FINAL SCIENTIFIC REPORT

on the NKFIH research grant K 132254 entitled

Atmospheric new aerosol particle formation and growth: the role of biogenic sources

for the period from 01–12–2019 to 30–11–2024

The major research results and conclusions obtained during the project are grouped and briefly summarised below in items 1–7 with references to the selected, most important publications of the project in the actual fields. In the last item no. 8, auxiliary but associated achievements and honours related to the research activities are listed. More detailed and further information can be found on the website of the Budapest platform for Aerosol Research and Training (BpART) Laboratory, which represents a solid and essential experimental foundation that enabled us to address the following composite scientific questions by combining new state-of-the-art experimental methods with novel evaluation and modelling studies.

1. We revealed the association between the relative occurrence frequency (f_{NPF}) of regional atmospheric new particle formation and growth (NPF) events and vegetation growth dynamics. The spring maximum of f_{NPF} often overlapped with the time intervals of positive T anomaly on vegetated territories as well. The link between the potential heat stress exerted on plants in sultry summer intervals and the summer f_{NPF} minimum could not be proved. The data set evaluated involved 1) modelled vegetation activity properties including gross primary production of biomass, leaf area index and stomatal conductance from an advanced regional biogeochemical model, 2) aerosol properties, air pollutants and meteorological data from in situ measurements at the BpART Lab, and 3) satellite-based product (NDVI index) recorded by MODIS on Terra. We also applied our coupled radiocarbon-levoglucosan marker method for source apportionment of the total carbon (TC=OC+EC, where OC and EC are organic carbon and elemental carbon in particles, respectively) into contributions from fossil fuel (FF) combustion, biomass burning (BB) and biogenic sources for a regional background environment of the Carpathian Basin, a suburban area and central part of Budapest in each season for 1 year. We showed that the biogenic sources changed radically over the seasons at all locations. Their contributions in winter were the smallest (though non-negligible); the biogenic sources and FF combustion were the largest two contributors in spring and autumn, whereas biogenic activities became the major source in summer.

   (Salma et al., Influence of vegetation on occurrence and time distributions of regional new aerosol particle formation and growth, Atmos. Chem. Phys., 21, 2861–2880, 2021; Salma et al., Fossil fuel combustion, biomass burning and biogenic sources of fine carbonaceous aerosol in the Carpathian Basin, Atmos. Chem. Phys., 20, 13767–13781, 2020.)

2. We proved that NPF phenomenon tends to occur also on the days traditionally characterized as non-event days. Using multiple-year measurement data, we were able to show that during such days, particle formation rates at 6 nm were 20% of those observed during the traditional NPF event days. The role of biogenic and anthropogenic sources was identified. This phenomenon – called as quiet NPF – contributes significantly to the production of secondary

carbonaceous particles in the atmosphere. We proposed a methodology of estimating secondary organic carbon (SOC) based on the high EC edge approach reinforced by traditional EC marker method for primary OC. The intercepts of the primary OC vs. EC were substantial above zero in spring and summer, which showed that the production of OC from additional sources than the FF combustion and BB emissions cannot be neglected. Contributions of SOC to OC was the smallest (30%) in winter, whereas the SOC prevailed in summer with a typical share above 50%.

*(*Kulmala et al., Quiet new particle formation in the atmosphere, *Front. Environ. Sci.*, 10, 912385, 2022; Salma et al., Secondary organic carbon and its contributions in different atmospheric environments of a continental region and seasons, *Atmos. Res.*, 278, 106360, 2022.)

3. We derived and interpreted droplet activation properties of urban aerosol particles and their consequences 1) on particle scavenging and fog formation, and 2) on particle deposition in the human respiratory system. The median concentrations of cloud condensation nuclei at water vapour supersaturations (*S*s) of 0.1, 0.2, 0.3, 0.5 and 1.0% were 0.59, 1.09, 1.39, 1.80 and 2.5×10^3 cm^{-3}, respectively. They represented from 7% to 27 % of the total particles. The relationships between the critical *S* and effective critical dry particle activation diameter suggested that the urban particles with a diameter larger than 130 nm showed similar hygroscopicity than the continental aerosol, while the smaller particles appeared to be less hygroscopic. These properties were related to size-dependent chemical composition and external mixtures of particles in cities. The urban aerosol was characterised by substantially smaller hygroscopicity parameters (κ values) than for regional or remote locations. Numerical simulations using a box model with a size resolving moving bin scheme showed that the aerosol particles can be washed out from the atmosphere more efficiently by Brownian motion and phoretic scavenging than by nucleation processes. The aqueous-phase chemistry also impacted the hygroscopicity of particles. The hygroscopic growth of urban-type particles with diameters of 50, 75, 110 and 145 nm determined by stochastic lung deposition mathematical model yielded decreased deposition fractions in all major parts of the healthy respiratory system. The deposited fractions were rising monotonically with particle size. The spatial distribution of the deposited particles within airway units remained highly inhomogeneous. For patients with severe chronic obstructive pulmonary disease, the hygroscopic growth in the conductive airways caused a substantial relative decrease in the deposition fractions. In contrast, the relative depositions of hygroscopic particles increased in the acinar region. These findings can have important consequences on drug administration by inhalation.

(Salma et al., Cloud activation properties of aerosol particles in a continental central European urban environment, *Atmos. Chem. Phys.*, 21, 11289–11302, 2021; Kumar et al., Numerical simulation of the micro-physics and liquid-phase chemical processes in fog using size resolving bin scheme, *Atmos. Res.*, 266, 105972, 2022; Farkas et al., Effects of hygroscopic growth of urban aerosol particles on their modelled regional and local deposition in healthy and COPD-compromised human respiratory system, *Sci. Total Environ.*, 806, 151202, 2022.)

4. We systematically determined and interpreted the oxidative potential (OP) of aerosol particles by adopting the most widely used ascorbic acid and dithiothreitol acellular assays at three locations in the Carpathian Basin. The source apportionment of aerosol constituents was

achieved by positive matrix factorisation (PMF) modelling. Biomass burning, suspended dust, road traffic, oil combustion, vehicle wear and mixed industrial/metal source were identified. The OP data sets were related to the main aerosol sources using multiple linear regression. In winter and autumn, the OP activity was dominated by BB at all sites. The effects of motor vehicles (road traffic and vehicle wear sources) and suspended dust prevailed in the non-heating period. The temporal coincidence between the most severe daily PM health limit exceedances in winter and the more toxic PM chemical composition from BB, and the spatial coincidence between the urban hotpots of particularly OP active transition metals from traffic and the high population densities indicate important consequences for more efficient air quality regulations. The OP values were related to air quality and secondary organic aerosol.

(Vörösmarty et al., Oxidative potential and its relationships in rural background, suburban and city centre environments in the Carpathian Basin, *Atmos. Chem. Phys.*, 23, 14255–14269, 2023.)

5. We performed source apportionment on size-segregated particle number concentrations (PNCs) in 27 size channels over a diameter range of 6–1000 nm augmented by air pollutants in Budapest for 11 full years. The input dataset was treated for the effect of the local meteorology by dispersion correction. Both the uncorrected and corrected datasets were evaluated using PMF in separate seasons. Six source types including nucleation, two road vehicle emission sources separated into a semi-volatile fraction and a solid core fraction, diffuse urban source, secondary inorganic aerosol, and ozone-associated secondary aerosol were identified, characterised and quantified. The dispersion correction did not considerably change the profiles and diel variations or patterns of the sources, while it substantially modified the relative shares of the nucleation source in all seasons. The mean relative contributions of the traffic emissions (of 60%) point that on-road motor vehicles were the leading source of particle numbers. The nucleation was responsible for 24% of the PNC annually as a lower estimate. It exhibited a compound character consisting of photochemically induced nucleation and traffic-related nucleation. The conditional bivariate probability function analysis showed considerable spatial variations in the source origin. The response of nucleation and road traffic semi-volatile fraction factors to COVID-19 lockdown restrictions was highly variable in European cities. Secondary aerosols exhibited extensive reductions in their mean PNCs. These variegated responses to lockdowns across Europe point to a complex network system of emission and precursor sources vs. aerosol sinks.

   (Vörösmarty et al., Attribution of aerosol particle number size distributions to main sources using a 11-year-long urban dataset, *Atmos. Chem. Phys.*, 24, 5695–5712, 2024; Rowell et al., Insights into the sources of ultrafine particle numbers at six European urban sites obtained by investigating COVID-19 lockdowns, *Atmos. Chem. Phys.*, 24, 5695–5712, 2024.)

6. We characterised and quantified the health and environmental consequences of the firework displays in Budapest on St. Stephen's Day above a larger urban territory over seven years. In the most impacted year, the time series of the measured total PNCs reached its maximum of 369×10^3 cm^{-3} in 5 min after the end of the firework, and it returned to the pre-event level within 45 min. The fireworks increased the hourly mean concentration by a factor of 5–6,

whereas the concentration in the most sensitively affected diameter range of 100–1000 nm was elevated by a factor of 20–25. A separate new size mode with a median diameter of 203 nm showed up in the size distributions. Spatiotemporal dispersion simulations revealed that there were substantial vertical and horizontal concentration gradients in the firework plume. The affected area made up a large part of the city. Not only the onsite spectators of the show and adjacent environments, but further inhabitants and more distant, larger and populous urban quarters located downwind of the displays were influenced by the plume and its fallout. The firework increased the deposition rate in the respiratory system of adults by a factor of 4. Children were more susceptible to the exposure than adults. Furthermore, we showed in general that the total number of particles, and the particle number concentrations in different size fractions are efficient quantities for air quality monitoring in urbanized areas. At low PM mass concentrations, PNCs express the aerosol health effects better than PM mass, and emphasizes the excess health risk from ultrafine particle (diameter <100 nm) exposures.

(Salma et. al., Firework smoke and its consequences on urban air quality and deposition in the human respiratory system, *Environ. Pollut.*, 320, 121612, 2023; Thén and Salma, Particle number concentration: a case study for air quality monitoring, *Atmosphere*, 13, 570, 2022.)

7. The on-line measurements by our particle mobility size spectrometer and the associated evaluations of particle number size distribution and further quantities continued all over 5 years of the research project with a daily data coverage of >95%. Our data now span over 13 full measurement years, which make them one of the longest urban nucleation data sets in the world. Other on-line measurements and samplings were accomplished for 1 year or several months. This background allowed us to actively participate in bilateral and international studies such as Research Infrastructures Services Reinforcing Air Quality Monitoring Capacities in European Urban and Industrial Areas (RI-URBANS), which resulted in several important comparative and integrating articles with a European perspective. Furthermore, Guidance Documents on Measurements and Modelling of Novel Air Quality Pollutants connected to new European Air Quality Directive were created, tested and recommended for urban areas by an international team, which we participated in.

   (Garcia-Marlès et al., Source apportionment of ultrafine particles in urban Europe, *Environ. Int.*, 194, 109149, 2024; Garcia-Marlès et al., Inter-annual trends of ultrafine particles in urban Europe, *Environ. Int.*, 185, 108510, 2024; Zhang et al., New particle formation event detection with convolutional neural networks, *Atmos. Environ.*, 327, 120487, 2024; Liu et al., Ambient air particulate lung deposited surface area levels in urban Europe, *Sci. Total Environ.*, 898, 165466, 2023; Trechera et al., Phenomenology of ultrafine particle concentrations and size distribution across urban Europe, *Environ. Int.*, 172, 107744, 2023; RI-URBAN Service Tools ST1 and ST11.)

8. The scientific contributions of the PI during the project were acknowledged by the Science Award 2020 of the Institute of Chemistry, Eötvös Loránd University, the Gold Medal for Talent Development of the Hungarian Scientific Students' Association (OTDT, 2021), the Széchenyi Prize (the highest Hungarian state distinction acknowledging outstanding scholarly achievements) from the President of Hungary (2023), and the Medal for the Environment (Ministry for Energy Affairs, 2024), and the PI has become an editor of the journal *Atmospheric Chemistry and Physics* (IF=5.2) on invitation in 2024.

Our research results and conclusions were published during the project in 21 peer-reviewed international articles (of them, 18 in D1, mostly open access journals) and in 3 papers in Hungarian journals with the acknowledgement of the NKFIH support. It is mentioned that an article published in Science (He et al., Role of iodine oxoacids in atmospheric aerosol nucleation, *Science*, 371, 589–595, 2021) was realised a joint result of our previous and present NKFIH grants (i.e., K116788 and K132254), and therefore, it is not dealt with in this final report. We presented 5 lectures and 4 posters at *European Aerosol Conferences* or *International Aerosol Conference* between 2021 and 2024. We also organised one international symposium (*Advances in Aerosol Science*) in Budapest with an invited leading lecturer from the Institute of Atmospheric Sciences and Climate, Lecce, Italy. The obtained outcomes were adopted into university courses as well; and six early-carrier researchers, PhD and master students were involved in the project. Our activities are in line with international research tendencies as demonstrated with our long-term and close international cooperations with leading aerosol research groups, e.g. with the University of Helsinki, University of Vienna, University of Ghent, Leibniz Institute for Tropospheric Research in Leipzig, University of Milan, Institute of Chemical Process Fundamentals in Prague, Institute of Atmospheric Sciences and Climate in Lecce, Grenoble Alps University, University of Birmingham, Institute of Environmental Assessment and Water Research in Barcelona, National University of Ireland in Galway, University of Eastern Finland in Kuopio, University of Tartu, Paul Scherrer Institute in Villigen, National Observatory of Athens, University of Rochester, NY, USA, and Nanjing University, China.

The achievements of the project correspond to the submitted research plan, and fulfil the expected results.

5 January 2025

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principal investigator