Response to Referee number 3

7 February 2019

The authors thank Referee #3 for his/her work. We have considered all comments thoroughly and profoundly. Unfortunately, many of them cannot be accepted at all or fully. Our specific responses are as follows, while the textual modifications are highlighted in red or by crossing out in the revised MS.

The paper summarizes a large body of data and tries to extract information on the underlying processes of new particle formation. This is rather difficult as the lowest particle size they measure is 6 nm and growth rates can only be determined around 10 nm.

1. The lower measurable particle diameter limit of DMPS/SMPS systems is important for identification of NPF and growth events and further data treatment. Evaluations of this type of atmospheric measurements are mostly based on particle diameter range <20 nm (e.g. Kulmala et al., Nat. Protoc., 7, 1651–1667, 2012). In order to separate reliably the NPF and growth events from huge emission peaks which can occur in cities and which can temporarily influence the size intervals down to even smaller diameters, it is highly preferable to have the lower limit below 10 nm. Our limit value of 6 nm was proved to be already satisfactory since it allows to identify and separate different particle generation processes (see e.g. Fig. S1b of the present MS and Salma et al., Atmos. Chem. Phys. 16, 7837–7851, 2016). It is also worth mentioning that from 6 urban cites involved in a recent global analysis of NPF over long-term measurements (Nieminen et al., Atmos. Chem. Phys., 18, 14737–14756, 2018), the lower diameter limit was 3 nm at 2 sites, it was 6 nm at 3 locations, while it was 11 nm at 1 of the sites, and both the J_{nuc} and GR were determined for the diameter interval of 10–25 nm. All these indicate that in atmospheric studies, our experimental systems and evaluation protocols seem completely adequate for the time being.

The authors do not provide much more insight than in the paper of Niemienen et al., where they are coauthors of, except that the results are now based on a larger data set.

2. The goals of the paper mentioned in the comment were largely different from our aims. We can list several important insights explicitly as examples which are part of the present MS and which were not dealt with in the referred paper. They primarily include 1) the evaluation

and discussion of monthly distributions of J_6 an GR_{10} together with their relationships with nucleation occurrence frequency and relevant atmospheric parameters, 2) timing properties of NPF and growth events, 3) refinements of *J* and GR calculations dedicated to urban environments, 4) statistical distributions of J_6 an GR_{10}, 5) occurrence and properties of extreme events and events with broad onset. These items represent a considerable piece of novelty and new knowledge. Furthermore, the results and conclusions are based on 247 quantifiable NPF and growth events in an urban environment, which means a rather strong background. Finally, we can quote from the Summary and conclusions section of the Nieminen et al. paper (p. 14750): "For future studies, it would be very valuable to make detailed investigations on the interdependencies among J_{nuc}, GR, and NPF event frequency, at both single measurement sites and among sites of seemingly similar environmental characteristics." This is exactly what we did in our MS. In addition to our arguments, we can offer to all persons involved a recent and excellent review paper of Kerminen et al., Environ. Res. Lett. 13 (2018) 103003, 2018 dedicated to field observations, which also gives a scientific outlook and summarizes future research needs, and which can help putting our present results and conclusions more adequately into a scientific frame of international atmospheric NPF and particle growth studies.

Also the fact that the sulphuric acid proxy does not correlate with J6 and GR10 has already been reported in an earlier paper. Although sulphuric acid does only contribute 12.3% to GR10 it does not mean that it is not relevant for NPF (line 608).

3. This conclusion was mentioned in the MS as a minor outcome of the study with the purpose of confirming earlier results (as explicitly stated in the line specified in the comment). The related sentence was modified now to emphasize the key role of H_2SO_4 in the nucleation process and early particle growth.

Many studies have shown a relation between NPF rates measured at small sizes and sulphuric acid, while the growth is dominated by organics. In Figure 4 the authors relate basically reciprocal (sulfuric acid proxy) versus reciprocal (sulfuric acid proxy) modulated by the GR. The linear relation is not surprising and does not lead to any conclusions. As the authors repeat several times in the paper NPF and growth is a complex process. Nevertheless, they test only relations of one single parameter with J6 or GR10. Why do the authors not make an attempt to combine parameters?

4. Figure 4 and the related discussion were removed from the MS to avoid any misunderstanding or incompleteness. The remaining part was also restructured, split into shorter pieces and clarified. 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. See also response no. 5.

It is known that low temperature stabilizes nucleating clusters and that organics promote growth and thus the survival probability. It might thus be worthwhile to look for a proxy representing condensing organics.

5. Chemical species including organics participating in the urban atmospheric NPF and growth were investigated in an intensive international measurement campaign in Budapest over March-May 2018 by deploying API TOF-MS with/without CI, PSM, AIS and DMPS systems. Some potential proxy values for condensing organics are under evaluation. This was mentioned in the Conclusions section, and it is further emphasized and explained in the revised version.

I also question if daily averages are the appropriate parameter to inquire NPF mechanisms. Although it is worth to report on this large data set, I find the paper does not provide much new information and I do not see what the authors' "consequences of dynamic and timing properties" are as announced in the title.

6. Daily averages were calculated for those variables which change slowly over a day (e.g. $[SO_2]$). For some other variables such as CS, we constrained the averaging for the time intervals from t_1 to t_2, thus over the nucleation process itself. Some other variables, such as the gas-phase H_2SO_4 proxy, were characterized by their daily maximum. They are accurate

specified in Section 4.1 Ranges and averages. As far as the novelty of the MS is concerned, we must refer again to the list in response no. 2. The main conclusions drawn from the dynamic and timing properties are readily collected in the Abstract.

Line 151 and 494: What is the detection limit of the SO2 detector? Are the low SO2 concentrations measured significantly above DL?

7. The limit of determination (LOD) of the SO_2 analyzer system applied is approximately 0.2 μg m^{-3}. More than 98% of the hourly-mean concentrations were above the LOD. The information is also included into the text now.

Line 318-319: I do not see a trend in particle concentrations.

8. The annual medians for the city centre in the measurement years 2008–2009, 2013–2014, 2014–2015, 2015–2016 and 2017–2018 are as follows: 11.5×10^3, 9.7×10^3, 9.3×10^3, 7.5×10^3 and 10.6×10^3 cm^{-3}, respectively. The first 4 data indicate unambiguously a decreasing tendency, while the last data point may look somewhat different. Rigorous statistical evaluation of the joint data set of particle number concentrations in various size fractions over a decennial time interval from 03–11–2008 to 02–11–2018 is in progress, and its preliminary results in the one hand, confirm the decreasing tendency, and in the other hand, reveal some fine structure to this dependency. This information was added to the revised MS.

Table 2: the authors use local time as time base. We know that photolysis is an important driver of sulfuric acid and oxidant production. Would it not be more appropriate to use time after sunrise for starting time?

9. The suggestion represents an option, which can be consider for specific studies. In the present MS, we selected the local time as the time base of most data on purpose and as a compromise because we had experienced in several earlier investigations (e.g. Salma et al., Atmos. Environ., 92, 154–161, 2014) that it is the daily activity time pattern of inhabitants that substantially influences or determines many atmospheric sources and important processes in Budapest. It was explained in lines 123–125 of the original MS, and a reference

for the statement was included as well. The timing parameters of the NPF were given in UTC+1.

Line 441: how can you conclude that NPF is not sensitive to temperature? Indeed the yearly average does not vary much, but is the yearly average really important? What matters more is the temperature during an event in combination with formation rates of nucleating and condensing vapors.

10. The sentence mentioned was replaced from its original location to section 4.2 Monthly distributions. It was largely corrected and extended to a discussion by involving the temperature profiles on nucleation and non-nucleation days, biogenic emissions, photochemistry and results from other international studies.

Line 498: What do you mean by "CO is less certain"?

11. The related sentence was modified to express our intention better that the variability of CO was without obvious tendentious temporal structure or feature.

Figure 2: Is the low value of H2SO4-proxy in May real or an artefact? What is the reason for that?

12. It is the monthly distribution of daily maximum gas-phase H_2SO_4 proxy that is shown in Fig. 2. The mean value for May represents 23 days. Its low value seems to be influenced by enhanced effect of multiplying relatively low GRad with relatively small [SO_2] for a few days particularly in 2015 (which was a strange year as far as the monthly distribution of nucleation frequency as well is concerned; see Fig. 1 of this response). The reliability of the monthly data is to be increased with the length of the overall data sets in the future. This additional information is added now in a synthetized manner to the text.

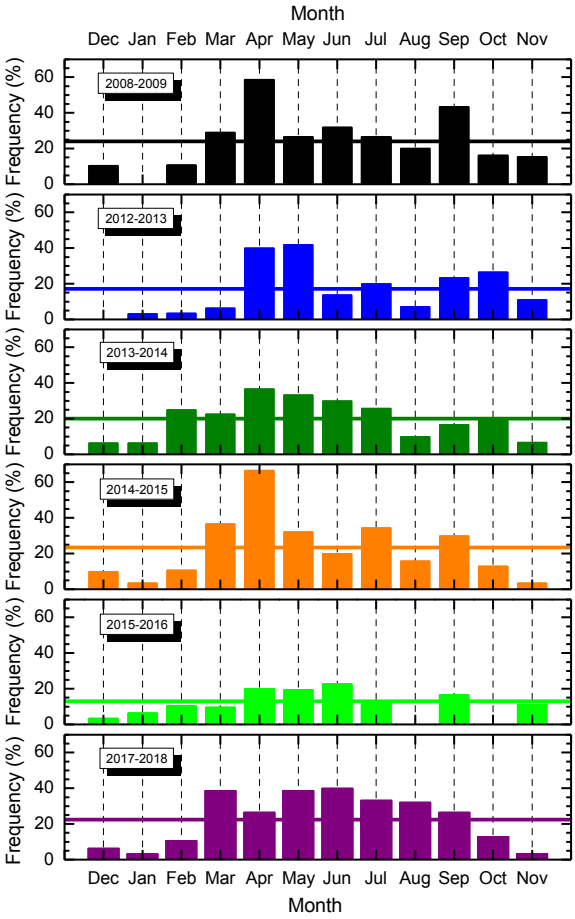

Figure 1. Monthly distribution of relative nucleation frequency in Budapest for measurement years of 2008–2009, 2012–2013, 2013–2014, 2014–2015, 2015–2016 and 2017–2018. The horizontal lines indicate the annual mean frequency. More information is given in the MS.

Line 545: This is not the line of equality. The units of each axis is different. There is also no discussion of this relation with respect to literature, e.g. Nieminen et al.

13. We used the expression “line of equality” in its broader sense, hence when the abscissa and ordinate are on the same scale even they do not have the same units. To the explicit request of Referee #3, however, we can change it to another expression, e.g. “line with a slope of 1”. We also amended the discussion of the relationship between *J* and GR at several places by considering the international results available in the literature.

Line 547: The difference between slopes for centre and near–city station is not very convincing. If the authors would also restrict the city centre plots to GR<10 nm/h I expect a large scatter of the slopes. The near-city data do not seem to be different from the other data.

14. We were aware of this inherent limitation mainly caused by smaller dynamic properties (and partly by shorter measurement time interval) in the near-city background than in the city centre, and expressed it by ourselves in lines 554–556 of the original MS. Now, we reformulated the statement completely and turned it from a conclusion into a working hypothesis because a rigorous statistical treatment would indeed require stronger/larger variability in the near-city background data.

Line 559: It should say "that leads to J6>0". J=0 cannot be measured and is meaningless.

15. The suggestion was accepted and adopted.

Line 565: what do you mean by "weak phenomena"?

16. The related sentence was modified to express that we mean the NPF events with relatively small particle formation rate (weak events).

Line 611ff: This explanation is unclear. Surely, GR need to be faster in urban areas but that does not mean that there could be no correlation. Simply speaking higher CS should lead to lower GR. Apparently, a positive correlation is found, isn't it? This would be counterintuitive.

17. The GR of newly formed particles to larger sizes is primarily coupled to 1) CS, which is further linked to the entire aerosol particle population (including the newly formed particles, thus the NPF itself), 2) to the total concentration and some physicochemical properties of non-volatile gaseous compounds and 3) to their production rate in the gas phase from aerosol precursor compounds (e.g. Kerminen et al., Environ. Res. Lett. 13 (2018) 103003, 2018). Understanding these couplings is essential when analyzing atmospheric observations. It is not fully plausible to make intuitive expectations on simplified paired relationships, for instance between CS and GR, under such complexity. Therefore, we stuck to the experimental data and are to contribute to the phenomenological picture on the system of relationships in this part of the MS, which will be eventually leading to a comprehensive and

qualitative explanation of the connections in the future. We extended the sentence briefly with these additional arguments and explanation.

Section 4.4 needs much improvement.

18. We split the section into shorter parts and clarified it by clearer formulations.

Line 739: Where does this number of contribution of NPF to total particle concentration come from? How was the analysis done?

19. Typical number of particles generated by an NPF and growth event on a nucleation day was roughly estimated by considering the median J_6 and median duration of nucleation, Δt (their distribution function is lognormal; see Table 2) and mean relative coagulation loss, F_{coag} (see Table S1) as: $J_6 \times \Delta t \times (1 - F_{coag}) = 4.6 \times 180 \times 60 \times 0.83 = 41 \times 10^3$ cm$^{-3} \approx 10^4$ cm^{-3}. This concentration is in line with other results achieved by nucleation strength factor according to which the particle number concentration due to NPF and growth process on a general nucleation day is increased by a factor of approximately 2 (Salma et al., Atmos. Chem. Phys., 17, 15007–15017, 2017). A more detailed description of the estimation process and the mathematical expression utilized are added now together with the last reference mentioned.

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.

Finally, we think that the comments of Referee #3 eventually helped us to formulate our thoughts and ideas better. We appreciate this. We wish, however, to emphasize that the major message of the MS lies in a considerable variety of contributions to the emerging research field of urban atmospheric NPF and growth, which have been becoming possible and increasingly recognized thank to gradually generating, several-year long, semi-continuous, critically evaluated, complex and coherent data sets. We further stressed this aspect of the MS now in the Conclusion section and added a new opening sentence to the Abstract as well.

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