Title: Improved retrieval of SO_2 from Ozone Monitoring Instrument: residual analysis and data noise correction

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We thank the reviewer #1 for carefully reading the manuscript and for providing constructive comments. All the comments are taken into account in the revised manuscript. We believe that in dealing with the comments put forward by the referee #1, the clarity of the manuscript will be greatly improved. We have attempted to address the comments below.

1. **Reviewer 1:** The paper needs significant copy editing to improve on the English – in some parts, it is really difficult to understand what the authors have done.

   Authors: We agree with this comment. We will take all necessary measures to improve English in the manuscript. Before final submission, we will send the text to professional organization for improving the English writing.

2. **Reviewer 1:** The methods used and the thresholds and criteria applied need to be described much clearer and more details need to be given. Some examples are

   - p 983, l8: "because of the invalid values…" – what are invalid values?
   - p 983, l9: "the nearest neighbour interpolation method" – is that linear interpolation from the neighbouring wavelengths or what is meant here?
   - p 985, l27: "when the selected pixels are near the terminator, we decrease the residual correction area" – what is "near the terminator" – 85° SZA? What is "decrease the residual correction area" – by how much?
   - p 985, l28: "bad pixels determined by residuals are filtered" – what are bad pixels, and how are they determined?
   - The entire discussion of the residuals for different latitude windows is difficult to follow and needs to be rewritten.

Authors: The text was amended as follows:

- p.983, l.8. The invalid value is the value with the solar irradiance value less than 0. We revised this sentence: “Because of the invalid value (solar irradiance value less than 0) in the solar irradiance data, the SO_2 column amount distribution shows a clear along-track stripe error (Fig. 2a–b).”.
- p.983, l.9. The nearest neighbour interpolation method is the linear interpolation from the neighbouring wavelengths. We revised this sentence: “We used the valid values from the neighbouring wavelengths to remove the invalid values in the solar irradiance data by the linear interpolation, and the clear stripe biases can be removed, as shown in Fig. 2c and Fig. 2d.”.
- p.985, l.27. Near the terminator is the area where the latitude is larger than 85°. “Decrease the residual correction area” means that near the terminator, because of no enough pixels covering 10° latitude area, we decrease the residual correction area to ensure the roughly equal pixels on either side of the selected pixels. Thus, near the terminator, the decrease of the residual correction area is not constant, changes with the latitude. We revised this sentence: “When the selected pixels for correction are near the terminator (latitude >85°), we decrease the residual correction area to ensure the roughly equal pixels on either side of the selected pixels.”.

  Please note that when the observations are near the terminator, the radiance measurements from the satellite contain larger error compared to normal viewing conditions. Consequently retrievals near the terminator are in general not useful.
- p.985, l.28. Bad pixel is defined as the pixel with residuals less than -150 at four wavelengths (310.8, 311.9, 313.2, and 314.4 nm). We revised this sentence: “Bad pixels with residuals less than -150 at four wavelengths (310.8, 311.9, 313.2, and 314.4 nm) are filtered before median residual value calculation.”.

- p.984, l.28. We agree with the reviewer that the discussion of the residuals for different latitude windows is difficult to follow. The text was revised for clarity. Note that this text will be sent to professional organization for improving the English writing. Thus, for the present, the text was revised as follows (in red font):

  By selecting one oceanic area with low SO_2 emission (0–30 °N, 140–170 °E), the comparison analyses of the different residual correction area effects were conducted on 16 January 2008 and 12 January 2009. We took the first wavelength pair as an example and selected the pixels with SO_2 slant column less than 2 DU for median residual. Figure 4 shows that on 16 January 2008, the median residuals from a sliding group pixels covering 20° and 10° latitude are consistent with those covering 30° latitude in the cross-track direction; meanwhile, the median residuals covering 5° latitude may be not sufficient to include the background errors for residual correction. However, from the residual analysis on 12 January 2009, the median residuals covering 30° latitude have an abnormally low value in the 40th pixel position at the cross-track direction, which largely deviates from the uncorrected residual at P1 wavelength pairs (Fig. 5). With such abnormally low median for residual correction, the SO_2 columns yields clear stripe biases on 12 January 2009 (Fig. 3). The median residuals from 20° latitude area also have a low value in the 40th pixel position, whereas 10° latitude median value did not record a low value. Therefore, the median value from 10° latitude residual correction area has a relatively better residual correction effect.

3. **Reviewer 1:** I'm not really sure I fully understood the method, but my understanding is that the main difference to the operational product is just the use of a 10° latitude band for the median background instead of the operationally used 30°. If that's the case, this is a small change and it should be stated clearly that not a new method was developed but rather one parameter of the existing method was tuned.

Authors: For clarity, the inappropriate words in the text were revised. For example:

- "a new optimization method" was revised as "an improved scheme for current BRD algorithm".
- "the modified algorithm results" was revised as "the optimized results".

4. **Reviewer 1:** The reason for using a large latitude window in the first place is that a small latitude window has the risk of removing not only artefacts but also reducing the high SO_2 values resulting from volcanic eruptions or pollution. This turns out to be the case as can be clearly seen in the comparison of the operational and the new data set – the latter is smaller by 20 – 40 % for the 2008 cases and also clearly lower for the 2009 examples. This is a serious problem and has to be discussed in detail. It would also be very useful to have the 1:1 line in the correlation plots as then the change in absolute values will be apparent.

   Authors: Yes, this comment is very valuable. We agree with the reviewer that a small latitude window has the risk of removing not only artefacts but also reducing the high SO_2 value resulting from volcanic eruptions or pollution. This can be seen from Fig. 6, Fig.7 and Fig. 8. The discussion about this underestimation was added at the end of section 3 in the revised manuscript. Notice that all the current SO_2 retrieval algorithms just enable the relative SO_2 distribution, because the precise SO_2 column amount is difficult to obtain by satellite technology. The relative spatial distribution of the optimized results is consistent with OMI level 2 SO_2 PBL product. The added discussion at the end of section 3 in the revised manuscript is as follows (in red font):

   Although the modified BRD SO_2 algorithm results are consistent with the OMI SO_2 PBL product in January 2008, having similar crest and trough variation (Fig. 6), the median residuals covering 10° latitude causes slight underestimation of SO_2 column amount in our optimized results in January 2008 in China. The extent of underestimation increases with rising SO_2 column. As shown in Fig. 6, compared with OMI SO_2 PBL product, the optimized results with SO_2 column larger than 10 DU are smaller by 10–40% in the cross-track direction. The underestimation can be also seen in Fig. 8. The slopes of regression lines are larger than 1, which prove the underestimation in January 2008. This underestimation is mainly related with high SO_2 emissions in China. When the selected area decreases from 30° latitude to 10° latitude in China, a higher proportion of pixels with high SO_2 columns is included for median residual; thus, increasing median residual results in the underestimation of SO_2 column amount. We recognize that the precise SO_2 vertical column is important, and our improved scheme may actually affect the absolute value of retrieved SO_2 column. However, the actual background errors in the residuals are dynamic over the world, and difficult to obtain. Mathematical methods for background errors correction may affect the retrieved SO_2 column to different degrees. Current SO_2 retrievals enable the relative SO_2 distribution, but not the absolute SO_2 results. Therefore, our improved method can be considered as an effective scheme for background errors correction.

Please note that the above added discussion will be further modified by the professional organization for improving the English writing.

The 1:1 line in the correlation plots was added. In addition, in order to present the correlation plots more clearly, we separated these correlation plots from Fig. 7, and add one new figure for these correlation plots in the revised manuscript. The new figure is named as Fig. 8 in the revised manuscript.

5. **Reviewer 1:** The discussion of other error sources both with respect to irradiance and radiance errors as well as in the conclusions chapter is very superficial and in my opinion does not go beyond what other studies have already reported earlier (and more clearly).

Authors: Thanks for pointing this out. We agree with the reviewer that this article does not include detailed error analysis. The discussion about the irradiance and radiance errors in section 2 was expanded. Two references were added to the revised manuscript. The corresponding text is edited as follows:

- p.983, lines 4–17. The text is revised as follows (in red font):

  OMI uses the sun-synchronous polar orbit with a daily global coverage; a solar irradiance corresponds to about 14 different earth radiances each day. Because of the invalid value (solar irradiance value less than 0) in the solar irradiance data, the SO_2 column amount distribution shows a clear along-track stripe error (Fig. 2a–b). We used the valid values from the neighbouring wavelengths to remove the invalid values in the solar irradiance data by the linear interpolation, and the clear stripe biases can be removed, as shown in Fig. 2c and Fig. 2d. However, after such reprocessing, the distribution of SO_2 column amount in Fig. 2c–d remains to have along-track biases. Different orbits in one day show the coincident stripes distribution, similarly performing at some certain viewing angles, as shown in Fig. 2c and Fig. 2d. Considering the OMI observation mode, these SO_2 along-track stripes can be mainly attributed to the solar irradiance noises, which is affected by dark signal, diffuser features, and signal noise (Veihelmann and Kleipool, 2006). The dark signal, caused by particle hitting on the detector, can influence the intensity measurement of each pixel of the detector array. The solar irradiance measured via a quartz volume diffuser (QVD) depends on the viewing angle, the goniometry (solar incident azimuth and elevation angles of the sun), and the column (wavelength) of the detector array (Dobber et al., 2008). The errors caused by the viewing angle dependence of the irradiance increase the stripe biases of SO_2 columns. The derivation of calibration parameters for the irradiance goniometry was based on the assumption that the changes in the sensor irradiance response over the course of the preceding years after launch were mainly attributed to the goniometry variations (Dobber et al., 2008). However, as the irradiance radiometric degradation increases, the inaccuracies caused by the irradiance radiometric degradation exceed those caused by irradiance goniometry, which make the assumption for irradiance goniometry calibration unstable. Thus, more noises are introduced into the irradiance data for serious irradiance radiometric degradation. The small wavelength dependence of irradiance data and signal noise are also the potential causes for stripes. These noises in solar irradiance are not constant; rather, they change over time.

- p.983, lines 18–26. The text is revised as follows (in red font):

Apart from solar irradiance noises, the earth radiance noises can also introduce cross-track biases into the SO_2 column amount results. The earth radiance data depends on the wavelength and the viewing angle. The viewing angle dependence causes the decrease of the earth radiance data at far off-nadir viewing angles, which can have a subtle but significant effect on the OMI-retrieved SO_2 results (Dobber et al., 2008). The absolute earth radiance cannot be calibrated because no accurate standards are readily available. In addition, the primary telescope mirror, which is used by the earth radiance observation mode, is subject to the optical degradation. Jaross has showed that no long-term degradation was observed in the earth radiance data from launch until about launch+3 years (from October 2004 to June 2007) (Jaross and Warner, 2008). However, as OMI sensor ages, the earth radiance data begin to be affected by the so-called row anomaly since June 2007. This anomaly affects the quality of the OMI science data for specific OMI cross-track positions (each cross-track position corresponds with a certain viewing angle). The row anomalies have four distinct errors on the OMI radiance spectra: a multiplicative error by blockage effect that decreases earth radiance; a wavelength shift error; a stray earthlight related additive error that increases earth radiance; and a stray sunlight related additive error that increases earth radiance (http://www.knmi.nl/omi/research/product/rowanomaly-background.php). These effects are dynamic, which means that the row anomalies vary with time.

In the conclusions chapter, we made a concise discussion about the other errors introduced by SO_2 retrieval algorithm model. Yes, this discussion is short. For this article, the objective of our article is to address the clear stripes biases in OMI level SO_2 PBL product since 2009. These obvious biases are mainly caused by solar irradiance and earth radiance noises, not by retrieval algorithm model. The description about other error in the conclusions chapter is used to present current other potential error in SO_2 column amount retrieval. We plan to do a much more detailed study on the errors analysis of SO_2 retrieval algorithm model. In addition, for clarity, we revised the conclusions chapter as follows:

- p.989, lines 4–19. The text is revised as follows (in red font):

  In addition to the measurement error, the errors caused by the retrieval algorithm model, which were not discussed in this study, can also bring about more complex biases in the retrieved SO_2 columns. Among current retrieval algorithms, the complex atmosphere conditions are simulated by a simple mathematics and physics model. However, the parameters that affect the radiative transfer process are complicated and variable, making a simple linear physical mathematical model difficult to use in simulating the nonlinear radiative transfer process. SO_2 inversion results can be distorted by O_3, surface albedo, aerosols, and vertical temperature profiles, etc. The linear algorithm model can result in SO_2 column underestimation. Therefore, the quantitative retrieval of SO_2 column amount, especially near-surface SO_2 concentration, needs further study in the future.

6. **Reviewer 1:** The figures are very small and difficult to read at least in the printout.

   Authors: Yes, the figures are really small and hard to read. Thanks for pointing this out. Fig. 6 was enlarged in the revised manuscript. In order to display more clearly, the correlation plots were separated from Fig. 7, and one new figure named as Fig. 8 was added in the revised manuscript.

7. Other changes: In addition to the above changes, we have also made some additions and updates to the paper to improve clarity. These changes will not influence the content and framework of the paper. And here we list the primary changes in the revised manuscript. Please see the revised manuscript for the detailed changes.

- One paragraph was added to the revised manuscript. The aim of this paragraph is to describe the structure of this article. The added paragraph is as follows (in red font):

  The paper is organized as follows: in section 2.1, the main noise in solar irradiance and earth radiance data is presented. In section 2.2, the different residual correction area is discussed, and an improved scheme for data biases correction is described. In section 3, the optimized results are compared with OMI level 2 SO_2 PBL product. In section 4, we summarize our findings.

- For clarity, we divided the section 2 into two sub-section, section 2.1 and section 2.2.

- p.986, lines 4–18. For clarity, the structure of this paragraph was rearranged. These changes will not influence the content of the manuscript.

- p.986, lines 19–25. This paragraph was revised and added to the end of section 3 for the underestimation discussion of our optimized results.

- p.988, lines 18–27. p.989, lines 1–3. After serious consideration, we think that it is more appropriate to delete these sentences. Because these sentences have relatively weak correlation with the objective of our article.

- Three references were added in the revised manuscript. They are as follows (in red font):

  Dobber, M., Kleipool, Q., Dirksen, R., Levelt, P., Jaross, G., Taylor, S., Kelly, T., Flynn, L., Leppelmeier, G., and Rozemeijer, N.: Validation of ozone monitoring instrument level 1b data products, J. Geophys. Res., 113, D15S06, doi: 10.1029/2007JD008665, 2008.

Eatough, D. J., Caka, F. M., and Farber, R. J.: The conversion of SO_2 to sulfate in the atmosphere. Israel Journal of Chemistry, 34(3-4), 301-314, 1994.

Jaross, G., and Warner, J.: Use of Antarctica for validating reflected solar radiation measured by satellite sensors, J. Geophys. Res., 113, D16S34, doi:10.1029/2007JD008835, 2008.