

Interactions between urban air and regional atmospheric nucleation

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New aerosol particle formation (NPF) and consecutive particle growth processes in the atmosphere were first identified in clean environments. Recently, NPF has been proved to be common in polluted environments including large cities as well, but the connections between the urban and regional air have remained unknown. We investigated the effect of regional NPF on urban aerosol load under well-defined atmospheric conditions. The Carpathian Basin, the largest orogenic basin in Europe, represents an excellent opportunity for exploring these interactions with its extension, moderate climate and topographically discrete character, and because it contains a large city, Budapest, at its central part. We characterized atmospheric conditions of the city centre and near-city background of Budapest by performing continuous measurements of aerosol, criteria air pollutant gases and meteorological data for two 1-year long time intervals. We completed similar measurements in the rural background at K-pusztá station representing the regional atmospheric conditions over the same time intervals.

The annual mean NPF occurrence frequencies of 24% and 28% for Budapest sites were smaller by factors of 1.3 and 1.4, respectively, than for K-pusztá station. Moreover, a continuously decreasing tendency from the rural background through the near-city background to the city centre was found. The seasonal variability of the NPF frequency was very similar at both sites with a minimum in winter and two local maxima, one in spring and the other in autumn. The spring maximum took place in April during the first year and in March during the second year. Such a shift can be caused by inter-annual differences in meteorological conditions or biogenic cycling through emissions of volatile organic compounds from vegetation. The two measurement sites seemed to respond identically to these influences. We found that NPF events at the urban site are usually delayed by >1 hour relative to the rural site or even inhibited above a critical condensational sink (CS) level. The mean values of J_6 and GR for central Budapest exceeded that for K-pusztá station by factors of 2 and 1.6, respectively. This is consistent with the idea that particles capable of escaping coagulation scavenging need to grow faster in polluted air compared to cleaner environments. Correlation analysis on the joint two-year long data set resulted in a significant relationship in the occurrence of the NPF at the 2 sites. In addition, air mass transport analysis indicated that the nucleating air masses only reached approximately one third of the geographical distance between Budapest and K-pusztá when the NPF

had already started at the downwind site. Hence, the NPF observed at the downwind site was not because of air mass advection from the upwind site, but rather took place simultaneously over large distances in the basin.

Based on long-term observations, we revealed that NPF seen in a central large city of the basin (Budapest) and its regional background occur in a consistent and spatially coherent way as result of a joint atmospheric phenomenon taking place over large horizontal scales.

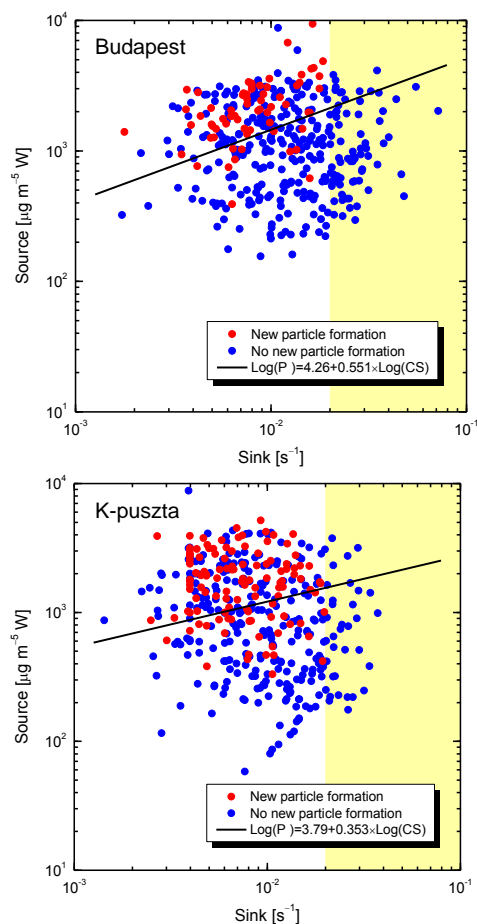


Figure 1. Major source and sink of gas-phase H_2SO_4 separately for quantifiable-event days and non-event days in Budapest and at K-pusztá station. The yellow areas indicate the CS range in which the NPF events were suppressed over the whole region.

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