Abstract

Im, Jung-Sun. Investigation of aerosol optical properties on regional climate forcing and Spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes. (Under the direction of Dr. V. K. Saxena)

Aerosols influence Earth’s heat budget both directly by scattering and absorbing sunlight, and indirectly by acting as nuclei for cloud droplets. To reduce the uncertainties of current estimates of aerosol climate forcing, aerosol optical properties relevant to the computation of direct radiative forcing were measured at a regionally representative site near Mount Mitchell, North Carolina. On the basis of these measurements and model calculations, we have studied (1) the effects of relative humidity (RH) on aerosol optical properties and direct aerosol radiative forcing and (2) the influence of long-range transport on black carbon (BC) concentrations, its seasonal and weekly patterns, and the effects of BC on the regional climate of the southeastern US. The light scattering of aerosol is strongly dependent on RH at which it is measured, due to hygroscopic growth nature of most atmospheric aerosols. In this study, the hygroscopic growth factor (ratio of total scattering coefficient at RH=80% to that at RH=30%) was calculated to be almost constant value of 1.60 +/- 0.01 for polluted, marine, and continental air masses. In addition, it was found that as the RH increased from 30% to 80%, the backscatter fraction decreased by 23%. The patterns of direct radiative climate forcing by aerosols for various values of RH were similar for the three air masses, but the magnitudes of the forcing were larger for polluted air masses than for marine and continental air masses by a factor of nearly 2 due to higher sulfate concentration in polluted air masses. The
averaged forcing for all the observed ambient RHs was -2.9 W m\(^{-2}\) (the negative forcing of -3.2 by aerosol scattering plus the positive forcing of +0.3 by aerosol absorption) for polluted air masses, -1.4 W m\(^{-2}\) (-1.5 plus +0.1) for marine air masses, and -1.5 W m\(^{-2}\) (-1.6 plus +0.1) for continental air masses. The BC mass concentration of the southeastern US showed the highest average concentration in polluted air masses and the lowest in marine air masses. During the winter, the overall average BC value was 74.1 ng m\(^{-3}\), whereas the overall summer mean BC value was higher by a factor of 3. The main reason for the seasonal difference may be enhanced thermal convection during summer, which increases transport of air pollutants from the planetary boundary layer of the surrounding urban area to this rural site. In the spring of 1998, abnormally high BC concentrations from the continental sector were measured. These concentrations were originating from a biomass burning smoke plume in Mexico. This was confirmed by the observations of the Earth Probe Total Ozone Mapping Spectrometer. The net aerosol radiative forcing (scattering effects plus absorption effects) per unit vertical depth at 2006 m MSL was calculated to be -1.4x10\(^{-3}\) W m\(^{-3}\) for the southeastern US. The magnitude of direct radiative forcing by aerosol scattering was reduced by 15 +/- 7 % due to the BC absorption.

SAGE II ozone and aerosol measurements and NCEP/NCAR potential vorticity and temperature fields during 1985-1999 were analyzed to study the spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes and to investigate the impact of polar stratospheric clouds (PSCs) on aerosol distribution and ozone depletion. During austral spring (Sep/Oct) there exist strong radial gradients
in ozone, aerosol, and temperature near the polar vortex edge region and an isolation of materials inside polar vortex, confirming that springtime polar vortex acts effectively in keeping out intrusions of materials from the exterior of the polar vortex. The analysis of vertical profiles of 1020-nm aerosol extinction and 525- to 1020-nm aerosol extinction ratio inside spring polar vortex indicates aerosol enhancement in 9-15 km layer and lack of larger particles in 15-20 km layer, which is consistent with the effects of gravitational sedimentation and subsequent evaporation of PSCs. For the spherical PSC particles for which the Reynolds number < 1, gravitational sedimentation velocity is estimated to be 0.11, 0.04, and 1.5 km/wk for Type Ia, Ib, II PSCs, respectively. The probabilities of the occurrence of T<195K (threshold temperature of Type Ia PSC formation), T<192K (Type Ib), and T<190K (Type II) at 50 mb pressure height are calculated in the region of 60-75 S during austral winter (Jul/Aug). The probabilities of occurrence for the three categories of temperature show increasing trends with time, whereas springtime stratospheric column ozone amount shows decreasing trend. During volcanically unperturbed years, the stratospheric column ozone in the spring is highly correlated with the probability of occurrence of temperature below 195 K in the previous winter (R=-0.88). The empirical relationship between stratospheric column ozone amount (O3) in the spring (Sep/Oct) and the probability (P) of occurrence of T<195K in the previous winter (Jul/Aug) is found to fit the equation of O3 = -360.38 x P + 446.38, which can be used to predict ozone depletion from the previous winter temperature.
Investigation of aerosol optical properties on regional climate forcing and spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes

by

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Biography

Jung-Sun Im was born in Seoul, Korea on February 17, 1966. After graduation from Ewha Womans High School, she applied and was accepted for an admission to Ewha Womans University. She received her B.S. degree in Science Education (Earth Science). After graduation, she was admitted to Seoul National University and received a M.S. degree in Atmospheric Sciences in 1991. In the fall of 1998, she entered the graduate school at North Carolina State University. Thereafter, she was inducted into the Phi Kappa Phi, the National Honor Society, in 2000. After completion of her another M.S. degree in August 2000, she went on to pursue a Ph.D. degree in Atmospheric Sciences at North Carolina State University.
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The deepest appreciation goes to her parents who have provided her with love and support throughout her life. Lastly, she would like to share her happiness with her husband and her son, Sang-Kyun Han.
# Table of Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>List of Tables</td>
<td>viii</td>
</tr>
<tr>
<td>List of Figures</td>
<td>ix</td>
</tr>
<tr>
<td>1. Introduction</td>
<td>1</td>
</tr>
<tr>
<td>1.1. Statement of problem</td>
<td>1</td>
</tr>
<tr>
<td>1.2. Objectives of this study</td>
<td>9</td>
</tr>
<tr>
<td>1.3. References</td>
<td>11</td>
</tr>
<tr>
<td>2. An assessment of hygroscopic growth factor for aerosols in the surface boundary layer for computing direct radiative forcing</td>
<td>20</td>
</tr>
<tr>
<td>2.1. Abstract</td>
<td>21</td>
</tr>
<tr>
<td>2.2. Introduction</td>
<td>23</td>
</tr>
<tr>
<td>2.3. Methodology</td>
<td>25</td>
</tr>
<tr>
<td>2.3.1. Experimental sites</td>
<td>25</td>
</tr>
<tr>
<td>2.3.2. Instrumentation</td>
<td>27</td>
</tr>
<tr>
<td>2.4. Results and discussions</td>
<td>30</td>
</tr>
<tr>
<td>2.4.1. Total Scattering</td>
<td>30</td>
</tr>
<tr>
<td>2.4.2. Single scatter albedo</td>
<td>31</td>
</tr>
<tr>
<td>2.4.3. Humidity and light scattering</td>
<td>34</td>
</tr>
<tr>
<td>2.4.4. Effects of relative humidity on direct radiative forcing</td>
<td>37</td>
</tr>
<tr>
<td>2.4.5. Direct radiative forcing at mean ambient relative humidity</td>
<td>41</td>
</tr>
<tr>
<td>2.5. Summary and conclusion</td>
<td>42</td>
</tr>
<tr>
<td>2.6. References</td>
<td>46</td>
</tr>
</tbody>
</table>
3. Temporal trends of black carbon concentrations and regional climate forcing in the southeastern United States ...............................................................65
  3.1. Abstract .......................................................................................................66
  3.2. Introduction .................................................................................................68
  3.3. Methodology ...............................................................................................69
    3.3.1. Experimental setup .............................................................................69
    3.3.2. Instrumentation ....................................................................................71
  3.4. Results and discussions ...............................................................................73
    3.4.1. Seasonal patterns of BC concentrations .............................................73
    3.4.2. Weekly patterns of BC concentration .................................................74
    3.4.3. Comparison with scattering measurements ........................................76
    3.4.4. BC effects on direct radiative climate forcing ....................................77
  3.5. Summary and conclusion ............................................................................81
  3.6. References ..................................................................................................83

4. Spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes: Impact of polar stratospheric clouds .........................................97
  4.1. Abstract .......................................................................................................98
  4.2. Introduction ................................................................................................100
  4.3. Data sets .....................................................................................................103
    4.3.1. SAGE II data ......................................................................................103
    4.3.2. NCEP/NCAR reanalysis data ..............................................................104
  4.4. Results and discussions ...............................................................................106
    4.4.1. Spatial and temporal distributions of aerosol and ozone ....................106
    4.4.2. Gravitational sedimentation of PSCs .................................................110
    4.4.3. Probability of temperature for PSC formation ....................................113
    4.4.4. Empirical relationships between ozone, temperature, and PV .........115
  4.5. Summary ....................................................................................................117
  4.6. References ..................................................................................................121
# List of Tables

<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Table 2.1</td>
<td>Sampling period and number of samples for instruments deployed at the valley and mountain sites during the summer of 1998.</td>
</tr>
<tr>
<td>Table 2.2</td>
<td>Aerosol radiative parameters and the direct radiative forcing calculated for various values of RH in the lowest 1km layer of the troposphere for three different air masses influencing the research sites during summer 1998.</td>
</tr>
<tr>
<td>Table 2.3</td>
<td>Estimated effects of RH on radiative parameters and the direct radiative forcing (which were defined as the ratio of the value at a given RH to the value at RH=30%) averaged for all three air masses in the lowest 1 km layer of the atmosphere.</td>
</tr>
<tr>
<td>Table 3.1</td>
<td>Comparison of temporal (day-of-week) distribution of black carbon concentrations at Mount Gibbes, NC, to those obtained in southwestern PA. The numbers represent mean ± standard deviation.</td>
</tr>
<tr>
<td>Table 4.1</td>
<td>Statistics for the linear regression of ozone on probability of temperature for PSC formation.</td>
</tr>
<tr>
<td>Table 4.2</td>
<td>Statistics for the linear regression of ozone on PV.</td>
</tr>
</tbody>
</table>
List of Figures

Figure 1.1. Geographical distribution of present-day annual-average radiative forcing (W m⁻²) due to (a) well-mixed greenhouse gases, (b) stratospheric ozone depletion, (c) increases in tropospheric ozone, (d) the direct effect of sulphate aerosol, (e) the direct effect of organic carbon and black carbon from biomass burning, and (f) the direct effect of organic carbon and black carbon from fossil fuel burning [IPCC, 2001]...........................................18

Figure 1.2. The global mean radiative forcing of the climate system for the year 2000, relative to 1750 [IPCC, 2001]........................................................................19

Figure 2.1. Map of the United States illustrating the geographical location and classification of the different air mass types influencing the research sites. ....................................................................................................................56

Figure 2.2. Average value of total scattering coefficient at 530nm measured by the nephelometer for three different air masses arriving at the valley and mountain sites during summer 1998..........................................................57

Figure 2.3. Diurnal variation of RH and total scattering coefficients at 530nm measured by the nephelometer for three air masses arriving at the valley site..............................................................................................................58

Figure 2.4. Average values of total scattering coefficient, absorption coefficient, and single scatter albedo for three air masses arriving at the mountain site during summer 1998. .........................................................................................59

Figure 2.5. Variation of averaged total scattering coefficient with averaged RH and fitting for three air masses arriving at the lowest 1 km layer of the troposphere during summer 1998. .............................................................60

Figure 2.6. Variation of averaged backscatter fraction with averaged RH measured from 3-wavelength nephelometer at the valley site during the period of July 14 – 24, 1998...........................................................................................................61

Figure 2.7. Variation of aerosol direct radiative forcing with RH for three air masses arriving at the lowest 1 km layer of the troposphere during summer 1998. ...........................................................................................................62
Figure 2.8. Influence of RH on the aerosol direct radiative forcing for three air masses arriving at the lowest 1 km layer of the troposphere during summer 1998. ..............................................................63

Figure 2.9. Average direct radiative forcing by scattering and absorption of aerosols in the lowest 1 km layer of the troposphere for the three air masses influencing the research site during summer 1998. .................................64

Figure 3.1. Map of the United States illustrating the geographical location and classification of the different air mass types influencing the research sites. .................................................................................................89

Figure 3.2. Average monthly black carbon concentrations for each sector at Mount Gibbes, NC..............................................................90

Figure 3.3. Average black carbon concentrations for each sector during the summer months (June, July, and August in 1998) and during the winter months (November and December in 1997 and December in 1998). ..............91

Figure 3.4. Earth Probe TOMS aerosol index maps for May 15 (top), May 16 (middle), and May 17 (bottom) 1998.........................................................92

Figure 3.5. Forty-eight-hour back-trajectories for polluted continental air mass cases arriving at the research site at 1300 EST during June and July 1998 ......93

Figure 3.6. Temporal (day-of-week) distribution of black carbon concentrations for the trajectories displayed in Figure 3.5. Each point represents the average concentration for 1000 EST to 1600 EST during the days indicated. ......94

Figure 3.7. Temporal (day-of-week) distribution of black carbon concentrations averaged for polluted continental air masses during the summer period (June, July, and August, 1998). The line with circles denotes the mean value and dashed lines denote 95% confidence interval (CI) for the mean estimate. ....................................................................................................95

Figure 3.8. Daily means of absorption coefficient and scattering coefficient measured from July 15 to August 13, 1998. P, C, and M denote polluted continental, continental, and marine air mass influences for specific times, respectively, based on 48 hr back trajectory analyses that were available during the period ........................................................................................................96
Figure 4.1. Potential vorticity (PV) distribution averaged for winter (July/August) and spring (September/October), on the 500 K isentropic surface in the southern hemisphere during 1985–1999. 1 PV unit = $-10^3$ m$^2$ s$^{-1}$ kg$^{-1}$ K. Latitude circles are denoted in 10 degree increments from 90 °S outward to 30 °S.

Figure 4.2. Seasonal variations of the vertical distributions of 9-year mean (1985–1990 and 1997–1999) stratospheric ozone number concentration in the region of 60 °S – 75 °S.

Figure 4.3. Seasonal variations of the vertical distributions of 10-year mean (1986–1990 and 1995–1999) aerosol extinction coefficient (at 1020 nm) in the region of 60 °S – 75 °S.

Figure 4.4. Vertical profiles of 525- to 1020-nm aerosol extinction ratio inside and outside the springtime polar vortex, averaged for 10-year period (1986–1990 and 1995–1999). The error bars denote standard error.

Figure 4.5. Schematic illustration of aerosol redistribution process through the gravitational sedimentation and subsequent evaporation of polar stratospheric clouds inside the polar vortex.

Figure 4.6. The gravitational sedimentation velocity as a function of the particle diameter for Type Ia PSCs, Type Ib PSCs, and Type II PSCs.

Figure 4.7. Probability of occurrence of T < 195 K, T < 192 K, and T < 190 K during winter (Jul/Aug) at the 50 mb pressure height in the region of 60 °S – 75 °S.

Figure 4.8. The stratospheric column ozone amount in the spring (Sep/Oct) and the probability of occurrence of temperature below 195 K at the 50 mb pressure height during the previous winter (Jul/Aug), in the region of 60 °S – 75 °S.

Figure 4.9. The spring (Sep/Oct) stratospheric column ozone amount plotted against the probability of occurrence of temperature below 195 K at the 50 mb pressure height during the previous winter (Jul/Aug), in the region of 60 °S – 75 °S. The individual years are indicated.

Figure 4.10. The spring (Sep/Oct) stratospheric column ozone amount plotted against the spring (Sep/Oct) potential vorticity on the 500 K isentropic surface, in the region of 60 °S – 75 °S. The individual years are indicated.
Plate 4.1.  SAGE II multi-year horizontal distributions of 1020-nm aerosol extinction at 20-km altitude in Jan/Feb. Latitude circles are denoted in 10 degree increments from 90 °S outward to 30 °S. .................................................................140

Plate 4.2.  SAGE II multi-year horizontal distributions of 1020-nm aerosol extinction at 20-km altitude in Sep/Oct (color dots) and NCEP/NCAR mean potential vorticity (PV) distributions on the 500 K isentropic surface for Sep/Oct. Contour interval is 3 PV units (1 PV unit = −10^{-5} \text{ m}^2 \text{ s}^{-1} \text{ kg}^{-1} \text{ K}). Latitude circles are denoted in 10 degree increments from 90 °S outward to 30 °S. .............................................................................................................141

Plate 4.3.  SAGE II multi-year horizontal distributions of stratospheric column ozone amount in Sep/Oct (color dots) and NCEP/NCAR mean potential vorticity (PV) distributions on the 500 K isentropic surface for Sep/Oct. Contour interval is 3 PV units (1 PV unit = −10^{-5} \text{ m}^2 \text{ s}^{-1} \text{ kg}^{-1} \text{ K}). Latitude circles are denoted in 10 degree increments from 90 °S outward to 30 °S........142
1. Introduction

1.1. Statement of the Problem

Since the beginning of the Industrial Revolution, mid-18th century, the impact of human activities on their environment has begun to extend to large scale, even to global scale. Human activities, in particular those involving the combustion of fossil fuels for industrial or domestic usage, and biomass burning, produce greenhouse gases and aerosols which alter the composition of the atmosphere. The emission of chlorofluorocarbons (CFCs) and other chlorine and bromine compounds has also led to the depletion of the stratospheric ozone layer. Such effects have a potential impact on regional and global climate.

It is widely recognized that anthropogenic aerosol particles influence the earth’s radiative balance directly by backscattering and absorption of solar radiation [e.g., Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Penner et al., 1994; Haywood and Shine, 1995; Chýlek and Wong, 1995]. Backscattering of solar radiation by aerosol particles, such as sulfates, can cause climate cooling, while absorption of solar radiation by aerosol particles, such as black carbon, can induce climate warming, depending upon the albedo of the underlying terrain and the height of the aerosol layer above the ground. In contrast to the long lifetime of greenhouse gases (decades to a century), atmospheric aerosol particles have a relatively short lifetime (couple of days to a week) in the troposphere. The short lifetime of aerosol particles, together with the highly non-uniform
distribution of aerosol sources, can result in very different patterns of radiative forcing and different climate response patterns. Numerical model calculations of radiative forcing of climate by sulfate aerosol indicate that a large magnitude of negative forcing occurs in local regions of the eastern United States, eastern China, and south central Europe [Charlson et al., 1991; Kiehl and Briegleb, 1993]. The examples of the geographical distribution of present-day annual-average radiative forcing (1750 to 2000) [IPCC, 2001] are shown in Figure 1.1. These are the estimates of the present day radiative forcing due to (a) well-mixed greenhouse gases including CO$_2$, CH$_4$, N$_2$O, CFC-11 and CFC-12 [Shine and Forster, 1999], (b) stratospheric ozone depletion over the period 1979 to 1994 given by WMO, 1995 [Shine and Forster, 1999], (c) increases in tropospheric O$_3$ [Berntsen et al., 1997; Shine and Forster, 1999], (d) the direct effect of sulphate aerosol [Haywood et al., 1997], (e) the direct effect of organic carbon and black carbon from biomass burning [Penner et al., 1998; Grant et al., 1999], (f) the direct effect of organic carbon and black carbon from fossil fuel burning [Penner et al., 1998; Grant et al., 1999]. The effects of spatial inhomogeneity in the distribution of the radiative forcing may lead to locally different responses in surface temperature indicating that the spatial distributions of the radiative forcing need to be accurately represented to improve regional estimates of surface temperature response and other physical parameters. A regional cooling trend in the mean maximum surface temperature over the southeastern United States from 1949 to 1994 [Saxena and Yu, 1998] is potentially an observational confirmation for these predictions. However, the lack of sufficiently detailed observations of aerosol optical properties leads to considerably large
uncertainties in calculating radiative forcing on climate [Charlson et al., 1992; Penner et al., 1994; Schwartz and Andreae, 1996; IPCC, 2001]. Figure 1.2 shows global mean radiative forcing of the climate system for the year 2000, relative to 1750 [IPCC, 2001]. As seen in Figure 1.2, the level of scientific understanding is very low in the radiative forcing for aerosols. There is much less confidence in the ability to quantify the total aerosol effect than that for the gases. To reduce the uncertainties of current estimates of aerosol climate forcing, more integrated studies of aerosols are needed and then it should be used in political decision regarding controls on emission of greenhouse gases and anthropogenic aerosols.

The discovery of the Antarctic ozone hole in the middle 1980s [Farman et al., 1985] stimulated great interests in the photochemistry and dynamics of the southern winter and spring stratosphere. It is now generally accepted that heterogeneous chemical reactions on the surface of polar stratospheric clouds (PSCs) are mainly responsible for springtime Antarctic ozone destruction [e.g., Solomon et al., 1986; Solomon, 1990], whereas polar vortex dynamics creates and maintains the unique environment suitable for the necessary chemical reactions to take place [e.g., Schoeberl and Hartmann, 1991]. In order for widespread ozone destruction to occur during Antarctic spring, the air temperature within the polar vortex must be lower than 195 K for a sufficiently long period so that the PSCs can affect the conversion of chlorine from the inactive reservoir species to the radical species that attack ozone [Schoeberl and Hartmann, 1991]. In addition to the production of active chlorine species by heterogeneous reactions on the PSC particles, another
importance of the PSCs is the sedimentation processes that irreversibly remove HNO$_3$ (denitrification) and water (dehydration) from the lower stratosphere. Denitrification slows the return of chlorine to its inactive forms and hence, enhances ozone destruction in spring season [Toon et al., 1986; Salawitch et al., 1993; Portmann et al., 1996]. However, many questions remain about PSCs because of difficulty of sampling. The microphysics questions involve determining the stable forms of PSCs under stratospheric conditions, the solubility of various gases on PSC surfaces, and how PSCs undergo phase changes over time. The chemistry questions relate to reaction rates and sensibility to variables such as temperature, surface area, and PSC composition. Because of difficulty of sampling PSCs, all of these areas are currently of tremendous research interest. In the stratosphere, aerosols take up nitric acid and water at cold temperature to form PSCs [Carslaw et al., 1997]. Because of the role the aerosols play in PSC formation and thus, indirectly, in heterogeneous chemical processes which promote the destruction of ozone [Poole and McCormick, 1988; Solomon, 1990], polar stratospheric aerosols are of interest. Aerosol physical properties measured by means of remote sensing may be good tracers of dynamical, chemical and physical processes in the lower part of the middle atmosphere.

1.1.1. Aerosol Hygroscopic Growth Factors

The aerosol light scattering and absorption properties depend on the aerosol particle number size distribution, chemical composition (refractive index) and shape of the particles, the degree of mixing of the particles (external and internal mixture), and the
wavelength of the incident light [Bohren and Huffman, 1983; Martins et al., 1998]. In addition, the light scattering of aerosol is strongly dependent on the relative humidity (RH) at which it is measured, because of the hygroscopic growth nature of most atmospheric aerosols [Tang and Munkelwitz, 1977; Tang, 1980; Svenningsson et al., 1994]. The RH dependence of aerosol light scattering (the so-called hygroscopic growth factor) has been studied experimentally [Charlson et al., 1984; Malm et al., 1994; Hegg et al., 1996; Carrico et al., 1998] and theoretically [Hänel, 1976; Hegg et al., 1993], and has been used in the computation of direct radiative climate forcing by aerosol [Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Boucher and Anderson, 1995; Kotchenruther and Hobbs, 1998; Kotchenruther et al., 1999].

In calculating the direct radiative forcing of climate by sulfates, Charlson et al. [1991,1992] used a value of 1.7 for the hygroscopic growth factor because the sulfate mass scattering efficiency increased by a factor of 1.7 when the RH rose from 50% to a mean RH of 75% – 80% for the near-surface atmosphere in the Northern Hemisphere. However, they did not consider the dependence of the upward scatter fraction on RH. Instead, a constant value of 0.29 for the upward scatter fraction for dry aerosols was used. Therefore, it is necessary to investigate an RH dependence of the upward scatter fraction as well as the total scattering to estimate the direct radiative forcing by aerosols.

### 1.1.2. Temporal Trends of Black Carbon

Black carbon (BC) is defined as the light absorbing component of carbonaceous aerosols. The effects of BC on climate forcing (i.e., a warming influence on the
atmosphere, by adding to greenhouse forcing or by decreasing cooling by the scattering components of aerosols), although recognized as potentially important, were estimated with large uncertainties [Penner et al., 1994; IPCC, 2001].

The BC is mainly produced by incomplete combustion of fossil and biomass fuels. It is ubiquitous and can be found in soils, ice, sediments and the atmosphere. Wet and dry deposition are known as important sinks for BC [Hansen et al., 1988; Ogren et al., 1984]. Since most of the BC particles are in the fine particle size mode [e.g., Ogren and Charlson, 1983; Penner et al., 1992], dry removal rate is small [Ogren et al., 1984]. Lifetime of BC is relatively short (40 hrs in rainy climates to 1 week in clean, dry regions) and controlled by several factors (e.g., the initial size distribution, the concentration of ambient particles, the frequency and duration of precipitation, and the efficiencies of removal mechanisms) [Ogren and Charlson, 1983]. BC is found in the atmosphere in both urban and remote regions. In general BC is assumed to be inert and non-volatile [Ogren and Charlson, 1983], thus the light absorbing aerosol component measured by optical methods is commonly used as a good tracer for combustion [e.g., Hansen et al., 1988; Allen et al., 1999]. Since the lifetime of the particles with diameters of greater than a few micrometers is quite short, it is necessary when using a global climate model or a regional climate model to concentrate on the fraction of the aerosols which is capable of long-range transport.

The presence of anthropogenic aerosol in a remote region may result in changes in the solar radiation balance. Therefore, the investigation of the seasonal variations and long-term trends in BC aerosol parameters as well as the indication of possible source
region are important. Our investigations of the influence of long-range transport on measured BC mass concentrations at a representative mountain-top site in North Carolina, its seasonal and weekly patterns, and the effects of BC on the regional climate of the southeastern United States will aid in understanding the meteorological transport of atmospheric aerosol and improve the knowledge of the climatology due to aerosol BC in the southeastern United States.

1.1.3. Aerosol, Polar Stratospheric Clouds, and Ozone in the Antarctic Stratosphere

Since PSCs form only under extremely cold conditions, they are observed occasionally in the winter polar vortex region of the Arctic and Antarctic stratosphere [McCormick et al., 1982; Poole and Pitts, 1994]. The maintenance of such a cold polar vortex requires a very weak transport of heat and conservative trace species from the midlatitudes into the inner vortex. However, observational studies did not lead to agreement on the horizontal mixing of the air across the edge of the polar vortex. From analysis of the data from the Airborne Antarctic Ozone Experiment, Hartmann et al. [1989] and Schoeberl et al. [1989, 1992] have concluded that the interior of the Antarctic polar vortex is isolated from the exterior of the vortex and consequently, the vortex acts as a chemical containment vessel. On the other hand, Proffitt et al. [1989] has argued that air inside the polar vortex is mixed continuously with air outside the polar vortex and thus, the vortex acts as a flowing processor. Aerosols in the Antarctic stratosphere can be used as tracers of the circulation within and across the polar vortex as well as tracers of sedimentation processes of PSCs. Therefore, an analysis of temporal and spatial
distributions of aerosol and ozone associated with the Antarctic polar vortex processes could aid in answering the question: “How do stratospheric trace constituents respond to polar vortex processes and influence the ozone depletion?”.

The temperature and potential vorticity of the lower stratosphere during winter and spring are the key factors in explaining the change of the magnitude of springtime Antarctic ozone depletion, since PSCs form under extremely cold conditions [e.g., McCormick et al., 1982; Pole and McCormick, 1988; Browell et al., 1990; Poole and Pitts, 1994] and the maintenance of such a cold polar vortex requires a high degree of isolation inside the polar vortex [e.g., Juckes and McIntyre, 1987; Schoeberl and Hartmann, 1991; Bowman, 1993; Norton, 1994]. However, no studies about how frequently the stratospheric temperature falls below threshold temperature for the formation of PSCs during winter have been reported so far. Therefore, it is necessary to investigate the probability of occurrence of temperature below threshold temperature for PSC formation in the stratosphere during winter. Furthermore, the quantitative relationships between temperature, potential vorticity, and ozone are needed to estimate the lower stratospheric ozone depletion associated with dynamical and physical processes in the Antarctic.
1.2. Objective of this Study

This study is focused on the role of atmospheric aerosols in determining the direct radiative forcing in the lower troposphere and the ozone depletion in the lower stratosphere as associated with atmospheric dynamical processes (i.e., long range transport processes in the surface boundary layer of the southeastern United States and polar vortex processes in the stratosphere over the Antarctic). The results will aid in understanding the microphysics of atmospheric aerosols, the meteorological transport of atmospheric aerosols, and the impacts of PSCs on aerosol distribution and ozone depletion, and ultimately improve the knowledge of the climatology in the southeastern United States and the Antarctic.

The specific objectives of this study are the following:

(1) To study effects of RH on aerosol optical properties and direct aerosol radiative forcing on the basis of field measurements and model calculations for the southeastern United States.

(2) To investigate the influence of long-range transport on measured BC mass concentrations at a representative mountain-top site in North Carolina, its seasonal and weekly patterns, and the effects of BC on the regional climate of the southeastern United States.

(3) To study the spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes and to investigate the impact of PSCs on aerosol
distribution and ozone depletion, by analyzing Stratospheric Aerosol and Gas Experiment II (SAGE II) satellite measurements of ozone and aerosol and the National Centers for Environmental Prediction and the National Center for Atmospheric Research (NCEP/NCAR) reanalysis data of potential vorticity and temperature for the period of 1985 – 1999.
1.3. References


Proffitt, M. H., K. K. Kelly, J. A. Powell, B. L. Gary, M. Loewenstein, J. R. Podolske, S. E. Strahan, and K. R. Chan, Evidence for diabatic cooling and poleward transport


Figure 1.1. Geographical distribution of present-day annual-average radiative forcing (W m\(^{-2}\)) due to (a) well-mixed greenhouse gases, (b) stratospheric ozone depletion, (c) increases in tropospheric ozone, (d) the direct effect of sulphate aerosol, (e) the direct effect of organic carbon and black carbon from biomass burning, and (f) the direct effect of organic carbon and black carbon from fossil fuel burning [IPCC, 2001].
The global mean radiative forcing of the climate system for the year 2000, relative to 1750

Figure 1.2. The global mean radiative forcing of the climate system for the year 2000, relative to 1750 [IPCC, 2001]
2. An assessment of hygroscopic growth factors for aerosols in the surface boundary layer for computing direct radiative forcing

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2.1. Abstract

Aerosol optical properties in the southeastern United States were measured at two research sites in close horizontal proximity but at different altitudes at Black Mountain (35.66 °N, 82.38 °W, 951 m msl) and Mount Gibbes (35.78 °N, 82.29 °W, 2006 m msl) to estimate the direct radiative forcing in the lowest 1 km layer of the troposphere during the summer of 1998. Measurements of light scattering and light absorption at ambient relative humidity (RH) are categorized by air mass type (polluted continental, marine with some continental influence, continental) according to 48-hour back-trajectory analysis. At a wavelength of 530 nm the average total scattering coefficient ($\sigma_{sp}$) measured at the valley site was $1.46 \times 10^{-4}$ m$^{-1}$ for polluted continental air masses, $7.25 \times 10^{-5}$ m$^{-1}$ for marine air masses, and $8.36 \times 10^{-5}$ m$^{-1}$ for continental air masses. The ratio of $\sigma_{sp}$ at the mountain site to $\sigma_{sp}$ at the valley site was 0.64, 0.58, and 0.45 for polluted continental, marine, and continental air masses, respectively. The hygroscopic growth factor ($\sigma_{sp}(\text{RH}=80%) / \sigma_{sp}(\text{RH}=30%)$) was calculated to be almost a constant value of $1.60 \pm 0.01$ for polluted continental, marine, and continental air masses. As the RH increased from 30% to 80%, the backscatter fraction decreased by 23%. On the basis of these measurements, direct radiative climate forcing ($\Delta F_R$) by aerosols in the lowest 1 km layer of the troposphere was estimated. The patterns of $\Delta F_R$ for various values of RH were similar for the three air masses, but the magnitudes of $\Delta F_R(\text{RH})$ were larger for polluted continental air masses than for marine and continental air masses by a factor of about 2 due to higher sulfate concentration in polluted continental air masses. The
average value of $\Delta F_R(\text{RH}=80\%) / \Delta F_R(\text{RH}=30\%)$ was calculated to be almost a constant value of $1.45 \pm 0.01$ for all three types of air masses. This implies little dependence of the forcing ratio on the air mass type. The averaged $\Delta F_R$ for all the observed ambient RHs, in the lowest 1 km layer during the 3-month summer period, was $-2.95 \text{ W m}^{-2}$ (the negative forcing of $-3.24 \text{ W m}^{-2}$ by aerosol scattering plus the positive forcing of $+0.30 \text{ W m}^{-2}$ by aerosol absorption) for polluted continental air masses, $-1.43 \text{ W m}^{-2}$ ($-1.55$ plus $+0.12$) for marine air masses, and $-1.50 \text{ W m}^{-2}$ ($-1.63$ plus $+0.14$) for continental air masses. The $\Delta F_R$ for polluted continental air masses was approximately twice that of marine and continental air masses. These forcing estimates are calculated from continuous \textit{in situ} measurements of scattering and absorption by aerosols without assumptions for Mie calculations and global mean column burden of sulfates and black carbon (in g m$^{-2}$) used in most of the model computations.
2.2. Introduction

It is known that aerosol particles influence the Earth’s radiative balance directly by backscattering and absorption of solar radiation [e.g., Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Penner et al., 1994; Haywood and Shine, 1995; Chýlek and Wong, 1995]. Backscattering of solar radiation by aerosol particles, such as sulfates, can cause climate cooling, while absorption of solar radiation by aerosol particles, such as black carbon, can induce climate warming, depending upon the albedo of the underlying terrain and the height of the aerosol layer above the ground. In contrast to the long lifetime of greenhouse gases (decades to a century), atmospheric aerosol particles have a relatively short lifetime (couple of days to a week) in the troposphere. The short lifetime of aerosol particles, together with the highly nonuniform distribution of aerosol sources, can result in very different patterns of radiative forcing and different climate response patterns. Numerical model calculations of radiative forcing of climate by sulfate aerosol indicate that a large magnitude of negative forcing occurs in local regions of the eastern United States, eastern China, and south central Europe [Charlson et al., 1991; Kiehl and Briegleb, 1993]. A regional cooling trend in the mean maximum surface temperature over the southeastern United States from 1949 to 1994 [Saxena and Yu, 1998] is potentially an observational confirmation for these predictions. However, the lack of sufficiently detailed observations of aerosol optical properties leads to considerably large uncertainties in calculating radiative forcing on climate [Charlson et al, 1992; Penner et al., 1994; Intergovernmental Panel on Climate Change (IPCC), 1994; Schwartz and
Andreae, 1996]. To reduce the uncertainties of current estimates of aerosol climate forcing, more integrated studies of aerosols are needed.

The aerosol light scattering and absorption properties depend on the aerosol particle number size distribution, chemical composition (refractive index) and shape of the particles, the degree of mixing of the particles (external and internal mixture), and the wavelength of the incident light [Bohren and Huffman, 1983; Martins et al., 1998]. In addition, the light scattering of aerosol is strongly dependent on the relative humidity (RH) at which it is measured, because of the hygroscopic growth nature of most atmospheric aerosols [Tang and Munkelwitz, 1977; Tang, 1980; Svenningsson et al., 1994]. The RH dependence of aerosol light scattering (the so-called hygroscopic growth factor) has been studied experimentally [Charlson et al., 1984; Malm et al., 1994; Hegg et al., 1996; Carrico et al., 1998] and theoretically [Hänel, 1976; Hegg et al., 1993] and has been used in the computation of direct radiative climate forcing by aerosol [Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Boucher and Anderson, 1995; Kotchenruther and Hobbs, 1998; Kotchenruther et al., 1999].

In calculating the direct radiative forcing of climate by sulfates, Charlson et al. [1991, 1992] used a value of 1.7 for the hygroscopic growth factor because the sulfate mass scattering efficiency increased by a factor of 1.7 when the RH rose from 50% to a mean RH of 75% – 80% for the near-surface atmosphere in the Northern Hemisphere. However, they did not consider the dependence of the upward scatter fraction on RH. Instead, a constant value of 0.29 for the upward scatter fraction for dry aerosols was used. To estimate the direct radiative forcing by biomass burning, Kotchenruther and Hobbs
[1998] considered an RH dependence of the upward scatter fraction as well as the total scattering. Their results showed a reduced magnitude of negative radiative forcing.

In this paper, we study effects of RH on aerosol optical properties and direct aerosol radiative forcing on the basis of field measurements and model calculations for the southeastern United States. The relationship of these aerosol optical properties and direct radiative forcing to different air mass types influencing the research sites is also investigated. By performing these continuous in situ measurements of scattering and absorption by aerosols and model computations, direct radiative forcing for different air masses in the lowest 1 km tropospheric layer is evaluated with various values of RH.

2.3. Methodology

2.3.1. Experimental Sites

The experimental sites are located in the Blue Ridge Mountains of western North Carolina. They consist of a valley site near the town of Black Mountain (35.66 °N, 82.38 °W, 951 m mean sea level (msl)) and a mountain-top site on the peak of Mount Gibbes (35.78 °N, 82.29 °W, 2006 m msl) in Mount Mitchell State Park. The horizontal separation between the two sites is 10 km. The measurements were obtained from June to August 1998. The air masses arriving at these mountain and valley research sites were classified into three different categories, i.e., polluted continental, marine with some continental influence, and continental, based on the SOx and NOx emission inventories obtained from the U.S. Environmental Protection Agency [1993] as shown in Figure 2.1. (Detailed descriptions on the sector division can be found in the work of Ulman and
To determine the sector of origin of an air mass, a 48-hour back trajectory, calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HY-SPLIT) model [Draxler and Hess, 1997], was drawn at the height of the mountain site every 6 hours during the entire sampling period. The total number of trajectories drawn for this study was 321. In situations where the back trajectories crossed the boundaries between adjacent sectors, the classification of air mass history became somewhat subjective and was based on the perceived amount of time spent in any one sector. Since we assume the air mass arriving at the research site is transported from its source of origin, the classification of air mass does not imply a pure air mass but rather a modified air mass.

Validation of the air mass sector classification was obtained through analysis of cloud water pH [Ulman and Saxena, 1997], the ionic concentration found in cloud water [Deininger and Saxena, 1997], and black carbon (BC) concentration [Bahrmann and Saxena, 1998]. Deininger and Saxena [1997] showed that cloud-forming air masses from the polluted continental sector were abundant in sulfates. They also found that air masses from the marine sector were characterized by the presence of sea salts, and air masses from the continental sector were characterized by the presence of calcium ions, although a major component in both marine and continental air masses was also sulfate. Ulman and Saxena [1997] reported the lowest average pH value in the polluted continental sector. Bahrmann and Saxena [1998] showed the highest average concentration of BC transported from the polluted continental sector, while the lowest values originate in the marine sector.
2.3.2. Instrumentation

A Radiance Research M-903 nephelometer (1-\(\lambda\) nephelometer) measured the total light scattering of aerosol, a Magee Scientific aethalometer measured the light absorption of black carbon (BC), and a TSI integrating nephelometer (model 3550/3560) (3-\(\lambda\) nephelometer) measured total light scattering and backscattering of aerosol. The sampling period and the number of samples for all instrument deployed at each research site are summarized in Table 1.

The M-903 portable nephelometer measures the total light scattering coefficients (\(\sigma_{sp}\)) at a wavelength of 530 nm (near the peak of the solar radiation spectrum) using the geometry of a standard integrating nephelometer under ambient RH at the mountain and valley sites. With general meteorological values (i.e., pressure, temperature, and RH), the values of \(\sigma_{sp}\) are monitored and recorded as 15-min averages during the entire research period. Calibration is accomplished by adjusting the span so that the indicated scattering coefficient matches the calculated value when sampling calibration span gas at local pressure and temperature conditions. Particle free air and Freon gas (CHClF\(_2\)) are used as zero reference and span gas, respectively. The optical and electrical background noise (usually wall scatter) is sufficiently low to allow measurement of \(\sigma_{sp}\) from less than 10\(^{-6}\) m\(^{-1}\) to greater than 10\(^{-3}\) m\(^{-1}\).

The TSI integrating nephelometer integrates over scattering angles from 7\(^\circ\) to 170\(^\circ\) for total scattering (\(\sigma_{sp}\)) and from 90\(^\circ\) to 170\(^\circ\) for backscattering (\(\sigma_{bsp}\)) at three wavelengths of 450, 550 and 700 nm. Meteorological data (temperature, pressure, and RH) as well as \(\sigma_{sp}\) and \(\sigma_{bsp}\) at ambient RH are recorded as 5-min averages. The
nephelometer detects the scattering properties of aerosol particles by measuring the light scattered by the aerosol and subtracting light scattered by the walls of the measurement chamber, light scattered by the gas, and electronic noise inherent in the detectors. A 75 W quartz-halogen lamp, with a built-in reflector, provides illumination for the aerosol. To calibrate scattered signals, the nephelometer uses a reference chopper making a full rotation 23 times per second. The chopper consists of three separate areas (signal, dark, and calibrate sections). Particle-free air and CO₂ gas are used as zero reference and span gas, respectively, adjusting for the appropriate scattering values. A high-efficiency particulate air (HEPA) filter is switched periodically in line with the inlet to subtract the light scattered by the gas portion of the aerosol. The backscatter fraction (β) is defined as the ratio of σ_{bsp} to σ_{sp}. This ratio was calculated from σ_{sp} and σ_{bsp} measurements obtained from the TSI-integrating nephelometer at the Black Mountain site during the period of July 14 – 24.

The aethalometer [Hansen et al., 1984; Hansen, 1996] was deployed at the Mount Gibbes site and provided real-time, continuous measurement of BC mass concentration. The BC measurements were recorded as 15-min intervals. It is designed to measure the fraction of the carbonaceous aerosol that absorbs light over a broad region of the visible spectrum by determining the attenuation of the light transmitted through the sample when collected on a fibrous filter. Gundel et al. [1984] have shown that optical absorption is proportional to BC mass concentration, such that it is frequently assumed that BC mass concentration (g m⁻³) is equal to the absorption coefficient (m⁻¹) divided by the absorption efficiency (m² g⁻¹). The value of absorption efficiency depends on the size of particles,
the incident light wavelength, and the type of mixing between BC and scattering component (such as organic matter and sulfates) [e.g., Waggoner et al., 1981; Rosen and Hansen, 1984; Lioussé et al., 1993; Chýlek et al., 1995; Martins et al., 1998]. In general, internally mixed particles composed of an absorbing core surrounded by a nonabsorbing shell have a greater BC mass absorption efficiency than pure BC particles or externally mixed particles [e.g., Ackerman and Toon, 1981]. This is due to the nonabsorbing shell increasing the total cross-section area of the particles and focusing light toward the absorbing core, causing the same amount of BC in the internally mixed structure to absorb more than pure BC particles [Martins et al., 1998]. Several authors [Clarke et al., 1987; Japar et al., 1986; Roessler and Faxvog, 1980] obtained values around 10 m² g⁻¹ for absorption efficiency of aerosol particles, and Gundel et al. [1984] reported a value of 25 m² g⁻¹ by comparing direct thermal measurements of ambient BC with measurements of light attenuation. In addition, Martins et al. [1998] reported absorption efficiencies measured for various types of biomass fires during the Smoke, Cloud, Radiation–Brazil (SCAR-B) experiment ranging between 5.2 and 19.3 m² g⁻¹ with an average value of 12.1 ± 4.0 m² g⁻¹. The value used in our aethalometer is 19 m² g⁻¹ (as determined by the manufacturer), and it is an empirical calibration factor based on a number of studies at different sites and under different conditions. The absorption coefficient (σap) was derived by applying the manufacturer-specified absorption efficiency value to the BC mass concentration measurements of the aethalometer. During November 1997 a field experiment comparison of the absorption coefficients as derived from the aethalometer and a soot absorption photometer demonstrated a reasonably consistent agreement. This
result indicates that the use of an absorption efficiency of 19 m$^2$ g$^{-1}$ is appropriate for our aethalometer.

2.4. Results and Discussions

During the measurement period, 56% of air masses arriving at the research site were transported from the polluted continental sector, 7% from the marine sector, and 37% from the continental sector. The use of trajectory analysis is only applicable for a general description of the flow field because the accuracy of these trajectories, as with all computed trajectories, is limited and depends on a variety of factors including wind field resolution and associated synoptic-scale meteorological conditions [e.g., Draxler, 1987; Stunder, 1996].

2.4.1. Total Scattering

The $\sigma_{sp}$ measured at the valley and mountain sites were averaged for each air mass type (Figure 2.2). The average value measured at wavelength of 530 nm was $1.46 \times 10^{-4}$ ($\pm 7.78 \times 10^{-5}$) m$^{-1}$ for polluted continental air masses, $7.25 \times 10^{-5}$ ($\pm 2.42 \times 10^{-5}$) m$^{-1}$ for marine air masses, and $8.36 \times 10^{-5}$ ($\pm 4.94 \times 10^{-5}$) m$^{-1}$ for continental air masses, at the valley site. The averaged values at the mountain site were smaller; $9.32 \times 10^{-5}$ ($\pm 5.15 \times 10^{-5}$) m$^{-1}$ for polluted continental air masses, $4.24 \times 10^{-5}$ ($\pm 3.79 \times 10^{-5}$) m$^{-1}$ for marine air masses, and $3.74 \times 10^{-5}$ ($\pm 2.88 \times 10^{-5}$) m$^{-1}$ for continental air masses. The ratio of $\sigma_{sp}$ at the mountain site to $\sigma_{sp}$ at the valley site was 0.64, 0.58, and 0.45 for polluted continental, marine, and continental air masses, respectively. These are much smaller values
compared to the ratio of pressure at the mountain site to pressure at the valley site (i.e., 0.87) calculated from mean pressure values of 932mb at the valley site and 807mb at the mountain site. This implies that aerosol mass concentration in the lower boundary layer does not necessarily follow the pressure ratio. Previously reported values of $\sigma_{sp}$ measured at different regions of the lower troposphere, for 500 – 550 nm wavelength at low RH are typically in the range $5 \times 10^{-5} – 3 \times 10^{-4} \text{ m}^{-1}$ for polluted continental (nonurban) case, $5 \times 10^{-6} – 3 \times 10^{-5} \text{ m}^{-1}$ for clean continental case, and $5 \times 10^{-6} – 2 \times 10^{-5} \text{ m}^{-1}$ for clean marine case [IPCC, 1994].

Diurnal variations of RH and $\sigma_{sp}$ at a wavelength of 530 nm measured by the nephelometer for polluted continental, marine, and continental air masses arriving at the valley site are shown in Figure 2.3. Since the diurnal variation in temperature causes a variation in RH with an inverse relationship, diurnal changes of RH similar to those in Figure 2.3 have been observed in many climates (e.g., Singapore [http://www.gov.sg/metsin/] and Florida [http://www.ksc.nasa/biomed/climate/climate.html/]). At our research site the maximum RH is observed in the early morning and the minimum RH is observed in the midafternoon. The trends of diurnal variation of $\sigma_{sp}$ were coincident with those of RH. This implies that high RH causes hygroscopic particles to grow into an optically active size range and hence increase scattering.

2.4.2. Single Scatter Albedo

The single scatter albedo ($\omega_0$) defined as the ratio of $\sigma_{sp}$ to ($\sigma_{sp} + \sigma_{ap}$), is the fraction of total attenuation by a single particle that is due to scattering. Figure 2.4 shows $\sigma_{sp}$, $\sigma_{ap}$,
and \( \omega_0 \) averaged for each air mass type at the mountain site. The average value of \( \sigma_{ap} \) was \( 6.10 \times 10^{-6} \, (\pm \ 2.82 \times 10^{-6}) \) m\(^{-1}\) for polluted continental air masses, \( 2.51 \times 10^{-6} \, (\pm \ 1.65 \times 10^{-6}) \) m\(^{-1}\) for marine air masses, and \( 2.81 \times 10^{-6} \, (\pm \ 1.90 \times 10^{-6}) \) m\(^{-1}\) for continental air masses. These are comparable to the value of \( 4.8 \times 10^{-6} \) m\(^{-1}\) measured in the polluted continental boundary layer in the Mexico City basin [Raga et al., 1998]. Typical ranges [IPCC, 1994] of \( \sigma_{ap} \) in the lower tropospheric aerosols are reported to be \( 5 \times 10^{-6} - 5 \times 10^{-5} \) m\(^{-1}\) for the polluted continental (nonurban) case, \( 1 \times 10^{-6} - 1 \times 10^{-5} \) m\(^{-1}\) for the clean continental case, and \( 1 \times 10^{-8} - 5 \times 10^{-8} \) m\(^{-1}\) for the clean marine case.

The calculated \( \omega_0 \) value was \( 0.93 \pm 0.04 \) for polluted continental air masses, \( 0.92 \pm 0.06 \) for marine air masses, and \( 0.91 \pm 0.06 \) for continental air masses. In general, when \( \sigma_{ap} \) is large, \( \sigma_{sp} \) is large. This implies that the anthropogenic aerosols that cause scattering and absorption are emitted simultaneously during fossil fuel combustion processes. Thus the mean values of \( \omega_0 \) show constancy for the three air mass types. However, because diurnal variations of \( \sigma_{ap} \) were not observed, it should be noted that the constancy of \( \omega_0 \) does not imply diurnal variations of \( \sigma_{ap} \) similar to those of \( \sigma_{sp} \). Indeed, the different characteristics of \( \sigma_{sp} \) and \( \sigma_{ap} \) with respect to RH induce the deviations from the mean value of \( \omega_0 \). In order to eliminate the diurnal variation of \( \sigma_{sp} \), daily average values of \( \omega_0 \) were recalculated from daily mean \( \sigma_{sp} \) and \( \sigma_{ap} \). As a result, the standard deviation of \( \omega_0 \) was calculated to be 0.02 for the three air mass types. The daily mean \( \sigma_{sp} \) and \( \sigma_{ap} \) were well correlated (correlation coefficient, \( R=0.78 \)), which suggest that aerosols originated from the same emission sources. Ichoku et al. [1999] also reported
that fine BC correlates with sulfur in an eastern Mediterranean arid environment, suggesting that they may have originated from the same anthropogenic sources or source regions.

In comparison, Ogren and Sheridan [1996] reported values of $\omega_0 = 0.88 - 0.96$ for aircraft measurement of tropospheric aerosols over the United States, Penner et al. [1992] estimated $\omega_0 = 0.92$ for biomass burning aerosols, Haywood and Shine [1995] calculated $\omega_0 = 0.8 - 0.96$ for plausible soot/sulfate aerosol composition using a Mie model, Tegen et al. [1996] reported values of $\omega_0 = 0.85 - 0.95$ for aerosols from disturbed soils in arid regions, and Yu et al. [2000] estimated $\omega_0 = 0.74 - 0.99$ for a ground albedo of 0.19 in the southeastern US.

In the study of radiative forcing and climate response, Hansen et al. [1997] calculated the critical single scatter albedo ($\omega_0^*$), at which the aerosol impact on the global mean surface temperature shifts from cooling to heating, to be 0.86 for fixed clouds. Values of $\omega_0$ averaged for the three different air masses influencing our research site are greater than this critical value of 0.86. In addition, 96.4% of polluted continental air masses arriving at the site had an $\omega_0 \geq 0.86$, 85.2% of marine air masses had an $\omega_0 \geq 0.86$, and 90.4% of continental air mass had an $\omega_0 \geq 0.86$. These results support the model prediction [e.g., Kiehl and Briegleb, 1993] that our research site is in a climate cooling region.
2.4.3. Humidity and Light Scattering

The light scattering of aerosol is strongly dependent on the RH at which it is measured, due to the hygroscopic growth nature of most atmospheric aerosols. These hygroscopic growth curves are generally described in the form of $\sigma_{sp}$ as a function of RH normalized to $\sigma_{sp}$ at 30% RH at which RH the aerosol is assumed dry with a very weak dependence of $\sigma_{sp}$ on RH. For assessment of the impact of aerosols on direct radiative forcing of climate, Charlson et al. [1991, 1992] used an RH of 80%, which they judged a reasonable mean value for the atmospheric boundary layer and selected the value of $1.7 \pm 0.3$ for the ratio of $\sigma_{sp}(\text{RH}=80\%)$ to $\sigma_{sp}(\text{RH}<50\%)$.

The average variations of $\sigma_{sp}$ with RH for the different air masses influencing the layer between the valley and the mountain sites (in which a mean value of $\sigma_{sp}$ measured at the valley and mountain sites was used to represent $\sigma_{sp}$ in the layer) are shown in Figure 2.5. These data were fitted to an expression of the general form derived by Kasten [1969]:

$$\sigma_{sp}(\text{RH}) = \sigma_{spd} \left(1 - \frac{\text{RH}}{100}\right)^{-g}$$

(1)

where $\sigma_{sp}$ is the total scattering coefficient at a given RH, $\sigma_{spd}$ is the fitted total scattering coefficient for the dry aerosol, and $g$ is another empirical fitting parameter.

The value of $g$ was calculated to be $0.38 \pm 0.03$ with an $R^2=0.94$ for polluted continental air masses, $0.37 \pm 0.05$ with an $R^2=0.84$ for continental air masses, and $0.38 \pm$
0.05 with an $R^2=0.85$ for marine air masses. Figure 2.5 also shows that $\sigma_{sp}$ for polluted continental air masses is $2.16 - 2.18$ times larger than $\sigma_{sp}$ for continental and marine air masses. However, the hygroscopic growth factor, the ratio of $\sigma_{sp}(\text{RH}=80\%)$ to $\sigma_{sp}(\text{RH}=30\%)$, was calculated to be 1.61 for polluted continental air masses, 1.61 for marine air masses, and 1.59 for continental air masses. These nearly constant values of hygroscopic growth factor calculated for different air mass types do not differ significantly from the constant value of 1.7 for urban and ground level measurements, selected by Charlson et al. [1991, 1992]. In general, the value of the hygroscopic growth factor decreases as the diameter of particle increases. That is, a smaller particle is more sensitive to increasing RH [Hegg et al., 1993, 1996; Boucher and Anderson, 1995]. However, our results indicate that the hygroscopic growth factor is nearly constant for the three different air masses. This may be because the mean particle sizes of air masses arriving at the site are similar for the three air mass types [Menon, 1998] and the major component in the three air mass types is sulfate [Deininger and Saxena, 1997].

On the other hand, the backscatter fraction ($\beta$) also has an RH dependence, that is, the value of backscatter fraction decreases as RH increases. Figure 2.6 shows the dependence of the backscatter fraction on RH as well as wavelength. Unlike characteristics of total scattering, the backscatter fraction decreases with decreasing wavelength and with increasing RH. This association can be explained by scattering patterns with increasing particle size parameter (defined as the ratio, $\pi \times$ particle diameter to wavelength of incident light). When particle diameter is much less than the wavelength of incident light (i.e., particle size parameter $<< 1$), Rayleigh-scattering
pattern is shown, and for this case, the value of $\beta$ becomes 0.5 (i.e., forward scatter fraction is equal to backward scatter fraction.). When particle diameter is on the order of the wavelength of incident light (i.e., particle size parameter $\equiv 1$), a Mie-scattering pattern is shown, and for this case, the value of $\beta$ becomes less than the value of forward scatter fraction (that is, the value of $\beta$ is less than 0.5). Thus for a fixed particle size, $\beta$ decreases as the wavelength decreases, and for a fixed wavelength, $\beta$ decreases as the particle size increases. In our study, the values of $\beta$ decrease linearly with increasing RH having the linear regression equation of $\beta = -0.0005 \times \text{RH} + 0.12$ with an $R^2 = 0.95$, and the humidification factor of $\beta$ defined as the ratio of $\beta(\text{RH}=80\%)$ to $\beta(\text{RH}=30\%)$ was calculated to the value of 0.77 for the wavelength of 550 nm. In a similar study, Kotchenruther et al. [1999] reported that $\beta$ decreased by 30% to 40% as the RH increased from 30% to 80% at the mid-Atlantic coast of the US in summer season.

Since the radiation backscattered by aerosols is a key parameter in direct radiative forcing, the RH dependence of $\beta$ is important. The $\beta$ calculated from measurements of the nephelometer can be considered as the aerosol light backscatter to space, assuming that the Sun is directly overhead. For direct radiative forcing computations, all Sun angles during the daytime must be considered. Therefore the average solar radiation backscattered to space during the 12 hours, the so-called upscatter fraction ($\beta$), was calculated from the related quantity $\beta$ using the Henyey-Greenstein phase function [Wiscombe and Grams, 1976]. At the wavelength of 550 nm the humidification factor of backscatter fraction, i.e., the ratio of $\beta(\text{RH}=80\%)$ to $\beta(\text{RH}=30\%)$, was calculated to be
0.77 and then the humidification factor of upscatter fraction, i.e., the ratio of $B_{\text{RH}=80\%}$ to $B_{\text{RH}=30\%}$, was calculated to be 0.87. This indicates that $B$ was less sensitive to increases in RH than $\beta$.

### 2.4.4. Effects of Relative Humidity on Direct Radiative Forcing

The direct radiative forcing by aerosols in the lowest 1 km layer of the atmosphere in the southeastern United States, $\Delta F_R$, can be estimated from the equation obtained by Chýlek and Wong [1995]:

$$
\Delta F_R = -\frac{S_o}{4} T_{\text{atm}}^2 (1 - A_c) \left[ (1 - R_s)^2 2B \tau_{\text{sc}} - 4R_s \tau_{\text{abs}} \right],
$$

where $S_o$ is the solar constant, $T_{\text{atm}}$ is the atmospheric transmission, $A_c$ is the fractional cloud amount, $R_s$ is the surface albedo of the underlying surface, $B$ is the fraction of average daily solar radiation scattered back to space (i.e., upscatter fraction computed from $\beta$), and $\tau_{\text{sc}}$ and $\tau_{\text{abs}}$ are the optical depth of the aerosol layer due to light scattering and absorption, respectively. For a nonabsorbing aerosol $\tau_{\text{abs}} = 0$, the above reduces to the equation used by Charlson et al. [1992], a box model applied for nonabsorbing sulfate aerosols. However, for absorbing aerosols the second term on the right-hand side of the above equation cannot be neglected. The advantage of the above equation is an explicit dependence on the individual parameters determining the direct radiative forcing. In general, the direct radiative forcing by aerosols refers to changes in radiative fluxes by
anthropogenic or episodic (e.g., volcanic) aerosols from a preindustrial or background condition. Since measurements in this research did not allow separation of the natural and anthropogenic aerosol components, the direct radiative forcing values presented here are the radiative flux perturbations due to the combined effects of natural and anthropogenic aerosols.

To calculate $\tau_{sc}$ and $\tau_{abs}$, the following equations were used:

\[
\tau_{sc} (\text{RH}) = \int_{z_1}^{z_2} \sigma_{sp} (\text{RH}) dz, \tag{3}
\]

\[
\tau_{abs} = \int_{z_1}^{z_2} \sigma_{ap} dz. \tag{4}
\]

Parameters used to calculate the direct radiative forcing in this study were $S_0 = 1370 \text{ W m}^{-2}$, $T_{\text{atm}} = 0.76$, $A_c = 0.61$, and $R_s = 0.15$ based on the values used in the box model [Charlson et al., 1992]. The values of $\tau_{sc}(\text{RH})$ and $\tau_{abs}$ in the lowest 1 km atmospheric layer were calculated by the following equations:

\[
\tau_{sc} = \left[ \frac{(\sigma_{sp})_m + (\sigma_{sp})_v}{2} \right] (z_m - z_v), \tag{5}
\]

\[
\tau_{abs} = (\sigma_{ap})_m (z_m - z_v), \tag{6}
\]
where \((\sigma_{sp})_m\) and \((\sigma_{sp})_v\) are \(\sigma_{sp}\) measured at the mountain site and the valley site, respectively and \((\sigma_{ap})_m\) is \(\sigma_{ap}\) measured at the mountain site, and \(z_m\) and \(z_v\) are heights of mountain and valley, respectively. The \(\tau_{abs}\) was calculated to be 0.0064 for polluted continental air masses, 0.0026 for marine air masses, and 0.0030 for continental air masses. The air mass dependence of \(B(RH)\) was ignored, which is a plausible assumption because the hygroscopic growth factor of \(\sigma_{sp}\) calculated in this study was similar for the three different air mass types. Parameters for various values of \(RH\), used to evaluate the direct radiative forcing are shown in Table 2, and the results of direct radiative forcing calculated for various values of \(RH\) are shown in Figure 2.7 and Table 2. The magnitude of \(\Delta F_R\) \((RH)\) for polluted continental air masses was found be larger than that of marine and continental air masses by a factor of \(\approx 2.14\) due to higher concentration of sulfates in polluted continental air masses. Since the magnitude of \(\tau_{sc}\) is much larger than that of \(\tau_{abs}\), the contribution of \(\tau_{sc}\) to \(\Delta F_R\) was dominant, resulting in negative forcing of \(\Delta F_R\) for the entire range of \(RH\) in our search site. The effects of \(RH\) on direct radiative forcing estimated for different air masses are shown in Figure 2.8, in which \(\Delta F_R(\text{RH}=30%)\) and \(\Delta F_R(\text{RH})\) are the values of the aerosol direct radiative forcing at 30\% \(RH\) and at a specified \(RH\), respectively. The influence of \(RH\) on the direct radiative forcing is nearly the same for polluted continental, marine, and continental air masses. For example, \(\frac{\Delta F_R(\text{RH}=80%)}{\Delta F_R(\text{RH}=30\%)}\) was calculated to be 1.46 for polluted continental air masses, 1.45 for marine air masses, and 1.44 for continental air masses with an overall average value of 1.45. The overall average value of effects of \(RH\)
on the radiative parameters and the direct radiative forcing are shown in Table 3. If the radiative parameters relevant to the direct radiative forcing are measured at RH=30%, the values of humidification factor in Table 3 can be used to compute $\Delta F_R$ for mean ambient atmospheric RH in the lower troposphere.

In comparison, Boucher and Anderson [1995] calculated the effective RH factor defined as the ratio between climate forcing by the hydrated aerosol using RH predicted by the General Circulation Model and climate forcing computed for a constant RH value of 30% to be between 1.19 and 1.71. The effective RH factors tested by the different chemical forms were shown to be 1.40 for $(\text{NH}_4)_2\text{SO}_4$, 1.39 for $(\text{NH}_4)\text{HSO}_4$, and 1.40 for $\text{H}_2\text{SO}_4$. These indicate little chemical dependence of the effective RH factor for sulfate aerosol. A compensating effect, linking increases in $\sigma_{sp}$ associated with decreases in B, also indicates low sensitivity to sulfate aerosol size in computation of the effective RH factor. Hence they suggested that three optical parameters used in the Charlson et al. [1992] model (i.e., the sulfate scattering efficiency at low RH, the upscatter fraction, and the increase in scattering from low RH to typical tropospheric values like 80%) may be combined into a single parameter (i.e., the backscattering efficiency at high RH) in order to reduce the model uncertainties. Consequently, they mentioned that low sensitivity to sulfate aerosol size and chemistry would mean that direct sulfate climate forcing can be incorporated in global climate models with only a knowledge of sulfate mass concentration. The results obtained in our study (i.e., nearly constant value of effective RH factor for different air mass types) support this suggestion.
The effective RH factor estimated by our observations is related to the change in RH due to diurnal variation of temperature, assuming constant H$_2$O amount in the atmosphere. In terms of global climate change, a change in RH due to an increase in H$_2$O vapor amount would increase not only negative forcing (i.e., cooling by aerosol due to hygroscopic growth effects) but also positive forcing (i.e., greenhouse warming effect), counteracting each other.

2.4.5. Direct Radiative Forcing at Mean Ambient Relative Humidity

The average direct radiative forcing (by scattering and absorption of aerosol) for all the observed ambient RHs, in the lowest 1 km layer of the troposphere for the three air masses influencing the research site during the 3-month summer period was calculated using equations (2), (5), and (6). The values of $S_o$, $T_{atm}$, $A_c$, and $R_s$ used in this computation were the same as those in the previous section. The calculated value of $\tau_{sc}$ was 0.127 for polluted continental air masses, 0.061 for marine air masses, and 0.064 for continental air masses; and the calculated value of $\tau_{abs}$ was 0.0064 for polluted continental air masses, 0.0026 for marine air masses, and 0.0030 for continental air masses. The $B$ used in the computation of $\Delta F_R$ was derived using the regression equation of $\beta$ in Figure 2.6 and Henyey-Greenstein phase function [Wiscombe and Grams, 1976] to be 0.23 at RH=68%. This RH is the mean ambient RH during the measurement and is consistent with the clear-sky daytime RH (68.6%) in the eastern United States during July, obtained by Boucher and Anderson [1995]. The direct radiative forcing by aerosol absorption was computed to be +0.30 W m$^{-2}$ for polluted continental air masses, +0.12 W
m$^{-2}$ for marine air masses, and $+0.14$ W m$^{-2}$ for continental air masses. In comparison, *Haywood and Shine* [1995] estimated the global mean radiative forcing due to fossil fuel derived soot aerosol to be between $+0.04$ and $+0.18$ W m$^{-2}$, assuming a soot/sulfate mass ratio of 0.075.

The direct radiative forcing by aerosol scattering back to space was calculated to be $-3.24$, $-1.55$, and $-1.63$ W m$^{-2}$ for polluted continental, marine, and continental air masses, respectively. Hence the net direct radiative climate forcing by aerosol was calculated to be $-2.95$ W m$^{-2}$ for polluted continental air masses, $-1.43$ W m$^{-2}$ for marine air masses, $-1.50$ W m$^{-2}$ for continental air masses (Figure 2.9). The net direct radiative forcing by aerosol for polluted continental air masses was approximately twice that of marine and continental air masses.

### 2.5. Summary and Conclusion

The aerosol optical properties suitable for evaluating their climate impacts were measured and the aerosol direct radiative forcing was calculated in this study. The results obtained are the following:

1. At a wavelength of 530 nm, the average value of $\sigma_{sp}$ was $1.46 \times 10^{-4}$, $7.25 \times 10^{-5}$, and $8.36 \times 10^{-5}$ m$^{-1}$ for polluted continental, marine, and continental air masses arriving at the valley site and $9.32 \times 10^{-5}$, $4.24 \times 10^{-5}$, and $3.74 \times 10^{-5}$ m$^{-1}$ for polluted continental, marine, and continental air masses arriving at the mountain site, respectively. The ratio of $\sigma_{sp}$ at the mountain site to $\sigma_{sp}$ at the valley site was 0.64, 0.58, and 0.45 for polluted
continental, marine, and continental air masses, respectively. These are much smaller values compared to the ratio of mean pressure at the mountain site to mean pressure at the valley site (i.e., 0.87) in which mean pressure was 932mb and 807mb at the valley and mountain sites, respectively. This implies that aerosol mass concentration in the lower boundary layer does not necessarily follow the pressure ratio.

(2) The averages of $\sigma_{sp}$ measured at the mountain site was $6.10 \times 10^{-6}$, $2.51 \times 10^{-6}$, and $2.81 \times 10^{-6}$ m$^{-1}$, and $\omega_0$ was calculated to be 0.93 ± 0.04, 0.92 ± 0.06, and 0.91 ± 0.06 for polluted continental, marine, and continental air masses, respectively, which were larger values than critical $\omega_0 (= 0.86)$ obtained by Hansen et al. [1997].

(3) The variation of $\sigma_{sp}$ with RH averaged for different air masses arriving at the layer between the valley and the mountain sites was fitted to the equation of

$$\sigma_{sp}(RH) = \sigma_{spd} \left(1 - \frac{RH}{100}\right)^{-g},$$

with g = 0.38, 0.37, and 0.38 for polluted continental, marine, and continental air masses, respectively, showing similar patterns but different magnitudes. The hygroscopic growth factor ($\sigma_{sp}(RH=80%)/\sigma_{sp}(RH=30%)$) was calculated to be almost a constant value of 1.60 ± 0.01 for polluted continental, marine, and continental air masses.

(4) $\beta$ decreased linearly with increasing RH having $\beta(RH=80%)/\beta(RH=30%) = 0.77$.

(5) The direct radiative forcing by aerosol in the lowest 1 km layer of the troposphere for various RH was estimated. The patterns of $\Delta F_R$ for various values of RH were similar
for the three air masses, but the magnitudes of $\Delta F_R(RH)$ were larger for polluted continental air masses than for marine and continental air masses by a factor of about 2 due to a higher-sulfate concentration in polluted continental air masses. The average value of $\Delta F_R(RH=80\%) / \Delta F_R(RH=30\%)$ was calculated to be almost constant value of $1.45 \pm 0.01$ for all three types of air masses. This implies little chemical, physical dependence of the effective RH factor (defined as $\Delta F_R(RH=80\%) / \Delta F_R(RH=30\%)$) in the southeastern US. Boucher and Anderson [1995] also mentioned that low sensitivity of climate forcing to sulfate aerosol size and chemistry (obtained by a plausible set of assumptions) would mean that direct sulfate climate forcing can be incorporated in global climate models with only a knowledge of sulfate mass concentration.

The averaged $\Delta F_R$ for all the observed ambient RHs in the lowest 1 km layer was $-2.95\; \text{W m}^{-2}$ (the negative forcing of $-3.24\; \text{W m}^{-2}$ by aerosol scattering plus the positive forcing of $+0.30\; \text{W m}^{-2}$ by aerosol absorption) for polluted continental air masses, $-1.43\; \text{W m}^{-2}$ ($-1.55$ plus $+0.12$) for marine air masses, and $-1.50\; \text{W m}^{-2}$ ($-1.63$ plus $+0.14$) for continental air masses. The $\Delta F_R$ for polluted continental air masses was approximately twice that of marine and continental air masses.

These forcing estimates at ambient RH are calculated from continuous in situ measurements of scattering and absorption by aerosols without assumptions for Mie calculations and global mean column burdens of sulfates and black carbon (in g m$^{-2}$) used in most of model computations.
Acknowledgments

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2.6. References


Lioussse, C., H. Cachier, and S. G. Jennings, Optical and thermal measurement of black
carbon aerosol content in different environments: Variation of the specific attenuation
cross-section sigma (σ), *Atmos. Environ.*, 27, 1203-1211, 1993.

Malm, W. C., J. F. Sisler, D. Huffman, R. A. Eldred, and T. A. Cahill, Spatial and
seasonal trends in particle concentration and optical extinction in the United States,

of black carbon content, particle size, and mixing on light absorption by aerosols

Menon, S., Role of sulfates in regional cloud-climate interactions, 238 pp., Ph.D. thesis,

Ogren, J. A., and P. J. Sheridan, Vertical and horizontal variability of aerosol single
scattering albedo and hemispheric backscatter fraction over the United States, in
paper presented at the 14th International Conference on Nucleation and Atmospheric
Aerosols, Helsinki, Finland, 1996.

Penner, J. E., R. E. Dickinson, and C. A. O’Neill, Effects of aerosol from biomass

Penner, J. E., R. J. Charlson, J. M. Hales, N. S. Laulainen, R. Leifer, T. Novakov, J.
Ogren, L. F. Radke, S.E. Schwartz, and L. Travis, Quantifying and minimizing


Table 2.1. Sampling period and number of samples for instruments deployed at the valley and mountain sites during the summer of 1998.

<table>
<thead>
<tr>
<th></th>
<th>Valley</th>
<th>Mountain</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1-λ nephelometer 3-λ nephelometer</td>
<td>1-λ nephelometer aethalometer</td>
</tr>
<tr>
<td>sampling period</td>
<td>6/1 - 8/31</td>
<td>6/1 - 8/31</td>
</tr>
<tr>
<td>number of samples</td>
<td>7911</td>
<td>2774</td>
</tr>
<tr>
<td></td>
<td>7/14 - 7/24</td>
<td>6/1 - 8/31</td>
</tr>
<tr>
<td></td>
<td>8771</td>
<td>8224</td>
</tr>
</tbody>
</table>
Table 2.2. Aerosol radiative parameters and the direct radiative forcing calculated for various values of RH in the lowest 1km layer of the troposphere for three different air masses influencing the research sites during summer 1998.

<table>
<thead>
<tr>
<th>R.H %</th>
<th>β</th>
<th>B</th>
<th>Polluted</th>
<th>Marine</th>
<th>Continental</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>σsp m(^{-1})</td>
<td>τsc</td>
<td>ΔF(_{sc}) *</td>
<td>ΔF (_R)</td>
<td>σsp m(^{-1})</td>
</tr>
<tr>
<td>30</td>
<td>0.110</td>
<td>0.244</td>
<td>8.72×10(^{-5})</td>
<td>0.092</td>
<td>-2.50 -2.20</td>
</tr>
<tr>
<td>35</td>
<td>0.107</td>
<td>0.242</td>
<td>8.97×10(^{-5})</td>
<td>0.094</td>
<td>-2.55 -2.25</td>
</tr>
<tr>
<td>40</td>
<td>0.105</td>
<td>0.241</td>
<td>9.25×10(^{-5})</td>
<td>0.097</td>
<td>-2.61 -2.32</td>
</tr>
<tr>
<td>45</td>
<td>0.102</td>
<td>0.239</td>
<td>9.56×10(^{-5})</td>
<td>0.101</td>
<td>-2.68 -2.39</td>
</tr>
<tr>
<td>50</td>
<td>0.100</td>
<td>0.238</td>
<td>9.91×10(^{-5})</td>
<td>0.104</td>
<td>-2.76 -2.47</td>
</tr>
<tr>
<td>55</td>
<td>0.097</td>
<td>0.234</td>
<td>1.03×10(^{-4})</td>
<td>0.109</td>
<td>-2.84 -2.54</td>
</tr>
<tr>
<td>60</td>
<td>0.095</td>
<td>0.231</td>
<td>1.08×10(^{-4})</td>
<td>0.114</td>
<td>-2.93 -2.63</td>
</tr>
<tr>
<td>65</td>
<td>0.092</td>
<td>0.228</td>
<td>1.14×10(^{-4})</td>
<td>0.120</td>
<td>-3.04 -2.74</td>
</tr>
<tr>
<td>70</td>
<td>0.090</td>
<td>0.225</td>
<td>1.20×10(^{-4})</td>
<td>0.127</td>
<td>-3.18 -2.88</td>
</tr>
<tr>
<td>75</td>
<td>0.087</td>
<td>0.219</td>
<td>1.29×10(^{-4})</td>
<td>0.136</td>
<td>-3.32 -3.02</td>
</tr>
<tr>
<td>80</td>
<td>0.085</td>
<td>0.213</td>
<td>1.41×10(^{-4})</td>
<td>0.148</td>
<td>-3.51 -3.21</td>
</tr>
<tr>
<td>85</td>
<td>0.082</td>
<td>0.209</td>
<td>1.57×10(^{-4})</td>
<td>0.165</td>
<td>-3.86 -3.56</td>
</tr>
<tr>
<td>90</td>
<td>0.080</td>
<td>0.206</td>
<td>8.44×10(^{-5})</td>
<td>0.089</td>
<td>-2.04 -1.92</td>
</tr>
</tbody>
</table>

* ΔF\(_{sc}\) refers to direct radiative forcing due to aerosol scattering.
Table 2.3. Estimated effects of RH on radiative parameters and the direct radiative forcing (which were defined as the ratio of the value at a given RH to the value at RH=30%) averaged for all three air masses in the lowest 1 km layer of the atmosphere.

<table>
<thead>
<tr>
<th>RH</th>
<th>$\sigma_{sp}(RH) / \sigma_{sp}(RH=30%)$</th>
<th>$\beta(RH) / \beta(RH=30%)$</th>
<th>$B(RH) / B(RH=30%)$</th>
<th>$\Delta F_{R}(RH) / \Delta F_{R}(RH=30%)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>40</td>
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<td>0.95</td>
<td>0.99</td>
<td>1.05</td>
</tr>
<tr>
<td>50</td>
<td>1.14</td>
<td>0.91</td>
<td>0.97</td>
<td>1.12</td>
</tr>
<tr>
<td>60</td>
<td>1.23</td>
<td>0.86</td>
<td>0.95</td>
<td>1.19</td>
</tr>
<tr>
<td>70</td>
<td>1.38</td>
<td>0.82</td>
<td>0.92</td>
<td>1.30</td>
</tr>
<tr>
<td>80</td>
<td>1.60</td>
<td>0.77</td>
<td>0.87</td>
<td>1.45</td>
</tr>
</tbody>
</table>
Figure 2.1. Map of the United States illustrating the geographical location and classification of the different air mass types influencing the research sites.
Figure 2.2. Average value of total scattering coefficient at 530nm measured by the nephelometer for three different air masses arriving at the valley and mountain sites during summer 1998.
Figure 2.3. Diurnal variation of RH and total scattering coefficients at 530nm measured by the nephelometer for three air masses arriving at the valley site.
Figure 2.4. Average values of total scattering coefficient, absorption coefficient, and single scatter albedo for three air masses arriving at the mountain site during summer 1998.
Figure 2.5. Variation of averaged total scattering coefficient with averaged RH and fitting for three air masses arriving at the lowest 1 km layer of the troposphere during summer 1998.
Figure 2.6. Variation of averaged backscatter fraction with averaged RH measured from 3-wavelength nephelometer at the valley site during the period of July 14 - 24, 1998.
Figure 2.7. Variation of aerosol direct radiative forcing with RH for three air masses arriving at the lowest 1 km layer of the troposphere during summer 1998.
Figure 2.8. Influence of RH on the aerosol direct radiative forcing for three air masses arriving at the lowest 1 km layer of the troposphere during summer 1998.

\[ y = 6 \times 10^{-9}x^5 - 2 \times 10^{-6}x^4 + 0.0002x^3 - 0.0088x^2 + 0.2211x - 1.1975 \]

\[ R^2 = 0.9997 \]
Figure 2.9. Average direct radiative forcing by scattering and absorption of aerosols in the lowest 1 km layer of the troposphere for the three air masses influencing the research site during summer 1998.
3. Temporal trends of black carbon concentrations and regional climate forcing in the southeastern United States

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Atmospheric Environment, 35, 3293-3302, 2000
3.1. Abstract

The effect of black carbon (BC) on climate forcing is potentially important, but its estimates have large uncertainties due to a lack of sufficient observational data. The BC mass concentration in the southeastern US was measured at a regionally representative site, Mount Gibbes (35.78 °N, 82.29 °W, 2006 m MSL). The air mass origin was determined using 48-hr back trajectories obtained from the Hybrid Single-Particle Lagrangian Integrated Trajectory model. The highest average concentration is seen in polluted continental air masses and the lowest in marine air masses. During the winter, the overall average BC value was 74.1 ng m⁻³, whereas the overall summer mean BC value is higher by a factor of 3. The main reason for the seasonal difference may be enhanced thermal convection during summer, which increases transport of air pollutants from the planetary boundary layer of the surrounding urban area to this rural site. In the spring of 1998, abnormally high BC concentrations from the continental sector were measured. These concentrations were originating from a biomass burning plume in Mexico. This was confirmed by the observations of the Earth Probe Total Ozone Mapping Spectrometer. The BC average concentrations of air masses transported from the polluted continental sector during summer are low on Sunday through Tuesday with a minimum value of 256 ng m⁻³ occurring on Monday, and high on Wednesday through Friday with a maximum value of 379 ng m⁻³ occurring on Friday. The net aerosol radiative forcing (scattering effects plus absorption effects) per unit vertical depth at 2006 m MSL is calculated to be $-1.38 \times 10^{-3}$ W m⁻³ for the southeastern US. The magnitude of
direct radiative forcing by aerosol scattering is reduced by $15 \pm 7\%$ due to the BC absorption.

Key word index: aerosol, black carbon, absorption, direct radiative climate forcing, temporal patterns
3.2. Introduction

The carbonaceous component of atmospheric aerosols is composed of two main fractions (i.e., organic carbon and black carbon). Organic carbon is generally associated with condensed organic compounds which is primarily a scattering medium, whereas black carbon (BC) is generally defined as the absorbing component of carbonaceous aerosols. Absorption of solar radiation by BC heats the atmosphere while reducing solar irradiance at the ground, thereby altering the vertical temperature profile. The effects of BC on climate forcing (i.e., a warming influence on the atmosphere, by adding to greenhouse forcing or by decreasing cooling by the scattering components of aerosols), although recognized as potentially important, were estimated with large uncertainties [IPCC, 1994; Penner et al., 1994].

The BC is mainly produced by incomplete combustion of fossil and biomass fuels. It is ubiquitous and can be found in soils, ice, sediments and the atmosphere. Wet and dry deposition are known as important sinks for BC [Hansen et al., 1988; Ogren et al., 1984]. Since most of the BC particles are in the fine particle size mode [e.g., Ogren and Charlson, 1983; Penner et al., 1992], dry removal rate is small [Ogren et al., 1984]. Barhmann and Saxena [1998] showed BC concentration decreased abruptly after precipitation due to scavenging effects. Lifetime of BC is relatively short (40 hrs in rainy climates to 1 week in clean, dry regions) and controlled by several factors (e.g., the initial size distribution, the concentration of ambient particles, the frequency and duration of precipitation, and the efficiencies of removal mechanisms) [Ogren and Charlson, 1983]. BC is found in the atmosphere in both urban and remote regions. In general BC is
assumed to be inert and non-volatile [Ogren and Charlson, 1983], thus the light absorbing aerosol component measured by optical methods is commonly used as a good tracer for combustion [e.g., Hansen et al., 1988; Allen et al., 1999]. Since the lifetime of the particles with diameters of greater than a few micrometers is quite short, it is necessary when using a global climate model or a regional climate model to concentrate on the fraction of the aerosols which is capable of long-range transport.

The presence of anthropogenic aerosol in a remote region may result in changes in the solar radiation balance. Therefore, the investigation of the seasonal variations and long-term trends in BC aerosol parameters as well as the indication of possible source region are important. In this study, we investigate the influence of long-range transport on measured BC mass concentrations at a representative mountain-top site in North Carolina, its seasonal and weekly patterns, and the effects of BC on the regional climate of the southeastern United States. The results could aid in understanding the meteorological transport of atmospheric aerosol and improve the knowledge of the climatology due to aerosol BC in the southeastern United States.

3.3. Methodology

3.3.1. Experimental Setup

The experimental site (North Carolina State University research station) is located on the peak of Mount Gibbes (35.78 °N, 82.29 °W, 2006 m MSL), in the Blue Ridge Mountains of western North Carolina. The research site lies approximately 4 km southwest of Mount Mitchell (2038 m MSL), the highest peak in eastern North America.
Detailed descriptions of the site are available in *Bahrmann and Saxena* [1998]. The air mass origin was determined using 48-hr back-trajectories obtained from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HY-SPLIT) model [*Draxler and Hess*, 1997]. Air mass source regions were classified as polluted continental, continental, and marine with some continental influence, based on the SO$_x$, NO$_x$ emission inventories obtained from the US Environmental Protection Agency (Figure 3.1). (Detailed descriptions on the sector division can be found in *Ulman and Saxena* [1997].) In situations where the back trajectories crossed the boundaries between adjacent sectors, the classification of air mass history became somewhat subjective and was based on the perceived amount of time spent in any one sector. Since we assume the air mass arriving at the research site is transported from its source of origin, the classification of air mass does not imply a pure air mass but rather a modified air mass. Validation of the air mass sector classification was obtained through analysis of cloud water pH [*Ulman and Saxena*, 1997] and the ionic concentration found in cloud water [*Deininger and Saxena*, 1997]. *Ulman and Saxena* [1997] reported the lowest average pH value to originate in the polluted continental sector. *Deininger and Saxena* [1997] showed that cloud-forming air masses from the polluted continental sector were abundant in sulfates. They also found that air masses from the marine sector were characterized by the presence of sea salts, and air masses from the continental sector were characterized by the presence of calcium, although a major component in both marine and continental air masses was also sulfate.
3.3.2. **Instrumentation**

A Magee Scientific aethalometer was deployed at the site and provided real-time, continuous measurement of BC mass concentration. The measurements were recorded at 15 min intervals and were obtained for Spring (5/4−5/31, 1998), Summer (6/1−8/31, 1998), and Winter (11/11−12/31, 1997 and 12/17−12/31, 1998) to investigate the seasonal patterns of BC concentration.

The aethalometer is designed to measure the fraction of the carbonaceous aerosol that absorbs light over a broad region of the visible spectrum by determining the attenuation of the light transmitted through the sample when collected on a fibrous filter [Hansen, 1996]. Gundel et al. [1984] have shown that optical absorption is proportional to BC mass concentration, such that BC mass concentration (g m\(^{-3}\)) is equal to the measured absorption (i.e., absorption coefficient, m\(^{-1}\)) divided by the absorption efficiency (m\(^2\) g\(^{-1}\)). The value of absorption efficiency depends on the size of particles, the incident light wavelength, and the type of mixing between BC and scattering component (such as organic matter and sulfates) [e.g., Waggoner et al., 1981; Rosen and Hansen, 1984; Liousse et al., 1993; Chýlek et al., 1995; Martins et al., 1998]. Several authors [e.g., Clarke et al., 1987; Japar et al., 1986] obtained values around 10 m\(^2\) g\(^{-1}\) for absorption efficiency of aerosol particles and Gundel et al. [1984] reported a value of 25 m\(^2\) g\(^{-1}\) by comparing direct thermal measurements of ambient BC with measurements of light attenuation. In addition, Martins et al. [1998] reported absorption efficiencies measured for various types of biomass fires ranging between 5.2 and 19.3 m\(^2\) g\(^{-1}\) with an average value of 12.1 ± 4.0 m\(^2\) g\(^{-1}\). The value used in our aethalometer is 19 m\(^2\) g\(^{-1}\) and
can be considered a first step approximation in determining BC mass concentration in our study. Internally mixed particles composed of an absorbing core surrounded by a nonabsorbing shell have, in general, a greater BC mass absorption efficiency than pure BC particles or externally mixed particles [e.g., Ackerman and Toon, 1981; Chýlek et al., 1995]. This is because the nonabsorbing shell increases the total cross-section area of the particles and focuses light toward the absorbing core, causing the same amount of BC in the internally mixed structure to absorb more than pure BC particles [Martins et al., 1998].

The aerosol scattering coefficient was also measured from July 15 to August 13, 1998. A Radiance Research M-903 nephelometer measures the total scattering coefficients at a wavelength of 530nm, using the geometry of a standard integrating nephelometer under ambient relative humidity (RH). With general meteorological values (i.e., pressure, temperature, and RH), the values of total scattering coefficient are monitored and recorded as 15 minute averages. Calibration is accomplished by adjusting the span so that the indicated scattering coefficient matches the calculated value when sampling calibration span gas at local pressure and temperature conditions. Particle free air and Freon gas (CHClF₂) are used as zero reference and span gas, respectively. The optical and electrical background noise (usually wall scatter) is sufficiently low to allow measurement of total scattering coefficient from less than $10^{-6} \text{ m}^{-1}$ to greater than $10^{-3} \text{ m}^{-1}$. 
3.4. Results and Discussions

3.4.1. Seasonal Patterns of BC Concentrations

To investigate the influence of long-range transport on measured BC mass concentrations, each BC sample was categorized by air mass type. The monthly average for each air mass type is shown in Figure 3.2. Except for May 1998, the highest average BC concentration values were found in polluted continental air masses and the lowest BC values were found in marine air masses. BC mass concentrations in summer 1998 agree well to those in summer 1996, 1997 [Bahrmann and Saxena, 1998]. The seasonal variation of BC mass concentrations was also investigated (Figure 3.3). During the summer season, the mean BC mass concentration is 327.4 ng m\(^{-3}\) for polluted continental air masses, 165.9 ng m\(^{-3}\) for continental air masses, and 103.3 ng m\(^{-3}\) for marine air masses. During the winter season, the mean BC mass concentration is 107.4 ng m\(^{-3}\) for polluted continental air masses, 86.7 ng m\(^{-3}\) for continental air masses, and 28.1 ng m\(^{-3}\) for marine air masses. In general, BC mass concentrations observed in other areas in winter are higher than those in summer, due to domestic and commercial heating in urban areas (e.g., 1.74 µg m\(^{-3}\) for winter compared to 0.54 µg m\(^{-3}\) for summer in Nova Scotia, Canada [Chýlek et al., 1999] and 2.0 µg m\(^{-3}\) for winter compared to 1.6 µg m\(^{-3}\) for summer over the Washington DC area [Malm et al., 1994]). However, rural regions such as our research site located at a higher altitude, are influenced by long-range transport of air pollutants, and show different seasonal patterns. One of main reasons for the seasonal difference is the enhanced thermal convection during the summer. This may increase transport of planetary boundary layer air pollutants from the surrounding urban area to
this rural area. Therefore, the mixing height in the site can be as high as or higher than 2006 m MSL during the summer months. On the other hand, we would expect lower mixing height in the winter. Hence, during the winter, the air on Mount Gibbes is cleaner, with an overall average value of 74.1 ng m$^{-3}$, whereas the overall summer mean value is higher by about a factor of 3. These results are in good agreement with the seasonal variations of daily median BC mass concentrations at a high-alpine research station during 1995 to 1997 [Lavanchy et al., 1999].

In the spring of 1998, abnormally high BC concentrations from the continental sector were measured (Figure 3.2). The monthly mean value of BC (375.0 ng m$^{-3}$) in May 1998 for continental air masses is even greater than that of polluted continental air masses (283.9 ng m$^{-3}$) and about twice as high as the concentrations in the continental air masses of May, 1997. A maximum daily average BC concentration is also shown in this continental air mass (May 16, 1998) with a value of 673 ng m$^{-3}$. This anomaly can be explained by a massive biomass burning in Central America and southern Mexico in the spring of 1998. The aerosols originating from the forest fire smoke plume in Mexico were transported to the southeastern US. The plume was also detected by the Earth Probe TOMS (Total Ozone Mapping Spectrometer) [NASA/GSFC, Laboratory for Atmospheres; http://toms.gsfc.nasa.gov/] and the most extensive smoke coverage at the research site occurred on May 16 (Figure 3.4). Therefore, these BC measurements obtained from continental sector air masses during May 1998 is evidence of the long-range transport of BC produced by biomass burning in Mexico.
3.4.2. Weekly Patterns of BC Concentration

To investigate day-of-week distribution of BC concentrations for the air masses transported from the polluted continental sector, 48-hr back-trajectories arriving at the research site at 1300 EST during June and July 1998 are drawn in Figure 3.5. The BC concentration is averaged for 3 Sundays, 2 Mondays, 1 Tuesday, 2 Wednesdays, 2 Thursdays, 2 Fridays, and 2 Saturdays displayed in Figure 3.5 to illustrate the day-of-week distribution (Figure 3.6). The mean BC concentration increases from Monday to Friday and decreases from Friday to Monday. The day-of-week distribution for the polluted continental air masses is also calculated for the entire summer period in order to generalize BC weekly patterns (Figure 3.7). This shows the mean BC concentration of the week along with 95% confidence intervals for the estimated mean values. As can be seen, the BC concentrations are low on Sunday to Tuesday with a minimum value of 256 ± 10 ng m\(^{-3}\) occurring on Monday and higher from Wednesday to Friday with a maximum value of 379 ± 13 ng m\(^{-3}\) occurring on Friday. Considering the transport time required from northern urban areas, the minimum occurring on Monday can represent lower BC emissions on Saturday and Sunday in the polluted source regions. The higher BC concentrations shown on Wednesday to Friday may be due to weekday pollution emissions in the source regions. The summer average of the BC concentration temporal (day-of-week) distribution measured at our site and that measured in southwestern Pennsylvania [Allen et al., 1999] are compared in Table 1. The major air pollution sources in the latter region were reported as the Hatsfield electric generating station, vehicular emissions, and open burning of residential and commercial trash allowed on
Saturdays and/or Wednesdays [Allen et al., 1999]. It is apparent that there is about a 1-day lag for peak and minimum BC concentrations at our site compared to one of BC emission source regions in the polluted sector. The overall average BC concentration at our site (322 ng m\(^{-3}\)) is reduced to about 25% of that in the southwestern PA (1270 ng m\(^{-3}\)). Although this comparison is insufficient to explain the exact mean time for transport to our site from southwestern PA, it can be used to approximate the time lag of the weekly BC concentration trend and BC amount transported from a source region.

3.4.3. Comparison with Scattering Measurements

Total aerosol scattering coefficient measurements at the site were compared with the BC absorption coefficient measurements. Scattering by aerosols is strongly dependent on the RH at which it is measured, due to the hygroscopic growth nature of most atmospheric aerosols [e.g., Tang and Munkelwitz, 1977; Malm et al., 1994; Hegg et al., 1996; Kotchenruther et al., 1999]. At our research site the maximum value of total scattering is observed just before sunrise and the minimum value is observed near 2 PM, which is coincident with the trend of RH. In order to eliminate the diurnal variation of total scattering, daily average values were calculated. Figure 3.8 illustrates daily means of total scattering and absorption coefficients from July 15 to August 13 1998. Although the magnitude of the absorption coefficients is less than that of total scattering coefficients at the site, the daily mean absorption coefficients correlate well (correlation coefficient, R=0.86) with the daily mean total scattering coefficients. This suggests that
the transported aerosols responsible for scattering and absorption have originated from the same emission sources.

### 3.4.4. BC Effects on Direct Radiative Climate Forcing

Numerous studies of the direct effect of tropospheric sulfate aerosols have shown an aerosol impact on climate through reflection of solar radiation back to space [e.g., Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Boucher and Anderson, 1995; Saxena and Menon, 1999]. Estimates of the globally averaged annual direct radiative forcing due to anthropogenic sulfates range from \(-0.3\) W m\(^{-2}\) [Kiehl and Briegleb, 1993] to \(-1.3\) W m\(^{-2}\) [Charlson et al., 1992]. The direct cooling effects of aerosols are reduced by the presence of BC [e.g., Chýlek and Wong, 1995; Chýlek et al., 1995]. The BC within the sulfate aerosol reduces the expected sulfate direct cooling effect by about 0.034 W m\(^{-2}\) for each 1\% of the BC to sulfate mass mixing ratio [Chýlek et al., 1995]. Haywood and Shine [1995] reported that fossil fuel derived BC caused positive global mean radiative forcing ranged from +0.03 to +0.24 W m\(^{-2}\) for an assumed BC/sulfate mass ratio of between 0.05 and 0.1.

With direct measurements of optical properties for aerosols in the southeastern US, aerosol direct radiative forcing is calculated using the equation obtained by Chýlek and Wong [1995]:

\[
\Delta F_R = -\frac{S_0}{4} \tau_{atm}^2 (1 - A_c) \left[ (1 - R_s)^2 2B\tau_{sc} - 4R_s\tau_{abs} \right]
\]  (1)
where \( S_0 \) is the solar constant, \( T_{\text{atm}} \) is the atmospheric transmission, \( A_c \) is the fractional cloud amount, \( R_s \) is the surface albedo of the underlying surface, \( B \) is the fraction of average daily solar radiation scattered back to space (i.e., upscatter fraction computed from backscatter fraction), and \( \tau_{sc} \) and \( \tau_{abs} \) are the optical depth of the aerosol layer due to light scattering and absorption, respectively.

To calculate \( \tau_{sc} \) and \( \tau_{abs} \) the following equations were used:

\[
\tau_{sc} = \int_{Z_1}^{Z_2} \sigma_{sp} \, dz \tag{2}
\]

\[
\tau_{abs} = \int_{Z_1}^{Z_2} \sigma_{ap} \, dz \tag{3}
\]

where \( \sigma_{sp} \) and \( \sigma_{ap} \) are the total scattering coefficient and absorption coefficient, respectively. To compare contributions of aerosol scattering and absorption to regional climate at a certain height, the values of \( \tau_{sc} \) and \( \tau_{abs} \) were calculated for unit vertical depth at 2006 m MSL. Parameters used to calculate the direct radiative forcing in this study were \( S_0 = 1370 \text{ Wm}^{-2} \), \( T_{\text{atm}} = 0.76 \), \( A_c = 0.61 \), and \( R_s = 0.15 \) based on the values used in the box model \cite{Charlson et al., 1992}. In addition, the value of \( B \) used in the computation of \( \Delta F_R \) was estimated using the regression equation of backscatter fraction \( (\beta = -0.0005 \times \text{RH} + 0.125) \) obtained from three-wavelength nephelometer at a nearby research station (35.66 N, 82.38 W, 951 m MSL) and Henyey-Greenstein phase function \cite{Wiscombe and Grams, 1976} to be 0.23 at \( \text{RH}=68\% \). This RH is the mean ambient RH during the measurement and is consistent with the clear sky daytime RH (68.6\%) in the eastern United States during July, obtained by \textit{Boucher and Anderson} \cite{1995}. Direct radiative
forcing per unit vertical depth ($W m^{-3}$) was computed for aerosol scattering and absorption at 2006 m MSL of the southeastern US during July 15 – August 13, 1998. The net aerosol direct radiative forcing (scattering effects plus absorption effects) per unit vertical depth was calculated to have a negative value of $-1.38 \times 10^{-3} W m^{-3}$. The magnitude of direct radiative forcing by aerosol scattering is reduced by $15 \pm 7\%$ due to the BC absorption. Thus, the presence of BC does not significantly change the estimates of aerosol direct cooling effect in the southeastern US. A regional cooling trend in the mean maximum surface temperature over the southeastern US from 1949 to 1994 [Saxena and Yu, 1998] is potentially an observational confirmation for the estimate.

*Haywood and Shine* [1995] expressed the aerosol forcing as a function of optical depth:

$$\Delta F = -DS_o T_{atm}^2 (1 - A_c) \omega_o B \delta \left[ (1 - R_s)^2 - \frac{2R_s}{B} \left( \frac{1}{\omega_o} - 1 \right) \right]$$  \hspace{1cm} (4)$$

where $\delta$ is the optical depth, $D$ is the fractional daylength (the value of $D = 0.5$ is used in this study), and $\omega_o$ is single scatter albedo (the ratio of scattering to absorption plus scattering for a thin aerosol layer). The parameters ($S_o$, $T_{atm}$, $A_c$, $B$, and $R_s$) are the same as those defined in equation (1). Rearranging the equation (4), the aerosol radiative forcing per unit optical depth (aerosol radiative forcing efficiency) is obtained as follows:
\[
\frac{\Delta F}{\delta} = -D S_o T_{\text{atm}}^2 (1 - A_e) \omega_o B \left[ (1 - R_s)^2 - \frac{2R_s}{B} \left( \frac{1}{\omega_o} - 1 \right) \right]
\]  

(5)

The direct radiative forcing per unit optical depth are calculated as a function of single scatter albedo during July 15 - August 13, 1998. In this computation, the value of 0.23 for upscatter fraction is used as mentioned previously. At our experimental site, 93% of the calculated single scatter albedo are in the range of 0.90 to 0.96 and the overall average direct radiative forcing per unit optical depth is calculated to be \(-20 \pm 2\) \(\text{W m}^{-2}\). The magnitude of this cooling estimate is smaller than that for dry aerosols in the range of \(-23\) to \(-33\) \(\text{W m}^{-2}\) over central and eastern North America [Sheridan and Ogren, 1999]. It is partially due to aerosol particle size growth with RH in our case considering the hygroscopic growth factor. That is, the smaller upscatter fraction value used in our study is responsible for this difference. Neglecting BC absorption effects (i.e., single scatter albedo = 1), the direct radiative forcing per unit optical depth is derived to be \(-25.6\) \(\text{W m}^{-2}\) which approaches to the value of \(-32\) \(\text{W m}^{-2}\) obtained by Charlson et al. [1991] when the value of 0.29 for upscatter fraction is used. In our study considering the effects of BC absorption and aerosol hygroscopic growth nature, the BC reduces the expected sulfate direct cooling effect per unit optical depth by 21\(\pm\)9 \%. 
3.5. Summary and Conclusion

The BC mass concentrations in the southeastern United States were measured at a regionally representative site near Mount Mitchell, North Carolina, the highest peak in eastern North America. The results obtained are the following:

(1) During the experiment, except for May 1998, the highest average BC concentration is seen in polluted continental air masses and the lowest in marine air masses.

(2) During the winter the air is cleaner with an overall average value of 74.1 ng m$^{-3}$, whereas the overall summer mean value is higher by about a factor of 3. The main reason for the seasonal difference may be enhanced thermal convection during summer, which increases transport of planetary boundary layer air pollutants from the surrounding urban area to this rural site.

(3) In the spring of 1998, abnormally high BC concentrations from the continental sector were measured. These concentrations were originating from biomass burning from the forest fire smoke plume in Mexico. This was confirmed by the observations of the Earth Probe TOMS.

(4) The mean BC concentrations of air masses transported from the polluted continental sector during summer are low from Sunday to Tuesday with a minimum value of 256 ng m$^{-3}$ on Monday and higher from Wednesday to Friday with a maximum value of 379 ng m$^{-3}$ on Friday.

(5) The daily mean BC absorption and scattering coefficients are well correlated (R=0.86), which suggests the aerosols originated from the same emission sources.
The net aerosol radiative forcing (scattering effects plus absorption effects) per unit vertical depth at 2006 m MSL was calculated to be $-1.38 \times 10^{-3}$ W m$^{-3}$ for the southeastern United States. The magnitude of direct radiative forcing by aerosol scattering is reduced by $15 \pm 7\%$ due to the BC absorption.

The effect of BC on climate forcing is important because of its high absorption of solar radiation, but the effect is estimated with a large uncertainty due to a lack of sufficient observational data. Therefore, the investigation of the seasonal variations and long-term trends in BC aerosol parameters as well as the indication of possible source area is important. The results obtained from continuous in situ measurements of BC aid in understanding the meteorological transport of atmospheric aerosol and improve the knowledge of the climatology due to aerosol BC in the southeastern United States.

**Acknowledgments**

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3.6. References


Table 3.1. Comparison of temporal (day-of-week) distribution of black carbon concentrations at Mount Gibbes, NC, to those obtained in southwestern PA. The numbers represent mean ± standard deviation.

| Day of week | Polluted air masses arriving at Mt. Gibbes, NC  
summer of 1998  | BC concentration (ng m$^{-3}$) | Air masses in southwestern PA  
summer of 1990 (Allen et al., 1999)  | BC concentration (ng m$^{-3}$) |
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<tr>
<td>Sunday</td>
<td>278 ± 107</td>
<td>647 ± 338</td>
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<tr>
<td>Monday</td>
<td>256 ± 108</td>
<td>1016 ± 271</td>
<td></td>
</tr>
<tr>
<td>Tuesday</td>
<td>290 ± 109</td>
<td>1294 ± 453</td>
<td></td>
</tr>
<tr>
<td>Wednesday</td>
<td>352 ± 130</td>
<td>1490 ± 600</td>
<td></td>
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<tr>
<td>Thursday</td>
<td>372 ± 157</td>
<td>1612 ± 486</td>
<td></td>
</tr>
<tr>
<td>Friday</td>
<td>379 ± 161</td>
<td>1409 ± 561</td>
<td></td>
</tr>
<tr>
<td>Saturday</td>
<td>324 ± 149</td>
<td>1421 ± 969</td>
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Figure 3.1. Map of the United States illustrating the geographical location and classification of the different air mass types influencing the research sites.
Figure 3.2. Average monthly black carbon concentrations for each sector at Mount Gibbes, NC.
Figure 3.3. Average black carbon concentrations for each sector during the summer months (June, July, and August in 1998) and during the winter months (November and December in 1997 and December in 1998).
Figure 3.4. Earth Probe TOMS aerosol index maps for May 15 (top), May 16 (middle), and May 17 (bottom) 1998.
Figure 3.5. Forty-eight-hour back-trajectories for polluted continental air mass cases arriving at the research site at 1300 EST during June and July 1998.
Figure 3.6. Temporal (day-of-week) distribution of black carbon concentrations for the trajectories displayed in Figure 3.5. Each point represents the average concentration for 1000 EST to 1600 EST during the days indicated.
Figure 3.7. Temporal (day-of-week) distribution of black carbon concentrations averaged for polluted air masses during the summer period (June, July, and August, 1998). The line with circles denotes the mean value and dashed lines denote 95 % confidence interval (CI) for the mean estimate.
Figure 3.8. Daily means of absorption coefficient and scattering coefficient measured from July 15 to August 13, 1998. P, C, and M denote polluted continental, continental, and marine air mass influences for specific times, respectively, based on 48 hr back trajectory analyses that were available during the period.
4. Spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes:

Impact of polar stratospheric clouds

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4.1. Abstract.

SAGE II ozone and aerosol measurements and NCEP/NCAR potential vorticity and temperature fields during 1985–1999 are analyzed to study the spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes and to investigate the impact of polar stratospheric clouds (PSCs) on aerosol distribution and ozone depletion. During austral spring (Sep/Oct) there exist strong radial gradients in ozone, aerosol, and temperature near the polar vortex edge region, and an isolation of materials inside polar vortex, confirming that springtime polar vortex acts effectively in keeping out intrusions of materials from the exterior of the polar vortex. The analysis of vertical profiles of 1020-nm aerosol extinction and 525- to 1020-nm aerosol extinction ratio inside the spring polar vortex indicates aerosol enhancement in 9–15 km layer and lack of larger particles in 15–20 km layer, which is consistent with the effects of gravitational sedimentation and subsequent evaporation of PSCs. For the spherical PSC particles for which Reynolds number < 1, gravitational sedimentation velocity is estimated to be 0.11 km/wk, 0.04 km/wk, and 1.5 km/wk for Type Ia, Ib, II PSCs, respectively. The probabilities of the occurrence of T<195 K (threshold temperature of Type Ia PSC formation), T<192 K (Type Ib), and T<190 K (Type II) at 50 mb pressure height are calculated in the region of 60–75 °S during austral winter (Jul/Aug). The probability averaged for the 15 year period is 0.62 ± 0.05 for T<195 K, 0.46 ± 0.06 for T<192 K, and 0.33 ± 0.06 for T<190 K. The probabilities of occurrence for the three categories of temperature show increasing trends with time, whereas springtime stratospheric column ozone amount shows decreasing trend. During volcanically
unperturbed years, the stratospheric column ozone amount in the spring is highly correlated with the probability of occurrence of temperature below 195 K in the previous winter ($R=−0.88$). The empirical relationship between the stratospheric column ozone amount ($O_3$) in the spring (Sep/Oct) and the probability ($P$) of occurrence of $T<195$ K in the previous winter (Jul/Aug) is found to fit the equation of $O_3 = -360.38 \times P + 446.38$ ($R^2=0.78$), which is a useful equation to predict ozone depletion from the previous winter temperature. In addition, the most severe ozone depletion observed in the late 1990s is consistent with the occurrence of the strong PV, and the empirical relationship between the $O_3$ and the PV strength ($|PV|$) during the spring is also found to fit the equation of $O_3 = -6.46 \times 10^6 |PV| + 651.92$ ($R^2=0.93$). These linear regression equations indicate that springtime ozone depletion is strongly correlated with the PV strength and the previous winter temperature for the formation of PSCs.
4.2. Introduction

The discovery of the Antarctic ozone hole in the middle 1980s [Farman et al., 1985] stimulated great interests in the photochemistry and dynamics of the southern winter and spring stratosphere. It is now generally accepted that heterogeneous chemical reactions on the surface of polar stratospheric clouds (PSCs) are mainly responsible for springtime Antarctic ozone destruction [e.g., Solomon et al., 1986; Solomon, 1990], whereas polar vortex dynamics creates and maintains the unique environment suitable for the necessary chemical reactions to take place [e.g., Schoeberl and Hartmann, 1991]. In order for widespread ozone destruction to occur during Antarctic spring, the air temperature within the polar vortex must be lower than 195 K for a sufficiently long period so that the PSCs can affect the conversion of chlorine from the inactive reservoir species to the radical species that attack ozone [Schoeberl and Hartmann, 1991]. In addition to the production of active chlorine species by heterogeneous reactions on the PSC particles, another importance of the PSCs is the sedimentation processes that irreversibly remove HNO₃ (denitrification) and water (dehydration) from the lower stratosphere. Denitrification slows the return of chlorine to its inactive forms and hence, enhances ozone destruction in spring season [Toon et al., 1986; Fahey et al., 1990; Salawitch et al., 1993; Portmann et al., 1996]. However, many questions remain about PSCs because of difficulty of sampling.

The stratospheric aerosols take up nitric acid and water at cold temperature to form PSCs [Carslaw et al., 1997]. Because of the role aerosols play in PSC formation and thus, indirectly, in heterogeneous chemical process that promotes the destruction of
ozone [Poole and McCormick, 1988; Solomon, 1990], polar stratospheric aerosols are of interest. Aerosol physical properties measured by means of remote sensing may be good tracers of dynamical, chemical and physical processes in the lower part of the middle atmosphere. Since PSCs form only under extremely cold conditions, they are observed occasionally in the winter polar vortex region of the Arctic and Antarctic stratosphere [McCormick et al., 1982; Poole and Pitts, 1994]. The maintenance of such a cold polar vortex requires a very weak transport of heat and conservative trace species from the midlatitudes into the inner vortex. However, observational studies did not lead to agreement on the horizontal mixing of the air across the edge of the polar vortex. From analysis of the data from the Airborne Antarctic Ozone Experiment, Hartmann et al. [1989] and Schoeberl et al. [1989, 1992] have concluded that the interior of the Antarctic polar vortex is isolated from the exterior of the vortex and consequently, the vortex acts as a chemical containment vessel. On the other hand, Proffitt et al. [1989] has argued that air inside the polar vortex is mixed continuously with air outside the polar vortex and thus, the vortex acts as a flowing processor. Aerosols in the Antarctic stratosphere can be used as tracers of the circulation within and across the polar vortex as well as tracers of sedimentation processes of PSCs. Therefore, an analysis of temporal and spatial distributions of aerosol and ozone associated with the Antarctic polar vortex processes could aid in answering the question: “How do stratospheric trace constituents respond to polar vortex processes and influence the ozone depletion?”.

Since PSCs form under extremely cold conditions [e.g., McCormick et al., 1982; Pole and McCormick, 1988; Browell et al., 1990; Poole and Pitts, 1994] and the maintenance
of such a cold polar vortex requires a high degree of isolation inside the polar vortex [e.g., Juckes and McIntyre, 1987; Schoeberl and Hartmann, 1991; Bowman, 1993; Norton, 1994], the temperature and potential vorticity of the lower stratosphere during winter and spring are the key factors in explaining the change of the magnitude of springtime Antarctic ozone depletion. However, no studies about how frequently the stratospheric temperature falls below threshold temperature for the formation of PSCs during winter have been reported so far. Therefore, it is necessary to investigate the probability of occurrence of temperature below threshold temperature for PSC formation in the stratosphere during winter. Furthermore, the quantitative relationships between temperature, potential vorticity, and ozone are needed to estimate the lower stratospheric ozone depletion associated with dynamical and physical processes in the Antarctic. In this paper we analyze Stratospheric Aerosol and Gas Experiment II (SAGE II) ozone and aerosol measurements and the National Centers for Environmental Prediction and the National Center for Atmospheric Research (NCEP/NCAR) reanalysis data of potential vorticity (PV) and temperature, (1) to study the spatial and temporal distributions of aerosol and ozone associated with the polar vortex processes over the southern hemisphere, (2) to investigate the impact of PSCs on aerosol distribution and ozone depletion, (3) to estimate gravitational sedimentation velocity of PSCs using Stokes’ equation, (4) to investigate the probabilities of occurrence of temperature below threshold for the formation of Type Ia, Type Ib, and Type II PSCs during the winter in the Antarctic lower stratosphere, and (5) to eventually derive empirical relationships between ozone, temperature, and PV.
4.3. Data Sets

4.3.1. SAGE II Data

The Stratospheric Aerosol and Gas Experiment II (SAGE II) was launched aboard the Earth Radiation Budget Satellite (ERBS) in October 1984. The instrument continues to provide high quality measurements of ozone, nitrogen dioxide, water vapor, and multi-wavelength (386, 452, 525, and 1020 nm) aerosol extinction from the mid-troposphere to as high as the lower mesosphere. The SAGE II instrument is a self-calibrating, limb-scanning sun photometer on board the ERBS that is a free-flying satellite with a 610-km circular orbit and a 57° angle of inclination. The SAGE II instrument contains 7 spectral channels centered at 1020, 935, 600, 525, 452, 448, and 386 nm wavelengths, which is designed to measure the intensity of solar radiation attenuated by atmospheric constituents in the earth’s atmospheric limb during each sunrise and sunset event encountered by the spacecraft as it orbits the earth. Such solar occultation technique used by the SAGE II provides 15 sunrise and 15 sunset measurements each day. Over a year the SAGE II instrument covers from 75 °S to 75 °N. More detailed information on the SAGE II program, instrument, and inversion algorithm can be found in McCormick [1987], Mauldin et al. [1985], and Chu et al. [1989], respectively.

Recently NASA Langley Research Center’s SAGE II Algorithm Team has completed a major revision of the processing algorithms for SAGE II and released Version 6.0. The fundamental changes to the algorithms occur in the Level 1 transmission algorithm and ultimately yield substantial improvement in altitude registration and scan co-registration. These improvements permit the change from 1-km to 0.5-km altitude resolution in the
species profiles. The Level 2 algorithm takes the transmission profiles and inverts them for profiles of the number density of ozone, nitrogen dioxide, and water vapor and aerosol extinction profiles at 4 wavelengths. Version 6.0 changes to the Level 2 algorithm are refinements of the changes included in the 1998 Stratospheric Processes And their Role in Climate (SPARC) Ozone Trend Assessment Version (5.96). These changes include the reduction of the interference of enhanced aerosol on the ozone measurements and improvements in the quality of reported species uncertainties. In this study we use the updated SAGE II Version 6.0 ozone and aerosol data covering the last decade from 1985 to 1999. Following Kent et al. [1993], extinction measurements in which only aerosols occur along the optical path have been obtained by examining the wavelength dependence of the extinction. Based on scattering calculations carried out using the Mie approximation, Kent et al. [1993] have shown that the ratio of the aerosol extinction at the SAGE II wavelengths 525 nm and 1020 nm is normally between 2 and 5, whereas scattering from cloud particles (having radius in the range from 1 to 5000 µm [Heymsfield and Platt, 1984]) has little wavelength variation. When 525- to 1020-nm aerosol extinction ratio is less than 2 at any height, it has been considered as a cloud contamination and the profile has been eliminated in the analysis.

### 4.3.2. NCEP/NCAR Reanalysis Data

This data set contains meteorological products from the National Centers for Environmental Prediction and the National Center for Atmospheric Research (NCEP/NCAR) reanalysis project [Kalnay et al., 1996]. The NCEP/NCAR reanalysis is
a continuous, consistent assimilation system and uses the NCEP spectral model and satellite temperature retrievals. The output products include over 80 different variables and 3 different coordinate systems. In this study we use temperature and Ertel’s potential vorticity (PV) from 1985 to 1999. The temperature data are available on pressure levels from 1000 to 10 hPa (17 levels) and a 2.5° longitude by 2.5° latitude horizontal grid. The PV data are available on isentropic surfaces from 270 to 650 K (11 levels) and a 2.5° longitude by 2.5° latitude horizontal grid. The PV area diagnostic method developed by Butchart and Remsberg [1986] has been used in many studies [e.g., Baldwin and Holton, 1988; Manney et al., 1994] for quantifying polar vortex variation. In the area diagnostic method, PV is considered as a tracer and the polar vortex is defined by sharp PV gradient on an isentropic surface. In this study the vortex boundary is defined by the contour with the greatest PV gradient on an isentropic surface. Figure 1 displays 15-year (1985–1999) mean PV distributions for winter (July/August) and spring (September/October), on the 500 K isentropic surface (approximately 20-km altitude) in the southern hemisphere. As seen in Figure 1, during both winter and spring, there exists a strong radial gradient of PV in the regions of PV unit of 4–7 (near 60 °S latitude circle) surrounding the Antarctic polar vortex, while a region with very weak gradient of PV is shown in the midlatitudes. This feature in the PV field was very common in the Antarctic stratosphere during the winter and spring for the period of 1985–1999. The strongest radial gradient is found near the 6 PV unit contour, which can be considered as polar vortex boundary and completely circumnavigates the continent of Antarctica including the end of the Antarctic
Peninsula. This structure was shown every year during this period of 1985–1999 and the high PV inside the spring polar vortex evolved more strongly in the late 1990s.

4.4. Results and Discussions

4.4.1. Spatial and Temporal Distributions of Aerosol and Ozone

Plate 1 and Plate 2 display cloud-free SAGE II multi-year horizontal distributions of 1020-nm aerosol extinction coefficients for summer (January/February) and spring (September/October), respectively, observed at 20 km altitude during the period of 1985–1999. For the spring the PV distributions are also shown, superimposed on the aerosol distributions. For this analysis period there were two huge volcanic eruptions at El Chichon (17.3 °S, 93.2 °W) in April 1982 and at Mt. Pinatubo (15.1 °N, 120.4 °E) in June 1991, which significantly perturbed the stratospheric aerosol level. In some area the Mt. Pinatubo plume was sufficiently opaque that the solar intensity reaching the SAGE II instrument was zero and thus the layers below the height of largest measurable extinction were truncated in measurement. Due to the frequent truncation of the layer below 20 km (indicating the most optically thick layers of the Mt. Pinatubo aerosol), the extinction measurement data at 20 km altitude was not available in the whole southern hemisphere during the summer of 1992 and in the lower latitudes during the summer of 1993 (Plate 1), and in the lower latitudes during the spring of 1991 and 1992 (Plate 2). In addition, it is not surprising to see that the extinction coefficients are unusually high during the summer of 1993 (Plate 1) and the spring in 1992 and 1993 (Plate 2). For the periods of 1985 – 1991 and 1994 – 1999, it is also shown that the stratospheric aerosol levels had
recovered toward background ones after the eruptions of El Chichon in April 1982 and Mt. Pinatubo in June 1991, respectively (Plate 1).

As seen in Plate 1, during the summer the horizontal distributions of stratospheric aerosols are roughly uniform in the south 60 °S. In contrast, springtime aerosol distributions (Plate 2) show strong radial gradients near the polar vortex edge region with very low and uniform levels of aerosol extinction inside the polar vortex. Consistent with Schoeberl et al.’s arguments [1989, 1992] and the results from modeling studies [e.g., Juckes and McIntyre, 1987; Bowman, 1993; Norton, 1994], Plate 2 indicates that the aerosols in the inner region of the polar vortex have been isolated in the absence of horizontal mixing processes across the boundary of the polar vortex. This suggests that the ozone loss chemical processes inside the polar vortex can proceed without replenishment by intrusions of ozone-rich air from midlatitudes. Inside the springtime polar vortex, indeed, the horizontal distributions of aerosol extinction in Plate 2 are very similar to those of springtime stratospheric column ozone in Plate 3. Plate 3 displays SAGE II multi-year horizontal distributions of stratospheric column ozone amount and the PV distributions superimposed on the column ozone, in the spring (Sep/Oct) during the 11-year period (i.e., volcanically unperturbed period of 1985–1990 and 1995–1999, excluding the period of the Mt. Pinatubo eruption influence). Since aerosol surface area density enhancements due to volcanic eruption may permit heterogeneous chemical reactions on the aerosols themselves and consequently enhance ozone depletion in the absence of PSCs [e.g., Brasseur et al., 1990; Solomon et al., 1993], in this study we consider only volcanically unperturbed years to emphasize the impacts of polar vortex
and PSCs on ozone depletion. Contrary to summer column ozone distributions (not shown) that indicate quite uniform distributions of ozone with no annual variation, springtime column ozone distributions (Plate 3) reveal the lowest amount of ozone inside the polar vortex and show significant interannual variation of ozone depletion with more severe depletion in the late 1990s. The horizontal distributions of stratospheric temperature during the spring (not shown) also show quite similar patterns to those of ozone, indicating that the vortex edge might block the heat transfer from midlatitudes so that the cold air temperature was maintained inside the polar vortex.

Figure 2 displays the vertical profiles of stratospheric ozone number concentration (cm\(^{-3}\)) averaged for spring (Sep/Oct) inside the polar vortex, spring (Sep/Oct) outside the polar vortex, summer (Jan/Feb), and fall (March), in the region of 60°S – 75°S during 1985–1990 and 1997–1999 (Data for the years of 1995 and 1996 was excluded in the analysis because of insufficient measurement coverage over the specified region.). Over the Antarctic region, SAGE II provides data for spring, summer, and fall (For winter the polar region falls into darkness after the autumnal equinox.). As shown in Figure 2, the profile outside the vortex for spring is quite similar to those for summer and fall. The maximum concentration occurs near 20-km altitude with magnitude of about 4.5 \times 10^{12} \text{ cm}^{-3}. As expected from the horizontal distributions of the stratospheric column ozone, Figure 2 indicates ozone depletion inside the polar vortex during the spring. The severe ozone depletion is observed in the layer of 12 to 23 km and the minimum concentration occurs in the layer of 15–16 km.
Since air parcels are relatively isolated inside the polar vortex (Plate 2), the vertical air motion inside the polar vortex is important for evaluating the evolution of the ozone hole. To estimate the vertical motion, vertical profiles of the aerosols from SAGE II are analyzed. We restrict our analysis to altitudes from 8 to 20 km because a relatively larger number of aerosol extinction measurements is available at this altitude range and the uncertainties of the measurements in the layer are relatively smaller [Chu et al., 1989; Wang et al., 1989]. Figure 3 illustrates the vertical profiles of 1020-nm aerosol extinction averaged for spring (Sep/Oct) inside the polar vortex, spring (Sep/Oct) outside the polar vortex, summer (Jan/Feb), and fall (March), in the region of 60 °S – 75 °S during 10-year period (1986–1990 and 1995–1999, i.e., volcanically unperturbed period). The 10-year mean vertical profiles of aerosol extinction exhibit two regions with different seasonal variations. In the layer of 15–20 km the extinction is the highest during the summer and fall, and the lowest inside the polar vortex during the spring. The seasonal variation of the aerosol in the layer of 9–15 km shows opposite trend from the 5-km layer above, with the highest inside the spring polar vortex and the lowest during the fall. In a relatively shallow layer of 8–9 km, aerosol enhancement is observed during the summer. A similar profile has been observed by the Stratospheric Aerosol Measurement (SAM) II satellite instrument [Wang et al., 2000]. The springtime vertical distributions of the aerosol inside the polar vortex are quite different from those outside the polar vortex and those of summer and fall. Specifically, the rapid decrease of the aerosol extinction in the layer between 15–20 km and an enhancement of the aerosols in the layer between 9–15 km are shown inside the polar vortex during the spring. The rapid decline in aerosol
extinction inside the springtime polar vortex above 15 km concomitant with the rapid enhancement of that below 15 km appears to be consistent with the effect of large scale subsidence air motion inside the polar vortex. The increase in aerosol optical depth for the layer between 9–15 km from fall to spring is calculated to be about $7.6 \times 10^{-4}$ inside the polar vortex. Assuming that downward transport of aerosol mass loading above 15 km causes the enhancement of aerosols in the layer between 9–15 km, the vertical descent rate inside the vortex is estimated to be about 1 km/month from March to Sep/Oct. Using vortex interior descent model, Rosenfield et al. [1994] reported that air parcels initialized at 18 km on March 1, descended 3 km by the end of October. This is equivalent to a descent rate of 0.38 km/month during 8-month period, which is much lower than our estimate. Therefore, further study of additional mechanism is needed to explain the observed springtime vertical profile of aerosol extinction inside the polar vortex.

4.4.2. Gravitational Sedimentation of PSCs

The aerosol extinction ratio of 525- to 1020-nm is a good measure of the particle size because of wavelength dependence of light extinction. It can be explained using Angstrom equation: $Ext_\lambda = c\lambda^{-\alpha}$, where $Ext_\lambda$ is the extinction coefficient at wavelength $\lambda$, $c$ is a constant, and $\alpha$ is the Angstrom coefficient. Typical values of $\alpha$ are nearly 4 for gases, on the order of 2 for aerosols, and close to 0 for very coarse aerosols or clouds. For large values of $\alpha$, therefore, there will be a large difference in extinction at different wavelengths, while for small values of $\alpha$ (for example near 0) extinction at different
wavelengths are nearly equal. Hence, as the particle size is smaller, the 525- to 1020-nm aerosol extinction ratio becomes larger. Figure 4 displays the volcanically unperturbed 10-year mean vertical profiles of 525- to 1020-nm aerosol extinction ratio inside and outside the polar vortex during the spring. As seen in Figure 4, in the layer of 15–20 km the extinction ratio inside the vortex is significantly greater than that outside the vortex in a statistical approach. This indicates that inside the springtime polar vortex, most of the aerosols in the layer of 15–20 km are in the fine particle size mode, which implies the lack of the larger particles in the layer. Since this layer corresponds closely to the layer of 16 to 22 km in which PSCs most frequently occur during winter [Poole and Pitts, 1994; Vömel et al., 1995], the lack of the larger particles in this layer during the spring may be due to irreversible loss of the larger particles through the gravitational sedimentation of PSCs from winter to spring. This feature is consistent with the result of Thomason and Poole [1993]. This process is schematically shown in Figure 5. The lack of larger particles in 15–20 km layer and the enhancement of aerosols in 9–15 km layer observed during the spring are consistent with the effect of gravitational sedimentation and subsequent evaporation of PSCs. As larger PSC particles (which were generated during the previous winter) sediment to the lower and warmer layer, they evaporate [Vömel et al., 1995] and as a result, lead to enhancement of aerosols in the layer between 9–15 km inside the polar vortex during the spring. The analysis of the vertical profiles of 1020-nm aerosol extinction and 525- to 1020-nm aerosol extinction ratio inside the springtime polar vortex indicates a preferential loss of larger aerosols that served as PSC nuclei in the layer above 15 km [Thomason and Poole, 1993], suggesting aerosol
redistribution through the gravitational sedimentation and evaporation of PSCs together with the subsidence air motion, and the consequent enhancement of aerosols in the upper troposphere and lower stratosphere (i.e., 9–15 km) inside the polar vortex during the spring.

For spherical PSC particles for which the Reynolds number < 1, the gravitational sedimentation velocity can be estimated using the Stokes’ equation [Arya, 1999]:

\[
V_g = \frac{1}{18} \frac{gd_p^2 \rho_p}{\mu}
\]

where \( V_g \) is the gravitational sedimentation velocity, \( g \) is the gravity, \( d_p \) is the diameter of spherical particle, \( \rho_p \) is the density of particle, and \( \mu \) is the dynamic viscosity of air. This equation is valid for particles of diameter smaller than about 20 \( \mu m \). According to the size, composition, and formation temperature, PSCs have been classified into Type Ia, Type Ib, and Type II [see Crutzen and Arnold, 1986; Toon et al., 1986, 1990; Hanson and Mauersberger, 1988; Poole and McCormick, 1988; Browell et al., 1990; Carslaw et al., 1994, 1997; Tabazadeh et al., 1994; Larsen et al., 1997]. It is generally assumed that Type Ia PSCs consist of crystals of nitric acid trihydrate (NAT), Type Ib PSCs are droplets of supercooled ternary solution, and Type II PSCs are water ice. In spite of disregard of atmospheric and cloud microphysical processes, the empirical classification of PSCs allows the assignment of the majority of observed PSCs to one of the three categories Ia, Ib, and II. To estimate \( V_g \) of the three types of PSC particles, we used \( \rho_p = \)
1125 kg/m³ for Type Ia PSCs (HNO₃ · 3H₂O clouds), ρₚ = 1510 kg/m³ for Type Ib PSCs (HNO₃-H₂SO₄-H₂O clouds), and ρₚ = 920 kg/m³ for Type II (H₂O ice clouds). Using the density values of HNO₃, H₂SO₄, and H₂O [Schafer, 1977; Rosen, 1971; Kundu, 1990], the ρₚ for Type Ia and Type Ib PSCs were calculated as weighted averages of the separate molecule’s densities (HNO₃ and H₂O for Type Ia and HNO₃, H₂SO₄, and H₂O for Type Ib). The dynamic viscosity of air, μ = 1.3 kg m⁻¹ s⁻¹ at 190 K [Rogers and Yau, 1989; Houghton, 1985] was used in this computation. Figure 6 shows the gravitational sedimentation velocity as a function of the particle diameter for the three categories of PSCs. For Type Ia PSCs the V₉ is estimated to be 0.0003 to 0.46 km/week in diameter range of 0.1 – 4.0 µm. The V₉ of Type Ib PSCs is in the range of 0.0004 to 0.61 km/week for diameter between 0.1 – 4.0 µm and Type II PSC’ V₉ is in the range of 0.02 – 9.3 km/week for diameter between 1.0 – 20 µm. As shown in Figure 6, the V₉ is estimated to be 0.11 km/week for the mean diameter of 2 µm [Toon et al., 1990] in Type Ia PSCs, 0.04 km/week for the mean diameter of 1 µm [Toon et al., 1990] in Type Ib PSCs, and 1.5 km/week for the mean diameter of 8 µm [Poole and McCormick, 1988; Vömel et al., 1995] in Type II PSCs.

4.4.3. Probability of Temperature for PSC Formation

To investigate the maximum possibilities of the formation of various types of PSCs in the Antarctic stratosphere during winter (July/August), the probabilities of occurrence of temperature below threshold for the formation of Type Ia, Type Ib, and Type II PSCs are calculated using NCEP/NCAR reanalysis daily (at 12Z) data at 50-mb pressure level...
We used three threshold temperatures to characterize the composition of the three types of PSCs, i.e., 195 K for Type Ia PSCs (NAT; HNO₃ · 3H₂O) [e.g., Hanson and Mauersberger, 1988; Larsen et al., 1997; Stein et al., 1999], 192 K for Type Ib PSCs (liquid ternary droplet; HNO₃-H₂SO₄-H₂O) [e.g., Carslaw et al., 1994, 1997; Drdla et al., 1994; Tabazadeh et al., 1994; Larsen et al., 1997; Stein et al., 1999], and 190 K for Type II PSCs (ice particle; H₂O) [e.g., Poole and McCormick, 1988; Browell et al, 1990]. Figure 7 displays the probability of occurrence of T < 195 K, T < 192 K, and T < 190 K in the region of 60 °S – 75 °S from 1985 to 1999. The total number of grid data points used in the computation of the probability for each year is 62496 (62 days × 144 points in longitude × 7 points in latitude = 62496 points). Since the formation of PSCs depends on the amount of constituent (e.g., H₂O, H₂SO₄, HNO₃, etc.) and the duration of the cold temperature in the stratosphere [Tabazadeh et al., 1996; Larsen et al., 1997], the probabilities of occurrence of temperature below threshold presented in this study imply the maximum possibilities of the formation of various types of PSCs. The 15-year mean probability of occurrence of temperature below threshold for the formation of PSCs is calculated to be 0.62 ± 0.05 for T < 195 K, 0.46 ± 0.06 for T < 192 K, and 0.33 ± 0.06 for T < 190 K. Figure 7 shows that probabilities of occurrence of the three categories of temperature increase with time. The increasing rate is calculated to be about 0.0074/year for T < 195 K, about 0.0095/year for T < 192 K, and about 0.0105/year for T < 190 K (The most rapid increase is found in the temperature category of T < 190 K.) Using the Statistical Analysis System (SAS), P-value for the changing trend is calculated. The changing trends in the probabilities of occurrence for T < 195 K,
T < 192 K, and T < 190 K are significant at the 98.3%, 99.7%, and 99.9% confidence level, respectively.

While the wintertime probabilities of occurrence of the three categories of temperature show increasing trend with time from 1985 to 1999, the springtime stratospheric column ozone amount shows decreasing trend during the volcanically unperturbed years. Figure 8 displays the stratospheric column ozone amount (DU) during the spring (Sep/Oct) and the probability of occurrence of temperature below 195 K at the 50 mb pressure height during the previous winter (Jul/Aug), in the region of 60°S − 75°S. As seen in Figure 8, the stratospheric column ozone amount in the spring is highly correlated with the probability of occurrence of temperature below 195 K in the previous winter, during the volcanically unperturbed years (R = −0.88).

4.4.4. Empirical Relationships between Ozone, Temperature, and PV

Based on the negative correlation shown in Figure 8, the empirical relationship between springtime ozone depletion and the maximum possibility of wintertime PSC formation is derived. Figure 9 displays the spring (Sep/Oct) stratospheric column ozone amount (O₃) versus the probability (P) of occurrence of temperature below 195 K at the 50 mb pressure height during the previous winter (Jul/Aug). As shown in Figure 9, the empirical relationship is found to fairly well fit the equation of $O₃ = -360.38 \times P + 446.38$ ($R^2=0.78$), where the unit of $O₃$ is DU. Approximately 78% of the total variation in the annual springtime stratospheric column ozone is explained by this linear regression on the probability of occurrence of temperature below 195 K. The P-value for the
estimate of slope is calculated to be 0.0016, so the null hypothesis (Ho: Slope=0) is rejected. A 95% confidence interval for the true slope of this linear regression equation extends from −531.12 to −189.63. The statistics from SAS for this linear regression model are summarized in Table 4.1. This linear regression quantitatively demonstrates that springtime ozone depletion is related with the previous winter temperature for the formation of PSCs. Furthermore, this equation can be used to predict springtime Antarctic ozone depletion from the previous winter temperature.

In addition, the most severe ozone depletion was observed in the late 1990s (as shown in Figure 8), which in turn was consistent with the occurrence of the strong PV during the spring. Figure 10 shows the spring (Sep/Oct) stratospheric column ozone amount plotted against the spring (Sep/Oct) PV on the 500 K isentropic surface. The empirical relationship between the stratospheric column ozone amount (O3) and the PV strength (|PV|) during the spring (Sep/Oct) is obtained to fit the equation of \[ O_3 = -6.46 \times 10^6 |PV| + 651.92 \] (R²=0.93), where the unit of O3 and |PV| is DU and m² s⁻¹ kg⁻¹ K, respectively. About 93% of the total variation in the annual springtime stratospheric column ozone is explained by this linear regression on |PV|, indicating a strong correlation between stratospheric column ozone amount and PV strength during the spring. The P-value for the estimate of slope is calculated to be < 0.0001, so the null hypothesis (Ho: Slope=0) is rejected. A 95% confidence interval for the true slope of this linear regression equation is calculated to be from −8.05 \times 10^6 to −4.87 \times 10^6. The statistics for this linear regression model are summarized in Table 4.2. This relationship provides strong evidence that springtime ozone depletion is related with springtime PV strength.
4.5. Summary

SAGE II ozone and aerosol measurements and NCEP/NCAR temperature and PV fields during the period of 1985–1999 are analyzed to study the spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes and to investigate the impact of PSCs on aerosol distribution and ozone depletion. The analysis results can be summarized as follows:

(1) The analysis of the austral spring (Sep/Oct) PV distributions on the 500 K isentropic surface indicates that the polar vortex completely circumnavigates the continent of Antarctica including the end of the Antarctic Peninsula every year and high PV inside the polar vortex evolves more strongly in the late 1990s.

(2) The horizontal distributions of stratospheric column ozone, aerosol extinction, and temperature are fairly uniform in the south 60 °S during the austral summer (Jan/Feb), whereas during the austral spring (Sep/Oct) there exist strong radial gradients in the vortex edge region and an isolation of materials inside the polar vortex. This confirms that the Antarctic polar vortex can act effectively in keeping out intrusions of materials from the exterior of the vortex.

(3) The vertical profiles of the Antarctic ozone number concentration reveal severe ozone depletions in the layer of 12–23 km inside the polar vortex during the spring with a minimum value in 15–16 km layer. The most severe ozone depletion is observed in the late 1990s during the volcanically unperturbed years and is consistent with the occurrence of the strong PV.
(4) The analysis of vertical profiles of 1020-nm aerosol extinction and 525- to 1020-nm aerosol extinction ratio inside the springtime polar vortex indicates aerosol enhancement in the upper troposphere and lower stratosphere (i.e., 9–15 km) and the lack of larger particles in 15–20 km layer, which is consistent with the effects of the gravitational sedimentation and subsequent evaporation of PSCs.

(5) For the spherical PSC particles for which the Reynolds number < 1, gravitational sedimentation velocity is estimated to be about 0.11, 0.04, and 1.5 km/week for Type Ia, Ib, II PSCs, respectively.

(6) The probability of the occurrence of T<195 K (threshold temperature of Type Ia PSC formation), T<192 K (Type Ib), and T<190 K (Type II) at 50 mb pressure height is calculated in the region of 60–75 °S during the austral winter (Jul/Aug). The probability averaged for the 15 year period is 0.62 ± 0.05 for T<195 K, 0.46 ± 0.06 for T<192 K, and 0.33 ± 0.06 for T<190 K. The probabilities of occurrence for the three categories of temperature show increasing trends with time, whereas springtime stratospheric column ozone amount shows decreasing trend.

(7) In volcanically unperturbed conditions, the stratospheric column ozone amount in the spring is highly correlated with the probability of occurrence of temperature below 195 K in the previous winter (R= −0.88). The empirical relationship between the stratospheric column ozone amount (O₃) in the spring (Sep/Oct) and the probability (P) of occurrence of T<195 K in the previous winter (Jul/Aug) is found to fit the equation of O₃ = −360.38 × P + 446.38 (R²=0.78), which can be used to predict ozone depletion from the previous winter temperature.
The empirical relationship between the \(O_3\) and the PV strength \(|PV|\) during the spring is found to fit the equation of \(O_3 = -6.46 \times 10^6 \, |PV| + 651.92\) \((R^2=0.93)\). About 93\% of the total variation in the annual springtime stratospheric column ozone is explained by this linear regression on \(|PV|\), indicating that the stratospheric column ozone amount in the Antarctic is strongly correlated with PV strength there during the spring.
Acknowledgments.

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4.6. References


M. Loewenstein, J. R. Podolske, S. E. Strahan, M. H. Proffitt, C. R. Webster, R. D.
Elkins, K. R. Chan, J. G. Anderson, Chemical loss of ozone in the Arctic polar vortex

Schafer, K. (Ed.), Landolt-Bornstein, Numerical Data and Functional Relationshils in

Schoeberl, M. R., and D. L. Hartmann, The dynamics of the stratospheric polar vortex

Schoeberl, M. R., L. R. Lait, P. A. Newman, R. L. Martin, M. H. Proffitt, D. L. Hartmann,
M. Loewenstein, J. Podolske, S. E. Strahan, J. Anderson, K. R. Chan, and B. Gary,
Reconstruction of the constituent distribution and trends in the Antarctic polar vortex

Schoeberl, M. R., L. R. Lait, P. A. Newman, and J. E. Rosenfield, The structure of the

Solomon, S., Process towards a quantitative understanding of Antarctic ozone depletion,

Solomon, S., R. R. Garcia, F. S. Rowland, and D. J. Wuebbles, On the depletion of

Solomon, S., R. W. Sanders, R. R. Garcia, and J. G. Keys, Enhanced chlorine dioxide and


Wang, P.-H., G. S. Kent, K. A. Powell, G. K. Yue, L. R. Poole, and M. P. McCormick, Properties of the SAM II Antarctic 1.0-μm extinction coefficients: Implications of

Table 4.1. Statistics for the linear regression of ozone on probability of temperature for PSC formation.

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<th>Source</th>
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| Variable | DF | Parameter Estimate | Standard Error | t for Ho: parameter=0 | Prob > |t| |
|----------|----|--------------------|----------------|-----------------------|--------|---|
| Intercept| 1  | 446.37973          | 44.26343       | 10.08                 | <.0001 |
| Slope    | 1  | -360.37694         | 72.19671       | -4.99                 | 0.0016 |

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<td>218.7270</td>
<td>-15.8751</td>
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Table 4.2. Statistics for the linear regression of ozone on PV.

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<th>Source</th>
<th>Degree of Freedom</th>
<th>Sum of Squares</th>
<th>Mean Square</th>
<th>F Value</th>
<th>Prob &gt; F</th>
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<td>6022.09217</td>
<td>6022.09217</td>
<td>92.26</td>
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<tr>
<td>Error</td>
<td>7</td>
<td>456.92719</td>
<td>65.27531</td>
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<tr>
<td>Corrected Total</td>
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<td>6479.01935</td>
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<td></td>
<td></td>
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</table>

Root Mean Square for Error 8.07931  
R-Square 0.9295

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<tr>
<td>Variable</td>
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<td>Intercept</td>
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<td>Slope</td>
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<th>Output Statistics</th>
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<td>9</td>
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</table>
Figure 4.1. Potential vorticity (PV) distribution averaged for winter (July/August) and spring (September/October), on the 500 K isentropic surface in the southern hemisphere during 1985 - 1999. 1 PV unit = -10^-5 m^2 s^-1 kg^-1 K. Latitude circles are denoted in 10 degree increments from 90 S outward to 30 S.
Figure 4.2. Seasonal variations of the vertical distributions of 9-year mean (1985-1990 and 1997-1999) stratospheric ozone number concentration in the region of 60 °S – 75 °S.
Figure 4.3. Seasonal variations of the vertical distributions of 10-year mean (1986-1990 and 1995-1999) aerosol extinction coefficient (at 1020 nm) in the region of 60 °S – 75 °S.
Figure 4.4. Vertical profiles of 525- to 1020-nm aerosol extinction ratio inside and outside the springtime polar vortex, averaged for 10-year period (1986-1990 and 1995-1999). The error bars denote standard error.
Figure 4.5. Schematic illustration of aerosol redistribution process through the gravitational sedimentation and subsequent evaporation of polar stratospheric clouds inside the polar vortex.
Figure 4.6. The gravitational sedimentation velocity as a function of the particle diameter for Type Ia PSCs, Type Ib PSCs, and Type II PSCs.
Figure 4.7. Probability of occurrence of $T < 195$ K, $T < 192$ K, and $T < 190$ K during winter (Jul/Aug) at the 50 mb pressure height in the region of $60^\circ$S – $75^\circ$S.
Figure 4.8. The stratospheric column ozone amount in the spring (Sep/Oct) and the probability of occurrence of temperature below 195 K at the 50 mb pressure height during the previous winter (Jul/Aug), in the region of 60 °S – 75 °S.


Figure 4.9. The spring (Sep/Oct) stratospheric column ozone amount plotted against the probability of occurrence of temperature below 195 K at the 50 mb pressure height during the previous winter (Jul/Aug), in the region of 60 °S – 75 °S. The individual years are indicated.

\[
O_3 = -360.38P + 446.38
\]

\[R^2 = 0.78\]
Figure 4.10. The spring (Sep/Oct) stratospheric column ozone amount plotted against the spring (Sep/Oct) potential vorticity on the 500 K isentropic surface, in the region of 60 °S – 75 °S. The individual years are indicated.
Plate 4.1. SAGE II multi-year horizontal distributions of 1020-nm aerosol extinction at 20-km altitude in Jan/Feb. Latitude circles are denoted in 10 degree increments from 90 S outward to 30 S.
Plate 4.2. SAGE II multi-year horizontal distributions of 1020-nm aerosol extinction at 20-km altitude in Sep/Oct (color dots) and NCEP/NCAR mean potential vorticity (PV) distributions on the 500 K isentropic surface for Sep/Oct. Contour interval is 3 PV units (1 PV unit = \(-10^{\text{-}5}\) m\(^2\) s\(^{-1}\) kg\(^{-1}\) K). Latitude circles are denoted in 10 degree increments from 90 S outward to 30 S.
Plate 4.3. SAGE II multi-year horizontal distributions of stratospheric column ozone amount in Sep/Oct (color dots) and NCEP/NCAR mean potential vorticity (PV) distributions on the 500 K isentropic surface for Sep/Oct. Contour interval is 3 PV units (1 PV unit = $10^{-5}$ m$^3$ s$^{-1}$ kg$^{-1}$ K). Latitude circles are denoted in 10 degree increments from 90 S outward to 30 S.
5. Conclusions and Directions for Future Work

5.1. Summary and Conclusions

Atmospheric aerosols play an important role not only in climate forcing but also in the depletion of the stratospheric ozone layer. Such effects have a potential impact on regional and global climate.

To reduce the uncertainties of current estimates of aerosol climate forcing, aerosol optical properties relevant to the computation of direct radiative forcing were measured at a regionally representative site near Mount Mitchell, North Carolina. On the basis of these measurements and model calculations, we have studied (1) the effects of relative humidity (RH) on aerosol optical properties and direct aerosol radiative forcing and (2) the influence of long-range transport on black carbon (BC) concentrations, its seasonal and weekly patterns, and the effects of BC on the regional climate of the southeastern US. The light scattering of aerosol is strongly dependent on RH at which it is measured, due to hygroscopic growth nature of most atmospheric aerosols. In this study, the hygroscopic growth factor (ratio of total scattering coefficient at RH=80% to that at RH=30%) was calculated to be almost constant value of 1.60 ± 0.01 for polluted, marine, and continental air masses. In addition, it was found that as the RH increased from 30% to 80%, the backscatter fraction decreased by 23%. On the basis of these measurements, direct radiative climate forcing ($\Delta F_R$) by aerosols in the lowest 1 km layer of the troposphere was estimated. The patterns of direct radiative climate forcing by aerosols
for various values of RH were similar for the three air masses, but the magnitudes of the forcing were larger for polluted air masses than for marine and continental air masses by a factor of nearly 2 due to higher sulfate concentration in polluted air masses. The average value of $\Delta F_R(\text{RH}=80\%) / \Delta F_R(\text{RH}=30\%)$ was calculated to be almost a constant value of $1.45 \pm 0.01$ for all three types of air masses. This implies little dependence of the forcing ratio on the air mass type. The averaged forcing for all the observed ambient RHs was $-2.9$ $\text{W m}^{-2}$ (the negative forcing of $-3.2$ $\text{W m}^{-2}$ by aerosol scattering plus the positive forcing of $+0.3$ $\text{W m}^{-2}$ by aerosol absorption) for polluted air masses, $-1.4$ $\text{W m}^{-2}$ ($-1.5$ plus $+0.1$) for marine air masses, and $-1.5$ $\text{W m}^{-2}$ ($-1.6$ plus $+0.1$) for continental air masses. The $\Delta F_R$ for polluted continental air masses was approximately twice that of marine and continental air masses. The BC mass concentration of the southeastern US showed the highest average concentration in polluted air masses and the lowest in marine air masses. During the winter, the overall average BC value was 74.1 $\text{ng m}^{-3}$, whereas the overall summer mean BC value was higher by a factor of 3. The main reason for the seasonal difference may be enhanced thermal convection during summer, which increases transport of air pollutants from the planetary boundary layer of the surrounding urban area to this rural site. In the spring of 1998, abnormally high BC concentrations from the continental sector were measured. These concentrations were originating from a biomass burning smoke plume in Mexico. This was confirmed by the observations of the Earth Probe Total Ozone Mapping Spectrometer. The BC average concentrations of air masses transported from the polluted sector during summer were low on Sunday through Tuesday with a minimum value of 256 $\text{ng m}^{-3}$ occurring on Monday, and high on
Wednesday through Friday with a maximum value of 379 ng m\(^{-3}\) occurring on Friday. The net aerosol radiative forcing (scattering effects plus absorption effects) per unit vertical depth at 2006 m MSL was calculated to be \(-1.38 \times 10^{-3}\) W m\(^{-3}\) for the southeastern US. The magnitude of direct radiative forcing by aerosol scattering was reduced by 15 \(\pm\) 7\% due to the BC absorption.

SAGE II ozone and aerosol measurements and NCEP/NCAR potential vorticity and temperature fields during 1985-1999 were analyzed to study the spatial and temporal distributions of aerosol and ozone associated with the Antarctic polar vortex processes and to investigate the impact of polar stratospheric clouds (PSCs) on aerosol distribution and ozone depletion. During austral spring (Sep/Oct) there exist strong radial gradients in ozone, aerosol, and temperature near the polar vortex edge region and an isolation of materials inside polar vortex, confirming that springtime polar vortex acts effectively in keeping out intrusions of materials from the exterior of the polar vortex. The analysis of vertical profiles of 1020-nm aerosol extinction and 525- to 1020-nm aerosol extinction ratio inside spring polar vortex indicates aerosol enhancement in 9-15km layer and lack of larger particles in 15-20km layer, which is consistent with the effects of gravitational sedimentation and subsequent evaporation of PSCs. For the spherical PSC particles for which the Reynolds number < 1, gravitational sedimentation velocity is estimated to be 0.11, 0.04, and 1.5 km/wk for Type Ia, Ib, II PSCs, respectively. The probabilities of the occurrence of T<195K (threshold temperature of Type Ia PSC formation), T<192K (Type Ib), and T<190K (Type II) at 50mb pressure height are calculated in the region of 60-75 °S during austral winter (Jul/Aug). The probabilities of occurrence for the three
categories of temperature show increasing trends with time, whereas springtime
stratospheric column ozone amount shows decreasing trend. During volcanically
unperturbed years, the stratospheric column ozone in the spring is highly correlated with
the probability of occurrence of temperature below 195 K in the previous winter (R=−
0.88). The empirical relationship between the stratospheric column ozone amount (O₃) in
the spring (Sep/Oct) and the probability (P) of occurrence of T<195 K in the previous
winter (Jul/Aug) is found to fit the equation of O₃ = −360.38 × P + 446.38 (R²=0.78),
which can be used to predict ozone depletion from the previous winter temperature. In
addition, the most severe ozone depletion observed in the late 1990s is consistent with the
occurrence of the strong PV, and the empirical relationship between the O₃ and the PV
strength (|PV|) during the spring is also found to fit the equation of O₃ = −6.46 × 10⁶ |PV|
+ 651.92 (R²=0.93). These linear regression equations indicate that springtime ozone
depletion is strongly correlated with the PV strength and the previous winter temperature
for the formation of PSCs.

Our results obtained from remote sensing and in situ measurements aid in
understanding the microphysics of atmospheric aerosols, the meteorological transport of
atmospheric aerosols, and the impacts of PSCs on aerosol distribution and ozone
depletion, and improve the knowledge of the climatology in the southeastern United
States and the Antarctic.
5.2. Recommendations

The Stratospheric Aerosol and Gas Experiment III (SAGE III) was launched aboard the Russian Meteor-3M satellite in December, 2001, which measures aerosols as well as ozone, nitrogen dioxide, nitrogen trioxide, chlorine dioxide, and water vapor, mainly in the Arctic region. The most important of these are the vertical distributions of aerosols and ozone from the upper troposphere through the stratosphere. The high northern latitude coverage (50 °N – 80 °N) will provide insight into the processes leading to ozone loss during winter and spring. Therefore, it is recommended to investigate the inter-hemispheric differences by contrasting the SAGE III measurements in the Arctic to the results obtained from this study in the Antarctic. The study of the behavior of the polar aerosols and ozone should prove valuable for parameterization in multidimensional photochemical and dynamical models whose use is essential for fully understanding observed Antarctic ozone depletions as well as for assessing the possibility of similar phenomenon occurring in the future in the Arctic.

In addition, the SAGE III instrument contains 12 spectral channels over the wavelength region between 280 nm and 1560 nm and is essentially an improved version of the seven-channel SAGE II instrument. The SAGE III provides aerosol extinction coefficients at 9 wavelengths (385, 448, 521, 596, 676, 754, 868, 1019, and ~1550 nm). Using these aerosol extinction coefficients, it is recommended to retrieve the aerosol physical properties (e.g., mass concentration, surface area density, effective radius, size distribution, etc.). These aerosol physical quantities are important as input in climate, photochemical, and radiative forcing models. To retrieve the aerosol physical properties,
an inversion technique (e.g., Randomized Minimization Search Technique (RMST) [Heintzenberg et al., 1981]; a method to find a size distribution whose optical properties have a minimum deviation from the measured optical properties) can be applied. Its initial application to the SAGE II data set was conducted by Lin and Saxena [1992] and the comparison of this algorithm to the constrained linear inversion (CLI) method [King et al., 1978] was performed by Anderson et al. [2000]. The aerosol measurements and the retrieved parameters can be utilized in comparison with other remote sensing and in situ measurements.
5.3. References


