Response to reviewer 1

This manuscript addresses the important problem of what can be implied from the observed relationship between aerosols and cloud properties from spaceborne sensors. Most studies have, either explicitly or implicitly, assumed that positive correlations between aerosol optical depth (AOD) and cloud cover imply a causal relationship whereby the cloud changes are a response to the differing aerosol state. But we know that aerosols are themselves affected by clouds, most importantly through their removal by precipitation (here termed "wet scavenging"). This study uses a combination of observations and numerical modeling to show that aerosol properties are significantly impacted by wet scavenging, but that much of the impact of clouds on aerosols is confined to the region below the clouds themselves, which would not be detectable from space because these rely on clear-sky conditions. This is a generally well-written manuscript, with a few exceptions, describing a strong piece of work. This is one of a handful of recent papers that are undertaking the noble task of trying to better understand what processes are driving the AOD-cloud correlations seen in spaceborne data. I support its publication in Atmospheric Chemistry and Physics subject to some revision.

MAIN POINTS:

1.: *For the modeling study, the obvious omission to me is a case without wet scavenging, or at least one where is it significantly perturbed. For example, the low aerosol extinction values below cloud in Fig. 5 could simply be caused by penetrative downdrafts bringing clean air from aloft. Such downdrafts are quite common in deep convection. In addition, the conceptual model of Houze (1989) has downward moving air passing toward the storm center from the rear, and this too would be bringing air from aloft down to lower levels. How do the authors know that the low extinction values below the center of the precipitating cloud system are caused by wet scavenging? This is a fundamental tenet of the study and no evidence is provided to support it.*

Reply: We have since performed simulations of the same region with the wet scavenging of aerosols turned off. We find that there is an increase in AOD at the centre of the storm (due to aerosol humidification), rather than the decrease observed when wet-scavenging is active. A composite similar to that shown in Fig. 5 is also now included in the results section (Fig. 6). We believe that this shows that wet scavenging (at least in this model) is the leading cause of the reduction in AOD at the centre of the composite convective system.

2.: *The authors point out important differences in the "all-sky" aerosol assessments (e.g. from MACC) with the "clear sky" aerosol from MODIS, but the study really would benefit from a simulation with a high enough resolution that it could be used to directly compare AOD-cloud relationships for all-sky with clear-sky aerosol.*

Reply: We agree that the ideal experiment to perform would be a high resolution, convection resolving simulation of the tropics so that the all-sky and the clear-sky aerosol could be directly compared. Unfortunately, we do not have access to a simulation of the necessary length, resolution and domain size to compare the AOD-precipitation relationships in this way. Additionally, the relationships between precipitation and aerosols are not well understood, so it is not clear that a modelling study would be able to represent all the necessary processes. We hope that such a study could be performed in the future to gain a more complete picture of the differences between all-sky and clear-sky AOD.

3.: *The shift from the modeling study (section 3.1) to observations (3.2) is too abrupt. The observations are not even from the same region as the modeling study, but seem to include the entire tropics, most of which is over the ocean rather than the land in the observational study. I find this switch to be quite confusing. I couldnt really follow the arguments about wet scavenging in section 3.2. The text in this section appears to fall into the "causality trap" that the authors are trying to warn against. Maybe I missed some key point, but I found this part very confusing as to what the authors are trying to say.*

Reply: We have separated the observational part into a new section and improved the link between the sections, providing a mini-introduction as to why it is necessary and what it is trying to show.

The observational part of this work tries to avoid meteorological covariations being responsible for the observed relationships (and the associated "causality trap") by normalising by CF at the time of the AOD retrieval (along with other meteorological parameters). This normalising by CF aims to reduce the influence of aerosol humidification, so that any underlying relationship between aerosol and precipitation can be examined (Gryspeerdt et al., 2014a). Here, we use MACC to provide a modelled "all-sky" AOD. Although we are limited from demonstrating that the "clear-sky" is definitely more useful by our inability to simulate the whole tropics at the necessary resolution, this observational study provides some evidence that the "clear-sky" AOD is more useful, as it is able to detect an effect consistent with aerosol invigoration, while the "all-sky" AOD is not (it is heavily affected by wet scavenging).

4.: *The post-storm "wake" of low AOD seen in Fig. 5 seems to warrant more mention. How much further behind the storm does it extend (it reaches the edge of the composite domain and so would be expected to extend further). Surely, these wet-scavenged clear sky AOD values would be observable from space. For example, P6870, line 6- 8. The authors appear to be arguing that using clear sky AOD is better for inferring aerosol impacts on precipitation. But dont such clear sky measurements include the storm wake, where scavenging has played an important role? The authors need to back up their assertion that clear sky AOD is more useful with model simulations that demonstrate it. It seems far too speculative to me. Why is meteorological covariation not an issue?*

Reply: It is hoped that these wet scavenged regions are visible from space and this is a topic of current research. However, the impact of the post-storm "wake" on the surrounding clear-sky AOD is much smaller than the impact of precipitation on the "cloudy-sky" AOD. Whilst the reduced AOD region make up around 30% of the "cloudy-sky" region, it is perhaps 10% of the "all-sky" region and a smaller proportion still of the "clear-sky" region. Some previous studies have shown that the clear-sky AOD correlates more strongly with precipitation than the all-sky AOD (Grandey et al., 2014), but much of this effect could be due to aerosol humidification or meteorological covariations. The "wake" itself does extend further than the edge of the domain, it keeps the same width to around 300 km behind the composite system, where it then becomes difficult to distinguish. However, it is unclear how far it actually extends in practice, as effects due to the finite size of the simulation domain start to play a role. A comment on this has been included in the discussion section (Sect. 4.1).

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5. P6871, line 17.: *What on earth is an invigoration-like effect? Is there a wet scavenging-like effect to parallel this? What would it be?*
Reply: This was a rather clumsy way of referring to the increased precipitation from the high AOD population at times after the AOD retrieval, which is consistent with, but not necessarily due to, aerosol invigoration.

OTHER ISSUES:

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1. P6853, line 27.: *Chand et al. (2012) concludes the same as the Quaas and Grandey papers.*
Reply: Added

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2. P6847, line 8-10.: *Why is the indirect effect of aerosols on clouds expected to be much weaker than the wet scavenging effects? I thought that the whole idea is that understanding the wet scavenging effects is needed to help understand the indirect effects. If the latter are negligible then what is the purpose of this study?*
Reply: We expect the influence of wet scavenging on aerosols to be stronger than the effects of aerosol on precipitation following previous studies investigating the AOD-CF and AOD-precipitation relationship using GCMs (Quaas et al., 2010; Grandey et al., 2013, 2014). As wet scavenging is the primary method for removing aerosols from the atmosphere, but aerosols are not thought to be the primary control on precipitation occurrence, we feel this is a reasonable assumption. The purpose of investigating wet scavenging in this work is to improve our understanding of how it controls the relationship between AOD and cloud properties, to better understand the difference between models and satellite observations. This then leads to improved estimates of the influence of aerosols on clouds.

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3. P6849, line 14.: *data are*
Reply: Amended

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4. Figure A2.: *I would have expected more water than dry aerosol mass. Radius growth factors of 2 are not uncommon, which implies almost an order of magnitude more water in the aerosol than other material.*
Reply: It is possible that the low water content is due to a large amount of hydrophobic aerosol in this region, as the main aerosol source is biomass burning. The weak relationship between AOD and precipitation rate in the Congo region

in Fig. 1 might also suggest that aerosol humid growth is weaker here (or wet scavenging is stronger), perhaps due to the aerosol content. This low growth factor would not affect the results in this work relative to wet scavenging, but might result in a lower increase in AOD due to aerosol humidification in the cloudy regions compared to the global average.

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5. P6864, line 15.: *What "regimes" are being discussed here?*
Reply: These are the different cloud regimes included in Fig. 6. This has now been amended.

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6. P6865, line 22:: *How exactly is wet scavenging "observed" here? Also, P6866 line 25 talks of "observation of wet scavenging", but I dont know what observation they are referring to.*
Reply: Unfortunately, we cannot observe the physical scavenging of the aerosol, so this is really "observations consistent with wet-scavenging". The observations referred to are the plots in Fig. 6. Clouds which are observed to have a low AOD at the time of the AOD retrieval are found to have a higher mean precipitation rate over the previous 12 hours than clouds with a high retrieved AOD (the blue line is above the red line). While this is not conclusive, it is the relationship that would be expected if wet scavenging was significantly impacting the aerosol population.

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7. P6866, line 3-4.: *How is the invigoration effect "observed" here? Are the authors concluding the opposite of their title statement? I was losing steam by this point and was a little confused about whether the authors are arguing for an invigoration effect in shallow cumulus which probably dont even contain ice (and therefore how can there be an invigoration effect as outlined by Rosenfeld?).*
Reply: We thank the reviewer for pointing this out. This part of the paper appears to be short on some of the explanation that was necessary in previous work. The important point here is that while the regimes are labelled as "shallow cumulus" and "thick mid level", these names only refer to the cloud types at the time of the aerosol retrieval (as that is also when the cloud properties are retrieved). The clouds can develop after the aerosol retrieval and the observed increase in precipitation is likely to come from the clouds that transition into the deep convective regime. As shown in Gryspeerdt et al. (2014a), there is little change in precipitation from clouds which remain below the freezing level, implying (as the reviewer has pointed out) that occurrence of ice is a important factor. As with the wet scavenging, this is only a result that is consistent with aerosol invigoration, rather than an observation of it. Further explanation has been included in the text to explain the names of the regimes and the possibility of regime transitions occurring after the AOD retrieval.

Response to reviewer 3

OVERVIEW This manuscript examines the relationship between AOD and precipitation in GCMs and satellite retrievals. Satellite retrievals show a positive correlation between clear sky AOD and adjacent precipitation in most tropical and subtropical locations, but AOD and precipitation retrievals are non-coincident. In GCMs, AOD is an “all sky” variable rather than a “clear sky” variable, and the AOD-precipitation relationship is different than in satellite retrievals. With the aid of a 3-week mesoscale simulation of deep convective systems in the Congo, the authors show that this is a result of wet scavenging of aerosols. The aerosols ingested into the deep convective updrafts come from nonscavenged regions, which supports the use of satellite clear sky retrievals of AOD in inflow regions adjacent to convective clouds and precipitation. Further support comes from the lack of a positive AOD-precipitation relationship in a high CF thick mid-level cloud regime when the MACC reanalysis AOD is used instead of MODIS AOD. The strongest part of the paper is the demonstration of wet scavenging producing different AOD-precipitation relationships in GCMs and satellite retrievals. The weakest is the hand wavy discussion of invigoration in shallow cumulus and thick mid-level cloud regimes, as discussed more in comments below. Overall, this is a manuscript on the important but complicated topic of aerosol impacts on precipitation, and it is worthy of publication in ACP, but only following revisions.

MAJOR COMMENTS

1.: *Sections 2.3 and 3.2 are confusing, and there needs to be better transitions from and connections to the simulation result sections (e.g., Sections 2.2 and 3.1). Here are a couple examples from the observations results that did not make sense to me:*

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a: . *You ensure that high and low AOD quartiles have the same distributions of CF and meteorology at T+0 for different cloud regimes and claim that changes in precipitation before and after T+0 are the result of aerosol interactions. Why cant meteorology or CF change before or after T+0 and be responsible for differences in precipitation rather than aerosols?*

Reply: This is a good point, which Gryspeerdt et al. (2014a) attempted to cover. The normalisation by CF is really intended to assist in normalising by meteorology, as it may be a better indicator of local relative humidity than the reanalysis RH. It is true that the normalisation at the time of the AOD retrieval may not actually restrict the local meteorology enough to conclude that the resulting precipitation development changes are due to an aerosol impact of cloud processes. However, it appears that the normalisation (especially by CF) does act to significantly reduce the impact of meteorological covariations. An analysis of the development of 500hPa vertical velocity before and after the AOD retrieval indicated that the normalisation by meteorological parameters removed any significant difference in the development of the meteorological parameters, indicating that normalising at T+0 is a reasonable attempt at removing the influence of meteorological covariations (Gryspeerdt et al., 2014a).

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b.: *Controlling for 850 hPa relative humidity, 500 hPa pressure vertical velocity, 10 m/s wind speed, and LTSS (from Gryspeerdt et al. 2014) does not control for all meteorological factors that impact precipitation. What about variables* *that directly impact rainfall such as precipitable water or variables that directly impact convection such as CAPE and vertical wind shear? Without controlling for these variables, the claim that an aerosol invigoration effect is occurring is unsubstantiated.*

Reply: Obviously to demonstrate that aerosol is responsible for the precipitation increase, it is necessary to normalise by all atmospheric variables. Unfortunately, this restricts the data volume too severely and so we had to make some choices. The variables chosen (850 hPa RH and 500hPa vertical velocity) were picked because of their previous links to both aerosol and cloud processes. While CAPE is important for convective cloud processes, its connection to aerosol and AOD is much more tenuous.

The normalisation by CF actually does the majority of the "work", providing a significant advance over previous attempts to account for meteorological covariations, although it is still possible that aerosol humidification is playing a role in the observed relationships. From previous labbooks, normalisation by ECMWF precipitable water was tried in Gryspeerdt et al. (2014a), but it had much the same effect as normalising by 850hPa RH and so was omitted from the final analysis.

c.: *If there were an aerosol invigoration effect, youd expect to see it in the deep convective regime where updrafts are strong enough to loft liquid into the mixed phase zone so that freezing can be enhanced. An increase in aerosols should reduce the probability of warm rain precipitation in shallow cumulus (since they dont contain ice) because increased CCN reduces cloud droplet size, so aerosol invigoration of precipitation doesnt make sense for these clouds. Thick mid-level clouds are presumably stratiform rainfall in mesoscale convective systems and frontal type systems. Again, little cloud water is lofted in this cloud type, so how does aerosol invigoration operate?*

Reply: This is due to the method of naming the regimes, something which isn't covered well in this paper (and so has been added to the methods section). The issue is that the cloud types are determined at the same time as the AOD retrieval. This is the only time they are guaranteed to be in this regime (and they often transition between regimes over a short period of time due to the diurnal cycle - Gryspeerdt et al., 2014b). The shallow cumulus clouds themselves probably aren't the ones generating the increase in precipitation, indeed, Gryspeerdt et al. (2014a) showed that if the cloud tops are restricted to being below the freezing layer, there is relatively little difference between precipitation development of the high and low AOD populations (highlighting the importance of cloud ice). It is also important to note that the clustering method used to determine the regimes does not limit the shallow cumulus regime to only shallow clouds, some clouds with higher tops are included.

d.: *To examine large deep convective systems in the Congo with a simulation and then global shallow cumulus and thick mid-level clouds regimes with observations is part of what contributes to the confusion when transitioning between sections of the paper. Since most of the precipitation in the tropics is from deep* *convection, and this is the regime where one would theoretically most expect an aerosol invigoration effect, why not include this regime in the observational analysis?*

Reply: As noted above the observations do study the precipitation from deep convective clouds, but this is not obvious, as the names of the regimes are determined by the cloud type retrieved at T+0. The explanation of this has been improved in the method and observations sections.

2.: *Can you clarify what you mean by an invigoration-like effect? Im assuming that you are referring to the dynamical invigoration of convection by increased lofting and freezing of cloud water (as hypothesized by Rosenfeld), but there are a lot of steps missing between what you have shown and concluding that this type of an effect is increasing precipitation. You can certainly hypothesize reasons for increased precipitation with increased clear sky AOD, but you shouldnt be so conclusive without more evidence shown, and you should more clearly lay out what invigoration means and how it might lead to more precipitation. In my mind, I dont see why meteorological conditions that you did not control for at T+0 (see 1b) cant be correlated with AOD and lead to increased precipitation just as easily as the aerosols themselves.*

Reply: The term “invigoration-like effect” is just a clumsy way of referring to the increased precipitation after T+0 in the high AOD population. It has been changed where it occurs to refer to an “apparent invigoration effect”.

3.: *Convective downdrafts bring down cleaner air from mid levels into the boundary layer. Can you show that this effect is small relative to wet scavenging in your WRF simulation? Also, although not an issue over the rainforest in the Congo, convective system outflow in arid regions (e.g., the Sahel) often generates large amounts of dust that can increase AOD (e.g., Flamant et al. 2007 (http://onlinelibrary.wiley.com/doi/10.1002/qj.97/abstract)), so the simulation in the Congo region may not be universally representative.*

Reply: A composite from a simulation without wet scavenging has been included in section 4 of the paper. It shows that without wet scavenging, the reduction in AOD at the centre of the storm is not present.

MINOR COMMENTS

1.: *This is not a major gripe, but I dont think the title of the paper fits the results you show. First, wet scavenging is an aerosol-cloud-precipitation interaction. Second, you are not examining all aerosol-cloud-precipitation interactions. You are primarily concerned with aerosol-precipitation interactions. Third, these interactions are not being detected by satellite. Correlations are being detected. And lastly, detection isnt limited in satellite retrievals. It is limited in GCMs. A more specific title would be something like “Wet scavenging limits the detection of aerosol effects on precipitation in GCMs” or “Wet scavenging produces different relationships between AOD and precipitation in satellite retrievals and GCMs”.*

Reply: Although it is discussed that this might limit the detection in GCMs, the results in this work show the detection of these relationships being limited in a reanalysis/observations combined study, so specifying GCMs in the title might not be correct. Indeed, due to their ability to run multiple realisations of the same climate with different aerosol perturbations, it is relatively straightforward to detect aerosol effects on precipitation in models. However, we agree with the point that wet scavenging is an interaction between aerosol and precipitation. The title has therefore been changed to “Wet scavenging limits the detection of aerosol effects on precipitation.”

2.: *You mention the uncertainty associated with modes of convection that are different than your composite mode from the WRF simulation, but what about uncertainty in the scavenging of aerosols that contribute to AOD that are not present in the Congo? Is scavenging of biomass burning aerosols representative of scavenging of other aerosols such as dust? Is scavenging of boundary layer aerosols representative of scavenging of free tropospheric aerosols plumes?*
Reply: This is a good point and one not really covered in this work. However, the previous work by Grandey et al. (2014) using a GCM suggests that wet scavenging is also important at larger scales. They find that when running a GCM with and without convective wet scavenging, the relationship between AOD and precipitation in the model is reversed, almost globally. As this occurs even over ocean, and far from the main aerosol sources, this would in turn suggest that wet scavenging is also important for aerosols that have been transported long distances or in the free atmosphere. This has now been noted in the results (Sect. 3.2).

3.: *Does your WRF simulation reintroduce aerosols into the atmosphere when cloud and rain droplets evaporate? If not, removal of aerosol could be overestimated.*
Reply: The convective transport scheme, which handles the convective wet scavenging, also reintroduces aerosol from evaporating rainwater.

4.: *Do you have a citation for the last sentence of the second paragraph in Section 4 (stating that aerosol hygroscopic growth generates much of the positive correlation between clear sky AOD and precipitation)?*
Reply: We have now included the citations (Boucher and Quaas, 2012; Grandey et al., 2014).

5.: *It seems strange to have a Section 4 (Discussion) and Section 4.1 (Comparison to GCM processes) without a 4.2. Maybe change Section 4.1 to Section 5 or have the first part of Section 4 as Section 4.1 with the comparison to GCM processes as Section 4.2.*
Reply: Amended

6.: *Can you explain what you mean by relative frequency of occurrence (RFO)?*
Reply: This term was from earlier work and has been removed as it occurs only once. The sentence now reads: “These high CF/strongly precipitating regimes occur rarely, with only 13% of the cloud regime occurring in the tropics

falling into the deep convective or thick mid-level regimes (Gryspeerdt and Stier, 2012)."

7.: *At the beginning of Section 4.1, you should change air is drawn into convective updraughts from non-precipitating regions to air is usually drawn into convective updraughts from non-precipitating regions.*
Reply: Amended

8.: *Can you provide a citation for the last sentence in the second paragraph of the conclusions (aerosol hygroscopic growth primarily causes the increased in AOD with increasing precipitation in scenes with low precipitation)?*
Reply: This is shown in supplementary Figure A1 (a reference to this figure is now included). There is not a specific study that we are aware of that covers this for low precipitation rate situations, but it is covered more generally by (Boucher and Quaas, 2012) and (Grandey et al., 2014) who examine the link between AOD and precipitation.

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Wet scavenging limits the detection of ~~aerosol–cloud–precipitation interactions~~aerosol effects on precipitation

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Abstract. Satellite studies of aerosol–cloud interactions usually make use of retrievals of both aerosol and cloud properties, but these retrievals are rarely spatially co-located. While it is possible to retrieve aerosol properties above clouds under certain circumstances, aerosol properties are usually only retrieved in cloud free scenes. Generally, the smaller spatial variability of aerosols compared to clouds reduces the importance of this sampling difference. However, as precipitation generates an increase in spatial variability, the imperfect co-location of aerosol and cloud property retrievals may lead to changes in observed aerosol-cloud-precipitation relationships in precipitating environments.

In this work, we use a regional-scale model, satellite observations and reanalysis data to investigate how the non-coincidence of aerosol, cloud and precipitation retrievals affects correlations between them. We show that the difference in the aerosol optical depth (AOD)-precipitation relationship between general circulation models (GCMs) and satellite observations can be explained by the wet scavenging of aerosol. Using observations of the development of precipitation from cloud regimes, we show how the influence of wet scavenging can obscure possible aerosol influences on precipitation from convective clouds. This obscuring of aerosol-cloud-precipitation interactions by wet scavenging suggests that even if GCMs contained a perfect representation of aerosol influences on convective clouds, the difficulty of separating the "clear-sky" aerosol from the "all-sky" aerosol in GCMs may prevent them from reproducing the correlations seen in satellite data.

1 Introduction

Aerosols have an important influence on cloud properties by providing cloud condensation nuclei (CCN). An increased number of CCN can lead to an increase in cloud droplet number concentration and a reduction in droplet size (Twomey, 1974), which in turn has been hypothesised to lead to a reduction in precipitation (Albrecht, 1989). Theoretical (Williams et al., 2002; Rosenfeld et al., 2008; Stevens and Feingold, 2009) and modelling studies (Khain et al., 2005; Tao et al., 2007) have suggested that under certain conditions, this liquid-phase suppression of precipitation may lead to an invigoration of convective clouds through the additional release of the latent heat of freezing. An invigoration of convective clouds may in turn lead to an increase in precipitation from the cloud in later stages of its lifecycle.

Observational studies have detected positive correlations between aerosols and precipitation that might indicate aerosol invigoration of convective clouds (Lin et al., 2006; Li et al., 2011; Koren et al., 2012; Niu and Li, 2012; Gryspeerdt et al., 2014b). These studies generally show an increase in precipitation with increase in a CCN proxy (aerosol optical depth (AOD – Andreae and Rosenfeld, 2008) or aerosol index (AI – Nakajima et al., 2001). However, given that precipitation is responsible for the removal of the majority of atmospheric aerosol (Textor et al., 2006), wet scavenging might be expected to generate a strong negative correlation between AOD and precipitation. Although this negative correlation is not observed in satellite studies, it can be observed in global models (e.g. Fig. 1c), especially in regions of high precipitation.

Correlations between aerosol and cloud properties have been shown to be strongly influenced by meteorological covariation and retrieval errors (Zhang et al., 2005; Mauger and Norris, 2007; Wen et al., 2007; Chand et al., 2012). Evidence from global models suggests that the positive correlation between AOD and precipitation rate (Fig. 1a) is largely due to aerosol hygroscopic growth, resulting in an AOD covariation with relative humidity (Boucher and Quaas, 2012; Grandey et al., 2014). Along with aerosol hygroscopic growth, retrieval errors such as cloud contamination of AOD retrievals (Zhang et al., 2005) can also lead to a positive correlation between AOD and cloud properties. Retrieval errors and aerosol hygroscopic growth together have been shown to be responsible for the majority of the positive correlation between AOD and cloud fraction (CF) ~~(Quaas et al., 2010; Grandey et al., 2013)~~ (Quaas et al., 2010; Chand et al., 2012; Grandey et al., 2013) . Influences on the AOD–CF correlation are particularly important, as the strong correlation between CF and other cloud parameters (including precipitation) can generate correlations between AOD and these cloud parameters (Gryspeerdt et al., 2014a).

While observational studies have shown an increase in precipitation with increasing AOD, this correlation is not always found when using general circulation models (GCMs). The difference between models and observations is demonstrated in Fig. 1, where each of the subplots shows the difference in precipitation rate between the highest and lowest quartiles of AOD over five years of data. Figs. 1a,b use precipitation data from the TRMM merged precipitation dataset (Huffman et al.,

2007) between 2003 and 2007, but different AOD products. Fig. 1a uses the MODIS AOD product (Remer et al., 2005) and Fig. 1b using the MACC reanalysis AOD (Morcrette et al., 2011). For comparison, the same analysis is performed on a 5-year simulation from the HadGEM3-UKCA GCM (Mann et al., 2014), showing similar results to the ECHAM-HAM GCM (Grandey et al., 2014).

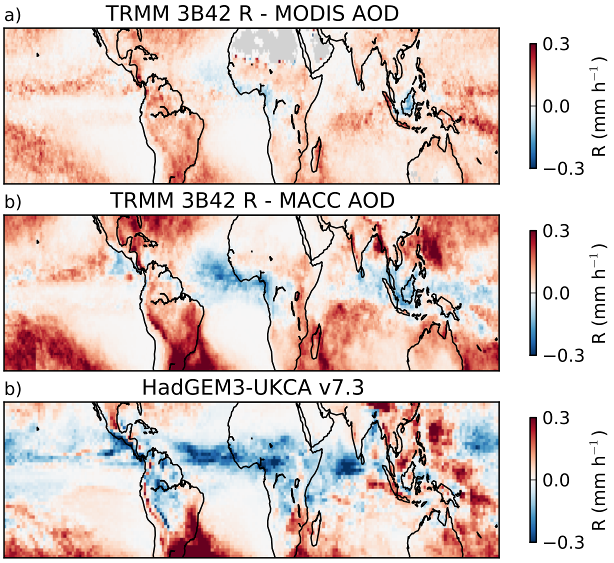

Figure 1. The difference in the TRMM merged precipitation rate between the highest and lowest AOD quartiles when using (a) MODIS and (b) MACC AOD between 2003 and 2007. c) The same as (a) but using 5 years of HadGEM-UKCA precipitation and AOD. Red(blue) indicates an increase(decrease) in precipitation for the high AOD population.

Meteorological covariations partially disguise the negative relationship between AOD and precipitation that exists as the result of the wet scavenging of aerosol (Quaas et al., 2010; Grandey et al., 2014). However, as models are expected to reproduce covariations between aerosol and cloud properties, these covariations are unlikely to be the cause of the difference in the AOD–precipitation correlation between models and observations seen in Fig. 1. Previous studies have suggested that the difference is due to the different sampling between models and observations (Grandey et al., 2013, 2014). Understanding the impact of sampling on modelled and observed aerosol–cloud–precipitation correlations (Fig. 1) is important for determining the strength of the aerosol influence on clouds and precipitation.

Whilst there are some instruments (e.g. Winker et al., 2007) and algorithms (Jethva et al., 2014) that can retrieve the properties of aerosols above or below cloud, the most commonly used satellite retrievals of aerosol properties are only performed in cloud-free skies. On the other hand, GCMs are able to determine the aerosol concentration in cloudy skies, and so can determine the AOD in cloudy or precipitating scenes. This variation in sampling means that the aerosol seen by a model or the "all-sky" aerosol may be very different from the satellite sampled or "clear-sky" aerosol, especially in strongly precipitating locations. Almost all observational studies of aerosol–cloud–precipitation interactions use "clear-sky" sampling and studies using GCMs use the "all-sky" sampling. This means it is vital to account for the discrepancies caused by the differing sampling if observational studies are to be used in constraining aerosol–cloud interactions in GCMs.

Most GCMs only carry the "all-sky" aerosol between timesteps, meaning that GCMs effectively assume that each gridbox is well mixed over a period equal to that of the model timestep (usually 10–30 minutes). Within this limitation, GCMs take some steps to determine a "clear-sky" AOD, taking into account the wet scavenging that has occurred during a model timestep to diagnose a "clear-sky" AOD. Some GCMs also take account of the variation in relative humidity (RH) between in-cloud and out-of-cloud locations when diagnosing the AOD (e.g. Stier et al., 2005), resulting in a difference between the "all-sky" and the "clear-sky" AOD within a GCM gridbox. However, as wet scavenging affects the CCN population rather than just the AOD, accounting for RH variations does not account for the underlying CCN (and AOD) variations caused by precipitation. Throughout this work we refer to the difference in sampling between GCMs and satellites, but any process which prevents the separation of "clear-sky" aerosol from "all-sky" aerosol in GCMs (such as assumptions about mixing) can generate these results.

This work focuses on possible aerosol interactions with precipitation from convective clouds, using regional-scale models, reanalysis and satellite data to investigate the impact of aerosol sampling on the AOD–precipitation relationship. A high resolution model is used to examine the impact of only retrieving AOD in cloud free locations on the mean AOD. A composite convective system from this model is used to examine the impact of heavily precipitating systems on AOD in the neighbourhood of these systems and to investigate the detectability of aerosol–cloud interactions by satellites. We use the precipitation development method of Gryspeerdt et al. (2014b) together with satellite and re-analysis (to provide a model-like observational product) AOD products to investigate the link between aerosol and precipitation in observations while accounting for meteorological covariations. Combining the results from these methods, we show how wet scavenging can impact the detectability of aerosol influences on precipitation from convective clouds.

2 Methods

2.1 Model setup

We use v3.4.1 of the Weather Research and Forecasting (WRF)-Chem model (Grell et al., 2005) with a 10 km horizontal grid length. Although this is not sufficient to resolve small-scale convective features, it is able to resolve the larger precipitating systems that impact aerosol in this region. A model grid length of 10 km requires a cumulus parametrisation, in this study we use the Grell 3D ensemble scheme (Grell, 2002). We use 30 vertical levels and the standard WRF stretched vertical

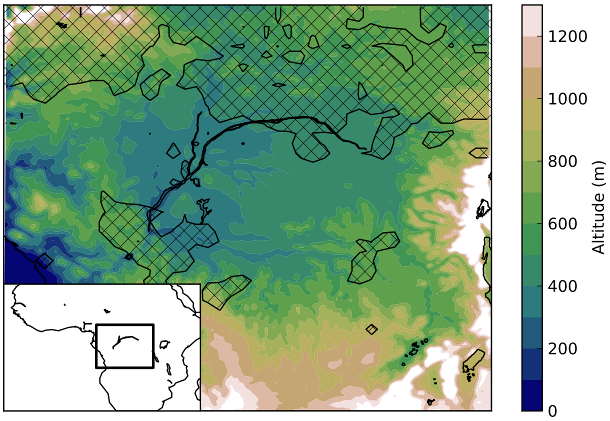

Figure 2. The domain used for the WRF-Chem simulations in this study. The colours indicate the altitude and the hatched areas indicate regions where MODIS detects more than one fire per 5,000 km^2 over the simulation time period. The inset shows the domain location over Africa.

grid, with grid spacing of about 100m in the lower levels, increasing towards the upper levels. This provides sufficient resolution to resolve the vertical structure of the aerosol and precipitation within our study region. To provide the atmospheric heat and moisture tendencies, microphysical rates and surface rainfall, we use the 5-class prognostic Lin microphysics scheme (which includes snow, graupel and mixed-phase processes – Lin et al., 1983). Longwave and shortwave radiation in the model are parametrised by the RRTM (Mlawer et al., 1997) and Goddard shortwave (Chou and Suarez, 1994) schemes respectively. The model domain covers a 2100 km by 2100 km region over the Congo Basin (Fig. 2), chosen due to the highly convective nature of this region and the strong sources of biomass burning aerosol (Fig. 2). The study region incorporates the Congo Basin and a large fraction of the biomass burning region to the north of it (Fig. 2). The model initial and boundary conditions are generated from NCEP reanalysis, starting at 0000 UTC on 01 March 2007 and updated every 6 hours over the three week simulation. The simulation period was selected due to the peak in precipitation in the Congo basin during March and April (Washington et al., 2013).

All of the aerosol in this semi-idealised setup is generated by emissions within the domain. Although simulations of a larger domain (not shown) indicate that a significant amount of the aerosol is transported into the study region from outside, there are sufficient aerosol sources inside the study region so that the influence of precipitation on AOD can be studied. We use the MADE-SORGAM aerosol module (Ackermann et al., 1998; Schell et al., 2001) and include cloud chemistry so that the wet scavenging of aerosols by the stratiform precipitation is represented. This allows aerosols to influence the cloud droplet number concentration. However, the influence of an aerosol indirect effect on the AOD-precipitation relationship in this study is expected to be small compared to wet scavenging. Convective wet scavenging of aerosol is included in the convection scheme. The main variability in emissions over the three week study period comes from biomass burning. Anthropogenic emissions using the EDGAR and RETRO databases and biomass burning emissions using daily updated MODIS fire counts (MCD14ML – Giglio et al., 2003) are generated using PREP-CHEM-SRC (Freitas et al., 2011). Biogenic emissions are generated using the Guenther emissions scheme (Guenther et al., 1994), but emissions from biomass burning dominate the AOD in this region.

2.2 Storm composites

To investigate the influence of precipitation on aerosols through wet scavenging, we identify regions of heavy precipitation, specifically convective storms. We then composite these storms, rotating them onto a common direction of travel, so that the properties of these systems and their influence on the AOD can be investigated.

We define our systems using the hourly accumulated precipitation field. We consider a heavy precipitation rate as greater than 2 mm hr^{-1}, which results in easily separated precipitating systems, without overly restricting the number of these systems. Heavily precipitating, four-connected (two gridboxes are considered joined if they share an edge, not if they only share a corner) gridboxes are then joined together to produce precipitating "blobs".

To determine the direction of travel of a system, the blobs are filtered to select cases that are easy to track, which removes the majority of detected blobs. Only blobs with an area greater than 3,000 km^2 and less than 15,000 km^2 are retained. Blobs are discarded if they are insufficiently independent of other blobs (forming less than 90% of the precipitating area within 50 km of the blob edge), if they are within 50 km of the domain edge or if they fail to meet circularity criteria. As the blobs are selected to be independent of each other, the position of the blob after one hour is selected as the largest blob within 100 km of the starting position. Over the 21-day simulation, 51444 blobs are found, of which 37 are retained to form the system composite. The direction of travel and velocity are determined from the motion of the storm over a single hour following its detection.

2.3 Observations

The strong link between AOD and CF can generate correlations between AOD and other cloud or precipitation properties (Gryspeerdt et al., 2014a). Here we use the precipitation development method of Gryspeerdt et al. (2014b), which is explicitly designed to account for these covariations, to examine the links between aerosol and precipitation.

The precipitation development method makes use of sub-daily time-resolved precipitation measurements and the diurnal cycle of precipitation, to investigate the link between satellite retrieved aerosol and precipitation. The data ~~is~~ are separated into different cloud regimes ~~(Gryspeerdt and Stier, 2012) and~~ using the clusters defined in Gryspeerdt and Stier (2012) . The high and low aerosol populations are determined as the highest and lowest AOD quartiles for each regime and season. Meteorological covariations and the strong influence of CF are accounted for at the time of the aerosol retrieval (T+0), by ensuring that the high and low AOD populations have the same distribution of CF and meteorological parameters, as described in Gryspeerdt et al. (2014c). This almost completely removes the correlation between AOD and precipitation at T+0, while the different development of precipitation at times before and after T+0 for the high and low aerosol populations demonstrates the interaction of aerosols with precipitation. This method reduces some of the largest confounding factors when studying aerosol–cloud interactions. A full description of the precipitation development method is given in Gryspeerdt et al. (2014b).

We use precipitation data from the TRMM 3B42 merged precipitation product (Huffman et al., 2007). This product merges precipitation estimates from radar, passive microwave, geostationary infra-red and surface rain gauges to give three-hourly estimates of the precipitation across the tropics. The cloud and aerosol data used are from the MODIS collection 5.1 (Platnick et al., 2003; Remer et al., 2005) level 3 product, with only the dark-target aerosol being used. These data are all gridded to 1° by 1° resolution. To increase the number of available aerosol retrievals in cloudy regions, AOD data is interpolated into gridboxes that have no AOD retrievals if those gridboxes have a neighbour where AOD data exists, following Koren et al. (2012). The interpolation does not generate AOD data for all overcast locations, but it does increase the number of available retrievals in cloudy regions. The MODIS data is used to determine cloud regimes at the time of the aerosol retrieval (T+0), separating cloud with different properties. High aerosol is defined as the highest AOD quartile and low as the lowest quartile. These quartiles are determined for each regime, location and season separately.

Defining the MODIS Aqua overpass time (1330 local solar time – LST) as T+0, we investigate the development of the precipitation for each of the regimes, at times before and after the AOD retrieval. The high and low AOD populations are sampled so that they have the same CF distribution (see. Gryspeerdt et al., 2014c) to remove the AOD–CF relationship at T+0. This is important due to the ability of the AOD–CF correlation to generate correlations between aerosol and other cloud properties (Gryspeerdt et al., 2014a). In this work, we consider only two regimes. The shallow cumulus regime is a low CF regime and the thick mid-level regime is a high CF regime. Both of these regimes showed evidence of the wet scavenging of aerosol and of possible aerosol invigoration of convection in previous work (Gryspeerdt et al., 2014b). It is important to note that the regimes are named for their properties at T+0, as this is when the cloud properties are retrieved. There are often transitions between the regimes over time, so several hours after T+0, a shallow cumulus regime may have transitioned into the deep convective regime (Gryspeerdt et al., 2014c) . An increase in precipitation after T+0 in the shallow cumulus regime is likely the result of transitions to more heavily precipitating regimes rather than an increase in precipitation from the shallow cumulus clouds themselves.

As we cannot use satellites to sample aerosol in cloudy regions in the same style as a GCM, we use the ECMWF MACC product (Benedetti et al., 2009) to provide an “all-sky” AOD product. The

MACC project assimilates aerosol information from MODIS into the ECMWF integrated forecast system, and so can also provide an AOD estimate in overcast or precipitating scenes where there is no MODIS AOD retrieval. In cloud free regions, MACC is largely similar to MODIS, but as the CF increases, MACC increasingly has to rely on its own modelled estimates of AOD, especially in overcast regions where there are no AOD retrievals to be assimilated. This makes it a suitable replacement for a study using only GCMs, as it provides a model-like "all-sky" AOD for the real world. Due to the resolution of the MACC product and instantaneous mixing of aerosol over each gridbox every timestep, the wet scavenging of aerosols effectively takes place across an entire gridbox. This prevents MACC from providing a separate "clear-sky" AOD..

As the MACC AOD product is specified at 0300 UTC (a three hour forecast from 0000 UTC), we interpolate consecutive days to generate a 1330 LST MACC AOD product. Although this interpolated product cannot reproduce the diurnal cycle of AOD, this cycle is much smaller than the diurnal cycle of precipitation (which is captured). Validating MACC (or interpolated MODIS) in cloud covered or precipitating regions is not the focus of this paper. As precipitation in global models can be unrealistic (Stephens et al., 2010), we use the TRMM 3B42 precipitation data to generate precipitation development plots when using the MACC AOD data.

3 Results

3.1 Regional relationships

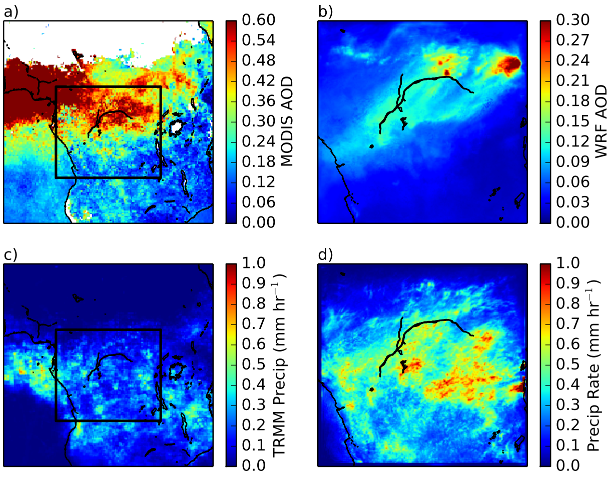

Figure 3. a) The mean Aqua MODIS AOD in equatorial Africa for March 2007. The box shows the study region. b) The mean AOD at 1330 local time for March 2007 from WRF-Chem. Note the change of color scale. c) The mean TRMM precipitation rate and (d) the mean WRF-Chem precipitation rate for March 2007.

The WRF-Chem simulation shows a strong aerosol plume heading diagonally from north-east (near the main biomass burning regions) to the south-west, following the direction of the prevailing wind (Fig. 3b). With a maximum AOD of around 0.3, this is lower than, although a similar order of magnitude to, the MODIS retrieved AOD. The spatial pattern is similar to MODIS, with a lower AOD in the southern part of the domain, although there is a noticeable difference due to the lack of aerosol being advected in from outside the domain in WRF-Chem. This semi-idealised setup does not influence our later results, as they depend on the interaction of precipitation and aerosol within the domain.

The precipitation rate in the study region is about double that observed in the TRMM 3B42 product for March 2007 (Figs. 3c,d). However, the spatial pattern shows some similarities, with a reduction of the precipitation towards the north of the domain. The increased precipitation in the model may be partly responsible for the lower AOD in the simulation compared to the MODIS AOD, through an increase in wet scavenging. It is also possible that the use of MODIS fire counts to determine the biomass burning emissions results in an underestimation of the emissions in the southern part of the domain, where cloud cover is higher.

While there are some shortfalls in the representation of the magnitude of the aerosol and precipitation rates in this simulation, the main aim of this work is to investigate the interaction between precipitation and aerosol within the domain. Given the somewhat idealised nature of this study, this simulation represents convective precipitation in an aerosol-laden environment to a sufficient extent for this study.

We investigate four different definitions of “precipitating” or “cloudy” when separating the “clear-sky” from the “all-sky” AOD in WRF-Chem. The first two rows in Fig. 4 show definitions of “precipitating” using the WRF-Chem surface precipitation rate. Whilst the “clear-sky” AOD is very similar between the different definitions of “precipitating”, the precipitating-sky AOD is much noisier when using the stricter definition ($>2\,\mathrm{mm\,hr^{-1}}$) of “precipitating” (Fig. 4d). For both definitions of “precipitating” (Fig. 4c,f), the AOD in the precipitating scenes is generally lower than that in “clear-sky” scenes. When only heavily precipitating scenes ($>2\,\mathrm{mm\,hr^{-1}}$) are counted as precipitating (Fig. 4f), the reduction in AOD for the precipitating scenes becomes even more pronounced. In regions of significant biomass burning to the north of the domain, part of the reduction in AOD comes from an impact of precipitation on biomass burning emissions. However, large reductions in AOD are also seen in regions further away from aerosol sources, as would be expected if wet scavenging is a major method of removing aerosol (and reducing AOD) in the atmosphere.

The conditions used in the bottom two rows of Fig. 4 are defined using the WRF cloud flag, summed vertically such that it is equal to the number of model layers where there is cloud. This integrated cloud flag (ICF) provides a measure of the geometrical thickness of a cloud. We again find that the “clear-sky” AOD is similar for both the lenient and more stringent cloudiness definitions (Fig. 4h,k) and that as the definition becomes more stringent, the cloudy-sky AOD becomes noisier

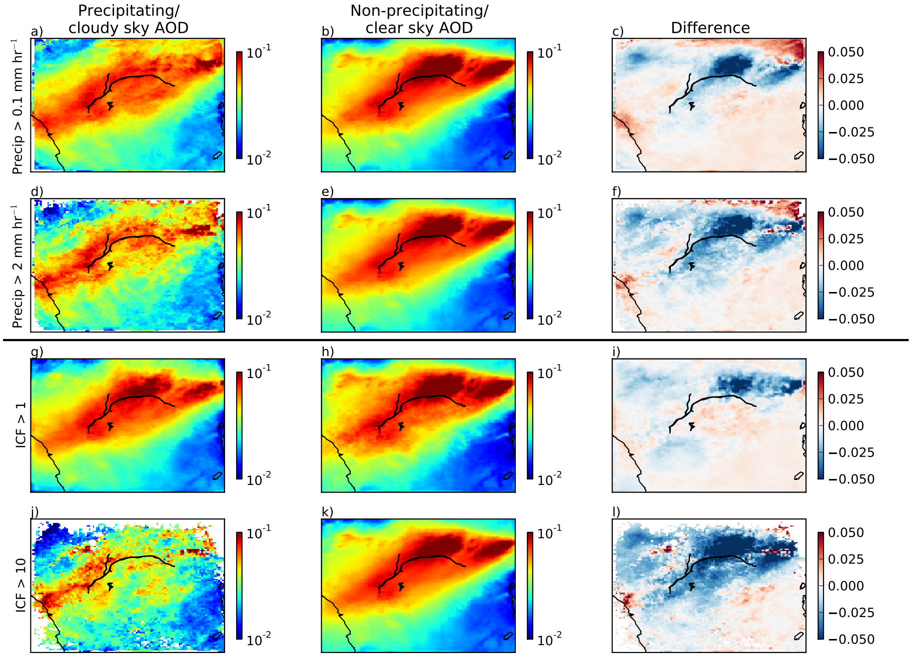

Figure 4. The influence of wet scavenging on AOD in WRF-Chem. Each of the rows uses a different criterion to define clear/cloudy sky. The top two rows use surface precipitation, with precipitation over 0.1 mm hr^{-1} and 2 mm hr^{-1}, respectively, defined as cloudy regions. The bottom two rows define cloudy as an integrated cloud flag of greater than 1 and greater than 10 respectively. From left to right, the columns show the cloudy-sky AOD, the clear sky AOD and the difference, with blue indicating a lower AOD in the cloudy sky.

(Fig. 4j). In general, there is a decrease in AOD in the cloudy scenes compared to the “clear-sky” regions, with this decrease becoming stronger if the cloudiness condition is made more stringent (Fig. 4i,l).

When using either the precipitation or the ICF criteria for separating the “clear-sky” AOD, there are several regions where there is an increase in AOD in the precipitating/cloudy sky, especially when using the less stringent condition (R >0.1 mm hr^{-1}, ICF>1). This is primarily due to an increase in relative humidity in these cloudy regions resulting in hygroscopic growth of the aerosols and increasing the AOD (Supplementary Fig. A1). The increase of AOD in cloudy and near-cloud locations is thought to be responsible for a large part of the AOD–CF relationship (Quaas et al., 2010).

3.2 Storm-centric composites

To further investigate the impact of precipitation on aerosol, we examine the properties of a composite of mid-sized convective systems and the surrounding aerosol from our WRF-Chem simulation. Fig. 5a shows a strong reduction in the column integrated AOD where the composite system is cur-

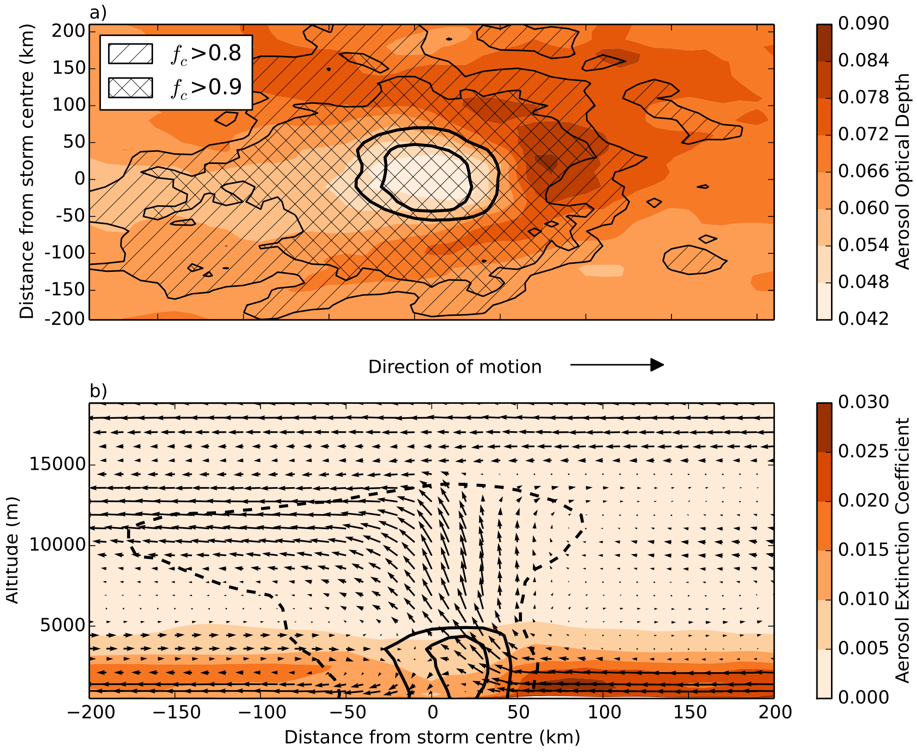

Figure 5. A composite of storms from a three week WRF-Chem simulation in March 2007 over the Congo Basin. The storm composite is moving from left to right on the above plots. *a)* A horizontal plot, with the orange filled contours showing the integrated aerosol optical depth and the hatched regions showing cloud covered regions in 80 and 90% of the storms making up the composite. The solid lines are the 2 and 5 mm hr^{-1} rainrate contours. *b)* A vertical cross section through the centre of the composite storm. The orange contours show the aerosol extinction coefficient and the arrows indicate the wind direction relative to motion of the storm centroid. The vertical wind has been enhanced by a factor of five to compensate for the different vertical and horizontal scales. The solid contours show the 0.2 and 0.8 g kg^{-1} levels of rainwater content and the dashed contour is the -20 dbZ radar reflectivity contour.

rently precipitating and along its previous trajectory (towards the left of the plot). There is also an increase in AOD towards the leading edge of the system, primarily due to aerosol humidification effects (Haywood et al., 1997; Redemann et al., 2009, Supplementary ~~Fig. 2~~Figs. A2). We also see that both the region where there is a reduction in AOD and that where there is an increase in AOD are obscured by higher cloud cover. As fractional cloud cover is not available in this simulation, f_c is the percentage of storms in the composite that have an ICF>1. The region with an f_c of greater than 90% extends slightly in the direction of travel of the system (towards the right of the figure), but trails further behind the system, hiding the main regions where wet scavenging is occurring.

In the vertical cross section (Fig. 5b), we see that the main contribution to the total AOD comes from below 5 km, with only a small amount coming from aerosol being lofted by vertical motion at the leading edge of the system. There is a clear reduction in aerosol in the centre of the system where the most significant precipitation is occurring. The bold black contours showing the location of rainwater within the cloud are displaced slightly from the storm centre, as they are instantaneous values and the storm centre is determined using precipitation values accumulated over one hour periods.

We have used the simulated -20 dbZ radar reflectivity contour to indicate the edge of the composite system. The storm composite shows a divergent anvil outflow at 10 km altitude. The -20 dbZ contour is also higher directly above the centre of the system, perhaps indicating overshooting tops.

Perhaps most importantly for possible aerosol effects on convective precipitation, the main updraughts in the storm composite contain air that is sourced from ahead of the storm (Fig. 5b). This means that the air ingested by the storm into the updraught areas (where the aerosol activation takes place) has not been affected by precipitation (as would be the case if the storm drew in air from regions it had just passed through). The structure of this composite storm is very similar to that previously observed in radar studies of convective systems (e.g. Houze et al., 1989). The composite displays a 'trailing stratiform' precipitation pattern (where the stratiform precipitation trails the convective updraught region), shown by the larger extent of the radar reflectivity and rain water content contours behind the composite storm than in front of it. This structure is more common than the ~~'leading stratiform'~~ "leading stratiform" structure, where the stratiform precipitation region leads the convective region (Parker and Johnson, 2000). We also observe a weak rear inflow of approximately 4 ms^{-1} relative to the motion of the composite (Smull and Houze, 1987). This inflow does not reach the centre of the storm, descending to the surface at the trailing edge of the heavily precipitating region (Fig. 5b). The exact structure of the composite depends on the parameters used for selecting the systems making up the composite. Slightly different values can result in a more symmetrical composite system, probably the result of combining leading and trailing stratiform systems. However, the region of main precipitation is still covered by cloud at the centre of the composite.

To demonstrate the importance of wet scavenging in reducing the AOD at the centre of the composite convective system. We also examine a composite convective system created from a simulation in which the wet scavenging of aerosols is disabled (Fig. 6). The lack wet scavenging leads to a higher overall AOD, due to the slower rate of aerosol removal. There is an increase in AOD at the centre of the composite system, rather than the decrease in AOD found in Fig. 5, which suggests that wet scavenging is indeed responsible for the reduction in AOD at the centre of the composite system. The increase in AOD at the centre of the composite constructed with wet scavenging absent (Fig. 6) is due to both the hygroscopic growth of the aerosol at the centre of the system and an increase in aerosol dry mass due to aerosol being lofted by the storm.

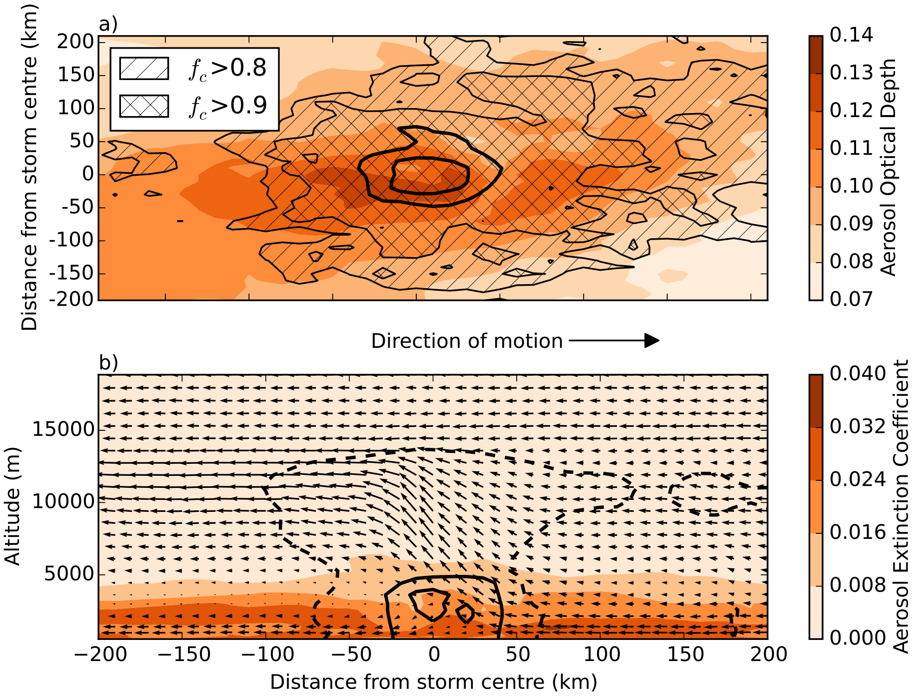

Figure 6. As Fig. 5 but constructed from a simulation with no wet scavenging of aerosol. This composite is composed of 22 individual convective systems.

As this simulation concerns mainly the wet scavenging of boundary layer aerosol, it is possible that the inclusion of free tropospheric aerosol from sources outside the simulation domain might change the relationship between AOD and precipitation in the composite. However, Grandey et al. (2014) showed that convective wet scavenging of aerosol is responsible for generating a negative AOD-precipitation correlation in a GCM throughout the tropics, even far from the main sources of aerosol. This would suggest that including free tropospheric aerosol this simulation would not impact the simulated AOD-precipitation relationships.

A simulation with a resolution of 10 km may not be able to resolve all of the important features in the convective systems that are part of this composite. However, the composite shows a qualitative similarity with a composite generated from a simulation at 4 km resolution without chemistry or aerosols (see ~~appendix~~Supplementary Information Section B). The updraughts are in the same location at the front of the storm, drawing in air from regions that have not previously experienced precipitation.

3.3 ~~Observations~~

4 Observational Consequences

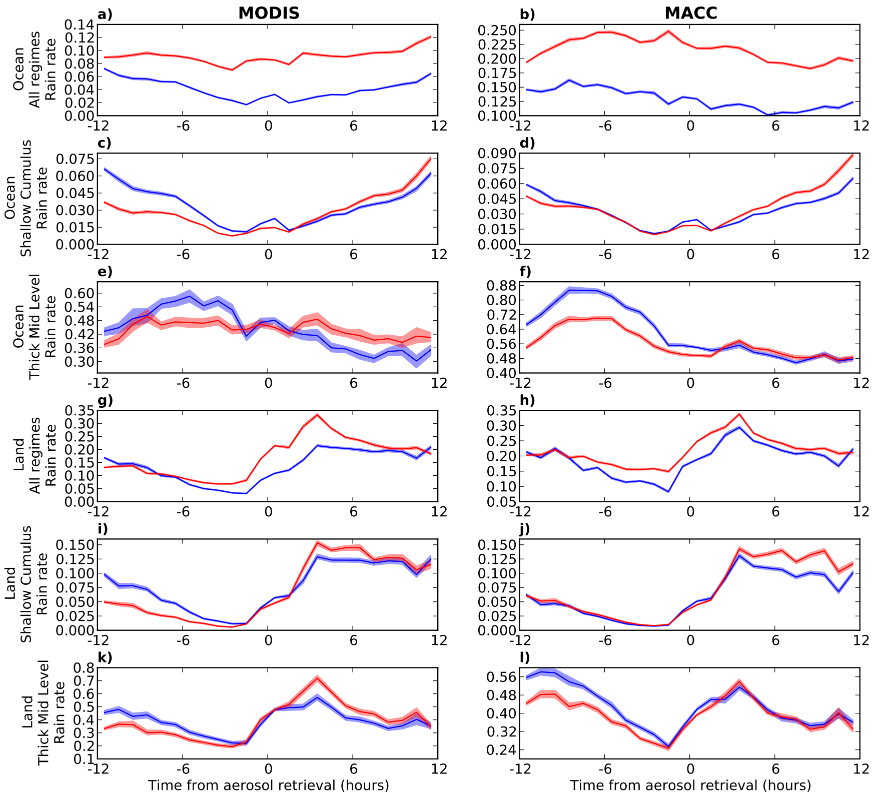

Figure 7. TRMM 3B42 precipitation development plots in the style of Gryspeerdt et al. (2014b) from 2003–2007 between 30°N and 30°S, comparing the use of MODIS AOD (left column) and MACC Total AOD at 550 nm (right column) as the aerosol product. This shows the development of the precipitation at times before and after the aerosol retrieval (1330 LST). The red line is the precipitation rate for the high AOD population and the blue for the low AOD population. Statistical errors are shown at the 95 % level. The plots are shown for ocean (a–f) and land (g–l) separately. For each product and surface type, data for all the regimes together is shown along with the shallow cumulus regime (as an example of a low CF regime) and the thick mid-level regime (as an example of a high CF regime). The cloud regimes are only specified at the time of the aerosol retrieval; transitions may occur between regimes at other times. TRMM 3B42 merged precipitation is used throughout this figure.

~~Given the~~ Large differences between clear-sky and all-sky aerosol relating to the occurrence of precipitation have been observed in the WRF-Chem simulation~~, we investigate the relationship between satellite retrieved AOD and precipitation in different cloud regimes. By using~~. However, it is unclear to what extent the inability to distinguish clear-sky and all-sky aerosol might impact observed aerosol–cloud–precipitation relationships. In this section, we use two aerosol products ~~,~~ ~~we~~ to investigate the importance of distinguishing the “clear-sky” AOD (MODIS AOD) from the “all-sky” AOD (MACC AOD) for observed aerosol–cloud–precipitation relationships. While the previous section has only shown the difference of the “clear-sky” and the “all-sky” AOD in the Congo Basin region, previous work using global models has indicated that they are important to distinguish throughout the tropics (Grandey et al., 2014) .

While some differences between the “all-sky” MACC AOD and the “clear-sky” MODIS AOD are shown in Fig. 1, the strong impact of aerosol humidification on the relationship obscures a possible influence of aerosol on precipitation. We use the precipitation development method from Gryspeerdt et al. (2014a) to reduce the influence of these meteorological covariations on the AOD-precipitation relationship while retaining the ability to detect a possible influence of aerosol on precipitation.

We find strong similarities in the precipitation development of the different cloud regimes when using MACC AOD and MODIS AOD (Fig. 7). When regimes and CF variations are not considered, both MODIS (Fig. 7a) and MACC (Fig. 7b) AOD show a strong link between precipitation and AOD over ocean, before, at and after T+0. This relationship is also seen over land, although to a lesser extent (Fig. 7g,h), with increased precipitation from the high AOD population (red line) compared to the low AOD population. This matches the effect seen in Fig. 1, where increased AOD is correlated to an increase in retrieved precipitation.

The diurnal cycle of precipitation is very similar between the plots using MODIS AOD and using MACC AOD, as the same precipitation dataset is used for both sets of plots. The absolute magnitude of the precipitation is larger when using MACC AOD, as MACC allows the sampling of overcast regions with a higher precipitation rate that MODIS cannot sample.

In the shallow cumulus regime (a low CF regime), the “all-sky” AOD is dominated by the “clear-sky” AOD. When using MODIS AOD (Fig. 7c,i), we see a higher precipitation rate for the low AOD population compared to the high AOD population at times before T+0, previously interpreted as wet scavenging (Gryspeerdt et al., 2014b). We also see an increase in the precipitation rate for the high AOD population compared to the low AOD population at times after T+0, over both land and ocean. This may indicate an aerosol invigoration of convective clouds (Gryspeerdt et al., 2014b). It is important to note that the cloud regime is only determined at T+0, with transitions between regimes occurring at other times. As such, the apparent invigoration of the shallow cumulus regime is not necessarily due to a change in precipitation from shallow cumulus clouds, but is likely due to transitions into more heavily precipitating regimes (Gryspeerdt et al., 2014c) .

When comparing the precipitation development plots using MACC AOD (Fig. 7d,j) to those using MODIS AOD, the shallow cumulus regime shows similar features. The increase in precipitation for the high AOD population compared to the low AOD population is still visible after T+0 over land. However, there is very little difference in the precipitation rate at times before T+0 between the high and low AOD populations (Fig. 7d,j). This contrasts strongly with the MODIS AOD results, where a wet scavenging signature is easily visible over both land and ocean for the shallow cumulus regime.

The thick mid-level regime is an example of a high CF regime, where MODIS AOD retrievals are less common and the “clear-sky” AOD is a much smaller proportion of the “all-sky” AOD. For both

MODIS (Fig. 7e,k) and MACC (Fig. 7f,l) we see a higher precipitation rate for the low AOD population before T+0, over both land and ocean. This indicates the wet scavenging of aerosol. The higher precipitation rates when using MACC AOD over ocean (Fig. 7f) are likely due to the increased sampling of overcast, precipitating locations that MACC allows for. ~~Whilst wet scavenging~~The higher precipitation rate from the low AOD population before T+0, consistent with wet scavenging, is observed when using both MACC and MODIS AOD, an increase in precipitation with increasing AOD after T+0 is only observed when using MODIS AOD. This increase in precipitation with increasing AOD observed when using MODIS is consistent with an aerosol invigoration of convective clouds. If this increase in precipitation is due to an aerosol invigoration effect, then this suggests that the use of MACC AOD obscures the aerosol influence on precipitation in these high CF, highly precipitating regimes.

5 Discussion

5.1 "All-sky" vs. "Clear-sky" AOD

The analysis of AOD for different precipitation rates in Fig. 4 generally shows a reduced AOD in cloudy/precipitating areas. The high CF in these locations would restrict the satellite retrieval of AOD. This can also be seen on the scale of an individual storm in the storm composite (Fig. 5), where the AOD is reduced in the precipitating region towards the centre of the storm. This reduction in AOD would be hard to retrieve with satellites due to the high cloud cover, while the AOD in lower f_c regions towards the edge of the storm has not been so strongly influenced by precipitation, remaining similar to the "clear-sky" AOD at the edge of the composited region.

This provides further evidence that the difference in the AOD–precipitation correlation between MODIS and the HadGEM-UKCA GCM shown in Fig. 1 is due to differences in sampling between the model and observations, as suggested in Grandey et al. (2014). In regions with a high precipitation rate (such as the tropics), wet scavenging dominates over aerosol hygroscopic growth when determining the relationship between the "all-sky" AOD and precipitation, explaining the negative correlation in Fig. 1c. Wet scavenging impacts the "clear-sky" aerosol via the post-storm "wake" that can be seen to the left of Fig. 5a. Although this "wake" would be visible to satellites, it is a very small proportion of the "clear-sky" aerosol when compared to the impact of wet scavenging on aerosol in the cloud covered regions of the composite. This means that precipitation exerts a much stronger control over the aerosol in cloudy regions compared to the "clear-sky" aerosol. As the "clear-sky" aerosol is not so heavily scavenged, wet scavenging does not play such a strong role in determining the correlation between "clear-sky" AOD and precipitation. In these situations, the influence of aerosol hygroscopic growth is more important (Boucher and Quaas, 2012; Grandey et al., 2014), generating much of the positive correlation between AOD and precipitation seen in Fig. 1c.

~~Although~~ While the correlations in Fig. 1 show the link between AOD and precipitation, they cannot provide evidence of aerosol invigoration of convective clouds due to the confounding effects of meteorological covariations (Boucher and Quaas, 2012; Grandey et al., 2013; Gryspeerdt et al., 2014c). The precipitation development plots in this work are designed to account for the influence of meteorological covariations when investigating aerosol–cloud interactions. The observation of both wet scavenging and possible aerosol invigoration when using MODIS suggests that aerosol invigoration could be responsible for an increase in precipitation from convective clouds under certain conditions (Gryspeerdt et al., 2014b). The increase in precipitation after T+0 and the wet scavenging effect are only observed in certain regimes when using MACC aerosol data. Given the different sampling of MACC and MODIS AOD, this suggests that the strong effect of wet scavenging on AOD in cloudy skies might be obscuring an aerosol influence on precipitation in some regimes.

In heavily precipitating regions, the "all-sky" AOD observed by a model (with a similar sampling to MACC) is significantly lower than the "clear sky" AOD, as seen in the WRF-Chem results (Fig. 4). A lower AOD is not itself enough to prevent the observation of an aerosol invigoration effect in the precipitation development plots, as they depend on the AOD having some predictive power of the future evolution of the storm, rather than the absolute magnitude of the AOD. However, in regions of high CF and strong precipitation, the "all-sky" AOD–precipitation correlation is controlled almost entirely by wet scavenging. In these regions, the control of the aerosol by precipitation means that the "all-sky" AOD then loses its predictive power over the future evolution of the storm, only reflecting the previous history of the airmass. This suggests that the "clear-sky" AOD, preferentially sampled by satellites, is more representative of the aerosol environment in the early stages of the formation of storms, as it is not so strongly affected by precipitation from those storms. While the influence of wet scavenging can affect satellite studies (Fig. 7), low resolution models are much more significantly affected as they are less able to separate the "clear-sky" aerosol from the "all-sky" aerosol.

The mixing of clear and cloudy sky aerosol populations explains why the wet scavenging of aerosols is visible in the precipitation development plots from the thick mid-level regime (high CF) when using MACC AOD but the increase in precipitation with increasing AOD after T+0 is not visible. In this high CF regime, the MACC AOD is strongly influenced by the model precipitation, as there are few MODIS AOD retrievals to assimilate. The MACC AOD is then much more strongly connected to the history of the precipitation rate than it is to the aerosol that is drawn into the cloud, preventing the ~~potential invigoration-like effect for~~ apparent invigoration of the thick mid-level regime over land (Fig. 7k) from being observed using MACC AOD (Fig. 7l).

In low CF regimes (such as the shallow cumulus), this is not an issue as the majority of the aerosol is "clear-sky" aerosol and so the "all-sky" AOD closely tracks the "clear-sky" AOD. This allows the ~~invigoration-like effect to be observed in~~ observation of an apparent invigoration of precipitation from the shallow cumulus regime when using both MACC (Fig. 7i) and MODIS AOD (Fig. 7j).

Wet scavenging obscuring the influence of aerosols on convective clouds also explains some of the results in previous work. Gryspeerdt et al. (2014c) investigated the links between aerosols and transitions between cloud regimes, finding that whilst increased transitions to deep convective-type clouds were observed with increases in MODIS aerosol index, this increase was not observed when using MACC AOD. The results in this work suggest that this is most likely due to the influence of wet scavenging and the sampling difference between MACC and MODIS AOD.

The influence of wet scavenging does not need to have a large effect on the total mean AOD to have a strong effect on the link between AOD and precipitation development within the strongly precipitating/high CF regimes. These high CF/strongly precipitating regimes ~~have a small combined relative frequency of occurrence (RFO) – deep convective and thick mid level have a combined RFO~~ occur rarely, with only 13% of the cloud regime occurring in the tropics ~~of approximately 13~~falling into the deep convective or thick mid-level regimes (Gryspeerdt and Stier, 2012). Even though the sampling varies between MACC and MODIS, the mean MACC AOD is very close to that determined using MODIS and other satellite instruments (Morcrette et al., 2011). This demonstrates that although the overall magnitude of the wet scavenging in MACC may be similar to that seen in observations, small sampling differences can impact correlations between aerosol and cloud properties.

5.2 Comparison to GCM processes

We have shown that the clear-sky sampling bias in satellite AOD data impacts the correlations between AOD and precipitation. Both the composite storm in Fig. 5 and previous radar-based studies of convective systems suggest that air is usually drawn into convective updraughts from non-precipitating regions. Coupled with the reduction in aerosol in cloudy skies due to wet scavenging, this suggests that the “clear-sky” AOD could be more closely related to the aerosol drawn into convective systems than the “all-sky” AOD (which is more strongly influenced by precipitation). GCMs assume that aerosol is mixed across a gridbox on a timescale of a model timestep (10–30 minutes), limiting their ability to distinguish the “clear-sky” aerosol from the “all sky” aerosol. This may make it difficult for GCMs to detect aerosol influences on precipitation using the precipitation development method.

A preference for using the “clear-sky” aerosol when investigating aerosol–cloud interactions is not likely to be the case for all precipitating clouds. As shown in previous work, models can reproduce the observed AOD–CF correlation more successfully in mid-latitude regions than they can near the equator (Grandey et al., 2014). This suggests that this sampling difference is not as important an issue where frontal precipitation is involved, perhaps due to the larger precipitation spatial scales involved. The intensity of the precipitation involved is also important, as the precipitation must be intense enough to remove the link between the “all-sky” and the “clear-sky” aerosol. Unlike the convective regimes, the development of the stratocumulus regime shows a similar correlation to

MACC AOD as it does to MODIS AOD (Gryspeerdt et al., 2014c), suggesting that the “all-sky” and the “clear-sky” aerosol are correlated for cloud regimes with low precipitation rates.

The storm composite in Fig. 5 is composed of storms approaching the size of a GCM gridbox that are independent from other storm systems. The filtering techniques used to select the storms for the storm composite may have introduced a bias into the composite so that is it not representative of convective storms in general. As noted earlier, the storm composite displays the more common trailing stratiform structure. However, leading stratiform structure storms may ingest air into convective updraughts from locations with recent precipitation or through the stratiform precipitation regions, reducing the link between the “clear-sky” AOD and the ingested aerosol. The requirement that the storms be independent of neighbouring precipitating systems may also bias the structure of the composite. In large groups of interacting individual convective systems, new systems may be triggered by the outflow from convective downdraughts (Thorpe et al., 1982; Wakimoto, 1982). This makes new convective systems more likely to ingest air that is part of the outflow from other systems. As the aerosol in the outflow has come from inside a cloud, the “all-sky” sampling may be more representative of the aerosol ingested by convective systems in these cases.

While there are some cases where the “clear-sky” AOD may not have an advantage over the “all-sky” AOD, the precipitation development results (Fig. 7) suggest that the “clear-sky” AOD has an advantage in detecting influences of aerosol on precipitation. If the “clear-sky” AOD can not be separated from the “all-sky” AOD, links between aerosol and precipitation development from convective systems can be obscured. Due to the difficulty in determining the “clear-sky” AOD in GCMs, this is may impact the detectability of aerosol influences on precipitation in GCMs using the precipitation development method.

6 Conclusions

In this work we have used the WRF-Chem model and satellite observations to examine how aerosol is affected in precipitating convective systems and how this impacts correlations between AOD and precipitation properties.

Using the WRF-Chem model, we have found that there is generally a reduction in AOD in precipitating regions (Fig. 4), with this reduction becoming more severe when more stringent conditions are used to define precipitating regions. We also find a decrease in AOD in cloud covered locations, due to the strong link between CF and precipitation in the Congo region. In scenes with a low (but non-zero) precipitation rate, there is an increase in AOD with increasing precipitation, primarily caused by the hygroscopic growth of aerosol in humid environments (Supplementary Fig. A1).

Creating a composite of mid-sized convective systems in our study region (Fig. 5), we show how aerosol interacts with precipitating systems on the storm scale. AOD is strongly reduced in the core of these systems, where the precipitation is strongest, although the reduction in AOD persists in locations where the system has previously been precipitating. The regions where the most significant reduction in AOD occurs are in locations that are usually covered by cloud, preventing their observation by satellites. This results in different sampling of AOD between satellites and models and help to explain the difference in the AOD–precipitation correlation between them (Fig. 1).

The composite also shows how air is drawn into convective systems relative to their direction of travel, such that the aerosol ingested by a system has not previously interacted with precipitation from the same storm system. This suggests that the aerosol drawn into such storm systems is more closely related to the "clear-sky" AOD observed by satellites than the "all-sky" AOD that is sampled by atmospheric models. The importance of the "clear-sky" aerosol relative to the "all-sky" aerosol varies by cloud regime, but for sufficiently spaced individual convective systems as analysed here, this would suggest that the satellite "clear-sky" sampling of AOD may be more suited to investigating aerosol–cloud interactions.

This is supported by observations using MODIS AOD and the TRMM merged precipitation product, along with MACC reanalysis AOD to provide a model-like "all-sky" AOD field. When looking at two specific regimes, the shallow cumulus (with a low CF) and the thick mid-level (with a high CF), we see an invigoration-like effect in both regimes when using MODIS AOD. When using MACC AOD, we only see the invigoration-like effect in the low CF regime, suggesting that the use of "all-sky" AOD in highly precipitating regimes masks the observation of a possible invigoration effect.

This work shows that the different sampling of aerosols by satellites and reanalysis models/GCMs can have a large effect on the correlations between aerosol and precipitation properties. When using the precipitation development method in highly-precipitating convective regimes, an increase in precipitation with increasing AOD seen when using MODIS AOD cannot be detected when using MACC reanalysis AOD. This suggests that even if a GCM has a perfect representation of aerosol effects on convective clouds, it may not be able to reproduce the correlations between AOD and precipitation in highly precipitating locations, due to the differences in AOD sampling between GCMs and satellites.

7 ~~Higher resolution simulations~~

~~The compositing methodology in this work has also been applied to a convection-permitting simulation of the same region and period at a resolution of 4km, running without a cumulus scheme. This run does not include aerosols or chemistry, but it does show the structure of the storm composite (Figs. ??, ??). Similar to the 10km simulation, updraughts are located at the front of the system, drawing air into the system from nearby, non-precipitating regions (Fig. ??). While the structure of the composite shows some differences, missing some of the inflow at the rear of the system (Fig. 5), the similarity of the composite system to the 10km simulation suggests that the conclusions drawn in this work~~

~~would be supported if the simulations were re-run at a higher resolution. This composite contains 22 separate systems.~~

~~Radar reflectivity structure of the storm composite from a one month run at 4km resolution. The direction of motion of the system is towards the right of the image.~~

~~a) A horizontal plot of the storm composite from the 4km resolution WRF simulation. The hatched areas indicate the percentage of storms going into the composite with cloud in that region. Note the different limits for the contours compared to Fig.5 due to the resolution dependence of cloud fraction. The solid lines are the 2 and 5mmhr^{-1} rainrate contours. b) A vertical cross section through the centre of the system. The arrows indicate the wind direction relative to the storm centroid, enhanced by a factor of five to compensate for the different vertical and horizontal scales. The solid contours show the 0.2 and 0.8gkg^{-1} levels of rainwater content and the dashed contour is the -20dbZ radar reflectivity contour.~~

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