STUDIES with the AEROSOL MASS SPECTROMETER

Thesis by

Roya Bahreini

In Partial Fulfillment of the Requirements for the

Degree of Doctor of Philosophy

California Institute of Technology

Pasadena, California

2005

(Defended February 1, 2005)

Acknowledgements

Looking back at the years I spent at Caltech for my graduate studies, I feel privileged that I got the chance to meet and interact with many individuals whom I greatly respect for their scientific insights, and I am thankful for their invaluable support and friendship.

First, I thank Profs. John Seinfeld and Rick Flagan. I could not have completed this path without their continuous support, scientific insights, and patience throughout the years. They always kept me motivated and provided me with productive and useful advice when I needed guidance.

I have also interacted to a great extent with many people at Aerodyne Research, Inc. who have been involved with the Aerosol Mass Spectrometer. I am especially grateful to Doug Worsnop and John Jayne for their generous support and valuable technical and scientific insights and advice throughout the years.

My parents, sister, and uncle have always been a great support in my life, and the past few years at Caltech have not been an exception. Although we were usually hundreds of miles apart, I am very grateful for their continuous support and encouragement.

I have been very fortunate to be working in such a friendly and nice environment at Caltech, and I owe this to all the past and present members of Seinfeld/Flagan research group, especially the “Roofians”- Jose, David, Tomtor, Melita, Sally, Jesse, Dara, Song, and Jason. They not only made the experience in the lab pleasant, but also kept me good company when I needed to do something aside from being in the lab. I will always remember our mountain-watchings, endless afternoon snackings, and “eating-outings”!

During my Ph.D. study, I had the opportunity to participate in several field campaigns. I would like to thank the CIRPAS crew, especially Haf Jonsson, Roy Woods,

Nava Roy, Mike Hubble, and Reggie Burch, for making this experience as smooth as possible and for their support during the field studies. I share many good memories from these campaigns, thanks especially to Tomtor, Tracey, and Bill.

Although I have not worked as closely with Serena, Tim, Thanos and the rest of the members of our research group, I greatly value their friendship and appreciate their support during these years.

Many thanks to Ann Hilgenfeldt who has been so helpful and patient in accommodating me with what was needed for research in the lab and field projects. I would also like to thank Fran Matzen, Linda Scott, and Elizabeth Hinojos for their great administrative work. Thanks to Mike Vondrus and Suresh Guptha who always provided me their excellent expertise on usually rushed machining requests and panicking computer-related issues.

Finally, throughout my years at Caltech, I was lucky to meet my three very good friends—Mariu, Maca, and Pilar. Their support and friendship played a big role in making it easier for me to deal with the different issues throughout these years, and I am grateful for that. Thanks also to other friends that I met while at Caltech who kept me outside the lab when I needed a break. I enjoyed our gatherings, outdoor activities, parties, salsa classes, etc. to a great extent and have many good memories.

Abstract

Fast time-response of the Aerodyne Aerosol Mass Spectrometer (AMS) makes it a well-suited instrument for ambient field measurements. On the other hand, laboratory chamber experiments provide the opportunity to study a specific system in a more controlled environment. The goal of this thesis is to provide a summary of laboratory and field measurements using the AMS.

During laboratory chamber photooxidation experiments of diiodomethane (CH_2I_2), particle nucleation was observed at CH_2I_2 concentrations down to 15 ppt, which is comparable to the total gas-phase iodine species measured at coastal areas. Iodine oxides and oxyacids were observed in the aerosol mass spectra obtained by the AMS, consistent with the known gas-phase chemistry.

Airborne measurements by the AMS during the ACE-Asia field study revealed that the non-refractory submicron aerosols in the pollution layers of the boundary layer up to 3700 m were mainly composed of sulfate, ammonium, and organics. These pollution plumes originated primarily from urban and industrial areas of China and Korea.

The laboratory chamber experiments of oxidation of cycloalkenes, terpenes, and *m*-xylene provided the opportunity to study the Secondary Organic Aerosol (SOA) forming potential, i.e., yield, and determine SOA effective density and chemical composition. Evidence of acid-catalyzed heterogeneous chemistry in the ozonolysis of α-pinene was observed since the total AMS organic mass during the experiments with acidic seed particles had a greater contribution of higher molecular weight fragments. The mixtures of SOA compounds produced from similar precursors studied resulted in broadly similar

AMS mass spectra. Thus, fragmentation patterns observed for biogenic vs. anthropogenic SOA can be potentially useful in determining the sources of ambient SOA.

Table of Contents

List of Tables

List of Figures