Response to Referee #1

The authors thank Referee #1 for his/her 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.

1. Two NSF factor have been used in the manuscript. The mixed usage of the two factors always confused me. I would like to suggest the authors to clearly state which factor they are referring to whenever possible, e.g., NSFs in Line 134-136. Also, is NSFnucl days larger than NSFall days? Looks to me that (N6-100/N100-1000)nul days is larger than (N6-100/N100-1000)all days by definition, isn't it? I would like to see more discussion on the relationship between the two factors.

We 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 all over the MS to assist their differentiation. The interpretation of their meaning regarding the limiting values is valid for both of them. We also extended the text with several new aspects on their relationships. The changes are highlighted in red.

2. In table 2, wouldn't I get (N6-100/N100-1000)nul days from the numbers listed? For example, (N6-100/N100-1000)nul days can be obtained (1.72/1.03 = ((N6-100)Nucl/(N6-100)Non-nucl)/((N100-1000)Nucl/(N100-1000)Non-nucl) = ((N6-100)Nucl/(N100-1000)Nucl)/((N6-100)Non-nucl/(N100-1000)Non-nucl) = (N6-100/N100-1000)nul days. However, my number is different from those listed in Table 3. What is the problem?

In data processing, concentration ratios were first derived from the individual data; later these ratios were averaged for nucleation and non-nucleation days for a specific time point considered, and finally the NSF values were calculated and averaged over the time interval selected. This does not necessarily and exactly results in the same final NSF value as that derived from the mean concentrations. The former data treatment is the correct method considering the dynamic character of atmospheric concentrations, uncertainties of the data, and the propagation of errors. This was actually utilised in the MS.

3. (Line 156-158), put "Data coverage for summer and autumn for the mean ratios" as a footnote of Table 2.

The requested footnote was added.

4. (Line 159-160) the lower background particle concentration on nucleation days in winter came from real measurements, right?

All the primary data presented were obtained experimentally, which was confirmed now in this section as well.

5. (Line 173) rephrase "its consideration in the averaging is justified".

The sentence was modified to clarify its meaning.

6. Clearly mark the vertical coordinates (NSFnucl days or NSFall days) in Figures 1 & 4, and also include NSFnucl days or NSFall days in the body instead of the title of Tables 3 & 4.

The requested changes were fully adopted. See also the response to Comment 1.

7. In table 2, what caused the behavior of N6-100 on NPF days? Especially, the peak at night. Also, please expand to discuss why N6-100 is significantly larger between 6-9 am on non-nucleation days? Does this mean that nucleation was hindered by the high concentrations of preexisting particles?

We assume that the Referee meant Figure 2 instead of Table 2. The late evening/night peak was observed from the beginning of the measurements in Budapest (see e.g. Salma et al., 2010), and can likely be related to the combined effect of burning and heating activities at residences and homes, and of local meteorology. They are also influenced by the daily cycling of the boundary layer mixing height and mixing intensity. The exact interpretation of the evening peak, however, needs further dedicated investigations. As far as the $N_{6–100}$ between 6 and 9 am is concerned, its higher level on non-nucleation days with respect to nucleation days is indeed related to higher pre-existing aerosol concentration level, and thus, larges condensation sink values, which hinder the chances for NPF. These explanations were now included into the text.

8. (Line 272) lower solar radiation in winter is understandable, but less biogenic pre-cursor gases are not justified. Is there any evidence that NPF in Budapest requires biogenic vapors?

There is indirect evidence that biogenic emissions contribute to the early stage of the growth process, and likely to the nucleation itself as well. Distribution of the monthly mean occurrence frequency, and relatively low (<20%) relative contribution of gas-phase H_2SO_4 to the growth rate of particles can indicate the role of biogenic precursors. The sentence was extended to include this information, and two specific references were given now for further details.

9. (Line 277-279) rephrase “this observation raises the question ... for calculating the NSF”.

The sentence was modified.

10. Regarding the health effects (Line 385-387), I would like to suggest the authors to be more conservative. The relative short lifetime is one aspect, but the toxicity per particle is another aspect. The authors just can’t evaluate the health effects of nanoparticles generated by NPF.

We can fully agree. The discussion on the health effects was modified to be more accurate, and the specific aspect raised by the Referee was included.

Imre Salma
27 November 2017

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 N6-100 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 N6-100 is exactly similar, but on the latter (called here a nucleation day just to show the point) N100-1000 is lower than on the first by a factor of 1.5, the NSF would 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 N6-100 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 it's 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 contradicts 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 N100-1000 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 (NH4)2SO4, NH4NO3 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, TC=EC+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 NSFs 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 dmps 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.

Imre Salma

27 November 2017

Quantification of atmospheric nucleation and growth process as a single source of aerosol particles in a city

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Abstract. Effects of new aerosol particle formation (NPF) and particle diameter growth process as a single source on atmospheric particle number concentrations were evaluated and quantified on the basis of experimental data sets obtained from particle number size distribution measurements in the city centre and near-city background of Budapest for 5 years. Nucleation strength factors separately for a nucleation day (NSF_{NUC}) and for a general day (NSF_{GEN}) were derived for seasons and full years. The former characteristics represents the concentration increment of ultrafine (UF) particles specifically on nucleation days with respect to accumulation-mode (regional background) ~~particle~~ concentration (particles with equivalent diameters of 100–1000 nm; $N_{100–1000}$) due solely to nucleation process. The latter factor expresses the contribution of nucleation to particle numbers on general days, thus it represents a longer time interval such as season or year. The nucleation source had the largest effect on the concentrations around noon and early afternoon as expected. During this time interval, it became the major source of particles in the near-city background. Nucleation increased the daily mean ~~particle number~~ concentrations on nucleation days by mean factors of 2.3 and 1.58 in the near-city background and city centre, respectively. Its effect was the largest in winter, which was explained with the substantially lower ~~background concentration~~ $N_{100–1000}$ levels on nucleation days than that on non-nucleation days. On an annual time scale, 37% of the UF particles were generated by nucleation in the near-city background, while NPF produced 13% of UF particles in the city centre. The differences among the annual mean values, and among the corresponding seasonal mean values were likely caused by the variability in controlling factors from year to year. The values obtained represent lower limits of the contributions. The shares determined imply that NPF is a non-negligible or substantial source of particles in near-city background environments and even in city centres, where the vehicular road emissions usually prevail. Atmospheric residence time of nucleation-mode particles was assessed by a decay curve analysis ~~of $N_{6–25}$ concentrations in time~~, and a mean of 2:30 was obtained. The present study suggests that the health-related consequences of atmospheric NPF and growth process in cities should also be considered in addition to its urban climate implications.

1 Introduction

Large-scale modelling studies suggest that new aerosol particle formation (NPF) and consecutive particle diameter growth process in the atmosphere (Kulmala et al., 2004, 2013) is the dominant source of particle number concentrations on global scale (Spracklen et al., 2006; Reddington et al., 2011; Makkonen et al., 2012; Yu et al., 2015). In addition, up to approximately 50% of all cloud condensation nuclei (CCN) can originate from NPF and growth (Spracklen et al., 2008; Merikanto et al., 2009), which relates the process to the climate system, and indicates its overall importance (Kerminen et al., 2012; Carslaw et al., 2013; Shen et al., 2017). New particle formation has also been proved to be common in large cities (Nieminen et al., 2017). Urban NPF can interact with and can be influenced by regional nucleation events at least under some geographic conditions, and can become part of a phenomenon with a much larger horizontal extension than the city (Salma et al., 2016b). At the same time, particle number concentrations in cities are strongly affected by high-temperature emission sources from different sectors (Paasonen et al. 2016) such as household and residential heating (e.g. Butt et al., 2016), industrial processes and power production (e.g. Xiao et al., 2015), and vehicular road traffic (e.g. Morawska et al., 2008). Their diurnal variation often show daily time-activity pattern of inhabitants (Dall'Osto et al., 2013). Relative contributions of primary and secondary particle sources – particularly in cities – change substantially in time and space (Pikridas et al., 2015; Posner and Pandis, 2015). Several

methods were proposed to distinguish the major production types of particles (e.g. Shi et al., 1999; Alam et al., 2003; Rodrígues and Cuevas, 2007; Qian et al., 2007; Park et al., 2008; Costabile et al., 2009; Brines et al., 2015). The share of NPF as a single source of ambient particle number concentrations specifically in cities remained, however, largely unknown despite the fact that there is often a spatial coincidence between the poorer air quality and population density (Samoli et al., 2016). Moreover, approximately 70–80% of total particles in cities belong to the ultrafine (UF) size range (with an equivalent diameter <100 nm; Putaud et al., 2010), and their inhalation can represent an excess health risk relative to coarse or fine particles with the same or similar chemical composition (Oberdörster et al., 2005; Braakhuis et al., 2014). An estimate on the relative contribution of primary and secondary formation processes is also required for efficient action plans to improve the air quality in cities. It is worth noting that indirect climate effects (due to CCN) become important for particles with diameters >50–100 nm, while the excess health effects are linked with diameters <100 nm.

Nucleation strength factor (NSF) was introduced to assess the contribution of NPF to UF particle number concentrations relative to accumulation-mode (regional background) concentration (particles with diameters of 100–1000 nm; $N_{100-1000}$) with respect to all other sources (Salma et al., 2014). The results derived from this approach correspond to the mode-segregated secondary particle load. By now, atmospheric concentration data sets are available for multiple years to study the applicability, ~~and~~ behaviour and interpretation of the NSF in detail. The major advantage of this quantification approach is that it only requires experimental data that can be readily derived from ordinary NPF (size distributions) measurements. The main objectives of this paper are to quantify and discuss the contribution of NPF events to ambient particle number concentrations in near-city and central urban environments of a Central European city considering five-year long data sets, to investigate and explain the meaning and further details of the NSF, and to interpret the consequences achieved for the urban air and air quality.

2 Methods

2.1 Experimental

The measurements were performed in Budapest, Hungary. Its population is approximately 2.5 million in the metropolitan area. The major pollution sources in terms of particle number include vehicular road traffic, residential heating and household burning activities. Contributions of passenger cars and buses to the vehicle fleet registered in Budapest and Pest County are 87% and 0.46%, respectively (OKJ, 2015). Diesel-powered vehicles shared 19% and 97% of the national passenger car and bus fleets, respectively. Wintertime median concentrations of particulate matter (PM) mass, elemental carbon (EC) and organic carbon (OC) in the $PM_{2.5}$ size fraction were 25, 0.97 and 4.9 μg m^{-3}, respectively in the related time interval (Salma et al., 2017). The mean contributions of EC and organic matter (OM, with an OM/OC mass conversion factor of 1.6) to the $PM_{2.5}$ mass and standard deviation (SD) were 4.8±2.1% and 37±10%, respectively, while the contribution of $(NH_4)_2SO_4$ and NH_4NO_3 derived from an earlier study in spring were 24% and 3%, respectively. The contributions of EC and OC from fossil fuel combustion to the total carbon contained in particles (TC=OC+EC) were 11.0% and 25%, respectively, and EC and OC from biomass burning were responsible for 5.8% and 34%, respectively of the TC, while the OC from biogenic sources made up 24% of the TC.

Two urban sites were involved in the study. Most measurements were performed at the Budapest platform for Aerosol Research and Training (BpART) facility (N 47° 28' 29.9", E 19° 3' 44.6", 115 m above mean see level (a.s.l.) of the Eötvös University (Salma et al., 2016a). The sampling inlets were set up at heights between 12 and 13 m above the street level. The location represents a well-mixed, average atmospheric environment for the city centre. The other location was situated at the NW border of Budapest in a wooded area of the Konkoly Astronomical Observatory of the Hungarian Academy of Sciences (N 47° 30' 00.0", E 18° 57' 46.8", 478 m a.s.l.). It represents the air masses entering the city since the prevailing wind direction in the area

is NW. The experimental data obtained for five full-year long time intervals, i.e. from 03–11–2008 to 02–11–2009, from 19–01–2012 to 18–01–2013, from 13–11–2013 to 12–11–2014, from 13–11–2014 to 12–11–2015 and from 13–11–2015 to 12–11–2016 were considered in the present study. Local time (UTC+1 and daylight saving time, UTC+2) was chosen as the time scale because the daily routine activities of the inhabitants in the city were primarily considered.

The key measuring instrument was a flow-switching type differential mobility particle sizer (DMPS; Salma et al., 2011). Its main components include a Ni-60 radioactive bipolar charger, a Nafion semi-permeable membrane dryer, a 28-cm long Vienna-type differential mobility analyser and a butanol-based condensation particle counter (TSI, model 3775). The system operates in an electrical mobility diameter range from 6 to 1000 nm in the dry state of particles (with a relative humidity RH<30%) in 30 channels with a time resolution of approximately 8 or 10 min at two sets of flows. The sample flow rate is 2.0 L min^{-1} in high-flow mode, and 0.31 L min^{-1} in low-flow mode with sheath air flow rates 10 times larger than for the sample flows. The DMPS measurements were performed according to the international technical standard (Wiedensohler et al., 2012). The DMPS data for the 1-year long time intervals in 2008–2009, 2012–2013, 2013–2014, 2014–2015 and 2015–2016 were available in 95%, 95%, 99%, 95% and 73% of the total number of days, respectively. Meteorological data were recorded by an on-site meteorological station (Salma et al., 2016a). Standardised meteorological measurements of air temperature (*T*), RH, wind speed and wind direction were recorded with a time resolution of 10 min. The coverage of the meteorological data was >80% in each year.

2.2 Data treatment

The overall treatment of the measured DMPS data was performed according to the procedure protocol by Kulmala et al. (2012). The inverted DMPS data were utilised to generate particle number size distribution surface plots showing jointly the variation in particle diameter and particle number concentration density in time. Identification and classification of NPF and growth events was accomplished from the surface plots by using the algorithm similar to that of Dal Maso et al. (2005) on a day-to-day basis into the following main classes: NPF event days, non-event days, days with undefined character, and days with missing data (for more than 4 h in the midday). Frequency of events was determined as the ratio of the number of event days to the total number of relevant (i.e. all–missing) days. Particle number concentrations in the diameter ranges from 6 to 1000 nm ($N_{6–1000}$), from 6 to 100 nm ($N_{6–100}$), from 6 to 25 nm ($N_{6–25}$) and from 100 to 1000 nm ($N_{100–1000}$) were calculated from the DMPS data. The major portion of the $N_{6–100}$ concentration (i.e. in the Aitken mode plus sporadically the nucleation mode) is essentially related to local source processes due to the limited atmospheric residence time (typically <10^1 h) of these particles, while the $N_{100–1000}$ (regional background concentration associated mainly with the accumulation mode) expresses larger ~~(background)~~ spatial and time scales because of much longer residence times (up to 10^1 d; Salma et al., 2011). To obtain mean diurnal variation of the concentrations and further properties derived from them (see later), the exact recording times belonging to the individual concentrations were rounded off to 5 min (in case of the time resolution of ca. 8 min) or 10 min (in case of the time resolution of ca. 10 min) time scale. These data were averaged by the time of day separately for nucleation and non-nucleation days. Finally, the averaging was also performed separately for different seasons, hence for spring (March-May), summer (June-August), autumn (September-November) and winter (December-February), and for the measurement year.

Two types of NSF (Salma et al., 2014) were derived in the present study by considering different conditions. The quantity:

$$\mathrm{NSF_{NUC}} = \frac{\left(N_{6–100}/N_{100–1000}\right)_{\mathrm{nucleationdays}}}{\left(N_{6–100}/N_{100–1000}\right)_{\mathrm{non\text{-}nucleationdays}}} \quad (1)$$

considers the $N_{6–100}/N_{100–1000}$ concentration ratios for nucleation days only. The numerator expresses the increase in $N_{6–100}$ relative to the ~~background concentration~~ $N_{100–1000}$ caused by all source sources. The denominator represents the same property due to all sources except for NPF. Hence, the NSF_{NUC} accounts for the concentration increment ~~in background particle concentration~~ on a nucleation day exclusively caused by NPF. It was implicitly assumed that the major emission and formation processes of UF particles except for NPF are uniformly present on both nucleation and non-nucleation days. It seems to be a reasonable condition for time intervals of several months, although the number of nucleation days during a time interval actually plays a more determining role than the length of the time interval. Winter, when the occurrence frequency shows the minimum (see Table 1), appears to be the most restrictive or critical season. The effect of the non-uniformly present sources is indicated by unusually larger scatter in the diurnal data points (see Sect. 3.2). It was also presumed that the production of particles larger than 100 nm was much smaller than the concentration of UF particles. This is ordinarily realised in cities, and can be justified from the contributions of UF particles to the total particle number (Putaud et al., 2010; Németh et al., 2017).

The other type of NSF was calculated for all days in the numerator, thus:

$$NSF_{GEN} = \frac{\left(N_{6–100}/N_{100–1000}\right)_{\text{all days}}}{\left(N_{6–100}/N_{100–1000}\right)_{\text{non-nucleation days}}} \quad (2)$$

It expresses the overall contribution of NPF to ~~particle numbers of~~ $N_{100–1000}$ ~~background concentration~~on longer time span, so in general. Since there are usually more non-nucleation days than nucleation days in a time interval of month or more (Nieminen et al., 2017), the assumptions for NSF_{GEN} are met easier than for NSF_{NUC}. The NSF_{NUC} characterises an ordinary nucleation day within e.g. a season, while the NSF_{GEN} quantifies the overall effect of NPF and growth on the atmospheric concentrations on a regular/average day over e.g. a season or year. The former is always larger than the latter due to their definition. They can be calculated for seasons or years. Despite their relatively simple mathematical definition, the exact meaning and rigorous interpretation of the NSF_{NUC} and NSF_{GEN} are complex, and they should be approach with care in particular, as far as the assumptions for their utilisation are concerned. If 1) NSF≈1 then the relative contribution of nucleation to particle number concentrations with respect to other sources is negligible, 2) 1<NSF<2 then its relative contribution as a single source is considerable, and 3) NSF>2 then the contribution of nucleation itself to particle number concentrations is larger than of any other source sectors together. The interpretation regarding the limiting values is valid for both types of NSF. Since the major phase of NPF and growth process takes place in most cases in one day, it is advantageous to express NSFs as daily mean values. The data for the undefined days were not taken into account for the present evaluation.

3 Results and discussion

Number of nucleation days for different seasons in each measurement year are summarised in Table 1. It is seen that the NPF frequency has an obvious seasonal variation. This can be obtained from its monthly dependency which exhibits an absolute and local minimum in January and August, respectively, and an absolute and local maximum in March or April, and September, respectively (Salma et al., 2016b). 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 ~~reported by~~ (Manninen et al., 2010).

Table 1. Number of nucleation days for seasons in the near-city background (in 2012–2013) and in the city centre (in 2008–2009, 2013–2014, 2014–2015 and 2015–2016) during 1-year long time intervals.

Environment	Time interval	Spring	Summer	Autumn	Winter
Background	2012–2013	35	20	24	17
Centre	2008–2009	34	21	22	6
Centre	2013–2014	28	20	13	11
Centre	2014–2015	41	19	14	7
Centre	2015–2016	15	9*	5*	6

* Low data coverage. See Sect. 2.1.

3.1 Seasonal atmospheric concentrations

Particle number concentrations in the related size fractions for different seasons in each measurement year are summarised in Table 2 for an overview.

Table 2. Median atmospheric concentration of particles with a diameter from 6 to 100 nm ($N_{6–100}$) and from 100 to 1000 nm ($N_{100–1000}$) in units of 10^3 cm^{-3} separately on nucleation (Nuc) days and non-nucleation (Nonuc) days for seasons in the near-city background (in 2012–2013) and in the city centre (in 2008–2009, 2013–2014, 2014–2015 and 2015–2016) during 1-year long time intervals. Mean ratios of median concentrations on nucleation days to that on non-nucleation days with standard deviation (SD) for the size fraction are also indicated.

Urban environment			Near-city backgr.		City centre				
Time interval	Size fraction	Day type	2012–2013	Ratio	2008–2009	2013–2014	2014–2015	2015–2016	Ratio ±SD
Spring	$N_{6–100}$	Nuc	4.8	1.72	11.2	9.7	10.0	8.6	1.37
	$N_{6–100}$	Nonuc	2.8		8.9	7.2	7.1	5.9	±0.09
	$N_{100–1000}$	Nuc	1.56	1.03	2.0	2.5	2.6	1.56	0.99
	$N_{100–1000}$	Nonuc	1.51		2.2	2.7	2.5	1.5	±0.07
Summer	$N_{6–100}$	Nuc	4.0	1.37	10.3	8.0	8.6	6.9*	1.17
	$N_{6–100}$	Nonuc	2.9		8.9	7.5	6.5	6.2*	±0.11
	$N_{100–1000}$	Nuc	1.27	0.89	1.36	2.0	2.5	1.40*	0.92
	$N_{100–1000}$	Nonuc	1.42		1.72	2.4	2.4	1.37*	±0.13
Autumn	$N_{6–100}$	Nuc	4.3	1.29	14.0	11.9	12.6	5.2*	1.41
	$N_{6–100}$	Nonuc	3.3		10.4	8.5	8.4	5.1*	±0.08
	$N_{100–1000}$	Nuc	1.67	0.74	2.0	3.4	2.7	1.6*	0.87
	$N_{100–1000}$	Nonuc	2.3		2.4	3.9	3.3	1.7*	±0.06
Winter	$N_{6–100}$	Nuc	3.9	1.10	6.9	10.5	5.6	7.7	0.87
	$N_{6–100}$	Nonuc	3.6		12.5	9.2	7.8	7.4	±0.28
	$N_{100–1000}$	Nuc	1.12	0.38	1.02	3.7	1.65	1.4	0.54
	$N_{100–1000}$	Nonuc	2.9		3.0	4.5	3.8	2.7	±0.22

* Low data coverage. See Sect. 2.1.

Data coverage for summer and autumn in 2015–2016 were low, and therefore, the corresponding concentration ratios were excluded from the averaging for the mean ratios. It can be seen that the $N_{6–100}$ were ordinarily larger on nucleation days than on non-nucleation days. This is most likely a direct effect of nucleation. At the same time, the $N_{100–1000}$ usually showed a constant level within approximately 10% except for winters and some autumns. The $N_{100–1000}$ ~~background concentrations~~ on nucleation days were, however, smaller (by factors of 0.4–0.5) than for non-nucleation days, particularly in winter. The differences are further discussed and explained in Sect. 3.2.

3.2 Concentration increment on nucleation days

Diurnal variation of the concentration increment due to NPF on nucleation days (i.e. of NSF_{NUC}) for the city centre and near-city background separately for different seasons are shown in Fig. 1 as representative examples. The curves exhibited a single

peak around noon with a longer tail on the decreasing side. The exact location of the peak is also influenced by setting the local daylight saving time in spring and autumn. The baseline of some peaks from 0:00 to 7:00 deviated systematically and substantially from unity although no nocturnal nucleation has been observed in Budapest. The mean values of this baseline in the city centre for spring, summer, autumn and winter (Fig. 1 upper panel) were 1.02, 1.15, 1.15 and 1.55, respectively, while they were 1.11, 1.18, 1.31 and 1.72, respectively in the near-city background (Fig. 1 lower panel). The elevated line can be explained by the fact that particle growth process could be traced till the late morning of the next day in several occasions, thus the NPF influenced the N_{6-100} concentrations over the next morning. This affected the baseline if a non-nucleation day followed a nucleation day, and particularly, in the seasons when NPF events occur well separated from each other in time, which is typical for winter. The elevated baseline is a real effect of the NPF, and it should be definitely included in deriving the mean NSF_{NUC}~~s consideration in the averaging is justified~~.

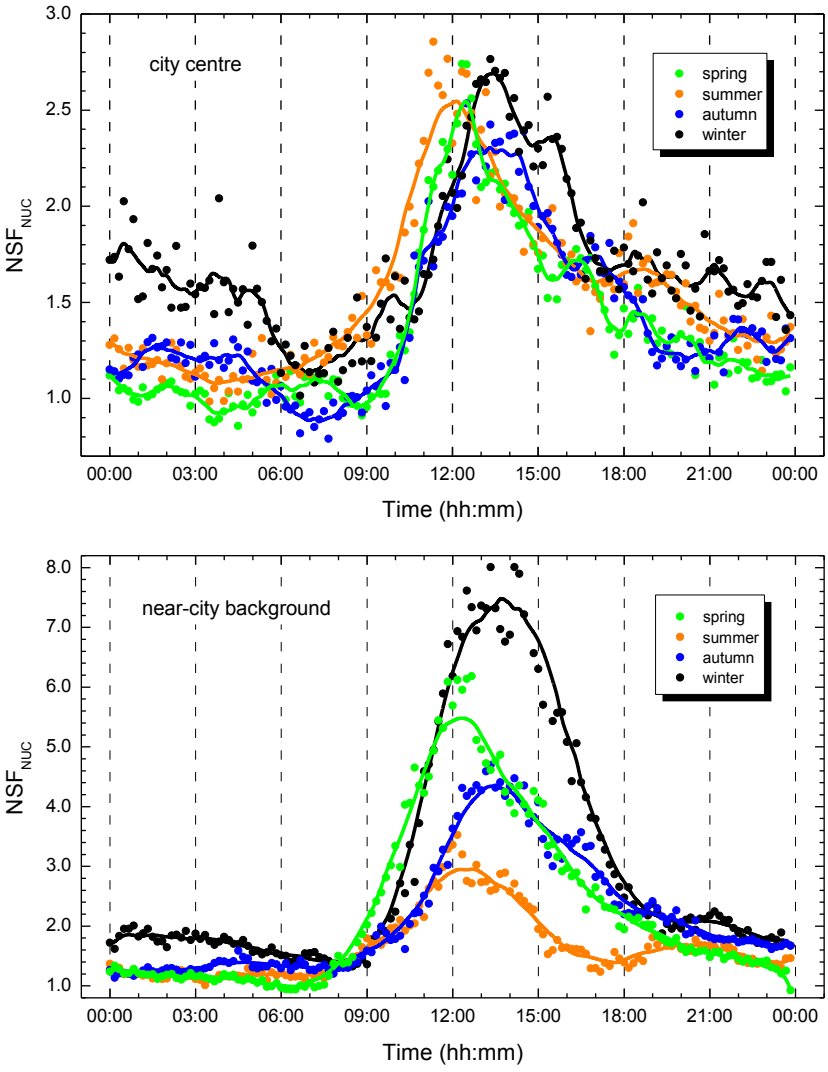

Figure 1. Diurnal variation of concentration increment on a nucleation day (NSF_{NUC}) due to new particle formation and growth for the city centre in 2008–2009 (upper panel) and in the near-city background in 2012–2013 (lower panel) separately for seasons. The solid lines represent 1-h smoothing to the data.

It was also observed in all years that the concentration increment on nucleation days due to NPF (i.e. NSF_{GEN}) was the largest for winter. This evidently showed up for the near-city background. It was followed by the other seasons which had similar importance to each other in the city centre, or which were ordered as spring, autumn and summer in the near-city background. To investigate these findings more closely, diurnal variation of the related particle number concentrations were derived and evaluated. Diurnal variation of the concentrations for summer and winter are shown in Figs. 2 and 3, respectively for the city

centre and near-city background. The dependencies in the city centre for the spring, autumn and other summer seasons are similar to Fig. 2 upper panel, while the corresponding seasonal curves for the near-city background resemble Fig. 2 lower panel. The diurnal patterns represented by Fig. 2 are coherent with the previous ideas on the NPF and growth events in the Budapest area (Salma et al., 2014, 2016b; Németh et al., 2017). They also confirm the basic assumptions of the NSF_{NUC} definition on the source intensities at both location types. The comparison of the $N_{6–100}$ curves for nucleation days and non-nucleation days already emphasizes the importance of NPF, and indicates that the phenomenon has larger relative effect in the near-city background than in the central urban parts - as it is expected. The late evening peak can be likely related to the combined effect of burning activities at residences and homes, of local meteorology, and they are also influenced by the daily cycling of the boundary layer mixing height and mixing intensity. As far as the $N_{6–100}$ between 06:00 and 09:00 is concerned, its higher level on non-nucleation days with respect to nucleation days is related to higher pre-existing aerosol concentration level, and thus, to larges condensation sink values, which hinder NPF. The ~~particle number concentrations for the background aerosol (~~$N_{100–1000}$~~)~~ curves appear close to each other within a relative uncertainty of 10–20%, which implies that the accumulation-mode~~background~~ concentrations affect the NPF occurrence and formation rate in a limited manner in these seasons.

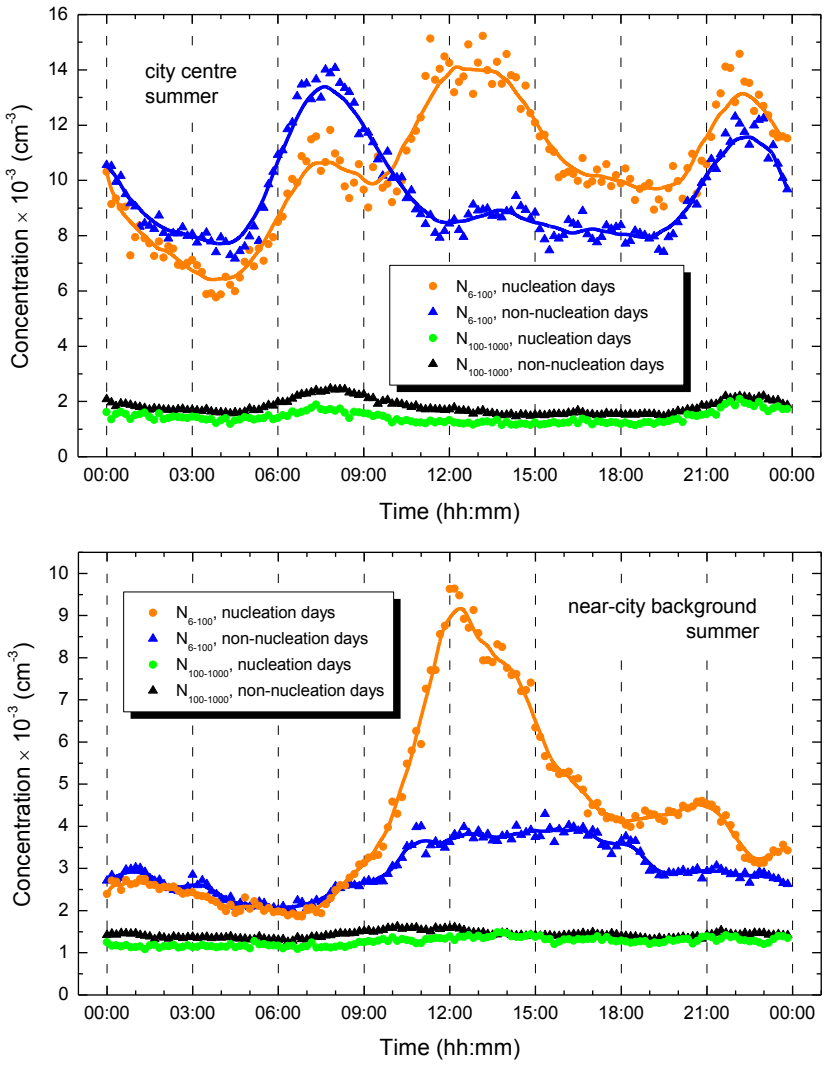

Figure 2. Diurnal variation of particle number concentrations in summer for the diameter ranges from 6 to 100 nm ($N_{6–100}$) and from 100 to 1000 nm ($N_{100–1000}$) in the city centre in 2008–2009 (upper panel) and in the near-city background in 2012–2013 (lower panel) separately for nucleation days and non-nucleation days. The solid lines represent 1-h smoothing to the data.

For winter, it is seen, however, that the $N_{100–1000}$~~background concentrations~~ were substantially different for the nucleation and non-nucleation days (Fig. 3). The mean non-nucleation/nucleation $N_{100–1000}$ ratios for the city centre (in 2008–2009) and near-

city background were 2.8 and 2.3, respectively. This implies that the NPF events preferably took place on those days when the particle number concentrations were generally smaller. It is understandable if we consider that the basic preconditions of NPF events are realised by competing source and sink for condensing vapours. The source strength in winter has a decreased tendency due to lower solar radiation intensities and less (biogenic) precursor gases in the air. There is indirect evidence that biogenic emissions contribute to the early stage of the growth process, and likely, to the nucleation itself as well (Salma et al., 2016b). Nevertheless, nucleation can occur at these small source terms if the (condensation and scavenging) sink - which is related to the concentration of pre-existing particles - is even smaller. This explains the differences in the $N_{100–1000}$ ~~background concentrations~~ on nucleation and non-nucleation days. Larger concentration increments (higher NSF_{NUC}, Fig. 3) for winter were simply caused by systematically smaller $N_{100–1000}$ ~~background concentrations~~ on nucleation days. In addition, the fluctuation in this concentration ~~of $N_{100–1000}$~~ for nucleation days in winter of the other years was sometimes larger than that shown in Fig. 3. This observation raises the question of ~~which is~~ the smallest number of NPF events in a time interval (e.g. season) that is ~~can be considered~~ sufficient for obtaining representative ~~mean diurnal concentration data for calculating the~~ NSF_{NUC}. A few NPF events during winters (see Table 1) may not be fully satisfactory for this purpose. The longer time intervals needed do not detract from the value of the quantification, because the health and environmental effects of NPF are important mostly on longer time scales.

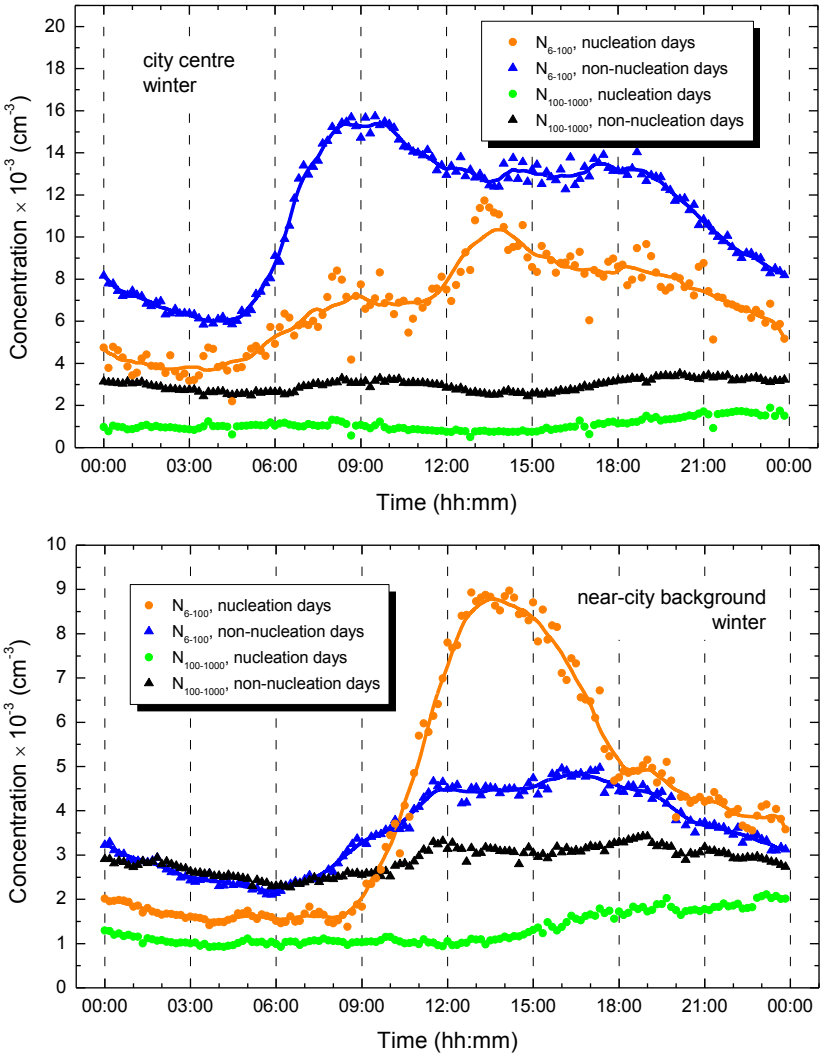

Figure 3. Diurnal variation of particle number concentrations in winter for the diameter ranges from 6 to 100 nm ($N_{6–100}$) and from 100 to 1000 nm ($N_{100–1000}$) in the city centre in 2008–2009 (upper panel) and in the near-city background in 2012–2013 (lower panel) separately for nucleation days and non-nucleation days. The solid lines represent 1-h smoothing to the data.

Seasonal and annual mean concentration increments on a nucleation day (hence the daily mean NSF_{NUC} values) for different years are summarised in Table 3. Nucleation as a single source increased the daily particle number concentrations by factors of 2.3 and 1.58 in the near-city background and city centre, respectively on an annual time scale. The differences among the annual mean values, and among the corresponding seasonal mean values were likely caused by the variability of controlling parameters from a year to year. As far as the seasonal variation is concerned, it is noted that the formation rate for particles with a diameter of 6 nm (J_6) showed only limited seasonal dependency in Budapest (Salma et al., 2011), which also contributed to similar mean increments for spring, summer and autumn.

Table 3. Seasonal and annual mean increments of ~~background concentration~~ $N_{100–1000}$ due to nucleation on a nucleation day (nucleation strength factor NSF_{NUC}) in the near-city background (in 2012–2013) and in the city centre (in 2008–2009, 2013–2014, 2014–2015 and 2015–2016) during 1-year long time intervals.

Urban environment	Time interval	NSF_{NUC}				
		Spring	Summer	Autumn	Winter	Year
Background	2012–2013	2.3	1.66	2.2	3.0	2.3
Centre	2008–2009	1.36	1.55	1.42	1.71	1.49
Centre	2013–2014	1.31	1.33	1.36	1.56	1.44
Centre	2014–2015	1.50	1.34	1.83	2.8	1.73
Centre	2015–2016	1.54	1.26	1.46	2.4	1.64

3.3 Contribution of nucleation to particle number concentrations

Diurnal variation of NSF_{GEN} is shown in Fig. 4. The curves exhibited a single peak with a maximum around noon and a longer tail in the early afternoon. The maximum values in the city centre represented concentration contributions from 30% to 60% due to nucleation for a limited time interval. The curve for the near-city background was the largest, as expected, and it even exceeded the value of 2 around noon for approximately 3 h. This all means that nucleation has an important contribution to UF particles during the midday in the city centre, while it even becomes the dominant source of particles directly after midday in the near-city background.

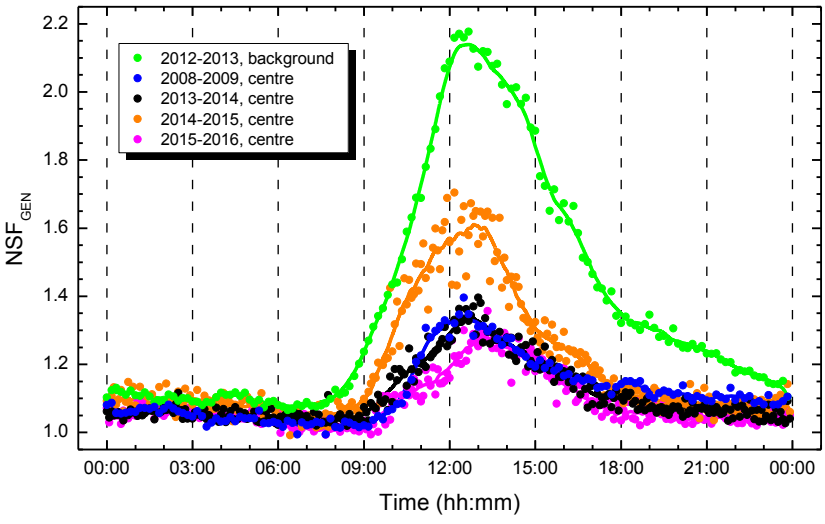

Figure 4. Diurnal variation of NPF contribution to particle number concentrations (NSF_{GEN}) in the near-city background (in 2012–2013) and in the city centre (in 2008–2009, 2013–2014, 2014–2015 and 2015–2016) during 1-year long time intervals. The solid lines represent 1-h smoothing to the data.

The importance of nucleation was also demonstrated for different seasons and years by the mean NSF_{GEN} values which are summarised in Table 4. In general, 37% of UF particles (more precisely of particle number increase) were produced by

nucleation as a single source in the near-city background. In the city centre, it generated 13% of UF particles. These values can be considered as lower limits since a considerable part of $N_{100–1000}$ ~~background~~ particles can be also produced by NPF from previous days. Nevertheless, the proposed method is capable of quantifies the relevance of particles from NPF relative to other sources for the first time, which is an unambiguous and important step forward in urban atmospheric studies. The differences among the annual mean values, and among the corresponding seasonal mean values were likely caused by the year-to-year variability similarly to the concentration increments on nucleation days (NSF_{NUC}). It is informative to compare the contribution values to the global share of various source sectors in primary UF particle number emission to have an idea on the relative extent of our results. It is stressed that our and the literature data are related to very different types of particles, nevertheless, some analogy can be found in the relative importance of the sources and source processes. Road transport, power production and residential combustion are the first three largest contributors to primary UF particles with the shares of 40%, 20% and 17%, respectively (Paasonen et al., 2016). The actual contributions can vary in different parts of the world and with economic development.

The effects of particles generated by NPF and growth in increased concentrations on human health and the environment are also influenced by the time interval for which the particles remain in the air. Nucleation-mode particles are primarily removed by coagulation with larger particles, agglomeration, diffusion and turbulent losses, scavenging and various aging processes. We showed in Sect. 3.2 and it can be also proved directly from the measured data that it is the NPF events that usually produce the $N_{6–25}$ concentrations in an overwhelming extent (the $N_{6–25}$ is increased by 1–2 orders of magnitude) in a relatively short time interval. The continual concentration decrease in time for several hours after the event can be utilised to assess the general atmospheric residence time of nucleation-mode particles. The decrease of this concentration after a nucleation burst could be approximated by an exponential function (first order kinetics). By a decay curve analysis of 15 selected NPF and particle diameter growth cases, the residence times were estimated from the slope of the concentrations $N_{6–25}$ in a natural logarithmic scale versus time. The residence time of nucleated particles varied from 1:30 to 4:15 with a mean and SD of 2:30±1:00. This suggests that the nucleation-mode particles (or in other words the nucleated particles in their very small sizes, or atmospheric nanoparticles) likely have limited health effects due to their relatively short existence in the air.

Table 4. Seasonal and annual mean contributions of nucleation to $N_{100–1000}$ ~~background concentration~~ (nucleation strength factor NSF_{GEN}) in the near-city background (in 2012–2013) and in the city centre (in 2008–2009, 2013–2014, 2014–2015 and 2015–2016) during 1-year long time intervals.

Urban environment	Time interval	NSF_{GEN}				
		Spring	Summer	Autumn	Winter	Year
Background	2012–2013	1.51	1.18	1.31	1.40	1.37
Centre	2008–2009	1.12	1.15	1.10	1.05	1.12
Centre	2013–2014	1.11	1.07	1.08	1.11	1.11
Centre	2014–2015	1.22	1.08	1.14	1.16	1.19
Centre	2015–2016	1.09	1.06	1.03	1.09	1.09

4 Conclusions

We showed in the present study that NPF and particle diameter growth process as a single source represents a considerable contribution to UF particles in a Central European city with respect to all other emission sources including vehicular road traffic. Nucleation was a major process that produced UF particles at noon and in the early afternoon, and its relative contribution was comparable to other production sources during this time period even in the city centre. Relative importance of nucleation as a source of particles decreased with anthropogenic influence. The NSFs were defined by utilising $N_{6–100}$ and $N_{100–1000}$ concentrations. There are several sensible and practical reasons for selecting these specific size fractions although

423 other dividing values are also imaginable. The quantifications in the present study are, therefore, subjected to certain inherent
424 uncertainty. Considering that the modes of the particle number size distribution are usually shifted to smaller diameters in
425 cities with respect to rural or remote areas, it seems realistic that these size fractions represent well the particles of urban (local)
426 origin and the aged particles (which characterize larger spatial or urban background area), respectively. The study also suggests
427 that particles from NPF events in cities are relevant not only for their effects on urban climate but because of their health risk
428 to inhabitants. At the same time, it should also be mentioned that ambient atmospheric aerosol which ordinary persists in the
429 air of cities contains particles in the largest abundance with a diameter between approximately 25 and 150 nm. Atmospheric
430 aerosol system containing smaller particles is thermodynamically not stable, and most of these particles are removed from the
431 air during relatively short time intervals. The exposures to freshly nucleated particles (d<25 nm) or ambient nanoparticles
432 (d<10 nm) are usually limited to several hours after the onset of the NPF.
433
434 Regulations of aerosol emissions and atmospheric concentrations are usually based on PM mass. The changes or reductions in
435 anthropogenic aerosol load are ordinary assessed by assuming similar relative tendencies in particle mass and particle number
436 concentrations. This generalisation may yield to tentative conclusions. According to current legislation scenarios, particle
437 number emissions are expected to decrease in most part of the world by 2030 mainly due to spreading the diesel particulate
438 filters (DPF) in cars and due to diesel fuels with ultralow sulphur content. In effect, this may imply that the relative share of
439 NPF in the particle number production is expected to increase above the levels estimated in the present study. By demonstrating
440 the relevance of NPF as an important single source of UF particles, we also raise the question of an international enhanced
441 particle mass and particle number inventory with precursor gas data that potentially includes the NPF and growth process as a
442 separate sector among the source types.

443 **5 Data availability**

444 The relevant observational data used in this paper are available on request from the corresponding author or at the website of
445 the Budapest platform for Aerosol Research and Training (http://salma.web.elte.hu/BpArt).

446 *Competing interests.* The authors declare that they have no conflict of interest.

447 *Acknowledgement.* Financial supports by the National Research, Development and Innovation Office of Hungary (contract
448 K116788) is appreciated.

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