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Interactive comment on "SO_2 and BrO observation in the plume of the Eyjafjallajökull volcano 2010: CARIBIC and GOME-2 retrievals" *by* K.-P. Heue et al.

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Interactive comments on "SO_2 and BrO observation in the plume of the Eyjafjallajökull volcano 2010: CARIBIC and GOME-2 retrievals" by K.-P. Heue et al.

We thank both referees for the helpful comments and suggestions, both referees suggested to shorten the paper, therefore we deleted 4 figures and 2 were modified. Also the DOAS sections were shortened. On the other hand a small discussion of the BrO to SO_2 ratio (1 new figure) is now included, and some details are clarified.
The specific answers to the comments are written below the comments in italics. Sometimes two comments are answered simultaneously.

Anonymous Referee #1
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This paper reports on satellite and aircraft measurements of SO_2 and BrO in a volcanic plume following the 2010 eruption of the Eyjafjallajökull volcano. It appears to be a carefully done piece of work. The measurements are of high quality and the methodology is rigorous. I believe this paper should be published, but after addressing a number of points. Specifically, I find the paper rather long and technical. In its current state, it would probably be more appropriate for Atmos. Meas. Tech. Although I understand this paper has been written to be part of the special issue dedicated to the atmospheric implications of the Eyjafjallajökull, I found the paper rather weak in the conclusions for a journal like Atmos. Chem. Phys.

General comments

1) I find disappointing that the authors limited the discussion to general statements, in substance 'SO_2 and BrO could be measured in the plume using CARIBIC' and 'Both SO_2 and BrO observations agree well with simultaneous satellite observations'. As there is currently substantial interest in measurements of volcanic BrO, I expected (before reading) that the authors would try to interpret their results a bit more. Personally, I would have found interesting to relate these observations to other published BrO results (e.g., Bobrowski et al., 2003, 2007) and say more on the chemistry in the plume at the time of the measurements (e.g., via the BrO: SO_2 ratio). Moreover, it is currently not fully clear in the scientific community why BrO is detected for some volcanoes and why not for others. As BrO could be detected using GOME-2 only for several days among more than a month of eruption of Eyjafjoll, it could be very interesting to relate the BrO observations to the different phases of the eruption, in order to better understand the origin of bromine in volcanic plumes.

A short section on BrO to SO_2 ratio was added to the discussion of the CARIBIC data. For a more detailed discussion of the BrO/SO_2 ratio the two observations with

15 km distance are not sufficient. The study suggested by the referee to correlate the GOME-2 BrO/SO_2 with the eruption phase would certainly be very interesting, and will hopefully be made soon; but it would not fit in the context of this "CARIBIC" paper. Moreover, the first results obtained by dividing the vertical column densities illustrate that this simple approach is not very useful.

2) From my knowledge, it is the first time that volcanic BrO has been detected from an aircraft DOAS instrument. In my opinion, the authors should briefly explain what is exactly/can be the added-value of such aircraft measurements with respect to other observations (using ground-based or satellite DOAS instruments) for (future) studies of volcanic bromine emissions.

Unfortunately, this not the first time BrO was observed with an airborne DOAS instrument, there is a paper by Bani et al. 2009. The reference is added to the manuscript, together with a short section about the advantages of aircraft measurements in volcanic plumes, i.e. high spatial resolution compared to satellite, in contrast to ground based observations measurements can be performed at various distances from the crater, quasi Lagrangian observations can be made. . . Using a DOAS instrument offers the additional advantage, that the aeroplane can fly over or under the plume (as done in this study as well) thereby the risk for the crew is minimized.

Bani, P., Oppenheimer, C., Tsanev, V. I., Carn, S. A., Cronin, S. J., Crimp, R., Calkins, J, A., Charley, D., Lardy, M., Roberts, T. R.:Surge in sulphur and halogen degassing from Ambrym volcano, Vanuatu, Bull. Volcanol., 71, 1159 – 1168, doi: 10.1007/s00445-009-0293-7, 2009.

3) In general, an effort must be done to improve the readability of the paper (see below for some specific comments). Although I understand that the authors would like to show their other (nice) CARIBIC data (mercury, hydrocarbons, ...), I am not convinced it really brings something to the discussion on SO_2 and BrO.

The referee is right, that the mercury and the ozone data not showing any significant

signal that might be attributed to the volcano and are not necessary. Hence these data are omitted from the overview and the discussion. The detailed discussion of the hydrocarbons will be given elsewhere so they were also removed. Although these findings are in themselves very interesting and the reduction of the NMHCs might be partly caused by volcanic bromine.

Specific comments

-As far as satellite detection of volcanic BrO is concerned, a reference to the work of Theys et al., 2009 (GRL) should be added.

Yes, definitely. The reference was added.

-In section 2.2 no details on RTM settings is given for the calculation of AMFs. It would be good to mention somewhere that the latter will be provided in sect 4.4 (3.2).

Done.

-An error of 7% is associated to the AMF but if I understand well it is a statistical error coming out of the Monte Carlo RTM. It doesn't include any error acting systematically due to parameters (albedo, clouds, ...) of the RT simulations. This error of 7% is then used directly to estimate the VCD error just like it would include all sources of errors. Please clarify.

The error in the AMF mentioned here is the statistical error only. The error caused by incorrect settings (wrong plume altitude or cloud cover) can be much higher, therefore section 3.2 documents all the available information.
A respective sentence is added in the paper. See also comment (1) from referee 2.

-Fig. 1: It is clear from the SO_2 residuals that remaining O_3 absorption structures are present. Although it probably doesn't affect too much the quality of the SO_2 SCD data (as the SO_2 signal is strong), it is not mentioned at all. For BrO, I am not convinced it is a good fit. I am concerned by what happens below 335 nm where O_3 signatures can clearly affect the quality of the fit. This must be addressed. The retrieval of O_4 looks

particularly bad in Fig 1!

Unfortunately, there is indeed a systematic structure in the residual of SO_2 fit which is probably caused by an imperfect O_3 fit, but as the referee wrote himself the SO_2 absorption is strong, and therefore the effect on the SO_2 column is low. Concerning the BrO fit we found no influence of the wavelength interval on the residual and the BrO SCD as long as the lower limit is above 320nm. There are better O_4 fits for the -10° viewing direction (here nadir is shown) but as the noise is higher in this viewing direction, we decided to show the fits for this spectrum. From my point of view all the fits should be from the same spectrum rather than showing a few strong absorptions for different spectra.

-Is there a specific reason why not using the same fitting window/settings for CARIBIC and GOME-2 (especially for SO_2)?

No, except, that the spectra are evaluated by different persons. We also did a comparison for the CARIBIC DOAS spectra and retrieved the SCD with the same settings as used for GOME-2, and found no significant difference.
Accordingly, a statement concerning the comparison was added.

-The reader has no possibility to evaluate the quality of the fitting results for GOME-2 (neither SO_2 nor BrO). A similar figure as Fig.1 should be added.

The focus of this paper should be the CARIBIC measurements; therefore a respective figure will not be included here. A publication on BrO and SO_2 observation from GOME-2 is in preparation, it might be included there.

-L 211: please further explain why an O_3 cross-section scaled with a polynomial is used? How this procedure can account for the dependence of the atmospheric light paths with wavelength?

The second O_3 reference spectrum was included to consider the wavelength dependence of the O_3 AMF caused by Rayleigh scattering (see e.g. Pukite et al., 2010).

Especially for strong ozone absorption at large SZA the fit residual could be reduced including this second O_3 reference spectrum.

Pukite, J., Kühl, S., Deutschmann, T., Platt, U., and Wagner, T.: Extending differential optical absorption spectroscopy for limb measurements in the UV, Atmos. Meas. Tech., 3, 631-653, doi:10.5194/amt-3-631-2010, 2010.
The reference was added to the explanation.

-L 244-248: it is unclear what AMF has been used for the satellite data (geometric AMF or RT sim).

Both. For the standard satellite products as shown in the figure 9 (formerly 11 and 12) the geometric AMF is used, for the comparison (section 4.2) new AMF were calculated based on the information retrieved from the CARIBIC observations and additional sources (e.g. MODIS).
This fact is clarified in the text.

-L288-end of sect 3.2: I don't understand why Fig. 7 shows only a comparison between O_4 SCDs* (calculated) and SO_2 SCDs (measured)? Although both data sets shows a clear anticorrelation which is a good point, there is no way for the reader to judge if the O_4 SCDs * agrees well with the measured O_4 SCDs!

First of all, the O_4 SCD are not the calculated column densities. The star was intended to distinguish between a column density of trace gas with a "known concentration" having the unit molec/cm^2 and O_4 with the absorption being linear to the square of the O_2 concentration, which results in the unit molec^2/cm^5. But as this only caused confusion and also in other publication O_4 is treated as normal trace gas, the star was removed from all O_4 SCDs.*
Results of the simulations are added to figure 6 (formerly 7).

-L 311: I am bit surprised by the value of 0.95 for the single scattering albedo. I would have expect a lower value as the ash emitted by Eyjafjoll are quite absorbing aerosols.

Please comment.

I was surprised myself, when I found, that the best agreement between the measurements and the simulation was found for such a highly reflecting aerosol. But then I read the paper by Schuman et al. where they said that a low or non absorbing aerosol was found. It also agrees well with the SSA estimates from the third CARIBIC volcano flight based on the OPC measurements.

-In my opinion, Figure 11 is useless. Simply state that many other satellite instruments could similarly observe the SO_2 plume during this period (and maybe add a reference to the SACS website or another site).
-Figure 12 could be displayed for the whole Western Europe (just as Figure 11). This would strengthen the GOME2 BrO results if one could identify BrO close to the volcano as well.

These two comments are bit contradicting, showing the SO_2 plume over north western Europe is useless, but at the same time the BrO plume for the same region should be included. We combined the two pictures, but still try to give the same information. Therefore four small pictures are shown, SO_2 and BrO for the complete plume, including Iceland, and a detailed zoom on the British Isles, that only shows the orbit being closest in time relative to the CARBIC observation. Unfortunately the picture of the OMI data had to be removed.

-I found that the GOME-2 BrO results were not very well used. I would find very interesting to study the BrO:SO_2 ratio based on GOME-2 elsewhere than in vicinity of the CARIBIC observations area (e.g. close to the volcano) and at other dates than the 16th November.

The study the referee suggested here would certainly be very interesting; however, the focus of this paper is on the CARIBIC observation and a short comparison to GOME-2. The suggested comparison will be an independent study and hence published elsewhere. For the small area around the CARIBIC observations, the BrO to SO_2 ratio

from the GOME-2 data are included.

-Add a short discussion on the BrO/SO_2 ratio estimate you obtain. How does it compare with other estimates in the literature.

The referee is right that the BrO/SO_2 ratio is of high interest for chemical simulation studies of volcanic plumes, mainly because in the short periods of several hours to a few days, the total SO_2 amount can be seen as constant, while BrO is build up in the plume. Hence SO_2 is a tracer for mixing in and BrO is the reactive species we are interested in. The ratio gets most interesting when it is studied along the plume in a "Lagrangian" experiment for a longer period. On the other hand, a combination of several individual measurements like this study can also be used for chemical transport simulations. A small section on the ratio was added and with $1.3 \cdot 10^{-4}$ it is within the range reported by Bobrowski and Platt (2007) for other volcanoes.

Bobrowski, N. and Platt, U.: SO_2/BrO ratios studied in five volcanic plumes, J. Volcanol. Geotherm. Res. 166, 147-160, doi:10.1016/j.jvolgeores.2007.07.003, 2007.

Minor comments

-section 2.2: I found perturbing to first give a background on DOAS, then introduce the instrument and then present the DOAS settings. It would help the reader to first introduce the instrument and then talk about DOAS (background and SO_2 and BrO settings).

I reorganised the DOAS sections and reduced the details for all the fit windows - including GOME-2 to one table.

-page 6: footnote - this sentence is a little bit ambiguous. What is "normal" O_4 SCD? Star means the true O_4 SCD? While 'no star' means measured O_4?

I am not sure the referee had downloaded the actual version of the manuscript as this comment refers to the first version submitted to ACPD, where the explanation was a footnote. {ß Anyway, as stated above the stars are now removed from all O_4 SCD.

-L194-201: at this stage of the paper, it is hard to understand the role of Figure2.

I am sorry about that, especially as I think, that showing figure 2 here, helps to understand figure 14 (formerly 17) and table 4. The picture is not changed.

-Fig 9 has to be adapted. It is difficult to read. Please increase the width of lines.

The line width was increased in the version on ACPD.

-Harmonize the color bars of Figs 6 and 13 to facilitate direct comparisons.

Both referees suggested to reduce the size of the paper (and the number of figures), therefore we removed figure 13. The differences in the column densities can also be seen in Figure 6.

Typos L30: "distriubtion" ! "distribution"
L121: "Togetehr"! "Together"
L122: "programm"! "program"
L133: "colud" ! "could"

Corrected.

Caption Fig. 4: "The flight pressure altitude"! "The flight altitude" (?)

It is not trivial to measure the altitude of an aeroplane, therefore the pressure is measured and an altitude is calculated using a standard atmosphere. That means the flight altitude is defined as the pressure altitude. The word "pressure" was removed.

L411: (Bobrowski et al., 2007, e.g.) ! (e.g., Bobrowski et al., 2007)
Caption Table 2 : BrO error of 1.8 ppb! 1.8 pptv

Corrected.

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