



*The Abdus Salam*  
**International Centre for Theoretical Physics**



**2256-15**

**Workshop on Aerosol Impact in the Environment: from Air Pollution to  
Climate Change**

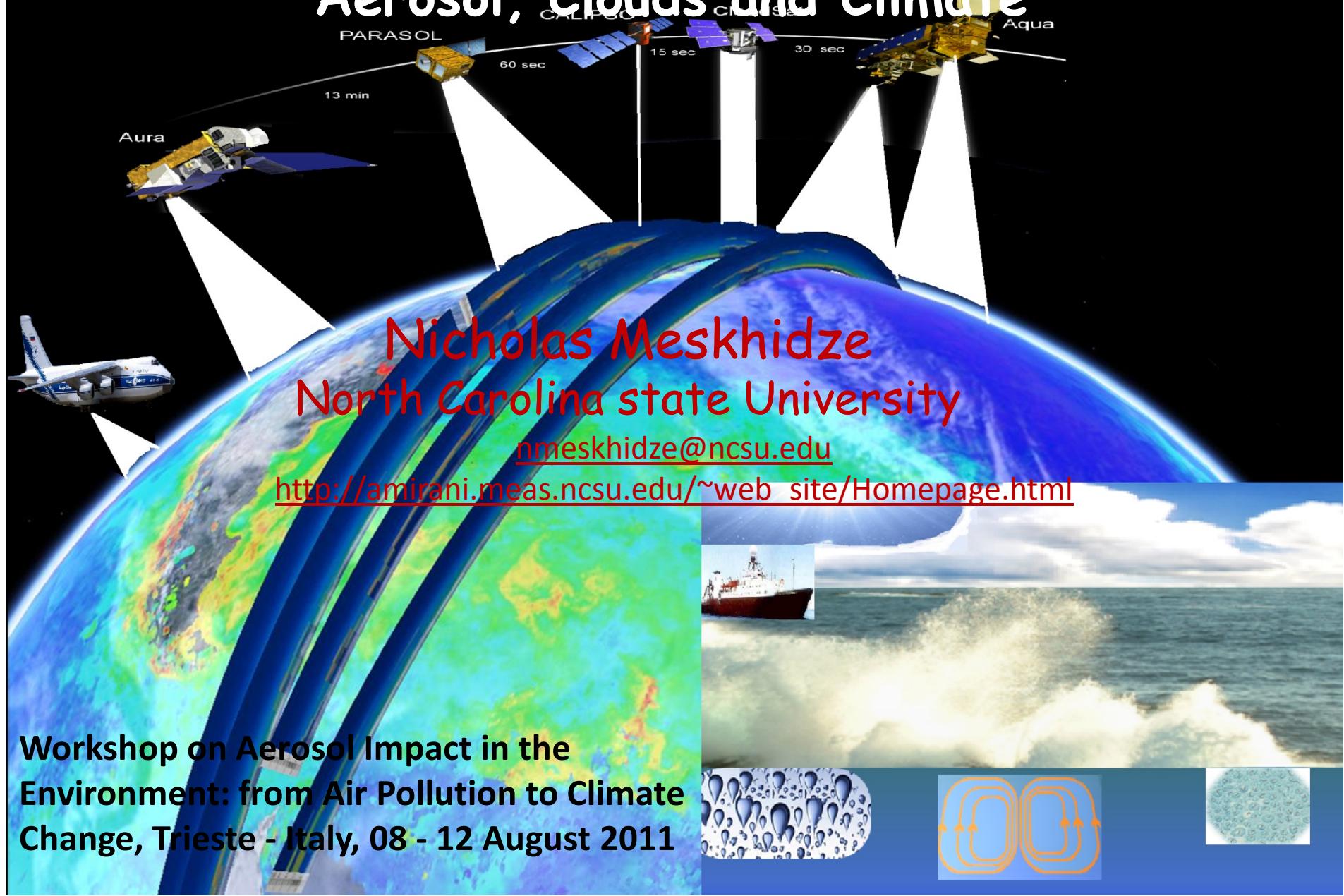
*8 - 12 August 2011*

**The Effect of Ocean Biogeochemistry on Aerosols, Clouds and Climate**

N. Meskhidze

*North Carolina State Univ. Raleigh  
USA*

# The Effect of Ocean Biogeochemistry on Aerosol, Clouds and Climate



# Outline

- 
- 1) Introduction
  - 2) Biogenic gas emissions from the ocean and their impact on aerosol/cloud interaction and radiative properties of the overlying atmosphere
  - 3) Sources, chemical composition and size distribution of ocean-derived primary organic aerosols
  - 4) Model results
  - 5) The future



Introduction

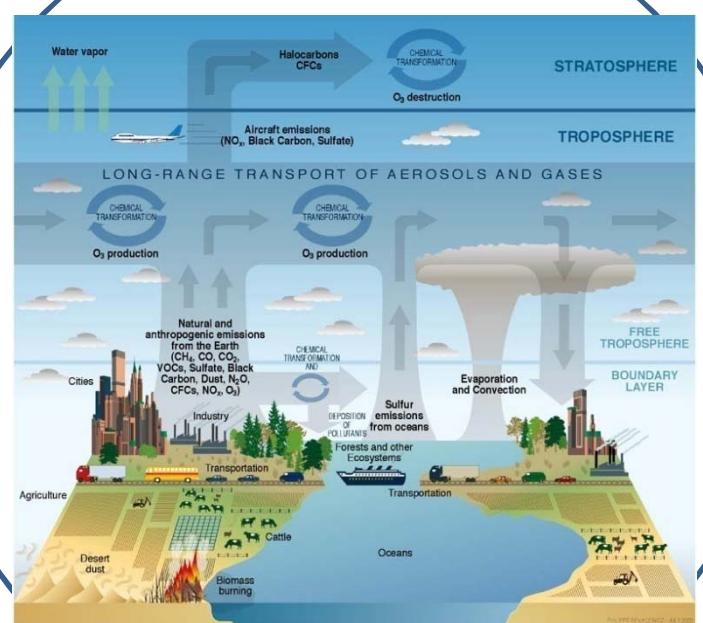
Gas emissions

POM emissions

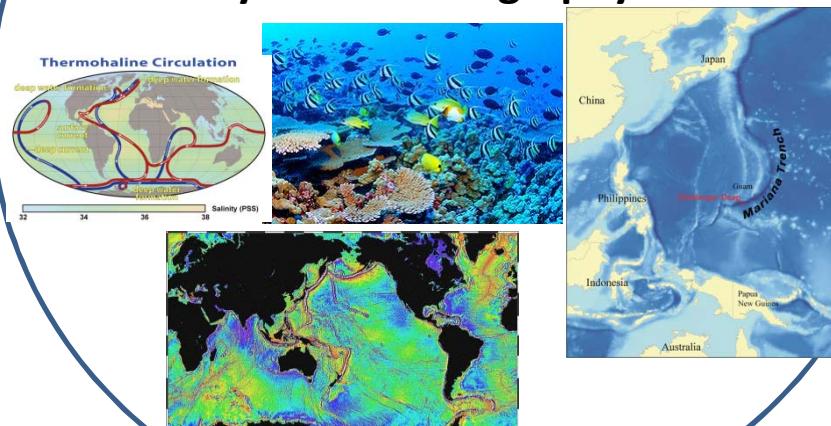
Model Results

Future

# We used to think about **Atmospheric Sciences** and **Oceanic Sciences** independently...



- Biological oceanography
- Chemical oceanography
- Geological oceanography
- Physical oceanography



Source: [www.wikipedia.org](http://www.wikipedia.org)



Introduction

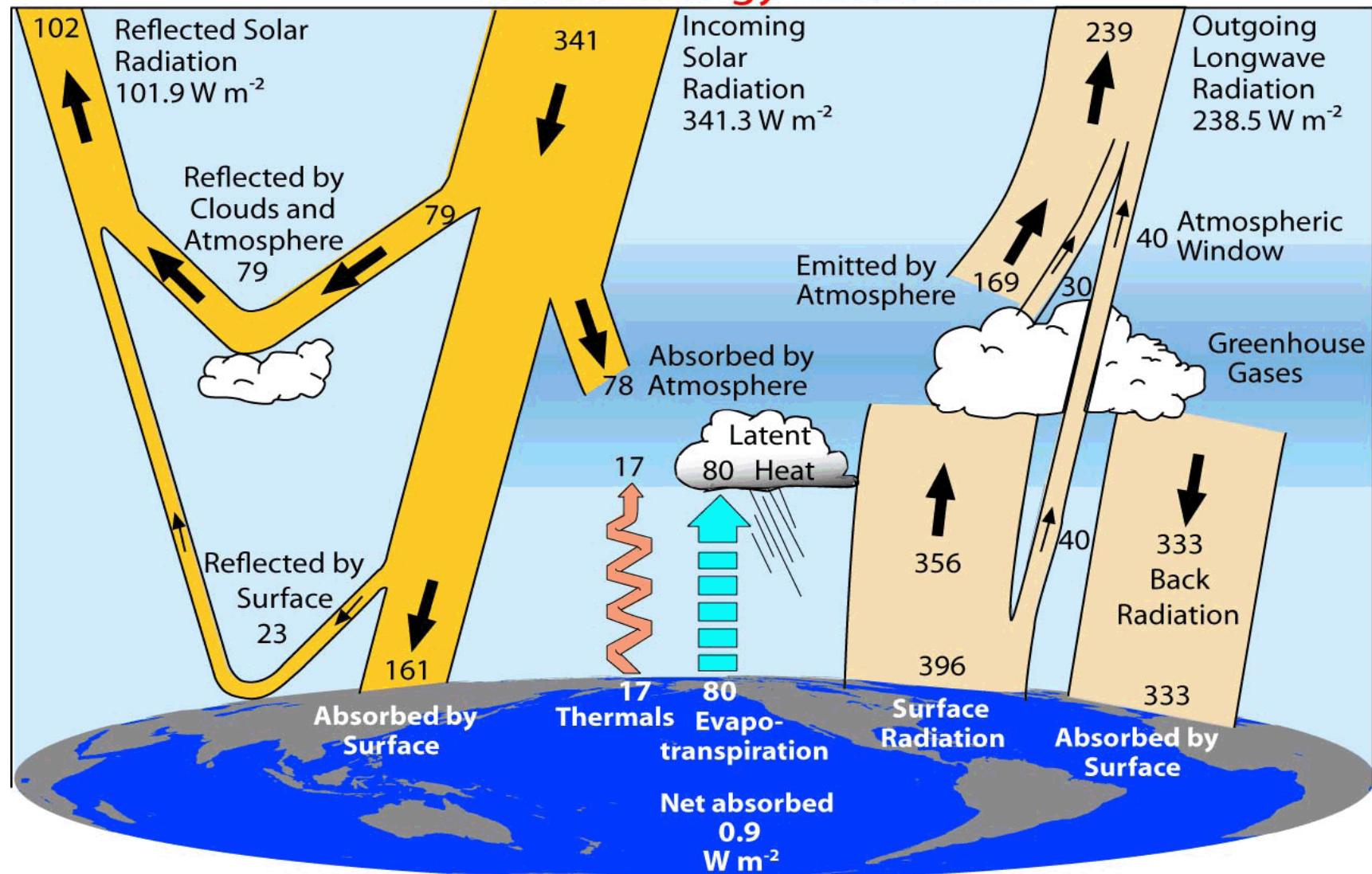
Gas emissions

POM emissions

Model Results

Future

## Global Energy Flows $\text{W m}^{-2}$



Introduction

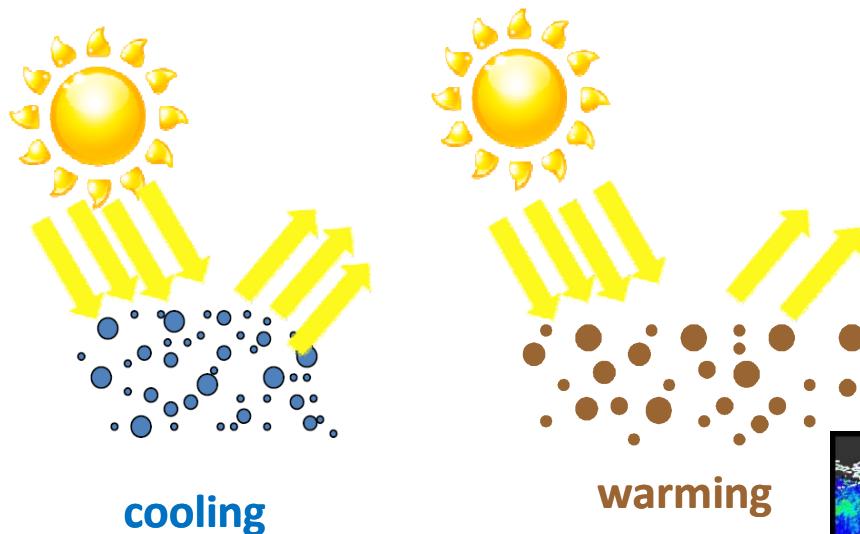
Gas emissions

POM emissions

Model Results

Future

# The Direct Radiative Effects of Aerosols



Aerosol Optical Depth (AOD)

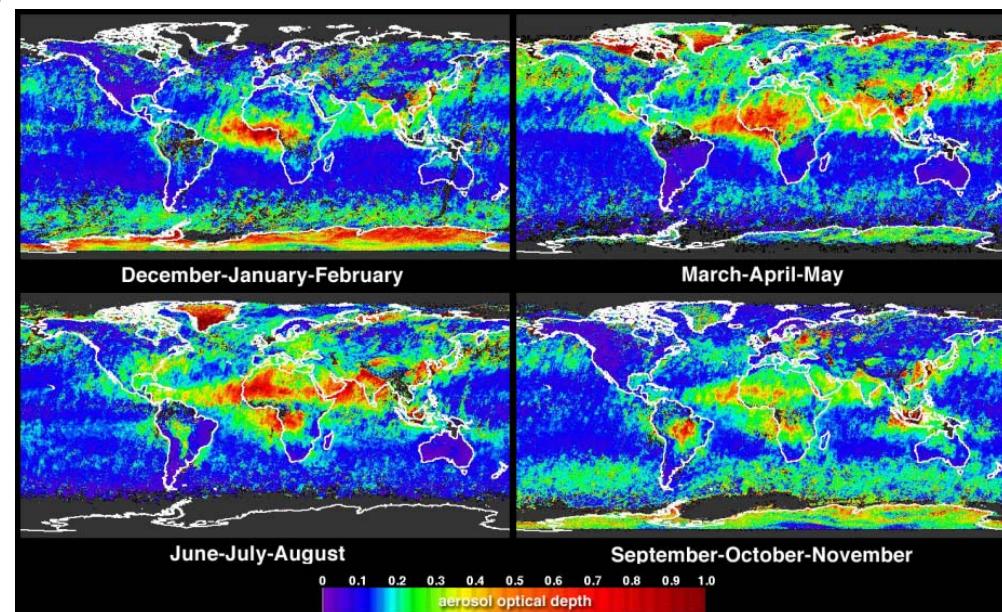


Image courtesy NASA/GSFC/LaRC/JPL, MISR Team  
([http://visibleearth.nasa.gov/view\\_rec.php?id=16472](http://visibleearth.nasa.gov/view_rec.php?id=16472))



Introduction

Gas emissions

POM emissions

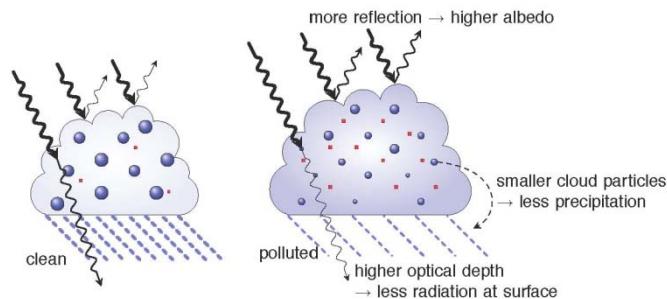
Model Results

Future

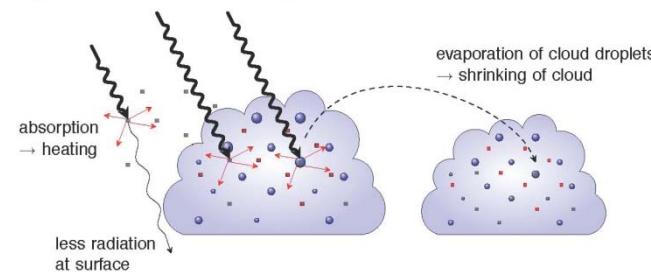
# The Indirect Radiative Effects of Aerosols

Ship tracks

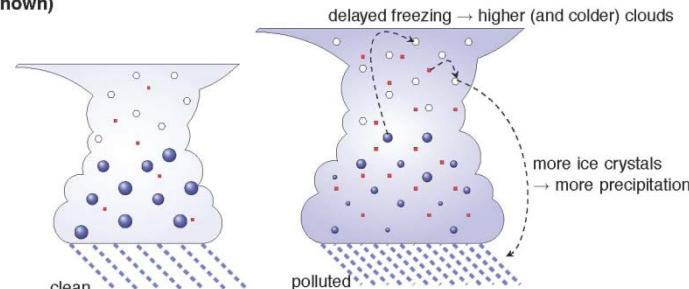
Cloud albedo and lifetime effect (negative radiative effect for warm clouds at TOA; less precipitation and less solar radiation at the surface)



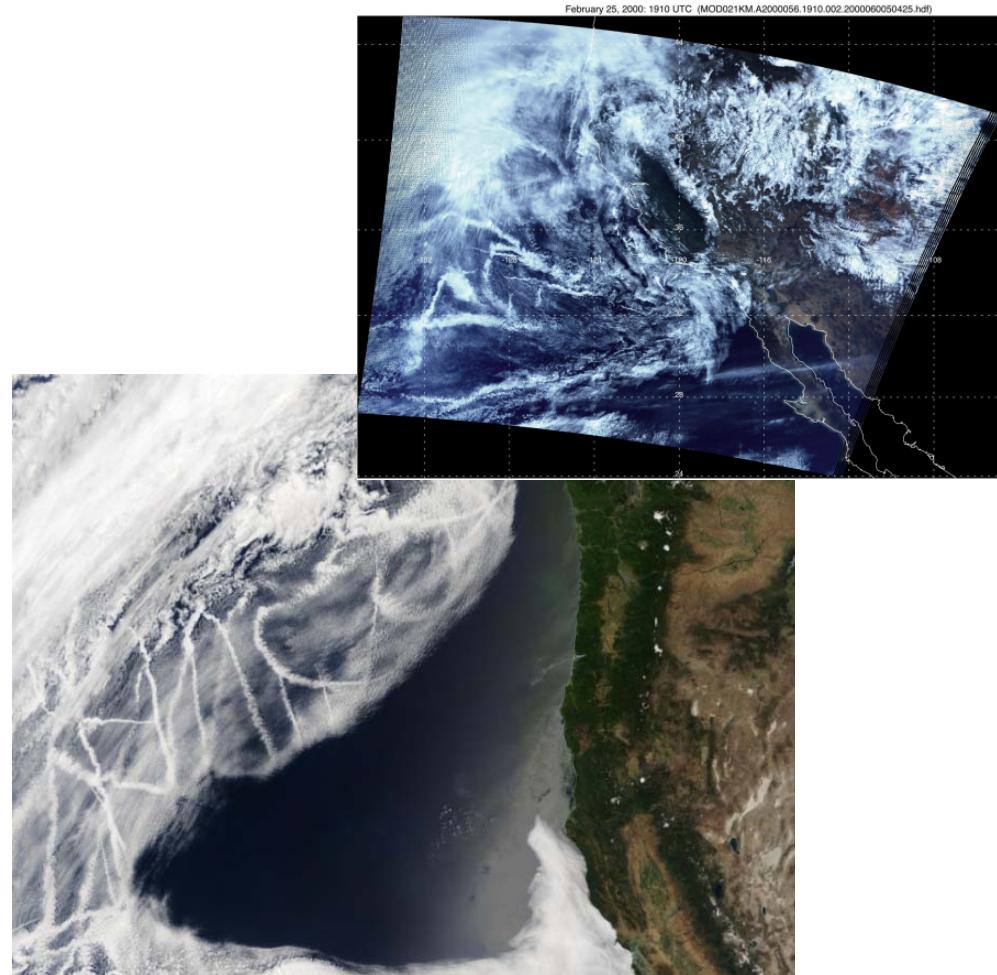
Semi-direct effect (positive radiative effect at TOA for soot inside clouds, negative for soot above clouds)



Glaciation effect (positive radiative effect at TOA and more precipitation), thermodynamic effect (sign of radiative effect and change in precipitation not yet known)



Source: Figure 7.20, Solomon et al., IPCC AR4 WG I, Climate Change 2007: The Physical Science Basis, 2007



Credit :MODIS Atmosphere Science Team  
[http://visibleearth.nasa.gov/view\\_rec.php?id=224](http://visibleearth.nasa.gov/view_rec.php?id=224)



Introduction

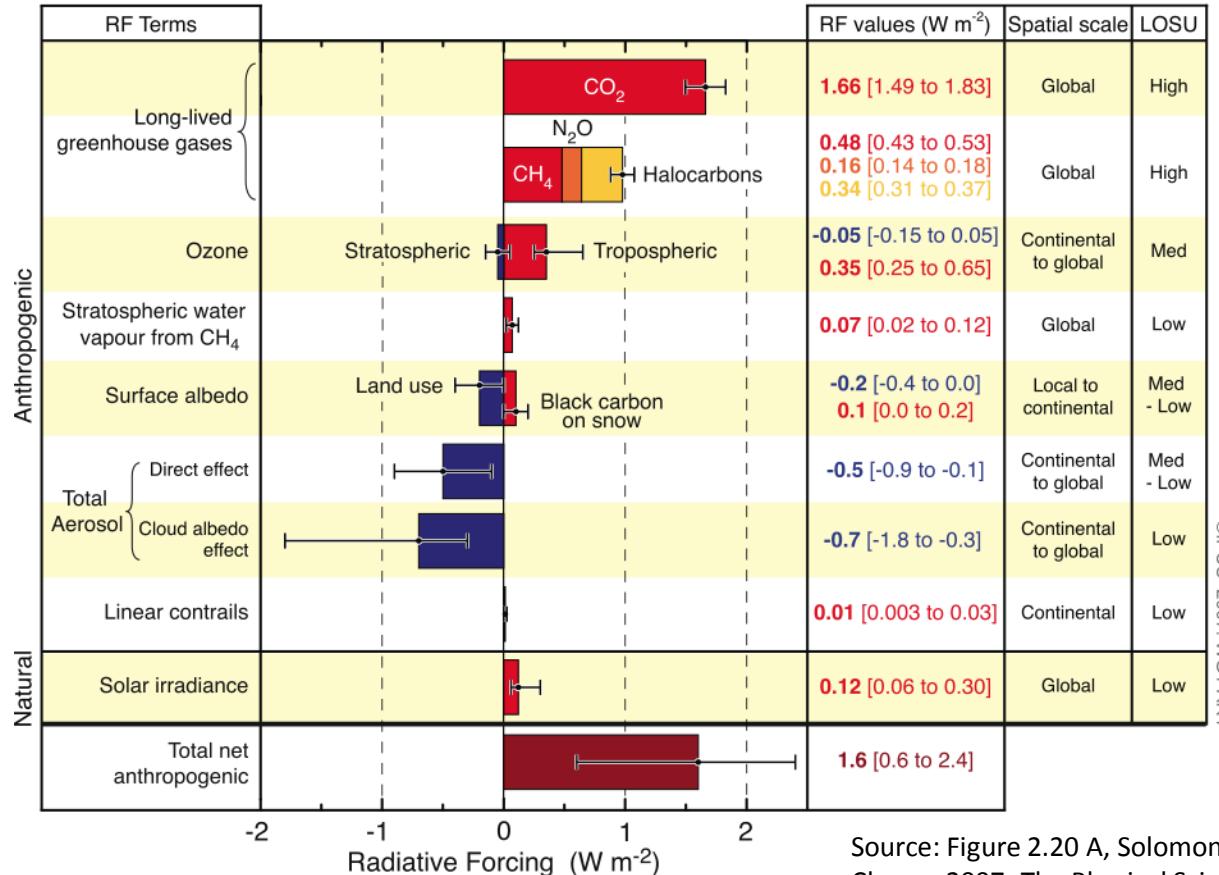
Gas emissions

POM emissions

Model Results

Future

# Global Average Radiative Forcing (RF) Estimates and Ranges



Source: Figure 2.20 A, Solomon et al., IPCC AR4 WG I, Climate Change 2007: The Physical Science Basis, 2007

➤ No marine aerosols, why?



Introduction

Gas emissions

POM emissions

Model Results

Future

# Why Do We Need Improved Quantification of Marine Aerosol?



- Anthropogenic activities seem to influence marine ecology

GEOGRAPHICAL RESEARCH LETTERS, VOL. 30, NO. 15, 1899, doi:10.1029/2003GL01681

**nature**

**LETTERS**

**Ocean primary production and climate: Global decadal changes**

Watson W. Gregg  
Laboratory for Hydrographic Processes, NASA-Goddard Space Flight Center, USA

Margarita E. Conkright  
Ocean Climate Laboratory, NOAA/National Oceanographic Data Center, USA

Paul Ginoux  
NOAA Geophysical Fluid Dynamics Laboratory, USA

John E. O'Reilly  
NOAA/National Marine Fisheries Service, USA

**Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and the inorganic carbon system**

Scott C. Doney<sup>a\*</sup>, Natalie Mahowald<sup>b</sup>, Ivan Lima<sup>a</sup>, Richard A. Feely<sup>c</sup>, Fred T. Mackenzie<sup>d</sup>, Jean-François Lamarre<sup>e</sup>, and Phil J. Rasch<sup>f</sup>

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**GSFC**

Vol 444 | 7 December 2006 | doi:10.1038/nature05317

**Human Impacts on the Global Marine Ecosystem**

Published by perrygeo at 9:06 am under Uncategorized, environment

We did it!

As some of you may know in 2005 through 2006, I was part of a research team at NCEAS, developing a global model of human impacts on the marine ecosystem or compiled 17 high-resolution global datasets of human-induced threats (land-based fishing, shipping, climate change, etc.) and 20 ocean habitat datasets. These were an impact index which models the cumulative level of human-induced stress on our oceans.

**Image 1**

**Goddard SPACE FLIGHT CENTER Yesterday's Vision, Tomorrow's Reality**

**Related Links**

**September 16, 2003** - (date of web publication)

**OCEAN PLANT LIFE SLOWS DOWN AND ABSORBS LESS CARBON**

Plant life in the world's oceans has become less productive since the early 1980s, absorbing less carbon dioxide from the atmosphere, which may in turn impact the Earth's carbon cycle, according to a study that combines NASA satellite data with NOAA

**Image 1**

The results were published today in *Science* magazine and presented yesterday at the AAAS Annual Meeting. To summarize, we found that the entire ocean is affected and 40% is heavily impacted. It is



Introduction

Gas emissions

POM emissions

Model Results

Future

# Why Do We Need Improved Quantification of Marine Aerosol?



- Modeling studies do not always consider proper quantification of natural/background aerosols
- Pristine conditions small changes can be important
- Large fraction of the Earth is covered by the oceans
- Prescribing lower bounds of CDNC in GCMs could introduce up to **80%** uncertainty in AIE (Lohmann et al., 2001; Ghan et al., 2001; Wang and Penner, 2008; Hoose et al., 2009)
- Model predicted extent of human-induced climate change



Introduction

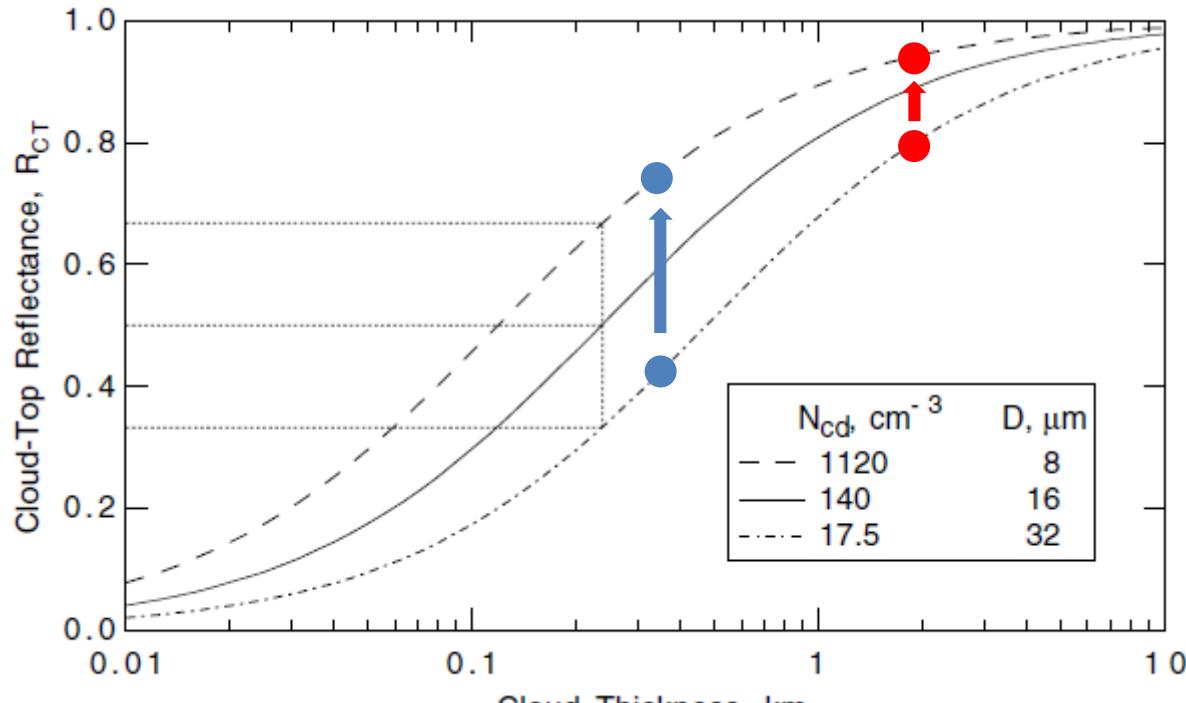
Gas emissions

POM emissions

Model Results

Future

# Cloud Susceptibility



[Source: Schwartz and Slingo, 1996]

For a nonabsorbing, horizontally homogeneous cloud the cloud albedo can be parameterized in terms of cloud optical thickness

A definition of cloud susceptibility has been proposed as a measure of the extent of CCN influence on cloud albedo.

albedo

$$\alpha_c = \frac{\tau}{7.7 + \tau}$$

Optical thickness

$$S = \frac{\partial \alpha_c}{\partial N_{d,\text{col\_avg}}} = \frac{\alpha_c(1 - \alpha_c)}{3N_{d,\text{col\_avg}}}$$



Introduction

Gas emissions

POM emissions

Model Results

Future

# Improved Quantification of Sea Spray



- Modeling studies do not always consider proper quantification of natural/background aerosols
- Pristine conditions V small changes can be important
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- **Model predicted extent of human-induced climate change**



Introduction



Gas emissions



POM emissions



Model Results

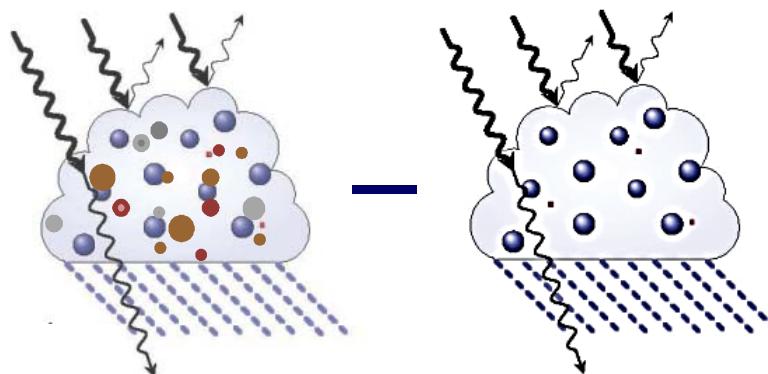


Future

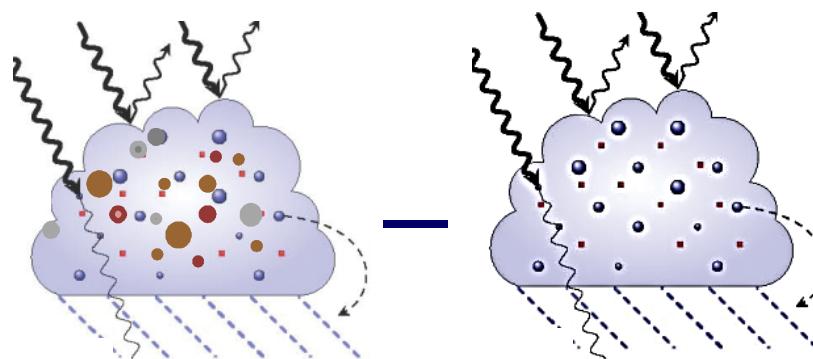
# Improved Quantification of Sea Spray



“Background” CCN/CDNC  $\sim$  10 to 20 cm $^{-3}$



“Background CCN/CDNC  $\sim$  30 to 40 cm $^{-3}$



Introduction

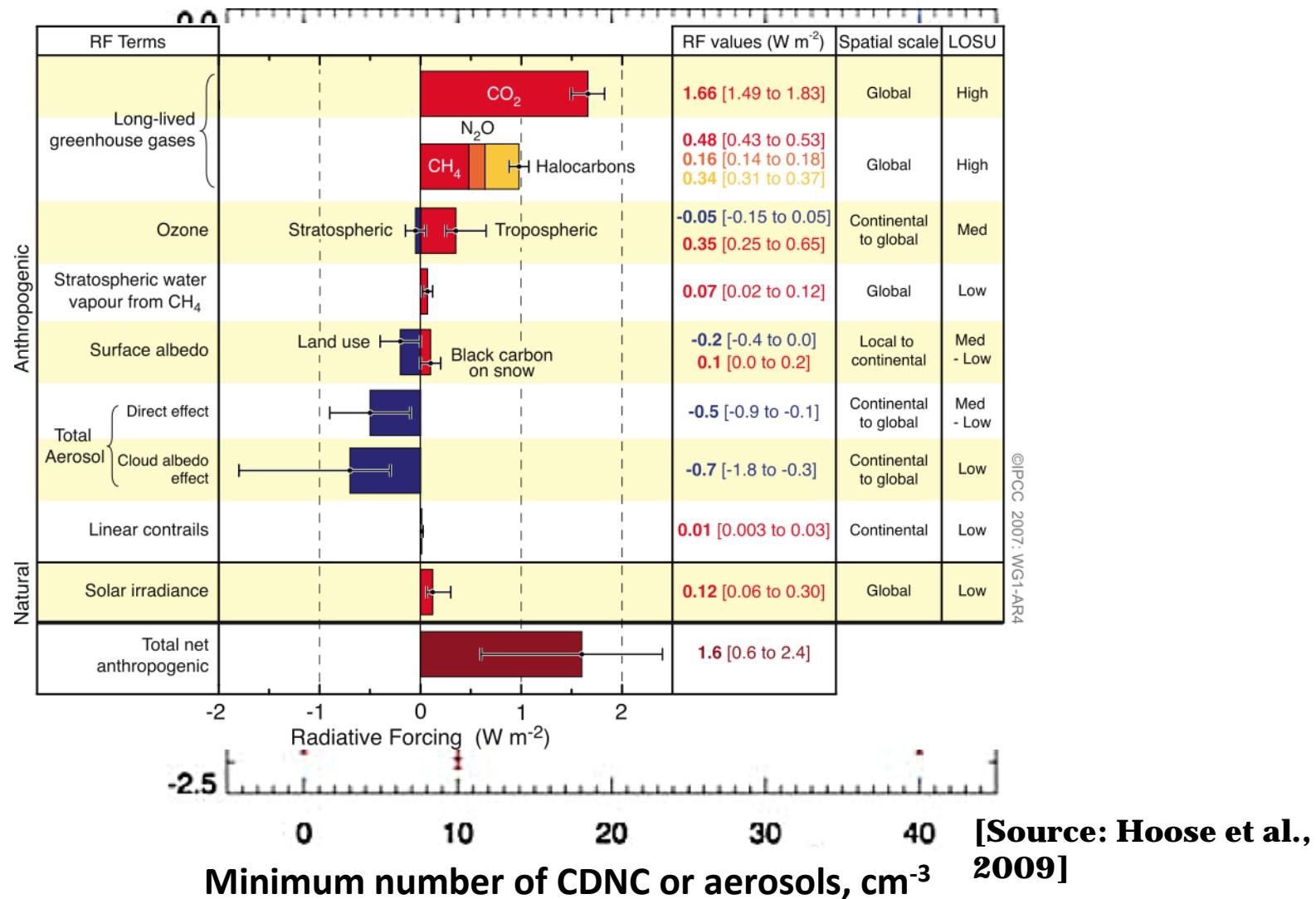
Gas emissions

POM emissions

Model Results

Future

# Difference in Short-wave Cloud Forcing (SWCF)



Introduction

Gas emissions

POM emissions

Model Results

Future

# Difference in Short-wave Cloud Forcing (SWCF)

- Improving *Ocean-Aerosol Interaction* will go a long way toward narrowing this large uncertainty
- Will also help reconcile the differences between the model predictions and satellite estimates/inverse calculations



Introduction

Gas emissions

POM emissions

Model Results

Future

# Terminology



**Chlorophyll - a: an index of phytoplankton biomass**

- The main photosynthetic pigment
- Present in all phytoplankton
- Present only in phytoplankton

Units : mg m<sup>-3</sup> ( $\equiv \mu\text{g L}^{-1}$ )

**Measurement methods:**

- ✓ Remote sensing (Chlorophyll a absorbs well at a wavelength of about 400-450 nm and at 650-700 nm)
- Colorimetric
- High Performance Liquid Chromatography
- Fluorometric



Introduction

Gas emissions

POM emissions

Model Results

Future

# Factors Controlling Primary Production



## External

- Nutrients
- Light
- Grazing pressure
- Temperature

## Internal

- Pigments
- Cell size
- Enzyme concentration
- Nutrient pool



➤ Ocean Net Primary Production (NPP) contributing roughly half of the biosphere's NPP!



Introduction

Gas emissions

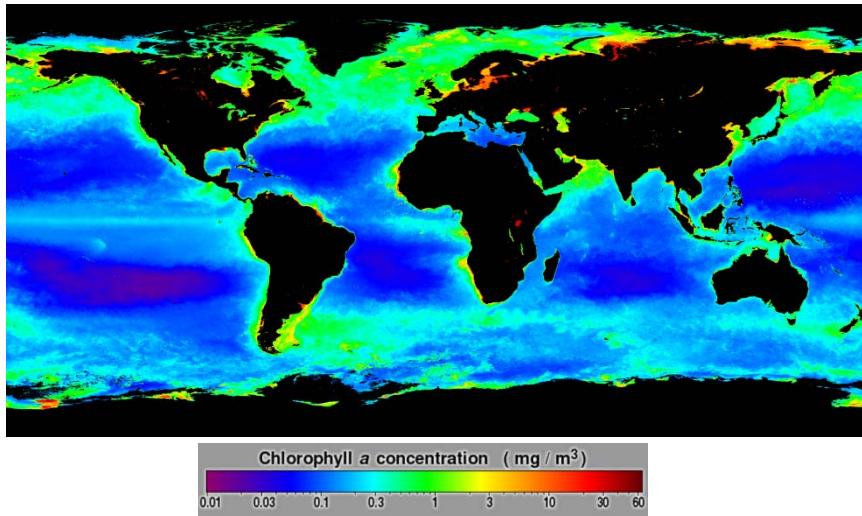
POM emissions

Model Results

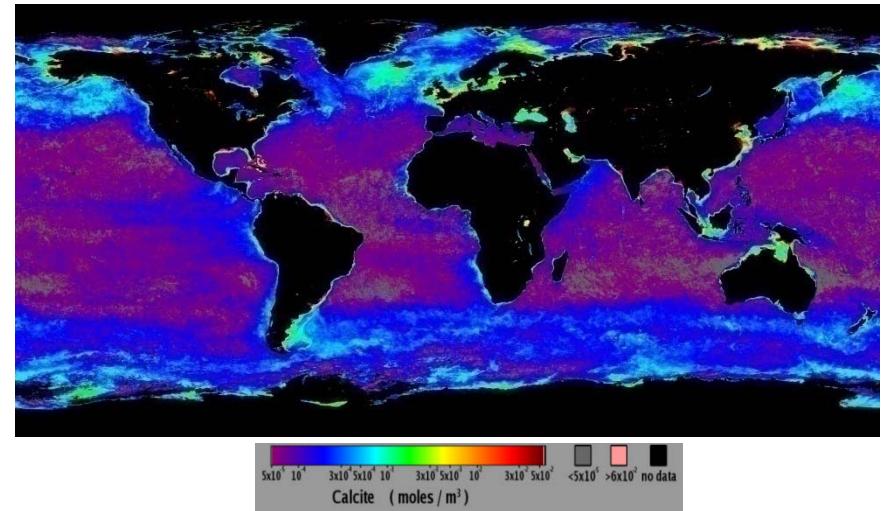
Future

# Phytoplankton Functional Groups

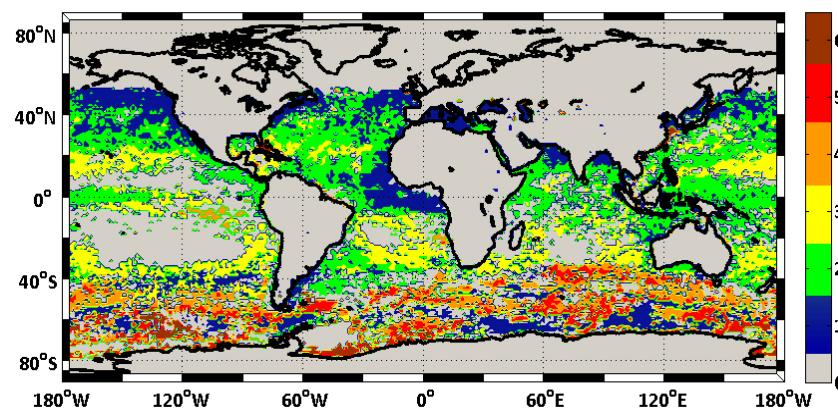
Aqua MODIS [Chl a] (year 2008)



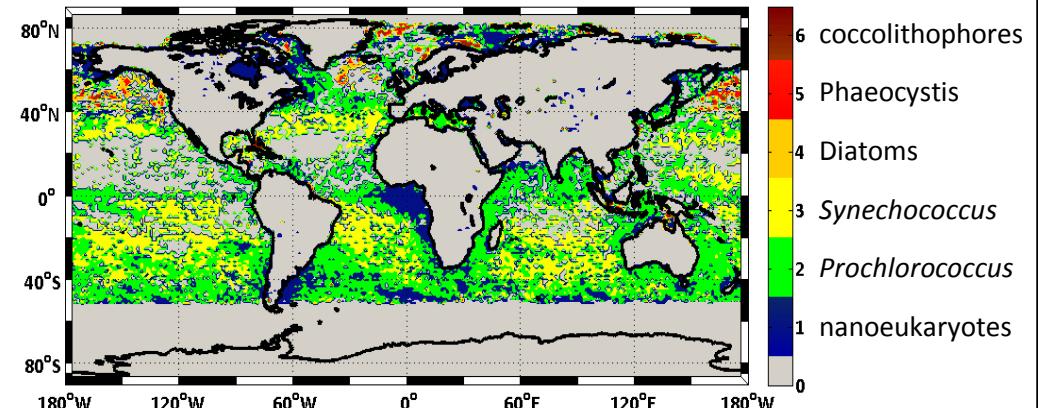
Aqua MODIS Calcite (year 2008)



January



July



Maps are created after Alvain, S., C. Moulin, Y. Dandonneau, and H. Loisel (2008), Seasonal distribution and succession of dominant phytoplankton groups in the global ocean: A satellite view, Global Biogeochem. Cycles, 22, GB3001.



Introduction

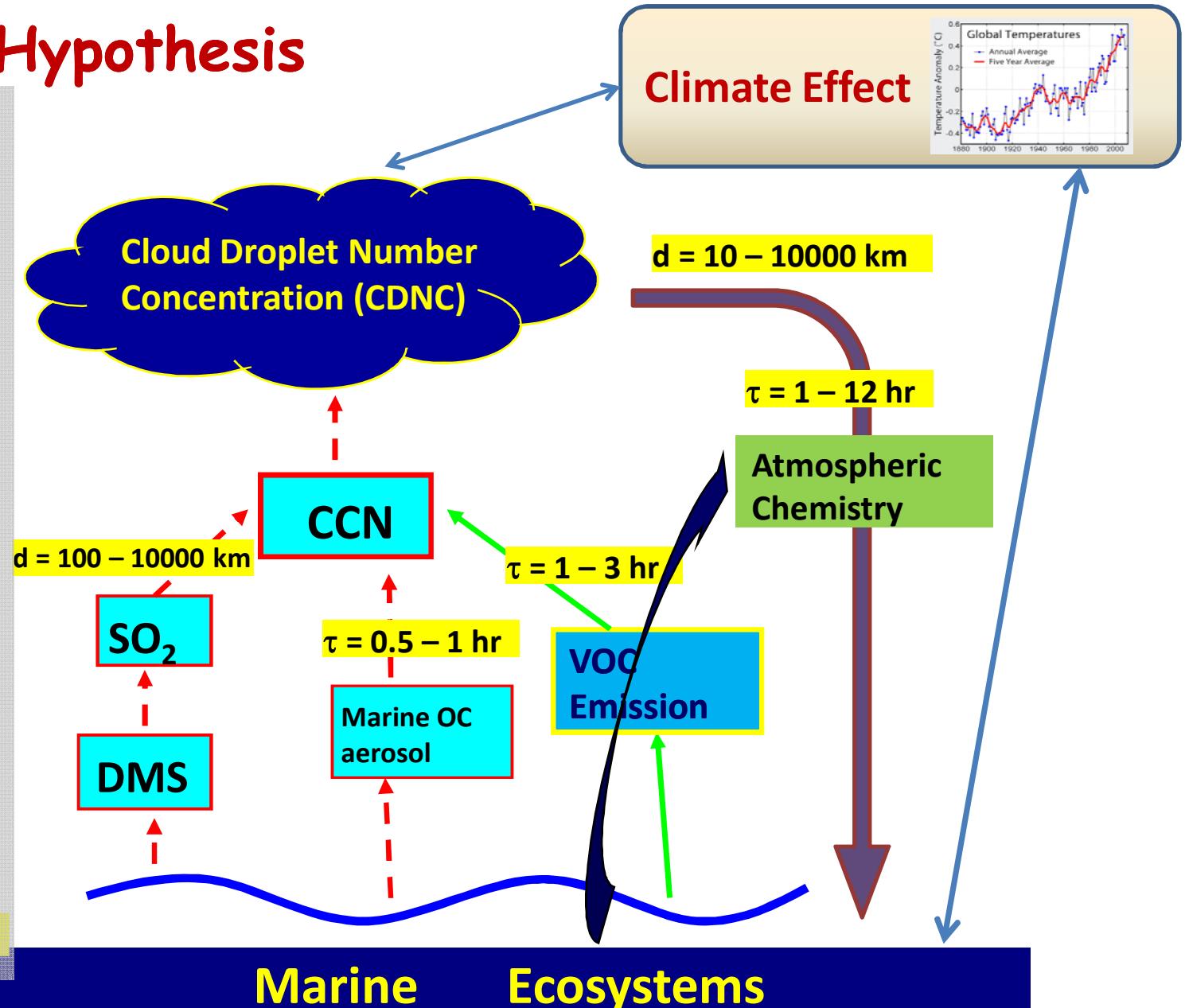
Gas emissions

POM emissions

Model Results

Future

# CLAW Hypothesis



Introduction

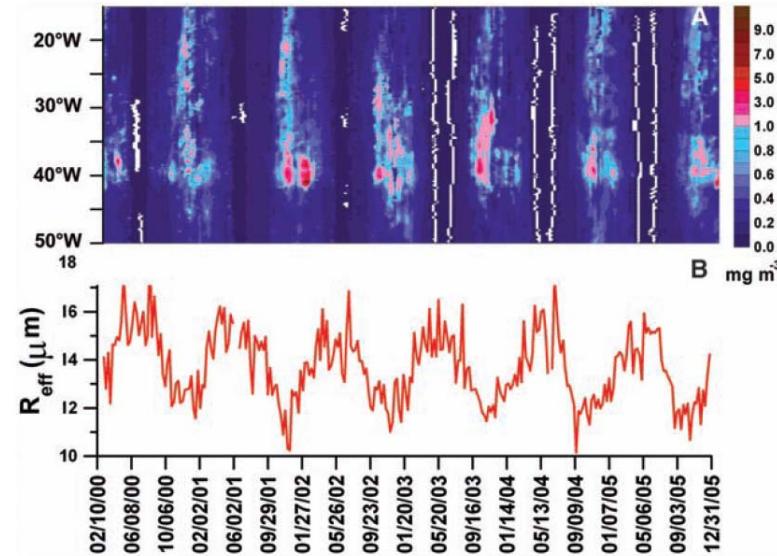
Gas emissions

POM emissions

Model Results

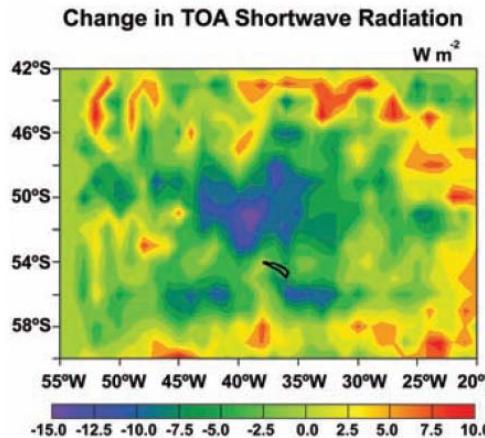
Future

# Extended CLAW



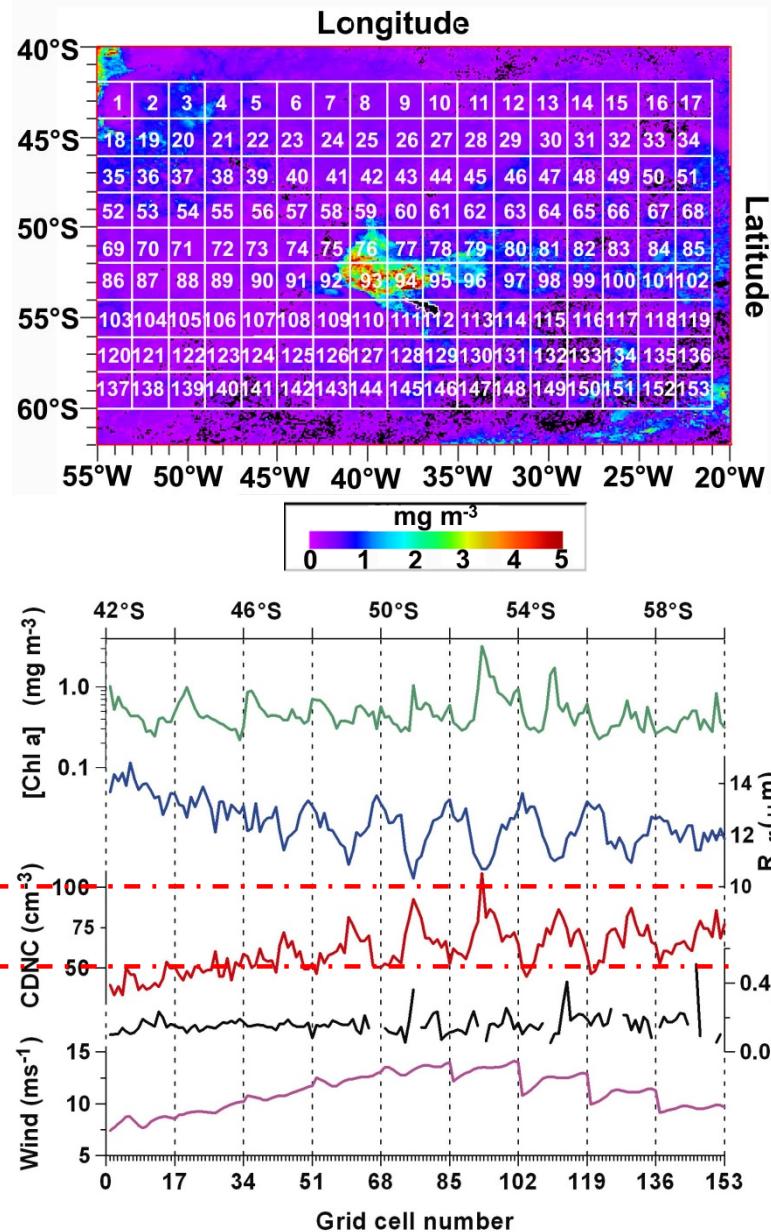
**Fig. 1.** The 8-day averaged (A) SeaWiFS-observed chlorophyll *a* and (B) MODIS-retrieved cloud effective radius. Data for [Chl *a*] is gridded at a resolution of 9 by 9 km and zonally averaged between 49°S and 54°S; data for  $R_{\text{eff}}$  is gridded at a resolution of 1° by 1° and averaged in the area of 49° to 54°S and 35° to 41°W. White areas in (A) indicate missing data.

SOCEX II - polluted



SOCEX II - "background"

**Fig. 3.** Change in TOA short-wave radiation. The radiative effect was evaluated for the change in albedo of warm marine clouds. Calculations are carried out using monthly averaged MODIS-observed data at 1° by 1° resolution and the GMI-supplied monthly averaged solar flux at 4° by 5° resolution.



Source: Meskhidze, N. and A. Nenes, Phytoplankton and Cloudiness in the Southern Ocean, Science, Vol 314, 2006.



Introduction

Gas emissions

POM emissions

Model Results

Future

# Outline



- 1) Introduction
- 2) Biogenic gas emissions from the ocean - impact on aerosol/cloud interaction and radiative properties of the overlying atmosphere
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- 5) The future



Introduction

Gas emissions

POM emissions

Model Results

Future

# Biogenic Gas Emissions From the Ocean

## Basic Concepts

$$F = K \Delta C$$

Gas flux ( $F$ ) across the interface is driven by a concentration difference ( $\Delta C$ ) between air and surface water.

$K$  is a proportionality constant known as gas transfer velocity or transfer coefficient or a piston transfer velocity (cm h<sup>-1</sup>)

$$\Delta C = \frac{C_a}{H} - C_w$$

Where  $C_a$  and  $C_w$  are the gas concentrations in air and water, respectively and  $H$  is Henry's Law constant

$$H = \frac{p_g}{C_w}$$

Tables: <http://www.mpch-mainz.mpg.de/~sander/res/henry.html>

Ratio of the gas in air to its concentration in unionized form dissolved in water



Introduction

Gas emissions

POM emissions

Model Results

Future

## Basic Concepts (continued)

It is often convenient to think in terms of reciprocal of the transfer velocity – a measure of the resistance to interfacial gas exchange

$$\frac{1}{K} = \frac{1}{\alpha k_w} + \frac{1}{H \cdot k_a}$$

$\alpha$  – is a factor accounting for the chemical enhancement of gas exchange due to gas-aqueous rxns

For trace gases which are chemically “unreactive” in the aqueous-phase (DMS, CH<sub>4</sub>, N<sub>2</sub>O, CO<sub>2</sub>, inert gases)  $\alpha=1$

For highly aqueous-phase gases (SO<sub>2</sub>, NH<sub>3</sub>, HCl)  $\alpha \approx 10^3$

$$R = r_w + r_a$$



Introduction

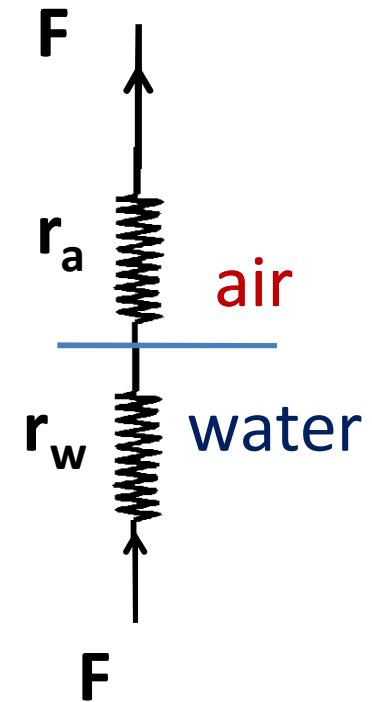
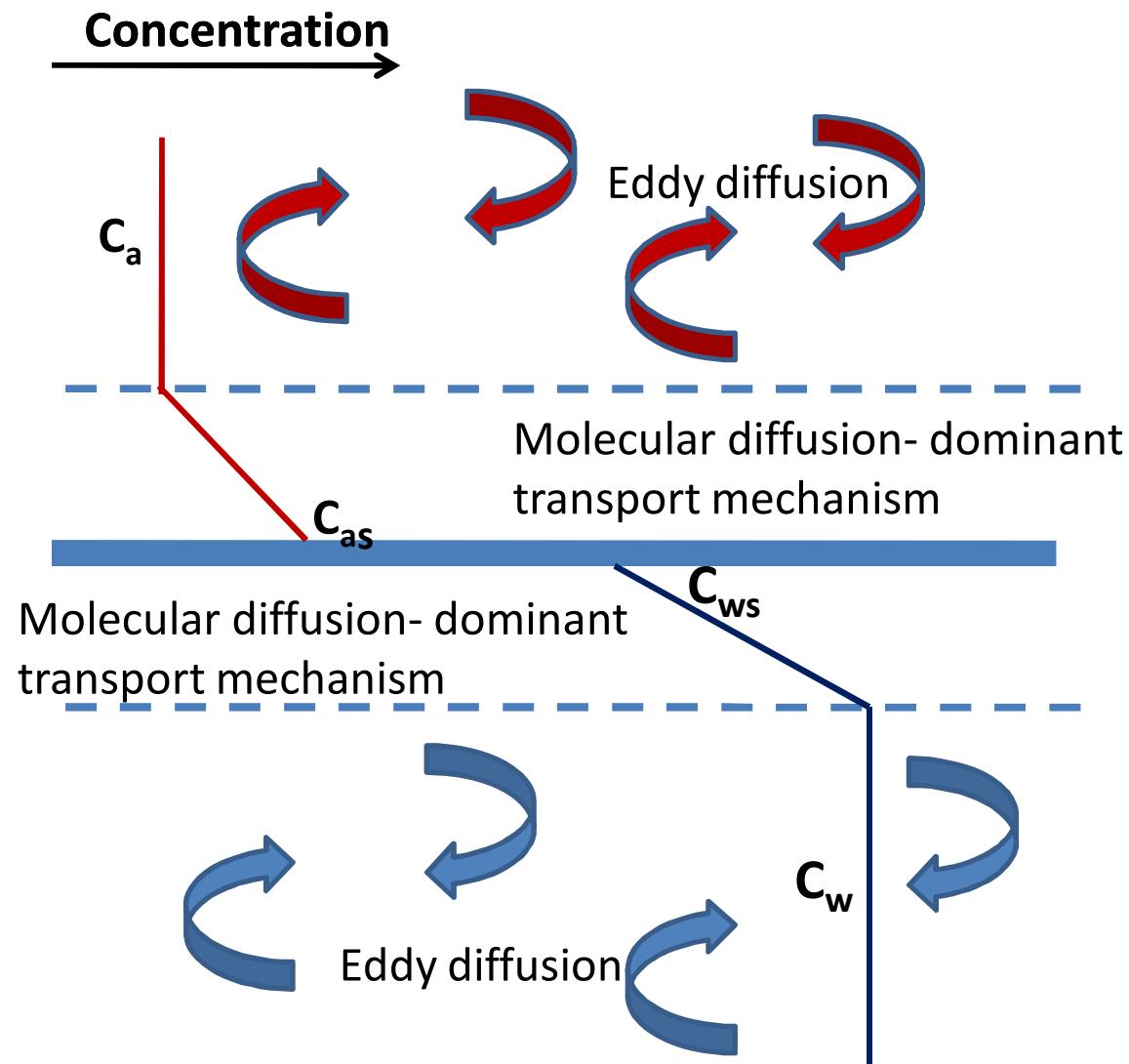
Gas emissions

POM emissions

Model Results

Future

# Two Film Model



Introduction

Gas emissions

POM emissions

Model Results

Future

## Basic Concepts (continued)

$$k_a = \frac{F}{(C_a - C_{as})}$$

$$k_w = \frac{F}{(C_w - C_{ws})}$$

For aqueous-phase reactive gases  $r_a \gg r_w \Rightarrow$  bulk of the resistance to the gas transfer is in the air-phase (i.e.,  $k_w \ll k_a$  and  $K \sim k_a$ ).

For “chemically unreactive” gases  $r_w \gg r_a \Rightarrow$  all of the resistance to the gas transfer is in water phase (i.e.,  $k_w \gg k_a$  and  $K \sim k_w$ )



$$C_w - C_{ws} = C_w - \beta C_a$$

$\beta$  – Bunsen solubility coefficient, v/v atm<sup>-1</sup>

$$F = K(C_w - \beta C_a)$$



Introduction

Gas emissions

POM emissions

Model Results

Future

# Measurement Techniques

1. Measurement of  $F$  in the air above the sea surface and  $\Delta C$  in the water to determine  $K$ . These methods are referred to as direct flux measurements or micrometeorological approaches and include covariance (or eddy correlation), eddy accumulation, atmospheric concentration profile, and inertial dissipation techniques.
2. Measurement of  $C_a$  (in air) and the change in  $C_w$  (in water) as a function of time. Assuming the water volume and surface area are known,  $F$  is then equal to the change in  $C_w$  multiplied by the ratio of the volume to the surface area. Then,  $K$  can be calculated with  $C_a$  and  $C_w$ . These bulk concentration techniques include mass-balance and perturbation studies where the concentration of gases in air and water are out of equilibrium through biological consumption/production, water heating/cooling ( $N_2$ ,  $O_2$ ,  $CO_2$ , noble gases), radioactive decay ( $^{222}Rn$ ), or by purposeful addition ( $^3He$ ,  $SF_6$ ).
3. Proxy techniques where a nongaseous tracer whose air-sea flux is more easily measured is used as a surrogate for a gas using the principle that all air-water transfer is controlled by the near surface hydrodynamics. In field applications, proxy methods are limited to thermographic methods that use heat.

Wanninkhof, R. et al., Advances in Quantifying Air-Sea Gas Exchange and Environmental Forcing, Annu. Rev. Mar. Sci. 2009. 1:213–44,  
10.1146/annurev.marine.010908.163742



Introduction

Gas emissions

POM emissions

Model Results

Future

# Basic Concepts - Schmidt number

$$\text{Schmidt number } (Sc) = \frac{\text{Kinematic viscosity of seawater}}{\text{Gas diffusivity}}$$

If  $K$  is known for any given gas, its value may be inferred for any other gas

Gas transfer velocities are typically normalized to  $Sc_{600}$  (the values for CO<sub>2</sub> in freshwater and seawater at 20 °C)

$$\frac{K_1}{K_2} = \left( \frac{Sc_2}{Sc_1} \right)^n$$

$n=-1$  for the thin film model

Value of  $n$  changes from 0 to -1.22 in different publications



Introduction

Gas emissions

POM emissions

Model Results

Future

# Variables Affecting K



1. Wind speed
2. Fetch
3. Breaking waves and bubbles
4. Surfactant films
5. Chemical enhancement of gas exchange
6. Temperature and humidity gradients
7. Rain



Introduction

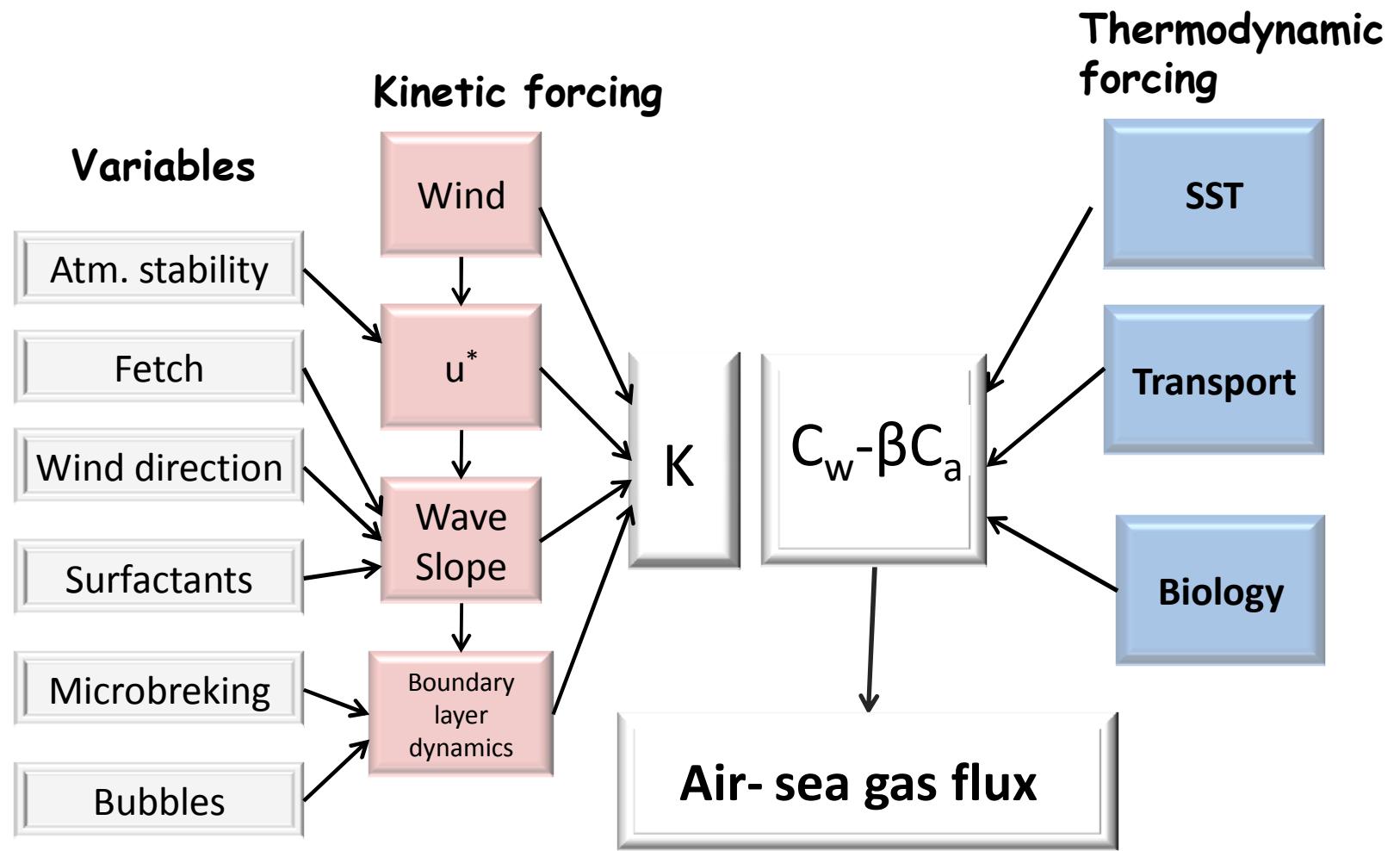
Gas emissions

POM emissions

Model Results

Future

# Factors Affecting Air-sea Fluxes of Weakly Soluble, Non-reactive Gases



Wanninkhof, R. et al., Advances in Quantifying Air-Sea Gas Exchange and Environmental Forcing, Annu. Rev. Mar. Sci. 2009. 1:213–44, 10.1146/annurev.marine.010908.163742



Introduction

Gas emissions

POM emissions

Model Results

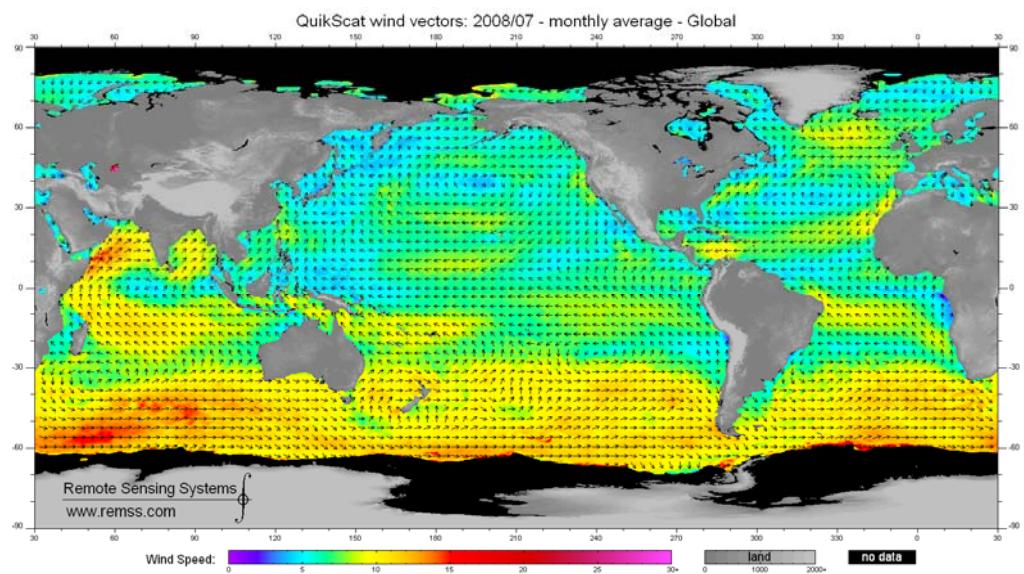
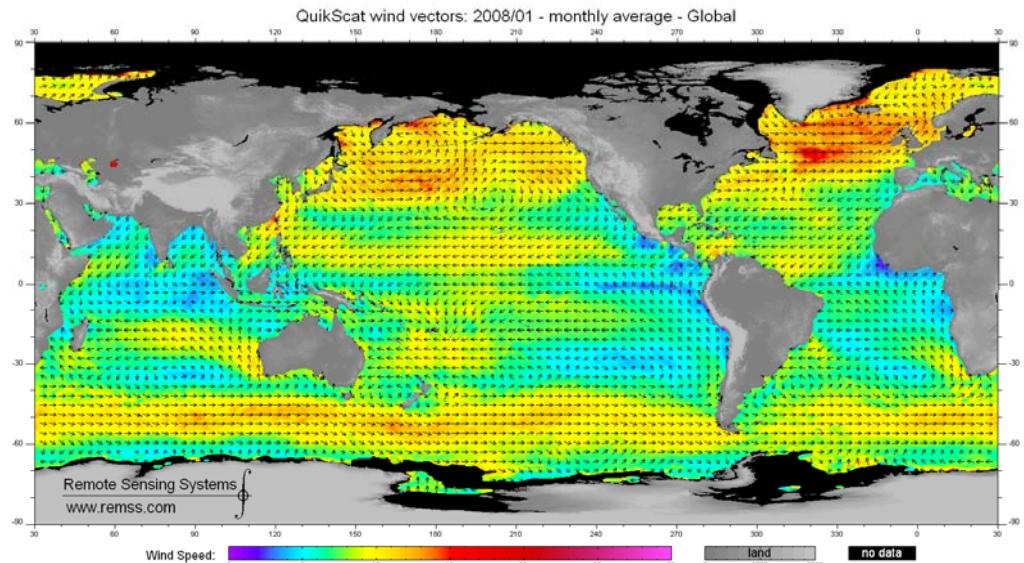
Future

# Gas Exchange - Wind Speed Relationships

Relationship	Equation
Liss and Merlivat, 1986	$K=0.17 \cdot U_{10} \cdot (600/Sc)^{0.67}$ ( $U_{10} < 3.6 \text{ m s}^{-1}$ ) $K=(2.85 \cdot U_{10} - 9.65) \cdot (600/Sc)^{0.5}$ ( $3.6 \text{ ms}^{-1} < U_{10} < 13 \text{ ms}^{-1}$ ) $K=(5.9 \cdot U_{10} - 49.3) \cdot (600/Sc)^{0.5}$ ( $U_{10} > 13 \text{ ms}^{-1}$ )
Wanninkhof, 1992	$K=0.39 (U_{10\text{ave}})^2 (660/Sc)^{0.5}$ (long term average winds) $K=0.31 (U_{10})^2 (660/Sc)^{0.5}$ (instantaneous wind speeds)
Nightingale et al., 2000	$K=0.333(U_{10})^2 + (600/Sc)^{0.5}$



# SeaWinds on QuikSCAT



<http://www.remss.com/qscat/qscat Browse.html>



<http://winds.jpl.nasa.gov/missions/quikscat/index.cfm>

Wind speed at 10 meters over  
the ocean surface ( $U_{10}$ )



Introduction

Gas emissions

POM emissions

Model Results

Future

## We Need a “Couple” of Other Parameters...

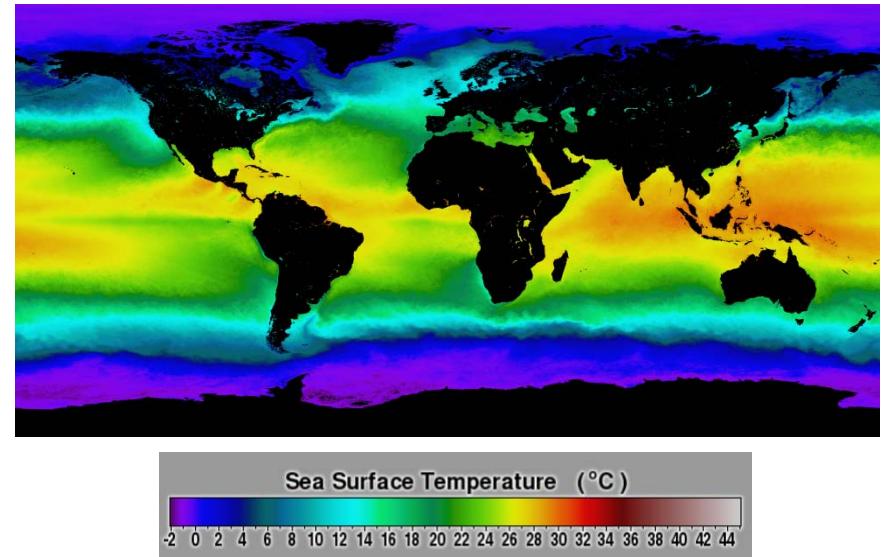
$$Sc = 3875.21 - 180.80T_c + 2.69T_c^2$$

Where  $T_c$  is sea-surface temperature in ( $^{\circ}\text{C}$ ) [Moore and Groszko, 1999]

Assume  $C_a \approx 0$

$$P = C_w \left( K - \sum_i K_{Chem,i} C_{xi} - K_{Biol} \right)$$

MODIS sea-surface temperature



Mean lifetime against rx with OH and  $\text{O}_2$  in seawater = 19 and 115 days

Mean lifetime against bacterial consumption 17 days

Palmer, P. I. and Shaw, S. L.: Quantifying global marine isoprene fluxes using MODIS chlorophyll observations, J. Geophys. Res., 32, L09805, doi:10.1029/2005GL022592, 2005.



Introduction

Gas emissions

POM emissions

Model Results

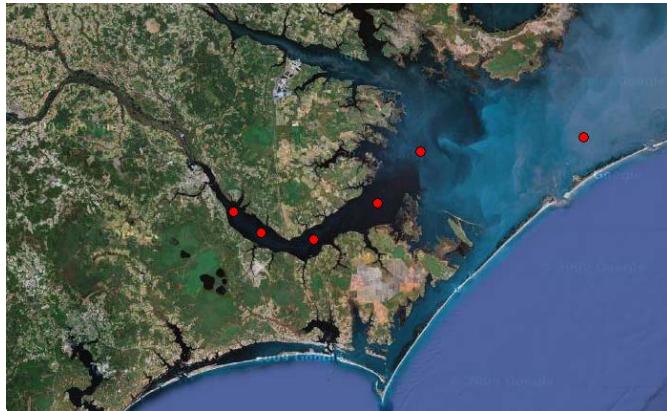
Future

# Light And Species Dependent Production Rates Of Marine Isoprene And Monoterpenes

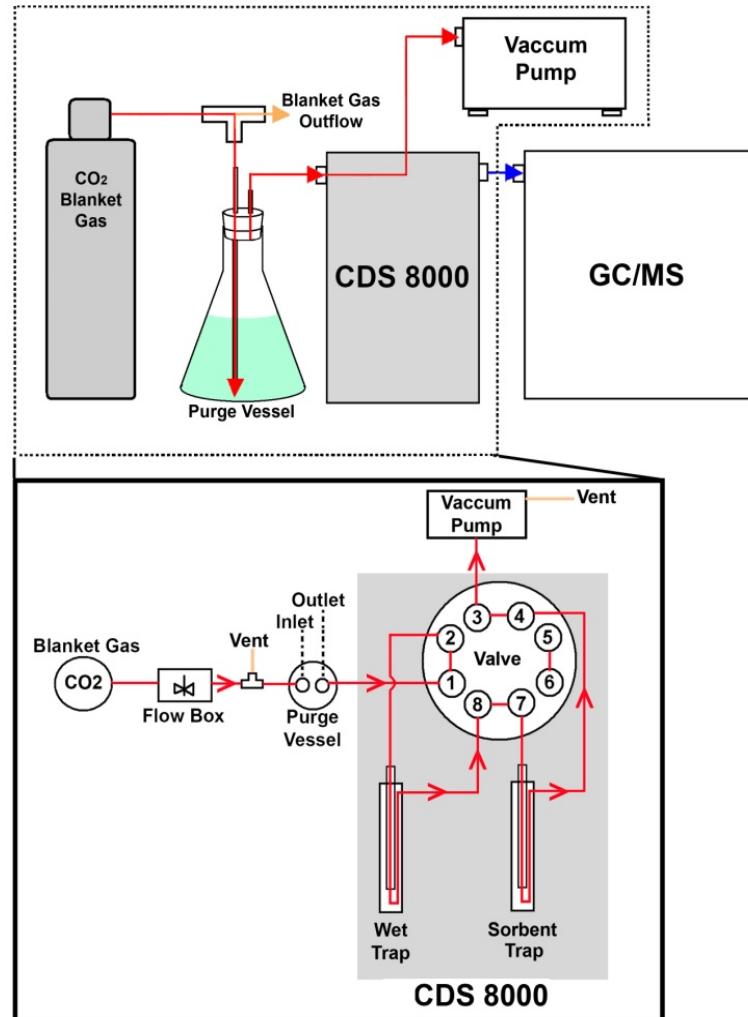
Laboratory grown phytoplankton monocultures: diatoms *Thalassiosira weissflogii* (*T. weiss.*) (CCMP 1336) and *Thalassiosira pseudonana* (*T. pseud.*) (CCMP 1335), prymnesiophyte strains- *Pleurochrysis carterae* (*P. carter.*) (CCMP 645); dinoflagellate strains- *Karenia brevis* (*K. brevis*) (CCMP 718, CCMP 2229) and *Prorocentrum minimum* (*P. minim.*) (CCMP 1329); cryptophyte strains- *Rhodomonas salina* (*R. salina*) (UTEX 2423)



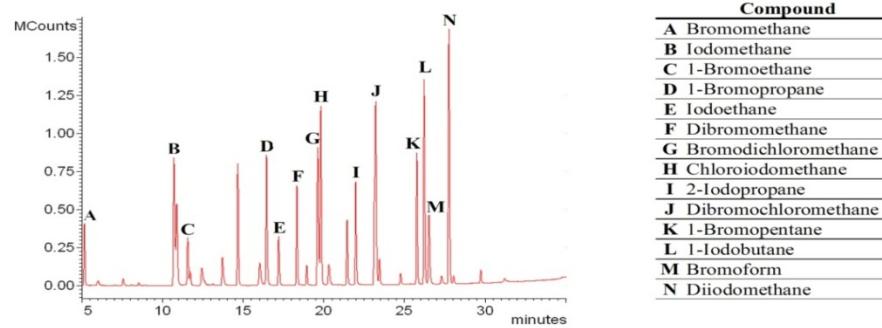
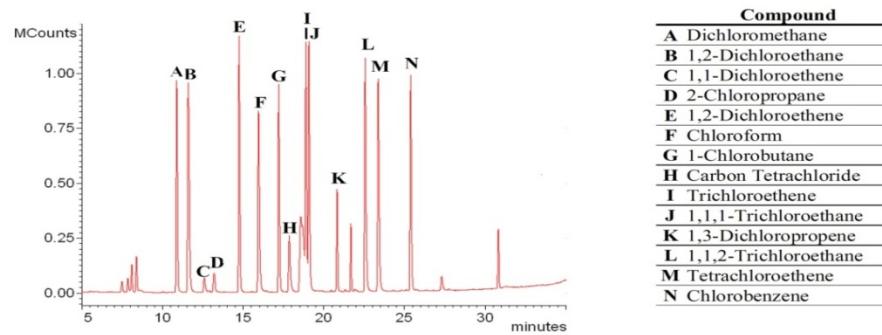
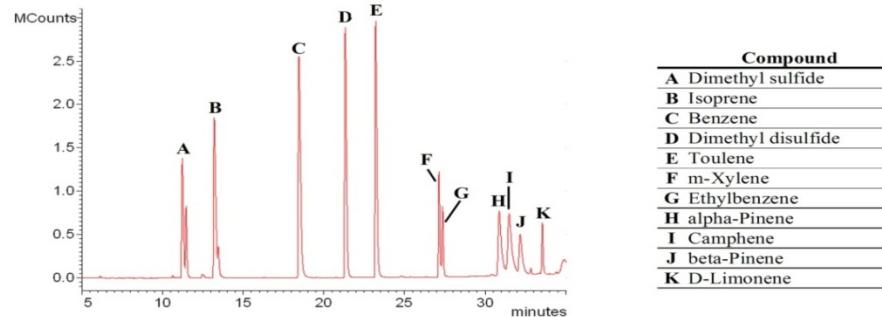
# Light And Species Dependent Production Rates Of Marine Isoprene And Monoterpenes



# Instrumentation Setup



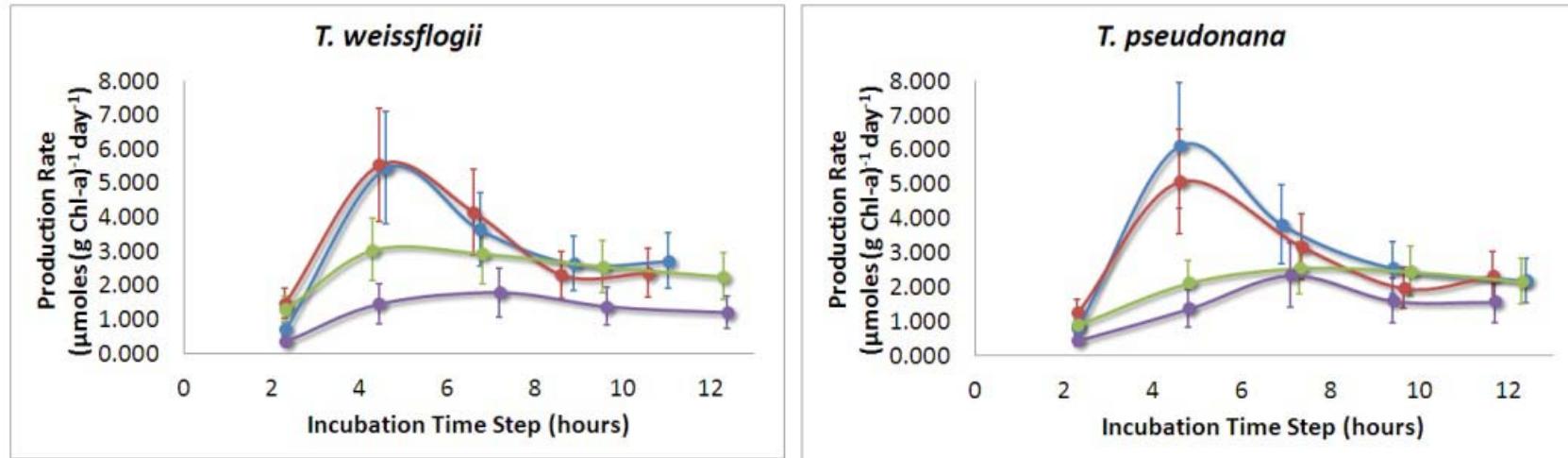
# Sample chromatographs for 38 BVOC



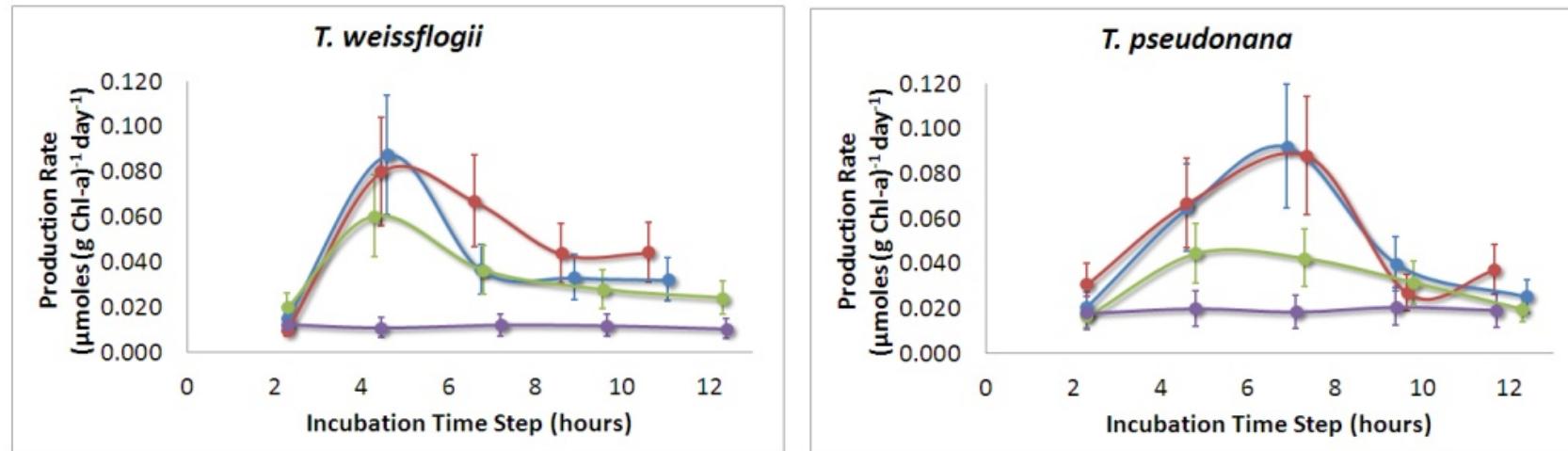
- We measured 38 different VOCs emitted from phytoplankton including  $\alpha$ - and  $\beta$ -pinene, d-limonene, camphene, halocarbons...

# Results Of Light Experiments

## Stress induced isoprene production as a function of time



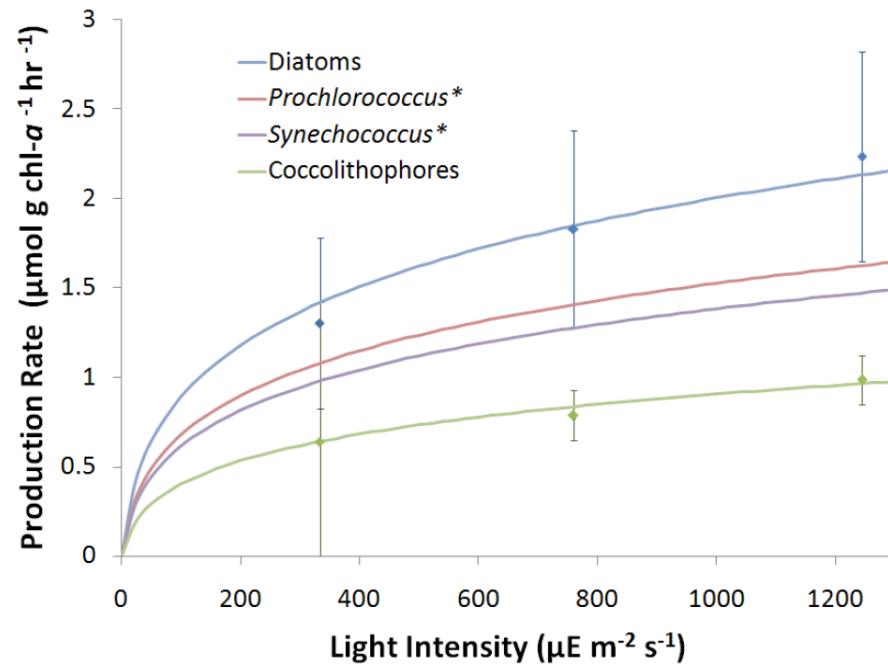
## Stress induced $\alpha$ -pinene production as a function of time



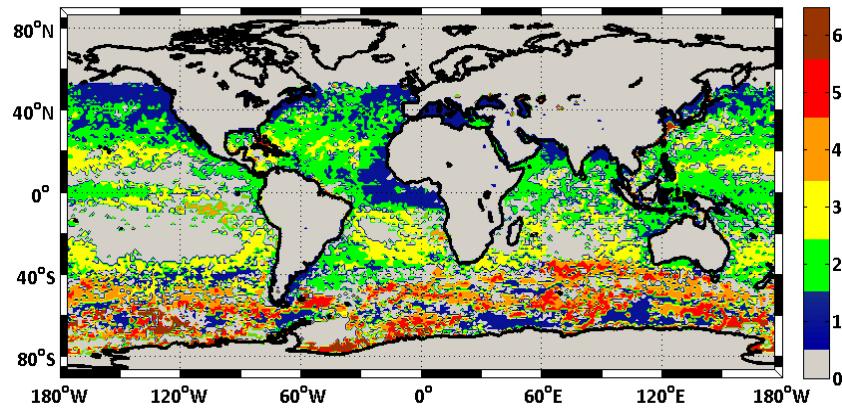
— 90  $\mu\text{E m}^{-2} \text{s}^{-1}$  — 150  $\mu\text{E m}^{-2} \text{s}^{-1}$  — 420  $\mu\text{E m}^{-2} \text{s}^{-1}$  — 900  $\mu\text{E m}^{-2} \text{s}^{-1}$

[Sabolis et al., in prep]

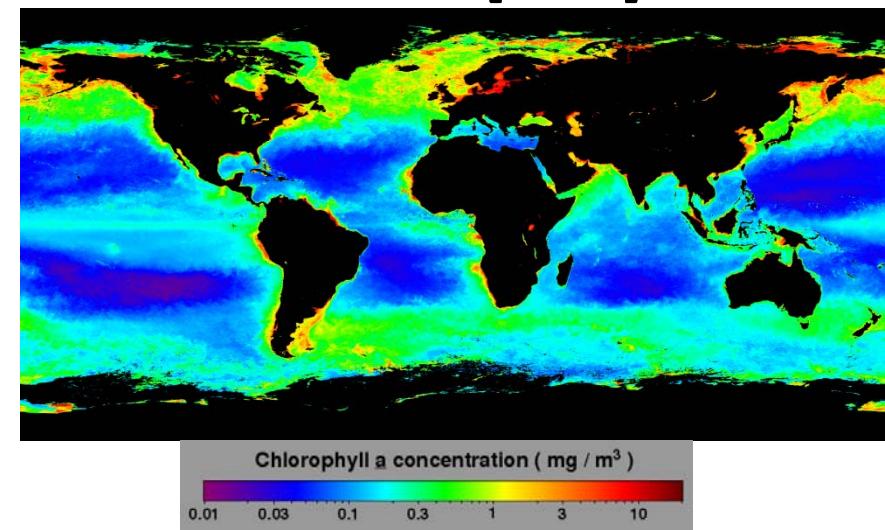
# Light Dependent Production Rates



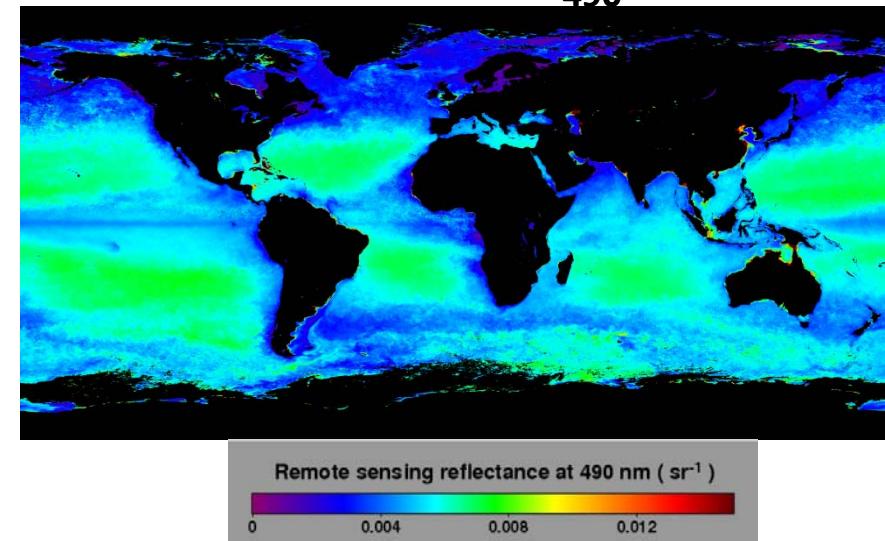
Source: Gantt, B., N. Meskhidze, and D. Kamykowski, Atmos. Chem. Phys., 9, 4915–4927, 2009



SeaWiFS [Chl a]



SeaWiFS  $K_{490}$



<http://oceancolor.gsfc.nasa.gov>



Introduction

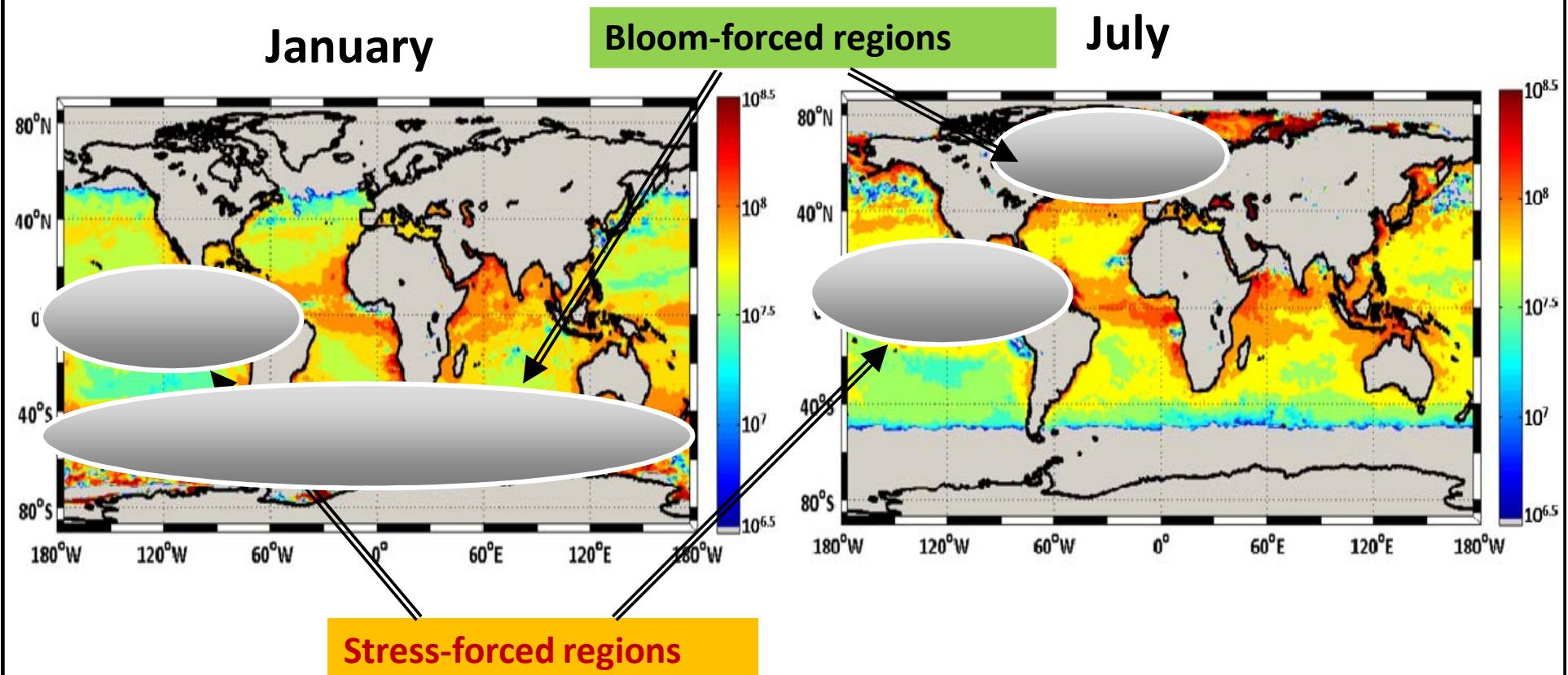
Gas emissions

POM emissions

Model Results

Future

# Marine Isoprene Emissions



Introduction

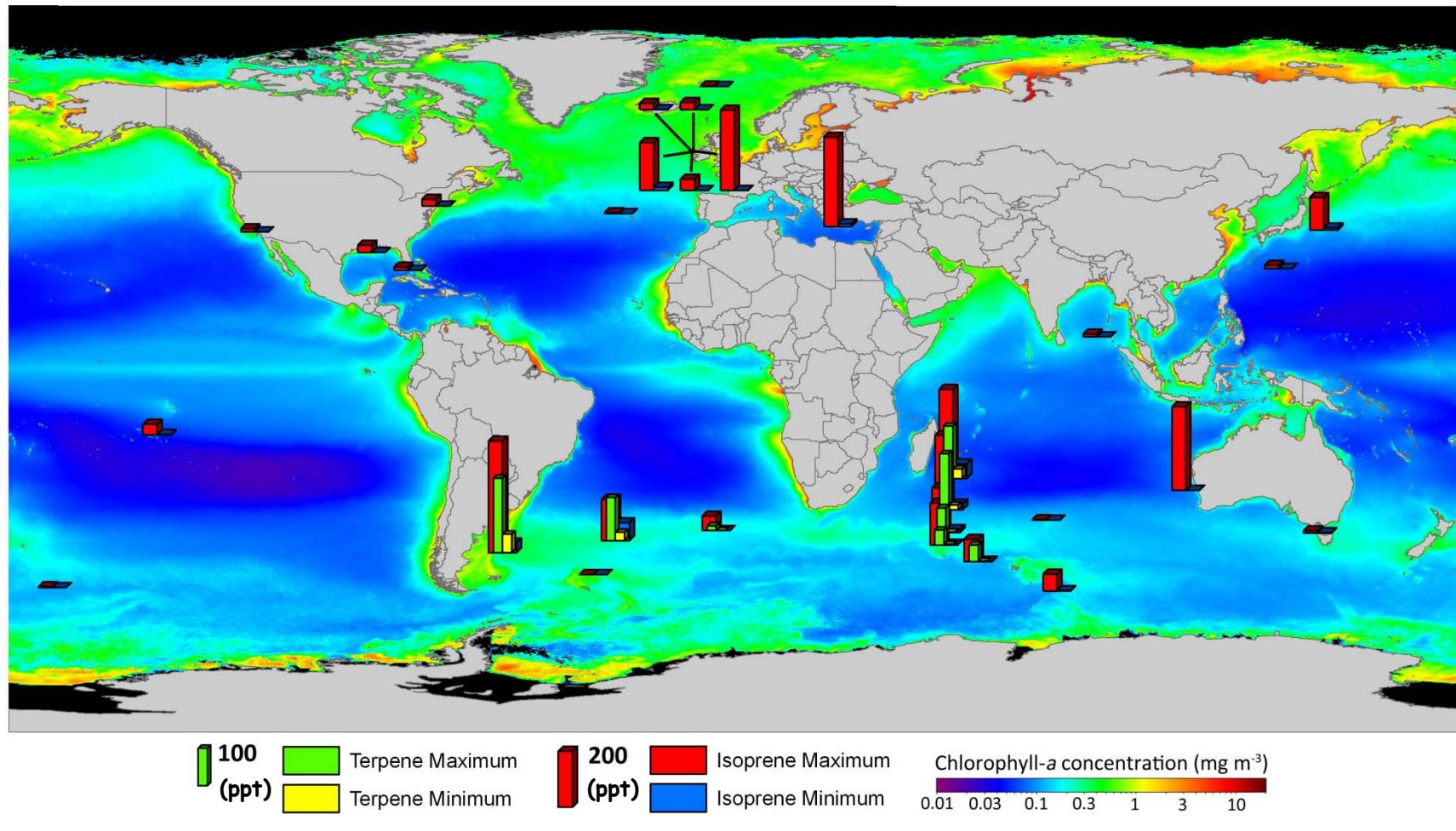
Gas emissions

POM emissions

Model Results

Future

# Observed Marine Boundary Layer Hydrocarbon Concentrations



[Shaw et al., 2010 AMET]

# Global Annual Total Marine Isoprene Emissions

Global Marine Isoprene Emissions (Tg C yr <sup>-1</sup> )	Source
1.1	Bonsang et al. (1992)
0.1	Palmer and Shaw (2005)
1.2	Sinha et al. (2007)
0.27	Arnold et al. (2008)
1.68	Arnold et al. (2008)
<b>0.92</b>	Gantt et al. (2009)
2.5	Luo and Yu (2009)

- Global terrestrial emissions ~ 440 to 660 Tg C yr<sup>-1</sup> (500 to 750 Tg isoprene)

# Primary and Secondary Organic Aerosol Formation and Growth

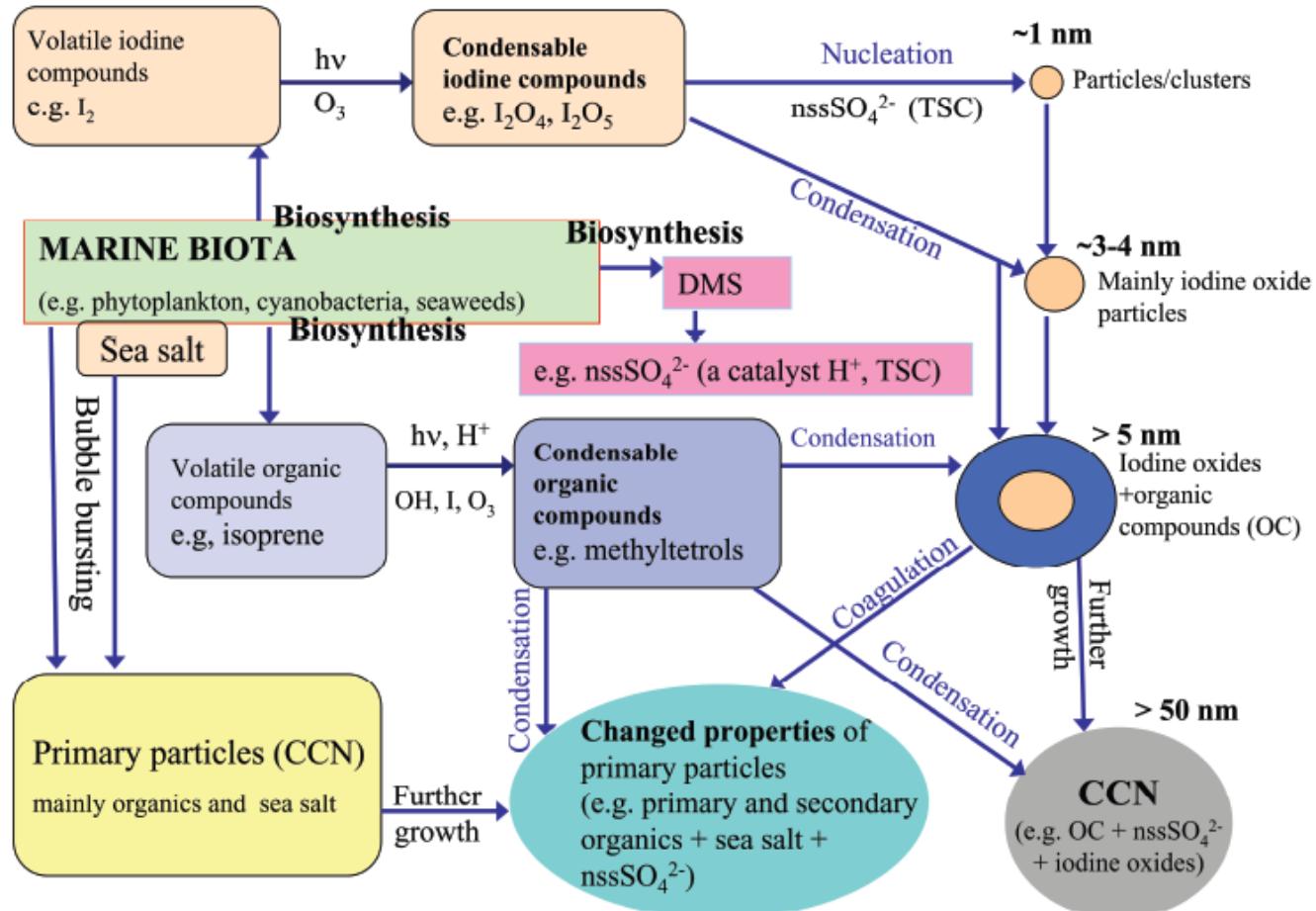


Fig. 13 Vaattovaara, P., Huttunen, P. E., Yoon, Y. J., Joutsensaari, J., Lehtinen, K. E. J., O'Dowd, C. D. & Laaksonen, A. 2006 The composition of nucleation and Aitken modes particles during coastal nucleation events: evidence for marine secondary organic contribution. *Atmos. Chem. Phys.* 6, 4601–4616.



Introduction

Gas emissions

POM emissions

Model Results

Future

# Outline



- 1) Introduction
- 2) Biogenic gas emissions from the ocean and their impact on aerosol/cloud interaction and radiative properties of the overlying atmosphere
- 3) Sources, chemical composition and size distribution of ocean-derived primary organic aerosols**
- 4) Model results
- 5) The future



Introduction

Gas emissions

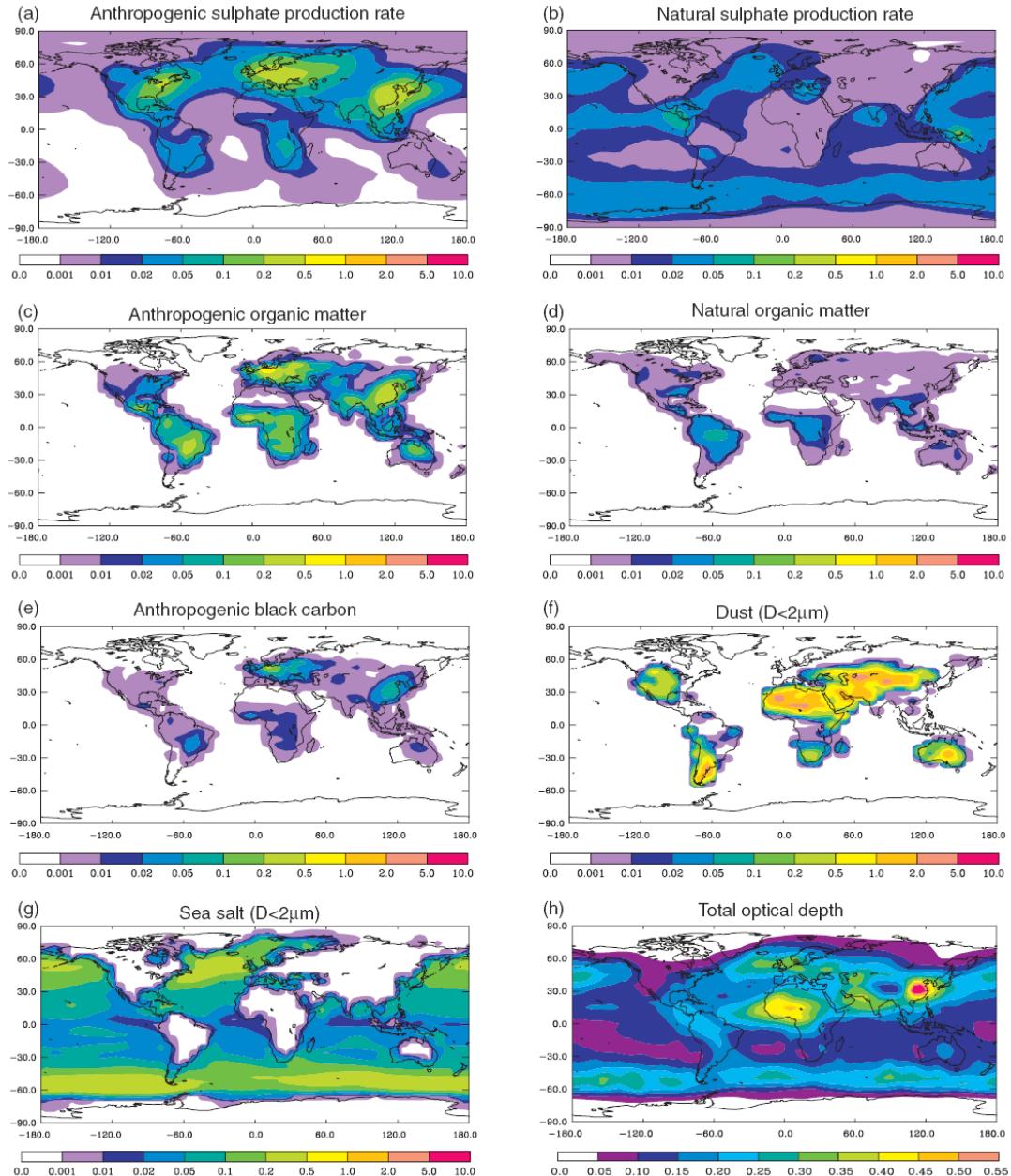
POM emissions

Model Results

Future

# Annual average source strength in four major aerosol types [kg km<sup>-2</sup> hr<sup>-1</sup>]

- Predicted sea salt emissions 3,340 Tg in 2000 could increase to 5,880 Tg in 2100
- The present day direct radiative impact of sea salt is -0.60 and -1.52 Wm<sup>-2</sup> (Ma et al., 2008)
- Total indirect forcing of sea salt is -2.9 Wm<sup>-2</sup> (Ma et al., 2008)
- Potentially important negative climate feedback



Source: Figure 5.2, Penner et al., Climate Change 2001: Working Group I: The Scientific Basis, IPCC 2001



Introduction

Gas emissions

POM emissions

Model Results

Future

# Global Emission Estimates for Major Aerosol Types

Table 5.3: Primary particle emissions for the year 2000 (Tg/yr)<sup>a</sup>.

	Northern Hemisphere	Southern Hemisphere	Global	Low	High	Source
Carbonaceous aerosols						
Organic Matter (0–2 µm)						
Biomass burning	28	26	54	45	80	Liousse <i>et al.</i> (1996), Scholes and Andreae (2000)
Fossil fuel	28	0.4	28	10	30	Cook <i>et al.</i> (1999), Penner <i>et al.</i> (1993)
Biogenic (>1µm)	—	—	56	0	90	Penner (1995)
Black Carbon (0–2 µm)						
Biomass burning	2.9	2.7	5.7	5	9	Liousse <i>et al.</i> (1996); Scholes and Andreae (2000)
Fossil fuel	6.5	0.1	6.6	6	8	Cooke <i>et al.</i> (1999); Penner <i>et al.</i> (1993)
Aircraft	0.005	0.0004	0.006			
Industrial Dust, etc. (> 1 µm)			100	40	130	Wolf and Hidy (1997); Andreae (1995) Gong <i>et al.</i> (1998)
Sea Salt						
d< 1 µm	23	31	54	18	100	
d=1–16µm	1,420	1,870	3,290	1,000	6,000	
Total	1,440	1,900	3,340	1,000	6,000	
Mineral (Soil) Dust <sup>b</sup>						
d< 1 µm	90	17	110	—	—	
d=1–2µm	240	50	290	—	—	
d=2–20µm	1,470	282	1,750	—	—	
Total	1,800	349	2,150	1,000	3,000	

<sup>a</sup> Range reflects estimates reported in the literature. The actual range of uncertainty may encompass values larger and smaller than those reported here.

<sup>b</sup> Source inventory prepared by P. Ginoux for the IPCC Model Intercomparison Workshop. Table 5.3, , Penner *et al.*, Climate Change 2001: Working Group I: The Scientific Basis, IPCC 2001



Introduction

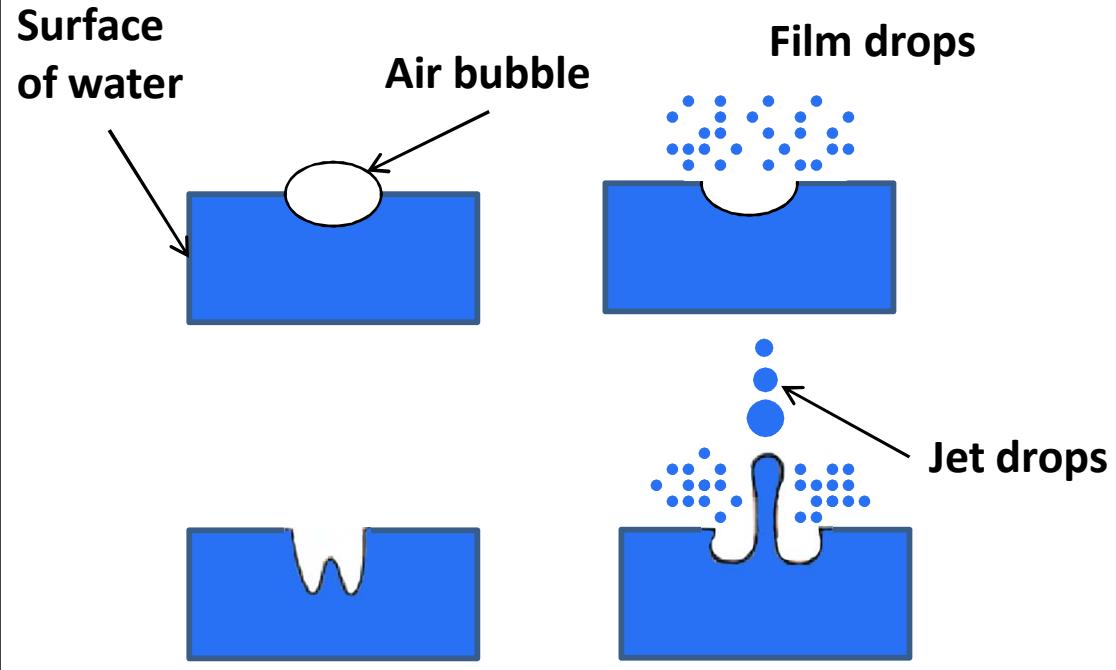
Gas emissions

POM emissions

Model Results

Future

# Sea Spray Aerosol Production



**Sea-salt production through bubble bursting:**

- ✓ **Film drops** (many and small) ; contain material from the surface microlayer
- ✓ **Jet drops** (fewer and larger) ; Mostly sea-salt



Introduction

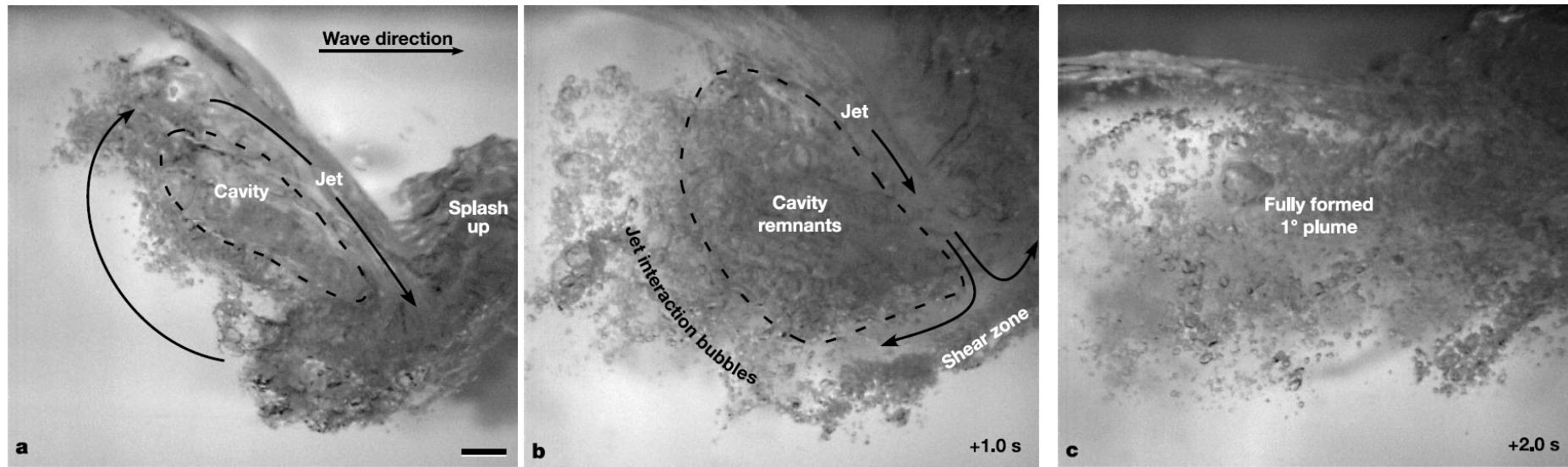
Gas emissions

POM emissions

Model Results

Future

# Sea Spray Aerosol Production



[Deane & Stokes, Nature, 2002]



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Introduction

Gas emissions

POM emissions

Model Results

Future

# Quantifying Sea Spray

- Bubble bursting experiments
- Aerosols observations combined with meteorology
  - Direct particle measurements with condensation particle counter (CPC) or differential mobility analyzer (DMA)
  - Indirect aerosol measurements of aerosol optical depth (AOD) with a sun photometer

[http://www.nuigalway.ie/ccaps/images/Mace\\_Head.jpg](http://www.nuigalway.ie/ccaps/images/Mace_Head.jpg)



CPC + sonic anemometer

Sun photometer + sonic anemometer

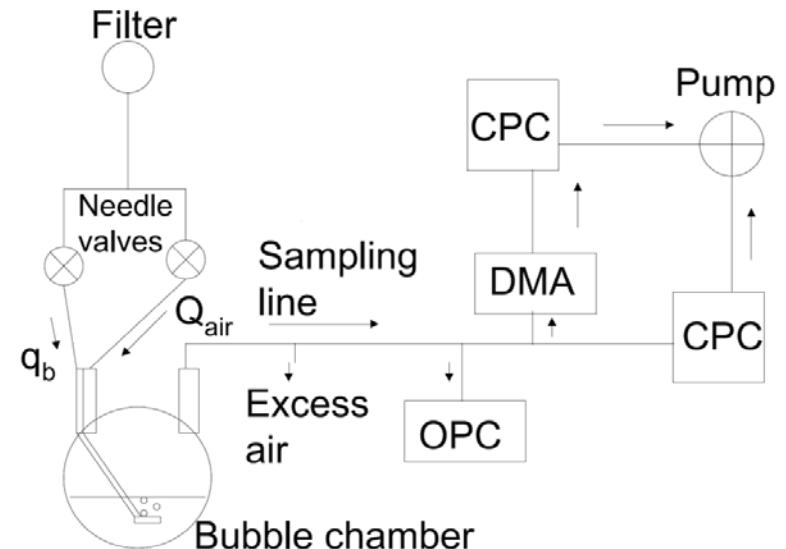


Figure 1. Martensson, E. M., E. D. Nilsson, G. de Leeuw, L. H. Cohen, and H.-C. Hansson, Laboratory simulations and parameterization of the primary marine aerosol production, *J. Geophys. Res.*, 108(D9), 4297, doi:10.1029/2002JD002263, 2003.



[http://mynasadata.larc.nasa.gov/images/AERONET\\_sunphotometer.jpg](http://mynasadata.larc.nasa.gov/images/AERONET_sunphotometer.jpg)



Introduction

Gas emissions

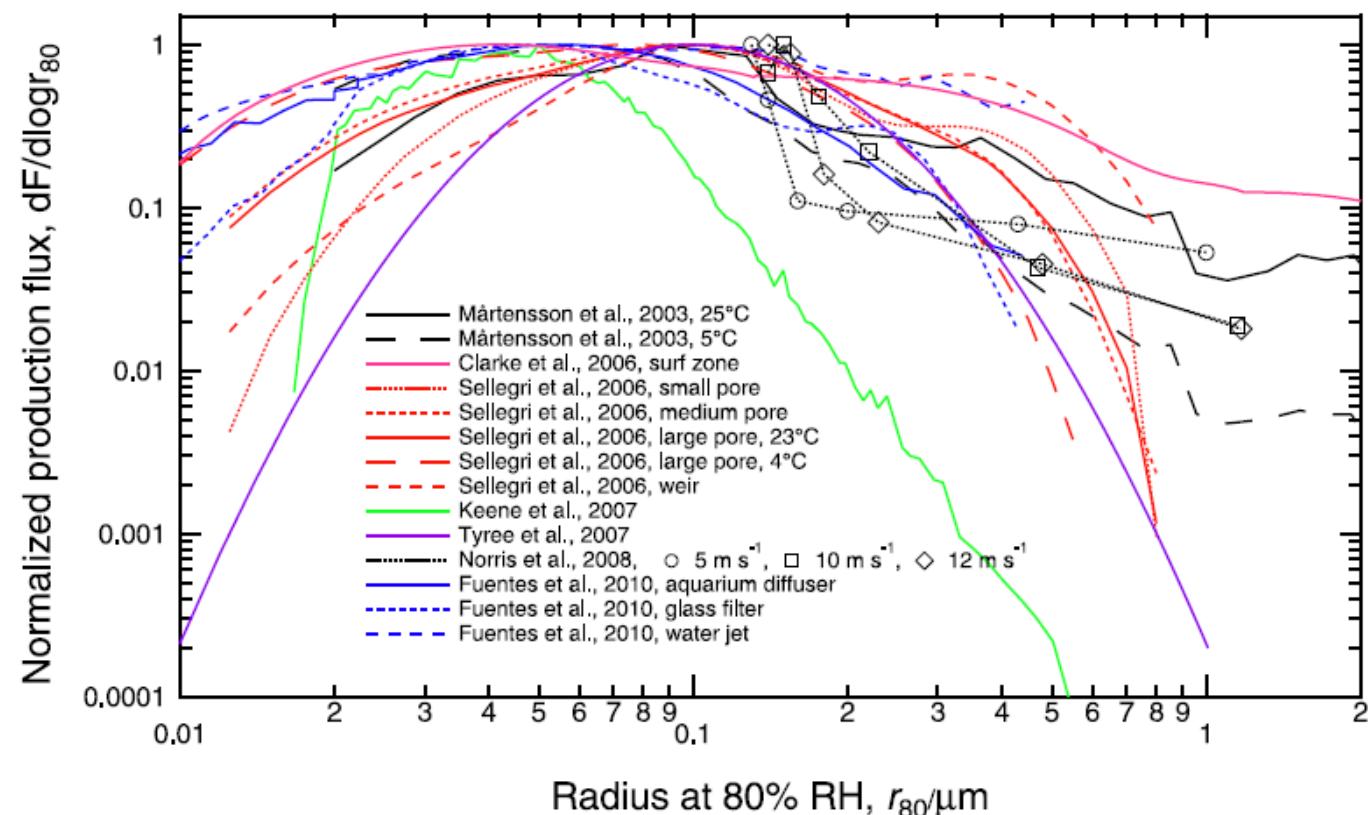
POM emissions

Model Results

Future

# Sea Spray Number Emissions

- Based on two relationships
  - Wind speed and white cap formation
  - White cap formation and sea spray
- Number emissions are predominantly in the sub-micron range, although there are significant numbers in the coarse mode



Introduction

Gas emissions

POM emissions

Model Results

Future

## Sea Spray Source Functions and Range of Applicability [O'Dowd and de Leeuw, 2007].

reference	method	Formulation ( $\text{m}^{-2}\mu\text{m}^{-1}\text{s}^{-1}$ )	Size ( $\mu\text{m}$ )	WS ( $\text{m s}^{-1}$ )	SST ( $^{\circ}\text{C}$ )
Monahan et al. (1986)	whitecap/lab	$dF_{M86}/dr = 1.373 U_{10}^{3.41} r^{-3} (1 + 0.057 r^{1.05}) \times 10^{1.19 e^{-B^2}}$ $B = (0.380 - \log r)/0.650$	$0.3 < r_{80} < 20$	n.a.	lab temp (~ 20)
de Leeuw et al. (2000)	whitecap/surf	$dF_N/dD = W \times 1.1 \times e^{0.23 \times U} \times D^{-1.65}$	$1.6 < D_0 < 20$	0–9	~ 16
de Leeuw et al. (2003)	model/field	$dF_N/dr = c \left( \sum_{i=1}^2 A_i e^{-c_i * \ln(r/r_1)^2} \right)$ units: $\mu\text{m}^{-1} \text{m}^{-2} \text{s}^{-1}$ $A_1 = 1.41 u + 0.98$ $A_2 = 0.51 u - 1.82$ $C_1 = -0.1 u + 1.69$ $C_2 = 1.09$ $c = (0.24 u + 0.4) \times 10^4$	$0.063 < R < 7.996$	0–9	~16
Vignati et al. (2001)	model/field	$dF_{v00}(\log r_{80})/d \log r = \sum_{i=1}^3 N_i / (\sqrt{2\pi} \log \sigma_i)$ $\exp(-(\log r_{80} - \log R_i)/2 \log^2 \sigma_i)$ $d \log r = 0.1$ $i=1; N_i = 10^{(0.095U+0.283)}; R_i = 0.2; \sigma_i = 1.9$ $i=1; N_i = 10^{(0.0422U-0.288)}; R_i = 2; \sigma_i = 2$ $i=1; N_i = 10^{(0.069U-3.5)}; R_i = 12; \sigma_i = 3$	$0.04 < r_{80} < 13$	6–17	~ 13
Märtensson et al. (2003)	whitecap/lab	$dF_0/d \log D_p = W(A_k T_w + B_k)$ , $T_w$ is water temperature in K $A_k = c_1 D_p^4 + c_2 D_p^3 + c_3 D_p^2 + c_4 D_p + c_5$ $B_k = d_1 D_p^4 + d_2 D_p^3 + d_3 D_p^2 + d_4 D_p + d_5$ $k=1$ , size ranges $j=1-13$ , $0.018-0.168 \mu\text{m}$ $k=2$ , size ranges $j=13-26$ , $0.168-0.949 \mu\text{m}$ $k=3$ , size ranges $j=26-38$ , $0.949-5.700 \mu\text{m}^b$	$0.018 < D_d < 20$	n.a.	2–25
Gong (2003)	modified Monahan/ lab	$dF_{M86}/dr = 1.373 U_{10}^{3.41} r^{-A} (1 + 0.057 r^{3.45}) \times 10^{1.19 e^{-B^2}}$ $A = 4.7(1 + \theta r)^{-0.017 r^{-1.44}}$ $B = (0.433 - \log r)/0.433$	$0.07 < r_{80} < 20$ $0.04 < r_d$	n.a.	see Monahan et al. (1986)
Clarke et al. (2006)	whitecap/surf	$(dF_N/d \log D_p) = w \sum_{i=1}^3 A_i$ $A_i = \beta_0 + \beta_1 D_p + \beta_2 D_p + \beta_3 D_p + \beta_4 D_p + \beta_5 D_p^c$	$0.01 < D_p < 8$	n.a.	~25

For Geever  $100 \text{ nm} < D < 1000 \text{ nm}$   $f(x) = ax^b$ ,  $f(x) = \text{Number flux}$ ,  $a = 1.854 \times 10^{-3}$ ,  $x = \text{wind speed } 1-20 \text{ m s}^{-1}$ ,  $r^2 = 0.8$ ,  $b = 2.706$ . For Geever  $10 \text{ nm} < D < 10000 \text{ nm}$   $f(x) = ax^b$ ,  $a = 9.204 \times 10^{-5}$ ,  $b = 4.1$ . SST, sea-spray temperature; WS, wind speed;  $W(U_{10}) = 3.84 \times 10^{-6} U_{10}^{3.41}$ , where  $U_{10}$  ( $\text{m s}^{-1}$ ) is the wind speed at 10 m.

# Sea-water Composition

**Table 6.** Composition of Seawater of Salinity 35

Species	Mass Fraction	Mass	Mole Fraction	Molarity		Ionic Strength		
		Ratio		(kg-sw) <sup>-1</sup>	g (kg-H <sub>2</sub> O) <sup>-1</sup>	mol (kg-sw) <sup>-1</sup>	mol (kg-sw) <sup>-1</sup>	equiv (kg-sw) <sup>-1</sup>
H <sub>2</sub> O	0.9648	964.83	1000.00	0.9795	53.558	55.510	—	—
Cl <sup>-</sup>	0.0194	19.35	20.06	0.0100	0.546	0.566	0.546	0.273
SO <sub>4</sub> <sup>2-</sup>	0.0027	2.71	2.81	0.0005	0.028	0.029	0.056	0.056
HCO <sub>3</sub> <sup>-</sup>	0.0001	0.11	0.11	< 0.0001	0.002	0.002	0.002	0.001
Br <sup>-</sup>	0.0001	0.07	0.07	< 0.0001	0.001	0.001	0.001	< 0.001
Na <sup>+</sup>	0.0108	10.78	11.18	0.0086	0.469	0.486	0.469	0.235
Mg <sup>2+</sup>	0.0013	1.28	1.33	0.0010	0.053	0.055	0.106	0.106
Ca <sup>2+</sup>	0.0004	0.41	0.43	0.0002	0.010	0.011	0.020	0.020
K <sup>+</sup>	0.0004	0.40	0.41	0.0002	0.010	0.011	0.010	0.005
minor species	< 0.0001	0.06	0.05	< 0.0001	0.001	0.001	0.001	0.001
Σ salts	0.0352	35.17 <sup>a</sup>	36.45	0.0205	1.120	1.162	1.211	0.698
<b>Total</b>	<b>1.0000</b>	<b>1000.00</b>	<b>1036.45</b>	<b>1.0000</b>	<b>54.678</b>	<b>56.672</b>	<b>54.769</b>	<b>0.698</b>

Source: Table 6, Ramaswamy et al., Climate Change 2001: Working Group I: The Scientific Basis, IPCC 2001

Why do we even need to even worry about anything other than sea salt?



Introduction

Gas emissions

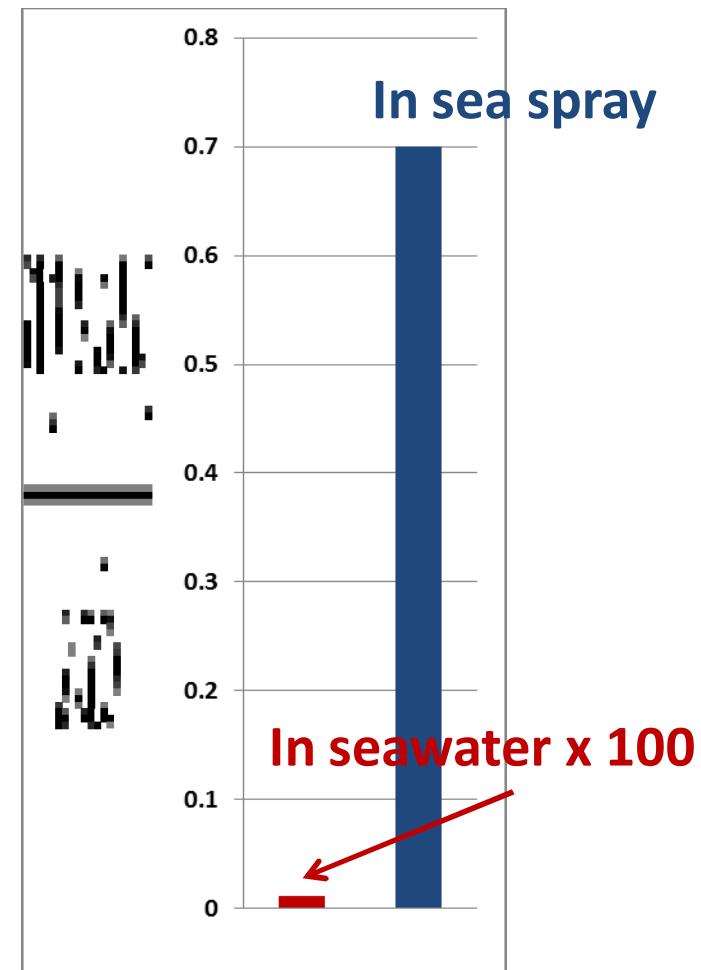
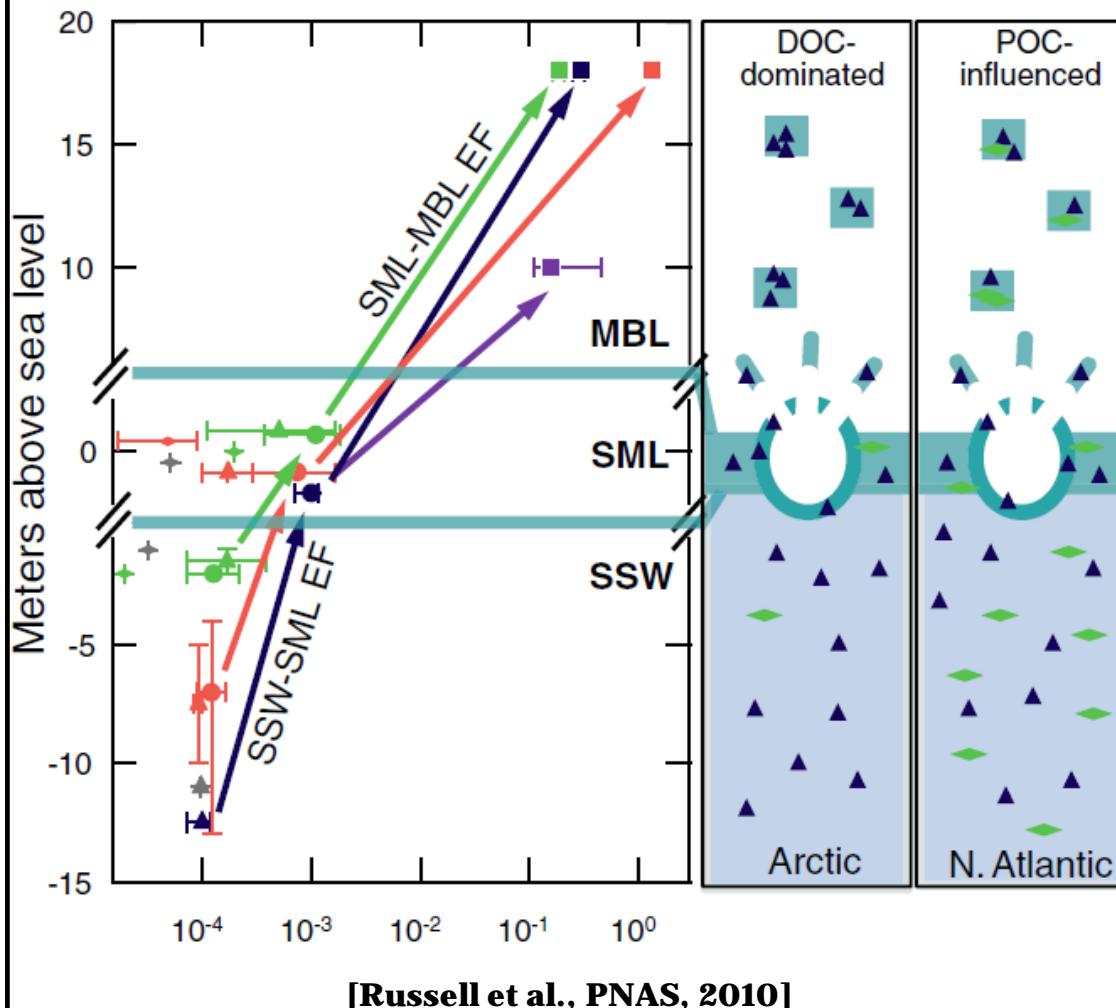
POM emissions

Model Results

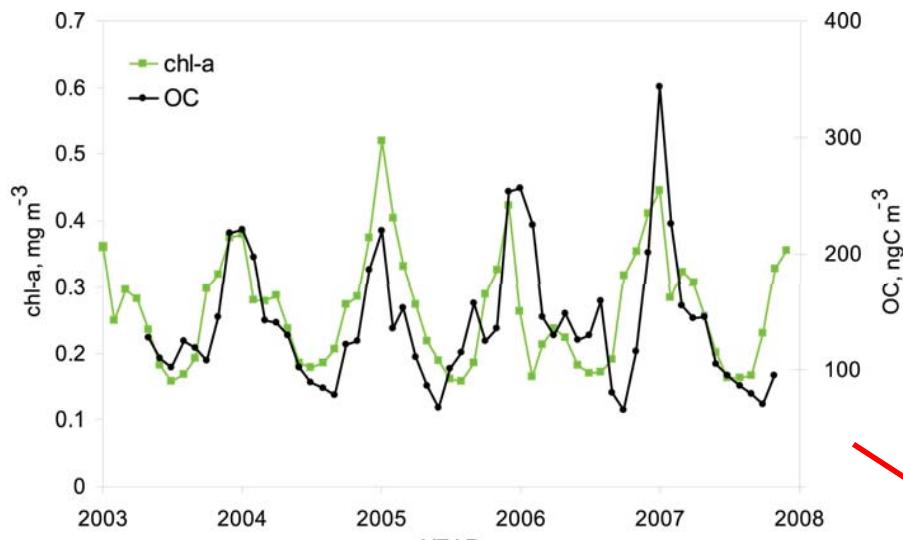
Future

# From Sea Water To Aerosol

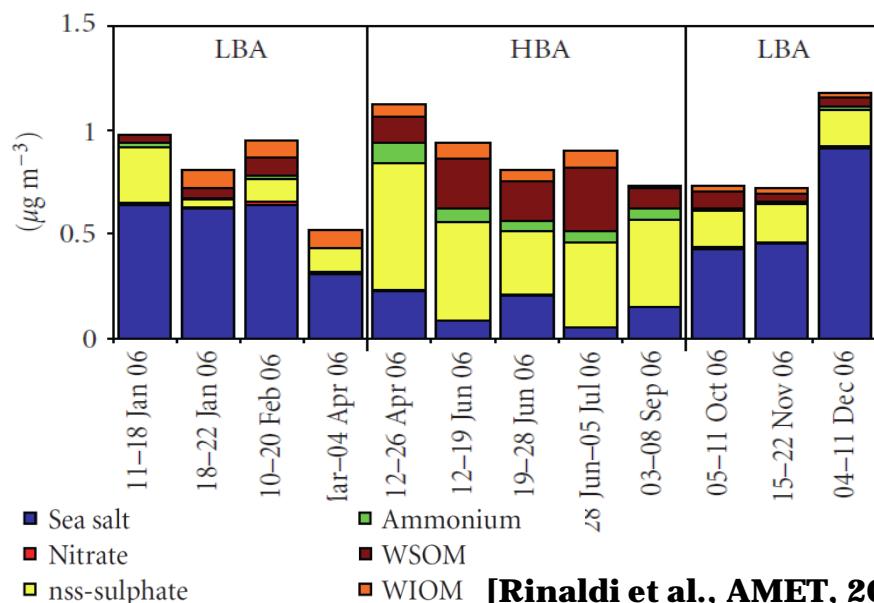
- Chemical composition?
- Controlling mechanism?
- Number or Mass?



# Marine Organic Aerosols: A Biological Source



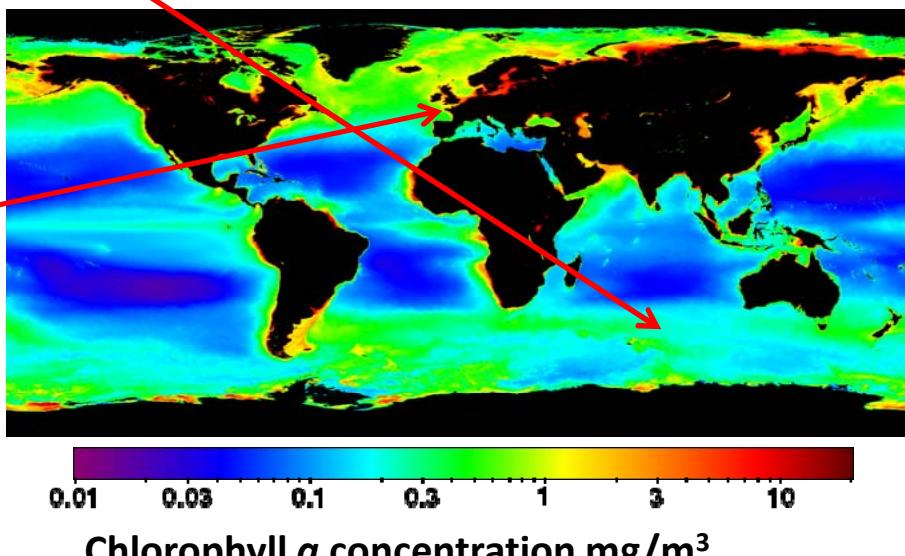
[Sciare et al., JGR, 2009]



[Rinaldi et al., AMET, 2010]

## A direct evidence

- Long term measurements of OC at Amsterdam Island
- Submicron marine aerosol chemical composition at Mace Head



# Flux of an ultrafine sea-salt or marine organic matter?



- "Dry" ( $RH = 40\%$ ) sea salt particle size distributions for  $0.01 \mu m \leq D_p \leq 8 \mu m$
- 60% of all sea salt particles were smaller than  $0.1 \mu m$  diameter

Clarke et al., 2006

- Airborne micro-organisms of marine origin and their fragments peaking at  $D_p$  of  $0.03 - 0.04 \mu m$  do not appear to have any association with sea salt
- The presence of airborne marine aggregates consisting of amorphous transparent exopolymer (EP) secretions (gels) of algae and bacteria
- Sea salt was not detected in particles  $< 0.2 \mu m$  diameter

Leck and Bigg, 2008

- During high biological activity organic fraction can contribute up to 60% of sub-micrometer mass
- During low biological activity periods fraction reduces to 10-15%

O'Dowd et al., 2007; Facchini et al., 2008

Surface films can be highly enriched in organics: "marine microcolloids", "mucus-like" or gel-like material, *Exopolymer gels*, airborne marine aggregates (AMA)



Introduction

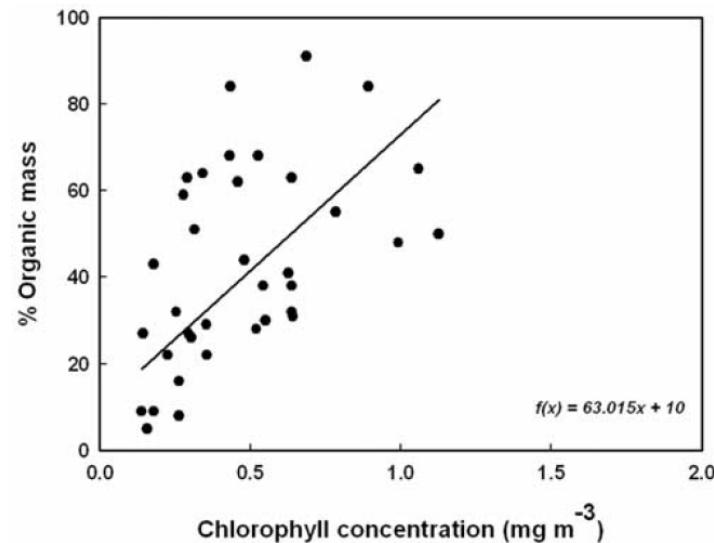
Gas emissions

POM emissions

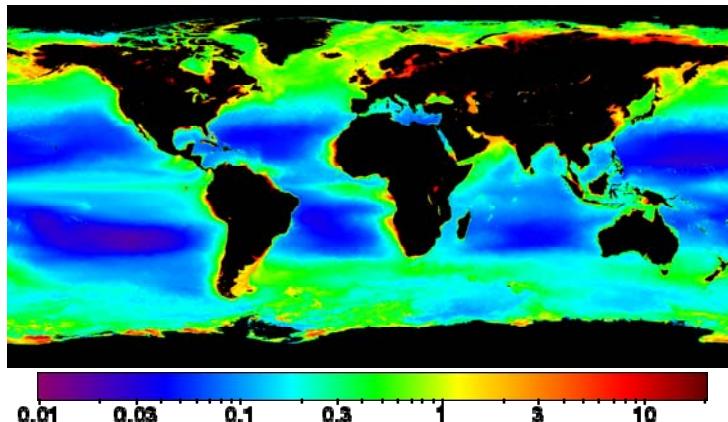
Model Results

Future

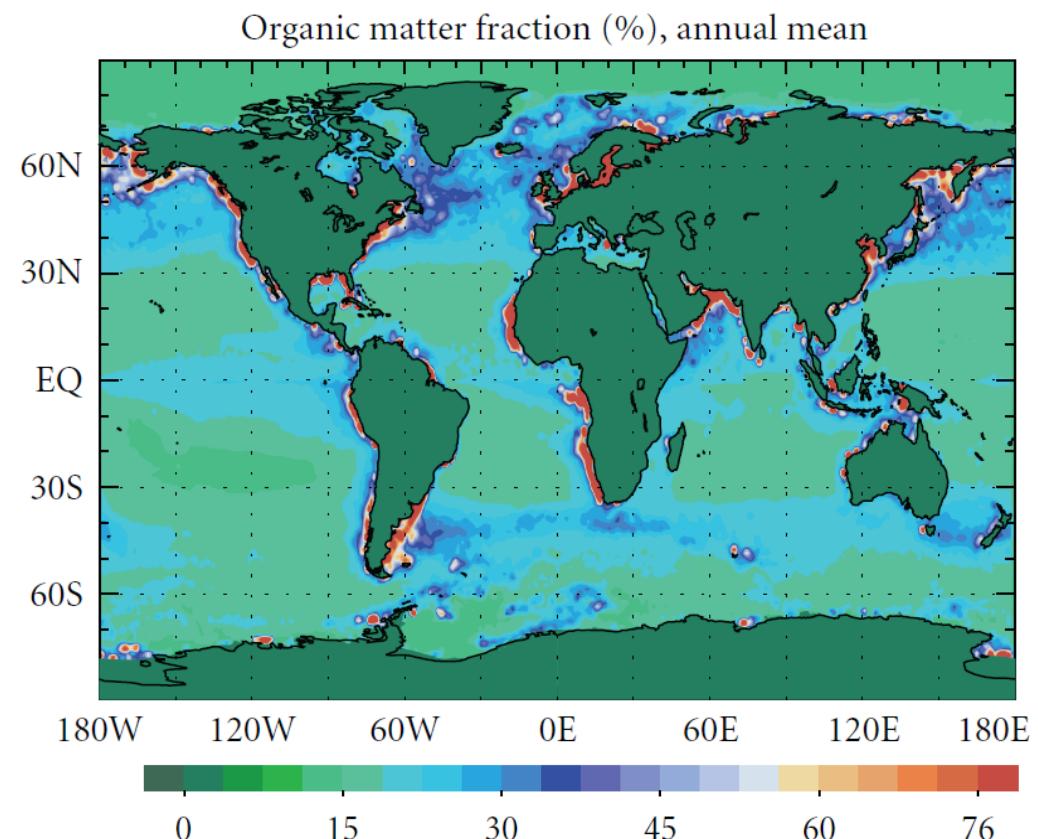
# Modeling Of Marine Primary Organic Aerosol (POM) Emissions



[O'Dowd et al., GRL, 2008]



[Myriokefalitakis et al., AMET, 2010]



Introduction

Gas emissions

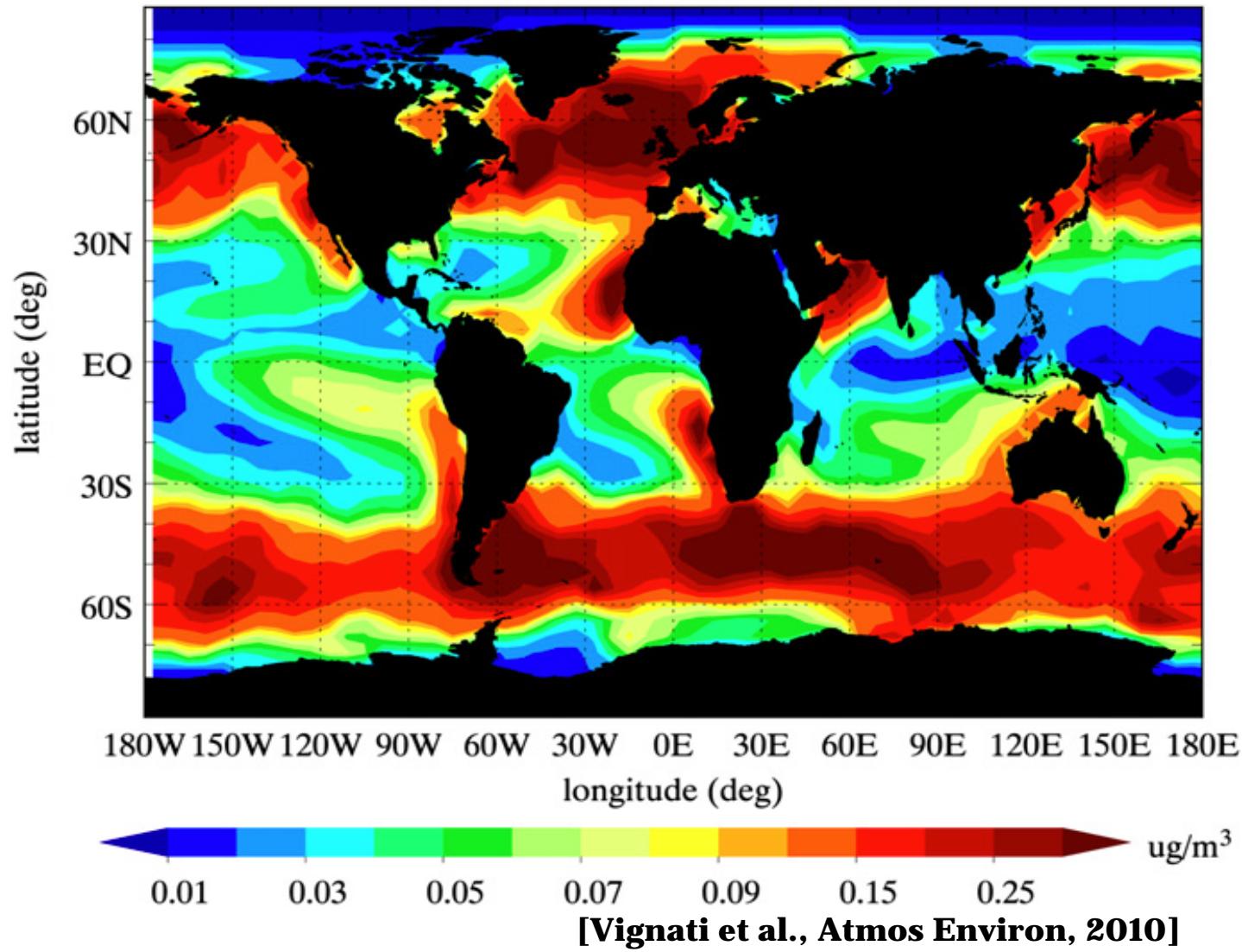
POM emissions

Model Results

Future

# Marine Primary Organic Aerosols

Primary Marine Organic Aerosols



Introduction

Gas emissions

POM emissions

Model Results

Future

# Marine Primary Organic Aerosols

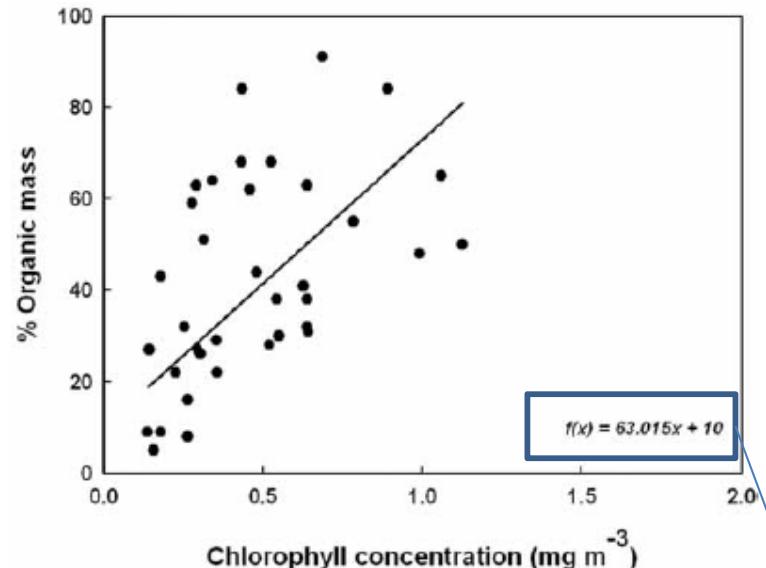
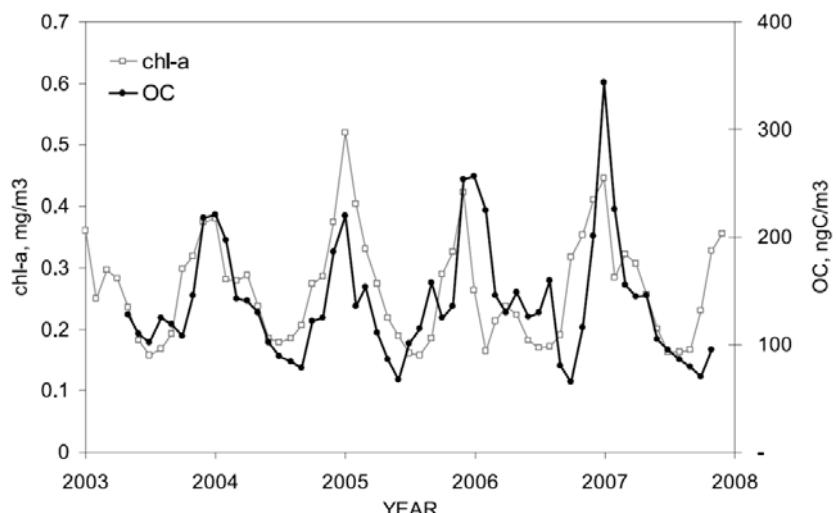


Figure 2. O'Dowd, C. D., B. Langmann, S. Varghese, C. Scannell, D. Ceburnis, and M. C. Facchini (2008), A combined organic-inorganic sea-spray source function, Geophys. Res. Lett., 35, L01801, doi:10.1029/2007GL030331.



- Strong observed relationship between [Chl a] and organic fraction of sea spray mass
- Two steps to calculate organic emissions
  - Convert sea spray number emissions to mass by integrating by particle size, assuming spherical shape, and using the apparent density
  - Multiply the sea spray mass emission rate by organic fraction determined by [Chl a]

$$F_{\text{sub}} = 0.63015 * [\text{Chl-}a] + 0.1$$
$$F_{\text{super}} = F_{\text{sub}} * 0.03$$

Figure 6. Sciare, J., O. Favez, R. Sarda-Este`ve, K. Oikonomou, H. Cachier, and V. Kazan (2009), Long-term observations of carbonaceous aerosols in the Austral Ocean atmosphere: Evidence of a biogenic marine organic source, J. Geophys. Res., 114, D15302, doi:10.1029/2009JD011998.



Introduction

Gas emissions

POM emissions

Model Results

Future

# Marine Organic Aerosol Emissions

- Two classes: primary and secondary
  - Primary
    - Formed by bubble bursting of organic surface layer
    - Mainly water insoluble
  - Secondary
    - Formed by condensation of organic gases (isoprene, monoterpenes, amines, etc)
    - Mainly water soluble
- Both tied to the productivity of the ocean with particular focus on [Chl-*a*]

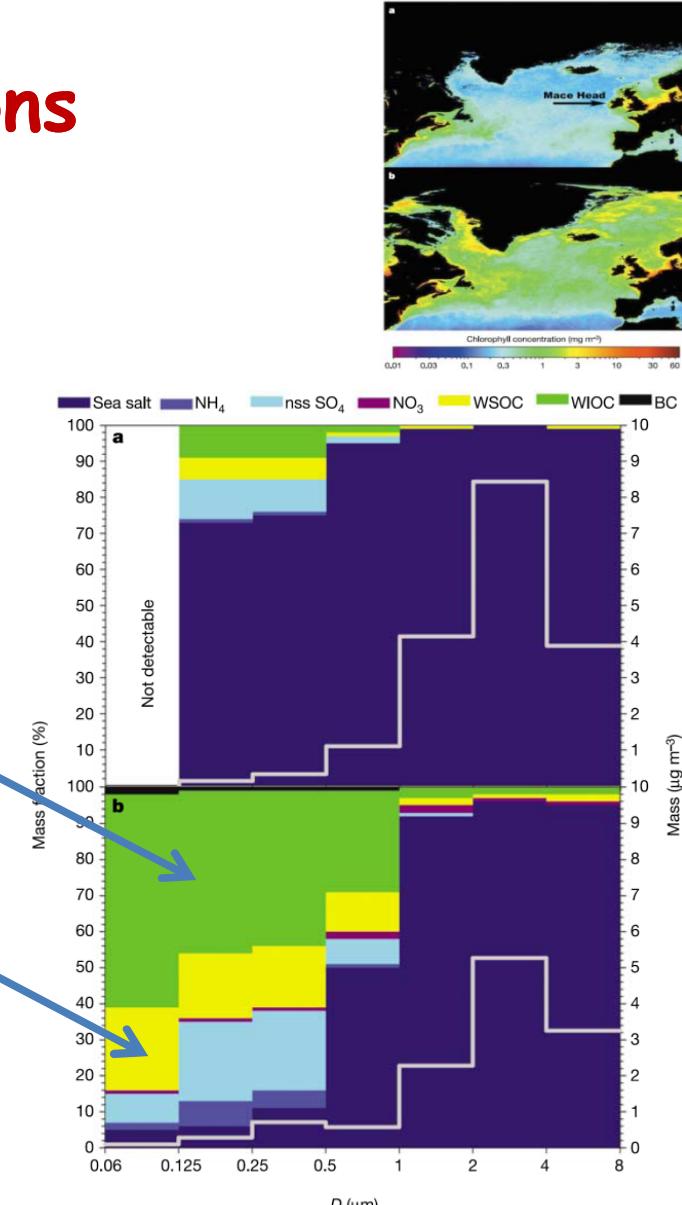


Figure 2. O'Dowd, C. D., M. C. Facchini, F. Cavalli, D. Ceburnis, M. Mircea, S. Decesari, S. Fuzzi, Y. J. Yoon, and J. P. Putaud (2004), Biogenically driven organic contribution to marine aerosol, *Nature*, 431, 676–680, doi:10.1038/nature02959



Introduction

Aerosol deposition

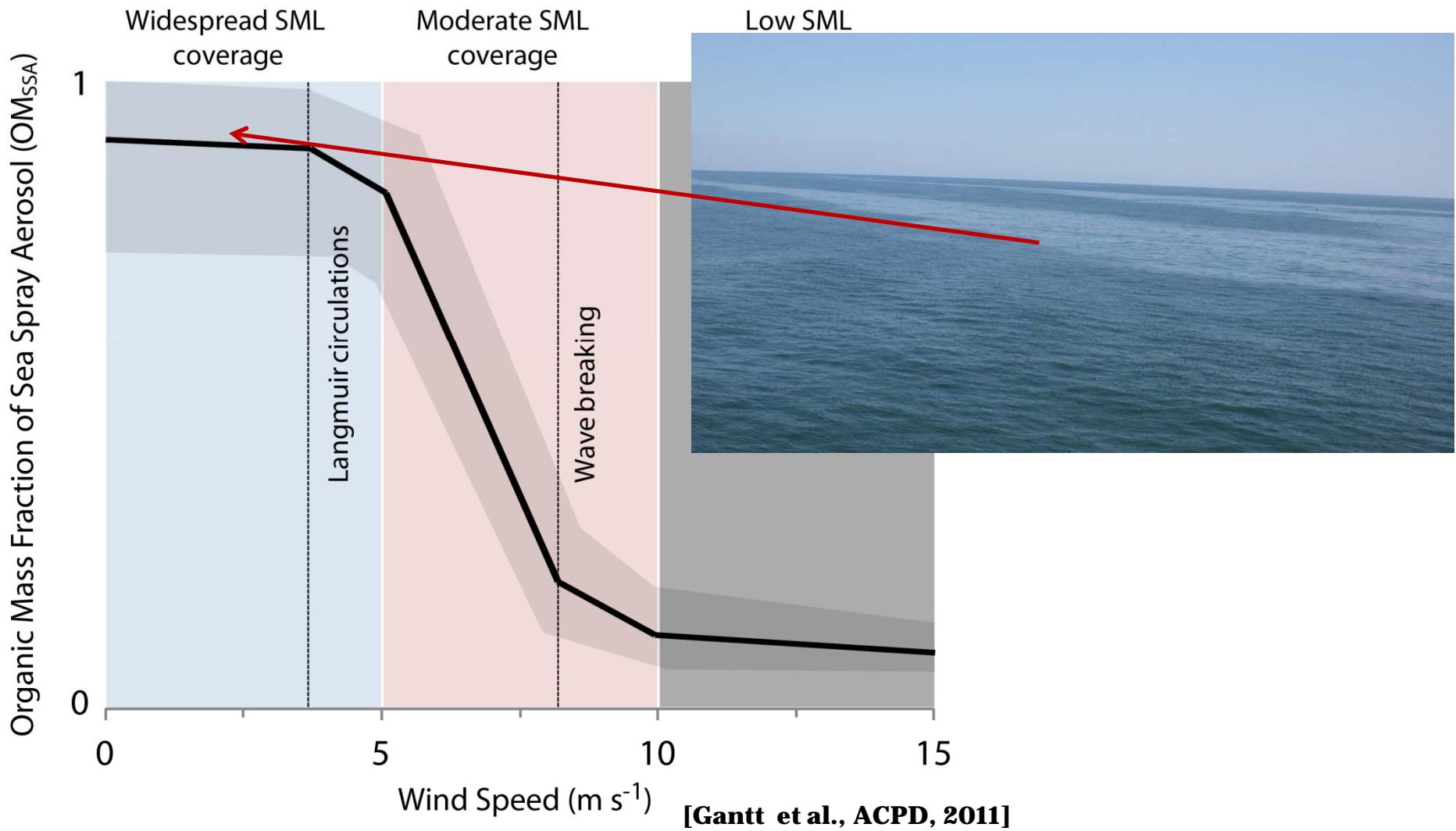
Gas emissions

POM emissions

Model Results

Future

# Organic Enrichment Of The Air-sea Interface And Surface Wind Speed



Introduction

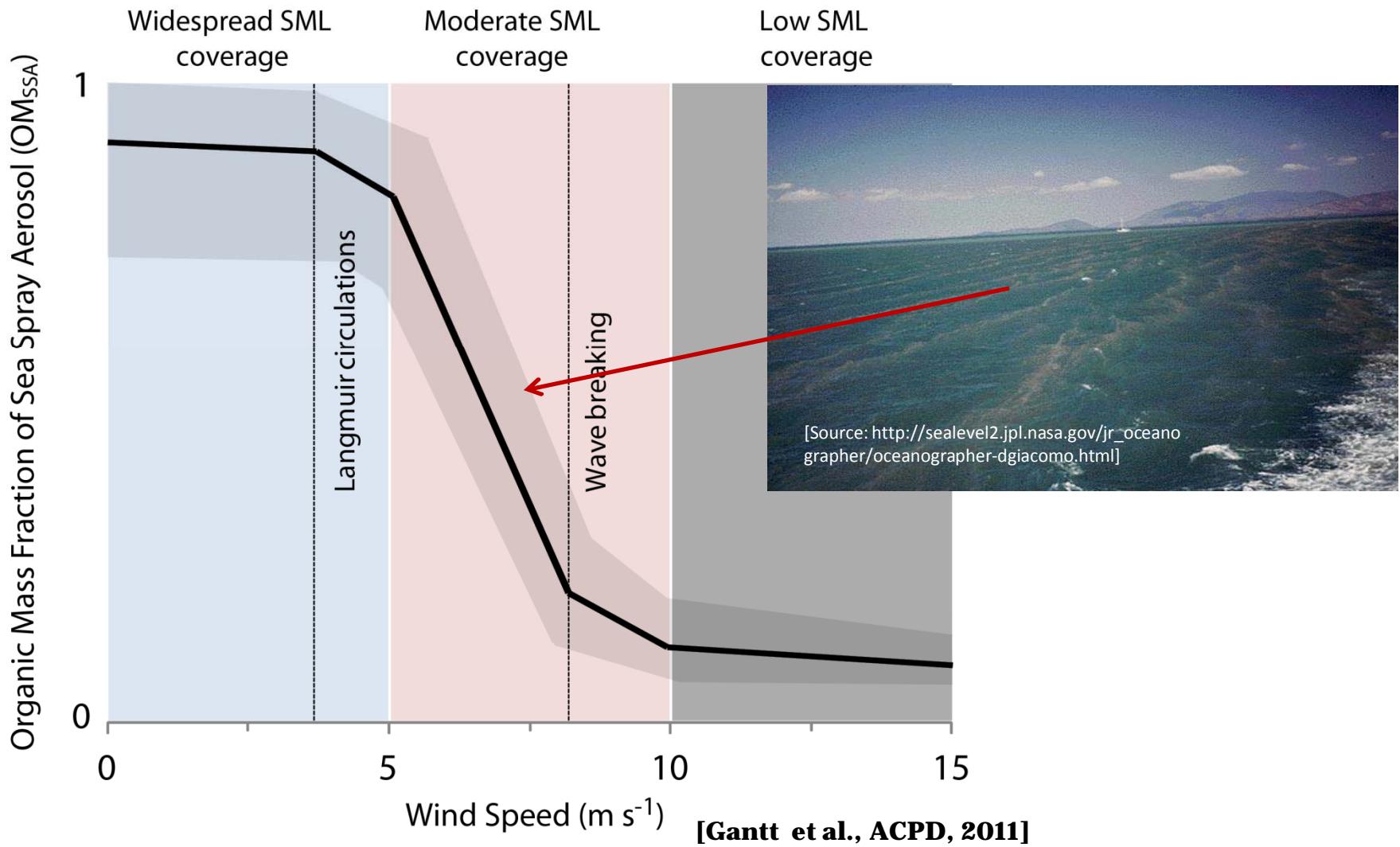
Gas emissions

POM emissions

Model Results

Future

# Organic Enrichment Of The Air-sea Interface And Surface Wind Speed



Introduction

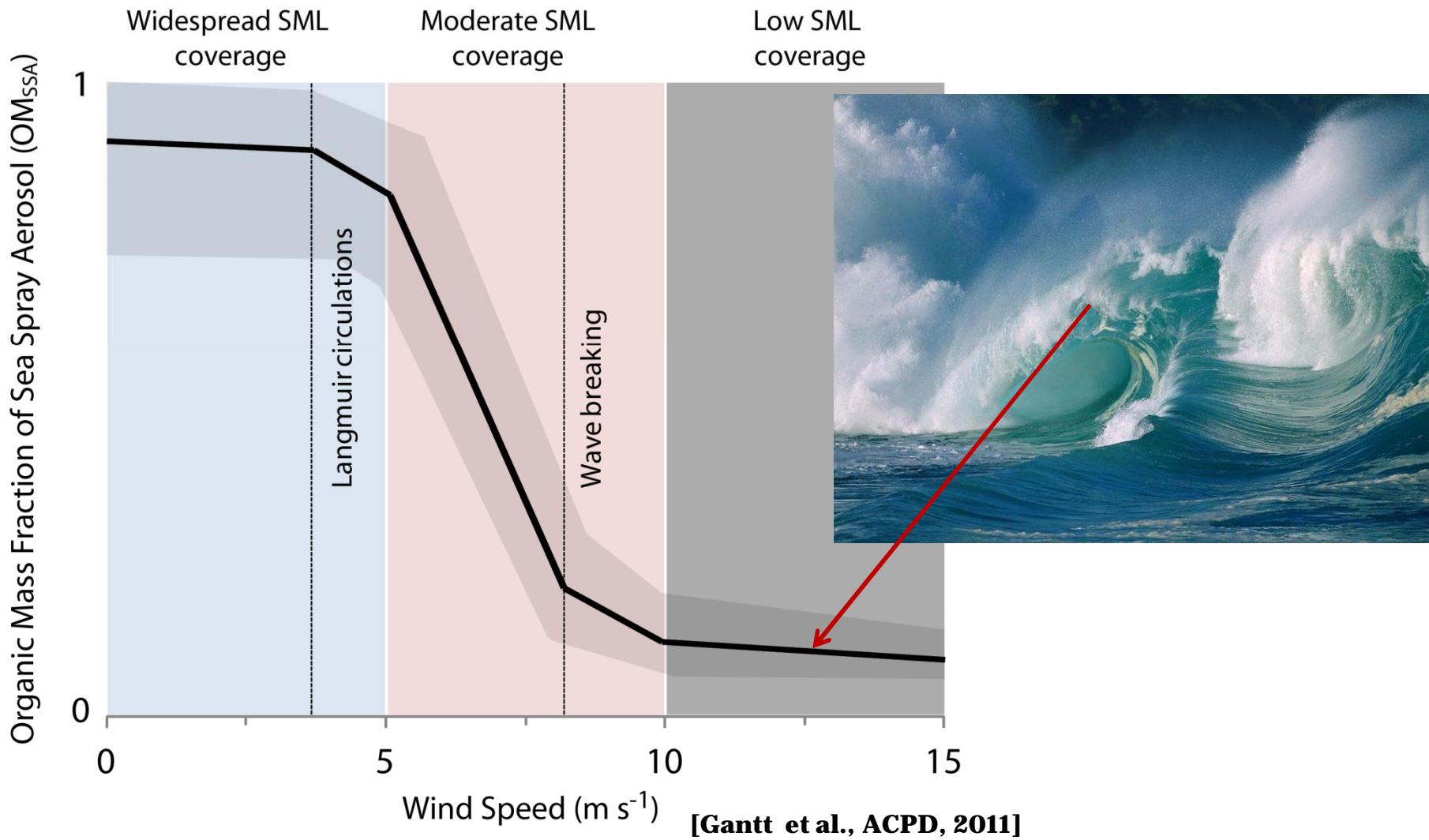
Gas emissions

POM emissions

Model Results

Future

# Organic Enrichment Of The Air-sea Interface And Surface Wind Speed



Introduction

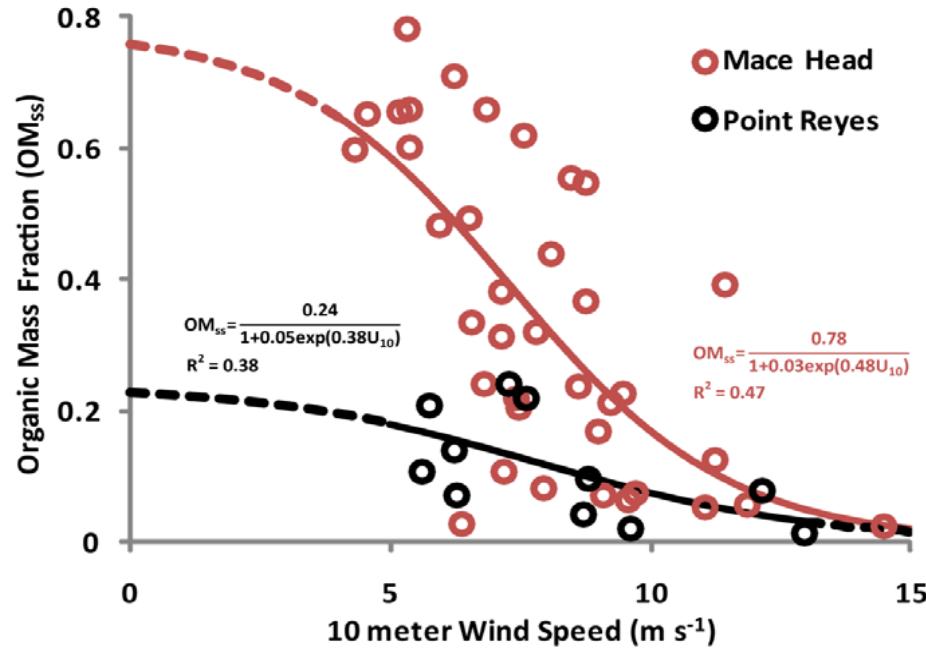
Gas emissions

POM emissions

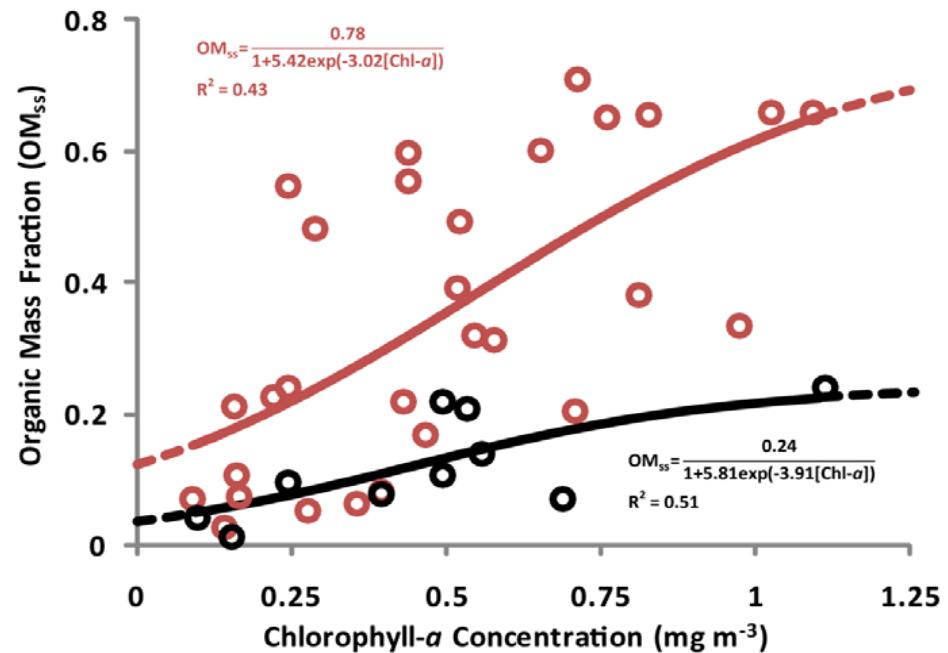
Model Results

Future

# Organic Mass Fraction of Sea Spray



[Source: Gantt et al., ACPD, 2011]



Introduction

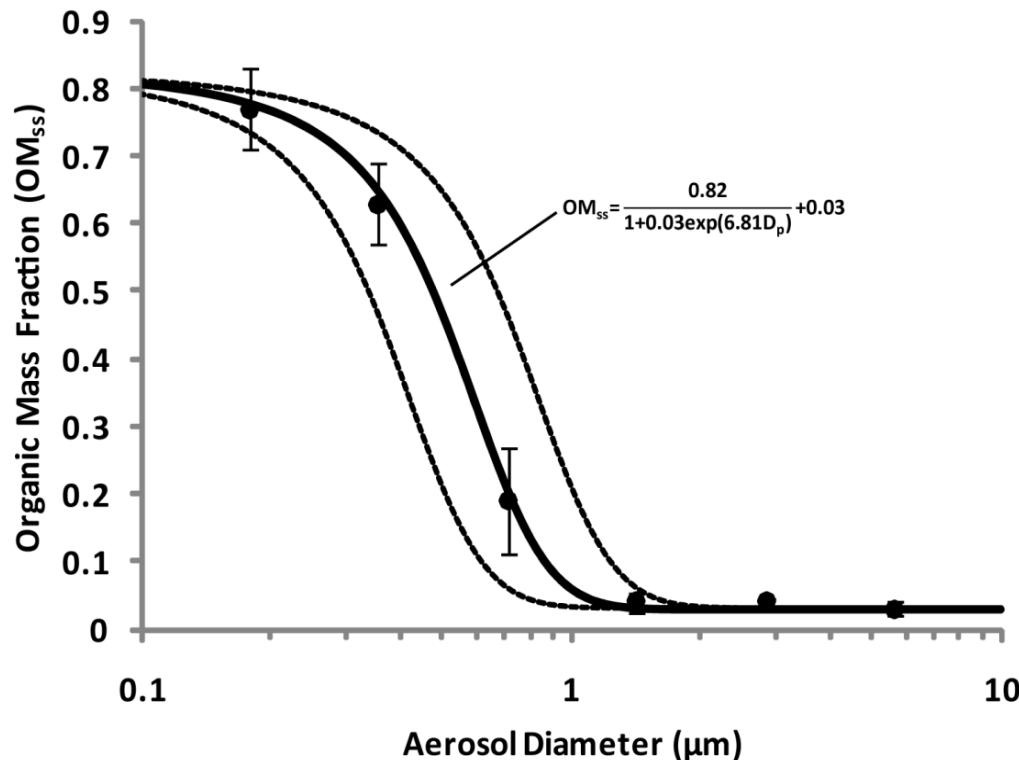
Gas emissions

POM emissions

Model Results

Future

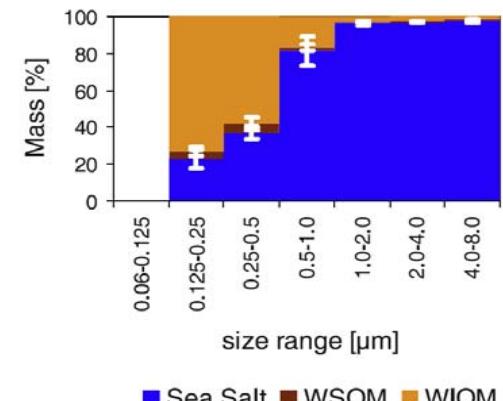
# $OC_{ss}$ as a Function of Ambient Aerosol Aerodynamic Diameter



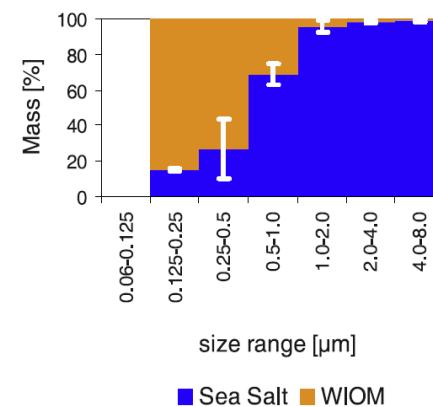
$$OC_{ss}(D_p) = \frac{OC_{ss}^{\max}(D_p)}{1+0.05\exp(6.64D_p)} + OC_{ss}^{\min}(D_p)$$

[Source: Gantt et al., ACPD, 2011]

Bubble bursting samples



Mace Head



[Facchini et al., GRL, 2008]



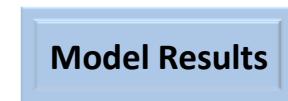
Introduction



Gas emissions



POM emissions



Model Results

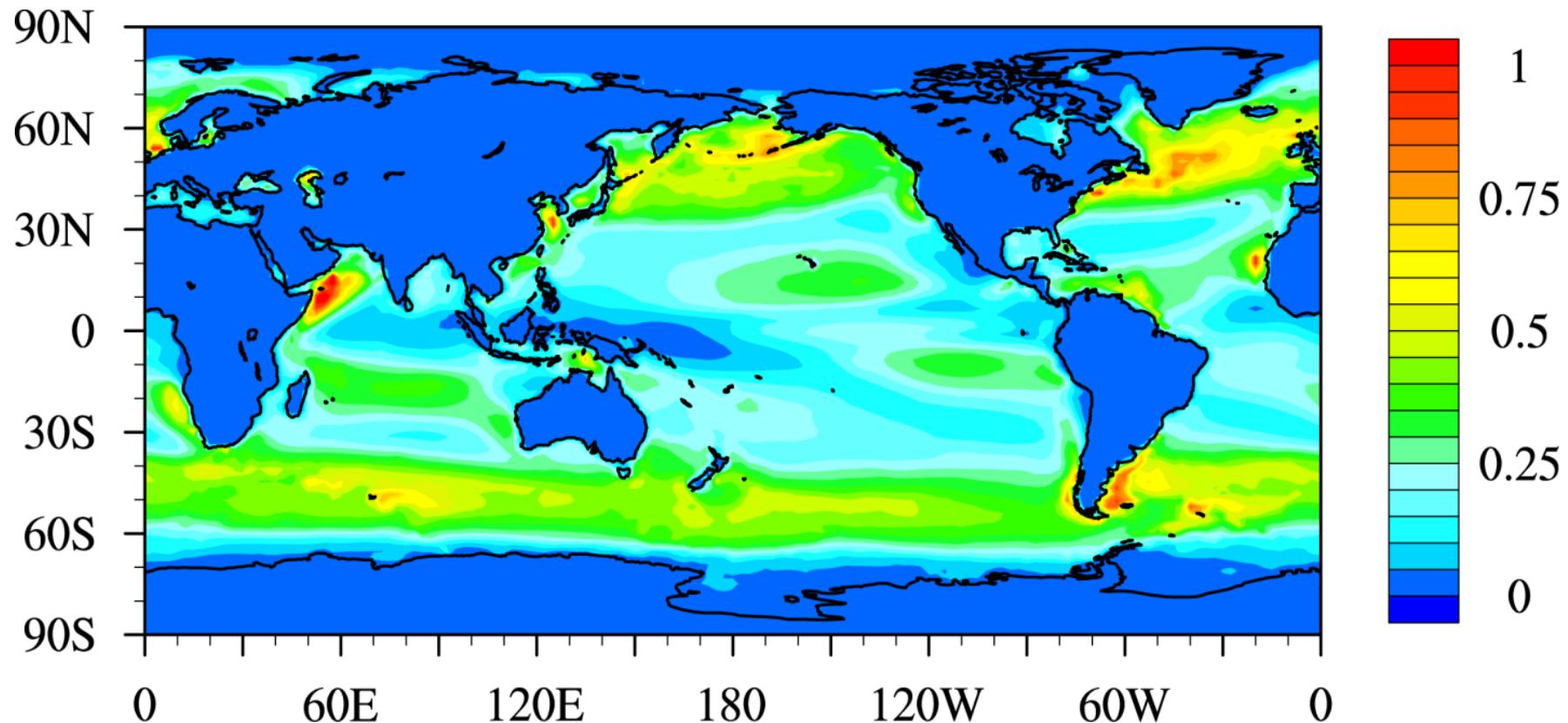


Future

# Annual Average Submicron Marine Primary OC Emission Rate

2.8 to 5.6 Tg C yr<sup>-1</sup>

Units: ng C m<sup>-2</sup> s<sup>-1</sup>



[Source: Gantt et al., ACPD, 2011]



Introduction

Gas emissions

POM emissions

Model Results

Future

# Global Annual Marine Primary Organic Aerosol Emission

Sub-micron Primary Organic Carbon (Tg C yr <sup>-1</sup> )	Source
5.5	Spracklen et al. (2008)
2.5	Langmann et al. (2008)
2.9	Gantt et al. (2009)
5.8	Vignati et al. (2010)
<b>2.8 to 5.6</b>	Gantt and Meskhidze (2011)
Super-micron Primary Organic Carbon (Tg C yr <sup>-1</sup> )	
75	Spracklen et al. (2008)
8	Roelofs (2008)
19.4	Gantt et al. (2009)
29	Long et al. (2011)
17.2	Vignati et al. (2010)

# Outline



- 1) Atmospheric aerosol deposition as a control of ocean biological productivity
- 2) Biogenic gas emissions from the ocean and their impact on aerosol/cloud interaction and radiative properties of the overlying atmosphere
- 3) Sources, chemical composition and size distribution of ocean-derived primary organic aerosols
- 4) Model results**
- 5) The future



Introduction

Aerosol deposition

Gas emissions

POM emissions

Model Results

Future

# Model Setup

Model	CMAQ V. 4.7
Time Period	June, July, August 2005
Domain	Western US, Pacific coast
Horizontal Resolution	12 x 12 km <sup>2</sup>
Vertical Resolution	14 layers from the surface to 100mb
Emissions	Anthropogenic 2005 NEI Natural: BEIS
Meteorology	MM5
Simulations	<ol style="list-style-type: none"><li>1) Baseline without marine emissions</li><li>2) Isoprene and primary emissions included</li><li>3) “Real” simulation using Southern Ocean (SO) data from Columb et al., (2009)</li></ol>



Introduction

Gas emissions

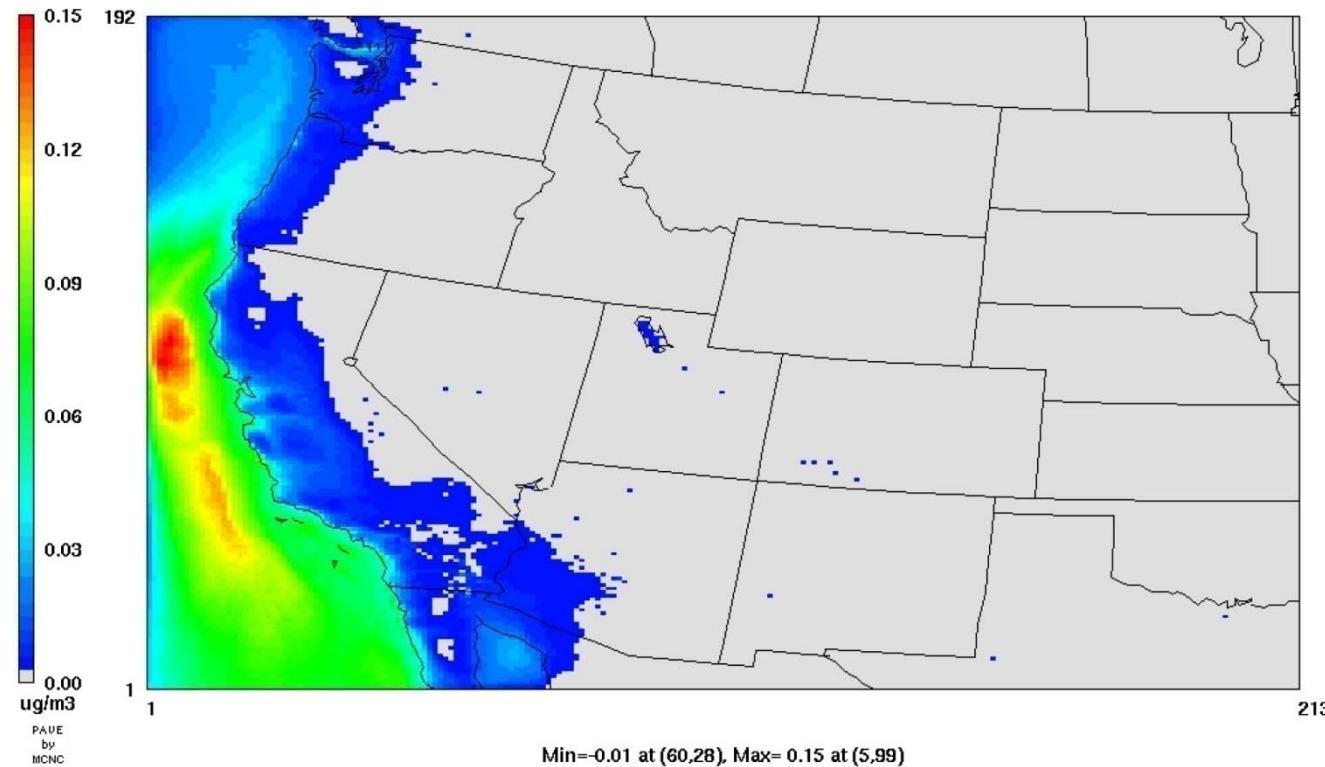
POM emissions

Model Results

Future

# Impact on PM<sub>2.5</sub>

Change in PM<sub>2.5</sub> concentrations due to marine emissions



- ✓ Greatest impact offshore where winds are the strongest
- ✓ Much of the California coast has an average concentration increase of  $0.1 \mu\text{g m}^{-3}$  (still small)



Introduction

Gas emissions

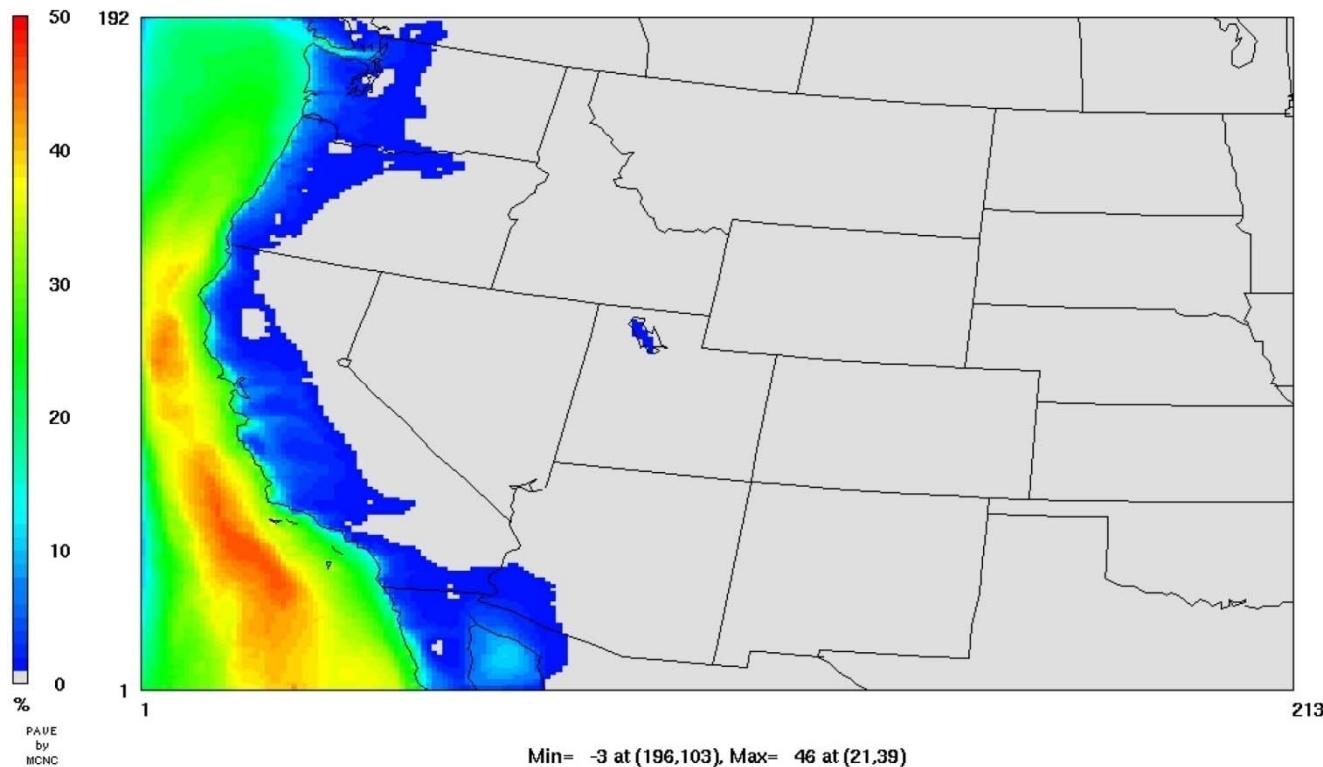
POM emissions

Model Results

Future

# Impact on OC Aerosol

Marine OC Aerosol Contribution to Baseline OC



- ✓ Marine OC aerosol (POM + marine isoprene-derived SOA) may contribute up to 20% of CMAQ predicted OC at the coastal regions and up to 10% inland (not small)



Introduction

Aerosol deposition

Gas emissions

POM emissions

Model Results

Future

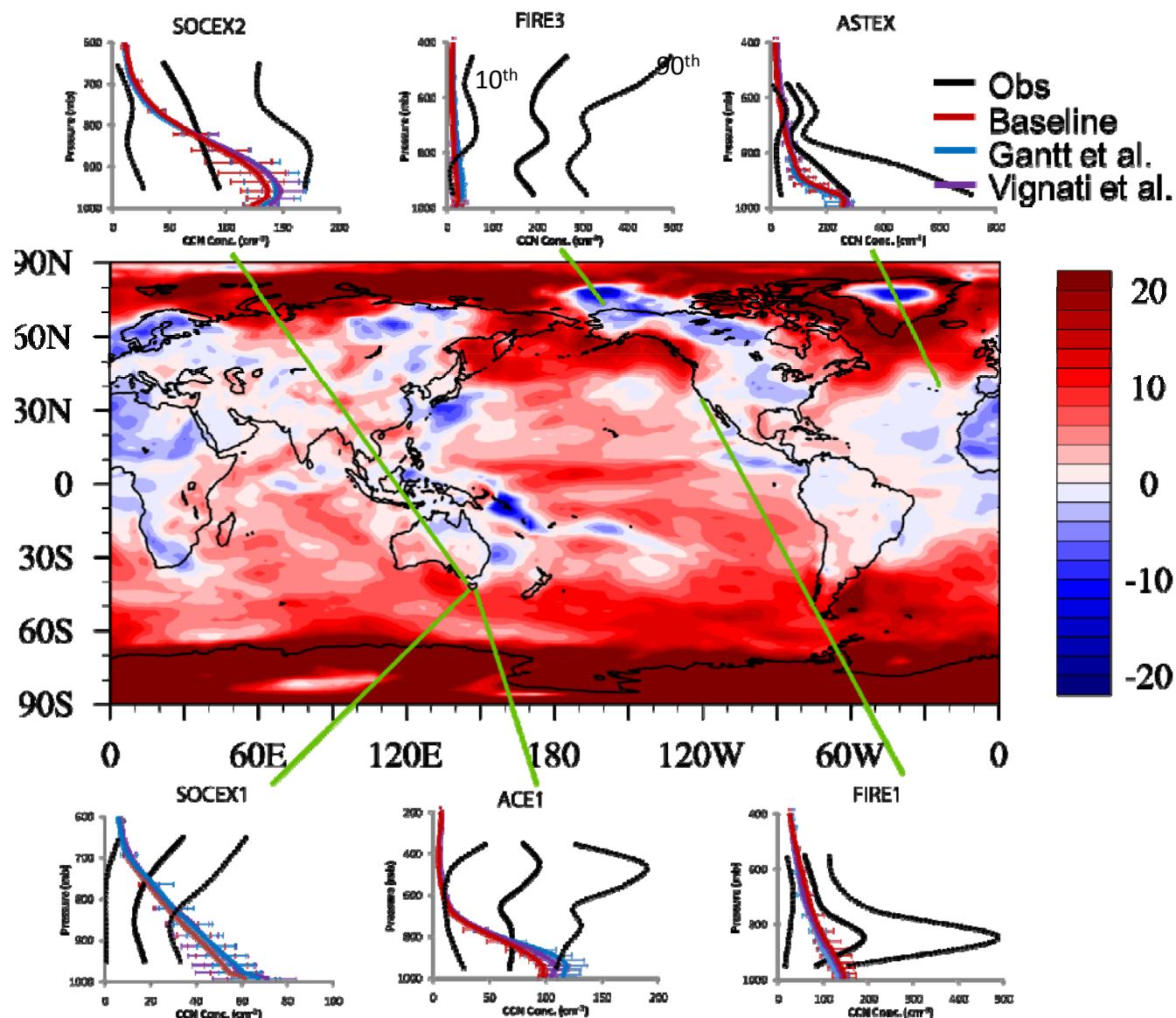
# Implementation Of Marine Carbonaceous Aerosols In CAM5

## Model Configurations

- Horizontal Resolution:  $1.9^\circ \times 2.5^\circ$ ; Vertical: 30 layers
- Aerosol: 5 sub- and 2 super-micron modes
- Simulation: 5 years; Spin up period: 3 months
- Mårtensson et al. [2003] for  $0.02 < D_p < 2.5 \mu\text{m}$
- Gong [2003] for  $2.5 < D_p < 20 \mu\text{m}$
- SOA from isoprene, monotrpenes & MSA ( $\text{CH}_3\text{SO}_3\text{H}$ )

Marine Organic Aerosols in CAM5							
Mode	Accumulation	Atiken	Primary Carbon	Sea salt	Fine Soil Dust	Coarse Sea salt	Coarse Soil Dust
Aerosol component	Sulfate, Ammonium, POM, SOA, BC, Sea salt, <b>Marine POM &amp; SOA</b>	Sulfate, Ammonium, SOA, Sea salt, <b>Marine POM &amp; SOA</b>	POM, BC	Sea salt, Sulfate, Ammonium, <b>Marine POM</b>	Dust, Sulfate, Ammonium	Sea salt, Sulfate, Ammonium, <b>Marine POM</b>	Dust, sulfate, Ammonium

# Difference In Annually Averaged CCN (0.2%)



- Up to 10% higher CCN in marine BL
- Slightly improved agreement with measurements



Introduction

Gas emissions

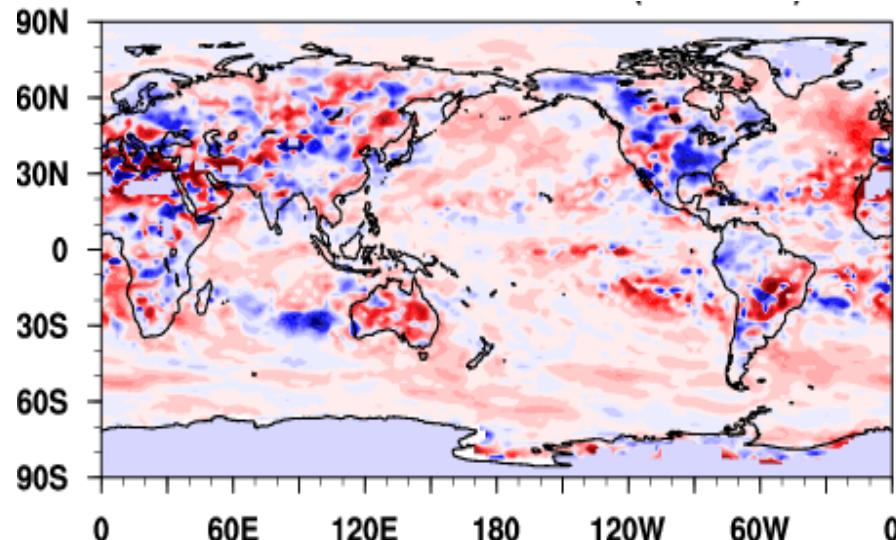
POM emissions

Model Results

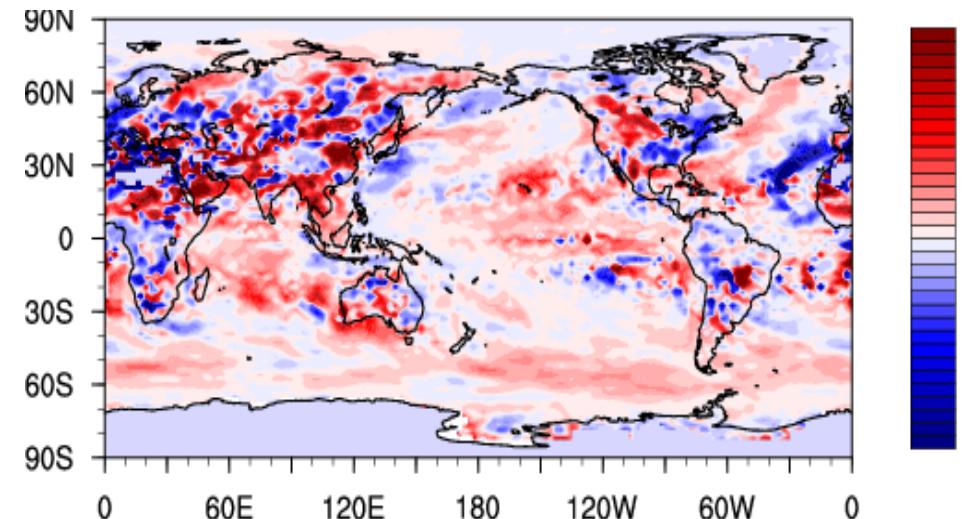
Future

# Change In Cloud Droplet Number Concentration

PI (w/marine – w/o)



PD (w/marine – w/o)



Introduction

Gas emissions

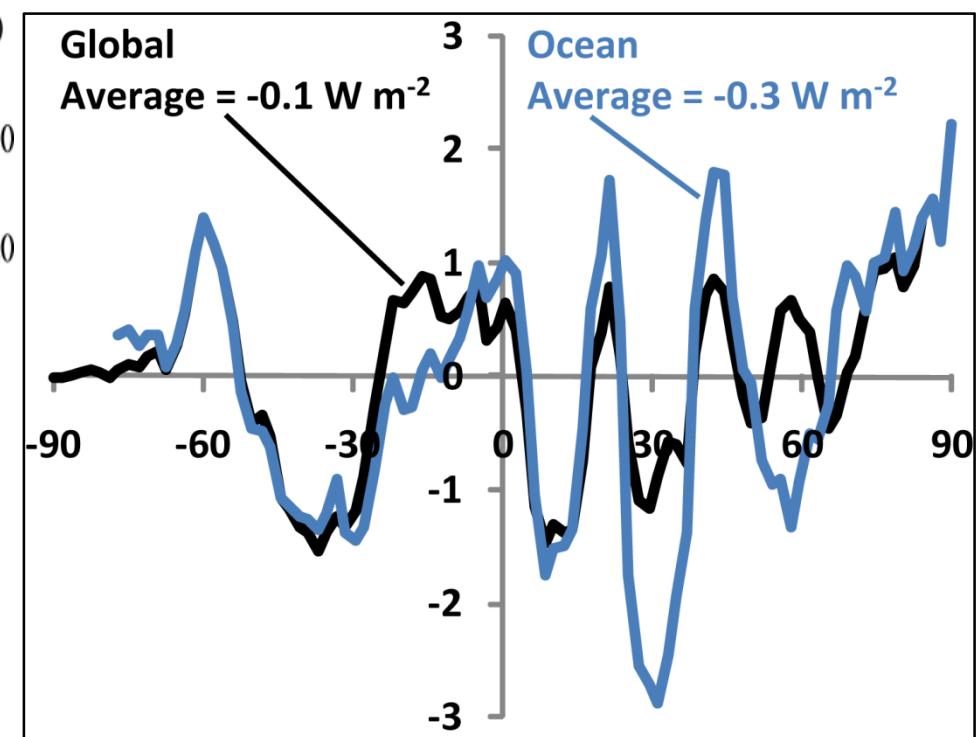
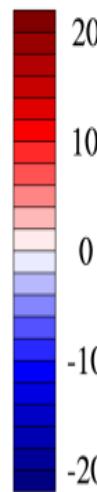
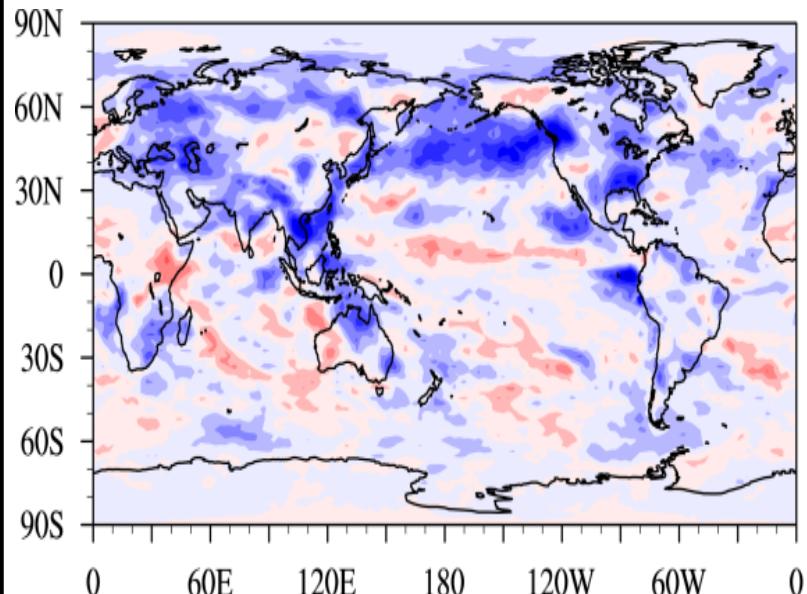
POM emissions

Model Results

Future

# $\Delta SWCF$ (PD-PI)

w/marine [W m<sup>-2</sup>]



Introduction

Gas emissions

POM emissions

Model Results

Future

# Conclusions



Updates to CAM5 resulted in:

- ✓ Increased shallow in-cloud **CDNC [cm<sup>-3</sup>]** from **0** to **1.5 (2.6%)** over the global oceans and from **0.2 (0.6%)** to **2.2 (5.4%)** over the Southern Ocean
- ✓ Increased **LWP [g m<sup>-2</sup>]** from **0.04** to **0.06 (~1%)** over the global ocean and by **0.03** to **0.2 (0.2 to 4%)** over the Southern Ocean
- ✓ Decreased cloud top **r<sub>e</sub>** by **<0.3%** over the global ocean and **<0.5%** over the Southern Ocean
- ✓ Increased global mean **SWCF [Wm<sup>-2</sup>]** from **0** to **-0.3** over the ocean and from **-0** to **-0.1** globally



Introduction

Gas emissions

POM emissions

Model Results

Future

# Outline

- 
- 1) Introduction
  - 2) Biogenic gas emissions from the ocean and their impact on aerosol/cloud interaction and radiative properties of the overlying atmosphere
  - 3) Sources, chemical composition and size distribution of ocean-derived primary organic aerosols
  - 4) Model results
  - 5) The future



[www.shortsshortsshortss.com/.../](http://www.shortsshortsshortss.com/.../)



Introduction

Gas emissions

POM emissions

Model Results

Future

# My Take on Near Future Marine Organic Aerosol Research



**"It is time to go and sample in pristine environments..."**

Jos Lelieveld (MPI Mainz)

- ❖ Extensive lab measurements for phytoplankton emitted biogenic volatile organic compounds (BVOC) under different light/ temperature regimes
- ❖ More field campaigns in pristine marine environments (e.g., Southern Ocean)
- ❖ Improved satellite retrievals of aerosols (low AOD), Ocean ecosystems ([Chl-a], phytoplankton functional groups, CDOM) and trace gases within boundary layer (Sulfur dioxide ( $\text{SO}_2$ ), formaldehyde (HCHO), glyoxal (CHOCHO), nitrous oxide ( $\text{N}_2\text{O}$ ), bromine monoxide (BrO), chlorine dioxide (OCIO), ozone ( $\text{O}_3$ ))
- ❖ Improved quantification of marine POM emissions
- ❖ Marine sources of SOA (e.g., monoterpene such as  $\alpha$ -pinene,  $\beta$ -pinene, myrcene, camphene, limonene and others)
- ❖ Finer model spatial resolution (estuary emissions)
- ❖ Future air quality studies should consider marine sources of OC aerosol



Introduction

Gas emissions

POM emissions

Model Results

Future

# Special Issue on Marine Aerosol-Cloud-Climate Interaction

<http://www.hindawi.com/journals/amet/osi.html>

The screenshot shows the homepage of the Hindawi Publishing Corporation website for the journal *Advances in Meteorology*. The header features the Hindawi logo (two overlapping circles in blue and green), the text "Hindawi Publishing Corporation", a search bar with "Advanced Search" and "Go" buttons, and navigation links for "Home", "Journals", and "About Us". A large banner image of a dramatic lightning storm over a field is prominently displayed. Below the header, there are links for "About this Journal", "Submit a Manuscript", and "Table of Contents". The main content area is titled "Marine Aerosol-Cloud-Climate Interaction" and lists several guest editor articles. To the right, there is a thumbnail image of the journal cover.

**Marine Aerosol-Cloud-Climate Interaction**

Guest Editors: Nicholas Meskhidze, Charles R. McClain, Markus D. Petters, Elisabetta Vignati, Olaf Stetzer, Chris Osburn, and David J. Kieber

- ▶ **Marine Aerosol-Cloud-Climate Interaction**, Nicholas Meskhidze, Charles R. McClain, Markus D. Petters, Elisabetta Vignati, Olaf Stetzer, Chris Osburn, and David J. Kieber  
Volume 2010 (2010), Article ID 250896, 2 pages
- ▶ **Primary and Secondary Organic Marine Aerosol and Oceanic Biological Activity: Recent Results and New Perspectives for Future Studies**, Matteo Rinaldi, Stefano Decesari, Emanuela Finessi, Lara Giulianelli, Claudio Carbone, Sandro Fuzzi, Colin D. O'Dowd, Darius Ceburnis, and Maria Cristina Facchini  
Volume 2010 (2010), Article ID 310682, 10 pages
- ▶ **Polysaccharides, Proteins, and Phytoplankton Fragments: Four Chemically Distinct Types of Marine Primary Organic Aerosol Classified by Single Particle Spectromicroscopy**, Lelia N. Hawkins and Lynn M. Russell

The thumbnail image shows the front cover of the journal issue. It features a dramatic photograph of a lightning strike over a dark, cloudy sky. The title "Marine Aerosol-Cloud-Climate INTERACTION" is printed in white capital letters across the top. Below the title, smaller text reads "Guest Editors: Nicholas Meskhidze, Charles R. McClain, Markus D. Petters, Elisabetta Vignati, Olaf Stetzer, Chris Osburn, and David J. Kieber".

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AMSR-E - [http://www.remss.com/amsr/amsr\\_browse.html](http://www.remss.com/amsr/amsr_browse.html)  
CALIPSO - [http://www-calipso.larc.nasa.gov/products/lidar/browse\\_images/show\\_calendar.php](http://www-calipso.larc.nasa.gov/products/lidar/browse_images/show_calendar.php)  
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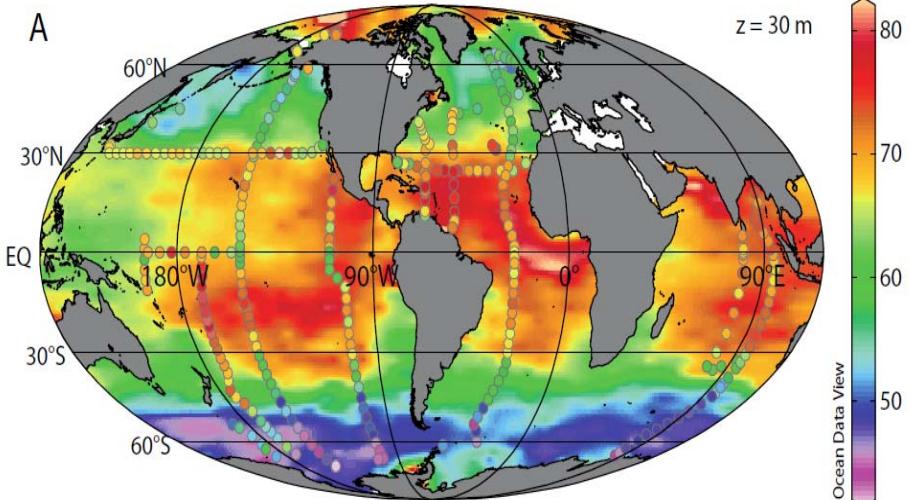
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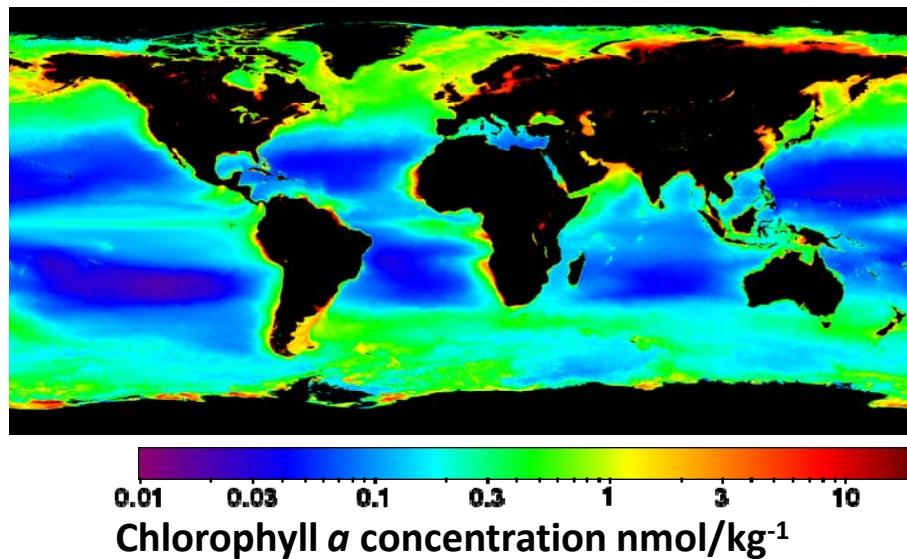
# Ocean Carbon Inventory



**0 – 200 m    DOC =  $47 \times 10^{15} \text{ C}$**

[Hansell et al., Oceanography, 2009]

Carbon inventory of ocean biomass  $\sim 3 \times 10^{15} \text{ C}$



Introduction

Gas emissions

POM emissions

Model Results

Future

# What Are The Right Parameters For Organic Enrichment Of Sea Spray ?

