Reply to the comments of Anonymous Referee #4

We thank Referee#4 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" indicate general comment, while with "MC" minor comment.

GC1: "**This manuscript describes the applicability of the Johnson SB distribution for fitting particle size distributions measured by optical particle counters operated at two sites in Italy. The paper first focuses on assessment of the merits of the expression form by examining the fraction of measurements that lie within the region on the skewness-kurtosis plane that is bounded by the Johnson SB envelope of possible solutions. The patterns of data point clusters on that plane are then linked with meteorological and environmental conditions to suggest the more general use in describing the sources and processing responsible for an observed size distribution. The manuscript is reasonably well-written but would require some editing prior to publication**."

We thank Referee#4 for this kind and general comment to our manuscript.

GC2: "**I have identified several specific concerns I have with the manuscript below. More generally though, this simply does not seem to be appropriate for ACP. The dataset described is very limited and rather uninteresting when not complemented by other aerosol and trace gas measurements. More importantly, the dataset is not really the focus of the paper, but rather the technique to describe the dataset is. Thus, in its current form this would be more appropriate for a journal such as AMT. If the authors chose to shift the emphasis more toward the size distributions I still feel that because of the limitations of the dataset this would be better suited for another journal**."

We thank Referee#4 for this comment. Here we want to clarify that in this work 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 used two datasets to illustrate the methodology. The methodology is quite general, and with general implications for the assessment of aerosol dynamics. We intend to apply this to other datasets in the near future, as explained in the next point. In this work, we have mainly focused on physical issues, rather than chemical issues, influencing the variability of PNSD. We will investigate chemical issues in a further study. In the revised version of the manuscript, we clarify this issue to improve the presentation of our work. We think that, for the wide breath of the work, ACP is the proper editorial place.

GC3: "**It could be that collaboration with researchers involved in more comprehensive measurement campaigns could be valuable for evaluating the utility of the techniques described here for understanding influences on size distributions. It seems the authors have considerable experience with statistical methods and data analysis, but not with air quality. The relevance of this is that there is far too much text describing rather fundamental details about aerosol sources and sinks and meteorology**."

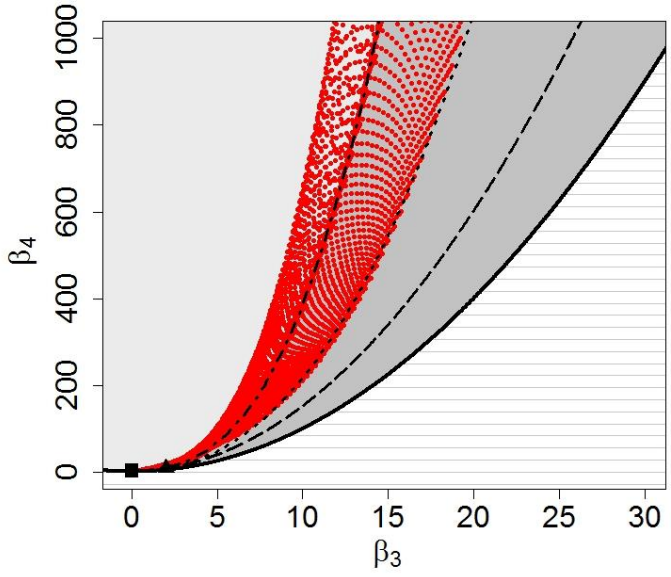
Thanks for this suggestion. We are planning to continue the collaboration with researchers belonging to Institute of Atmospheric Sciences and Climate –ISAC, National Research Council, Italy. They are regularly involved in comprehensive field campaigns (see e.g. activities documented at the website http://actris-cimone.isac.cnr.it/) measuring aerosol dynamics from the physical and chemical point of views, in addition to meteorological conditions. So, in the near future, we intend to analyze the data of existing comprehensive field campaigns in order to confirm and extend the results obtained in this work.

GC3: "**The averaged and example size distributions shown in Figures 1 and 3 reveal a common characteristic of distributions measured by OPCs - erroneous peaks and troughs that are often linked with features in the scattering intensity vs. size relationship for the optical geometry of the instrument. The fact that they are retained in the distributions suggests the authors didn't invest much time in calibration of the instruments and processing of the data. But more relevant for this paper, those features will influence any fit of the distributions and the location on the S-K diagrams. There is no discussion of these features or their impact**."

We thank Referee#4 for this comment, also pointed out by Referee#3 (see SC9). In the revised version of the manuscript we will fix this issue, i.e., the inflections in the distributions between about 0.4 um and 0.8 um, due to ambiguities in the scattering function for the particular angle of the OPC. We will operate an averaging across the bins, as suggested by Referee#3. So, in the revised version of the manuscript we will report the new figures (1 and 3), and remake the representation of data in the skewness-kurtosis plane.

GC4: "**The authors argue that the Johnson SB distribution is more appropriate for fitting the particle size distributions than more commonly used forms such as the lognormal. But they neglect to discuss the utility of the lognormal because of the direct connection of the parameters describing it with physically meaningful elements of the aerosol distribution (i.e., N, Dp_mean, SD) and the ability to describe variation of those parameters accompanying things such as atmospheric processing. Furthermore, the manuscript largely dismisses lognormals based on the difference between the data points and the single lognormal point on the S-K diagrams. But does the representation as a point presume that only one lognormal is used to fit the distribution? In practice, multiple lognormals are almost always used**."

We thank Referee#4 for pointing at our attention this interesting comment. In the revised version of the manuscript, we would like to address this issue by reporting in the skewness-kurtosis plane, the domain of a mixture of two lognormals (indicated with red dots). According to the OPC size particle classes, a mixture of two distributions is sufficient to keep the modes of the analyzed datasets. We have compared the Johnson SB domain (in dark grey) with the domain of a mixture of two lognormals, as reported here in the figure. This is an original issue never investigated in the literature and we are happy to deal it in the revised manuscript.



In the revised manuscript, we plan to add an appendix where we describe how we have calculated the β3-β4 domain of the mixture. From the figure, it is possible to see that 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.

Minor issues:

MC1: "**Page 4, line 1: Grimm model what**?"

The GRIMM model used at Oga San Colombano is "GRIMM 107 Environcheck" as well as at Pascal-Città Studi. We will specify this in the revised manuscript.

MC2: "**Page 4, line 7: What is the basis for the assertion that the composition is different between the two sites. It undoubtedly is, but this still needs some support**."

In the revised version of the manuscript, we will add some support to this sentence. Specifically, we would like to write "Oga San Colombano shows a higher relative contribution of organic aerosol, likely of secondary origin, as suggested by a higher organic to elemental carbon ratio (Sandrini et al., 2014). In addition, Milano shows a higher nitrate to sulfate ratio, in agreement with a stronger impact from combustion sources, such as traffic and industrial emissions (Perrone et al., 2012")."

Perrone M.G. (2012). Sources of high PM2.5 concentrations in Milan, Northern Italy: Molecular marker data and CMB modelling, Science of the Total Environment 414, 343–355.

Sandrini S. et al. (2014). Spatial and seasonal variability of carbonaceous aerosol across Italy, Atmospheric Environment, 99, 587-598.

MC3: "**Page 4, line 26: Nitrogen dioxide and nitric oxide are not aerosol compounds**."

We acknowledge the Referee for pointing out the mistake. The sentence in modified as follows:

"The influence of primary aerosol sources and meteorology on PNSD has been investigated for the site of Milan. To study the effect of pollutant concentration we have used nitrogen dioxide (NO2) and nitrogen oxide (NO) measurements collected with a chemiluminescence technique following the requirements of European Standard EN 14211: 2005: Ambient Air Quality".

MC4: "**Page 6, line 24: Total particle count is meaningless to readers. I assume the authors simply need to divide this by the product of flow rate and sample time to report it in concentration. Additionally, it seems there is confusion about the upper threshold value because it is written both as 10ˆ4 and as 100000 (=10ˆ5)**."

Thank you for the comment. There was an error in Table 2: the threshold value is 10^5 (100000) and not 10^4. Regarding the total particle count, we think that this statistical and physical measure is not meaning less, because it allows the readers to have a direct and simple measure of the load of aerosol particles that can be recorded in a minute and to compare the different cases, changing season and/or site.

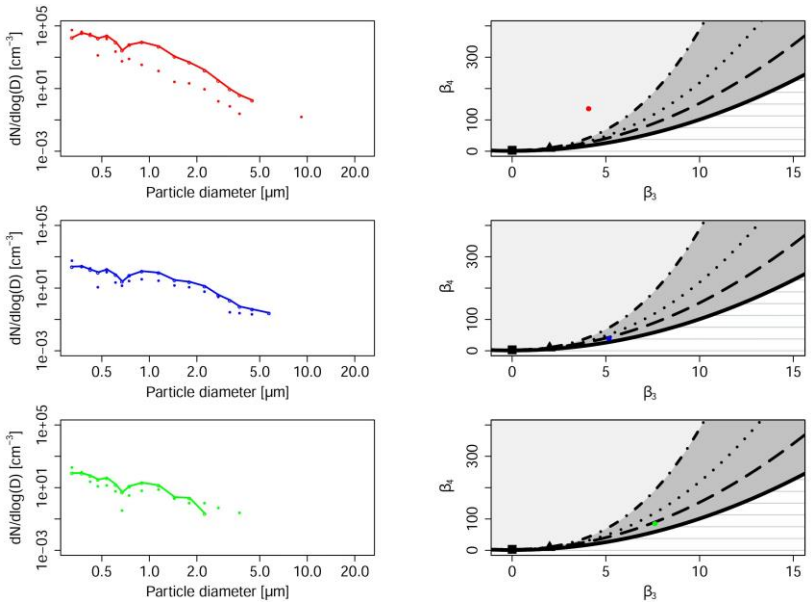
MC5: "**Page 10, top: The NO2 to NOx ratio will be largely dependent on time of day, which will confound the interpretation of its influence on the patterns in the S-K diagrams**."

We agree with the Referee that the NO2 to NOx ratio depends on the time of the day, as primary emissions from traffic do as well. In addition, previous measurements at the urban site here investigated show that the NO2 to NOx ratio anti-correlates with black carbon to PM1 ratio (a marker of primary traffic emissions in this area) and correlates with the ratio of secondary to primary aerosol species (i.e. secondary organic and inorganic aerosol to black carbon plus primary organic aerosol ratio). Unfortunately, during the presented experiment no data on aerosol chemical composition was available to direct evaluate the contribution of primary and secondary components. Thus, we decided to use the NO2 to NOx ratio as a proxy of polluted air mass ageing. To improve clarity, we would like to modify the manuscript as follows:

"It follows that the NO2 to NOx ratio can provide a measure of the oxidative capacity of the atmosphere (Rao and George, 2014; Fernández-Guisuraga et al., 2016). In addition, measurements performed in Milan during different field experiments show that NO2 and NOx ratio anti-correlates with black carbon to PM1 ratio, confirming that the NO2 to NOx in urban area is an indicator of the relevance of secondary pollutant formation over primary traffic emissions.
In Fig. 4 we have again reported the skewness-kurtosis plane, where we have plotted in black the data points of MI1 (upper panel) and MI2 (lower panel). Then, we have selected the data points belonging to minutes characterized by values of the ratio NOx/NO2 between 1 and 1.1 (red dots – highly oxidizing atmosphere), 1.1 and 1.5 (orange dots – slightly oxidizing atmosphere), 1.5 and 3 (yellow – little oxidizing atmosphere), greater than 3 (green - no oxidizing atmosphere). Both the two datasets are characterized by high aerosol numbers and high percentages of data points outside JSB domain (74 % and 65% respectively). The percentages of data points characterized by a ratio NOx/NO2 greater than 3 are around 50 %, indicating a prevalence of the primary traffic aerosol contribution. If we select only the data points outside the JSB domain, the percentage of data points with ratio greater than 3 (strong prevalence of primary aerosols) is 56 % for MI1 and 50 % for MI2. While, the percentage of data points with ratio greater than 1.5 (light or strong prevalence of primary aerosols) is 88 % for MI1 and 67 % for MI2. These findings support our hypothesis that in urban sites during winter season the increase of primary traffic contributes to the shifts of (β_3, β_4) couples in the skewness-kurtosis plane."

MC6: "**Figure 3: The use of an unnecessarily large y-axis range obscures the information in the distributions and the quality of the fits**."

Thank you for the comment. We will modify Fig.3 following your suggestion.



MC7: "**Figure 4: The differences among these graphs are pretty modest**."

In our opinion, the difference between the graphs are not so modest. The discrepancies are highlighted by the numbers showed in the legends and by the explanation of Paragraph 4.2.