Supplement of

On the sources and sinks of atmospheric VOCs: An integrated analysis of recent aircraft campaigns over North America

Xin Chen, Dylan B. Millet et al.

Correspondence to: Dylan B. Millet (dbm@umn.edu)

Table S1. Gas-phase species and instrumentation used here.

Campaign	Species involved in this work	Measurement technique	Measurement reference
CalNex	isoprene, monoterpenes, benzene, toluene, C8 aromatics methanol, acetone, methyl ethyl ketone, formaldehyde, acetaldehyde, MVK+MACR acetonitrile	PTR-MS	de Gouw and Warneke (2007)
	2-$BuONO_2$, 3-$PenONO_2$, 2-$PenONO_2$, 3-methyl-2-$BuONO_2$ C_2H_6, C_3H_8, i-butane, n-butane, i-pentane, n-pentane, n-hexane, n-heptane, n-octane, 2,3-dimethylbutane, 2-methylpentane, 3-methylpentane, C_2H_4, propene, 1-butene, trans-2-butene, cis-2-butene, C_2H_2	WAS	Colman et al. (2001), Schauffler et al. (2003)
	PAN	CIMS	Zheng et al. (2011), Osthoff et al. (2008)
	NO, NO_2, NO_y	Gas-phase chemiluminescence	Pollack et al. (2010), Ryerson et al. (1999), Ryerson et al. (1998)
DC3 DC-8	isoprene, monoterpenes, benzene, toluene, xylene methanol, acetone, acetaldehyde, MVK+MACR acetonitrile	PTR-MS	Wisthaler et al. (2002)
	formaldehyde	DFGAS (base)	Richter et al. (2015), Weibring et al. (2010)
	formaldehyde	ISAF-LIF (sens)	Cazorla et al. (2015), DiGangi et al. (2011), Hottle et al. (2009)
	ISOPOOH+IEPOX, glycolaldehyde, hydroxyacetone, peroxyacetic acid, CH_3OOH ISOPN, PROPNN	CIT-CIMS (CF_3O^-)	Crounse et al. (2006), St Clair et al. (2010)
	2-$BuONO_2$, 3-methyl-2-$BuONO_2$, 3-$PenONO_2$, 2-$PenONO_2$ C_2H_6, C_3H_8, i-butane, n-butane, i-pentane, n-pentane, 2,3-dimethylbutane, 2-methylpentane, 3-methylpentane, n-hexane, n-heptane, 2,4-dimethylpentane, 2-methylhexane, 2,3-dimethylpentane, 3-methylhexane, 2,2,4-trimethylpentane, C_2H_4, propene, trans-2-butene, cis-2-butene, cyclopentane, methylcyclopentane, cyclohexane, methylcyclohexane, C_2H_2	WAS	Blake et al. (2003)
	PAN, PPN	CIMS	Huey (2007), Kim et al. (2007), Slusher et al. (2004)
	MPN	TD-LIF	Wooldridge et al. (2010)
	NO, NO_2, NO_y	Gas-phase chemiluminescence	Pollack et al. (2010), Ryerson et al. (1999), Ryerson et al. (1998)
DC3 GV	i-butane, n-butane, i-pentane, n-pentane, n-hexane, n-heptane, benzene, toluene, ethylbenzene+m,p-xylene, o-xylene, isoprene, alpha-pinene, camphene, beta-pinene, limonene methanol, ethanol, methyl butenol, acetone, MEK, propanal, butanal, acetaldehyde, MVK, MACR acetonitrile	TOGA	Apel et al. (2010)
	formaldehyde	CAMS	Fried et al. (2011)
	CH_3OOH	PCIMS	O'Sullivan et al. (2018)
	NO, NO_2	Gas-phase chemiluminescence	Weinheimer et al. (1994)
SENEX	isoprene, monoterpenes, benzene, toluene, xylene methanol, acetone, methyl ethyl ketone, acetaldehyde, MVK+MACR acetonitrile	PTR-MS	de Gouw and Warneke (2007)
	C_2H_6, C_3H_8, i-butane, n-butane, i-pentane, n-pentane, n-hexane, C_2H_4, propene, C_2H_2	WAS	Lerner et al. (2017), Gilman et al. (2009)
	formaldehyde	ISAF-LIF	Cazorla et al. (2015), DiGangi et al. (2011), Hottle et al. (2009)
	ISOPOOH+IEPOX ISOPN, MVKN+MACRN HCOOH, $C_2H_4O_3$, $C_3H_4O_3$, $C_3H_4O_4$, $C_5H_8O_4$, $C_9H_{14}O_4$, $C_{10}H_{16}O_3$	UW CIMS (sens)	Lee et al. (2014)
	HCOOH	NOAA CIMS (base)	
	glyoxal	ACES	Min et al. (2016)

	PAN, PPN	CIMS	_ENREF_28, Zheng et al. (2011), Osthoff et al. (2008), Slusher et al. (2004)
	NO, NO_2, NO_y	Gas-phase chemiluminescence	Pollack et al. (2010), Ryerson et al. (1999), Ryerson et al. (1998)
SEAC[4]RS	isoprene, monoterpenes, benzene, toluene methanol, acetone, acetaldehyde, MVK+MACR acetonitrile	PTR-MS	Wisthaler et al. (2002)
	formaldehyde	CAMS (base)	Fried et al. (2011)
	formaldehyde	ISAF-LIF (sens)	Cazorla et al. (2015), DiGangi et al. (2011), Hottle et al. (2009)
	ISOPOOH+IEPOX, hydroxyacetone, peroxyacetic acid ISOPN, PROPNN, MVKN+MACRN $C_4O_4H_7N$, $C_5O_5H_9N$, BUTENE_HN	CIT-CIMS (CF_3O^-)	Crounse et al. (2006), St Clair et al. (2010)
	2-BuONO_2, 3-methyl-2-BuONO_2, 3-PenONO_2, 2-PenONO_2 C_2H_6, C_3H_8, i-butane, n-butane, i-pentane, n-pentane, neopentane, n-hexane, 2,3-dimethylbutane, 2-methylpentane, 3-methylpentane, n-heptane, C_2H_4, propene, trans-2-butene, cis-2-butene, 1-butene, i-butene, 1-pentene, C_2H_2	WAS	Blake et al. (2003)
	PAN, PPN	PAN-CIMS	Huey (2007), Kim et al. (2007), Slusher et al. (2004)
	MPN	TD-LIF	Wooldridge et al. (2010)
	NO, NO_2, NO_y	Gas-phase chemiluminescence	Pollack et al. (2010), Ryerson et al. (1999), Ryerson et al. (1998)
DISCOVER-AQ	formaldehyde	DFGAS	Richter et al. (2015), Weibring et al. (2010)
	non-methane VOCs	PTR(-ToF-)MS	Müller et al. (2016), Müller et al. (2014)
	ethane (DISCOVER-AQ CO)	TILDAS	Yacovitch et al. (2014)
	NO, NO_2, NO_y	Gas-phase chemiluminescence	Weinheimer et al. (1994)
FRAPPÉ	formaldehyde, ethane	CAMS	Fried et al. (2011)
	monoterpenes, acetonitrile methanol, acetaldehyde, acetone, isoprene, MVK+MACR, methyl ethyl ketone, benzene, toluene, C8 aromatics	PTR-MS (sens)	Kaser et al. (2013)
	methanol, acetaldehyde, acetone, isoprene, MVK, MACR, methyl ethyl ketone, benzene, toluene, ethylbenzene-m-p-xylene+o-xylene ethanol, propanal, butanal, methyl butenol C_3H_8, i-butane, n-butane, i-pentane, n-pentane, n-hexane,2-methylpentane, 3-methylpentane, n-heptane tert-butylnitrate, 2-butylnitrate-n-butylnitrate, 2-pentylnitrate-3-pentylnitrate	TOGA (base)	Apel et al. (2010)
	CH_3OOH, HCOOH, Acetic acid	PCIMS	Treadaway et al. (2018)
	C_2H_4, propene, cyclopentane, methylcyclopentane, cyclohexane, methylcyclohexane, C_2H_2	WAS	Blake et al. (2003)
	PAN, PPN	PAN-CIMS	Zheng et al. (2011)
	NO, NO_2	Gas-phase chemiluminescence	(Weinheimer et al. (1994))

Table S2. Sensitivity of spatially aggregated model performance for total VOC-carbon to the use of data from alternate instruments for co-measured VOCs.

	Base case[a]	DC3 LIF HCHO	SEAC4RS LIF HCHO	SENEX UW CIMS HCOOH	FRAPPÉ PTRMS VOCs	All
NMB						
FT (>3km)	-63.7%	-62.0%	-63.8%	-63.7%	-63.7%	-62.1%
PBL (<2km)	-37.2%	-36.3%	-37.3%	-37.2%	-37.3%	-36.3%
R^2						
FT (>3km)	0.066	0.097	0.067	0.065	0.066	0.098
PBL (<2km)	0.361	0.445	0.362	0.369	0.360	0.449

[a]Base case uses DC3 DFGAS HCHO, SEAC4RS CAMS HCHO, SENEX NOAA CIMS HCOOH, FRAPPÉ TOGA methanol, acetaldehyde, acetone, isoprene, MVK, MACR, MEK, benzene, toluene, C8 aromatics.

Table S3. Sensitivity of spatially aggregated model performance for total VOC reactivity to the use of data from alternate instruments for co-measured VOCs.

	Base case[a]	DC3 LIF HCHO	SEAC4RS LIF HCHO	SENEX UW CIMS HCOOH	FRAPPÉ PTRMS VOCs	All
NMB						
FT (>3km)	-62.6%	-61.9%	-63.2%	-62.6%	-62.6%	-62.5%
PBL (<2km)	-33.9%	-32.7%	-34.0%	-33.7%	-33.9%	-32.8%
R^2						
FT (>3km)	0.043	0.045	0.046	0.043	0.043	0.048
PBL (<2km)	0.542	0.629	0.542	0.547	0.542	0.631

[a]Base case uses DC3 DFGAS HCHO, SEAC4RS CAMS HCHO, SENEX NOAA CIMS HCOOH, FRAPPÉ TOGA methanol, acetaldehyde, acetone, isoprene, MVK, MACR, MEK, benzene, toluene, C8 aromatics.

Table S4. Sensitivity of campaign-aggregated mixing ratio and model bias to the use of data from alternate instruments for co-measured VOCs

		PBL			FT		
		Mixing ratio (ppbC)	Model bias (ppbC)	Model bias (%)	Mixing ratio (ppbC)	Model bias (ppbC)	Model bias (%)
Formaldehyde	base	1.930	-0.401	-22.6	0.230	-0.048	-27.8
	sens	1.944	-0.424	-23.3	0.250	-0.072	-36.9
Formic acid	base	1.006	-0.721	-76.4	0.430	-0.330	-86.3
	sens	0.972	-0.709	-75.8	0.244	-0.153	-73.5
Methanol	base	4.238	-2.135	-59.9	1.299	-0.924	-77.7
	sens	4.191	-2.062	-58.8	1.303	-0.927	-77.7
Acetaldehyde	base	1.162	-0.545	-57.3	0.185	-0.156	-91.0
	sens	1.212	-0.580	-59.3	0.186	-0.159	-91.1
Acetone	base	6.671	-2.150	-34.7	3.459	-1.761	-52.3
	sens	6.803	-2.272	-36.0	3.466	-1.776	-52.4
Isoprene	base	0.259	-0.116	-74.1	0.019	-0.018	-100.0
	sens	0.309	-0.155	-78.1	0.020	-0.019	-100.0
MVK+MACR	base	0.424	-0.165	-58.5	0.035	-0.031	-99.7
	sens	0.450	-0.177	-58.5	0.035	-0.031	-99.7
MEK	base	0.636	0.240	43.4	0.167	-0.077	-75.5
	sens	0.697	0.187	34.9	0.168	-0.078	-76.4
Benzene	base	0.291	-0.095	-38.9	0.108	-0.073	-72.9
	sens	0.298	-0.105	-40.6	0.108	-0.074	-73.0
Toluene	base	0.210	0.041	13.6	0.026	-0.021	-97.8
	sens	0.221	0.039	8.6	0.026	-0.021	-97.8
Xylene	base	0.112	-0.039	-59.5	0.014	-0.013	-100.0
	sens	0.126	-0.052	-62.9	0.015	-0.015	-100.0

Table S5. Sensitivity of campaign-aggregated reactivity and model bias to the use of data from alternate instruments for co-measured VOCs

		PBL			FT		
		Reactivity (s^{-1})	Model bias (s^{-1})	Model bias (%)	Reactivity (s^{-1})	Model bias (s^{-1})	Model bias (%)
Formaldehyde	base	0.343	-0.071	-22.6	0.023	-0.004	-26.7
	sens	0.346	-0.076	-23.3	0.026	-0.007	-35.8
Formic acid	base	0.009	-0.006	-76.4	0.003	-0.002	-86.3
	sens	0.009	-0.006	-75.8	0.002	-0.001	-73.5
Methanol	base	0.080	-0.039	-59.2	0.010	-0.008	-77.2
	sens	0.080	-0.038	-58.2	0.010	-0.008	-77.2
Acetaldehyde	base	0.191	-0.087	-57.4	0.018	-0.015	-90.7
	sens	0.198	-0.093	-59.3	0.018	-0.016	-90.8
Acetone	base	0.008	-0.003	-34.6	0.002	-0.001	-52.1
	sens	0.008	-0.003	-36.0	0.002	-0.001	-52.3
Isoprene	base	0.111	-0.050	-74.1	0.006	-0.005	-100.0
	sens	0.130	-0.066	-78.1	0.006	-0.005	-100.0
MVK+MACR	base	0.052	-0.021	-58.5	0.003	-0.003	-99.7
	sens	0.055	-0.022	-58.2	0.003	-0.003	-99.7
MEK	base	0.004	0.001	40.6	0.001	-0.000	-72.3
	sens	0.005	0.001	34.7	0.001	-0.000	-73.2
Benzene	base	0.001	-0.000	-38.1	0.000	-0.000	-73.3
	sens	0.001	-0.000	-40.6	0.000	-0.000	-73.4
Toluene	base	0.004	0.001	13.7	0.000	-0.000	-97.7
	sens	0.004	0.001	8.6	0.000	-0.000	-97.8
Xylene	base	0.007	-0.002	-59.5	0.001	-0.000	-100.0
	sens	0.008	-0.003	-62.9	0.001	-0.001	-100.0

Table S6. Correlation between observed OVOCs and model-derived biogenic ($\mathcal{B}_{OVOC}$) and anthropogenic ($\mathcal{A}_{OVOC}$) contributions.

	SEAC^{4}RS	SENEX	DISCOVER-AQ TX	Other campaigns
Acetaldehyde	0.71	0.62	0.65	
Formaldehyde	0.71	0.71	0.63	
Acetone	0.87	0.70	0.68	
MEK	n/a	0.68	0.62	
PAA	0.85	n/a	n/a	
Methanol	0.55	0.73	0.53	
MVK	0.74	0.75	0.82	<0.50
MACR	0.75	0.75	0.78	
HCOOH	n/a	0.79	n/a	
Acetic acid	n/a	n/a	0.44	
Glyoxal	n/a	0.79	n/a	
Glycolaldehyde	n/a	n/a	0.63	
Hydroxyacetone	0.80	n/a	n/a	

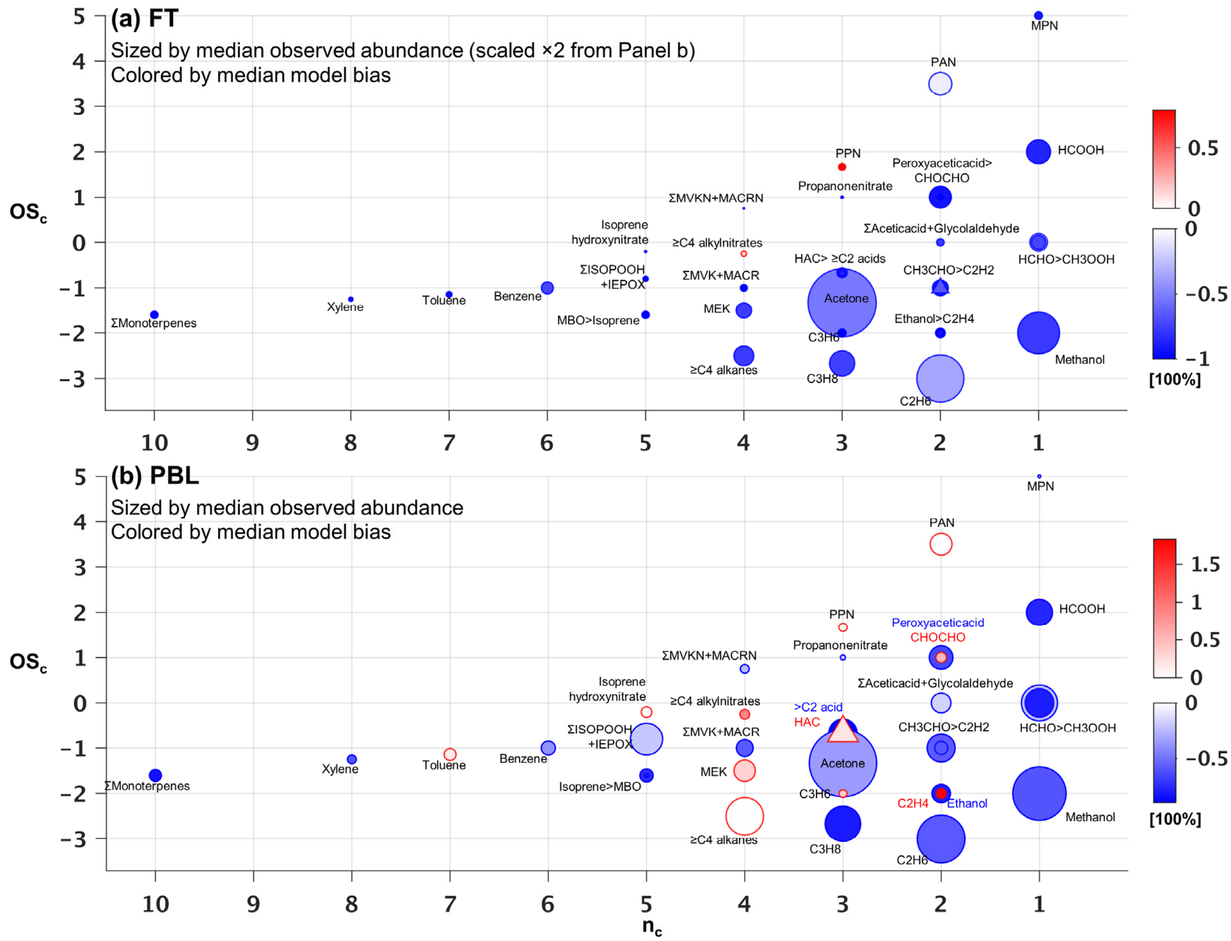

Figure S1. Same as Figure 5, but with the color shaded indicating the median relative model bias.

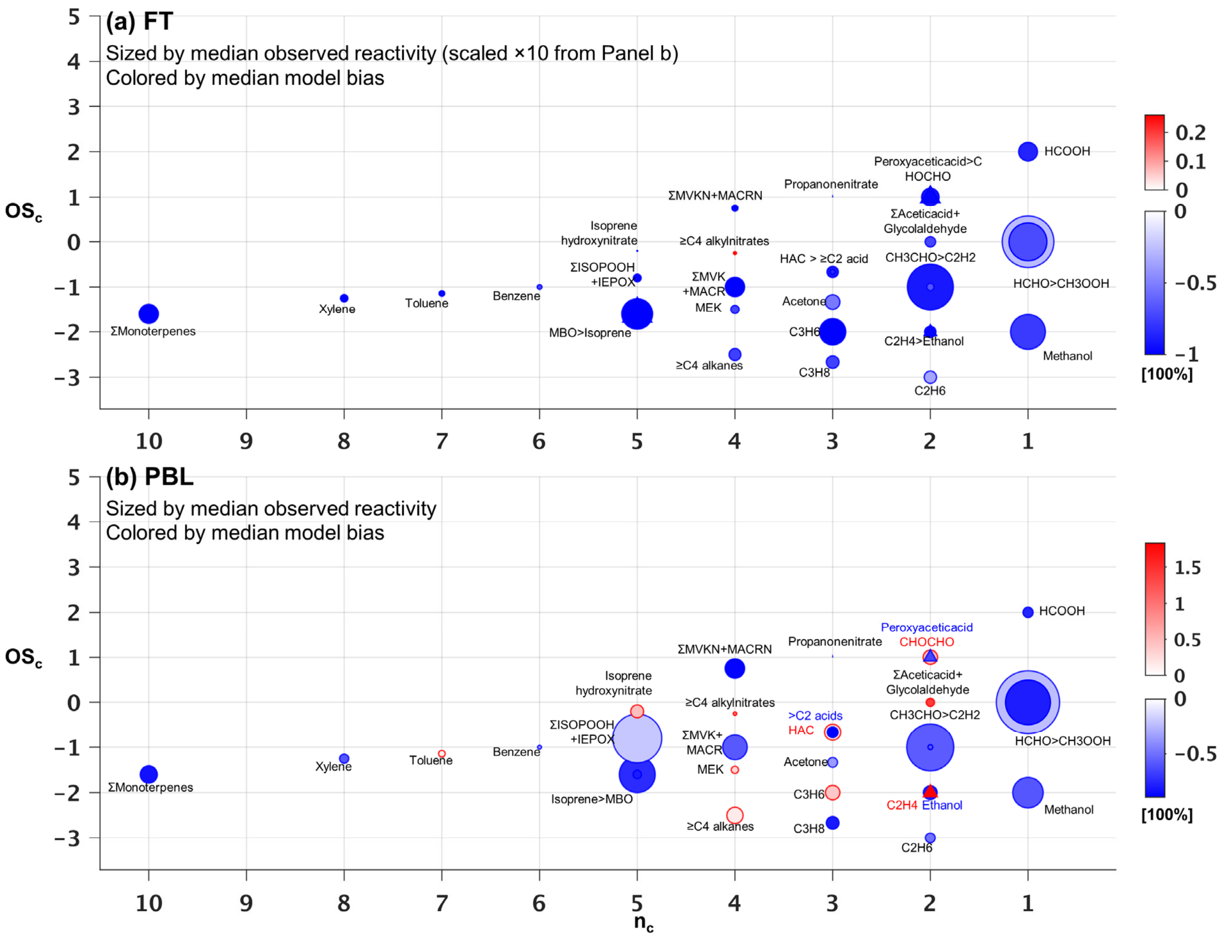

Figure S2. Same as Figure 6, but with the color shaded indicating the median relative model bias. Note that the relative model bias in VOC-carbon for a given species is identical to that for OH reactivity, except in the case of lumped or co-measured species.

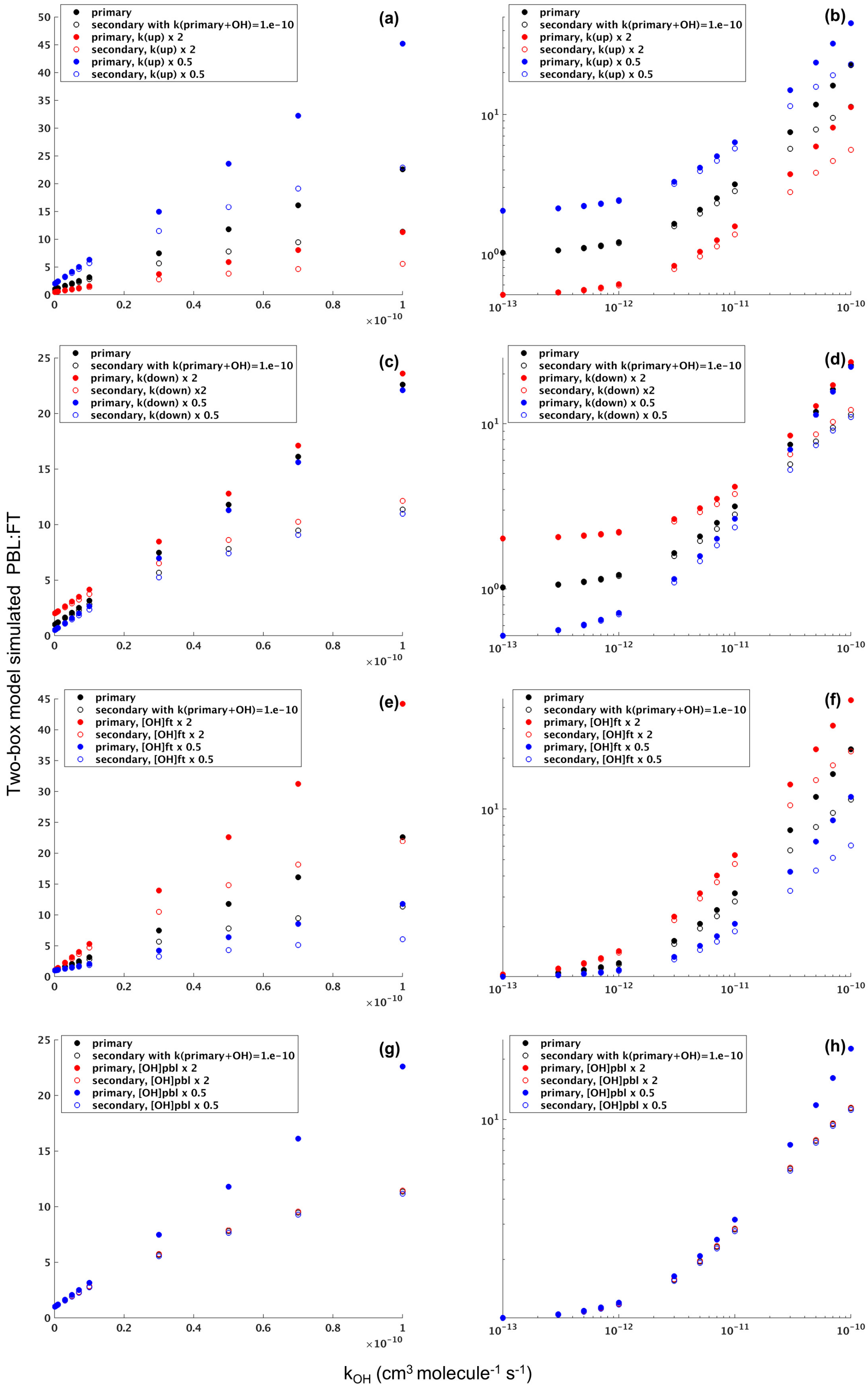

Figure S3. Sensitivity of the PBL:FT concentration ratio for primary and secondary VOCs to PBL-FT exchange rates, OH, and reactivity, based on a simple two-box model. See main text.

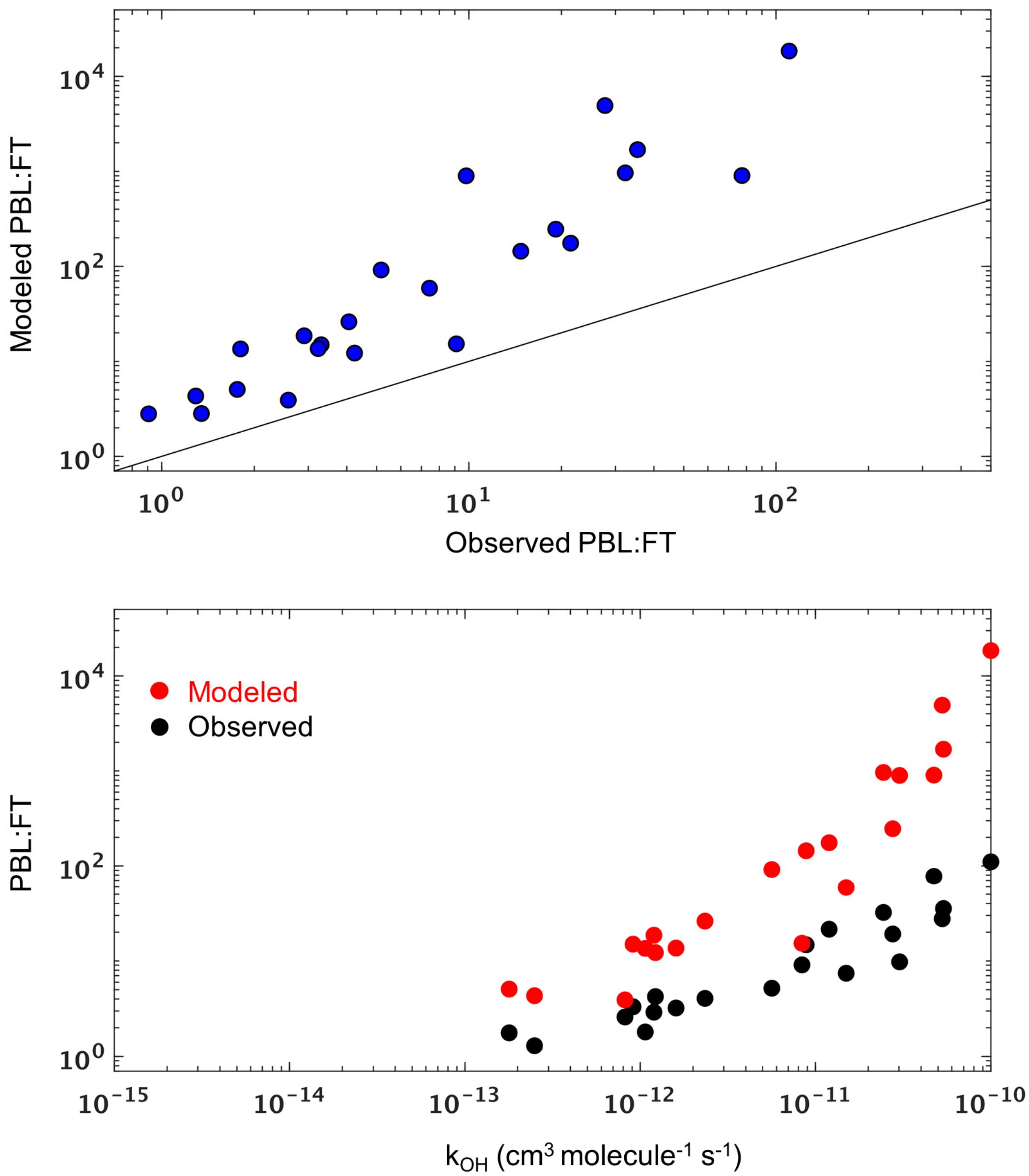

Figure S4. Same as Figure 9, but with model results from a sensitivity simulation with 40% reduced PBL depths.

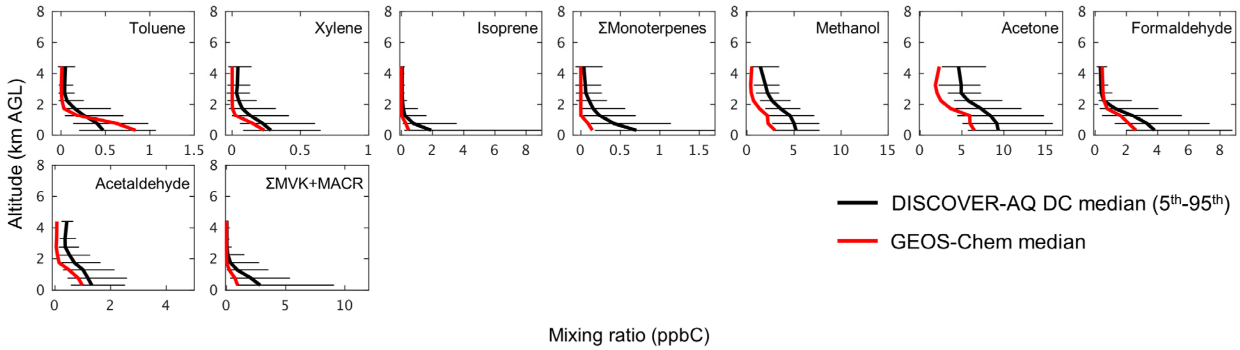

Figure S5. Vertical VOC profiles as measured and simulated by GEOS-Chem during the DISCOVER-AQ DC aircraft campaign. Plotted are the observed (black) and predicted (red) median profiles (in ppbC), with horizontal bars indicating the 5th - 95th percentiles measured for each bin. The vertical bin resolution is 0.5km below 3km and 1km above 3km. Fresh biomass burning and pollution plumes have been filtered out as described in-text.

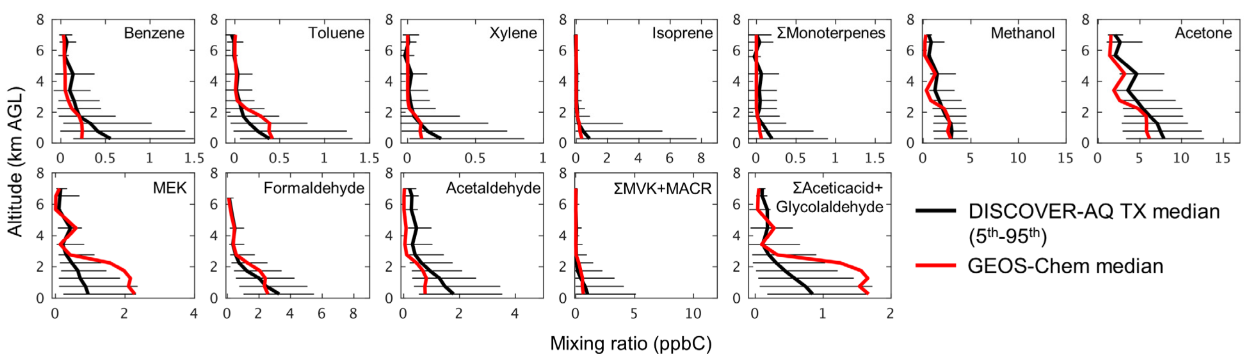

Figure S6. Same as Figure S5 but for the DISCOVER-AQ TX aircraft campaign.

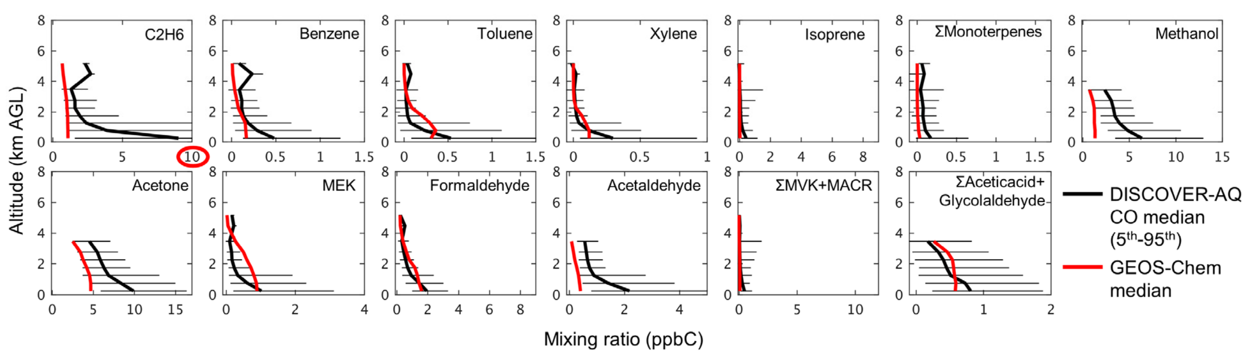

Figure S7. Same as Figure S5 but for the DISCOVER-AQ CO aircraft campaign. Red circles indicate axis scales that differ from others for the same compound in Fig. S5, 6, 9, 10.

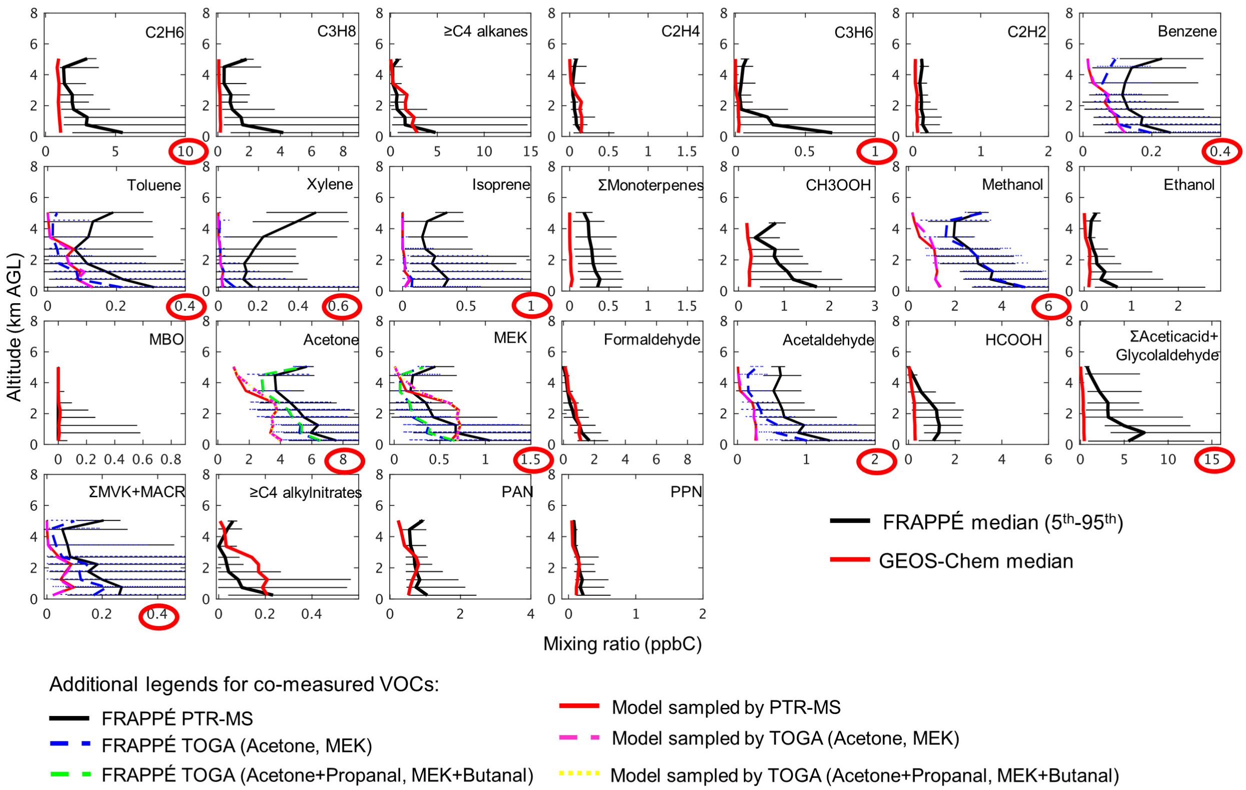

Figure S8. Same as Figure S5 but for the FRAPPÉ aircraft campaign. Red circles indicate axis scales that differ from others for the same compound in Fig. S5, 6, 9, 10.

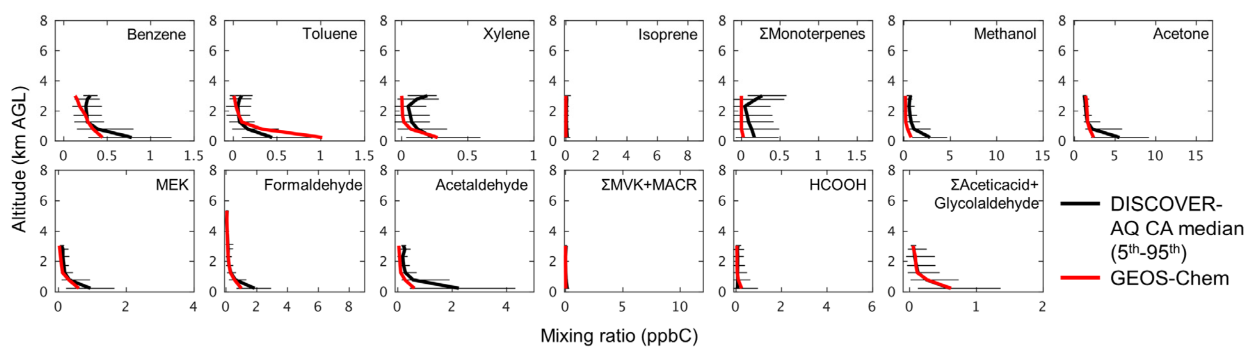

Figure S9. Same as Figure S5 but for the DISCOVER-AQ CA aircraft campaign.

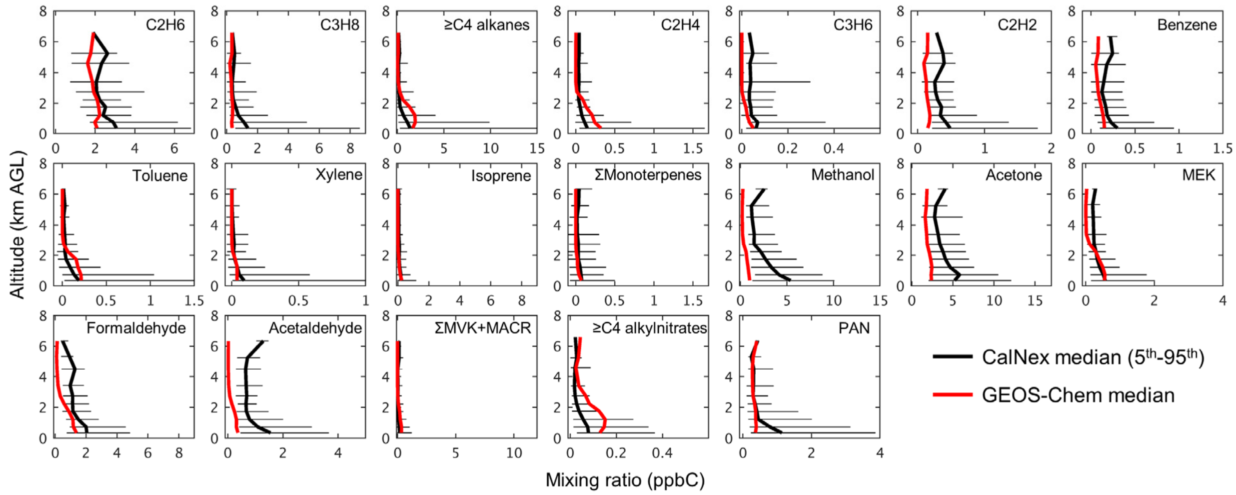

Figure S10. Same as Figure S5 but for the CalNex aircraft campaign.

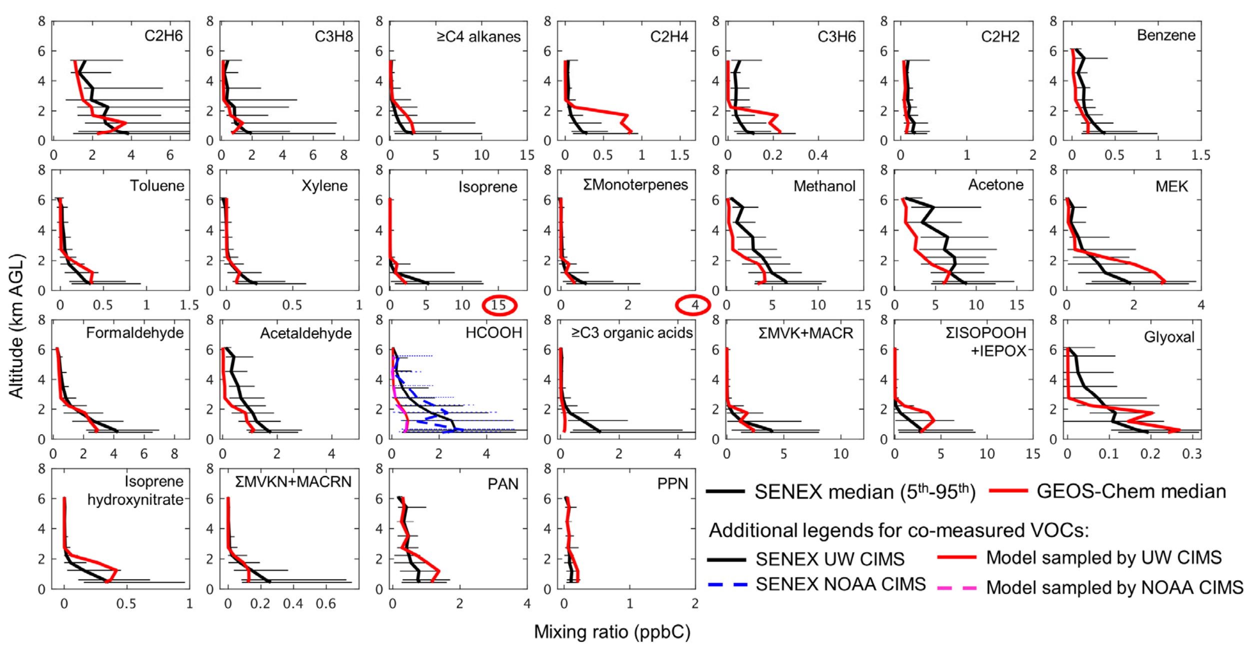

Figure S11. Same as Figure S5 but for the SENEX aircraft campaign. Red circles indicate axis scales that differ from others for the same compound in Figures S5, 6, 9, 10.

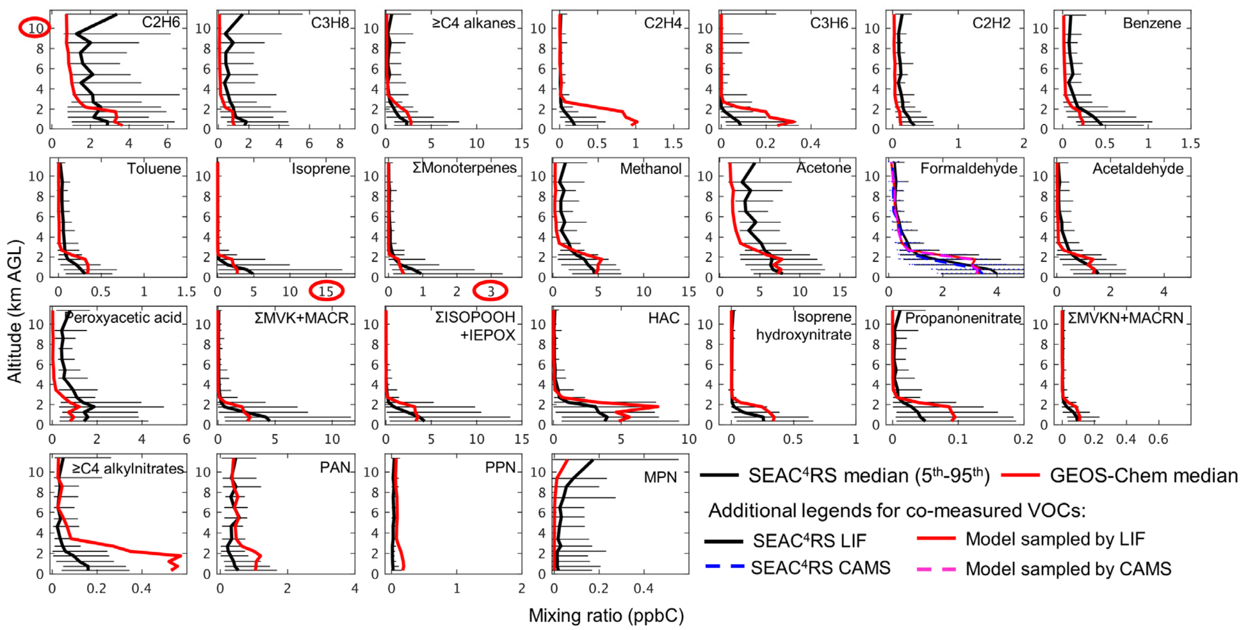

Figure S12. Same as Figure S5 but for the SEAC^{4}RS aircraft campaign. Red circles indicate axis scales that differ from others for the same compound in Figures S5, 6, 9, 10.

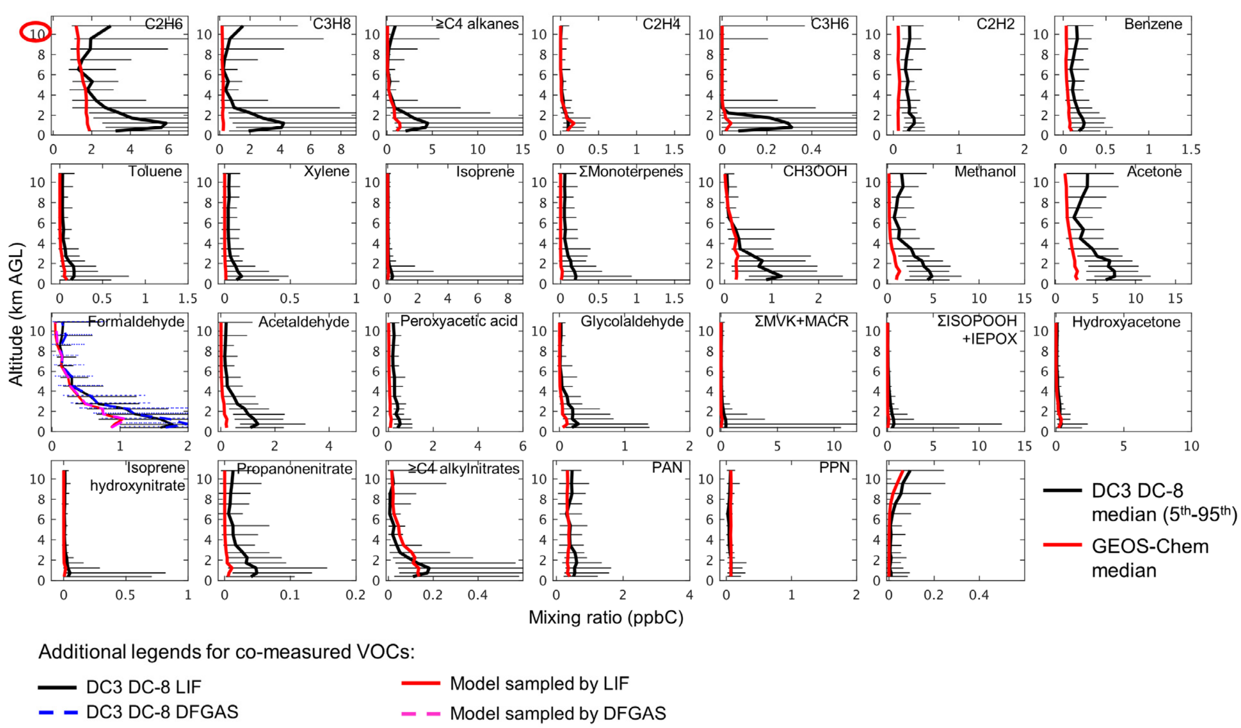

Figure S13. Same as figure S5 but for the DC3 DC-8 aircraft observation. Red circles indicate axis scales that differ from others for the same compound in Figures S5, 6, 9, 10.

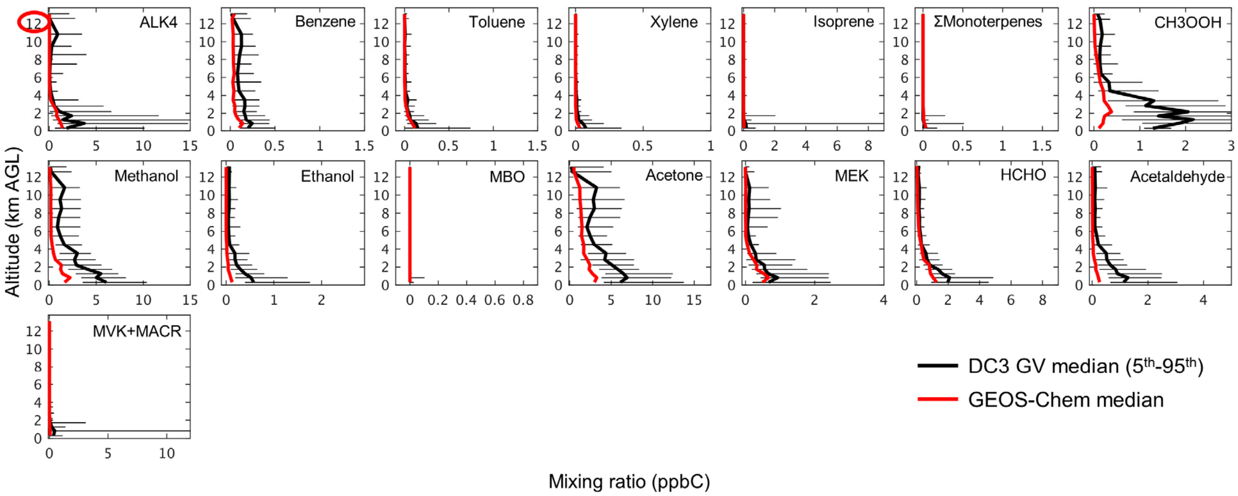

Figure S14. Same as figure S5 but for the DC3 GV aircraft observation. Red circles indicate axis scales that differ from others for the same compound in Figures S5, 6, 9, 10.

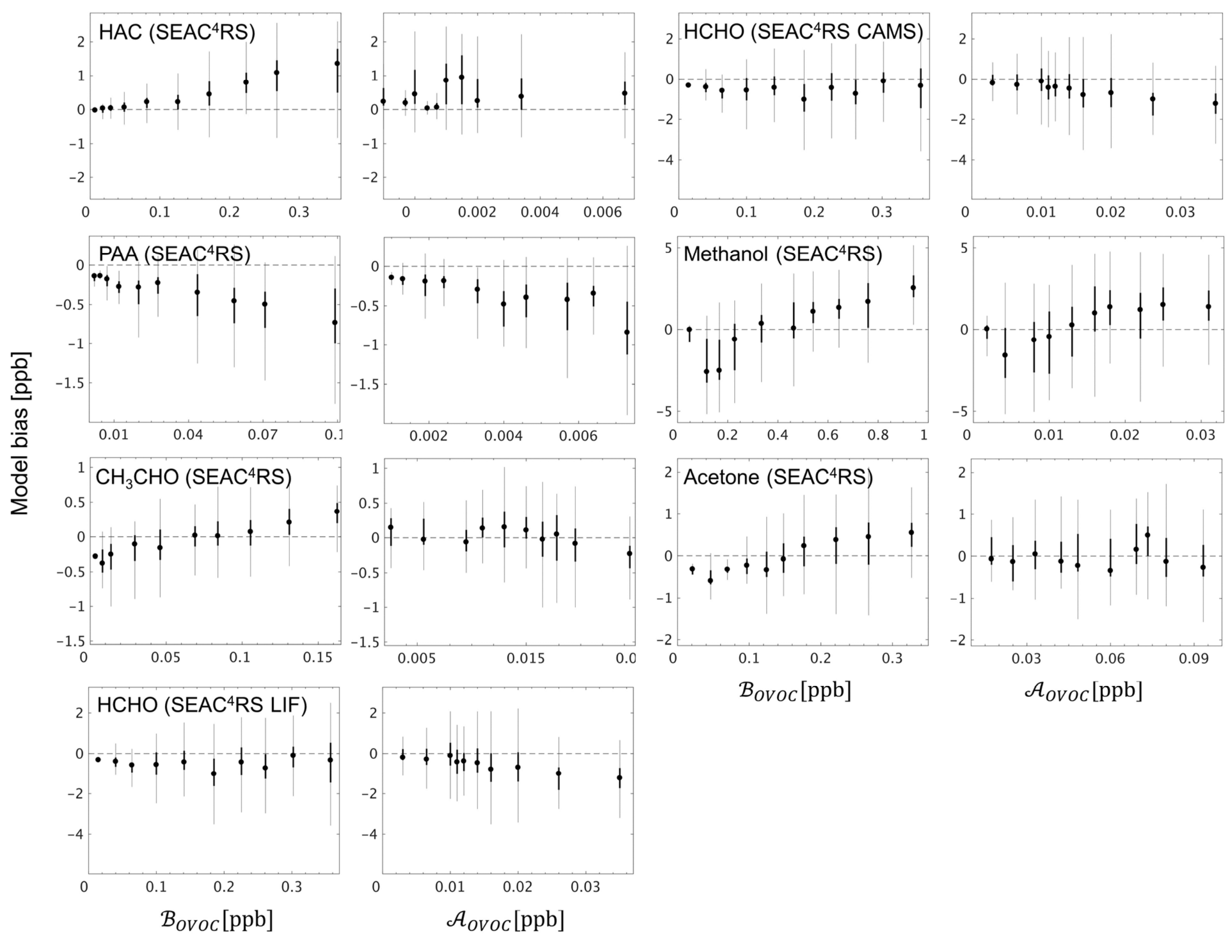

Figure S15. Same as Figure 10 but for the SEAC^{4}RS campaign.

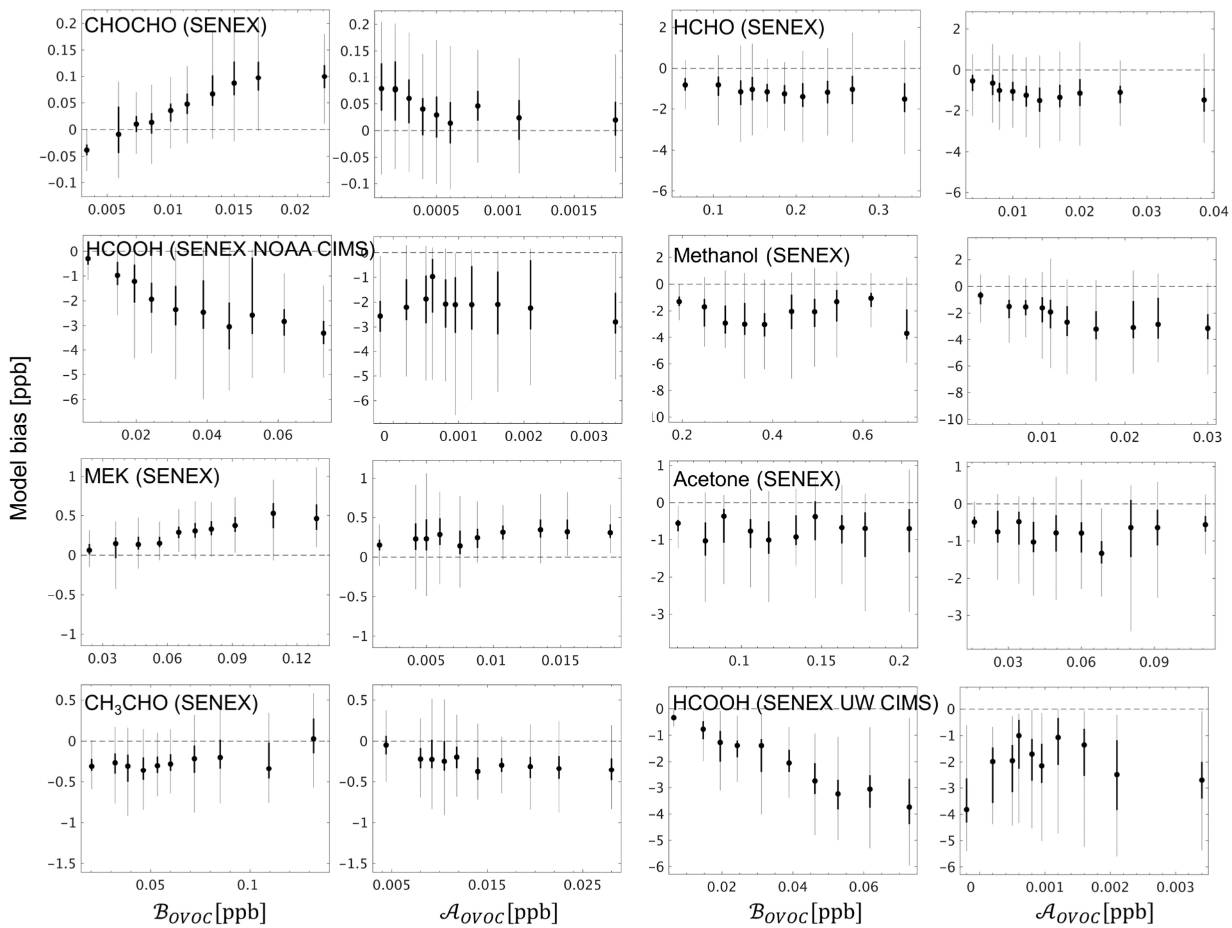

Figure S16. Same as Figure 10 but for the SENEX campaign.

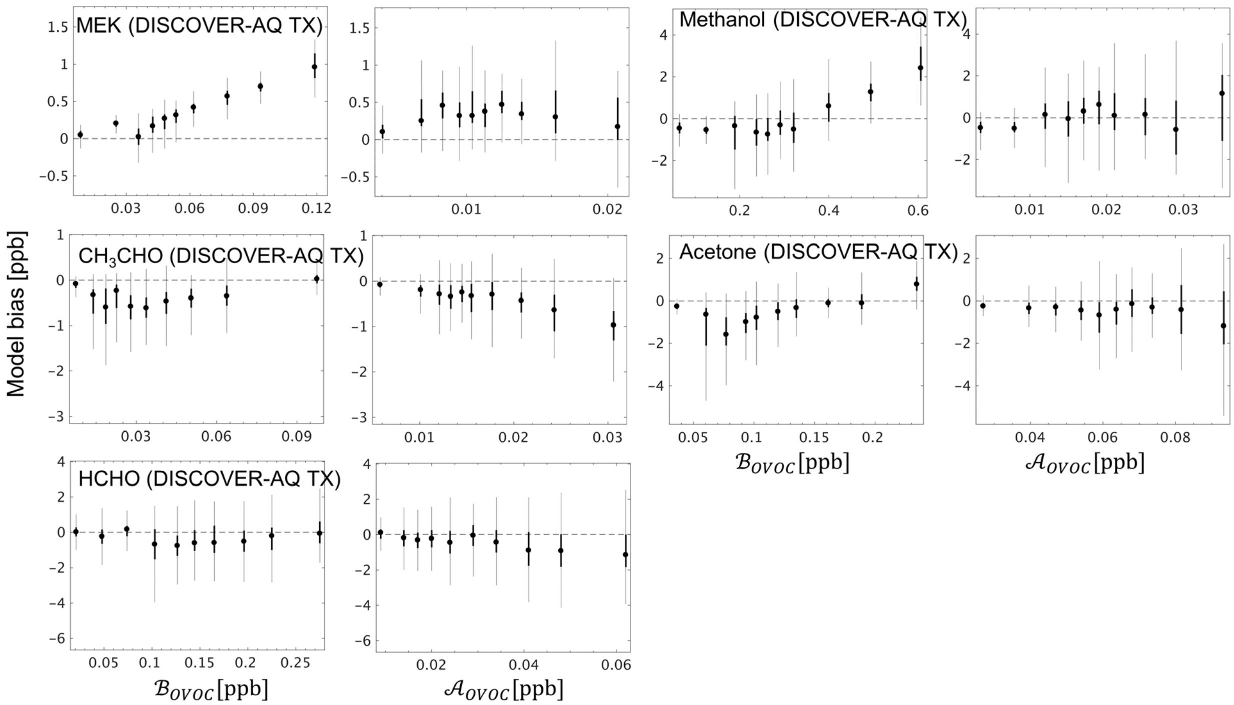

Figure S17. Same as Figure 10 but for the DISCOVER-AQ TX campaign.

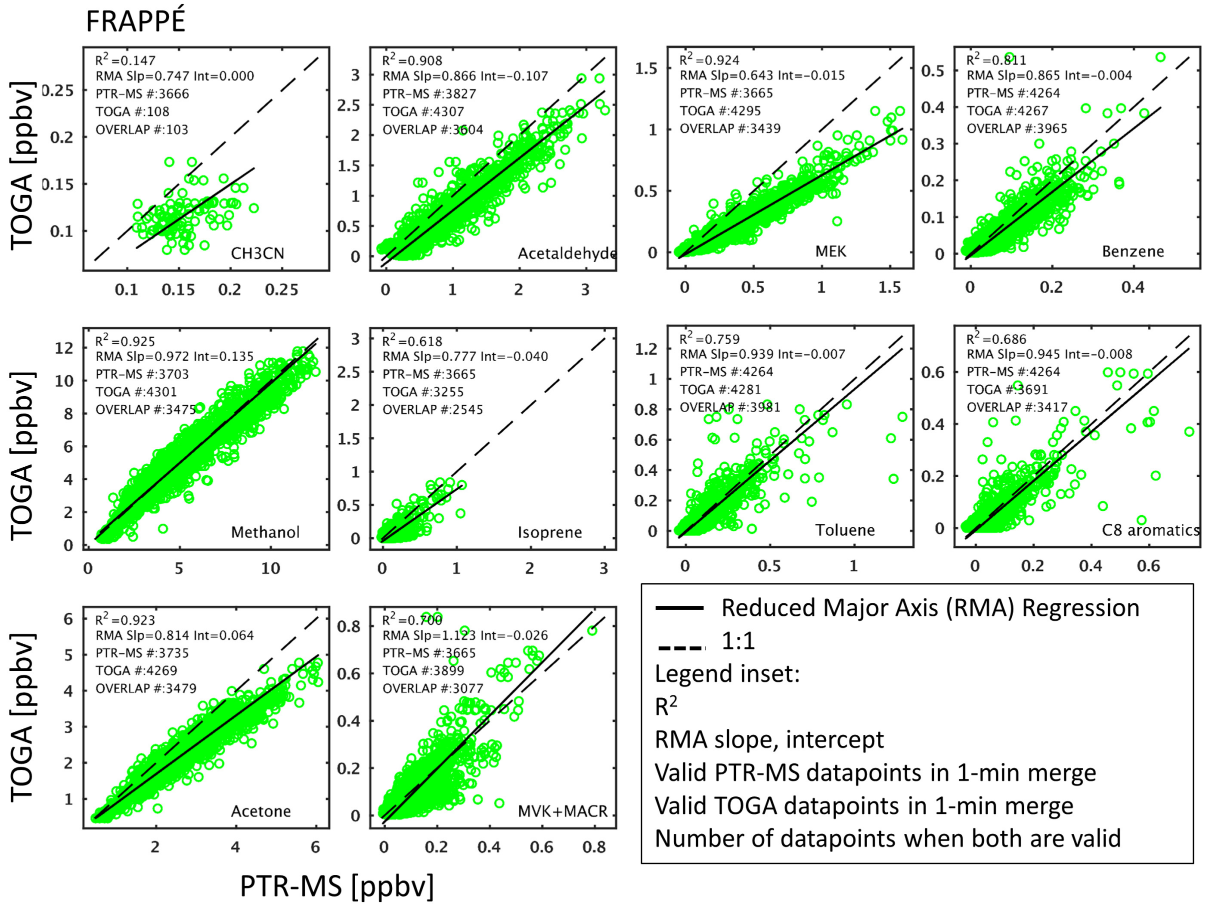

Figure S18. Inter-comparison of co-measured VOCs from FRAPPÉ.

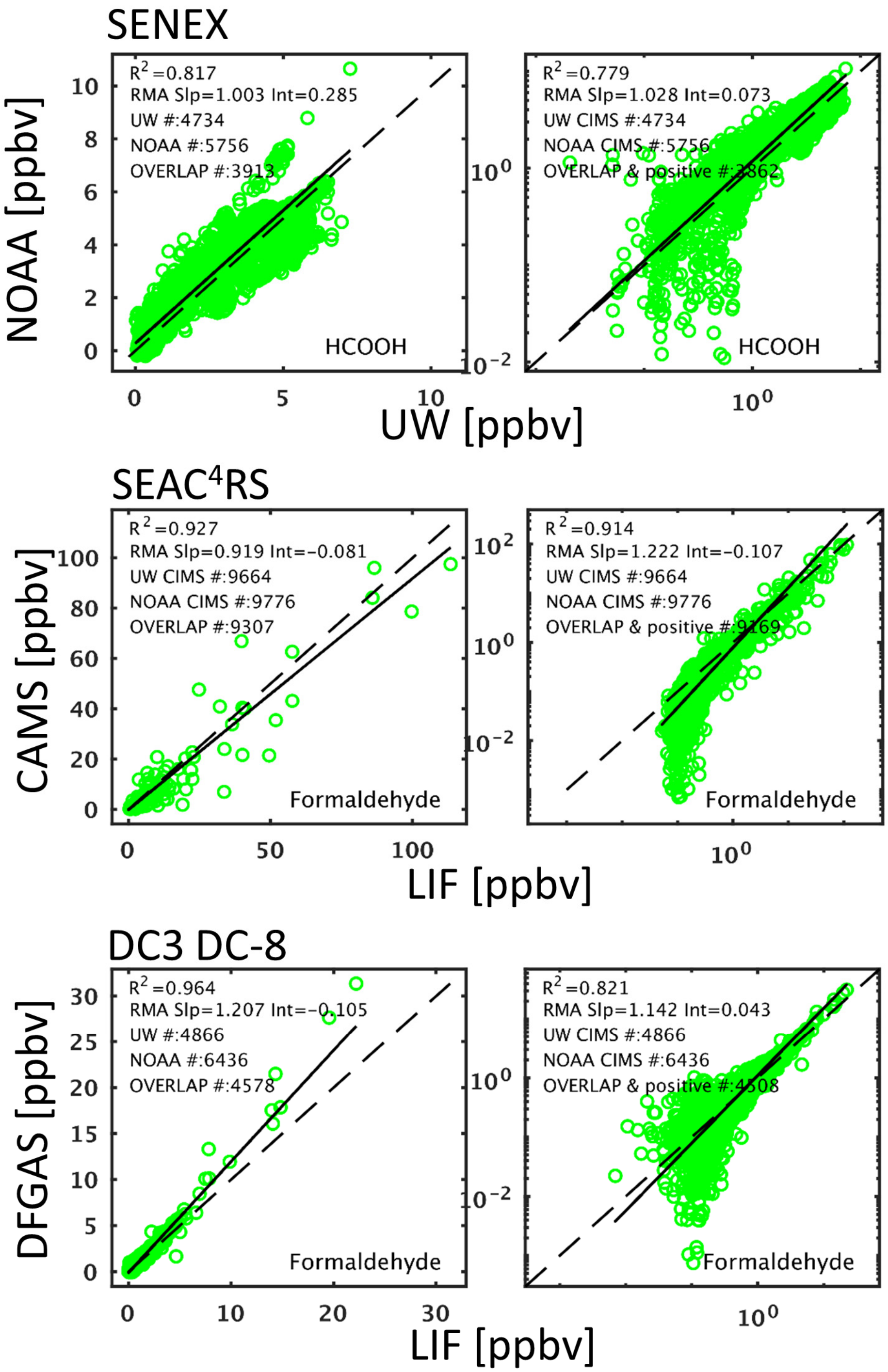

Figure S19. Inter-comparison of concurrent HCOOH measurements during SENEX and of concurrent formaldehyde measurements during DC3 DC-8 and SEAC^{4}RS. See legends in Figure S18.

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