Preface to topical collection on new particle formation in Atlanta

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[1] The Aerosol Nucleation and Real Time Characterization Experiment (ANARChE) took place in Atlanta, Georgia, during late July and through August 2002. The study was aimed at providing new insights into several discrepancies that arose from our previous observations on atmospheric new particle formation. These include (1) rates of new particle formation near the Earth’s surface are often orders of magnitude higher than are predicted by the binary theory of nucleation for sulfuric acid and water; (2) new particle formation rates often vary in proportion to the first or second power of sulfuric acid concentration, rather than the much stronger dependence predicted by nucleation theories; and (3) the growth rates of freshly nucleated particles are typically two to ten times faster than can be explained by the condensation of sulfuric acid and its associated water and ammonia. Established methods were used to measure sulfuric acid vapor concentration and particle size distributions down to 3 nm. Ammonia concentrations were measured with high time resolution by chemical ionization mass spectrometry to explore ammonia’s role in particle production. Two new instruments were developed to enable measurements aimed at explaining the above observations. These include the thermal desorption chemical ionization mass spectrometer (TDCIMS) for measuring the composition of nanoparticles down to 7 nm and the nanometer tandem differential mobility analyzer (Nano TDMA) for measuring the volatility and hygroscopicity of 4–10 nm particles. Measurements by the latter instruments allowed the primary growth species to be identified, and therefore a direct comparison could be made between precursor concentration and growth rates.

[2] ANARChE measurements were carried out at the Jefferson Street site located about 4 km northwest of downtown Atlanta [Solomon et al., 2003]. We chose this site because continuous measurements of size distributions from August 1998 through August 2000 showed that new particle formation often occurs there, especially in late summer; that the concentrations of new sub-10 nm particles produced can be extremely high; and that new particle formation is associated with elevated levels of SO2 [Woo et al., 2001; McMurry and Woo, 2002]. Thus this study gave us the opportunity to investigate new particle formation in a polluted environment where SO2 is likely a major precursor. Also, measurements of meteorological parameters and gas and particle concentrations that are carried out routinely at the Jefferson Street site as a part of the SEARCH and ARIES programs were made available to us.

[3] Our results show that the collision-controlled theory for nucleation in a chemically reacting system [McMurry, 1983] (also referred to as “kinetic” or “barrierless” nucleation) provides an upper limit to the number of particles produced. However, the number of particles observed is typically less than the number predicted by this theory, by factors that range from about ten to one thousand. We found some evidence for an association between ammonia and the number of particles produced, but ammonia apparently did not account for much of the observed variability. In addition, three separate observations (direct measurements of composition, measurements of nanoparticle hygroscopicity and volatility, and comparison of measured and predicted growth rates) support the conclusion that sub-40 nm particles that were recently formed by nucleation consisted primarily of sulfates neutralized to some extent by ammonia. However, growth rates of particles larger than 40 nm were about a factor of five greater than could be explained by the condensation of sulfuric acid. We did not measure the composition of nucleated particles as they grew past 40 nm during this study, but such measurements are clearly needed.

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