Response to Referee 3- Rejano et al.:

We thank the reviewer for his/her comments and suggestions that have highly contributed to improve the quality of the manuscript. A point-by-point response to the reviewer's comments is included below. Reviewer'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.

Reviewer #3: This study deals with the complex dependence of CCN activity on the hygroscopicity of organic aerosols. Since CCN activity determines the indirect effect of aerosols on radiative forcing, the subject is of great interest. Using chemical and size distribution measurements, the authors study the role of OA hygroscopicity on CCN activity, for different atmospheric conditions. The positive matrix factorisation method was used to recover the relative contribution of OA with different oxidation levels, showing that medium and low oxidised OA are predominant, and that their contribution varies as a function of the vertical transport of PBL to the site. The originality of this study lies in the use of a neural network model to predict the amount of CCN using aerosol size distribution data, the fraction of OA and a factor of PMF, and radiation. This innovative tool gave the best results compared with assumptions about global chemical composition. The authors conclude by stressing the importance of taking into account the complexity of the aerosol and in particular its internal/external mixing. The manuscript is well structured and well written. The conclusion is clear and the message to be retained is correctly underlined. I consider that the manuscript can be published, after minor revision and responses to the following points:

Specific comments

1. Please add the equation which links κ, Dcrit and SS at the CCN activation in section 3.2 (κ -Kohler theory). You could then cite it after (eg. L 435).

We included the aforementioned equation in the manuscript and cited it accordingly.

2. L 281. Could you explain why the eBC increase starts earlier than the inorganics and OA increase during the day?

It's something that we are not able to give a conclusive explanation of this fact based on our measurements. However, considering all the possibilities to explain this fact we point out different issues:

The eBC particles are considered primary aerosol particles, while IA and OA are mainly dominated by secondary particles. For that reason, we can expect some delay between the increase of these species. In Figure 3 we can observe that eBC starts to increase at the same time as nucleation mode concentration. Since nucleation mode particles are difficult to form during NPF events at night and the eBC increase coincides with the nucleation mode increase, we can assume that there is some influence of local primary emission. On the other hand, the IA and OA increase during morning hours is caused by vertical transport. Therefore, IA and OA increase may take longer time to be observed.

We have modified the manuscript to clarify this discussion in lines 284-292:

"Based on these diurnal patterns, inorganic species are most likely transported from the Granada urban area due to upslope mountain breezes and the increase of the PBL height during daytime. OA also increased at midday, but the increase is sharper, reaching a maximum between 12:00-

16:00 UTC. OA exhibits a larger increase in concentration at midday hours compared to the other species (Figure 1b), which might suggest the influence of upslope transport but also additional sources of OA in the vicinity of the measurement site (such as local emissions or secondary processes as nucleation). Finally, eBC mass concentration increased more gradually, starting at 3:00 UTC and reaching a maximum at 11:00 UTC. The earlier increase of eBC with respect to IA and OA species might be related to some local primary emissions during the early morning, although most of the eBC observed at SNS is due to upslope transport at midday (Rejano et al., 2021)."

3. L 324. What are the wind direction and speed like during the two identified periods ?

At the measurement site, the wind direction is predominantly from the west and low wind speeds predominated during the campaign. Figure R1 shows the diurnal pattern of the wind speed (left panel) and direction (right panel) during the two periods. The wind direction was predominantly from the west. For the first half of the campaign, we can observe a clearer diurnal pattern and more influence of other wind directions (lower angles values respect the north direction during nighttime hours). During the second half of the campaign, a more constant wind direction is observed. As we can see in Figure R1 for the wind speed, the diurnal pattern shape is very similar during the whole campaign with higher wind speeds during the evening and lower at the central hours of the day. During the second half of the campaign, we can observe higher values of the wind speed respect to the first half of the campaign. A detailed analysis of the air masses and wind influence in aerosol composition is shown in Jaén et al. (2023), for the same measurement period.

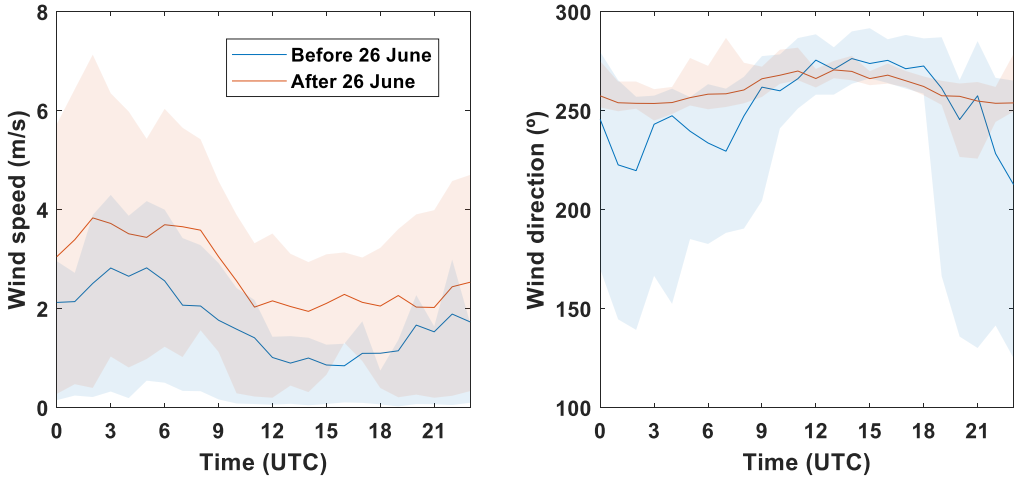

Figure R1. Mean diurnal pattern of the wind speed (left panel) and direction (right panel) during the whole campaign, before and after 26th June. The shaded area represents the interquartile range for each variable.

Despite the significance differences in the wind direction and speed between periods (confirmed by Wilcoxon rank-sum test), CCN predictions are barely affected by the different periods of the campaign. As we can see in Figure R2, we cannot observe a difference in the agreement between N_{CCN} observed and N_{CCN} predicted when considering the two-time period separately For the current analysis, we don´t consider that the changes in the wind conditions can explain the bias between measured and predicted CCN concentrations.

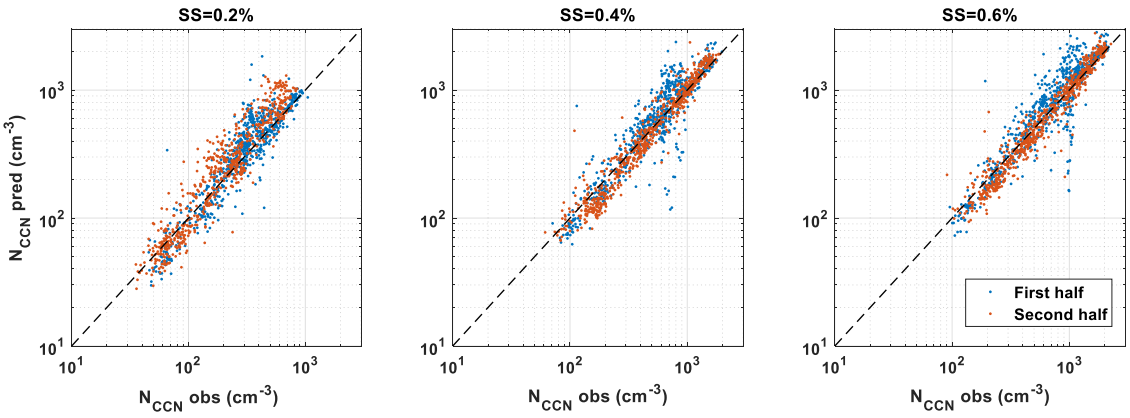

Figure R2. Scatter plot of predicted CCN concentrations (N_{CCN} pred) using OA scheme 3 as a function of observed CCN concentrations (N_{CCN} obs) during the two main periods of the campaign at different SS values.

L 365. What about the solar radiation diurnal profile role in the photochemical oxidation ?

We only mentioned additional factors that could contribute to photochemical oxidation, but we didn't mention solar radiation, that is the main factor that control these reactions. Therefore, we have added the solar irradiance diurnal pattern to Figure S3b (see below) and have updated the manuscript as follows:

Line 374-375: "*by SOA formation linked to photochemical oxidation induced by high solar irradiance values and high concentration of O_3 and NO_x (Figure S3a) together with high temperatures (Figure S3b)".*

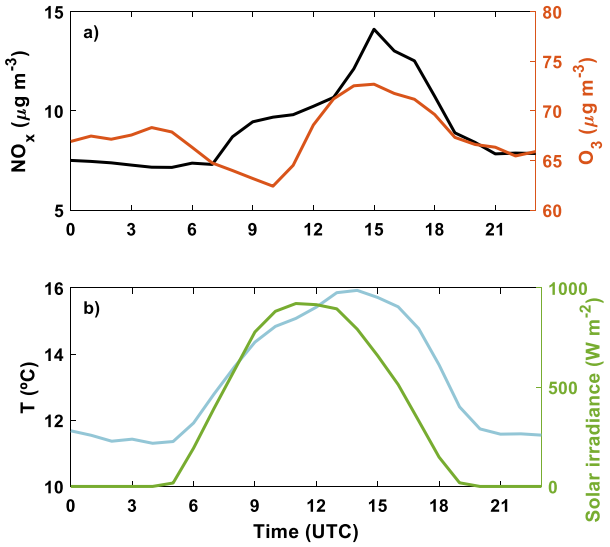

Figure S2. Mean diurnal evolution of a) NOx (left Y axis) and O3 concentration (right Y axis) and b) temperature (left Y axis) and solar global irradiance (right Y axis) along the campaign.

Also, we have corrected the manuscript according to the actual physical variable measured by the pyranometer during the campaign which is the global solar irradiance instead of total radiation.

L 413. What do you mean by "Time-dependent" ? If I've well understood, you described previously that in Scheme 3 specific κ values for LO-OOA and MO-OOA have been used to

take into account their relative contribution at SNS, but these values are fixed, and not varying as a function of the time. Could you please clarify this ?

According to reviewer's comment, we want to clarify that for Scheme 3 the calculation of κ_{OA} is obtained from the relative contribution of each OA factor. It means that κ_{OA} value changes over time since it is calculated including the OA temporal variability (each OA factor has a different hygroscopicity and each OA factor has a different contribution to the overall kappa during the day). In contrast, the overall κ computation of scheme 1 does not include the OA variability since that scheme assumes a constant κ_{OA} of 0.1. Also, for Scheme 2 we can state almost the same since the only difference is including the time variability of HOA in the overall κ calculation, which represents the smallest fraction of OA. We acknowledge that the term "time-dependent κ_{OA}" can be confusing and for that reason we have modified the manuscript in lines 421-423 to clarify this statement:

"*Scheme 3 exhibits a clearly different data distribution compared to schemes 1 and 2 due to the assumption of different κ values for the LO-OOA and MO-OOA factors.*"

L 443. Could you add the Dcrit values for SS= 0.2 and 0.6% to compare it with Dcrit=72 nm?

We agree with the useful reviewer comment, and we have added the mean Dcrit value at 0.2% and 0.6%. The text is modified in lines 451-452 as follows:

"In this case, the predicted N_{CCN} values overestimate the measurements at low SS (mean D_{crit} is 111±21 nm) and underestimate the measurements at high SS level (mean D_{crit} is 58±16 nm). At SS=0.4% the mean D_{crit} is 72±18 nm and…"

L 451 to 461. Did you observe a difference in the agreement between NCCN observed and NCCN calculated when considering the two-time period separately (before and after June 26th)?

We have observed some difference in some aerosol properties and atmospheric conditions in the two periods, but we have not observed a clear impact in the CCN closure agreement. To support this statement, we show the scatter plot of predicted N_{CCN} using OA scheme 3 respect to observed N_{CCN} for each time period for all three SS considered (Figure R2, discussed above). There are hardly any differences between the two measurement periods of the campaign in terms of CCN closure agreement. For that reason, we decided during the manuscript preparation not to focus on the differences between the two periods and focus on the importance of chemical composition and other variables (N80, radiation) which are measured at high-time resolution and provide better insight into the CCN predictions.

Technical corrections:

Please check the units notation in the text and the figures (eg. µg m-3 instead of µg/m3)

Thanks for the recommendation, we have corrected this issue along the manuscript.

Figure 4 : please add a note on the shaded areas representing the PDF of each variable

We thank the reviewer´s comment and we have updated the Figure 4 caption as follows:

"Figure 4. Violin plot of κ distribution data for the chemical schemes (κ_{chem}) and the CCN calculation at different SS values (κ_{CCN}). The boxes represent the interquartile distance, and the

asterisk is the mean value. The shaded area for each variable represents the probability density function (PDF)".

L 489. "is associated with"

Done

Figure 6: Please add a) and b) on the different panels.

We have modified Figure according to the reviewer's comment.

L 513.: please give the values used for κIA and κBC

According to that comment, we don´t use a specific value for κ_{IA}, it is derived from ToF-ACSM measurements and the hygroscopicity of each inorganic compound provided in the Table S1. The κ_{BC} is always assumed as 0. We have modified the manuscript to avoid misunderstanding. See the new text in lines 523-524 in the revised version:

"In this sub-section we calculate κ_{OA} from Equation 5 using the overall aerosol hygroscopicity as κ_{CCN}, κ_{IA} obtained from ToF-ACSM measurements assuming specific hygroscopicity values for each inorganic compound shown in Table S1 and κ_{BC} assumed as 0 (Cerully et al., 2015; Kuang et al., 2020; Thalman et al., 2017):"

Figure 8a. : Could you sort data by f44 values such that the points with the lowest f44 are plotted in first and the points with the highest f44 and plotted over (ie. yellow points overlapping the blue points) ?

According to the reviewer's comment, Figure R3 shows the implementation of his/her suggestion. Data are sorted according to the f44 values if they are below/above f44=0.25. Then, we plot small markers over datapoints with f44 value below 0.25 which are used in the filtered fit. Anyway, we prefer not to include this update in the revised version since we prefer to include the color-coded scale in this figure.

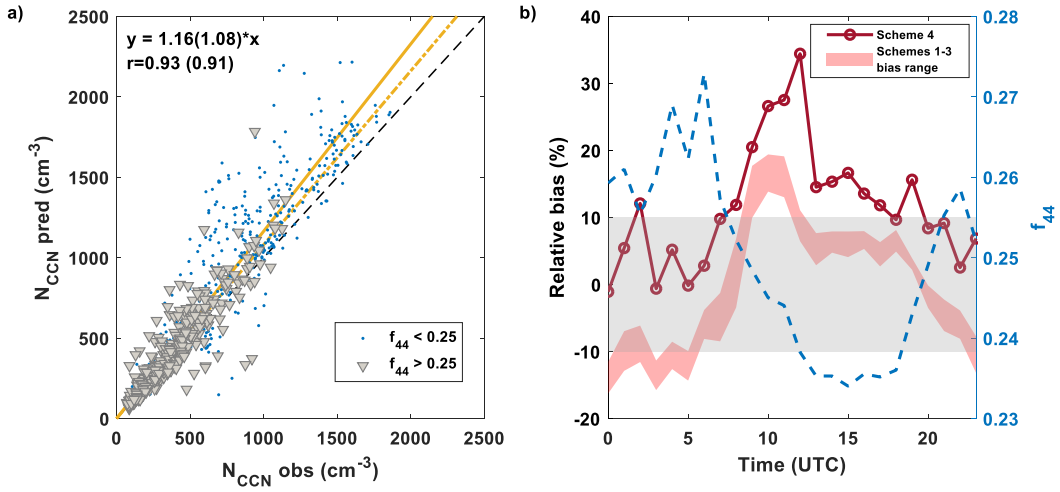

Figure R3. a) Scatter plot of predicted CCN concentrations (NCCN pred) as a function of observed CCN concentrations (NCCN obs) using the OA scheme 4. Datapoints with f44>0.25 are represented as grey triangles and f44<0.25 as small blue dots. The black dash line represents the 1:1 line. The linear equation and Pearson correlation coefficient (r) are also included for all data and for filtered data in parenthesis. 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 f44 (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.

L 588. Slopes and correlation coefficients don't correspond to the ones in Fig. 9a.

Thanks to the reviewer´s comment we have corrected this mistake in the manuscript and updated it with the correct values presented in Figure 9a.

L 603. R2 = 0.88 or 0.94 (cf. Fig. 9a) ?

We again thank the reviewer; the R value is 0.94 and we have corrected the manuscript according.

References

Jaén, C., Titos, G., Castillo, S., Casans, A., Rejano, F., Cazorla, A., Herrero, J., Alados-Arboledas, L., Grimalt, J. O., and van Drooge, B. L.: Diurnal source apportionment of organic and inorganic atmospheric particulate matter at a high-altitude mountain site under summer conditions (Sierra Nevada; Spain), Science of the Total Environment, 905, https://doi.org/10.1016/j.scitotenv.2023.167178, 2023.