

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

The comments of the Anonymous Referee #2 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 font.

The interpretation of the temporal variations in the BrO/SO₂ molar ratio in terms of magma dynamics is not straightforward as the abundance of BrO results from a complex combination of atmospheric and magmatic processes. A deconvolution of these various processes is required to improve our understanding of the mechanisms triggering changes in the temporal evolution of the eruptive dynamics or volcano activity. However, since the discovery of its existence of BrO in volcanic plumes, BrO has been traditionally measured during short-term field experiments. Very few time-series of the BrO/SO₂ molar ratio in the plume are available on a period of time sufficient to apprehend in depth the meaning of BrO variations in terms of magma dynamics. For now, no algorithms presenting an automatic procedure for the BrO trace gas species retrieval have been neither proposed yet to robustly process long data time-series.

In light of the above, the paper of Lübcke et al. is very welcome as it forges ahead to break new ground in the perspective of the potential future development of new tools for more efficient volcano monitoring. This paper presents a nice, long time-series, covering months/years of variations in the BrO/SO₂ molar ratio in the plume of the active Nevado del Ruiz volcano. As mentioned earlier, very few such long data sets are shown in the existing literature, which demonstrates the irrefutable interest of this study. This work is essentially a technical paper, as except the observation/result that a decrease of the BrO/SO₂ ratio preceded of several weeks the eruption of Nevado del Ruiz in June 2012, the discussion of the mechanisms (which could be of volcanological, atmospheric or meteorological type) that could explain such temporal variations is very limited. This is unfortunate as such a dataset is really intriguing. Nevertheless, the authors do not pretend to have the goal of exploring such processes. However, the publication of a technical study in ‘Solid Earth’ is questionable and could be more naturally expected in a journal publishing technical developments such as AMT (Atmospheric Measurement Techniques) for example. This said, whether it is worth publishing this study in ‘Solid Earth’ is obviously in the hands of the Editor.

Lübcke et al. explore a technical aspect, which consists in proposing an algorithm which could be further implemented in an automatic mode to process long time series of the BrO abundance.

The paper is well-written with meaningful and clear figures.

We thank the reviewer for his opinion and his helpful suggestions. We agree with reviewer that the time-series of the BrO/SO₂ could be of large interest for the volcanological community. We answer the specific comments of the referee below.

BrO is a trace gas species, whose abundance is often close to the level of detection. The determination of the BrO abundance requires consequently a refined but also robust and cautious spectroscopical retrieval, as retrieved abundance may be

significantly impacted by the retrieval process in itself. My main concern is that different stages in the retrieval algorithm which is presented (listed in the following) could produce artefactual variations in the BrO/SO₂ ratio related to the retrieval algorithm. As detailed below, more evidences and quality control steps in the algorithm procedure are required to demonstrate the robustness of the proposed algorithm. This is a mandatory step before allowing for the interpretation, with confidence, of BrO/SO₂ ratio variations in terms of magma dynamics or atmospheric/meteorological processes.

If the authors can provide more robust evidences of the algorithm robustness/reliability, the paper would be greatly improved. Other suggested edits are mentioned in the following. For more clarity, some suggestions of re-organisation of some parts of the manuscript are also proposed.

We added a section in which we compare the BrO/SO₂ ratio with seismic activity. We also added several discussion points that help to prove the robustness of the data evaluation.

We will answer the questions below:

Major comments:

- There is no thermal stabilisation of the spectrometers used in this study. This fact is presented as a considerable advantage (line 2 of p1849) as thermal stabilisation is energy consuming and its development may be more difficult for the monitoring of remote volcanoes. However, this presentation could appear somehow fallacious as the authors do not mention simultaneously on the same lines the impact of the absence of thermal stabilisation on the gas spectroscopical retrieval and the associated uncertainty on the results. As detailed below, this uncertainty is not quantified. The main concern is that non negligible artefactual variations of the BrO/SO₂ molar ratio could result from temperature variations. At least, in the line of the information provided in the paper, it is impossible to ascertain that this effect is negligible.

We agree with the reviewer that we should mention that the lack of thermal stabilization can be a disadvantage in terms of spectral quality.

Two different problems can occur: 1) the wavelength-pixel-mapping of the instrument changes and 2) the shape of the instrument line function (ILF) varies. The first point is not a problem during the evaluation of spectra from a single scan since the Fraunhofer Reference Spectrum is calibrated with help of a high-resolution solar atlas and a shift of the trace gas cross sections (linked all together) is allowed in the fitting procedure. Problems might occur during co-adding of spectra from several scans. The intensities of the different spectra are co-added for each channel. If the wavelength-pixel-mapping changes between different scans this could lead to problems. However, when evaluating spectra created from co-adding of scans we observe a BrO fit error that is only 50% of the fit error for spectrum created from a single scan. We thus conclude, that the fit error reduces according to the ideal case of a photon statistics limited instrument, and that temperature effects seem to be a minor (neglectable) problem here.

Variations of the ILF could have an impact on our data evaluation. However, the largest problems occur if FRS and measurement spectrum are not recorded at

the same temperature. This effect does not occur in our retrieval, since FRS and measurement spectra are recorded in close time proximity (5-15 minutes) and thus at the same temperature.

A problem that actually might influence our retrieval is that the ILF, which was used for the convolution of the absorption cross sections, differs from the ILF of the instrument during the measurement. To assess this issue, we performed a DOAS retrieval on synthetic spectra. A Gaussian profile with an FWHM of 0.65 nm was used for the convolution of the absorption cross sections, for the synthetic spectra a Gaussian profile with varying FWHM was assumed. The FWHM for the synthetic spectra was varied in steps of 0.025 nm up to between 0.525 nm and 0.775 nm (which covers a little more than the range of ILF of the instruments used in the NOVAC network as given in Pinardi et al., 2007). DOAS retrievals of the synthetic spectra showed that variation of the ILF lead to variations of the BrO/SO₂ ratio that were below 15%.

We added a discussion of the temperature effects to the Methods section (Section 2.3).

A more detailed explanation for the studies on synthetic spectra was added as Appendix 1:

“We simulated synthetic spectra as outlined in Vogel et al., 2011. Measurement and FRS spectra were simulated. The BrO content of the measurement spectrum was 1.5×10^{14} molecules/cm² and the SO₂ content was 1×10^{18} molecules/cm². The synthetic spectra used the Chance and Kurucz, 2011 Solar Atlas. Beer-Lambert’s Law was applied on the wavelength grid of the solar atlas using stratospheric absorbers and in the case of the measurement spectrum additionally BrO and SO₂. Then the high resolution spectra were convolved to a lower resolution with a Gaussian profile with varying FWHM. The FWHM was varied between 0.525 - 0.775 nm in steps of 0.025 nm. For each FWHM a set of measurement and FRS spectrum (convolved with the same Gaussian profile) were evaluated using the DOAS retrieval as outlined in Section 2. The absorption cross sections were convolved with a constant Gaussian profile with a FWHM of 0.65 nm. This approach mimics the situation in NOVAC. While a constant FWHM is used for the convolution of the absorption cross sections, the ILF of both spectra varies with temperature. Figure 7 shows the deviation of the BrO/SO₂ ratio from the BrO/SO₂ ratio retrieved with both spectra using the same ILF. The results show that the error in the BrO/SO₂ ratio is below 15% for variations of the ILF of up to 0.125 nm.”

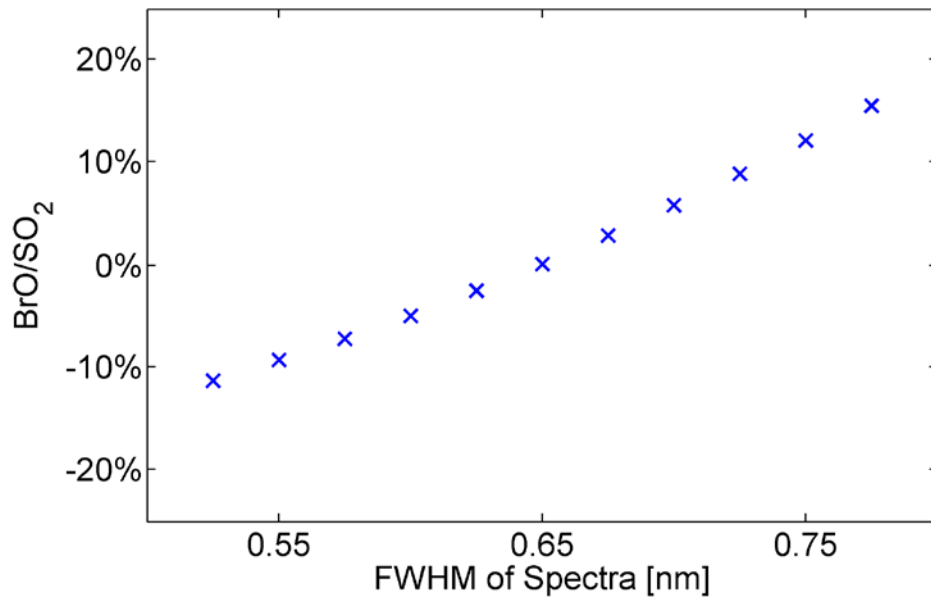


Figure 1 Variation of the BrO/SO₂ ratio with varying FWHM of the synthetic spectra. The FWHM used for the convolution of the absorption cross sections had an FWHM of 0.65 nm. (Figure 7 in the updated manuscript).

The effect of temperature changes is discussed shortly later in the manuscript, associated with Figure 5. This representation in Fig. 5 (shift or squeeze versus temperature, using half a year of data) does not provide any information on the time variations of temperature which are observed and the associated uncertainty on retrieved gas abundances. Lines in the 'Conclusion' section suggest that the authors themselves are aware of this matter. However, the discussion which is provided appears largely insufficient as this temperature effect may be a major issue to assess the robustness of retrieved BrO/SO₂ molar ratio time series. Given the goal of the paper and the uncertainty on the provided results, this question is central and cannot be relegated to new lines of investigation for the future.

We have now added that variations due to temperature induced changes of the ILF are below 15%. Furthermore, we investigated only daily average values of the BrO/SO₂ ratio. Considering the location of Nevado del Ruiz, close to the equator, the temperature variations over the course of a year are minor compared to the diurnal cycle of the temperature. We therefore conclude that temperature variations might cause some of the scatter that can be observed in the BrO/SO₂ time series, but does not lead to significant a significant bias over larger time scales as observed here.

To give confidence in the results, is there a record of the instrument temperature available, or at least of the atmospheric temperature? If so, a first step could be to check that variations in temperature are not correlated with variations in the BrO/SO₂ molar ratio (at least during the more sensible period of time, in the weeks/months preceding the eruption of Nevado del Ruiz in June 2012). In a second stage, a more detailed exploration of the modifications of the instrumental line shape with temperature could be done.

We did not observe correlations between variations of the temperature and the BrO/SO₂ ratio. Also as mentioned above the temperature is rather similar throughout the year due to the geographical location and no extraordinary

temperature events were observed for the first half of 2012. The quality of the SO₂ DOAS fit given by the size of the fit error, correlates with the instrument temperature.

Similarly to the impact of temperature changes that could create artefactual variations of the BrO/SO₂ molar ratio, could there be an impact of the meteorological / volcanological conditions that could affect artefactually the gas retrieval, such as the cloudiness, ash content in the plume, etc.. Is there a way to evaluate these impacts? Could these parameters affect differently the retrieved BrO and SO₂ abundances as the retrieval is performed on a different wavelength range? (if so, would not it be more robust to retrieve SO₂ and BrO on a common wavelength range?) Other independent observations, which may not be available, could be needed to answer these questions. Nevertheless, even if the magnitude of the uncertainties associated to these impacts is unknown, it would be important to mention it in the text.

We agree with the reviewer that the impact of clouds and ash is an important topic to discuss in the manuscript. As mentioned in the answers to Referee #1, we only expect a minor influence of ash or clouds on the gas retrieval. Evaluating both trace gases in a common wavelength range is tempting in order to diminish radiative transfer influences. However, the costs for this is a much larger DOAS retrieval error and possible fit instabilities because the larger SO₂ absorption features at lower wavelengths are not taken into account. Nevertheless, we evaluated both trace gases in largely coinciding wavelength ranges (SO₂: 326.5 – 353.3 nm, BrO: 327 – 346 nm) and found similar variations of the BrO/SO₂ ratio). See Figure 10 (bottom).

- The choice of the background spectrum could also affect the quality of the spectroscopical retrieval. The authors mention the stack of in-plume and out-plume spectra to improve the signal to noise ratio. How is done the selection of these spectra? More precisely, which is the criterion used by the authors to be certain to be in (or out) the plume. A spread of the plume would not produce the nice bell shaped profile of the SO₂ column density versus viewing angle during a scan shown in Fig. 2. How typical is the profile of Fig. 2? How do the authors deal with such scans in the procedure? How are the authors confident that the background spectrum is not polluted? A pollution of the background spectrum, whose magnitude may vary with time, could indeed also produce artefactual variations of the BrO/SO₂ molar ratio.

As outlined in the text, we defined the area of 10 consecutive spectra with the highest SO₂ column density as the in-plume area and the 10 spectra with the lowest SO₂ content as the out-plume area. We did not look at the exact plume shape and summed up spectra from a rather broad field of view (~36°) for the measurement spectra as well as the FRS spectra.

However, a contaminated background spectrum does only influence the BrO/SO₂ molar ratio if it has a different BrO/SO₂ ratio than the measurement spectrum. If the BrO/SO₂ ratio is similar in both spectra, the ratio does not change and only the signal-to-noise ratio decreases. Some measurements performed from our institute (Bobrowski et al., 2006, Louban et al., 2009, General et al., 2014) indicate that the BrO/SO₂ ratio might vary towards the edges of the volcanic plume. In the following text we explain why this is only a minor problem for the data presented in this manuscript. The step size between

two viewing directions is $\sim 3.6^\circ$ in NOVAC, therefore most of the added up spectra are either inside of the volcanic plume or outside of the plume. In case that a spectrum is recorded directly at the edge of the plume (where the BrO/SO₂ ratio might be higher) the two neighboring spectra are either inside or outside of the plume as they are 3.6° apart from the “edge spectrum”. Therefore, only one out of 10 spectra is assumed to lead to variations of the BrO/SO₂ ratio and might explain to a small degree the scatter that is observed in our dataset, which is further mitigated by the summing over several spectra.

To further assess this issue, we used an approach similar to Salerno et al., 2009 and evaluated all plume and background spectra with a high resolution Solar Atlas as an FRS (publication in preparation). We used the resulting SO₂ column densities to create a filter for the data-set. All scans for which an SO₂ content of the FRS was above 25% of the SO₂ content of the plume spectrum, were removed (see Figure 2). The running mean of the filtered data-set is presented in orange. To compare this with our original data-set the running mean from the original dataset is shown as the black line in Figure 2. It can be seen, that while there are fewer data points, the running mean only shows small variations from the original plot, mainly caused by the decreased number of data points. It is therefore assumed, that contaminated background spectra only lead to small differences in our retrieval, and the complete (unfiltered) data-set is used in the manuscript for sake of better statistics.

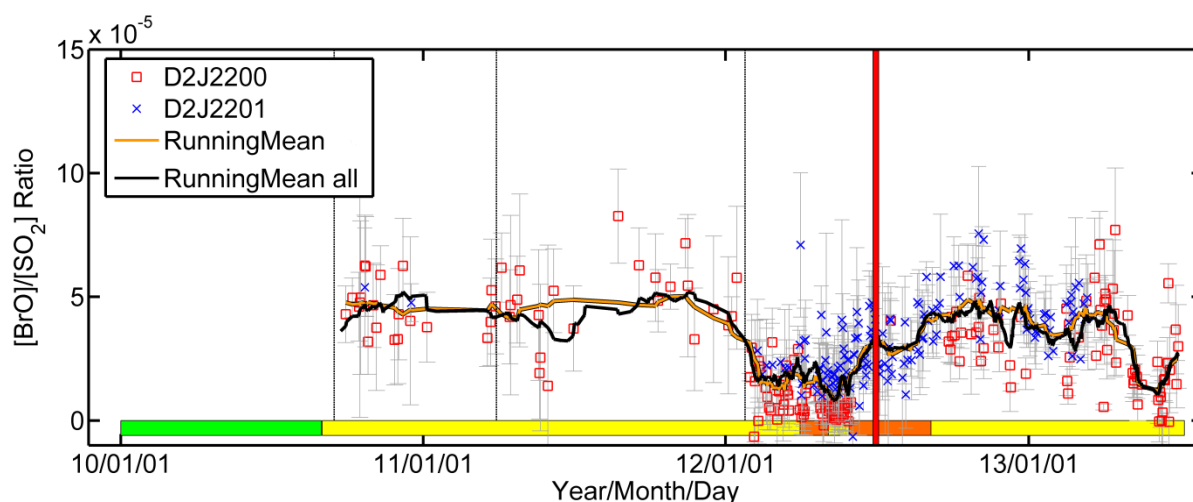


Figure 2 BrO/SO₂ ratios for all spectra that contain less than 30% SO₂ in the FRS and with a resulting SO₂ column density above 7×10^{17} molecules/cm². The black line shows the running mean for the data set containing all spectra.

- A few questions arise from the presented results of the DOAS retrieval: 1) Is the SO₂ column amount retrieved on the SO₂ retrieval window used as a constraint to correct for the contribution of SO₂ in the BrO retrieval window?

No, using the SO₂ column density retrieved in one wavelength interval as a constraint in another wavelength interval might lead to erroneous results. Generally, radiative transfer effects, like the light dilution effect, can cause a wavelength dependency of the retrieved SO₂ column density. In our revised manuscript we included radiative transfer modelling, which show that there are

indeed very small differences in the radiative transfer in both wavelength ranges.

2) It would be of interest to see the abundances of the other components of the plume which are retrieved. Is there a significance to include in the retrieval gas species such as NO₂ and CH₂O? Other studies have shown that the lifetime of NO₂ may be very short in volcanic plumes, as quickly converted to NO_y species. The point is that the inclusion of additional species (and a polynomial offset) in the retrieval could produce an artefactual retrieval of BrO if the cross sections of the various involved species can be linearly related. To explore this question, are similar values of BrO and SO₂ column amounts obtained when the cross sections of NO₂ and CH₂O removed from the retrieval? 3) In the same line of idea, is the retrieved amount of O₃ relevant?

We fully agree with the reviewer, that incorrectly included trace-gases in combination with completely free shift and squeeze values can lead to problems in the DOAS retrieval (see Salerno, 2010 and the corresponding comments by C. Kern and L.Vogel). However, the large difference between our evaluation and the evaluation of Salerno, 2010 is, that we do not allow the different absorption cross sections to shift against each other (also see Comment 5 below), which largely reduces the risk of artefactual BrO retrievals.

We did not find any significant amounts of NO₂ or CH₂O in our values and no clear correlation between these trace gases with BrO or SO₂. However, while these trace gases might not be apparent in the volcanic plume they could be found elsewhere in the atmosphere due to other sources, biomass burning, industrial pollution or cars, e.t.c. Not including a trace-gas in the DOAS retrieval, which absorption structures influence the spectra can lead to large errors in the retrieval.

We also chose the same evaluation range for BrO as determined by Vogel, 2011, which has the lowest cross interferences with other trace gases (in particular NO₂, HCHO).

As examples the correlation plots between BrO and different absorbers for all spectra with an SO₂ CD above 7×10^{17} molec/cm² (which was used as a criterion to select spectra for the BrO/SO₂ ratio time-series) are shown in Figure 3.

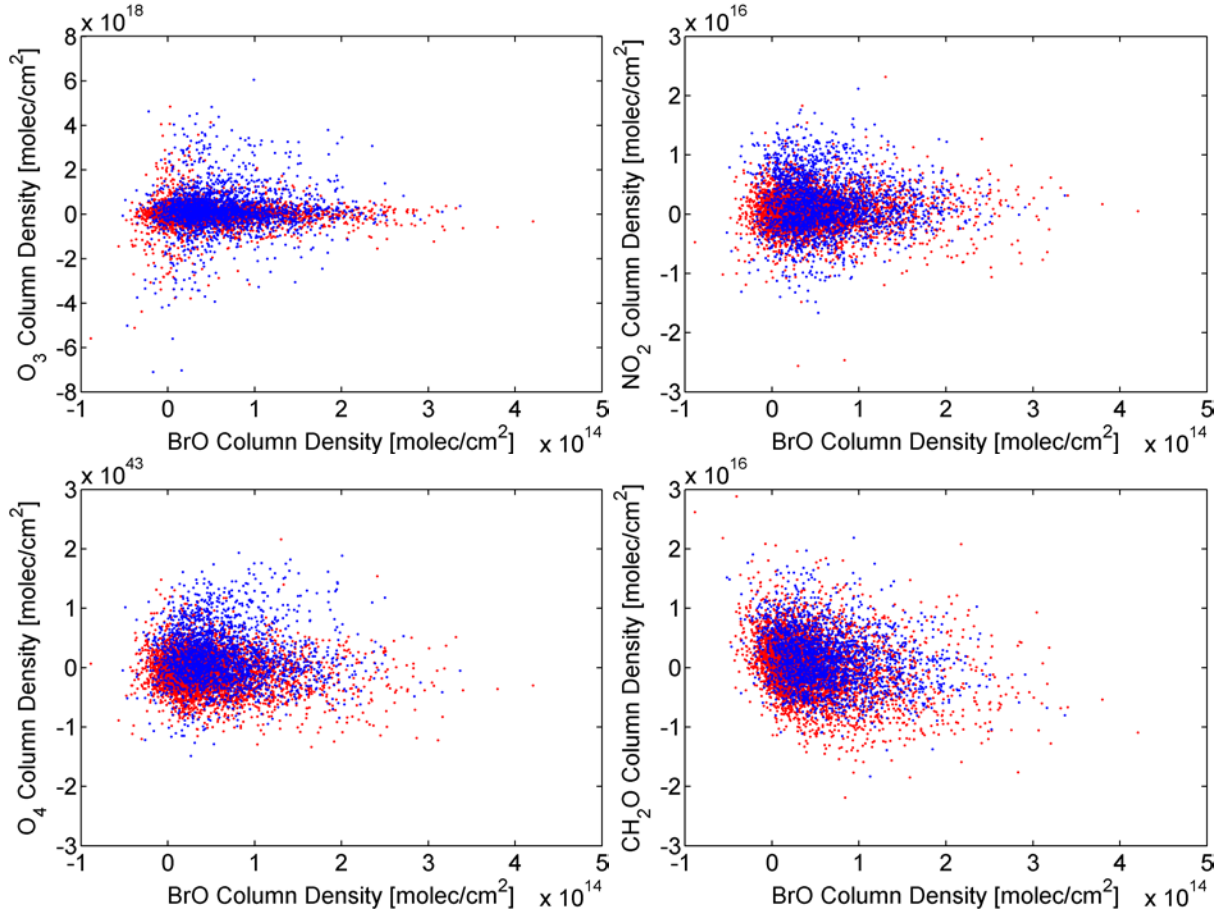


Figure 3 Correlation plots between BrO and O₃, NO₂, O₄ and CH₂O.

4) Why is an additional polynomial offset included in the retrieval algorithm? This gives an additional degree of freedom in the retrieval procedure which may jeopardize the robustness of the retrieval algorithm. (It seems that the authors mentioned a wavelength independent offset (Line 10 p1850) whereas a different label is displayed on Fig. 3 ('offset polynomial'). Is it a mistake?)

The offset polynomial allowed in the intensity space was included in the fit to account for spectrometer stray-light. We chose a polynomial of order 0, which is a constant. The wavelength dependency seen in Figure 3 is due to the fact that the offset polynomial is added in intensity space. Assuming a measurement spectrum $I_{meas}(\lambda)$ and an FRS $I_{0,meas}(\lambda)$. Both spectra have an additional constant stray-light term C that distinguishes them from the ideal measurement spectra:

$$\begin{aligned} I_{meas}(\lambda) &= I(\lambda) + C \\ I_{0,meas}(\lambda) &= I_0(\lambda) + C \end{aligned}$$

Writing down the optical density and using the fact that usually $C \ll I(\lambda)$ we can expand the optical density:

$$\begin{aligned} \ln\left(\frac{I_{meas}(\lambda)}{I_{0,meas}(\lambda)}\right) &= \ln\left(\frac{I(\lambda) + C}{I_0(\lambda) + C}\right) = \ln\left(\frac{I(\lambda)}{I_0(\lambda)}\right) + \ln\left(1 + \frac{C}{I(\lambda)}\right) - \ln\left(1 + \frac{C}{I_0(\lambda)}\right) \\ &\approx \ln\left(\frac{I(\lambda)}{I_0(\lambda)}\right) + \frac{C}{I(\lambda)} - \frac{C}{I_0(\lambda)} \end{aligned}$$

Therefore a constant stray-light term in intensity space does not appear constant in the plot, which shows the optical densities (and the complete stray-

light term). The reviewer is right that it might be confusing that we named it wavelength independent offset and in the Figure 4 it is named offset polynomial. This is the case because DOASIS provides the possibility also to include a wavelength dependent polynomial and not only a constant in the intensity space. We changed the labeling in Fig 4 to “Offset constant”.

5) Which is the range of values permitted for shift/squeeze? If well understood, shift/squeeze are the same for the reference and Ring spectrum, but it may be different to the shift/squeeze permitted for gas absorption cross-sections? What is the physical reason for this choice? It would be interesting to see if the same BrO abundance is obtained when permitting the same shift/squeeze for reference spectrum/Ring spectrum/gas cross sections. It would be also important to show the time variations of shift/squeeze with time and compare them with variations in the BrO/SO₂ molar ratio. Globally, the retrieval procedure requires more control steps to ensure a stability and robustness of the retrieval

We allowed a shift of ± 0.2 nm and a squeeze between 0.98 and 1.02. The Ring spectrum is calculated from the Reference spectrum. Therefore, it has to have the same calibration as the Reference spectrum. It should be noted, that the FRS was recorded at the same temperature as the measurement spectrum. Therefore shift and squeeze could also be restricted to 0 or 1, respectively. Our evaluation did not show any considerable shift/squeeze for the FRS and Ring Spectrum. The absorption cross-sections on the other hand were convolved on the wavelength grid of the Mercury emission lines. Therefore, the absorption cross sections might have a slightly different wavelength dependency than the measurement spectrum. Therefore, a shift and squeeze of all absorption cross-sections as one set was 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.”

- The authors show a decrease in the BrO/SO₂ ratio before the 30 June eruption of Nevado del Ruiz, which would suggest a deep source for the observed degassing. Is this in agreement with other volcanological observations at Nevado del Ruiz? Even if the goal of the paper is not to provide a detailed exploration of the volcanological processes, some additional discussion on this point could be given.

To our current state of knowledge, the solubility of bromine in comparison to sulphur is not well known. It is therefore speculative the decrease of the BrO/SO₂ ratio is caused by a shallow or a deep source. We added a subsection (Section 3.3) in the results section, in which we compare the observed BrO/SO₂ ratios with the number of volcano-tectonic and long-period seismic events per month. This section also includes a brief discussion of possible degassing interpretation. In this section we also compare our observed BrO/SO₂ ratios with data on seismic activity that was taken from the technical reports of the Servicio Geológico Colombiano (see Figure 4 and 5).

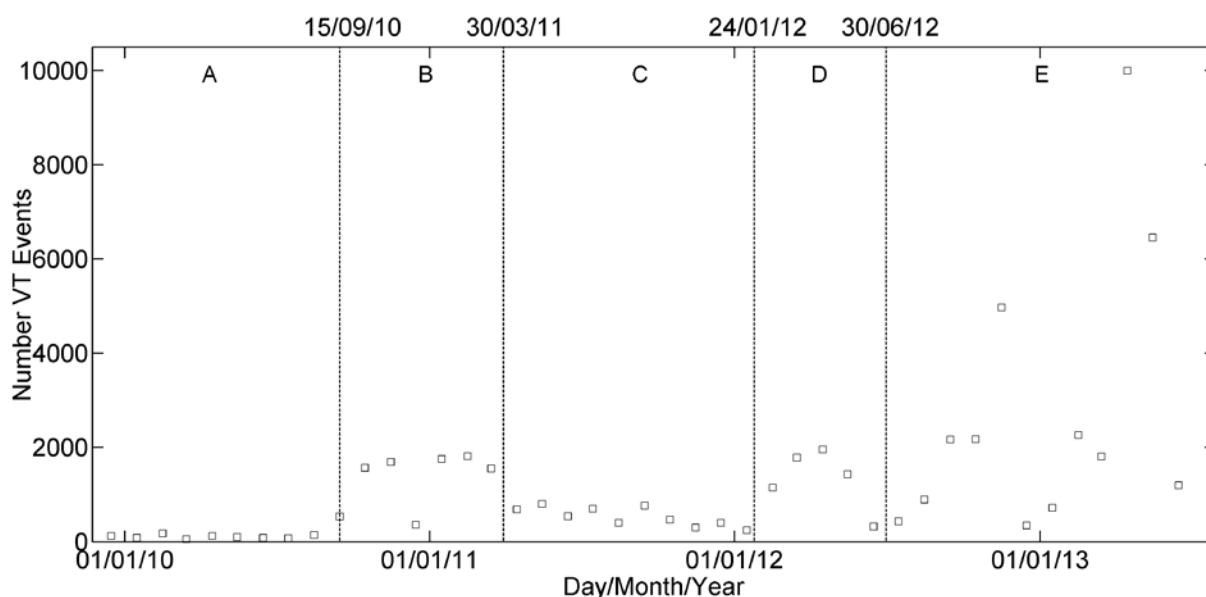


Figure 4 Number of volcano tectonic events per month. Data taken from the SGC reports from <http://www.sgc.gov.co/>.

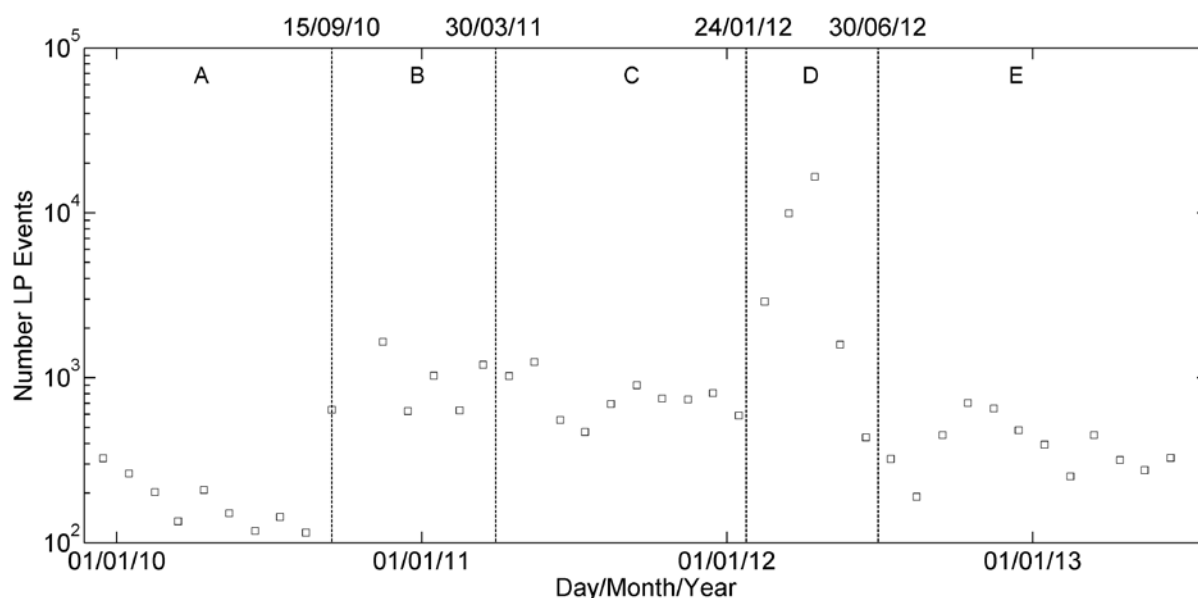


Figure 5 Number of long period seismic events per month. Data taken from the SGC reports from <http://www.sgc.gov.co/>.

Minor comments:

- To allow for an easy comparison of the time-evolutions of different parameters (SO₂ flux, SO₂ column amount, BrO column amount (which are presently displayed for on different figures), it would be better to merge different figures by making sub-plots, such as Figures 6, 7 and 8. Similarly, for an easy comparison of the different results, it would be better to merge Figures 9 and 10.

We agree with the reviewer and adapted the figures according to his suggestions. Figure 9 in the new manuscript merged figures 6, 7 and 8 from the old manuscript. For a clearer presentation the scale of the SO₂ emission rates was changed to a logarithmic scale. Figure 10 in the new manuscript merges Figure 9 and 10 from the old manuscript.

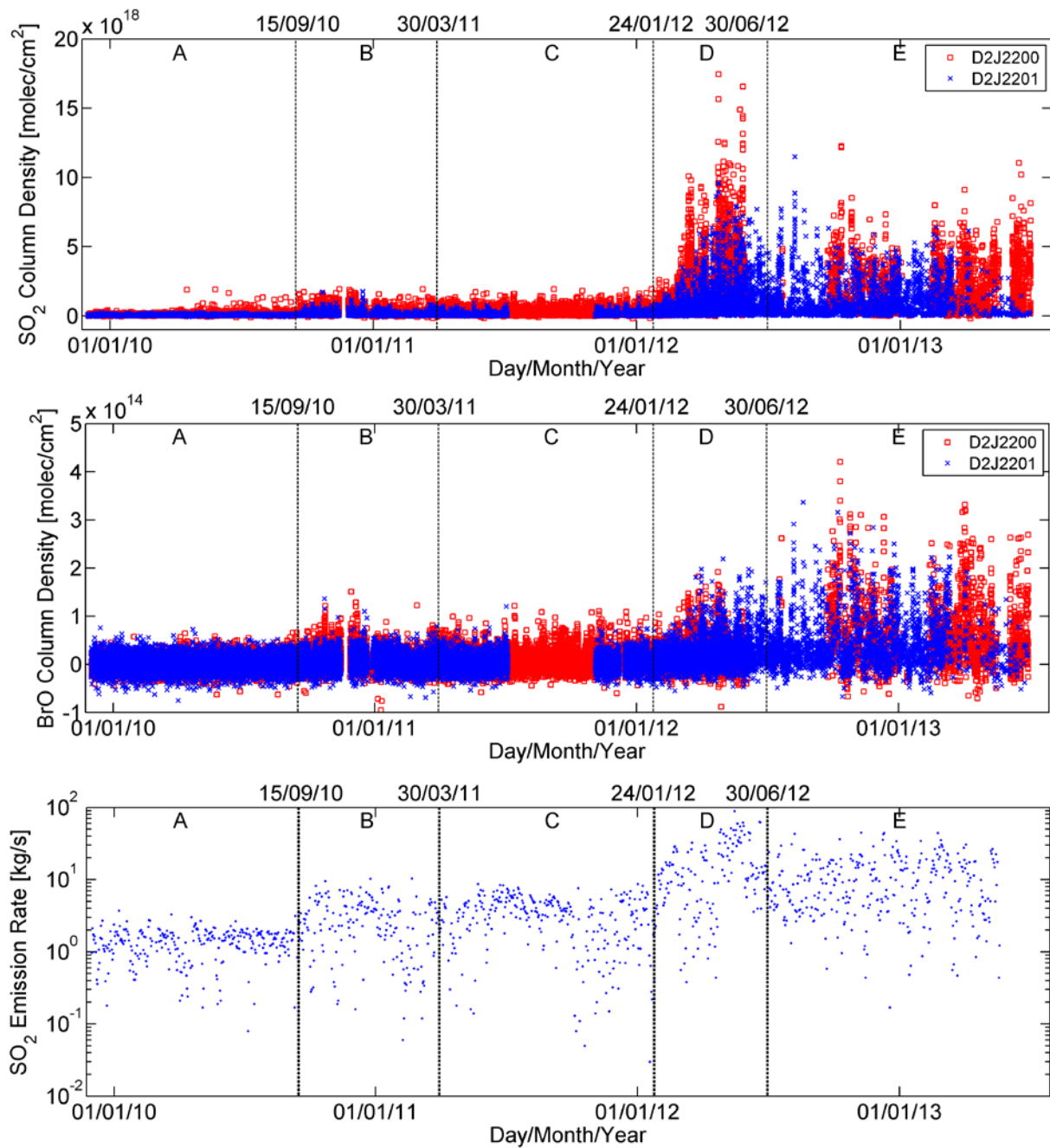


Figure 9 Time-series from the two instruments at Nevado del Ruiz. The instrument D2J2200 is located in a way that measures elevated SO₂ and BrO column density values more frequently (see Fig. 1). The spectra evaluated were created from co-adding reference and plume spectra from four consecutive scans. Top: SO₂ CDs Centre: BrO CDs Bottom: Daily averages of the SO₂ emission rates. The wind-speed was taken from the ECWMF database, more details in Galle et al. (2010) and Arellano et al. (2013).

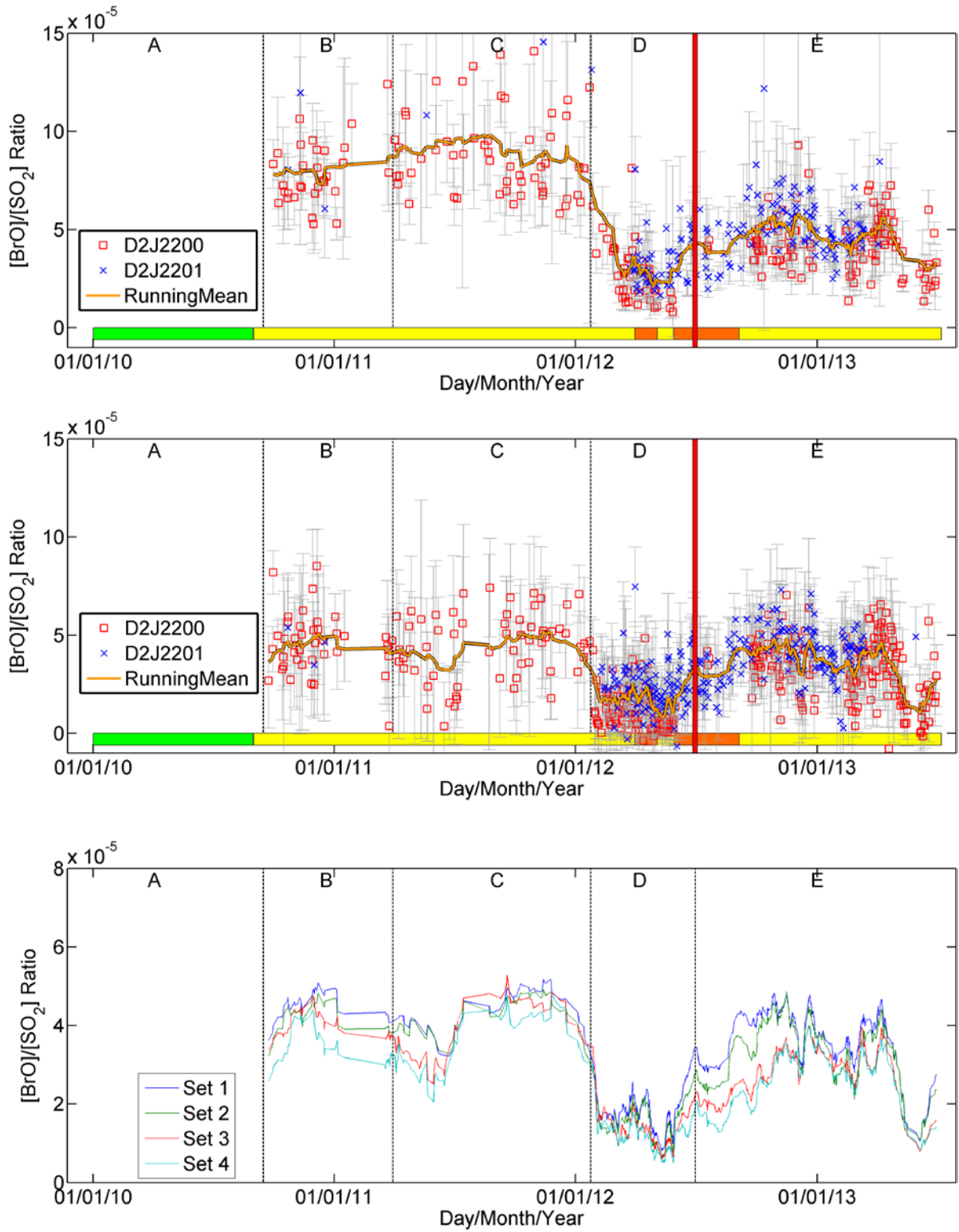


Figure 10 Daily averages of the BrO/SO₂ molar ratios from Nevado del Ruiz from November 2009–June 2013. The ratio drops down to values below 5×10^{-5} , and increases again after the eruption on 30 June 2012 (red vertical bar). Also shown at the bottom of the image is the volcanic activity risk level defined by the Colombian Geological Survey (coloured horizontal bars). The black-orange line shows a running mean around 7 data points (from both instruments). Top: The criterion for a valid measurement is a BrO column density above four times the BrO DOAS retrieval error. Centre: For this figure an SO₂ threshold of 7×10^{17} molecules cm⁻² was chosen as the criterion for valid data points. Bottom: BrO/SO₂ time series retrieved in different wavelength ranges, with an SO₂ threshold of 7×10^{17} molecules cm⁻² as the criterion for valid data points.

- Line 1 p1854: Ash emissions are mentioned before June 2012? If ash is observed, this means that magma has reached the surface and has erupted. Eruptive episodes are consequently recorded before June. This is not clear in the short volcanological background which has been given in the manuscript. This would require more developments as this has an impact on the interpretation of the BrO/SO₂ molar ratio time variations in terms of magma dynamics.

As mentioned in Garzón et al. (2013), ash emissions were already observed at the end of February 2012. To our knowledge this is also not a unique situation that ash emission is observed without an eruption or even lava emissions. Ash emission can be also caused e.g. explosions below the surface, caused for instance by old material slighting inside the crater and blocking for a short moment the degassing until the increasing pressure push it into the air – resulting in ash observations – there are many other possibilities like that.

Section 2 and 3 should be merged in a 'Method section' with sub-sections. In this new section, a sub-section describing in more details the activity of Nevado del Ruiz in the last years would be welcome. The present lines 16-21 of p 1848, which are irrelevant at the end of the Introduction section, could be moved to this new sub-section.

We combined Section 2 and 3 to a 'Method section'. However we included the discussion of the volcanic activity in the results section.

Lines 5-9 of p1854: should be moved elsewhere.

We removed Lines 5-9 on p1854, as the information is redundant and can be found on p1852 Lines 9 – 16.

Section 5 also includes some discussion of the results and could consequently be named differently than 'Conclusions and outlook'.

We renamed the section to “Discussion of results and outlook”

References:

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