# Optical and Cloud Nucleating Properties of Aerosols Measured onboard R/V Atlantis during CalNex

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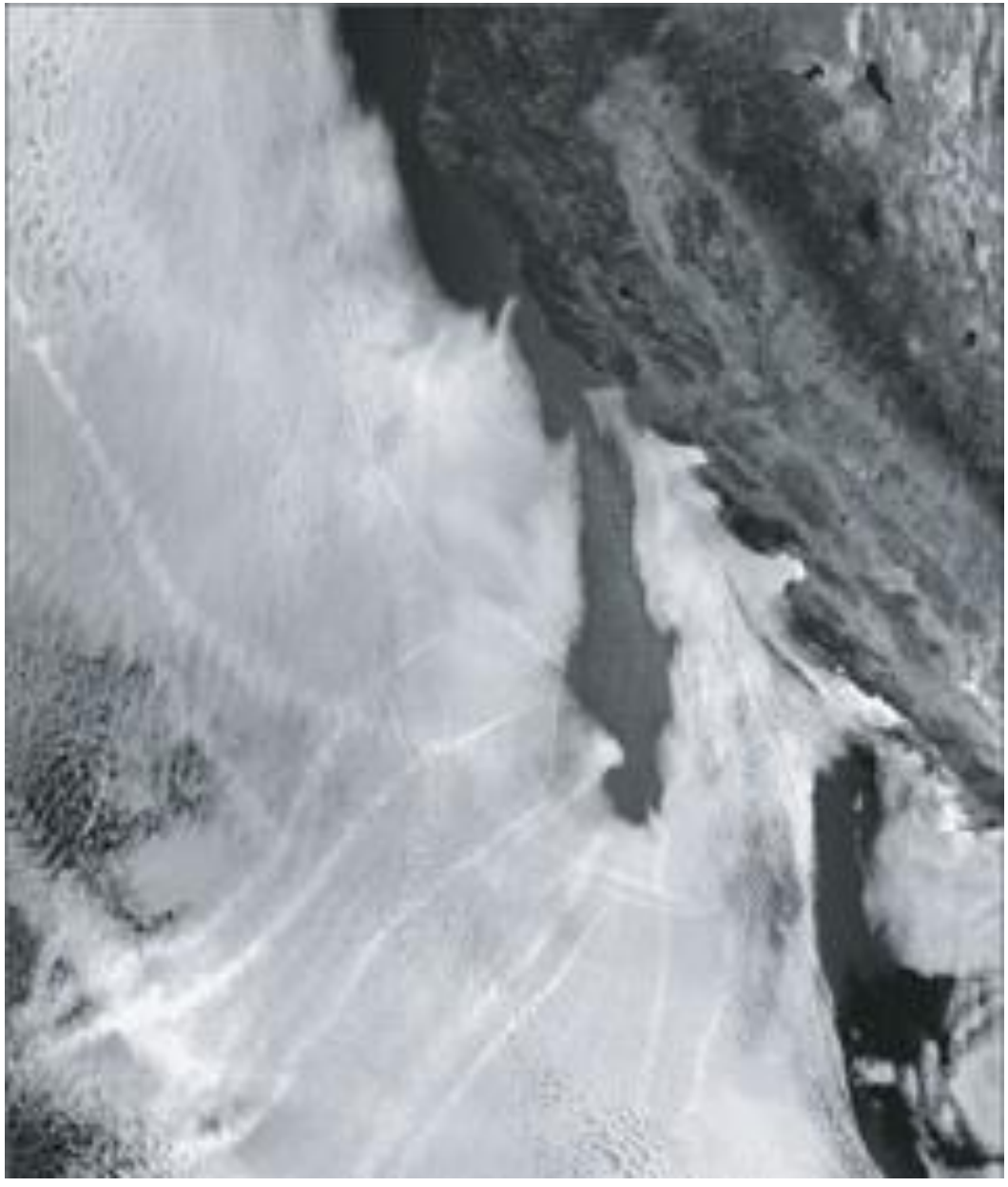
(NOAA PMEL)

# Impact of particle size and composition on cloud microphysics and albedo



Studying the aerosol that is entrained into clouds, how the clouds respond, and all factors affecting the cloud response is essential for improving the understanding of radiative forcing by aerosols.

Aerosol sources and aging processes

↓

Particle size and composition

↓

Ability of a particle to nucleate a cloud droplet

# Impact of particle size and composition on regional aerosol direct radiative forcing

Measurements of the response of aerosol optical properties to changes in particle composition are required for an improved understanding of aerosol direct radiative forcing.

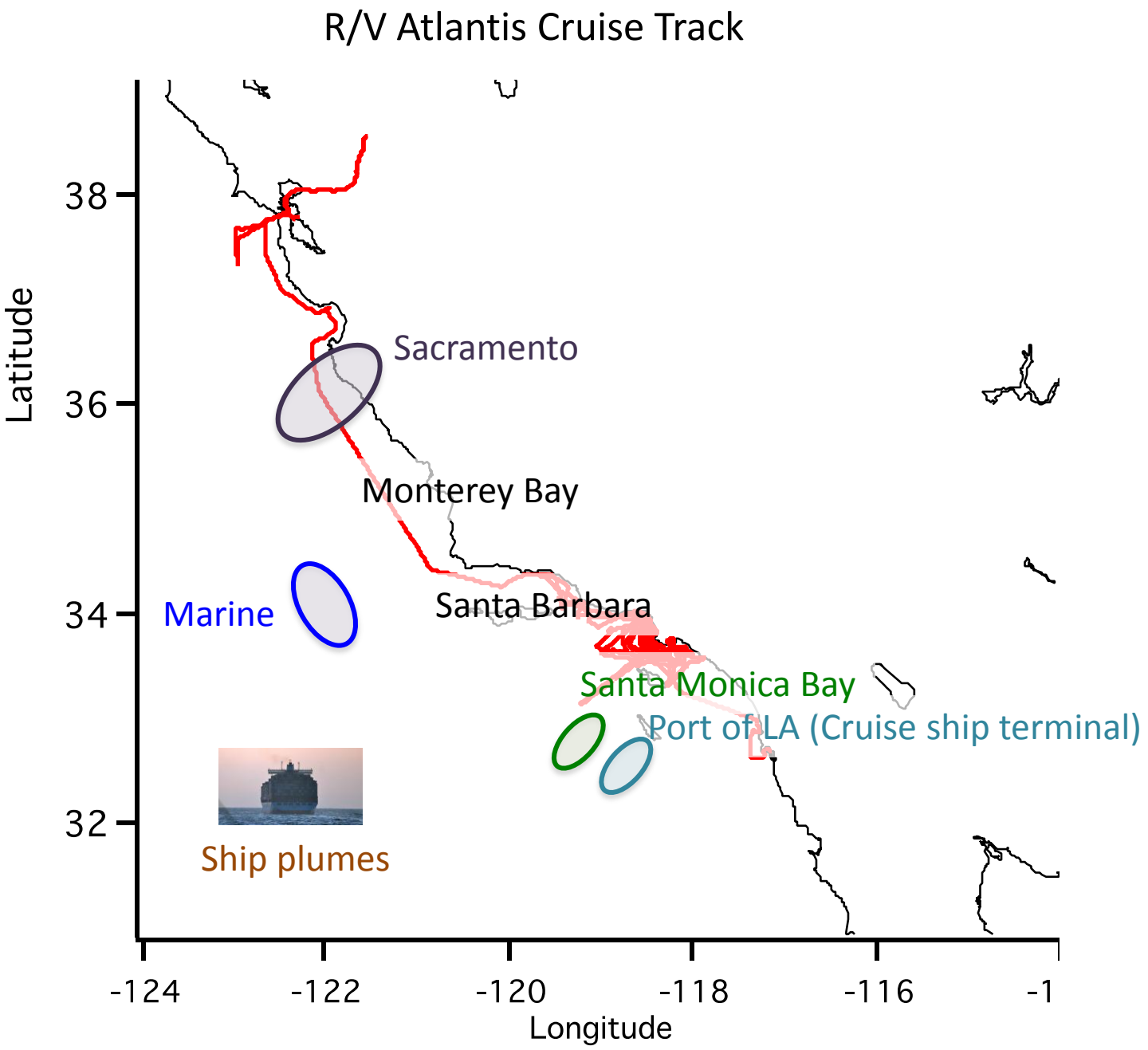
Aerosol sources and aging processes

↓

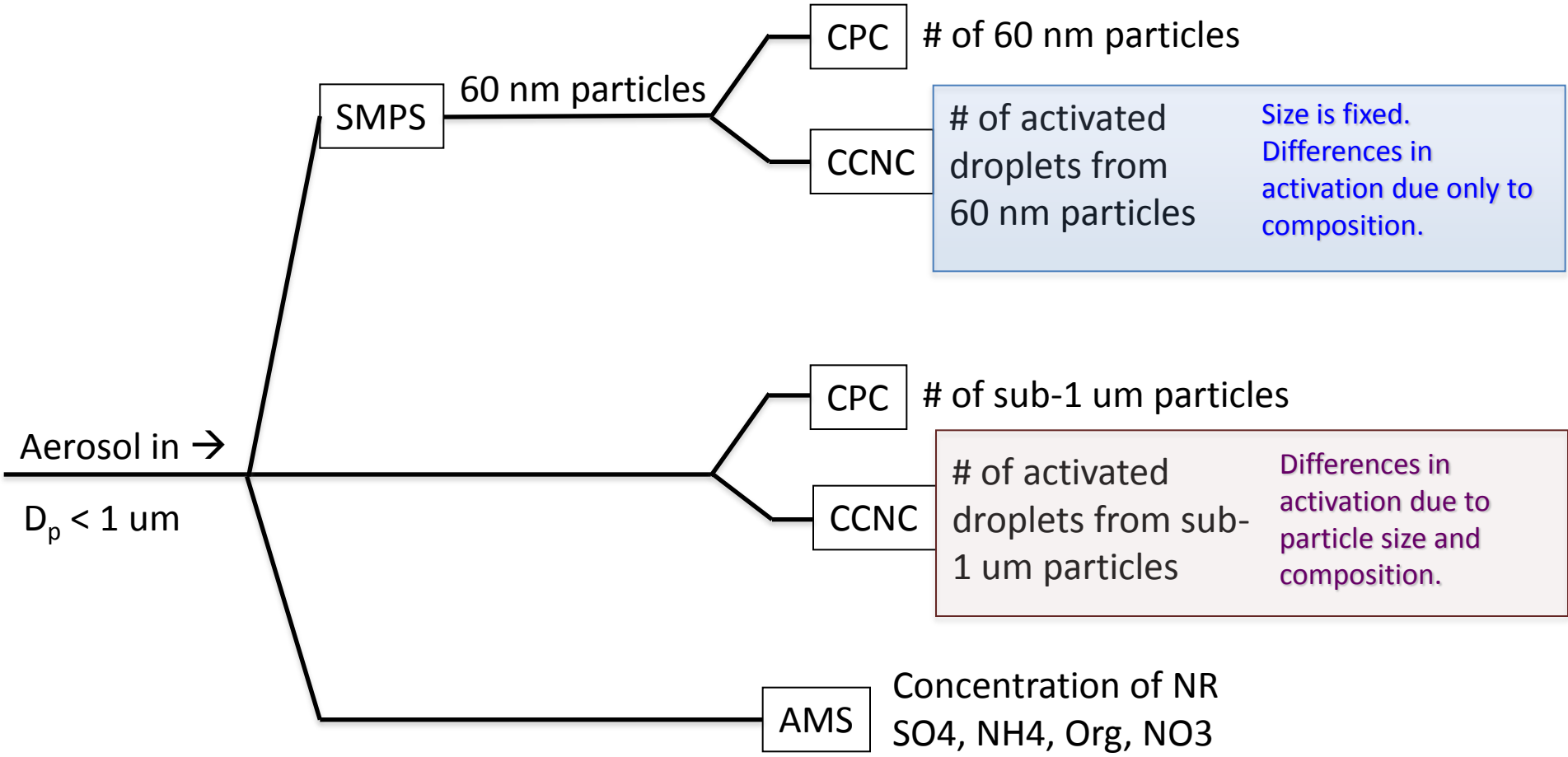
Particle size and composition

↓

Dependence of aerosol light scattering on RH

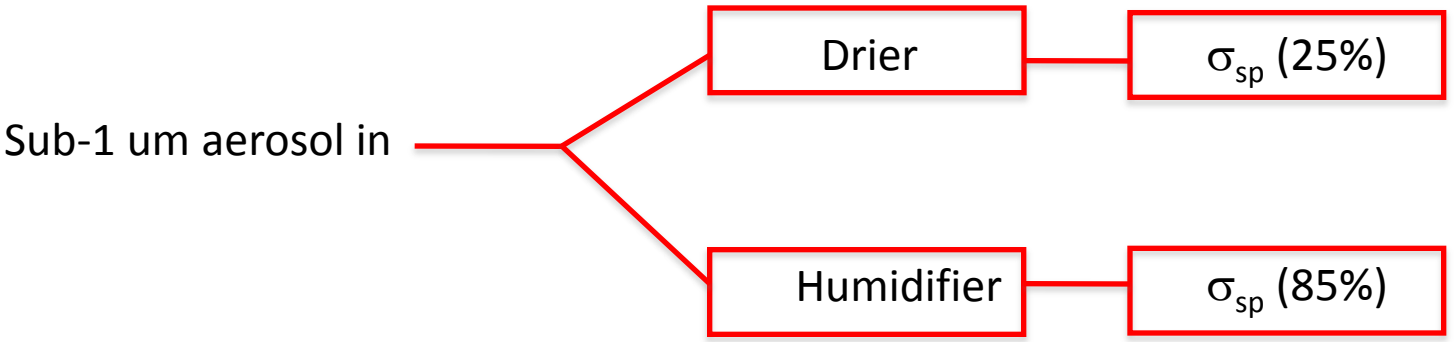

R/V Atlantis Cruise Track
Latitude
38
36
34
32
Sacramento
Monterey Bay
Marine
Santa Barbara
Santa Monica Bay
Port of LA (Cruise ship terminal)
Ship plumes
-124
-122
-120
-118
-116
-1
Longitude

# Measurements to assess the dependence of cloud drop activation on particle size and composition



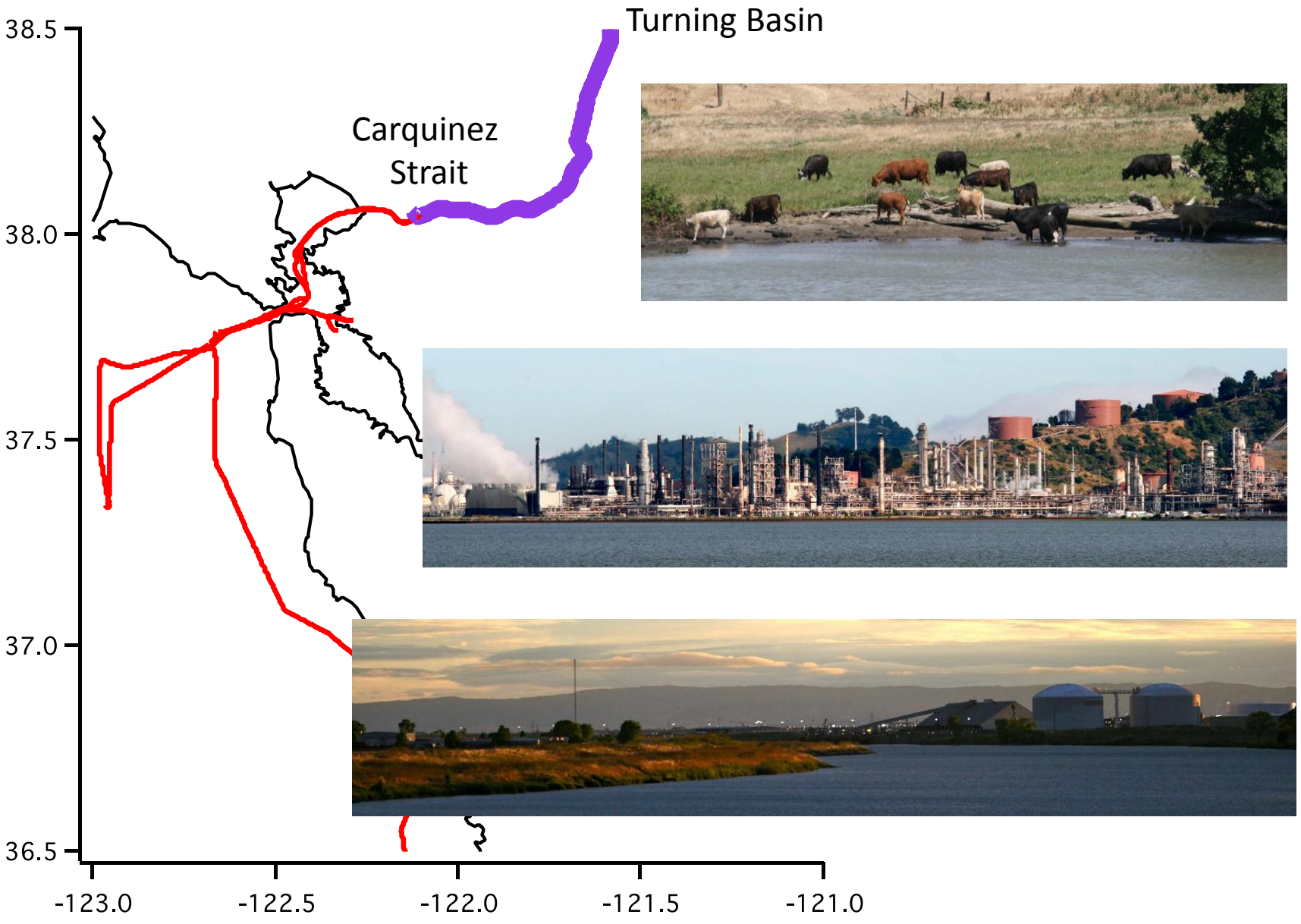

- Activation ratio (CCN / CN) for 60 nm and sub-1 um aerosol
- Supersaturation = 0.3, 0.4, 0.5, 0.6, and 0.7%

# Measurements to assess the impact of particle composition on the relative humidity dependence of scattering

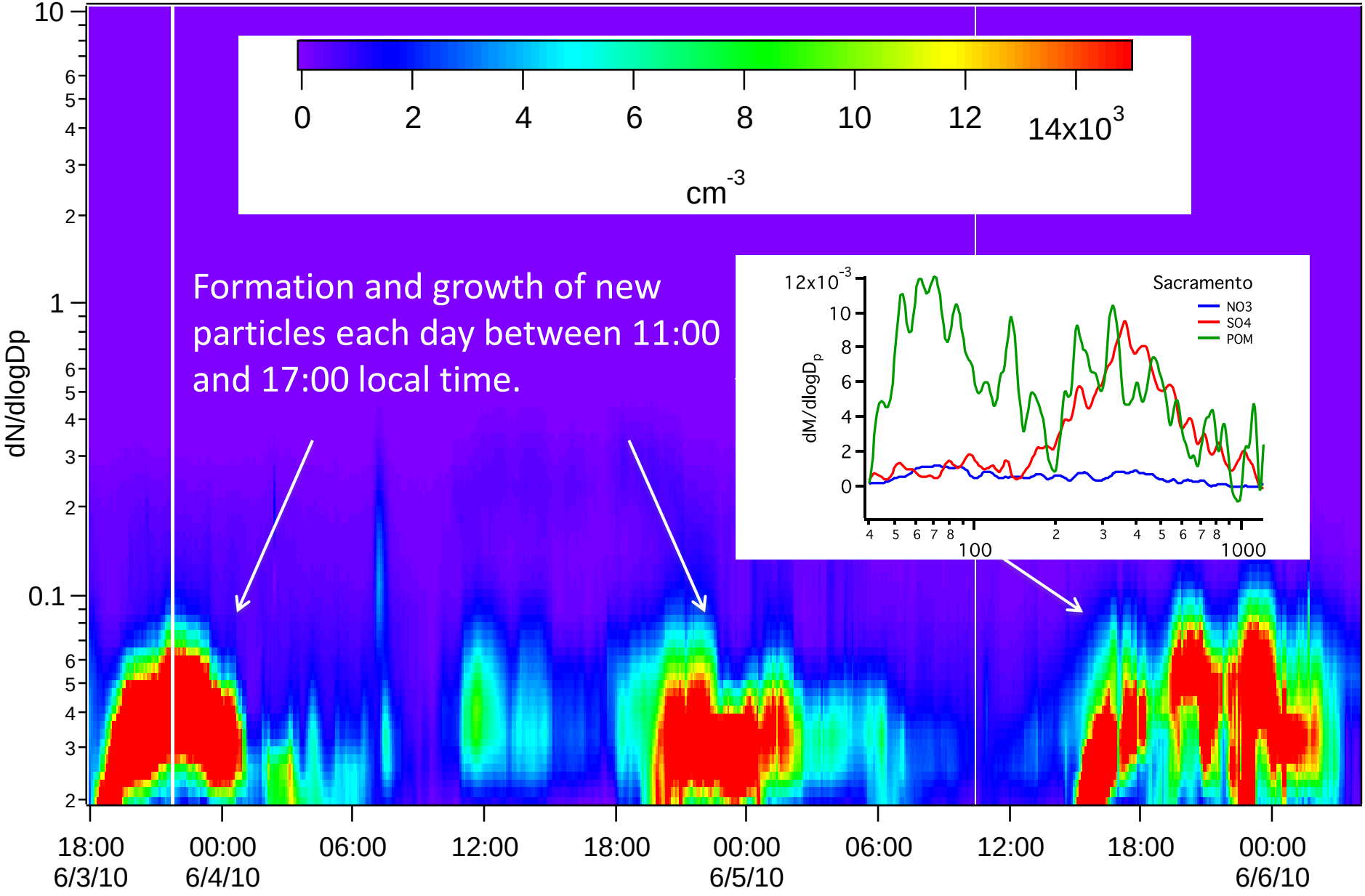


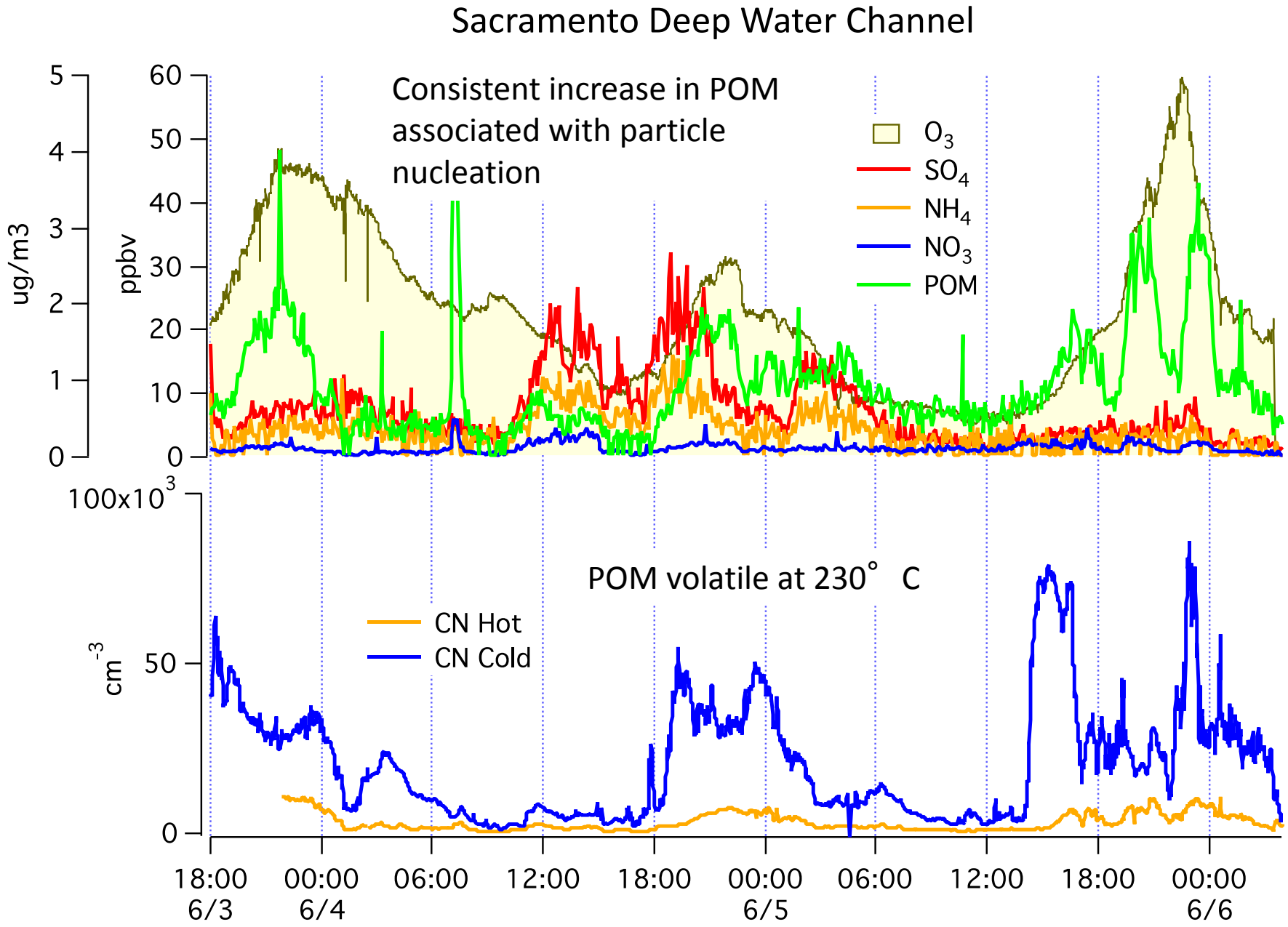

$$fs_{sp}(RH) = \frac{s_{sp}(RH)}{s_{sp}(RH_{ref})}$$

Sacramento Deep Water Channel



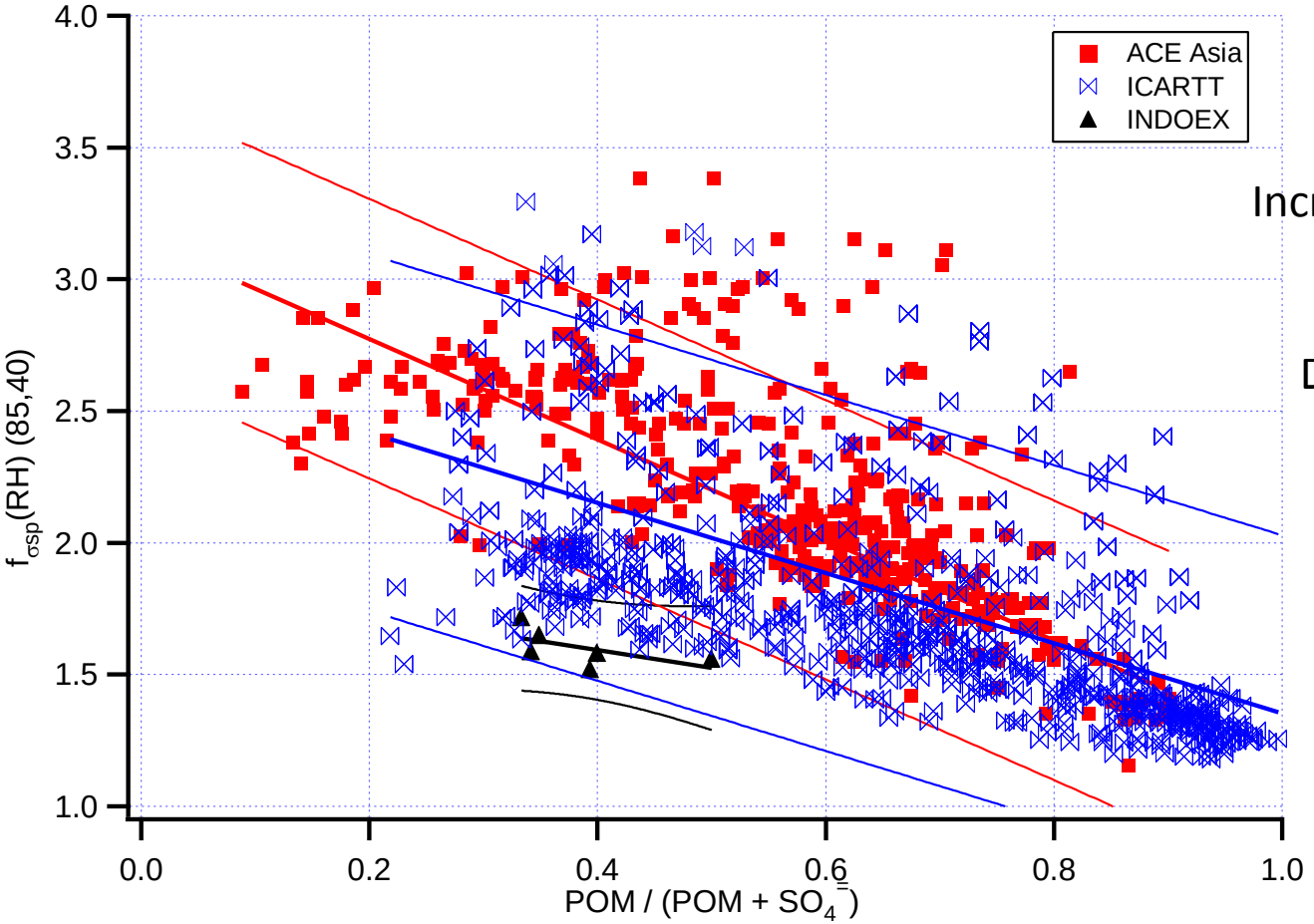
# Sacramento Deep Water Channel
# Particle Number Size Distribution




Sacramento Deep Water Channel
Consistent increase in POM associated with particle nucleation
O3
SO4
NH4
NO3
POM
ug/m3
ppbv
POM volatile at 230° C
CN Hot
CN Cold
100x10^3
cm^-3
18:00 6/3
00:00 6/4
06:00
12:00
18:00
00:00 6/5
06:00
12:00
18:00
00:00 6/6

# Impact of aerosol composition on f(RH):

## Previous results from ACE Asia, ICARTT, and INDOEX




Increase in POM mass fraction

↓

Decrease in water uptake

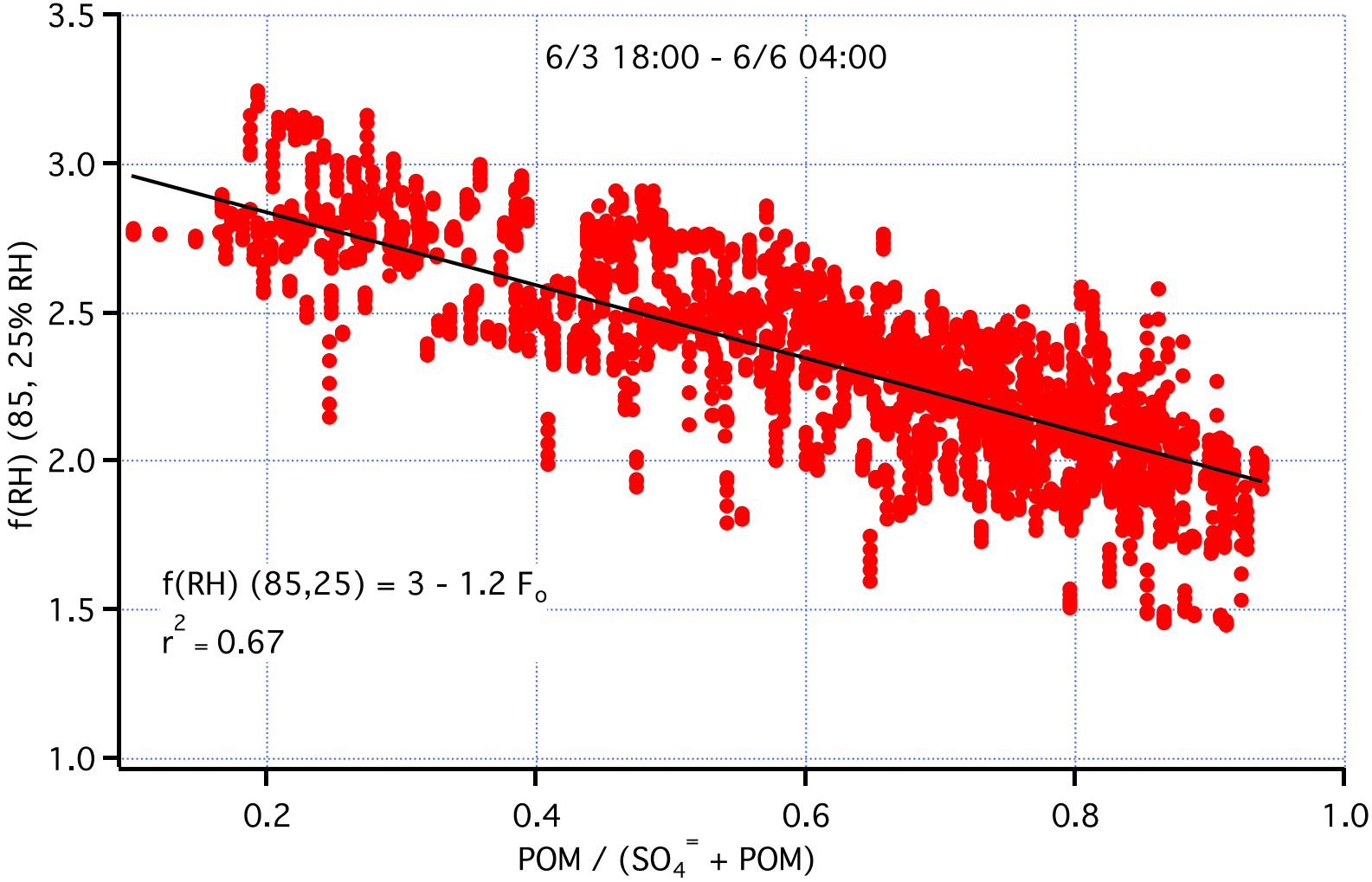
↓

Decrease in f(RH)

Empirically-based parameterization that quantifies the POM mass fraction – f(RH) relationship for use in radiative transfer and air quality models either as input or as validation.

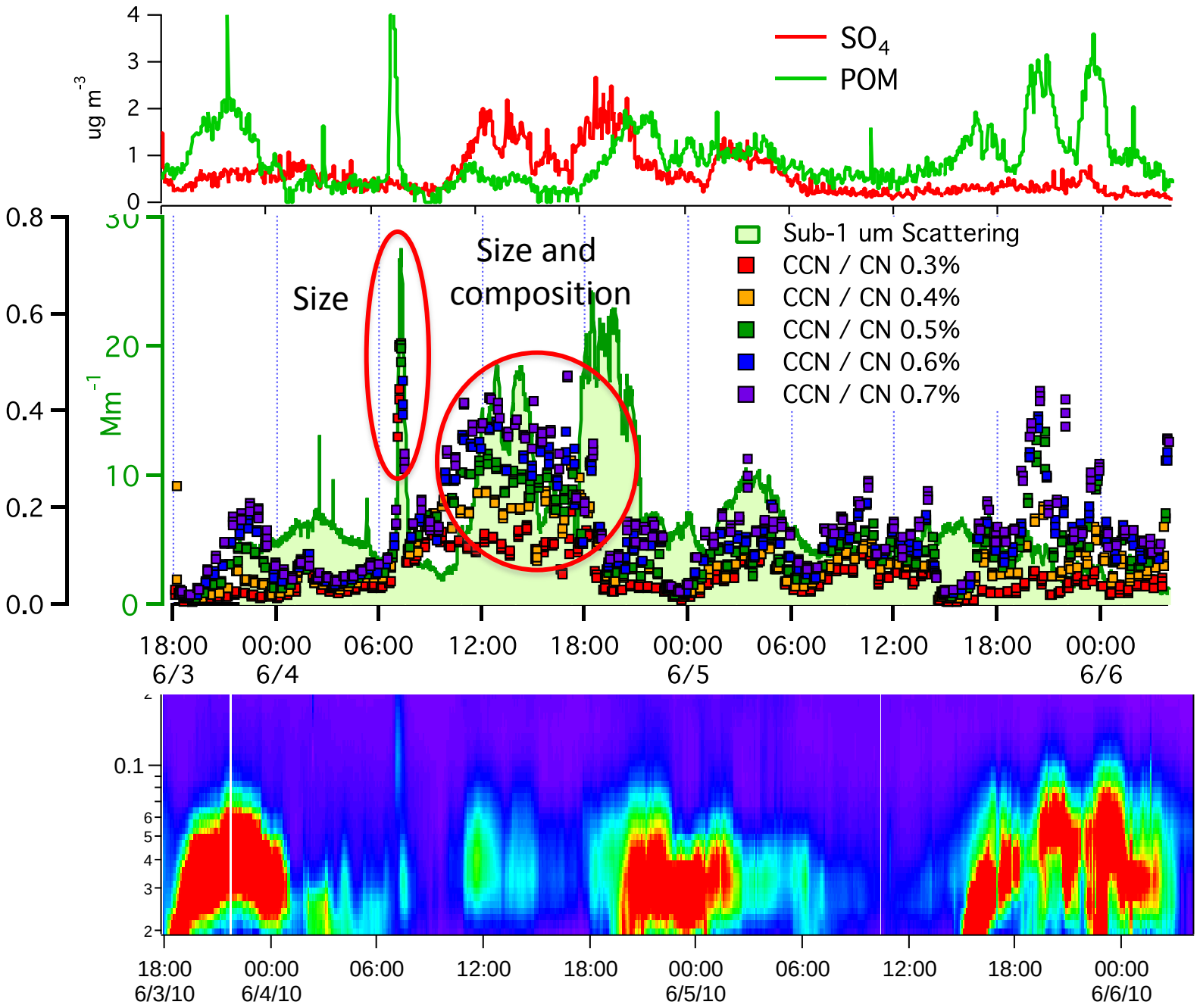
Quinn et al., 2005.

# Sacramento Deep Water Channel
## Dependence of f(RH) on Aerosol Composition



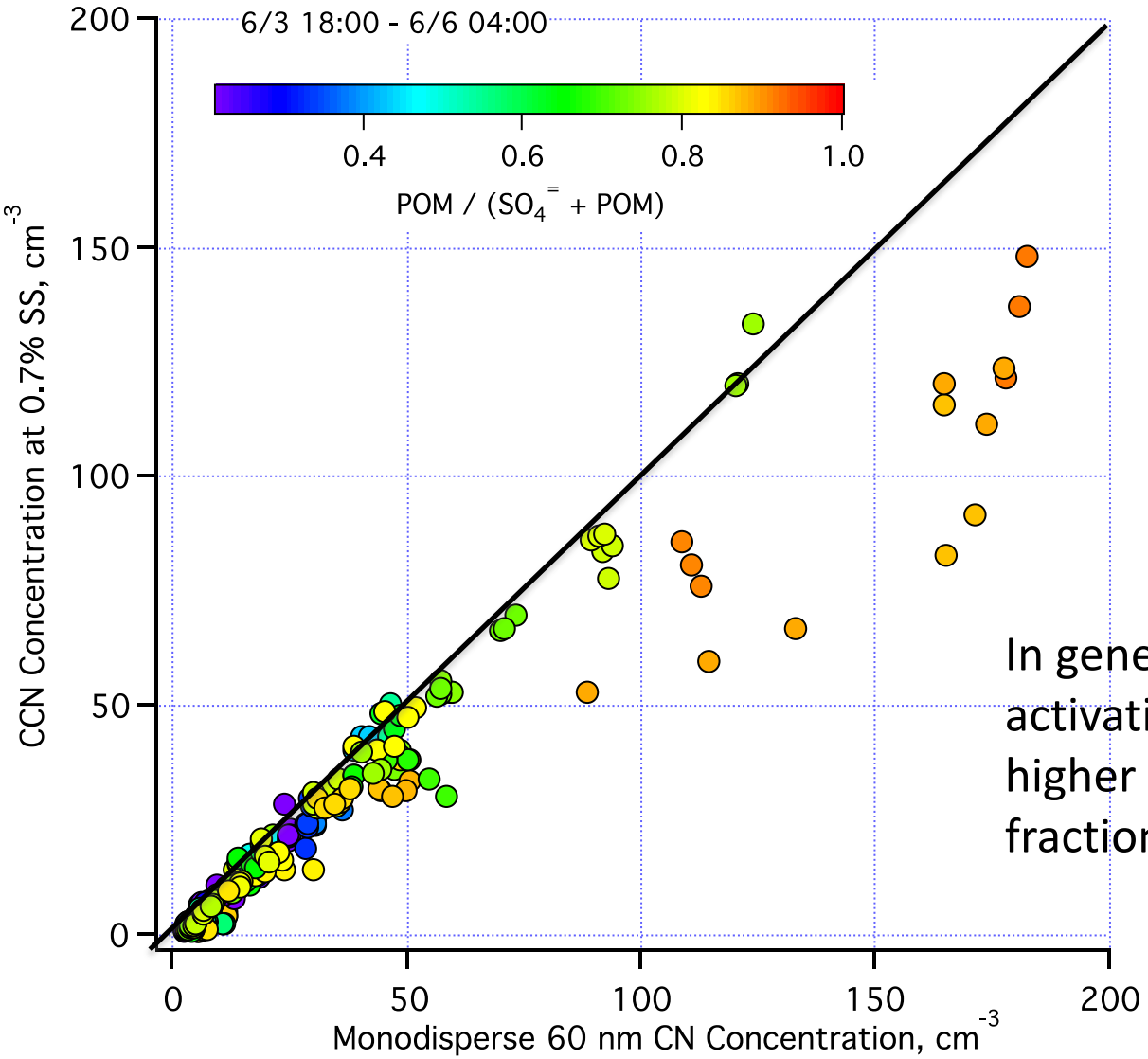

70 % of the variance in f(RH) can be explained by chemical composition.
Need to also assess impact of particle size.

# Sacramento Deep Water Channel: CCN Activation Ratio for $D_p$ < 1 um



# Sacramento Deep Water Channel

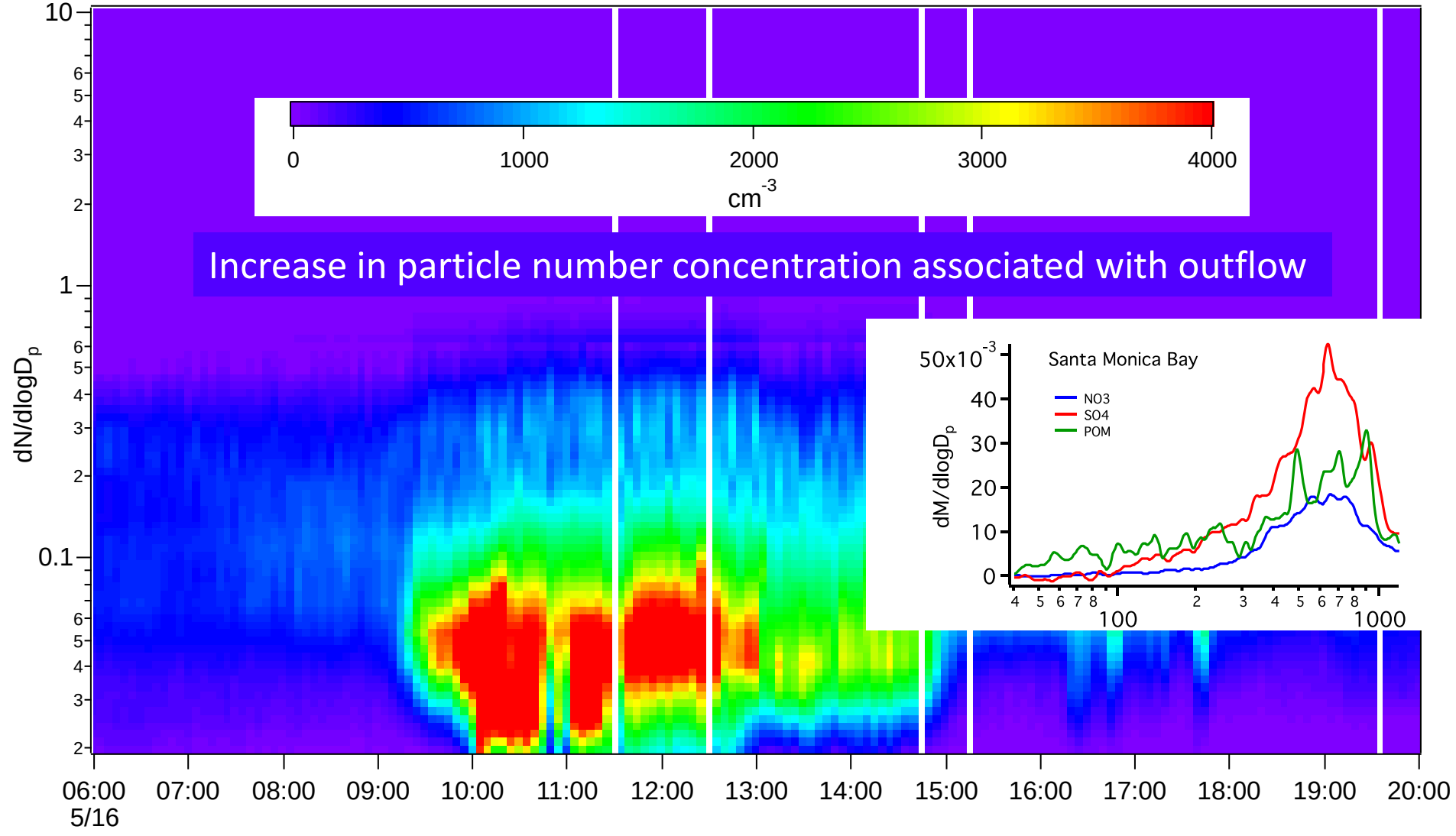
## CCN Activation Ratio for $D_p$ = 60 nm as a function of POM mass fraction

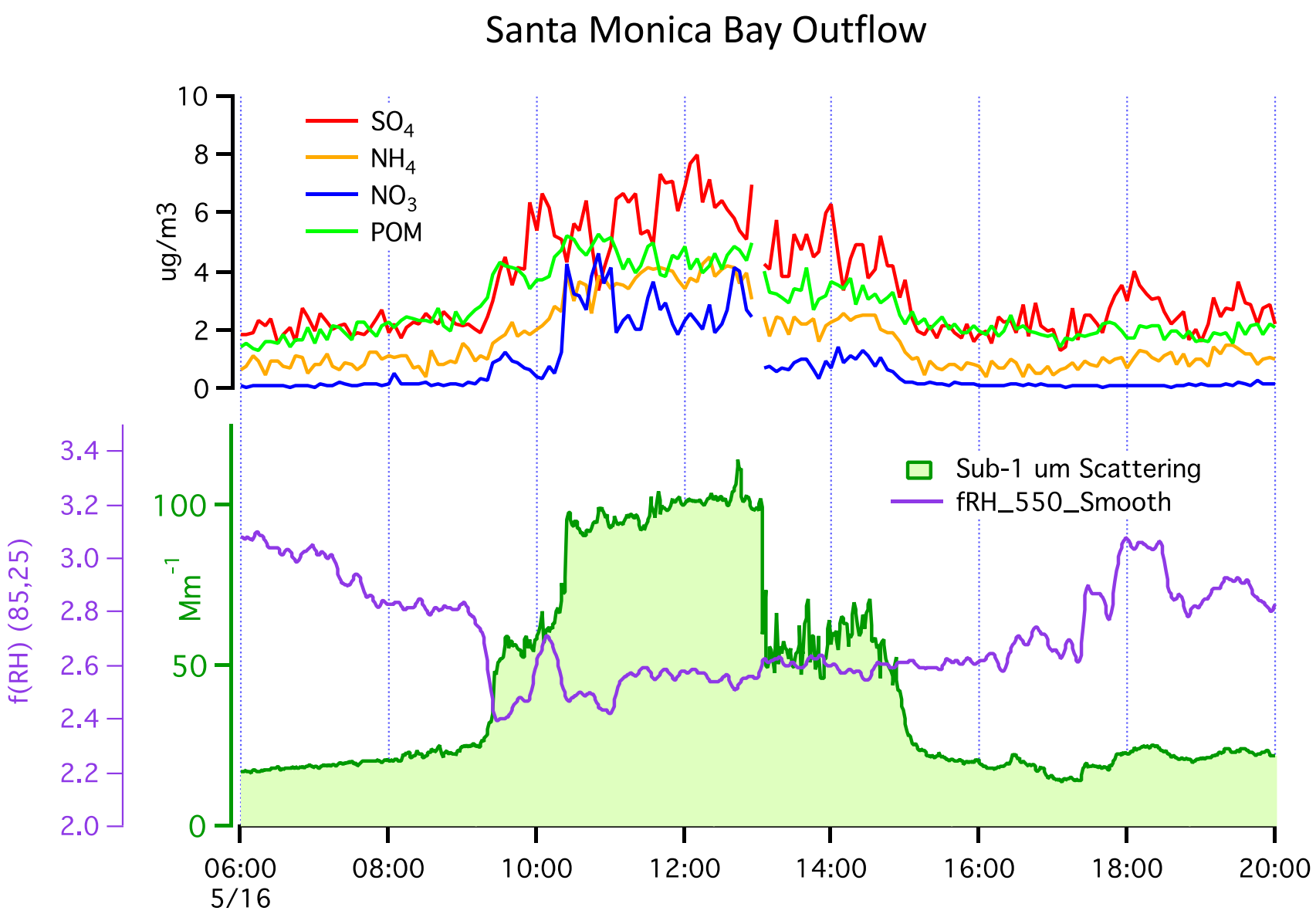


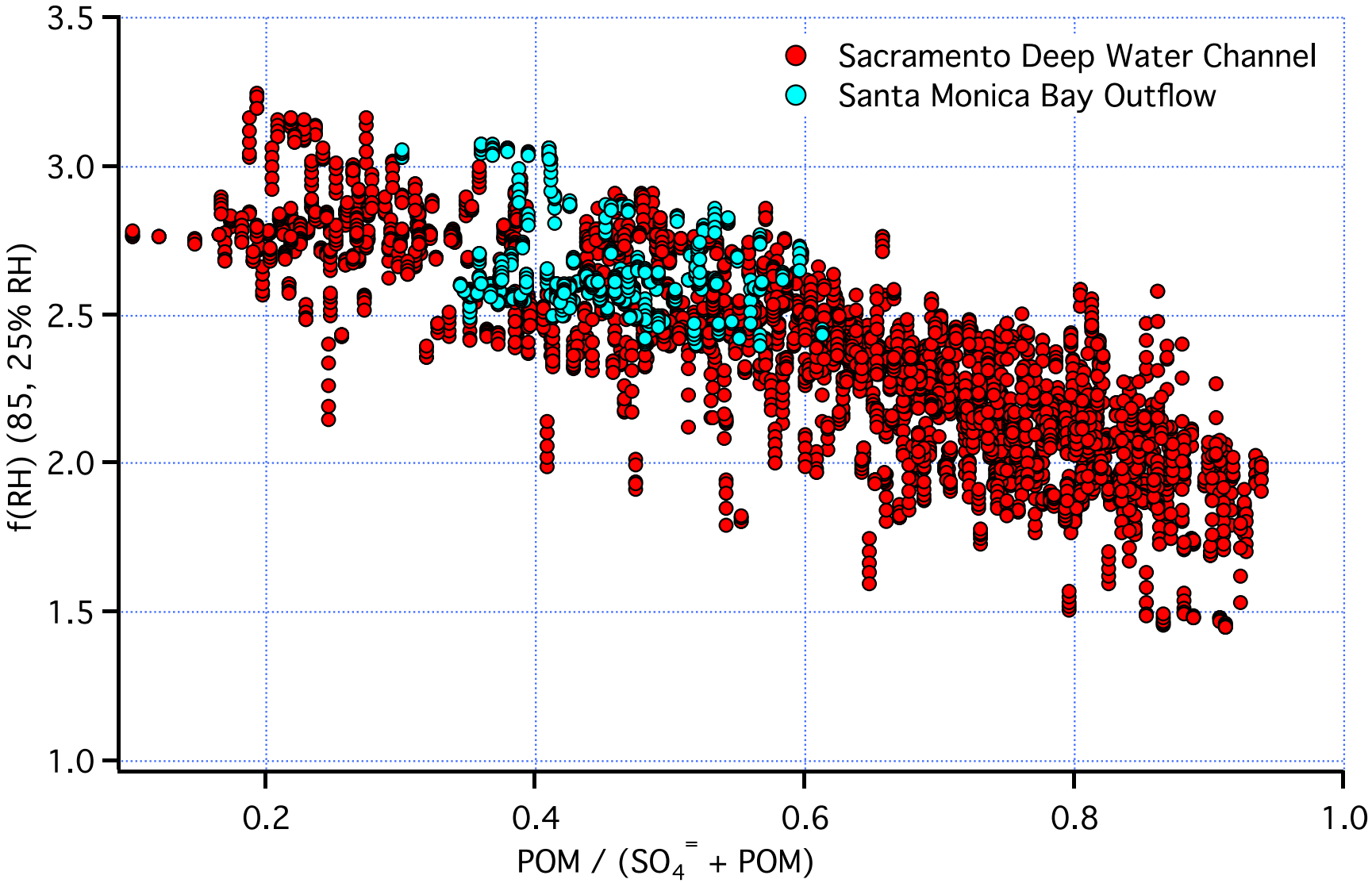

In general, less activation with a higher POM mass fraction

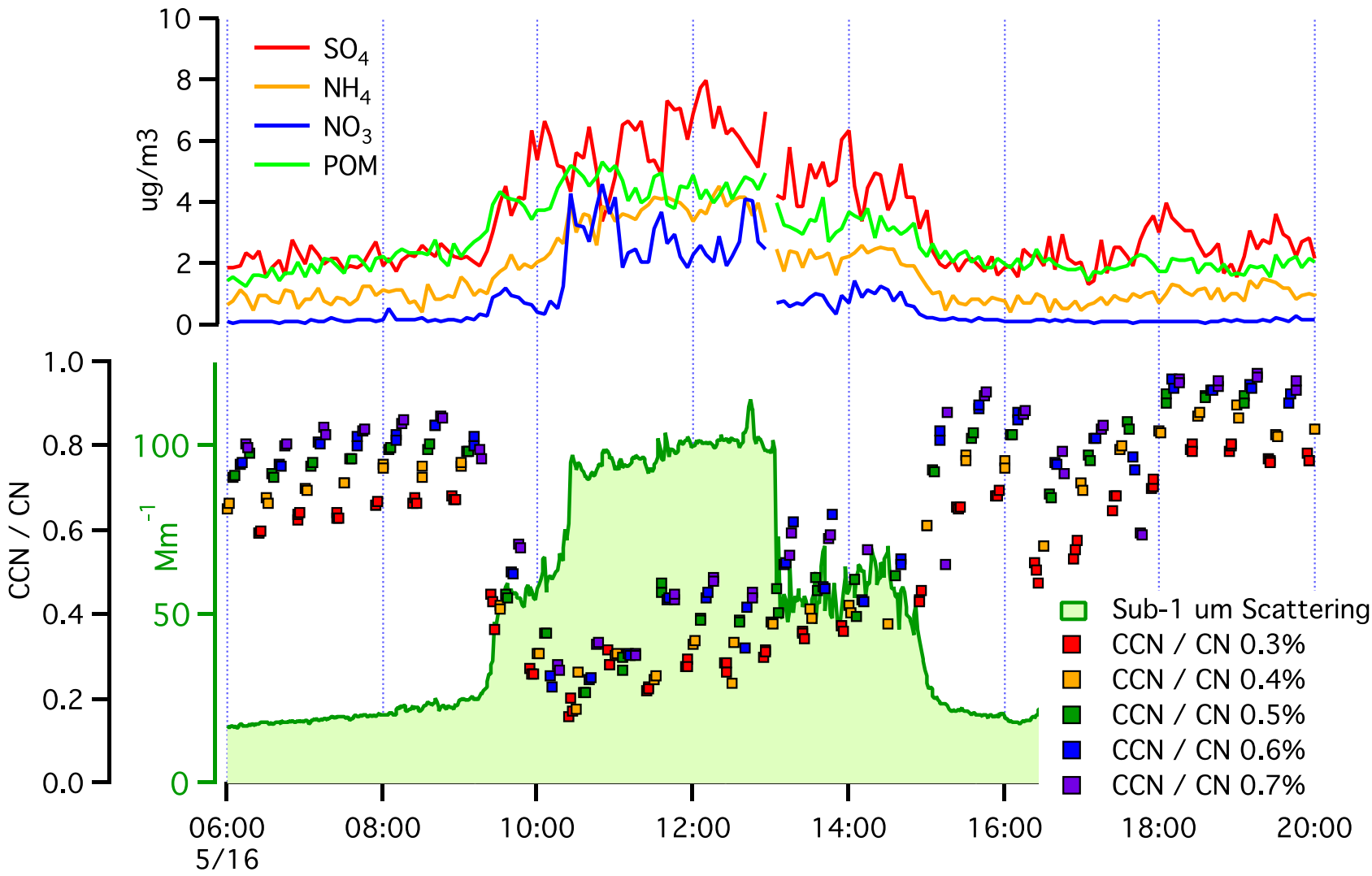

Santa Monica Bay Outflow
Typically see outflow starting early in the morning and tapering off around 10:00 am (local time)
34.2
34.0
33.8
33.6
-118.6
-118.4
-118.2
-118.0
6:00 PM
3:00 PM
12:00 PM
9:00 AM
5/16/10 6:00 AM

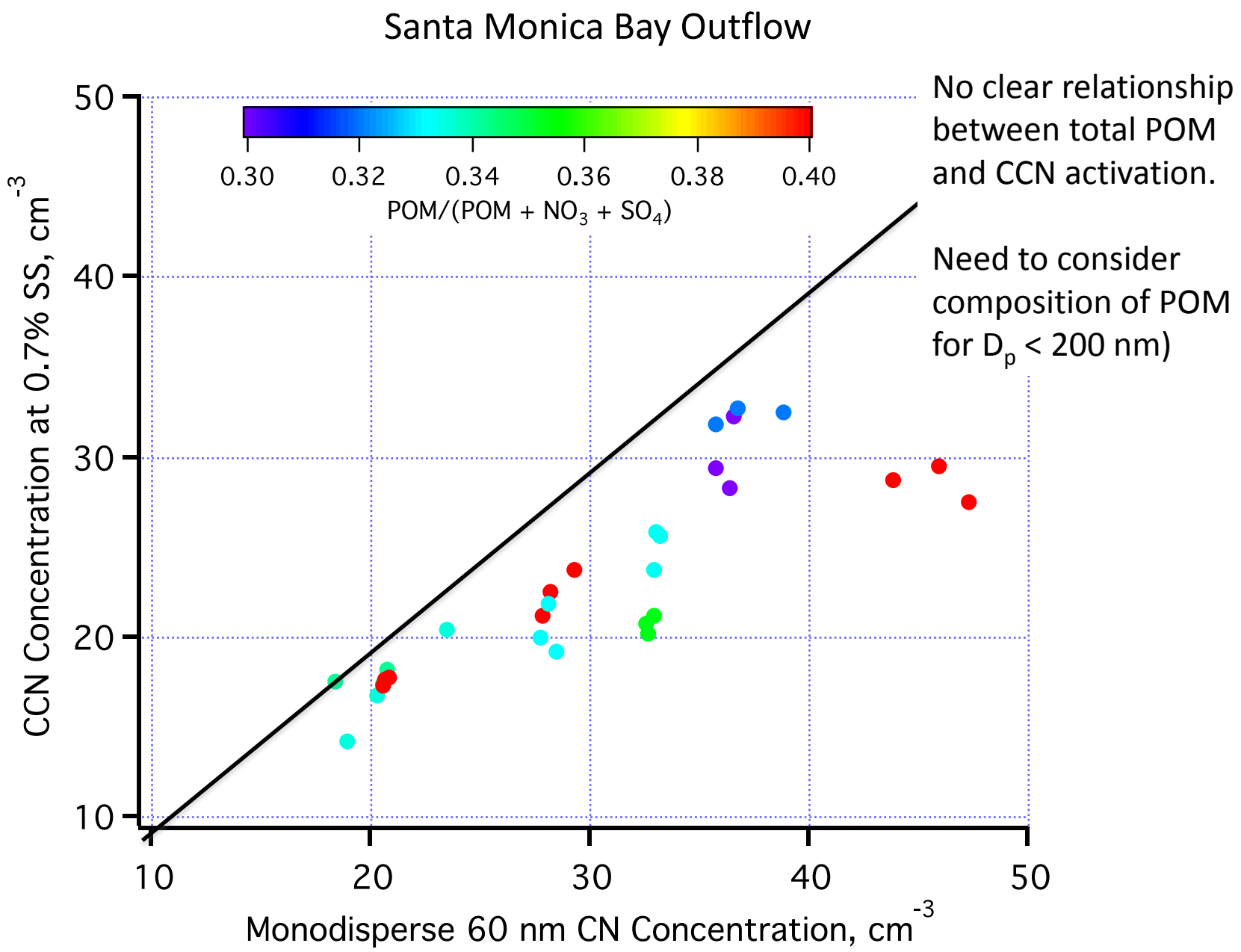
# Santa Monica Bay Outflow

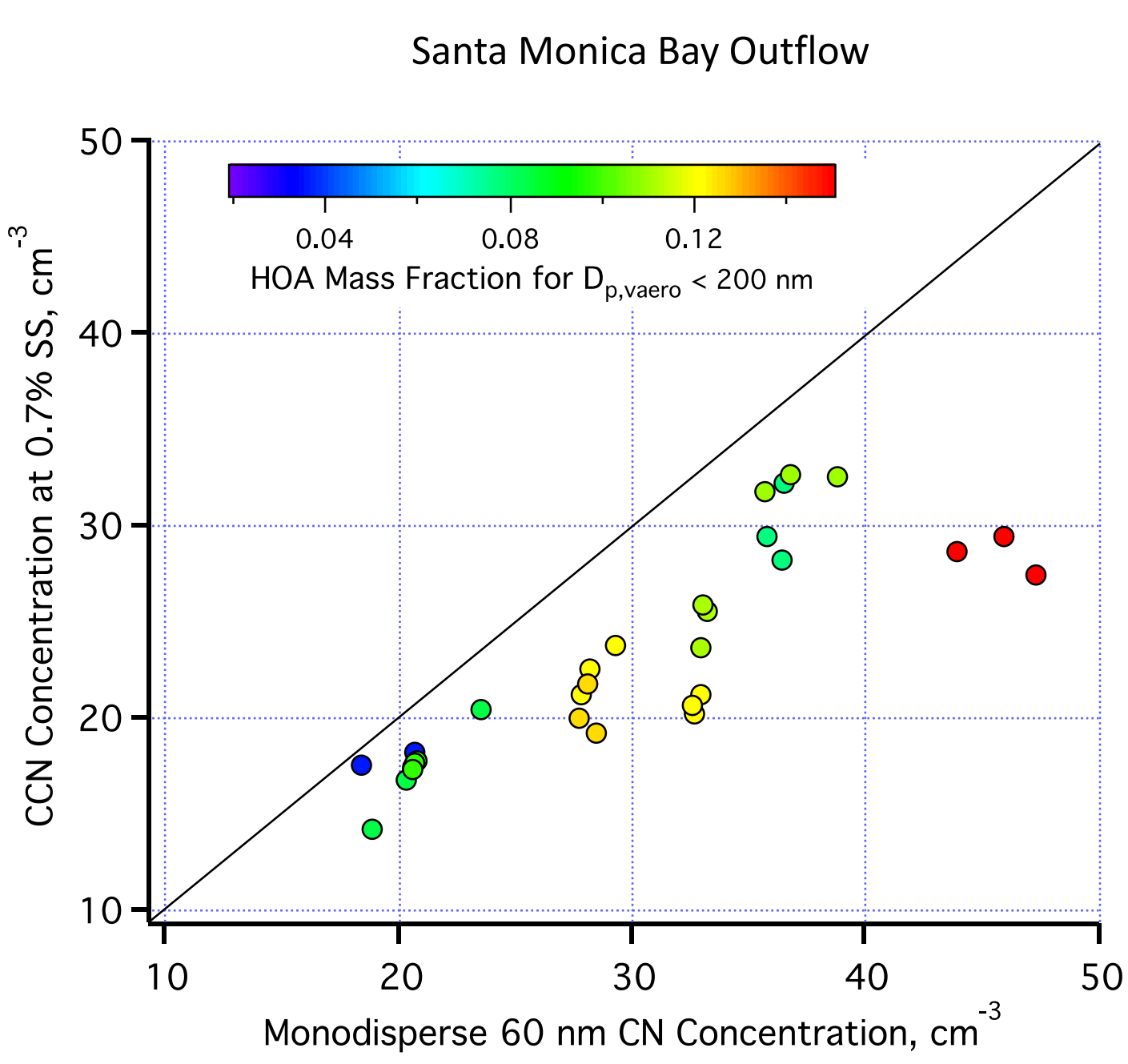



Santa Monica Bay Outflow
ug/m3
10
8
6
4
2
0
SO4
NH4
NO3
POM
f(RH) (85,25)
3.4
3.2
3.0
2.8
2.6
2.4
2.2
2.0
Mm-1
100
50
0
Sub-1 um Scattering
fRH_550_Smooth
06:00
5/16
08:00
10:00
12:00
14:00
16:00
18:00
20:00


Santa Monica Bay Outflow
Dependence of f(RH) on Aerosol Composition
Sacramento Deep Water Channel
Santa Monica Bay Outflow
f(RH) (85, 25% RH)
3.5
3.0
2.5
2.0
1.5
1.0
0.2
0.4
0.6
0.8
1.0
POM / (SO4= + POM)


Santa Monica Bay Outflow
ug/m3
10
8
6
4
2
0
SO4
NH4
NO3
POM
CCN / CN
1.0
0.8
0.6
0.4
0.2
0.0
Mm-1
100
50
0
Sub-1 um Scattering
CCN / CN 0.3%
CCN / CN 0.4%
CCN / CN 0.5%
CCN / CN 0.6%
CCN / CN 0.7%
06:00
5/16
08:00
10:00
12:00
14:00
16:00
18:00
20:00

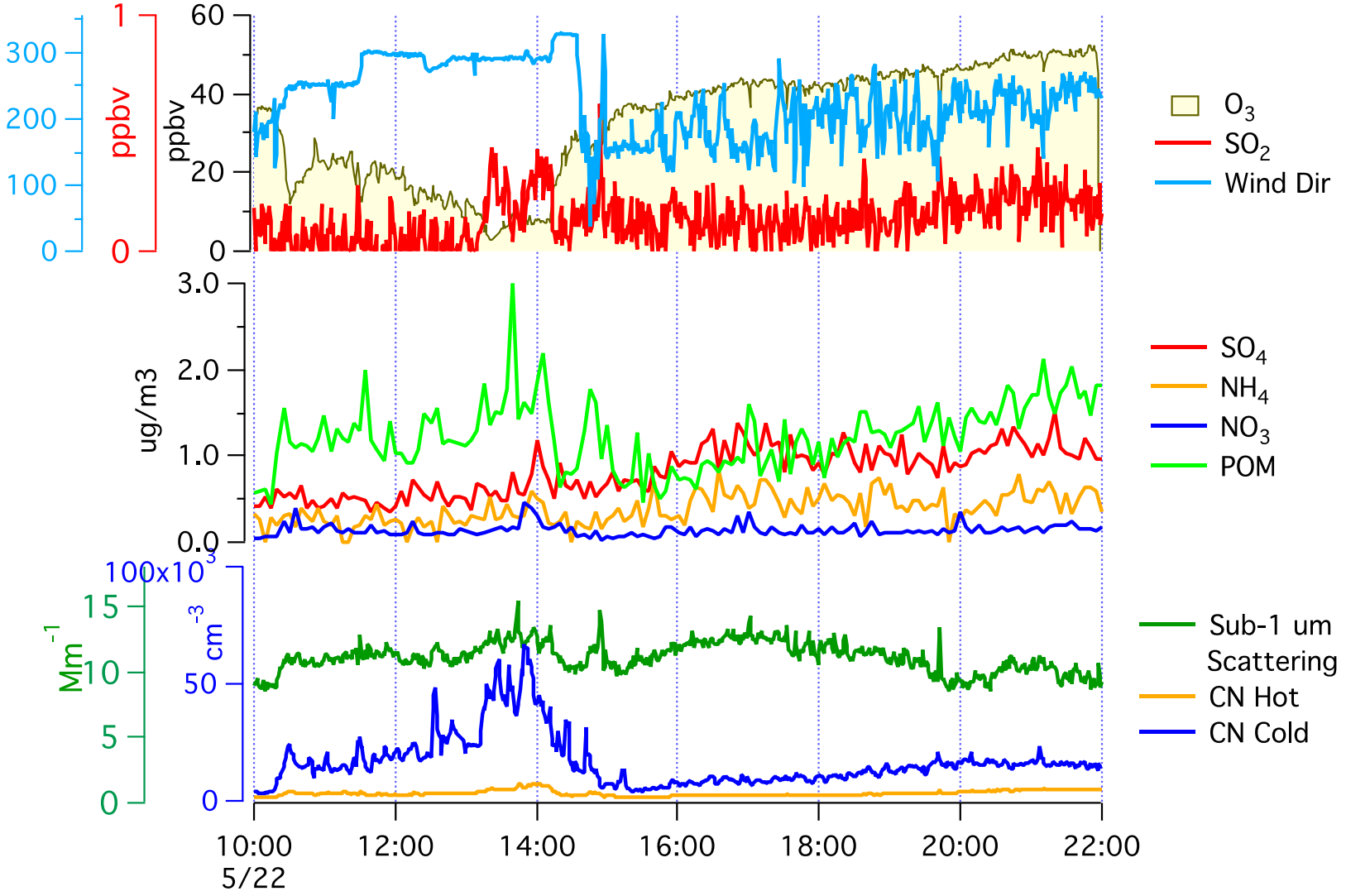

Santa Monica Bay Outflow
No clear relationship between total POM and CCN activation.
Need to consider composition of POM for $D_p$ < 200 nm)
0.30
0.32
0.34
0.36
0.38
0.40
POM/(POM + $NO_3$ + $SO_4$)
CCN Concentration at 0.7% SS, $cm^{-3}$
Monodisperse 60 nm CN Concentration, $cm^{-3}$
10
20
30
40
50

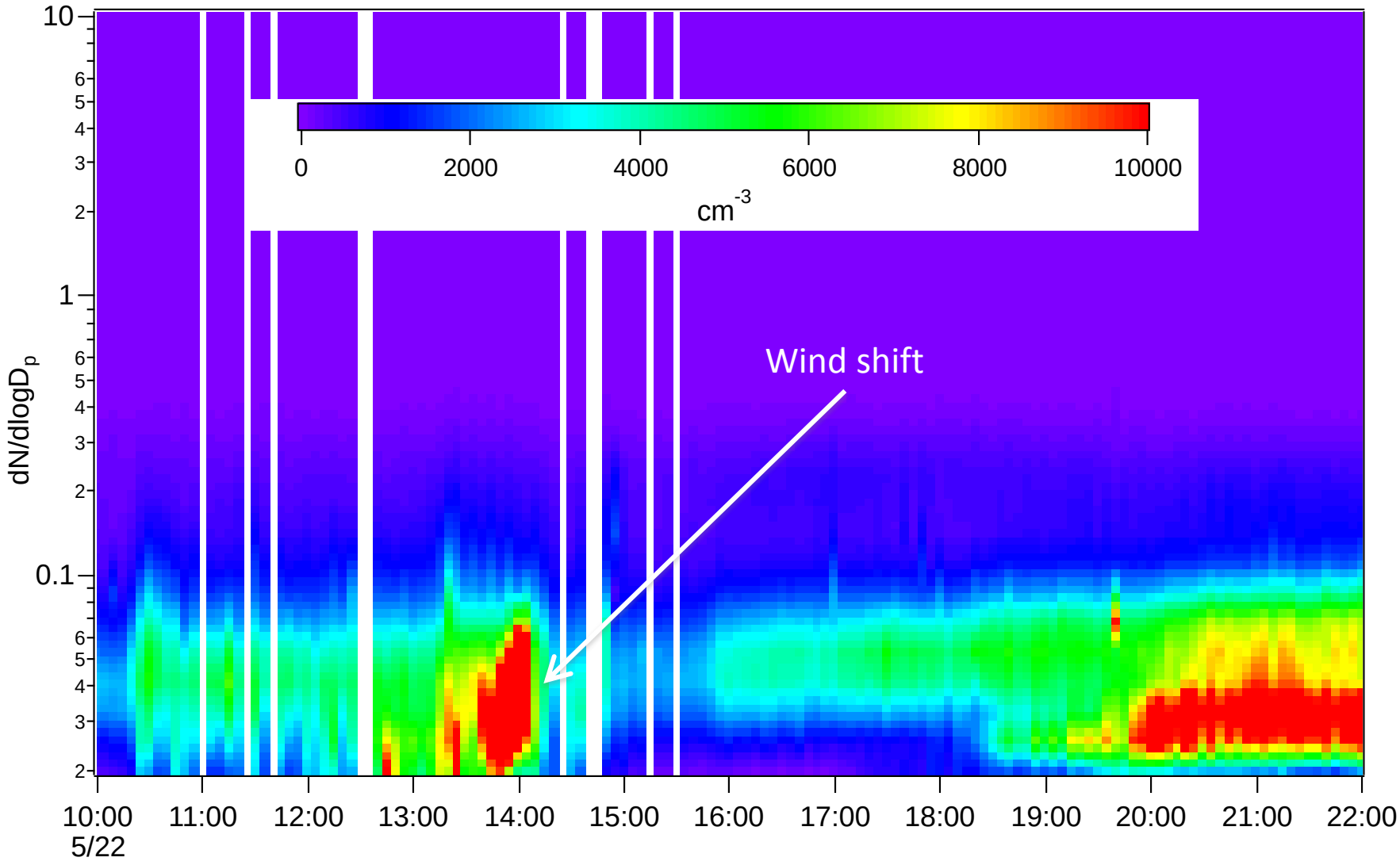

Santa Monica Bay Outflow
0.04
0.08
0.12
HOA Mass Fraction for $D_{p,vaero}$ < 200 nm
CCN Concentration at 0.7% SS, $cm^{-3}$
Monodisperse 60 nm CN Concentration, $cm^{-3}$
10
20
30
40
50

# Cruise Terminal Port of LA

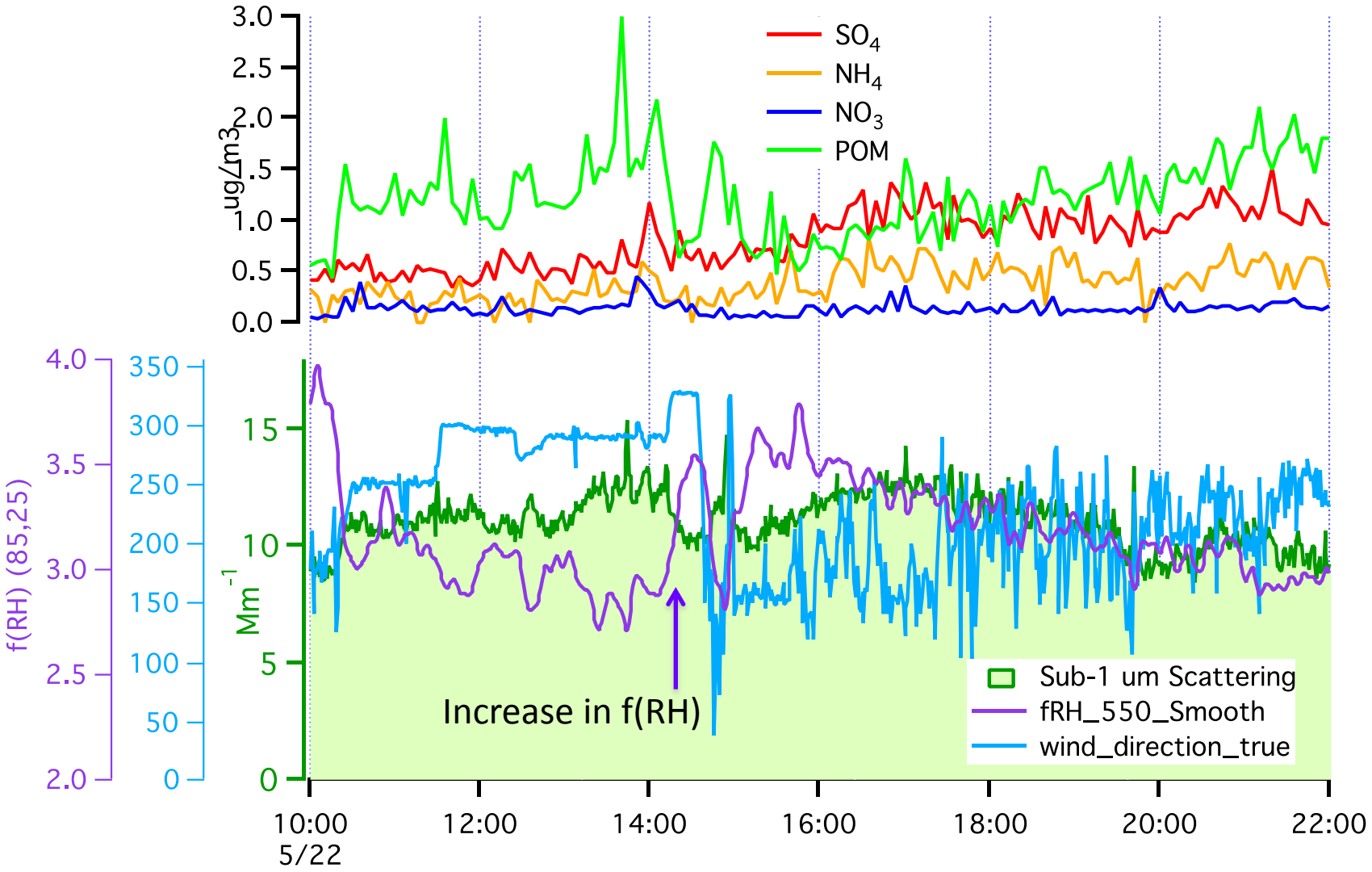
Cruise Terminal Port of LA

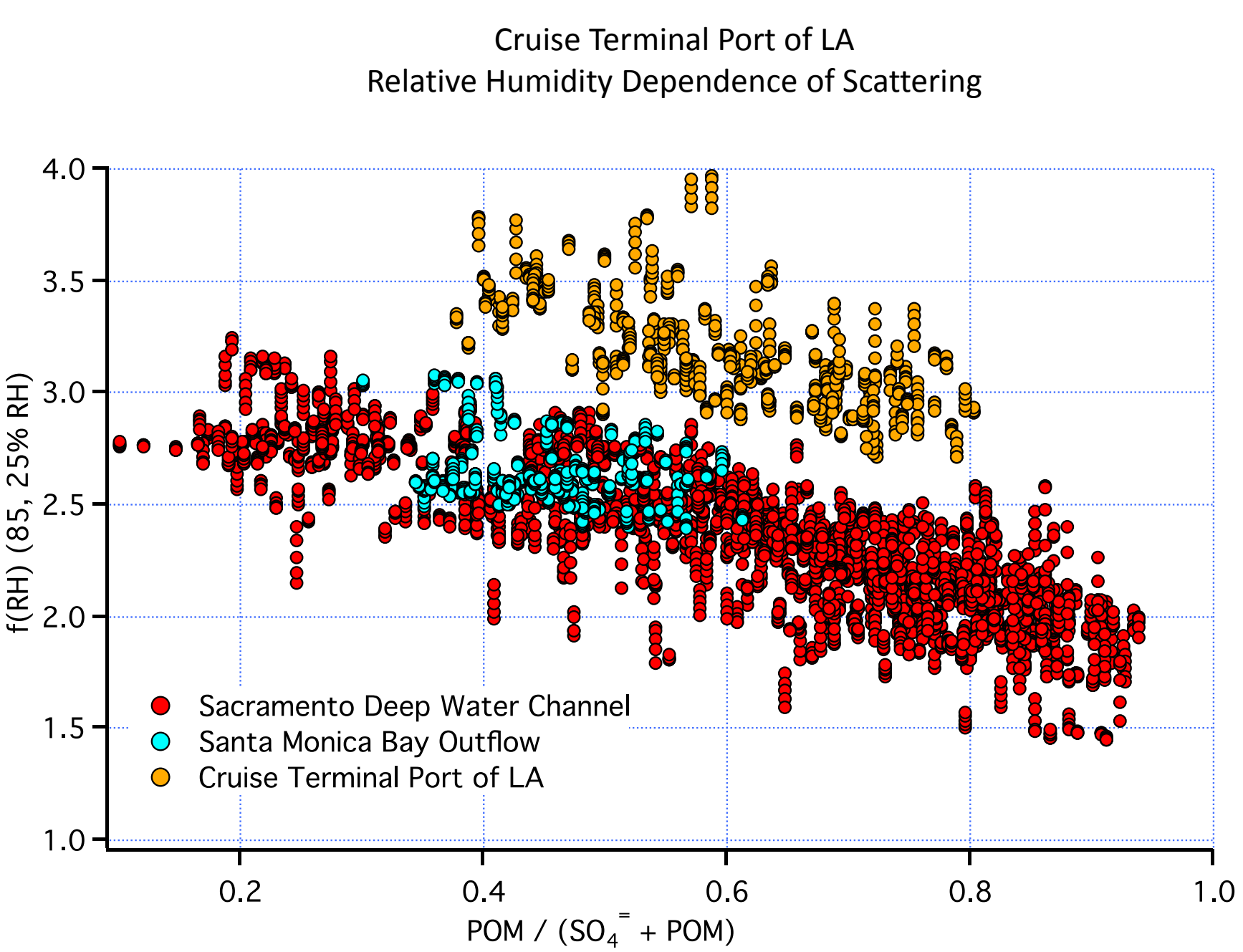
Wind shift at 14:30 UTC led to lower particle number concentrations, decrease in POM, and an increase in $SO_4^=$

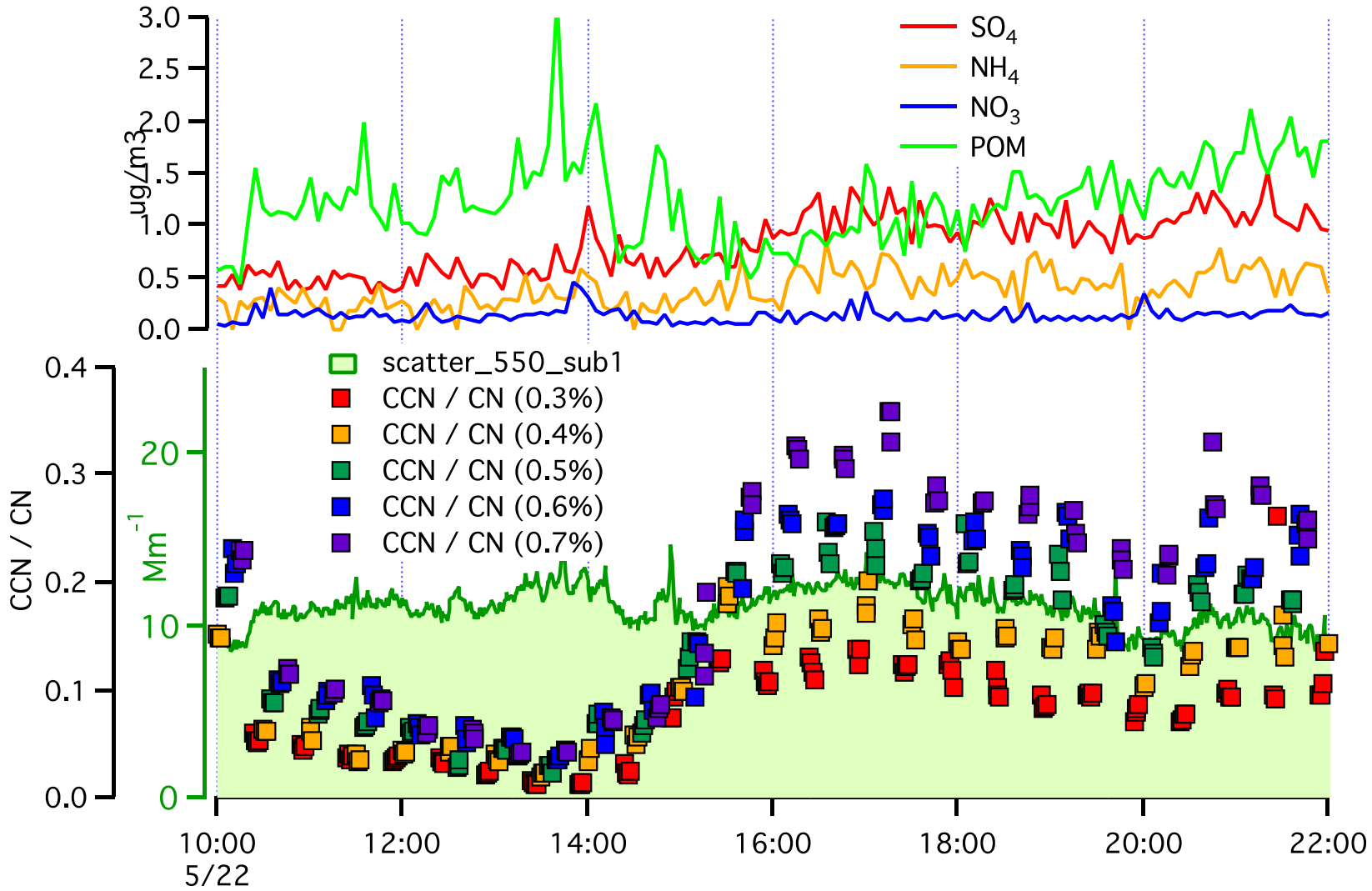



Cruise Terminal in Port of LA
Particle number size distribution
10
6
5
4
3
2
1
6
5
4
3
2
0.1
6
5
4
3
2
dN/dlogDp
0
2000
4000
6000
8000
10000
cm-3
Wind shift
10:00
5/22
11:00
12:00
13:00
14:00
15:00
16:00
17:00
18:00
19:00
20:00
21:00
22:00

# Cruise Terminal Port of LA
# Relative Humidity Dependence of Scattering

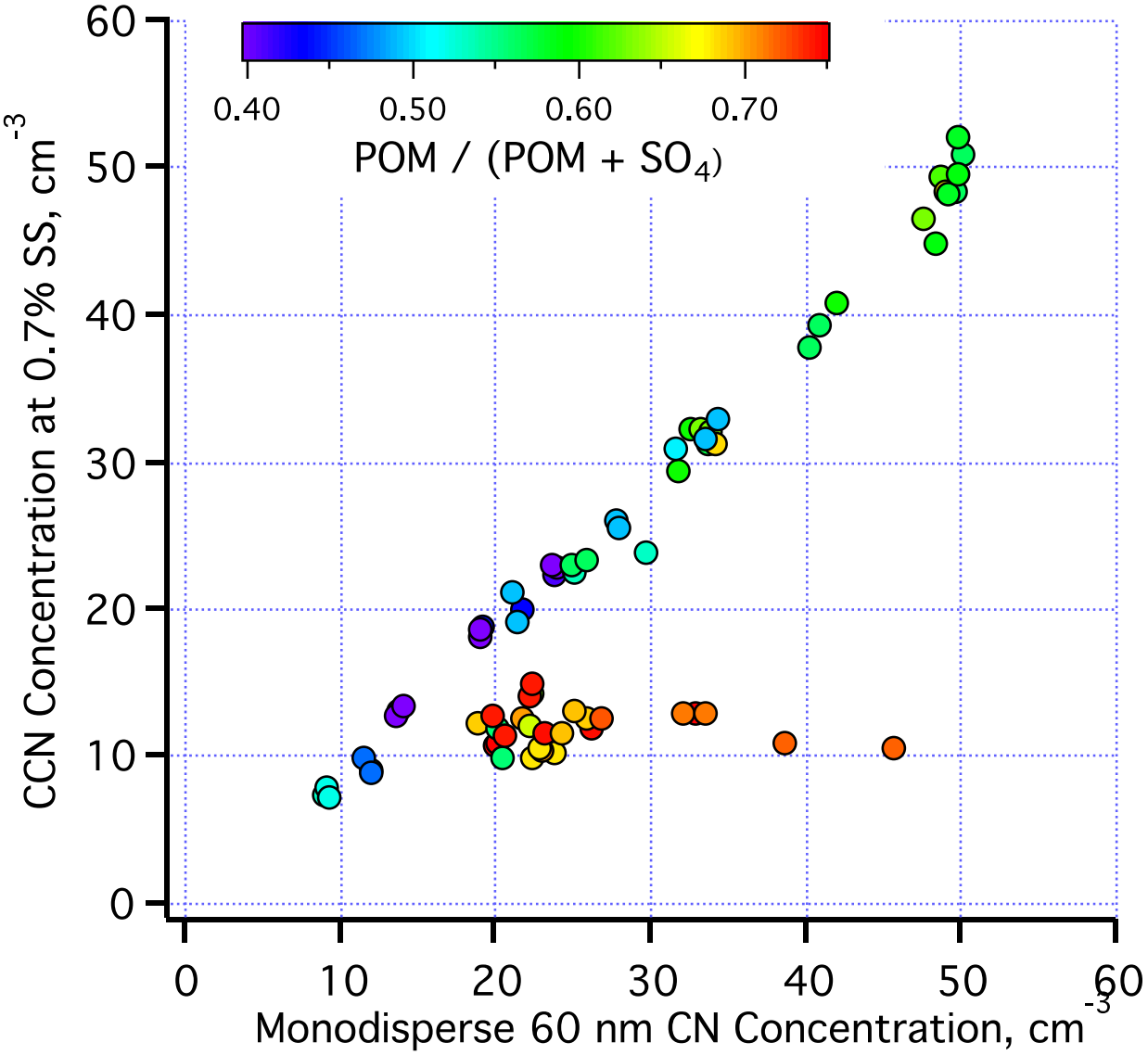



Cruise Terminal Port of LA
Relative Humidity Dependence of Scattering
f(RH) (85, 25% RH)
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.2
0.4
0.6
0.8
1.0
POM / (SO4= + POM)
Sacramento Deep Water Channel
Santa Monica Bay Outflow
Cruise Terminal Port of LA

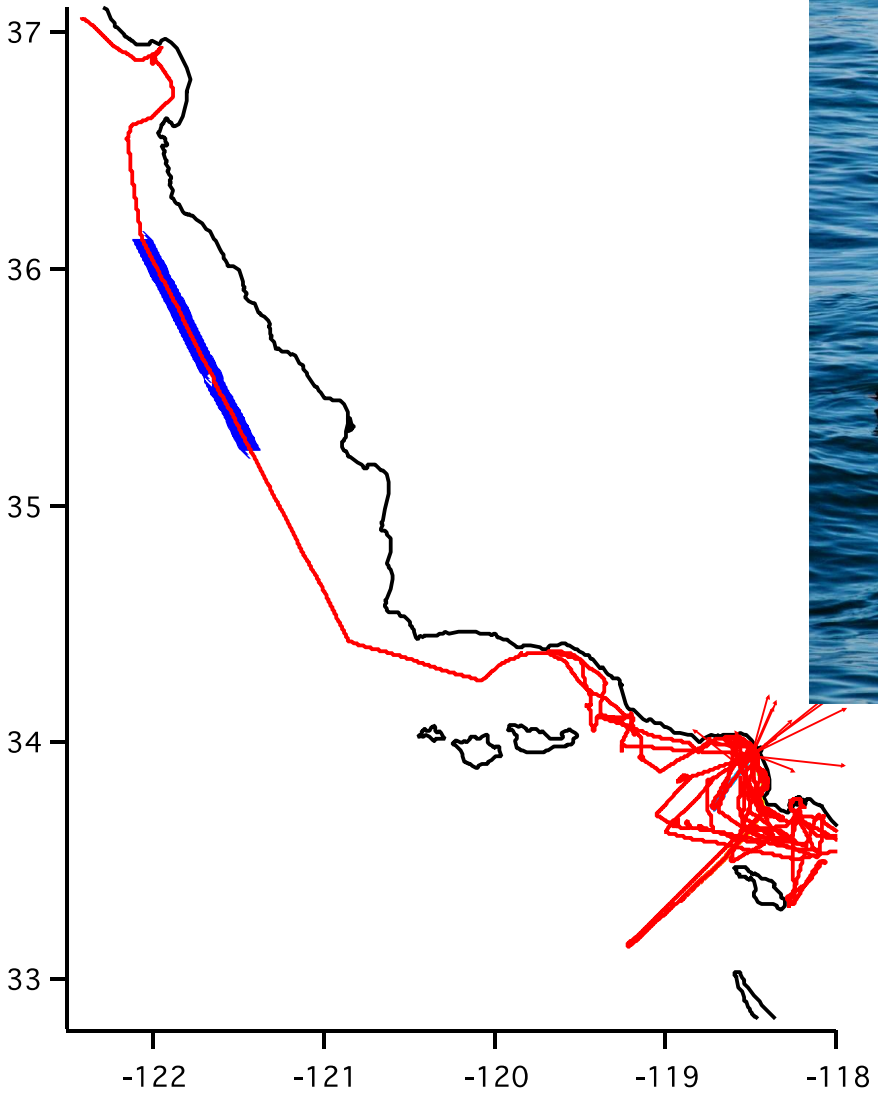

Cruise Terminal Port of LA
CCN Activation Ratio of Sub-1 um Aerosol
ug/m3
3.0
2.5
2.0
1.5
1.0
0.5
0.0
SO4
NH4
NO3
POM
scatter_550_sub1
CCN / CN (0.3%)
CCN / CN (0.4%)
CCN / CN (0.5%)
CCN / CN (0.6%)
CCN / CN (0.7%)
CCN / CN
0.4
0.3
0.2
0.1
0.0
Mm-1
20
10
0
10:00
5/22
12:00
14:00
16:00
18:00
20:00
22:00

Cruise Terminal Port of LA

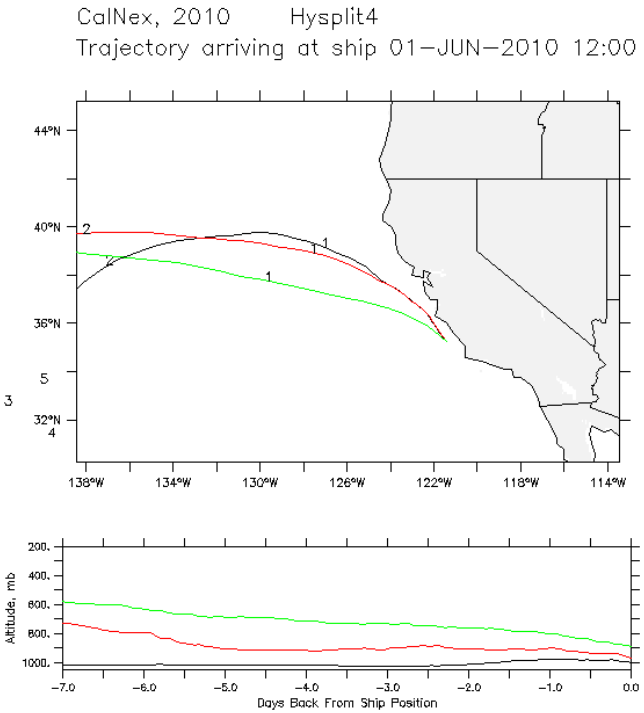
CCN Activation Ratio of 60 nm Aerosol as a function of POM mass fraction

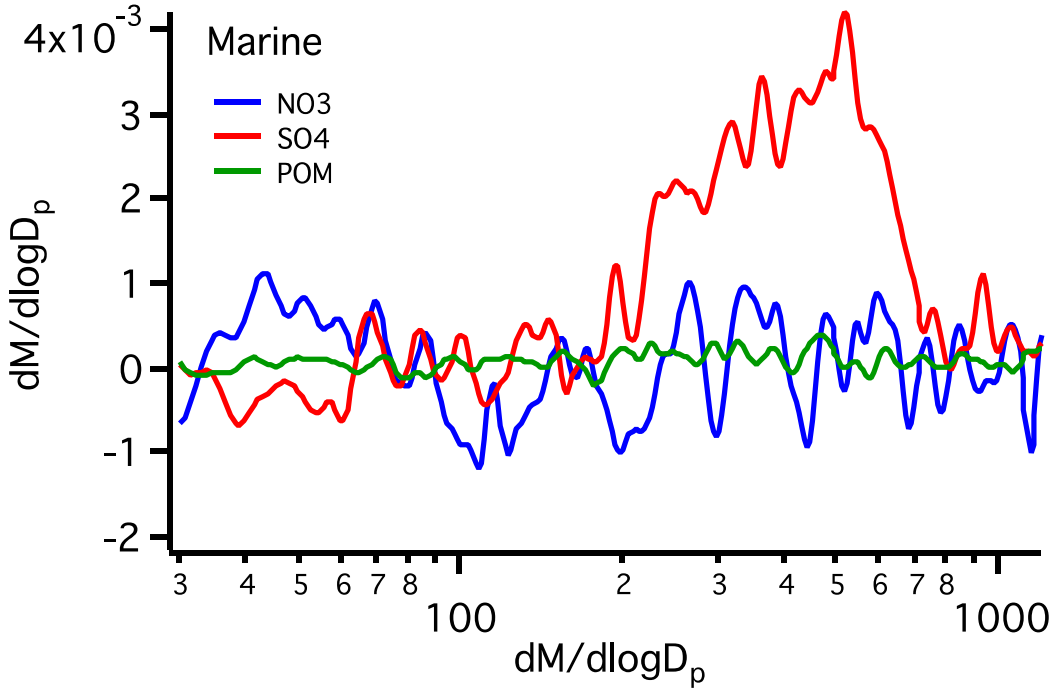


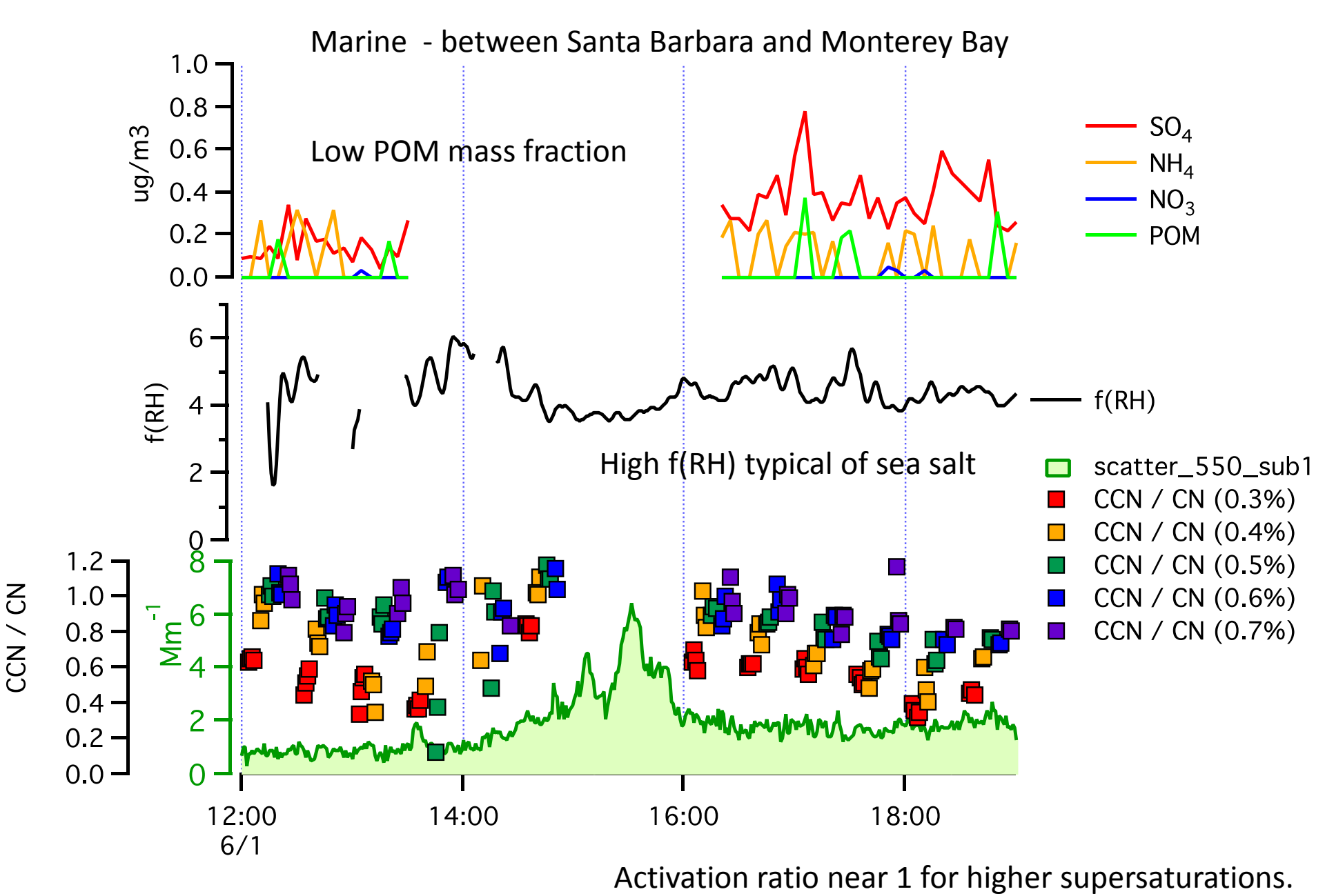
# Marine - between Santa Barbara and Monterey Bay



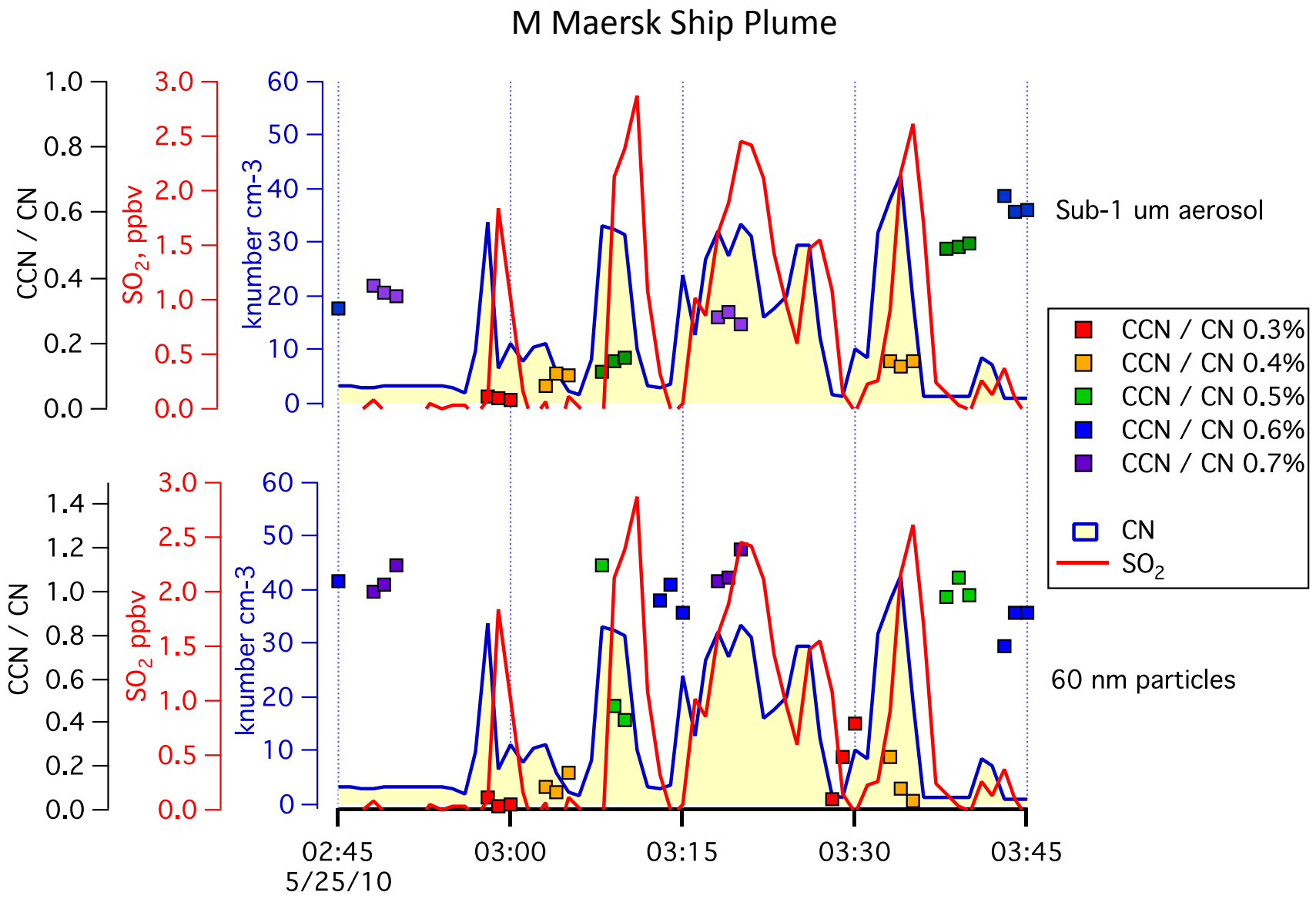
# Marine - between Santa Barbara and Monterey Bay



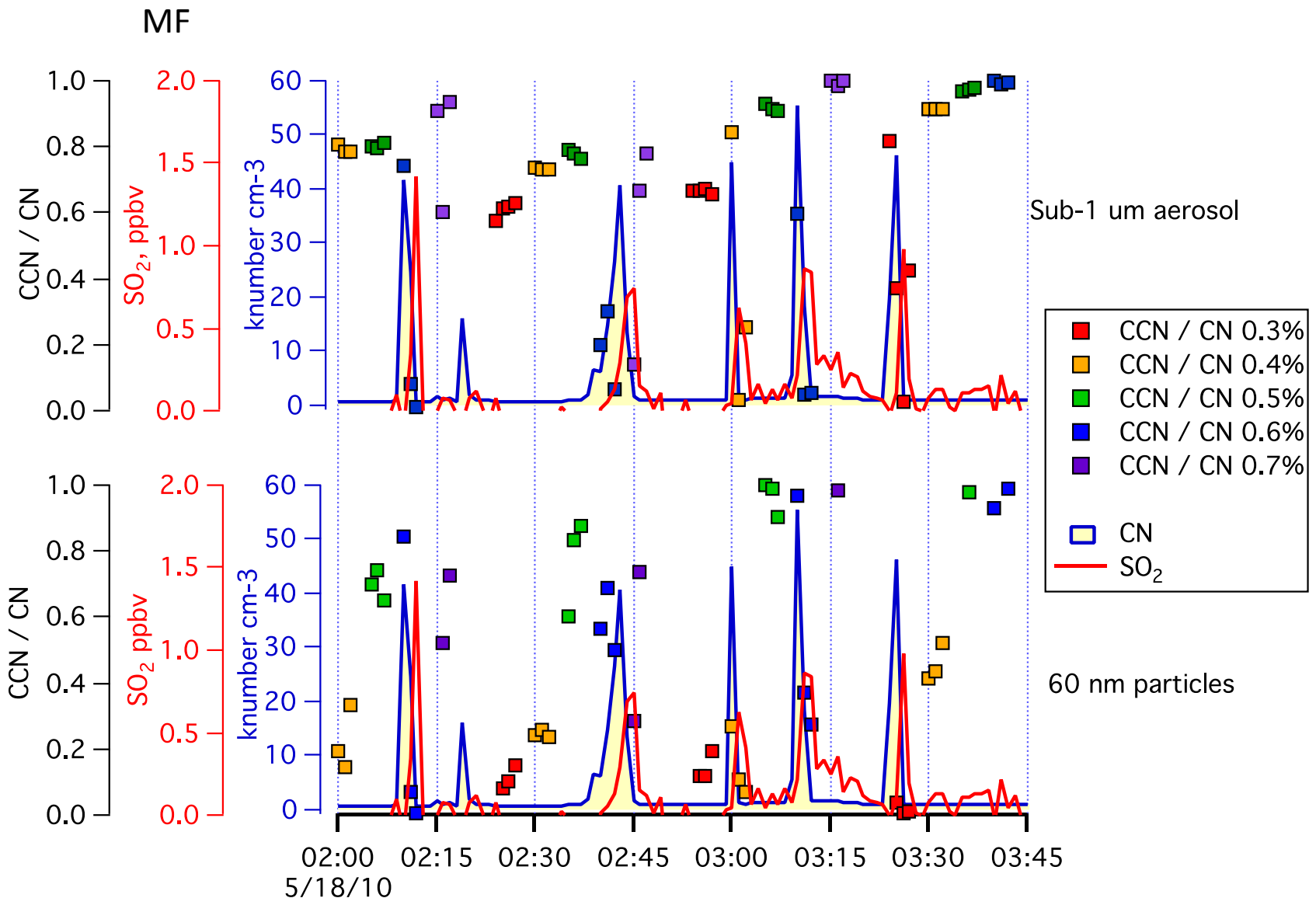




Marine - between Santa Barbara and Monterey Bay
ug/m3
1.0
0.8
0.6
0.4
0.2
0.0
Low POM mass fraction
SO4
NH4
NO3
POM
f(RH)
6
4
2
0
f(RH)
High f(RH) typical of sea salt
scatter_550_sub1
CCN / CN (0.3%)
CCN / CN (0.4%)
CCN / CN (0.5%)
CCN / CN (0.6%)
CCN / CN (0.7%)
CCN / CN
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Mm-1
8
6
4
2
0
12:00
6/1
14:00
16:00
18:00
Activation ratio near 1 for higher supersaturations.

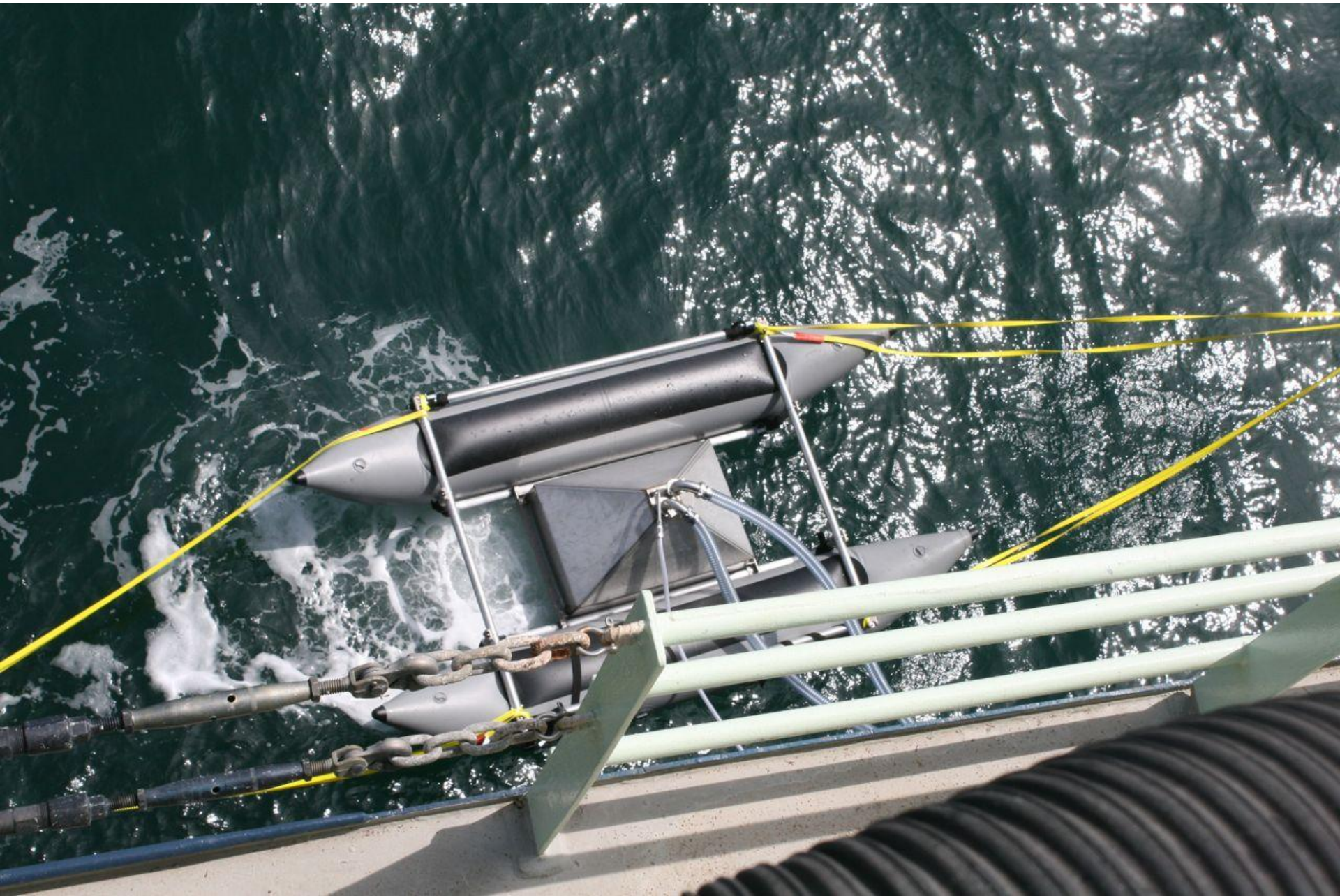
M. Maersk

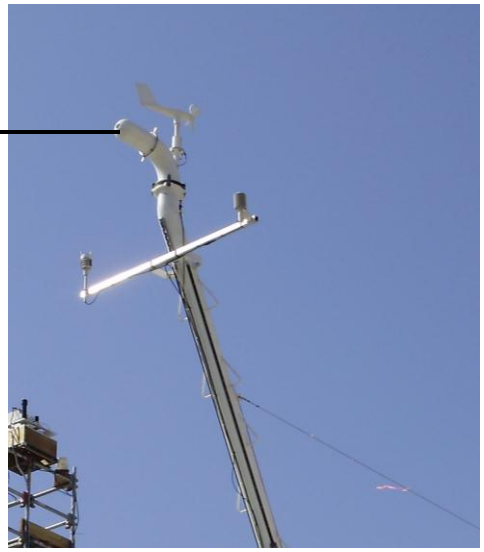

M Maersk Ship Plume
CCN / CN
SO2, ppbv
knumber cm-3
Sub-1 um aerosol
CCN / CN 0.3%
CCN / CN 0.4%
CCN / CN 0.5%
CCN / CN 0.6%
CCN / CN 0.7%
CN
SO2
60 nm particles
02:45
5/25/10
03:00
03:15
03:30
03:45

Miller Freeman


MF
CCN / CN
SO2, ppbv
knumber cm-3
Sub-1 um aerosol
CCN / CN 0.3%
CCN / CN 0.4%
CCN / CN 0.5%
CCN / CN 0.6%
CCN / CN 0.7%
CN
SO2
60 nm particles
02:00
02:15
02:30
02:45
03:00
03:15
03:30
03:45
5/18/10

# Seasweep: Generation of sea spray aerosol at the ocean surface





Inlet to sampling mast

Particle-free air

Air Curtain

Air Curtain to seal off hood from ambient air

Frit

Frit ( 1 m below the surface)

Relationship between particle size, composition, and supersaturation required for a particle to act as a CCN

$$s_c \approx \sqrt{\frac{4}{\kappa D_p^3}\left(\frac{4\sigma M_w}{3RT\rho_w}\right)^3}$$

$S_c$ = critical water vapor supersaturation

$\kappa$ = hygroscopicity parameter

$D_p$ = dry particle diameter

$\kappa$ can be determined from measured CCN concentrations at a given $S_c$ and the particle number size distribution

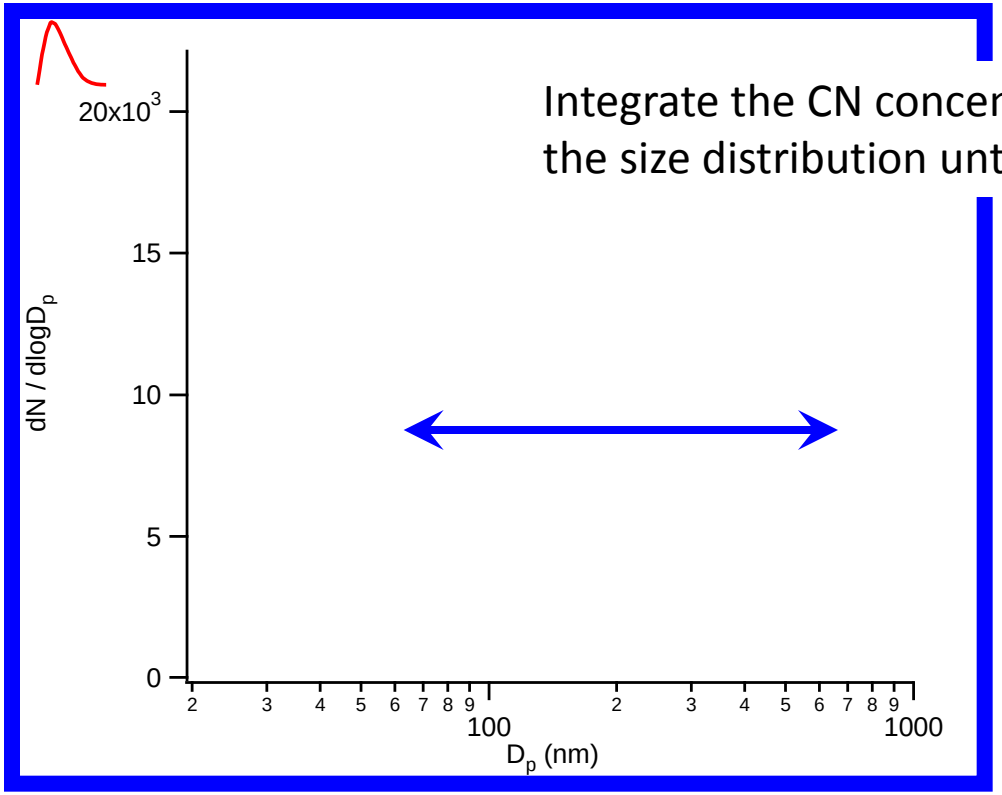
# Relationship between particle size, composition, and supersaturation required for a particle to act as a CCN

$$s_c \approx \sqrt{\frac{4}{\kappa D_p^3}\left(\frac{4\sigma M_w}{3RT\rho_w}\right)^3}$$

$S_c$ = critical water vapor supersaturation

$\kappa$ = hygroscopicity parameter

$D_p$ = dry particle diameter

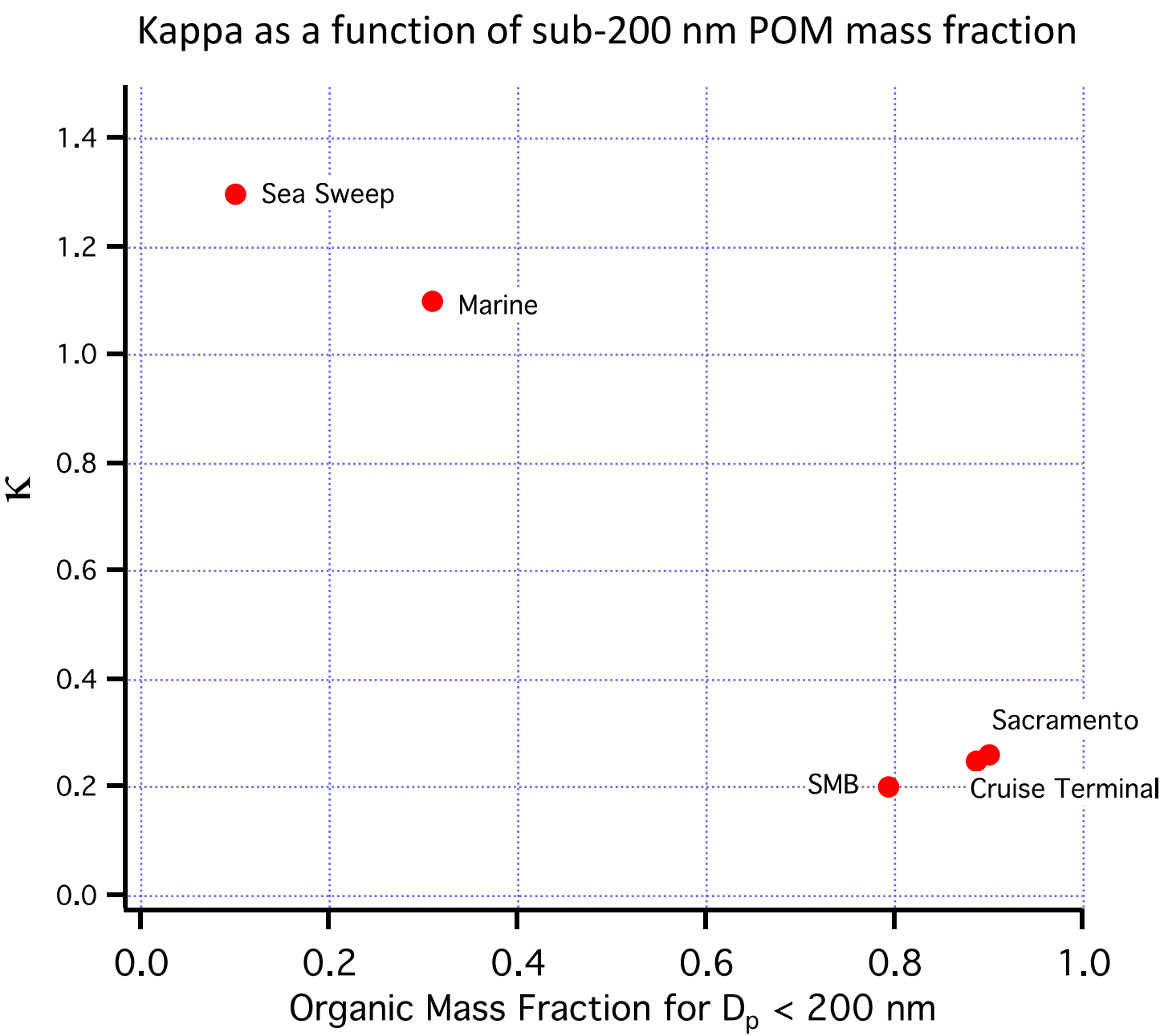



**Critical diameter** is defined as the diameter at which:

$CCN_{measured}$ = CN Concentration

$D_p = D_c$

| CCN Source | Kappa |
|---|---|
| Miller Freeman | 0.025 |
| M. Maersk | 0.13 |
| Santa Monica Bay  Outflow | 0.20 |
| Cruise Terminal – LA | 0.25 |
| Sacramento | 0.26 |
| Marine | 1.1 |
| Sea Sweep | 1.3 |


Kappa as a function of sub-200 nm POM mass fraction
κ
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Sea Sweep
Marine
Sacramento
SMB
Cruise Terminal
0.0
0.2
0.4
0.6
0.8
1.0
Organic Mass Fraction for $D_p$ < 200 nm

## Conclusions to date

• We observed the same influence of aerosol composition (POM mass fraction) on f(RH) as has been reported for other marine regions downwind of continental aerosol sources. Now we need to do a more detailed assessment of the impacts of the composition of the POM.

• The measurement of monodisperse 60 nm particles allowed for the isolation of the influence of composition on the CCN activation ratio.

• We observed that higher POM mass fractions led to lower CCN activation ratios and kappa values. The next step is to assess the impacts of the composition of the POM and to generalize results across all collected marine data sets.

• The measurements of CCN concentrations in ship plumes allow for the derivation of CCN emission factors (Lack et al., 2011).