

**HEALTH AND ENVIRONMENTAL CONSEQUENCES OF ATMOSPHERIC NANO-AEROSOL
RESEARCH REPORT – PD124283**

Introduction

Ultrafine atmospheric aerosol particles (UF, $d < 100$ nm) have increasingly become the subject of scientific investigations. They are distinguished as primary and secondary particles according to their formation process. The primary particles are directly emitted by high temperature processes such as vehicle traffic, burning, residential heating and cooking. The main formation process of the secondary particles is the atmospheric nucleation. It is a major source of particle number concentration on global scale. New particle formation was first identified at remote sites, later it was observed at various geographical locations, from the free troposphere to large cities. The ultrafine atmospheric aerosol particles have been in the focus of scientific interest due to their effects on public health in cities and on climate.

Concentration of UF particles can reach up to 10^4 – 10^5 cm^{-3} in cities, and it varies in time by 2 orders of magnitude. Their contribution to the total particle concentration is 70–90%. The UF particles entering into the human respiratory system represent excess health risk compared to the fine or coarse particles with similar chemical composition due to their very small size and hence higher biological activity.

Aerosol particles affect directly the climate of the Earth by scattering (e.g. sulphate particles) or absorbing (e.g. soot particles) the solar radiation. The UF particles are able to grow and later act as cloud condensation nuclei (CCN) in the presence of supersaturation of water vapor in the atmosphere. It is the atmospheric nucleation that produces up to 50% of the particles, which act as CCN. This is an indirect effect, that has the highest single uncertainty among the climate forcing factors.

This PD project focused on the effects of atmospheric ultrafine particles and the measurements were carried out in Budapest according to the project proposal.

Results

Research project covered 4 subtopics related to urban new particle formation, and its properties, dynamics. According the project proposal, one subtopic was changed. Dynamic properties of NPF were analyzed and evaluated on a global scale instead of the hygroscopicity related lung deposition of UF particles (see 4. result). This was in connection with the fact that an ad-hoc organized international research cooperation made feasible to integrate and evaluate the urban Budapest NPF data into a global database. The Budapest site was one out of the 6 urban, and one out of the 36 global measurement sites that were discussed in the project. The authors of the paper published on this topic are leading experts

in the field of NPF. The PD related subtopics are discussed in detail in the original research articles. Therefore, a short summary on each subtopic is presented here.

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

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 nucleation strength factor (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 particles, and the amount of them is comparable to the directly emitted UF particles. The diurnal variation curves of the NSFs had similar shapes for the three cities. The peaks can be assigned to NPF events. 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. 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.

2. Hygroscopicity and volatility of atmospheric ultrafine particles

Hygroscopic growth factors (GFs), volatility GFs and hygroscopicity parameters were quantified for ambient aerosol particles with dry diameters of 50, 75, 110 and 145 nm in situ by using a VH-TDMA system in central Budapest during two months in winter. The measurements were supported by a DMPS system and meteorological sensors, which were operated in parallel. The urban aerosol showed distinct bimodality with respect to both hygroscopic and volatile properties, which indicated that the urban

aerosol contained an external mixture of particles with a diverse chemical composition. Vehicular road traffic had significant influence on both the hygroscopic and volatile properties, and contributed substantially to the particles in the nearly hydrophobic (NH) and less hygroscopic (LV) modes. These two modes were associated with each other, and both followed the typical diurnal pattern of road traffic and its workday/weekend variation. The other hygroscopic mode, the LH mode was composed of moderately transformed aged combustion particles consisting of partly oxygenated organics, inorganic salts and soot, and typically exhibited a volatility GF of approximately 0.6. Both the hygroscopic GFs and volatility GFs showed modest size dependent behaviour, while the fraction of particles in the modes exhibited much stronger size dependency. Smaller particle diameters were associated with a larger number fraction of non-volatile and hydrophobic particles than the larger diameters. This can be explained by assuming that the larger particles grew by condensation of organic vapours, and it is also supported by the small size dependency in the κ values with respect to the dry particle size. The 50-nm particles, however, had a considerably lower κ value and showed 30 larger volatility during daytime in the LH mode with respect to the larger diameters. This suggests that these particles have a different chemical composition than the larger particles. The transformation process of soot particles from hydrophobic to hydrophilic in the atmosphere is still not sufficiently understood. This measurement emphasizes the importance of the mixing state of particles on influencing their hygroscopic properties. The ambient conditions during the campaign were typical for wintertime Budapest. Since there are strong seasonal variations in both the anthropogenic and natural components, the hygroscopic and volatile properties are also expected to change. Therefore, further similar in situ measurements should be carried out in different seasons in the future together with on-line chemical characterisation of particles to better quantify and understand the properties, relevance and role of urban aerosol.

3. Dynamic properties and timing of urban new particle formation in Budapest

Magnitude of the particle number concentration level produced solely by NPF and growth can roughly be estimated by considering the median J_6 , median duration of nucleation (Δt), and the mean coagulation loss of these particles (F_{coag}). In central Budapest, it yields a concentration of 10.000 cm^{-3} . This is in line with another result achieved using the nucleation strength factor. More importantly, the estimated concentration is comparable to the annual median atmospheric concentrations. This example shows that the phenomenon is relevant not only for aerosol load and climate issues on regional or global spatial scales, which were first recognized. It is sensible also to study the effects of NPF and growth events on urban climate and health risk for inhabitants since they produce a large fraction of particles, even in cities. Similar recognitions have led urban atmospheric nucleation studies to emerge. As part of this international progress, we have presented a considerable variety of contributions, which became feasible due to gradually generating, multi-year-long, critically evaluated, complex and coherent data sets. Dynamic and timing properties of 247 NPF and growth events were studied together with supporting

aerosol properties, meteorological data and pollutant gas concentrations in the near-city background and city centre of Budapest for 6 years. The results and conclusions derived from an important component that is based on atmospheric observations. This research has been considered as the first step toward a larger and more comprehensive statistical evaluation process.

4. International, comprehensive analysis of dynamic properties of new particle formation on a global scale

A data base on continuous particle number size distribution measurements at 36 continental sites worldwide was realized, the research study investigated the overall and seasonal behavior of regional new particle formation in five very different environmental regimes ranging from polar and other remote areas to heavily-polluted megacities. The study showed that the regional NPF events to take place at all the measurement sites throughout the year, being most common (median of site-median values of about 30%) during the northern-hemisphere spring and least common (less than 10%) during the winter. No clear environmental pattern in the frequency of NPF was observed, except that NPF events seemed to be most rare in polar areas during most seasons. It turned out that the formation rates of 10–25 nm particles (J_{nuc}) during the NPF events tended to increase with an increasing degree of anthropogenic influence, being one to two orders of magnitude higher in urban areas compared with most of the remote and polar sites. The seasonal variability of J_{nuc} was quite modest at most of the sites. There was not any systematic environmental pattern for the growth rate (GR) of 10–25 nm particles during the NPF events. For the vast majority of the sites, the seasonal-median values of GR were the highest during the local summer and the lowest during the local winter. The observed values of J_{nuc} , GR and NPF indicate that regional NPF can explain a dominant fraction of the total particle number concentration, and give an important contribution to a cloud condensation nuclei population, in both remote and heavily-polluted continental locations. The research revealed that the connection between J_{nuc} , GR and NPF event frequency was at best moderate between the different measurement sites, as well as between the sites belonging to some environmental regime. The apparent lack of a strong relation between these three quantities is understandable due to the environmental and seasonal variability in the dominant new-particle formation mechanisms, in the abundancies of compounds contributing to the initial steps of NPF and subsequent particle growth, and in the prevailing meteorological conditions.

Checkpoints

The original research proposal and the realized results, checkpoints are in good agreement in terms of checkpoints. In terms of original research articles in high impact factor and class Q1, peer-reviewed scientific journals, 2 papers a year were submitted and accepted. The cumulative impact factor for the published articles is approx. 21.0. In the 2 years of the project, at least 2 abstracts for international conferences a year were submitted and accepted.

International conferences:

Zoltán, Németh; Tamás, Weidinger; Ernő, Keszei; Pasi, Aalto; Tuuka, Petäjä; Markku, Kulmala; Imre, Salma, Multi-year long new particle formation in urban environments (2017), 20th International Conference on Nucleation and Atmospheric Aerosols, Helsinki, Finland

Imre, Salma; Zoltán, Németh; Veronika, Varga; Markku, Kulmala, Special features and relevance of new aerosol particle formation and growth process in cities (2017), 20th International Conference on Nucleation and Atmospheric Aerosols, Helsinki, Finland

Zoltán, Németh; Veronika, Varga; Tamás, Weidinger; Imre, Salma, Multi-year long measurement of urban new aerosol particle formation and its relation to local meteorology (2018), EMS Annual Meeting Abstracts, Budapest, Hungary

Santtu, Mikkonen; Zoltán, Németh; Veronika, Varga; Tamás, Weidinger; Ville, Leinonen; Taina, Yli-Juuti; Imre, Salma, Time trends of particle number concentrations in a Central European city between 2008 and 2018 (2019), European Aerosol Conference, Gothenburg, Sweden

Imre, Salma; Zoltán, Németh, Dynamic and timing properties of new aerosol particle formation and consecutive growth events, (2019), European Aerosol Conference, Gothenburg, Sweden

Other project related publications:

Salma, I., Németh, Z., Varga, V., Weidinger, T., Légekőri nukleáció és következményei, Magyar Kémiai Folyóirat 125 (2019) 64–69.

Németh, Zoltán; Bernadette, Rosati; Naděžda, Zíková; Salma, Imre; Bozó, László; Regina, Hitzenger; Jaroslav, Schwarz; Vladimír, Ždímal; Anna, Wonaschütz, Újrézecske-képződés jelentősége három közép-európai városi környezetben, 6. Környezetkémiai Szimpózium, Bakonybél, Magyarország (2017)

Németh, Zoltán; Bernadette, Rosati; Naděžda, Zíková; Salma, Imre; Bozó, László; Regina, Hitzenger; Jaroslav, Schwarz; Vladimír, Ždímal; Anna, Wonaschütz, A légekőri nukleáció jelentősége közép-európai városi környezetekben, XIII. Magyar Aeroszol Konferencia, Pécs, Magyarország (2017)

Salma, Imre; Németh, Zoltán; Weidinger, Tamás; Blumberger, Zoltán, Légekőri nukleáció városi környezetekben, XIII. Magyar Aeroszol Konferencia, Pécs, Magyarország (2017)

Varga, Veronika; Németh, Zoltán; Salma, Imre, A nukleáció mint egyedi forrás jelentősége légekőri részecskék képződésében Budapesten, 6. Környezetkémiai Szimpózium, Bakonybél, Magyarország (2017)

Varga, Veronika; Németh, Zoltán; Salma, Imre, A diffúziós veszteség meghatározása és a légekőri nukleáció jelentősége Budapesten, XIII. Magyar Aeroszol Konferencia, Pécs, Magyarország (2017)

Summary

Ultrafine aerosol particles are in the focus of scientific interest due to their environmental and health effects since the last decade. This PD project contributed to the better understanding of the atmospheric urban new particle formation. The project was successfully realized with 4 original research articles with a cumulative impact factor of 21.0 and several international conferences.

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