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Interactive comment on "Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment" by I. Salma et al.

I. Salma et al.

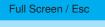
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The authors thank Referee #1 for his/her detailed comments for further clarifying and improving the ACPD paper. We have considered all recommendations and revised the MS substantially. The most important alterations include:

- condensation sink, gas-phase H2SO4 proxy and residence times for H2SO4 vapour and freshly formed particles were calculated, and a whole new section (including a new figure and a table) on their importance, relevance and results was added, as requested by some of the Referees and the handling Editor;

- diurnal variation of particle number concentrations in two size fractions calculated



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separately for event days and non-event days were prepared, and the results were included into the interpretation as a new section including a new figure, as requested by one of the Referees;

- the section on mean number size distributions was substantially revised, new results and additional interpretation were included;

- further evidence was provided at many places in the text, e.g., to support that the Fig. 4b type contour plots are related to direct emissions, to explain the smaller time variation in daily mean number concentrations with respect to PM10 mass concentrations;

- the statement on the presence of aged aerosol before new particle formation event starts (that was considered to be not fully justified) was completely removed;

- improved and explicit interpretation at several places with more detailed background information, firmer arguments and better explanations;

- Conclusions were reformulated substantially and were clarified.

We believe that the MS contains a large number of valuable information, and that its major weaknesses were all removed.

Response to Specific Comments

Response to Comment P13690 L16 The sentence on the presence of aged aerosol was removed from the Abstract and text.

Response to Comments P13690 L19 and L20 The sentence was clarified by a short explanation of the related concentration ratio which was added into the Abstract as well. The exact meaning of the ratio is further explained in the text.

Response to Comment P13691 L23 The sentence was modified to express that the indicated size interval refers to the median diameter (of an aerosol population), and

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two related references were now included.

Response to Comment P13692 L9 The maximum vehicle circulation and maximum ultrafine particle concentrations from morning rush hours in Budapest usually occur between 7:30 and 9:00. Expression "early morning" was changed to this time period.

Response to Comment P13692, L25 Primary ultrafine particles refer to the aerosol particles with an electric mobility diameter <100 nm directly emitted into the air. Their major emission sources in cities include road traffic, fossil fuel burning and biomass burning. We mainly use this expression to differentiate these particles from the secondary ultrafine particles, i.e., from the particles that are formed by nucleation.

Response to Comment P13694 L19 The specific research objectives of the MS were formulated on P13694 L18–22. Characterization of ultrafine particles involving new particle formation and growth has not been performed in many cities in the world so far. It is thought that each large city has its own specialities which can have implications on public health risk for its inhabitants. The measurement site in Budapest was described on P13964 L25 – P13965 L11. Wind channel that is often formed above the river Danube and that passes the central part of the city belongs to these unique aspects. Moreover, long-term continuous measurements in all types of environments are desirable for better understanding and quantifying the general mechanisms, the extent of production, and the role of ultrafine particles in more detail. Our paper contributes to this goal.

Response to Comment P13694 L20 A sentence was added now to emphasize that the paper indeed contains the first information on the production, growth and properties of ultrafine particles in the whole region over a longer-run (yearly) time scale.

Response to Comments P13696 L14 and P13701 L24 The size separation in the DMA was tested and calibrated by using spherical PSL particles with a diameter of 400 nm. Size separation depends sensitively on the air flow rates. The flows were checked and were adjusted almost every week. Moreover, we performed several comparative

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performance tests with the DMPS during the measurement campaign, and found satisfactory agreements. These all mean that the instrumental error (e.g., miscalibration of larger particles) on the extent of the observed shift (100 nm vs. 150–250 nm) can be rejected. We think that the shift is related to several other factors. The fact that we measure dry diameters, the presence of multiple Aitken modes from different sources and their overlap with the accumulation mode, together with the consequence of this to the fitting method can all contribute to the explanation. It turned out in a recent comparative investigation that the median diameters of the accumulation mode for Prague, Czech Republic obtained by a different evaluation method were very similar to our results. This indicates that the shift could be a specificity of urban environments. As to the wet diameters, we have no experimental information on the hygroscopic properties of (ultrafine) aerosol particles in Budapest, and this is to be one of our next research steps. We completed the text now by a list of contributing explanations.

Response to Comment P13696 L19 Complex meteorological characterization of the measurement site would enlarge the length of the paper, and it is more related to the reasons of new particle formation. The description was completed now by a brief overview on basic meteorological parameters.

Response to Comment P13700 L17 We agree with the Reviewer that the explanation for the relatively small time variation in the daily median particle number concentration was missing from this part. It was actually given later on P13702 L14–17: "New particle formation events occur when the number concentration levels are relatively low, and they increase the concentrations considerably. Combination of these two effects smoothes the variability in the daily average concentrations mentioned in Sect. 3.1." We added now a reference to the latter explanation at this particular point as well. This part of the MS was reformulated.

Response to Comments P13701 L6 and P13706 L24 A whole new section was included on the condensation sink. Effects of atmospheric concentrations of condensing species and driving forces for condensation were discussed in more detail within this

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section. Reasons for new particle formation are to be dealt with in a separate paper.

Response to Comment P13701 L24 See Response to Comment P13696 L14.

Response to Comment P13702 L6 The sentence in question was removed.

Response to Comment P13702 L12 The explanation of the concentration ratio was improved. It is the ratio of the mean area of the nucleation mode to the total number concentration measured just before the new particle formation event. The value of this ratio can be larger or smaller than unity. The ratio expressively indicates the contribution of new particle formation events to the total particle number.

Response to Comment P13703 L3 As suggested by the Reviewer, we added a figure that shows the diurnal variation of the mean ultrafine and N_100–1000 concentrations for non-nucleation and nucleation days, and their conclusions were discussed now in the text. We also included new related information dealing with the diurnal vehicle circulation in Budapest, added a related reference, and complemented the site description (Sect. 2.1) with the basic information on the composition of vehicle fleet in Hungary. The fact that the particle concentration level in the afternoon rush hours remains high for longer time than for the morning rush hours can be related to several factors. More stable boundary layer mixing height is only one of them. Larger mixing in the afternoon and early evening is another important factor. The afternoon rush hours take longer than the morning peak, and smaller wind speed during late afternoons can also contribute. A more complex interpretation was included now to explain this.

Response to Comment P13703 L5 We removed the adjective local from the sentence and emphasized now its photo-chemical nature, i.e., the growth due to photo-chemical formation of condensing gases.

Response to Comment P13703 L9 Formulation of the sentences was modified. (By writing "they" we referred originally to the authors of the cited paper.) Our interpretation was substantially refined, more explicit formulations were adopted, and it was clarified

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that the arguments are to be considered as an alternative explanation.

Response to Comment P13703 L11 Mean concentrations of some markers for photochemical activity – and expectedly, concentrations of other photo-chemically derived compounds – were larger on these days than the yearly averages. The information was included into the text.

Response to Comment P13703 L19 At present, we only have indirect evidences to back the outlined interpretation, which is regarded as working hypothesis. We short-ened our statement now and expressed this clearly.

Response to Comment P13703 L23 By the wide onset of the new particle formation, we meant that it took several hours. A further explanation was added to clarify this, and the word "probably" was removed.

Response to Comment P13703 L29 The sudden shift in the time evolution of the nucleation mode diameter occurs due to an artefact of the fitting program. When the growing nucleation mode gets close to the Aitken mode, they cannot be resolved into two peaks any more, and the shift happens. A brief clarification was added now to the text.

Response to Comment P13704 L21 In total, 83 new particle formation events were identified (see Table 1). Of them, 42 were classified as Nucleation event class 1. This class represents processes without disruptions in the contour plot, and, therefore, they could be further evaluated. Of them, nucleation mode was so well developed that the time evolution of its modal parameters could be calculated with reasonable uncertainty for 31 days. The later subset is usually called Nucleation event class 1A. Table 1 was extended by several further details to avoid any misunderstanding.

Response to Comment P13704 L24 Days with undefined feature and missing data were left out from the classification as stated on P13705 L1–2. This is the ordinary action to distinguish days with evident nucleation and days with evident non-nucleation. The possible reasons for the undefined days and the estimated influence of this action

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on the conclusions were discussed on P13704 L26– P13705 L3, and on P13697, L15– 29.

Response to Comment P13704 L29 The sentence was reformulated to clarify that the air masses near the measurement site seem to be spatially inhomogeneous, and therefore, the shape of the growth curve do not depend very much on the wind direction. This could be associated with the wind channel above the river.

Response to Comment P13705 L14 Unfortunately, we only have access to the seasonal variation of solar radiation for our site, and, therefore, the issue raised is beyond the scope of our MS. This part of the MS was shortened.

Response to Comment P13705 L24 The suggestion was accepted without notes.

Response to Comment P13706 L24 The following sentence was added as explanation: "The larger summertime growth rates probably reflect the larger concentrations of condensing vapours due to enhanced photochemical activity, and also to increased biogenic organic precursor emissions." See also Response to Comment P13701 L6.

Response to Comment P13707 L21 Formulation "regional" event was changed to "meso-scale" event because this represents better the horizontal dimensions we dealt with in the present paper.

Response to Comment P13708 L10 Several sentences were reformulated to express our original intention that the urban-type sites show several drawbacks in comparison with background and rural sites.

Response to Comments P13708 L19 and on Fig. 3 The Conclusions was modified and clarified. Separation of modes in the size distributions by the fitting method was explained now more carefully, and it was discussed in section Data treatment. Number of peaks required to fit multiple peaks is mainly constrained by the width parameters of the individual peaks. Assumption that the modes can be fitted by log-normal distribution was justified by cumulative log-probability graphs of some selected individual ACPD 10, C9715–C9722, 2010

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distributions as well as monthly mean size distributions.

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