Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-115-AC1, 2016

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Regional effect on urban atmospheric nucleation" *by* Imre Salma et al.

Imre Salma et al.

salma@chem.elte.hu

Received and published: 9 June 2016

The authors thank Referee #1 for his/her detailed and valuable comments to further improve and clarify the MS. We have considered all recommendations, and made the appropriate alterations. Our specific responses to the comments are as follows.

Comment 1.i Determining the quantities used in the analysis: GR: -How much does the choice of the method for assessing GR (the log-normal fitting method) affect the obtained values? The maximum concentration method (see e.g. Kulmala et al., 2012) is also very often used; would the values change if this method was used instead? -Is GR corrected for coagulational losses, as proposed by Leppä et al. (Atmos. Chem. Phys. 11, 4939–4955, 2011)? -Are the GRs from literature (page 5, lines 39-41) determined for the same size range (and with the same method)?

Response to Comment 1.i All GR values were determined by an identical method, i.e.

by the log-normal fitting method, and the possible influence of the different calculation methods was not studied. Uncertainties and possible systematic difference in dynamic properties caused by various evaluation approaches represent a relevant issue, which could be an objective of a separate dedicated study. The GR calculations were based on Kulmala et al., 2012, and the coagulation losses were not considered. The GR values cited from literature were not necessarily determined for the same size range and with the same method, and they serve for comparative purposes as the first orientation in the available data sets. It is also important to be aware of that there are banana plots with a broad onset of up to 3–4 hours, and the termination of their dynamic properties accurately represents a larger challenge than selection of the calculation method itself. In addition, it is worth mentioning here that an overview study on the global picture of observationally based estimate on NPF is under preparation by an international expert team, which is to handle the issue raised by the Referee.

Comment 1.ii J: -It is not clear how J is in practice determined. The first paragraph of Section 2.3 lists the works by Kulmala et al. (2012), Kulmala et al. (2001) and Dal Maso et al. (2002) as references to how the DMPS data analysis was done; however, all these give somewhat different approaches for determining a formation rate J. -Does J depend on the determined GR as in the expression of Kulmala et al. (2012)? If so, the dependence should be brought up in the discussion for clarity. -Is J determined for dp = 6 nm for all the data? Information on the size is missing in e.g. Section 2.3, and Figs. 6 and 7. If the size is always 6 nm, how is J determined for the K-puszta site, for which the lower limit of the DMPS is 10 nm?

Response to Comment 1.ii Formation rate J_d of particles with a diameter d nm was computed as:

J_d=dN_nuc/dt+CoagS_nuc x N_nuc + GR/delta(d) x N_nuc,

where N_nuc is the number concentration of nucleation-mode particles, CoagS_nuc is their coagulation scavenging efficiency, GR is the growth rate in the size range [d,

d+delta(d)], and t is time. The nucleated particles were estimated by N_6–25. It was assumed that the intensity of the NPF is constant for a certain time interval, and, hence, dN_6–25/dt was determined as the slope of the linear function N6–25 versus time within an interval where the dependence could be approximated by a linear fit. The referred publications indicate the continuous evolution of the calculation concept, and its final version (Kulmala et al., 2012) was adopted in the present MS. The text was modified now to express this accurately, and to emphasize also that J indeed depends on GR. Formation rates were determined for particles with a diameter of 6 nm, and it was specified or corrected at several places in the text and figures.

Comment 1.iii t1: It is stated that "The time t1 also includes the time shift that accounts for the particle growth from the stable neutral cluster mode at (1.5+-0.4) nm to the smallest detectable diameter limit of the DMPS systems". How was this done exactly? Presumably this calculation requires a growth rate for the sub-detection sizes; which values were assumed? How much does the inclusion of this time shift affect the determined times t1?

âĂČ Response to Comment 1.iii The time shift – that accounts for the particle growth from approximately 2 nm of the stable cluster mode (Kulmala et al., 2013) to the smallest detectable diameter limit of the DMPS systems (6 nm) – was calculated by adopting the GR value in the size window nearest to it in size space. This approximation can result in an underestimation of the shift by up to 30% since GR increases with d in this size range (Kulmala et al., 2012). It is noted that the shifts were mostly smaller than 30–40 min, which is acceptable with respect to the uncertainty of the starting time parameter t1, to the time resolution of the DMPS system, and to the ordinary dynamics of atmospheric processes. The text was extended to include these pieces of information. See the highlighted part of the marked-up MS.

Comment 2 Page 4, lines 23-26: The quantity tau is used to assess if the air mass was transported from one site to the other for tau = 1 and tau Âń 1; for tau > 1, it is only stated that " tau > 1 is often caused by large (>7 m s–1) WSs." Can anything be

hypothesised about the origin of the air mass in the last case? Also, tau isn't really discussed in the Results section; could it be e.g. added to Table 2?

Response to Comment 2 Quantity tau has sensible meaning for 1 column and 2 rows of Table 2, and therefore, it was not added there. Instead, we extended its discussion in the text. See the highlighted part of the marked-up MS. It is difficult to arrive at any solid conclusion for tau >1 since the related cases of very high WS happened only twice, and so, a representative evaluation could not be achieved.

Comment 3 Figure 3 and discussion on page 6: What does the dividing line describe, i.e. is there a physical reason to fit a line to the (sink, source)-data? (Isn't it quite clear also without the line that most of the red dots corresponding to events are at higher source values?)

Response to Comment 3 The dividing line in Fig. 3 was calculated by discriminant analysis. It was determined on one hand by the middle point between the arithmetic mean of the data subset for nucleation days and that for non-nucleation days, and on the other hand, it is perpendicular to the connecting interval between these 2 means (Hamed et al., 2010). We would like to keep the line in the figure since it is advantageous in fast and unambiguous orientation and serves as a discrete visual limit.

Comment 4 Page 7, lines 21-22: Discussion on the effect of the condensation sink on NPF events: "This implies that the CS affected the NPF in the Budapest area, and that it can have preventing influence on the events. In contrast, the mean CS values for Kpuszta station showed much less or even little effect." Why is the effect smaller in the K-puszta site? Are the absolute values of CS lower than in Budapest?

Response to Comment 4 Median values of CS for Budapest and K-puszta station are shown in Table 4 for four combinations of conditions, i.e for the time intervals when NPF events were identified in both Budapest and K-puszta station (BpY&KpY), event in Budapest and no event at K-puszta station (BpY&KpN), no event in Budapest and event at K-puszta station (BpN&KpY), and no event in both Budapest and K-puszta station

(BpN&KpN). It turns out from them that the CS varies in a broader range in Budapest among these cases and that its high values are associated with no NPF, while CS changes in a smaller interval at K-puszta station. The CS depends sensitively on the concentration and size-distribution of pre-existing aerosol particles. At K-puszta station, the average particle number concentrations are substantially smaller, and hence, the CS values and their changes are smaller as well. The text was extended to express these more precisely. See the highlighted part of the marked-up MS.

Comment 5 Page 8, line 23: "Fig. 8" should read "Fig. 7". Moreover, this Figure is only briefly mentioned in one sentence; it should be discussed more in the text to justify its existence.

Response to Comment 5 The figure number was corrected, and it was also more discussed now. See the highlighted part of the marked-up MS.

âĂČ Comment 6 Figures: i) Figs. 4, 5 and 7: It would be useful for the reader to estimate the H2SO4 proxy also in units molec./cm3 (e.g. as an additional y-axis), as well as the source in units molec./cm3/s (Fig. 3). ii) It would be useful to have also the hours, not only the day of year, in the time axis of Fig. 5. Also, in the bottom panel, the scaling factors could be written in the y-axes labels for clarity, and the legend could be removed (as now both the y-labels and the legend give essentially the same information)?

Response to Comment 6 The gas-phase H2SO4 proxy value was calculated as [SO2]$\times$GRad/CS for intensities >10 W m–2. Its average or extreme values for Budapest and K-puszta station were also expressed in absolute concentrations for several cases as well by using the scaling factor k between the proxy value and H2SO4 concentration of k=1.4$\times$10ˆ–7$\times$ GRadˆ–0.70 (Petäjä et al., Sulfuric acid and OH concentrations in a boreal forest site, Atmos. Chem. Phys. 9, 7435–7448, 2009). These concentrations were given in Table 4 or on page 6, lines 23–24. As far as the figures are concerned, we would prefer using the proxy without adopting the scaling factor

since it was derived specifically for a remote boreal site as an empirical relationship. Urban areas are expected to differ from remote regions (Mikkonen et al., A statistical proxy for sulphuric acid concentration, Atmos. Chem. Phys. 11, 11319–11334, 2011), and the GRad involved implicitly in the scaling factor can distort the relationships and trends investigated in the figures. We emphasize this by a separate note which was added now into the text, and with a new reference. See the highlighted part of the marked-up MS. In addition, we also think that Fig. 5 showing the size distribution surface plot and 8 related meteorological, pollutant gas an aerosol data in 3 panels would possibly become over-sophisticated to follow by an extra axis. The minor ticks of the time axis on Fig. 5 represent 3 hour time intervals, and thus, the hourly dependency for a day can be recognised. We originally selected the way of indicating the figure scaling factors for various independent variables in the figure legends. This seems virtually equivalent with the solution suggested by the Referee.

Technical comments: -Page 1: The sentence "Despite the fact that NPF..." starting on line 28 should be tied to the previous sentence; on its own it doesn't mean anything. Also, the following sentence should be somehow modified, as it's not entirely clear to what kind of studies the expression "for such studies" refers. -Page 5, lines 26-27: The English of the sentence "At present knowledge, advection of nucleating air masses cannot be excluded only in a few cases" is somewhat unclear; please modify. -Page 7, lines 17-18: Tie the sentence "In spite of the fact that the estimated reduction..." to the previous sentence. -Reference list: Seven references to works of one of the authors seems a bit unbalanced; the authors should cite also works other than their own in these occurrences.

Response to Technical comments The whole MS was checked for typing and language errors. The specific examples mentioned by the Referee were, naturally, all corrected. See the highlighted part of the marked-up MS. There are a few and recent publications on the NPF in the Carpathian Basin. They were cited since they were regarded to be relevant for the context and background of the special objective of the present paper.

Their number is, however, not extraordinary large in comparison with the total number of references.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-115, 2016.