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Effect of coarse marine aerosols on stratocumulus clouds

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[1] In contrast to fine anthropogenic aerosols (radii ~<0.5 μm), large aerosol particles are thought to enhance cloud droplet growth, promote precipitation formation and reduce cloud albedo. While shown in cloud simulation models, the impact of coarse aerosols on marine stratocumulus clouds lacks observational evidence. Here, by combining data from AMSR-E and MODIS, both aboard NASA's satellite Aqua, we link the amount of coarse marine aerosols emitted to the atmosphere through wind-driven processes with the size of cloud droplets, at the world's largest deck of marine stratocumulus clouds over the southeastern Pacific. For constrained meteorological conditions, approximately 1/2 of the change in droplet effective radius (r_{eff}) is attributed to increase in coarse marine aerosol optical depth (τ_{cm}), as surface winds intensify. Accordingly, a two-fold increase in τ_{cm} is associated with a 1.4 μm +/−0.11 increase in r_{eff}. Our results suggest that climatic changes in surface winds may play an important role not only over land for wind-power estimation but also over the oceans by changing clouds reflectance and lifetime. **Citation:** Lehahn, Y., I. Koren, O. Altaratz, and A. B. Kostinski (2011), Effect of coarse marine aerosols on stratocumulus clouds, *Geophys. Res. Lett.*, *38*, L20804, doi:10.1029/2011GL048504.

1. Introduction

[2] Forming large decks over the eastern part of subtropical oceans, marine stratocumulus clouds (Figure 1a) are important constituents of the climate system. By increasing the earth's shortwave albedo compared with the underlying ocean, while radiating in the longwave at approximately the same temperature as the surface, these shallow marine boundary clouds have a significant cooling contribution to the Earth's radiative balance [*Hartmann and Doelling*, 1991]. Marine stratocumulus clouds have two stable formations: open and closed cells [*Krueger and Fritz*, 1961]. The dynamics of the two formations and the roles of precipitation and aerosols in these processes are subject to extensive observational and modeling research [e.g., *Wood et al.*, 2008; *Feingold et al.*, 2010].

[3] The radiative impact of marine stratocumulus is susceptible to perturbations by maritime and terrestrial aerosols that modulate cloud microphysical and optical properties. The outcome of different cloud-aerosol interactions strongly depends on the size of aerosol particles involved in the process [*Dusek et al.*, 2006]. When focusing on the optical effects, increase in fine aerosol loading, usually from anthropogenic sources, results in more numerous and smaller droplets competing for the same amount of available water vapor. When other factors being held constant, this enhances the cloud reflectivity (the first aerosol indirect effect) [*Twomey*, 1974] and, by reducing the warm rain formation efficiency, increases the cloud lifetime (the second aerosols indirect effect) [*Albrecht*, 1989]. The impact of increased fine aerosol concentration on cloud droplets size have been explored using both regional and global satellite datasets [e.g., *Breon et al.*, 2002; *Kaufman et al.*, 2005].

[4] In contrast to the indirect cooling effect by small anthropogenic aerosols, it was suggested that large aerosol particles, acting as giant cloud condensation nuclei (GCCN), may exert an opposite effect of *reducing* cloud albedo by *enhancement* of cloud droplet growth and *promotion* of precipitation formation through more efficient collision coalescence process [*Johnson*, 1982; *Feingold et al.*, 1999; *Rosenfeld et al.*, 2002; *Posselt and Lohmann*, 2008]. There are various definitions in the literature for GCCN. In this work we will refer to coarse marine aerosols (CMA, radii >0.5 μm) that are emitted from the surface of the ocean through wind driven processes. The population of wind-induced CMA in the marine boundary layer is dominated by soluble sea salt particles whose size distribution, concentration and optical properties strongly depend on surface wind speed (W), with *stronger* winds promoting *more* emission of *larger* particles [*Lewis and Schwartz*, 2004].

[5] While explored in various modeling studies [e.g., *Feingold et al.*, 1999; *Posselt and Lohmann*, 2008], satellite observations of CMA effect on shallow marine clouds are rare [*Yuan et al.*, 2008; *L'Ecuyer et al.*, 2009] and do not provide sufficient quantitative information on the expected change in cloud microphysical properties.

[6] In this work we make use of satellite data to quantify the link between wind induced CMA and the size of marine stratocumulus cloud droplets, near the top of marine stratocumulus clouds at the largest and most persistent subtropical marine stratocumulus deck found over the Southeast Pacific [*Klein and Hartmann*, 1993], off the coast of Chile and Peru. The Southeastern Pacific stratocumulus cloud field (Figure 1), was recently subject to extensive study in the framework of the VOCALS project [*Wood et al.*, 2011].

2. Data and Methods

[7] A major reason for the lack of observational evidence to the coarse marine aerosol effect is the inherent difficulty of acquiring simultaneous satellite measurements of the two components (i.e. CMA and cloud microphysical properties) in densely covered decks of marine stratocumulus. To overcome this obstacle we use satellite measurements of surface wind speed (W) as a proxy to coarse marine aerosol optical

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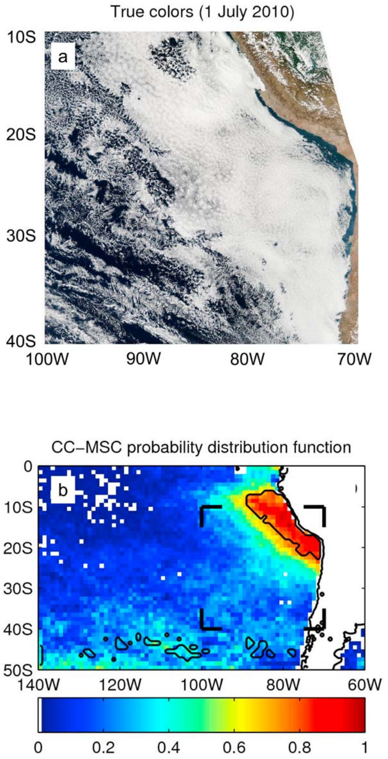

Figure 1. (a) True color MODIS-Aqua image showing a dense closed cells marine stratocumulus (CC-MSC) deck. The image is taken from the MODIS rapid response system (http://rapidfire.sci.gsfc.nasa.gov/). (b) Probability distribution function of CC-MSC occurrence during June–August 2008. Data points are classified as CC-MSC if they are associated with low (CTT > 280 K) and dense (CF > 0.9) clouds. The black frame denotes the area covered by the true color image in Figure 1a. The black contour marks the area where the probability of occurrence is higher than 0.7, which defines the regions of interest for Figures 2 and 3.

depth (τ_{cm}), which represent the amount of coarse marine particles found in the atmosphere [*Lehahn et al.*, 2010]. Since radiometric measurements of W are not affected by clouds, this concept enables "observation" of CMA in locations otherwise obscured by clouds.

[8] Atmospheric and oceanic parameters throughout a 5 years period (2004–2008) were obtained from two sensors aboard NASA's satellite Aqua: the Advanced Microwave Scanning Radiometer NASA's - Earth Observing System (AMSR-E) and the Moderate Resolution Imaging Spectroradiometer (MODIS), both providing daily global observations of the ocean and atmosphere. The MODIS dataset consists of daily, 1°, level 3 datasets of $r_{\mathrm{eff},}$ cloud fraction, and cloud top tmp.

[9] AMSR-E data, which is obtained from Remote Sensing Systems (RSS), Inc. (http://www.remss.com), is used for extracting daily, 0.25°, observations of liquid water path (LWP), sea surface temperature (SST) and surface wind speed. Based on the wind speed we estimate coarse marine aerosol optical depth, τ_{cm}, using the equation recently derived by *Lehahn et al.* [2010]:

$$\tau_{\mathrm{cm}} = 0.009 \times (\mathrm{W} - 4) + 0.03$$

with W corresponding to surface wind speed (independently of wind direction) higher than 4 m/s. The suggested algorithm, which is based on comparison between satellite retrievals of W and aerosols over 6 tropical and subtropical oceanic regimes, was validated against ground data from several AERONET stations and is in agreement with other studies linking surface wind speed and aerosol optical depth.

3. Aerosols or Meteorology?

[10] Clouds depend on both the local meteorological conditions and aerosol properties. In most cases it is hard to decouple the effect of aerosols on clouds from changes in other environmental conditions. In our case, stronger surface wind that enhances marine coarse particles formation might be an indication for less stable meteorological conditions. Recent study on shallow cumulus clouds showed a link between wind speed and precipitation, attributing it to boundary layer humidity while acknowledging the possible effect of sea salt aerosols [*Nuijens et al.*, 2009].

[11] In order to estimate the effect of aerosols on clouds, one has to constrain the meteorological variance and examine clouds and aerosols for a given meteorological state. Consistent trends for different meteorological states may suggest an aerosol effect, independently of meteorological influence [*Koren et al.*, 2010]. Here, to isolate the coarse aerosol effect, we stratify the analysis to similar environmental conditions and similar macrophysical cloud properties. The first step in that direction is removing the impact of annual cycle by analyzing a 3 month period during the austral winter (June–August). Secondly, we try to avoid variations associated with changes in cloud field morphology by focusing on decks of Closed Cells marine stratocumulus (CC-MSC) as the one seen in Figure 1a. A preliminary classification is performed based on the cloud top height (represented by cloud top temperature, CTT) and spatial density (cloud fraction, CF), with pixels identified as being dominated by CC-MSC, when they are both shallow (CTT > 280 K) and dense (CF > 0.9) clouds. Based on this preliminary classification, a region of interest (ROI) is defined as the area of highest probability (P > 0.7) for having CC-MSC throughout the 3 months period (black contour in Figure 1b). Following that we further reduce meteorological and macrophysical variance, by restricting the analysis to narrow ranges of clout top temperature (281–280 K), cloud fraction (0.9–1) and temperature difference between cloud top and the sea surface (ΔT, 8–12 K). In addition, the analysis is stratified to data associated with three bands of LWP (25–50, 50–75 and 75–100 gr/m^2), which, in marine stratocumulus, is a parameter that captures much of the cloud sensitivity to environmental conditions not related to aerosol effects [*Korolev*, 1993; *Pawlowska and Brenguier*, 2003]. Similarly to CMA, LWP is positively correlated with W via stronger evaporation and thicker clouds. On the other hand, one expects the liquid water path to correlate negatively with the entrainment speed, as the dry air overlying the cloud dilutes cloudy air [*Stevens*, 2006; *Wood*, 2007]. Since surface and entrainment speeds affect LWP, as a first approximation, slicing the data to bins of equal LWP accounts for both effects, while minimizing meteorological variance.

4. Results and Discussion

[12] The link between coarse marine aerosols and southeast Pacific stratocumulus cloud droplet size is clearly seen in

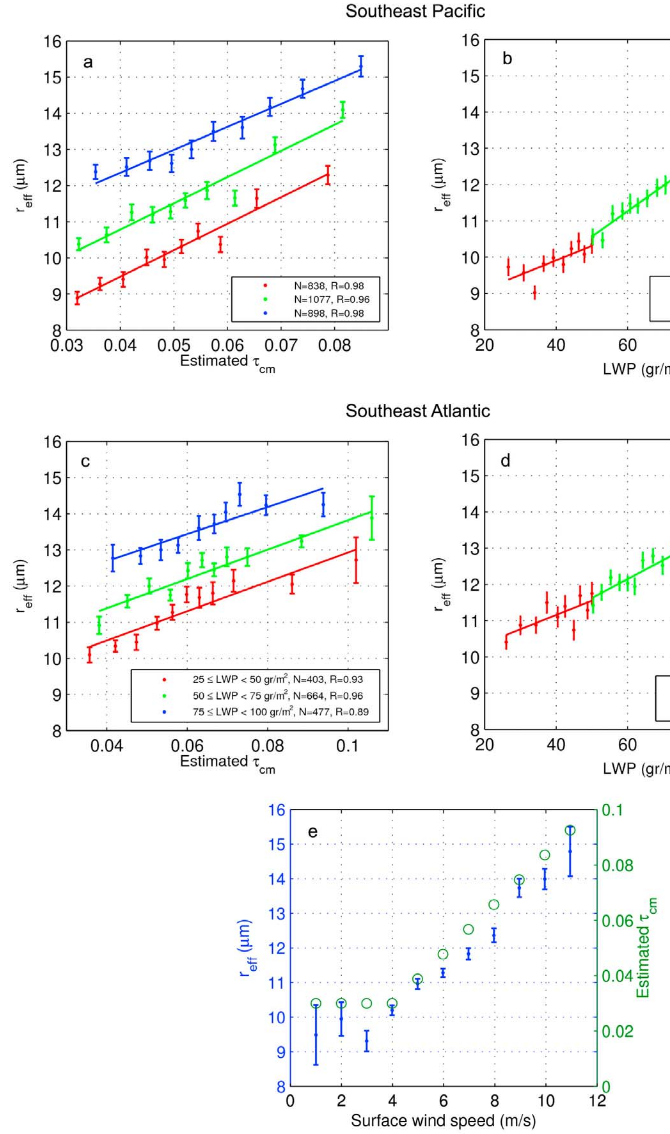

Figure 2. Cloud droplet effective radius, r_{eff}, plotted against (a, c) estimated τ_{cm}, (b, d) LWP, and (e) W. Green data points in Figure 2e show the empirically derived τ_{cm} dependence on W from *Lehahn et al.* [2010]. N denotes number of independent samples, each corresponding to one day and one pixel. In Figures 2a, 2b, 2c and 2d the data are plotted for different ranges of LWP (25–50, 50–75 and 75–100 gr/m^2, colored respectively in red, green and blue), while in Figure 2e only data associated with 50 < LWP < 75 gr/m^2 is shown. The analysis is performed over the area of high CC-MSC occurrence probability (see Figure 1) and is restricted to narrow ranges of CTT (281–283°k) CF (0.9–1) and ΔT(8–12 K). Data are sorted by τ_{cm} (Figures 2a and 2c), LWP (Figures 2b and 2d) and W (Figure 2e) and divided into 10 bins. Dots and error bars represent, respectively, the mean and standard error of r_{eff} within each bin.

Figure 2a, where r_{eff} is plotted against wind-based estimates of τ_{cm} over the region dominated by closed cells marine stratocumulus (black contours in Figure 1b). For the three LWP ranges r_{eff} is linearly correlated with τ_{cm}, with a remarkably consistent slope of 69.82 μm^{-1} ± 5.41, corresponding to average r_{eff} increase of 2.79 μm ± 0.22 for a twofold increase in τ_{cm}. Over the whole range of τ_{cm}, averaged increase in r_{eff} per LWP band is 3.38 μm ± 0.21. The shift between the regression lines, associated with the effect of changes in meteorological conditions (manifested in LWP variability), is 1.34 μm ± 0.07. Further examination of the relative contributions to cloud droplet size variability is done by correlating r_{eff} and LWP (Figure 2b). In agreement with the above, the increase in r_{eff} within a given LWP band (ΔLWP = 25 gr/m^2) is 1.6 μm ± 0.54. This suggests that approximately 1/2 of the r_{eff} variance can be attributed to aerosol perturbations and 1/2 to changes in meteorological conditions manifested in LWP variability. Accordingly, doubling of τ_{cm} corresponds to a 1.4 μm ± 0.11 increase in r_{eff}.

[13] How does the effect of coarse marine aerosols compares with that of fine anthropogenic aerosols? Similar analysis performed over the southeastern Atlantic - an area dominated by fine aerosols from biomass fires in southern Africa [*Kaufman et al.*, 2005] - shows a weaker effect of CMA on stratocumulus cloud droplet size (Figures 2c and 2d). Here, the averaged increase in r_{eff} per LWP band is 2.47 μm ± 0.44 (corresponding to a slope of 39.54 μm^{-1} ± 1.86), and the shift between the regression lines is 1.07 μm ±

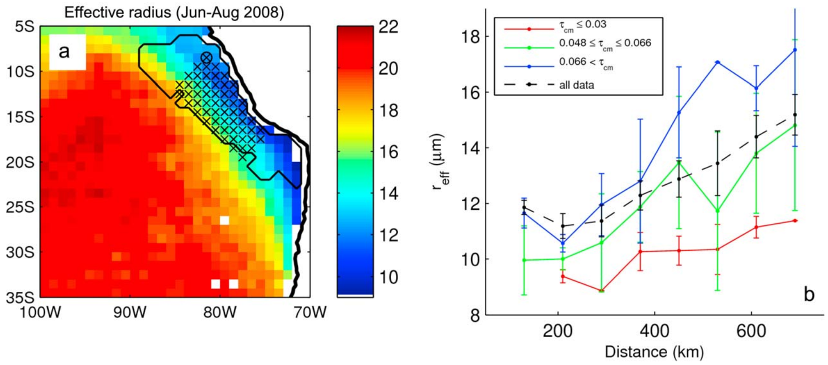

Figure 3. (a) 3 months mean (June–August 2008) of r_{eff}, emphasizing a distinct coast to open sea gradient. The black contour marks the area of high CC-MSC occurrence probability (see Figure 1), and the crosses mark the pixels used for extracting the cross-sections in Figure 3b. (b) Offshore cross sections of r_{eff} for different ranges of estimated τ_{cm} (solid lines, data is restricted to the narrow ranges of meteorological parameters detailed in Figure 2) and for the entire marine stratocumulus dataset (dashed line, data associated with CTTc > 280 K and CF > 0.9). Dots and error bars represent respectively the mean and standard deviation of r_{eff} at a given distance.

0.2 (Figure 2c). The increase in r_{eff} within a given LWP band is 1.13 μm ± 0.16 (Figure 2d), indicating that here also meteorology and CMA contribute equally to r_{eff} variance.

[14] Does the observed r_{eff} variance result from meteorological effects not filtered out through stratification to constrained environmental conditions? Wind induced production of CMA is expected to occur only when surface wind speed exceeds 4 m/s (for weaker wind conditions, breaking waves are rare if not entirely absent [*Lewis and Schwartz*, 2004]). Consequently, when wind intensity is below 4 m/s the measured background levels of τ_{cm} do not depend on the wind, while for stronger winds the two fields are well correlated (green circles in Figure 2e, corresponding to results from *Lehahn et al.* [2010]). We use this dependency pattern, which characterizes coarse marine aerosols in equatorial and mid-latitude regions, as a "sanity check" to our analysis results: similarly to τ_{cm}, at wind speed of ~4 m/s there is a distinct transition from no to almost linear dependency of marine southeast Pacific stratocumulus r_{eff} on W (blue data points in Figure 2e). This, together with the stratification to constrained meteorological conditions, implies that the observed increase in r_{eff} is indeed linked to CMA emitted to the atmosphere through wind driven processes.

[15] While the meteorological and coarse aerosol effects discussed above are the outcome of high frequency interactions (typical time scales smaller than ~1 day), r_{eff}, as well as other microphysical properties, are also subject to low frequency variabilities associated with regional changes in environmental conditions. For the southeastern Pacific marine stratocumulus cloud field, the regional scale variability is expressed by a distinct r_{eff} gradient, with averaged values ranging from approximately 11 μm near the coast to 15 μm at the far most edge of the seasonal cloud field (Figure 3a and dashed line in Figure 3b) during June–August 2008.

[16] As recently noted by *Twohy et al.* [2010], this offshore gradient is likely to result from a combined effect of anthropogenic aerosols increasing droplet number concentration near the coast and meteorological factors decreasing LWP. In view of the apparent importance of regional scale meteorology to r_{eff} variance, further examination of the coarse aerosol effect is done by plotting the change in r_{eff} as a function of distance from the coast for 3 bands of τ_{cm} (Figure 3b). The cross sections are extracted from data bounded by the same meteorological constrains used in Figure 2, with LWP ranging between 50 and 75 g/m^2. For a given distance, the increase in CMA loading is associated with increase in cloud droplet size. When comparing intense ($0.066 < \tau_{cm}$, blue curve) to negligible ($\tau_{cm} < 0.03$, red curve) coarse aerosol loading, the average change in r_{eff} is 4.2 μm ± 2.0. As suggested above, approximately 1/2 of this change is attributed to the effect of coarse marine aerosols.

[17] In contrast to the effect of fine aerosol particles, it is shown here that increase in coarse marine aerosol loading (estimated from surface wind speed), is associated with increase in droplets size of marine stratocumulus clouds. In accordance with results from different model studies [e.g., *Feingold et al.*, 1999], we propose that the shown association is due to the enhanced particles. Since aerosol activation strongly depends on aerosol size, the increase in concentration of large soluble sea salt particles is likely to produce larger cloud droplets. At a later stage, those bigger droplets are likely to grow more efficiently by collision-coalescence processes. Apart from the direct effect of reducing cloud reflectivity, increase in droplets size can also affect the efficiency of rain processes [*Comstock et al.*, 2004]. Precipitation has an important impact on the properties of marine stratocumulus clouds, and atmospheric aerosol is suggested to have a key role in precipitation modulations [*Feingold et al.*, 2010; *Wood et al.*, 2011]. These results strongly suggest that any attempt to quantify the impact of anthropogenic and biogenic marine aerosols on marine boundary layer clouds should take into account the opposing effect of wind-induced coarse marine particles.

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[19] The Editor thanks the anonymous reviewer.

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