

**1 Analyzing signatures of aerosol-cloud interactions**  
**2 from satellite retrievals and the GISS GCM to**  
**3 constrain the aerosol indirect effect**

Surabi Menon,<sup>1</sup> Anthony D. Del Genio,<sup>2,3</sup> Yoram Kaufman,<sup>4</sup> Ralf Bennartz,<sup>5</sup>

Dorothy Koch,<sup>2,3</sup> Norman Loeb,<sup>6</sup> and Daniel Orlikowski<sup>7</sup>

---

S. Menon, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, MS90KR109, Berkeley,  
CA 94720, USA. (smenon@lbl.gov)

<sup>1</sup>Lawrence Berkeley National Laboratory,

**Abstract.**

Evidence of aerosol-cloud interactions are evaluated using satellite data from MODIS, CERES, AMSR-E, reanalysis data from NCEP and data from the NASA Goddard Institute for Space Studies climate model. We evaluate a series of model simulations: (1) Exp N- aerosol direct radiative effects; (2) Exp C- Like Exp N but with aerosol effects on liquid-phase cumulus and stra-

---

Berkeley, CA, USA

<sup>2</sup>NASA Goddard Institute for Space Studies, New York, NY, USA.

<sup>3</sup>Columbia University, New York, NY, USA.

<sup>4</sup>Formerly at NASA Goddard Space Flight Center, Greenbelt, Maryland, USA, now deceased.

<sup>5</sup>Department of Atmospheric and Oceanic Sciences, University of Wisconsin, Madison, WI, USA

<sup>6</sup>NASA Langley Research Center, Hampton, VA, USA

<sup>7</sup>Lawrence Livermore National Laboratory, Livermore, CA, USA

10 tus clouds; (3) Exp CN- Like Exp C but with model wind fields nudged to  
11 reanalysis data. Comparison between satellite-retrieved data and model sim-  
12 ulations for June to August 2002, over the Atlantic Ocean indicate the fol-  
13 lowing: a negative correlation between aerosol optical thickness (AOT) and  
14 cloud droplet effective radius ( $R_{eff}$ ) for all cases and satellite data, except  
15 for Exp N; a weak but negative correlation between liquid water path (LWP)  
16 and AOT for MODIS and CERES; and a robust increase in cloud cover with  
17 AOT for both MODIS and CERES. In all simulations, there is a positive cor-  
18 relation between AOT and both cloud cover and LWP (except in the case  
19 of LWP-AOT for Exp CN). The largest slopes are obtained for Exp N, im-  
20 plying that meteorological variability may be an important factor. The main  
21 fields associated with AOT variability in NCEP/MODIS data are warmer  
22 temperatures and increased subsidence for less clean cases, not well captured  
23 by the model. Simulated cloud fields compared with an enhanced data prod-  
24 uct from MODIS and AMSR-E indicate that model cloud thickness is over-  
25 predicted and cloud droplet number is within retrieval uncertainties. Since  
26 LWP fields are comparable this implies an under-prediction of  $R_{eff}$  and thus  
27 an over-prediction of the indirect effect.

## 1. Introduction

28 The largest uncertainty in climate forcing from the pre-industrial (PI) time period to the  
29 present day (PD) arises from estimates of aerosol-cloud interactions [*Intergovernmental*  
30 *Panel on Climate Change*, 2007]. These aerosol-cloud interactions include the first and  
31 second aerosol indirect effects (AIE) [*Twomey*, 1991; *Albrecht*, 1989]. While these effects  
32 are often described as a climate forcing, feedbacks associated with the response of cloud  
33 properties to changes in the dynamics and the thermodynamic state need to be isolated  
34 in order to quantify cloud reflectivity changes due solely to aerosols. Given this ambiguity  
35 and the large uncertainty in PD and PI aerosol distributions, predictions of the AIE remain  
36 highly uncertain, spanning a range from  $-0.2$  to  $-4.4 \text{ Wm}^{-2}$  [*Menon*, 2004; *Lohmann and*  
37 *Feichter*, 2005].

38 Satellite observations (such as those from the Moderate Resolution Imaging Spectroradi-  
39 ometer (MODIS)) can potentially decipher cloud responses to aerosol changes [*Kaufman*  
40 *et al.*, 2005a] (hereafter KF05) and thereby constrain model parameterizations of aerosol-  
41 cloud interactions [*Lohmann et al.*, 2006; *Quaas and Boucher*, 2005; *Quaas et al.*, 2005;  
42 *Chylek et al.*, 2006; *Storelvmo et al.*, 2006]. Such satellite based comparisons [*Lohmann*  
43 *and Lesins*, 2002; *Quaas and Boucher*, 2005] have been used to suggest that the AIE is  
44 closer to the smaller magnitude of the range of current predictions ( $>-1 \text{ Wm}^{-2}$ ). With  
45 observationally-based constraints on PD simulations, predictions of the AIE in future  
46 decades appear feasible [*Menon et al.* [2007], in preparation].

47 With a view to constraining future AIE predictions, we evaluate PD AIE simulations  
48 obtained with the NASA Goddard Institute for Space Studies (GISS) global climate model

49 (ModelE) using satellite data from MODIS and the Clouds and the Earth's Radiant  
50 Energy System (CERES). We focus our analyses on the Atlantic Ocean region for the  
51 summer season using the same data set from MODIS as analyzed by KF05. KF05 chose  
52 the Atlantic since this region is significantly influenced by aerosols of different types at  
53 different latitudes: marine aerosols for the 30 to 20S region, biomass aerosols for 20S to  
54 5N, dust for the 5 to 30N region and polluted aerosols for 30 to 60N.

55 We simulate aerosol effects on liquid-phase cumulus and stratiform clouds and compare  
56 to a control simulation that includes only aerosol direct effects. In addition, to test the  
57 sensitivity of our results to errors in the GCM general circulation, we conduct another  
58 simulation with winds nudged to reanalysis data. Section 2 describes the methodology,  
59 satellite data and model simulations; Section 3 compares results from satellite data to  
60 model simulations; and in Section 4 reanalysis data from NCEP are examined to evaluate  
61 the influence of meteorological errors on cloud properties. Finally in Section 5 we present  
62 the summary of our study.

## 2. Methodology

63 MODIS-Terra data used in this study are the aggregated  $1^\circ$  daily resolution data for  
64 June to August 2002 for the Atlantic Ocean region (30S-60N, 40E -100W) for liquid-phase  
65 shallow clouds (cloud top pressure (CTP)  $>640\text{hPa}$ ). Simultaneously retrieved aerosol and  
66 cloud properties are available for partly cloud covered  $1^\circ \times 1^\circ$  areas. We specifically examine  
67 aerosol optical thickness (AOT), cloud droplet effective radius ( $R_{eff}$ ), liquid water path  
68 (LWP), water cloud optical thickness ( $\tau_c$ ), cloud cover (CC), cloud top pressure (CTP) and  
69 cloud top temperature (CTT). For the GCM, in addition to these we also analyze cloud  
70 droplet number concentration (CDNC) and shortwave cloud radiative forcing (SWCRF)

71 fields. LWP is estimated from the product of  $R_{eff}$  and  $\tau_c$ . An error in MODIS's retrieval  
 72 procedure that may cause it to report the presence of clouds for large AOT necessitated  
 73 removal of values for  $AOT > 0.6$  (3% of the data). A similar constraint was also placed on  
 74 CERES and simulated data. Additionally, meteorological fields from the NCEP reanalysis,  
 75 namely temperature, horizontal winds and vertical velocity fields at various pressure levels  
 76 are also examined.

Although MODIS retrievals do not distinguish between types of aerosols, the fractions  
 in the submicron mode allow some distinction between aerosol types as suggested in  
 KF05. Since the contribution of dust aerosols to cloud properties (dependent in part on  
 solubilities assumed and its mixing with other aerosols), is not well known, we estimate  
 the dust contribution to total AOT in the dust zones (5 to 30N) and subtract the dust  
 AOT from the total AOT following *Kaufman et al.* [2005b]. The dust AOT ( $AOT_{du}$ ) is  
 calculated as:

$$AOT_{du} = \frac{[AOT(f_{an} - f) - AOT_{ma}(f_{an} - f_{ma})]}{(f_{an} - f_{du})} \quad (1)$$

77 where  $f$ , the fine mode fraction is obtained from retrievals and  $f_{ma}$ ,  $f_{an}$ , and  $f_{du}$  are the  
 78 marine, anthropogenic and dust components, respectively, of the fine mode fraction.  $f$  is  
 79 bounded by  $f_{an}$  and  $\min[f_{an}, f_{du}]$  and  $f_{an} = 0.9 \pm 0.05$ ;  $f_{du} = 0.5 \pm 0.05$ ;  $f_{ma} = 0.3 \pm 0.1$  and  
 80  $AOT_{ma} = 0.06$ . The assumed values for the fine mode fraction for the different aerosol  
 81 types are obtained from MODIS aerosol measurements in regions with high concentrations  
 82 of dust, smoke and maritime aerosols. For values of  $AOT_{du} > 0.1$  errors are estimated to  
 83 be upto 10 to 15% as described in *Kaufman et al.* [2005b].

84 As a check on the MODIS retrieved aerosol and cloud products, particularly  $R_{eff}$ , since  
 85 MODIS retrievals may overestimate  $R_{eff}$ , we use data from CERES that include AOT,

86  $R_{eff}$ ,  $\tau_c$ , LWP and CC. These fields are then compared to data from MODIS as well  
87 as model simulated fields. CERES data used here are subject to similar constraints as  
88 are MODIS fields for AOT values ( $AOT < 0.6$ ) and we examine liquid-phase low level-  
89 clouds ( $CTP > 640\text{hPa}$ ) only. The CERES AOT values are determined directly from the  
90 MODIS aerosol data product for  $10 \times 10 \text{ km}^2$  domains that are simply averaged into CERES  
91 footprints by convolving them with the CERES point-spread function. Cloud properties  
92 are obtained by applying a cloud retrieval algorithm to MODIS radiances following the  
93 methodology of *Minnis et al.* [2003]. These cloud algorithms are different from the ones  
94 used to retrieve MODIS cloud properties. While LWP values from both CERES and  
95 MODIS are based on the product of  $R_{eff}$  and  $\tau_c$ ,  $R_{eff}$  for MODIS is based on retrievals  
96 from the 2.1 micron channel compared to the 3.7 micron channel used for CERES retrieved  
97  $R_{eff}$ . Additionally, for CERES data, a log average value for mean  $\tau_c$  over a grid box is  
98 used compared to a linear average used by MODIS. This essentially results in lower  $\tau_c$   
99 values for CERES data.

100 To validate some of the simulated cloud properties, we also use enhanced data-sets  
101 described in *Bennartz* [2007] that include CDNC and cloud thickness inferred from MODIS  
102 data (onboard Aqua), LWP,  $\tau_c$ ,  $R_{eff}$  and CC for assumed adiabatically stratified clouds.  
103 The derived LWP product from MODIS is compared to LWP retrievals from the passive  
104 microwave Advanced Microwave Scanning Radiometer (AMSR-E) that is co-located with  
105 MODIS-Aqua. CDNC and cloud thickness are obtained from independent retrievals of  
106 LWP, CC and  $\tau_c$  along with a few parameters (condensation rate, scattering efficiency  
107 and dispersion factor for  $R_{eff}$ ) that may impact retrieval accuracy depending on the  
108 assumptions made. *Bennartz* [2007] estimates a retrieval uncertainty of better than 80%

109 and 20% for CDNC and cloud thickness, respectively, for cloud fraction  $>0.8$  and higher  
110 uncertainties for low LWP and cloud fractions. Furthermore, a difference of a constant  
111 factor of 0.83 is expected in LWP estimates based on the vertically homogeneous versus  
112 adiabatically stratified cloud assumptions. At low values of LWP, AMSR-E values exceed  
113 those from MODIS and at high values the opposite is true. An in-depth explanation of  
114 the derivation of the enhanced data products and the retrieval uncertainties are given in  
115 *Bennartz [2007]*. The Bennartz products differ from the standard MODIS products we use  
116 in several ways: the passage time of Aqua (1:30 pm) is different from that of Terra (10:30  
117 am), adiabatically stratified clouds are assumed as opposed to a vertically homogeneous  
118 cloud for the standard MODIS retrievals, and retrievals are only performed by Bennartz  
119 for  $CC > 50\%$ . Thus, we restrict our analysis to a shorter subset of fields: CDNC,  $R_{eff}$ ,  
120 LWP,  $\tau_c$  and cloud thickness.

121 For simulations, we use the newly developed GISS GCM (ModelE) [*Schmidt et al.*,  
122 2006] ( $4^\circ \times 5^\circ$  and 20 vertical layers) that includes a microphysics based cumulus scheme  
123 [*Del Genio et al.*, 2005], coupled to an on-line aerosol chemistry and transport model [*Koch*  
124 *et al.*, 2007, 2006]. Aerosols simulated include sulfates, organic matter (OM), black carbon  
125 (BC) and sea-salt [*Koch et al.*, 2007, 2006], with prescribed dust [*Hansen et al.*, 2005].  
126 A description of the aerosol emissions, processes treated and schemes used to couple the  
127 aerosols with the clouds is given in *Koch et al. [2007]* and *Menon and Del Genio [2007]*.  
128 PD simulations use emission data from 1995 [*Koch et al.*, 2007], meant to reflect current  
129 day conditions. We perform several sets of simulations, mainly to illustrate changes to  
130 cloud properties for different representations of aerosol effects on cloud properties.



Table 1 lists the parameterization assumptions used in simulations for CDNC and autoconversion. We calculate  $R_{eff}$  as in *Liu and Daum* [2002]:

$$R_{eff} = R_{vol}\beta \quad (2)$$

where  $R_{vol}$ , the volume-weighted mean droplet radius is

$$R_{vol} = \left( \frac{3\mu}{4CDNC\pi\rho_w} \right)^{\frac{1}{3}} \quad (3)$$

and  $\beta$  is an increasing function of the relative dispersion of the cloud drop size distribution (ratio of standard deviation to mean radius) given as

$$\beta = \frac{(1 + 2 * (1 - 0.7 * \exp(-0.003 * CDNC))^2)^{\frac{2}{3}}}{(1 + (1 - 0.7 * \exp(-0.003 * CDNC))^2)^{\frac{1}{3}}} \quad (4)$$

The  $\tau_c$  is then calculated as

$$\tau_c = \frac{1.5\mu\Delta H}{R_{eff}\rho_w} \quad (5)$$

131 Here,  $\mu$  is the cloud liquid water content (LWC),  $\rho_w$  is density of water and  $\Delta H$  is the  
132 cloud thickness.

133 In simulation Exp N, we do not let aerosols affect cloud microphysics, but we do allow  
134 for direct radiative effects of aerosols. In the second simulation, Exp C, we allow aerosols  
135 to modify liquid-phase stratus and shallow cumulus clouds, through changes in CDNC and  
136 autoconversion as described in Table 1. *Menon and Rotstajn* [2006] performed sensitivity  
137 studies with two climate models and found large differences in the AIE and in condensate  
138 distributions when including aerosol effects on cumulus clouds. These were related to  
139 specific model processes used to distribute cumulus condensate as precipitation or as  
140 anvils. Suppression of precipitation in cumulus clouds leads to an increase in detrained  
141 condensate especially over ocean regions that in turn increases moisture and condensed

142 water available for the creation of stratus clouds. Thus, aerosol effects on cumulus clouds  
143 indirectly affect LWP and precipitation in stratus clouds. We also perform an additional  
144 simulation that mirrors Exp C (Exp CN), except that model horizontal wind fields are  
145 nudged to reanalysis data. All runs use climatological mean sea-surface temperatures and  
146 are run for 6 years (including a spin up of one year). To compare model fields with satellite  
147 retrievals, we use instantaneous values of model fields sampled once every day at cloud  
148 top for the last year of the simulation. Model sampling times are chosen to coincide either  
149 with data from MODIS on Terra or that from MODIS on Aqua. All data are analyzed  
150 for the June to August (JJA) time period.

### 3. Analysis of aerosol and cloud fields

151 As in KF05 we examine low-level clouds with average CTP of 866 hPa, between 30S to  
152 60N and 40E to 100W, over oceans. We do not separate the regions based on latitudinal  
153 distribution as in KF05, but rather examine differences in fields over the whole domain.  
154 Characteristics in AOT and cloud properties from MODIS, CERES and AMSR-E are  
155 compared with model simulations as follows:

#### 3.1. Aerosol Optical Thickness

156 Figure 1 indicates the clear-sky AOT from MODIS, CERES, Exp C and CN. Exp N is  
157 comparable to Exp C. The top and middle panels indicate total AOT at  $0.55 \mu\text{m}$  from  
158 MODIS and CERES without and with the dust contribution. The bottom panel indicates  
159 instantaneous clear-sky total visible AOT without dust from Exp C and CN since we use  
160 prescribed dust fields and do not let dust modify cloud properties via its effects on CDNC.  
161 If dust contributions are included, higher values of AOT are observed near 5 to 30N (as in

162 Fig.1 of KF05). A difference in cloud algorithms between MODIS and CERES will lead to  
163 sampling differences over regions and days that could cause differences in the AOT values  
164 used since the data are sampled for partly-cloudy conditions for simultaneous retrievals  
165 of AOT and cloud products. For days and locations that coincide, values are similar for  
166 both CERES and MODIS as expected. Major differences between CERES and MODIS  
167 AOT are over the dust regions, where differences in total AOT and fine fraction (mainly  
168 due to the sampling differences and assumptions used in Eq. (1)) add to produce larger  
169 differences in the AOT product filtered for dust. Without the dust filtering, AOT values  
170 over the dust zone are fairly similar as shown.

171 Excluding the larger values of AOT usually found in the dust zones (5 to 30N), the  
172 major aerosol regions are off the west coast of Africa (20S to 5N), from biomass source  
173 regions, and off the east coast of North America, where the sources are the industrial and  
174 transportation sectors. *Kaufman et al.* [2005c] provide an in-depth analysis on MODIS  
175 AOT error estimates over the ocean for various issues such as aerosol growth, cloud con-  
176 tamination, sun glint, etc. While cloud contamination causes an error of  $0.02 \pm 0.005$  in  
177 MODIS AOT, side-scattering from clouds was not found to cause an artificial increase in  
178 AOT and is not considered a major issue for analyzing aerosol impacts on cloud micro-  
179 physics with MODIS [*Kaufman et al.*, 2005c]. A general bias between MODIS AOT and  
180 model estimates of AOT of about 0.04 in the mean values for ocean regions is reduced  
181 to 0.02 when accounting for aerosol growth [*Kaufman et al.*, 2005c]. The standard error  
182 in MODIS AOT over the ocean for non-dust aerosols is  $\delta\text{AOT} = \pm 0.05 \text{ AOT} \pm 0.03$  with  
183 slightly higher errors for dust (KF05).

184 Model estimates of AOT are usually underestimated when compared to observations,  
185 especially over tropical oceans [Kinne *et al.*, 2006], and our simulations are no excep-  
186 tion. Over the biomass burning areas (west coast of Africa) model AOT is especially  
187 underestimated compared to MODIS. With nudged winds, the sea-salt production rate  
188 increases since it depends on wind speed, and the overall increase in AOT is about 20%,  
189 with increases over most of the domain especially near the biomass zone, due to increased  
190 advection of aerosols from the continent (based on wind directions shown in Fig. 7). A  
191 previous comparison of model aerosol fields (with similar aerosol effective radii as used  
192 in this work but different spatial distributions) with several satellite retrievals indicates  
193 that the spatial and seasonal variability are comparable to satellite retrievals, but that  
194 the assumed aerosol sizes in the GCM may lead to an underestimation in AOT [Liu *et al.*,  
195 2006]. While assumed aerosol sizes can lead to a factor of two difference in AOT, a defi-  
196 ciency of natural aerosols in southern tropical regions [Koch *et al.*, 2006] can also lead to  
197 the lower bias in simulated AOT. However, this should not affect CDNC prediction, that  
198 modulates GCM cloud properties, since our CDNC formulation is based on aerosol mass.

### 3.2. Cloud property changes due to aerosols

199 In this section we compare model mean cloud property fields with MODIS and CERES.  
200 Table 2 indicates mean values and standard deviations of several properties from MODIS,  
201 CERES and simulations. While simulated LWP and CC are comparable to MODIS and  
202 CERES (except the high/low LWP for Exp N/CERES), simulated AOT values are much  
203 lower than MODIS and CERES. Simulated  $R_{eff}$  agrees better with CERES than MODIS.  
204 Reasons for the differences in these products are discussed as follows:

#### 3.2.1. Variation in cloud droplet size and liquid water path with AOT

206 Figure 2 shows the  $R_{eff}$  distributions from MODIS, CERES and Exp C, as well as the  
207 simulated CDNC from Exp C. Although model AOT is underestimated, there is clear  
208 evidence of a change (larger values) in CDNC (dependent on mass-based estimates of  
209 aerosols) between the North and South Atlantic, and to some extent along the continental  
210 edges, where  $R_{eff}$  is also smaller, somewhat similar to the changes evident in MODIS AOT  
211 retrievals. In general, model cloud fields exhibit smaller  $R_{eff}$  and larger CDNC (except for  
212 Exp N since CDNC is constant) in the more polluted North Atlantic sector (sulfate and  
213 carbonaceous aerosols from fossil- and biofuel are more dominant in the North Atlantic  
214 and sea-salt and carbonaceous aerosols from biomass are more prevalent in the South  
215 Atlantic).

216 Simulated  $R_{eff}$  is largely underestimated compared to that retrieved from MODIS, and  
217 around  $1 \mu\text{m}$  smaller compared to CERES, as shown in Table 2 and Fig. 2. Similar results  
218 for comparison of model simulated  $R_{eff}$  fields with MODIS were obtained from other  
219 studies [*Storelvmo et al.*, 2006; *Lohmann et al.*, 2006]. For bumpy inhomogeneous cloud  
220 fields MODIS may over-predict  $R_{eff}$  and under-predict  $\tau_c$ , though this should not preclude  
221 using the dataset to examine changes in  $R_{eff}$  for changing AOT conditions (KF05). Values  
222 retrieved from CERES are much lower than MODIS, especially along the eastern parts  
223 of the Atlantic. Differences in retrievals from the 2.1 versus 3.7 micron channel used for  
224 MODIS and CERES, respectively, alone cannot account for the differences in retrieved  
225  $R_{eff}$  and exact reasons for the differences are not known and is beyond the scope of this  
226 analysis.

227 In general,  $R_{eff}$  in Fig. 2 is smaller in polluted regions than in cleaner regions in both  
228 datasets and in Exp C and CN. The same is not true for Exp N (not shown). By definition

of the first AIE, an increase in AOT can lead to a decrease in  $R_{eff}$  if LWC stays unchanged. LWC estimates are not available from satellite, but the spatial relationships we observe are at least consistent with an AIE signal. Since model differences in  $R_{eff}$  for increases in AOT for Exp C and Exp CN are smaller than those from MODIS and CERES, we analyze the variability between AOT and  $R_{eff}$  for different ranges of LWP, since varying LWP may influence the  $R_{eff}$ -AOT relationship. Figure 3 shows the correlation coefficients for  $R_{eff}$ -AOT versus LWP averaged over selected LWP bins ( $20 \text{ gm}^{-2}$  for  $LWP < 100 \text{ gm}^{-2}$ ;  $50 \text{ gm}^{-2}$  for  $100 < LWP < 350 \text{ gm}^{-2}$ ; and for  $LWP > 350 \text{ gm}^{-2}$ ) for CERES, MODIS, and Exp N, C and CN. For cases where LWP values are roughly similar, the negative correlations between  $R_{eff}$  and AOT should prevail if aerosols influence  $R_{eff}$ . As shown in Fig. 3, both MODIS and CERES indicate a negative correlation between  $R_{eff}$  and AOT, except at the higher ranges in LWP where CERES indicates a positive correlation for  $R_{eff}$ -AOT. For simulations, Exp C is mostly negative, whereas Exp CN and Exp N are more positive. For Exp N, since LWP values are rather large and CDNC is fixed,  $R_{eff}$  also increases since we have no aerosol-induced modification of cloud properties (autocoverison is a function of condensate only) that may alter the distribution of LWP that may be more determined by non aerosol-cloud effects.

Thus, the positive correlations we find cannot simply be explained as that due to varying LWP. Modifications to the precipitation efficiency may result in situations where LWP may increase or decrease with increasing aerosols. This was found to depend on the humidity conditions above cloud and the entrainment of dry air, such that only for moist overlying air masses with low CDNC does cloud water increase with aerosols; and for cases with enhanced entrainment of dry air, cloud water decreases with an increase in CDNC

252 [*Ackerman et al.*, 2005]. Spatial distributions of the correlation between LWP and AOT for  
253 MODIS, CERES and simulations indicate an overall positive relationship with a negative  
254 correlation found in biomass regions and the eastern North Atlantic region for MODIS  
255 and to some extent for CERES. The increase in LWP with AOT is more pronounced in  
256 Exp N, indicating that non aerosol-cloud effects play a stronger role in modulating LWP  
257 over the ocean. Since LWP is a derived product and may mask liquid water variability  
258 if cloud thickness varies, a more conclusive reasoning for spatial variations between  $R_{eff}$   
259 with AOT is hard to obtain.

260 Thus, observational signals to evaluate the first and second AIE are complicated, since  
261 these include changes to LWP and CC that may even be more obscured by feedbacks  
262 or meteorological variability. As shown in Table 2, mean LWP fields for Exp C and CN  
263 are somewhat comparable to MODIS (about 5% higher), but are higher than CERES.  
264 The lower LWP values for CERES compared to MODIS may partly be related to the log  
265 average values used for  $\tau_c$  and the lower  $R_{eff}$ . However, since LWP is a derived product  
266 for both CERES and MODIS, evaluation of this field may be obscured if there are biases  
267 in  $\tau_c$  and  $R_{eff}$ . Since we cannot evaluate retrieval uncertainties in these products within  
268 the scope of our analysis, to at least understand if biases exist in simulated  $R_{eff}$  and  $\tau_c$ ,  
269 the standard  $(\tau_c, R_{eff})$  and enhanced data products, such as CDNC and cloud thickness,  
270 derived from MODIS (on Aqua) with collocated retrievals of LWP from AMSR-E from  
271 *Bennartz* [2007] are used to evaluate some of the cloud microphysics products from Exp  
272 C.

### 273 **3.2.2. Simulated cloud microphysical fields versus those derived from satellite**

274 Here, we perform an analysis of cloud microphysical fields using the derived data set from  
275 *Bennartz* [2007] that includes cloud thickness, CDNC,  $\tau_c$ ,  $R_{eff}$  and LWP from MODIS  
276 (onboard Aqua) versus those simulated for Exp C. Also included are LWP retrievals from  
277 AMSR-E (also onboard Aqua). These data sets (both for retrievals and simulations) are  
278 obtained at a different time interval than those used in the prior sections and do not  
279 include AOT fields. Figure 4 shows CDNC, cloud thickness,  $R_{eff}$  and  $\tau_c$  inferred from  
280 MODIS and that from Exp C. Figure 5 shows LWP inferred from MODIS, obtained from  
281 AMSR-E and that from Exp C. In general, we note that model CDNC values are within  
282 retrieval uncertainties (though lower by 46% compared to the average value inferred from  
283 MODIS) and cloud thickness is over-predicted by a factor of 1.5 compared to the average  
284 values obtained from retrievals. The apparent differences in CDNC fields may in part  
285 be related to assumptions used in CDNC calculations for simulations, that are based on  
286 empirical observations and do not capture the higher values, especially near continental  
287 edges, and the higher uncertainty in CDNC estimates from retrievals (80%), especially at  
288 low LWP values found here (see for example Fig. 3 in *Bennartz* [2007]).

289 LWP values for Exp C (average of  $76 \text{ gm}^{-2}$ ) are comparable to MODIS and AMSR-E ( $70$   
290  $\text{gm}^{-2}$ ), thus suggesting that liquid water contents in the model may be under-estimated  
291 since LWP is the vertical integral of LWC over cloud thickness. However, since the  
292 uncertainty in cloud thickness retrievals are small (20%) and models in general tend to  
293 over predict cloud thickness (coarse resolution being one aspect of the problem since all  
294 simulations have similar cloud thickness values), the over-prediction of simulated cloud  
295 thickness must imply lower LWC values for simulations that include aerosol-induced cloud  
296 modifications.



297 Estimates for  $R_{eff}$  for Exp C (average of  $12.2 \mu\text{m}$ ) are about  $2 \mu\text{m}$  smaller than that  
298 retrieved for MODIS ( $14.3 \mu\text{m}$ ) and  $\tau_c$  values for Exp C (9.2) were comparable to MODIS  
299 (8.6). Closer agreement between MODIS and Exp C indicated here, compared to values  
300 shown in Table 2, may be related to the uncertainties in the simulated diurnal cycle of  
301 the clouds or retrieval issues that are more difficult to verify. Retrieval assumptions for  
302 vertically homogeneous versus adiabatically stratified clouds should not lead to differences  
303 in  $R_{eff}$  and  $\tau_c$  retrievals nor should differences in the dispersion term used to convert  $r_{vol}$   
304 to  $R_{eff}$  for MODIS and Exp C (an average value of  $1.08 \pm 0.06$  is used by *Bennartz* [2007],  
305 and for Exp C the value for dispersion (given by the  $\beta$  term in Eq. 1) varies between 1.1  
306 and 1.6 with a central value of  $1.14 \pm 0.05$ ). Based on the above comparisons we find that  
307 simulated CDNC is within retrieval uncertainties but low biases exist in simulated cloud  
308 liquid water (based on the over-estimation of cloud thickness) and thus,  $R_{eff}$ .

### 309 **3.2.3. Estimating the response of cloud property changes to AOT**

310 Patterns of correlations between all the variables examined here (from MODIS-Terra,  
311 CERES and simulations) with AOT are shown in Fig. 6 and provide a visual analysis of  
312 trends across simulations, MODIS and CERES (CERES values for CTT are not available  
313 here and are indicated as 0). MODIS does indicate an increase in CC and  $\tau_c$  and a decrease  
314 in  $R_{eff}$  with increasing AOT as does CERES. Other variables, such as CTT and CTP  
315 appear to be more correlated to CC (negative correlations) than AOT, with a somewhat  
316 positive association between warmer clouds and AOT and a negative correlation between  
317 CTP and AOT. However, CERES indicates a positive relationship between CTP and  
318 AOT similar to simulations. In all simulations, an overall increase in LWP (except for  
319 Exp CN),  $\tau_c$  and CC with aerosols is observed, especially for Exp N.

320 Since the relationships between cloud properties and aerosols are not necessarily linear,  
321 we examine the magnitudes of slopes based on log-log [Sekiguchi *et al.*, 2003] or log-linear  
322 relationships, depending on the range and best fit line to the data. Table 3 shows the  
323 slopes between AOT and the variables of interest for MODIS, CERES and simulations. We  
324 note that model slopes for  $R_{eff}$  and AOT are severely underestimated w.r.t. MODIS and  
325 CERES. Only Exp N (without aerosol-induced changes to cloud microphysics) exhibits a  
326 positive correlation between AOT and  $R_{eff}$  (due to the higher LWP and fixed CDNC).  
327 For LWP versus AOT, the positive slopes for Exp N and Exp C are in contrast to the  
328 negative slopes from MODIS, CERES and Exp CN. However, only the slopes for Exp N  
329 and CERES were significant at the 95% level. The larger slope for Exp N indicates that  
330 meteorological effects play a role in increasing LWP in areas with high AOT.

331 For CC versus AOT, slopes from all simulations are positive, similar to MODIS and  
332 CERES, but a few factors lower. Since all simulations had fairly similar slopes, we note  
333 that meteorological variability or non-aerosol-cloud effects appear to explain most of the  
334 increase in CC with AOT, similar to the results in Lohmann *et al.* [2006] that indicate a  
335 more dominant non aerosol-cloud effect on CC increase with AOT. As CC increases, so  
336 does relative humidity in the clear regions adjacent to the clouds, resulting in an increase  
337 in AOT and an apparent correlation between AOT and CC. Recent 3D Monte Carlo  
338 simulations of side-scattering from clouds qualitatively capture both increases in AOT  
339 with CC and the spectral dependence in AOT with CC seen in the satellite retrievals  
340 [Wen *et al.*, 2007]. This may explain some of the larger slopes seen in MODIS and perhaps  
341 CERES. Additionally, changes in CC and AOT over regions subject to different dynamical  
342 forcings and different aerosol sources may cause an apparent correlation between AOT and

343 CC that may be misinterpreted as aerosol-cloud interactions. Thus, based on simulations  
344 and the uncertainty in retrievals, correlated changes in CC and aerosols may in large part  
345 be related to meteorological and aerosol humidification effects.

346 Comparing  $\tau_c$ -AOT slopes between model and MODIS/CERES indicates that model  
347 values for Exp N and Exp C are higher than MODIS and CERES, primarily due to the  
348 lower AOT and the higher  $\tau_c$  and the variability in LWP. To understand the changes in  
349 radiative fields, we compare the slopes of SWCRF-AOT amongst simulations. CERES  
350 derived values for SWCRF were not directly comparable to simulated values and hence  
351 is not compared to simulations. For SWCRF versus AOT, Exp C is of similar magnitude  
352 but of opposite sign compared to Exp N. Exp CN is a factor of 1.5 greater than Exp  
353 C. Thus, changes in the radiative fields (SWCRF) from aerosol-induced changes to cloud  
354 microphysics are a factor of 2 to 3 higher than that obtained from non aerosol-cloud  
355 effects. Interestingly, *Lohmann et al.* [2006] find that aerosol-induced changes to cloud  
356 microphysics account for 25% of the change in SWCRF, for simulations with and without  
357 aerosol-cloud interactions. Using  $\tau_c$ -AOT and CC-AOT slope differences between Exp N  
358 and Exp C, we estimate that non aerosol-cloud effects accounts for 57% of the increase in  
359  $\tau_c$  simulated by Exp C and completely dominate the CC increase.

360 Though the mean values for the various properties are similar in Exp C and Exp CN  
361 (except for SWCRF), as shown in Table 2, overall the magnitude of the slopes for Exp  
362 CN are in slightly better agreement with MODIS and CERES than are the slopes for  
363 Exp C (as shown in Table 3). Thus, nudging to observed wind fields with aerosol induced  
364 modification to cloud properties creates conditions that are in closer agreement to satellite-  
365 based retrievals. Clearly, wind-fields and their effects on the response of  $R_{eff}$ , LWP and

366 thus  $\tau_c$  and SWCRF to AOT are different that may be due to AOT fields themselves that  
367 increase slightly with nudged winds, probably resulting in more aerosols advected from  
368 the continent.

369 Thus, in general, model slopes for  $R_{eff}$  and CC are underestimated compared to MODIS  
370 and CERES and the  $\tau_c$ -AOT slope is generally overestimated (probably due to the under-  
371 prediction of  $R_{eff}$  as noted in Sec.3.2.2, and AOT). The largest uncertainty in such an  
372 inference relates to the LWC and meteorological variability with AOT.

#### 4. Meteorological influence on aerosol and cloud properties

373 To further explore the influence of meteorology on cloud properties, we evaluate tem-  
374 perature, wind and vertical velocity fields from NCEP and model simulations. Figure  
375 7 shows temperature and wind fields from NCEP and Exp C at 1000 hPa. Mean tem-  
376 perature fields (at 1000 hPa) from NCEP indicate higher values in the northern tropics  
377 along the east coast of S. America and higher values at 750 hPa along the dust (10-30N)  
378 and biomass (10-20S) zones. NCEP wind fields indicate the presence of easterly winds  
379 between 0 to 15N and south-easterly winds from 20S to 0, transporting dust and biomass  
380 layers towards S America. For the N. Atlantic sector, between 40 to 60N, air masses  
381 (perhaps polluted) from N. America are transported towards Europe. The simulations do  
382 capture the spatial distribution of the temperature fields, with higher values over the trop-  
383 ical areas compared to NCEP. The prevailing wind fields are also comparable to NCEP,  
384 except for weaker westerlies in the N. Atlantic sector. The wind field strength increases  
385 in simulations with aerosol-cloud interactions (especially for the nudged case) compared  
386 to Exp N.

387 NCEP vertical velocity fields indicate uniformly low subsidence over most of the do-  
388 main at 750 hPa (and a bit more so at 500 hPa) except near the equator, where ascent is  
389 observed. Figure 8 shows the probability density function for geometric vertical velocity  
390 at 750 hPa (positive upward) from NCEP and from the simulations. Simulated subsidence  
391 rates are weaker for all model simulations than for NCEP; nudging of winds has only a  
392 minimal effect.

393 To understand changes to aerosol and clouds fields due to meteorological influences,  
394 KF05 performed multiple regression analyses to judge the relative influence of the various  
395 fields and found temperature, followed by wind fields to be more important. We perform  
396 similar analysis, using NCEP and model fields, but instead characterize differences based  
397 on the probability density distributions for particular AOT conditions (above or below  
398 the baseline value of 0.06 for AOT). Figure 9 shows the probability density distributions  
399 for temperature, the U and V component of the horizontal wind fields at 1000 hPa and  
400 vertical velocity fields at 750 hPa, for AOT values below and above 0.06 for MODIS and  
401 the three simulations. Results were similar at other levels (750 and 500 hPa), unless noted  
402 otherwise. Results from Fig.9 indicate an increase in warmer conditions for higher values  
403 of AOT ( $>0.06$ ). This may be simply related to location of aerosol source regions (e.g.  
404 higher dust and biomass sources near the tropics). For simulations, only a slight tendency  
405 towards higher temperature was obtained for differences in AOT. For the high AOT cases,  
406 the mean temperature from NCEP and simulations were similar, but for the low AOT  
407 cases, the mean temperature was about 2 degrees warmer for simulations compared to  
408 NCEP. For wind fields, for low ( $AOT < 0.06$ ) and high AOT cases, NCEP indicates a  
409 slight tendency for easterly and southerly components for the higher AOT cases, and the

410 simulations (especially Exp CN) follow the NCEP distribution for the low AOT case but  
411 the southerly component for the high AOT case is not well simulated.

412 Vertical velocity fields for both NCEP and simulations are similar and exhibit no signif-  
413 icant changes for differences in AOT values. To investigate the association of cloudiness  
414 and pollution with regions of subsidence that could lower the PBL height and trap pol-  
415 lution, we further separate the vertical velocity fields to areas of negative velocities only.  
416 We find no strong evidence of increased subsidence strength associated with clean or less  
417 clean cases from simulations. However, NCEP data do indicate a factor of 2 increase in  
418 subsidence strength for the less clean compared to the clean cases. In subsidence regions  
419 CC does increase for MODIS (62%) and all simulations (about 9%) for the less clean cases.  
420 The increase is similar to that found for all conditions (positive and negative vertical ve-  
421 locity regions). Further analysis of CC changes in areas of greater subsidence (subsidence  
422 values greater than the mean) do not indicate any significant changes in CC based on  
423 changes in subsidence strengths.

## 5. Summary

424 To evaluate model predictions of the aerosol indirect effect, we compare a series of  
425 model simulations with and without aerosol effects on cloud microphysics with data from  
426 MODIS, CERES, AMSR-E and NCEP for the Atlantic Ocean region for June to August  
427 2002. Cloud response to aerosols for liquid-phase shallow clouds are studied in the differ-  
428 ent simulations that include the aerosol direct effect (Exp N), aerosol effects on stratus  
429 and cumulus clouds (Exp C), and for a simulation that mirrors Exp C but with model  
430 horizontal winds nudged to reanalysis data (Exp CN). Analysis of model simulations using  
431 correlation matrices and slopes indicate that simulations without aerosol-induced changes

432 to cloud microphysics (Exp N) did not capture the reduction in  $R_{eff}$  with increasing AOT  
433 seen in satellite data since the less clean cases have a large increase in the LWP fields,  
434 from meteorological effects that dominates the changes in  $R_{eff}$ , and CDNC is fixed in  
435 these simulations. For Exp N, LWP was positively correlated to AOT in contrast to the  
436 negative relationship found for MODIS (not significant) and CERES. The correlation be-  
437 tween LWP and AOT for Exp C (positive) and Exp CN (negative) were not significant.  
438 While both MODIS and CERES data did indicate a strong increase in CC with AOT, all  
439 simulations capture a similar increase, though of lesser magnitude, with non aerosol-cloud  
440 effects dominating CC changes. Although features in Exp CN are also present in Exp C,  
441 nudging to wind fields results in simulations with different dynamics and these simula-  
442 tions improve the response of cloud properties to AOT (based on the comparison of slopes  
443 obtained for simulations versus that for CERES and MODIS shown in Table 3). This ap-  
444 pears to be due to slightly higher values of AOT in Exp CN (nudging to wind fields helps  
445 advect more aerosols from the continent to the ocean thereby reducing the generally low  
446 model AOT bias). However, based on the signs of the slopes, these changes are smaller  
447 than are changes associated with not including aerosol-induced cloud modifications.

448 An association between warmer temperature and higher AOT was found for NCEP  
449 and to a somewhat weaker extent in all simulations. We find a slight increase in the  
450 easterly and southerly wind fields with an increase in AOT (more so for NCEP than  
451 the GCM) and no association between vertical velocities and AOT. While there was no  
452 association between subsidence strength and pollution for the simulations, NCEP/MODIS  
453 did indicate an increase in subsidence strength (factor of 2) for the less clean versus the  
454 clean case. An increase in CC with aerosols in areas of subsidence was found for both

455 NCEP/MODIS fields and simulations that was of similar strength as that obtained for  
456 cases without separating the data into subsidence only regions, indicating that aerosols  
457 were more influential than large-scale subsidence in changing CC.

458 Comparing the magnitudes of the slopes between  $R_{eff}$ -AOT for MODIS/CERES and  
459 Exp C, as a measure of the relative changes in cloud properties due to aerosols, we note  
460 that model slopes are underestimated. However, the  $\tau_c$ -AOT slope is overestimated by  
461 the model (except for Exp CN) compared to MODIS and CERES, and this relates to the  
462 variability and slope of the LWP-AOT relationship that was different between MODIS,  
463 CERES and the simulations, especially Exp N. Clearly, the variability in LWP and an  
464 independent accurate measure of liquid water are critical to AIE estimates.

465 Constraining cloud properties (cloud thickness and CDNC) simulated by Exp C with  
466 those inferred from the enhanced MODIS data set used here (onboard Aqua) and based on  
467 estimates of co-located LWP fields (MODIS and AMSR-E), we conclude that the model  
468 CDNC fields are within retrieval uncertainties but the model significantly over-predicts  
469 cloud thickness (factor of 1.5). Since simulated LWP values are comparable to satellite  
470 estimates, this could imply that simulated LWC and  $R_{eff}$  are also under-predicted. Cloud  
471 changes –increase with an increase in aerosols– are quite robust in MODIS and CERES  
472 data. While cloud changes with aerosols were not as strong in simulations, similar val-  
473 ues found for all simulations suggest that meteorological variability may play a stronger  
474 role in modulating CC.  $\tau_c$ -AOT and CC-AOT slope differences between Exp C and Exp  
475 N indicate that meteorological variability accounts for a 57% increase in  $\tau_c$  and domi-  
476 nates the CC increase. We estimate changes in the SWCRF fields from aerosol-induced  
477 modifications to cloud properties are a factor of 2-3 greater than without aerosol-induced



478 changes to cloud properties, based on the estimated slopes between SWCRF and AOT  
479 for the three simulations (Exp N, C and CN), due to the stronger first AIE.

480 For Exp C, we obtain an annual global average AIE value (defined as the difference  
481 in net cloud radiative forcing between Year 2030 –for the IPCC midline A1B scenario  
482 described in *Unger et al.* [2006]– and Year 2000) of  $-0.68 \text{ W m}^{-2}$  [*Menon et al.*, 2007].  
483 The average value for June to August for the Atlantic Ocean region studied here is  $-0.50$   
484  $\text{Wm}^{-2}$ . Using the best-guess estimate from retrievals ( $R_{eff}$  from CERES,  $\tau_c$  from MODIS  
485 and CC from both), we attempt to evaluate if our AIE is over or under- predicted for Exp  
486 C, based on changes in  $\tau_c$ ,  $R_{eff}$  and CC with AOT. From Table 3, we find that:

- 487 (1) the slope of CC w.r.t. AOT is underestimated by  $\sim 80\%$  and  $70\%$ , compared to MODIS  
488 and CERES, respectively;
- 489 (2) the slope of  $\tau_c$  w.r.t. AOT is about a factor of 2.2 higher compared to MODIS; and  
490 (3) the slope of  $R_{eff}$  w.r.t AOT is underestimated by  $90\%$  compared to CERES.

491 Thus, as a rough approximation we estimate that Exp C may slightly over-predict the  
492 indirect effect compared to best-guess MODIS/CERES estimates.

493 Summarizing the main points of our study, in spite of several caveats present in satellite  
494 and model fields analyzed here, we find that:

- 495 (1)  $R_{eff}$  decreases with an increase in AOT, averaged over the entire domain, are robust  
496 in MODIS and CERES retrievals and are present to some extent in simulations where  
497 aerosols modify cloud properties;
- 498 (2) CC increases with AOT are especially robust in MODIS and CERES retrievals and are  
499 also noted in model simulations, with meteorological variability providing the dominant  
500 signal for simulated CC changes;

501 (3)  $\tau_c$  increases with an increase in AOT in MODIS and CERES are smaller compared to  
502 simulations;

503 (4) association between a small subset of large-scale meteorological fields examined here  
504 (temperature, horizontal winds and vertical velocity) and AOT, from NCEP and sim-  
505 ulations indicate warmer temperatures in areas of higher AOT ( $>0.06$ ), more related  
506 to location of source regions, and an increase in subsidence strength with pollution for  
507 NCEP/MODIS;

508 (5) nudging to wind fields in simulations that include aerosol-induced changes to clouds  
509 improves the response of cloud properties to differences in AOT (based on slopes between  
510 Exp C, CN, MODIS and CERES shown in Table 3) probably due to improved AOT dis-  
511 tributions themselves;

512 (6) our standard simulation (Exp C) predicts CDNC within retrieval uncertainties but  
513 under-predicts LWC compared to data inferred from MODIS and AMSR-E and thus may  
514 under-predict  $R_{eff}$ ; that may explain the overestimation in  $\tau_c$  and SWCRF.

515 We believe that the above analyses can only be considered as a very broad approximation  
516 or a first guess attempt to constrain the AIE magnitude. Contextualizing the major  
517 objective of this work, constraining present-day AIE simulations to better predict the  
518 future, it appears that our values for Exp C, our standard simulation, may only be slightly  
519 overestimated for the ocean region. To better understand the global-scale implications of  
520 the above analysis since land signals are different compared to ocean signals (AOT and  
521 CDNC values and thus AIE are higher over land), ongoing future work will extend the  
522 present analysis globally with an emphasis on variations of key features of aerosol-cloud

523 interactions isolated for specific meteorological regimes with co-located MODIS, AMSR-E  
524 and radiation data from CERES.

525 **Acknowledgments.** We gratefully acknowledge funding from the NASA MAP and  
526 Radiation Sciences Program managed by Don Anderson and Tsengdar Lee. This paper is  
527 dedicated to the memory of Yoram Kaufman. We gratefully acknowledge the contributions  
528 from S. Bauer, whose reanalyses techniques have been used in model simulations, and I.  
529 Koren and S. Mattoo for help with interpreting MODIS and NCEP data used in this  
530 study.

## References

- 531 Ackerman, A. S., M. P. Kirkpatrick, D. E. Stevens, and O. B. Toon, The impact of  
532 humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, *432*, 1014–  
533 1017, 2005.
- 534 Albrecht, B. A., Aerosols, cloud microphysics, and fractional cloudiness, *Science*, *245*,  
535 1227–1230, 1989.
- 536 Bennartz, R., Global assessment of marine boundary layer cloud droplet number concen-  
537 tration from satellite, *J. Geophys. Res.*, *112*, doi:10.1029/2006JD007547, 2007.
- 538 Chylek, P., M. Dubey, U. Lohmann, V. Ramanathan, Y. Kaufmann, G. Lesins, J. Hudson,  
539 G. Altmann, and S. Olsen, Aerosol indirect effect over the Indian Ocean, *Geophys. Res.*  
540 *Lett.*, *33*, L06,806, 2006.
- 541 Del Genio, A. D., M.-S. Yao, W. Kovari, and K.-W. Lo, A prognostic cloud water param-  
542 eterization for global climate models, *J. Climate*, *9*, 270–304, 1996.

- 543 Del Genio, A. D., W. Kovari, M.-S. Yao, and J. Jonas, Cumulus microphysics and climate  
544 sensitivity, *J. Climate*, *18*, 2376–2387, 2005.
- 545 Hansen, J., M. Sato, R. Ruedy, L. Nazarenko, and A. L. et al., Efficacy of climate forcings,  
546 *J. Geophys. Res.*, *110*, doi:10.1029/2005JD005776, 2005.
- 547 Intergovernmental Panel on Climate Change, *Climate Change 2007: The Physical Science*  
548 *Basis*, Cambridge University Press, (S. Solomon, D. Qin, M. Manning, Z. Chen, M.  
549 Marquis, K.B. Averyt, M.Tignor and H.L. Miller , Eds.), 2007.
- 550 Kaufman, Y., I. Koren, L. Remer, D. Rosenfeld, and Y. Rudich, The effect of smoke, dust,  
551 and pollution aerosol on shallow cloud development over the Atlantic Ocean, *Proc. Natl.*  
552 *Acad. Sci.*, *32*, 11,207–11,212, 2005a.
- 553 Kaufman, Y., I. Koren, L. Remer, D. Tanre, P. Ginoux, and S. Fan, Dust trans-  
554 port and deposition observed from the Terra-Moderate Resolution Imaging Spec-  
555 troradiometer (MODIS) spacecraft over the Atlantic Ocean, *J. Geophys. Res.*, *100*,  
556 doi:10.1029/2003JD004436, 2005b.
- 557 Kaufman, Y., et al., A critical examination of the residual cloud contamination and diurnal  
558 sampling effects on modis estimates of aerosol over ocean, *IEEE Trans. Geosci. Remote*  
559 *Sens.*, *43*, 2886–2897, 2005c.
- 560 Kinne, S., M. Schulz, C. Textor, S. Guibert, and U. L. et al., An aerocom initial assessment  
561 - optical properties in aerosol component modules of global models, *Atmos. Chem. Phys.*  
562 *Discuss.*, *6*, 1815–1834, 2006.
- 563 Koch, D., G. Schmidt, and C. Field, Sulfur, sea salt and radionuclide aerosols in GISS,  
564 ModelE, *J. Geophys. Res.*, *111*, d06206,doi:10.1029/2004JD005550, 2006.

- 565 Koch, D., T. Bond, D. Streets, N. Unger, and G. van der Werf, Global impacts  
566 of aerosols from particular source regions and sectors, *J. Geophys. Res.*, *111*,  
567 d02205,doi:10.1029/2005JD007024, 2007.
- 568 Liu, L., A. Lacis, B. Carlson, M. Mishchenko, and B. Cairns, Assessing a GCM aerosol  
569 climatology using satellite and ground-based measurements: A comparison study, *J.*  
570 *Geophys. Res.*, *111*, doi:10.1029/2006JD007334, 2006.
- 571 Liu, Y., and P. Daum, Anthropogenic aerosols: Indirect warming effect from dispersion  
572 forcing, *Nature*, *419*, 580–581, 2002.
- 573 Lohmann, U., and J. Feichter, Global indirect aerosol effects: A review, *Atmos. Chem.*  
574 *Phys. Discuss.*, *5*, 715–737, 2005.
- 575 Lohmann, U., and G. Lesins, Stronger constraints on the anthropogenic indirect aerosol  
576 effect, *Science*, *298*, 1012–1016, 2002.
- 577 Lohmann, U., I. Koren, and Y. Kaufman, Disentangling the role of microphysical and  
578 dynamical effects in determining cloud properties over the Atlantic, *Geophys. Res. Lett.*,  
579 *33*, L09,802, 2006.
- 580 Menon, S., Current uncertainties in assessing aerosol effects on climate, *Ann. Rev. Envi-*  
581 *ron. Resour.*, *29*, 1–30, 2004.
- 582 Menon, S., and A. Del Genio, *Evaluating the impacts of carbonaceous aerosols on clouds*  
583 *and climate. In Human-induced climate change: An interdisciplinary assessment*, Cam-  
584 bridge University Press, (M. Schlesinger et al., Eds.), 2007.
- 585 Menon, S., and L. Rotstayn, The radiative influence of aerosol effects on liquid-phase  
586 cumulus and stratus clouds based on sensitivity studies with two climate models, *Clim.*  
587 *Dyn.*, *27*, 345–356, 2006.

- 588 Menon, S., N. Unger, D. Koch, D. Streets, D. Shindell, A. Del Genio, and J. Hansen,  
589 Aerosol-cloud interactions and climate change for 1980-2030, *Env. Res. Lettr.*, to be  
590 submitted, 2007.
- 591 Minnis, P., D. F. Young, S. Sun-Mack, P. W. Heck, D. R. Doelling, and Q. Trepte, CERES  
592 cloud property retrievals from imagers on TRMM, Terra, and Aqua, *Proc. SPIE 10th*  
593 *Int. Symp. on Remote Sensing: Conf. on Remote Sensing of Clouds and the Atmosphere*  
594 *VII, Barcelona, Spain*, pp. 37–48, 2003.
- 595 Quaas, J., and O. Boucher, Constraining the first aerosol indirect radiative forcing in  
596 the LMDZ GCM using POLDER and MODIS satellite data, *Geophys. Res. Lett.*, *32*,  
597 L17,814, 2005.
- 598 Quaas, J., O. Boucher, and U. Lohmann, Constraining the total aerosol indirect effect  
599 in the LMDZ and ECHAM4 GCMs using MODIS satellite data, *Atmos. Chem. Phys.*  
600 *Discuss.*, *5*, 9669–9690, 2005.
- 601 Rotstayn, L. D., and Y. Liu, A smaller global estimate of the second indirect aerosol  
602 effect, *Geophys. Res. Lett.*, *32*, doi:10.1029/GL021922, 2005.
- 603 Schmidt, G. A., R. Ruedy, J. E. Hansen, I. Aleinov, and N. B. et al., Present day atmo-  
604 spheric simulations using GISS ModelE: comparison to in-situ, satellite and reanalysis  
605 data, *J. Climate*, *19*, 153–192, 2006.
- 606 Sekiguchi, M., T. Nakajima, K. Suzuki, K. kawamoto, A. Higurashi, D. Rosenfeld,  
607 I. Sano, and S. Mukai, A study of the direct and indirect effects of aerosols us-  
608 ing global satellite data sets of aerosol and cloud parameters, *J. Geophys. Res.*, *108*,  
609 doi:10.1029/2002JD003359, 2003.

- 610 Storelvmo, T., J. Kristjansson, G. Myhre, M. Johnsrud, and F. Stordal, Combined obser-  
611 vational and modeling based study of the aerosol indirect effect, *Atmos. Chem. Phys.*  
612 *Discuss.*, *6*, 3757–3799, 2006.
- 613 Twomey, S., Aerosols, clouds and radiation, *Atmos. Environ.*, *25*, 2435–2442, 1991.
- 614 Unger, N., D. Shindell, D. Koch, and D. Streets, Cross influences of ozone and sulfate  
615 precursor emissions changes on air quality and climate, *Proc. Natl. Acad. Sci.*, *103*,  
616 4377–4380, 2006.
- 617 Wen, G., A. Marshak, R. Cahalan, L. Remer, and R. Kleidmand, 3d aerosol-cloud radia-  
618 tive interaction observed in collocated MODIS and ASTER images of cumulus cloud  
619 fields, *J. Geophys. Res.*, in Review, 2007.

**Table 1.** Expressions used to obtain the cloud droplet number concentration (CDNC) and autoconversion for simulations.  $N_a$  is the aerosol concentration obtained from the aerosol mass for a log-normal distribution as described in *Menon and Rotstayn* [2006].

Variable	Exp N	Exp C-Stratus	Exp C-Cumulus
CDNC-land	175	$-598+298 \log(N_a)$	$174.8 + 1.51 N_a^{0.886}$
CDNC-ocean	60	$-273+162 \log(N_a)$	$-29.6+4.92N_a^{0.694}$
Autoconversion	f(condensate) [ <i>Del Genio et al.</i> , 1996]	f(droplet threshold size) [ <i>Rotstayn and Liu</i> , 2005]	f(droplet threshold size) [ <i>Menon and Rotstayn</i> , 2006]

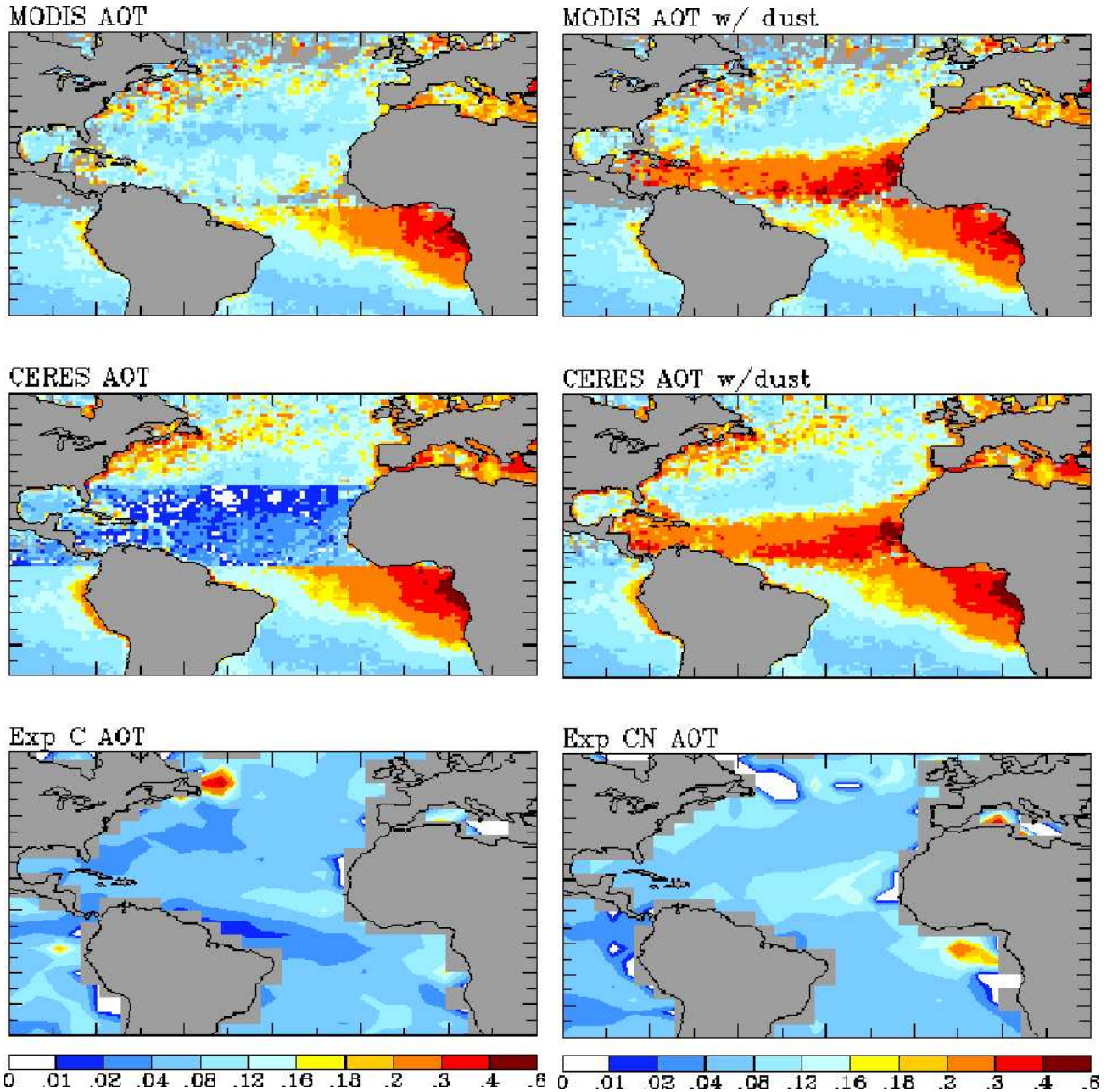


**Table 2.** Average and standard deviations for aerosol optical thickness (AOT), cloud droplet effective radii ( $R_{eff}$ ) ( $\mu\text{m}$ ), liquid water path (LWP) ( $\text{gm}^{-2}$ ), cloud cover (CC) (%), cloud optical depth ( $\tau_c$ ), cloud top temperature (CTT) (K) and cloud top pressure (CTP) (hPa) for MODIS, CERES and the three simulations. Also included for model simulations are shortwave cloud radiative forcing (SWCRF) ( $\text{Wm}^{-2}$ ) values.

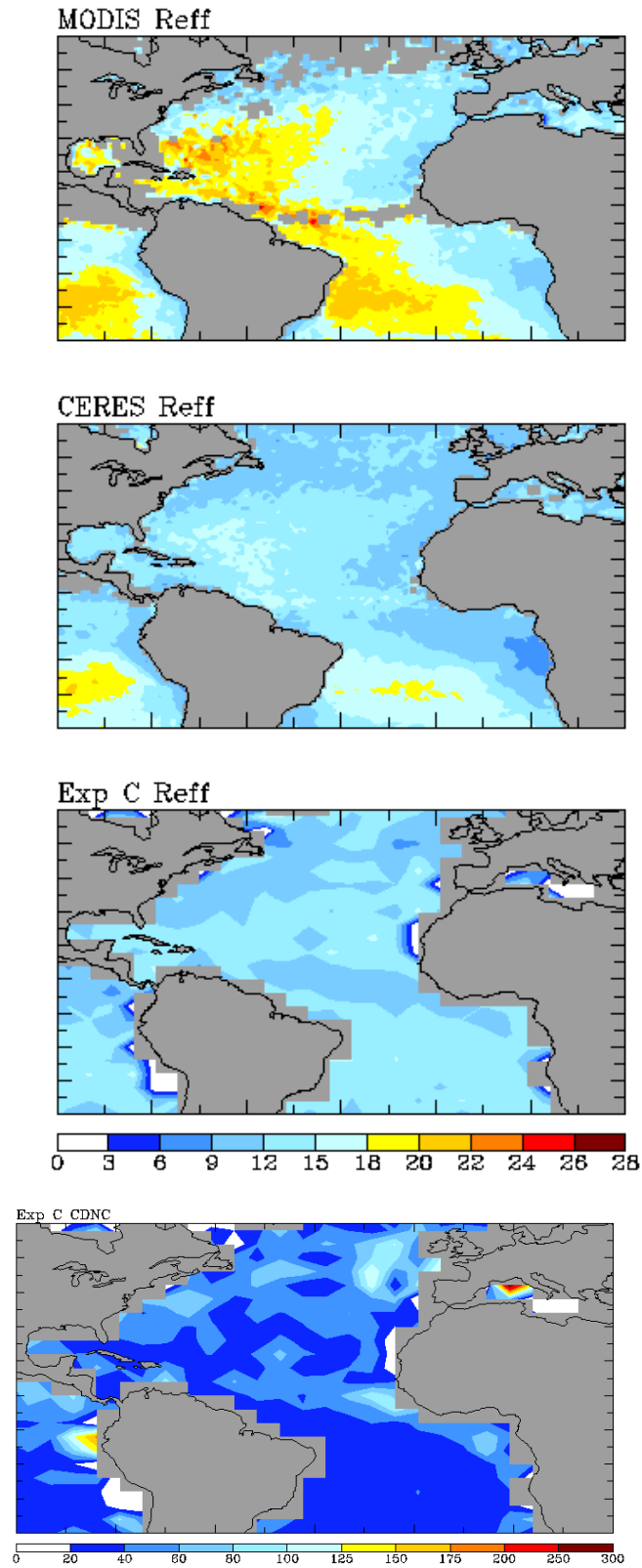
Values	MODIS	CERES	Exp N	Exp C	Exp CN
AOT	0.13±0.09	0.13±0.11	0.06±0.05	0.06±0.05	0.07±0.05
$R_{eff}$	16.7±4.70	13.7±4.36	13.1±4.22	12.6±3.34	12.3±4.02
LWP	67.4±46.7	43.8±37.6	134±167	71.9±65.2	70.6±68.8
CC	41.0±31.6	54.1±32.3	44.9±19.8	46.5±19.3	45.7±20.0
$\tau_c$	5.82±3.52	3.10±3.00	12.8±9.79	8.77±9.17	8.96±10.1
CTT	288±3.62	NA	289±5.38	289±5.33	290±5.41
CTP	866±67.7	878±50.9	896±54.9	895±58.2	898±56.9
SWCRF	NA	NA	-101 ±134	-103±129	-89.9±120

**Table 3.** Summary of slopes between cloud droplet effective radii ( $R_{eff}$ ), liquid water path (LWP), cloud cover (CC), cloud optical depth ( $\tau_c$ ) and shortwave cloud radiative forcing (SWCRF) versus aerosol optical thickness (AOT) for log-log (1) and log-linear (2) relationships for MODIS, CERES and model simulations. Values that are not significant ( $p < 0.05$ ) based on the Student's t-test are indicated in italics.

Slope	MODIS	CERES	Exp N	Exp C	Exp CN
$R_{eff}$ -AOT (1)	-0.11 $\pm$ 0.001	-0.17 $\pm$ 0.001	0.06 $\pm$ 0.01	-0.02 $\pm$ 0.008	-0.06 $\pm$ 0.01
LWP-AOT (1)	<i>-0.004 <math>\pm</math> 0.003</i>	-0.07 $\pm$ 0.03	0.09 $\pm$ 0.04	<i>0.005 <math>\pm</math> 0.03</i>	<i>-0.04 <math>\pm</math> 0.04</i>
CC-AOT (1)	0.40 $\pm$ 0.005	0.23 $\pm$ 0.004	0.07 $\pm$ 0.01	0.07 $\pm$ 0.01	0.05 $\pm$ 0.02
$\tau_c$ -AOT (2)	0.61 $\pm$ 0.01	0.75 $\pm$ 0.01	1.12 $\pm$ 0.24	1.95 $\pm$ 0.22	0.60 $\pm$ 0.28
SWCRF-AOT (2)	NA	NA	15.2 $\pm$ 3.27	-13.0 $\pm$ 3.09	-33.1 $\pm$ 3.29



**Figure 1.** Aerosol optical thickness (AOT) for June-July-August (JJA) without and with the dust contribution to AOT from MODIS (top panel), CERES (middle panel), and AOT without the dust contribution as simulated by the model for Exp C and Exp CN (bottom panel).



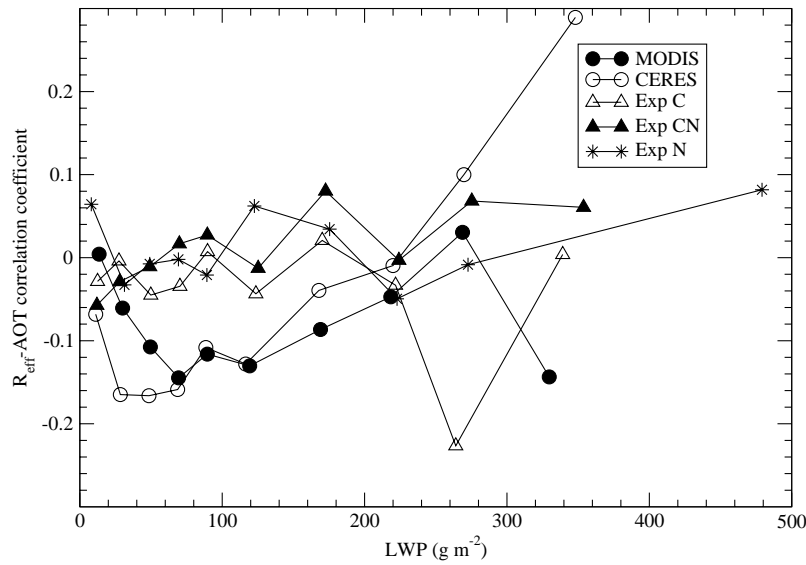
**Figure 2.** Cloud droplet effective radii ( $R_{eff}$ ) ( $\mu\text{m}$ ) for June-July-August (JJA) as retrieved from MODIS, CERES and as simulated by the model for Exp C. Also shown is the cloud droplet

number concentration (CDNC) ( $\text{cm}^{-3}$ ) for Exp C.

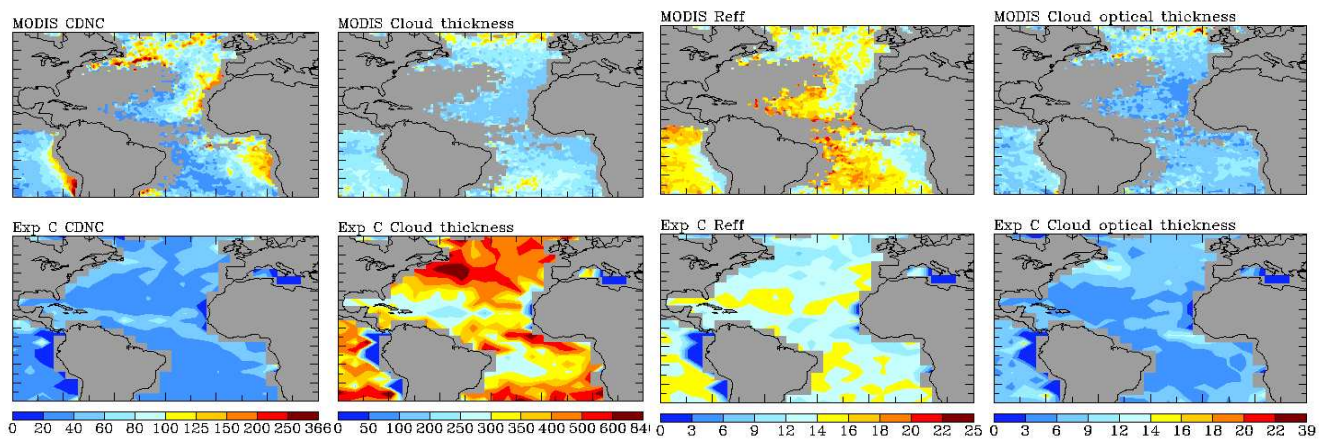
D R A F T

October 1, 2007, 9:31am

D R A F T

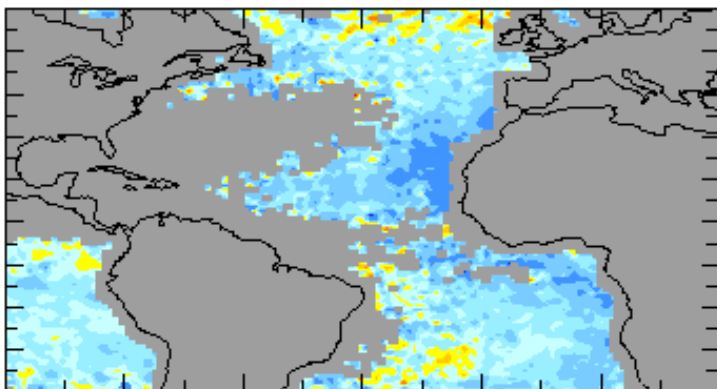


**Figure 3.** Correlation coefficients between cloud droplet effective radii ( $R_{eff}$ ) and aerosol optical thickness (AOT) versus liquid water path (LWP) for June-July-August (JJA) as obtained from MODIS, CERES and as simulated by the model for Exp C, CN and N. Each point represents the average values over a given LWP range.

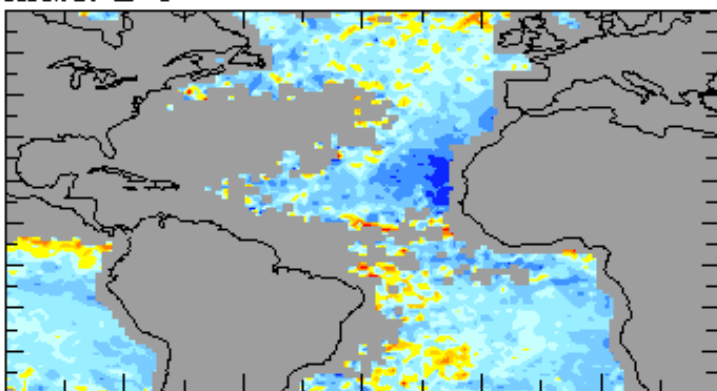


**Figure 4.** Cloud droplet number concentration (CDNC) ( $\text{cm}^{-3}$ ), cloud thickness (m), cloud droplet effective radii ( $R_{eff}$ ) ( $\mu\text{m}$ ) and cloud optical thickness for June-July-August (JJA) as inferred from MODIS (onboard Aqua) and as simulated by the model for Exp C.

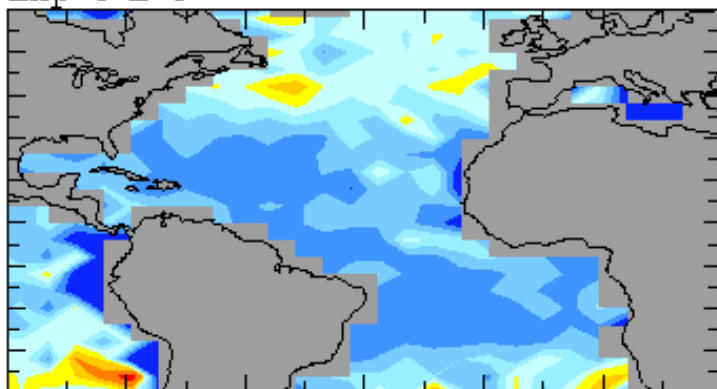
## MODIS LWP



## AMSR LWP

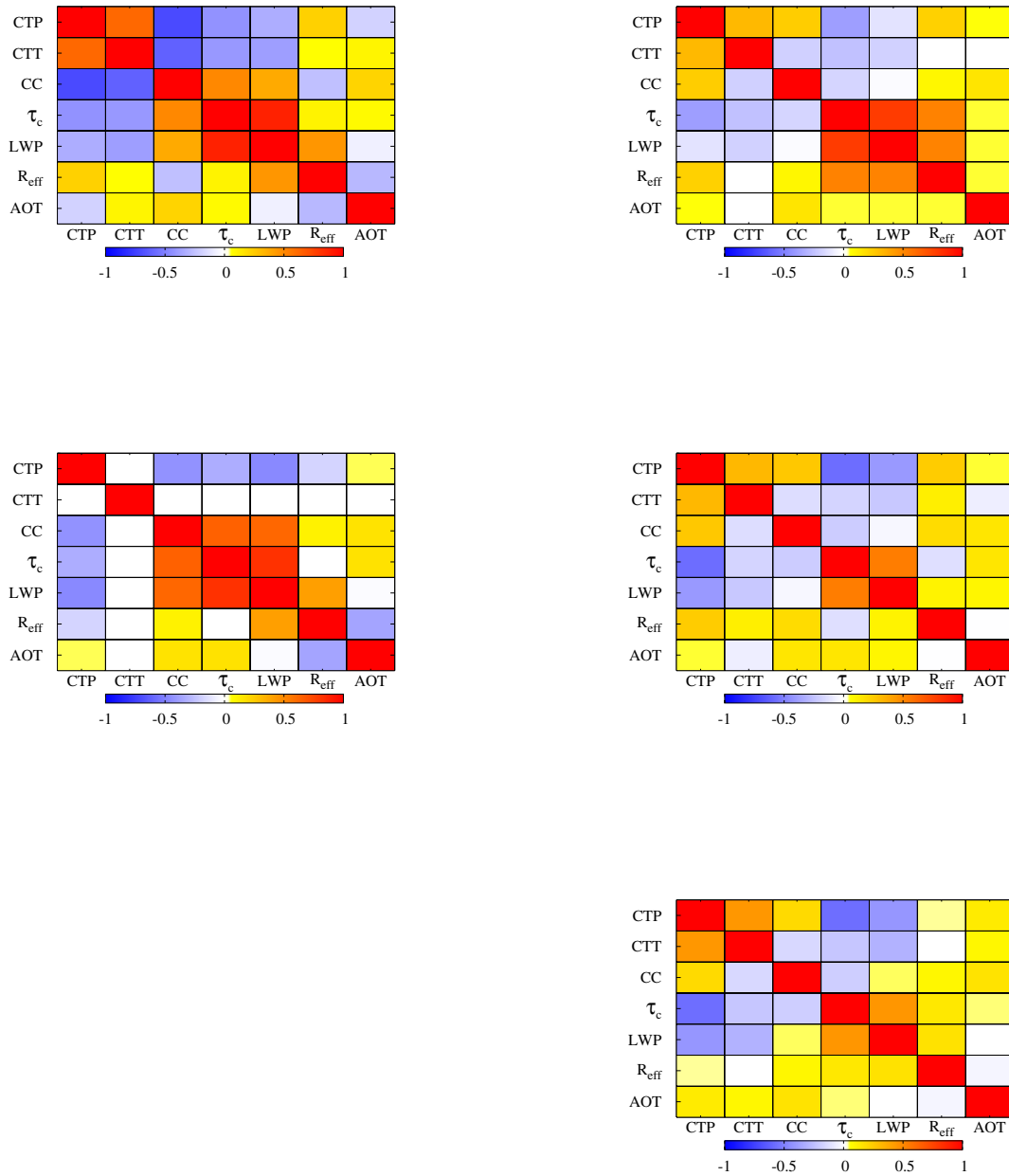


## Exp C LWP



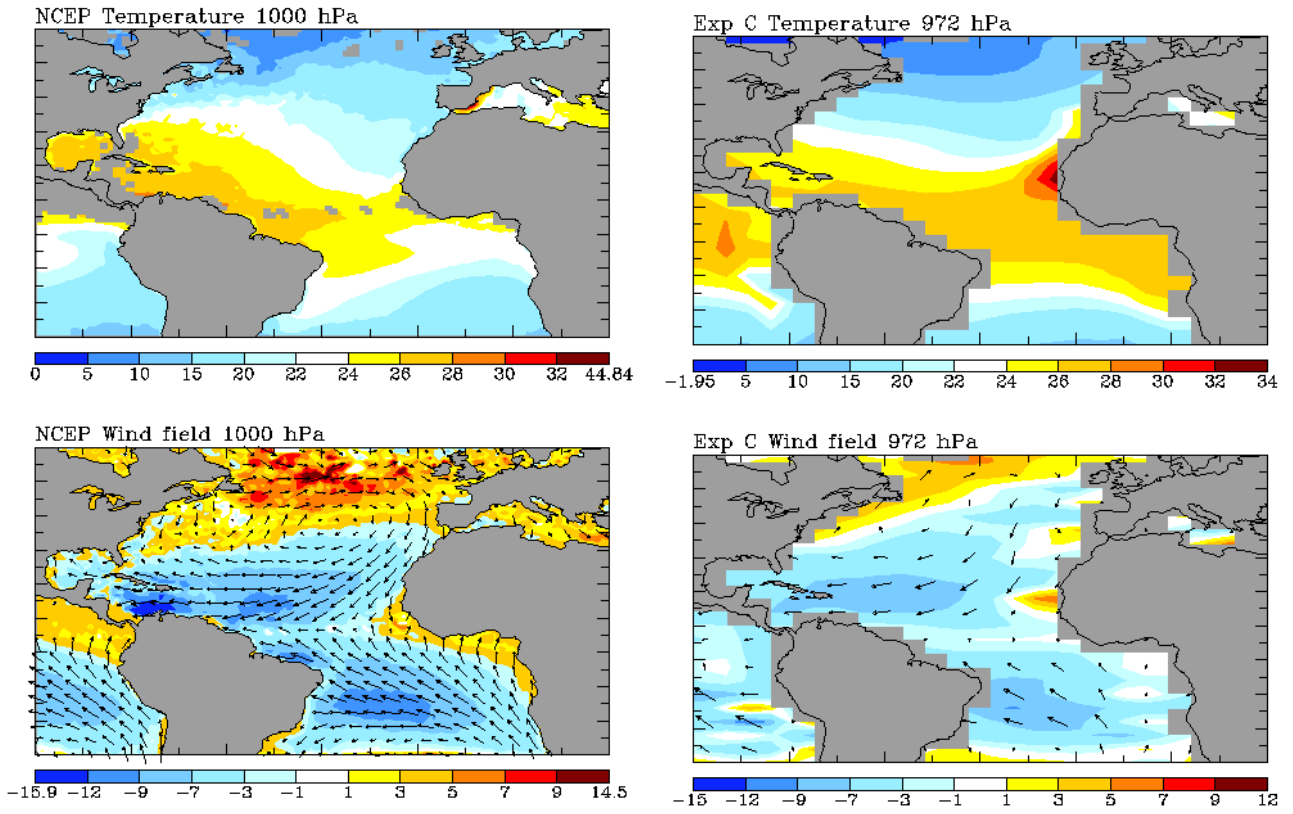
0 20 40 60 80 100 125 150 200 250 300

**Figure 5.** Liquid water path (LWP) ( $\text{gm}^{-2}$ ) for June-July-August (JJA) as obtained from MODIS (onboard Aqua), AMSR-E and as simulated by the model for Exp C.

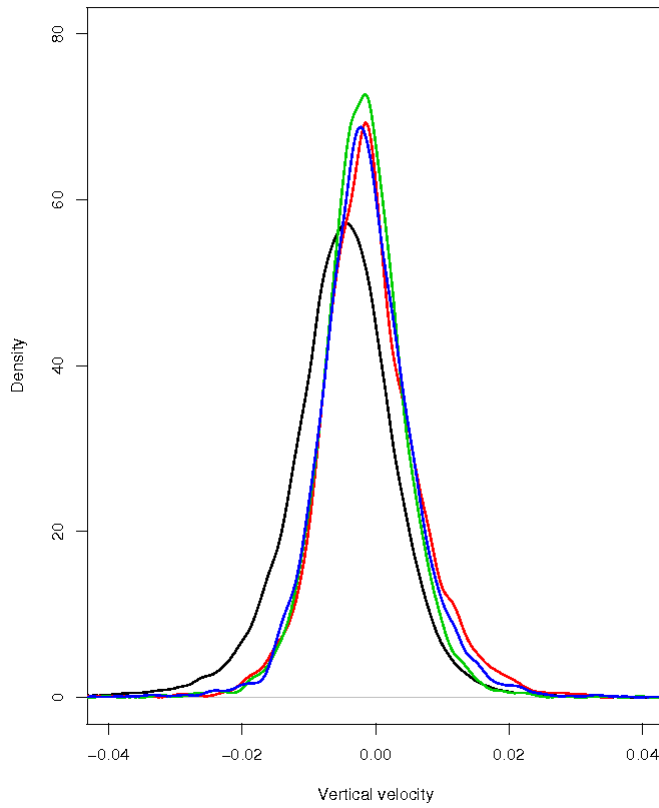


**Figure 6.** Correlation coefficients for the seven variables of interest for MODIS (top left), CERES (bottom left), Exp N (top right), Exp C (middle right) and Exp CN (bottom right). Values were significant at the 95% level for all data except for (1) Exp N: CTT-AOT, CTT- $R_{eff}$ , (2) Exp C:  $R_{eff}$ -AOT, significant at the 90% level and (3) Exp CN: LWP-AOT, CTT- $R_{eff}$ .

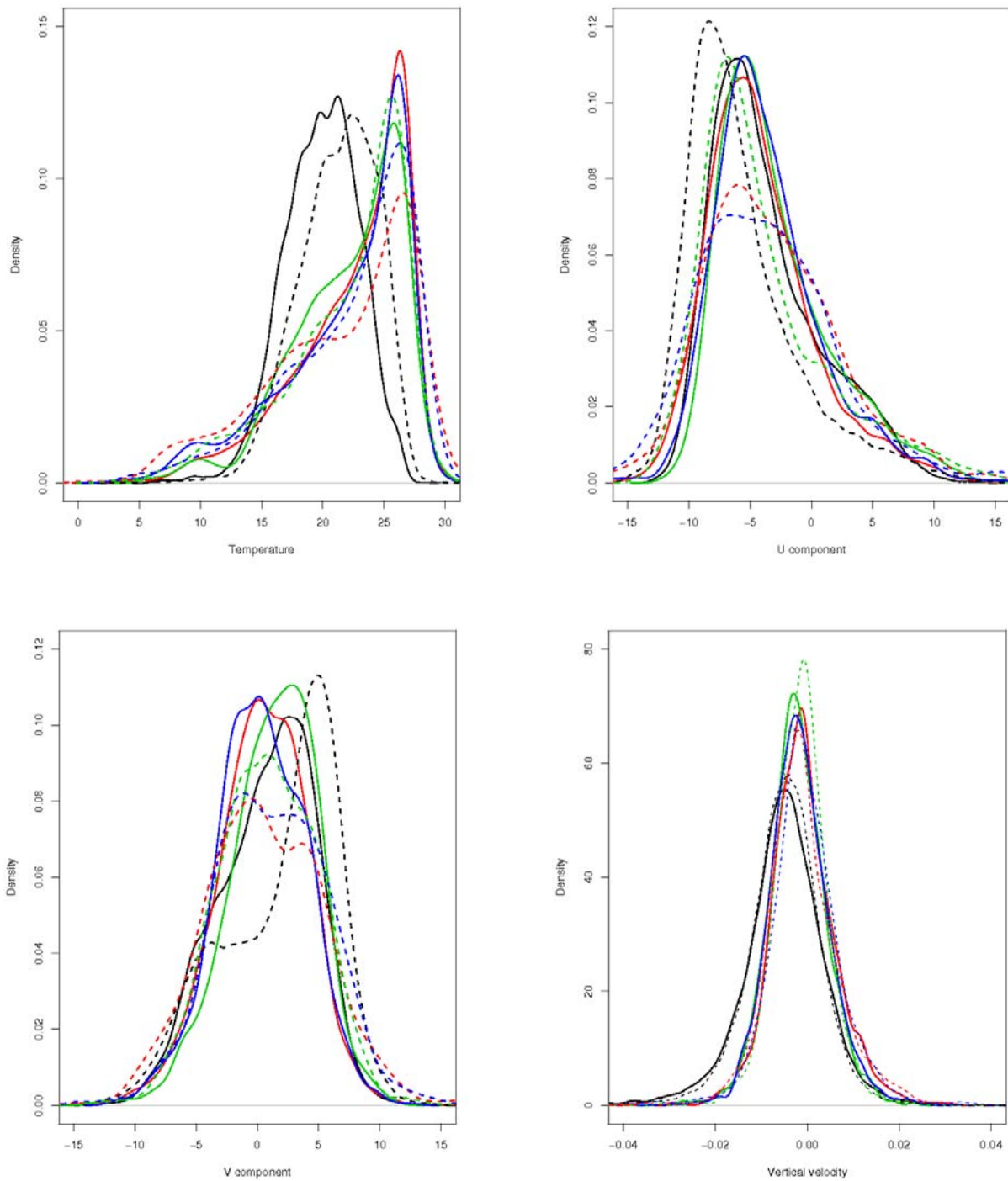




**Figure 7.** Temperature (C) and wind fields (ms<sup>-1</sup>) from NCEP and Exp C for June-July-August (JJA).



**Figure 8.** Probability density distribution of vertical velocity ( $\text{ms}^{-1}$ ) at 750 hPa for June-July-August (JJA) as obtained from reanalysis data (NCEP) (black solid line) and as simulated by the model for all three simulations: Exp N (blue), Exp C (red) and Exp CN (green). Values are positive for upward direction.



**Figure 9.** Probability density distributions for temperature (C), U and V components of winds (m/s) at 1000 hPa, and vertical velocities (m/s) at 750 hPa, for AOT < 0.06 (solid) and AOT > 0.06 (dashed) for NCEP (black), Exp N (blue), Exp C (red) and Exp CN (green) for June-July-August (JJA).