Sources of ultrafine particles over Germany, an airborne survey

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Ultrafine particles in the planetary boundary layer were considered to be climate and health relevant. The climate effect is based on their ability to act as cloud condensation nuclei with an impact on cloud microphysics and rainfall temporal, spatial and intensity distribution (Junkermann *et al*, 2011). Possible health effects are suspected to be due to the high mobility and deep penetration into the lungs where chemical components bound to the aerosols can be deposited (Frank *et al*, 2011). Although a network for monitoring of ultrafine particles (GUAN) now exists for several years in Germany (Birmili *et al*, 2009) the knowledge about sources and 3D- distribution in the planetary boundary layer is still limited. Particle number concentrations and size distributions are extremely variable and most probably dependent on local to regional scale transport and vertical mixing of the planetary boundary layer. Thus ground based stations are often affected by local pollution. Sources are assumed to be located in the planetary boundary layer as well as in elevated layers of the lower atmosphere. Anthropogenic as well as biogenic emissions are discussed as contributions to formation and growth from nm sizes towards size ranges important for aerosol cloud interactions (Kulmala *et al*, 2013) Although sulphur compounds seem to be important for the production of ultrafine particles the significant reduction of sulphur emissions in Germany since 1990 did not lead to a concurrent reduction of ultrafine particle concentrations (Hamed *et al*, 2010). In contrast, the number concentrations of these particles increased, indicating that anthropogenic emission patterns were changing. Also important but not well established is the ratio of primary emission to secondary production in the atmosphere from gas to particle conversion.

Source apportionment and source characterization are rather difficult from ground based field sites although the size distribution at least allows a rough estimate of the age of the particles within the first hours of growth. Too many different processes have to be considered. Thus within this project airborne measurements were performed using small aircraft flying low and slow to characterize ultrafine particle number concentrations all over Germany from the Bavarian Alps towards the Northern Sea, to characterize 'typical' background levels and to identify the major sources and to estimate their source strength and possible contribution to the overall ultrafine particle budget. Airborne measurements have the advantage that local pollution from small emitters is normally not disturbing the investigations, and within a short time both the planetary boundary layer and the lower free troposphere can be probed. Also larger particle sources are directly visible from plume passages and their spatial extend (Bigg, *et al,*1978).

Background as well as maximum concentrations and size distributions varied widely over Germany depending on the origin and (pollution) history of the air masses encountered. Dominating single sources were also identified emitting primary ultrafine particles which were generally related to burning and processing of sulphur containing fossil fuel, highlighting the major role of sulphur compounds for the generation of ultrafine particulate matter. These large sources are able to affect the planetary boundary layer particle size and number distributions over regional scale distances up to more than 200 km.

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