Transformation of organic aerosol in Sacramento during CARES**

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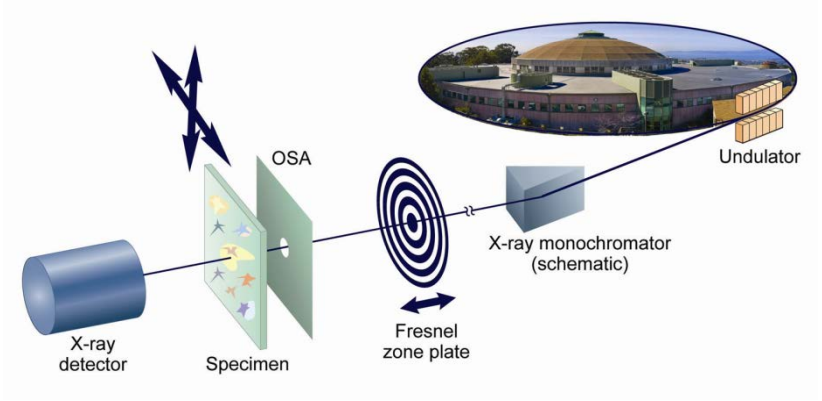
**See poster #71 & ACPD paper

Motivation Background

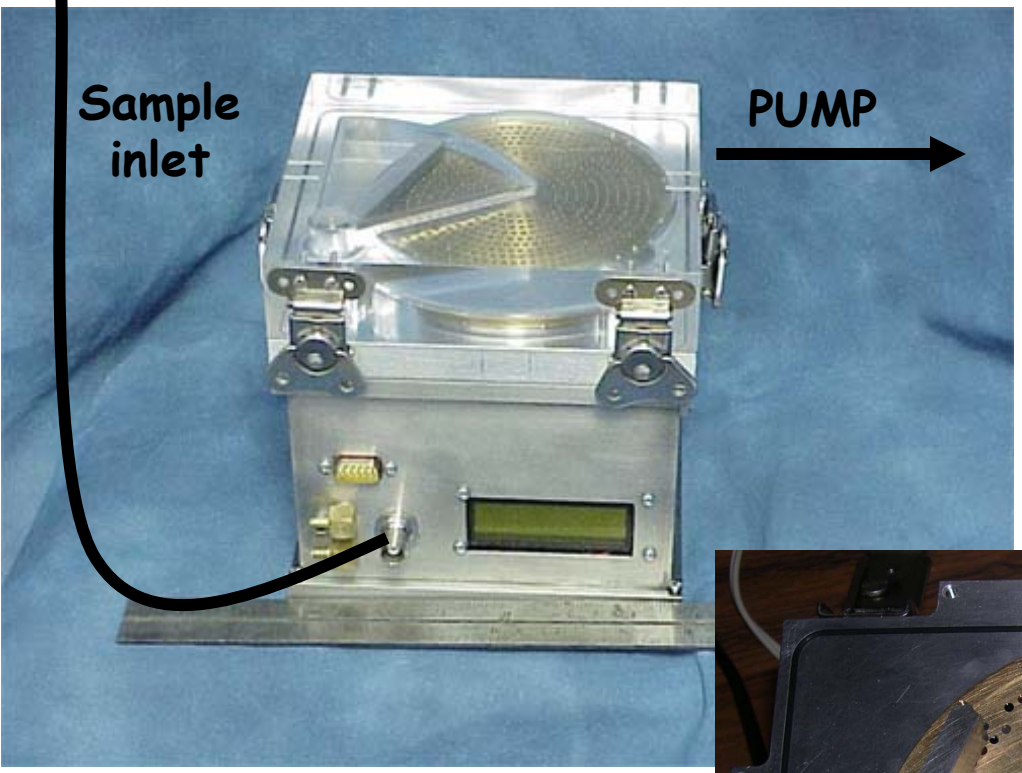
- Mixing state evolution (Soot, Organics, Inorganics)
- Direct effects: Does mixing state affect optical properties?
- Indirect effects: Water uptake, ice nucleation
- Microscopic methods:

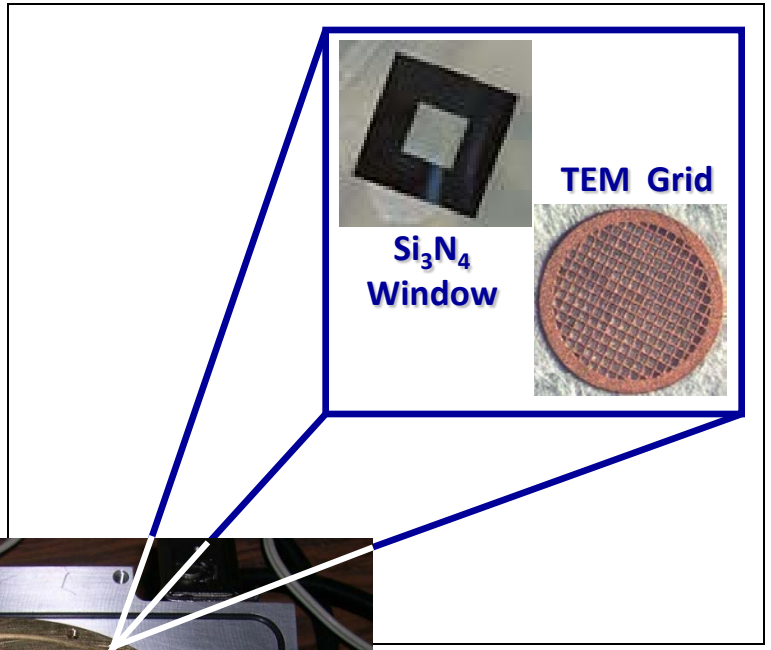
  - Scanning electron microscopy

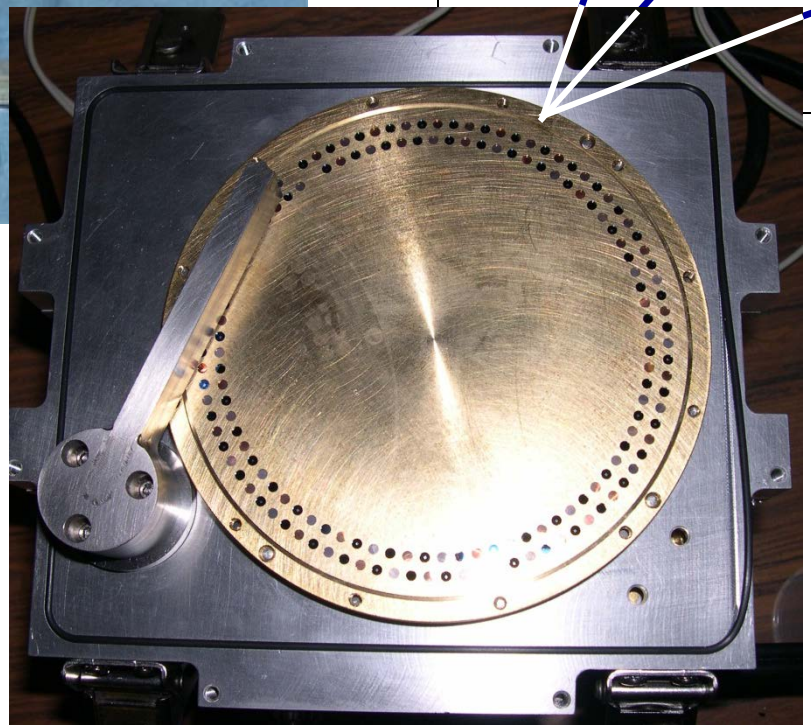
  - Scanning transmission microscopy
- Further studies on water uptake and ice nucleation (See Knopf poster 236, O'Brien poster 237, Room 1)



Time Resolved Aerosol Collection







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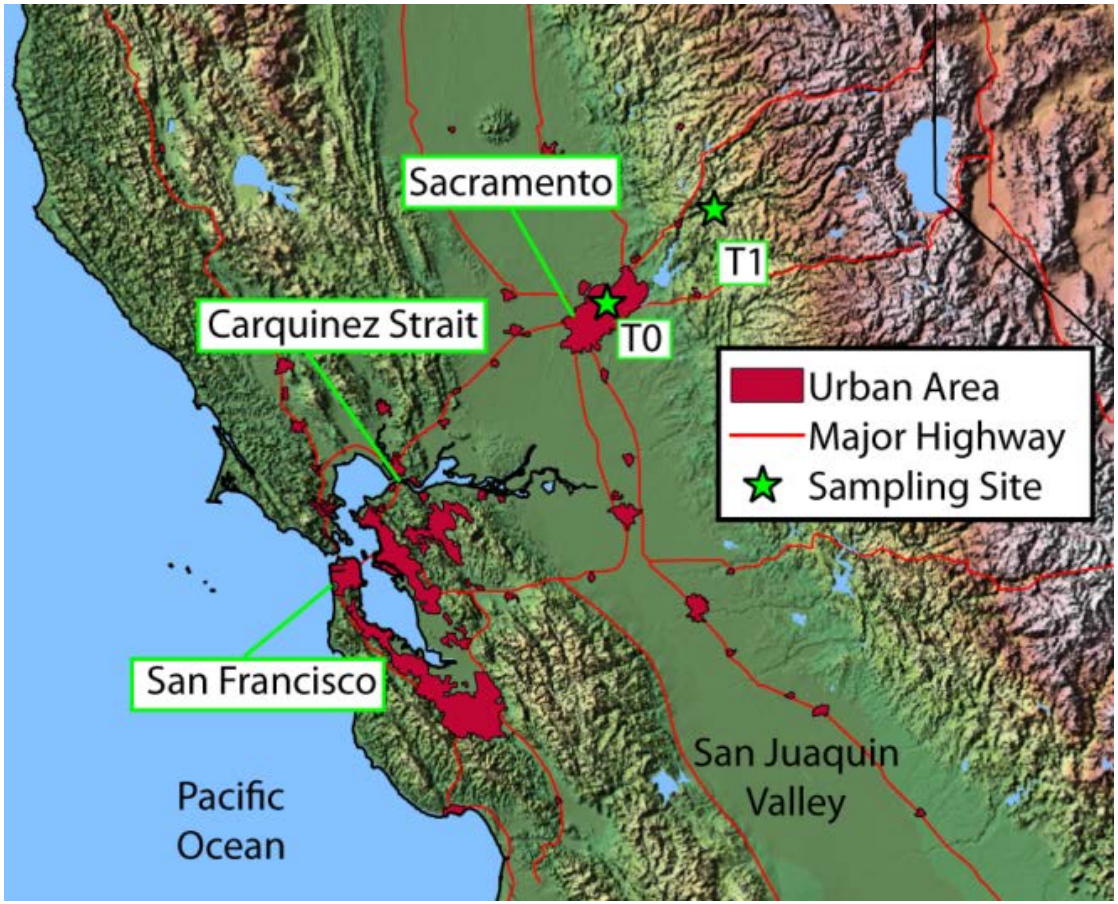
The Carbonaceous Aerosols and Radiative Effects Study (CARES)



Sampling sites (T0 ant T1) selected progressively further from the urban area.

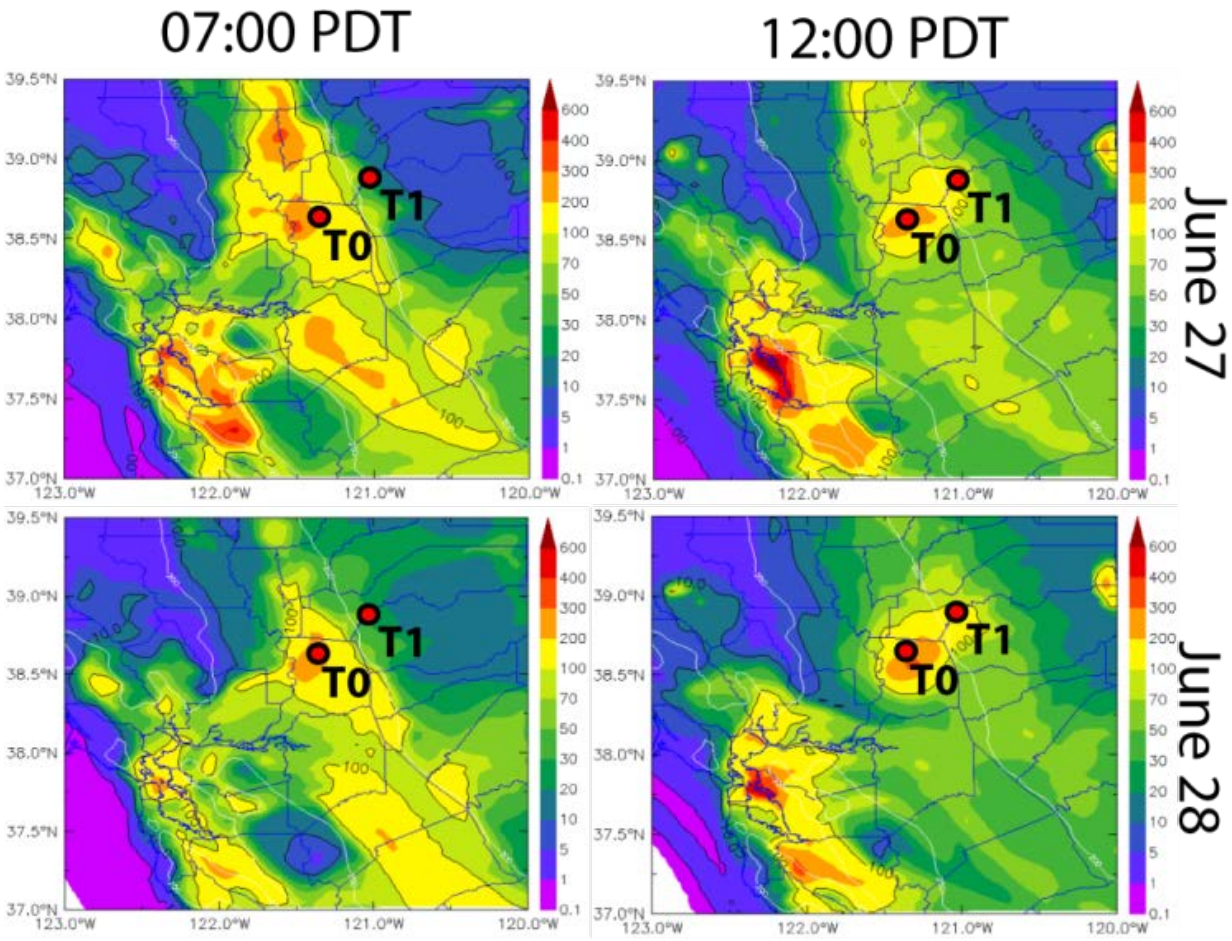
Microscopy impactors set up at each site

Extensive suite of instrumentation gave information on aerosol chemical and physical properties*.

Meteorological modeling provided guidance for measurements.

*Zaveri et al., Atmos. Chem. Phys., 12, 7647-7687, 2012

Anthropogenic Tracer Forecasts



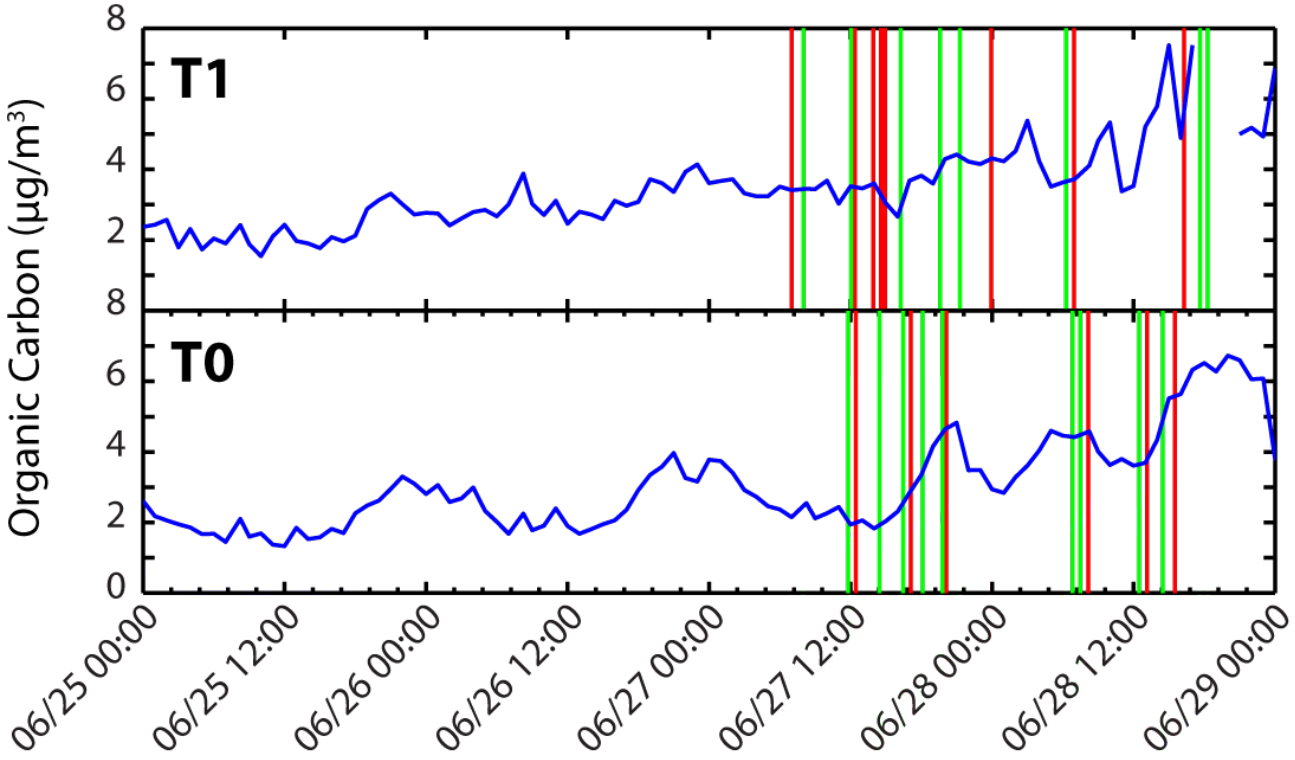
Colorscale proportional to CO concentration.

As boundary layer increased during the day, pollution was advected from T0 to T1.

Unique stagnant conditions towards the end of the campaign

Ground based measurements indicated marked change in aerosol properties.

Organic Carbon Mass "Buildup" at Campaign End

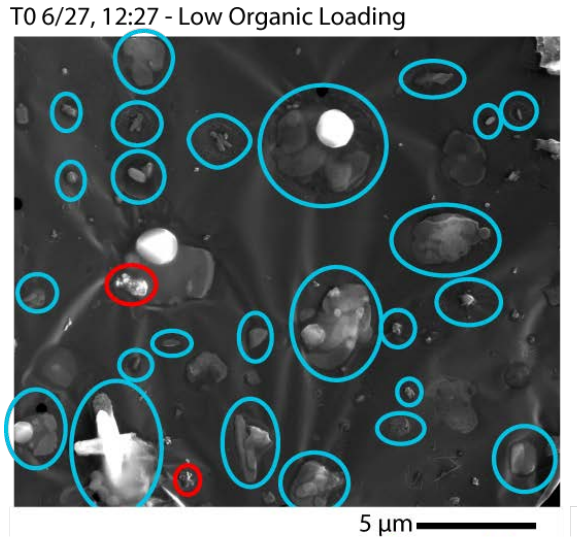


particulate organic carbon mass at the T0 and T1 sites

Organic mass increased towards end of the campaign

Green and red lines indicate x-ray and electron microscopic samples

General Particle Characteristics – An SEM View



At T0 during the low organic period, large inorganic particles were observed

Many of these particles were sea salt and sulfates*

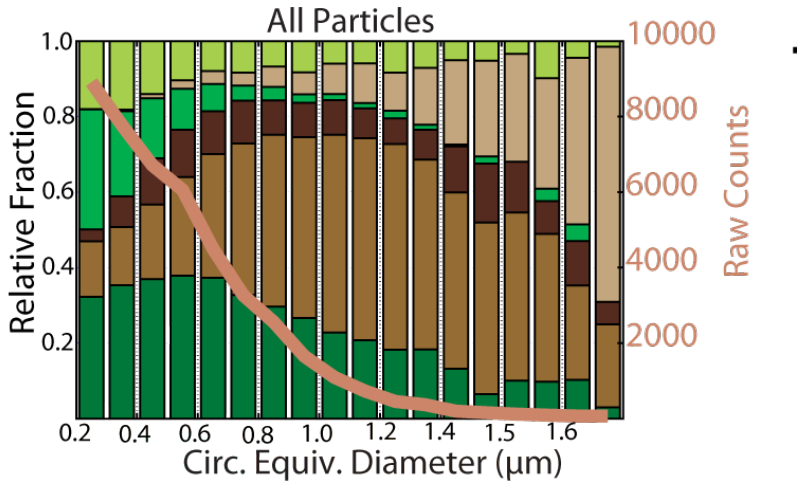
At T1, during the period of high organic loading, more secondary OC types were observed

At T0, biological brochosomes were frequently observed

Statistical numbers of particles were observed using computer controlled data aquistion.

*Laskin et al., JGR, 117, doi:10.1029/2012JD017743

Cluster Analysis of Computer Controlled SEM Data



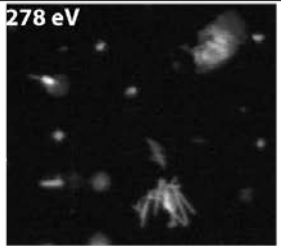
Green particle types: small and probably mostly organic

Brown particle types: larger and dominated by inorganic tracers

Cluster 1 Nitrogen Containing Carbonaceous
Cluster 2 Coarse Carbonaceous/Nitrogenous
Cluster 3 CONa Dust/Seasalt
Cluster 4 Secondary Carbonaceous
Cluster 5 CaMgAlSi Dust/Seasalt
Cluster 6 Refractory Carbonaceous

Low & High OC Loading – A STXM View

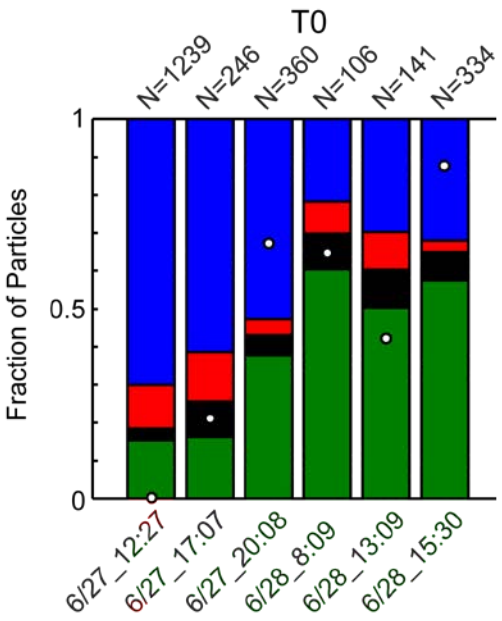
Low OC Loading

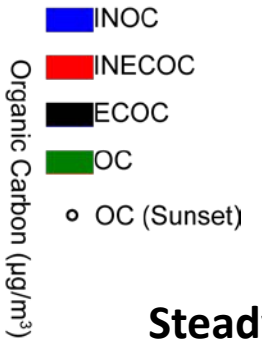


2 µm

285 295 305 315
Energy (eV)

STXM Microscopy Particle Classes



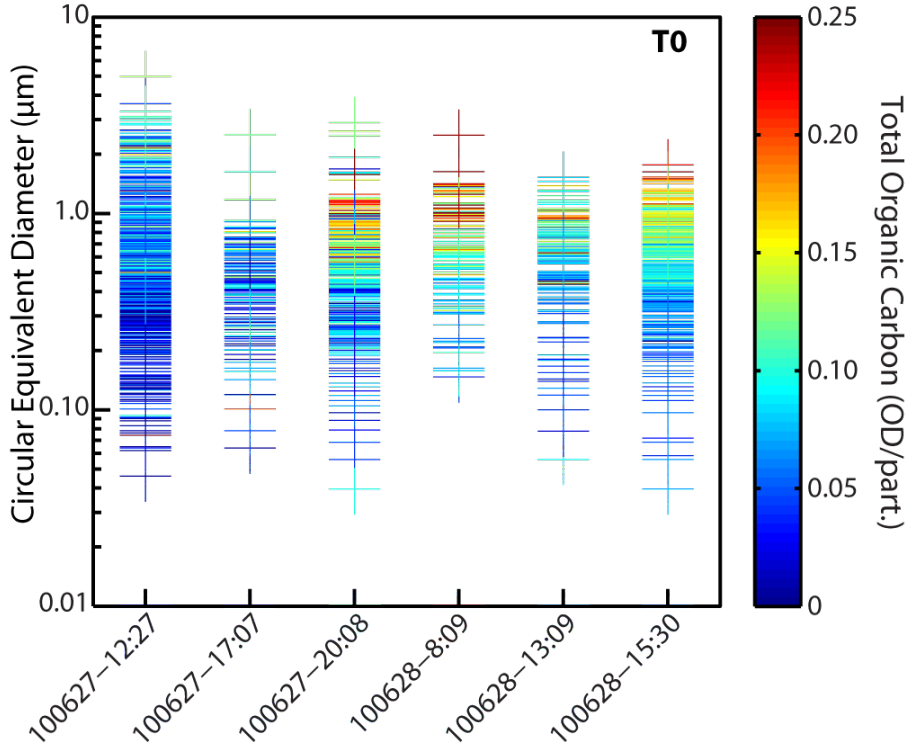


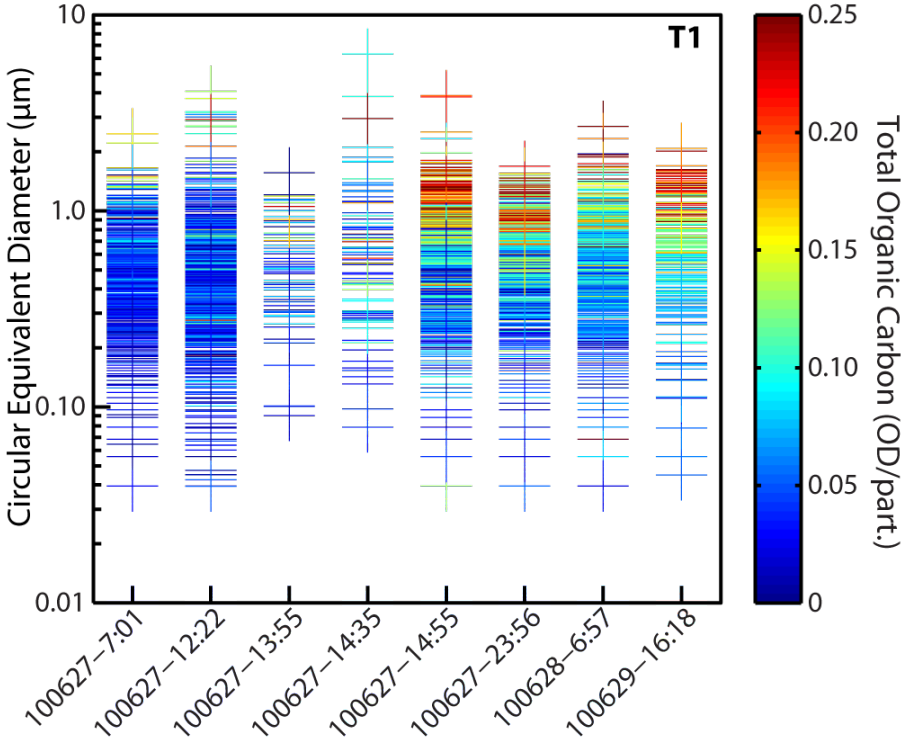
Steady increase of organic particle types at T0 reflects "buildup"

Variable amounts of OC at T1 indicative of boundary layer movements

Good correlation of single particle total OC and bulk OC indicates a stronger internal mixture

Single Particle Total OC – Trends with Size and Time





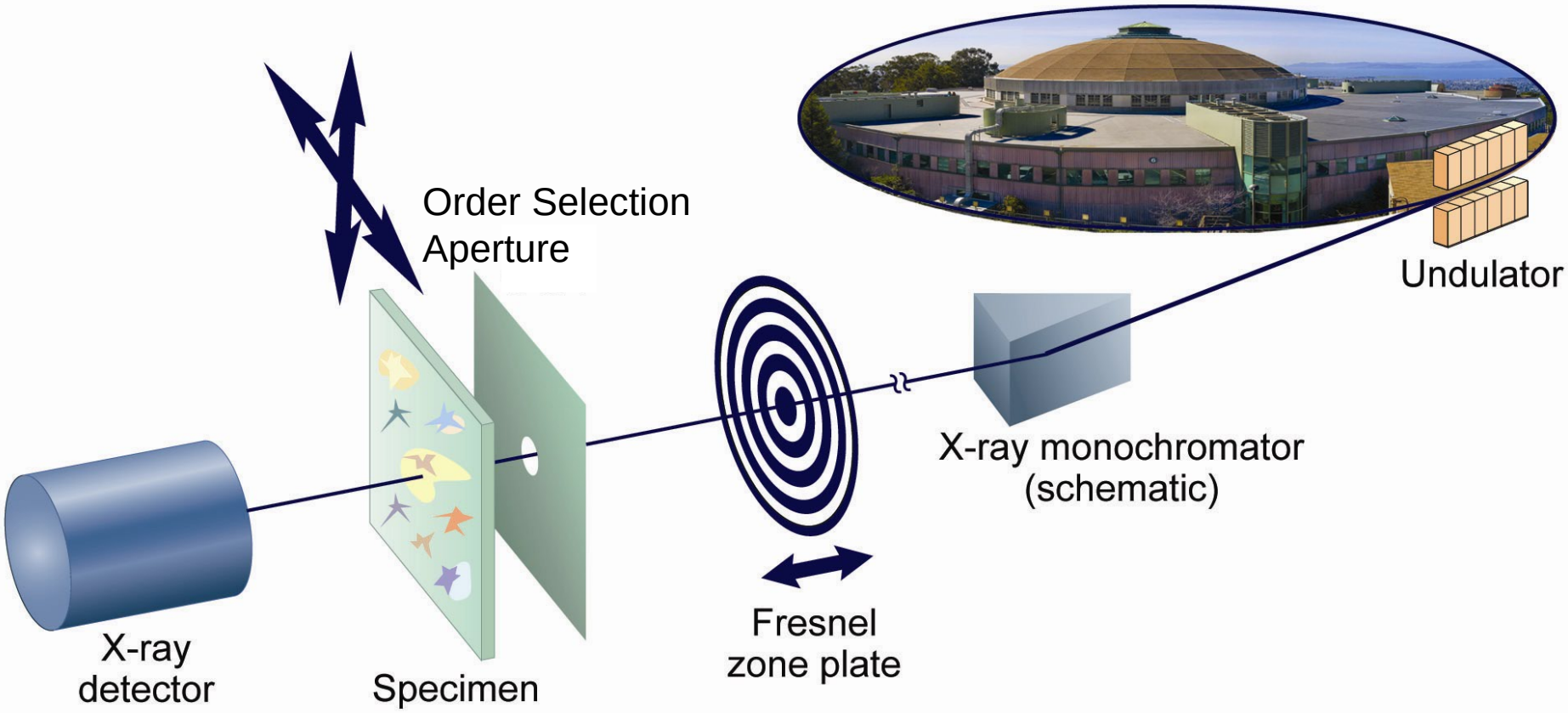
Conclusions from CARES via Comparison with Mexico City (MILAGRO)

- Trends between T0 and T1 in Central California (CARES) is similar to those observed in Mexico City (MILAGRO)
- Total single particle carbon in during CARES was 2-3 times lower than in Mexico City
- Aging processes expected to be different (biogenic/anthropogenic interactions)
- Organic Carbon in Mexico City had significantly more C=C (%sp^2~0.12) than California (%sp^2~0.05)
- Fraction of soot particles identified by STXM not significantly different between Mexico City and Sacramento.

Acknowledgements

- Mary Gilles, Alex Laskin, and Rahul Zaveri
- DOE ASR Program for funding + Ashley Williamson
- Tobias Henn, Tobias Rodel, Peter Sprau (DAAD exchange program)
- Shruti Prakash, BingBing Wang, Steve Kelly, Greg Caroll, Florent Karsenty
- Entire DOE ASR Team for field study logistics.

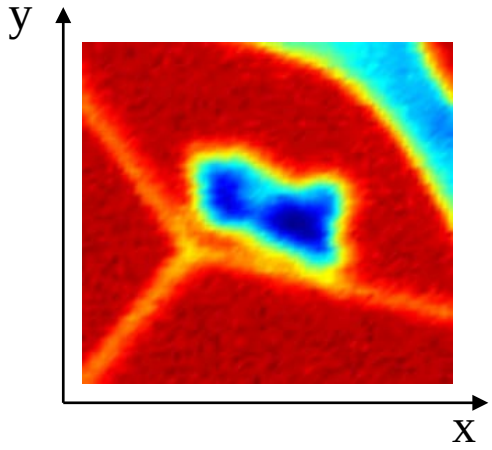
Scanning Transmission X-Ray Microscopy (STXM)

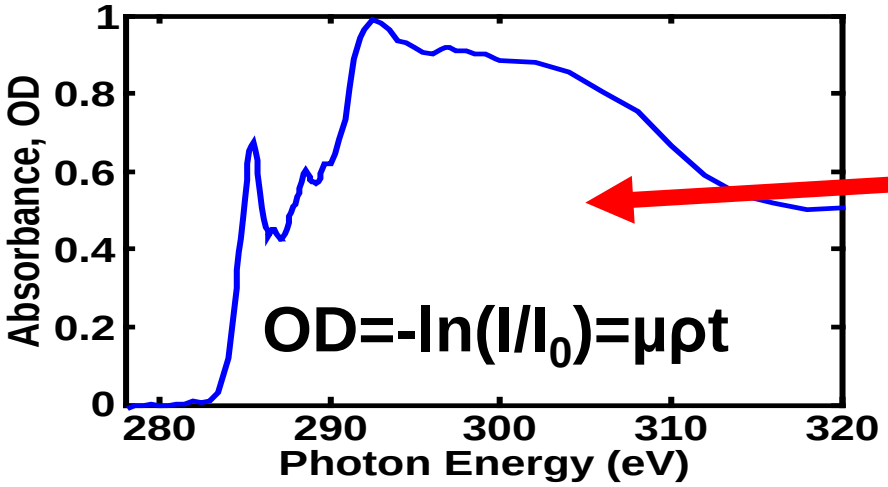


Data Acquisition Modes

- **Image at a single energy:**

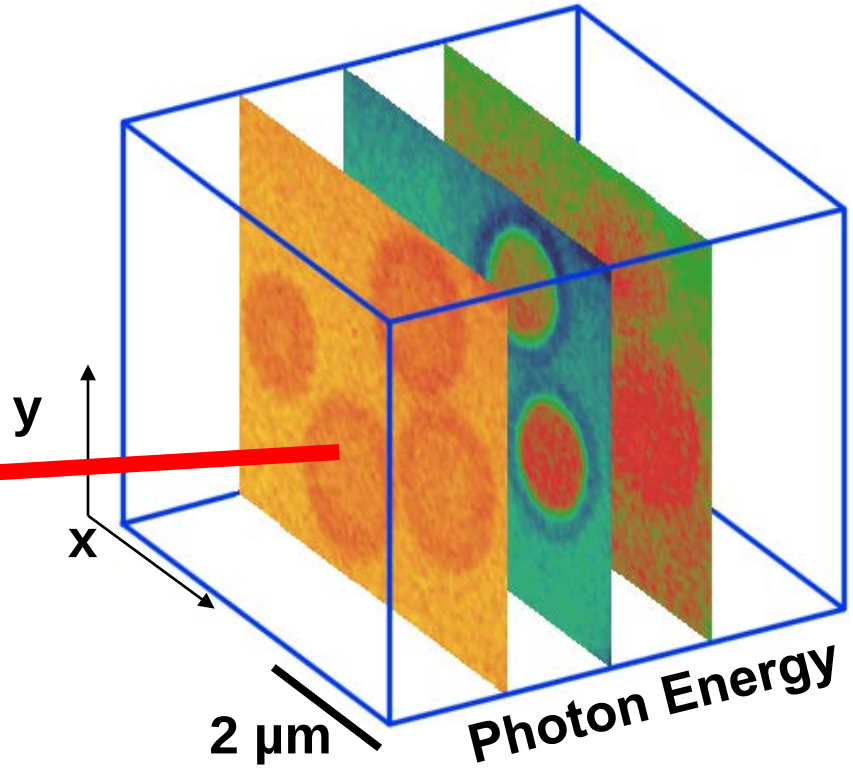
raster scan sample at a fixed energy



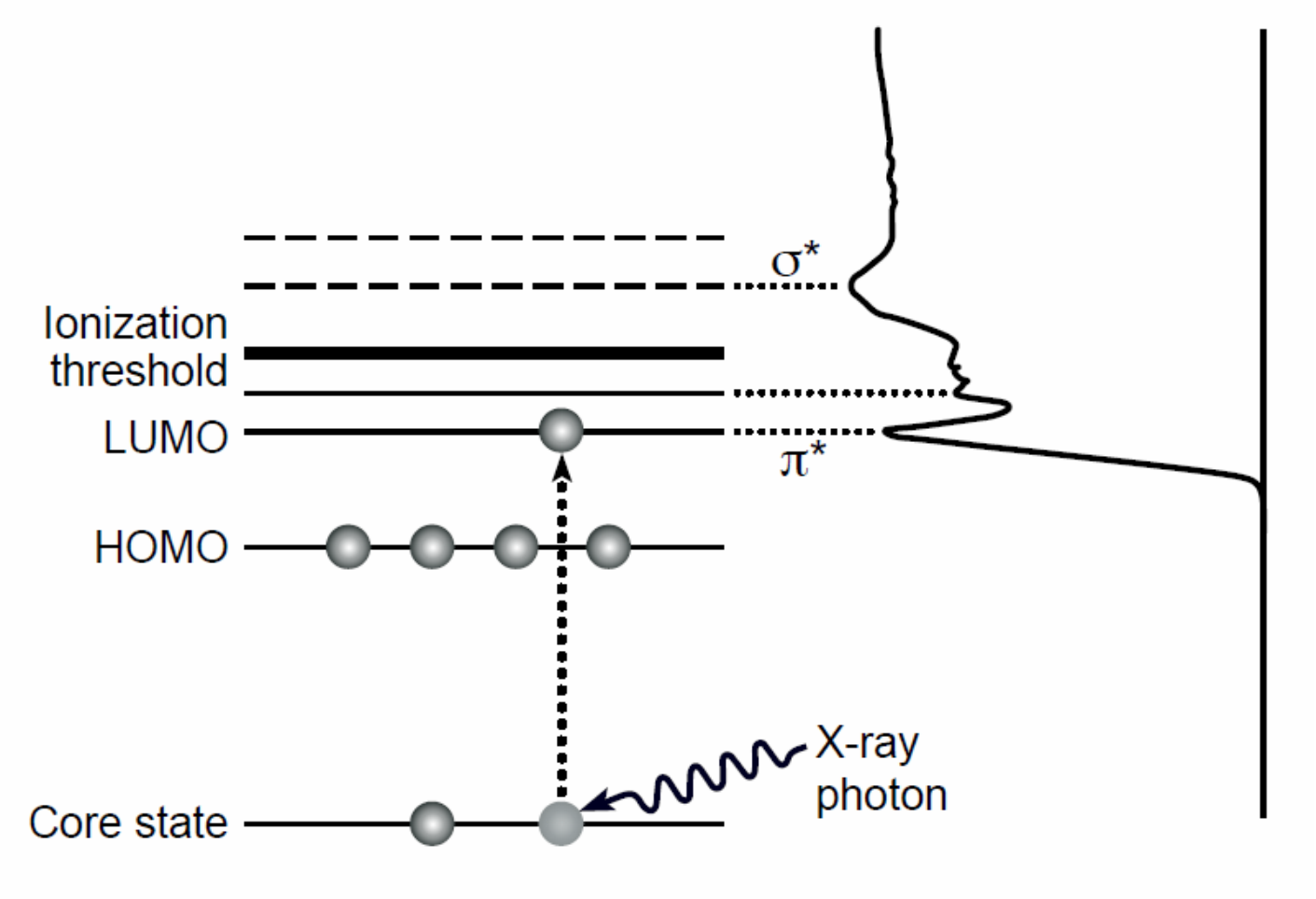


- **Stack:**

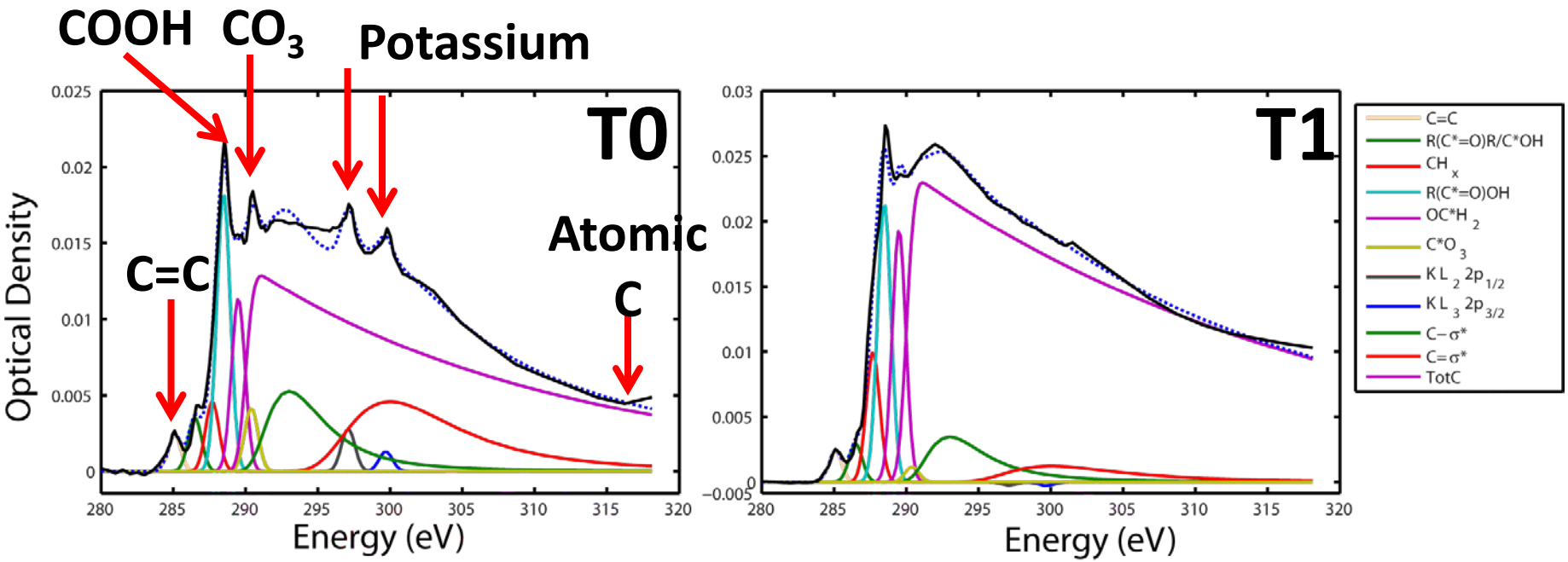
Images as a function of energy→
a spectrum at each image pixel or
a region of interest



Near Edge X-Ray Absorption Spectroscopy (NEXAFS)



Particle Averaged Carbon Speciation



From the T0 perspective:

more potassium (biomass burning/seasalt/dust)

More CO_3 (sea salt/aqueous?)

less carbon (Its T0 for a reason ☺)

less C=C

More pronounced COOH

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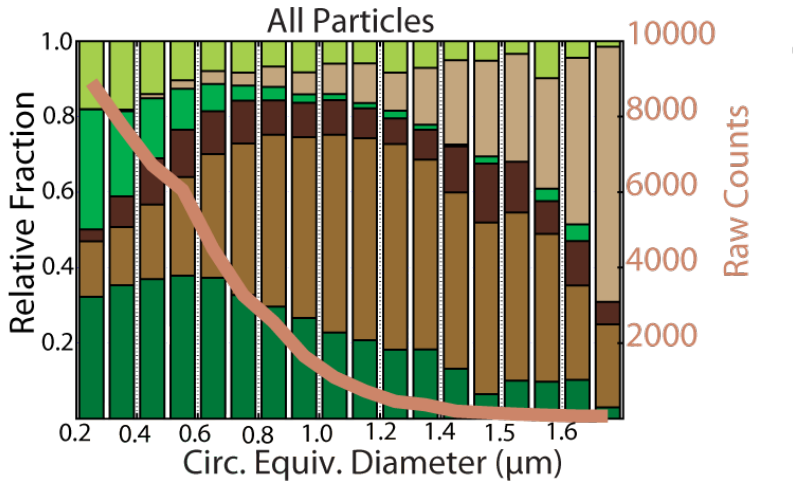

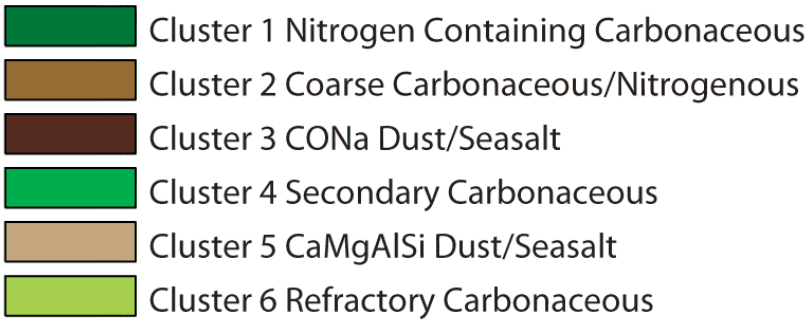

Green particle types: small and probably mostly organic

Brown particle types: larger and dominated by inorganic tracers

More inorganic types at T0 (similar to Mexico City)

More organic particle types at T1, where more biogenic VOCs reside