

Answer to Interactive comment on “Retrieval of absolute SO₂ column amounts from scattered-light spectra – Implications for the evaluation of data from automated DOAS Networks” by Peter Lübcke et al.

Dear Editor,

We like to thank referee #3 for the comments and suggestions. We implemented most of the suggestions into the revised version of our manuscript and give detailed reasons in the cases where we could not follow the reviewer’s suggestions. With these changes (and those in response to the other reviewers’) we are confident that the revised manuscript has improved considerably. Please find the comments (in normal face) and our detailed answers (in bold face) below. The page and line numbers in our response are reflecting the page and lines numbers of the original manuscript:

Anonymous Referee #3

Received and published: 13 May 2016

Review of Manuscript AMT-2016-24: Retrieval of absolute SO₂ column amounts from scattered-light spectra - Implications for the evaluation of data from automated DOAS Networks by P. Lübcke et al.,

Recommendations I recommend publication in AMT only after a MAJOR REVISION of the manuscript in line with criteria requirements of the Journal.

General comments This paper discuss the implementation of a modelled Fraunhofer Reference Spectrum (FRS) for retrieving SO₂ Column Density (CD) from open-path ultraviolet spectra by scanning ultraviolet spectrometer. The authors carry out a number of specific statistical analysis (e.g., PCA) such as to identified instrumental features of spectrometers and explore confidence in the retrieval. Data from NOVAC network at Nevado del Ruiz (Colombia) and Tungurahua (Ecuador) between 2010 and 2012, and 2009 and 2011, respectively were reduced applying both the standard NOVAC (e.g., Galle et al., 2010) and the FRS approach. Comparing the results gathered from both methods, the authors observed a large difference between the two, with the NOVAC underestimating SO₂ CDs more than a factor of two. At volcanology observatories self-reliance from robotic system is a first priority for real-time multidisciplinary surveillance and monitoring purpose. NOVAC is largely spread at active volcanoes worldwide representing a unique trail for the understanding of volcanoes and their gas emission impact in climatology. Hence, the application of the modelled FRS at NOVAC represents a step forward for both monitoring purpose and revaluation of global volcanic emission rates inventory.

In the appreciation of our study, the referee mentions the discrepancy shown in our study of more than a factor of two in the SO₂ - SCD between the standard evaluation and our proposed method. It should be stressed, however, that such a difference does not necessarily propagate to the final emission rate estimation, calculated by the robotic surveillance systems, because in most of those cases, the NOVAC algorithm would discard the scan measurements by evaluating its shape (see Galle et al., 2010 for an exact definition of the completeness value). The observatories will have no flux determination under those conditions. The main contribution of our analysis is to identify the reasons of

such data gaps (it would mean that there are low plumes or high but wide plumes, instead of no gas, changing completely the interpretation of the emissions), but the flux calculation will remain challenging in such conditions.

We added the information on the completeness value on page 18, line 11.

I believe this paper is in line with the scope of AMT. However, there are some aspects of the work that are questionable, and in which a strong revision is required before the manuscript is suitability for publication in AMT. There are also several minor points to address, which I have described with line references after my general comments.

The main issue concern the originality of this study. This research is not a novel idea in SO₂ measurements at active volcanoes by scanning spectrometer system. In particular, the pioneering application of a modelled FRS in NOVAC data at the Piton de la Fournaise has been not mentioned and/or discussed (Hibert et al., 2015). In this work, the authors successfully retrieved SO₂ CD and flux from the NOVAC scanning network without invoke different ILS as in the manner of Salerno et al., 2009a and no laborious PCA analysis for accounting of instrumental effects in the spectrometers.

Nevertheless, Lubcke et al., set the originality of their study in the PCA analysis.

We thank the referee for correctly pointing out that a reference to the study of Hibert et al., 2015 was missing from the manuscript. We apologize for having missed this article before, and we now properly reference it on Page 2, line 30-32.

However, we sharply disagree with the reviewer's statement that this influences the originality of our study in any way. On the contrary, the article by Hibert et al. 2015 highlights the need for in depth discussion of the topic for the following reasons: Hibert et al. 2015 only mention the use of a modelled FRS as a side note, without giving any details on the technical implementation. Their article concentrates on a different topic and analyses only very few spectroscopic data. Figure 8. in Hibert et al, is characterized by several data gaps and only shows data from six days. The authors of Hilbert et al., 2015 compared the SO₂ emission rates from an evaluation using a spectral catalog FRS approach with NOVAC-style evaluations. However, the mixing of spectroscopic retrieval and geometric and meteorological considerations for the emission rate calculation makes it difficult to single out the effect of the spectroscopic retrieval. It is also not possible to judge from the Hilbert et al., 2015 manuscript, how successful the application of their approach actually is with respect to the quality of the spectroscopic retrieval. Furthermore, discrepancies in the results of Hibert et al., 2015 show the need for studies like ours (or the recent manuscript by Burton and Sawyer, 2016 on a similar topic): The FRS evaluations presented in Hibert et al. yield lower SO₂ emission rates than the standard NOVAC approach. Applying a solar catalog spectrum should always result in SO₂ SCDs that are at least as high as the SO₂ SCDs derived by the standard NOVAC approach (or higher in case of contaminated reference spectra).

Our manuscript demonstrates that the PCA approach that we used in our study to identify instrumental effects can greatly improve the robustness of the retrieval and thus lead to reliable results. SO₂ has a strong absorption cross-section in the UV, combined with the high abundance in volcanic plumes this results in optical densities that can easily be much

larger than the residual structure found by the PCA in our analysis. Therefore, the SO₂ absorption structure might be large compared to the residual, but the combination of different absorbers (in particular O₃), FRS and Ring Spectrum might lead to interferences that can lead to large errors if instrumental effects are not taken into account (e.g. by the PCA approach).

The exact technical implementation of the FRS approach is an important and major point of our study, but it is not the only original idea presented in our manuscript. We want to point out that no other study before has reported actual figures on the influences of SO₂ containing references on the long-term SO₂ emissions of active volcanoes. We studied Tungurahua and Nevado del Ruiz, - two different volcanic systems with different settings of the instruments around these strong or medium level emitting volcanoes. While Salerno et al., 2009b compares a modelled FRS approach with traverse measurements and finds good agreement at Mt. Etna, the authors do not state which kind of error would be introduced by using an FRS measured from the same instrument (before the scan). Our study showed the magnitude of possible influences as well as the variability of the effect. SO₂ containing reference spectra occurred in particular at strong degassing volcanoes, where scanning systems are installed at relatively close distance to the active crater and under weak wind condition.

We also like to stress that only our approach offers the possibility to study this issue without revisiting the field instruments and it therefore offers the possibility to apply this method on the whole, already existing, NOVAC data set. A characterization of the field-deployed NOVAC instruments to determine their specific properties, such as detector quantum efficiency and the pixel-to-pixel variation of the detector sensitivity, but also the characteristic changes of their instrument slit function with temperature, is not feasible. Moreover, a number of instruments are not functional anymore and thus will be forever unavailable for characterization. Thus our method is ideally suited to reprocess already existing datasets of such instruments.

The authors carried out this statistical test in free-volcanic gas spectra selected from the database of Nevado del Ruiz and Tunguraha NOVAC network. Selected 7-days data between Sept-Oct at Nevado del Ruiz and 10-days/year between 2009 and 2011 at Tunguraha. Results, were assumed reflecting the instrumental features of the entire network. However, as reported by Pinardi et al., 2007, instrumental effect dramatically may change over time due to temperature drift, and these are different between spectrometers (eg., Instrumental line function, stray lights).

We chose a set of 7 or 10 days to determine the instrumental effects for each spectrometer individually (see Fig. 3). As correctly mentioned by the referee, we created a set of principal components at Tungurahua for each year (and each instrument) since we observed long term drifts in the instruments response, leading to a decrease of the retrieval performance, when performing only one PCA (per instrument) for the entire data-set. We did not imply that our results show the exact features of the entire NOVAC network. However, as all studies on evaluating SO₂ absorption with a modelled FRS, we assume that the spectrometers (which after all are all of the same type) behave in an essentially similar way and we believe that this approach can be adapted to other instruments.

We discussed the instrumental features the referee is referring to such as temperature (see below) and long-term drifts in our manuscript. Figure 5 clearly shows the long term drifts for one instrument at Tungurahua, while Figure 4 shows, that the principal components are more constant at Nevado del Ruiz. The variation of the first two principal components for the three other instruments are shown in Figures 17 – 19 (or Figs. D1 – D3 in the appendix of the revised manuscript).

To clarify that the PCA was performed for each instrument individually, we changed the sentence on Page 6, Line 1-2 from “Information about these absorbers were obtained from the spectra themselves by using a PCA” to “Information about these absorbers were obtained for each instrument individually from the spectra themselves by using a PCA..” We also added a discussion of the long-term variation of the SO₂ Fit error for instrument I2J8548 to Section 4.4.

As the NOVAC instruments are not thermo-stabilised (Galle et al., 2010) variable instrumental drifts may take place at different magnitude and time scale. This raises the hesitation to believe that the PCA results gathered from few data and from a unique spectrometer (per network) may be assumed representative of the instrumental features of entire scanning networks.

As mentioned above, the PCA was performed for each instrument individually. The variation of the principal components fit coefficient with time and temperature are shown in Figures 4 and 5 and D1- D3 in the appendix. Section 4.2 was edited in order to clarify this point.

Regarding the existence of temperature drifts, the referee is absolutely correct. However, the instruments in the NOVAC network are typically exposed to a wide variety of temperatures during each day (low temperatures in the morning and increasing temperatures during the day). We showed that some of the main PCA components scale within the observed temperature range linearly with temperature. Thus the temperature variation is at least partially captured by the PCA and in this way taken into account by our evaluation. Figure 12 in our manuscript shows that the quality of the SO₂ fit indeed degrades to some extent for temperatures below 10°C. However, this is most likely not caused by problems with the principal components but rather due to variations of the ILF as shown in Pinardi et al., 2007. This is reflected in our manuscript on Page 5, Lines 15-19 and Page 19, Line 13 – Page 20, Line 3.

The authors, attempt to explore this issue by comparing the residual structures and optical depth (Fig. 3, 4, and 5) obtained by retrieving the entire records of data from the two networks.

Figures 4 and 5 are only examples at this position in the manuscript. The same type of illustration for the remaining instruments were shown in Figs. 17 – 19. These figures are now moved to Figs. D1 – D3 in the appendix of the revised manuscript.

They found a reasonable agreement between the instruments. This finding arises anyway a further question. In detail, since the results of the comparison relates to the PCA results and as instrumental drifts are taking place physically in different way, the question that come up is whether the statistical PCA has a fundamental role in the framework of the paper.

We do not follow the concerns of the reviewer. First, we fail to understand how a reasonable agreement obtained by statistically relevant numbers of measurements should make the statistical importance of the PCA questionable? The referee has addressed the fact that different instruments at different locations may be subject to physically different processes. The Ocean Optics S2000 spectrometers at all volcanoes are of the same design, and it should come as no surprise that they behave similar with temperature changes (as was also shown in Pinardi et al., 2007). However, while the instruments show similar behavior overall, subtle differences between the instruments are present. Trying to evaluate data from one instrument with the principal components obtained from a different instruments results in extremely poor quality of the fit.

Additionally, (as mentioned on Page 6, Line 7 in our manuscript) another potential source influencing the principal components are uncertainties in the Solar Atlas (as suggested by Burton and Sawyer, 2016), which should lead to a similar effect for all instruments. Furthermore, we also reported influences of long-term and temperature drifts (for example Page 10, Line 1-3). If the referee compares Fig. 4 for (the individual instrument) D2J2201 with figures 12 (SO₂ Fit Error as a function of time) and 13 (SO₂ Fit Error as a function of temperature) he/she could observe, that there are indeed variations with instrument temperature as well as long term drifts. Similarly drifts of the principal components can be observed at Tungurahua as well (see Fig. 5 and Figs. D2 and D3 in the appendix). We added a discussion of the SO₂ fit error for instrument I2J8548 to Section 4.4 to show an example of an instrument with long term drifts.

There are also some comments about the structures of the article. The paper needs to be restructured, reducing the number of paragraphs and length of the article itself. For instance, the detailed description of the DOAS technique is unnecessary. This technique is a very well established spectroscopy approach and the theory largely published. I really suggest the authors to replace details and equation with references (Platt and Stutz, 2008). Moreover, some paragraph might be eliminated or merged with others, e.g 4.2 consist of only five lines. Instead, I'd suggest the authors to include a small paragraph describing the NOVAC networks object of the study. Finally, a graph of the SO₂ CDs retrieved by FRS throughout the years at both Nevado del Ruiz and Tungurahua should be displayed for completeness of the study.

We thank the referee for his/her suggestion and rewrote/shortened the paragraph regarding the DOAS technique. However, we feel that a minimal discussion of the technique is necessary for the understanding of the PCA in the DOAS context. Page 3, Line 19 – Page 4, Line 7 were replaced by a shorter summary that refers to the literature. Regarding the reviewer's suggestion to add a graph of the SO₂ CDs retrieved by the two approaches: We feel that this would lead to an even longer article without improving its clarity and therefore decided to include these figures in the appendix of the manuscript as Figs. B1 – B5 (The figures are also shown at the end of this reply). We added a more detailed description of the NOVAC installations at the volcanoes that were subject to this study at the beginning of the result section.

Other comments Abstract: Pag 1, Section 05, line 1: define here the scanning DOAS, change this sentence with ‘ : : : : ..scanning ultraviolet spectrometers network, also named as scanning DOAS ’.

We changed the sentence on Page 1, Line 1 from “Scanning Differential Optical Absorption Spectroscopy (DOAS) networks using scattered solar radiation have become an increasingly important tool for monitoring volcanic sulphur dioxide (SO₂) emissions.” to “Scanning spectrometer networks using scattered solar radiation in the ultraviolet spectral region have become an increasingly important tool for monitoring volcanic sulphur dioxide (SO₂) emissions. Most frequently, measured spectra are evaluated using the Differential Optical Absorption Spectroscopy (DOAS) technique.” as suggested by the referee.

Pag 1, Section 10, line 3: specify volcanoes.

The volcanoes are specified one line below.

Pag 1, section 20, line 1: ‘complicated instrumental calibrations’ please report which kind of calibrations are required in field ‘eg.,.....’ .

We added the following text at the above mentioned location: “...instrumental calibration in the field (e.g., by employing calibration cells or broad-band light sources)...”.

In our understanding the methods described in Salerno et al., 2009 is based on empirically tuning some of the retrieval parameters in order to reproduce the SO₂ column densities of known calibration cells. The method described in Burton and Sawyer (2016) needs measurements with a broadband light source to characterize the instrument.

Pag 2, section 05, line 3-4: It’s not clear if the word ‘New’, is related to the FRS or the entire study carried out in the paper. In case it relates to the FRS method, as mentioned in the general comment, I’d suggest to remove ‘new’ because it is not a novel approach.

We deleted “new” from the sentence, however obtaining instrumental effects from the residual structures of the spectra in a systematic manor is a novel approach.

Introduction Pag 2, section 10, line 2: add reference Williams-Jones, et al, 2008 and Oppenheimer, 2011

We thank the referee for this suggestion, but think that the COSPEC is appropriately introduced with the two given references. Even more since we are trying to reduce the length of the article (as demanded by the reviewer in a comment above).

Pag 2, section 10, line 2-3: More recently the availability of miniature spectrometers allowed: : : : :’: I suggest to delete more recently because it’s now more than twelve years after the first application of this technique at active volcanoes by Galle et al., 2003.

We thank the referee for this suggestion and deleted “more recently”

Pag 2, section 10, line 5: add reference ‘Elias et al, 2006.

We added the reference to Elias et al., 2006.

Pag 2, section 15, line 2-3: change 'One of the first installations were scanning DOAS instruments at Montserrat volcano (Edmonds et al., 2003)' with 'The first installations of scanning-DOAS network was developed at Montserrat volcano (Edmonds et al., 2003)'.
We changed the sentence to "the first installations of ... were done Montserrat volcano (Edmonds et al., 2003)"

Pag 2, section 20, line 1: add reference Salerno et al., 2009b.
We added the reference to Salerno et al., 2009b.

Pag 3, section 15, line 1-4: specify large NOVAC database of the two Colombian and Ecuadorian volcanoes object of this research.
We changed the sentence to : "Data from the NOVAC database at Nevado del Ruiz and Tungurahua gives us the...."

Pag 3, section 30, line 5-7: move and change 'First steps towards the here described approach were taken in Lübcke (2014), where measurements from NOVAC instruments at Nevado del Ruiz were evaluated for SO₂ with a modelled background spectrum'. With 'this approach was successively adopted by Lübcke (2014) and Hibert et al., 2015 for evaluating NOVAC data collected at Nevado del Ruiz and at Piton de la Fournaise Reunion, respectively'.
We changed the sentence with small variations: "Different variations of this approach were used by Lübcke (2014) and Hibert et al., 2015 for evaluating NOVAC data collected at Nevado del Ruiz and at Piton de la Fournaise Reunion, respectively."

Pag 3, section 10-15, line 3-3: move and rephrase this section in Pag. 2 section 25-30 while talking about the use of the modelled FRS. Report that limitations and uncertainty of the standard NOVAC retrieval has been already discussed and overtaken applying a modelled background FRS by Hibert, C., et al. 2015.

We have added a reference to Hibert et al. 2015 on page 3, Line 5-7 (see answer above). However, we must stress the great differences between Hibert et al., 2015 and this work. The authors of Hibert et al., 2015 focus on calculated emissions and do not further elaborate on the use of modeled FRS. This was apparently applied however to individual spectra forming the bases of subsequent flux calculations. This leaves a great room for debate on the source of observed differences in fluxes, and which one better captures reality.

We agree, and point out in our manuscript, that at times significant differences between the standard NOVAC evaluation and the modeled FRS method exists in the column density derivations. However, the spectroscopic retrieval methods must be discussed in detail and require close scrutiny to allow repeatability and application in retrieval algorithms. This is where the extra value of our manuscript lies, while Hibert et al. 2015 has an entirely different focus.

Furthermore, the difference of both methods could also be caused by radiative transfer causing a fake SO₂ signal in FOV directions outside the plume, which would lead to an error prone value using the modelled FRS approach, overestimating the real flux. As far as we know this hasn't been discussed in literature before but goes to the point that it might be difficult to judge which of the two methods leads to more accurate results. Comparison

with traverse measurements cannot help this issue, since these measurements could theoretically be influenced by the same problem.

2 Background spectra for scanning DOAS instrument networks at volcanoes

As reported in the general comments, the theory behind the DOAS evaluation is widely published. Thus allocate paragraph to a detailed description of the physics/equation of DOAS does not provide any advantages to the article. I'd suggest to replace part of this section with Platt and Stutz, 2008.

We thank the referee for this suggestion. Our intention was to give a more accessible introduction in order to guide the reader to the modelled FRS. Following the referee's suggestions we deleted Page 3, Line 19 – 29 and Page 4, Line 1 – 8. We added an abbreviated introduction to the topic.

Pag 4, section 30, line 7-8: The experiment reported in Salerno et al., 2009a does not imply any routinely specific spectroscopy retrieval operations to be performed in field.

The abstract of Salerno et al., (2009a) says “We describe the method here and tune it empirically by collecting clear, zenith sky spectra using calibration cells containing known amounts of SO₂.”

In order to tune the method for a new instrument (as the referee pointed out correctly above, the instruments are not exactly the same) a new set of calibration cell measurements is necessary. While Salerno et al, 2009a showed that the parameters found in their study worked for other instruments installed at Mt. Etna this does not necessarily imply that they are also valid for instruments installed at other volcanoes. This follows directly from the argument given by the referee himself on the individuality of instruments and difference in temperature variations.

For NOVAC stations that are already installed in the field the approach of Salerno et al. (2009a) would result in retrieval operations performed in the field. These operations should be performed on a routinely basis since long-term drifts could occur, as was also correctly pointed out by the referee.

SO₂ calibrated quartz cells spectra were employed for exploring and validate at different timescale the application of a modelled FRS for reducing ultraviolet open-path spectra from scanning ultraviolet spectrometers. The comparison of three year of SO₂ CDs an flux form scanning system and traverses (Salerno et al., 2009b) provide a further quantitative constrain on the efficiency of the approach developed in Salerno et al., 2009a.

See discussion directly above. We also think that our approach delivers the high level of accuracy that is necessary in order to make a quantitative comparison of the SO₂ CDs from different methods that we wanted to achieve in our manuscript. In order to validate the influence of the spectroscopic retrieval itself, it is more straightforward to compare the retrieval methods on one common data-set. This approach also allowed to identify systematic deviations between the dSCDs from the two retrieval methods (see Fig. 6 and Table 1) as well as differences between the SCDs (Figs. 7, 9, 14 and 15).

Pag 5, section 10, line 1-5: rephrase this section and delete math equations.

We rephrased the section and deleted the math equation.

Pag 5, section 15, line 4-5: As reported in the paper (pag 6, section 20) SO₂ retrieval was performed between 310.0 - 326.8 nm, report the reason for which high resolution laboratory cross-sections were convolved using the Hg line at 334.15 nm and not for instance 313.16 nm.

We are surprised by this suggestion: The Hg line recordings for the 313.16 nm line show that the spectral resolution of the installed instruments is not sufficient to separate it from the 312.57 so this pair of mercury lines appears as one single peak in the spectra and therefore doesn't provide a possibility to use it as instrumental line function.

3 Data evaluation 3.1 Settings of the DOAS retrieval Pag 6, section 15, line 5: 'for both retrieval methods (see below)' define here the two retrievals.

We moved the definition of the retrieval methods from Page 6, Line 27 – Page 7, Line 10 to the beginning of the section on Page 6, Line 11.

Pag 6, section 25-30, line 4-9: detail of standard NOVAC retrieval has been already reported in the introduction, delete this section or synthesise.

While we give a general description in the introduction, these lines explain in more detail what was done in the data retrieval of this manuscript. Additionally, in our opinion it greatly helps the reader of the manuscript to have these lines in the definition of the different methods.

Pag 7-8, section 30 – 30: replace DOAS theory with references.

Assuming that the referee refers to Page 7, Line 31 – Page 8, Line 3, we disagree. While these methods are standard approaches in the DOAS community they are oftentimes unknown to users at volcanoes and consequently ignored. Additionally, these lines give an exact description of what was done in the work of this manuscript (e.g. that the SCD for the I₀ corrected convolution of both O₃ cross-sections was the O₃ SCD retrieved at 221K...), which is necessary to reproduce the results.

3.3 Principal Component Analysis for Method B Pag 9, section 10-20, the authors report that PCA needs to be run using clean-gas spectra. However, due to volcanic activity this is not so straightforward. Therefore, only few dataset of spectra were selected from both Nevado del Ruiz and Tungurahua database. Nevertheless, at Pag 14 paragraph 4.3 the requirements of clean-gas spectra arise again for evaluating the sensitivity of the retrieval as zero CDs. As reported in this paragraph at section 5, line 2-4 a greater number of data-days were discriminated at both volcanoes respect what have been selected for the PCA analysis (e.g, at Nevado del Ruiz 7 days vs 73 days). Please, rephrase the two section in case of mistake or clarify this conflict issue.

We identified a small set of days (7 or 10) that is sufficient for creating the principal components combining spectral analysis and information on the level of degassing activity from the observatories. This subset was used as our training dataset for the PCA. After

evaluation (including the PCAs) we looked at the results and found a larger number of days without significant SO₂ emissions.

Using all gas free days for the creation of the principal components could have resulted in overfitting. It is a quite common approach to train a model based on a small subset of data and evaluate its performance using a different dataset.

We added to Page 9 Line 32 – Page 10, Line 4: “Choosing only a small set of a few days for the PCA has several advantages. For one, it allows us to investigate the performance on gas free days that are not part of the PCA sample. Using all gas free days for the PCA would by definition shift the average SO₂ CD to zero on these days. The second advantage is of a more practical nature, in real-life it is not always feasible to manually investigate a large amount of data to find gas-free days, and only a few days have to be sufficient.”

Additionally, we changed “sample data” on Page 9, Line 31 and Page 10, Line 3 to “training data” in order to use more common terminology and clarify our approach.

Pag 9, section

15, line 5: constrain in a quantitative scale the meaning of ‘little degassing activity’. Pag 9-10, section 30-5, define period of selected data at Tungurahua. Pag 10, section 30, line 7: constrain in a quantitative scale the meaning of ‘low volcanic degassing activity’.

We chose days on which the majority of spectra had an SO₂ CD that fulfilled “Only spectra where the absolute value of the SO₂ CD was smaller than twice the DOAS fit error were considered in the PCA.” (Page 9, Line 20-21).

3.4 Implementation of the new algorithm

I would suggest the authors to remove this paragraph. It is a summary of what has been already reported in the manuscript.

We moved this section into the appendix. The spectroscopic evaluation is quite complex and we believe that this summary will help fellow scientists to reproduce the steps necessary for the evaluation.

4 Results

Pag 11, section 25, line 1: delete brackets in Lubcke et al., 2013.

We deleted the brackets in the reference.

Pag 14, section 05: see comments Pag 9, section 10-20. Conflict or erroneous explanation of this issue.

See explanation above.

Pag 17-18, section 05. Method A and B retrieve the same spectra, explain in more detail why the effect of stray light should affect the two retrieval performed with method A and B in a different way.

As the referee correctly points out, the evaluated spectra are the same, however, for Method A also the FRS is exposed to spectrometer stray light. Thus, while in Method A the amount of stray light between FRS and plume measurement might differ, only the plume measurement is influenced for Method B. In Method B the fit has to completely compensate for stray-light (which is done by an additional polynomial in intensity space)

and may be mitigated if structures are captured by the PCA. We added on Page 18, Line 1: “While the measurement spectra are similar for both methods, only the FRS of method A is influenced by stray-light.”

4.2 DOAS fit example This paragraph consist of only five lines, please remove it or include in a different paragraph.

We combined these lines with section 4.4 and renamed Section 4.4 to “Fit quality and SO2 fit error for Method B”.

Figure Fig 9: change ‘which is similar to the standard NOVAC approach’ with ‘standard NOVAC approach’.

Other parameters for the selection of valid spectra were used in this study when compared to the standard NOVAC approach. The formulation used in the caption of Fig. 9 is thus more accurate.

Appendix:

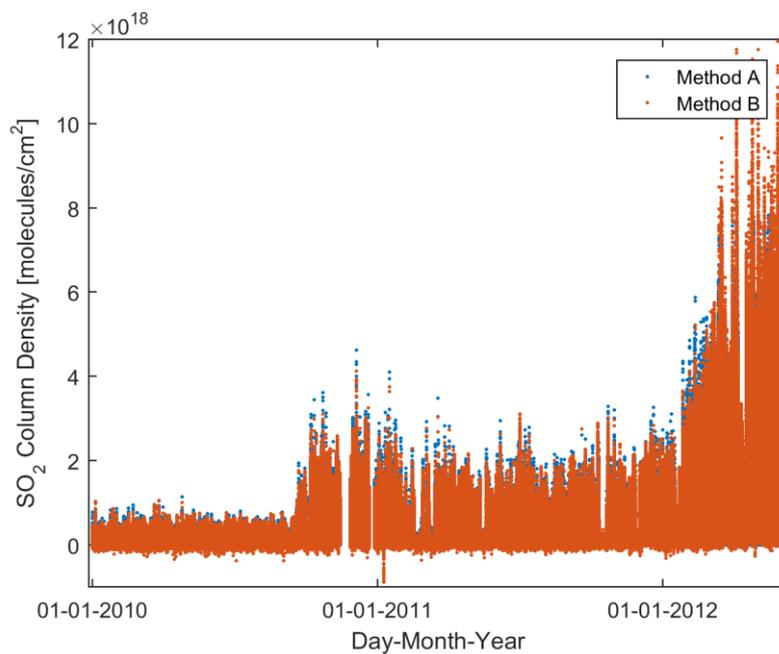


Fig. 1 Time-series of the SO₂ CDs for instrument D2J2200 at Nevado del Ruiz.

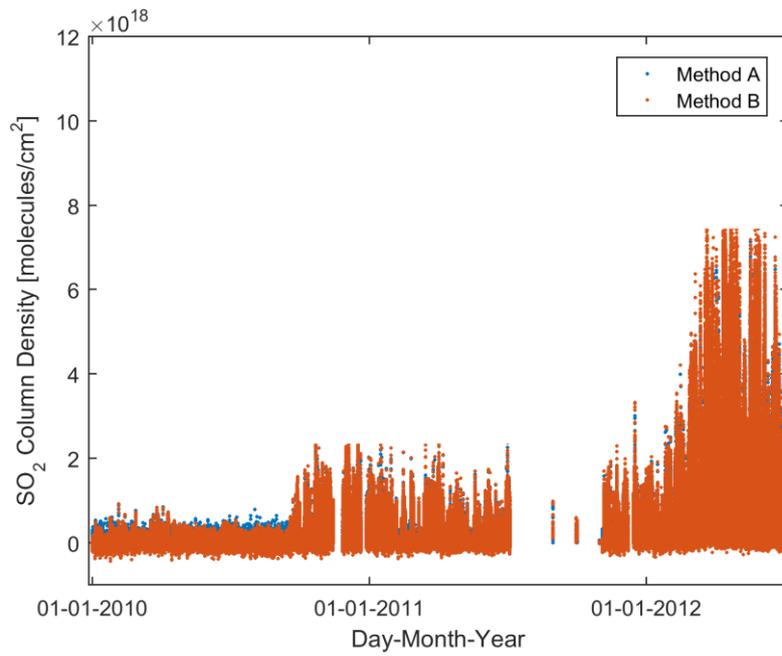


Fig. 2 Time-series of the SO₂ CDs for instrument D2J2201 at Nevado del Ruiz.

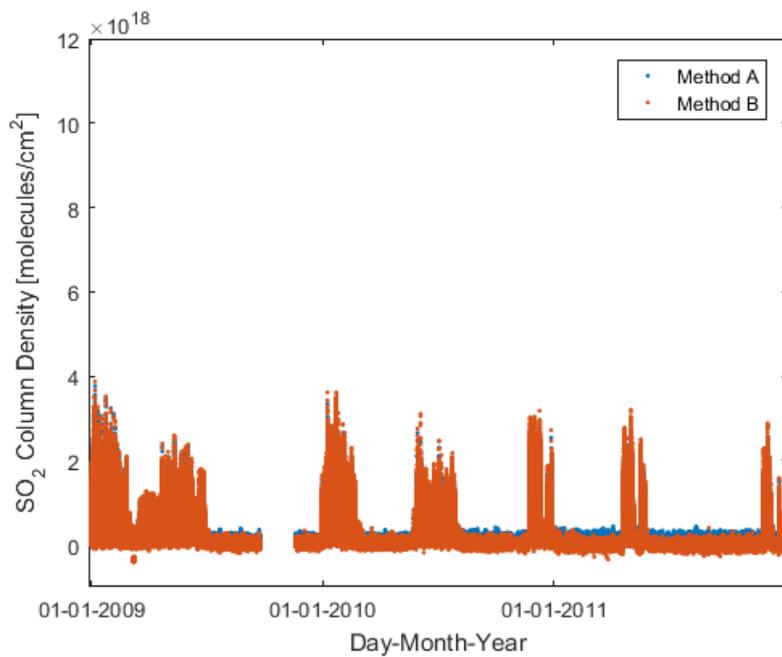


Fig. 3 Time-series of the SO₂ CDs for instrument D2J2140 at Tungurahua.

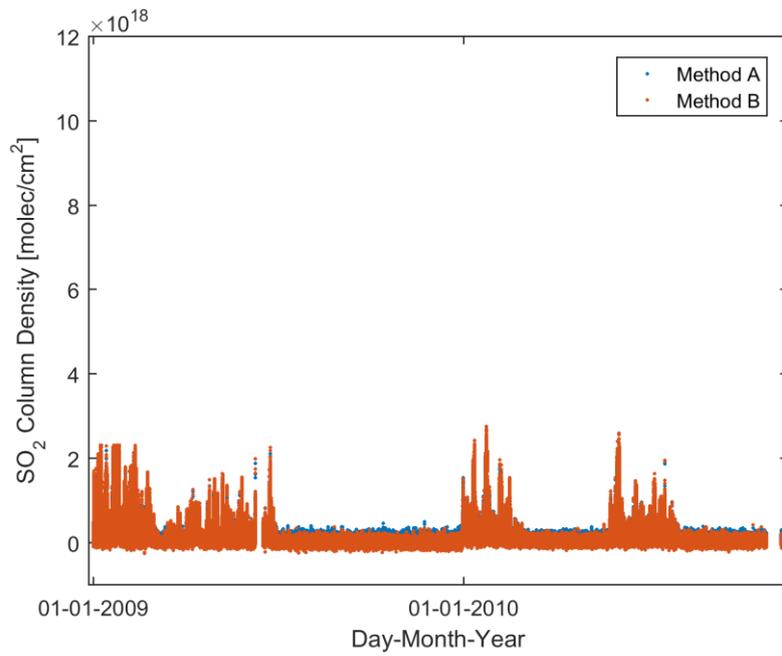


Fig. 4 Time-series of the SO₂ CDs for instrument I2J8546 at Tungurahua.

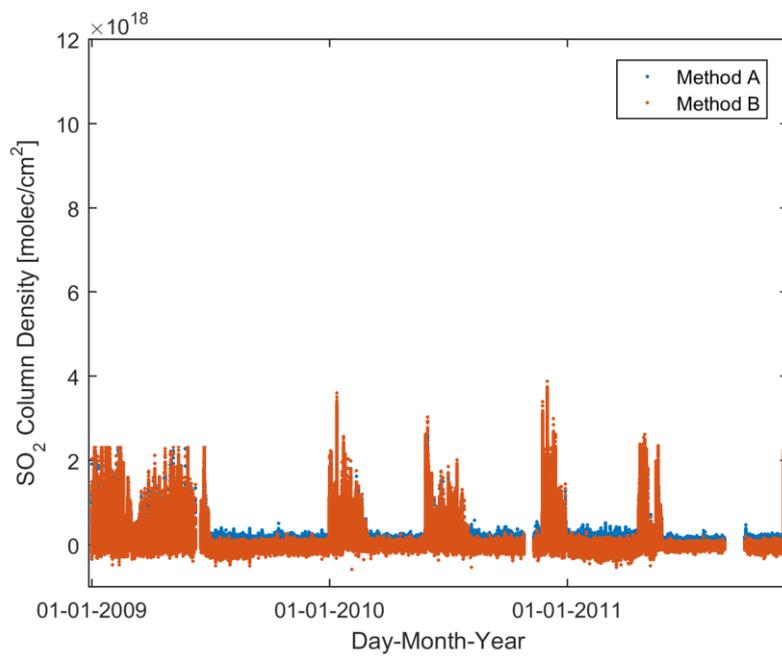


Fig. 5 Time-series of the SO₂ CDs for instrument I2J8548 at Tungurahua.