HCOOH in the remote atmosphere: Constraints from Atmospheric Tomography (ATom) airborne observations

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Supporting Information:

S1.Tables S1-S3; Figures S1-S4

Table S1. Airborne HCOOH measurements in the remote atmosphere*

Campaign	Aircraft platform	Location	Timeframe	Technique	References
PEM-WEST A	DC-8	Northwest Pacific	Sep-Oct 1991	Mist Chamber/Ion Chromatography (MC/IC)	Hoell et al. (1996)[1]
TRACE-A	DC-8	South Atlantic	Sep-Oct 1992	MC/IC	Fishman et al. (1996)[2]
PEM-WEST B	DC-8	Northwest Pacific	Feb-Mar 1994	MC/IC	Hoell et al. (1997)[3]
PEM-Tropics A	DC-8	South tropical Pacific	Aug-Sep 1996	MC/IC	Hoell et al. (1999)[4]
SONEX	DC-8	North Atlantic	Oct-Nov 1997	MC/IC	Singh et al. (1999, 20000)[5, 6]
INTEX-B	C-130	Northeast Pacific	Apr-May 2006	CIT-CIMS (CF_3O^-)	Kleb et al. (2011)[7]; Singh et al. (2009)[8]; Crounse et al. (2006)[9]

*Flight tracks at: https://www-gte.larc.nasa.gov/GTE2/missions/Aircraft_Based_Missions/Mission_Maps/Mission_Maps.htm

Table S2. Reported surface seawater + lakewater concentrations of HCOOH and formate

Timeframe	Location	Seawater concentration	Reference
1958-1959	Northeast Pacific surface water	Formic acid: 3.91 (0.65-8.26) µM	Koyama and Thoivipson (1964)[10]
	Inshore surface water	Formic acid: 10 (0.13-22.39) µM	
Oct 1977	Lake Kizaki	Formic acid: 1.67-2.5 µM	Hama and Handa (1981)[11]
Mar, Apr, Nov 1987	Biscayne Bay seawater	Formate: 0.19-0.8 µM	Kieber et al. (2002)[12]
	Oligotrophic Sargasso seawater Biscayne Bay coastal seawater	Formate: 0.08-0.19 µM Formate: 0.21-0.29 µM	Vaughan and Mopper (1990)[13]
Jul 1988	Black Sea	Formate: <10 µM	Mopper and Kieber (1991)[14]

Table S3. Global HCOOH sinks

	Paulot et al. (2011)[15]	Millet et al. (2015)[16]	This work
Simulation year	2004-2008 average	2013	2017
Total sink	56.7 Tg/y	61.3 Tg/y	44.1 Tg/y
Dry deposition	24.7 Tg/y	20.8 Tg/y	15.0 Tg/y (14% over ocean)
Boundary layer lifetime against dry deposition			4 d (over ocean) 3.3 d (over land)
Wet deposition	20.1 Tg/y	29.8 Tg/y	21.0 Tg/y (17% over ocean)
Boundary layer lifetime against wet deposition			2.4 d (over ocean) 2.4 d (over land)

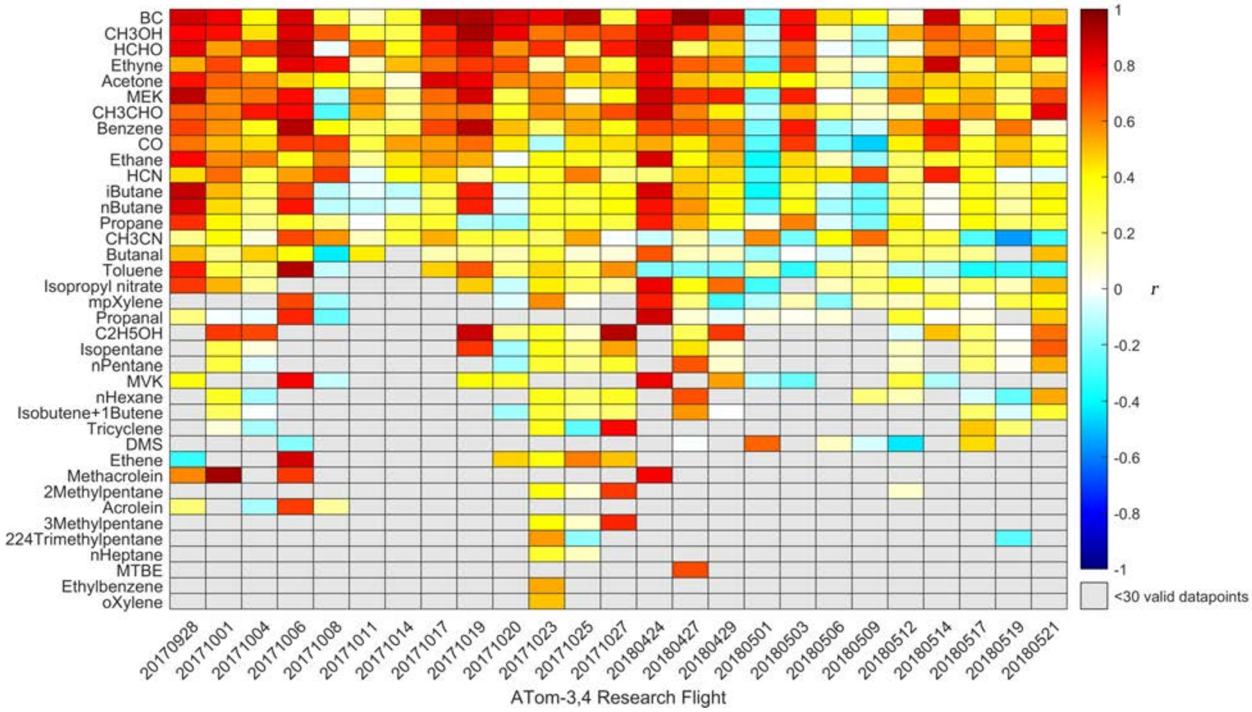


Figure S1. Pearson correlation coefficient (r) between HCOOH and other observed species for each flight during ATom-3 and 4. Grey cells indicate compounds with <30 valid datapoints (i.e., excluding missing data and values below the instrumental limit of detection) for a given flight. Instrumental coverage is as described in **Section 2.1.2** of the main text. Rows are sorted by the number of flights meeting the data threshold (first) and by the mean species-HCOOH r value across flights (second).

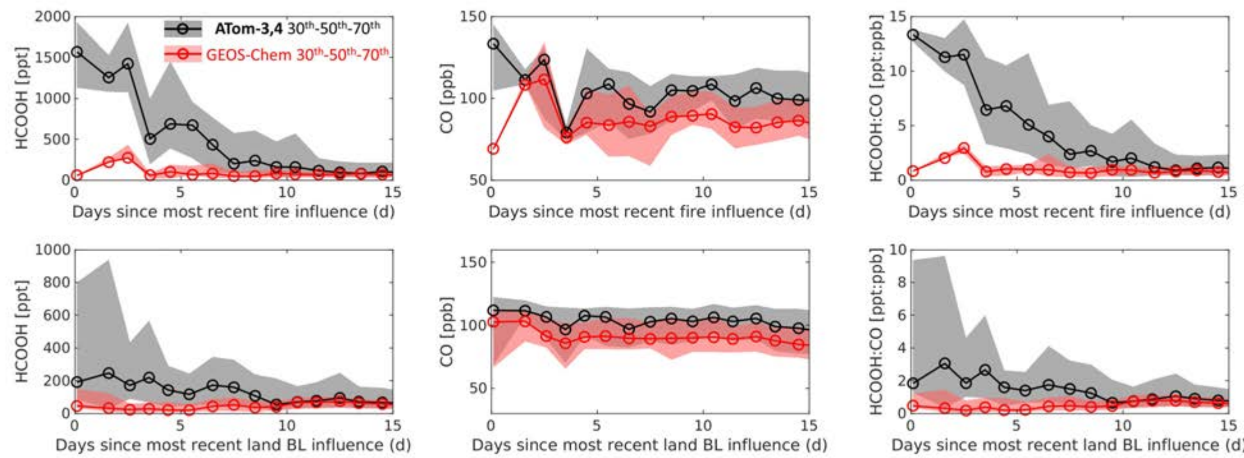


Figure S2. Back trajectory-derived influence from fires and the land boundary layer (BL) on HCOOH, CO, and the HCOOH:CO ratio during ATom-3,4. Data are plotted as the median of the 1-min observed (black) and predicted (red) values, binned to the most recent day since a given influence process. The time since most recent influence is calculated based on the average across a 245-trajectory cluster released each minute of the flight. Shaded regions indicate the 30th-70th percentile range. Data for 05/14/2018 are removed from this analysis due to a mid-flight instrument operational change.

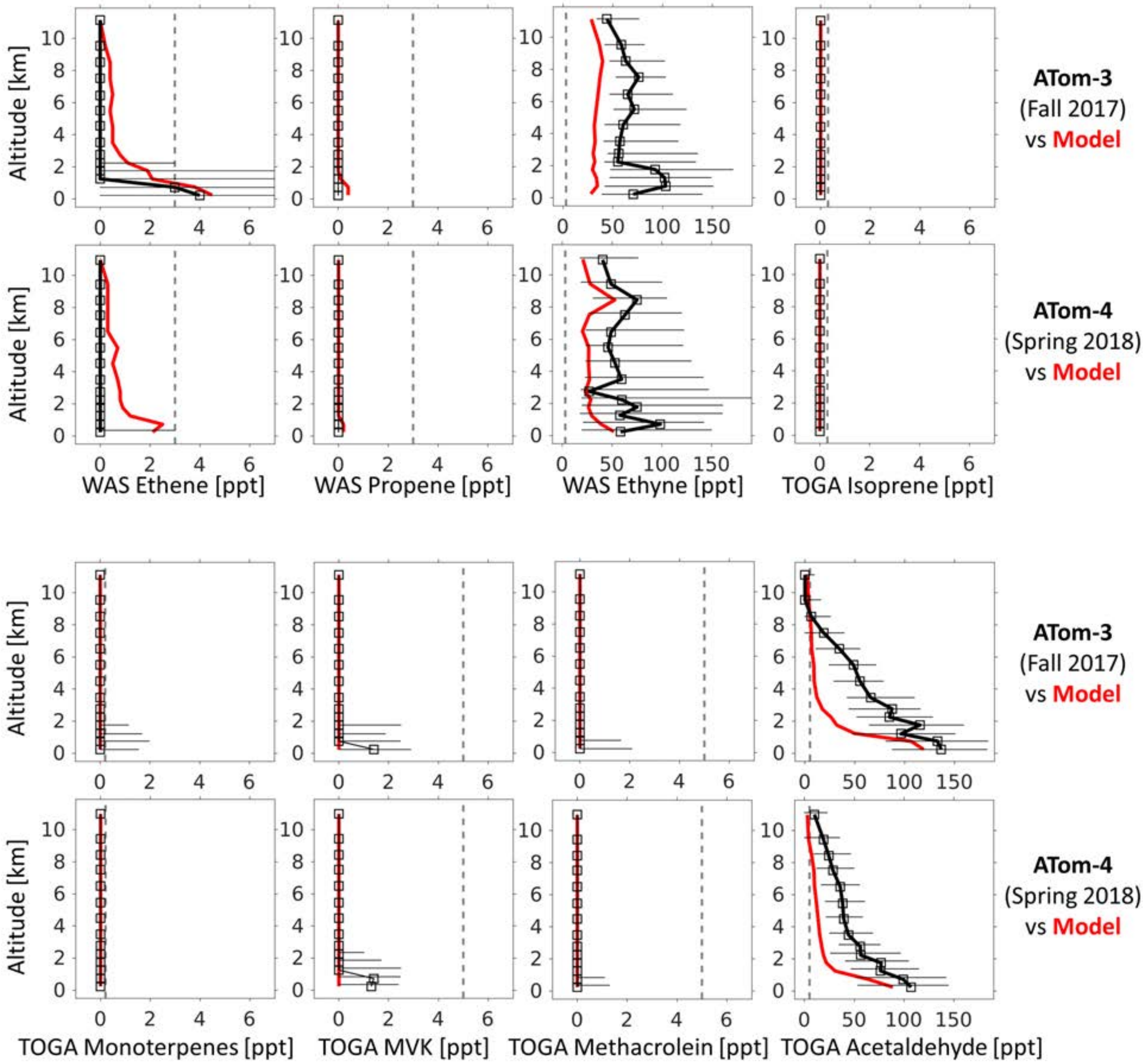


Figure S3. Campaign median vertical profiles for key HCOOH precursors as observed (black) and simulated by GEOS-Chem (red) during ATom-3 and ATom-4. Horizontal lines (black) indicate the measured interquartile range (IQR). Dashed vertical lines (gray) indicate the instrumental LOD (the TOGA LOD for individual monoterpenes varies between 0.1 and 20 ppt). Data flagged as below LOD are treated as zero when computing medians and IQRs.

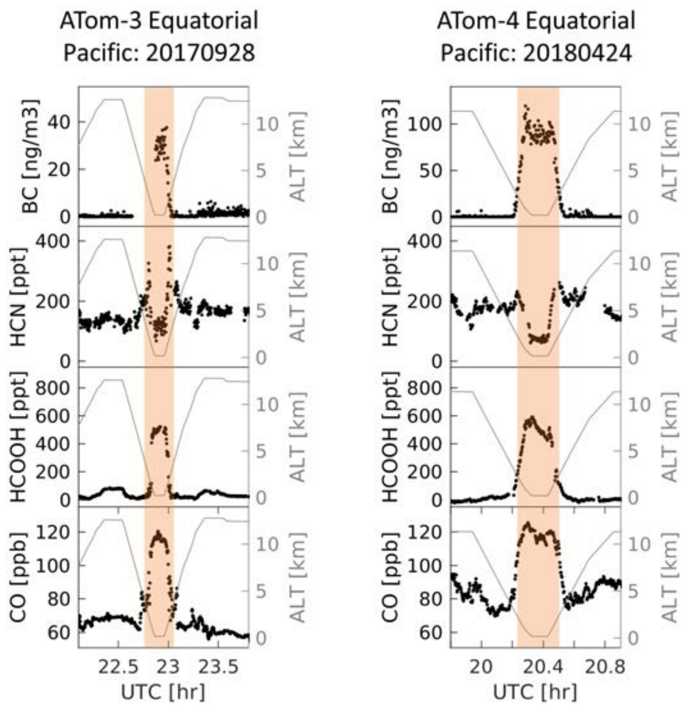


Figure S4. Example timeseries during DC-8 profiling showing MBL enhancements of HCOOH, along with black carbon (BC), hydrogen cyanide (HCN), CO, and altitude (ALT, grey, secondary y axis).

S2. Plume attribution

We identified 53 fire or mixed plume intercepts as described in the main text and summarized in Table S4. Some large scale plumes were penetrated more than once in consecutive ascending and descending maneuvers (shaded in Table S4) and we treat these separately in our analysis. The following sub-sections S2.1-2.4 detail the plume identification and source attribution process with plots showing tracer timeseries, NEMR regressions, trajectory cluster distribution of plume age, and additional evidence used for plume source attribution (e.g., ΔBC/ΔCO, flight-level HCOOH-CO scatter plots). Plume intercepts are numbered by time of sampling and are grouped into four categories. The plume intercept number and associated flight date are indicated in the title for each sub-section.

Plume identification — Flight timeseries: For each individual or group of plume intercepts being characterized we present the corresponding flight timeseries for black carbon (BC), hydrogen cyanide (HCN), HCOOH, CO, PALMS biomass burning particle number fraction (hereinafter "PALMS BB fraction")[17], NO_x ($NO+NO_2$), and altitude (ALT, grey, secondary y axis), with plume intercept windows shaded in orange and plume intercept numbers marked in the top (or second) panel. Dashed horizontal lines show thresholds employed in plume identification (BC: 100 ng/m^3, HCOOH: 1 ppb, CO: 175 ppb) and also indicate 50% fire influence for the PALMS BB fraction.

NEMR calculation — Plume HCOOH-CO regression: For each individual plume intercept, we show the reduced major axis regression (fit line in orange) of HCOOH and CO mixing ratios during the plume intercept (data points in blue) along with relevant statistical information. The NEMR of HCOOH relative to CO is obtained as the slope of this regression.

Plume age determination — Distribution of plume age in back trajectory clusters: We define the physical age of the plume as the time since most recent fire influence (t_f) according to the back trajectories. As described in **Section 2.3** of the main text, a cluster of 245 particles are released every minute along the flight track, generating up to 245 t_f estimates (typically less as some trajectories do not encounter any fire plumes). We then visualize the t_f distribution for each cluster within the plume intercept window. Based on these distributions, each plume intercept is assigned to one of seven age classes: <1 day, 0-5 days, 1-5 days, 1-10 days, 5-10 days, >10 days, or not determined (ND). For plumes in the first six groups, we illustrate below 1-2 representative distributions within the corresponding plume intercept window. The ND age group contains fire plumes without a well-defined age distribution or source regions (see below).

Plume source identification — Fire influence probability map: For each plume intercept below, a fire influence probability map is shown for the same trajectory cluster highlighted in the plume age distribution plot. Colored grid cells in these maps denote the fire source locations identified through the back trajectory analysis. Maps also indicate the flight tracks (in orange) and plume intercept locations (red). For plume intercepts with no clear fire influence in the back trajectories, we do not estimate an age or source region.

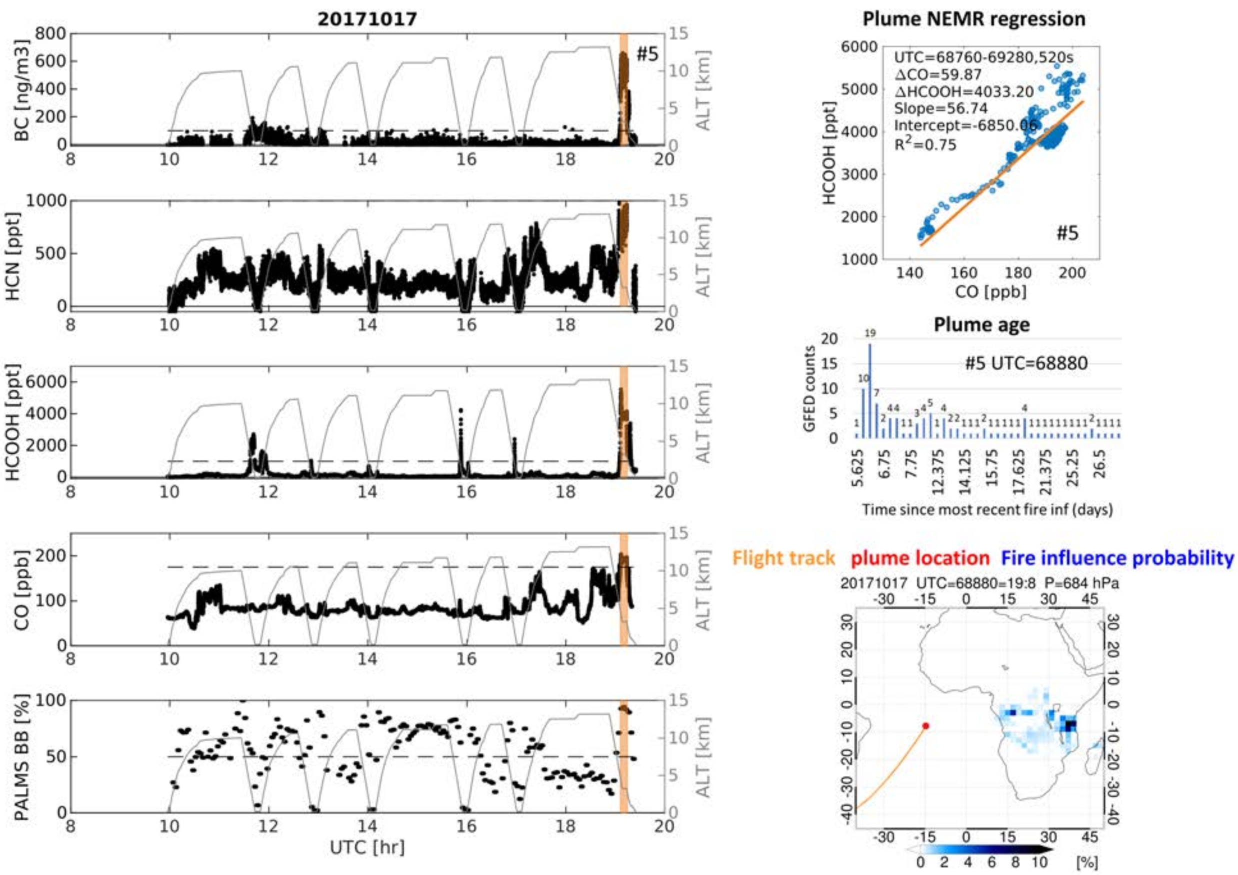
Table S4. Summary of the 53 identified plume intercepts. Consecutively shaded plume intercepts were identified in consecutive ascents and descents.

Plume intercept #	Flight	Time window (UTC in seconds)	HCOOH:CO NEMR (ppt/ppb CO)	Fire age category (days)	Fire source region	Source attribution	See details in SI page #
1	20170928	[52590, 53020]	63.9	> 10	N. America	Fire w/ potential anthro influence	p25
2	20171006	[99780, 100300]	11.2	< 1	Fiji	Fire w/ potential anthro influence	p26
3	20171017	[41870, 42000]	168.8	1-5	S. America	Fire w/ potential anthro influence	p16
4	20171017	[42570, 42720]	136.4	1-5	S. America	Fire w/ potential anthro influence	p16
5	20171017	[68760, 69280]	56.7	5-10	S. Africa	Fire	p11
6	20171019	[31200, 31480]	19.5	1-10	S. Africa	Fire	p12
7	20171019	[33430, 33750]	16.6	1-10	S. Africa	Fire	p12
8	20171019	[34320, 34600]	19.2	1-10	S. Africa	Fire	p12
9	20171019	[41200, 41400]	42.8	1-5	N. Africa	Fire	p12
10	20171019	[42370, 43600]	36.6	1-5	N. Africa	Fire	p12
11	20171019	[46670, 47260]	62.5	1-5	N. Africa	Fire	p12
12	20171020	[33700, 34130]	22.2	1-10	N. Africa	Fire	p13
13	20171020	[37740, 37980]	35.1	1-5	N. Africa	Fire	p13
14	20171020	[39500, 39620]	33.6	1-5	N. Africa	Fire	p13
15	20171020	[42700, 43040]	26.1	1-5	N. Africa	Fire	p13
16	20171020	[45240, 45520]	30.0	1-10	N. Africa	Fire	p13
17	20171025	[74040, 74240]	9.8	5-10	NE. Asia	Fire	p17
18	20171027	[68170, 68220]	4.2	ND	ND	Fire	p18
19	20171027	[70670, 70910]	3.4	ND	ND	Fire	p18
20	20180424	[53120, 53240]	60.7	1-5	NE. Asia + N. America	Fire w/ potential anthro influence	p27
21	20180424	[77400, 77660]	20.1	5-10	C. America	Fire	p19
22	20180424	[86760, 87130]	24.5	0-5	NE. Asia + N. America	Fire w/ potential anthro influence	p27
23	20180427	[57840, 58010]	19.1	< 1	N. America	Fire w/ potential anthro influence	p28
24	20180427	[71420, 71560]	58.9	ND	E. Asia	Fire	p20
25	20180427	[74460, 74900]	37.8	1-5	E. Asia	Fire	p20
26	20180427	[76560, 77040]	47.5	1-5	E. Asia	Fire	p20
27	20180427	[93700, 93800]	43.2	1-10	E. Asia	Fire	p20
28	20180429	[69560, 70600]	38.2	1-10	Eurasia	Fire	p21
29	20180429	[70880, 71180]	31.8	1-10	Eurasia	Fire	p21
30	20180429	[77180, 77360]	10.3	5-10	Eurasia	Fire	p21
31	20180429	[77420, 77500]	43.2	5-10	Eurasia	Fire	p21
32	20180429	[78420, 78640]	24.1	5-10	Eurasia	Fire	p21
33	20180503	[92840, 92960]	57.2	5-10	SE. Australia	Fire w/ potential anthro influence	p29
34	20180503	[93620, 93720]	70.8	5-10	SE. Australia	Fire w/ potential anthro influence	p29

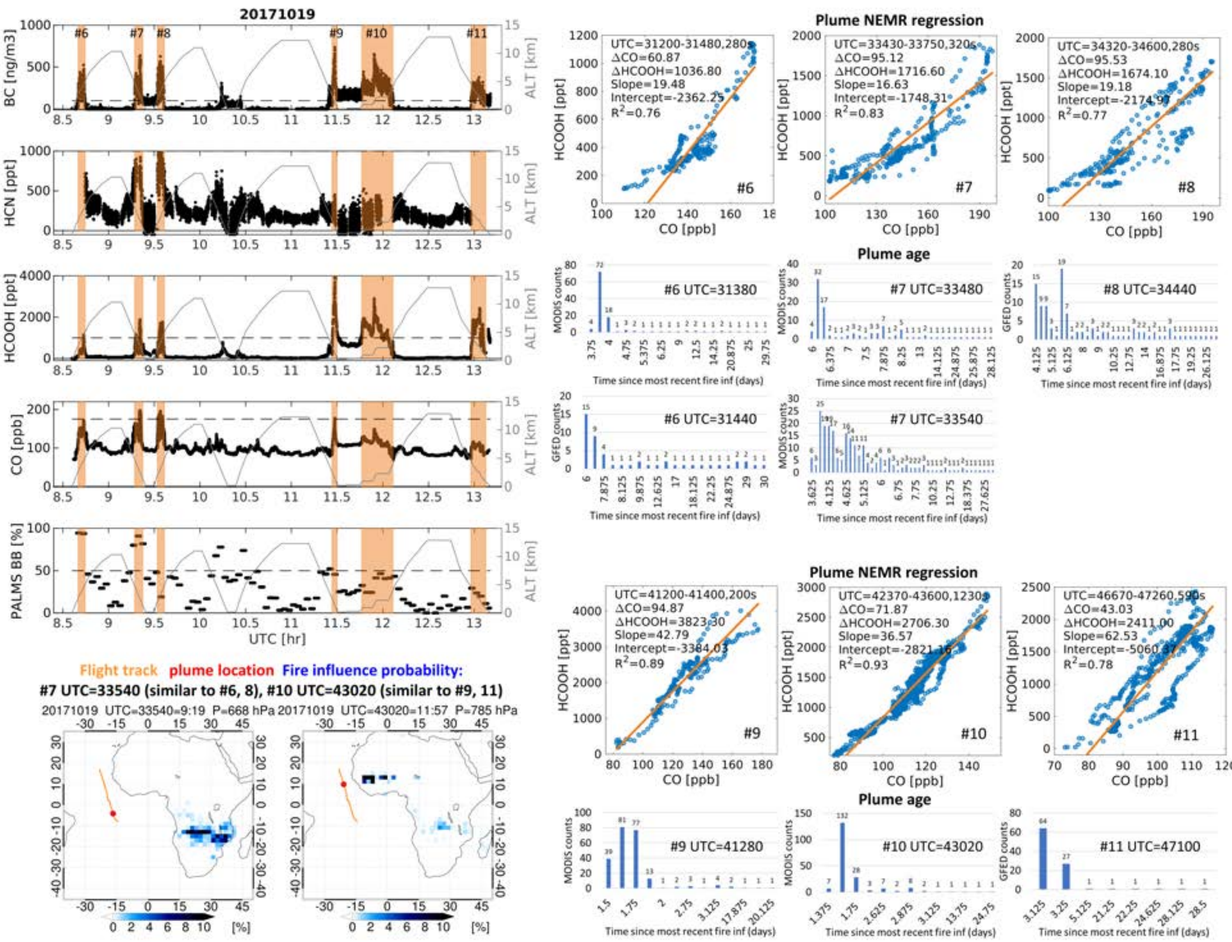
35	20180503	[95760, 95900]	49.3	1-10	SE. Australia	Fire w/ potential anthro influence	p28
36	20180503	[96200, 96760]	26.5	1-5	SE. Australia	Fire w/ potential anthro influence	p28
37	20180514	[48820, 49400]	47.9	1-10	N. Africa	Fire	p13
38	20180514	[49700, 50080]	43.0	1-10	N. Africa	Fire	p13
39	20180514	[53740, 54010]	139.4	1-10	N. Africa	Fire	p13
40	20180514	[57240, 57600]	112.3	1-10	N. Africa	Fire	p13
41	20180514	[57960, 58260]	129.3	1-10	N. Africa	Fire	p13
42	20180514	[60690, 60900]	97.8	ND	N. + C. Africa	Fire	p13
43	20180514	[61620, 61780]	96.0	ND	N. Africa	Fire	p13
44	20180517	[38400, 38510]	11.2	ND	ND	Fire	p21
45	20180517	[51090, 51210]	24.2	ND	ND	Fire	p21
46	20180517	[53170, 53240]	26.1	ND	ND	Fire	p21
47	20180519	[73440, 73620]	54.1	ND	Eurasia	Fire	p22
48	20180519	[74260, 75020]	31.0	ND	Eurasia	Fire	p22
49	20180521	[73160, 73280]	68.2	ND	Eurasia	Fire	p23
50	20180521	[81980, 82410]	87.4	0-5	N. America + E. Asia	Fire w/ potential anthro influence	p14
51	20180521	[82500, 82960]	55.3	< 1	N. America + E. Asia	Fire w/ potential anthro influence	p14
52	20180521	[83220, 83420]	241.4	ND	N. America + E. Asia	Fire w/ potential anthro influence	p14
53	20180521	[84200, 84620]	206.5	ND	N. America + E. Asia	Fire w/ potential anthro influence	p14

S2.1 African fire plume intercepts: #5 (20171017), #6-11 (20171019), #12-16 (20171020), #37-43 (20180514)

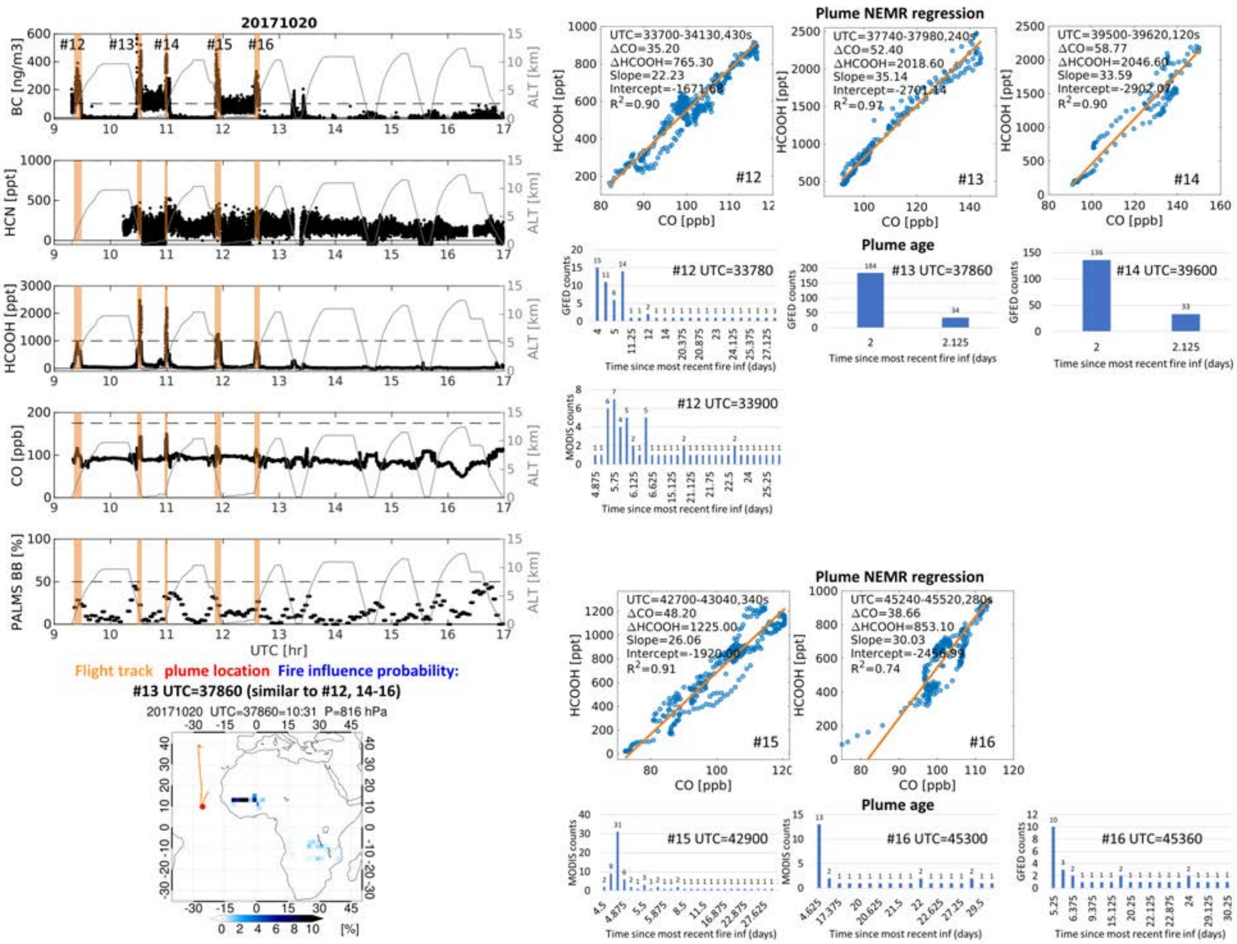
Plume intercept #5 (20171017): This plume was intercepted prior to landing in Ascension Island. Elevated values of the PALMS BB fraction and individual fire tracers reflect a dominant fire influence from southern Africa. Classification: *fire plume*.



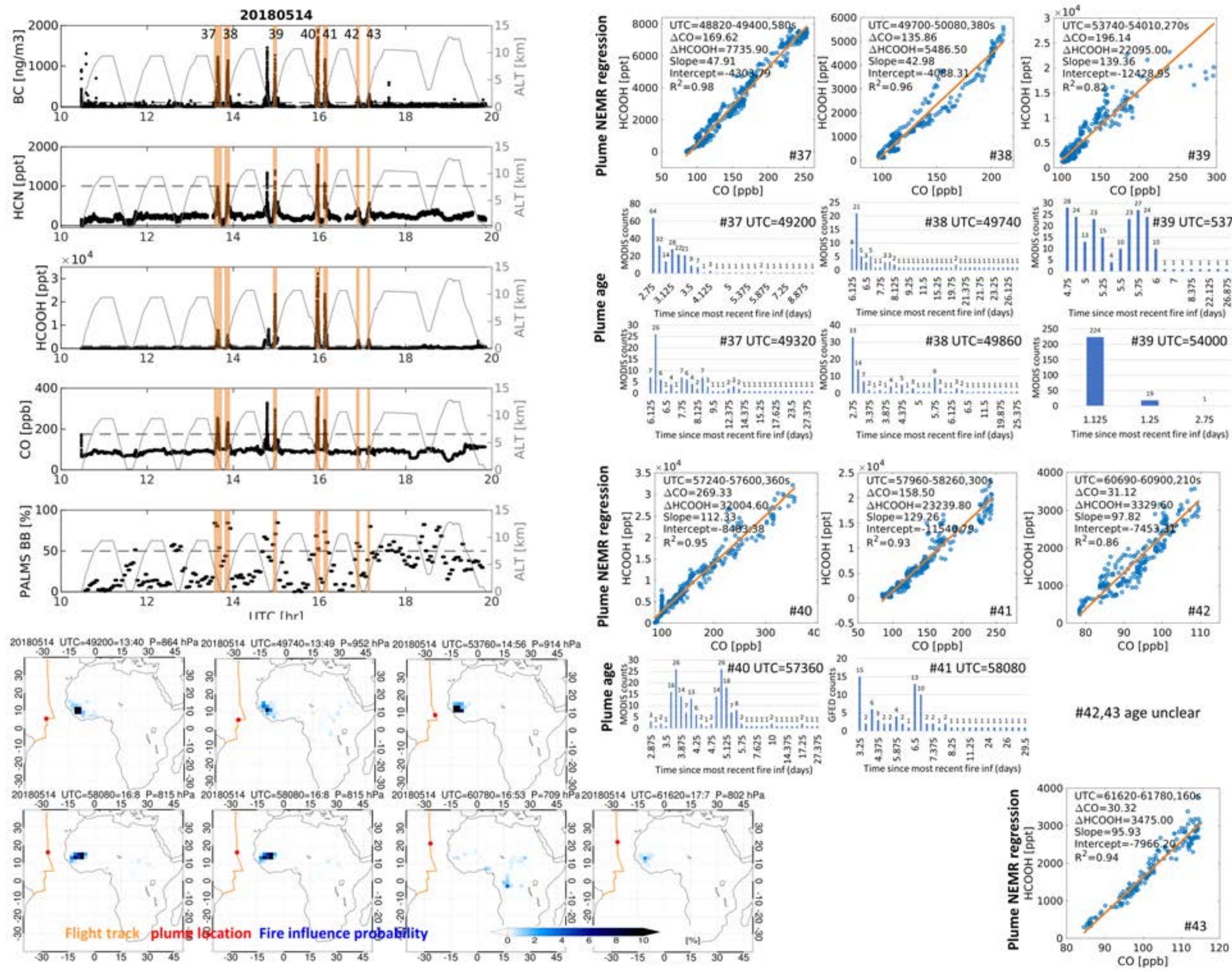
Plume intercepts #6-11 (20171019): These plumes were intercepted during the flight from Ascension Island to Cabo Verde. Back trajectories indicate that plume intercepts #6-8 originated in southern Africa, while plume intercepts #9-11 mainly originated in northern Africa. Although the PALMS BB fraction suggests low to moderate fire influence for plume intercepts #8-11, back trajectory analysis shows clear fire influence for plume intercepts #9-11 in particular. In addition, BC and CO are highly correlated in each plume intercept, with consistent ΔBC/ΔCO regression slopes (5.8-8.3 ng BC/m^3/ppb CO) that are within the range of BC:CO emission ratios for major African fire types reported by [18]—i.e., savanna: 8.1 ng BC/m^3/ppb CO, tropical forest: 5.1 ng BC/m^3/ppb CO, agricultural residue: 5.8 ng BC/m^3/ppb CO. Classification: *fire plumes*.



Plume intercepts #12-16 (20171020): These plumes were encountered near northwestern Africa. Despite only moderate enhancements in the PALMS BB fraction, back trajectories indicate clear fire influence (in particular for plume intercepts #13-14). The ΔBC/ΔCO regression slopes for these plume intercepts (5.8-9.1 ng BC/m^3/ppb CO) are close to the corresponding emission ratios for relevant fire types (5.1-8.1 ng BC/m^3/ppb CO; Andreae, 2019). Classification: *fire plumes*.



Plume intercepts #37-43 (20180514): Plumes intercepted by this flight are clearly identified as fire plumes from northwestern Africa, with consistent enhancements for the PALMS BB fraction and individual fire tracers. The ΔBC/ΔCO regression slopes for these plumes (6.6-8.9 ng BC/m^3/ppb CO) are close to the corresponding emission ratios for relevant fire types (5.1-8.1 ng BC/m^3/ppb CO; Andreae, 2019). Classification: *fire plumes*.

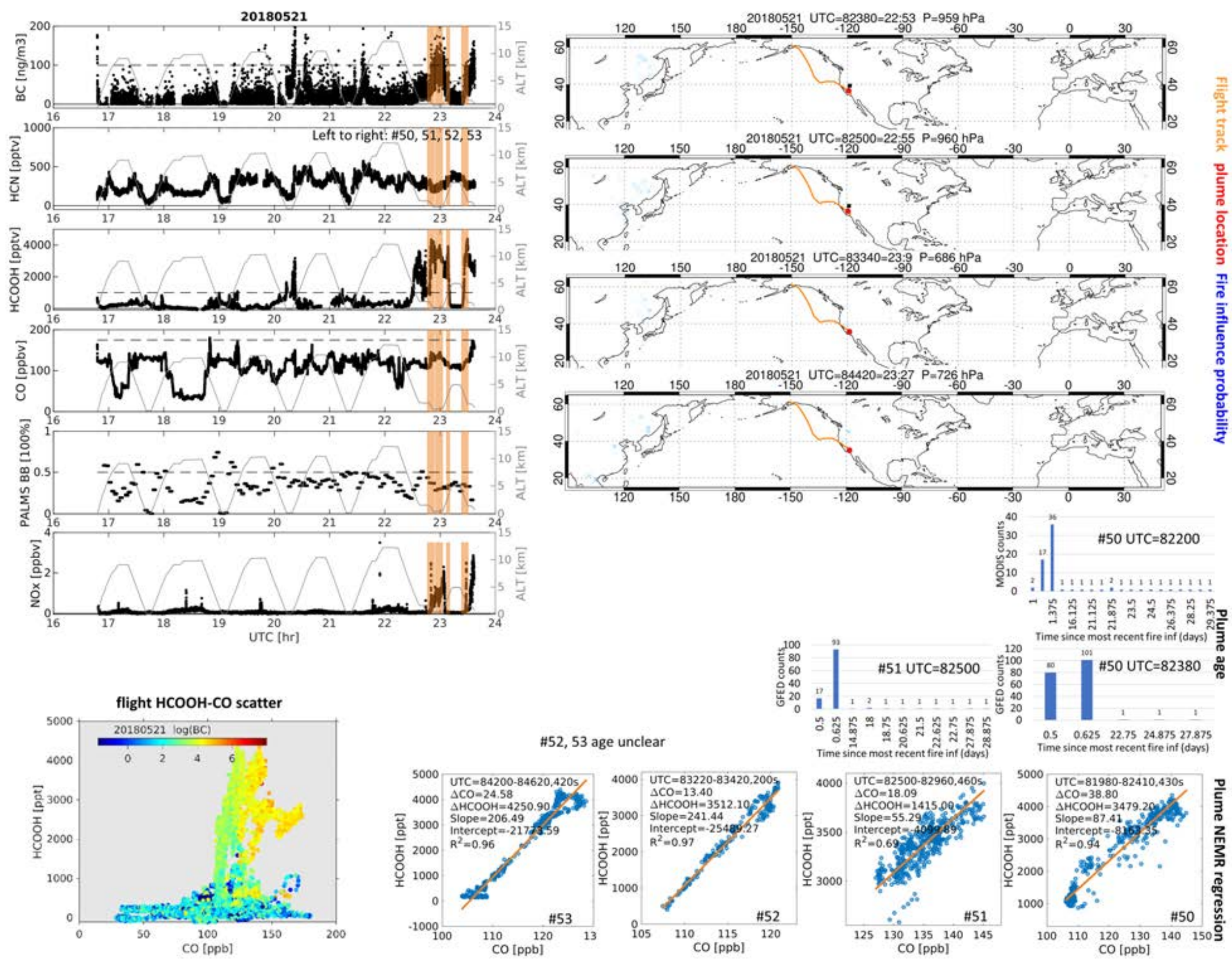


S2.2 Plumes with unusually high HCOOH:CO NEMRs: #50-53 (20180521), #3-4 (20171017)

Plume intercepts #50-53 (20180521): This is the final ATom flight. Prior to landing in CA, large HCOOH enhancements (reaching several ppb above background) are observed near and within the boundary layer. However, CO is only slightly elevated above background, resulting in quite high HCOOH:CO NEMR values of 55-241 ppt/ppb CO.

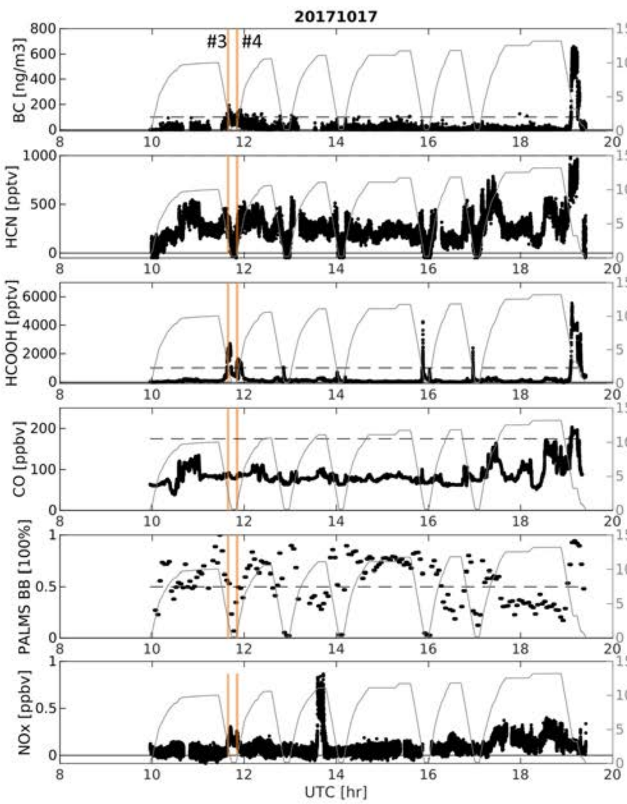
While the PALMS BB fraction suggests only low-to-moderate fire influence, back trajectories point to California fire sources for plume intercepts #50-51. Urban emissions may also contribute to the observed enhancements, but alone would only be expected to yield ΔHCOOH/ΔCO ratios of up to ~20 ppt/ppb CO[19]. Prior work by Hecobian et al. (2011)[20] pointed out the high oxidative potential in mixed plumes over California containing both urban and wildfire emissions, with accompanying elevated NEMRs for both NO_x and NO_y. The unusually high HCOOH NEMRs in these ATom plume intercepts could therefore reflect chemistry associated with fire-urban pollution interactions—specifically, VOC-rich biomass burning emissions mixing with high levels of urban NO_x[21].

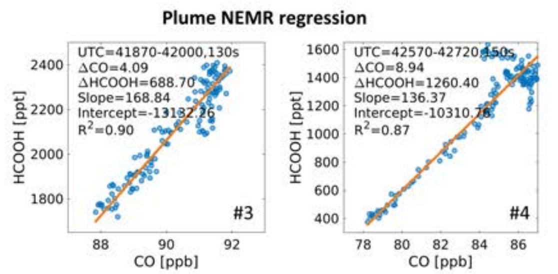
Classification: *fire plumes with potential urban/anthropogenic influence*

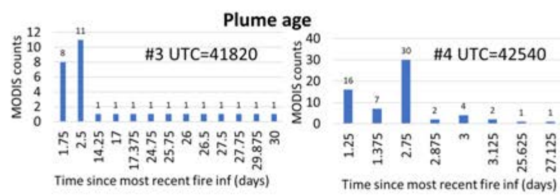


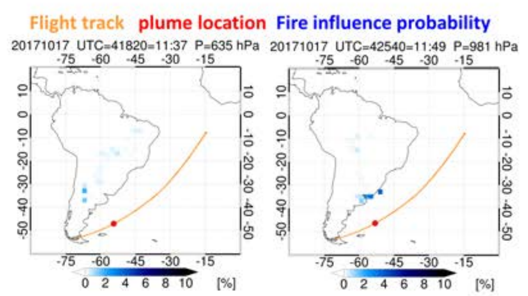
Plume intercepts #3-4 (20171017): These two plume intercepts were intercepted downwind of South America during the first vertical profile of the flight. The PALMS BB fraction is ambiguous as to the extent of fire influence. The encountered plume intercepts exhibit highly elevated methanol (> 6 ppb) and ethanol (> 1 ppb). The > 1ppb ethanol is very high for a remote ocean sample, and much higher than would be found in a fire plume. However, ethanol is used extensively as a fuel additive in parts of South America. The isopentane:nPentane ratio in this profiling maneuver ranges from 1 to 1.9, further implying anthropogenic influence.

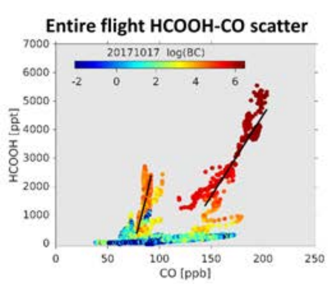
The ΔHCOOH/ΔCO NEMR values in these plume intercepts are unusually high (as also evident in the HCOOH-CO scatter plot for the entire flight). Classification: *fire plumes with potential urban/anthropogenic influence*.





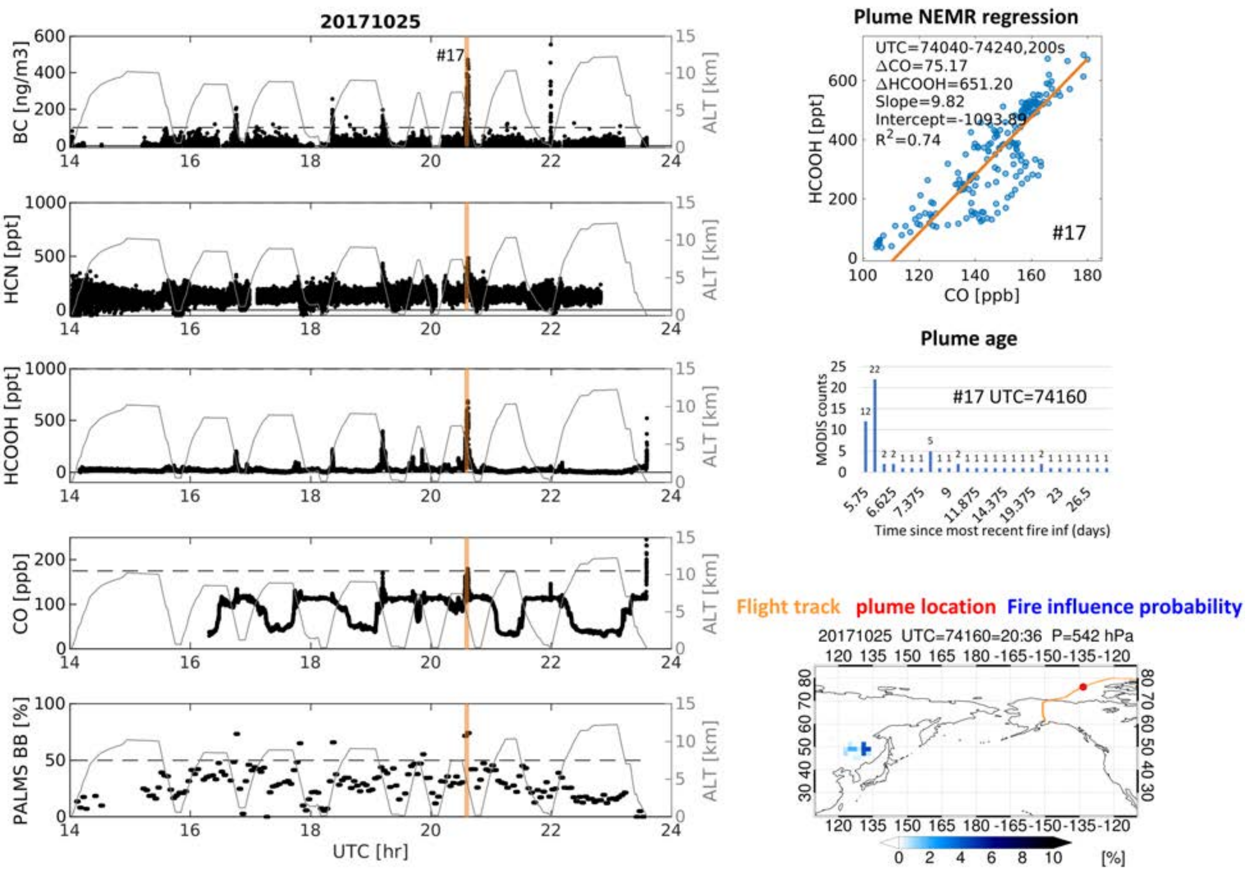




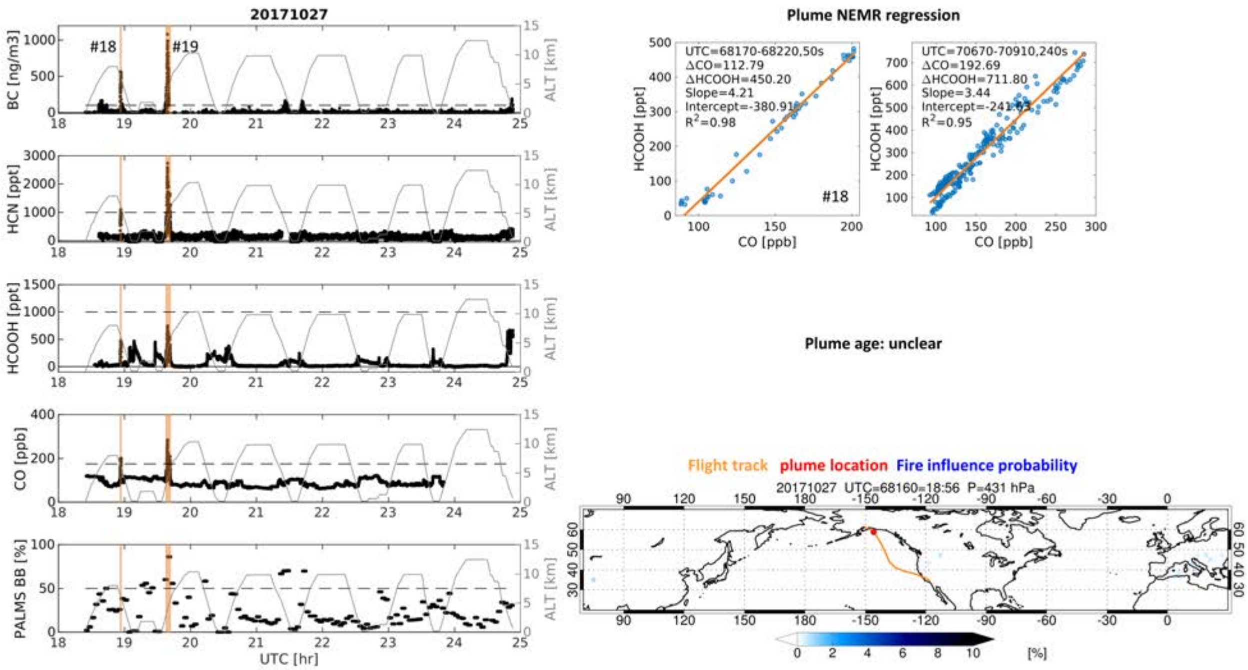


S2.3 Other fire plume intercepts: #17 (20171025), # 18-19 (20171027), #21 (20180424), #24-27 (20180427), #28-32 (20180429), #44-46 (20180517), #47-48 (20180519), #49 (20180521)

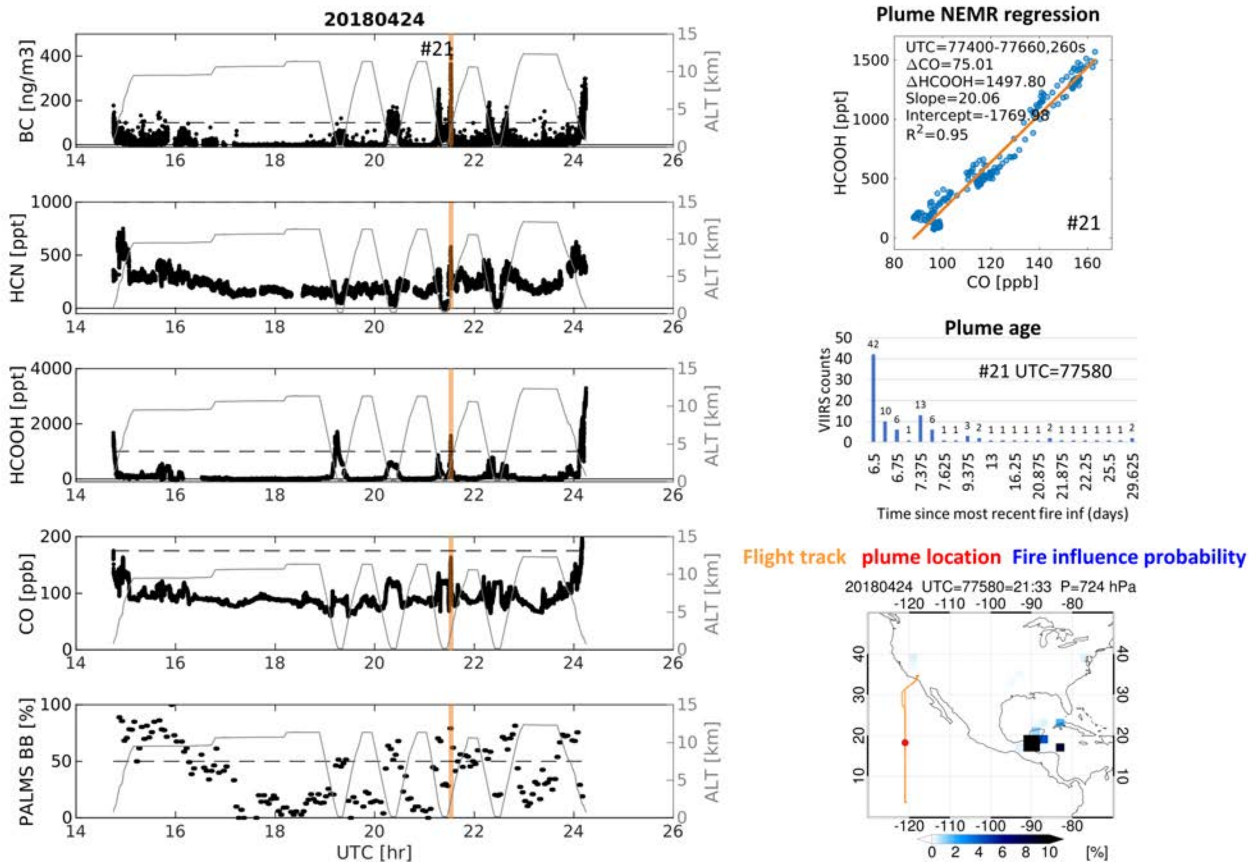
Plume intercept #17 (20171025): This plume was encountered at high altitude in the Arctic. Back trajectory analysis indicates fire influence from northeast Asia. Co-enhancements for fire tracers and the PALMS BB fraction suggest significant fire influence. Classification: *fire plume*.



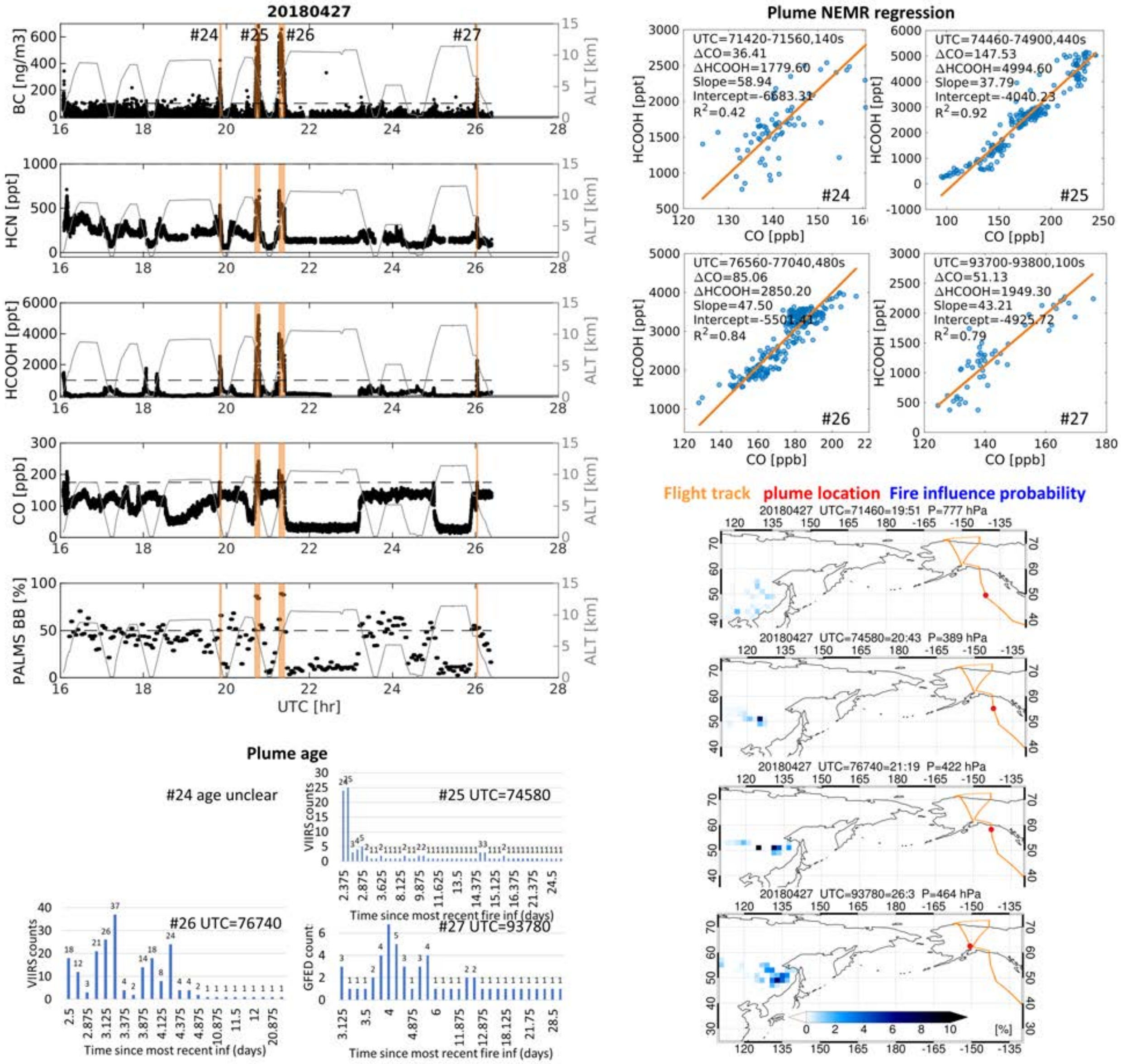
<u>Plume intercepts #18-19 (20171027)</u>: These two plume intercepts were intercepted at high altitude near Alaska. Despite a lack of detected fire signal in the back trajectories, the PALMS BB fraction suggests moderate to high fire influence. Elevated levels of HCN and other fire tracers are also observed. Plume intercept #19 exhibited the highest HCN mixing ratio but lowest HCOOH NEMR seen during ATom-3 and 4. Classification: *fire plumes*.



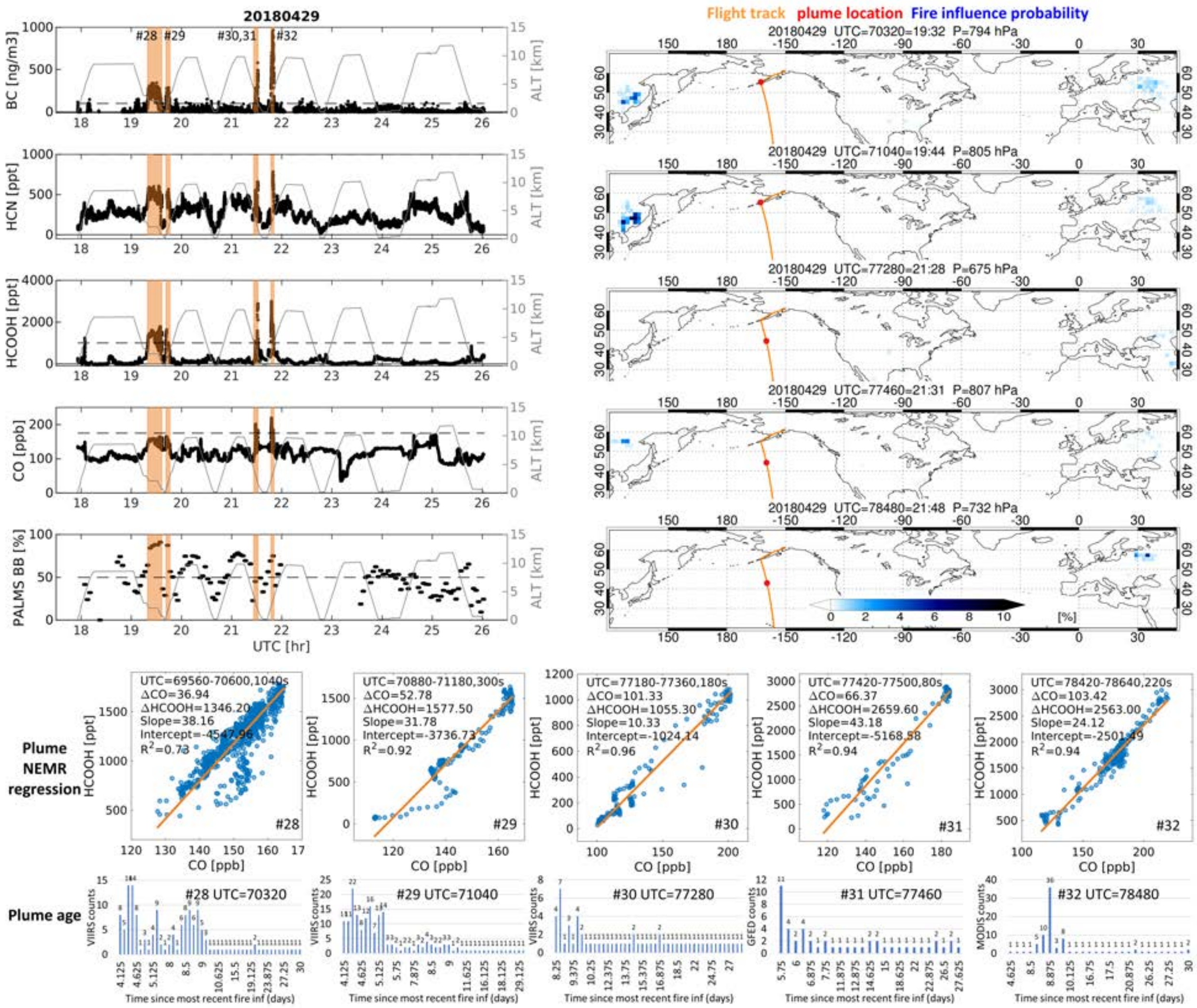
Plume intercept #21 (20180424): This plume was intercepted during the first equatorial flight of ATom-4. The PALMS BB fraction and many individual fire tracers point to significant fire influence. Back trajectories indicate a fire source in Central America. Classification: *fire plume.*



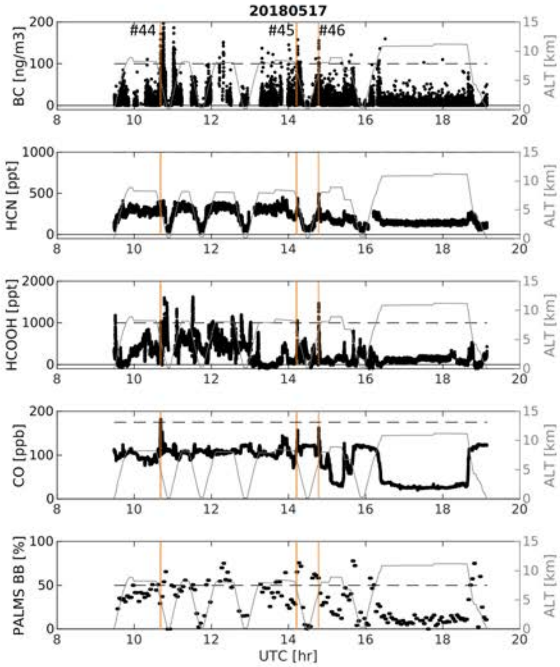
<u>Plume intercepts #24-27 (20180427)</u>: These plume intercepts were intercepted during the ATom-4 flight from California to Alaska. Co-enhancements for the PALMS BB fraction and individual fire tracers (e.g., HCN) suggest that these are fire plumes. Back trajectory analysis points to a fire source in northeast Asia. Classification: *fire plumes.*

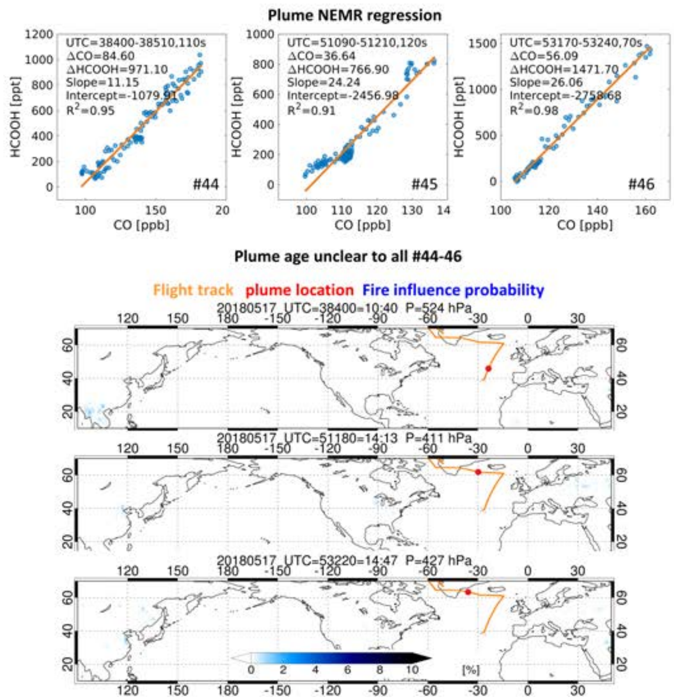


Plume intercepts #28-32 (20180429): These plumes were intercepted over the high-latitude Pacific. Moderate to large enhancements in the PALMS BB fraction and for individual fire tracers (e.g., HCN) indicate that these are fire plumes. Back trajectory analysis shows significant fire influence from northeast Asia for plume intercepts #28-29. Classification: *fire plumes.*

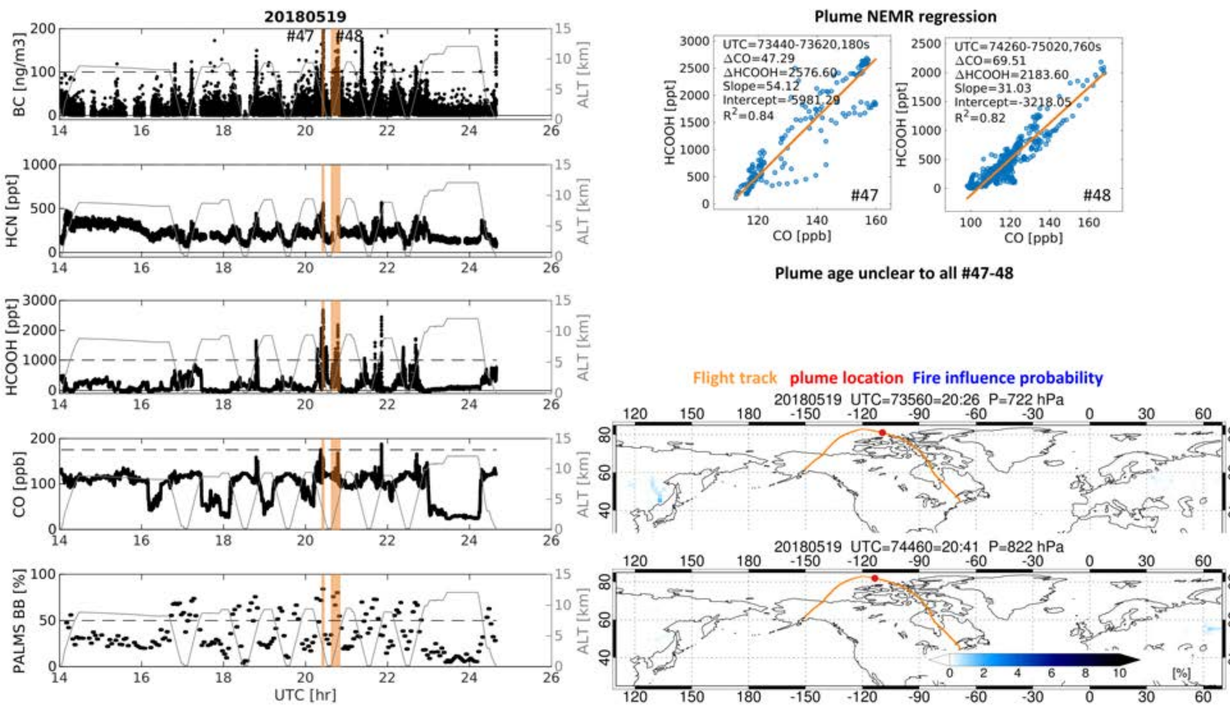


Plume intercepts #44-46 (20180517): These plumes were encountered at high altitude over the North Atlantic. Back trajectory analysis does not reveal any clear fire influence, but co-enhancements for fire tracers and the PALMS BB fraction suggest clear fire influence Classification: *fire plumes*.

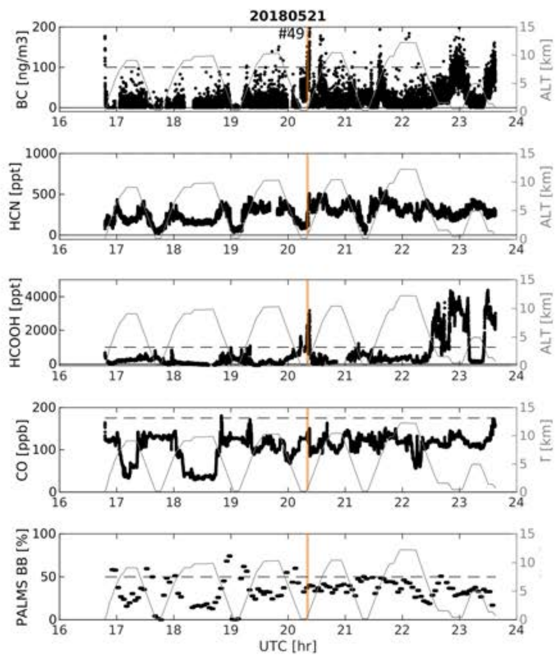


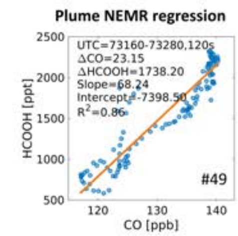


<u>Plume intercepts #47-48 (20180519)</u>: These plumes were encountered over the Arctic. The PALMS BB fraction and individual fire tracers (e.g., HCN) suggest clear fire influence, with back-trajectories pointing to a source region in northeast Asia. Classification: *fire plumes.*

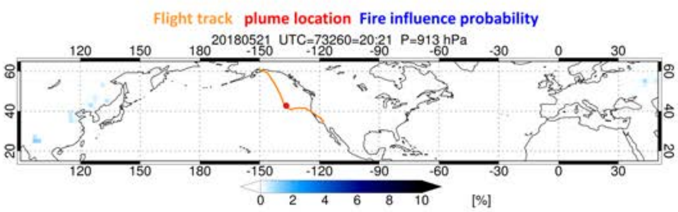


<u>Plume intercept #49 (20180521)</u>: The PALMS BB fraction suggests moderate fire influence, with an east Asian origin based on back trajectory analysis. Classification: *fire plume.*



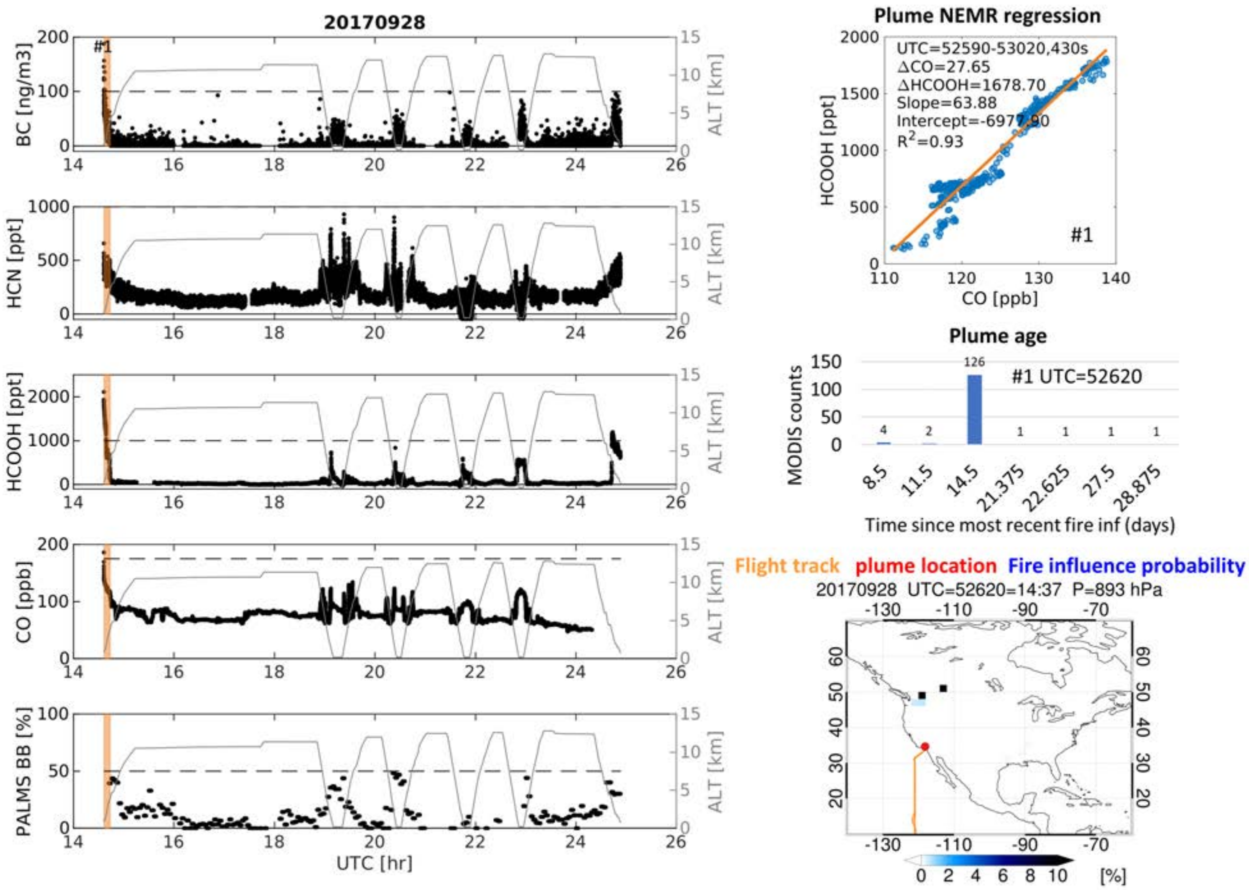


Plume age unclear

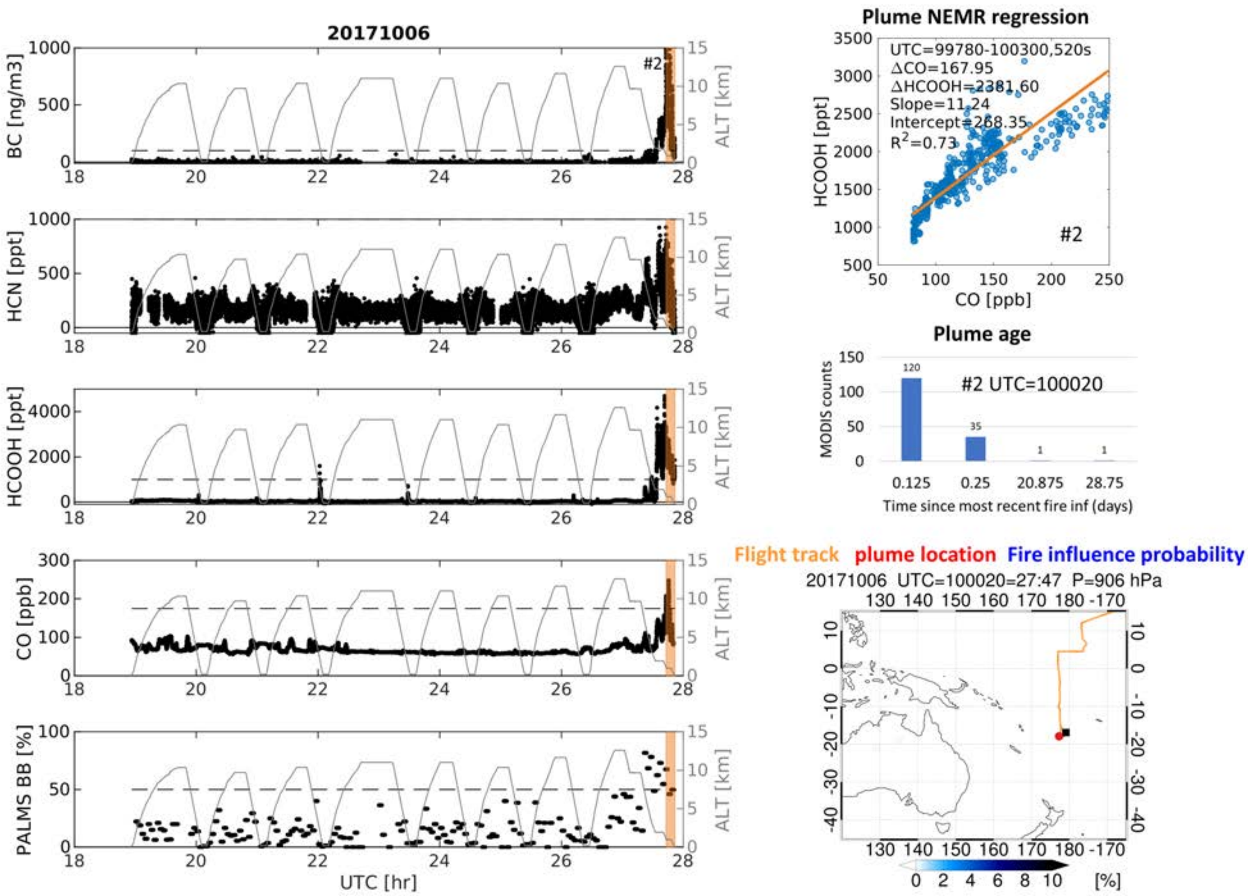


S2.4 Other fire plume intercepts with potential urban/anthropogenic influence: #1 (20170928), #2 (20171006), #20 and #22 (20180424), #23 (20180427), #33-36 (20180503)

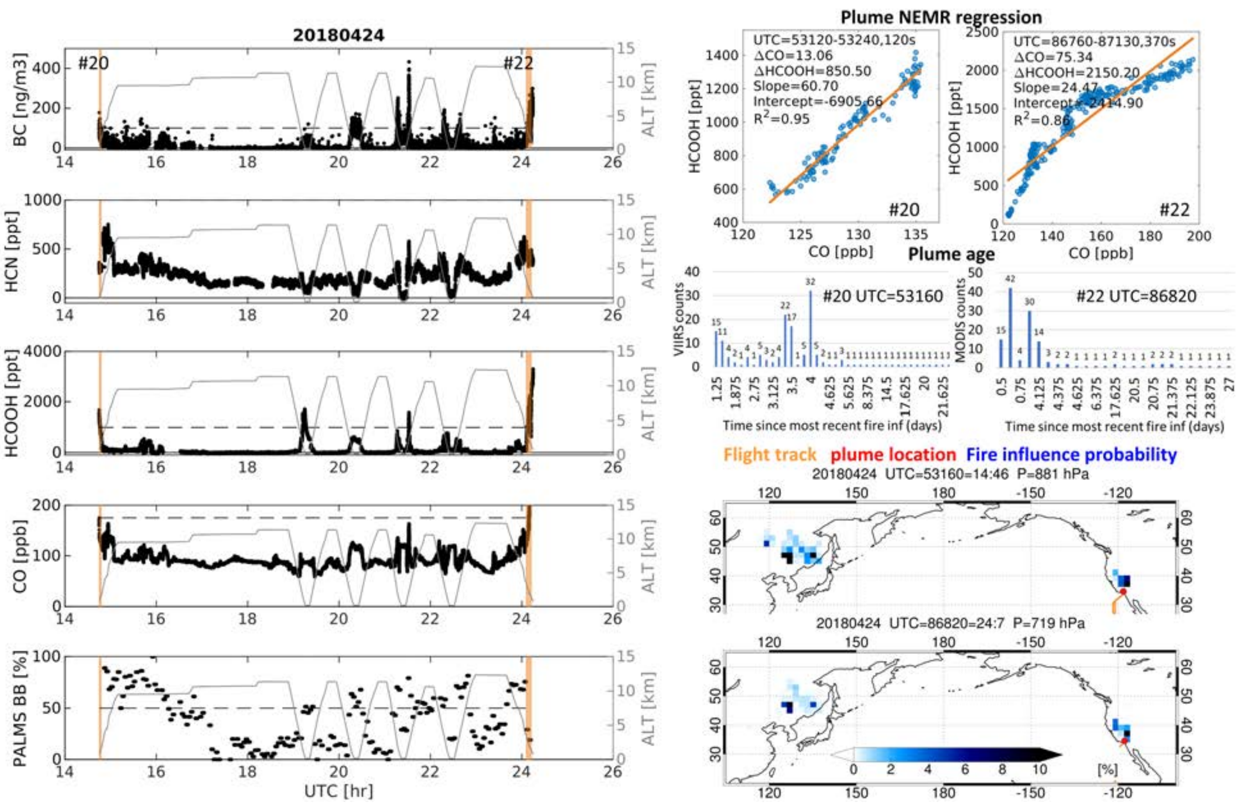
Plume intercept #1 (20170928): The PALMS BB fraction suggests moderate fire influence. Back trajectory analysis implicates fire sources in western Canada. The observed ΔBC/ΔCO NEMR (3.7 $ng/m^3/ppb$) is consistent with reported boreal forest emission ratios (e.g., 3.7 $ng/m^3/ppb$ CO; Andreae, 2019). The plume was sampled shortly after take-off and therefore may also contain an anthropogenic contribution. Classification: *fire plume with potential urban/anthropogenic influence.*



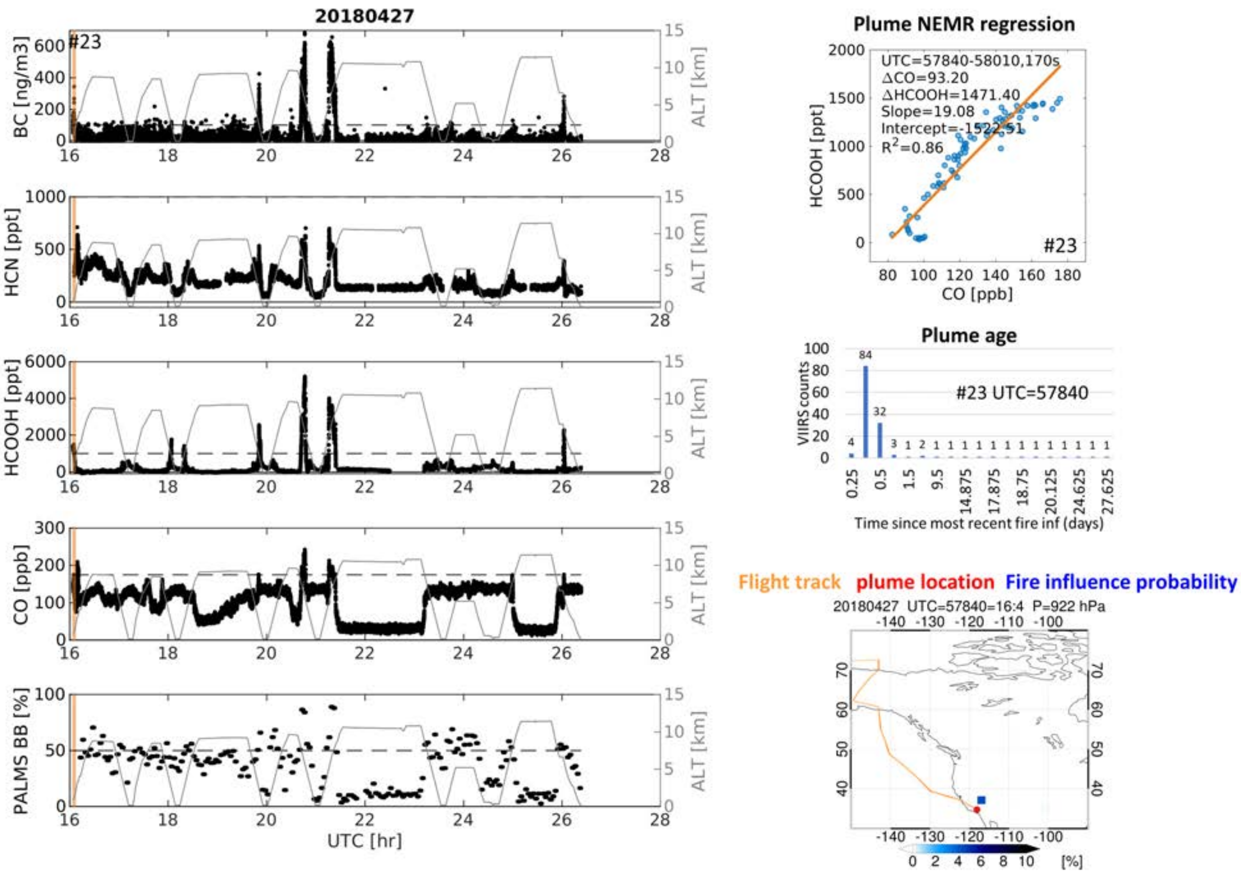
Plume intercept #2 (20171006): This plume was encountered prior to landing in Fiji. The PALMS BB fraction suggests moderate fire influence (~50%) in the selected time window, but strong fire influence more broadly during the final descent. Back trajectory analysis indicates significant local fire influence. Classification: *fire plume with potential urban/anthropogenic influence*.



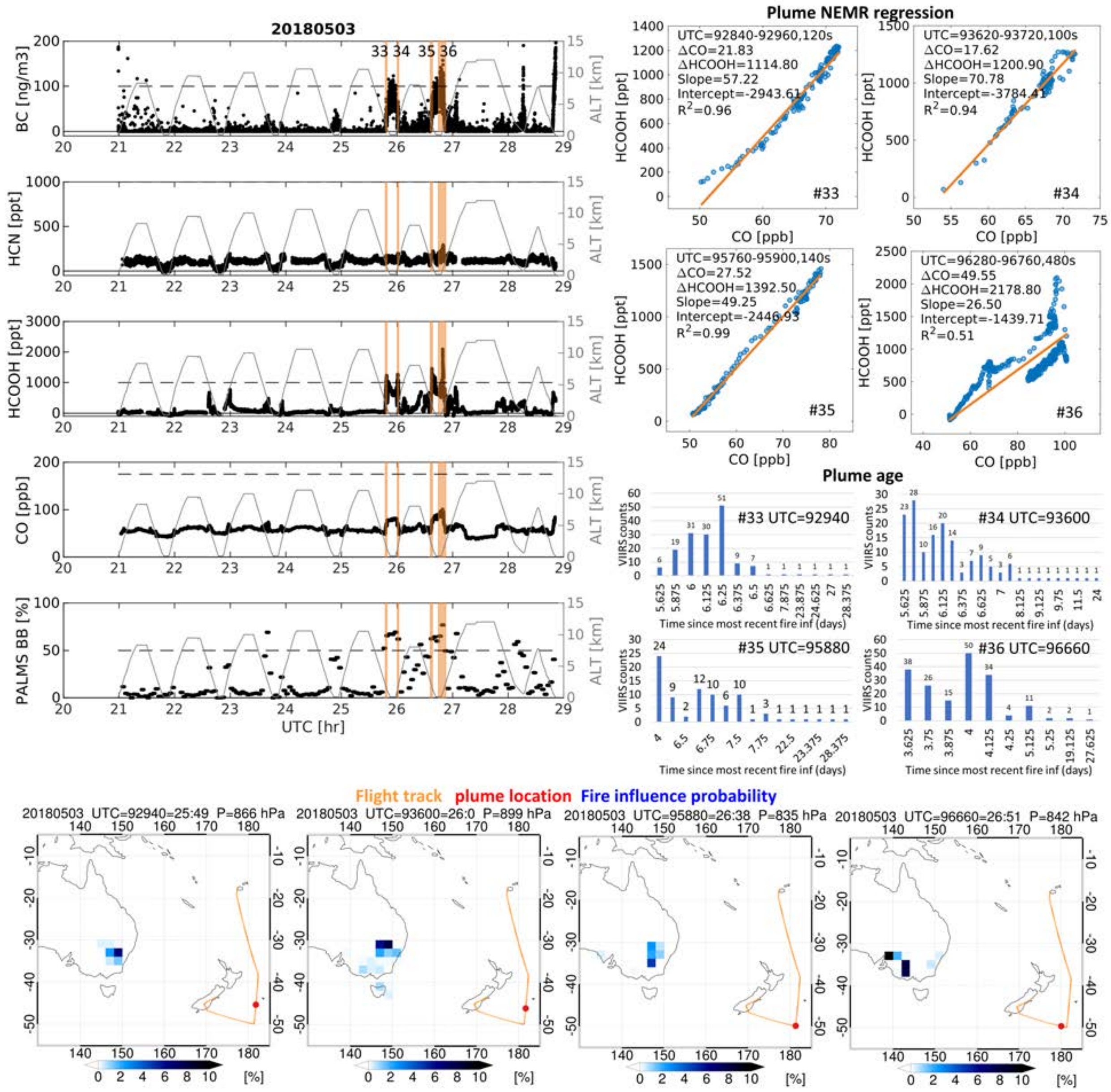
<u>Plume intercepts #20 and #22 (20180424)</u>: These two plumes were sampled during take-off and landing segments over California. The PALMS BB fraction suggests low to moderate fire influence for plume intercept #22. Back trajectory analysis suggests a combined influence from California and northeast Asia fires for both plumes. Classification: *fire plumes mixed with urban/anthropogenic pollution*.



Plume intercept #23 (20180427): This plume was sampled shortly after takeoff from California. Fire influence is indicated by enhancements in fire tracers such as HCN. Classification: *fire plume with potential urban/anthropogenic influence*.



Plume intercepts #33-36 (20180503): This flight encountered a large pollution layer with chemical tracers exhibiting enhancements throughout the MBL (e.g., BC) or above the MBL (e.g., HCOOH). Strong fire influence is implied by the PALMS BB fraction and by back trajectory analysis. This portion of the flight is also influenced by Asian/Australian anthropogenic pollution from the free troposphere through the MBL. Classification: *fire plumes with potential urban/anthropogenic influence.*



S3. ROC related tables and figures

Tables S5 lists the species included in the ROC pie charts shown in Figures 8 and S4. Pie charts are only generated for the 21 plume intercepts with sufficient TOGA sample coverage (i.e. ≥3 datapoints) to compute NEMR values by major axis regression. Of these, 9 plume intercepts also have sufficient WAS sample coverage (also ≥3 datapoints). Inclusion of the WAS species then introduces differences in measurement coverage across the 21 plume intercepts. **Figure 8** in the main text omits VOCs only measured by WAS for consistency across plumes; **Figure S5** includes such species when available.

The resulting 21 plume intercepts (those featuring **WAS coverage in bold**) are the following: 20170928 #1, 20171006 #2, 20171017 #5, 20171019 #6, **20171019 #10**, **20171019 #11**, 20171020 #12, 20171020 #15, 20171020 #16, **20180424 #22**, 20180427 #25, 20180427 #26, **20180429 #28**, **20180503 #36**, **20180514 #37**, 20180514 #38, **20180514 #40**, 20180514 #41, **20180519 #48**, 20180521 #50, **20180521 #51**.

For a given species-plume combination, we either list the derived NEMR (in ppt C/ppb CO, correlation coefficient in parentheses), or indicate why a NEMR is not calculated (text in grey). Instrument names are abbreviated as T (TOGA), W (WAS), F (ISAF), CC (CF_3O^- CIMS), IC (Iodide CIMS), and H (HR-AMS). Below are a few examples to demonstrate the notation:

- **T n/a:** Species not reported by TOGA. Likewise for W n/a and F n/a.
- **W LLOD:** Species reported by WAS, but does not have enough above-detection-limit values to derive an NEMR by major axis regression. Likewise for T LLOD.
- **T NaN:** Species reported by TOGA, but lacks sufficient non-missing data values to derive an NEMR by major axis regression. Likewise for W NaN, F NaN, CC NaN, and H NaN. This mainly occurs for organic nitrate and halogen species that are not major ROC contributors in these plumes.
- **CC NEMR<0:** Species reported by CF_3O^- CIMS with enough valid datapoints for NEMR regression. However, the species-CO correlation is negative in the plume intercept. Likewise for T NEMR<0 and W NEMR<0.
- **H r<0.3:** Species reported by HR-AMS with enough valid datapoints for NEMR regression. However, the in-plume species-CO correlation is low (r<0.3). Likewise for T r<0.3, W r<0.3, F r<0.3, CC r<0.3.
- **W <3 data:** Species reported by WAS but the plume window is too short to yield enough datapoints for NEMR regression.

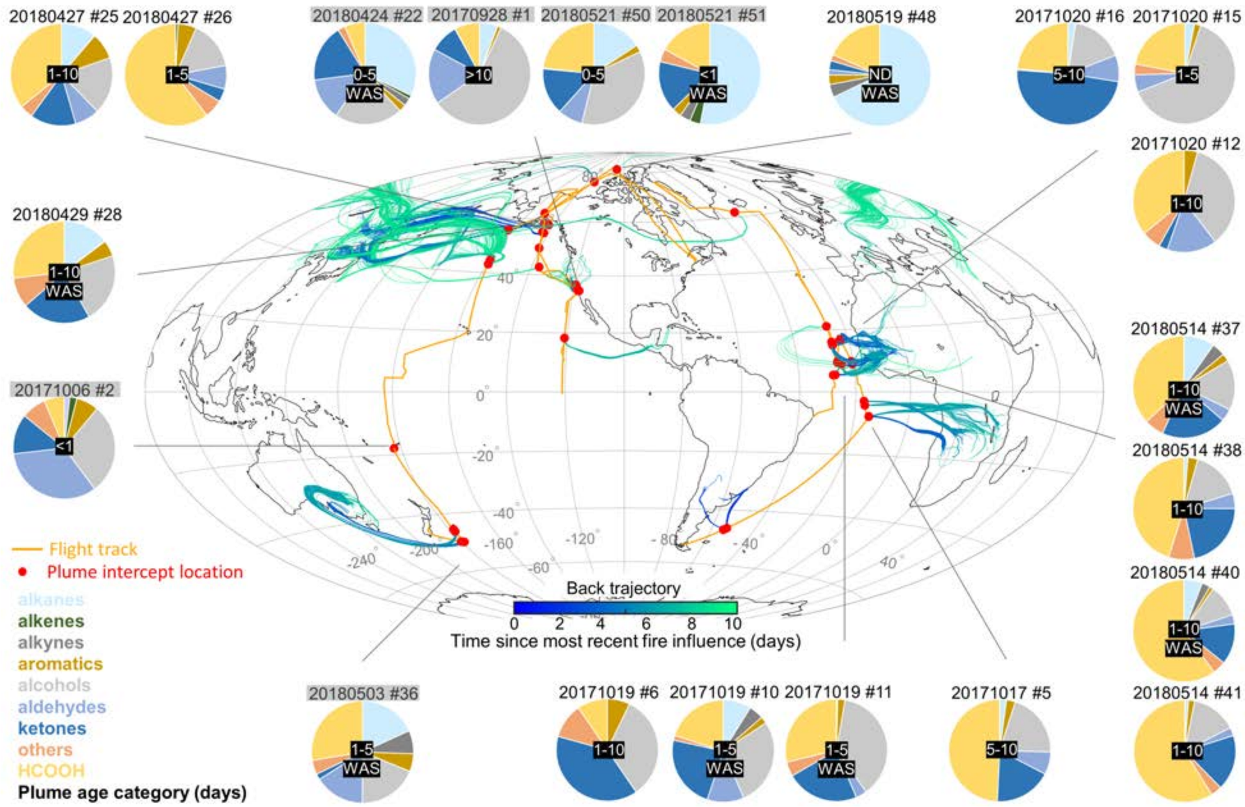


Figure S5. Same as **Figure 8**, but including those species measured only by WAS for the 9 plume intercepts with sufficient coverage for NEMR regression (**Sect. S2.4**).

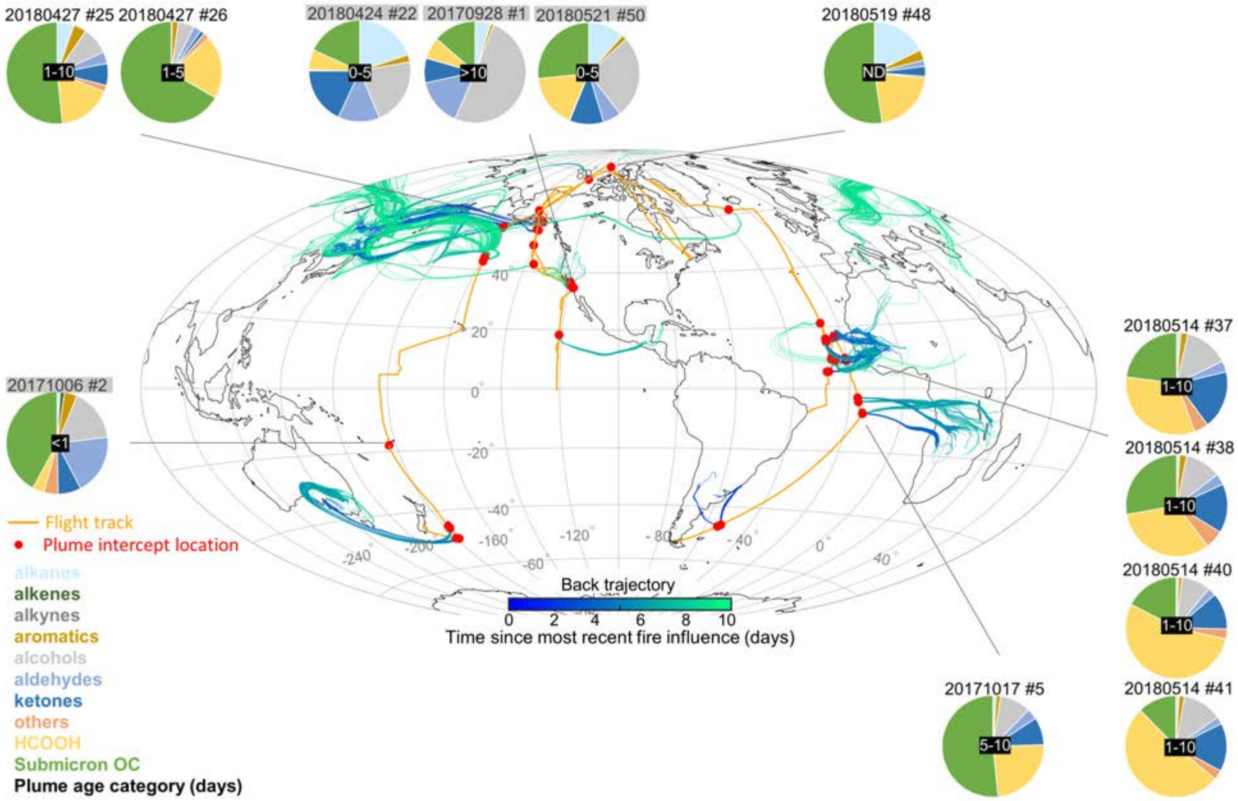


Figure S6. Same as **Figure 8**, but including submicron aerosol OC.

Table S5. ROC speciation in Figures 8, S4 and S5 (ppt C/ppb CO)

Classes	Plume intercept # / Species	1	2	5	6	10	11	12
Alkanes	Ethane	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 10.63 (0.77)	T n/a, W NEMR<0	T n/a, W <3 data
	Propane	T 23.33 (0.98)	T 2.66 (0.94)	T 2.29 (0.82)	T LLOD, W <3 data	T NEMR<0, W 2.28 (0.83)	T NEMR<0, W NEMR<0	T NEMR<0, W <3 data
	iButane	T 4.55 (0.95)	T NEMR<0, W <3 data	T 0.05 (0.88)	T LLOD, W <3 data	T 0.79 (0.25)	T NEMR<0, W NEMR<0	T NEMR<0, W <3 data
	nButane	T 4.90 (0.81)	T NEMR<0, W <3 data	T 0.15 (0.85)	T LLOD, W <3 data	T 1.47 (0.39)	T r<0.3, W NEMR<0	T NEMR<0, W <3 data
	Isopentane	T 5.82 (0.99)	T r<0.3, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T 0.44 (0.62)	T 0.92 (0.34)	T LLOD, W <3 data
	nPentane	T 4.30 (0.99)	T 0.17 (0.55)	T LLOD, W <3 data	T LLOD, W <3 data	T 0.40 (0.41)	T LLOD, W LLOD	T LLOD, W <3 data
	nHexane	T 0.87 (1.00)	T 0.11 (0.96)	T LLOD, W <3 data	T LLOD, W <3 data	T 0.12 (0.62)	T LLOD, W LLOD	T LLOD, W <3 data
	nHeptane	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
	2Methylpentane	T 1.42 (0.85)	T 0.10 (0.58)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
	3Methylpentane	T LLOD, W <3 data	T 0.04 (0.82)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
	224Trimethylpentane	T 1.09 (0.90)	T NEMR<0, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Alkenes	Ethene	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W LLOD	T n/a, W <3 data
	Propene	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W LLOD	T n/a, W LLOD	T n/a, W <3 data
	Isobutene+1Butene	T 1.05 (0.36)	T 1.38 (0.85)	T LLOD, W n/a	T LLOD, W n/a	T 0.10 (0.33)	T LLOD, W n/a	T LLOD, W n/a
	Isoprene	T LLOD, W <3 data	T r<0.3, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
	aPinene	T LLOD, W <3 data	T 2.63 (0.99)	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Tricyclene	T NEMR<0, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Camphene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	bPineneMycene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	LimoneneD3Carene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Alkynes	Ethyne	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 8.41 (0.95)	T n/a, W 9.66 (0.59)	T n/a, W <3 data
Aromatics	Benzene	T 3.90 (0.97)	T 8.82 (1.00)	T 3.23 (1.00)	T 13.87 (0.59)	T 3.73 (0.95)	T 4.80 (1.00)	T 2.67 (0.94)
	Toluene	T 4.19 (0.95)	T 2.75 (0.99)	T LLOD, W <3 data	T 0.27 (0.89)	T 0.29 (0.81)	T 0.21 (0.73)	T r<0.3, W <3 data
	Ethylbenzene	T LLOD, W <3 data	T 0.36 (0.95)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
	mpXylene	T 1.13 (0.85)	T 0.36 (0.94)	T LLOD, W <3 data	T 0.47 (0.75)	T LLOD, W LLOD	T LLOD, W LLOD	T NEMR<0, W <3 data
	oXylene	T LLOD, W <3 data	T 0.13 (0.97)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
Alcohols	CH3OH	T 100.69 (0.90)	T 24.93 (1.00)	T 40.57 (0.78)	T 67.73 (0.30)	T 40.28 (0.83)	T 81.34 (0.79)	T 21.85 (0.78)
	C2H5OH	T 375.55 (0.89)	T 25.55 (0.10)	T 2.96 (0.30)	T LLOD, W n/a	T 10.34 (0.64)	T r<0.3, W n/a	T r<0.3, W n/a
Aldehydes	HCHO	F 72.31 (0.84)	F 26.76 (0.90)	F 5.56 (0.48)	F r<0.3, T NEMR<0	F 11.58 (0.78)	F 7.67 (0.38)	F 9.24 (0.31)
	CH3CHO	T 61.38 (0.90)	T 18.53 (0.99)	T 2.66 (0.50)	T NEMR<0, W n/a	T 8.36 (0.43)	T NEMR<0, W n/a	T r<0.3, W n/a
	Propanal	T LLOD, W n/a	T 2.87 (0.88)	T LLOD, W n/a	T LLOD, W n/a	T 0.80 (0.90)	T LLOD, W n/a	T NaN, W n/a
	Butanal	T 4.86 (0.99)	T 0.37 (0.78)	T NEMR<0, W n/a	T NEMR<0, W n/a	T 0.57 (0.28)	T NEMR<0, W n/a	T 0.59 (0.40)
	Acrolein	T LLOD, W n/a	T 6.00 (0.97)	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Methacrolein	T 4.17 (0.91)	T 3.99 (0.95)	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Ketones	Acetone	T 51.81 (1.00)	T 11.13 (0.94)	T 19.44 (0.82)	T 79.06 (0.86)	T 37.59 (0.88)	T 44.41 (0.68)	T NaN, W n/a
	MEK	T 11.60 (1.00)	T 1.63 (1.00)	T 1.47 (0.95)	T r<0.3, W n/a	T 4.99 (0.74)	T 5.65 (0.51)	T 1.71 (0.44)
	MVK	T 10.79 (0.88)	T 9.49 (0.97)	T LLOD, W n/a	T LLOD, W n/a	T r<0.3, W n/a	T LLOD, W n/a	T LLOD, W n/a
Other OVOCs	MTBE	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Other VOCs	OCS	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W 2.4 (0.63)	T n/a, W <3 data
	DMS	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD	T LLOD, W <3 data
	HCN	CC r<0.3, T r<0.3	CC 8.15 (0.59)	CC r<0.3, T r<0.3	CC 15.21 (0.42)	CC r<0.3, T r<0.3	CC NEMR<0, T r<0.3	CC NaN, T 2.12 (0.70)
	CH3CN	T 1.32 (0.93)	T 3.49 (0.96)	T r<0.3, W n/a	T 6.57 (0.65)	T 1.99 (0.50)	T 4.33 (0.94)	T 1.66 (0.95)

Table S5. Continued

Classes	Plume intercept # / Species	1	2	5	6	10	11	12
Organic nitrates	Methyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.03 (0.57)	T n/a, W 0.02 (0.37)	T n/a, W <3 data
	Ethyl nitrate	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W 0.03 (0.94)	T NaN, W r<0.3	T NaN, W <3 data
	Isopropyl nitrate	T NEMR<0, W <3 data	T LLOD, W <3 data	T r<0.3, W <3 data	T LLOD, W <3 data	T 0.09 (0.50)	T r<0.3, W NEMR<0	T LLOD, W <3 data
	nPropyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.01 (0.98)	T n/a, W NEMR<0	T n/a, W <3 data
	2Butyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.11 (0.68)	T n/a, W NEMR<0	T n/a, W <3 data
	3Pentyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/aW 0.07 (0.99)	T n/a, W 0.05 (0.87)	T n/a, W <3 data
	2Pentyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.09 (0.98)	T n/a, W 0.04 (0.85)	T n/a, W <3 data
	3Methyl2butyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.05 (1.00)	T n/a, W 0.04 (0.66)	T n/a, W <3 data
Halogens	CFC12	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.21 (0.12)	T n/a, W 0.58 (0.92)	T n/a, W <3 data
	CFC11	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W 0.20 (0.67)	T n/a, W <3 data
	CFC113	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W 0.02 (0.88)	T NaN, W 0.13 (0.53)	T NaN, W <3 data
	CFC114	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.01 (0.59)	T n/a, W 0.04 (0.47)	T n/a, W <3 data
	HFC152a	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W NaN	T n/a, W <3 data
	HFC134a	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.10 (0.83)	T n/a, W 0.45 (0.96)	T n/a, W <3 data
	HFC365mfc	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W NaN	T n/a, W <3 data
	HCFC124	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W NaN	T n/a, W <3 data
	HCFC22	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.03 (0.41)	T n/a, W 0.40 (0.91)	T n/a, W <3 data
	HCFC142b	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.05 (0.64)	T n/a, W r<0.3	T n/a, W <3 data
	HCFC141b	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.03 (0.53)	T n/a, W 0.07 (0.64)	T n/a, W <3 data
	H1301	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W 0.01 (0.41)	T n/a, W <3 data
	H2402	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W 0.00 (0.68)	T n/a, W <3 data
	H1211	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W 0.01 (0.56)	T n/a, W <3 data
	CH3CCl3	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.00 (0.54)	T n/a, W r<0.3	T n/a, W <3 data
	CCl4	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W 0.07 (0.99)	T n/a, W <3 data
	CHCl3	T NEMR<0, W <3 data	T NEMR<0, W <3 data	T r<0.3, W <3 data	T 0.21 (0.65)	T 0.03 (0.34)	T r<0.3, W NEMR<0	T 0.03 (0.44)
	CH2Cl2	T NEMR<0, W <3 data	T 2.07 (0.86)	T NEMR<0, W <3 data	T 0.74 (0.99)	T r<0.3, W NEMR<0	T r<0.3, W NEMR<0	T NEMR<0, W <3 data
	C2Cl4	T NEMR<0, W <3 data	T 0.01 (0.95)	T NEMR<0, W <3 data	T r<0.3, W n/a	T NEMR<0, W 0.01 (0.65)	T r<0.3, W r<0.3	T NEMR<0, W <3 data
	CH3Cl	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W r<0.3	T NaN, W 1.17 (0.98)	T NaN, W <3 data
	CH3Br	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W <3 data	T NaN, W NEMR<0	T NaN, W NEMR<0	T NaN, W <3 data
	CH3I	T 0.00 (0.63)	T 0.00 (0.76)	T r<0.3, W <3 data	T r<0.3, W <3 data	T 0.03 (0.32)	T 0.02 (0.52)	T NaN, W <3 data
	CH2Br2	T NEMR<0, W <3 data	T 0.00 (0.79)	T NEMR<0, W <3 data	T NEMR<0, W <3 data	T r<0.3, W r<0.3	T r<0.3, W NEMR<0	T NEMR<0, W <3 data
	CHBrCl2	T LLOD, W <3 data	T NEMR<0, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T NEMR<0, W r<0.3	T LLOD, W r<0.3	T LLOD, W <3 data
	CHBr2Cl	T NEMR<0, W <3 data	T r<0.3, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T NEMR<0, W r<0.3	T LLOD, W NEMR<0	T LLOD, W <3 data
	CHBr3	T NEMR<0, W <3 data	T NEMR<0, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T r<0.3, W r<0.3	T r<0.3, W NEMR<0	T r<0.3, W <3 data
	CH2ClCH2Cl	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W r<0.3	T n/a, W <3 data
	ClBenzene	T NEMR<0, W <3 data	T r<0.3, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T NEMR<0, W n/a	T LLOD, W n/a	T LLOD, W n/a
	CH2ClI	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	CH2BrI	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	CH2I2	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
HCOOH	HCOOH	IC 63.88 (0.93)	IC 11.24 (0.73)	IC 56.74 (0.75)	IC 19.48 (0.76)	IC 36.57 (0.93)	IC 62.53 (0.78)	IC 22.23 (0.90)
OC	OC	H 128.75 (0.70)	H 127.71 (0.93)	H 123.13 (0.88)	H NaN	H r<0.3	H r<0.3	H r<0.3

Table S5. Continued

Classes	Plume intercept # / Species	15	16	22	25	26	28	36
Alkanes	Ethane	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 46.05 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 19.24 (0.61)	T n/a, W 10.28 (1.00)
	Propane	T NEMR<0, W <3 data	T NEMR<0, W <3 data	T 22.49 (1.00)	T 6.26 (0.77)	T r<0.3, W <3 data	T r<0.3, W r<0.3	T 2.53 (0.95)
	iButane	T 1.33 (0.77)	T 1.29 (0.75)	T 8.62 (0.99)	T 1.24 (0.89)	T r<0.3, W <3 data	T NEMR<0, W R<0.3	T 0.72 (0.89)
	nButane	T 2.32 (0.51)	T 1.84 (0.59)	T 12.53 (1.00)	T 2.30 (0.94)	T r<0.3, W <3 data	T NEMR<0, W R<0.3	T 1.26 (0.96)
	Isopentane	T LLOD, W <3 data	T LLOD, W <3 data	T 15.49 (1.00)	T 0.81 (0.95)	T r<0.3, W <3 data	T r<0.3, W 1.05 (0.39)	T 1.90 (1.00)
	nPentane	T LLOD, W <3 data	T LLOD, W <3 data	T 4.95 (1.00)	T 0.78 (0.80)	T r<0.3, W <3 data	T r<0.3, W NEMR<0	T 0.76 (0.92)
	nHexane	T LLOD, W <3 data	T LLOD, W <3 data	T 1.54 (0.98)	T 0.11 (0.99)	T r<0.3, W <3 data	T 0.23 (0.31)	T LLOD, W LLOD
	nHeptane	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD
	2Methylpentane	T LLOD, W <3 data	T LLOD, W <3 data	T 4.22 (0.99)	T r<0.3, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD , W LLOD
	3Methylpentane	T LLOD, W <3 data	T LLOD, W <3 data	T 1.28 (1.00)	T 0.05 (0.31)	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD
	224Trimethylpentane	T LLOD, W n/a	T LLOD, W n/a	T 4.07 (0.99)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a
Alkenes	Ethene	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 3.90 (0.94)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W LLOD
	Propene	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.34 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W LLOD	T n/a, W LLOD
	Isobutene+1Butene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T 0.16 (0.40)	T NEMR<0, W n/a	T NEMR<0, W n/a	T LLOD, W n/a
	Isoprene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD
	aPinene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Tricyclene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T 0.66 (0.30)	T 0.37 (0.53)	T LLOD, W n/a
	Camphene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	bPineneMycene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	LimoneneD3Carene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Alkynes	Ethyne	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 9.43 (0.98)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 8.78 (0.92)	T n/a, W 7.03 (0.99)
Aromatics	Benzene	T 2.21 (0.87)	T NEMR<0, W <3 data	T 4.06 (1.00)	T 7.50 (0.98)	T 4.95 (0.93)	T 7.20 (0.62)	T 4.16 (1.00)
	Toluene	T 0.18 (1.00)	T 0.00 (1.00)	T 4.42 (0.98)	T 1.13 (1.00)	T 0.91 (0.21)	T r<0.3, W LLOD	T 1.20 (0.97)
	Ethylbenzene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T 0.16 (0.99)	T LLOD, W <3 data	T LLOD , W LLOD	T LLOD, W LLOD
	mpXylene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W <3 data	T NEMR<0, W LLOD	T LLOD, W LLOD
	oXylene	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T 0.05 (0.98)
Alcohols	CH3OH	T 57.65 (0.44)	T r<0.3, W n/a	T 45.03 (0.74)	T 17.02 (0.95)	T 13.11 (0.68)	T 31.37 (0.41)	T 18.08 (0.34)
	C2H5OH	T 18.10 (0.46)	T 21.41 (0.72)	T 36.58 (1.00)	T 1.79 (1.00)	T 0.97 (0.02)	T r<0.3, W n/a	T NEMR<0, W n/a
Aldehydes	HCHO	F 6.66 (0.67)	F 9.90 (0.48)	F 29.16 (0.96)	F 2.28 (0.45)	F NaN, T 1.57 (0.90)	F r<0.3, TOGA r<0.3	F 10.37 (0.70)
	CH3CHO	T NEMR<0, W n/a	T r<0.3, W n/a	T 18.39 (1.00)	T 5.65 (0.98)	T 4.06 (0.40)	T r<0.3, W n/a	T 4.58 (0.99)
	Propanal	T LLOD, W n/a	T LLOD, W n/a	T 1.27 (0.35)	T NEMR<0, W <3 data	T NEMR<0, W n/a	T r<0.3, W n/a	T LLOD, W n/a
	Butanal	T NEMR<0, W n/a	T 0.88 (0.90)	T 0.61 (0.87)	T 0.24 (1.00)	T 0.73 (0.50)	T NEMR<0, W n/a	T 0.34 (0.96)
	Acrolein	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Methacrolein	T LLOD, W n/a	T LLOD, W n/a	T 0.71 (0.80)	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Ketones	Acetone	T r<0.3, W n/a	T 53.48 (0.75)	T 57.38 (0.96)	T 10.58 (0.27)	T r<0.3, W n/a	T 27.08 (0.34)	T r<0.3, W n/a
	MEK	T r<0.3, W n/a	T 8.97 (0.99)	T 9.31 (1.00)	T 4.53 (0.99)	T 3.48 (0.86)	T 3.66 (0.33)	T 1.74 (0.61)
	MVK	T LLOD, W n/a	T LLOD, W n/a	T 1.92 (0.80)	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Other OVOCs	MTBE	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T 0.08 (0.40)	T NEMR<0, W n/a	T 1.06 (0.53)	T LLOD, W n/a
Other VOCs	OCS	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	DMS	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W LLOD
	HCN	CC r<0.3, T NEMR<0	CC r<0.3, T NEMR<0	CC NEMR<0, T NEMR<0	CC 3.52 (0.59)	CC 4.94 (0.64)	CC 8.49 (0.47)	CC r<0.3, T 1.84 (0.75)
	CH3CN	T 3.46 (0.89)	T NEMR<0, W n/a	T NEMR<0, W n/a	T 0.72 (0.46)	T NEMR<0, W n/a	T 2.18 (0.47)	T r<0.3, W n/a

Table S5. Continued

Classes	Plume intercept# / Species	15	16	22	25	26	28	36
Organic nitrates	Methyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.05 (0.97)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W 0.37 (0.94)
	Ethyl nitrate	T NaN, W <3 data	T NaN, W <3 data	T LLOD, W 0.08 (0.99)	T NaN, W <3 data	T NaN, W <3 data	T NaN, W 0.05 (0.64)	T NaN, W 0.11 (0.98)
	Isopropyl nitrate	T LLOD, W <3 data	T 0.26 (0.71)	T 0.42 (0.90)	T 0.07 (0.42)	T 0.06 (0.42)	T r<0.3, W 0.12 (0.62)	T 0.25 (0.39)
	nPropyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.05 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W 0.02 (1.00)
	2Butyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.52 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.23 (0.41)	T n/a, W 0.08 (1.00)
	3Pentyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.28 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.13 (0.65)	T n/a, W 0.04 (0.85)
	2Pentyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.48 (0.99)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.20 (0.30)	T n/a, W 0.06 (0.93)
	3Methyl2butyl nitrate	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.67 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W 0.05 (1.00)
Halogens	CFC12	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.07 (0.79)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	CFC11	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.02 (0.81)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	CFC113	T NaN, W <3 data	T NaN, W <3 data	T NaN, W 0.02 (0.99)	T NaN, W <3 data	T NaN, W <3 data	T NaN, W NEMR<0	T NaN, W 0.02 (0.54)
	CFC114	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W 0.02 (0.76)
	HFC152a	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 2.27 (1.00)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W r<0.3
	HFC134a	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 2.03 (0.98)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W r<0.3
	HFC365mfc	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W NaN
	HCFC124	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NaN	T n/a, W NaN
	HCFC22	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 1.08 (0.93)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W r<0.3
	HCFC142b	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.06 (0.83)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W NEMR<0
	HCFC141b	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.18 (0.87)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	H1301	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	H2402	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.00 (0.56)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.00 (0.80)	T n/a, W NaN
	H1211	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.01 (0.94)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	CH3CCl3	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.01 (0.99)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W r<0.3
	CCl4	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.03 (0.94)	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W NEMR<0
	CHCl3	T 0.19 (0.80)	T 0.10 (0.99)	T 0.09 (0.99)	T 0.04 (0.87)	T NEMR<0, W <3 data	T r<0.3, W 0.20 (0.65)	T 0.07 (1.00)
	CH2Cl2	T 0.38 (0.76)	T 0.47 (0.99)	T 0.36 (0.78)	T NEMR<0, W <3 data	T NEMR<0, W <3 data	T NEMR<0, W 0.17 (0.42)	T 0.06 (0.98)
	C2Cl4	T NEMR<0, W <3 data	T r<0.3, W <3 data	T 0.11 (0.99)	T 0.00 (0.84)	T NEMR<0, W <3 data	T r<0.3, W r<0.3	T 0.01 (0.46)
	CH3Cl	T NaN, W <3 data	T NaN, W <3 data	T NaN, W NEMR<0	T NaN, W <3 data	T NaN, W <3 data	T NaN, W NEMR<0	T NaN, W 1.24 (0.65)
	CH3Br	T NaN, W <3 data	T NaN, W <3 data	T NaN, W 0.01 (0.62)	T NaN, W <3 data	T NaN, W <3 data	T NaN, W r<0.3	T NaN, W 0.02 (0.67)
	CH3I	T 0.03 (0.69)	T NEMR<0, W <3 data	T 0.00 (0.84)	T r<0.3, W <3 data	T 0.01 (0.70)	T NEMR<0, W r<0.3	T 0.01 (0.82)
	CH2Br2	T 0.01 (1.00)	T NEMR<0, W <3 data	T 0.00 (0.91)	T 0.00 (0.99)	T NEMR<0, W <3 data	T NEMR<0, W r<0.3	T 0.02 (0.77)
	CHBrCl2	T NaN, W <3 data	T LLOD, W <3 data	T LLOD, W 0.01 (1.00)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W 0.00 (0.38)	T LLOD, W 0.00 (0.86)
	CHBr2Cl	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W 0.01 (1.00)	T LLOD, W <3 data	T LLOD, W <3 data	T LLOD, W 0.00 (0.37)	T LLOD, W 0.00 (0.81)
	CHBr3	T r<0.3, W <3 data	T 0.02 (1.00)	T LLOD, W 0.02 (0.98)	T LLOD, W <3 data	T 0.01 (0.66)	T NEMR<0, W r<0.3	T 0.01 (0.60)
	CH2ClCH2Cl	T n/a, W <3 data	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W <3 data	T n/a, W 0.62 (0.40)	T n/a W NEMR<0
	ClBenzene	T LLOD, W n/a	T LLOD, W n/a	T 0.01 (1.00)	T LLOD, W n/a	T 0.01 (0.58)	T NEMR<0, W n/a	T NEMR<0, W n/a
	CH2ClI	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	CH2BrI	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	CH2I2	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
HCOOH	HCOOH	IC 26.06 (0.91)	IC 30.03 (0.74)	IC 24.47 (0.86)	IC 37.28 (0.82)	IC 51.33 (0.79)	IC 38.16 (0.74)	IC 26.50 (0.51)
OC	OC	H r<0.3	H r<0.3	H 68.88 (0.90)	H 111.60 (0.59)	H 170.06 (0.66)	H r<0.3	H r<0.3

Table S5. Continued

Classes	Species \ Plume intercept#	37	38	40	41	48	50	51
Alkanes	Ethane	T n/a, W 10.75 (1.00)	T n/a, W <3 data	T n/a, W 9.38 (0.91)	T n/a, W <3 data	T n/a, W 87.84 (0.40)	T n/a, W <3 data	T n/a, W 70.75 (0.84)
	Propane	T 1.39 (0.98)	T 1.20 (1.00)	T 1.30 (0.67)	T 1.92 (0.99)	T 19.19 (0.36)	T 22.56 (0.98)	T 37.55 (0.47)
	iButane	T 0.18 (0.99)	T 0.12 (0.94)	T 0.11 (0.88)	T 0.25 (0.98)	T 2.93 (0.56)	T 6.35 (0.97)	T 26.38 (0.69)
	nButane	T 0.38 (0.99)	T 0.27 (0.92)	T 0.14 (0.57)	T 0.41 (0.98)	T 4.32 (0.58)	T 8.43 (1.00)	T 8.80 (0.90)
	Isopentane	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T 14.30 (1.00)	T 11.52 (0.88)
	nPentane	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T 3.40 (0.90)
	nHexane	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T 3.16 (0.80)
	nHeptane	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T 0.50 (0.65)
	2Methylpentane	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T 4.86 (1.00)	T 4.13 (0.77)
	3Methylpentane	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T 1.81 (0.75)
	224Trimethylpentane	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T 2.68 (1.00)	T 2.61 (0.79)
Alkenes	Ethene	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD	T n/a, W <3 data	T n/a, W 2.71 (0.90)
	Propene	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD
	Isobutene+1Butene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T 1.84 (0.90)
	Isoprene	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD , W <3 data	T 6.29 (0.65)
	aPinene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Tricyclene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Camphene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	bPineneMycene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T NEMR<0, W n/a
	LimoneneD3Carene	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T r<0.3, W n/a
Alkynes	Ethyne	T n/a, W 5.45 (0.99)	T n/a, W <3 data	T n/a, W 4.93 (0.94)	T n/a, W <3 data	T n/a, W 6.86 (1.00)	T n/a, W <3 data	T n/a, W 10.99 (0.84)
Aromatics	Benzene	T 3.23 (1.00)	T 2.99 (1.00)	T 2.27 (1.00)	T 4.25 (0.98)	T 5.08 (0.95)	T 3.71 (0.98)	T 3.34 (0.92)
	Toluene	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T NaN, W LLOD	T 4.63 (1.00)	T 5.08 (0.72)
	Ethylbenzene	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T 1.11 (0.87)
	mpXylene	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T r<0.3, W LLOD
	oXylene	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T r<0.3, W LLOD
Alcohols	CH3OH	T 20.68 (0.99)	T 15.07 (1.00)	T 19.21 (0.96)	T 31.42 (0.96)	T NEMR<0, W n/a	T 58.82 (0.99)	T NEMR<0, W n/a
	C2H5OH	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD	T 76.10 (0.95)	T NEMR<0, W n/a
Aldehydes	HCHO	F n/a, T 2.32 (0.89)	F n/a, T 1.74 (0.79)	F n/a, T 3.29 (0.68)	F n/a, T 3.24 (0.95)	F n/a, T r<0.3	F n/a, T 7.23 (1.00)	F n/a, T r<0.3
	CH3CHO	T 2.63 (0.80)	T 2.51 (0.53)	T 1.99 (1.00)	T 2.43 (0.88)	T 2.86 (0.63)	T 21.86 (0.99)	T NEMR<0, W n/a
	Propanal	T NEMR<0, W n/a	T 0.25 (0.73)	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T NEMR<0, W n/a
	Butanal	T 0.18 (0.56)	T 0.10 (0.78)	T 0.10 (0.36)	T 0.14 (0.97)	T LLOD, W n/a	T 0.56 (0.91)	T r<0.3, W n/a
	Acrolein	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
	Methacrolein	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T r<0.3, W n/a
Ketones	Acetone	T 25.27 (0.99)	T 19.32 (0.99)	T 22.91 (0.83)	T 36.45 (0.97)	T r<0.3, W n/a	T 49.25 (0.99)	T 47.00 (0.88)
	MEK	T 1.51 (0.99)	T 1.32 (0.99)	T 1.32 (0.99)	T 2.15 (1.00)	T 4.78 (0.44)	T 6.51 (1.00)	T 4.82 (0.87)
	MVK	T r<0.3, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T r<0.3, W n/a
Other OVOCs	MTBE	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a
Other VOCs	OCS	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 3.46 (0.40)
	DMS	T LLOD, W NEMR<0	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W LLOD	T LLOD, W <3 data	T LLOD, W NEMR<0
	HCN	CC 4.40 (0.91)	CC 5.83 (0.95)	CC 5.20 (0.95)	CC 4.13 (0.94)	CC r<0.3, T r<0.3	CC NEMR<0, T NEMR<0	CC NEMR<0, T NEMR<0
	CH3CN	T 1.96 (0.96)	T 1.71 (0.95)	T 1.28 (0.91)	T 3.55 (0.95)	T NEMR<0, W n/a	T NEMR<0	T NEMR<0, W n/a

Table S5. Continued

Classes	Plume intercept # / Species	37	38	40	41	48	50	51
Organic nitrates	Methyl nitrate	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.32 (0.90)	T n/a, W <3 data	T n/a, W 0.05 (0.96)
	Ethyl nitrate	T LLOD, W 0.03 (0.54)	T NaN, W <3 data	T NaN, W 0.03 (0.46)	T NaN, W <3 data	T NaN, W 0.18 (0.84)	T NaN, W <3 data	T NaN, W 0.06 (0.51)
	Isopropyl nitrate	T 0.05 (0.94)	T 0.05 (0.94)	T 0.07 (0.94)	T NEMR<0, W <3 data	T 0.38 (0.85)	T 0.39 (0.93)	T 0.51 (0.85)
	nPropyl nitrate	T n/a, W 0.85 (0.85)	T n/a, W <3 data	T n/a, W 0.01 (0.45)	T n/a, W <3 data	T n/a, W 0.07 (0.41)	T n/a, W <3 data	T n/a, W 0.03 (0.99)
	2Butyl nitrate	T n/a, W 0.03 (0.92)	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.48 (0.33)	T n/a, W <3 data	T n/a, W 0.24 (0.84)
	3Pentyl nitrate	T n/a, W LLOD	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.05 (0.30)	T n/a, W <3 data	T n/a, W 0.23 (0.90)
	2Pentyl nitrate	T n/a, W LLOD	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.27 (0.33)
	3Methyl2butyl nitrate	T n/a, W LLOD	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.70 (0.83)
Halogens	CFC12	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.11 (0.59)	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.52 (0.52)
	CFC11	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.03 (0.61)	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W R<0.3
	CFC113	T LLOD, W NEMR<0	T NaN, W <3 data	T NaN, W 0.03 (0.49)	T NaN, W <3 data	T NaN, W 0.12 (0.86)	T NaN, W <3 data	T NaN, W 0.31 (0.63)
	CFC114	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.07 (0.66)	T n/a, W <3 data	T n/a, W r<0.3
	HFC152a	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W NaN
	HFC134a	T n/a, W NEMR<0	T n/a, W <3 data	T N/A, w r<0.3	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 1.36 (0.97)
	HFC365mfc	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD	T n/a, W <3 data	T n/a, W NaN	T n/a, W <3 data	T n/a, W NaN
	HCFC124	T n/a, W LLOD	T n/a, W <3 data	T n/a, W LLOD	T n/a, W <3 data	T n/a, W NaN	T n/a, W <3 data	T n/a, W NaN
	HCFC22	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.14 (0.76)	T n/a, W <3 data	T n/a, W 0.63 (0.98)	T n/a, W <3 data	T n/a, W 1.55 (0.89)
	HCFC142b	T n/a, W r<0.3	T n/a, W <3 data	T n/a W 0.01 (0.66)	T n/a, W <3 data	T n/a, W 0.14 (0.63)	T n/a, W <3 data	T n/a, W 0.13 (0.72)
	HCFC141b	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.17 (0.99)	T n/a, W <3 data	T n/a, W 0.38 (0.99)
	H1301	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.00 (0.70)	T n/a, W <3 data	T n/a, W 0.01 (0.89)	T n/a, W <3 data	T n/a, W NEMR<0
	H2402	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.00 (0.58)	T n/a, W <3 data	T n/a, W 0.00 (0.81)
	H1211	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.00 (0.92)	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W R<0.3
	CH3CCl3	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.04 (0.48)	T n/a, W <3 data	T n/a, W 0.02 (0.97)
	CCl4	T n/a, W NEMR<1	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W 0.24 (0.48)
	CHCl3	T 0.03 (0.55)	T 0.04 (1.00)	T r<0.3, W NEMR<0	T NEMR<0, W <3 data	T 0.19 (0.86)	T 0.12 (0.95)	T 0.11 (0.66)
	CH2Cl2	T 0.20 (0.63)	T 0.15 (0.94)	T 0.06 (0.58)	T r<0.3, W <3 data	T 0.36 (0.41)	T 0.34 (0.66)	T 0.61 (0.58)
	C2Cl4	T 0.01 (0.37)	T 0.01 (0.98)	T 0.00 (0.35)	T NEMR<0, W <3 data	T 0.06 (0.51)	T 0.09 (0.71)	T r<0.3, W 0.03 (0.99)
	CH3Cl	T LLOD, W 0.51 (1.00)	T NaN, W <3 data	T NaN, W 0.88 (0.83)	T NaN, W <3 data	T LLOD, W NEMR<0	T NaN, W <3 data	T NaN, W 1.51 (0.29)
	CH3Br	T LLOD, W r<0.3	T NaN, W <3 data	T NaN, W LLOD	T NaN, W <3 data	T LLOD, W r<0.3	T NaN, W <3 data	T NaN, W r<0.3
	CH3I	T 0.01 (0.81)	T 0.01 (0.97)	T 0.00 (1.00)	T 0.01 (0.93)	T r<0.3, W NEMR<0	T 0.01 (0.97)	T 0.01 (0.37)
	CH2Br2	T R<0.3, W 0.00 (0.41)	T 0.00 (0.88)	T r<0.3, W NEMR<0	T NEMR<0, W <3 data	T 0.01 (0.66)	T 0.00 (0.33)	T NEMR<0, W 0.01 (1.00)
	CHBrCl2	T LLOD, W r<0.3	T LLOD, W <3 data	T LLOD, W r<0.3	T LLOD, W <3 data	T LLOD, W 0.01 (0.80)	T LLOD, W <3 data	T LLOD, W 0.01 (1.00)
	CHBr2Cl	T LLOD, W r<0.4	T LLOD, W <3 data	T LLOD, W r<0.3	T LLOD, W <3 data	T LLOD, W 0.01 (0.73)	T LLOD, W <3 data	T NEMR<0, W 0.01 (0.99)
	CHBr3	T r<0.3, W r<0.3	T 0.00 (0.80)	T NEMR<0, W r<0.3	T 0.00 (0.31)	T 0.02 (0.89)	T 0.01 (0.80)	T 0.02 (0.71)
	CH2ClCH2Cl	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W NEMR<0	T n/a, W <3 data	T n/a, W r<0.3	T n/a, W <3 data	T n/a, W 0.52 (0.56)
	ClBenzene	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T 0.02 (0.69)	T 0.01 (0.97)	T 0.04 (0.82)
	CH2ClI	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a
	CH2BrI	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a
	CH2I2	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W n/a	T LLOD, W <3 data	T LLOD, W n/a
HCOOH	HCOOH	IC 47.91 (0.98)	IC 42.98 (0.96)	IC 112.33 (0.95)	IC 129.26 (0.93)	IC 31.03 (0.82)	IC 87.41 (0.94)	IC 55.29 (0.69)
OC	OC	H 33.38 (0.88)	H 36.69 (0.89)	H 36.00 (0.94)	H 31.03 (0.91)	H 78.34 (0.64)	H 136.29 (0.72)	H r<0.3

Table S6. Primary emission ratios (ERs, ppt C/ppb CO) for fire-emitted ROC species

Classes	ATom reported species[a]	Reference	Savanna & grassland	Agricultural residues	Tropical forest	Temperate forest	Boreal forest
Alkanes	Ethane	Andreae 2019	11.24	19.25	15.80	11.50	14.93
	Propane	Andreae 2019	3.70	4.19	9.78	4.67	4.64
	iButane	Andreae 2019	0.21	0.41	0.27	0.52	0.83
	nButane	Andreae 2019	0.58	1.09	0.76	1.38	1.76
	Isopentane	Akagi 2011	0.06	0.51	0.19	n/a	0.61
	nPentane	Andreae 2019	0.18	1.06	0.27	0.58	0.81
	nHexane	Andreae 2019	0.51	0.81	0.59	0.55	0.87
	nHeptane	Andreae 2019	0.45	0.80	0.44	0.50	0.34
	2Methylpentane	Akagi 2011	n/a	n/a	n/a	n/a	n/a
	3Methylpentane	Akagi 2011	n/a	n/a	n/a	n/a	n/a
	224Trimethylpentane	n/a					
Alkenes	Ethene	Andreae 2019	24.06	26.10	21.22	19.75	25.37
	Propene	Andreae 2019	13.37	12.37	16.55	10.63	11.10
	Isobutene+1Butene	Andreae 2019	1.78	2.12	1.75	1.83	1.75
	Isoprene	Andreae 2019	3.01	4.59	4.34	1.83	1.26
	aPinene Tricyclene Camphene bPineneMycrene LimoneneD3Carene	Reported as terpene group in Andreae 2019	3.09	0.72	2.96	21.35	25.96
Alkyne	Ethyne	Andreae 2019	9.73	7.69	7.30	5.94	5.02
Aromatics	Benzene	Andreae 2019	10.27	7.63	7.85	8.03	10.12
	Toluene	Andreae 2019	5.87	4.70	3.90	5.16	6.19
	Ethylbenzene	Andreae 2019	0.67	1.22	0.87	0.76	0.66
	mpXylene oXylene	Reported as xylene group in Andreae 2019	2.62	2.91	1.75	2.98	1.84
Alcohols	CH3OH	Andreae 2019	17.08	37.86	23.50	17.08	16.80
	C2H5OH	Andreae 2019	0.64	0.79	0.79	0.82	0.58
Aldehydes	HCHO	Andreae 2019	16.60	22.03	21.49	16.90	13.46
	CH3CHO	Andreae 2019	15.45	30.04	27.59	13.67	8.50
	Propanal	Andreae 2019	1.11	3.42	1.39	1.11	2.91
	Butanal	Andreae 2019	2.56	3.41	1.89	1.51	1.97
	Acrolein	Andreae 2019	10.31	12.23	9.35	4.52	4.14
	Methacrolein	Andreae 2019	2.31	5.87	2.30	1.99	1.45
	Hydroxyacetaldehyde[a]	Andreae 2019	1.78	38.92	2.92	3.23	2.92
Ketones	Acetone	Andreae 2019	9.84	13.48	8.75	9.77	18.98
	MEK	Andreae 2019	2.94	11.78	7.46	3.14	2.04
	MVK	Andreae 2019	5.32	10.07	5.99	2.34	1.31
	Hydroxyacetone[a]	Andreae 2019	9.12	46.38	19.71	11.38	19.71
Other OVOCs	HCOOH	Andreae 2019	1.83	4.45	2.83	4.90	5.22
	MTBE	Use methyl vinyl ether from Akagi 2011	3.35	1.52	n/a	n/a	n/a
Other VOCs	OCS	Akagi 2011	n/a	n/a	0.11	n/a	1.77
	DMS	Andreae 2019	0.05	0.27	0.01	0.06	0.01
	HCN	Andreae 2019	6.60	5.71	4.38	5.89	4.53
	CH3CN	Akagi 2011	2.17	3.76	5.37	2.70	3.47
Organic nitrates	Methyl nitrate	Akagi 2011	0.00	n/a	0.03	n/a	0.01
	Ethyl nitrate	Akagi 2011	n/a	n/a	0.03	n/a	0.01
	Isopropyl nitrate	Akagi 2011	n/a	n/a	0.01	n/a	0.02
	nPropyl nitrate	Akagi 2011	n/a	n/a	0.00	n/a	0.00
	2Butyl nitrate	Akagi 2011	n/a	n/a	0.01	n/a	0.03
	3Pentyl nitrate	Akagi 2011	n/a	n/a	n/a	n/a	n/a
	2Pentyl nitrate	Akagi 2011	n/a	n/a	n/a	n/a	n/a
	3Methyl2butyl nitrate	Akagi 2011	n/a	n/a	n/a	n/a	n/a
Halogens	CFC12	n/a					
	CFC11	n/a					
	CFC113	n/a					
	CFC114	n/a					
	HFC152a	n/a					
	HFC134a	n/a					
	HFC365mfc	n/a					
	HCFC124	n/a					
	HCFC22	n/a					

	HCFC142b	n/a					
	HCFC141b	n/a					
	H1301	n/a					
	H2402	n/a					
	H1211	n/a					
	CH3CCl3	n/a					
	CCl4	n/a					
	CHCl3	Akagi 2011	0.04	n/a	0.00	n/a	n/a
	CH2Cl2	n/a					
	C2Cl4	n/a					
	CH3Cl	Andreae 2019	0.50	1.21	0.15	0.21	0.27
	CH3Br	Andreae 2019	0.01	0.00	0.02	0.00	0.01
	CH3I	Andreae 2019	0.00	0.00	0.01	0.00	0.00
	CH2Br2	Akagi 2011	n/a	n/a	n/a	n/a	0.00
	CHBrCl2	n/a					
	CHBr2Cl	n/a					
	CHBr3	n/a					
	CH2ClCH2Cl	n/a					
	ClBenzene	n/a					
	CH2ClI	n/a					
	CH2BrI	n/a					
	CH2I2	n/a					
ER of reported VOC			201.01	351.37	242.67	199.17	224.16
OC		Andreae 2019	101.19	149.91	98.48	225.71	113.45
ER of reported ROC			302.20	501.28	341.14	424.88	337.61
ER of unmeasured+unidentified ROC[b]			201-402	351-702	243-486	199-398	224-448
ER of reported+unmeasured/unidentified ROC			503-704	852-1203	584-827	624-823	562-786
ER from known gas-phase HCOOH precursors			91.18	188.62	118.00	96.64	107.80

[a] Hydroxyacetone and hydroxyacetaldehyde are not reported but included here as known HCOOH precursors

[b] Unmeasured + unidentified ROC are estimated at 1-2× the total reported VOC[22]

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