Exploring the Variability of Stratospheric Aerosol

Ryan Reynolds Neely III
University of Colorado at Boulder, rrmiii@me.com

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EXPLORING THE VARIABILITY OF STRATOSPHERIC AEROSOL

by

RYAN REYNOLDS NEELY III

B.A., North Carolina State University, 2008

M.S., University of Colorado, 2010

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This thesis entitled:
Exploring the Variability of Stratospheric Aerosol
written by Ryan Reynolds Neely III
has been approved for the Department of Atmospheric and Oceanic Sciences.

Prof. Jeffrey P. Thayer, PhD, Chair

Prof. O. Brian Toon, PhD

Prof. Susan Solomon, PhD

R. Michael Hardesty, PhD

Karen Rosenlof, PhD

Prof. Cora Randall, PhD

Date

The final copy of this thesis has been examined by the signatories, and we find that both the content and the form meet acceptable presentation standards of scholarly work in the above mentioned discipline.
Neely III, Ryan Reynolds (Ph.D., Atmospheric and Oceanic Sciences)

Exploring the Variability of Stratospheric Aerosol

Thesis directed by Professor Jeffrey P. Thayer

The variability in stratospheric aerosol from 2000 to 2010 is described. This period is unique because it is the first opportunity to observe decadal variability of stratospheric aerosol unperturbed by colossal volcanic eruptions. Analyses of observations from lidars located in Lauder, NZ, Mauna Loa, HI and Boulder, CO show an increase in aerosol above 20 km that is modulated by a strong annual cycle with a wintertime peak; but observations alone are unable to conclusively partition the source of this increase between volcanic and anthropogenic emissions. Thus, a version of the Whole Atmosphere Community Climate Model coupled to the Community Aerosol and Radiation Model for Atmospheres, structured for sulfate aerosol and meteoritic smoke, is used to help attribute the sources of variability.

Comparisons of baseline simulations, using anthropogenic emissions of SO$_2$ and a boundary layer of OCS from 2000, with observations show agreement above 20 km. The comparison also reveals a bias in lidar retrievals that are due to the presence of meteoritic smoke. The bias results from the assumption that the region around 35 km is aerosol free in order to estimate the molecular component of the total observed backscatter. Meteoritic smoke is identified as a major source of extinction above 30 km and needs to be considered in the scattering properties for lidar retrievals.

To explore the decadal trends, increases of SO$_2$ from known anthropogenic sources and volcanic eruptions were added to the model. Comparisons of simulation results and observations suggest that from 2000 to 2010, above 20 km, volcanic injections are the principle driver of
stratospheric aerosol variability. Trends in anthropogenic emissions are found only to increase stratospheric aerosol annually in the late summer and fall in the northern hemisphere in the region near the tropopause.
Dedication

For my best friend and wife, Emily.
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CHAPTER I: INTRODUCTION

1.1 Motivation

The stratosphere has not been perturbed by a large volcanic eruption since the Pinatubo volcanic eruption in June of 1991 (Hofmann et al., 2009; Solomon et al., 2011). Thus, 2000 to 2010 is an idle volcanic period that yields a unique window to study the “background” variability of the stratospheric aerosol layer. As shown in Figure 1, the radiative forcing of stratospheric aerosol from 2000 to 2010 is not negligible, as previously realized in many climate model simulations, and must be taken into account when examining the variability of the atmosphere (Sato et al., 1993; Ammann et al., 2003; Solomon et al., 2011).

The observations from which the variability in radiative forcing from stratospheric aerosol was initially derived came from ground-based lidars located at Lauder, NZ, Mauna Loa, HI and Boulder, CO. These instruments observed an increase of 4-6% per year in the stratospheric aerosol volume backscatter coefficient between 2000 and 2010 (Hofmann et al., 2009; Nagai et al., 2010). Combined satellite datasets of aerosol extinction have confirmed these trends (Vernier et al., 2011a). These observations present many questions about the mechanisms controlling the stratospheric aerosol layer and how moderate volcanic eruptions, anthropogenic emissions, and changes in dynamics may be impacting the layer over time.
Figure 1: Total radiative forcing for various amounts of stratospheric aerosol. Forcings scenarios include: no stratospheric aerosol forcing (solid blue-green line), background aerosol excluding volcanic eruptions (dash-dotted green line), stratospheric aerosol from the GISS optical depth record transitioning to negligible levels of stratospheric aerosol forcing after 2000 (black line), forcing from GISS until 1998 plus aerosol loading inferred from the global satellite optical depths until 2010 (black followed by blue line), three future projections starting from satellite record (dashed purple lines). Adapted from Solomon et al. (2011).

1.2 Questions

The goal of this research is to delineate the mechanisms behind the increases in stratospheric aerosol that occurred from 2000 to 2010 as observed by ground-based lidars. Of the possible causes, the two that will be examined here are 1) small periodic volcanic emissions of aerosol to the stratosphere and 2) increases in anthropogenic sulfur gas emissions in the troposhere (Hofmann et al., 2009; Niwano et al., 2009; Vernier et al., 20011a and b). Previous analysis suggested that a large increase in coal burning since 2002, primarily in China and India, is the probable source of sulfur dioxide that ultimately ends up as the sulfate aerosol responsible for the increased volume backscatter coefficient in the stratospheric aerosol layer (Hofmann et al., 2009). This idea was also supported by the work of Randel et al. (2010) that examined the transport of pollution into the stratosphere driven by convection in the Asian monsoon. Satellite observations have also shown the existence of an Asian tropopause aerosol layer (Vernier et al., 2009; 2011a). More recently, analysis of satellite data showed that small to moderate volcanic
eruptions (1 to 2 orders of magnitude smaller than Pinatubo) coincided with short-term peaks in aerosol extinction (Vernier et al., 2011b). Unfortunately, as will be shown, lidar volume backscatter coefficient observations and other measurements of extinction cannot separate the sources of stratospheric aerosol and therefore cannot attribute proportion of the observed increase to sources. Thus, for this thesis, the main questions to be addressed are as follows:

1. What do lidar observations from 2000 to 2010 reveal about stratospheric aerosol variability?
2. What role do small volcanic eruptions and anthropogenic emissions have in the observed variability in the stratospheric aerosol record during the last ten years?

It is hypothesized that the long-term trends are a combination of both anthropogenic and volcanic emission, but that volcanoes are the major contributor and have a larger impact on short-term variability. It should be also noted that both sources of aerosol are heavily modulated by atmospheric dynamics as indicated by the role of the quasi-biennial oscillation (QBO) (Hofmann et al., 2009; Niwano et al., 2009; Randel et al., 2010; Sunilkumar et al., 2011).

In the attempt to answer these questions, many other questions arose about the annual cycles of stratospheric aerosol (a large component of variability in the lidar observations), the sensitivity of the aerosol layer to the variations in volcanic injections of aerosol, and details of how lidars observe stratospheric aerosol. Most of these concerns stemmed from trying to understand the details of the variability in the lidar observation record and are, therefore, included throughout the discussion of the observation analysis and the comparison of lidar observations to model output. Specifically, these questions included: 1) Do annual cycles in stratospheric aerosol exist or are the observed cycles due to biases in lidar volume backscatter coefficient retrievals? 2) How do errors in lidar retrievals manifest themselves in the
observational record? 3) Can annual cycles be observed when the stratospheric aerosol layer is perturbed by small volcanic injections or anthropogenic emission? 4) How do the location, height, magnitude and timing (in relation to the phase of the stratospheric oscillation) of small volcanic eruptions impact their ability to modulate the stratospheric aerosol layer?

1.3 Organization

This work has been a synthesis of observation and modeling (Figure 2). The observations initially drove the science questions being examined here while the modeling allowed for numerical experimentation that illuminated the possible sources of the variability. Through the course of this work I have taken many lidar observations and helped develop a global stratospheric aerosol microphysical model for use in these studies. Through this fusion of retrieval and analysis, both the lidar observations and aerosol modeling have improved. As such, this thesis is divided into two main topics: observations and modeling. Chapter 2 discusses lidar observations of stratospheric aerosol. Chapters 3 and 4 discuss the modeling conducted to further the understanding of these observations. Chapter 3 contains a description of the model used and validation of the model output with observations. Chapter 4 discusses the use of this model to explore the variability of the stratospheric aerosol layer by examining the steady state of the aerosol layer and the impact of various perturbations compared to the baseline layer. Chapter 5 provides a summary and points out the major findings of this work.

Figure 2: Intersection of observation and modeling.
Chapter 2 first discusses the relevant methodology of observing the stratospheric aerosol layer with ground-based lidars. Much of the information presented in Chapter 2 was acquired practically through the development of a water vapor profiling channel at the Arctic Lidar Technology (ARCLITE) facility and with the Cloud, Aerosol and Polarization Lidar (CAPABL) (Thayer et al., 1997; Neely and Thayer, 2011; Neely et al., 2012). A description of CAPABL and an example of its observations may be found in Appendix A. The details of the work on developing a water vapor channel in ARCLITE may be found in Appendix B, which is based on the work published by Neely and Thayer (2011). Chapter 2 then discusses the analysis of the observational record from three specific lidars. The Chapter includes a discussion of the methods employed in the analysis of the lidar data. Finally Chapter 2 concludes with a discussion of the limitations of the observations in answering the questions of this thesis.

Chapter 3 introduces the specifics of the model used to help explore the observed variability of stratospheric aerosol. It then describes the methods employed to compare the model output data to the lidar observations. Next, runs from the base model configuration are compared to mean lidar volume backscatter coefficient aerosol profiles in the attempt to validate the model’s baseline state. The section also includes a discussion of how biases in lidar retrievals impacted the comparison and suggestions for improved stratospheric aerosol volume backscatter coefficient observations. This work is based on the publication “Implications of extinction due to meteoritic smoke in the upper stratosphere” by Neely et al. (2011).

The analysis of stratospheric variability is discussed in Chapter 4. First, observed and modeled annual cycles are compared. Next, the main source of the stratospheric aerosol increase from 2000 to 2010 is identified by experimenting with volcanic and anthropogenic source schemes and comparing to observations. The sources and implementation of the schemes used
are described in detail. Discussion of the analysis includes a comparison of the annual cycle of stratospheric aerosol under perturbed conditions to observations and the comparison of several model runs, which varied the injection magnitudes and heights of the simulated volcanic eruptions. It also includes a sensitivity analysis of the height and magnitude of the volcanic injections on the aerosol loading of the stratosphere.

Finally, the concluding chapter highlights the main findings of this work. It also discusses the limitations and concerns of the current analysis and makes suggestions about the next steps in understanding the variability of stratospheric aerosol.
CHAPTER II: GROUND-BASED LIDAR OBSERVATIONS OF STRATOSPHERIC AEROSOL

2.1 General Overview of The Rayleigh/Mie Lidar Technique

The first application of a ruby optical MASER (predecessor to all modern LASERs, Light Amplification by Stimulated Emission of Radiation) to the detection and ranging of a particle was made by Smullin and Fiocco (1962). Though the particle happened to be the Moon, Smullin and Fiocco (1962) paved the way to all modern LiLight Detection And Ranging (lidar) instruments. In 1967, Grams and Fiocco used the newly created ruby laser to create the first atmospheric lidar, then referred to as optical radar. Using this instrument, Grams and Fiocco (1967) made the first lidar observations of a volcanic eruption (Agung, Bali) and stratospheric aerosol. Currently, lidar is in wide use to make observations of various atmospheric variables. In this work, the observations discussed come primarily from Rayleigh/Mie (elastic) volume backscatter coefficient lidars that were specifically built and maintained to monitor the stratospheric aerosol layer (Barnes and Hofmann, 1997; Hofmann et al., 2003; Hofmann et al., 2009; Nagai et al., 2010).

2.1.1 Physical Description of the Elastic Scatter Lidar Measurement

The instrument design and retrieval method for aerosol volume backscatter coefficient (described originally by Fernald et al. (1972) and Klett (1981)) is in wide use (Russell et al., 1979; Fernald, 1984; Thayer et al., 1997; Hofmann et al., 2003; Pappalardo et al., 2004; Wandinger, 2005) and is only paraphrased here to describe the aspects of the retrieval process which are important to ascertain information about stratospheric aerosol.

The traditional elastic lidar measurement technique is dependent on a high-powered, pulsed laser transmitter (See Figure 3). The laser transmitter rapidly fires short, intense pulses of
light into the atmosphere and the individual photons then interact with the atmosphere. In this work, the only interaction under consideration is the elastic scatter from the molecular atmosphere constituents and aerosol. The transmitter is co-located and aligned with a large telescope receiver that collects a small fraction of the light scattered back to the instrument. The collected light is then transferred to a detector that converts photons into electronic pulses that are counted and accumulated into sequential time bins by a data acquisition system. The laser pulses and data acquisition are controlled by a precise timer so that the range of the backscattered light may be determined with an accuracy of meters using the time-of-flight principle. Thus, relative intensities of the backscattered light are accumulated separately from all altitude intervals over an averaging period. The basic lidar measurement technique is based on the assumption of single scattering. The time of flight principle is only valid for calculating the range or rather the distance the pulse of light travels from the transmitter to the scattering target, which are the molecular atmosphere and aerosol.

The raw data products of lidar measurements are profiles of detector counts (Figure 4). The retrieval used to turn these counts into information about the atmosphere is based on the fact that these counts are linearly proportional to the intensity of the backscattered light and the scattering volume consists of both molecules and aerosol. Detector saturation may cause this proportionality to have an altitude dependence and must be addressed in the retrieval. The molecular signal can be separated from the total volume backscatter coefficient signal by employing retrieval algorithms that contain a priori information about the molecular density of the atmosphere. Once the molecular scatter is removed, any residual signal is attributed to the presence of aerosol and an aerosol volume backscatter coefficient or scattering ratio can be determined. Additional derivables from the lidar volume backscatter coefficient data, which
involve assumptions about the aerosol, are aerosol extinction profiles, integrated volume backscatter coefficient and aerosol optical depth.

Figure 3: Basic lidar schematic showing the necessary components of an elastic lidar. Adapted from: www.mlo.noaa.gov/programs/gmdlidar/mlo/gmdlidar mlo.html.

Figure 4: Example of raw lidar measurements from NOAA’s lidar located in Boulder, CO. The lidar has two photon counting detectors in order to maximize the dynamic range of the measurement. Data were collected in March 2012.
2.1.2 Description of the Aerosol Elastic Volume Backscatter Coefficient Retrieval

Any elastic volume backscatter coefficient lidar transmitting light at wavelength $\lambda$, assuming single scattering, is described by the lidar equation (1):

$$N(\lambda, z, t) = N_L[\beta(\lambda, z, t)\Delta R]\frac{A}{z^2} \exp\left\{-2 \int_0^z \alpha(\lambda, z') \, dz'[\eta(\lambda, z, t)G(\lambda, z)] + N_B(\lambda, z, t)\right\}$$  (1)

Here $N(\lambda, z, t)$ is the total number of scattered photons detected by the receiver, $z$ is altitude above the surface (adjusted for the pointing angle of the lidar), $N_L$ is the number of transmitted photons, $\Delta R$ is the range resolution of the lidar, $\beta$ is the volume backscatter coefficient of the scatterers, $\alpha$ is the extinction coefficient of the atmosphere, $G$ is the geometric overlap function of the transmitter and receiver, $\eta$ is a system efficiency parameter, $N_B$ is the background number of photons detected and $t$ is time.

The volume backscatter coefficient and extinction coefficients may be separated into molecular ($m$) and aerosol ($a$) components:

$$\beta(\lambda, z, t) = \beta_a(\lambda, z, t) + \beta_m(\lambda, z, t)$$  (2)
$$\alpha(\lambda, z, t) = \alpha_a(\lambda, z, t) + \alpha_m(\lambda, z, t) + \sum_{i=1}^{n} \sigma_{X_i}(\lambda, z)N_{X_i}(z, t)$$  (3)

The molecular volume backscatter coefficient in the atmosphere may be determined empirically through the hydrostatic equation and quantum mechanics, but the volume backscatter coefficient from aerosol is quite variable and must be determined for each measurement (Elterman, 1964; Russell et al., 1979). The extinction coefficient has an additional term that accounts for extinction due to absorption at the transmit wavelength by different trace molecules in the atmosphere. This term is represented as summation of $n$ trace species with cross section $\sigma_{X_i}$ and volume number density $N_{X_i}$. 
The lidar volume backscatter coefficient ratio (LBSR) is defined as the ratio of the total volume backscatter coefficient to the molecular volume backscatter coefficient. Physically, the lidar LBSR is defined as:

\[
LBSR(\lambda, z, t) = \frac{\beta_a(\lambda,z,t)}{\beta_m(\lambda,z,t)}
\]

(4)

where \(\beta_a(\lambda, z, t)\) and \(\beta_m(\lambda, z, t)\) are the aerosol and molecular volume backscatter coefficients, respectively. The molecular volume backscatter coefficient is calculated from temperature and pressure profiles obtained twice daily from co-located radiosonde launches. The scattering ratio is retrieved from lidar signals by evaluating

\[
LBSR(\lambda, z, t) = \frac{S(\lambda,z,t)T^2(\lambda,z,t)}{C(\lambda)\beta_m(\lambda,z,t)T^2(\lambda,z,t)}
\]

(5)

where \(S(\lambda,z,t)\) is the background subtracted lidar signal \((N_s(\lambda,z,t) - N_B(\lambda,z,t))\), \(T^2(\lambda,z,t)\) (evaluated as \(\exp[-2 \int_0^z \alpha(\lambda,z')]\)) is the two-way atmospheric transmittance and \(C\) is a system constant determined by normalizing the right-hand side of the equation to an expected minimum value of \(LBSR(\lambda,z,t)\) over a specified calibration altitude range \((z^*)\) where the LBSR is small and known or can be assumed to be insignificant \((LBSR_{min} = 1)\) (Russell et al., 1979). The calibration term is shown explicitly here because the impact of errors in the \(LBSR_{min}\) term and the molecular profile used to separate the aerosol component from the total volume backscatter coefficient will be discussed at length in Chapter 3.

\[
C(\lambda) = \frac{N_s(\lambda,z^*,t)(z^*)^2}{LBSR_{min}\beta_m(\lambda,z^*,t)T^2(\lambda,z^*,t)}
\]

(6)

The transmittance is calculated from a combination of radiosonde derived molecular extinction model, lidar-derived aerosol extinction and modeled ozone absorption. During periods of moderate to heavy aerosol loading, aerosol extinction must be scaled to the aerosol volume backscatter coefficient using a model extinction to volume backscatter coefficient ratio (Jäger.
and Deshler, 2002). The lidar aerosol extinction to volume backscatter coefficient ratio is assumed to be between 35 and 60 sr according to the values derived from in situ balloon-borne measurements (Jäger and Deshler, 2002; 2003). Differences in the lidar ratio are not critical to derive the volume backscatter coefficient in small aerosol loading conditions, but after a large volcanic perturbation the extinction to volume backscatter coefficient ratio is very important to derive the extinction coefficient from the volume backscatter coefficient due to differences in the size distribution of the aerosol. Under background conditions in the stratosphere it has been traditionally assumed that $LBSR_{\text{min}}$ may represent only molecular scattering (i.e. it is defined as 1). Using this information, Equation (5) is solved iteratively from the top of the profile down, using an updated value of aerosol extinction for each iteration (Fernald et al., 1972 and Klett, 1981). Aerosol volume backscatter coefficient ($\beta_a$) profiles may then be derived by scaling the portion of LBSR attributed to aerosol scattering by the derived molecular volume backscatter coefficient

$$\beta_a(z, t) = (LBSR - 1)\beta_m(\lambda, z, t). \quad (7)$$

Analyses of the errors in the retrieval presented here are discussed at length by Russell et al. (1979). Note that small deviations in the normalization factor attributed to aerosol scattering can easily cause biases in the aerosol profiles. Russell et al. (1979) and, as will be discussed later, Neely III et al. (2011) show that simply including a small correction to account for additional aerosol scattering at the calibration altitude can improve the retrieved aerosol profiles when a topside calibration altitude is chosen erroneously to be aerosol free.

### 2.2 Lidars and Sources of Data Used within this Study

The data in this study comes from three ground-based Rayleigh/Mie lidar systems. The three lidars are located (See Figure 5) at Boulder, CO (40° N, 105° W, 1655 m, Mauna Loa
Observatory, Hawaii (20° N, 155° W, 3394 m) and Lauder, New Zealand (45° S, 170° E, 370 m), (Barnes and Hofmann, 1997; Hofmann et al., 2003; Barnes et al. 2008; Hofmann et al., 2009; Nagai et al., 2010). The lidar data record began at Lauder in 1992, Mauna Loa in 1994 and Boulder in 1999. NOAA’s Global Monitoring Division (GMD) of the Earth Systems Research Laboratory operates the lidars in Mauna Loa and Boulder. New Zealand’s National Institute of Water and Atmospheric Research (NIWA) operates the lidar in Lauder. Each of these systems employs frequency doubled Nd:YAG (532 nm) pulsed laser transmitters and an optical receiver arranged in a parallel arrangement.

Figure 5: Location of lidar systems whose data were used in this study.

These sites were chosen for their high data quality and long records. The quality and length requirements for inclusion in this study consisted of a collection of individual profiles to be taken every 7-10 days over the majority of the background aerosol period and error due to signal noise lower than 5% over the entire aerosol layer for each individual lidar profile. These
requirements stem from the work of Hofmann et al. (2009), which stated the annual variation in aerosol volume backscatter coefficient is two and a half times larger than the magnitude of the observed long-term trends. Therefore, the trend would be difficult to detect by any method that could not accurately resolve the annual variation. Figure 6 through Figure 8 show the aerosol volume backscatter coefficient from each lidar site for the entirety of the respective instrument’s data collection period.

Figure 6: Full record of aerosol volume backscatter coefficient from Boulder, CO. The white line represents the tropopause.

Figure 7: Full record of aerosol volume backscatter coefficient from Mauna Loa, HI. The white line represents the tropopause.

Figure 8: Full record of aerosol volume backscatter coefficient from Lauder, NZ. The white line represents the tropopause.
Several volcanic events briefly affected the region of the aerosol layer from 2000 to 2010. These events (volcanoes of volcanic explosivity index (VEI) ≤ 4) were once considered to be minor, but as later will be discussed, they play a large role in the variability of the stratospheric aerosol layer (Newhall and Self, 1982; Hofmann et al., 2009; Vernier et al. 2011a; Solomon et al., 2011). In order to ensure that data were not skewed by the direct injection of aerosol created by these small volcanic events, lidar profiles for two weeks after each of these events were not included in the data set. The data were also filtered for other unusual profiles that could have been caused by instrument error or atypical distributions of aerosol. The filtered data are shown in Figure 9 through Figure 11.
Figure 10: Filtered aerosol volume backscatter coefficient record from Maun Loa, HI. The white line represents the tropopause.

Figure 11: Filtered aerosol volume backscatter coefficient record from Lauder, NZ. The white line represents the tropopause.
2.4 Methods of Lidar Observation Analysis

For this study, the aerosol volume backscatter coefficients from the three lidar sites were analyzed to describe the observed annual cycles and trends in the aerosol. To accomplish the analysis, it was broken into three parts. First, the lidar data were collected from the three lidar sites, compiled into a coherent dataset and checked for errors. Then, the lidar data were analyzed for annual cycles and trends using mathematical regression techniques described here. Finally, these results were compared to model output as described in Chapters 3 and 4.

To quantitatively examine the annual cycles and trends (Figure 12), the volume backscatter coefficient data from each site were integrated using the technique depicted in Figure 13. Variable integration limits were used rather than constant limits so that the layers being compared over each lidar would be sampled from similar air masses with respect to the tropopause. It should be noted that though the upper and lower limits for each integration may vary, the thickness of the layers being compare was constant and the same for each site. The integrated aerosol volume backscatter coefficient (IABS) of each site was then fitted using a series expansion of sine functions. Fitting with a series of separate sine functions allowed for easy selection and removal of annual cycles and long-term trends from other harmonics in the data by comparing the frequency of each sine term to known geophysical cycle timescales. The technique used in this work is based on the method of Thoning et al. (1989) and was employed in a similar analysis by Hofmann et al. (2009). The annual harmonic was then removed from the original IABS so that a linear fit could be made to assess the long-term trend of the data. An example of the analysis applied to data from Mauna Loa may be seen in Figure 12.
Figure 12: Example of lidar data analysis using data from Mauna Loa, HI. The black circles represent the individual integrated aerosol volume backscatter coefficient observations (raw). The dark blue line is the smoothed dataset derived from the raw points. The yellow line represents the sum of sines fit to the blue line with the annual mode removed from the full fit. The green dashed line is the linear fit to the yellow line. The light blue is the residual of the smoothed data set and yellow line. The dashed red line is the fit of the light blue line using a singular sine function. The dashed pink line is a linear fit to the light blue data. Figure 12 is just for reference for the discussion of the analysis methods below. More detail is given below for each of these lines.

The integrated aerosol volume backscatter coefficient (IABS) is defined traditionally as the integral of the aerosol volume backscatter coefficient (BS) from the bottom of layer of interest to the top of the layer (Barnes and Hofmann, 2001). IABS is defined as

$$IABS_x(z, t) = \int_{Bottom~of~Layer}^{Top~of~Layer} \beta_a(z, t)dz \quad (8)$$

following the traditional definition used by Hofmann et al. (2003). IABS is analogous in interpretation to an aerosol optical depth (AOD). Here $x$ represents the lidar site, $z$ is altitude and $t$ is time. In Figure 12, the IABS is represented by the black circles.

The limits for integration were chosen in order to not bias the annual cycle with annual motions of the tropopause (Figure 13). It was decided that limits that used the tropopause at the time of the collection of each profile would be a better basis than integrating between two constant altitude limits throughout the dataset. Without the integration limit correction, the shift
in the column due to the annual changing position of the tropopause causes an oscillation that could modulate the true annual cycles in the aerosol (Figure 14-16). The correction does not assume that the location of the center of mass of the aerosol follows the motion of the tropopause. Nor does the technique presented here try to follow the motion of the aerosol layer. The intention of this technique is only to more accurately compare air masses throughout the annual oscillations and over the entire observational period.

In order to equally compare the data from one site versus the other an additional correction was made by integrating the layer above each site with a common center point. Preferably, the comparison would have been done on a pressure grid and these corrections would not have had to be made but since the native grid of the data is geometric altitude and no temperature data is routinely taken, conversion to a pressure grid was not done. The integration limits used in this study were found by taking the difference in the mean tropopause height and the top and bottom of the layer of interest and then adding these differences to the tropopause of each lidar profile and integrating between these new limits. See Figure 13 for a depiction of the integration scheme.

Figures 14-16 depict the bias that is introduced when integrating between constant limits. Figure 16 shows that the change in limits does not affect the mean or decadal trend but does have a significant effect on the amplitude of the annual cycle. It is also noted that the difference in the two schemes contains no trend.
3.2 Lidar Data Analysis

Chapter 3: Methods of Data Analysis

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Figure 3.2: Example of the lidar data analysis using data from Mauna Loay HI

Figure 3.3x3.5 depict the bias that is introduced when integrating between conx

The limits for integration were chosen in order to not bias the seasonal cycle
integrated from 20km to 25km but the top and bottom levels of the integration are
Figure 3.4: Lidar data from Boulder integrated from constant limits of 20km to

Figure 3.5: Lidar data from Boulder integrated from 4km to 9km above the

Integrated Aerosol Backscatter ($1E-6 \, 1/sr$)

Top = 25 km

Bottom = 20 km

Stratosphere

Troposphere

Surface

Mean Tropopause

Actual Tropopause

Annual Change

Figure 13: Diagram depicting limits of aerosol volume backscatter coefficient integration used in
this analysis

Boulder, CO Lidar IABS from 20km to 25km

Figure 14: Lidar data from Boulder integrated from constant limits of 20 km to 25 km.
Figure 15: Lidar data from Boulder integrated from 4 km to 9 km above the tropopause (the mean tropopause height at Boulder is ~16 km). The integration is centered at the same altitude range as the data integrated from 20 km to 25 km but the top and bottom levels of the integration are allowed to oscillate with the tropopause.

Figure 16: Time series of the difference in the lidar data from Boulder integrated from 4 km to 9 km above the tropopause and the data integrated using constant limits at 20 km and 25 km. The difference depicts the bias in the annual cycle that can be introduced to the integrated volume backscatter coefficient by not allowing for changes in the tropopause height.
Numerically the calculation is accomplished by turning the integral into a Riemann sum of the aerosol volume backscatter coefficient from the bottom to the top of the layer and multiplying the sum by the range resolution of the lidar instrument (Equation 9).

$$IABS_x(t) = \sum_{z=Bottom}^{z=Top} BS_x(z, t) \Delta z$$ \hspace{1cm} (9)

The dataset of the IABS for each site was then smoothed using a moving average as a low pass filter, to remove higher frequency oscillations from the data that could be due to instrumental differences between individual lidar profile collections and geophysical changes that take place on time scales of less than a week. The smoothed IABS (SIABS, blue line in Figure 12) was calculated over an interval of five profiles, approximately one month’s worth of observations. Experiments with averaging showed that inclusion of more than 5 data points removed natural variability on a time scale similar to the annual cycle. The smoothing function takes the form of Equation 10.

$$SIABS_x(t) = \frac{\sum_{t=2}^{t=\text{Length of Data Set}} IABS_x(t)}{(t+2)-(t-2)}$$ \hspace{1cm} (10)

After deriving the SIABS of the layer of interest, the data were fitted using a sum of sines regression model of the form:

$$Sum \ of \ Sines \ Model_x(t) = \sum_{i=1}^{g} a_{i,x} \sin(b_{i,x}t + c_{i,x}).$$ \hspace{1cm} (11)

From the series of sine functions, the annual mode may be identified by looking at the frequency, $b$, of each harmonic from the fit. This harmonic may then be removed leaving the remaining terms:

$$Sum \ of \ Sines \ Mode \ sans \ Annual \ Mode_x(t) =$$

$$\sum_{i=1}^{g} a_{i,x} \sin(b_{i,x}t + c_{i,x}) - a_{Annual,x} \sin(b_{Annual,x}t + c_{Annual,x}).$$ \hspace{1cm} (12)
The yellow curve represents the results of Eq. 12. In the legend the fit parameters displayed (root mean square of the fitting error, RMSE, and $r^2$, the correlation coefficient, RSQR) correspond to the original model fit.

A linear regression (green dashed line) is then applied to the sum of sines fit with the annual mode removed to determine the long-term trend of the layer. The linear model is defined in Eq. 12.

$$Linear Model_x(t) = a_{Linear,x}(t) + b_{Linear,x}$$  \hspace{1cm} (13)

The annual mode must be removed from the data before the trend may be described using a linear model because the amplitude of the annual cycle is 2-3 times larger than the increase caused by the trend (Hofmann et al., 2009). When fitting to data where the annual cycle has not been removed, the trend becomes dependent on the temporal limits of integration. This can even lead to fits of the data showing falsely negative trends. Using the trend analysis described above allows for the linear fit to be independent of the start and stop points of the data. The value for the trend and associated fit errors are located in the legend (Figure 17-Figure 25).

To create a robust model of the annual cycle that does not contain the effects of the long-term trend, the sum of sines fit with the annual mode removed is first subtracted from the SIABS data.

$$Annual Mode Residuals_x(t) =$$

$$SIABS_x(t) - Sum\ of\ Sines\ Mode\ sans\ Annual\ Mode_x(t)$$  \hspace{1cm} (14)

Equation 14 leaves only the residual of the annual mode within the data (light blue line in Figure 13) that can be fit with a singular sine function to give a numerical description of the frequency, amplitude and offset of the annual cycle (red dashed line). To check whether the residuals did contain any trend, a linear fit was applied to the residuals (pink dashed line). If the analysis is
done correctly, the fit results in a line with zero slope. When this is true, the residuals may be fit with the sine function with confidence that the result should only describe the annual cycle. The results from the linear fit to the residuals are also found in the legend of the plot.

Other regression and fitting analyses were explored, including a Fourier analysis, but the fitting procedure described above statistically produced the best results for explaining the variability of the data as determined through minimization of the residuals. A Fourier analysis of the spectral power density of the integrated layers agrees with the strong annual mode identified with the sum of sines fits but is not shown here.

2.5 Results of Lidar Observation Analysis

For each instrument being examined in this study, three layers of integrated aerosol volume backscatter coefficient – centered about the geometric layers of 20-25 km, 25-30 km and 30-35 km – have been analyzed for annual cycles and trends in an attempt to understand the processes occurring throughout the stratospheric aerosol layer. Each plot contains the raw data being examined as well as the numerical description of the long-term trend and annual cycle. Various other layers were also examined but it was found that the main trends and behavior of the entire aerosol layer are captured by the three layers defined previously. These layers may be considered the lowermost (20-25 km), middle (25-30 km) and uppermost (30-35 km) sections of the sulfate aerosol layer. Below the results of the analysis for each site are discussed.

Analysis of the stratosphere from the tropopause to 20 km is omitted here because the model used in the comparisons in the following chapters does not include the complete array of tropospheric aerosol necessary to correctly simulate the aerosol distribution near the tropopause.
2.5.1 Mauna Loa, HI

The analysis for Mauna Loa is presented in Figures 17-19. The analysis shows an annual cycle in the integrated aerosol volume backscatter coefficient peaking in winter for each of the layers. The annual cycle is strong in the 20-25 km and 25-30 km layers which both display a cycle with a 20% amplitude. The layer from 30-35 km shows a cycle of slightly smaller magnitude and the cycle is much noisier. The long-term trends of the layers decrease with altitude from 3.3% in the lowest layer to 1.0% in the top layer. Upon comparison, the analysis here agrees with the annual variation in the stratospheric aerosol volume backscatter coefficient observed at Mauna Loa Observatory by Barnes and Hofmann (2001) after the decay of the Pinatubo aerosol in 1996 and the trend analysis in the more recent work of Hofmann et al. (2009). The trends reported here are slightly smaller than those reported by Hofmann et al. (2009) due to the slight decrease in aerosol volume backscatter coefficient observed after 2007, which was not fully captured in data available in 2009. In addition to the annual cycle and long-term trend, there is a biennial variation in the data. The variation is most likely related to the quasi-biennial oscillation (QBO) in tropical winds as shown by Barnes and Hofmann (2001) and Hofmann et al. (2009).
Figure 17: Integrated aerosol volume backscatter coefficient for the layer centered around 20-25 km above Mauna Loa.

Figure 18: Integrated aerosol volume backscatter coefficient for the layer centered around 25-30 km above Mauna Loa.
Figure 19: Integrated aerosol volume backscatter coefficient for the layer centered around 30-35 km above Mauna Loa.

2.5.2 Boulder, CO

Figure 20 through Figure 22 contain the results from the Boulder lidar. The analysis shows a strong annual cycle in the integrated aerosol volume backscatter coefficient peaking in winter for each of the layers. The annual cycle is strongest in the 25-30 km and 30-35 km layers, which both display a 34% amplitude. The layer from 20-25 km contains a smaller annual cycle with an amplitude of 14% and is not as distinct due to nosier data. The long-term trends of the layers vary with altitude from 4.4% in the lowest layer to 5.7% in the middle layer but then decreases significantly to almost no trend in the top layer. Upon comparison, the analysis of the middle layer agrees well with the annual variation and long-term trend observed for Boulder by Hofmann et al. (2009). Unlike Mauna Loa, Boulder does not show a significant decrease in aerosol content after 2007. In addition to the annual cycle and long-term trend, there is also a biennial variation in the de-annualized data as observed in Mauna Loa as described by Hofmann et al. (2009).
Figure 20: Integrated aerosol volume backscatter coefficient for the layer centered around 20-25 km above Boulder.

Figure 21: Integrated aerosol volume backscatter coefficient for the layer centered around 25-30 km above Boulder.
2.5.3 Lauder, NZ

The results of the analysis for Lauder are contained in Figure 23 through Figure 25. Similar to Boulder and Mauna Loa, the analysis reveals an annual cycle in the integrated aerosol volume backscatter coefficient peaking in late winter for each of the layers. The absolute magnitude of the cycle is smaller in Lauder than observed in the other sites. Like Boulder, the cycle is strongest in the 25-30 km and 30-35 km layers, which both display a cycle with a 37% amplitude. The layer from 20-25 km shows a cycle of slightly smaller amplitude and, like Boulder, is much noisier. The magnitude of the long-term trends of the layers decreases with altitude from 8.9% in the lowest layer to 6% in the top layer. Upon comparison, the analysis of Lauder’s data shown here agrees with the annual variation in the stratospheric aerosol volume backscatter coefficient and the trend analysis found in the recent work of Nagai et al. (2010). The biennial variability in the Lauder data, with the annual cycle removed, is not as apparent as observed in the Mauna Loa and Boulder data but the sum of sines regression of the data does contain a term which accounts for the variability in the data that may be attributed to the QBO.
Figure 23: Integrated aerosol volume backscatter coefficient for the layer centered around 20-25 km above Lauder.

Figure 24: Integrated aerosol volume backscatter coefficient for the layer centered around 25-30 km above Lauder.
2.6 Discussion

The long-term trends and amplitudes of the annual cycles for all layers (labeled using the mean center height of the layer) analyzed at each site are summarized in Table 1:

<table>
<thead>
<tr>
<th>Site</th>
<th>Layer Center (km)</th>
<th>Linear Trend Increase (%)</th>
<th>Fit Error (%, NRMSE)</th>
<th>Annual Cycle Peak (DOY)</th>
<th>Period (Days)</th>
<th>Amplitude (%)</th>
<th>Fit Error (%, NRMSE)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Boulder</td>
<td>22.5</td>
<td>4.41</td>
<td>0.24</td>
<td>51</td>
<td>364.8</td>
<td>14.3</td>
<td>1.05</td>
</tr>
<tr>
<td></td>
<td>27.5</td>
<td>5.72</td>
<td>1.70</td>
<td>19</td>
<td>364.4</td>
<td>34.8</td>
<td>0.98</td>
</tr>
<tr>
<td></td>
<td>32.5</td>
<td>0.14</td>
<td>0.97</td>
<td>320</td>
<td>366.1</td>
<td>34.2</td>
<td>0.81</td>
</tr>
<tr>
<td>Mauna Loa</td>
<td>22.5</td>
<td>3.27</td>
<td>2.00</td>
<td>363</td>
<td>364.4</td>
<td>20.7</td>
<td>0.88</td>
</tr>
<tr>
<td></td>
<td>27.5</td>
<td>2.46</td>
<td>2.06</td>
<td>339</td>
<td>364.6</td>
<td>20.3</td>
<td>1.52</td>
</tr>
<tr>
<td></td>
<td>32.5</td>
<td>1.01</td>
<td>2.47</td>
<td>289</td>
<td>372.3</td>
<td>19.7</td>
<td>1.23</td>
</tr>
<tr>
<td>Lauder</td>
<td>22.5</td>
<td>8.92</td>
<td>1.72</td>
<td>245</td>
<td>365.3</td>
<td>24.4</td>
<td>1.55</td>
</tr>
<tr>
<td></td>
<td>27.5</td>
<td>4.42</td>
<td>2.27</td>
<td>221</td>
<td>363.4</td>
<td>37.3</td>
<td>1.29</td>
</tr>
<tr>
<td></td>
<td>32.5</td>
<td>5.99</td>
<td>2.05</td>
<td>214</td>
<td>363.2</td>
<td>36.7</td>
<td>1.48</td>
</tr>
</tbody>
</table>

Table 1: Summary of lidar observation analysis. Parameter estimates come from Figure 17 through Figure 25. The error estimates come from the normalized root mean square of the fit residuals of each parameter.

Most notable in this analysis is the symmetry between the annual cycles in Lauder and Boulder. Both have wintertime peaks in their annual cycles that vary with altitude. The
amplitudes of the two mid-latitude annual cycles in the top two layers are equal while the bottom layer has a diminished annual cycle. The similarity is most likely due to the fact that the annual aerosol variability in the stratosphere is due to transport by the mean meridional circulation. The transport creates a clean annual signal at higher altitudes while intrusions into the lower stratosphere from summertime convection create the observed noise at lower levels (Holton et al., 1995; Niwano et al., 2009). These two topics will be expanded upon in the discussion of the annual variation of stratospheric aerosol in Chapter 4. The overall results of the lidar analysis presented here agree with the global distribution of stratospheric aerosol described by Niwano et al. (2009), using data from Stratospheric Aerosol Gas Experiment (SAGE II), and Vernier et al. (2009; 2011a), using data from SAGE II, the Global Ozone Monitoring by Occultation of Stars (GOMOS) and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP). Both of these studies suggest that the annual cycle of stratospheric aerosol is caused by the slow ascent and poleward motion of aerosol in the tropics associated with the Brewer-Dobson circulation and the QBO modulation of the meridional transport toward the subtropics shown by Hitchman and McKay (1994).

Unlike the two mid-latitude sites, at Mauna Loa the annual cycle is stronger in the lower two layers. The higher amplitude, lower region may be explained by the relatively higher tropopause at Mauna Loa and Mauna Loa’s position in the tropics. It is well known that many species with tropospheric origins have a tape recorder that shows alternating patterns of high and low mixing ratios, propagating upward (Mote et al., 1996; Plumb, 1996; Park et al., 2004; Schoeberl et al., 2006; Pumphrey et al., 2008). These are caused by annual variations in their source to the stratosphere that are then transported upwards. The observations at Mauna Loa suggest the aerosol exhibit a tape recorder effect similar to that of stratospheric water vapor
(Vernier et al., 2011b). It should also be noted that Mauna Loa is located such that the atmosphere above the site is tropical in nature some of the time, and more mid-latitude in nature at other time times, depending on the position of the sub-tropical jet. It is possible that the annual cycle observed here may be biased as a result of sampling these different air masses at different times of year. This point needs further examination than the scope of this study entails.

In addition to the annual cycle that has been discussed, the lidar observations show significant positive trends in aerosol volume backscatter coefficient over the last decade. The long-term trends range from negligible to 8.0% ± 2% per year over the altitude range of data examined. No apparent global pattern is observed except for a decrease in the positive trend with respect to height, though the decrease in the trend is not strictly monotonic for each site. The observed altitude dependence is consistent with the results of earlier studies (Barnes and Hofmann, 2001; Hofmann et al., 2009) and trends shown in stratospheric AOD reported by Vernier et al. (2011a) (Figure 26). Currently, there are at least three possible hypotheses for these trends.
One possible mechanism for the observed enhancement in aerosol volume backscatter coefficient includes the recent increases in SO$_2$ emissions from sources near the region of the Asian monsoon. Satellite observations show that pollution is being transported to the
stratosphere during the boreal summer over the Himalayan plateau. Such observations suggest a preferential injection of Asian pollution into the stratosphere during the Asian monsoon. These findings are based on the studies of Fu et al. (2006) and Park et al. (2007) that showed observations of variations in water vapor, CO and ozone that correlated with the intensity of underlying deep convection of the Asian monsoon. Randel et al. (2010) also demonstrated the possibility through observations of HCN being transported deep into the stratosphere by convection associated with the monsoon. This was further confirmed by observations from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) of an Asian tropopause aerosol layer that correlated with the existence of the Asian Monsoon (Vernier et al. 2011b).

A second potential source of the observed increases is a string of smaller volcanic eruptions over the last decade as noted by Vernier et al. (2009) in data from CALIOP. Vernier et al. (2009; 2011a) focused on the tropics and more investigation needs to be done to understand the long-term global effect that small eruptions can have on a global scale. Some of the specific questions that need to be answered include 1) how tropical versus high latitude volcanoes affect the stratosphere and 2) how critical is the maximum injection height of the volcano to impacting the global stratosphere. The influences of these volcanoes are quite evident on short timescales within the lidar observations but the long-term effects, especially in the mid-latitudes, are not well understood. The persistence of the long-term trend in the Boulder and Lauder lidar datasets – while Mauna Loa has shown a recent decrease – may be evident of this fact. As suggested by Hofmann et al. (2009), the only way to discern the source of the trends is through modeling the background sulfate aerosol in the stratosphere. As described by Hofmann et al. (2009), the model must include the latest SO$_2$ emission data, tropical deep convection, complete tropospheric sulfur chemistry, and aerosol growth under changing vapor pressures in the stratosphere in order to
delineate important mechanisms. As per this suggestion, the next two chapters describe the modeling done to attribute the observed aerosol variability to a source.

Pyrocumulonimbus (pyroCb) events are similar in nature to volcanic injection of aerosol to the stratosphere (Fromm, 2008). For the purposes of this work the injections are acknowledged as another source of variability in the aerosol record but are not dealt with directly. PyroCb are ignored here because their ability to impact the middle and upper stratosphere (above 20 km) is negligible (Fromm, 2008). Vernier et al. (2011a) show that even the massive Victoria, Australia fire in February 2009 had minimal impact on the stratosphere in comparison to the volcanic eruptions from 2000 to 2010. More observations of pyroCb will be needed before they can be analyzed with simulations similar to those in this work (Fromm, 2008).

It has also been suggested that the stratospheric circulation will be impacted by greenhouse gas warming (Butchart et al., 2010). If the cross tropopause upward flux of tropical air has increased, it will loft more COS, more SO\textsubscript{2} and more aerosol into the stratosphere. This would also cause an upward aerosol trend. In the absence of compensating radiative processes, an increase in the upward flux of tropical air would also cause a decrease in tropical tropopause temperatures, which would increase nucleation rates of sulfate aerosol at the point of injection into the stratosphere (Hamill et al., 1997; Yue, 1982). Evidence for such a mechanism over the last decade may be found in the work of Rosenlof and Reid (2008). This process will not be examined in this work due to the limitations of the current model.

As indicated earlier, there is also a QBO signal present in the data for each site. A consistent relationship between increases in aerosol volume backscatter coefficient at high altitudes and the QBO has been observed at both Mauna Loa and Boulder by Hofmann et al.
(2009) but has not been noted in the literature for Lauder. Hofmann et al. (2009) show that the aerosol volume backscatter coefficient growth rate consistently increases during the westerly phase and decreases during the easterly phase of the QBO. The modulation in aerosol abundance is most likely due to increased tropical upwelling during the westerly phase of the QBO (Hofmann et al., 2009; Randel et al., 2010). In this study, the QBO impact is noted but not investigated further because – though it modulates the annual cycle – the QBO analysis is thought to be more important when looking at the long-term trends of the data. Observations of the QBO effect in the lidar data provide further motivations for future numerical experiments to examine how enhanced tropical upwelling and increases of SO$_2$ near areas with deep tropical convection may perturb aerosol, and if this could be the cause for the observed increases in aerosol as noted above.

2.7 Conclusions

The lidar observations show clearly that decadal variability of stratospheric aerosol consists of an increasing linear trend of up to 8% per year superimposed upon variability that is due to fluctuations in transport processes such as the mean meridional circulation and the QBO. Figure 27, derived from satellite observations of stratospheric aerosol, clearly indicates the influence of volcanic injections on the stratosphere from 2002 to 2009. It also shows enhancements in aerosol extinction during periods of no volcanic eruptions, as in 2007. As with the lidar observations, a clear partitioning of the sources of the long-term trends cannot be made. Ultimately the ambiguity of the observations leads to the question of how different types of sources (slow constant emissions from the surface versus quick direct volcanic injections) impact the stratosphere. Other questions also remain as to whether an annual cycle exists or if the cycle is an instrumental artifact. Because observations from ground based lidars and other instruments...
alone will not conclusively answer these questions, as suggested by Hofmann et al. (2009), the modeling effort described in the rest of this work was conducted to evaluate the questions posed by these observations (Vernier et al., 2009; 2011a; 2011b).

Figure 27: Monthly mean extinction ratio (525 nm) profiles evolution in the tropics (20°N-20°S) from January 2002 to December 2010 derived from SAGE II, GOMOS and CALIOP. Black contours represent the extinction ratio in log-scale from 0.1 to 100. The position of each volcanic eruption occurring during the period is displayed with its first two letters on the horizontal axis, (tropical eruptions are in red). These other eruptions are listed in Table 2 in Chapter 4. Plot provided through personal communication with Jean Paul Vernier.
CHAPTER III: MODELING STRATOSPHERIC AEROSOL

3.1 Overview
To elucidate the differing roles of transport, chemistry and microphysical processes on stratospheric aerosol, the results from Chapter 2 are compared to model simulations. The model used is a version of the Whole Atmosphere Community Climate Model 3 (WACCM) coupled to the Community Aerosol and Radiation Model for Atmospheres that has been developed to examine processes in the stratospheric aerosol layer (English et al., 2011). In this section the model is described and a baseline run is compared to observations to validate the model’s use for exploring the variability of stratospheric aerosol.

3.2 WACCM: Whole Atmosphere Community Climate Model
WACCM is a comprehensive chemistry-climate model that spans from the Earth’s surface to the thermosphere. We use WACCM3 version 3.1.9 tag 9 with 30-minute time steps at 4° latitude by 5° longitude horizontal resolution with 66 vertical levels based on hybrid-sigma coordinates and a model top near 140 km. The grid setup results in the model simulating levels with vertical spacing of 1-1.75 km in the stratosphere. The model version has been used to simulate the middle and upper atmosphere in many studies and a full description may be found in the work of Garcia et al. (2007).

The dynamical and transport equations are solved using a finite volume dynamical core from NCAR’s Community Atmosphere Model (Lin, S.-J., 2004). WACCM takes into account sub-grid scale dynamical forcing through parameterized orographic gravity waves. A spectrum
of non-stationary gravity waves is also included (Garcia et al., 2007). The inclusion of gravity waves produces a more realistic zonal mean wind field. The model used here is specifically configured with tuning parameters for the gravity waves that have been adjusted to produce a realistic stratospheric temperature distribution. These calculations also use the interactive land and ice models integrated in WACCM, and prescribed sea surface temperatures as described by Mills et al. (2008) and Bardeen et al. (2008a; b; 2010).

The chemistry module is derived from the 3D chemical transport Model for OZone And Related chemical Tracers (MOZART). A full description of MOZART is found in Horowitz et al. (2003). For this study, WACCM uses a 63-species chemistry module that is implemented to represent the span of chemical and physical processes in the stratosphere. WACCM’s standard 56-species chemical package includes O$_x$, NO$_x$, HO$_x$, ClO$_x$, and BrO$_x$ chemical families along with CH$_4$ and its products and 7 ions (Kinnison et al., 2007). Seven sulfur-bearing gases are added to the chemistry scheme to accurately simulate sulfate aerosol and are described more fully in Mills et al. (2005). The sulfur species are S, SO, SO$_2$, SO$_3$, HOSO$_2$, H$_2$SO$_4$, and OCS. The reaction rates and photo-dissociation rates for these sulfur species are provided in English et al. (2011).

The model includes emissions of carbonyl sulfide (OCS) and sulfur dioxide (SO$_2$), which are the two primary sulfur emissions of importance to the stratospheric aerosol layer. OCS is specified with a constant surface concentration of 510 pptv. SO$_2$ is specified from a two-dimensional monthly mean surface emissions dataset representative of the year 2000 (Lamarque et al., 2010; Smith et al., 2011). In Figure 28 and 29 the January and July SO$_2$ surface emission is shown to illustrate the annual global variability of the base SO$_2$ emissions. The base model configuration does not include emissions of dimethyl sulfide (DMS) or tropospheric volcanic
SO₂ (including injections into the upper atmosphere from explosive eruptions and slower surface emissions from effusive eruptions), which would make a minor contribution to the background stratospheric aerosol. In the next chapter, volcanic injections into the upper troposphere and lower stratosphere and trends in anthropogenic emissions from the last decade are described and added to the model for comparison against the observed trends in the lidar data.

Figure 28: January WACCM baseline SO₂ emissions.

Figure 29: July WACCM baseline SO₂ emissions.

Wet deposition for all constituents (including the aerosol bins from CARMA) is calculated using WACCM’s existing techniques (Barth et al., 2000). All of the aerosol bins are assumed to have a constant 0.3 solubility parameter. WACCM treats dry deposition of gases
(Barth et al., 2000), while dry deposition of aerosol is not treated in this model, since it is not important for stratospheric aerosol.

3.3 CARMA: Community Aerosol and Radiation Model for Atmospheres

CARMA 2.3 has been coupled to WACCM and configured to handle all aerosol microphysics. CARMA is a sectional aerosol model (i.e. the model creates a size distribution by defining many discrete size bins in which a concentration of aerosol may exist) based on the three dimensional model originally described by Toon et al. (1988). CARMA originated from a one-dimensional stratospheric aerosol code first developed by Turco et al. (1979) and Toon et al. (1979) that included both sulfur chemistry and aerosol microphysics. Since this first work, extensive updates to the aerosol microphysics and numerical calculations have been made (Bardeen et al., 2008a; b; 2010; English et al., 2011).

WACCM has been coupled to CARMA in a manner similar to that used in the meteoritic-smoke studies by Bardeen et al. (2008a; b; 2010) and in a study on ozone loss after a nuclear conflict by Mills et al. (2008). The coupling of CARMA to WACCM is accomplished by integrating a single column version of CARMA into the physics routine of WACCM using the same vertical grid. This means that at every model time step, the model’s state is passed to CARMA one column at a time. Then CARMA calculates changes to the constituents within these columns from the microphysical processes specified within CARMA. These changes are then passed back to WACCM, which adjusts its state accordingly before taking another time step. (In the configuration of the model used here, the aerosol in CARMA are not coupled to the radiation code in WACCM so interactive aerosol heating does not adjust the dynamics and chemistry of WACCM). For this to work with WACCM’s framework, each aerosol size bin is added to WACCM as a unique constituent. Thus, WACCM controls the advection, vertical
diffusion and wet deposition of each of the aerosol, while CARMA calculates production by nucleation, sedimentation, growth, evaporation and coagulation of the aerosol.

CARMA calculates the effect of coagulation of particles of equivalent aerosol size using the numerical approach described in Toon et al. (1988). Binary homogeneous nucleation of sulfuric acid and water is calculated using the numerically efficient method of Zhao and Turco (1995). Split-time stepping is enabled to allow for simultaneous solution of nucleation and growth routines when sulfuric acid is supersaturated. Coagulation coefficients are calculated to include Brownian, convective and gravitational effects. A sticking coefficient of 1 is used, which assumes that all particles stick together upon colliding. A correction for the impact of inter-particle van der Waals forces on coagulation is included (Chan and Mozurkewich, 2001).

Three types of aerosol are considered in this configuration of CARMA to model the stratospheric aerosol layer completely. They are composed of pure sulfate, mixed sulfate (sulfate aerosol with smoke cores) and meteoritic smoke. Other types of aerosol were not included, as sulfates and meteoritic smoke are believed to be the primary aerosol in the upper stratosphere (Murphy et al., 2007). For the microphysical calculations this version of CARMA is set up such that the size distribution of the aerosol is spread across thirty-six aerosol bins for each type of aerosol (two sets are needed for the mixed sulfate in order to separately handle the necessary microphysics of the sulfate shell and smoke core). Each set ranges in dry radius from 0.2 nm to 1100 nm. During every time step of the model, each of these sets of bins goes through the full set of microphysical calculations. These calculations sometimes take many time sub-steps in order to properly handle the large changes in concentration of gases and aerosol. Each set of bins is also allowed to transfer mass into and out of each other type of aerosol bin though, for conservation purposes, particles in each corresponding bin of the different sets have equal mass.
After the model run is complete, concentration data from each set of bins, which is output on monthly time steps for this study, is used as a size distribution to compute extinction and volume backscatter coefficient from Mie scattering calculations so that the modeled aerosol optical properties may be compared to lidar observations.

The meteoritic smoke source in this model is based on the work of Bardeen et al. (2008a; b) who used WACCM to make the first three-dimensional simulation of meteoric smoke. Bardeen et al. (2008a; b) showed that the mesospheric meridional circulation creates an annual pattern in meteoric smoke concentration with a winter maximum. The smoke is emitted as 0.2 nm particles (the smallest bin) using flux values from previous studies of micrometeorite ablation to specify the total mass emitted as described in Bardeen et al. (2008a; b). All newly emitted smoke enters the model between the altitude levels of 0.01 hPa (75km) and 0.0001 hPa (110km). The global mean mass flux of the meteoritic smoke is 16 kt yr$^{-1}$. Figure 30 and 31 show the global annual cycle of the smoke mixing ratio at 0.0179 hPa.

Careful consideration of the composition of meteoritic smoke and the sulfate aerosol is important for the optical calculations made to compare the modeled aerosol size distributions to observations because the refractive index depends strongly on the particle composition. Limited observations and modeling show that meteoric smoke is primarily composed of olivine and hematite (Hervig et al., 2009). For the purposes of the calculations here it is not necessary to specify the exact composition of the meteoritic material, but it is important to correctly model the observed extinction at the wavelength being examined. Thus, for these purposes optical constants were chosen from the work of Jäger et al. (2003) and Huffman and Stapp (1973) that produced similar extinction profiles as those observed by Hervig et al. (2009). The index of refraction for the sulfate aerosol is better understood due to the less complex composition of the aerosol. As
Figure 30: January mass density of meteoritic smoke at 0.956 hPa.

Figure 31: July mass density of meteoritic smoke at 0.956 hPa.
suggested by Palmer (1975), the real part of the index of refraction was allowed to vary as a function of the weight percent of sulfuric acid. Absorption was ignored due its small imaginary component at visible wavelengths. Because the sulfate bins do not include the contribution from water, the equivalent sulfate aerosol size (including sulfuric acid and water) is determined in post processing as described by Tabazadeh et al. (1997), which calculates weight percent sulfuric acid as a function of temperature and water activity. Weight percent sulfuric acid is assumed to be independent of particle size.

### 3.4 Scattering Calculations of Model Output

To compare the model output to observations, aerosol volume backscatter coefficient and extinction were calculated from the model’s size distribution, at the appropriate wavelengths and assuming the composition described above, using Mie scattering code. The Mie calculations used in the analysis of this work use the results of Bohren and Huffman (1983) that were adapted for use in MATLAB by Mätzler (2002).

For the pure sulfate and mixed sulfate aerosol the particles were assumed to be spherical and, for particles with a smoke core, the core was assumed to be optically identical to the sulfate. It is suspected that the cores are often dissolved in the sulfuric acid, so the approximation may not be significant. The complex component of the index of refraction was also assumed to be zero because of the lack of data for the smoke particles at this wavelength. Thus, the optical calculations of the modeled sulfate aerosol ignore absorption, which may cause an over-estimate of their true volume backscatter coefficient. The real component was allowed to vary as a function of the mass percent of \( \text{H}_2\text{SO}_4 \) present within the aerosol. Data for this dependence is found in the work of Palmer (1975). A fit was applied to the data so that a continuous function for the index of refraction as a function of the mass percent of \( \text{H}_2\text{SO}_4 \) could be made and used to
determine the index of refraction for the complete range of concentration values found in the aerosol within the model. The index of refraction of the aerosol should also be a function of temperature and pressure but for the wavelengths of the observations examined here (532 nm, 525 nm and 1037 nm) the variation is small compared to that caused by the variation in H$_2$SO$_4$ content (Redemann et al., 2000; Massie, 1994; Myhre et al., 2003; Muller et al., 1999; Zhao et al., 1997).

To convert the model output into volume backscatter coefficient and extinction coefficients, the integral for calculating the volume backscatter coefficient over a continuous size distribution of particles (Equation 15) was converted into a Riemann sum to account for the discrete number of radius bins used in the model (Equation 16). The calculation was done for each type of aerosol element separately and combined later.

\[
\beta(\lambda, z) = \pi \int_0^\infty r^2 Q_{ext}(\tilde{m}, x)n(r, z)dr \quad (15)
\]

\[
\beta(\lambda, z) = \pi \sum_{n=1}^{36} r_{bin}^2 Q_{ext}(\tilde{m}, x)n(r_{bin}, z)dr \quad (16)
\]

The size distribution \( n(r_{bin}, z) \) is calculated using the concentration of each of the 36 radius bins used for each of the three types of aerosol. In both equations \( x \) is the size parameter, defined as \( x = \frac{2\pi r}{\lambda} \), and \( \tilde{m} \) is the complex index of refraction used in the Mie calculation of the scattering efficiency \( Q \). The volume backscatter coefficient efficiency \( Q_\pi \) (Equation 17) is a special case of the extinction efficiency, \( Q_{ext} \) (Equation 18), for a scattering angle equal to \( \pi \) and may be defined using only the size parameter and a series of the a and b Mie coefficients (Mätzler, 2002; Bohren and Huffman, 1983).

\[
Q_\pi = \frac{1}{x^2} \left| \sum_{n=1}^{\infty} (2n + 1)(-1)^n (a_n - b_n) \right|^2 \quad (17)
\]

\[
Q_{ext} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n + 1) Re(a_n + b_n) \quad (18)
\]
Here $n = \infty$ is approximated with $n_{\text{max}} = x + 4x^{1/3} + 2$ as suggested by Bohren and Huffman (1983).

To aid the speed of these calculations, a look up table of scattering efficiency terms, $Q_{\text{ext}}$ and $Q_{\pi}$, was compiled as a function of the sulfate particle size parameter and index of refraction for the range of values represented in the model output data. Values of $Q_{\text{ext}}$ and $Q_{\pi}$ at three different indices of refraction are shown in Figure 32 and 33. The sulfate look up table has $Q$ values corresponding to 10000 radius size parameters and 107 indices of refraction corresponding to the mass percent of sulfuric acid in the particle. For the actual scattering calculation of the sulfate aerosol the closest scattering efficiency term above and below the actual size parameter and above and below the actual mass percent of $\text{H}_2\text{SO}_4$ of the aerosol were averaged to reduce errors.

A similar method is employed for the meteoritic smoke particles. Unlike for the sulfate aerosol, a single index of refraction, $2+1.2i$, was used for all the meteoritic smoke calculations, as described in the section discussing the aerosol composition, to create a look up table of $Q$ values (Figure 34). For these calculations, the two closest values of corresponding particle radius are used to find $Q$ values that are then averaged and used in the scattering calculation.
Figure 32: Extinction efficiency ($Q_{ext}$) coefficient for three indices of refraction representing the full range of values used in the sulfate aerosol extinction calculations.

Figure 33: Volume backscatter coefficient ($Q_{\pi}$) coefficient for three indices of refraction representing the full range of values used in the sulfate aerosol extinction calculations.
The scattering calculations are performed for monthly average values for all latitude, longitude and levels of the model. The individual volume backscatter coefficient components were then summed (Equation 19) and the results from the closest model grid box to the location of each lidar were used for the comparison.

\[
\beta(\lambda, z)_{Total} = \beta(\lambda, z)_{Pure Sulfate} + \beta(\lambda, z)_{Mixed Dust–Sulfate} + \beta(\lambda, z)_{Dust}
\]  

(19)

Figure 34: Extinction and volume backscatter coefficient efficiency coefficient used for the scattering calculations involving meteoritic smoke.
3.5 Baseline Model Run Comparison to Lidar Observations

For comparison to the lidar observations, the baseline model was run for 15 years to allow adequate time to reach equilibrium and have ten years to create a climatology over an equal period of time to that of the lidar data. This was done so that the variability could be equivalently compared. The climatology for the lidar observations and model output was created by making monthly means of the data. These monthly means were then used to create a climatological year that was used for the comparison below.

3.5.1 Model Comparison to Lauder, NZ

![Lauder Seasonal Profile Comparison](image_url)

Figure 35: Seasonal mean aerosol volume backscatter coefficient profiles from the lidar and model over Lauder, NZ. Error bars represent two standard deviations of the lidar observations.

Overall, the seasonal means of the model and the lidar data at Lauder agree within two standard deviations of the variability in the lidar observations (Figure 35). Figure 36 and 37 show the monthly mean volume backscatter coefficient for the entire year. In these Figures, both model and data show that the time of peak volume backscatter coefficient retreats as altitude...
increases. They also agree that the peak in the aerosol concentration occurs in September and October.

In the 25 to 30 km region, WACCM tends to have slightly higher volume backscatter coefficient than the data. The increased aerosol in the model is most likely due to over estimation of the amount of meteoritic smoke, or an error in the optical properties of the smoke, which results in too much volume backscatter coefficient. It could also be due to an overestimation in the assumed molecular profile used to retrieve the lidar data. The error could also result in overestimation of the aerosol backscatter volume coefficient if the molecular profile was underestimated. The most distinct difference occurs above 35 km. The departure is either due to the signal to noise limit of the lidar or underestimation of the calibration volume backscatter coefficient ratio. At these altitudes, the aerosol volume backscatter coefficient is less than 1% greater than Rayleigh scatter so the ability of the lidar retrieval algorithms to discern aerosol from the total volume backscatter coefficient becomes questionable. The issue of low signal to noise is especially important in Lauder where the background aerosol loading is smaller than at the other two sites in this study.
4.2 Comparison of Lidar and Model

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Figure 36: Mean monthly lidar aerosol volume backscatter coefficient from Lauder, NZ.

Figure 37: Mean monthly WACCM aerosol volume backscatter coefficient over Lauder, NZ.
3.5.2 Model Comparison to Mauna Loa, HI

Figure 38: Seasonal mean aerosol volume backscatter coefficient profiles from the lidar and model over Mauna Loa, HI. Error bars represent two standard deviations.

The seasonal means of the model and lidar data over Mauna Loa are within the lidar’s expected variation between 22 km and 28 km (Figure 38). Figure 39 and 40 show the monthly mean volume backscatter coefficient for the entire year above Mauna Loa. Figure 39 and 40 show similar seasonal variation in aerosol concentration but the distribution of aerosol is quite different. The largest discrepancy is the fact that the model shows a much larger extent of aerosol into the upper stratosphere than the observations, especially in the summer.

The larger model values of volume backscatter coefficient above 22 km may be attributed to overestimating the production of sulfur from OCS, which is the main source of sulfur at these altitudes in the tropics or overestimation of the volume backscatter coefficient by the meteoritic smoke. The lidar could also be underestimating the observations due to an over-appropriation of the observed scatter to the molecular atmosphere.
Figure 39: Mean monthly lidar aerosol volume backscatter coefficient from Mauna Loa, HI.

Figure 40: Mean monthly WACCM aerosol volume backscatter coefficient from Mauna Loa, HI.
The most distinct difference in the model and data occurs below 22 km. Below 17 km, the difference may be attributed to tropospheric aerosol that the lidar observes but are not included in the model. It is known from measurements by Murphy et al. (2007) that approximately 60% of the aerosol in the upper troposphere and lower stratosphere are organics, which are currently not included. Above the tropopause, the model would be influenced to a lesser extent by missing aerosol in the upper troposphere. However, doubling the mass will also make the particles larger, which in turn would increase their scattering cross section significantly since these particles have optical size parameters less than one.

3.5.3 Model Comparison to Boulder, CO

Overall, the Boulder comparison in Figure 41-43 is very similar to that of Mauna Loa. The model and the lidar data over Boulder agree within two standard deviations of the lidar between 22 km and 30 km but large departures are seen above 30 km and below 22 km (Figure 41). Figure 42 and Figure 43 show a similar seasonal peak in the winter but the model indicates much higher aerosol in the upper stratosphere. The over-estimation of aerosol at Boulder, like Mauna Loa, is especially significant in the summer.

Above 30 km, like the Mauna Loa comparison, the model overestimates the amount of volume backscatter coefficient and this is either due to the over-estimation of the contribution of meteoritic smoke to the total volume backscatter coefficient in the model or the lidar profiles are biased due to an underestimation of the volume backscatter coefficient within the calibration range of the lidar.

The largest discrepancy occurs below 22 km. Unlike the other two sites, the underestimation of the model could not totally be because of tropospheric aerosol because it is too far above the tropopause. Moreover English et al. (2011) find that their model, which simulates the
aerosol layer using only sulfate particles, matches the SAGE II data down to the tropopause. The difference between the two model comparisons with observations needs further examination.

Figure 41: Mean annual volume backscatter coefficient profiles from the lidar and model over Boulder, CO. Error bars represent two standard deviations.
Figure 42: Mean monthly lidar aerosol volume backscatter coefficient from Boulder, CO.

Figure 43: Mean monthly WACCM aerosol volume backscatter coefficient from Boulder, CO.
3.5.4 Discussion of Baseline Model Run Comparison to Lidar Observations

The model simulations compared well to the lidar observations at all three sites between 20 and 30 km. Below 20 km the observations at all three sites were higher than the model output. This discrepancy is mostly due to the fact that the model omits all non-sulfate tropospheric aerosol that could make up as much as 60% of the amount of aerosol below 20 km (Murphy et al., 2007). At Mauna Loa and Boulder the lidar observations were lower than the model simulations above 30 km, while Lauder tended to be slightly higher. These differences are thought to be due to either errors simulating the meteoritic smoke or in the lidar retrievals. Errors in the upper portion of the profile may be propagating through the retrieval and causing biases down low as well.

As for the differences above 30 km, English et al. (2011) show that a similar stratospheric aerosol model using only sulfates to model the stratospheric aerosol greatly underestimates the volume backscatter coefficient above 30 km. Comparison of the model output from this work with the results from English et al. (2011) and other supporting observations of meteoritic smoke indicate the need for meteoritic smoke when simulating the stratospheric aerosol layer.

In Figure 44, the vertical profile of the fraction of the aerosol containing smoke from annually averaged model simulations is compared to the observations presented by Murphy et al. (2007). The in situ measurements reported by Murphy et al. (1998, 2007) suggest that the meteoritic component of stratospheric aerosol increases with height. These observations come from three aircraft measurement campaigns flown from Houston, TX, Key West, FL and San Jose, Costa Rica onboard the NASA WB-57F, which we averaged. Model output is averaged over a similar geographic region as the observations. The measurements represent the fraction of the total number of particles that have positive ions from both sulfate and meteoritic smoke (as defined by Murphy et al., 2007). The model fraction is derived from the total number of particles
contained in the mixed sulfate-smoke size bins compared to the total number of aerosol (only particles with wet radii larger than 300 nm were counted from the model in order to match the detection ability of the instruments used by Murphy et al., 2007). The comparison displays overall agreement and demonstrates the model’s ability to capture the increasing importance of meteoritic smoke with height and the complexities of the interactions of meteoritic smoke with sulfate aerosol.

Figure 45 provides further evidence for the existence of meteoritic smoke in extinction measurements and for the ability of this version of WACCM/CARMA to correctly model its distribution. In Figure 45, the observations originate from the study by Hervig et al. (2009), which uses measurements from the Solar Occultation for Ice Experiment (SOFIE) on board the Aeronomy of Ice in the Mesosphere (AIM) satellite. A zonal mean profile from observations from all of June 2008 and a single profile from June 20, 2008 taken from 65.2° S are compared to a zonal mean of 10 years June monthly mean output from 66° S to 62° S. The index of refraction of the meteoritic smoke in the simulations shown here is based on the work of Hervig et al. (2009) so it is not surprising that the absolute values of the model output compare well to the SOFIE observations. It is more important for the comparison in Figure 45 to note that the shape of the three extinction profiles matches well because the shape is dependent on the distribution of aerosol concentrations, which is what the model is attempting to simulate correctly, rather than the composition of the aerosol.
Figure 44: Comparison of the fraction of mixed sulfate-meteoritic smoke aerosol to total aerosol by number between the model and observations reported by Murphy et al. (2007).
In Figure 46, model calculations that include and exclude meteoritic smoke are compared to SAGE II aerosol extinction retrievals averaged from 2000 to 2005 in the tropics (20° S to 20° N) and northern mid-latitudes (45° N to 70° N). Both sets of model output compare well to SAGE II observations between 20 and 30 km. The agreement from 20 to 30 km suggests that sulfates dominate aerosol extinction in this region. However, above 30-35 km the two models diverge beyond the standard deviation of the observations in the mid-latitudes. The model including the smoke source much more closely resembles the observations above 30-35 km. The model without smoke diverges from the SAGE II observations at a lower altitude in mid-latitudes than in the tropics. (The southern hemisphere follows similar trends to the northern hemisphere, but is on average 15% lower in the amount of extinction caused by aerosol.)

The latitudinal dependence of meteoritic smoke above 30 km is further illustrated in Figure 47, which depicts the zonal averaged percentage of extinction due to meteoritic smoke.
compared to the total amount of extinction due to all aerosol in the model (meteoritic, mixed sulfate-meteoritic, and pure sulfates). Poleward of 30 degrees in latitude, the role of meteoritic smoke above 35 km is significant. In the tropics, meteoritic smoke dominates above about 40 km. Of course the smoke also modifies the extinction due to the mixed particles, which is not included in 7. As can be judged from Figure 46, adding smoke changes the extinction at mid-latitudes by about a factor of 2 at 25 km. Much of the change comes from the impact of meteoritic smoke particles on the observable properties of the sulfate aerosol. The results of English et al. (2011) also lead to the conclusion that the discrepancy observed in the comparison above is due to the lidar retrieval, which does not take the contribution of smoke into account within the calibration region.

A comparison of annual mean profiles from the lidar, SAGE II and WACCM datasets shows agreement within the standard deviation or error of each up to 30 km over each lidar location (Figure 48-Figure 50). Above 30 km, the volume backscatter coefficient due to meteoritic smoke becomes relatively more important and there are large deviations of the lidar data from the SAGE II and WACCM profiles at Mauna Loa and Boulder. Comparison of SAGE II and the lidar profiles to WACCM profiles with the meteoritic smoke component removed (Figure 51-Figure 53) shows a much higher agreement with the lidars above 30 km over Boulder, CO and Mauna Loa, HI. The comparison with SAGE II suggests that the retrievals of the Boulder and Mauna Loa data may be underestimating the amount of aerosol volume backscatter coefficient present in the upper stratosphere. The biases caused by not including meteoritic smoke in the lidar retrievals are further discussed in Section 3.6.
Figure 46: Globally averaged model aerosol extinction profiles (525 nm) are compared to aerosol extinction profiles (total extinction profiles with Rayleigh scattering subtracted) from Stratospheric Aerosol and Gas Experiment (SAGE) II satellite retrievals (Chu et al., 1989) at a wavelength of 525 nm. The SAGE data are averaged from years 2000–2005. Simulations are 10-yr averages. WACCM data points have been converted from a vertical model pressure grid to geometric altitudes (using model derived temperature profiles) for the comparison. The Rayleigh scattering lines are shown as a comparison for the amount of scattering caused by the molecular atmosphere.

Figure 47: Percentage of modeled zonal average total aerosol extinction at 532 nm due to meteoritic smoke alone. This was calculated by taking the ratio of the extinction caused by meteoritic smoke particles alone to the total amount of extinction from all aerosol (meteoritic smoke plus mixed and pure sulfates) included in the model. Average comes from 10 years of model run time.
Figure 48: Annual mean monthly total extinction profiles from the lidar, SAGE II, and WACCM with meteoritic smoke over Lauder, NZ.

Figure 49: Annual mean monthly total extinction profiles from the lidar, SAGE II, and WACCM with meteoritic smoke over Mauna Loa, HI.

Figure 50: Annual mean monthly total extinction profiles from the lidar, SAGE II, and WACCM with meteoritic smoke over Boulder, CO.
Figure 51: Annual mean monthly total extinction profiles from the lidar, SAGE II, and WACCM without meteoritic smoke over Lauder, NZ.

Figure 52: Annual mean monthly total extinction profiles from the lidar, SAGE II, and WACCM without meteoritic smoke over Mauna Loa, HI.

Figure 53: Annual mean monthly total extinction profiles from the lidar, SAGE II, and WACCM without meteoritic smoke over Boulder, CO.
While the focus of this discussion has been on the discrepancy above 30 km, English et al. (2011) show that the models also tend to depart from the data below 20 km. There are several reasons for these departures as discussed by English et al. (2011). The observations presented by Murphy et al. (2007) suggest that differences between models and observations from SAGE II and lidar in the section of the profile from the tropopause to 20 km could be due to the exclusion of carbonaceous aerosol in the model. Carbonaceous aerosol has tropospheric sources and may account for a significant fraction of the total aerosol near the tropopause.

3.6 Discussion of Lidar Retrieval Error

In the course of the baseline comparison an error was identified in the lidar observations that has implications for all lidar observations of stratospheric aerosol volume backscatter coefficient. The error in the aerosol retrieval is due to an assumption that the region above 35 km is aerosol free and could be used as a pure molecular calibration region for the lidar. By underestimating the aerosol present in the calibration altitude (represented as the $LBSR_{min}$ term in Equation 6), the retrieval effectively removes the component of volume backscatter coefficient due to meteoritic smoke.

A second possibility for the discrepancies observed in the comparison above is an error in the molecular model used to separate the molecular scatter from the aerosol scattering in the Mauna Loa and Boulder lidars. Both type of errors will be discussed below. A simulation of lidar stratospheric aerosol retrieval will also be discussed to help elucidate how these two types of errors impact the retrieval of information about the aerosol layer differently.

3.6.1 The Impact of Meteoritic Smoke on Lidar Retrievals

In Figure 54, modeled aerosol volume backscatter coefficients and derived lidar aerosol volume backscatter coefficients are compared to demonstrate how neglecting the impact of
meteoritic smoke can lead to an underestimate of scattering due to aerosol. The model derived Rayleigh volume backscatter coefficient profile is included to demonstrate the total amount of volume backscatter coefficient expected in an aerosol-free atmosphere. The lidar profiles are mean profiles taken from data spanning from 2000 to 2010. Modeled profiles come from an average of ten years of model simulation.

![Graph showing comparison of aerosol volume backscatter coefficient](image)

Figure 54: Comparison of the NOAA Mauna Loa, HI lidar aerosol volume backscatter coefficient (total volume backscatter coefficient minus Rayleigh volume backscatter coefficient) averaged from 2000 to 2010 to mean aerosol volume backscatter coefficient profiles derived from 10 years of model simulations, with and without the meteoritic smoke source, taken from the model grid box closest to Mauna Loa. Error bars represent 1σ standard deviation. The arrow represents the altitude where an 8% adjustment to the calibration scattering ratio was made to the original lidar data to account for the presence of meteoritic smoke. Adapted from Neely et al. (2011b).

In Figure 54, the original lidar profile most closely resembles the modeled profile without smoke emissions. The model aerosol volume backscatter coefficient including smoke exceeds the original lidar-retrieved aerosol volume backscatter coefficient above 25 km, suggesting the current retrieval underestimates the amount of aerosol scattering. The result is a consequence of
the lidar aerosol retrieval method. The procedure used by the Mauna Loa aerosol lidar to separate the aerosol component of the volume backscatter coefficient from the molecular part was pioneered by Fernald et al. (1972) and Klett (1981) and is in wide use within the lidar community (Russell et al., 1979; Thayer et al., 1997; Hofmann et al., 2003; Pappalardo, G. et al., 2004). The method uses an altitude range near the top of the raw lidar volume backscatter coefficient profile (determined by a trade off between signal strength and where aerosol impacts were thought to be negligible) to provide a reference point for a molecular density profile derived from a model or other observations. The adjusted Rayleigh profile is then projected down through the lower altitudes of the volume backscatter coefficient profile and any deviations of the actual lidar profile from the calibrated Rayleigh profile may be attributed to aerosol. Russell et al. (1979) showed how small deviations in the top of the profile used to derive the molecular profile, attributed to aerosol scattering, can cause definite biases in the aerosol profiles. Russell et al. (1979) also show that including a small correction to account for additional aerosol scattering at the calibration altitude can improve the retrieved aerosol profiles, as compared to those obtained when the topside calibration altitude is erroneously assumed to be aerosol free. Furthermore, recent observations from CALIOP by Vernier et al. (2009) suggest an increase in aerosol to Rayleigh ratio in the tropical stratosphere that ranges from 2-12% with an average of 6%. Based on these observations and model results, we apply an 8% adjustment to $LBSR_{min}$.

In Figure 54, the impact of the adjustment in the retrieval process as suggested by Russell et al. (1979) and Vernier et al. (2009) is evident. The modeled aerosol volume backscatter coefficient when smoke is included now more closely compares to the lidar observations, suggesting that such an adjustment is required to take into account the effect of meteoritic smoke
at the lidar calibration altitude. Note that the altitude level at which meteoritic smoke dominates the extinction declines with increasing latitude as shown in Figure 47.

3.6.2 Simulation of Lidar Retrieval Errors

Further investigation of the error created by the omission of meteoritic smoke in lidar retrievals was conducted with the use of a lidar simulation code. The code was designed to use output from the WACCM/CARMA baseline model results and simulate a lidar that is representative of the Mauna Loa lidar discussed above. The point of the simulation was to show the difference between errors in $LBSR_{min}$ and errors in the molecular profile used to separate the aerosol backscatter from the total observed backscatter. The lidar signal and error simulations are based on the methodology presented by Russell et al. (1979) and applied to the specific case of retrieval of stratospheric aerosol using lidar system parameters that would simulate signals equivalent to the Mauna Loa lidar system (Barnes et al., 2008).

The simulated lidar retrievals are based on returns calculated from the elastic volume backscatter coefficient lidar equation (Equation 1). Here, $N(\lambda, z, t)$ is the total number of scattered photons detected by the simulated receiver, the wavelength of the transmitted light ($\lambda$) is 532 nm, $z$ is altitude above the surface, $N_L$ is based on a laser transmitter with 500 mJ pulses, $\beta$ and $\alpha$ come from the aerosol volume backscatter coefficient calculated from the profiles of annual mean size distributions and molecular density in the baseline WACCM output in the closest grid square to Mauna Loa, $G$ is ignored for simplicity, $\eta$ is based on standard detector and mirror efficiencies and $N_B$ is modeled after the background signal observed in typical measurements from Mauna Loa. The resulting profile of simulated photons counts is shown in Figure 55.
Figure 55: Simulated raw lidar signals. The simulated lidar performance was based on the raw signals of the Mauna Loa lidar. The blue line represents the signal from one pulse from the simulated lidar system without any simulated background signal. The red line represents the simulated signal with background noise integrated for 1 hour (representative of a typical single data collection period). The dashed green line is the background subtracted integrated lidar signal.

As described in Chapter 1, the molecular volume backscatter coefficient is determined empirically and then the aerosol component may be separated from the total observed volume backscatter coefficient (Elterman, 1964; Russell et al. 1979). As shown in Equations 5, 6 and 7, the separation of the aerosol volume backscatter from the total observed volume backscatter is accomplished by deriving the $LBSR$ and calibrating the lidar in a region with known scattering properties. As was previously noted, small deviations in the calibration factor ($LBSR_{min}$ from Equation 6) attributed to aerosol scattering can easily create large biases in the region near the calibration altitude of the retrieved aerosol profile. Small errors in the molecular profiles used to separate the aerosol scattering from the total scattering can also cause biases in the profile. Unlike errors in the calibration term, errors in the molecular profile bias the entire profile.
because of their impacts on the extinction term ($\alpha$) in the retrieval as well as the scattering term ($\beta$).

Analyses of the errors in these simulated retrievals are based on the work of Russell et al. (1979). The result of the method described by Russell et al. (1979), applied to the specific requirements of a retrieval of stratospheric aerosol, is summarized and applied to the simulated signals in Figure 55 to point out the major sources of error and how they impact the retrieval.

As shown in Russell et al. (1979), by using standard propagation of error techniques the total error in the derived aerosol volume backscatter coefficient profile may be derived from Equations 5, 6 and 7.

$$\left( \frac{\delta \beta_a}{\beta_a} \right)^2 = \left( \frac{\delta \beta_m}{\beta_m} \right)^2 \left\{ \left( \frac{\delta N_z}{N_z} \right)^2 + \left( \frac{\delta q}{q} \right)^2 + \left( \frac{\delta LBSR_{\text{min}}}{LBSR_{\text{min}}} \right)^2 + \left( \frac{\delta \beta_m}{\beta_m} \right)^2 - 2 \frac{c_{\text{mm}}^2}{LBSR \beta_m \beta_m} \right\} LBSR^2 + \left( \frac{\delta \beta_m}{\beta_m} \right)^2 \}$$

In Equation 20, $q$ represents the two way transmission of the transmitted light which is defined using extinction ($\alpha$) in Equation 1 as $\exp[-2 \int_0^z \alpha(\lambda, z') dz']$ and * means the quantity is derived from the calibration altitude. The sources that contribute to the total error may be categorized into four parts. Three of the four sources are directly dependent on altitude while the other (error due to the molecular profiles used in the retrieval) is dependent on independent measurement or model error. For the simulation below the relative contribution of the molecular term is assumed constant for simplicity (Figure 56).

The main source of error at higher altitudes comes from the total amount of signal detected (blue line in Figure 56). This source is also referred to as “shot noise”. For photon counting detectors, as are utilized by the lidars this study, shot noise follows Poisson statistics and is defined as

$$\frac{\delta N_z}{N_z} = \sqrt{\frac{N}{N_z}}. \quad (21)$$
Recall that \( N \) is the total detected photons (signal and background) and \( N_s \) is the background subtracted photon counts. Shot noise is largely determined by the power of the lidar system’s transmitter, length of measurement integration and altitude of calibration. As such, it is easily reduced to minimize the impact of signal error on the retrieval.

The second largest error comes from the assumptions required to calculate the two-way transmission of the transmitted light (green line in Figure 56).

\[
\left( \frac{\delta q}{q} \right)^2 = 4 \left\{ \left[ \delta \tau_a(\lambda, z, z') \right]^2 + \left[ \delta \tau_m(\lambda, z, z') \right]^2 + \left[ \delta \tau_{O_3}(\lambda, z, z') \right]^2 \right\}
\] (22)

The optical thickness uncertainties described in this error term depend on atmospheric variability and the assumptions within the models used to derive the components of the transmission term. Russell et al. (1979) suggest the following values (Equations 23-25) for each of the terms based on analysis of typical models of the atmosphere suitable for use in lidar retrievals of 532 nm light.

\[
\delta \tau_{O_3}(\lambda, z, z') = 0.2 \tau_{O_3}(\lambda, z, z') \] (23)

\[
\delta \tau_m(\lambda, z, z') = 0.5 \tau_m(\lambda, z, z') \] (24)

\[
\delta \tau_a(\lambda, z, z') = 0.1 \tau_a(\lambda, z, z') \] (25)

The third source of error comes from the molecular density profile used within the retrieval. Error in the molecular profile is dependent on whether or not the density is measured with a co-located measurement or is derived from a model.

\[
\frac{\delta \beta_m}{\beta_m} \approx 0.01 \text{ (measured)} \] (26)

\[
\frac{\delta \beta_m}{\beta_m} \approx 0.03 \text{ (modeled and or interpolated)} \] (27)

The final source of error comes from the assumed minimum scattering ratio used in the calibration of the retrieval, \( \left( \frac{\delta LBSR_{min}}{LBSR_{min}} \right)^2 \). The expected errors for this term vary depending on the
part of the atmosphere being measured and the amount of aerosol present. Russell et al. (1979) summarize typical values of error associated with $LBSR_{min}$ when used in lower stratospheric and tropospheric measurements. The values presented by Russell et al. (1979) show the importance of the $LBSR_{min}$ term but are not appropriate for calibration of any lidar observations above 30 km.

Lidar observations of the upper stratosphere have typically neglected to account for any scattering in the $LBSR_{min}$ term due to the low amount of aerosol above 35 km but; as shown by the observations and simulations presented in this work, the assumption that the $LBSR_{min}$ is singular can cause biases in retrievals near the calibration region. As such, $LBSR_{min}$ must be evaluated for each measurement through an independent observation of aerosol scattering.

The total relative error of the aerosol volume backscatter coefficient also contains a covariance term, $C_{mm*}^2$, for molecular volume backscatter coefficient values at different heights, $z$ and $z^*$. $C_{mm*}^2$ is only important for photon counts in the region of the retrieval that are closer to the calibration altitude than any of the data points used to obtain the molecular profile or if the molecular model must be interpolated onto the same vertical grid as the lidar measurement. As such it is not important in my simulation because the molecular profile resolution matches exactly with the resolution of photon counts. The term can also become important if there is a systemic error in the model or measurement. The covariance term is further described by Russell et al. (1979).

The shot noise and total error for the integrated simulated signal are shown in Figure 56. The shot noise and total error are shown as reference for the type of error that is inherent in stratospheric aerosol volume backscatter retrievals and to provide a baseline that the error
resulting in erroneous assignment of $LBSR_{\text{min}}$ or the molecular model may be compared to in Figure 56.

Figure 56: Expected percent error from the simulated integrated lidar signal. The blue line is the error from the shot noise or the detected photon counts. The green line represents the error due to the transmission. The constant magenta line represents the expected error from a typical molecular model. The red line is the expected error due to choosing an appropriate $LBSR_{\text{min}}$ value. The black line represents the total error from the simulated volume backscatter coefficient retrieval.

Using the retrieval method discussed in Chapter 1 and the error analysis described above several different retrieval error cases were examined (Figures 57 and 58). The difference in the profiles with a $LBSR_{\text{min}}$ of 1 versus profiles with a $LBSR_{\text{min}}$ of 1.00345 is notably close to the calibration region. The difference seen in the comparison (using the WACCM/CARMA meteoritic smoke and sulfate aerosol profiles) also corresponds well with the difference in the lidar aerosol backscatter with a change in calibration discussed above in Figure 54. The
sensitivity of the retrieval to the extent seen in Figure 57 is not evident from the discussion of the errors terms above (Figure 56). The sensitivity comes from Equation (7), which may also be written as:

\[ \beta_a(z) = \frac{z^2}{(z^*)^2} \frac{N(z)}{N(z^*)} \frac{T^2(z)}{T^2(z^*)} \beta_m(z^*)LBSR_{min} - \beta_m(z). \]  

(28)

In Equation 28, the direct impact of \( LBSR_{min} \) is more easily recognized. \( LBSR_{min} \) obviously plays a role in the whole profile but it plays a much larger role at the calibration altitude because the first three terms approach unity, leaving only the guess in \( LBSR_{min} \) as the difference between the values of the molecular profile and the derived aerosol profile (Russell et al., 1979). The errors described by Russell et al. (1979) for \( LBSR_{min} \) and the error analysis above (Figure 56) do not capture the bias created by using an inappropriate \( LBSR_{min} \) value because they are both based on standard propagation of error analysis techniques which describe precision, not accuracy. Further error assessments of \( LBSR_{min} \) should include a term similar to the error described by Russell et al. (1979) as well as an additional term to describe the bias.

A distinct difference can also be seen between the retrieval that contains an error in the molecular and ozone profiles and the retrieval that has an error in the minimum scattering ratio. The error in assuming a scattering ratio of 1.0 in the presence of aerosol causes a sharp difference near the calibration region but becomes less significant lower in the profile. The relation of the bias error caused by neglecting scattering in \( LBSR_{min} \) is much different than the shift of the entire profile resulting from errors in the molecular and ozone profiles used in the retrieval. This behavior is also evident from Equation 28.

The sharp bias in the top of the profile from assuming too low a scattering ratio to calibrate the lidar signal causes the retrieved aerosol volume backscatter coefficient to resemble the pure sulfate aerosol profile (Figure 57). Assuming a \( LBSR_{min} \) of unity also slightly biases
the lower part of the profile (Figure 58). As supporting observations from SOFIE and Murphy et al. (2007) have shown, meteoritic smoke is a major component of stratospheric aerosol and causes significant extinction above 35 km. Thus, meteoritic smoke must be taken into account to avoid biases in lidar profiles that would negate its existence.

Figure 57: Aerosol volume backscatter coefficient derived from the simulated lidar signals using different assumptions to make the retrieval. The yellow line is the total aerosol volume backscatter coefficient calculated directly from the WACCM size distributions and molecular profile. The black dashed line is the volume backscatter coefficient from the WACCM molecular profile alone. The blue crosses are the profile of WACCM aerosol volume backscatter coefficient (contain pure and mixed sulfates as well as meteoritic smoke) used to simulate all the lidar signals. The blue circles are shown for reference as an aerosol profile that only accounts for sulfate aerosol. The solid blue profile is the simulated lidar aerosol volume backscatter coefficient assuming a minimum scattering ratio of 1 at the calibration altitude. The green profile is the simulated lidar aerosol volume backscatter coefficient assuming a minimum scattering ratio of 1.00345 at the calibration altitude (the amount needed to account for the scattering of meteoritic smoke at 37 km). The dashed pink line represents a lidar retrieval assuming a minimum scattering ratio of 1.00345 and an error in the molecular and ozone profile.
Figure 58: Percent Difference in aerosol volume backscatter coefficient derived from the simulated lidar signals using different assumptions to make the retrieval from the actual aerosol volume backscatter coefficient similar to the raw lidar signal. The lines correspond to the retrievals in Figure 56.

3.7 Summary

Chapter 3 described and demonstrated WACCM/CARMA’s ability to model the mean state of the background stratospheric aerosol layer. Through comparison with ground based lidar and SAGE II observations, it was determined that the model accurately simulates the bulk of the stratospheric aerosol layer above 20 km.

Below 20 km, because the model used here ignores all non-sulfate aerosol from the troposphere, the comparisons show that the model underestimates the extinction and volume backscatter coefficient. The model compares favorably in the region below 20 km to aircraft observations of aerosol compositions. Specifically, the model correctly simulates the abundance of the meteoritic component of aerosol in the UTLS region.

Above 30 km, the model results compare well with observations from SAGE II but not to the lidar observations. Corroborating evidence from observations of stratospheric aerosol
composition in the lower stratosphere made by Murphy et al. (2007) and extinction of the upper stratosphere and mesosphere from SOFIE compiled by Hervig et al. (2009) (as well as the work of Rosinski and Snow (1961), Hunten et al. (1980), Megner et al. (2008), Bardeen et al., (2008a; b) and English et al. (2011)) indicate the need for the inclusion of meteoritic smoke to correctly model the stratospheric aerosol layer. Thus, it was concluded that an error existed in the lidar retrievals that was causing a bias resulting in the lidar aerosol backscatter to be underestimated. Further analysis of the bias in $LBSR_{min}$ suggested that application of a small correction to the calibration used in the retrieval (that accounts for the scattering of meteoritic smoke at the calibration altitude) would fix the observed biases in the lidar aerosol volume backscatter coefficient profiles.

A full analysis of the impacts on the observed radiative forcing caused by meteoritic smoke and the associated errors it may cause in lidar retrievals is not within the scope of this work. However, it is thought that the heating by micrometeorites will be insignificant compared to other sources of heating in the stratosphere. Based on the recalibration of CALIOP by Vernier et al. (2009), we assume the error in the LBSR from improperly calibrating lidar data is on the order of 8-12% below the calibration region (this translates to an error of 50% to 100% in the aerosol volume backscatter coefficient). The assumption is supported by the lidar simulation (below 34 km in Figure 58). The difference in the CALIOP calibration value also corresponds to the adjustment observed in the recalibration of the lidar profile in Figure 54 needed to match the modeled aerosol.

The initial comparison of the base model to observations shown here suggests that the model is suitable for use in the following numerical experiments to isolate the source of decadal trends. It also emphasizes the importance of accounting for meteoritic smoke to correctly model
the stratospheric aerosol layer and in the retrieval of aerosol backscatter in lidar observations, especially above 30 km. Though the lidar observations shown here have not taken the impact of meteoritic dust into account, the results of analysis in Chapter 2 and the comparisons made with integrated lidar backscatter throughout this work should not be impacted due to the minimal impact of the retrieval in the bulk of the stratospheric aerosol below 30 km.
CHAPTER IV: ANALYSIS OF ANNUAL CYCLES AND TRENDS IN STRATOSPHERIC AEROSOL

4.1 Introduction

In Chapter 2, observations from ground-based lidar and several other instruments provided evidence that the optical depth of stratospheric aerosol increased from 2000 to 2010, agreeing with studies by Hofmann et al. (2009), Nagai et al., (2010), Trickl et al. (2010) and Vernier et al. (2011a). Unfortunately, observations alone have been unable to conclusively partition the source of this increase between volcanic injections and anthropogenic emissions (Hofmann et al., 2009; Vernier et al., 2011a and b). The observations from ground based lidar also indicated that the overall trend in stratospheric AOD is superimposed upon other variability, including annual cycles. Here, WACCM/CARMA is used to help identify the sources of variability in stratospheric aerosol.

For the purposes of this analysis, the variability in stratospheric aerosol is broken into two parts: the annual variability and a decadal scale trend spanning the observations from 2000 to 2010. The annual cycle is analyzed by comparing the lidar signals described in Chapter 2, satellite observations from the SAGE II (1985-2005), GOMOS (2002-2009) and CALIOP (2006-2011) instruments and aircraft data from Wilson et al. (2008) to output from the baseline model described in Chapter 3 (Vernier et al., 2011a). The baseline model is used to analyze the annual cycles because the repeating yearly emissions of the baseline model should provide the clearest signal of the annual cycles in stratospheric aerosol without interference from trends and other perturbations.
To examine the trends, perturbations representative of the increases in anthropogenic emissions and volcanic injections from 2000 to 2010 were applied to the emission scheme of the baseline model. The output from these simulations is compared to the lidar observations included in this work and observations of AOD from SAGE II, GOMOS and CALIOP (Vernier et al., 2011a). Unlike the current observations, this numerical experiment allows for the isolation of the two different sources. The ability to isolate the sources is thought to be the only way to clearly attribute the impact of each source on the stratosphere.

In both of these analyses the atmosphere is more conveniently thought of as three parts: the underworld, the middleworld and the overworld as outlined by Hoskins (1991) and Holton et al. (1995) (Figure 59). The underworld (approximately the troposphere at mid and high latitudes, and the portion of the tropical troposphere with potential temperature below 300K) is defined as the region that contains no isentropes that cross the tropopause. The middleworld is the layer from 300 to 380 K where isentropic mixing can exchange air between the troposphere and stratosphere. The overworld is defined as the region above 380 K where isentropic mixing is exclusively stratospheric. Tracer species enter the overworld through diabatic ascent across the isentropes, determined by large-scale transport via the Brewer-Dobson circulation, mixing processes and chemical production and destruction. Their distribution is largely determined by large-scale ascent through the tropical pipe, removal through the high-latitude middle world and horizontal transport through the mean meridional circulation.
4.2 Annual Cycle Analysis

4.2.1 Validation of Model Results with SAGE II and Aircraft Observations

Niwano et al. (2009) used background (1998-2004) aerosol extinction observations from SAGE II to examine seasonal cycles in stratospheric aerosol. Here baseline model output is compared to the results of Niwano et al. (2009) for validation of the model’s simulation of annual cycles. The comparison is shown in Figure 60. The modeled AOD at 30 km compares favorably to the observations, especially for the northern and southern hemispheres. At 18 km and 19 km, the model captures the phase and magnitude of the annual cycle described by Niwano et al. (2009) but the absolute value of the observed AOD is ~30% lower than the observations. This offset is expected because the model omits all aerosol other than sulfates and meteoritic smoke. The most notable of these missing contributions is carbonaceous aerosol, which can make up a third of the total concentration of aerosol in this region (Murphy et al., 2007). A distinct difference in the annual pattern of optical depth is observed between 30 km
and 18 km at mid-latitudes of the southern hemisphere. The 30 km annual cycle peaks in winter while the 18 km cycle peaks in summer.

Figure 60: Comparison of stratospheric AOD monthly mean climatologies from SAGE II (1998–2004) (Adapted from Niwano et al., 2008) and ten years of model baseline model output. Shading represents one standard deviation of the inter-annual variation. Colored lines and shading are from observations while all black lines are based on model output. Plots with red shading are from 25° S, green shading are from the Equator, and blue are from 25° N. The top three panels are taken from 30 km and the bottom three are taken at 18 km for the northern and southern hemispheres and 19 km at the Equator.
4.2.2 Lidar Observation-Model Comparison Results

In the process of comparing the annual cycles in stratospheric IABS from the lidar observations presented in Chapter 2 to satellite observations and the baseline model a large error was found in the lidar observations at Mauna Loa, HI and Boulder, CO. Figure 2 compares the lidar observations at Mauna Loa, HI to zonal mean model output and satellite observations within 5° of the lidar site. Large annual cycles with winter time peaks are observed in the lidar’s IABS (pink, integrated from 15 to 40 km). The lidar results are also presented in Chapter 2. In comparison, the amplitude of the annual satellite observations and model are typically 50-75% smaller and slightly earlier peaks. The difference is especially evident in 2003 and 2004. (The ability of the satellite instruments to discern the annual cycles from the variability caused by volcanic eruptions will be discussed below.)

Figure 61: Comparison of the zonal mean of three baseline model runs (green) and satellite observations to the Mauna Loa Lidar observations (pink) from 2000 to 2010 integrated from 15 to 40 km (model and satellite observations taken from within 5° of Mauna Loa, HI). The model output was converted to aerosol volume backscatter at 532 nm, as described in Chapter 3, to match the lidar observations. The satellite data was converted from aerosol extinction to aerosol volume backscatter using a extinction to backscatter ratio of 50 sr ((Jäger and Deshler, 2002; 2003).
The discrepancy in the observations led to an re-examination of the Mauna Loa lidar retrieval in comparison to the retrieval used by CALIOP. The result of this comparison is shown in Figure 62. Below 33 km, the NOAA air density is ~98% of the density used in the CALIOP retrieval. And above 33 km, in the summer, a similar ratio is observed. In winter, there is a sharp discontinuity at 33 km. This height also corresponds with the region where data from radiosondes launched from Hilo, HI are spliced to air densities taken from the MSIS model to create the full molecular profile needed to separate the aerosol volume backscatter from the total observed volume backscatter (Personal Communication, John E. Barnes).

The large part of the discrepancy in Figure 61 is caused by a discontinuity in the molecular profile used by the Mauna Loa aerosol retrieval at 33 km in the winter. The discontinuity causes an error similar in nature to, but larger than, the bias created by meteoritic smoke in the lidar calibration region discussed in Chapter 3. The result of the error in the molecular profile is a bias in the annual signal of aerosol volume backscatter coefficient at Mauna Loa. Similar results are also found for the Boulder lidar because it uses a related retrieval scheme (Personal Communication, John E. Barnes and Jean Paul Vernier). Due to the bias created by the error in the lidar retrieval used at Mauna Loa, HI and Boulder, CO further direct comparison of the annual cycle to the lidar data and the model is not done. It should also be noted that the description of the annual cycle in the lidar data in Chapter 2 is biased due to this error in the retrieval. The observed difference in the lidar data may be fixed by using an appropriate molecular model but is not within the scope of this work to address.
4.2.2 Comparison of Model Results with Satellite Observations

The model was shown to capture the annual cycle in stratospheric aerosol in the previous section. Here, the model is used as comparison to identify annual cycles in the observational record, which may currently be obscured by other sources of variability. In Figure 63, the tropical overworld is examined. The model depicts an annual cycle of ~10% of the total AOD from 20 to 30 km with a peak in the late summer and fall. The observations in the tropical overworld exhibit large variability due to volcanic injections. The volcanic increases obscure any smaller variation in the observations that may be due to annual cycles. Analysis of Figure 64 (same as Figure 63 but the integration of AOD has been extended down to 15 km) reveals a similar result, though the model estimates the amplitude of the annual cycle to be as much as 25% of the total AOD integrated from 15 to 30 km. The difference in the amplitude exhibited by the model in Figure 63 and Figure 64 suggests that the amplitude of the annual cycles decreases with height as seen in lidar data in Chapter 2. The cycle’s dependence on height is also in agreement with the observed tracer behavior of aerosol in Wilson et al. (2002) and Niwano et al. (2009).
Figure 63: Tropical (20° S to 20° N) stratospheric AOD integrated from 20 to 30 km. Three baseline model runs are shown in green and the satellite record is in black.

Figure 64: Tropical (20° S to 20° N) stratospheric AOD integrated from 15 to 30 km. Three baseline model runs are shown in green and the satellite record is in black.

Figure 65 and Figure 66 show the comparison of observed and modeled AOD in the northern and southern mid-latitudes (30° N/S to 50° N/S) integrated from 15 km to 30 km. The annual cycle in the model has peaks in local winter and the annual amplitude for both hemispheres is 30-40% of the total AOD. At mid-latitudes in both hemispheres, from 2000 to 2004, (when there was a minimum in volcanic activity) the observational record exhibits behavior similar to the baseline model. At the beginning of 2009 a low in AOD is also observed. The low corresponds to the minimum in the annual cycle in the model. The coincidence of the two minima also suggests that the annual oscillations in transport plays a role
in the variability of aerosol in the lower mid-latitude stratosphere in the presence of mid-latitude and polar volcanic eruptions. However, tropical volcanic injections perturb the annual signal sufficiently to cause the annual cycle to be obscured in the measurements. The impact of annual variability in the mid-latitudes is further suggested by Figure 66. Because the volcanic eruptions from 2008 to 2010 were located in the northern hemisphere, the observations of the southern hemisphere (Figure 66) show an annual cycle similar to the model; differing from the northern hemisphere except during periods of high tropical volcanic loading.

The difference in the phase of the peak AOD observed in the mid-latitudes versus the tropics is indicative of the transport of aerosol from the tropics to the mid-latitudes. The difference in amplitudes between the tropics and mid-latitudes is explained by greater variations in the meridional transport process at higher latitudes, especially in the northern hemisphere, as discussed by Holton (1995). The mean background (2000 to 2004) AOD in the northern hemisphere (Figure 65) is also noted to be slightly lower than the mean background AOD in the southern hemisphere (Figure 66). The difference in AOD is thought to be related to the differences in transport in the two hemispheres and the preferential downwelling of the meridional transport in the northern hemisphere (Holton, 1995). The difference in the “background” of the two hemispheres and its relation to annual transport of aerosol should be further examined because it may have an impact on long-term trends.
Figure 65: Northern mid-latitude (30° N to 50° N) stratospheric AOD integrated from 15 to 30 km. Three baseline model runs are shown in green and the satellite record is in black.

Figure 66: Southern mid-latitude (30° S to 50° S) stratospheric AOD integrated from 15 to 30 km. Three baseline model runs are shown in green and the satellite record is in black.

4.2.3 Summary of Annual Cycle Analysis

Here the annual variability of stratospheric aerosol has been explored using baseline output of WACCM/CARMA. The model was found to compare well with observations from SAGE II and capture the tracer behavior of stratospheric aerosol observed by Wilson et al. (2002). This validation led to a comparison of the observed time series in stratospheric AOD from the lidars in Chapter 2 and satellites to baseline model output in order to identify seasonal cycles in the observational record.
The lidar comparison revealed an error in the lidar retrieval that led to a bias in the seasonal cycle. The comparison with lidar data also shows the sensitivity of aerosol measurements with lidar. Care must be taken in lidar retrievals to not bias annual variability with cycles in the molecular model used to separate the aerosol volume backscatter from the total observed volume backscatter.

Comparisons of model results with the satellite record suggest that in the tropics, above 20km, a small annual cycle exists but it is not revealed in the satellite record because the aerosol variability is obscured by episodic volcanic injections. Comparison with satellite observations suggested a seasonal cycle does exist in the northern and southern hemisphere satellite data records but is episodically hidden by volcanic injection of aerosol. It is thought that the cycle is more easily observed at higher latitudes due to fewer volcanic perturbations and the larger annual variations in meridional transport.

As noted in Chapter 3, this model does not couple the aerosol formed in CARMA to the radiation code of WACCM. Thus, WACCM’s atmosphere is not heated by the aerosol shown here and will not react accordingly. The changes caused by aerosol heating may impact the results of this work, especially in regions of high aerosol loading, and should be explored before attribution of the mechanism of the annual cycle in stratospheric aerosol can be fully clarified. The comparison of annual cycles in the observations and model does indicate that the variability observed in stratospheric aerosol is largely determined by volcanic injections in the tropics. In the mid-latitudes, the comparison shows that the increased aerosol from volcanic injections largely obscures the annual cycle but in particularly volcanic quiescent periods, such as 2000 to 2004, the annual cycle plays a role in the observed variability.
4.3 Numerical Experiments to Explore Decadal Trends

To explore the decadal trends (Chapter 2) of stratospheric aerosol the baseline model was used to conduct numerical experiments to examine the response of the stratospheric aerosol layer to different emissions. The emission schemes simulated in this experiment include the increasing anthropogenic emissions from China and India and the volcanic injections observed from 2000 to 2010. These two sources of SO\(_2\) were used because they represent the current leading theories that attempt to explain the trends. This section will describe the methodology of the experiment, the methods used to derive and implement the two emissions schemes and the results of the model runs.

There are two main theories that have attempted to attribute the trends to sources. The first was the proposal by Hofmann et al. (2009). In this work it was argued that recent increases in Asian SO\(_2\) injected into the stratosphere by the Asian monsoon and by other convection in the tropics could lead to the observed trends. The second theory, illustrated by Vernier et al. (2011a), suggests that the trends are the result of the combined ejections by the string of moderate volcanic eruptions from 2000 to 2010.

Figure 67 and Table 2 summarize the methodology of this experiment. To start the model simulations, the baseline model was allowed to “spin-up” to equilibrium by running it for five years (model years “1995” to “2000”). The output from the end of the spin-up model run was used as the initial conditions for all the model runs in the experiment. The runs to be used in the comparison consist of 1) 3 baseline model runs, 2) two runs with increased anthropogenic emissions over China and India, 3) one run with ten times (10x) the amount of observed anthropogenic emissions and 4) a run with volcanic injections representative of the eruptions from 2000 to 2010. All simulations ran from model years “2000” to “2010”. Because WACCM is not forced to match observations of dynamics from 2000 to 2010 (i.e. the model is free
running) the best result that can be hoped for in the comparison shown here is a similarity in variability from one of the modeled emissions schemes to the observed variability.

Figure 67: Schematic representing the model runs done for the numerical experiment to attribute the trends in aerosol to emissions.

<table>
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<th>Model Run</th>
<th>Emissions</th>
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<tr>
<td>Anthropogenic</td>
<td>Base + Chinese and Indian Trend from Lu et al. (2010)</td>
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<td>10x Anthropogenic</td>
<td>Base + Ten times Chinese and Indian Trend from Lu et al. (2010)</td>
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<tr>
<td>Volcanic</td>
<td>Base + Injections from volcanoes listed in Table 3 and Vernier et al. (2011a)</td>
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Table 2: Definition of labels used to describe the three types of model runs done to explore the impact of various sources on trends in stratospheric aerosol.

4.3.1 Model Inputs

Central to the experiment are the sources of SO₂, which lead the two theories attempting to explain the increases in aerosol. To that end, great care was taken to accurately depict the contribution of SO₂ from both. In the sections below (4.3.1.1-2) the sources of data for the anthropogenic emissions and the volcanic injections are described in detail.
4.3.1.1 Increased Anthropogenic Surface Emissions

To isolate and represent the trends in anthropogenic emissions discussed by Hofmann et al. (2009) an emission scheme was created that added the increases in Chinese and Indian SO$_2$ to the baseline model emissions (year 2000 repeating) (Smith et al., 2011; Lu et al., 2011). The increases added were the result of observations and inventories described by Lu et al. (2011). To create the increased emissions, the grid squares representing the industrial regions in China and India are multiplied by a factor at each time step that results in a total annual emission from 2000 to 2010 described by Lu et al. (2011). Once this model input was created, an additional scheme was simulated with the model where the additional Chinese and Indian emissions were multiplied by a factor of ten in order to discern possible future anthropogenic impacts. The additional (above 2000 levels) global annual emission of SO$_2$ for the two model schemes and the data on which the schemes were based is shown in Figure 68.

Figure 69 is shown as a comparison of the resulting vertical distribution of SO$_2$ from a baseline model run, an anthropogenic model run and the 10x anthropogenic emission run. The simulated Manam volcanic injection is also shown as a juxtaposition of the emission types (surface source versus direct stratospheric injection). The anthropogenic emissions elevate the SO$_2$ burden from 4 km to 18 km by ~20%. Above 18 km, no difference in the SO$_2$ burden is made by the anthropogenic emissions. The 10x anthropogenic emissions run increases the lower tropospheric burden even further but matches the actual anthropogenic emissions above 16 km. The volcanic injection does not influence the SO$_2$ profile except in the region of the emission and directly below. Above the forced injection region (~22 km) the SO$_2$ concentrations are undisturbed. Below 10 km, the volcano has no impact.
Figure 68: Observations of anthropogenic emissions (Lu et al. 2011) and model input from China and India of SO\(_2\) above year 2000 emissions documented by Smith et al. (2011).

Figure 69: Zonal, monthly mean SO\(_2\) profiles at the equator during model month 61 (January, 2005 and also month of Manam eruption). The volcanic profile is taken from over the grid
square containing Manam on the day of the eruption. The black dashed line represents Manam eruption modeled as closely to observation parameters as the model grid will allow (this profile is taken from the grid square containing Manam). The black solid line is from a baseline emission model run. The blue line is from an anthropogenic model run. The red line is from the simulation with ten times the observed anthropogenic SO$_2$ emitted.

### 4.3.1.2 Volcanic Injections

To simulate the volcanic eruptions from 2000 to 2010 that were found to impact stratospheric aerosol formation, an inventory was made of all the volcanoes with volcanic explosivity index (VEI) of 3 or greater during this period. The information used to model the volcanoes was gathered from various publications on each eruption, which contained information about the height and the amount of SO$_2$ of each injection as well as from satellite observations. The summary of the data for each volcano may be found in Table 3. Using this information, each volcano was modeled using observations of injection height and total emitted SO$_2$. Each injection was modeled at the closest corresponding model grid square to the actual volcano’s location as a Gaussian shaped plume with a maximum emission at the model level most closely matching the observed injection height and the full injection spread over 5 model levels (~4-6 km). This formulation of modeling each volcano is supported by the recent observations by Doeringer et al. (2012). This work shows that the majority of the SO$_2$ produced by the Sarychev eruption in 2009 forms a Gaussian shaped plume centered slightly above the region of highest observed aerosol extinction observed just after the eruption. The simulated Manam injection is shown as an example in Figure 69.
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Table 3: Volcanic emission database used for simulated eruptions in the model runs done as part of the numerical experiment. Parameters were derived from information gathered by the Global Volcanism Program maintained by the Smithsonian Institution and the citations shown.
As part of the effort to build the volcanic emission database and simulate each volcano accurately within the framework of the model, an analysis of the sensitivity of the model to variations in the volcanic injections was done. The analysis was done by bracketing the simulated Manam eruption based directly on observations with injections one model level higher and one model level lower as well as injections at the same model level but with double and half the observed SO$_2$ emission. Each simulation was started with the same initial conditions from a previous baseline run. The results from this analysis are shown in Figure 70 and 72.

Figure 70 and 72 show the resulting zonal mean tropical AOD from 15 to 20 km (middleworld) and 20 km to 30 km (overworld) for each of the simulated injections. Obvious in both plots is that the AOD is linear in the injection magnitude. Several studies have shown that the AOD is generally not linear with injected mass for large injections because as mass is added the particle radius increases (Pinto et al., 1989; English et al., 2012). With larger radius there is generally lower optical depth per unit mass, and a shorter lifetime. However, here we are dealing with very small injections, which produce minor changes in particle size, so that AOD and mass are linearly related.

Varying the height of the injection changes the altitude region in which injection occurs. So lowering some of the injections that were in the middleworld into the troposphere reduces the optical depth in the middle world. Aerosols have longer lifetimes in the overworld than in the middleworld, because they cannot mix out of the stratosphere along isentropic surfaces. Therefore moving more of the injection into the overworld can ultimately extend the lifetime of the perturbation.
Figure 70: Zonal mean, monthly mean AOD at 525 nm from model months 59 to 67, latitudes 20° S to 20° N and 15 km to 20 km in altitude (middleworld). Simulated Manam eruption occurs in month 61. The black line represents eruption modeled as closely to observation parameters as the model grid will allow. The red line is a simulation with the peak of the eruption one level lower than the observations and the blue is one level higher. The green and purple lines represent simulated eruptions with injection at the same peak height of the observations but with half and double, respectively, the total amount of observed SO₂ emitted.

Figure 71: Zonal mean, monthly mean AOD at 525 nm from model months 59 to 67, latitudes 20° S to 20° N and 20 km to 30 km in altitude (overworld). Simulated Manam eruption occurs in month 61. The black line represents eruption modeled as closely to observation parameters as the model grid will allow. The red line is a simulation with the peak of the eruption one level lower than the observations and the blue is one level higher. The green and purple lines represent simulated eruptions with injection at the same peak height of the observations but with half and double, respectively, the total amount of observed SO₂ emitted.
4.3.2 Results and Discussion

Figure 72-75 show the collective results from the modeling experiment compared to observations of stratospheric AOD (integrated from extinction profiles from 20 to 30 km in the tropics and 15 to 30 km in the mid-latitudes) averaged monthly from 2000 to 2010 from SAGE II, GOMOS and CALIOP (dashed black line with x’s marking monthly values). This comparison shows the three baseline model runs in green, the two anthropogenic runs in blue, the 10x anthropogenic emission run in dashed blue, and the model run with volcanic injections in red.

In Figure 72, it is evident that the model run containing volcanic emissions compares quite favorably to the observations from 20 km to 30 km in the tropics. In Figure 73 and Figure 74 (mid-latitudes) the volcanic run does not compare as well to observations but it is obvious that the variability

![Graph showing monthly averaged AOD at 525 nm averaged from 20° S to 20° N and integrated from 20 km to 30 km from model experiment runs and observations (black dashed line) from SAGE II (2000 to Aug. 2005), GOMOS (Mar 2002) and CALIOP (Apr. 2006 onward). Baseline model runs are in green. Model runs with increasing anthropogenic emissions from 2000 to 2010 are in blue. A model run with ten times the increasing anthropogenic emissions from 2000 to 2010 is represented by the dashed blue line. The model run with volcanic emissions is in red. The black X’s and letters along the bottom of the plot represent the volcanic eruptions during this period that were included in the model run.](image-url)
Figure 73: Monthly averaged AOD at 525 nm averaged from 30° N to 50° N and integrated from 15 km to 40 km from model experiment runs and observations (black dashed line) from SAGE II (2000 to Aug. 2005), GOMOS (Mar 2002) and CALIOP (Apr. 2006 onward). Baseline model runs are in green. Model runs with increasing anthropogenic emissions from 2000 to 2010 are in blue. A model run with ten times the increasing anthropogenic emissions from 2000 to 2010 is represented by the dashed blue line. The model run with volcanic emissions is in red. The black X’s and letters along the bottom of the plot represent the volcanic eruptions during this period that were included in the model run.

Figure 74: Monthly averaged AOD at 525 nm averaged from 30° S to 50° S and integrated from 15 km to 40 km from model experiment runs and observations (black dashed line) from SAGE II (2000 to Aug. 2005), GOMOS (Mar 2002) and CALIOP (Apr. 2006 onward). Baseline model runs are in green. Model runs with increasing anthropogenic emissions from 2000 to 2010 are in blue. The dashed blue line represents a model run with ten times the increasing anthropogenic emissions from 2000 to 2010. The model run with volcanic emissions is in red. The black X’s and letters along the bottom of the plot represent the volcanic eruptions during this period that were included in the model run.
created by the simulated volcanoes compares more favorably to the observations than do the runs with the anthropogenic emissions. In all three of these plots (Figure 72-Figure 74) the AOD in the anthropogenic runs are indiscernible from the baseline emission runs. Note these runs are being done with a free running model, not one forced by observed winds. Hence one cannot expect perfect agreement between the model and observations because transport from the locations of the eruptions is important in determining the aerosol abundances.

Figure 75 compares results of model runs to the annual averaged integrated aerosol volume backscatter coefficient from each of the three lidar sites discussed in Chapter 2. The results from the volcanic model also compare much more favorably to the observations than the anthropogenic runs, which show essentially no difference from the baseline model. It is also interesting to note the increases in AOD attributed to 10x the actual anthropogenic emissions in Figure 72-75. In the tropics, the increased anthropogenic simulation shows increases in AOD and IABS comparable to the volcanic simulation and suggests further increased emissions may have a larger impact on the stratospheric aerosol layer.

WACCM was not forced to the observed dynamics from 2000 to 2010. Therefore, the point of this experiment was not to try to exactly match observations. Rather, the point was to examine the difference in stratospheric aerosol variability when forced by a string of volcanic injections of SO$_2$ into the stratosphere versus sources representative of increases in surface emissions of SO$_2$ from China and India. It is unexpected that the volcanic simulation in Figure 72 matches the observations to such a high degree. The results in Figure 73 and Figure 74 are more representative of what was expected from the comparison due to differences in transport and the limitations imposed on these simulations due to the coarse grid of the model and the sensitivity of the resulting aerosol to the height of the volcanic injections due the discrete vertical
resolution. The differences in transport in the model are more noticeable in the mid-latitudes because aerosol is transported strongly along isentropes, as well as through the diabatic transport process observed in the tropics (Holton et al. 1995). The variability resulting from the volcanic runs and the relatively large optical depths reached following eruptions much more closely matches the observations than the variability and peak optical depths of any of the other runs. Hence, simulations presented here directly support the suggestion of Vernier et al. (2011a) that volcanic eruptions are responsible for the trends in stratospheric optical depth seen from 2000 to 2010. Yet, observations by Randel et al. (2010) and Vernier et al. (2011b) suggest the Asian monsoon may be transporting aerosol into the lower stratosphere (middle world). This is further explored in Figures 77-80.

Figure 76 depicts a zonal mean plot of SO$_2$ and aerosol mass for January and July. Model output was taken from the tenth year of a baseline run. In January there is a clear demarcation between the troposphere and stratosphere for both species. In July, no clear boundary exists. A large amount of SO$_2$ is found in the UTLS region in the northern mid-latitudes extending into the tropics. A similar increase in aerosol mass is seen in the UTLS at Northern mid-latitudes extending into the tropics in July. These UTLS concentrations of SO$_2$ and aerosol suggest that emissions of SO$_2$ in the northern hemisphere could act as a source of aerosol to the middle-world region. Thus, any increases in this source should also manifest themselves in this region of the stratosphere. Figure 77 and Figure 78 show that the source in anthropogenic SO$_2$ is largely centered in China and India.
Figure 75: Annually averaged integrated aerosol volume backscatter coefficient at 532 nm from model experiment runs and observations (black dashed line) from the Boulder (a), Mauna Loa (b) and Lauder (c) lidars. Baseline model runs are in green. Model runs with increasing anthropogenic emissions from 2000 to 2010 are in blue. A model run with ten times the increasing anthropogenic emissions from 2000 to 2010 is represented by the dashed blue line. The model run with volcanic emissions is in red. The black X’s and letters along the bottom of the plot represent the volcanic eruptions during this period that were included in the model run.
Figure 76: Monthly averaged SO$_2$ (a and c) and aerosol mass (b and d) for January (a and b) and July (c and d) from 2010 in the baseline model run.
Figure 77: Monthly averaged SO$_2$ for January 2009 at 313.5 mb in an anthropogenic model run.

Figure 78: Monthly averaged SO$_2$ for July 2009 at 313.5 mb in an anthropogenic model run.
In Figure 79, the AOD from 12 km to 40 km is plotted for 30° N to 50° N. The only difference in this plot from Figure 72 is the extension of the altitude range into the middleworld region. The inclusion of the lower stratosphere causes the AOD for each of the runs to become heavily modulated by an annual cycle that peaks in late fall, rather than the winter time peak observed in the over-world region. A small separation may also emerge after 2005 between the baseline runs and the anthropogenic runs.

Figure 79: Monthly averaged AOD from 30° N to 50° N from 12 km to 30 km from model experiment runs. Baseline model runs are in green. Model runs with increasing anthropogenic emissions from 2000 to 2010 are in blue. A model runs with ten times the increasing anthropogenic emissions from 2000 to 2010 is represented by the dashed blue line. The model run with volcanic emissions is in red.

Figure 80 further depicts the separation of the anthropogenic runs from the baseline runs by only looking at the AOD from 17 km to 21 km from 20° N to 50° N. The separation is especially seen in 2005, 2006 and 2007 when anthropogenic emissions peaked (Figure 68). In 2006, the peak separation is approximately 20%. To examine the possible impact of the anthropogenic aerosol sources further, zonal mean plots of the difference between the mean of
the anthropogenic and baseline runs are shown for March and September of 2000 and 2009 to illustrate the spatial extent of these differences (Figure 81).

Figure 80: Monthly averaged AOD from 20° N to 50° N from 17 km to 21 km from model experiment runs. Baseline model runs are in green. Model runs with increasing anthropogenic emissions from 2000 to 2010 are in blue.

The methodology described by Mahlstein et al. (2011) is used to test for significance (grey shading in Figure 81) between these means. Here, the Kolmogorov-Smirnov test (K-S test) is employed at the 90% level to discern whether the two ensembles’ mean AOD (at each point in a monthly zonal mean) comes from a different distribution. Similar results were also found using a Student t-test but because the K-S test makes no assumptions about the nature of the underlying distributions being tested it is a more appropriate test. Figure 81 shows that no significant difference is found between the baseline and anthropogenic emission schemes for March and September 2000. In 2009, areas of significance emerge in March. The most important of these changes is located from 50° N to 90° N along the tropopause (black line). This difference (~10%) is thought to be representative of the long-term change in AOD due to the increases of anthropogenic emissions. In September 2009, a much larger area of significant increase is seen along the tropopause from 40°S to 90°N. A large increase is also seen up to 16 km in the mid-
latitudes and 21 km in the tropics. These differences are thought to be mostly due to the annual increases in SO$_2$ in the middleworld region from enhanced summer-time convective uplift and seasonal emission increases in the northern hemisphere. These increases are mostly diminished in winter but as shown by the plot depicting March 2009, a residual increase remains in the lower stratosphere of northern high latitudes. These increases are representative of ~10-15% increase in AOD (integrated from 17 to 30 km, in the northern hemisphere) above the baseline simulation.

Figure 81: Mean differences in aerosol extinction between the mean of the 2 anthropogenic model runs and the three baseline model runs for March (a and c) and September (b and d) 2000 (a and b) and 2009 (c and d). Grey shading represents areas with a significant difference at the 90% level. Regions in (a) designated by Roman numerals correspond to the four time series in Figure 75. The black line is the zonal monthly mean tropopause.
Emergence of the significance of the anthropogenic impacts over the course of the modeled decade was also analyzed in the four regions designated by Figure 81a. To do this, the monthly mean probability that the mean of the two ensembles is different is plotted for the length of the simulations. The probability is derived as the mean of 1 minus the p-value resulting from the K-S test conducted on all points in the specified region for each month. The over-world (from 20 km to 30 km) is defined as region I. In Figure 82, as expected from Figure 81, it is evident that the two emission schemes do not result in any significant difference for the entire simulation in region I. Regions II and IV are representative of the northern hemisphere middleworld. In both cases the differences become significant after 2005 during the late summer and fall. Region IV also exhibits the most significant and clearly cyclic difference from all the regions examined. This cyclic difference suggests that the aerosol is being preferentially transported to region IV during the summer. Because of the differences in Figure 81d, region III was also examined. An increasing trend is seen in the probability of the difference between baseline and anthropogenic simulations being significant but no clear signal emerges above the 90% level. Region III is the southern hemisphere equivalent to region II but, because the source of aerosol is in the northern hemisphere, it is thought that preferential transport along isentropes will only lead to significant increases of AOD in the middle world of the northern hemisphere.
4.3.2 Summary of Trend Analysis

The observed decadal increases in AOD from 2000 to 2010 were examined by using the model to conduct a numerical experiment. The experiment was accomplished by simulating emissions from the string of volcanic injections during this period and the increases in Indian and Chinese emissions of \( \text{SO}_2 \), which likely peaked in 2008. In this study, an attribution of stratospheric aerosol variability was made possible by the ability to isolate the human and volcanic sources; an ability that observations from 2000 to 2010 lack. The results of these simulations suggest that the overworld is not significantly impacted by anthropogenic emissions of \( \text{SO}_2 \) from Asia and that volcanoes are the main drivers of aerosol variability (above the natural annual variability) in this region. Of course anthropogenic emissions of COS are important to the stratosphere, as is the global emission of \( \text{SO}_2 \) and sulfate. In the middleworld, anthropogenic emissions were found to increase stratospheric aerosol annually in the late summer and fall in the northern hemisphere, especially noticeable in the region of the tropopause. During the rest of
2009, slight increases (above the baseline model) in the mean AOD were also observed. The increase in the background level is thought to be due to the slow residual build up of aerosol from the increasing annual source while the rainout of aerosol remains constant. These results are in agreement with the suggestion of Vernier et al. (2011a and b) that volcanoes are responsible for stratospheric AOD trends from 2000-2010 and largely contradict the theory suggested by Hofmann et al. (2009) that rising emissions of SO2 in Asia are responsible.
CHAPTER V: SUMMARY

5.1 Main Findings and Implications

Observations from ground-based lidar from 2000 to 2010 reveal large variability in the stratospheric aerosol layer, a period unperturbed by colossal volcanic eruptions. The goal of this research was to examine the annual and decadal components of the observed variability and to delineate their mechanism or source. Thus, the main questions set forth by this work were:

(1) What do lidar observations from 2000 to 2010 reveal about stratospheric aerosol variability?

(2) What role do small volcanic eruptions and anthropogenic emissions have in the observed variability in the stratospheric aerosol record during the last ten years?

In the attempt to answer the first question it was found that the lidar observations from Boulder, CO, Mauna Loa, HI, and Lauder, NZ show strong wintertime annual peaks in aerosol volume backscatter and a 4-7% increase per year between 20 and 30 km. This result was found to match well with the observations presented in other work (Hofmann et al., 2009; Nagai et al., 2010; Trickl et al., 2010; Vernier et al. 2011a). In the analysis shown here, it was found that observations of lidar volume backscatter coefficient and other measurements of extinction convolve the separate sources of stratospheric aerosol and therefore cannot be used to attribute a proportion of the observed variability to a mechanism or source.

Because of the ambiguity in observations, a model of stratospheric aerosol was employed to further evaluate the second question. The model chosen for this work is a version of
WACCM/CARMA specifically configured to simulate stratospheric aerosol. Through comparison with ground based lidar observations and observations from SAGE II, it was determined that the model, in its baseline configuration, accurately simulates the background stratospheric aerosol layer above 20 km. Below 20 km, because the model ignores all non-sulfate aerosol from the troposphere, the comparisons show that the model underestimates the aerosol extinction and volume backscatter but does accurately simulate the sulfate and meteoritic components of the total aerosol. Above 30 km, the model results compare well with observations from SAGE II but not to the lidar observations. Corroborating evidence from observations of stratospheric aerosol composition in the lower stratosphere made by Murphy et al. (2007) and extinction of the upper stratosphere and mesosphere from SOFIE observed by Hervig et al. (2009) support the inclusion of meteoritic smoke to correctly model the stratospheric aerosol layer. Thus, it was concluded that an error existed in the lidar retrievals that was causing a bias that resulted in the lidar aerosol backscatter to be underestimated mainly above 30 km.

Comparisons of observations to model derived extinction profiles in the stratosphere with and without the inclusion of smoke suggest that the extinction caused by meteoritic smoke and its interaction with the sulfate aerosol is significant down to altitudes as low as 25-30 km at mid-latitudes and 35 km in the tropics. Hence the presence of smoke needs to be considered in remote sensing retrievals. When this extinction is omitted, lidar measurements appear to be biased through a significant depth of the stratosphere, down to near 30 km in the tropics. Further analysis of the lidar error suggested that application of a small correction to the calibration used in the retrieval (that accounts for the scattering of meteoritic smoke in the region of the lidar
calibration altitude) would fix the observed biases in the lidar aerosol volume backscatter coefficient profiles.

Possible solutions to achieve more accurate lidar measurements include calibrating instruments within regions of the atmosphere in which information about the extinction of meteoritic aerosol is known and may be included within retrievals. SAGE II and other similar observations (including lidar observations after addressing the concerns described in this work) above about 30 km altitude reflect the behavior of meteoritic smoke, and could be mined to learn more about temporal trends and latitudinal behavior of meteoritic smoke as well as its impact upon the sulfate aerosol layer. Because of the strong latitudinal dependence in meteoritic smoke, further analysis of polar lidar observations may also reveal new information and serve as a check for this analysis.

The annual cycle of stratospheric AOD in the model was first validated through a comparison of observations from SAGE II and from aircraft data (Wilson et al., 2008) to results from the baseline model runs. The baseline model was used because the repeating yearly emissions of the baseline model provide the clearest signal of the annual cycles in stratospheric aerosol without interference from trends and other variability. The model was found to compare well with observations from SAGE II and capture the tracer behavior of stratospheric aerosol depicted by the correlation of aerosol abundance with N₂O by Wilson et al. (2008).

The baseline model results were then compared to the record of stratospheric IABS from the lidar observations presented in Chapter 2 and a combined satellite observation record from SAGE II, GOMOS and CALIOP. The comparison of the annual cycle in the lidar observations revealed an error in the lidar retrieval that led to a bias in the observed annual cycle. Comparison with satellite observation suggested a seasonal cycle does exist in the northern and southern
hemisphere satellite observations but is also hidden by the large increases caused by the episodic volcanic injection of aerosol, though the annual variability may play a larger role in the total variability of stratospheric AOD.

As for the second question of this thesis, there are two main theories that have attempted to explain the observed trends in aerosol. The first argued that recent increases in Asian SO\textsubscript{2} injected into the stratosphere by the Asian monsoon and other convective processes in the tropics could lead to the observed trends (Hofmann et al., 2009). The second theory suggests that the trends are the result of the combined impact of several moderate volcanic injections into the stratosphere from 2000 to 2010 (Vernier et al., 2011a).

To examine the trends, perturbations that are representative of the increases in anthropogenic emissions and volcanic injections from 2000 to 2010 were applied to the standard emission scheme of the baseline model. These results were compared to the lidar observations and observations of AOD from SAGE II and CALIOP. Unlike the observations, the numerical experiment provided the ability to isolate the impact of the two different sources on the stratospheric aerosol layer.

The results of these simulations suggest that volcanoes are the main drivers of aerosol variability above 20 km from 2000 to 2010. They also definitively show that the overworld has not been significantly impacted by anthropogenic emissions of SO\textsubscript{2} in Asia from 2000 to 2010, though these emissions are part of a global source. These results are in agreement with those of Vernier et al. (2011a; b). In the northern mid-latitude middleworld, anthropogenic emissions were found to increase stratospheric aerosol cyclically, in the late summer and fall in the northern hemisphere, by as much as 20% above the baseline simulations. These increases were especially noticeable in the region of the tropopause. A smaller increase (~10%) in aerosol in
this region was also observed during other months at the end of the anthropogenic simulations and is thought to be due to the slow residual build up of aerosol from the increasing annual source.

It should be noted that the argument for a volcanic source of stratospheric aerosol is somewhat circular: injections of aerosol into the stratosphere led to increases in stratospheric aerosol. Because the formation of sulfate aerosol from its constituents takes on the order of minutes, the observations from CALIOP of aerosol backscatter used to model injection height are thought to be robust for the purposes of this work. Further examination of the volcanic injection height and the lofting of aerosol formed in the troposphere needs to be examined with a model which couples CARMA radiatively to transport to more clearly attribute the sources of the increased aerosol.

The importance of the aerosol trend in terms of radiative forcing is described in Solomon et al. (2011). This work suggests that as much as 0.1 W/m² of cooling from 2000 to 2010 may be attributed to the increase in stratospheric aerosol. The cooling from aerosol in the overworld may now be attributed to natural sources. It should also no longer be thought of as a trend. Rather the stratospheric aerosol layer should be treated as a natural source of cooling that includes a background (base) amount of aerosol that is episodically perturbed by injections from volcanoes. This work does suggest that increases in aerosol in the lower stratosphere may be impacted by anthropogenic emissions but, at most, only 20% of the observed increases in AOD in the extra-tropical northern hemisphere (Figure 80) could be explained by current emissions of SO₂ from China and India.
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APPENDIX A: Polarization LIDAR at Summit, Greenland for the Detection of Cloud Phase and Particle Orientation


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Abstract

Precise measurements of cloud properties are necessary to document the full range of cloud conditions and characteristics. The Cloud, Aerosol Polarization and Volume backscatter coefficient Lidar (CAPABL) has been developed to address this need by measuring depolarization, particle orientation and the volume backscatter coefficient of clouds and aerosol. The lidar is located at Summit, Greenland (72.6° N, 38.5° W; 3200 m. a. s. l.) as part of the Integrated Characterization of Energy, Clouds, Atmospheric State and Precipitation at Summit project and NOAA’s Earth System Research Laboratory’s Global Monitoring Division’s lidar network. Here the instrument is described with particular emphasis placed upon the implementation of new polarization methods developed to measure particle orientation and improve the overall accuracy of lidar depolarization measurements. Initial results from the lidar are also shown to demonstrate the ability of the lidar to observe cloud properties.
A1 Introduction

Clouds and aerosol modulate the surface energy and ice mass budgets in polar regions (Francis and Hunter, 2006; Kay et al., 2008; van den Broeke et al., 2009). Any alteration in the current climatology of clouds and aerosol will have large impacts on their role in both of these budgets. Remote sensing of the vertical distribution, volume backscatter coefficient and linear volume depolarization of particles in the atmosphere has been shown to greatly contribute to this knowledge by constraining their radiative parameters (Sassen et al., 2003; Hansen et al., 2011).

Currently, lidar measurements allow for continuous observation of the thermodynamic phase of atmospheric particles from the surface to the stratosphere (Russell et al., 1979; Sassen, 2005). As with all remote sensing techniques, the determination of thermodynamic phase by linear polarization lidar is subject to many uncertainties from measurements and assumptions in the retrieval of physical parameters from raw data (Russell et al., 1979; Sassen, 2005; Hu et al., 2009; Nott et al., 2011; Hayman and Thayer, 2009). A significant assumption in conventional lidar depolarization ratio methods is that the observed particles are randomly oriented (Kaul et al., 2004; Hu et al., 2009). This assumption has been shown to bias cloud phase estimates towards higher assignment of liquid water and inhibit accurate quantitative measurement and analysis of cloud phase by depolarization lidar (Hu et al., 2009; Takano, 1989; Chepfer and Brogniez, 1998; Hayman and Thayer 2012). It has also been well documented, through observations of optical phenomena such as sun dogs and halos, that this assumption is clearly not valid for a proportion of observations (Lynch et al., 1994; Hu et al., 2009; Noel and Chepfer, 2010).

It should be noted that only certain habits of ice crystals will orient horizontally because orientation is a bulk property of a population of ice crystals with similar aerodynamic properties. The habits of ice crystals are quite variable and are strongly dependent on the super-saturation of
ice and temperature where the crystal is formed and grows (Magono and Lee, 1966; Bailey, 2009). Kaul et al. (2004) found that the orientation of particles with large azimuthal diameters are more probable to be horizontally oriented, but observations of these particles are obscured by a larger population of smaller randomly oriented particles. This leads current observations to determine mean particle parameters that under characterize the amount of particle orientation in clouds. The horizontally oriented ice crystals (HOIC) discussed here are ideally thought of as hexagonal plates but the observations shown here, in reality, result from populations of ice crystals that are plate-like (i.e. flatter) around their preferred orientation.

With conventional depolarization lidars, cloud phase is often classified by interpreting the ratio of two perpendicular polarization channels in conjunction with the relative amount of observed volume backscatter coefficient. This ratio, conventionally assigned to the symbol \( \delta \), is often called the depolarization ratio. The ability to identify cloud phase using the depolarization ratio was recognized by Schotland et al. (1971) and is based on the assumption that near-spherical liquid water droplets will produce low linear depolarization ratios while non-spherical ice crystals will produce high linear depolarization ratios. Implicit to this interpretation is that the scattering volume is optically thin, such that multiple scattering of spherical particles is not the cause for higher depolarization ratios (e.g., Pal and Carswell, 1973), and that the particles within this volume are randomly oriented, such that volume backscatter coefficient from oriented ice crystals are not the cause for low depolarization ratios (e.g., Platt et al., 1978). These issues have been generally treated by correlating the depolarization ratio with the relative amount of observed volume backscatter coefficient. Post-processing algorithms developed for the CALIOP mission have been implemented and demonstrated to help distinguish multiple scattering and oriented ice crystals effects in characterizing cloud phase (Hu et al., 2009; Noel and Chepfer,
The data set from near-nadir (0.3°) CALIOP observations indicated a clear population of oriented ice crystals when organized into the scattering regime of high volume backscatter signals with low depolarization ratios. In comparison, an off-nadir (3°) CALIOP data set no longer observed this characteristic scattering regime. This proved to be a useful approach in identifying oriented ice crystals. However, the off-nadir measurement does not preclude the presence of oriented ice crystals from the data set but moves them into another regime of the volume backscatter coefficient to depolarization scattering relationship that is not clearly separable from randomly oriented ice crystals. It is known that when horizontal ice crystals are viewed obliquely the volume backscatter coefficient strength decreases substantially while the depolarization ratio increases (Platt et al., 1978). Thus, when viewed obliquely the scattering characteristics of horizontally oriented ice crystals are similar in volume backscatter coefficient strength and depolarization ratio to randomly oriented ice crystals. In fact, many lidar systems operate at oblique angles to avoid the strong specular signal by horizontally oriented ice crystals and the adverse effects these signals have on detector performance. Therefore, polarization lidar systems measuring only depolarization ratios and operating obliquely are not capable of resolving HOIC from randomly oriented ice crystals through depolarization and volume backscatter coefficient measurements. Those lidar systems operating normal to HOIC can use indirect methods of correlating volume backscatter coefficient strength to depolarization ratios but, because of the large dynamic range of the spectral and non-spectral signals, the performance of the system is often compromised. A scanning lidar system can monitor the change in depolarization ratio with incident angle to help identify HOIC but still relies on interpretation and assumptions of the scattering volume (Noel and Sassen, 2005).
Recently, Hayman and Thayer (2012) addressed this issue by exploring the general polarization properties of atmospheric scatterers and particularly of HOIC. They showed through forward polar decomposition of scattering matrices that HOIC can display the polarization properties of depolarization, retardance and diattenuation. They note that the commonly estimated depolarization ratio does not make any distinction between scattering matrix types but only indicates whether the polarization properties have changed. It is not until the scattering matrix is truly of a randomly oriented, non-spherical particle description that the depolarization ratio has a physical polarization definition. The scattering matrix for oriented particles cannot be attributed to any single polarization effect. However, using forward polar decomposition, the oriented scattering matrix can be decomposed into the differing polarization effects of depolarization, retardance and diattenuation. Hayman and Thayer (2012) indicate diattenuation, which is a polarization dependent scattering efficiency, is displayed by oriented particles when viewed at oblique scattering angles. This is a property that cannot be exhibited by randomly oriented particles, and thus, can be used to identify HOIC in oblique lidar volume backscatter coefficient observations. The goal of this paper is to describe a polarization lidar system designed to measure HOIC and to demonstrate the benefits of such a configured lidar system for cloud and aerosol studies.

CAPABL is located at Summit, Greenland (72.6° N, 38.5° W; 3200 m. a. s. l.) as part of the Integrated Characterization of Energy, Clouds, Atmospheric State and Precipitation at Summit (ICECAPS) project (Shupe and Coauthors, 2012). While its primary objective is to identify tropospheric cloud phase, the ability of the lidar to identify HOIC removes ambiguity in the interpretation of linear depolarization ratios. The lidar is also part of NOAA’s Earth System Research Laboratory’s Global Monitoring Divisions (GMD) lidar network. In this role, the lidar
uses traditional volume backscatter coefficient retrieval methods to measure aerosol profiles into the lower stratosphere (Russell et al., 1979; Barnes and Hofmann, 1997; Hofmann et al., 2009). CAPABL was deployed to Summit in June of 2010 and has run continuously (24 hour operation) since that time except for short periods of maintenance and further instrument development. CAPABL is the newest addition to the Arctic network of lidars and due to its location on top of the Greenland Ice Sheet (GIS) is one of the few lidars that will be minimally impacted by regional change (Nott and Duck, 2011). From June 2010 to April 2011, CAPABL was run in a near-zenith pointing direction to represent traditional polarization lidar measurements and then in May 2011 was tilted about 11° off zenith to search for non-zero diattenuation values in the backscattered signals from HOIC.

A1.1 Tropospheric Cloud Measurements and ICECAPS

Despite the importance of clouds to Greenland’s climate, recent studies indicate that little is known about the true cloud cover characteristics over Greenland (Griggs and Bamber, 2008). Currently, the only observations of cloud microphysics made at Summit were reported in 1993 (Borys et al., 1993). The large uncertainty in cloud fraction and the lack of information on cloud microphysical properties inhibit our understanding of cloud radiative effects on the surface (Starkweather, 2004). As a result, current models poorly represent clouds over the Arctic and more specifically over the GIS, and simulations of surface energy budgets and precipitation continue to be highly uncertain.

The goal of ICECAPS is to make measurements of the cloud, atmosphere, precipitation and radiation properties over the GIS to address these issues (Shupe and Coauthors, 2012). Alongside the polarization lidar discussed here, an instrument suite consisting of a cloud radar, two microwave radiometers, an Atmospheric Emitted Radiance Interferometer, an X-band
precipitation sensor, a ceilometer, a micro-pulse lidar, a sodar and a twice-daily radiosonde program contribute to the ICECAPS dataset. Data from CAPABL will be used in conjunction with the other instruments at the observatory to help improve the understanding of Arctic clouds and climate processes by quantifying cloud occurrence, vertical distribution, microphysical composition and radiative properties. This information can then be used to better constrain the next generation of forecast and climate models. Current data and analysis software may be found at: http://www.esrl.noaa.gov/psd/arctic/observatories/summit/browser/.

A1.2 Aerosol Volume backscatter coefficient Measurements

The stratospheric aerosol layer has been monitored by lidars operated by GMD at Mauna Loa, Hawaii and Boulder, Colorado since 1975 and 2000, respectively (Russell et al., 1979; Barnes and Hofmann, 1997; Hofmann et al., 2009). From 2000 to 2010, a 6% per year increase in the stratospheric aerosol volume backscatter coefficient has been observed by many globally located ground-based lidar systems as well as several satellite systems (Barnes and Hofmann, 1997; Hofmann et al., 2009; Solomon et al., 2011; Vernier et al., 2011). CAPABL complements GMD’s other lidars by providing a polar record to the aerosol dataset. This new record will help complete GMD’s goal of creating a long-term holistic understanding of stratospheric aerosol by revealing new information about the aerosol layer in the polar downwelling segment of the mean meridional circulation (Holton, 1986; Holton et al., 1995). Data from CAPABL will primarily be used to understand the latitudinal dependence of the stratospheric aerosol layer and its associated cycles. Analysis will also be done on injections of aerosol into the upper troposphere and lower stratosphere by annually occurring pyrocumulonimbus clouds created by boreal forest fires in Canada and Russia and episodic high latitude moderate volcanic injections, such as the recent eruptions from Kasatochi and Sarychev, which may have a larger impact on the stratospheric
aerosol layer and decadal climate variability than previously concluded (Fromm, 2005; Fromm et al., 2007; 2008; Vernier et al., 2011; Solomon et al., 2011).

A2 Location

Summit Station is the only dedicated atmospheric observatory operating continuously at high altitudes in the Arctic. Because of its elevation, Summit allows for almost direct access to the free troposphere and is relatively free of local influences that could corrupt free tropospheric climatic records. The high altitude also allows CAPABL to easily observe the lower stratosphere. Summit is situated ideally for studies attempting to identify and understand long-range, intercontinental transport, such as the long-range transport of boreal forest fire smoke. While other Arctic atmospheric research observatories, such as those at Barrow, Alert, Ny Alesund, Tiksi and Cherski, lie at sea level near coastal and continental influences, Summit is free of regional effects from increased shipping, melting ice and thawing permafrost. Thus, changes in the observational record from Summit should generally represent widespread Arctic trends or events that are sufficiently significant to have large-scale effects. This makes Summit an ideal location for observing processes that may be applicable to the larger GIS and the Arctic region.

A3 System Description

CAPABL is based on NOAA’s Earth System research Laboratory’s Chemical Science Divisions (CSD) Depolarization and Volume backscatter coefficient Unattended Lidar (DABUL) (Alvarez et al., 1998; Intrieri et al., 2002; Turner, 2005). The structure of DABUL was unchanged, but the transmitter and the receiver were reconfigured to enable measurement of diattenuation for detecting HOIC. Modifications most notably include the use of a Meadowlark liquid crystal variable retarder (LCVR) and a new data acquisition system.

The CAPABL transmitter consists of a frequency-doubled, diode-pumped, neodymium-
doped, yttrium lithium fluoride (Nd:YLF) laser operating at 523.5 nm (Fig. (A1)). The transmitted signal is first passed through a half-wave plate (HWP) and polarizing beam splitter (linear output polarization) to ensure maximum linear polarized power output. After the polarizer, the beam passes through an 80 times expander to achieve a divergence of 0.165 mrad. A back reflection of the beam, from a beam sampler, is then used to trigger a photodiode, which initiates the data acquisition. Two folding mirrors aligned in periscope configuration direct the transmitted beam above the receiver secondary. This allows for a full overlap with the receiver field-of-view at 200 m, which is required for tropospheric studies. The several folding mirrors in the transmitter generally disrupt the linearly polarized input state. However, there must be a linear input polarization that produces a linear output polarization. The HWP located directly in front of the laser is rotated so that the output polarization is purely linear above the convergence transmit mirror (located above the receivers secondary mirror). A final HWP above the convergence mirror allows for the rotation of this linearly polarized signal and defines the polarization axes of the lidar (all further references to transmitted or received polarization are in reference to this axis which is at zero degrees when the polarization is in the s-plane or horizontally polarized). To achieve the polarization measurements of diattenuation, described in the next section, the lidar output polarization is rotated (using the outgoing HWP) 45° to the reference horizontal polarization plane (see Fig. A2).
The receiver consists of a F/14.3 Dall-Kirkham telescope with 508 cm focal length and 35.6 cm aperture (see Table A1). This telescope design uses symmetric low angles of incidence on the mirrors, which minimizes polarization effects caused by the system. The collected light is then collimated by a 30 mm negative lens and passed through a horizontal quarter-wave plate (QWP), the LCVR oriented at 45° and a polarizing beam splitter (horizontal polarizer). The
combination of these polarization elements creates a rotating analyzer with polarization angle controlled by the phase of the LCVR. After the polarizer, 10% of the signal is passed to the low altitude PMT with low gain to avoid saturation from high intensity signals below 1 km in altitude. The remaining 90% of the signal is passed to the high gain channel for upper tropospheric and lower stratospheric data collection.

<table>
<thead>
<tr>
<th>Transmitter</th>
<th>Receiver</th>
<th>Signal Processing</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Spectra-Physics EL2-523Q Diode pumped Nd:YLF)</td>
<td>(Dall-Kirkham cassegrain telescope configuration)</td>
<td>(Photon Counting Data Acquisition)</td>
</tr>
<tr>
<td>Wavelength: 523.5 nm (frequency doubled)</td>
<td>Receiver Aperture: 35.6 cm</td>
<td>Data System: Fast Comtec P7882</td>
</tr>
<tr>
<td>Pulse Energy: 25 µJ</td>
<td>Filter bandwidth: 0.3 nm</td>
<td>Range Bin Size: 100 ns (30m)</td>
</tr>
<tr>
<td>Pulse Rate: 2000Hz</td>
<td>Number of Channels: 2 (High, Low)</td>
<td>One-line integration: 5 s</td>
</tr>
<tr>
<td>Divergence: 0.165 mrad</td>
<td>Field of View: .3 mrad, .7 mrad</td>
<td>PMTs (2): EMI 9863B/100</td>
</tr>
</tbody>
</table>

Table A1. CAPABL system specifications.

A LabVIEW program controls the lidar operation and data acquisition system. The entire system (including data transfer and processing) is fully autonomous and runs continuously. Control of the lidar may also be done remotely. An operator is only required for hardware maintenance and modifications.

The current observation specifications (including maximum range and resolution) of CAPABL for the observations of clouds in the troposphere and aerosol volume backscatter coefficient from the stratosphere may be found in Table A2 and A3, respectively. Observations of the different polarization channels are made sequentially with a 5 second integration on each. Between each integration, 0.5 s is needed to download the data from the data acquisition system. This results in a total 16.5 s to measure three polarization planes. This observation scheme was based on an analysis of the signal-to-noise requirements, measurement duty cycle and anticipated temporal variability of the clouds. CAPABL has the ability to make measurements at a much faster rate but due to the hard limit of 0.5s between each integration and slow temporal variation
of Arctic stratiform clouds a longer duty cycle was made default for observations in order to increase the signal-to-noise of each observation and reduce the dead time between them (Shupe et al., 2011).

<table>
<thead>
<tr>
<th>Property</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Range</td>
<td>5 km (Above Summit)</td>
</tr>
<tr>
<td>Vertical Resolution</td>
<td>30 m</td>
</tr>
<tr>
<td>Temporal Resolution</td>
<td>16.5 s per polarization</td>
</tr>
<tr>
<td>Observed Polarizations</td>
<td>4</td>
</tr>
<tr>
<td>Uncertainty in Linear Depolarization Ratio</td>
<td>2.5%</td>
</tr>
</tbody>
</table>

Table A2. Polarization observation ranges and limits.

<table>
<thead>
<tr>
<th>Property</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Maximum Range</td>
<td>25 km a. m. s. l.</td>
</tr>
<tr>
<td>Vertical Resolution</td>
<td>300 m</td>
</tr>
<tr>
<td>Temporal Resolution</td>
<td>4 hour</td>
</tr>
<tr>
<td>Observed Polarizations</td>
<td>2 (summed to obtain total backscatter signal)</td>
</tr>
<tr>
<td>Uncertainty in due to Shot Noise</td>
<td>8%</td>
</tr>
</tbody>
</table>

Table A3. Stratospheric aerosol observation ranges and limits.

**A4 Polarization Method**

A polarization lidar system is completely described using the Stokes Vector Lidar Equation (SVLE), which relates the Stokes vector of the transmitted light to the received photon counts in each observed polarization channel (Hayman and Thayer, 2012). Besides the sought after polarization properties of the scatterers in the atmosphere, all of the optical components within the lidar must be included to account for the possibility of polarization modification and cross talk by the optical system. Thus, the SVLE takes the form of

\[
\vec{N} = OM_{RX} \left\{ \left( G(R) \frac{A}{R^2} \Delta R \right) T_{atm}(R) F(\vec{k}_{\|}, \vec{k}_{\perp}, R) T_{atm}(R) M_{TX} S_{TX} + \vec{S}_B \right\}, \tag{A1}
\]

where \( \vec{N} \) is a vector of the photon counts from the observed planes of polarization, \( O \) is the output matrix describing the LCVR and measurement channels, \( M_{RX} \) is the receiver’s Mueller matrix, \( T_{atm} \) is the Mueller matrix accounting for atmospheric transmission, \( F(\vec{k}_{\|}, \vec{k}_{\perp}, R) \) is the
scattering phase matrix for the incident wavenumbers $\vec{k}_i$ and $\vec{k}_s$ at range $R$ and $M_{tx}$ is the Mueller matrix of the transmitter. For randomly oriented particles observed by a mono-static lidar, such as CAPABL, the scattering phase matrix assumes the form of (van de Hulst, 1981; Flynn et al., 2007; Gimmestad, 2008):

$$F(\pi) = \begin{bmatrix} f_{11} & 0 & 0 & 0 \\ 0 & f_{22} & 0 & 0 \\ 0 & 0 & -f_{22} & 0 \\ 0 & 0 & 0 & f_{11} - 2f_{22} \end{bmatrix}. \quad (A2)$$

Linear depolarization due to randomly oriented scatterers may be characterized from this scattering phase matrix by observing the parallel and perpendicular polarization components of the backscatter light (Gimmestad, 2008). The resulting photon count vector of Eq. (A1) takes the form

$$\vec{N} = \begin{bmatrix} N_\perp \\ N_\parallel \end{bmatrix}. \quad (A3)$$

The volume linear depolarization ratio, $\delta$, takes the form

$$\delta = \frac{f_{11} - f_{22}}{f_{11} + f_{22}} = \frac{N_\perp}{N_\parallel}. \quad (A4)$$

For randomly oriented scatterers, this measurement fully characterizes the depolarizing effect of the scattering volume. When HOIC are present in the observed scattering volume off-diagonal elements of Eq. (A2) become non-zero. Furthermore, the depolarization ratio, $\delta$, no longer retains the traditionally assumed physical meaning derived from Eq. (A2), as additional polarization effects contribute to the diagonal elements as well (Hayman and Thayer, 2012). When a scattering volume contains some mixture of oriented and randomly oriented nonspherical particles, the backscattered light will not only be depolarized but may also contain diattenuation and retardance. Thus, for proper interpretation of depolarization data, the orientation state of the scatterers must be known.
To collect the additional information needed to evaluate the assumption of randomly oriented scatterers, additional terms from the scattering matrix must be observed. The scattering phase matrix for a volume containing HOIC is given in Eq. (A5) (Kaul et al., 2004; Hayman and Thayer, 2012). One notable difference between the randomly oriented scattering phase matrix (Eq. A2) and the oriented scattering phase matrix in Eq. (A5) is the additional off-diagonal elements of $F_{12}$ and $F_{34}$. These elements represent diattenuation properties of the scatterer (polarization dependent scattering efficiency) and retardance respectively and a non-zero measurement of either of these terms would signify the presence of HOIC. It is also important to note the different notation between elements in Eq. (A2) and (A5) with the diagonal elements in Eq. (A2) not necessarily equal to the diagonal elements in Eq. (A5).

$$F(k^t_i, -k^r_s) = \begin{bmatrix} F_{11} & F_{12} & 0 & 0 \\ F_{12} & F_{22} & 0 & 0 \\ 0 & 0 & F_{33} & F_{34} \\ 0 & 0 & F_{34} & F_{44} \end{bmatrix}$$  \quad (A5)$$

The $F_{12}$ element represents linear diattenuation and to observe this term requires another polarization measurement in addition to the perpendicular and parallel polarization measurements needed to estimate depolarization. We perform this measurement at 45° compared to the parallel polarization channel. Therefore, CAPABL’s full vector of observed backscattered light is represented by

$$\vec{N} = \begin{bmatrix} N_\perp \\ N_{45} \\ N_{\parallel} \end{bmatrix}.$$  \quad (A6)$$

A depiction of CAPABL’s transmitted light and received polarizations in reference to a HOIC is found in Fig. A2. This measurement arrangement has been entitled the Parallel-45-
Perpendicular (P45P) technique (Hayman, 2011). From this measurement an assessment of the linear diattenuation term, \( D_q \), may be defined in terms of the observable values of the lidar,

\[
D_q = \frac{F_{12}}{F_{11}} = \frac{2N_{15}}{N_{||} + N_{\perp}} - 1. \quad (A7)
\]

Eq. (A7) represents the normalized linear diattenuation of the scattering matrix. If \( F_{12} \) is zero, the scattering matrix will take the form of Eq. (A2). In the case of HOIC, \( F_{12} \) is non-zero and linear diattenuation exists. For CAPABL’s particular geometry the linear depolarization ratio, \( \delta \), takes the form

\[
\delta = \frac{F_{11} + F_{33}}{F_{11} - F_{33}} = \frac{N_{\perp}}{N_{||}}, \quad (A8)
\]

where \( F_{33} < 0 \) due to the \( \pi \) phase shift from the volume backscatter coefficient. This value is observed using the traditional polarization ratio method, as in Eq. (A4), but should be interpreted with consideration that \( F_{33} \) may depend on depolarization, diattenuation and retardance (Hayman and Thayer, 2012). Though symmetry conditions of oriented scatterers also allow for a non-zero \( F_{34} \), results reported by Kaul et al. (2004) suggest this term is generally small compared to linear diattenuation.
Figure A2. Illustration of the polarization terms in reference to the lidar beam and horizontal scatterers. The lidar tilt angle ($\rho$, $11^\circ$ for CAPABL) is measured relative to zenith (vertical axis) and the polarization angle, $\psi$, is measured relative to the linear polarization that lies in the horizontal plane (plane perpendicular to the $\vec{k}$ of the transmitted light). The dashed oval represents the plane perpendicular to the vector of the transmitted beam.

The ability of the instrument to discern diattenuation is dependent on the aspect angle of the ice crystal axis of symmetry and the $\vec{k}$ of the lidar. For CAPABL, $11^\circ$ was chosen to optimize range within the bounds of previous estimates of HOIC concentrations (Noel and Sassen, 2005; Noel and Chepfer, 2010). It was also shown that observations of diattenuation can only be accomplished by tilting the beam off zenith because diattenuation signature of HOIC is zero when the beam is directed normal to the crystal surface.

The P45P method allows the lidar to forego the assumption that scatterers in the observation volume are randomly oriented. The presence of linear diattenuation unambiguously
shows when this assumption is invalid. In the presence of HOIC, the data may still be observed and described in terms of the linear depolarization ratio, $\delta$, but it may not uniquely depend on one term from the scattering matrix and may not equally be compared to the $\delta$ of randomly oriented scatterers.

**A5 Polarization Operation and Sources of Error**

CAPABL uses a LCVR sandwiched between a QWP and horizontal polarizer to create a polarization analyzer that can perform the three polarization measurements necessary for the observation method (Fig. (A1)). The LCVR has no capability to rotate in its mount, so its orientation defines the $45^\circ$ polarization plane within the analyzer setup. The Mueller matrix for this system of optics is

$$A\left(\frac{\Gamma_{wp}}{2}\right) = P(0)V(\Gamma_{wp}, 45^\circ)Q(0) = \frac{1}{2} \begin{bmatrix} 1 & \cos \Gamma_{wp} & \sin \Gamma_{wp} & 0 \\ 1 & \cos \Gamma_{wp} & \sin \Gamma_{wp} & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{bmatrix},$$

(A9)

where $\Gamma_{wp}$ is the voltage controlled phase shift of the LCVR. This allows the polarizer to select any linear polarization component by adjusting $\Gamma$. The Mueller matrix in Eq. (A9) differs from a rotated linear polarizer because the output polarization is always horizontal. This property is a result of CAPABL’s specific setup. The single output polarization means all optics after the analyzer always experiences the same polarization. Thus, the measurements are only dependent on the intensity of the observed signal and the polarization effects of the optical components after the analyzer do not impact the accuracy of CAPABL.

Accurate alignment of CAPABL’s polarization analyzer is necessary to obtain the high polarization accuracy required for diattenuation measurements of HOIC. For this reason, particular attention is given to the alignment of the QWP, LCVR and polarizer that make up the
polarization analyzer in the receiver to which all other components must be aligned. Clear sky observations have demonstrated that the minimum linear depolarization ration observable by CAPABL is 0.025. This value may be interpreted as the error (maximum resolution) of CAPABL’s polarization measurements due to the polarization effects of the instrument.

A5.1 Accuracy of Observed Polarizations

The LCVR imposes a voltage controlled phase shift to change the observed polarization mode. However the phase shift created in the optical medium of the LCRV drifts as a function of temperature. CAPABL is contained in a temperature controlled room at Summit, but some temperature drift does occur in the room. A sensitivity analysis of diattenuation measurements for a relative phase shift error of $\Delta \Gamma_{wp}$ was performed. For a depolarizing medium described by Eq. (A2), the received photon counts as a function of phase shift on the LCVR are given by

$$N_{RX} (\Gamma_{wp}) = \frac{N_0}{2} \left[ f_{11} - f_{22} \sin \Gamma_{wp} \right]$$  \hspace{1cm} (A10)

where $N_{RX}$ are the photons detected, $N_0$ are the photons incident on the LCVR, $f_{22}$ is the (2,2) element of the scattering matrix describing axially symmetric randomly oriented scatterers from Eq. (A2) and $\Gamma_{wp}$ is the phase shift of the LCVR.

The objective of diattenuation measurements with CAPABL is to identify oriented scatterers. Thus, the primary concern is avoiding false positives (i.e. non-zero diattenuation) in the presence of strictly randomly oriented scatterers. Because the room where CAPABL operates is heated through standard HVAC systems, the LCVR retardance error is generally small. The received photon counts for a target phase shift $\Gamma_{wp}$ and phase shift error $\Delta \Gamma_{wp}$ is expanded from Eq. (A10) to first order
Here, the second term corresponds to error in the detected photon counts. The first order error sensitivity of the perpendicular and parallel photon counts (at $\Gamma_{wp} = -\pi$ and $\Gamma_{wp} = \pi$, respectively) is zero. Higher order terms are always equal and opposite between $-\pi$ and $\pi$ and because two terms are summed in diattenuation calculations, the error terms cancel. Thus, diattenuation measurements are relatively insensitive to LCVR phase error in the perpendicular and parallel measurements. To maintain accurate phase shifts corresponding to perpendicular and parallel polarizations, only periodic recalibration of the LCVR voltage settings is required.

The 45° polarization measurement is made at $\Gamma_{wp} = 0$, where Eq. (A11) has maximum sensitivity to $\Delta \Gamma_{wp}$. The measured diattenuation of the randomly oriented scatterers as a function of LCVR error in the 45° plane is given by evaluating Eq. (A7) with Eq. (A11) at each corresponding phase shift, resulting in

$$D_q = -f_{22}\Delta \Gamma_{wp}.$$  \hspace{1cm} (A12)

Because randomly oriented scatterers are only being considered in this analysis, any non-zero value of $D_q$ must be strictly related to the error. Deviations from zero may therefore be used to determine the phase error of the LCVR. This error is then used in a feedback loop to control the LCVR voltage for 45° polarization measurements.

The feedback loop for LCVR control is closed using part of the lidar profile. However, to make precise corrections to the LCVR voltage, the uncertainty from the signal-to-noise of the photon counts must be low and we must assume that this part of the profile contains no actual diattenuating particles. Therefore, low altitude signals, where photon counts are high, are used to close the feedback loop. To obtain a diattenuation uncertainty of less than 1%, individual profiles
are further integrated in the feedback loop. Longer integration has the effect of reducing the control bandwidth but since phase shift drift caused by temperature fluctuations is slow, the controller bandwidth is only a concern with regard to settling time when first starting the loop. Currently the feedback loop takes 10 to 20 minutes to lock on the appropriate value of $\Gamma_{wp}$ when the instrument begins it measurement. After the loop has settled on this value, its time response is able to react quickly enough to normal operational changes in building temperature to adapt $\Gamma_{wp}$ during the duration of the measurement without corrupting the observations.

A5.2 Nonlinear Photon Counting

Nonlinear photon counting due to detector saturation is also a large concern for the depolarization and diattenuation measurements as these are functions of the parallel channel photon counts (Eq. (A4), Eq. (A7) and Eq. (A8)) (Donovan et al. 1993; Liu et al. 2009). It should be noted that the impact of saturation is not unique to this method and may have especially large impact on observations that attempt to identify HOIC by high specular volume backscatter coefficient and low depolarization (Sassen et al., 2003; Hu et al., 2009; Sassen, 2005; Noel and Chepfer, 2010). To verify the CAPABL’s diattenuation measurement, a fourth polarization measurement was added in October 2011 to diagnose the effects of detector saturation of the $F_{12}$ term.

CAPABL obtains parallel, perpendicular and 45° measurements using $\Gamma_{wp}=-\pi, 0, \pi$, respectively (where $F_{33} < 0$). By adding a fourth measurement at an arbitrary $\Gamma_{wp}$ ($\pi<2|\Gamma_{wp}|<\pi$), diattenuation may be calculated where the presence of saturation has an opposing effect compared to its impact on Eq. (A7). Saturation causes diattenuation to increase in Eq. (A7), while when using the fourth measurement, it causes the diattenuation to decrease. This relationship provides a useful filter and diagnostic for saturation in our measurement set.
Detector nonlinearity has been identified as the most significant contributor to error in CAPABL’s diattenuation measurements. If any of the component measurements have lower photon count gain due to detector nonlinearity the diattenuation estimates will be biased. For diattenuation to be measured as zero, the observed photon counts in the 45° channel must represent 50% of the full received power. When the parallel channel reports less than the actual number of received photons, the measured diattenuation shifts positive. Thus, detector nonlinearity presents an issue for false positives in detection of HOIC. To test our confidence in cloud data, the corresponding photon count rate of diattenuating signals are checked to determine if they are within the linear range of the detectors.

A second issue relating to detector nonlinearity arises when low altitude stratus clouds are present. These events result in very high volume backscatter coefficient levels at low altitudes as well as high extinction, so volume backscatter coefficient data cannot be retrieved above such clouds. In these cases, saturation effects corrupt the feedback signal of the LCVR controller. When this happens, the feedback loop controls the profile to cancel the apparent positive diattenuation induced by detector nonlinearity. An attempt has been made to turn off the feedback loop when diattenuation is present in low altitude clouds and corrupts the feedback signal. However, drift in the LCVR phase shift can occur and it is difficult to support findings of oriented scatterers under these conditions. For this reason, diattenuation data is generally ignored when the feedback signals are corrupted by saturation and where no clear zero diattenuation baseline exists in the observed profile.

A6 Volume Backscatter Coefficient Retrieval
An additional data product from CAPABL is the lidar volume backscatter coefficient ratio associated with the presence of aerosol. CAPABL uses the retrieval method for aerosol volume
backscatter coefficient originally employed and described by Fernald et al. (1972) and Klett (1981). This method is in wide use (Russell et al., 1979; Fernald, 1984; Thayer et al., 1997; Hofmann et al., 2003; Pappalardo et al., 2004) and is only paraphrased here to explicitly describe the method employed for this lidar.

Any elastic volume backscatter coefficient lidar transmitting light at wavelength \( \lambda \), assuming single scattering, is described by the lidar Eq. (A13):

\[
N(\lambda, z, t) = N_L [\beta(\lambda, z, t) \Delta R] \frac{\lambda}{2 \pi} \exp \left[ -2 \int_0^z \alpha(\lambda, z') \, dz' \left[ \eta(\lambda, z, t) \right] \right] G(\lambda, z, t) + N_B(\lambda, z, t). (A13)
\]

For CAPABL this equation is a scalar version of Eq. (A1), where the parallel and perpendicular polarized components of the received signal have been summed into a single profile. Here \( N(\lambda, z, t) \) is the total number of scattered photons detected by the receiver, \( \lambda \) is the wavelength of the transmitted light, \( z \) is altitude above the surface (adjusted for the pointing angle of the lidar), \( N_L \) is the number of transmitted photons, \( \beta \) is the volume backscatter coefficient of the scatterers, \( \alpha \) is the extinction coefficient of the atmosphere, \( G \) is the geometric overlap function of the transmitter and receiver, \( \eta \) is a system efficiency parameter, \( N_B \) is the background number of photons detected and \( t \) is time.

The volume backscatter coefficient and extinction coefficients may be separated into molecular (\( m \)) and aerosol (\( a \)) components:

\[
\beta(\lambda, z, t) = \beta_a(\lambda, z, t) + \beta_m(\lambda, z, t) \quad (A14)
\]

\[
\alpha(\lambda, z, t) = \alpha_a(\lambda, z, t) + \alpha_m(\lambda, z, t) + \sum^n \sigma_{X_i}(\lambda) \sigma_{X_i}(r) \quad (A15)
\]

The molecular volume backscatter coefficient in the atmosphere may be determined empirically through the hydrostatic equation and quantum mechanics, but the volume backscatter coefficient
from aerosol is quite variable and must be determined for each measurement (Elterman, 1964; Russell et al. 1979). The extinction coefficient has an additional term that accounts for extinction due to absorption by different trace molecules in the atmosphere.

The volume backscatter coefficient ratio is defined as the ratio of the total volume backscatter coefficient to the molecular volume backscatter coefficient. Physically, the lidar volume backscatter coefficient ratio (LBSR) is defined as:

$$LBSR(\lambda, z, t) = \frac{\beta_a(\lambda,z,t) + \beta_m(\lambda,z,t)}{\beta_m(\lambda,z,t)} \quad (A16)$$

where $\beta_a(\lambda, z, t)$ and $\beta_m(\lambda, z, t)$ are the aerosol and molecular volume backscatter coefficient coefficients, respectively. The molecular volume backscatter coefficient is calculated from temperature and pressure profiles obtained twice daily from co-located radiosonde launches. The scattering ratio is retrieved from raw lidar by evaluating

$$LBSR(\lambda, z, t) = \frac{CS(\lambda,z,t)z^2}{\beta_m(\lambda,z,t)T^2(\lambda,z,t)} \quad (A17)$$

where $S(z)$ is the background subtracted lidar signal, $T^2(z)$ is the two-way atmospheric transmittance and $C$ is a system constant determined by normalizing the right-hand side of the equation to an expected minimum value of LBSR over a specified altitude range.

Russell et al., (1979) showed how small deviations in the top of the profile used to derive the molecular profile, attributed to aerosol scattering, can cause biases in the aerosol profiles. Russell et al., (1979) and Neely III et al., (2011) also show that simply including a small correction to account for additional aerosol scattering at the calibration altitude can improve the retrieved aerosol profiles when a topside calibration altitude is chosen erroneously to be aerosol free. For CAPABL, extinction measurements from the Optical Spectrograph and InfraRed Imager System (OSIRIS) aboard the Odin spacecraft where used to constrain the minimum
LBSR value in the calibration region (Bourassa et al., 2011). This was done by converting the observed OSIRIS extinction to LBSR during periods when both OSIRIS and CAPABL coincidentally observed clear sky above Summit (within 5°). LBSR values were obtained at these times at the highest altitude to which CAPABL observed photon counts with signals to noise error less than 5% (Ansmann et al., 1992; Jäger and Deshler, 2002; 2003). This process was performed several times in different seasons and annually averaged R values are applied to the dataset to derive the LBSR for all observations (Ansmann et al., 1992). As observations continue, this process will be repeated to help reduce error in the derived LBSR.

The transmittance is calculated from a combination of a radiosonde-derived molecular extinction model, lidar-derived aerosol extinction and modeled ozone absorption. During periods of moderate to heavy aerosol loading, aerosol extinction must be scaled to the aerosol volume backscatter coefficient (Jäger and Deshler, 2002). Under background conditions in the stratosphere, it can be ignored. Eq. (A17) is then solved iteratively from the top of the profile down, using an updated value of aerosol extinction for each iteration. The lidar extinction to volume backscatter coefficient ratio is assumed between 35 and 60 sr according to the values derived from in situ balloon-borne measurements (Jäger and Deshler, 2002; 2003). Differences in the lidar ratio are not critical to derive the volume backscatter coefficient in small aerosol loading conditions, but after a large volcanic perturbation the value is very important to derive the extinction coefficient from the volume backscatter coefficient due to differences in the size distribution of the aerosol.

A7 Example Measurement

CAPABL has run nearly continuously since June 2010, collecting polarization data utilizing the P45P technique. During the first year of observations the lidar was fixed in a near-
zenith pointing direction. In October 2011, the lidar was tilted about 11° from zenith. A set of example observations made on 15 November 2010 (zenith pointing) and 18 February 2012 (11° tilt) are shown in Fig. A3 and A4. The clear-air normalization region was set at 7 km for both of these observations (all observations are shown with heights relative to the elevation of Summit Station). A 7 km altitude was chosen due to the signal to noise limit set by the current volume backscatter coefficient retrievals. A geometric overlap correction (up to a range of 200 m) is applied to aerosol volume backscatter coefficient data products based on obtaining a volume backscatter coefficient ratio of unity during a very clean air episode.

Figure A3. Example of the total linear depolarization observations made by CAPABL with a vertical resolution of 30 m and a temporal resolution of 110s. This observation is from November 15, 2010; a day that included several cloud systems with precipitating snow and ice broken in the middle of the day by clear sky. Of particular note is a seeder-feeder cloud system at 20:00 UTC. The color threshold in the total linear depolarization scale is set at a value of 0.08, which is the typical level used to distinguish between liquid and ice for linear depolarization observed from randomly oriented particles (Sassen, 1992). To help illuminate areas of liquid water (characterized by high volume backscatter coefficient and low linear depolarization ratio) the LBSR is also shown for this day.
Figure A4. Time evolution of a strong variation in diattenuation (indicated by the light blue regions from 3:00 to 6:00 UTC) thought to be a possible HOIC detection on February 18, 2012. The top panel is the total linear depolarization observation made simultaneously with the diattenuation measurement using the 45° channel shown here with a vertical resolution of 30 m and a temporal resolution of 110 s. Again, the color threshold in the total linear depolarization scale is set at a value of 0.08. This draws a definitive line between areas containing scatters of pure liquid (spheres) and scatterers containing ice (non-spherical scatterers). A similar threshold at ±0.1 was set for the diattenuation plot to help distinguish the HOIC event from non-diattenuating signals and noise.
Figure A3 shows the linear volume depolarization ratio, $\delta$, and the LBSR with a vertical resolution of 30 m and a temporal resolution of 30 s for 15 November 2010. This day included several cloud systems with precipitating snow and ice with a period of clear air in the middle of the day. Of particular note is a seeder-feeder cloud system at 20:00 UTC (Reinking and Boatman, 1986). The distinction between a liquid layer and ice precipitation is made when comparing the depolarization ratios with the LBSR in a manner similar to previous work. Liquid water is characterized by high volume backscatter coefficient and low depolarization ratio while the ice has relatively lower LBSR and much higher depolarization ratios. At the time of this observation the lidar was pointing near zenith. Therefore, ambiguity exists in determining cloud phase because an oriented ice cloud would produce similarly high LBSR and low depolarization values as liquid water. Analysis of diattenuation using all three polarization measurements is close to zero but is inconclusive as HOIC observed at near-normal incidence will produce a value of zero as will liquid water. This expectation was used as a diagnostic to understand potential false positives in oriented scatterer detection within our entire dataset. The example shown in Fig. A3 demonstrates the ability of CAPABL to determine the phase of hydrometeors in various situations assuming randomly oriented scatterers prevail.

Over the entire observational period, a large effort has been made to establish the measurement setup and conditions required to reliably observe diattenuation with CAPABL. This has been done to ensure confidence that detection of a non-zero diattenuation signature is attributable to HOIC. During this process, CAPABL was tilted from 2° to 11° off zenith and the HWP was added above the last steering mirror in the transmitter. After tilting the lidar, several oriented scattering occurrences were observed.

On 18 February 2012 (Fig. A4), CAPABL observed two diattenuation signatures that
coincided with clouds at altitudes between 3500 and 4500 m from 2:00 to 5:00 UTC and between 1000 and 2500 m from 5:00 to 6:30 UTC. During this same period, signals at lower altitudes depict variable linear depolarization with no concomitant diattenuation. The observations in Fig. A4 suggest that the diattenuation signature is not caused by detector nonlinearity. First, the depolarization calculations contain the stronger parallel signal in the denominator. If detector nonlinearity is occurring, this would cause the denominator to be smaller than its actual value (due to the inhibited ability of the detector to record high count rates) and result in increased depolarization. Second, we observe some detector nonlinearity in the diattenuation profiles at many times close to the surface. Using the filters developed during periods of known clean sky and using the fourth polarization channel, these data have been removed. In this example, the photon count rate of the diattenuating clouds is much lower than count rates observed near the surface where impacts of detector nonlinearity are detected and filtered out. Therefore, it is unlikely that detector nonlinearity is responsible for the diattenuation signature observed. This observation was also confirmed by use of the fourth polarization channel. That measurement produced a diattenuation profile consistent with the observations shown in Fig. (A4) during the HOIC event and behaved oppositely, as described in section A5b, when encountering the impacts of saturation near the surface.

This behavior is also shown in Figure A5, which contains an integrated profile from 5:30 to 6:00 UTC on February 18, 2012 (a section from the whole day shown in Fig. A4). The two diattenuation profiles demonstrate how CAPABL’s separate measurements help determine the difference between false positive diattenuation due to detector saturation and actual variations in diattenuation. Regions where both diattenuation profiles track together, above the error limits, contain positive detection of diattenuating scatterers. Regions where the two diattenuation
profiles behave oppositely, as is seen in the bottom of the profile, is due to detector saturation as described above.

This observation provides a demonstration of operationally detecting HOIC by direct polarization determination. Furthermore, CAPABL can simultaneously determine the cloud phase of randomly oriented scatterers and assess variation in the diattenuation of the scatterers, which may be used to interpret the presence of HOIC. The measurements presented in Figures A3 and A4 illustrate the benefits of a multi-polarization channel lidar in studying polar atmospheric processes; particularly those involving the phase of water.

Figure A5. Time integrated profile (30 m vertical resolution) from 5:30 to 6:00 UTC on February 18, 2012 (a section from the whole day shown in Fig. 4). The left panel shows diattenuation calculated from the 45° channel and the 4th channel. On the right is the total linear depolarization observation made simultaneously with the diattenuation measurements. The dashed lines represent the error associated with each derived observation obtained through standard propagation of error techniques of the signal to noise error associated with the observation from each polarization.
A8 Summary

Recent rapid melting of Arctic sea ice is likely influenced by changes in cloud cover, radiation and circulation (Francis and Hunter, 2006; Kay et al., 2008; van den Broeke et al., 2009; Shupe et al., 2011). Signatures of climate change are known to be most evident in the polar regions (Washington and Meehl, 1989). Thus, it is not surprising that concurrent with the dramatic sea-ice losses, the GIS is experiencing similar rapid melting (Rignot and Kanagaratnam, 2006). Detailed information on cloud amount and type is needed to accurately determine the effect of climate change on snowmelt by using energy-balance in global climate models (Cawkwell and Bamber, 2002). Many shortcomings in numerical models are likely caused by unrealistic assumptions or parameterizations of cloudiness due to a shortage of observations over the GIS (Cawkwell and Bamber, 2002). Accurate quantitative measurements of atmospheric aerosol, especially the determination of the thermodynamic phase of hydrometeors, is essential to further our understanding of the effects of clouds and aerosol on the radiative budget of the GIS.

CAPABL uses recent advances in polarization theory (Hayman and Thayer, 2012) to develop techniques that better quantify the information needed to help understand the microphysical properties of clouds and how these properties are changing climatically over the GIS. Observations indicate that CAPABL can accurately determine altitude profiles at high spatial and temporal resolution of the aerosol volume backscatter coefficient ratio, the linear depolarization ratio, and a new data product called linear diattenuation through the combination of three polarization channels. Through careful configuration of the lidar system, polarization effects of the system are minimized and errors in depolarization and diattenuation estimates are below a few percent and primarily limited by photon counting statistics. The first observations of diattenuation in atmospheric scatters are demonstrated and shows promise in unequivocal
detection of horizontally oriented ice crystals. CAPABL incorporates self-verification of atmospheric diattenuation by incorporating a fourth polarization channel to check against false positive diattenuation detection due to detector saturation. This ability has also been used, when no diattenuation is present, to access the impact of saturation in other observations.

Since the measurement period began in June 2010, the necessary configuration to reliably measure diattenuation with CAPABL has been established. This has ensured confidence that detection of a non-zero diattenuation signature is attributable to atmospheric scatterers. The data shown here demonstrate CAPABL’s ability to detect and quantify polarization signatures that may be used to assess the occurrence of HOIC. A successful campaign detecting HOIC can have broad implications for our understanding of the radiative budget. HOIC lead to increased cloud albedo, which leads to a proportional reduction in the surface solar flux (Sassen et al., 2003). Thus, a climatology of HOIC, in conjunction with the full array of cloud parameters collected by ICECAPS, is needed to understand the consequences orientation may have for the heating of the atmosphere and the surface. Also, HOIC, when scattering normal to its surface, can exhibit low linear depolarization ratios that can result in erroneous classification of thermodynamic phase. Diattenuation measurement enables lidar systems to detect oriented scatterers within the same dynamic range as other cloud signals. This new observational method therefore allows for an easier and more certain means of collecting comprehensive climatological observations of clouds, oriented particles and their radiative impact.

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APPENDIX B: Raman Lidar Profiling of Tropospheric Water Vapor over Kangerlussuaq, Greenland


Abstract
A new measurement capability has been implemented in the Arctic Lidar Technology (ARCLITE) system at the Sondrestrom upper atmosphere research facility near Kangerlussuaq, Greenland (67.0° N, 50.9° W) enabling estimates of atmospheric water vapor through the troposphere. A balloon campaign was simultaneously conducted to calibrate and validate the new lidar water vapor measurements. Initial results show that height-resolved profiles up to 10 km with better than 10% statistical error are obtained with 30 minute integration and 250 m height resolution. Comparison of the lidar observations with water vapor profiles retrieved by the AIRS instrument on board the AQUA satellite agree within the error associated with each measurement. These new observations offer more routine measurements of water vapor in the Arctic to complement measurements related to the Arctic’s hydrologic cycle.
B1 Introduction

Though water vapor is a minor species in the upper troposphere and lower stratosphere, its impact on the Earth’s atmospheric radiative and chemical budget makes it of major importance. Its distribution influences many physical and chemical properties of the troposphere and stratosphere including polar stratospheric clouds and the Arctic and Antarctic ozone holes. Water vapor is particularly important to Earth’s energy budget, influencing both incoming solar radiation and outgoing infrared radiation. Variations in the total amount of atmospheric water vapor are natural and normal but changes in long-term trends in its vertical distribution, especially in the upper troposphere and lower stratosphere, may be indicative of changes in the Earth’s climate (Houghton et al., 2001). Global trends in stratospheric water vapor concentrations have been identified as a significant contributor to both stratospheric cooling and tropospheric warming (Forster and Shine, 2002). Recently upper tropospheric water vapor has also been recognized as an important driver of decadal global surface climate change and decreases in water vapor over the last decade may have acted to slow the rate of global warming (Solomon et al. 2010).

In addition to its role in the radiation budget, long-term trends in water vapor also play a role in the destruction of ozone through the HOX cycle. This may also cause changes the NOX/ClOX family, which could further deplete ozone through catalytic reactions during the formation of the Arctic ozone hole (Stenke and Grewe, 2005). Also, because of the indirect effect water vapor has on the radiation budget through cloud formation, changes in water vapor can cause changes in heterogeneous ozone chemistry through polar stratospheric cloud formation (Stenke and Grewe, 2005).

Water vapor profiles of the Arctic atmosphere, where no long-term records exist and current measurements are scarce, are particularly important because climate change has had the
largest effect to date in this region (Houghton et al., 2001; Blanchet and Girard, 1995; Solomon et al., 2007). More observations are particularly needed to understand the complex set of feedback cycles that involve water vapor as the Arctic atmosphere responds to climate change.

The Arctic Lidar Technology (ARCLITE) facility, a Rayleigh/Mie/Raman lidar system, has been in operation at the Sondrestrom Upper Atmospheric Research Facility, near the town of Kangerlussuaq (67.0° N, 50.9° W), since 1993 (Thayer et al., 1997). Molecular and aerosol volume backscatter coefficient is measured at 532 nm to retrieve temperature profiles from 35 km to 80 km and aerosol information from the troposphere to the mesosphere. A Raman channel for molecular nitrogen (608 nm) was implemented in 2008 to help determine aerosol extinction values. This allowed the derivation of temperature profiles to be extended into the troposphere. Most recently a Raman water vapor (661 nm) channel was added in February, 2010 to measure water vapor mixing ratio profiles into the upper troposphere and lower stratosphere. Currently there are few ground-based measurements of water vapor in the upper troposphere and lower stratosphere in the Arctic and none exist which could be compiled into a climatology. This new capability of the ARCLITE system fills this void in the face of numerous questions about the role of water vapor in climate change. The water vapor analysis of the Raman signals will be the focus of section B2 and section B3, and the derived lidar estimates of water vapor will be calibrated with balloon profiles of water vapor in section B4. In section B5, comparisons will be shown with additional balloon flights and profiles retrieved by the AIRS satellite.

**B2 System Description**

The ARCLITE system employs a monostatic lidar configuration with the transmitter and the center of the telescope separated by about 1.3 meters. The ARCLITE transmitter consists of a 30-Hz, 42-W Spectra-Physics Nd:YAG laser, with injection seeding, a second harmonic...
generator producing 530 mJ pulses at 532.0 nm, and a 5x beam expander reducing the laser beam divergence to better than 0.1 mrad. The receiver consists of a 92-cm diameter, f/2.2 Newtonian telescope and a side-mounted optical breadboard that holds receiver optics and detectors. The telescope field of view is adjustable but typically set to 0.5 mrad. This basic layout remains similar to that described by Thayer et al. (1997).

Over time, the ARCLITE system has undergone modifications to enhance its measurement capabilities, including addition of cross-polarized 532 nm channels for aerosol polarization ratio estimates, and 608 and 661 nm Raman receiver channels. The Raman signals will be the focus of this present work. Figure B1 illustrates a portion of the ARCLITE receiver path that includes the 532 nm Rayleigh/Mie receiver channel and the Raman receiver channel. The salient features for the Raman receiver path are numbered in Figure B1 and defined in Table B1.
Figure B1. Depiction of modifications made to the original ARCLITE design, described in Thayer et al. (1997), to enable profiling of water vapor mixing ratio. Characteristics of optics used in the two signal channels for the measurement of water vapor mixing ratio. All values are reported as the percent transmission of the particular signal through the optic, unless noted.
Table B1. Characteristics of optics used in the two signal channels for the measurement of water vapor mixing ratio. All values are reported as the percent transmission of the particular signal through the optic, unless noted.

As the 532 nm pulses serve as the excitation wavelength for the Raman-shifted return signals from molecular nitrogen and water vapor, the first dichroic beam splitter, labeled 1 in Figure B1, reflects the 532 nm signal while transmitting longer wavelengths. A folding mirror directs the longer wavelength signals through a field iris and a mechanical chopper. The mechanical chopper is synchronized with the laser transmitter and blocks near-field signals that may exceed the linearity of the photomultiplier tubes (PMTs). For the Raman signals under study, the near-field signals are geometrically limited by the laser beam/telescope field of view overlap function. The overlap function becomes unity at ranges in excess of 2 km and the Raman signals above those altitudes are weak enough to insure a linear response by the PMTs. Thus, the chopper is not used in this measurement scheme.

The second dichroic beam splitter reflects the 608 nm molecular nitrogen Raman signals and passes them through a filter stack that rejects any potential signal contamination by 532 nm signals, as detailed in Table 1. The filter stack consists of a long-pass filter, a 532 nm band-stop filter, and a bandpass filter (0.5 nm centered on 607.7 nm). The 661 nm water vapor Raman signal is transmitted through the dichroic beam splitter, passes through its filter stack, and detected by a new Hamamatsu detector, model H7422P-40. The 661 nm filter stack is similar to the 608 nm filter stack except the bandpass filter is 0.5 nm centered at 660.8 nm.
The associated signal transmissions for the three primary wavelengths are detailed in Table B1. A large net rejection (>23 optical density) of the 532 nm (Rayleigh and aerosol) backscattered light insures little contamination of the Raman signals by the 532 nm signal. This is an important factor when trying to measure the much weaker ($10^3 - 10^5$ less volume backscatter coefficient) Raman scattered light and derive physical quantities from these measurements on the order of a few parts per million. Without this precaution contamination could easily bias the derived water vapor mixing ratio. The two Raman optical paths are designed to be symmetric so that both channels will have the same near-range overlap function with the telescope. Thus, upon taking the ratio of the two Raman signals, the geometric overlap function, in principle, cancels out. The two PMTs are different and a calibration procedure is discussed in the next section to address this disparity in signal response.

**B3 Retrieval Algorithm**

In the Raman water vapor lidar technique (see Whiteman, 2003a; b; Sherlock et al., 1999a), the ratio of Raman volume backscatter coefficient from water vapor and molecular nitrogen is proportional to the water vapor mixing ratio. This is possible due to the well mixed nature of molecular nitrogen in the atmosphere below 80km. Following the traditional approaches developed by Whiteman (2003a; b) and Sherlock et al. (1999a), the water vapor mixing ratio as a function of height, $q(z)$, may be expressed in terms of the two lidar signals ($N_x$), a calibration constant ($C$) and a differential transmission term, $\Gamma(z)$:

$$ q(z) = C \Gamma(z) \frac{N_{H_2O} - N_{B,H_2O}}{N_{N_2} - N_{B,N_2}},$$  \hspace{1cm} (B1)$$

where $N_x$ is the total photon counts from each detector and $N_{B,x}$ is the estimated noise counts caused by background skylight and thermal noise of each detector (Sherlock et al., 1999a). $\Gamma(z)$ is an atmospheric differential transmission term, (B2), accounting for the wavelength differences
in relative extinction of the two Raman volume backscatter signals. This term is traditionally defined as:

\[ \Gamma(z) = \frac{\exp\left[-\int_{z_0}^{z} \alpha(\lambda_{N_2}, z')dz'\right]}{\exp\left[-\int_{z_0}^{z} \alpha(\lambda_{H_2O}, z')dz'\right]}, \]  

(B2)

where \( \alpha \) is the wavelength-dependent extinction coefficient (Whiteman, 2003a; b; Sherlock et al., 1999a). According to Sherlock et al. (1999a) the error associated with the omission of this term is \( \Gamma(z)^{-1} \). In the planetary boundary layer, this error is less than 5% and above it drops to less than 0.2%. Because it is such a small factor compared to the relative error from other sources and the fact that reliable simultaneous measurements or models are not available to derive this value, it is assumed to be one for the actual retrieve profiles shown here.

The calibration constant, \( C \), that accounts for differences in optical path and transmission for the two wavelengths, differing Raman cross sections and physical constants related to water vapor, air, and molecular nitrogen needed to derive the mixing ratio (B1), may be defined as:

\[ C = \frac{\int L_{N_2}(\lambda) \frac{M_{H_2O}}{M_{dry\,air}} \frac{n_{N_2}}{n_{dry\,air}}}{\int L_{H_2O}(\lambda) \frac{M_{H_2O}}{M_{dry\,air}} \frac{n_{H_2O}}{n_{dry\,air}}}, \]  

(B3)

Here, \( L_{i}(\lambda) \) accounts for differential transmission of the receiving optics (Sherlock et al., 1999a). The differential Raman volume backscatter cross section, \( \sigma(\lambda) \), of a particular wavelength is in principle a function of temperature and pressure and will vary under different atmospheric conditions. In this retrieval the ratio of the molecular nitrogen to water vapor cross section from Penney and Lapp (1976) is used to derive the calibration constant. Though this parameter is known to vary with atmospheric conditions, principally with temperature (e.g., (Whiteman, 2003a)), this dependence has been omitted. The last two constants pertain to the conversion or relative numbers of photons to the mixing ratio of water vapor (Whiteman, 2003a; b; Sherlock et al. 1999a). \( \frac{M_{H_2O}}{M_{dry\,air}} \) is the molecular mass ratio of water vapor to dry air and
\[ \frac{n_{N_2}}{n_{\text{dry air}}} \] is the number density fraction of molecular nitrogen in dry air assuming a well mixed atmosphere (Whiteman, 2003a; b; Sherlock et al., 1999a).

An additional correction may be needed to account for the overlap between the telescope and laser for ranges close to the instrument (Halldorsson and Langerholc, 1978). The calibration constant, C, is needed to account for uncertainties in transmissions, reflectivities and sensitivities of the optical and electronic components. Two balloon launched Vaisala RS-80 radiosonde profiles have been used to independently derive this single constant. In addition, a separate calibration technique using only the nitrogen signal and physical properties of the measurement was developed. This independent estimate will be shown to have accuracy with error of similar order to the balloon calibration technique and may be performed routinely to check degradation of the signal paths and detectors over time.

The relative error (B4) of the derived mixing ratio profile (B1) was found by employing standard propagation of error techniques to the lidar signal and retrieval algorithm (Thayer et al. 1997).

\[
\frac{\sigma_g(x)}{q(x)} = \sqrt{\frac{\sigma_{C^2(x)}^2}{C^2(x)} + \frac{\sigma_{R_4}^2(x)}{R_4^2(x)} + \frac{\sigma_{r^2(x)}^2}{r^2(x)}} \quad (B4)
\]

The terms contributing to the net relative error are the calibration constant relative error, the lidar signal ratio relative error, and the relative error associated with the uncertainty in the differential transmission; all summed in quadrature assuming the errors are independent and random. The net relative error is predominantly affected by the signal ratio error and the calibration error. The signal ratio error is statistical and assessed assuming Poisson statistics for the lidar signals, where the variance is the mean of the signal counts. Thus, this error relates to the amount of water vapor and molecular nitrogen in the atmosphere and, in general, increases
with altitude due to decreasing signal levels. The calibration error relates to the method used to determine the calibration constant. This is discussed in more detail in the next section.

**B4 Calibration**

The sensitivity of the Earth’s radiation budget to water vapor variations requires accuracy on the order of 3%-10% to fully understand and quantify water vapor related radiative impacts on climate change (Leblanc and McDermid, 2008b). This required accuracy demands signal counts to be large in order to keep the signal ratio percent error in single-digit percentages. This is accomplished by the high power-aperture product of the ARCLITE system and through temporal and range integration. As will be shown, signals integrated for 30 minutes with a range resolution of 250 meters achieve single-digit percent error through the troposphere. The calibration error must also be single-digit percentages to achieve useful water vapor estimates but has the added complication of possibly introducing systematic errors to the estimate. A common calibration method is to use balloon-launched radiosondes in the local vicinity of the lidar beam to independently estimate water vapor and, thus, retrieve the calibration constant by forcing the lidar estimate to equate to the balloon estimate (Sherlock et al., 1999b; Whiteman et al., 2000). However, balloon instruments have their own inherent set of problems that must be considered when calibrating the system. Therefore, a system calibration procedure was developed to provide a second method for calibration to constrain the systematic effects introduced by the balloon measurement. The second method evaluates lidar system variables that contribute to the calibration constant by performing a signal assessment of the two Raman receiving channels and then deriving the calibration constant. Both methods and their results are described below.

The first attempt at deriving a calibration constant for the new lidar channel was
accomplished by using coincident radiosonde, RS80-Hs, measurements and forcing the lidar signal to match the balloon profile within an altitude range of relatively high signal to noise and homogenous water vapor signal. The accuracy of the humidity measurement on radiosondes has been shown to vary with the sensor type and individual instruments. Especially noteworthy is a dry bias at low humidities and a time lag at low temperatures (Ferrare et al., 1995; Miloshevich et al., 2001). Coincident flights with RS80-H and cryogenic frost point hygrometers at NOAA’s Mauna Loa Observatory in Hawaii have shown that the RS80-H deviates where the temperature drops below -55°C (Vömel et al., 2003; 2007). In order to minimize this effect as well as the dry bias, the radiosondes were flown on relatively wet nights and the upper tropospheric data were not used for the calibration of the lidar. The altitude range from 3 to 7 km from two balloon flights were used to determine the calibration constant. Between 1 and 3 km the lidar has incomplete overlap with the telescope so data from this region are not used for calibration. Differences between sonde and lidar derived profiles may be due to the spatial and temporal inhomogeneity of water vapor in the atmosphere. This causes physical differences in the measurement of the sondes and the lidar—the balloons covered a flight path of over 50 km horizontally—which may cause large differences in the concurrent profiles.

An independent calibration is needed to truly determine the accuracy of the lidar measurement (Whiteman et al., 2000; Leblanc and McDermid, 2008a). In our second calibration approach, an independent assessment of the two Raman channel detectors and their optical paths was conducted by measuring a redundant nitrogen Raman profile through the optical path of the water vapor channel. This allowed for differences in the optical path, geometric overlap, and PMT gains to be isolated and removed using the relative intensities of the two profiles. When measuring the nitrogen signal along the water vapor path, the final dichroic mirror was removed
and the water vapor bandpass filter was replaced by the nitrogen filter (Figure B1). The error involved in this calibration includes the wavelength differential in the sensitivity of the optics and water vapor PMT to the nitrogen signal. From this information, a calibration constant was derived using only the nitrogen signal. This technique avoids errors associated with the sonde profile calibration technique caused by inhomogeneity of water vapor in the atmosphere because it only depends upon the nitrogen signal, which may be assumed to be well mixed. Though this is not a true absolute calibration of the system, as discussed by Sherlock et al. (1999b) and Whiteman et al. (2000), this method is an alternative to deriving the calibration constant from the traditional radiosonde method. Comparison of the separately derived constants showed less than 5% difference. Due to the need for long-term accuracy, as discussed by Leblanc and McDermid (2008a), and difficulty of regularly launching radiosondes at this location, the second calibration method discussed above will be implemented on a regular basis to check for system degradation that could cause measurement biases.

![Figure B2](image)

Figure B2. Water vapor mixing ratio profile comparison between a balloon flight and 30 minute integration of lidar signal on 2/15/2010. The lidar has been integrated spatially to 250m to match the balloon data. The subplot is the corresponding percent error in the signal of the lidar profile. Dashed lines represent the uncertainty associated with each measurement.
**B5 Validation**

Six additional balloon flights occurred during February 2010 after the new water vapor channel was installed and calibrated. These flights followed similar procedures as described by Barnes et al. (2008) and Leblanc and McDermid (2008b). One of the validation flights conducted during February 2010 is shown in Figure 2. The radiosondes relative humidity and temperature have been used to calculate mixing ratio, parts per million by volume (ppmv), using the Vaisala recommended conversion (Hyland and Wexter, 1983). The results of this flight are typical of other inter-comparison flights.

The dashed lines in Figure B2 represent the total error involved in each measurement and for the majority of the profile the lidar and radiosonde agree within these bounds. From 2 to 5 km the lidar tracks the layers seen by the radiosonde. This suggests that the feature is a stable layer within the free troposphere. Below 1 km, the deviation of the comparison increases due to the larger variability of water vapor in the planetary boundary layer. A curtain plot of mixing ratio that passed through the lidar beam during the balloon flight is shown in Figure B3 with an approximate location of the balloon’s height with time. The temporal variation we see in this plot may account for the discrepancy seen in comparing the individual balloon profile with the lidar profile in Figure B2. Given the data, error and variability, no low-altitude geometric overlap correction was deemed necessary and none has been applied in this analysis.
An illustration of how the lidar-derived water vapor estimates compare with the six validation balloon flights is given in Figure B4 in terms of a percent difference. The average percent difference from all six lidar-balloon comparisons is close to zero indicating no significant bias exists in the calibration constant. Individual balloon-lidar comparisons show deviations (independent with height below 3km) below 10%. Above 3 km, the lidar tends to become wetter (>2% at 7km) compared to the balloon measurements. This could be due to either a dry bias in the balloons at the colder temperature in the upper polar wintertime troposphere, the omission of a temperature dependence in the volume backscatter coefficient cross sections used to calibrate the lidar or could be due to the small number of samples included in the comparison (Whiteman, 2003b; Ferrare et al., 1995; Miloshevich et al., 2001). Furthermore, geophysical variance in water vapor may contribute to the lidar-balloon differences. This comparison leads to the conclusion that the lidar estimate for water vapor concentration is well within 10% statistical
A comparison with version 5 of the atmospheric humidity data collected by the AQUA Atmospheric Infrared Sounder (AIRS)(Olsen et al., 2007) was conducted as another validation of ARCLITE water vapor profiles. AIRS is a spectrally resolved infrared sounder with 2378 channels covering 650–2675 cm\(^{-1}\) that was launched on the EOS Aqua satellite on May 4, 2002 and flies in a satellite formation known as the afternoon, A train (Read et al., 2007; Olsen et al. 2007). AIRS retrieves \(\mathrm{H}_2\mathrm{O}\) on 28 height levels and, on the basis of radiosonde comparisons, the accuracy of the AIRS humidity data is 15% at 250mb(Read et al., 2007). Further information and validation of this data product may be found in the work of Read et al. (2007). For the comparison, the AIRS data was screened by the instrument’s recommended criteria to insure the data used were of high fidelity.

A mean AIRs profile is created from profiles taken on February 15, 2010 within one degree of latitude and longitude of the lidar site and within six hours of the mid point of the lidar data collection. Comparing AIRS data with the lidar and radiosonde data show fairly good agreement (Figure B5). The coarser resolution of AIRS fails to distinguish the detailed structure of the water vapor profile as observed by the radiosonde and lidar, but the AIRS profile does capture the mean trend of the water vapor profile. A comparison with lidar data collected ten months later (December 21, 2010) shows a similar result between the lidar and AIRS data, see Figure 6. No balloon was launched for this comparison and the calibration used to derive water vapor from ARCLITE was the same as the one derived during the February calibration and validation campaign. This comparison was done to demonstrate the stability of the lidar calibration used in these retrievals.

Figure B5 Comparison of balloon-sonde, ARCLITE and AIRS during validation campaign on February 15, 2010. AIRS profile is within one degree of the lidar site’s latitude and longitude.
Figure B5 Comparison of ARCLITE and AIRS on December 21, 2010. AIRS profile is within one degree of the lidar site’s latitude and longitude.

B6 Conclusion and Summary

We have described the design, calibration and validation of the Raman water vapor profiling channel in the ARCLITE system at the Sondrestrom Upper Atmosphere Research Facility near Kangerlussuaq, Greenland. These initial results suggest the current water vapor profiling setup will provide precise and accurate long-term measurements of water vapor in the upper Arctic troposphere and lower stratosphere. Currently results show that the lidar is within the range of the 3%-10% levels of accuracy needed to quantify changes in water vapor in order to assess impacts on climate change, as stated by Leblanc and McDermid (2008b). Due to the dry conditions of the Arctic atmosphere this level of uncertainty is not unexpected and will be validated at a later date with a second balloon comparison campaign and routine cross measurements of the Raman PMTs will be made to check for detector degradation. The aim of
this measurement is to build a climatology of water vapor profiles in order to assess the
dynamics of water vapor in the upper Arctic troposphere, a region that has few water vapor
measurements but may experience the largest effects. This measurement is of particular
importance in light of the recent findings of Solomon et al. (2010) who suggest that stratospheric
water vapor is an important driver in global temperature trends.

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