

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Composition and evolution of volcanic aerosol from eruptions of Kasatochi, Sarychev and Eyjafjallajökull in 2008–2010 based on CARIBIC observations

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Received: 12 April 2012 – Accepted: 4 June 2012 – Published: 22 August 2012

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper | Discussion Paper

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Abstract

Large volcanic eruptions impact significantly on climate and lead to ozone depletion due to injection of particles and gases into the stratosphere where their residence times are long. In this the composition of volcanic aerosol is an important but inadequately studied factor. Samples of volcanically influenced aerosol have been collected following the Kasatochi (Alaska), Sarychev (Russia) and also during the Eyjafjallajökull (Iceland) eruptions in the period 2008–2010. Sampling was conducted by the CARIBIC platform during regular flights at an altitude of 10–12 km as well as during dedicated flights through the volcanic clouds from the eruption of Eyjafjallajökull in spring 2010. Elemental concentrations of the collected aerosol were obtained by accelerator-based analysis. Aerosol from the Eyjafjallajökull volcanic clouds was identified by high concentrations of sulfur and elements pointing to crustal origin, and confirmed by trajectory analysis. Signatures of volcanic influence were also used to detect volcanic aerosol in stratospheric samples collected following the Sarychev and Kasatochi eruptions. In total it was possible to identify 17 relevant samples collected between 1 and more than 100 days following the eruptions studied. The volcanic aerosol mainly consisted of ash, sulfate and included a carbonaceous component. Samples collected in the volcanic cloud from Eyjafjallajökull were dominated by the ash and sulfate component (~45 % each) while samples collected in the tropopause region and LMS mainly consisted of sulfate (50–77 %) and carbon (21–43 %). These fractions were increasing/decreasing with the age of the aerosol. Because of the long observation period, it was possible to analyze the evolution of the relationship between the ash and sulfate components of the volcanic aerosol. From this analysis the residence time ($1/e$) of sulfur dioxide in the studied volcanic cloud was estimated to be 45 days.

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1 Introduction

Despite its modest size, the eruption of Eyjafjallajökull volcano in the spring of 2010 caused considerable disruption of European air traffic due to the ash and sulfate aerosol it produced. In particular ash particles are hazardous, since they can damage jet engines and disrupt avionics and navigation systems (Casadevall, 1994), whereas sulfate aerosol have been reported to cause crazing of aircraft windows (Carn et al., 2009). Above all the complex effects of volcanoes on atmospheric chemistry and physics are also of concern for climate (e.g. Ammann et al., 2003). Not only the amount of ejected material, the location of the volcano and the force of the eruption, but also the properties of volcanic aerosol play a considerable role in these contexts. Unfortunately, our present understanding of the composition of volcanically influenced atmospheric aerosol is limited since quantitative measurements in the free troposphere and in the stratosphere are scarce. To improve this situation, we report here on the investigation of the elemental composition of volcanic aerosol sampled directly in the volcanic cloud from the Eyjafjallajökull eruption in 2010, and moreover sampled in the upper troposphere/lowermost stratosphere (UT/LMS) following eruptions of the volcanoes Kasatochi (Alaska, 2008) and Sarychev (Russia, 2009).

The scattering and absorbing properties of volcanic aerosol affect the Earth's radiation budget and thus result in temperature gradients that perturb circulation patterns and impact climate (IPCC, 2007). After the eruption of Mount Pinatubo in 1991, with a large Volcanic Explosivity Index (VEI) (Newhall and Self, 1982) of 6, the global, tropospheric temperature was estimated to have been 0.5 °C lower than the climatological average temperature. Another large eruption in Tambora (1815, VEI 7) caused an estimated drop in global mean temperature of 0.4 to 0.7 °C (McCormick et al., 1995). Stratospheric aerosols also act as surfaces for heterogeneous reactions that affect the distribution of ozone and other trace gases (IPCC, 2007). Not only massive eruptions such as those mentioned above are of importance to the stratospheric aerosol load. A study by Vernier et al. (2011) based on satellite-borne lidar observations shows

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that eruptions of lower explosivity also are important sources of stratospheric aerosol. Their effect is visible in the increase of the stratospheric aerosol layer that has occurred since 2002 after a period with little volcanic influence. This increase in the stratospheric aerosol load has also been observed in other data sets, however, anthropogenic influence cannot be ruled out (Hofmann et al., 2009; Solomon et al., 2011).

In the absence of volcanic eruptions, stratospheric aerosol is mainly found at altitudes of 20–30 km (Junge et al., 1961). This aerosol layer, referred to as the Junge layer, mainly consists of sulfate aerosol that is chiefly formed by sulfur dioxide (SO₂) produced from photo-dissociation of carbonyl sulfide (OCS) transported from the troposphere (Crutzen, 1976). However studies indicate that OCS is not enough to explain the observed aerosol load (Chin and Davis, 1995), and direct transport of SO₂ or sulfate aerosol have been suggested as important contributions to stratospheric aerosol (Pitari et al., 2002; Myhre et al., 2004). Volcanic injections however makes it difficult to determine the background state of the stratospheric aerosol layer (Solomon et al., 2011), and thereby the importance of different sources for its production. A carbonaceous component of the UT/LMS aerosol was identified by Murphy et al. (1998), which was subsequently found to be a large fraction of the aerosol (Nguyen and Martinsson, 2007). Martinsson et al. (2009) found that volcanic aerosol contains a large carbonaceous component, and recently Friberg et al. (2012) found that volcanic eruptions can be important contributors to the carbonaceous aerosol also during time periods close to background conditions. Additional sources contributing to the aerosol load in the UT/LMS, include air traffic (Ferry et al., 1999; Kjellström et al., 1999), meteorites (Cziczo et al., 2001) and boundary layer aerosol and precursor gases transported across the tropopause (Papaspiropoulos et al., 2002; Köppe et al., 2009). Especially aerosol from forest fires can be brought into the UT/LMS by extreme convection, however, the frequency and global contribution of such events is poorly understood (Fromm et al., 2004, 2008). On the other hand do volcanic eruptions often contribute to stratospheric aerosol mass of similar magnitude as OCS does (Vernier et al., 2011), and in a few

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events per century volcanism is by far the strongest source of stratospheric aerosol (Ammann et al., 2003).

Volcanic eruptions inject large quantities of ash and gases into the atmosphere. Sulfur dioxide is the third most abundant gas in volcanic emissions, after water vapor and carbon dioxide (von Glasow et al., 2009). It is oxidized in the atmosphere thus leading to sulfate aerosol. Enhanced concentrations of stratospheric aerosol following the eruptions of El Chichón (1982) and Pinatubo (1991) had a residence time ($1/e$) of 10.3 and 12.0 months respectively (Jäger, 2005). Especially the number concentration of particles larger than $1\ \mu\text{m}$ diameter was observed to increase substantially following the Pinatubo eruption (Deshler, 2008). The directly emitted ash particles exhibit a size of 2 mm or less (by definition) (Heiken and Wohletz, 1985), and show a large span in size with particle diameters down to less than $1\ \mu\text{m}$ (Rose and Durant, 2009; Mather et al., 2003). Large particles sediment quickly while very fine ash particles ($<15\ \mu\text{m}$) have been found to have a residence time of days to weeks in the UT/LMS (Rose and Durant, 2009; Niemeier et al., 2009). Although research has shown that fine ash particles are spread and deposited over large areas (Rose and Durant, 2009), we know little about the atmospheric fate (change in composition, lifetime) of volcanic particles in the micrometer size range.

Most research into atmospheric influence of volcanic eruptions is based on remote sensing from the surface or from satellite to follow the dispersal of volcanic SO_2 clouds or to investigate the influence of eruptions on the stratospheric aerosol load. The actual composition of volcanic aerosol has been investigated by aircraft-based measurements by Martinsson et al. (2009) and Schmale et al. (2010) following the Kasatochi eruption in 2008 and recently by Schumann et al. (2011) in the volcanic cloud from Eyjafjallajökull. However these studies only consider aerosol composition from single eruptive events and do not address evolution of the composition of volcanic aerosol in the atmosphere. In the study by Martinsson et al. (2009) a subset of the data presented in this study was used to investigate the development of the sulfurous and carbonaceous components of the volcanically influenced aerosol.

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Here we present and discuss the multi elemental composition of aerosol from three eruptions with VEI 4. The volcanic aerosol was collected between one and over 100 days after the eruptions, which provides a unique opportunity to investigate the evolution of the aerosol. Aerosol sampling and measurements of trace gases were performed by the CARIBIC (Civil Aircraft for Regular Investigation of the atmosphere Based on an Instrument Container) platform operating on a passenger aircraft (Brenninkmeijer et al., 2007), <http://www.caribic-atmospheric.com>.

2 Experimental methods

Samples of volcanic aerosol particles were obtained from the CARIBIC platform during regular long-distance passenger flights in the UT/LMS following large eruptions of the Kasatochi (2008) and Sarychev (2009) volcanoes, as well as during special flights at 4–12 km altitudes on 16 May and 19 May 2010, which were conducted to investigate the composition of the volcanic clouds produced by the Eyjafjallajökull eruption (Rauthe-Schöch et al., 2012). The CARIBIC measurement container is mounted in the forward cargo bay of a Lufthansa Airbus 340-600 during four sequential flights (Brenninkmeijer et al., 2007) on a monthly basis. From Frankfurt (Germany) destinations in North and South America, South Africa and South and East Asia, are reached thus covering a large geographical area mainly in the Northern Hemisphere. Instruments in the container automatically measure concentrations of many trace gases, aerosol number concentrations and the aerosol size distribution. In addition sampling of air and aerosol for laboratory analysis takes place. This study concentrates on volcanically influenced aerosol samples collected between 2008 and 2010, but also uses measurements of the background aerosol collected from 1999 to 2002 (Martinsson et al., 2005) and somewhat volcanically influenced samples from 2005 to 2008 (Friberg et al., 2012).

Aerosol particles of 0.08–2 μm were collected in a multi-channel aerosol sampler on 0.2 μm thick polyimide foils (Nguyen et al., 2006) by impaction. The upper size limit is determined by a cyclone separator placed between the aerosol inlet and the

sampler, and the lower limit by the cutoff diameter of the aerosol sampler. The sampler has 16 sampling channels with 14 for sequential collection and 2 for integral samples used to monitor contamination by comparing the integral samples to the sum of the sequential samples. The sampling time for each sequential sample is typically 100 min, corresponding to a flight distance of approximately 1500 km at cruising speed, and a sampling volume of approximately 0.25 m³ STP (Standard Temperature (273K) and Pressure (1013 hPa)). Sampling is suspended when the outside pressure is above 350 hPa. However, during the special flights through the Eyjafjallajökull volcanic cloud, collection of aerosol at lower altitudes was allowed and the sampling time was reduced to 50 min.

Aerosol samples were analyzed by accelerator-based techniques at the Lund ion beam analysis facility using two methods to obtain elemental concentrations: PIXE (Particle-Induced X-ray Emission) and PESA (Particle Elastic Scattering Analysis). In both methods the samples were mounted in a high vacuum chamber and irradiated with a beam of 2.55 MeV protons. Concentrations of elements with atomic numbers of 16 (*S*) or more were obtained by PIXE (Johansson and Campbell, 1988), and concentrations of hydrogen, carbon, nitrogen and oxygen by PESA (Nguyen and Martinsson, 2007). PESA was implemented for analysis of samples collected after 2005. Detection limits reached from several down to 0.1 ng m⁻³ STP, depending on element. The accuracy for elemental determination by both methods is estimated to be 10 % (Nguyen and Martinsson, 2007). The PIXE technique can also detect silicon (Si) when the concentrations are high, such as in aerosol samples which are dominated by aerosol particles from volcanic eruptions. In samples with low concentrations of Si problems arise due to interference with sulfur, which is often dominant. In samples with a *S* / Si ratio below a critical value, these effects have been corrected for, but for larger *S* / Si ratios corrections were not possible and the measured Si concentrations were excluded from the result. Elements that usually are below the detection limit (Sr, Zr) were detectable in samples collected in the Eyjafjallajökull volcanic clouds, using longer than usual analysis times.

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We further use ozone measurements performed by two instruments on the CARIBIC platform. A UV-photometer is used for accurate determination of the ozone concentration by absorption of UV light and also serves as a standard for a fast chemiluminescence detector which enables ozone detection with high temporal resolution. The accuracy is estimated to be 0.3–1 % at typical mixing ratios at a measurement frequency of 10 Hz (Zahn et al., 2012).

Also measurements of SO₂ concentrations and particle size distributions during the flights in the volcanic cloud from Eyjafjallajökull were used in this study. Concentrations of SO₂ are obtained from the CARIBIC DOAS (Differential Optical Absorption Spectroscopy) instrument (Dix et al., 2009), detecting NO₂, HCHO, HONO, BrO, OClO, O₃, SO₂ and O₄ simultaneously by measuring scattered or reflected sunlight, collected with three telescopes pointing to –82°, –10° and +10° relative to the horizon, with a temporal resolution of 8 s. During the volcanic flights the instrument was only functioning properly on 16 May, measuring in the –82° and –10° directions (Heue et al., 2011). Particle size distributions are measured with an integrated OPC (Optical Particle Counter), which measures particles with a diameter in the range of approximately 0.1–1 μm. OPC measurements from the volcanic flight on 19 May were used in this study (Rauthe-Schöch et al., 2012). A time resolution of 3 min was used to calculate the OPC size distributions during this flight.

Potential vorticity (PV) at the location of the aircraft was used to determine whether collected particles were of tropospheric or stratospheric origin, the tropopause region was defined as the region between 1.5–3 PVU (Potential Vorticity Unit; 1 PVU = 10⁻⁶ Km² kg⁻¹ s⁻¹). PV was derived from archived ECMWF (European Centre for Medium-range Weather Forecast) analyses with a resolution of 1 × 1° in the horizontal direction and 91 vertical hybrid sigma-pressure model levels. To obtain PV at the aircraft position the PV values was interpolated linearly in latitude, longitude, log pressure and time for each sample.

The recent history of probed air was investigated by means of 5-days backward air mass trajectories, calculated every third minute along the flight route using the

trajectory model TRAJKS (Scheele et al., 1996) and the horizontal and vertical wind fields provided by ECMWF.

Complementary to trajectory analysis, lidar measurements from the CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations) satellite were used. The lidar is equipped with a Nd:YAG laser producing highly polarized co-aligned beams with wavelengths of 532 and 1064 nm. Clouds and different types of aerosol particles can be identified since the shape and size of particles and droplets cause different scattering properties at these wavelengths (Winker et al., 2009).

3 Results

3.1 Identification of volcanic aerosol

A first indication of volcanic influence on aerosol samples is high concentrations of sulfur. These concentrations correspond to sulfate aerosol produced from SO_2 emitted during eruptions. Such influence is mainly seen in stratospheric samples, where the residence time of aerosol particles is long compared to the troposphere. In Fig. 1 the ratio of S/O_3 is used as an indicator of volcanic influence on stratospheric aerosol. Concentrations of S and O_3 show a correlation in absence of volcanic influence, since both sulfate and ozone are produced in the stratosphere with the aid of shortwave radiation. Consequently, high concentrations of sulfur are usually observed in association with downward transport in the stratosphere, together with elevated O_3 levels (Martinsson et al., 2009). Explosive volcanic eruptions disturb this ratio by injecting large amounts of additional sulfur. In Fig. 1, volcanic influence is indicated by elevated S/O_3 ratios following major eruptions (VEI 4) of the Kasatochi (52.177° N, 175.508° W), Redoubt (60.485° N, 152.742° W) and Sarychev (48.092° N, 153.20° E) volcanoes.

The sulfurous and carbonaceous components of the aerosol related to the Kasatochi eruption were discussed in detail in the study by Martinsson et al. (2009). Figure 1 shows that elevated S/O_3 ratios prevailed for at least four months after the Kasatochi

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eruption and were just reaching background levels when elevated concentrations of sulfur again were observed after the Redoubt and Sarychev eruptions in 2008. The small effects of the Eyjafjallajökull (63.63° N, 19.62° W) eruption in 2010 can be explained by the fact that the emissions were poor in SO₂ and that only a small fraction of the plume reached the stratosphere (BGVN, 2010; Thomas and Prata, 2011). However, very high concentrations of crustal elements were found in three aerosol samples collected during the special flights on 16 May and 19 May 2010, strongly indicating volcanic influence. These samples were collected in the troposphere and are thus not included in Fig. 1, which only shows samples collected in the stratosphere or the tropopause region.

Further connection to the eruptions was established by using 5-days, and in one case 8-days, backward trajectory analysis for samples following the Kasatochi (Martinsson et al., 2009), Sarychev and Eyjafjallajökull eruptions. The first samples following the eruption of Redoubt were collected more than a month after the first explosion; hence trajectory analysis could not be used to verify volcanic origin. In addition, these samples show less elevation in their S/O₃ ratio than the samples following Sarychev and Kasatochi. The eruptions of Sarychev and Kasatochi emitted approximately 1.2 Tg (Haywood et al., 2010) and 2 Tg (Yang et al., 2010) of SO₂, respectively, into the atmosphere which can be compared to only 0.08 Tg by the eruption of Redoubt (Lopez et al., 2009). It is thus unlikely that the sulfate aerosol produced by the Redoubt eruption had large enough influence for a clear identification in sampled aerosol. It is possible that the elevated concentrations in the samples instead are due to downward transport of an upper branch of the aerosol produced in the preceding eruption of Kasatochi. The volcanically influenced aerosol also contains a crustal component, which will be further discussed in Sect. 3.2. Stratospheric concentrations of crustal elements such as potassium (K) and iron (Fe) show a seasonal dependence with high concentrations in spring (March to June) that seem to be connected with transport across the tropopause from the troposphere (Martinsson et al., 2005). The samples collected following the Redoubt eruption were collected at this time of the year; therefore it cannot be excluded that the

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crustal component of the aerosol has been transported from the boundary layer rather than been injected by the eruption. Due to the uncertainty of the source of the aerosol following the Redoubt eruption, these samples are excluded from further consideration.

Trajectory analysis performed for each of the three samples with large crustal components from the Eyjafjallajökull volcanic clouds showed that the sampled air had passed over the volcano. The transport time from the volcano to the aircraft position, and thus the age of the aerosol, was estimated to have been about 40 and 45 h for the samples collected 16 May and about 25 h for the sample collected 19 May. The time for sampling within the volcanic cloud, and thereby the most likely transport path, was determined from peaks in the SO₂ and particle mass concentrations obtained from the CARIBIC DOAS and OPC instruments (Heue et al., 2011; Rauthe-Schöch et al., 2012).

Trajectory analysis conducted for samples following the Sarychev eruption was combined with lidar measurements from CALIPSO (Fig. 2). The large amount of sulfate produced during this eruption made it possible to follow the volcanic aerosol in the lidar measurements up to more than a month after the eruption. Aerosol layers detected by CALIPSO at an altitude between 10 and 13 km were identified, and this information was put together with the path of calculated air mass trajectories. By this method aerosol produced in the eruption earlier than the 5 days backward time span of the trajectories could be linked to the eruption, by investigating if the trajectories intersected these aerosol layers. Three samples collected during the first flight after the Sarychev eruption, taking place 12 days after the start of the eruption, had trajectories that clearly traversed aerosol layers. Also samples collected during the next flight, 6 weeks after the eruption, could be connected to the aerosol produced in the eruption by this method, because the volcanic cloud could still be identified in the CALIPSO measurements.

3.2 Ash composition

The high concentrations of crustal elements in three of the samples collected during the Eyjafjallajökull eruption are important indications of volcanic origin. To further investigate this component of the aerosol it was compared to the composition of a

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fall-out sample of volcanic ash from the eruption site (Sigmundsson et al., 2010). The composition of the three CARIBIC samples shows good agreement with the fallout sample for crustal elements (Fig. 3). The higher aerosol potassium (K) content in the CARIBIC samples could have been caused by fractioning in concentration between different sizes of ash particles in the samples analyzed, due to a larger surface area to mass ratio of small particles. Small ash particles remain in the plume longer than larger particles, and cool faster, which favors condensation of volatiles onto the smaller particles (Witham et al., 2005). Among the elements presented in Fig. 3, K happens to be the most volatile species found enriched in volcanic gases, due to degassing from the magma (Hinkley et al., 1994; Rubin, 1997). Also the zirconium (Zr) content is higher in the aerosol than in the fallout sample. Zirconium was found in enhanced concentrations in deposits from a volcanic plume compared to the magma (Moune et al., 2005), indicating that also this element could have been abundant in the gas phase and condensed onto the ash particles.

The oxygen content (corrected for the amount of oxygen assumed bound to sulfurous aerosol in the form of sulfate) is also high compared to that of the fallout sample. The amount of oxygen in the fallout sample was not measured (indicated by grey symbols in Fig. 3), but are expressed as generalized oxygen proportions to the corresponding elements. It should however represent the approximate oxygen content, indicating that there is more oxygen in the aerosol samples. A likely explanation for the extra oxygen is that part of it is bound to the carbonaceous fraction of the aerosol (see Sect. 3.3).

The ash component of the Eyjafjallajökull aerosol is also similar to Earth's crustal composition (Rudnic and Fountain, 1995). Although analysis of rare elements such as the chalcophile metals (Bi, Cd, Cu, In, Pb and Tl), found in volcanic plumes from degassing of silicate melts (Hinkley et al., 1994), would be necessary to distinguish volcanic ash from this type of source, the high concentrations of crustal elements together with elevated sulfur and carbon concentrations in the aerosol samples are strong evidence of volcanic origin. On a final note, all of the elements, except manganese (Mn), in these three samples have the highest concentrations noted in the entire CARIBIC

data set taken over a 10 yr period. Therefore the composition of crustal elements in the Eyjafjallajökull aerosol samples (Fig. 3) is used to define the ash component of volcanic aerosol.

The ash component of the sampled aerosol influenced by the Kasatochi and Sarychev eruptions was identified by comparing it to the ash composition of the Eyjafjallajökull samples. Since this composition cannot be clearly distinguished from Earth's crustal composition, it was used as an indicator of volcanic ash in those samples with elevated S/O_3 levels, and for early collected samples, where trajectory analysis indicate volcanic origin. In addition the identified samples from the Kasatochi and Sarychev eruptions were collected in the summer to winter period when little influence of crustal particles in the LMS is seen (Martinsson et al., 2005). Only the three first samples following the Sarychev eruption were collected in spring when concentrations usually are elevated. For these samples however there is strong evidence of volcanically influence from the trajectory analysis and aerosol composition. Also meteoritic material can contribute to elements indicative of crustal material. Samples which are mainly influenced by meteorites can be identified by their Fe/Ni ratio (Kopp, 1997), which was used to exclude them from this study.

In Fig. 4 we include all samples having a crustal component classified to be of volcanic origin. These samples have a geometric residual sum of squares (RSS) in their elemental to iron ratios of less than two compared to the geometric mean of the Eyjafjallajökull aerosol samples (shown in Fig. 3). A limit of at least three detected elements was applied to be able to examine the composition. Also the detection limit of undetected elements in relation to the iron content was taken into account. Samples were excluded if more than one of the elements K, Ca, Ti or Mn were missing and were expected to have had a concentration above the detection limit in relation to the content of Fe in the samples. One missing element was accepted to account for the risk of contamination due to mixing in the atmosphere and since it is likely that the composition of ash from different eruptions is not exactly the same. The ash composition depends on the magma type as well as on the absorption of volatiles onto the ash particles which

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is controlled by a number of factors such as eruption type, concentration of gases and particles and particle size (Witham et al., 2005). As can be seen from Fig. 4, Si is only represented in a few samples which is due to problems with interference with S as described in Sect. 2. Also Mn is represented only in a few samples since its concentrations in ash is low hence falling below the detection limit in samples of low ash content.

The composition of samples collected 8 days after the Kasatochi eruption and 12 days after the Sarychev eruption (indicated by dark circles in Fig. 4) show good agreement with the composition of the Eyjafjallajökull samples, except for lower concentrations of K in the sample from the Kasatochi eruption. Also samples collected one month or more after the eruptions are similar in their composition but ratios show more scatter. The larger deviations in composition in these samples indicate that the aerosols are more mixed and influenced by other sources. The identified samples, classified as volcanic aerosol both by elevated S/O_3 ratios and composition of the crustal (ash) component, range in age between 8 to 128 days and 12 to 77 days from the start of the explosive phase of the Kasatochi and Sarychev eruptions, respectively. The length of these eruptions adds an uncertainty to these age estimates (1 and 5 days of explosive eruptions in the case of Kasatochi and Sarychev, respectively, BGVN, 2009; Waythomas et al., 2010).

3.3 Major components

The main components of the samples classified as volcanic aerosol are carbon, sulfate and ash (Fig. 5). Samples influenced by Kasatochi and Sarychev are grouped with respect to age, where the compositions of aerosol collected at approximately the same time from the eruptions are presented as mean values. The carbonaceous component is presented as the mass of the measured carbon concentrations, since the stoichiometry of the carbon aerosol is unknown. The mass of sulfate (SO_4) was calculated by adding the amount of oxygen corresponding to the measured mass of sulfur, assuming that all sulfur is in the form of SO_4 . In a similar manner the mass of the

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ash components in the Eyjafjallajökull samples were obtained by adding the mass of oxygen according to the fallout sample presented in the table by Sigmundsson et al. (2010). Elements found in ash that could not be detected, primarily sodium (Na), magnesium (Mg), aluminum (Al) and phosphorus (P), were calculated according to the composition of the fallout sample using the relation to the iron content. The entire ash components of samples influenced by the Sarychev and Kasatochi eruptions were estimated based on their iron content since many of the elements were below their detection limits. In these samples the mass of Fe, S and C was corrected for stratospheric background concentrations calculated as averages of samples with concentrations below the 95 percentile collected during the period from 1999 to the eruption of Kasatochi in August 2008. For S and C this correction was done with respect to the O₃ mixing ratios measured during the sampling time since they show a correlation with O₃. For Fe, the background correction was done with respect to season, divided into December–February, June–August and September–November (no samples were obtained in the period March–May). Since the samples from the Eyjafjallajökull eruption originate in lower altitudes than the CARIBIC platform usually measures, no such correction was made for them. Therefore C and S components should be considered to represent maximum estimates. However the concentrations were very high so the background influence most likely was small.

Figure 5 reveals that the samples collected following Eyjafjallajökull's eruption show little variation in their compositions, they mainly consist of ash and sulfate in about equal amounts. In contrast samples influenced by the eruptions of Kasatochi and Sarychev to a large extent consist of sulfate and carbon, while the ash component is well below 10%. The carbonaceous fractions in the volcanic samples are highly elevated compared to background conditions, indicating that they are connected to volcanism (Martinsson et al., 2009).

The aerosol from Sarychev shows a strong dependence of the carbon-to-sulfur ratio on latitude, with increasing C/S ratio with increasing latitude, while the iron-to-sulfur ratio shows no such dependence (not shown). The origin of the carbonaceous

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component in volcanic aerosol is not known (Martinsson et al., 2009). Possibly changing properties of the eruptions that persisted for several days and/or differences in carbonaceous aerosol precursor gases combined with transport patterns from the source can explain this latitudinal dependence. The time evolution of C/S can thus not be given. Instead the average C/S over the volcanically influenced samples taken from June to October 2009 is used for Sarychev in Fig. 5 together with the relation between Fe and S for the respective time period after the eruption. A decreasing fraction of ash with time is observed from Sarychev. The aerosol from the Kasatochi eruption has a simpler C-to-S relation. The sulfurous fraction shows an increase with time, whereas the ash and carbonaceous fractions decline.

4 Discussion

To further evaluate the evolution of the volcanic aerosol the sulfate and ash components were studied. In Fig. 6 the ratio of Fe/S shows the relation between the ash and sulfate components as a function of time after the eruptions. A decreasing trend can be seen in the ratios, which likely is explained by the deposition velocity of the ash and sulfate particles as well as by the rate of conversion of SO₂ into sulfate aerosol. Since all samples older than two weeks were taken in the stratosphere or the tropopause region, where little wet scavenging occurs, the solubility of the particles has had no, or only a small influence on the rate of deposition. If the difference in sedimentation velocity between ash and sulfate particles in the 0.08–2 μm size range is assumed to be small, the conversion rate of SO₂ into SO₄ can be estimated from the decrease in the Fe/S ratios. From the reaction of SO₂ with OH, which mainly controls the conversion into sulfate aerosol (Weisenstein et al., 1997), an exponential decay rate of the number concentrations of SO₂ can be derived (Seinfeld and Pandis, 2006)

$$C_{\text{SO}_2}^n(t) = C_{\text{SO}_2}^n(0) \cdot e^{-kt}, \quad (1)$$

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where k is the loss rate constant. Since the depletion rate $C_{\text{SO}_2}^n(t)/dt$ of SO_2 molecules equals the production rate of SO_4 , the mass concentration ratio of Fe and S can be expressed as;

$$\frac{C_{\text{Fe}}^n}{C_{\text{SO}_4}^n(t)} \propto \frac{C_{\text{Fe}}^m}{C_{\text{S}}^m(t)} = \frac{A}{1 - e^{-kt}} \quad (2)$$

where C^n and C^m denotes number respective mass concentrations. The constant A is the mass concentration ratio of Fe from ash and total S from SO_2 that is eventually converted to sulfate. Since the composition of volcanic aerosol depends on the eruption characteristics (magma composition, explosivity, temperature etc. (Mather et al., 2003)) aerosol from different eruptive events cannot be expected to have similar Fe/S ratios. Still most samples collected following the Sarychev and Kasatochi eruptions show large similarities in their Fe/S ratio with respect to age. Thus Eq. (2) was fitted to the Fe/S ratio in Sarychev and Kasatochi samples, assuming similar emission ratio of Fe/S in the eruptions. The logarithm of the Fe/S ratios was used in the fitting to avoid over representing the large ratios appearing the first two weeks after the eruptions. Changed emissions during the eruptions as well as atmospheric conditions that affect mixing and transport, also influence the composition of the aerosol. As stated earlier, the ash component was rather simple to recognize within two weeks after the eruption, and thereafter it was more influenced by other sources. Such influences could explain some of the deviations from the decreasing trend in Fig. 6. Also deviations from the average background concentrations used in the corrections have to be considered. Samples taken more than 100 days after the eruptions show large deviations from the general tendency, probably as the result of dilution of the volcanic cloud and mixing, and are excluded from the fit. One younger Sarychev sample shows large deviation compared to other samples collected at approximately the same time, and has therefore also been excluded.

Samples from the Eyjafjallajökull volcanic cloud was not included in this fit. As was seen in Fig. 5, the relative concentration of sulfate is low in samples from the

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Eyjafjallajökull eruption compared to the samples following Sarychev and Kasatochi, likely due to the large emissions of fine grained ash in the Eyjafjallajökull eruption (BGVN, 2010). In addition these samples were collected close to the eruption which implicates short time for conversion of SO₂ into sulfate aerosol. However the residence time of SO₂ in the troposphere where these samples were collected are considerably shorter than in the stratosphere, only hours to few days (McGonigle et al., 2004; Carn et al., 2011), leading to rapid conversion into sulfate aerosol. These samples should therefore not follow the same decay as those collected at higher altitudes. In addition no background correction was made for the Eyjafjallajökull samples as mentioned earlier.

The residence time of SO₂ ($1/k$) in the tropopause region and LMS after the eruptions was estimated from the fitted curve to be (mean \pm standard error) 45 ± 22 days. The value of A , representing the ratio of total Fe and S was estimated to 0.0036 corresponding to a final ash/SO₄ mass ratio of approximately 0.016 for particles in the 0.08–2 μm size range considered here. The obtained residence time can be compared to earlier estimated stratospheric residence times of 62 days (Jurkat et al., 2010) following the Kasatochi eruption as well as 35 (Bluth and Doiron, 1992) and 33 days (Niemeier et al., 2009) following Pinatubo (1991). Thus these residence times are within the span of the present estimate. Longer residence times following the eruptions of Sarychev and Kasatochi could be caused by differences in the abundance of OH. The latitudinal variability could thereby be of significance as Pinatubo is located in the tropics (15.13° N, 120.35° E) where OH concentrations are higher than at the mid-latitudes (Gross and Khalil, 2000) where the Kasatochi and Sarychev volcanoes are located. Since the eruptions occurred in June (Pinatubo and Sarychev) and August (Kasatochi), there should be negligible differences in the OH concentration due to seasonal variation in the UV flux. A more important factor are likely to be the altitude at which these estimates were made, since OH concentrations increase fast above 20 km (Gross and Khalil, 2000). The results obtained in this study and by Jurkat et al. (2010) are based on aircraft measurements at altitudes of 7–12 km. The estimated residence time by Bluth and Doiron (1992) was based on TOMS satellite measurement of the

Pinatubo volcanic coloud, which was injected well into the stratosphere to altitudes of 25 km (Guo et al., 2004). The model result by Niemer et al. (2009) were calculated for an injection heighth of 24 km.

It is possible that the residence time obtained in this study is underestimated due to a higher sedimentation velocity of ash than sulfate particles. The impact of the different sedimentation velocities was investigated by estimating the settling velocity of ash and sulfate particles. In the computations the particle diameters that carry most of the mass according to the particle mass distributions were applied. For sulfate spherical particles with a diameter of 0.6 μm (Martinsson et al., 2005) (corresponding to 0.76 μm aerodynamic diameter) and a density of 1600 kg m^{-3} (Yue et al., 1994) were used in the calculations. The diameter of the sulfate aerosol estimated by Martinsson et al. (2005) was obtained from the effective radius of stratospheric aerosol from lidar observations (Bauman et al., 2003) during a period with low volcanic activity. Thus the radius is underestimated for volcanic aerosol which usually exhibit larger diameters (Bauman et al., 2003; Deshler et al., 2003). Ash particles usually have substantially larger diameters (Schumann et al., 2011). The 2 μm aerodynamic diameter upper size limit of the aerosol sampler was thus used to estimate the upper limit in settling velocity of the ash. The thickness of the volcanic aerosol layers from the Sarychev and Kasatochi eruptions, through which the volcanic aerosol has to settle, was estimated by locating the altitude of volcanic aerosol during the first week after the eruptions using CALIPSO lidar measurements. After some time the identified volcanic aerosol layers are assumed to have been horizontally mixed and contributed to a more or less homogeneous aerosol layer. Therefore a cumulative altitude distribution of the volcanically influenced air from 10 km altitude and upwards was calculated, by summing all events when volcanic aerosol was observed at a certain altitude, see Fig. 7. About 85 % of the identified aerosol following the Sarychev eruption was injected to a height between 10 and 15 km. Therefore a 5 km thick layer was assumed to have been produced. The aerosol layer formed by the Kasatochi eruption was less homogenous with 75 % of the aerosol located between 10-13 km and the remaining aerosol extending up to

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20 km. During the 2.5 months considered here, the 2 μm ash particles fall a distance of less than 15 % of thickness of the volcanic aerosol layer produced by Sarychev and about 20 % of the lowest 3 km of the volcanic aerosol produced by Kasatochi. Since the aerosol samples were collected in the lower part of these layers, at 10–11.3 km, there should thus still be large amounts of 2 μm ash particles, originating in the upper part of the volcanic aerosol layers. Baring in mind that the sulfate sedimentation was underestimated and that of ash was overestimated, this simple estimation suggests that the difference in deposition velocity between ash and sulfate particles only has minor importance for the relation between the measured ash and sulfate concentrations on the timescale considered here. Thus, sedimentation only has a weak influence on the estimation of the residence time of SO_2 .

5 Conclusions

Volcanic aerosol have been collected in the tropopause region and lowermost stratosphere following major eruptions of the Kasatochi and Sarychev volcanoes, and in the troposphere in the volcanic cloud from the Eyjafjallajökull eruption in 2010. The main components of the volcanic aerosol were found to be sulfate, ash and a carbonaceous component. In samples collected in the volcanic cloud from Eyjafjallajökull ash and sulfate contributed approximately equal amounts to the total aerosol mass ($\sim 45\%$). In samples collected following Sarychev and Kasatochi ash was a minor part of the aerosol (1–7 %) while sulfate (50–77 %) and carbon (21–43 %) were dominating. These fractions changed with the age of the aerosol. In this study we could follow the evolution of volcanic aerosol during more than twice the residence time of SO_2 in a volcanic cloud. The first samples collected 1 and 1.5 weeks after the eruption of Kasatochi and Sarychev consisted of 14 % and 5 % ash relative to the mass of sulfate, respectively. After 10 and 11 weeks the ash component was still identifiable, but had decreased to about 2 % relative to the sulfate mass for both eruptions. The ash / sulfate ratio of aerosol collected following the Kasatochi and Sarychev eruptions showed a

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decreasing trend. From this decay the residence time of SO₂ in the tropopause region and lowermost stratosphere was estimated to be 45 ± 22 days.

Acknowledgements. We thank all members of the CARIBIC team, especially C. Koepfel, D. S. Scharffe and S. Weber. The collaboration with Lufthansa and Lufthansa Technik is gratefully acknowledged. Aerosol measurements from CALIPSO were produced by NASA Langley Research Center. Financial support from the Swedish Research Council and the Swedish Research Council for Environments, Agricultural Sciences and Spatial Planning under grants 621-2007-4639 and 214-2009-613 is gratefully acknowledged.

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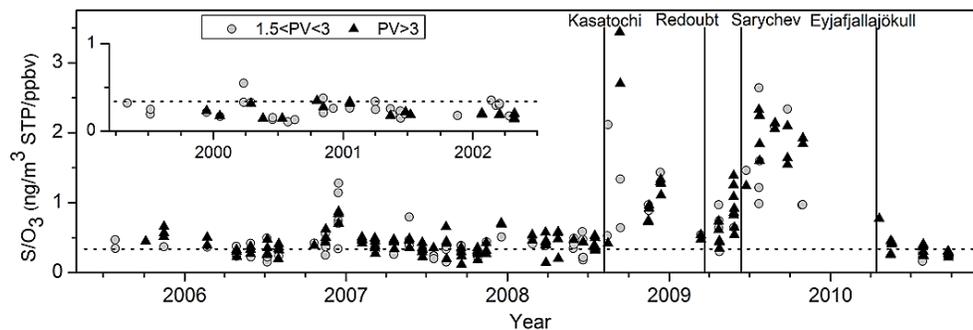


Fig. 1. Ratio of particulate sulfur to ozone concentration in aerosol samples collected in the tropopause region (grey circles) and the lowermost stratosphere (black triangles). The dotted line indicates the geometrical mean of samples collected before the eruption of Kasatochi in August 2008. The occasions of major eruptions are marked by vertical lines.

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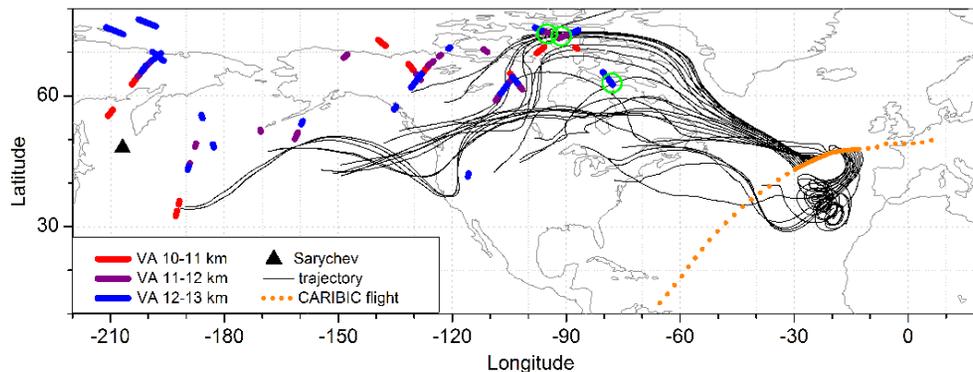


Fig. 2. Air-mass back-trajectories (thin black lines) calculated for one sample collected during the first CARIBIC flight from Frankfurt to Caracas (orange dotted line) following the eruption of Sarychev, combined with volcanic aerosol (VA) layers obtained from CALIPSO lidar images at 10–13 km altitude (colored lines). Green circles indicate where trajectories and aerosol layers intersect horizontally and vertically.

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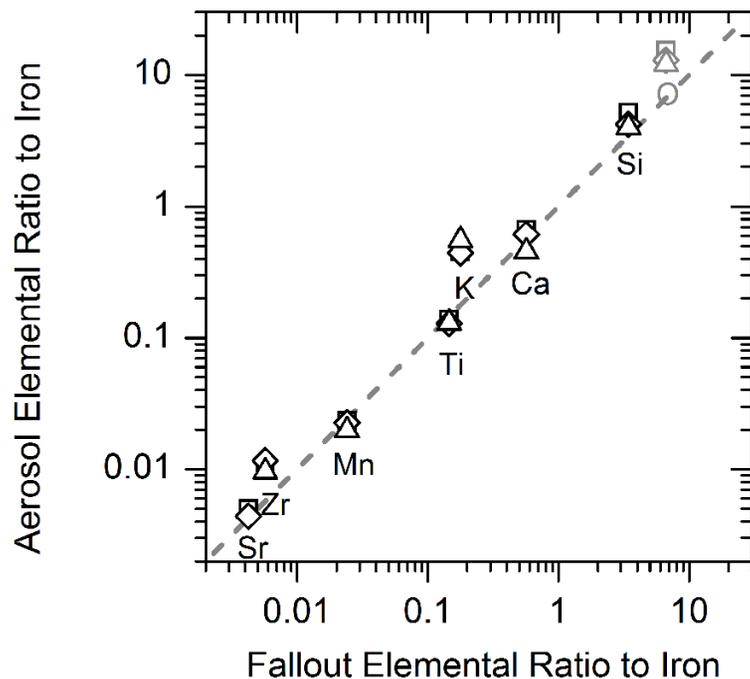


Fig. 3. Elemental ratio to iron in three aerosol samples (shown by different symbols) collected by the CARIBIC platform vs. fall out sample (SRG 5a, Sigmundsson et al., 2010). The dashed line indicates equal ratio. The oxygen content in the fallout sample is not measured and therefore indicated by grey symbols (see text for further details).

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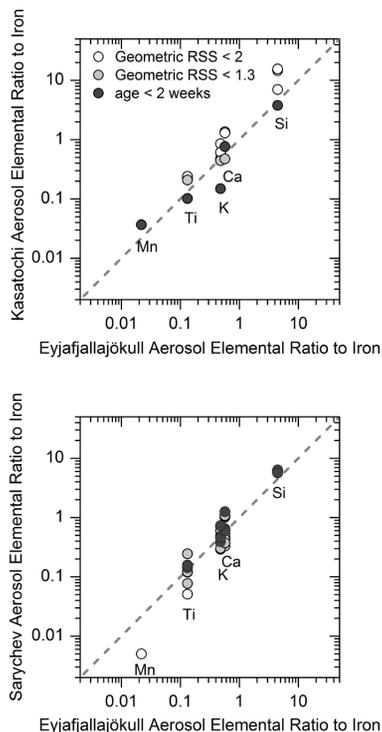


Fig. 4. Elemental ratio to iron in aerosol samples collected following eruptions of Sarychev and Kasatochi vs. geometric mean of volcanic aerosol collected in the volcanic cloud from Eyjafjallajökull. The dashed line indicates equal ratio. Samples with a Residual Sum of Squares (RSS) of less than two are included in the figures. Samples collected less than two weeks after the start of the eruptions are marked by dark circles.

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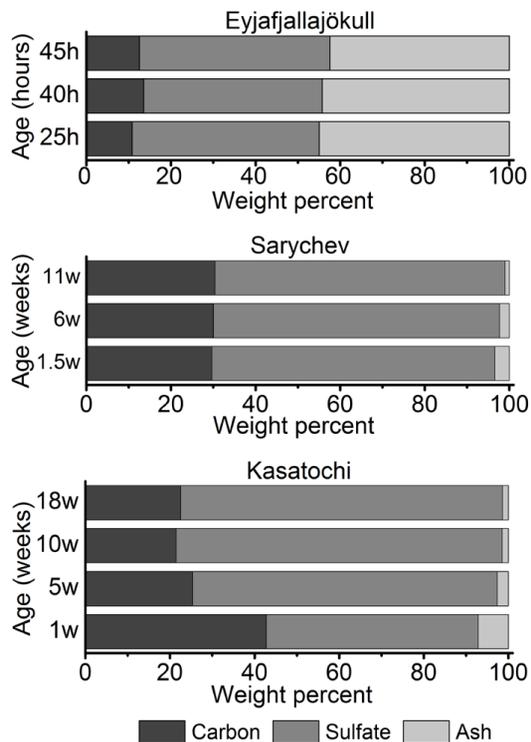


Fig. 5. Major components of aerosol samples collected following eruptions in Eyjafjallajökull, Sarychev and Kasatochi. Aerosol collected 1.5, 6 and 11 weeks after the eruption in Sarychev are shown as averages of 3, 6 and 2 samples respectively. The composition of aerosol collected 18 weeks after the Kasatochi eruption are an average of 3 samples. The remaining aerosol compositions are each represented by only one sample. The carbonaceous fraction is averaged over all samples of the aerosol from Sarychev, see text for further details.

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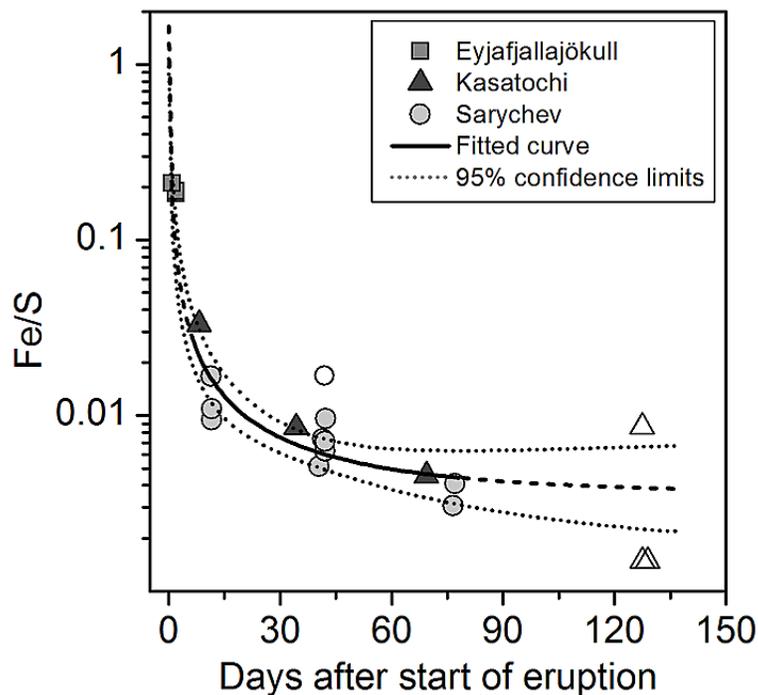


Fig. 6. Mass ratio of iron to sulfur in aerosol samples influenced by three volcanoes (marked by different symbols) vs. time since the start of the eruptions. The line shows a fit to the Fe/S ratio for the samples influenced by the Sarychev and Kasatochi eruptions. Samples shown with open symbols have been excluded from the fit.

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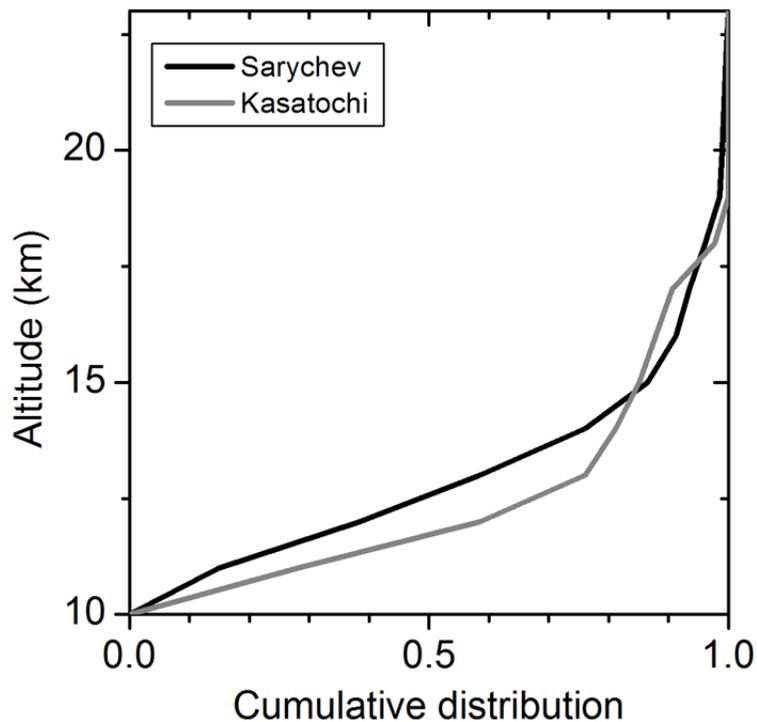


Fig. 7. Cumulative altitude distributions of volcanic injections above 10 km the first week after the Sarychev and Kasatochi eruptions. The distributions was deduced from the number of observations of volcanic aerosol layers in CALIPSO lidar measurements as a function of altitude, see the text for further details.

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