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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Composition and evolution of volcanic aerosol from eruptions of Kasatochi, Sarychev and Eyjafjallajökull in 2008–2010 based on CARIBIC observations" *by* S. M. Andersson et al.

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General comments

Source attribution of aerosol samples collected during the CARIBIC flights is complex given the lack of a priori knowledge of the history of air masses prior to encounter. In this study, the authors calculate back-trajectories and highlight aerosol composition deviations from background to imply a volcanic origin. The aircraft encounters with air masses contended to be of volcanic origin occurred 1000s km from source, many

days or weeks after emplacement. For this reason there is a strong likelihood of mixing with other potential aerosol sources such as wildfires. The authors use temporal measurement of Fe and SO4 in aerosol samples to calculate a SO2 lifetime. However, the underlying assumptions of this calculation include a constant source term with respect to ash and SO2 emission, and negligible fractionation during transport, may be unjustified. Some specific comments follow.

Specific comments

(1) 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

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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.

(2) 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 analysed the Eyjafjallajökull volcanic clouds and determined that ash and sulphur species may co-exist or be mutually exclusive.

(3) 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.

(4) 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

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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 SO2. On P.21501 L.1, "During the 2.5 months considered here, the 2 μm ash particles fall a distance of less than 15

(5) Enhanced fallout through ash aggregation has implications for the SO2 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 SO2 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 SO2 erupted remains constant throughout the eruption so downwind the proportions remains the same except the change expected as SO2 is converted to SO4; (3) the ash and SO2 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 SO2 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 SO2 to simply as a function of variability in the source term. What value was used for the loss rate constant? Please add more information. Would this vary as a function of height in the atmosphere and latitude (related to T and water content)? Also, the Fe/S ratios shown in Figure 6 span approximately half an order of magnitude (or more) for a given

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time step so the fit does not appear to be particularly strong.... what is the R2 value? The resulting error in the estimated SO2 lifetime is large (approx. 50 %), which is not altogether unexpected based on the apparent fit. 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 (SO2 retrievals) of exactly the same clouds.

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"

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 21481, 2012.