



## Lack of correlation between chlorophyll *a* and cloud droplet effective radius in shallow marine clouds

Matthew A. Miller<sup>1</sup> and Sandra E. Yuter<sup>1</sup>

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[1] The hypothesis that areas of high oceanic productivity affect the physical properties of shallow marine clouds via the production of secondary organic aerosols is evaluated using satellite data. The correlation between chlorophyll *a* concentrations, an indication of oceanic productivity, and low cloud droplet liquid phase effective radius ( $R_e$ ) is examined for several ocean regions and time periods. While a strong correlation between chlorophyll *a* and low  $R_e$  can occur for specific periods in some locations, the correlation is not reproducible in other regions and time periods. The intermittent correlation between high concentrations of chlorophyll *a* and low  $R_e$  is a coincidence and is not representative of a dominant, monotonic, causative relation between secondary organic aerosols and marine shallow cloud properties. **Citation:** Miller, M. A., and S. E. Yuter (2008), Lack of correlation between chlorophyll *a* and cloud droplet effective radius in shallow marine clouds, *Geophys. Res. Lett.*, 35, L13807, doi:10.1029/2008GL034354.

### 1. Introduction

[2] Shallow, liquid phase, marine clouds are important to global climate because they reflect a substantial amount of incoming shortwave radiation but have only a small effect on net outgoing longwave radiation compared to the sea surface. The interactions among aerosols, cloud properties, boundary layer dynamics, surface processes, and radiative effects in these shallow marine clouds are complex and can be non-monotonic [Ackerman *et al.*, 2004; Jiang and Feingold, 2006; Xue *et al.*, 2008; Wood, 2007].

[3] Meskhidze and Nenes [2006] (hereinafter referred to as MN06) explored the effect of oceanic biological productivity on the properties of shallow marine clouds. MN06 hypothesized that secondary organic aerosols (SOA), produced as a byproduct of biological production by phytoplankton, can affect the number concentration and size distribution of cloud droplets in marine clouds. They used the concentration of chlorophyll *a*, estimated by the Sea-viewing Wide Field-of-view Sensor (SeaWiFS) on the Seastar satellite, as a proxy for the productivity of phytoplankton. Cloud droplet liquid phase effective radius ( $R_e$ ), estimated by the Moderate Resolution Imaging Spectroradiometer (MODIS) on the Terra satellite, was monitored to assess any changes in cloud properties. MN06 used the correlation between high concentrations of chlorophyll *a* and low  $R_e$  as evidence to support a causative

link between oceanic biological productivity and marine cloud characteristics. MN06 argue that increases in chlorophyll *a* concentration lead to increased concentrations of SOA, which in turn cause a decrease in  $R_e$  and an increase in cloud droplet concentration. MN06 examined other meteorological controls that could alter  $R_e$  and concluded that SOA produced as a byproduct of biological production by phytoplankton blooms is the dominant factor influencing  $R_e$  and cloud droplet concentration properties.

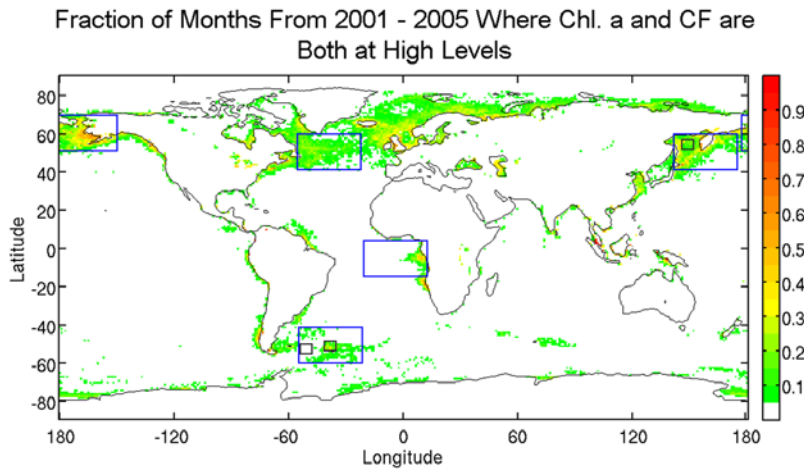
[4] The crux of the MN06 argument relies on the high anticorrelation of chlorophyll *a* to  $R_e$  in areas with high chlorophyll *a* concentrations. We tested the reproducibility of this correlation by examining not only MN06's southern Atlantic study region but other oceanic areas that experience phytoplankton blooms. Figure 1 shows the fraction of months from 2001–2005 where the concentration of chlorophyll *a* was high enough to indicate strong phytoplankton blooms and the cloud fraction was high enough to ensure reliable  $R_e$  measurements. Chlorophyll *a* concentrations above  $1 \text{ mg m}^{-3}$  are considered sufficient to indicate a phytoplankton bloom, and cloud fraction values above 0.7 are considered sufficient for reliable  $R_e$  measurements. Each of the blue boxes in Figure 1 indicates an area where the spatial correlation between chlorophyll *a* and  $R_e$  was evaluated. The smaller black boxes indicate areas where the time series of chlorophyll *a* and  $R_e$  were evaluated. The potential effects of cloud fraction on the reliability of  $R_e$  measurements are discussed in section 5. Table 1 summarizes the spatial correlation statistics for the areas indicated in Figure 1.

### 2. Spatial Correlation Methodology

[5] To evaluate the spatial correlation between phytoplankton and marine cloud properties, we define the high chlorophyll *a* bloom area as the subset of  $1^\circ \times 1^\circ$  grid boxes where the average concentration of chlorophyll *a* was greater than or equal to  $1 \text{ mg m}^{-3}$ . The remaining grid boxes were considered non-bloom areas. In contrast, MN06 defined the latitude band between  $48^\circ\text{S}$  and  $56^\circ\text{S}$  within the larger geographic area between  $55^\circ\text{W}$  to  $21^\circ\text{W}$  and  $42^\circ\text{S}$  to  $60^\circ\text{S}$  as inside the phytoplankton bloom area and the areas to the north and south as outside the bloom. MN06 calculated correlations of oceanic and atmospheric parameters for  $2^\circ \times 2^\circ$  grid boxes over their latitude bands as defined above (MN06, Table 1). For the latitude band definition of the bloom area to be applied to other blooms, the size, shape, and geographic surroundings of the bloom would have to be similar to the bloom from the MN06 case.

[6] The time period 11 December 2001–8 January 2002 corresponds to the period studied in MN06 (MN06,

<sup>1</sup>Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Raleigh, North Carolina, USA.



**Figure 1.** The fraction of months from 2001–2005 where chlorophyll *a* concentrations were above 1 mg m<sup>-3</sup> and cloud fraction was above 0.7. The data were averaged to a 1° × 1° grid. The blue boxes denote the phytoplankton bloom regions examined in this study. The smaller black boxes denote areas where time series were calculated.

Figure 3a). Using the 1° grid box threshold as opposed to the latitude band definition of bloom and non-bloom areas yields higher correlations between chlorophyll *a* concentration and  $R_e$  inside the bloom area ( $-0.67 \pm 0.19 - 0.13$  versus MN06  $-0.49 \pm 0.09$ ) and slightly lower correlations outside the bloom area ( $-0.02 \pm 0.08$  versus MN06  $0.18 \pm 0.14$ ) than were found in MN06. For that event, we agree with MN06 that inside the bloom area there is a strong negative correlation between chlorophyll *a* concentrations and  $R_e$  (Table 1). The comparable correlations indicate that the 1° grid box threshold methodology used in this study is acceptable for qualitative comparison to the results of the MN06 study.

### 3. Temporal Variation of $R_e$ and Phytoplankton

[7] Figure 2 shows the time series of area-averaged chlorophyll *a* concentration from SeaWiFS monthly products and  $R_e$  from MODIS monthly products for two different areas in the southern Atlantic Ocean for the period from March 2000–December 2005. The area-averaged time series in Figure 2b is for the same time period and area used in MN06 (their Figure 2) except that it is compiled from monthly rather than eight-day averaged data. We use monthly

data for computational ease since the time series in question are presented for qualitative comparison and analysis.

[8] For the bloom area in Figure 2,  $R_e$  oscillates on an annual cycle that coincides with the annual phytoplankton blooms. The time series for a nearby area with low chlorophyll *a* to the west and upwind of the bloom area is shown in Figure 2c. Comparison of the time series of  $R_e$  for this nearby area with low chlorophyll *a* to the area with high chlorophyll *a* shows that  $R_e$  oscillations have similar frequencies and amplitudes regardless of the presence of phytoplankton blooms.

[9] The Sea of Okhotsk, which lies northeast of Japan, adjacent to Russia’s Kamchatka Peninsula (Figure 3), experiences intense but short-lived phytoplankton blooms during the local spring and summer. During blooms in the Sea of Okhotsk, levels of chlorophyll *a* increase by a factor of ten. As with the area off South Georgia Island [Ward *et al.*, 2005], the phytoplankton blooms in the Sea of Okhotsk are predominantly composed of diatoms [Sorokin and Sorokin, 1999]. This area also has low-cloudiness characteristics similar to the MN06 original study area [Norris, 1998].

[10] The five-year time series of  $R_e$  and chlorophyll *a* concentration for the boxed area in the Sea of Okhotsk shows the annual oscillation in  $R_e$  and the local maxima in

**Table 1.** Correlations of the Concentration of Chlorophyll *a* to  $R_e$  Inside and Outside the Bloom Areas for Five Different Events Using the 1° Grid Box Threshold Definition of Bloom and Non-bloom Area as Outlined in Section 2<sup>a</sup>

Location (Date Range)	Correlation Between Chlorophyll <i>a</i> and $R_e$	
	Non-Bloom Area Chl. <i>a</i> Concentration (Chl. <i>a</i> < 1 mg m <sup>-3</sup> )	Bloom Area Chl. <i>a</i> Concentration (Chl. <i>a</i> > 1 mg m <sup>-3</sup> )
Southern Atlantic (11 Dec 2001–8 Jan 2002)	$-0.02 \pm 0.08$	$-0.67^{+0.19}_{-0.13}$
Southern Atlantic (11 Dec 2003–8 Jan 2004)	$0.15^{+0.08}_{-0.09}$	$-0.1^{+0.30}_{-0.29}$
Sea of Okhotsk (26 June 2003–27 July 2003)	$-0.13 \pm 0.12$	$-0.02^{+0.18}_{-0.17}$
Northwest Atlantic (4 Aug 2003–4 Sep 2004)	$-0.18 \pm 0.08$	$-0.09^{+0.33}_{-0.31}$
West African Coast (1 Jan 2002–1 Feb 2002)	$0.13^{+0.09}_{-0.10}$	$0.02^{+0.36}_{-0.37}$
Bering Sea (7 Apr 2004–8 May 2004)	$-0.1^{+0.17}_{-0.16}$	$-0.13 \pm 0.11$

<sup>a</sup>The 95% confidence intervals are given (asymmetry of confidence intervals is a function of correlation and sample size). The correlations for the Northwest Atlantic event are for the area from 42°N to 60°N and 55°W to 21°W. The correlations for the Western African Coast event are for the area from 4°N to 14°S and 20°W to 14°E. The correlations for the Bering Sea event are for the area from 52°N to 70°N and 176°E to 150°W.

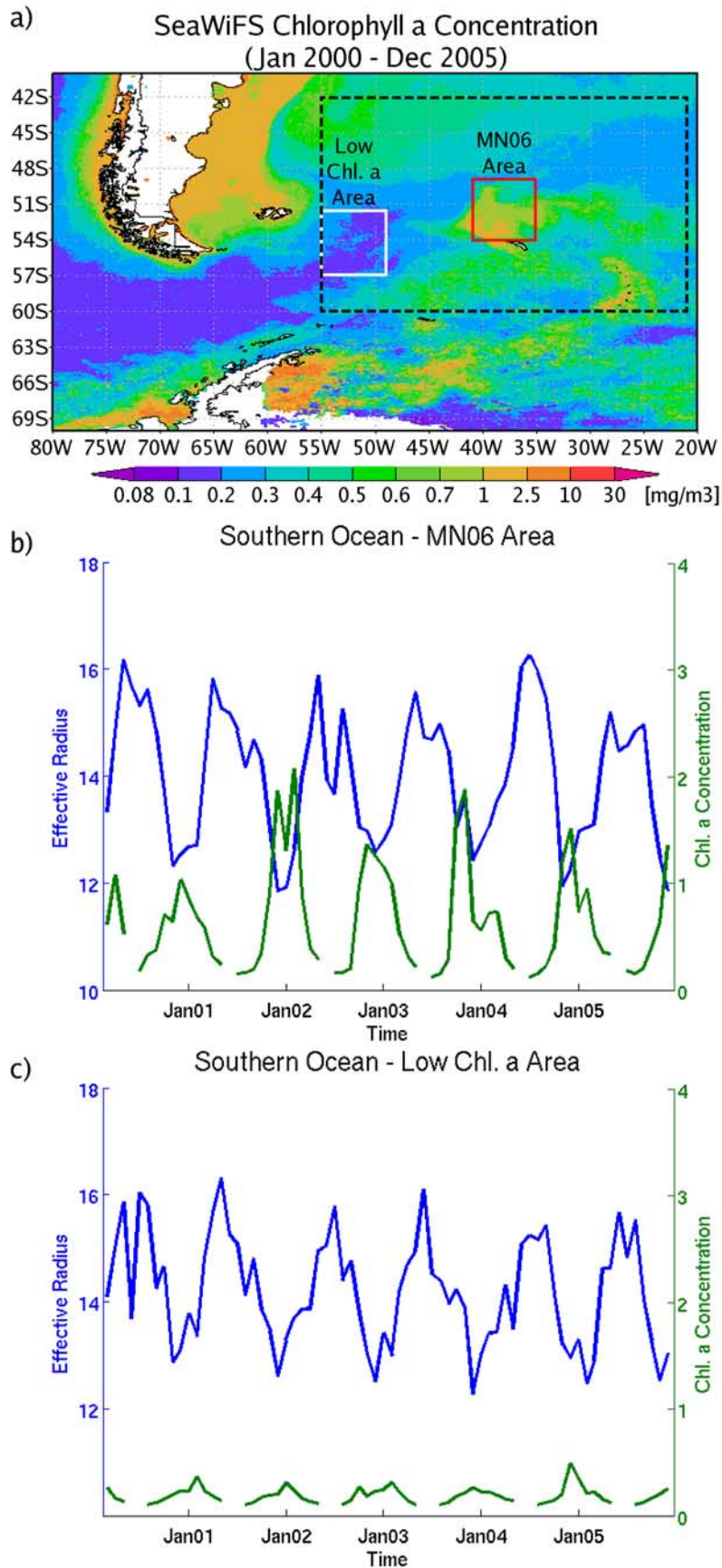
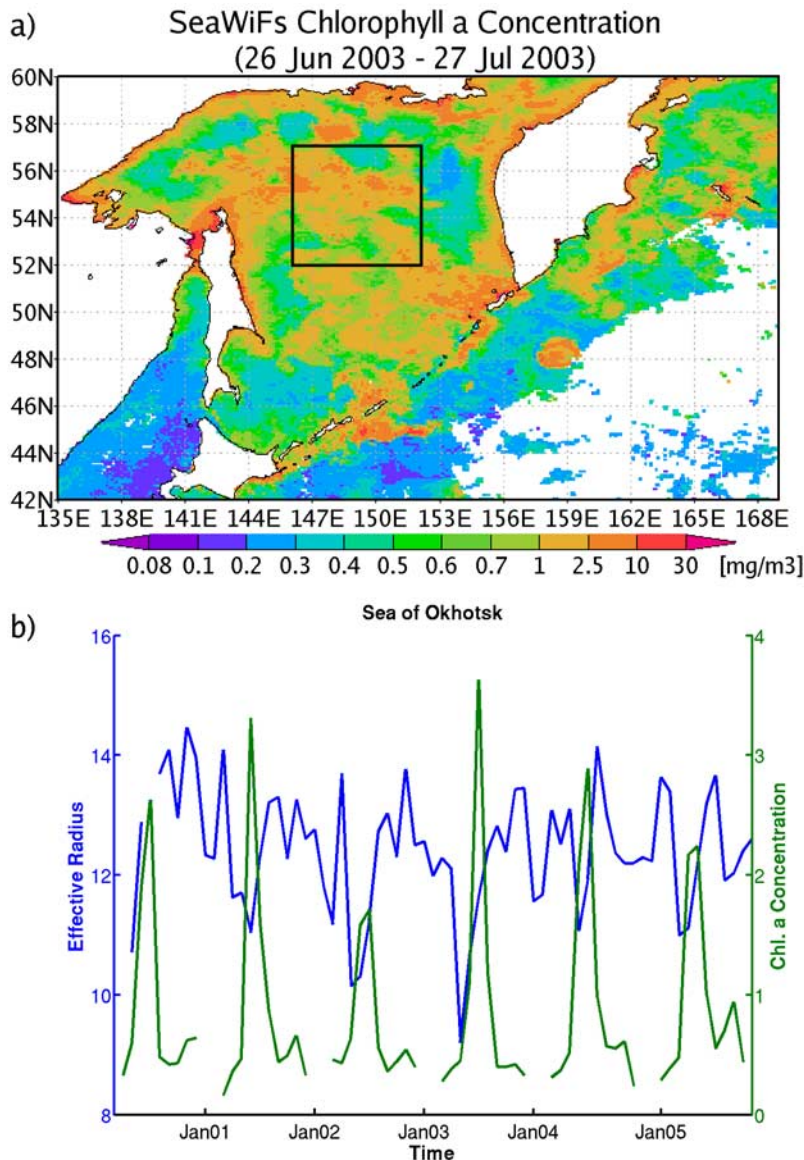


Figure 2



**Figure 3.** (a) Sea of Okhotsk regional map of SeaWiFS-derived chlorophyll *a* concentration derived from 8-day averaged data for a 4-week period for 26 June 2003–27 July 2003. The black box indicates the area from which the time series in Figure 3b is derived. White areas denote land or missing data. (b) Time series of monthly chlorophyll *a* concentration and  $R_e$  as observed by SeaWiFS and MODIS, respectively, for an area averaged from 57°N to 52°N and 146°E to 152°E.

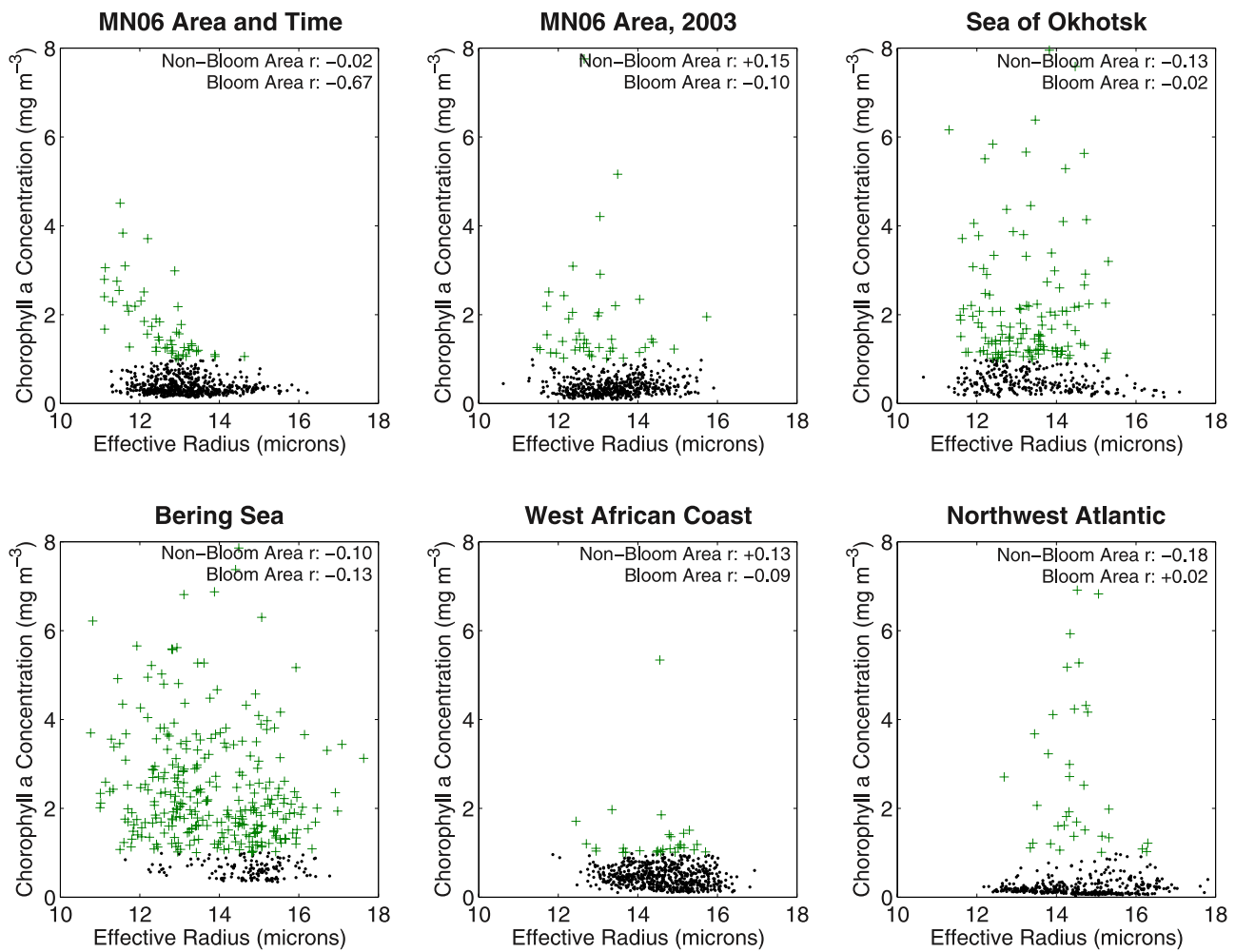
chlorophyll *a* concentration that denote the annual phytoplankton blooms (Figure 3). The chlorophyll *a* concentrations and  $R_e$  do not have the proper phase relationship to produce a negative correlation that would support the MN06 theory. The minimum in  $R_e$  precedes the spike in chlorophyll *a* concentration by  $\sim 2$  months, suggesting that the phenomena which alter the marine cloud properties on

an annual basis in the Sea of Okhotsk act before the phytoplankton blooms occur.

#### 4. Spatial Correlation of $R_e$ and Phytoplankton

[11] Examination of phytoplankton bloom events across multiple areas facilitates the evaluation of the relationship

**Figure 2.** (a) South Atlantic regional map of SeaWiFS-derived chlorophyll *a* concentration derived from monthly data for January 2000–December 2005. Dashed box indicates area where spatial correlations were calculated. Red box indicates MN06's bloom area, and the white box indicates low-bloom area from which time series in Figures 2b and 2c are respectively derived. White areas denote land or missing data. (b) Time series of monthly chlorophyll *a* concentration and  $R_e$  as observed by SeaWiFS and MODIS, respectively, for an area averaged from 49°S to 54°S and 41°W to 35°W (red box). (c) Time series of monthly chlorophyll *a* concentration and  $R_e$  as observed by SeaWiFS and MODIS respectively for an area averaged from 52°S to 57°S and 55°W to 49°W (white box).



**Figure 4.** Scatter plots of  $R_e$  and chlorophyll  $a$  for bloom (dots) and non-bloom (crosses) areas for the events described in Table 1. The spatial correlation values from Table 1 are shown in upper right of each plot.

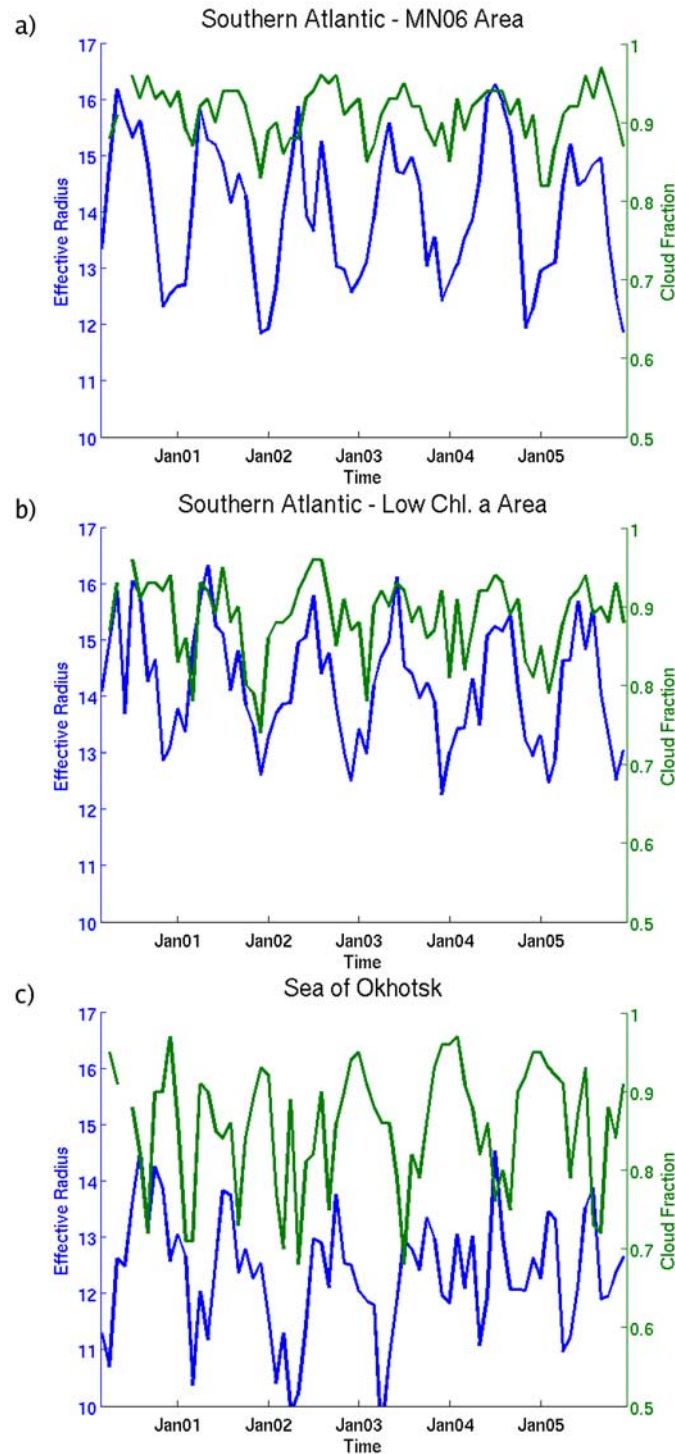
between phytoplankton and marine cloud properties in a variety of meteorological backgrounds with conditions both similar and different to the southern Atlantic Ocean. Figure 4 shows scatter plots of chlorophyll  $a$  concentrations and  $R_e$  for six bloom events within five geographic areas depicted in the blue boxes in Figure 1.

[12] As was stated in section 2, we agree with MN06 that inside the Southern Atlantic bloom area for the time period 11 December 2001–8 January 2002, there is strong negative correlation between chlorophyll  $a$  concentrations and  $R_e$  ( $-0.67 \pm 0.19 - 0.13$ ). However, this relationship is not reproducible in other cases. The correlation between  $R_e$  and chlorophyll  $a$  is weak ( $-0.1 \pm 0.30 - 0.29$ ) inside the bloom area over the same area that was examined in MN06, but for a bloom from 11 December 2003–8 January 2004 of similar size and intensity. The correlation between SeaWiFS estimated chlorophyll  $a$  concentrations and MODIS estimated  $R_e$  for a bloom in the Sea of Okhotsk for 26 June 2003–27 July 2003 was almost zero ( $-0.02 \pm 0.18 - 0.17$ ). Furthermore, the spatial correlations from bloom events in the Northwest Atlantic (4 August 2004–4 September 2004), off the western African Coast (1 January 2002–1 February 2002), and in the Bering Sea (7 April 2004–8 May 2004)

all show a poor relationship between chlorophyll  $a$  concentrations and  $R_e$  (Figure 4). The bloom and non-bloom area spatial correlations for each case are given in Table 1. With the exception of the bloom event examined in the MN06 study, none of the other events – from different locations and time periods – demonstrate a strong correlation between chlorophyll  $a$  and  $R_e$  in bloom areas.

## 5. Potential Observational Errors

[13] There are several potential sources of error in satellite estimation of  $R_e$ ; chief among them are cloud fraction and view angle. The view angle should not represent a source of error that would differentially impact the satellite  $R_e$  retrievals since the MODIS satellite’s sun-synchronous orbits allows for a constant view angle. Several studies have examined the impacts of cloud fraction on the accuracy of MODIS  $R_e$  estimations [Coakley *et al.*, 2005; Cornet *et al.*, 2005; Kato *et al.*, 2006].  $R_e$  retrievals are based on 1-D radiative transfer theory that neglects horizontal variations of cloud properties. As a result,  $R_e$  is often overestimated. The magnitude of the overestimate is a function of cloud type and decreasing cloud fraction.  $R_e$  estimates are most reliable where clouds are more homogeneous in the



**Figure 5.** Area-averaged time series of MODIS cloud fraction and  $R_e$  from March 2000–December 2005 for (a) the Southern Atlantic region in the red box in Figure 2, (b) the Southern Atlantic region in the white box in Figure 2, and (c) the Sea of Okhotsk region in the black box in Figure 3.

horizontal (e.g. stratus) and less reliable where clouds are less homogeneous in the horizontal (e.g. cumulus).

[14] Figure 5 shows area-averaged time series of cloud fraction and  $R_e$  for the three study areas depicted in Figures 2 and 3. The area-averaged cloud fraction for both the high and low phytoplankton areas in the Southern Atlantic oscillates to a value no lower than 0.84. The cloud fraction

for the Sea of Okhotsk is more variable with area-averaged cloud fraction sometimes dropping just below 0.6 but with a mean above 0.8. If cloud fraction represented a significant source of error in the MODIS  $R_e$  estimations, we would expect to see a local maxima in  $R_e$  coinciding with minima in cloud fraction representing the bias where low cloud fraction leads to overestimation of  $R_e$ . Examination of the

plots in Figure 5 illustrates that this is not that case. For the cases examined in this study, cloud fraction does not appear to represent a significant source of error.

## 6. Conclusions

[15] We have shown that the correlation between low  $R_e$  and high chlorophyll  $a$  shown by MN06 is not reproducible in other regions and time periods (Table 1). MN06's theory that phytoplankton derived SOA is the dominant influence on  $R_e$  in marine clouds during plankton blooms requires that high chlorophyll  $a$  systematically correlate with low  $R_e$ . We have shown in Figure 4 that this is not the case for the southern Atlantic region in December 2003–January 2004, the Sea of Okhotsk region in June–July 2003, the Northwest Atlantic region in August–September 2004, off the Western African Coast in January 2002, and the Bering Sea region in April–May 2004. The strong correlation between high chlorophyll  $a$  concentration and low  $R_e$  for the area off South Georgia Island for the time period studied by MN06 is a coincidence and not representative of a widespread trend.

[16] A recent letter by Wingenter [2007] further questions the conclusions of MN06 based on MN06's three-order-of-magnitude overestimation of SOA levels in the phytoplankton bloom area. In their reply, Meskhidze and Nenes [2007] state that this correction does not alter their conclusions because isoprene air-sea fluxes can vary by two orders of magnitude. The isoprene fluxes used in their cloud parcel model simulations represent the high end of measured values within phytoplankton blooms in the Southern Ocean.

[17] It is well established that SOA are a source of cloud condensation nuclei [Wallace and Hobbs, 2006]. While SOA are likely a contributing factor to shallow marine cloud  $R_e$  and number concentration properties, further evidence does not corroborate a direct and strong link between phytoplankton and clouds. The correlation between chlorophyll  $a$  and  $R_e$  is not systematically reproducible and hence is likely a coincidence.

[18] **Acknowledgments.** The authors greatly appreciate the advice and assistance of N. Riemer and A. Aiyer. This research was supported by NASA grant NNG04GA65G and a NASA Earth and Space Science Fellowship.

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M. A. Miller and S. E. Yuter, Department of Marine, Earth, and Atmospheric Sciences, North Carolina State University, Campus Box 8208, Raleigh, NC 27695, USA. (mamille4@ncsu.edu)

## ERRATA

### Description and explanation of the following corrections

The following corrections are for Table 1 and its caption. The listed date ranges for the rows describing data in the Northwest Atlantic and Bering Sea were corrected to fix typographical errors in the original submission. The original caption only defined the geographic locations of 3 of the 5 regions. The revised caption provides the locations for all 5 of the regions. MODIS and SeaWiFS product version numbers are added for clarity. Also, a minor typographical error in section 1 paragraph 2 is corrected.

### Original Table 1 and Caption:

**Table 1.** The correlations of the concentration of *chlorophyll a* to  $R_e$  inside and outside the bloom areas for five different events using the  $1^\circ$  grid box threshold definition of bloom and non-bloom area as outlined in sect. 2. The 95% confidence intervals are given (asymmetry of confidence intervals is a function of correlation and sample size). The correlations for the Northwest Atlantic event are for the area from  $42^\circ$  N to  $60^\circ$  N and  $55^\circ$  W to  $21^\circ$  W. The correlations for the Western African Coast event are for the area from  $4^\circ$  N to  $14^\circ$  S and  $20^\circ$  W to  $14^\circ$  E. The correlations for the Bering Sea event are for the area from  $52^\circ$  N to  $70^\circ$  N and  $176^\circ$  E to  $150^\circ$  W.

### Correlation Between *Chlorophyll a* and $R_e$

Location (Date Range)	Non-Bloom Area <i>Chl. a</i> Concentration ( <i>Chl. a</i> < 1 mg m <sup>-3</sup> )	Bloom Area <i>Chl. a</i> Concentration ( <i>Chl. a</i> > 1 mg m <sup>-3</sup> )
Southern Atlantic (11 Dec 2001 – 8 Jan 2002)	-0.02±0.08	-0.67 <sup>+0.19</sup> <sub>-0.13</sub>
Southern Atlantic (11 Dec 2003 – 8 Jan 2004)	0.15 <sup>+0.08</sup> <sub>-0.09</sub>	-0.1 <sup>+0.30</sup> <sub>-0.29</sub>
Sea of Okhotsk (26 June 2003 – 27 July 2003)	-0.13±0.12	-0.02 <sup>+0.18</sup> <sub>-0.17</sub>
Northwest Atlantic (4 Aug 2003 – 4 Sep 2004)	-0.18±0.08	-0.09 <sup>+0.33</sup> <sub>-0.31</sub>
West African Coast (1 Jan 2002 – 1 Feb 2002)	0.13 <sup>+0.09</sup> <sub>-0.10</sub>	0.02 <sup>+0.36</sup> <sub>-0.37</sub>
Bering Sea (7 Apr 2004 – 8 May 2004)	-0.1 <sup>+0.17</sup> <sub>-0.16</sub>	-0.13±0.11



**Corrected Table 1 and Caption:**

**Table 1.** The correlations of the concentration of *chlorophyll a* to  $R_e$  inside and outside the bloom areas for five different events using the 1° grid box threshold definition of bloom and non-bloom area as outlined in sect. 2 using data from Terra MODIS version 5 and SeaWiFS version 5.1. The 95% confidence intervals are given (asymmetry of confidence intervals is a function of correlation and sample size). The correlations for the Southern Atlantic events are for the area from 42° S to 60° S and 55° W to 21° W. The correlations for the Sea of Okhotsk event are for the area from 60° N to 42° N and 135° E to 169° E. The correlations for the Northwest Atlantic event are for the area from 42° N to 60° N and 55° W to 21° W. The correlations for the Western African Coast event are for the area from 4° N to 14° S and 20° W to 14° E. The correlations for the Bering Sea event are for the area from 52° N to 70° N and 176° E to 150° W.

**Correlation Between *Chlorophyll a* and  $R_e$**

<b>Location (Date Range)</b>	<b>Non-Bloom Area <i>Chl. a</i> Concentration (<i>Chl. a</i> &lt; 1 mg m<sup>-3</sup>)</b>	<b>Bloom Area <i>Chl. a</i> <i>Chl. a</i> Concentration (<i>Chl. a</i> &gt; 1 mg m<sup>-3</sup>)</b>
<b>Southern Atlantic (11 Dec 2001 – 8 Jan 2002)</b>	-0.02±0.08	-0.67 <sup>+0.19</sup> <sub>-0.13</sub>
<b>Southern Atlantic (11 Dec 2003 – 8 Jan 2004)</b>	0.15 <sup>+0.08</sup> <sub>-0.09</sub>	-0.1 <sup>+0.30</sup> <sub>-0.29</sub>
<b>Sea of Okhotsk (26 June 2003 – 27 July 2003)</b>	-0.13±0.12	-0.02 <sup>+0.18</sup> <sub>-0.17</sub>
<b>Northwest Atlantic (4 Aug 2004 – 4 Sep 2004)</b>	-0.18±0.08	-0.09 <sup>+0.33</sup> <sub>-0.31</sub>
<b>West African Coast (1 Jan 2002 – 1 Feb 2002)</b>	0.13 <sup>+0.09</sup> <sub>-0.10</sub>	0.02 <sup>+0.36</sup> <sub>-0.37</sub>
<b>Bering Sea (6 Apr 2004 – 7 May 2004)</b>	-0.1 <sup>+0.17</sup> <sub>-0.16</sub>	-0.13±0.11

**Original Sentence from Section 1 Paragraph 2:**

... The potential effects of *cloud fraction* on the reliability of  $R_e$  measurements are discussed in section 4. ...

**Corrected Sentence from Section 1 Paragraph 2:**

... The potential effects of *cloud fraction* on the reliability of  $R_e$  measurements are discussed in section 5. ...