MODIS Observations of Global Tropospheric Aerosols in 2001

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ABSTRACT

It is of interest to monitor aerosols (i.e. desert dusts and sulfate aerosols from industrial emissions) over land and ocean because aerosol particles influence the temperature structure and chemistry of the Earth’s troposphere. MODIS (Moderate Resolution Imaging Spectroradiometer) observations of vertical column aerosol optical depths during 2001 were analyzed on a global and seasonal basis, and compared to regional sources of industrial tropospheric pollution. There was a general correspondence between annually averaged MODIS aerosol optical depth and that of the ECHMAN/GRANTOUR computer model. Both maps indicated high aerosol content over Africa and Asia. Furthermore, a general positive correlation was found when MODIS aerosol optical depth data for year 2001 were compared to regional sulfur dioxide (SO$_2$) emission values of Smith et al. (2001). Analysis of TOMS aerosol index, however, demonstrated that regional values of MODIS aerosol optical depth were influenced by the presence of desert dusts. Regional values were also influenced by the transport of aerosols from one region to another (e.g. from China to Japan). These results implied that future work, that will accurately distinguish aerosol types and factor in wind effects, is needed to use the MODIS instrument to study the impact of anthropogenic industrial SO$_2$ emissions upon tropospheric aerosol amount.

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Section 1: INTRODUCTION

Aerosols are tiny liquid or solid particles suspended in the air. The presence of aerosols in the atmosphere has several environmental consequences including acid deposition and stratospheric ozone depletion. Aerosols also interact with the Earth’s radiation budget and can greatly alter the climate in two ways: (1) direct radiative forcing by absorbing and scattering solar and infrared radiation, and (2) indirect radiative forcing due to changing cloud properties (IPCC Report, 2001). The effects of tropospheric aerosols on radiative balances, in particular, are currently being studied to improve the quality of future climate scenarios. To accurately quantify the radiative forcing of aerosols, however, much research is still needed about the sources and sinks of aerosols, their lifecycles in the atmosphere, and their distributions around the globe. Present knowledge of global aerosol climatology and variability is by no means complete.

Satellites are suitable for aerosol studies since they provide global observations of aerosol optical depths. An aerosol optical depth is a measure of the opacity of air. In other words, satellite measurements of aerosol optical depth indicate how opaque (or transparent) the atmosphere is in a given area. Several earlier satellite instruments such as the Meteosat, Geostationary Operational Environmental Satellite (GOES), Advanced Very High Resolution Radiometer (AVHRR), Along Track Scanning Radiometer (ATSR), and the Ocean Color and Temperature Scanner (OCTS) retrieved aerosol optical depths over the oceans in clear-sky conditions. These aerosol data, however, were limited in two respects. First, these satellite measurements were limited to reflectance measurements in only one or two spectral channels. Second, these instruments were capable of retrieving aerosol content over the oceans, but not over the land. In order to obtain complete measurements and observations of tropospheric aerosols, a satellite instrument with a wide spectral range and spatial resolution is necessary.

The Moderate Resolution Imaging Spectroradiometer (MODIS) was designed for this purpose. The MODIS experiment on the Earth Observing System (EOS) Terra and Aqua satellites takes daily measurements of atmospheric emission and reflection in 36 spectral bands in the visible and infrared spectrum. The MODIS science team solves for the total aerosol optical depth and many other atmospheric parameters from these observations. Aerosol spectral optical thickness measured by a global ground-based instrument, the Aerosol Robotic Network (AERONET), validates the reliability of the MODIS experiment by demonstrating that Terra aerosol measurements can represent the annual average value within 2% error (Kaufman et al., 2000).

The long term interest of this study is to monitor regional industrial emissions using a variety of datasets, including MODIS measurements of aerosol optical depth. The increasing concentration of atmospheric pollutants such as carbon monoxide (CO), sulfur dioxide (SO₂), oxides of nitrate (NOₓ), and aerosol particles is a growing problem today. As
a possible solution to this pollution problem, international agreements that will control industrial emission levels of regions are expected in the future. The implementation of such treaties will, however, require satellite observations to monitor pollutants worldwide. Therefore, the goal of this research was to find out whether aerosol information retrieved from MODIS and from other data sources will become useful to study the pollution problem.

In this study, several datasets were used to examine the global distribution of tropospheric aerosols. The 2001 annually averaged MODIS aerosol optical depth data were correlated with 2000 regional anthropogenic SO$_2$ emissions calculated by Smith et al. (2001). Because SO$_2$ is a major component of anthropogenic pollutants, sulfate aerosols produced from SO$_2$ provided a good measure of regional industrial activity. Furthermore, monthly averages of aerosol optical depths, retrieved from MODIS and from other data sources, were examined for each region to understand the seasonal variations of global aerosol distribution.

Section 2 provides an overview of the MODIS instrument—its mission, data products, and their retrieval methods. This is followed by descriptions and analysis techniques of four additional datasets, in Section 3. The Total Ozone Mapping Spectrometer (TOMS) aerosol index was investigated to study the distribution of desert dust. This information was used to separately examine global desert dust by comparing TOMS measurements of UV-absorbing aerosols (i.e. desert dust, smoke) with MODIS measurements of all tropospheric aerosols. Regional SO$_2$ emissions data from Smith et al. (2001) were correlated with the MODIS aerosol optical depths to determine the relationship between SO$_2$ emission and sulfate aerosol amount. The European Center for Medium-Range Weather Forecasts (ECMWF) wind field data were used to study the effects of winds on aerosol distribution. High aerosol content over an area observed by MODIS may be caused by wind transport of aerosols from surrounding areas, and not necessarily by natural or anthropogenic emissions from the surface below. In Section 4, the results of the analyses are discussed. Lastly, the main conclusions and future research goals are presented in Section 5.

Section 2: METHODOLOGY

2.1 Data Description

This section introduces the four datasets used in this study. It gives an overview of the type of information the datasets provide, their data retrieval methods, and the reliabilities and uncertainties of the data.

2.1.1 MODIS Experiment

MODIS (Moderate Resolution Imaging Spectroradiometer) is a key instrument on two Earth Observing System (EOS) satellites (Figure 1). The first MODIS experiment was launched into space on the EOS Terra satellite on December 18, 1999. Terra's sister ship,
Aqua, was launched on May 4, 2002. The MODIS instrument was built to National Aeronautics and Space Administration (NASA) specifications by the Hughes Santa Barbara Research Center to gather information about Earth's land, oceans, and atmosphere (Conboy, 2002). The mission objective is to provide a comprehensive series of global observations for constructing models of Earth's processes, so that better predictions can be made in the future. Principal MODIS data products include the following (Kaufman and Tanre, 1998):

- Cloud coverage and properties
- Vegetation and land-surface cover
- Snow and sea-ice cover and reflectance
- Ocean color
- Concentration of chlorophyll a
- Chlorophyll fluorescence
- Surface/cloud/atmospheric temperatures
- Aerosol concentration and optical properties

The last data product was studied for the purpose of this research.

*Figure 1. The MODIS instrument on the two Earth Observing System satellites, (Conboy, 2002).*
MODIS views the whole globe every one to two days using a 2,330 km-wide viewing swath. It acquires data in 36 spectral bands ranging in wavelength from 0.4 $\mu m$ to 14.4 $\mu m$ with a spatial resolution ranging from 250 m to 1,000 m. These characteristics give MODIS a great advantage over previous satellite measurements such as those of GOES, METEOSAT, and the AVHRR, whose reflectance measurements are limited to one or two channels. Kaufman et al. (1990) further demonstrated the benefit of MODIS over other satellite instruments by comparing the high sensitivity of MODIS 3.8 $\mu m$ channel to pollution aerosol or smoke, and the rather poor sensitivity of the corresponding AVHRR 3.7 $\mu m$ channel. In addition, past satellites did not view aerosol content over the land because of the difficulty incorporating sunlight reflectivity of variable land surfaces into the measured signal.

The MODIS Aerosol Product (MOD 04) monitors the aerosol optical thickness over the oceans and moist parts of the continents, simultaneously. The global remote sensing of aerosol uses 250 m resolution 0.66 $\mu m$ channel, the 500 m resolution 0.47 $\mu m$ and 2.1 $\mu m$ channels, and the 1000 m resolution 3.8 $\mu m$ and 11 $\mu m$ channels (Kaufman and Tanre, 1998). Aerosol retrieval over the oceans is done by a look-up table (LUT) approach (Kaufman and Tanre, 1998). The LUT contains radiative transfer values that have been calculated by a computer model, based on known aerosol and surface characteristics. These “pre-calculated values” from LUT are compared with the measured spectral radiance until two closely matching values are found. A collection of best fit values are then used to invert the measured radiance into aerosol optical thickness and volume distribution. To retrieve aerosol content over the land, dark surfaces must first be identified in the blue (0.47 $\mu m$) and red (0.66 $\mu m$) channels. After the dark surfaces are identified, the surface reflectance of the dark pixels are estimated and used to derive the optical thickness in these two channels. (For a more complete algorithm of MODIS remote sensing of aerosols, refer to Kaufman and Tanre, 1998.)

Polar orbiting satellites, like the EOS-MODIS satellite, are suitable for remote sensing of aerosols because they provide good spatial and spectral resolution while maintaining global coverage. Although data from Terra and Aqua are collected at specific times of the day only, Kaufman et al. (2000) found that their aerosol measurements can represent the daily average optical thickness value within 2% error. This has been confirmed by global ground-based instruments such as the Aerosol Robotic Network (AERONET) that are being used to supplement satellite records.

One of the questionable aspects of MODIS is its ability to distinguish aerosol types. The MODIS science team leader, Yoram Kaufman, of NASA, commented on this limitation during his talk at the Summer Colloquium at National Center for Atmospheric Research (NCAR) on July 15, 2002. Kaufman stated that he feels “pretty confident” on the MODIS instrument’s ability to distinguish aerosol types over the oceans, but that “more validation is
needed” over the continents. The accuracy of this aspect of MODIS needs further investigation.

2.1.2 TOMS Experiment

The Total Ozone Mapping Spectrometer (TOMS) data were used to supplement MODIS observations of desert dust. TOMS instrument monitors UV-absorbing aerosols over land and water. Since it does not measure properties of non-UV-absorbing aerosols, like sulfate aerosols, TOMS observations were useful in locating areas of abundant desert dust and/or smoke. Together with the MODIS total aerosol optical depth data, TOMS aerosol index was used to study the distributions and concentrations of dust and/or smoke, in comparison to other non-UV-absorbing aerosols.

The primary purpose of the TOMS experiment is to retrieve ozone. Other UV-absorbing aerosols detected by TOMS include smoke and soot aerosols from biomass burning and forest fires, volcanic ash clouds, and desert mineral dust. TOMS is able to observe global distributions of these tropospheric absorbing aerosols using reflectivity differences between the 340 nm and 380 nm channels. A table of backscattered Earth radiance was created as a function of total ozone or UV-absorbing aerosol. The computed radiances for a given set of conditions were then used to extrapolate from the table, the total ozone or UV-absorbing aerosol value in a given atmospheric column (Herman et al., 1996). Some of the uncertainties of TOMS values are listed below (Herman et al., 1996):

- Errors in the radiance measurements
- Errors in the values of input physical quantities obtained from laboratory measurements
- Errors in the parameterization of atmospheric properties used as input to the radiative transfer computations
- Limitations in the way the computations represent the physical atmospheric processes

However, these data uncertainties consisted of errors that are less than +/- 3 percent. Substantial calibrations have been done to test the accuracy and reliability of TOMS data products.

2.1.3 Regional SO₂ Emissions Data

The correlation between MODIS aerosol optical depth and regional SO₂ emission was studied to quantify the impact of human emissions on the atmosphere.

Table 1 lists eleven continental regions and their corresponding total anthropogenic SO₂ emissions (in teragrams of sulfur, TgS, per year) for 2000, estimated by Smith et al. (2001). The emissions were calculated using country-level emission inventories and regional fossil fuel sulfur content information. Smith et al. also included 1999 data from the Energy Information Administration (EIA) about changes in oil and coal consumption of
developing regions, such as Latin America and Africa, as well as changes in emission reduction controls for developed regions, such as North America and Europe.

All various sources of sulfur emissions were claimed reliable. Major uncertainties of Smith’s analysis were caused by the questionable emissions from coal, petroleum products, industrial processes, and biomass burning. In particular, emissions from developing regions were uncertain because complete and reliable information on energy-use to fuel properties was lacking. Uncertainties in the energy data, the sulfur content of fossil fuels, and the extent of de-sulfurization efforts of fuels may have caused errors in Smith’s estimates. There is a global uncertainty of +/- 10% for 2000 values.

Table 1. Estimated Regional SO$_2$ Emissions for Year 2000 for 11 Continental Regions from Smith et al. (2001)

<table>
<thead>
<tr>
<th>Regions</th>
<th>Total anthropogenic sulfur dioxide emissions (TgS per year)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Japan</td>
<td>0.4</td>
</tr>
<tr>
<td>Australia/New Zealand</td>
<td>1.0</td>
</tr>
<tr>
<td>Canada</td>
<td>1.4</td>
</tr>
<tr>
<td>Middle East</td>
<td>1.6</td>
</tr>
<tr>
<td>Africa</td>
<td>3.4</td>
</tr>
<tr>
<td>Western Europe</td>
<td>5.1</td>
</tr>
<tr>
<td>Latin America</td>
<td>6.7</td>
</tr>
<tr>
<td>USA</td>
<td>8.1</td>
</tr>
<tr>
<td>South and East Asia</td>
<td>9.4</td>
</tr>
<tr>
<td>FSU*/Eastern Europe</td>
<td>10</td>
</tr>
<tr>
<td>China/CPA**</td>
<td>18</td>
</tr>
</tbody>
</table>

*FSU = Former Soviet Union, **CPA = Centrally Planned Asia; See Smith et al. (2001) for regional details.

2.1.4 ECMWF Wind Field Data

The European Centre for Medium Range Weather Forecasts (ECMWF) wind field data were analyzed to study how wind effects MODIS aerosol measurements over each of the defined regions. This information was necessary because high aerosol optical depth over a certain region may have been a result of aerosols transported by wind from adjacent areas.

This dataset provided wind velocities over the entire globe. The observing system consisted of a mixture of synoptic and non-synoptic observations from satellites, buoys, aircrafts, and drifting balloons (Bengtsson et al., 1982).

Errors in the ECMWF wind field data may have occurred while assigning the heights of high-level satellite winds.

2.2 Data Analysis

Each of the datasets described in the previous sections were used accordingly to explain several tropospheric aerosol features. First, global distributions of aerosols were
studied by mapping global values of MODIS aerosol optical depth with IDL computer programming. The MODIS observation-based map of aerosol optical depth was then compared with a computer model-derived map of aerosol optical depth, to see if the maps agreed with each other. This was done so that locations of high aerosol optical depth, indicated by both maps, could be identified for regional investigation. Second, anthropogenic SO$_2$ emissions of eleven continental regions in 2000 from Smith et al. (2001) were correlated with their annually averaged MODIS aerosol optical depth. This correlation illustrated the relationship between regional industrial emissions and their corresponding aerosol amount. To examine the emissions and aerosol amount in a regional scale, eleven continental regions needed to be defined by their latitude and longitude boundaries. Third, seasonal variations in MODIS aerosol optical depth were examined for each of the defined regions to observe how aerosol content changed in 2001. Not only did this offer an understanding of the lifecycle of aerosols in the atmosphere, but it also allowed us to focus our investigation on months with extreme high or low aerosol optical depth.

A general correlation between industrial emission and optical depth was anticipated. However, aerosol optical depth was not expected to be a function of anthropogenic SO$_2$ emission alone. Since other factors also play a role in determining the presence of aerosols over a region, additional datasets were needed to explain the complicated relationship between industrial emission and optical depth, depending on a region’s geographic location and topographic features. For example, TOMS aerosol index was necessary to study the global distribution of desert dust because the presence of desert dust can enhance MODIS signals of aerosols. It would be misleading to correlate this enhanced MODIS aerosol optical depth value with regional SO$_2$ emission. The ECMWF wind field data were also important for identifying windblown aerosols from adjacent regions.

2.2.1 Latitude and Longitude of Regions

Using the anthropogenic SO$_2$ emission source regions chosen by Smith et al. (2001) as a starting guideline, latitudes and longitudes of seventeen oceanic and continental regions were defined (Table 2). Six oceanic regions were also added to Smith’s tabulation of regions. The latitude-longitude boundaries of continental regions were drawn so that all of the world’s major land masses were included. Oceanic regions encompassed the open ocean with few small islands.

These regions were carefully defined so that the annual averages of MODIS aerosol optical depth could be calculated for each region. These values were then used to correlate with Smith’s regional anthropogenic SO$_2$ emissions in order to quantify the impact of human emissions on the atmospheric aerosol content.
Table 2. Latitude-longitude Grids for 16 Continental and Oceanic Regions

<table>
<thead>
<tr>
<th>Region</th>
<th>Latitude</th>
<th>Longitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>Japan</td>
<td>30°N – 46°N</td>
<td>129°E – 147°E</td>
</tr>
<tr>
<td>Australia/New Zealand</td>
<td>11°S – 47°S</td>
<td>113°E – 178°E</td>
</tr>
<tr>
<td>Canada</td>
<td>45°N – 83°N</td>
<td>53°W – 141°W</td>
</tr>
<tr>
<td>Middle East</td>
<td>11°N – 40°N</td>
<td>32°E – 75°E</td>
</tr>
<tr>
<td>Africa</td>
<td>35°S – 33°N</td>
<td>18°W – 51°E</td>
</tr>
<tr>
<td>Western Europe</td>
<td>36°N – 70°N</td>
<td>10°W – 15°E</td>
</tr>
<tr>
<td>Latin America</td>
<td>56°S – 32°N</td>
<td>117°W – 35°W</td>
</tr>
<tr>
<td>USA</td>
<td>25°N – 49°N</td>
<td>67°W – 125°W</td>
</tr>
<tr>
<td>South and East Asia</td>
<td>10°S – 35°N</td>
<td>70°E – 154°E</td>
</tr>
<tr>
<td>FSU/Eastern Europe</td>
<td>40°N – 80°N</td>
<td>15°E – 170°E</td>
</tr>
<tr>
<td>China/CPA</td>
<td>20°N – 52°N</td>
<td>75°E – 135°E</td>
</tr>
<tr>
<td>North Pacific Ocean</td>
<td>30°N – 60°N</td>
<td>150°W – 180</td>
</tr>
<tr>
<td>South Pacific Ocean</td>
<td>0 – 50°S</td>
<td>180 – 81°W</td>
</tr>
<tr>
<td>West Pacific Ocean</td>
<td>0 – 30°N</td>
<td>130°E – 180</td>
</tr>
<tr>
<td>North Atlantic Ocean</td>
<td>10°N – 45°N</td>
<td>60°W – 18°W</td>
</tr>
<tr>
<td>South Atlantic Ocean</td>
<td>0 – 50°S</td>
<td>35°W – 10°E</td>
</tr>
<tr>
<td>Indian Ocean</td>
<td>10°S – 50°S</td>
<td>50°E – 113°E</td>
</tr>
</tbody>
</table>

To separately look at the contributions of desert dust and sulfate aerosols from desert and industrial regions of China, two sub-regions were defined under China (Figure 2, Table 3). Details on the analysis of these sub-regions are discussed in Section 3.

![Map of China and defined regions](image)

Figure 2. A map of China (Expedia.com Travel) and the defined desert and industrial regions.

Table 3. Latitude-longitude Grids for Sub-regions of China

<table>
<thead>
<tr>
<th>Sub-regions</th>
<th>Latitude</th>
<th>Longitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>Desert China</td>
<td>35°N – 50°N</td>
<td>75°E – 110°E</td>
</tr>
<tr>
<td>Industrial China</td>
<td>20°N – 52°N</td>
<td>110°E – 130°E</td>
</tr>
</tbody>
</table>

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2.2.2 **MODIS Aerosol Optical Depth Data**

A global map 2001 annually averaged MODIS aerosol optical depth was created using IDL. The MODIS aerosol optical depth map was then compared with an optical depth map derived by a computer model to see how closely the observations matched with model predictions. The model used in this study, the ECHAM/GRANTOUR model, is similar to the coupled climate and chemistry model described by Chuang et al. (1997), but with updated cloud scavenging and convective processes. This model was chosen because of its reference use in the Intergovernmental Panel on Climate Change (IPCC) Report (2001).

In addition to creating a global map of aerosol optical depth and using it for a comparison with a model-derived map, regional maps of MODIS aerosol optical depth were also created to examine areas of high optical depth in more detail.

2.2.3 **Regional SO$_2$ Emissions Data vs. MODIS Aerosol Optical Depth Data**

For each of the seventeen regions, annually and spatially averaged MODIS aerosol optical depth was graphed as a function of Smith's regional anthropogenic SO$_2$ emission to study the correlation between industrial emissions and aerosol amount. We assumed that oceanic regions do not emit anthropogenic SO$_2$ (i.e. zero SO$_2$ emissions). Outlier regions whose data points lay considerably higher or lower than the general trendline were noted for further analysis. (See Section 2.3 on Sulfur Chemistry for an explanation on the mechanism of sulfate aerosol production from SO$_2$ oxidation process.)

2.2.4 **Seasonal Variations**

Monthly averages of MODIS aerosol optical depth were graphed for all regions to examine the seasonal variations of aerosols. These graphs were compared with each other for similarities and differences that help explain the relationship of aerosol properties between adjacent regions. Months of maximum and minimum aerosol optical depth were noted to compare the results of other datasets during those months. The comparisons offered possible causes for why aerosol optical depth was high or low, increased or decreased, throughout the year.

2.2.5 **TOMS Aerosol Index**

The TOMS aerosol index was used to obtain a map of the global distribution of desert dust. This map was visually compared with the MODIS aerosol optical depth map to see if areas of high optical depth and high aerosol index corresponded with each other. If a region observed high measurements from both TOMS and MODIS instruments, then this indicated the presence of desert dust and/or smoke over that particular region. If MODIS alone measured a high value of optical depth, then the signal was most likely coming from sulfate aerosols, the predominant non-UV-absorbing aerosol.
Daily measurements of TOMS aerosol index were created for all seventeen regions to study the seasonal variations of UV-absorbing aerosols. As with the seasonal variation graphs of MODIS aerosol optical depth, these graphs were used for comparison purposes. Similar seasonal variations suggest a close relationship between adjacent regions in terms of aerosol type and properties. Unlike monthly averages, daily measurements provided more detailed and specific characteristics of the seasonal variation. Regions such as Africa, the Middle East, and China were particularly of interest because of their large desert areas.

2.2.6 Wind Field Effects

Monthly wind field vectors at 500 hPa (5×10^4 Pa) were mapped globally. Regional maps were also produced for certain regions that demanded further investigation due to high or low optical depth, relative to its anthropogenic SO₂ emission. An image of the wind fields of several regions combined illustrates a possible transport mechanism of materials (e.g. aerosols) from one region to another.Regional maps of the wind field during the months of maximum and minimum aerosol optical depth were examined comparatively to see if wind speed and/or direction differed between those months. This may or may not explain the driving force behind the presence of aerosol over a region, as well as its seasonal variation.

2.3 Sulfur Chemistry

The majority of sulfate aerosols are anthropogenic, and produced mainly by combustion of sulfur containing fuels (coal and oil) and smelting of sulfur containing ores (copper, lead, and zinc) (Jacob 249). Dimethyl sulfide (DMS) produced by marine plankton and sulfur compounds emitted by volcanoes also contribute to the global sulfate aerosol reservoir. Nevertheless, even when the two sources are combined, they comprise less than half of the fossil fuel and industry related sulfate aerosol emissions (Table 4; IPCC, 2001). Therefore, in this study, we assumed that the contribution of non-anthropogenic sources to sulfate aerosols were negligible.

<table>
<thead>
<tr>
<th>Source</th>
<th>Northern Hemisphere</th>
<th>Southern Hemisphere</th>
<th>Global</th>
<th>Range</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fossil fuel and industry</td>
<td>68</td>
<td>8</td>
<td>76</td>
<td>60-100</td>
</tr>
<tr>
<td>Volcanoes</td>
<td>6.3</td>
<td>3.0</td>
<td>9.3</td>
<td>6-20</td>
</tr>
<tr>
<td>DMS</td>
<td>11.6</td>
<td>13.4</td>
<td>25.0</td>
<td>12-42</td>
</tr>
</tbody>
</table>
SO₂ emitted in the atmosphere reacts with various constituents under gaseous and aqueous processes. The oxidation reaction of gaseous SO₂ is as follows (Jacob 249):

\[
\begin{align*}
SO_2 + OH & \rightarrow HSO_3 \\
HSO_3 + O_2 & \rightarrow SO_3 + HO_2 \\
SO_3 + H_2O & \rightarrow H_2SO_4
\end{align*}
\]

Reactions (2) and (3) are fast, but because reaction (1) is relatively slow, the lifetime of gaseous SO₂ is relatively long, or one to two weeks.

The lifetime of aqueous SO₂, however, is much shorter. The timescale of aqueous SO₂ oxidation is on the order of hours due to its faster reaction with hydrogen peroxide (H₂O₂). The oxidation reaction of SO₂ in the aqueous phase is as follows (Seinfeld and Pandis 348):

\[
\begin{align*}
SO_2 (g) + H_2O & \rightarrow SO_2 \cdot H_2O \\
SO_2 \cdot H_2O & \rightarrow HSO_3^- + H^+ \\
HSO_3^- + H_2O_2 (aq) + H^+ & \rightarrow SO_4^{2-} + 2H^+ + H_2O
\end{align*}
\]

The aqueous-phase oxidation process takes place in cloud droplets. Field measurements and theoretical studies have confirmed that reaction (6) occurs very fast that SO₂ and H₂O₂ do not normally coexist in clouds (Seinfeld and Pandis 818-9).

Another compound that reacts with SO₂ is ozone, O₃. Although SO₂ reaction with O₃ is slower than its reaction with H₂O₂, this process is still much quicker than the gaseous-phase processes.

\[
SO_2 + O_3 \rightarrow SO_4^{2-} + O_2
\]

Langner and Rodhe (1991) found that more than 70% of global oxidation of SO₂ to SO₄²⁻ occurs by means of aqueous phase processes. Chuang et al. (1997) also predicted that aqueous-phase SO₂ oxidation in clouds is the most important mechanism for producing sulfate on a global scale (Table 4). The amount of sulfate aerosols in the atmosphere, therefore, is determined by the availability of reactants (or oxidants) and cloud coverage. It is generally known that atmospheric sulfate concentrations are seasonally variable because of OH production, and consequently, O₃ and H₂O₂ production, varies throughout the year. The concentrations of OH and of its by-products peak during the summer and decrease during the winter.
Table 5. Estimated Annual Fluxes of Sulfate (in TgS per year) (Chuang et al., 1997)

<table>
<thead>
<tr>
<th>Sources</th>
<th>Northern Hemisphere</th>
<th>Southern Hemisphere</th>
<th>Global</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gaseous oxidation</td>
<td>4.6</td>
<td>1.5</td>
<td>6.1</td>
</tr>
<tr>
<td>Aqueous oxidation</td>
<td>29.8</td>
<td>12.5</td>
<td>42.3</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Sinks</th>
<th>Northern Hemisphere</th>
<th>Southern Hemisphere</th>
<th>Global</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry deposition</td>
<td>3.7</td>
<td>1.5</td>
<td>5.2</td>
</tr>
<tr>
<td>Wet deposition</td>
<td>30.7</td>
<td>12.5</td>
<td>43.2</td>
</tr>
</tbody>
</table>

Sulfate aerosols are removed from the atmosphere by either wet or dry deposition. The annual fluxes of sulfate for wet and dry deposition are listed on Table 5 (Chuang et al., 1997). The mechanism of removal depends on the size of the aerosol particle. Dry deposition or sedimentation is dominated by large (or coarse) particles that have diameters more than 1 μm. Small (or fine) particles with diameters between 0.1 μm and 1 μm are deposited via wet processes such as precipitation. Lifetimes of aerosols are dependent upon the removal mechanism and particle size (see Figure 4.16 in Brasseur 143).

Sulfate aerosols fall into the fine (less than 1 μm radius) aerosol category and are the largest constituent (about 37%) of all fine tropospheric aerosols (Heintzenberg, 1989). There are, however, coarse (1 μm radius or larger) aerosols in the troposphere as well. The coarse aerosols are primarily represented by desert dust. Table 6 lists the emissions and loadings, or the amount in the atmosphere, of these predominant fine and coarse aerosol types. The different sources of sulfur have been specified (in italics) under the total sulfur emissions and loadings.

Table 6. Emissions and Loadings of Desert Dust and Sulfur

<table>
<thead>
<tr>
<th>Aerosol</th>
<th>Emissions (Tg per year)</th>
<th>Loadings (Tg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Desert Dust*</td>
<td>3307</td>
<td>46.2</td>
</tr>
<tr>
<td>Total Sulfur**</td>
<td>100.5</td>
<td>99.6</td>
</tr>
<tr>
<td>(Anthropogenic)</td>
<td></td>
<td>37</td>
</tr>
<tr>
<td>(Biomass burning)</td>
<td></td>
<td>1.6</td>
</tr>
<tr>
<td>(Dimethyl Sulfide/DMS)</td>
<td>18</td>
<td>25</td>
</tr>
<tr>
<td>(Volcanoes)</td>
<td>14</td>
<td>36</td>
</tr>
</tbody>
</table>

* (Luo et al., 2002), ** (IPCC Report, 2001)

This table illustrates that even though the emission of desert dust is much higher than the emissions of total sulfur, sulfur is more commonly found in the atmosphere than desert dust. This is probably because much of the desert dust is deposited back on the surface due to its large size. Furthermore, the main source of sulfur is anthropogenic, which underlines the importance of SO₂ emissions and the oxidation processes that yield sulfate aerosols.
Section 3: RESULTS & DISCUSSION

3.1 Map Comparison

Annual averages of MODIS aerosol optical depth were mapped (Figure 3a) and compared to the ECHAM/GRANTOUR (IPCC Report, 2001) computer model-derived map of aerosol optical depth (Figure 3b) to see how well MODIS observations agreed with model predictions. One apparent difference between the two maps was that the MODIS map was missing data over some continental regions (e.g. Northern Africa and the Middle East region) whereas the model-derived map displayed a more complete and continuous image of aerosol optical depth. The MODIS instrument experienced difficulty retrieving aerosol data over these areas due to the high reflectivity of their deserts (Kaufman, 2002). MODIS cannot retrieve aerosol data over regions with high reflectance because the signal contrast between aerosol and land/ocean surface is too small. For the same reason, MODIS also suffers lack of data over the oceans at certain times of the day because of sea surface glints.

Although observations from the MODIS instrument were incomplete, its aerosol optical depth generally corresponded with those of the ECHAM/GRANTOUR model. For example, both maps indicated high optical depths over West Africa, the Iran-Pakistan region, China, Japan, and the North Pacific Ocean. (The reasons for their high optical depths were investigated, and are explained in Section 3.2) There was also an agreement over the Himalayas, where values dropped significantly due to their high elevation. Because the air column over high mountains is shallow, the atmosphere over mountainous regions cannot hold as many aerosols as in an atmosphere over lowlands that have deeper air columns above them. Another resemblance between Figures 3a and 3b was the contrast between aerosol optical depths of northern and southern hemispheres. Higher optical depths were displayed in the northern hemisphere compared to the southern hemisphere. This was partly because the northern hemisphere has vast land and desert coverage that emit vegetation debris and dust in the atmosphere. Larger population and higher industrial activity, which add more aerosols to the atmosphere, are also concentrated in the northern hemisphere.

3.2 SO₂ Emission and Aerosol Optical Depth Correlation

Regional anthropogenic SO₂ emissions (in TgS/yr) from Smith et al. (2001) were graphed against annual averages of MODIS aerosol optical depth (Figure 4). In general, the two variables were positively correlated. China, which had the highest anthropogenic SO₂ emission (18 TgS/yr) of all regional emissions, observed the second highest aerosol optical depth value (0.325) after the Middle East (0.370). On the other hand, oceanic regions with no anthropogenic SO₂ emissions had low values of aerosol optical depth. MODIS aerosol optical depths of regions with intermediate SO₂ emissions, such as those of the USA and Europe, lie neatly within the trendline. These results were reasonable and agreed with our
Figure 3a. Annually averaged MODIS observation of aerosol optical depth in 2001.

Figure 3b. ECHAM/GRANTOUR computer model of aerosol optical depth (IPCC Report, 2001). (Note: the color bar for Figure 3a also applies to this figure.)
Figure 4. MODIS total aerosol optical depth as a function of regional anthropogenic SO$_2$ emissions from Smith et al. (2001).
understanding of sulfur chemistry (see Section 2.3); under the same atmospheric conditions, the amount of sulfate aerosols in an air column largely depends on the amount of SO₂ that are available for oxidation in the atmosphere. Regional aerosol optical depths reflected this relationship fairly well.

Nevertheless, not all regions fit the trendline as precisely as others. Three outlier regions that stood out were Japan, the North Pacific Ocean, and the Middle East. Africa’s aerosol optical depth was also considerably higher than its regional SO₂ emission would have predicted. The explanations for why MODIS observed such high aerosol optical depths over each of these regions, despite their low SO₂ emissions, are described in Sections 3.2.2 – 3.2.5.

3.2.1 China

It is evident from Figure 4 that high aerosol content is observed over China. The simple explanation behind this observation is that MODIS was detecting sulfate aerosols that were being produced by the oxidation of China’s large anthropogenic SO₂ emission. However, the explanation may not be as simple as that. Do China’s aerosols consist purely of industry-related sulfate aerosols and no other aerosol type? Apparently, this is not true.

In China, the third largest nation in the world with an area of 9.6 million square kilometers, there are deserts and heavily industrialized cities that are each concentrated in different parts of the country (Figure 4). Populated cities that emit SO₂ and produce large quantities of sulfate aerosols, including Beijing and Shanghai, are located on the east coast of China. Desert regions that are sources for windblown mineral dust are found further inland. For instance, the Gobi Desert is by the China-Mongolia border and the Taklimakan Desert is located in the western part of China. Aerosols observed by MODIS over eastern China, therefore, are likely to be a combination of sulfate aerosols produced in the east and desert dust transported from the west.

To quantify the contribution of two types of aerosols to the aerosol optical depth separately, industrial and desert regions of China were first defined (Figure 2, Table 3). The seasonal variations of monthly averaged MODIS aerosol optical depth in 2001 were then graphed separately for these regions (Figure 5a,b). The desert region observed a gradual decline in aerosol optical depth values from February through December. The industrial region observed a peak in the springtime and declined thereafter. The maximum optical depth of industrial China suggested an enhanced industrial activity during the springtime, but additional data analyses are necessary to prove this point.

The optical depths of the industrial region were slightly higher than those of the desert region. Since the desert region is located in a remote area of the Asian continent, the only contribution to the aerosols found over western China would be desert dust. On the other hand, aerosols over industrial regions of China are likely to be a combination of sulfate aerosols from industrial emissions and desert dust transported from western China.
Figure 5a. Seasonal variation in MODIS aerosol optical depth of desert China.

Figure 5b. Seasonal variation in MODIS aerosol optical depth of industrial China.
Therefore, monthly averaged aerosol optical depths over industrial China were probably an overestimate, due to the contribution of mineral dust from desert China.

Daily measurements of TOMS aerosol index in 2001 over industrial and desert regions of China were graphed to quantify the contribution of desert dust to the aerosol index of both regions (Figure 6a,b). According to Figures 6a and 6b, seasonal variations were highly correlated. If aerosol transport from the desert region to the industrial region takes approximately two days, as calculated from an average observed wind speed of 15m/s, and if most of the airborne dust from deserts in the west are being transported to the east, it makes much sense that the seasonal variations of the east and of the west are similar, if not alike. In other words, it is very likely that desert dust from western China contributed substantially to the total aerosol content over industrialized eastern China, thereby causing similar peaks and valleys in their seasonal variations.

These analyses tell us that China’s high aerosol optical depth observed by MODIS and predicted by the model was due to a combination of sulfate aerosols from anthropogenic SO₂ emission of industrialized eastern China and desert dust transported from desert China in the west.

3.2.2 Japan

Figure 7 shows the seasonal variation in monthly averaged MODIS aerosol optical depth in 2001 for Japan. High aerosol optical depths were observed in the springtime, with the maximum value in April. Low values were observed in the fall, during November and December.

To find out why the aerosol optical depth over Japan was so large in April, despite its low SO₂ emission, the source region of Japan’s aerosols needed to be determined. MODIS aerosol optical depth map of Japan-China region in April clearly illustrated a streak of aerosols, stretching from China to Japan, eventually reaching the North Pacific Ocean (Figure 8a). This continuous streak of aerosols suggested that aerosols were being transported from China to adjacent regions to its east. Since SO₂ is quickly converted to sulfate in a matter of hours by aqueous-phase processes (Section 2.3), it was expected that sulfate aerosols produced by China’s SO₂ emission would influence Japan’s aerosol optical depth. The ECMWF wind field data for Japan-China region in April 2001 further supported this hypothesis of aerosol transport from China (Figure 8b). Winds were clearly westerly, capable of transporting materials from China to Japan. In December, when optical depth was at its minimum over Japan, the aerosol band was not easily definable (Figure 9a). However, westerly winds were again observed during this time of year (Figure 9b). Wind direction, therefore, could not have been the driving force behind Japan’s seasonal variation.

If aerosols were indeed transported from China to Japan by fairly constant westerly winds throughout the year, Japan’s seasonal variation in aerosol optical depth should match
Figure 6a. Seasonal variation in TOMS aerosol index of desert China.

Figure 6b. Seasonal variation in TOMS aerosol index of industrial China.
Figure 7. Seasonal variation in MODIS aerosol optical depth of Japan.
Figure 8a. MODIS observation of aerosol optical depth of Japan in April 2001.

Figure 8b. ECMWF wind fields of Japan in April 2001.
Figure 9a. MODIS observation of aerosol optical depth of Japan in December 2001.

Figure 9b. ECMWF wind fields of Japan in December 2001.
with that of China. As expected, the seasonal variation in MODIS aerosol optical depth observed over Japan (Figure 7) corresponded well with that of industrial China (Figure 5a). The correlation was weaker with the seasonal variation of desert China (Figure 5b) because sulfate aerosols were not yet added to the total aerosol budget. Daily measurements of TOMS aerosol index for Japan (Figure 10), on the other hand, matched well with both the graphs of desert and industrial China (Figure 6a, b). This indicated that both the desert dust from western China and the sulfate aerosols produced over eastern China had an effect on the aerosol content over Japan.

In general, these results confirmed that both the desert dust from western China and the sulfate aerosols produced over eastern China were transported to Japan by wind, adding to the total amount of aerosols over Japan.

3.2.3 North Pacific Ocean

The North Pacific Ocean observed high aerosol optical depth for the same reason as Japan. The seasonal variation in monthly averaged MODIS aerosol optical depth of the North Pacific Ocean (Figure 11) was similar to that of industrial China and Japan (Figure 5b and 7). Aerosol optical depth values peaked in April and declined during the winter months. In April, the long streak of aerosols from China reached as far as the North Pacific Ocean (Figure 8a). The ECMWF wind fields also indicated southwesterly and westerly flow over the North Pacific Ocean that supported the hypothesis that aerosols were being transported from the continental regions (Figure 8b, 9b).

Furthermore, the seasonal variation in MODIS aerosol optical depth observed over the North Pacific Ocean (Figure 11) corresponded well with that of industrial China (Figure 5b) and Japan (Figure 7), but not of desert China (Figure 5a). Daily measurements of TOMS aerosol index for the North Pacific Ocean (Figure 12) was also very similar to those of industrial and desert China, and of Japan. These results indicated that the influence of desert dust from western China and sulfate aerosols over eastern China extended as far east as the North Pacific Ocean.

3.2.4 Middle East

Figure 13 shows the seasonal variation in monthly averaged MODIS total aerosol optical depth in 2001 for the Middle East region. According to the graph, the highest and the lowest aerosol optical depths occurred in July and December, respectively. The MODIS aerosol optical depth map for July clearly indicated a patch of high optical depth over the northern Arabian Sea (Figure 14a); whereas in December, the patch was not visible (Figure 14b). Unfortunately, aerosols were not detected over land for the entire Middle East region because of the high reflectivity of deserts discussed earlier.

To determine the type of aerosols observed by MODIS over the northern Arabian Sea
Figure 10. Seasonal variation in TOMS aerosol index of Japan.
Figure 11. Seasonal variation in MODIS aerosol optical depth of the North Pacific Ocean.

Figure 12. Seasonal variation in TOMS aerosol index of the North Pacific Ocean.
Figure 13. Seasonal variation in MODIS aerosol optical depth of the Middle East.
Figure 14a. MODIS observation of aerosol optical depth of the Middle East in July 2001.

Figure 14b. MODIS observation of aerosol optical depth of the Middle East in December 2001.
(i.e. sulfate aerosols, desert dust, or sea salts), daily measurements of TOMS aerosol index over the Middle East region for July 2001 were examined. Since the TOMS experiment only detects UV-absorbing aerosols, its aerosol index indicated the presence of smoke, soot, volcanic ash clouds and/or desert dust, but not of sulfate aerosols. Global maps of TOMS aerosol index for periods between July 5, 2001 and July 7, 2001 are shown in Figures 15a-c. Similar to the monthly averaged MODIS aerosol optical depth map of the Middle East region for July 2001 (Figure 14a), high values of TOMS aerosol index were observed over the northern Arabian Sea. This indicated that aerosols observed by MODIS over the northern Arabian Sea in July 2001 were not sulfate aerosols. Considering the location of the high aerosol patch over the sea, directly south of the deserts of Iran and Pakistan, it was highly likely that MODIS and TOMS satellite instruments were observing windblown desert dust.

To test this hypothesis, July 2001 ECMWF wind field data were examined for the Middle East region (Figure 16a). Clearly, the observed winds in the Middle East region were blowing from the coast of Iran and Pakistan toward the Arabian Sea, suggesting the presence of windblown dust from land to sea. In contrast, wind fields in December were westerly, and were not transporting dust into the Arabian Sea from its northern end (Figure 16b). It was, therefore, reasonable to conclude that aerosols observed by MODIS over northern Arabian Sea in July were mineral dust that were blown southward from the deserts of Iran and Pakistan. This explained why the aerosol optical depth value of the Middle East was much higher than predicted by its anthropogenic SO₂ emission.

3.2.5 Africa

Figure 17 shows the seasonal variation in monthly averaged MODIS aerosol optical depth in 2001 over Africa. According to this graph, high optical depths were observed twice in 2001, during early fall (August, September) and winter (January, February). The lowest optical depth was observed in April. MODIS observations of maximum and minimum aerosol optical depths over Africa were examined (Figure 18a, b). In order to determine the type of aerosols they were, TOMS aerosol index was again observed for the month of September and April (Figure 19a, b). It is evident from Figures 18 and 19 that TOMS observations of UV-absorbing aerosols over Africa during September and April corresponded well with MODIS observations of aerosols. Therefore, as done with the Middle East analysis, we were able to identify Africa’s aerosols observed by MODIS as desert dust that were distributed locally within Africa. They were not sulfate aerosols produced from Africa’s anthropogenic SO₂ emission or emissions of other adjacent continental regions.

Unfortunately, the MODIS instrument did not retrieve aerosol data over northern Africa. MODIS measurements of aerosols, therefore, underestimated the total amount of aerosols found over Africa, as stated by Yoram Kaufman (NCAR Summer Colloquium, 15 July 2002). The optical depth value of Africa in Figure 4 would, therefore, be higher, if
Figure 15a. TOMS aerosol index on July 5, 2001.

Figure 15b. TOMS aerosol index on July 6, 2001.

Figure 15c. TOMS aerosol index on July 7, 2001.
Figure 16a. ECMWF wind fields of the Middle East in July 2001.

Figure 16b. ECMWF wind fields of the Middle East in December 2001.
Figure 17. Seasonal variation in MODIS aerosol optical depth of Africa.
Figure 18a. MODIS observation of aerosol optical depth of Africa in August 2001.

Figure 18b. MODIS observation of aerosol optical depth of Africa in April 2001.
Figure 19a. TOMS aerosol index on August 2001.

Figure 19b. TOMS aerosol index on April 2001.

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MODIS did not have the aerosol retrieval problem over the Saharan Desert. Nevertheless, the investigation of aerosol characteristics observed over Africa showed that the MODIS observation of Africa’s aerosol optical depth was relatively high because of abundant airborne dust from its deserts.

Section 4: CONCLUSIONS

MODIS observations of aerosol optical depth in 2001 illustrated the global distribution of tropospheric aerosols. A comparison with the ECHAM/GRANTOUR computer model showed that there was a general correspondence between the observation-based map and the model-derived map of the total aerosol optical depth. For example, both maps indicated low values of aerosol optical depth over major ocean basins. The maps also showed an agreement over regions in Africa and Asia, where high aerosol optical depths were observed, although for different reasons. By using additional datasets, such as the TOMS aerosol index and the ECMWF wind field data, we found that aerosols over the Middle East and Africa were mainly desert dust, whereas aerosols over Japan and the North Pacific Ocean were primarily sulfate aerosols (and desert dust) that were transported from China.

When annually averaged MODIS observations of aerosol optical depth were compared to the regional anthropogenic SO$_2$ emissions, a positive correlation was found. This correlation between MODIS aerosol optical depths and regional industrial emissions suggested the future use of the MODIS instrument to monitor regional pollution contribution. However, the MODIS aerosol dataset alone is not sufficient to illustrate the relationship between industrial emission and aerosol amount of certain regions. When the high aerosol optical depths of the Middle East, Japan, the North Pacific Ocean, and Africa were investigated and analyzed, we found that aerosol type information and atmospheric conditions (i.e. wind field) during measurements needed to be quantified. These parameters had to be taken into account for a better and accurate correlation between industrial emissions and aerosol optical depths.

One reason for the complications of the correlation is the limitations of the current MODIS instrument. Although its attempt to retrieve aerosol data over land is an improvement upon many previous satellite instruments, various images of MODIS aerosol optical depth showed that global measurements of aerosols are still incomplete. It is evident from the incomplete images that MODIS is having difficulty retrieving aerosol data over some parts of land with high reflectivity, such as the deserts of Africa and the Middle East. For this reason, Kaufman points out that the MODIS measurements of aerosols over land are underestimating the true values of total aerosol content (NCAR Summer Colloquium, 15 July 2002). In addition to improving aerosol retrieval over land, the MODIS team needs to
develop a way to better distinguish various aerosol types. These comments made by Kaufman confirmed our understanding of the limitations and capabilities of the current MODIS instrument.

An improved understanding of the MODIS instrument is simply a gateway to extensive aerosol studies of the future. One way of accomplishing this goal is to develop a satellite instrument that combines several datasets, including information from TOMS, lidars, and polarimeters. Once this Aerosol Satellite (AEROSAT), or what Kaufman describes as the “dream satellite,” is developed, the global distribution and atmospheric properties of various aerosol types need to be studied separately. If, for example, sulfate aerosols and desert dust are partitioned accurately, industrial emissions will be better correlated with anthropogenic aerosol content. In addition to the TOMS aerosol index and the ECMWF wind field data, the outgoing longwave radiation (OLR) dataset should also be analyzed in future work because this dataset indicates the presence of convective systems. Deep convection can lift large quantities of surface aerosols into the atmosphere, which will enhance the aerosol optical depth value observed by MODIS. The effects of these additional datasets need to be analyzed and quantified to modify the regional values of aerosol optical depth.

In conclusion, the study of 2001 MODIS observations of global tropospheric aerosols will be useful in setting the stage for future work that will attempt to use the MODIS instrument to study the impact of anthropogenic SO$_2$ emissions upon tropospheric aerosol amount.

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