<u>Report on the "*k* Conundrum Workshop", Norwich, UK, 6-7 February 2008</u>

Thirty-five specialists in air-sea exchange from Europe and North America were brought together for a workshop on gas and particle transfer at the sea surface in Norwich, UK over the two days of 6-7 February. This workshop was instigated and developed by the COST Action 735 Working Group 2, which focuses on development of parameterization for gas and particle transfer velocities. Funding was provided by COST-735, the UK Surface Ocean Lower Atmosphere Study (SOLAS) via the UK Natural Environment Research Council (NERC). The workshop was organized through the SOLAS International Project Office (IPO) at the University of East Anglia in Norwich.

The intention of the workshop was to synthesize the available air-sea gas flux transfer velocity and particle generation (sea salt production) and deposition velocity parameterizations with a view to making recommendations toward the COST-735 goals, i.e. the creation of global flux datasets for various climate-relevant gases and particles. The workshop considered in-depth studies of the processes of air-sea exchange, but with the practical and immediate objectives at the forefront. The workshop included many formal presentations of research, but discussion and dialogue was key to the success of the meeting.

Two major incentives for the workshop were the wealth of new information arising from air-sea exchange science and the diversity of interpretations of historical and new data. In the case of the air-sea exchange of poorly soluble gases, there are a number of very diverse parameterizations that try to encapsulate current knowledge of the exchange processes in a transfer velocity, *k*. Recent published estimates of the net transfer of gases (ranging from global climatologies to local flux studies) use a variety of parameterizations and apply these in significantly different ways. Non-specialists are faced with a genuine conundrum in dealing with parameterizations offered by the experts on the transfer coefficients. Among the experts, there has been much open-ended debate without agreeing "a best guess" and "known uncertainties". We were determined to rectify this situation as far as possible.

Early sessions included presentations on major recent measurement campaigns, the issues of real and apparent variability in measured transfer coefficients and the historical development of gas transfer velocity parameterizations. There is a wealth of data, with good prospects of better data and more insights, but interpretation of the results remains quite contentious. A particular issue is whether apparent variability is only a product of measurement error or whether it reveals the need to include additional physics or chemistry. An example is wind-speed dependence, of for example gas transfer velocity or sea salt generation. It is clear that transfer is sensitive to wind speed, but experimental results show a significant spread at each wind speed. Is this just measurement error or does it signify that wind speed alone is an imperfect predictor of transfer? Undoubtedly, measurement errors are generally too large and prevent confident interpretation of the results. There are a number of experiments that suggest "additional physics" particularly in low winds and high winds. In low winds, there is evidence of significant gas transfer where traditional parameterizations would predict negligible transfer. There are too few measurements at high wind speeds to allow easy interpretation and while some of the results are closely clustered, other results are widely divergent.

Most of the commonly-used parameterizations are based on a fairly straightforward empirical interpretation of experimental results. Historically, many of these results are from laboratory experiments or lake studies, as the methods necessary for ocean studies took much time and effort to develop. Limited understanding of differences between laboratory tanks, lakes and the ocean still undermine attempts to synthesize all results. Two primary tasks are undertaken by parameterizations of gas transfer velocity. First, to describe the wind forcing of transfer, traditionally in terms of wind speed, but alternatively in terms of the friction velocity of the wind or other wind and/or sea state parameters. Secondly, to describe the dependence on gas properties – so that experimental results for one gas could be used for predictions of another gas. Additional tasks include accounting for possible effects of surface films or chemical reaction on gas transfer. It is generally understood that wind speed is an imperfect predictor, but the extent of this problem and whether there is an acceptable substitute were debated at length with only limited consensus. Most parameterizations take a fairly simple approach to gas-property-dependence (for weakly soluble gases), based on the molecular diffusivity of dissolved gases or Schmidt number (kinematic viscosity divided by diffusivity), though early laboratory results already suggested this may be insufficient at high wind speeds.

A more recent approach to parameterization has been to include more explicitly the known physics of all transfer across boundary layers at either side of the air-sea interface. One advantage of this approach is it can bring gas and particle exchange into a common framework with momentum and heat flux, which is conceptually appealing and enables a cross-fertilization of ideas between the “flux” and “exchange” communities. More narrowly it applies a similar framework to all gases, whereas traditionally, weakly soluble gases and more soluble or reactive gases have been considered separately. The “NOAA COARE” parameterization of heat fluxes has been extended to gas transfer. This immediately includes consideration of all boundary layers and includes the modification of wind-forcing by temperature gradients. It has been necessary to include a parameterization of bubble-mediated gas transfer, which significantly enhances the exchange of poorly soluble gases at moderate and high wind speeds. Areas of development within this framework include extending the scope to sea salt generation and particle exchange, experimenting with wave breaking or wind wave energy dissipation formulations (in place of simple wind forcing) and perfecting the description of buoyancy effects.

Throughout the meeting, there were provocative presentations describing radically new methods, surprising or unique measurements which demonstrate insight into important but neglected processes. We were made aware of some important gaps in our understanding, while also gaining some glimpses of a way forward. It was clear that while we might agree a consensus, or “best guess”, thus enabling the development of better flux climatologies, there also must be caveats and uncertainty bounds on these products. These studies also helped to determine where effort could be wisely invested in the future, with a reasonable expectation of removing caveats and reducing uncertainties in the near to medium future. Early measurements of gas exchange or sea-salt production were rarely accompanied by parallel measurements of the underlying processes; recent and future measurements should benefit from some of the new methods. Actual measurements of gas flux have improved but remain difficult. Heat is treated as a proxy of gas and enables detailed study of process, but there are significant problems in scaling to gases (and to a lesser extent, between gases).

Specific sessions on the second day were devoted to "aerosols", "high winds" and "low winds". Again, significant discussion by COST-735 Working Group 2 members and other attendees provided stimulating dialog and development of consensus on various issues of interest.

Actual measurements of flux above the sea surface are a relatively recent innovation for aerosols. There has been a narrowing in the uncertainty range for sea salt production (primary aerosol source function), but the methods are still quite new and very significant unknowns remain.

High winds are the conditions where sea-salt production is highly active and gas transfer rates are particularly high. It is also a condition when gas exchange does not seem to be simply regulated by wind speed, with sea-state development likely playing a significant role. Though fluxes are high, high winds are infrequent and make measurements difficult. There is a theoretical basis for proposing that parameterizations of gas transfer need to be more sophisticated in high winds and some measurements to support that contention, but more and better measurements are required. It is also important that methodologies are developed to measure the highly energetic processes active in high winds, especially associated with wave breaking and bubble-mediated transfer. Bubble-mediated transfer in principle can greatly complicate gas transfer. Bubble-mediated transfer is preferentially aligned to the injection (as opposed to simple exchange) of gases and has a unique and poorly known dependence on gas properties. In addition all transfer associated with large breaking waves and wave energy dissipation is modulated by sea state rather than following a simple wind-speed-dependence. Much of the above is still theoretical, but there is an emerging body of evidence on, for example, high transfer in strong winds, the supersaturating effects of bubbles and solubility-dependent transfer. The measured divergent behaviour of DMS and carbon dioxide at high winds is notable and strengthens the case for bubble-mediated transfer and parameterizations (e.g. NOAA COARE) that incorporate it. The development of methods applicable to very high winds and even hurricanes is welcome and promises strong future progress.

Traditionally it is supposed that sea salt generation will not occur in low winds and other gas and particle exchange will be very weak. There is however an expectation and limited evidence that gas transfer can be fairly strong in low winds, where surface cooling or rain drives turbulence. A vertical structure of temperature, density and gas concentration also can complicate low-wind studies and requires a careful appraisal of experimental methods and of calculation methods when estimating averaged daily fluxes. Rain hitting the sea surface is also another significant source of turbulence, which may be particularly significant in low winds when other sources of turbulence are weak. Surface films and chemical enhancement are also likely to be relatively significant in low winds.

It was the crucial objective of this workshop to enable the production of new flux climatologies by the best methods possible. To this purpose, on both days of the workshop we considered the practical tasks involved. One prerequisite for gas or particle global climatologies is that the underlying global fields of variables within the chosen parameterization must be available and validated. Wind-speed-based parameterizations have the significant virtue that climatologies of wind speed are now very mature. The difficulty then becomes in choosing the wind-speed parameterization. A "cubic + constant" dependence of gas transfer velocity on wind speed has significant merits, but a "square law" dependence remains the most supported. It was noted that a recent modification of the original Wanninkhof '92 parameterization "square law" on the basis of carbon isotopes by Sweeney and parameterizations based on purposeful tracer release are virtually indistinguishable. More demanding parameterizations such as NOAA COARE require further fields and the maturity of these fields is variable. The data necessary for including rain and buoyancy forcing is sufficient to merit inclusion. An area of considerable concern is the uncertainty surrounding wave energy dissipation, wave breaking and bubble distributions. While a number of methods – from wind-wave modelling to satellite-based measurement of foam through its microwave brightness temperature – are available, these are largely unvalidated. Any practical advantage of a greater understanding of the effects of sea state development on gas and particle exchange will be lost unless there is parallel progress on global fields (modelling and/or earth observation).

The assembly concluded that we should recommend (1) a "best current method" for constructing climatologies, (2) a "quick method" and (3) a clear expression of the limitations and known uncertainties of both methods. These outputs will need to be developed and written up carefully, but we debated the general form of our recommendations. We judge that NOAA COARE should be the basis of carefully constructed climatologies. The necessary flux fields are available, are already used for heat flux climatologies and this can be readily extended to gas transfer velocities. The "Sweeney modification of Wanninkhof '92" is recommended as a quick method; this requires only wind speed and the Schmidt number of the gas to calculate the transfer velocity. This simple method may be useful for carbon dioxide and less soluble gases, but cannot be recommended for significantly more soluble gases such as DMS. It is also likely that it will under-predict transfer in low wind regions. More generally the practical discrepancies between the two methods require further investigation. Various "known unknowns" were identified. In due course, additional physics can be drawn into the NOAA COARE framework, but first gaps in knowledge must be closed. For example, it is suspected that "fetch" and "sea state development" are important – with associated differences in transfer between coastal and open ocean conditions, or between hurricanes and more extensive storm events – but this requires further investigation. Low winds, buoyancy effects and the effects of surface films were also identified as worthy of detailed investigation.

Following on from the workshop, we will need to construct and promote the 3 outputs described above. We must also explore how best to tackle the identified knowledge gaps, including national and international research initiatives.

We must be realistic that "the conundrum is not solved". The methods for estimating fluxes will need to be revisited, but we believe that we have established a process for enabling the best practice and realistic estimates. Future research and workshops should take this process forward.

Meeting Agenda Day One

8:30	Introduction	Woolf
Recent/upcoming field campaigns		**Chair: Woolf**
8:40	DOGEE/SEESAW/HIWASE	Brooks/Goddard/Yelland
9:00	GasEx3	Ho
9:15	Discussion on measurements / expected data sets	
Variability effects		**Chair: Wanninkhof**
9:25	Natural vs Experimental Variability	Asher
9:45	pCO2 variability	Rutgersson
10:00	CO2 fluxes and background variability	Sempreviva
10:15	Carioca observed variability	Boutin
10:30	Coffee Break	
Historical perspectives		**Chair: Liss**
11:00	Liss/Merlivat	Liss
11:15	COARE algorithm	Fairall
11:40	Measurement and modeling of gas exchange	Zappa
11:55	Discussion on parameterization development	
Provocatives		**Chair: Hare**
12:15	Fluxes in unstable conditions	Rutgersson
12:25	Chemical enhancement of fluxes	Liss
12:35	Lunch	
Challenges for global air-sea flux fields		**Chair: Bell**
13:30	Challenges	Wanninkhof
14:00	Alternatives to wind speed	Woolf
14:15	Project Integration	Bell
14:45	Discussion on global fluxes	
Provocatives		**Chair: Hare**
15:00	Polarimetric methods	Zappa
15:15	Isotope methods	Andreas
15:30	Coffee Break	
Commonalities of species in parameterizations		**Chair: Fairall**
16:00	Commonalities gas and heat exchange	Rutgersson
16:15	Commonalities of species over a lake	Kugler
16:30	Similarities for low winds	Piskozub
16:45	Modeling efforts	Jenkins
17:00	Heat as a proxy	Garbe
17:15	Heat as not a proxy	Asher
17:30	Discussion on commonalities	
18:00	End	

19:30 Dinner at Maids Head

Meeting Agenda Day Two

High Winds		**Chair: McGillis**
8:30	Unique observations in high winds	McNeil
8:50	Shortcomings of measurements	Drennan
9:00	UK-SOLAS efforts	Brooks/Yelland
9:15	Spume in high winds	Fairall
9:30	Parameterizing bubbles	Woolf
9:45	Discussion on high wind parameterizations	
Provocatives		**Chair: Hare**
10:00	Waterside covariance measurements	McNeil
10:15	Uncertainties in fluxes	Huebert
10:30	Coffee Break	
Aerosols		**Chair: Andreas**
11:00	Source theory / parameterization	Fairall
11:15	Source measurement	de Leeuw
11:30	Errors in aerosol flux measurement	Norris
11:45	Discussion on aerosol fluxes	
12:00	Lunch	
Low Winds		**Chair: Ho**
13:00	Modeling efforts in low winds	Woolf
13:15	Measurements in low winds	Ward
13:30	Thermographic measurements of exchange	Garbe
13:45	Discussion on low wind parameterization	
Priorities for Future Work		**Chair: Bell**
14:00	Strategies for observations	Huebert
14:30	Strategies for flux synthesis	Bell
14:45	Discussion on strategic measurements / flux fields	
15:30	Coffee Break	
16:00	Discussion on wind speed only parameterizations	Wanninkhof
16:30	Discussion on improvements to parameterizations	Woolf
17:00	Discussion on scaling for parameterizations	Brooks
17:30	Discussion on products from this meeting	Hare
18:00	End	