Atmospheric Environment 178 (2018) 191–197

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Comparison of atmospheric new particle formation events in three Central European cities

Zoltán Németh[a], Bernadette Rosati[b,e], Naděžda Zíková[c], Imre Salma[a], László Bozó[d], Carmen Dameto de España[b], Jaroslav Schwarz[c], Vladimír Ždímal[c], Anna Wonaschütz[b,*]

[a] Institute of Chemistry, Eötvös University, Budapest, Hungary
[b] Faculty of Physics, University of Vienna, Vienna, Austria
[c] Laboratory of Aerosol Chemistry and Physics, Institute of Chemical Process Fundamentals, Prague, Czech Republic
[d] Hungarian Meteorological Service, Budapest, Hungary
[e] Department of Chemistry, Aarhus University, Aarhus, Denmark

ARTICLE INFO

Keywords:
Urban environment
Ultrafine particles
New particle formation
Particle number concentration
Particle number size distribution
Nucleation strength factor

ABSTRACT

Simultaneous particle number size distribution measurements were performed in the urban environment of Budapest, Vienna, and Prague, three Central European cities located within 450 km of each other. The measurement days from the continuous, 2-year long campaign were classified for new particle formation (NPF) events using an adapted classification scheme for urban sites. The total numbers of NPF event days were 152 for Budapest, 69 for Vienna, and 143 for Prague. There were 12 days when new particle formation took place at all three sites; 11 out of these 12 days were in spring and in summer. There were only 2 (Budapest-Vienna), 19 (Budapest-Prague), and 19 (Vienna-Prague) nucleation days, when NPF did not occur on the third site. The main difference was related to source and sink terms of gas-phase sulphuric acid. Air mass origin and back-trajectories did not show any substantial influence on the atmospheric nucleation phenomena. The relative contribution of particles from NPF with respect to regional aerosol to the particles originating from all sources was expressed as nucleation strength factor. The overall mean nucleation strength factors were 1.58, 1.54, and 2.01 for Budapest, Vienna, and Prague, respectively, and showed diurnal and seasonal variations. The monthly mean NSF varied from 1.2 to 3.2 in Budapest, from 0.7 to 1.9 in Vienna, and from 1.0 to 2.3 in Prague. This implies that the new particle formation in cities is a significant source of ultrafine (UF) particles, and the amount of them is comparable to the directly emitted UF particles.

1. Introduction and objectives

Ultrafine particles (UF, $d < 100$ nm) have the largest contribution to the total particle number concentrations on a global scale (Kulmala et al., 2004; Spracklen et al., 2006). Their major sources include direct emissions (vehicular exhaust, heating, burning) and atmospheric nucleation (Wåhlin et al., 2001; Kulmala et al., 2013). The UF particles enter into the human body by breathing, and approx. 60% of them can be deposited in the respiratory system (Geiser et al., 2005; Salma et al., 2015). They can cause adverse health effects mainly by inflammation or oxidative stress (Oberdörster et al., 2005; HEI Review Panel, 2013). Concentration of UF particles was recently linked to hospitalization in cities (Samoli et al., 2016).

New particle formation (NPF) events have been identified at various locations from clean, remote sites to polluted environments (Dal Maso et al., 2002; Holmes, 2007; Manninen et al., 2010; Brines et al., 2015; Dameto de España et al., 2017). Urban atmospheric nucleation has been increasingly investigated due to the superposition of freshly formed particles on the already existing UF particles resulting in high particle number concentrations (up to 10^4–10^5 cm^{-3}) (Kumar et al., 2014). The spatial extent of the urban NPF phenomena has been investigated more recently (Dall'Osto et al., 2013; Zhu et al., 2014). Németh and Salma (2014) found that the nucleating air mass in regional NPF events may originate as far as several hundred kilometers away from the measurement location. Urban NPF and particle growth processes can occur as a part of a regional type NPF event, which leads to comparable NPF occurrence frequency at various locations within the region (Salma et al., 2016a).

The Central European cities of Budapest, Vienna, and Prague are located within 450 km from each other, and are comparable in size, population, and climate. Atmospheric NPF events in the three cities were independently studied previously (Řimnáčová et al., 2011; Salma

* Corresponding author.
E-mail address: anna.wonaschuetz@univie.ac.at (A. Wonaschütz).

https://doi.org/10.1016/j.atmosenv.2018.01.035
Received 21 July 2017; Received in revised form 20 December 2017; Accepted 18 January 2018
Available online 31 January 2018

et al., 2011; Wonaschütz et al., 2015). Previous studies on the regional extent of NPF events imply that simultaneous events could be expected between the locations (Németh and Salma, 2014; Salma et al. (2016a). UF particle number concentrations in the cities were compared in a joint paper (Borsós et al., 2012). As a continuation, we focus on the similarities and differences of NPF events in the three cities in this study. The main objectives of the paper are 1) to investigate the possible simultaneous occurrence of NPF events in three cities in the Central European region, 2) to determine the significance and relative contribution of atmospheric NPF events to UF particles in urban environments.

2. Methods

2.1. Experimental

The measurements were carried out in Budapest, Vienna and Prague, three capital cities in the Central European region. Budapest is located within the Carpathian Basin, Vienna at its northwestern boundary, whereas Prague is topographically more separated by the Bohemian Massif and other orographic features. Prague and Vienna are located about 440 km and 210 km to the northwest of Budapest, respectively, and the distance between Prague and Vienna is 250 km. The cities are similar in terms of area (525 km^2 for Budapest, 415 km^2 for Vienna, and 496 km^2 for Prague) and population (1.8, 1.9 and 1.3 million inhabitants respectively). The three cities have continental climate and similar urban characteristics discussed earlier in more detail by Borsós et al. (2012).

Particle number size distributions (PNSD) were measured continuously in the three cities from 1 January 2014 to 1 January 2016. In Budapest, the measurements were performed at the Budapest Platform for Aerosol Research and Training (BpART, 47°28′29.86″N, 19°03′44.76″E 115 m above mean sea level, a.m.s.l.) by a flow switching-type Differential Mobility Particle Sizer (Salma et al., 2011). Particles with an electrical mobility diameter range of 6–1000 nm were measured with a time resolution of ~8 min. The location is in the city centre, near the river Danube and represents well mixed urban air. Concentrations of air pollutants (SO_2, NO, NO_2, CO, O_3, PM_{10}) were obtained from the closest station of the National Air Quality Network located at a distance of 1.6 km in NW direction from the research platform. Global radiation (GRad) data were measured by the Hungarian Meteorological Service at a distance of 10 km in E direction. The time resolution of these data was 1 h.

In Vienna, PNSDs were measured at an urban background site at the University of Vienna (48°14′54″N, 16°21′42″E, 190 m a.m.s.l.). The site is located on a rooftop, and separated from major traffic routes by courtyards and two infrequently travelled small roads. Situated slightly above the neighborhood's roof level, the site receives air masses from all directions. Measurements were performed with two different set-ups during the campaign. A Vienna-type differential mobility analyzer (DMA) coupled with a CPC 3775 (TSI, USA) was operated from January 2014 to May 2015. The set-up recorded particles in the diameter range of 10–926 nm with a time resolution of ca. 10 min (Burkart et al., 2011). From June 2015 to January 2016, a DMA 3081 (TSI, USA) with a CPC 3772 or CPC 3775 were operated. Particles were measured in the electrical mobility diameter range of 10–505 nm and 13–673 nm with an increased time resolution of 5 min. The comparability of the two set-ups was tested by running them in parallel from April to June 2015. The temporal evolution of the size spectra was well comparable and the total number concentrations agreed within 20%. Meteorological parameters including GRad with 1-h resolution were measured at a distance of 1 km to the North of the site. The data were provided by the Zentralanstalt für Meteorologie und Geodynamik (ZAMG). Concentrations of O_3, SO_2, NO_x and NO_2 were obtained at a distance of 1.7 km to the South of the measurement site, and were provided by the Viennese Environmental Department monitoring network.

In Prague, PNSDs were measured from 1 January 2014 to 1 January 2016 at an urban background station located in the campus of Institute of the Chemical Process Fundamentals in Suchdol (50°7′36.47″N, 14°23′5.51″E, 277 m a.s.l.) by a Scanning Mobility Particle Sizer (SMPS 3034, TSI, USA) upgraded to the ACTRIS standard (Skrabalova et al., 2015). The site is located in a suburban area of Prague away from major roads. The SMPS measured size distributions between 7 and 540 nm with a time resolution of 5 min (Pfeifer et al., 2014). Meteorological data were measured with a time resolution of 10 min at the automated immission monitoring (AIM) station of the Czech Hydrometeorological Institute (CHMI) located directly in the campus. Air pollutant concentrations (SO_2, NO, NO_2, CO, O_3) were obtained from the same station, also from the CHMI, with a time resolution of 10 min.

2.2. Data treatment

Classification of the measurement days into different nucleation categories was performed according to a modified decision scheme based on Dal Maso et al. (2005). The original classification scheme was developed for clean, remote atmospheric environments without major pollution sources. The polluted, urban environments have a more complex character (e.g. rush hours due to traffic, ship emissions). This includes higher concentration levels, which also means larger fluctuation of concentration with spatial and temporal changes than at rural or remote sites. The Aitken mode and the whole size distribution is usually shifted to smaller diameters in cities. Special emission sources connected to anthropogenic activities (e.g. burning, grass cutting) in cities have been shown more recently (Skrabalova et al., 2015; Karl et al., 2016; Salma et al., 2016b). These features appear in the size distributions and should be taken into account to avoid misclassifications and to reduce the uncertainty of the classification. The modification of the original scheme was performed with respect to the sub-25 nm particles resulting the urban classification scheme (Fig. 1.). The classification was performed based on daily contour plots according to Kulmala et al.

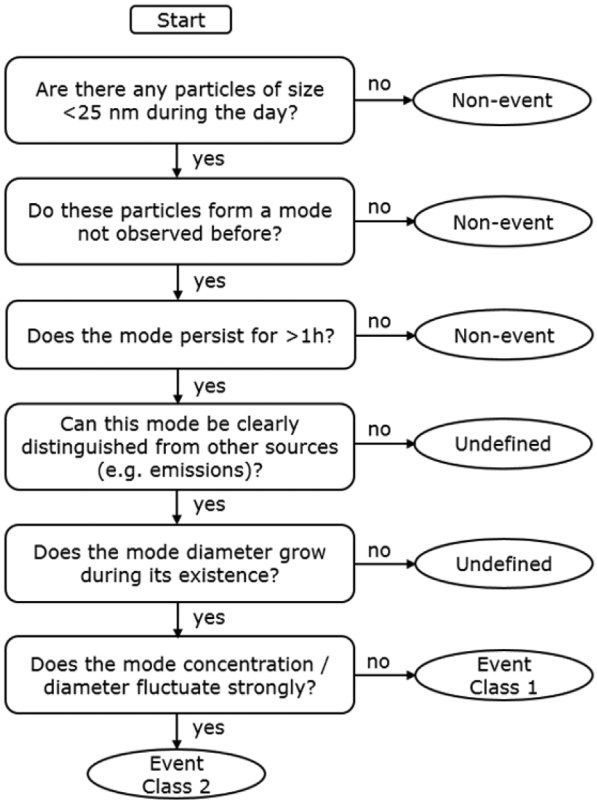

Fig. 1. Nucleation classification scheme specifically adapted for urban environments.

(2012). The distinction of the sub-25 nm particles from other sources was the key step instead of the observation of the mode itself. For instance, a clear traffic pattern - peaks in sub-25 nm particles in the morning and the evening - would be classified as "non-event", in spite of the sub-25 nm mode potentially lasting longer than an hour (an indication for "undefined" according to Dal Maso et al., 2005). As there is long-standing experience in identifying urban emission patterns in all three locations, the identification of a local source pattern in the daily contour plots was also occasionally cross-checked with previously used identification methods of traffic emissions: patterns in time series of integrated number concentrations of particles < 30, < 100 and < 300 nm (Prague), black carbon measurement time series (Vienna), and meticulous logbook entries (Budapest). This results a more reliable classification in urban environments. Thus, non-event days, undefined events, NPF event days with well-developed and interrupted shape (Class 1 and Class 2), and missing days were distinguished. The frequency of the different classes was calculated with respect to relevant days, when quantifiable data were collected.

Size distributions in a diameter range from 10 to 500 nm as the overlapping interval for the 3 cities were chosen for the evaluation. Hourly median particle number concentrations in the diameter ranges from 10 nm to 100 nm (N_{10-100}), from 100 to 500 ($N_{100-500}$) and from 10 to 500 nm (N_{10-500}) were generated to calculate the nucleation strength factor (NSF; Salma et al., 2014). NSF was defined as the particle number concentration ratio of ultrafine particles to the regional aerosol in accumulation mode on nucleation day to non-nucleation days, respectively:

$$\mathrm{NSF} = \frac{\left(N_{10-100}/N_{100-500}\right)_{\text{nucleation days}}}{\left(N_{10-100}/N_{100-500}\right)_{\text{non-nucleation days}}} \quad (1)$$

The NSF indicates the relative concentration increment of particles from NPF with respect to regional aerosol to the particles originating from all sources on event days. The importance of atmospheric nucleation is negligible if NSF < 1.0. In case of 1.0 < NSF > 2.0, NPF shows comparable importance to all other sources. When NSF > 2.0, the NPF has the highest contribution to UF particles than any other sources.

Hourly averaged meteorological data and air pollutant concentrations were obtained. The hourly median condensation sink (CS) for vapour molecules condensing onto the surface of existing aerosol particles was computed for discrete size distributions consisting of i channels according to Dal Maso et al. (2005):

$$\mathrm{CS} = 2\pi D \sum_i \beta_M(d_{p,i}) d_{p,i} N_i, \quad (2)$$

where D is the molecular diffusion coefficient of the condensing H_2SO_4 vapour in the air, β_M is the transition correction factor, $d_{p,i}$ and N_i are the diameter and number concentration of particles, respectively, for the size channel i. The condensation sink values were further utilized for calculating the H_2SO_4 proxy according to Petäjä et al. (2009):

$$[H_2SO_4] \alpha \frac{[SO_2] \times \mathrm{GRad}}{\mathrm{CS}}. \quad (3)$$

All three cities are in the same time zone, and so all the data were expressed in local time. This also made it feasible to compare the incidence of the NPF process at the 3 sites.

Air mass back-trajectories were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory Model 4 (HYSPLIT_4; Draxler and Rolph, 2013; Stein et al., 2015) with the Global Data Assimilation System (GDAS) meteorological database with a spatial resolution of 0.5° × 0.5°. The 60-h long backward trajectories were calculated for arrival heights of 100 m and 500 m above ground level starting at UTC 12:00. The modelling was performed for those days, when NPF took place on all three sites, or when the nucleation occurred in two cities while the day was unambiguously classified as a non-event day at the third site. Moreover, the Hess-Brezowsky catalogue (Hess and Brezowsky, 1952) was used to investigate the prevailing synoptic weather types in the region representative for the three investigated sites.

Table 1
Number (n) and relative contribution (f, in %) of NPF event, undefined, non-event, and missing days for the 2-year long campaign in Budapest, Vienna, and Prague.

	Budapest		Vienna		Prague	
	n	f	n	f	n	f
Nucleation days	152	21.2	69	12.4	143	30.2
Class 1 days	118	16.5	38	6.8	41	8.6
Class 2 days	34	4.7	31	5.6	102	21.5
Non-nucleation days	516	72.1	394	71.0	136	28.7
Undefined days	48	6.7	92	16.6	195	41.1
Missing days	14	–	175	–	256	–

3. Results and discussion

3.1. Classification and simultaneous NPF events

Classification of the days into the various categories is summarized in Table 1. The relative nucleation frequency over the 2-year long time interval was 21% in Budapest, 12% in Vienna and 30% in Prague, respectively. The nucleation frequencies showed a different seasonal distribution. Most NPF events occurred unambiguously in late spring and in early summer in Vienna and Prague. The monthly distribution of nucleation frequency in Budapest showed two distinct maxima, one in spring and one in autumn. The inter-annual variability did not show substantial differences in the cities. This enables the joint evaluation of the 2-year long dataset. The missing days in Prague were a result of measurement failures and had uniform distribution over the 2 years. In Vienna, 109 of the missing days occurred in winter, 14 in spring, 20 in summer, and 32 in fall. The nucleation frequency of 12% for Vienna reported in this study may therefore be a slight over-estimate, but is comparable to an earlier study for the same site (Wonaschütz et al., 2015), where a nucleation frequency of 11.5% was reported. The number of simultaneous NPF events taking place at all three sites was 12. Five out of the 12 days occurred in April, and 11 out of the 12 days took place in spring and in summer. There were two consecutive days, when NPF happened at all sites. Pairwise investigation was also performed, meaning that NPF events took place in 2 cities and there was a non-event day in the third city. This showed that there were 2, 19, and 19 days with nucleation in Budapest-Vienna, Budapest-Prague, and Vienna-Prague, respectively (Table 2). Their distribution was inhomogeneous during the 2 years. The number of these pairwise NPF events is unexpected, as Budapest and Vienna are located closer to each other than Budapest and Prague.

3.2. Atmospheric concentrations

Overall median particle number concentrations of N_{10-500} were 8889 cm^{-3} in Budapest, 5395 cm^{-3} in Vienna, and 5844 cm^{-3} in

Table 2
Number of days with NPF in all cities, in 2 cities and without NPF.

City with NPF	City without NPF	Number of days
–	Budapest, Vienna, Prague	55
Budapest, Vienna, Prague	–	12
Budapest, Vienna	Prague	2
Budapest, Prague	Vienna	19
Vienna, Prague	Budapest	19

Table 3
Statistical overview of daily median concentrations in cm^{-3} in Budapest, Vienna, and Prague.

	Budapest			Vienna			Prague		
	$N_{10\text{-}25}$	$N_{10\text{-}100}$	$N_{100\text{-}500}$	$N_{10\text{-}25}$	$N_{10\text{-}100}$	$N_{100\text{-}500}$	$N_{10\text{-}25}$	$N_{10\text{-}100}$	$N_{100\text{-}500}$
Min	367	1107	321	10	273	124	297	750	138
Median	2096	6449	2663	1058	3945	1414	1625	4660	1298
Max	6093	17238	9380	4423	16350	14953	7829	14127	6967
Mean	2170	6695	2882	1137	4439	1756	1893	5017	1505
St. dev.	862	2597	1497	832	2318	1590	1080	2282	966

Prague. The present choice of $N_{10\text{-}500}$ as total particle number concentration due to the overlapping size distribution range in the 3 cities was 20% lower compared to $N_{10\text{-}1000}$ (Salma et al., 2011). Budapest with a measurement site in the city centre had the highest total particle concentration, while the other two locations, characterized as urban background, showed moderate concentrations (Table 3). Significant seasonal variations of the concentrations were not observed at any of the sites. The contributions of UF particles to the total particle number concentration (expressed as $N_{10\text{–}100}/N_{10\text{–}500}$) were rather high, 0.71 in Budapest, 0.73 in Vienna, and 0.78 in Prague, and did not seem to have significant temporal variation. This can be explained by the phenomenon that primary emissions and NPF take place in an alternating way in the urban air: NPF occurs usually on days having smaller condensation sink values and thus cleaner air mass than days dominated by primary emissions. This leads to a more or less constant level of UF particles (Salma et al., 2011).

The diurnal variation of UF particles showed complex characteristics (Fig. 2). Two distinct peaks occurred in Prague on non-event days,

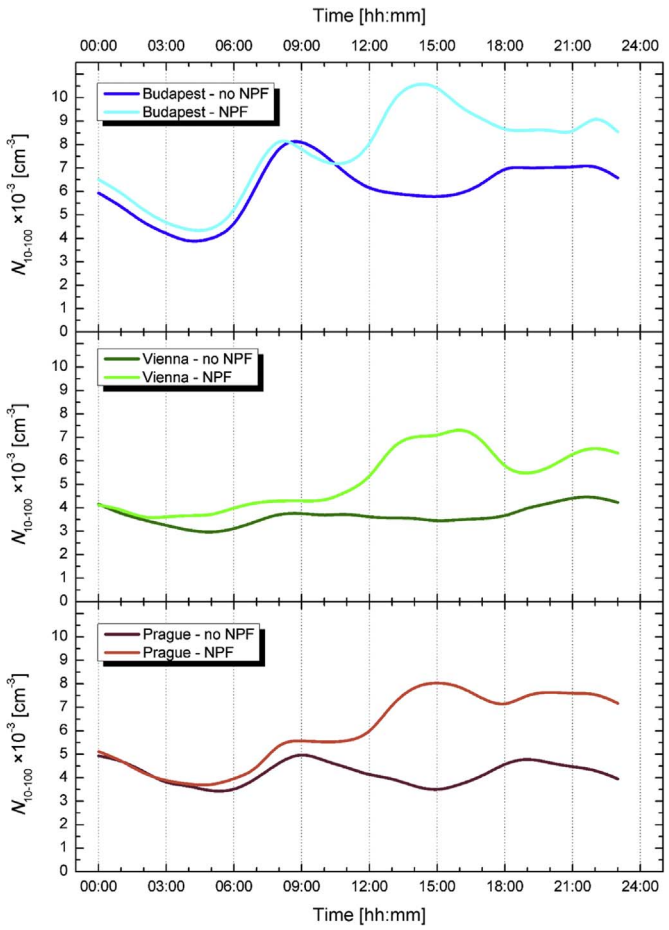

Fig. 2. Diurnal variation of UF particles for the Budapest, Vienna, and Prague.

one in the morning and the other one in the late afternoon. This was associated with traffic emissions. The concentration increased around 7:00 and rose again in the late afternoon in accordance with the rush hours. In the city centre of Budapest, the concentration of UF particles was higher than in the other two cities. Primary particles emitted by traffic had a more significant effect on the diurnal variation. The peak caused by the UF particles originating from NPF was superposed on the curve of non-event days. The peak of UF particles from NPF was relatively higher than the two other traffic peaks. This is due to the fact that the source and sink terms of available gas-phase H_2SO_4 determine together whether nucleation takes place or not (Salma et al., 2016b). The sink term (surface of pre-existing aerosol) is higher in the more polluted city centre, and thus a stronger source term is generally needed for the new particle formation to happen.

3.3. Relevance of atmospheric nucleation in cities

Overall mean nucleation strength factors of 1.58, 1.54, and 2.01 were obtained for Budapest, Vienna, and Prague, respectively. This implies the considerable contribution of atmospheric nucleation to urban particle sources. The NSF values in Budapest varied from 1.31 to 1.73. They followed the occurrence of NPF frequency pattern, except in winter. The latter difference is partially caused by the usually low number of NPF events in winter. The other cause was two very strong nucleation events, when the monthly mean NSF reached the value of 2.8 (2014 Dec), and 4.1 (2015 Dec) due to the unusually low background particle number concentrations on those particular days. The seasonal NSF values had the same pattern in Vienna and Prague (Fig. 3.), they vary from 1.14 to 1.62 (Vienna), and from 1.25 to 2.09 (Prague). This is connected to the seasonal dependence of the NPF frequency for these cities. It is worth noting that the NSF calculation with $N_{100\text{-}500}$ instead of the usual $N_{100\text{-}1000}$ resulted in a 5% increase in the NSF values. This means that the NSF is not very sensitive in the particle diameter range from 500 nm to 1000 nm, which allows using it as a quantifying parameter by instruments with different upper cut-off sizes.

The diurnal variation curves of the NSFs had similar shapes for the three cities (Fig. 4.). The peaks can be assigned to NPF events. All the three NSF curves have a monotonously increasing start after 9:00 in the morning. This feature can be also seen on the contour plots or by examining the particle number concentration time series. The diurnal NSF curves can be affected by special types of banana curve as well, including multiple-onset NPF events, shrinkage of the newly formed particles, broad onsets when the formation process takes place for several hours, or nocturnal events (Skrabalova et al., 2015; Salma et al., 2016b; Salimi et al., 2017). The particles originating from NPF become the dominant source of UF particles in the time interval of 11–17 h. This is the typical time interval for NPF events, which underlines the importance of characteristic time parameters. These characteristic time parameters and their determination were introduced in a previous work for Budapest (Németh and Salma, 2014).

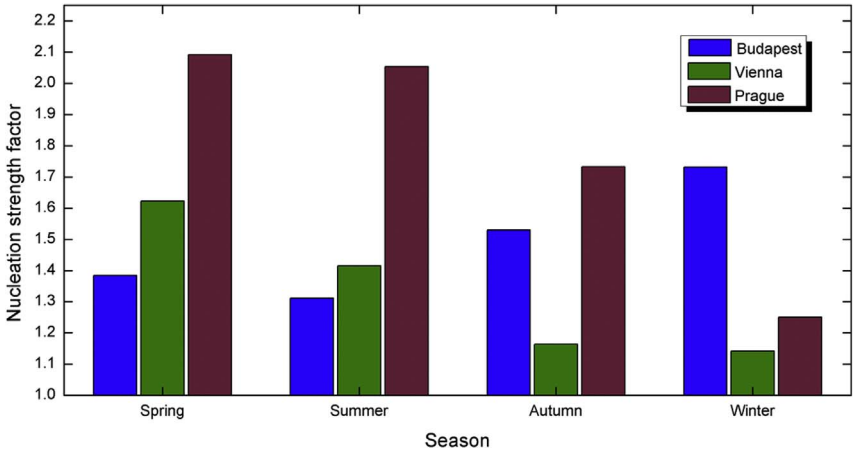

Fig. 3. Nucleation strength factors for various seasons in Budapest, Vienna, and Prague.

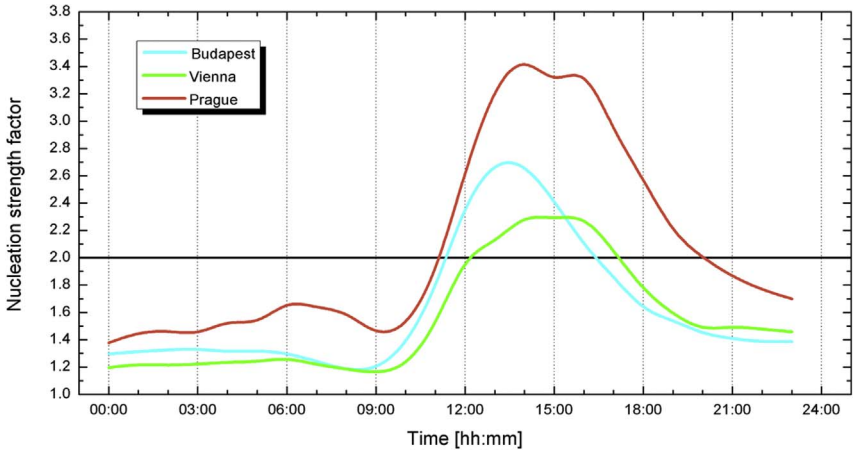

Fig. 4. Diurnal variation of the nucleation strength factors for Budapest, Vienna, and Prague.

Table 4
Median gas-phase H_2SO_4 proxy on nucleation, undefined and non-nucleation days in Budapest, Vienna, and Prague.

	Budapest	Vienna	Prague
	Proxy × 10^{-3} [μg m^{-5} W s]	Proxy × 10^{-3} [μg m^{-5} W s]	Proxy × 10^{-3} [μg m^{-5} W s]
Nucleation days	134	190	106
Undefined days	122	98	52
Non-nucleation days	56	82	34

3.4. Simultaneous new particle formation events

The general weather situation over the region was examined using the Hess-Brezowsky typization method. On 5 days (04 April 2015, 29 April 2015, 10 July 2015, 11 July 2015 and 26 July 2015) of the 12 nucleation days, when NPF occurred at all sites (Table 2.), high pressure systems prevailed. The other 7 days (20 April 2014, 24 May 2014, 26 May 2014, 20 June 2014, 09 April 2015, 21 April 2015 and 30 December 2015) were diverse, showing low as well as high pressure situations. The full analysis of the 2 years showed that there was no preferred Hess-Brezowsky synoptic type on NPF event days compared to non-events days. The analysis also revealed clear inter-annual variations of synoptic types. Therefore, the general weather situation over Central Europe does not seem to play a crucial role for NPF events in the three cities.

Air mass back-trajectories on the above mentioned 5 out of 12 days with NPF in all cities were similar and indicated northerly and northwesterly air mass origins. On the other 7 days the back-trajectories showed no clear trend. Investigation of air mass back-trajectories for the days, when NPF occurred in 2 out of 3 cities was also performed. On the 2 days, when Budapest and Vienna (but not Prague) had NPF events, there was no clear difference in air mass origins for all three cities. In the case of simultaneous NPF in Budapest and Prague (but not Vienna), only 1 day featured similar back-trajectories for Budapest and Prague that clearly differed from the one for Vienna. There were also 19 days, when NPF occurred in Vienna and Prague, and Budapest had non-event days. On 5 of them, the air mass origins (mainly originating from the North) were comparable for Vienna and Prague, and clearly different to those for Budapest. Therefore, the differences among sites could not clearly be explained by air mass trajectories.

The local meteorology related to NPF was investigated on an hourly basis. The median gas-phase H_2SO_4 proxy showed substantial differences for event, undefined and non-event days for the 2 year-long interval (Table 4.). The median diurnal variations of the proxy for nucleation and non-nucleation days were calculated as well (Fig. 5.). The maximum of the curves varied substantially, and was 125×10^3 W m^{-2} μg m^{-3} s in Budapest, 202×10^3 W m^{-2} μg m^{-3} s in Vienna, and 79×10^3 W m^{-2} μg m^{-3} s in Prague on non-event days. On event days, the peak of the diurnal proxy curve was 324×10^3 W m^{-2} μg m^{-3} s in Budapest, 471×10^3 W m^{-2} μg m^{-3} s in Vienna, and 269×10^3 W m^{-2} μg m^{-3} s in Prague. The main driver of the higher proxy values in Vienna, as compared to Prague and Budapest, are the consistently higher global radiation values. Similarly, the low proxy values for Prague mostly reflect the consistently lower solar radiation.

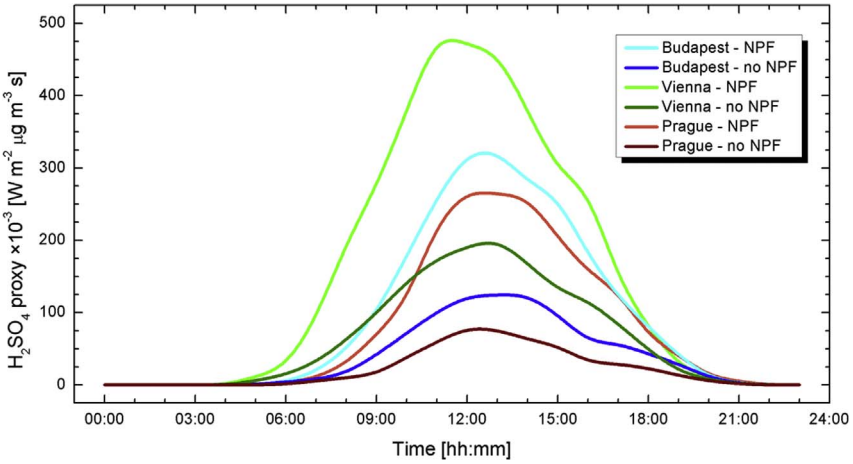

Fig. 5. Median diurnal variation of gas-phase H_2SO_4 proxy for NPF event and non-event days in Budapest, Vienna, and Prague.

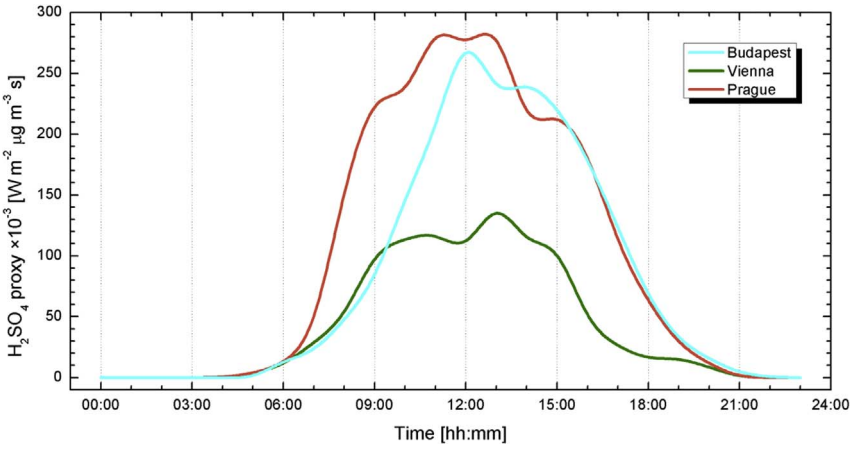

Fig. 6. Diurnal variation of gas-phase H_2SO_4 proxy expressed as a difference between the mean proxy of the days when nucleation took place only in Budapest and Prague, and the 2 year-long mean proxy for non-event days.

Median CS values are lower in Prague and Vienna than in Budapest. Median SO_2 concentrations for non-event days in Vienna are lower than those in Prague and Budapest, whereas on event days, median SO_2 for Vienna is comparable to that of Budapest. Due to the different proxy values in the 3 cities, the proxy was expressed for comparative purposes as a difference between the mean proxy of the days when nucleation took place only at 2 sites and the 2 year-long mean proxy on non-event days. When Budapest and Prague had NPF event and Vienna had non-event days, Budapest and Prague exhibited high proxy values which were favorable for NPF (Fig. 6.). On these days, Vienna still had photochemically more active air mass than on the other non-event days, when NPF did not occur simultaneously in Budapest and Prague. This pattern was recognized for the other 2 cases (when only Budapest-Vienna and Vienna-Prague had NPF event days) as well. The difference of proxy values was higher by a factor of approx. 2.0 on event days compared to non-event days. The shape of the curves was similar for all cities.

4. Summary and conclusions

Particle number size distributions were measured in 3 Central European cities continuously for 2 years. The total particle number concentrations were found to be of similar order of magnitude, but the city centre environment (Budapest) showed elevated levels compared to the urban background sites (Vienna and Prague). The contribution of UF particles to the total aerosol population was more than 70% in all three cities. NPF events occurred on 152 (Budapest), 69 (Vienna), and 143 (Prague) days in the 2 year-long period. There were 12 days, when NPF occurred in all three cities. On 2 (Budapest-Vienna), 19 (Budapest-Prague), and 19 (Vienna-Prague) event days, NPF did not occur at the third site. Regional and local differences in the atmospheric environment played significant roles as far as NPF is concerned. The differences in NPF occurrence in the 3 cities were explained mainly by the diurnal variation of gas-phase H_2SO_4 proxy.

Acknowledgements

Financial supports by the Hungarian Scientific Research Fund (contract K116788 and PD124283), by the European Union's Horizon 2020 research and innovation programme ACTRIS-2 (grant agreement No. 654109), ACTRIS - CZ (num. LM2015037), and ACTRIS – CZ RI (CZ.02.1.01/0.0/0.0/16_013/0001315) are appreciated. This work was supported by GINOP-2.3.2-15-2016-00055 Project through the National Research, Development and Innovation Office, Hungary. The authors thank the Austrian Central Institute for Meteorology and Geodynamics (ZAMG) for providing meteorological data and the Viennese Environmental Department (MA 22) for air quality monitoring network data.

References

Borsós, T., Řimnáčová, D., Ždímal, V., Smolík, J., Wagner, Z., Weidinger, T., Burkart, J., Steiner, G., Reischl, G., Hitzenberger, R., Schwarz, J., Salma, I., 2012. Comparison of particulate number concentrations in three Central European capital cities. Sci. Total Environ. 433, 418–426.

Brines, M., Dall'Osto, M., Beddows, D.C.S., Harrison, R.M., Gómez-Moreno, F., Núñez, L., Artíñano, B., Costabile, F., Gobbi, G.P., Salimi, F., Morawska, L., Sioutas, C., Querol, X., 2015. Traffic and nucleation events as main sources of ultrafine particles in high-insolation developed world cities. Atmos. Chem. Phys. 15, 5929–5945.

Burkart, J., Steiner, G., Reischl, G., Hitzenberger, R., 2011. Long-term study of cloud condensation nuclei (CCN) activation of the atmospheric aerosol in Vienna. Atmos. Environ. 45, 5751–5759.

Dal Maso, M., Kulmala, M., Lehtinen, K.E.J., Mäkelä, J.M., Aalto, P.P., O'Dowd, C., 2002. Condensation and coagulation sinks and formation of nucleation mode particles in coastal and boreal forest boundary layers. J. Geophys. Res. 107 (19D), 8097. http://dx.doi.org/10.1029/2001jd001053. 2002.

Dal Maso, M., Kulmala, M., Riipinen, I., Wagner, R., Hussein, T., Aalto, P.P., Lehtinen, K.E.J., 2005. Formation and growth of fresh atmospheric aerosols: eight years of aerosol size distribution data from SMEAR II. Hyytiälä, Finland. Boreal Env. Res. 10, 323–336.

Dall'Osto, M., Querol, X., Alastuey, A., O'Dowd, C., Harrison, R.M., Wenger, J., Gómez-Moreno, F.J., 2013. On the spatial distribution and evolution of ultrafine particles in Barcelona. Atmos. Chem. Phys. 13, 741–759.

Dameto de España, C., Wonaschütz, A., Steiner, G., Rosati, B., Demattio, A., Schuh, H., Hitzenberger, R., 2017. Long-term quantitative field study of New Particle Formation (NPF) events as a source of Cloud Condensation Nuclei (CCN) in the urban background of Vienna. Atmos. Environ 164, 289–298.

Draxler, R.R., Rolph, G.D., 2013. HYSPLIT (HYbrid Single-particle Lagrangian Integrated Trajectory) Model. NOAA Air Resources Laboratory, College Park, MD. http://www.arl.noaa.gov/HYSPLIT.php (last access: 16 May 2017).

Geiser, M., Rothen-Rutishauser, B., Kapp, N., Schürch, S., Kreyling, W., Schulz, H., Semmler, M., Im Hof, V., Heyder, J., Gehr, P., 2005. Ultrafine particles cross cellular membranes by nonphagocytic mechanisms in lungs and in cultured cells. Environ. Health Perspect. 113, 1555–1560.

HEI Review Panel on Ultrafine Particles, 2013. Understanding the Health Effects of Ambient Ultrafine Particles. HEI Perspectives 3. Health Effects Institute, Boston.

Hess, P., Brezowsky, H., 1952. Catalogue of European Large-scale Weather Situations (Katalog der Grosswetterlagen Europas) (in German). Berichte des Deutschen Wetterdienstes in der US-Zone 33. Bad Kissingen, Germany.

Holmes, N.S., 2007. A review of particle formation events and growth in the atmosphere in the various environments and discussion of mechanistic implications. Atmos. Environ. 41, 2183–2201.

Karl, M., Kukkonen, J., Keuken, M.P., Lützenkirchen, S., Pirjola, L., Hussein, T., 2016. Modeling and measurements of urban aerosol processes on the neighborhood scale in Rotterdam, Oslo and Helsinki. Atmos. Chem. Phys. 16, 4817–4835.

Kulmala, M., Vehkamäki, H., Petäjä, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W., McMurry, P., 2004. Formation and growth rates of ultrafine atmospheric particles: a review of observations. J. Aerosol Sci. 35, 143–176.

Kulmala, M., Petäjä, T., Nieminen, T., Sipilä, M., Manninen, H.E., Lehtipalo, K., Dal Maso, M., Aalto, P.P., Junninen, H., Paasonen, P., Riipinen, I., Lehtinen, K.E.J., Laaksonen, A., Kerminen, V.-M., 2012. Measurement of the nucleation of atmospheric aerosol particles. Nat. Protoc. 7, 1651–1667. http://dx.doi.org/10.1038/nprot.2012.091.

Kulmala, M., Kontkanen, J., Junninen, H., Lehtipalo, K., Manninen, H.E., Nieminen, T., Petäjä, T., Sipilä, M., Schobesberger, S., Rantala, P., Franchin, A., Jokinen, T., Järvinen, E., Äijälä, M., Kangasluoma, J., Hakala, J., Aalto, P.P., Paasonen, P., Mikkilä, J., Vanhanen, J., Aalto, J., Hakola, H., Makkonen, U., Ruuskanen, T., Mauldin III, R.L., Duplissy, J., Vehkamäki, H., Bäck, J., Kortelainen, A., Riipinen, I., Kurtén, T., Johnston, M.V., Smith, J.N., Ehn, M., Mentel, T.F., Lehtinen, K.E.J., Laaksonen, A., Kerminen, V.-M., Worsnop, D.R., 2013. Direct observations of atmospheric aerosol nucleation. Science 339, 943–946.

Kumar, P., Morawska, L., Birmili, W., Paasonen, P., Hu, M., Kulmala, M., Harrison, R.M., Norford, L., Britter, R., 2014. Ultrafine particles in cities. Environ. Int. 66, 1–10.

Manninen, H.E., Nieminen, T., Asmi, E., Gagné, S., Häkkinen, S., Lehtipalo, K., Aalto, P., Vana, M., Mirme, A., Mirme, S., Hörrak, U., Plass-Dülmer, C., Stange, G., Kiss, G., Hoffer, A., Törő, N., Moerman, M., Henzing, B., de Leeuw, G., Brinkenberg, M., Kouvarakis, G.N., Bougiatioti, A., Mihalopoulos, N., O'Dowd, C., Ceburnis, D., Arneth, A., Svenningsson, B., Swietlicki, E., Tarozzi, L., Decesari, S., Facchini, M.C., Birmili, W., Sonntag, A., Wiedensohler, A., Boulon, J., Sellegri, K., Laj, P., Gysel, M., Bukowiecki, N., Weingartner, E., Wehrle, G., Laaksonen, A., Hamed, A., Joutsensaari, J., Petäjä, T., Kerminen, V.-M., Kulmala, M., 2010. EUCAARI ion spectrometer measurements at 12 European sites - analysis of new-particle formation events. Atmos. Chem. Phys. 10, 7907–7927.

Németh, Z., Salma, I., 2014. Spatial extension of nucleating air masses in the Carpathian Basin. Atmos. Chem. Phys. 14, 8841–8848.

Oberdörster, G., Oberdörster, E., Oberdörster, J., 2005. Nanotoxicology: an emerging discipline evolving from studies of ultrafine particles. Environ. Health Perspect. 113, 823–839.

Petäjä, T., Mauldin III, R.L., Kosciuch, E., McGrath, J., Nieminen, T., Paasonen, P., Boy, M., Adamov, A., Kotiaho, T., Kulmala, M., 2009. Sulfuric acid and OH concentrations in a boreal forest site. Atmos. Chem. Phys. 9, 7435–7448.

Pfeifer, S., Birmili, W., Schladitz, A., Müller, T., Nowak, A., Wiedensohler, A., 2014. A fast and easy-to-implement inversion algorithm for mobility particle size spectrometers considering particle number size distribution information outside of the detection range. Atmos. Meas. Tech 7, 95–105.

Salimi, F., Rahman, Md. M., Clifford, S., Ristovski, Z., Morawska, L., 2017. Nocturnal new particle formation events in urban environments. Atmos. Chem. Phys. 17, 521–530.

Salma, I., Borsós, T., Weidinger, T., Aalto, P., Hussein, T., Dal Maso, M., Kulmala, M., 2011. Production, growth and properties of ultrafine atmospheric aerosol particles in an urban environment. Atmos. Chem. Phys. 11, 1339–1353.

Salma, I., Borsós, T., Németh, Z., Weidinger, T., Aalto, P., Kulmala, M., 2014. Comparative study of ultrafine atmospheric aerosol within a city. Atmos. Environ. 92, 154–161.

Salma, I., Füri, P., Németh, Z., Balásházy, I., Hofmann, W., Farkas, Á., 2015. Lung burden and deposition distribution of inhaled atmospheric urban nanoparticles as the first step in their health risk assessment. Atmos. Environ. 104, 39–49.

Salma, I., Németh, Z., Kerminen, V.-M., Aalto, P., Nieminen, T., Weidinger, T., Molnár, Á., Imre, K., Kulmala, M., 2016a. Regional effect on urban atmospheric nucleation. Atmos. Chem. Phys. 16, 8715–8728.

Salma, I., Németh, Z., Weidinger, T., Kovács, B., Kristóf, G., 2016b. Measurement, growth types and shrinkage of newly formed aerosol particles at an urban research platform. Atmos. Chem. Phys. 16, 7837–7851.

Samoli, E., Andersen, Z.J., Katsouyanni, K., Hennig, F., Kuhlbusch, T.A.J., Bellander, T., Cattani, G., Cyrys, J., Forastiere, F., Jacquemin, B., Kulmala, M., Lanki, T., Loft, S., Massling, A., Tobias, A., Stafoggia, M., 2016. Exposure to ultrafine particles and respiratory hospitalisations in five European cities. Eur. Respir. J. 48, 674–682.

Skrabalova, L., Zikova, N., Zdimal, V., 2015. Shrinkage of newly formed particles in an urban environment. Aerosol Air Qual. Res. 15, 1313–1324.

Spracklen, D.V., Carslaw, K.S., Kulmala, M., Kerminen, V.-M., Mann, G.W., Sihto, S.-L., 2006. The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales. Atmos. Chem. Phys. 6, 5631–5648.

Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J.B., Cohen, M.D., Ngan, F., 2015. NOAA's HYSPLIT atmospheric transport and dispersion modeling system. Bull. Am. Meteorol. Soc. 96, 2059–2077.

Wåhlin, P., Palmgren, F., Van Dingenen, R., 2001. Experimental studies of ultrafine particles in streets and the relationship to traffic. Atmos. Environ. 35, 63–69.

Wonaschütz, A., Demattio, A., Wagner, R., Burkart, J., Zíková, N., Vodička, P., Ludwig, W., Steiner, G., Schwarz, J., Hitzenberger, R., 2015. Seasonality of new particle formation in Vienna, Austria - influence of air mass origin and aerosol chemical composition. Atmos. Environ. 118, 118–126.

Zhu, Y.J., Sabaliauskas, K., Liu, X.H., Meng, H., Gao, H.W., Jeong, C.-H., Evans, G., Yao, X.H., 2014. Comparative analysis of new particle formation events in less and severely polluted urban atmosphere. Atmos. Environ. 98, 655–664.

Řimnáčová, D., Ždímal, V., Schwarz, J., Smolík, J., Řimnáč, M., 2011. Atmospheric aerosols in suburb of Prague: the dynamics of particle size distributions. Atmos. Res. 20, 539–552.