

Reviewer #1

Interactive comment on Comparison of south Atlantic aerosol direct radiative effect overclouds from SCIAMACHY, POLDER and OMI/MODIS by Martin de Graaf et al.

This paper examines satellite retrievals of the radiative effect of absorbing aerosols that overlie clouds (here termed the DRE). Retrievals from OMI+MODIS, POLDER and SCIAMACHY are compared. The latter can observe at many different wavelengths, but has low resolution. POLDER can observe the degree of polarization of the reflected light, which allows extra information about the aerosol and cloud to be obtained and minimizes the retrieval assumptions that need to be made. It is found that OMI+MODIS and SCIAMACHY agree reasonably well, but that POLDER produces larger DRE and cloud optical thicknesses (COT). Some of this difference is attributed to sampling issues (mainly arising from the different resolutions of the instruments) and some due to the larger optical depths retrieved by POLDER.

The study should be useful to other researchers since it would be useful to know how large this warming effect is (can it offset a significant amount of aerosol-cloud cooling?) and whether the models get it right. It also seems like the POLDER approach has some promise, particularly if it can be combined with more conventional instruments on e.g., the upcoming METOP-SG 3MI platform. As such I think it should be published after the suggested revisions.

However, the arguments are often a bit muddled and it would be good to see the reasons for the larger POLDER COT values explored a little more, as well as some more investigation into the effect of the low resolution retrievals from the other instruments. The paper talks a lot about ‘sampling errors’ for OMI+MODIS and SCIAMACHY, but this seems to assume that all such errors are just from averaging of the final DRE or COT values, whereas it seems likely that some retrieval errors may be introduced by the averaging effects of the reflectances to low resolution, particularly if the relationship between the reflectances and the retrieved quantities are non-linear. Such effects occur for MODIS retrievals of effective radius and COD for example (Zhang; doi:10.1029/2012JD017655, 2012). It would be good to discuss this and to look into this possibility. It would even be possible to test what effects the averaging of reflectances to lower resolutions might have using synthetic higher resolution reflectances. On a similar note – considering just the ‘sampling effect’ (i.e., just the effect of averaging the retrieved quantities, rather than the reflectances), it should be possible to quantify this effect by degrading the POLDER retrievals to the coarser grids, rather than the other way round, as is currently done.

Section 3.2.3 needs some checking as some of the statements regarding the POLDER optical depth being smaller seemed to contradict the results. The explanations were also not clear.

The reviewer is thanked for the careful and thorough review of the manuscript. Many valuable suggestions were made, which were followed unless stated otherwise, in which case a motivation is given. In particular, the regridding of POLDER data to the coarser grid of OMI was performed, to improve the comparison. This was not done the first time, because SCIAMACHY has the coarsest grid, and regridding to

SCIAMACHY, and especially requiring SCIAMACHY collocation, yielded too sparse datasets. However, if only POLDER and OMI-MODIS are compared and collocated on the OMI grid, the analysis is much improved.

The manuscript was rewritten, to better distinguish between sampling issues and retrieval uncertainties. Sampling issues arise from the fact that different sensors in different orbits see different parts of the the atmosphere, and that different filter settings yield different pixels taken into account. This can be solved by requiring strict collocation of the considered pixels.

However, such collocation also requires resampling and regridding of data that are originally on different spatial resolutions. As the reviewer points out, nonlinear effects play a role here, and we have included a discussion on the role of the plane parallel bias for heterogeneous clouds. In our analyses MODIS radiances were added and resampled on the OMI footprint, while POLDER COT are averaged over the footprint. This has effects on the COT and CER averaging in a satellite footprint, and can account for the differences found between POLDER averaged COT and OMI-MODIS COT. We have tried to explain and quantify differences that we find. The resulting DRE differences are now explained in terms of the uncertainties in the AOT and COT retrievals. Additional improvements of the measurements can then improve the DRE retrieval, but this is not the focus here.

All the issues raised by the reviewer are addressed below:

Specific statements

p.1 L15 Aerosol-cloud-radiation interactions currently present the largest uncertainty in our understanding of Earth's climate (Boucher et al., 2013). The effects of atmospheric aerosols are especially uncertain.

The second sentence here reiterates the first and does not really make sense. It should be removed, or else made more clear what it is referring to. Do you mean that the effects of aerosols alone are especially uncertain (compared to cloud-aerosol interactions)? However, think that it is hard to argue that this case.

Aerosol effect in global climate models are currently the largest uncertainty in global climate change attribution. However, the poor phrasing was also noted by reviewer #2, and the introduction was rewritten to better reflect the current state of aerosol climate science, and to clarify the text.

p. 1 L18 – ‘The presence of clouds has a strong influence on the DRE from the light absorbing species in smoke at TOA.

It's hard to understand what you mean here. I think you mean something like this :

–
‘The DRE (at TOA) due to the light absorbing species in smoke is strongly affected by the presence of clouds.’ Although maybe it would be good to introduce the idea of light absorption (rather than just scattering) affecting the DRE before this sentence. Or maybe this sentence isn't necessary given what follows?

I think an introductory sentence improves the paragraph, and the suggestion by the reveiwer was adopted as given.

p. 1 L20 – ‘Over clouds, on the other hand, scattering by aerosols is negligible’ –

this is not quite correct I think. The scattering due to aerosols overlying cloud would be quite high – it is the cloud that is doing less scattering in this case because of this. I think you mean that the addition of aerosols above a cloud has negligible extra impact on scattering relative to that which the cloud is already causing.

Agreed. The text has been modified to read: ‘Over clouds, on the other hand, scattering by aerosols hardly contributes to the upwelling radiation at TOA, since the scattering by clouds is dominant. However, the aerosols absorb radiation, lowering the planetary albedo, resulting in a positive direct effect (warming).’

p.5, L2 – ‘CER was derived from collocated MODIS measurements.’ Would it not be better for POLDER to retrieve the CER? Is this retrieval not possible? Could MODIS CER be biased by the overlying aerosol, or by inhomogeneous clouds, etc.?

POLDER does not have measurement in the near infrared. MODIS CER is retrieved primarily from the 2.1 μm channel over the ocean. It can potentially be biased by the presence of aerosols above clouds. However, in the region of interest, the aerosols typically observed above clouds (i.e. biomass burning aerosols) are characterised by a large Ångström exponent. Therefore, their contribution to the signal at 2.1 μm is expected to be negligible. This is the same argument that is used for the (OMI-)MODIS retrievals, except at 1.2 μm . At 2.1 μm the effect will be much smaller. Regarding the 3D effect, several filters are used on the POLDER AAC products in order to reject inhomogeneous clouds (Waquet *et al.*, 2013b, GRL)

p.5 L30 – ‘MODIS, on-board the Aqua satellite, flies in formation with Aqua in the A-Train, leading Aqua by about 15 minutes’ Should this be MODIS flies in formation with and leads Aura?

Correct, it should be (and is now): MODIS, on-board the Aqua satellite, flies in formation with Aura in the A-Train, leading Aura by about 15 minutes.

p.6 L14 - ‘Note however, that such an estimate is often missing, while methods other than DAA are moreover highly uncertain due to their dependence on the correct characterization of the spectral properties of the overlying aerosols.’

This doesn’t quite make sense. Do you mean that often such an error estimate is not made in other studies (does this only apply to those that use DAA)? Please correct if so. The part after should probably be a separate sentence.

This is correct, an estimate on the individual measurements (of DRE in this case) is often missing. Many satellite products are delivered without uncertainty estimate on the individual measurements, e.g. relevant for this manuscript: OMI, MODIS, and CALIOP above cloud AOT. Uncertainty estimates are obtained from comparison with other datasets, like is done in this manuscript. However, we argue that error and uncertainty estimates can, and should, also be given on the basis of assumptions and uncertainties of the input parameters, which lead to measurement uncertainties. In that case, comparisons like the current one, can be performed in light of the uncertainties of the measurements.

Here, we have tried to quantify the uncertainties in aerosol DRE in terms of uncertainty estimates in above cloud AOT and COT for POLDER, and relate the difference between OMI-MODIS and POLDER DRE in terms of those uncertainties.

p.6 L16 – ‘Other minor error sources for the DAA method are the uncertainty in input parameters, the influence of the smoke on the estimated cloud fraction, cloud optical thickness and cloud droplet effective radius, an uncertainty in the anisotropy factor (de Graaf et al., 2019), and the uncertainty of estimating the COT and CER at SWIR wavelengths.’

In Section 3.2.3 you say that the DRE depends very strongly on the COT. So, wouldn’t the COT uncertainty be likely to have a larger contribution to the error than indicated here? Also, this sentence needs to use semi-colons to make it clearer to become :-

‘Other minor error sources for the DAA method are the uncertainty in input parameters; the influence of the smoke on the estimated cloud fraction, cloud optical thickness and cloud droplet effective radius; an uncertainty in the anisotropy factor (de Graaf et al., 2019); and the uncertainty of estimating the COT and CER at SWIR wavelengths.’
 Yes and no. The (large) effect of the COT uncertainty on DRE is in the uncertainty estimate of the DRE by applying it to aerosol-free cloud scenes, which yields the rather large uncertainty of 13 Wm^{-2} for OMI-MODIS DRE. The additional cloud uncertainties investigated in De Graaf et al. (2012) are the effect of *smoke* on the cloud parameters, and uncertainty of estimating CER and COT in the SWIR *instead of in the visible*. However, it is agreed that this is not clear from the text. Furthermore, we show in this paper that the effects of COT and AOT are coupled and COT uncertainties will have larger effects at larger above clouds AOT, which was not estimated. This was added to the manuscript.

p. 8, L2 – ‘the instantaneous aerosol DRE over clouds was normalized by dividing by the cosine of the solar zenith angle.’

Have you checked whether the DREs scale linearly with the cosine of the angle (presumably a proxy for the incoming SW)? This could be checked with a radiative transfer code. If not then this might introduce some bias. Presumably there is a lower limit for the solar zenith angle allowed?

The DRE is defined as the difference in upwelling flux at TOA for a cloud scene and a cloud with aerosol scene, which can be written as

$$\text{DRE}_{\text{aer}} = F_{\text{cld+aer}}^{\uparrow} - F_{\text{cld}}^{\uparrow} = \mu_0 \int_{\text{SW}} E_0(\lambda)(A_{\text{cld}} - A_{\text{cld+aer}}) d\lambda \quad (1)$$

where A is the local planetary albedo, defined in terms of the reflectance $R = \pi I / \mu_0 E_0$ as $A(\mu_0, \phi_0, \lambda) = \frac{1}{\pi} \int_0^{2\pi} \int_0^1 R(\lambda; \mu, \phi; \mu_0, \phi_0) \mu d\mu d\phi$. Neglecting the small effect on the planetary albedo (which is the effect of the BRDF of the cloud scenes, that has been treated in De Graaf et al. (2012) and De Graaf et al. (2019)), the fluxes can be noon-normalised by dividing DRE by μ_0 to cancel the effect of different solar incoming fluxes $\mu_0 E_0$ on the DRE computation during the overpasses of SCIAMACHY (10:00 LT) and OMI, MODIS and POLDER (around 13:30 LT).

The solar zenith angle has no lower limit, since this does not produce any problems. High solar zenith angles may introduce biases, but these do not occur, because

the considered area of the south-east Atlantic basin is near the equator.

p. 8, L26 – ‘The main reason for the much larger area-averaged POLDER DRE on 12 August 2006 is the smaller coverage of the area by POLDER, compared to that by OMI/MODIS, due to a smaller swath. This was illustrated in Figures 1d and 1e.’

This is part of the reason, but it seems that generally POLDER gives considerably higher values for the same regions. You could help demonstrate the magnitude of the differences caused by the different swath area vs that of POLDER values being higher by giving the collocated averages in Table 1. It would be better to change ‘The main reason’ to ‘One of the reasons’.

All the sampling issues are resolved when the OMI-MODIS data are regridded to POLDER or vice versa, which is shown in the manuscript. The description here is merely to illustrate the main difference between the two datasets on 12 August 2006, which is not the day with the most extreme DRE values, but the day with the largest difference between the area-averages. The main reason is the sampling, which is very different for OMI-MODIS and POLDER, as shown in Figure 1d–e. When the sampling issue is removed the average for that day reduced from 74 Wm^{-2} difference to 33 Wm^{-2} difference. So the sampling issue is the main reason for the large discrepancy.

The numbers are added to table 3.

p. 8, L33 – ‘This means that a (dense) plume may be sampled once by a far off-center pixel, or by 15 nadir pixels, all of them receiving the same high values, depending on the satellite track.’

For this to have an effect on the average it would require that the values retrieved from the mean reflectances over the larger pixel did not produce the correct average DRE value – i.e., there is a non-linear relationship between reflectance and the retrieved products, so that the result is dependent on the averaging scale (pixel size). It would be worth nothing this here. Also, the sentence would be clearer without ‘all of them receiving the same high values,’.

This unclear statement has been removed. The text has been changed to just state the different pixel sizes. The discussion has been changed to showing the effect of regridding, both SCIAMACHY and OMI-MODIS to the high-resolution POLDER grid, as POLDER to the OMI grid.

p.10, L20 – ‘This issue could be resolved if all values were regridded to the coarsest available. However, since this is the SCIAMACHY grid, not many grid cells would remain.’

Although you could do it for the OMI grid vs POLDER, which would be useful?

Yes. The main addition to the new manuscript is the regridding of POLDER DRE to the coarser OMI grid. The text was rewritten to include this analysis. Without the SCIAMACHY collocation requirement the coverages of both datasets are still very good after collocation and sampling issues are removed. It shows that regridding to POLDER grid or OMI grid does not change the results very much, but the removal of the SCIAMACHY collocation requirement ensures much better statistics.

p.11 L19 – ‘A comparison of SCIAMACHY, OMI/MODIS and POLDER COT histograms (not shown) revealed a slightly higher COT from SCIAMACHY and OMI/MODIS compared to POLDER (up to 42 for POLDER and 48 for OMI/MODIS (Schulte, 2016)), but the maximum of POLDER is restricted due to LUT limits.’

It’s not clear here where or when these histograms apply to. I see that it is likely to refer to the 19 August case (Table 3), but it needs to be mentioned in the text. Also, ‘a slightly higher COT from SCIAMACHY’ should be changed to ‘a slightly higher maximum COT from SCIAMACHY’ since it otherwise it sounds like you are referring to mean values. However, visually it looks from Figure 5 like POLDER has higher maxima in general? You should also explain the part about the LUT limits in the context of the statement on p.10 L24 (‘The POLDER DRE is dependent on the retrieved AOT and COT, which in principle are both unbounded.’).

The discussion on COT was completely rewritten and this text was removed. We believe the COT analysis is now much clearer, the relationship between POLDER COT and OMI-MODIS COT is now shown in Figure 6, with POLDER COT being systematically higher.

The statement on p.10 L24 is rephrased as: The POLDER DRE is dependent on the retrieved AOT and COT, which in principle are both unbounded (although in the LUT for POLDER THE COT is limited to 42).

p. 11 L28 – ‘Even though the OMI/MODIS data are regridded to a high resolution grid, the values are obviously still more smoothed compared to the COT on the native high resolution POLDER grid. Therefore, even though POLDER COT and POLDER DRE are generally smaller than from OMI/MODIS on average, the extreme values and averages are higher.’

The second sentence seems to contradict the rest of the paper – from the tables and figures POLDER has a generally larger DRE and COT?

The discussion on COT was completely rewritten and this text was removed.

Table 3 – it should be made clear in the table caption that the DRE was calculated using the POLDER AOT in both cases.

Agreed, this was added to the caption, and the value itself was placed in the center, so it is clear it was not retrieved for OMI-MODIS.

p.12 L8 – ‘It shows that the difference between these two quantities disappears completely for these instruments, and the slope is even reversed.’

- It has reduced a lot, but not disappeared completely! Plus, saying that the slope has reversed is a bit unclear. Perhaps better to say it went from <1 to >1.

The discussion on COT was completely rewritten. Only OMI-MODIS and POLDER data were used to examine the DRE differences. The relationship between POLDER COT and OMI-MODIS COT is now shown in Figure 6.

p. 12 L20 – ‘The aerosol DRE from POLDER is completely independent. It correlates well with SCIAMACHY and OMI/MODIS DRE for moderate values, but is larger than SCIAMACHY and OMI/MODIS DRE for high values. This is caused by a larger COT retrieved by POLDER, and to a lesser degree by an underestimation of the aerosol

DRE using DAA, which by definition assumes a zero AOT at SWIR wavelengths.

The largest contribution to the difference between SCIAMACHY, OMI/MODIS and POLDER DRE are sampling issues.'

- It seems that the last sentence contradicts the ones before where it says that larger COT retrievals by POLDER are the cause. Is it the COT differences or the sampling issues that are most important? Or are they equally important? See also p.13 L8. Also, L9 in the abstract says that sampling issues are the most important – is this actually the case and can you point to the evidence that shows that it such errors are larger than the COT errors?

Sampling differences are the main source for DRE differences, which is now clearer from the case of 12 August 2006, see above. Only after sampling is removed an analysis of the differences in terms of the input parameters and assumptions makes sense. After removing sampling issues, COT differences are the largest contribution to the found differences. AOT differences have a smaller but still important effect, and AOT and COT influences are coupled. The new manuscripts makes these conclusions more apparent, thanks to the suggestions of the reviewers.

p.13 L5 – 'This approach removes issues related to selecting high positive DRE values by filtering on COT and CF, which introduce large differences in the average DRE.'

It's not clear what you are referring to here regarding filtering of COT and CF – is this a method that has been suggested in the literature (please say so and give a reference if so). Or from this paper – again this needs to me made clear.

The sampling by the three instruments is different. Not only because they sense different areas, but also because even if the same limits on permitted cloud fractions and COT are used, the results will be different, simply because the retrieved CFs and COTs may be different. Therefore, even perfectly aligned instruments with exactly the same filter settings will sample different parts of the Earth. These issues are also resolved with the collocation requirement used in the analysis.

A sentence explaining this was added to the manuscript: 'Even if the same filtering is used for the CF and COT for all instruments, different areas will be sampled, because the CF and COT retrieved by the different instruments may be different.'

p. 13 L13 – 'Normally, MODIS COT retrievals at 0.8 and 1.2 microns retrievals' - Doesn't the usual MODIS retrieval over oceans use the 0.86 and 2.1um bands? Correct, this has been changed to 2.1 microns.

Figures *Fig. 2 – The linewidths of the monthly mean lines need to be quite a bit thicker for the colour and dash style to be visible.*

Agreed.

Fig.3 – the legend lines need to be thicker to be able to see the different colours.
agreed

Typos

The word 'microns' is used a lot, but also the symbol 'µm'. I think that the latter is

the ACP standard for units.
Agreed. All are changed to μm .

p. 7, L20 – ‘when the comparison between the instrument is worst’ -> ‘when the comparison between the instruments are worst’
‘instrument’ was changed to ‘instruments’. (the comparison ..) ‘is’ was retained.

p. 7, L30 – ‘Here, we show the effect of ignoring the sampling effect, even of area averages of, in this case, aerosol DRE over clouds.’
The text was changed.

– this doesn’t quite make sense. How about something more simple like ‘Here we show the effect of ignoring the sampling differences between instruments’?
Agreed. I also don’t understand this sentence. The reviewer’s suggestion was adopted.

p.10 L23 – ‘possibly’ -> ‘possible’.
Correct. Text was changed.

p. 11 L12 – ‘This way, an AOT at 1.2 μm can be found between 0.15 and 0.35’ -> ‘In this way an AOT at 1.2 μm of between 0.15 and 0.35 can be found’
Agreed.

p. 11, L22 – ‘However, the spectral variation in COT is very small. Only for very small cloud droplets the COT at 0.87microns is about 4% smaller than the COT at 1.2microns for cloud droplet effective radii of 4 microns, and this reduces for larger droplets.’

- This would be better as :- ‘However, the spectral variation in COT is very small and is only significant for very small droplets. For example, for cloud droplet effective radii of 4 microns the COT at 0.87microns is about 4% smaller than the COT at 1.2microns and this reduces for larger droplets.’
Gladly accepted.

p. 13 L23 – ‘Comparing AOT over clouds POLDER with MODIS and CALIOP, showed POLDER to be high, but not necessarily overestimated’ – insert ‘from’ between ‘clouds’ and ‘POLDER’.
Agreed. Text was changed.

Reviewer #2

Interactive comment on Comparison of south Atlantic aerosol direct radiative effect overclouds from SCIAMACHY, POLDER and OMI/MODIS by Martin de Graaf et al.

This short study is a comparison of above-cloud aerosol direct radiative effects estimated by three methods applied to three satellite sensors or combinations of sensors (POLDER, SCIAMACHY, and OMI/MODIS). Looking at two days in August 2006 and at daily averages over 4 months in 2006, the authors find sizeable differences between the three sets of estimates, with POLDER retrievals producing significantly stronger radiative effects. Those differences are reduced when correcting for sampling differences. The remaining differences can be explained by differences in aerosol and cloud optical thickness, with cloud optical thickness being the dominant cause. The study is of interest to the wider aerosol community because aerosol modellers have now begun to use above-cloud aerosol retrievals to compare against their models, and large differences between observation-based estimates weaken observational constraints. This study is hopefully a first stage to eventually reconciling the different estimates. The paper is generally well-written, although language editing will help in places, and Figures and Tables illustrate the discussion well. My main criticism of the study is that it does not attempt to bring additional information to resolve the disagreement. The discussion can also be improved in places. I recommend major revisions because addressing my main comment will probably require additional analyses.

1 Main comment *The study concludes that differences are mostly caused, once the effect of sampling has been accounted for, by differences in cloud optical thickness (COT) retrievals between the instruments. Differences in aerosol optical thickness (AOT) also play a role, especially at longer wavelengths. But it would be most useful to know which dataset does best. Retrievals of AOT in nearby clear-sky regions, or using CALIOP, or even nearer the sources by AERONET should help determine whether the large AOTs (almost 2) retrieved by POLDER are realistic. Similarly, differences in retrieved COT are large enough to determine whether POLDER is realistic or not by comparing to CALIOP or passive retrievals, e.g. from SEVIRI. Adding such an analysis would make the study a more ambitious, and ultimately more useful, contribution.*

The reviewer is thanked for the careful evaluation of the manuscript. We have changed many of the discussions, which were indeed sometimes vague, because the results were not always clear. After reanalysis of the sampling issues, by gridding POLDER to OMI and disregarding SCIAMACHY to get a good comparison between two datasets with a good coverage, the results are much clearer, which are -hopefully- clearer presented in the new manuscript.

After sampling, the remaining differences between POLDER and OMI-MODIS DRE are explained in terms of AOT and COT and the uncertainties in those. The uncertainties in COT retrieved by POLDER and OMI-MODIS for polluted clouds are difficult to establish, because there is little to compare with, 'normal' COT retrievals being biased by overlying smoke. Both POLDER COT and OMI-MODIS COT retrievals show continuous behaviour from polluted to unpolluted areas. We show the difference in COT retrieval (9% on average, no extreme differences) and the effect on

the DRE.

For AOT, the comparison with more established datasets is also difficult, because these are all in clear sky. However, an inspection of several AERONET sites showed high AOT during the biomass burning seasons, but never as high as 2 over the Atlantic. Ascension Island (almost 3000 km from the source) has no measurements during 2006, but in other years AOT is measured up to 1 (UV). e.g. in 2016, which was also an anomalously extensive biomass burning season. St. Helena has few measurements, São Tomé has measurements in 2017 and 2018 up to 1.5 (UV). Only over Gabon, which is most likely the source region or in the path of the smoke towards the ocean, AOT at 340 and 280 nm of more than 2.0 was found in August 2016 (data start in 2014).

We have compared the above cloud AOT (ACAOT) from several sources that are currently available, from MODIS, OMI, and CALIOP, all of them science datasets, i.e. no proper validation has been performed for these datasets. High ACAOT up to 1.5 or 2 is common, with POLDER being in line with the highest retrievals. The discussion in the manuscript was extended with these numbers from the literature.

The DRE results and differences are explained in terms of these findings, but no conclusion was given on which dataset is 'best', because the truth is not known.

2 Other comments *Page 1, line 15: The statement 'The effects of atmospheric aerosols are especially uncertain' repeats the first sentence and can be deleted.*

Agreed. The introduction was rewritten to be more clear and correct.

Page 1, line 21: I acknowledge that the terminology of aerosol direct, indirect, and semi-direct effects is now well known by the wider atmospheric science community, but I recommend defining them anyway for the sake of completeness.

Agreed. The introduction was rewritten to include this.

Page 2, line 2: 'which can be characterized relatively well' sounds like an instance of concluding too quickly!

We derive a direct effect of aerosols over clouds including an uncertainty estimate. I think this a relatively good performance, given that semi-direct effects are not even estimated at all from measurements.

Page 2, line 5: Caution: the use of 'forcing' in the sense of Forster et al. 2007 implies that the unperturbed values correspond to pre-industrial conditions. In the present study however, unperturbed values are for an aerosol-free atmosphere, so to avoid confusion I recommend avoiding the word "forcing".

In 2007, Forster *et al.* defined 'radiative forcing' as the net broadband irradiance change ΔF at a certain level with and without the forcing constituent, after allowing for stratospheric temperatures to readjust to radiative equilibrium, but with tropospheric and surface temperatures and state held fixed at the unperturbed values (chapter 2.2.). This was quoted in the manuscript, with 'radiative forcing' changed to 'radiative effect', because the terminology has changed since then in favour of 'forcing' as the change since pre-industrial times, and 'effect' as the instantaneous change. However, one instance of 'radiative forcing' on p2,17 was overlooked, and this was changed to

‘radiative effect’ in the new manuscript.

Page 2, line 33: Myhre et al. (2013) is not the correct reference for that statement, as that paper only refers to global averages and does not isolate cloudy- sky radiative effects. I think the authors mean Figure 2 of Zuidema et al. (2016) doi:10.1175/BAMS-D-15-00082.1 . The same comment applies to Page 13, line 28.

Agreed.

Page 3, line 15: ‘Finally, the . . . using an RTM.’ That has been said already.

Agreed.

Page 3, line 16: ‘highest yet’. What do you mean? Over which period are you making that statement?

Over the south-east Atlantic in 2006, as stated in the manuscript.

Page 5, line 4: ‘(from models)’. Be more specific.

This has been removed.

Page 6, section 2.4: Isn’t it possible to get an error/uncertainty for the POLDER product?

The main source of error for the aerosol DRE over clouds from POLDER is the assumption on the aerosol refractive index. In the first step of the algorithm, an assumption on the refractive index is used in order to retrieve the above-cloud scattering AOT. In the second step, the imaginary part is modified in order to retrieve the absorption AOT from total reflectances, assuming the same real part of the refractive index as in the first step. The impact of the refractive index assumption on the DRE has been analysed in Peers et al (2015) and a maximum error of 10W.m-2 has been observed. Finally, an error on the CER can cause a bias of up to 10% on the COT.

This was added to the manuscript.

Page 7, lines 3–4: How were the two cases selected?

The first case shows the situation during the largest difference between the datasets, and the second case the situation one week later, when the differences are small. During 2006 all instruments performed well, and August is the peak of the biomass burning season in southern Africa.

This was added to the manuscript.

Page 7, section 3.2: That section is confusing. It goes back and forth between case studies and monthly averages. I suggest starting with case studies, then discussing the implications for longer time averages.

The discussion now starts with the cases, and the discussion on the area-averaged DRE goes back to the cases, which are part of the dataset. In Figure 2, the 12th and 19th are indicated more clearly, so the reader understands that Figure 1 and 2 are connected, and where.

Page 7, lines 30–31: ‘even of area-averages’: I do not understand that statement.

As stated, sampling effects are often treated by averaging. Here, we show that this is not sufficient for the sparse DRE over the Atlantic. The text ‘even of area-averages’ was removed.

Page 9, section 3.2.1: The comparison protocol is unusual. The usual method is to regrid higher resolution datasets on to the coarser grids. The reason for doing like that is that the higher resolution represents variability within the coarser grid- box, so it is safe to make an average. But the authors do the other way around, replicating coarser values to fill the higher-resolution grid. Why that choice?

The reason was to avoid the very coarse SCIAMACHY grid. However, another analysis was added without SCIAMACHY and with POLDER gridded to OMI, which is the ‘normal’ way, and still has a large coverage from both instruments. This improved the comparison considerably. Both the reason for the first choice, and the new comparison were added to the manuscript.

Page 11, line 3: ‘it has been shown’ requires a reference.

The statement was from the reference just before the sentence. The reference was moved to include this statement as well.

Page 11, section 3.2.3: Why not show 12 Aug 2006 on Figure 5? The DRE difference is even larger on that day, which should help identify differences in COT as the main cause.

Agreed, the figure was changed.

Comparison of ~~south~~-~~south-east~~ Atlantic aerosol direct radiative effect over clouds from SCIAMACHY, POLDER and ~~OMI/MODIS~~OMI-MODIS

Martin de Graaf¹, Ruben Schulte², Fanny Peers³, Fabien Waquet⁴, L. Gijbert Tilstra¹, and Piet Stammes¹

¹Satellite Observations Department, Royal Netherlands Meteorological Institute (KNMI), De Bilt, The Netherlands

²Geosciences & Remote Sensing Department, Delft University of Technology (TUD), Delft, The Netherlands

³University of Exeter, Exeter, United Kingdom

⁴Université des Sciences et Technologies de Lille 1, Lille, France

Correspondence: Martin de Graaf (martin.de.graaf@knmi.nl)

Abstract. The Direct Radiative Effect (DRE) of aerosols above clouds has been found to be significant over the south-east Atlantic Ocean during the African biomass burning season due to elevated smoke layers absorbing radiation above the cloud deck. So far, global climate models have been unsuccessful in reproducing the high DRE values measured by various satellite instruments. Meanwhile, the radiative effects by aerosols have been identified as the largest source of uncertainty in global climate models. In this paper, three independent satellite datasets of DRE during the biomass burning season in 2006 are compared to constrain the south-east Atlantic radiation budget. The DRE of aerosols above clouds is derived from the spectrometer SCIAMACHY, the polarimeter POLDER, and from collocated measurements by the spectrometer OMI and imager MODIS. All three confirm the high DRE values during the biomass season, underlining the relevance of local aerosol effects. Differences between the instruments can be attributed mainly to sampling issues. When these are accounted for, the remaining differences can be completely explained by the higher cloud optical thickness derived from POLDER compared to the other instruments. Additionally, a neglect of AOT at SWIR wavelengths in the method used for SCIAMACHY and ~~OMI/MODIS accounts for 26%~~ OMI-MODIS accounts about a third of the difference between POLDER and ~~OMI/MODIS DRE.~~ OMI-MODIS DRE, which is mainly evident at high values of the aerosol DRE. The datasets from POLDER and OMI-MODIS effectively provide lower and upper boundary for the aerosol DRE over clouds over the south-east Atlantic, which can be used to challenge Global Circulation Models. Comparisons of model and satellite datasets should also

account for sampling issues. A combination of the DRE retrievals from OMI-MODIS and POLDER may benefit from upcoming satellite missions that combine spectrometer and polarimeter measurements.

35

Copyright statement.

1 Introduction

~~Aerosol-cloud-radiation interactions currently present the largest uncertainty in our understanding of Earth's climate (Boucher et al., 2013). The effects of atmospheric aerosols are especially uncertain. Aerosols can absorb and scatter longwave and shortwave radiation, depending on their internal and external composition. These effects can be strongly amplified depending on the atmospheric composition. Clouds especially can alter the local radiation field, amplifying the aerosol effects and even changing the sign of the net effect~~

40

45

During the monsoon dry season in Africa, biomass burning from wildfires produces huge amounts of carbonaceous aerosols, or smoke (de Graaf et al., 2010). The smoke that is transported over the south-east Atlantic Ocean overlies one of the planet's major stratocumulus cloud decks (Swap et al., 1996). Smoke is a light-absorbing aerosol and the instantaneous change in radiative flux by the scattering and absorption of sunlight is known as the aerosol direct radiative effect (DRE). The absorption of

55

sunlight by aerosols adds heat to the atmosphere at the aerosol layer height, changing the atmospheric stability and the amount of radiation received at the surface (Yu *et al.*, 2002), which in turn affects the development of clouds (Feingold *et al.*, 2005) and precipitation (Sorooshian *et al.*, 2009). Absorbing aerosols in or near clouds may evaporate cloud droplets (Ackerman *et al.*, 2000), while absorbing aerosols above marine stratocumulus clouds may increase the temperature inversion, thickening the cloud (Johnson *et al.*, 2004; Wilcox, 2010). These rapid adjustments to radiative flux changes are known as aerosol semi-direct climate effects. Furthermore, aerosols impact the formation of clouds by acting as cloud condensation nuclei, known as the aerosol indirect effect. Aerosol climate impacts are expected to counteract a significant part of the greenhouse gas-induced global warming, which is estimated at $+2.8 \pm 0.3 \text{ Wm}^{-2}$, but the large uncertainty of aerosol-radiation interactions, ranging from 0 to -0.9 Wm^{-2} , limits our ability to attribute climate change and improve the accuracy of climate change projections (?).

~~Although many aerosol effects have been identified~~ Constraining aerosol effects in model studies, ~~constraining these models~~ remains a challenge as observations of aerosol direct, indirect, and semi-direct effects are scarce. The main problems are the complexities involved in untangling the observations of aerosols, clouds and radiation in the real world. In this paper, we focus on the direct effect of aerosols above clouds, which can be characterized relatively well due to recent developments in retrieval techniques from a number of different satellite instruments.

The radiative effect of an atmospheric constituent can be defined as the net broadband irradiance change ΔF at a certain level with and without the forcing constituent, after allowing for stratospheric temperatures to readjust to radiative equilibrium, but with tropospheric and surface temperatures and state held fixed at the unperturbed values (Forster *et al.*, 2007). For tropospheric aerosols as the forcing agent, stratospheric adjustments have little effect on the radiative forcing effect and the instantaneous irradiance change at the Top Of the Atmosphere (TOA) can be substituted. The instantaneous aerosol Direct Radiative Effect (DRE) at TOA is therefore defined as the change in net (upwelling minus downwelling) irradiance, due to the introduction of aerosols in the atmosphere. Since at TOA the downwelling irradiance F^\downarrow is the incoming solar irradiance F_0 for all scenes, for a cloud scene the aerosol DRE can be determined from the difference between the upwelling irradiance in an aerosol-free cloud scene F_{cld}^\uparrow and the upwelling irradiance of a scene with the same clouds plus aerosols $F_{\text{cld+aer}}^\uparrow$:

$$\text{DRE}_{\text{aer}} = (F^\downarrow - F^\uparrow)_{\text{cld}} - (F^\downarrow - F^\uparrow)_{\text{cld+aer}} = F_{\text{cld+aer}}^\uparrow - F_{\text{cld}}^\uparrow. \quad (1)$$

A radiative transfer model (RTM) is commonly used to, given the atmospheric constituents in the atmosphere, simulate the scene twice; once with and once without the aerosols. To do this for a scene with aerosols overlying a cloud, the optical and physical properties of both the aerosols and the clouds have to be determined, and to a lesser extent the light absorption and scattering properties of the air and the surface reflectance.

The ~~presence of clouds has a strong influence on the DRE at TOA from DRE (at TOA) due to~~ the light absorbing species in smoke is strongly affected by the presence of clouds. Over the dark ocean, in cloud-free scenes, the upwelling radiation at TOA is dominated by the scattering from aerosols and the planetary albedo is increased by the presence of aerosols, resulting in a negative direct effect (cooling). Over clouds, on the other hand, scattering by aerosols hardly contribute to the upwelling radiation at TOA, since the scattering by clouds is dominant. However, the aerosols absorb radiation, lowering the planetary albedo, resulting in a positive direct effect (warming). E.g. an average change in forcing efficiency (DRE divided by AOT) from $-25 \text{ Wm}^{-2}\tau^{-1}$ in cloud-free scenes to $+50 \text{ Wm}^{-2}\tau^{-1}$ in fully clouded scenes was found by Chand *et al.* (2009). The DRE changed sign at a critical cloud fraction of about 0.4 for scenes over the south-east Atlantic Ocean. Similarly, simulations show that the DRE changed sign at a critical cloud optical thickness (COT) of about 4–8, a higher COT resulting in a higher DRE (Feng and Christopher, 2015).

The south-east Atlantic has been a strong focus of modeling and observational studies of the aerosol DRE over clouds. The ocean west of the African continent, where sea surface temperatures are low due to upwelling of cold deep sea water, is covered by a semi-permanent cloud deck. During the austral winter months (July – October), which is the dry season on the adjacent African continent, a myriad of vegetation fires produces immense amounts of smoke ($\sim 25 \text{ Tg}$ black carbon per year), resulting in the largest source of black carbon and natural carbonaceous species in the atmosphere worldwide (van der Werf *et al.*, 2010).

The combination of large areas of boundary layer clouds and overlying smoke proved to be a huge challenge for global climate models (GCMs) to simulate consistent aerosol DRE values at TOA. A comparison of sixteen GCMs showed a large range of aerosol DRE over the south-east Atlantic, from strongly negative (cooling) to strongly positive (warming) for the same experiment (Myhre *et al.*, 2013) (Zuidema *et al.*, 2016), depending on the models' details on cloud and aerosol microphysical properties. It also shows that aerosol radiative effects can be very important on the local scale, near the source areas, even if the contribution to the global radiative budget can be small.

Observations are needed to constrain the model simulations. This can be challenging, because ground observations are sparse and scarce, and satellite observations of COT and aerosol optical thickness (AOT) are difficult to disen-

tangle. Satellite COT observations in the common visible spectral region are biased by absorption by aerosols, resulting in a biased DRE estimation (Haywood *et al.*, 2004; Coddington *et al.*, 2010). Satellite AOT retrievals are commonly performed only in cloud-free scenes, hampering the computation of the aerosol DRE in cloud scenes.

One way of separating cloud and aerosol scattering is the use of active (lidar) instruments, which produce vertically high resolution backscatter profiles, e.g. CALIOP onboard CALIPSO (Chand *et al.*, 2009; Meyer *et al.*, 2013; Zhang *et al.*, 2014). Unfortunately, the spatial coverage of a lidar is limited. Another solution is the use of polarimeter measurements. The different effects of spherical water droplets and irregularly shaped aerosol particles on the polarisation of light can be used to separate the cloud and aerosol contribution to the radiation at TOA. This was applied to POLDER measurements (Waquet *et al.*, 2013a). The absorption from the aerosol layer and the COT is retrieved using reflectances measured in the visible and shortwave infrared. Knowing the COT, and AOT ~~over-of~~ overlying aerosols, the aerosol DRE in cloud scenes can be computed using an RTM twice, simulating the upwelling radiation for the cloud scene with ($F_{\text{cld+aer}}^{\uparrow}$) and without the aerosols ($F_{\text{cld}}^{\uparrow}$). The monthly averaged instantaneous DRE values from POLDER for aerosols over clouds over the south-east Atlantic in August 2006 have been the highest yet, up to 125 was 33 Wm^{-2} (Peers *et al.*, 2015).

The absorption by small smoke aerosols is especially strong in the UV. Several methods use this principle to separate the cloud scattering from the aerosol absorption and scattering. The strong UV absorption can be quantified by the UV Aerosol Index (UV-AI) (de Graaf *et al.*, 2005, 2007; Wilcox, 2012), while the reduction in reflectance in the UV and visible channels can be simulated using LookUp Tables (LUTs). This was used to retrieve AOT of smoke above clouds in the south-east Atlantic, and the COT of the clouds underneath simultaneously, using OMI measurements (Torres *et al.*, 2011). A similar method was applied to MODIS measurements to retrieve AOT and COT simultaneously, using measurements in the visible (Jethva *et al.*, 2013).

These methods all rely on the quantification of the optical properties of the aerosols. However, light absorption by smoke is highly variable and the spectral dependence (quantified by the Ångström parameter) is much larger than often assumed (Jethva and Torres, 2011) and not necessarily unique (Bergstrom *et al.*, 2007). The AOT over clouds in the south-east Atlantic derived from POLDER, CALIOP and MODIS measurements were compared in Jethva *et al.* (2014), showing a general agreement, but large differences in the details.

Spectral information of the aerosol and cloud properties is needed to correctly specify the aerosol-cloud-radiation interactions at all wavelengths. Measurements from six wavelength channels from MODIS (from 0.47–1.24 μm) have

been used to retrieve COT and cloud droplet effective radius (CER) for clouds with overlying aerosols, simultaneously with the above-cloud AOT, and subsequently aerosol DRE (Meyer *et al.*, 2015). However, here also the aerosol spectral properties have to be assumed. To circumvent the use of aerosol optical property models altogether, the spectral dependence of aerosol absorption can be measured with hyperspectral satellite instruments like SCIAMACHY (de Graaf *et al.*, 2012). The principle here is that the absorption by the aerosols is captured entirely by the radiance measurements at TOA in the UV, visible and SWIR spectral regions (measured $F_{\text{cld+aer}}^{\uparrow}$), and only the aerosol-free atmosphere is simulated in an RTM (simulated $F_{\text{cld}}^{\uparrow}$). The cloud properties can be retrieved in the SWIR where small particles like smoke have little to no effect on the COT and CER. The DRE is then retrieved from a difference in simulated and measured reflectance, and the difference is attributed to absorption by aerosols. Hence it is termed differential aerosol absorption (DAA) method. ~~The DRE retrieved in this way monthly averaged instantaneous DRE values from SCIAMACHY for aerosols over clouds over the south-east Atlantic in August 2006 was 23~~ Wm^{-2} . The DRE from SCIAMACHY was compared to Hadley Centre Global Environmental Model version 2 (HadGEM2) climate model simulations, showing that even this GCM, which simulated a large warming over the south-east Atlantic, still fell short in simulating the UV-absorption by smoke (de Graaf *et al.*, 2014). The DAA method was recently applied to a combination of OMI and MODIS reflectance measurements. The monthly averaged instantaneous DRE values from OMI-MODIS for aerosols over clouds over the south-east Atlantic in August 2006 was 25 Wm^{-2} (de Graaf *et al.*, 2019).

The main challenge in comparing satellite data is the wide range in spatial resolution and sampling of different instruments. To resolve this, many papers report area- and time-averaged DRE values and compare them to other average values of the aerosol DRE. In this paper, the DRE derived from POLDER measurements are compared to the DRE from SCIAMACHY and to DRE derived from a combination of OMI and MODIS measurements, accounting explicitly for sampling issues. POLDER reports consistently high values of AOT, COT and DRE compared to other instruments, and we show that the DRE values agree to within the error uncertainty estimates when sampling issues are accounted for and the differences in AOT and COT with other instruments are taken into account.

2 Methods

2.1 POLDER DRE

POLDER is a passive optical imaging radiometer and polarimeter on-board the Polarization and Anisotropy of Re-

fectances for Atmospheric Science coupled with Observations from a Lidar (PARASOL). PARASOL was launched in December 2004 and was part of the A-Train satellite constellation for five years. After 2009, PARASOL's orbit was lowered, and it fully exited the A-Train in 2013. POLDER provides radiances in nine spectral bands between 443 and 1020 nm and polarisation measurements at 490, 670 and 865 nm. The ground spatial resolution is about $5.3 \times 6.2 \text{ km}^2$ and the swath width about 1100 km (Deschamps *et al.*, 1994). All measurements of POLDER are projected on a fixed global reference grid of $6 \times 6 \text{ km}^2$.

The POLDER method retrieves the above-cloud AOT, the aerosol Single Scattering Albedo (SSA) and the COT in two steps. The first one consists of using the polarization radiance measurements to retrieve the scattering AOT and the aerosol size distribution in a cloudy scene. Aerosols affect the polarisation in a cloudy scene in two ways. Firstly, the large peak of the signal around a scattering angle of 140° , caused by the liquid cloud droplets, is attenuated. Secondly, an additional signal at side scattering angles is created. The effect of absorption is assumed to be very weak at these angles and mostly treated as a scattering process. In the second step, the spectral contrast and the magnitude of the total radiances measured in the visible and SWIR are used to retrieve the absorption AOT and COT simultaneously. Therefore, the retrieval of the aerosol properties is done with minimal assumptions and with the cloud properties corrected for the overlying aerosol absorption. To ensure the quality of the products, several filters are applied, which include the removal of inhomogeneous clouds, broken clouds, cloud edges, clouds with COT lower than 3 and cirrus (Waquet *et al.*, 2013b; Peers *et al.*, 2015).

The POLDER DRE is finally calculated over the south-east Atlantic for aerosols over clouds in 2006 using the retrieved AOT, SSA and COT with the method described in section 3 of Peers *et al.* (2015). POLDER apparent O_2 cloud top pressures were used to constrain the cloud layer height, although the cloud top pressure has been shown to have a negligible effect on the TOA radiation (less than 1% for a change of 200 hPa (Ahmad *et al.*, 2004; de Graaf *et al.*, 2012)). CER was derived from collocated MODIS measurements. The DRE was derived for all scenes with a geometric cloud fraction (CF) of 1.0 and a COT larger than 3.0. The surface reflectance was computed taking surface winds (from models) into account (Cox and Munk, 1954), but since only scenes with a minimum COT of 3 were used, the influence of the surface reflectance on the total radiation field will be small. The ozone and the water vapour content were obtained from meteorological reanalysis.

2.2 SCIAMACHY DRE

The DAA method was developed for reflectance spectra from the SCanning Imaging Absorption spectroMeter for Atmospheric CHartography (SCIAMACHY). SCIAMACHY

was part of the payload of the Environment Satellite (EnviSat), launched in 2002, into a polar orbit with an equator crossing time of 10:00 ~~LT for the descending node~~ local solar time in a descending (southward) direction, but stopped delivering data in 2012. SCIAMACHY observed radiation in two alternating modes, nadir and limb, yielding data blocks called states, approximately $960 \times 490 \text{ km}^2$ in size. In nadir mode, SCIAMACHY measured continuous reflectance spectra from 240–2380 nm with a spatial resolution of about $60 \times 30 \text{ km}^2$ and a spectral resolution of 0.2–1.5 nm (Bovensmann *et al.*, 1999). This unique spectral range from the UV to the shortwave infrared (SWIR) contains 92% of the incoming solar irradiance. The DRE was determined from SCIAMACHY reflectance spectra of cloud scenes in 2006 over the south-east Atlantic. Cloud properties were determined at 1.2 and $1.6 \mu\text{m}$, where absorption by smoke is assumed to be negligible. Effective CF and cloud pressure (CP) were determined from (FRESCO) ~~O_2 -A band retrievals~~ Wang *et al.* (2008) O_2 -A band retrievals (Wang *et al.*, 2008). All scenes with effective CF > 0.3 , CP $> 850 \text{ hPa}$ and COT > 3.0 were used to select pixels with sufficient water clouds only. The ocean surface albedo was assumed to have a small, spectrally dependent, constant value. Total ozone was accounted for, but this has a negligible impact on the DRE. See de Graaf *et al.* (2012) for details.

2.3 DRE from combined OMI-MODIS reflectances

The absorption of radiation by aerosols is spectrally dependent, but since the particles vary in size and composition, the spectral dependence is smooth, as opposed to absorption by (trace) gases, which is strongly peaked in absorption lines. Therefore, the DRE data record from SCIAMACHY was continued using a combination of spectrally high-resolution OMI reflectances and low-resolution MODIS reflectances, which are sufficient to capture the spectral dependence of the absorption in the visible and SWIR.

OMI (Levelt *et al.*, 2006), on-board the Aura satellite, was launched in 2004 ~~in a polar orbit, crossing the equator around 13:30 local solar time in an ascending (northward) direction~~, to measure the complete spectrum from the UV to the visible wavelength range (up to 500 nm) with a high spatial resolution, similar to SCIAMACHY. The Earth shine radiance is observed in a swath width of about 2600 km, covering almost the entire Earth in one day. The spatial resolution of OMI is typically about $15 \times 23.5 \text{ km}^2$ at nadir to about $42 \times 126 \text{ km}^2$ for far off-nadir (56 degrees) pixels. Since 2008, OMI suffers from progressive degradation, especially in far off-nadir pixels, called the row anomaly.

MODIS, on-board the Aqua satellite, flies in formation with Aqua-Aura in the A-Train, leading Aqua-Aura by about 15 minutes (in 2006, while PARASOL was placed in between these two instruments). MODIS measures radiances in broad bands (typical about 20–50 nm) from the visible to SWIR, with a typical spatial resolution of 250–500 m. Spec-

trally, OMI overlaps with MODIS at 459–479 nm (central wavelength 469 nm), which can be used to match the OMI reflectances in the visible channel and the MODIS reflectance in band 3 (de Graaf *et al.*, 2016). This way, a continuous low-resolution spectrum at OMI resolution is available to which DAA can be applied (de Graaf *et al.*, 2019).

The DRE was determined from OMI pixels over the southeast Atlantic in 2006. COT and CER were determined at 1.2 and 2.1 μm , because of a reduced sampling in MODIS/Aqua 1.6 μm band due to nonfunctional detectors (Meyer *et al.*, 2015). CP and effective CF are available from OMI $\text{O}_2\text{-O}_2$ retrievals. All scenes with $\text{COT} > 3.0$, effective $\text{CF} > 0.3$ and $\text{CP} > 850$ hPa were selected. The ocean surface albedo was assumed to have a small, spectrally dependent, constant value.

2.4 Error budget

The largest uncertainty for the DRE derives from the assumption that the aerosol-free cloud scene can be simulated using an RTM, which is assumed in all methods. For SCIAMACHY and ~~OMI/MODIS-OMI-MODIS~~ scenes this was actually tested, by applying the technique to measured aerosol-free cloud scenes and determining the DRE, which should be zero by definition. This provides an easy verification of the method. For each instrument and area this can be determined separately, by screening cloud scenes with overlying absorbing aerosol using the aerosol UV index, which is highly sensitive to UV-absorbing aerosols. The (average) deviation of the DRE from zero, determined for aerosol-free cloud scenes, is a good estimate of the uncertainty of the method, which can be substantial. Such an estimate is often missing for other methods, even if this can be done for scenes with small or negligible AOT. Methods other than DAA are moreover highly uncertain due to their dependence on. Moreover, the correct characterization of the spectral properties of the overlying aerosols, while this is circumvented by DAA. Other minor error sources for the DAA method are the uncertainty in input parameters, the influence of the smoke on the estimated cloud fraction, cloud optical thickness and cloud droplet effective radius, an uncertainty in the anisotropy factor (de Graaf *et al.*, 2019), and the uncertainty of estimating the COT and CER at SWIR wavelengths. The error on the aerosol DRE from SCIAMACHY is about 8 Wm^{-2} (de Graaf *et al.*, 2012) and from ~~OMI/MODIS-OMI-MODIS~~ about 13 Wm^{-2} (de Graaf *et al.*, 2019).

The main source of error for the aerosol DRE over clouds from POLDER is the assumption on the aerosol refractive index. In the first step of the algorithm, an assumption on the refractive index is used in order to retrieve the above-cloud scattering AOT. In the second step, the imaginary part is modified in order to retrieve the absorption AOT from total reflectances, assuming the same real part of the refractive index as in the first step. The impact of the

Table 1. Maximum and average values of ~~OMI/MODIS-OMI-MODIS~~, SCIAMACHY, and POLDER DRE on 12 and 19 August 2006 for the areas shown in Figures 1(d–f) and 1(a–c).

12 August 2006	Max DRE	$\langle \text{DRE} \rangle$
POLDER	303.8	109.1
OMI/MODIS-OMI-MODIS	120.0	35.5
SCIAMACHY	112.5	28.4
19 August 2006		
POLDER	190.3	43.0
OMI/MODIS-OMI-MODIS	94.0	11.4
SCIAMACHY	71.3	18.1

refractive index assumption on the DRE has been analysed in (Peers *et al.*, 2015) and a maximum error of 10 Wm^{-2} has been observed. Finally, an error on the CER can cause a bias of up to 10% on the COT.

3 Results

3.1 Case studies in August 2006

The aerosol DRE retrievals over clouds from the various satellite instruments are first introduced in Figure 1 using two cases in August 2006. ~~August is the peak of the biomass burning season in southern Africa, and an extended smoke plume, originating from the African continent, drifted over the south-east Atlantic Ocean in an elevated layer above a stratocumulus deck in the boundary layer. The absorption of radiation by the smoke above the stratocumulus cloud deck is indicated by high DRE values, in cloud scenes only. On the left the situation on 19 August 2006 is given, which shows a good correlation between the instruments. Figure 2006, on the 12th and the 19th. The first case shows the situation during the largest difference between the datasets, the second case the situation one week later, when the differences are moderate. Figures 1a–c show the same data as Figures 1d–f, only one week earlier and from a slightly different area, centered on the MERIS and SCIAMACHY overpass. Figures 1a shows and d show the POLDER DRE overlaid on a MODIS RGB image acquired around 13:10–13:20 UTC, Figure 1b the OMI/MODIS and e the OMI-MODIS DRE over the same MODIS RGB image, and Figure 1c and f the SCIAMACHY DRE overlaid over a MERIS RGB image, both on EnviSat. EnviSat is in a morning orbit, and the SCIAMACHY and MERIS measurements were taken around 9:30 UTC. Clearly, the 30–9:45 UTC. The clouds are more extensive in the latter image, because clouds in this area break up as the day progresses and the solar radiation intensifies (Bergman and Salby, 1996).~~

During 2006 all instruments performed well, and August is the peak of the biomass burning season in southern Africa.

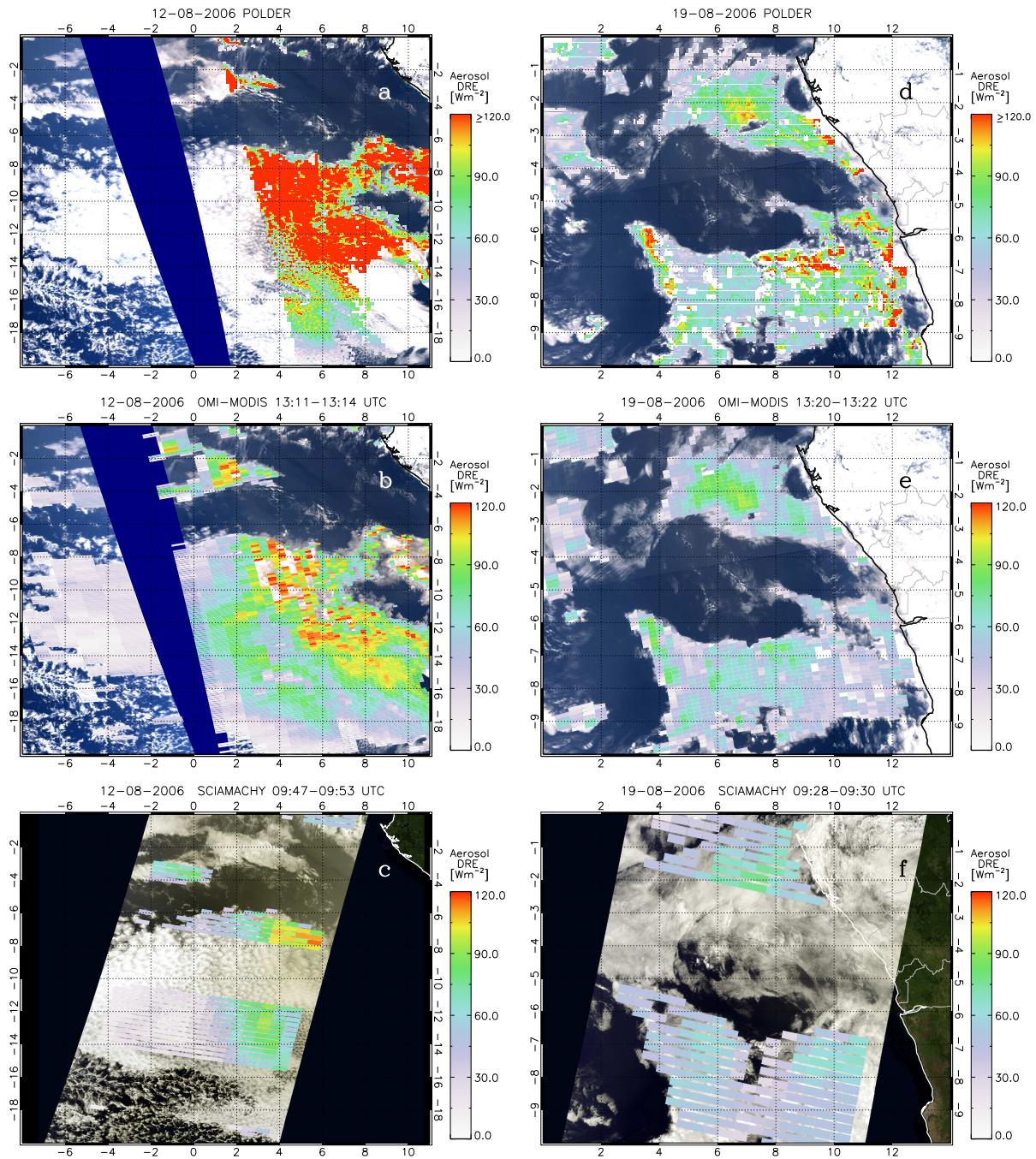


Figure 1. (a) Instantaneous Aerosol Direct Radiative Effect (DRE) over clouds on ~~19~~12 August 2006 from POLDER, overlaid over a MODIS RGB image; (b) Aerosol DRE over clouds on the same day from a combination of OMI and MODIS reflectances, overlaid over the same MODIS RGB image; (c) Aerosol DRE over clouds from SCIAMACHY on the same day, overlaid over a MERIS RGB image; (d–f) same as (a–c) for ~~12~~19 August 2006. The areas are centered over the MERIS/SCIAMACHY overpasses.

An extended smoke plume, originating from the African continent, drifted over the south-east Atlantic Ocean in an elevated layer above a stratocumulus deck in the boundary layer. The absorption of radiation by the smoke above the

stratocumulus cloud deck is indicated by high DRE values, in cloud scenes only.

Obviously, the spatial coverage of SCIAMACHY is much lower than OMI and MODIS, measuring in nadir mode only

half of the time, and having larger pixels. Consequently, the ~~OMI/MODIS~~ OMI-MODIS DRE is smoother with a better coverage. However, the most striking feature is the much higher values from POLDER compared to the other two instruments, even though the general DRE patterns for the three instruments are quite similar. ~~The POLDER DRE~~ On 12 August 2006, the POLDER DRE is very large, reaching values up to 304 Wm^{-2} . The OMI-MODIS DRE reaches up to 120 Wm^{-2} , much lower than the POLDER DRE. The maximum SCIAMACHY DRE was 113 Wm^{-2} . On 19 August 2006 the differences are smaller, but still obvious. The POLDER DRE reaches values up to 190 Wm^{-2} in parts where smoke from the African continent is abundant. The values drop off to zero over clouds where the smoke plume is thinning. The DRE from the two other instruments, on the other hand, is never larger than 100 Wm^{-2} . The DRE values for these cases are summarized in Table 1

~~On the right, the figure shows the situation for~~ Furthermore, a much higher area-averaged POLDER DRE on 12 August 2006, when the comparison between the instrument is worst is found then for the other instruments, which is not only due to higher individual values. Figures 1d–f show the same data as Figures 1a–c, only one week earlier and from a slightly different area, centered on the MERIS and SCIAMACHY d and 1e show that due to a smaller swath compared to OMI and MODIS, POLDER samples an area near the continent that has by coincidence only very high DRE values; the entire left part of the area in Fig. 1a is not sampled. This yields a much higher area-averaged DRE for POLDER than from the other instruments. OMI and MODIS sample the entire basin, where large parts have only very low to zero aerosol DRE values. In the case of SCIAMACHY only about 1/6 of the area is covered by SCIAMACHY nadir measurements, which obviously makes it very sensitive to the sampling of an aerosol plume during one overpass.

~~Obviously, there are clear differences between the retrievals. The POLDER DRE is very large, reaching values up to 304 Wm^{-2} . The OMI/MODIS DRE is larger than~~ Additionally, the SCIAMACHY and OMI large pixel sizes smooth the high DRE values that are found by POLDER. Pixel sizes from SCIAMACHY are about 50 times as large as those from POLDER, which will result in a smoothing of small scale features, like local high values. OMI pixel sizes vary between nadir and the far off-nadir, being about 10 times larger than POLDER at nadir, up to 147 times larger at a viewing zenith angle of 56 degrees.

Lastly, on 19–12 August 2006, reaching up to 120 Wm^{-2} , but still much lower than the POLDER DRE. The SCIAMACHY DRE shows similar values and patterns as OMI/MODIS DRE, but the coverage is rather poor. The maximum SCIAMACHY DRE was 113 Wm^{-2} . 2006 at some places where the highest values of aerosol DRE can be expected, the OMI-MODIS retrievals failed (Figure 1d), probably due to broken cloud scenes in combination with

very high aerosol loadings, which resulted in low scene reflectances which were not marked as clouded scenes. Furthermore, cloud filtering can be different for the three instruments, due to the use of different cloud filters (use of effective or geometrical cloud fractions), which may have a strong influence on the (average) DRE.

The differences between the datasets will be explained below by a closer inspection of the data and the retrievals.

3.2 Area-averaged DRE

~~When~~ Clearly, sampling is an issue that needs to be considered when comparing datasets, and datasets and simulations, sampling is a serious issue. Often, area- and time-averages are compared, to reduce the effects of sampling differences. Here, we show the effect of ignoring the sampling effect, even of area-averages of, in this case, aerosol DRE over clouds differences between instruments.

In Figure 2a, the area-averaged instantaneous aerosol DRE over clouds from all three instruments is given for all available data in the area 10°N – 20°S , 10°W – 20°E , between 1 June and 1 October 2006. This is the biomass burning season and the area where often area-averaged DRE values have been reported during this season (e.g. Chand et al., 2009; de Graaf et al., 2014; Meyer et al., 2013; Peers et al., 2015). Since the instruments have different overpass times, the instantaneous aerosol DRE over clouds was normalized by dividing by the cosine of the solar zenith angle. Therefore, the quantity in Figure 2a represents the instantaneous aerosol DRE at noon for an overhead Sun (at noon), which is generally higher than the instantaneous aerosol DRE measured during the overpass. Figure 2a shows the evolution of the biomass burning season in 2006, with low DRE values in June, high values in July, extreme values in August and moderate values in September.

The area-averaged DRE of smoke over clouds reaches values up to 100 Wm^{-2} and more in mid-August 2006, according to SCIAMACHY and POLDER. The events during this period have been investigated often before (e.g. Chand et al., 2008; Jethva and Torres, 2011; Yu and Zhang, 2013). The SCIAMACHY DRE values were compared to model calculations from GCMs, particularly HadGEM2 (de Graaf et al., 2014). Models were not able to replicate these extremely high aerosol direct radiative effects. The emission of smoke from Africa was possibly strongly peaked in August, but even accounting for such episodic emissions in models did not explain the difference in aerosol effects in models and observations by SCIAMACHY. And Figure 2a shows that the aerosol DRE values from POLDER are even higher than those from SCIAMACHY. On the other hand, the average OMI/MODIS OMI-MODIS DRE is never higher than about 60 Wm^{-2} . The largest difference between the datasets was found on 12 August 2006.

The differences between the instruments are illustrated using histograms of all noon-normalized aerosol

DREs, see Figure 3a. Clearly, the average POLDER aerosol DRE is almost twice as large as that from ~~OMI/MODIS-OMI-MODIS~~ and SCIAMACHY (24.9 Wm⁻² for ~~OMI/MODIS-OMI-MODIS~~, 28.4 Wm⁻² for SCIAMACHY and 46.6 Wm⁻² for POLDER). The statistics of the distributions are given in Table 2. In only the month August, the average aerosol DRE was 27.5 Wm⁻² for ~~OMI/MODIS-OMI-MODIS~~, 36.8 Wm⁻² for SCIAMACHY and 49.7 Wm⁻² for POLDER. This is a somewhat larger difference between POLDER and SCIAMACHY than found by Peers *et al.* (2015) (about 10.5 Wm⁻² difference between SCIAMACHY and POLDER), but there POLDER DRE was averaged over a much larger area containing more small values of DRE.

The histograms show that the DRE from POLDER is higher than the DRE from ~~OMI/MODIS-OMI-MODIS~~ mainly due to more ~~large-high~~ DRE values. This is indicated by the larger positive skewness for POLDER, a measure for the asymmetry of the distribution, where the other instruments show a more symmetric distribution.

~~The main reason for the much larger area-averaged POLDER DRE on 12 August 2006 is the smaller coverage of the area by POLDER, compared to that by OMI/MODIS, due to a smaller swath. This was illustrated in Figures 1d and 1e. In 1d the entire left part of the image is not sampled. In this case, this results in a sampling by POLDER of only the very high DRE values that are found near the continent, while OMI and MODIS sample the entire basin, which has large parts with very low to zero aerosol DRE. In the case of SCIAMACHY only about 1/6 of the area is covered by SCIAMACHY nadir measurements, which obviously makes it very sensitive to the sampling of an aerosol plume during one overpass. OMI and MODIS have a much better coverage, sampling most of the area during an overpass.~~

~~However, OMI pixel sizes vary between nadir and the far off-nadir pixels by a factor of 15. This means that a (dense) plume may be sampled once by a far off-center pixel, or by 15 nadir pixels, all of them receiving the same high values, depending on the satellite track. The pixel sizes of SCIAMACHY are even larger. The POLDER pixels sizes, on the other hand, are constant on a relatively fine 6×6 km² grid. The different pixel sizes produce a difference in the extreme values. Due to the larger pixel sizes, small features in OMI/MODIS and SCIAMACHY will be more smoothed out compared to POLDER data.~~

~~Additionally, on 12 August 2006 at some places where the highest values of aerosol DRE can be expected, the OMI/MODIS retrievals failed (Figure 1d), probably due to broken cloud scenes in combination with very high aerosol loadings, which resulted in low scene reflectances which were not marked as clouded scenes. Furthermore, cloud filtering can be different for the three instruments, due to the use of different cloud filters (use of effective or geometrical cloud fractions), which may have a strong influence on the (average) DRE.~~

Table 2. DRE Statistics of the different instruments before and after collocation for the area 10°N–20°S;10°W–20°E in the south-east Atlantic.

Native grid	Mean	Median	Std. Dev
POLDER	46.60	38.74	39.00
OMI/MODIS-OMI-MODIS	24.88	21.92	30.27
SCIAMACHY	28.42	26.11	24.62
Collocated <u>POLDER grid</u>			
POLDER	46.96-47.11	37.88-38.04	41.01-40.90
OMI/MODIS-OMI-MODIS	34.50-37.13	31.48-33.74	27.69-26.15
SCIAMACHY	39.47-39.50	36.23	24.68-24.66
Collocated <u>OMI grid</u>			
<u>POLDER</u>	<u>43.66</u>	<u>36.30</u>	<u>35.91</u>
<u>OMI-MODIS</u>	<u>35.63</u>	<u>32.29</u>	<u>24.96</u>

3.2.1 Sampling

Clearly, these different spatial scales limit the ~~usefulness~~ usefulness of a comparison of average values from satellite instruments. In order to correct for the issues described above, the ~~OMI/MODIS-OMI-MODIS~~ and SCIAMACHY measurements were regridded onto a regular lat/lon grid, of 6666×3333 grid points. This corresponds to a 6 km × 6 km grid at the equator (reducing to 5.6×5.6 km² at 20°S). All regular grid cells covered by a SCIAMACHY or OMI pixel were given the value of that SCIAMACHY or ~~OMI/MODIS~~ OMI-MODIS DRE measurement. This gave SCIAMACHY and ~~OMI/MODIS-OMI-MODIS~~ OMI-MODIS DRE values on a grid similar to the POLDER grid (albeit smoothed per OMI or SCIAMACHY pixel), ~~so that values can be compared on a pixel-per-pixel basis.~~ The individual POLDER DRE values were then compared to the ~~OMI/MODIS-OMI-MODIS~~ and SCIAMACHY DRE values in the grid cell that was closest to the POLDER grid cell. In Figure 2b the noon-normalised area-averaged instantaneous DRE over clouds over the south-east Atlantic is shown, like in Figure 2a, but using only those pixels that are covered by all three instruments. This effectively removes all sampling issues and differences due to different cloud screening strategies for the instruments. Note that at a number of days no values were available, since there were ~~simply~~ no areas with DRE that are sampled by all three instruments¹. This underlines the importance of sampling, even for such a fairly large area. The number of pixels over which was averaged per day is shown in the lower panel of Figure 2b.

The correlation between the noon-normalised area-averaged instantaneous DRE from the three instruments is now significantly improved compared to Figure 2a. The aerosol DRE from ~~OMI/MODIS-OMI-MODIS~~ follows the aerosol DRE from SCIAMACHY very closely for almost the entire period shown. Note that the maximum DRE from ~~OMI/MODIS-OMI-MODIS~~ is now increased to almost 10090 Wm⁻², which was due to removing many pixels with a moderate to low DRE during mid-August, that were not

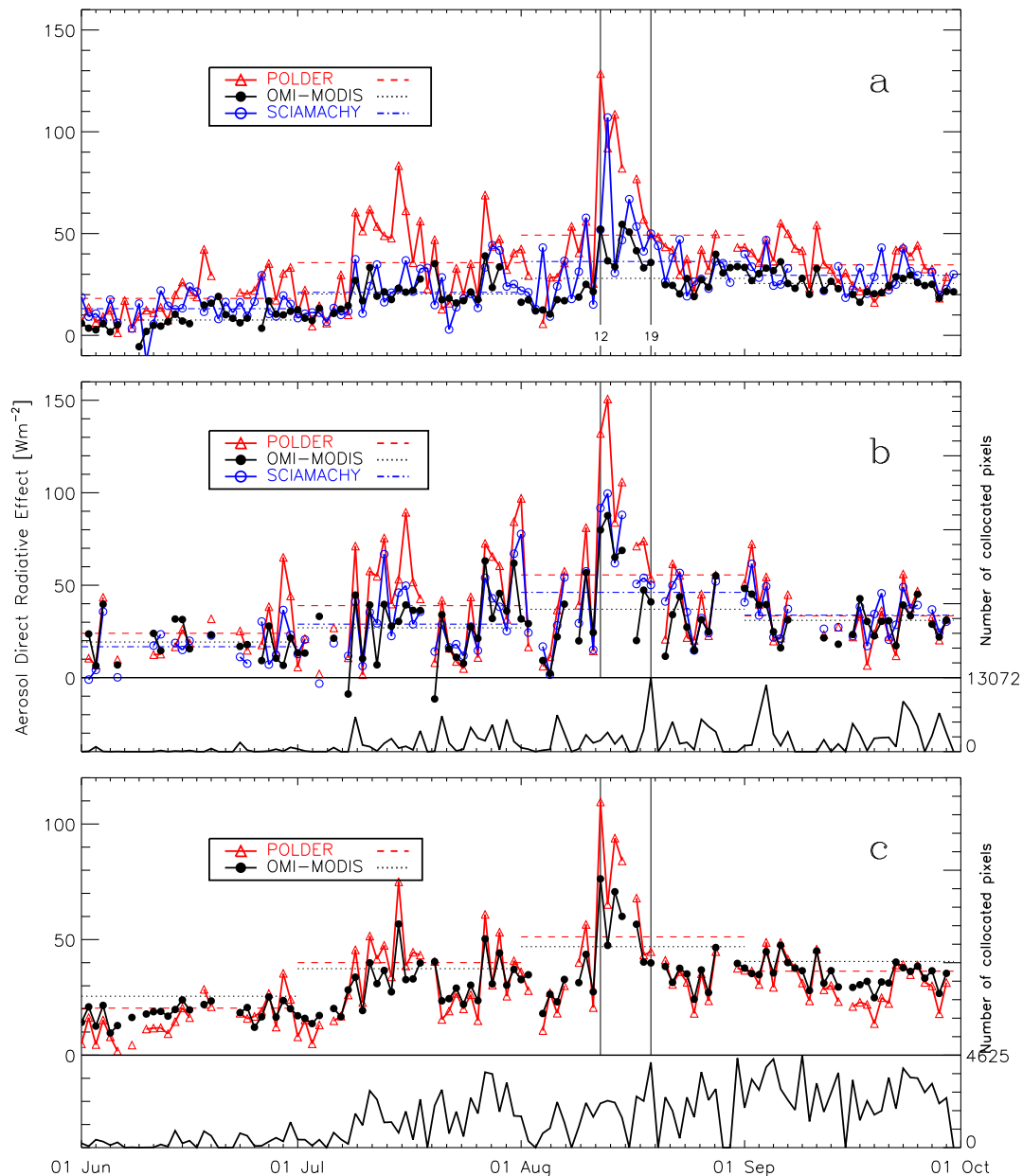


Figure 2. a) Noon-normalized instantaneous aerosol DRE over clouds from combined [OMI/MODIS-OMI-MODIS](#) reflectances (black), SCIAMACHY reflectances (blue) and POLDER AOT and COT retrievals (red) from 1 June - 1 October 2006, averaged over the area $10^{\circ}\text{N}-20^{\circ}\text{S}; 10^{\circ}\text{W}-20^{\circ}\text{E}$ in the south-east Atlantic. The average monthly aerosol DRE over clouds are given by the coloured straight lines during each month. b) Same as a), but for collocated POLDER, regridded [OMI/MODIS-OMI-MODIS](#) and regridded SCIAMACHY pixels only. The number of collocated pixels that are covered by all three instruments is given in the lower panel in b). c) [Area-averaged instantaneous aerosol DRE from OMI-MODIS and POLDER regridded to the OMI footprint](#). Note that because SCIAMACHY is omitted the number of pixels is much larger than in a) and b), and furthermore, the DRE is not noon-normalised, because the overpass time of OMI, POLDER and MODIS are similar.

covered by POLDER and SCIAMACHY, [as illustrated in Figures \(cf. Figures 1d-f-a-c\)](#). Also note that the day with the largest average values does not occur on the 12th but on

[13 August 2006 for all instruments, because SCIAMACHY samples closer to the continent that day, where smoke plumes are generally thicker](#). The difference in average DRE be-

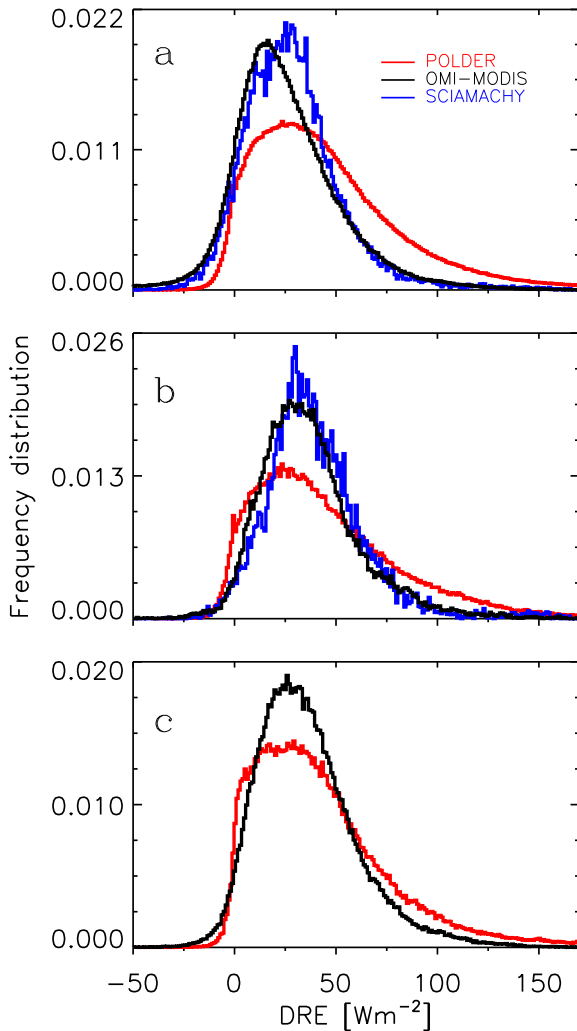


Figure 3. a) Histograms of aerosol DRE over clouds in the Atlantic Ocean during June – September 2006 from POLDER AOT and COT retrievals (red), combined OMI/MODIS OMI-MODIS reflectance spectra (black) and SCIAMACHY reflectance spectra (blue) and POLDER AOT and COT retrievals (red). b) Same as a) but for collocated POLDER, OMI-MODIS regridded OMI/MODIS to POLDER grid and regridded SCIAMACHY regridded to POLDER grid pixels only. c) Same as a) but for collocated POLDER regridded to OMI grid and OMI-MODIS pixels only.

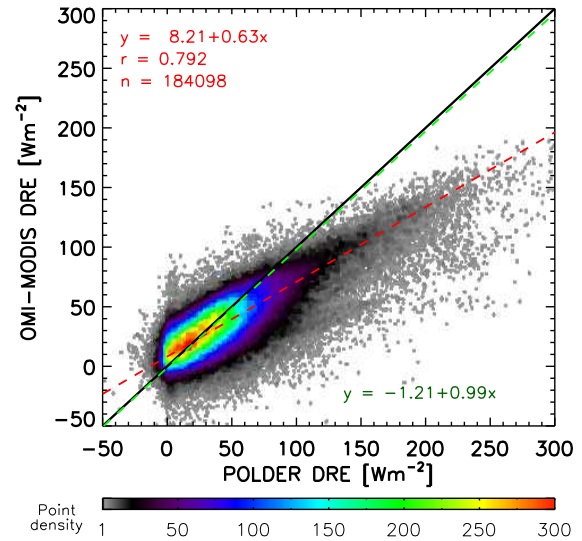


Figure 4. (a) Scatterplot of POLDER DRE gridded to the OMI grid versus DRE from regridded OMI/MODIS OMI-MODIS data from June-September 2006 over the south-east Atlantic. The green red dashed line shows an unweighted linear least-squares fit, the red dashed. The green line shows a the linear least-squares fit weighted by the distance to the majority average value of the points in the center 25 Wm⁻². (b) same as (a), but for the quantity DRE/COT.

tween the instruments is also greatly reduced, see Figure 3b, which shows the histograms for only overlapping regridded pixels pixels regridded to the POLDER grid, and its statistics in Table 2. The average DRE from POLDER is still about 47.0 Wm^{-2} for only overlapping pixels, while the average DRE from OMI/MODIS OMI-MODIS has increased to 34.5 37.1 Wm^{-2} and 39.5 Wm^{-2} for SCIAMACHY regridded pixels.

The skewness of the OMI/MODIS DRE distribution Additionally, the sampling was checked by gridding the finer POLDER data to the coarser OMI grid and sampling only pixels that were covered by both OMI-MODIS and POLDER. In this case SCIAMACHY was omitted, so as not to lose too many POLDER and OMI-MODIS pixels because of the poor SCIAMACHY sampling. Figures 2c and 3c show the area-averaged instantaneous aerosol DRE over clouds from collocated OMI-MODIS and POLDER pixels sampled on the OMI grid, and the histograms of both datasets. The statistics are given in Table 2. Obviously, gridding to the OMI grid instead of to the POLDER grid doesn't change the results very much, but without SCIAMACHY the large number of pixels that are collocated results in a very high consistency between OMI-MODIS and POLDER DRE. Furthermore, without SCIAMACHY the noon-normalisation is no longer necessary because the overpass times of OMI, MODIS and POLDER are very close, and Figures 2c and 3c show the instantaneous

local DRE as retrieved by the instruments, which helps the comparison discussion later on. The figures show that POLDER DRE is still larger than OMI-MODIS DRE, especially for high values, but also lower for low values. This is illustrated by the skewness of the OMI-MODIS DRE distribution, which is now closer to the skewness of the distribution of POLDER DRE, but still the POLDER distribution is dominated by high values, more high and low values.

This is also clear from a scatterplot of collocated POLDER DRE vs. OMI/MODIS OMI-MODIS DRE for regridded OMI/MODIS POLDER pixels, shown in Figure 4a. Note that for this plot only a POLDER and OMI/MODIS overlap was required, which yielded a significantly higher number of pixels than when also SCIAMACHY overlap was required. The figure shows a good correlation of aerosol DRE for most measurements, but at high values of the DRE, the OMI/MODIS DRE is systematically underestimated compared to POLDER DRE. The between collocated POLDER and OMI-MODIS DRE, but with higher values for POLDER, especially for DRE larger than 100 Wm^{-2} . An average ratio of OMI/MODIS OMI-MODIS DRE to POLDER DRE is of 0.82 can be found from Table 2, while a normal linear least-squares fit (shown by the green-red line in Figure 4a) yields a slope of OMI/MODIS OMI-MODIS to POLDER ratio of only 0.560.63. This is because the fit is dominated by the large values, while the large majority of points are moderate values around 25 Wm^{-2} . When a fit is drawn which is weighted to the majority of the points deviation from this moderate value (shown by the red-green line), a slope of 0.74 0.99 is found, which is closer to the average ratio showing that the aerosol DRE over clouds is the same from POLDER and OMI-MODIS for moderate values.

SCIAMACHY DRE is very similar to OMI/MODIS OMI-MODIS DRE for all pixels sampled by these instruments (not shown).

Several reasons for the smaller SCIAMACHY and OMI/MODIS DRE compared to POLDER DRE exist. First, a sampling issue still remains, since the regular grid cells contain DRE values from larger OMI and SCIAMACHY pixels. Therefore, the high resolution grid cells receive smoothed values. This issue could be resolved if all values were regridded to the coarsest available. However, since this is the SCIAMACHY grid, not many grid cells would remain.

3.2.2 AOT differences

Another explanation is an underestimation of

3.3 Effects of differences in AOT and COT

To explain the difference between the OMI-MODIS DRE and the POLDER DRE, the differences in AOT and COT retrieved by the different instruments and their effects on the DRE are investigated. The effects on the DRE of a difference

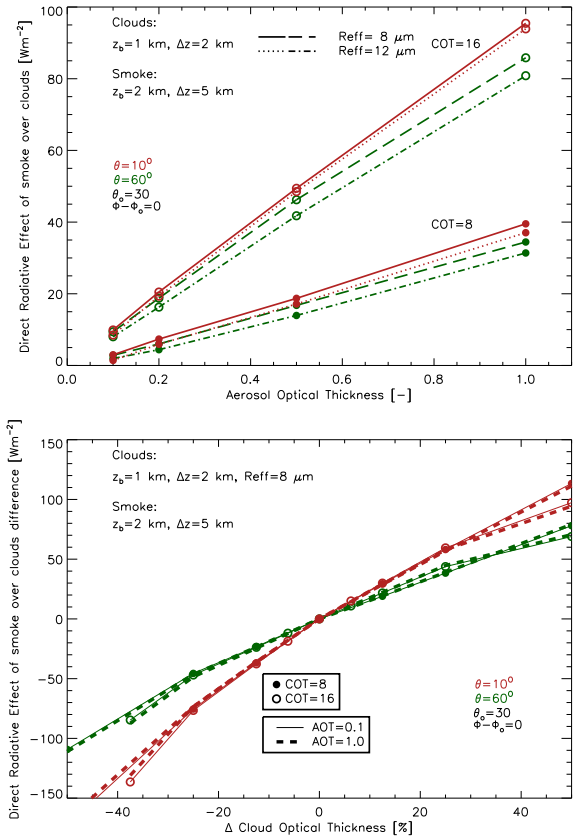


Figure 5. (a) Aerosol DRE for simulated scenes with clouds between 1-2 km and smoke aerosol between 2-5 km as a function of AOT at 550 nm. The COT was 8 or 16, the effective cloud droplet radius 8 or $12 \mu\text{m}$. SZA was 30° , VZA was 10° or 60° , RAZI was 0° . (b) Aerosol DRE for simulated scenes as in (a), as a function of relative error in the retrieved COT.

in AOT and COT are first investigated using simulated TOA spectra scenes of aerosols above clouds. An RTM was used to simulate the aerosol DRE at TOA of a scene with varying AOT, above a cloud deck with varying COT. For the simulations, a cloud was placed between 1 and 2 km and an aerosol layer between 2 and 5 km altitude. The clouds were simulated assuming a single-mode gamma particle size distribution with effective radius $r_{\text{eff}} = 16 \mu\text{m}$ and an effective variance $\nu_{\text{eff}} = 0.15$. For the aerosols, a bi-modal log-normal size distribution model was used, based on the ‘very aged’ biomass plume found over Ascension Island during SAFARI 2000 (Haywood et al., 2003). A refractive index of $1.54 - 0.018i$ was used for all wavelengths longer than 550 nm. However, for the UV spectral region the imaginary refractive index was modified so that the absorption Ångström exponent was 2.91 in the UV, which fits satellite observations better (Jethva and Torres, 2011). The geometric radii for this haze plume used in the simulations

were $r_c = 0.255 \mu\text{m}$ and $r_f = 0.117 \mu\text{m}$ for the coarse and fine modes, with standard deviations $\sigma_c = 1.4$ and $\sigma_f = 1.25$, respectively. The fine mode number fraction was 0.9997. These numbers are the same as used in de Graaf *et al.* (2012) to estimate the anisotropy change in the DRE calculation and in de Graaf *et al.* (2019) to study the BRDF of a scene with the aerosols above clouds.

The effects of varying AOT and varying COT on the aerosol DRE over clouds are illustrated in Figure 5a for an AOT between 0.1 and 1, and a COT of 8 and 16, with cloud effective radii of 8 and $12 \mu\text{m}$. The solar zenith angle (SZA) in the simulations shown was 30° , the relative azimuth angle (RAZI) was 0° and two viewing zenith angles of 10° and 60° are shown, which span a typical range of viewing angles for OMI. The figure clearly shows a linear relationship between AOT and DRE, with an increasing aerosol DRE with increasing AOT, as expected. However, as known, the increase in DRE with AOT depends mainly on the COT of the underlying clouds. With larger COT, the amount of light at TOA increases, and the amount of absorption by the aerosols above the clouds also increases, increasing the DRE. Clearly, the effect of AOT by SCIAMACHY and OMI/MODIS and a possibly overestimation of the AOT by POLDER. The POLDER DRE is dependent on the retrieved AOT and COT, which in principle are both unbounded. When the algorithm retrieves very large values for both, the derived DRE can also become very large. In mid-August, DRE above 300 on DRE are coupled. At a still relatively modest COT of 16, an increase of AOT from 0.1 to 1 increases the DRE from 10 to 95 Wm^{-2} were often reached, up to more than 400, for high AOT of 1, a doubling of COT from 8 to 16 increases the DRE from 40 to 95 Wm^{-2} .

Accurate AOT and COT retrievals are clearly essential for an accurate aerosol DRE over clouds. For DAA, the effect of an error in the COT is estimated using simulated reflectances as above, shown in Figure 5b. Irrespective of AOT or COT, an error of 20% in COT can lead to an error in DRE of 50 Wm^{-2} for the SZA-corrected DRE. This is 30% of the maximum incoming solar irradiance. The POLDER AOT. A note for DAA is in order here: because the simulated aerosol-free cloud spectrum is computed from the COT and CER retrieved from the measured aerosol-cloud spectrum, the spectra are the same in the SWIR, cancelling retrieval errors in the COT. A test with OMI-MODIS spectra and POLDER COT regridded to the OMI grid yielded erratic DRE, even though the POLDER COT retrieved in the visible is probably superior to the OMI-MODIS COT retrieved in the SWIR. However, in such a case a simulated cloud spectrum using POLDER COT is often different from the spectrum by MODIS in the SWIR, yielding aerosol effects even without aerosols. For the POLDER DRE calculation this effect is different, because the DRE is computed using the scene twice with the same retrieved COT.

3.3.1 AOT differences

A part of the difference in DRE between OMI-MODIS (and SCIAMACHY) and POLDER can be attributed to differences in AOT, even if AOT is not explicitly retrieved for OMI-MODIS and SCIAMACHY. However, it is assumed to have negligible effect in the SWIR from about $1.2 \mu\text{m}$, which may be an underestimation (in AOT). POLDER AOT, on the other hand, can be overestimated. Waquet *et al.* (2013a) estimated an overestimation of AOT for mineral dust above clouds of about 6% due to plane parallel RTM computations, for smoke no estimate was given. Comparisons between AOT above clouds for smoke from several instruments show POLDER to be consistently on the high side, although not necessarily overestimated. On 12 August 2006, POLDER AOT at 550 nm on 12 August 2006 was 1.1, averaged over the entire area, with individual values up to 1.9, which is extremely high. However, high AOT for this plume was found before. Chand *et al.* (2009) found. From CALIOP data an AOT of up to 1.5 (532 nm) using CALIOP data, was found for this day in the same area (Chand *et al.*, 2009), while Jethva *et al.* (2013) found above-cloud AOT observed from MODIS up to 2.0 for this day. A comparison between AOT above clouds from several instruments (Jethva *et al.*, 2014) showed POLDER to be on the high side, but not necessarily strongly overestimated. Comparisons with CALIOP observations revealed that the CALIOP. On 13 August 2006, the maximum OMI above cloud AOT was about 1.3, the maximum MODIS above cloud AOT about 1.5, the same as for POLDER (Jethva *et al.*, 2014). Also from this day, POLDER above-cloud AOT from the operational retrieval are underestimated, while the were about 11% higher than the otherwise well correlated above-cloud AOT from POLDER and the CALIOP depolarisation ratio method are well correlated (Deaconu *et al.*, 2017).

When aerosols are mixed into the cloud layer the polarization signal may be enhanced, and POLDER AOT may possibly be high-biased. Also, when the smoke has a high real refractive index ($m_r > 1.47$) the AOT is overestimated by POLDER (Peers *et al.*, 2015). However, it has been shown that the real part of the refractive index has mostly an impact on the scattering AOT. In the case of biomass-burning aerosols above clouds, the absorption AOT, which is retrieved by POLDER with a better accuracy, has a larger influence on the DRE calculation.

The DRE from SCIAMACHY and OMI/MODIS is limited to about 200–250 (Deaconu *et al.*, 2017). The effect of a high AOT can have a large effect on DRE for a high COT according to Figure 5a. The average POLDER COT on 12 August 2006 was 12.9. A 6% overestimation in AOT can lead to an overestimation in DRE of about 10 Wm^{-2} for individual pixels. Since the DRE for these instruments is determined by assuming an AOT of zero at $1.2 \mu\text{m}$ and calculated as a fraction of the incoming local irradiance, it

is unlikely to reach extremely high values. This assumption of that COT.

For SCIAMACHY and OMI-MODIS, on the other hand, the DRE is computed assuming negligible AOT at longer wavelengths, which is valid for sufficiently small particles, but. This assumption may break apart at very high AOT, or for larger particles. The AOT at, leading to an underestimation of the AOT, especially for higher values, limiting the DRE. The assumption can be tested by estimating the $1.2 \mu\text{m}$ may be estimated from the POLDER AOT from POLDER AOT retrieved at 550 nm AOT, using an Ångström parameter of 1.45, which was found in the spectral region from 325 to 1000 nm for African biomass burning aerosols from SAFARI 2000 observations (Bergstrom et al., 2007; Russell et al., 2010). This way, an AOT at $1.2 \mu\text{m}$ can be found between 0.15 and 0.35 was found during the smoke peak in mid-August 2006, occasionally even reaching 0.6 (Schulte, 2016). The effect on DRE of neglecting

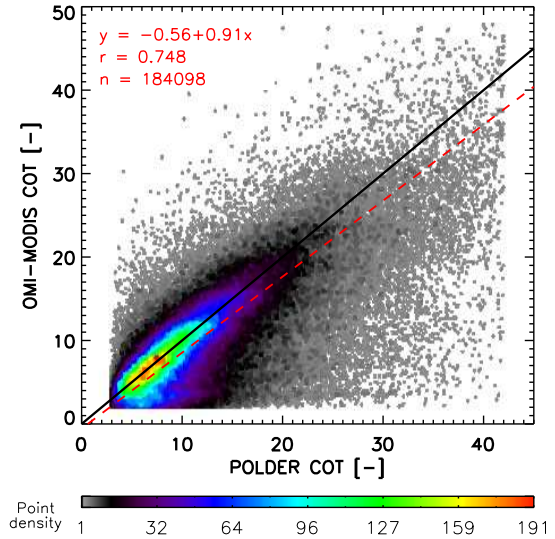


Figure 7. Scatterplot of POLDER COT gridded to the OMI footprint versus COT from OMI-MODIS data. The red dashed line shows the linear least-squares fit.

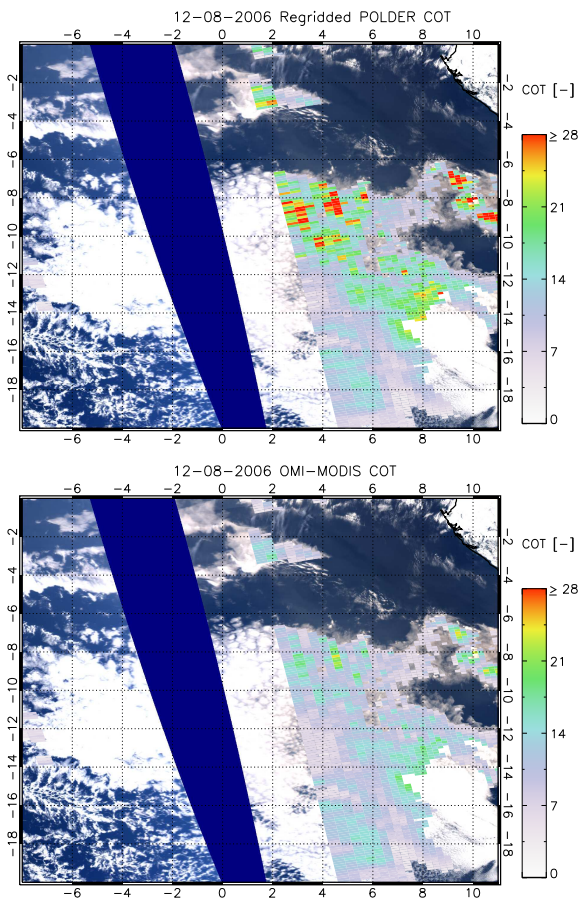


Figure 6. Cloud optical thickness (COT) overplotted on a MODIS RGB image on 12 August 2006 for POLDER at $0.87 \mu\text{m}$ regressed to OMI grid (upper panel) and OMI-MODIS at $1.2 \mu\text{m}$ (lower panel).

a non-zero AOT at $1.2 \mu\text{m}$ in DAA the DRE was estimated at $21.7 \text{ Wm}^{-1} \tau^{-1}$ (de Graaf et al., 2012), so these, by correcting the retrieved COT for the additional AOT (de Graaf et al., 2012), since this effect is essentially an underestimation of the COT. The effect is linear in AOT so the AOTs given by POLDER would result in an underestimation of the DRE by both SCIAMACHY and OMI/MODIS OMI-MODIS of up to 13 Wm^{-2} . This would explain about 26% of the difference of up to 50 Wm^{-2} between the DRE from POLDER and SCIAMACHY or OMI/MODIS shown in Figure 2.

3.3.2 COT differences

The DRE depends more strongly dependence of DRE on the COT of the cloud underlying the smoke than can be large, depending on the AOT of the smoke. A comparison of SCIAMACHY, OMI/MODIS and POLDER COT histograms (not shown) revealed a slightly higher COT from SCIAMACHY and OMI/MODIS compared to POLDER (up to 42 for POLDER and 48 for OMI/MODIS (Schulte, 2016)), but the maximum of POLDER is restricted due to LUT limits. Figure 6 shows the COT retrievals from POLDER and OMI-MODIS on 12 August 2006, when the differences were the largest. POLDER COT is clearly higher peaked than OMI-MODIS COT. SCIAMACHY results are not shown, but similar to those from OMI-MODIS, although the COT may also be different because of the overpass by SCIAMACHY in the morning, when the cloud cover is systematically thicker than in the afternoon.

Note that POLDER COT is retrieved at 0.87 μm , while COT from ~~OMI/MODIS and SCIAMACHY~~ OMI-MODIS is retrieved at 1.2 μm , which effectively is the MODIS channel. However, the spectral variation in COT is very small. Only and is only significant for very small cloud droplets. For example, for cloud droplet effective radii of 4 microns the COT at 0.87 μm is about 4% smaller than the COT at 1.2 μm and this reduces for larger droplets. For SCIAMACHY, the COT may also be different because of its morning overpass, when the cloud cover is systematically thicker than in the afternoon. In Figure 6, the COT from POLDER is compared to the

A comparison between MODIS and POLDER COT is presented in Figure 7 for all collocated OMI-MODIS and POLDER pixels regridded to the OMI footprint from June to September 2006. It shows that the COT from POLDER correlates well with the COT retrieved by MODIS but is higher by about 9% on average. Here, POLDER COT is regridded, while the MODIS radiances are averaged in the OMI footprint and one COT is retrieved for that OMI pixel. Therefore, the difference in COT derived from OMI/MODIS on 19 presented here can be caused by the plane parallel bias, which arises due to cloud heterogeneities and the nonlinear dependence of the cloud albedo (or reflectance) on water content (or optical thickness) (e.g. Oreopoulos and Davies, 1998). This effect is particular important in marine stratocumulus, which appear as plane parallel clouds, but are characterised by strong internal turbulent variability. Internal variability is largest at a cloud fraction of one and can have a stronger effect on the average (meso-scale) cloud optical thickness estimates than cloud fraction itself for marine stratocumulus clouds (e.g. Cahalan *et al.*, 1994).

Again, the POLDER retrieval (of COT) is high, but not necessarily overestimated. An overestimation of 9% at an average COT of about 13 and an AOT of 0.94 on 12 August 2006, regridded to a $6 \times 6 \text{ km}^2$ regular grid. It shows that the high DRE values in Figure 1 are highly correlated with high COT values (which makes sense, since the AOT varies rather smoothly over the area) 2006 would lead to a change of about 9 Wm^{-2} . The average difference in COT is within the error estimate of the POLDER COT, but is more likely the result of the plane parallel bias. However, it also shows that the POLDER COT peaks are much larger than those from OMI/MODIS. Even though the OMI/MODIS data are regridded to a high resolution grid, the values are obviously still more smoothed compared to the COT on the native high resolution POLDER grid. Therefore, even though POLDER COT and POLDER DRE are generally smaller than from OMI/MODIS on average, the extreme values and averages are higher.

To illustrate Finally, the effect of a higher different COT on the DRE retrieval, the average was also computed using an RTM as used for the POLDER DRE calculations for

Table 3. Average values of the ~~OMI/MODIS~~ OMI-MODIS COT and the POLDER COT and above-cloud AOT on 12 and 19 August 2006 between $20\text{--}0^\circ\text{S}; 8^\circ\text{W--}14^\circ\text{E}$. The DRE was calculated at noon using the average AOT from POLDER and the average COT values for both OMI-MODIS and POLDER, assuming a cloud droplet effective radius-CER of $8 \mu\text{m}$, an aerosol SSA of 0.840 at 550 nm and an aerosol geometric radius of $0.1 \mu\text{m}$.

	Max COT	$\langle \text{COT} \rangle$	$\langle \text{COT} \rangle$	$\langle \text{AOT} \rangle$	POLDER
OMI GRID	COT			AOT	
12 August 2006					
POLDER	42.0	41.6	14.8	12.9	0.938
OMI/MODIS	24.1	37.5	10.1	10.0	
19 August 2006					
POLDER	42.0	41.6	11.8	10.5	0.477
OMI/MODIS	47.7		9.4	8.9	

the average values on the two selected days. The average COT from both POLDER and OMI/MODIS on the case studies OMI-MODIS on 12 and 19 August 2006 for collocated pixels were compared. Then, determined and the above-cloud DRE have been calculated for OMI/MODIS was calculated for OMI-MODIS and POLDER using their mean COT and the mean AOT retrieved by POLDER. Results are summarized in Table 3. On 12 August 2006, the mean COT from OMI/MODIS was 9.4 (max. 48), while from POLDER the mean COT was 11.8 (max. average COT from POLDER was 12.9, and from OMI-MODIS 10.0, while the average POLDER AOT was 0.94. This results in a DRE of 42). Based on a mean AOT of 0.578, an aerosol DRE over clouds of 83134 Wm^{-2} and 67 for POLDER and 107 Wm^{-2} have been obtained from the average COT for OMI-MODIS, while the average DRE on this day was 110 Wm^{-2} from POLDER and OMI/MODIS, respectively. On 12-76 Wm^{-2} from OMI-MODIS. On 19 August 2006, the average mean COT from POLDER was 14.8, and from OMI/MODIS 10.1, while the average POLDER AOT was 1.25. This results in a DRE of 18410.5, while from OMI-MODIS the mean COT was 10.5. Based on a mean AOT of 0.48, an aerosol DRE over clouds 63 Wm^{-2} for POLDER and 134 and 53 Wm^{-2} for OMI/MODIS. This suggest was obtained from POLDER and OMI-MODIS, respectively, while the average values for that day were 45 and 40 Wm^{-2} , respectively. Although the simulated average DRE is quite a bit larger than the average DRE found by the instruments, it suggests that the COT difference can completely account for account for about 80% of the difference of up to 5033 Wm^{-2} between the DRE from POLDER and SCIAMACHY or OMI/MODIS or OMI-MODIS shown in Figure 2.

To show the effect of the COT on the DRE retrieval, in Figure 4b the DRE divided by the COT is shown for OMI/MODIS vs. POLDER. It shows that the difference between these two quantities disappears completely for these instruments, and the slope is even reversed. Clearly, the COT

has the largest impact on the computation of the aerosol DRE over clouds. Since the POLDER instrument retrieves higher values of COT in smaller pixels, the DRE is subsequently higher.

The errors in AOT and COT are not independent. In the DAA method, when the assumption of negligible AOT at longer wavelengths is no longer valid (large concentration of aerosols and/or large particles), the estimated COT is biased, resulting in a bias in the DRE. A better estimate of the DRE from the DAA method could be obtained when an unbiased retrieval of the COT was used, like e.g. from POLDER.

Cloud optical thickness (COT) overplotted on a MODIS RGB image on 19 August 2006 for (a) POLDER at 550 nm and (b) OMI/MODIS at 1.2 micron, regridded to $6 \times 6 \text{ km}^2$ grid boxes.

4 Conclusions

In this paper, the aerosol direct radiative effect product is presented for cloud scenes in the south-east Atlantic, retrieved from SCIAMACHY reflectances, combined reflectance measurements from OMI and MODIS, and POLDER COT and AOT measurements in 2006. During this year, the production of smoke from vegetation fires in Africa was very large, and all instruments performed well. The average DRE from SCIAMACHY and OMI/MODIS/OMI-MODIS, both retrieved using DAA, correspond/correlate very well, even though OMI/MODIS/OMI-MODIS DRE has a much better resolution and coverage. The aerosol DRE from POLDER is completely independent. It correlates well with SCIAMACHY and OMI/MODIS/OMI-MODIS DRE for moderate values, but is larger/higher than SCIAMACHY and OMI/MODIS/OMI-MODIS DRE for high values. This is caused by a larger COT retrieved by POLDER, and to a lesser degree by an underestimation of the aerosol DRE using DAA, which by definition assumes a zero AOT at SWIR wavelengths. The POLDER DRE is dependent on the retrieved AOT and COT, which in principle are both unbounded (although in the LUT for POLDER the COT is limited to 42). When the algorithm retrieves very large values for both, the derived DRE can also become very large. In mid-August, DRE above 300 Wm^{-2} were often reached, up to more than 400 Wm^{-2} for the SZA-corrected DRE. This is 30% of the maximum incoming solar irradiance. The DRE from SCIAMACHY and OMI-MODIS is limited to about $200\text{--}250 \text{ Wm}^{-2}$ for individual pixels.

The largest contribution to the difference between SCIAMACHY, OMI/MODIS/OMI-MODIS and POLDER DRE are sampling issues. Regridding SCIAMACHY and MODIS/OMI to the native POLDER grid and selecting only pixels sampled by all three instruments improved the comparison considerably. This approach removes issues related to selecting high positive DRE values by filtering on COT and CF, which introduce large differences in the av-

erage DRE. Only Even if the same filtering is used for the CF and COT for all instruments, different areas will be sampled, because the CF and COT retrieved by the different instruments may be different. After sampling, only smoothing due to the large footprints of SCIAMACHY and OMI remains, which is reflected in the less extreme COT and DRE values compared to POLDER.

After removing sampling issues, the largest remaining differences in DRE are caused. This difference was removed by gridding POLDER to the coarser OMI grid, improving the comparison between OMI-MODIS DRE and POLDER DRE. Because SCIAMACHY was not considered in this analysis, the statistics were much better than when SCIAMACHY collocation was required, because SCIAMACHY's spatial coverage is rather poor. The largest average difference after removing sampling issues between OMI-MODIS and POLDER DRE was 33 Wm^{-2} on 12 August 2006, which can be explained by different estimates of the AOT and COT using the various instruments. Since the bright background of clouds determines the measured reflectance to a very large degree, the DRE is strongly dependent on the COT. COT can change on small spatial scales. This is reflected in the higher positive skewness of the POLDER DRE. The POLDER DRE distribution is less symmetric with larger tails than those from SCIAMACHY and OMI/MODIS, due to the high spatial resolution of the-

In DAA, the AOT is assumed to be zero at $1.2 \mu\text{m}$, but was estimated from POLDER to be up to 0.6 in extreme cases, which results in an underestimation of the DRE in DAA of 13 Wm^{-2} . For POLDER AOT, comparisons with OMI, MODIS and CALIOP AOT over clouds in the literature consistently show POLDER to be on the high side. POLDER AOT may be high-biased when aerosols are mixed into the cloud layer, enhancing the polarization signal. Also, when the smoke has a high real refractive index ($m_r > 1.47$) the AOT is overestimated by POLDER. However, the real part of the refractive index mostly impacts the scattering AOT (Peers *et al.*, 2015), while the DRE calculation, in the case of biomass burning aerosols above clouds, is influenced mainly by the absorption AOT. The underestimation of the AOT for high values can explain about a third of the difference in DRE between POLDER and OMI-MODIS on 12 August 2006, an overestimation of AOT by POLDER is difficult to establish.

The COT has a strong influence on the aerosol DRE over clouds. The average POLDER measurements. The POLDER COT is systematically about 9% higher than that from OMI/MODIS and SCIAMACHY/OMI-MODIS in 2006. This difference can be caused by the plane parallel bias. Normally, MODIS COT retrievals at 0.8 and 1.2 microns retrievals are very 2.1 μm retrievals are close to POLDER COT for fully clouded scenes with liquid water clouds (Zeng *et al.*, 2012) (not considering overlying smoke). However, to avoid biases from smoke absorption, the MODIS channels at 1.2 and 2.1 microns μm are used to derive COT and CER for OMI/MODIS DRE retrievals. In our retrievals the MODIS

reflectances at a resolution of $1^\circ \times 1^\circ$ are first aggregated to the OMI spatial grid, and for this analysis regridded back to the POLDER grid. This will smooth extreme values. When the DRE was divided by the retrieved COT, the difference between the instruments is reversed, OMI/MODIS DRE/COT being larger than POLDER DRE/COT. This shows that a correct COT is essential for the determination of the direct radiative effect of aerosols above clouds. The difference in average COT from OMI/MODIS and POLDER can explain 100% OMI-MODIS DRE retrievals, which may further influence the results. The difference between COT from OMI-MODIS and POLDER on 12 August 2006 can explain about 80% of the difference in DRE on 12 August 2006, that day.

The AOT is assumed to be zero at 1.2 microns in DAA, but was estimated from POLDER to be up to 0.6 in extreme cases, which resulted in an underestimation errors in AOT and COT are not independent. In DAA, when the assumption of negligible AOT at longer wavelengths is no longer valid (large concentration of aerosols and/or large particles), the estimated COT is biased, resulting in a bias in the DRE. A better estimate of the DRE in DAA of 13 Wm^{-2} . Comparing AOT over clouds POLDER with MODIS and CALIOP, showed POLDER to be high, but not necessarily overestimated. The underestimation of the AOT for high values can explain about 26% of the difference in DRE between POLDER and OMI/MODIS on 12 August 2006, from the DAA method could be obtained when an unbiased retrieval of the COT was used, like e.g. from POLDER. However, a test using this approach using DDA on OMI-MODIS spectra using POLDER COT yielded very erratic results. The reason is that the aerosol-free clouds spectrum simulated with POLDER COT can be quite different from the OMI-MODIS measured spectrum, yielding spectral differences that are interpreted as aerosol absorption. This may be improved if POLDER radiances were used.

This analysis shows that the aerosol direct effect of aerosols above clouds can be significant on the local scale when smoke is present over clouds. So far, model simulations have been unable to reproduce the high values, and many models underestimate the signal and even simulate a cooling (Myhre *et al.*, 2013) (Zuidema *et al.*, 2016), where the datasets in this analysis clearly show that the positive effect is significant and real. However, when observations and model simulations of local effects are compared, sampling issues should be properly accounted for, because area-averaging and time-averaging does not work well for episodic events like smoke plumes, which are short-lived and localized.

The analysis also shows the strengths and weaknesses of the DRE retrieval algorithms for POLDER, SCIAMACHY and OMI/MODIS. Clearly, the latter two still suffer from a From the analysis here, we conclude that the aerosol DRE from OMI-MODIS and SCIAMACHY are likely

underestimated, due to the bias in the cloud parameter retrieval when smoke is abundant, providing a lower limit of the aerosol DRE over clouds. POLDER DRE, POLDER on the other hand, takes advantage of the polarization measurements to accurately estimate the COT, CER and AOT, without interdependent biases. However, for the spectral dependence of the aerosol absorption in the UV, there is still a dependence on the choice of aerosol model, and the aerosol DRE from POLDER may be on the high side. The two datasets from POLDER and OMI-MODIS most likely provide a high and low bound for the aerosol DRE in the south-east Atlantic, respectively, which can be used to challenge GCMs and test their aerosol intrinsic properties and aerosol-cloud-radiation interaction schemes. However, when observations and model simulations of local effects are compared, sampling issues should be properly accounted for, because area-averaging and time-averaging do not work well for episodic events like wildfire smoke plumes, which are short-lived and localized.

The analysis has shown the strengths and weaknesses of the DRE retrieval algorithms for POLDER, SCIAMACHY and OMI-MODIS. A combination of the two methods, DAA and DRE based on polarization measurements, could provide very accurate measurements of aerosol DRE over clouds, which is feasible for upcoming missions like METOP-SG 3MI (Marbach *et al.*, 2015). This mission combines spectral imaging METOP-SG-A and B (Marbach *et al.*, 2015). These missions combine spectral imaging from a UV-VIS spectrometer Sentinel-5 and polarization measurements from a multi-angle polarimeter 3MI on one platform. The DAA method would benefit from unbiased COT retrievals, that could be provided with polarization measurements. The assumptions on the spectral dependence of the aerosol absorption in the POLDER-like retrieval can be assessed and improved by the DAA method in a closure study using the instruments on the METOP-SG 3MI platform platforms. This would allow time-dependent retrievals of UV-absorption by aerosols above clouds.

The POLDER, SCIAMACHY and OMI/MODIS DRE products provide datasets that can be used to challenge GCMs and test their aerosol intrinsic properties and aerosol-cloud-radiation interaction schemes.

Data availability. The data used in this study are available from the authors. OMI-MODIS DRE are available online <https://doi.org/10.21944/omi-modis-aerosol-direct-effect>

Author contributions. MdG, LGT and PS are responsible for the DRE datasets from SCIAMACHY and OMI-MODIS. FP and FW are responsible for the POLDER dataset. RS initially compared the datasets.

Competing interests. The authors declare no competing interests.

Acknowledgements. The work by MdG was funded by the Dutch National Programme for Space Research User of the Netherlands Space Office (NSO), project number ALW-GO/12-32.

5 References

- Ackerman, A. S., Toon, O. B., Stevens, D. E., Heymsfield, A. J., Ramanathan, V., and Welton, E. J.: Reduction of Tropical Cloudiness by Soot, *Science*, 288, 1042–1047, <https://doi.org/10.1126/science.288.5468.1042>, 2000.
- 10 Ahmad, Z., Bhartia, P. K., and Krotkov, N.: Spectral properties of backscattered UV radiation in cloudy atmospheres, *J. Geophys. Res.*, 109, D01201, <https://doi.org/10.1029/2003JD003395>, 2004.
- Bergman, J. W. and Salby, M. L.: Diurnal Variations of Cloud Cover and Their Relationship to Climatological Conditions, *J. Climate*, 9, 2802–2820, [https://doi.org/10.1175/1520-0442\(1996\)009<2802:DVOCCA>2.0.CO;2](https://doi.org/10.1175/1520-0442(1996)009<2802:DVOCCA>2.0.CO;2), 1996.
- 20 Bergstrom, R. W., Pilewskie, P., Russell, P. B., Redemann, J., Bond, T. C., Quinn, P. K., and Sierau, B.: Spectral absorption properties of atmospheric aerosols, *Atmos. Chem. Phys.*, 7, 5937–5943, <https://doi.org/10.5194/acp-7-5937-2007>, 2007.
- Boucher, O., Randall, D., Artaxo, P., Bretherton, C., Feingold, G., Forster, P., Kerminen, V.-M., Kondo, Y., Liao, H., Lohmann, U., 25 Rasch, P., Satheesh, S., Sherwood, S., Stevens, B., and Zhang, X.: Clouds and Aerosols, in: *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P., Cambridge Univ. Press, Cambridge, UK and New York, NY, USA, 2013.
- Bovensmann, H., Burrows, J. P., Buchwitz, M., Frerick, J., Noël, S., Rozanov, V. V., Chance, K. V., and Goede, A. P. H.: 35 SCIAMACHY: Mission Objectives and Measurement Modes, *J. Atmos. Sci.*, 56, 127–150, <https://doi.org/10.1175/1520-0469.1999>.
- Cahalan, R. F., Ridgway, W., Wiscombe, W. J., Bell, T. L., and Snider, J. B.: The Albedo of Fractal Stratocumulus Clouds, 40 *J. Atmos. Sci.*, 51, 2434–2455, 1994.
- Chand, D., Anderson, T. L., Wood, R., Charlson, R. J., Hu, Y., Liu, Z., and Vaughan, M.: Quantifying above-cloud aerosol using spaceborne lidar for improved understanding of cloud-sky direct climate forcing, *J. Geophys. Res.*, 113, D13206, 45 <https://doi.org/10.1029/2007JD009433>, 2008.
- Chand, D., Wood, R., Anderson, T. L., Satheesh, S. K., and Charlson, R. J.: Satellite-derived direct radiative effect of aerosols dependent on cloud cover, *Nat. Geosci.*, 2, <https://doi.org/10.1038/NGEO437>, 2009.
- 50 Coddington, O. M., Pilewskie, P., Redemann, J., Platnick, S., Russell, P. B., Schmidt, K. S., Gore, W. J., Livingston, J., Wind, G., and Vukicevic, T.: Examining the impact of overlying aerosols on the retrieval of cloud optical properties from passive remote sensing, *J. Geophys. Res.*, 115, D10211, <https://doi.org/10.1029/2009JD012829>, 2010. 55
- Cox, C. and Munk, W.: Measurement of the Roughness of the Sea Surface from Photographs of the Sun's Glitter, *J. Opt. Soc. Am.*, 44, 838–850, <https://doi.org/10.1364/JOSA.44.000838>, <http://www.osapublishing.org/abstract.cfm?URI=josa-44-11-838>, 1954. 60
- de Graaf, M., Stammes, P., Torres, O., and Koelemeijer, R. B. A.: Absorbing Aerosol Index: Sensitivity Analysis, application to GOME and comparison with TOMS, *J. Geophys. Res.*, 110, D01201, <https://doi.org/10.1029/2004JD005178>, 2005.
- de Graaf, M., Stammes, P., and Aben, E. A. A.: Analysis of reflectance spectra of UV-absorbing aerosol scenes measured by SCIAMACHY, *J. Geophys. Res.*, 112, D02206, <https://doi.org/10.1029/2006JD007249>, 2007. 65
- de Graaf, M., Tilstra, L. G., Aben, I., and Stammes, P.: Satellite observations of the seasonal cycles of absorbing aerosols in Africa related to the monsoon rainfall, 1995 - 2008., *Atmos. Environ.*, 44, 1274–1283, <https://doi.org/10.1016/j.atmosenv.2009.12.03>, 2010. 70
- de Graaf, M., Tilstra, L. G., Wang, P., and Stammes, P.: Retrieval of the aerosol direct radiative effect over clouds from spaceborne spectrometry, 75 *J. Geophys. Res.*, 117, <https://doi.org/10.1029/2011JD017160>, <http://dx.doi.org/10.1029/2011JD017160>, 2012.
- de Graaf, M., Bellouin, N., Tilstra, L. G., Haywood, J., and Stammes, P.: Aerosol direct radiative effect of smoke over 80 clouds over the southeast Atlantic Ocean from 2006 to 2009, *Geophys. Res. Lett.*, <https://doi.org/10.1002/2014GL061103>, <http://dx.doi.org/10.1002/2014GL061103>, 2014.
- de Graaf, M., Sihler, H., Tilstra, L. G., and Stammes, P.: How big is an OMI pixel?, 85 *Atmos. Meas. Tech.*, <https://doi.org/10.5194/amt-9-3607-2016>, <http://www.atmos-meas-tech.net/9/3607/2016/>, 2016.
- de Graaf, M., Tilstra, L., and Stammes, P.: Aerosol direct radiative effect over clouds from synergic OMI and MODIS reflectances, *Atmos. Meas. Tech. Disc.*, 90 2019, 1 – 21, <https://doi.org/10.5194/amt-2019-53>, <https://www.atmos-meas-tech-discuss.net/amt-2019-53/>, 2019.
- Deaconu, L. T., Waquet, F., Josset, D., Ferlay, N., Peers, F., Thieuleux, F., Ducos, F., Pascal, N., Tanré, D., Pelon, J., and Goloub, P.: Consistency of aerosols above 95 clouds characterization from A-Train active and passive measurements, *Atmos. Meas. Tech.*, 10, 3499–3523, <https://doi.org/10.5194/amt-10-3499-2017>, 2017.
- Deschamps, P. Y., Breon, F. M., Leroy, M., Podaire, A., Bricaud, A., Buriez, J. C., and Seze, G.: The POLDER mission: instrument characteristics and scientific objectives, *IEEE Transactions on Geoscience and Remote Sensing*, 32, 598–615, <https://doi.org/10.1109/36.297978>, 1994. 100
- Feingold, G., Jiang, H., and Harrington, J. Y.: On smoke suppression of clouds in Amazonia, *Geophys. Res. Lett.*, 32, <https://doi.org/10.1029/2004GL021369>, 2005. 105
- Feng, N. and Christopher, S. A.: Measurement-based estimates of direct radiative effects of absorbing aerosols above clouds, *J. Geophys. Res.*, 120, 6908–6921, <https://doi.org/10.1002/2015JD023252>, 2015. 110
- Forster, P., Ramaswamy, V., Artaxo, P., Bernsten, T., Betts, R., Fahey, D. W., Haywood, J., Lean, J., Lowe, D. C., Myhre, G.,

- Nganga, J., Prinn, R., Raga, G., Schulz, M., and Van Dorland, R.: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, in: *Climate Change 2007: The Physical Science Basis.*, edited by Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K., Tignor, M., and Miller, H., p. 996, Cambridge Univ. Press, Cambridge, UK and New York, NY, USA, 2007.
- Haywood, J. M., Osborne, S. R., Francis, P. N., Neil, A., Formenti, P., Andreae, M. O., and Kaye, P. H.: The mean physical and optical properties of regional haze dominated by biomass burning aerosol measured from the C-130 aircraft during SAFARI 2000, *J. Geophys. Res.*, 108, D13, <https://doi.org/10.1029/2002JD002226>, 2003.
- Haywood, J. M., Osborne, S. R., and Abel, S. J.: The effect of overlying absorbing aerosol layers on remote sensing retrievals of cloud effective radius and cloud optical depth, *Q. J. R. Meteorol. Soc.*, 130, 779–800, <https://doi.org/10.1256/qj.03.100>, 2004.
- Jethva, H. and Torres, O.: Satellite-based evidence of wavelength-dependent aerosol absorption in biomass burning smoke inferred from Ozone Monitoring Instrument, *Atmos. Chem. Phys.*, 11, 10 541–10 551, <https://doi.org/10.5194/acp-11-10541-2011>, 2011.
- Jethva, H., Torres, O., Remer, L. A., and Bhartia, P. K.: A Color Ratio Method for Simultaneous Retrieval of Aerosol and Cloud Optical Thickness of Above-Cloud Absorbing Aerosols From Passive Sensors: Application to MODIS Measurements, *IEEE T. Geosci. Remote*, 51, 3862–3870, <https://doi.org/10.1109/TGRS.2012.2230008>, 2013.
- Jethva, H., Torres, O., Waquet, F., Chand, D., and Hu, Y.: How do A-train Sensors Intercompare in the Retrieval of Above-Cloud Aerosol Optical Depth? A Case Study-based Assessment, *Geophys. Res. Lett.*, 41, <https://doi.org/10.1002/2013GL058405>, 2014.
- Johnson, B. T., Shine, K. P., and Forster, P. M.: The semi-direct aerosol effect: Impact of absorbing aerosols on marine stratocumulus, *Q. J. R. Meteorol. Soc.*, 130, 1407–1422, <https://doi.org/10.1256/qj.03.61>, 2004.
- Levelt, P. F., van den Oord, G. H. J., Dobber, M. R., Mälkki, A., Visser, H., de Vries, J., Stammes, P., Lundell, J. O. V., and Saari, H.: The ozone monitoring instrument, *IEEE T. Geosci. Remote*, 44, 1093–1101, 2006.
- Marbach, T., Riedi, J., Lacan, A., and Schluessel, P.: The 3MI Mission: Multi-Viewing -Channel -Polarisation Imager of the EUMETSAT Polar System -Second Generation (EPS-SG) dedicated to aerosol and cloud monitoring, *Proc SPIE*, 9613, 10–1, <https://doi.org/10.1117/12.2186978>, 2015.
- Meyer, K., Platnick, S., Oreopoulos, L., and Lee, D.: Estimating the direct radiative effect of absorbing aerosols overlying marine boundary layer clouds in the southeast Atlantic using MODIS and CALIOP, *J. Geophys. Res.*, 118, <https://doi.org/10.1002/jgrd.50449>, 2013.
- Meyer, K., Platnick, S., and Zhang, Z.: Simultaneously inferring above-cloud absorbing aerosol optical thickness and underlying liquid phase cloud optical and microphysical properties using MODIS, *J. Geophys. Res.*, 120, 5524–5547, <https://doi.org/10.1002/2015JD023128>, 2015.
- Myhre, G., Samset, B. H., Schulz, M., Balkanski, Y., Bauer, S., Bernsten, T. K., Bian, H., Bellouin, N., Chin, M., Diehl, T., Easter, R. C., Feichter, J., Ghan, S. J., Hauglustaine, D., Iversen, T., Kinne, S., Kirkevåg, A., Lamarque, J.-F., Lin, G., Liu, X., Lund, M. T., Luo, G., Ma, X., van Noije, T., Penner, J. E., Rasch, P. J., Ruiz, A., Seland, Ø., Skeie, R. B., Stier, P., Takemura, T., Tsigaridis, K., Wang, P., Wang, Z., Xu, L., Yu, H., Yu, F., Yoon, J.-H., Zhang, K., Zhang, H., and Zhou, C.: Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations, *Atmos. Chem. Phys.*, 13, 1853–1877, <https://doi.org/10.5194/acp-13-1853-2013>, 2013.
- Oreopoulos, L. and Davies, R.: Plane Parallel Albedo Biases from Satellite Observations. Part I: Dependence on Resolution and Other Factors, *J. Climate*, 11, 919–932, [https://doi.org/10.1175/1520-0442\(1998\)011<0919:PPABFS>2.0.CO;2](https://doi.org/10.1175/1520-0442(1998)011<0919:PPABFS>2.0.CO;2), 1998.
- Peers, F., Waquet, F., Cornet, C., Dubuisson, P., Ducos, F., Goloub, P., Szczap, F., Tanré, D., and Thieuleux, F.: Absorption of aerosols above clouds from POLDER/PARASOL measurements and estimation of their direct radiative effects, *Atmos. Chem. Phys.*, 15, 4179–4196, <https://doi.org/10.5194/acp-15-4179-2015>, 2015.
- Russell, P. B., Bergstrom, R. W., Shinozuka, Y., Clarke, A. D., DeCarlo, P. F., Jimenez, J. L., Livingston, J. M., Redemann, J., Dubovik, O., and Strawa, A.: Absorption Angstrom Exponent in AERONET and related data as an indicator of aerosol composition, *Atmos. Chem. Phys.*, 10, 1155–1169, <https://doi.org/10.5194/acp-10-1155-2010>, 2010.
- Schulte, R.: A quantitative comparison of spaceborne spectrometry and polarimetry measurements of the aerosol direct radiative effect over clouds, Tech. Rep. KNMI IR–2016–09, Royal Netherlands Meteorological Institute (KNMI), 2016.
- Sorooshian, A., Feingold, G., Lebsock, M. D., Jiang, H., and Stephens, G. L.: On the precipitation susceptibility of clouds to aerosol perturbations, *Geophys. Res. Lett.*, 36, <https://doi.org/10.1029/2009GL038993>, 2009.
- Swap, R., Garstang, M., Macko, S. A., Tyson, P. D., Maenhaut, W., Artaxo, P., Källberg, P., and Talbot, R.: The long-range transport of southern African aerosols to the tropical South Atlantic, *J. Geophys. Res.*, 101, D19, <https://doi.org/10.1029/95JD01049>, 1996.
- Torres, O., Jethva, H., and Bhartia, P. K.: Retrieval of Aerosol Optical Depth above Clouds from OMI Observations: Sensitivity Analysis and Case Studies, *J. Atmos. Sci.*, 69, 0022–4928, <https://doi.org/10.1175/JAS-D-11-0130.1>, 2011.
- van der Werf, G. R., Randerson, J. T., Giglio, L., Collatz, G. J., Mu, M., Kasibhatla, P. S., Morton, D. C., DeFries, R. S., Jin, Y., and van Leeuwen, T. T.: Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997–2009), *Atmos. Chem. Phys.*, 10, 11 707–11 735, <https://doi.org/10.5194/acp-10-11707-2010>, 2010.
- Wang, P., Stammes, P., van der A, R., Pinardi, G., and van Roozendaal, M.: FRESCO+: an improved O₂ A-band cloud retrieval algorithm for tropospheric trace gas retrievals, *Atmos. Chem. Phys.*, 8, 6565–6576, <https://doi.org/10.5194/acp-8-6565-2008>, 2008.
- Waquet, F., Cornet, C., Deuzé, J.-L., Dubovik, O., Ducos, F., Goloub, P., Herman, M., Lapyonok, T., Labonnote, L. C., Riedi, J., Tanré, D., Thieuleux, F., and Vanbuauc, C.: Retrieval of aerosol microphysical and optical properties above liquid clouds from POLDER/PARASOL po-

- larization measurements, *Atmos. Meas. Tech.*, 4, 991–1016, <https://doi.org/10.5194/amt-6-991-2013>, 2013a.
- Waquet, F., Peers, F., Ducos, F., Goloub, P., Platnick, S., Riedi, J., Tanré, D., and Thieuleux, F.: Global analysis of aerosol properties above clouds, *Geophys. Res. Lett.*, 40, 5809–5814, <https://doi.org/10.1002/2013GL057482>, 2013b.
- Wilcox, E. M.: Stratocumulus cloud thickening beneath layers of absorbing smoke aerosol, *Atmos. Chem. Phys.*, 10, 11 769–11 777, 2010.
- 10 Wilcox, E. M.: Direct and semi-direct radiative forcing of smoke aerosols over clouds, *Atmos. Chem. Phys.*, 12, 139–149, <https://doi.org/10.5194/acp-12-139-2012>, 2012.
- Yu, H. and Zhang, Z.: New Directions: Emerging satellite observations of above-cloud aerosols and direct radiative forcing, *Atmos. Environ.*, 72, 36–40, <https://doi.org/http://dx.doi.org/10.1016/j.atmosenv.2013.02.017>, 2013.
- 15 Yu, H., Liu, S. C., and Dickinson, R. E.: Radiative effects of aerosols on the evolution of the atmospheric boundary layer, *J. Geophys. Res.*, 107, <https://doi.org/10.1029/2001JD000754>, 2002.
- Zeng, S., Cornet, C., Parol, F., Riedi, J., and Thieuleux, F.: A better understanding of cloud optical thickness derived from the passive sensors MODIS/AQUA and POLDER/PARASOL in the A-Train constellation, *Atmos. Chem. Phys.*, 12, 11 245–11 259, <https://doi.org/10.5194/acp-12-11245-2012>, 2012.
- 25 Zhang, Z., Meyer, K., Platnick, S., Oreopoulos, L., Lee, D., and Yu, H.: A novel method for estimating short-wave direct radiative effect of above-cloud aerosols using CALIOP and MODIS data, *Atmos. Meas. Tech.*, 7, 1777–1789, <https://doi.org/10.5194/amt-7-1777-2014>, 2014.
- 30 Zuidema, P., Redemann, J., Haywood, J., Wood, R., Piketh, S., Hipondoka, M., and Formenti, P.: Smoke and Clouds above the Southeast Atlantic: Upcoming Field Campaigns Probe Absorbing Aerosol's Impact on Climate, *Bull. Am. Meteor. Soc.*, 97, 1131–1135, <https://doi.org/10.1175/BAMS-D-15-00082.1>, 2016.
- 35