Chapter 1

Introduction

Introduction

Aerosol particles have gained much attention in recent years due to their role in reducing visibility, cloud formation and lifetime, climate change, and health related issues. Most of these effects are driven by physical and chemical properties of the aerosols, for example, aerosol size, concentration, and chemical composition. These factors vary from region to region depending on the sources of the aerosols. For example, sea-salt and dust particles dominate the coarse mode aerosols, whereas sulfate and organic particles dominate the sub-micron aerosols. Furthermore, particulate mass of fine aerosols is dominated by sulfate particles in the eastern U.S. and by organic and nitrate particles in the western U.S. (*1*). Absorption and condensation of different gas-phase species onto the aerosols as they are transported away from their source also affect their physical and chemical properties by changing their size and composition. Oxidation of inorganic gas-phase pollutants such as sulfur dioxide (SO_2) and nitrogen oxides (NO_x), which are dominantly emitted by coal-fired power plants and fossil fuel combustion in vehicles, leads to partitioning of their low volatility oxidation products into the particle phase. Furthermore, when gas phase organic species are oxidized in the atmosphere by ozone, hydroxyl, or nitrate radicals, they often form products that have low vapor pressure and tend to nucleate to form new particles or condense on pre-existing aerosols. These secondary organic aerosols (SOA) are a significant contributor to the total ambient organic aerosol loading in urban areas as well as regionally and globally. Traditionally, controlled laboratory chamber experiments have been carried out to determine the SOA forming potential, i.e., organic yield, of volatile hydrocarbons that are, or have a chemical structure close to, atmospherically relevant compounds and to identify specific compounds formed as SOA.

A good understanding of atmospheric processes and the corresponding effects on aerosol properties is essential to better understand the importance of ambient aerosols and to incorporate the results into future regulations. However, our understanding of these properties is limited by the capabilities of instruments for aerosol measurements. Although instruments capable of real-time aerosol size distribution measurements have been available for more than a decade, the ability to obtain chemical composition information with high time resolution has emerged only in the last few years. The simultaneous size and chemical composition measurements of non-refractory components of submicron aerosols provided by the Aerodyne Aerosol Mass Spectrometer (AMS) make it a powerful instrument in probing aerosol properties in real-time with high time resolution.

Here, in Chapters 2-3, results of laboratory chamber experiments of particle formation from photooxidation of diiodomethane-- an organic iodine compound emitted from algae at the coastal areas-- are presented. The measurements included number, size, chemical composition, and hygroscopicity measurements of the aerosols formed using Condensation Particle Counters (CPCs), Differential Mobility Analyzers (DMA), the AMS, and a tandem DMA. Furthermore, a kinetic model was developed to help understand the chemical mechanism leading to particle formation.

Chapter 4 summarizes the results of the AMS measurements made during the ACE-Asia (2001) field campaign-- the first aircraft deployment of the instrument. The goal of this study was to characterize the properties of aerosols present in the eastern Asian outflow. In Chapter 5, AMS observations are combined with other aerosol measurements made during the CRYSTAL-FACE (2002) field campaign to investigate a study of the aerosol-cloud closure.

Chapters 6-10 are devoted to laboratory chamber studies of SOA formation from oxidation of cycloalkenes, terpenes, and *m*-xylene. Results of measured SOA yield of various cycloalkenes and the effect of different hydroxyl radical scavengers on the yield are summarized in Chapters 6-7. Chapters 8-9 describe the results of chemical analysis of bulk filter samples collected during the oxidation experiments of cycloalkenes and α-pinene. Chapter 10 summarizes specifically the AMS observations made during oxidation experiments of cycloalkenes, terpenes, and *m*-xylene. Supplementary material for Chapters 7-9 are presented in Appendices A-C (Chapters 11-13). Finally, Chapter 14 provides a summary of the thesis.

Reference:

(1) Malm, W. C.; Schichtel, B. A.; Pitchford, M. L.; Ashbaugh, L. L.; Eldred, R. A. Spatial and monthly trends in speciated fine particle concentration in the United States. *J. Geophys. Res.*, 2004, *109*, D03306, doi: 03310.01029/02003JD003739.