Aerosol Composition, Chemistry, and Source Characterization during 2008 VOCALS

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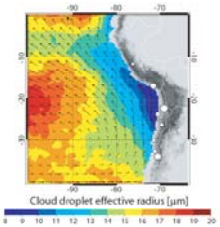
Primary objective of VOCALS*:

To improve understanding of the southeastern Pacific coupled ocean-atmosphere-land system on diurnal to inter-annual timescales.

Location: Coastal marine atmospheres off northern Chile
Time: October 15 to November 15, 2008

*VAMOS Ocean-Cloud-Atmosphere-Land Study

Why aerosol chemical composition Matters?



To understand aerosols regarding:
- Sources and formation pathways
- Optical properties
- Cloud nucleating properties
- Mixing state
- Cloud-aerosol interactions

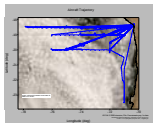
Increased aerosol loading near the shore inferred from decreased cloud droplet effective radius based on satellite observations

Experimental Section:

Cloud and aerosol microphysical properties were characterized using instrumented DOE Gulfstream-1 (G-1) aircraft flying below-, in-, and above-clouds.


G-1, Arica, Chile

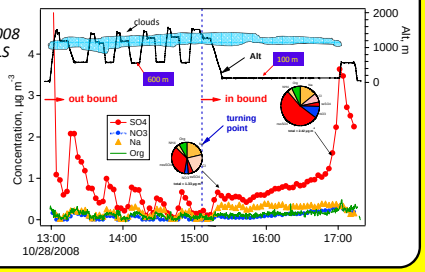

cToF-AMS and PILS


Composite flight tracks

Aerosol composition measurement techniques deployed on G-1

PILS-IC (3.0 min, bulk)	Na^+, Cl^-, $CH_3SO_3^-$, Mg^{2+} NO_3^-, SO_4^{2-}, NH_4^+ K^+, Ca^{2+}
cToF-AMS (22 sec, size resolved)	NO_3^-, SO_4^{2}, NH_4^+, Org (non-refractory)

Aerosol composition observed on Oct 28, 2008 flight, typical of VOCALS

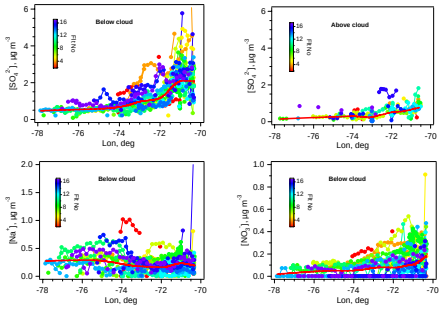


Findings:

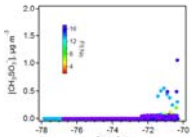
Composition, Chemistry, and Mixing State:

✓ SO_4^{2-} dominating, followed by NaCl, with NH_4^+, NO_3^- and organics each contributing only ≤ 10%

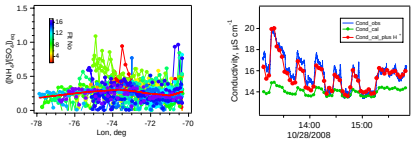
✓ Aerosol loading much higher in marine boundary layer (MBL) than in free troposphere, contrary to model predictions



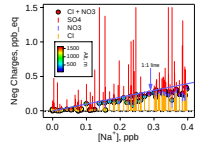
✓ SO_4^{2-} of terrestrial origin as ocean-emitted dimethylsulfide (DMS) and its product, $CH_3SO_3^-$, were practically non-existent



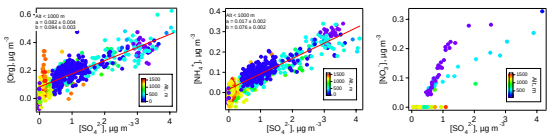
✓ SO_4^{2-} aerosols strongly acidic : $(NH_4^+/SO_4^{2-})_{eq}$ ~ 0.25; presence of H_3O^+ verified by conductivity measurement



✓ Sea-salt and SO_4^{2-} aerosols externally mixed as NO_3^- deposited on the former was detected by PILS, but not AMS



✓ SO_4^{2-} aerosols well mixed in MBL, but not sea-salt particles, consistent with size and mixing state



Source Identification:


Copper smelters – major SO_2 sources


Atacama desert, devoid of biogenic activities

Terrestrial:
- smelters
- power plants and urban emissions

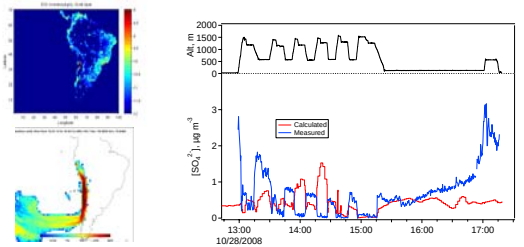
Less important:
- Agricultural activities
- Biogenic emissions
- dust

Oceanic:
- sea-salt

Unimportant:
- DMS chemistry

Model vs Observations:

Flexpart calculations driven by NCEP 0.5 x 0.5 deg met data with an up-to-date emission inventory for South America show underestimation of SO_4^{2-} in MBL near coast, and overestimation in free troposphere



Conclusions:

- Aerosols in marine atmospheres off northern Chile coast are dominated by anthropogenic sulfate and sea-salt particles
- The particles are highly hygroscopic due to high sulfate, strong acidity, and low organics
- Enhanced aerosol direct effect is expected because of the large growth factors of H_2SO_4 and NaCl on RH
- Aerosol cloud nucleating properties are governed by size
- Discrepancies between observed and predicted SO_4^{2-} aerosol concentrations need to be resolved


VOCALS DOE G-1 Participants

BROOKHAVEN NATIONAL LABORATORY

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