Interactive comment on “Absolute calibration of the colour index and O$_4$ absorption derived from Multi-AXis (MAX-) DOAS measurements and their application to a standardised cloud classification algorithm” by Thomas Wagner et al.

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Reply to Anonymous Referee #2

The paper gives two main results: 1) new calibration methods for absolute calibration of colour index (CI) and O4 absorption and 2) location-independent threshold values for an earlier developed cloud-classification scheme. The calibration methods developed in this paper together with the new threshold values and the adapted colour index are an important step towards a uniform cloud classification scheme for DOAS measurements of scattered sunlight, as the authors state correctly. The paper is well-written and the method is well-demonstrated. Therefore, I recommend to have this paper accepted for publication in AMT, after considering the minor comments below.

Author reply: We thank the reviewer for his/her positive assessment of our manuscript and for many very good suggestions. We addressed almost all of the points raised by the reviewer as outlined in detail below.

General comments: - The authors make extensive use of radiative transfer calculations. The method or model used for that should be described.

Author reply: We added the following information at the beginning of section 2: ‘In this study (like in Wagner et al., 2014) we use the Monte-Carlo radiative transfer model MCARTIM (Deutschmann et al., 2011) for the simulations of radiances, CI and O4 absorption. The settings used for the radiative transfer simulations used in this study are described in section 2.2 of Wagner et al. (2014).’

- Section 4 is difficult to read, since it assumes the reader to be very familiar with the original cloud-classification scheme, and in addition is a mixture of actual new methods, comparison to the old scheme, discussion, and recommendations for future research. I recommend to restructure and shorten this section considerably, and add a separate section "discussion". It should be made clearer what is new, and what contributes to a more uniformly applicable classification, and what not. Also a table summarising all differences between the old and the new classification scheme, would be illuminating.

Author reply: We completely restructured section 4. The old sub-sections 4.4.1 and 4.7 are moved to the new section 5, which is now named ‘Further improvements of the classification scheme’. We added an updated version of Fig. 14 from Wagner et al. 2014 (new Fig. 9) which summarises the general structure of the classification scheme. We further included a new table (Table 1) which compares the old and new thresholds and normalisation procedures of the individual decisions.
Specific comments: - The introduction contains some text that does not belong there. The reader expects only the context, history, and objectives here: a) The text between page 2, line 29 and page 3, line 5 contains important references to previous methods or suggestions for absolute calibration, but are mixed with statements on what is or is not being done in this study. I recommend to remove these statements, and rephrase so that it is a discussion on previously published methods for the absolute calibration of the important parameters.

Author reply: We changed the text to: 'The proposed CI calibration comprises the determination of a proportionality constant, which converts the measured values into well-defined quantities (i.e. radiance ratios for the selected wavelengths). Similar suggestions for the calibration of the CI were already presented by Gielen et al. (2014) and Wagner et al. (2014). For the O4 measurements the calibration comprises the determination and correction of an additional offset (the O4 absorption of the Fraunhofer reference spectrum, FRS) like in Wagner et al. (2014). Already in Wagner et al. (2014) the measured CI and O4 were calibrated based on selected clear sky measurements. In contrast, here we develop standardised calibration algorithms for CI and the O4 absorption, which can be applied to other MAX-DOAS measurements in a consistent way.'

b) The text between page 3 lines 6 and 14 provides information on important modifications to the cloud-classification scheme. I recommend to make a separate section for this.

Author reply: We inserted a new sub-section (1.1 Update of the cloud classification scheme). At the beginning of this sub-section we added the following text: 'After applying the new calibration algorithms to the measurements the calibrated CI and the O4 absorption differ slightly from the calibrated values of the original classification scheme. Thus the threshold values of the cloud classification scheme have to be adapted accordingly. In addition to these changes, further improvements to the original classification scheme (Wagner et al., 2014) are introduced: . . .'

c) Page 3, lines 15 and 16 belongs in a conclusion, or can be rephrased as an objective of this study.

Author reply: We changed the text to: 'One major aim of this study is to provide a universal cloud classification scheme for MAX-DOAS measurements based on the new calibration procedures for the CI and the O4 absorption and the updated threshold values.'

- Page 4, line 7 refers to the "original CI". Some explanation should be added here, either the reference to the Wagner et al, 2014, paper or the old wavelengths used.

Author reply: We changed the sentence to: 'In the following the original CI (based on the wavelength pair 320 nm and 440 nm, see Wagner et al. 2014) is indicated by CI_orig and the new CI by CI_new.'

- Page 4, line 28-29, "Instead, the low CI values are probably caused by 3D effects of broken clouds": Please explain what is meant by 3D-effects and how would they lead to lower CI values.

Author reply: We added the following information to the text: 'If for example the side of a cloud is illuminated by the direct solar beam, the composition of the light, which enters the cloud might change compared to horizontally homogeneous clouds. The relative fraction of the diffuse sky radiation (which is blueish) compared to those of the direct solar beam might decrease, because the cloud side is illuminated by only part of the downwelling diffuse sky radiation. This effect would lead to a decrease of the CI.'

- Page 4, line 29-31, "However, even the lowest measured CI are close to the simulated minimum": this is not completely true, there is one clear outlier. Please also quantify the percentage of measurements with CI lower than the simulated minimum.

Author reply: We inspected the spectrum, for which the very low CI are found (around 18:00). From the spectral analysis (large residual) it is evident that this spectrum is of bad quality. But the specific reason for the bad quality is not clear. We added the
We quantified the percentage of the CI below the minimum: -20.0 % for CI_orig -20.1 % for CI_new

We added this information to the text: 'Interestingly, about 20% of all measurements are slightly lower than the simulated minimum values.'

Page 5, line 5, "Their maxima directly represent the ... proportionality constant": This is not necessarily true for locations where there are mostly clear skies. It might be necessary to remove clear-sky data from this analysis.

Author reply: At the end of section 2 we added the following information: 'It should be noted that for measurements at locations with very low or very high cloud probability a larger time period than for our method might be needed to obtain a sufficient number of both cloudy and cloud-free measurements. In extreme cases, an accumulation point might even exist for CI representing clear sky conditions (if also the AOD stays constant over an extended time period). In such cases, clear sky measurement might be identified by visual inspection and be removed before the frequency distribution is calculated.'

Section 3: This section determines the O4 AMF for one single FRS. How would this method change if different reference spectra are used throughout the data set? Also only one VCD is used for the Cabauw data set, and it is unclear if this is also the case for the Wuxi data set (a complete year). What is the expected error of using one single VCD?

Author reply: We added the following text at the end of section 3: 'Finally, two important aspects should be mentioned: a) For long term measurements, it might be necessary to use different FRS for different parts of the whole time series. In such cases the calibration procedure has to be applied for each selected FRS. b) The O4 VCD depends on atmospheric temperature and pressure. Thus it varies with time. Depending on the weather conditions and season, such changes can exceed 10%. The variation of the O4 VCD leads to a similar variation of the measured O4 DAMFs. In addition, the temperature dependence of the O4 cross section probably further increases this variability. So far, these effects are not explicitly considered in most studies, and also here we assumed the O4 VCD to be constant. Thus part of the scatter of the measured O4 AMFs in Figs. 7 and 8 might be caused by the variation of the O4 VCD and temperature dependence of the O4 cross section. However, the current version of the algorithm is only slightly affected by the corresponding uncertainties of the derived O4 DAMFs, because they are used only for the identification of optically thick clouds and fog. Future studies might take the effects discussed above into account when retrieving the O4 DAMFs from the measured spectra.'

Page 7, line 6-7 "A very similar value for AMFFRS (1.75) was also found by Wagner et al. (2014).": Please explain what is different between the methods used in Wagner et al 2014 and the method used in this study.

Author reply: We changed the text to: 'In the original version of our algorithm (Wagner et al. 2014) AMFFRS was determined based on selected clear sky days with similar AOD. In spite of the different procedures, a very similar value for AMFFRS (1.75) was found.'

Page 7, line 11-12: "... under similar conditions." Please describe what makes them similar.

Author reply: We changed the text to: 'The derived value for the O4 AMF of the FRS is almost the same as for the Cabauw measurements as both FRS were recorded at similar AOD and SZA.'

Page 8, line 5-6: "Simultaneous AERONET measurements": please quantify, or include in figure caption.
Author reply: We added the information that the blue curves represent simulation results for the AOD measured by the AERONET instrument in the morning of 24 June 2014 to the text: 'During the morning the measured CI are similar to the simulation results (blue lines) for the AOD obtained from the simultaneous AERONET measurements.'

In the figure caption we added the following information: ‘The blue curves represent CI for the AOD measured by the AERONET instrument on the morning of 24 June 2009. The red curves represent CI for the threshold values used in the new classification scheme.’

- In Figure 10, there is a distinct different behaviour between the black line and the other lines for small solar zenith angles. Is there just a mistake in the legend? Otherwise this needs more explanation.

Author reply: Indeed, there was an error in the legend. The black line showed the AERONET results. Thus the agreement of the results of the different versions of the cloud classification schemes is actually much better. Nevertheless, Fig 10 (and Figures 11 – 13) were removed in the revised version. Instead a quantitative comparison for the individual classification results (new Fig. 14) was added.

- There is no need for an appendix, the table and Figures in the appendix are important enough to be included in the main paper.

Author reply: We partly followed the suggestion and moved Fig. A2 to the main text (new Fig. 12). But we prefer to leave Table A1, Fig. A1 and Fig. A3 in the appendix in order to limit the length of the main part of the paper. We also added a new figure (new Fig. A3) to the appendix. It shows simulation results of the O4 AMF for different surface albedos.

- Section 4.2: A polynomial for TSI thresholds for zenith view is provided in Tables 1 and A1, although in the text it is explained that it can not be universally used. Therefore, I recommend to leave it out of the Tables. The authors should give a clear discussion on how this influences their objective to come to a uniform cloud-classification scheme. An alternative is to leave out Section 4.2 entirely. In this respect also the statement in the conclusion (page 13, lines 18-20) needs some refinement.

Author reply: We modified the definition of the TSI: in the new definition (new equation 7) the time (differences) of the measurements are not explicitly considered anymore. The advantage of the new TSI is that it is much less dependent on the specific instrument properties or measurement protocols. It can thus be used in a universal way in our cloud classification scheme. We compared the new TSI with the older versions (new Fig. 11) and found good agreement. The corresponding SZA-dependent threshold values are provided in Table 2 and Table A1.

- Section 4.3, including 4.3.1: I can not see what is new here. The threshold value for spread in CI is the same as in Wagner et al (2014). This section further only contains discussion and recommendation for future research, and is too long. See my general comment on Section 4 above.

Author reply: We made it more clear in section 4.3 (also new Table 1) that in contrast to the original algorithm, in the new algorithm the spread of the CI is not normalised anymore. We shortened section 4.3 and moved the recommendations for future research to the conclusions. The sub-section 4.3.1 was moved to the new section 5.

- Section 4.1.-4.3, Figure 10-13: The comparison between the old and new algorithm is done on fraction of measurements with a certain classification. However, if the old and the new algorithm result in the same fraction, that does not necessarily mean that the classification for individual measurements is the same. What is interesting is whether individual measurements get the same classification. Please quantify and discuss how many classifications were different between the two algorithms, and in what way.

Author reply: We agree with the request (and a similar request of the other reviewer) and removed Fig. 10 – 13. Instead a quantitative comparison for the individual classi-
fication results (new Fig. 14) was added. The comparison results are discussed in the new sub-section 4.6.

- Section 5.2: I recommend to change the title, since this is a discussion on the effect of surface albedo, which can be high also at moderate or low latitudes. Please adapt the text in this section accordingly.

Author reply: We changed the title to ‘Observations at high latitudes and over bright surfaces’ We prefer to keep ‘high latitudes’ in the title, because part of the discussion in this section is about the restriction of the SZA at high latitudes. We added the sentence to the text: ‘Here it should be noted that also at mid latitudes the surface might be covered by ice and snow during part of the year.’

- Section 5.2: From Figs 6 and 17 it is clear that the SZA for which the spread in the O4 AMF is almost independent of AOD is different for different surface albedo. It would be interesting to see a figure of this “optimal” SZA as a function of surface albedo. As already suggested by the authors, it seems straightforward to make the method applicable for regions with high surface albedo as well. The only difference is in the selection of data points from which the O4 AMF for the FRS is determined. For low surface albedo the SZA between 30 and 50 degree are used, for high surface albedo the SZA between, e.g., 70 and 90 degree, following from the above-mentioned dependency. For each location the data can be filtered for periods with similar surface albedo (snow or no snow).

Author reply: We added a new figure (Fig. 19) to the manuscript. It shows that interestingly, for all albedo values a specific SZA exists, for which the O4 AMFs become independent from the AOD. The dependence of this SZA on the surface albedo is almost linear. Individual simulation results for the O4 AMF for different values of the surface albedo are added to the appendix (new Fig. A3).

Tecnical comments: page 3, line 7: change "were" to "are"

Corrected

page 3, line 10: change “420” to “440”
corrected

page 5, line 8: change “cloud sky” to “suspected broken cloud” (or clarify)
‘cloud sky’ was replaced by ‘clear sky’.
page 6, line 19: change “at he” to “at the”
corrected

page 6, line 27: change “applied to the remove” to “applied to remove”
corrected

page 6, line 27: remove " from the O4 AMF”
removed

page 7, line 5: change “poor statistics” to “low number of observations”
corrected

page 8, line 4: change “CI for AOD” to “CI for AOD=0.75 and 0.85 (red lines)”
Table 1: The coefficients in the table should have less significant digits, and should be written in scientific notation. In addition the normalisation of S should be given in the caption (1 degree?). I recommend to normalise S with 90 degrees, so that all coefficients have similar orders of magnitude, and to have a maximum of 5 or 6 significant digits. The first polynomial would read, e.g., y = 8.666s5+6.364s4+6.804s3+0.464s2+0.180s + 0.540, with s between 0 and 1.

Author reply: Many thanks for this good suggestion. We changed the representation of the polynomials as suggested.

page 8 line 30: different spelling of "Angstrom"
Angstrom is changed to Ångström
page 9 line 10: change "versions of or algorithm" to "versions of the algorithm"
The part of the section with this sentence was removed
page 11 line 19: "compared for and elevation angle" to "compared for an elevation angle"
corrected
page 10, line 23: threshold of old algorithm is 0.74 (table 2 in Wagner et al, 2014) instead of 0.8
corrected
Figure 1: make the distinction between aerosols and clouds visible in the legend, e.g., AOD=0.2, and COD=2.
corrected
Figure 1, caption: change "Heyey" to "Henyey", and "Asymmetry" to "asymmetry".
corrected
Figures 1, 2, and A2 caption: put a symbol for radiance (I) in the formula for CI (e.g. $I_{320nm} = I_{340nm}$ instead of $320nm / 340nm$)
corrected
Figure 6, caption, line 5: change "to the left" to "to the left". Quantify which aerosol layer height and surface albedo are used for the simulations.
Author reply: Corrected, the information on aerosol layer height (1000m) and surface albedo (5%) was added.
Figure 8, caption: change "represents" to "represent", and "Cabuw" to "Cabauw".
corrected
Figure 9, caption, line 5: change "The the simulations" to "The simulations". Explain that the red line represents the threshold value distinguishing clear-sky from cloudy.
corrected
Figure 14, caption: specify wavelength of radiance
Author reply: The wavelength (360nm) was added to the caption.