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## Increase in background stratospheric aerosol observed with lidar at Mauna Loa Observatory and Boulder, Colorado

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[1] The stratospheric aerosol layer has been monitored with lidars at Mauna Loa Observatory in Hawaii and Boulder in Colorado since 1975 and 2000, respectively. Following the Pinatubo volcanic eruption in June 1991, the global stratosphere has not been perturbed by a major volcanic eruption providing an unprecedented opportunity to study the background aerosol. Since about 2000, an increase of 4–7% per year in the aerosol backscatter in the altitude range 20–30 km has been detected at both Mauna Loa and Boulder. This increase is superimposed on a seasonal cycle with a winter maximum that is modulated by the quasi-biennial oscillation (QBO) in tropical winds. Of the three major causes for a stratospheric aerosol increase: volcanic emissions to the stratosphere, increased tropical upwelling, and an increase in anthropogenic sulfur gas emissions in the troposphere, it appears that a large increase in coal burning since 2002, mainly in China, is the likely source of sulfur dioxide that ultimately ends up as the sulfate aerosol responsible for the increased backscatter from the stratospheric aerosol layer. The results are consistent with 0.6–0.8% of tropospheric sulfur entering the stratosphere. **Citation:** Hofmann, D., J. Barnes, M. O'Neill, M. Trudeau, and R. Neely (2009), Increase in background stratospheric aerosol observed with lidar at Mauna Loa Observatory and Boulder, Colorado, *Geophys. Res. Lett.*, *36*, L15808, doi:10.1029/2009GL039008.

### 1. Introduction

[2] Major volcanic eruptions inject large amounts of sulfur dioxide (SO<sub>2</sub>) into the stratosphere increasing the optical thickness of the stratospheric sulfate aerosol layer that has both climatic (cooling) effects and stratospheric ozone layer (heterogeneous chemical) implications [Hofmann and Solomon, 1989]. SO<sub>2</sub> is oxidized primarily by OH to sulfuric acid vapor with a lifetime of 30–40 days [McKeen et al., 1984] that condenses into an H<sub>2</sub>SO<sub>4</sub> - H<sub>2</sub>O liquid aerosol in the stratosphere [Rosen, 1971]. Major volcanic eruptions (e.g., Pinatubo in June 1991) can increase the stratospheric aerosol mass by more than an order of magnitude. This eruption produced approximately  $-3 \text{ Wm}^{-2}$  radiative forcing, cooling the troposphere by about 0.5°C [Hansen et al., 1997], and decreased stratospheric ozone by ~1–2%. However, the perturbation is transient because the

super-cooled droplets fall out of the stratosphere with a characteristic time of 1–2 years. During non-volcanic periods, the stratospheric sulfate layer is sustained at low levels by SO<sub>2</sub>, COS and other sulfurous gases that provide sulfur to the stratosphere with entry mainly in the tropics. The period following the decay of the Pinatubo aerosol [Barnes and Hofmann, 1997], from 1996 to the present, is the longest period free of major volcanic eruptions since the sulfate layer was discovered in 1959 [Junge et al., 1961]. This lull in major volcanic activity has provided an unusual opportunity to study the background stratospheric aerosol.

### 2. Observations

[3] Backscatter of Nd:YAG lidar (532 & 1064 nm) laser light pulses is used to detect the stratospheric aerosol layer at Mauna Loa Observatory (19°N) in Hawaii and Boulder, Colorado (40°N). These instruments began monitoring at these locations in 1994 and 2000, respectively.

[4] Prior to the Nd:YAG lidar operation at Mauna Loa, a ruby laser-based lidar was operated beginning in about 1975. While the ruby lidar was inferior in terms of power and shot frequency it provided an adequate record of stratospheric aerosol in the northern sub-tropics [Barnes and Hofmann, 1997], in particular the effects of the major volcanic eruptions of El Chichón in 1982 and Pinatubo in 1991.

#### 2.1. Seasonal Variation

[5] A seasonal variation in the stratospheric aerosol backscatter, peaking in winter, was observed at Mauna Loa Observatory from the Nd:YAG lidar data after the decay of the Pinatubo aerosol in 1996 [Barnes and Hofmann, 2001]. Figure 1 shows seasonal average aerosol backscatter ratio profiles from approximately weekly lidar soundings at Mauna Loa Observatory and Boulder. The backscatter ratio is defined as the ratio of the total backscatter (aerosol plus the molecular backscatter) to the molecular backscatter. For each sounding, the molecular scattering is determined from daily National Weather Service radiosonde soundings of pressure and temperature at Hilo, HI and Denver, CO for the two sites. The seasonal variation is clear in Figure 1 with the winter backscatter ratio from aerosols alone (the Backscatter Ratio in Figure 1 minus 1) being about 60% larger than the summer aerosol backscatter ratio at 25 km.

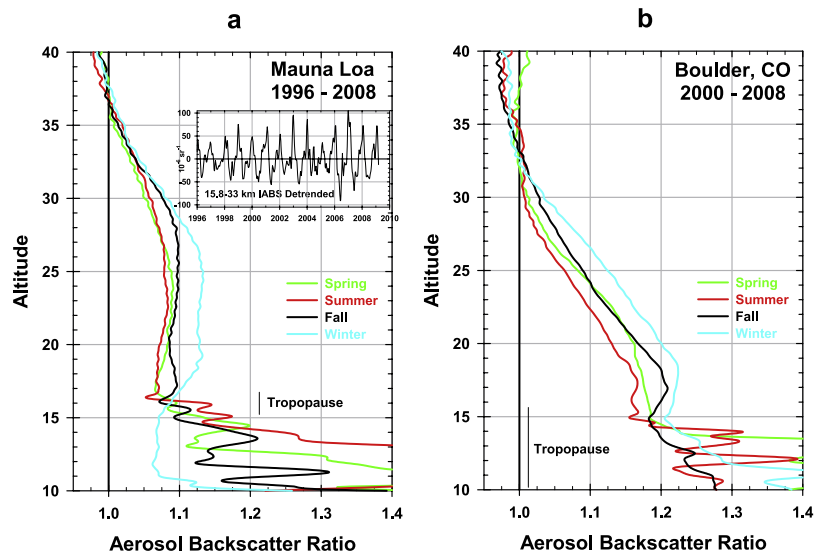
#### 2.2. Decadal Trends

[6] Two minor volcanic events briefly affected the 20 km region during the 2000–2008 period. These were from the Tungurahua (Ecuador – July, 2006) eruption observed at Mauna Loa and the Kasatochi (Alaska – August, 2008)

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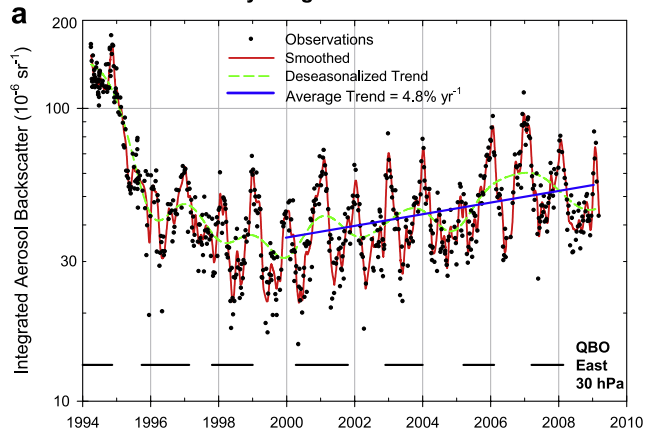


**Figure 1.** Seasonal average aerosol backscatter ratio profiles at (a) Mauna Loa Observatory and (b) Boulder, Colorado. The backscatter ratio is defined as the ratio of the total backscatter (aerosol plus molecular backscatter) to the molecular backscatter. A ratio of 1.0 indicates pure atmospheric molecular scattering. The inset in Figure 1a shows the seasonal cycle amplitude versus time.

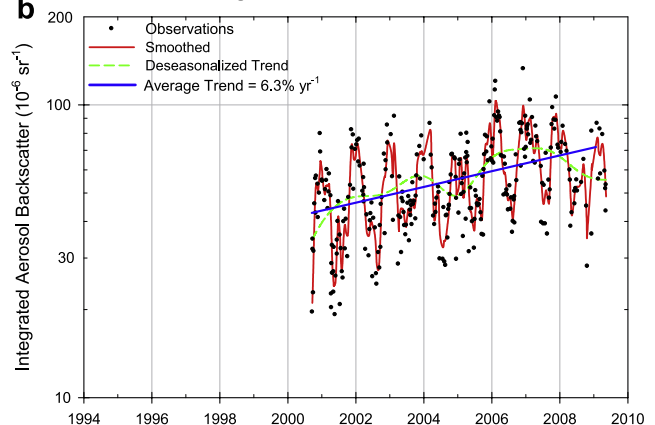
eruption observed at Boulder. Three soundings shortly after each of these events were not included in the trend data. Figure 2a shows the Mauna Loa Observatory Nd:YAG lidar 20–25 km integrated backscatter data from 1994, when the lidar began operating, to early 2009. The data have been analyzed using the technique of *Thoning et al.* [1989] to smooth the data, remove the seasonal variation, and determine the trend curve and growth rate (determined by differentiating the deseasonalized trend curves). There is a biennial component in the deseasonalized trend in Figure 2a, likely related to the quasi-biennial oscillation (QBO) in tropical winds, as will be discussed later. From 1994 to 1996 the decay of aerosol from the 1991 Pinatubo eruption dominates the data [Barnes and Hofmann, 1997]. From 1996 to 2000 there was a slightly decreasing trend at Mauna Loa, possibly due to remnants of the Pinatubo eruption. However, after 2000 there is a decidedly increasing aerosol backscatter trend. The magnitude of the aerosol backscatter trend at Mauna Loa Observatory varies with altitude. The maximum trend occurs in the 20–25 km region with an average value of 4.8% per year, and about 3.3% per year for the total column for the 2000–2009 period (the standard error in determining these trends is about  $\pm 5\%$  of the trend value). Figure 2b, for the 20–25 km range at Boulder, indicates an increasing average trend of 6.3% per year for the 2000–2009 period.

[7] It is important to note that the seasonal increase in aerosol backscatter (summer to winter) is about 2.5 times larger than the backscatter magnitude of the 2000–2009 trend. Therefore, the trend would be difficult to detect by any method that cannot resolve the seasonal variation. We are not aware of other surface-based or satellite lidar or satellite limb extinction instruments that have reported observing the background aerosol seasonal variation or a long-term trend. Finally, since 1996, the peak-to-peak magnitude of the detrended, smoothed annual cycle at Mauna

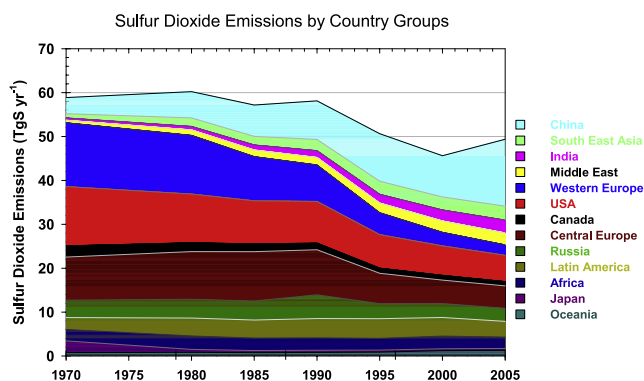
**Mauna Loa Observatory Integrated Lidar backscatter 20–25 km**



**Boulder Integrated Lidar Backscatter 20–25 km**



**Figure 2.** Integrated backscatter for the 20–25 km altitude range at (a) Mauna Loa Observatory and (b) Boulder, Colorado.



**Figure 3.** Sulfur dioxide emissions by country groups. Data are from Smith (personal communication, 2009).

Loa has been increasing at a rate of  $5 \pm 2\%$  per year (see inset in Figure 1a).

### 3. Discussion

#### 3.1. QBO Relation

[8] As indicated earlier, there is a quasibiennial (QBO) signal in the trend curves. While the aerosol backscatter magnitude appears to peak around the middle of the easterly phase (see Figure 2a), a consistent relationship between the aerosol backscatter *growth rate* at high altitude (25–33 km) and the QBO has been observed at both Mauna Loa and Boulder. The aerosol backscatter growth rate consistently increases during the westerly phase and decreases during the easterly phase. This is likely related to an enhancement in aerosol mass from the tropical deep convection region during the westerly phase of the QBO.

#### 3.2. Backscatter Conversion

[9] Lidar backscatter can be converted to aerosol mass, extinction (optical depth) and particle surface area given the aerosol size distribution and assuming liquid spherical droplets, as is appropriate for the stratospheric aerosol. *Jäger and Hofmann* [1991] used balloon-borne stratospheric aerosol particle counter profiles to calculate the lidar conversions for both background and volcanic aerosol conditions and for several wavelengths and altitude intervals. Average one sigma errors for these estimates are about 7, 11, and 25% for mass, extinction and surface area, respectively. The seasonal variation in integrated backscatter at Mauna Loa Observatory for the total (15.8–33 km column) background aerosol of about  $1 \times 10^{-4} \text{ sr}^{-1}$  requires an aerosol column mass source of about  $2 \times 10^{-3} \text{ g m}^{-2}$  to replenish each year. It represents an optical depth change of about 0.006, a change in radiative forcing of about  $-0.3 \text{ W m}^{-2}$ , using the conversion given for sulfate by *Charlson et al.* [1992], and an average particle surface area of  $1.3 \mu\text{m}^2 \text{cm}^{-3}$  in the total column, increasing to  $2.2 \mu\text{m}^2 \text{cm}^{-3}$  in the 20–25 km column.

[10] By assuming global uniformity an estimate of the required source strength can be obtained, which is about 0.4 TgS per year for a 75%  $\text{H}_2\text{SO}_4$ , 25%  $\text{H}_2\text{O}$  aerosol, as is characteristic of the background aerosol composition [*Rosen*, 1971]. If the influx to the stratosphere were not globally uniform, for example, owing to the geographical distribution of tropical deep convection [*Jiang et al.*, 2004],

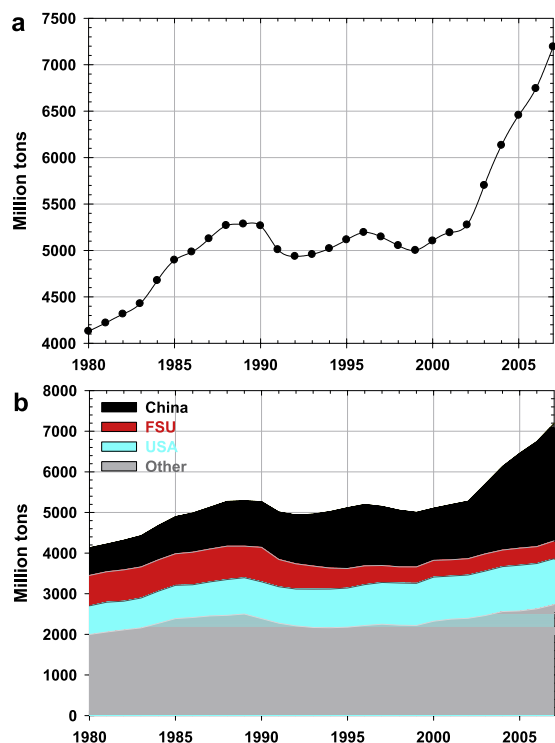
then a value somewhat less than 0.4 TgS per year would be required. A range of 0.3–0.4 TgS per year is appropriate. For comparison, the sulfur flux required to balance the background stratospheric aerosol in a three-dimensional global aerosol model (*Kjellström*, 1998) was 0.21 TgS per year. In that model, about 85% of the sulfur that entered the stratosphere came from tropospheric  $\text{SO}_2$  and only about 10% came from COS. Although the lifetime of  $\text{SO}_2$  in the stratosphere is 30–40 days, it is believed to be only a few days in the troposphere. If not injected explosively by volcanoes, the sulfur from  $\text{SO}_2$  for the non-volcanic background aerosol likely enters the stratosphere in an oxidized form ( $\text{HSO}_3$ ,  $\text{H}_2\text{SO}_4$ , or  $\text{SO}_4$ ).

[11] The increase in total (15.8–33 km) backscatter owing to the aerosol increase observed at Mauna Loa Observatory from 2000 to early 2009 is about  $4.3 \times 10^{-6} \text{ sr}^{-1}$  per year or an optical depth (radiative forcing) change of about 0.0003 per year ( $0.015 \text{ W m}^{-2}$  per year). The required mass flux is  $8.6 \times 10^{-5} \text{ g m}^{-2}$  per year of  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  aerosol or about 0.015 – 0.02 TgS per year globally.

#### 3.3. Source of the Aerosol Increase

[12] There are at least three mechanisms that can affect the stratospheric aerosol level, major volcanic activity, increased tropical upwelling (Brewer-Dobson circulation), and an increase in sulfurous gases in the troposphere. Since there has not been a major volcanic eruption that perturbed the global stratosphere since 1991, the first mechanism can be ruled out in explaining the 2000–2009 aerosol increase. The second mechanism, increased circulation, has been proposed to explain a decrease in stratospheric water vapor of the order of 10% after about 2001 [*Randel et al.*, 2006] associated with a colder tropopause ( $-1 \text{ K}$ ). The cause was proposed to be an increase of about 20% in the Brewer-Dobson circulation. Satellite water vapor data and radiosonde temperature data indicated that the change occurred abruptly in the beginning of 2001 and while this cannot explain the more gradual change in aerosol backscatter after 2000, it cannot be ruled out as a contributing factor. Related to changes in stratospheric water vapor are changes in the  $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$  aerosol weight percent. However, the water vapor change in 2001 was small and in the wrong direction to cause aerosol swelling and increased backscatter. Long-term cooling of the stratosphere related to climate change or ozone depletion, on the other hand, would affect the vapor pressure under which the aerosol droplets grow, but is probably a secondary effect.

[13] The third mechanism, an increase in sulfurous gases in the troposphere, would affect the stratospheric aerosol through deep convection in the tropics. While COS is a component of the background stratospheric aerosol [*Crutzen*, 1976] with a sulfur input of about 0.03 TgS per year [*Chin and Davis*, 1995], its concentration did not increase between 2000 and 2006 [*Montzka et al.*, 2007]. Aircraft emissions of  $\text{SO}_2$  have been proposed as a possible source of stratospheric sulfate [*Hofmann*, 1991]. The magnitude of this source was estimated to be about 0.06 TgS per year in 1992 [*Hofmann*, 1991; *Intergovernmental Panel on Climate Change*, 1999], which would have increased to about 0.10 TgS per year by 2008. Figure 3 shows  $\text{SO}_2$  emissions by country groups and although emissions have declined since about 1980 in Europe and the U.S., emissions in China, Southeast Asia, India and



**Figure 4.** (a) World coal consumption from 1980 to 2007. Since 2002, coal consumption has increased dramatically. (b) The increase in world coal consumption after 2002 is mainly occurring in China. FSU = Former Soviet Union. Data are from the U.S. Energy Information Administration. 1 million tons = 0.907 Tg.

the Middle East increased after 1980 (S. J. Smith, personal communication, 2009). China's annual  $\text{SO}_2$  emissions (all sources) increased from 9.4 to 15.3 TgS from 2000 to 2005 (an average increase of 1.2 TgS or 10.2% per year). The 50 TgS global total emissions in 2005 suggests that about 0.6–0.8% of the sulfur would need to enter the stratosphere to sustain the 0.3–0.4 TgS aerosol seasonal variation.

[14] We can extend the  $\text{SO}_2$  record by using the burning of coal as a proxy for  $\text{SO}_2$  emissions. Coal burning is responsible for 70–80% of global anthropogenic  $\text{SO}_2$  emissions with the highest percentage in China and East Asia at 85% [Smith *et al.*, 2001]. As indicated in Figure 4a, world coal consumption began increasing about 2002 and has shown no letup through 2007. Most of this increase is occurring in China, as Figure 4b indicates. We can estimate the sulfur emission index for coal burning in China. In 2000, 1163 Tg of coal produced about 8.2 TgS or about 0.007 gS/gCoal. The increase in coal consumption for the 2002–2007 period was 1740 Tg (36.3%), mostly in China, giving an estimated increase in  $\text{SO}_2$  emissions due to coal burning of about 2.4 TgS per year or a 5.2% per year increase since 2002. The inferred increase compares favorably with the 4–7% per year increase in stratospheric background aerosol backscatter observed at Mauna Loa Observatory and Boulder from 2000–2009. This would suggest that of the 2.4 TgS per year increase, about 0.6–0.8% (0.015–0.02 TgS per year required to explain the lidar increasing trend) reaches the stratosphere for aerosol formation, the same fraction required from the global sulfur

burden in order to sustain the stratospheric background aerosol seasonal variation, which is expected since the same process is responsible.

[15] While detailed global  $\text{SO}_2$  emissions beyond 2005 are not available, future Chinese emissions estimates are given in a Technical Assistance Report [Asian Development Bank, 2008]. Chinese coal production accounted for 38% of global production in 2005; this figure is projected to increase to 49% by 2030. Chinese coal demand increased from 446 Mt in 1980 to 1,563 Mt in 2005, and is projected to increase to 3,462 Mt by 2030. The 2000–2005 Chinese five year plan targeted an  $\text{SO}_2$  emission reduction of 10%; instead, emissions increased by 27% (5.4%/yr).

[16] The value estimated earlier for the  $\text{SO}_2$  emission rate increase from coal consumption (mainly in China) of 2.4 TgS (5.2%) per year from 2002 to 2007 is similar to the reported increase in Chinese  $\text{SO}_2$  emissions of 5.4% per year from 2000–2005. It is projected that China's coal use will double in about 22 years or 3.2% per year [Asian Development Bank, 2008]. If this happens without controls on  $\text{SO}_2$  emissions it could result in a doubling of the 2000 “normal” background stratospheric aerosol mass by 2022 (about  $2.4 \times 10^{-4} \text{ sr}^{-1}$  integrated lidar backscatter, optical depth of about 0.015), or a level of about 5% of the Pinatubo volcanic aerosol maximum. Climate effects include an increase in solar backscatter resulting in a small tropospheric cooling effect while the ozone layer would be slightly depleted through increased heterogeneous chemistry. Based on observed perturbations following the Pinatubo eruption, the order of magnitude is estimated to be only about a 0.03 °C reduction in tropospheric temperatures and about a 0.1% reduction in stratospheric ozone.

#### 4. Conclusions

[17] Lidar measurements of stratospheric aerosol at Mauna Loa Observatory and Boulder, Colorado suggest the following:

[18] 1) Background stratospheric aerosol conditions have existed since about 1996 (12 years).

[19] 2) There is a large annual cycle (> 50%) in the aerosol backscatter above 20 km with a maximum in winter, requiring 0.3–0.4 TgS per year to sustain.

[20] 3) There is a QBO effect at high altitude with increasing aerosol backscatter growth rate during the westerly phase.

[21] 4) There is an increasing average trend in aerosol backscatter above 20 km after 2000 of about 4–7% per year (0.015–0.02 TgS  $\text{yr}^{-1}$ ).

[22] 5) An increase in global coal consumption, mainly in China, is estimated to have resulted in increased  $\text{SO}_2$  emissions of about 5.2% per year from 2002 to 2007, and is proposed to be the main source of the background stratospheric aerosol increase since about 2002.

[23] 6) China's estimates of future coal use without removal of  $\text{SO}_2$  could result in a doubling of the “normal” 2000 level of background stratospheric aerosol by about 2022, resulting in a small perturbation to tropospheric temperatures and stratospheric ozone with a stratospheric background aerosol level comparable to about 5% of the Pinatubo volcanic aerosol maximum.

[24] 7) Both the seasonal cycle and the long-term trend require about 0.6 – 0.8% of tropospheric sulfur to enter the stratosphere each year.

[25] These results suggest that a renewed modeling effort should be made to determine the background sulfate aerosol in the stratosphere including the latest SO<sub>2</sub> emission data, tropical deep convection, complete tropospheric sulfur chemistry, and aerosol growth under changing vapor pressures in the stratosphere in order to delineate important mechanisms.

[26] **Acknowledgments.** The lidar observations are partially supported through the U.S. National Oceanic and Atmospheric Administration (NOAA) Climate Program. Updated IPCC SO<sub>2</sub> emission data were kindly provided by Steve Smith. Discussions with Terry Deshler and Susan Solomon are appreciated.

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