CHAPTER 2

BACKGROUND

# 2.1 Introduction

Stratospheric aerosol plays an important role in the global radiative forcing balance by scattering solar irradiation and causing an overall cooling effect that depends on the particle size distribution and the concentration (*Kiehl and Briegleb*, 1993; *Stocker et al.*, 2013). These climate effects are an important and recent focus of research due to the potential contribution of stratospheric aerosol to the so-called global warming hiatus (*Solomon et al.*, 2011; *Haywood et al.*, 2014; *Fyfe et al.*, 2013) and efforts to quantify the variability and trends in the global stratospheric aerosol load are underway with various ground-based and satellite data sets (*Rieger et al.*, 2015; *Ridley et al.*, 2014).

Since its discovery with stratospheric balloon observations (*Junge et al.*, 1961), stratospheric aerosol has been measured with various techniques, although due to the variability of physical composition and particle size, the observations are always limited to some degree. In-situ balloon observations continue to be used and have provided highly valuable data sets, including most notably the long time series of optical particle counter measurements from Laramie, WY (*Deshler et al.*, 2003; 2008; *Kovilakam et al.*, 2015). Aircraft-borne nephelometers (*Beuttell and Brewer*, 1949; *Charlson et al.*, 1969) acquire detailed in-situ measurements, providing, for example, plume composition (*Murphy et al.*, 2014), but are spatially limited to the aircraft track. Ground based lidars have been used to do detailed studies of the extent of volcanic aerosol plumes (*Chazette et al.*, 1995; *Sawamura et al.*, 2012) and provide valuable insight into long term local variability and trends in the aerosol layer. For example, lidar observations were used by *Hofmann et al.* (2009) to first report the observed increase in stratospheric aerosol over approximately the last decade. However, the global distribution, which can only really be obtained with satellite observations, provides invaluable insight into aerosol processes and variability. A good example of this is the use of satellite observations by *Vernier et al.* (2011b) to determine that the increased stratospheric aerosol load reported by *Hofmann et al.* (2009) was in fact due to a series of relatively minor, mostly tropical, volcanic eruptions.

Several recent studies have highlighted the requirement for continued global stratospheric aerosol observations and especially the need to resolve, both vertically and horizontally, aerosol in the lowermost stratosphere and the upper troposphere. This is the case for tracking the evolution of aerosol from volcanic eruptions, which can have a substantial effect on the aerosol optical depth in the lowermost stratosphere (*Ridley et al.*, 2014; *Andersson et al.*, 2015). Furthering the understanding of the transport of aerosol near and across the tropopause would also benefit from higher spatial and temporal resolution observations. This is evident in the case of volcanic plumes, such as that from Nabro in 2011, the transport and origin of which has been studied extensively and somewhat controversially (*Bourassa et al.*, 2012c; 2013; *Vernier et al.*, 2013; *Fromm et al.*, 2013; 2014; *Fairlie et al.*, 2014; *Clarisse et al.*, 2014). However, this is also the case for the formation of background-level aerosol, particularly in the region of the Asian and North American monsoons, which have been identified as a source of substantial, seasonal and highly structured aerosol formation from precursor tropospheric source gases (*Vernier et al.*, 2011a; *Neely et al.*, 2014; *Thomason and Vernier*, 2013). .

Continued stratospheric aerosol observations from space are drastically needed though few, if any, planned missions with such capability are underway. In this work, we present the design and test of a prototype instrument for potential future satellite-based stratospheric aerosol observation. The Aerosol Limb Imager (ALI) concept is a relatively small, low-cost, low-power, passive instrument, suitable for microsatellite deployment with the capability to provide high spatial resolution measurements, both vertically and horizontally, of the visible/NIR aerosol extinction coefficient. The basic idea is to leverage the clear advantages of the limb scatter technique as a passive, and therefore low mass and low power, means to obtain daily global coverage, with a two dimensional hyperspectral imager for filling cross-track observation.

The ALI instrument concept is built around the use of an Acousto-Optic Tunable Filter (AOTF), which is a novel filtering technology that provides the ability to rapidly select the central wavelength of an image with no moving parts. These filters, which have recently been developed as large aperture imaging quality devices, operate very efficiently in the red and near infrared spectral range, which is a well matched spectral range for limb scatter sensitivity to aerosol and cloud (*Rieger et al.*, 2014). Additionally, the spectral bandpass of the AOTF, which is typically between 3-6 nm at these wavelengths, is very suitable for the broadband scattering characteristics of the aerosol limb signal. The two dimensional imaging nature of the design provides the capability to achieve at least sub-kilometer resolution at the tangent point, which is on the order of the scale size of the upper troposphere and lower stratosphere (UTLS) aerosol features mentioned above.

It should be noted that the basic instrument design concept of ALI is very similar to that of the Atmospheric Limb Tracker for the Investigation of the Upcoming Stratosphere (ALTIUS) (*Dekemper et al.*, 2012), which is a Belgian instrument concept from the Belgian Institute for Space Aeronomy (BIRA). ALTIUS is designed to measure limb scattered sunlight; however, it also has solar, stellar, and planetary occultation modes and is scientifically focused on trace gas measurements, particularly for ozone, whereas ALI is optimized for aerosol observation.

# 2.2 Stratospheric Aerosol

The atmosphere of earth is a complex and complicated system and is effected by human activities. In the late 18th century, it was known that atmospheric temperature decreased with altitude and a theory had been raised that at specific certain altitudes the temperature must eventually go to absolute zero. This lead to a series of balloon campaigns to discover this mysterious altitude in the atmosphere which were noisy and unreliable due to the technology available. However, in the late 19th century the technology used in these sounding balloon had improved to a point where the atmospheric temperature could be accurately measured and it was found that at approximately 12 km an inversion point was discovered where the temperature started to increase and thus the tropopause, which separates troposphere and the stratosphere, was discovered (*Hoinka*, 1997). The stratosphere is the region of the atmosphere above the temperature inversion of the troposphere, where atmospheric temperature increases and the lower bound of the stratosphere is in-between 10 and 16 km from the high latitude to the tropics (*Andrews*, 1987). The stratosphere, which extends up to approximately 50 km, is thermodynamically stable and fairly dry (*Boucher*, 2015). The characteristic stability of the region limits vertical transport of the stratosphere, leading to long lifetimes, spanning from months to years, for non-volatile species (*Volk et al,* 1997; *Brasseur and Solomon*, 2005)

The stratosphere undergoes exchange of air with the troposphere though a series of dynamical processes including tropical convection, the polar vortex, and tropopause folding (*Holton et al.,* 1995) and circulation within the stratosphere is dominated by the slow Brewer-Dobson circulation (*Plumb and Eluszkiewicz*, 1999). Some chemicals can cross the tropopause thermal barrier into the stratosphere which allows for chemicals reactions to occur. One such reaction forms stratospheric sulfate aerosol, discovered by *Junge et al* (1961) though stratospheric balloon sondes, which are droplets of hydrated sulfuric acid (H­2­SO­4) formed from the oxidation of sulfate compounds, primary OCS and SO2 (*Brock et al.*, 1995). This stable layer of aerosol exists in the stratosphere from the altitude of the tropopause to approximately 30 km.

## 2.2.1 Aerosol Sources and Microphysics

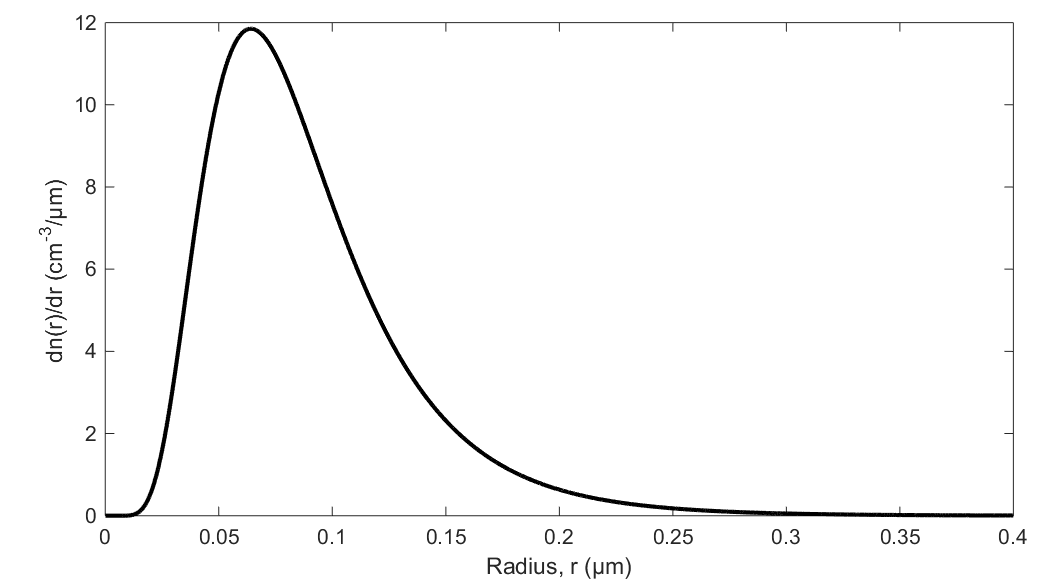
The sources for sulfate aerosol are primarily produced in the troposphere and formed though both natural and anthropogenic processes. One primary source of this sulfur is OCS which originates from marine processes, biomass burnings, and industry (*Kettle et al.*, 2002; *Notholt et al.*, 2003). OCS has a long lifetimes in the troposphere and low solubility allowing for significant portion of to reach there stratosphere and there some of it oxidizes and hydrates to form sulfate aerosol (H2SO4) and form add to the background aerosol layer (*Crutzen*, 1976).

Another source of sulfur is SO2 and one source originates in the troposphere though industry form the burning of fossil fuels. Sulfur dioxide (SO2) has a short lifetime and its concentration various regionally (*Thomason and Peter*, 2006) which also hydrates into sulfate aerosol. A second source of SO2 is from volcanic eruptions which is highly variable in location and time and can insert a large amount of sulfur directly into the atmosphere. The sulfur amounts that can be injected by volcanos can be so large they in fact dominate the stratospheric aerosol layer perturbing from the background levels. Examples of this perturbation of the aerosol layer were noted during the volcanic eruptions of El Chichon in 1982 (12-20 Tg of sulfur) (*McCormick and Swissler*, 1983; *Hofmann and Rosen*, 1983) or Mount Pinatubo in 1991 (20-30 Tg of sulfur) (*McCormick and Veiga*, 1992). However, after the Mount Pinatubo eruption a volcanically quiescent period occurred where aerosol layers returned to background. Following a series of smaller volcanic eruptions have increased the background aerosol layer in the amount of 4-7% per year from 2000 to 2009 (*Vernier et al.*, 2011).

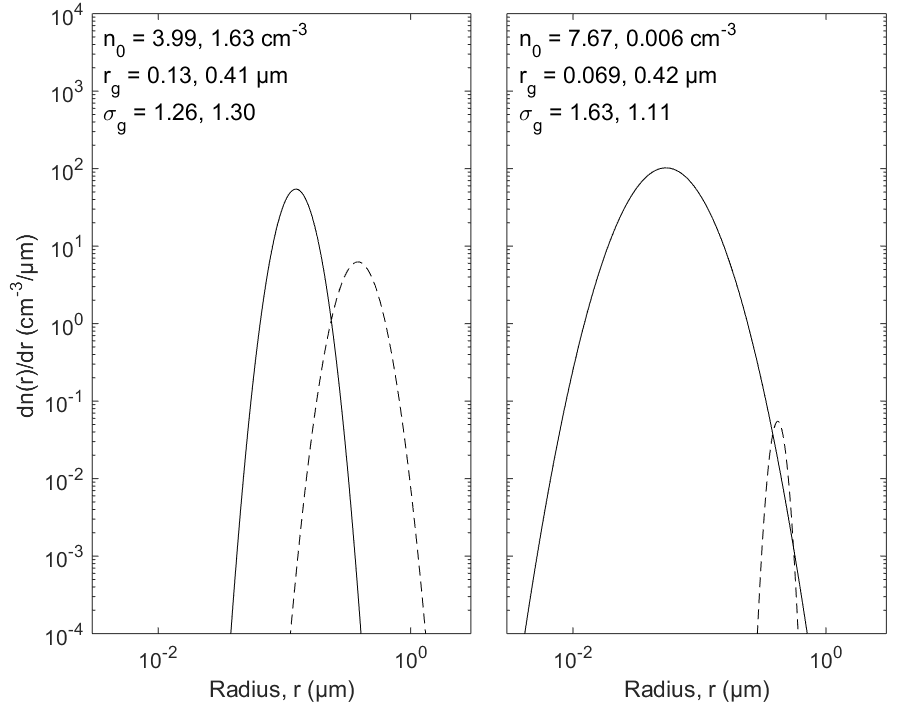
These sulfur sources undergo a series of reactions to be conversion into H2SO­4 and have been found to form droplets of aerosol that consists of approximately 25% H2O and 75% H2SO4 (*Rossen*, 1971; Wang *et al.*, 1989). These aerosol particles form droplets of various sizes that undergo that grow to various sizes on the order of 0.05 to 1.0 µm depending on the processes of nucleation, evaporation and condensation that is underwent (*Junge et al.* 1961; *Brock et al.*, 1995; *Bingen et al.*, 2004). A log-normal distribution generally describes the particle size of aerosol in the form of

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|  | (2.1) |

where is the aerosol number density, is the mode radius, and is the mode width (*Jäger and Hofmann,* 1991; *Hamill et al.* 1997). The particles are distributed in a normally over the logarithm of the radius. To fully describe aerosol for a single log-normal distribution the number density, mode radius and mode width is required and for a non-volcanic background aerosol layer an approximate mode radius and mode width of 0.08 µm and 1.6 respectively have been determined (*Thomason*, 1991) shown in Figure 2-1.



**Figure 2-1**: Log-normal distribution for non-volcanic background aerosol layer.



**Figure 2-2**: Bimodal particle size distributions fits from optical particle size. a) Distributions from a volcanic period after the Mount Pinatubo eruption recorded in 1993. b) Distributions from a background aerosol period eruption recorded in 1999. Both of the aerosol distribution measurement are from 20 km altitude with the solid line being the fine mode and the dashed line is the coarse mode. Figure is recreated from figure 5 of *Deshler et al.* (2003).

Optical particle counter have been used on board Laramie, Wyoming over the past 40 years to measure particle sizes in bin in between 0.15 to 2.0 µm. The composition of the particle size distributions have been primarily unimodal, known as a fine mode, over non-volcanic periods and can be used as an acceptable distribution approximation during background periods. But during volcanic episodes a bimodal log-normal distribution of aerosol particles is more representative of the stratospheric aerosols with the addition of a coarse mode (*Deshler et al.*, 2003; 2008; *Kovilakam et al.*, 2015). The coarse mode has larger particles than the fine mode and complicates determining full aerosol microphysical parameters since the number of required has increased to six: a number density for both the fine and coarse mode, two mode radii, and two mode widths. Figure 5 from *Deshler et al.* (2003), recreated in Figure 2-2, demonstrated two bimodal particle size distributions from balloon optical particle counters. The first distribution is from a volcanic period in 1993 after the Mount Pinatubo eruption and from a background period in 1999.

## 2.2.2 Climate Effects

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# 2.3 Aerosol Measurements

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## 2.3.1 Ground based and in-situ

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## 2.3.2 Occultation

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## 2.3.3 Lidar

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## 2.3.4 Limb Scatter

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# 2.4 Radiative Transfer

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## 2.4.1 Scalar Radiative Transfer

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## 2.4.2 Vector Radiative Transfer

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## 2.4.3 Rayleigh Scattering

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## 2.4.4 Mie Scattering

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# 2.5 ALI Prototype and Stratospheric Balloon Flight

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