GCA1-Meeting, Harvard-NUIST

Atmospheric aqueous-phase SOA formation: Laboratory investigations and field observations

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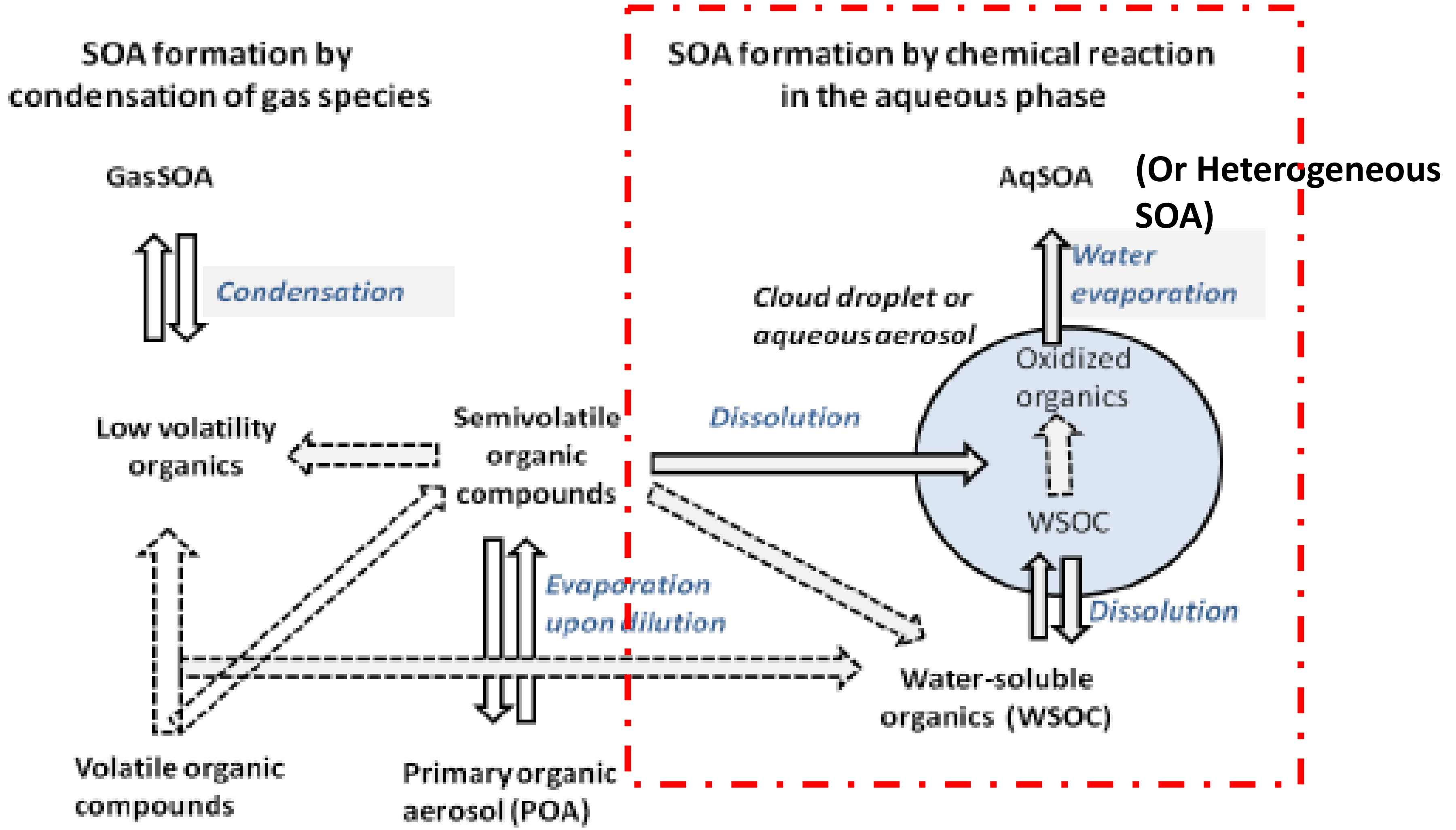
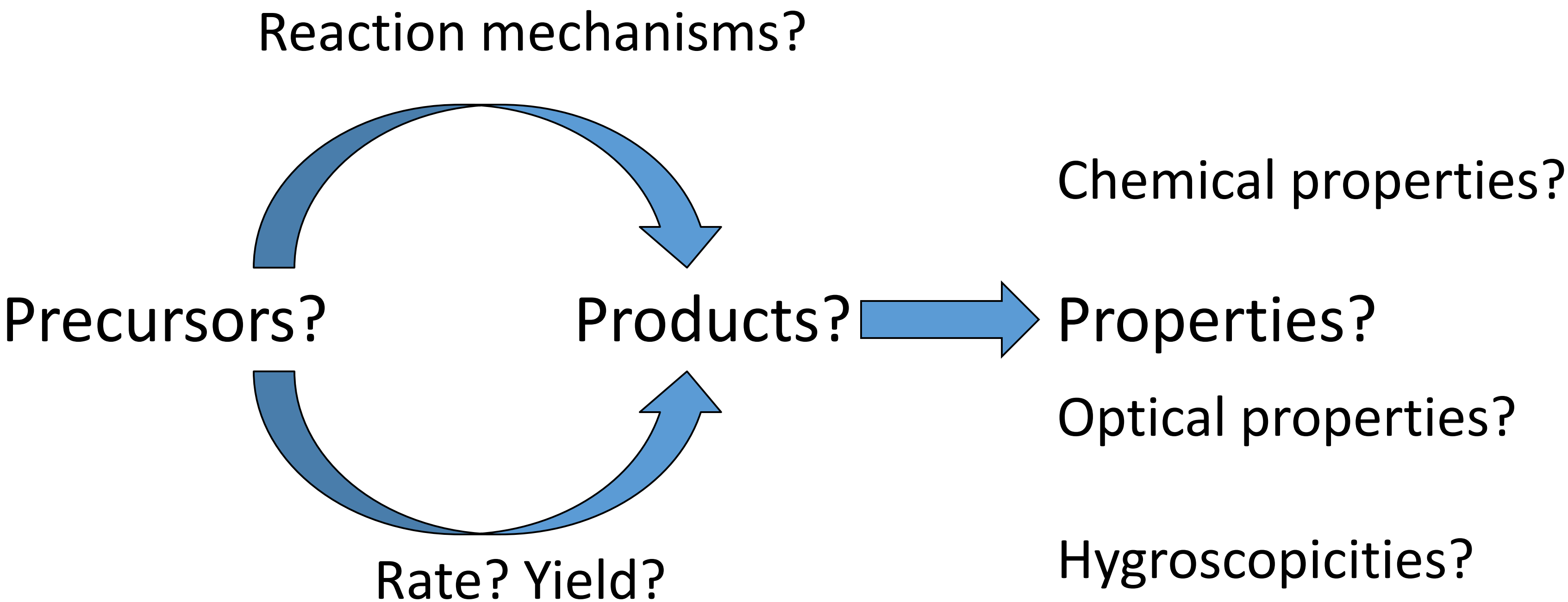
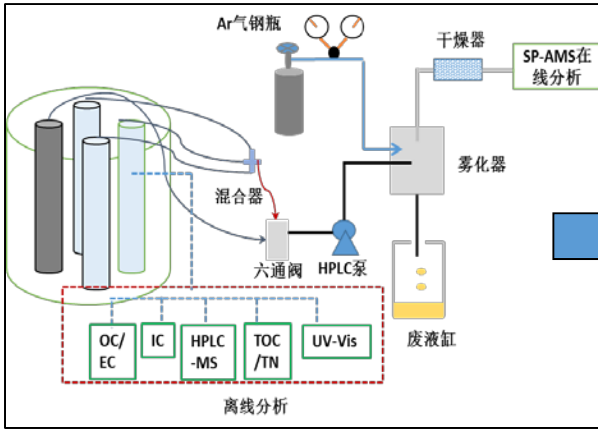
1. Secondary Organic Aerosol(SOA) formation

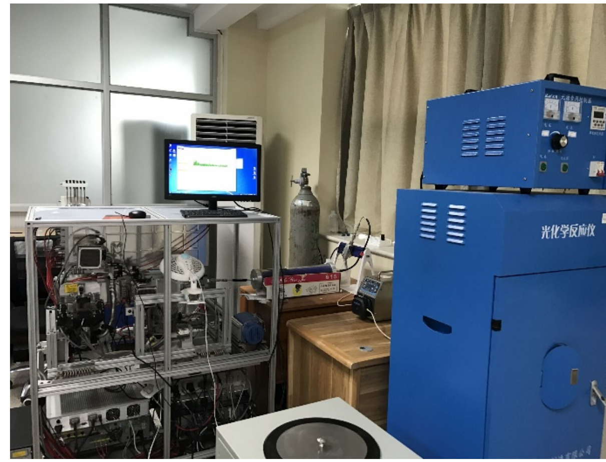


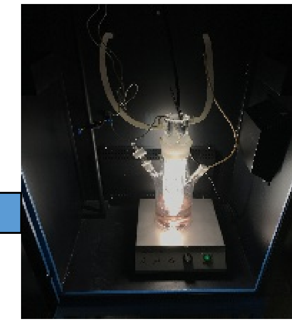
AqSOA formation is less clear, might be significant to reconcile model-observation discrepancies.


Reaction mechanisms?
Chemical properties?
Precursors?
Products?
Properties?
Optical properties?
Hygroscopicities?
Rate? Yield?

2. Laboratory experiments



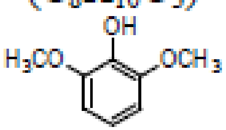




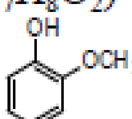
Online/offline characterization: Aqueous-phase reactor + SP-AMS

On biomass burning compounds

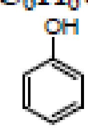
Syringol ($C_8H_{10}O_3$)



Guaiacol ($C_7H_8O_2$)

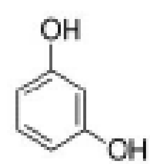


Phenol (C_6H_6O)

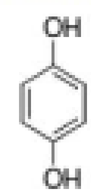


4-Ethylphenol ($C_8H_{10}O$)

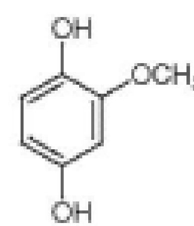
Resorcinol $C_6H_6O_2$



Hydroquinone $C_6H_6O_2$

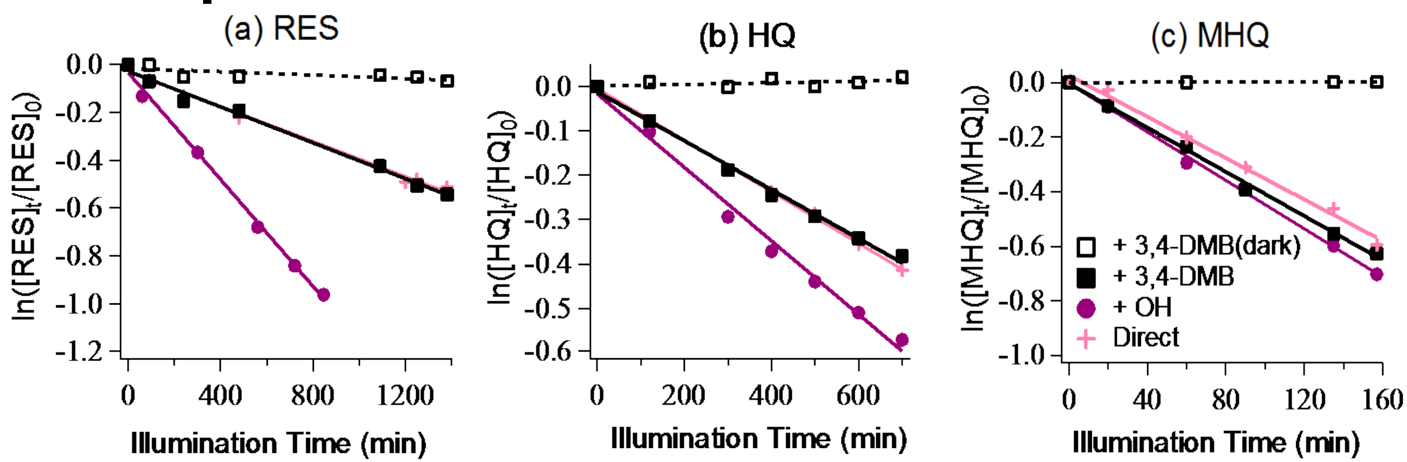


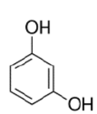
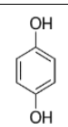
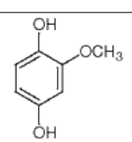
Methoxyhydroquinone $C_7H_8O_3$



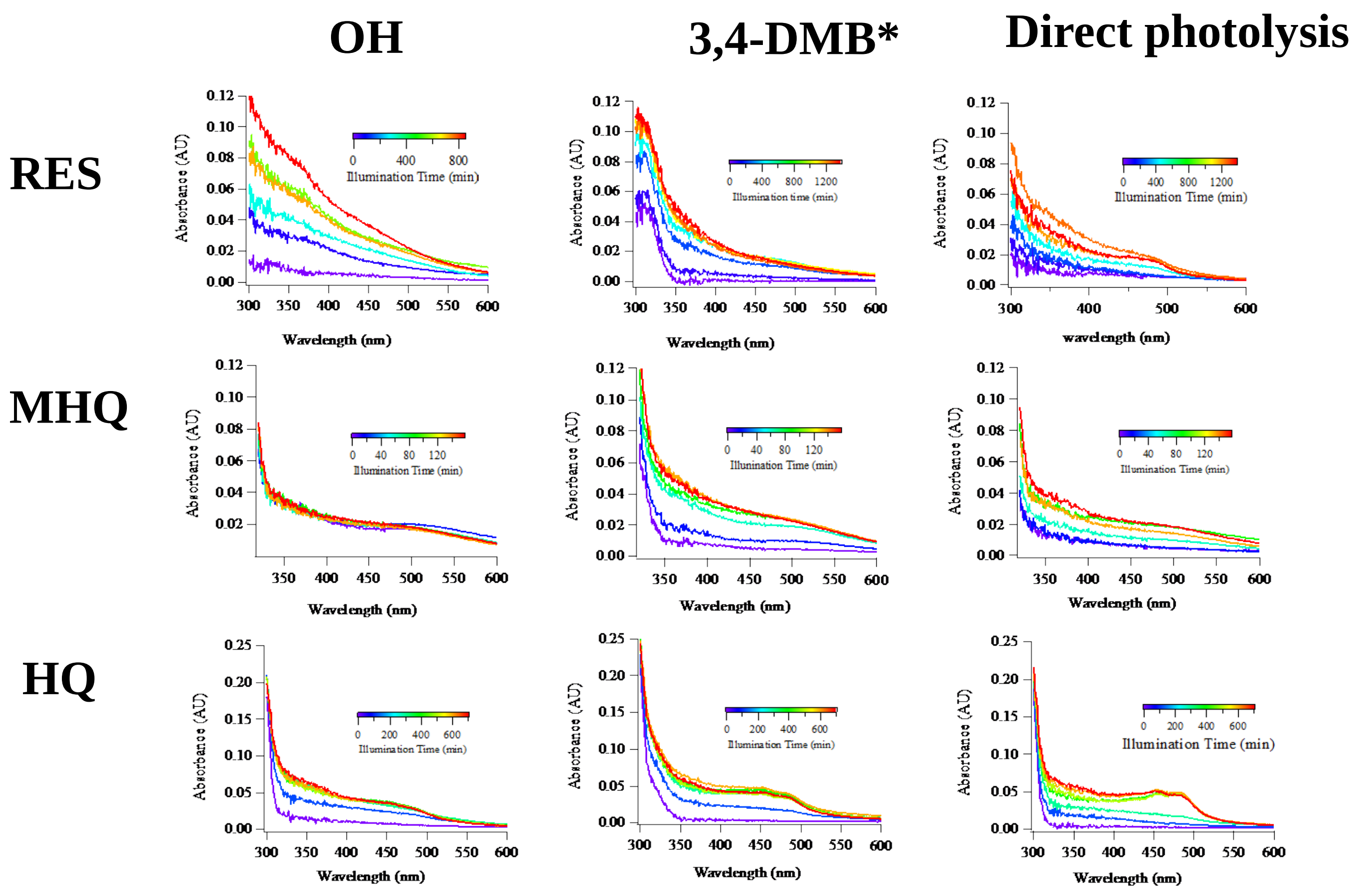
2.1 Photolysis rate

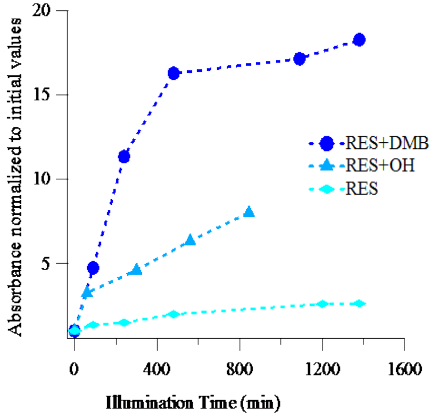
Direct photolysis, OH- and in particular $^3C^*$-initiated reactions (triplet excited states, a unique group of oxidants in interfacial and bulk phases)

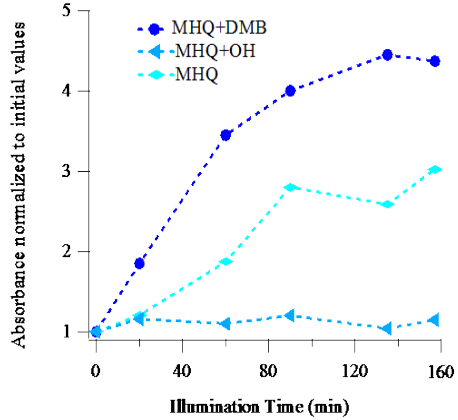


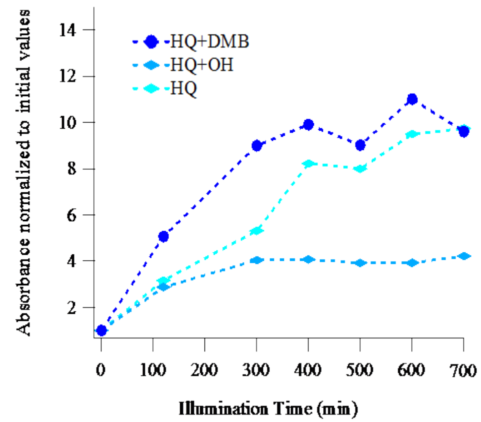
Sample information				
Precusor		Oxidant	$t_{1/2}$ (min)	Rate constant k_d (h^{-1})
Resorcinol $C_6H_6O_2$ 110.11		·OH	763	0.067
		$^3C^*$	1697	0.023
		——	1825	0.022
Hydroquinone $C_6H_6O_2$ 110.11		·OH	967	0.050
		$^3C^*$	1384	0.033
		——	1208	0.035
Methoxyhydroquinone $C_7H_8O_3$ 140.14		·OH	154	0.26
		$^3C^*$	159	0.24
		——	197	0.23

2.2 Light absorption (“Brown Carbon”?)



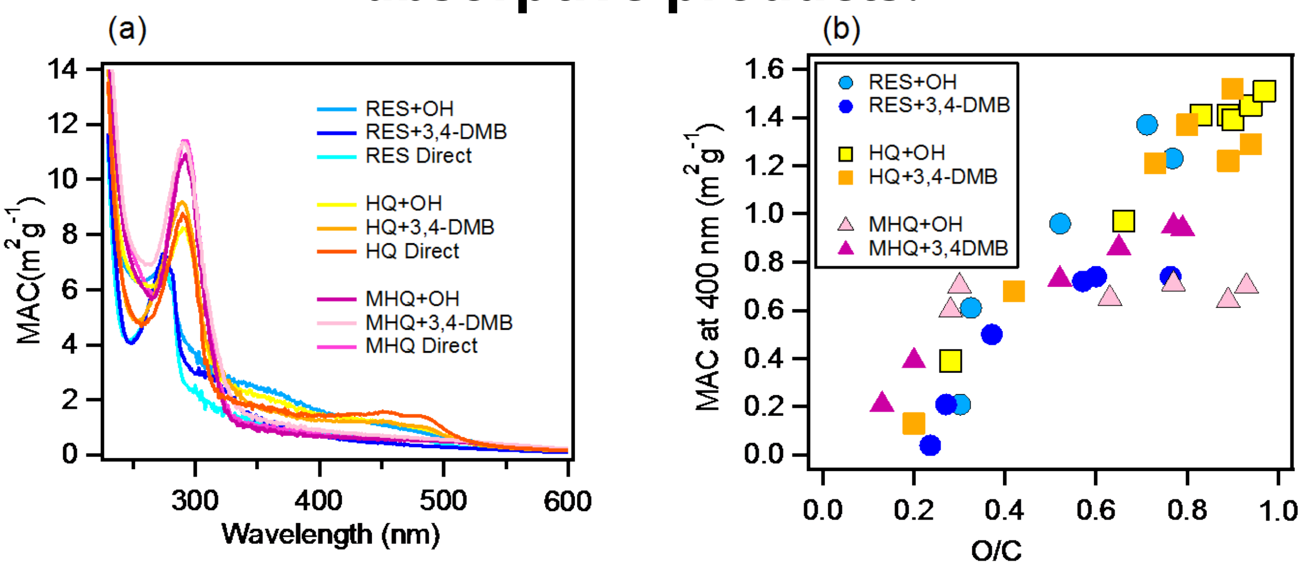






Absorption at 400 nm

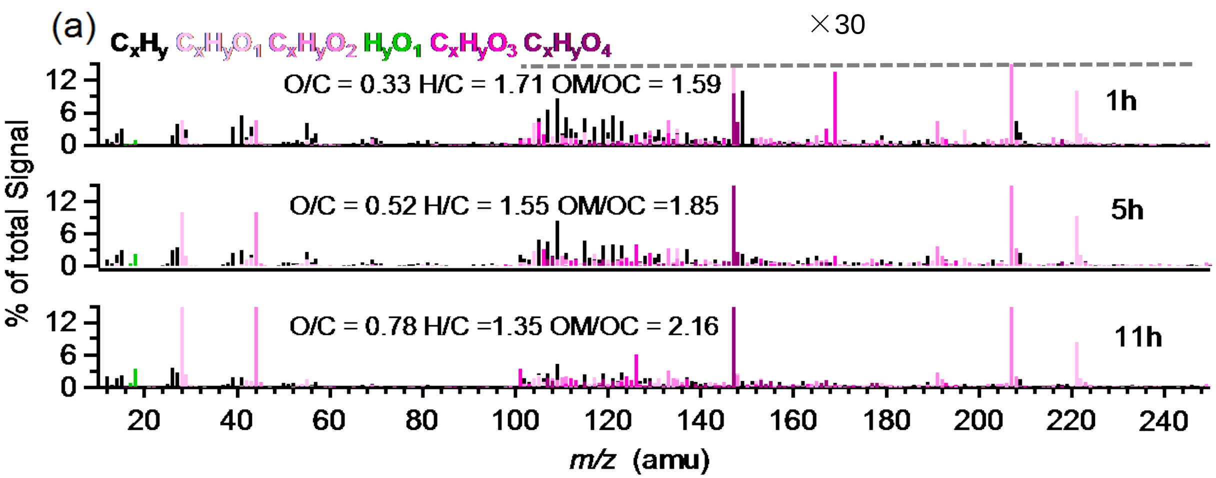
$^3C^*$-oxidation reactions produces more light-absorptive products!



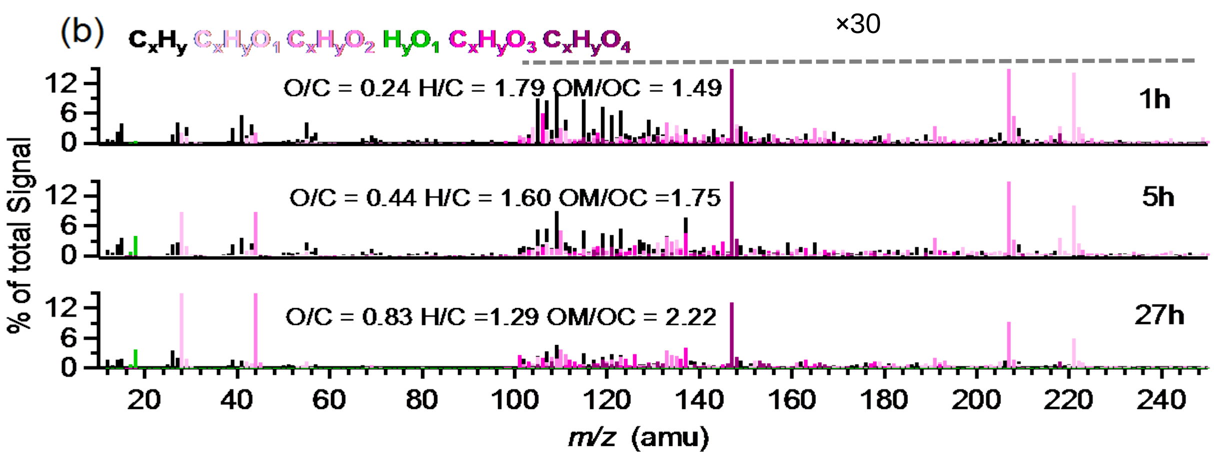
Light absorption increases with SOA oxidation degrees

2.3 Chemical compositions (AMS mass spectra)

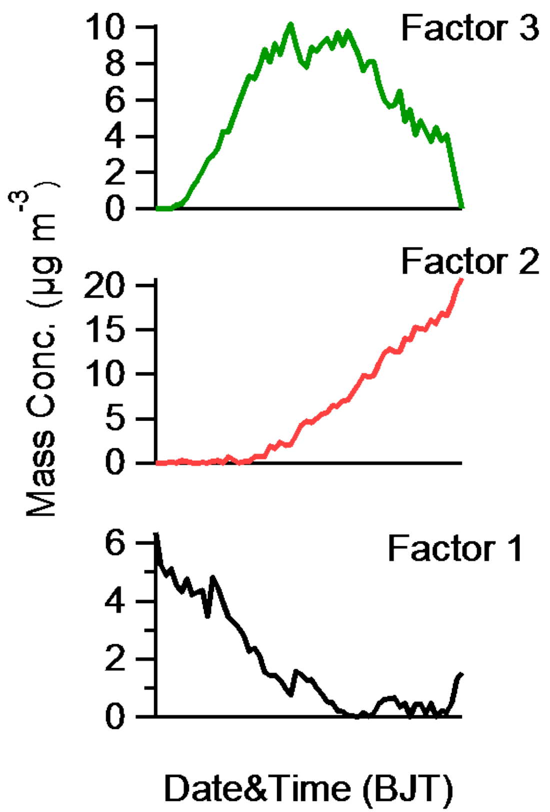
RES+OH

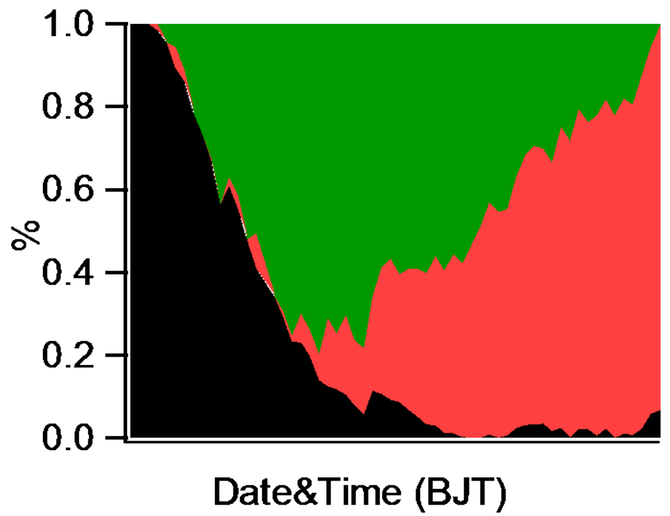


RES+3,4-DMB



PMF analyses





MHQ+OH oxidation

Factor 3— Second-generation (oligomerization then fragmentation).

Factor 2—Third-generation (more small highly oxygenated species)

Factor 1— first-generation

2.4 aqSOA yields

Offline：

$$Y_{SOA}=\frac{\text{mass of illuminated sample}-\text{mass of dark sample}}{\text{mass of phenol reacted}}$$

for 4-EPhOH

10 mL solutions:

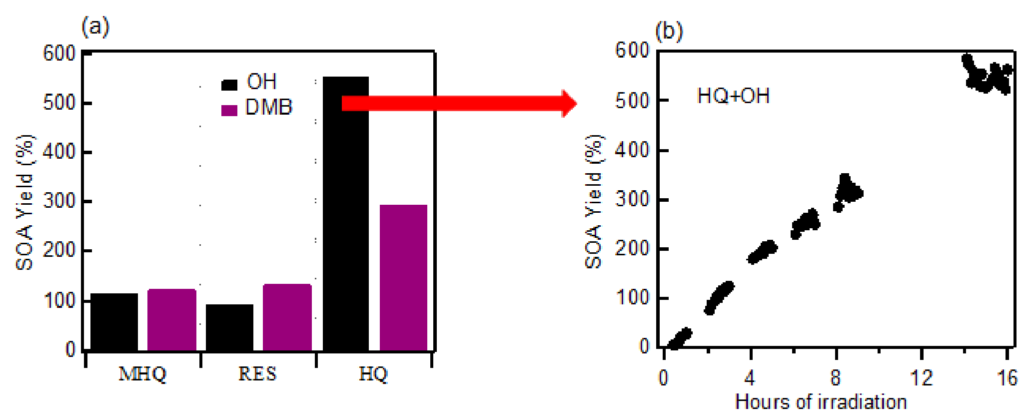
$$Y_{SOA}=\frac{(0.28-0.21)\times10^{3}\mu g}{(0.01/1)\times100\times122.17\times52.68\%}=1.09$$

for GUA

10 mL solutions:

$$Y_{SOA}=\frac{(0.31-0.24)\times10^{3}\mu g}{(0.01/1)\times100\times124.14\times0.5513\mu g}=1.02$$

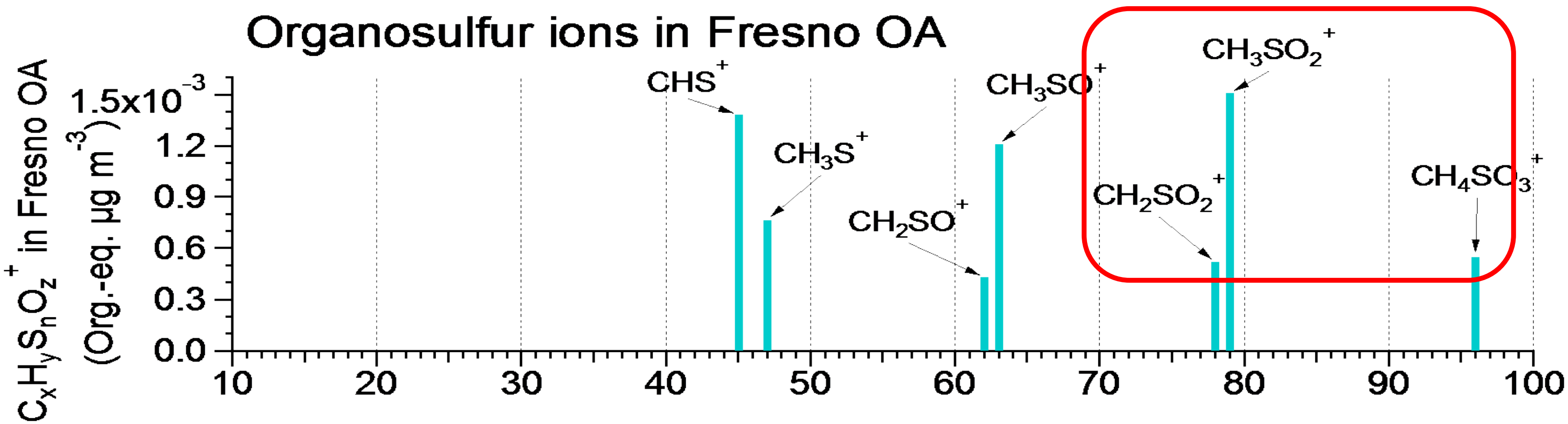
Online：



3. Field observations

aqSOA – Higher O/C, larger size, positive correlations with RH or LWC

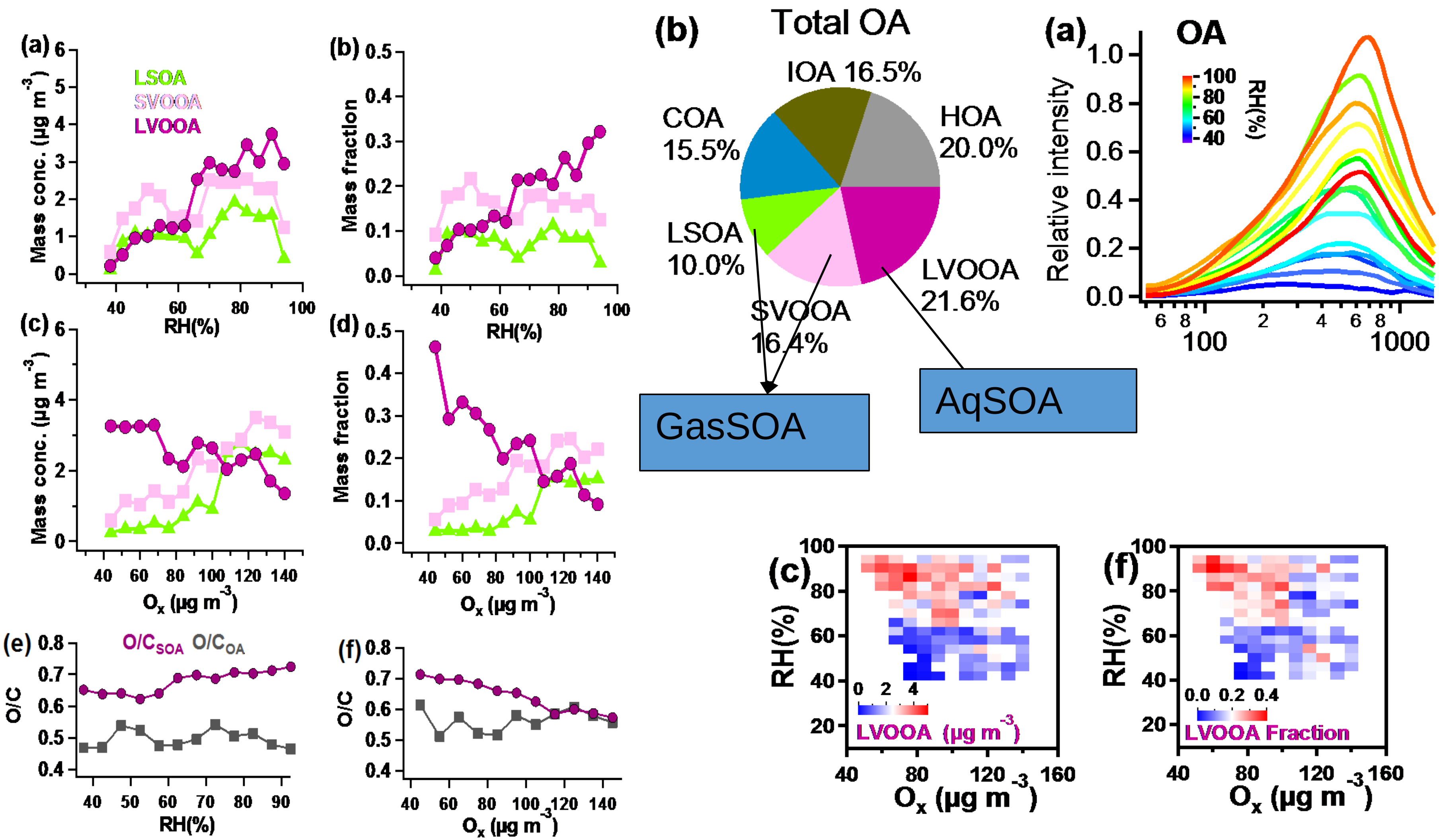
MSA - aqSOA Tracer (Ge et al., EC, 2012)



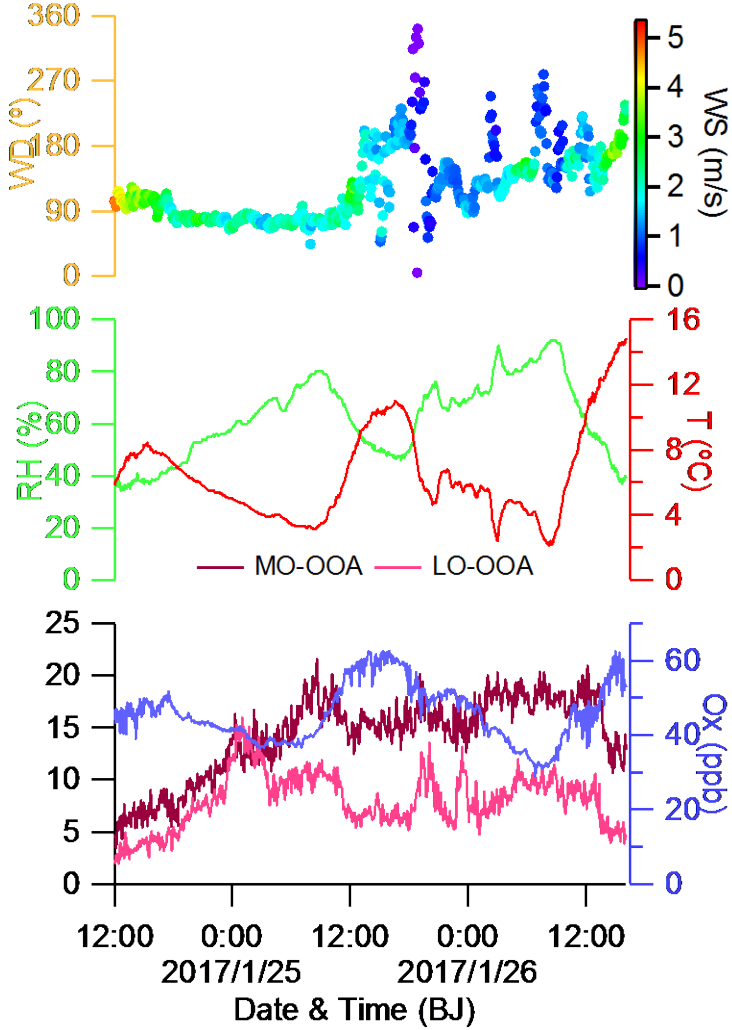
Italy — *Gilardoni et al., PNAS, **2016**, 113, 10013-10018*

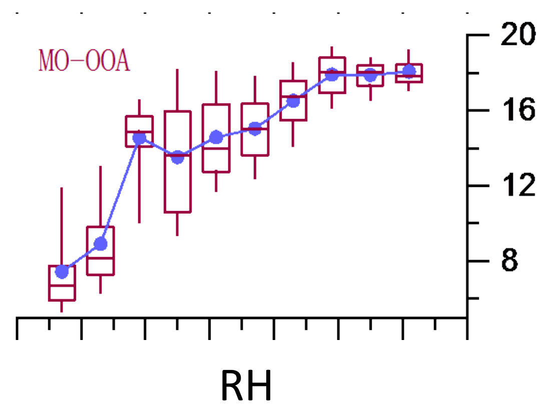
Beijing Winter — *Sun et al., Atmos. Chem. Phys. **2016**, 16, 8309-8329.*

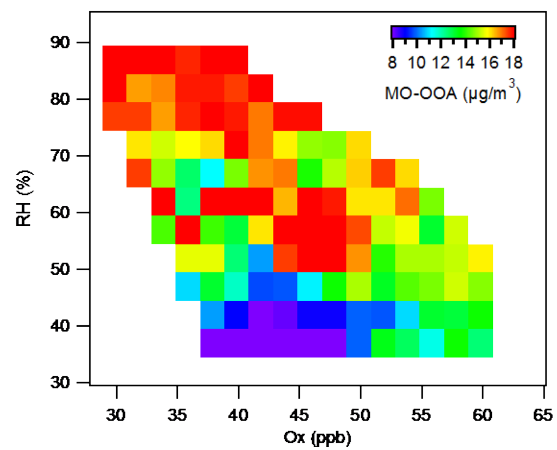
Nanjing 2015 winter



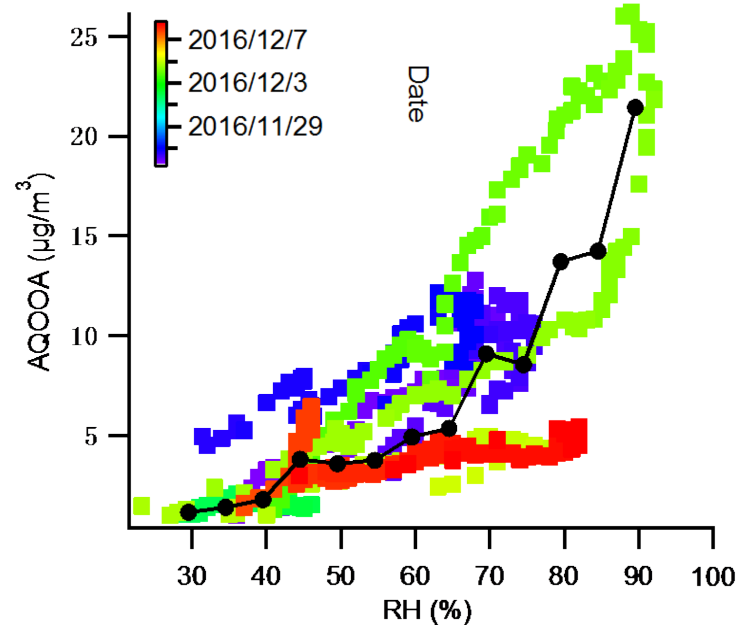
Nanjing 2017 Winter



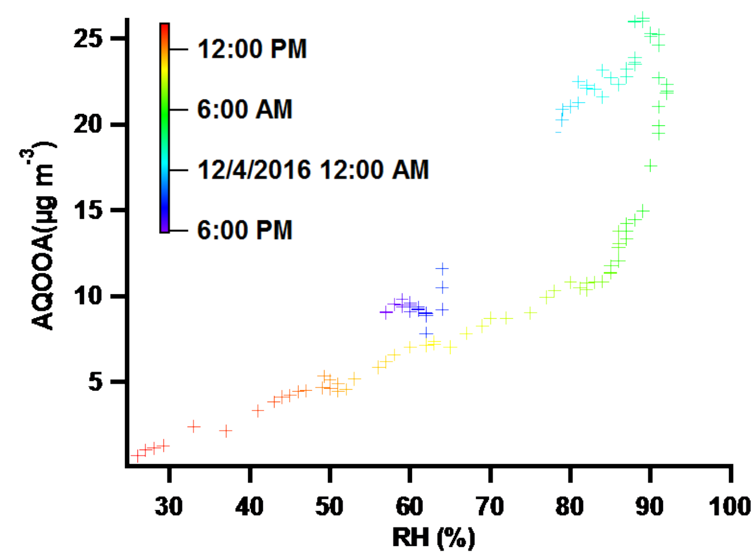




Beijing 2016 Winter

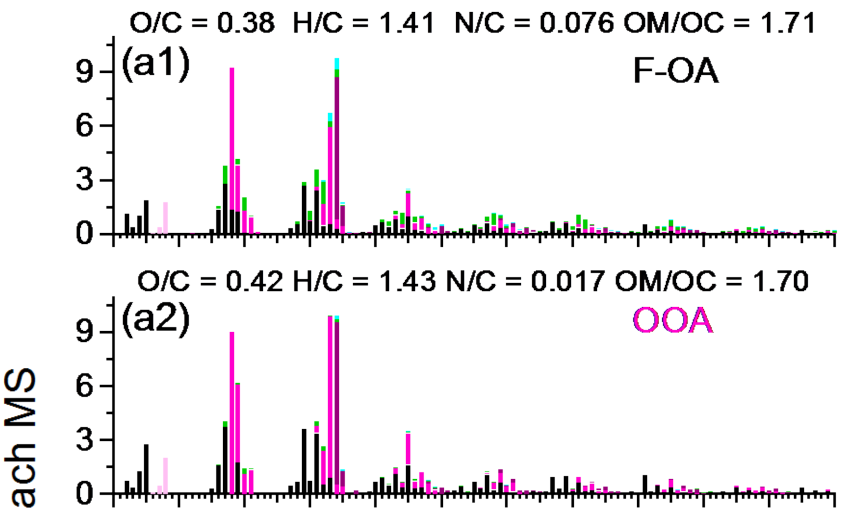


APPH Beijing Winter campaign (2016.11.15-12.14)
SP-AMS for BC-particles

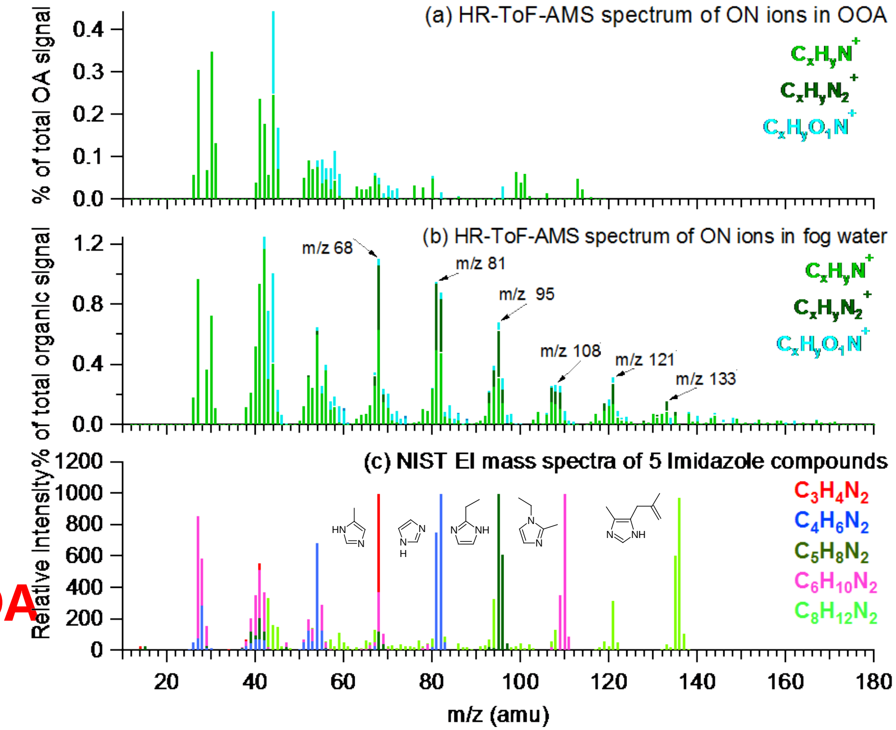


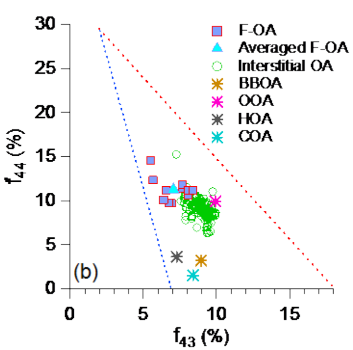
2010 Fresno Winter

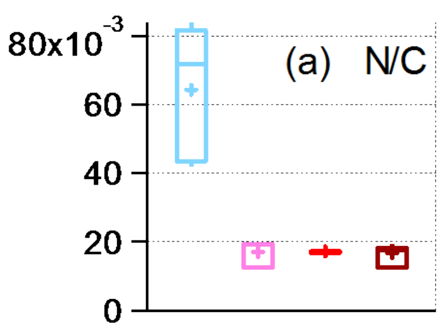
AMS PM_1 + Fogwater



Fog-OA is highly similar to PM_1-OOA

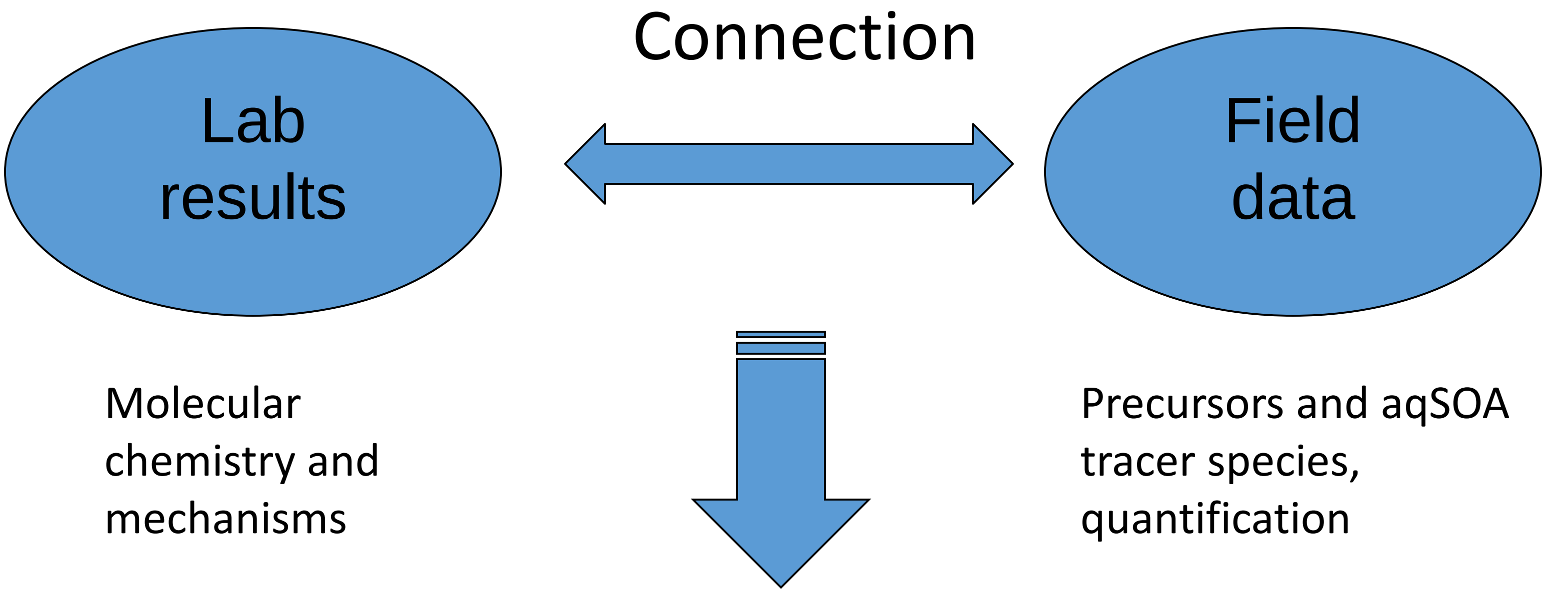






More N-organics in Fogwater-OA, imidazoles (likely from Aq-rxns)

4. Future work



Model updates and improvements

Thanks！

Group members

Aerodyne SP-AMS

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