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

**Commented by A. J. Durant**

We thank the referee for the valuable comments which have helped to improve the manuscript. Below we address the given specific comments. Replies are given in red.

## Specific comments

The authors state that there is a correlation between C present in solid aerosol particles sampled by the CARIBIC platform and a volcanic origin for the aerosol. Can the authors discuss the origin of C in the volcanic system? To my knowledge, carbon will only be present in significant quantities in erupted products if the magma interacts extensively with carbonates in the crust prior to eruption. This is not a common process – a well-known example is Ol Doinyo Lengai, Tanzania, a cabonatite volcano. One natural process the authors do not discuss is wildfires. The Martinsson 2009 GRL paper cited presents an analysis of carbonaceous material in the Kasatochi August 2008 aerosol, but again stops short of explaining where the C is derived from. According to the National Oceanic and Atmospheric Administration National Climatic Data Center, approximately 7,000 acres (2,833 ha) of the U.S. were burned by wildfires during August 2008 (see http://www.ncdc.noaa.gov/sotc/fire/2008/8). It is known that wildfires can generate plumes that penetrate the tropopause and carry particulate matter to the stratosphere (Fromm et al. 2000; Fromm et al. 2006; Fromm and Servranckx 2003). It is possible that the C measured by CARIBIC is related to wildfires or other combustion sources, and not at all to the volcanic eruption. It is also possible that airborne soot particles generated from wildfires in North America are either entrained by the volcanic column during eruption, or that wildfire soot particulates and volcanic emissions mix during the dispersion of the volcanic cloud; the soot may have pre- or post-dated the volcanic aerosol. Furthermore, the Icelandic aerosol contains less C than the Alaskan/Kamchatkan volcanoes. Perhaps this can be explained due to the prevalence of wildfires over North America in close proximity to these other volcanoes, versus Iceland, due to the greater distance from source. In summary, the value of using C as a tool for volcanic source attribution in the IAGOS flights is not immediately obvious or relevant.

We agree with the referee that the C component in the volcanically influenced aerosol need to be covered more in detail, and have therefore inserted a section on this in the revised manuscript, were we point to the air that is entrained into the volcanic jet and plume as a possible source. This section follows below (last in this section). We don't consider direct injection of C by wildfires as a likely source of the measured C as pyro convection is not a very common phenomenon, and only a few events per year result in transport of the plume above 8 km altitude, see table 1 in Guan et al. (2010). Also none of the identified events where aerosol is transported into the stratosphere in Guan et al. (2010) coincides with our measurements of volcanic influence. It seems unlikely that such events would produce high concentrations of C in the UT/LMS simultaneously as we see other volcanic signatures (i.e. high $S/O_3$ ratios and air mass trajectories intersecting the eruption area/volcanic clouds). On the other hand, aerosol from wildfires at lower altitude could be entrained during the eruption and that way affect the carbon concentration. However, we do not see clearly elevated concentrations of potassium, in excess of the ash concentration, which would be expected from wildfires.

The C component in the Eyjafjallajökull samples is actually of similar concentration as in the samples collected after the other two eruptions; only the large amount of ash makes the C component look small. This has been clarified in the revised manuscript.

Finally we would like to point out that the C content has not been used for volcanic source attribution; it is only presented as a part of the *volcanically influenced* aerosol. For the identification of volcanic aerosol we have used $S/O_3$ ratios, trajectory analysis for samples close in time to eruption, and in a second stage composition of the mineral component.

*For implementation in revised manuscript:*

The studied volcanic clouds contain substantial amounts of particulate carbon, this despite the fact that only for few volcanos lava can interact with carbonates in the crust prior to eruption. The volcanoes considered here do not belong to this category. This is corroborated by the measurements because the stoichiometric relations between carbon, oxygen, sulphur and ash elements in the sampled aerosol do not permit such an oxygen-rich form of carbon. Therefore explanations other than direct volcanic emissions need to be considered. One alternative explanation is the carbon content of the air that is entrained into the volcanic jet and lifted with the volcanic effluents. Mixing with air creates the buoyancy needed for the formation of a volcanic plume that can reach the stratosphere (Suzuki and Koyaguchi, 2010).

To obtain an estimate on the organic aerosol concentration in the region of the volcanoes studied, we consider the conditions in the boundary layer, where large numbers of observations are available. Boundary layer air contains high concentrations of carbonaceous aerosol from anthropogenic and from natural sources. Jimenez et al. (2009) report average urban organic aerosol concentrations up to 30 000 ng/m$^3$ (Beijing, China) and, even at remote sites like Hyytiala, Finland the organic concentration exceeds one thousand ng/m$^3$. In a study focusing on the USA, urban regions average concentrations of several thousand ng/m$^3$ were found and in rural areas the organic concentration was of the order 1000 ng/m$^3$ (Hand et al., 2012). Interestingly, the Alaskan rural average organic aerosol concentration peaks in August, the month of the Kasatochi eruption, at approximately 3 000 ng/m$^3$. Organic aerosol sources extend also to the oceans. Regional and seasonal variation in oceanic biological activity can be derived from SeaWiFS sensor on the OrbView2 satellite. The activity shows strong geographical and seasonal variations (Stramska, 2009). Measurements in Maze Head (Ireland) show organic aerosol concentration of several hundred ng/m$^3$ connected with biologic activity in the ocean during spring to autumn (O'Dowd et al., 2004;Yoon et al., 2007). All three volcanoes studied here erupted in the biologically active part of the year. Modelling of the global distribution of yearly average organic aerosol with the ocean source included indicate high, to a large degree ocean-derived, organic aerosol concentration over the ocean surrounding island volcanoes Sarychev, Kasatochi and Eyjafjallajökull. For Sarychev a concentration of approximately 1000ng/m$^3$ was found, the same or somewhat lower for Kasatochi and a few hundred ng/m$^3$ for Eyjafjallajökull (Spracklen et al., 2008). Additional organic material can be derived from gaseous precursors. Volcanic halogen emissions organic chemistry is important in the often OH poor volcanic clouds (von Glasow et al., 2009). In the volcanic cloud of Eyjafjalljökull chlorine radicals rapidly depleted organic trace gases to levels well below background concentrations (Baker et al., 2011). Such processes can further add particulate carbon to the volcanic cloud.

The LMS particulate carbon concentration measured by CARIBIC outside directly injected volcanic clouds is approximately 100 ng/m$^3$ at STP. In volcanic clouds the concentration typically is a few hundred ng/m$^3$ STP, reaching more than one thousand ng/m$^3$ STP in three observations of the volcanic cloud from Sarychev. The regional average particulate carbon concentrations in the boundary layer around the volcanoes studied here are thus comparable to the observations at high altitude (free troposphere, UT and LMS) in this study. Although sources in direct connection to the volcano, such as burning vegetation and sedimentary layers, also could contribute to carbonaceous aerosol, we hypothesize that organic material in entrained air constitutes a significant fraction of the particulate carbon observed in volcanic clouds.

On P.21490 the authors state, “A first indication of volcanic influence on aerosol samples is high concentrations of sulfur”. It should be noted (and stated alongside this statement) that this observation is not always true. For example, Thomas and Prata (2011) carefully analyzed the Eyjafjallajökull volcanic clouds and determined that ash and sulphur species may co-exist or be mutually exclusive.

The statement of the referee is correct, however all samples except those taken during the special flights during the Eyjafjallajökull eruption have been collected more than one week after the eruptions. In these measurements we have not identified volcanic clouds other than those where the sulfate component is clearly dominating. Since the sentence well describes the method that we used, and since the air would be volcanically influenced even if there were no ash present due to separation, we would like to keep this sentence. However to clarify that this does not include the early collected samples after the Eyjafjallajökull eruption, "aerosol sample" has been replaced with "CARIBIC UT/LMS aerosol samples". Also separation of ash and $SO_2$ is now considered in the introduction.

In Figure 4 the authors compare measured Sarychev and Kasatochi aerosol composition to the Eyjafjallajökull aerosol. . . why is the comparison relative to Eyjafjallajökull? Also why aren't Sarychev and Kasatochi compared in the same way? And furthermore, why is a single sample of fallout used for ground-truthing? This single sample represents an instantaneous composition of the eruption (although the source varied over time). The single sample was derived from an initial particle distribution erupted at the vent that would have been subject to fractionation during transport and sedimentation in the atmosphere. For example, see Figure 7 of Carey and Sigurdsson (1982) for variation in the composition of fallout from the well-studied Mount St. Helens 1980 eruption. In summary, this sample is fairly arbitrary and should not be used alone to define the geochemistry of the Eyjafjallajökull eruption.

Chemical analysis of six independent samples from the Eyjafjallajökull eruption is available at http://earthice.hi.is/eyjafjallajokull_2010_chemical_composition. Even though these samples represent different types of eruption products they are similar in composition. Thus this composition seems to represent the erupted material well, at least at "close" distance to the volcano. Not all of the analysis results presented on the web are published, thus we choose one of the samples that is published in (Sigmundsson et al., 2010). The change in ash composition with distance from the volcano is an important aspect that now has been included in the revised manuscript. However our analysis for elemental composition give us no possibility to see changes in mineral constituents or the "type" of ash particles except a variation in concentration of the elements. Interestingly, the aerosol samples fit well with the Eyjafjallajökull fallout sample. To identify ash in aged volcanic clouds we need to define some criteria for the classification of ash. Our best way to do this is to use the ash samples that were collected after the Eyjafjallajökull eruption, as no analyses of fallout samples were available from Kasatochi and Sarychev. However, the comparison of the Kasatochi and Sarychev samples to the Eyjafjallajökull aerosol samples shows that the composition of the ash component is similar. There is thus no obvious reason to compare the Sarychev and Kasatochi samples in the same way.

On P.21498 L.16 the authors discuss ash particle lifetime and note that after 2 weeks in the atmosphere it was challenging to identify the volcanic ash fraction based on composition of the aerosol. At this point please refer to Rose et al. (2001) – this paper describes satellite-constrained observations of the lifetime of volcanic ash clouds. It is known that ash aggregation causes much of the fine ash fraction (<63 microns) to settle to the surface at rates orders of magnitude faster than predicted by gravitational settling of single particles (Brown et al. 2012), and the majority of the ash fraction is removed in the first day or so. Therefore it will be expected that the volcanic aerosol will quickly evolve away from an ash-dominated composition to a sulphate aerosol dominated composition as aggregation and sedimentation remove ash particles faster than sulphate aerosol generated due to the oxidation of $SO_2$. On P.21501 L.1, "During the 2.5 months considered here, the 2 μm ash particles fall a distance of less than 15

We thank the referee for bringing up this important aspect. We have now included a section on the initial evolution in a volcanic plume in the introduction, including Rose et al. (2001), to better describe the development of volcanic ash clouds from an early state. The processes described by Rose et al. (2001) are however mainly taking place before we encounter the volcanic cloud for the first time one week after the eruptions, and dilution is more likely the main cause for the less significant ash (and sulfate) signal in later collected samples than the processes described by Rose et al. (2001).

Since we do not follow the volcanic clouds during the first week after the eruptions in these samples, we have changed from “During the 2.5 months considered here” to “During the 2 months we followed these volcanic clouds” and inserted new estimates of sedimentation. This also means that initial rapid aggregation and fallout does not affect our measurements. Coagulation in a volcanic cloud acts as a self-limiting effect on the number concentration as high concentrations lead to fast coagulation and growth of the particles, while coagulation is less efficient if the number of particles is small (Pinto et al., 1989). The particle concentrations in the volcanic clouds measured here are low. As an example the ash and sulfate concentrations measured during the first plume encounter after the Kasatochi eruption are ~22 ng/m$^3$ and ~156 ng/m$^3$, respectively. Assuming only 2μm (aerodynamic diameter) ash particles of typical density, thermal coagulation would reduce the number concentration of ash by only ~0.004% during two months, following Hinds (1999). The coagulation rate increases if, as more realistically, a poly dispersed aerosol is assumed, but the rate of aggregation remains small in such low concentrations. The most efficient loss of ash particles < 2μm would be by coagulation with larger particles. However we find no reason to believe that they would exist in considerable larger concentrations than in our measurement range. Instead the most likely interaction is with the sulfate particles that are larger in number and smaller in size. In two months coagulation between 0.6 μm (0.76 μm aerodynamic diameter) sulfate particles and 2 μm ash particles (concentration as above) leads to a change in number concentration by only ~0.3%. In addition coagulation between a 2 μm ash particle and a 0.76 μm sulfate particle would only increase the aerodynamic diameter of the 2 μm particle with ~1.8%, which would barley affect our measurements. To summarize we conclude that coagulation of ash (and sulfate) particles does not have any large effect on the measurements. This motivation is included in the revised manuscript.

Enhanced fallout through ash aggregation has implications for the $SO_2$ lifetime calculation presented in equation (2): the abundance of Fe (presumably associated with the silicate particle component of the aerosol) is related to the abundance of SO4 in the aerosol sample to derive a $SO_2$ lifetime. The underlying assumptions (P.21497 L.19) are: **(1)** the difference in sedimentation rates of ash particles and sulphate particles in the size fraction 0.08-2 microns is assumed to be negligible; **(2)** the amount of ash versus $SO_2$ erupted remains constant throughout the eruption so downwind the proportions remains the same except the change expected as $SO_2$ is converted to SO4; **(3)** the ash and $SO_2$ fractions do not separate during transport in the atmosphere; and **(4)** the cloud can be modelled as a closed system (all mass conserved). The study by Thomas and Prata (2011) demonstrates that this is not the case for the Eyjafjallajökull clouds. Another example can be drawn from extensive study of the Pinatubo 1991 eruption: while there is abundant evidence that the Pinatubo sulphate aerosol persisted for years, it is not clear that fine ash particles (<2 microns) in the aerosol persisted beyond 1 year (e.g., Pueschel et al. 1994). The main issue for the CARIBIC measurements is using successive encounters on different days that sample different parts of the cloud each with a different history of emission and transport. The relative proportions of ash and $SO_2$ co-emitted instantaneously at source vary by a large amount as a function of time during a given eruption. Sometimes gas dominates the mixture, other times more ash is generated. Therefore the source term is not constant, so one would expect far-field spatial variation in proportions of ash and $SO_2$ to simply as a function of variability in the source term.

Below we address the given assumptions (1- 4) above:

(1) Our calculations on the sedimentation velocity shows that this is the case during the 2 months considered, if coagulation is negligible, which we estimate it to be.

(2) Initial fallout that affects the concentrations downwind up to one week does not affect the estimate since our calculations only concern the period after this initial phase. Changed emissions during the eruption leading to different ratios of Fe to S in different part of the resulting volcanic cloud do affect our estimate. This has already been mentioned in the manuscript (P21498 L14-15) and is likely to have caused some variability in the plot. However if this effect were large, the derived function should not be possible to fit to the Fe/S ratios.

(3) According to (1) separation is not likely the case for such small particles and particle concentrations. The Eyjafjallajökull samples are not included in the fit.

(4) Since we use relative values (Fe/S) mixing is not a problem as long as there is no additional source above the background concentrations. Mixing could instead play an important role in "smoothing" variability in the emissions of ash and $SO_2$. At longer time from the eruptions dilution and mixing becomes a larger problem as the concentrations of volcanic material declines towards background conditions. This is the reason that the last samples influenced by the Kasatochi eruption are excluded.

We don't see how the measurements by Pueschel et al (1994) lead to the conclusions that ash and sulfate could not be measured simultaneously during the two months that we consider. The fact that they measure ash up to almost one year after the eruption supports the assumption that separation by sedimentation is slow, instead of the opposite.

What value was used for the loss rate constant? Please add more information.

The loss rate constant for oxidation of $SO_2$ is given by $1/\tau$ ($= 0.222\ s^{-1}$) and was obtained as the result of the fit.

Would this vary as a function of height in the atmosphere and latitude (related to T and water content)?

The altitude and latitude dependence is already thoroughly discussed in the manuscript.

Also, the Fe/S ratios shown in Figure 6 span approximately half an order of magnitude (or more) for a given time step so the fit does not appear to be particularly strong. . .. what is the R2 value?

The $R^2$ value of the fit is 0.71, however we have chosen to give the confidence bounds as an error estimate instead of an $R^2$ value for this nonlinear fit, and wish to continue to do so.

The resulting error in the estimated $SO_2$ lifetime is large (approx. 50 %), which is not altogether unexpected based on the apparent fit.

There are a few published estimates of the residence time of $SO_2$ in the volcanic cloud of Kasatochi. These estimates span from 9 days (Krotkov et al., 2010), to 18 days (Karagulian et al., 2010) and up to 62 days (Jurkat et al., 2010). Thus there is a large spread in estimated residence times. The residence time of $SO_2$ following the Sarychev eruption has been estimated to 11 days (Haywood et al., 2010). Estimations of the conversion rate of $SO_2$ following the Pinatubo eruption is less scattered; 35 days (Bluth et al., 1992), 33 days (Read et al., 1993) and 25±5 as well as 23±5 days (Guo et al., 2004). Except in Guo et al. (2004), uncertainty of the estimated residence time has not been given. Thus it is difficult to compare the validity of our estimate to others. There is a rather large scatter in our data which we cannot change. While earlier estimates obtained for the Pinatubo eruption (satellite based) and by aircraft observations for Kasatochi (Jurkat et al., 2010) lies within our uncertainty limits, the results based on satellite observations of the Kasatochi and Sarychev volcanic clouds are considerably lower. The spread in estimated residence time, both between similar and dissimilar methods, motivate that it is valuable to perform calculations of $SO_2$ residence in different volcanic clouds and by different methods, and not only rely on the more consentient residence time following Pinatubo.

Finally, in order to evaluate the lifetime formulation presented in equations 1 and 2, it would be instructive to compare lifetimes determined using aircraft measurement to lifetimes calculated from satellite remote sensing observations ($SO_2$ retrievals) of exactly the same clouds.

We have now included the estimates of the residence time of $SO_2$ clouds by satellite following the Kasatochi (Krotkov et al., 2010; Karagulian et al., 2010) and Sarychev (Haywood et al., 2010) eruptions given above. It seems like the aircraft observations of the Kasatochi and Sarychev volcanic clouds (Jurkat et al., 2010; this study) result in longer residence times than the satellite based measurements (see above). Haywood et al. (2010) argue that the detection limit of the IASI satellite measurements could lead to somewhat underestimated residence time (less than or ~50%). Measurements following the Kasatochi eruption based on particle detection by OSIRIS onboard the satellite Odin corresponded to a $SO_2$ residence time of approximately 30 days (Bourassa et al., 2010). This points to that the difference between the Kasatochi measurements is connected to whether the measurements are based on detection of $SO_2$ or particles rather than satellite or *in-situ*. Heard et al. (2012) modeled $SO_2$ and aerosol optical depth (AOD). Their results were compared with satellite observations from IASI and OSIRIS for the eruption of Sarychev. In their model $SO_2$ remained longer and the particle concentration rose faster than in the observations. The detection limit of the satellite $SO_2$ retrieval was suggested as a possible cause for the faster decrease of $SO_2$ in observed data, while for the AOD the inadequately modeling of nucleation were proposed as the main reason for the earlier peak of the AOD in the model.

One important difference between the aircraft and satellite observations is that while the *in-situ* measurements are constrained to a well-defined altitude, the remote sensing instruments observes the whole atmospheric column. The altitude of the Kasatochi plume is estimated to 12.5±4 km by Karagulian et al. (2010) and 10-12 km by Krotkov et al. (2010). Mixing with tropospheric air, where residence times are shorter, cannot be excluded and would have affected the satellite based estimates. Krotkov et al. (2010) suggest the rater low altitude of the Kasatochi plume and the dynamic UT/LS in the extra tropics as an explanation for their shorter residence time (9 days) than that observed after the Pinatubo eruption. For the evaluation of the residence time estimation in our work, the discussion in the two last sections highlights important aspects that should be included in the revised manuscript.

## Technical corrections

P.21490 L.12 (and elsewhere) Please use UK spelling of "sulphur" for this European Journal

P.21492 Please add some dates/times that the CALIPSO observations correspond to on Figure 2

P.21494 L.13 change "volcanically" to "volcanic"

P.21498 L.8 The composition of the aerosol is most strongly affected by the gases released during the eruption (derived from magmatic volatiles) – these aren't mentioned.

P.21501 L.6 Change "baring" to "bearing"

The requested corrections for have been performed.

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