### Major influence of tropical volcanic eruptions on the stratospheric aerosol layer during the last decade

J.-P. Vernier,<sup>1,2</sup> L. W. Thomason,<sup>1</sup> J.-P. Pommereau,<sup>2</sup> A. Bourassa,<sup>3</sup> J. Pelon,<sup>2</sup> A. Garnier,<sup>2</sup> A. Hauchecorne,<sup>2</sup> L. Blanot,<sup>2,4</sup> C. Trepte,<sup>1</sup> Doug Degenstein,<sup>3</sup> and F. Vargas<sup>5</sup>

Received 25 March 2011; revised 21 April 2011; accepted 30 April 2011; published 30 June 2011.

[1] The variability of stratospheric aerosol loading between 1985 and 2010 is explored with measurements from SAGE II, CALIPSO, GOMOS/ENVISAT, and OSIRIS/Odin space-based instruments. We find that, following the 1991 eruption of Mount Pinatubo, stratospheric aerosol levels increased by as much as two orders of magnitude and only reached "background levels" between 1998 and 2002. From 2002 onwards, a systematic increase has been reported by a number of investigators. Recently, the trend, based on ground-based lidar measurements, has been tentatively attributed to an increase of SO<sub>2</sub> entering the stratosphere associated with coal burning in Southeast Asia. However, we demonstrate with these satellite measurements that the observed trend is mainly driven by a series of moderate but increasingly intense volcanic eruptions primarily at tropical latitudes. These events injected sulfur directly to altitudes between 18 and 20 km. The resulting aerosol particles are slowly lofted into the middle stratosphere by the Brewer-Dobson circulation and are eventually transported to higher latitudes. Citation: Vernier, J.-P., et al. (2011), Major influence of tropical volcanic eruptions on the stratospheric aerosol layer during the last decade, Geophys. Res. Lett., 38, L12807, doi:10.1029/2011GL047563.

### 1. Introduction

[2] Eruptions of Volcanic Explosivity Index (VEI) larger than 5 (e.g., Mount Pinatubo in June 1991) have the potential to inject large amounts of sulfur, primarily in the form of sulfur dioxide (SO<sub>2</sub>), into the stratosphere. The SO<sub>2</sub> is oxidized into H<sub>2</sub>SO<sub>4</sub>, which, after homogeneous nucleation and/or condensation onto existing aerosol, results in an increase in the mass of liquid sulfate aerosols that can persist for years [*SPARC*, 2006]. The enhanced aerosol layer can have a significant impact on the radiative and chemical processes of the atmosphere and can lead to ozone destruction [*McCormick et al.*, 1995; *Hofmann and Solomon*, 1989]. For example, large temperature changes were observed after the 1991 Mount Pinatubo eruption that led to a warming of the stratosphere by about 1°C and a cooling of the global surface

<sup>4</sup>ACRI-ST, Sophia-Antipolis, France.

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temperatures in the following few years [*Robock*, 2000]. As the aerosols are removed by sedimentation and stratospheretroposphere exchange [*Hamill et al.*, 1997], the stratosphere gradually approaches a non-volcanic background condition [*Deshler et al.*, 2006]. In the absence of volcanic events, the existence of a permanent stratospheric aerosol or Junge layer [*Junge et al.*, 1961] is generally attributed to sulfuric gas precursors (SO<sub>2</sub>, OCS, DMS) emitted at the surface, further oxidized (more or less rapidly) during their transport into sulfuric acid, and finally transformed by homogeneous nucleation into a H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O liquid aerosol mixture [*Brock et al.*, 1995].

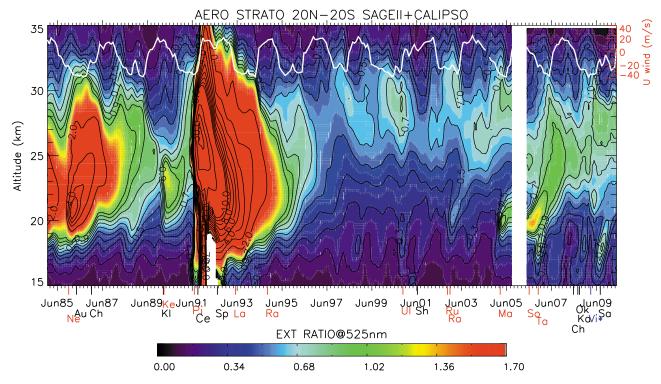
[3] Several periods of near-background aerosol conditions have been identified since the advance of reliable stratospheric aerosol observations. These include 1976 to 1981 (following the Fuego eruption and before the Mount St Helens/El Chichòn eruptions), 1989 to 1991 (ending with the Mount Pinatubo eruption), and post-1998. However, a detailed analysis that accounted for the long recovery from volcanic events did not reveal a statistically significant trend at any altitude or latitude in this 30-year period [Deshler et al., 2006]. More recently, ground-based lidar measurements at Mauna Loa, Hawaii, Boulder, Colorado [Hofmann et al., 2009], Lauder, New Zealand [Nagai et al., 2010] and Gadanki, India [Kulkarni et al., 2008] have been used to infer a trend in aerosol backscatter of 4-7%/yr since 2000 in the altitude range of 20 to 30 km. As no large volcanic eruptions have occurred since Mount Pinatubo in 1991, the trend was attributed by Hofmann et al. [2009] to an increase of SO<sub>2</sub> emission from coal burning in Southeast Asia, especially in China, conveyed into the stratosphere by transport processes associated with deep convection [Notholt et al., 2005; Randel et al., 2010]. Hofmann et al. [2009] specifically excluded a volcanic contribution to the trend even though a number of small eruptions were reported throughout this period. For example, a significant eruption of VEI 4 occurred on 20 May 2006 at Soufriere Hills in Montserrat which was observed by the CALIPSO lidar. These measurements showed a plume near 19-20 km, followed by its slow ascent in the tropical stratosphere as it was carried by the Brewer-Dobson circulation [Vernier et al., 2009]. Other stratospheric volcanic events were also reported by SAGE II [Thomason et al., 2008] and GOMOS [Vanhellemont et al., 2010] following the eruption of Ruang in Indonesia in September 2002, Reventador in Ecuador in November 2002 and Manam in Papua New Guinea in January 2005. These observations suggest that the stratospheric impact by smaller eruptions (VEI  $\leq$  4) is not uncommon. In this study, we use a set of global satellite observations including CALIPSO, SAGE II, GOMOS and OSIRIS to show that the contribution of medium VEI events

<sup>&</sup>lt;sup>1</sup>NASA Langley Research Center, Hampton, Virginia, USA.

<sup>&</sup>lt;sup>2</sup>LATMOS, CNRS, INSU, Université de Versailles Saint Quentin, Université de Paris 6, Guyancourt, France.

<sup>&</sup>lt;sup>3</sup>Institute of Space and Atmospheric Studies, University of Saskatchewan, Saskatoon, Saskatchewan, Canada.

<sup>&</sup>lt;sup>5</sup>Institute for Research and Development, Paraiba Valley University, São José dos Campos, Brazil.



**Figure 1.** Monthly mean extinction ratio (525 nm) profile evolution in the tropics [20°N-20°S] from January 1985 to June 2010 derived from (left) SAGE II extinction in 1985–2005 and (right) CALIPSO scattering ratio in 2006–2010, after removing clouds below 18 km based on their wavelength dependence (SAGE II) and depolarization properties (CALIPSO) compared to aerosols. Black contours represent the extinction ratio in log-scale from 0.1 to 100. The position of each volcanic eruption occurring during the period is displayed with its first two letters on the horizontal axis, where tropical eruptions are noted in red. The eruptions are listed in Table 1. Superimposed is the Singapore zonal wind speed component at 10 hPa (white line).

to the increase in the stratospheric aerosol load over the last decade is significant.

## 2. Aerosols Measurements in the Tropical Stratosphere From Space-Borne Platforms

[4] Stratospheric aerosols have been monitored since the early 1970's [SPARC, 2006; Deshler et al., 2006] by (i) solar occultation with the Stratospheric Aerosol and Gas measurements (SAGE) series of instruments [SPARC, 2006], (ii) a network of ground-based lidars that are a part of the Network for Detection of Atmospheric Composition Change (NDACC) and (iii) balloon borne Optical Particles Counters [Deshler et al., 2003]. Since 2002, those measurements have been complemented by a new generation of satellite instruments that include limb scatter measurements made by the Optical Spectrograph and InfraRed Imaging System (OSIRIS) onboard the Odin satellite [Llewellyn et al., 2004], stellar occultation measurements made by the Global Ozone Monitoring by Occultation of Stars (GOMOS) on ENVISAT [Vanhellemont et al., 2010] and by the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) aboard the CALIPSO platform since 2006 [Winker et al., 2010].

[5] Figure 1 shows a composite time history of the monthly mean Extinction Ratio (ER) profile at 525 nm in the tropics (20S to 20N) since 1985 based on SAGE II and CALIPSO measurements. The ER, which is the ratio between the particulate and molecular extinction and is analogous to an aerosol mixing ratio, was constructed using

SAGE II 525-nm aerosol extinction coefficient (1985–2005) and CALIPSO backscatter measurements (2006–2010) using a 532-nm lidar ratio (i.e., the aerosol extinction-tobackscatter ratio). This ratio varies between 30 and 70 sr as a function of latitude and altitude and was determined using collocated CALIPSO backscatter values and GOMOS extinction between 2006 and 2009 then applied throughout the whole period of the CALIPSO measurements (see auxiliary material).<sup>1</sup> These values are in good agreement with theoretical calculations [*Chazette et al.*, 1995] given the ~10% uncertainty in the retrieved CALIOP extinction coefficients and optical depths. On Figure 1, we have superimposed the Singapore zonal wind speed component at 10 hPa (white line) which is indicative of the phase of the Quasi-Biennial Oscillation (QBO).

[6] Several strong volcanic events can be seen between 1985 to 1995 (see Table 1) including: Nevado del Ruiz in Colombia in November 1985, Kelut in Indonesia in February 1990, and the dominant event of the past 30 years: the VEI 5– 6 Mount Pinatubo (Philippines) eruption in June 1991. The Pinatubo eruption resulted in an increase of the aerosol loading by a factor of 100 at altitudes up to 35 km. Several eruptions with stratospheric influence occurred during this period, including Cerro Hudson in October 1991 and Rabaul in Papua New Guinea in September 1994, but their less

<sup>&</sup>lt;sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2011GL047563.

**Table 1.** List of Volcanic Eruptions of VEI  $\geq$  4 Between 1984 and 2010 as Reported by the U.S. Geological Survey<sup>a</sup>

Volcano	Date	Latitude	VEI
Nevado del Ruiz (Ne)	14-Nov-85	5°S	4-5 ?
Augustine (Au)	27-Mar-86	59°N	4 ?
Chikurachki (Ch)	20-Nov-86	50°N	4
Kliuchevskoi (Kl)	30-Jan-90	56°N	4
Kelut (Ke)	10-Feb-90	8°S	4
Pinatubo (Pi)	15-Jun-91	15°N	5-6
Cerro Hudson (Ce)	12-Aug-91	46°S	5+
Spur (Sp)	27-Jun-92	61°N	4
Lascar (La)	19-Apr-93	23°S	4
Rabaul (Ra)	19-Sep-94	4°S	4 ?
Ulawun (Ul)	29-Sep-00	5°S	4
Shiveluch (Sh)	22-May-01	56°N	4 ?
Ruang (Ru)	25-Sept-02	2°N	4 ?
Reventador (Ra)	3-Nov-02	0°N	4
Manam (Ma)	27-Jan-05	4°S	4
Soufrière Hills (So)	20-May-06	16°N	4 ?
Tavurvur (Ta)	07-Oct-06	4°S	4 ?
Chaiten (Ch)	2-May-08	42°S	4 ?
Okmok (Ok)	12-juil-08	55°N	4
Kasatochi (Ka)	07-Aug-08	55°N	4
Fire/Victoria (Vi*)	07-Feb-09	37°S	
Sarychev (Sa)	12-Jun-09	48°N	4 ?

<sup>a</sup>VEI, Volcanic Eruption Index. Report from U.S. Geological Survey available at http://www.volcano.si.edu/reports/usgs/. Volcanoes in the tropics are in boldface.

intense plumes are effectively masked by the long-lived Pinatubo-derived aerosol enhancement. As shown in Table 1, there was no eruption of VEI 4 or greater reported between 1994 and 2000. The largest event during this period was the VEI 3 eruption of Shishaldin in 1999 [Rizi et al., 2000]. Between 2000 and 2002, only a few volcanic events with very minor stratospheric impact occurred, including Ulawun in September 2000 and Shiveluch in May 2001. The overall stratospheric aerosol loading between 1998 and 2002, shown in Figure 1, is relatively steady with no discernable volcanic enhancement. We consider this to be the only quasi-background aerosol period during the SAGE II lifetime (1984–2005). During this quiescent period, the altitude of the highest ER values in the stratospheric aerosol layer is 4–5 km higher than observed after volcanic eruptions. The maximum ER density of the Junge layer is modulated by the QBO such that it reaches a maximum altitude, around 28-30 km, during its easterly phase [Trepte and Hitchman, 1992]. Note the maximum altitude of the layer during the entire period occurs in summer 2005 near 30 km and may be related to the increased tropical upwelling (Brewer-Dobson circulation) after 2001 suggested by Randel et al. [2006]. The aerosol loading at altitudes below 20 km shows a seasonal modulation that has been attributed to the cleansing of the lower stratosphere by convective overshooting of clean air in the Southern tropics during the SH summer season [Vernier et al., 2011a]. This contrasts with the transport of additional aerosols in the Northern tropics during the Asian Monsoon in the NH summer [Vernier et al., 2011b].

[7] The background aerosol period ends in 2002 with the beginning of a series of tropical volcanic eruptions with small but significant stratospheric impact. These are noted in Table 1 and include: Ruang in September 2002 and Reventador in November 2002 [*Thomason et al.*, 2008], followed by Manam in January 2005 [*Vanhellemont et al.*, 2010], Soufriere Hills in May 2006 [*Prata et al.*, 2007]. Additional volcanic

plumes, followed by the eruptions of Okmok, Kasatochi [*Bourassa et al.*, 2010] and Sarychev [*Haywood et al.*, 2010] respectively in July 2008, August 2008 and June 2009, can be seen in the tropics after their horizontal transport from the Northern polar region. Another aerosol enhancement can be seen in February 2009 at 22 km. Those aerosols are not of volcanic origin but instead are associated with the "Black Saturday" forest fire in Southern Australia on 7 February 2009 which was lofted into the stratosphere [*Trepte et al.*, 2009].

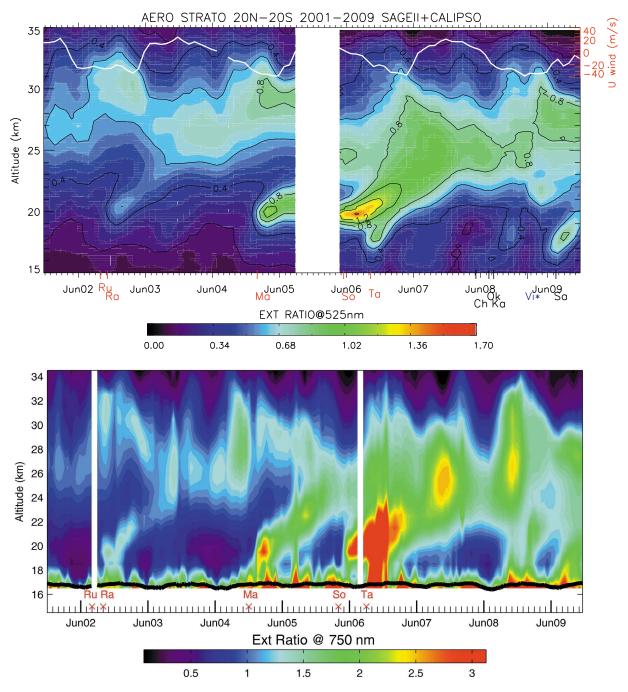
# 3. Impact of Minor Volcanic Eruptions on the Aerosol Loading of the Tropical Stratosphere

[8] For a better characterization of the impact of volcanic plumes due to minor eruptions on the stratospheric aerosol load, we show in Figure 2 (top) an enlargement in 2002-2009 of the SAGE II/CALIPSO extinction ratio of Figure 1 together with the Odin-OSIRIS extinction ratio at 750 nm during the same period (Figure 2, bottom). The picture is dominated by the relatively large volcanic plume from Soufriere Hills that slowly rises up into the middle stratosphere within one year. Other plumes from Ruang/Reventador and Manam show very similar behavior. Although of lower vertical resolution, the OSIRIS extinction ratio confirms the SAGE/CALIPSO picture associated with i) the stratospheric aerosol layer between 22-34 km ii) the modulation of the top altitude of the Junge layer by the QBO, and iii) the slow ascent of the plumes lofted within one year from the altitude of their injection up to 25 km. Despite its episodic nature, the overall behavior of the aerosol transport process is strongly reminiscent of the well-known water vapor tape recorder [Mote et al., 1998]. It should be noted that some differences exist between OSIRIS and SAGE II/CALIPSO in the middle stratosphere during the winters 2005, 2007 and 2008. They are very likely associated with the OSIRIS aerosol retrieval process that requires the assumption of an aerosol scattering phase function. The scattering phase function is a relatively weak function of stratospheric aerosol particle size at 750 nm [McLinden et al., 1999], but an increase in particle size associated with the volcanic perturbations will systematically impact the retrieved extinction from the limb scatter measurements.

[9] Overall, these minor eruptions are seen to have a clear impact on the tropical stratospheric aerosol load. As an example, the amount of SO<sub>2</sub> injected in the stratosphere by the Soufriere Hills was estimated to be 0.1 TgS ( $10^9$  g of sulfur) by the Ozone Monitoring Instrument (OMI) onboard AURA [*Prata et al.*, 2007]. Assuming that SO<sub>2</sub> is entirely transformed into gaseous H<sub>2</sub>SO<sub>4</sub> and further condensed into a 75%-25% H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O solution, the total aerosol mass added by this single volcanic event would be 0.25 TgS. This is ten times more than the 0.01–0.02 Tg S/year required to explain the average aerosol increase of 4–7%/year after 2002 estimated by *Hoffman et al.* [2009]. Moreover, injections from the other smaller volcanic injections events in the last decade in the tropics are of a similar magnitude.

#### 4. Aerosol Trend Since 2000

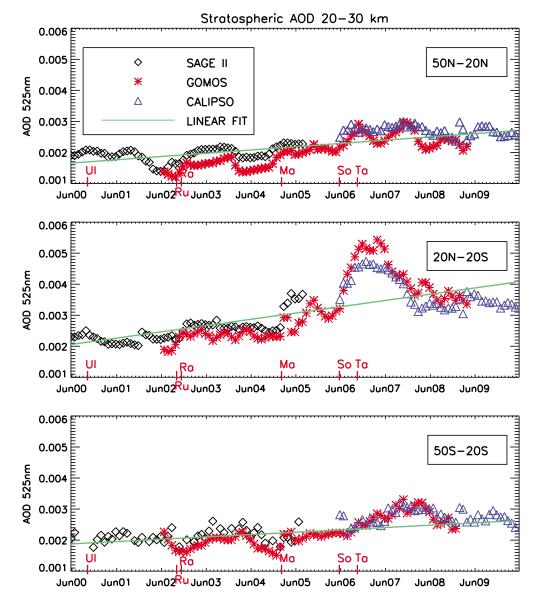
[10] Figure 3 shows the evolution of the Stratospheric Aerosol Optical Depth (SAOD) at 525-nm integrated from 20 to 30 km. It was derived from SAGE II, GOMOS and



**Figure 2.** (top) Enlargement of Figure 1 in 2002–2009. (bottom) Odin/OSIRIS aerosol extinction at 750 nm zonal monthly mean profile at 20N-20S since the beginning of the mission.

CALIPSO within the 50°N-20°N (Figure 3, top), 20°N-20°S (Figure 3, middle) and 20°S-50°S (Figure 3, bottom) latitude bands. The GOMOS SAODs are consistent with those of SAGE II before 2005 and those inferred from CALIPSO after 2006. Between 20°S-20°N in the tropics, the stratospheric aerosol load does not display a linear trend but instead a sequence of impulses of various amplitudes and durations associated with the small volcanic eruptions. The persistence of the volcanic signature is enhanced by the 1 to 2 years required for the ascent of the aerosol to 25 km by the upwelling branch of the Brewer-Dobson circulation. Compared to the tropics, the mid-latitude SAOD records do

show smoother increases with enhancements of smaller amplitude delayed by an additional year due to the time required for transport from the tropics to mid-latitude. This is illustrated by the series of monthly mean zonal averages of CALIPSO scattering ratio following the Soufriere Hills eruption shown in Figure 4. The first in July 2006 (Figure 4a) shows the dense volcanic plume of the Soufriere Hills at 19–21 km with the remnants of the Manam eruption aerosols between 22 and 27 km. Six months later (Jan-07, Figure 4b), the Soufriere Hills plume has ascended to the stratosphere and is mostly confined in the tropical reservoir [*Plumb*,1996; *Hitchman et al.*, 1994]. During the same period, the plume

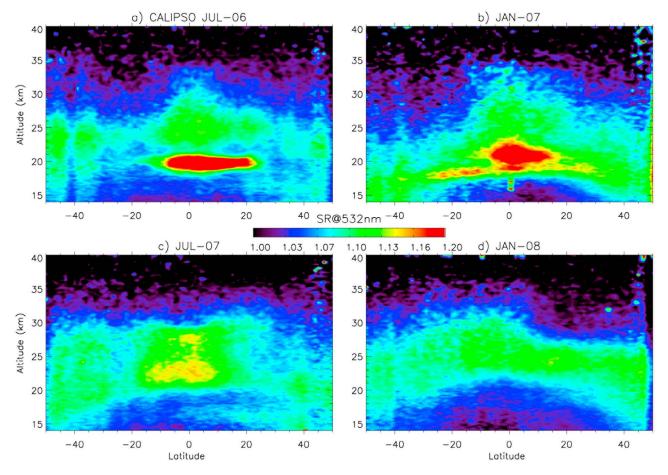


**Figure 3.** Monthly mean stratospheric 525 nm Aerosol Optical Depth (AOD) evolution between 20–30 km since 2000 from SAGE II (black diamonds), CALIPSO (blue triangles), GOMOS (red stars); (top) 50°N-20°N, (middle) 20°N-20°S and (bottom) 20°S-50°S. Linear fits over the all period are plotted in green. The rate of increase in stratospheric AOD deduced from the linear fits is  $10.9 \times 10^{-5}$ /year for the latitude band 50°N-20°N,  $20.4 \times 10^{-5}$ /year for 20°N-20°S and  $7.7 \times 10^{-5}$ /year for 50°S-20°S.

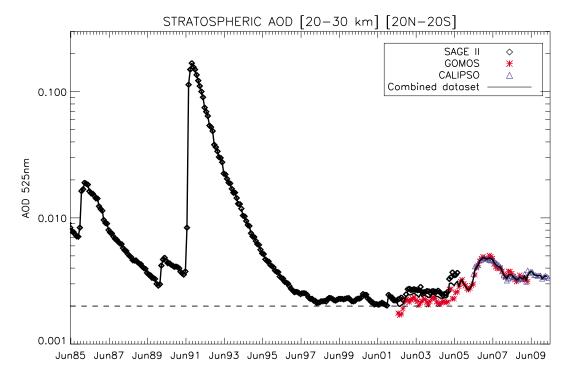
at lower levels that followed the eruption of Tavurvur in October is rapidly transported towards mid-latitudes. In July 2007 (Figure 4c), the tropical stratospheric reservoir is filled by aged volcanic aerosol, spanning from 22 to 28 km. The release of those aerosols at mid-latitudes is dictated by the circulation in the form of the so-called "horns" (Figure 4c) typically observed during westerly phase of the QBO [*Trepte and Hitchman*,1992]. Additionally, the Annual Oscillation also drives the transport of aerosol toward mid-latitudes alternatively southward during NH summer (Figure 4c) and northward during NH winter (Figure 4d). As a consequence of these transport of the Soufriere Hills plume toward mid-latitudes shown in Figure 3 is delayed by several months and spread over a longer period than in the tropics.

[11] The apparent trends of 5–10%/yr shown by the linear fits in Figure 3 are consistent with the estimates of *Hofmann et al.* [2009] and *Nagai et al.* [2010]. However, they are mainly explained by the sequence of volcanic eruptions in the tropics (Figures 1 and 2) followed by the dilution and the time required for the transport of their plumes to the mid-latitudes (Figure 4).

[12] The 25-years record of stratospheric aerosol optical depth reconstructed from SAGE II, GOMOS and CALIPSO displayed in Figure 5 shows that after the major eruptions of the Nevado del Ruiz and Pinatubo volcanoes in 1986 and 1991, the stratospheric aerosol reached a quasi stable background level of 0.002-0.0025 optical thickness until 2002. Afterward, the burden of stratospheric aerosol has increased mainly due to the impact of tropical volcanic



**Figure 4.** Monthly mean of the CALIPSO zonal scattering ratio at 532 nm in (a) Jul 2006, (b) Jan 2007, (c) Jul 2007 and (d) Jan 2008. Cloudy pixels in the upper troposphere are removed using the depolarization channel.



**Figure 5.** Mean stratospheric Aerosol Optical Depth in the tropics [20°N-20°S] between 20–30 km since 1985 from SAGE II (black diamonds), GOMOS (red stars), CALIPSO (blue triangles) and combined satellites (black line).

eruptions of VEI 4. Although the last decade was thought to be an ideal period to study the effect of anthropogenic emission of sulfur on the stratospheric aerosol layer, these satellites observations demonstrate that the changes in the stratospheric aerosol load have been mainly driven by tropical volcanic eruptions.

### 5. Conclusion

[13] A 4–7%/yr increase in the stratospheric aerosol load has been reported from ground-based lidar observations between 2000-2009. This was tentatively attributed to increased SO<sub>2</sub> emissions from coal burning in Southeast Asia, particularly China. However, the SAGE II, GOMOS, OSIRIS, and CALIPSO aerosol measurements show that the increase is largely due to a series of volcanic eruptions of medium explosive index (VEI  $\leq 4$ ) occurring after a quiescent period between 1998-2002. The plumes injected by these volcanoes at around 18-20 km altitude in the lower stratosphere are lofted into the mid-stratosphere up to around 25 km within one year by the upwelling tropical branch of the Brewer-Dobson circulation, resulting in a tape recorder like feature of the aerosol evolution. Compared to the 0.002-0.0025 stratospheric aerosol optical depth (AOD) reported during the 1998-2002 minimum, which is indicative of the possible contribution of natural and anthropogenic gas precursors, minor volcanic eruptions are shown to enlarge the AOD by 0.002-0.004 depending on the year. We do not exclude the possibility that a human-derived component to stratospheric aerosol trends exists. However, given the subtle but clear volcanic influence to stratospheric aerosol levels, a human-derived trend to stratospheric aerosol cannot be inferred and any such trend must be significantly smaller than those reported by Hofmann et al. [2009].

[14] Acknowledgments. This work was support by the NASA Post-doctoral Program at Langley Research Center, administrated by Oak Ridge Associated Universities. It is also a part of the PhD thesis of JPV financed by the Centre National la Recherche Scientifique at LATMOS/UVSQ. CALIOP and ECMWF data were made available at the ICARE data center (http://www-icare.univ-lille1.fr/) and processed at NASA LaRC. The authors want to thank the Meteorological Institute at Free University of Berlin for providing the zonal wind at Singapore used to study the QBO (http://www.geo.fu-berlin.de/en/met/ag/strat/produkte/qbo/index.html). The authors want also to thank people in charge of the ground-based lidars for their discussion, including, C. David, J. Jumelet and P. Keckhut for OHP (France), J.E Barnes for Boulder and Mauna Loa (US), T. Nagai for Lauder (New Zeland) and H. Gadhavi for Gadanki (India). The authors also want to acknowledge J. C Lebrun for the processing of the GOMOS stratospheric aerosol dataset and P. Lucker for her help in the processing of the CALIPSO data. Work on OSIRIS is funded in part by the Canadian Space Agency.

[15] The Editor thanks two anonymous reviewers for their assistance in evaluating this paper.

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L. Blanot, ACRI-ST, 260 Rte. du Pin Montard, BP 234, F-06904 Sophia-Antipolis CEDEX, France.

A. Bourassa and D. Degenstein, Institute of Space and Atmospheric Studies, University of Saskatchewan, 116 Science Pl., Saskatoon, SK S7N 5E, Canada.

A. Garnier, A. Hauchecorne, J. Pelon, and J.-P. Pommereau, LATMOS, CNRS, INSU, Université de Versailles Saint Quentin, Université de Paris 6, 11 Blvd. d'Alembert, F-78280 Guyancourt CEDEX, France.

L. W. Thomason, C. Trepte, and J.-P. Vernier, NASA Langley Research Center, 23a Langley Blvd., Hampton, VA 23666, USA. (jeanpaul.vernier@ nasa.gov)

F. Vargas, Institute for Research and Development, Paraiba Valley University, Av. Shishima Hifumi, 2911, São José dos Campos, SP 12244-000, Brazil.