Interactive comment on “BrO/SO2 molar ratios from scanning DOAS measurements in the NOVAC network” by P.Lübcke et al.

The comments of Anonymous Referee #1 are printed in normal black font, our answers are printed in bold font. Text that was changed or added to the revised manuscript is printed in italic.

In this work the authors describe a procedure they implemented to automatically process UV spectrometer data obtained by automated NOVAC type I ground-based mini DOAS stations for BrO and SO2 retrievals. A remarkable 3-year-long record of observations collected by two NOVAC stations located at Nevado del Ruiz volcano, Colombia, is included as a demonstration dataset. The obtained timeseries covers a very interesting period where Ruiz experienced significant unrest and based on their observations of changes in the BrO/SO2 ratio before and after a small eruption in June 2012, the authors suggest that plume BrO/SO2 ratios may eventually be useful in tracking volcanic activity. While the authors choose not to discuss possible causes for changes in BrO/SO2 at Ruiz, their stated intent is to apply similar techniques to other NOVAC datasets in the future and to introduce automated BrO retrievals as an operational volcano monitoring parameter. There is a lot to like in this manuscript: the authors (and field personnel) are congratulated on collecting an outstanding dataset that is of high value and clearly should be published. Also, the manuscript is in general well-written and clearly presented, despite some language and organizational issues and a few minor editorial mistakes that deserve attention.

We would like to thank the reviewer for his helpful review that greatly helped improving the manuscript. We answered the comments of the reviewer below.

My main concerns with the manuscript in its present state are twofold: i) the data processing procedure and the discussion of possible pitfalls in automating the procedure are not as robust as they could be (discussed below), and ii) the example dataset is – at present – the most novel aspect of this work and its greatest strength but alas, it is not discussed in detail since it is simply used as a tantalizing example of an extremely beautiful DOAS dataset. If the data processing procedure were stronger (the purpose of the paper) I wouldn’t feel so let down by the lack of discussion of the Ruiz dataset. Since developing a processing procedure is the main goal of the study, I wonder if a more specialized journal might be better suited for publication (e.g. in the spirit of Bobrowski et al., 2010) that would allow for the approach described here to be more fully developed. If the emphasis is placed more on describing and interpreting the dataset from Ruiz, I believe the choice in publication is appropriate but then the authors must substantially re-think the purpose of the present work. As is stands, I feel the manuscript shows considerable promise and touches on two important subjects – each important and worthy of attention – but that neither is considered as fully as they deserve. My main recommendation is to re-work the data processing procedure to be more fully developed (see comments below). Also, the authors might consider submitting the manuscript to another, more specialized journal. My final recommendation would be to start writing up a detailed analysis of the Ruiz dataset since I’ll be very interested to read it! Please see below for further comments.

The reviewer expresses some doubts about the robustness of the automation of our novel evaluation procedure and goes on to suggest that sending the paper to another journal more specialised on technology should be considered or - alternatively the data of Nevado del Ruiz should be more comprehensively discussed. We do not think that the algorithm is lacking robustness at all. However, we appreciated very much the suggestions and comments by the reviewer which helped us to improve our manuscript and to better illustrate the validity of the algorithm. We changed the manuscript in the points suggested by the reviewer and are convinced that this additional discussion helps to better show the robustness of the method. We reflected the question regarding our choice of journal and still think this is the right place to publish this kind of results. In particular because the algorithm is developed for these robust, and low budget instruments which are currently mainly applied for the remote sensing of "high" volcanic gas emissions. Although in parts rather technical, we are convinced that the manuscript fits in the scope of Solid Earth because it addresses ongoing research and monitoring efforts of the volcanological community. The reported observations of variations in BrO/SO_2 ratios are of great interest and make this data set accessible for further research. The presented method to obtain BrO/SO_2 ratios from the NOVAC spectra by co-adding of spectra is a new approach. A proper explanation of the method is necessary before an interpretation in a volcanological context is possible. We now added a comparison of the observed BrO/SO_2 ratio with seismic measurements that were taken from the homepage of the Servicio Geológico Colombiano that indicate that variations of the BrO/SO_2 ratio coincide with changes of seismic activity (Section 3.3). However, at this point more interpretation would be speculative and we therefore waive this idea.

Specific comments:

My main concerns regarding the algorithm developed here is that a few important issues are not discussed and that the procedure should, in my opinion, be more generalized and contain better assessment elements to assure quality control. For example: is any consideration given to meteorologic clouds and how they may affect retrievals or be identified in the data? My concern stems from the fact that the summit of Ruiz (and ~6000 m, the likely transport altitude of the plume) is oftentimes very cloudy. What impact would clouds have on the retrievals and how are cloudy data identified and dealt with? Is it possible to compare cloudy vs. non-cloudy data to see if there are impacts? I am also wondering if any consideration is given to ash in the plume and its effects on the retrievals? Ash must be an issue since it covered the solar panels and temporarily knocked the stations offline.

We fully agree with the referee that meteorological clouds and ash are important factors to be considered during the remote sensing of volcanic gas emissions - in principle. However, in contrast to SO_2 emission rate measurements (where clouds can severely affect the results), looking at the ratio of two trace gases is often much more robust. Broad-band effects (like those produced by the presence of clouds or ash) have long been known to influence the retrieved SO_2 column density in remote-sensing measurements (e.g., Milan, 1980, Mori et al., 2006, Kern et al., 2010, Kern et al, 2012). However these effects usually do not directly influence the DOAS retrieval, but rather the light-path, the path that photons have travelled before being detected by the

instrument. Increased aerosol abundance (e.g. meteorological clouds) is known to increase the light-path and might thus lead to an increased column density (e.g. Pfeilsticker et al. 1998). On the other hand a meteorological cloud or a strongly condensed plume can lead to the effect that most radiation that is scattered into the instrument's field of view has not penetrated the entire volcanic plume. This would lead to a reduced trace gas column density. The advantage when looking at the ratio of two gases (especially when the DOAS retrieval only contains a relatively narrow wavelength interval as in our case) is that many of the radiative transfer effects appear for both gases and thus mostly cancel out for the ratio.

We answer the latter question first: how are cloudy data identified and dealt with? Is it possible to compare cloudy vs. non-cloudy data to see if there are impacts?
Recent publications concerning the detection of clouds in MAX-DOAS observations suggest that a colour-index (radiation intensity at two different wavelengths), the absorption of the oxygen dimer (O_4) or the Ring-effect can be used to identify clouds (Wagner et al., 2013). One particular problem, when applying these findings to our data-set is, that volcanic plumes may contain aerosol (water droplets or ash), which may be difficult to distinguish from meteorological clouds as described above.

However, this is not a weakness but strength of looking at the trace-gas ratios, which will become apparent when answering "What impact would clouds have on the retrievals…?"
The evaluation ranges of SO_2 and BrO are relatively close in wavelength and thus both evaluations are influenced by clouds (or aerosols in the volcanic plume) in a very similar way. A meteorological cloud between the instrument and the plume would lead to a considerable reduction of the detected column densities. But the observed reduction is very similar for both gases. To assess this issue in a more quantitative way, we investigated two approaches:

1. **We evaluated SO_2 in a second wavelength range, between 326.5 - 335.3 nm (in addition to the standard range of 314.6 to 326.8 nm), this wavelength range was, e.g., chosen by Hörmann et al., 2013 as an alternative fit range for SO_2. As this fit range is less influenced by strong SO_2 absorption bands and radiative transfer effects (e.g., the light dilution effect) and even closer to the BrO retrieval range it can serve as an excellent proxy for the quality of the results. It should be noted though, that the SO_2 retrieval error in the alternative fit range is roughly a factor of 10 higher than in the standard range. Figure 1 shows the correlation of the SO_2 column densities retrieved in the two fit ranges. We observed ~30 - 35% higher SO_2 column densities in the alternative wavelength range and a linear correlation between the SO_2 column densities retrieved in the two wavelength ranges.**

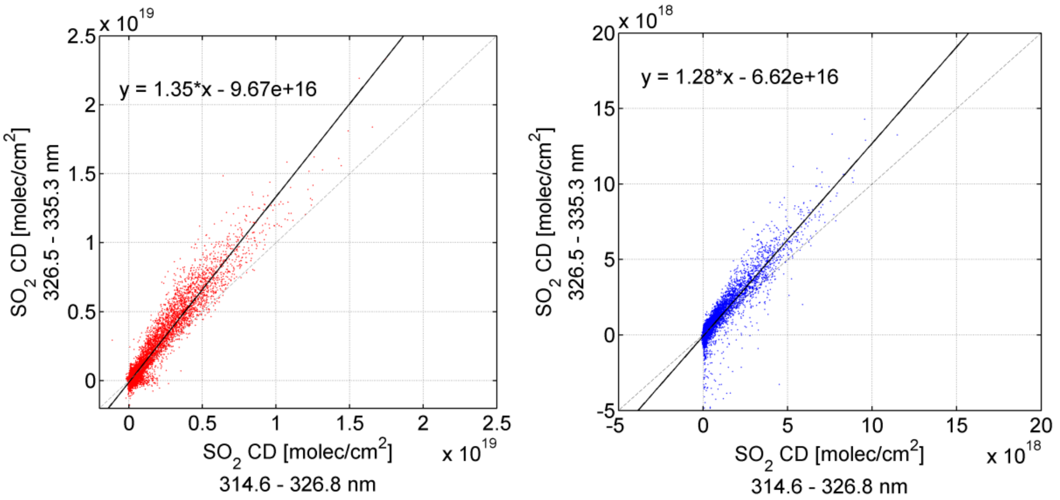

Figure 1 SO_2 **column densities retrieved betwen 314.6 - 326.8 nm (x-axis) plotted vs. the SO2 column densities retrieved between 326.5 - 335.3 nm. Left side: Instrument D2J2200, right side: D2J2201.**

We also re-evaluated BrO in the wavelength range between 327-347 nm as suggested by the referee. The retrieved BrO column densities are largely the same (see Figure 2). However, the BrO retrieval error in the fit range between 327-347 nm was approximately 30% higher, than the retrieval error in our standard fit range.

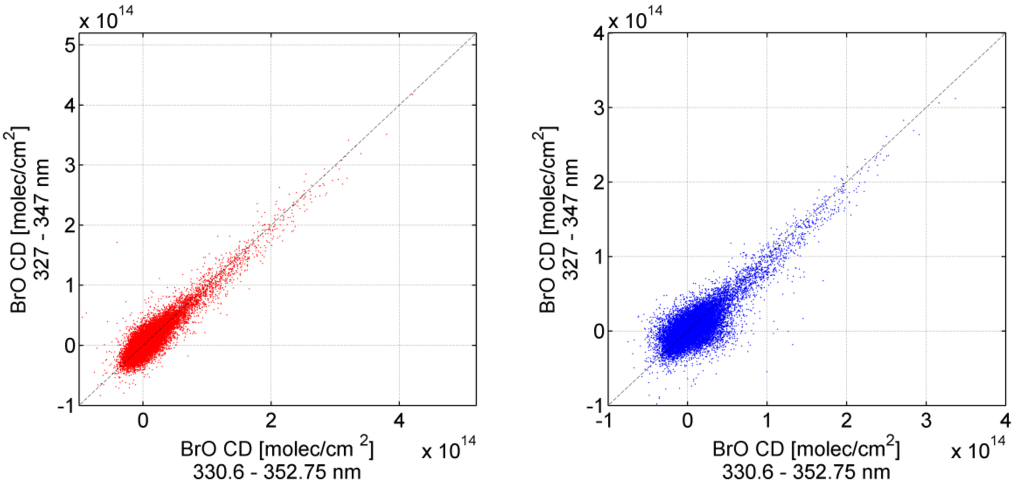

Figure 2 BrO column densities retrieved betwen 330.5 - 352.8 nm (x-axis) plotted vs. the BrO column densities retrieved between 327 - 347 nm (y-axis). Left side: Instrument D2J2200, right side: D2J2201. The BrO retrieval error in the fit range between 327-347 nm is approximately 30% higher, than the retrieval error in our standard fit range. The grey-dashed line shows the identity line.

To assess the influence of the different retrieval ranges on the BrO/SO_2 **ratios we compared the results from the different retrieval ranges. For a clearer presentation we compared the running mean values of the BrO/SO2 ratio for the four possible combinations of the retrieval ranges:**

1. ***Set 1*: The standard wavelength ranges,** SO_2 **was evaluated between 314.6 – 326.8 nm, BrO between 330.5 – 352.75 nm.**
2. ***Set 2*:** SO_2**: 314.6 – 326.8 nm, BrO: 327 – 347 nm**
3. ***Set 3*:** SO_2**: 326.8 – 353.5 nm, BrO: 330.5 – 352.75 nm**
4. ***Set 4*:** SO_2**: 326.8 – 353.5 nm, BrO: 327 – 347 nm**

The results (see Figure 3) were added to the new Figure 10 that combines Figure 9 and 10 with the results from the different evaluation ranges. It can be seen that evaluating SO_2 in the alternative range (which is closer to the BrO evaluation range) leads to larger SO_2 column densities and thus slightly lower BrO/SO_2 ratios. However, the variations of the BrO/SO_2 ratio are similar.

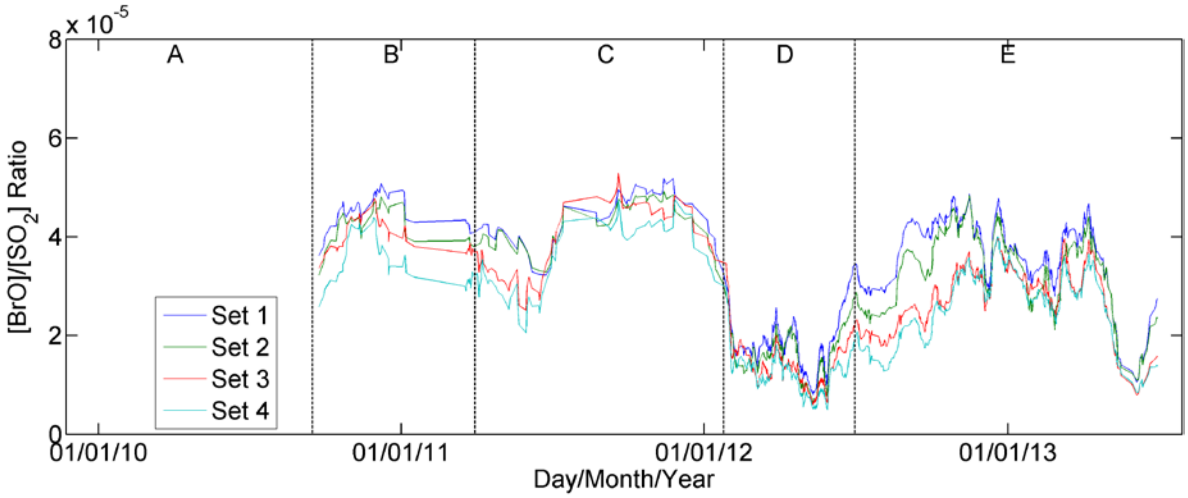

Figure 3 BrO/SO_2 time-series from Nevado del Ruiz. The four different sets use different wavelength ranges for the DOAS retrieval of BrO and SO_2 (see text for details).

2. **We performed radiative transfer simulations using the 3d radiative transfer model McArtim (Deutschmann, 2011). The simulations were set-up to mimic the geometry described by the referee. A volcanic plume with a circular cross-section shape with diameter 1 km in the x-z plane was simulated. The plume was simulated at a height between 5.5 and 6.5 km and was assumed to be infinitely extended in the y-direction (see Figure 2). Typical atmospheric profiles of the trace gases O_3, O_4 and NO_2 and a Rayleigh atmosphere were used to account for non-volcanic absorbers. Trace-gas concentrations of SO_2 and BrO were set to $1x10^{13}$ molecules/cm³ and $1x10^{9}$ molecules/cm³, respectively, inside the volcanic plume. This leads to column densities of $1x10^{18}$ molecules/cm² for SO_2 and $1x10^{14}$ molecules/cm² for BrO, assuming the light-path is a straight line through the plume centre. The resulting BrO/SO_2 ratio is $1.0x10^{-4}$. The instrument was placed below the plume at a height of 4.5 km, different lateral distances were simulated. The viewing direction of the instrument was always directed towards the centre of the volcanic plume.**

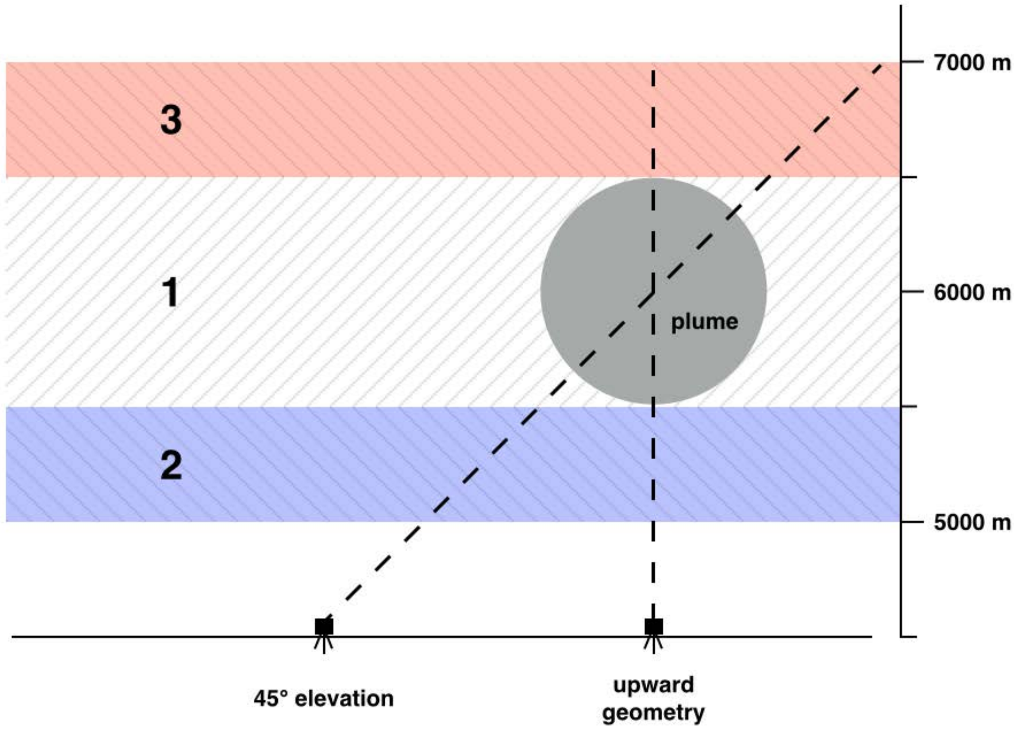

Figure 4 Measurement geometries and the three different cloud layers that were simulated for the radiative transfer simulations.

We first investigated the influence of an increasing lateral distance (between 0 km and 6 km from the plume) on the obtained BrO and SO_2 column densities. The column densities for SO_2 and BrO were simulated in the approximate middle of the DOAS fit window (320 nm for SO_2, 340 nm for BrO). Both, the retrieved SO_2 and BrO column densities clearly decline with increasing lateral distance. However, as the left side of Figure 5 shows, the BrO/SO_2 ratio only varies by less than 7%, even for a lateral distance of 6 km.

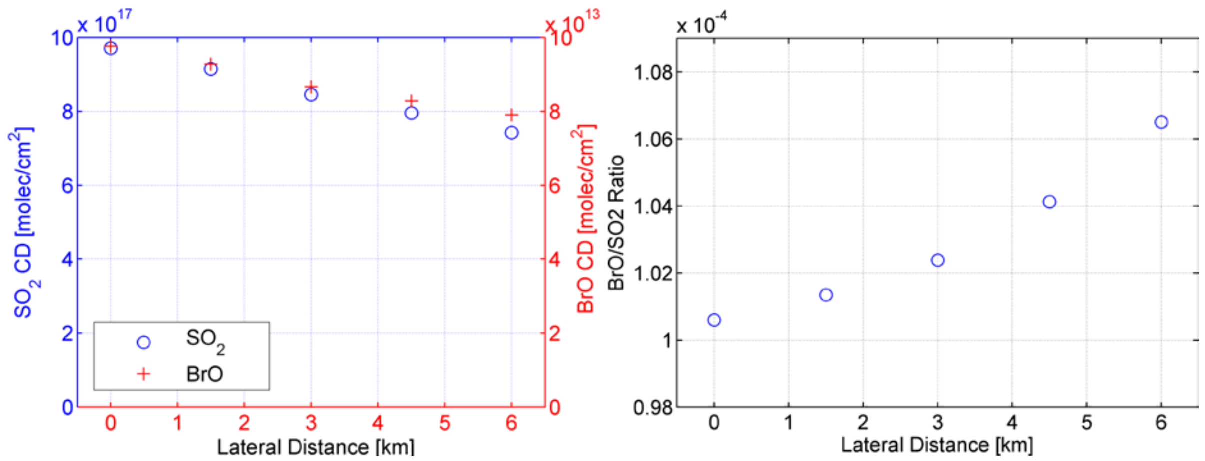

Figure 5 BrO and SO_2 CDs for different lateral distances of the instrument. b) shows that the SO2/BrO ratio is only slightly influenced by an increasing distance.

In order to investigate the influence of clouds, we simulated clouds in different heights. Three different cloud layers with heights between 5.5 – 6.5 km (the height of the volcanic plume), 5 – 5.5 km (a cloud layer between instrument and volcanic plume) and 6.5 - 7 km (a cloud layer above the volcanic plume) were simulated. An aerosol with an Ångström exponent of 1, a SSA of 1 and an asymmetry parameter g=0.85 was used in simulating the cloud (see, e.g.,

Wagner et al., 2013). Two different aerosol extinction coefficients were simulated to assess variations of the cloud optical depth. The aerosol extinction coefficient was chosen to obtain an AOD of 4 or 12 at 340 nm (for an upward looking instrument) and of 4.25 or 12.75, respectively, at 320 nm. Additionally as a fourth scenario no meteorological cloud layers were assumed and it was assumed to only have an enhanced aerosol content inside the volcanic plume. For these four simulations only two geometries, an instrument below the volcanic plume with an upward looking telescope, and an instrument at a lateral distance of 1.5 km with an elevation angle of 45° were simulated. The results are summarized in Table 1 and 2. Although for the 45° elevation angle geometry in cloud scenario 2, only ~30% of the SO_2 and BrO column densities are retrieved, the BrO/SO_2 ratio varies by only 2.5%.

		SO_2 [molec/cm^2] $\times 10^{18}$	BrO [molec/cm^2] $\times 10^{14}$	BrO/SO_2 Ratio	Deviation [%]
0	90°	0.97	0.98	1.007×10^{-4}	0.7
	45°	0.92	0.93	1.014×10^{-4}	1.4
I	90°	1.09	1.09	9.998×10^{-5}	−0.0
	45°	1.04	1.05	1.013×10^{-4}	1.3
II	90°	0.95	0.97	1.029×10^{-4}	2.9
	45°	0.28	0.27	9.755×10^{-5}	2.5
III	90°	0.96	0.96	1.000×10^{-4}	0.0
	45°	1.01	1.02	1.008×10^{-4}	0.8
IV	90°	1.08	1.08	9.992×10^{-5}	−0.1
	45°	1.07	1.09	1.013×10^{-4}	1.3

Table 1 SO_2, BrO CDs and the BrO/SO_2 ratios for the different cloud scenarios and a cloud optical density of 4 at 340 nm. (0) is a cloud free atmosphere, I a cloud layer between 5.5 and 6.5 km, II a cloud layer between 5 and 5.5 km, III a cloud layer between 6.5 and 7 km, and IV an enhanced aerosol abundance coinciding with the volcanic plume.

		SO_2 [molec/cm^2] $\times 10^{18}$	BrO [molec/cm^2] $\times 10^{14}$	BrO/SO_2 Ratio	Deviation [%]
0	90°	0.97	0.98	1.007×10^{-4}	0.7
	45°	0.92	0.93	1.014×10^{-4}	1.4
I	90°	1.37	1.36	9.997×10^{-5}	−0.0
	45°	1.17	1.20	1.023×10^{-4}	2.3
II	90°	0.81	0.83	1.034×10^{-4}	3.4
	45°	0.22	0.22	9.964×10^{-5}	−0.4
III	90°	0.99	0.99	9.989×10^{-5}	−0.1
	45°	1.03	1.04	1.006×10^{-4}	0.6
IV	90°	1.26	1.26	9.991×10^{-5}	−0.1
	45°	1.20	1.22	1.024×10^{-4}	2.4

Table 2 SO_2, BrO CDs and the BrO/SO_2 ratios for the different cloud scenarios and an cloud optical density of 4 at 340 nm. (0) is a cloud free atmosphere, I a cloud layer between 5.5 and 6.5 km, II a cloud layer between 5 and 5.5 km, III a cloud layer between 6.5 and 7 km, and IV a cloud layer coinciding with the volcanic plume.

Thus we can conclude that the influence on the retrieved BrO/SO_2 ratios is minimal. We added the above discussion on radiative transfer calculations and the evaluations in different wavelength ranges to the manuscript and are confident that we can convince the reviewer of the robustness of our evaluation procedure regarding the presence or absence of clouds.

We added on p1850, Line 22:
"To verify the results both trace gases were also evaluated in additional wavelength ranges. SO_2 was additionally evaluated between 326.5-353.3 nm a wavelength range already used by Hörmann et al., 2013 as an additional wavelength range for the SO_2 retrieval. BrO was additionally evaluated between 327 – 347 nm. This wavelength range was used by Kelly et al., 2013 for the BrO retrieval."

A subsection on Radiative Transfer Model calculations was added to the method sections (Section 2.4). The results for both, different evaluation ranges and for the RTM calculations were added as a separate subsection in the results section (Section 3.2).

With regard to quality assurance, I appreciate the discussion on the effects of temperature. However, might some other additional methods prove advantageous for assuring measurement quality? For example, the present procedure fits BrO in the region 330.6 – 352.75 nm (Section 3, pg 1850, line 15). Could additional BrO fits in other regions (e.g. 327-347, 327-357, etc.) be used as in internal check on the fitting procedure and to assure nothing is being missed? The same goes for the SO2 retrieval; if an additional window is available (e.g. that described in Bobrowski et al., 2010) could that be used to determine if the data are impacted by ash, the distance to the plume, etc.? What tools are included to assure the procedure is working?

SO_2 and BrO were now both evaluated in additional wavelength ranges (see discussion about the different retrieval ranges above). An SO_2 retrieval in the wavelength range described by Bobrowski et al., 2010 is not possible as the filters that are used to block straylight (Hoya U330) also block radiation above 360 nm (Galle et al., 2010). We also added a discussion on temperature effects on the DOAS retrieval (see Reviewer #2 comments).

All of these thoughts are motivated by my concerns that the procedure outlined in the manuscript seem somewhat "tuned" to the Ruiz case and do not seem sufficiently generalized for application to other sites. For example, the threshold values chosen (e.g. Section 4, pg 1853, line 6) seem somewhat arbitrary and dictated by the present data set.

The presented data is in no way especially tuned for Nevado del Ruiz. In fact, we evaluated 2 years of data at Galeras, Colombia. While the BrO retrieval error is comparable, in general emissions were found to be too low to retrieve (non-zero) BrO/SO_2 ratios. However, with the approach outlined in our paper other observatories have a guideline at hand, on how to evaluate BrO for their instruments.
The threshold value is obviously (apart from the detection limit of BrO) dictated by the BrO/SO_2 ratios and the total gas amount. However changing the SO_2 threshold to lower or higher values does not significantly change the observed

signal. Additionally, to discuss the influence the choice of threshold levels can have on the BrO/SO_2 ratio, we included both time series (with an SO_2 CD or a BrO error-based threshold) in the manuscript.

The present work would be much stronger if it outlined a more robust, generalized approach to processing long DOAS timeseries. Such an approach should include quality assurance steps and methods to try to identify data compromised by technical or environmental conditions. Along the same lines, I am surprised that a toolkit of sorts or some kind of collection of scripts is not included as an online supplement to the work. Such a toolkit would be of considerable value and would provide a basis for further code development, especially since the authors plan to implement an automated retrieval routine at observatories that host NOVAC instruments (Section 5, pg 1855, lines 14-15). More fully developing the procedure to assure measurement quality and providing a toolkit would substantially increase the value of the present work.

We apologize for the misunderstanding. We do not intend to implement the current version of the algorithm as an operational product at the observatories. This work should be rather seen as a starting point (a proof of the possibility to evaluate BrO with these low cost, robust instruments which are tuned to obtain SO_2 emission rates with a high time resolution and therefore have a lower signal to noise ratio) for the continuous observation of BrO/SO_2 ratios. We will consider publishing the scripts at a later time. Much work on the documentation of the scripts is needed before publishing them online. After all there is a large difference, between a working script and a script that yields reliable results for inexperienced users in all situations.

Minor comments

Abstract, pg 1846, line 2: I disagree; the ratio of BrO to SO2 is not like other commonly measured halogen sulfur ratios (e.g. HCl/SO2) since BrO is not a primary product emitted from volcanoes. I realize that the operational use of BrO/SO2 for monitoring is somewhat out the scope of the present work, but I would advise extreme caution in interpreting this parameter in terms of volcanic activity. Of course, this is the question we all hope to address…

While the referee correctly points out that BrO is not primarily emitted from volcanoes, other halogen ratios can be influenced by the environment as well. For example HCl/SO_2 ratios are also known to be influenced by local conditions (meteoric water, ect). Since we are aware that BrO is not a primary product we were very careful with our interpretation. However the amount of BrO that is detected will probably not be independent from the emitted amount of HBr. Announcing it only as a "possible" precursor and observing for the second time (second volcano besides Etna, Italy, Bobrowski and Giuffrida, 2012) a similar coinciding pattern with volcanic activity is a careful enough formulation in our opinion.

A flow chart illustrating the data processing procedure would be helpful.

We added a flow chart (see Figure 6) to illustrate the data processing and on p.1849, line 17 "*A flow chart of the data processing procedure is shown in Fig. 3."*

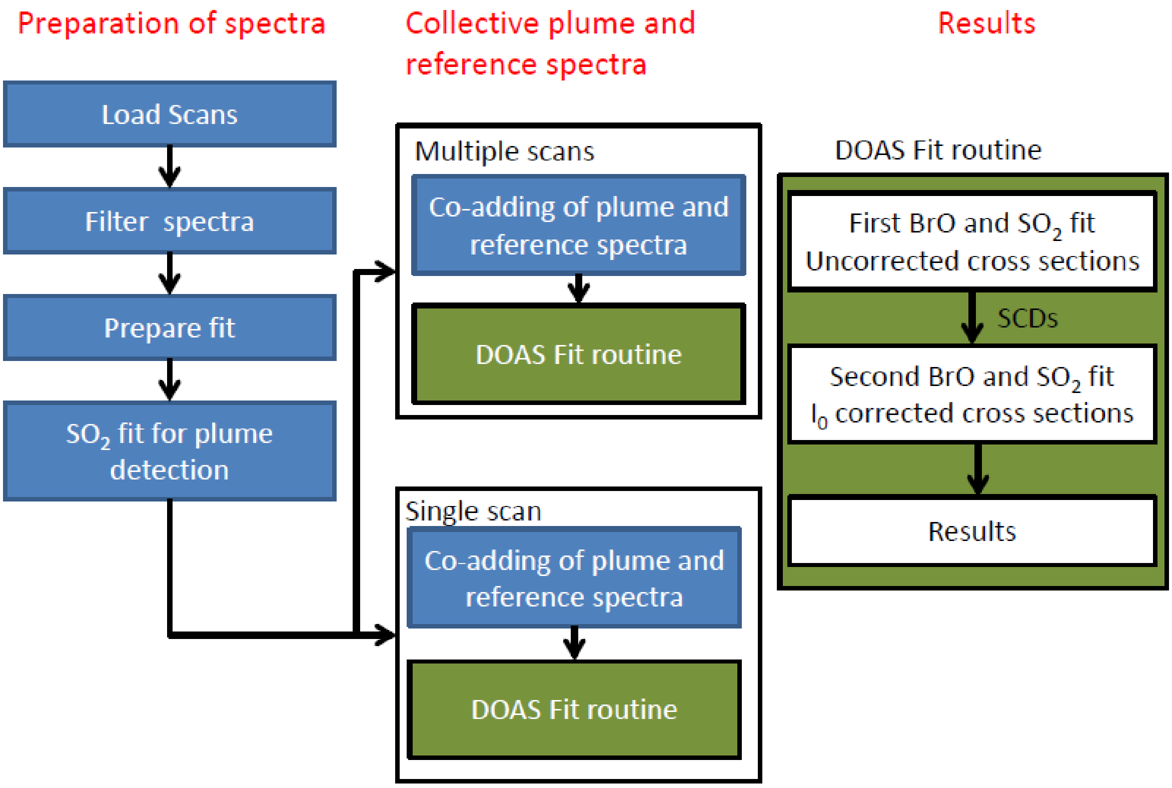

Figure 6 Flow chart of the data evaluation to automatically retrieve BrO/SO_2 ratios.

Section 2: please specify the wavelength range of the instruments.

We added: *"(with a wavelength range of 280-425 nm and an optical resolution of ~0.6 nm)*" **on p1849, line 2.**

Section 3, lines 29-32: please specify how much shift and squeeze are allowed.

We added the information on shift and squeeze on p.1851, Line 4: "*A shift of ±0.2 nm and a squeeze between 0.98 – 1.02 were allowed.***"**

Pg 1847, line 20: I would suggest replacing "reach" with "approach." Other geophysical methods operate at second to sub-second sampling frequencies; only SO2 camera can truly be said "reach" these levels.
We agree with the reviewer and replaced "*reach***" with "***approach***".**

Pg 1851, line 11: How long does it take the system to make 4 consecutive scans? I.e. what is the time resolution?

The time resolution depends on the time of day, in the morning and evening hours one scan (from horizon to horizon) might take up to 15 minutes, while during the middle of the day one scan is performed within 4-5 minutes. We

added the following sentence on p1849, line 16: "*Depending on the time of day four consecutive scans are typically recorded within 15 – 60 minutes.***"**

Pg 1852, line 3: "is thought to not be influenced by these temperature issues" – is there a reference or other means to substantiate this?

Yes, we showed in Figure 5 (Figure 6 in the new manuscript) that our DOAS fit error is decreasing by adding up spectra according to the photon statistics, which indicates that the influence of temperature changes is of minor importance here.

Pg 1852, line 23: "ratioing" is misspelled and should probably be replaced by "Taking the ratio of two values: : :"

We replaced "*ratioing***" with "***Taking the ratio of two values…***".**

I would suggest integrating much of the "pitfalls" discussion in Section 5 into Section 2 or 3, especially the issue concerning BrO line shape (Section 5, pg 1856, lines 24-30). In particular, since this issue seems fundamental could it be addressed in the present work? Also, introducing new data or ideas not previously mentioned in the article should be avoided in the conclusions.

We added a subsection on effects from the temperature to the Methods section. DOAS retrievals on synthetic spectra indicated that the error of the BrO/SO_2 ratio caused by temperature variations in the NOVAC instruments is below 15%. We replaced p.1856, lines 8 – 29 with:
"*Another possible approach to further improve the accuracy of our algorithm is taking temperature effects of the instruments, which were discussed in the Methods section, into account. This could further help to improve the quality of data evaluation.***"**

Figure 8: The daily average SO2 emission rate values appear to be very different than daily maximum values that have been available elsewhere (presumably from the same dataset, e.g. Fig. 4 in Herrick). Can you comment on the discrepancy? For example, the highest SO2 emission rate shown in Figure 8 is _90 kg/s or _8000 tonnes/day. Herrick Fig 4 shows maxima up to 33,000 tonnes per day (around 400 kg/s). Is there really that much daily variation to drag down the emission rates? Also, is there a way to assess the model wind speeds and how well they are performing? Can you provide the url from where were the data accessed? Perhaps it's just a scaling issue, but it appears as though the flux data are cut off earlier than the column densities displayed in Fig 6.

The average daily SO_2 emission rates are indeed different from the daily maximum SO_2 emission rate. For example, if an explosion happens large SO_2 emission rates can be observed, however, before the explosion emission rates might be suppressed nearly completely. The average would thus be considerably lower than the maximum value for a short time. When looking at single scans we can observe comparable SO_2 emission rates as shown in Herrick, 2012. Wind data was taken from the ECMWF Reanalysis (http://www.ecmwf.int/). More detailed information on the SO_2 emission rates will be published in Arellano, 2014 (manuscript in preparation).

References:

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