FOCUS 2

Exchange Processes at the Air-Sea Interface and Role of Transport and Transformation in Atmospheric and Oceanic Boundary Layers

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The first section of the plan outlines a set of field campaigns that provide the platform for the required experiments to be conducted. The second section details the fieldwork, modelling and remote sensing needed in these experiments. The proposed field campaigns are part of the nine fieldwork campaigns envisaged across all three SOLAS Foci over the next 10 years. The proposed fieldwork locations are not meant to be prescriptive, but instead to allow the broad sweep of experiments called for in this plan to make use of common infrastructure.

Introduction

The SOLAS Focus 2 objective is to gain a quantitative understanding of processes controlling the air-sea exchange of mass, momentum and energy to permit accurate simulations of regional and global gas and aerosol fluxes. This requires establishing the dependence of these interfacial transfer mechanisms on physical, biological and chemical factors within the atmospheric and oceanic boundary layers, and the transport and transformation processes that regulate these exchanges. This implementation plan proposes activities over roughly 10 years, and will evolve with our needs and abilities.

A successful completion of the SOLAS Focus 2 goals and objectives will require development of new infrastructure and technology to complement our present capabilities. The implementation of the programme will be achieved with a combination of process studies, sustained time-series observations, and model-observational comparisons and reconciliation. Regional and seasonal air-sea fluxes of climate relevant compounds (CRCs) will be studied through international coordination between ocean and atmospheric physicists, biologists and chemists.

Knowledge of mechanisms underlying air-sea exchange is essential to create realistic numerical simulations of biogeochemical and physical processes and feedbacks within the context of changing forcing. The large range of scales associated with the mechanics of air-sea exchange (from micrometer to mega meter) necessitates that such models contain as much physics as possible, and that any unavoidable parameterisations are based on physical and biogeochemical principles. If these parameterisations are not well founded, the models will have limited skill and predictive capacity with respect to climate or other environmental changes. Such improvement requires quantitative measurements of the exchange (including wet and dry deposition) of number concentration (aerosols), mass (gases, aerosols and water vapour), momentum, and energy (including heat) across the air-sea interface, as well as the biogeochemical and physical parameters that characterise the interface and drive the processes. Simultaneous study of the physics and biogeochemistry of the air-sea interface will lead to improved understanding of biogeochemical fluxes, momentum, and energy. SOLAS Focus 2 aims to significantly improve the numerical simulation of air-sea exchange processes, thus allowing more accurate estimations of regional and global air-sea flux fields within Foci 1 and 3 Activities.

An important goal of Focus 2 is to learn enough about the exchange mechanisms so that straightforward measurements of mean quantities can be used to infer air-sea exchange on regional and global scales. This will require a coordinated programme of comprehensive field observations, laboratory experiments and modelling, including small-scale studies of transport near and across the air-sea interface. It should include turbulence resolving models as well as larger-scale modelling of mesoscale transport extending through both the oceanic and atmospheric boundary layers. In the atmosphere, the effects of boundary-layer clouds on transport, wet deposition and chemical transformation also need to be addressed. Remote sensing will be used for measurements of halogen oxides, aerosols (sea spray, dust, biomass burning aerosol, etc.), whitecaps, and sea surface characterisation (wave height, wind, sea surface roughness, SST, chlorophyll).

The SOLAS Science Plan identifies deficiencies in our understanding of gas and material transport in each of the layers into which the MABL can be roughly divided: the "wave layer" adjacent to the interface; the "constant flux" layer where concentration gradients are significant and sea-spray intense during high winds; the "mixed layer" of MABL-scale motions and vertical convective transport, with the whole capped by clouds whose character and influence are cross-linked to processes going on below.

The vertical continuum of transport processes implies a need to characterise the MABL through its entire depth, and to consider the spatial variability of both atmosphere and ocean over a region commensurate with this scale. In practice, this suggests an experimental campaign involving the complementary sampling strategies for ships and aircraft, and raises the possibility of combining with similar intensive field programmes suggested in Activity 2.1. Experiment 1 of Activity 2 is designed to study the air-sea transfer process to develop parameterisations of air-sea gas and aerosol fluxes and, among other things, to intercompare geochemical and micro-meteorological methods for determining transfer velocities. Experiment 2 aims to understand the relationship between environmental variables (e.g., wind stress, wave characteristics, precipitation) and the forcing mechanism for air-sea gas and aerosol transfer. The observational requirements of Activities 2.1.1 and Activity 2.3, are clearly complementary and interdependent, so implementation strategies should take advantage of this. While more closely targeted studies of specific gas and aerosol transfer processes both at the interface and in the MABL may follow, it is suggested that such a major joint field campaign should be undertaken early in the life of SOLAS.

The MABL observations should be designed to address the science questions set out above in support of improved BL models. If possible they should include observations to help understand the influence of rainfall on the transport of gas and aerosols. In addition to the measurement of basic environmental variables, the routine direct determination of physical surface fluxes (energy, momentum, and radiation) should accompany those of the gas and aerosol species under investigation. Other physical processes that need attention are the long-standing problem of the connection between wind, waves and surface roughness; the validity over the ocean of flux/gradient relationships and turbulence parameterisations developed over land; and diurnal warming of the upper ocean. These are areas where our understanding of the physical processes is inadequate, and to which the transport of gases and aerosols is closely linked. For example, heat and momentum fluxes are parameterised in the COARE version 3.0 bulk flux algorithm (Fairall et al., 2003), so the data obtained would enable the improvement of the algorithm. Field experiments should be designed to test and improve the transfer velocity parameterisation of Fairall et al. (2000).

Proposed Fieldwork Campaigns and Detailed Recommendations for Focus 2 Experiments

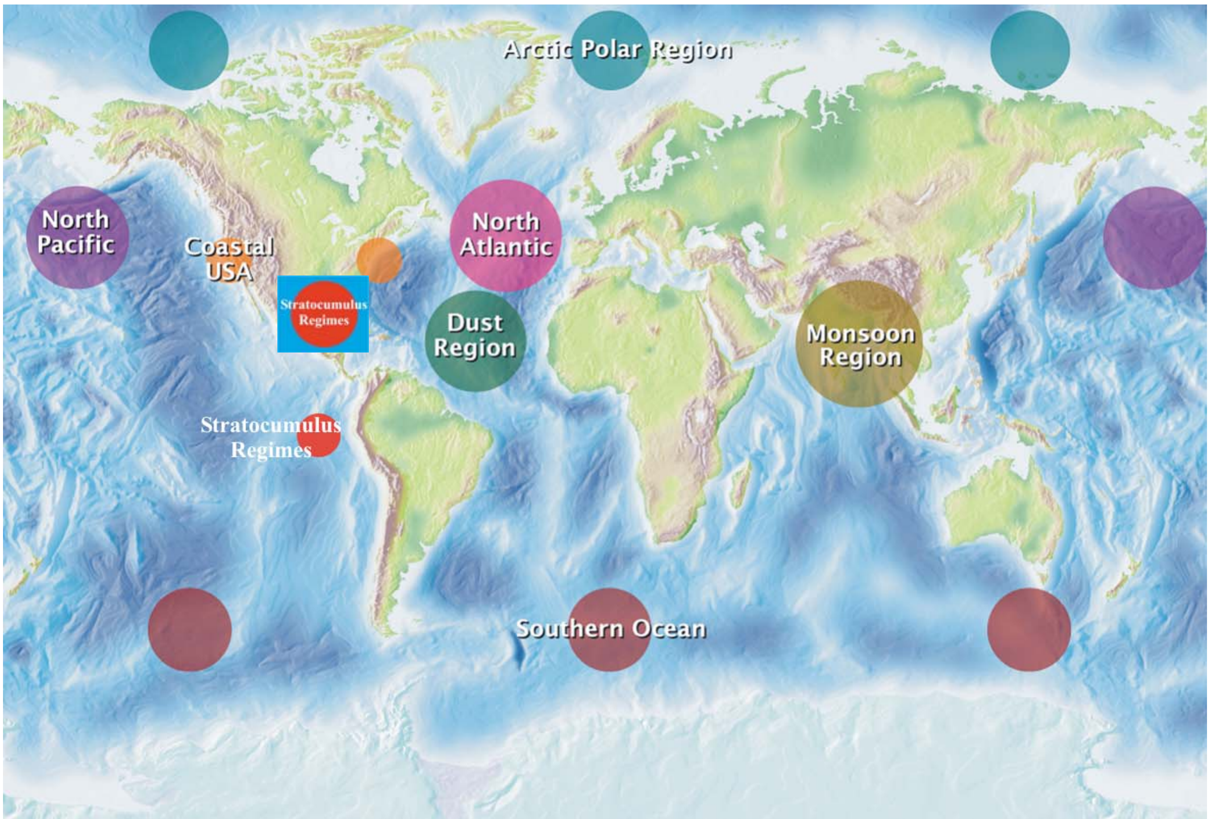

Figure 1: Global provinces for air-sea interactions studies.

1. Regional air-sea exchange campaigns

SOLAS envisages nine major field campaigns spanning all three foci during its 10-year lifetime (see introduction). These will combine sustained observations, intensive process studies, remote sensing and modelling. Focus 2 will have strong role to play in at least seven of these campaigns (Figure 1), which are designed to allow the broad sweep of experiments called for in this plan to converge. In particular, a set of regional air-sea exchange experiments would allow investigation of non-wind speed forcing of air-sea exchange such as surfactants, fetch, rainfall regime, wave field and bubbles, which are often geographically unique.

The studies should be done in areas where there is a significant flux of climate relevant gases and where the "auxiliary" forcing is distinct and separable from wind forcing. Seven target studies are described that take advantage of infrastructures, or other programmes that have a strong interest in flux parameterisations.

The scope of the studies is ambitious and provides infrastructure and synergism with several other efforts within SOLAS and other international programmes. It is envisioned that these studies would occur in concert with other regional process studies. Table 1 provides a list of synergistic process studies for the gas exchange efforts advocated here.

Table 1: Description of global provinces for air-sea interactions studies.

Study Region	Infrastructure	Processes	CRCs	Programme Linkages
Coastal	Sustained-tower fluxes	Surfactants; Fetch; Bottom-boundary friction; air-sea temperature difference; stratification; heterogeneity.	CO_2 fluxes; CH_4 fluxes; Halocarbon fluxes	LOICZ II; Carbo Europe; NACP; OCCC; CA and US ORION, EU-FP6
Southern Ocean	Large buoys may need to be developed and deployed.	High wind; Swell; understand unknown CO_2 variability.	CO_2 fluxes Bubble mediated flux Aerosol	SO CLIVAR
North Pacific	VOS; Buoys.	Coastal upwelling; Ekman cells	Bubble mediated fluxes; Aerosol	The OOI; NEPTUNE; NACP
Monsoon Asia	Asian monitoring systems and Research Vessels.	Coastal upwelling; Seasonal forcing; Capture Episodic events; riverine inputs.	CO_2 fluxes; N_2O fluxes	ENSO
Tropical Pacific	High heat fluxes, high CO_2 outgassing.	Persistent low winds; ENSO, Often oligotropic;	CO_2 fluxes	TAO, ENSO
Polar Regions		Transport in polynas, leads, ice melts.	DMS, CO_2, BrO, Ozone, POPs, aerosols	IPY; OASIS
Stratocumulus regimes	VOCALS radiator fin, aircraft, buoys	Particle formation, CCN. Light/DMS response	DMS, Halogens, Aerosols	CLIVAR/VAMOS

The study design will have many commonalities including: the need to capture a range of forcings; measurement of gas fluxes with micro-meteorological support; independent means to constrain gas fluxes through mass balance approaches; and accurate characterisation of the forcing parameters. Comprehensive background information should be available or should be obtained prior to the studies to characterise the scales of variability and heterogeneity in surface concentrations and forcing. Remotely sensed information is particularly useful in this regard.

The studies also have commonality in experimental design in that they should cover a representative spectrum of spatial and temporal variability encountered in the regime. The processes to be studied are often strongly non-linear such that episodic events can have a disproportionate effect on the fluxes. Several of the designs include a nested approach of intensive (shipboard) studies at locations of sustained flux and surface water measurements on autonomous platforms. Measurements from the autonomous platforms should cover one to two years to capture phenomena associated with the seasonal cycles such as winter storms, monsoons and spring blooms.

a. Atlantic coastal study

The heterogeneous nature and local focus of coastal studies have hampered quantitative and integrative understanding of this region. Programmes such as LOICZ I and II have made major inroads into upscaling and extrapolating biogeochemical fluxes. However, one area where SOLAS can make a unique contribution is improved constraints of air-sea fluxes through development of robust algorithms.

The gas fluxes from coastal areas are poorly quantified. The air-water disequilibrium anomalies of climate relevant gases such as CO_2, CH_4, and N_2O are often large due to enhanced biological productivity and remineralisation in water column and sediments. Gas fluxes are not well constrained with magnitude and sometimes even the directions of fluxes on regional scale are in doubt.

Coastal regimes have several unique features that will likely yield different flux algorithms compared to the open ocean. Several of the parameters that affect coastal gas transfer, such as fetch and surfactants, also operate in the open ocean but at smaller scales. Therefore the information gleaned in the coastal area where certain processes dominate will lead to refinements in open ocean bulk algorithms.

Surfactants are known to have a significant effect on gas transfer by suppressing formation of capillary waves. For the same reason, surfactants also have a significant effect on primary marine aerosol formation, while in addition the surfactants are concentrated at the bubble surface and the ejected film and jet drops are significantly enriched with surfactant material. Surfactants are ubiquitous in the coastal ocean such that surfactant effects will be pronounced. Surfactants are a generic description of a large number of compounds with varying levels of hydrophobicity. Studies should incorporate determination of key physical parameters of the surfactants in conjunction with good measurements of surface stress, near surface turbulence and (capillary) wave field. Because of the ephemeral nature of surfactants and patchiness, direct flux measurements with high frequency are essential in this context.

The coastal environment is heavily influenced by fetch effects. Depending on location and wind direction fetches can range from below 1km to 1000s of km. There is a dramatic influence of fetch on friction velocity and presumably air-sea gas fluxes. The same applies for sea-spray aerosol. Wind direction is the controlling factor on fetch effects and direction can change on daily scales. Therefore direct flux measurements at high frequency are again the preferred method of determining air-sea fluxes as well as sea spray aerosol.

The protocol for the execution of experiments must take the small scales of variability into account and preferably capture the full range of forcing. A design that includes an Eulerian component to capture the temporal variability over month to year timescales along with a survey "intensive" component to address spatial variability is recommended. The design follows the successful ASGAMAGE study but with measurements covering longer timescales. Utilizing a tower for the Eulerian component is optimal from a logistical perspective as it provides a stable platform for micrometeorological and near surface measurements and many towers have sufficient power for sustained measurements. An optimal time scale to capture most scales of variability would be two years. Several surveys should be undertaken to capture the spatial variability during the study. During these intensives, measured frequency and measured parameters should increase to the level of the shipboard campaign.

Tower measurements for the two-year observation period should include surface water measurements temperature, salinity, pCO_2, DMS, chlorophyll and O_2. High-resolution meteorological measurements should be taken, including radiation measurements. Continuous flux measurements of DMS, CO_2 and aerosols should be performed by one or more of the micrometeorological techniques.

During the intensives both the tower and ship should augment these measurements with surface and profile measurements of nutrients, inorganic carbon, and productivity estimates. Measurements on tower and survey should include concentration of CO_2 and DMS in bulk surface water at 10 minutes or less. Automated water profiling systems would be advantageous.

As with all the proposed gas exchange studies, the framework is such that many of the efforts outlined in other sections of the implementation plan can be performed synergistically, augmenting the focus of the studies that aim to provide robust physically based parameterisations of gas transfer with environmental forcing. The North Sea and North Eastern US continental shelf are suggested as prime locations because of the accessibility of towers and the proximity to major research institutions.

b. Southern Ocean study

The Southern Ocean has several unique characteristics that warrant dedicated studies. It is a region with frequent high wind events and long fetch, which leads to large swells. The remoteness of the region has made it one of the last unexplored frontiers in oceanography. Many of the discrepancies and inconsistencies in mass balances and biogeochemical rate estimates are in this region. Numerical global circulation and biogeochemical models are very sensitive to parameterisations of processes within the Southern Ocean. Of particular note for SOLAS is the significant discrepancy between uptake estimates of CO_2 in the Southern Ocean determined from atmospheric and oceanic (inversion) models and measurement based estimates obtained from estimates of ΔpCO_2 (charge in fugacity between air and sea at the interface) and gas transfer velocity with the direct methods, which yield significantly higher estimates. The direct estimates suffer from dearth of pCO_2 observations and lack of a firm knowledge of gas transfer velocities in this energetic region.

The process study to parameterise transfer velocities in the Southern Ocean will be centred on the deployment of a large surface mooring as part of the CLIVAR and GEO mooring network. The proposed 12m diameter mooring with possible diesel generators will have sufficient space and power to install the relevant flux and marine air and surface water measurements. Ship based process studies providing higher resolution and a larger suite of measurements will provide proper details for interpretation.

To elucidate and quantify the controls on gas transfer at high winds from the buoy, accurate wind and friction velocity measurements must be made along with pertinent and automated measurement of wave parameters such as wave height and period. Much of the instrumentation for measurement of physical forcing is available as part of the high resolution meteorological packages such as the ASIMET system (http://uop.whoi.edu/) and would be provided by the main project. Robust gas flux measurement systems with low power requirements that can be operated autonomously for 6 to 12 months must be improved. Autonomous measurement of CO_2 fluxes by eddy correlation and eddy accumulation methods should be attainable with a modest development effort. Instrumentation for measurement of DMS fluxes and fluxes of other climate relevant gases are not at the level for sustained autonomous measurements and would have to be done during the ship component of the study. Water column measurements should include CO_2, DMS, O_2 and total gas tension. The last two parameters can be used to assess the effect of bubble dissolution and gas transfer.

Performing the ship component as part of an iron fertilisation study as recommended in the SOLAS Focus I Implementation Plan would be an optimal utilisation of resources and would provide several important assets to the gas exchange studies. By adding ^3He to the SF_6 tag of the fertilised patch, the gas transfer velocity can be determined over 1 to 3 day timescales, offering a strong constraint to the higher frequency but noisier direct flux measurements (as done in the New Zealand SAGE experiment). The fertilisation also will cause a drawdown of CO_2 improving the fidelity of the direct CO_2 flux measurements. Previous iron fertilisation studies have also shown significant increases in DMS, halo and hydrocarbons such as terpenes. Flux measurements of these compounds by gradient and conditional sampling techniques will provide additional constraints on the effect of solubility and diffusivity on gas exchange, particularly in the presence of bubbles.

c. North Pacific study

The proposed NEPTUNE cabled network provides an excellent opportunity for deployment of sensors with significant power and data transfer requirements. The funded Canadian deployment and the proposed US design will cross the coastal upwelling regime that is characterised by large concentration anomalies of climate relevant gases. The region also experiences strong currents where the effect of wind-wave interaction of gas transfer can be studied in greater detail. An augmented study design patterned after the Canadian SOLAS Station Papa mooring is recommended. A series of moorings is proposed covering the different coastal regimes including a near coast riverine dominated system that experiences large salinity gradients, strong buoyancy fluxes and low pCO_2 levels. The second regime would be in the active upwelling areas characterised by low SST and high pCO_2. The third regime would be just beyond the upwelling regime where there are frequently strong currents both parallel to the coast and occasionally perpendicular (coastal) jets. The sites of these phenomena are liable to change location over time as a result of changing winds, river flow and other forcing factors such that the moorings will not always be at the center of the regime of interest. The regions can often be distinguished from satellite SST, colour, and radar images. Optimal locations for deployment along the NEPTUNE network can be determined from perusing past satellite data. The advantage of the longer duration studies is that the particular regime can be studied in detail despite the spatial variability.

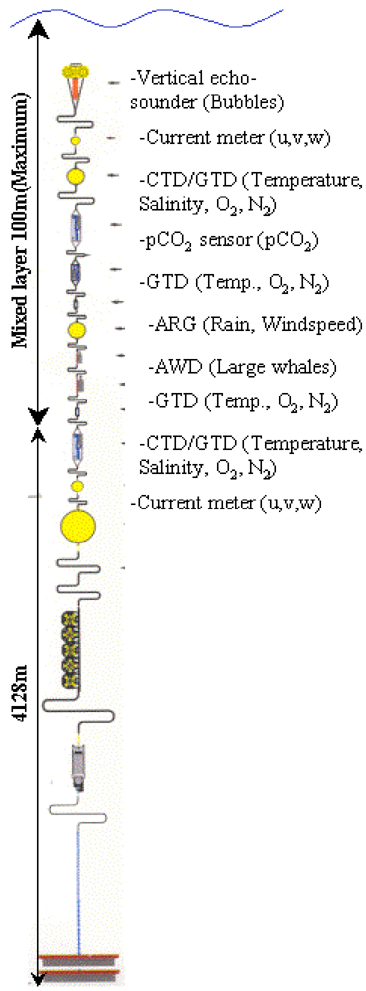

Figure 2: A Canadian-SOLAS mooring. The mooring is instrumented with both physical and biogeochemical sensors within the mixed layer to allow for studies of air-sea gas exchange processes for long periods in a region with high wind and sea states. Results are being interpreted in conjunction with a coupled atmosphere-ocean boundary layer model. Schematic courtesy of Svein Vagle.

Of particular interest in the coastal work along the western boundaries of continents is the effect of near shore air-sea fluxes on modifying air masses that flow across the continent. Some of the most robust estimates of sources and sinks of long-lived climate relevant gases such as CO_2 are obtained from inverse models. In these models the inferred magnitude of sources and sinks are directly related to the concentration of the particular compound of interest. Current inverse models use large regions and do not account for strong fluxes in smaller areas that can influence the adjacent boxes. For the North American continent the concentration in the air masses entering over the west coastal region must be measured in a sustained fashion. Thus aside from the very precise air measurements necessary for the direct flux measurement, the concentration must be measured at great accuracy as well.

The Canadian SOLAS mooring (Figure 2) is designed with a series of temperature, current, and gas tension sensors strung along a subsurface mooring line. For air-sea interaction studies, a surface manifestation is essential to capture the environmental forcing and to measure the gas, heat, water vapour, and momentum fluxes. Although deploying moorings in energetic environments is challenging, improved mooring and cable designs (e.g., the Tsunami early warning moorings) have led to successful deployments in a variety of stormy environments.

d. Monsoon Asia study

The Monsoonal Regime study will focus on air-sea gas fluxes in an area with extreme variation in forcing due to the monsoonal effects of changes in the wind direction, wind speed, and rainfall.

Significant air-sea interaction and biogeochemical information has been obtained in the Arabian Sea over the past decades. In particular the International JGOFS study and associated work on an air-sea interaction buoy funded by ONR has provided important seasonal information. However, no dedicated air-sea gas transfer process studies were performed.

The focus of this gas exchange study will be on the contrasting fluxes due to monsoonal forcing. To capture the effects of rain on air-sea gas transfer and boundary layer stability, the project area would be situated off the Indian Coast. Direct flux instrumentation will have to be adapted to perform measurements under rain conditions. Little is known about the stabilisation of the water column due to formation of a fresh water lens and its effect on air-sea gas fluxes. Water-side stability can be determined with profiling floats.

Aside from surface pCO_2 measurements with levels exceeding 1000 µatm, elevated CH_4 and N_2O levels are encountered near the coast during the monsoonal period. Although the three gases have some similar characteristics, they also have important differences that can be used to elucidate gas transfer mechanisms. CO_2, CH_4, and N_2O have similar Schmidt numbers such that their gas transfer velocity over a smooth and wavy surface should be the same. CH_4 has a much lower solubility than CO_2, such that in comparison with CO_2 transfer velocities, it can be used to separate gas transfer enhancement into turbulence and bubbles components. N_2O has similar physical properties as CO_2, but N_2O is not buffered in the water column nor will it experience any possible chemical enhancement effects due to hydration like CO_2 does. Therefore studying N_2O and CO_2 gas transfer simultaneously will offer insights into possible chemical influence on air-sea CO_2 exchange.

e. Tropical Pacific study

The Equatorial Pacific is one of the best-studied regimes because of its importance in the global heat, water and carbon budgets along with the large interannual variability caused by the ENSO cycle. From a SOLAS perspective the regime has many unique attributes that warrant quantification. The large fluxes of heat and several CRCs facilitate the measurements by improving the signal to noise for measurement of trace gases. The large heat fluxes and surface currents combined with low to intermediate winds make this an ideal environment to study parameters other than wind that control gas fluxes.

The process study in the Equatorial Pacific will build on several studies of sulphur, carbon, halocarbon and productivity limitation that have been performed in the last decade. The objectives will include:

- Quantifying DMS and SO_2 fluxes and gas transfer velocities using micro-meteorological techniques
- Investigating the response of the ecosystem to increased iron availability with focus on DMS, and halocarbon production
- Discerning the environmental forcing that affects exchange of CRCs
- Quantifying the effects of near surface chemical and physical gradients on the fluxes
- Validation, extension and tuning of air-sea exchange models such as the TOGA COARE model
- Modelling fluxes of CRCs for the Equatorial Pacific regime using the results from the SOLAS process study, previous process studies, and *in situ* and remote synoptic observations

The recommended study will be truly integrative over different CRCs and time scales through multi-platform execution, which take advantage of the large infrastructure available through the TAO/TRITON project. The area and scale will call for an international endeavour with the scope of the COARE study. The study would include an iron fertilisation component that perturbs the ecosystem in a systematic fashion and follows the response over its full perturbation back to background state. This has been lacking in the previous iron perturbation studies performed in the Equatorial Pacific and other regions. During the study, comprehensive micrometeorological measurements of heat, water vapour, momentum, CO_2, and halocarbons would be performed as well as detailed near surface profiles of temperature, salinity, and pH (and other parameters that can be measured at sampling frequency of about once per second). These studies would be done in conjunction with SOLAS Focus 1 Implementation Plan, which would focus on the biogeochemical response in the water column.

f. Polar regions

OASIS will lead this regional activity coincident with the international polar year. The polar region study is aimed at filling major gaps in our knowledge of the physical and chemical variables involved with Polar Ocean surface ozone and mercury depletion and radiatively-active trace-gas budgets. For the Arctic Ocean, we will collaborate with the International Arctic Buoy Project, the North Pole Environmental Observatory, and the proposed Arctic Ocean Observing System. Coordination is also envisaged with the satellite remote sensing community involved with measurements of halogen oxides, sea surface characteristics, and other chemical and physical parameters. SOLAS will encourage the setup of coordinated ice camps, icebreaker and aircraft studies of OASIS chemical exchange, for species to include DMS, CO_2, VOCs, O_3, $NO_{x/y}$, Hg, RGM, and particulate phase Hg, POPs (persistent organic pollutants), halogen oxides and molecular halogens, OVOCs, organo-halogen compounds, snow-phase ions; snow, ice and ocean bacteria and micro-algae; aerosols. The impact on, and by, the physical state of the local environment will be a key topic of these studies, as will cloud optical properties, and meteorological parameters.

g. Stratocumulus regimes

VOCALS Biogeochemical/Physical Observation/Model Study addresses interactions between the South American continent and the Southeast Pacific (SEP) Ocean which are extremely important for both the regional and global climate system. The Andes Cordillera form a sharp barrier to zonal flow, resulting in strong winds parallel to the coasts of Chile and Peru. This drives intense oceanic upwelling along these coasts, bringing cold, deep, nutrient/biota-rich waters to the surface. The resulting cold SST (in combination with warm, dry air aloft) is ideal for the formation of marine stratocumulus clouds, and supports the largest, most persistent, and least observed subtropical stratocumulus deck in the world (Klein and Hartmann 1993). The radiative properties of these clouds are strongly influenced by air-sea gas exchange.

VOCALS (the CLIVAR-VAMOS Ocean-Cloud-Atmosphere-Land Study) is an international programme in which modeling, extended-time observations, and intensive field observations in the SEP are being coordinated to address these issues over the period 2003-2010. A VOCALS "radiator fin" campaign will be conducted during a four week period in October/November 2007. It was named in recognition of a proposed cruise track and the characterisation of the subtropical subsidence regions as the "radiator fins" of the tropics, in which the free troposphere is very dry and infrared radiation efficiently cools the earth system. CLIVAR has formally requested collaboration with SOLAS in VOCALS.

Observations in the SEP made during the EPIC 2001 field campaign (Bretherton et al. 2004) suggest a direct link between drizzle and cloudiness in MBL clouds that is manifest through regions of broken cloud, called "open cellular convection", embedded within otherwise overcast stratocumulus (Figure 3). These regions have been termed POCs, or "pockets of open cells." Measurements suggest that POCs tend to be associated with low aerosol concentration (Petters et al. 2004), and intense drizzle production. This link between drizzle production and cloudiness is central to the hypothesis of Albrecht (1989), namely that increases in anthropogenic aerosol may lead to a reduction in precipitation and a corresponding increase in global cloud cover and thickness. We need to conduct detailed observational studies of POCs, with co-located aircraft, in-situ measurements, and ground/shipborne remote sensing, to determine whether POCs do indeed evince a fundamental mechanism whereby aerosols influence MBL cloudiness.

SOLAS interests include the role of biogenic gases (DMS) in forming the aerosols that control the marine cloud properties, the impact of light-level changes on biological activity and exchange coefficients, and studies of the many physical factors that control air-sea exchange. We need to quantify the links between the ocean chemistry and biology that modulate DMS (and other trace gas) production using integrated biological studies, DMS flux and exchange velocity measurements, and characterisation of natural and anthropogenic aerosols in the SEP MBL. SOLAS will assist CLIVAR in assembling an aerosol measurement team to study the chemical and physical properties of the aerosols inside and outside of POCs. The very low aerosol concentrations expected in POCs will no doubt lead to nucleation of new particles, so this team will need to measure ultrafine CN and DMA size distributions down to a few nm. One of the central questions is how rapidly these newly nucleated particles can grow to the size where they can act as CCN, thus allowing the POCs to again fill with clouds. To address that we will measure the supply of the precursor gas DMS and the nanoparticle growth rate in POCs. If the aerosol growth is faster than DMS fluxes can explain, then other sources, such as organics, must be important.

VOCALS offers an excellent opportunity to study the factors that control gas exchange velocities. To achieve this we must measure non-wind factors such as wave spectra (sea state), surfactant films, whitecap coverage, and other controlling factors alongside the gas exchange ve locities.

2. Sustained observations on research vessels (RVs) and buoys

Present parameterisations of SOLAS-relevant air-sea exchange (trace gases and aerosols) as implemented in global models have been principally derived from a combination of laboratory work and a few very limited field studies. This is in stark contrast to more conventional heat, moisture and momentum fluxes where tens of thousands of hours of direct observations are available to refine the parameterisations (e.g., Fairall et al., 2003). We anticipate that the intensive process studies planned for SOLAS (as outlined in this document) will lead to rapid improvement in parameterisations. However, these process studies are likely to be limited in time and spatial extent. A more extensive verification strategy is required because the parameterisations will eventually be applied globally and in a wide range of surface conditions.

In the realm of physical air-sea fluxes, Ocean Reference Stations (ORS) are the primary *in situ* measurement systems for long-term observation of surface forcing in a wide range of conditions. The ORS are large surface buoys designed to obtain a complete set of high quality meteorological variables for estimation of air-sea fluxes using bulk formulas. Arrays of buoys are used to obtain relevant spatial information. The buoy data are critical in evaluating fluxes in various climate regimes (McPhaden et al., 1998), intercomparisons for operational forecast models (Josey et al., 2002), and efforts to retrieve fluxes from satellite information (Curry et al., 2004).

Since networks of ORS buoys are not expected to meet all air-sea flux scientific needs (Taylor and Yelland, 2001; Cronin et al., 2002), plans are being developed by the High Resolution Marine Meteorology (HRMM) community to supplement them with high-quality measurements from research vessels (R/Vs; Smith, 2004). The vessels of the R/V fleet ply the world's oceans for 200-300 days a year, often over routes not visited by commercial Volunteer Observing Ships, and represent an untapped platform resource for obtaining geographically and seasonally diverse data. The HRMM initiative seeks to improve the accuracy of the meteorological and oceanographic measurements made on these ships, by establishing calibration and operational protocols, and providing educational information to those responsible for installing and maintaining the equipment. The evolving strategy for ORS and R/V AWS conventional flux observations suggests an obvious parallel pathway for SOLAS. There are two quite different goals: (1) comparison of model/satellite estimates of fluxes to those derived from local high quality *in situ* estimates and (2) actual verification of parameterisations using direct measurements. The first goal assumes that state-of-the-art parameterisations are accurate and evaluates the ability of GCMs or satellite methods to accurately reproduce the inputs of the parameterisations, to implement the parameterisations (which are often simplified compared to the detailed, research grade parameterisations), or both. The second goal is a straightforward evaluation of the state-of-the-art parameterisations in a broad context.

The first goal can be implemented by supplementing the existing buoy or VOS (SAMOS - The Shipboard Automated Meteorological Oceanographic System) observations with relevant bulk chemical or particle concentration measurements (those appropriate for bulk flux calculations such as ΔpCO_2). In some cases, additional measurements of parameterisation-critical variables that may not be routinely measured (e.g., wave height, whitecap coverage, IR SST, surfactants) would add greatly to this enterprise. However, because this is principally a verification effort, auxiliary measurements would be much less comprehensive than the intensive field studies. The second goal will require instrumenting buoys or R/Vs for direct covariance flux (or some other direct flux technology). For most SOLAS variables, this will require considerable sensor/system development to fit within the SAMOS concept. Implementation on ORS will be more challenging than on R/Vs because they have additional constraints of low power and unattended operations.

Finally, SOLAS can make use of existing near-shore platforms for this type of scientific activity. The 600m long pier of the FRF in Duck, NC, USA would also be an excellent facility for sustained observations. Oceanographic and meteorological support measurements are now available and it is feasible at this location to perform long term unattended flux measurements. In addition, the Chesapeake Bay and Woods Hole Air-Sea Interaction Tower are two other platforms to consider.

3. High wind process studies

The gas and particle fluxes that are of central importance in SOLAS have stronger wind speed dependencies (on the order of the third power of wind speed for breaking wave processes) than conventional energy and momentum fluxes (first or second power). Thus, measurements of gas and aerosol fluxes at high wind speeds will be of increased significance for SOLAS. Field observations under these conditions present increasing levels of operational and technological difficulties as wind speeds increase. Statistically significant sampling of the fluxes at high wind conditions offers additional problems because the experimental scatter tends to be larger and high wind events are generally episodic with relatively long time periods in between events. An additional complication is that the storms that are generally associated with high wind speeds are often accompanied by precipitation and higher wave states, making the high frequency atmospheric turbulence measurements required for the micrometeorological flux methods even more difficult.

There are also issues associated with the choice of measurement platform when conducting field observations at high wind speeds. Flux measurements from ships are complicated by wave-induced motions and spray generated from the wake. Fixed platforms are easier to work from because platform motion and spray generated by the wake are not issues, and an undisturbed flow is easier to achieve. Ocean towers have disadvantages in terms of the sampling statistics of the high wind events (i.e., mobile platforms can move to high wind areas). For very strong wind conditions, aircraft are ideal for getting to the experimental location but are not appropriate for use with *in situ* sensors to make concurrent surface-layer measurements. There are also safety concerns in operating aircraft close to the ocean surface during conditions of high winds and intermittent areas of low visibility. There is no single perfect option, so a combination of fixed platform, ship, and aircraft observations should be used during SOLAS to obtain the required flux measurements at high wind speeds.

Obviously, high wind processes must be studied in locations with reasonable expectations of high winds: mid-latitude storm tracks, tropical cyclone areas, regions with orographic/thermobaric winds, and monsoon jet regions. For gas exchange, the study region must be an area with large air-sea gas concentration disequilibrium if the gas fluxes are to be made using micrometeorological methods. Aerosol flux measurements are simplified if the study region is not heavily impacted by nearby terrestrial aerosol sources.

Ship-based atmospheric observations must be hardened to yield usable data in the presence of wind-driven rain. Fast-response gas sensors using closed-path technologies are preferred. Sample tube corrections for these sensors will be needed, so some open-path sensors should also be used during rain-free periods. Contamination of sonic-anemometer turbulence signals must be removed during processing or a different fast wind sensor technology (e.g., modernised K-gill propellers) must be used. Present day ship-motion correction methods are inadequate for gas flux applications for winds exceeding 15 m/s for typical research vessels. Improvements in turbulence sensors, motion sensing technology, and processing algorithms are required. Most of these points also apply to buoy-based observations.

Observations at heights on the order of 100 m can be used to infer true interfacial fluxes, and aircraft could be very useful for the gas exchange, investigation. However, such observations will require innovative measurements of other environmental variables for the purpose of parameterisation development. The recent advancement in whitecap imaging methods is one good example. Aircraft might be useful for very strong wind sampling of sea spray concentrations, but some form of a nadir active remote sensing system (Doppler radar, lidar, etc.) will probably be required to obtain useful concentration profiles. Modern high frequency pulsed Doppler radars (95 GHz) have marginal vertical resolution (30 m) for the sea spray problem. To date, FMCW Doppler techniques are unworkable in the presence of scattering from the sea surface. Thus, very high frequency radar in the 100s of GHz will need to be developed.

Oceanic observation systems on the ocean side of the wave boundary layer are relatively more developed than meteorological sensors for high wind conditions. However, measurement systems based on umbilical-powered small catamarans, tethered to a nearby research vessel have become unworkable above 10 m/s with today's technology. Conventional discus and spar buoys have been used up to 15 m/s, but some considerations for much stronger winds and larger waves must be evaluated.

Activity 2.1 - Exchange across the Air-Sea Interface

Understanding physical and biogeochemical processes near the air-sea interface is critical for predicting the air-sea exchange of gases and aerosol particles and for determining how these processes will affect and be affected by global change.

Introduction

The air-sea exchange of gases and aerosol particles is controlled by many interdependent and interacting biogeochemical, hydrodynamic and aerodynamic processes (Figure 4). Activity 2.1 aims to quantify the functionality and impact of these processes on the magnitude and variability of gas and particle transfer rates. Of particular interest is the development of transfer parameterisations in terms of measurable environmental properties and a characterisation of the accuracies of these models.

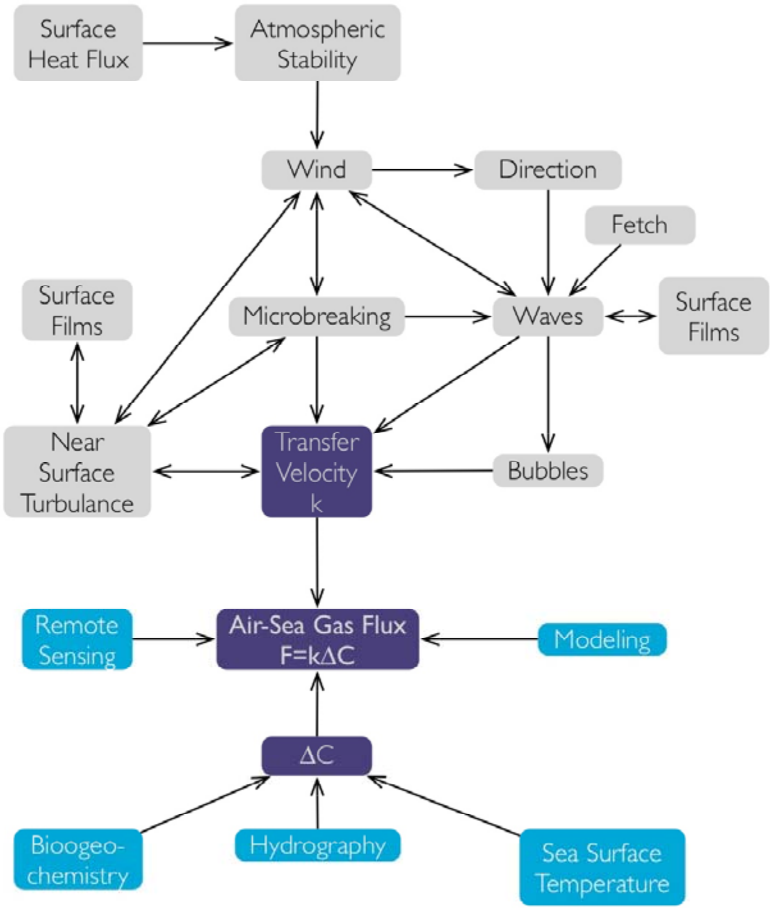

Figure 4: Key air-sea gas transfer properties and processes to be measured in order to extrapolate to global and regional scales. Also shown are the tools of remote sensing and modelling that are crucial to scaling up air-sea fluxes from small temporal and spatial scales.

Such a parameterisation would quantify the dependence of the gas and aerosol fluxes on near-surface atmospheric and oceanic turbulence, atmosphere/ocean density gradients, air/water temperatures (and gradients in each phase), bulk/skin seawater temperature difference, salinity, sea-ice cover, sea state, surfactant concentrations, and bubble concentrations (and their sub-surface dynamics) at wind speeds up to 25 m/s. It is essential to understand the sensitivity of climate and ecological models to the air-sea gas and aerosol fluxes derived from these parameterisations. Modelling studies will be conducted in concert with the experimental efforts so that the model results can be used for determining the focus of the evolving experimental activities.

2.1.1 - Predict gas and aerosol exchange rates for commonly observed environmental conditions. This requires determination and quantification of the processes that control mass exchange rates. The errors of these exchange rate estimates should be comparable to those for fluxes of momentum, heat and moisture

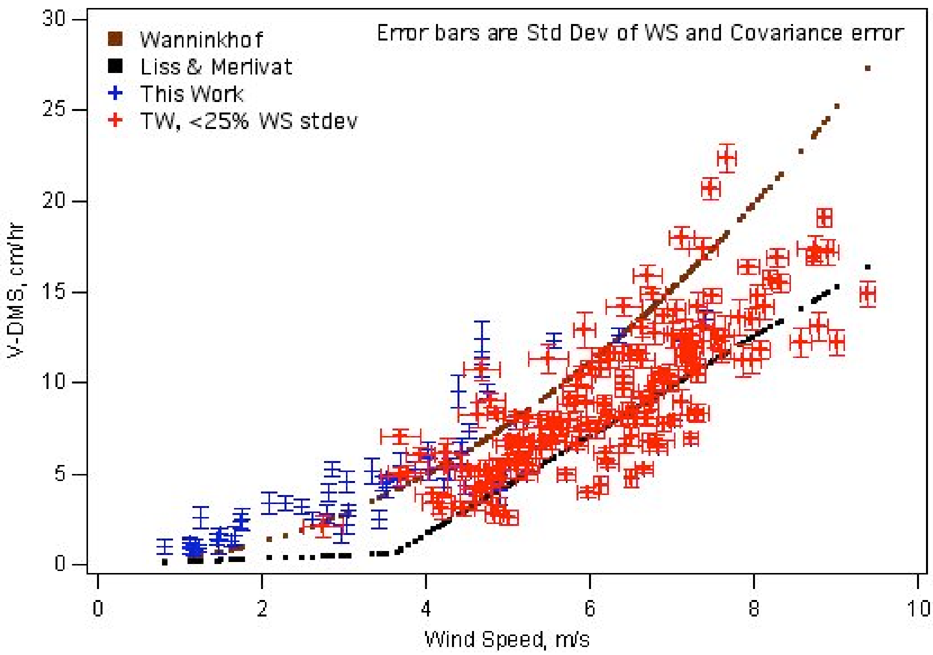

Figure 5: Gas transfer velocity versus wind speed measured using direct covariance DMS measurements over the ocean (Huebert et al. 2004).

The first determinations of gas exchange rates relied on geochemical mass balance techniques on a range of spatial-temporal scales. These range from purposeful gaseous tracer experiments (e.g., Nightingale et al., 2000a; Wanninkhof et al., 2004) and radon disequilibrium determinations (e.g., Smethie et al., 1985) to global, ocean-basin scale and regional radiocarbon uptake estimates (e.g., Broecker et al., 1986). Developing improved parameterisations requires measuring the air-sea fluxes over the same time scales as the variability in the environmental forcing functions, on the order of tens of minutes. Recent advancements in micrometeorological methods of gas flux measurement will be invaluable to Activity 2.1. Results from the ASGAMAGE (Jacobs et al., 1999), GasEx-98 (McGillis et al., 2001) and GasEx-2001 (McGillis et al., 2004) studies have demonstrated that it is now practical to measure the air-sea flux of CO_2 with a time resolution on the order of 30 minutes by eddy covariance. Direct eddy correlation measurements of the air-sea flux of dimethylsuphide (DMS) are also now possible from airplanes, towers and ships (Huebert et al. 2004, Figure 5). These micrometeorological gas flux methods are an integral component of the Activity 2.1 field experiments.

Achieving Activity 2.1.1 also requires the identification and quantification of the processes that control the fluxes. Given the large number of these variables and the fact that many of them have multiple controlling environmental factors (e.g., near-surface oceanic turbulence is generated by wind stress, wave breaking, convective overturning, current shear, and rain), it is unlikely that comprehensive parameterisations can be developed using data from a single field experiment. Understanding some processes will require carefully designed laboratory experiments. Thus, field and laboratory experiments must be designed to complement or validate each other. Implementation of Activity 2.1 requires a series of coordinated activities ranging from microprocess studies of single factors to large-scale field experiments with coordinated sampling from ships and aircraft.

2.1.1.a Conduct laboratory and field measurements on the impact of surfactants

Naturally occurring surface-active organic material apparently modulates gas exchange, surface waves and turbulence, so characterisation of the surface excess of dissolved organic matter should be a routine measurement. Although currently available methods for collecting ocean surface microlayer samples are successful (Frew et al., 2002), they are costly and cumbersome. We must develop more easily deployed methods for collecting and analysing surface microlayer samples as well as techniques for the *in situ* determination of organic surface concentrations. Assessing the effects of surfactants is complicated by the fact that it is not clear how to determine which environmental variables should be measured to parameterise the effect.

Understanding the effects of surfactants on gas transfer, aerosol generation, and aerosol chemical composition requires measuring gas fluxes, aerosol fluxes, and droplet compositions for known concentrations of naturally occurring surface active material. Although laboratory experiments of this type have been carried out in the past, synthesising these data into a usable parameterisation of the effect of surfactants on fluxes has been problematic. Laboratory process studies should have a strong emphasis on relating the results to field conditions. They should be designed to provide guidance for the field measurements of surfactant impacts on ambient fluxes. The laboratory measurements must use surfactant concentrations typical of those found in the ocean and ensure that the surface rheology of the laboratory surfactants are similar to those of naturally occurring surfactants.

2.1.1.b Improve aerosol eddy covariance methods and study bubble-mediated aerosol production in the laboratory

Direct eddy correlation measurement of aerosol fluxes has been demonstrated by Nilsson et al. (2001). This includes size resolved and chemically specified fluxes, which provides extremely useful data of high priority. Other promising new techniques are aircraft measurements to establish the role of fetch in aerosol fluxes (Reid et al., 2001) and application of transport models (Vignati et al., 2001) and laboratory experiments of particle production from artificial bubble bursting (Mårtensson et al., 2003). Key factors for parameterisation of the sea-spray include wind speed and wind speed variability, thermal stability, wave steepness, water temperature, bubble concentrations and spectral shape, and surface tension. Simultaneous measurement of bubble and sea-spray size distributions, covering a variety of atmospheric and oceanographic conditions, in combination with techniques to derive the sea-spray, provide a promising approach to better parameterise the production of sea-salt aerosol.

Since aerosol particles generated by breaking waves depend on both air-phase and water-phase processes, parameterising the air-sea aerosol flux is a problem of even greater complexity than gas flux. Understanding aerosol production requires understanding of how the bubbles that are created by breaking waves form spray droplets. Then, the air-phase motions that are responsible for advecting the spray away from the water surface must be quantified. Current methods for measuring aerosol production from single bubbles and bubble plumes cannot be easily adapted to quantify oceanic droplet generation mechanisms. Therefore, as is the case for the whitecap-mediated gas flux, detailed investigations of droplet production by breaking waves and bursting bubbles may be most effectively conducted in the laboratory. See 2.3.1 for droplet generation and aerosol flux characterisation methods.

2.1.1.c Develop an accurate method for estimating the whitecap-mediated gas flux and steady-state bubble-mediated gas supersaturation using field-measurable environmental parameters and eddy-correlation gas fluxes

The influence of wave breaking and bubbles on air-sea gas flux is best understood by simultaneous measurement of bubble populations and dynamics, subsurface turbulence, wave properties, and exchange rates of gases with different molecular diffusivities and solubilities. Whitecaps work to increase the gas flux through both turbulence and bubbles, and the relative importance of each mechanism is not clear at present. Because there has been a limited suite of gases whose air-sea fluxes can be measured with short timescales, the physiochemical hydrodynamics of whitecap-mediated gas exchange have been studied using laboratory experiments in wave basins, tanks, and wind-wave tunnels (Asher et al. 1995; 1996; 1997; De Leeuw et al., 1999; 2002). Further studies to develop an accurate method for estimating the whitecap-mediated gas flux and steady-state bubble-mediated gas supersaturation should employ eddy-correlation gas fluxes, detailed bubble spectra, and field-measurable environmental parameters. Laboratory studies on the effect of breaking waves on gas fluxes should be conducted in saltwater using wave basins, wind-wave tunnels, or bubble-plume tanks. Experiments should include a modelling component in which the data collected are used to synthesize a verified parameterisation of the bubble-mediated gas flux that includes the measured invasion/evasion flux asymmetry and steady-state supersaturation. Bubble microprocess studies must measure not only the gas fluxes, but also the size-segregated bubble populations and their subsurface dynamics from their initial creation to their dissolution or rise back to the surface.

2.1.1.d Develop methods for quantifying the impact of rainfall on gas and aerosol fluxes

The importance of reliable and appropriate precipitation observations in field campaigns needs to be emphasised. Rainfall contributes directly to the air-sea heat and momentum fluxes (Gosnell et al., 1995; Caldwell and Elliott, 1971), and affects the surface roughness and drag coefficient. It influences air-sea gas transfer in a number of ways: changing the temperature and salinity at the surface (with a consequent effect on solubility and reaction rates), physically disturbing surface films, changing the density stratification of the upper mixed layer, and suppressing turbulent mixing at the surface. During light winds and in the tropics, freshwater lenses can persist on the ocean surface for considerable periods.

Rain-rate knowledge is central to studies of wet deposition. However, rain measurements at sea are problematical on two counts: wind flow distortion by the ship can seriously impair rain-gauge performance, and the typical variability of rain storms means that the point measurement may not be representative of the average rainfall over the study region. The first defect can be mitigated by using multiple instruments that work on different principles and by carefully choosing locations. The second problem can be addressed with the aid of radar and satellite-derived spatial rainfall patterns. Improved algorithms for TRMM and other remote rain sensors are currently being developed. Broad-scale rainfall estimates recently have been obtained successfully as the residual of freshwater budget calculations from profiling CTD surveys (Feng et al., 1998; Godfrey et al., 1999). Regional rainfall estimates may also be obtained from atmospheric moisture budgets when the field programme includes an array of radiosonde stations (Johnson and Ciesielski, 2000). APL (Jeff Nystuen, Washington State University) has achieved some success in measuring rainfall over the ocean by detecting its acoustic signature with an instrument mounted at depth on a mooring.

2.1.1.e Develop and validate thermographic methods to infer gas fluxes

Infrared imagery is being used to study air-sea exchange of heat and gases (Jähne et al., 1989; Haußecker et al., 1995; Garbe et al., 2004; Schmipf et al., 2004). These thermographic methods may provide a means for measuring air-water gas transfer rates, air-water net heat fluxes, and bulk/skin water temperature differences with unprecedented spatial and temporal resolution. Recent results show some of the efficacy of using heat as a proxy tracer for gas transfer (Asher et al., 2004; Atmane et al., 2004; Zappa et al., 2004). These methods must be verified using field measurements of trace gas fluxes. This technology is a strong complement to other mass-balance and micrometeorological techniques.

2.1.1.f Conduct integrated, intensive process studies in which ships, towers, and aircraft are used to measure fluxes and controlling factors

Integrated intensive process studies can provide mechanistic information on one or more controlling factors for parameterising fluxes. These process studies will probably also have objectives from Focus 1 or 3 in which flux measurements are needed for understanding geochemical cycles. Comprehensive major process studies should include intercomparison of several different flux measurement techniques, including both geochemical and micrometeorological methods, so that inter- and intra-method variability can be assessed. Field efforts must include measurement of the forcing functions such as wind stress, wave properties (including breaking extent and subsurface bubble populations), surfactant concentrations, sea state, and near-surface turbulence to enable analysis of controlling factors. Whenever possible, these studies should include development of new methodology and techniques for direct flux measurements of additional species using micrometeorological techniques for gases and aerosols. Specific emphasis should be given to techniques providing information on particle chemical composition (De Leeuw et al., 2003; Nilsson et al., 2003).

These intensive campaigns should be conducted in several regions with characteristic oceanographic, biological and micrometeorological properties (see Proposed Fieldwork Campaigns and Detailed Recommendations for Focus 2 Experiments) with particular emphasis on understanding the dominant flux mechanisms at high wind speeds, the flux variability due to differences in surfactant levels, and how the fluxes can be most accurately expressed in terms of easily measured environmental parameters. Parameterising the aerosol flux will require special attention to the effect of breaking wave properties on bubble plume types and the resulting aerosol production. Aircraft measurements would be desirable to get spatial variability in aerosol and gas fluxes while ships are needed to measure controlling factors and air/sea concentration differences. Tower experiments will also have a role in these process studies, provided they can be suitably instrumented and are in relevant locations.

2.1.1.g Conduct time-series flux measurements

Long-term measurements should be made of air-sea fluxes and the relevant forcing functions at a number of sites that cover a wide range of biogeochemical-physical conditions (see 3. Sustained observations on research vessels and buoys, and 2.4.2). This will provide the data required to both validate and refine flux parameterisations developed as part of Activity 2.1 and to understand how seasonal cycles and periodic phenomena such as ENSO and climatic changes affect the air-sea fluxes and their parameterisations. These observations must be planned and carried out cooperatively with ongoing programmes responsible for long-term observations. Where possible, other SOLAS experiments can be conducted at these sites, to tie the process studies to time-series observations. As many flux forcing functions as possible should be measured concurrently with these long-term air-sea fluxes.

2.1.2 - Modelling gas and aerosol exchange rates on local, regional and global scales. Quantify the sensitivity of models used in SOLAS to a range of gas flux and primary marine aerosol radiative forcing scenarios spanning at least twice the range of uncertainties determined from known measurements

Parameterisations of air-sea transfer processes are used in various kinds of models such as meteorological, climate (GCM), and local and regional-scale chemical transport models. For instance, sea-spray is used in GCMs to estimate the top-of-atmosphere, global-annual radiative forcing due to sea salt. The interaction between the modelling communities (as users of the parameterisations from the SOLAS air-sea exchange studies) and the experimentalists will provide input on the required accuracy and limitations of the results that can be expected from SOLAS in the next decade. Achieving this goal will require testing the parameterisations of air-sea transfer processes that are used in various kinds of models such as meteorological, climate and local and regional-scale chemical transport models.

3-D models are at the top of the hierarchy of models applied in SOLAS. They include regional and global chemistry-climate models and chemistry-transport models coupled to ocean surface layer models. Coupled chemistry-climate models allow investigations of interactions and feedbacks between the dynamics, physics and chemistry of the atmosphere, while chemistry-transport models prescribe the meteorology and may represent the atmospheric chemical processes in more detail. Parameterisations for 3-D models will be optimised using both SOLAS observations and the results from process models.

In general, the accuracy of the model's representation is assessed by comparing modelled trace gas and particle concentrations and fluxes with SOLAS observations. Some SOLAS measurements may be organised in a Lagrangian way, to evaluate model processes and assess their uncertainties. Budget analyses provide insights into the contributions of dynamical, physical and chemical processes that affect marine chemical cycles. For example, comparison of the simulated budgets of the marine sulphur cycle for different conditions, (low vs. high wind velocity, cloud-free vs. cloudy sky, advection of clean vs. polluted air, etc.) may provide insights into the response of the chemical system to perturbations of the climate.

In summary, SOLAS supports modelling the interfacial processes for gas and aerosol transfer (e.g., aerosol deposition, spume droplet production, bubble bursting processes). The employment of inverse modelling to determine surface fluxes using assimilation of surface observations and satellite derived data (aerosols, gases, surface characteristics) in chemical transport models. This activity should be conducted in collaboration with CLIVAR, WGCM and WGNE, and the development of a parameterisation of k as a function of satellite-derived data such as wind stress and surface roughness (Glover et al., 2002).

Activity 2.2 – Processes in the Oceanic Boundary Layer

Understanding upper ocean boundary layer physical and biogeochemical processes that regulate changes in near surface concentrations and air-sea exchange of gases and materials is critical for determining how such exchanges and processes will affect and be affected by global change.

Introduction

The upper ocean is intermittently mixed and re-stratified through the combined effects of wind, surface buoyancy fluxes and penetrating radiation. Mixing reduces vertical gradients close to the air-sea interface. In the oceanic boundary layer, upper ocean circulations combine with *in situ* biogeochemical transformations to control exchanges with the atmosphere. Wind mixing, wave action and current shear represent constantly changing inputs of kinetic energy. This physical forcing operates on variable time and length scales. Biogeochemical transformations have yet another set of distinctive time scales that are not generally matched to the physical scales. The interaction of these different and interdependent processes produce spatially and temporally varying properties within the ocean surface layer.

The ocean-atmosphere interface, which controls exchange, is influenced both by processes occurring on short time scales and by longer-term controls of oceanic boundary layer properties. Transport and transformation of properties within the oceanic boundary layer influence air-sea exchange on time scales from diurnal to seasonal. Moreover, oceanic boundary layer physical processes exert fundamental controls on the character and overall rates of biogeochemical processing within the upper layers, which in turn play a role in modifying the characteristics of the interface itself. The boundary layer is often defined in physical (circulation and mixing layer) or biological (euphotic zone) terms. The coupling of this boundary layer to deeper layers is important, particularly with regard to the return of remineralised nutrients and the subsequent regulation of biological productivity and trace gas production. Modification of that mixing may be part of one climate feedback mechanism.

The dynamics of the ocean mixed layer exert an important control on gas transport. Langmuir circulation, bubble formation and transport, and surface layer stratification are among the controlling factors that are not adequately understood.

2.2.1 - Accurately measure gradients of gases and their variability within the upper ocean

We need to develop a dynamic coupled biogeochemical - upper-ocean mixed layer model with time resolution of milliseconds and vertical resolution near the surface of the order of 100 microns, capable of predicting near-surface concentrations and gradients of salinity, nutrients, O_2, CO_2, DMS, NH_3, DIC, DOC and Fe species, with contemporary process formulations.

The major unresolved issue relating to this goal of Activity 2.2 is how physical transport and biogeochemical processes in the ocean surface boundary layer regulate near-surface concentrations of biogeochemically important materials (gases and nutrients). Understanding this regulation is central to predicting gas exchange across the air-sea interface, the goal of Activity 2.1.

This work will be addressed through coordinated observational and modelling studies. The observational approach will include time series and spatial gradient measurements of biogeochemically important elements and compounds within the context of a process study resolving the relevant upper ocean physics. The modelling approach will develop and test coupled physical-biogeochemical models constrained through initial and boundary conditions and validated by the observations. The strategy will be to obtain data sets suitable for model initialisation and validation in a variety of biogeochemical and physical regimes where a particular process dominates variability on a certain time scale. Models would then be exercised by attempting to reproduce salient features of the observations in these regimes.

The importance of small-scale processes in driving upper ocean biogeochemical activity requires the development of very high resolution numerical models and a concomitant observational programme to design, parameterise and test those models. Critical tests of model fidelity include not only the simulation of the bulk properties and long-term evolution of the surface boundary layer, but also small-scale variability and statistics of physical properties and biogeochemically important species that will be taken up, transformed and/or released in the surface ocean on short time scales by chemical (e.g., photochemical transformation) and biological (e.g., phytoplankton, zooplankton and bacteria) processes.

Modelling activities at different levels of complexity will be needed. On the local scale, one-dimensional (1-D) vertical column models should reproduce physical processes on diurnal to seasonal time scales and include coupling to relevant biogeochemical processes. On the global scale, models should be able to reproduce the major features of present ocean circulation and water mass properties, as well as capture the alteration in upper ocean circulation on interannual to decadal time scales due to climate change. Models must describe several sources of nutrients, physical processes, nitrogen fixation by cyanobacteria, and the photochemical inhibition of biological degradation. Research will be required to determine the best parameterisations for global scale models of processes explicit in local scale models.

Oceanic measurements with high temporal and vertical resolution will be critical to the observational component. Processes of interest will occur on time scales from minutes to seasons and on vertical scales from millimetres to a few hundred meters. A unique emphasis under Goal 1 will be to consider the coupling of the oceanic mixed layer with the underlying pycnocline. A classic example is the entrainment of pycnocline water during mixed layer deepening driven by surface forcing. However, a complete study must also include physical coupling from below (e.g., through turbulent mixing and entrainment modulated by internal waves in the pycnocline) and biogeochemical coupling that is independent of mixing and entrainment (e.g., through particle fluxes or grazing by vertically migrating zooplankton).

Examples of focus areas for process study experiments, and sites where they might be conducted are listed below. The examples are meant to be indicative rather than prescriptive, and are not mutually exclusive (e.g., a eutrophic vs. oligotrophic study could be done at two sites that both exhibit strong seasonal cycling of vertical stratification and have episodic eddy activity).

2.2.1.a Time series experiments at high-latitude sites with strong seasonal variability in vertical stratification and mixed layer depth and a distinct biological response

High latitude air-sea interaction time series are required to capture this energetic state. Observing systems such as Ocean Weather Station (OWS) Papa in the Northeast Pacific or Ocean Station Mike (Norwegian Sea), have long histories of physical observations and may serve as canonical test cases for 1-D mixed layer models (Martin, 1985; Large et al., 1994) and biogeochemical modelling work (Denman and Pena, 2001). The subarctic North Atlantic also exhibits strong seasonal cycling, and is distinguished from the Northeast Pacific by a more dramatic spring phytoplankton bloom. There is a history of coupled physical/biological experiments in this region (e.g., the Marine Light – Mixed Layers experiment (Marra, 1995) and the JGOFS North Atlantic Bloom experiment (Ducklow and Harris, 1993)).

2.2.1.b Long-term studies of upwelling

The equatorial upwelling regime in the Eastern Equatorial Pacific is of interest in the study of CO_2 flux and climate (e.g., JGOFS EqPac, Murray et al. 1995, Feely et al., 2002). This activity is cross-linked with Focus 3. A long-term study in this region could be accomplished by deployment of biogeochemical sensors on ships and moorings in coordination with ongoing operations of the TAO/TRITON array. Such an approach was taken in a MBARI pilot study (www.mbari.org/bog/Projects/EQPAC/) and currently being expanded in a collaborative effort between MBARI and PMEL. In a coastal upwelling regime (e.g., the Peru or California coast), an experiment spanning several upwelling events could be conducted. A traditional moored array, supplemented by observations from ships and autonomous vehicles could be used. A more innovative approach would establish links with cabled observatories such as NEPTUNE, which may extend southward along the west coast of the United States from its origins on the Juan de Fuca plate.

2.2.1.c Winter experiments at a site characterised by deep mixed layers driven by strong surface forcing

Winter mixed layers several hundred meters deep can be found in the subarctic North Atlantic, affording possible synergy with a seasonal cycling site. OWS Bravo in the Labrador Sea is of particular interest to climate studies because it monitors variability of various North Atlantic Deep Water properties (Lazier, 1980; 1988) and is a principal source of the carbon flux into Atlantic deep and intermediate waters. Research Vessels and ruggedized moorings are required for this component of SOLAS. A synergy with CLIVAR would be highly beneficial.

2.2.1.d Measurement of property fluxes caused by eddies and fronts

Mesoscale features are associated with space/time variability in physical, biological and chemical processes that may drive significant property fluxes and affect local, regional, and basin-scale biogeochemical budgets. These features represent substantial observational and modelling challenges. The ubiquity of mesoscale processes means that most time-series sites will eventually be affected by them (e.g., Letelier et al., 2000; Sweeney, 2003). Specific sites that might be chosen with these processes in mind would include the Gulf Stream and Kuroshio extension regions, the northwest Arabian Sea, the Iceland-Faroes front, and the Atlantic subtropical convergence zone.

2.2.1.e Comparison of eutrophic and oligotrophic ecosystems

Observations from a high-latitude eutrophic site, an upwelling site or an eddy/frontal area should be compared with those from a canonical oligotrophic site near the center of a sub-tropical gyre. It is notable that some biogeochemical observations are already being made at oligotrophic sites through programmes such as the Bermuda Atlantic Time-series Study (BATS) and the Hawaiian Ocean Time-series (HOT) (Karl and Lukas, 1996). There are also a variety of coastal sites where contrasting oligotrophic and eutrophic conditions can be found in relatively close geographic proximity. The role of surface films may be a crucial difference between these systems. Diatom and phyocystis blooms may have a significant impact on local surfactant levels. Surface film manipulation using an SF_6 labelled diatom culture is one possible strategy.

2.2.1.f Development of models to guide experimental design to include coupled physical and biogeochemical processes

Model requirements for SOLAS depend on the specific application. However, a priority for SOLAS includes the incorporation of biogeochemistry into physical models. At the local scale envisioned for many of the SOLAS observational studies, models representing detailed mixing and transport processes will need to be developed to guide and integrate the detailed measurements. Such models exist for physical measurements, but current versions have little or no representation of biogeochemical processes. Some Atmospheric Single Column Models (ASCM) include extensive chemistry relevant to SOLAS. There is a requirement to take the knowledge and understanding gained from combining models and observations of processes at the local scale to develop improved parameterisations of those processes for inclusion in models at the regional to global scale. This scaling-up process must be iterative and interdisciplinary.

2.2.1.g Time-series observations of inert species to isolate physical processes

It is presently difficult to identify the effect of individual physical processes on gas exchange even for a chemically and biologically non-reactive gas, where the mixed layer concentration is determined only by advection, entrainment from below, and exchange across the air-sea interface. Due to the complexity of the coupling of physical and biogeochemical processes, initial studies may be in regions where vertical processes dominate. Substantial progress can be made in such regions using traditional moored time-series techniques along with judicious use of gas concentration and biogeochemical measurements.

2.2.1.h Development and deployment of biogeochemical sensors on a variety of platforms

While ships provide important capacity for the use of large, complex instruments and techniques that may require large-volume material sampling or human intervention, one key to observationally coupling physical and biogeochemical processes will be to incorporate established and newly developed biogeochemical sensors capable of deployment on moorings, drifters, towed devices and autonomous vehicles. The vertical resolution and extent of the gas concentration observations should approach that of the physical properties.

An increasing number of bio-optical and chemical parameters can be measured from autonomous platforms. Optical approaches are arguably the most mature, and include beam attenuation, absorption, scattering, chlorophyll fluorescence and Photosynthetically Available Radiation (PAR). Multiple-wavelength sensors allow phytoplankton to be distinguished from chromophoric dissolved organic material (CDOM) and may allow identification of phytoplankton community groups. More sophisticated sensors such as flow cytometers have been packaged for autonomous deployment. *In situ* chemical analysis for constituents including nitrate, nitrite, phosphate, silicate, and iron, can be done using underway systems on ships and from self-contained systems suitable for deployment on moorings and vehicles. A useful review of emerging techniques applicable to multi-disciplinary studies as envisioned for SOLAS Activity 2.2 is given by Dickey (2003).

2.2.1.i Use of chemical tracers to study physical transport and mixing processes

Chemical tracers can be used to improve understanding of physical transport and biogeochemistry in the upper ocean mixed layer and in particular the coupling of this mixed layer with the underlying pycnocline. The concentrations of the noble gases within the oceanic mixed layer are controlled only by physical processes such as diffusive and bubble-mediated gas exchange, temperature changes and to a lesser extent atmospheric pressure deviations (Hamme and Emerson, 2002). The concurrent measurement of dissolved oxygen together with noble gas concentrations therefore allows the determination of the physical processes influencing oxygen saturation, hence the gross production/respiration of oxygen can be calculated and this information can be used in turn to estimate export production (e.g., Emerson et al., 1995). Time series measurements of noble gases also represent useful tools to investigate air-sea gas exchange because of their wide range of solubilities and diffusivities. Chemical tracers can also provide important information on rates of ocean mixing and ventilation.

Another important application of tracers that could be employed in SOLAS is the use of tritium, chlorofluorocarbons (CFCs) and bomb-produced radiocarbon to model water mass circulation over decadal timescales. These chemical tracers can also be used for model evaluation and validation although a significant uncertainty in the use of CFCs for this purpose is their air to sea gas transfer rates (England and Maier Reimer, 2001). It is envisaged that time series measurements of the noble gases, their isotopes, and CFCs will form an important component of most of the process studies proposed under Activity 2.2. None of these tracers can now be measured *in situ*, but have to be determined either on board a ship or on the return of samples to the laboratory. The development of shipboard mass spectrometers capable of measuring argon in near real time suggests that use of O_2/Ar ratios will be a powerful technique for use in SOLAS field experiments.

2.2.1.j Quantification of atmospheric boundary layer forcing

Although this activity focuses on oceanic processes, there is a need for accurate local surface forcing as well as regional-scale atmospheric boundary layer properties and surface fluxes in order to drive the models. This can be accomplished in the context of a field experiment by (1) measurement of surface meteorology and bulk fluxes from a ship or buoy along with (2) observations of baseline boundary layer properties (vertical structure of temperature and humidity, wind profiles, PBL top, integrated water vapour and liquid water) using a combination of radiosondes, lidar cloud ceilometer, wind profiling radar and microwave radiometer, and (3) satellite estimates of shortwave and longwave radiation, rainfall, cloud properties and whitecap fraction. These observations would be used as boundary conditions for ocean column models and to constrain the output of large-scale atmospheric models for regional studies.

2.2.1.k Use cabled observatories for high data rates or significant power demands for long periods

As observational requirements become more complex, field studies are encouraged to use emerging cabled observatory infrastructures because of the opportunities to sample physical (e.g., bubbles) and biogeochemical variables at high data rates for long periods (e.g., sampling at one minute intervals for months to years). Specialised instruments such as multi-frequency echosounders, optical bubble counters, and horizontally scanning Doppler sonars tend to require high data rates and power requirements that may be better suited to operation from a cabled observatory system than a long-term mooring. The observatory infrastructure may also afford opportunities to use autonomous vehicles to more effectively sample horizontal processes.

2.2.2 - Develop parameterisations for atmospheric, surface, and upper ocean environmental conditions, including: bubble generation, subsequent aerosol production, and sea ice. These should be based on wind speed, sea state, and the nature of surface-ocean and marine boundary layers

During wave breaking, large numbers of bubbles are formed. These bubbles are pulled down into the water by turbulent motions and by more coherent flows, such as Langmuir circulation. The bubble size distribution is time dependent, because the distribution is quickly modified by bubble sorting due to buoyancy. Buoyancy, dissolution and the local flow field control the subsequent development of the bubble field.

Under increasing hydrostatic pressure, the transfer of gases across the bubble-water interface is controlled by physico-chemical transport across yet another interfacial layer, the properties of which evolve and are modified by surfactants and microparticles. The gaseous composition of the bubbles therefore evolves in a complex manner along their individual trajectories. Depending on sea state and biogeochemical factors, varying fractions of the bubble population are forced into solution, whereas the remainder rise to the surface and burst. Thus, bubbles are a unique and potentially important extension of the air-sea interface, particularly for relatively insoluble gases such as N_2 and O_2.

When rising bubbles reach the surface, they burst and produce jet drops and film drops. A third type of drops, spume drops, are formed by direct tearing from the wave tops at wind speeds >9 m/s. These droplets enhance the effective surface area for exchange of constituents that are transferred across the air-water interface. Film, jet and spume droplets are collectively referred to as sea-spray aerosol. Small droplets can be dispersed throughout the boundary layer and can act as cloud condensation nuclei.

The highly turbulent flow field within a breaking wave will control the initial evolution of the bubble plume. At larger time scales (>2 s), buoyancy, dissolution, and coherent flows in the mixed layer will dominate the dynamics of the bubble field. The buoyant rise speed is highly bubble size dependent and is also influenced by surfactants adsorbing on bubble surface shortly after their generation. Surfactants, hydrostatic pressure, and the local air saturation levels in the mixed layer control bubble dissolution. The bubble size distribution will change with time in any given storm because of increased bubble-induced saturation levels.

One of the fundamental goals here is to determine how the generation and evolution of surface ocean bubble fields control the production of primary marine aerosols and the rates of gas transfer.

Sea spray will also carry with it the enhanced concentrations of biological and chemical constituents of the air-sea interface, in particular, natural and anthropogenic organic compounds, including gases. Sea spray therefore serves as a medium for transporting these properties up into the boundary layer and ultimately into the free atmosphere.

Long thought to be an impermeable cap on air-sea gas exchange, sea ice is now recognised as an active player in the marine carbon cycle (Delille et al., 2004; Papakyriakou et al., 2004; Semiletov et al., 2004), and under at least some conditions, CO_2 fluxes from the atmosphere into sea ice can be larger than into open water. The magnitude of the fluxes coupled with measurements of seawater pCO_2 below the ice have indicated that the ice is actively participating in the fluxes, not acting as a passive conduit, and current theories of the transport mechanisms in play are focussed on brine channel dynamics and $CaCO_3$ precipitation/dissolution within the brines, as well as on algal and microbial production cycles. To date, significant CO_2 fluxes have been observed either above or within sea ice in both the Arctic (Point Barrow, the Canadian Archipelago, and the southern Beaufort Sea) and in the Antarctic (the western Weddell Sea and the Australian sector). With the exception of the Beaufort Sea study, all of these measurements have been limited to spring, and we still do not know how representative the measurements have been and what the net annual flux may be. Of particular importance is whether CO_2 absorbed by sea ice, as has been consistently observed in spring data sets, is eventually released into the water or back into the atmosphere when the ice melts.

2.2.2.a Characterise breaking waves and associated whitecaps to enable the parameterisation of bubble and aerosol generation

Statistics such as breaking probability, whitecap coverage, and energy dissipation are highly dependent on parameters like wind speed and fetch. These relationships need to be determined in suitable field experiments. Concurrent measurements of turbulence and particles (sea-spray aerosols on the atmospheric side and bubbles on the oceanic side) are also required at time scales related in the breaking process.

2.2.2.b Measure sea spray aerosols by direct covariance (DC), mean vertical gradients, and budget residuals (see Activity 2.3)

The application of micrometeorological methods to measure sea spray aerosol fluxes has been explored by several investigators. Vertical gradients were measured by De Leeuw et al. (2000) to determine the production of sea spray aerosol in the surf zone. Particle size distributions were measured at three heights at a short distance down wind from the surf zone. The background aerosol distribution advected from elsewhere was accounted for by subtraction of particle size distributions measured just upwind from the surf. This method worked because the surf is a localised and continuous source. Over the open sea, where the source is distributed, the profiles often have no distinct gradient (G. de Leeuw, unpublished results from measurements from the FPN platform in the North Sea). Eddy covariance measurements were explored by Nilsson et al. (2001), who correlated total particle concentrations measured with a CPC and vertical wind speed measured with a sonic anemometer. This package was complemented with an optical particle counter to measure the size segregated fluxes (Geever et al., 2005). Equipping the particle counters with an inlet tube heated to 300 °C allows for the selective sampling sea spray aerosol particles (sea spray volatilises at ~600°C, whereas most other constituents are more volatile); (De Leeuw et al., 2003; 2005; Nilsson et al., 2003). Due to both inlet losses and the requirement to have relatively high particle concentrations for statistical reasons, the eddy covariance technique can mainly be applied to particles smaller than a few µm. The relaxed eddy accumulation (REA) method has also been suggested as a possible technique. REA would have the advantage that size-resolved chemical analysis could be used to add a chemical dimension to the fluxes. Fluxes of specific organic compounds or groups will be of high priority. This applies both to natural organic compounds with a biological origin, and anthropogenic compounds.

2.2.2.c Evaluate the role of surfactants in bubble formation and evolution

Studies of bubbles and turbulence in breaking waves should also include measurements of surface-active materials to determine their effects on the breaking and bubble generation processes. Measurements of the main microlayer constituents will be required and *in situ* measurements of surface tension (a proxy for the surfactant concentration) should be attempted. Other approaches include optical spectroscopy and mass spectrometry to determine constituents of the microlayer. Both laboratory measurements and oceanic studies in several locations with different concentrations of surfactants are needed.

2.2.2.d Use both optical and acoustic methods to quantify the evolution of bubble plumes

The penetration of near-surface bubble clouds can be determined from single-frequency acoustic sounders in the 50-200 kHz range. These instruments are capable of autonomous sampling at rates of order 10 seconds for up to a year. These basic bubble observations should be augmented where possible by multi-frequency acoustic sounders for more detailed resolution of bubble clouds and by optical sensors for determination of bubble size distributions. Small-scale (1-100 m) horizontal variability in bubble distributions caused by convergence from coherent mixed layer motions such as Langmuir circulation should also be resolved. Acoustic backscatter and acoustic Doppler profilers operated in a surface scanning mode (analogous to side scan sonar) are capable of making such measurements.

2.2.2.e Improve methods for measuring whitecap fraction

The fractional coverage of the ocean's surface by whitecaps is often used as a surrogate for the increase in gas transfer caused by the greater interfacial area when bubbles are present. Although relatively simple video cameras can be used to estimate this fraction, they give little or no information about the depth of the bubble field and the size distribution of the bubbles, both of which are important for controlling the rate of gas and particle exchange.

2.2.2.f The following sensors should be used to study wave breaking and the resultant bubble distributions:

- Surface-following floats for near-surface measurements of temperature, salinity, turbulence, bubbles and gases.

- Acoustical measurements of near surface flow field using the upper ocean bubbles as tracers.
- Acoustic Doppler measurements to obtain upper-ocean coherent and turbulent flows.
- Measurements of the directional wave field.
- Measurements of surface roughness (mean square slope of short wind waves).
- Bottom mounted sonars for bubble clouds (for shallow water only or from towers or moorings).
- Thermistor arrays with 1-second response to determine the ocean Mixed Layer Depth (MLD).
- Gas Tension Devices (GTD) for saturation levels of O_2, N_2, and pCO_2 as function of time and depth.
- Passive acoustic measurements of noise generated by the breaking.
- Concurrent atmospheric measurements of sea-spray aerosols from a tower or ships, including both physical and chemical size distributions.
- Aerosol Flux measurements from tower, ship or buoys.
- Photographic measurements of whitecap coverage from a tower or blimp.
- Near-surface vertical measurements with high-resolution profilers.

2.2.2.g Determine the role of sea ice in controlling air-sea CO_2 fluxes

Measurements of CO_2 fluxes above sea ice utilizing both direct covariance time series and spot measurements with flux chambers on bore holes in the ice are needed under as many different conditions as opportunity and logistical constraints allow. In addition, we must develop methods for examining and monitoring carbon system dynamics within sea ice. Oxygen micro-optodes (Mock et al., 2002) are used to monitor the biological system within ice brine channels, but microprobes that can also measure pCO_2 at low temperatures are also required. Other methods for measuring CO_2 in ice brines currently in development are limited in temporal resolution, but nonetheless have the capacity to provide useful information, particularly in the absence of effective pCO_2 microprobes. Non-destructive methods for monitoring pCO_2 in sea ice will also be amenable to deployment on ice buoys. All investigations of the carbon system within the ice must be coupled with studies of the physical microstructure of the ice in order to understand how temperature and brine channel structure contribute to the fluxes. High-resolution (both vertical and temporal) pCO_2 measurements are also needed in the water just below the ice in order to evaluate the efficiency of carbon export from the ice via excluded brines. Finally, laboratory measurements to define the carbonate system parameters at the low temperatures and high salinities of ice brines are required not only to estimate $CaCO_3$ precipitation and CO_2 evolution within the ice, but also to model the seasonal development of the carbon system in sea ice. Many of the fundamental transport questions to be addressed with regard to CO_2 will also be relevant to other gases in sea ice, such as DMS.

2.2.2.h The techniques above should be deployed from a variety of experimental platforms:

- Surface-following instruments can be tethered to ships. An advantage is that movement to different locations is relatively easy (for different upwelling regions or different concentrations of surfactants). Flux systems should be extended ahead of the ship beyond the internal boundary layer of the ship. Disadvantages of the ship-based platforms include: time constraints, inability to function at high wind speeds, possibility of ship emissions influencing data, disturbed flow, and the need to correct flux calculations for platform motions.

- Cabled observatories such as the NEPTUNE deep ocean site. Different nodes might be suitable for different regimes, for example, upwelling regions, low-high natural surfactants concentrations, coastal versus offshore. The NEPTUNE infrastructure would permit sampling over a large range of spatial and temporal scales.
- Selected coastal sites with towers extending above the internal boundary layer and a favourable foot print with as few islands, banks, rocks, etc. as possible.
- Autonomous Ocean Sampling Network. Brings together sophisticated new robotic vehicles. The operational system includes data collection by smart and adaptive platforms and sensors that relay information to shore in near real time.
- Platforms and towers in shallow water, such as LEO-15, Martha's Vineyard Coastal Observatory (MVCO), Venice, and Baltic platforms. These generally offer easy access for maintenance of equipment.
- Long piers, such as those at the Field Research Facility at Duck, NC, USA and in several other countries.

Table 2: Physical Observations required on the ocean side of the air-sea interface.

Parameter/Process	Time scale	Spatial scale	Instrumentation
Bubble size distribution in plumes (*in situ)*	Milliseconds to seconds	Millimeters	Optical bubble counters
Bubble size distributions in tenuous clouds (*in situ)*	Seconds	Centimeters	Acoustical bubble counters (Resonators, travel time sonars)
Bubble clouds	Minutes	Meters	Single or multifrequency backscatter sonars
Turbulence dissipation	<Seconds	Centimeters	ADV, Dopbeams, and microstructure profilers
Gas saturation levels (N_2, O_2, Ar, other inert gases, and noble gases)	Minutes	Meters	GTD
Mixed layer depth	Minutes	Meters	Thermistor arrays, profiling CTDs
Air-sea temperature difference	<Seconds	Micrometers	Thermister profilers.
Surfactants	Seconds	Micrometers	surface tension meters; Langmuir troughs;

Activity 2.3 – Processes in the Atmospheric Boundary Layer

Understanding atmospheric boundary layer physics and biogeochemical processes is critical for predicting the air-sea exchange of gases and particles and in determining how such exchanges and processes will affect and be affected by global change.

Introduction

The lower atmosphere, specifically the boundary layer, links processes occurring at the air-sea interface to the rest of the atmosphere. To quantify the vertical transport and fate of gases and particles in the atmosphere, it is necessary to understand the physical and chemical mechanisms involved. In general, the atmospheric boundary layer (ABL) is more difficult to observe over water than over land. Although there are important differences, a few model parameterisations still use values derived from measurements over land. In coastal regions the surface properties clearly change at the land-sea transition. Not surprisingly, the greatest observational difficulties arise at high wind speeds, when gas and particle transport may be especially important because of the increased role of sea spray (see Activities 1.1). The air-sea transports described here represent a sensitive part of the coupling between the ocean and atmosphere. Changes in climate variables such as temperature and wind speed will produce feedbacks through this coupling. Thus, understanding these processes is essential to the development of reliable predictions of the effects of global change.

Transport of energy, momentum and mass in the ABL is achieved through a variety of processes. Wet and dry deposition, sea-spray dynamics and atmospheric heterogeneity are important factors to consider. From the perspective of aerosol effects on climate, mass is most important for the direct aerosol effect (Mie-scattering), while number concentration is the key parameter for the indirect effect (cloud albedo and lifetime). Trace species are incorporated into cloud droplets by condensation, coagulation, and diffusive (Brownian) capture to droplets. The size spectra and associated hygroscopic properties of cloud condensation nuclei are critical in controlling the microphysical, chemical, and optical properties of clouds, which in turn affect chemical processing, wet deposition and boundary layer turbulence.

Table 3: Observations required on the atmospheric side of the air-sea interface.

Parameter	Time scale	Spatial scale	Instrumentation
Wind speed	Minutes	Meters	variety
Sensible and latent heat fluxes	Minutes	Meters	variety
Air-sea temperature difference	Minutes	Centimeters-Meters	variety
Aerosol fluxes (size resolved and chemically specified)	Seconds-Minutes	Centimeters-Meters	Eddy covariance; relaxed eddy accumulation; profile technique.
Complementary chemical and physical aerosol properties	Minutes-Days	Meters-Kilometers	Aerosol hygroscopicity, morphology, chemical analysis by filter or impactor sampling of components with low concentrations, e.g., Detailed or-

			ganic specification, number size resolution <100 nm diameter.
Wave field	Minutes		Wave wires, lasers, microwave.
Gas flux	10 Minutes		EC-APIMS (Eddy Covariance systems, Atmospheric Pressure Ionisation Mass Spectrometer systems)

2.3.1 - Determine a quantitative algorithm describing the generation of sea spray at each size between 0.1 and 50 μm radius as a function of wind speed and sea state to within a factor of three and to wind speeds of greater than 20 m/s

This goal requires the measurement of sea spray generated particle fluxes and other key variables that are expected to constitute the basis for parameterisation. Spray droplets are produced by white-cap bubble bursting (both film and jet droplet mechanisms) and direct wind-spume removal from the wave surface. Both mechanisms are principally associated with wave breaking.

The parameterisation of spray production to date has principally featured a dependence on wind speed or scaling with whitecap fraction. Bubble mediated production has been represented in terms of the bubble spectrum, which in turn may be parameterised in terms of wind speed and whitecap fraction. More recently, physically-based parameterisations have been developed using wave breaking energy flux, water surface tension, slope of the breaking wave, particle fall velocity, friction velocity, and the distribution of wind fluctuations near the breaking wave top.

The complicated physics of droplet production suggests that a comprehensive characterisation of oceanic droplets using only field measurements will require a measurement programme of unprecedented complexity. Characterisations of wave breaking statistics (breaking probability distribution, whitecap fraction, energy dissipation, etc.) and droplet/aerosol size distributions clearly require field measurements. In contrast, essential details of turbulence and bubble concentration profiles may be more tractable through carefully designed laboratory experiments. Similarly, aspects of the problem such as a hypothesised dependence of whitecap fraction on water temperature may also be amenable to laboratory studies.

For sea salt particles, the evidence suggests that the smallest particles of interest are about 0.01-μm dry radius (film drops), although even smaller particles may serve as nuclei for particle growth. The largest drops of relevance are on the order of 50 μm radius. These larger droplets are produced by both jet and spume mechanisms, with spume becoming more important at sizes greater than 10 μm and winds speeds exceeding about 15 m/s.

2.3.1.a Measure or infer aerosol particle fluxes under a variety of environmental conditions using direct eddy covariance, mean vertical gradients, budget residuals, and other methods

Micrometeorological techniques such as eddy covariance, vertical profiles and relaxed eddy accumulation (REA) were discussed in section 2.2.2.b.

The eddy-covariance method is essentially direct, but counting statistics limit the application to either high concentrations or to high volume sampling. Thus, this method is presently applicable to particles that are less than about 3 μm. For measurements above the source region, additional assumptions about deposition processes and near-surface profiles are required to relate such measurements to the source function. There are also some uncertainties in the nature of the vertical distribution of the source (e.g., droplet ejection heights versus the effects of disrupted flow patterns over breaking waves) that require experimental investigation. Particle fluxes can also be determined by inverse modelling, where the source function is adjusted to match observed size distributions (Vignati et al., 2001; De Leeuw et al., 2003). See 2.1.1.b.

2.3.1.b Measure and model the many factors controlling sea spray production alongside the particle flux measurements over a wide range of conditions

The observations needed for the field aspect of this work feature a concentration of sensors near the interface to characterise turbulence, bubble populations and droplets. We are also interested in obtaining data over a wide range of wind speeds with a range of other parameterisation variables which, given the statistical difficulties presented by breaking waves, implies measurement campaigns of unusually long duration. The combination of these requirements suggests use of a fixed tower approach, such as a coastal observatory. Certain aspects of the problem, such as breaking wave probabilities and statistical distributions, might best be approached with selected aircraft operations. Other aspects associated with particle/bubble dynamics in the wave boundary layers are best accomplished with specialised high-resolution numerical models. Here the emphasis will be on interpreting field measurements and establishing the sensitivity of simplified source characterisations (e.g., one effective source height versus a vertically distributed source) to details of the production, ejection and flow properties. The determination of what degree the primary aerosol source contributes to the observed marine aerosol and CCN spectra should be studied using detailed aerosol models that take into account the aging of the aerosol, other sources (including secondary aerosol formation: nucleation and condensation), sinks (wet and dry deposition), and cloud processing.

2.3.2 - Quantify the DMS flux from the surface ocean to the atmosphere and the transport of sulphur compounds through the MABL to permit modelling of temporally and spatially varying generation of CCN from DMS outgassing (with Activity 1.3)

Here the focus is on the physics of processes in the MABL that are pertinent to quantifying the transport of sulphur and halogen compounds through the MABL and on modelling the temporally and spatially varying generation of CCN from DMS outgassing from the ocean. The conceptual foundation for addressing this goal is a process model, which describes the distribution and transformation rates of sulphur species (and sulphur-derived CCN) that are derived from DMS generated in the surface ocean.

2.3.2.a Use direct eddy covariance measurements of the DMS flux to improve models of its flux

The flux of DMS will be developed from knowledge of the competing biotic and abiotic pathways leading to, and affecting, DMS in the surface ocean (Activity 1.2) and from parameterisation of the DMS exchange flux at the surface (Activity 2.1). High frequency measurements of DMS make direct eddy correlation now possible, allowing this component of the model to be constrained or tested by direct observations (Huebert et al., 2004).

2.3.2.b Physically-based based laboratory kinetic studies and numerical models should be used to design field studies of the conversion of DMS into other sulphur gases and aerosols (complementary with Activity 1.3)

The transformations of DMS in the atmosphere involve a combination of gas phase chemical reactions and heterogeneous processes involving cloud water, precipitation and aerosol particles, particularly sea salt droplets. Parameterisation of these complex processes in the model will be a considerable challenge. For example, the reaction pathways and kinetic constants for gas-phase transformations of DMS through to SO_2, SO_3, MSA and H_2SO_4 are poorly known and involve many untested assumptions. Thus, laboratory experiments are required to better refine this information so that kinetic models can provide more accurate knowledge of each reaction pathway. This will constrain the time scales for observations of the various intermediate products in the atmospheric sulphur cycle.

2.3.2.c. Challenge the physical transport processes controlling DMS transformation models with intensive process study measurements of as many sulphur species and oxidant concentrations as possible on diurnal time scales (complementary to Activity 1.3)

The process model of DMS transformation through SO_2 to CCN should be tested and refined through intensive process studies that measure the flux of DMS and the concentrations of the main products of DMS oxidation, including CCN. These should take place in regions of high DMS production without nearby volcanic and anthropogenic sulphur sources such as the equatorial Pacific near Christmas Island, where the source is uniquely homogenous. These studies should, as a minimum, incorporate direct measurements of concentrations and fluxes of DMS and SO_2, both of which have now been demonstrated. Measurements of CCN size spectra would also be needed to validate the model output. This intensive programme should include measurement of parameters required to constrain photochemical transformations (OH, Ozone, NO_x, halogen oxides, etc) and meteorological processes (clouds, soundings, etc.).

These process studies need to encompass a variety of environmental conditions relevant to the MABL sulphur cycle; i.e., cloud conditions, temperature, and sunlight intensity. Ideally, they would be performed in areas of known high DMS emission rates. For logistical reasons, they might also be conducted in parallel with process studies of DMS emission (complementary to Activity 1.2).

The chemistry of DMS transformation becomes more complex once reactive gas species are formed that can interact with cloud water, aerosol and sea salt droplets. This will be particularly true of SO_2. Thus, both the model and the observational process studies will need to constrain these factors. This will require knowledge of atmospheric water, aerosol and sea salt content, and the composition of these heterogeneous phases that will affect SO_2 after absorption. Two components of the transformation pathways can be identified as requiring special attention because of our current lack of knowledge. The first is the loss function of SO_2 from the MABL through processes other than formation of sulphate aerosols (and ultimately CCN) such as direct dry deposition, rainout and scavenging by sea salt droplets. The second is the transformation of SO_2 to sulphate aerosol and thence to CCN.

Limited process studies focused on the transformation of SO_2 to CCN (and competing loss processes) can be carried out in oceanic regions of high atmospheric SO_2; e.g., near volcanoes. Suitable examples of such volcanic sources exist, for example the Miyake-Jima volcano, Japan (Uematsu et al., 2004). This study would determine the concentrations of SO_2, sulphate aerosol and CCN downstream of the source region. This information, coupled with a dispersion model for SO_2 emissions, should allow refinement of SO_2 transformation rates. DMS should also be measured, along with MSA, to obtain information on the oceanic emission source strength.

Ozone deposition into the oceans represents a significant loss from the atmosphere (Figure 6). An accepted model for the description of dry deposition relies on the resistance approach (Wesely and Hicks, 2000). For gases which are destroyed by chemical reaction after deposition, the flux equation reduces to the form $F=-V_{dx}\ f_{xa}$, where the deposition velocity depends on different resistance terms, with $V_{dx} = (R_{at} + R_{am} + R_{cw})^{-1}$, where R_{at} is the turbulent/aerodynamic resistance reflecting the turbulent transport to the ocean surface, which is a function of sea surface roughness, wind speed and atmospheric stability.

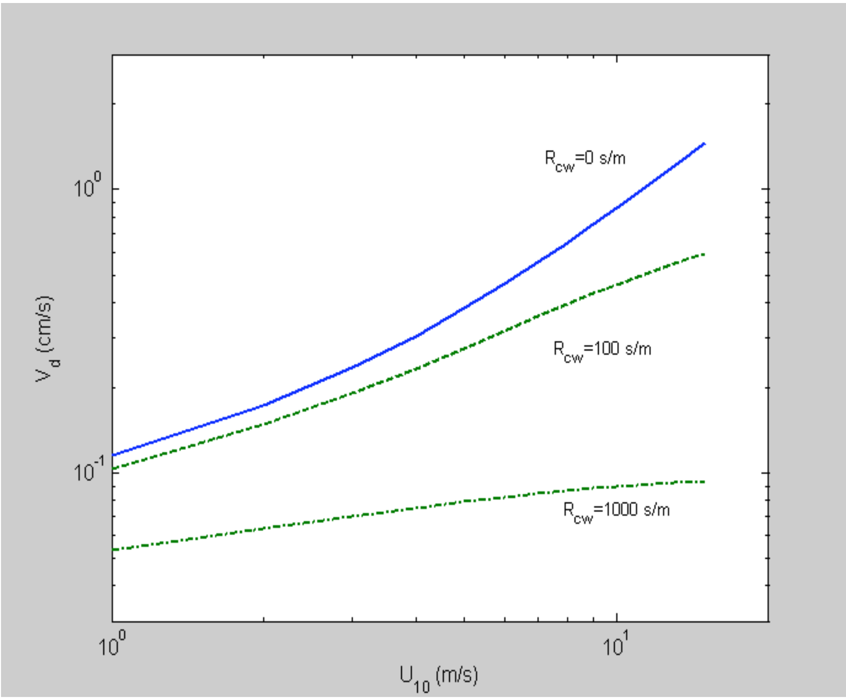

Figure 6: Ozone deposition velocity as a function of 10-m wind speed using the COARE bulk algorithm. The different lines are for different specifications of the oceanic resistance.

2.4 Cross Cutting Activities

2.4.1 - Data management

Data management and services within Focus 2 encompasses several needs:

- To serve data from individual process studies in a timely manner
- To provide model code and pertinent output from the models to compare with field studies
- To disseminate bulk parameterisations and algorithms with appropriate descriptions and disclaimers (metadata)
- To provide easy access to global forcing fields and climatologies

The following section outlines the individual components and resources needed to implement a data management system. A central tenet in SOLAS Data Management is to make available the output of the data and model results to the community at large. This will engage the larger group of investigators in the studies and provide a larger scope of approaches to interpreting the data that are critical for the interdisciplinary nature of the programme. The data management approach should be pro-active where data managers provide, or act as the liaison to obtain necessary background data for the process studies including: historical field data, larger scale climatological and remotely sensed data, and relevant data output.

2.4.1.a Make available data from individual process studies in a timely manner

A basic requirement will be a data management system that can provide data between investigators of field programmes proposed in this implementation plan. The well received approach of a two stage data release of a protected data site for investigators involved in the study followed by an open release is advocated. Following the approach of several large programmes in the past, a central data site seems most appropriate. Data from different investigators should be merged at the earliest stage and/or data should be easily linkable through unequivocal common fields with a set format such as year day and location (x, y, z). The files would be accompanied with a metadata set that clearly indicates the release dates and versions of the data in the appropriate fields. Enhancements such as visualisation software and automatic means of combining datasets will be valuable. Decisions on software packages used to serve the data should be made at the initial stages of the programme. Data quality flags linked to metadata on the accuracy/precision and level of quality control are considered critical.

2.4.1.b Provide model code and pertinent output from the models to compare with field studies

An important component of SOLAS will be to provide parameterisations and validation data for models. However, model output can also provide key background information and hypotheses of the effect of SOLAS processes on the pertinent biogeochemical cycles. A critical step for developing each process study will be investigation of the models describing the relevant processes. The data management system should be set up to conveniently provide the data output. Model description and model code should be readily available through the data management system.

2.4.1.c Disseminate bulk parameterisations and algorithms with appropriate descriptions and disclaimers

The data management system should provide a broader function than simply managing data from the SOLAS community. It should also serve as an easy and consistent access point of the bulk parameterisations for heat, momentum, water vapour and climate relevant gases that are developed in the programme as well as canonical parameterisations. Metadata should be provided with the parameterisations describing their proper use, units and limitations. Limited access and confusion about parameterisations is a common source of errors in interpretation of data and sometimes also in execution of models.

2.4.1.d Provide easy access to global forcing fields and climatologies

The SOLAS field studies are performed in the context of larger scale forcing and response. Increasingly these fields are available is near real time. However, the fields are in different locations and not always in a user-friendly format. Access to these sites through the data management system, possibly with reformatting, will be an important service of the system. For example, global wind forcing fields are presented at the different levels and with different units, including breaking down the u and v component and expressing winds in terms of quantities such as wind stress. For specific applications such expressions have merit, but for the larger SOLAS community, easily understandable, documented fields are important. Products such as gas transfer velocity fields for specific gases might be of interest. A query to the community of desired fields and intended use would be appropriate.

The data management functions are greater than those provided historically and will require greater resources. Compared to previous programmes in the last decade, much of the data infrastructure is in place and many of the merging and data visualisation schemes are well developed, making it possible to get the system up and running expediently. The basic infrastructure should be set up, up front with an appreciation of the data handling capacities necessary for the next decade of SOLAS. A central data dissemination system for all of SOLAS is advocated.

2.4.2 - Infrastructure and technology

The achievement of the SOLAS Focus 2 goals will require use of that infrastructure and technology that presently exists and of some that will be necessary to develop. The SOLAS Focus 2 Implementation Plan requires a wide range of observational and modelling studies of the upper-ocean, lower atmosphere and air-sea interface. The implementation of the programme will be performed with a combination of well-defined process studies, sustained observations, and model-observational comparisons and reconciliation.

Through this plan, the SOLAS project will study regional and seasonal air-sea climate relevant compound (CRC) fluxes. These studies will be performed through international coordination between ocean and atmospheric physicists, biologists and chemists. These activities will help explore components of small-scale feedbacks that effect local climate.

FOCUS 2 will make use of the following existing and development infrastructure:

- Unmanned high-wind platforms

- Satellite remote sensing (e.g., Space-based Air-Sea Exchange of CO_2 (SPACE) Integration of remote sensing and *in situ* measurements and modelling studies)
- Aircraft and platform remote sensing
- Wave follower
- ML Lidar
- Profilers
- Spar buoy development
- Ocean Stations Mike, Papa, etc
- Dedicated SOLAS meteorological flux packages are recommended for research vessels, buoys and Ocean Observatories Initiative (OOI) and Ocean Research Interactive Observatory Networks (ORION)
- Cape Verde Atmospheric and Oceanic Monitoring Station.

2.4.2.a SOLAS and the Global Ocean Observation Initiatives for coastal, regional, and global measurements that complement SOLAS

The OOI has three primary elements: 1) a regional cabled network consisting of interconnected sites on the seafloor spanning several geological and oceanographic features and processes, 2) relocatable deep-sea buoys that could also be deployed in harsh environments such as the Southern Ocean, and 3) new construction or enhancements to existing systems leading to an expanded network of coastal observatories.

The OOI will begin building an openly accessible network of ocean observatories to facilitate the collection of long time-series data sets needed to understand the dynamics of biological, chemical, geological and physical processes. The primary infrastructure for components of the OOI includes dedicated fiber-optic cables to shore and moorings capable of two-way communications with a shore station. Moorings are envisaged to be either freestanding, as for the global array of buoys, or attached to fiber-optic cables to provide the capability for water column investigations. Seafloor junction boxes connected to this primary infrastructure will support individual instruments or instrument clusters at varying distances from cables as well as the moorings. These junction boxes include undersea connectors that provide not only the power and two-way communication needed to support seafloor instrumentation, but also the capability to exchange instrumentation *in situ* when necessary for conducting new experiments or for repairing existing instruments.

2.4.3 - Satellite remote sensing

Satellites also provide whitecap cover, halogens, methane, clouds, SST, Chlorohyll, aerosolwind speeds, roughness, wave height, from various satellites (e.g., MERIS, AATSR, etc) Satellites will be quite important in the future, and in SOLAS we have a good representation.

One promising technique to estimate a globally averaged CO_2 flux over the ocean is to integrate in space and time the air-sea exchange of local CO_2 flux. Error sources for these estimates are mainly linked to uncertainties concerning the exchange coefficient and the space-time distribution of ΔpCO_2. Traditionally, the surface ocean carbon dioxide fields are taken from Takahashi et al. (1999), monthly data assimilations and much of the research in the past has focused on the determination of the air-sea gas transfer velocity.

Several parameterisations exist for the gas transfer velocity, which are typically expressed as a function of wind speed at a 10m height. Some common parameterisations include formulae provided by Liss and Merlivat (1986), Wanninkhof (1992), Wanninkhof and McGillis (1999), and Nightingale et al. (2000b). While similar in their general dependence on the wind speed, the models differ in their prediction of the CO_2 concentration difference across the atmospheric and oceanic boundary layers and assume different transport mechanisms that are involved in the transfer.

Regarding the use of satellites to infer gas fluxes between the ocean and the atmosphere from space, several issues emerge:

- Most existing flux parameterisations assume the wind to be the sole factor controlling the gas transfer. As such, the associated global and regional estimates of ocean-atmosphere gas fluxes are limited by the uncertainties in current wind speed based gas transfer velocity parameterisations (Boutin et al., 2002). The validity of such a relationship is vigorously debated since it is poorly constrained by the field data and produces widely varying global ocean flux estimates (e.g., in the case of CO_2 from 1.2 to 2.7 GtC/yr). Nevertheless, the first step toward satellite determinations of gas fluxes should satellite wind and SST information in existing parameterisations of the gas transfer velocity.
- It is known that wind is not the only controlling factor but that many other factors affect the air-sea flux of gases, such as sea surface roughness and the near surface turbulence. In particular, it was shown that the gas transfer rate is correlated with the Mean Square Slope (MSS) of short wind waves (e.g., Jähne, 1980; Bock et al., 1999). Jackson et al. (1992) showed that the wave MSS in the gravity-capillary region of the surface slope spectrum is a robust predictor of transfer velocity and can be estimated from nadir-looking microwave altimeters using geometric optics specular scattering model. Along those lines (Glover et al., 2002) developed an algorithm for estimating regional and global air-sea gas exchange rates using the dual-frequency TOPEX and Jason-1 radar altimeter return pulses. The differential scattering of the dual frequency altimeter (Ku- and C-band) allows the estimation of small-scale MSS from which the transfer velocity can be computed using sea surface roughness parameterisation. Those estimates of transfer velocities are similar to those obtained from commonly used wind speed parameterisations, but the global climatology yields an average global transfer velocity that is approximately 30% lower than that predicted by the Wanninkhof (1992) wind speed model. However, the degree of this difference depends mostly on the inherent discrepancy in the underlying wind speed that is not known and needs to be tested using satellite data of surface roughness and wind speed or stress.
- It is known that surface films may also lead to a strong decrease in transfer rate (e.g., Frew et al., 1990; 2004; Bock et al., 1999), and that bubbles mediate transport through air entrainment due to wave breaking (e.g., Woolf, 1997; Farmer et al. 1993). Information about surface films and surface roughness can also be gained from SAR instruments. This information could be used, in principle, to provide global scale gas flux using SAR images. However, their availability on a routine basis on a global scale is hindering progress at the present time. SAR instruments may

additionally be used to infer information about surface waves. The role of capillary waves in enhancing the gas process have also been investigated extensively (e.g., Kitaigorodskii and Donelan, 1984; Coantic, 1986; Back and McCready, 1988; Csanady, 1990; Szeri, 1997; Saylor and Handler, 1997; Bock et al., 1999).

Estimation of air-sea gas fluxes require more information than gas concentration gradient, including net heat and freshwater fluxes. Air-sea fluxes also include dust that is advected over the ocean from continents and that fertilises the upper ocean biosphere. Dust clouds over the ocean can be detected from satellite visible observations, and the respective information can be combined with observations of oceanic primary productivity to estimate its effect on the stimulation of plankton growth. In the context of SOLAS, the role of biology and changing the surface pCO_2, and thus changing the air-sea flux of CO_2, needs to be considered. The amount of biomass in the upper ocean can be estimated from satellites using instruments such as SeaWifs or MODIS. For the most comprehensive description of upper ocean processes and their effect on air-sea fluxes, a biogeochemical model needs to be combined with a physical model to estimate the fluxes of interest using satellite and *in situ* data as well as the method of data assimilation for parameter estimation.

2.4.4 - Modelling

Most of the SOLAS efforts in the field will be spent on local studies, sampling individual sites either intensively for a limited period of time, or at larger intervals over several years. Such local studies make sense in a global project only if their conclusions can be extrapolated to the rest of the ocean. For example, the carbon system, in the surface ocean is complicated and varies rapidly in space and time, and global averages inferred by interpolation from shipboard surveys are unreliable. Only if there is some underlying order that varies much more slowly will SOLAS goals be attainable. Because SOLAS will focus primarily on small-scale processes, there is a need for process-level models that will integrate across the MABL and upper ocean layer. Such models will be used to formulate and evaluate parameterisations, especially for gas and other material fluxes. The models and parameters will then be used to the scaling up of the local studies by contributing to flux calculations over large regions of the ocean.

Modelling, a key tool for interpretation and testing scenarios, extrapolation and prediction, will play an essential role in this scaling up exercise. Satellite remote sensing is another method potentially capable of delivering synoptic descriptions of the ocean's state on a basin or global scale. Simultaneity of a suite of ocean colour sensors (i.e., SeaWiFS, MERIS, MODIS Aqua/Terra and of the tandem mission of the TOPEX/POSEIDON and JASON altimeters) provides the opportunity to analyze the large-scale distribution of phytoplankton biomass in relation to the forcing caused by the physical environment. Eclectic assimilation of both *in situ* and remotely sensed data help SOLAS achieve some of the Focus 2 goals; to integrate and parameterise the small scale processes-oriented studies so they can be applied to larger regional, and even global scale studies. Therefore, an integrated quantitative view of the biogeochemical cycles of the major biogenic gases in the surface ocean and lower atmosphere, incorporating the role of biota, physical transport, air-sea exchange and including some estimate of uncertainty will be obtained. This effort should also aim to assess the responses of oceanic ecosystems and the marine atmosphere to anthropogenic forces of environmental change.

The modelling activities planned for Focus 2 have the dual objectives of scaling up from small-scale process studies and improving the boundary parameterisations used in regional and global-scale coupled climate models. They should be developed simultaneously as an iterative process linking diverse, complementary approaches. We must ensure that we do not restrict investigations to one particular modelling strategy. Therefore this work can be structured along the three following lines:

2.4.4.a Models for assimilation of sparse observations

SOLAS process studies and time series station measurements are limited in space and/or time, and as such are difficult to interpret in terms of regional or global changes without any extra information. To obtain a coherent description of the physical state of the ocean, an ocean circulation model can be brought into consistency with all available data. If done in a mathematically rigorous way, the results will be dynamically consistent descriptions of the changing ocean that takes into account uncertainties of models and data alike. For that purpose many approaches are being used today to assimilate available satellite and *in situ* observations into regional and global ocean models. These techniques are now well developed in the WCRP community for estimating the physical state of the large scale ocean (Stammer and Chassignet, 2000; Stammer et al., 2002).

However, for ocean state estimation in general, resolution remains a major limitation. The role of large-scale to small-scale coupling in general circulation models remains to be established which is of importance especially for SOLAS applications. Moreover, the SOLAS studies will require developing the appropriate procedures needed to perform data assimilation into a coupled physical-biological system. Although a hierarchy of models and data assimilation approaches have been developed for that purpose, assimilation using coupled physical-chemical models remains a quantitative challenge. As mentioned earlier, eclectic data assimilation approaches, either through sequential estimation or inverse methods together with a description of the general circulation of the ocean, will be valuable tools to provide increased knowledge and insight of the role of transport and transformation of trace gases and nutrients in the oceanic and atmospheric boundary layers. The existing JGOFS, WOCE, CACGP and CLIVAR data sets along with all remotely sensed data provide the unique opportunity to gain insight into the functioning of coupled transport-chemistry models and to identify their deficiencies. A valuable approach is to investigate budgets of the large-scale circulation rather than simulating and investigating the small scale processes alone. The challenge for SOLAS is to combine existing and future physical, chemical and biological data sets with satellite information and with state-of-the-art dynamical coupled models, along with the use of full 4D assimilation schemes to fulfil Focus 2 integration objectives. A close cooperation of SOLAS with existing groups involved in estimating the physical state is an important element to success. Those efforts have been developed in the international CLIVAR and GODAE context (e.g., HYCOM, ECCO, MERSEA). They now need to be extended to incorporate biogeochemical elements and to provide maximum return from SOLAS measurements.

2.4.4.b Coupled Boundary Layer Models (CBLMs)

There is a need for intermediate-scale models to bridge the process scales of SOLAS observational projects and the regional and global 3-D models. In SOLAS, we plan to develop "test bed" 1-D CBLMs, where various parameterisations of air-sea exchange processes can be developed and evaluated prior to implementation into 3-D regional and global-scale coupled models. These CBLM models should consist of generalised oceanic mixed layer models, coupled to atmospheric "single column" models, extending from the air-sea interface to a height well above the atmospheric boundary layer so that cloud effects and mesoscale convective structures can be included. The one-dimensional column model assumes horizontal homogeneity and ignores or parameterises the coherent mixed layer processes, such as surface gravity waves and Langmuir circulation.

A candidate oceanic mixed layer module is the General Ocean Turbulence Model (GOTM), which was developed as a community upper ocean model (Burchard and Bolding, 2001; Burchard, 2002), where different oceanic turbulent mixing schemes (e.g., Mellor-Yamada, PWP, KPP, kappa-epsilon) could be implemented with a simple software switch. A potential atmospheric boundary layer module might be a single column atmospheric model, such as those developed in collaboration with the U.S. National Center for Atmospheric Research (SCCM) and the Canadian Centre for Climate Modelling and Analysis, which have identical physical processes as in the NCAR CCM and the CCC AGCM (e.g., Lohmann et al., 1999). The Canadian model includes extensive chemistry of sulphur and other climatically important substances.

The two modules need to be coupled by two-way transfer of information sufficient to calculate the air-sea fluxes on the time scales required by the exchange processes (of order 10 minutes for the parameterised processes). The GOTM modules do not currently contain gas phase chemistry. So, using CO_2 as an example, dissolved inorganic carbon chemistry, including alkalinity, would have to be built into the model. Similarly, gas flux formulations would have to be incorporated into the air-sea flux transfer module.

2.4.4.c Regional and global 3-D models

The 3-D model is at the top of the hierarchy of models that should be applied in SOLAS, and several potential routes can be followed. The 3-D process models resolve the spatial variability, can be used to test the hypotheses and the processes observed, and can be used to systematically evaluate the sub-grid scale parameterisations employed in the 1-D column model.

Two types of 3-D process model developments can be undertaken: the Large Eddy Simulation (LES) model of the atmospheric and oceanic boundary layers, and the Direct Numerical Simulation (DNS) model of the surface microlayer.

The methodology of LES, which was first introduced in atmospheric boundary layer research in the 70s, is increasingly adopted in the study of ocean mixed layer processes. An ocean LES model is formulated by calculating the dynamical processes of length scales ranging approximately from the mixed layer depth down to the grid size of the computation. This range encompasses the dominant coherent turbulent structures induced by the surface layer forcing and the vertical stratification. The surface layer forcing consists of the near-surface turbulence generated by wave breaking and the Langmuir circulation that arises from wave-current interaction. The ocean LES model will need to parameterise these two processes associated with the dynamics of surface gravity waves. The turbulent production generated by wave breaking can be modelled by applying a random surface forcing with TKE flux corresponding to the observational level. A common approach to imposing the process of Langmuir circulation is to introduce an additional advection by Stokes drift and a "vortex force" in the momentum equation.

Cross comparison of the computed parameters, such as the turbulence dissipation and the mixed layer depth, with other SOLAS observation activities is required for model diagnosis and validation. In parallel with the development of the coupled ocean-atmosphere column model, the oceanic mixed layer LES model will be coupled with the atmospheric boundary layer. An on-going effort to improve the existing ocean LES model is to incorporate a "large wave simulation" scheme, which resolves the motions of large wind-generated waves and swells, and parameterises the subgrid shorter waves.

The impact of surfactants on impeding the air-sea exchange is through the enhancement of laminar sublayer, induced by inhomogeneous surfactant distribution. Across the induced sublayer, the vertical flow processes are dominated by molecular transports. Improved parameterisations of the surfactant effect in the air-sea flux formulations call for a better understanding of the underlying mechanism of the interaction between the surfactants and the interfacial turbulent flow. Incorporating the surfactant effect into the lower atmosphere – upper ocean coupled models, including the 1-D column model as well as the 3-D LES model – also requires parameterisation of the surfactant effects on the sea-surface temperature, the surface stress, the evaporation, etc., in addition to the prevalent notion of transfer velocity reduction rate. To resolve the millimetres to centimetres microprocesses, a DNS model has been developed. The computational domain of the model is boundary fitted, as the upper boundary evolves with the effect of surface gravity waves. No subgrid parameterisations are employed, and the model resolution is fine enough to resolve the surface capillary waves, the molecular microlayer, and the Kolmogorov turbulent microscale. The surfactant effect is incorporated into the model through the dependence of the surface tension on the surfactant concentration. The computational domain of a direct numerical simulation model only extends several grids of the 1-D or 3-D upper ocean models. Accordingly, the modelled results can be used in *a priori* tests for validating and improving the subgrid scale parameterisations of upper ocean models.

2.4.5 - Approach, activities, timeline, and linkages to WGSF

The Implementation Group 2 has international representation spanning a broad range of oceanic and atmospheric research. Focus 2 is integrated with both Focus 1 and 3, the SOLAS Data Management Task Team (DMTT). In addition, Focus 2 will work closely with the World Climate Research Program's (WCRP), Working Group on Surface Fluxes (WGSF), and will develop strong interactions with other IGBP, SCOR and WCRP sponsored international programmes such as IMBER, LOICZ, CLIVAR and Joint Carbon Project (JCP). Implementation Group 2 has identified targeted global study sites that are essential to quantify air-sea exchange processes and elucidate the factors controlling interfacial transfer. These studies will be done in areas where there is a significant flux of climate relevant gases and where the oceanic and atmospheric environmental forcing is distinct and separable from just wind forcing.

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ACRONYMS

AATSR	Advanced along track scanning radiometer
ABL	Atmospheric boundary layer
ADV	Acoustic doppler current velocimeter
APL	Applied physics laboratory (university of Washington)
ASCM	Atmospheric single column model
ASGAMAGE	Air-sea gas exchange mage
ASIMET	Air-sea interaction improved meteorological
BATS	Bermuda Atlantic time-series study
BL	Boundary layer
CACGP	Commission on atmospheric chemistry and global pollution
CBLM	Coupled boundary layer model
CCC AGCM	Canadian centre for climate atmospheric general simulation model
CCN	Cloud condensation nuclei
CDOM	Chromophoric dissolved organic matter
CLIVAR	Climate variability and predictability research programme
CN	Condensation nuclei
COARE	Coupled ocean atmosphere response experiment
CPC	Condensation particle counter
CRC	Climate relevant compound
CTD	Conductivity/temperature/depth
DC	Direct covariance
DIC	Dissolved inorganic carbon
DMA	Differential mobility analyser
DMS	Dimethylsulphide
DMTT	Data management task team
DNS	Direct numerical simulation
EC-APIMS	Eddy covariance systems, atmospheric pressure ionisation mass spectrometer systems
ECCO	Estimating the circulation and climate of the ocean
ENSO	El niño- southern oscillation
EPIC	Eastern Pacific investigation of climate processes
FMCW	Frequency modulated continuous wave
FP6	Sixth framework programme
FPN	Forschungsplatform Nordsee (Research Platform North Sea Experiment)

GCM	General circulation model
GEO	Global environment outlook or Group on earth observations
GODAE	Global ocean data assimilation experiment
GOTM	General ocean turbulence model
GTD	Gas tension devices
HOT	Hawaii ocean time-series
HRMM	High resolution marine meteorology
HYCOM	Hybrid coordinate ocean model
IGBP	International geosphere- biosphere programme
IMBER	Integrated marine biogeochemistry and ecosystem research project
IPY	International polar year
JCP	Joint carbon project
JGOFS	Joint global ocean flux study
LES	Large eddy simulation
LOICZ	Land- ocean interactions in the coastal zone project
MABL	Marine atmospheric boundary layer
MBARI	Monterey bay aquarium research institute
MERIS	Medium resolution imaging spectrometer instrument
MERSEA	Marine environment and security for the European area
MODIS	Moderate resolution imaging spectroradiometer
MSA	Methane sulphonic acid
MSS	Mean square slope
MVCO	Martha's Vineyard coastal observatory
NACP	North American carbon program
NCAR CCM	National center for atmospheric research community climate model
NEPTUNE	North-East Pacific time-series undersea networked experiments
OASIS	Ocean-atmosphere-seafloor integration study
OCCC	Ocean carbon and climate change
ONR	Office of naval research
OOI	Ocean observatories initiative
ORION	Ocean research interactive observatory networks
ORS	Ocean reference stations
OVOC	Oxygenated volatile organic chemical
OWS	Ocean weather station
PAR	Photosynthetically available radiation

PMEL	Pacific marine environmental laboratory
POC	Particulate organic carbon
POP	Persistent organic pollutant
REA	Relaxed eddy accumulation
RGM	Reactive gaseous mercury
SAGE	SOLAS air-sea gas exchange
SAMOS	Shipboard automated meteorological oceanographic system
SAR	Synthetic aperture radar
SCCM	Single-column community climate model
SCOR	Scientific committee on oceanic research
SeaWiFS	Sea- viewing wide field of view sensor
SEP	Southeast Pacific
SPACE	Space-based air-sea exchange
TAO	Tropical atmosphere ocean project
TKE	Turbulent kinetic energy
TOGA	Tropical ocean global atmospheres
TOPEX	Topography experiment
TRITON	Triangle trans-ocean buoy network
TRMM	Tropical rainfall measuring mission
VAMOS	Variability of the American monsoon systems
VOCALS	VAMOS ocean-cloud-atmosphere-land study
VOS	Volunteer observing ship
WCRP	World climate research programme
WGCM	Working group on coupled modelling
WGNE	Working group on numerical experimentation
WGSF	Working group on surface fluxes
WOCE	World ocean circulation experiment