Reply to the comments of Anonymous Referee #5

We thank Referee#5 for the comments provided to our manuscript. Here we try to reply to the comments at our best, indicating the changes we are going to make in the revised version of the manuscript. With "GC" we indicate the general comments.

GC1: "**The manuscript proposes that the skewness-kurtosis plane (the size distribution projected into the third and fourth moments) can be used to follow changes to the particle number size distribution (PNSD), and that four-parameter Johnson SB (JSB) distribution as being sufficient for observing changes to the PNSD in a way that maps to the skewness-kurtosis plane. The authors present PNSDs from four measurement campaigns under different NOx and meteorology conditions. This manuscript includes a few interesting ideas that are less well-known to the broader ACP community, and has potential for novelty and impact**."

We thank Referee#5 for this kind and encouraging comment to our manuscript.

GC2: "**However, at present time the manuscript is strongly recommended for revision and re-submission. The reason for this is recommendation that each of these ideas introduced are not fully developed. As a result, the reader is mostly left with an impression that what is demonstrated is that the size distribution changes when there are changes in meteorology or emission sources, which could be characterized more informatively using traditional approaches (number concentration, modes, etc.)**."

We thank Referee#5 for the comment. In the revised version of the manuscript will improve both the analyses and the presentation. In this manuscript we want to propose a new methodology of analysis of PNSD data, based on the skewness-kurtosis plane and the Johnson SB domain, which can be used also to summarize statistically the aerosol dynamics under meteorological conditions. We want to provide a tool to describe statistically and quantitatively the PNSD variation with meteorological conditions and emission sources. We used two datasets to describe the performance of such a tool. We will improve the presentation of our work, and add additional analyses as explained in the next point.

GC3: "**The conclusion that the JSB can be used to represent PNSDs does not appear to be well-supported by the material that is presented. As one of the other reviewer notes, PNSDs can be multimodal, and representing each of these modes well is in itself a challenge. There is no indication regarding the modality or quality of fit permitted by JSB. What is presented seems to be that the range of skewness and kurtosis in observed PNSDs fall within the range that can be represented by the JSB distribution except at high concentrations. Furthermore, it is not demonstrated that JSB outperforms other parametric distributions for representing PNSDs (except for the reason of having four fitting parameters), and the authors even note in the conclusions that the other parametric representations may be adequate**."

Thanks again for this comment, also raised by Referee#4 (GC4). We know that the lognormal or the mixture of lognormals are generally used in the literature to represent the PNSD data. We agree with the Reviewers that it is a nice idea to compare JSB distribution with those commonly used in the literature, in order to see if the JSB outperforms (or not) other parametric distributions in representing PNSDs. We have analyzed this topic and in the revised version of the manuscript, we would like to address this issue.

Lets consider a mixture of two lognormals. According to the OPC size particle classes, a mixture of two lognormals is sufficient to keep the modes of the analyzed datasets. Let assume that X follows a mixture of two lognormal distributions. Its density $f_X(x)$ is

$$f_X(x) = \sum_{i=1}^{2} \pi_i f_i(x)$$

Where $f_i(x)$ and π_i are respectively the (lognormal) density and the weight of the i-th component. The k-th order moment respect the origin of the mixture

$$E[X^k] = \mu^{(k)}$$

has been calculated as

$$\mu^{(k)} = \sum_{i=1}^{2} \pi_i \mu_i^{(k)}$$

where $\mu_i^{(k)}$ is the k-th order moment respect the origin of the i-th component. $\mu_i^{(2)}$, $\mu_i^{(3)}$, $\mu_i^{4)}$ can be written in terms of mean $\mu_i^{(1)}$, standard deviation σ_i, skewness β_3 and kurtosis β_4 of the i-th component as

$$\mu_i^{(2)} = \sigma_i^2 + (\mu_i^{(1)})^2 \quad (1)$$

$$\mu_i^{(3)} = \beta_{3_i}\sigma_i^3 + 3\mu_i^{(1)}\sigma_i^2 + (\mu_i^{(1)})^3 \quad (2)$$

$$\mu_i^{(4)} = \beta_{4_i}\sigma_i^4 + 4\mu_i^{(1)}\beta_{3_i}\sigma_i^3 + 6(\mu_i^{(1)})^2\sigma_i^2 + (\mu_i^{(1)})^4 \quad (3)$$

The skewness β_3 and kurtosis β_4 of the mixture are respectively

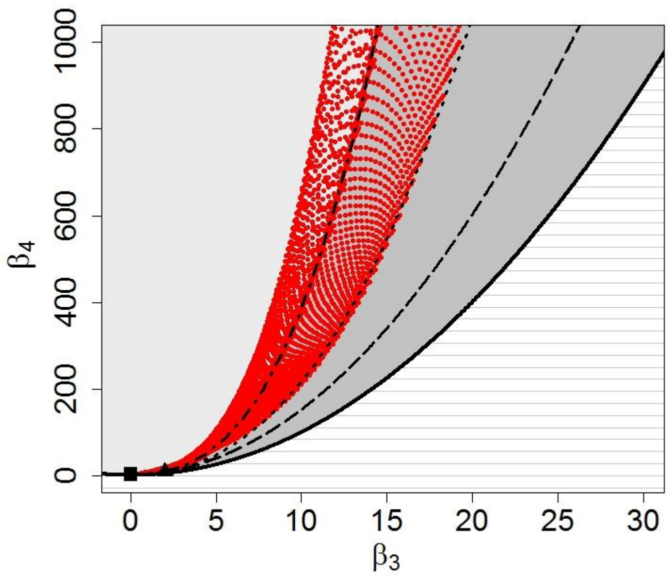
$$\beta_3 = \frac{\mu^{(3)} - 3\mu^{(1)}\mu^{(2)} + 2(\mu^{(1)})^3}{(\mu^{(2)} - (\mu^{(1)})^2)^{3/2}} \quad (4)$$

$$\beta_4 = \frac{\mu^{(4)} - 4\mu^{(1)}\mu^{(3)} + 6\mu^{(2)}(\mu^{(1)})^2 - 3(\mu^{(1)})^4}{(\mu^{(2)} - (\mu^{(1)})^2)^2}. \quad (5)$$

Substituting Eq.s (1-3) into Eq.s (6,7), it is possible to express skewness and kurtosis of the mixture as a function of mean, standard deviation, skewness and kurtosis of each component.

In the skewness-kurtosis plane, the domain of a mixture of two Lognormal distributions has been determined numerically through a Montecarlo simulation by simulating couples of lognormal distributions with parameter μ in the range (-10:0.1:10), parameter σ in the range (0:0.1:10) and weight π in the range (0:0.1:10). The skewness and the kurtosis can be calculated by using equations (4,5).

We compared the Johnson SB domain (in dark grey) with the domain of a mixture of two lognormals (indicated with red dots), as reported here in the figure. This is an original issue never investigated in the literature and we will be happy to deal it in the revised manuscript.



As shown in the figure, the Johnson SB distribution has a wider domain respect to the mixture of two lognormals, indicating that the Johnson SB distribution is more versatile respect to the mixture of two lognormals in representing the OPC data.

GC4: "**Regarding the use of the skewness-kurtosis (S-K) plane, does it provide more information that cannot be achieved by examining other parameters of the PNSD conventionally used (e.g., first and second moments of lognormally transformed data)? Given the long history of modeling PNSDs, the mode gives some indication of whether the dominant source is likely anthropogenic or biogenic; the geometric standard deviation may be related to the extent of atmospheric dispersion. It is not clear from the results presented whether 1) changes in PSNDs in the S-K plane cannot be detected in a conventional parameter space, and 2) any approximate delineations can be proposed that link physical processes to regions in S-K that could demonstrate its usefulness**."

Thank you for this comment, we will improve the description of the skewness-kurtosis plane in the revised manuscript, accordingly.

The S-K has been used here as a powerful and easy-to-use statistical tool for the identification of the statistical distributions which best represent the PNSDs. The peculiarity of the S-K moment-ratio diagram is that every theoretical distribution occupies a specific domain in the plane, that can be a point, a line or an area. Thus, this plane can be used as a diagnostic tool for the identification of distributions able to model given datasets, comparing the domain of the theoretical distributions and the sample variability. For these reasons we can state that the use of this plane provides different information respect the examinations of other statistical parameters, conventionally used for the analysis of PNSDs. In the manuscript we have described our first tentative to study the changes in PNSDs pattern in the S-K plane related to physical processes, in particular the influence of primary and secondary aerosol particles. In the future, we would like to examine more in depth this very interesting and, in our opinion, very promising analysis.