

ASP/VOCALS

A proposal for DOE/ASP Participation in

VOCALS-SouthEast Pacific Regional

Experiment (REx)

VOCALS-REx

P. H. Daum, Y. Liu, Y.-N. Lee, R. L. McGraw, J. Wang, L. I. Kleinman,
S. R. Springston, G. I. Senum

Brookhaven National Laboratory

J. Hudson
Desert Research Institute

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1. Introduction

Marine stratus clouds are an important, yet under-sampled component of the Earth's climate system. These clouds are known to have at least two stable modes: one with relatively large cloud droplets and relatively large drizzle rates and another with relatively smaller cloud droplets and little or no drizzle. Aerosol loading is thought to play a critical role in determining which stable mode is observed with stable long-lived non-precipitating clouds associated with high aerosol loading, and drizzling clouds with low aerosol loading. Marine stratus clouds also exhibit a strong diurnal cycle due to a pronounced cloud and radiation feedback involving changes in the net radiative flux at cloud top.

Extensive sheets of stratus and stratocumulus clouds are a common feature of the eastern boundary current upwelling regions of the world's oceans during summer. These clouds are known to exert a large scale cooling effect on the ocean surface and to be an important component of global cloud forcing. The large-scale cloud structure often fluctuates between solid stratus sheets with a high albedo, and thin, broken patches with low albedo (sometimes termed rift zones). These fluctuations are often observed over regions where the large-scale subsidence rate is relatively constant, which implicates microphysical processes or local thermodynamic processes as a possible cause of the large observed variations in albedo. Despite a rich theoretical underpinning, relatively few in-situ measurements of marine stratus properties have been made. This lack of relevant observational data has limited research progress over the past decade.

Because of the importance of marine stratus clouds to the climate system, and the potential for these coastal clouds to be influenced by anthropogenic aerosols, the DOE Atmospheric Science Program (ASP) conducted an airborne study of marine stratus over the eastern Pacific Ocean during the summer of 2005 (MASE study) in collaboration with the DOE Atmospheric Radiation Measurements (ARM) Program, and the California Institute of Technology which instrumented and directed flights of the CIRPAS Twin Otter. ARM's mobile facility was

located at Pt. Reyes National Seashore just north of San Francisco. The two aircraft sampled clouds over the Pt. Reyes site as well as over the coastal Eastern Pacific Ocean. Although very productive scientifically, the combination of prevailing meteorology and aerosol sources was such that nearly all of the clouds that were sampled during MASE were heavily influenced by anthropogenic aerosol and we were unable to explore the relationships between aerosol loading and cloud microphysics over a climatologically relevant range of conditions (Daum et al, 2006). Thus, there remains the need to conduct a similar experiment in a region where there is the possibility of sampling clouds with a more pristine character to put our observations of the relationship between aerosol and cloud properties on a broader foundation, and to provide more rigorous tests of the applicability of parameterizations that we have recently developed for the representations of cloud microphysics in large-scale models. Such improved parameterizations are needed to provide more accurate estimates of the magnitude of both the first and second indirect aerosol effects, which together are among the most uncertain components of the climate system.

For this reason we are proposing that ASP participate in the VAMOS Ocean-Cloud-Atmospheric-Land Study (VOCALS). VOCALS is an NSF sponsored international field program designed to develop an understanding of the physical and chemical processes central to the climate system of the Southeast Pacific (SEP). Details regarding this program may be found at <http://www.eol.ucar.edu/projects/vocals/>. The experiment will be conducted in a climatologically important, but poorly understood region of the globe where extensive areas of marine clouds exist (coverage ~70% during the month of October). Figure 1 shows the proposed VOCALS experimental area off the west coast of Chile. This region is the location of one of the largest and most persistent subtropical stratocumulus cloud decks in the world. It is formed because the Andes Mountains provide a sharp barrier to the generally westerly zonal flow in the region resulting in strong winds parallel to the coasts of Chile and Peru. This causes an intense upwelling of deep water to the surface causing the sea surface temperature (SST) to be low. The low SST, in combination with warm dry air aloft, results in the formation of a persistent layer of marine stratocumulus

clouds. This cloud layer helps maintain the cool SST resulting in tight couplings between the upper ocean and the atmosphere. The VOCALS experiment is scheduled for a one-month period, beginning during the month of October and ending in November of 2008.

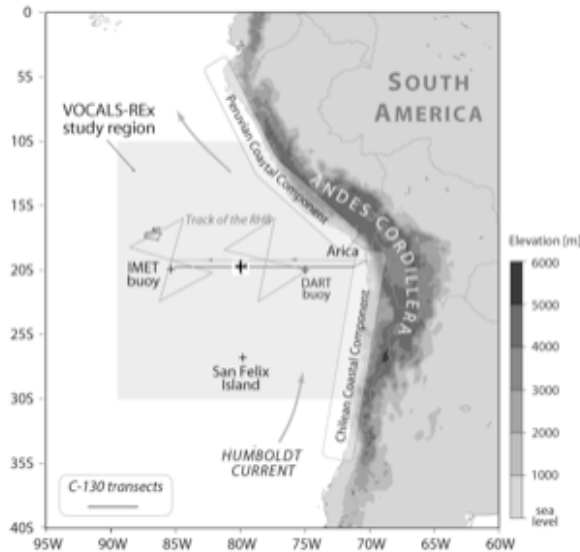


Figure 1. Proposed location of the VOCALS Experiment

The persistent cloud layer exhibits some interesting properties. Satellite observations reveal that the cloud droplet effective radius decreases sharply from west to east near the Chilean coast implying an increase in the cloud droplet number concentration, Figure 2. These gradients in droplet microphysics appear to be associated with the SO_2 emissions from the Chilean copper smelting industry whose SO_2 emissions of $\sim 1.5 \text{ TgS yr}^{-1}$ are comparable to the SO_2 emissions of countries such as Mexico and Germany and are directly upwind of the region where the sharpest gradient in effective radius is observed. These variations in the cloud droplet effective radius are associated with an increase in cloud reflectivity as observed by satellite, which results in a significant increase in cloud radiative forcing. If the relation between the anthropogenic SO_2 emissions and the increased cloud radiative forcing is verified, this constitutes an excellent example of the first indirect aerosol effect in action.

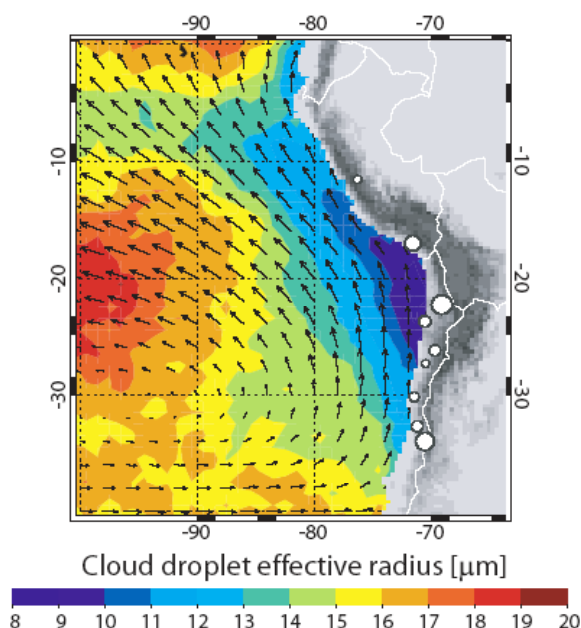


Figure 2. Austral spring season mean cloud droplet effective radius from MODIS; mean surface winds from Quicksat. Location of major copper smelters in the region are shown as white circles (relative area proportional to emissions). Taken from the VOCALS Scientific Program Overview.

Gradients in drizzle concentration have also been observed, and these may also be associated with concurrent gradients in aerosol concentration. Preliminary shipboard observations of drizzle in the project region show that regions of enhanced drizzle are associated with periods of reduced cloud droplet number concentration. In regions such as the SEP where cloud macrophysical characteristics are thought to be relatively constant, this implies that the variation in cloud droplet number concentration is associated with variation in the accumulation mode particle concentration. Such variations could arise from a number of possible sources. One possibility is that variations in aerosol loading are caused by variations in biological productivity and consequent variation in DMS concentrations. But, it is more likely that these variations are caused by perturbations in the aerosol loading from anthropogenic sources and there is some preliminary evidence to support this view.

The SEP cloud deck also exhibits “pockets of open cells” (POCs) that can persist for days. Although the cause of formation of these POCs is not known, it is hypothesized that they may be associated with regions of low aerosol concentration. In this view clouds that form in regions of low aerosol concentration have a smaller number of droplets that are larger in size, than clouds that form in regions of higher aerosol concentration. Such clouds are more likely to drizzle and it is thought that this drizzle results in depletion of CCN, which in turn, is responsible for maintenance of the POCs for significant periods of time. POCs are less frequently observed closer to shore where aerosol concentrations are higher lending credence to this hypothesis. POC’s and their potential association with aerosol loading are an example of the second indirect aerosol effect wherein cloud lifetime is lengthened by the suppression of drizzle due to alterations in cloud microphysics by anthropogenic aerosols.

The presence of a persistent stratus/stratocumulus cloud layer, and the possibility of significant alteration of essentially background clouds by fairly large scale anthropogenic sources make the SEP an ideal venue for the study of the indirect effect of aerosols on clouds. Offshore clouds are generally with little anthropogenic influence. Close to coast however, clouds can be influenced by aerosols from urban areas and from industrial sources such as smelters that are located both inland and along the coast, Figure 2. Thus, conditions will prevail such that we can sample the properties of clouds that are truly background, and those that are subjected to varying degrees of anthropogenic influence. This contrasts to the situation that we have encountered in our previous US based cloud studies (NARE and MASE) where clouds that were sampled were nearly always perturbed by substantial quantities of anthropogenic aerosols. The transition from pristine to anthropogenically influenced clouds and the presence of POC make the region especially useful for study of the second indirect aerosol effect, one of the most poorly understood phenomena in the entire climate system. VOCALS also provides an opportunity to examine indirect effects in an environment where clouds are thought to be most sensitive to the effects of anthropogenic aerosols.

2. Objectives

It is proposed that ASP/VOCALS be focused on aerosols, and how their chemical and microphysical properties, and their ability to act as CCN differ between remote marine air-masses and marine air-masses that have been influenced to varying degrees by anthropogenic aerosols, and how these differences in aerosol properties influence the microphysical properties of the clouds that form in these different environments. *The data collected during this study will allow examination of the relationship between aerosol composition, size, and CCN activity; between CCN loading and activity and cloud droplet microphysics; between cloud droplet microphysics and cloud radiative properties (first indirect effect studies); and between cloud droplet microphysics and the formation of drizzle (second indirect aerosol effects studies).* The data will also be used to examine the validity of recently developed parameterizations of cloud microphysical processes and properties designed for use in GCMs, and to develop the physical insight needed to develop more complete and sophisticated parameterizations of these quantities.

It is worth pointing out that although studies have been made of aerosol, CCN and cloud properties under conditions that are arguably similar to those that will be encountered during VOCALS, significant advances have been recently made in our ability to conduct the relevant measurements that should provide new insight into the complex relationships between aerosol and cloud properties. In particular we note that most previous studies have been limited by the availability of adequate data on aerosol composition, and by the lack of high time- resolution measurements of aerosol size distributions for particle diameters $< \sim 200$ nm. Most studies have either assumed a composition, or used data that is highly averaged (e.g., from filter samples or impactors that sampled for extended periods of time). Here, we propose to obtain detailed data on aerosol composition using a combination of the Particle into Liquid Sampler (PILS) which measures soluble inorganic and organic aerosol constituents, and the Aerodyne ToF-AMS which provides high time resolution measurements of aerosol size and the concentration of non-refractory aerosol constituents. Size

distributions below ~200 nm have typically been measured with DMA based systems with time resolution on the order of 60s; our new Fast Integrated Mobility Spectrometer (FIMS), can make similar measurements with a time resolution of ~ 1 s (Kulkarni and Wang 2006a; 2006b). Specific sets of objectives divided into three general categories are outlined below.

2.1 Aerosol and CCN Properties.

The primary objective here is to assess the extent to which it is necessary to specify aerosol composition in addition to size, in order to adequately predict the supersaturation at which aerosol particles will activate to form cloud droplets. It is our expectation that VOCALS will be a near ideal venue for examining the role of composition/size in determining the CCN behavior of ambient aerosols owing to the fact that a wide range of aerosol properties is likely to be encountered, ranging from remote marine aerosols, mixtures of remote marine aerosols and sulfate aerosols from anthropogenic sources, to situations where the aerosols are dominated by anthropogenic sulfate. Indeed it is highly likely that measurements during VOCALS will shed significant insight into the current controversy regarding whether it is more important to represent aerosol size distributions or composition in GCMs (Dusek et al., 2006).

Thus, one focus of ASP/VOCALS will be to devise and carry out a sampling strategy such that we are able to characterize the size distribution, chemical composition and CCN behavior of the major aerosol types that are important in this region of the globe.

Essential measurements required to conduct this characterization include the Desert Research Institute and Droplet Measurement Technologies measurements of CCN, AMS and PILS measurements of aerosol composition, and DMA and PCASP measurements of aerosol size distribution. A particularly useful set of measurements will be the critical supersaturation of size-selected aerosol particles in the different environments that will be encountered. Similar

experiments during the 2005 MASE study showed that the composition and CCN properties of the above-cloud aerosol was very different from than the composition and CCN properties of the below-cloud aerosol. We expect similar differences will exist between remote marine aerosol, and marine aerosols that have been heavily influenced by anthropogenic sources. Such information is critical for improved representation of aerosol particles in GCMs.

We will also take the opportunity to examine several additional issues. First, we plan to conduct measurements to examine the relative importance of natural *vs* anthropogenic aerosol precursors and how that varies as a function of distance offshore. Essential measurements for this task include measurements of sulfate, methanesulfonate, and NaCl in the aerosol phase (PILS and ToF-AMS), and DMS, SO₂ and various organic species in the gas phase (PTRMS, and pulsed fluorescence SO₂). Second, we plan to see whether organic aerosols in substantial excess of model predictions are observed at this location, and whether there is a layer of organic enriched aerosols of high concentration just above cloud-top as observed during MASE.

2.2 Aerosol/CCN/Cloud microphysics relationships

A key issue with regard to both the first and second indirect aerosol effects is the relationship between pre-cloud aerosol number concentrations, size distributions, composition, and cloud droplet microphysics including the cloud droplet number concentration (N_{cd}) and properties of the droplet size distribution such as the spectral dispersion, ϵ (ϵ , is defined as the ratio of the standard deviation to the mean droplet radius). These together determine the cloud droplet effective radius, r_e , which is the quantity used to specify cloud droplet microphysics in climate models. The primary goal here is to develop an understanding of the chemical, physical and dynamical quantities needed to specify N_{cd} , ϵ , and r_e , and to incorporate that understanding into parameterizations that can be used by models of various scales to predict cloud properties and to provide better estimates of indirect aerosol effects. We note that we, and others have been active in developing new and more physics based

parameterizations (e. g. Liu and Daum, 2000a; 2000b; Liu et al, 2006; Nenes and Seinfeld, 2004) of N_{cd} and r_e and plan to use the data collected during ASP/VOCALS to evaluate and extend these parameterizations. Although such evaluations could be done nearly anywhere, VOCALS presents a unique opportunity to sample a climatologically important region of clouds that that exhibits a broad range of microphysical properties.

The broad range of conditions in this region will be especially useful for examining the effects of aerosol loading on the spectral dispersion ϵ , and the latter on the magnitude of the first indirect aerosol effect. We have recently shown (Liu and Daum, 2002) that the addition of anthropogenic aerosols to marine clouds not only increases the cloud droplet number concentration, as indicated by the theory of the first indirect aerosol effect, but also increases ϵ . This increase in ϵ exerts a warming effect on the climate that acts to offset the first indirect aerosol effect by 10 to 80%, depending on the relationship between ϵ and N_{cd} (Liu and Daum, 2002; Rotstajn and Liu 2003; Peng and Lohmann, 2003). But, the relationship between ϵ and aerosol loading is highly uncertain because there are a number of factors that determine ϵ (e.g., updraft velocity, entrainment) besides aerosol loading, and because there is little published data on values of ϵ in remote marine clouds, to provide the reference point for the effect.

Thus, an important component of ASP/VOCALS will be sampling of clouds over a range of conditions such that the aerosol loading varies between remote background and polluted air. The cloud sampling strategy will be similar to the successful cloud sampling strategy that was employed during the recent MASE project. This strategy involved flying between two navigational points about 100 km apart at various altitudes starting with a below cloud leg, in-cloud legs at successively higher altitudes spaced by 100 – 200 m, and then an above cloud leg. This sampling strategy allowed us to examine the relationships and the variability between the properties of the pre-cloud aerosol and cloud droplet microphysics, and between various properties of cloud droplet microphysics such as r_e and ϵ , and cloud dynamics.

Analysis techniques and products will be similar to those that we employed in examining the MASE data (Daum et al, 2006). Our overall goal will be to use the data to understand the relationship between aerosol loading and cloud microphysics including N_{cd} , r_{cr} and ϵ . We will try to distinguish between the effects of aerosols, and the effects of updraft and turbulence on the cloud droplet spectrum by analyzing the characteristics of the relationship between ϵ and N_{cd} and ϵ and γ ($\gamma = L/N_{cd}$ where L is the cloud liquid water content) from the in-situ data. Physical reasoning suggests that these relationships will behave differently in response to cloud-scale updrafts and turbulent mixing. For example, an increase in updraft velocity will lead to increases in N_{cd} and γ , but a decrease in ϵ , which equates to increasing negative slopes in the corresponding ϵ - N_{cd} and β - γ (β is a parameter related to ϵ) relationships. Conversely, an increase in pre-cloud aerosol loading results in an increasing positive slope in the ϵ - N_{cd} relationship and an increasing negative slope in the β - γ relationship. Identifying these relationships from the in situ data therefore “fingerprints” the combination of processes that have contributed to the prevailing state of the cloud droplet structure. These “fingerprints” provide a mechanism for separating the effects aerosols from dynamics by identifying periods in which one or the other is dominant, whereupon the signal may be deconvolved in cases where both changes in aerosol loading and changes in updraft velocity are occurring simultaneously. Additional analyses will focus on evaluating parameterizations that we have developed to quantify the effects of dynamics on N_{cd} and ϵ .

The effect of aerosol loading on ϵ and the cloud droplet number concentration will be studied using measurements of pre-cloud aerosols and/or CCN measurements (e.g., at cloud base) together with corresponding droplet concentrations and size distributions. Similar to the studies we and others have conducted (Daum et al. 2006; Novakov 1994; Leaitch et al. 1996; Daum et al, 1987), *we will examine the relationship between pre-cloud aerosol (or CCN) number concentration and droplet concentration for similar types of clouds so*

that variation in cloud dynamics will not be a significant cause of variability in droplet concentration. We will also examine the relative dispersion of the aerosol size distribution and its relationship to N_{cd} and ϵ . The above results will be combined to evaluate expressions relating N_{cd} and ϵ to both pre-cloud aerosol properties (e.g., aerosol number concentration N_a and relative dispersion ϵ_a) and cloud dynamics (updraft and turbulence).

2.3 Aerosols, cloud microphysics, and the formation of drizzle

It is well known that the formation of drizzle is a strong function of cloud microphysics with drizzle being more probable in clouds with small numbers of large droplets than in clouds with large numbers of small droplets. This behavior forms the basis for the so-called second indirect aerosol effect wherein clouds that form under conditions influenced by anthropogenic aerosols exhibit larger numbers of small droplets than background clouds; leading to a lowered probability of drizzle formation, and thus a longer cloud lifetime, which in turn leads to an enhanced planetary albedo. Because it is difficult to measure differences in cloud lifetime due to this effect, observational estimates of the magnitude of the second indirect aerosol effect are highly uncertain. Estimates using models are equally uncertain, at least in part because parameterizations of the conversion of cloud drops to drizzle (generally termed the autoconversion process), and of the drizzle rate are very crude (see Liu et al, 2006a for an analysis). *So despite the fact that drizzle can exert a powerful effect on the structure, thickness and coverage of marine stratus/stratocumulus clouds, and is thought to be an important mechanism by which aerosols can impact the radiative balance of the Earth, little is known about how important this effect might be.*

Our primary objective here is to evaluate and extend several new physics-based parameterizations of the autoconversion process that we have recently developed (Liu and Daum, 2004; Liu et al, 2004; Liu et al, 2005, Liu et al, 2006a; Liu et al, 2006c) that hold great promise for improving the representation of the

drizzle formation process in GCMs. Preliminary tests of these parameterizations using data that was collected during the recent Marine Stratus Experiment (MASE) are very promising. But, the clouds that we sampled during MASE did not exhibit a wide range of microphysical properties, were generally strongly influenced by anthropogenic sources, and generally did not contain much drizzle. In contrast, it is highly likely that the clouds present in the VOCALS region, together with studies of POCs will span the range of microphysical properties needed to adequately test the parameterizations that we have developed.

Our examination of the observational data will be guided by the recent work that we have done on the theory of the drizzle formation (McGraw and Liu, 2003; 2004) and on parameterizations of the autoconversion process that we have recently developed (Liu and Daum, 2004; Liu et al, 2004; Liu et al, 2005, Liu et al, 2006a; Liu et al, 2006c). In the former case, drizzle formation is identified as a statistical barrier-crossing phenomenon that transforms cloud droplets to drizzle size with a rate dependent on turbulent diffusion, droplet collection efficiency, and lower-order moments of the droplet size distribution. For purposes of testing the theory, one requires measurements of only certain average properties of the cloud droplet spectrum, the most important of these being the cloud droplet number concentration, N_{cd} , and liquid water content, L . In addition to suggesting a resolution to the puzzle of long initiation times, this theory makes very interesting microphysical predictions concerning the stability behavior of clouds, and provides new insights into the threshold behavior of drizzle (Liu et al., 2004) and the second indirect effect (McGraw and Liu, 2003; 2004) -- all predictions that can be readily tested through field measurements of N_{cd} , L , and cloud turbulence. Cloud turbulence is currently represented in the theory using a single turbulent diffusion parameter, D_v , related to fluctuations in local supersaturation and droplet growth (McGraw and Liu, 2006). We will investigate several independent approaches to estimation of D_v . One approach is to estimate this parameter from concurrent measurements of L and N_{cd} as described recently (Liu et al., 2004). Another approach is to estimate D_v and updraft velocities from the G-1 gust probe measurements and use these together

with additional assumptions and model calculations, to back out fluctuations in supersaturation and droplet size.

We will also use the in-situ cloud data to examine the dependence of the autoconversion rate on droplet concentration. In the various parameterizations that have been developed, this dependence varies between no dependence (Kessler, 1969) to N_{cd}^{-1} (Liu and Daum, 2004), to $N_{cd}^{-3.3}$ (Beheng, 1994). To address this issue we will simulate the change in drizzle water content with time (autoconversion rate) using a stochastic collection equation model as has been done to obtain empirical parameterizations based on model simulations (Beheng, 1994), but will use measured droplet size distributions instead of assumed distributions to drive the model. The output of the model will be then compared to the different parameterizations. We will also address this issue by comparing measured vertical profiles of drizzle water content, to those calculated from a simple model using the different autoconversion parameterizations (e.g., Wood, 2000).

3. Major collaborators

Two major platforms form the core of the NSF VOCALS: (a) the NSF C-130 aircraft; (b) the NOAA R/V Ronald H Brown (RHB). The C-130 will provide detailed *in-situ* measurements of cloud microphysics, gas and aerosol physicochemical properties, lower tropospheric structure, and MBL turbulence, in addition to passive and active cloud and precipitation remote sensing measurements. A major focus will be on lagrangian experiments designed to document the lifecycle of POCs. The RHB will provide atmospheric, oceanographic and air-sea exchange datasets. Atmospheric measurements will include *in-situ* surface meteorology, rawinsonde profiles, and gas/aerosol physicochemical properties. Remote sensing measurements of cloud and precipitation will be made from the RHB using active millimeter and centimeter radars, a ceilometer, and a passive microwave radiometer. Oceanographic measurements from the RHB include the towed SeaSoar platform that will provide upper ocean vertical profile sampling, XBTs, surface drifters and

thermistor chains, an ADCP, and high time resolution sea-water DMS concentration.

In addition, a number of other platforms will provide additional important datasets especially at the land ocean boundary. A comprehensive near-coastal sampling strategy is planned for October 2007, which includes atmospheric thermodynamic and dynamic measurements with a light aircraft (Chilean AirForce Twin Otter), a Chilean Servicio Hidrográfico y Oceanográfico de la Armada de Chile (SHOA) research vessel, an elevated land site at a site, El Tofo, on the Chilean coastal range to measure cloud and aerosol microphysical properties, and enhanced meteorological observations at sites along the Chilean seaboard. Funding for most of these activities will be requested from FONDECYT, the Chilean science funding agency, with support from the Chilean Airforce (Twin Otter).

As part of the international contribution to VOCALS-REx, a group of Peruvian researchers are proposing a set of enhanced atmospheric measurements and an oceanographic/atmospheric coastal cruise with a Peruvian research vessel. The primary contribution to VOCALS-REx will consist of a 30 day cruise organized by the Instituto del Mar del Perú (IMARPE) to sample oceanography and meteorology along the Peruvian coastal zone from 4-18°S. It is also planned to request the NSF Deployment Pool 915 MHz wind profiler and Radio Acoustic Sounding System (RASS) on the cruise to provide high time resolution profiling observations of the horizontal wind and virtual temperature in the marine boundary layer and lower free troposphere. The wind profiler is also sensitive to precipitation-sized hydrometeors in low clouds, and will be used to characterize the occurrence of drizzle falling from the near-coastal stratocumulus clouds. Details regarding the instrument packages on these various platforms and how they will be deployed can be found at www.eol.ucar.edu/projects/vocals/

4. G-1 Flight plans

This program is viewed primarily as an experiment to sample the persistent layer of marine stratus/stratocumulus clouds that is present off the

west coast of South America. On the basis of satellite data and the limited number of measurements that have been made previously in the project area it is anticipated that clouds near the coast will be heavily influenced by the aerosol and SO₂ emissions from smelters, but that clouds further offshore will be more pristine in character, and our flights will be designed to sample this gradient in cloud properties. With an instrument package on the G-1 similar to the one carried during MASE, we should be able to fly as far as 500-700 km offshore and this should be sufficient to sample the desired gradient in cloud properties. The flight strategy will be similar to the successful G-1 strategy that was employed during MASE. In that program flights included a below-cloud leg to measure pre-cloud aerosol properties, in-cloud flights at several altitudes to examine the relationships between pre-cloud aerosol properties, cloud dynamics and cloud droplet microphysics, and their variation with altitude, and sampling through and above cloud top to define the vertical dimensions of the cloud, to document the thermodynamic structure of the atmosphere, and to sample the properties of the above-cloud aerosols.

We envision that these flights will be conducted both independently, and in conjunction with the Ron Brown, and the NCAR C-130. Although discussions with these other two platforms are only at a preliminary stage, we can envision joint flights with the C-130 wherein they would fly above cloud measuring cloud optical properties such as albedo, and cloud microphysical and dynamical properties using their onboard cloud radar, while the G-1 is measuring the in-cloud microphysical properties. Overflights of the Ron Brown will be useful because they are also planning to deploy a cloud-radar that will be useful for the detection of drizzle, as well as for measuring cloud dynamics. Thus we can imagine an experiment in which we sampled below, in and above a cloud-layer that was located above the Ron Brown. But as stated above, discussions regarding joint flights have just been initiated.

5. Proposed Instrumentation

Table 1 gives a list of instrumentation proposed for deployment on the G-1 during ASP/VOCALS beyond what is normally supplied as part of the aircraft. The latter includes atmospheric state parameters, winds from a 5-point gust probe, and aircraft velocity attitude, etc. The complement of instrumentation that will be added to the aircraft is similar to the one carried on the G-1 during the MASE project. It consists of instrumentation needed to characterize the chemical and microphysical properties of the aerosols, and the microphysical properties of clouds. Trace gas instrumentation is limited to instruments that measure aerosol precursors (SO_2), or gas phase species (CO , O_3 , DMS, and SO_2) that can aid us in estimating the anthropogenic contribution of aerosols and aerosol precursors in the air-mass that is being sampled.

Table 1 Proposed G-1 Instrumentation

Parameter	Instrument	Source
Aerosol Size distribution 0.1 – 3 μm	PCASP	PNNL
Aerosol size distribution 30 – 120 nm	FIMS	BNL
Aerosol concentration $d > 10$ nm	TSI 3010	PNNL
Aerosol concentration $d > 3$ nm	TSI 3025	PNNL
Aerosol composition Soluble inorganic and organic	PILS	BNL
Aerosol composition	Aerodyne ToF-AMS	BNL/PNNL
CCN Spectrum	DRI spectrometer	Hudson/DRI
CCN @ 2 fixed supersaturations	2xDMT CCN	BNL
TSI 3 integrating neph.	Aerosol scattering	PNNL
Aerosol absorption	Aethelometer, photothermal, or photoacoustic instrument	BNL?, PNNL? DRI?
O_3	Thermo Electron 49-100	BNL
CO	UV Fluorescence	BNL
SO_2	Thermo Electron 43S modified	BNL
DMS/Organics	PTRMS	PNNL
Cloud droplet and drizzle number size distribution	DMT CAPS	BNL
Cloud liquid water content	Gerber Probe/CAPS	PNNL/BNL

6. References

Albrecht, B. A., (1989), Aerosols, cloud microphysics and fractional cloudiness, *Science*, 245, 1227-1230.

Alexander et al (2005), Observations of enhanced organic aerosol concentrations above cloudtops in Marine Stratus Experiment off the Northern California Coast. *Eos Trans. AGU*, 86(52) Fall Meet. Suppl. Abstract A13B-0924.

Beheng, K. D. (1994): A parameterization of warm cloud microphysical conversion processes. *Atmos. Res.*, 33, 193-206

Daum et al (2006), Microphysical Properties of Stratus/stratocumulus Clouds During the 2005 Marine Stratus/Stratocumulus Experiment (MASE), *Eos Trans. AGU*, 87(52), Fall Meet. Suppl., Abstract A22A-04

Daum, P. H., et al., 1987: Chemistry and physics of a winter stratus cloud layer: a case study. *J. Geophys. Res.*, D92, 8426-8436.

Dusek, U., et al., *Science* **312**, 1375 (2006)

Hudson, J. G., and S. S. Yum, 1997: Droplet spectral broadening in marine stratus. *J. Atmos. Sci.*, 54, 2642-2654.

Kessler, E., 1969: On the distribution and continuity of water substance in atmospheric circulation. *Meteor. Monogr.*, **10**, No. 32, 84pp.

Kulkarni, P and J. Wang New fast integrated mobility spectrometer for real-time measurement of aerosol size distribution- I: Concept and theory *Aerosol Science* 37 (2006) 1303-1325)

Kulkarni, P and J. Wang New fast integrated mobility spectrometer for real-time measurement of aerosol size distribution- II: Design, calibration, and performance characterization *Aerosol Science* 37 (2006b) 1326-1339)

Leaith, W. R., C. M., Banic, G. A. Isaac, M. D. Couture, P. S. K. Liu, I. Gultepe, S. Liu, L. Kleinman, P. H. Daum, and J. I. MacPherson, 1996: Physical and chemical observations in marine stratus during the 1993 North Atlantic Regional Experiment. Factors controlling cloud droplet number concentration, *J. Geophys. Res.*, D101, 29123-29135.

Liu, Y., and P. H. Daum, 2000a: Spectral dispersion of cloud droplet size distributions and the parameterization of cloud droplet effective radius. *Geophys. Res. Lett.* 27, 1903-1906.

Liu, Y., and P. H. Daum, 2000b: Which size distribution function to use for studies related to effective radius. 13th International Conf. on Clouds and

Precipitation, Reno, USA.

Liu, Y., and P. H. Daum, 2002: Warm effect from dispersion forcing. *Nature*, 419, 580-581.

Liu, Y., and P. H. Daum, 2004: Parameterization of the autoconversion process. Part I: Analytical formulation of the Kessler-type parameterizations. *J. Atmos. Sci.*, 61, 1539-1548.

Liu, Y., P. H. Daum, and R. McGraw, 2004: A new analytical expression for predicting the critical radius in the autoconversion parameterization. *Geophys. Res. Lett.*, 31, L06121, doi:10.1029/2003GL019117.

Liu, Y., P. H. Daum, and R. McGraw, 2005: Size Truncation Effect, Threshold Behavior, and a New Type of Autoconversion Parameterization. *Geophys. Res. Lett.* 32, L11811, doi:10.1029/2005GL022636.

Liu, Y., P. H. Daum, R. McGraw, and R Wood, 2006a: Parameterization of the autoconversion process. Part II: Generalization of Sundqvist-type parameterizations. *J. Atmos. Sci.* 63, 1103-1109.

Liu, Y., P. H. Daum, and S. Yum, 2006b: An analytical expression for predicting relative dispersion of the droplet size distribution. *Geophys. Res. Lett.*, 33, 102810, doi:10.1029/2005GL024502.

Liu, Y., P. H. Daum, R. McGraw and M. Miller, 2006c: Generalized threshold function accounting for the effect of relative dispersion on threshold behavior of autoconversion process. *Geophys. Res. Lett.*, 33, L11804, doi:10.1029/2005GL025500.

Lohmann, U., and J. Feichter, 2001: Can the direct and semi-direct aerosol effect compete with the indirect effect on a global scale? *Geophys. Res. Lett.*, 28 (1), 159-161.

Martin, G. M., D. W. Johnson, and A. Spice, 1994: The measurement and parameterization of effective radius of droplets in the warm stratocumulus clouds. *J. Atmos., Sci.*, 51, 1823-1842.

McGraw, R. and Y. Liu, 2003: Kinetic potential and barrier crossings: A model for warm cloud drizzle formation. *Phys. Rev. Lett.*, 90(1), 018501-1 - 018501-4.

McGraw, R. and Y. Liu, 2004: Analytical formulation and parameterization of the kinetic potential theory for drizzle formation. *Phys. Rev. E.* 70, 031606-1-13.

McGraw, R. and Y. Liu, 2006: Brownian drift-diffusion model for evolution of droplet size distributions in turbulent clouds. *Geophys. Res. Lett.*, 33 103832, doi:10.1029/2005GL1023545.

Nenes, A. and Seinfeld, J. H., 2003: Parameterization of cloud droplet formation in global climate models, *J. Geophys. Res.*, 108, doi:1029/2002JD002911.

Novakov, T., 1994: The effects of anthropogenic sulfate aerosols on marine cloud droplet concentrations. *Tellus*, 46B, 132-141.

Peng, Y. R., and U. Lohmann, 2003: Sensitivity study of the spectral dispersion of the cloud droplet size distribution on the indirect aerosol effect. *Geophys. Res. Lett.*, 30 (10), Art. No. 1507.

Rotstajn, L. D., and Y. Liu, 2003: Sensitivity of the first indirect aerosol effect to an increase of cloud droplet spectral dispersion with droplet number concentration. *J. Climate*, 16, 3476-3481.

Rotstajn, L. D., and Y. Liu, 2005: A smaller global estimate of the second indirect aerosol effect. *Geophys. Res. Lett.*, 32, L05708-1-4.

Twomey, S. (1974), Pollution and the planetary albedo, *Atmos. Environ.*, 8, 1251-1256

Wood, R., Parameterization of the effect of drizzle upon the droplet effective radius in stratocumulus clouds, *Quart. J. Royal Meteor. Soc.*, 126, 3309-3324 (2000).

7. Budget