The Effect of Smoke, Dust and Pollution Aerosol on Shallow Cloud Development Over the Atlantic Ocean

Yoram J. Kaufman[1], Ilan Koren[1,2], Lorraine A. Remer[1], Daniel Rosenfeld[3] and Yinon Rudich[4]

[1] NASA Goddard Space Flight Center, Greenbelt Maryland 20771, USA,
[2] JCET, University of Maryland, Baltimore County, Baltimore, MD, USA
[3]Institute of Earth Sciences, The Hebrew University of Jerusalem, Jerusalem 91904, Israel
[4]Department of Environmental Sciences, Weizmann Institute, Rehovot 76100, Israel

ABSTRACT

Clouds developing in a polluted environment tend to have more numerous, but smaller droplets. This may lead to suppression of precipitation and longer cloud lifetime. Absorption of incoming solar radiation by aerosols, however, can reduce the cloud cover. The net aerosol effect on clouds is currently the largest uncertainty in evaluating climate forcing. Using large statistics of 1 km resolution MODIS satellite data, we study the aerosol effect on shallow water clouds, separately in 4 regions of the Atlantic Ocean, for June through August 2002: marine aerosol (30°S-20°S), smoke (20°S-5°N), mineral dust (5°N-25°N) and pollution aerosols (30°N-60°). All 4 aerosol types affect the cloud droplet size. We also find that the coverage of shallow clouds increases in all the cases by 0.2-0.4 from clean to polluted, smoky or dusty conditions. Co-variability analysis with meteorological parameters associates most of this change to aerosol, for each of the 4 regions and 3 months studied. In our opinion, there is low probability that the net aerosol effect can be explained by coincidental, unresolved changes in meteorological conditions that also accumulate aerosol, or errors in the data, though further in situ measurements and model developments is needed to fully understand the processes. The radiative effect at the top of the atmosphere incurred by the aerosol effect on the shallow clouds and solar radiation is: -11±3 W/m^2 for the 3 months studied, 2/3 of it is due to the aerosol-induced cloud changes, and 1/3 due to aerosol direct radiative effect.

INTRODUCTION

During June through August, the Atlantic Ocean is covered by varying concentrations of several aerosol types, each covering a separate latitude belt (see Fig. 1). The Southern Tropical Atlantic (30°S-20°S) is dominated by clean maritime air. The region between 20°S-5°N is a relatively well-defined region covered by smoke from biomass burning in Africa[1,2]. The Northern Tropical Atlantic (5°N-30°N) is under heavy influx of dust from Africa[3] and the Northern Atlantic (30°N-60°N) is impacted by anthropogenic pollution aerosol from North America and Europe. These aerosols absorb and reflect solar radiation to space[4], thereby affecting the regional atmospheric energy balance. Clouds that form in air laden by high aerosol concentrations tend to contain more numerous but smaller droplets that reflect sunlight, and cool the Earth[5]. The smaller cloud droplets reduce the efficiency of droplet growth by collision coalescence, which at least under some conditions[6] reduce precipitation formation and increase cloud lifetime[7,8]. However, there is a second pathway for aerosols to affect clouds; Smoke, pollution and dust aerosols absorb solar radiation, heat the atmosphere and reduce evaporation from the surface[9,10,11]. As a result, smoke over the Amazon or pollution aerosol over the Indian Ocean, can inhibit cloud formation[12-13]. This "semi-direct effect"[14,15]

was initially predicted to lead to a net global warming effect, but recent studies questioned this conclusion[16,17]. Cloud-resolving models show that absorbing aerosols located above stratiform clouds can strengthen the temperature inversion, thus increasing the moisture and liquid water content of the cloud layer[18]. Here we present new observations of yet a stronger effect of aerosols on clouds and climate, namely, a substantial increase in shallow cloud coverage due to high aerosol concentrations.

The contradictory pathways by which aerosols can affect clouds, and the large natural variability of cloud properties, represent the largest uncertainty in understanding climate change forcing. A better understanding of the effect requires large-scale systematic measurements in order to resolve the effect of aerosol on the hydrological cycle and distinguish it from natural variability. Aerosol - cloud interactions over the Atlantic Ocean and in other regions were explored in field experiments[19,20]. In the Atlantic Ocean region, these studies demonstrated connections between aerosol concentration and cloud microphysics. The experiments were intensive in aerosol and cloud physical and chemical characterizations, but limited in their spatial and temporal extents. Satellite data were used on a global scale to measure the effect of aerosol on cloud droplet size[21,22], liquid water content and cloud cover[23]. Sekiguchi et al[23] found a 0.10 increase in global cloud cover between pristine and hazy (high aerosol concentration) conditions. However, these studies used satellite data with limited spatial resolution (4-6 km) that cannot resolve smaller clouds, more susceptible to the aerosol effect. They also left open the question of whether the changes in the cloud cover are due to the aerosol effect or due to other atmospheric changes that can influence both clouds and aerosol. Here, using the new MODIS-Terra satellite data of aerosol and clouds with resolution of 1 km we analyze 3 months (June-August, 2002) of data covering millions of km^2 of shallow (stratiform and trade cumulus) clouds, and the aerosol in their immediate vicinity, and apply multiple regression to distinguish the aerosol impact on clouds from that of coincidentally changing meteorological conditions.

Shallow water clouds have a critical role in the climate system; an increase in shallow cloud cover by only 0.04 is enough to offset 2-3°K of greenhouse warming[24]. By reflecting sunlight back to space, stratiform clouds are[25,26] "the vast *climate refrigerator* of the tropics and subtropics". They are difficult to model because they are only a few hundred meters thick, capped by a strong temperature inversion and are controlled by small scale physical processes. Using state of the art satellite data we show, for the first time, that the aerosol concentration is linked to the development, microphysics and coverage of shallow clouds, thereby generating a large radiative forcing of climate.

ANALYSIS OF THE SATELLITE DATA

We use the MODIS (MODerate resolution Imaging Spectroradiometer) data on the Terra satellite to measure the daily aerosol column concentration and its correlation to the local stratiform and trade cumulus cloud cover and properties. MODIS observes detailed aerosol and cloud properties with resolution of 0.5-1 km. The data are summarized into a daily 1°x1° latitude and longitude grid. Simultaneous observations of aerosols in cloud-free regions of the grid box and clouds in the cloudy regions of the grid box are possible [see http://modis-atmos.gsfc.nasa.gov/]. Aerosol non-homogeneity has a spatial scale of 50-400 km[27], allowing the 1° resolution study. MODIS measures the aerosol optical thickness, τ (in cloud-free, sun-glint free conditions), representing the aerosol column concentration[28], that we use as a surrogate for the concentration of aaerosol that interact with

the cloud layer. MODIS also measures the following cloud properties: cloud cover, optical depth, liquid water content, cloud top effective radius and cloud top pressure[29-31].

The 1°x1° latitude and longitude data were classified as shallow water clouds if the average cloud top pressure is higher than 640 hPa and all the clouds in the given grid box and in its surrounding neighboring pixels are water clouds (no ice). The average cloud top pressure of the shallow clouds is 870 hPa, corresponding to 1200 m. For the region impacted by smoke, 53% of the 1°x1° grid boxes were classified as shallow clouds (see Table 1). This corresponds to 10 million km^2 with average of 50 daily observations during the 3 months of investigation. For the region impacted by dust it corresponds to 6 million km^2 of observations (see definition of the studied region in Table 1).

RESULTS

During June through August, smoke, dust and pollution aerosols are confined to separate latitude belts of the Atlantic Ocean (Figures 1a, 1b), allowing separate analysis of their effect, and that of pure marine air on the prevailing clouds (see Table 1). Figure 2 shows the longitudinal distribution of changes in the shallow cloud cover and in the effective radius (R_{eff}) from clean to aerosol-laden conditions. The fraction of the shallow clouds decreases from east to west due to transition from shallow to convective clouds (see Fig. 1). The largest changes in cloud cover and R_{eff} are observed in regions with high aerosol concentrations near the continental sources. The cloud liquid water path (LWP) increases in all but the biomass burning zone, in agreement with theory[7]. In the smoke zone the LWP decreases.

The satellite data show a systematic increase in the shallow cloud coverage as a function of the aerosol concentration across the Atlantic Ocean for all 4 aerosol types (see Table 1). For a given value of cloud fraction (0.30), the spatial coverage of shallow clouds extends ~2000 km further to the west for heavy smoke or dust in comparison to the clean conditions (Fig. 2). The shallow clouds also form closer to the African coast in smoke-laden conditions. Can the observed changes in the cloud cover be associated with aerosol effects?

CAUSE AND EFFECT

In Fig. 2 and Table 1 we showed the relationship between shallow cloud cover and the presence of aerosols in all 4 geographical zones analyzed separately for each of the 3 months of this study. Cloud resolving models predict an increase in stratiform cloud cover with an increase in the aerosol concentration[32]. However cloud properties also change due to variation in large-scale atmospheric circulation that may also affect aerosol concentrations. For example, low atmospheric pressures are convergence zones that tend to accumulate aerosol and water vapor and generate conditions favorable for cloud formation[33].

To untangle the effect of aerosol and large-scale meteorology on cloud properties, we use linear multiple regression. Note that the aerosol indirect effect cannot be untangled with high degree of confidence until regional models can predict cloud evolution with high precision. Here we are mainly trying to eliminate the influence of large-scale meteorological parameters that can impact simultaneously both aerosol concentration and cloud development, generating false correlation between them. The regression analyzes the dependence of the measured cloud properties (cover, droplet effective radius, and optical thickness) on: (1) MODIS measurements: aerosol optical thickness (AOT), total precipitable water vapor (indicator of convergence); (2) NCEP (National

Center for Environmental Prediction) generated meteorological fields that include air temperature at 1000 hPa, temperature difference of 850 and 1000 hPa, and 750-1000 hPa, winds at 3 altitudes (1000 hPa, 750 hPa, 500 hPa), broad scale vertical motion at 850 hPa & 500 hPa based on the continuity equation, sea surface temperature, equivalent potential temperature difference between 500 and 950 hPa[34], and low static stability, $(1/\Theta_e)(d\Theta_e/dz)$, where the differential is defined as a finite difference between 850 and 950 hPa. Logarithm of the AOT is used to reduce nonlinearity in the regression. Logarithmic dependence is expected from cloud condensation theory[35], and was found to be appropriate here. Nonlinearity in the relationships among the parameters may reduce the efficiency of the multiple regression. The analysis is used to address the following questions:

- *What is the sensitivity of the cloud cover to independent variations in meteorological and aerosol parameters?* We find that (Table 2) cloud cover is affected mainly by air temperature at 1000 hPa, temperature difference 1000-750 hPa, the aerosol optical thickness, sea surface temperature, and the winds. The influence of aerosol is similar to the influence of these meteorological parameters. This influence of meteorological parameters on MODIS clouds, as expected, shows that the NCEP data are relevant to assess simultaneous effects of synoptic meteorological variables on clouds and aerosol.

- *Can changes in the meteorological parameters increase the cloud cover while increasing the aerosol concentration?* We check the systematic change of the meteorological parameters from clean to hazy conditions. The main systematic residual is in the dust region (Table 2) by the air temperature difference 1000-750 hPa and the strength of the Easterly winds at 750 hPa. As a result, the multiple regression suggests that 70% of the change in the cloud cover between clean and dusty conditions is due to the actual dust influence. In the other regions the change in the cloud cover from clean to hazy conditions is similar to the change associated to aerosol (see Table 1).

The associated error in the net aerosol effect within the 95th percentile confidence level, based on the multiple regression is 4-8% of the effects mentioned. Errors in the meteorological parameters or nonlinearity in the effects could shift some of the dependencies of the cloud cover to aerosol, however we do not expect the errors to be more than double from the multiple regression results.

RADIATIVE FORCING

Here we compute the radiative impact resulting from the aerosol enhancement of cloud cover, and compare it with the aerosol indirect radiative effects due to the increase in cloud droplet concentration and liquid water path (LWP). Note that cloud droplet concentration is proportional to[5] $R_{eff}^{-1/3}$ for fixed LWP. Results are summarized in Table 1. The calculations are done in several sequential steps: Preparatory stage: the cloud droplet density, LWP and the cover are all scaled to the baseline clean conditions[37] of AOT=0.06 from the actual AOT in each grid box of 1°x1°. The scaling uses the multiple regression-derived dependences of these cloud properties on the AOT. Then, using the M.-D. Chou radiative transfer model[36] we calculate the reflected sunlight at the top of the atmosphere for the clean conditions. Step 1: for each grid box of 1°x1°, we replace R_{eff} and the cloud droplet density from their values for the clean conditions to the actual values, and compute the change in the reflected sunlight (column ΔN_c in Table1); Step 2: we replace the clean condition LWP with the actual value (column $\Delta N_c+\Delta LWP$ in Table1); Step 3: we replace the cloud cover with the

actual value (column $\Delta N_c+\Delta LWP+\delta cl$ in Table1); Step 4: We add the direct aerosol effect, assuming that most of the aerosol is above the shallow clouds, since both the dust and the smoke are observed to be at 3 km altitude in this period of the year[1-3]. The aerosol properties were taken from the aerosol climatology of Dubovik et al.[4]. The aerosol effect for the entire study area is shown in Fig. 3.

The results in Table 1 show that the effect of the aerosol induced change in the cloud cover generates a radiative effect of -3 to –8 W/m^2, or 3-8 times larger than the effect of aerosol induced changes in the droplet concentration and LWP. The results are comparable to the radiative effects over the Mediterranean Sea derived from field experiment data of -7 W/m^2 at the top of the atmosphere (TOA)[11]. By including aerosol direct effect on solar radiation, the total aerosol radiative effect in the north Atlantic is -8 to -14 W/m^2. This strong radiative effect is not counteracted by the thermal radiative effect due to the low altitude of the clouds. The thermal effect is < 0.2 W/m^2.

The satellite measurements are performed at 10:30 am ±30 minutes, local time. However the diurnal cycle of the shallow clouds in this region was shown to be of amplitude of 0.03 in cloud fraction, corresponding to an error in the diurnal average[38] of 7%.

The radiative effect at the surface due to the aerosol and aerosol-cloud interaction is a combination of the radiative effect at the TOA + absorption by the aerosol. We estimate the aerosol absorption using the aerosol climatology of Dubovik et al.[4]. The results, shown in Table 1, indicate that the surface radiative effect is -9 to -14 W/m^2.

Note that these large radiative effects are found for the season with highest aerosol loading at this region. Myhre et al.[39] applied similar analysis for the whole globe, and found a global aerosol indirect forcing of –1.8 W/m^2 through similar processes.

UNDERSTANDING THE PROCESSES

The satellite analysis shows that in all four geographical zones of the Atlantic Ocean, and independently for the 3 months, each with different aerosol properties and meteorology, aerosols systematically increase the shallow cloud cover. In the marine aerosol zone, in the very clean conditions, clouds have difficulties forming. Cloud resolving models that simulate[32] this condition, show an increase in stratiform cloud cover with the increase of aerosol concentration. Aerosols supply the condensation nuclei needed to form cloud droplets. Further increases in aerosol concentrations reduce the size of the droplets and delay or inhibit the formation of precipitation, increasing the cloud cover in the process.

In the smoke covered zone, the processes are more complex. Further south, Haywood et al[1] observed that the stratiform clouds are detached from the overlaying smoke layer, with a vertical separation of a few hundred meters. A recent modeling study[18] showed that under such conditions. the absorption of sunlight by the smoke alone can influence the underlying stratiform clouds even without physical interaction. Solar radiation heats the smoke layer, increasing the strength of the inversion that prevents entrainment of dry air into the stratus clouds below, and thus increases the moisture and the cloud liquid water content in the stratus deck. However not all the clouds in this latitude zone are shallow stratiform clouds. Some are trade cumulus clouds that penetrate the smoke layer at 800 hPa. Probably we observe a combination of the increase in cloud cover predicted by Johnson et al.[18], and microphysical effects in the trade cumulus as indicated by the strong reduction of the droplet effective radius. Note that we measure the total aerosol column and correlate it with the presence of low shallow clouds. Therefore we can see correlation both in case of aerosol modifying the cloud microphysics and in case of aerosol affecting the clouds through modifying the radiation field.

Ackerman et al[40] show that the inhibition of precipitation is expected to increase entrainment of air from above the clouds. If the air above the cloud is dry the entrainment may reduce the cloud water content. However over the Atlantic Ocean the humid conditions are expected to increase the cloud liquid water content[40], in agreement with our findings.

DATA QUALITY

The satellite analysis of the aerosol optical thickness was evaluated against independent ground based measurements of the AERONET sun photometers[41] for **~30** stations on islands and coast-lines around the world[42]. This validation, using 2000 points, shows that the standard error in the satellite optical thickness is $\Delta\tau=\pm0.03\pm0.05\tau$, with a bias of $\Delta\tau\sim$**0.008**. The MODIS aerosol cloud screening over the oceans is based on rigorous spatial variability of the reflectances at 0.86 μm and 1.38 μm (cirrus channel)[43]. Can residual cloud contamination still affect the data significantly?

We performed two studies to answer the question[44]. In the first we calculated the change in the average aerosol fine fraction (fraction of the optical thickness contributed by fine aerosols) between clear and hazy conditions. Cloud contamination, with its flat spectrum, would have been interpreted by the inversion as coarse aerosols. For smoke or dust the fine fraction increases with the transition from clear oceanic air to high dust or smoke concentrations, contrary to what can be expected due to cloud contamination. For the pollution zone, the fine fraction increases with τ till $\tau\leq0.3$, and decreases for higher τ.

In the second study we check, if the AERONET validation with the low bias of $\Delta\tau\sim$**0.008,** mentioned above, could have missed cases with cloud contamination or cloud illumination of the aerosol path. In the validation a point is selected if there are at least 2 AERONET measurements during one hour around the satellite overpass time and at least 5 ocean measurements out of 25 possible in a 50 km zone around the AERONET station. Does this sampling bias the validation to clear skies? We simulated 900 AERONET validations for different cloud conditions to find out. The simulation shows that for an average cloud fraction of 50%, the selected validation data set has an average cloud fraction of only 27%. Therefore, the true cloud contamination and illumination should be roughly twice the contamination observed in the validation data set, or on average doubling the bias to $\Delta\tau\sim$ **0.016**.

We also studied to what degree cloud detection can be affected by the presence of aerosol. We found it to be independent of the presence of aerosols[45] for aerosol optical thickness $\tau < 0.5$. For $\tau >$ 0.6, the aerosol fields affected the cloud classification, significantly.

The methodology to derive the cloud droplet effective radius and optical thickness is based on calculations for spatially homogeneous and smooth clouds[30,31]. In reality the cloud bumpiness and inhomogeneities result in overestimation of the effective droplet radius and underestimation of the cloud' optical thickness[46]. However these effects do not depend significantly on the presence of aerosol. Therefore the MODIS retrievals are adequate for studying the correlations between changes in the cloud cover, droplet size and cloud optical thickness and changes in the surrounding aerosol concentration.

A layer of smoke or dust above the cloud can obscure the cloud properties from the satellite observations. Haywood et al.[1,2] evaluated the cloud retrievals in the presence of African dust and smoke aerosol. They found that the MODIS droplet effective radius (using the 0.86 and the 2.1 μm channels) is not affected by overlying aerosol. We expect the cloud optical thicknesses to be accurately derived (within 10%) in the presence of dust, since dust does not absorb sunlight at 0.86 μm. However smoke can reduce the observed cloud optical thickness by 10-20%.

The point here is to acknowledge several sources of uncertainty in deriving both aerosol and cloud parameters from satellite. However, none of these sources of error can explain the systematically significant relationships we find between aerosol optical thickness and cloud fraction.

DISCUSION AND CONCLUSIONS

Three months of daily observations of clouds and aerosol over the Atlantic Ocean show independently that for each month and for each of the four regions, each dominated by a different aerosol type, aerosols have a large effect on the coverage and properties of shallow clouds. The shallow cloud cover increases systematically by 0.20-0.40 with increases in the aerosol column concentration, which is represented by increase in the optical thickness from 0.03 to 0.5. This increase in cloud cover also extends the coverage of shallow clouds thousands of kilometers west in the smoke and dust dominated regions. The changes are accompanied with reduction in cloud droplet size by 10-30%. In most of the regions (all but the smoke region) the liquid water content increases as well. All these observations are in agreement with the suggestion that inhibition of precipitation by aerosol plays a critical role in the formation, cover, spatial extent and properties of Atlantic shallow clouds. Multiple regression analysis associated most of the increase in the cloud cover with increase in the presence of aerosol and only a small part to changing large-scale meteorological conditions. However the large scale and linear nature of the multiple regression analysis leaves uncertainties in the cause and effect that can be resolved with further development of regional cloud resolving models. The 95th percentile confidence limit on the aerosol effect on cloud fraction is only 4-8% lower than the stated values.

The systematic influence of high aerosol concentrations on clouds generates large radiative effects over the Atlantic Ocean that may regionally counteract the greenhouse warming. Here we can expect that the non-marine aerosol have doubled in the last 50-100 years, due to expansion of population and economic activity by factor of 3, and a doubling in Saharan dust production[47]. The smoke and pollution effect on cloud cover generates a radiative forcing at the top of atmosphere which is about half of today's aerosol effect or -6 W/m^2, and reduction of sunlight available for evaporation from the ocean by 7 W/m^2, thus dominating climate change in this region. This is in line with the observation of a global dimming of sunlight at the surface over the land in the last 50 years[48] of 10-20W/m^2, taking into account the higher aerosol concentrations over the land, near the sources than over the ocean[1,49]. Recent papers show that the dimming effect reversed in the mid 1980s and a brightening resumed[50,51]. This reversal is also associated with similar reversal in cloud-free transmission of sunlight in Europe and Japan, which is a measure of the aerosol optical thickness. A rough estimate based on Wild et al[51] figure S4 gives an increase till the mid 1980s and decrease till 2000 of AOT of 0.01-0.02 per decade respectively. Scaling the radiative effects in Table 1 to these changes in AOT gives radiative effects of 2-3 W/m^2 in agreement with the measurements[48-51]. This suggests that the aerosol indirect effect and in particular the increase of cloud cover can serve as a possible explanation to the observed changes in surface illumination.

The forcing observed by aerosol induced increase in cloud coverage exceeds that due to aerosol induced changes in cloud drop concentrations alone by a factor 3 to 5. These findings clearly demonstrate that traditional estimates of aerosol-cloud forcing, which focused on cloud top brightness may be inadequate and severely underestimate the aerosol climatic effects.

The aerosol inhibition of precipitation derived indirectly in this study, and the drastic influence, in particular of smoke and dust on the shallow stratiform and trade cumulus clouds, spatial cover and radiative forcing, leaves open the question to what extent these aerosols control the circulation and climate of the Atlantic Ocean. The influence can be expected to be significantly large.

Acknowledgement

This work was supported by NASA, the Israeli Space Agency, and Technology and Israel Science Foundation.

REFERENCES

1. Haywood JM, Osborne SR, Francis PN, et al., *J. Geophys. Res.,* 108 (D13): Art. No. 8473 FEB 18 2003;
2. Haywood JM, Osborne SR, Abel SJ, *J. Royal Meteor. Soc.* 130 (598): 779-800 Part A APR 2004
3. Prospero, J. M., and T. N. Carlson, *J. Geophys. Res.*, *77* , 5255-5265, 1972.
4. Dubovik, O., B.N. Holben, T. F. Eck, et al, *J. Atmos. Sci,* 59, 590-608, 2002
5. Twomey, S., Piepgrass, M. & Wolfe, T.L., *Tellus* 36B, 356-366, 1984
6. Warner, J., J. Appl. Meteor., 7, 247-251, 1968.
7. Albrecht B. A., Aerosols, *Science* 245, 1227-1230 (1989);
8. Rosenfeld, D., *Science* 287, 1793-1796, 2000
9. Ramanathan, V. *et al*., *J. Geophys. Res.* 106, 28371-28398, 2001;
10. Ramanathan V., et al., , *Science*, 294, 2119-2124, 2001
11. Lelieveld J, Berresheim H, Borrmann S, et al., Science 298 (5594): 794-799 Oct. 25 2002
12. Ackerman, A.S. Toon OB, Stevens DE, *et al*., *Science* 288, 1042-1047 (2000);
13. Koren, I., Y. J. Kaufman L. A. Remer and J.V. Martins, *Science*, 303, 1342-1344, 2004
14. Hansen, J., Sato, M. & Ruedy, R., *J. Geophys. Res.* 102, 6831-6864 (1997);
15. Jacobson, M. Z., *Nature* 409, 695-697, 2001
16. Penner JE, Zhang SY, Chuang CC, *J. Geophys. Res.,* 108 (D21): Art. No. 4657 Nov. 1 2003;
17. Lohmann U, Feichter J., *Geophys. Res. Lett.,* 28 (1): 159-161 JAN 1 2001
18. Johnson, B.T., Quart. *J. Royal Meteor. Soc.* 130, 1407-1422, doi:10.1256/QJ.03.61, 2004
19. Bates T. S., et al., *J. Geophys. Res.* 103, 16297-16318 (1998);
20. Raes F, Bates T, McGovern F, et al., *Tellus-B* 52 (2): 111-125 APR 2000
21. Nakajima, T., A. Higurashi, K. Kawamoto, and J. E. Penner, *Geophys. Res. Lett.*, 28, 1171–1174, 2001
22. Bréon, F.-M., D. Tanré & S. Generoso, *Science*, 295, 834-838, 2002
23. Sekiguchi, M., T. Nakajima, K. Suzuki, et al., *J. Geophys. Res*, 108, NO. D22, 4699, doi:10.1029/2002JD003359, 2003
24. Randall, D. A., J. Coakley, C. Fairall, et al., *Bull. Amer. Meteor. Soc.*, 65, 1290 – 1301, 1984.
25. Bretherton, C.S., T. Uttal, C.W. Fairall et al., *B.Amer. Meteor. Soc.,* July, Doi: 10.1175/BAMS-85-7-967, 2004;
26. Klein, S.A. and D.L. Hartmann, *J. Climate, 6,* 1587-1606, 1993.
27. Anderson T.L., Charlson RJ, Winker DM, et al., *J. Atmos. Scie.* 60 (1): 119-136 JAN 2004
28. Tanré, D., Y. J. Kaufman, M. Herman and S. Mattoo, *JGR-Atmospheres*, 102, 16971-16988, 1997.
29. Ackerman, S. A., K. I. Strabala, W. P. Menzel, et al., *J. Geophys. Res.*, *103*, 32141-32157, 1998;

30. King MD, Menzel WP, Kaufman YJ, et al., *IEEE TGRS* 41 (2): 442-458 FEB 2003;
31. Platnick S, King MD, Ackerman SA, et al., *IEEE TGRS* 41 (2): 459-473 Feb. 2003
32. Ackerman A.S, Toon OB, Stevens D.E, et al., *Geophys, Res. Lett.,* 30 (7): Art. No. 1381 APR 5 2003
33. Chou, M-D, Chan PK, Wang MH*, J. Atmos. Scie*. 59 (3): 748-757 2002
34. Matsui T, Masunaga H, Pielke RA, et al., *Geophys. Res. Lett*., 31 (6): Art. No. L06109, 2004
35. Feingold, G., L.A. Remer, J. Ramaprasad and Y. J. Kaufman, *J. Geophys. Res*., 106, 22907-22922, 2001
36. Chou, M-D., *J. Atmos. Sci*. 49, 762, 1992.
37. Kaufman, Y. J., A. Smirnov, B. N. Holben and O. Dubovik, *Geophys. Res. Lett*., 28, 3251-3254, 2001
38. Rozendaal, M.A., C.B. Leovy and S.A. Klein, J. of Climate, 8 (7): 1795-1809, 1995
39. Myhre, G., F. Stordal, M. Johnsrud, et al., submitted to Nature.
40. Ackerman, A.S., M.P. Kirkpatrick, D.E. Stevens and O.B. Toon, Nature, 432, 1014-1017, 2004.
41. Holben, B. N., T. F. Eck, I. Slutsker, et al., Rem. Sens. Environ., **66**, 1-16, 1998.
42. Remer, L. A., D. Tanré, Y. J. Kaufman et al., *Geoph. Res. Lett*., 29 (12): art. no. 1618 JUN 15 2002;
43. Martins, J. V., D. Tanré, L.A. Remer, et al., Geophys. Res. Lett., 29(12),10.1029/2001GL013252, 2002.
44. Kaufman Y.J., L. A. Remer, D. Tanré, et al., A critical examination of the residual cloud contamination and diurnal sampling effects on MODIS estimates of aerosol over ocean, submitted to IEEE TGRS44.
45. Brennan, J.I. Y. J. Kaufman, R.-R. Li, and I. Koren, submitted to *IEEE TGRS*, 2004
46. Várnai T. and A. Marshak, *J. Atmos. Sci*., 59,1607-1618, 2002
47. Prospero JM, Lamb PJ, *Science* 302 (5647): 1024-1027 NOV 7 2003
48. Stanhill, G. S. Cohen, *Agric. and Forest Metor*. 107 (4): 255-278 APR 19 2001.
49. Ichoku, C., L. A. Remer, Y. J. Kaufman, et al., *J. Geophys. Res.,* 108 (D13): art. no. 8499 APR 12 2003
50. Pinker R.T., Zhang B., Dutton E.G., Science, 308 (5723): 850-854 MAY 6 2005
51. Wild M., Gilgen H., Roesch A., et al., Science 308 (5723): 847-850 MAY 6 2005

Table 1: Results of the analysis for 4 regions in the Atlantic Ocean. Columns from left: location; fraction of the region classified as shallow clouds; shallow cloud fraction; range of the aerosol optical thickness in the analysis (5^{th} - clean and 95^{th} - hazy percentile) & <u>average value</u>; Δcl-aer - change in the cloud cover from the clean and hazy conditions; δcl-aer - partial change in the cloud cover associated with aerosol by the multiple regression; % change in the cloud effective radius (R_{eff}) from the clean and hazy conditions; % change in the cloud liquid water content (LWP); change in the cloud top pressure. For each value the variability among the 3 months of analysis (June-August) is given. The average radiative effects due to change in the aerosol optical thickness from the base oceanic value of 0.06 associated with: increase in cloud droplet concentration (ΔN_c) due to reduction in R_{eff}; + change in the column cloud water content (ΔLWP); + change in the cloud cover (δcl); + direct aerosol radiative effect; ΔAbs - absorption of sunlight by aerosol. Note that the sum of the last 2 columns is the total aerosol radiative forcing at the surface. The radiative effect was calculated as 1/2 of the effect for solar zenith angle of 60° only for 1° latitude grid boxes characterized as shallow clouds. The uncertainty in the aerosol measurements from MODIS is ~10%, cloud fraction ~ 3% and cloud effective radius 20%. The 95^{th} percentile confidence limit of the multiple regression is 4-8% off the stated values. The overall error in the radiative effects calculations is therefore ~20%. Absorption computations depend on the validity of the assumed single scattering albedo with uncertainty of 50%. No calculations are given for the marine region, since the average AOT is too close to the baseline value.

Region and Dominant aerosol	Fract. of region	Shallow cloud cover	Range of aerosol optical thickness / <u>average</u>	Δcl-aer	δcl-aer	% change in R_{eff}	% change in LWP	change in CLTP hPa	Radiative effects (W/m^2) due to:				
									ΔN_c	ΔN_c+ Δ LWP	ΔN_c+ ΔLWP + δcl	Total forcing TOA	ΔAbs
30°N-60°N Pollution	0.17	0.07	0.03-0.19 <u>0.102</u>	0.20 ±0.06	0.19 ±0.03	-12 ±10	6 ±34	-39 ±20	-1.0	-1.1	-4.5	-8.0	0.7
5°~-30°N Saharan Dust	0.26	0.11	0.03-0.46 <u>0.174</u>	0.36 ±0.12	0.25 0.04	-12 ±13	9 ±34	-66 ±13	-0.7	-0.9	-6.8	-14.0	0.7
20°S-5°N biomass burning	0.53	0.29	0.03-0.43 <u>0.152</u>	0.31 ±0.07	0.31 ±0.04	-32 ±3	-21 ±8	-55 ±11	-1.5	-1.0	-9.5	-11.3	2.9
30°S-20°S Marine	0.47	0.27	0.02-0.24 <u>0.085</u>	0.45 ±0.10	0. 45 ±0.04	-19 ±7	35 ±22	-72 ±18	---	---	---	---	---

Table 2: Multiple regression analysis of the influence of meteorological parameters and dust optical thickness on the cloud fraction analyzed in the 5°-30°N region of the Atlantic Ocean. The analysis was carried for 3 months independently – June through August, and the table shows the average and variability among the 3 months. The parameters influencing the cloud fraction are ordered by order of importance based on the correlation with the cloud cover (second column). The next 2 columns give the correlation of the parameter with the aerosol optical thickness (AOT) and the change in the cloud fraction associated by the multiple regression with changes in the meteorological parameters or AOT from the 5th to 95th percentile average values. Note that while the AOT is one among 7 parameters affecting the cloud fraction, it is by factor 5 the dominant parameter affecting the change in the cloud fraction from clean (AOT=0.03) to hazy (AOT=0.40) conditions. In the smoke, pollution or marine regions the aerosol effects where even stronger. The main meteorological parameters that affected the difference in the cloud cover from clean to hazy conditions are the air temperature at 1000 hPa, the difference in the temperature between 1000 and 750 hPa and the Easterly wind speed at 750 hPa.

Parameter	correlation to cloud fraction	correlation to dust **AOT**	Change in cloud fraction clean to hazy
Temperature at 1000 hPa	**-0.32±0.09**	**0.22**	**-0.04±0.08**
Temperature difference, 1000-750 hPa	**-0.31±0.27**	-0.13	**0.05±0.07**
Ln(AOT)	**0.29±0.14**	0.97	**0.25±0.03**
Sea Surface Tempe00rature	**-0.28±0.09**	-0.04	0.00±0.03
Northern wind at 1000 hPa	**-0.28±0.26**	-0.17	0.01±0.01
Temperature difference, 1000-850 hPa	**-0.21±0.15**	-0.17	-0.01±0.01
Difference in potential temp at 500-950 hPa	**0.19±0.07**	-0.06	0.01±0.01
Low Static Stability at 850 and 950 hPa	-0.17±0.05	0.04	-0.01±0.02
Eastern wind at 1000 hPa	-0.10±0.08	**-0.20**	0.00±0.01
Northern wind at 750 hPa	-0.09±0.11	0.02	0.00±0.00
Eastern wind at 750 hPa	-0.08±0.07	**-0.24**	**0.06±0.02**
Total Column Precipitable Water Vapor	-0.05±0.08	**0.20**	0.01±0.01
Absolute Vorticity at 1000 hPa	0.04±0.06	-0.14	0.00±0.00
Eastern wind at 500 hPa	-0.02±0.05	**-0.29**	-0.01±0.03

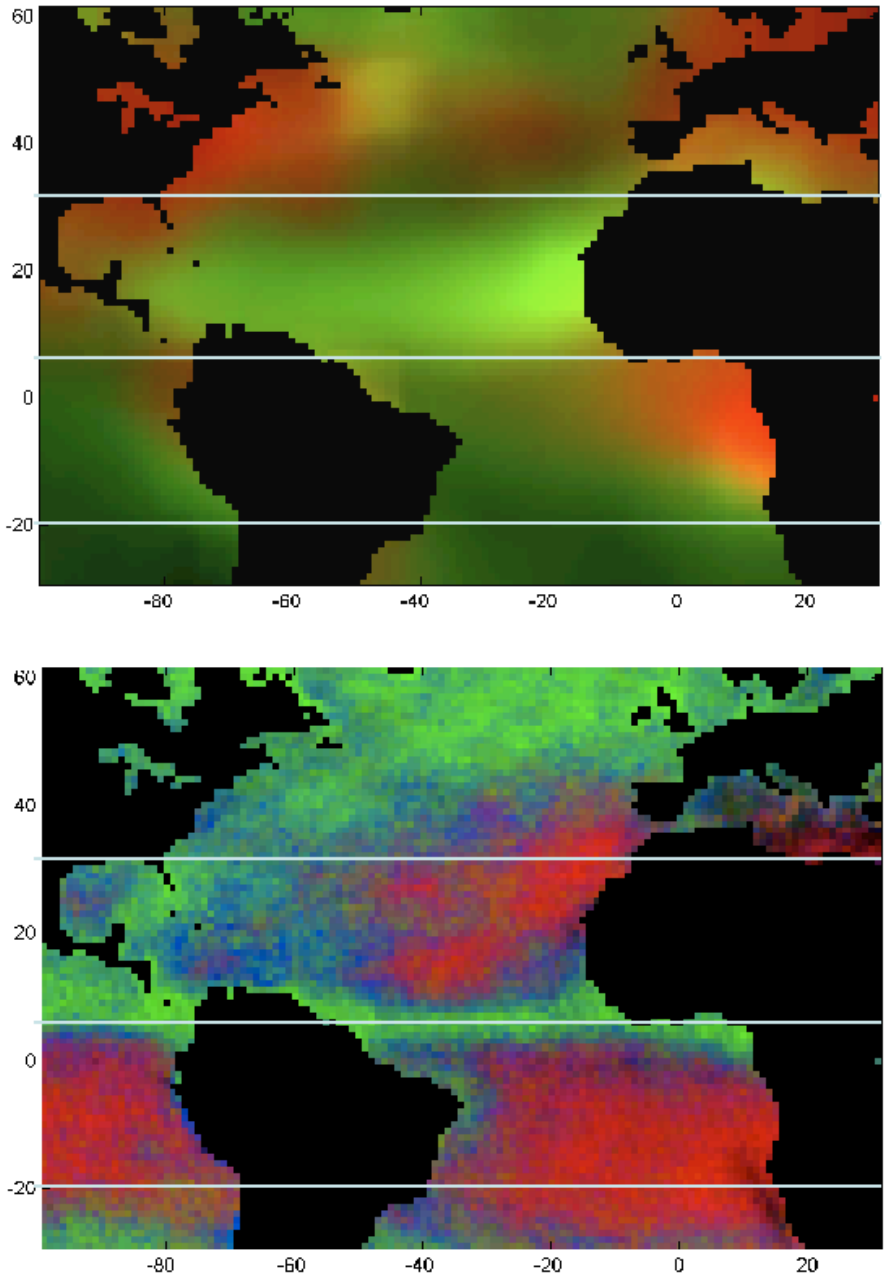

Fig 1: Spatial distribution of aerosol and clouds over the Atlantic Ocean from MODIS data for June-August 2002.

a) Spatial distribution of the aerosol column concentration (expressed as the optical thickness) and type (given by the fraction of the aerosol in the sub-micron mode) over the Atlantic Ocean for the June-Aug period. The optical thickness is represented by the brightness of the image, the aerosol type by the color, red - dominance by submicron particles - smoke from central Africa and pollution from Europe and North America, and green – dominance by dust from Africa or sea salt in regions with high winds.

Fig 1 (b) Spatial distribution of shallow (red), deep convective (green) and mixed (blue) cloud cover on a 1°x1° longitude x latitude grid. Black - continental regions. The data are averaged for June-August 2002.

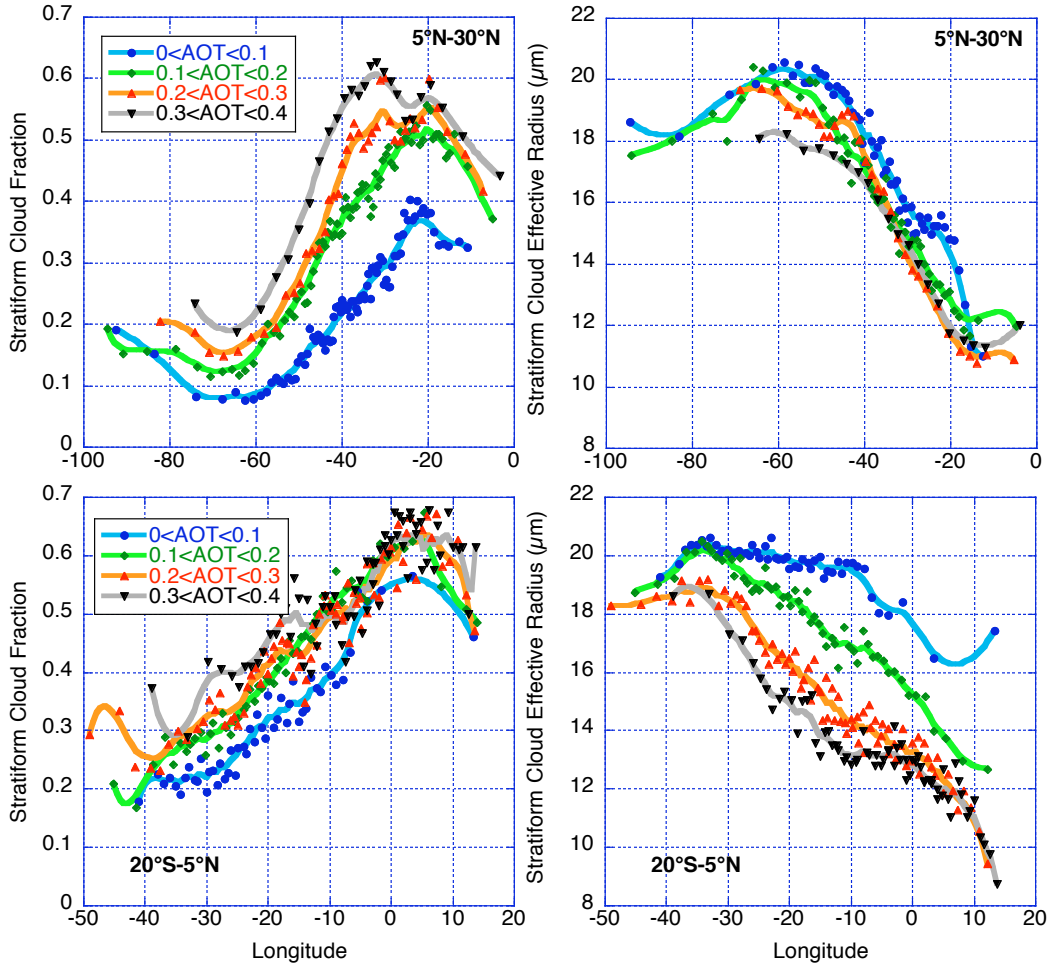

Fig. 2: Longitudinal dependence of the shallow cloud fraction (left panels) and droplet effective radius (right panels) for the Northern tropical Atlantic with dust intrusions (top panels) and Southern tropical Atlantic with smoke intrusion (bottom panels). The results are shows for 4 ranges of the aerosol optical thickness (AOT) given in the figure. The dots are average of 50-200 1°x1° grid boxes located in similar longitude location and for the same aerosol optical thickness range.

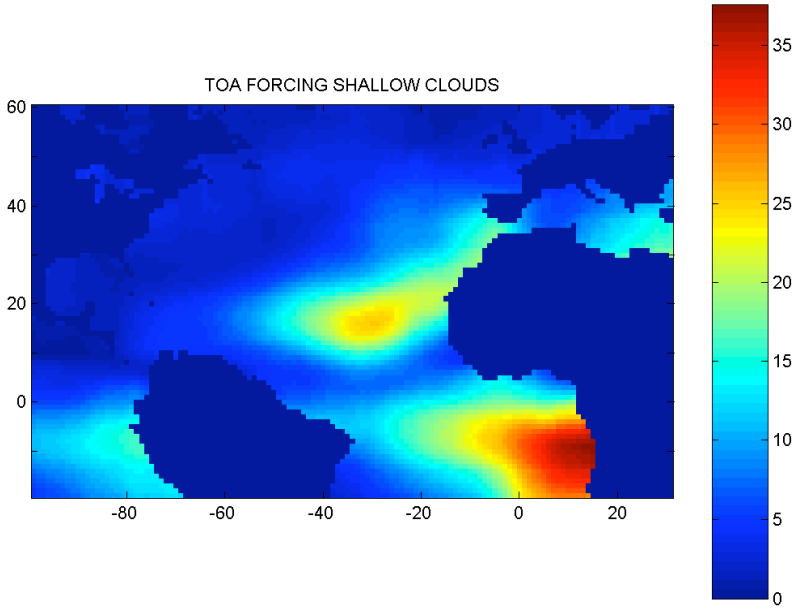

Fig. 3: The aerosol reflected solar flux at the top of the atmosphere due to combined effect on cloud cover and microphysics and on reflection of solar radiation. The radiative effect is calculated as the difference of MODIS observations from the conditions for baseline aerosol with optical thickness of 0.06. The results are weighted by the frequency of detection of shallow clouds in the 1° latitude x longitude daily grid boxes. The color bar shows the values in W/m^2