Electronic Supplementary Material (ESI) for Environmental Science: Atmospheres.

SUPPLEMENTARY INFORMATION

Investigating the evolution of water-soluble organic carbon in evaporating cloud water

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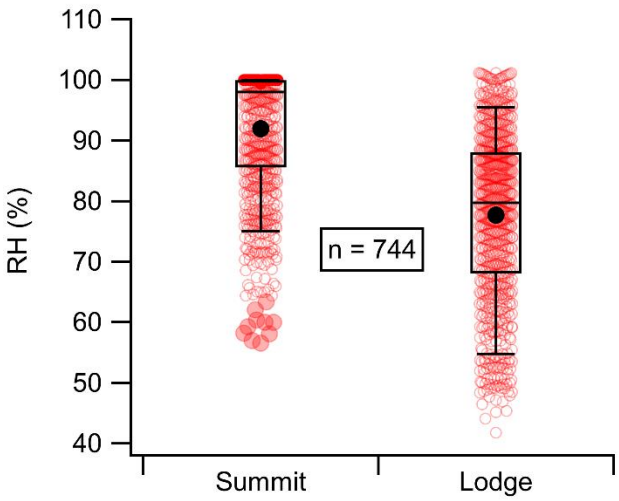

Figure S1: Box-plots of relative humidity (RH) measured at the WFM summit and the downwind lodge site in the month of August 2017. Circular symbols are respective means.

Table S1: Analytical parameters of cloud water sample from bypass channel (unperturbed)

Sample id	Date (dd/mm/yy)	Time (hrs)	Avg Temp (^oC)	LWC (g m^{-3})	CO (ppbv)	NOx (ppbv)	O3 (ppbv)	SO2 (ppbv)	*$WSOC_{byp}$ (µg-C L^{-1})	*Sulfate (µg L^{-1})	*Oxalate (µg L^{-1})	*Abs_{byp} (A.U.)
CW1	18/8/17	0500-0800	14.1	1.23	119.5	0.53	31.3	0.030	77	29.1	14.7	0.0191
CW2	18/8/17	0900-1000	15.5	1.35	102.0	0.33	34.0	0.00	76	40.7	14.8	0.0205
CW3	13/8/17	0500-0800	9.1	0.62	103.8	0.18	34.0	0.025	50	11.2	14.4	0.0161
CW4	13/8/17	0800-0900	8.4	0.42	81.0	0.18	37.0	0.020	59	15.9	14.2	0.0270
CW5	12/8/17	0500-0800	12.3	1.12	114.8	0.28	41.5	0.015	167	149.5	17.7	0.0279

* Measurements performed at UMBC lab by aerosolizing bulk cloud water samples. Other measurements were performed at WFM during cloud water sampling

Table S2: Measured parameters of 12-hour composite cloud water samples by Adirondack Lake Survey Corporation (ALSC)

Equivalent Sample Id*	Date (dd/mm/yy)	Time (hrs)	Net sample collection time (hrs)	Temp (°C)	Wind direction	pH	NH_4 (µg L^{-1})	SO_4 (µg L^{-1})	NO_3 (µg L^{-1})	K (µg L^{-1})	TOC (µg-C L^{-1})
CW1	18/08/2017	6:00	6.09	15.6	237	4.793	887.6	1834.9	1456.1	17.3	4653.1
CW1+CW2	18/08/2017	18:00	7.22	14.0	255	5.542	1831.0	2491.4	3255.8	52.8	3874.1
CW3	13/08/2017	6:00	4.60	8.9	293	4.969	92.1	476.1	301.2	18.6	2319.2
	12/08/2017	18:00	10.11	11.2	258	4.918	2465.3	4124.5	3513.9	68.4	5944.4
CW5	12/08/2017	6:00	6.16	13.1	247	4.135	1541.0	4951.2	3163.6	64.4	6562.0

*An equivalent sample id is provided to these samples to ease in understanding, even though these are 12-hour composite samples.

Text S1: Cloud water analysis

The details of the cloud water samples, analytical results and other measurements (gases and meteorological) performed at WFM during the cloud water sampling are presented in Table S1. CW1, CW3 and CW5 were 3-hour composite samples, while CW2 and CW4 were 1-hour samples collected in the morning. Note the results in table S1 are from the measurements of PILS samples collected from the aerosolized cloud water samples. Therefore, even though the measurements are representatives of cloud water samples, they are not concentrations in the CW. Due to limited sample volume, we were unable to conduct comprehensive analysis of CW. Table S2 list the measurements performed by Adirondack Lake Survey Corporation (ALSC) on 12-hour composite samples. The 12-hour composite samples were from 6:00 am to 6:00 pm and 6:00 pm to 6:00 am (next day). Based on the sample collection times, equivalent identities are provided to 12-hr composite samples consistent with samples analyzed at UMBC (Table S1 and S2). The results in table S2 represent the actual concentrations of species in CW. Even though 12-hr composite samples are not exact representation of samples analyzed at UMBC, they can still provide some qualitative inferences. For example, it can be seen in table S1 that in CW5, all measured species were significantly large, suggesting a much stronger anthropogenic influence on this cloud water sample. The observation is consistent with the results of equivalent CW5 (6:00 am sample) listed in table S2 where the sulfate concentration and pH were significantly high and low respectively. Similarly, CW3 (Table S1) and equivalent CW3 (Table S2) are consistent with each other and appear to have least anthropogenic influence within the sample set.

Figure S2 shows the trend of various species plotted for the month of August. Note here the $PM_{2.5}$ was measured at the marble mountain lodge, which is located on WFM at 604 m a.s.l.

$PM_{2.5}$, NOy and O_3 were correlated during the month (R^2 = 0.271 – 0.670) suggesting their regional transportation. The $PM_{2.5}$ data reveals a relatively highly polluted air mass arrived at WFM on August 11 (peak $PM_{2.5}$ = 25 μg m^{-3}), and on the next day early morning CW5 sample was collected (Table S1 and S2). The significantly large values of the species measured in CW5 were likely due to that pollution event.

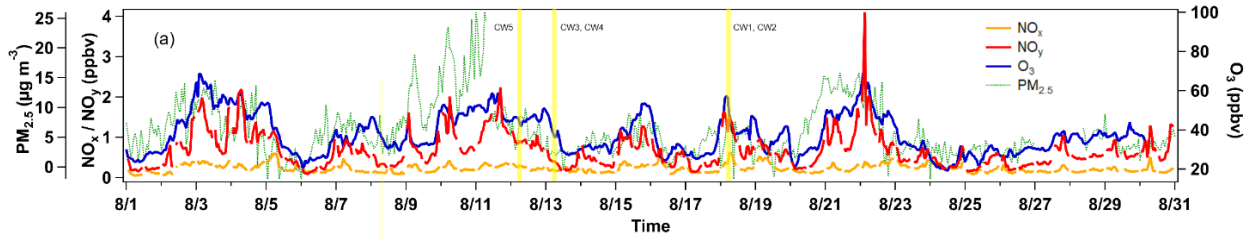

Figure S2 Time series of NOx, NOy, O_3 and $PM_{2.5}$ (lodge) in the month of August. The yellow bands are when our cloud water samples were collected. Note CW1 and CW2 are combined in one band as the samples collected were close in time. Similarly, CW3 and CW4 are also combined in one band.

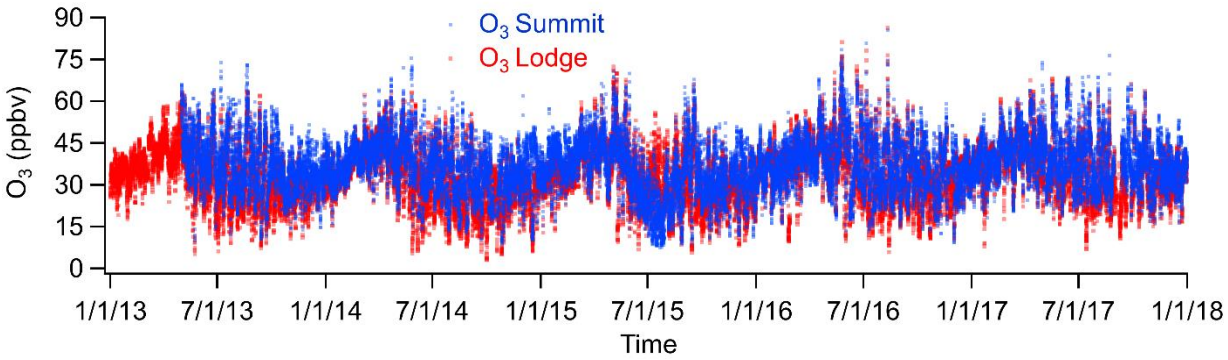

Figure S3: Five year (2013-2017) hourly O_3 measurements performed at the WFM summit and WFM lodge. WFM lodge is often downwind of the WFM summit (Lance et al., 2020).[1] The

strong relationship in trend between O_3 data at both sites suggest that the air-parcel sampled at the lodge is likely processed at the summit (upwind). The data is available at the following link: http://atmoschem.asrc.cestm.albany.edu/~aqm/

References:

1 S. Lance, J. Zhang, J. J. Schwab, P. Casson, R. E. Brandt, D. R. Fitzjarrald, M. J. Schwab, J. Sicker, C.-H. Lu, S.-P. Chen, J. Yun, J. M. Freedman, B. Shrestha, Q. Min, M. Beauharnois, B. Crandall, E. Joseph, M. J. Brewer, J. R. Minder, D. Orlowski, A. Christiansen, A. G. Carlton and M. C. Barth, Overview of the CPOC Pilot Study at Whiteface Mountain, NY: Cloud Processing of Organics within Clouds (CPOC), *Bull. Am. Meteorol. Soc.*, 2020, **preprint**, 1–63.