A Comprehensive Analysis of New Particle Formation Across the Northwest Atlantic: Analysis of ACTIVATE Airborne Data

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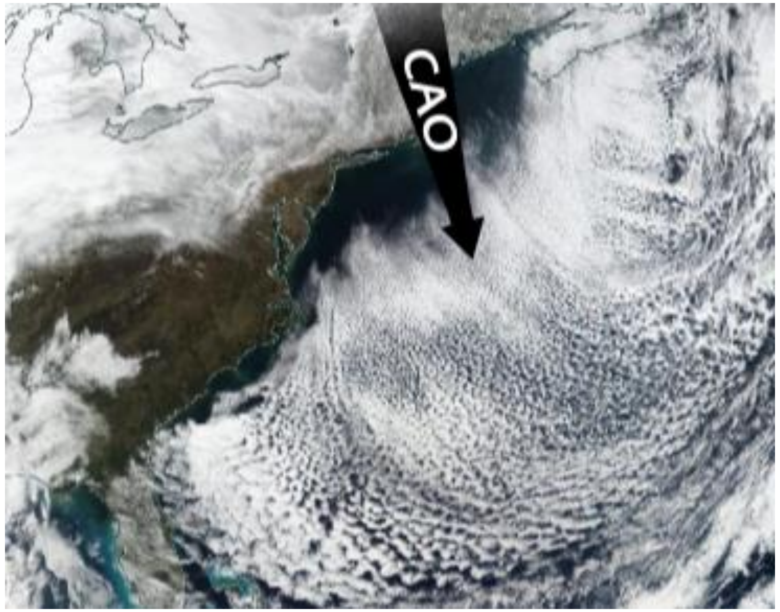
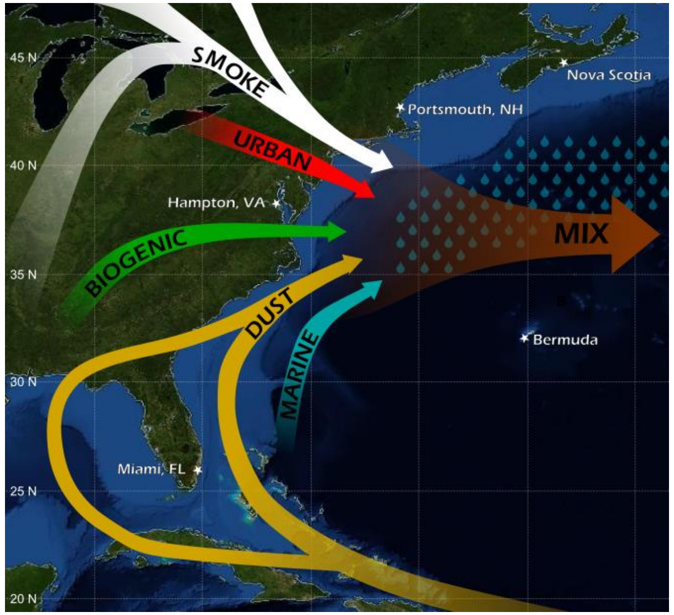
Key Question and Motivation

- Motivation: New particle formation (NPF) is crucial for understanding aerosol number and size distributions, and cloud formation owing to influence on the CCN budget.
- Driving question: What are the spatial, vertical, and seasonal characteristics of NPF over the northwest Atlantic?





- What promotes NPF?
- High precursor vapor levels (e.g., ammonia, sulfuric acid, organics) and water
- Low aerosol surface area
- Enhanced incident solar radiation

Relevant recent results over Atlantic Ocean (Zheng et al., 2021; Corral et al., 2022):

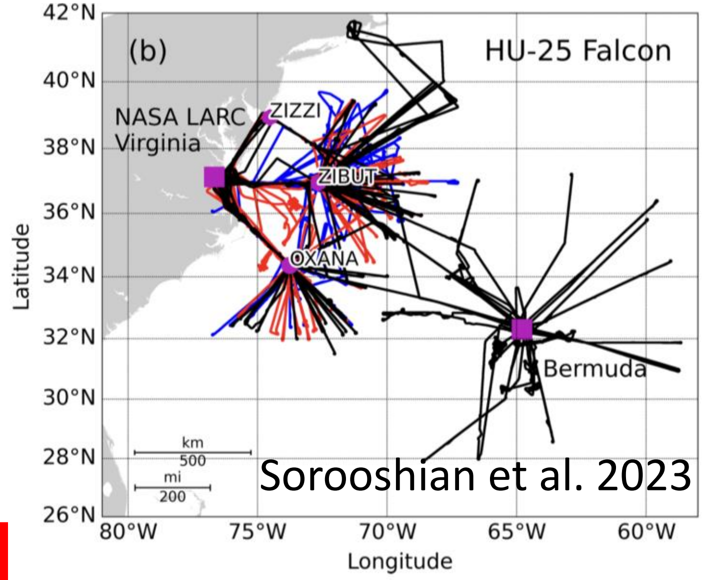
- NPF occurs in the upper marine boundary layer after cold fronts.
- Cold Air Outbreaks promote NPF offshore the U.S. East Coast.
- NPF attributed to cold temperatures, low aerosol surface area, and high actinic fluxes in broken cloud fields.

There is a need for more studies over marine regions

ACTIVATE Strategy

- NASA's Aerosol Cloud meTeorology Interactions oVer the western ATlantic Experiment (ACTIVATE) [2020-2022]
    - Two spatially coordinated aircraft sampling variables relevant to aerosol-cloud interactions
    - Aimed to maximize data volume (179 total research flights)
    - Publicly available dataset: https://doi.org/10.5067/SUBORBITAL/ACTIVATE/DATA001

Blue = 2020; Red = 2021; Black = 2022

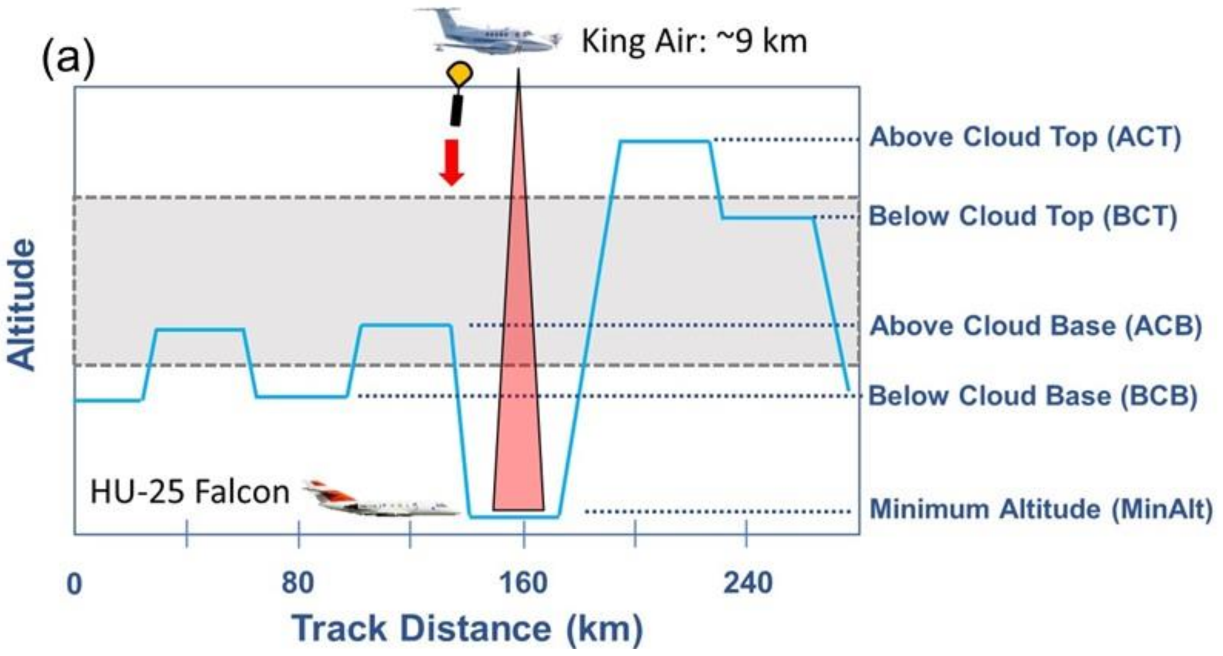


Sorooshian et al. 2023

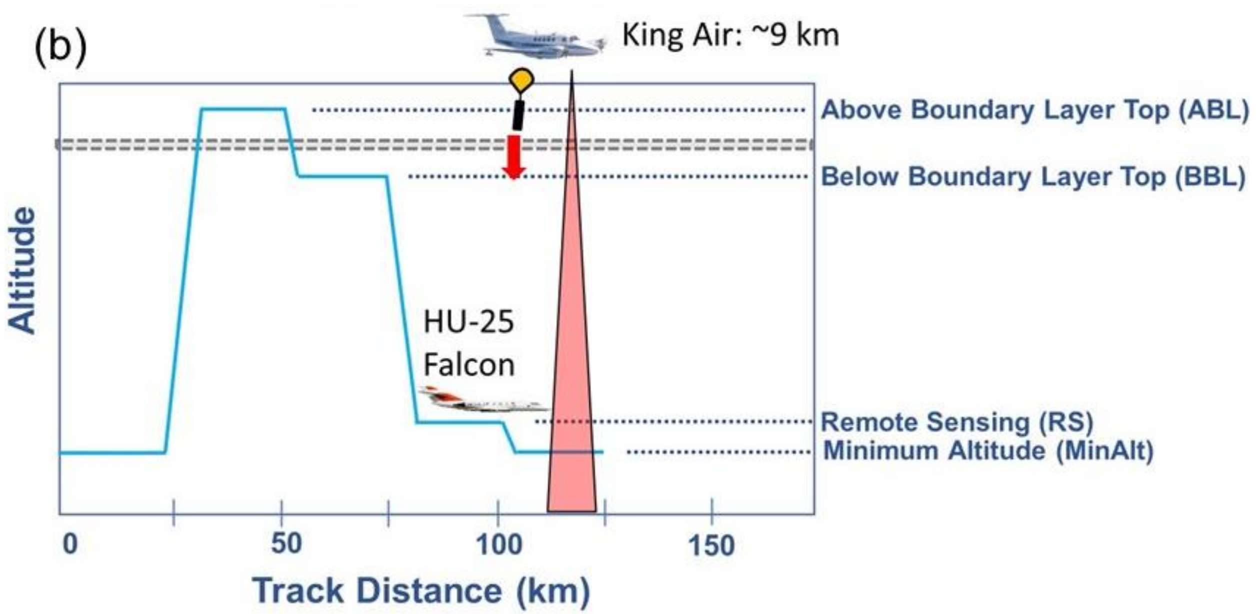
What are ensembles?

	Research flights			Flight hours		Joint ensembles	
	HU-25 Falcon	King Air	Joint	HU-25 Falcon	King Air	Cloudy	Clear
Winter 2020 (14 February–12 March)	22	17	17	73	59	43	28
Summer 2020 (13 August–30 September)	18	18	18	60	67	58	36
Winter 2021 (27 January–2 April)	17	19	15	56	66	47	25
Summer 2021 (13 May–30 June)	32	32	32	106	108	103	74
Winter 2021–2022 (30 November–29 March)	55	54	53	182	193	198	72
Summer 2022 (3 May–18 June)	30	28	27	97	98	86	46

Cloudy Ensemble



Clear Ensemble



- Ensembles: Facilitate easier data analysis by conducting flights in a routine and prescriptive manner where data can be easily combined and aggregated based on factors such as altitude relative to cloud and MBL top
- Both aircraft within ~5 min and ~6 km for 73% of the time (e.g., Schlosser et al., 2024)
- This Study: Focused on in situ data just from the lower flying HU-25 Falcon

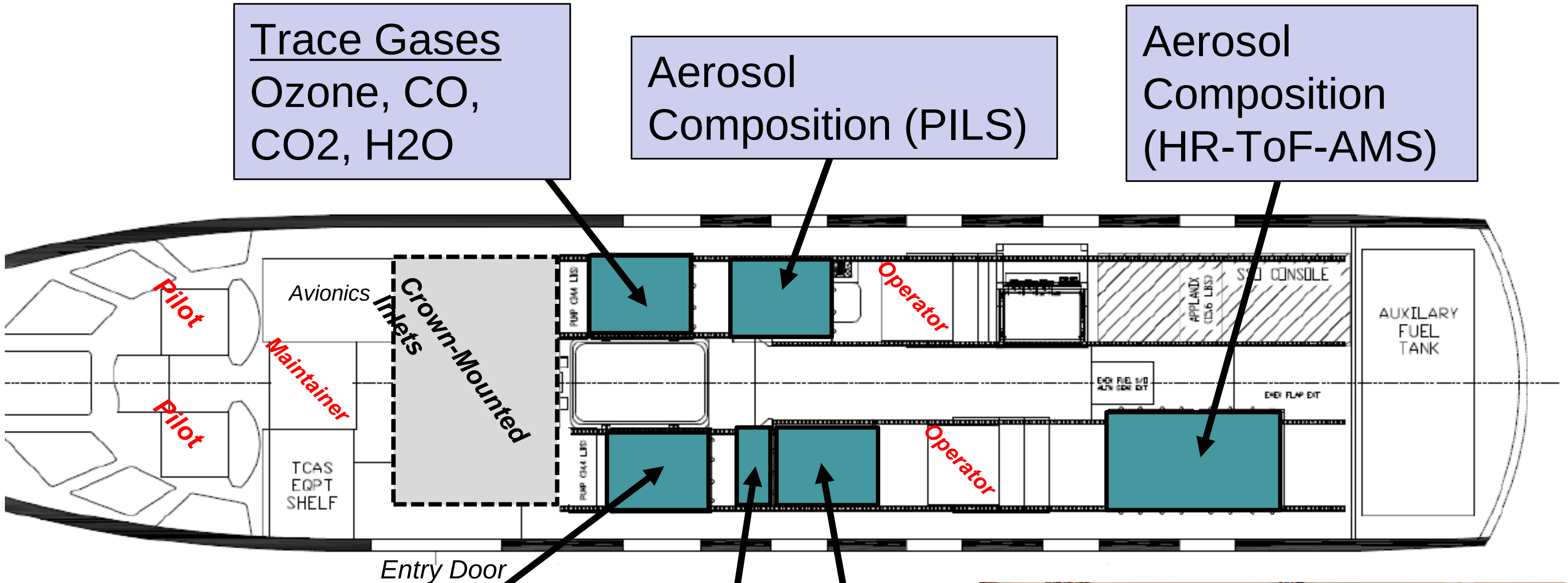
Payload: Falcon In-Situ Measurements

<u>Most relevant instruments used</u>

- Two condensation particle counters (> 3 nm and > 10 nm) that measure total particle concentration (N)

<u>Indicator of NPF</u>

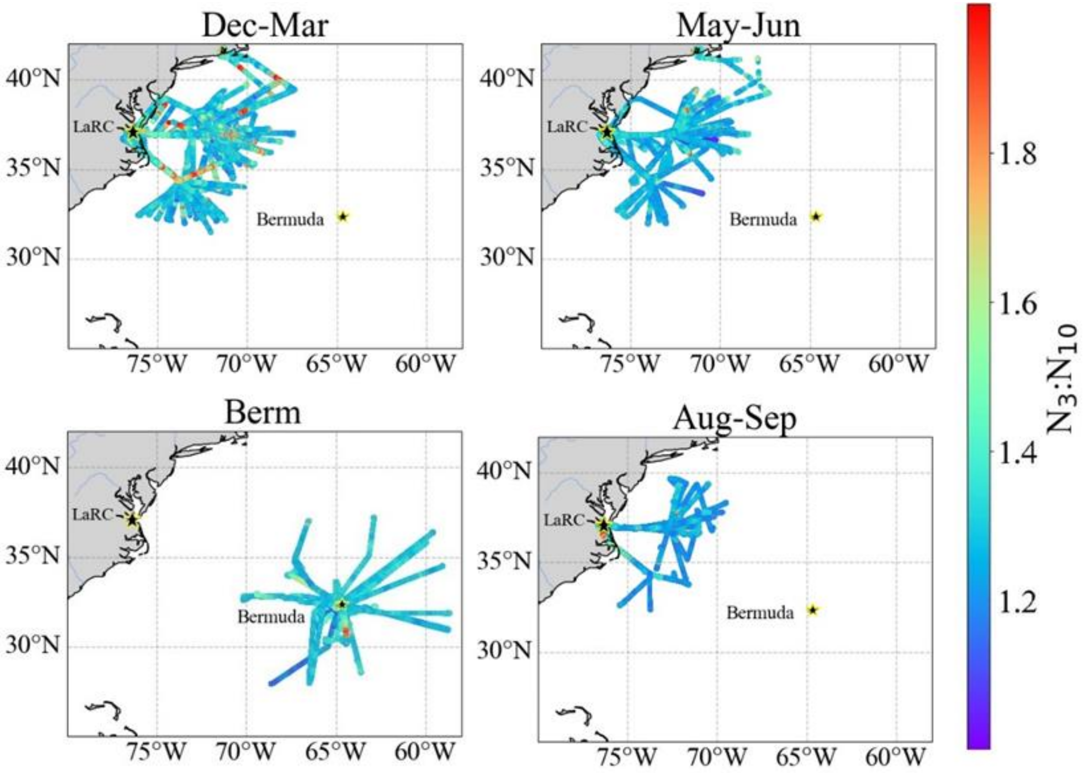
- Ratio of N_3:N_{10}
- Ranges from 1.0 (no clear NPF signature) to higher values (i.e., particles between 3-10 nm)
- Caution: High ratios don't necessarily indicate NPF occurred exactly at the point of measurement



Relevant Datasets

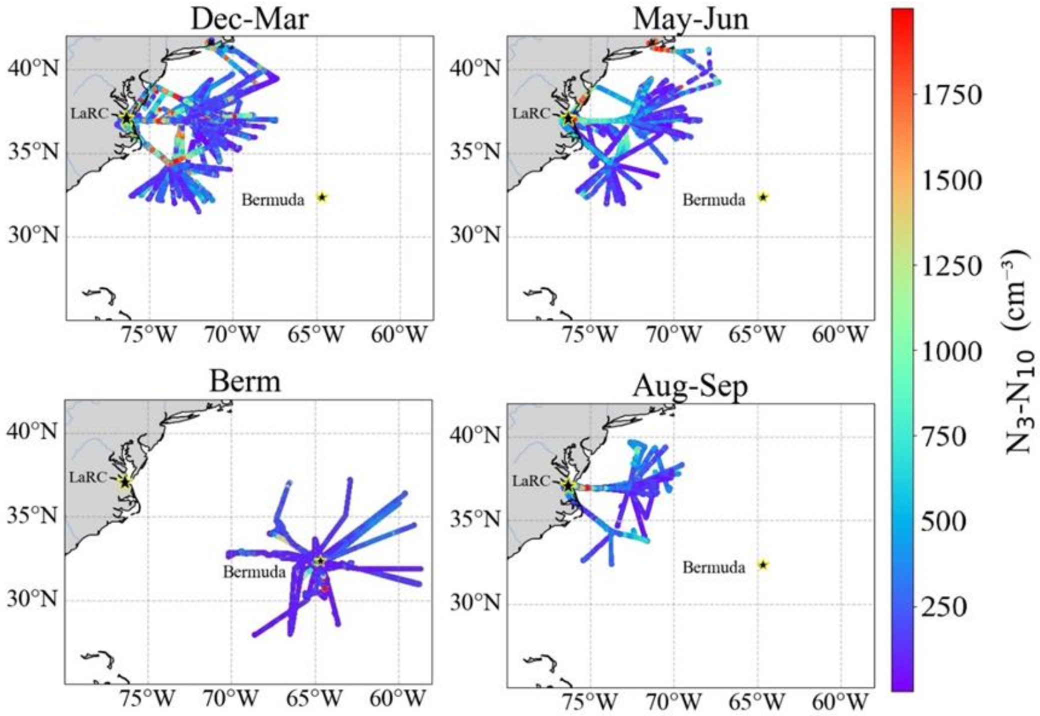
Dataset	Relevant Variable(s)
Applanix navigational data	Basic navigational and motion data
Diode Laser Hygrometer	Water vapor measurements
PICARRO G2401-m	CO2, CH4, CO measurements
2B Technologies Inc. O3 monitor	Ozone (O_3) measurements
Turbulent Air Motion Measurement System (TAMMS)	3D winds, temperature, and pressure
Condensation Particle Counters (CPCs)	Particle concentration (>3 nm and >10 nm)
Scanning Mobility Particle Sizer (SMPS)	Size distribution of ultrafine particles (3-100 nm)
High Resolution Time of Flight Aerosol Mass Spectrometer (HR-ToF-AMS)	Chemical composition of aerosol particles
Fast Cloud Droplet Probe (FCDP)	Droplet size distributions (3-50 µm): used for cloud screening
MERRA-2	SO_2

Seasonal Maps: N_3:N_{10} ratio



- Highest N_3:N_{10} observed during winter months near the U.S. East Coast
- Elevated N_3:N_{10} are also found in remote areas like Bermuda

Seasonal Maps: N_3-N_{10}

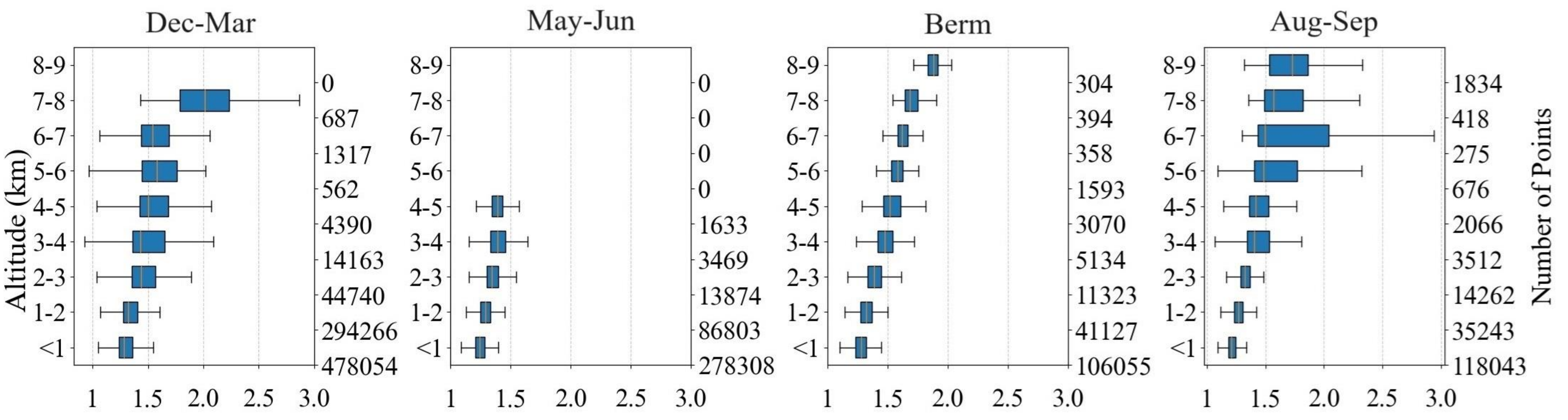


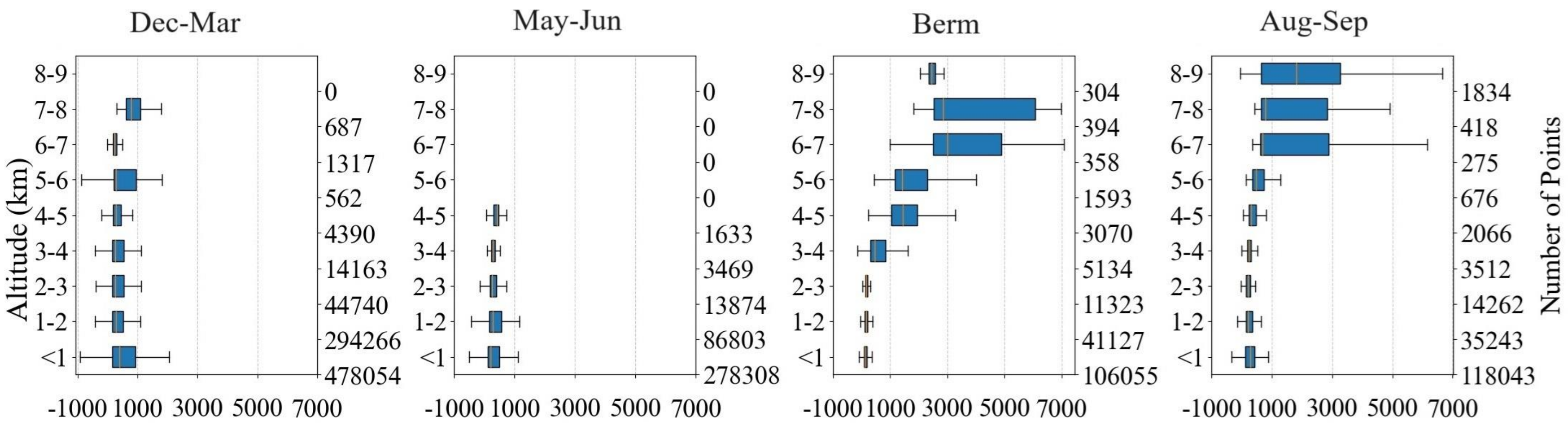
- Highest particle concentrations are near the U.S. East Coast
- Particles represented by "N_3-N_{10}" grow via condensation and coagulation, potentially contributing to the cloud condensation nuclei (CCN) budget

Results: Vertical Distribution

$N_3:N_{10}$

- Gradually increases with altitude.
- Of all ensemble legs, ACT consistently showed highest ratio for all seasons



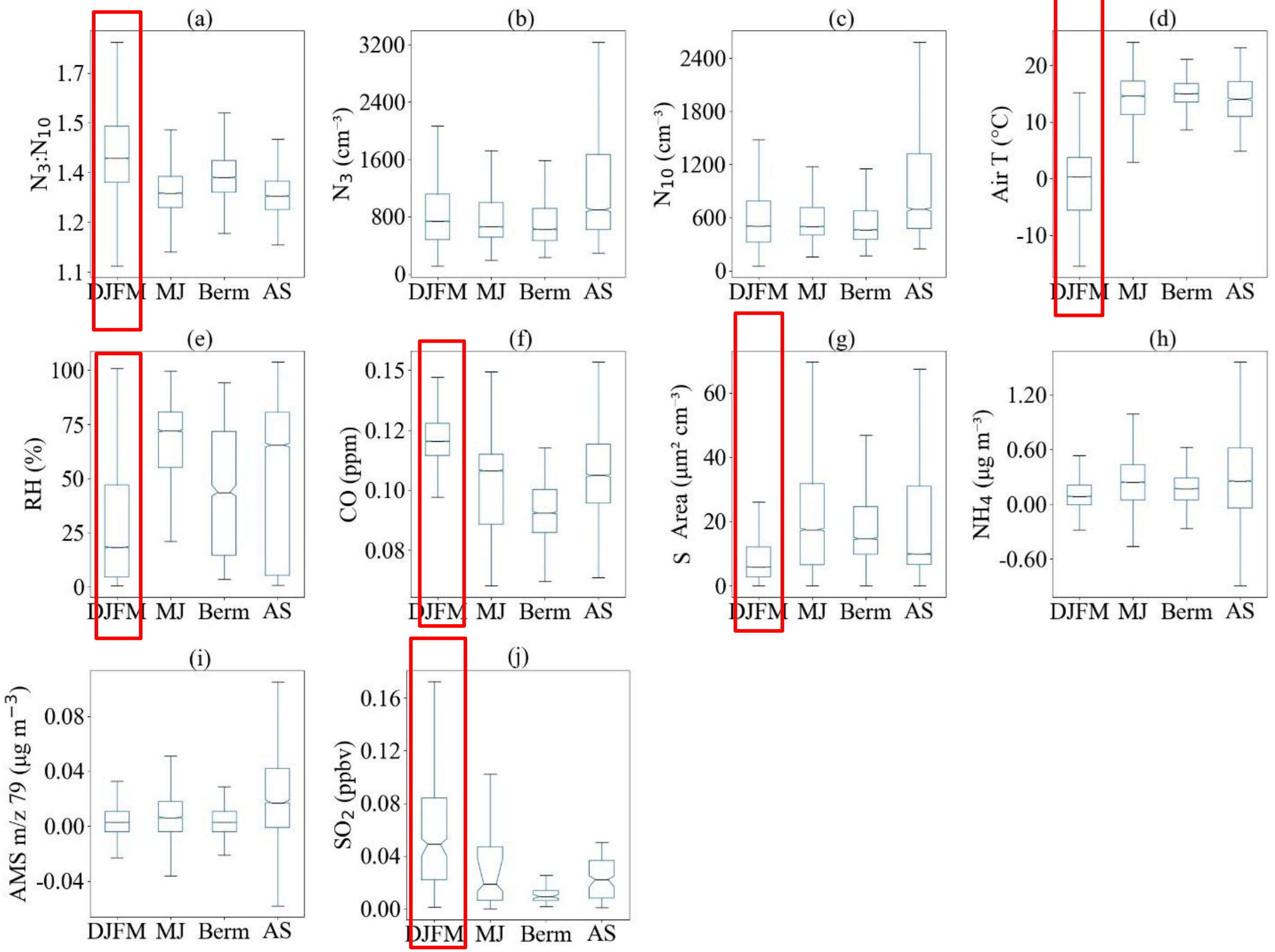


N_3-N_{10}

- Fairly uniform with altitude but shows the most significant increases above 6 km for Aug-Sep and above 3 km for Bermuda.

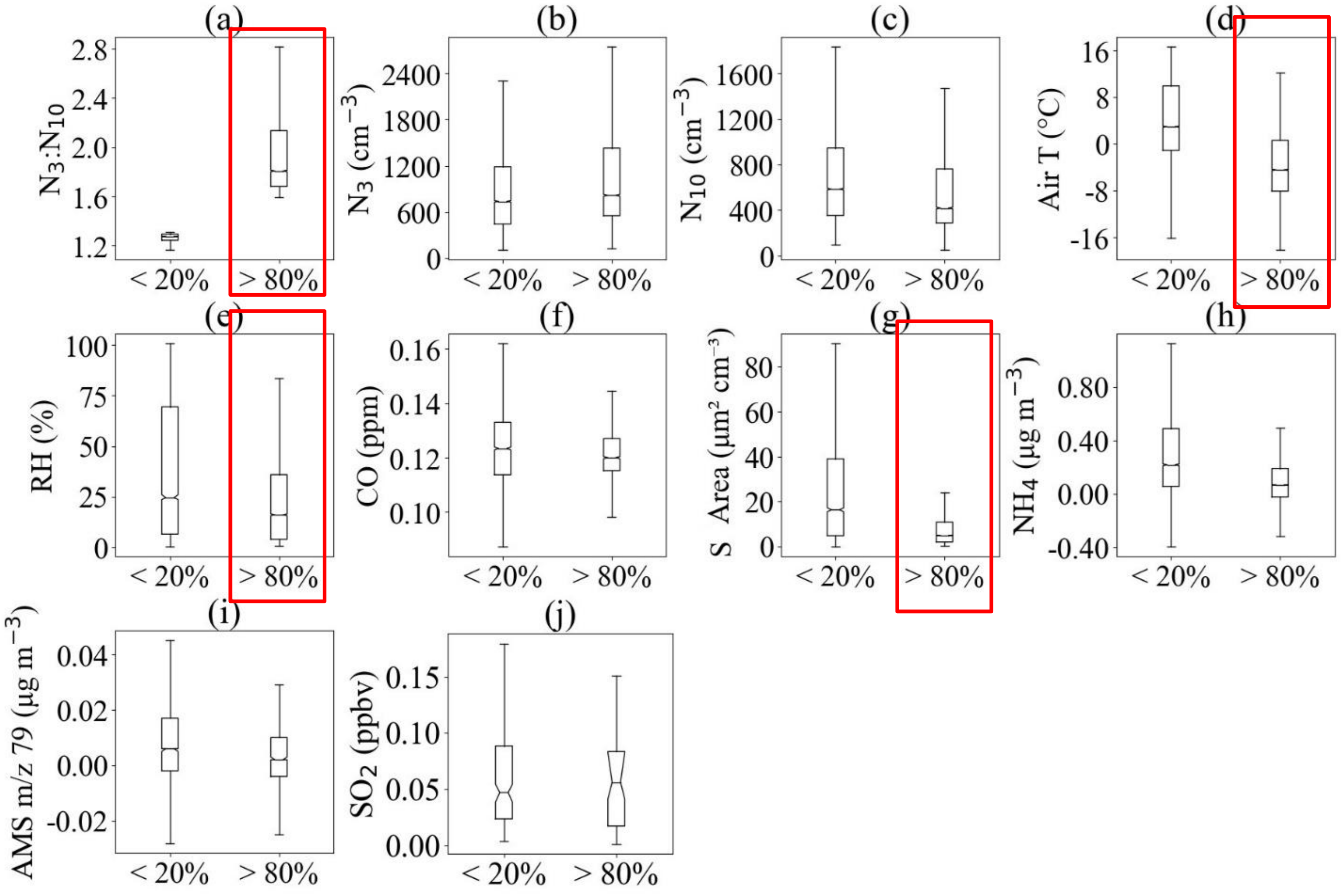
Results: Related Environmental Variables

- Winter (Dec-Mar; DJFM) exhibits higher N_3:N_{10} ratios coincident with lower T/RH/S-Area and higher tracer levels for continental pollution (CO, SO_2)



Seasonal Variation in N_3:N_{10} and Environmental Factors in the Above Cloud Top (ACT) Leg

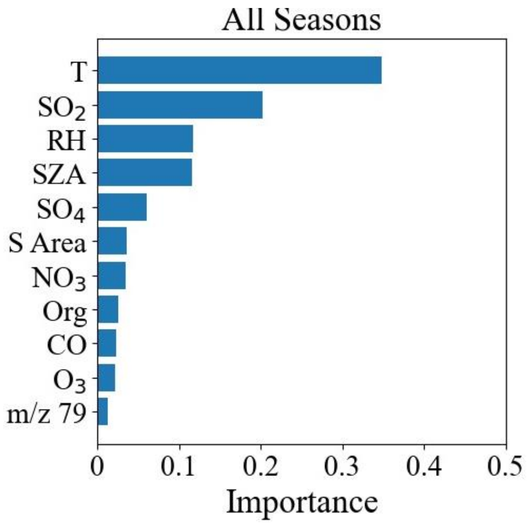
- Focusing now just on the Above Cloud Top leg in winter season, the top 80% of N_3:N_{10} ratios also coincide with:
  - Lower T, RH, S-Area

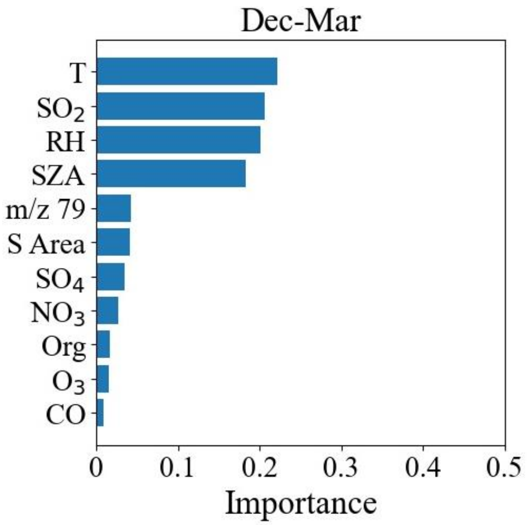


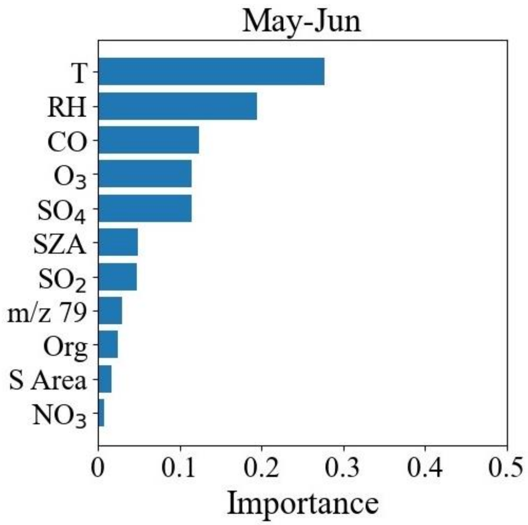
Variation in N_3:N_{10} and Environmental Factors in Dec-Mar in the ACT Leg

Results: Gradient Boosted Regression Tree Modeling

GBRT: A machine learning model that predicts N_3:N_{10} ratios by addressing non-linear relationships between environmental variables.

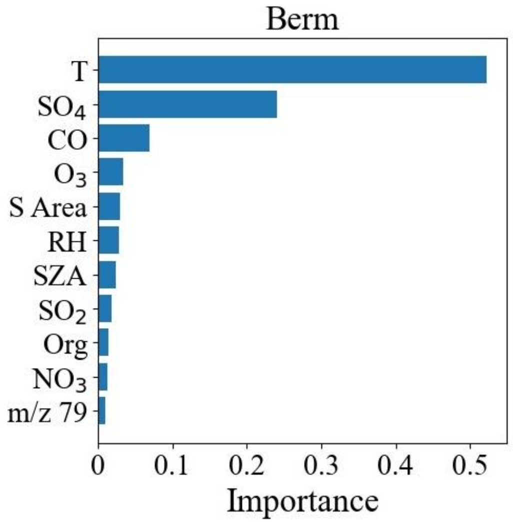


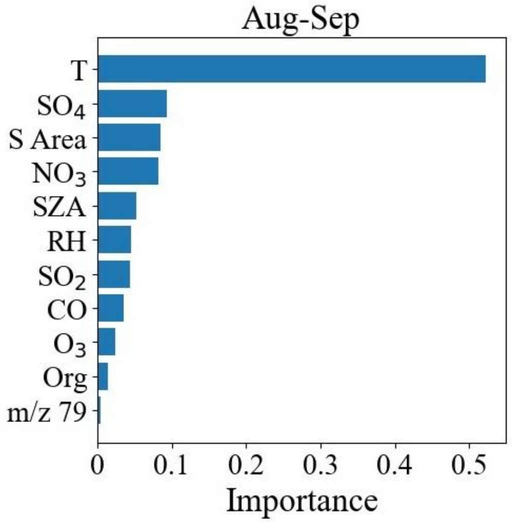




(R^2/MSE):

- all seasons = 0.44/0.035,
- Dec-Mar = 0.43/0.057;
- May-Jun = 0.44/0.004;
- Berm = 0.77/0.002,
- Aug-Sep = 0.71/0.004.



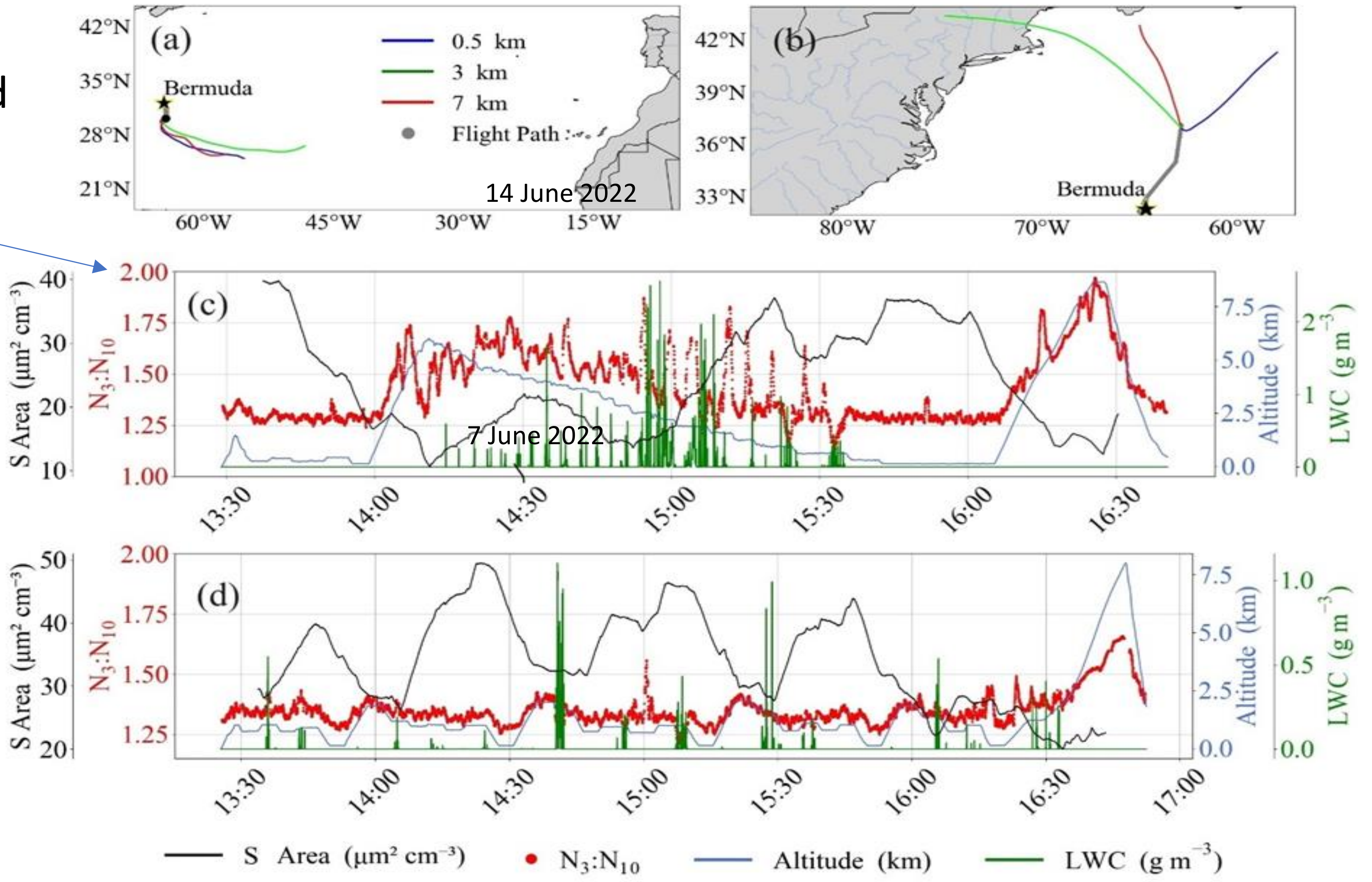


- GBRT confirms that T is the best predictor, which obviously co-varies with altitude.
- SO_2/SO_4, RH, and aerosol surface area also rank well as predictors

Case Study Flights

14 June:

- Growing cumulus cloud; Falcon conducted ~21 level legs at different cloud levels!
- African air mass with dust
- High N_3:N_{10} ratios (>1.5) during the "wall" pattern when Falcon was outside clouds.
- Precipitation acted as an aerosol removal process, promoting NPF
- High actinic flux during outside clouds supported NPF activity.



17 June:

- Clear sky
- N_3:N_{10} < 1.5
- Influenced by an air mass from North America and the North Atlantic.
- Control case flight in same area as 14 June suggesting clouds may be important for more NPF.

Findings and Limitations

Findings

•Seasonal Patterns: Highest in winter, especially above clouds.

•Environmental Drivers: High N_3:N_{10} coincides with low temperature, low relative humidity, and low aerosol surface area.

•High N_3:N_{10} values around Bermuda despite lower pollution levels.

Limitations

•Lack of Precursor Data: No direct measurements of key precursors like NH_3, SO_2, and organic vapors.

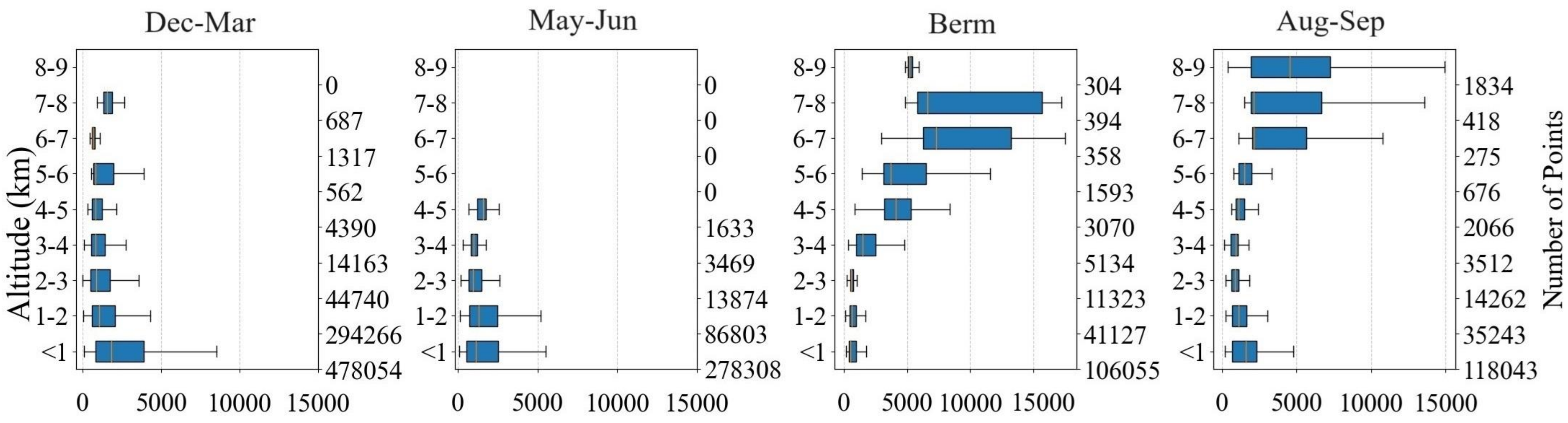
•Regional Limitation: Results focused on the northwest Atlantic.

ACTIVATE Science Team, November 2023
Credit: Xiaojian Zheng

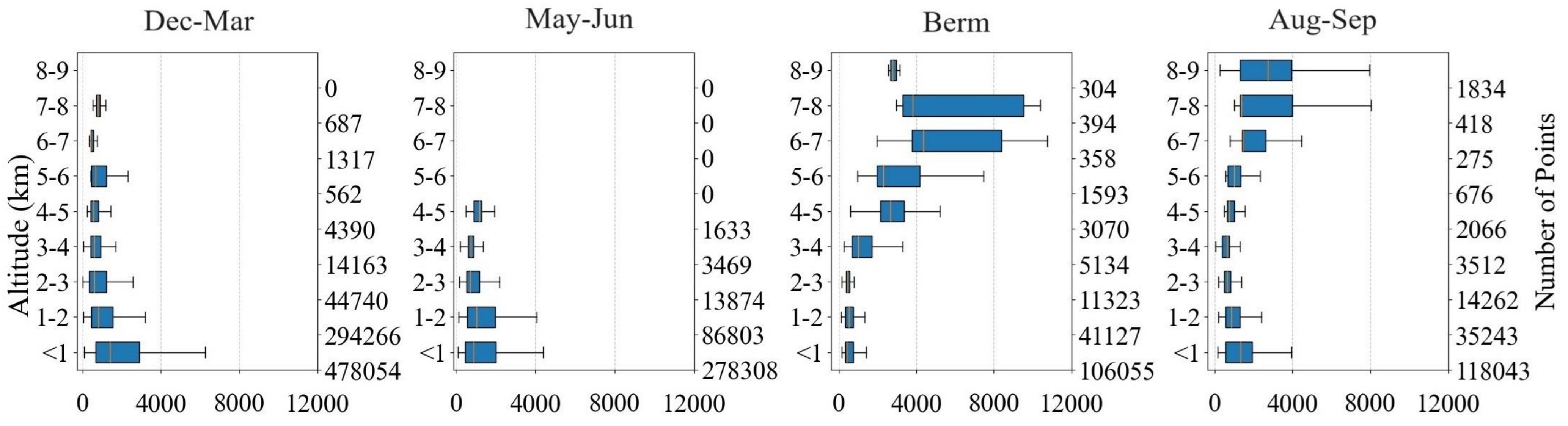
Namdari, S., et al. (2024). A Comprehensive Analysis of New Particle Formation Across the Northwest Atlantic: Analysis of ACTIVATE Airborne Data. Atmospheric Environment. https://doi.org/10.1016/j.atmosenv.2024.120831.

Back Up Slides

N_3


Dec-Mar
May-Jun
Berm
Aug-Sep
Altitude (km)
Number of Points
8-9
7-8
6-7
5-6
4-5
3-4
2-3
1-2
<1
0
5000
10000
15000
0 687 1317 562 4390 14163 44740 294266 478054
0 0 0 0 1633 3469 13874 86803 278308
304 394 358 1593 3070 5134 11323 41127 106055
1834 418 275 676 2066 3512 14262 35243 118043

N_{10}


Dec-Mar
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Altitude (km)
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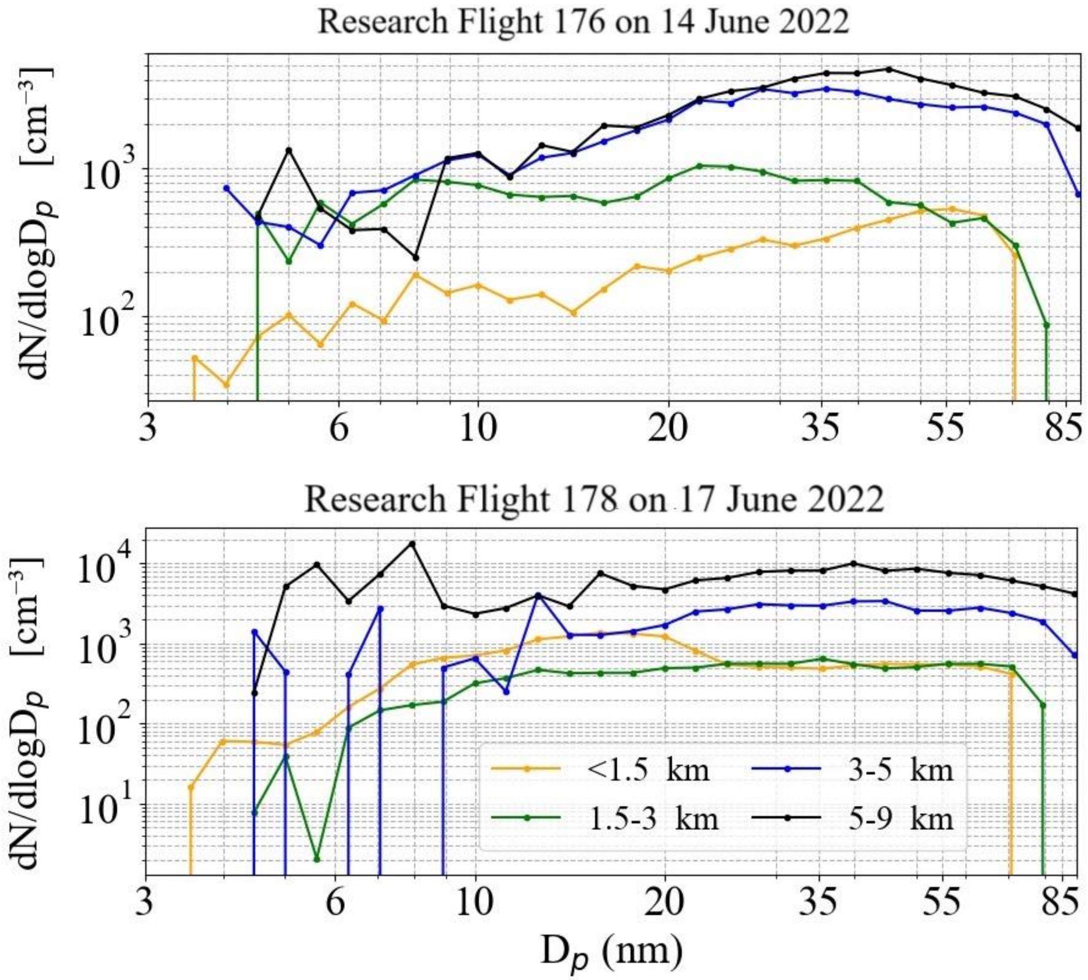
1-2
<1
0
4000
8000
12000
0 687 1317 562 4390 14163 44740 294266 478054
0 0 0 0 1633 3469 13874 86803 278308
304 394 358 1593 3070 5134 11323 41127 106055
1834 418 275 676 2066 3512 14262 35243 118043

	O3	**T**	**Ca**	**WS**	**Lat**	**Lon**	**Chl**	**NH4**	**Org:SO_4**	**RH**	**CO**	**DMS**	**BCphil**	**BCphob**	**OCphil**	**OCphob**	**QV**	**SO_2**	**Org**	**SO_4**	**NO_3**	**Alt**	**S Area**	**SZA**	**m/z 42**	**m/z 79**
O_3	1.00	**-0.39**	**-0.03**	0.22	0.22	-0.34	0.05	0.23	0.18	-0.47	0.64	-0.20	0.13	0.21	0.09	0.22	-0.55	0.23	0.28	**-0.12**	0.21	0.26	0.14	0.20	0.28	0.22
T	**-0.39**	1.00	-0.19	-0.19	-0.27	0.20	0.03	0.04	**0.07**	0.33	-0.43	0.35	0.05	0.04	0.08	0.11	0.87	-0.01	0.35	0.47	-0.22	-0.39	0.20	-0.49	0.34	0.32
Ca	**-0.03**	-0.19	1.00	0.17	0.05	-0.07	0.04	-0.07	**-0.06**	**0.04**	-0.02	-0.01	-0.05	**-0.10**	-0.05	-0.10	-0.18	-0.14	-0.13	**-0.10**	-0.04	0.05	-0.05	0.04	-0.12	-0.11
WS	0.22	-0.19	0.17	1.00	0.24	-0.30	**0.02**	0.02	0.05	-0.16	0.16	0.10	0.06	0.09	0.10	0.14	-0.18	0.04	0.04	**-0.10**	0.01	0.21	0.04	0.18	0.03	0.03
Lat	0.22	-0.27	0.05	0.24	1.00	-0.43	0.08	0.19	**0.12**	-0.11	0.35	**0.00**	0.11	0.37	**0.06**	**0.12**	-0.25	0.32	0.16	**0.01**	0.27	-0.15	0.08	0.20	0.17	0.16
Lon	-0.34	0.20	-0.07	-0.30	-0.43	1.00	-0.07	-0.21	-0.13	0.26	-0.40	0.12	-0.13	-0.31	-0.08	-0.22	0.26	-0.34	-0.21	**-0.05**	-0.26	**0.10**	-0.12	-0.25	-0.21	-0.20
Chl	0.05	0.03	0.04	**0.02**	0.08	-0.07	1.00	0.27	**0.10**	0.13	0.20	**0.07**	0.03	0.16	0.01	0.06	**0.05**	0.15	0.24	0.17	0.27	-0.07	0.26	0.01	0.23	0.22
NH_4	0.23	0.04	-0.07	0.02	0.19	-0.21	0.27	1.00	**0.11**	0.13	0.43	0.00	0.08	0.35	0.02	0.14	0.05	0.44	0.56	0.63	0.69	-0.18	0.49	-0.05	0.55	0.46
Org:SO_4	0.18	**0.07**	**-0.06**	0.05	**0.12**	-0.13	**0.10**	**0.11**	1.00	-0.09	0.19	**-0.01**	0.07	0.23	0.06	0.18	0.00	0.23	0.38	-0.02	0.14	**0.02**	0.23	**-0.04**	0.36	0.38
RH	-0.47	0.33	**0.04**	-0.16	-0.11	0.26	0.13	0.13	-0.09	1.00	-0.19	0.26	-0.10	-0.01	-0.09	-0.09	0.55	-0.14	0.10	0.39	0.00	-0.30	0.13	-0.20	0.10	0.08
CO	0.64	-0.43	-0.02	0.16	0.35	-0.40	0.20	0.43	0.19	-0.19	1.00	-0.16	0.08	0.48	0.01	0.30	-0.50	0.51	0.32	0.00	0.48	-0.23	0.28	0.42	0.30	0.28
DMS	-0.20	0.35	-0.01	0.10	**0.00**	0.12	**0.07**	0.00	**-0.01**	0.26	-0.16	1.00	-0.02	0.02	-0.01	0.00	0.45	**-0.01**	**0.08**	0.18	-0.09	-0.25	0.11	-0.20	**0.07**	**0.08**
BCphil	0.13	0.05	-0.05	0.06	0.11	-0.13	0.03	0.08	0.07	-0.10	0.08	-0.02	1.00	0.34	0.99	0.37	-0.03	0.14	0.13	0.04	0.07	-0.21	0.09	-0.04	0.13	0.12
BCphob	0.21	0.04	**-0.10**	0.09	0.37	-0.31	0.16	0.35	0.23	-0.01	0.48	0.02	0.34	1.00	0.27	0.74	**0.00**	0.67	0.41	0.15	0.38	-0.30	0.36	0.08	0.39	0.40
OCphil	0.09	0.08	-0.05	0.10	**0.06**	-0.08	0.01	0.02	0.06	-0.09	0.01	-0.01	0.99	0.27	1.00	0.39	0.01	0.07	0.10	0.03	0.01	-0.13	0.07	**-0.06**	0.10	0.10
OCphob	0.22	0.11	-0.10	0.14	**0.12**	-0.22	0.06	0.14	0.18	-0.09	0.30	0.00	0.37	0.74	0.39	1.00	0.02	0.40	0.25	**0.05**	0.14	-0.16	0.27	0.05	0.23	0.23
QV	-0.55	0.87	-0.18	-0.18	-0.25	0.26	**0.05**	0.05	0.00	0.55	-0.50	0.45	-0.03	**0.00**	0.01	0.02	1.00	-0.07	0.30	0.54	-0.19	-0.35	0.18	-0.48	0.29	0.27
SO_2	0.23	-0.01	-0.14	0.04	0.32	-0.34	0.15	0.44	0.23	-0.14	0.51	**-0.01**	0.14	0.67	0.07	0.40	-0.07	1.00	0.42	0.20	0.51	-0.26	0.34	0.13	0.39	0.40
Org	0.28	0.35	-0.13	0.04	0.16	-0.21	0.24	0.56	0.38	0.10	0.32	**0.08**	0.13	0.41	0.10	0.25	0.30	0.42	1.00	0.55	0.35	-0.14	0.63	**-0.25**	0.98	0.91
SO_4	**-0.12**	0.47	**-0.10**	**-0.10**	**0.01**	**-0.05**	0.17	0.63	-0.02	0.39	0.00	0.18	0.04	0.15	0.03	**0.05**	0.54	0.20	0.55	1.00	0.20	-0.37	0.44	-0.32	0.54	0.43
NO_3	0.21	-0.22	-0.04	0.01	0.27	-0.26	0.27	0.69	0.14	0.00	0.48	-0.09	0.07	0.38	0.01	0.14	-0.19	0.51	0.35	0.20	1.00	**-0.09**	0.36	0.16	0.33	0.31
Alt	0.26	-0.39	0.05	0.21	-0.15	**0.10**	-0.07	-0.18	**0.02**	-0.30	-0.23	-0.25	-0.21	-0.30	-0.13	-0.16	-0.35	-0.26	-0.14	-0.37	**-0.09**	1.00	-0.13	-0.07	-0.12	-0.13
S Area	0.14	0.20	-0.05	0.04	0.08	-0.12	0.26	0.49	0.23	0.13	0.28	0.11	0.09	0.36	0.07	0.27	0.18	0.34	0.63	0.44	0.36	-0.13	1.00	-0.09	0.61	0.57
SZA	0.20	-0.49	0.04	0.18	0.20	-0.25	0.01	-0.05	**-0.04**	-0.20	0.42	-0.20	-0.04	0.08	**-0.06**	0.05	-0.48	0.13	**-0.25**	-0.32	0.16	-0.07	-0.09	1.00	**-0.25**	**-0.22**
m/z 42	0.28	0.34	-0.12	0.03	0.17	-0.21	0.23	0.55	0.36	0.10	0.30	**0.07**	0.13	0.39	0.10	0.23	0.29	0.39	0.98	0.54	0.33	-0.12	0.61	**-0.25**	1.00	0.90
m/z 79	0.22	0.32	-0.11	0.03	0.16	-0.20	0.22	0.46	0.38	0.08	0.28	**0.08**	0.12	0.40	0.10	0.23	0.27	0.40	0.91	0.43	0.31	-0.13	0.57	**-0.22**	0.90	1.00

SMPS Size Distribution:

14 June: Higher concentrations of 3-10 nm particles with increasing altitude.

17 June: Elevated particle concentrations at 5-9 km, contrasting with lower altitudes.



Median number size distributions from SMPS for different altitude bins