Thanks referee #1 for his time and interest in this work.

Minor comments:

1/ Page 3 "experimental evidence", maybe better "observation-based evidence" and "drives" is awkward maybe "can illustrate".

We agree that these expressions are more appropriate. They will be used in the revised version

2/ Page 4 I am not convinced that smoke particles have solubility that is (relatively to what)? High.

Here we mean that smoke has a significantly higher solubility than dust, the other main aerosol type present in this region. As a consequence, smoke may provide the largest fraction of CCN.

Aerosol released from forest and cropland fires (generally referred to biomass burning aerosol) mostly contains organic carbon (OC) with various amounts of black carbon (BC, emitted primarily in efficient flaming fires) depending on the particular fuel, oxygen availability and combustion phase (Andreae and Merlet, 2001). The inorganic component biomass burning aerosol is made of some insoluble dust and ash material, and soluble salts, while half of the organic matter (major component) is considered to be water soluble (Reid et al. 2005; Hoffer et al., 2005; Decesari et al. 2006).

The amount of oxygen at the surface of soot particles depends on combustion conditions, with more efficient combustion regimes resulting in higher abundance of oxygen (Chughtai et al., 2002; Su et al., 2004) which in turn increases the chemical reactivity of the particles and their wettability in the atmosphere (Andreae and Gelencsér, 2006.). While soot is usually thought to be insoluble in water and organic solvents (this is definitely true for pure graphite) more atmospherically relevant soot types behave differently (Medalia and Rivin, 1982).

In conclusion, as a result of its chemical internal mixing, aerosol from biomass burning consists in a large fraction of soluble (organic and inorganic) material and is already able to act as CCN immediately over the fire (Andreae and Rosenfeld, 2008)

<u>Additional references not present in the paper Bibliography:</u>

Andreae, M. O. and Gelencsér, A.: Black carbon or brown carbon? The nature of light-absorbing carbonaceous aerosols, Atmos. Chem. Phys., 6, 3131-3148, doi:10.5194/acp-6-3131-2006, 2006.

Andreae, M. O. and Rosenfeld, D.: Aerosol-cloud-precipitation interactions. Part 1: The nature and sources of cloud-active aerosols, Earth Sci. Rev., 89(1–2), 13–41,

doi:10.1016/j.earscirev.2008.03.001, 2008.

Chughtai, A. R., Kim, J. M., and Smith, D. M.: The effect of air/fuel ratio on properties and reactivity of combustion soots, J. Atmos. Chem., 43, 21–43, 2002.

Decesari, S., Fuzzi, S., Facchini, M. C., et al.: Characterization of the organic composition of aerosols from Rondonia, Brazil, during the LBA-SMOCC 2002 experiment and its representation through model compounds, Atmos. Chem. Phys., 6, 375–402, 2006.

Hoffer, A., Gelencser, A., Guyon, P., Kiss, G., Schmid, O., Frank, G., Artaxo, P., and Andreae, M. O.: Optical properties of humic-like substances (HULIS) in biomass-burning aerosols, Atmos. Chem. Phys. Discuss., 5, 7341–7360, 2005.

Medalia, A. I. and Rivin, D.: Particulate carbon and other components of soot and carbon black, Carbon, 20, 481–492, 1982.

Reid, J. S., Koppmann, R., Eck, T. F., and Eleuterio, D. P: A review of biomass burning emissions part II: intensive physical properties of biomass burning particles, Atmos. Chem. Phys., 5, 799–825, 2005.

Su, D. S., Muller, J. O., Jentoft, R. E., Rothe, D., Jacob, E., and Schlogl, R.: Fullerene-like soot from EuroIV diesel engine: con-sequences for catalytic automotive pollution control, Topics in Catalysis, 30–31, 241–245, 2004.

3/ Page 8 the derivation of the relationships in equation 9 is not quite clear (sub-steps would help).

The idea is to firstly substitute eq 8 in 6: H^2 results proportional to CDR ^3, N and H. We isolate H, expressed as a function of CDR and N, and substitute it in equation 7 that would now relate COT, CDR and N.

We absolutely agree with referee#1 that the derivation of eq 9 is not clear, mostly because in equation 9 there is a mistake in the exponent of N, which is equal to -0.4 and not to 0.2 ! This correction may probably be enough to clarify the derivation of equation 9.

4/ Page 8 “use used”

Ok.

5/ Page 9 what is done to detect AOD above the stratocumulus, if there are not cloud-free scenes (as we would expect for stratocumulus decks), which are required for MODIS AOD retrievals?

We consider only cases of broken cloud conditions, when MODIS is able to retrieve aerosol properties between clouds or near clouds edges. This could be seen as a strong limitation of our

analysis. In practice, the analysis of MODIS products show that there are aerosol retrievals in the vicinity of the cloud parameter retrievals is most cases.

6/ Page 9 significant uncertainty to the microphysical cloud properties (reff, COT, LWC..) can be expected not only by inhomogeneity but also by the presence of absorbing wildfires aerosol above those clouds. I hope the discussions address this issue. Cloud top altitude overestimates (in case inversions) should be discussed also in the context of CALIOP data.

We do not expect a strong underestimation of cloud properties (CDR and COT) in case of absorbing aerosol above cloud top, using the 0.86 μm and 2.12 μm MODIS channels. For what concerns CDR, in Costantino and Breon (2010), we make use of POLDER instrument (on board of PARASOL satellite) to estimate of cloud droplet effective radius and we find a very similar trend of CDR with increasing AI to that shown in this paper, in case of mixed and unmixed cloud-aerosol layers. Using the 0.86/2.1 μm combination of wavelengths, Haywood et al. (2004) find a maximum error in retrieved COT up to 10-20% in case of smoke.

For what concerns cloud top pressure errors, in case of strong inversion we report of some studies showing a maximal error in MODIS retrievals up to 200 hPa, compared to Lidar determination of mono-layer cloud top altitudes (Menzel et al., 2008; Garay et al., 2008; Harshvardan et al., 2009). The error in cloud top height can be as much as 1000-3000 km. Over the selected area, the bias in MODIS retrieved cloud top pressure error can be particularly high. In our analysis, however, we account only for relatively low clouds. Looking for MODIS-CALIPSO coincidences, cases of MODIS retrievals with top pressure lower than 600 hPa are not considered. We then expect that large errors in CTP estimates, placing cloud top altitude in elevated layers of the atmosphere, are mostly avoided.

7/ Page 10 "than in?" (upper troposphere?) Page 10 I wonder on cloud-top agreement by ground-based lidar (must be thin clouds).

We say "with a higher resolution in the lower atmosphere than in and the upper layers" to mean than (a part from negative altitude values) CALIPSO level 1 data have different spatial resolutions for different altitude ranges, decreasesing monotonically with increasing altitude as shown in Table 1 (Winkler et al., 2004).

Altitude Range [km]	Horizontal Resolution [Km]	532 nm Vertical Resolution [m]	1064 nm Vertical Resolution [m]
30.1 to 40.0	5.0	300	---

20.2 to 30.1	1.67	180	180
8.2 to 20.2	1.00	60	60
-0.5 to 8.2	0.33	30	60
-2.0 to -0.5	0.33	300	300

Table 1: CALIPSO spatial resolution.

Kim et al. (2007), considering both geometrically thin and thick clouds (up to several km), find a good agreement between ground base and CALIPSO estimates of top and bottom cloud layer altitudes, within 0.1 km (this is true also for aerosol top and bottom layer altitudes). They show a strong discrepancy in aerosol layer altitudes between ground base and CALIOP lidar, when aerosol is located below thick clouds. In this paper, however, cases of aerosol below clouds are not considered.

In a more recent work of Kim et al. (2011), cloud top altitudes retrieved from the Cloud Profiling Radar (CPR, on board of CloudSat satellite) and CALIOP for thick tropospheric clouds show good agreement with each other. Discrepancies between the cloud base altitude obtained by the CPR and that acquired by the CALIOP for thick opaque clouds arise from the strong CALIOP signal attenuation (Kim et al. 2011).

Again, however, we recall that, in this paper, CALIPSO is used only to provide information on the cloud top altitude and the aerosol (between or above clouds) base altitude.

Additional reference:

Kim, Sang-Woo , Chung, Eui-Seok , Yoon, Soon-Chang , Sohn, Byung-Ju and Sugimoto, Nobuo (2011) 'Intercomparisons of cloud-top and cloud-base heights from ground-based Lidar, CloudSat and CALIPSO measurements', International Journal of Remote Sensing, 32: 4, 1179 — 1197.

8/ Page 11 clear-sky cases are not considered (as there are no cloud data). Alternately are completely cloudy cases also thrown out, because there are not aerosol data, or are then aerosol data 'borrowed' from neighboring grid-points? Filtering data may introduce biases and limit the value of results.

Completely cloudy cases are only considered if there is some aerosol next to the cloud edge, within a 10 km radius from CALIPSO observation. Aerosol layers are assumed to be rather homogeneous spatially (correlation scale much larger than 10 km) as they are observed rather far from the sources.

9/ Page 12 there are a lot of cases that are excluded for focus. Maybe a simple graph can illustrate the selection. And with so many exclusions I wonder if the remaining cases are sufficient for good statistics.

We should probably put a table showing data selection rules relative to each relationship reported in the paper.

The number concentration of coincident MODIS-CALIPSO retrievals for all aerosol regimes, in the region within [2 S, 15 S; 14W, 18 E], is shown in figure 12. Scatter-plot (not shown in the paper, but shown in the Figure 1 - below - for CDR) of the different relationship between cloud properties and AI, indicates that there is a sufficient number of observation points for the statistical analysis. Measurements of CDR, COT, etc.. are quite homogeneously distributed along the different aerosol regimes so that, when averaging cloud properties over constant bin of AI, statistical uncertainties are relatively small.

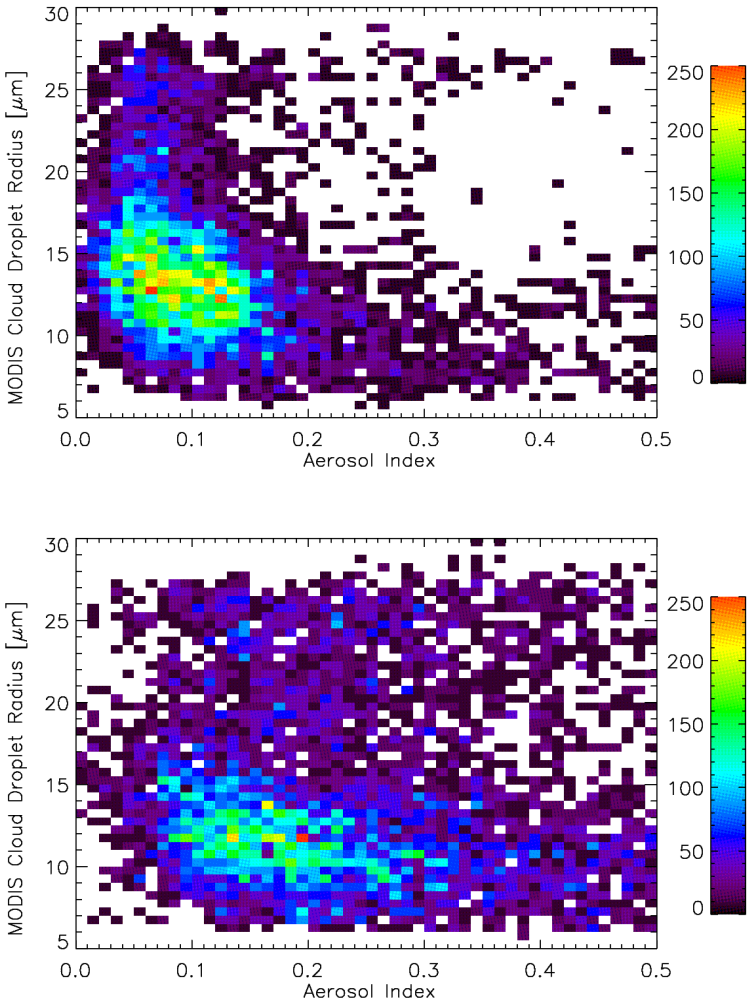

Figure 1: Histogram of coincident MODIS-CALIPSO retrievals of CDR and AI, in case of mixed and interacting cloud and aerosol layers (top image) and well separated layer (bottom image), within [2S, 15S] of latitude. Unmixed case cloud-aerosol minimum distance threshold is 750 m. Color scale represents arbitrary units, proportional to the number of points in a box of ΔAI = 0.01 and ΔCDR = 0.5 μm.

10/ Page 13 I am not quite sure why so much detail is needed about the larger region even north of the equator when only the area off Nambia and Angloa is of interest (this section cloud be shortened) or do we address in the analysis the Gulf of Guinea as there are not significant cloud amounts below most aerosol.

The South-East Atlantic, from the Gulf of Guinea to the open ocean away from the Namibian coast is a region of particular interest in the study of aerosol, aerosol-cloud interaction and their radiative impact on atmosphere and climate. In addition to aerosol and cloud statistics, we were also interest in providing an overall and comprehensive description of this region and its climatology.

11/ Page 15 I am not so happy with the seasonal choices. The biomass burning in southern Africa (e.g. Zambia) that is largely responsible for the elevated larger AOD values off Namibia is between August and October. Thus, the selected seasons are unfortunate.

African Southern Hemisphere (SH) burning starts in April (Edwards et al., 2006) and last up to October, as shown in Figure 2. By October, fire occurrence has decreased with respect to previous months. The definition of a "biomass burning season" is then a bit arbitrary. In this paper we define this season (then a three months time period) in agreement with some previous works (Ichoku et al, 2000; Myhre et al. 2003; Swap et al., 2003; Eck et al. 2003) according to which the peak of the African SH dry season biomass burning activity is from July to September;

Additional references:

Cattani, E., M. J. Costa, F. Torricella, V. Levizzani, and A. M. Silva, 2006: Influence of the aerosol particles from biomass burning on cloud microphysical properties and radiative forcing. Atmos. Res., 82, 310-327.

Eck, T. F., et al., Variability of biomass burning aerosol optical characteristics in southern Africa during the SAFARI 2000 dry season campaign and a comparison of single scattering albedo estimates from radiometric measurements, J. Geophys. Res., 108(D13), 8477, doi:10.1029/2002JD002321, 2003.

Myhre, G., T.K. Berntsen, J.M. Haywood, J.K. Sundet, B.N. Holben, M. Johnsrud, and F. Stordal, 2003, Modelling the solar radiative impact of aerosols from biomass burning during the Southern African Regional Science Initiative (SAFARI-2000) experiment, J. Geophys. Res.,108, 8501, doi:10.1029/2002JD002313.

Swap, R. J., H. J. Annegarn, J. T. Suttles, M. D. King, S. Platnick, J. L. Privette, and R. J. Scholes (2003), Africa burning: A thematic analysis of the Southern African Regional Science Initiative (SAFARI 2000), J. Geophys. Res., 108(D13), 8465, doi:10.1029/2003JD003747.

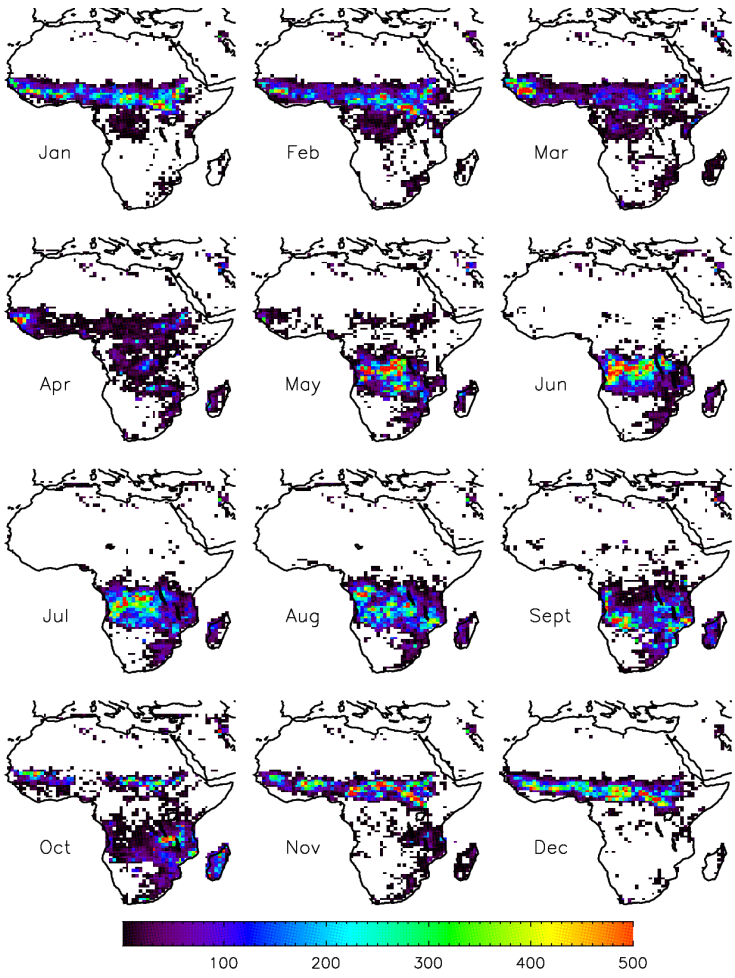

Figure 2: fire occurrence for 2005, according to MODIS Active Fire Product. Color-scale represents the number of active fires detected each month, at a nominal resolution (at nadir) of 1 km, within a 1°×1° gird box.

12/ Page 15 'figure 2' should be 'figure 3

We do not find 'figure 2' in page 15. Probably, this error has been yet corrected in the last ACPD uploaded version.

13/ Page 16 I wonder about the likelihood of MODIS aerosol retrievals in the presence of stratocumulus decks. How much is this a contributing factor for the low AI values off Namibia for Oct-Dec? Similar are the Jul to Sep values largely defined by Jul values, as much fewer samples are expected for Aug and Sep?

In our analysis, we did not consider cases with aerosols below the cloud deck. This is because this case is believed to be always interacting with the cloud (so no possibility to differentiate the two cases as in the main body of the paper). In case of aerosol above cloud, we do not expect that cloud contamination causes large aerosol in AOD (Kaufman et al., 2005; Loeb and Shuster, 2008).

In a side study we tried to quantify the so-called blueing effect, due to cloud side scattering adjacent to aerosol retrievals. Marshak et al. (2008) find out that enhancement in column radiance is more pronounced at shorter wavelength (scattered light by clouds in cloud free pixel is further scattered upward by molecules, Wen et al., 2008). According to Varnai and Marshak (2009) this effect may

lead to significant over estimation of AOD retrievals in cloud-free pixels, as far as 15 km away from cloud. At the same time, as it is supposed to be stronger at shorter wavelengths, it can affect the spectral dependence of AOD and lead to increased estimates of Angstrom exponent, near cloud edges. This would result in an apparent increase of aerosol fine mode fraction in the vicinity of clouds

As, MODIS products return the pixel position of each retrieved aerosol (10 km grid box) and cloud (1 km gird box) parameter. It is then possible, to a certain extent, to analyze the AOD difference between MODIS and CALIPSO (Δ_{AOD}) as a function of cloud-aerosol pixel distance (CAD), as shown in Figure 3. CALIPSO retrievals are not expected to be affected by cloud side-scattering. A decrease of Δ_{AOD} with increasing CAD may reasonably reflect an average decrease in MODIS AOD estimates, due to the diminution of adjacent effect as AOD is retrieved farther from cloud.

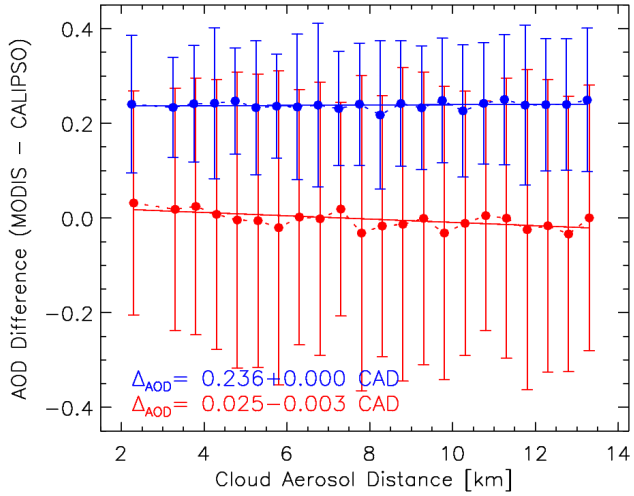

Figure 3: AOD retrieval differences, between MODIS and CALIPSO, averaged over constant bin of cloud-aerosol distance, by step of 0.5 km, in case of mixed cloud-aerosol layers (red) and aerosol above cloud top (blue). Linear regressions are performed for each case. Error bars represent the Δ_{AOD} standard deviation within the bin.

In case of mixed layers, atmospheric conditions are very similar to those considered by Wen et al. (2008) in their experiment (aerosol overlying a dark surface and trapped in the PBL; clear troposphere above cloud top) and Δ_{AOD} shows a small but negative dependence on CAD. The difference between MODIS and CALIPSO estimates is reduced by about 0.034, when average cloud-aerosol pixel distance increases from 2 to 13.5 km. This is consistent with the expected decreased in MODIS retrieved column radiance contamination by photons scattered by adjacent cloud and after by molecules above cloud top, over clear pixels located near the cloud edges.

When aerosol and cloud are well separated, with aerosol located in the low free troposphere above cloud top, Δ_{AOD} (blue) does not show any sensible dependence on cloud-aerosol pixel distance (the difference between MODIS and CALIPSO AOD is much larger than in case of mixed layers, but constant with increasing CAD). We do not dispose of an in-depth and detailed analysis of 3D

radiation processes in case of aerosol above cloud top, as such atmospheric condition is out of the assumption made by Wen et al. (2008). We hypothesize, however, that upward scattered radiation by molecules gets attenuated by overlying aerosol and does not affect MODIS retrievals.

In conclusion, results suggest that MODIS estimates of aerosol optical depth are biased high, only when the aerosol layer is very close to clouds and at the same altitude (mixed case). The resulting bias in AOD seems rather small. In addition, the typical distance between coincident aerosol-cloud pixels ranges approximately between 10 and 20 km (mean CAD is equal to 14.3±6.5 km), which is mostly larger the radius of significant influence. For these reasons, we argue that Rayleigh enhancement of MODIS retrieved radiance in cloud-free pixel is not significant enough to produce any spurious correlation between aerosol concentration and cloud fraction.

Note, however, that AOD differences between MODIS and CALIPSO single estimates are subjected to an extremely high variability that may depend on different factors. We do not think that resulting statistics, obtained by averaged all Δ_{AOD} values within a same CAD interval, can be interpreted as a conclusive quantification of cloud adjacent effect.

14/ Page 17 Is Figure 5 for data of the entire year or just one (biomass) season?
It refers to the whole dataset of MODIS-CALIPSO coincidences. We use data acquired from June 2006 to December 2010.

15/ Page 17 '-14E' is strange ... suggest to use '14W' (similarly later)
There is no -14E in the last version that was uploaded on the ACPD website.

16/ Page 18 I assume that the LWC investigation for the same (2S-15S, 14W-18E) region the text only mentioned latitude boundaries.
Yes, the latitude is from 14W to 18E. This will be added to the final version.

17/ Page 18 can we speculated on the LWC reductions (e.g. loss of water by drizzle or dry-air entrainment)?
In the paper we do argue an important dry air entrainment, as proposed by Ackerman et al. (2004), which is consistent with the local meteorology and the South Atlantic aerosol climatology.

18/ Page 19 The 4N-30S (and I assume 14W-18E) is much larger. I am in doubt that this larger region is really relevant due to the low frequency of cases of aerosol within low level clouds outside the core region. In addition, the statement confuses. There is little interest, if there is a higher value at AI=0 (this would be expected since now more tropical regions are included) but if there is a change in slope.
Small regions are desirable to avoid spurious correlation between local meteorology and AI values. On the other hand, small regions leads to few observations points that are not sufficient to derive statistical relationship. When we dispose of a particular methodology that allows to minimize the effect of spatial heterogeneity of local meteorology (eg. CTP sorting or not considering cases with low AI), but as the same time decreases the number of measurements, we increase the retrieval domain to dispose of a sufficient number of points to decrease the large statistical uncertainties.

19/ Page 20 delete 'A' before 'statistical'
Ok

20/ Page 21 ‘computed’ ! ‘compute’, what is the ‘whole SE Atlantic’ be more specific.
Ok (whole SE Atlantic region: [4 N, 30 S; 14W, 18 E]). This will be corrected

21/ Page 22 captions in Figure 8 and 9 confuse with northern latitudes ! must be wrong.
This was corrected in the last uploaded version.

22/ Page 24 By using the larger area in order to increase stat significant, the focus on ‘aerosol above stratus’ cases is weakened.
We agree with referee#1 that in tropical regions and especially in the Intertropical Convergence Zone (ITCZ), the occurrence of high clouds or strongly vertically developed and convective clouds is larger. However, we try to avoid such cases not considering MODIS retrievals of clouds with cloud top pressure smaller than 600 hPa (as well as COT larger than 35 and LWP larger than 300 gm^{-2}).

23/ Page 25 ‘for WHAT is very large’
Not found, probably corrected in last version.

24/ Page 27 the separation into COT regimes is smart to address potentially delayed precipitation. On the other hand the COT class frequency for mixed and un-mixed clouds would be nice addition. I also would like to know, who this graph would look like for the smaller focus region (2S-15S) in the Aug-Oct frame. (In that context I am still confused as to the time-period of the data considered in the most plots. I know that they are multi-year but are there considerations of seasons (or even months)?

In all multi-year relationships between cloud parameters and aerosol index, months and seasons are mixed together with no difference between different time period. [Answer continues in the comment to the next question]

25/ Page 28 Figure 12 is very important. Maybe it is possible to offer this plots by season and then sub-sample regions (and seasons?) with relatively high frequency of low altitude stratus.

We do agree with referee#1 that a study of the different time period and hence the seasonal variability of aerosol effect would be very important. However, our attempts toward this suggestion show that the dataset becomes too small to derive meaningful relationship.
Similarly, the need of a very large dataset to derive multi-regression analysis of CLF-AI relationship (sorted by CTP) did not allow to use the smaller South-Atlantic area within [2 S, 15 S; 14 W, 18 E], instead of larger one within [4 N, 30 S; 14 W, 18 E], as observed in a preliminary study.
In addition, it may be extremely interesting to sub-sample those relationships by cloud type. This would probably allow to address several issues on aerosol signature on different cloud types.
Such analysis will become meaningful when the CALIPSO/MODIS dataset will get larger. Further work is needed and we do believe that the combination of co-located vertical and horizontal observations of the atmosphere would help to clarify several aspects of aerosol-cloud interaction that are still largely uncertain.

26/ Page 32 ‘in function’ ! ‘as function’
27/ Page 33 ‘present’ ! ‘presence’
28/ Page 34 ‘precesses’ ! ‘processes’
OK