Answer to comments by referee #1

Retrieval interval mapping, a tool to visualize the spectral retrieval range in differential optical absorption spectroscopy

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Anonymous Referee #1:

Remote sensing via differential optical absorption spectroscopy (DOAS) has become a standard technique to identify and quantify trace gases in the atmosphere. Due to the wide range of measurement conditions, atmospheric compositions and instruments used, a specific challenge of a DOAS retrieval is to optimize the parameters for each specific case and particular trace gas of interest. For example, a well chosen evaluation wavelength range is crucial to the DOAS technique. Although for many trace gases the overall dependence of common DOAS retrieval on the evaluation wavelength interval is known, a systematic approach to find the optimal retrieval wavelength range and qualitative assessment is missing. Here the authors present a novel tool to determine the optimal evaluation wavelength range. It is based on mapping retrieved values in the retrieval wavelength space and thus visualizing the consequences of different choices of retrieval spectral ranges. This visualization allows one to readily identify retrieval wavelength intervals which are likely to lead to erroneous results, or might lead to greater variability of retrieved values due to a large gradient of the retrieval interval dependency. The technique is demonstrated using the examples of a theoretical study of BrO retrievals for zenith-sky stratospheric BrO measurements and for BrO measurements in volcanic plumes. For both examples, the effects of: (1) the I0-effect, (2) cross correlations between RCSs (reference absorption cross sections) and (3) added noise are examined using synthetic spectra. For zenith-sky DOAS, the tests confirmed the evaluation wavelength range 346-359 nm as suggested by Aliwell et al. (2002). This retrieval wavelength interval of BrO offers the least dependency on the I0-effect although an I0 correction of RCSs is still mandatory. BrO retrievals including lower wavelengths are not advised since strong O3 absorption and slightly insufficient I0 correction of RCSs may yield highly erroneous values even in this \best-case" scenario. BrO retrievals for measurement of volcanic plumes show a much lower I0 dependency of the BrO SCDs on the retrieval wavelength interval, mostly due to an about 100-times weaker total O3 SCDs. Whereas the fit applying I0 corrected RCSs shows a good agreement with the true BrO SCD at most wavelengths, misleading SCDs are retrieved for uncorrected RCSs. An evaluation wavelength interval with a lower wavelength limit between 320-335 nm is found to be optimal because here the differences are smallest between retrievals applying uncorrected and I0 corrected RCSs. In practice, the absorber strength at short wavelengths is most variable because of the great variability of volcanic SO2 emissions and O3 absorptions in early and late hours of the day. To ensure a comparable evaluation of data, measurements should be performed at the upper range of suggested interval.

However, a specific retrieval wavelength interval can not be recommended for the evaluation of BrO in volcanic plumes here. Advanced modelling of synthetic spectra (including realistic simulations of the atmospheric radiative transfer and the Ring effect) in comparison with measured spectra are needed in order to advise the specific retrieval wavelength interval to be chosen. At the example of synthetic spectra of passive DOAS measurements, the novel tool of retrieval wavelength mapping was introduced. However, the method is not limited to these types of instruments but can be applied to any DOAS measurement. Thus it enables a systematic study of important retrieval parameters, can highlight pitfalls in any DOAS retrieval and allows for an encompassing motivation of applied parameters.

Referee's recommendation

As outlined above, this paper describes and tests a new method of analyzing the quality of BrO DOAS spectral fits and may be applied to any type of DOAS measurement. It may be implemented using a PC with moderate specifications and shows great promise in helping one to determine the best fit range for a given trace gas, among other parameters. I recommend this discussion paper for full publication, once the following comments have been addressed satisfactorily and minor grammatical changes to both the text and figures/tables have been made.

Answer:

The authors would like to thank the referee for his/her efforts and kind comments. In the following, these comments will be addressed. It is important, however, to keep in mind that the authors aimed at presenting a novel tool and its application to certain simplified measurement scenarios. Therefore, our intention was not to present extensive studies including all imaginable effects.

Many of the here addressed questions originate from this gap between the synthetic spectra used as an example, best possible case, and measured data. Thus, this study can only act as a starting point, enabling the reader to optimize the evaluation of his/her data set depending on the respective choice of parameters.

Many questions remain unanswered, and one of the most pressing is the influence of the Ring-effect. So far the software tools are missing to calculate the Ring-effect in a 3 dimensional atmosphere, which is needed to model it for volcanic plumes. Also such a study would need to include measured data, preferably a comparison between different instruments and retrieval approaches. This would have exceeded the scope of this manuscript.

The authors will gladly assist in further studies (e.g., combining the theoretical approach with measured spectra) by providing existing algorithms and software. A supplement is added to the manuscript which includes all necessary files and description to perform Test 1 for both measurement scenarios.

In the following, the authors address the Referee's comments referencing page and line of the original AMTD publication.

Scientific Questions/Comments

Page 4200

Comment:

line 8: In the DOAS community the meaning of χ^2 is known, but maybe you should define it here briefly anyway?

Answer:

The authors abstained from a more detailed introduction of DOAS and its retrieval algorithms because the paper is specifically addressed to the DOAS community. In order to explain the χ^2 value in a meaningful way, it would be best to also introduce common DOAS fitting algorithms. This however would exceed the scope of the manuscript, especially since it is already quite lengthy.

With this in mind, the authors added:

"…(e.g. fit error, χ^2 value as a measure of the overall goodness of the fitting algorithm's results, possible shifts in wavelength calibration by the algorithm)…"

(P4200, L8)

Comment:

GENERAL POINT: The authors chose 2 case studies: 1 - stratospheric BrO and 2 - volcanic BrO. Why not an additional tropospheric case utilizing MAX-DOAS (ie. MAX-DOAS BrO Arctic measurement such as seen in Wagner 2007 or Hoenninger 2004b)?

Answer:

We aimed at presenting the novel tool together with as few examples as possible. In principle, it should be quite simple to study additional tropospheric cases. However, if all tests would be repeated for additional scenarios, and discussed properly, the manuscript would become even longer. Therefore, we restricted ourselves to only two scenarios.

Page 4203

Comment:

line 5: The Ring Effect is not considered. As you have stated, this may greatly affect results so why not create a synthetic Ring spectrum (as commonly done in MAX-DOAS retrievals) and include that when fitting your synthetic spectra, then compare these results with those without the Ring Effect correction? How were the Fraunhofer reference spectra determined and used in the fitting of the spectra?

Answer:

The Ring-Effect may arguably be one of the major sources of error in common DOAS evaluations of scattered sun light measurements. To fully discuss this issue, however, an extensive study including advanced radiative transfer modelling and comparison with measured data would be necessary. The authors focus in the here presented work on the novel visualization.

The approach to study the Ring-Effect as proposed by the referee was actually used in Vogel 2011, and the results differ from the simple scenarios presented here. To fully discuss this issue however, an extensive study including advanced radiative transfer modelling and comparison with measured data would be necessary. Up to date, the tools are missing to calculate the Ring effect in a 3 dimensional atmosphere in order to simulate the effect for volcanic plumes. Especially when high SCDs of trace gases and a condensed and/or ash laden plume are present also wavelength dependent optical paths need to be considered. Therefore the authors refrained from including the Ring effect in the study and restricted themselves to a presentation of the visualization tool.

The above reasoning was included in the manuscript by adding the sentences:

"For a correct assessment of the Ring effect however, an extensive study including advanced radiative transfer modelling and comparison with measured data would be necessary. Up to date, the tools are missing to calculate the Ring effect in a 3-dimensional atmosphere in order to simulate the effect for volcanic plumes. Furthermore, retrieval wavelength ranges yielding highly erroneous SCDs without considering the Ring effect will not yield true SCDs when the effect is included."

(P4203 L7)

"… and the Ring effect in a 3-dimensional atmosphere once the respective tools are created) … "

(P4220 L24)

As Fraunhofer reference spectra I_0 acted the Fraunhofer spectrum without any absorption structures (Kurucz, 2005), which was convolved to the instrument resolution. This was stated on page 4204, line 6. To clarify, a sentence was added at the end of line 7:

"The in this way constructed spectrum I_0 was used as a Fraunhofer reference spectrum in all evaluations."

(P. 4204, L6)

<u>Page 4204</u>

Comment:

line2: What is your rationale for the slit function chosen (ie. why was the resolution chosen to be 0.65 nm)? Why was the detector chosen to have 1024 pixels, don't most DOAS setups employ at least 2048 pixels nowadays? Are these choices important?

Answer:

We chose the respective settings since they reflect common spectrographs. Although detectors with 2048 might be used more frequently, 1024 are also incorporated into many instruments (e.g., Ocean Optics QE65000 spectrograph). An optical resolution of 0.65nm is common for instruments of 1024 and 2048 pixels. In fact, the presented method is independent of the number of pixels chosen. The resolution of 0.65nm at a dispersion of 0.1nm per pixel was chosen so that undersampling effects can be disregarded and the results can be transferred to instruments with a similar optical resolution and equal or higher number of pixels.

A half sentence was added at P4205 L5

"… possible under-sampling effects (Roscoe, Platt, Chance…), and the results can be transferred to spectrograph of similar optical resolution and equal or higher number of pixels."

(P4205, L5)

Comment:

line 11: You say that the optical density is greater than 0.1. Strictly speaking I believe that optical density is $\ln(I_0/I)$, so I would re-word this sentence, unless I have missed something.

Answer:

We agree with the Referee and rectified the typing error to

"The convolution also introduces an error for absorbers at high optical densities ($\ln(I_0/I) > 0.1$)."

(P4204, L11)

<u>Page 4207</u>

Comment:

line 8: You chose a second order polynomial. Is this typical for these types of BrO fits in the field? In the field there are more broad band extinctions to worry about right? Would this be something that you would have to worry about when applying this technique to measurements performed in the field?

Answer:

The applied polynomial degree to correct for broad band extinctions is an important parameter in a DOAS retrieval. Especially, since a polynomial of higher degree is usually necessary for a wider retrieval interval. A polynomial of the same degree might lead to erroneous retrieval results for smaller retrieval interval ranges, if the differential absorptions of trace gases can be mimicked by the polynomial. The presented tool could be easily applied to field measurements for a study of the influence of different degrees of the DOAS polynomial, and the authors encourage the user to do so.

To better highlight this valid point, the authors added a sentence at the end of line 10:

"If the presented tool is applied to field measurements however, the effect on the retrieval results by different degrees of DOAS polynomials should be studied as well."

(P4207, L8)

Page 4208

Comment:

line 15: How do you chose the assumed SCD value when calculating your I_0 corrected absorption cross sections? Will this make the I_0 correction less accurate at times? Is there anything you can do to limit this source of error in the field? It seems a difficult task when you don't have any idea of the true SCD value.

Answer:

For this study, always the original SCDs of all trace gases were used in the correction of the I_0 effect, i.e. 10^{19}molec/cm^2 for O3 even if O3 was varied by 1% in scenario zenith DOAS. The relative deviations in the corrected O3 RCSs are less than 5 10^{-5} (RCSs corrected with 1.01 10^{19}[molec/cm^2] / RCSs corrected with 10^{19}[molec/cm^2]). The error induced by only using a first order I_0 correction (see comment below) is dominating over the error by the incorrect assumption of respective trace gas SCDs in the I_0 correction.

Finding the correct SCDs for I0 correction of absorption cross sections for field measurements may indeed pose a problem. One way would be the use of an iterative algorithm on the respective trace gas SCDs. However, tests should be performed on the sensitivity of the retrieval to the a priori assumptions in this case. If a wavelength retrieval range can be identified by the here presented tool, which is least dependent on the I_0 effect, these errors will be diminished as well.

Comment:

Secondly, in your Equation 2 for the I0 correction you mention that the term $I_{k;0}$ in the denominator should consider all gases except the gas of interest. In the field you cannot correct this but you could for this study. Why not do so for a sensitivity study to examine the potential source of error due to this effect? Is it really a major source of error?

Answer:

The authors performed sensitivity tests with RCSs corrected in this manner, and the results are mentioned on P4218 L6. If RCSs are used in the fit which have been truly corrected for the I0 Effect, deviations for all retrieved trace gas SCDs over all retrieved wavelength ranges are negligible. This noteworthy point is now highlighted by adding two sentences at the end of Test I, P4207 L17:

"As mentioned in Sec. 3.1, a true I_0-correction can only be achieved if the denominator $I_{K,0}$ in Eq. (2) includes absorptions of all trace gases except the one which cross section is to be corrected. This was tested for both synthetic measurement spectra, and correct SCDs could be retrieved for all trace gases at all retrieval wavelength ranges."

For field measurements, iterative approaches may yield improved I0 corrections, but care should be taken not to induce other artefacts in the fit. Of course, this can only be achieved if significant computational power is available.

Comment:

line 20 Why were the 1% and 10% limits set? You refer to Table 2, but what is your rationale behind these choices (maybe a certain reference(s))?

Answer:

These choices are motivated by the different measurement conditions. Changes in the measured trace gas SCDs are mainly induced by changes in SZA for the zenith DOAS scenario, whereas for the volcanic plume scenario sudden changes in measured trace gases may occur by sudden changes in emission strength of the volcano. Of course different trace gas SCD may vary on a different scale, but for the sake of simplicity the same deviations were chosen for all trace gases in the respective measurement scenario. The 1% changes for the zenith DOAS scenario have been chosen, because they represent the mean deviation from the true O_3 SCDs observed in Aliwell et al. 2002, scenario B2 and C2 (see page 4, table 3), which correspond to the retrieval approach chosen in this publication.

The motivation of 10% deviation for the volcanic plume setup is that rapid changes of observed trace gas SCDs may occur in volcanic plumes, which may well exceed 10%. Trace gas SCDs in the ambient atmosphere may vary as well, but not to such an extent. For the sake of simplicity, the authors chose for 10% all trace gas variations representing mean variations off all trace gases in the scenario.

Therefore, the authors added the following text to manuscript starting at P4208 L20:

"These choices are motivated by the different measurement conditions:

For the zenith-sky DOAS, changes in the measured trace gas SCDs are mainly induced by changes in SZA. The 1% changes for the zenith DOAS scenario have been chosen, because they represent the mean deviation from the true O_3 SCDs observed in Aliwell et al. (2002), scenario B2 and C2 (see page 4, table 3), which correspond to the retrieval approach chosen in this publication. In case of the volcanic plume scenario, the motivation of 10% deviations of true trace gas SCDs is, that rapid changes of observed trace gas SCDs may occur due to changes in volcanic emission strength, which may well exceed 10%. If aerosols or ash are present in the plume, also trace gas SCDs of the ambient atmosphere may vary due to different optical light paths observed. Different trace gas SCDs may vary on a different scale, but for the sake of simplicity the same deviations were chosen for all trace gases in the respective measurement scenarios."

<u>Page 4212</u>

Comment:

lines 9-11: Why does a mistake in the I_0 correction of O3 make a difference when NO2 is varied in Figure 5???

Answer:

The residual structures induced in by the slightly incorrect I_0 correction of O3 are present in both measurement spectra with varying NO2 SCDs. Malicious features like these can affect the retrieval in unexpected ways, in this case leading to the observed cross correlations between RCSs. It is not unambiguously possible to assign the cross correlations to certain trace gases.

In order to clarify this point and regarding comments from Referee #2, the text in section 5.2, P4218 L22-23 was changed to:

"The only sources of error remaining are malicious spectral features induced by the insufficient I0-correction of RCSs. This hypothesis is strengthened by the observations for the zenith-DOAS measurement scenario (Fig. 5). The similarities of the structures observed for variations of O_3 and NO_2 SCDs indicate that the structures observed in the NO_2 plot may originate from the insufficient correction of the I0-effect of the O_3 RCSs."

(P4218, L23)

Page 4215

Comment:

lines 24-25: BrO shows the least dependence on changes in HCHO absorber strength. In the field you say this is different, and much higher anti-correlations are expected. Could you create a scenario that more closely models what is experienced in the field to determine a better BrO wavelength retrieval range for future studies? What is the suggested reason for this difference between your synthetic data and those in the field (perhaps comparing your results to the work done in (Vogel, 2011))?

Answer:

The construction of a scenario reflecting real measurement conditions is mandatory for a final recommendation on a suitable wavelength evaluation range.

In this work, it is shown that the I0 effect is a major source of error at the presence of strong absorbers (measurement scenario zenith DOAS). These strong absorptions are usually not present when measuring volcanic plumes in common retrieval ranges for BrO (wavelengths > 325nm).

However, when studying volcanic plumes, a Ring effect on the order of several percent optical density is commonly encountered. If not corrected properly, the remaining residual structures may interfere with the retrieval of other trace gases (here BrO, HCHO). In Vogel, 2011, the Ring effect was treated as an additional absorber, which represented a more realistic approach but did not capture all the variations observed in field measurements. As mentioned previously, this approach may not simulate reality if a condensed and/or ash-laden plume is observed (for example the additional wavelength dependence of the optical path). The introduced residual structures lead to a cross correlations between HCHO and BrO, which were partly observed in the measured data as well. Since the software tools are missing to simulate a 3 dimensional condensed volcanic plume together with the Ring effect, further efforts are necessary.

General Questions

Comment:

(1)Why was the synthetic BrO SCD set to 1.5 10^{14} molec cm^{-2}? Is this near or at the detection limit or simply a common (average) retrieved value?

Answer:

A BrO SCD of 1.5 10^{14} molec/cm^2 was chosen because it is a common SCD measured for both zenith DOAS (see Aliwell et al.) and may also be measured in volcanic plumes. (see P4205 L 13 and P4206 L9)

Comment:

(2)Throughout the paper you often refer to (Aliwell, 2002) as your bench mark BrO study? What is your reasoning? Is it simply because this is known as the most comprehensive study to date and is confirmed by Theys, 2007?

Answer:

It is correct that the study by Aliwell et al., 2002 is a very comprehensive study. It is not the intension of the authors to repeat or validate these previous efforts. The zenith DOAS scenario was chosen to demonstrate the abilities of the presented tool at the example of a known study to allow easy comparison.

Comment:

(3)In this paper you use synthetic spectra but you know that spectra in the field behave differently and other effects need to be considered (ie. Ring effect). How can/will you modify your technique to make your determinations close to the “real-world" conditions in the future? Are you planning on including advanced modelling of synthetic spectra including radiative transfer effects and the Ring effect in the future?

Answer:

The authors would like to distinguish between the novel tool itself and its possible application. In general, the tool can be easily applied to any DOAS measurement and thus give a quick and comprehensive overview over possible pitfalls in DOAS evaluations. The tool in itself is ready to be used by anyone, and the authors gladly supply the respective software.

The application to synthetic spectra, however, strongly depends on the way they are generated. Here, a straightforward approach was chosen. For a more advanced generation of synthetic spectra the authors would like to direct the interested reader to Vogel 2011. However, for a truly realistic modelling of volcanic plumes, efforts need to be taken to include a correct modelling of the ring effect in present 3-D radiative transport modelling algorithms, especially as for volcanic plumes a 3 dimensional modelling of the atmosphere is necessary.

Thus the authors primarily wanted to introduce the presented tool to the DOAS community at this point. Further work with more advanced generated synthetic spectra is ongoing.

A respective clarification has been added at P4203 L7, see also answers to comments of Referee #2.

Technical Corrections

The authors incorporated the suggested changes and would like to thank the reviewer for his/her consequent proof reading.

The following lists grammatical suggestions and technical corrections to the text, listed by page and line number:

Page 4196

line 5: remove comma (, respectively.)

Done

line 6: change ground based to ground-based (be consistent throughout the text)
Done

line 6: change trace-gases to trace gases

Done

lines 14-17: cut ALSO and add later. start sentence with Instrumental limitations.....ALSO need to be taken into account.

Done

line 15: change wavelength depending to wavelength dependent

Done

line 15: change sources of errors to sources of error

Done

line 20: replace find with finding

Done

line 21: here do you mean qualitatively or QUANTITATIVELY?? Changed to quantitative

line 23: change visualize to visualizing

Done

lines 23-24: change consequence to consequences

Done

line 24: cut e.g.

Done

GENERAL POINT: the authors tend to use e.g. frequently throughout the text, try to limit it when not entirely necessary since it helps the text flow better

line 24: change text....of different choices of spectral retrieval ranges...

Done

line 26: cut the ... demonstrated using examples of a .....

Done

line 27: cut FOR ... stratospheric BrO and BrO measurements in volcanic....

Done

line 27: optional in brackets ...stratospheric BrO (measurements) and ... I would cut it since it not necessary to state it twice in the same sentence

Done

Page 4197

line 2: revise as ....of DOAS retrieval (active or passive), or alternatively as ... of (active or passive) DOAS retrieval.

Done

line 6: change VIS to Vis

Done

lines 7-8: change -law to -Law

Done

line 13: change a trace gases to various gases

Done

line 15: change e.g. to such as

Done

line 18: change amount to amounts

Done

line 25: add the ...lead to small errors in THE determination of the strong...

Done

Page 4198

line 1: repetition of word DOAS not required, I would cut it in the following passage...active and passive (DOAS) instruments

Done

line 2: ...artificial OR natural light sources...

Done

line 3: remove comma (,respectively)

Done

line 3: change star light to starlight

Done

line 3: GENERAL POINT: throughout the text the authors often omit a word (e.g. one or us) when phrasing a sentence like this: Active DOAS instruments allow to compare spectra.... it should be something like: Active DOAS instruments allow ONE to compare spectra....

Done

line 4: replace passes with passed

Done

line 6: ...due to the necessity of THE stable deployment of A light source (or A...THE)

Done

line 7: ...as well AS

Done

line 11: ...without THE necessities OF an additional emitter and reflectors.

Done

line 13: change ground based to ground-based

Done

line 14: Are car traverses using MAX-DOAS? If so, include it in the brackets ...balloon, ships and car traverses)??

Car traverses of volcanic plumes are usually performed using a fixed telescope looking to zenith. Since other configurations or multiple telescopes with different viewing angles are possible, “car traverses” has been moved inside the brackets.

line 15: Why is this a separate paragraph? I would include it at the end of the last paragraph??

Done

line 15: ...overview OF different DOAS...

Done

line 22: change interference to interferences "interference "

It has been replaced by “cross correlations” throughout the manuscript (see also comments of the second reviewer).

line 23: I suggest replacing involves with introduces

Done

line 23: ...incorporating THE strong absorption...

Done

line 25: cut e.g.

Done

line 27: ...IN the presence of higher aerosol...

Done

line 27: You did not state in implicitly in your text, but I assume that you did not consider cloud effects in this paper??

Correct, cloud effects were not studied

Page 4199

line 1: change RCS to RCSs. Moreover, the retrieval....

Done

line 4: ...the problem only A few attempts...

Done

line 4: Can you provide some references for these attempts here or maybe just refer to Table1?

To prevent repetition, the authors would like to keep the reference to Table 1.

line 6: ..the retrieval wavelength ranges APPLIED are only motivated by a SINGLE comparison TO ONE other retrieval wavelength range.

Done

line 9: This is illustrated IN..

Done

line 10: change which to THAT... (BrO) that have been used in the past. (just so you are not repeating which in the same sentence)

Done

line 13: ...one wonders whether A total of ....

Done

line 16: ...systematic studies ON the retrieval...

Done

line 18: replace ground based with ground-based

Done

line 22: ... only two, relatively weak, ...

Done

lines 24-25: The results of this study (Do you mean Theys et al. 2007 or Aliwell et al. 2002? I think you mean Aliwell et al., right?)

Since both studies yield the same results, “this study” was changed to “these studies”

line 25: replace different with ANY

Done

lines 26-27: ...time difference between THE measurement of THE...

Done

line 28: Again, do you have any references or maybe refer to Table 1??

Added : (for NO_2 e.g. Roscoe et al. 2010)

Page 4200

line 3: ... we introduce a novel tool THAT is suitable FOR systematically and quantitatively studyING the ....

Done

lines 10-11: I would re-word this sentence as follows: ....of THE results for a large set of evaluation wavelength ranges and offerS an intuitive tool FOR showING how certain....

Done

line 22: cut (the subsequent)

Done

line 23: ..concluding remarks in Sect. 6.

Done

GENERAL POINT: At times there is a overload of bracketed material. It often makes the text harder to read. I have made suggestions such as in line 23 to limit the use of brackets in your text for this reason.

Done

Page 4201

line 7: ...and allows US to study...

inserted “…one…”

line 8: replace E.g., with For example, or For instance, ... interval dependency of:
Removed “E.g.,“

line 13: THE influence of instrumental...
Done

line 14: THE dependency of ...
Done

line 15: I0-effect and Ring effect (keep spelling constant)...passive DOAS...
Done

line 23: The PLOTTED results are ...
Done

line 23-24: either use colour-coded or colour coded (keep spelling constant)
Done

line 25: The resulting maps allow ONE to ...
Done

Page 4202
line 1: ..to A greater variability of THE retrieved values...
Done

line 11: no comma needed (, respectively)
Done

line 13: ... is apparent. In a first ....
Done

line 14: ... they yield results THAT DEVIATE quite strongly from the true value,
WHILE others....
Done

line 19: THE computational...

Done

line 20: All individual maps SHOWN ...

Done

line 21: ... within 30 minS for THE given wavelength evaluation ranges....

Done

line 22: ... this novel tool allows ONE to give a ...

Done

line 24: IN THE TITLE -recommend changing at to FOR line 25: ...of the method described above...

Done

line 26: ...stratospheric BrO...

Done

Page 4203

line 3: ...allowing ONE to study...

Done

line 4: ...neglected. The most important of these is the Ring Effect...

Done

line 10: ...This IMPOSES certain limits on extending (applying) the results from the synthetic measurements studied to spectra measured in the field. (Is this what you mean??) I would re-word the sentence as above or something to that effect.

Done

line 11: The prime focus OF this study..

Done

line 17: replace RCS with RCSs

Done

line 18: sunlight

Done

line 23: ...multiplied BY the respective
Done

line 25: ..according to THE Lambert-Beer Law:
Done

Page 4204
line 4: ...This allows ONE to ...
Done

line 6... cut out THUS
Done

line 12: ..Lambert-Beer Law...
Done

line 22: I0-effect
Done

line 26: ...effort IF applied to ...
Done

Page 4205
line 3: ground-based
Done

line 6: possibly cut e.g. Is it needed?
Done

line 8: ...are the strong O3 absorption structures ENCOUNTERED at ...
Done

lines 10-11: change to balloon and satellite (no capitals required) ... in THE limb-viewing direction...

Done

line 11: RCSs

Done

line 15: ... AT higher resolution...

Done

line 20: Since BrO's optical density...

Done

line 23: ..seen in results presented later ...

Done

Page 4206

line 2: The Fraunhofer REFERENCE spectrum...

Done

lines 3-4: I understand what you mean but I would re-phrase this sentence to make it more clear, something like this: ...and it is assumed to be taken in close proximity in time with measurements at the lower elevation angles, and thus...

Done

line 7: cut comma (, both)

Done

line 10: cut e.g.

Done

line 12: cut e.g.

Done

line 19: Possible....HCHO must ALSO be taken into account due to THE similar..

Done

line 20: ...and optical density ENCOUNTERED.
Done

line 22: ..to allow FOR...
Done

lines 24-25: A set of .....ARE APPLIED.
Done

Page 4207
line 1: replace fitted with FIT
Done

line 2: add a comma: ... , instead...
Done

line 3: wavelength-pixel mapping
Done

line 4: between THE Fraunhofer reference
Done

line 5: cut e.g.,
Done

line 7: THE stability of...
Done

line 8: RCSs ...spectra
Done

line 14: WITH which..
Done

lines 15-16: This test ALSO..
Done

line 19: Beyond (STUDYING)..
Done

line 21: ... to small variations IN the amount of ...
Done

line 24: replace ...are depending... with DEPEND
Done

Page 4208
line 1: ...cross correlations. The original sets of...
Done

line 2: replace fitted with FIT
Done

line 3: ..of THE retrieved...
Done

line 4: ...expressed as:
Done

line 9: ...range (RELATIVE) to changes...
Done

line 10: ...estimated by:
Done

line 15: ...are resulting from: (1)....
Done

Page 4209

line 5: ...to an optical density of 3 10^4

Done

line 7: you should not start a sentence with a number... Reword (for example)...Iterations of 10 and 50 correspond...

Done

line 9: For one BrO absorption band with an average FWHM of 1.5 nm....

Done

line 12: remove comma (, respectively)

Done

line 15: RCSs

Done

line 20: ..scenarios OF zenith..

Done

Page 4210

line 4: ...In THE case...

Done

line 9: replace an with A (...A lower limit of ...)

Done

line 10: ...can be MAINLY attributed TO A slightly ERRONEOUS...

Done

line 19: ...the results DEPEND mostly on...

Done

line 19: This AGAIN is ...

Done

line 20: ... OF the inuence ...

Done

line 21: replace (are decreasing) with DECREASE

Done

line 22: ...CHANGE WITH THE retrieval wavelength interval INCLUDING THE BrO..

Done

line 24: (cut the example of) and replace with: ...and residuals at three different wavelength ranges...

Done

line 25: cut (both)

Done

Page 4211

line 1: ...and THE use of ...

Done

line 4: replace than with ASIDE FROM

Done

line 6: replace test with TESTS ... Aliwell et al. (2002), ...

Done

line 7: ...both in the retrieved BrO SCDS and shapeS of THE residual.

Done

line 12: .. ,otherwise the retrieval...

Done

line 20: ...WITH a lower limit...

Done

line 28: The differences SHOWN ...

Done

Page 4212

line 1: replace are resulting with RESULT

Done

line 3: ..cross correlation of absorption cross sectionS..

Done

line 4: ...varying THE NO2 SCD ...

Done

line 11: ...Indications OF THIS...

Done

line 28: ...regardless OF WHETHER the RCSs are corrected for the I0-effect OR NOT.

Done

Page 4213

line 4: ...spectra exhibit wavelength DEPENDENT structures...

Done

line 5: .. systematic wavelength DEPENDENT...

Done

line 6: (more the word also) ... are ALSO more apparent ...

Done

Page 4214

line 17: cut whereas

Done

line 26: ... to THE different absorbers THAT interfere the most with the BrO....
Done

Page 4215
line 1: cut ,e.g., It shows that the observed....
Done

line 2: ..between THE RCSs ...
Done

line 15: deviationS
Done

line 19: I think you are referring to Fig.10. here not Fig. 11.?
Done

Page 4216
line 7: THE area...
Done

line 8: ..between THE lower limits of...
Done

Page 4217
line 1: ...THE amplitude..
line 25: .. the zenith-sky DOAS SCENARIO).
Done

Page 4218
line 19: ... wavelength DEPENDENT..

Done

line 23: ..the RCSs...

Done

line 24: change depending to DEPENDENT

Done

line 25: cut , e.g.,

Done

line 26: DEPENDENT

Done

line 27: Ring Effect (be consistent)

Done

line 27: .. in THE case of passive measurements. ONE SUCH example IS THE cross correlations...

Done

Page 4219

line 7: ... these findings ALSO remained true...

Done

line 13: change ..are present to WERE present

Done

line 16: ... appear to DEPEND on ... (or to be dependent on)

Done

line 16: replace by the with DUE TO THE

Done

Page 4220

line 7: ... THE least dependency..

Done

line 13: I would replace about with OR rephase as: due to a total O3 SCD that is about 100 times weaker.

Done

line 25: ...order to advise ONE ON choosing a specific retrieval...

Done

Page 4221

lines 4-6: (e.g. instrumental features...radiative transfer.) or replace e.g. with SUCH AS,

Done

line 6: Previous publications WERE motivated by the COMPARISON of the applied retrieval wavelength interval WITH typically one ....

Done

line 15: THE best values of THE resulting SCDs.

The Authors replaced the sentence with *“Furthermore, the method is not limited by studying the dependency of the retrieved SCDs on the evaluation wavelength intervals.”*

line 16: replace e.g. with SUCH AS, ...THE effect...

Done

line 17: ...errors in THE I0 correction, ...

Done

line 22: replace where as with WHEREAS

Done

line 25: ..I0-effect...

Done

Page 4222

line 6: cut E.g. and use For example, for certain measurement...

Done

line 8: reference spectra

Done

line 9: nighttime

Done

line 11: FOR the example...

Done

line 19-20: ....corrected and uncorrected RCSs ARE DISCUSSED.

Done

Page 4223

line 2: why do you say white colour code? I would cut out white since other colours are present in deviations up to 1%?

Apologies, to clarifiy the sentence it was changed to:

“Their respective true value in the range of ±1% is denoted in white. Deviations will influence the retrieval as a whole.”

line 13: ...errors due TO NO2 BECOME more pronounced...

Done

line 14 ..increasES AND an overestimation of SCD occurs (see Fig. 13).

Done

line 17: cut thus

Done

line 25: replace at with UNDER

Done

Comments for the Figures

Fig. 1. CAPTION...Deviations from the true BrO SCD are displayed on A logarithmic colour-coded scale.

Done

Fig. 2. CAPTION: remove comma (, respectively)........WITH AN amplitude of 3 10^4.

Done

Fig. 3. CAPTION: ...for THE measurement scenario.......with THE uncorrected RCSs......

Done

Fig. 4. FIGURE: On the left hand side of the figure for case 9 you have the range listed as 345-359 nm, I believe this should read 346-359 nm, according to your text?? Absolutely, the depicted retrieval was performed from 346-359nm, done.

Fig. 4. CAPTION: Shown ABOVE ... The fit of THE BrO RCS and the residuum ....Regardless OF which...

Done

Fig.5. CAPTION: ...zenith-sky DOAS scenario. Changes in ......the strong O3 absorptions are CLEARLY dominating...

Done

Fig. 6. CAPTION: ...on THE zenith-sky DOAS measurement scenario....on THE noise applied...THE second till fourth column depict..and calculated correction factorS USED to retrieve..

Done

Not sure about your wording for the correction factor here? Do you mean: ....and calculated correction factor (defined as the standard deviation divided by the fit error)....

The sentence was changed as suggested.

Fig. 9. CAPTION: Fit examples FOR selected...

Done

Fig. 9. FIGURE: On the left hand side of the figure for case 9 you again have the range of 345-359 nm, I believe this should read 346-359 nm??

Absolutely, the depicted retrieval was performed from 346-359nm, done.

Fig. 13. CAPTION: ...from the true SCDs of trace gases OTHER than BrO in test I of THE measurement scenario OF volcanic plumes...

Done

Comments for the Tables

Table 2. CAPTION:replace Zenith with zenith.

Done

Table 3. CAPTION: ...absorption CROSS SECTION multiplied BY the respective SCD...

Done