IN SITU AND SOLAR RADIOMETER MEASUREMENTS OF ATMOSPHERIC AEROSOLS IN BOZEMAN, MONTANA

by

Michael John Thomas

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ii

APPROVAL

of a thesis submitted by

Michael John Thomas

This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citation, bibliographic style, and consistency and is ready for submission to The Graduate School.

Dr. Joseph A. Shaw

Approved for the Department of Electrical Engineering

Dr. Robert C. Maher

Approved for The Graduate School

Dr. Carl A. Fox
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iv

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# TABLE OF CONTENTS

1. INTRODUCTION .................................................................................................................. 1
   - Background ......................................................................................................................... 1
   - Aerosol Overview ............................................................................................................... 1
   - Overview of the Thesis ........................................................................................................ 10

2. INSTRUMENTS AND CALIBRATION .................................................................................. 12
   - Nephelometer .................................................................................................................... 12
   - Measurement .................................................................................................................... 13
   - Measurement Characteristics ............................................................................................ 13
   - Physical Characteristics .................................................................................................. 14
   - Power ............................................................................................................................... 14
   - Internal Data Storage ....................................................................................................... 14
   - Operation ........................................................................................................................ 14
   - Calibration Procedure .................................................................................................... 17
   - MetOne Optical Particle Counter .................................................................................... 20
   - Calibration ....................................................................................................................... 21
   - Scanning Mobility Particle Sizer ....................................................................................... 22
   - Solar Radiometer ............................................................................................................ 26
   - Calibration ....................................................................................................................... 27

3. ANNUAL AEROSOL CHARACTERISTICS IN BOZEMAN, MONTANA ......................... 30
   - Introduction ...................................................................................................................... 30
   - Annual Scattering Coefficient ......................................................................................... 30
   - Aerosols Optical Depth in Bozeman ................................................................................ 40
   - Wind and Aerosols ........................................................................................................... 42
   - Conclusion ....................................................................................................................... 47

4. EXAMINING AERONET’S REAL REFRACTIVE INDEX IN LOW AEROSOL OPTICAL DEPTH
   CONDITIONS ......................................................................................................................... 48
   - Introduction ...................................................................................................................... 48
   - Method .............................................................................................................................. 49
   - Results and Discussion .................................................................................................... 57
   - Conclusions ...................................................................................................................... 65
TABLE OF CONTENTS – CONTINUED

5. AEROSOL SIZE DISTRIBUTION COMPARISONS OF SMPS VS AERONET .......................... 67
   Introduction ....................................................................................................................... 67
   Method .............................................................................................................................. 68
   Results and Discussion ................................................................................................. 74
   Conclusions .................................................................................................................... 84

6. GENERAL CONCLUSIONS .......................................................................................... 85

REFERENCES CITED ........................................................................................................... 89
LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1. Effective climate forcings used in the GISS climate model</td>
<td>4</td>
</tr>
<tr>
<td>3.1. AOD annual averages for several locations across the world</td>
<td>41</td>
</tr>
</tbody>
</table>
## LIST OF FIGURES

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.1</td>
<td>Earth’s annual energy budget</td>
</tr>
<tr>
<td>1.2</td>
<td>Idealized aerosol size distribution</td>
</tr>
<tr>
<td>1.3</td>
<td>Color intensity measured size distribution example</td>
</tr>
<tr>
<td>1.4</td>
<td>Measured size distribution example</td>
</tr>
<tr>
<td>2.1</td>
<td>Photo of Radiance Research M903 integrating nephelometer</td>
</tr>
<tr>
<td>2.2</td>
<td>Drawing of M903 nephelometer with labeled parts</td>
</tr>
<tr>
<td>2.3</td>
<td>Internal drawing of M903 nephelometer</td>
</tr>
<tr>
<td>2.4</td>
<td>Photo of MET ONE 327B optical particle counter</td>
</tr>
<tr>
<td>2.5</td>
<td>Optical particle counter operation</td>
</tr>
<tr>
<td>2.6</td>
<td>Photo of Scanning Mobility Particle Sizer</td>
</tr>
<tr>
<td>2.7</td>
<td>Drawing of TSI 2071A differential mobility analyzer</td>
</tr>
<tr>
<td>2.8</td>
<td>Drawing of TSI 3010 condensation particle counter</td>
</tr>
<tr>
<td>2.9</td>
<td>Photo of CIMEL 318A solar radiometer</td>
</tr>
<tr>
<td>2.10</td>
<td>Langley plots done in Bozeman, MT</td>
</tr>
<tr>
<td>3.1</td>
<td>Google map for layout of Gallatin Valley</td>
</tr>
<tr>
<td>3.2</td>
<td>Annual Scattering coefficients for 2010</td>
</tr>
<tr>
<td>3.3</td>
<td>Candle stick plot of annual scattering coefficients for 2010</td>
</tr>
<tr>
<td>3.4</td>
<td>Color intensity size distribution plot for a smoke event</td>
</tr>
</tbody>
</table>
# LIST OF FIGURES – CONTINUED

<table>
<thead>
<tr>
<th>Figure</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.5. Daily scattering coefficient plot for a smoke event</td>
<td>37</td>
</tr>
<tr>
<td>3.6. Photo of trapped pollution layer in the Gallatin Valley</td>
<td>38</td>
</tr>
<tr>
<td>3.7. Aerosol extinction for the United States</td>
<td>39</td>
</tr>
<tr>
<td>3.8. Compass Rose for wind direction for 2010</td>
<td>43</td>
</tr>
<tr>
<td>3.9. Daily wind direction and speed for a diurnal event</td>
<td>44</td>
</tr>
<tr>
<td>3.10. Color intensity size distribution plot for a diurnal event</td>
<td>45</td>
</tr>
<tr>
<td>3.11. Aerosol concentration vs. wind speed and direction plot</td>
<td>46</td>
</tr>
<tr>
<td>4.1. Flow chart of experiment procedure</td>
<td>50</td>
</tr>
<tr>
<td>4.2. SMPS and fit size distribution plots</td>
<td>54</td>
</tr>
<tr>
<td>4.3. Time series plots of measured and modeled scattering coefficients</td>
<td>58</td>
</tr>
<tr>
<td>4.4. Scatter plot of measured vs. modeled scattering coefficients</td>
<td>59</td>
</tr>
<tr>
<td>4.5. Scatter plots of measured vs. modeled scattering coefficients with adjusted refractive indices</td>
<td>60</td>
</tr>
<tr>
<td>4.6. Refractive index values for Bozeman and Mauna Loa during study period</td>
<td>63</td>
</tr>
<tr>
<td>5.1. Flow chart of experiment procedure</td>
<td>70</td>
</tr>
<tr>
<td>5.2. Peak radius values of accumulation modes for AERONET and the SMPS</td>
<td>75</td>
</tr>
<tr>
<td>5.3. Size distribution plots from SMPS and AERONET</td>
<td>77</td>
</tr>
<tr>
<td>5.4. Scatter plot of accumulation mode widths from AERONET and the SMPS</td>
<td>80</td>
</tr>
<tr>
<td>5.5. Time series plots of effective boundary layer height calculations</td>
<td>81</td>
</tr>
<tr>
<td>Figure</td>
<td>Page</td>
</tr>
<tr>
<td>--------</td>
<td>------</td>
</tr>
<tr>
<td>5.6. Scatter plot of effective boundary layer height calculations</td>
<td>83</td>
</tr>
</tbody>
</table>
Atmospheric aerosols play a very important role in Earth’s climate system. Aerosols provide one of the most significant uncertainties in the climate system, mainly due to aerosols differing greatly in different regions. Measuring these aerosols accurately and effectively is very important. Inversions of solar irradiance and sky radiance measured by solar radiometers, for example those within the AErosolROboticNETwork (AERONET), provide aerosol properties within different regions across the world. Aerosol properties retrieved by AERONET are generally found to be very accurate and have been a very reliable source for aerosol measurements, but there are concerns about the inversion accuracy in very clean environments with low aerosol concentration.

In this thesis, accuracy assessments are done on some of the aerosol properties retrieved by the AERONET inversion using in situ instruments making aerosol measurements at the ground. First, Bozeman, Montana and the surrounding Gallatin Valley’s aerosol characteristics are assessed to show why this area is a good place to do these studies. Then, the real part of the refractive index retrieved from AERONET is assessed and shown to be potentially about 3.5% too high during the study period for all aerosol concentration conditions. Lastly, the aerosol size distribution that is retrieved by AERONET is assessed and found to be very accurate under well-mixed conditions in the boundary layer. Overall, the AERONET inversions are very accurate during these very clean conditions, with evidence of small (~3.5%) linear biases in the real part of the refractive index.
Aerosols are a very important component of the earth’s climate system. Different kinds of aerosols affect heating and cooling of the earth in different ways. Aerosols occur both naturally as well from manmade sources and are highly variable in the troposphere. The ability to make accurate aerosols measurements is very important to climate studies. Aerosol measurements have been taken in Bozeman, Montana by a solar radiometer as part of the NASA-led world-wide AErosol RObotic NETwork (AERONET) [Holben et al. 1998] since January 2008. In situ measurements of parameters such as the aerosol size distribution and the scattering coefficient have been taken since August 2009. These measurements give a good description of the type of background aerosol properties that exist in Bozeman. A good understanding of the aerosols in Bozeman can lead to more in-depth studies on how to make the aerosol measurements with improved accuracy. This chapter will give a brief overview of general aerosol properties, especially those properties that will be discussed throughout this thesis, as well as describe an overview of some questions that are answered in the following chapters.

An aerosol is defined as a suspension of fine solid or liquid particles in a gas [Seinfeld 2006]. Atmospheric aerosols are particles that range in size from a few
nanometers (nm) to tens of micrometers (μm). Aerosols can be emitted directly as particles or formed in the atmosphere by gas-to-particle conversion processes. Aerosols are present almost everywhere from the stratosphere to the surface of the earth.

Aerosols generally have a short residence time in the atmosphere, usually around a few days to a few weeks. The main sources of these aerosols are natural, such as volcanoes, sandstorms, fires, and sea salt. The other small portion of aerosols is manmade. These aerosols come from processes such as fossil fuel combustion and biomass burning. The two primary mechanisms of aerosols being removed from the atmosphere are deposition at the earth’s surface and incorporation into cloud droplets. Because of their relatively short lifespan and spatially variable sources, tropospheric aerosols vary widely in concentration and composition over the Earth. This variability is one reason for the importance of studying aerosols, especially considering the important role of aerosols in the global climate system.

The earth’s climate is controlled by the amount of solar radiation intercepted by the planet and the fraction of that energy that is absorbed. Total solar irradiance is the dominant driver of global climate, and both natural and anthropogenic aerosols are climatically important constituents of the atmosphere that can contribute to either cooling or heating, depending on their location and optical properties. Figure 1.1 shows earth’s annual global energy budget and the sources of forcing [Trenbert 2009].
Figure 1.1 – The global annual mean of Earth’s energy budget for March 2000 to May 2004 (W/m²).

Figure 1.1 shows that during the period of March 2000 to May 2004 the net global effect of aerosols were to cool the Earth’s surface by scattering the incoming solar radiation back into space. Table 1.1 shows the estimated changes in climate forcings during the period of 1880-2003 [Hansen et al. 2005]. A positive change means a contribution toward climate warming, and a negative change means a contribution toward climate cooling.
Table 1.1 – Effective climate forcing (W/m²) used to drive the 1880 to 2003 simulated climate change in the GISS climate model [Hansen et al. 2005].

<table>
<thead>
<tr>
<th>Forcing agent</th>
<th>Forcing (W/m²)</th>
</tr>
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<tbody>
<tr>
<td>Greenhouse gases (GHGs)</td>
<td>--</td>
</tr>
<tr>
<td>Well-mixed GHGs</td>
<td>2.75</td>
</tr>
<tr>
<td>Ozone</td>
<td>0.24</td>
</tr>
<tr>
<td>CH₄-derived stratospheric H₂O</td>
<td>0.06</td>
</tr>
<tr>
<td>Total: GHGs</td>
<td>3.05 ± 0.4</td>
</tr>
<tr>
<td>Solar irradiance</td>
<td>0.55 (x2)</td>
</tr>
<tr>
<td>Land use</td>
<td>-0.09 (x2)</td>
</tr>
<tr>
<td>Snow albedo</td>
<td>0.14 (x2)</td>
</tr>
<tr>
<td>Aerosols</td>
<td>--</td>
</tr>
<tr>
<td>Volcanic aerosols</td>
<td>0.00</td>
</tr>
<tr>
<td>Black carbon</td>
<td>0.43</td>
</tr>
<tr>
<td>Reflective Tropospheric aerosols</td>
<td>-1.05</td>
</tr>
<tr>
<td>Aerosol indirect effect</td>
<td>-0.77</td>
</tr>
<tr>
<td>Total: aerosols</td>
<td>--</td>
</tr>
<tr>
<td>Sum of individual forcings</td>
<td>-1.39 ± 0.7</td>
</tr>
<tr>
<td>All forcings at once</td>
<td>--</td>
</tr>
<tr>
<td></td>
<td>1.80 ± 0.85</td>
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Table 1.1 shows that the estimated magnitude of the net aerosol forcing and its current uncertainty are comparable to those of the sum of all climate forcings [Mishchenko et al. 2007]. This shows that aerosols generally have a cooling effect on the Earth’s climate, but the large error indicates more need for accurately measuring and understanding aerosol effects on climate.

To further predict the impact aerosols have on climate change, it is important to be able to accurately measure and understand aerosols in all of Earth’s regions. To do this, knowledge of certain aerosol properties is needed. These properties include size,
refractive index, shape, and chemical composition. These properties can tell everything about how an aerosol affects scattering and absorption of light.

The size distribution of aerosols is one of the primary properties in understanding how aerosols are affecting solar radiation. An idealized aerosol size distribution is shown in figure 1.2, along with typical processes that lead to particle formation and transition from one mode to another.

Figure 1.2 – Idealized schematic of the distribution of particle surface area of an atmospheric aerosol [Whitby 1978]. Principal modes, sources, and particle formation and removal mechanisms are indicated.
Aerosols can be classified into four modes: the nucleation, the Aitken, the accumulation, and the coarse mode. The nucleation mode describes particles up to about 10 nm in diameter. The Aitken mode includes particles from 10 nm up to about 100 nm (0.1 µm). Particles in these two size ranges usually make up a small amount of the total number of particles. Particles in these size ranges are less stable and tend to have smaller life spans because they tend to coagulate with other particles and grow into larger size ranges. The accumulation mode contains particles with diameters from 0.01 µm to 2.5 µm. Particles in this mode are mainly made up of particles from the nucleation and Aitken modes growing into its size range from coagulation and from condensation of water vapor onto existing particles. This mode accounts for a large number of particles as well as a substantial part of the aerosol mass. The accumulation mode gets its name because particle removal mechanisms are least efficient in the regime, causing particles to accumulate here. The coarse mode consists of particles that have a diameter larger than 2.5 µm. These are particles that consist of dust and sand formed by mechanical processes. These particles tend to settle out of the atmosphere in reasonably short time periods.

Figure 1.3 is an intensity-modulated plot of an aerosol size distribution, to get a better idea of what a real measured aerosol size distribution looks like.
The intensity-modulated plot shows the size distribution vs. time, where the intensity modulation color represents the aerosol concentration. This size distribution shows sizes from the Aitken mode through the accumulation mode (no coarse mode).

Throughout this day, there are two distinct modes that are displayed as bands. Figure 1.4 is a slice of this plot taken at 03:36 Mountain Daylight Time (UTC-6). The two modes that look like bands in the intensity-modulated plot can be seen as two lognormal modes in Figure 1.4.
Aerosol extinction is another important property in understanding aerosol effects on solar radiation. Extinction can be broken down into two parts: scattering of light by aerosols and absorption of light by aerosols. Aerosol scattering and absorption coefficients (extinction) are functions of the particle size, complex refractive index, and the wavelength $\lambda$ of the incident light. A widely used model for scattering and absorption by small spherical particles is called Mie theory. Mie theory is very accurate in calculating scattering and absorption of spherical particles and can be used to calculate these values for most aerosols. The complex refractive index describes a particle’s optical density relative to the surrounding medium. With aerosols, the surrounding medium is air, which has a refractive index very close to 1 ($1.00029 - 0i$), so...
the ratio of the refractive index of the aerosols to air is basically identical to that of the aerosols. The complex refractive index of a particle is directly related to the scattering and absorption properties of that particle. The real part represents the scattering properties, while the imaginary part describes the absorption properties of a particle.

Particles with diameters between 0.1 \( \mu \text{m} \) and 1 \( \mu \text{m} \) (middle of the accumulation mode) scatter visible light most efficiently (visible light has a wavelength range of approximately 0.38 – 0.68 \( \mu \text{m} \)). The complex refractive index also has very specific effects on scattering and absorption properties. As the real part of the refractive index increases, the amount of light scattered increases and the peak of the scattering efficiency moves towards smaller particles. The range of the real part of the refractive index is around 1.3 to 2 for most aerosols. Increasing the imaginary part of the refractive index increases absorption, but the peak of the absorption efficiency is weakly affected. A value of zero for the imaginary part of the refractive index refers to zero absorption, whereas a value of around 0.79 is the imaginary part of the refractive index for black carbon (highly absorbing).

Aerosol optical depth (AOD) is another very useful measurement when looking at how aerosols affect light. Optical depth is a unitless and dimensionless quantity that describes the amount of total extinction in an integrated medium. In this case, it describes the total extinction of solar irradiance by the atmosphere. Optical depth varies with wavelength. Optical depth can be used to describe many different sources of extinction such as aerosol scattering and absorption, Rayleigh scattering, absorption
from gases, ozone, clouds, etc. AOD is obtained by taking the total optical depth (TOD) and subtracting out the optical depth from all other sources except the aerosols. AOD therefore describes the total extinction (scattering and absorption) through the atmospheric path length from aerosols.

**Overview of the Thesis**

The motivation behind this study was to answer some very important questions. These questions include the following:

- How clean is Bozeman and the surrounding Gallatin Valley?
- What causes us not to be clean and when do these events occur?
- How accurate is the AERONET real refractive index retrieval in very clean conditions (low AOD)?
- How accurate is the AERONET size distribution retrieval in very clean conditions?
- How well do in situ measurements and AERONET retrievals agree?

Studies were done to answer these questions as closely as possible. First, the instruments used in the study will be explained in detail as to understand what is making the measurements and the method of measurement that is being used. Next, a general overview of aerosols in Bozeman, MT will be discussed to understand normal conditions as well as different aerosol events throughout the year that may cause a rise in aerosol concentrations. Then, the accuracy of the real part of the complex refractive index retrieved by AERONET is assessed under very clean conditions to find out how
well these measurements can be made in Bozeman as well as how well these measurements represent the conditions at the ground. Lastly, the aerosol size distribution retrieved by AERONET is compared to the size distribution measured at the ground to find out how well these measurements agree, especially in very clean conditions.
Nephelometer

Figure 2.1 – Photo of the Radiance Research M903 integrating nephelometer (J. Shaw photo).

A nephelometer is an instrument that measures the optical scattering coefficient of aerosols contained in an air sample in the instrument’s internal chamber. The Radiance Research M903 Nephelometer measures light scattering at one wavelength, which is 530 nm. The M903 is a lightweight, portable, low-power instrument that measures light scattering using the geometry of a standard integrating nephelometer. Pressure and temperature of the air sample are monitored to allow calculation of Rayleigh scattering for automatic corrections in real time. The light source is a variable
flash lamp with an optical filter at the detector with a bandpass centered at 530 nm with full width at half maximum of approximately 60 nm [Mitchell et al. 2009]. The measurements are recorded in $m^{-1}$.

The system specifications are as follows:

**Measurement**

- **Parameter**: Light scattering extinction coefficient
- **Ranges**: 0 to $> 1 \text{ km}^{-1}$
- **Lower Detection**: $< 0.001 \text{ km}^{-1}$ at 30 sec average
- **Outputs**: 4 Analog (0 to 5 VDC) and RS 232 serial
- **Time constant**: Adjustable: 2 seconds to several minutes

**Measurement Characteristics**

- **Principle**: Integrating nephelometer
- **Electronics**: Computer based, MD68HC11 @ 8 MHz
- **Optics**: No lenses. Reference brightness measurement and chopper stabilized span. Chopper rate adjustable (typical, 20% duty cycle)
- **Wavelength**: 530 nm
- **Pressure**: Microswitch absolute, 1%
- **Temperature**: Thermistor, 0.2%
Physical Characteristics

- Sample Volume 0.44 liter
- Weight 2.6 kg (about 6 lbs)
- LxWxD 56 cm x 12 cm x 17 cm

Power

- Operating Voltage 12 VDC @ 1 amp
- Power Usage 2.5 – 3 watts

Internal Data Storage

- Selected averaging periods 20 sec to 1 hour
- Diagnostic parameters, zero and span settings and time stored in RAM
- Internal Clock
- 21 days of 5 minute averages can be stored before downloading; longer or shorter depending on averaging period.

Operation

The nephelometer (figure 2.2) works by illuminating the particles in a chamber with a diffuse light source (flash lamp) located on one side of the chamber (figure 2.3). The photomultiplier tube detector, located on the bottom of the device, views a dark trap through several baffles with small holes in them. The baffles block any light from reaching the detector except the light scattered directly through the holes in the baffles.
The light that reaches the photomultiplier is very nearly proportional to the scattering coefficient of the air sample in the chamber. The scattering coefficient describes the total amount of light scattered into all angles. Aerosols on the left-hand side of the nephelometer chamber contribute the forward-scattering portion of the integrated scattering coefficient, while aerosols on the right-hand side of the chamber contribute the back-scattering portion of the integrated scattering coefficient. As indicated by the dashed lines in figure 2.3. The range of angles actually included in this measurement excludes the angles very near 0° and 180° (i.e. exact forward and backward scattering, respectively).
M903 Integrating Nephelometer - Rh sensor version

Identifying Parts
1. Power switch
2. Switches to change display modes and operating parameters used in conjunction with panel display.
3. 20 x 4 back lighted panel display
4. Analog outputs - DB9p connector - pins 1-4 outputs (0-5 vdc), pins 6-9 grounds.
5. Serial connector - DB9s connector (direct PC connection).
6. Power connector - 5.5 mm OD, 2.1 mm ID, center positive VDC.
7. Aerosol inlet - Threaded 1/2 inch NPT.
8. Outlet - Vacuum source, threaded 1/2 inch NPT.
9. Flash lamp assembly - Flash lamp and flash lamp circuit board are installed in aluminum cover.
10. Purge port - used for clean air and span gas sources.

Figure 2.2 - Drawing of the M903 integrating nephelometer with parts labeled [Radiance Research M903 Manual].
**M903 Optical Design**

![M903 Optical Design Diagram](image)

Figure 2.3 – Internal view of the M903 nephelometer [Richards 2003].

### Calibration Procedure

Calibration of the nephelometer is done by filling the chamber with two different gases with known scattering coefficients. Particle free air is used as a zero reference with Rayleigh scattering being the known coefficient. The other gas that is used is Carbon Dioxide (CO₂).

The calibration requires some tubing, a filter, CO₂ gas, and a pump. Long tubing is preferred to prevent condensation from the CO₂ tank to enter the filter. The filter that is used in the calibration of this instrument is a Pall Life Sciences HEPA capsule, product number 12144. The CO₂ that is used is a Coleman grade 99.999% pure CO₂.
Lastly, a pump that can supply 10 to 20 liters per minute (lpm) with the filter in line is needed.

The procedure time for the calibration should take no longer than 30 minutes. Any of the operation modes on the M903 (fast, slow, or log) can be used to calibrate. Fast mode is the best option because of its faster flash rate which decreases the stabilization time. To switch the instrument to fast mode, use the display switch to change to the Mode menu. Then use the Item-slow switch to change to mode to Fast. The power switch must then be cycled for the change to take effect.

After the instrument is in the desired mode for calibration, the inlet and outlet must be blocked off. Use some tape to completely block off the outlet (lower). The inlet (upper) needs to be only partially blocked off to allow air flow through the chamber.

On the lower part of the instrument, there is a purge port. Turn the pump on with the filter inline and measure the flow rate blowing from the filter. A valve can be used to control the flow rate through the filter. Adjust the flow rate until about 10 lpm is flowing out of the filter. Now attach the air flow to the port with the filter inline from the pump. The chamber should be filling up with particle free air.

Press the Display switch until the Zero screen is displayed. Press and release the Reset-average switch to reset the bscat value on the screen and wait for the value to stabilize. The zero offset parameter can be adjusted at different speeds depending on how far the zero parameter needs to be adjusted. Holding the Item switch up while
pressing the Parameter switch up or down causes the parameter to be adjusted faster. Holding the Item switch down while pressing the Parameter switch up or down causes the parameter to be adjusted slower. Adjust this parameter until the bscat value is less than ±0.05e-5. Press the reset-average switch to re-stabilize the bscat value to double check the calibration. This concludes the Zero parameter adjustment.

Turn the pump off and move the tube from the pump to the CO$_2$ source. Leave the filter inline to prevent any unwanted particles to enter the chamber. Press the Display switch until the Span screen is displayed. Slowly blow the CO$_2$ into the chamber. Allow a small amount of time for the chamber to be completely filled with the CO$_2$ to prevent a bad calibration. The next steps are very similar to adjusting the zero parameter. Adjust the span value until the bscat value is equal to the gas value that is displayed on the second row of the screen. This is a value that is calculated from the CO$_2$/Air ratio and current air density determined for pressure and temperature. Keep an eye on the Wall scattering value. If the value goes above 100% before bscat is equal to the gas value, the chamber may need to be cleaned or there could be a problem with the alignment of the optics. If the span value was significantly off, the zero parameter may need to be readjusted.

Turn off the flow from the CO$_2$ source. Remove the tape from the inlet and outlet along with the tube attached to the purge port. All of the original tubes and pump can be hooked back up and the nephelometer can resume normal operation.
MetOne Optical Particle Counter

Figure 2.4 - Photo of MetOne optical particle counter (J. Shaw photo).

The MET ONE 237B optical particle counter is a portable device that includes the sensor, pump, and electronics in a compact enclosure [Hach Ultra 2008]. The particle counter operates at a flow rate of 0.1 cfm or 2.83 LPM. The sample air passes through a laser beam in the sensor. The flashes of scattered light are counted and the pulses are directly proportional to the amount of scatter to make a correlation to particle size. The particle counts are reported in size bins. These size bins are > 0.3, 0.5, 0.7, 1, 3, 5 µm. The numbers in the bins correspond to number of particles larger than that diameter. Figure 2.5 is a drawing of the general workings of an optical particle counter to get an idea of how an optical particle counter works.
The optical particle counter works by detecting the amount of light scattered by a particle. When a particle passes through the laser source, light is scattered by that particle and is detected by a photo detector at some angle (90° in the case of the example in figure 2.5). The amplitude of the light that is scattered by that particle can be used to estimate the particle’s size based on calibrated thresholds, and is counted in the appropriate size bin.

**Calibration**

The MET ONE 237B is calibrated to ISO 21501 standards [Hach Ultra 2008]. The particles that are used for calibration are polystyrene particles with a refractive index of...
1.59 @ 589 nm (25° C). The calibration is done by passing a continuous stream of standard, mono-sized particles through the optical particle counters sensor, which results in a stream of electrical pulses, each pulse being proportional to the size of each particle. The mono-sized standard particles produce a distribution of pulse heights, the median of which is typically regarded as the appropriate channel calibration threshold for that size. The calibration of these instruments with particles of a fixed refractive index introduces some uncertainties when the actual aerosols have a different index. However, the calibration particle index of 1.59 is quite typical of the values found for refractive index from inversions of the Montana State University solar radiometer data.

Scanning Mobility Particle Sizer

Figure 2.6 - Photo of the scanning mobility particle sizer setup (J. Shaw photo).
A scanning mobility particle sizer (SMPS) spectrometer is an instrument that is used to determine a size distribution for an aerosol sample. It uses a continuous, fast scanning technique that provides high-resolution measurements at a fast rate. The SMPS works based on the principle of the mobility of a charged particle in an electric field. There are three pieces that make up a SMPS. They are a static classifier, a particle counter, and software.

The electrostatic classifier is a TSI 3071A. The electrostatic classifier inputs polydisperse aerosols, and outputs monodisperse aerosols of known size. The instrument contains a Kr-85 source that produces bipolar ions and applies a Fuchs equilibrium charge distribution to the aerosols. After the particles are neutralized, they enter a differential mobility analyzer (DMA) where the aerosols are classified according to their electrical mobility. The mobility is the ratio of the steady state velocity to the product of electric field strength and charge on the ion. In the case of electrically charged particles in a stream of flowing air, this ratio is proportional to the particle radius (Shaw 2011).

Particles of various sizes are therefore selected by varying the electric field inside the chamber. The charge in the DMA is controlled by a negatively charged rod located in the center of a cylinder. The particles move along the side of the cylinder. Particles with positive charge move towards the central rod at a speed determined by their electric mobility. The voltage applied to this rod is scanned to produce a changing electric field. This causes particles with a certain mobility (i.e., particles with a specific
radius or diameter) to enter a narrow slit and pass to the output of the instrument. The size of the particles being output by the instrument is determined by charge, central rod voltage, and the flow in the DMA.

Figure 2.7 –Drawing of the TSI 2071A DMA [TSI 2002].

The particle counter is a TSI 3010 condensation particle counter. It works by enlarging the particles to a detectible size so they are able to be counted by a laser diode and photo detector.
The alcohol reservoir is filled with butanol and is heated. As the particles flow through, the alcohol evaporates into the sample stream. The sample then passes through a vertical thermoelectrically cooled condenser. The alcohol vapor then supersaturates and condenses onto all particles larger than 10 nm in diameter. As the droplets exit the condenser, they pass through a thin ribbon of laser light. Light scattered by these droplets is collected by optics and is focused onto a photo detector. The photo detector converts the light signal into an electrical pulse, which is recorded as a particle count.

The SMPS software is a TSI product. This software controls the scanning voltage of the electrostatic classifier and records the particle counts at each diameter size. It
then stores the size distributions to a file. The program can look at the size distribution in different ways such as a count or volume distribution. The size distributions are measured from 0.012 µm to 0.5 µm in diameter.

**Solar Radiometer**

A solar radiometer (also often called a sun photometer) is an instrument that measures the extinction of direct solar spectral irradiance (W m\(^{-2}\)) [Shaw et al. 1973, Shaw 1983]. This allows determination of the amount of sunlight absorbed and scattered by gases and particles. Wavelength bands are defined by optical interference filters. The data product that is most directly inferred from the relative irradiance measurements is optical depth. Optical depth is a unitless and dimensionless quantity that describes the amount of total extinction in an integrated medium, which in this case is the earth’s atmosphere. Aerosol optical depth describes the total amount of light scattered and absorbed by aerosols. Aerosol optical depth values can depend highly on wavelength. Solar radiometers can also produce a number of very useful data products derived from inversions [King et al. 1978, Shaw 1979, Nakajima et al. 1983 &1996, Wang and Gordon 1993, Dubovik and King 2000]. Most modern retrievals are done using almucantar and principal plane radiance scans at the solar zenith and azimuth angles of the sun, respectively. Retrieved data products include size distribution, refractive index, single scattering albedo (SSA), phase functions, and more.
The solar radiometer used in this project is a CIMEL Electronique 318A. It is a solar powered, robotically pointed sun and sky solar radiometer. The instrument uses 9 spectral bands: 340, 380, 440, 500, 670, 870, 940, 1020, and 1240 nm. This instrument is also part of the AERONET program (site 400). Established by NASA, AERONET is a large collection of these ground-based solar radiometers with locations across the world [Holben et al. 1998].

**Calibration**

Calibration of this instrument is done at the NASA Goddard Space Flight Center by AERONET personnel, side by side with another calibrated CIMEL. The calibration coefficients for each channel are determined by matching the AOD to those from the reference instrument that was calibrated previously at the Mauna Loa Observatory. Radiance calibrations are also done by using an integrating sphere to ensure accurate inversion products.
The method used to calibrate the AERONET reference radiometers, which we also have used to validate the AERONET calibration of the MSU solar radiometer, uses Langley plots to determine the calibration coefficients for each wavelength [Shaw et al. 1973]. This method is based on the following equation for measured solar irradiance $I_m$ as a function of optical depth $\tau$ and zenith angle $\theta_z$ for an exo-atmospheric irradiance $I_0$:

$$I_m = I_0 e^{-\tau \sec(\theta_z)}. \quad (2.1)$$

Taking the natural logarithm of this equation gives

$$\ln(I_m) = \ln(I_0) - \tau \sec(\theta_z). \quad (2.2)$$

Rewriting this in terms of a measured voltage that is proportional to irradiance gives

$$\ln(V_m) = \ln(V_0) - \tau \sec(\theta_z). \quad (2.3)$$

This is the equation of a straight line describing $\ln(V_m)$ as a function of $\sec(\theta_z)$ with intercept $\ln(V_0)$ and slope $\tau$. In accordance with this, a Langley plot is a plot of the natural logarithm of the measured signal vs. the airmass, $\sec(\theta_z)$. The calibration coefficient $\ln(V_0)$ describes the natural log of the solar irradiance the radiometer would see looking at the sun outside the earth’s atmosphere (no atmospheric effect on the light). A Langley plot is determined by taking measurements while the sun is rising through decreasing air mass, plotting these values on a natural log scale, and fitting a line through those measurements. Air mass describes the optical path length through the atmosphere relative to the zenith path length (a zenith path through the entire atmosphere is an air mass of 1). Where that line crosses the zero of the x axis (zero air mass) is the $\ln(V_0)$ or calibration coefficient of that channel. Once $V_0$ is known, optical
depth can be calculated by using equation 2.4.

\[
\tau = \frac{\ln(V_0) - \ln(V_m)}{\sec(\theta_2)}
\]  

(2.4)

Examples of Langley plots at two different wavelengths for the MSU solar radiometer are shown in figure 2.10.

Figure 2.10 – Langley plots for the MSU solar radiometer at 440 nm (top) and 500 nm (bottom). The \( V_0 \) value and \( r^2 \) values are shown in the title of each plot.
ANNUAL AEROSOL CHARACTERISTICS IN BOZEMAN, MT

Introduction

Aerosols vary vastly at different location across the world. This has to do with a number of causes, such as differences in climate (e.g. desert to tropical, etc.), population concentration, pollution, geographic location (e.g. inland or coastal), etc. Each of these different kinds of aerosols affects solar radiance in different ways and it is important to understand everything about this interaction in all regions of the world. Some regions have very high aerosol concentrations, while some are very low, even in populated areas. Bozeman, MT tends to have very clean and pristine conditions year round with episodic wildfire smoke in late summer and a small amount of pollution build up from temperature inversions in the winter. This chapter gives an overview of the aerosols in Bozeman, MT by looking at scattering and AOD throughout the year, as well as how wind affects the aerosols. It also looks at some of the scattering and AOD values from other locations to give an idea of how Bozeman compares to other well known areas.

Annual Scattering Coefficients

Overall, Bozeman, MT (45.67°N, 111.05°W) and the surrounding Gallatin valley is a very clean area devoid of any significant man-made aerosol sources and therefore devoid of high aerosol concentrations, especially those that reduce visibility from high
light scattering. Bozeman is immediately surrounded by mountains to the south, east, and north, and the Gallatin Valley extends nominally downhill to additional mountains approximately 75 km to the west (the entire valley is tilted with its highest elevations to the southeast and lowest elevations to the northwest). Figure 3.1 is a Google maps image of Bozeman to give an idea of the layout of the valley.

Figure 3.1 – Layout of Bozeman in the Gallatin Valley. ©2010 Google – Imagery ©2010 TerraMetrics, Map data ©2010 Google.
Figure 3.2 – Scattering coefficient for the year 2010, smoothed with a daily mean filter.

The daily mean scattering coefficients measured by a nephelometer for the entire year of 2010 in Bozeman, MT are shown in figure 3.2. The nephelometer is measuring the scattering coefficient at 530 nm wavelength. All scattering coefficients referenced for the Bozeman area in this chapter refer to that at 530 nm and include only scattering from aerosols (Rayleigh scattering is subtracted out). A daily mean filter is applied to the data to smooth the plot and allow for yearly trends to be seen. Figure 3.3 is the same plot but plotted to show maxima, minima, and standard deviations of the scattering coefficient for each day. In a year, there are usually three different periods of aerosol events. The first, most pristine and clean, periods are in late spring to early summer (April to late June) and fall (from September into November). The next period
is mid-to-late summer (July and August), where episodic forest fire smoke events occur (in dry years, this can continue well into September). Lastly, during the winter (late November through February), local pollution tends to be trapped by temperature inversions, leading to a higher concentration of locally generated aerosols.

The very clean periods represent Bozeman’s normal background aerosol makeup. These periods occur when the temperatures are sufficiently warm to encourage mixing and when there are very few sources of pollution or forest fire smoke. Figures 3.2 and 3.3 show that during these time periods the scattering coefficient rarely goes above 15 Mm\(^{-1}\) and there is very little daily fluctuation. The aerosol size distribution during these periods tends to have a very small accumulation mode, which would contribute significantly to optical scattering if present. The small accumulation mode is outweighed by Aitken mode particles, which cause very little optical scattering. These very clean conditions lead to very high visibility. The 2010 yearly average scattering coefficient measured in Bozeman was approximately 10 Mm\(^{-1}\).

The higher scattering coefficients that occur in the late summer mainly come from forest fire smoke. Bozeman is surrounded by mountains and national forests that have a very high risk for natural fires. Bozeman also receives periods of aged forest fire smoke transported from other natural fires from areas such as Washington, Oregon, California, and even southern Canada. Figures 3.2 and 3.4 show that during the later summer months of 2010, the scattering coefficient was often above 15 Mm\(^{-1}\), and in some cases much higher (it should be noted that summer 2010 was somewhat wetter
than normal, with a corresponding smaller number of smoky days). The very high spikes in the scattering coefficient are usually caused by local or regional fire events where the smoke is still fresh and the aerosol concentration is still very high.
Figure 3.3 – Candle stick plot of the scattering coefficient for 2010 in Bozeman, MT. The thin line represents the max and min for the day, where the thick line represents the standard deviation for that day.
Figure 3.4 shows an aerosol size distribution from two days in late September 2009 where a fresh smoke event occurred near Bozeman. This figure shows that the smoke starts being measured around 12:00 Mountain Daylight Time (MDT = UTC-6 hours) on September 25\textsuperscript{th}. The smoke particle diameters tend to center around 0.15 to 0.18 \(\mu\)m. Particles in this size range have a very large effect on optical scattering. Figure 3.5 shows the scattering coefficient measured by the nephelometer for these two days. The scattering coefficient rises around 12:00 on the 25\textsuperscript{th} while the smoke is coming in. At about 13:00 MDT on September 26\textsuperscript{th} winds from the west pick up and blow the smoke out from around Bozeman as can be seen in figures 3.4 and 3.5.

![Figure 3.4 – SMPS plot \(\frac{dN}{d\log(D_p)}\) for September 25\textsuperscript{th} and 26\textsuperscript{th} 2009 during a smoke event that comes in around 12:00 on the 25\textsuperscript{th} and is blown out briefly around 14:00-17:00 on the 26\textsuperscript{th}. Color intensity is in the units of \(1/cm^3\).]
Aerosols in Bozeman are very different in winter than they are in summer. Aerosol concentrations can change significantly from day to day and are usually higher than they are during the early summer. This most likely comes from temperature inversions during the very cold weather trapping aerosols and allowing local pollution to build up. Local pollution comes from wood burning stoves and cars, as well as a small amount of industrial pollution from businesses located west of Bozeman. These scattering coefficients are not significantly high compared to many populated areas, but they are highly variable compared to the summer months.

An example of a day when a temperature inversion trapping aerosols was in effect is shown in figure 3.6. A layer of trapped aerosols can be seen in the valley below
hanging over Bozeman. This occurred on December 24\textsuperscript{th}, 2009 at 11:22 am Mountain Standard Time (UTC-7). The temperature at the airport located about 13 northwest of where the picture was taken was \(-15^\circ\text{C} (4.5^\circ\text{F})\) at an elevation of 1,363 meters. The temperature on the Cobleigh Hall roof at Montana State University was around \(-9^\circ\text{C} (15.8^\circ\text{F})\) at an elevation of 1,495 meters.

Figure 3.6 – Photograph of a pollution layer trapped in the Gallatin Valley by severe temperature inversion on December 24\textsuperscript{th}, 2009 at 11:22 am Mountain Standard Time (UTC-7). This photo is looking northwest from the top of Painted Hills Road southeast of Bozeman (J. Shaw photo).

The average scattering value measured by the nephelometer for that day was 25.2 Mm\textsuperscript{-1}. To put this in perspective, this value is around half of what the yearly average on the west coast is. These inversion layers do not happen every day in the winter months, but when they do, the aerosols tend to stay around a few days while they mix back out of the boundary layer.
Figure 3.7 shows optical scattering coefficients based on three-year averages (mainly from National Parks) across the country. This is based on work done by the Interagency Monitoring of Protected Visual Environments (IMPROVE) network of visibility monitors.

Aerosol Extinction

These scattering coefficients are measured by a transmissometer, which measures light extinction at more than the single wavelength measured by the integrating nephelometer that was used in Bozeman. Figure 3.7 shows that the eastern United States has much higher scattering values than the rest of the country, and as a result, there is much lower visibility and noticeable aerosol concentrations. High humidity in
the east can contribute to this by causing aerosols to swell to larger sizes. The west coast has lower scattering coefficients than the east coast, but they are still much higher than those in the western continental United States. Figure 3.7 shows that Bozeman is located near the cleanest part of the country. The data collected at Bozeman and reported here indicate that it may be even cleaner than the IMPROVE data would suggest (perhaps because Bozeman is located between IMPROVE measurement sites, or possibly because of differences in transmissometer and nephelometer measurements).

**Aerosol Optical Depth in Bozeman**

Aerosol optical depth (AOD) is another very useful measurement of how aerosols affect light. AOD is different than a scattering coefficient, in that a scattering coefficient is an *in situ* measurement at a single point, while AOD measures the extinction caused by aerosols of solar irradiance through a column of the atmosphere. This is useful because AOD includes the effect of all aerosols in the atmosphere above any location (i.e., it can include aerosols that are not present at the ground).

Bozeman’s yearly average AOD at 500 nm wavelength is approximately 0.062 for the year 2010. Table 3.1 shows yearly averages of some different locations across the world to put this in broader context (these data are taken from the AERONET website).
Table 3.1 – AOD annual averages for several locations across the world.

<table>
<thead>
<tr>
<th>Location</th>
<th>Annual average AOD at 500nm during 2010</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bozeman, MT</td>
<td>0.062</td>
</tr>
<tr>
<td>Mauna Loa, HI</td>
<td>0.037</td>
</tr>
<tr>
<td>Cal Tech (Pasadena, CA)</td>
<td>0.143</td>
</tr>
<tr>
<td>Goddard Space Flight Center (Greenbelt, MD)</td>
<td>0.271</td>
</tr>
<tr>
<td>Gandhi College, India</td>
<td>0.697</td>
</tr>
<tr>
<td>Beijing, China</td>
<td>0.7</td>
</tr>
</tbody>
</table>

Mauna Loa, HI is considered one of the cleanest places on earth because of both its isolated location far from any sources of industrial pollution and its 3394-m high elevation, which is often above the boundary layer. In fact, in early summer, the Bozeman AOD is at times very close to that at Mauna Loa. AOD at many locations on the west coast, such as Pasadena, CA, have averages that are over twice what is seen in Bozeman and highly populated areas in the northeast United States have even higher AODs (perhaps because of the higher population concentration and aerosols there swell to larger sizes in the more humid air). For example, the average AOD at Goddard Space Flight Center in Greenbelt, MD is over four times higher than in Bozeman. Areas with high population and industry densities in Asia are showing average 500 nm AOD around
0.7. When interpreting these AOD values, it is important to remember that the aerosol optical transmittance is given by \( \exp(-\text{AOD}) \), so a factor of 2.3 higher AOD in Pasadena implies an aerosol-induced atmospheric transmittance that is only 10% of the mean value in Bozeman. Or, to put it in even starker contrast, the annual mean AOD value of 0.7 in Beijing implies an aerosol-induced atmospheric transmittance that is only 0.001% of the mean value in Bozeman. In other words, the visibility degrades exponentially with higher optical depth.

**Wind and Aerosols**

Wind patterns have a very large effect on the aerosols observed at any given site. The winds can control where the aerosols come from and also clean out aerosols that have been building up in a location. The unique geographical layout of Bozeman and the Gallatin Valley leads to up-slope and down-slope winds that have a very distinctive impact on the observed aerosols. Figure 3.8 is a compass rose plot of annual wind direction for 2010. The plot shows how often winds come from a specific direction. The longer the bars are for each direction, the more the wind came from that direction.
Figure 3.8 – Compass rose for annual wind direction occurrence for 2010 (data measured by the ZERT weather station, just west of the Montana State University campus in Bozeman, MT: orsl.eps.montana.edu/weather/zert).

Figure 3.8 shows that the wind most often blows from the southwest, with many occurrences of the wind blowing anywhere from the west to the south. The wind also comes in often from the southeast. Most storm systems and weather come into the valley from the southwest. The wind rarely blows in from the north.

Many of the winds over an annual cycle come from the southeast direction as can be seen in figure 3.8. Some of this wind pattern can be explained by something that is seen during warm weather conditions. At times, when the temperature is warm and there is solar heating in the valley, the day time winds can be calm and come out of the
westerly direction causing air to move up the valley. Near or at sunset, with the reduction of solar heating, the air cools rapidly, causing winds blow out of the east to southeast at reasonably high speeds. This diurnal cycle does not happen always, but it can be common during the summer months. Depending on what the conditions are like in the valley, the aerosol makeup may or may not change with the winds out of the east. There have been many instances where the aerosol makeup does not change, but there have also been times where significant aerosol size distribution change occurs with these winds. Figure 3.9 shows an example of this diurnal wind shift while figure 3.10 shows an SMPS plot displaying a drastic change in the aerosol size distribution when the winds out of the east pick up.

Figure 3.9 – Wind direction and speed plot showing high winds blowing out of the east as the sun starts to go down at about 18:00, bringing in cool air.
Figure 3.10 – SMPS intensity plot \( \frac{dN}{d\log(D_p)} \) showing the aerosol makeup drastically changing as the winds from the east come in.

To understand how aerosol concentration relates to the winds, an experiment is done using data from late August 2010 to early October 2010. This is analyzed by looking at the total aerosol concentration between 0.01 µm to 0.52 µm in diameter (SMPS measurements). This total aerosol concentration is plotted against the wind speed in Figure 3.11. The data points are color coded to represent wind direction.
Figure 3.11 shows that the concentration of aerosols tends to be related to the wind direction. One significant thing to note about the relationship between the winds and aerosols is, whenever the wind is blowing out of the south-southeast (from the nearby tall mountains with no nearby towns), the concentration of aerosols is usually low, regardless of wind speed (which is highly variable). Also, the winds from the east tend to have similar wind speeds, and the aerosol concentrations tend to stay between $0.2 \times 10^{-8} \text{ m}^{-3}$ and $0.4 \times 10^{-8} \text{ m}^{-3}$. The last thing to note is that when the winds do blow out of the north, across the main area of the city of Bozeman, the wind speed tends to be very low, but the aerosol concentration varies significantly. Very high concentrations are most likely caused by stagnation when there is little or no wind.
Conclusion

This chapter discussed annual aerosol characteristics of Bozeman and gave some examples of aerosol conditions in other areas to provide a context for the Bozeman conditions. Bozeman generally provides a very clean environment, and is arguably one of the cleanest parts of the continental United States. Bozeman has small amounts of pollution from wood burning and cars in the winter and some episodic smoke events in the late summer. This makes Bozeman a very interesting laboratory to do aerosol studies, especially the very important studies under conditions of low aerosol concentration.
EXAMINING AERONET’S REAL REFRACTIVE INDEX IN LOW AEROSOL OPTICAL DEPTH CONDITIONS

Introduction

Understanding the scattering and absorption properties of aerosols and how accurately they can be measured is the focus of this study. Inversions of Aerosol Robotic network (AERONET) solar spectral irradiance measurements produce column-averaged aerosol size distributions, scattering phase functions, and complex refractive indices. The complex refractive index is a very important piece of information for understanding aerosols, what they consist of, and how they affect the scattering and absorption of light. The real part of the refractive index that is retrieved from the AERONET inversion using a solar radiometer’s full sky scans has undergone very few accuracy assessments under clean conditions. The accuracy of the refractive index during very low aerosol optical depths and pristine conditions is very important for research at Montana State University, as the Gallatin valley is rarely polluted when there is no forest fire smoke present. This study compares measured scattering extinction coefficients with Mie-theory modeled scattering extinction coefficients derived from both in situ measured sized distribution and the AERONET’s retrieved complex refractive index. This comparison is used to assess the accuracy of the real part of the refractive index. The goals of this experiment are to look at how accurately the refractive index can be retrieved and if AERONET’s refractive index represents the refractive index of aerosols at the ground in clean, pristine conditions.
Method

During this study, measurements of aerosols were taken by many instruments that all contributed to the results. The *in situ* instruments included a scanning mobility particle sizer (SMPS), a single-wavelength integrating nephelometer that measured the scattering extinction coefficient at 530 nm, and a Met One optical particle counter. A solar radiometer that is part of the AERONET network makes direct solar irradiance measurements as well as full-sky radiance measurements that are inverted to create useful data products that describe aerosol properties, the most interesting one for this study being the complex refractive index.

The SMPS and the Met One particle counter measure the number of aerosols at specific particle sizes. The SMPS measures aerosol size distributions in the size range of approximately 0.0053 \( \mu \text{m} \) to 0.26 \( \mu \text{m} \) in radius. This accounts for particles in the Aitken and accumulation modes of the aerosol size distribution, but does not account for particles in the coarse mode size range. The Met One particle counter measures total number of particles in six size bins. These size bins are 0.15 to 0.25 \( \mu \text{m} \), 0.25 to 0.35 \( \mu \text{m} \), 0.35 to 0.5 \( \mu \text{m} \), 0.5 to 1 \( \mu \text{m} \), 1 to 2.5 \( \mu \text{m} \), and 2.5 \( \mu \text{m} \) and greater. These particle sizes are mostly part of the coarse mode in the aerosols size distribution.

Figure 4.1 is a flow chart of the procedure for this study. It shows all data products used and what processes they went through, including how to make a useful
coarse mode size distribution with the Met One particle counter measurements. The procedure shown in this flow chart will be explained in more detail below.

Figure 4.1 - Flow chart of experiment procedure.
The Mie model used to solve for the scattering extinction coefficient from aerosols uses two properties of the aerosols: the aerosol size distribution and the aerosol’s complex refractive index. The size distribution is measured by the SMPS and the refractive index data product comes from the AERONET inversion. Some initial models used only the SMPS data with no particles in the coarse mode included. The SMPS creates a very high-resolution size distribution for the smaller particles, but because the size bins of the Met One particle counter are so wide, there is no direct way to create a useful coarse mode size distribution from these measurements by using the counts alone. After some early results, it became clear the coarse mode particles generated a significant enough part of the scattering to justify adding them to the model, and were needed to obtain accurate results. Even though the coarse mode particles contribute little to the overall scattering coefficient, they still contribute enough to make the model incomplete without them. The particles in the size range that are measured by the SMPS create most of the scattering, but the larger particles (measured by the MET One particle counter) do have an effect on the scattering. The Met One particle counter has wide size bins and is difficult to produce a useful size distribution with just the measured particle counts. To account for these coarse mode particles, some simple polygonal fits were done to the size bins to create a coarse mode size distribution. These fits resulted in a large overestimation of very large particles (greater than 1 µm), which caused the scattering model to produce non-believable results. This resulted in a more complex approach that would create a believable size
distribution across the coarse mode size ranges while still accurately accounting for the total number of particles counted. Using the conventional assumption that the aerosols follow a log normal size distribution [Seinfeld 2006], log normal curves were fit to the SMPS data and a similar curve was fit to the measured particle counts from the Met One particle counter to account for the scattering from the larger particles.

The approach used to create a fit to the size distribution is a least-squares non-linear solver. This minimizes the difference between the measured aerosols and a modeled size distribution fit in count space ($\frac{dN}{dnr}$). The modeled size distribution was made up of five lognormal distributions all summed together. Each lognormal distribution has three variable inputs: the total concentration, the median radius, and the standard deviation. These three inputs to the nonlinear solver were changed until the differences between the fit and the measured data were minimized.

The five lognormal distributions were chosen to represent different parts of the aerosol size distribution: one for the nucleation mode, two for the Aitken mode, one for the accumulation mode, and one for the coarse mode. The nucleation mode was modeled because certain distributions measured by the SMPS during the study period displayed the tail from a mode centered below 10 nm in radius, which is the lower limit of the SMPS size range. This will be seen in some of the figures. The Aitken mode is modeled with two log normal distributions because there were several situations where 2 modes could be seen growing through the Aitken size ranges with their peaks at different sizes. This is also done because the Aitken mode changes rapidly and tends to
not be lognormal at times. A single lognormal distribution is sufficient to accurately model the accumulation mode. A modeled coarse mode distribution is the main goal of this fitting process because of the very wide particle count bins that the Met One particle counter measures. The fit is a good way to account for all of the particles counted while shaping the particles into a believable log normal distribution. Even though the fits to the SMPS data were not used as an input to the Mie code, it is important to include all the particles in the fit, because the tail of the accumulation mode overlaps with the coarse mode size range. This helps to mesh the size distributions measured from two instruments at their size limits and create a more accurate fit to the particles.

Some examples of the size distribution fits are shown in figure 4.2. Distributions are plotted in three ways: the number, volume, and cross section size distributions. The cross section distribution helps to show where most of the scattering comes from. An interesting thing to note about the size distributions from this sample period is that the Aitken mode particles almost always outweigh the accumulation mode (median radius around 0.07 μm). Still, these smaller particles contribute little to the scattering, but this shows how clean the Bozeman site is. In a more polluted site, the accumulation mode would typically highly outweigh the Aitken mode particles. An example of a tail from a smaller mode, in the nucleation size ranges, can be seen in the number distributions measured on August 24th and September 29th. There are also two modes growing through the Aitken size ranges on the plot from August 24th.
Figure 4.2 - SMPS size distribution (blue) plotted with the size distribution fit (red) from the solver. The coarse mode comes from the lognormal distribution fit to the Met One particle counter data.
The size distribution fit through the coarse mode allows a more accurate size distribution to be input into a Mie model. Mie theory describes the scattering and absorption of light by a spherical particle. (For the purposes of this study, the particles are assumed to be spheres.) There are three main parameters that affect the scattering and absorption of light by a particle. These parameters are the wavelength of the incident light, the size of the spherical particle, and the complex refractive index of the particle relative to the surrounding medium [Seinfeld 2006]. The complex refractive index consists of a real and imaginary part. The real part is related to the scattering component while the imaginary part relates to the absorption of a particle. The refractive index is a very important property for understanding aerosols and their effect on scattering and absorption. It also can suggest chemical properties and makeup of the aerosols.

The Mie code used in this experiment was developed by Christina Mătzler [Mătzler 2002] to calculate the scattering properties of a single particle. Code written by Nathan Pust [Pust 2011] implements this Mie model to calculate the scattering properties of a size distribution.

The inputs into the Mie model are the wavelength of interest (530 nm), the SMPS measured size distribution with the coarse mode fit attached to it, and the refractive index from the AERONET inversions during the same period. The refractive index from AERONET at the wavelength of 530 nm is obtained by interpolating between the measurements of the two closest wavelengths (500 and 670 nm). The refractive
index from AERONET is interpolated to the same time resolution as the size distribution input. The 530 nm wavelength of the model was chosen to match the wavelength of the nephelometer. The scattering coefficient is modeled for each of the size distributions taken by the SMPS, which has a 5 minute resolution.

The Mie model is also used to calculate truncation errors from the nephelometer. These errors come from limitations of the nephelometer to measure near-forward and near-backward scattered light [Moosmüller and Arnott 2003]. For the nephelometer used in this study, the truncation angle is estimated to be about 10° (measures scattering from 10° to 170°) [Heintzenberg et al. 2005]. This truncation error can have a significant effect on large particles (diameters greater than 1 µm) with errors in the range of 20% - 50% because of the large forward scattering component from large particles. The truncation errors in this study with volume mean diameters smaller than 1 µm, the errors are much smaller (around 5% - 10%).

The modeled scattering coefficient is then compared to the truncation error corrected scattering coefficient measured by the nephelometer, which is taking the same sample of aerosols as the SMPS. For the AERONET refractive index to be used with the ground-based instrument data, some important assumptions need to be made. The most important of them is well mixed conditions; that is, a uniform distribution of aerosols throughout the boundary layer. This is important because the solar radiometer is looking through the atmosphere at the sun. The data products from the inversion are
the integration through that column, whereas the *in situ* instruments measure conditions at the ground and are unaware of any elevated layers of aerosols.

**Results and Discussion**

This study was done for 9 full clear sky days between August 24, 2010 and September 30, 2010. The aerosol optical depths of the clear days during the study period are between 0.028 and 0.075 at 500 nm. The model for each day starts with the first AERONET inversion and ends with the last AERONET inversion from that day. This prevents interpolation between the last data point of the previous day and the first point of the current day. Some examples of the scattering comparisons versus time for two of the study days are shown in figure 4.3. The plots show the measured scattering coefficient from the nephelometer along with the modeled scattering coefficient using the SMPS data only (accumulation mode alone) and SMPS data with the coarse mode fit attached to it (accumulation and coarse modes). The addition of the coarse mode to the model produces a small increase in scattering, but does not have a significant effect on the August 26th and most of the September 19th results (figure 4.3). However, for the afternoon of September 19th there is a spike in the scattering coefficient for a couple hours. When only the SMPS data are used in the model, that spike cannot be seen, but when the coarse mode is added, the scattering spike is accounted for. This shows why it is important to know something about the larger particles and how they are affecting the scattering.
Figure 4.3 - Examples of time series plots of the modeled scattering coefficient compared to the measured.
The scattering models with the fit coarse mode show good agreement with measured values from the nephelometer. The modeled scattering coefficients are mostly within 20% of each other and track each other very reliably, as seen in figure 4.3. There are many reasons that can cause the 20% difference in the measured and modeled scattering coefficients that will be discussed below.

Figure 4.4 shows the results of the measured versus modeled scattering coefficient from the unaltered AERONET refractive index. Each dot represents a data point, with the solid line being the ideal fit and the dashed line representing the robust fit of the data. The figure shows that the modeled scattering is consistently high compared to the measured data product.

![Figure 4.4 - Scatter plot of measured scattering versus modeled scattering using SMPS and coarse mode fit and unaltered refractive index. The solid line is the ideal fit, and the dashed line is the robust fit of the data.](image)

To bring the modeled and measured values closer together, some changes were made to the real part of the refractive index retrieved from AERONET. The real index was decreased by 4% and 3.5% to see the resultant changes to the modeled scattering coefficient versus the measured values.

Figure 4.5 shows the results from the decrease of the real part of the refractive index. Again, each dot represents a data point with the solid line being the ideal fit and the dashed line representing the robust fit of the data. The scatter plots show that decreasing the real part of the refractive index decreases the modeled scattering coefficient and moves the robust fit toward from the ideal fit.

![Figure 4.5 - Scatter plots of measured scattering versus modeled scattering with changes made to the real part of the refractive index. The solid line is the ideal fit, and the dashed line is the robust fit of the data. top) Decrease in real refractive index by 4%. bottom) Decrease in real refractive index by 3.5%.]
The results from lowering the real part of the refractive index from the AERONET inversion bring the modeled scattering coefficient in line with the measured data from the nephelometer. For the refractive indices through this study, a 5% decrease in the real part of the refractive index causes about a 30% decrease in the scattering coefficient. This shows how sensitive the scattering is to small changes in the refractive index. With the modeled scattering coefficient falling in line with the measured value in relatively high and low scattering situations, the AERONET inversion may be overestimating the real refractive index by about 3.5% (0.06 to 0.07) during this study. Because the AERONET refractive index varied little over the experiment (from 1.5 to 1.6), it is difficult to tell whether there is a steady 3.5% overestimation over all refractive indices, or if the overestimation is a constant of 0.06 to 0.07 for these conditions. With a longer study period, the overestimation amount could be better understood. The small amount of outliers in the comparison suggests that the aerosols are likely evenly distributed throughout the boundary layer. It seems unlikely that the difference between the measured and modeled scattering is consistently affected by an elevated layer during the more-than-one-month-long study. It also suggests that as the scattering coefficient and AOD changes (especially to very low values), the accuracy of the real part of the refractive index is not changing drastically with lower concentration of aerosols (lower AOD).

There are many potential causes for the errors in the retrieval of the real part of the refractive index. One reason is an unmixed boundary layer. Though unlikely, there
still could be elevated layers that the *in situ* instruments are not detecting. Non-spherical particles can also have a large effect on the refractive index, but for this study, the sphericity measurements from AERONET are very high (almost always 99%). Previous work has shown that non-spherical particles can cause errors in the size distribution as well as an unrealistically strong decrease of the real part of the refractive index with decreasing wavelength [Dubovik et al. 2002]. Other work with polarized solar radiometers has shown that, without the higher inversion sensitivity provided by polarization, radiance-only inversions may exhibit real refractive index estimation errors [Z. Li et al. 2009].

Errors in AERONET retrievals can also be caused by errors in surface albedo [Sinyuk et al. 2007]. The underestimation of surface albedo can cause a large underestimation (around 75% to 85%) in the imaginary part of the refractive index, which is compensated for by the inversion in the real part (0.07 or about 5% increase). There is also a known issue of errors in the retrievals as the aerosol optical depth (AOD) decreases to low values [Dubovik et al. 2000]. This is a valid concern for the Bozeman area since the AOD at 500 nm rarely rises above 0.1 unless there are smoky conditions from fires. The results show that the refractive index can be accurately retrieved in these clean conditions but may have small overestimations of the real refractive index.

Stratospheric aerosols also start to be a factor at these very low AODs. During this study, the average 500-nm AOD measurements at the Mauna Loa Observatory were about 0.009, while the lowest 500-nm average AOD day used in this study from
Bozeman was 0.028, with the highest being 0.075. This shows that during some of the lowest AOD points, the stratospheric aerosols make up as much as 30% and as little as 12% of that optical depth. If these aerosols have a much different refractive index than those in the boundary layer, this could affect the overall refractive index through the column. Figure 4.6 shows a plot of real refractive index versus time during this study for Bozeman and Mauna Loa. Because the solar radiometer at Mauna Loa is looking largely at stratospheric aerosols during clear conditions, this will show how different or similar the refractive indices of these particles are compared to looking through the boundary layer and stratospheric aerosols. This could indicate whether stratospheric aerosols tend to bias the refractive index either high or low.

Figure 4.6 - Refractive index (real part) of Bozeman and Mauna Loa for the study time period. This is used to get an idea of how different the stratospheric aerosol's refractive index is.
The figure shows that there is no systematic bias that shows up between the locations. There tends not to be a trend of the real part of the refractive index from Mauna Loa being consistently high or low compared to the Bozeman refractive index. The lower accuracy in AERONET inversions during very low AOD conditions is also a problem with the Mauna Loa data (AOD of 0.007 at 500 nm). It is clear from the figure that 1.6 is the upper constraint of the AERONET retrieval on the real part of the refractive index. Figure 4.6 shows that during the study, there were many inversions that resulted in the refractive index being at that upper constraint for the Mauna Loa data, as well as some for the Bozeman data. This is more evidence that the inversion has a difficult time converging when there is a very small concentration of aerosols, especially at Mauna Loa where there are almost none. This low sensitivity can cause errors in the Bozeman data as well. The possibility of these errors in the inversion leads to investigation into other sources of information on the stratospheric aerosol refractive index.

The stratosphere contains a natural layer of aerosols at heights of 12-30 km. These aerosols consist mainly of small sulfuric acid droplets on the order of 0.2 µm diameter [Seinfeld 2006]. These supercooled aerosol particles are approximately 75% H₂SO₄ and 25% H₂O [Yue et al. 1994]. Studies done by Yue et al. show that the real part of the refractive index through the stratosphere at a wavelength of about 530 nm varies between 1.435 and 1.458. Other studies have shown that the real part of the refractive index in the stratosphere varies from 1.4 to 1.42 over a latitude range from 70° S to 50°
N and from 1.34 to 1.46 over a vertical range from 4 – 20 km [Baumgardner et al. 1996].

As can be seen from figure 4.6, these refractive index values of aerosols in the stratosphere are lower than almost all of the AERONET retrieved refractive indices from the study period in Bozeman. This suggests that if the stratospheric aerosols are skewing the AERONET refractive index values, they would most likely cause a decrease in the real part of the refractive index when compared to the boundary layer aerosol refractive index. This is the opposite of the effect shown during this study. Model results have shown that the AERONET refractive index may be too high, whereas the stratospheric aerosols—if they are having a significant effect—likely would be lowering the AERONET refractive index compared to the boundary layer value. Therefore, it is unlikely that the stratospheric aerosols are responsible for the higher refractive indices.

**Conclusions**

Using measurements from *in situ* instruments and a solar radiometer, the accuracy of the real part of the refractive index from the AERONET inversion during very clean conditions was assessed with a Mie scattering model. The motivation for this study was to find out how well the refractive index could be retrieved, as well as to determine whether the real part of the refractive index from AERONET can represent the refractive index at the ground during clean, well-mixed conditions. The analysis showed that the real part of the refractive index is retrieved with reasonably high accuracy, but with a small constant overestimation of about 5% throughout this study.
This is reassuring because the inversion products from AERONET are extremely useful for additional aerosol studies in the Bozeman valley. The AERONET retrievals can represent the refractive index of aerosols at the ground during this study if we assume that the real part is being overestimated by 3.5%. A longer study period would be needed to more accurately assess how well the refractive index from AERONET can generally represent the aerosols at the ground.

Although the results show that the AERONET-retrieved real part of the refractive index could be overestimated during this study period, several sources for errors need to be considered besides AERONET. These issues include:

- Boundary layer aerosols not being well mixed.
- Stratospheric aerosols causing a bias in the refractive index during very low AOD conditions.
- Surface albedo errors in AERONET inversions.
- Aerosols containing non-spherical particles (although AERONET products show they are spherical).
- Low sensitivity of solar radiometer at low aerosol optical depths.
- Low sensitivity of nephelometer at low scattering coefficients (< 1 Mm\(^{-1}\)).
- Errors in \textit{in situ} instrument calibrations (nephelometer, SMPS, and particle counter).
Introduction

The focus of this study is to do a direct comparison of the aerosol size distribution measured by two different instruments. These measurements are from an inversion product from a solar radiometer that is part of the Aerosol Robotic Network (AERONET) and a scanning mobility particle sizer (SMPS). These instruments are located close to each other on the Montana State University campus. The solar radiometer measures solar spectral radiance, from which a column-averaged size distribution is retrieved, while the SMPS measures the size distribution at ground level. The aerosol size distribution is a very important part of aerosol studies. It describes the sizes of the aerosols and concentration of aerosols as a function of size. The size strongly influences the optical scattering caused by the aerosols, and also can help identify the substance aerosols are made of. This study directly compares the accumulation mode from these two measurements by looking at the concentration, the shape and width of the mode, and the peak radius for the mode. The goal of this experiment is to consider how accurately the aerosol size distribution can be measured and how well AERONET’s retrieved size distribution represents the size distribution at the ground during well mixed, clean conditions.
For this study, several different instruments measuring aerosols in the Gallatin valley were used. The \textit{in situ} instruments include a scanning mobility particle sizer (SMPS), and a single-wavelength integrating nephelometer that measures the scattering extinction coefficient at 530 nm. The primary remote sensor used in the study is a solar radiometer operating as part of the AERONET network. This instrument directly measures solar spectral irradiance, as well as skylight spectral radiance along the principal plane and along almucantar scans (through 360° of azimuth at the solar elevation angle). The sky radiance measurements are inverted to create useful data products that describe aerosol properties, such as the aerosol size distribution, the complex refractive index, and the aerosol scattering phase function. The product that is the focus of this study is the aerosol size distribution.

The SMPS measures the number of aerosols at specific particle sizes to create a size distribution. The aerosol size distributions from the SMPS are in the size range of approximately 0.0053 µm to 0.26 µm in radius. This accounts for particles in the Aitken and accumulation modes of the aerosol size distribution, but does not account for particles in the coarse mode size range. The AERONET size distribution models the size distribution using the sky radiance scans from the solar radiometer. The size distributions from the AERONET inversions range from about 0.08 µm to 15 µm in radius. These size ranges account for the accumulation mode as well as the coarse mode of the aerosol size distribution. Since the SMPS does not measure particles in the
coarse mode, while the AERONET-inverted size distribution is not sensitive to particles in the Aitken size ranges, the accumulation mode is the only part of the aerosol size distribution that the two instruments share in common. Therefore, the aerosols in the accumulation mode are the primary interest for this study. The solar radiometer also infers aerosol optical depth (AOD) using the direct sun irradiance measurements that are used in this study.

Figure 5.1 is a flow chart outlining the procedure of this study. This chart shows all data products used in the study, as well as the data processing steps and what is being compared. The procedure shown in the flow chart is explained in more detail below.
Figure 5.1 - Flow chart of experiment procedure.
The primary goal for this study is to compare the accumulation mode of the AERONET-retrieved size distribution with the accumulation mode of the SMPS-measured size distribution. To do this, three parts of these log normal modes are compared: the total volume concentration, the mode width, and the peak radius of the mode. These comparisons are done with direct and indirect comparisons. All of these comparisons are done on the volume size distribution \( \frac{dv}{dn} \). For the comparisons to be accurate, the aerosols must be well mixed throughout the boundary layer. This is important because the solar radiometer is looking through the column of aerosols in the atmosphere, while the SMPS and nephelometer are measuring aerosols at the ground level.

Two of the parameters compared are the peak radius and the width of the mode. The peak radius describes the radius at which the peak of the log normal mode is located. This parameter can be compared directly. As shown in more detail in the results section, the AERONET size distribution radii have very low resolution when compared to the SMPS. This is not too big of a concern, but it can lead to difficulty in knowing exactly where the peak radius is when viewing AERONET data. The second comparison is the accumulation mode width. This is done by looking at how wide the log normal distributions of the accumulation modes are at half max.

The third comparison is the concentration of the two size distributions in volume space. This comparison is indirect and involves calculating the effective boundary layer height different ways using multiple instruments. Note that the effective boundary
layer height is different than the planetary boundary layer because there are aerosols above the planetary boundary layer. These aerosols are seen by the solar radiometer, but the concentration of these aerosols is usually much smaller than those in the lower boundary layer. The effective boundary layer height is defined as the altitude at which all of the aerosols in the atmosphere could be compressed down while being evenly distributed based on the measured aerosols properties. This means that the effective boundary layer height that is being calculated using aerosol measurements is expected to be higher than the actual planetary boundary layer during well-mixed conditions.

The first method to calculate the effective boundary layer height is to divide the peak volume concentration of the AERONET size distribution by the peak volume concentration of the SMPS size distribution. This gives an effective height because the AERONET size distribution is a column size distribution. This means the size distribution measured includes all particles from ground level up to the top of the atmosphere. The AERONET size distribution is in units of volume per unit area, where the SMPS measures volume per unit volume. This causes the ratio to have units of distance, which is the effective boundary layer height.

The second method that the effective boundary layer height is calculated is to divide the scattering aerosol optical depth (AOD) at the nephelometer wavelength by the scattering coefficient that is measured by the nephelometer. The AOD is measured by the solar radiometer using a direct sun irradiance measurement. This AOD can be split into two parts: the scattering AOD and the absorption AOD. The absorption AOD is
retrieved by the inversion of the full sky scans (same inversion that retrieves the size
distribution). To get the scattering AOD, the absorption AOD is subtracted from the
direct sun measured AOD. Since the nephelometer measures the scattering of light by
aerosols and not the absorption, the absorption product of the AOD needs to be
removed. The AOD is a unitless data product that describes the amount of light that
aerosols prevent from passing through a column of the atmosphere. This means that
the more light that is scattered and absorbed, the higher the AOD. The nephelometer
measures the scattering coefficient in inverse meters. Again, when these data products
are divided, the result is a distance, which is the effective boundary layer height.

These two effective boundary layer heights are then compared. The effective
boundary layer heights should be in the 0.5 to 5 km range to be believable. The data
products from the solar radiometer are derived using a column measurement through
the atmosphere (AOD and size distribution), but the data products are retrieved in very
different ways using direct sun and sky radiance measurements respectively. The direct
sun AOD is very accurate and is calculated using a very simple formula and a carefully
calculated calibration. The effective height obtained using the scattering coefficient and
the AOD should be accurate in well-mixed conditions. The accuracy of the volume
concentration of the AERONET size distribution can be inferred because, if the two size
distribution’s ratio is very close to the scattering and AOD ratio (effective heights), then
the AERONET volume concentration can be said to be very accurate.
Results and Discussion

This study was done using data taken from the instruments described above between August 24, 2010 and October 4, 2010. The aerosol optical depths from this time period average about 0.05 at 500 nm. The first comparison considered is the peak radius of the accumulation mode of the SMPS-measured size distribution and the AERONET-retrieved volume size distributions. Figure 5.2 shows the peak radius for each size distribution for both instruments during the study time period. The time resolution is that of the AERONET inversion. Each AERONET accumulation mode peak radius is plotted with its corresponding SMPS accumulation mode peak radius. There are spaces in the data because the AERONET inversion requires a clear-sky measurement for accurate retrievals.
Figure 5.2 – Peak radius values for the accumulation mode of the two size distributions (AERONET and SMPS).

The AERONET size distribution is much more limited in size resolution, as can be seen in figure 5.2. Throughout the study, the SMPS accumulation mode peak radius is consistently bound by the AERONET accumulation mode peak radius. The only period where this is not true is around September 11th and 12th. These are the two cleanest days during this study (AOD average of 0.028 at 500 nm). The smaller peak radius of the accumulation mode for the SMPS data on those two days is most likely caused by a few reasons. One is that the AERONET size distribution has a limited resolution, especially at smaller radii. The other is that instrument sensitivity limits are being reached with very small concentrations of aerosols, and the inversions are becoming less accurate [Dubovik et al. 2000]. Also, the lower tail of the accumulation mode for the AERONET
inversion size distribution has the slope constrained [Dubovik and King 2000]. This limits how small the peak radius of the accumulation mode can be from the inversion. Overall, the peak radii of the accumulation mode from the AERONET inversions are very accurate during this study period.

The accumulation mode width is examined by looking at the width at half max for the SMPS size distribution vs. the AERONET size distribution. Figure 5.3 shows some size distribution plots with both the SMPS and AERONET inversion data. These plots show only the accumulation mode and are normalized to get an idea of how well they compare to each other.
Figure 5.3 – Size distribution plots for some specific times during the study. (continued on next page)
The first two plots in figure 5.3 are from September 11th and 12th, 2010. Figure 5.2 shows that during these two days, the peak radius of the SMPS accumulation mode was lower than that of the AERONET accumulation mode. Figure 5.3 shows examples of
the size distributions from these days. The two modes look to have about the same width, but the x axis is in log scale, so the mode that is smaller in size will have a much smaller width. Again there are a few reasons that these differences in the size distributions occur. First, the solar radiometer is a light sensing instrument. Since the smallest particles measured here have much less effect on light scattering, the instrument is not sufficiently sensitive to these particles. These two days were very clean and had a very limited amount of light scattering particles in the atmosphere. This causes these smaller particles, which have much less effect on light scattering, to outweigh the larger particles in the accumulation mode. The SMPS can measure the smaller particles very well and is probably a more accurate measurement during these times. The second reason that these smaller particles do not have an effect on the AERONET measurement is that the slope of the size distribution at the small particle sizes is constrained in the inversion [Dubovik and King 2000]. With these constraints, the inversion will not display higher volumes at the very small particle sizes. Lastly, the aerosols being unevenly distributed throughout the boundary layer or elevated aerosol layers can contribute to the differences in the size distributions.

The last two plots in figure 5.3 show good agreement in the accumulation modes between the two size distributions. With higher volumes of particles that have a large effect on light scattering, the two size distributions tend to agree much better. During this study, there were a lot of very clean days, which makes the width comparisons very difficult. When the peak radii are not close to each other, the widths cannot be
compared accurately. To get around this, the widths of the two accumulation modes are compared only for measurements taken when the peak radius of the two modes are within 0.015 µm of each other. Figure 5.4 shows a scatter plot, using the peak radius constraint, of the two accumulation mode widths. The black solid line represents the one to one and the blue dots represent the widths of the accumulation mode made by the two instruments at similar times.

![Image](image.png)

**Figure 5.4** – Accumulation mode with at half max for the SMPS data vs. the AERONET size distribution. These data points are for the peak radii being within 0.015 um of each other.

Figure 5.4 shows that the widths cluster around the one-to-one line and show no significant bias towards either of the measurements. This shows that during conditions
where the accumulation mode contains a larger volume of light scattering particles, the widths tend to agree well.

The concentration of the volume size distribution is examined by calculating the effective boundary layer height using two approaches. The first approach is the ratio of the AERONET accumulation mode concentration peak and the SMPS accumulation mode concentration peak, and the second is the ratio of the AOD from scattering and the scattering coefficient measured at the ground. This part of the study uses 9 clear-sky days to ensure the maximum amount of AERONET inversions throughout the day. Time series plots of the effective boundary layer height on some of the clear-sky days are shown in figure 5.5.

![Effective boundary layer height for 08-25-2010](image.png)

Figure 5.5 – Time series plots of effective boundary layer height calculations. (continued on next page)
Figure 5.5 continued – Time series plots of effective boundary layer height calculations.
These time series plots show very good agreement between the two methods of calculating the effective boundary layer heights. The heights are very believable for the boundary layer, and they track each other very well throughout the day including capturing increasing boundary layer heights in the afternoon, most likely caused by convective lifting. Figure 5.6 is a scatter plot comparing the two boundary layer heights. The scatter plot includes all the data points from the 9 clear-sky days.

Figure 5.6 – Scatter plot of the two effective height calculations. ($r^2 = 0.59$)

The scatter plots show very good agreement between the two effective boundary layer heights throughout the study. There is no large bias showing one height being always higher or lower than the other. This suggests that the volume concentration for the AERONET size distributions is very accurate during this study.
Conclusions

Using measurements from *in situ* instruments and a solar radiometer, the accuracy of aerosol size distribution from the AERONET inversion during very clean conditions was assessed. The primary goal was to determine how well the SMPS size distribution compared to the AERONET retrieved version and if that size distribution could represent the size distribution at the ground level. The volume concentration and the peak radius for the most part were very accurate throughout the study despite the limited size resolution of the AERONET inversion. The effective boundary layer height comparisons were also very reassuring while assessing the size distribution accuracy. The widths of the accumulation mode compared reasonably well when the peak radii were very close to each other and showed no bias. Since these modes are log normal, they cannot be compared when the peak radii are not close, though. During this study period with well mixed, clear sky conditions; the AERONET size distribution generally represents the size distribution at the ground.
The annual aerosol characteristics of Bozeman were discussed and compared with other areas to provide a context for the Bozeman conditions. Bozeman generally provides a very clean environment, and is arguably one of the cleanest parts of the continental United States. Bozeman has small amounts of pollution from wood burning and cars in the winter and some episodic smoke events in the late summer. This makes Bozeman a very interesting laboratory to do aerosol studies, especially the very important studies under conditions of low aerosol concentration. Measurements of aerosols in Bozeman were taken from August 2009 and are still continuing.

Studies were then done on some specific time periods to look at the accuracy of different aerosol data products. Measurements taken between August 24, 2010 and October 4, 2010 were the main focus of these studies.

Using measurements from *in situ* instruments and a solar radiometer, the accuracy of the real part of the refractive index from the AERONET inversion during very clean conditions was assessed with a Mie scattering model. The motivation for this study was to find out how well the refractive index could be retrieved, as well as to determine whether the real part of the refractive index from AERONET can represent the refractive index at the ground during clean, well-mixed conditions. The analysis showed that the real part of the refractive index is retrieved with reasonably high accuracy, but with a small constant overestimation of about 5% throughout this study. This is reassuring because the inversion products from AERONET are extremely useful
for additional aerosol studies in the Bozeman valley. The AERONET retrievals can represent the refractive index of aerosols at the ground during this study if we assume that the real part is being overestimated by 3.5%. A longer study period would be needed to more accurately assess how well the refractive index from AERONET can generally represent the aerosols at the ground.

Although the results show that the AERONET-retrieved real part of the refractive index could be overestimated during this study period, several sources for errors need to be considered besides AERONET. These issues include:

- Boundary layer aerosols not being well mixed.
- Stratospheric aerosols causing a bias in the refractive index during very low AOD conditions.
- Surface albedo errors in AERONET inversions.
- Aerosols containing non-spherical particles (although AERONET products show they are spherical).
- Low sensitivity of solar radiometer at low aerosol optical depths.
- Low sensitivity of nephelometer at low scattering coefficients (< 1 Mm$^{-1}$).
- Errors in \textit{in situ} instrument calibrations (nephelometer, SMPS, and particle counter).

Using measurements from \textit{in situ} instruments and a solar radiometer, the accuracy of aerosol size distribution from the AERONET inversion during very clean conditions was assessed. The primary goal was to determine how well the SMPS size
distribution compared to the AERONET retrieved version and if that size distribution
could represent the size distribution at the ground level. The volume concentration and
the peak radius for the most part were very accurate throughout the study, despite the
limited size resolution of the AERONET inversion. The effective boundary layer height
comparisons were also very reassuring while assessing the size distribution accuracy.
The widths of the accumulation mode compared reasonably well when the peak radii
were very close to each other and showed no bias. Since these modes are log normal,
they cannot be compared when the peak radii are not close, though. During this study
period with well mixed, clear sky conditions, the AERONET size distribution generally
represents the size distribution at the ground.

There is still a large opportunity for future work in these studies. The
groundwork for \textit{in situ} and remote sensing instrument measurement comparisons have
been laid out, but some improvements can be made. Something that would open up
more routes in this type of study would be an improvement in the availability of
instruments, as well as some upgraded instruments. These include a three-wavelength
nephelometer to look more closely at scattering across the visible spectrum, and an
absorption-measuring instrument to allow ground level complex refractive index
measurements to be made. Another idea to look at in the future is splitting up the fine
and coarse mode complex refractive indices to get more accurate scattering results
since the fine and coarse mode aerosols are made up of potentially different materials.
This work could also be done under higher AOD conditions to determine if similar results would be obtained.
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