



Influence of the 2008 Kasatochi volcanic eruption on sulfurous and carbonaceous aerosol constituents in the lower stratosphere

B. G. Martinsson,¹ C. A. M. Brenninkmeijer,² S. A. Carn,³ M. Hermann,⁴ K.-P. Heue,² P. F. J. van Velthoven,⁵ and A. Zahn⁶

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[1] Influences on stratospheric aerosol during the first four months following the eruption of Kasatochi volcano (Alaska) were studied using observations at 10700 ± 600 m altitude from the CARIBIC platform. Collected aerosol samples were analyzed for elemental constituents. Particle number concentrations were recorded in three size intervals together with ozone mixing ratios and slant column densities of SO_2 . The eruption increased particulate sulfur concentrations by a factor of up to 10 compared to periods before the eruption (1999–2002 and 2005–August 2008). Three to four months later, the concentration was still elevated by a factor of 3 in the lowermost stratosphere at northern midlatitudes. Besides sulfur, the Kasatochi aerosol contained a significant carbonaceous component and ash that declined in time after the eruption. The carbon-to-sulfur mass concentration ratio of the volcanic aerosol was 2.6 seven days after the eruption and reached 1.2 after 3–4 months. **Citation:** Martinsson, B. G., C. A. M. Brenninkmeijer, S. A. Carn, M. Hermann, K.-P. Heue, P. F. J. van Velthoven, and A. Zahn (2009), Influence of the 2008 Kasatochi volcanic eruption on sulfurous and carbonaceous aerosol constituents in the lower stratosphere, *Geophys. Res. Lett.*, 36, L12813, doi:10.1029/2009GL038735.

1. Introduction

[2] Explosive volcanic eruptions can impact climate and endanger aviation by creating elevated concentrations of volcanic ash and sulfate aerosol over large regions for long periods (leading to excessive wear of turbine parts, window crazing). When such eruptions penetrate the tropopause they can increase the aerosol load in the stratosphere on a hemispheric to global scale. This study deals with the August 7–8, 2008 eruption of Kasatochi volcano in Alaska, USA (52.17° N, 175.51° W) and the effect it had on the concentration and composition of aerosol in the lower stratosphere.

[3] The Kasatochi eruption interrupted a period of low volcanic influence on the stratosphere after the waning of effects from the 1991 eruption of Mount Pinatubo. In periods

of low volcanic activity stratospheric sulfate aerosol [Bauman *et al.*, 2003; Martinsson *et al.*, 2005; Deshler, 2008] develops as a result of the Brewer-Dobson circulation. Air from the troposphere containing precursor gases is lifted into the tropical stratosphere, forming sulfate aerosol that is transported diabatically to lower altitudes at mid and high latitudes [Crutzen, 1976; Chin and Davis, 1995]. Volcanic activity is the main cause for variability in stratospheric aerosol concentration, thereby affecting solar radiation through absorption and scattering and inducing heating of the stratosphere and influencing heterogeneous ozone chemistry [Intergovernmental Panel on Climate Change, 2007]. Ammann *et al.* [2003] estimated the climate impact of volcanism during the 20th century, reaching peak radiative forcing of approximately -1 W/m^2 or more during five events. The influence from an eruption persists for a few years in the stratosphere. This negative forcing effect has inspired some researchers to suggest a deliberate loading of the stratosphere with sulfate aerosol to counteract greenhouse warming [Crutzen, 2006]. Model estimates indicate that 1.5 Tg S/yr would, well placed, balance a doubling of the atmospheric CO_2 concentration [Rasch *et al.*, 2008].

[4] Large volcanic eruptions thus induce scientific interest in their stratospheric and climatic effects but also serve as natural experiments for those investigating geo-engineering solutions to the climate problem. In this study of the Kasatochi eruption, in-situ measurements of aerosol constituents and trace gases in the upper troposphere (UT) and lowermost stratosphere (LS) are presented. Most of the measurements were made from the CARIBIC (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container) platform [Brenninkmeijer *et al.*, 2007].

2. Experimental Methods

[5] Monthly *in situ* measurements of aerosol particles and trace gases in the UT/LS are undertaken from the CARIBIC platform (www.caribic-atmospheric.com) during intercontinental flights with a Lufthansa Airbus A340-600 [Brenninkmeijer *et al.*, 2007]. Aerosol samples were collected at 10700 ± 600 m altitude with multichannel aerosol samplers [Nguyen *et al.*, 2006]. The samples were analyzed for elemental constituents by an accelerator-produced beam of 2.555 MeV protons. Samples taken in 1999–2002 were analyzed by particle-induced X-ray emission (PIXE) for sulfur and heavier elements. In a second period, spanning 2005–present, the aerosol samples were analyzed with PIXE and particle elastic scattering analysis (PESA) to obtain in addition concentrations of light elements [Nguyen and Martinsson, 2007]. The accuracy in the elemental determinations is estimated to be 10% [Martinsson *et al.*, 2001;

¹Division of Nuclear Physics, Lund University, Lund, Sweden.

²Division of Atmospheric Chemistry, Max Planck Institute for Chemistry, Mainz, Germany.

³Department of Geological and Mining Engineering and Sciences, Michigan Technological University, Houghton, Michigan, USA.

⁴Leibniz Institute for Tropospheric Research, Leipzig, Germany.

⁵Royal Netherlands Meteorological Institute, De Bilt, Netherlands.

⁶Institute of Meteorology and Climate Research, Forschungszentrum Karlsruhe, Karlsruhe, Germany.

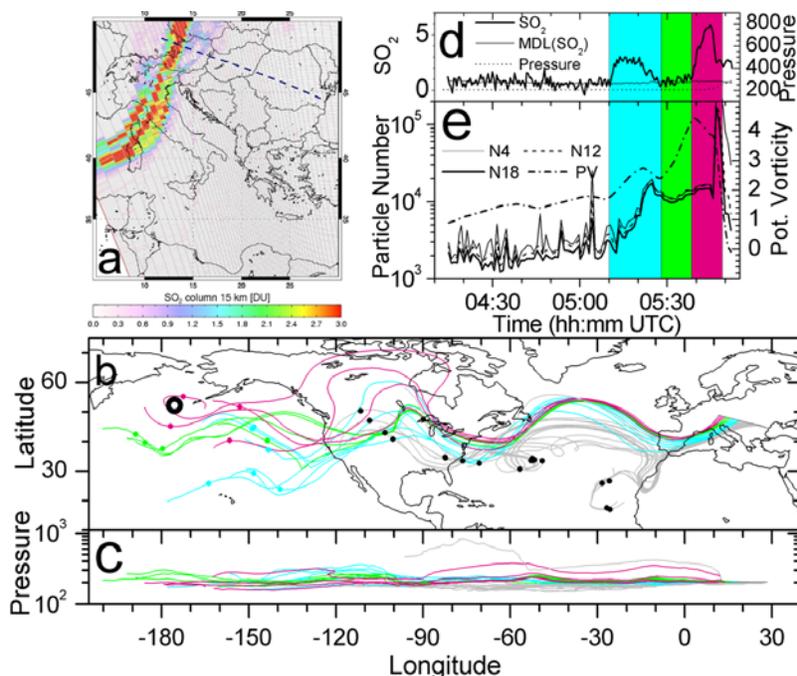


Figure 1. (a) Aura/OMI measurement of SO₂ on August 15, 2008 at 11:51–11:58 UTC (orbit 21730). The dashed line indicates the CARIBIC flight track on the same day, around 6 hours before the OMI overpass. (DU (Dobson Unit): 2.69×10^{16} molecules per cm².) (b and c) Eight-day air-mass back-trajectories (computed for every 3 minutes) to the position of the CARIBIC aircraft for the time period of the aerosol sampling (04:14–05:52 UTC, August 15, 2008) during the first encounter with the volcanic plume. Colors correspond to time periods indicated in Figures 1d and 1e. The circle indicates the location of Kasatochi volcano and dots show the position along the trajectories at the time of the eruption. (d and e) Slant column density of SO₂ (10^{17} molecules/cm²), minimum detection limit (MDL) of SO₂, atmospheric pressure level (hPa), particle number concentrations (cm^{-3} STP) in three size intervals (>4 nm (N4), >12 nm (N12), >18 nm (N18)) and potential vorticity (PV) for the same time period as in Figures 1b and 1c.

Nguyen and Martinsson, 2007]. The analysis was undertaken in a high vacuum at room temperature, implying that compounds of vapor pressure similar to or more volatile than n-tetracosane might in part have escaped analysis [Martinsson, 1987].

[6] Particle number concentrations were measured using three condensation particle counters (CPC) with size intervals: >4 nm (N4), >12 nm (N12) and >18 nm (N18) diameter. The uncertainty is estimated to be 10–35% depending on the size range of interest [Brenninkmeijer et al., 2007]. All particle concentrations are reported at standard temperature and pressure conditions (STP; 273.15 K and 1013.25 hPa). Two systems are used for ozone measurements; an accurate, dual-beam UV-photometer serving as a calibrated standard instrument, and a solid-state chemiluminescence detector giving additional high-resolution data. The accuracy is estimated to be 1% for 10 Hz measurements [Brenninkmeijer et al., 2007]. Slant column densities (SCD) of sulfur dioxide (SO₂) were obtained by a differential optical absorption spectrometer (DOAS) telescope (wavelengths 312–333 nm) pointing -10° relative to the horizon. DOAS is sensitive to air masses close to the aircraft and up to 10 km distant [Brenninkmeijer et al., 2007; Dix et al., 2009].

[7] This study also makes use of SO₂ measurements from the Ozone Monitoring Instrument (OMI) onboard NASA's Aura satellite. OMI is a UV/VIS (270–500 nm) nadir solar backscatter spectrometer in polar orbit. The instrument provides global mapping of O₃, SO₂ and other trace gases

with a nadir spatial resolution of 13×24 km. OMI's UV-2 channel (306–380 nm), which is used for SO₂ retrievals, has an average spectral resolution of 0.45 nm [Levelt et al., 2005].

[8] The origin of probed air masses was investigated using eight-day back trajectories that were calculated each third minute along the CARIBIC flight track based on horizontal and vertical wind components from the European Centre for Medium-Range Weather Forecast (ECMWF). Potential vorticity (PV) at the CARIBIC aircraft location was calculated from temperature and wind fields in archived ECMWF analyses with a resolution of $1 \times 1^\circ$ in the horizontal and 91 hybrid sigma-pressure model levels.

3. Results

[9] The first CARIBIC contact with the Kasatochi volcanic cloud was over central Europe on August 15, 2008, one week after the eruption. High concentrations of aerosol components such as sulfur and carbon indicated an unusual aerosol source. A plume of SO₂ was observed over Europe on the same day by OMI (Figure 1a). Figures 1b and 1c show air mass back trajectories spanning eight days for the CARIBIC aerosol sample collected between 04:14 to 05:52 UTC on August 15, indicating that air from the area of the eruption was sampled for part of the sampling time (cyan/green/red trajectories in Figures 1b and 1c concerning air masses intercepted 05:10–05:50). DOAS measurements onboard the CARIBIC aircraft registered strongly elevated SO₂

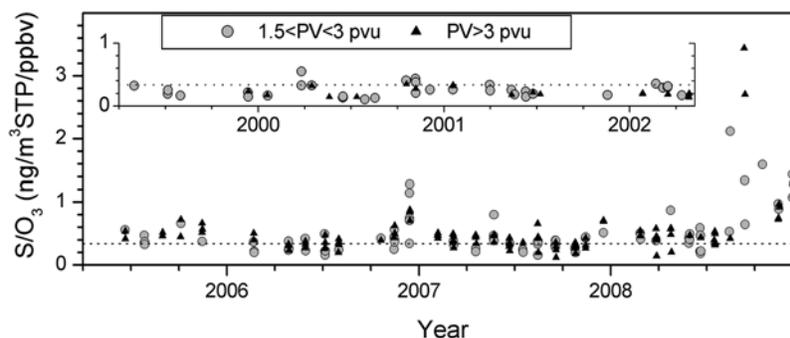


Figure 2. Ratio of the mass concentration of particulate sulfur to the ozone mixing ratio in the tropopause region (circles) and the lowermost stratosphere (triangles) during the time periods of CARIBIC measurements. The dotted line indicates the geometrical mean of the ratio for pre-eruption conditions.

concentrations between 05:10–05:30 and 05:40–05:50 (Figure 1d), thus indicating that the plume was split into two parts. The number concentration of particles larger than 18 nm diameter showed an unusual increase starting at 05:10 as the aircraft passed from lower to higher potential vorticity in the tropopause region (Figure 1e). The dip in particle number concentration between 05:30–05:40 is less clear than for SO_2 . Few particles were recorded in the 4–12 nm size interval, indicating no formation of new particles during the last day [Williams *et al.*, 2002]. About 15% of the particles were between 12–18 nm, indicative of new particle formation or injection during the foregoing week, which agrees well with the timing of the eruption. The actual plume passage most likely lasted approximately 30 minutes of this 98 minute sample.

[10] Stratospheric aerosol during background as well as volcanically influenced conditions contains a large fraction of sulfate aerosol [Deshler, 2008]. Similarly to ozone production, sulfate background aerosol production from carbonyl sulfide requires short-wavelength radiation that is available in the stratosphere. Diabatic, downward transport brings down large quantities of ozone and sulfate aerosol to the lowermost stratosphere, forming strong gradients in ozone [Zahn *et al.*, 2002] and sulfate [Martinsson *et al.*, 2005] concentrations. A correlation between these constituents, due to coincident production regions, thus appears in the lowermost stratosphere, which is evident in samples taken before the eruption. After the Kasatochi eruption, a large fraction of the data points significantly deviated from that correlation with deviations being particularly strong during August and September. In order to capture the volcanic influence on the aerosol concentration, the ratio of particulate sulfur to ozone mixing ratio was calculated. This particulate sulfur/ozone ratio shows moderate variations around its geometrical mean of $0.34 \text{ ng/m}^3\text{STP/ppbv}$ during pre-eruption conditions, 95% and 99% of the observations being smaller than 0.7 and $0.87 \text{ ng/m}^3\text{STP/ppbv}$, respectively. After the eruption this ratio was low for all except one UT sample taken at PV of less than 1.4 PVU. That particular sample was taken in September. None of the samples taken in the UT later during the autumn were notably affected, probably as the result of efficient aerosol removal in the troposphere. In contrast, all samples taken at PV values higher than 1.4 PVU are around or higher than the 95% probability level of the pre-Kasatochi distribution, thus indicating volcanic influence on the particulate sulfur concentration. The varia-

tion in time of the samples taken in the tropopause region (TP; $1.5 < \text{PV} < 3 \text{ PVU}$) and LS ($\text{PV} > 3 \text{ PVU}$) is shown in Figure 2 for the two available CARIBIC measurement periods (1999–2002, 2005–2008). A minor anomaly in the tropopause region in December 2006 indicates an unusual source, which we tentatively identify as Nyamuragira volcano (DR Congo) that erupted on November 27, 2006. The August 2008 Kasatochi eruption strongly influenced the sulfur-to-ozone ratio, which shows large variability in the first month after the eruption. In November and December 2008, the ratio shows less variability, likely as a result of mixing. The average over these two months was $1.04 \text{ ng/m}^3\text{STP/ppbv}$. This corresponds to an increase of the particulate sulfur concentration by a factor of 3.1 over stratospheric background conditions. The samples from the tropopause region and stratosphere were taken during 10 flights between Frankfurt, Germany, and Chennai, India. They cover a broad latitude range over central and eastern Europe and western Asia. Since all samples were affected, it can be concluded that the Kasatochi eruption affected at least the latitude and PV ranges of the observations, which were $25\text{--}50^\circ \text{ N}$ from the tropopause region up to a PV level of at least 5 PVU.

[11] In addition to establishing the extent of the volcanic influences based on the major component (particulate sulfur), the CARIBIC data also provide unique insight into the composition of the aerosol. Concentrations of sulfurous and carbonaceous aerosol are represented by elemental concentrations. Figure 3 shows the cumulative frequency distribution of carbon and sulfur of all pre-eruption CARIBIC measurements in the UT/TP/LS and all post-eruption measurements taken in the TP or LS. It is clear that all previously measured concentrations of carbon and sulfur from the CARIBIC platform were exceeded in the measurements taken seven days after the eruption. Two of the four TP/LS measurements taken one month after the eruption also exceeded all previously observed carbon and sulfur concentrations. Carbon and sulfur concentrations remained elevated throughout the autumn.

[12] The aerosol sample collected seven days after the eruption contained a substantial carbonaceous fraction, being a factor of 2.3 higher in mass concentration than particulate sulfur (C_S). It also contained a component probably originating from volcanic ash represented by silicon ($0.07C_S$) and iron ($0.03C_S$), which was significantly depleted already a month after the eruption. The relative importance of the carbonaceous component declined to on average $1.0C_S$

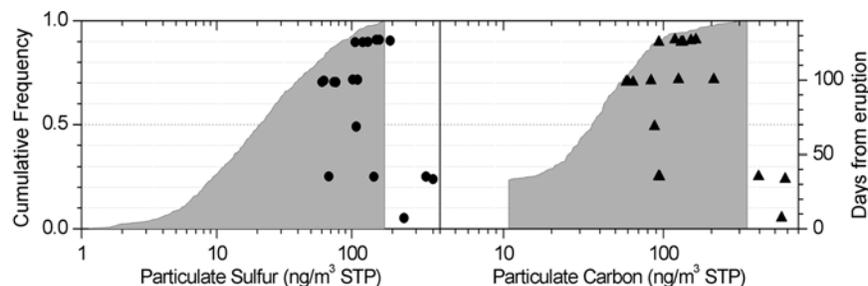


Figure 3. Cumulative frequency distribution of mass concentrations of UT/LS particulate sulfur and carbon for pre-eruption conditions (continuous distribution in gray). The step in the carbon distribution is caused by samples that did not exceed the minimum detection limit. Symbols indicate individual post eruption sample concentrations in the tropopause region and lowermost stratosphere as a function of the number of days elapsed since the eruption (sulfur: circles; carbon: triangles). Note that pre eruption frequency for a post eruption concentration is obtained in the vertical direction of the graph.

3–4 months after the eruption. This corresponds to an average increase of carbon concentration by a factor of 3.6 compared with stratospheric background conditions. The volcanic fraction of the aerosol thus had a carbon-to-sulfur mass concentration ratio of 1.2 after 3 to 4 months. This should be compared to the same ratio shortly after the eruption, which was 2.6. A likely cause for this reduction of the carbon-to-sulfur mass ratio of the volcanic aerosol is conversion of SO_2 to sulfate.

4. Discussion

[13] Making the assumption that all the particulate sulfur was in the form of sulfate would imply a factor of 1.2 and 2.5 excess of sulfate relative to carbon 7 days and 3–4 months after the eruption, respectively. A recent review of stratospheric aerosol describes the composition of volcanic aerosol as predominantly sulfuric acid and water [Deshler, 2008]. The composition of stratospheric aerosol has been studied by various methods including transmission electron microscopic morphology studies [Sheridan et al., 1992] and studies based on volatility [Deshler et al., 1992]. These methods are however non-specific; morphological patterns are difficult to interpret chemically [Nguyen et al., 2008], and vaporization after heating to a given temperature can indicate the presence of a range of compounds. Other studies, based on elemental analysis by X-ray detection in electron microscope, were specific but do not include the carbonaceous fraction in the analysis [Junge et al., 1961]. Strikingly few measurements of stratospheric aerosol include the carbonaceous component. Measurements by Murphy et al. [1998] and Nguyen et al. [2008] do show that stratospheric aerosol contains carbonaceous matter, but they apply to periods with no or little volcanic influence. The lack of measurements after previous eruptions that are sensitive to the carbonaceous component makes it difficult to assess whether the aerosol from the Kasatochi eruption is unusual in its chemical composition.

5. Conclusions

[14] Aerosol in the upper troposphere (UT) and the lowermost stratosphere (LS) was studied from the CARIBIC platform before and after the eruption of the Kasatochi volcano. Whereas the eruption's influence on the UT was

limited, persistent effects on the aerosol load in the tropopause region (TP) and the LS were observed. The ratio of particulate sulfur to the ozone mixing ratio was found to be within narrow bounds during periods of little volcanic influence on the aerosol in the TP/LS. This ratio was considerably elevated after the eruption in all TP and LS observations, covering latitudes 25 to 50° N and a potential vorticity range 1.5–5 PVU. Three to four months after the eruption the average particulate sulfur and carbon concentrations were elevated by factors of 3.1 and 3.6 relative to periods of low volcanic influence. The Kasatochi volcanic aerosol contains a significant fraction of carbonaceous matter, with a carbon-to-sulfur ratio of 2.6 measured one week after the eruption, decreasing to 1.2 after 3–4 months. Due to a lack of detailed aerosol composition measurements from previous volcanic eruptions it remains unclear if the carbonaceous component of the Kasatochi aerosol is unique.

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- C. A. M. Brenninkmeijer and K.-P. Heue, Division of Atmospheric Chemistry, Max Planck Institute for Chemistry, Postfach 3060, D-55020 Mainz, Germany.
- S. A. Carn, Department of Geological and Mining Engineering and Sciences, Michigan Technological University, 1400 Townsend Drive, Houghton, MI 49931-1295, USA.
- M. Hermann, Leibniz Institute for Tropospheric Research, Permoserstrasse 15, D-04318 Leipzig, Germany.
- B. G. Martinsson, Division of Nuclear Physics, Lund University, P.O. Box 118, SE-22100 Lund, Sweden. (bengt.martinsson@nuclear.lu.se)
- P. F. J. van Velthoven, Royal Netherlands Meteorological Institute, P.O. Box 201, NL-3730 AE De Bilt, Netherlands.
- A. Zahn, Institute of Meteorology and Climate Research, Forschungszentrum Karlsruhe, P.O. Box 3640, D-76021 Karlsruhe, Germany.