



Supplement of

Organic enrichment in droplet residual particles relative to out of cloud over the northwestern Atlantic: analysis of airborne ACTIVATE data

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Table S1. Average concentrations of submicrometer aerosol species measured by an airborne 23 24 AMS for different seasons associated with ACTIVATE deployments 1-4. Also shown are 25 mass fractions of each AMS species relative to total AMS mass, in addition to the ratio of 26 m/z 44 to total organic mass (i.e., f₄₄). Non-CAO and CAO categories include samples 27 collected between January and March. CVI = droplet residual particle measurements in 28 cloud; BCB = below cloud base, ACT = above cloud top, BBL = below boundary layer top, ABL = above boundary layer top. Corresponding standard deviations and number of points 29 are provided in Table S2. 30

_	(Non-CAO/CAO/May-Jun/Aug-Sep)						
_	CVI	BCB	ACT	BBL	ABL		
Organic (µg m ⁻³)	-	1.07/0.67/1.49/3.27	0.61/0.19/2.62/3.04	2.59/1.16/3.49/4.46	0.94/0.57/5.28/5.57		
Sulfate ($\mu g m^{-3}$)	-	0.93/0.79/1.71/1.35	0.53/0.26/1.23/1.11	0.80/0.57/1.17/1.77	0.51/0.45/1.26/2.13		
Nitrate ($\mu g m^{-3}$)	-	0.40/0.21/0.07/0.16	0.19/0.05/0.14/0.11	0.79/0.93/0.17/0.21	0.14/0.32/0.26/0.19		
Ammonium ($\mu g m^{-3}$)	-	0.45/0.32/0.36/0.36	0.28/0.10/0.41/0.37	0.67/0.65/0.38/0.53	0.26/0.30/0.51/0.63		
Chloride ($\mu g m^{-3}$)	-	0.03/0.02/0.03/0.03	0.02/0.01/0.02/0.02	0.05/0.01/0.02/0.02	0.01/0.01/0.02/0.02		
OrganicMF	0.55/0.60/0.68/0.61	0.40/0.34/0.35/0.48	0.28/0.29/0.42/0.51	0.50/0.39/0.63/0.57	0.44/0.32/0.65/0.54		
SulfatemF	0.24/0.19/0.14/0.14	0.39/0.45/0.53/0.39	0.42/0.46/0.43/0.34	0.24/0.20/0.26/0.33	0.35/0.36/0.24/0.35		
NitratemF	0.05/0.05/0.05/0.05	0.08/0.07/0.02/0.03	0.08/0.07/0.03/0.03	0.11/0.22/0.03/0.03	0.06/0.14/0.03/0.03		
AmmoniumMF	0.09/0.08/0.07/0.09	0.13/0.13/0.10/0.08	0.20/0.16/0.12/0.10	0.14/0.18/0.08/0.07	0.14/0.16/0.07/0.08		
Chloridem	0.06/0.08/0.06/0.10	0.01/0.01/0.01/0.01	0.01/0.02/0.01/0.01	0.01/0.01/0.01/0.00	0.01/0.03/0.00/0.00		
f 44	0.33/0.34/0.24/0.37	0.15/0.13/0.11/0.14	0.26/0.16/0.12/0.15	0.16/0.14/0.12/0.14	0.17/0.14/0.11/0.14		

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- 33 Table S2. Standard deviations and the number of points for parameters measured by the
- 34 AMS instrument. Note that numbers of points refer to entire legs for which calculations
- 35 were conducted using raw data rather than the raw data points per leg. Non-CAO and
- 36 CAO categories include samples collected between January and March. CVI = droplet
- 37 residual particle measurements in cloud; BCB = below cloud base, ACT = above cloud top,
- **BBL** = below boundary layer top, ABL = above boundary layer top. Corresponding
- 39 average values are provided in Table S1.

	(Non-CAO/CAO/May-Jun/Aug-Sep)						
	CVI	BCB	ACT	BBL	ABL		
Organic (µg m ⁻³)	-	0.63/0.43/1.84/3.93	1.06/0.14/5.08/4.50	2.13/0.69/2.58/3.25	1.01/0.57/3.92/5.86		
Sulfate ($\mu g m^{-3}$)	-	0.52/0.50/0.68/0.83	0.45/0.16/1.04/1.30	0.38/0.35/0.53/0.82	0.27/0.42/0.71/2.10		
Nitrate ($\mu g m^{-3}$)	-	0.82/0.44/0.07/0.16	0.37/0.07/0.27/0.15	1.05/0.82/0.19/0.18	0.25/0.48/0.25/0.18		
Ammonium ($\mu g m^{-3}$)	-	0.52/0.33/0.25/0.37	0.32/0.08/0.50/0.45	0.58/0.50/0.22/0.38	0.21/0.36/0.35/0.67		
Chloride ($\mu g m^{-3}$)	-	0.03/0.02/0.02/0.03	0.05/0.01/0.02/0.03	0.06/0.01/0.02/0.02	0.01/0.01/0.01/0.02		
Organic _{MF}	0.15/0.16/0.18/0.20	0.14/0.11/0.21/0.23	0.21/0.15/0.21/0.21	0.16/0.09/0.15/0.16	0.17/0.15/0.18/0.23		
Sulfate _{MF}	0.12/0.14/0.13/0.12	0.15/0.15/0.21/0.22	0.18/0.16/0.18/0.19	0.15/0.07/0.16/0.18	0.15/0.16/0.17/0.22		
Nitrate _{MF}	0.05/0.06/0.06/0.05	0.10/0.08/0.01/0.02	0.09/0.07/0.03/0.03	0.08/0.11/0.02/0.02	0.04/0.09/0.03/0.02		
Ammonium _{MF}	0.10/0.09/0.10/0.14	0.06/0.06/0.05/0.07	0.10/0.08/0.08/0.09	0.05/0.04/0.04/0.04	0.08/0.06/0.04/0.05		
Chloride _{MF}	0.08/0.08/0.08/0.14	0.01/0.01/0.01/0.01	0.02/0.01/0.01/0.02	0.01/0.01/0.01/0.01	0.01/0.05/0.00/0.00		
\mathbf{f}_{44}	0.14/0.21/0.51/0.59	0.12/0.04/0.07/0.11	0.19/0.11/0.09/0.11	0.02/0.04/0.03/0.04	0.09/0.03/0.03/0.08		
no. points	180/96/386/228	32/21/70/41	31/22/67/41	24/8/45/27	24/12/97/53		

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43 Figure S1. Concentration weighted trajectory maps for organic aerosol concentrations as

44 measured by an AMS on the Falcon during different ACTIVATE deployments (All data,

January-March 2020 and 2021, May-June 2021, August-September 2020). These are based
on 29,164 cloud-free AMS data points. The pink stars represent NASA Langley Research

on 29,164 cloud-free AMS data points. The pink stars represent NASA Langley Research
Center (Hampton, Virginia) and Bermuda for reference. Color bar scales differ to show

48 variability better within a given panel.







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61 Figure S3. Time series from research flight 10 (28 February 2020) of (top) AMS mass

62 concentrations of sulfate and organic in addition to f₄₄, and (bottom) Falcon altitude and

63 FCDP droplet number concentration. The CVI Flag horizontal bar indicators represent

64 when aerosol sampling was conducted downstream of the CVI, with the vertical orange

bars indicating specifically those CVI times where sampling was out of cloud (i.e., data

66 serving as blanks).