



Supplement of

Consistent estimates of carbonyl sulfide and methane stratospheric lifetimes retrieved from AirCore profiles at different latitudes

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S1 QCLS operating parameters and achieved precisions

Table S1: QCLS operating parameters and achieved precisions.

Parameter	Value
Temperature	$\sim 298 \pm 0.002$ K
Pressure	$\sim 50 \pm 0.005$ Torr (66 hPa)
Sample mass flow	~ 50 mL min ⁻¹
Measurements frequency	1 Hz
Geometric cell volume	150 cm ³
Effective cell volume	~ 10 cm ³
N ₂ O measurement precision	0.12 ppb
CH ₄ measurement precision	0.6 ppb
CO ₂ measurement precision	0.2 ppm
CO measurement precision	1 ppb
COS measurement precision	20 ppt
H ₂ O measurement precision	20 ppm
O ₃ measurement precision	0.1 ppm

S2 Data selection process for calculating the slope from COS-N₂O correlation

5 In this section, the data selection process for the COS-N₂O slope estimation will be presented for all flights except TRN2, which is reported in the main text.

Table S2: Summary of refined data used for the tracer-tracer regressions after the selection procedure explained in Sect. 3.1.2. For TRN1 (*) no temperature nor humidity data was available and the tropopause was assigned arbitrarily at 11.0 km.

Date/period	Data origin	Thermal tropopause (km)	Altitude range of the selection for the COS-N ₂ O regression (km)	Data points for the COS-N ₂ O regression	Altitude range of the selection for the CH ₄ -N ₂ O regression (km)	Data points for the CH ₄ -N ₂ O regression
17/06/2019	TRN1	11.0 *	17.3–20.9	77	17.3–20.0	62
18/06/2019	TRN2	11.5	18.3–22.2	47	18.3–21.0	36
18/06/2019	TRN3	10.8	17.9–21.5	64	17.9–20.4	48
20/06/2019	TRN4	10.6	17.3–20.6	43	17.3–19.4	31

13/08/2021	KRN-a	10.5	14.9–20.3	96	14.9–19.4	86
13/08/2021	KRN-b	10.5	14.9–20.3	93	14.9–19.4	82
02/08/2023	SOD1	10.9	16.1–20.7	116	16.1–20.0	102
05/08/2023	SOD3-a	9.2	16.8–18.4	22	16.8–18.4	22
05/08/2023	SOD3-b	9.2	16.5–16.5	1	16.5–16.5	1
08/08/2023	SOD5	10.6	16.8–20.9	66	16.8–20.2	53
June to September, 2012–2023	Polar (65°–69° N) average, ACE-FTS	/	15.5–20.5	6	15.5–18.5	4
June to September, 2012–2023	Mid-latitude (45°–49° N) average, ACE-FTS	/	17.5–21.5	5	17.5–20.5	4

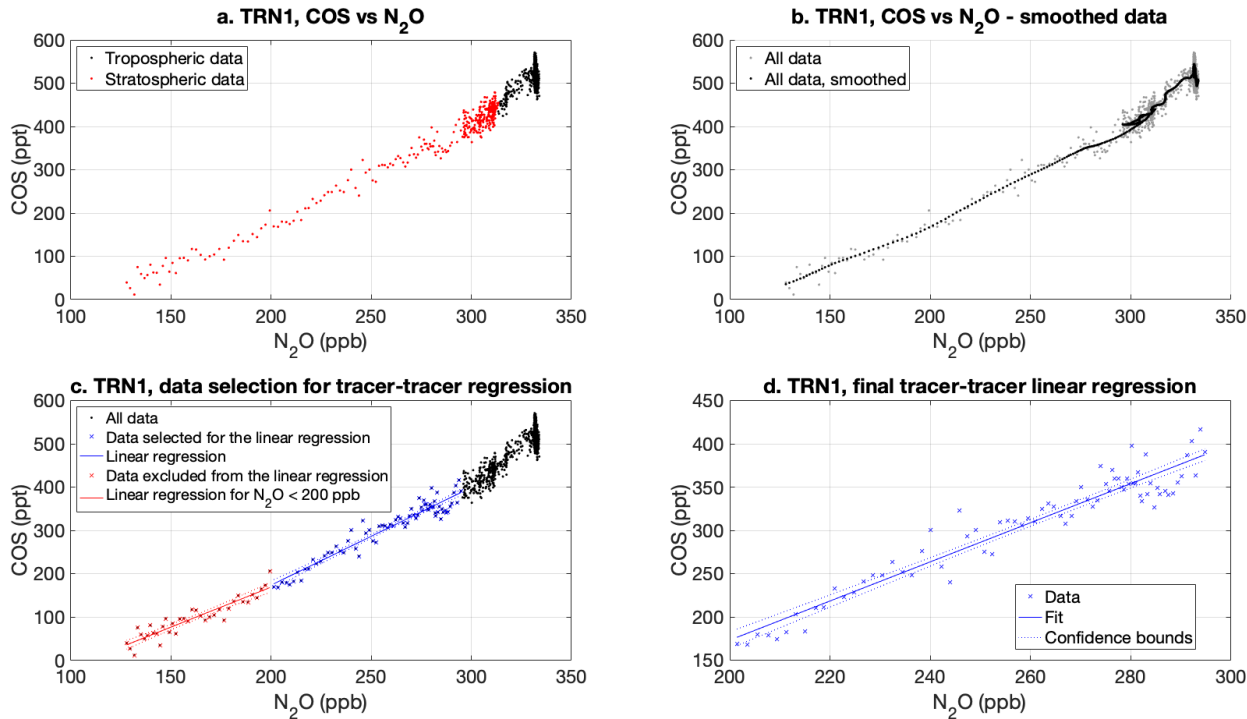


Figure S1: Data selection steps for the linear regression between COS and N_2O , TRN1 AirCore data.

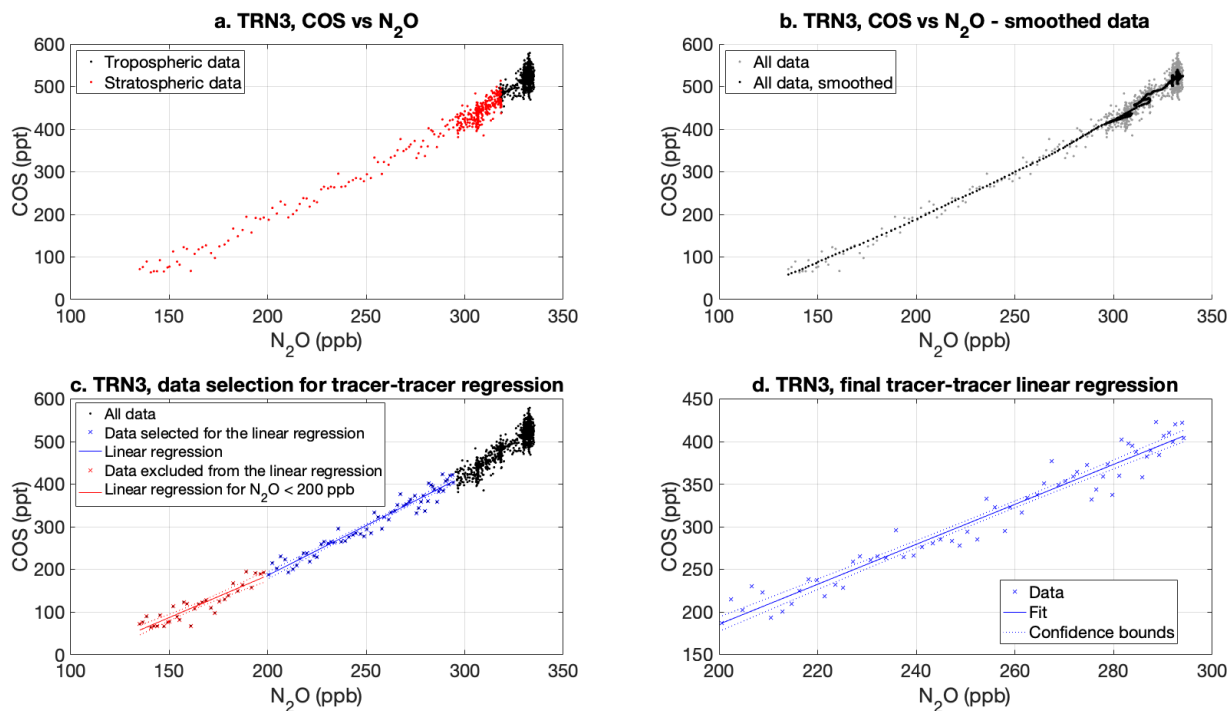


Figure S2: Data selection steps for the linear regression between COS and N_2O , TRN3 AirCore data.

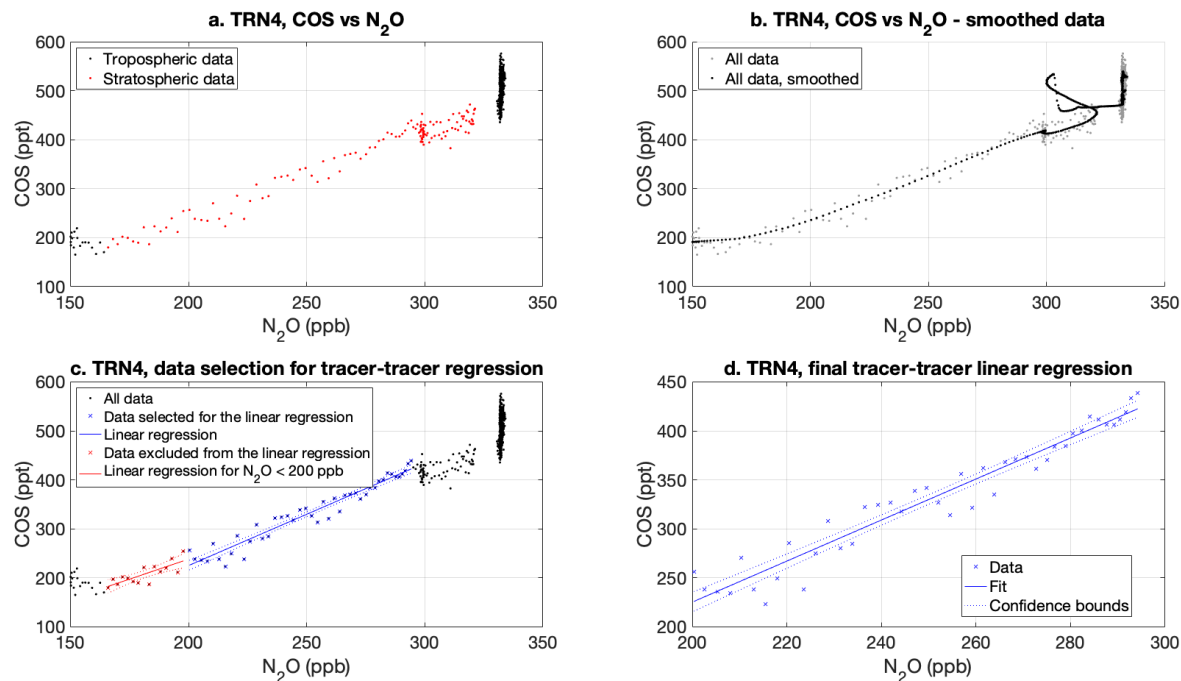


Figure S3: Data selection steps for the linear regression between COS and N_2O , TRN4 AirCore data.

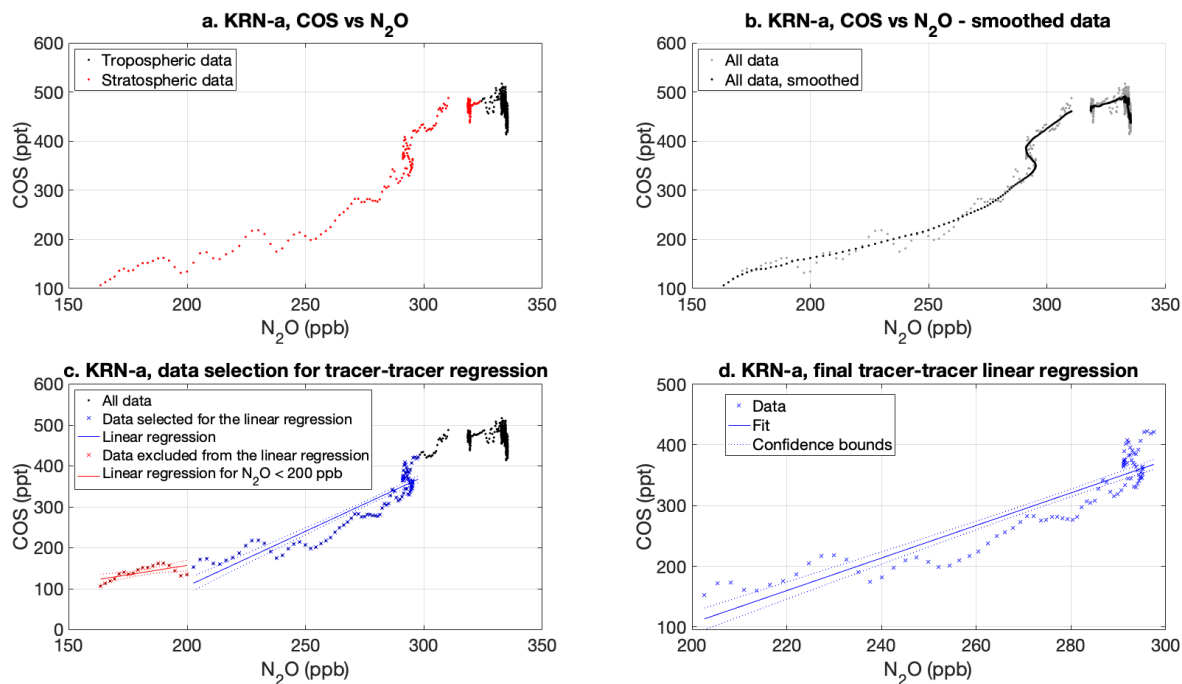


Figure S4: Data selection steps for the linear regression between COS and N_2O , KRN-a AirCore data.

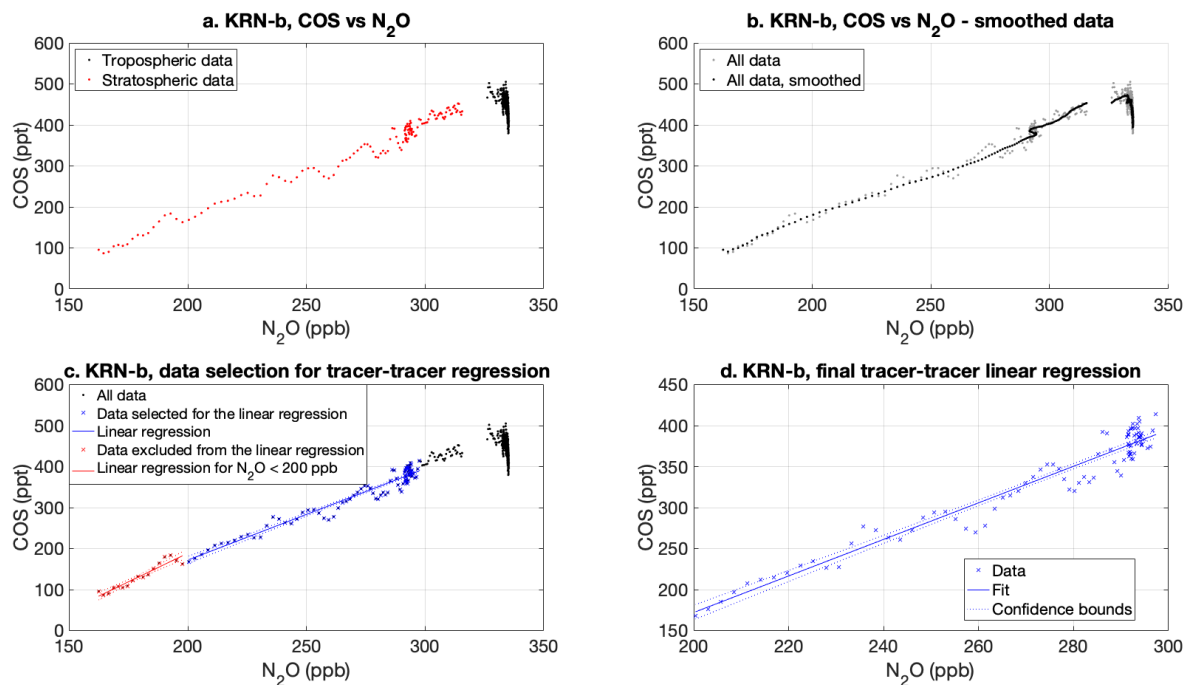


Figure S5: Data selection steps for the linear regression between COS and N_2O , KRN-b AirCore data.

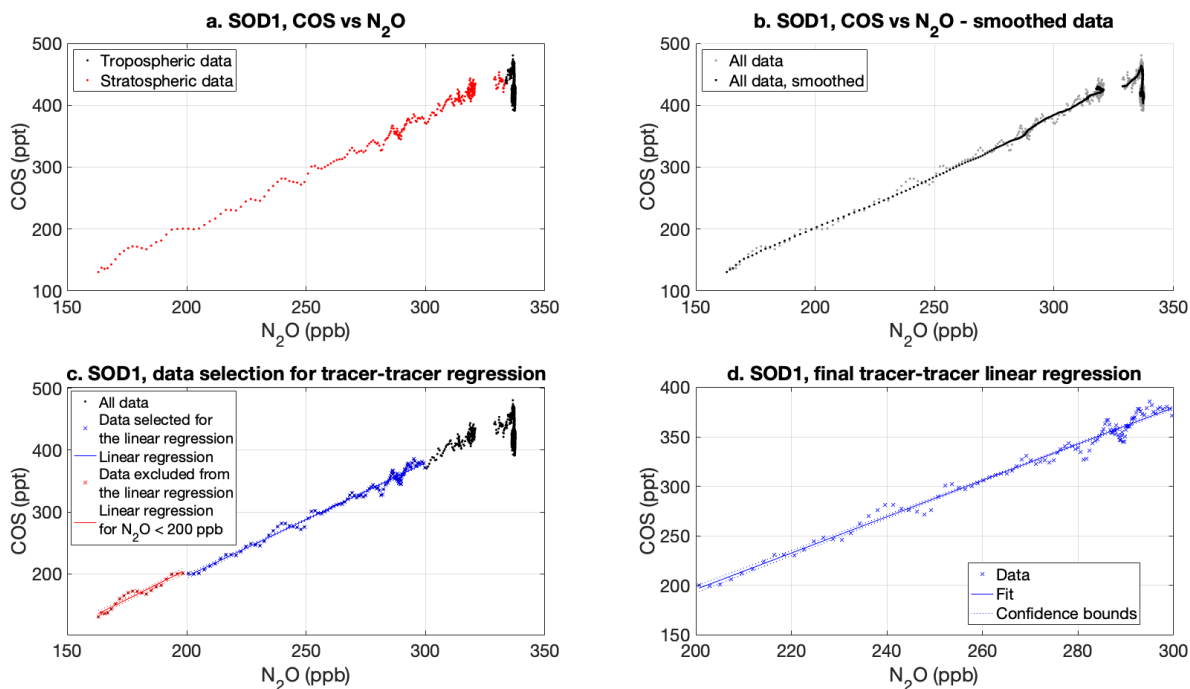


Figure S6: Data selection steps for the linear regression between COS and N_2O , SOD1 AirCore data.

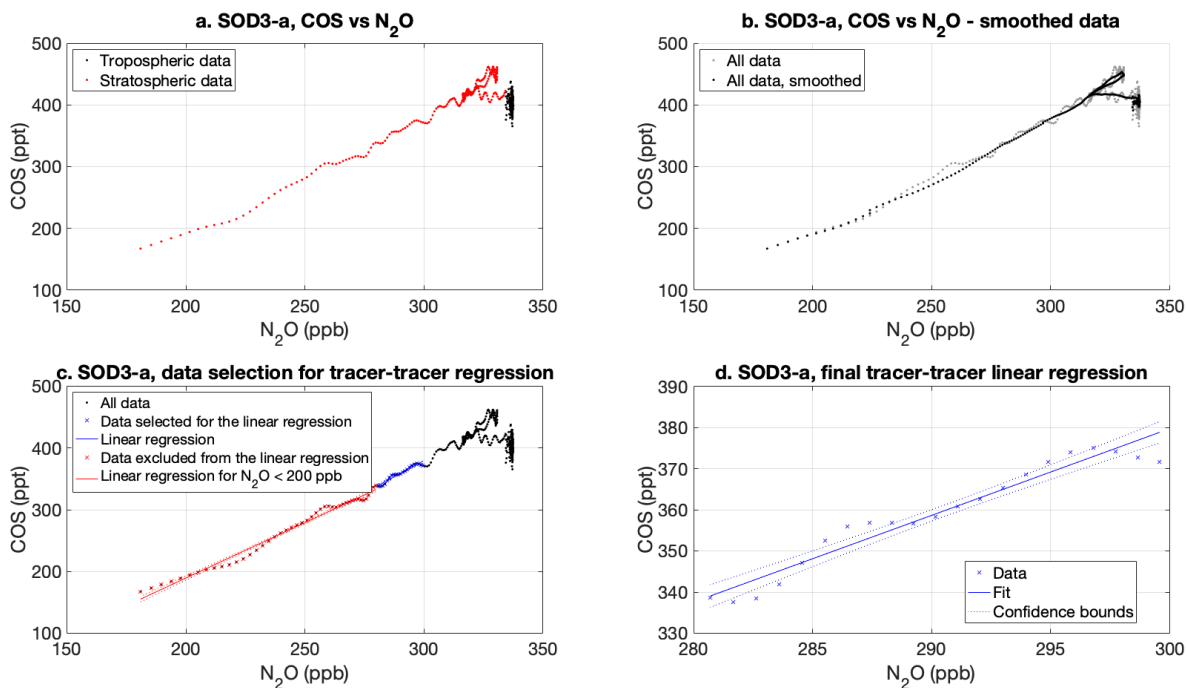
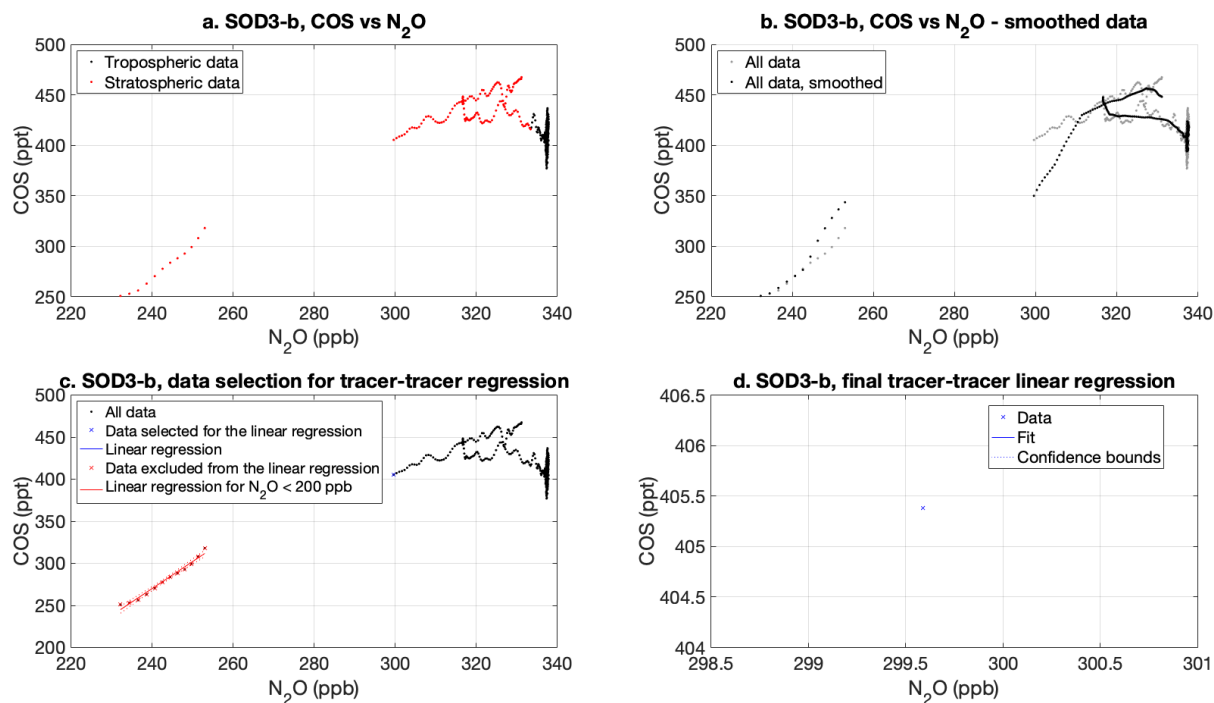


Figure S7: Data selection steps for the linear regression between COS and N_2O , SOD3-a AirCore data.



25 **Figure S8:** Data selection steps for the linear regression between COS and N_2O , SOD3-b AirCore data.

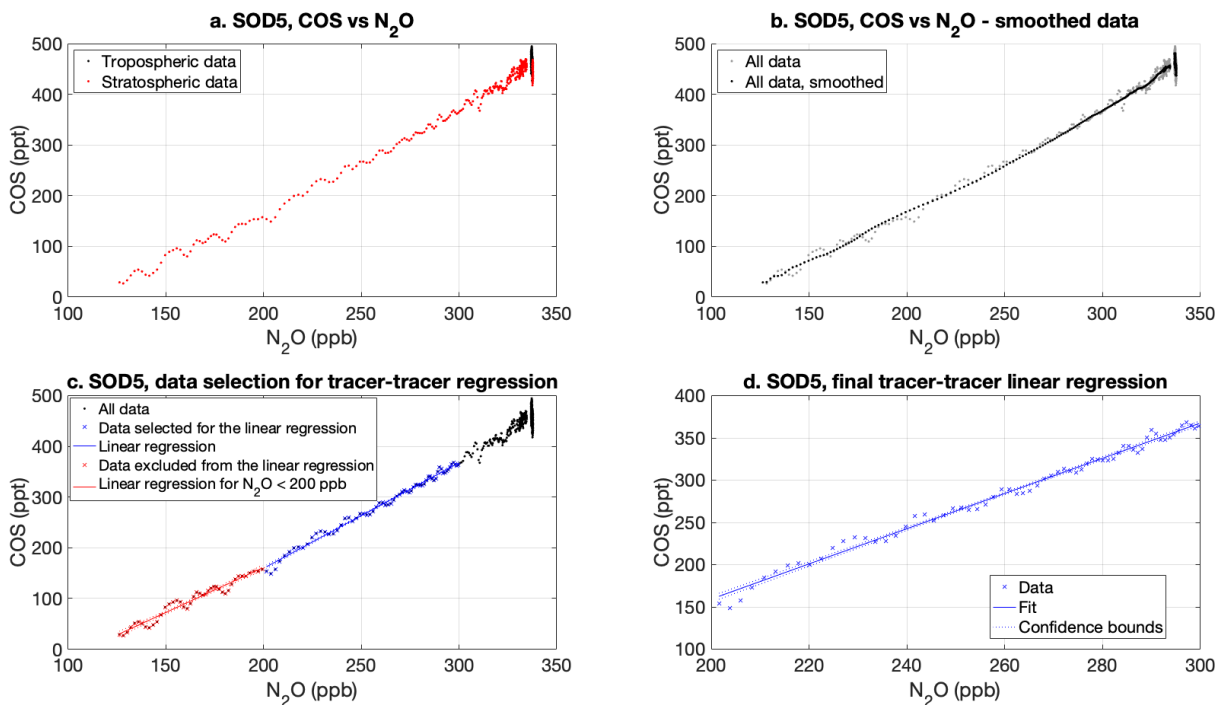


Figure S9: Data selection steps for the linear regression between COS and N_2O , SOD5 AirCore data.

S3 Data selection process for calculating the slope from CH₄-N₂O correlation

30 In this section, the data selection process for the CH₄-N₂O slope estimation will be presented for all flights except TRN2, which is reported in the main text.

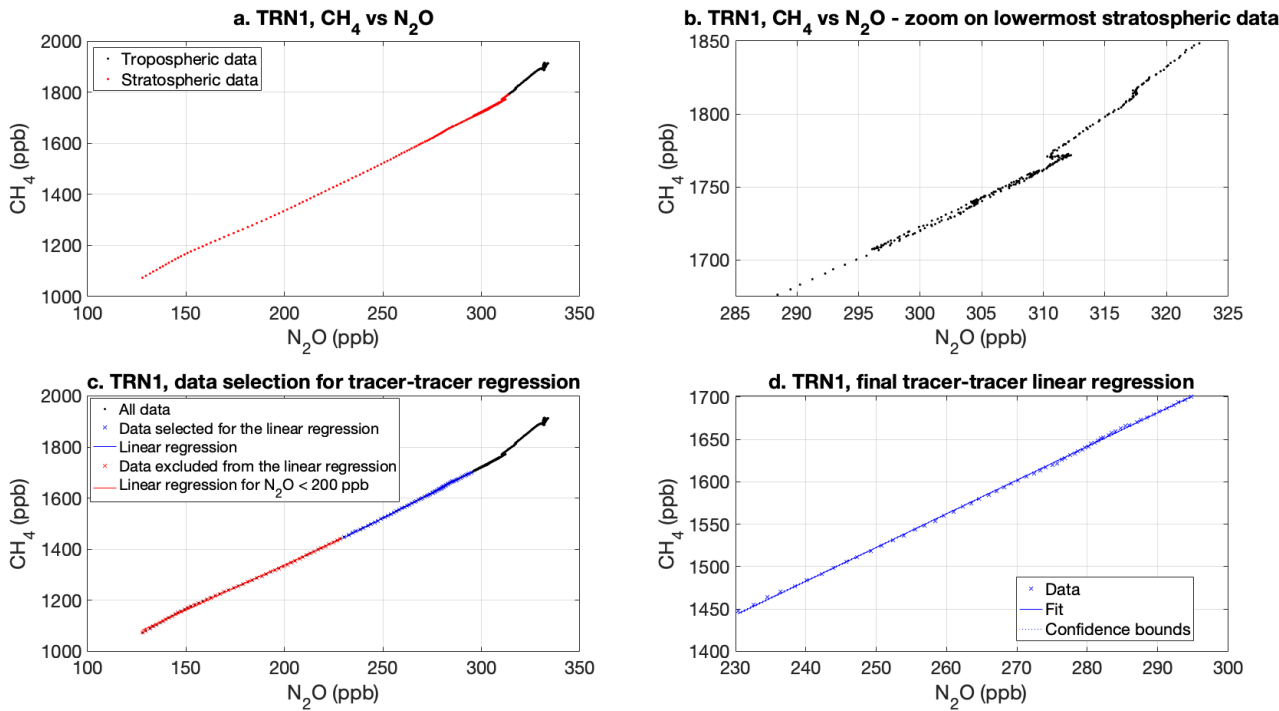
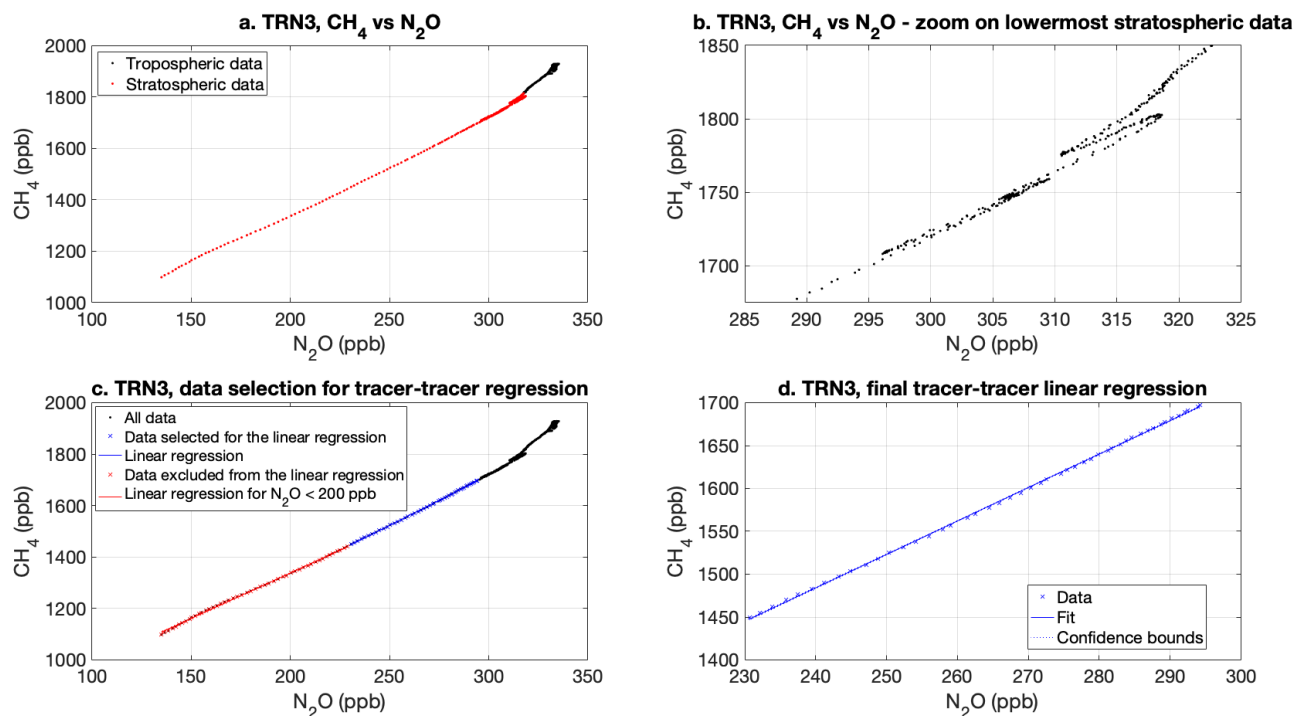


Figure S10: Data selection steps for the linear regression between CH₄ and N₂O, TRN1 AirCore data.



35 **Figure S11:** Data selection steps for the linear regression between CH_4 and N_2O , TRN3 AirCore data.

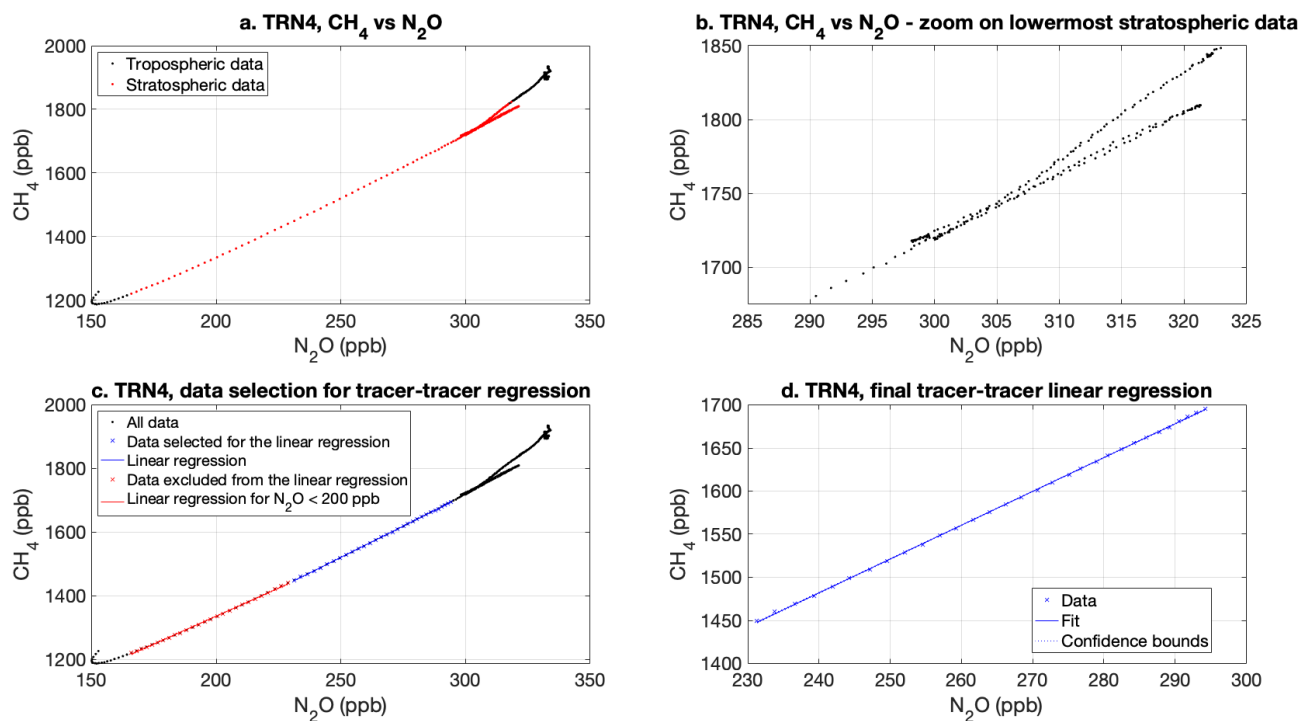


Figure S12: Data selection steps for the linear regression between CH_4 and N_2O , TRN1 AirCore data.

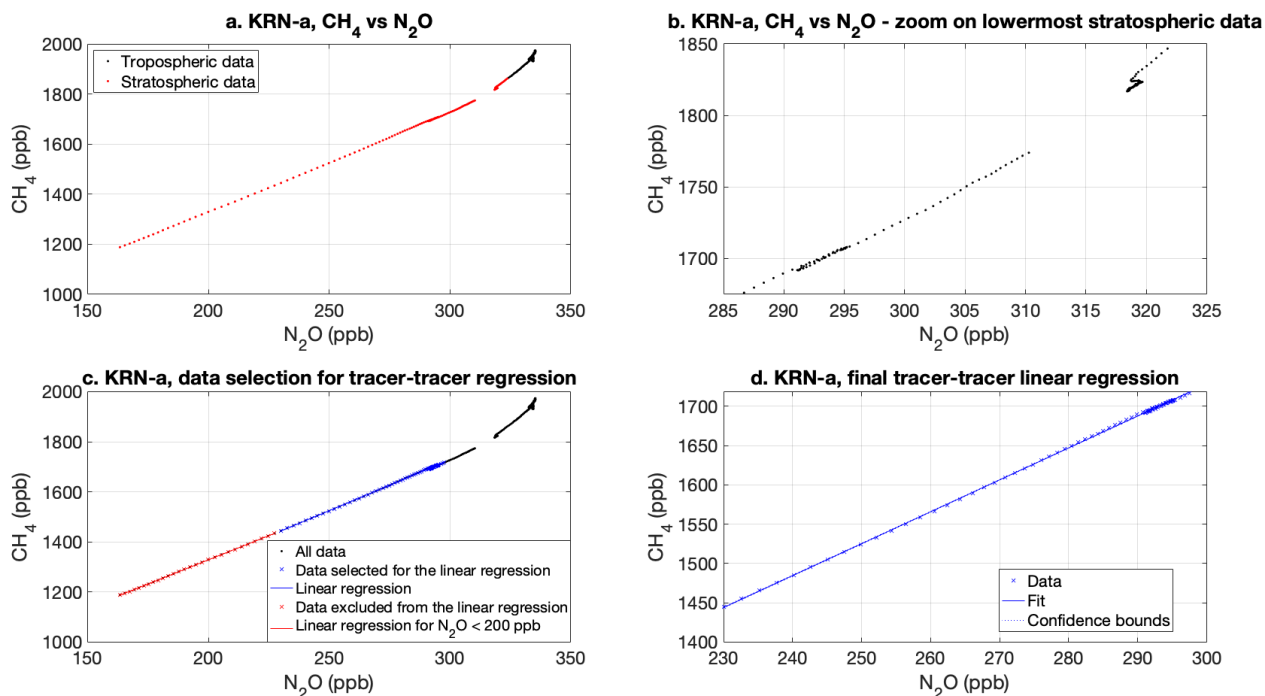


Figure S13: Data selection steps for the linear regression between CH_4 and N_2O , KRN-a AirCore data.

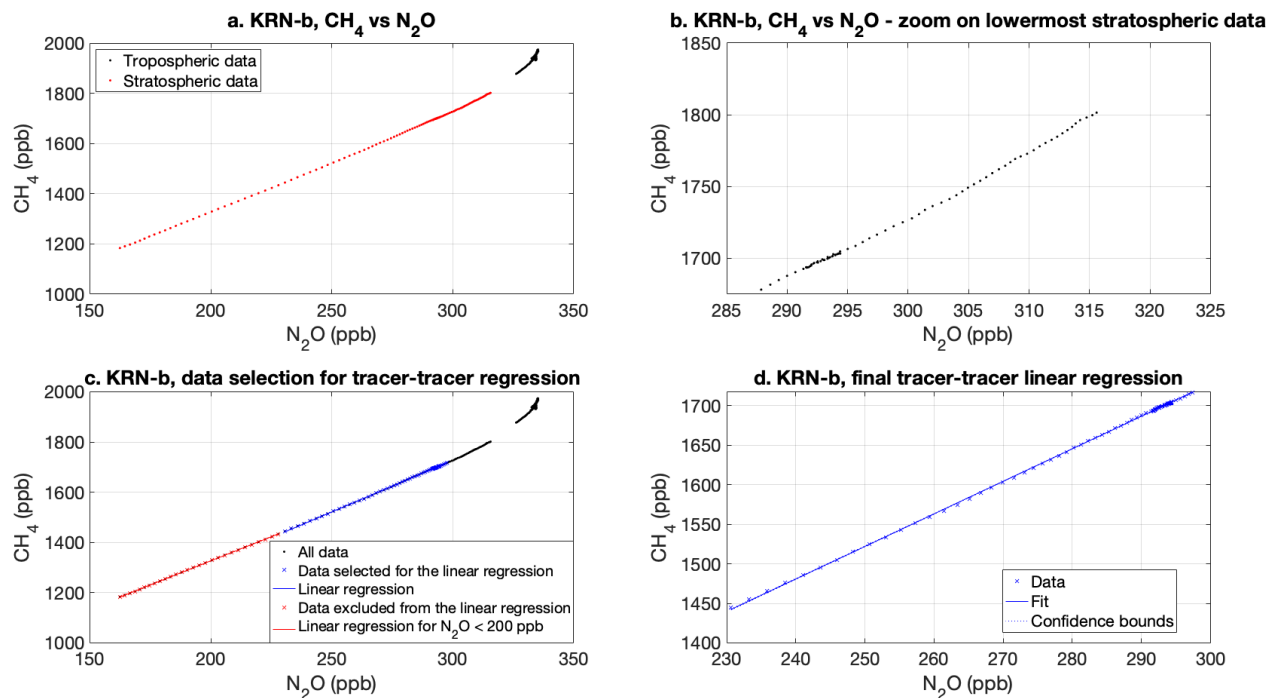


Figure S14: Data selection steps for the linear regression between CH_4 and N_2O , KRN-b AirCore data.

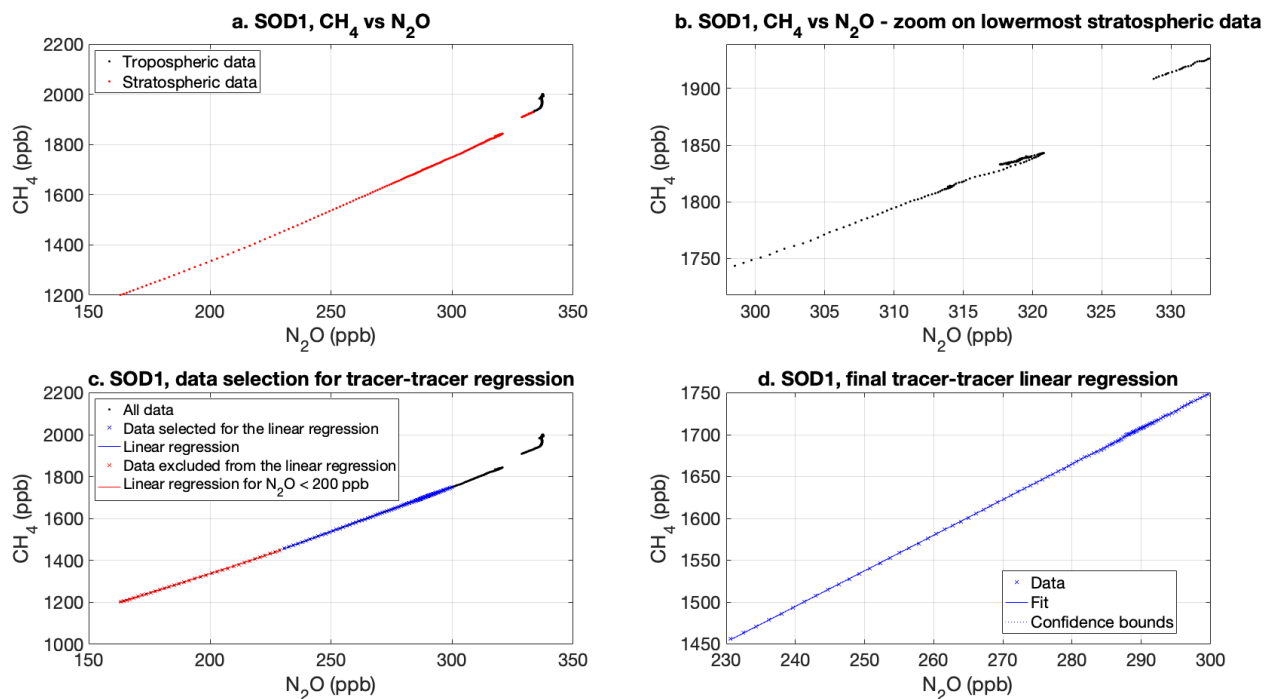


Figure S15: Data selection steps for the linear regression between CH_4 and N_2O , SOD1 AirCore data.

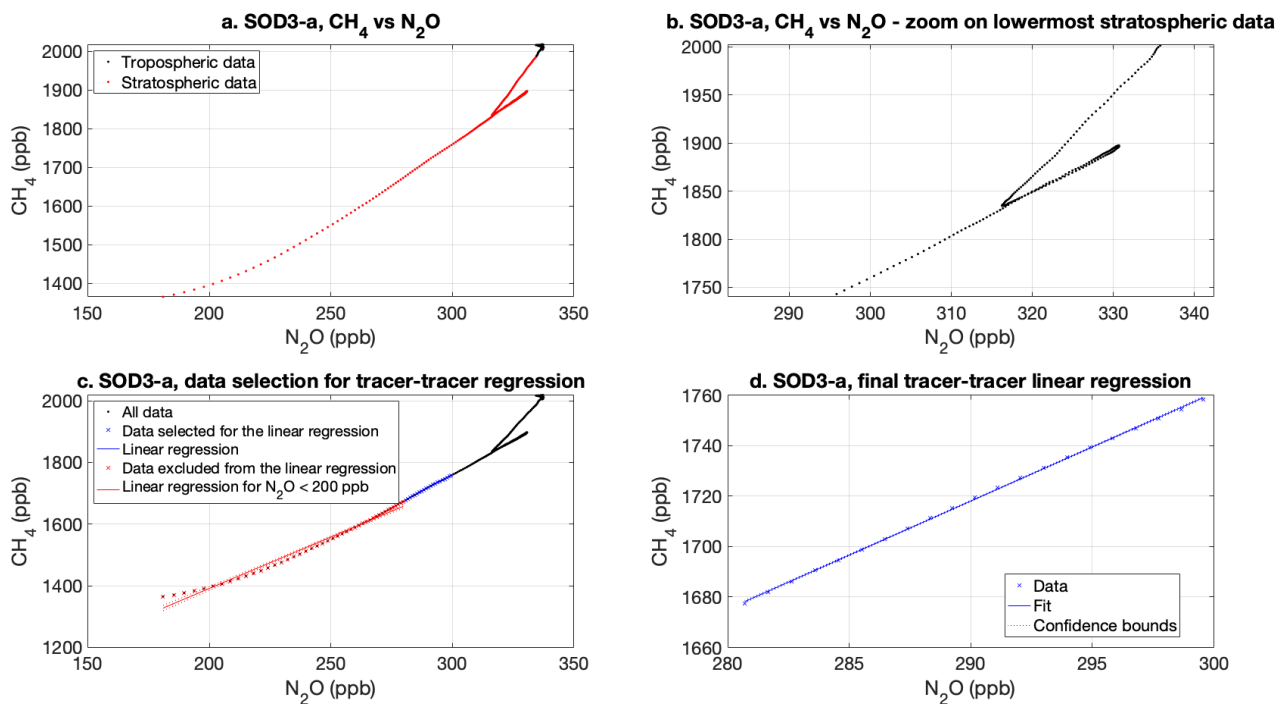


Figure S16: Data selection steps for the linear regression between CH_4 and N_2O , SOD3-a AirCore data.

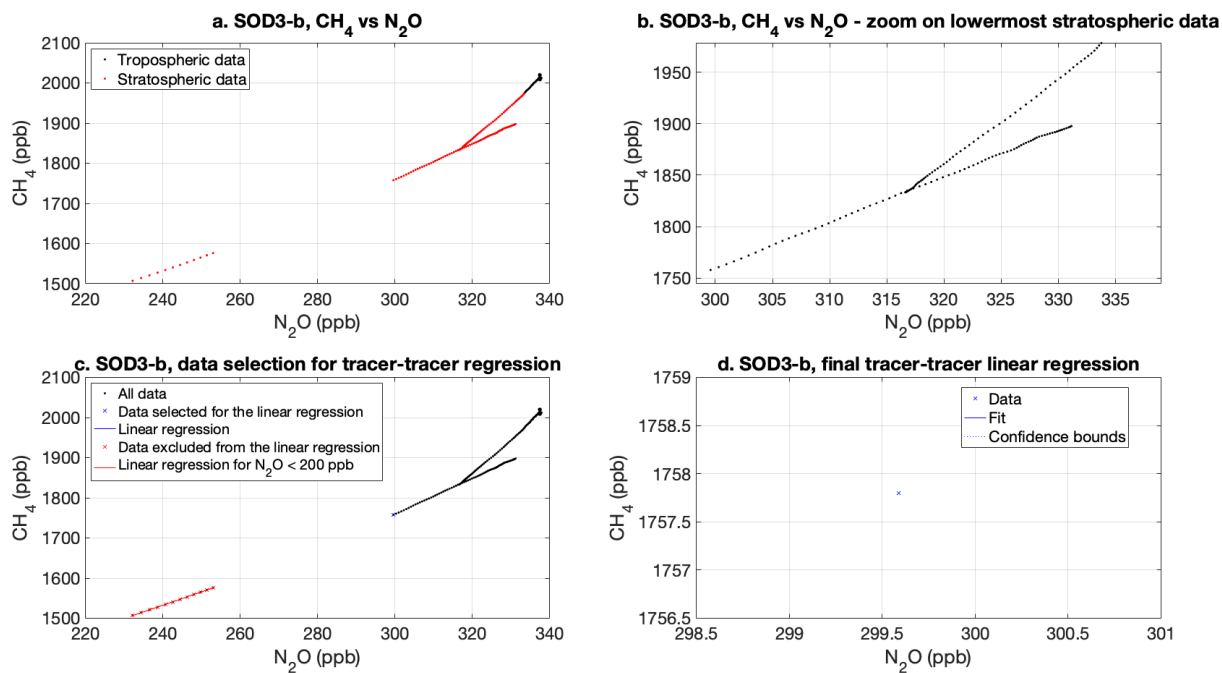


Figure S17: Data selection steps for the linear regression between CH_4 and N_2O , SOD3-b AirCore data.

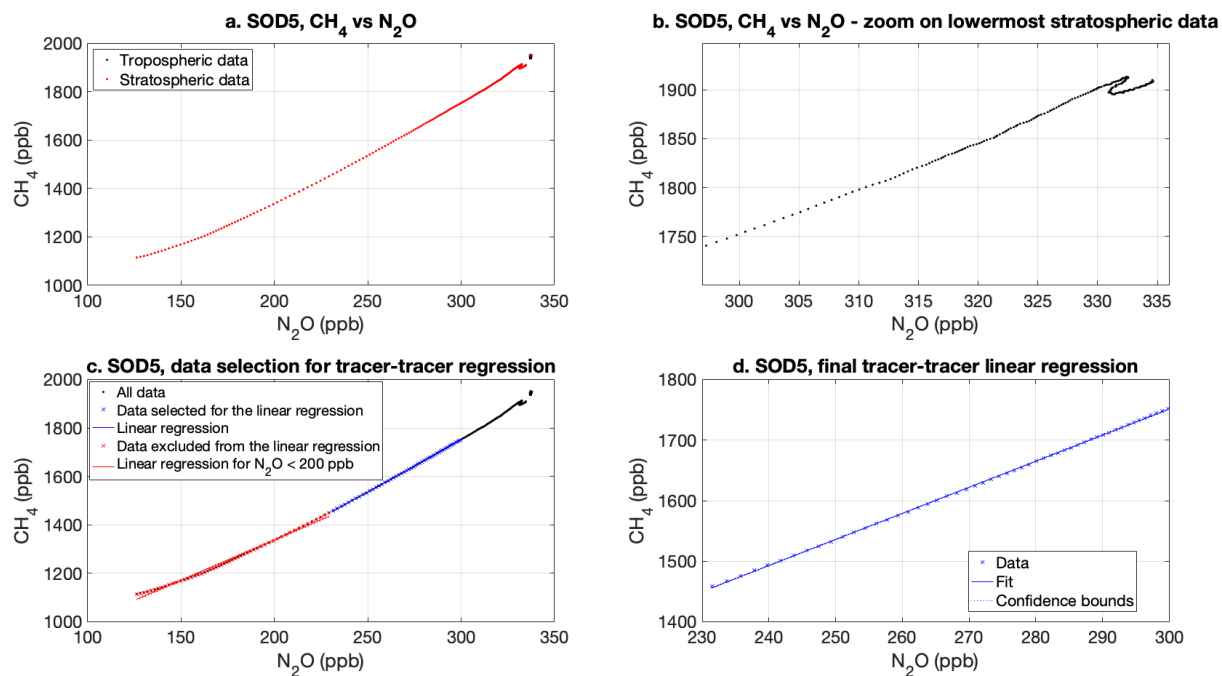


Figure S18: Data selection steps for the linear regression between CH_4 and N_2O , SOD5 AirCore data.

S3 Burden estimation example

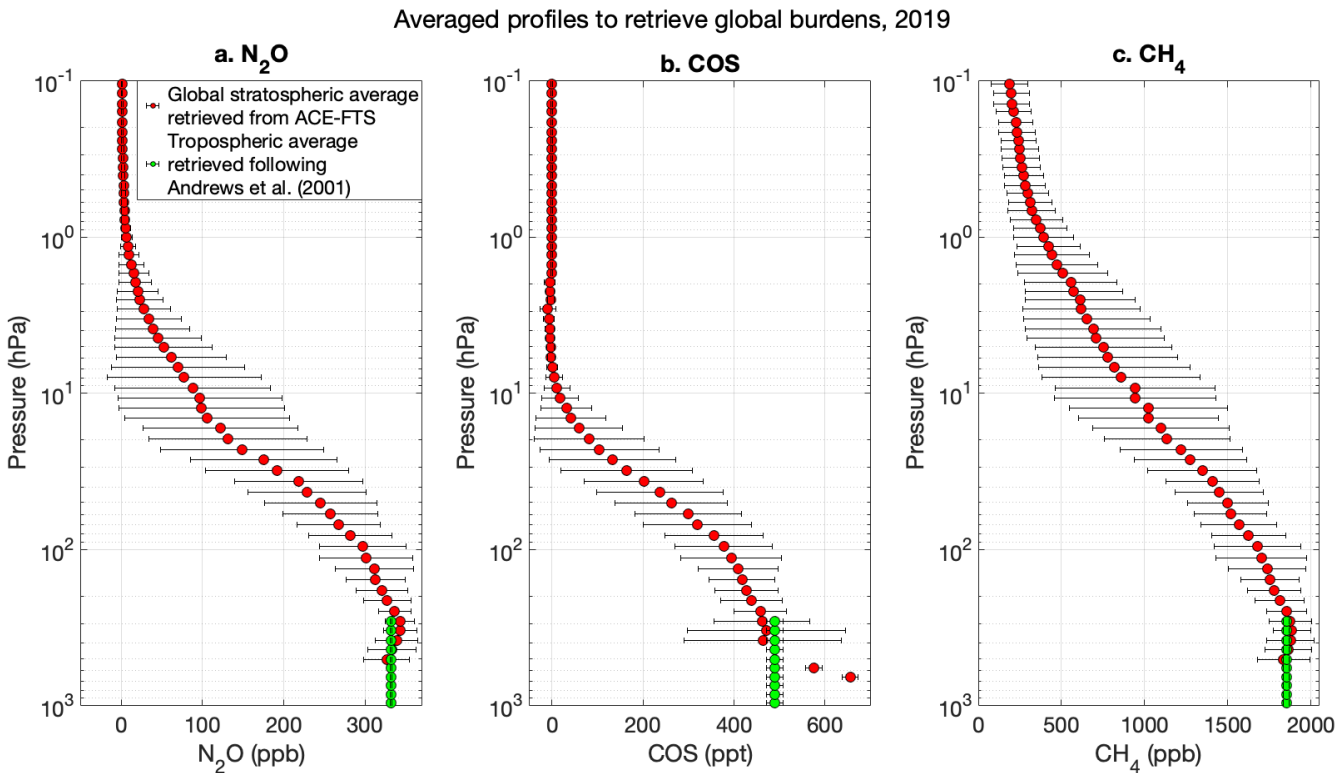


Figure S19: The average profiles used to calculate the global average burdens for 2019. The red dots represent globally averaged ACE-FTS data, while the green dots are the tropospheric averages retrieved following Andrews et al. (2001). It should be noted that ACE-FTS observations are biased towards mid and high latitudes. Where both data are present, only the averages retrieved with Andrews et al. (2001) method were considered.

S4 COS stratospheric lifetime and sink estimates

Table S3: Results of tropospheric burdens and stratospheric linear regression between COS and N₂O (Plumb and Ko, 1992) for all AirCore samples and ACE-FTS averages. The results marked by * shall be considered outliers.

Date/period	Data origin	COS tropospheric burden (ppt)	N ₂ O tropospheric burden (ppb)	N ₂ O COS ⁻¹ stratospheric slope (ppb ppt ⁻¹)	N ₂ O COS ⁻¹ R-squared
17/06/2019	TRN1	490 ± 18	332.0 ± 0.7	0.409 ± 0.014	0.924
18/06/2019	TRN2			0.463 ± 0.017	0.945
18/06/2019	TRN3			0.405 ± 0.012	0.949
20/06/2019	TRN4			0.448 ± 0.018	0.938

13/08/2021	KRN-a	479 ± 21	334.6 ± 1.1	0.316 ± 0.013 *	0.847
13/08/2021	KRN-b			0.423 ± 0.011	0.943
02/08/2023	SOD1	487 ± 18	336.9 ± 2.0	0.535 ± 0.007	0.982
05/08/2023	SOD3-a			0.448 ± 0.024	0.947
05/08/2023	SOD3-b			- *	-
08/08/2023	SOD5			0.475 ± 0.006	0.989
June to September, 2012-present	Polar (65°-69° N) average, ACE-FTS	487 ± 18	336.9 ± 2.0	0.456 ± 0.004	0.999
June to September, 2012-present	Mid-latitude (45°-49° N) average, ACE-FTS	490 ± 18	332.0 ± 0.7	0.436 ± 0.003	0.999

60

Table S4: Average global atmospheric burdens of COS and N₂O and COS stratospheric lifetime and sink estimates, retrieved following the Volk et al. (1997) method. The missing results are related to slopes at the tropopause that were marked as outliers (signalled by *, see Sect. 3.1.3).

Date	Data origin	N ₂ O COS ⁻¹ slope at the tropopause	COS global atmospheric average	N ₂ O global atmospheric average	COS lifetime (years)	COS sink (GgS yr ⁻¹)
17/06/2019	TRN1	0.419 ± 0.011	468 ± 9 ppt	322.3 ± 2.5 ppb	71 ± 9	38 ± 6
18/06/2019	TRN2	0.490 ± 0.025			83 ± 13	32 ± 6
18/06/2019	TRN3	0.403 ± 0.011			68 ± 9	39 ± 6
20/06/2019	TRN4	0.728 ± 0.047 *			-	-
13/08/2021	KRN-a	0.198 ± 0.011 *	458 ± 15 ppt	324.2 ± 2.4 ppb	-	-
13/08/2021	KRN-b	0.346 ± 0.016 *			-	-
02/08/2023	SOD1	0.498 ± 0.011	457 ± 9 ppt	324.0 ± 3.6 ppb	81 ± 11	32 ± 5
05/08/2023	SOD3-a	0.502 ± 0.024			82 ± 13	32 ± 6
05/08/2023	SOD3-b	0.276 ± 0.056 *			-	-
08/08/2023	SOD5	0.446 ± 0.010			73 ± 9	36 ± 5
June to September, 2012-2023	Polar (65°-69° N) average, ACE-FTS	0.483 ± 0.007	457 ± 9 ppt	324.0 ± 3.6 ppb	79 ± 10	33 ± 5
June to September, 2012-2023	Mid-latitude (45°-49° N) average, ACE-FTS	0.429 ± 0.008	468 ± 9 ppt	322.3 ± 2.5 ppb	72 ± 9	37 ± 5

65 **Table S5:** Calculated COS lifetime and sink for all AirCore campaigns and ACE-FTS observations following the method described by Plumb and Ko (1992). The missing results are the ones related to outliers (see Sect. 3.1.3).

Date	Data origin	COS tropospheric burden (TgCOS)	COS lifetime (years)	COS stratospheric sink (GgS yr ⁻¹)
17/06/2019	TRN1	5.29 ± 0.10	70 ± 11	40 ± 8
18/06/2019	TRN2	5.29 ± 0.10	79 ± 12	35 ± 7
18/06/2019	TRN3	5.29 ± 0.10	69 ± 10	41 ± 7
20/06/2019	TRN4	5.29 ± 0.10	77 ± 12	36 ± 7
13/08/2021	KRN-a	5.15 ± 0.12	-	-
13/08/2021	KRN-b	5.15 ± 0.12	70 ± 10	39 ± 7
02/08/2023	SOD1	5.20 ± 0.10	90 ± 12	31 ± 5
05/08/2023	SOD3-a	5.20 ± 0.10	75 ± 13	37 ± 8
05/08/2023	SOD3-b	5.20 ± 0.10	-	-
08/08/2023	SOD5	5.20 ± 0.10	80 ± 11	35 ± 6
June to September, 2012-2023	Polar (65°-69° N) average, ACE-FTS	5.20 ± 0.10	76 ± 10	36 ± 6
June to September, 2012-2023	Mid-latitude (45°-49° N) average, ACE-FTS	5.29 ± 0.10	75 ± 9	37 ± 6

70 **Table S6:** Tropospheric burdens (Sect 3.1.1) and stratospheric linear regression results (Sect. 3.1.2) between CH₄ and N₂O for all AirCore samples and ACE-FTS averages.

Date	Data origin	CH ₄ tropospheric burden (ppb)	N ₂ O tropospheric burden (ppb)	N ₂ O CH ₄ ⁻¹ stratospheric slope (ppb ppb ⁻¹)	N ₂ O CH ₄ ⁻¹ R-squared
17/06/2019	TRN1	1844.6 ± 9.9	332.0 ± 0.4	0.2516 ± 0.0006	0.999
18/06/2019	TRN2	1844.6 ± 9.9	332.0 ± 0.4	0.2523 ± 0.0005	0.999
18/06/2019	TRN3	1844.6 ± 9.9	332.0 ± 0.4	0.2562 ± 0.0007	0.999
20/06/2019	TRN4	1844.6 ± 9.9	332.0 ± 0.4	0.2549 ± 0.0006	0.999
13/08/2021	KRN-a	1864.3 ± 10.2	333.3 ± 0.4	0.2463 ± 0.0004	0.999
13/08/2021	KRN-b	1864.3 ± 10.2	333.3 ± 0.4	0.2432 ± 0.0004	0.999
02/08/2023	SOD1	1899.2 ± 9.9	335.9 ± 0.3	0.2352 ± 0.0002	0.999
05/08/2023	SOD3-a	1899.2 ± 9.9	335.9 ± 0.3	0.2334 ± 0.0010	0.999

05/08/2023	SOD3-b	1899.2 ± 9.9	335.9 ± 0.3	-	-
08/08/2023	SOD5	1899.2 ± 9.9	335.9 ± 0.3	0.2322 ± 0.0004	0.999
June to September, 2012-2023	Polar (65°-69° N) average, ACE-FTS	1899.2 ± 9.9	335.9 ± 0.3	0.2217 ± 0.0053	0.999
June to September, 2012-2023	Mid-latitude (45°- 49° N) average, ACE-FTS	1844.6 ± 9.9	332.0 ± 0.4	0.2647 ± 0.0118	0.996

Table S7: Calculated CH₄ lifetime and sink for all AirCore campaigns and ACE-FTS observations following Plumb and Ko (1992).

Date	Data origin	CH ₄ tropospheric burden (PgCH ₄)	CH ₄ lifetime (years)	CH ₄ stratospheric sink (TgC yr ⁻¹)
17/06/2019	TRN1	5.25 ± 0.03	163 ± 15	24 ± 2
18/06/2019	TRN2	5.25 ± 0.03	163 ± 15	24 ± 2
18/06/2019	TRN3	5.25 ± 0.03	166 ± 15	24 ± 2
20/06/2019	TRN4	5.25 ± 0.03	165 ± 15	24 ± 2
13/08/2021	KRN-a	5.30 ± 0.03	161 ± 15	25 ± 3
13/08/2021	KRN-b	5.30 ± 0.03	159 ± 15	25 ± 3
02/08/2023	SOD1	5.40 ± 0.03	155 ± 15	26 ± 3
05/08/2023	SOD3-a	5.40 ± 0.03	153 ± 15	27 ± 3
05/08/2023	SOD3-b	5.40 ± 0.03	-	-
08/08/2023	SOD5	5.40 ± 0.03	153 ± 15	27 ± 3
June to September, 2012-2023	Polar (65°-69° N) average, ACE-FTS	5.40 ± 0.03	146 ± 17	28 ± 4
June to September, 2012-2023	Mid-latitude (45°-49° N) average, ACE-FTS	5.25 ± 0.03	171 ± 23	23 ± 3

Table S8: Slopes at the tropopause, average global atmospheric burdens of CH₄ and N₂O and CH₄ stratospheric lifetime and sink estimates, retrieved following the Volk et al. (1997) method.

Date	Data origin	N ₂ O CH ₄ ⁻¹ slope at the tropopause	CH ₄ global atmospheric average	N ₂ O global atmospheric average	CH ₄ lifetime (years)	CH ₄ sink (TgC yr ⁻¹)
17/06/2019	TRN1	0.2473 ± 0.0005	1806.6 ± 14.1 ppb	322.3 ± 2.5 ppb	161 ± 15	24 ± 2
18/06/2019	TRN2	0.2394 ± 0.012			156 ± 15	25 ± 3
18/06/2019	TRN3	0.2425 ± 0.0014			158 ± 16	24 ± 3

20/06/2019	TRN4	0.2374 ± 0.0018			154 ± 16	25 ± 3
13/08/2021	KRN-a	0.2481 ± 0.0008	1825.5 ± 15.3 ppb	324.2 ± 2.4 ppb	162 ± 16	24 ± 3
13/08/2021	KRN-b	0.2401 ± 0.0008			157 ± 15	25 ± 3
02/08/2023	SOD1	0.2361 ± 0.0002	1845.9 ± 20.5 ppb	324.0 ± 3.6 ppb	156 ± 16	25 ± 3
05/08/2023	SOD3-a	-			-	-
05/08/2023	SOD3-b	-			-	-
08/08/2023	SOD5	0.2260 ± 0.0003			149 ± 15	26 ± 3
June to September, 2012-2023	Polar (65° - 69° N) average, ACE-FTS	0.1948 ± 0.0098	1825.5 ± 15.3 ppb	324.0 ± 3.6 ppb	127 ± 18	31 ± 5
June to September, 2012-2023	Mid-latitude (45° - 49° N) average, ACE-FTS	0.2343 ± 0.0169	1806.6 ± 14.1 ppb	322.3 ± 2.5 ppb	152 ± 25	25 ± 4

Table S9: Effects of different estimations of tropospheric burdens on the final lifetime and sink estimations.

Source	Parameters used in the source study	COS lifetime in the source study (years)	COS sink in the source study (GgS yr ⁻¹)	Parameters following the methods of this study	COS lifetime according to the new parameters (years)	COS sink according to the new parameters (GgS yr ⁻¹)
SPIRALE, Kryzstofiak et al. (2015)	$\sigma_{COS} = 550 \pm 40$ ppt	70 ± 20 (polar latitudes) 58 ± 14 (tropical latitudes) 64 ± 17 (global)	54 ± 14 (global)	$\sigma_{COS} = 509 \pm 23$ ppt	66 ± 27 (polar latitudes) 54 ± 16 (tropical latitudes) 60 ± 19 (global)	44 ± 20 (polar latitudes) 54 ± 18 (tropical latitudes) 48 ± 22 (global)
	$\sigma_{N_2O} = 328 \pm 8$ ppb			$\sigma_{N_2O} = 321 \pm 1$ ppb		
	$T_{N_2O} = 117 \pm 20$ years			$T_{N_2O} = 116 \pm 9$ years		
Karu et al. (2023)	$\sigma_{COS} = 415 \pm 32$ ppt	39 ± 15 (global, limited to 12 km), potential	55 ± 23 (global)	$\sigma_{COS} = 502 \pm 23$ ppt	46 ± 23 (global, limited to 12 km)	62 ± 34 (global)
	$\sigma_{N_2O} = 324.0 \pm 0.2$ ppb			$\sigma_{N_2O} = 330 \pm 2$ ppb		

	$T_{N_2O} = 116 \pm 9$ years	vorticity tropopause		$T_{N_2O} = 116 \pm 9$ years		
	$\sigma_{COS} = 416 \pm 35$ ppt	55 ± 18 (global, limited to 12 km), thermal tropopause	/	$\sigma_{COS} = 502 \pm 23$ ppt	65 ± 25 (global, limited to 12 km)	44 ± 19 (global)
	$\sigma_{N_2O} = 323.7 \pm 0.2$ ppb			$\sigma_{N_2O} = 330 \pm 2$ ppb		
	$T_{N_2O} = 116 \pm 9$ years			$T_{N_2O} = 116 \pm 9$ years		
Barkley et al. (2008)	$\sigma_{COS} = 500 \pm 20\%$ ppt	64 ± 21 (global)	34–66 (global)	$\sigma_{COS} = 508 \pm 23$ ppt	65 ± 24 (global)	34–66 (global)

80 S5 Equivalent latitude against altitude

The figures in this section show each tracer’s profiles plotted against the calculated equivalent latitudes of every flight.

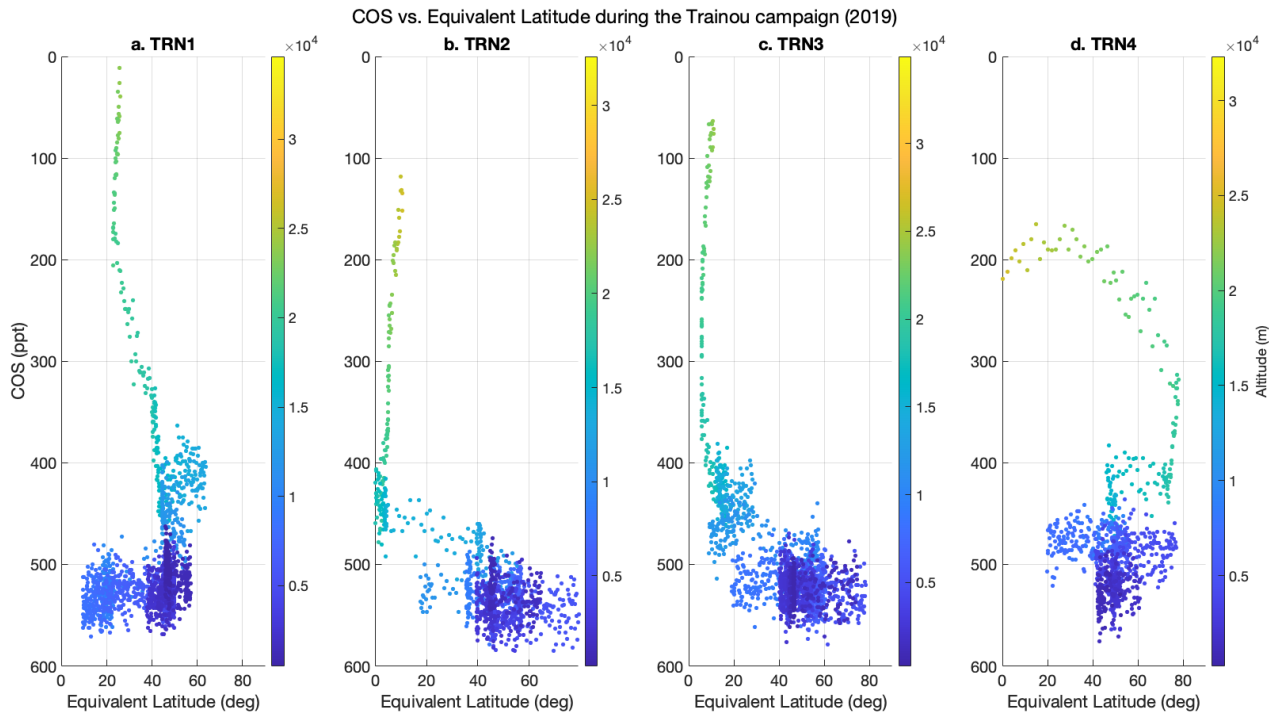


Figure S20: COS AirCore profiles against equivalent latitude in TRN (2019).

N_2O vs. Equivalent Latitude during the Trainou campaign (2019)

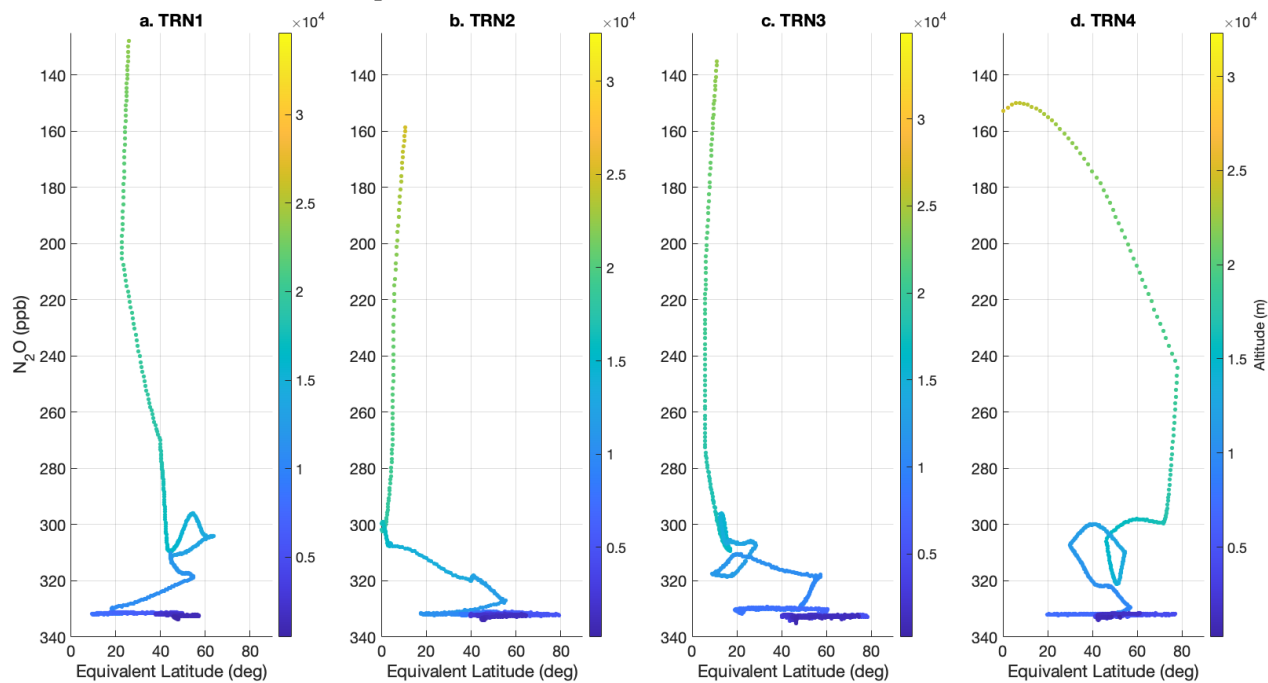


Figure S21: N_2O AirCore profiles against equivalent latitude in TRN (2019).

CH_4 vs. Equivalent Latitude during the Trainou campaign (2019)

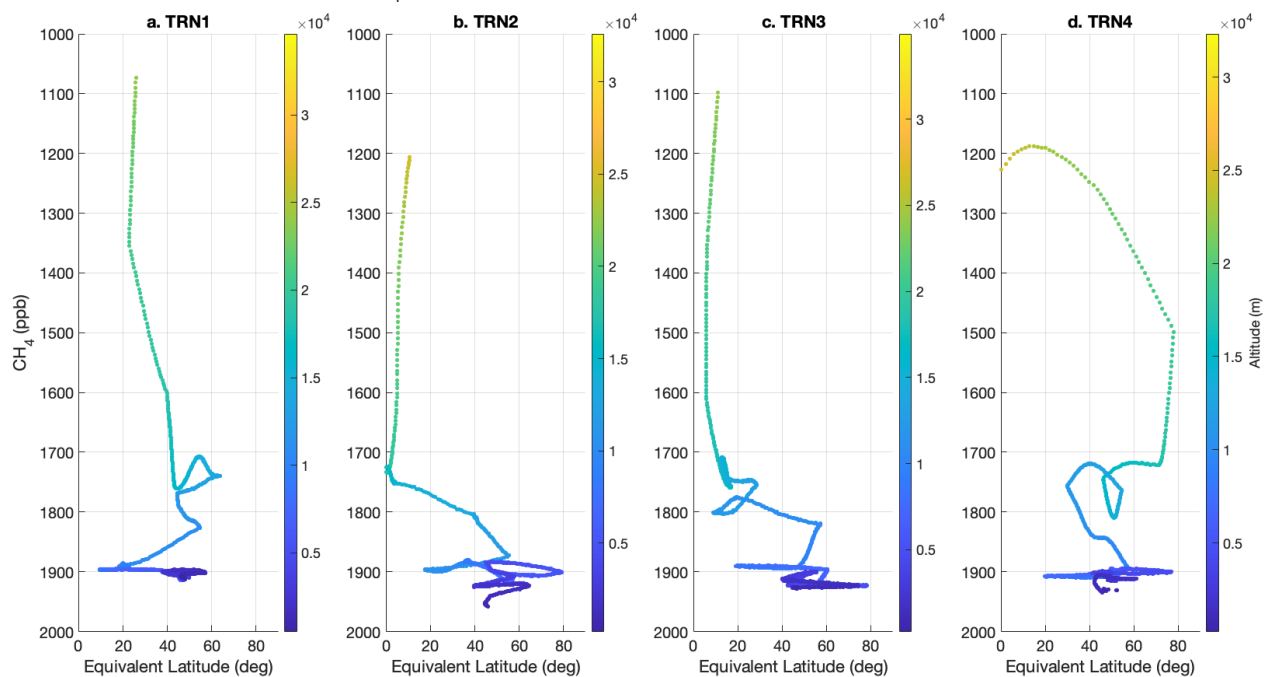
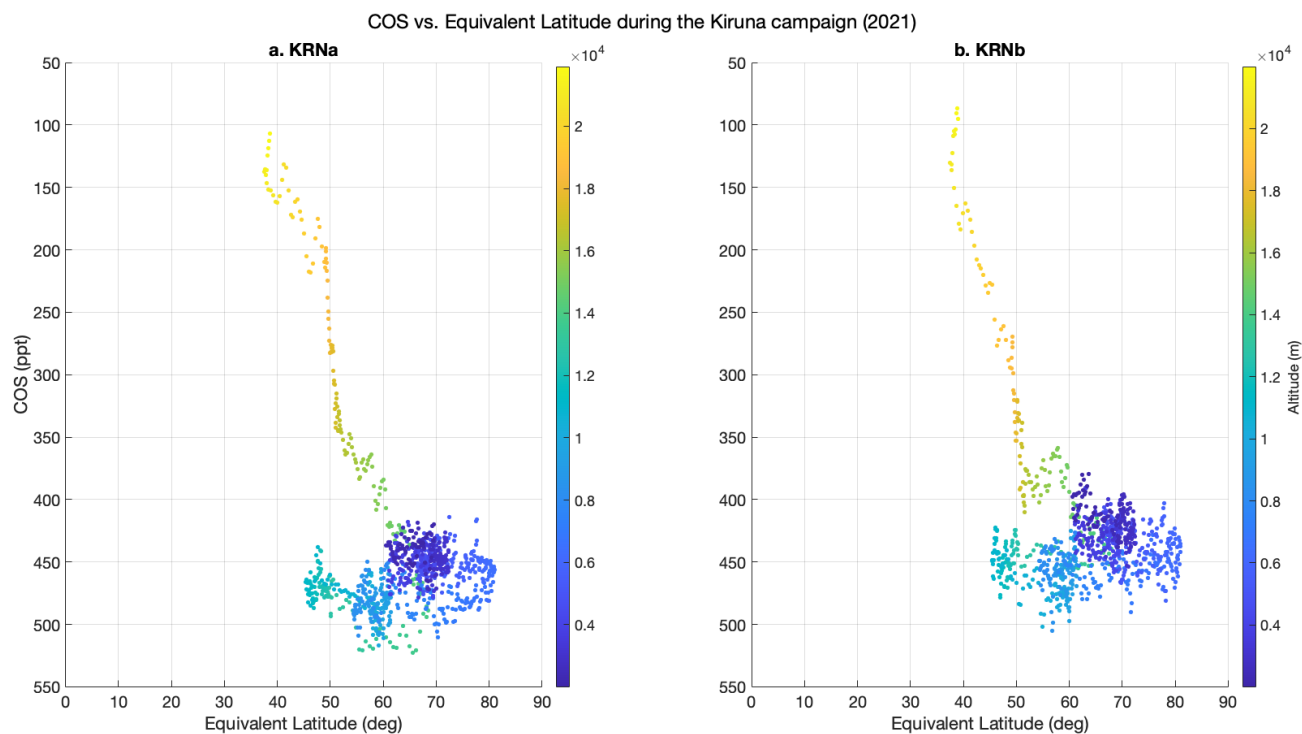


Figure S22: CH_4 AirCore profiles against equivalent latitude in TRN (2019).



90 **Figure S23:** COS AirCore profiles against equivalent latitude in KRN (2021).

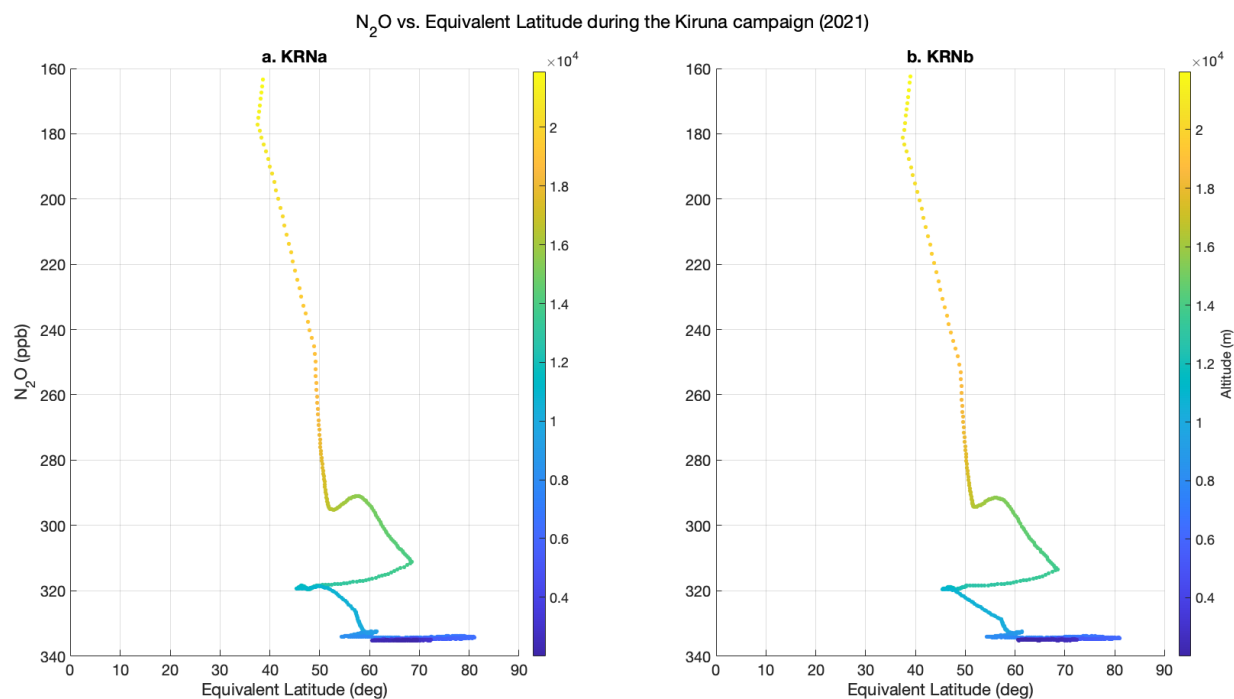


Figure S24: N_2O AirCore profiles against equivalent latitude in KRN (2021).

CH₄ vs. Equivalent Latitude during the Kiruna campaign (2021)

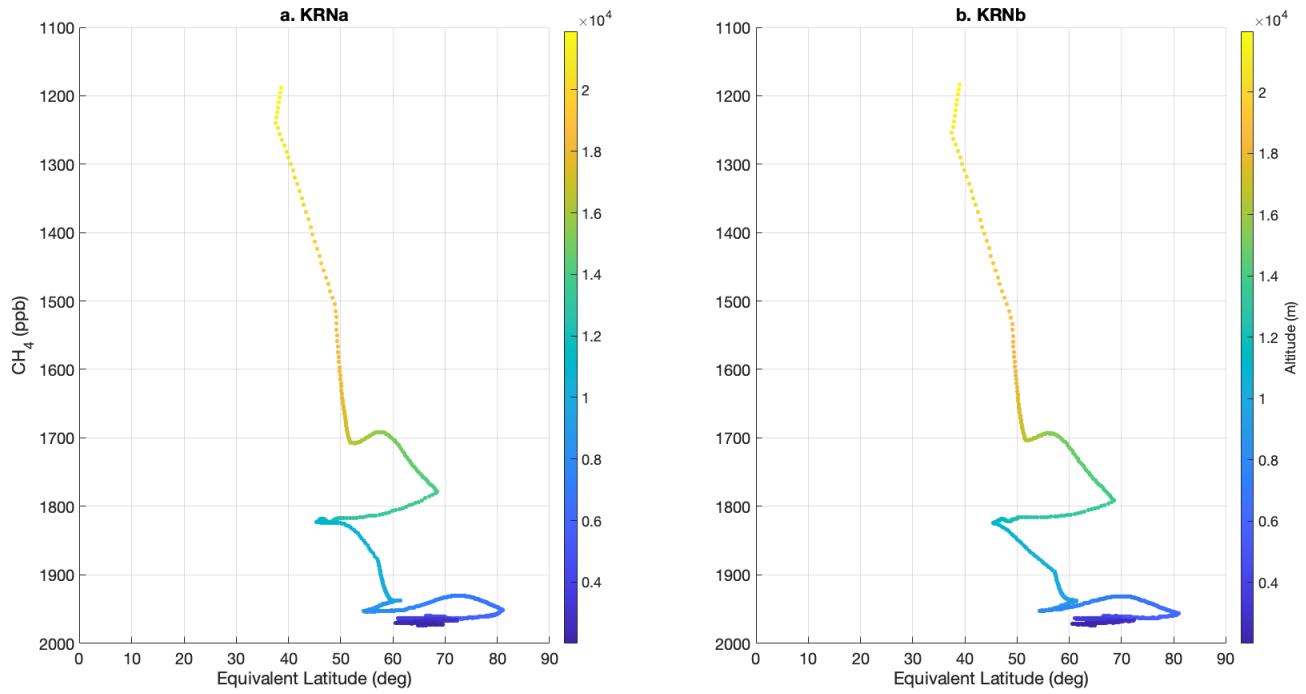


Figure S25: CH₄ AirCore profiles against equivalent latitude in KRN (2021).

COS vs. Equivalent Latitude during the Sodankylä campaign (2023)

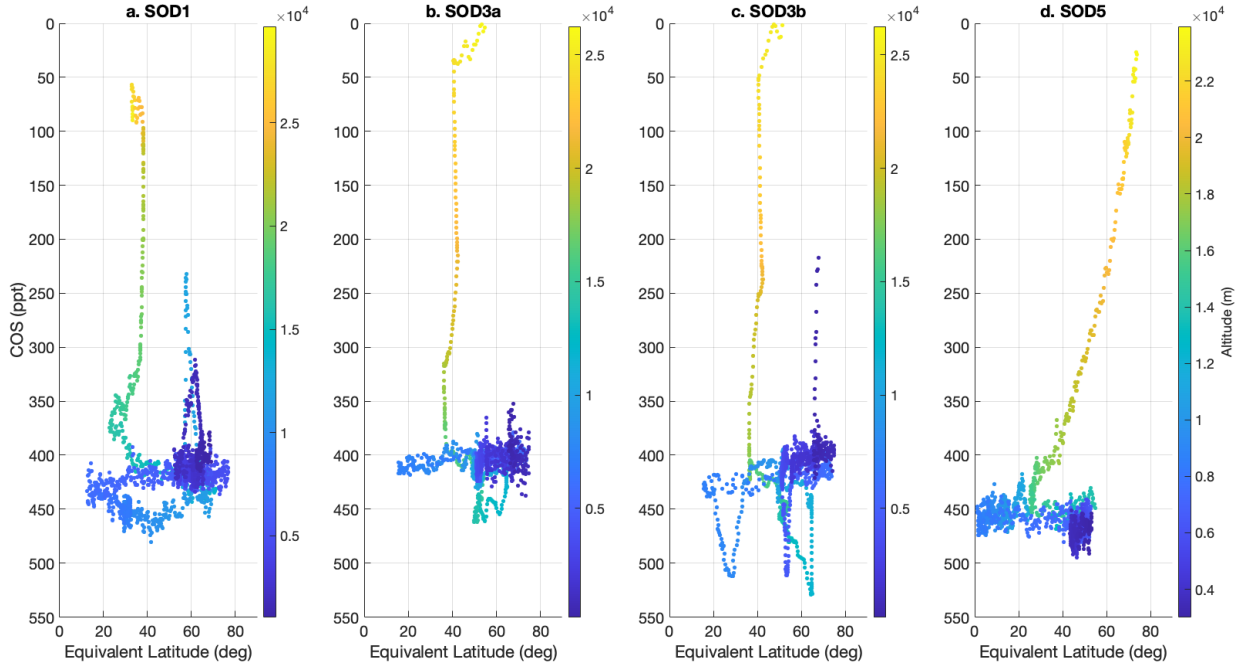


Figure S26: COS AirCore profiles against equivalent latitude in SOD (2023).

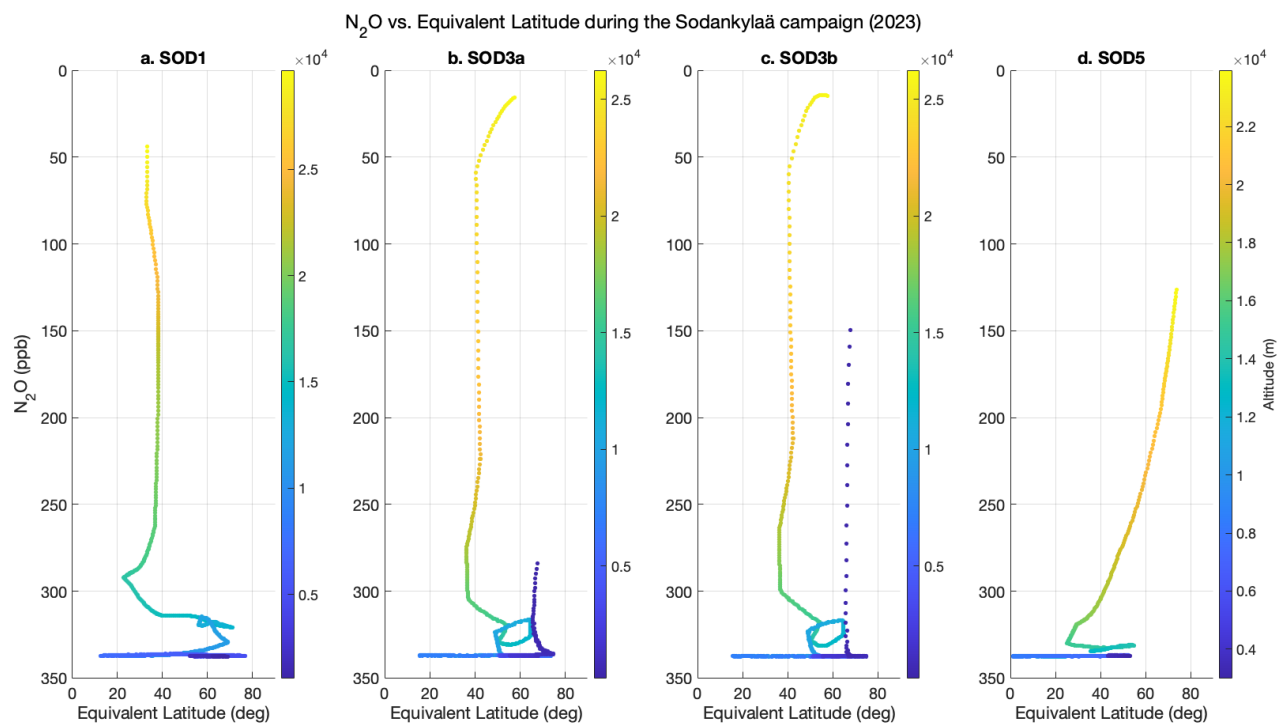


Figure S27: N_2O AirCore profiles against equivalent latitude in SOD (2023).

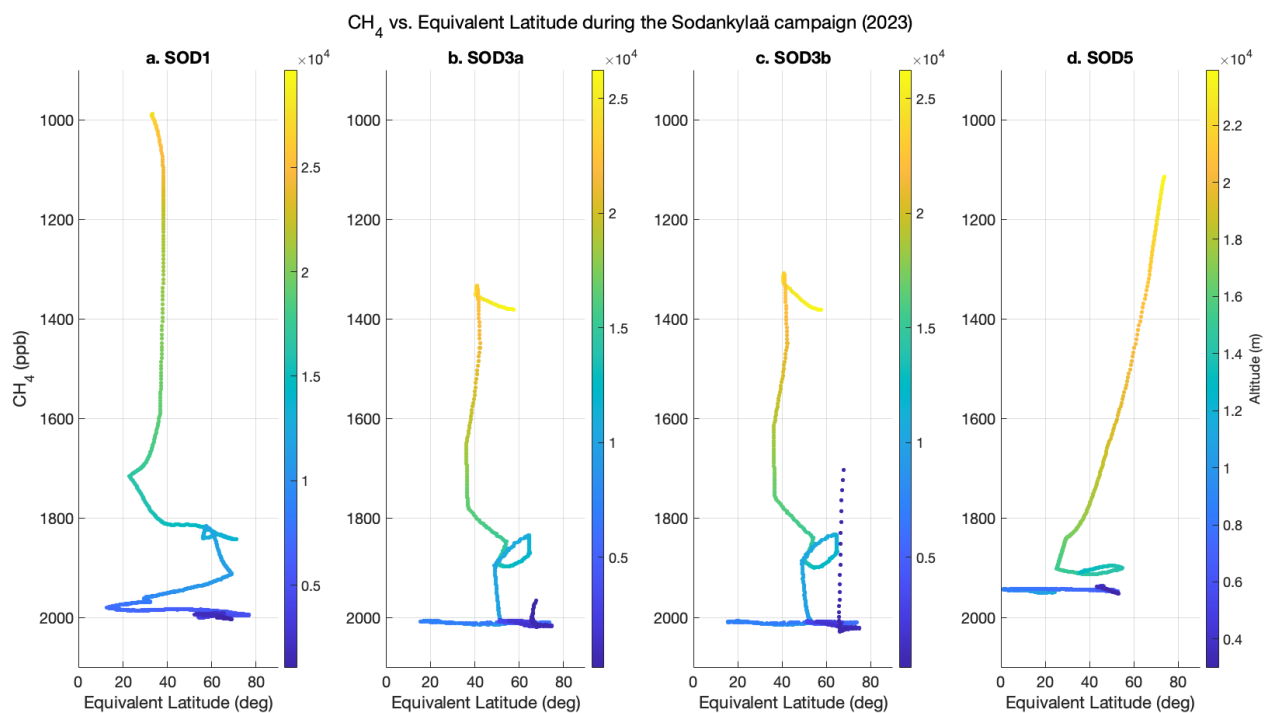


Figure S28: CH_4 AirCore profiles against equivalent latitude in SOD (2023).