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## Recent anthropogenic increases in SO<sub>2</sub> from Asia have minimal impact on stratospheric aerosol

R. R. Neely III,<sup>1,2,3</sup> O. B. Toon,<sup>1,4</sup> S. Solomon,<sup>5</sup> J.-P. Vernier,<sup>6,7</sup> C. Alvarez,<sup>2,3</sup> J. M. English,<sup>8</sup> K. H. Rosenlof,<sup>2</sup> M. J. Mills,<sup>8</sup> C. G. Bardeen,<sup>8</sup> J. S. Daniel,<sup>2</sup> and J. P. Thayer<sup>9</sup>

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[1] Observations suggest that the optical depth of the stratospheric aerosol layer between 20 and 30 km has increased 4–10% per year since 2000, which is significant for Earth's climate. Contributions to this increase both from moderate volcanic eruptions and from enhanced coal burning in Asia have been suggested. Current observations are insufficient to attribute the contribution of the different sources. Here we use a global climate model coupled to an aerosol microphysical model to partition the contribution of each. We employ model runs that include the increases in anthropogenic sulfur dioxide (SO<sub>2</sub>) over Asia and the moderate volcanic explosive injections of SO<sub>2</sub> observed from 2000 to 2010. Comparison of the model results to observations reveals that moderate volcanic eruptions, rather than anthropogenic influences, are the primary source of the observed increases in stratospheric aerosol. **Citation:** Neely R. R. III., O. B. Toon, S. Solomon, J.-P. Vernier, C. Alvarez, J. M. English, K. H. Rosenlof, M. J. Mills, C. G. Bardeen, J. S. Daniel, and J. P. Thayer (2013), Recent anthropogenic increases in SO<sub>2</sub> from Asia have minimal impact on stratospheric aerosol, *Geophys. Res. Lett.*, 40, 999–1004, doi:10.1002/grl.50263.

### 1. Introduction

[2] The stratospheric aerosol layer, also known as the Junge Layer, plays a key role in the radiative balance and chemistry of the atmosphere [Junge *et al.*, 1961]. The layer has long been thought to be largely composed of sulfuric acid droplets produced from gas precursors [carbonyl sulfide (OCS), dimethyl sulfide (DMS), sulfur dioxide (SO<sub>2</sub>)]

emitted at the surface by natural and anthropogenic sources with occasional increases associated with colossal volcanic eruptions such as the 1991 Mt. Pinatubo eruption [Thomason and Peter, 2006; Newhall and Self, 1982]. Recently, organic aerosol, meteoritic smoke, and black carbon were found to comprise a small but significant portion of stratospheric aerosol as well [Murphy *et al.*, 1998, 2007; Neely III *et al.*, 2011]. Recent work suggests the importance of the variability of stratospheric aerosol in global radiative forcing, even in periods not dominated by colossal volcanic eruptions [Solomon *et al.*, 2011]. Specifically, as much as 25% of the radiative forcing driving global climate change from 2000 to 2010 may have been counterbalanced by the increases in stratospheric aerosol loading over this period [Solomon *et al.*, 2011]. Stratospheric aerosol also impacts heterogeneous chemistry involving catalytic cycles affecting the ozone layer [Solomon *et al.*, 1998]. Given the importance of the stratospheric aerosol for climate and ozone chemistry, a delineation of natural and anthropogenic contributions to stratospheric aerosol variability is needed.

[3] Recent studies using ground-based lidar and satellite instruments document an increase in stratospheric aerosol of 4–10% per year from 2000 to 2010 [Vernier *et al.*, 2011a; Hofmann *et al.*, 2009; Nagai *et al.*, 2010; Trickl, 2010]. Notably, this period is the first with extensive observations in the absence of any colossal volcanic eruptions since the layer was first observed [Junge *et al.*, 1961; Hofmann *et al.*, 2009]. As such, it provides a unique window to understand the background processes responsible for the maintenance of the stratospheric aerosol layer. The years from 2000 to 2010 span a period when observations and inventories indicate that China and India increased emissions of SO<sub>2</sub> by ~ 60% (Supporting Information and Figure S1) [Smith *et al.*, 2011]. It should be noted that the increase in emissions from China and India also dominates the global increase in SO<sub>2</sub> since 2000 [Smith *et al.*, 2011]. Due to the proximity of China and India to the Asian monsoon region, a gateway for tropospheric pollution to the stratosphere, the increase in Asian emissions is a plausible source of the observed trend [Park *et al.*, 2004, 2007; Hofmann *et al.*, 2009; Randel *et al.*, 2010; Vernier *et al.*, 2011b].

[4] In contrast, satellite observations have recently shown that the increase in stratospheric aerosol was linked to the combined impact of several moderate tropical volcanic eruptions that each injected on the order of a megaton or less of SO<sub>2</sub> into the lower stratosphere [Vernier *et al.*, 2011a]. Until recently, only eruptions involving many megatons of SO<sub>2</sub> were thought to be capable of significantly perturbing the stratospheric aerosol layer, so this finding was a major change in the understanding of the sources of aerosol

Additional supporting information may be found in the online version of this article.

<sup>1</sup>Department of Atmospheric and Oceanic Sciences, University of Colorado, Boulder, Colorado, USA.

<sup>2</sup>NOAA, Earth System Research Laboratory, Boulder, Colorado, USA.

<sup>3</sup>Cooperative Institute for Research in Environmental Sciences, Boulder, Colorado, USA.

<sup>4</sup>Laboratory for Atmospheric and Space Physics, University of Colorado, Boulder, Colorado, USA.

<sup>5</sup>Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, Cambridge, Massachusetts, USA.

<sup>6</sup>Science Systems and Applications, Inc., Hampton, Virginia, USA.

<sup>7</sup>NASA, Langley Research Center, Hampton, Virginia, USA.

<sup>8</sup>Earth System Laboratory, National Center for Atmospheric Research, Boulder, Colorado, USA.

<sup>9</sup>Department of Aerospace Engineering Sciences, University of Colorado, Boulder, Colorado, USA.

Corresponding author: R. R. Neely III, Department of Atmospheric and Oceanic Sciences, University of Colorado, Boulder, CO, USA. (Ryan.Neely@colorado.edu)

loading of the stratosphere [Hofmann *et al.*, 2009; Vanhellemont *et al.*, 2010; Vernier *et al.*, 2011a]. While the volcanic signal is clear in the satellite data, it is not possible from these observations to completely rule out or quantify anthropogenic influences in the total observed aerosol variability in the middle and upper stratosphere.

[5] As a significant factor in climate, it is crucial to disentangle the role of the various sources of stratospheric aerosol to fully comprehend and separate the natural and anthropogenic contributions to climate. This is of particular importance in the northern hemisphere, where SO<sub>2</sub> from large anthropogenic sources can potentially impact the stratosphere via troposphere-to-stratosphere transport processes which include convective lofting, slow radiative ascent and isentropic transport [Vernier *et al.*, 2011b; Park *et al.*, 2004, 2007; Randel *et al.*, 2010; Smith *et al.*, 2011].

## 2. Method

[6] The Asian monsoon is a transport mechanism that is variable from year to year, making estimates of transport of sulfate aerosol into the stratosphere through this pathway is difficult to quantify without the use of a global circulation model. It has been proposed that the only way to quantify the separate sources of the observed increases is through modeling the background sulfate aerosol in the stratosphere [Hofmann *et al.*, 2009]. The model used in this study includes the latest SO<sub>2</sub> emission data, sulfur chemistry, and aerosol growth under changing vapor pressures in the stratosphere in order to delineate important mechanisms.

[7] We isolate the main anthropogenic and volcanic sources using a three-dimensional general circulation model with sulfur chemistry (Whole Atmosphere Community Climate Model, version 3; WACCM3) coupled to a sectional aerosol microphysical model (Community Aerosol and Radiation Model for Atmosphere, CARMA) [Turco *et al.*, 1979; Toon *et al.*, 1979, 1988; Garcia *et al.*, 2007; Bardeen, 2008; Mills *et al.*, 2008; English *et al.*, 2011; Neely III *et al.*, 2011]. (Please see Supporting Information for more details on the model and optical calculations used to make comparisons with the observations.) English *et al.* [2011] and Neely III *et al.* [2011] document the ability of the model to accurately depict the processes responsible for the stratospheric aerosol layer, including the essential role of surface emissions of SO<sub>2</sub> in accurately modeling the lower stratospheric aerosol layer.

[8] To examine the trends, perturbations representative of the increases in anthropogenic emissions in China and India (Supporting Information, Figure S1) and the observed moderate volcanic injections (Supporting Information, Table S1) of SO<sub>2</sub> from 2000 to 2010 were used in conjunction with the standard emission scheme of the model, which was based on observations from 2000 [Smith *et al.*, 2011; English *et al.*, 2011; Neely III *et al.*, 2011]. The uncertainty, described by the 5–95% confidence interval, in the inventory estimate of the emissions of SO<sub>2</sub> from China and India in 2000 is ~7.6 Tg [Smith *et al.*, 2011]. Modeled eruptions were based on detailed observations of injection height and SO<sub>2</sub> total column concentration for each eruption (Supporting Information, Figure S2, Figure S3, and Table S1). Thus, modeled sulfate aerosol, anthropogenic and volcanic, evolves freely from the supplied sources of SO<sub>2</sub> as defined by the chemical and microphysical schemes

represented in the model. The model runs are done with a free running model, not forced by observed winds. Hence, we do not expect perfect inter-annual or seasonal agreement between the model and observations because transport from the locations of the eruptions is important in determining the aerosol abundances.

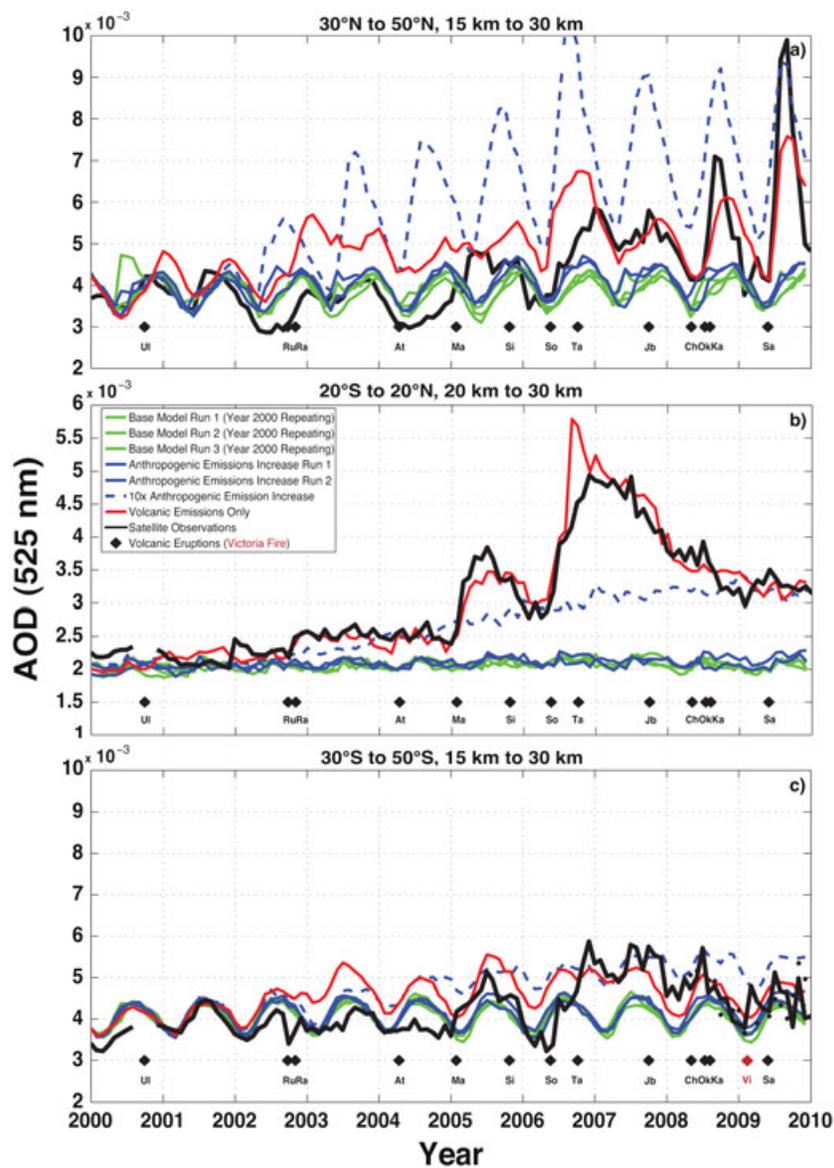
## 3. Result

[9] Figure 1 shows the collective results from the modeling experiment along with observations of stratospheric aerosol optical depth (AOD, black) (integrated from extinction profiles from 20 to 30 km in the tropics and 15 to 30 km in the mid-latitudes) averaged monthly from 2000 to 2010 from space-borne instruments including SAGE II (2000–2005), GOMOS (2002–2010), and CALIOP (2006–2010) [Vernier *et al.*, 2011a]. Below 20 km, data sets, which are contaminated by ice clouds, are filtered [Vernier *et al.*, 2009; Thomason and Vernier, 2012]. While the use of different data sets can produce artificial trends due their inhomogeneity, overlapping periods between those data were used to verify their consistency. The differences between the simulations and observations are obvious—the simulation with volcanic eruptions (red) depicts temporal trends of stratospheric AOD most similar to the observations, while simulations with only the increases in anthropogenic surface emissions (blue) do not appreciably alter stratospheric AOD. Simulated tropical stratospheric AOD (Figure 1b) from volcanic eruptions most closely follows the observations, while mid-latitude AOD (Figure 1a and 1c) has some divergence, but it is clear that the optical depths due to the imposed volcanic emissions are more similar to the observations than the runs with the anthropogenic emissions alone.

[10] In each of the panels of Figure 1, the AOD in the anthropogenic runs is indiscernible from the baseline emission simulations (green), though the model run with ten times (10×), the anthropogenic emission increase shows increases in aerosol comparable to the volcanic emissions. This unrealistically enhanced anthropogenic emission run is included only as a demonstration of the response of the stratospheric aerosol layer to an order of magnitude of change in Asian surface emissions. If SO<sub>2</sub> emissions increased by this amount, ~120 Tg, China and India, combined, would be emitting SO<sub>2</sub> at levels comparable to total global emissions in the 1970s [Smith *et al.*, 2011; Granier *et al.*, 2011].

[11] In Figure 2, we show the vertical cross-section of zonally averaged volume extinction ratio of the aerosol observed by satellite (Figure 2a) and simulated in a volcanic model run with no increase in anthropogenic emissions (Figure 2b) for the tropics (20°S to 20°N) from 2000 to 2010. Extinction ratio is defined as the ratio between the particulate and molecular extinction and may be interpreted as a measure of aerosol mixing ratio. This comparison further reveals the ability of moderate volcanic eruptions to influence the distribution of aerosol in the stratosphere, with far larger impact than anthropogenic sources.

[12] A similar simulation, done in conjunction with the ones described in this study, where all anthropogenic emissions of SO<sub>2</sub> were turned off, and the model was allowed to equilibrate, suggest that all global anthropogenic emissions in 2000 contribute ~50% of stratospheric AOD in

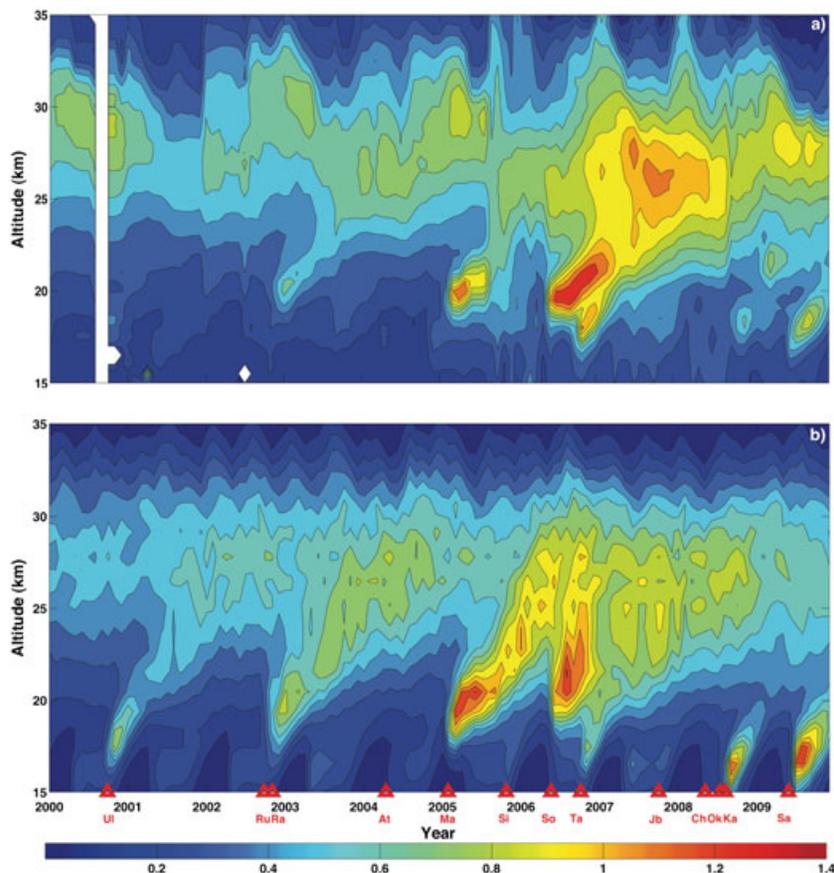


**Figure 1.** Observed and modeled time series of stratospheric AOD from three latitude bands. a) Monthly averaged AOD at 525 nm from 30°N to 50°N integrated from 15 km to 30 km, b) 20°S to 20°N integrated from 20 km to 30 km, c) 30°S to 50°S integrated from 15 km to 30 km from satellite observations (black line) from SAGE II (2000 to August 2005), GOMOS (March 2002 onward) and CALIOP (April 2006 onward) and the simulation results. Baseline model runs are in green. Model runs with the increase in anthropogenic emissions from China and India are in blue. The dashed blue line depicts a model run with 10× the actual increase in anthropogenic emissions. The model run with volcanic emissions is in red. The black diamonds and initials along the bottom of the plot represent the volcanic eruptions that were included in the model run (See Supporting Information, Table S1 for further information). Figure 1c also denotes the Victoria Fire in red.

the tropics from 20 km to 30 km. The remainder is contributed by OCS, tropospheric volcanic injections. (Compared to OCS and SO<sub>2</sub>, dimethyl sulfide (DMS) is a small source of stratospheric aerosol and is not considered in this model.)

[13] Additional model runs where the model was allowed to reach equilibrium with all anthropogenic emissions on except for China and India suggest that emissions of SO<sub>2</sub> from China and India contribute ~17% of AOD in the tropical stratosphere from 20 km to 30 km. According to *Smith et al.* [2011], the global emission of SO<sub>2</sub> in 2000 was ~111 Tg while China and India combined emitted ~27 Tg (i.e., ~25% of global emissions). In 2005, these values

change to ~120 Tg for global emissions with China and India contributing ~39 Tg combined (~32% of global emissions) [*Smith et al.*, 2011]. Hence, China and India drove emissions up by ~9 Tg between 2000 and 2005, against a fall in the rest of the world by ~3 Tg. So the expected increase in AOD due to the emissions from China and India less the decrease in the rest of the world is ~4% over 5 years. A 4% change in the stratospheric AOD is much smaller than observed, or simulated when volcanic emissions are included. Thus, it is not expected that the small increase in anthropogenic emissions from China and India, against the falling emissions in the rest of the world, would have a measurable impact on the stratospheric AOD. This assessment



**Figure 2.** Evolution of monthly mean extinction ratio (525 nm) profiles in the tropics (20°N–20°S) from January 2000 to December 2009 derived from a) SAGE II, GOMOS, and CALIOP, b) a model run with the input from the volcanoes listed in Table S1. The position of each volcanic eruption occurring during the period is displayed with its first two letters on the horizontal axis.

is consistent with the modeling results shown in Figure 1, as the model runs including the increased emissions from China and India show no trends that exceeds the variability displayed by the baseline model runs. This result suggests that the anthropogenic contribution from 2000 to 2010 is much smaller than the background variability of the upper majority of the stratospheric aerosol layer.

#### 4. Discussion

[14] While Figure 1 and 2 show remarkable agreement in many features of the stratospheric aerosol record, some differences are also apparent. Among these are the spike seen in the observations shown in Figure 1c at the beginning of 2009. Previous work has explained this increase in AOD as a result of the Victoria Fire in Australia during February 2009, which resulted in a pyrocumulonimbus event that injected biomass burning aerosol into the stratosphere [Vernier *et al.*, 2011a]. Such events were not included in the model runs shown here because they are thought to add to the observed aerosol variability without dominating the overall trend.

[15] Larger deviations between the observations and the volcanic model run, in Figures 1a and 1c relative to Figure 1b, may be explained by the fact that WACCM was not forced to match the observed dynamics from 2000 to 2010. As a specific example, in Figure 1a, the volcanic model run for the Northern mid-latitudes does not match

well when the volcanic injections are in the tropics such as in 2006. Though the Jebel at Tair (Jb) eruption in 2007 does match well, this may be due to the confluence of aerosol from the previous eruptions and the seasonal cycle. Yet, Figure 1a does a much better job of matching the large changes in aerosol caused by the northern mid-latitude eruptions in 2008 and 2009 which have no impact on the tropics or southern hemisphere. This is due to the fact that these eruptions directly impact the northern hemisphere and are little influenced by the differences in transport between the model and observations. However, the aerosols from tropical eruptions are sensitive to transport into the Northern mid-latitude. In general, in Figure 1c simulations and data are closer than in Figure 1a, but this is mostly due to the reduced amount of aerosol variability in general in the southern hemisphere.

[16] Similar transport related discrepancies in volcanic aerosol simulations are seen in English *et al.* [2013] for the Mt. Pinatubo eruption, a large well documented event. This paper also shows that the data often have large error bars that are hard to quantify. For example, the CALIOP optical depths involve conversion factors, which are particle size dependent. More significantly, the tropical simulations are not very sensitive to transport, because the Pinatubo injection was in the tropics. However, mid-latitude simulations depend on the transport out of the equatorial zone, which is known to be episodic.

[17] Year to year variability in transport should also be considered. This issue is reflected in how much of the aerosol is moved out of the zone where it is initially deposited, where it is moved to and when it is moved (in both the observations and model runs). This is the most likely reason that the results from the mid-latitudes (Figures 1a and 1c) are not as good as the results taken from the tropics (Figure 1b).

[18] The purpose of this modeling effort was not to accurately duplicate the observations at all latitudes and times, which would have required using observed winds from each year. Rather, our objective was to compare the stratospheric aerosol variability when forced by a series of realistic volcanic injections of SO<sub>2</sub> into the stratosphere with variability arising from sources representative of increases in surface emissions of SO<sub>2</sub> from China and India. The volcanic simulation in Figure 1b was not expected to match the observations to such a high degree and the agreement may be somewhat fortuitous. The minor deviations between mid-latitude results (Figure 1a and 1c) met our expectations due to the known differences in transport, limitations imposed on these simulations due to the coarse model grid (4° × 5°) and the sensitivity of the resulting aerosol load to the imperfectly known height of the volcanic injections. Nevertheless, the key point is that the variability in the volcanic simulations and the relatively large optical depths reached following eruptions much more closely matches the observations in all regions examined than the variability and peak optical depths related to anthropogenic SO<sub>2</sub> emission changes of any of the other realistic simulations.

## 5. Conclusion

[19] The results of these simulations unambiguously show that moderate volcanic eruptions are the main drivers of stratospheric aerosol variability from 2000 to 2010 as observed by Vernier *et al.* [2011a]. They also indicate that the middle and upper stratosphere (above the region of isentropic transport from the troposphere and containing the bulk of the ozone layer) is not measurably impacted by increased anthropogenic emissions of SO<sub>2</sub> in Asia from 2000 to 2010, though these emissions are part of a global source. Due to the variability of the lowermost stratosphere, the small signal of sulfate aerosol produced from anthropogenic sources and the incomplete treatment of tropospheric aerosol by this model, an assessment of anthropogenic influence on the lower-most stratosphere (the region of isentropic transport between the troposphere and stratosphere) is not shown. The significant portion of the radiative forcing due to increases in stratospheric aerosol from 2000 to 2010, interpreted as a mechanism of global cooling [Solomon *et al.*, 2011], may now be completely attributed to volcanic sources and should not be considered a trend. Rather, the stratospheric aerosol layer should be treated as a natural source of radiative forcing that is continuously perturbed by volcanic injections of a range of sizes, and potentially other sources such as large fires. Due to the small signal of the anthropogenic contribution to the stratosphere, future modeling is needed, specifically examining the lower stratosphere, that looks much more closely at the details of the model and its ability to correctly treat the movement of SO<sub>2</sub> into the stratosphere, so that such

small signals may be reliably distinguished from the natural variability of the aerosol layer.

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