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## 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.

## Specific comments

- -Introduction: please explain why the BrO/SO2 ratio is an important parameter.
- -Page 5, I143: 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.

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

- -Section 3.5: the SCD threshold value of 1x1018 molec/cm<sup>2</sup> is arbitrary for the reader. Please justify.
- -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).
- \*Table 2 is not necessary.
- -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.
- -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.
- \*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).
- -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. 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.
- -Section 5:
- \* p-value?

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- \*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 r² but this is the case because there are only few points. This makes the definition of the categories (Table 3) questionable.
- \*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.
- \*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). What is the cause of no-correlation? different heights? different timing in the (SO2-BrO) emissions?
- \*section 5.4 (and Fig 15): not necessary.
- -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.
- \*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.
- \*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.

Minor comments

- -P2, I22: please add a reference on stratospheric ozone depletion; Barrie et al and Simpson et al deals with ODEs in the troposphere only.
- -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.
- -P3, l62-65: please reformulate. Suggestions: "long-term development"  $\rightarrow$  "long-term dataset", "supposed"  $\rightarrow$  "argued", "volcano's state"  $\rightarrow$  something else (too vague).
- -P4, I74: "named"→"investigated".
- -P4, l81: "troposphere"→"atmosphere"
- -P8, I203:"(very unlikely)" → "(probably unlikely)".
- -P12, I313:"the volcanic BrO signal is superimposed.." $\rightarrow$  "..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 ( $\rightarrow$  adapt the text). As for the polar tropospheric BrO, the 2-D correction is unable to correct for this signal $\rightarrow$  I wouldn't mention this aspect.
- -Sect. 4.5, P24, I480: please add a reference to Waythomas et al., JGR, 2010
- -Sect 7, p41, I831: "Here, the corresponding BrO/SO2 ratios have been estimated to were below"→ "Here, the corresponding BrO/SO2 ratios are estimated to be below".
- -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,..).

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 29325, 2012.

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