

Response to Referee number 4

7 February 2019

The authors thank Referee #4 for his/her detailed, expertise and valuable comments to further improve and clarify the MS. We have considered all recommendations and made appropriate alterations. Our specific responses to the comments are as follows, while the detailed textual modifications are highlighted in red or by crossing out in the revised MS.

Page8, line 256: the mean new-to-old rate ratios of J_6 were 1.23 for city center and 1.20 for near-city background. I would expect that traffic emission causes overestimation of formation rates because it is a source of nanoparticles. Please specify why correcting traffic emission in formation rates calculation gives higher J_6 .

1. Several modifications were simultaneously adopted in the revisited and refined calculations protocol of the new set of J_6 for the measurement years of 2008–2009 (city centre) and 2012–2013 (near-city background). They include the subtraction of particle number concentrations emitted by road traffic from N_{6-25} , which usually leads to a decrease in the coagulation loss and loss due to growth out from the diameter range of 6–25 nm, and which can sensitively influence the slope of the concentration change in time (dN_{6-25}/dt) in a positive or negative manner depending on the actual time evolution of perturbing emission source. In addition to that, the time interval in which this slope is considered to be constant was determined within a completely new treatment. We would also like to mention the mean relative contribution of the concentration increment, coagulation loss and growth out from the diameter interval to J_6 have different weights of 71%, 17% and 12%, respectively (lines 246–249 of the original MS, and Table S1). Furthermore, J_6 itself also depends on GR_{10} , which makes the relationships even more complex. These explain why the overall effect of urban influence generally resulted in increased dynamic properties. The mean new-to-old ratio for J_6 was larger for the city centre (1.23) than for the near-city background (1.20). It should also be emphasized that the re-calculation mainly affected the individual dynamic properties with relatively small absolute values. The whole process is considered as a methodological improvement over the years of research. The MS was amended by a more detailed description of Equation (1) and by a brief explanation of the issues above.

Section 4.2: Discussion on NPF events frequency should include conditions of NPF days as well as non-NPF days. Properties discussed in the section are only based on events days. This could be misleading because non-events day conditions are not discussed.

2. Information on the average CS (calculated for whole days), gas-phase H₂SO₄ proxy, GRad, air *T* and concentration of some criteria pollutants on non-nucleation days were partly included now. Many properties are, however, biased by the seasonal cycle of solar electromagnetic radiation via the seasonal variation of new particle formation frequency, and therefore, their interpretation needs special attention. They are to be fully utilized and explained for investigating the changes in annual pattern of relative nucleation frequency over the years, and a more comprehensive evaluation and discussion is to be realized in a future study outlined in response no. 5.

Line 484 conclude gas-phase H₂SO₄ are unlikely to be the limiting factor of NPF occurrence in Carpathian Basin including Budapest from the misalignment between the monthly occurrence frequency and the other properties. To make this statement solid, H₂SO₄ proxy for events days and non-events days is needed.

3. Averages of several atmospheric properties involved in the H₂SO₄ proxy were derived separately for the event days and non-event days, their effects were briefly discussed, and as a result of it, the statement mentioned was removed.

Page18, line548: Direct compare the numbers of J and GR or saying something contribute equally to the formation of particle and to their growth don't make sense because they are different physical variables. Correlation between J and GR are expected but comparison of the regression line with $J6=GR10$ doesn't give any useful information.

4. The sentence was modified to express clearly that we mean that the chemical species available in the air affect the formation rate and growth rate differently at the 2 urban sites. This could partially be caused by differences in chemical composition. We reformulated the whole statement completely and turned it from a conclusion into a working hypothesis because a rigorous statistical treatment would indeed require larger variability in the near-city background data.

Page20: Lacking correlation with single parameters to J/GR doesn't tell too much as NPF is controlled by multiple parameters. With the size of the data set, authors could perform analysis on subsets of the data with certain constrains like temperature or H2SO4 proxy.

5. Evaluation of the overall data set by multistatistical methods is indeed planned. This comprehensive evaluation is, however, to be accomplished after some markers or proxies for biogenic emission sources (such as e.g. photosynthetical activity) are also included. The extension of the present MS by this comprehensive statistical analysis would not fit among the present objectives and would not be advantageous or feasible considering both the length and timing of this MS as well. The present study can be considered as the first step in a larger statistical evaluation process and which supplied orienting ideas on the specific directions to proceed in. This perspective further study is very briefly mentioned in the Conclusions section now.

Page 20, line 625 to 636 and figure 4: $GR/H_2SO_4 \text{ proxy} = b * (1/H_2SO_4 \text{ proxy}) + a$ is equivalent to $a * H_2SO_4 \text{ proxy} + b = GR$. A negative 'a' means the higher H2SO4, the lower the GR. This is contradictory to the interpretation of increasing gas-phase H2SO4 related to larger contribution of other vapors to particle growth. Another concern would be special care should be taken when combine H2SO4 proxy at sub-urban site and urban site as the VOCs and NOx condition could be totally different but not taken into consideration.

6. Figure 4 and the related discussion were removed from the MS to avoid any misunderstanding or incompleteness. The remaining part of the section was restructured, split into shorter pieces and clarified.

Page 24, line 739: To make the full potential of the data set, more detailed studies on the contribution of NPF to regional particle concentration could be performed.

7. We fully agree on this item and will proceed in that direction in the future.

Spelling

Line 113: mean see level-> mean sea level

Line 751: cloud -> CLOUD

8. The typing errors were corrected.

In addition to the issues above, we also adopted some smaller changes and added a few recent papers as references to further improve the MS.

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