Interactive comment on "Systematic investigation of bromine monoxide in volcanic plumes from space by using the GOME-2 instrument" by C. Hörmann et al.

Anonymous Referee #1
Received and published: 18 December 2012

This paper provides an extensive study of BrO emissions from volcanoes using satellite data. It appears to be a carefully done piece of work and the results are of high quality. I believe this paper should be published, but after addressing a number of (minor) points. Specifically, I find the paper rather long and it includes a lot of repetitions. The English could also be improved.

We would like to thank referee #1 for the detailed and helpful comments and suggestions he/she made to improve the quality and clarity of our manuscript. As suggested by the referee, we now merged the discussion and conclusions sections to avoid repetitions. Furthermore, the text was amended in several places to improve speech quality, as listed below.

For reference, the original comments (**black**) are always included below followed by our response (**red**). Modifications of the manuscript are indicated in **green**.

Specific comments

-Introduction: please explain why the BrO/SO2 ratio is an important parameter.

The introduction already contained a segment on the ability of the BrO/SO_2 ratio to indicate the activity state of a volcano as it has been suggested by Bobrowski and Giuffrida (2012). While this application was, so far, restricted to ground-based measurements close to the crater of a passively degassing volcano, satellite observations of the BrO/SO_2 ratios during volcanic eruptions may lead to important additional conclusions regarding the applicability of BrO/SO_2 long-term measurements for the possible prediction of volcanic eruptions. Our results show that the BrO/SO_2 ratios during minor/major eruptions are not significantly different from the ones from ground-based measurements at non-erupting active volcanoes. However, future observations of volcanic emissions by both ground-based and satellite observations will be necessary to improve our knowledge on bromine chemistry in volcanic plumes and possible conclusions regarding the application of this parameter to predict volcanic eruptions.

We further added the following explanation to the introduction

„As SO_2 is relatively inert compared to BrO, it can e.g. be used as a tracer to investigate ongoing BrO chemistry in ground-based measurements close to a volcano. The BrO/SO_2 ratio can be used as an indicator for the evolution of BrO in a plume, as aspects like plume dilution by ambient air, plume dispersion and varying emission strengths of the volcano are similar for both species and thus cancel out (Vogel, 2012)."

Additional reference:

Vogel, L.: Volcanic plumes: Evaluation of spectroscopic measurements, early detection, and bromine chemistry, PhD thesis, Combined Faculties for the Natural Sciences and for Mathematics, University of Heidelberg, 2012.

and additionally to the discussion (Sect. 6.2):

"Satellite observations of the BrO/SO_2 ratios during volcanic eruptions may nevertheless lead to important additional conclusions regarding the applicability of BrO/SO_2 long-term measurements for the possible prediction of volcanic eruptions."

-Page 5, l143: a second O3 cross-section is used which is the original cross section scaled by a 4th order polynomial: this last step is unclear. Please provide a reference.

We updated the text to:

„In addition to the original O_3 absorption cross section, a second one (the original cross section scaled with λ^4) was included to consider the wavelength dependence of the O_3 AMF caused by Rayleigh scattering (e.g. Van Roozendael et al., 2006a and Puķīte et al., 2009). By including this second O_3 reference spectrum, the fit residual can be reduced appreciably, especially for strong ozone absorption at large SZA."

Additional reference:

Puķīte, 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, 2010, doi:10.5194/amt-3-631-2010

-Section 2.2: I think the explanation on why SO2 SCDs are underestimated in the SR is misleading. The statement "only the outermost layers of the volcanic plume are actually penetrated by the incident sunlight, and no light from inner parts of the plume or below is detected within the analyzed wavelength range" is incorrect. Instead, I would say that "For large SO2 column amounts, the atmosphere cannot be considered as optically thin as for the SO2 absorption. The penetration of light in the plume becomes strongly wavelength dependent, causing an underestimation of the retrieved SO2 SC in the SR". I would also avoid using the word "saturation". Before encounter saturation effects, first "non-linear retrieval effects" are faced.

We agree with the referee that the explanation might be misleading and that the SO_2 concentrations in volcanic plumes are usually not high enough to prevent the penetration of a plume's inner parts. However, we also think that such situations are indeed possible during major volcanic eruptions. Radiative transfer studies for the Kasatochi eruption by Bobrowski et al. (2010) showed that the AMF can become essentially zero for SO_2 concentrations larger than $1 \cdot 10^{13}$ molec/cm³. Nevertheless, this effect is typically reduced by light that is scattered by aerosol particles, which are usually present during such strong events. We therefore changed the text as suggested by the referee:

„For such cases, the atmosphere cannot be longer considered as optically thin for the standard SO_2 wavelength fit range (in this study 312.1-324 nm). The penetration of light in the plume becomes strongly wavelength dependent, resulting in low-biased SO_2 SCDs due to non-linear effects. In extreme cases, only the outermost layers of the volcanic plume might be actually penetrated by the incident sunlight, and no light from inner parts of the plume or below is detected within the analysed wavelength range. However, this effect is usually reduced by light that is scattered by aerosol particles, so that the SO_2 absorption signal is very weak but not zero (Bobrowski et al., 2010)."

Later on, the sentence "Also, usually insufficient knowledge on aerosol and cloud properties is available, which also affect the measured spectra." might make the reader think that aerosols and clouds are not important effects in the 326.5-335.3 nm range (which I think is not what you meant).

We agree with the referee that this sentence might be misleading and removed it from the manuscript.

-Section 3.3: The offset correction may be corrupted by elevated SO2 distributed zonally (e.g. Kasatochi). Of course, taking the median is already mitigating the error, but still, it can have an effect for very concentrated plumes of SO2. Please provide an estimate of the maximum error associated to this correction.

The initial offset correction applied to the gridded GOME-2 data may indeed be corrupted by locally strong enhanced SO_2 VCDs. However, this "first" or "initial" offset correction in Section 3.3 is used as a rough estimation for the (mostly) latitudinal dependent offset that is caused by spectral interference with ozone. The roughly offset-corrected gridded data is therefore only used to determine the greater area of measurements that is affected by a volcanic plume.
In extreme cases, a volcanic plume with very high SO_2 SCDs that extends along large parts of a certain latitude may lead to the problem that another volcanic event at the same latitude and with much lower SO_2 SCDs might not be detected by the algorithm. However, such cases are extremely unlikely, because large plumes usually have strong horizontal gradients.

-Section 3.4:
* What brings the SO2 2D correction, knowing that there is already an offset correction applied (section 3.3)? No real information is given. In its current form, this correction seems a bit redundant. The SO2 distribution has (in principle) no background varying in latitude and longitude (in contrast to BrO for which a 2D correction makes a lot of sense).

There seems to be the misunderstanding that the 2D correction is applied additionally to the initial offset correction in Section 3.3. This is not the case:
In contrast to the offset correction that is applied to the gridded GOME-2 data (please compare to previous comment), the 2D offset correction in Section 3.4 is locally applied to the uncorrected original GOME-2 measurements (not the gridded data from Section 3.3). As the calculation of the 2D polynomial first excludes all satellite measurements with enhanced SO_2 VCDs, regional effects are taken into account more precisely (especially for the BrO offset). Finally, this 2D offset corrected data set is used for the BrO/SO_2 analysis.

However, we tried to make the differences of both offset correction processes clearer by minor modifications of the associated text.

„In contrast to the rough background correction process for the gridded satellite data in Sect. 3.3, the lat-/longitudinal offset was now corrected in a more sophisticated way."

*The extraction of the BrO data relies on the extraction of the SO2 columns (exceeding 3 sigma). By doing this, one would then ignore pixels with significant BrO and near zero SO2. How does it impact the analysis of Section 4 (BrO/SO2 ratios)?

We agree that by using SO_2 as a tracer for the plume affected measurements, possibly enhanced BrO VCDs at plume regions with near-zero SO_2 VCDs would be ignored. Nevertheless, we think that enhanced SO_2 VCDs are the best available indicator for the presence of a volcanic plume, as SO_2 is usually one of the dominant gaseous species. In addition, a visual inspection of our GOME-2 data set showed only one possible case of enhanced volcanic BrO in absence of SO_2 (Etna on 14 May 2008; events #97, Figure 6 in the discussion paper). However, it should be noted that the associated BrO SCDs are of the same magnitude as the background signal further away from the plume area and a clear verification of the enhancement therefore remains difficult (compare to our reply to referee #2).

We added the following text (Sect. 4.1):

„On closer examination, it can further be seen that a small part of the plume close to the volcano shows enhanced BrO SCDs in absence of enhanced SO_2 and is therefore not included in the BrO/SO_2 analysis. According to a visual inspection of all volcanic plumes investigated within the scope of this paper, this is a one-time-only event. As the corresponding BrO SCDs are close to the BrO detection limit, a clear verification of the enhancement remains difficult."

Why not extracting also the BrO data based on the BrO values in the PEBs (e.g. exceeding 2-3 sigma)?

For the systematic analysis of all investigated volcanic plumes (Section 5), the abundance of small clusters of enhanced BrO VCDs >2 sigma is already one key parameter to positively identify enhanced BrO columns within the plume area. As the offset correction is also applied to the plume pixels (and not only the reference area), the analysed satellite measurements may include pixels that show no enhanced BrO. However, this does not significantly influence the BrO/SO_2 ratio analysis by the linear fit as the BrO SCDs are scattered around zero for such cases.

-Section 3.5: the SCD threshold value of $1x10^{18}$ molec/cm² is arbitrary for the reader. Please justify.

Based on the experiences for a case study on the influence of different SO_2 evaluation wavelength ranges for the Kasatochi eruption in 2008 (Bobrowski et al., 2011), we found $1x10^{18}$ molec/cm² to be a plausible threshold.

We added a detailed discussion on the determination of the SO_2 SCD threshold in the Supplementary Material and a reference to that material in the text.

-Section 4: Fig. 5.

*Time-series of total masses rather than VC would make more sense here (it would also be complementary to the scatter plots shown throughout the paper).

We added a new figure to the manuscript that shows total SO_2 and BrO masses for all volcanic events (new Figure 6). In order to give an estimate on the significance of the results, we highlighted all BrO total masses that were found to be more than twice as large as the associated error in blue, while other data appear in red.
In a similar way, the maximum BrO VCDs at more than two times the individual fit error were indicated in Fig. 5. For the SO_2 data, this condition was fulfilled for all volcanic events.

The text was adjusted accordingly.

*Table 2 is not necessary.

Table 2 is supposed to give a short overview of the subsequently discussed volcanic plume events. We think that it helps to link the maximum VCD (total masses) time series to the following examples and therefore decided to keep it in the paper.

-Section 4.4. The BrO/SO2 ratio of 1.8e-5 is much lower than the previous estimates (Sects 4.1-4.3). In the meantime, there are two plumes for Nabro (on 16.06.2011) indicating two different altitudes. Therefore VCDs (corrected for the effect of altitude) should be used for the scatter plots, instead of SCDs. I wonder if it might give a BrO/SO2 ratio more in line with the previous estimates.

We agree that the SO_2 plume for Nabro on the 16 June 2011 mainly consists of two plume layers at different altitudes:

1) the plume that reaches towards western direction and then extends over the Asian continent at ~10-12 km
2) a plume that extends towards southern direction at ~4-5 km and were most of the enhanced BrO column densities showed up

as it has been shown e.g. by Theys et al. (2013).

A precise separation of both plumes remains difficult, especially in the overlapping area close to the volcano. However, as long as the BrO/SO_2 ratio is not significantly different for both plumes, the AMFs of both species mainly cancel out if the ratio is calculated. Nevertheless, we reanalysed our data for the very southern part of the plume (where an overlap with another plume layer can be excluded), but found no significant differences for the BrO/SO_2 ratio (respectively the correlation plot) compared to the consideration of the whole area with enhanced BrO SCDs.

We updated the text as followed (including additional suggestions by Robin Campion):

“Another possible reason for the different distributions of both species might be that the plume close to the volcano consists of two layers at different altitudes (Theys et al., 2012). Most of the enhanced BrO SCDs belong to a plume layer that is located at lower altitudes (4-5 km) and extends towards southern direction, while the dominating part of the plume is located at 10-12 km and no BrO was detected. A precise separation of both plume layers remains difficult, especially in the overlapping area close to the volcano. However, the data were reanalysed for the very southern part of the plume (where an overlap with the plume layer at higher altitudes could be excluded), but no significant differences for the BrO/SO_2 ratio (respectively the correlation plot) were found in comparison to the consideration of the whole area with enhanced BrO SCDs. The plume composition at different altitudes might vary due to different volcanic processes, such as energetic lava fountains during strong explosions (for the plume at higher altitudes) and residual degassing of lava flows (plume at low altitudes). Such mechanisms are known to produce differences in the ratios of SO_2 and halogen species like HCl or HF (e.g. Burton et al., 2003; Bobrowski and Giuffrida, 2012; Ohno et al., 2013), so that this might also be possible for the BrO/SO_2 ratio.

Additional references:

Burton, M., Allard, P., Mure, F., and Oppenheimer, C.: FTIR remote sensing of fractional magma degassing at Mount Etna, Sicily, Geological Society, London, Special Publications, 213, 281 –293, doi:10.1144/GSL.SP.2003.213.01.17, 2003.

Ohno, M., Utsugi, M., Mori, T., Kita, I., Kagiyama, T., and Tanaka, Y.: Temporal variation in the chemical 1000 composition (HCl/SO_2) of volcanic gas associated with the volcanic activity of Aso Volcano, Japan, Earth, Planets and Space, 65, e1–e4, doi:10.5047/eps.2012.11.003, 2013.

Theys, N., Campion, R., Clarisse, L., Brenot, H., van Gent, J., Dils, B., Corradini, S., Merucci, L., Coheur, P.-F., Roozendael, M. V., Hurtmans, D., Clerbaux, C., Tait, S., and Ferrucci, F.: Volcanic SO_2 fluxes derived from satellite data: a survey using OMI, GOME-2, IASI and MODIS, Atmos. Chem. Phys. Disc., 12, 31 349–31 412, doi:10.5194/acpd-12-31349-2012, 2012.

-Section 4.5. Kasatochi:

*The differences in the BrO and SO2 patterns are due partly to the differences in injection profiles. At least, this should be mentioned.

We agree that the differences in the BrO and SO_2 patterns are possibly caused by different injection profiles and added the following sentence:

„Generally, the differences between the SO_2 and BrO distributions during the eruption may also be partly caused by differences in the injection profiles of the individual explosions."

*for complex events as Kasatochi (and others investigated in this paper), it would make sense to look at the BrO/SO2 ratio in terms of total number of molecules (i.e. integrated) rather than the individual columns. Also interesting is to investigate how this BrO/SO2 ratio is evolving with time (age of the plume).

We thank the referee for this good suggestion and added BrO/SO_2 ratios in terms of total number of molecules for the individual determined volcanic plume pixels to all correlation plots and tables within the manuscript and the Supplementary Material as an additional estimate for the mean BrO/SO_2 ratio within the volcanic plumes. We further agree that the investigation of the BrO/SO_2 ratios evolving with time would be a very interesting task, but think that this should be done within another study in the future.

-Section 4.6. Sarychev: other examples are shown in the auxiliary material also showing different BrO-SO2 patterns. Maybe good to mention in the text.

We added „(other days during the eruption showed also different BrO/SO_2 patterns; see Supplementary Material)" to the text.

I found the last sentences of Section 4.6 rather vague. There is no indication that meteorological conditions explain the differences in patterns between SO2 and BrO. Instead, the author should consider differences in injection heights as a real option.

We reformulated the last sentences as followed:

„One explanation for this behaviour could be that different volcanic mechanisms were involved during the individual explosions (compare to Sect. 4.4.), leading to different injection heights and variable plume compositions. Furthermore, the bromine chemistry could be influenced by the meteorological parameters at different altitudes (e.g. temperature and relative humidity), which might have a crucial influence on the BrO formation process in addition to plume conditions like the presence of aerosol particles."

-Section 5:

* p-value?

The p-value "*...is associated with a test statistic. It is the probability, if the test statistic really were distributed as it would be under the null hypothesis, of observing a test statistic (as extreme as, or more extreme than) the one actually observed*" (Davidson and MacKinnon, 1993).

The smaller the p-value, the more strongly the test rejects the null hypothesis. In the context of the BrO/SO_2 analysis, the p-value indicates the result for testing the hypothesis of no correlation. Each p-value is the probability of getting a correlation as large as the observed value by random chance, when the true correlation is zero. The smaller the p-value, the more significant the correlation gets. Unlike the r^2 value, the p-value takes into account the total number of measurements. However, the statistical significance that was used for the different categories was empirically determined according to typical values that we found for volcanic events with clearly enhanced BrO SCDs at the same area as the enhanced SO_2 SCDs.

Additional reference added to the text:

Davidson, R. and MacKinnon, J. G.: Estimation and Inference in Econometrics, Oxford University Press, 1993.

*for plumes with small number pixels and/or measured BrO VC close to the detection limit (Fig 14 as an example among others), it is really hard to be conclusive (if not impossible). Please provide error bars on the BrO/SO2 ratios that takes into account reasonable values for the scatter on the BrO and SO2 SCDs. Also, sometimes one have very large r^2 but this is the case because there are only few points. This makes the definition of the categories (Table 3) questionable.

We included error bars to all correlation plots within the manuscript and the Supplementary Material by using the individual SO_2 and BrO fit errors of the DOAS analysis (the systematic errors mainly cancel out by taking the BrO/SO_2 ratio). Furthermore, the fit errors were also included in the bivariate linear fit analysis, yielding an estimate of the BrO/SO_2 ratios error which is now given inside the correlation plots and all tables throughout the manuscript and the Supplementary Material. For plumes with a small number of pixels and/or measured BrO SCDs close to the detection limit, the now included BrO/SO_2 ratio errors indicate the uncertainties of the ratio determined from the linear fit method.

*The categories are a bit surprising as a given volcanic event can be classified in two categories at the same time no matter if the eruption was strong or not.

The classification is strictly based on the phenomenological behaviour of the SO_2 and BrO distribution inside the plume area. As a very large number of volcanic events were investigated, it had been necessary to define a rather simple classification scheme. A detailed investigation of all identified cases depending on more complex parameters like e.g. the volcano's type or eruption circumstances (interaction with water/ice, extensive lava flows, volcanic explosivity index) might be an interesting task for future analyses.

*section 5.3: for this category, the BrO/SO2 ratio for individual measurements makes no sense. It would be better to investigate integrated BrO/SO2 ratio (see comment above).

The referee is right. We added BrO/SO_2 ratios from the total integrated molecule numbers (see reply above).

What is the cause of no-correlation? different heights? different timing in the (SO2-BrO) emissions?

The reason for no-correlation is so far unknown, but such cases might be caused by differences of the ambient atmospheric conditions and therefore meteorological parameters (such as temperature or relative humidity) at different heights. Furthermore, the composition of volcanic plumes during major eruptions (which are often caused by several individual explosions) may differ due to differences in the involved volcanic mechanisms, which could have a major impact on the ongoing halogen chemistry (please have also a look on our answer to the last comment of referee #2).

*section 5.4 (and Fig 15): not necessary.

We think that it is important to give an example for a volcanic eruption showing rather high SO_2 SCDs and no detectable enhanced BrO SCD in the plume area as the major part of all investigated plume showed comparable results. Therefore we prefer to keep this section as also recommended by the other referee.

-Section 6:

*Generally speaking, the Section 6 is too long (with a lot of repetitions of the findings already discussed in previous sections). The author might consider to merge Sects. 6 and 7.

We agree with the referee and merged Sections 6 and 7 into a "Discussion and Conclusions" section (new Sect. 6).

*It would be very useful to have an additional figure summarizing the BrO and SO2 observations. A world map showing all investigated eruptions with e.g. blue triangles at the locations of the volcanoes where only SO2 was measured and red triangles for the volcanoes where both SO2 and BrO have been detected. It would make a nice summary and link to the supplementary material.

We thank the referee for this good idea. A corresponding additional figure can now be found in the manuscript (Sect. 6) and is mentioned in the text:

*as another figure (or sub-figure), it would be good to summarize the range of BrO/SO2 ratios obtained in this study for the different volcanoes side-by-side with the reported values from the literature (GB, Aircraft) and discuss on that basis.

We added a new table to Sect. 6.2. of the manuscript, that summarises all worldwide reported volcanic BrO observations for different measurement geometries including the results from this study and shortly discussed the results. The results were sorted by the responsible volcano and the month of the corresponding measurements. In case of several measurements during a certain month, the maximum BrO/SO_2 is given.

Minor comments

-P2, l22: please add a reference on stratospheric ozone depletion; Barrie et al. And Simpson et al deals with ODEs in the troposphere only.

Two additional references on stratospheric ozone depletion were included:

1) Solomon, S.: Stratospheric ozone depletion: A review of concepts and history, Reviews of Geophysics, 37, 275, doi:10.1029/1999RG900008, 1999.

2) Rowland, F. S.: Stratospheric ozone depletion, Philosophical Transactions of the Royal Society B: Biological Sciences, 361, 769 –790, doi:10.1098/rstb.2005.1783, 2006.

-P3, l61: "This confirmed the suggestion that the reaction cycle is photolytically driven". The link with the previous paragraph is unclear. The fact that you measure only BrO during daytime is independent of the precise (gas-phase or heterogeneous) chemistry involved.

The fact that the GOME-2 measurements only take place during daytime is of course independent of the involved chemistry, but rather linked to the presence of scattered UV sunlight, which is not available during nighttime. However, the findings by Kern et al. (2008) were only mentioned to complete the paragraph on all previous studies on the volcanic BrO formation process. These measurements were important, as they confirmed that sunlight is crucially needed for a key step in the bromine explosion reaction cycle (the photolysis of Br_2). During the night most reactive bromine is therefore present as Br_2 and significant amounts of BrO are only regenerated after sunrise (von Glasow, 2010).

-P3, l62-65: please reformulate. Suggestions: “long-term development”! “long-term dataset” , “supposed”! “argued”, “volcano’s state” ! something else (too vague).

1) „long-term development“ was changed to „long-term development“
2) „supposed“ was changed to „argued“
3) „volcano’s state“ was changed to „… volcano’s state respectively a precursor/indicator for the onset of eruptive activity“

-P4, l74: “named”!”investigated”.

„named“ was changed to „investigated“

-P4, l81: “troposphere”!”atmosphere”

„troposphere“ was changed to „atmosphere“

-P8, l203:”(very unlikely)” !“(probably unlikely)”.

„very unlikely“ was changed to „probably unlikely“

-P12, l313:”the volcanic BrO signal is superimposed..”! “..is affected by the stratospheric BrO contribution” (+provide a refer). Please note that the longitudinal variation of stratospheric BrO is often larger than the latitudinal variation (! adapt the text). As for the polar tropospheric BrO, the 2-D correction is unable to correct for this signal! I wouldn’t mention this aspect.

The referee is in principle right that, especially for mid- and high-latitudes, strong longitudinal variations may appear. If such variations are very localised (and therefore small compared to the overall considered area for the calculation of the 2D polynomial), the 2D-correction is unable to correct entirely for this signal. However, if the variations are on a larger spatial scale, there is no reason why the 2D-correction should not work properly.

Like it was also recommended by the other referee, we added a short notice on the absolute range of the BrO VCD variations in dependence of the lat-/longitudinal position:

„...the volcanic BrO signal is affected by the stratospheric BrO distribution (Theys et al., 2009b), which systematically depends on latitude (the BrO VCDs typically increase from ~ $2x10^{13}$ molec/cm² at equatorial regions up to ~$7x10^{13}$ molec/cm² towards the poles, depending on season), but to a smaller degree also on longitude (small variations at the equatorial regions, but relatively strong variations of ±$2x10^{13}$ molec/cm² for the VCDs at mid- and high latitudes).“

Furthermore we added:

„It should be noted that the 2D correction only removes a smooth background signal, but cannot completely remove non-volcanic BrO with strong spatial gradients. However, such events typically only occur at high latitudes.“

-Sect. 4.5, P24, l480: please add a reference to Waythomas et al., JGR, 2010

Done

-Sect 7, p41, l831: “Here, the corresponding BrO/SO2 ratios have been estimated to were below”! “Here, the corresponding BrO/SO2 ratios are estimated to be below”.

Done

-References list: please rearrange. The list should be ordered alphabetically and then per year (for a given first author). If an author has published several papers in one year, please use letters (e.g., 2009a, 2009b,..).

Done

Additional corrections by the authors:

- We named the Redoubt volcano „Mt. Redoubt“ in the manuscript, which seems to be an often-made mistake. However, the volcano is correctly named „Redoubt“ or „Redoubt volcano“ and we therefore changed the name to „Redoubt“.

- Event #278 (the first observation of the Redoubt eruption in May/April 2009) was wrongly assigned to 11 March 2009. The date has been corrected to 23 March 2009.

- Due to an incorrect assumption of the BrO fit error in the discussion paper, the BrO/SO_2 ratios from the linear fit had to be recalculated. All events were once again analysed by using the individual (correct) SO_2 and BrO fit errors from the DOAS analysis. However, for the vast majority of all positively identified BrO events, the recalculated BrO/SO_2 remain essentially the same if the error of the linear fit is taken into account. Larger differences only showed up for some of the more complex events (Kasatochi, Sarychev), where the BrO/SO_2 ratio from the linear fit is probably not a good quantity as the distribution of the two species (at least partly) differ from each other (as mentioned above by the referee).