

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 Referee #1

We thank the referee for the positive review of our work, and for posing important questions. We answer these in the text below (in red).

This manuscript describes the collection and chemical analysis of volcanic aerosol in the lower stratosphere /upper troposphere using an airborne platform and presents an approach to estimate the atmospheric residence time of volcanic sulfur dioxide from the experimental data. Considering the general difficulty of obtaining volcanic aerosol samples directly in a plume, the CARIBIC platform represents an ideal tool for such investigations, the collected data set is very comprehensive and of large value for the scientific community. The description of the applied techniques as well as the presentation of the data is scientifically sound, and I recommend the manuscript for publication in ACP after addressing one critical issue regarding the interpretation of the data. The authors calculate the residence time of SO₂ with help of the Fe/S mass ratio. They argue correctly that the obtained residence time is underestimated due to a higher sedimentation velocity of ash compared to the sulfate particles. Furthermore, the authors also point out that e.g. within the Eyjafjallajökull plume, the ash mode diameter was considerably higher even far away from the source (larger than 2 micrometers, see e.g. airborne measurements by Schumann et al. (2010) and Bukowiecki et al. (2011, same ACP special issue, airborne measurements) than measurable with the size cutoff of the CARIBIC instrumentation (2 micrometers). This means that, at least for the Eyjafjallajökull data, only the lower size end of the ash mode was analyzed. Doesn't this have a drastic influence on the Fe/S mass ratio which is subsequently used for the calculation of residence times?

As the referee correctly infers only a fraction of the size distribution of the ash is measured, especially in the Eyjafjallajökull volcanic cloud. However the residence time calculation is based on samples influenced by Kasatochi and Sarychev. These samples were collected more than one week after the eruptions and it is likely that most of the large particles have been removed. On the other hand it does not affect our calculations that only a fraction of the ash is measured, as long as the size distribution does not change considerably since our method demands that we sample the same aerosol fraction in each measurement. This is one important reason to exclude the Eyjafjallajökull samples from the calculation of residence time, as the high concentrations in these samples imply that interaction between particles and thereby fractionation is more efficient than in the low concentrations measured after the Kasatochi and Sarychev eruption.

Furthermore, what does a change of Fe mass from one filter to the next mean? Is it a) due to a change in plume concentration (dilution), or is it b) rather due to a change in size distribution (fractionation)? What impacts do these two scenarios a) and b) have on the overall findings, and is it possible at all to separate these two situations?

A change in measured concentration is most likely caused by dilution of the volcanic cloud and/or by how long the aircraft encounters it, i.e. how large fraction of the sampling that is performed in volcanically influenced air. This does not affect our results as we use relative values (Fe/S). We have no method to measure if there is a change in the size distribution, but estimate that this is of minor importance for the measured concentrations. Coagulation in a volcanic cloud act as a limiting effect on the number concentration as high concentrations leads to fast coagulation and growth of the particles, while coagulation is less efficient if the number of particles are small (Pinto et al., 1989). Particle concentrations in the first sample taken more than one week following the eruptions of Kasatochi are ~22 ng/m³ ash and ~156 ng/m³ sulfate. This leads to very low coagulation rates, especially between the larger ash particles which would else cause them to quickly grow outside the collection range. The effect of thermal coagulation can be calculated following Hinds (1999), in the simplest case assuming only 2µm (aerodynamic diameter) ash particles of typical density, which leads to a decrease in number concentration by only ~0.004% during the two months considered in the residence time calculations.

The most efficient sink for ash particles $< 2\mu\text{m}$ would be by larger particles. However we find no reason to believe that they would exist in considerable larger concentrations than in our measurement range. Changed size distribution by coagulation is an important aspect that has now been included in the manuscript.

These questions need to be addressed in more detail by the authors; otherwise the deduced findings remain somewhat diffuse. I suggest including a estimation of uncertainty (at least semi-quantitative), if this is not possible the limitations of the interpretation should be clearly emphasized throughout the manuscript (also in the abstract).

An error estimate of the residence time is already given, this is now included also in the abstract. We have also included a description on fractionation. The ash concentrations are low in the Kasatochi and Sarychev volcanic clouds encountered. Based on calculated coagulation rates its effect on the estimate of the SO_2 conversion rate is estimated to be minor.

References

Hinds, W. C.: Aerosol technology: properties, behavior, and measurement of airborne particles, Book, John Wiley & Sons, New York, 1999.

Pinto, J. P., Turco, R. P., and Toon, O. B.: Self-limiting Physical and Chemical Effects in Volcanic Eruption Clouds, J. Geophys. Res., 94, 11165-11174, 1989.