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U.S. DEPARTMENT OF ENERGY

Improving single particle quantification through hardware and data analytics

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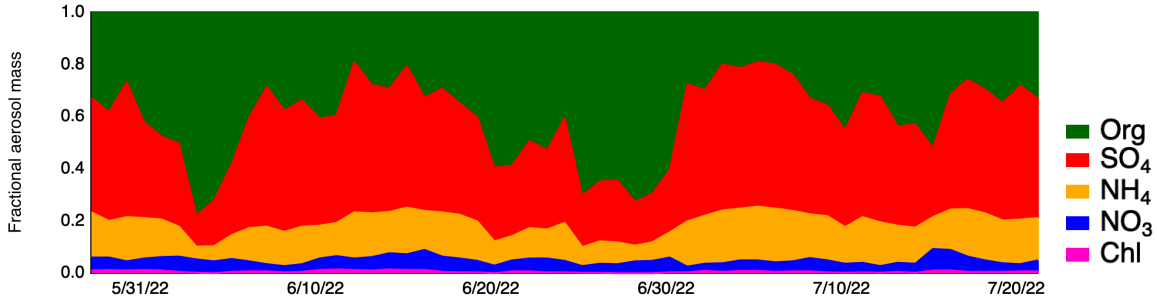
Aerosol chemistry measurements are an essential for accurate source apportionment and aerosol processes studies

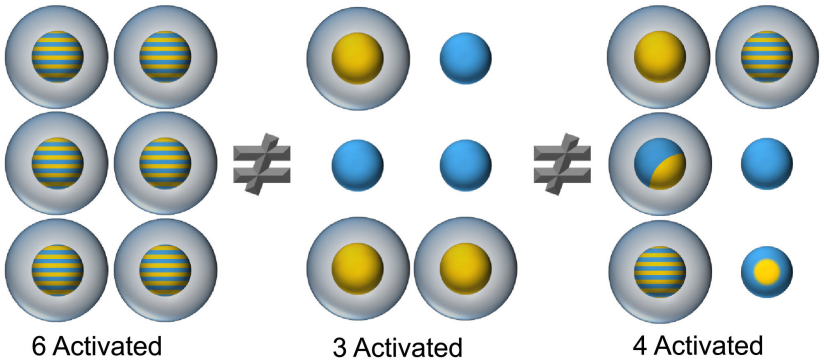


Aerosol composition is a frequently requested ARM capability.

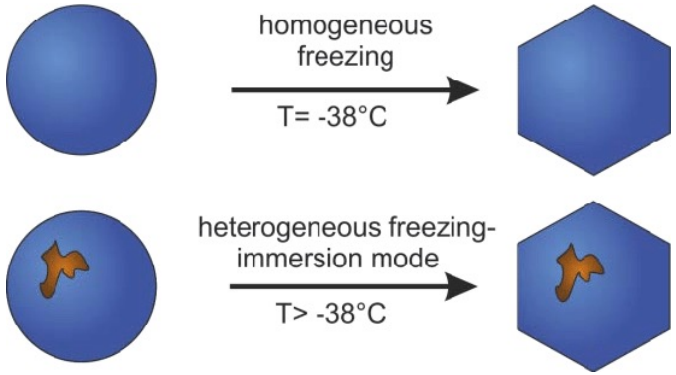
The currently used technique, the ACSM only provides bulk non-refractory submicron aerosol composition

Single-particle resolving models improve model accuracy for aerosol-cloud interactions

CCN activation



Heterogeneous ice nucleation



Many atmospheric processes, especially within the realm of aerosol-cloud interactions, depend strongly on properties of individual aerosol particles

Riemer, et al. (2019)

"Bulk parametrizations" can result in model inaccuracies

For example, Zaveri, et al. (2010) found that averaging the CCN composition resulted in overestimates of 40% in CCN efficiency compared to a particle-resolving model.

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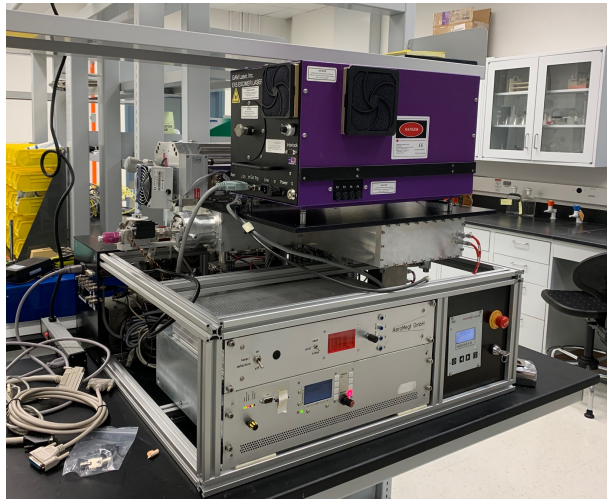
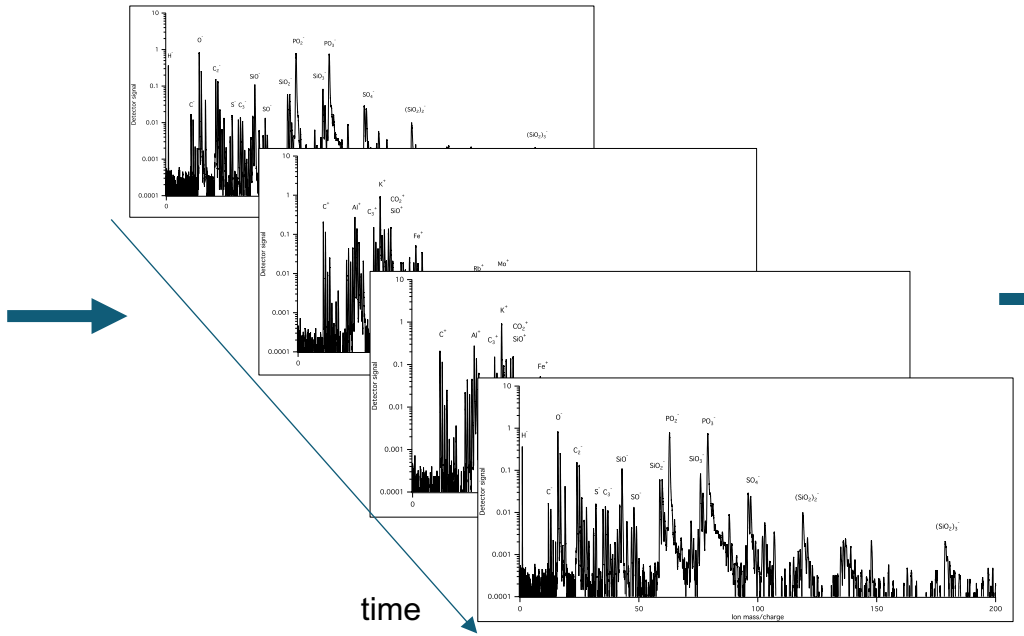
New project: single-particle mass spectrometry at BNL

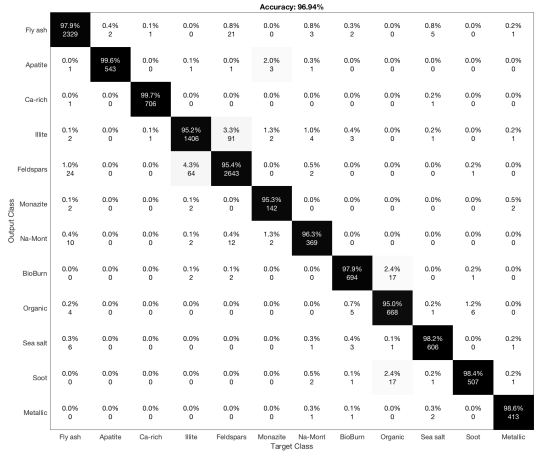
- Aims for the most comprehensive characterization of single particle composition to date through both hardware improvements and computational advances



Single-particle mass spectrometer



Multivariate datasets of aerosol composition



Machine learning algorithms

Building the hardware: laser desorption and ionization one particle at a time

Aerosol *in-situ* sampling

↓

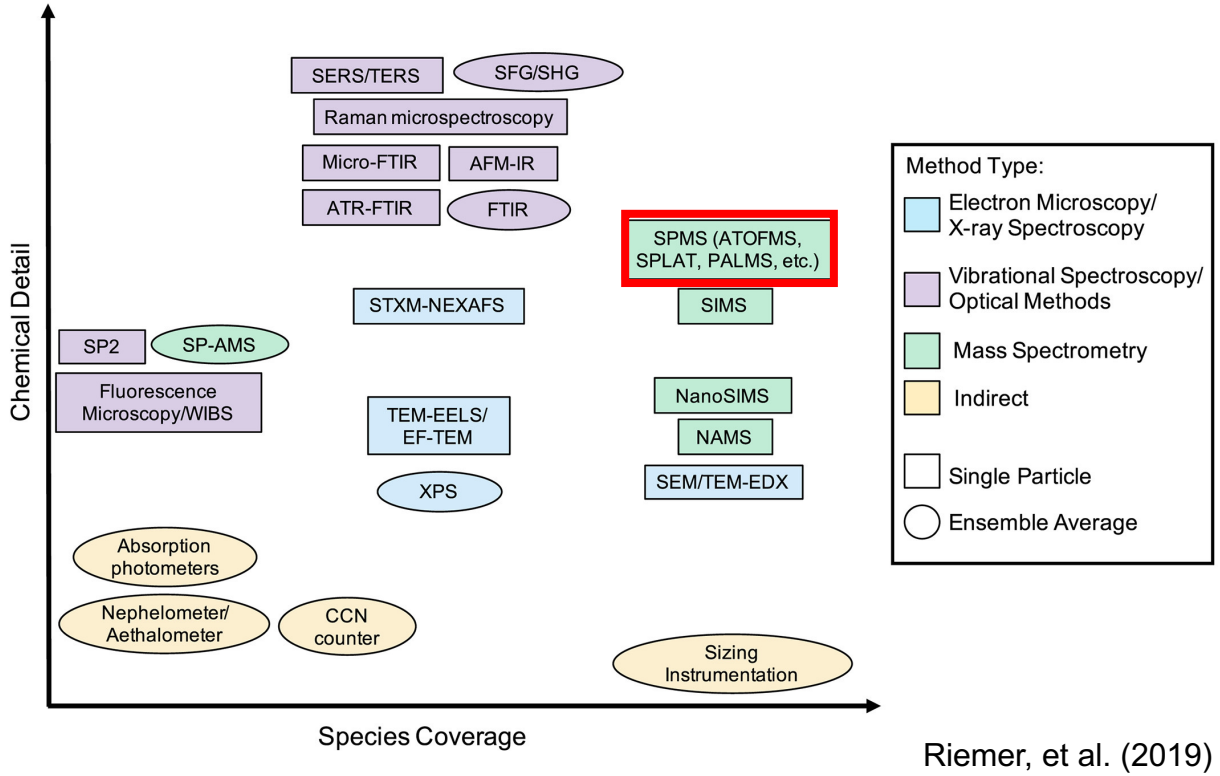
Measurement of vacuum aerodynamic diameter

↓

Single-step laser desorption and ionization

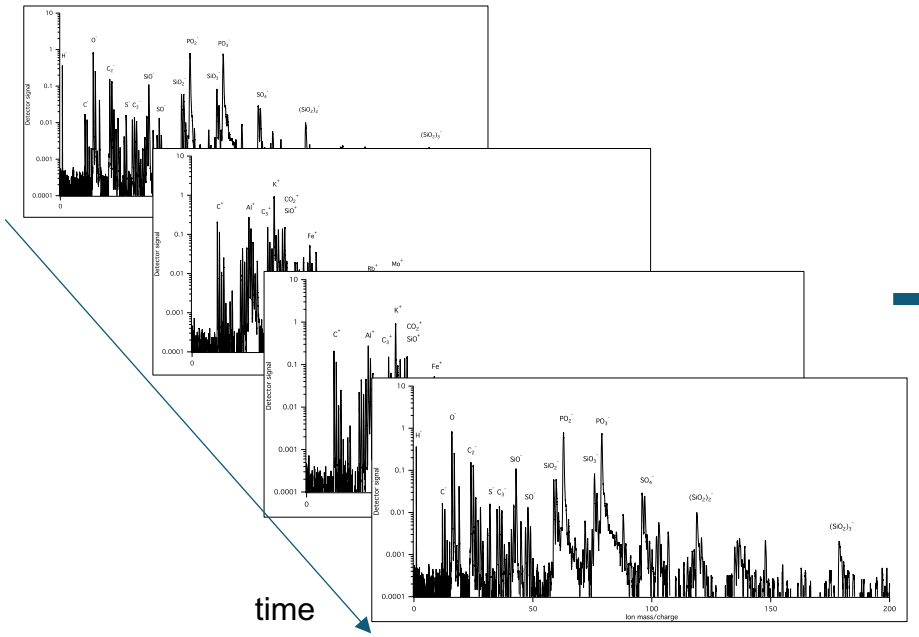
↓

Mass spectrometry



Riemer, et al. (2019)

Using supervised and semi-supervised machine learning classifiers to improve single particle measurements



Accuracy: 96.94%

Output class

	Fly ash	Apatite	Ca-rich	Illite	Feldspars	Monazite	Na-Mont	BioBurn	Organic	Sea salt	Soot	Metallic
Fly ash	97.9% 2329	0.4% 2	0.1% 1	0.0% 0	0.8% 21	0.0% 0	0.8% 3	0.3% 2	0.0% 0	0.8% 5	0.0% 0	0.2% 1
Apatite	0.0% 1	99.6% 543	0.0% 0	0.1% 1	0.0% 1	2.0% 3	0.3% 1	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0
Ca-rich	0.0% 1	0.0% 0	99.7% 706	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.2% 1	0.0% 0	0.0% 0
Illite	0.1% 2	0.0% 0	0.1% 1	95.2% 1406	3.3% 91	1.3% 2	1.0% 4	0.4% 3	0.0% 0	0.2% 1	0.0% 0	0.2% 1
Feldspars	1.0% 24	0.0% 0	0.0% 0	4.3% 64	95.4% 2643	0.0% 0	0.5% 2	0.0% 0	0.0% 0	0.0% 0	0.2% 1	0.0% 0
Monazite	0.1% 2	0.0% 0	0.0% 0	0.1% 2	0.0% 0	95.3% 142	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.5% 2
Na-Mont	0.4% 10	0.0% 0	0.0% 0	0.1% 2	0.4% 12	1.3% 2	96.3% 369	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0
BioBurn	0.0% 0	0.0% 0	0.0% 0	0.1% 2	0.1% 2	0.0% 0	0.0% 0	97.9% 694	2.4% 17	0.0% 0	0.2% 1	0.0% 0
Organic	0.2% 4	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.7% 5	95.0% 668	0.2% 1	1.2% 6	0.0% 0
Sea salt	0.3% 6	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.3% 1	0.4% 3	0.1% 1	98.2% 606	0.0% 0	0.2% 1
Soot	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.5% 2	0.1% 1	2.4% 17	0.2% 1	98.4% 507	0.2% 1
Metallic	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.0% 0	0.3% 1	0.1% 1	0.0% 0	0.3% 2	0.0% 0	98.6% 413

Target class

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