Replies to the comments by the Referee Professor Giuseppe Petrucci:

First of all, we would like to thank Professor Giuseppe Petrucci from the University of Vermont for reviewing our manuscript and for his helpful comments to improve it. In the following we will comment on the individual points. The referee's comments are shown in black and our answers in blue. The referee's comments refer to the version of the manuscript submitted for review and our answers refer to the revised version of the manuscript.

Comments and Replies:

This is a well written manuscript describing thorough experimental results for a significantly improved method of measuring INPs.

1) Fig. 10: I don't understand why there are two distinct branches for aerosol generation.

   The two separate branches for aerosol generation are required for the INP measurement of the externally mixed Snomax and NaCl particles (as described in Sect. 4.1.1 and Sect. 5). Referee#2 has also pointed out that the reference to Fig. 10 in Sect. 4.1.3 is missing. The corresponding reference to Fig. 10 has now been added in Sect. 4.1.3 (pg. 15, line 329).

2) Please elaborate How robust are the fits to equations 4 and 5? There are so many adjustable parameters in each, I wonder whether similarly good fits could be obtained with diverse sets of fitted parameters? In other words, is the fit a true global minimum in error or could there possibly be several local minima?

   The fit according to Eq. 4 is well constrained, since the parameters are quite independent. To describe a scan we need to specify the position and background (z_0 or y_0 for detection laser 1 or detection laser 2 and b_g) values. These are quite trivial to determine in a complete scan. Further we use only two parameters to represent the width and the height of the signal, since there is no reason why the width of the particle beam (σ_p) should be dependent on the width of the laser beam (r_{DL}) and vice versa. Additionally, these two parameters represent real physical parameters, which help to choose reasonable initial values and to judge the results. We tried out randomly different meaningful initial values, and we always ended up at the same result. Further, we randomly set either the fit parameters for the effective laser width or the particle beam width as constant. However, by these tests no similarly good fits could be obtained.
   In contrast, the fit according to Eq. 5 requires further conditions for the determination of the fitted parameters (Sect. 4.4), since the effective detection width at the ablation spot ($r_{DL(AL)}$) and the effective width of the ablation laser (r_{AL}) cannot be considered independently of each other. To avoid this problem, the parameters for the particle beam width (σ_p), effective width of the detection (r_{DL}) and the y_0-position for the distribution of the detection efficiency (y_0) were first determined at the second detection laser using Eq. 4 before the application of Eq. 5. Subsequently, the resulting particle beam width and effective detection width could be scaled up

to the distance to the ablation laser, resulting in $r_{DL(AL)}$ and $\sigma_{p(AL)}$. Together with y_0 (included in $y_{\overline{DA}}$, which is the difference between the center position of the detection y_0 and the center position of the hit rate $y_{0(AL)}$), the values of the three parameters were kept constant during the fits using Eq. 5. Thus, the fit again uses only independent parameters: 1) the effective width of the ablation laser, 2) the position of the ablation spot and 3) a background value.

Because the values of the fit parameters in Eq. 4 and Eq. 5 are real physical parameters, they are confined to certain boundaries. Within the confined parameter space, different combinations for initial values were tested. To the best of our knowledge, the given set of parameters are the global minima in this confined parameter space. In Sect. 4.2.2 (pg 20, line 411-414) and Sect. 4.4 (pg 28, line 598-605) we have added further explanations to the parameters of Eq. 4 and Eq. 5.

3) Some of the figures don't use symbols (lines are different colors) or use the same symbol (but, again, lines of different colors) to distinguish between experiments. This may be problematic for someone who is visually impaired or doesn't have access to a color version of the manuscript. I would suggest using different symbols for each experiment to facilitate understanding the figures.

   Many thanks for the hint. We modified Fig. 13, Fig. 19 and Fig. 22 such that the different measurement series can be better recognized by the symbols.

4) I found the use of "coincidences" terminology to be confusing. In my experience with this field, "coincidence" is typically reserved for when a second particle enters the timing region before the first particle has passed through second detection laser for estimating particle aerodynamic diameter. In other words, I have always seen this word used to describe a possible experimental complication or error.

   To avoid irritation, we decided to use the term "sized particles" as an alternative term to "coincidence" and replaced $d_{50(coinc)}$ by $d_{50(SizedP)}$. The terms were exchanged in the text of the new manuscript:
   - in Sect.2, pg 5 line 100-101
   - in Sect.4.2.1, pg 17 line 348
   - in Sect.4.3.1, pg 21 line 439, 447-454
   - in Sect.4.3.1, pg 22 Fig. 13 caption
   - in Sect.4.3.1, pg 22 line 461-464
   - in Sect.4.3.1, pg 23 line 478-480
   - in Sect.4.3.2, pg 25 line 527
   - in Sect.4.4, pg 29 line 612, 613
   - in Sect.4.5.6, pg 36 line 767-768
   - in Sect.6, pg 40 line 837, 838

   In the Supplement:

- in Sect.S5, pg 6 line 91-95
- in Sect.S6, pg 6-7 line 103-110
- in Sect.S9, pg 8 line 129-135
- in Sect.S9, Fig S7 caption
- in Sect.S9, pg 9 line 138-145
- in Sect.S15.1, pg 15 line 263

5) Sec. 4.2.1 is unclear to me, although I am not familiar with these types of counting instruments. Why sum OPC size channels from 0.65 to3.0 µm to represent a smaller range of particle sizes (1.8 to 2.6 µm)? Similarly, for particles greater than 2.6 µm?

   We agree that Sect. 4.2.1 "Definition of particle detection efficiency" was a bit misleading. Also for Referee #2 the background of the size selection in the OPC was not clearly visible from the text. Basically the summation over several size channels in the OPC was only performed for PSL particles with known size, because despite monodisperse PSL particles a size mode over several size channels could be observed in the OPC. First of all, a selection of size channels is not necessary as long as a DMA was used. With increasing size of the supermicron-particles it becomes more difficult to use our DMA in a suitable way. For monodisperse particles the applicable size range of the DMA can be extended, but not to all particle sizes we used. Therefore, the large particle sizes were measured without DMA. Without DMA, however, we have the effect that in the OPC a second size mode was observed in the smallest size channels. We attribute this second mode to small droplets or substances dissolved in the PSL suspension. To avoid that these unwanted small particles have an influence on our measurements, we removed this mode by selecting the size channels in the OPC and corrected the particle concentration measured with the OPC accordingly. The use of the size selection in the OPC is now described in more detail in Sect. 4.2.1 (pg.17, line: 357-364).

6) The authors refer several times to a size-dependent particle beam shift. Why is that? If particles are focused onto the ALS central axis, what causes beam pointing differences for different diameters?

   It can be observed from the measurements that the particle beam focus shifts slightly depending on size. The main reason for this is probably production-related imperfections in real lenses. Similar observations were made by other groups, e.g. Huffman et al. (2005; Sect. 3.4.; page: 1152; DOI: 10.1080/02786820500423782). The reference to Huffman et al. (2005) was added in Sect. 4.5.2, pg 31, line 662.