Second Year Report: Sea-salt Aerosol in Hawaii and the Influence of Large Organized Structures (LOS) or Rolls on Fluxes and Visibility

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LONG-TERM GOAL

Our long-term goal is to establish an improved understanding of the factors that control the marine aerosol properties and concentrations as they relate to generation processes, mixing processes, their dependence on oceanic and environmental conditions and physicochemical evolution in the marine boundary layer. We expect these efforts to lead to improved predictability of marine aerosol concentrations and optics.

OBJECTIVES

Our recent ONR efforts characterized sea-salt aerosol (SSA) size distributions and production from breaking waves through measurements in the "natural wind tunnel" between the Big Island of Hawaii and Maui. Here SSA production can be studied under accelerating wind conditions that also control its vertical mixing. These experiments revealed the typical presence of Large Organized Structures (LOS) or rolls aligned along the wind in the channel both with and without visible cloud streets. This organized mixing process can include entrainments of air to/from the buffer layer (BuL), the free troposphere (FT) and the mixed boundary layer (BL) that can directly influence aerosol concentrations, fluxes and visibility. Our activities this past year include aircraft studies to investigate these LOS and their role in aerosol processes including vertical mixing of sea-salt and associated optical extinction.

In 2007 we deployed our aerosol instrumentation during PASE (Pacific Atmospheric Sulfur Experiment) mission on flights in the marine boundary layer near Christmas Island to examine aerosol dynamics and its relation to entrainment, cloud processes and SSA. This experiment was prompted by the studies of the sulfur cycle and aerosol dynamics made at Christmas Is. a decade ago under ONR support and published in GRL [*Clarke et al.*, 1996]. PASE was completed in September 2007 and included a channel flight near Hawaii to study SSA aerosol production in the Alenuihaha channel and numerous flights near equator (Christmas Island) with and without MBL rolls structures.

APPROACH

We focus our ONR efforts upon characterizing the marine aerosol full size distributions, the associated source function and optical properties [*Clarke et al.*, 2003; *Clarke et al.*, 2006; *Clarke and Kapustin*,

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14. ABSTRACT
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2003; *Kapustin et al.*, 2006; *Shinozuka et al.*, 2004]. Our instrumentation includes total and submicrometer nephelometers, particle size spectrometers (DMA, LDMA, OPC, APS), Hot/cold CN, size-resolved volatility (DMA and OPC) and Time of Flight Aerosol Mass Spectrometry (ToF-AMS). The AMS was obtained through an ONR supported DURIP proposal for marine aerosol studies and was deployed for the first time by our group on the NCAR C-130 for the PASE mission.

WORK COMPLETED THIS YEAR

Our ONR related efforts this year include analysis of aerosol data measured on the NCAR C-130 as part of PASE. We examined our PASE aircraft data and found numerous events of flying through rolls in the marine boundary layer (MBL). This major aircraft study benefits from the full complement of the aerosol measurements listed above along with the fast 25 Hz gas phase and meteorological data measurements (including 3D wind velocities) and is valuable for pursuing our ONR objectives. We are also analyzing rolls encountered over the Gulf of Mexico during the 2006 Milagro experiment.

Some examples of the rolls and their link to MBL processes, aerosol concentrations and fluxes are illustrated in the figures below. These are being examined for presentation at the PASE 2008 Workshop, AGU meeting in San Francisco 2008 and as a core of our papers in preparation.

A. Rolls or Large Organized Structures (LOS) as a common feature of tropical / subtropical MBL environment. LOS observations during PASE

Large Organized Structures (or horizontal convective rolls) are evident in almost all flow regimes around Hawaii. Even cloud-free regions in satellite imagery include indications of these rolls organized along the wind in the channel as evident in majority of observations in our lidar (ceilometer) data (see our ONR annual reports 2006-2007). The typical rolls wavelength related to the depth **H** of the MBL is ~ 2.8***H**

During PASE (NCAR C-130) campaign we encountered numerous periodic structures related to horizontal convective rolls (HCRs) on our flight paths. HCRs commonly form over the ocean surface when some vertical wind shear is present. They are counterrotating pairs of helices oriented horizontally within the boundary layer and tend to be aligned parallel to main flow. Although HCRs have been clearly seen in satellite photographs as cloud streets for the past 30 years, their development and evolution is poorly understood due to lack of direct observational data. HCRs are significant to the vertical transport of momentum, heat, moisture, aerosol and gases within the boundary layer.

Figure 1 show GOES-11 images overlaid with PASE flights 14 and 8 flight tracks (yellow and red lines). Intense cloud streets (rolls) are clearly visible for flight 14 (top) and almost don’t exist in flight 8 track region (bottom).

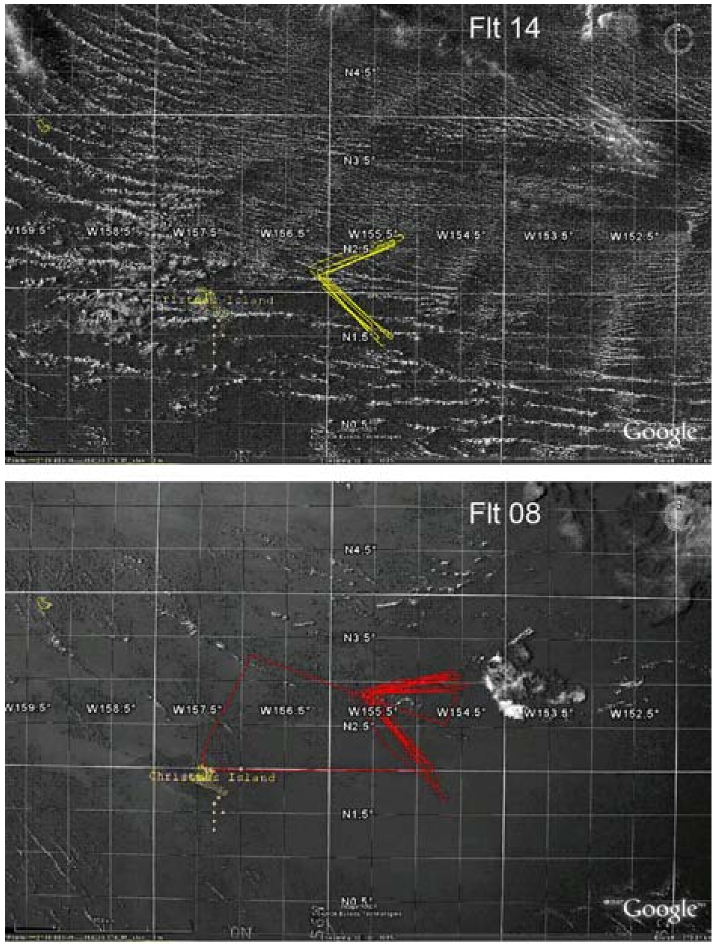

Figure 1. GOES-11 images overlaid with PASE flights 14 and 8 flight tracks (yellow and red) showing intense cloud streets (rolls) for flight 14 (top) and almost no LOS for flight 8 (bottom).

State of the art instruments were used during PASE to make accurate and high speed airborne determinations of SO2, DMS, DMSO, DMSO2, MSA, H2SO4, OH, HO2, NH3, H2O, O3, H2O2, liquid water, temperature, pressure and wind velocity. DMS, SO2, H2O and O3 were determined at 25 samples per second allowing eddy correlation determination of vertical fluxes of these species [Faloona et al., 2005]. We are also exploring the possibility of using our 1Hz aerosol data (condensation nuclei (CN), light scattering) to determine an aerosol vertical fluxes by eddy correlation.

Much of the PASE was conducted in the convective boundary layer (CBL) of the marine atmosphere east of Christmas Island (Kiritimati) during August-September 2007. The overall flight patterns used during PASE were determined by the primary goal of investigating the chemistry and physics (primarily of sulfur) in a cloud free CBL. Because the characterization of CBL turbulence structure was an integral part of this goal, numerous turbulence stacks and vertical profiles were made (see **figures 1 and 2**). Nevertheless, some of the flight legs were not ideally situated for the study of roll circulation influence on MBL aerosol parameters. For example, consistent Top of a Buffer Layer (BuL) / Free Troposphere stacks were practically missing.

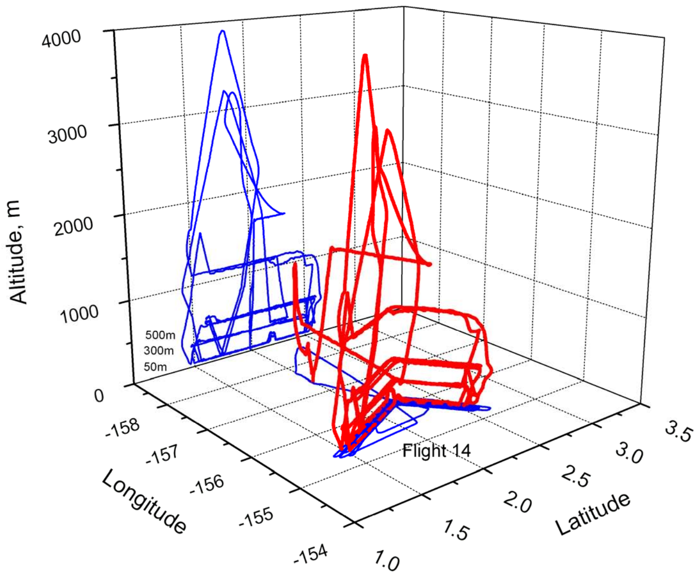

Figure 2. Three-dimensional display of a typical C-130 flight pattern during PASE field experiment (flight 14).

The examples included in this report consist of the flights made in the presence and absence of roll structures. The typical flight includes 3-4 sets of L-shaped horizontal flight legs across and along the mean wind and stacked vertically. The lowest level run was usually made at an altitude of 50m. Other levels were chosen with reference to vertical profiles made prior and after horizontal runs, typically in the subcloud layer (300m - see **figure 2**), near cloud base (500m) and near a trade wind inversion (TWI) (~1100m on **figure 2**).

B. ***Power spectra and scales of the rolls; Stability parameters; Mean boundary layer conditions, aerosol and roll characteristics.***

Our identification of large-scale roll structures involved the following stages:

a. 30min time resolution sets of 1km_ch1_vis GOES-11 imagery were used for direct observation of cloud streets (**figure 1**).

b. The power spectra of time series of downwelling IR radiation (IRTC) were used to clearly define the presence and scales of the rolls (cloud streets) (**figure 3a**). Frequencies **f1** and **f2** on figure 3a mark the roll-scale wavelength range. Frequency **f** can be converted to wavelength **λ** by **λ = c/f** with the aircraft speed during horizontal legs **c** ~ 100 m/s. Maximum is corresponding to a cloud streets scale range of approximately 2-4km. (after correction for angle of attack – see figure 1, top)

c. The cospectra of Vertical Wind component (WIC) with IRTC, CN concentration (CNCold and CNHot) or other meteorological and aerosol parameters were useful to identify possible roll cases (**figure 3b**) and to show significance of different parameters in vertical transport (**figure 3c**) .

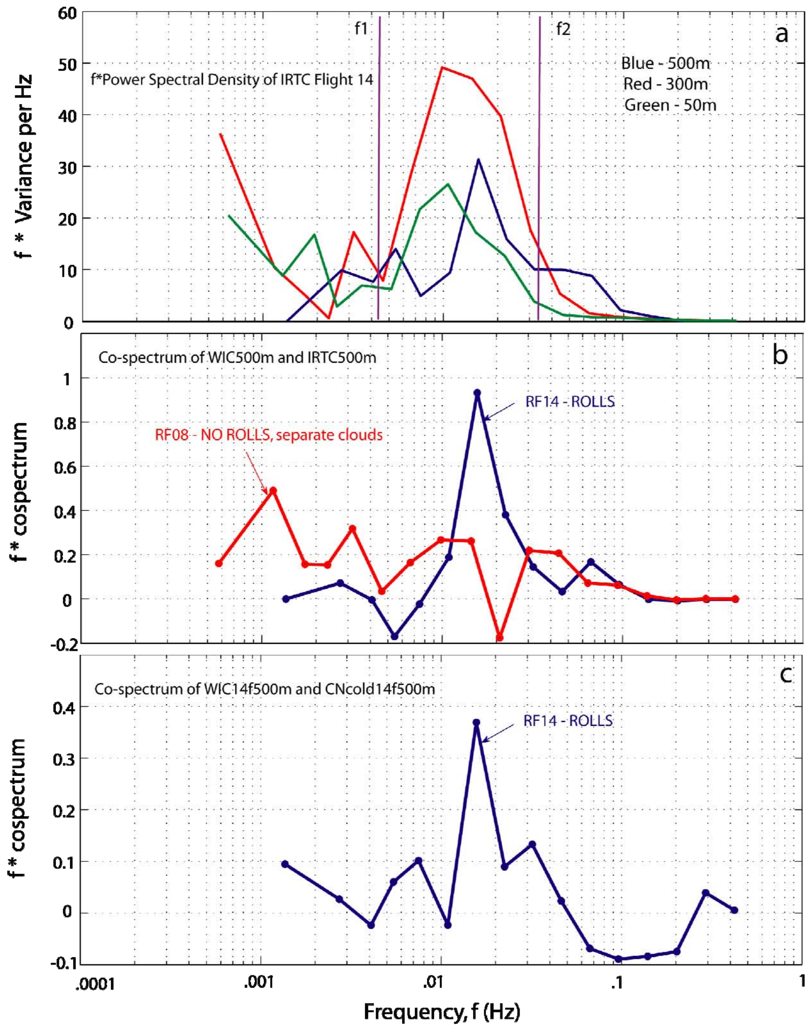

Figure 3 (a) Variance spectra of IR radiation weighted by frequency f and calculated from time series measured during 3 flight legs within a field of roll convection. Frequencies f1 and f2 mark the roll-scale wavelength range. Frequency f can be converted to wavelength λ by λ = c/f with the aircraft speed during horizontal legs c ~ 100 m/s. Maximum is corresponding to a scale range of approximately 7-10km; (b) Cospectra of Vertical Wind component (WIC@500m) with Top Infrared Irradiance (IRTC@500m) for flights 8 (red - no rolls) and 14 (blue - rolls); (c) Cospectra of Vertical Wind component (WIC@500m) with CNCold concentration (CNCold@500m) for flight 14 (blue - rolls).

Flight 14 which was identified as a case with a roll pattern shows high correlation of different meteorological, aerosol and gas parameters like vertical wind velocity (WIC), humidity mixing ratio (MRLA), CNHot (CN@350C) concentration – an indicator of refractory sea-salt number (**figure 4a**); or horizontal wind component (UIC), MRLA and CNCold (CN@40C) concentration (**figure 4b**).

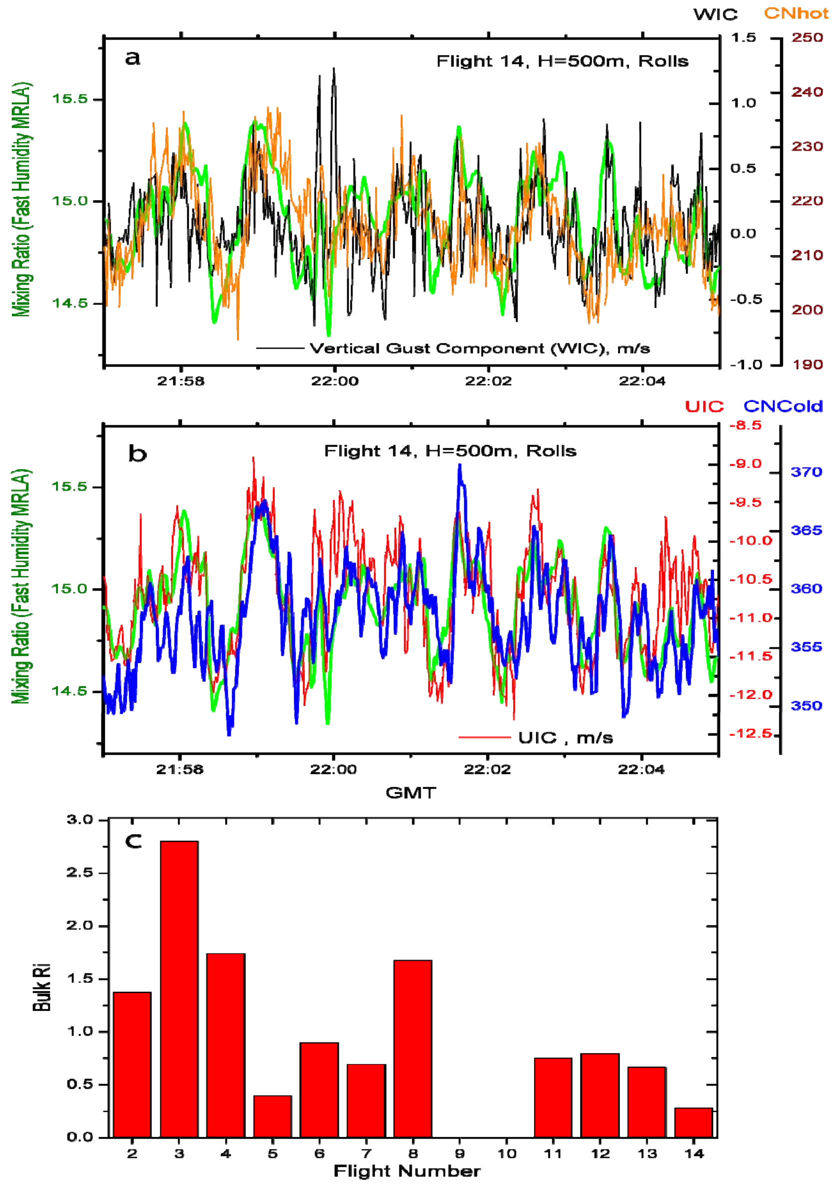

Figure 3. (a) Time series of vertical wind velocity (WIC), humidity mixing ratio (MRLA) and CNHot (CN@350C) concentration for rolls encountered on flight 14, H=500m; (b) same, but for one of the horizontal wind components (UIC), MRLA and CNCold (CN@40C) concentration; (c) Bulk Richardson number [Brümmer (1999] for PASE flights. Lowest Ri corresponds to a well developed roll structure (flight 14).

Ri = (g/Θv)*(ΔΘv*H/Ug^2); ΔΘv is the difference between values of virtual potential temperature at the top and bottom of the roll layer; Θv is the mean value across the full depth of the roll layer H; Ug is the wind just above the roll layer.

To identify rolls presence we can also use a stability parameter - bulk Richardson number as shown on **Figure 4c** for some PASE flights. Lowest Ri corresponds to a well developed roll structure (flight 14) [Brooks and Rogers, 1997]. Here we were using bulk Ri definition of Brümmer (1999): Ri = (g/Θv)*(ΔΘv*H/Ug^2) where ΔΘv is the difference between values of virtual potential temperature at the top and bottom of the roll layer; Θv is the mean value across the full depth of the roll layer H; Ug is the wind just above the roll layer.

Due to well mixed nature of BL, vertical profiles normalization by mean BL value of profiled parameter (Potential Temperature, DMS, Aerosol Concentration etc.), will simplify case to case profiles comparison.

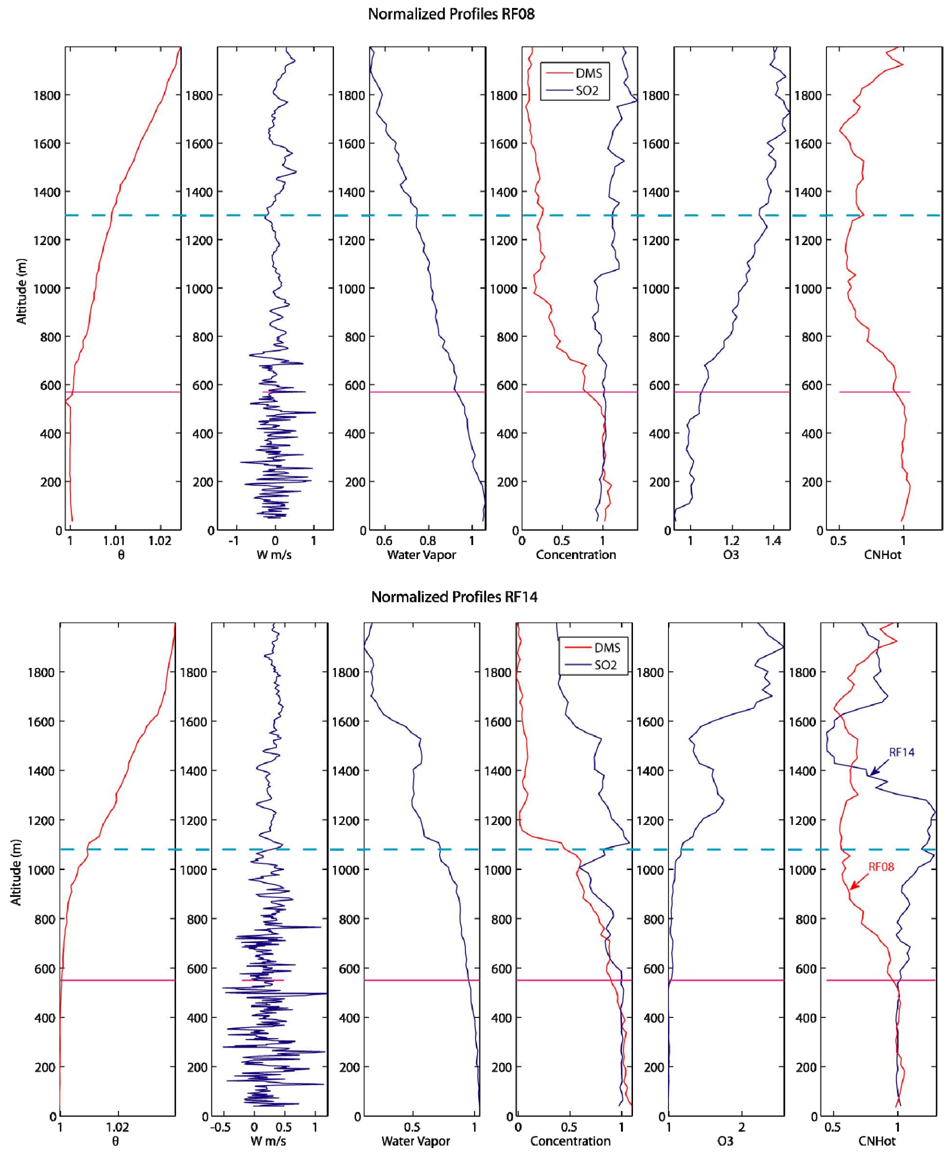

Figure 5. (top) Flight 8 (no rolls) Normalized profiles of Potential Temperature, Vertical Wind Speed (not normalized), Water Vapor, DMS, SO2, O3 and Hot CN concentrations. Cyan line – top of BuL, Magenta line- top of BL; (bottom)- same but for flight 14 (rolls).

Figure 5 shows the normalized (by mean BL value) profiles of Potential Temperature, Water Vapor, DMS, SO2, O3 and Hot CN concentrations for flights 8 (top figure – no rolls case) and 14 (bottom figure – strong rolls case). Vertical Wind Speed profiles are not normalized. Cyan lines on figures represent a top of Buffer layer (BuL) (trade wind inversion – TWI), magenta lines show a top of Boundary Layer (BL). An obvious difference between flight 8 and 14 is that for flight 14 both BL and BuL are well mixed with enhanced vertical wind speed in BuL. Also flight 14 shows larger CNhot in

BuL compared to BL due to entrainment from FT, where Amazon biomass burning plume near TWI was present (compare blue (flight 14) and red (flight 8) lines for CNhot concentration on **figure 5** (bottom, right)).

C. Aerosol vertical distribution and aerosol fluxes in the presence of rolls

Figure 6a represents another example of strong differences between aerosol parameters for flights with (flt14) and without (flt8) rolls. Normalized (by mean BL value) profiles of CNCold for Flights 8 (red) and 14 (blue) are similar in BL where we have strong mixing for both flights. In BuL aerosol concentration is steadily decreasing with altitude for Flight 8 which is indicative of less intense mixing from below and entrainment of cleaner air from FT. For Flight 14, normalized CNCold concentration increases steadily in BuL, which is indicative of both enhanced mixing from below and entrainment of biomass burning aerosol from FT. As before, cyan line represents a top of BuL (TWI), magenta line is a top of BL (Zi).

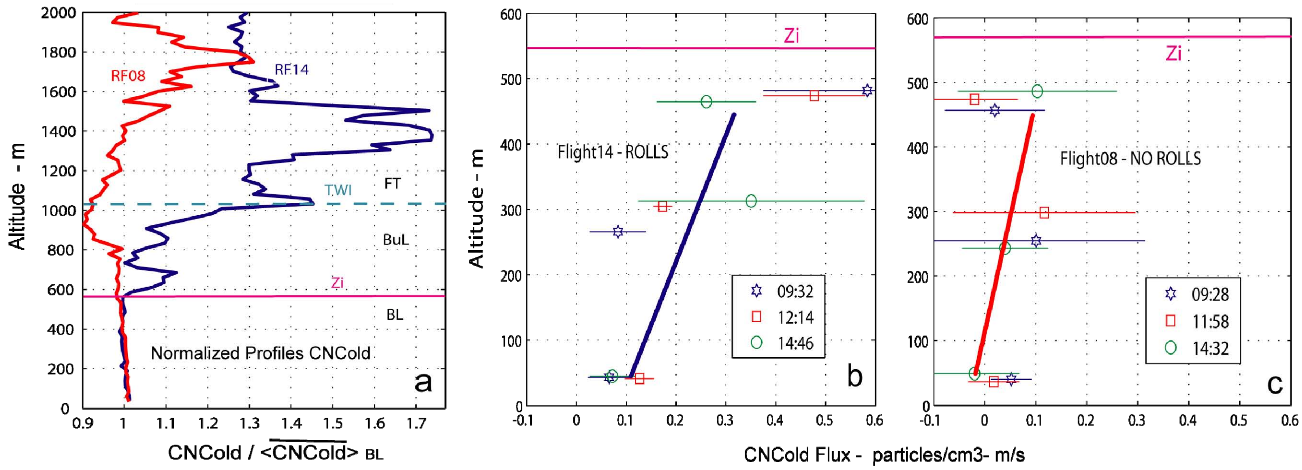

Figure 6. (a) Normalized profiles of CNCold for Flights 8 (red - no rolls) and 14 (blue – rolls) Cyan line – top of BuL (TWI) , Magenta line- top of BL (Zi); (b)-CNCold flux for flight 14. Symbols - the mean CNCold fluxes measured on each horizontal leg (see figure 2) along with the standard deviation of the mean. Solid blue line is the weighted least squares fit through the Flight 14 fluxes at all 3 altitude levels; (c) Same as b, but for Flight 8. Solid red line is the weighted least squares fit through the Flight 8 fluxes at all 3 altitude levels

Figure 6b,c show the observed vertical fluxes of CNCold concentration. Symbols are the mean CNCold fluxes measured on each horizontal leg (see **figures 1 and 2**) along with the standard deviation of the mean. Solid blue line is the weighted least squares fit through the Flight 14 fluxes at all three altitude levels; solid red line is the weighted least squares fit through the Flight 8 fluxes. The detailed study of aerosol fluxes and entrainment rates is currently under way and will be a subject of next year project activity.

RESULTS

Roll structures investigated in diverse MBL settings demonstrate that these can play an active role in the redistribution of aerosol, gas and water vapor in the MBL compared to non-roll cases. Depending upon the thermodynamic profiles, roll size, altitude and temporal duration, these LOS can have a marked effect on the exchange of air masses between the BuL, BL and FT. This will lead to changes in the horizontal extinction in these layers relative to regions not influenced by these LOS rolls. Hence, the evolution of aerosol optical properties in the near surface mixed layer will be affected by rolls and the conditions that stimulate them. These can occur with or without associated cloud features.

We are actively studying LOS influence on aerosol evolution in the MBL and will be presenting our studies at the fall AGU in San Francisco. Some ongoing issues include the following: what are the aerosol optics differences in updraft and downdraft caused by RH, aerosol concentration and entrainment; what are implications of roll structure for extinction measurements and remote sensing; what are implications of LOS on the redistributions of aerosol in MBL?

The results of our ongoing analysis will be the foundation for related papers to be written in 2008-2009.

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PUBLICATIONS

No new publications in 2008.