Insights Into Atmospheric Organic Aerosols Using An Aerosol Mass Spectrometer

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No portion of the work referred to in this thesis has been submitted in support of an application for another degree or qualification of this or any other university, or other institution of learning.

To my family, and to the memory of Ghadeer

Abstract

Atmospheric aerosols have significant effects on issues such as human health, air quality and global climate. At present, the radiative effects of aerosols represent the largest uncertainties in quantifying climate forcing due to man-made changes in the composition of the atmosphere. A significant fraction of atmospheric aerosols consists of organic substances. A clear understanding of the chemical composition and processes that control the formation and transformation of organic aerosols in the atmosphere has been limited by the measurement techniques employed to study them. Over the past decade, aerosol mass spectrometry has gained considerable interest as a means of correlating the size and chemical composition of aerosol particles in real-time. The Aerodyne aerosol mass spectrometer (AMS) has represented a significant advance in this area of research. The AMS is capable of providing quantitative information on the chemical composition of the submicron non-refractory fraction of aerosol particles with a high size and time resolution. The instrument employs an aerodynamic lens, differential pumping, aerodynamic sizing, thermal vaporisation, electron impact ionisation and quadrupole mass spectrometry, where particles are analysed within a few milliseconds of entering the vacuum. Work to characterise and validate the ability of this instrument to measure the organic fraction of the aerosol is presented in this thesis. The utilisation of the AMS to characterise the chemical and physical properties of the organic fraction of atmospheric aerosols in various locations around the world representing urban, rural and remote settings is discussed. Results show that organics represent a major fraction of the measured aerosol mass at all of these locations. Organics show a distinct bimodal distribution at urban locations, where combustion sources are responsible for the emission of hydrocarbon-dominated particles in the small mode. On the other hand, highly oxidised organic compounds dominate the accumulation mode at all locations, including the urban ones. The chemical and physical characteristics of secondary organic aerosols formed from the photooxidation of anthropogenic and biogenic precursors in a reaction chamber are also studied. The results indicate that photooxidation of the biogenic precursor explains more of the chemical signature of organic particulate observed at the various locations compared to the anthropogenic precursor. Finally, the role of atmospheric aerosols, particularly its organic fraction, in the activation of cloud droplets is investigated.

Content

CHAPTER ONE: Atmospheric Aerosols

CHAPTER TWO: Aerosol Measurements

CHAPTER THREE: The Aerodyne Aerosol Mass Spectrometer (AMS)

CHAPTER EIGHT: Conclusions and Future Work