

Response to Referee #2

The authors thank Referee #2 for his/her valuable comments. We utilized all of them to further improve and clarify the MS, and made several extensions and alterations. Our responses to the comments are as follows.

Major comment

1. The actual meaning of the NSF is not clear, due to the normalization of nucleation + Aitken mode concentration with the accumulation mode (why not e.g. with the mean N_{6-100} from 6 to 9 am on the same morning?). The applied normalization may cause unintended signals: for example, if we consider two days during which the N_{6-100} is exactly similar, but on the latter (called here a nucleation day just to show the point) $N_{100-1000}$ is lower than on the first by a factor of 1.5, the NSF would be by 1.5. This would not only be a false signal but also to the wrong direction: during lower sink conditions the equal source should lead to higher concentrations, and if equal N_{6-100} was observed, the source should be weaker and thus NSF smaller than 1. This does not necessarily mean that the applied definition of NSF would not make sense, but its behaviour with the applied data set should be analysed and its meaning explained much more in detail.

We can agree with the Referee that the applicability and exact meaning of the nucleation strength factors (NSFs) is complex despite their relatively simple mathematical definition. We extended the corresponding part of the text with several new aspects and made the existing explanations more explicit and clear to avoid any misunderstanding. We further emphasized the assumptions for their utilization and their rigorous interpretation, and also named the two versions of the NSF differently (as NSF_{NUC} for the concentration increment on a nucleation day, and NSF_{GEN} for the concentration increase on a general day) to assist their differentiation. The changes are highlighted in red. The conclusion of the Referee based mainly on two specific examples in this paragraph, however, cannot be accepted. A) The normalisation of N_{6-100} cannot be performed e.g. to the mean N_{6-100} from 06:00 to 09:00 on the same morning (as suggested by the Referee) because the effect of NPF and particle growth process can continue till the next morning for some events, and therefore, it can contribute to an elevated N_{6-100} in mornings, which would disturb the correct quantification. This effect shows up as an elevated baseline in the morning in Fig. 1, and its consequences were further discussed in section 3.2. B) One of the basic assumptions for the NSFs is that the major emission and formation processes of the UF particles except for NPF are uniformly present on both nucleation and non-nucleation days

(lines 119–120 of the original MS). This can be fulfilled by taking into account concentration data for several days, and it can be misleading to deal with just two specific days, i.e. with one nucleation day and one non-nucleation day. More importantly, these days cannot be characterised by identical N_{6-100} at all because this would seriously contradict with the equality of all sources except for NPF (you simply cannot have identical N_{6-100} on a non-nucleation day and on a nucleation days if the other sources - except for NPF - are equal). The basic assumption of the NSF is evidently not met for this specific example, and the final conclusion drawn by the Referee is then inaccurate. As far as the minimum number of days (more exactly, the minimum number of NPF events during a time interval) sufficient for obtaining representative NSF values is concerned, it was discussed in section 3.2 that 2–3 weeks in winter (which is the most unfavourable interval from this point of view, see Table 1) might not be fully satisfactory for this purpose. At the same time, the longer time interval needed does not detract from the value of the quantification, because the health and environmental effects of NPF are important mostly on longer time scales.

Minor comments, on the terminology

2. The word nucleation is used in the manuscript for regional new particle formation events. It is misleading, since many of the anthropogenic particles also are formed in through nucleation processes, as the authors know. This should be revisited through the manuscript.

The word nucleation is used in the MS to express the regional- and urban-type NPF. The particles generated by these processes are formed in the ambient air from precursor gases, and are of secondary character. The particles which are formed inside a localised source or within a plume are emitted directly into the air from their emission sources, and are regarded as primary particles. We follow this pragmatic concept throughout our publications.

3. The word background seems to be applied with (at least) two different meanings, one for the background site and one for the background concentrations (e.g. lines 159-161, lines 226-227), which here, if I understood correct, refers to concentration of accumulation mode particles in general. Additionally, it seems that on lines 322-323 and 329 the term background means the concentration without nucleation event, otherwise “increment of background concentration on nucleation days” would mean higher $N_{100-1000}$ than on non-nucleation days. Why not simply use the term accumulation mode?

We can agree with the Referee, and modified the whole text at many places to distinguish between spatial (near-city) background and aerosol concentration background ($N_{100-1000}$). We worked with size intervals of 6–100 nm and 100–1000 nm, and mentioned it explicitly now

that the intervals estimate the nucleation + Aitken modes, and accumulation modes, respectively in most cases.

Minor comments, specific notices

4. Lines 41-43, should these sentences be one?

The two sentences were joined as requested.

5. Line 68-70: Open $(\text{NH}_4)_2\text{SO}_4$, NH_4NO_3 and TC

The meaning of the first two chemical formulae is unambiguous (ammonium sulfate and ammonium nitrate, respectively). We explained the abbreviation of the TC now as total carbon contained in particles (TC , $\text{TC}=\text{EC}+\text{OC}$).

6. Line 105: N6-100 referred to as Aitken mode, should be nucleation + Aitken mode, compare e.g. to lines 22-23.

The request was adopted.

7. The time over which the daily mean is calculated for seasonal or annual NSF's should be mentioned in the methodology part (perhaps line 130), now it appears only on lines 322-323.

The request was adopted.

8. Line 131: reference for the site in question, in some/many locations more nucleation days.

An overview paper by Nieminen et al., Global analysis of continental boundary layer new particle formation based on long-term measurements to be submitted very soon was added as a reference for this general property.

9. Lines 143-144: describe shortly the "second group"

Main features of the referred group of the monthly mean nucleation frequency distribution were described now explicitly as: The seasonal variation of the nucleation frequency fits into the second group of the measurement sites - which is characterised by the highest number of nucleation events in spring and the lowest in winter, with relatively high total number of events (Manninen et al., 2010).

10. Lines 159-160: What are the background concentrations meant here? In the context of the table it is logically connected to background site, but from the sentence it seems not to be so.

The whole text was modified at many places to distinguish between spatial (near-city) background and concentration background ($N_{100-1000}$).

11. Line 277: maybe “of what” instead of “which”, or modify the sentence otherwise.

The sentence was modified as requested.

12. Lines 384-387: It seems that the growth of these particles is considered as a loss of these particles in this analysis. The share of particles grown out of nucleation mode size range should be possible to determine from the dm_{ps} measurements with the normal methodologies (e.g. Kulmala et al., 2012, referred to in the manuscript).

The residence time of particles with diameters 6–25 nm determined in this way indeed includes the major sinks, namely the coagulation of particles and growth out of particles from the specified size range. Their relative contributions (mean coagulation rate and growth out rate with respect to the formation rate J_6) in Budapest are evaluated in an ongoing study, and they are to be reported and discussed in a separate MS.

13. Lines 404-405: I don’t believe the authors mean the particles of e.g. 20 nm diameter are thermodynamically unstable.

The statement was indeed meant for the aerosol system containing particles with diameters below 20 nm, and not for the particles themselves. The sentence was corrected accordingly.

Finally, we would like also to mention that we considered several options and size intervals for quantifying the NPF as a single source of particles, and found that the quantities presented in the MS are the most advantageous and expressive possibilities. We are aware that the treatment introduced has some limitations. These are discussed explicitly and in detail in the revised paper. Nevertheless, the proposed method is capable of quantifying the relevance of particles from NPF relative to other sources, e.g. to road traffic emissions in cities for the first time, which is an unambiguous and important step forward in urban atmospheric studies.

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27 November 2017