Southern Ocean phytoplankton increases cloud albedo and reduces precipitation

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Received 17 December 2010; revised 14 February 2011; accepted 16 March 2011; published 26 April 2011.

[1] Effects of natural and anthropogenic aerosol particles on the radiation budget in cloudy atmospheres are still a major research topic. For example, can an increase or decrease in aerosol particle number, originating from changed dimethylsulfide (DMS) and isoprene emissions by marine phytoplankton, impact the earth radiation budget via increasing or decreasing planetary albedo and lifetime of clouds? And if so, is a shifted cloud droplet spectrum accompanied by a regional change in precipitation? Here, we show by a synergistic analysis of satellite observations (MODIS, SeaWiFS, AIRS, SSM/I and CERES) that the phytoplankton related emission of the mentioned gases into the atmosphere strongly influences cloud properties within a broad latitude belt in the Southern Hemisphere during the austral summer. For this season we detected indirect aerosol effects over the Southern Ocean from 45°S to 65°S, especially in regions with plankton blooms, indicated by high chlorophyll-a concentration in seawater. The strong increase in cloud condensation nuclei column content from 2.0×10^8 to more than 5.0×10^8 CCN/cm^2 for a chlorophyll increase from 0.3 to about 0.5 mg/m^3 in these regions decreases cloud droplet effective radius and increases cloud optical thickness for water clouds. Consequently, the upward short-wave radiative flux at the top of the atmosphere increases. Our analysis also reveals reduced precipitation over the Antarctic Polar Frontal Zone during strong plankton blooms. We suggest that due to fine particles formed in the atmosphere originating from gaseous DMS and possibly isoprene emissions the reduction of precipitation is caused by delayed homogeneous freezing in water clouds.
Citation: Krüger, O., and H. Graßl (2011), Southern Ocean phytoplankton increases cloud albedo and reduces precipitation, *Geophys. Res. Lett.*, *38*, L08809, doi:10.1029/2011GL047116.

1. Introduction

[2] Since the ocean covers over 70% of the Earth surface, biologically produced gases dissolved in oceanic waters and their subsequent emission into the marine troposphere play an important role in the global climate system. In particular molecular oxygen, sulphur containing compounds and possibly isoprene produced by phytoplankton often supersaturate in surface waters. These by-products of phytoplankton metabolism emanate into the atmosphere especially through the wave mediated bubble-whitecap mechanism and/or the rain mediated splash-bubble mechanism [*Blanchard and Woodcock*, 1957; *Monahan et al.*, 1983; *Stramska et al.*, 1990]. The magnitude of this marine aerosol production and the nature of its influence on climate via modulation of cloud optical properties are under discussion since 1983 [*Charlson et al.*, 1987; *Shaw*, 1983; *Ayers and Cainey*, 2007; *Andreae et al.*, 1995; *Meskhidze and Nenes*, 2006].

[3] The Sea-viewing Wide Field-of-View Sensor (SeaWiFS) and the Moderate Resolution Imaging Spectroradiometer (MODIS) allow monitoring of spatial and temporal distribution of phytoplankton chlorophyll in oceanic waters. These observations point to biologically very active waters in the Southern Ocean during austral summer, especially within the latitude belt of 45°S–65°S (Figure S1 of the auxiliary material).[1]

[4] In addition to being biologically active, this part of the Southern Ocean also experiences strong surface winds. This leads beside the abundance of bubbles and the related release of dimethylsulfide and isoprene also to the emission of sea salt particles. Therefore the atmosphere over these biologically very active oceanic waters is an ideal natural laboratory to detect and quantify indirect aerosol effects caused by marine aerosols.

2. Analysis of Satellite Data

[5] Two different data sets for chlorophyll-a concentration in the Southern Ocean from SeaWiFS and the NASA Ocean Biogeochemical Model (NOBM) [*Gregg and Casey*, 2007] show a remarkably clear increase of chlorophyll concentration with cloud condensation nuclei (CCN) column content (Figure 1a).

[6] The CCN numbers derived from MODIS [*Tanre et al.*, 1997; *Remer et al.*, 2005] data clearly show an annual cycle with more than 2.5 times as many CCN during austral summer (Figure S2 of the auxiliary material). The aerosol optical thickness [*Remer and Kaufmann*, 2006] and aerosol fine mode fraction undergo the same seasonal cycle showing values twice and five times as high during summer respectively. Since the values of both parameters and also the Angström exponent are anti-correlated with wind speed the increase of CCN numbers cannot be due to the increased production of sea salt particles [*Vallina et al.*, 2006]. In addition, the high values of the Angström exponent which are also frequently observed by the Aerosol Robotic Network (AERONET) stations close to the Southern Ocean indicate the strong contribution of fine mode particles. This leads to the conclusion that the formation of fine particles from DMS and potentially isoprene emission is trebling CCN number. The finding above is in line with the seasonal

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0094-8276/11/2011GL047116

[1]Auxiliary materials are available in the HTML. doi:10.1029/2011GL047116.

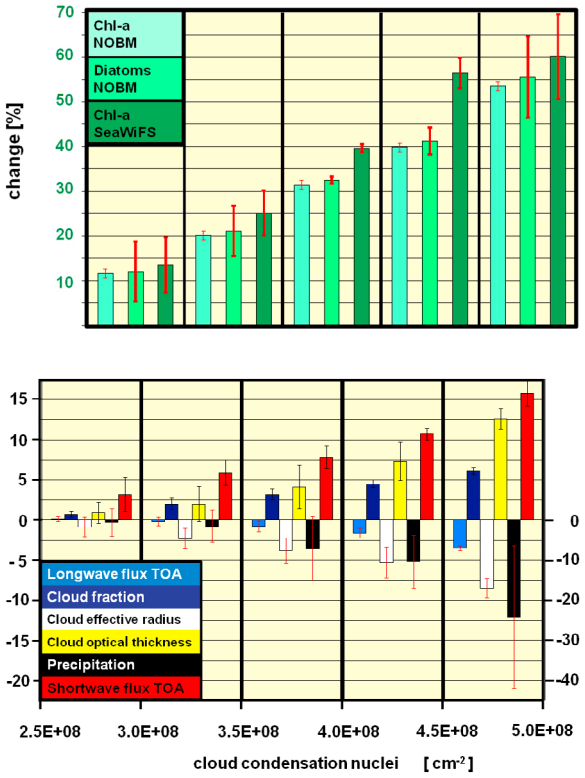

Figure 1. (a) Changes (in %) of chlorophyll-a concentration (Chl-a) from SeaWiFS and NASA Ocean Biogeochemical Model (NOBM), diatoms concentration ($C_{diatoms}$) from NOBM, (b) cloud effective radius for water clouds (R_e) from MODIS (in white, left ordinate), cloud fraction (C_f) from MODIS-TERRA (in dark blue, left ordinate), cloud optical thickness for water clouds (τ) from MODIS-TERRA (in yellow, left ordinate), upward shortwave radiative flux (SWF_U, in red, left ordinate) and upward longwave radiative flux (LWF_U, in light blue, left ordinate) at top-of-atmosphere from CERES and precipitation amount (P, in black, right ordinate) from HOAPS-3 relative to the three year mean range in CCN number per cm^2 from 2.0×10^8 to 2.5×10^8 (Chl-$a_{SeaWiFS}$ = 0.31 mg/m^3, Chl-a_{NOBM} = 0.30 mg/m^3, $C_{diatoms}$ = 0.29 mg/m^3, R_e = 13.9 μm, C_f = 89%, τ = 13.2, SWF_U = 326 W/m^2, LWF_U = 220 W/m^2, P = 55 mm/month) for 2003, 2004 and 2005. The standard deviation is also shown. The relative changes are calculated for a grid size of 1° longitude and 1° latitude within the latitude belt of 45°S–65°S. Results are shown for five intervals ranging from 2.5×10^8 – 3.0×10^8, 3.0×10^8 – 3.5×10^8, 3.5×10^8 – 4.0×10^8, 4.0×10^8 – 4.5×10^8 CCN/cm^2. While Chl-$a_{SeaWiFS}$, Chl-a_{NOBM}, C_{diatom}, C_f, τ and SWF_U are increasing with increasing CCN, the values for R_e and LWF_U and P are decreasing.

variation of H_2SO_4 deposition found in firn samples collected on the ice sheet of Antartica [*Aristarain et al.*, 1982]. The significant seasonal relationship of CCN numbers and aerosol methanesulphonate measured in marine air at Cape Grim (41°S) confirms a strong contribution of DMS [*Ayers and Gras*, 1991].

[7] The comparison of seasonal cycles derived from MODIS, SeaWiFS, SSM/I and CERES indicates that solar radiation is another important parameter within the negative feedback loop described by *Charlson et al.* [1987]. This is supported by marine in-situ measurements [*Vallina and Simó*, 2007], which clearly point to the influence of solar irradiance on DMS production in the surface ocean. Furthermore, solar irradiance is also needed for the formation of high CCN numbers. The remarkably high seasonal correlation of CCN number with both the downward shortwave radiation and the chlorophyll-a concentration in the water substantiates the hypothesis that DMS produced by plankton is released to the atmosphere, where it oxidizes to form small sulphate aerosol particles [*Bigg et al.*, 1984].

[8] The observed high CCN numbers over phytoplankton blooms stimulated our study whether the atmospheric part of the CLAW hypothesis, i.e. an increase of cloud albedo caused by increased CCN number, operates over the Southern Ocean. In order to identify the aerosol effects on clouds we firstly studied the effective radius of cloud droplets and the optical thickness of water clouds with MODIS [*Platnick et al.*, 2003] and CERES [*Chahine et al.*, 2006].

[9] However, MODIS data clearly show that the analysis of seasonal cycles alone does not allow to identify indirect aerosol effects, which is demonstrated by the missing seasonal anticorrelation of effective radius with optical thickness of water clouds (Figure S2 of the auxiliary material).

[10] Therefore, we investigated the Southern Ocean belt in more detail for austral summer by dividing it into 720 boxes of 1° latitude by 1° longitude. In our study we analyzed only boxes with a monthly mean CCN number higher than 2.0×10^8 CCN/cm^2. These monthly mean CCN numbers are higher than the median of the frequency distribution for the austral summer half year (ONDJFM, Figure S3 of the auxiliary material). Our analysis reveals that the chlorophyll-a concentration increases from 0.3 mg/m^3 to about 0.5 mg/m^3 for an increase in CCN number from 2.0×10^8 CCN/cm^2 to more than 5.0×10^8 CCN/cm^2 both for the satellite chlorophyll and the model assimilating these concentrations (shown in Figures 1a and S3). At the same time the wind speed decreases from 10.6 m/s to 5.1 m/s. The decreasing wind speed points to a decreasing production of sea salt particles [*O'Dowd et al.*, 1997; *Yoon and Brimblecombe*, 2002] while the chlorophyll-a concentration in seawater increases. Thus, we conclude that the increasing CCN numbers are due to phytoplankton-related emissions.

[11] The aerosol effects on clouds should lead to a cloud effective radius decrease and simultaneously to a cloud optical thickness increase at high CCN numbers. Just this dependence, the so called first indirect aerosol effect - sometimes called radius effect - is the result. With monthly mean CCN numbers doubling from 2.5×10^8 to 5.0×10^8 cm^{-2} the effective radius of cloud droplets decreases from 13.9 μm to 12.5 μm and the optical thickness for water clouds increases from 13.2 to 14.7 (Figure 1b). Hence, the cloud albedo is enlarged, causing a negative radiative forcing for areas with cloud cover. This finding is in agreement with previous studies [*Boers et al.*, 1998; *Nakajima et al.*, 2001; *Meskhidze and Nenes*, 2006; *Myhre et al.*, 2007].

[12] In order to determine the change in shortwave upward radiative flux for water clouds over the Southern Ocean we analysed data from another satellite instrument – NASA's CERES [*Chahine et al.*, 2006]. We found the change in CCN numbers from 2.5×10^8 to 5.0×10^8 is increasing reflection of solar radiation by clouds up to 15%

(see Figure 1b). We notice that this is a remarkably strong aerosol effect on clouds if compared to metropolitan regions where clouds form under the influence of strong anthropogenic emissions of air pollutants [*Krüger and Graßl*, 2002, 2004; *Krüger et al.*, 2004].

[13] Interestingly, for higher CCN numbers the CERES data also show a slightly decreased longwave upward flux at TOA while at the same time a decreased cloud top temperature is measured by MODIS. In principle this could be due to elevated cloud top height caused by higher CCN number as numerical modelling studies indicate [*Teller and Levin*, 2005], however to date the phenomenon has been observed only for clouds in polluted atmospheres [*Andreae et al.*, 2004; *Devasthale et al.*, 2006; *Krüger*, 2006].

[14] The radiation effects motivated us to investigate another aerosol effect on clouds, an increase of cloud lifetime with increased CCN numbers. As the droplet spectrum shifts towards smaller radii it could slow down coalescence and lower precipitation [*Rosenfeld*, 2000], hence extending cloud lifetime and perhaps increasing cloud top height.

[15] We analysed MODIS data to see if also a change in cloud cover depending on CCN number occurs. Amazingly, also for cloud fraction this indirect aerosol effect becomes visible: Cloud fraction increases by about 5% to more than 90% for doubling CCN number. Since optically thick haze layers are absent we can exclude misclassification [*Myhre et al.*, 2007] as sometimes seen for AOD values higher than 0.2. Moreover, the increase of cloud fraction occurs during both day and night. Our conclusion that higher abundance of aerosol particles alters cloud cover and cloud top height is supported by studies dealing with cloud development over the Atlantic Ocean [*Kaufman et al.*, 2005; *Koren et al.*, 2005].

[16] In order to get more evidence for aerosol effects on clouds we investigated whether a higher CCN number decreases the precipitation amount. To answer the question we analyzed data from two different precipitation climatologies, namely the Global Precipitation Climatology Project (GPCP) [*Adler et al.*, 2003] and the Hamburg Ocean Atmosphere Parameters and Fluxes from Satellite Data (HOAPS-3). Preceding studies for midlatitude cyclones confirm that precipitation structures and intensities in HOAPS-3 are in good agreement with observations of various precipitation types, including postfrontal lows (PFL) [*Klepp et al.*, 2005].

[17] What we found clearly supports our conclusion that the aerosol effects on clouds are driven by Southern Ocean phytoplankton: Precipitation amount decreases in both precipitation data sets with a percentage reduction of more than 10% for GPCP and of about 25% for HOAPS-3 data (see Figure 1b) at doubled CCN number. The result is of special importance because the influences of aerosols on the energy and the water cycle are observed at the same time.

[18] We summarize: For high CCN numbers 6 atmospheric parameters, cloud effective radius in water clouds, cloud optical thickness in water clouds, cloud fraction, upward longwave radiative flux at top of the atmosphere, upward shortwave radiative flux at top of the atmosphere and precipitation amount derived from 3 satellite instruments (MODIS, CERES and SSM/I) show a clear indication for aerosol effects on water clouds over the remote Southern Ocean during austral summer.

[19] In order to substantiate our results we tested whether they can be reproduced by dividing our study area, the Southern Ocean belt from 45°S to 65°S latitude into 4 smaller subareas (45°S–50°S, 50°S–55°S, 55°S–60°S, 60°S–65°S). Furthermore we included wind speed and Angström coefficient in our analysis allowing a characterisation of the aerosol which causes the effects. We found similar results for all the smaller belts except for precipitation north of 55°S latitude. For the belts south of 55°S latitude, where the precipitation amount is strongly reduced, the analysis of the Angström coefficient shows an abundance of fine mode aerosol particles (Figure S4 of the auxiliary material). Since the CCN number increases with decreasing wind speed we can rule out that additional sea salt particles cause the observed changes. Hence, we conclude that fine particles originating from increasing DMS and potentially isoprene emissions increase cloud albedo and reduce precipitation.

3. Discussion and Conclusions

[20] Even though it was discussed that the formation of new particles originating from isoprene chemistry may influence the cloud properties [*Meskhidze and Nenes*, 2006], there is experimental evidence that isoprene is not likely to induce nucleation [*Verheggen et al.*, 2007]. Nevertheless, it can contribute to secondary organic aerosol (SOA) by acid-catalyzed particle phase reactions [*Jang et al.*, 2002] and also via cloud processing [*Lim et al.*, 2005]. However, the annual cycle of the isoprene SOA source is found to be out of phase with the observed cycle of organic carbon aerosol in the remote Southern Ocean [*Arnold et al.*, 2009] pointing to a reduced importance.

[21] The precipitation loss could be due to the suppressed ice nucleation processes in combination with slowed down coalescence. While lowest optical thickness of ice clouds is observed for lower cloud top temperature and higher CCN number, coincident with higher optical thickness of water clouds, precipitation initiation seems to be affected by the reduced ice-crystal process.

[22] Since ice nuclei are expected to be widely absent over the remote Southern Ocean strongly supercooled water droplets will initiate freezing. On the other hand supersaturation of H_2SO_4 has been observed [*Zorn et al.*, 2008] in these oceanic regions. The smaller supercooled droplets likely contain a higher H_2SO_4 concentration near to blooming areas of plankton. Since it has been found in the laboratory that H_2SO_4 lowers the freezing point of small droplets [*Vortisch et al.*, 2000] the process of homogeneous ice formation needed for precipitation initiation could be suppressed. The analysis of the optical thickness and the effective particle size of ice clouds from MODIS supports this explanation (Figure S5 of the auxiliary material).

[23] Where are the high CCN numbers and blooming areas of phytoplankton located? The strongest aerosol effects on clouds occur along the Antarctic Polar Frontal Zone where strong upwelling of nutrient rich cold water takes place (Figure S6 of the auxiliary material). We find the highest CCN numbers over the blooming areas of phytoplankton with highest chlorophyll-a concentrations close to the cyclone track of the Southern Ocean as depicted by NASA's AIRS data for the 925 hPa geopotential height (Figure S7 of the auxiliary material). There, upwelling of

nitrate and silicate-rich deep water [*Falkowski et al.*, 1998] leads to high concentration of diatoms as also simulated by the NASA Ocean Biogeochemical Model (NOBM). Since we also find nearly constant lapse rates in the troposphere below 700 hPa for all CCN numbers from AIRS measurements (Figure S8 of the auxiliary material) we conclude that the changed stability of the atmosphere has no influence.

[24] Finally we investigated the dependence of our results on cloud liquid water path (L_p) using AIRS data. Aerosol effects occur for quite different L_p. However, the effect is strongest for low values of L_p (Figure S9 of the auxiliary material).

[25] In conclusion we observe a marine biology influence on the Earth's radiation budget over the Southern Ocean's plankton blooms (for a schematic see Figure S10). In addition, we found that not only the radiation budget at top of the atmosphere is reduced but also cloud cover is increased and precipitation amount is lowered over the Southern Ocean close to oceanic phytoplankton blooms. The strongest indirect aerosol effects are observed over the Antarctic Polar Frontal Zone at 60°S where nutrient rich water stimulates primary production in the austral summer half year.

[26] **Acknowledgments.** Images and data used in this study were acquired using the GES-DISC Interactive Online Visualization and Analysis Infrastructure (Giovanni) as part of NASA's Goddard Earth Sciences (GES) Data and Information Services Center (DISC). The authors thank R. Marks for valuable discussions and A. Devasthale for providing satellite data.

[27] The Editor thanks two anonymous reviewers for their assistance in evaluating this paper.

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