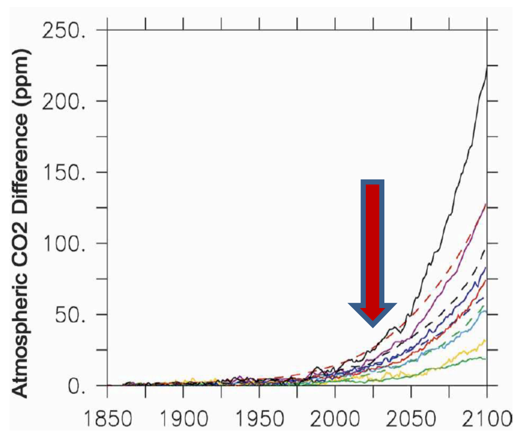
Studies of Aerosol - Ocean Ecosystems - Cloud Interactions by the ACE Mission

Projections of future climate over the next century remain an important scientific goal for much of the earth science community. A large fraction of the uncertainty in predicting climate change at 2100 lies in the uncertainties associated with feedbacks (figure 1) in the carbon cycle (Gregory et al., 2009) and aerosol forcing (Forster et al., 2007). These feedbacks are the result of land-atmosphere-ocean natural and anthropogenic interactions. For example, ocean uptake of carbon dioxide represents a large negative feedback of the carbon cycle onto atmospheric perturbations (Gruber et al., 2009). This uptake can be strongly modified by aerosol inputs of iron resulting in enhancement of nitrogen fixation (Falkowski et al., 1998) and mitigation of iron limitation (Martin, 1990) in large regions of the ocean. In addition, oceans can be strong sources of atmospheric aerosols and in the last 30 years a number of biogenic sources of aerosols have been identified (Shaw, 1983; Charlson et al., 1987; Middlebrook et al., 1998; Leck and Bigg, 2005; O'Dowd et al., 2002; Meskhidze and Nenes, 2006). The combined effect of these marine aerosols results in a climate perturbation through direct obstruction of sunlight as well as through changes to cloud reflective and precipitation properties.



While potentially important, the exact mechanisms and strengths of these feedbacks are poorly understood. For example, current climate warming has caused increased stratification in the upper ocean that may prevent the surface ocean from mixing with the deep ocean, thereby decreasing the nutrient fluxes from below (Levitus et al., 2000; Sarmiento and Gruber, 2006) and affecting oceanic primary production and efficiency of CO_2 transfer (Behrenfeld et al., 2006). This climate-warming induced situation makes oceanic primary production more dependent on the nutrient input from external sources, such as atmospheric Fe deposition. But the same warming may have desertified continents and increased the dust load of air masses whose long range transport eventually locates them over potentially nutrient-deficient ocean waters (Mahowald,2007).

Many questions remain regarding the processes linking the atmosphere and ocean ecosystems under the current climate warming situation:

- What is the flux of aerosols to the ocean and their temporal and spatial distribution?
- What are the physical characteristics and the source of aerosols deposited into the oceans?
- How are the physical and chemical characteristics of deposited aerosols altered in the atmosphere?

- How do ocean ecosystems respond to aerosol deposition?
- What is the spatial and temporal distribution and fluxes of aerosols and gases emitted from the ocean and how are these fluxes regulated by ocean ecosystems?
- What are the feedbacks among ocean emissions of aerosols and gases, microphysical and radiative properties of the overlying aerosols and clouds, aerosol deposition, ocean ecosystems and the Earth's climate, and how is humankind changing these feedbacks?

Atmosphere-Ocean Interactions: What do we know?

The detailed mechanisms and the radiative feedbacks in the earth climate system include mutually interacting processes (Figure 2) many of which remain poorly understood:

Atmosphere to Ocean interactions: Aerosol deposition over the ocean and the impacts on marine productivity. Phytoplankton growth in surface waters is limited by the supply of light and by the availability of various nutrients including nitrogen, phosphorus and iron. It is now clear that nutrients derived from terrestrial sources can play a role in biological activity in all pelagic regions of the global ocean. In high-nitrate, low-chlorophyll regions (HNLC), such as the sub-arctic Pacific Ocean, the equatorial Pacific Ocean and the Southern Ocean, iron availability can be the major factor regulating primary productivity (de Baar and Boyd, 2000). It is also possible that other trace metals delivered to the oceans primarily from the atmosphere (e.g. Mn, Co, and Zn), may influence primary productivity in some regions and times. Interactions also exist between phytoplankton and atmospheric nitrogen deposition. One of the most significant recent advances was the experimental demonstration of the importance of iron supply in regulating key biogeochemical interactions and feedbacks between the ocean and atmosphere (Martin et al., 1994; Coale et al*.,* 1996; Boyd et al., 2000). While dust is globally a rather small fraction of the total iron input to the ocean (typically<2% of the total deposited iron is available to primary production, (Jickells and Spokes, 2001)), it is disproportionately important in open ocean waters where availability

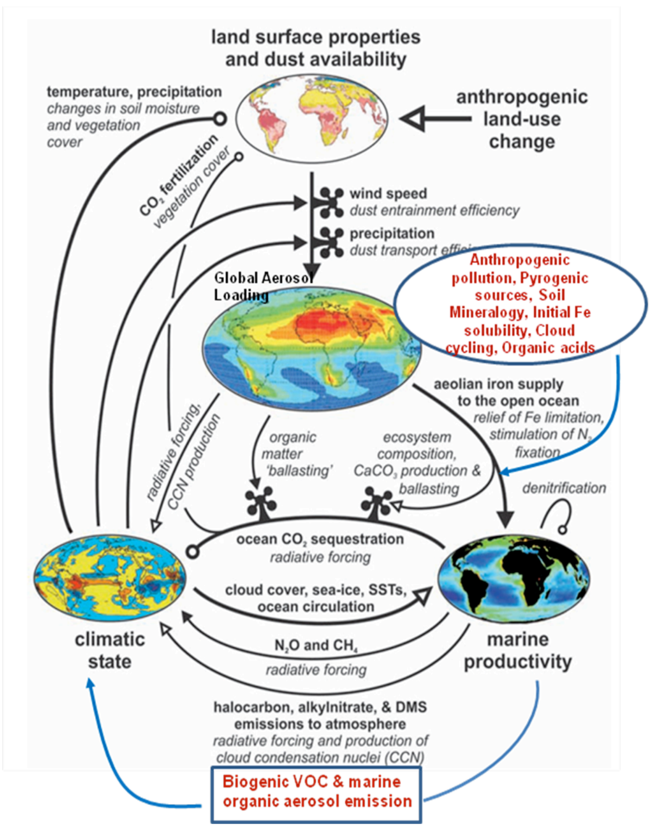


Figure 2: Diagram illustrating the different atmospheric, ocean and land interactions and feedbacks (Adapted from Jickells et al.,2005)

of micronutrient iron might limit phytoplankton productivity (Jickells et al., 2005). In general, the highest atmospheric concentrations of dust over marine areas are found in the Northern Hemisphere (e.g. over the tropical North Atlantic Ocean, the northern Indian Ocean including the Arabian Sea, and the western North Pacific Ocean (Duce et al., 1991)). Season, vegetation and soil aridity in the source area , however, modify the pattern and magnitude of delivery of dust containing iron varies (Mahowald et al., 2009). Dust deposition depends on natural climate variability, human land disturbance, local and regional scale weather and global atmospheric circulation. Changes in atmospheric inputs of dust will likely impact phytoplankton processes and in turn, alter the exchange of climatically important and biologically produced trace gases between the atmosphere and oceans, providing potential climate feedbacks.

Accurate measurements of dust and associated nutrients deposition over the ocean are very difficult. Dust is distributed unevenly, with lowest deposition to ocean regions remote from land (these approximately correspond to the HNLC areas). During atmospheric transport of aerosols, iron solubility can be changed due to its interaction with natural (Zhu et al., 1993) and anthropogenic trace gases (Solmon et al., 2009), cloud cycling and photochemical reactions (Fan et al., 2006). Combustion processes may also be a good source of soluble iron to the oceans (Mahowald et al., 2009). Details of the chemistry and photochemistry of iron in aerosols and cloud droplets are minimal to date (Journet et al; 2008). Further, once in seawater, the oxidation conditions of the upper waters and iron chemical state may influence iron availability to primary producers (Johnson et al., 1997).

In addition, there are a number atmosphere to ocean processes for which there is very little knowledge but initial studies suggest they are potentially important. Significant quantities of nitrogen species are delivered from the land to the ocean via the atmosphere (Duce et al., 2008). Much of this atmospheric nitrogen is from anthropogenic sources (primarily the combustion of fuels and the utilization of fertilizers), and it is subject to future changes, both in amount and geographical distribution, depending on population and industrial growth in various regions. Delivery of atmospheric nitrogen to coastal regions in Europe and North America is estimated to have increased by 50% to 200% during the past 50 years (Paerl, 1995). This deposition increases pressure on coastal ecosystems already stressed by a wide range of other human activities. As humans significantly perturb the nitrogen cycle, limitation by phosphorus becomes more likely. While less well studied, the impact of riverine and atmospheric phosphorus and human perturbations to these processes may also be important (Mahowald et al., 2008). Observations suggests atmospheric deposition of phosphorus can influence ocean ecosystems (Mills et al, 2004) and depending on toxicity, atmospheric deposition can also potentially inhibit phytoplankton growth (Paytan et al.,2009).

Marine biology to Atmosphere interactions: Production of marine aerosols and impacts on clouds. There are significant gaps in our understanding on how clouds self-regulate and adjust in the remote marine environment. The understanding of this balance is crucial because the global radiative balance is dominated by clouds over the oceans and any anthropogenic impact will be changing this balance. Following Shaw (1983), the CLAW hypothesis (Charlson et al., 1987) proposed a role for the marine biogeochemical sulfur cycle in altering global climate. An increase in ocean DMS emissions could lead to increases in CCN resulting in longer-lived clouds with higher droplet density (enhanced albedo). An increase in global albedo in turn leads to less solar radiation reaching the sea surface, thereby mitigating the effects of climate change. This DMS-CCN-albedo feedback could act as "planetary thermostat"- a negative feedback loop which would tend to stabilize the climate against perturbations such as warming due to anthropogenic production of greenhouse gases. However, the sign (direction) of the feedback modulation was left ambiguous and it remains uncertain today whether such feedbacks exist or what their strengths might be. Although it is well established that clouds with more particles have a higher albedo, the optical impact of various types of clouds on the underlying ocean biogeochemistry is still poorly understood and very few observational constrains exist for model simulations. Although the study of the sulfur cycle provides a plausible explanation for marine cloud modulation, alternative sources of aerosols with markedly different chemical properties have recently been found and proposed to act synergistically with the established mechanisms, leading to changes in marine aerosol chemical composition and to important impact on clouds (O'Dowd et al 2002; Meskhidze and Nenes, 2006; Facchini et al. 2008).

Release to the atmosphere of highly reactive trace gases by the marine ecosystem. The ocean is a source of trace gases, some which are known to have climatic impacts. For example, the ocean is ubiquitously supersaturated with CO_2 with respect to the atmosphere. However, the total annual emission to the atmosphere is small compared to current estimates from both terrestrial natural and anthropogenic sources (Bates et al., 1995). Non-methane hydrocarbons are also produced in surface seawater possibly by photochemical mechanisms, phytoplankton activity and/or microbial breakdown of organic matter (Bonsang et al., 1988 and 1992; Yassaa et al., 2008; Colomb et al., 2009). While it has been shown that some of the ocean produced biogenic volatile organic compounds (BVOC) can lead to the formation of secondary organic aerosol, and therefore influence the radiative properties of overlying atmosphere, the significance of ocean-atmosphere flux of BVOCs needs to be further explored. In addition, methyl halides are produced and consumed biologically and photochemically in surface ocean waters (Cota and Sturges 1997; Moore and Wang 2006). Many halogenated gases have climatic implications through their chemistry or radiative effects, especially in polar regions (Barrie and Platt, 1997). When gases are produced and destroyed in seawater and exchanged with the atmosphere on similar time scales, their exchange with the atmosphere can be controlled in part by their biogeochemical cycling in seawater.

Approach proposed

The detailed mechanisms and the radiative impact feedbacks in the earth climate system are best understood through the combination of in situ data, satellite remote sensing and models. For the problem of aerosol-ocean interactions, a new satellite mission is required to provide the increased number of parameters and improved signal resolution necessary to advance our understanding of these important processes and to improve future projections of climate. This satellite mission will be closely tied to field

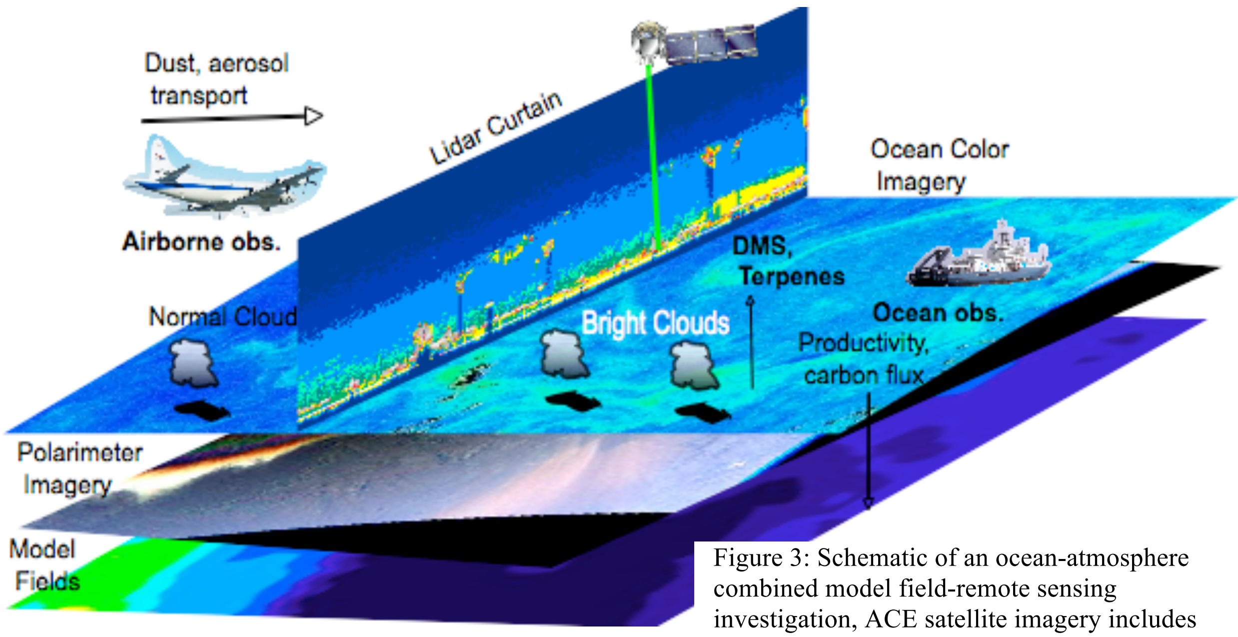


Figure 3: Schematic of an ocean-atmosphere combined model field-remote sensing investigation, ACE satellite imagery includes ocean color cloud and aerosol columnar and profiling characteristics. Concurrent field measurements can include trace gases such as DMS and precursors, and various cal/val parameters, as well as subsurface ocean measurements and nutrients/ hydrography. Field sampling can be guided by concurrent imagery and model outputs to provide context, such as the mesoscale field depicted. *(SeaWiFS, POLDER, and CALIPSO example imagery shown here)*.

studies and model development, to maximize the scientific impact of the satellite data collections (Figure 3).

Satellite measurements of appropriate aerosol and ocean ecosystem properties are required as well as supporting measurements for understanding their changes. For aerosol properties these include aerosol type (dust, smoke, etc.), optical thickness, complex index of refraction, and height and size distributions with a 2-day global coverage to resolve temporal evolution of plumes. Although oceanic aerosol sources appear to produce aerosol and gas concentrations in the near noise level of existing satellite platforms, estimates of natural biogenic concentrations over the ocean are essential.

For ocean ecosystem properties these include phytoplankton functional type and pigment absorption spectra, colored dissolved organic matter (CDOM) absorption, total and phytoplankton carbon concentration, ocean particle size distribution, phytoplankton and CDOM fluorescence, phytoplankton growth rates and rates of net primary production. Many of these determinations can be made by sampling the top of the atmosphere radiance spectra and polarized radiance spectra for selected UV, visible and SWIR bands. Active (lidar) measurements of aerosol properties along the orbit track are needed to refine height distribution and composition and to provide independent measurements of ocean particle scattering and its vertical distribution within the water column. Many supporting satellite measurements are needed to assess environmental conditions affecting aerosols and hydrosols including sea surface temperature, wind speed and direction, ice cover, humidity and temperature profiles and precipitation rates. In particular, measurements of drizzle detection and precipitation rates coincident with the ACE lidar and polarimeter observations are required supporting the need for a precipitation radar as a component of ACE. It is envisioned that many of the other supporting global products will come from operational satellite assets such as NPOESS or other Decadal Survey missions.

Simultaneous determinations of tropospheric concentrations of several trace gas species will be important for linking ocean – aerosol processes. These species include but are not limited to formaldehyde (CH_2O), glyoxal ($C_2H_2O_2$), IO, BrO, NO_2, and SO_2. It is hoped that these determinations will come from future satellite systems like the GeoCAPE mission to be deployed in geostationary orbit and is in the second tier of NASA's Decadal Survey plans. Further these observations will be available in a global form from the Global Atmospheric Composition Mission (GACM) which is in the third tier of the Decadal Survey plans.

Field observations are considered an integral part of the ACE mission from the pre-launch period onward. In situ measurements fulfill the dual role of calibrating and validating satellite sensors and product retrievals, and making essential observations that are not possible from satellite instruments. Field measurements will range from solar radiation observations, to in-water chemical, biological, and optical properties, to chemical characterization of aerosols. The unique capabilities of the ACE mission flight instrumentation will also require ongoing development of new and improved field measurements. Two types of field campaigns are envisioned: sustained time-series observations from fixed locations (e.g. the BATS and HOT oceanographic time-series sites, and the AERONET sunphotometer network) and mobile sites (Marine Aerosol Network, Smirnov et al., 2009), and intensive field campaigns to address particular science questions. Both types of field campaigns will contribute valuable data for calibration and validation as well as required data to answer the focused questions raised in the Science Traceability Matrix.

Some possible topics of field campaign studies that address the questions of the aerosol-ocean interactions STM include:

- Southern Ocean and DMS – A Southern Ocean (SO) study would be on the dimethylsulfide - cloud connection. Given that oceanic gases are probably the dominant CCN precursors over the SO, this study is potentially of the greatest climatic significance.
- North Atlantic Bloom Aerosol Production – A study focusing on comparing/contrasting the atmospheric imprint of coccolithophore and/or *Phaeocystis* blooms, and examining the hypothesis that the North Atlantic bloom is a major source of fine particle organic aerosols.
- North Pacific Asian Outflow Impact – An examination of the impact of Asian dust and pollutant outflow on oceanic productivity, trace gas emissions, and aerosol/cloud properties.

Because cloud cover in high latitude regions limits ocean color and aerosol satellite retrievals, the field campaigns with cooperative efforts are likely to be key components.

To this end the ACE mission science team should interact with national and international coordinating groups such as the SOLAS (Surface Ocean Lower Atmosphere Studies) and OCB (Ocean Carbon and Biogeochemistry) working groups in order to select and plan process studies as described above.

Another major element of this approach is the role of model simulations. Models permit to explore hypothesis about the processes controlling dust deposition, such as the human perturbation to dust over the anthropocene (e.g. Luo et al., 2008) or controls on soluble iron deposition over the oceans (Solmon et al., 2009). In addition, models allow us to explore the impact of changes on the climate system (e.g. the impact of changes in dust input on ocean biogeochemistry (Moore et al.,2006), which is not possible to quantitatively explore with only observations. Finally, models can explore and identify feedbacks in the system, which is not possible otherwise. While models are not perfect, they are a valuable tool for understanding atmosphere-ocean interactions, and will be utilized within the ACE-framework.

Impacts and Relevance

Over the past decade, in situ measurement, satellite remote sensing and modeling efforts have substantially improved our understanding of the temporal and spatial distribution of aerosols and trace gases, their physical and chemical characteristics and controlling effects on ocean ecosystems. However, past research has also revealed inherent complexity of aerosol-ocean ecosystems-cloud interactions with multiple forcings and feedbacks. Narrowing the gap in the current understanding of anthropogenic and natural contribution to a changing climate will require development of new space-based, field, laboratory instruments and modeling capabilities. This should include execution of focused field studies examining the aerosol fluxes to and from the ocean and subsequent changes to marine ecosystems in various oceanic regimes around the globe. By expanding available satellite-borne sensors to allow encompassing aerosol forcing of ocean biological systems and cloud processes, it will be possible to capture some potentially important feedbacks with implications on atmospheric radiative effects and climate. Models, in addition to represent current climate, will be able to better capture the changes that have occurred over the past century and predict the climate changes that would result from different future emission strategies. Achieving such confidence critically depends upon more realistic simulations of the aerosol- ocean ecosystems - cloud system with forcings and feedbacks operating on multiple spatiotemporal scales.

The societal implications are important. Current acidification of the ocean and its biological adaptation are a response to climate change. Yet, our current estimations of future climate effects are based on model approaches where many of the feedbacks processes are not included or poorly described. The few climate models that do incorporate more realistic feedback processes show significant impacts in future projections of surface temperatures (figure 1). The ACE mission will have an essential role in improving climate predictions by providing information of processes that are poorly constrained in climate models.

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