Vertical profiles of cloud condensation nuclei, aerosol hygroscopicity, water uptake, and scattering across the United States

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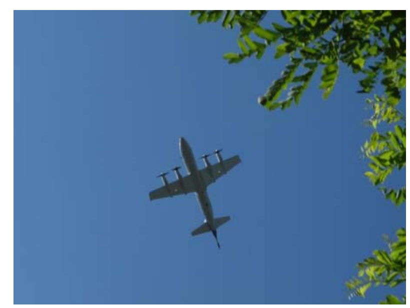


Introduction

Motivation:

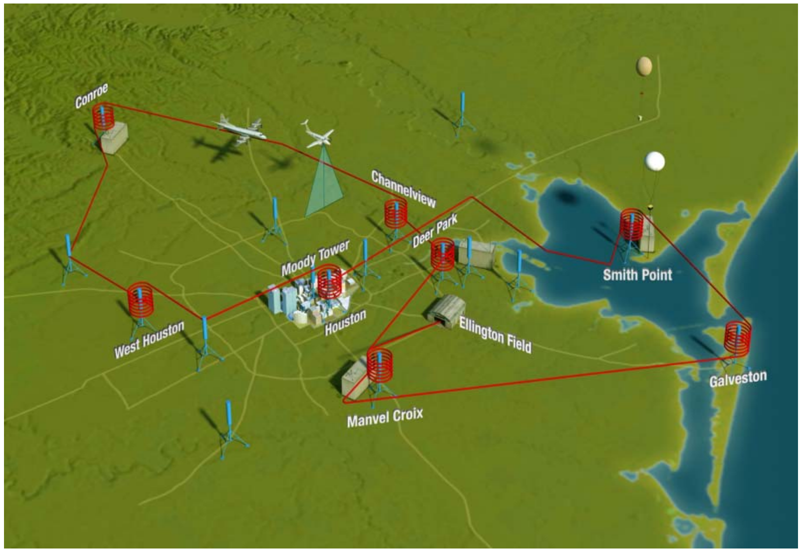
- Near surface pollution is difficult to diagnose from satellite-borne observations.
- Evolution of vertical distributions of aerosol properties are important for air quality and radiative transfer.
- Water uptake has a critical impact on aerosol optical depth and its radiative impacts (2-3 times the aerosol dry mass globally; Liao and Seinfeld, 2005).

Objectives:

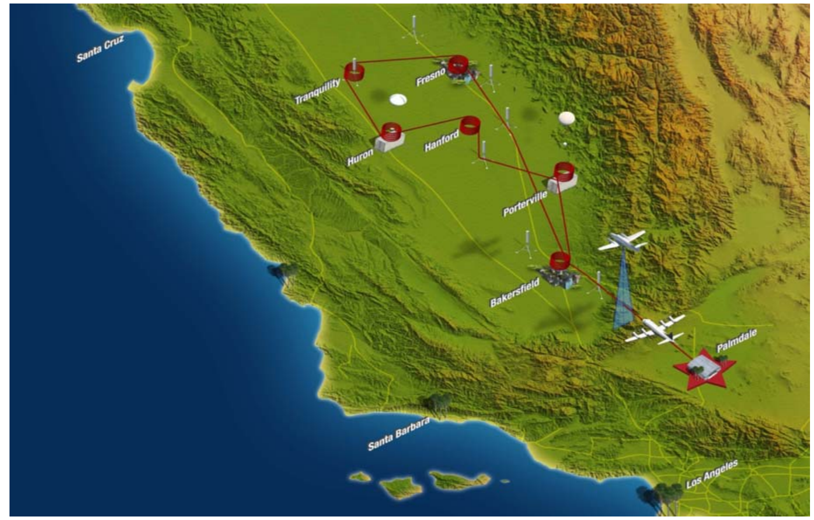
- *Vertical profiles of cloud condensation nuclei (CCN) and water uptake properties.*
- *Evaluate measurements of water uptake against predictions.*
- *Quantify the major contributors of LWC variability , particularly the relative role of organic vs. inorganic species.*

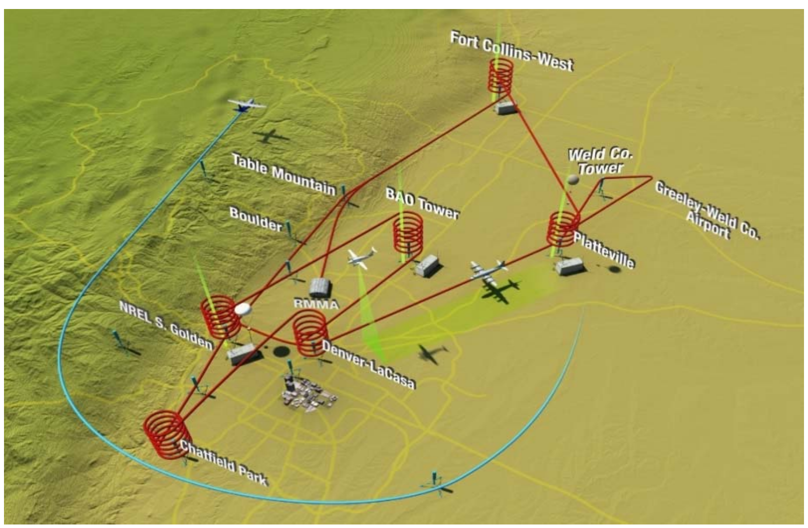
DISCOVER-AQ Datasets

Baltimore-Washington (July 2011)

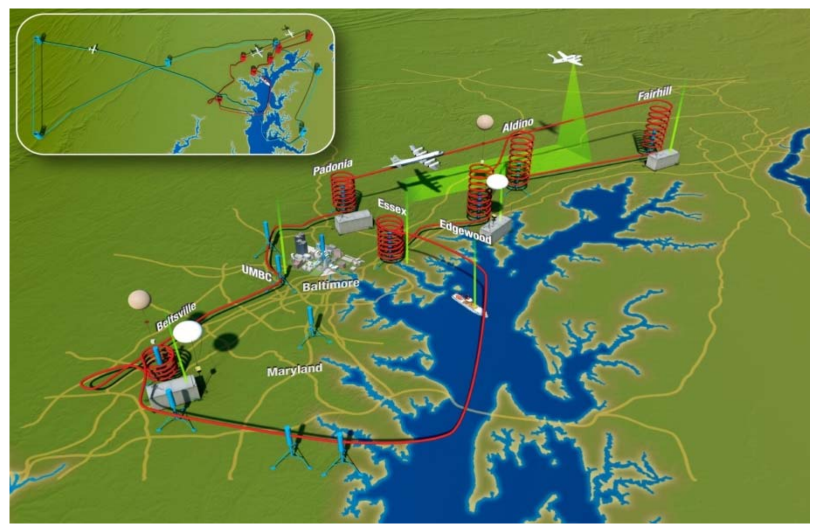


San Joaquin Valley (Jan-Feb 2013)



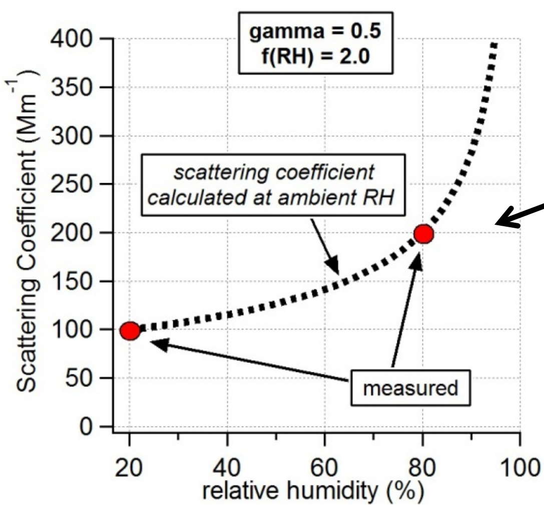


Denver, Colorado (July-August 2014)



Houston, Texas (September 2013)

Experimental methods: Data from DISCOVER-AQ



Aerosol Concentrations:

- Total and Non-Volatile Particles
- CCN counter (activation efficiency)

Aerosol Sizes (10 nm - 5 μm):

- SMPS, UHSAS, OPC & APS

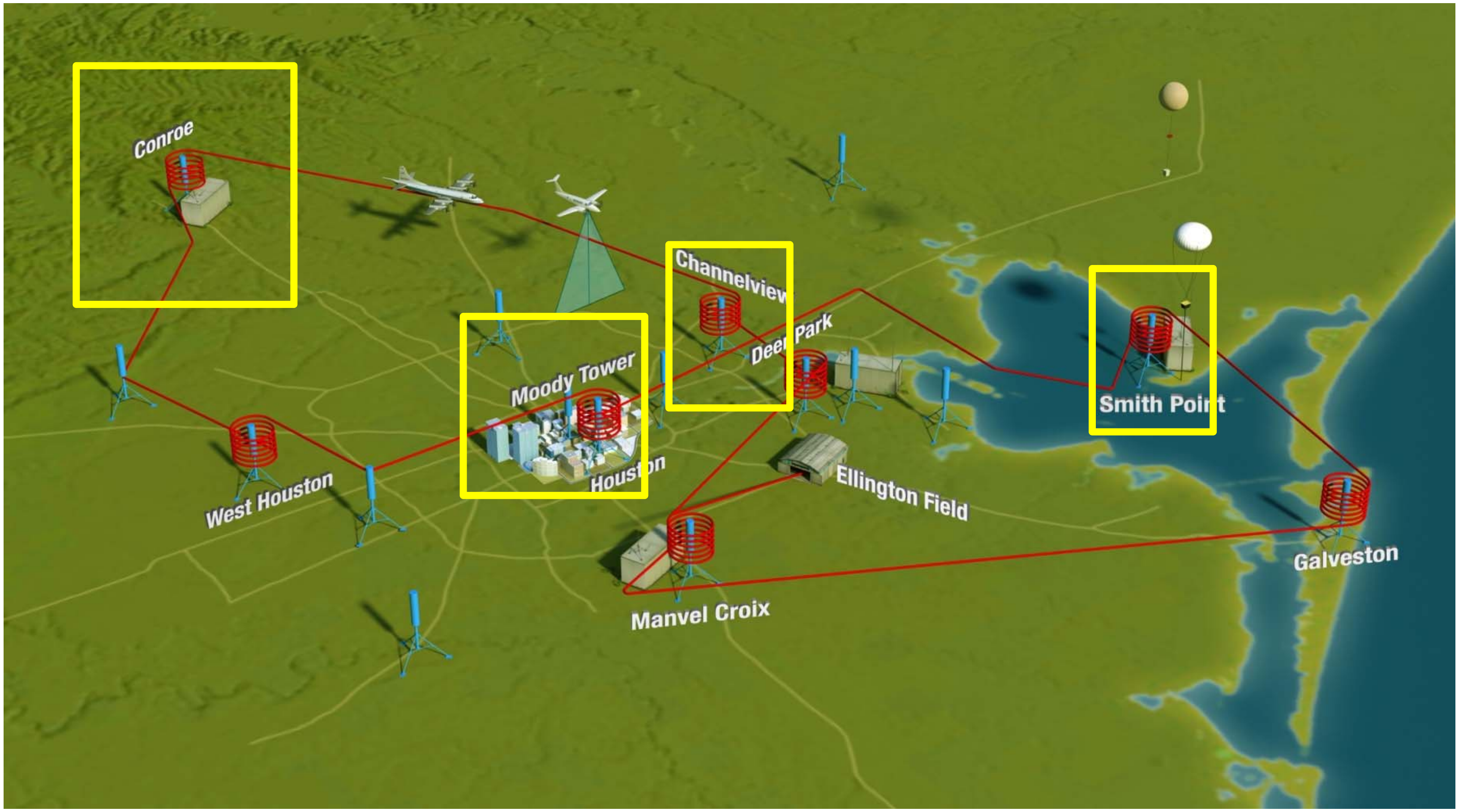
Optical Properties:

- Scattering & Absorption Coefficients
- Single Scattering Albedo
- Angstrom Exponent
- $f(RH)_{80/20}$ (effects of humidity on scattering)

Composition:

- Black Carbon Mass (SP2)
- Particle-Into Liquid Sampler (PILS, 4 min. resolution)

Focus on DISCOVER-AQ Houston Flights



- Unlike in other phases, Houston displayed a complex and heterogeneous vertical structure.
- Above boundary layer you had layers of smoke transported from east; sometimes aerosol in BL less concentrated than aloft.

Data used for analysis

PILS-IC (Particle-Into-Liquid-Sampler coupled with Ion Chromatograph) → water soluble ions in particles (SO_4^{2-}, NO_3^-, Cl^-, Br^-, NO_2^-, PO_4^{3-}, NH_4^+, Na^+, Ca^{2+}, Mg^{2+}, K^+, etc.).

PILS-TOC (Particle-Into-Liquid-Sampler, Total Organic Carbon) → water soluble organic carbon.

AMS (HR-ToF-AMS) → non-refractory components of submicron aerosols (primarily organic aerosol mass).

SMPS, UHSAS → aerosol size distribution

CCNc → particle hygroscopic parameter ($\boldsymbol{\kappa}$).

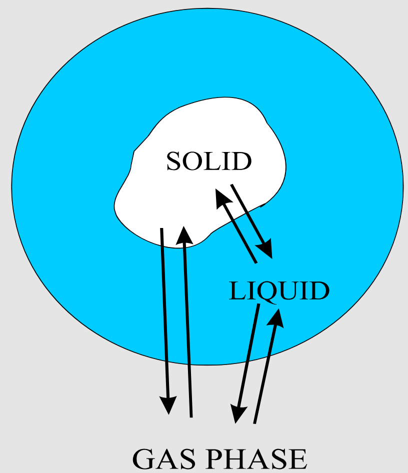
Nephelometers → ambient and dry aerosol light scattering coefficients (σ_{sp}), used to infer LWC.

$$f(RH) = \frac{\sigma_{sp}(wet)}{\sigma_{sp}(dry)} \qquad LWC = [f(RH)^{1.5} - 1]m_{dry}/\rho_p$$

Analysis methods - LWC calculations

Inorganic species: ISORROPIA-II (Fountoukis and Nenes, 2007)

Liquid: Na^+, NH_4^+, H^+, OH^-, HSO_4^-, SO_4^{2-}, NO_3^-, Cl^-, H_2O, $HNO_{3(aq)}$, $HCl_{(aq)}$, $NH_{3(aq)}$, **Ca^{2+}, K^+, Mg^{2+}**



T, RH

Solids: $NaHSO_4$, NH_4HSO_4, Na_2SO_4, $NaCl$, $(NH_4)_2SO_4$, $(NH_4)_3H(SO_4)_2$, NH_4NO_3, NH_4Cl, $NaNO_3$, **K_2SO_4, $KHSO_4$, KNO_3, KCl, $CaSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $MgSO_4$, $MgCl_2$, $Mg(NO_3)_2$**

Gas: HNO_3, HCl, NH_3, H_2O

Organic species: κ-Köhler theory (Petters and Kreidenweis, 2007)

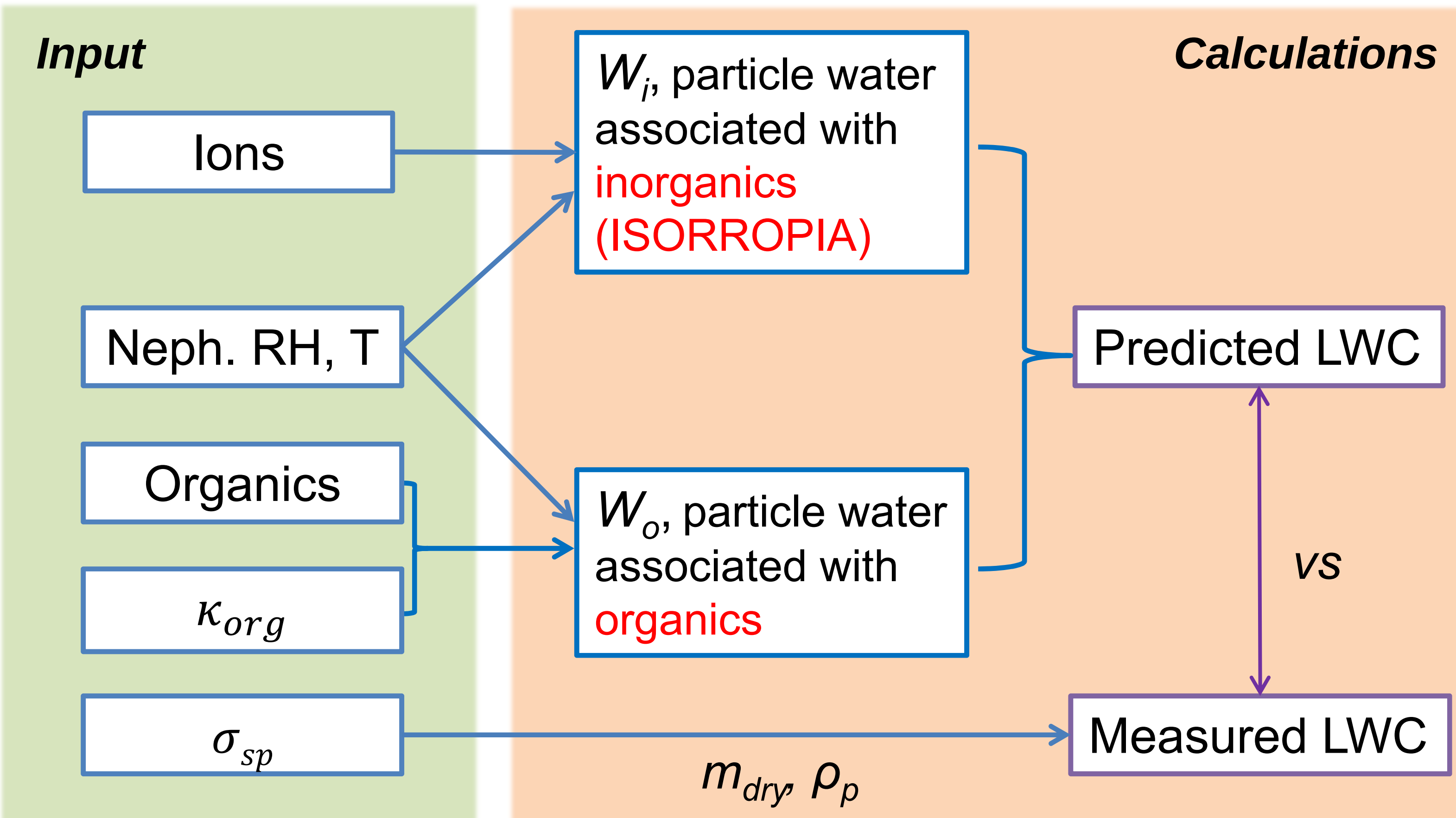
$$W_o = \frac{m_o}{\rho_p}\frac{\kappa_o}{(1-\mathrm{RH})}$$

m_o: aerosol mass
ρ_p: aerosol density
k_o: hygroscopicity parameter

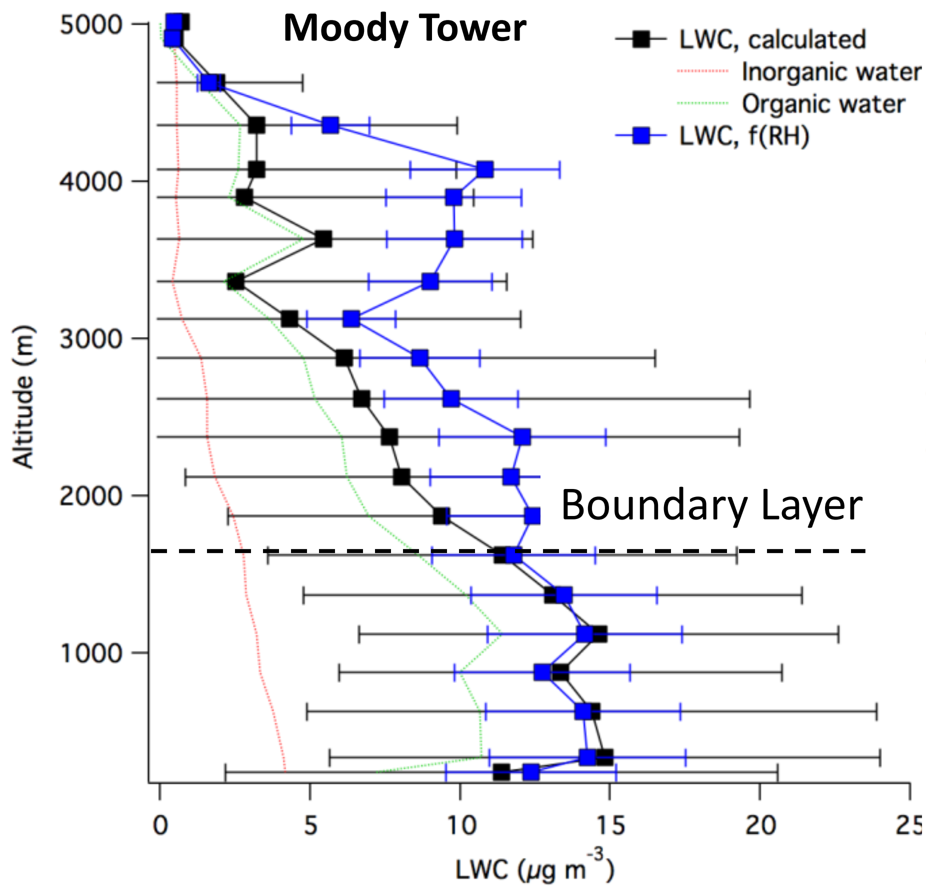
Analysis method: LWC/hygroscopicity closure

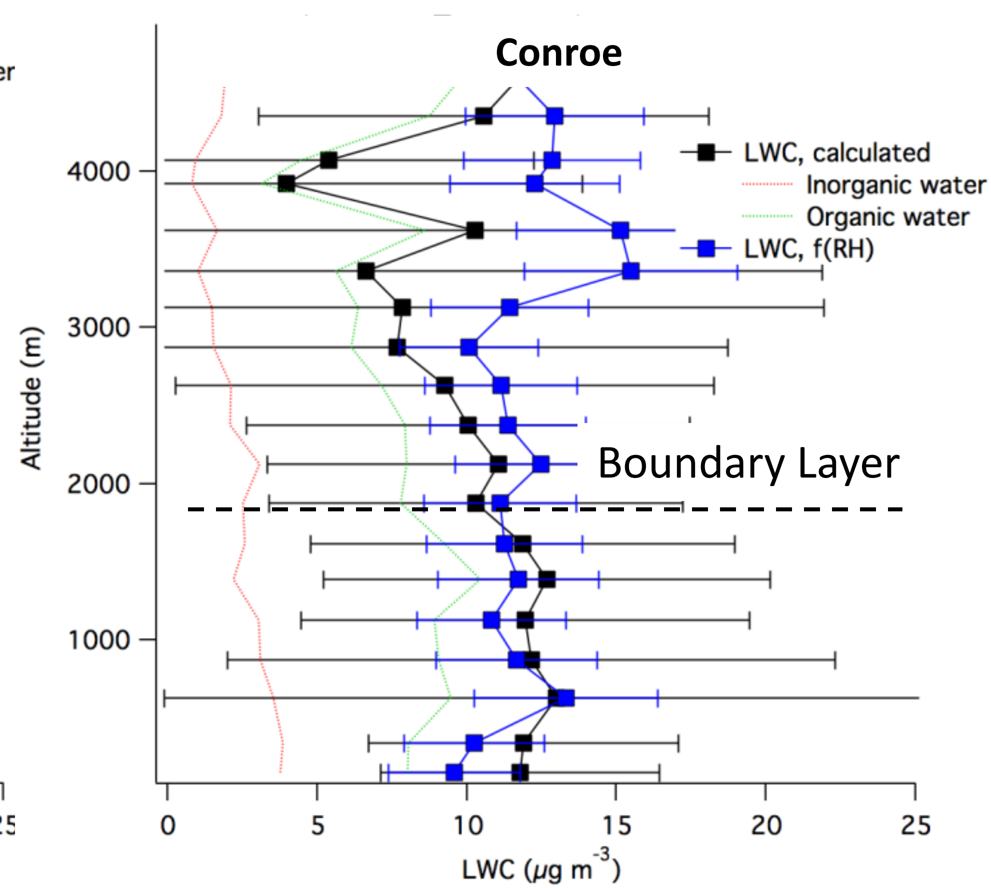
Input data includes:

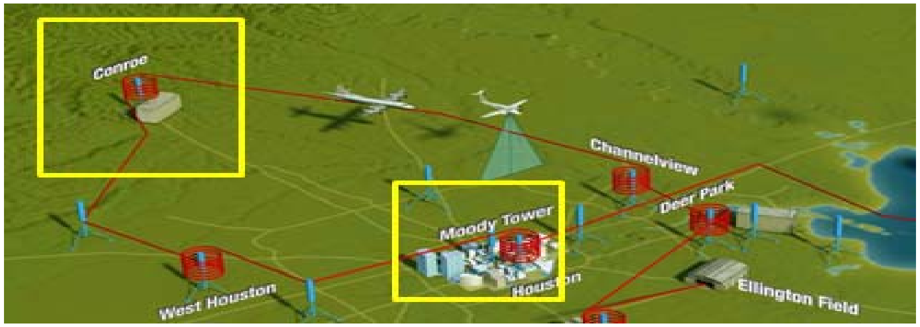
- *Particle ions (**SO_4^{2-}, NH_4^+**, NO_3^-, Cl^-, Na^+, K^+, Ca^{2+}, Mg^2);*
- ***Total organics**, κ_{org} **and f(RH)***
- ***Nephelometer RH** and T*



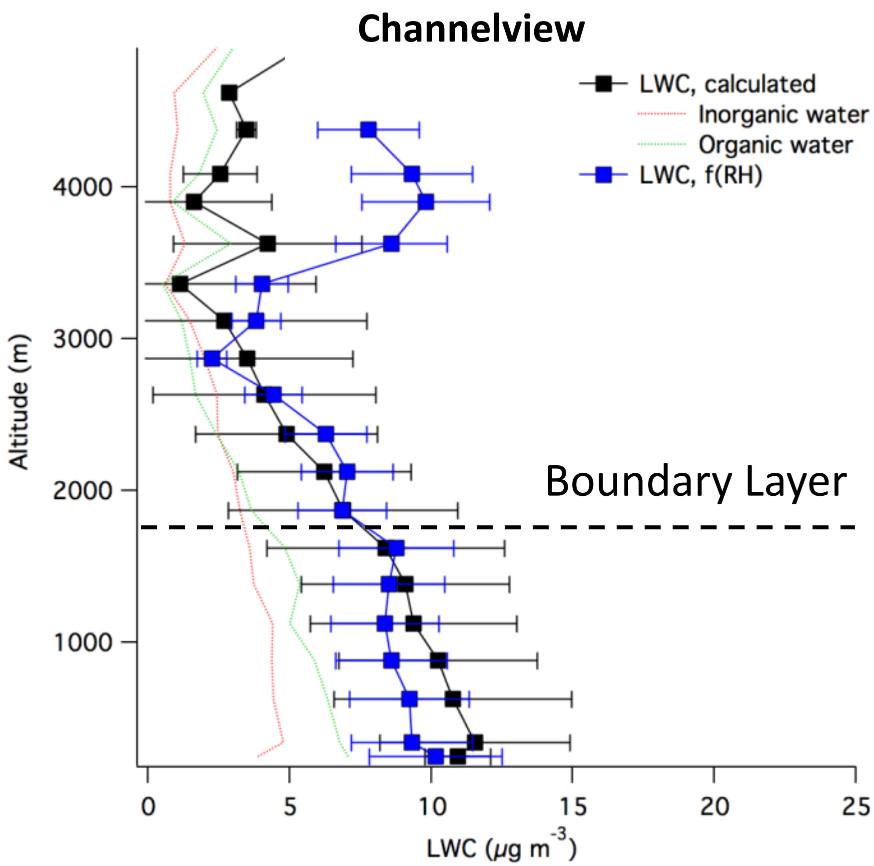
Analysis method: LWC/hygroscopicity closure

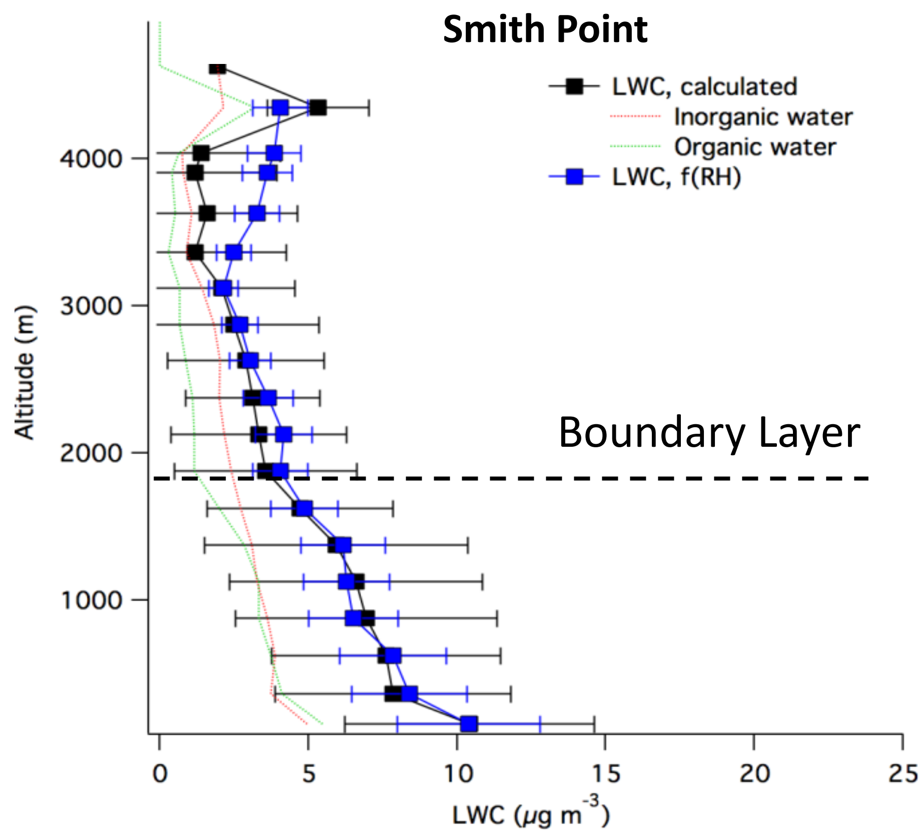






Analysis method: LWC/hygroscopicity closure

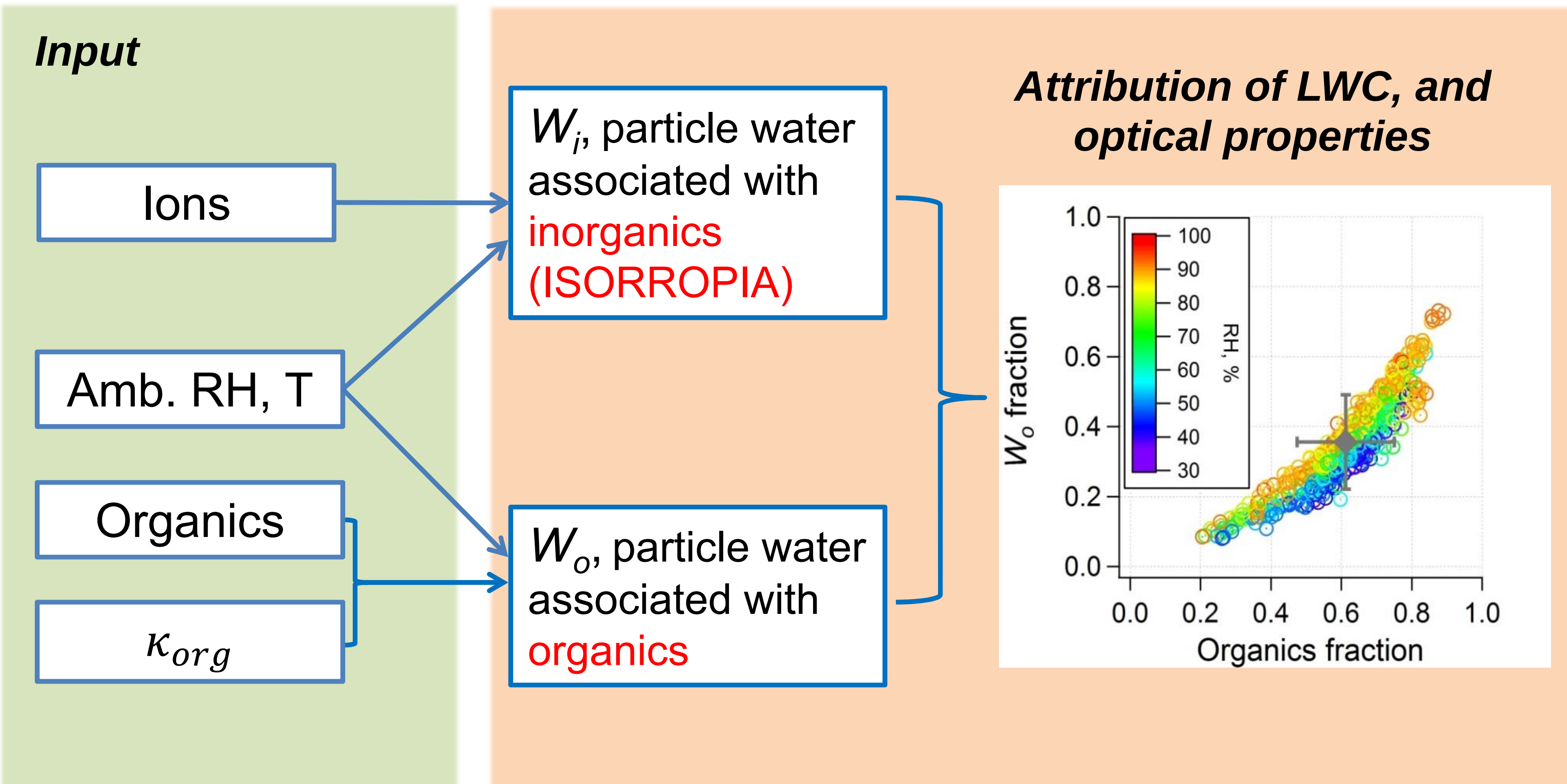




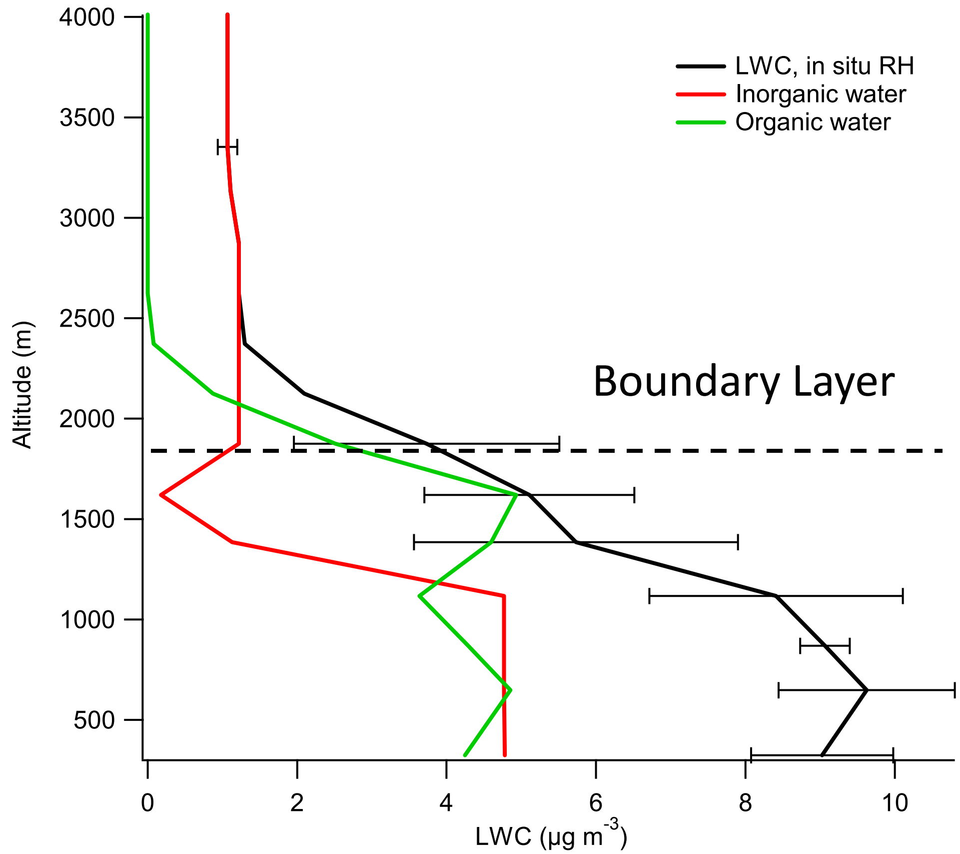
Analysis method: LWC attribution for ambient RH

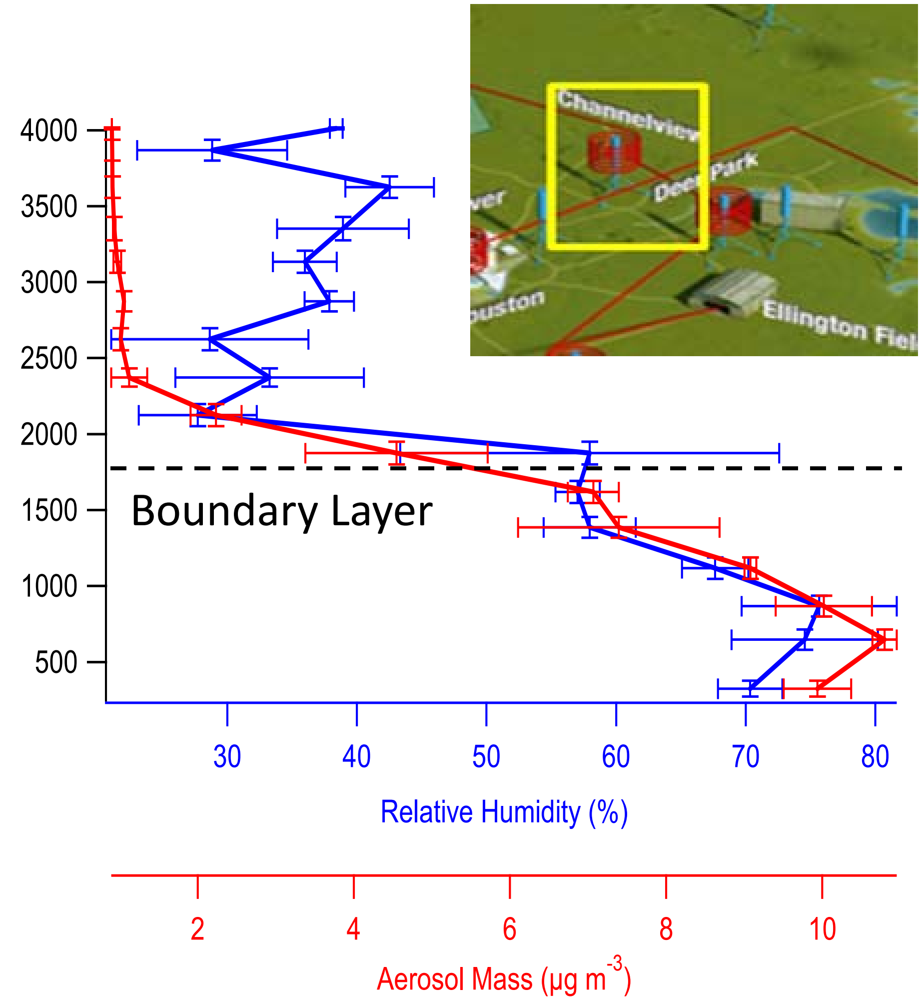
Input data includes:

- *Particle ions (**SO_4^{2-}, NH_4^+,** NO_3^-, Cl^-, Na^+, K^+, Ca^{2+}, Mg^2);*
- ***Total organics**, κ_{org} **and f(RH)***
- ***Ambient RH** and T*



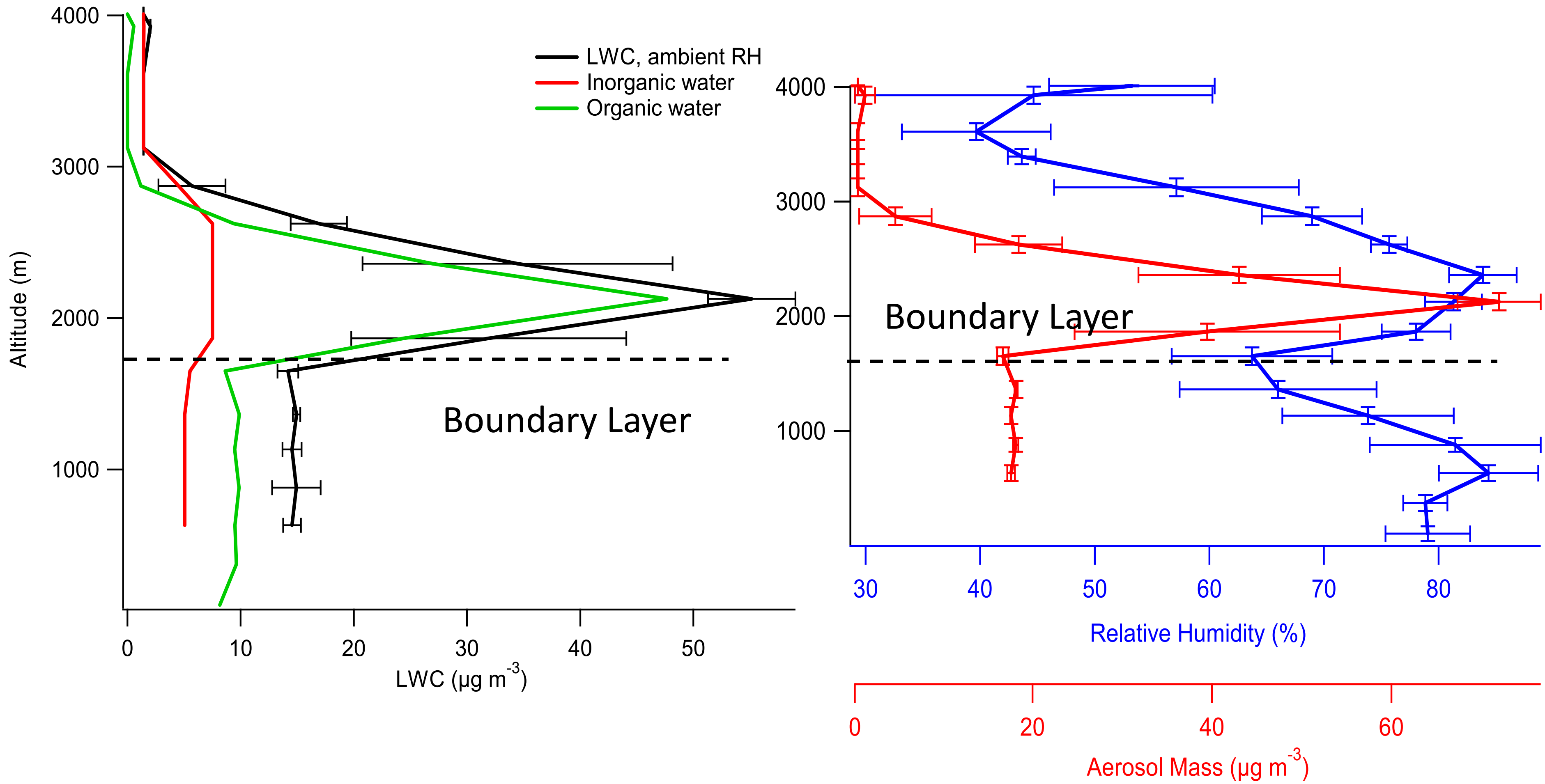
LWC attribution: Channelview





Organics contribute comparable (or more) water than inorganics
Most of the dry (and wet) aerosol mass in the boundary layer

LWC attribution: Galveston



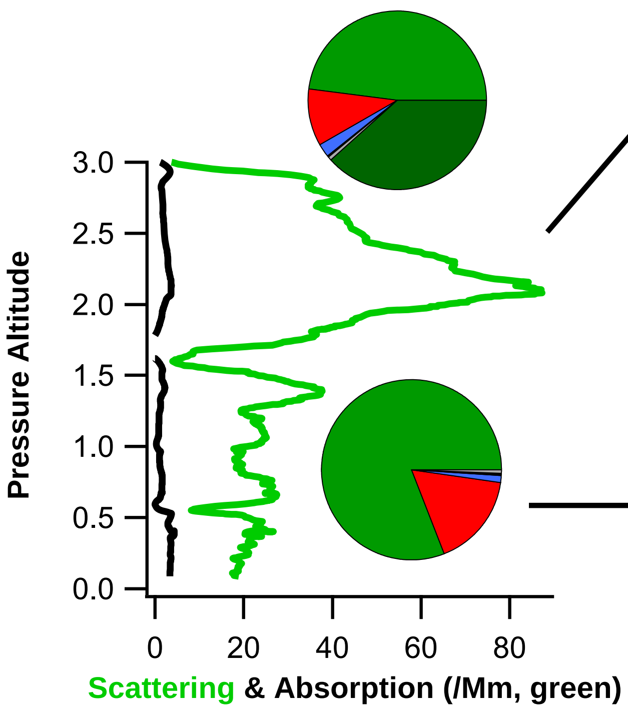
- Organics contribute comparable (or more) water than inorganics
- Smoke above boundary layer that dominates the aerosol (+water) mass in the column.

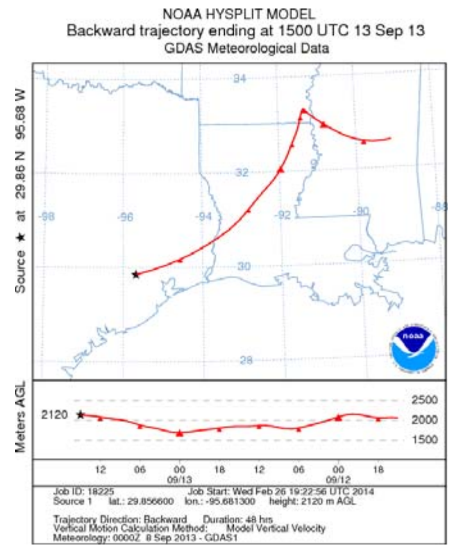
Biomass burning influence above boundary layer?

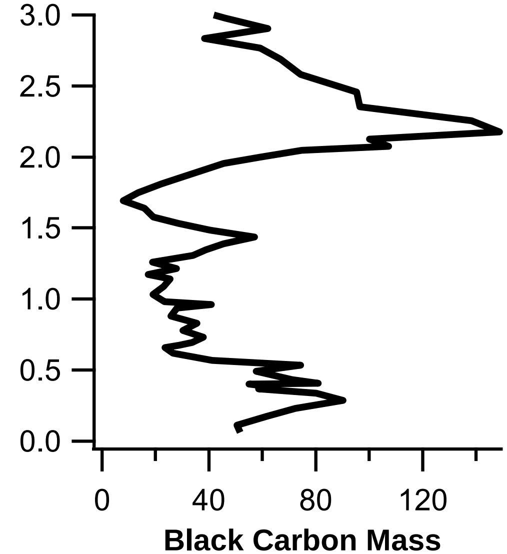
Sept. 13th, Circuit #1

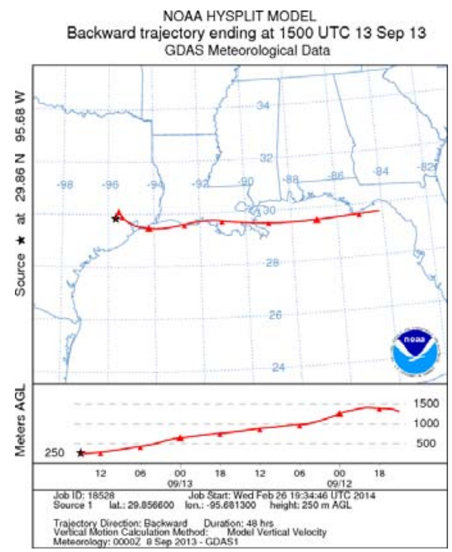
Highest aerosol loadings at 2 km in the northern portion of the circuit

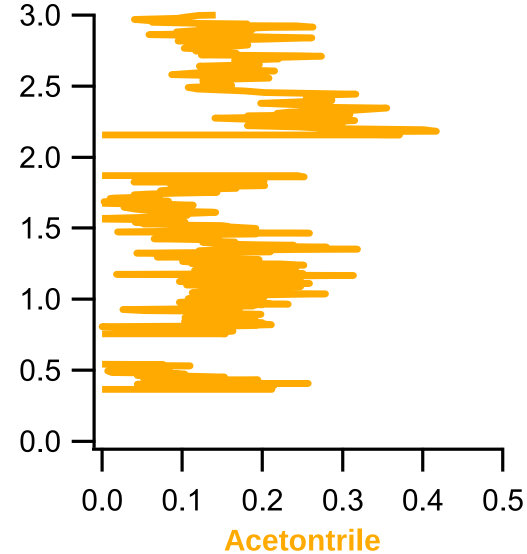
- Northeastern back-trajectories
- Acetonitrile ~ 300 pptv
    - Possible indication of smoke







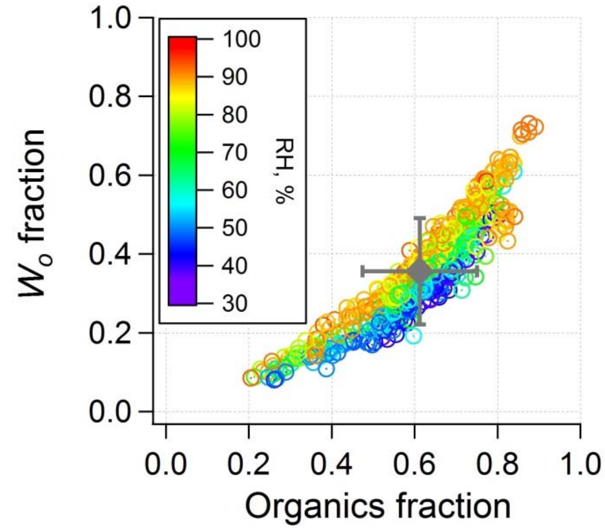




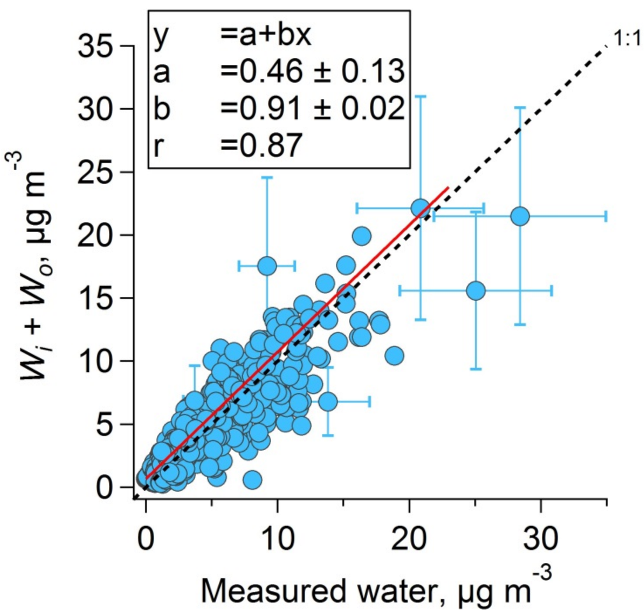
Comparison against SOAS (Jun-Jul 2013)

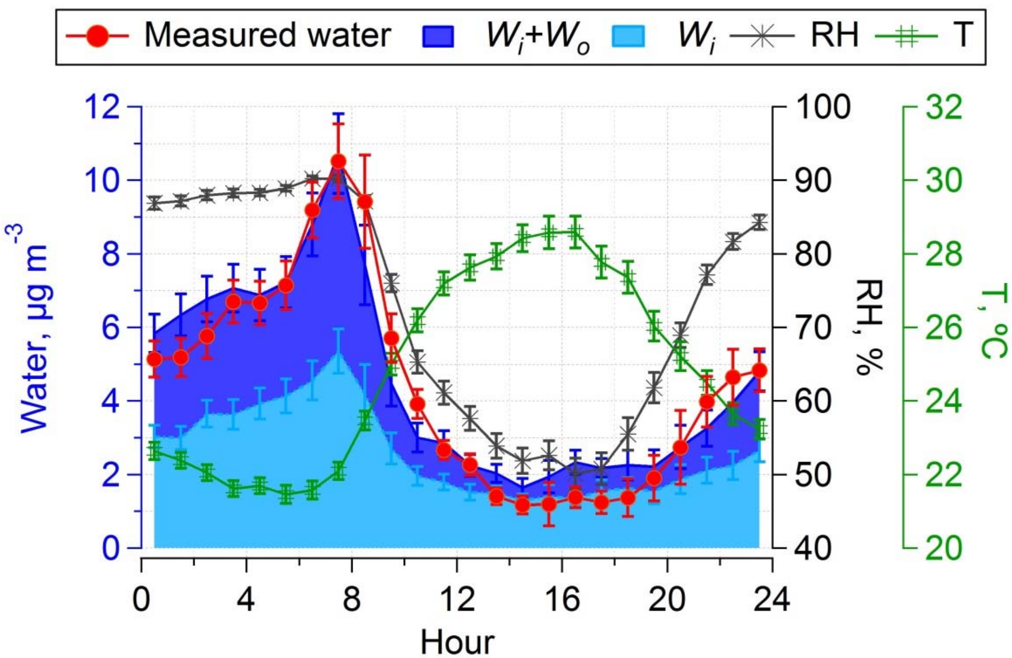
- W_i: LWC associated with inorganics
  W_o: LWC associated with organics
- Total predicted water ($W_i + W_o$) matches measured water very well (at ambient RH)
- LWC diurnal ratio (max/min) is **5**.
- W_o was significant, **29-39%** of total LWC at all sites. *(See Guo et al., 2014, ACPD)*

Fraction of organic water



Liquid Water: Predicted vs Measured





Take home messages

- Thermodynamic prediction of LWC verified by f(RH) and hygroscopicity measurements.
- Organics (mostly water-soluble) dominated the aerosol composition.
- Water associated with organic species is significant: 20-90%.
- The effect of organic water is higher in the BL but still significant above. Sometimes even more important (BB).
- The importance of organic water is not episodic but seems to be regional (SE US).
- This has important implications for aerosol chemistry .
- Aerosol loadings at ground-level (Houston) were low but high altitude aerosol layers contributed significantly (hence AOD).

THANK YOU!