Aerosol enhancements in the upper troposphere over the Amazon forest: Do Amazonian clouds produce aerosols?

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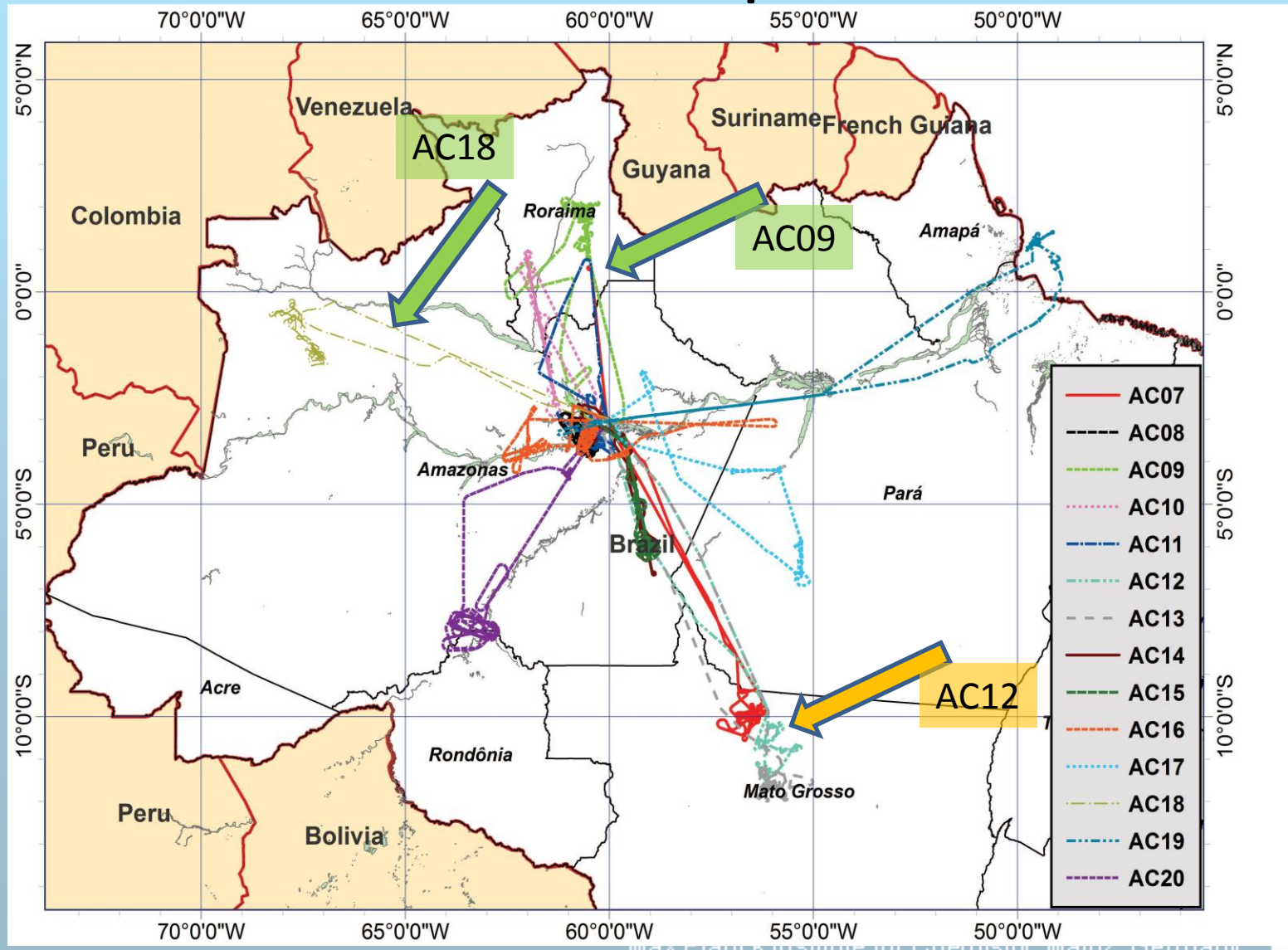
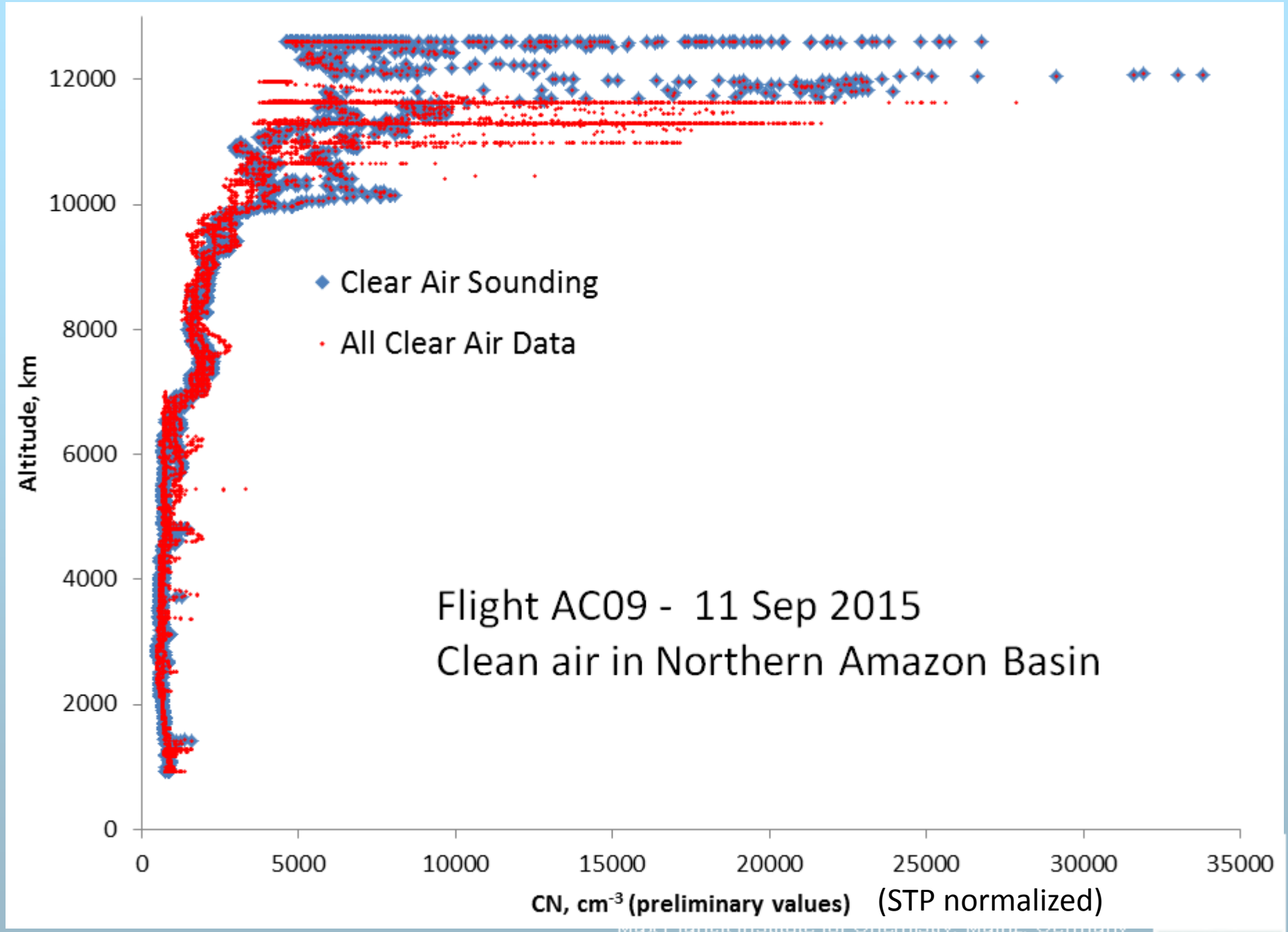
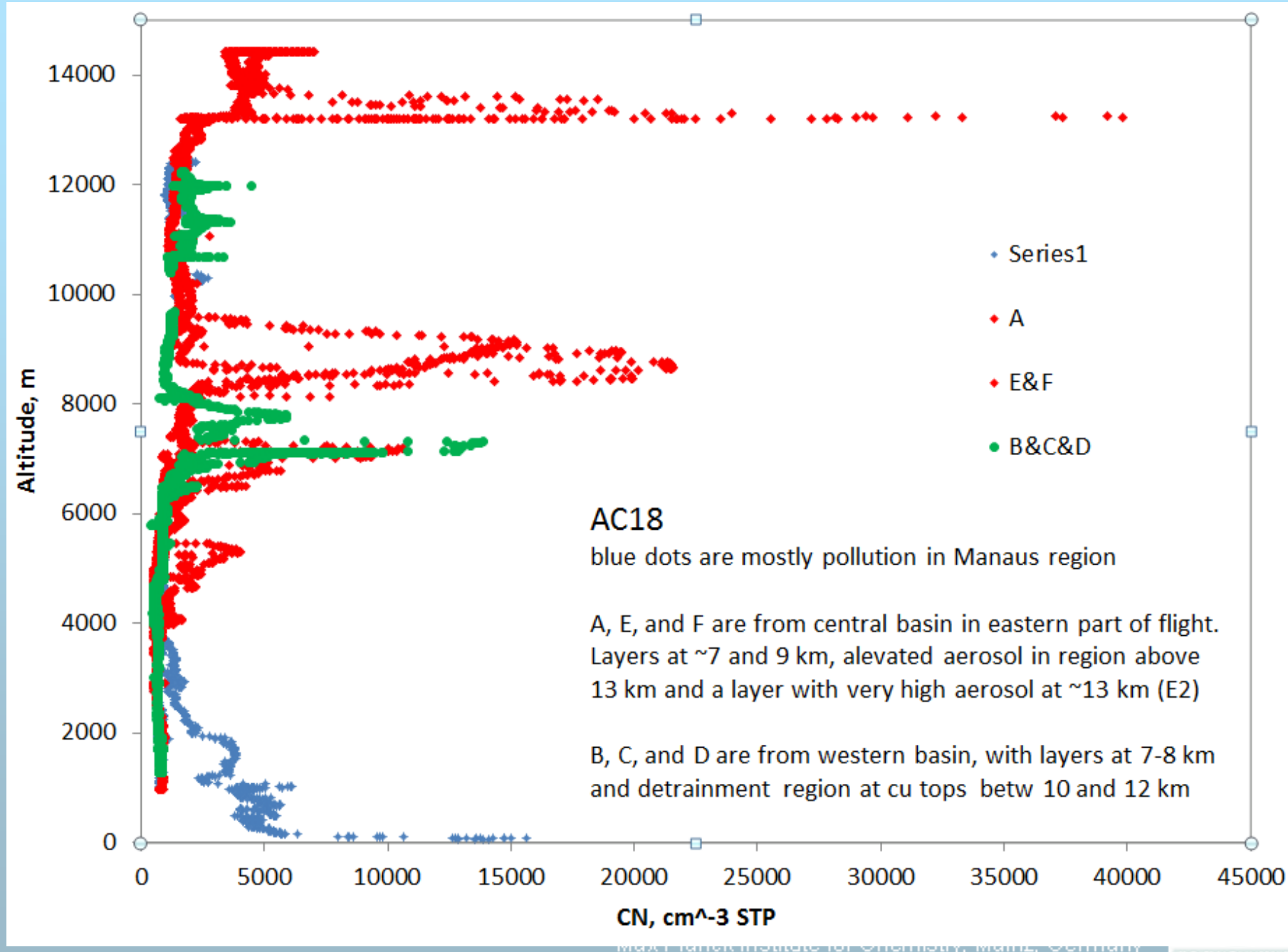
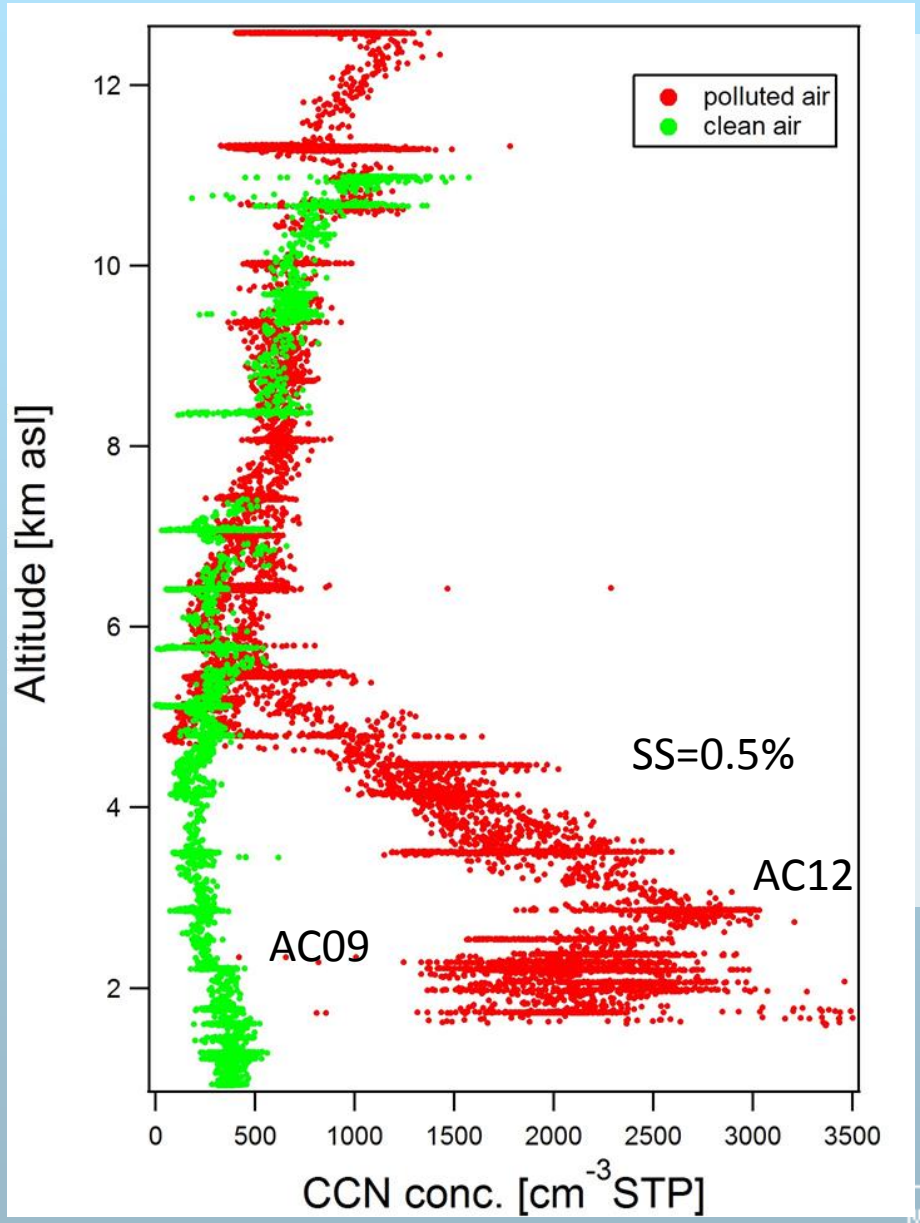
ACRIDICON-CHUVA on the HALO Aircraft Amazon Basin – September 2014




Altitude, km
0
2000
4000
6000
8000
10000
12000
0
5000
10000
15000
20000
25000
30000
35000
CN, cm-3 (preliminary values)
Clear Air Sounding
All Clear Air Data
Flight AC09 - 11 Sep 2015
Clean air in Northern Amazon Basin
(STP normalized)


Altitude, m
14000
12000
10000
8000
6000
4000
2000
0
0
5000
10000
15000
20000
25000
30000
35000
40000
45000
CN, cm^-3 STP
Series1
A
E&F
B&C&D
AC18
blue dots are mostly pollution in Manaus region
A, E, and F are from central basin in eastern part of flight. Layers at ~7 and 9 km, alevated aerosol in region above 13 km and a layer with very high aerosol at ~13 km (E2)
B, C, and D are from western basin, with layers at 7-8 km and detrainment region at cu tops betw 10 and 12 km

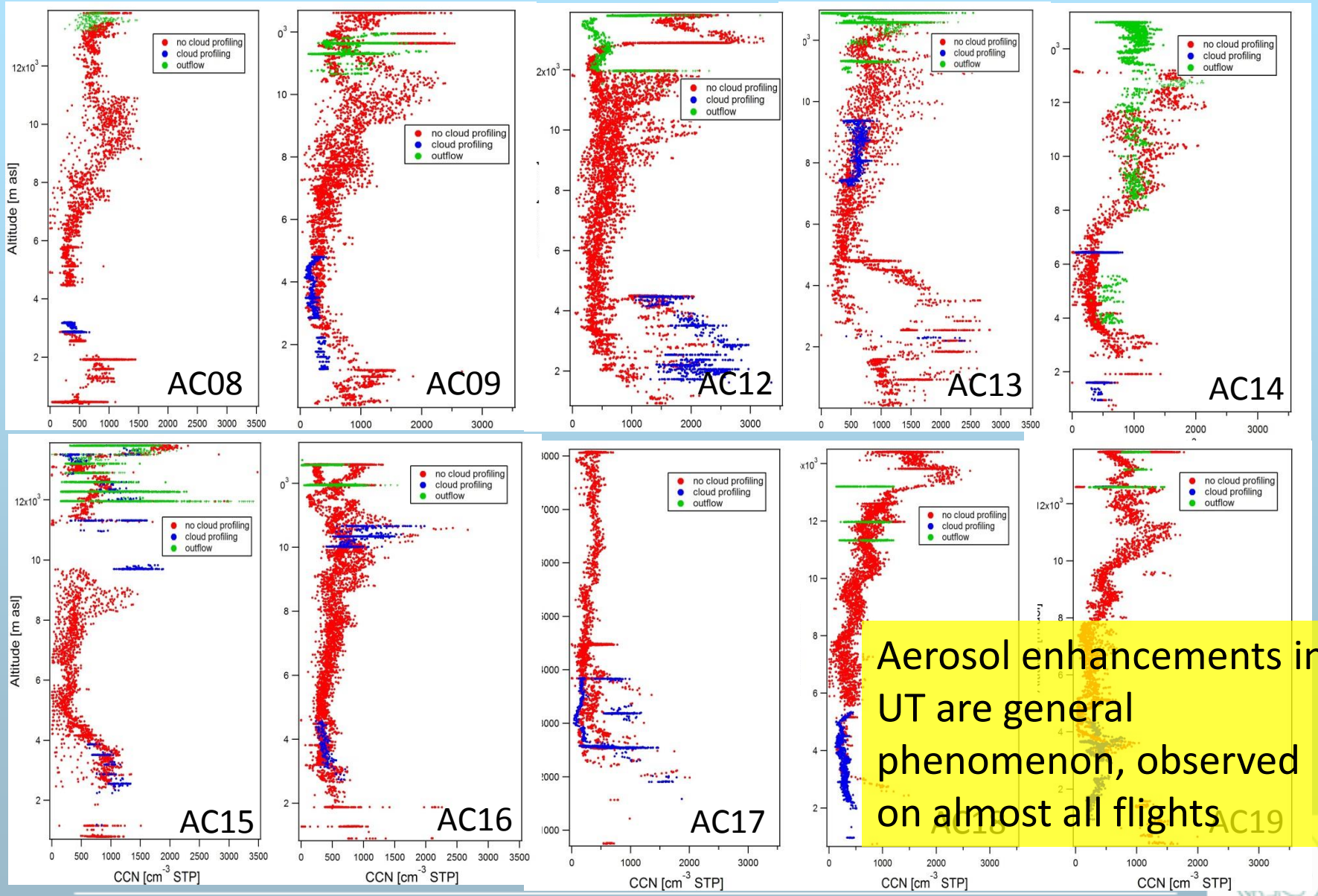
CCN Enhancement



CCN are also enhanced in UT, but not as strongly as CN: smaller particles and/or less hygroscopic

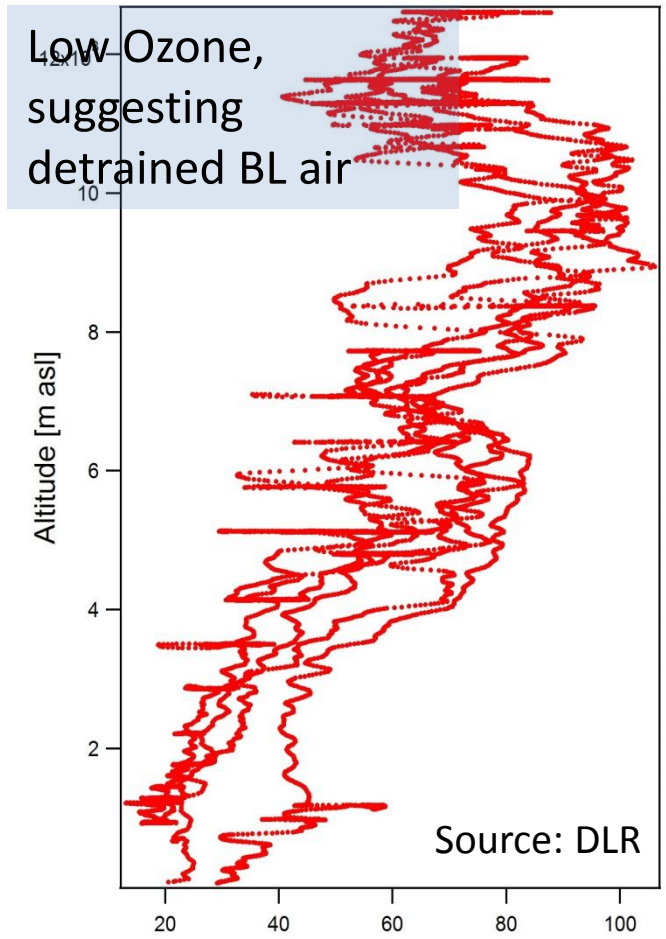
Enhancements in UT similar over clean area and polluted area: Aerosol is not result of pollution, but likely product of particle production from clean BL air – BVOC oxidation products

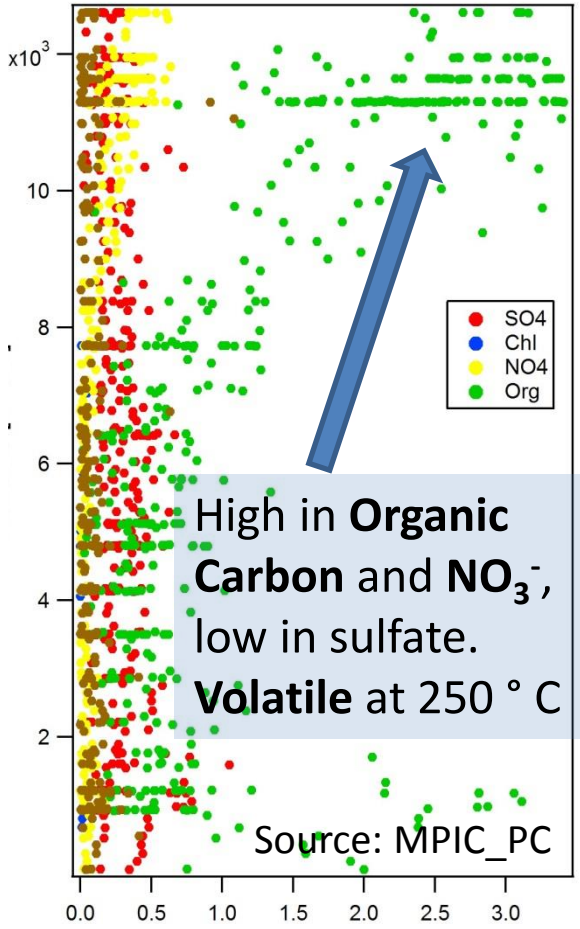
HALO ACRIDICON-CHUVA campaign Sep. 2014

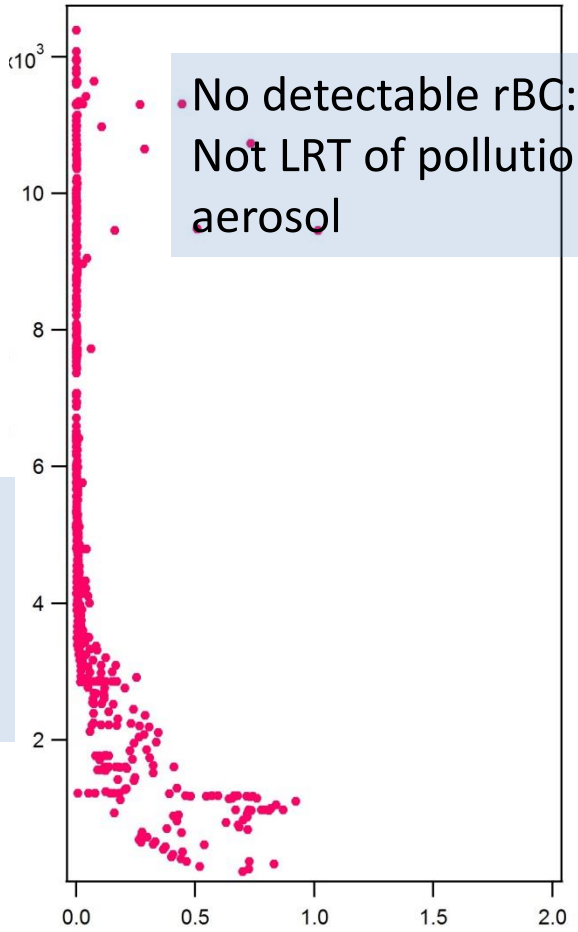


Aerosol enhancements in UT are general phenomenon, observed on almost all flights

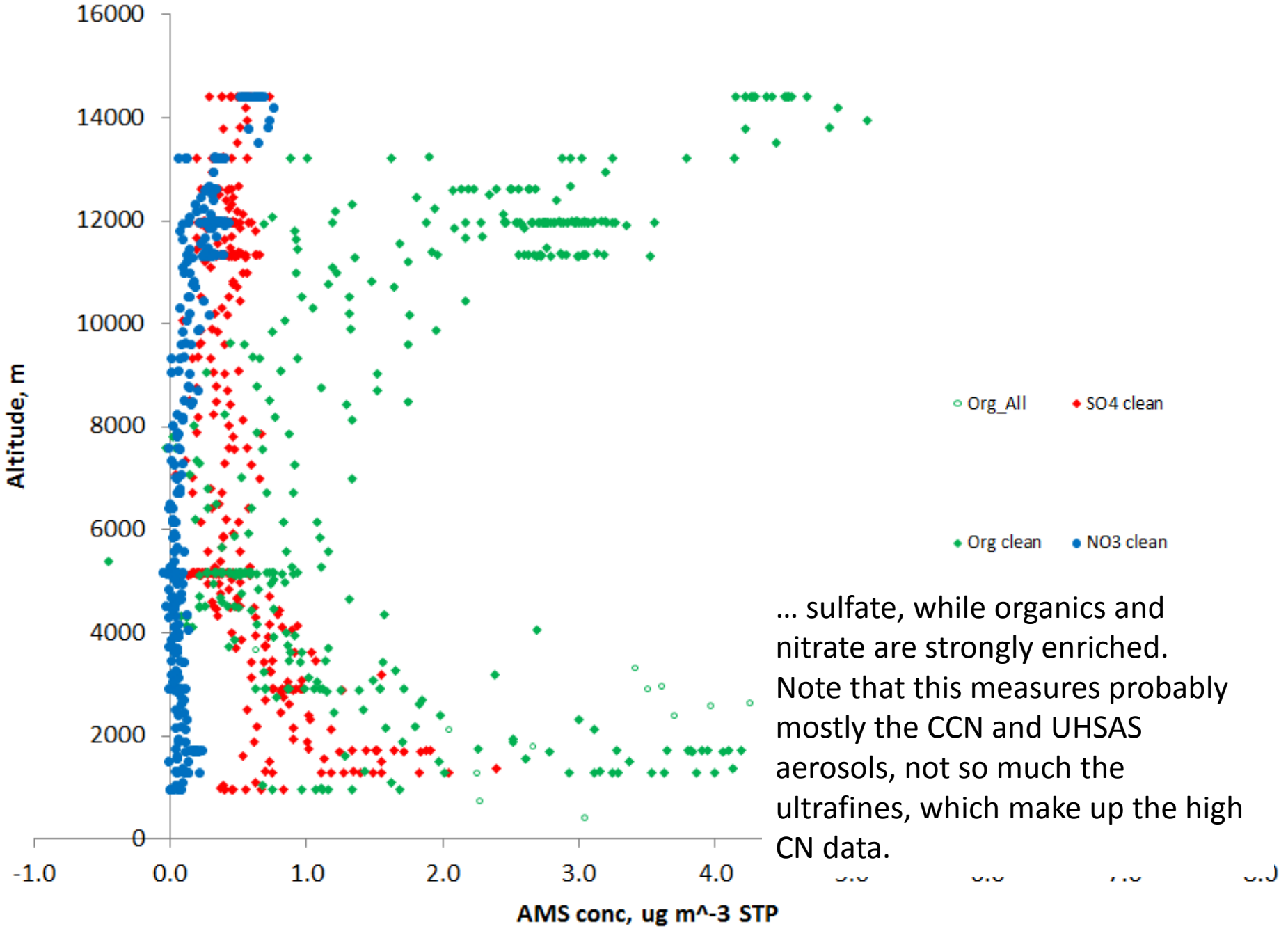
Flight AC09 - Ozone, Aerosol Composition





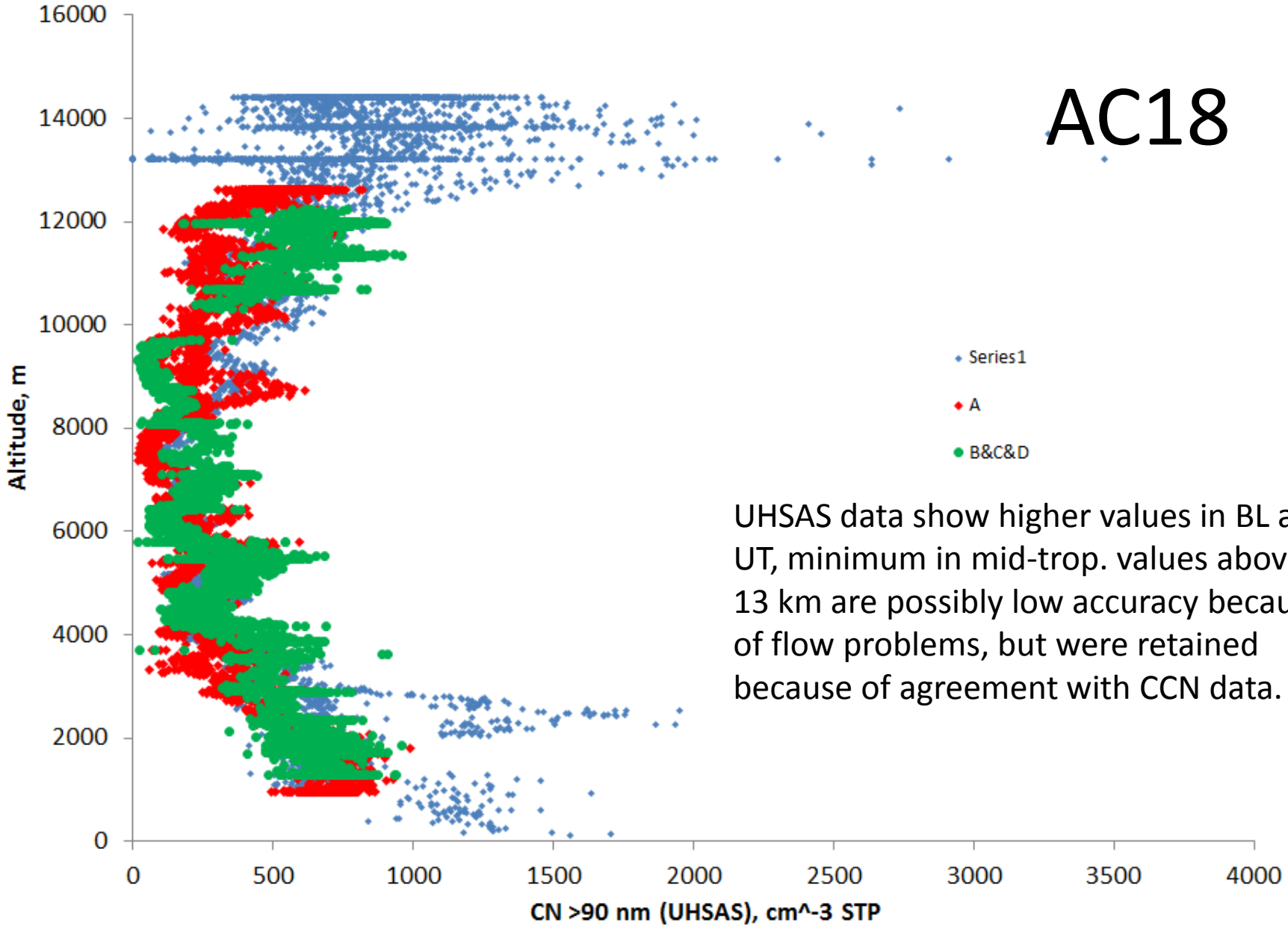


(STP normalized)



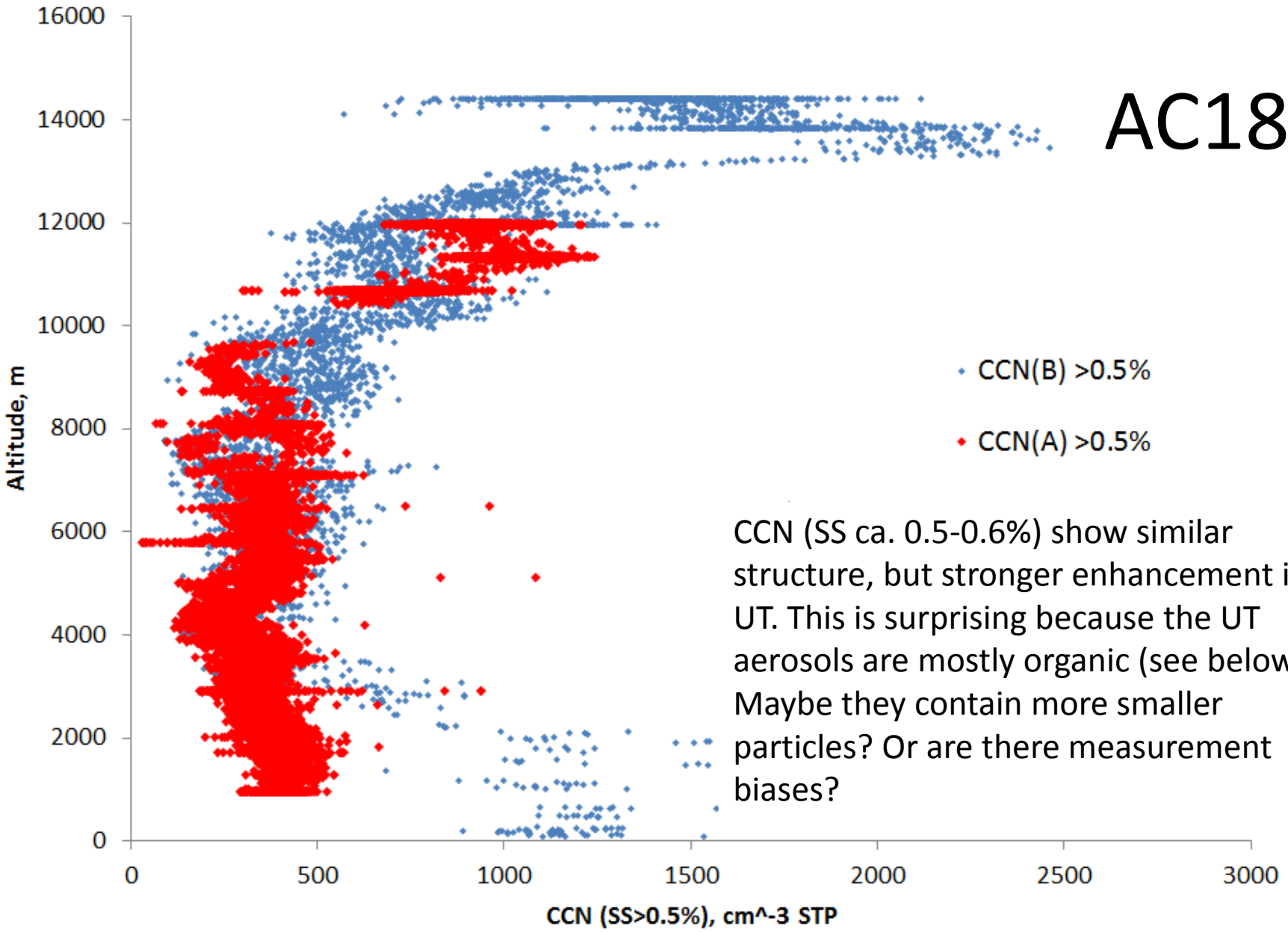
… sulfate, while organics and nitrate are strongly enriched. Note that this measures probably mostly the CCN and UHSAS aerosols, not so much the ultrafines, which make up the high CN data.

AC18

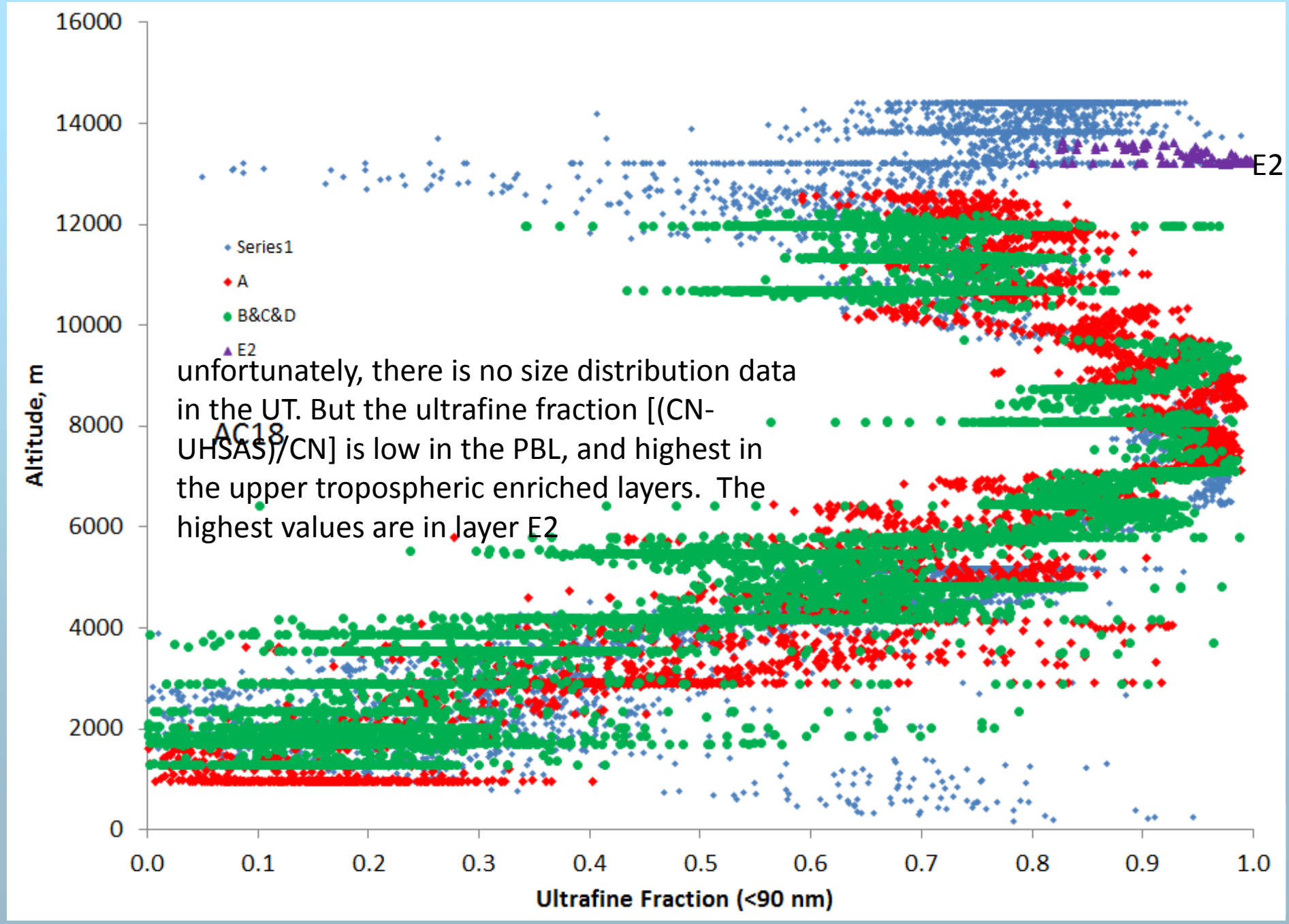


UHSAS data show higher values in BL and UT, minimum in mid-trop. values above 13 km are possibly low accuracy because of flow problems, but were retained because of agreement with CCN data.

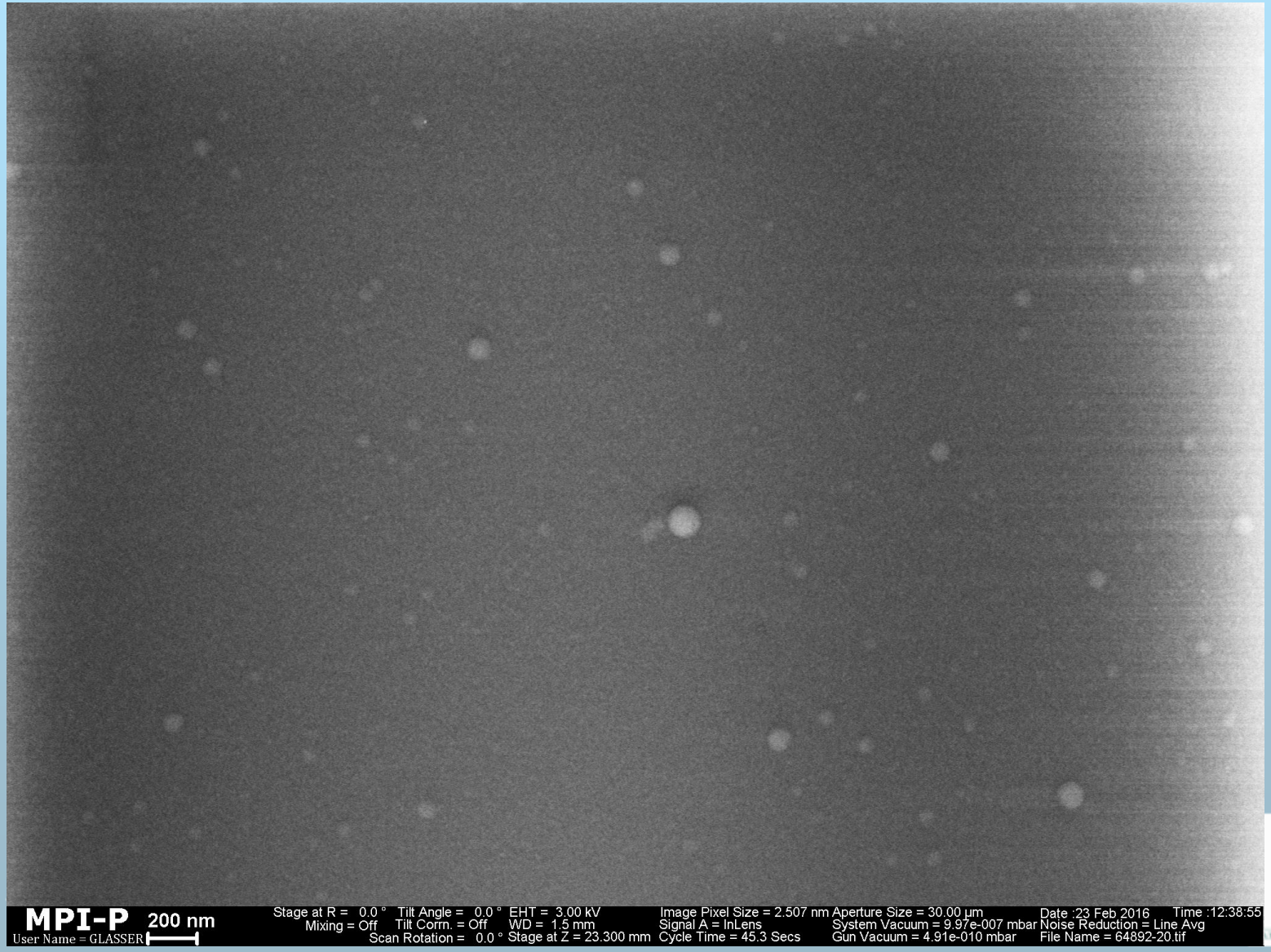
AC18

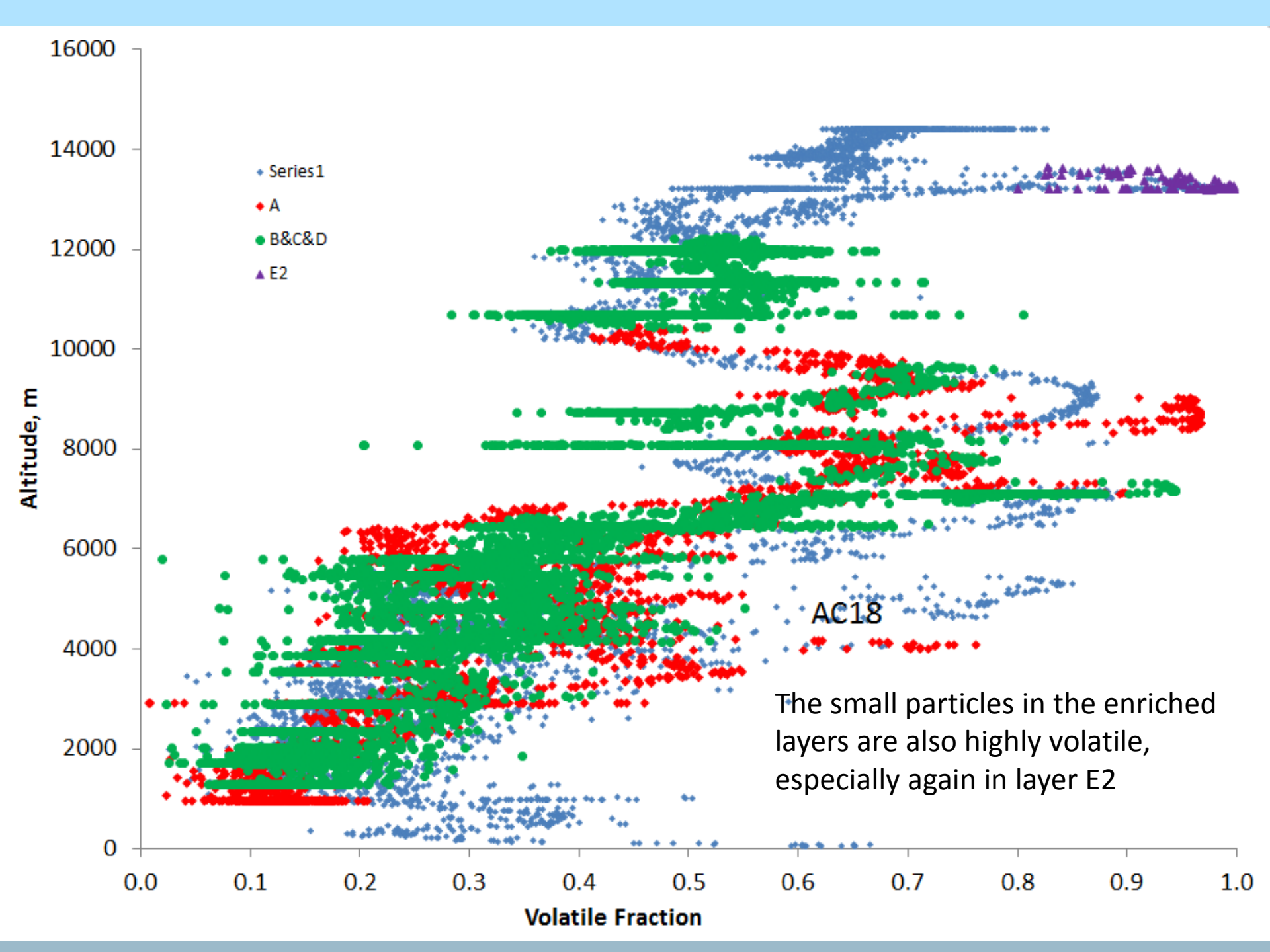


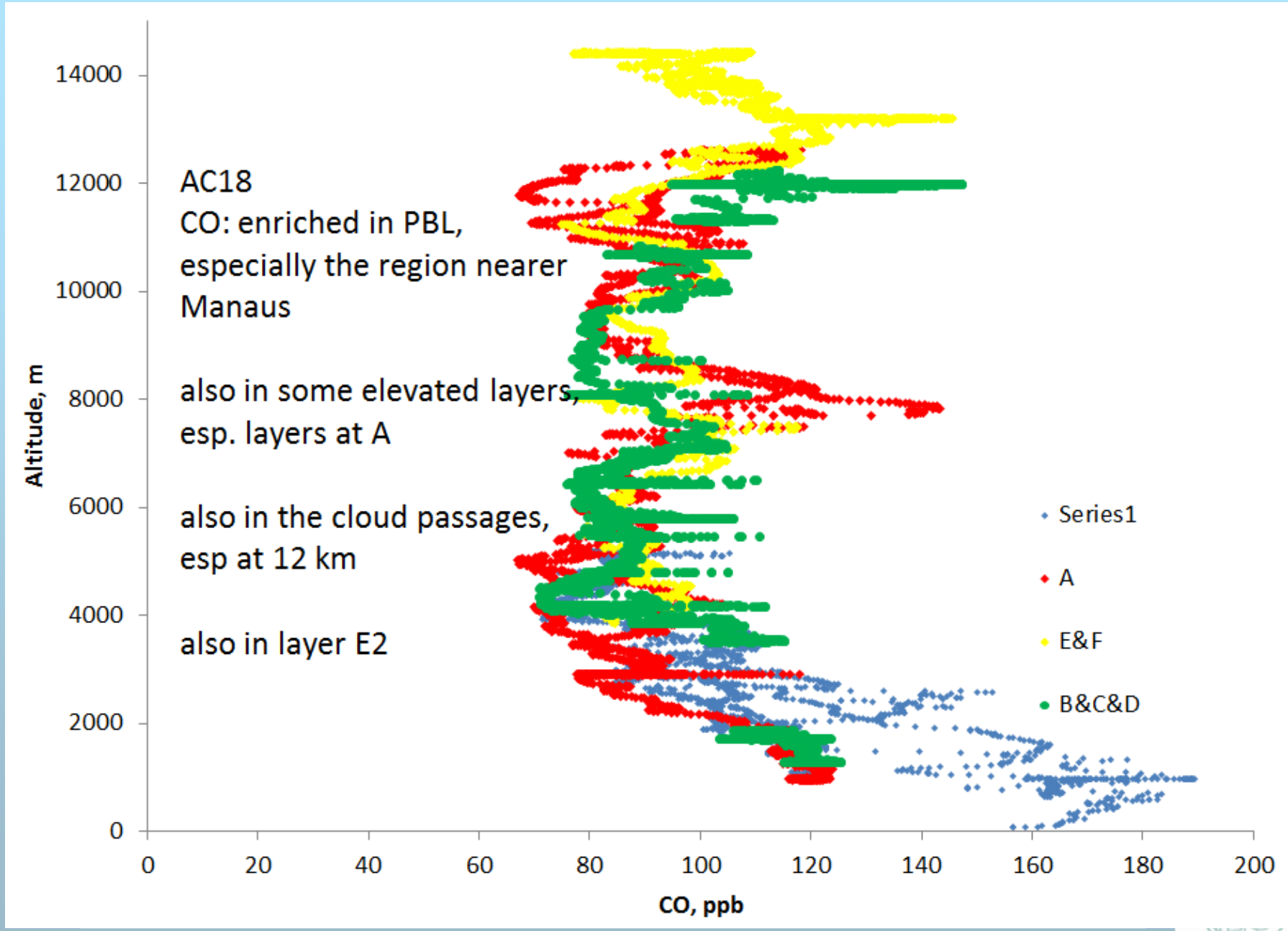
CCN (SS ca. 0.5-0.6%) show similar structure, but stronger enhancement in UT. This is surprising because the UT aerosols are mostly organic (see below). Maybe they contain more smaller particles? Or are there measurement biases?

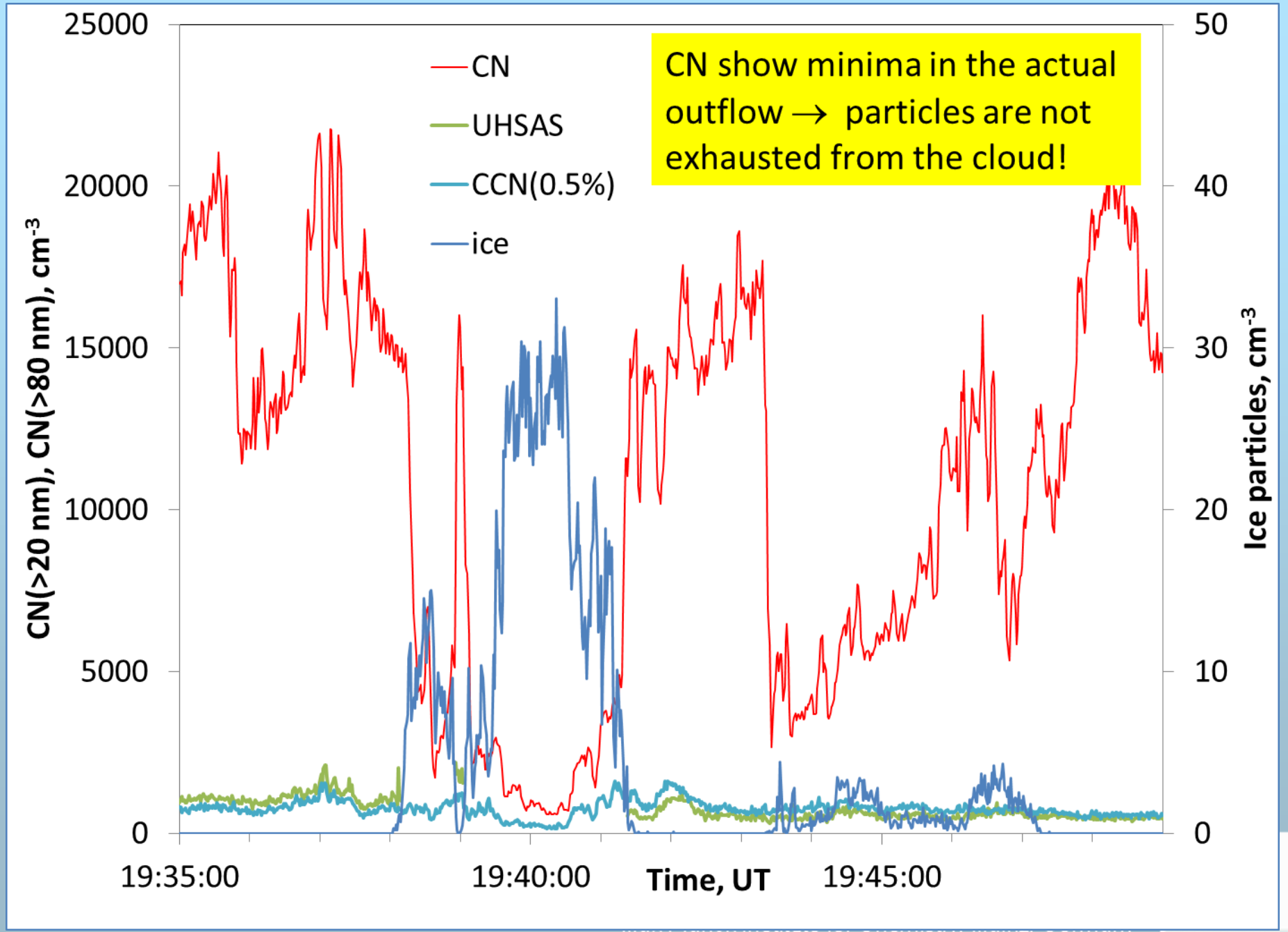

16000
14000
12000
10000
8000
6000
4000
2000
0
Altitude, m
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
Ultrafine Fraction (<90 nm)
Series1
A
B&C&D
E2
E2
AC18
unfortunately, there is no size distribution data in the UT. But the ultrafine fraction [(CN-UHSAS)/CN] is low in the PBL, and highest in the upper tropospheric enriched layers. The highest values are in layer E2

SEM Image AC12

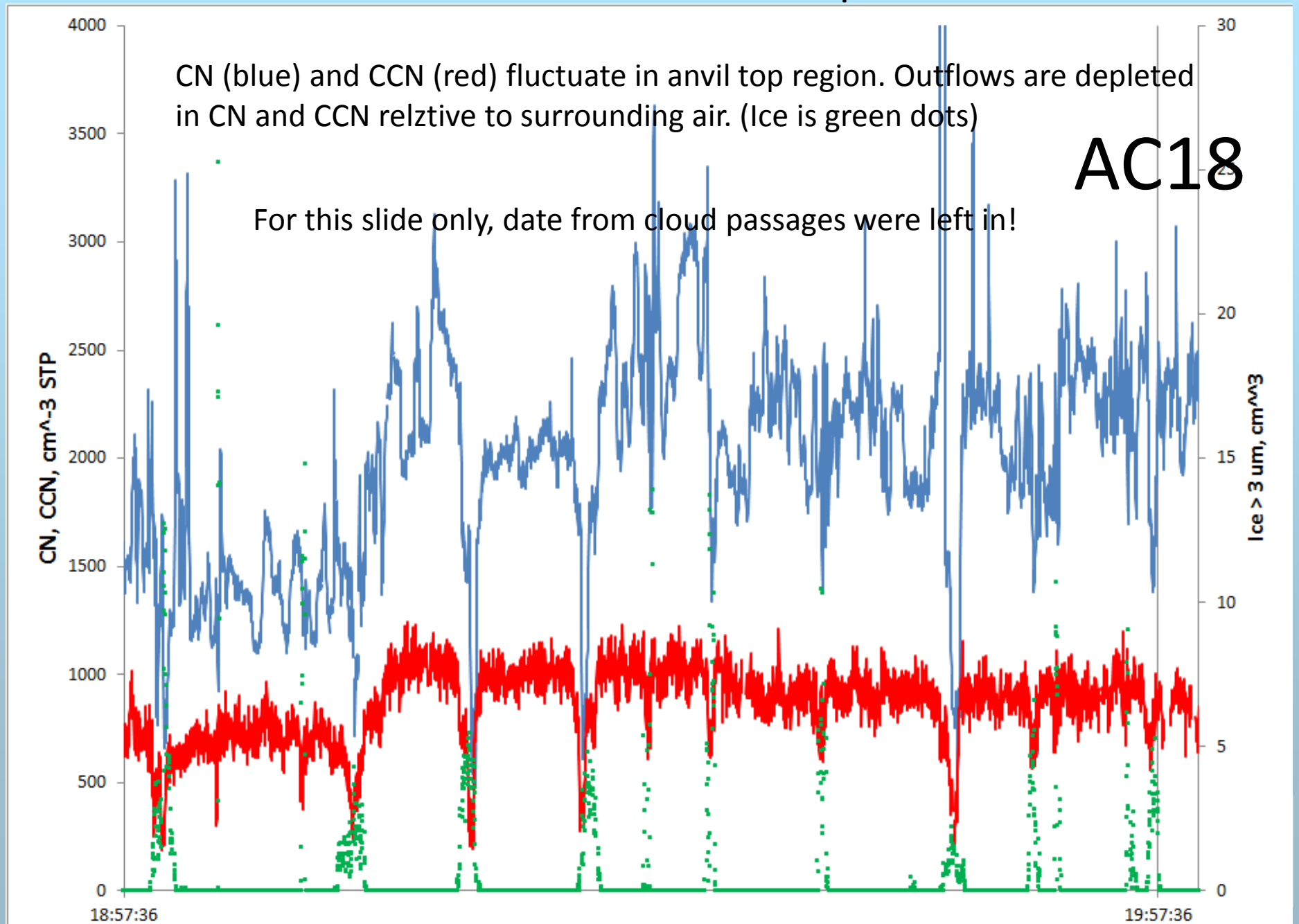


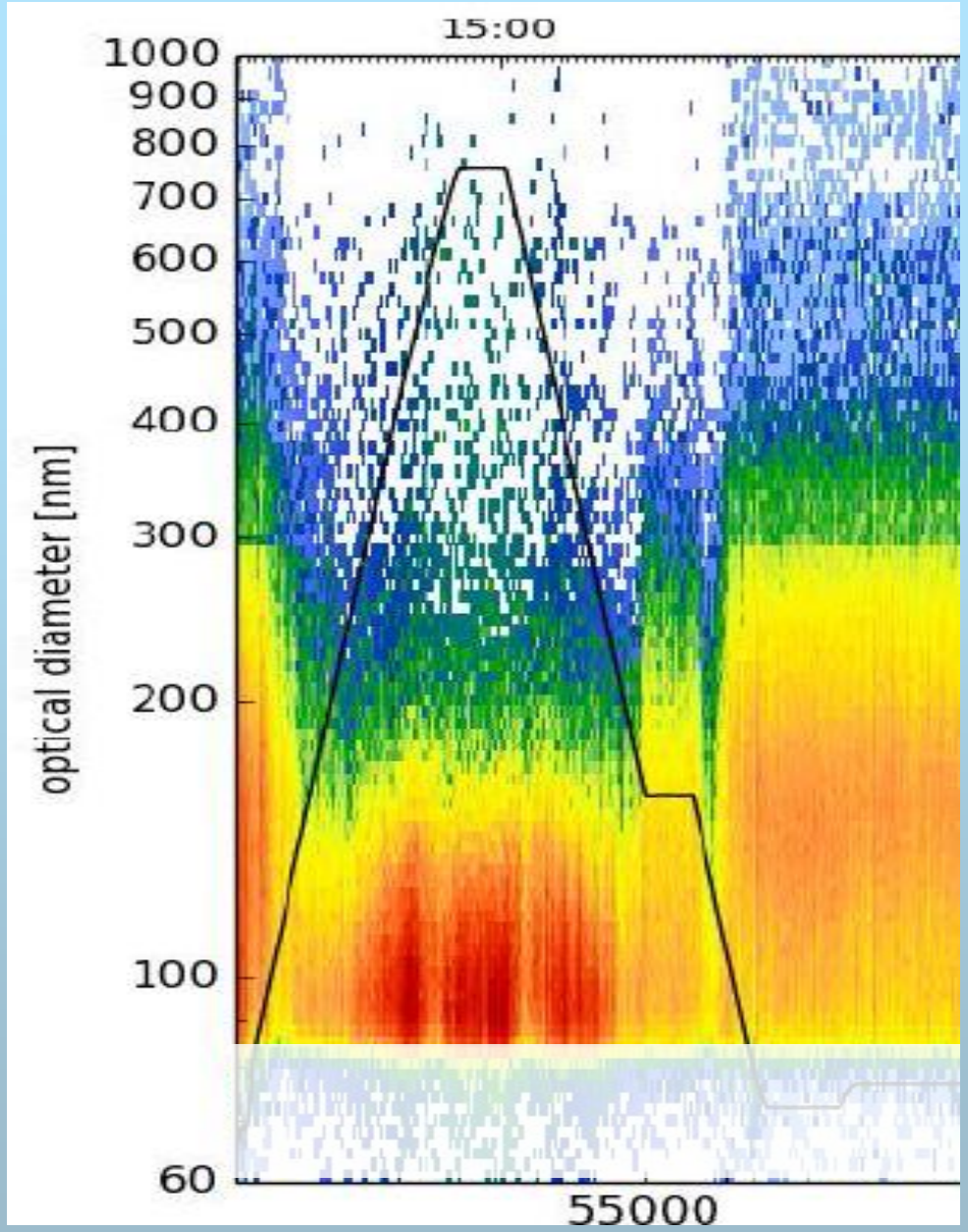

16000
14000
12000
10000
8000
6000
4000
2000
0
Altitude, m
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
Volatile Fraction
Series1
A
B&C&D
E2
AC18
The small particles in the enriched layers are also highly volatile, especially again in layer E2


AC18
CO: enriched in PBL, especially the region nearer Manaus
also in some elevated layers, esp. layers at A
also in the cloud passages, esp at 12 km
also in layer E2
Altitude, m
CO, ppb
0
2000
4000
6000
8000
10000
12000
14000
0
20
40
60
80
100
120
140
160
180
200
Series1
A
E&F
B&C&D


CN show minima in the actual outflow → particles are not exhausted from the cloud!
CN
UHSAS
CCN(0.5%)
ice
CN(>20 nm), CN(>80 nm), cm-3
Ice particles, cm-3
Time, UT
25000
20000
15000
10000
5000
0
50
40
30
20
10
0
19:35:00
19:40:00
19:45:00

CN and CCN at anvil tops





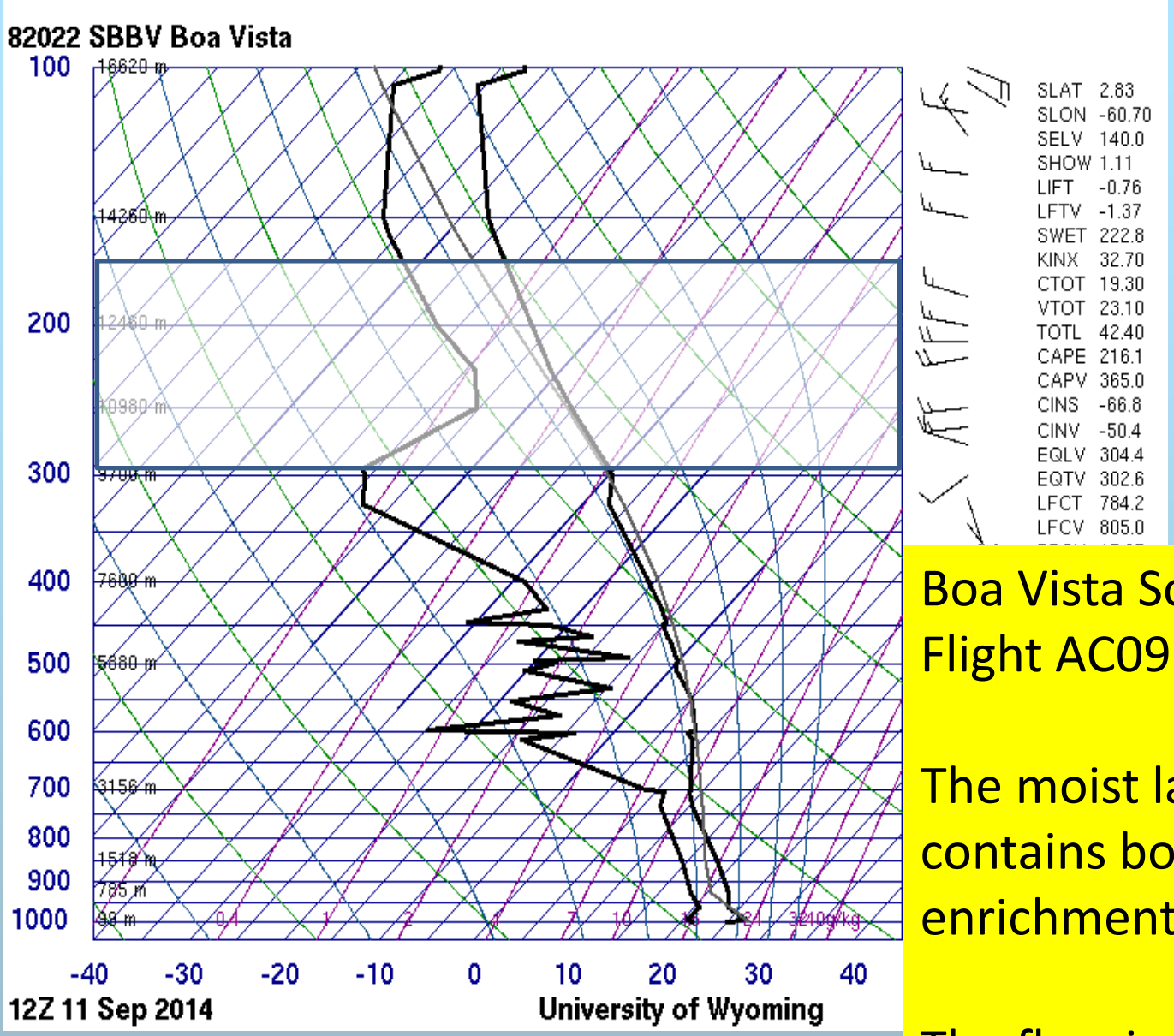
Data from a profile far from convective activity show same characteristics

UHSAS shows that particles in the UT are much smaller than in the PBL, and much more numerous (Unfortunately, there are no SMPS data available!)

Particles are **not**:

- From PBL by convection
- From pollution LRT
- Detrained from clouds

Where are they from??

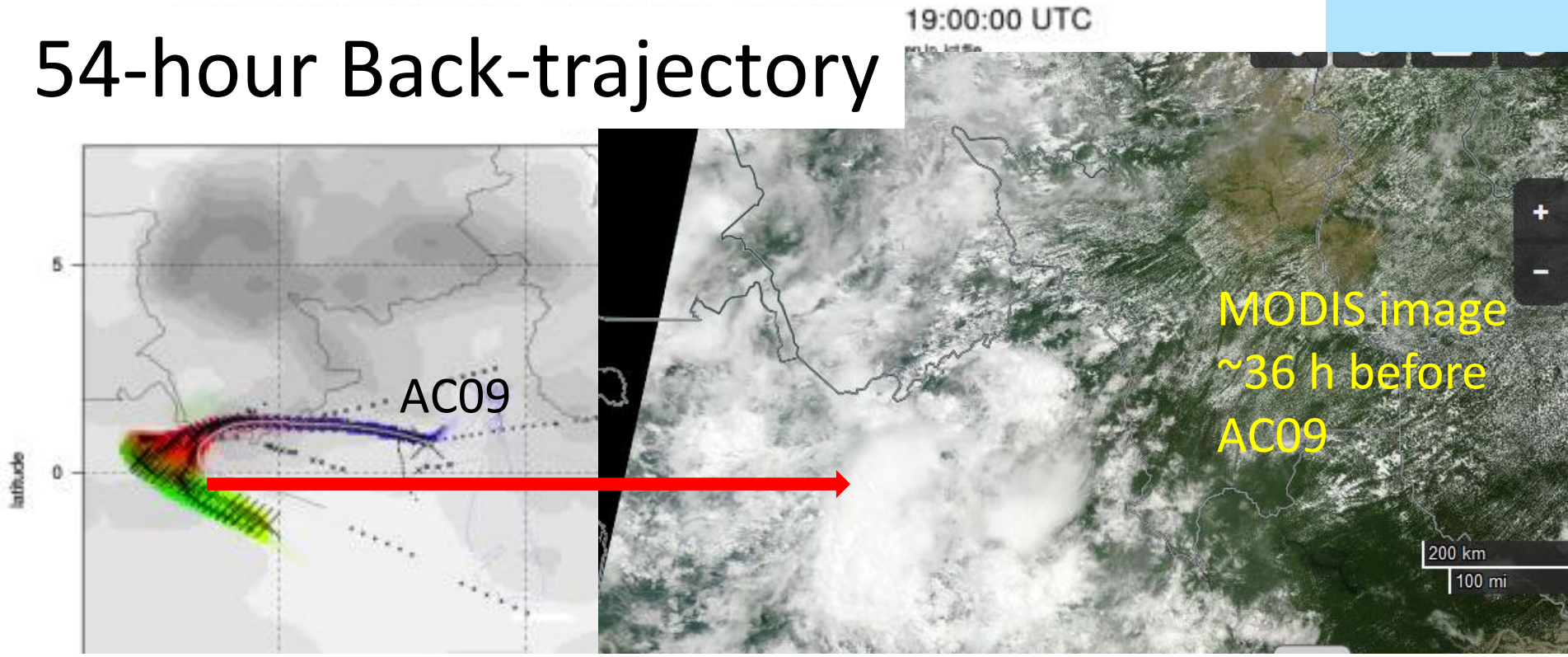


Boa Vista Sounding during Flight AC09

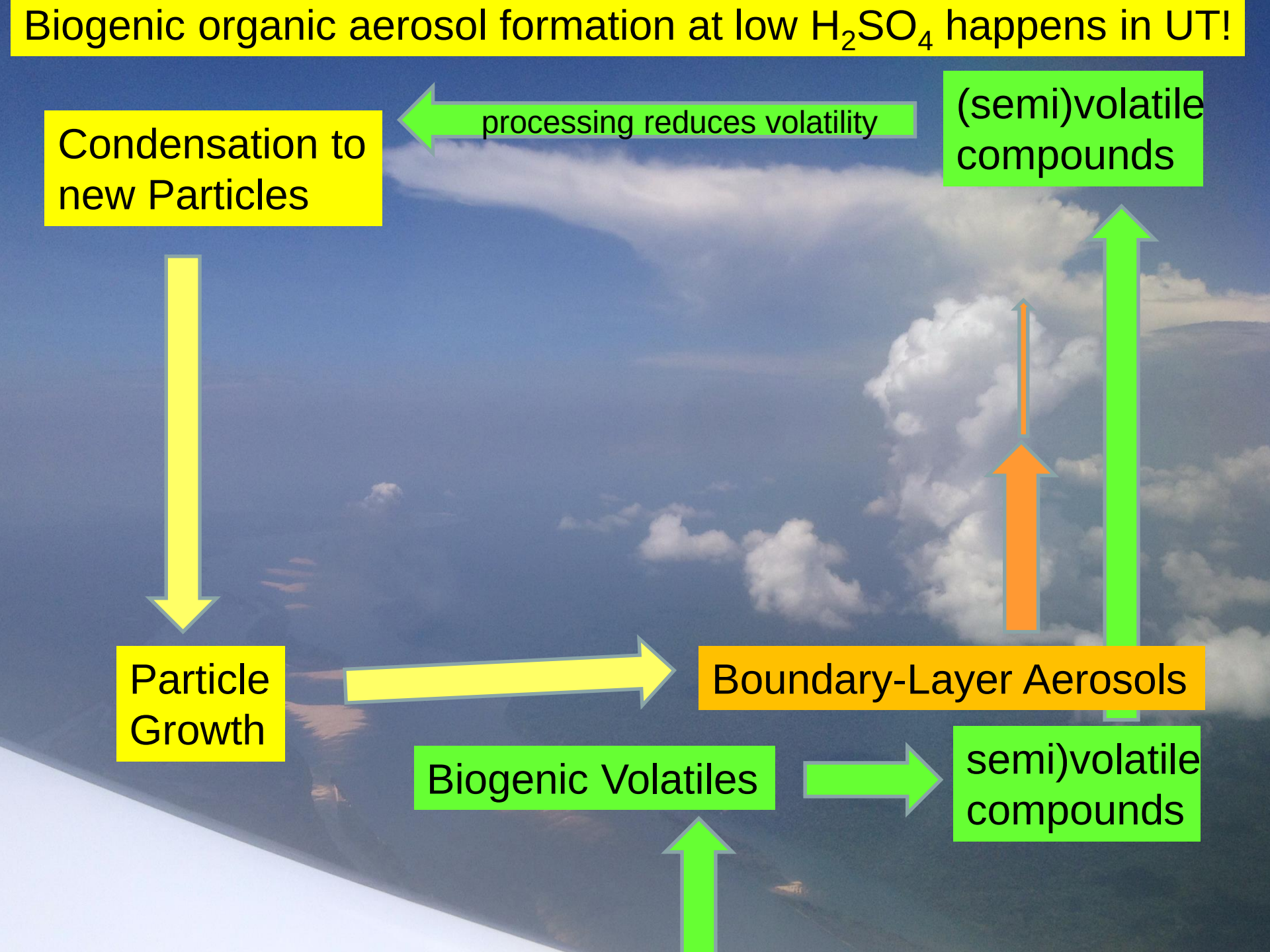
The moist layer above 10 km contains both the aerosol enrichment and lower O_3.

The flow in this layer is westerly.

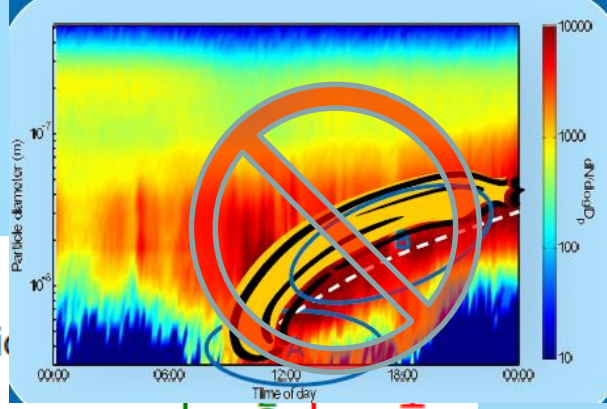
54-hour Back-trajectory

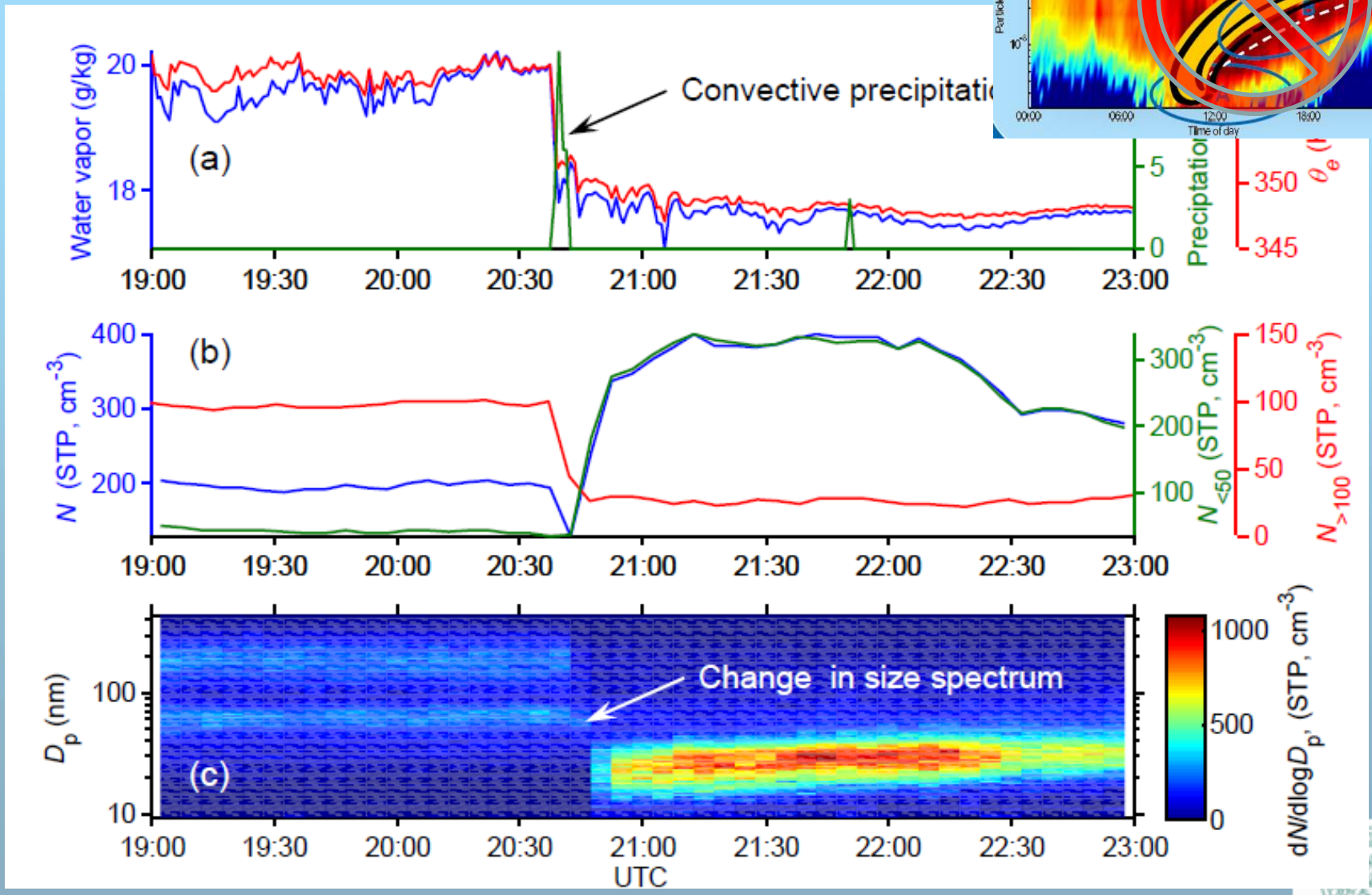


About 24-36 hours before being encountered by HALO, this air was in a highly convective region near the Venezuela/Colombia border!


Biogenic organic aerosol formation at low H_2SO_4 happens in UT!
Condensation to new Particles
processing reduces volatility
(semi)volatile compounds
Particle Growth
Boundary-Layer Aerosols
Biogenic Volatiles
semi)volatile compounds

Yes, we have no bananas!





Wang et al. (2016, under review)

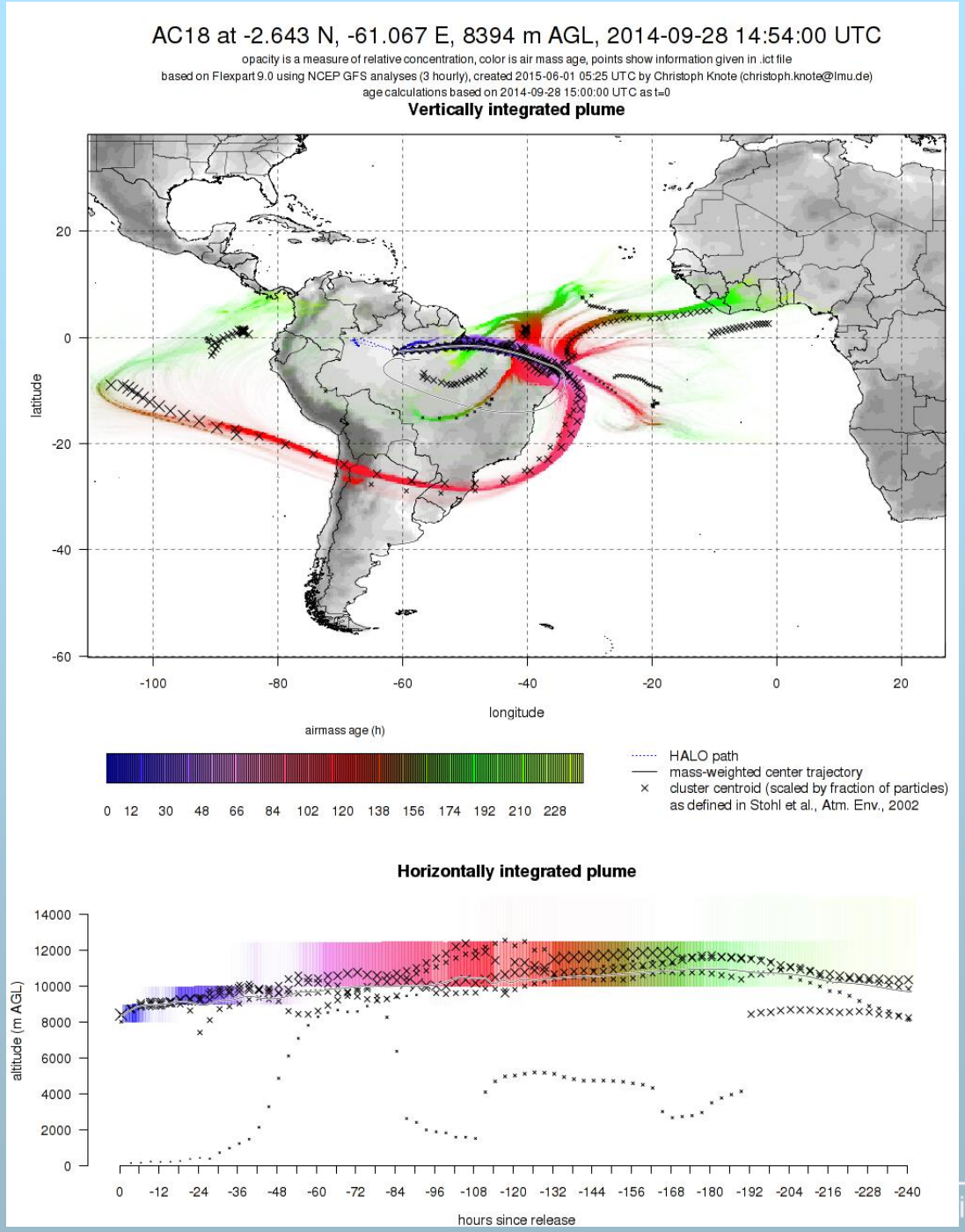
Conclusions

- Regions with strongly elevated aerosol concentrations were found in the upper troposphere all across the Amazon Basin
- These aerosols are distinct from the PBL aerosols by smaller size, high organic carbon and nitrate, low sulfate, absence of BC, and much higher particle concentrations
- They occur in moist layers with low O_3, which originate in regions with convective activity in the preceding days
- Convective transport removes preexisting aerosols and brings (S)VOCs into regions with low temperature, conducive to nucleation and new particle formation even w/o H_2SO_4
- Following detrainment, aerosols condense over a 24-36 h period, producing small particles, but also CCN sized aerosols
- Downward mixing supplies small particles to PBL, where they grow into the CCN range

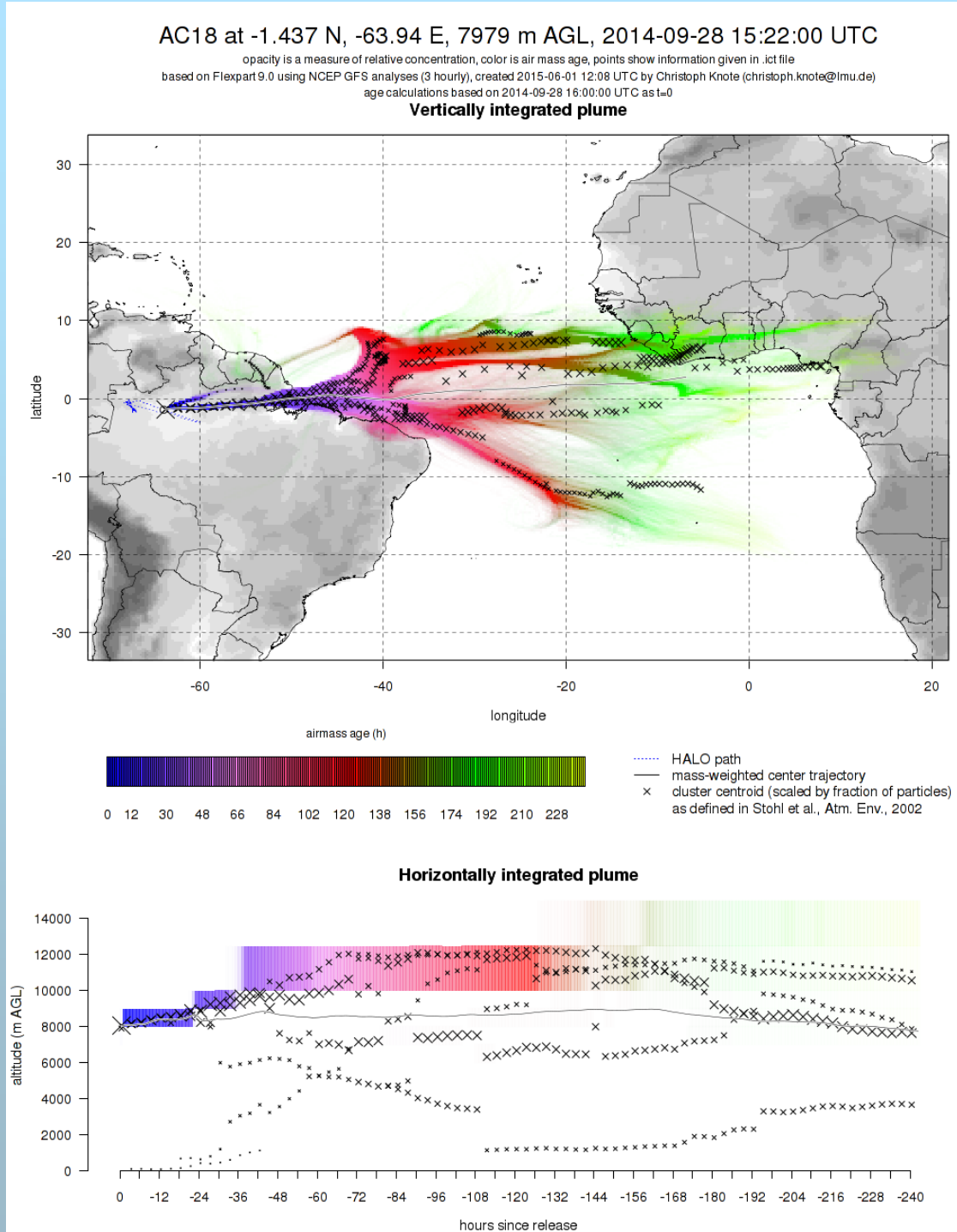
Thank you for your attention!

Airmass histories of layers

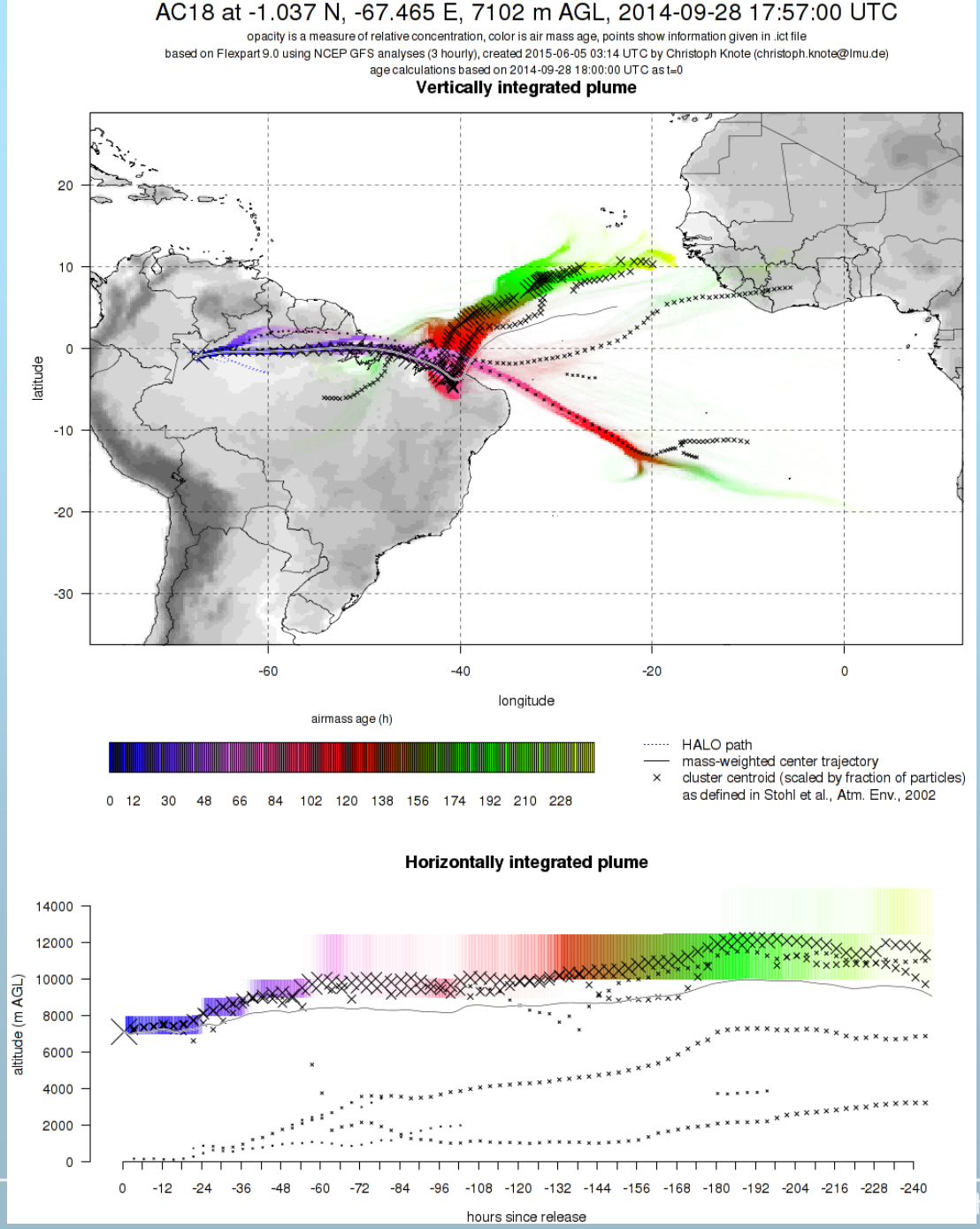
- next pages have trajectories taken with start point at aircraft location at center of layers
- Trajectories from layers A, B, and C do not intercept obvious convection on MODIS images, but obs on previous flights suggest convection in central basin when trajectories pass through it. Luiz, Rachel, can you please check for convection along these trajectories?
- Trajectories from other layers pass trough convection during preceding 1-3 days



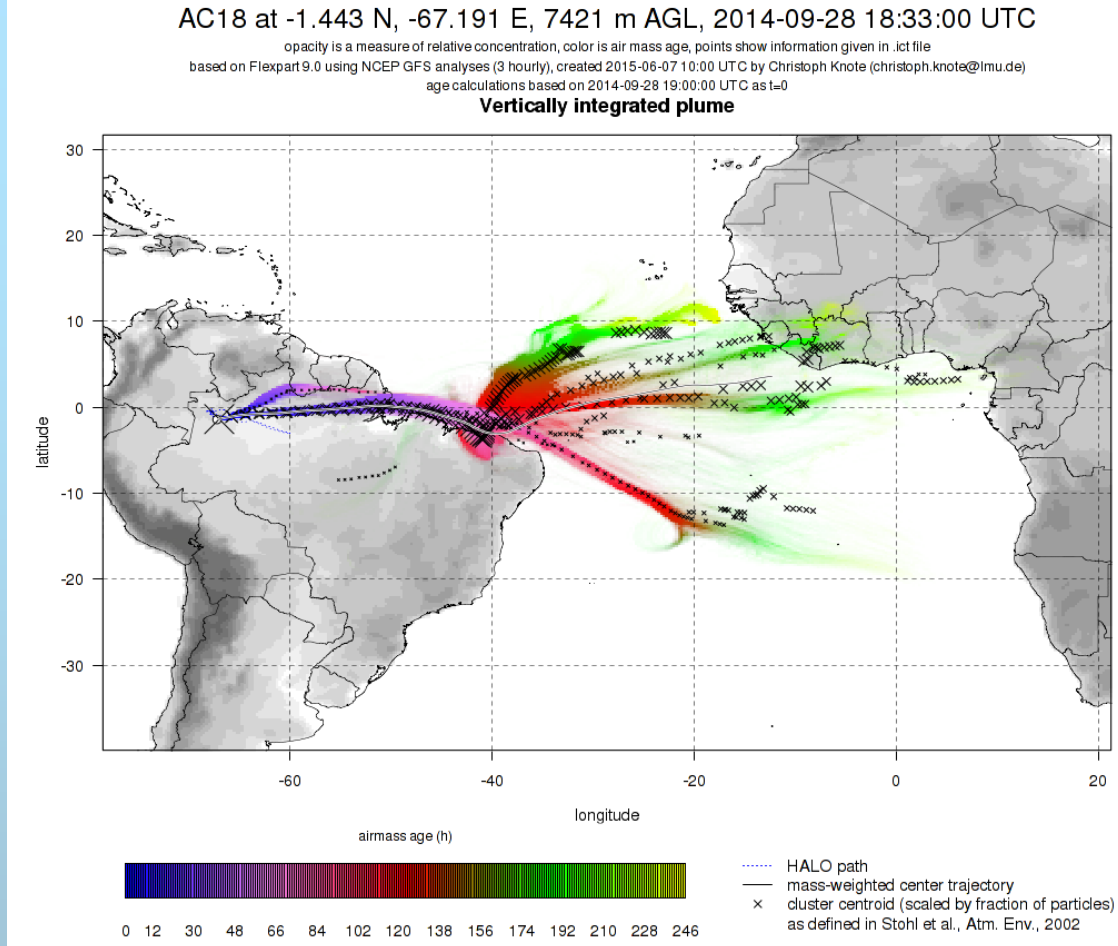
Layer A1, climb



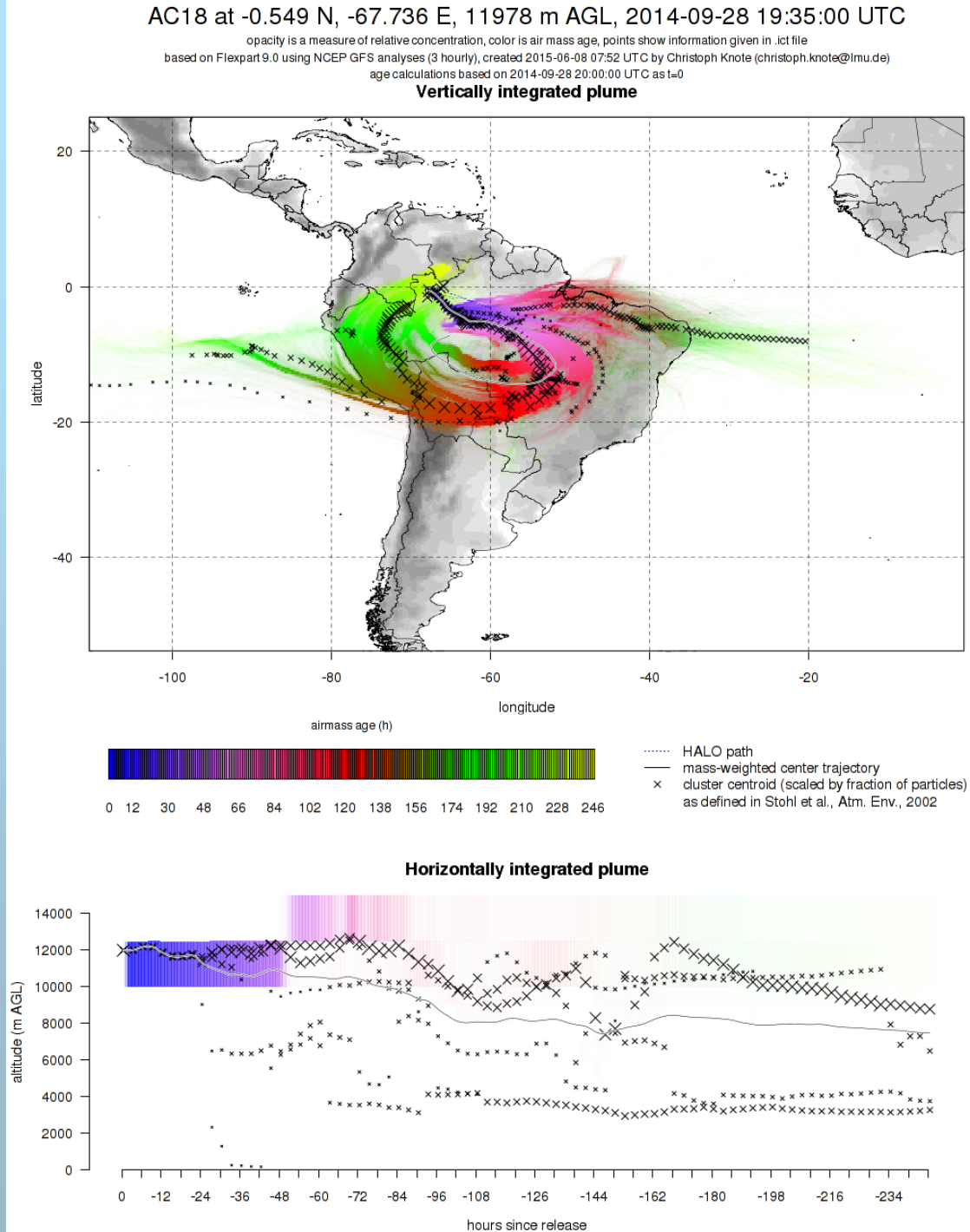
Layer A2, descent


AC18 at -1.037 N, -67.465 E, 7102 m AGL, 2014-09-28 17:57:00 UTC
opacity is a measure of relative concentration, color is air mass age, points show information given in .ict file
based on Flexpart 9.0 using NCEP GFS analyses (3 hourly), created 2015-06-05 03:14 UTC by Christoph Knote (christoph.knote@lmu.de)
age calculations based on 2014-09-28 18:00:00 UTC as t=0
Vertically integrated plume
latitude
longitude
airmass age (h)
0 12 30 48 66 84 102 120 138 156 174 192 210 228
HALO path
mass-weighted center trajectory
cluster centroid (scaled by fraction of particles)
as defined in Stohl et al., Atm. Env., 2002
Horizontally integrated plume
altitude (m AGL)
hours since release

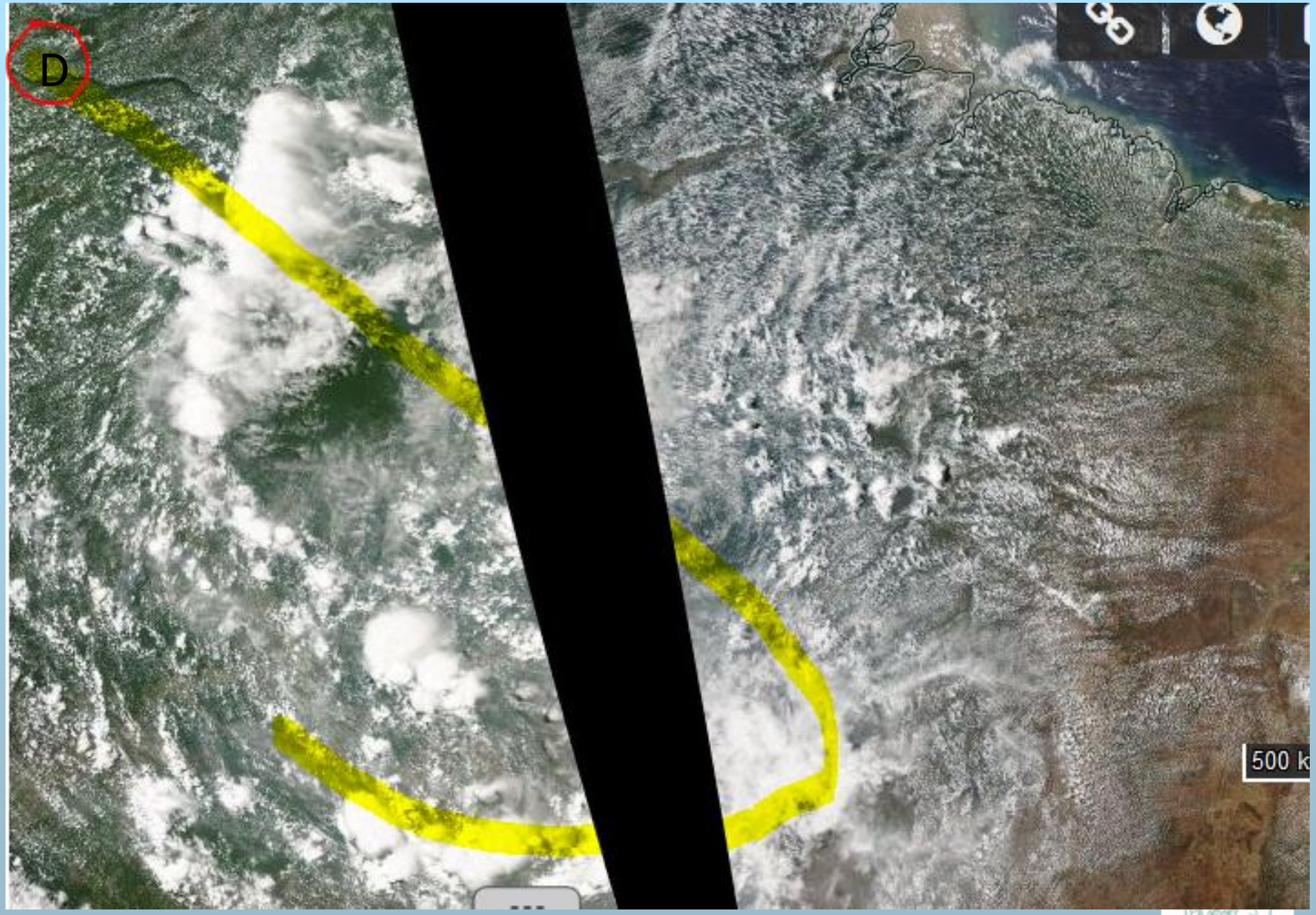
Layer B


AC18 at -1.443 N, -67.191 E, 7421 m AGL, 2014-09-28 18:33:00 UTC
opacity is a measure of relative concentration, color is air mass age, points show information given in .ict file
based on Flexpart 9.0 using NCEP GFS analyses (3 hourly), created 2015-06-07 10:00 UTC by Christoph Knote (christoph.knote@lmu.de)
age calculations based on 2014-09-28 19:00:00 UTC as t=0
Vertically integrated plume
latitude
longitude
airmass age (h)
0 12 30 48 66 84 102 120 138 156 174 192 210 228 246
HALO path
mass-weighted center trajectory
cluster centroid (scaled by fraction of particles)
as defined in Stohl et al., Atm. Env., 2002
Horizontally integrated plume
altitude (m AGL)
hours since release

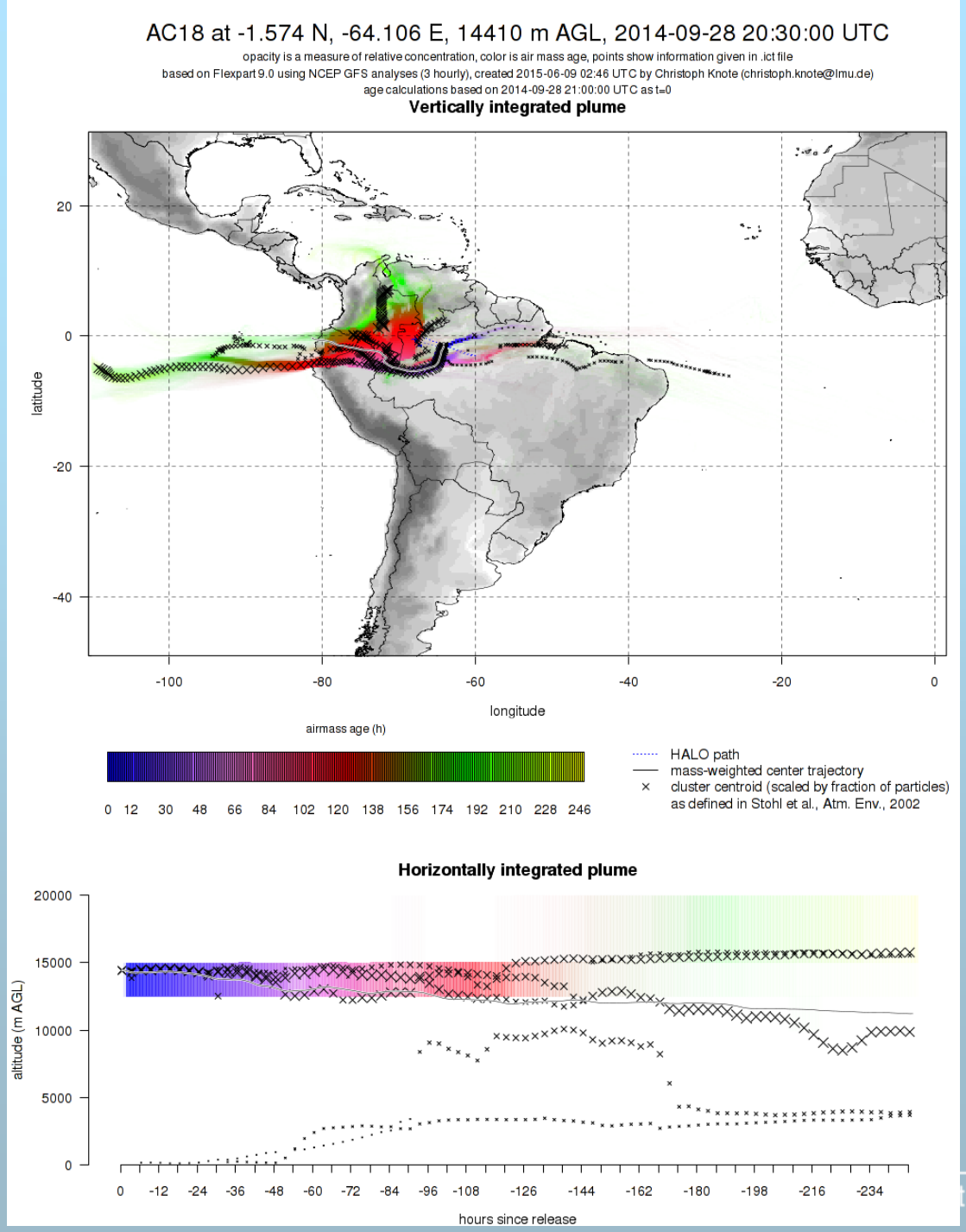
Layer C



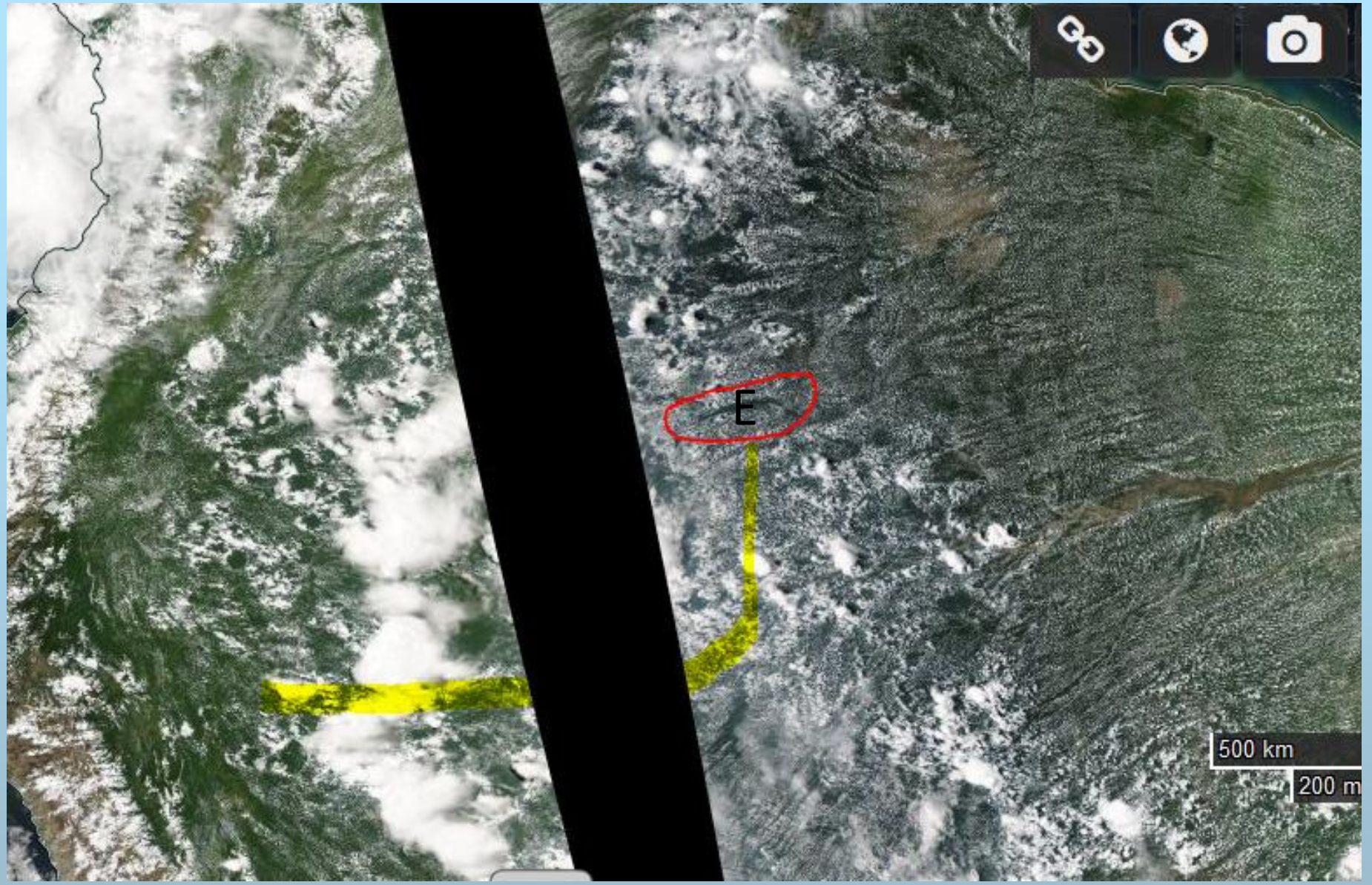
Layer D, cloud top region



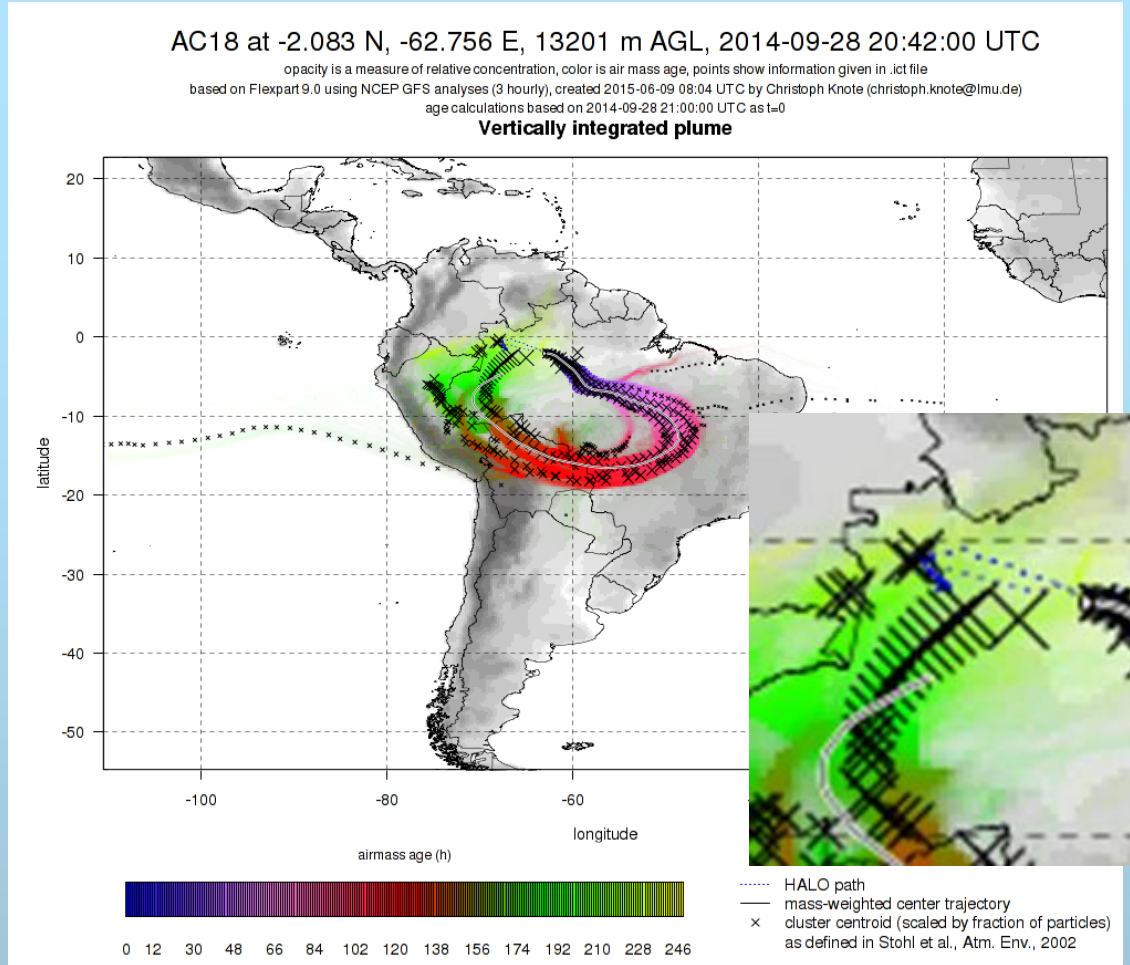
AQUA, 24 Sept.

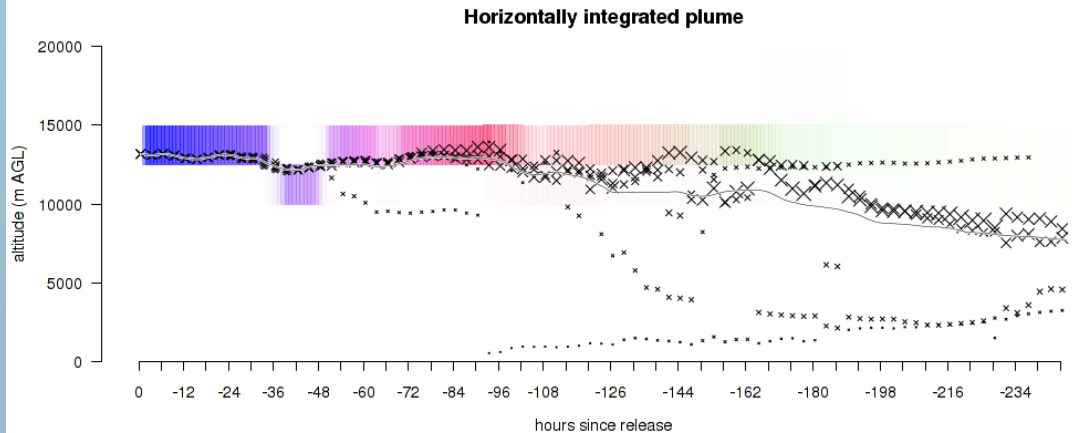


Layer E, uppermost later, around 14 km



E: uppermost layer comes from convection in westernmost part of Basin, consistent with cleaner air (low O3 and CO)

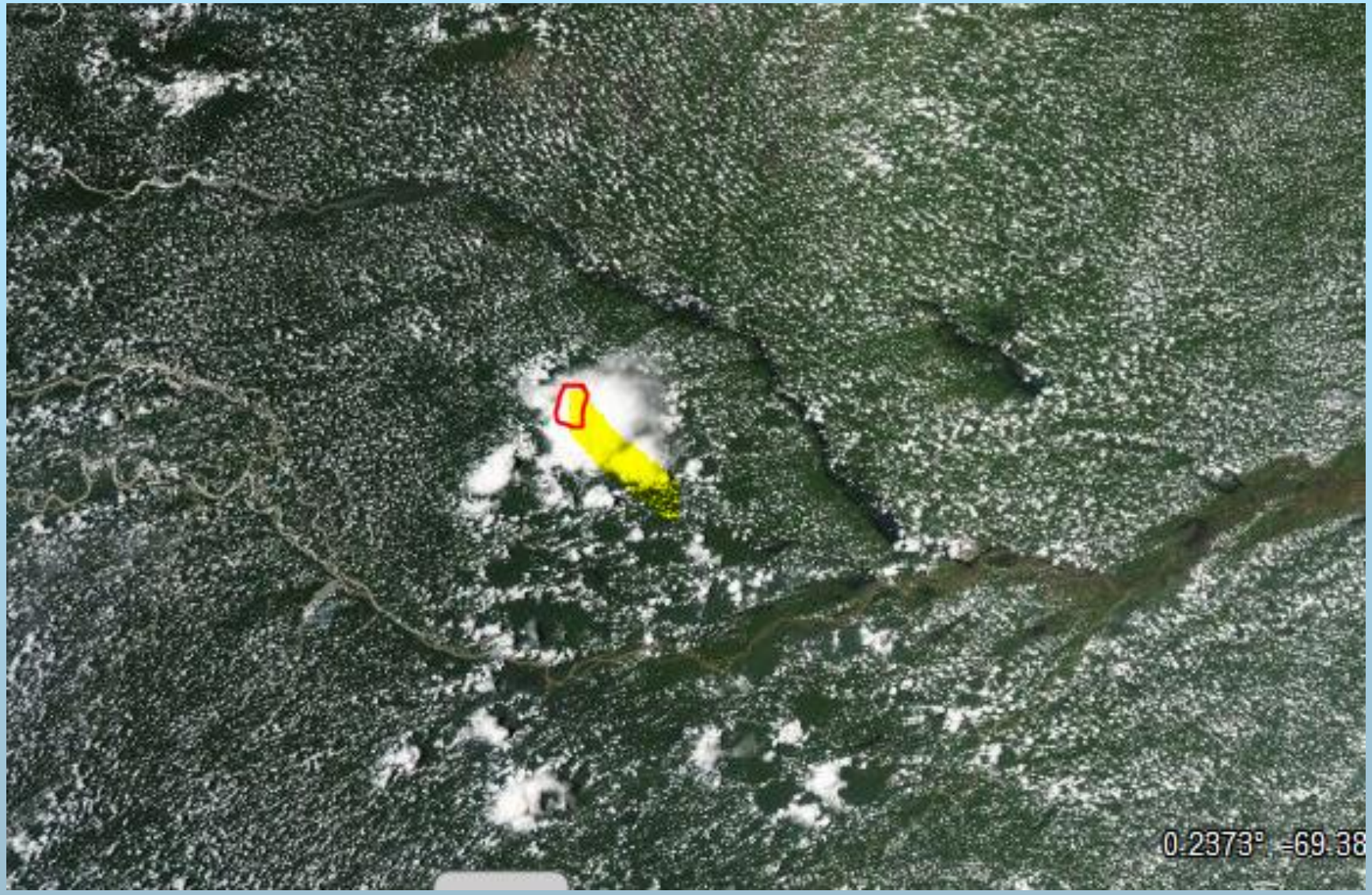




Layer E2, thin layer with very high aerosol conc. at ~13 km

new thoughts

- aerosol results from oxidation of volatiles in the UT (Jian Wang)
- rapid HOM formation by mixing w/ O3 rich air (Ehn et al. 2014; Jokinen et al 2015)
- rapid HOM formation by reaction w/ OH (Berndt et al., Science, submitted)
- Gordon et al, PNAS submitted
- presence of elevated SO2 in UT



E2 might have come directly from the super-large cB that we were passing on its northern side. Very high NO suggests recent origin or air.