Reviewer #1

2nd Review of de Graaf, et al. 2019.

This is my second review of this manuscript. I thank the authours for taking onboard many of the comments from my and the other review, and for improving the manuscript. It is easier to follow now, although there are still improvements that could be made to make it clearer (see the specific suggestions below, but there are likely more things that could be done). As well as these, a table showing the resolutions of the different instruments would be useful given the frequent re-gridding. Proof reading for grammar is also needed due to a few grammatical errors that hamper the reading somewhat.

The reviewer is thanked for the amount of time invested to improve the paper. All issues raised by the reviewer have been considered and addressed below. All changes to the manuscript are indicated for each answer. The desired table (see Table 1) was added to the new manuscript. The manuscript was carefully spell-checked and checked for grammar and errors by several co-authors.

Table 1: Spatial and temporal resolution of the different satellite instruments as used in this paper. Grid sizes of SCIAMACHY and OMI are those at nadir, grid sizes of

POLDER and MODIS are fixed.

Instrument	Platform	Local equator	Global cov-	Pixel size	Operation period
		crossing time	erage (days)	$(km \times km)$	
POLDER	PARASOL	13:33	1	6×6	2004 – 2013
SCIAMACHY	EnviSat	10:00	6	60×30	2002 - 2012
OMI	Aura	13:38	1	13×24	2004 – present
MODIS	Aqua	13:30	1	0.5×0.5	2002 – present

Specific suggestions

p.2, L19 "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 about a third of the difference between POLDER and OMI-MODIS DRE, which is mainly evident at high values of the aerosol DRE."

- These two sentences don't seem to agree with each other. Perhaps the first should be modified to "the remaining differences can be almost completely explained", or similar

The idea here is that the uncertainties in the cloud retrievals are already sufficient to explain the differences found between the DRE from the different instruments. Additionally, there are uncertainties in the AOT retrievals, which also contribute to the differences. Added together, the uncertainties are more than the differences found, so there are cancelling errors, or the error estimates are too large. The text was rephrased as follows:

When these are accounted for, the remaining differences can be explained by a higher cloud optical thickness (COT) derived from POLDER compared to the other instruments, and a neglect of AOT at SWIR wavelengths in the method used for SCIA-MACHY and OMI-MODIS. The higher COT from POLDER by itself can explain the difference found in DRE between POLDER and the other instruments. The AOT underestimation is mainly evident at high values of the aerosol DRE and accounts for about a third of the difference between POLDER and OMI-MODIS DRE.

p.3, L56 -" 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)."

- It would be good to quote the warming from the model, or the degree of underestimate.

Agreed. The following text was added, quoted from the paper: "Simulated monthly averaged aerosol DRE from HadGEM2 were a factor of 5 lower than SCIAMACHY observations.

From the first review: - 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 2.1µm 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 2.1µm is expected to be negligible. This is the same argument that is used for the (OMI-)MODIS retrievals, except at 1.2µm. At 2.1µm 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).

I was thinking of the direct retrieval of CER by POLDER using the separation of the peaks in the polarized scattering phase function (Breon, *et al.*, 2005). This retrieval is considered to bypass some of the potential biases in visible+near-infrared based retrievals (e.g., MODIS, etc.). There are such POLDER CER datasets available, although they may be limited to regions with more homogeneous clouds. A comparisons in some regions should be possible. It may be beyond the scope of this study, but should be mentioned at least.

In this paper the POLDER DRE data that are described in Peers *et al.* (2015) are compared with the other datasets. In this version of the data the cloud droplet sizes come from MODIS measurements as mentioned above. An application of CER by POLDER as the reviewer suggests is certainly interesting, and could be considered for a new version of the DRE retrieval.

A note was added to the text:

"CER may be retrieved directly by POLDER for specific cases using the separation of the peaks in the polarized scattering phase function (Bréon *et al.*, 2015), but here we use the data as described in Peers *et al.* (2015)."

And the reference was added.

p.5, L10 "Such an estimate is often missing." Better as "Such estimates are rarely given in the literature.", in order to make it clearer that you are referring to previous studies. Agreed, the text was changed accordingly.

p.5, L11 - Moreover, the correct characterization of the spectral properties of the overlying aerosols is circumvented by DAA." Not sure what you mean here. How is it circumvented? This doesn't seem to fit with the previous sentences.

This is a correct observation. It doesn't fit the previous statement, and should be a separate statement. DAA circumvents any aerosol property assumptions by using the spectral measurements instead of running a model twice (once with and once without (modelled) aerosols). A new paragraph was started and the text was changed as follows:

"The dependence on uncertanties in the spectral properties of the overlying aerosols is small by DAA, because in this method the spectral measurements are used, not a model."

Section 3.1 -You should quote the area average for OMI-MODIS when collocated to the POLDER grid (in the text and in Table 1) since this would give an idea of how much difference the restricted retrievals of POLDER makes and would give support to the statements in the text that this makes a big difference (rather than the larger individual values).

The desired information is available from Table 3 and Figure 2, which can be directly compared to Table 1. The introductory section 3.1 discusses the two selected cases before any analysis, using the data as they are, showing a large difference on one day and more moderate on the other. To quote here values from the regridded cases would confuse readers, since the regridding is not mentioned up to this point. Therefore, these values are discussed at the end of section 3.2.2.

Fig. 2 & Table 2 -In caption for (b) and in Table 2 (for second set of values) it would be better to say that OMI-MODIS and SCHIMACHY were re-gridded to a 6km grid and that averages are only calculated for the 6km grid points for which all 3 instruments produced a measurement.

Agreed. The caption was changed as follows:

"b) Same as a), but for OMI-MODIS and SCIAMACHY pixels that were regridded to the 6×6 km² POLDER grid. Averaged values were only calculated from grid points that were covered by all three instruments. The number of collocated pixels that are covered by all three instruments is given in the lower panel in b)."

p.9, L24 -"Additionally, the sampling was checked by gridding the finer POLDER data to the coarser OMI grid" Please make it clear in the text how this was done. Was an

average over each OMI-MODIS grid-box taken, or was just one value sampled for each OMI-MODIS box? However, if the latter then this might make the statistics less robust. Doing an average of lots of POLDER values for each box would be better.

The smaller POLDER pixels were sampled over the OMI footprint using an advanced 2D Gaussian weighting function, in the same way as MODIS pixels are weighted in the OMI footprint for the OMI-MODIS DRE computation. This is indeed much more accurate than a simple nearest neighbour sampling. The following was added to the text:

"The smaller POLDER pixels were averaged over the OMI footprint using a 2D Gaussian weighting function. This procedure is exactly the same for the averaging of MODIS pixels in an OMI footprint in the OMI-MODIS DRE computation, and described in detail in De Graaf *et al.* (2016)."

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p.9, L28 " "loose" - > "lose" Indeed. Thank you.
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p.10, L38 ""SCIAMACHY DRE is very similar to OMI-MODIS DRE for all pixels sampled by these instruments (not shown)." This is a strange blanket statement that seems to suggest that SCIAMACY and OMI agree for all pixels, which is surely not the case. This needs to be more quantitative, or show some results (e.g, a scatter plot). Agreed, the sentence on SCIAMACHY data doesn't really add anything to this paragraph and was removed.

Fig. 5a "there are 4 lines for each group of COT values, but only two entries in the legend (reff=8 and 12).

The legend shows that each group of two values refer to either 10° or 60° viewing zenith angle. The legend in the figure was changed to show also this color.

p.11 L21 -"Irrespective of AOT or COT, an error of 20% in COT can lead to an error in DRE of 50 Wm"-2." This is written in a slightly odd way that makes it seem like the error is always 50 W/m2 no matter what the COT or AOT values are. In fact it looks like the error value ranges from about 30 to 80 W/m2. I suggest :- "An error of 20% in COT leads to an error in DRE of between 30 and 80 Wm-2 for the COT values tested (COT=8 and COT=12), irrespective of the AOT."

The reviewer is correct that this is not for any COT and AOT value, but only those that are shown. However, the error range that the reviewer suggest is far too large. Therefore, the text was changed as follows:

An error of 20% in COT can lead to an error in DRE of *about* 50 Wm-2, for COT in the range of 8–16, irrespective of the AOT.

p.11, L23 ""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-MODISCOT 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."

This is difficult to follow and understand "it could do with a rewrite and a more careful explanation of what you mean.

The text was rewritten as follows:

The DRE is computed from the difference between a measured and simulated spectrum, which both have exactly the same COT and CER (since the simulation is done with the COT and CER retrieved from the measured spectrum). Therefore, any errors in the COT and CER retrieval have no influence on the difference between the two spectra and do not show in the DRE. However, if the COT or CER for the simulation were taken from a different measurement, however accurate, the simulated and measured spectra may be very different, giving rise to large DRE values, even without overlying aerosols. This was observed in a test where POLDER COT, regridded to the OMI grid, was used in the DRE computation, instead of the COT from the OMI-MODIS spectra. Even though the POLDER COT was probably more accurate than the OMI-MODIS COT, the derived DRE was very erratic.

p.11, L41 " "However, it is assumed to have negligible effect in the SWIR from about $1.2 \, \mu m$, which may be an underestimation (in AOT)."

This sentence doesn't really make sense. The first part of the sentence seems to say that AOT errors have a negligible effect, but does not say on what. The second part suggests there is an underestimation of AOT, but it is not clear.

Agreed. The text was changed as follows:

"However, the AOT of small particles like smoke is assumed to be negligible in the SWIR from about 1.2 μ m, which may be an underestimation.

p.12, L26 -"Note that POLDER COT is retrieved at 0.87 μ m, while COT from OMI-MODIS is retrieved at 1.2 μ m, which effectively is the MODIS channel."

MODIS has lots of channels, so I'm not sure I understand the last part of the sentence. Also, MODIS can retrieve COT using the 0.86um channel (as for POLDER), although combined with the 2.1 um or 3.7 um channel. However, most of the signal comes from the 0.86um channel. Or does the possibility of the above-cloud aerosol prevent the use of this?

"MODIS channel" was changed to "MODIS measurement".

Indeed, the whole idea of retrieving COT in the SWIR is to avoid biases due to aerosol absorption in the UV-VIS channels.

p.12, L38 " "Here, POLDER COT is regridded, while the MODIS radiances are averaged in the OMI footprint and one COT is retrieved for that OMI pixel."

It's not clear at this point in the manuscript why this is being done.

It is strange to read this remark, when the reason for doing this was that the reviewer suggested this in the first review. In the first draft the OMI-MODIS values were resampled to the POLDER grid, in the current manuscript the gridding is to the coarser OMI grid, which is more accurate, but does not yield fundamentally different results. However, "Here, POLDER COT is regridded," was changed to "Here, POLDER COT values were averaged over the OMI footprint,"

It is interesting to compare the effect of averaging the (1km resolution, or higher? Please state) MODIS radiances to the OMI resolution before doing the COT retrieval. But it needs some introduction at the start of the paragraph about the reasons for doing it. Is this is what is done to get the OMI-MODIS COT values used in the DRE retrieval? MODIS L1B reflectances have a spatial resolution of 250 or 500 m, depending on the channel. They have to be first co-added to 500 m and then gridded. The gridding over the OMI footprint is done using a 2D weighting function over the OMI footprint, described in De Graaf *et al.* (2016) and De Graaf *et al.* (2019).

Or are the 1km MODIS COT values averaged over each OMI gridbox? It should be mentioned in Section 2.3 how this is done.

No, this is not done. In section 3.2.2 it states: "for MODIS the radiances were averaged and one COT is retrieved for that OMI pixel."

If the MODIS COT values are averaged, you should also show a scatter plot of the POLDER vs OMI -MODIS COT values where the POLDER values are averaged to the OMI grid and the MODIS COT values are averaged.

But this is not done.

Also, please state what you do with the POLDER COT values. Do you average them over the OMI grid -boxes?

Yes, as mentioned previously, the POLDER values are not taken in a nearest neighbour fashion, but properly averaged using a weighting function over the OMI footprint, and described in De Graaf *et al.* (2016) and De Graaf *et al.* (2019). This was added to the manuscript.

Table 3 " it would be good to add that the POLDER data is gridded to the coarser OMI-MODIS grid in the caption. And whether the POLDER data was averaged over the OMI boxes, or interpolated/sampled.

This is already explained in the text. In the table are three headers: native grid, collocated POLDER grid, collocated OMI grid, which should make it clear which values belong to which analysis.

p.13, L30 -"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. 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."

My issue here from the 1st review was that it wasn't clear what filtering by CF and COT was done. Looking through the manuscript I see that this probably refers to the requirement of minimum CF and COT values for a DRE retrieval in Sections 2.1-2.3. However, since this information is in the methods section and not mentioned since, it wasn't clear here what was being referred to. Before talking about CF and COT filtering here it would be good to reiterate that different minimum CF and COT values were used for the different instruments. Also, I think here you mean "This approach removes issues related to filtering based on COT and CF, which can select high positive DRE values and lead to large differences in the average DRE."

The suggestion of the reviewer was adopted and "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." was changed to "This approach removes issues related to filtering based on COT and CF, which can select high positive DRE values and lead to large differences in the average DRE." The filter settings are mentioned in the methods section under the different instrument descriptions, which seems the correct place for this information and thus not changed.

p.13, L38 "This difference was removed by gridding POLDER to the coarser OMI grid, improving the comparison between OMI-MODIS DRE and POLDER DRE." This wouldn't completely remove the effect because the effect of averaging the reflectances from the coarser resolution instrument (and then doing the retrieval) would still remain "this causes differences due to the non-linearity between the reflectances and retrieved product. I.e., it's not just a case of averaging the POLDER values to a coarser grid.

Agreed; "removed" was changed to "reduced".

p.13, L62 "The underestimation of the AOT for high values can explain about a third of the difference in DRE between POLDER and OMI-MODIS om 12 August 2006, an overestimation of AOT by POLDER is difficult to establish."

The last part of this sentence (after the 1comma) doesn't fit with the rest. Also "om 12 August".

Agreed. Changed to:

"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. POLDER AOT may be overestimated, but this is difficult to quantify."

p.13, L69 "Normally, MODIS COT retrievals at 0.8 and 2.1 μ m 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 μ m are used to derive COT and CER for OMI-MODIS DRE retrievals, which may further influence the results."

- This information should also be in the methods section to explain why the usual

MODIS channels are not used for COT. Also, the second "retrievals" word should be removed.

"retrievals" was removed. The method mentions: "Cloud properties were determined at 1.2 and 1.6 μ m, where absorption by smoke is assumed to be negligible."

p.13, L76 " "The difference between COT from OMI-MODIS and POLDER on 12 August 2006 can explain about 80% of the difference in DRE on that day."

- Please state whether this is before or after the footprints have been collocated. This is after sampling issues have been removed. This is described in the text and the conclusion section, because that states that sampling issues are the largest contribution to the differences and should be removed first.

p.13, L85 " "However, a test using this approach using DDA on OMI-MODIS spectra using POLDER COT yielded very erratic results." — > "However, a test of this approach using DDA on OMI-MODIS spectra but with POLDER COT yielded very erratic results." (too many instances of "using").

Agreed.

p. 13, L87 ""clouds spectrum" – > "cloud spectrum". Agreed.

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

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

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 radi-5 ation 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 cli-10 mate 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**Scanning Imaging 15 Absorption Spectrometer for Atmospheric CHartographY (SCIAMACHY), the polarimeter **POLDER**Polarization and Directionality of the Earth's Reflectances (POLDER), and from collocated measurements by the spectrometer OMI and imager MODISOzone Monitoring Instrument 20 (OMI) and the imager Moderate Resolution Imaging Spectroradiometer(MODIS). All three datasets 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 is-25 sues. When these are accounted for, the remaining differences can be completely explained by the explained by a higher cloud optical thickness (COT) derived from POLDER compared to the other instruments. Additionally, and a neglect of AOT at SWIR aerosol optical thickness (AOT) at 30 shortwave infrared (SWIR) wavelengths in the method used

for SCIAMACHY and OMI-MODISaccounts about a third

of the difference. The higher COT from POLDER by itself

can explain the difference found in DRE between POLDER and OMI-MODIS DRE, which the other instruments. The AOT underestimation is mainly evident at high values of the aerosol DRE and accounts for about a third of the difference between POLDER and OMI-MODIS 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 (GCMs). Comparisons of model and satellite datasets should also account for sampling issues. A combination of the The complementary DRE retrievals from OMI-MODIS and POLDER may benefit from upcoming satellite missions that combine spectrometer and polarimeter measurements.

Copyright statement.

1 Introduction

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 sunlight by aerosols adds heat to the atmosphere at the aerosol layer height, changing the atmospheric stability and the amount of radiation re-

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

ceived 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., 5 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 10 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±0.3 Wm⁻², but the large uncertainty of aerosol-₁₅ 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 (Boucher et al., 2013).

Constraining aerosol effects in model studies 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 al-30 lowing 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 effect 35 and the instantaneous irradiance change at the Top Of the Atmosphere (TOA) can be substituted. The instantaneous aerosol Direct Radiative Effect (DRE) direct radiative effect at TOA is therefore defined as the change in net (upwelling minus downwelling) irradiance, due to the introduction of 40 aerosols in the atmosphere. Since at TOA the downwelling irradiance F^{\downarrow} is the incoming solar irradiance F_0 for all scenes, the aerosol DRE for a cloud scene the aerosol DRE can be determined from the difference between the upwelling irradiance in an aerosol-free cloud scene $F_{\rm cld}^{\uparrow}$ and the upwelling $_{45}$ irradiance of a scene with the same clouds plus aerosols $F_{cld+aer}^{\uparrow}$:

$$DRE_{aer} = (F^{\downarrow} - F^{\uparrow})_{cld} - (F^{\downarrow} - F^{\uparrow})_{cld+aer} = F^{\uparrow}_{cld+aer} - F^{\uparrow}_{cld}. \tag{1}$$

A radiative transfer model (RTM) is commonly used to, given the atmospheric constituents in the atmosphere, simusolate 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 extend the light absorption and scattering properties of the air and the surface reflectance.

The 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 65 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 $+50~{\rm Wm^{-2}}\tau^{-1}$ 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 ($\sim 25~{\rm Tg}$ 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) 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 (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 disentangle. 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 com-

monly 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 ver-5 tically 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 10 droplets and irregularly shaped aerosol particles on the polarization 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 15 using reflectances measured in the visible and shortwave infrared. Knowing the COT, and AOT 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 $(F_{\rm cld+aer}^{\uparrow})$ and without the aerosols $(F_{\rm cld}^{\uparrow})$. The 20 monthly averaged instantaneous DRE values from POLDER for aerosols over clouds over the south-east Atlantic Ocean in August 2006 found in this way 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 Absorbing Aerosol Index (UV-AIAAI) (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 In this way, the AOT of smoke above clouds in-was retrieved over the south-east Atlantic, and with the COT of the clouds underneath retrieved simultaneously, using OMI measure-35 ments (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 Ångtröm parameterngström exponent) 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 in- teractions at all wavelengths. Measurements from six wavelength channels from MODIS (from $0.47-1.24\mu m$) 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 $F_{cld+aer}^{\uparrow}$), and only the aerosolfree atmosphere is simulated in an RTM (simulated F_{cld}^{\uparrow}). The cloud properties can be retrieved in the SWIR where 65 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 monthly averaged instantaneous DRE values from SCIA-MACHY for aerosols over clouds over the south-east Atlantic in August 2006 found in this way was 23 Wm⁻². The DRE from SCIAMACHY was compared to Hadley Centre Global Environmental Model version 2 (HadGEM2) cli-75 mate model simulations (de Graaf et al., 2014). Simulated monthly averaged aerosol DRE from HadGEM2 were a factor of 5 lower than SCIAMACHY observations, showing that even this GCM, which simulated a large warming over the south-east Atlantic, still fell short in simulating the 80 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 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 Reflectances for Atmospheric Science coupled with Observations from a Lidar (PARASOL) (Deschamps et al., 1994).

Instrument	Platform	Local equator	Global cov-	Pixel size	Omanation maniad	
msu ument		crossing time	erage (days)	$(km \times km)$	Operation period	
POLDER	PARASOL	13:33	1	6×6	2004 – 2013	
SCIAMACHY	EnviSat	10:00	<u>6</u> ~	60×30	2002 - 2012	
$ \underbrace{OMI} $	Aura	13:38	1	3×24	2004 – present	
MODIS	Aqua	13:30	1	0.5 imes 0.5	2002 – present	

Table 1. Spatial and temporal resolution of the different satellite instruments as used in this paper. Grid sizes of SCIAMACHY and OMI are those at nadir, grid sizes of POLDER and MODIS are fixed.

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 polarization measurements at 490, 670 and 865 nm. The ground spatial resolution is about 5.3 × 6.2 km² and the swath width about 1100 km (Deschamps et al., 1994). All measurements of POLDER are projected on a fixed global reference grid of 6×6 km².

The POLDER method retrieves Using POLDER measurements, the above-cloud AOT, the aerosol Single Scattering Albedo (SSA) and the COT are retrieved in two steps. The first one consists of using the polarization radiance measurements to retrieve the scattering AOT and 15 the aerosol size distribution in a cloudy scene. Aerosols affect the polarization 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 20 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. 25 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 30 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 southeast 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 O₂ 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. CER may be retrieved directly by POLDER for specific cases using the separation of the peaks in the polarized scattering phase function

(Bréon and Doutriaux-Boucher, 2005), but here we use the data as described in (Peers et al., 2015).

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 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 vapor content were obtained from meteorological reanalysis.

2.2 SCIAMACHY DRE

The DAA method was developed for reflectance spectra from 55 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 local solar time in a descending (south- 60 ward) direction, but stopped delivering data in 2012. SCIA-MACHY observed radiation in two alternating modes, nadir and limb, yielding data blocks called states, approximately 960×490 km² in size. In nadir mode, SCIAMACHY measured continuous reflectance spectra from 240–2380 nm with 65 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 70 cloud scenes in 2006 over the south-east Atlantic. Cloud properties were determined at 1.2 and 1.6 μ m, where absorption by smoke is assumed to be negligible. Effective CF and cloud pressure (CP) were determined from (FRESCO) O2-A band retrievals (Wang et al., 2008). All scenes with effective 75 CF > 0.3, CP > 850 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 80 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 5 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 15×23.5 km² at nadir to about 42×126 km² 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 Aura in the A-Train, leading 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. Spectrally, 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 μ m, because of a reduced sampling in MODIS/Aqua 35 1.6 μ m band due to nonfunctional detectors (Meyer et al., 2015). CP and effective CF are available from OMI O₂-O₂ retrievals. All scenes with COT > 3.0, effective CF > 0.3 and CP > 850 hPa were selected. The ocean surface albedo was assumed to have a small, spectrally dependent, constant 40 value.

The temporal and spatial resolutions of the various instruments compared in this paper are summarized in Table 1.

2.4 Error budget

45 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 SCIA-MACHY and OMI-MODIS scenes this was actually tested, by applying the technique to measured aerosol-free cloud
 50 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 absorb-

Table 2. Maximum and average values of OMI-MODIS, SCIA-MACHY, 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	⟨DRE⟩
POLDER	303.8	109.1
OMI-MODIS	120.0	35.5
SCIAMACHY	112.5	28.4
19 August 2006		
POLDER	190.3	43.0
OMI-MODIS	94.0	11.4
SCIAMACHY	71.3	18.1

ing aerosol using the aerosol UV index.AAI, 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.

Moreover, the correct characterization of the estimates are rarely given in the literature.

The dependence on uncertainties in the spectral properties of the overlying aerosols is eireumvented by DAAsmall by DAA, because in this method the spectral measurements are used, not a model. 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 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 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, on the 12th and the 19th 12th and the 19th. The first case shows the situation during the

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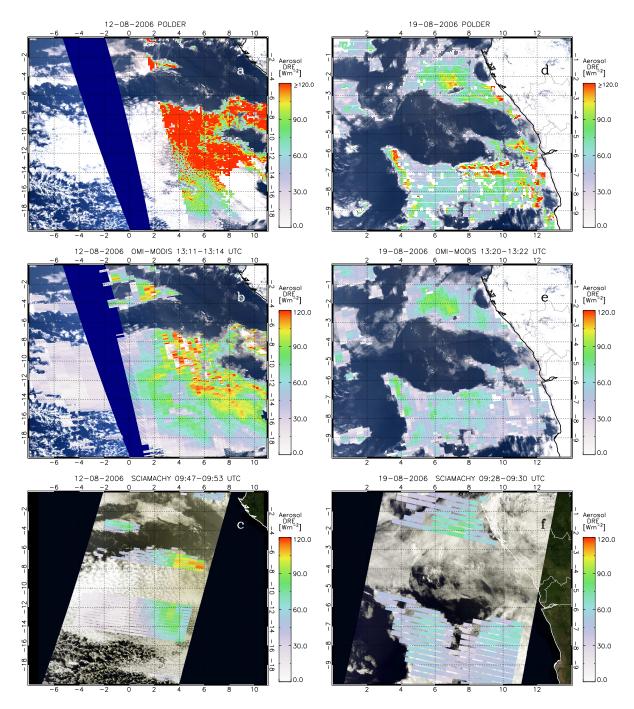


Figure 1. (a) Instantaneous Aerosol Direct Radiative Effect (DRE) over clouds on 12 August 2006 from POLDER, overlaid over a MODIS RGB image; (b) Aerosol DRE over clouds on the same day from a combination of OMI and MODIS reflectances, overlaid over the same MODIS RGB image; (c) Aerosol DRE over clouds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

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 and d show the POLDER DRE overlaid an over a MODIS RGB image acquired around 13:10–13:20 UTC, Figures 1b and e show the OMI-MODIS DRE over the same MODIS RGB

image, and Figures 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 9:30–9:45 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.

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 DRE is smoother with a better coverage. How-20 ever, 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. On 12 August 2006, the POLDER DRE is very large, reaching values up to 304 Wm⁻². The OMI-25 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 30 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 2.

Furthermore, a much higher area-averaged POLDER DRE on 12 August 2006 is found then for the other instruments, which is not only due to higher individual values. Figures 1d 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.

Additionally, the SCIAMACHY and OMI large pixel sizes smooth the high DRE values that are found by POLDER. Fixel 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 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.

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

3.2 Area-averaged DRE

Clearly, sampling is an issue that needs to be considered when comparing datasets, and datasets and simulations. Often, area- and time-averages are compared, to reduce the effects of sampling differences. Here we show the effect of ignoring the sampling differences between instruments.

In Figure 2a, the area-averaged instantaneous aerosol DRE over clouds from all three instruments is given for all 75 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., 80 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 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 95 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 100 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 105 OMI-MODIS DRE is never higher than about 60 Wm^{-2} .

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

Table 3. DRE Statistics of the different instruments before and after
collocation for the area $10^{\circ}N20^{\circ}S;10^{\circ}W20^{\circ}E$ in the south-east
Atlantic.

Native grid	Mean	Median	Std. Dev	Skew
POLDER	46.60	38.74	39.00	1.57
OMI-MODIS	24.88	21.92	30.27	0.62
SCIAMACHY	28.42	26.11	24.62	1.26
Collocated POLDER grid				
POLDER	47.11	38.04	40.90	1.84
OMI-MODIS	37.13	33.74	26.15	1.65
SCIAMACHY	39.50	36.23	24.66	1.25
Collocated OMI grid				
POLDER	43.66	36.30	35.91	1.62
OMI-MODIS	35.63	32.29	24.96	0.94

Figure 3a. Clearly, the average POLDER aerosol DRE is almost twice as large as that from OMI-MODIS and SCIA-MACHY (24.9 Wm⁻² for OMI-MODIS, 28.4 Wm⁻² for SCIAMACHY and 46.6 Wm⁻² for POLDER). The statistics of the distributions are given in Table 3. In only the month August, the average aerosol DRE was 27.5 Wm⁻² for 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 mainly due to more 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.

20 **3.2.1** Sampling

Clearly, these different spatial scales limit the usefulness of a comparison of average values from satellite instruments. In order to correct for the issues described above, the OMI-MODIS and SCIAMACHY measurements were regridded 25 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 DRE measurement. 30 This gave SCIAMACHY and OMI-MODIS DRE values on a grid similar to the POLDER grid (albeit smoothed per OMI or SCIAMACHY pixel). The individual POLDER DRE values were then compared to the OMI-MODIS and SCIA-MACHY DRE values in the grid cell that was closest to the 35 POLDER grid cell. In Figure 2b the noon-normalized areaaveraged 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 no areas with DRE that are sampled by all three instruments. 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-normalized areaaveraged instantaneous DRE from the three instruments is now significantly improved compared to Figure 2a. The aerosol DRE from OMI-MODIS follows the aerosol DRE from SCIAMACHY very closely for almost the entire period 50 shown. Note that the maximum DRE from OMI-MODIS is now increased to almost 90 Wm⁻², which was due to removing many pixels with a moderate to low DRE during mid-August, that were not covered by POLDER and SCIA-MACHY (cf. Figures 1a-c). Also note that the day with the 55 largest average values does not occur on the 12th 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 between the instruments is also greatly reduced, see Figure 3b, 60 which shows the histograms for only overlapping pixels regridded to the POLDER grid, and its statistics in Table 3. The average DRE from POLDER is still about 47.0 Wm⁻² for only overlapping pixels, while the average DRE from OMI-MODIS has increased to 37.1 Wm⁻² and 39.5 Wm⁻² for 65 SCIAMACHY regridded pixels.

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 70 to lose lose too many POLDER and OMI-MODIS pixels because of the poor SCIAMACHY sampling. The smaller POLDER pixels were averaged over the OMI footprint using a 2D Gaussian weighting function. This procedure is exactly the same for the averaging of MODIS pixels in 75 an OMI footprint in the OMI-MODIS DRE computation, and described in detail in de Graaf et al. (2016). 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 80 datasets. The statistics are given in Table 3. Obviously, gridding to the OMI grid instead of to the POLDER grid doesn't does not change the results very much, but without SCIA-MACHY the large number of pixels that are collocated results in a very high consistency between OMI-MODIS and 85 POLDER DRE. Furthermore, without SCIAMACHY the noon-normalization 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,

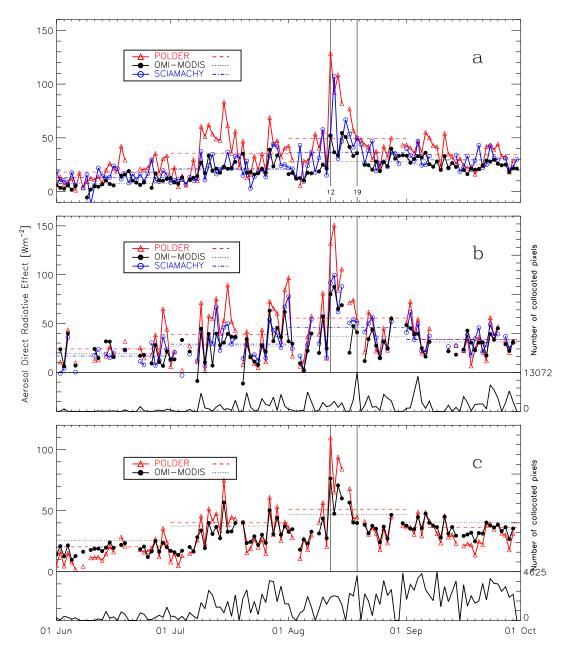


Figure 2. a) Noon-normalized instantaneous aerosol DRE over clouds from combined OMI-MODIS reflectances (black), SCIAMACHY reflectances (blue) and POLDER AOT and COT retrievals (red) from 1 June - 1 October 2006, averaged over the area $10^{\circ}N-20^{\circ}S;10^{\circ}W-20^{\circ}E$ in the south-east 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 and regridded SCIAMACHY pixels that were regridded to the 6×6 km² POLDER grid. Averaged values were only calculated from grid points that were covered by all three instruments. 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-normalized, because the overpass time of OMI, POLDER and MODIS are similar.

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 more high and low values.

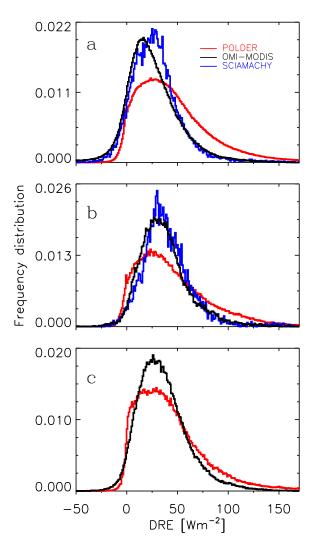


Figure 3. a) Histograms of aerosol DRE over clouds in the Atlantic Ocean during June – September 2006 from POLDER AOT and COT retrievals (red), combined OMI-MODIS reflectance spectra (black) and SCIAMACHY reflectance spectra (blue). b) Same as a) but for collocated POLDER, OMI-MODIS regridded to POLDER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

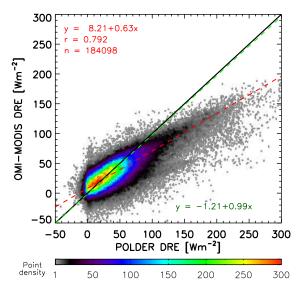


Figure 4. Scatterplot of POLDER DRE gridded to the OMI grid versus DRE from OMI-MODIS data from June-September 2006 over the south-east Atlantic. The red dashed line shows an unweighted linear least-squares fit. The green line shows the linear least-squares fit weighted by the distance to the average value of $25~\mathrm{Wm}^{-2}$.

This is also clear from a scatterplot of collocated POLDER DRE vs. OMI-MODIS DRE for regridded POLDER pixels, shown in Figure 4. The figure shows a good correlation 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 DRE to POLDER DRE of 0.82 can be found from Table 3, while a normal linear least-squares fit (shown by the red line in Figure 4) yields a slope of OMI-MODIS to POLDER ratio of only 0.63. This is because the fit is dominated by the large 10 values, while the large majority of points are moderate values around 25 Wm⁻². When a fit is drawn which is weighted to the deviation from this moderate value (shown by the green line), a slope of 0.99 is found, showing that the aerosol DRE over clouds is the same from POLDER and OMI-MODIS for 15 moderate values.

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

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 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

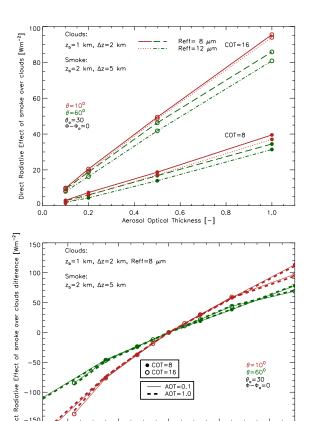


Figure 5. (a) Aerosol DRE for simulated scenes with clouds between 1-2 km and smoke aerosol between 2-5 km as a function of AOT at 550 nm. The COT was 8 or 16, the effective cloud droplet radius 8 or 12 μ m. SZA was 30°, VZA was 10° or 60°, RAZI was 0°. (b) Aerosol DRE for simulated scenes as in (a),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

20 0 20 Δ Cloud Optical Thickness [%]

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 parti- $_{\text{5}}$ cle size distribution with effective radius $r_{\text{eff}}=16\mu\mathrm{m}$ and an effective variance $\nu_{\rm eff} = 0.15$. 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 in-10 dex 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 geomet-15 ric radii for this haze plume used in the simulations were $r_{\rm c}=0.255~\mu{\rm m}$ and $r_{\rm f}=0.117~\mu{\rm m}$ 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 25 effective radii of 8 and 12 μ 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 30 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 and COT 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, for high AOT of 1, a doubling of COT from 8 to 16 increases the DRE from 40 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 45 AOT or COT, an An error of 20% in COT can lead to an error in DRE of about 50 Wm⁻², for COT in the range of 8-16, irrespective of the AOT. A note for DAA is in order here: because the simulated aerosol-free cloud spectrumis computed from The DRE is computed from the difference between a measured and simulated spectrum, which both have exactly the same COT and CER (since the simulation is done with the COT and CER retrieved from the measured aerosol-cloud spectrum spectrum). Therefore, any errors in the COT and CER retrieval have no influence on the difference between the two spectra and do not show in the DRE. However, if the COT or CER for the simulation were taken from a different measurement, however accurate, the simulated and measured spectra may be very different, giving rise to large DRE values, even without overlying aerosols. This was observed in a test where POLDER COT, 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 grid erratic DRE, even, was used in the DRE computation, instead of the COT from 65 the OMI-MODIS spectrum. Even though the POLDER COT retrieved in the visible is probably superior to was probably more accurate than the OMI-MODIS COTretrieved 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 acrosol effects even without aerosols, the derived DRE was very erratic. 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 5 SCIAMACHY) and POLDER can be attributed to differences in AOT, even if AOT is not explicitly retrieved for OMI-MODIS and SCIAMACHY. However, it the AOT for small particles like smoke is assumed to have negligible effect be negligible in the SWIR from about 1.2 μ m, which 10 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. For smoke no estimate was given. Comparisons between 15 AOT above clouds for smoke, but comparisons between AOT for smoke over clouds 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 20 the entire area south-east Atlantic, with individual values up to 1.9. From CALIOP data an AOT of up to 1.5 (532 nm) 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. On 13 August 2006, the maximum 25 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 abovecloud AOT were about 11% higher than the otherwise well correlated above-cloud AOT from the CALIOP depolariza-30 tion ratio method (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 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. This assumption may break apart for larger particles, especially at very high AOT, or for larger particles, leading 40 to an underestimation of the AOT , especially for at higher values, limiting the DRE. The assumption can be tested by estimating the 1.2 μ m AOT from POLDER AOT retrieved at 550 nm, using an Ångström parameter exponent of 1.45, which was found in the spectral region from 325 to 1000 nm 45 for African biomass burning aerosols from SAFARI 2000 observations (Bergstrom et al., 2007; Russell et al., 2010). This way, an AOT at 1.2 μ m 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 the DRE ₅₀ was estimated at 21.7 Wm⁻¹ τ ⁻¹, 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 SCIA-MACHY and OMI-MODIS of up to $13~{\rm Wm}^{-2}$.

3.3.2 COT differences

The dependence of DRE on the COT of the cloud underlying the smoke can be large, depending on the AOT of the smoke. 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 $0.87 \mu m$, while COT from OMI-MODIS is retrieved at $1.2 \mu m$, which effectively is the MODIS channel measurement. However, the spectral variation in COT is very small and is only signifi-

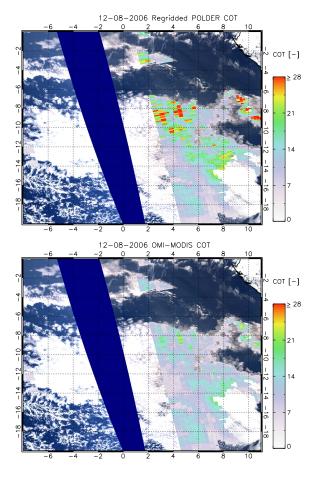


Figure 6. Cloud optical thickness (COT) overplotted on a MODIS RGB image on 12 August 2006 for POLDER at 0.87 μ m regridded to OMI grid (upper panel) and OMI-MODIS at 1.2 μ m (lower panel).

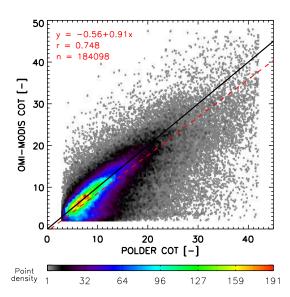


Figure 7. Scatterplot of POLDER COT gridded to the OMI footprint versus COT from OMI-MODIS data. The red dashed line shows the linear least-squares fit.

cant for very small droplets. For example, for cloud droplet effective radii of 4 microns the COT at 0.87 μ m is about 4% smaller than the COT at 1.2 μ m and this reduces for larger droplets.

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 10 higher by about 9% on average. Here, POLDER COT is regridded, while the MODIS radiances are averaged in the OMI footprint values were averaged over the OMI footprint, while for MODIS the radiances were averaged and one COT is retrieved for that OMI pixel. Therefore, the difference in 15 COT 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 stratocu-20 mulus, which appear as plane parallel clouds, but are characterized 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 stra-25 tocumulus 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 would lead to a change of about 9 Wm⁻². The average difference in COT is within the error estimate of the

Table 4. Average values of the OMI-MODIS COT and the POLDER COT and above-cloud AOT on 12 and 19 August 2006 between $20-0^{\circ}$ S; 8° W- 14° E. The DRE was calculated using the average AOT from POLDER and the average COT values for both OMI-MODIS and POLDER, assuming a CER of 8 μ m, an aerosol SSA of 0.840 at 550 nm and an aerosol geometric radius of 0.1 μ m.

	Max	⟨COT	POLDER	simulated	(DRE)
	COT	(CO1	$^{\prime}$ $\langle AOT \rangle$	DRE	\DKL/
OMI GRID					
12 August 2006					
POLDER	41.6	12.9	0.938	134.2	109.5
OMI-MODIS	37.5	10.0	0.938	107.1	76.3
19 August 2006					
POLDER	41.6	10.5	0.477	63.0	44.8
OMI-MODIS	47.7	8.9	0.4//	53.1	39.9

POLDER COT, but is more likely the result of the plane parallel bias.

Finally, the effect of different COT on the DRE retrieval was also computed using an RTM as used for the POLDER DRE calculations for the average values on the two se- 35 lected days. The average COT from both POLDER and OMI-MODIS on 12 and 19 August 2006 for collocated pixels were determined and the above-cloud DRE was calculated for OMI-MODIS and POLDER using their mean COT and the mean AOT retrieved by POLDER. Results are summa- 40 rized in Table 4. On 12 August 2006, the 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 134 Wm⁻² for POLDER and 107 Wm⁻² for OMI-MODIS. while the average DRE on this day was 110 Wm⁻² from 45 POLDER and 76 Wm⁻² from OMI-MODIS. On 19 August 2006, the mean COT from POLDER was 10.5, 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 and 53 Wm⁻² was obtained from POLDER and OMI-MODIS, 50 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 account for about 80% of the difference of 33 Wm⁻² between the 55 DRE from POLDER or OMI-MODIS shown in Figure 2.

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, both retrieved using DAA,

correlate very well, even though 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 DRE for moderate values, but is higher than SCIAMACHY and OMI-MODIS DRE for high values. 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 noon-normalized DRE. This is 30% of the maximum incoming solar irradiance. The DRE from SCIAMACHY and OMI-15 MODIS is limited to about 200–250 Wm⁻² for individual pixels.

The largest contribution to the difference between SCIA-MACHY, OMI-MODIS and POLDER DRE are sampling issues. Regridding SCIAMACHY and MODIS/OMI 20 OMI-MODIS 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 filtering based on COT and CF, which introduce can select high 25 positive DRE values and lead to large differences in the average DRE. 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 30 large footprints of SCIAMACHY and OMI remains, which is reflected in the less extreme COT and DRE values compared to POLDER. This difference was removed reduced by gridding POLDER to the coarser OMI grid, improving the comparison between OMI-MODIS DRE and POLDER DRE. Be-35 cause 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 40 DRE was 33 Wm⁻² on 12 August 2006, which can be explained by different estimates of AOT and COT using the various instruments.

In DAA, the AOT is assumed to be zero at $1.2~\mu m$, but was estimated from POLDER to be up to 0.6 in extreme ⁴⁵ 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 ($m_T > 1.47$) 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 ⁵⁵ 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 om on 12 August 2006, an overestimation of AOT by POLDER 2006. POLDER AOT may be overestimated, but this is difficult to establish quantify.

The COT has a strong influence on the aerosol DRE over clouds. The average POLDER COT is about 9% higher than that from OMI-MODIS in 2006. This difference can be caused by the plane parallel bias. Normally, MODIS COT retrievals at 0.8 and 2.1 μ m 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 μ m are used to derive COT and CER for 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 that day.

The 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 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 of this approach using DDA on OMI-MODIS spectra using but with POLDER COT yielded very erratic results. The reason is that the aerosol-free clouds cloud 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 90 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 (Zuidema et al., 2016), where the datasets in this analysis clearly show that the positive effect is significant and real. 95 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. POLDER on the other hand, takes advantage of the polarization measurements to accurately esti- 100 mate 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- 105 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 lo- 110

cal 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, 10 which is feasible for upcoming missions like 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 un-15 biased 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 plat-20 forms. This would allow time-dependent retrievals of UVabsorption by aerosols above clouds.

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

25 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.

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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.
- 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,

- 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., 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.: SCIAMACHY: Mission Objectives and Measurement Modes, J. Atmos. Sci., 56, 127–150, https://doi.org/10.1175/1520-0469(1999)056<0127:SMOAMM>2.\(\text{\text{\text{Q}}}\).CO;2, 1999.
- Bréon, F.-M. and Doutriaux-Boucher, M.: A comparison of cloud droplet radii measured from space, IEEETGR, 43, 1796–1805, https://doi.org/10.1109/TGRS.2005.852838, 2005.
- Cahalan, R. F., Ridgway, W., Wiscombe, W. J., Bell, T. L., and 70 Snider, J. B.: The Albedo of Fractal Stratocumulus Clouds, 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, 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, 80 https://doi.org/10.1038/NGEO437, 2009.
- 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.
- 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, 90 http://www.osapublishing.org/abstract.cfm?URI=josa-44-11-838, 1954.
- 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, 95
 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.
- 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,

https://doi.org/10.1175/1520-0442(1996)009<2802:DVOCCA>2.0.CO;22010. 1996.

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- de Graaf, M., Tilstra, L. G., Wang, P., and Stammes, P.: Retrieval of the aerosol direct radiative effect over clouds from spaceborne spectrometry, 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 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?, Atmos. Meas. Tech., https://doi.org/10.5194/amt-9-3607-2016, http://www.atmos-meas-tech.net/9/3607/2016/, 2016.
- 15 de Graaf, M., Tilstra, L., and Stammes, P.: Aerosol direct radiative effect over clouds from synergic OMI and MODIS reflectances, Atmos. Meas. Tech. Disc., 2019, 1 21, https://doi.org/10.5194/amt-2019-53, https://www.atmos-meas-tech-discuss.net/amt-2019-53/, 2019.
- 20 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 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.
- 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.
- 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.
- Forster, P., Ramaswamy, V., Artaxo, P., Berntsen, 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., Av-
- eryt, 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 dom-
- inated 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 wavelengthdependent aerosol absorption in biomass burning smoke in-

- ferred from Ozone Monitoring Instrument, Atmos. Chem. Phys., 60 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, 80 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, 90 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, 95 https://doi.org/10.1002/2015JD023128, 2015.
- 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.GO;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 POLD-ER/PARASOL measurements and estimation of their di- 105 rect 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 110 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.
- 5 Swap, R., Garstang, M., Macko, S. A., Tyson, P. D., Maenhaut, W., Artaxo, P., Kållberg, 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, 11707–11735, https://doi.org/10.5194/acp-10-11707-2010, 2010.
- 20 Wang, P., Stammes, P., van der A, R., Pinardi, G., and van Roozendael, M.: FRESCO+: an improved O₂ Aband cloud retrieval algorithm for tropospheric trace gas retrievals, Atmos. Chem. Phys., 8, 6565–6576, https://doi.org/10.5194/acp-8-6565-2008, 2008.
- 25 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 Vanbauce, C.: Retrieval of aerosol microphysical and optical properties above liquid clouds from POLDER/PARASOL polytics.
- 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, 11769– 11777, 2010.
- 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,
- 45 https://doi.org/http://dx.doi.org/10.1016/j.atmosenv.2013.02.017, 2013.
 - 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, 11245–11259, https://doi.org/10.5194/acp-12-11245-2012, 2012.
- Zhang, Z., Meyer, K., Platnick, S., Oreopoulos, L., Lee, D., and Yu, H.: A novel method for estimating shortwave 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.
- 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, 65 2016.