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

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

Aerosol composition measurement techniques deployed on G-1
PILS-IC (30 min, bulk)
Na+, Cl-, CH3SO3-, Mg2+, NO3-, SO42-, NH4+, K+, Ca2+
ToF-AMS (22 sec, size resolved)
NO3-, SO42-, NH4+, Olg (non-refractory)

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

Findings:
Composition, Chemistry, and Mixing State:
• SO42- dominating, followed by NaCl, with NH4+, NO3- and organics each contributing only ≤ 10%
• Aerosol loading much higher in marine boundary layer (MBL) than in free troposphere, contrary to model predictions

• SO2+ aerosols of terrestrial origin as ocean-emitted dimethylsulfide (DMS) and its product, CH3SO3-, were practically non-existent

• SO42- aerosols strongly acidic: (NH4+/SO42-) ≈ 0.25; presence of H2O+ verified by conductivity measurement

• Sea-salt and SO42- aerosols externally mixed as NO3- deposited on the former was detected by PILS, but not AMS

• SO42- aerosols well mixed in MBL, but not sea-salt particles, consistent with size and mixing state

Source Identification:
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 SO2+ 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 H2SO4 and NaCl on RH
• Aerosol cloud nucleating properties are governed by size
• Discrepancies between observed and predicted SO2+ aerosol concentrations need to be resolved

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