

## ***Interactive comment on “Measurement, growth types and shrinkage of newly formed aerosol particles at an urban research platform” by I. Salma et al.***

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Received and published: 5 June 2016

The authors thank Referee #2 for his/her detailed and valuable comments to further improve and clarify the MS. We have considered all recommendations, and made the appropriate alterations. We would also like to mention that there was a scatter in some properties of particle shrinkage on an event-by-event basis, which was likely caused by the complex relationships between the particular property and the atmospheric conditions. The fluctuation is expected to be decreased by separating the shrinkage events into groups according to major atmospheric conditions or some other selection principles, and by evaluating them separately for these groups. At the moment, this is unfortunately hindered by their limited number. We plan a dedicated measurement

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campaign and evaluation of particle shrinkage on a longer data sets. Our specific responses to the comments are as follows.

Comment 1 Page 3, lines 38–40: How representative are these gas-phase measurements for BpART (has there been inter-comparison measurements between BpART and the air quality site 1.6 km away)? Could there be larger differences in the concentrations between the two sites when the air masses do not pass both sites?

Response to Comment 1 The municipal air quality measurement stations measure criteria air pollutant at several locations in Budapest, and the BpART research platform has not been compared to them, or more specifically to the closest municipal station in upwind prevailing wind direction yet. It could be realized in the future. It is noted, however, that SO<sub>2</sub> concentration is ordinary distributed without larger spatial differences within the city (Salma et al., Comprehensive characterisation of atmospheric aerosols in Budapest, Hungary: physicochemical properties of inorganic species, Atmos. Environ. 35, 4367–4378, 2001), and, therefore, its actual value at the research platform is less influenced by air mass directions. In addition, an important advantage of the selected research location at the river Danube (mentioned also in the MS) is that it receives a well-mixed, average air masses from the city centre. The text was extended to include this information, and the reference was added. See page 4, lines 1–2.

Comment 2 Page 7, lines 8–10: What are these properties related to NPF events that varied substantially?

Response to Comment 2 We referred to N<sub>6–25</sub> and gas-phase H<sub>2</sub>SO<sub>4</sub> proxy. They were listed explicitly now.

Comment 3 Page 7, lines 37–39: There seems to be a growing nucleation mode present after 9 p.m (Fig 11b). Could this growing NPF also be related to the grass cutting?

Response to Comment 3 The particle growth observable in Fig. 11b around 21 UTC+2

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cannot be considered as NPF event because 1) it does not fulfil the requirements for NPF (Dal Maso et al., Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II, Hyytiälä, Finland, *Boreal Environ. Res.* 10, 323–336, 2005), and 2) it also appeared after the sunset. There were no nocturnal NPF events identified at all in Budapest since 2008. The growth was likely related to condensation on existing (Aitken-mode) particles which was caused by changes in local meteorology (e.g. decrease in air temperature and/or air mixing) and atmospheric chemistry. A similar example was discussed earlier (Salma et al., Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment, *Atmos. Chem. Phys.* 11, 1339–1353, 2011). A sentence was added to include this argument. See page 8, lines 1–4.

Comment 4 Page 8, lines 27–29: What is the typical time that particles were in high-vacuum conditions in the TEM, without evaporating?

Response to Comment 4 The freshly nucleated particles could withstand the conditions of high vacuum without evaporating for several tens of minutes. It was specified explicitly in the MS together with a reference. See page 8, line 36.

Comment 5 Page 8, lines 34–36: What does it mean that the growth and shrinkage phases “were selected by linear approximation”?

Response to Comment 5 The GR and SR of the nucleation-mode particles were derived as the slope of the fitted linear line of several particle number median diameter (NMD) data in their time evolution plot (Kulmala et al., Measurement of the nucleation of atmospheric aerosol particles, *Nat. Protoc.* 7, 1651–1667, 2012). These two time intervals were utilised separately as the time intervals for averaging aerosol-related and meteorological properties characteristic for the growth and shrinkage conditions. They are shown in Table 3. The text was modified accordingly. See page 8, lines 40–page 9, line 2.

Comment 6 Page 9, lines 35–37: In the conclusions the decreasing global radiation

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and H<sub>2</sub>SO<sub>4</sub>- proxy concentration are highlighted as the main reasons for the observed particle shrinkage events. However, the shrinkage episodes seem to be quite rare (8 out of 178 NPF events), whereas the global radiation and the proxy typically decrease in the afternoons of every day. Could the authors clarify their conclusion? Are the change rates of global radiation and proxy during the shrinkage episodes (reported in Table 3) much higher than on a typical afternoon of an NPF-day without particle shrinkage (for example on the day shown in Fig 8a)?

Response to Comment 6 The explanation was extended by condensable vapours in general since the mean contribution of H<sub>2</sub>SO<sub>4</sub> condensation to the particle GR was estimated to be 12.3% in Budapest (Salma et al., Regional effect on urban atmospheric nucleation, *Atmos. Chem. Phys. Discuss.* doi:10.5194/acp-2016-115, in review, 2016). Oxidation products of VOC are expected to play a more important role in the growth of relatively larger particles (with a diameter above ca. 20 nm). The shrinkage is likely caused by a joint effect of multiple factors including substantially decreased concentration of condensable vapours and GRad. Sudden changes in GRad could inhibit NPF or reverse the ongoing particle growth (Baranizadeh et al., The effect of cloudiness on new-particle formation: investigation of radiation levels, *Boreal Environ. Res.* 19, 343–354, 2014.) See also the arguments in the introductory part of this Authors’ response. The text of the MS was modified to express these details in more detail. See page 10, lines 2–4.

Comment 7 Page 15, Table 3: What is the reason for using both mean and median values in Table 3? The numbers in the last column are the mean G/S ratios of medians over growth and shrinkage phases, and as such are somewhat different from the ratios of the reported mean values over the growth and shrinkage phases.

Response to Comment 7 The atmospheric properties in Table 3 were expressed by their mean values. The ratios of these properties for the growth and shrinkage phases were, however, calculated by using their median values since the latter values are less sensitive to large fluctuations than the means, and therefore, they seem more

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advantageous for reliability and propagation of errors.

Comment 8 Page 15, Table 3: Reporting the H<sub>2</sub>SO<sub>4</sub> proxy in the units molec/cm<sup>3</sup> instead of  $\mu\text{g m}^{-3} \text{ W s}$  would make it easier to compare the proxy values to those from other sites. I know that the scaling coefficient in Petäjä et al. (2009) is based only on measurements in Hyytiälä, Finland, but it is still widely used in the literature, and would thus enable at least a semi-quantitative comparison of the Budapest proxy values with other sites.

Response to Comment 8 The gas-phase H<sub>2</sub>SO<sub>4</sub> proxy value was calculated as  $[\text{SO}_2] \times \text{GRad}/\text{CS}$  for intensities  $>10 \text{ W m}^{-2}$ . We would prefer using it without the scaling factor  $k$  between the proxy value and H<sub>2</sub>SO<sub>4</sub> concentration since the scaling factor was derived specifically for a remote boreal site as an empirical relationship:  $k=1.4 \times 10^{-7} \times \text{GRad}^{-0.70}$  (Petäjä et al., Sulfuric acid and OH concentrations in a boreal forest site, *Atmos. Chem. Phys.* 9, 7435–7448, 2009). Urban areas can/are expected to differ from the remote regions (Mikkonen et al., A statistical proxy for sulphuric acid concentration, *Atmos. Chem. Phys.* 11, 11319–11334, 2011), and the GRad involved in the scaling factor can distort the relationships investigated. As far as the comparison of our data to other sites is concerned, we referred now to a recent paper (Salma et al., Regional effect on urban atmospheric nucleation, *Atmos. Chem. Phys. Discuss.* doi:10.5194/acp-2016-115, in review, 2016) in which the scaling factor was adopted for the Budapest H<sub>2</sub>SO<sub>4</sub> data.

Response to Technical comments All technical comments were accepted and adopted.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, doi:10.5194/acp-2016-239, 2016.