Supplement of Atmos. Chem. Phys., 21, 15153–15170, 2021
https://doi.org/10.5194/acp-21-15153-2021-supplement

Atmospheric Chemistry and Physics
Open Access
EGU

Supplement of

Temporary pause in the growth of atmospheric ethane and propane in 2015–2018

Hélène Angot et al.

Correspondence to: Hélène Angot (helene.angot@epfl.ch)

Table S1: 2008-2010 response factors (RF), along with the associated relative standard deviation (RSD), used to calculate mixing ratios at GEOSummit.

	2008 (Days 150-365)		2009 (Days1-149)		2009 (Days 150-201)		2009 (Days 202-251)		2009 (Days 252-302)		2009 (Days 303-365)	
	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)
ethane	7.6	5%	7.4	3%	7.7	3%	7.6	5%	7.6	5%	7.8	6%
propane	7.2	6%	6.8	2%	7.3	4%	7.1	6%	7.1	7%	7.3	7%
i-butane	6.2		5.5		6.2		6.2		6.1		6.2	
acetylene	6.1		4.5		5.8		6.0		6.6		6.4	
n-butane	6.5		6.0		6.6		6.5		6.4		6.5	
i-pentane	4.5	1%	4.2	5%	4.4	4%	4.8		4.4	1%	4.3	1%
n-pentane	4.8	1%	4.9	1%	4.6	3%	5.1	1%	4.6	4%	4.6	5%
hexane	1.3		2.8		2.1		2.5		2.0		1.9	
benzene	2.7	10%	4.5	13%	2.6	13%	3.0	9%	2.5	6%	2.4	5%
toluene	1.4	2%	2.1	16%	1.3		1.6	6%	1.4	4%	1.2	1%

	2010 (Days1-149)		2010 (Days1-49)		2010 (Days50-99)		2010 (Days100-149)		2010 (Days150-200)	
	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)
ethane	7.65	8%	7.89	8%	8.00	10%	8.48	15%	7.49	1%
propane	7.38	7%	7.44	10%	7.60	10%	7.80	8%	7.29	1%
i-butane	6.47	5%	6.25		6.56		6.67		6.79	
acetylene			6.08		6.47		6.92		6.43	
n-butane	6.82	5%	6.56		6.86		7.08		7.18	
i-pentane	5.33	15%	4.55	2%	4.91	5%	5.73	14%	6.21	5%
n-pentane	5.90	15%	4.98	8%	5.44	7%	7.19	4%	6.79	8%
hexane	4.69	50%	3.05	45%	3.16	38%	5.90	23%	6.44	31%
benzene	3.52	40%	2.65	6%	1.64	85%	4.62	40%	4.17	48%
toluene	1.71	77%	1.43	7%	0.64	141%	1.83	102%	1.87	119%

Table S2: 2012-2020 monthly response factors (RF), along with the associated relative standard deviation (RSD), calculated from the repeated analysis of two independently prepared and cross-referenced standards in use at any given time.

	January		February		March		April		May		June		July		August		September		October		November		December	
	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)	RF	RSD (%)
2012																								
ethane									7.1	0.7	7.2	0.9	7.1	1.2	7.4	2.3	7.4	2.4	7.4	0.5	7.4	0.9	7.5	0.8
propane									6.8	0.7	6.8	0.8	6.7	1.2	7.0	2.7	7.0	2.9	7.0	0.6	7.0	0.9	7.1	0.7
i-butane									6.3	0.8	6.4	1.4	6.3	1.3	6.5	2.0	6.6	2.2	6.6	0.7	6.4	1.4	6.5	1.7
acetylene									3.3	12.6	4.1	13.2	4.7	8.0	5.2	30.0	5.5	16.0	5.8	13.2	6.0	6.2	5.9	6.0
n-butane									6.8	1.4	6.8	0.6	6.7	1.2	7.2	2.5	7.1	3.8	7.1	0.7	7.2	1.5	7.0	0.6
i-pentane									6.5	1.4	6.6	1.0	6.5	1.7	6.7	2.5	6.6	2.8	6.7	0.5	6.6	1.2	6.7	0.9
n-pentane									6.9	0.8	7.1	0.7	6.9	1.3	7.0	2.2	6.9	2.7	7.0	0.5	6.9	0.9	6.9	1.1
hexane									6.3	1.2	6.3	1.9	6.3	1.5	6.6	1.2	6.3	2.3	6.3	1.5	6.3	2.5	6.3	2.1
benzene									6.9	19.1	6.7	12.5	6.5	12.8	6.7	20.6	6.6	15.6	6.7	9.5	6.5	22.1	6.6	5.4
toluene									6.7	21.1	6.8	19.4	6.6	97.5	6.1	23.2	6.2	62.8	6.4	19.6	6.1	71.4	6.1	14.4
2013																								
ethane	7.4	0.6	7.5	0.7	7.5	0.6	7.4	0.8	7.2	1.3	7.4	0.5	7.5	1.2	7.4	3.0	7.6	1.4	7.5	2.0	7.3	1.9	6.3	2.9
propane	7.0	1.8	7.1	1.5	7.1	0.8	7.1	0.7	7.0	1.3	7.1	0.7	7.1	0.8	7.1	2.8	7.2	1.7	7.2	1.9	6.8	2.3	5.8	3.1
i-butane	6.5	2.4	6.6	1.8	6.6	4.7	6.5	2.5	6.4	4.2	6.7	1.1	6.6	3.2	6.6	1.6	6.6	1.8	6.6	2.9	6.3	1.4	5.3	2.4
acetylene	6.0	1.9	6.4	3.8	6.3	3.7	6.0	5.4	5.9	1.9	5.7	0.4	5.7	5.9	6.1		6.1		6.1		6.1		5.2	
n-butane	6.9		6.9		7.3		7.1		6.8		7.0		6.9		7.0	2.0	7.1	1.7	7.1	2.3	6.8	1.7	5.8	2.6
i-pentane	6.7	2.3	6.7	0.8	6.7	2.0	6.6	1.4	6.6	0.6	6.7	0.6	6.6	1.9	6.6	2.1	6.7	1.2	6.6	1.7	6.3	2.5	5.3	2.8
n-pentane	6.8	2.1	6.9	2.6	6.9	3.6	6.9	1.9	6.7	2.0	6.8	0.1	6.9	2.5	6.9	3.3	6.9	1.2	6.8	2.9	6.6	2.3	5.5	2.9
hexane	6.3	39.4	6.3	0.0	6.3	5.3	6.3	5.2	6.6	3.4	6.6	11.1	6.7	3.8	6.6	1.5	6.7	1.4	6.5	3.4	6.2	1.3	5.2	3.3
benzene	6.6	4.3	6.7	1.4	6.6	1.6	6.6	1.7	6.6	1.3	7.0	1.2	6.8	1.7	6.7	1.1	6.8	4.5	6.6	2.6	6.4	10.2	4.9	11.1
toluene	6.0	3.9	6.4	1.3	6.2	2.4	6.1	2.8	6.1	3.1	6.2	2.6	6.2	3.3	6.2	3.3	6.2	1.2	6.0	2.9	6.5	9.6	4.7	11.4
2014																								
ethane	6.4	1.0	6.3	0.5							7.6	1.7	7.5	0.8	7.6	3.0	7.7	3.1	7.6	1.1	7.7	3.8	7.7	1.9
propane	5.9	1.5	5.9	0.6							7.3	0.6	7.1	0.9	7.2	2.7	7.5	2.4	7.4	1.2	7.3	3.9	7.4	3.6
i-butane	5.3	1.0	5.3	0.7							6.7	4.3	6.5	0.8	6.8	5.5	7.0	3.2	7.0	0.1	6.8	3.5	6.8	4.6
acetylene	5.2		5.2										7.2	2.6	8.0	7.6	7.5	1.9	7.9	7.4	7.5	5.0	7.3	4.0
n-butane	5.9	1.2	5.8	0.7							7.3	2.9	7.2	1.2	7.2	3.6	7.5	1.9	7.4	7.0	7.0	2.3	7.1	3.4
i-pentane	5.4	1.4	5.4	0.3							6.8	2.2	6.7	0.7	6.8	4.4	7.1	1.6	7.1	1.8	6.8	4.8	6.9	2.1
n-pentane	5.5	1.6	5.6	1.7							7.1	1.4	7.0	1.1	7.1	3.0	7.3	0.9	7.3	2.3	7.1	2.7	7.2	2.8
hexane	5.1	1.0	5.3	1.5							6.8	1.3	6.8	0.7	6.9	3.1	7.1	0.7	7.0	1.4	6.9	0.7	6.9	2.1
benzene	5.0	2.0	4.9	1.2							7.0	0.7	6.8	0.3	7.0	3.5	7.3	1.0	7.2	1.7	7.1	2.3	7.0	1.2
toluene	4.5	1.2	4.4	1.5							6.3	1.2	6.2	0.7	6.3	2.8					6.5	2.2	6.5	1.6
2015																								
ethane	7.7	2.6	7.7	1.8	7.7	1.0	7.8	1.1	7.7	0.8	7.6	1.0	7.5	0.5	7.7	2.0	7.7	1.1	7.6	1.0	7.8	0.6	7.7	0.8
propane	7.3	2.3	7.4	1.4	7.4	1.4	7.4	1.6	7.4	1.2	7.3	1.1	7.2	0.5	7.3	1.9	7.2	0.5	7.2	0.8	7.2	1.1	7.2	0.5
i-butane	6.7	3.4	6.8	2.4	6.8	2.1	6.8	1.7	6.9	1.7	6.9	3.1	6.6	3.2	6.9	4.7	6.3	3.4	6.6	1.0	6.5	2.2	6.6	1.6
acetylene	7.6	4.8	7.6	2.7	7.8	1.5	7.6	2.1	6.8	1.5	6.7	4.1	6.1	5.8	5.5	7.4	5.7	4.6	5.8	3.0	5.3	2.4	5.4	1.4
n-butane	7.1	3.4	7.2	3.0	7.2	3.9	7.3	0.9	7.5		7.2		7.0		7.2		6.9		7.1		7.0		7.0	
i-pentane	6.9	1.8	6.9	2.3	7.0	1.6	7.0	1.9	7.1	2.8	7.0	2.1	6.9	1.8	7.0	2.9	6.7	3.6	6.8	1.1	6.5	1.2	6.5	0.9
n-pentane	7.1	2.9	7.2	2.4	7.2	1.7	7.2	1.4	7.2	3.2	7.2	2.4	7.1	1.8	7.1	2.5	6.6	2.7	6.8	1.7	6.6	0.5	6.7	1.8
hexane	6.9	1.8	6.9	1.6	7.0	1.1	7.0	1.0	6.9	1.9	6.8	1.3	6.6	0.5	6.7	3.1	5.6	4.8	6.3	3.8	6.3	2.1	6.4	2.1
benzene	7.0	2.7	7.1	1.1	7.1	1.7	7.2	1.1	7.2	3.2	7.0	2.7	7.3	1.5	7.3	4.2	6.6	3.0	7.0	2.8	6.6	1.2	7.0	0.9
toluene	6.5	1.6	6.5	1.9	6.6	2.0	6.5	0.9	6.9	5.4	6.7	3.7	6.2	2.9	6.1	7.3	4.7	6.4	5.4	6.6	5.9	2.0	5.9	3.6
2016																								
ethane	7.7	0.8	7.7	0.5	7.6	0.9	7.6	0.9	7.8	0.7	7.4	0.7	7.4	1.2	7.3	1.3	7.2	0.6	7.2	0.5	7.2	0.9	7.2	0.9
propane	7.3	0.7	7.3	0.8	7.2	1.7	7.2	1.2	7.3	0.9	7.3	0.7	7.3	0.8	7.3	1.9	7.3	0.9	7.4	0.6	7.3	0.8	7.4	0.8
i-butane	6.5	2.0	6.7	0.8	6.6	1.4	6.6	2.0	6.6	1.2	6.4	3.3	6.5	2.0	6.6	3.2	6.7	0.8	6.7	1.0	6.6	2.4	6.7	2.4
acetylene	6.2	1.7	6.3	3.5	6.0	2.6	6.1	2.7	5.7	2.1	5.9	22.3	5.9	2.8	5.6	7.5	5.9	4.5	6.1	2.6	6.4	1.5	6.4	2.4
n-butane	7.1		7.1		6.8		7.1		7.0		6.9		7.0		7.0		7.2		7.3		7.2		7.0	
i-pentane	6.6	2.0	6.7	1.4	6.6	1.1	6.7	3.1	6.2	3.7	5.9	9.7	6.1	4.3	6.4	3.0	6.8	1.8	6.9	1.3	6.7	1.0	6.8	0.7
n-pentane	6.8	2.2	7.0	1.7	6.6	1.4	6.8	2.8	6.2	5.3	5.7	15.6	6.1	6.5	6.4	3.7	7.0	1.6	7.0	1.2	7.0	1.7	6.8	1.2
hexane	6.5	2.2	6.6	1.1	6.7	2.5	6.4	1.7	5.1	8.0	4.6	57.7	5.6	8.0	6.0	13.4	6.7	1.0	6.8	0.8	6.7	0.7	6.9	1.0
benzene	7.2	1.7	7.1	0.4	7.0	1.6	7.0	0.9	5.5	9.3	5.2	9.8	6.3	10.7	7.3	11.8	7.5	2.2	7.4	0.6	7.3	0.8	7.3	0.8
toluene	6.8	7.3	6.5	2.7	5.9	3.4	5.8	3.1	4.5	12.9	5.2	38.4	4.6	2.2	5.5	5.5	6.6	3.1	6.3	2.3	6.1	2.7	6.0	2.6

(continued)

2017																				
ethane	7.2	1.1	7.2	0.4	7.2	0.9	7.2	0.7	7.2	0.9	7.3	0.7	7.2	0.5	7.2	0.7			7.5	0.6	7.5	0.8
propane	7.4	1.5	7.4	1.1	7.4	1.0	7.4	0.6	7.4	0.9	7.5	1.0	7.5	0.6	7.5	0.9			7.0	0.7	7.0	0.8
i-butane	6.7	2.0	6.6	0.8	6.7	1.0	6.7	0.6	6.6	1.2	6.7	0.7	6.6	0.9	6.7	1.5			6.7	1.3	6.7	1.1
acetylene	6.4	3.3	6.3	2.2	6.6	1.0	6.6	1.3	6.5	1.6	6.5	0.8	6.6	1.2	6.6	1.7			6.7	1.2	6.7	1.1
n-butane	7.3		7.2		7.3		7.3		7.3		7.3		7.3		7.3				7.1		7.1	
i-pentane	6.7	1.2	6.7	1.4	6.8	0.8	6.8	1.5	6.8	1.6	6.9	2.5	6.9	2.8	6.9	1.9			6.8	1.1	6.8	1.1
n-pentane	7.0	0.9	6.9	1.2	7.0	0.9	7.0	1.7	6.9	1.5	6.8	2.0	7.0	1.9	6.9	0.4			6.9	1.1	6.9	1.7
hexane	6.7	1.7	6.6	1.6	6.6	0.8	6.6	1.3	6.6	1.4	6.7	1.6	6.6	2.0	6.6	1.3			6.5	1.7	6.5	0.6
benzene	7.3	1.2	7.2	0.8	7.2	0.8	7.2	1.0	7.2	0.9	7.2	0.8	7.2	0.8	7.2	0.9			7.6	0.8	7.6	0.6
toluene	5.9	1.9	5.9	1.6	5.9	1.2	5.8	1.5	6.4	10.1	6.1	2.0	5.9	1.8	5.8	1.0			5.7	2.4	5.7	1.5
2018																				
ethane	7.5	0.7	7.4	0.8	7.4	0.8	7.4	1.1	7.3	0.5	7.3	0.7	7.0	1.4	7.1	0.8	7.1	0.4	7.3	1.6	7.2	1.3	7.4	1.9
propane	7.1	0.7	7.0	0.8	7.0	0.5	6.9	1.0	6.8	0.7	6.9	0.9	6.6	0.5	6.7	1.1	6.7	0.6	6.8	0.8	6.7	0.9	6.8	1.0
i-butane	6.7	1.0	6.7	0.7	6.6	1.2	6.6	1.1	6.5	0.8	6.5	1.2	6.3	1.2	6.4	1.0	6.3	1.1	6.4	1.3	6.3	2.0	6.4	1.5
acetylene	6.8	1.3	6.5	1.6	6.3	2.5	6.3	2.1	6.4	2.1	6.2	2.3	6.5	1.3	6.8	1.3	6.7	2.4	6.5	2.9	6.4	2.4	6.5	2.9
n-butane	7.1		7.0		7.0		7.0		6.8		6.9		6.6		6.7		6.6		6.9		6.4		6.6	
i-pentane	6.8	0.8	6.7	0.9	6.7	2.0	6.8	2.2	6.7	2.7	6.6	2.2	6.4	0.8	6.5	1.1	6.5	1.1	6.5	1.5	6.4	1.1	6.6	1.8
n-pentane	6.9	1.7	6.8	0.7	6.7	4.0	6.8	3.4	6.7	4.1	6.7	2.6	6.4	0.9	6.5	2.9	6.5	1.4	6.5	1.4	6.2	1.5	6.4	1.8
hexane	6.5	1.2	6.4	0.9	6.4	2.3	6.5	1.8	6.4	2.4	6.3	0.8	5.9	1.1	6.1	1.2	6.1	0.8	6.1	1.4	6.3	1.5	6.4	1.7
benzene	7.5	0.9	7.4	0.8	7.4	0.7	7.4	1.0	7.4	0.8	7.3	1.0	6.5	1.0	6.8	1.7	6.6	0.7	6.7	1.7	6.6	1.0	6.8	1.3
toluene	5.6	1.4	5.5	1.1	5.4	2.4	5.6	2.3	5.5	1.0	5.5	0.9	4.4	4.5	4.7	5.0	4.5	1.8	4.2	7.3	4.4	5.6	4.3	7.3
2019																				
ethane	7.5	1.2	7.4	1.8	7.5	0.9	7.4	0.0	7.4	1.4	7.4	0.8	7.4	0.8	7.5	1.7	7.0	3.8	7.0	2.0	7.0	2.1	7.0	16.1
propane	6.9	0.7	6.9	1.9	7.0	1.1	6.9	0.5	7.0	1.1	6.9	1.4	6.9	0.9	7.0	0.7	6.8	1.2	6.8	1.0	6.8	1.7	6.8	16.1
i-butane	6.6	1.4	6.5	2.1	6.7	1.1	6.6	1.5	6.7	1.7	6.6	2.5	6.6	1.1	6.7	1.2	7.0	1.8	7.0	1.6	7.0	2.1	7.1	16.3
acetylene	6.5	2.0	6.9	4.2	6.9	3.6	7.1	2.8	7.0	1.9	7.2	2.3	7.3	1.5	7.4	2.7	6.2	4.5	6.1	2.0	6.2	3.4	6.0	15.7
n-butane	6.9		7.2		7.2		7.1		6.8		6.8		6.9		6.9		6.9		6.9		6.9		6.9	
i-pentane	6.7	1.5	6.7	2.2	6.7	0.9	6.7	1.2	6.8	1.4	6.7	1.9	6.7	0.8	6.7	1.9	7.0	1.2	7.0	1.6	7.0	2.4	7.0	15.2
n-pentane	6.7	1.7	6.8	3.5	6.8	1.4	6.8	1.1	6.8	1.7	6.8	1.8	6.7	1.7	6.6	3.8	6.3	1.7	6.2	2.8	6.2	2.3	6.3	15.4
hexane	6.3	3.2	6.4	0.8	6.4	0.7	6.4	1.6	6.4	1.6	6.3	2.0	6.2	2.2	6.0	2.8	5.9	1.9	6.1	1.2	6.0	2.4	5.8	16.6
benzene	6.9	1.2	7.0	5.3	7.1	2.0	7.1	1.3	7.0	1.1	6.8	1.7	6.8	1.3	6.6	2.5	6.1	2.3	5.7	2.0	5.6	2.0	5.5	16.3
toluene	4.5	6.0	4.7	2.5	4.7	4.4	5.0	4.3	5.0	3.9	4.9	5.3	4.5	6.7	3.7	7.4	5.0	6.7	4.8	3.0	4.6	5.1	4.0	16.7
2020																				
ethane	7.5	0.9	7.4	2.9	7.5	14.5														
propane	6.9	0.8	6.9	2.1	7.0	14.5														
i-butane	6.6	1.1	6.5	1.5	6.7	13.7														
acetylene	6.5	3.5	6.9	4.8	6.9	14.5														
n-butane	6.9		7.2		7.2															
i-pentane	6.7	1.6	6.7	1.3	6.7	12.9														
n-pentane	6.7	1.5	6.8	2.1	6.8	13.3														
hexane	6.3	3.5	6.4	3.6	6.4	14.2														
benzene	6.9	3.4	7.0	4.1	7.1	14.2														
toluene	4.5	9.7	4.7	9.8	4.7	15.4														

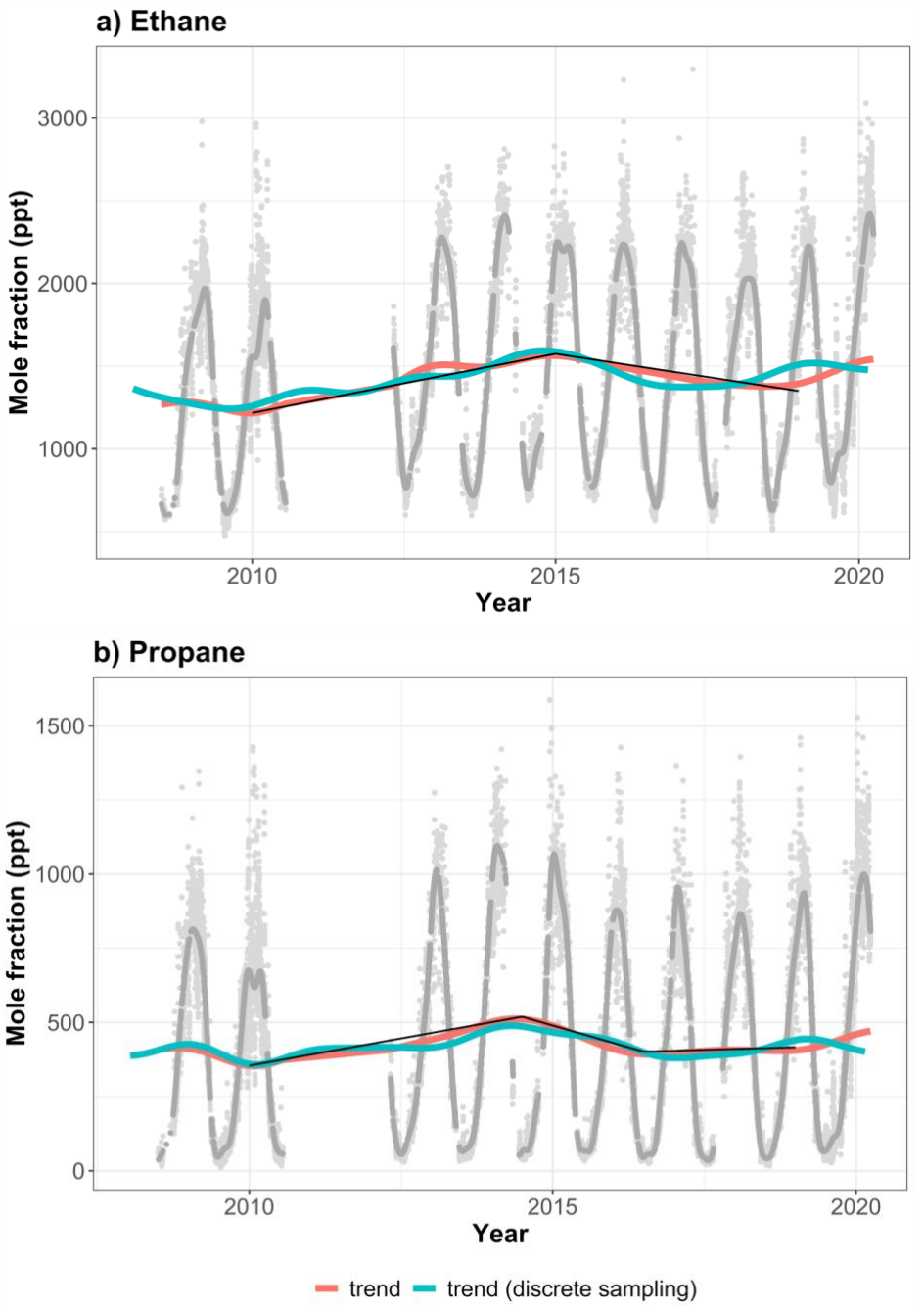

Figure S1: a) Ethane, and b) propane time-series at GEOSummit from July 2008 to March 2020. Individual data points are shown in light grey, smoothed fit in dark grey, and trends in salmon. Trends inferred from discrete flask sampling are shown in turquoise. The slopes given by the Theil-Sen test fro trend analysis are shown in black.

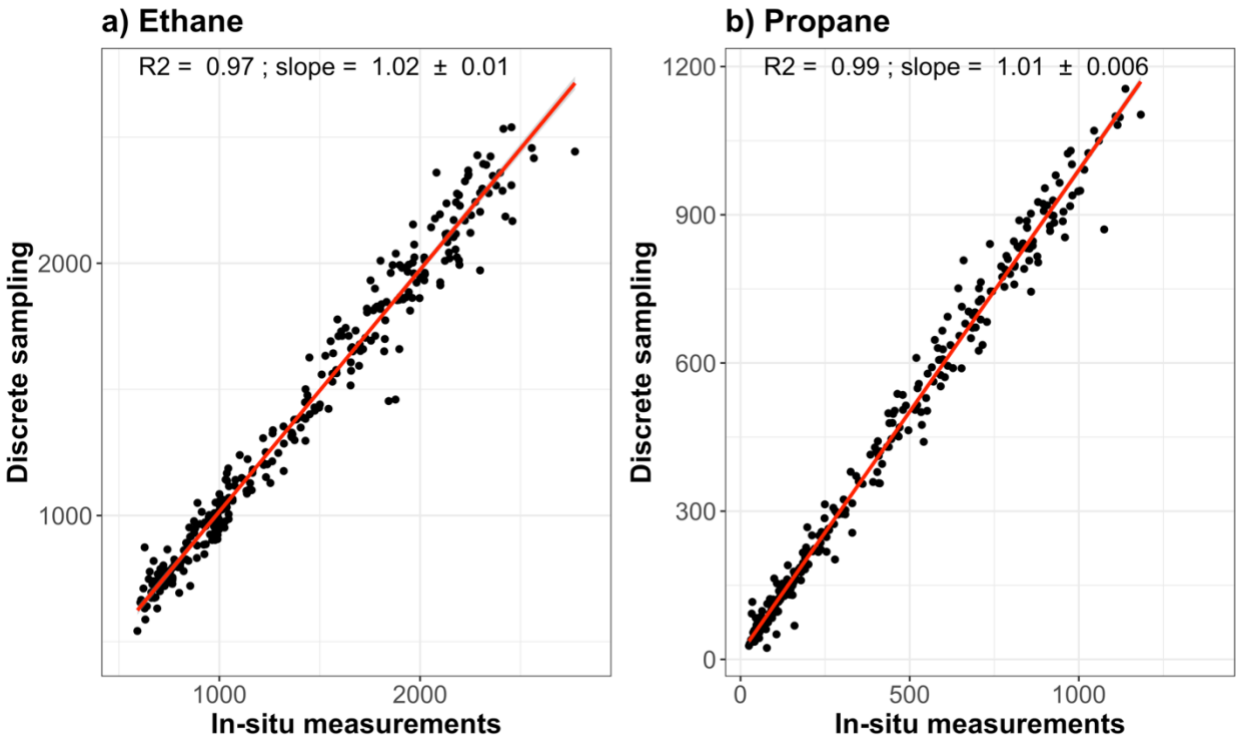

Figure S2: Scatter plot of daily averaged in-situ GC-FID measurements (x-axis) and discrete samples (y-axis) for **a)** ethane and **b)** propane. The red line gives the fitted linear regression. The slope (±95 % confidence interval) and correlation coefficient are given at the top. Note that these are not the same samples being compared: discrete samples are 5 minute-snapshots while in-situ GC-FID measurements are daily averages of 6-12 20 minute-samples.

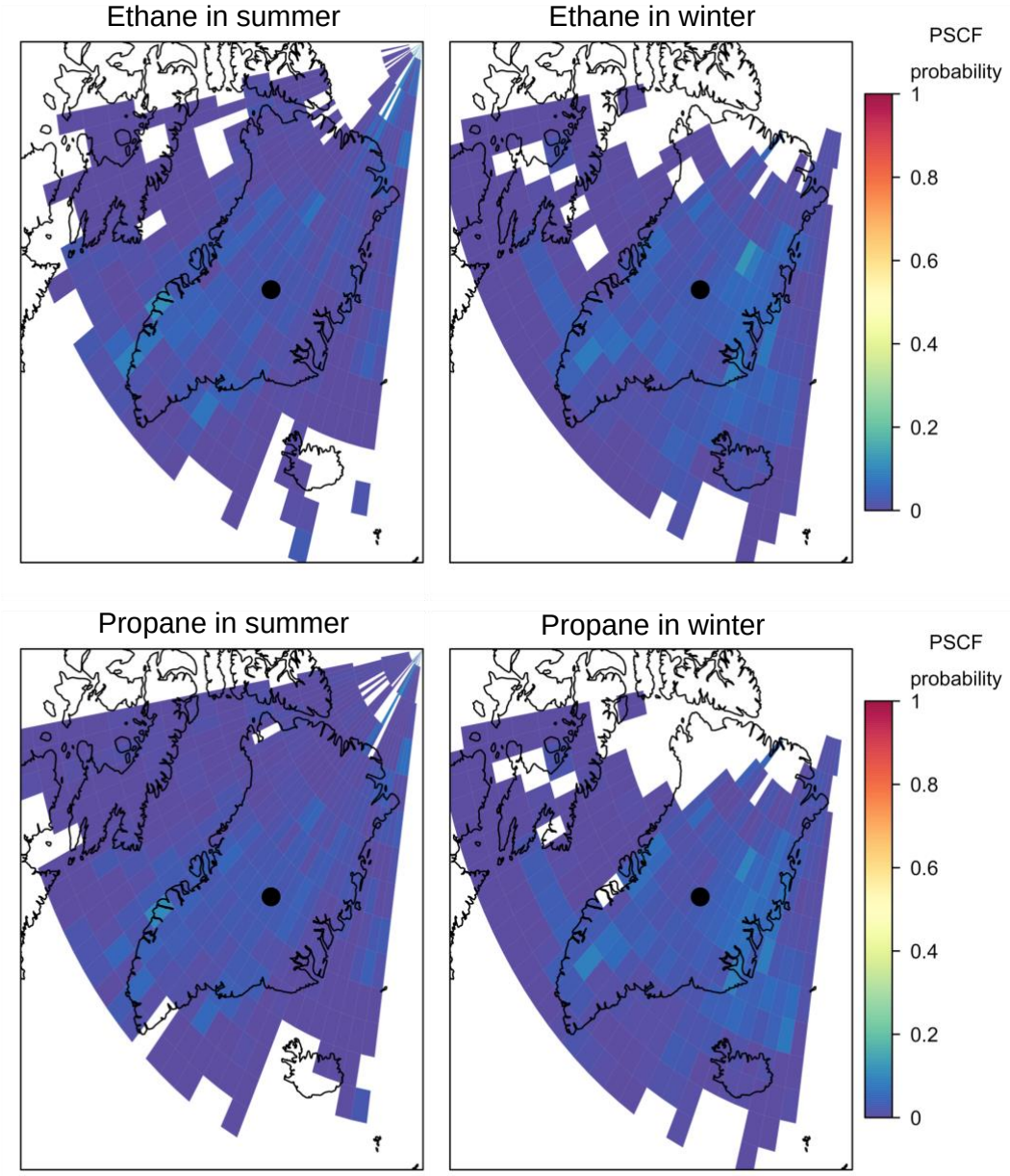

Figure S3: PSCF results for ethane and propane residuals greater than the 90th percentile for April 2012-June 2019.

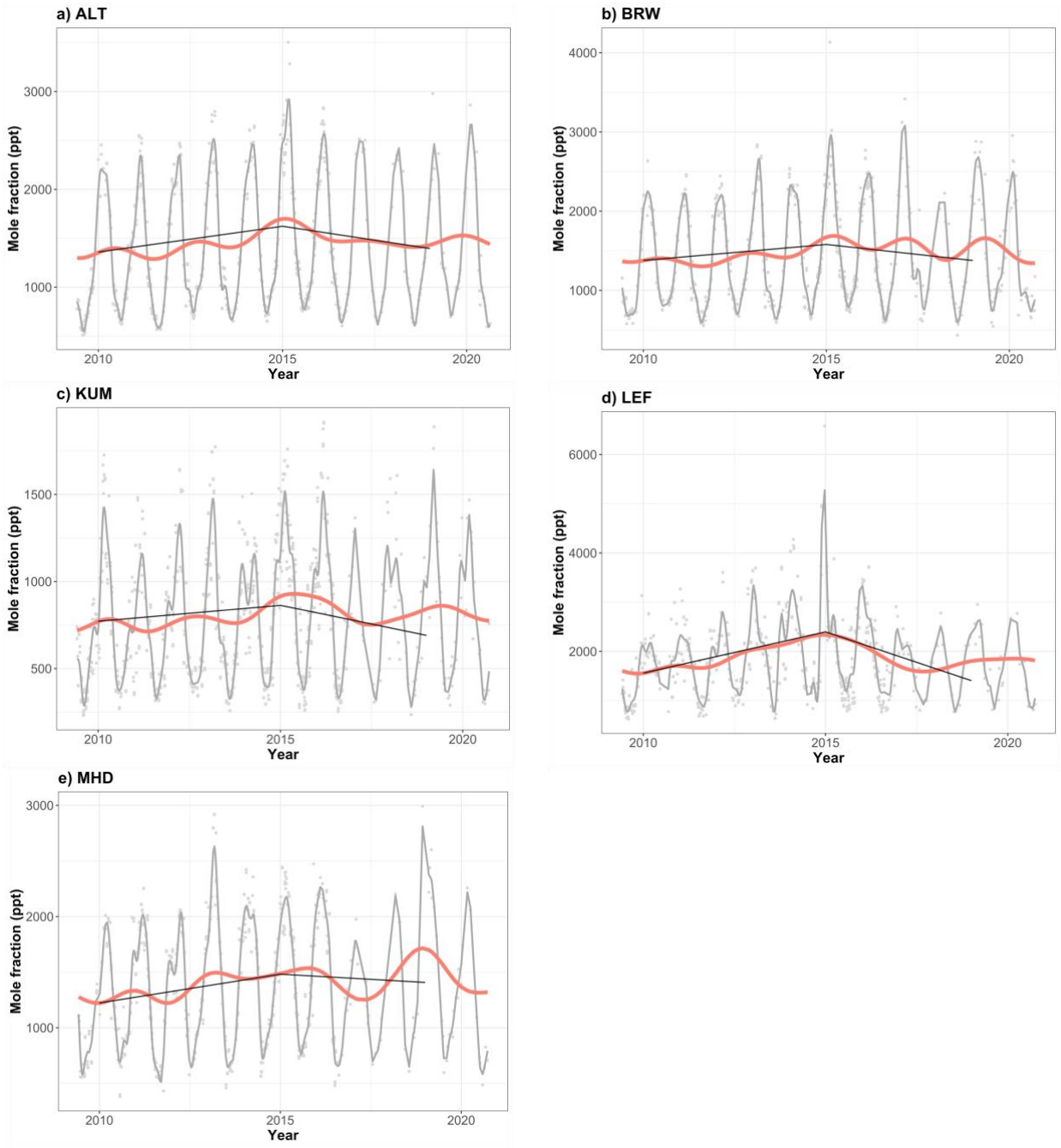

Figure S4: Ethane time-series at a) Alert (ALT), b) Utqiagvik (formerly known as Barrow (BRW)), c) Cape Kumukahi (KUM), **d)** Park Falls (LEF), and **e)** Mace Head (MHD) from June 2009 to March 2020 as inferred from discrete flask sampling. Individual data points are shown in light grey, smoothed fit in dark grey, and trends in salmon. The slopes given by the Theil-Sen test for trend analysis are shown in black (see also Table 1).

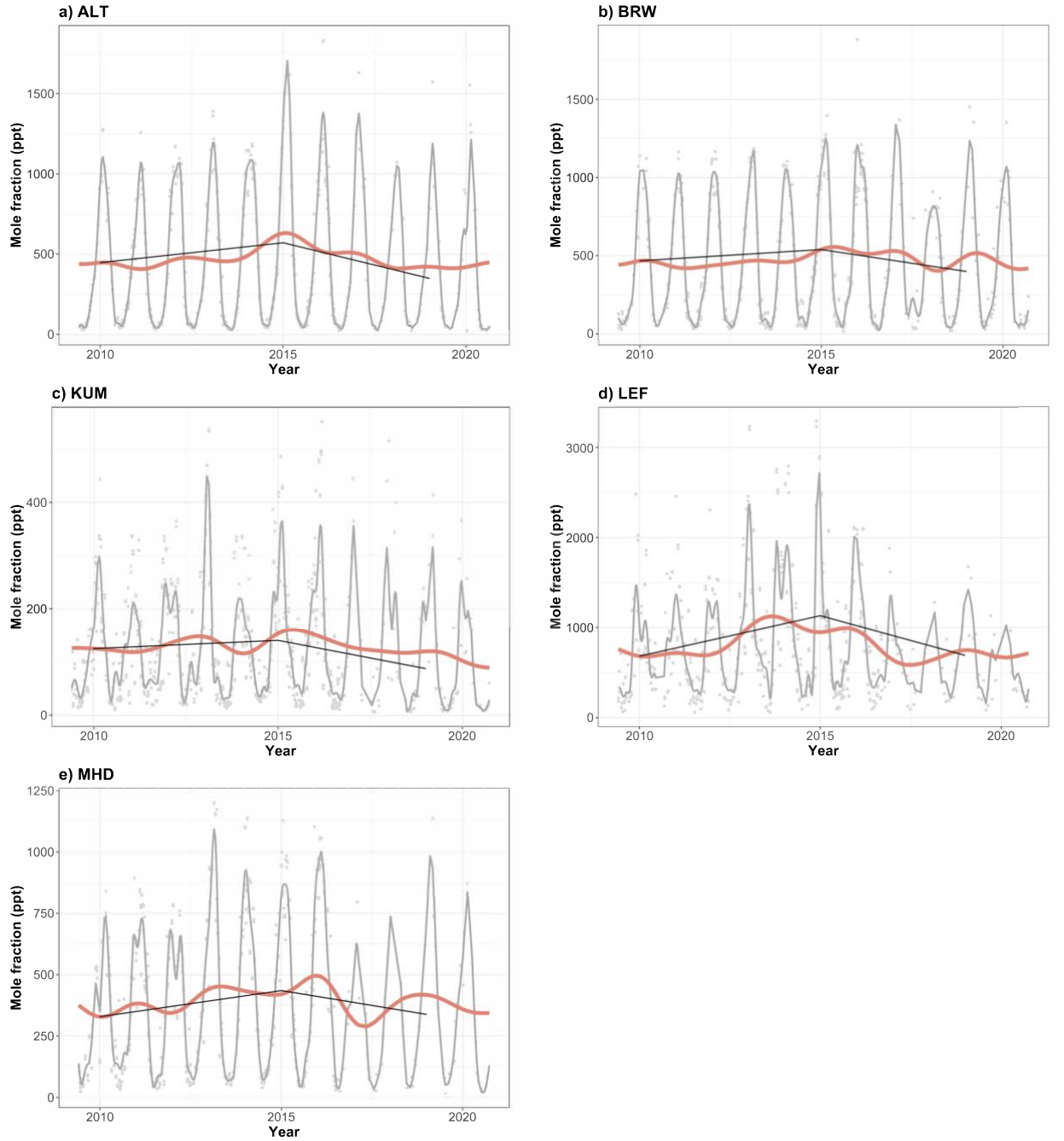

Figure S5: Propane time-series at **a)** Alert (ALT), **b)** Utqiagvik (formerly known as Barrow (BRW)), **c)** Cape Kumukahi (KUM), **d)** Park Falls (LEF), and **e)** Mace Head (MHD) from June 2009 to March 2020 as inferred from discrete flask sampling. Individual data points are shown in light grey, smoothed fit in dark grey, and trends in salmon. The slopes given by the Theil-Sen test for trend analysis are shown in black (see also Table 1).

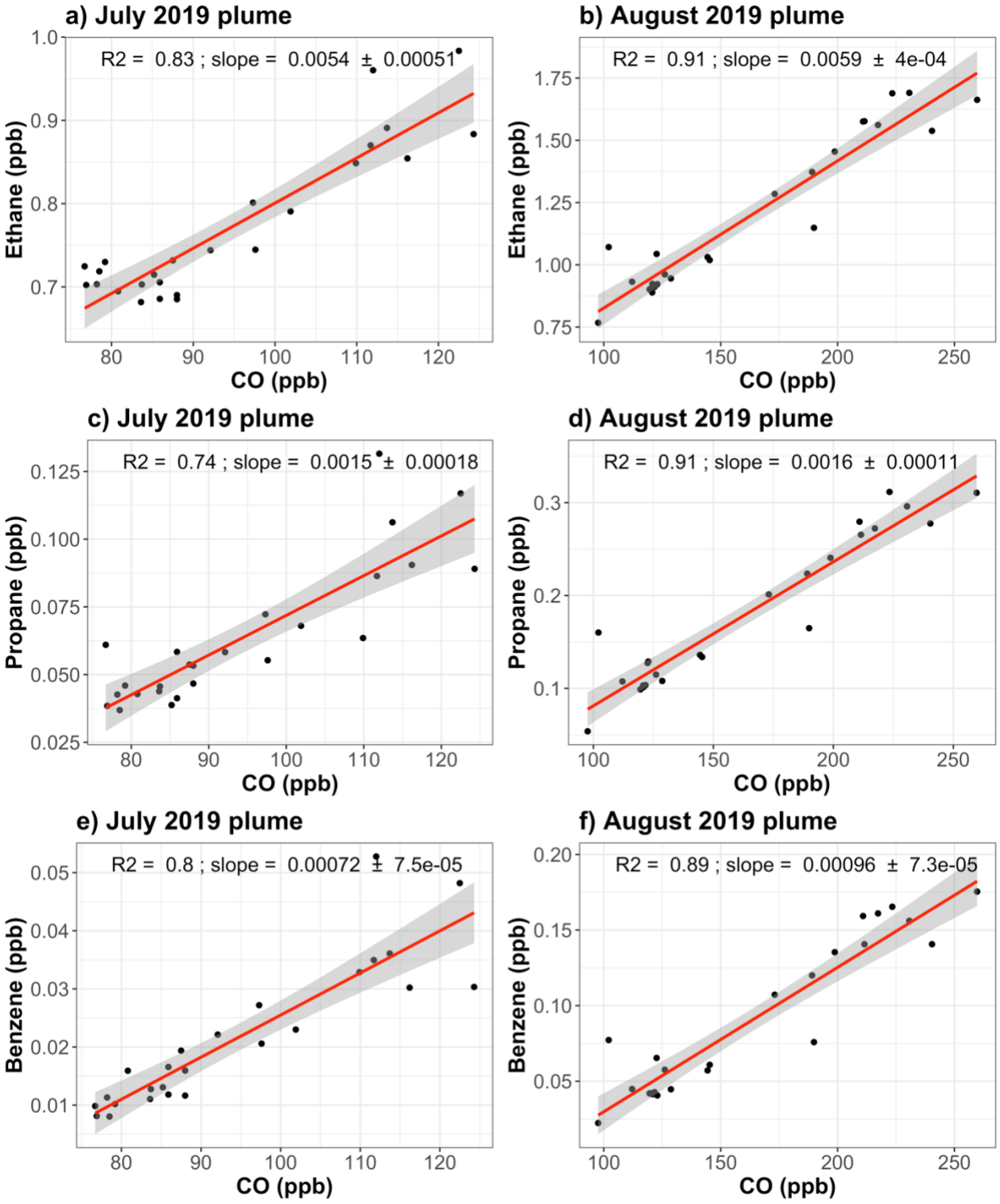

Figure S6: Scatter plot of ethane, propane, and benzene vs. carbon monoxide (CO) mixing ratios in the July and August 2019 biomass burning plumes observed at GEOSummit. The red line gives the fitted linear regression line with the 95 % confidence interval (grey shaded region). The slope, given at the top, gives the emission ratio (amount of compound emitted divided by that of a reference compound (CO here)). This figure was made using data from July 14-23, 2019 and from August 15-23, 2019 for the July and August biomass burning plumes, respectively.

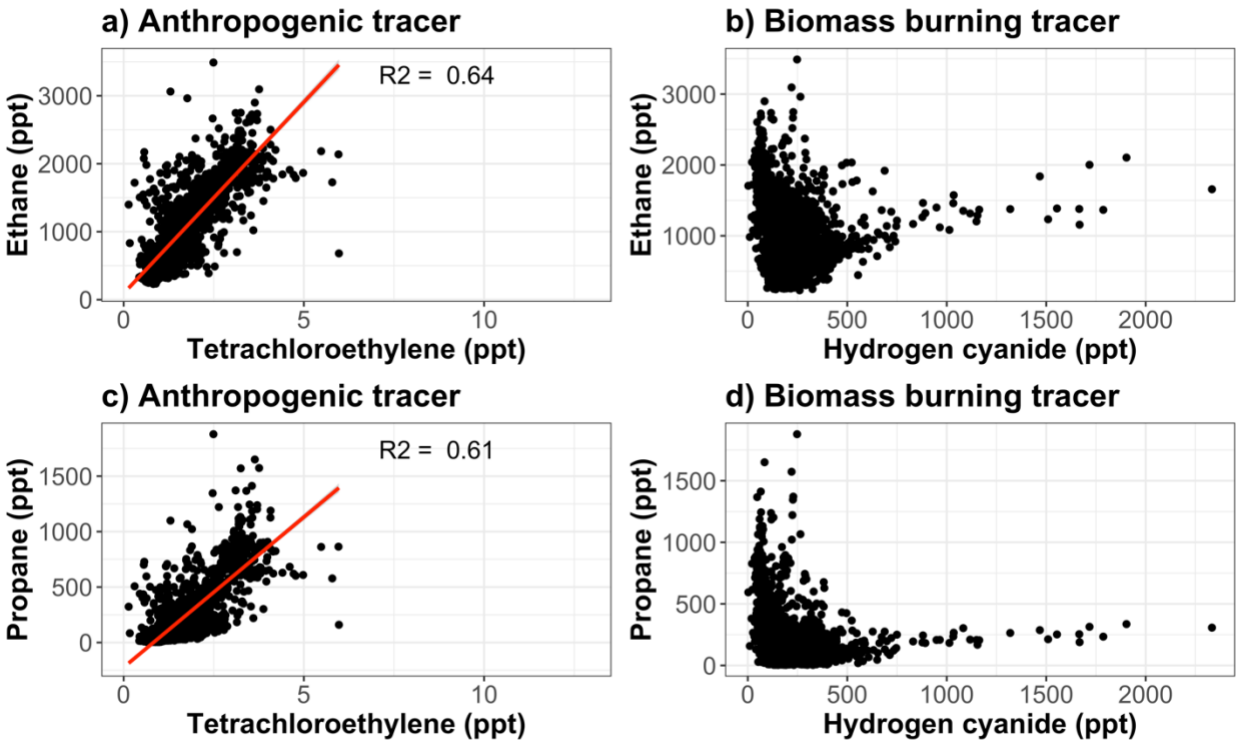

Figure S7: Scatter plot of ethane **(a, b)** and propane **(c, d)** vs. tetrachloroethylene and hydrogen cyanide mixing ratios in the remote free troposphere during the global-scale aircraft mission ATom. Tetrachloroethylene and hydrogen cyanide are used as tracers of anthropogenic and biomass burning emissions, respectively. The red line gives the fitted linear regression line with the 95 % confidence interval (grey shaded region). The correlation coefficient is given at the top.