

# Exchanges

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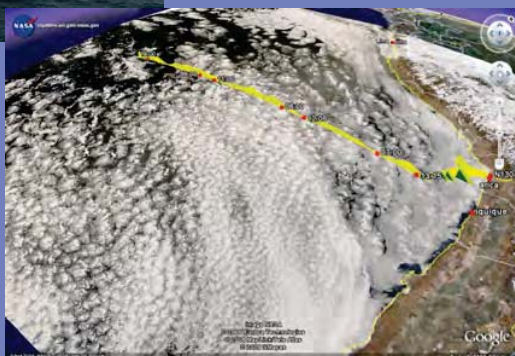
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## CLIVAR VAMOS Ocean-Cloud-Atmosphere-Land Study VOCALS



"The NSF C-130 flies by the NOAA Ronald H Brown underneath the stratocumulus during a research mission far off the Chilean coast on the 11th November 2008. Photo courtesy Cameron McNaughton, University of Hawaii"

"C-130 Flight track overlaid on a MODIS satellite image."



"Students from the Universidad de Arturo Prat, launch one of the four-times daily radiosondes from their shoreside campus in Iquique, Chile. Photo courtesy Carlye Calvin, UCAR"



"The NSF/NCAR C-130 preparing for a night-time take-off. Photo: Robert Wood"



"The Peruvian IMARPE R/V Jose Olaya"

CLIVAR is an international research programme dealing with climate variability and predictability on time-scales from months to centuries. CLIVAR is a component of the World Climate Research Programme (WCRP). WCRP is sponsored by the World Meteorological Organization, the International Council for Science and the Intergovernmental Oceanographic Commission of UNESCO.

proportional to friction velocity  $u^*$ , a measure of momentum transport from wind to the water surface.

Above 10 or 12 m/s the winds begin to cause the breaking of long waves, covering more of the surface with bubbles and making the interface ambiguous. The fraction of stress that is tangential to the interface is reduced as more and more of the wind stress contributes to "form drag," wind pushing on waves. Flow separation in the lee of waves further reduces the tangential stress that dominated in the mid-range. Bubbles are now the primary exchange agent for most gases, as they scavenge gas from the bulk and deliver it to the surface. Since the tendency of gases to partition into bubbles is a strong function of gas solubility, relatively insoluble gases like  $\text{CO}_2$  will experience much greater bubble enhancement than relatively soluble ones like DMS. Thus, the  $k$  vs  $U$  curves for various gases diverge in this high wind regime.

Although we did not encounter high winds during VOCALS, we were able to confirm that  $k_{\text{DMS}}$  in the mid-range was proportional to friction velocity. This confirms that the physical formulation is realistic under these conditions. Very little high-wind flux data exists, leaving that physics largely untested.

### Summary

The VOCALS DMS data set has educated us about many steps in the relationship between DMS production and aerosol formation. The production of DMS is spatially heterogeneous, with large increases at interfaces between eddies and currents. Dynamics create the chemical and physical environments that favor DMS-producing phytoplankton. The details of this control await further study.

The evasion of SW DMS to the atmosphere is controlled both by the amount of DMS in the water and by stress, the transport of wind momentum to the ocean's surface. We were able to validate a physical description of gas exchange in the mid-range of winds during VOCALS. Much more work is needed to test physical models in high winds.

This natural emission of DMS to the atmosphere dominates the sulfur budget in at least the non-coastal portions of the VOCALS region. While we still have to determine the extent over which upwind anthropogenic sulfur sources may have produced aerosol mass, it is clear that DMS is a major contributor.

Finally, measurements of DMS flux and concentration were used to quantify the project-average effective OH concentration. Since OH is the major oxidant of DMS (in this region, at least), this will be a valuable check on photochemical models leading to NSS and CCN formation.

DMS flux measurements illuminated a range of processes in the VOCALS program. However, many of these insights were qualitative, and therefore of limited utility to climate modelers. The role of dynamics on DMS production, in particular, demonstrates an intriguing connection between physical, biological, and chemical oceanography. No one discipline can understand DMS production in isolation.

### Acknowledgements

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## The Eastern Pacific Ocean is a source for short lived atmospheric gases: Glyoxal and Iodine Oxide

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Oceans cover 70% of the Earth surface, yet measurements of reactive gases like glyoxal, or iodine oxide have in the past been conducted mostly over land, or in coastal areas (see Figure 1). The open ocean marine atmosphere is among the most poorly probed atmospheric environments of our planet. Glyoxal is a short-lived gas that forms climate cooling secondary organic aerosol (SOA). Iodine oxide forms by destroying tropospheric ozone, and can nucleate new particles. The Atmospheric Trace Molecule Spectroscopy Laboratory (ATMOSpeclab) at the University of Colorado Boulder has designed, and assembled a prototype Ship Multi Axis Differential Absorption Spectroscopy instrument (CU SMAX-DOAS) that is optimized for the use from ships, and capable of measuring glyoxal, and iodine oxide, among other gases sensitively, selectively and directly in the atmosphere. This unique instrument was first deployed as part of the VOCALS-Rex field experiment aboard the NOAA Research Vessel Ron Brown from October through December 2008 over the Eastern tropical Pacific Ocean, and as part of other cruises aboard NOAA Research Vessels as part of the Tropical Atmosphere Ocean Program. The cruise tracks are also shown in Figure 1. We accomplished unambiguous spectral proof for the presence of glyoxal and iodine oxide in elevated concentrations inside the marine boundary layer more than 3000 km from land. This is surprising, because both gases are very short lived (atmospheric lifetime of seconds to hours), and cannot be transported from land sources. Ours are the first direct measurements of glyoxal and iodine oxide over the remote open ocean. Current atmospheric models fail to explain our observations.

The release of climate active gases from biologically active upwelling regions is a subject of ongoing research. Over the open tropical Pacific Ocean some satellite data showed concentrations of glyoxal (CHOCHO) (Kurosu et al., 2005; Wittrock et al., 2006; Vrekoussis et al., 2009) and iodine oxide (IO) (Schoenhardt et al., 2008). However, satellite inferences remain ambiguous. For glyoxal, the amounts differ between two satellite instruments, i.e., OMI and SCIAMACHY. Further, the GEOS-Chem and TM4 global models do not predict any glyoxal over the oceans (Myriokefalitakis et al., 2008; Fu et al., 2008). Finally, at tropical and sub tropical latitudes, the spatial patterns of glyoxal closely resemble those of chlorophyll-a in the surface waters. Chlorophyll-a affects ocean color in the blue spectral range, which is also used by satellites to measure glyoxal and iodine oxide. This spatial correspondence could either indicate ocean sources of glyoxal, or be the result of spectral artifacts caused by light absorption from chlorophyll-a or collocated chromophoric dissolved organic matter (CDOM). For iodine oxide, satellite measurements are inconclusive: two retrievals using the same SCIAMACHY data give different results over the tropical Pacific Ocean. One study finds iodine oxide is enhanced (Schoenhardt et al., 2008), the other study finds a lower upper limit column abundance here, and concludes that iodine oxide is not detectable over the tropical Pacific Ocean (Saiz-Lopez et al., 2007).

Measurements of glyoxal and iodine oxide are generally scarce. Near-surface observations of glyoxal are currently limited to air masses where the glyoxal source is from coastal, and/or

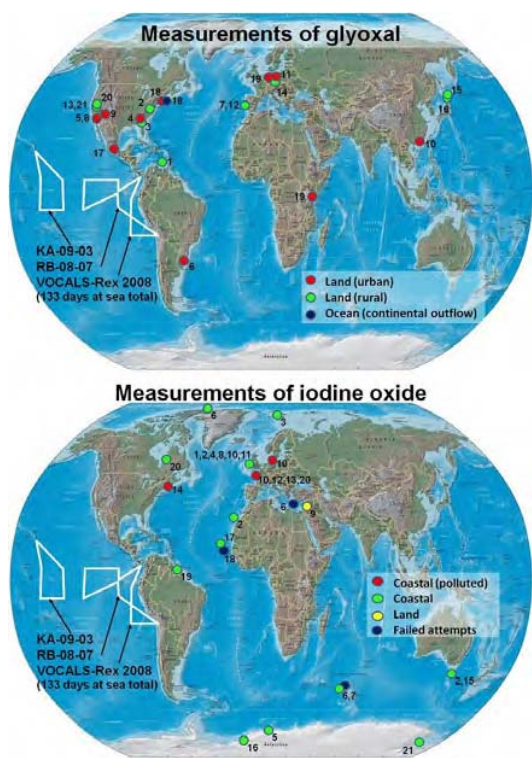


Figure 1: Ship cruise tracks from the VOCALS-REx, and two TAO cruises. Previous glyoxal and iodine oxide measurements exist only over land and in coastal areas. Top: (1) Zhou and Mopper (1990a,b); (2) Munger et al. (1995); (3) Lee et al. (1995); (4) Lee, et al. (1998); (5) Grosjean et al. (1996); (6) Grosjean and Grosjean (1999); (7) Borrego et al. (2000); (8) Kawamura et al. (2000); (9) Jing et al. (2001); (10) Ho and Yu (2002); (11) Moortgat et al. (2002); (12) Cerqueira et al. (2003); (13) Spaulding et al. (2002, 2003); (14) Mueller et al. (2005); (15) Matsunaga et al. (2004); (16) Ieda et al. (2006); (17) Volkamer et al. (2005); (18) Sinreich et al. (2007); (19) Wittrock et al. (2006); (20) Seaman et al. (2006); (21) Huisman et al., (2008). Bottom: (1) Alicke et al. (1999); (2) Allan et al. (2000); (3) Wittrock et al. (2000); (4) Carpenter et al. (2001); (5) Frieß et al. (2001); (6) Hönninger (2002); (7) Sebastian (2004); (8) Saiz-Lopez and Plane (2004); (9) Zingler and Platt (2005); (10) Peters et al. (2005); (11) Saiz-Lopez et al. (2006); (12) Whalley et al. (2007); (13) Wada et al. (2007); (14) Stutz et al. (2007); (15) Caine et al. (2007); (16) Saiz-Lopez et al. (2007); (17) Read et al. (2008); (18) Martin et al. (2009); (19) Butz et al. (2009); (20) Mahajan, ULeeds, pers. comm. (2010); (21) Kreher, NIWA, pers. comm. (2010).

terrestrial processes over land (see Figure 1). There is currently no unambiguous direct observation of either glyoxal or iodine oxide over the remote open ocean. This gap in our physical database creates bias in the way we think about the sources of these reactive gases as being mostly from coastal and/or terrestrial sources.

### Ship Multi Axis Differential Optical Absorption Spectroscopy (SMAX-DOAS)

In order to assess the validity of satellite retrievals of glyoxal and iodine oxide over the open ocean, the ATMOSpeclab has designed, and assembled an innovative Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS) instrument, the University of Colorado Ship MAX-DOAS (CU SMAX-DOAS), that is optimized for the use from ships. As solar stray light satellites, MAX-DOAS is a passive remote sensing technique (use of solar stray light) based on the well established DOAS technique. DOAS measures atmospheric trace gases by use of their specific narrow band (<5 nm) ultraviolet-visible light absorption structure in the open atmosphere, separating trace gas absorption from broadband molecule and aerosol extinction. MAX-DOAS is well developed for probing the atmosphere from land; however, past ship deployments did either not quantify ship movements, or suffered from a

variable field of view due to ship movements which affect the quantitative interpretation of these measurements. The CU SMAX-DOAS actively couples a set of two clinometers that measure pitch and roll of the ship, and compensates the telescope pointing position for ship movements. It has very good signal to noise ( $\sim 10^{-4}$  RMS), and features a very low dispersion telescope (0.3 degrees) that is optimized for measurements below clouds. The CU SMAX-DOAS consists of a 19" rack, a laptop PC, two clinometers (all mounted indoors, weight: 30kg, power: 350W, single 110V outlet, size: 93x108 cm<sup>2</sup> bench top space); and a telescope that is mounted outdoors on a railing (weight: 10kg, powered from indoor components; size: 56x13cm<sup>2</sup>). The telescope is coupled to the indoor components via flexible optical fibers of up to 22m length, allowing an adaptation of the CU SMAX-DOAS onto virtually any ship platform. For a picture of the instrument during VOCALS-REx, and a more detailed description see (Volkamer et al. 2009).

SMAX-DOAS is particularly well suited to test satellite retrievals over the open ocean, see Figure 2A. In contrast to the NADIR view from space, SMAX-DOAS collects scattered sunlight at a series of different elevation angles above the horizon. This makes SMAX-DOAS essentially insensitive to light absorption from chlorophyll, and/or ocean color effects. Over a low surface albedo environment like the ocean (typically <5%), atmospheric absorbers are inherently separated from ocean color effects, because the number of photons that enter the ocean, are scattered back into the atmosphere, and scattering into the SMAX-DOAS telescope is greatly reduced compared to the NADIR view from space (most photons that exit the ocean escape to space); indeed virtually all photons collected by SMAX-DOAS have never "seen" the ocean. Further, the satellite sensitivity varies over the height of the column: satellites need to make assumptions about the vertical distribution of a trace gas, and cannot retrieve information about the vertical distribution of the trace gas directly. In the case for SMAX-DOAS, trace gas column amounts recorded for sets of different elevation angles can be processed into a vertical profile of the atmospheric trace gas. Finally, the sensitivity of satellites is greatly reduced inside the marine boundary layer because of (1) the low surface albedo of water, (2) masking of the marine boundary layer absorbers from clouds, or (3) photons that are scattered back to space at higher altitudes (Rayleigh and Mie scattering). In contrast, SMAX-DOAS is especially sensitive inside the boundary layer. This increased sensitivity stems from the increased path length of the scattered light through the surface layer if the telescope is pointed close to the horizon. Clouds are not necessarily a limitation for SMAX-DOAS. The primary effect of clouds is that they increase the SMAX-DOAS sensitivity inside the cloud volume as the photon path length is greatly increased due to multiple scattering inside a cloud. The photon path length distribution is inherently constrained by observing absorption features of oxygen dimers that have a known distribution in the atmosphere. This enables to constrain the effect of aerosols on radiation fields by means of radiative transfer modeling.

### Spectral proof for the presence of glyoxal and iodine oxide during three ship cruises

In order to investigate the presence of glyoxal and iodine oxide over the remote Pacific Ocean, the CU SMAX-DOAS was deployed over the tropical Pacific Ocean in the frameworks of two projects. The first one was the VAMOS Ocean-Cloud-Atmosphere-Land Study - Regional Experiment (VOCALS-Rex, see <http://www.eol.ucar.edu/projects/vocals/>). Leg 1 of the cruise of the RV Ron Brown for VOCALS started on October 14, 2008, in Panama City heading south into the Pacific Ocean (see Figure 1). At 20° S and 85° W the ship stopped for a buoy replacement and headed Westwards to Arica, Chile, where leg 1 ended (November 3, 2008) and the ship stayed a couple of days in the harbour. Leg 2 (November 9 - December 3, 2008) consisted of going west to this buoy and back to Arica, with some deviations to North and South. The second project, which started after a couple of days, was cruise RB-08-07 as part of

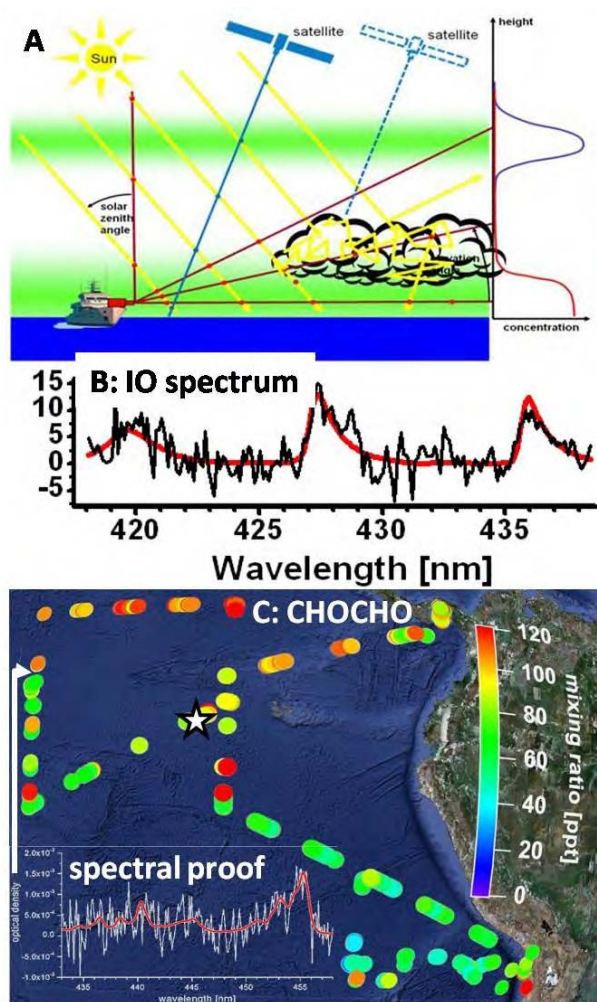


Figure 2: Measurement principle of SMAX-DOAS (panel A, see text), and spectral proof for the presence of glyoxal (B: ~70 ppt) and iodine oxide (C: ~2 ppt) over the tropical Pacific Ocean. The red line shows the scaled reference spectrum; overlaid in grey (black) is the instrument noise. The map shows VOCALS-Rex and TAO-2008 cruise tracks color coded by the mixing ratio of glyoxal (1ppt =  $2.46 \cdot 10^7$  molec  $\text{cm}^{-3}$ ).

the Tropical Atmosphere Ocean (TAO) program (<http://www.pmel.noaa.gov/tao/>). RB-08-07 started from Arica (December 9, 2008) and headed straight to the buoy line at  $95^\circ\text{W}$  servicing a buoy line from  $8^\circ\text{S}$  to  $0^\circ\text{S}$ . Then, the RV Ron Brown cruised to the  $110^\circ\text{W}$  buoy line continuing with buoys located from  $8^\circ\text{S}$  through  $8^\circ\text{N}$ . Lastly, the buoys between  $8^\circ\text{N}$  and  $2^\circ\text{N}$  on the  $95^\circ\text{W}$  buoy line were serviced before the vessel went straight back to Panama City and arrived there on January 11, 2009. In Figure 2C the whole measurement cruise is plotted with parts of South America on the right.

We succeeded to detect glyoxal and iodine oxide on a regular basis. Our data give unambiguous spectral proof for the presence of up to 120 ppt glyoxal, and up to 3.5 ppt iodine oxide over biologically active upwelling regions of the Pacific Ocean (Figs. 2B and 2C). This is surprising, because global models do not predict any glyoxal over the open ocean. The atmospheric lifetime of glyoxal is very short (few hours), and significant concentrations indicate local sources from the ocean. The source of the glyoxal as well as of IO is currently not understood, and is subject to ongoing activities in the ATMOSpeclab. On global scales, the isoprene oxidation is thought to be the most relevant glyoxal source (Myriokefalitakis et al., 2008; Fu et al., 2008), accounting for about one third of the terrestrial glyoxal source. We find similar or higher glyoxal concentrations over the remote tropical Pacific

Ocean than have been observed in remote air masses over land with abundant emissions of biogenic VOC precursor gases (see references in Fig 1). Assuming that 120ppt of glyoxal are produced from isoprene oxidation, an isoprene concentration in excess of one ppb is needed (1ppb = 1000 ppt). Typical isoprene concentrations over phytoplankton bloom areas are a factor of 2-35 lower (Meskhidze and Nenes, 2006); another source of glyoxal appears plausible. Ours is the first direct detection of glyoxal and iodine oxide over the open ocean. Our measurements locate both gases inside the marine boundary layer, and demonstrate for the first time unambiguously that the remote tropical Pacific Ocean is a source for reactive iodine species, and oxygenated hydrocarbons. Our data enable time synchronized comparisons with satellite data, as a prerequisite to assess the quantitative use of satellites to measure these reactive gases reliably, and assess their variability and predictability on global scales. Our data also enables to assess with confidence how these gases cool climate chemically, by (1) destroying the greenhouse gas ozone, (2) forming new particles that reflect a portion of the incoming solar radiation back to space, and (3) growing small particles into larger sizes that activate to form cloud droplets much more efficiently.

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### Predicting the Climate of the Coming Decades: A Workshop Summary and Directions for Research

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There is general scientific consensus that, because of increasing anthropogenic greenhouse gases, on average, by the middle of this century, the world will be warmer and sea level will be higher. However, because of natural fluctuations of the Earth system unrelated to greenhouse gases, the climate of each successive year may not be warmer than the previous one. Thus there may be clear and robust projections for anthropogenic influence in a particular region, but it is very likely that the climate in any given year or even decade between now and mid-century could be quite different from the projected anthropogenically forced trend. Given these dual 'natural' and anthropogenic influences on climate, how do we best plan on these different timescales?

To begin to tackle this inherently interdisciplinary challenge, we convened a workshop at the Rosenstiel School of Marine and Atmospheric Science of the University of Miami on January 11-14, 2010. The goal of the workshop was to bring together academics and practitioners who have shared interests in predicting the climate of the coming decades. This included researchers who were involved in developing climate prediction systems, studying decision making processes, developing applications of climate information, regional resource managers, and representatives from the insurance industry. This cross disciplinary approach to climate prediction and use is becoming increasingly important as the nations of the world develop new or enhance existing national climate services. Presentations covered three general topics: (1) status of decadal climate prediction efforts, (2) assessing user needs of decadal timescale climate information and (3) constraints on decision making. Here we summarize the main points made during presentations and discussions for each of these topics (all presentations and a list of participants can be found at

[http://www.clivar.org/organization/decadal/rsmas\\_decadal/rsmas\\_talks.php](http://www.clivar.org/organization/decadal/rsmas_decadal/rsmas_talks.php)), and suggest ways forward in preparing for the climate of the coming decades.

#### Status of decadal climate prediction efforts:

The decadal prediction problem represents a new 'frontier' in climate modeling. There is currently relatively high understanding and predictability at the seasonal to interannual timescales. On the longer end, we have a fairly good understanding of how the Earth will respond to greenhouse gas forcing, and have made projections of anthropogenic climate change that are generally consistent with the observed 20th century trends. The decadal timescale is a challenge because it contains both elements of natural fluctuations and anthropogenic change. This is largely uncharted territory in terms of climate prediction and includes a great deal of scientific uncertainty.

A number of presentations addressed the status of efforts to predict climate on a decadal timescale. There is currently a world-wide effort to examine whether including information about the present state of the climate (oceans, land, atmosphere and cryosphere) can introduce some predictability in the 'near-term' (i.e. the coming decades). This is coordinated through the Coupled Model Intercomparison Project (CMIP5), and results will be included in the next scientific assessment of the Intergovernmental Panel on Climate Change (IPCC). Many of the modeling centers involved in this effort were represented at the meeting, including those from the US (IRI, GFDL, NCAR, COLA, U. Miami), Australia (CAWCR-BoM/CSIRO), UK (Hadley Centre), and Canada. The presentations generally focused separately on predictability in the Atlantic and Pacific basins. In the Atlantic, the source of predictability is thought to come from the Atlantic Meridional Overturning