reason, the next section will explore a non-analytical approach with the aim of improving the CCN prediction throughout the day."

In lines 675-680: "Second, when the aerosol population consists of a complex mixture of particles, which at SNS can be observed during PBL influence conditions, the underlying assumptions for estimating CCN concentrations based on internally mixed aerosol particles can introduce an intrinsic bias and κ_{OA} assumptions have a secondary role. Since the methodology used here is limited and cannot directly infer the mixing state of the aerosol population, we are not able to quantify this effect on the CCN predictions. Moreover, during morning and midday hours related to more complex conditions, the relationship between variables might change over time and can have a non-linear nature."

3. Adding possibly supporting figure (e.g. in supplement) to strengthen the statement in lines L410-411: "The main difference in the data distribution between both schemes is observed at low hygroscopicity values, which have been identified as periods of higher HOA contribution (i.e., during HOA peak events). ", as was suggested.

We have added the following figure to the supplement of the manuscript to support the statement in lines L410-411 as suggested by the reviewer and the Editor. Lines 430-431 of the revised version have been updated as follows:

"The main difference in the data distribution between both schemes is observed at low hygroscopicity values, which have been identified as periods of higher HOA contribution (i.e., during HOA peak events) as can be observed in Figure S5."

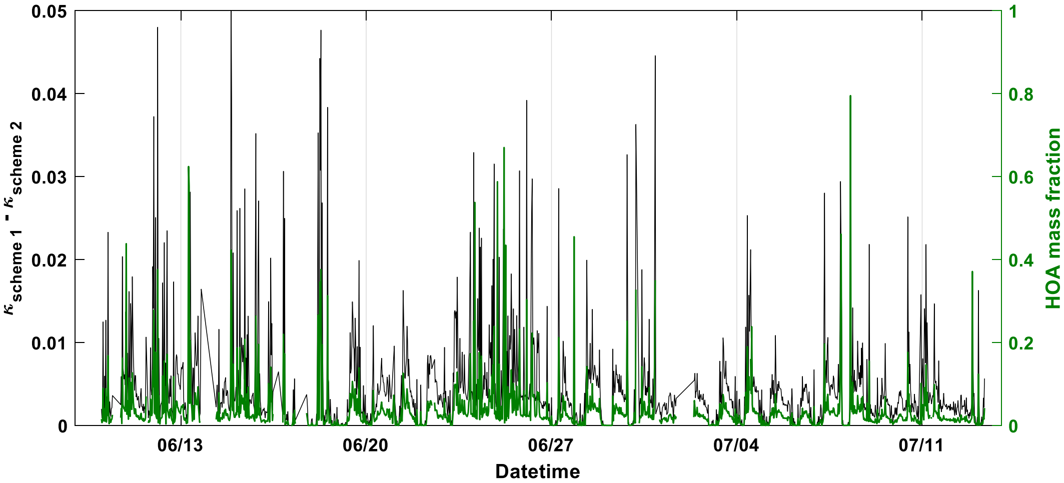

Figure S5. Time series along the field campaign of hygroscopicity parameter difference between both schemes (left Y-axis) and HOA mass fraction (right Y-axis).

4. clarifying the wind conditions, since this question was raised by both reviewers.

To clarify the discussion about the wind conditions during the campaign, we have added the following sentences in lines 330-340 of the revised version of the manuscript and the figure attached below in the supplement (Figure S2):

"Figure S1 shows the timeseries of meteorological variables (temperature, pressure, and relative humidity) during the campaign. The second half of the campaign is characterized by higher temperatures, higher pressure and lower relative humidity compared to the first half. In addition, we observed that the diurnal pattern of wind speed is very similar during the whole campaign (with higher wind speeds during the evening and lower at the central hours of the day), but the second half of the campaign shows higher values of the wind speed respect to the first half of the campaign (Figure S2). The wind direction was also predominantly from the west for the whole campaign, but we observed that the first half of the campaign shows a more pronounced diurnal pattern and more influence of other wind directions (Figure S2). During the second half of the campaign, a more constant wind direction is observed suggesting a continuous transport of aerosol from the valley to the mountain. The significant difference in the meteorological conditions between both halves of the campaign can be associated to a more efficient transport of aerosol from the valley to the mountain during the second half of the campaign, which would involve differences in the aerosol physico-chemical properties, and in particular in the predominance of the OA factors (50% contribution of LO-OOA and 48% of MO-OOA during the second half compared to 26% and 72% during the first half). A detailed analysis of the air masses and wind influence in aerosol composition during BioCloud campaign is shown in Jaén et al. (2023)."

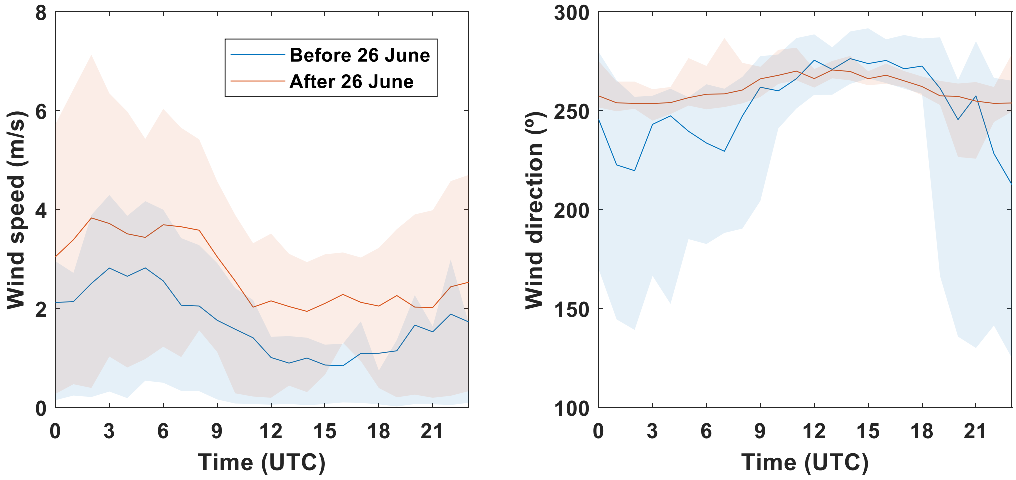

Figure S2. Mean diurnal pattern of the wind speed (left panel) and direction (right panel) during the whole campaign, before and after 26th June. The shaded area represents the interquartile range for each variable.