New methodology for deriving PM2.5 chemical composition using GEOS-Chem & High Spectral Resolution Lidar

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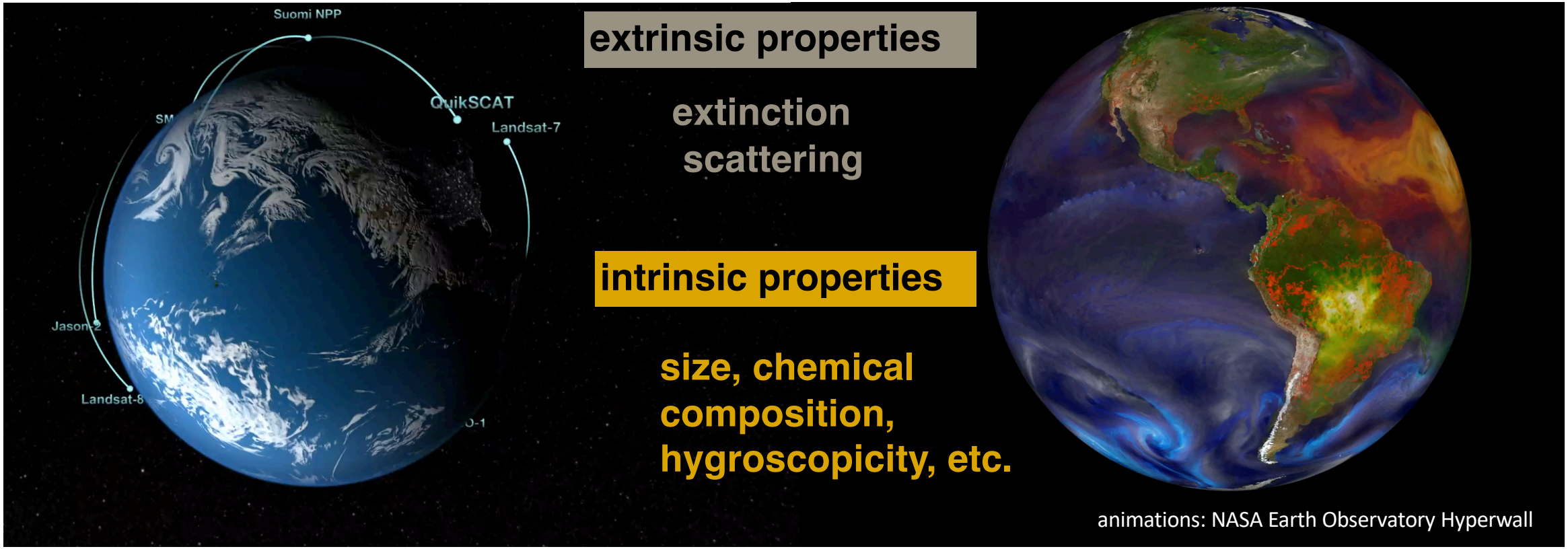
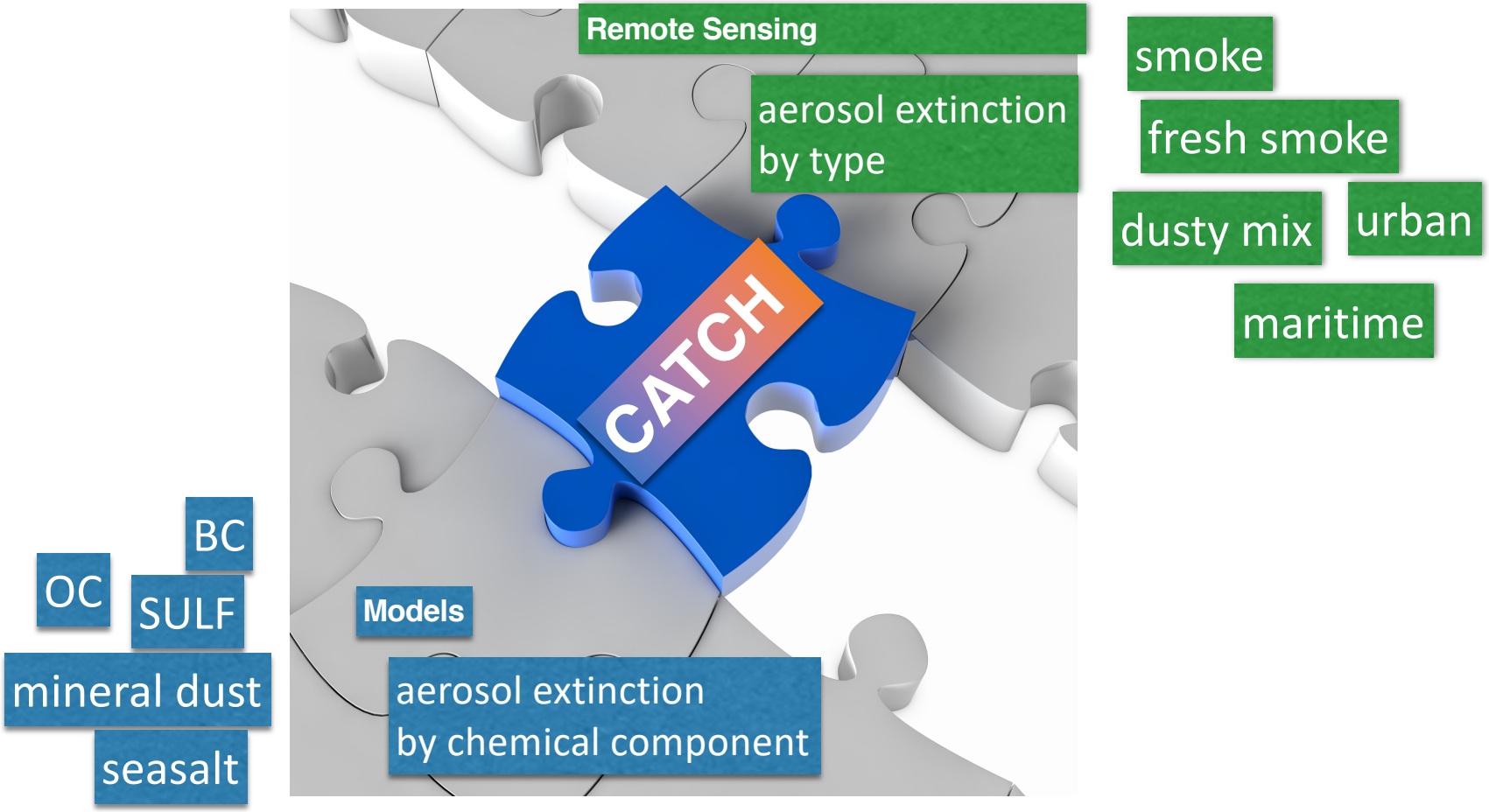
Satellites vs models



There is a constraint at the top (AOD) and through the column (extinction), but not for aerosol microphysics and not for chemical composition

To create a link, we derive aerosol types with GEOS-Chem



Creating **A**erosol **T**ypes from **CH**emistry (CATCH)

GEOS-Chem → M_{OC}; M_{BC}; M_{SULF}; M_{DUST}; M_{SS}

HSRL Aerosol Types

Unique groups ("clusters") are quantitatively described by the group's mean and covariance matrix

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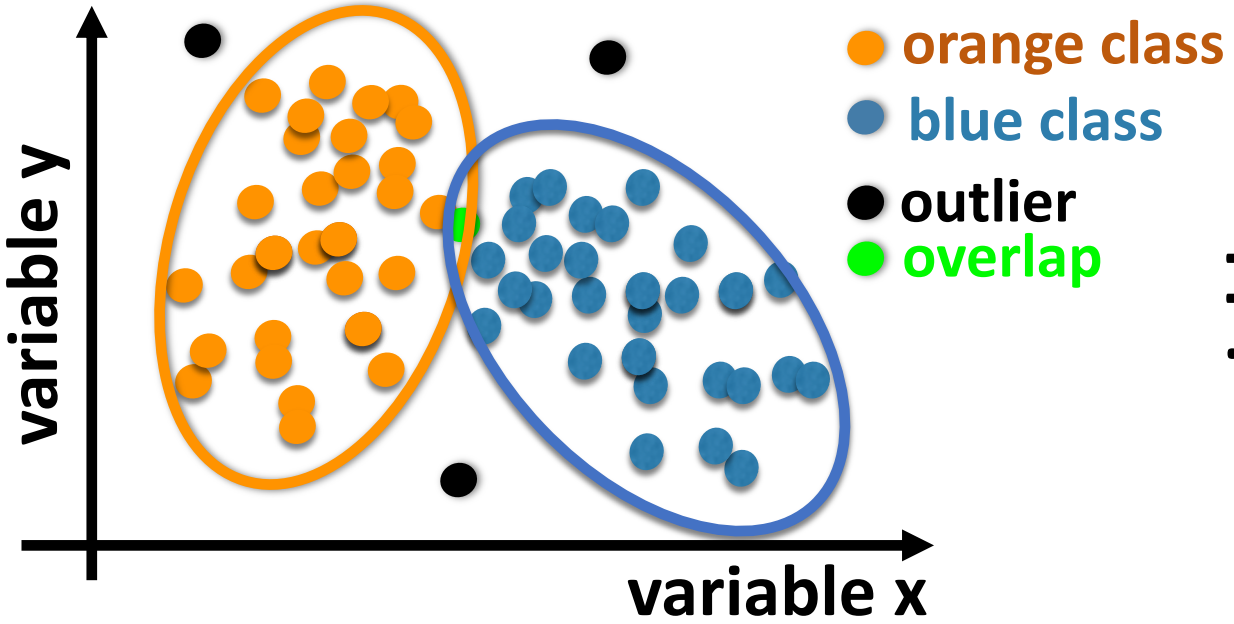
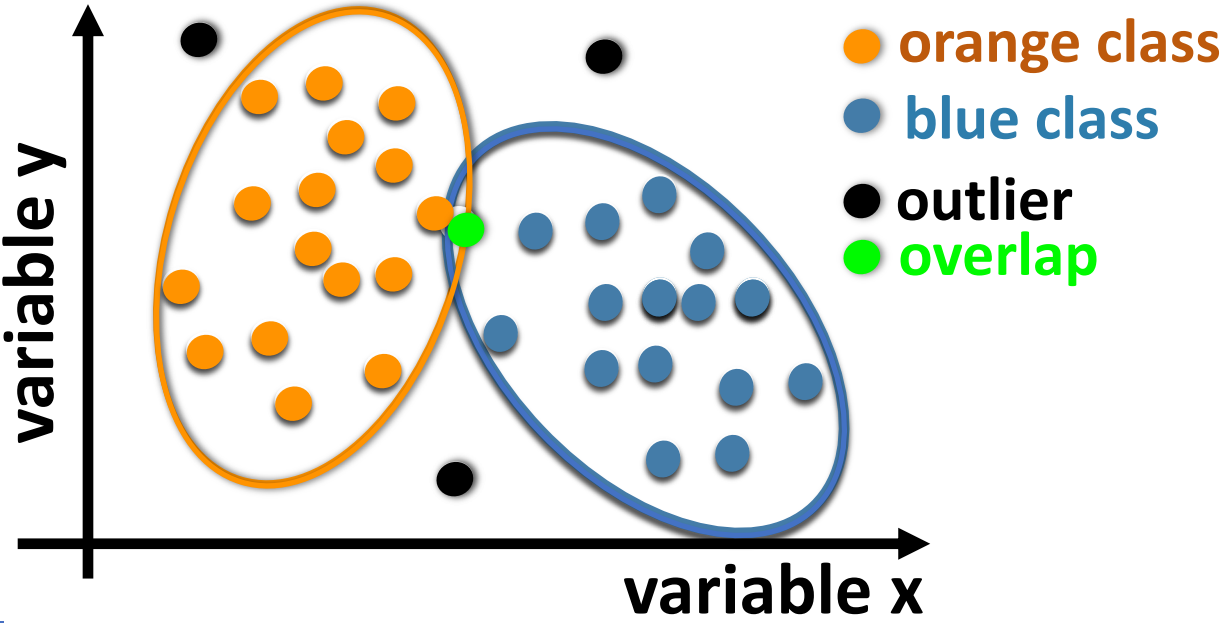
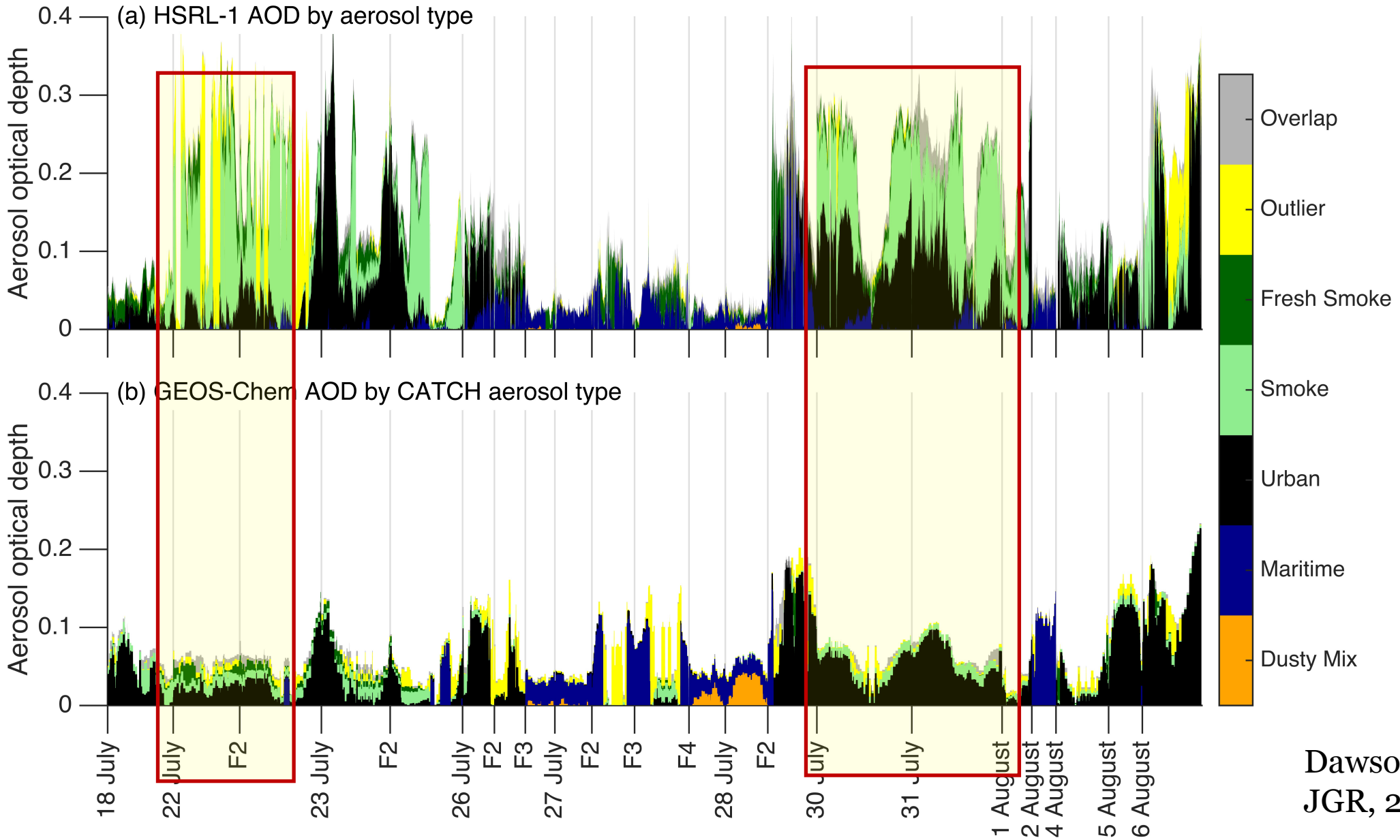


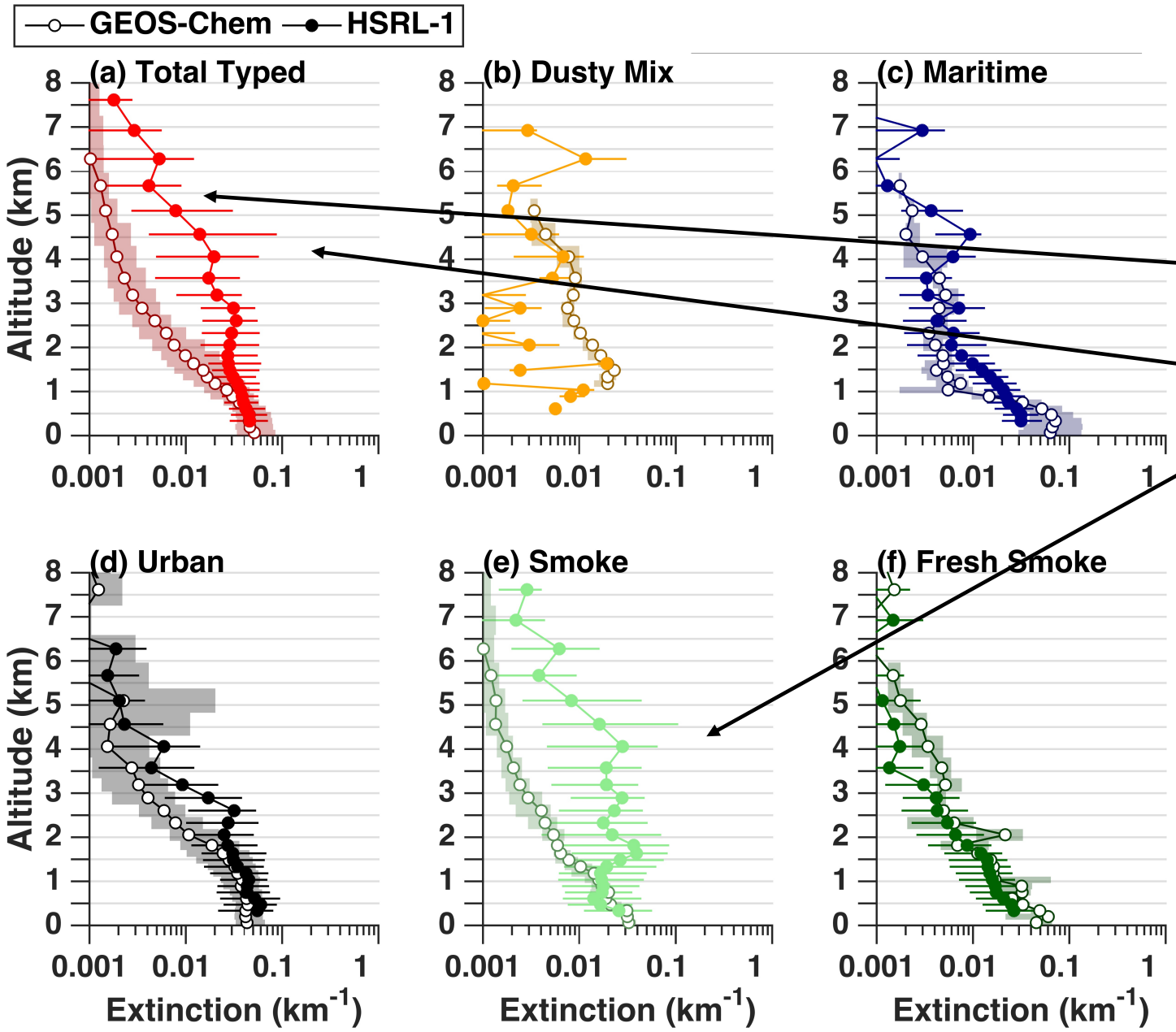


CATCH apportioned aerosol optical depth



Dawson et al., JGR, 2017

Type-specific aerosol extinction

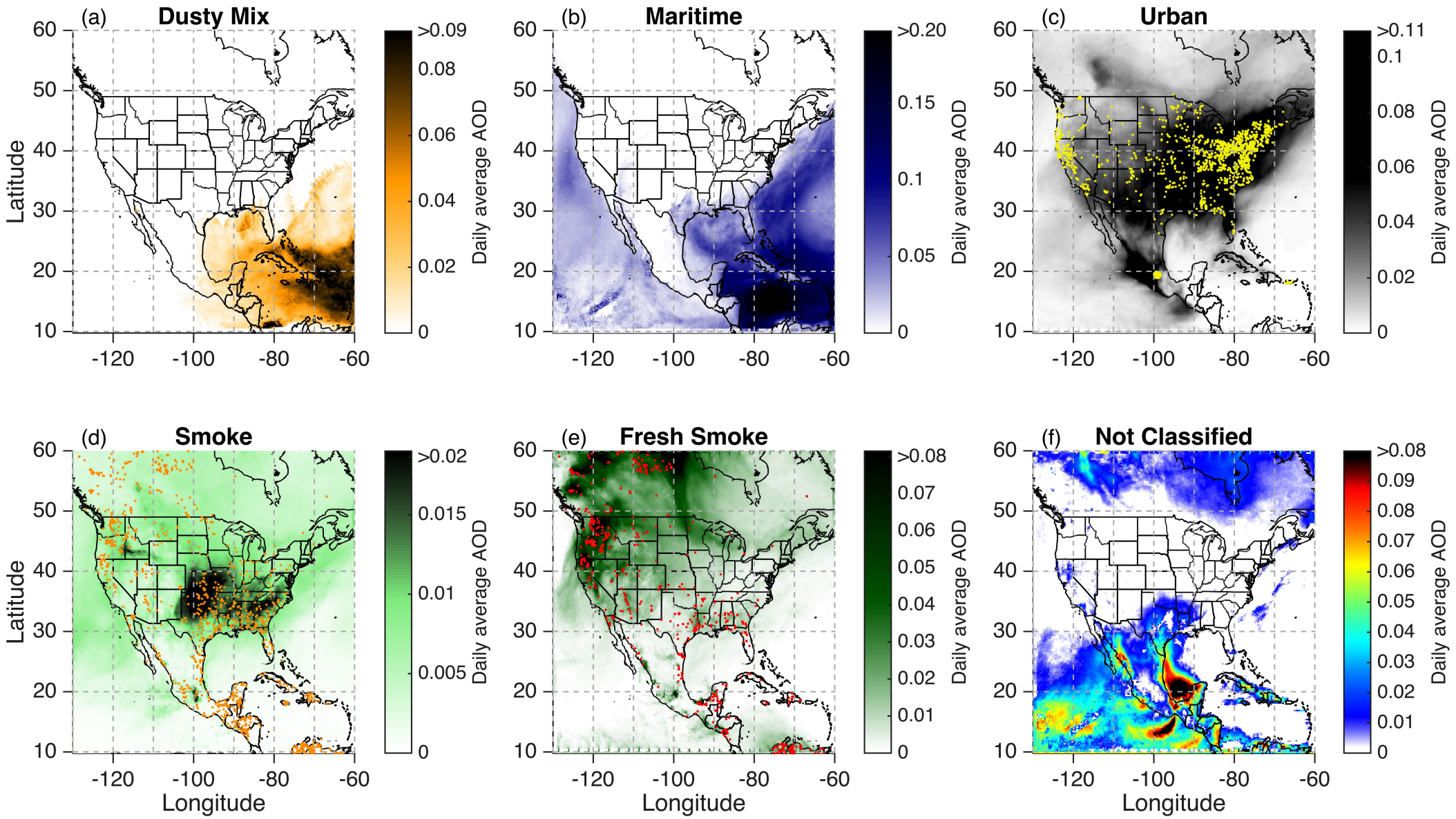


Considerable differences in the extinction

The differences in the extinction are largely due to smoke advecting above the boundary layer

Dawson et al., JGR, 2017

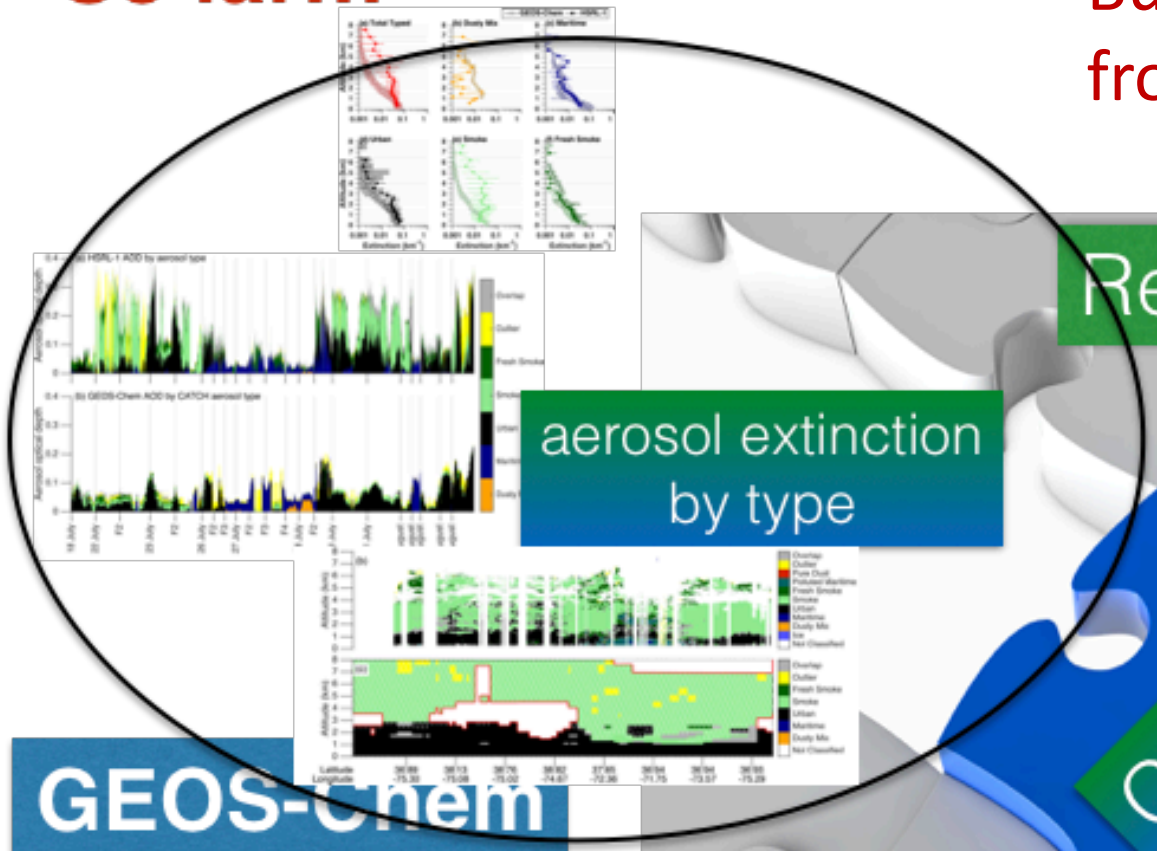
CATCH-derived aerosol types over the continental US



Dawson et al., JGR, 2017

So far…

But we can also get chemical composition from the model for each aerosol type



aerosol extinction by type

GEOS-Chem CTM

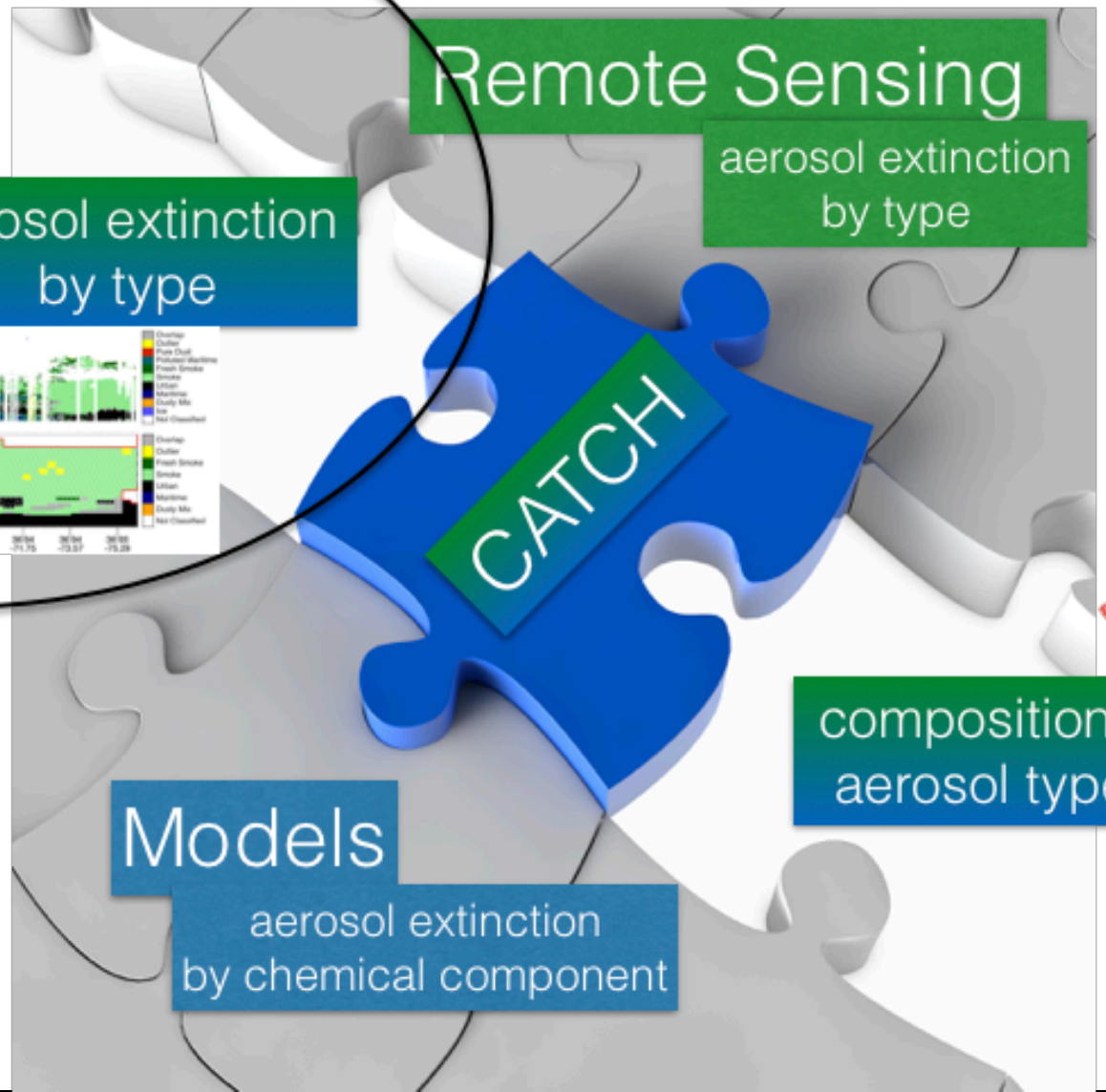
BC

OC

SO4

mineral dust

seasalt



composition of aerosol types

smoke

fresh smoke

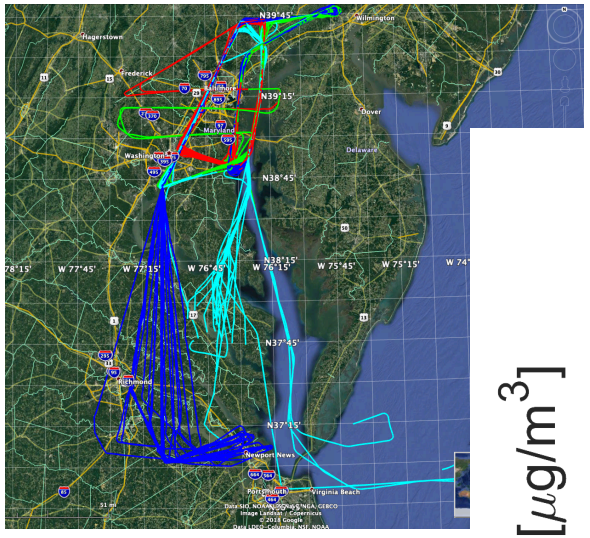
urban

dusty mix

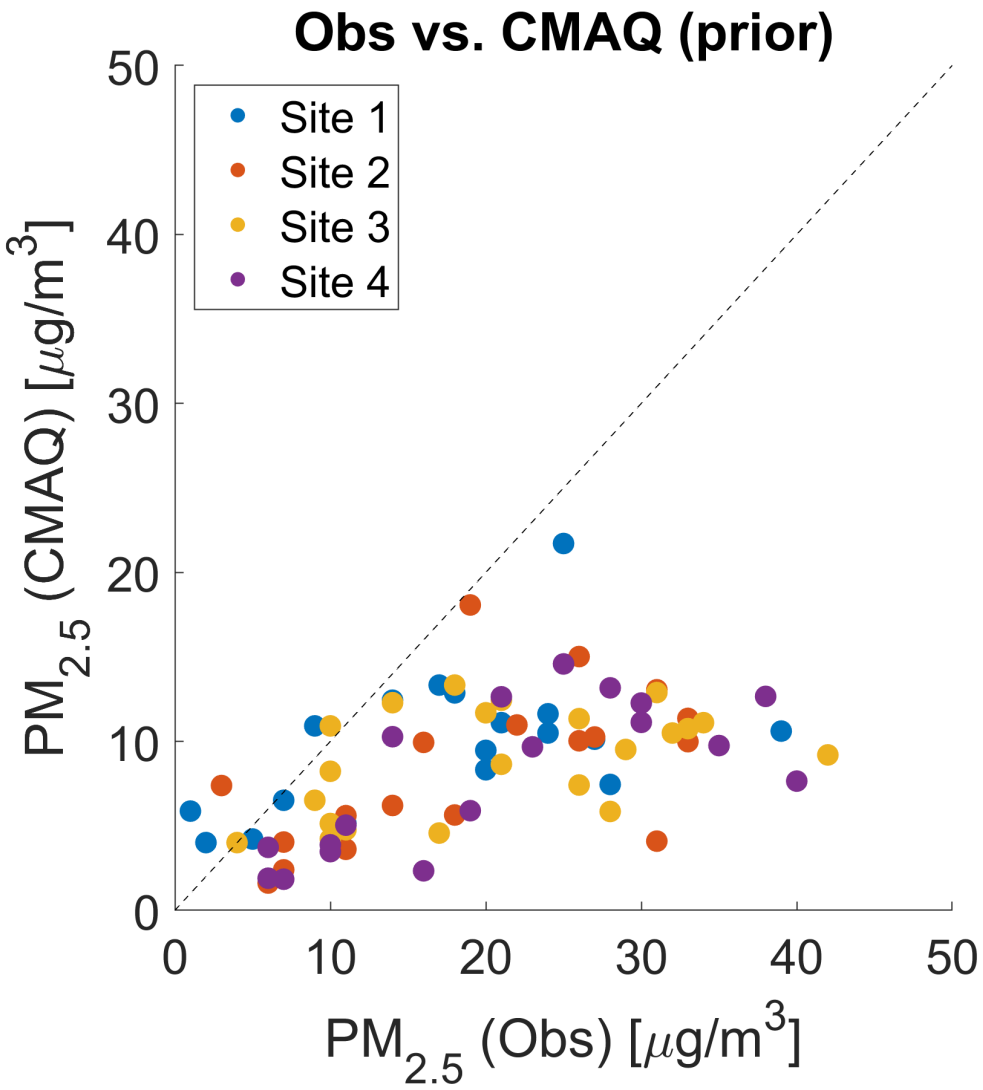
maritime

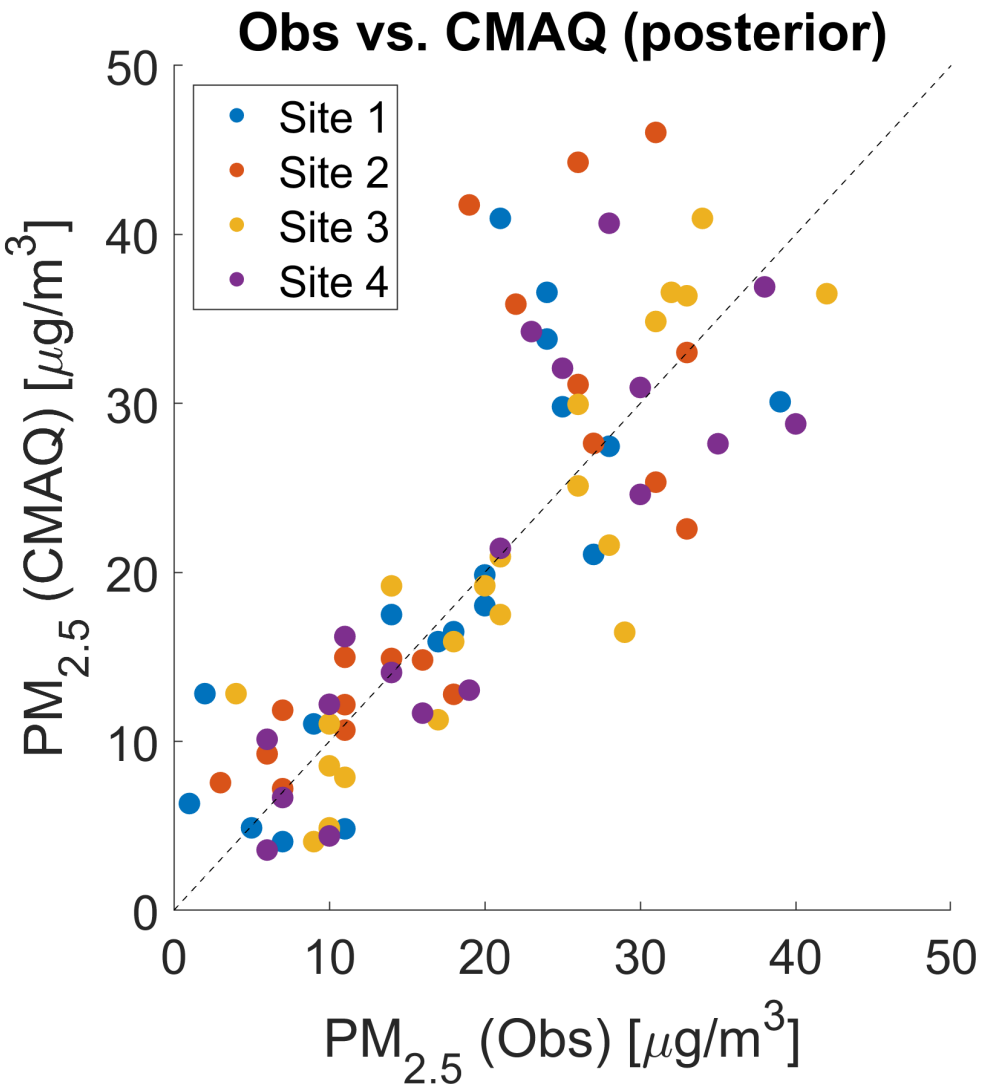
HSRL-1 LiDAR

Discover AQ Baltimore-Washington D.C. 2011



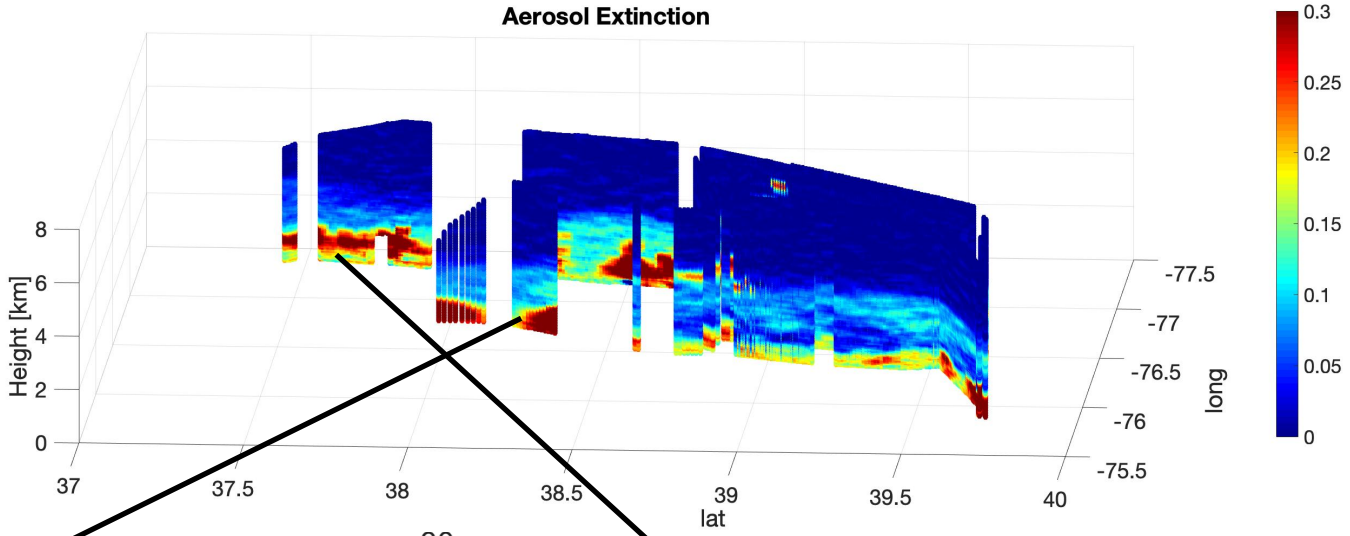
1 hr $PM_{2.5}$ data from the AQS network

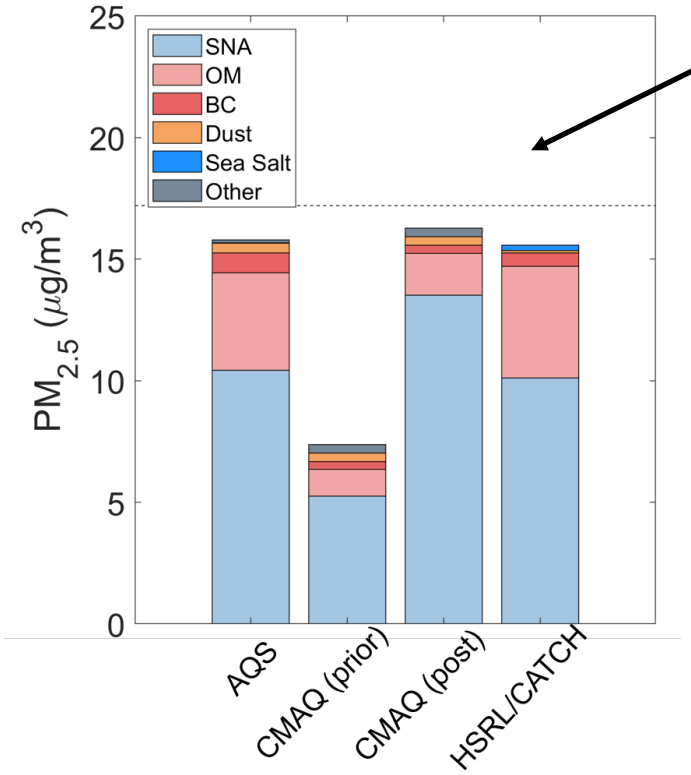


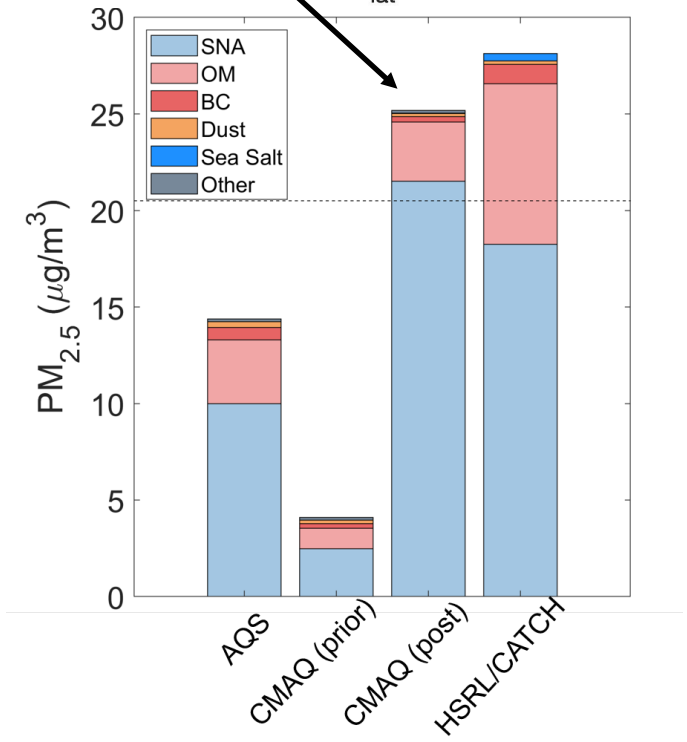


Comparison of PM2.5 composition

Flight 2 on July 20, 2011







In summary

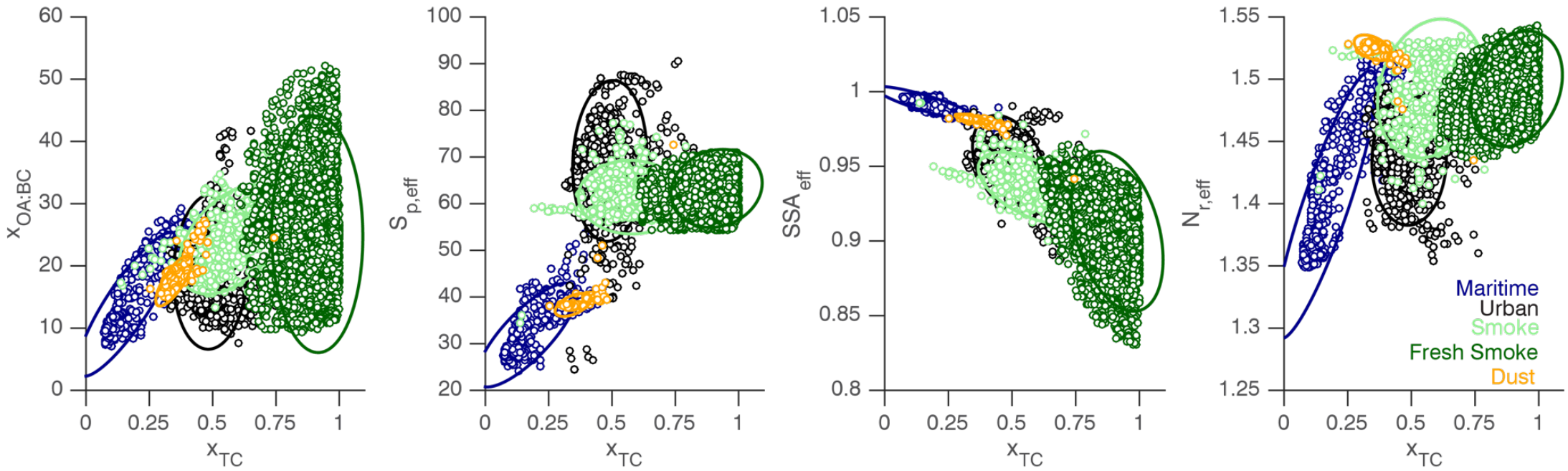
- ✓ The CATCH algorithm can link remotely sensed aerosol types and model-predicted chemical composition
- ✓ CATCH can be used to diagnose model-remote sensing discrepancies in AOD on an aerosol type level
- ✓ The CATCH algorithm (written for GEOS-Chem) can be adapted to any regional/global model

➢ CATCH-derived type-specific information combined with HSRL retrievals of aerosol type and extinction profile can be used to

- *improve* the air quality model-derived $PM_{2.5}$ concentrations
- *predict* the ground level $PM_{2.5}$ and its chemical proxies

Additional Slides

GEOS-Chem derived parameters are then clustered according to the HSRL-retrieved aerosol types to be used by CATCH



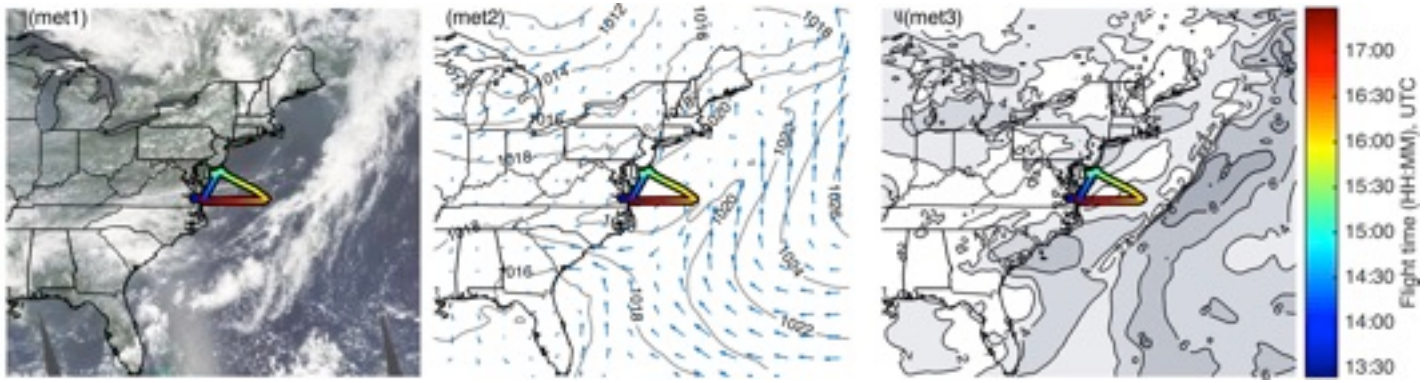
5-D class-to-class Mahalanobis distance indicates ease of classification:

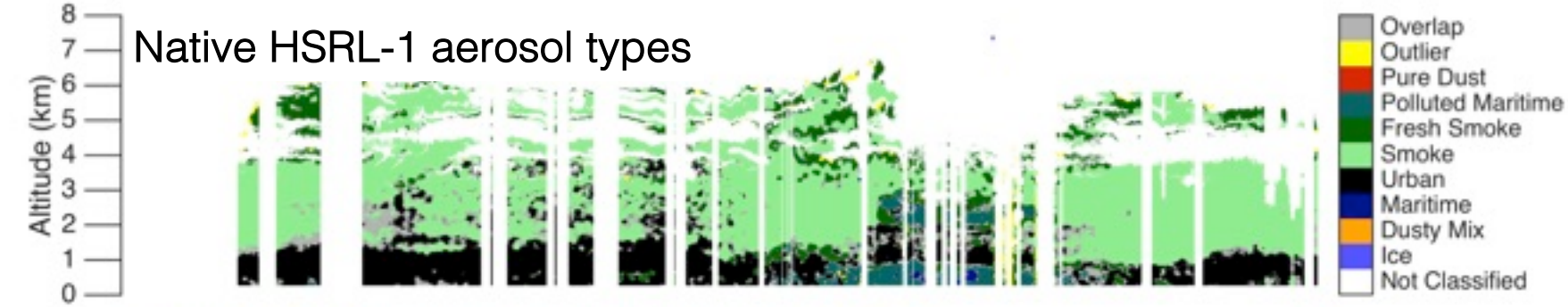
Classes	Dusty Mix	Maritime	Urban	Smoke	Fresh Smoke
Dusty Mix	0	41.2	24.0	40.5	64.3
Maritime	41.2	0	17.7	29.1	38.2
Urban	24.0	17.7	0	3.1	6.6
Smoke	40.5	29.1	3.1	0	5.1
Fresh Smoke	64.3	38.2	6.6	5.1	0

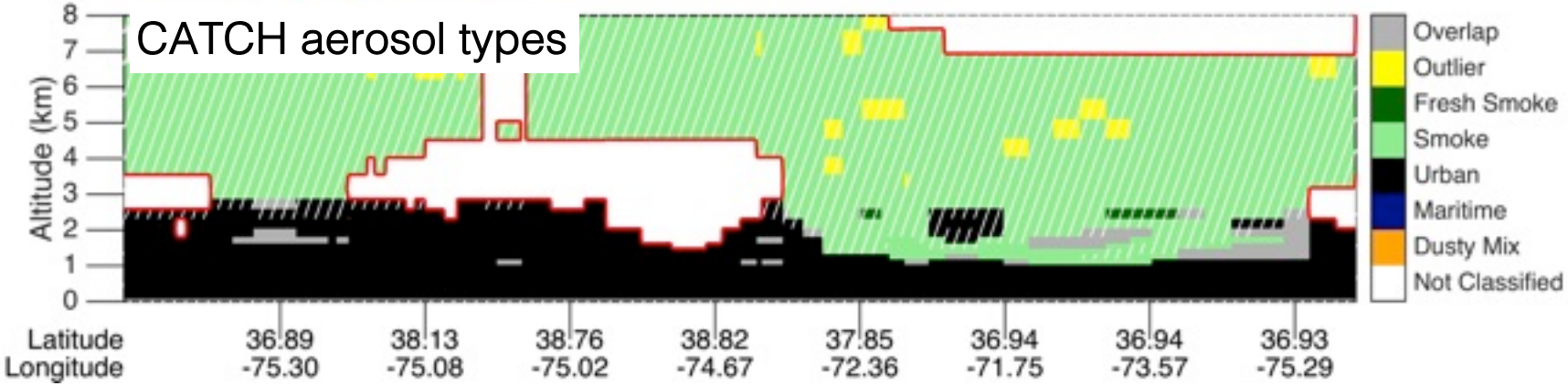
Dawson et al., JGR, 2017

Comparison of HSRL-1 and CATCH-derived aerosol types for SABOR campaign

31 July 2014



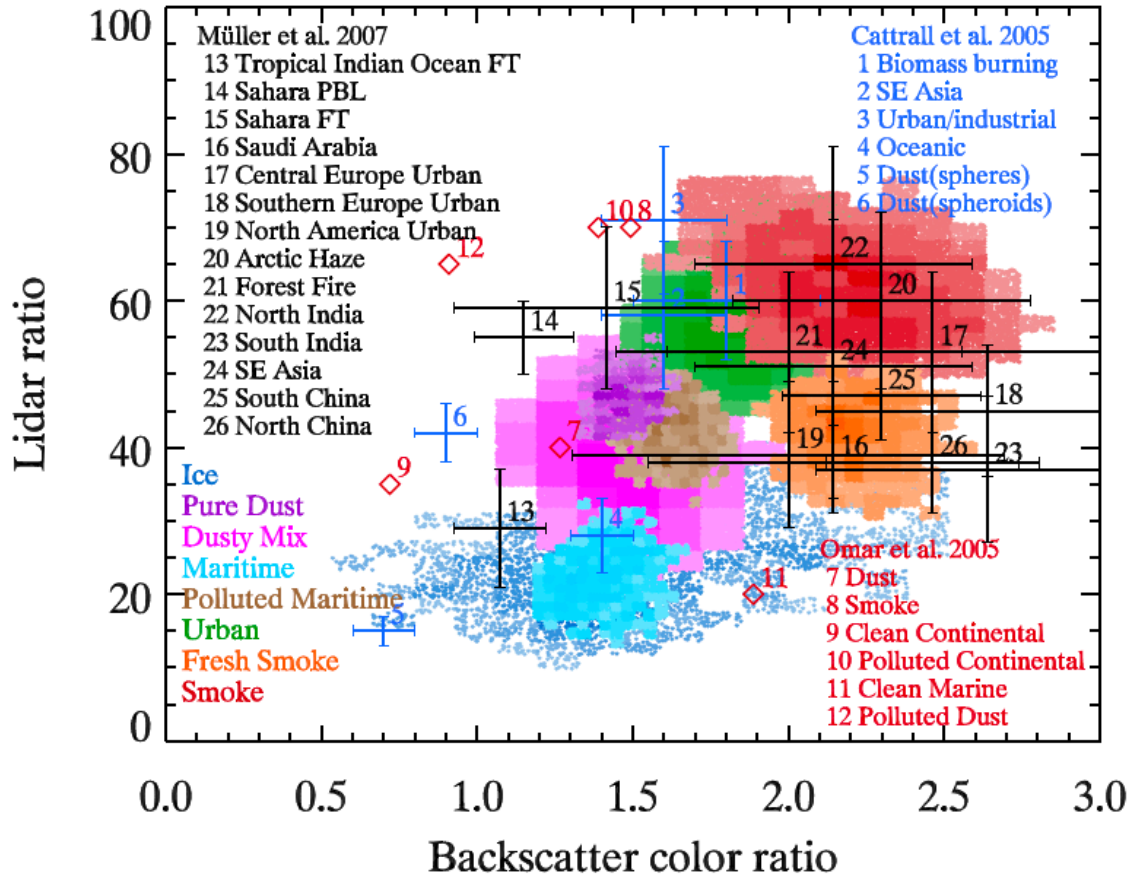


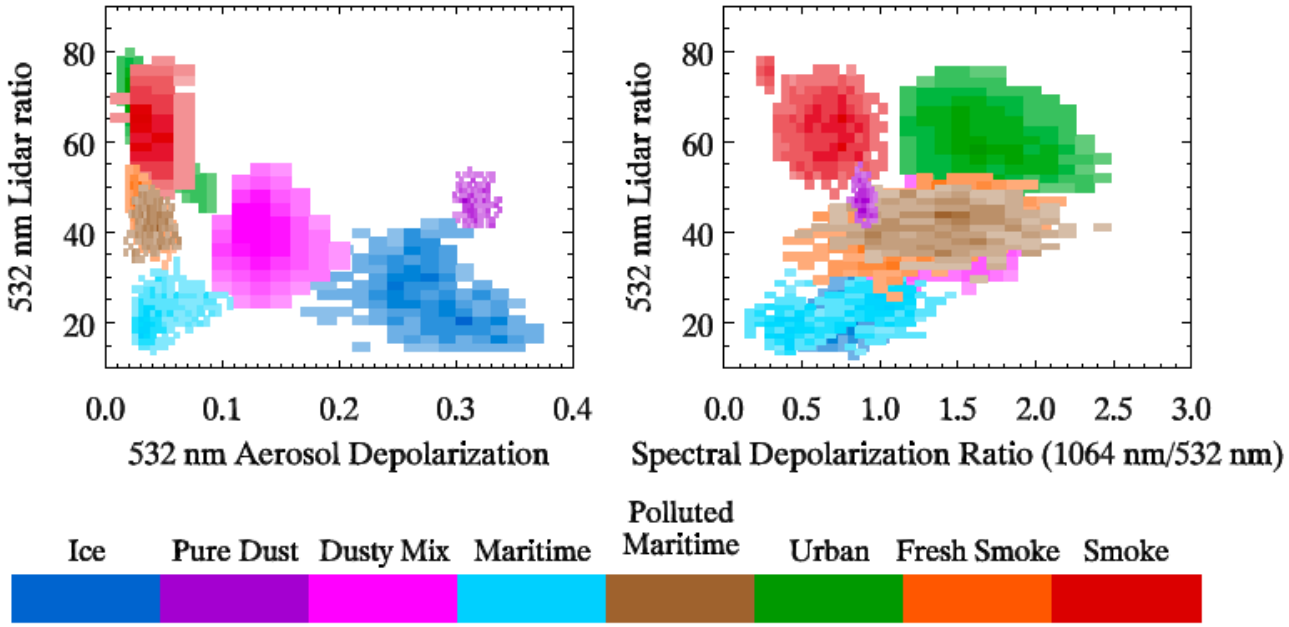


Dawson et al., JGR, 2017

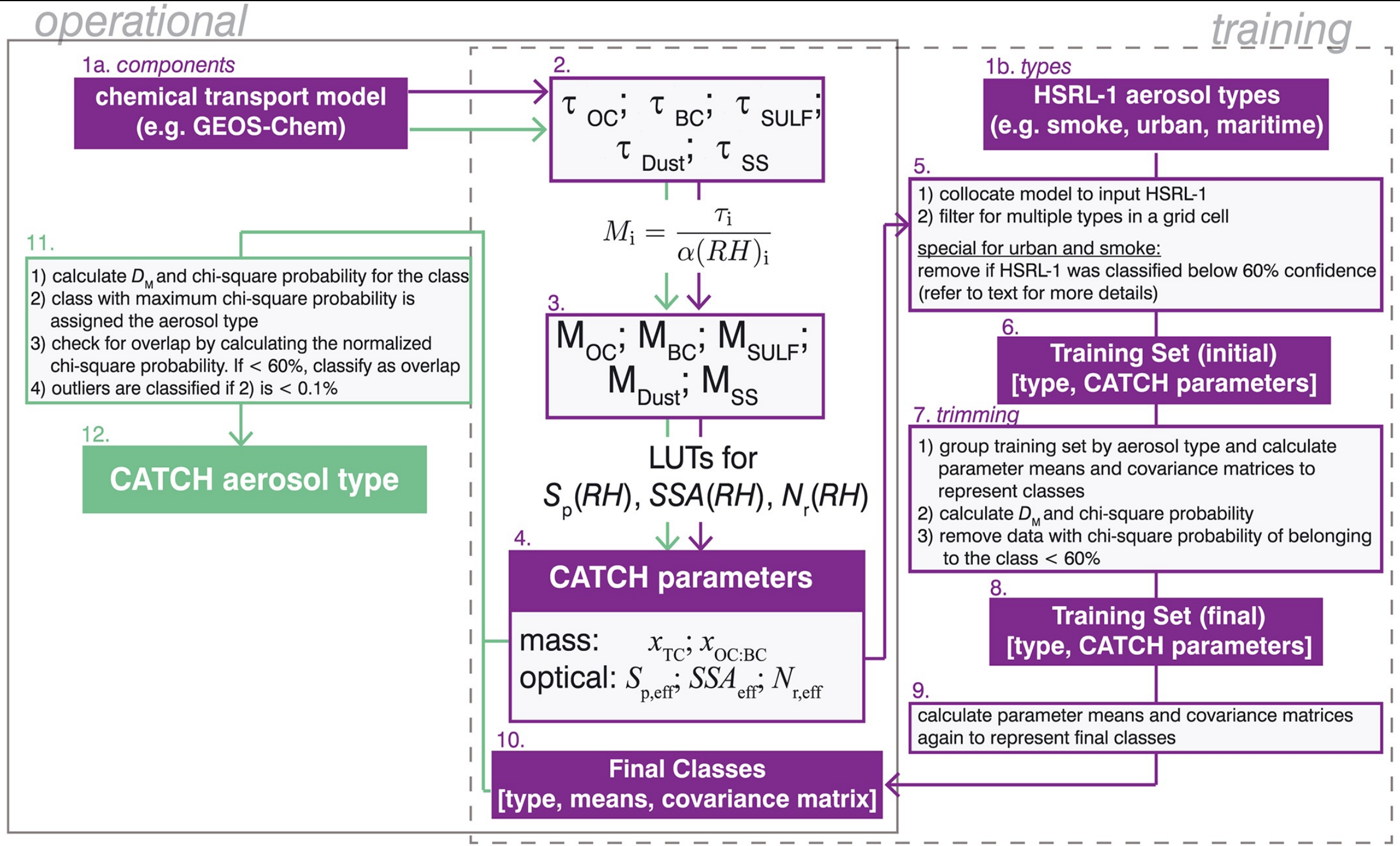
Aerosol Classification Using Airborne High Spectral Resolution Lidar (HSRL) Measurements

- Particles of different shape, chemical composition, etc. interact with light in different ways and that allow for unique identification of groups representing types of particles





[*Burton et al.*, 2012]


operational
training
1a. components
chemical transport model (e.g. GEOS-Chem)
2.
τ_{OC}; τ_{BC}; τ_{SULF}; τ_{Dust}; τ_{SS}
$M_i = \frac{\tau_i}{\alpha(RH)_i}$
3.
M_{OC}; M_{BC}; M_{SULF}; M_{Dust}; M_{SS}
LUTs for
$S_p(RH)$, $SSA(RH)$, $N_r(RH)$
4.
CATCH parameters
mass: x_{TC}; $x_{OC:BC}$
optical: $S_{p,eff}$; SSA_{eff}; $N_{r,eff}$
10.
Final Classes
[type, means, covariance matrix]
11.
1) calculate D_M and chi-square probability for the class
2) class with maximum chi-square probability is assigned the aerosol type
3) check for overlap by calculating the normalized chi-square probability. If < 60%, classify as overlap
4) outliers are classified if 2) is < 0.1%
12.
CATCH aerosol type
1b. types
HSRL-1 aerosol types (e.g. smoke, urban, maritime)
5.
1) collocate model to input HSRL-1
2) filter for multiple types in a grid cell
special for urban and smoke:
remove if HSRL-1 was classified below 60% confidence (refer to text for more details)
6.
Training Set (initial)
[type, CATCH parameters]
7. trimming
1) group training set by aerosol type and calculate parameter means and covariance matrices to represent classes
2) calculate D_M and chi-square probability
3) remove data with chi-square probability of belonging to the class < 60%
8.
Training Set (final)
[type, CATCH parameters]
9.
calculate parameter means and covariance matrices again to represent final classes

Mass Fractions Used For Uncertainty Calculations In Aerosol Microphysics

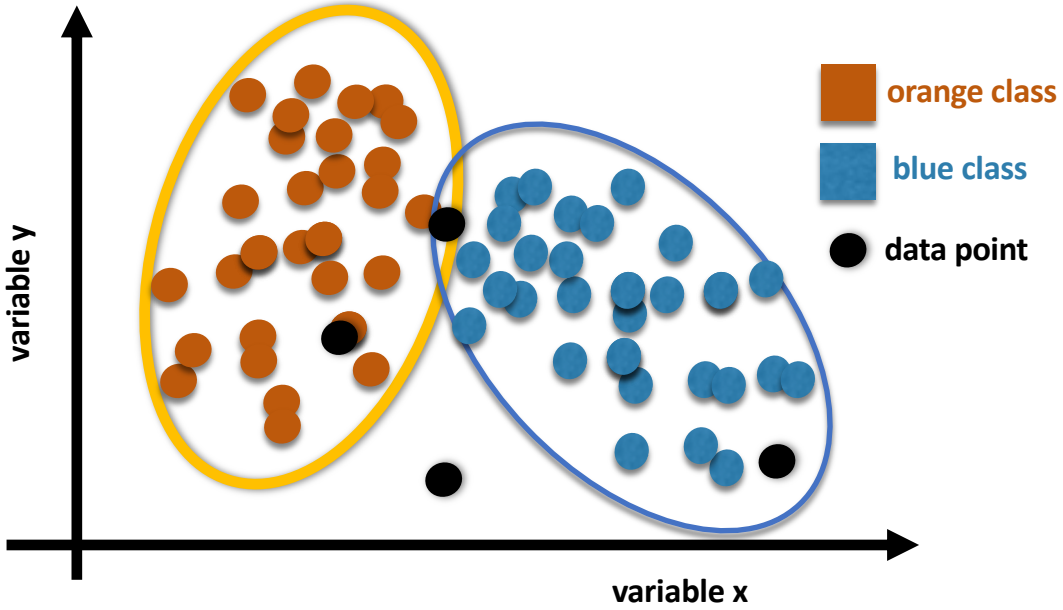
	DustyMix	Marine	Urban	Smoke	FreshSmoke
OM	0.762	0.226	27.693	32.988	48.6
BC	0.104	0.058	3.346	2.994	6.91
SO_4	3.677	5.93	60.849	59.795	40.434
Dust	94.756	4.578	0.603	2.612	1.031
SeaSalt	0.686	88.894	1.249	0.534	0.362

Remote sensing methods use statistical grouping of optical properties to create unique "clusters"

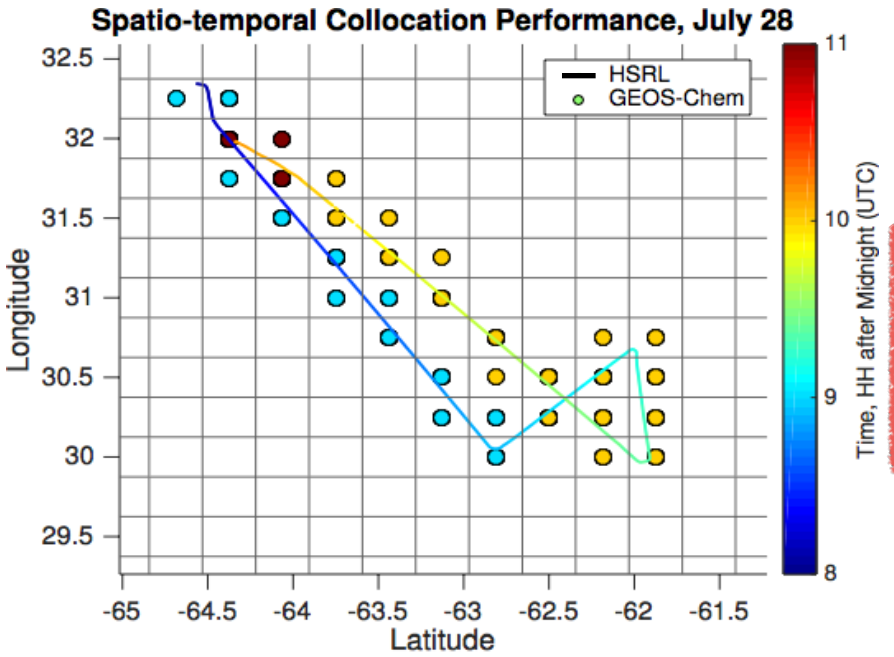
- First, variables are selected based on clustering potential and assuming approximate normal distributions

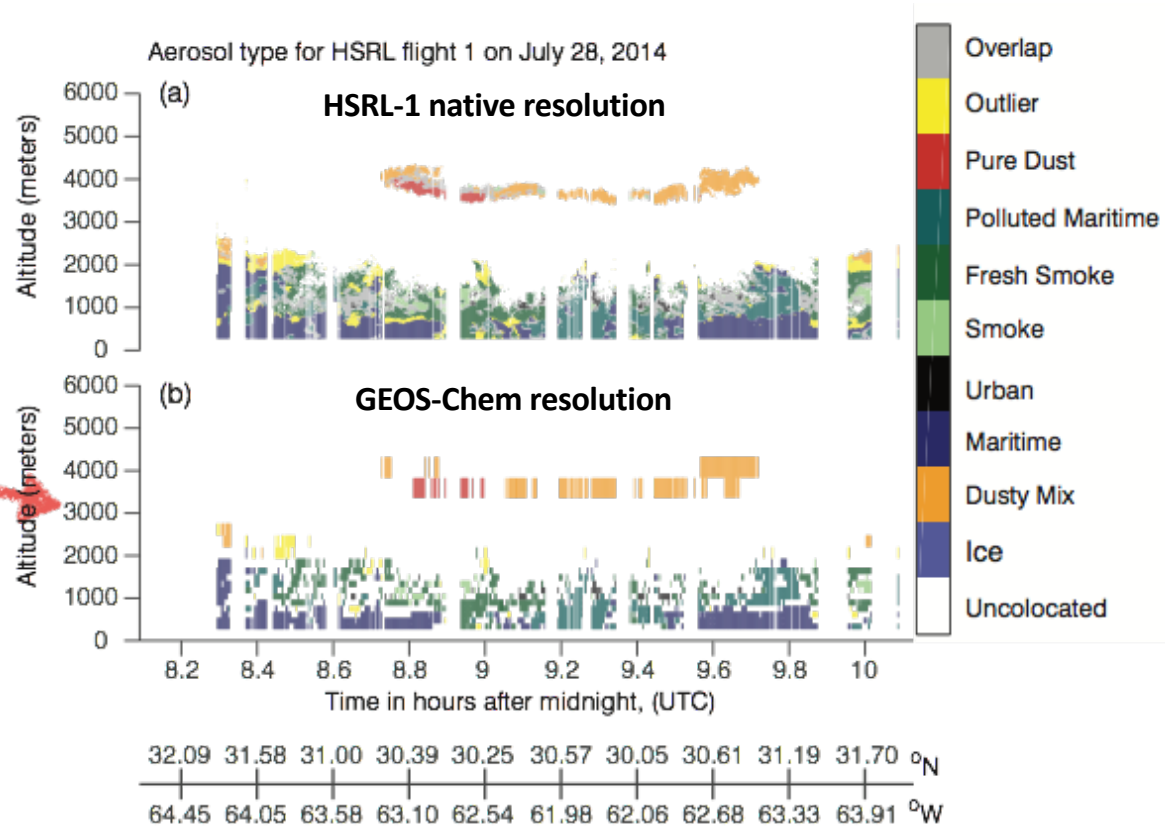
$$D_M^2 = (\mathbf{x} - \mu_\mathbf{x})^T \mathbf{S}^{-1} (\mathbf{x} - \mu_\mathbf{x})$$

- Mahalanobis distance (D_M) is a multivariate metric that combines variable means (μ_x) and covariance (**S**) to assign a probability of belonging to a particular cluster



HSRL-1 aerosol types are collocated and gridded to GEOS-Chem resolution





Retained data has:

1) consistent aerosol types for the model vertical grid
2) >60% probability of belonging to the HSRL-assigned type
3) >30% of the model vertical grid classified as an aerosol type

Now group the collocated model parameters by filtered HSRL—retrieved aerosol types