

Pre-Cloud Aerosol, Cloud Droplet Concentration, and Cloud Condensation Nuclei from the VAMOS Ocean-Cloud-Atmosphere Land Study (VOCALS) Field Campaign

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Contents

1 Introduction	1
2 Data Files	2
3 Results	4
3.1 Aerosol composition and size distribution	
3.2 Sub-cloud aerosol and comparison with cloud droplet number concentration	
3.3 Aerosol activation	
4. References	8

1. Introduction

During October and November, 2008, Brookhaven National Laboratory (BNL) participated in VOCALS (VAMOS Ocean-Cloud-Atmosphere Land Study), a multi agency, multi-national atmospheric sampling field campaign conducted over the Pacific Ocean off the coast of Arica, Chile. Support for BNL came from DOE's Atmospheric Science Program (ASP) which is now part of the Atmospheric System Research (ASR) program following a merger with DOE's Atmospheric Radiation (ARM) program. A description of the VOCALS field campaign can be found at <http://www.eol.ucar.edu/projects/vocals/>.

Measurements made from the DOE G-1 aircraft are being used to assess the effects of anthropogenic and biogenic aerosol on the microphysics of marine stratus. Aerosols affect the size and lifetime of cloud droplets thereby influencing the earth's climate by making clouds more or less reflective and more or less long-lived. Climatic impacts resulting from interactions between aerosols and clouds have been identified by the IPCC (2007) as being highly uncertain and it is toward the improved representation of these processes in climate models that BNL's efforts are directed.

In this, the first of a series of Program Metric Reports, we 1) describe archived data from the DOE G-1 aircraft, 2) illustrate several relations between sub-cloud aerosol, CCN, and cloud droplets pertinent to determining the effects of pollutant sources on cloud properties, and 3) post to the data archive an Excel spreadsheet that contains cloud and corresponding sub-cloud data. The Excel spreadsheet, [Cloud_Subcloud_Comparison.xls](#), contains the data that satisfies the **1st Quarter Metric**: "Tabulate and make available a set of VAMOS Ocean-Cloud-Atmospheric Land Study showing pre-cloud aerosol size distributions, cloud condensation nuclei (CCN) number concentrations, and cloud droplet distributions." The Excel spreadsheet is derived from data in the main archive but differs in that an association has been made between pre-cloud and in-cloud observations.

2. Data Files

The parent data set from which the Excel spreadsheet has been derived is archived at: ftp://ftp.asd.bnl.gov/pub/ASP%20Field%20Programs/2008VOCALS/Processed_Data

Table 1. Archived data.

Data	Link	Last Modified
AMS and PILS	Aerosol_Comp	5/20/2009
Cloud droplet spectra	CASPart	5/20/2009
Drizzle spectra	CIPPart	5/20/2009
Aerosol spectra	DMAPart	7/8/2009
Data summaries and graphs	Data_PDF_Files	5/21/2009
Chemistry data set & CCN	General	5/21/2009
CCN: Stable conditions	CCN	12/17/2009
PCASP A spectra	PCASPPart	5/21/2009

PCASP B spectra	PCASP_BPart	5/21/2009
Position information	PositionPart	5/21/2009

Data are archived as ASCII files. A file contains data from one or more instruments for a single flight. The first 39 rows of each file contain meta data including heading, units, contact information, and comments. Flights are identified by yymmddL where L=a, b, or c for the 1st, 2nd, or 3rd flight in a day. In VOCALS, there were no instances of multiple flights in a day, so L=a. All measurements are referenced to a common one second time base. The 1st column of each file contains Date in UTC, the 2nd column Time in UTC. Data acquisition rates vary by instrument. In the case of the DMA, PILS, and AMS with acquisition times longer than 1s, the instrument reading is repeated at 1s intervals from the start of a data acquisition period to the end. Each measurement is provided on a 1 second time base indicated by a file name that ends in _1.txt and averaged to a 10s time base indicated by _10.txt. For both times bases, “Time” is always the beginning of a measurement interval. Typical flight names are 081017a_1.txt and 081017a_10.txt for measurements made on the first (and only) flight of Oct. 17, 2008, put on a 1 or 10s time base. What is in these files is given by the name of the folder that they came from and by the meta data in the files. Files that contain data from instruments that give a size distribution (e.g. CASPart, CIPPart, DMAPart, PCASPPart, and PCASP_BPart) will have a series of columns giving particle number concentration for each size bin, followed by columns that give ancillary information such as total particle concentration, surface area, and volume. Files in the folders Aerosol_Comp, General, PositionPart contain measurements from multiple instruments. CCN data can be found in two places. The “General” files contain a complete record, while the CCN file picks out time periods in which the instruments were run under stable conditions. The use of a common time base facilitates combining data from different files.

Although specifically addressed in the meta data, past experience is that there are two points of confusion. First, all aerosol measurements are presented at ambient conditions of temperature and pressure. Second, AMS concentrations are given at a collection efficiency of one.

Table 2 indicates data availability on 17 flights. All results presented below are obtained from the archived data. Data from flight 081018a were not used because of problems with research power.

INSTRUMENT GUIDE FOR VOCALS 2008

File: Instrument Guide.xls
 Contact: Stephen R. Springston srs@bnl.gov

Created: October 6, 2008
 Last Update: May 14, 2009

This table is a summary of all measurements made aboard the DOE G-1 Research Aircraft Facility (RAF) during VOCALS 2008, a part of the VAMOS field campaign. The anonymous ftp site: <ftp.asd.bnl.gov/pub/ASP%20Filed%20Programs/2008VOCALS> is the primary source for data from the RAF. This table is only a guide and the actual files for each flight should be consulted for further details. The latest revision history is provided in the Read Me.txt file on the ftp site.

[Overview of G-1 Flight Maps and I Guide:](#)
[Summary of the Final-Form Flight Data Files](#)

■ Valid measurements taken, data in ASP ftp site
■ Measurement validity suspect, being reviewed or data not yet merged in ASI
■ Valid measurements not available (and not expected in future)

Flight Designation (yyymmdd(flight of day letter))	081014a	081017a	081018a	081022a	081023a	081025a	081026a	081028a	081029a	081101a	081103a	081104a	081106a	081108a	081110a	081112a	081113a
Date Take Off (UTC, yyyy-mm-dd)	10/14/08	10/17/08	10/18/08	10/22/08	10/23/08	10/25/08	10/26/08	10/28/08	10/29/08	11/1/08	11/3/08	11/4/08	11/6/08	11/8/08	11/10/08	11/12/08	11/13/08
Time Take Off (UTC, hh:mm:ss)	15:52:30	13:00:13	13:07:06	16:32:29	12:49:15	13:03:36	13:01:21	12:58:43	15:58:05	12:57:08	12:58:25	11:57:30	11:57:32	12:56:51	13:02:17	13:20:22	12:54:17
Time Landing (UTC, hh:mm:ss)	18:53:40	16:50:14	15:16:13	19:57:40	16:35:08	17:07:39	16:36:43	17:18:04	19:33:08	16:57:59	16:51:40	16:02:35	16:21:05	16:31:51	16:50:05	16:55:07	16:42:00
Channel	Power failure																
Latitude, Longitude, Altitude	TANS-Vector																
Latitude, Longitude, Altitude	DSM																
Ambient Temperature	Total Temperature Probe																
Potential Temperature	Calculated																
Static Pressure	Pressure Port																
Pressure Altitude	Calculated																
Dew Point	Chilled Mirror Hygrometer																
Ambient Temperature	Reverse Flow Probe																
Infrared Temperature	PRT-5																
Relative Humidity, H ₂ O Mixing Ratio	Calculated																
H ₂ O Mixing Ratio	TDLAS																
Wind Speed, Direction	Calculated (vector subtraction)																
Turbulence Dissipation	Gust Probe																
UV sky	Eppley Pyranometer																
UV and	Eppley Pyranometer																
Liquid Water Content	Gerber Probe																
3-Axis Acceleration	BNL Accelerometer																
N (30 bins, 0.1 - 3 um) nose	PCASP X100 w/ SPP 200 electronics																
N (30 bins, 0.1 - 3 um) cabin	PCASP X100 w/ SPP 200 electronics																
N (20 bins, 0.6 - 60 um)	CAPS Probe (CAS section)																
N (60 bins, 25 - 1550 um)	CAPS Probe (CIP section)																
GASEOUS SYSTEMS Forward Facing Inlet																	
O ₃	TEI Model 49-100																
O ₃	BNL Basic Fast Ozone Monitor																
CO	BNL VUV Fluorescence Analyzer																
SO ₂	TEI Model 43S																
Organic Compounds	Proton Transfer Mass Spectrometer																
PARTICLE SYSTEMS Isokinetic Aerosol Inlet																	
Aerosol Composition	Inlet Control																
Aerosol Composition	AMS																
Aerosol Composition	PILS																
Aerosol Absorbance	PTI																
Aerosol Composition	PSAP																
Particle scattering	Nephelometer (s _{sp})																
Condensation nuclei	DMT single CCN																
Condensation nuclei	DMT dual CCN																
N (21 bins, 16 - 444 nm)	TSEMS																
Aerosol Composition	TRAC																
PARTICLE SYSTEMS Reverse Facing Inlet(s)																	
N D _p >2.5 nm	CPC/TSI 3025																
N D _p >10 nm	CPC/TSI 3010																
N (35 - 110 nm)	FIMS																

*Test flights only

Table 2 Data availability

Data from Table 1 were used to create an Excel spreadsheet [Cloud_Subcloud_Comparison.xls](#) containing cloud droplet and sub (i.e., pre) - cloud aerosol concentration in a form useful for examining the fraction of aerosol that gets activated and thereby contributes to the climatically important first aerosol indirect effect. This file has been archived and is described in Section 3.2, below.

3. Results

An objective of BNL's participation in VOCALS is to understand the effect that sub-cloud aerosol has on cloud droplet number concentration (CDNC). Activation depends on aerosol composition and size distribution. Cloud dynamics is the other important variable, but this report stops short of that consideration.

3.1 Aerosol composition and size distribution

As an introduction to the VOCALS aerosol data set, composition and size distributions are presented in Figs. 1 and 2. Data for these figures are obtained from [Aerosol_Comp](#) and [DMAPart](#), respectively, and encompasses all of the below-cloud-level portions of 16 flights.

The chemical composition of sub micrometer aerosol measured below cloud level was remarkably uniform as a function of longitude as indicated by AMS data presented in Fig. 1. Within a longitude grouping there are only small fluctuations (not shown) around an average composition of 25% neutralized H_2SO_4 ($H_{1.5}(NH_4)_{0.5}SO_4$) plus a 10% admixture of organics. The amount and longitudinal gradient of aerosol sulfate, and a consideration of the locations of Cu smelters and power plants in Chile, strongly suggest that the sub micron aerosol is dominated by anthropogenic emissions.

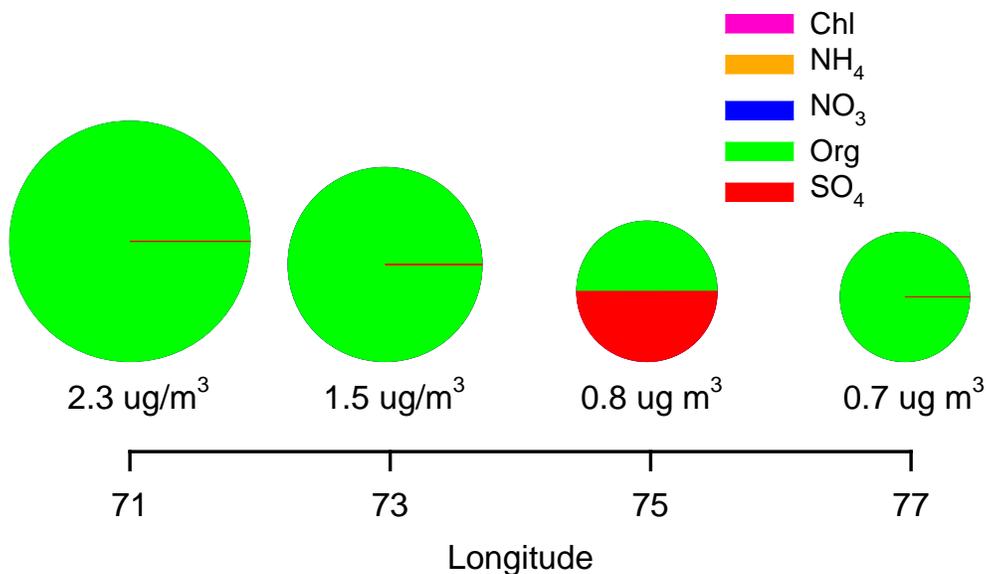


Figure 1. Below-cloud-level aerosol composition determined from AMS measurements averaged into 2 degree longitude bins. Aerosol particles measured by the AMS have a maximum diameter of $\sim 0.5 \mu\text{m}$. Collection efficiency for the AMS equals one. Within the size range of the AMS, chloride concentrations were very low, and are not resolved in the pie diagrams. Sea salt and nitrate makes a significant contribution to larger particles as indicated by data from the PILS (Particle into Liquid Sampler) which measured the composition of particles smaller than $\sim 2.5 \mu\text{m}$ (Lee et al., 2009).

Aerosol size distributions shown in Fig. 2 have a two mode structure characteristic of marine aerosol. This structure has been shown to develop because of in-cloud chemistry forming aerosol sulfate from gas phase precursors (Hoppel et al., 1986). A fraction of aerosol are activated to form cloud droplets. The aerosol that is formed upon cloud droplet evaporation is larger than the initial aerosol because of mass gained by in-cloud chemistry. Thus the accumulation mode centered at $\sim 170 \text{ nm}$ represents aerosol that has been activated at some point its history, while the Aitken size particles centered at $40 - 60 \text{ nm}$ represent aerosol that has not been activated.

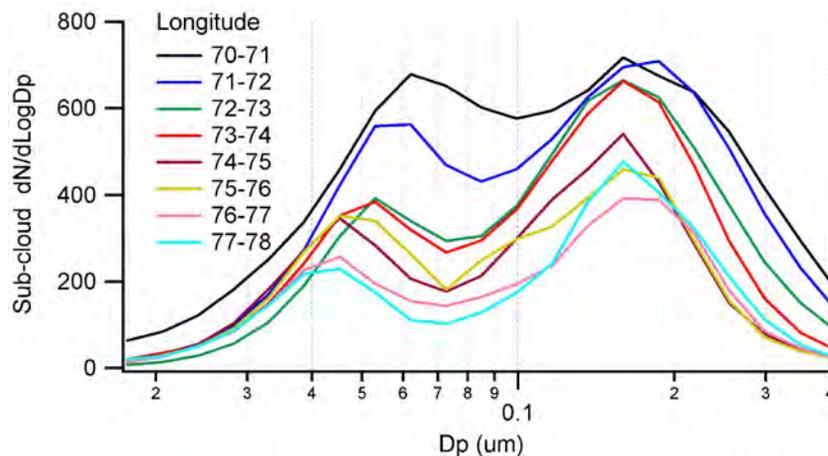


Figure 2 Size distribution of below-cloud-level aerosol as a function of longitude, which is very nearly proportional to distance from the Chilean coast.

3.2 *Sub-cloud aerosol and comparison with cloud droplet number concentration*

Our approach is to identify a time period with unbroken cloud and compare the number concentration of cloud droplets to the number concentration of aerosol particles below the cloud in air that will eventually be circulated into the cloud. In the VOCALS environment, below cloud aerosol concentrations are relatively homogeneous on horizontal spatial scales of 10s of km, through the depth of the boundary layer, so an exact equivalence between a cloud location and pre-cloud aerosol is not required. In so far as there are inhomogenities in the spatial distribution of aerosol, these will be greatest near the coastline.

Because we are interested in the size distributions of particles smaller than 100 nm we rely on the DMA (size range $15 - 440 \text{ nm}$) and consider average values over the

nominal 1 minute time period for the DMA to complete a size scan. Having identified a cloud penetration (criteria that liquid water content $> 0.05 \text{ g/m}^3$) we identify below cloud DMA scans that satisfy screening criteria including the specification that no 1s period contains liquid water greater than 0.01 g/m^3 and that the below cloud point be within 15 km of the cloud point. In order to eliminate clouds with holes, cloud droplet concentrations are calculated for that fraction of the 1 minute DMA scan that has 1 second LWC (liquid water content) $> 0.05 \text{ g/m}^3$. This procedure yielded 543 in-cloud data points (i.e. almost 8 hours of data) in which a cloud and corresponding sub-cloud DMA scan could be identified. In the usual case that more than one sub-cloud point corresponded to a cloud penetration, results were averaged over the several below cloud points.

The Excel spreadsheet [Cloud_Subcloud_Comparison.xls](#) added to the data archive as part of this report contains the time and position of the in-cloud DMA scans. On the cloud side, LWC, CDNC and a size spectrum ($dN/d\text{Log}D_p$) of cloud droplets are provided. For the corresponding sub-cloud aerosol, number concentration, size spectra ($dN/d\text{Log}D_p$), and CCN at 0.2% supersaturation are given.

Cloud droplet number concentration as a function of pre-cloud aerosol are shown in Fig. 3. Results are within the range of values obtained at different locations as depicted in Fig. 5 of Ramanathan et al (2001). The common tendency is for CDNC to increase linearly with sub cloud aerosol (N_a) at low aerosol concentration and then for a less steep increase at higher values of N_a . The 2 sets of outliers with CDNC below the main groupings come from measurements where the aerosol was unusually non-homogeneous.

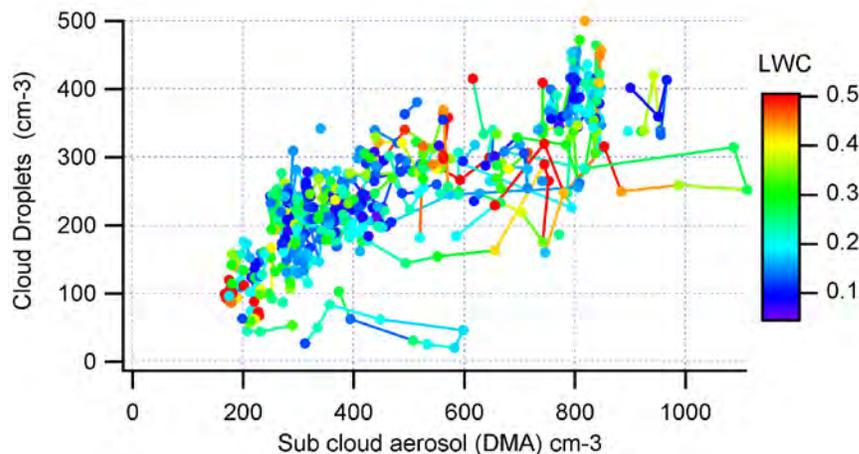


Figure 3. Comparison of cloud droplet number concentration with sub-cloud aerosol for the entire VOCALS campaign. Data points that are connected are sequential in time. Breaks appear because aircraft flight patterns consisted of transects and vertical profiles in and out of cloud.

If the only independent variable in our data set is sub-cloud aerosol concentration (i.e. meteorological variability can be ignored) then Figure 3 provides an important result on how clouds respond to a perturbation in anthropogenic aerosol, which with known

liquid water path can be used to determine changes in cloud albedo. The color coding by LWC, qualitatively suggests that in this case the relation between aerosol and cloud droplets is not strongly affected by LWC.

3.3 Aerosol activation

Figure 4 shows the number concentration of aerosol activated at $\sim 0.2\%$ supersaturation as a function of the total aerosol number concentration.

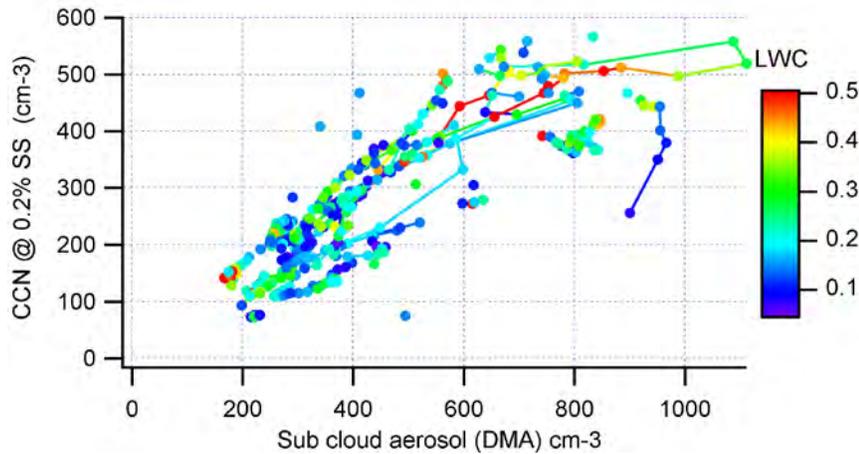


Figure 4. Number concentration of aerosols activated at a nominal supersaturation of 0.2% versus the total aerosol number concentration measured by DMA.

Aerosols with a composition illustrated in Fig. 1 are expected to have a CCN forming potential comparable to $(\text{NH}_4)_2\text{SO}_4$, which is among the most easily activated, atmospherically relevant aerosol. An $(\text{NH}_4)_2\text{SO}_4$ particle with a dry diameter of 90 nm activates at a supersaturation of 0.2% (Seinfeld and Pandis, 1998). We have compared the CCN measurements at 0.2% supersaturation with DMA spectra to determine a “critical diameter” above which 100% activation accounts for the CCN observations. For the data points in Fig. 4, we obtain an average critical diameter of 97 nm (median = 91 nm) with a standard deviation of 44 nm, very close to that predicted for $(\text{NH}_4)_2\text{SO}_4$, albeit with considerable scatter. The scatter is probably not due to composition differences. In contrast to other locations where aerosols have large and variable insoluble or sparingly soluble components (Wang et al., 2008), aerosol chemical composition in VOCALS is relatively constant (Fig. 1).

Given that a Hoppel minimum is formed by preferential activation of large particles, it is relevant that the size distributions in Fig. 2 show a separation between accumulation mode and the smaller, less easily activated Aitken mode between 70 and 100 nm.

If CDNC data from Fig. 3 were added to Fig. 4, one would see that CDNC is generally less than CCN at 0.2% supersaturation. This could be because the controlling supersaturation is less than 0.2% or it could have a cloud dynamic explanation. Both possibilities are being investigated.

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