

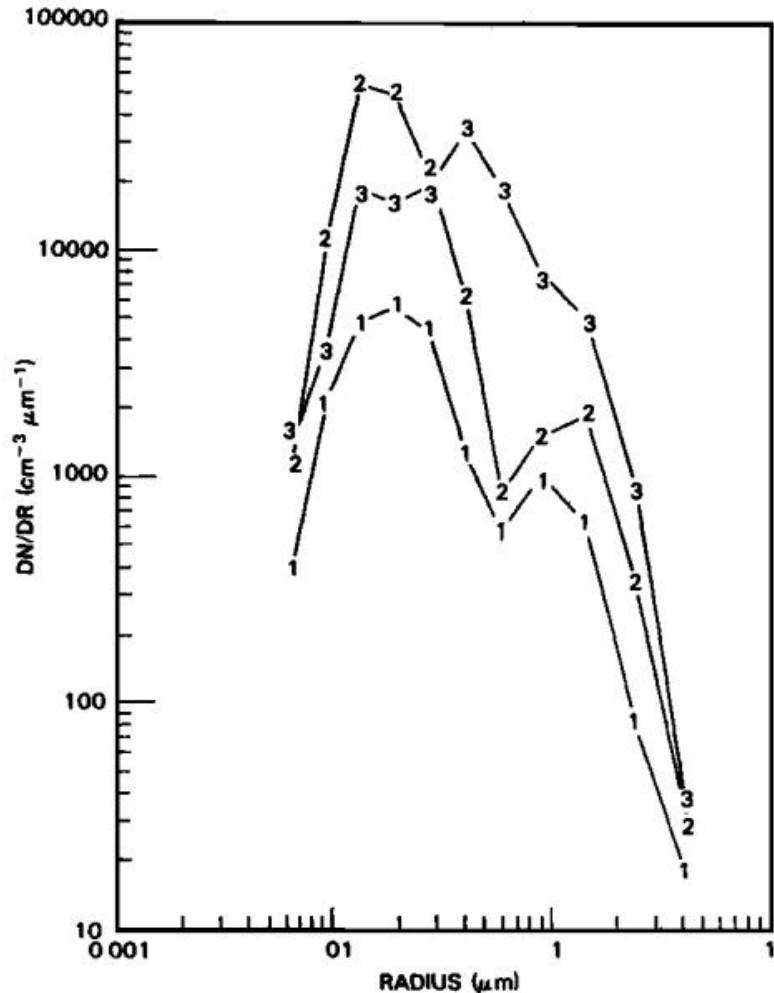
Exploration of cloud chemistry questions during EPCAPE-CCC

DOE-ASR Science Team Meeting

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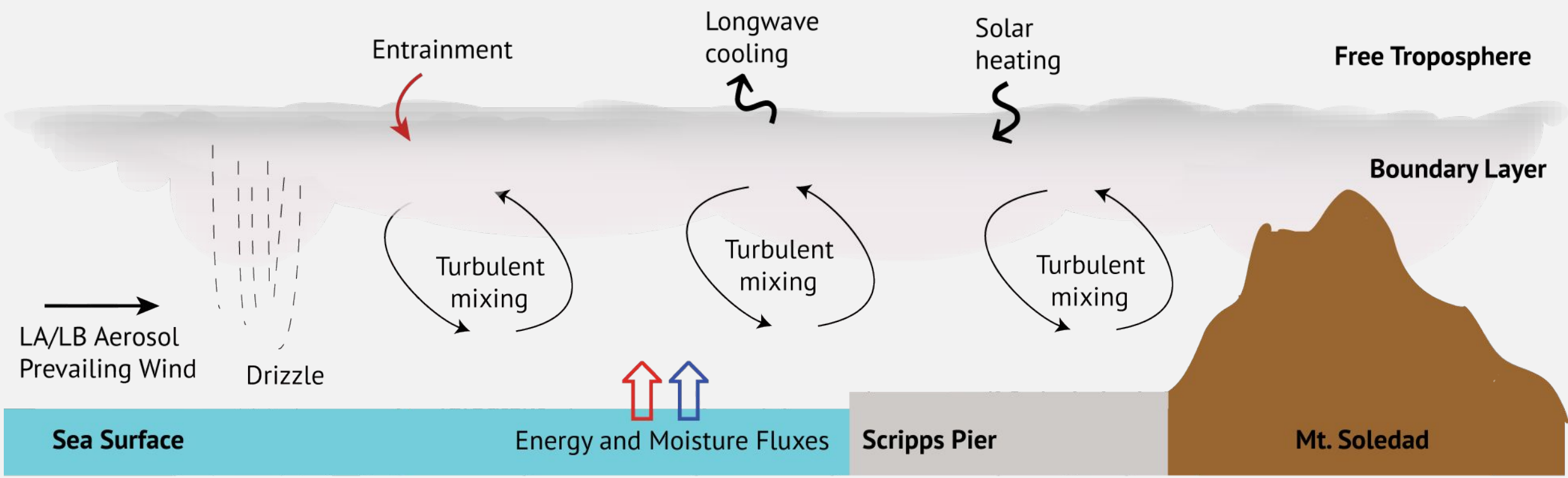
Effect of non-precipitating clouds on the aerosol size distribution in the marine boundary layer



“We believe that the data and arguments presented above make a strong case for the importance of nonprecipitating clouds in forming the size distribution. Nonprecipitating cloud cycles are probably the major mechanism for accumulating mass in the 0.08 to 0.5 μm size range where particles are very efficiently removed by Precipitation scavenging. **This mechanism has a tendency to produce a steep slope at sizes greater than about 0.1 μm and to produce a doubly peaked distribution in geographical regions where there is no direct particle injection in the 0.04 to 0.08 μm range.**”



Schematic diagram showing observable cloud processes at Mt. Soledad



Adapted from Wood [2012, Monthly Weather Review]

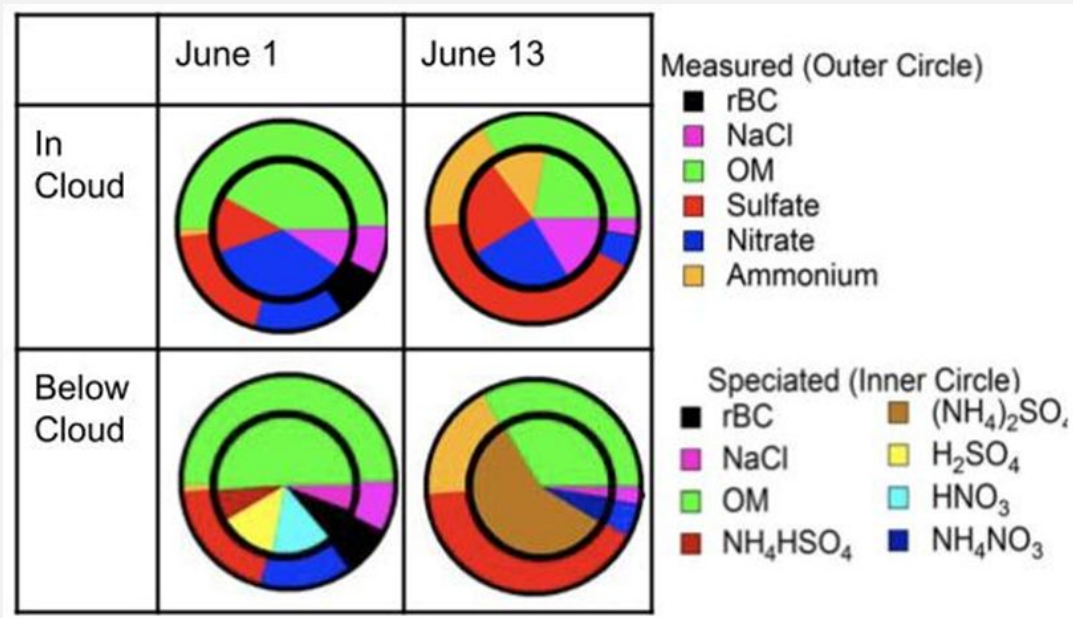
Proposes sampling from CVI and Isokinetic Inlet at Mt. Soledad



Instrument	Contact	Description	Inlet
Brechtel Counterflow Virtual Impactor (CVI)	Wheeler	Evaporates cloud droplets and provides residual particles to other instruments	N/A
Brechtel Differential Mobility Analyzer (DMA)	Russell	Number distribution of particles (0.02-0.9 μm)	Switched
*DMT Cloud Condensation Nuclei (CCN) Counter	Petters	CCN number concentration and supersaturation spectra of particles for 0.07-0.6% supersaturation	Switched
Mini Handix CCN (5)	Petters	CCN number concentration and supersaturation spectra of particles for 0.1-1% supersaturation	Both
Printed Optical Particle Spectrometer (POPS)	Petters	Aerosol number distribution (0.15-3 μm)	Switched
TSI Aerodynamic Particle Sizer (APS)	Russell	Number distribution of particles (0.5-10 μm)	Isokinetic
Aerodyne High-Resolution Aerosol Mass Spectrometer (HR-AMS) with Event Trigger (ET)	Russell	NR organic, sulfate, nitrate, chloride, ammonium mass fragment concentrations (0.07-0.8 μm) every 5 min	Switched
DMT Single-Particle Soot Photometer (SP2)	Wheeler	BC mass and number distribution (0.08-1 μm)	Switched
Aerodyne Iodide Chemical Ionization Mass Spectrometer (CIMS)	Liggio	Gas-phase compounds	Switched
Fog Droplet Monitor	Chang	Number size distribution of fog (cloud) droplets	N/A
DMT Photoacoustic Extinctionmeter (PAX)	Lee	BC concentration, aerosol light scattering and absorption coefficients	Switched
*Direct-to-Liquid Cloud Droplet OH Burst (DtL -OH)	Paulson	Hydroxyl radical formation by particles using direct-to-liquid sampling and fluorescence	Switched
*Filters for transition metals and OH burst	Paulson	Soluble metals by ICPMS and OH burst	Switched
Filters for FTIR and XRF	Russell	Organic functional group and element concentrations	Both

Hypotheses to be tested: Chemical Signatures of Cloud Processing

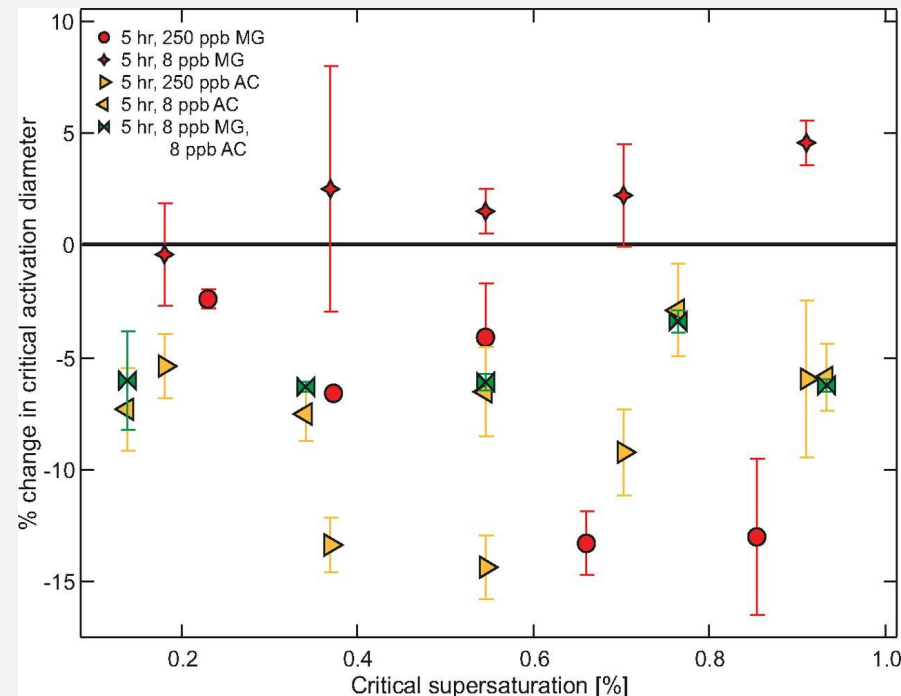
H1: The subset of the aerosol population that is activated to cloud droplets is chemically distinct from the unactivated particles, and its size-resolved chemical composition is further differentiated by adding aqueous-oxidized components that can be measured by single-particle mass fragments.



- Measured (outer circle) and simulated (inner circle) chemical composition for particles collected below cloud and in cloud.
- The simulated in-cloud droplet composition contains more nitrate, more salt, and less BC than is measured in the interstitial aerosol.
- The mixing state was unknown. Single-particle measurements of sulfate, nitrate, and carbon components are needed to accurately initialize simulations.

Hypotheses to be tested: Influence of gas-phase species on CCN activation

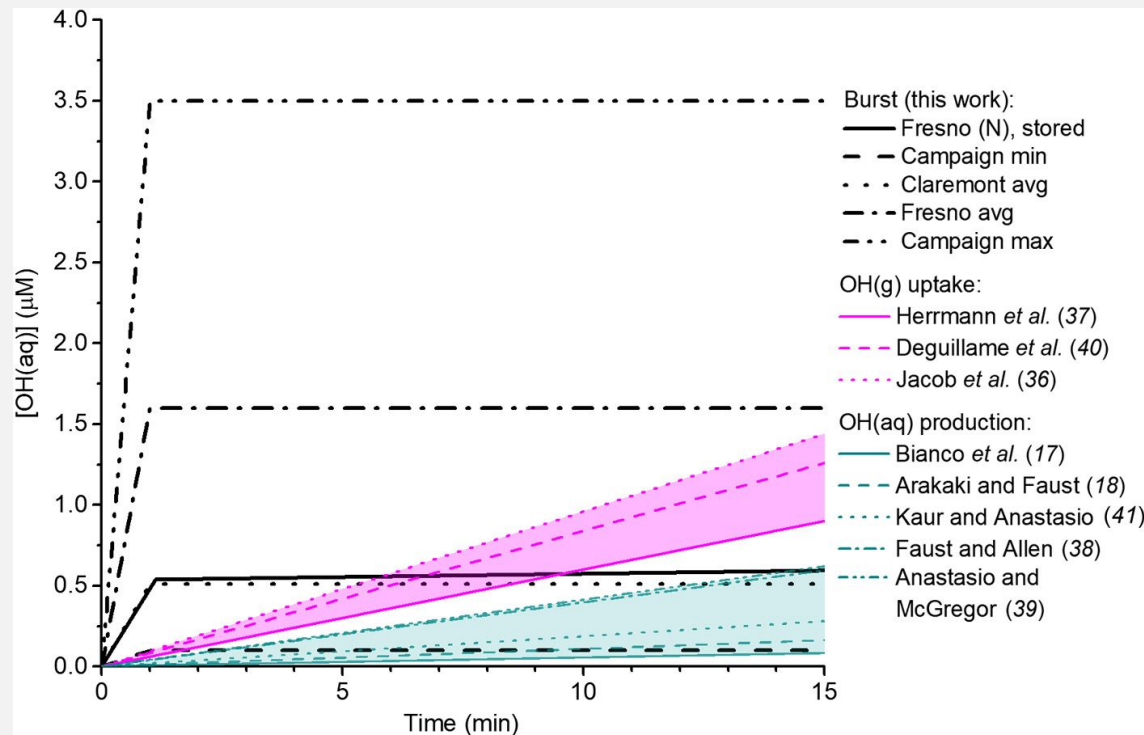
H2: Gas-phase compounds that are removed by denuding will lower the supersaturation required for activation of each particle by enhancing water solubility during the uptake process.



- Humidified ammonium sulfate aerosols were exposed to gas-phase methyl glyoxal or acetaldehyde in a Teflon reaction chamber.
- The critical dry diameters observed for each experiment as a function of instrument supersaturation are compared with the ammonium sulfate control to demonstrate the effect of organics.
- The data show a decrease in the activation diameter for particles exposed to methyl glyoxal and acetaldehyde.
- Other gases including nitric acid and semi-volatile organic vapors, such as those formed during secondary organic aerosol formation by oxidation are thought to have similar effects

Hypotheses to be tested: Aqueous phase production of hydroxyl radicals due to wetting

H3: Particles with longer times between cloud cycles will produce a larger OH burst, and as a result, larger changes to particle chemical composition during cloud cycling.

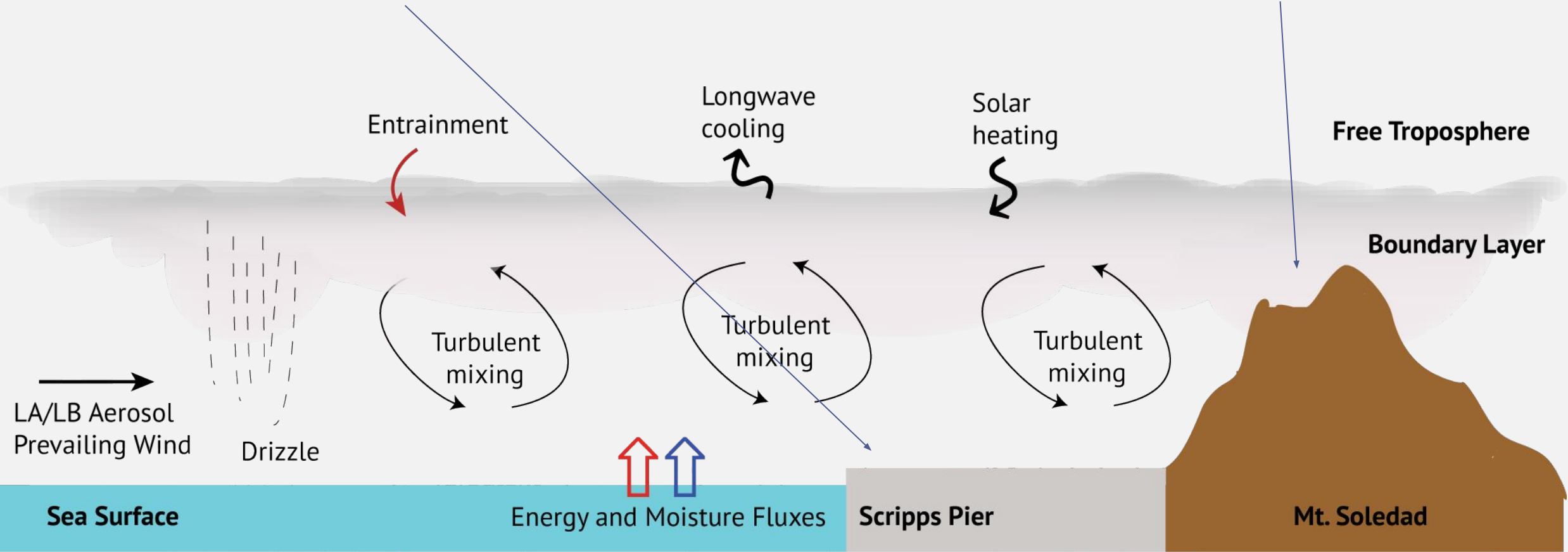


- Measured production from OH “bursts”. The solid line is a typical time-resolved burst from a Fresno stored sample.
- Dashed lines represent the minimum, average, and maximum cumulative concentrations from fresh samples.
- Magenta lines: Uptake of OH(g) into droplets, based on estimates from three cloud chemistry models.
- Green lines: Measured OH(aq) production in authentic cloud and fog water samples from five previous studies

Synergy with DOE-EPCAPE Deployment

EPCAPE-CCC
size distributions, detailed aerosol chemistry, comprehensive CCN measurements, CDNC estimates

AMF 1
in-situ aerosol measurements + remote sensors



EPCAPE-CCC will provide additional information about the vertical mixing of aerosol, aerosol properties near cloud base, cloud droplet number concentrations, and in-cloud supersaturations.