



*Supplement of*

**A very limited role of tropospheric chlorine as a sink of the greenhouse gas methane**

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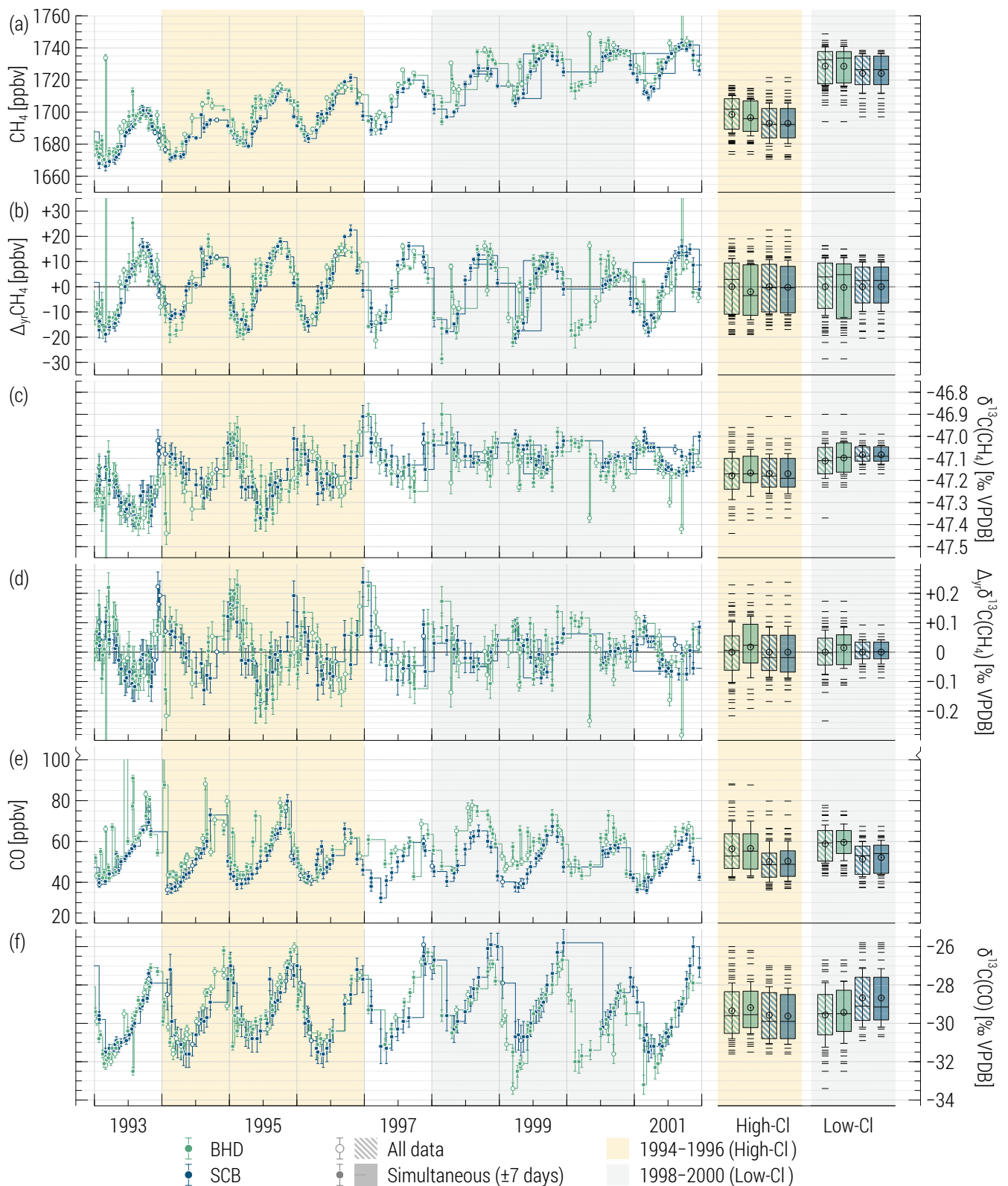


Fig. S2 Time series (left) and statistics (right, box-and-whisker plots) of the observations from Baring Head (BHD) and Scott Base (SCB) scrutinised in this study. Panels (a, c) present the mixing ratios and  $\delta^{13}\text{C}$  of  $\text{CH}_4$ ; panels (b, d) show anomalies with respect to the annual averages (denoted with “ $\Delta_{\text{yr}}$ ”). Panels (e, f) display the mixing ratios and  $\delta^{13}\text{C}$  of CO. The number of samples in each subset is presented in the manuscript (Fig. 1, panel (g)). Shaded areas denote the ETSH MBL high-Cl (orange shaded) and low-Cl (grey shaded) periods hypothesised by A07 (see text for details). Step lines navigate through the entire time series at each station. Boxes and whiskers present the median/interquartile range and  $\pm 1\sigma$  (of the population) of the selected data. Circles and minus symbols denote the averages and samples falling outside  $\pm 1\sigma$ , respectively. Solid symbols/boxes refer to the data when  $\text{CH}_4$  and CO samples were taken simultaneously (up to 7 days apart); hollow symbols/hatched boxes refer to all data.

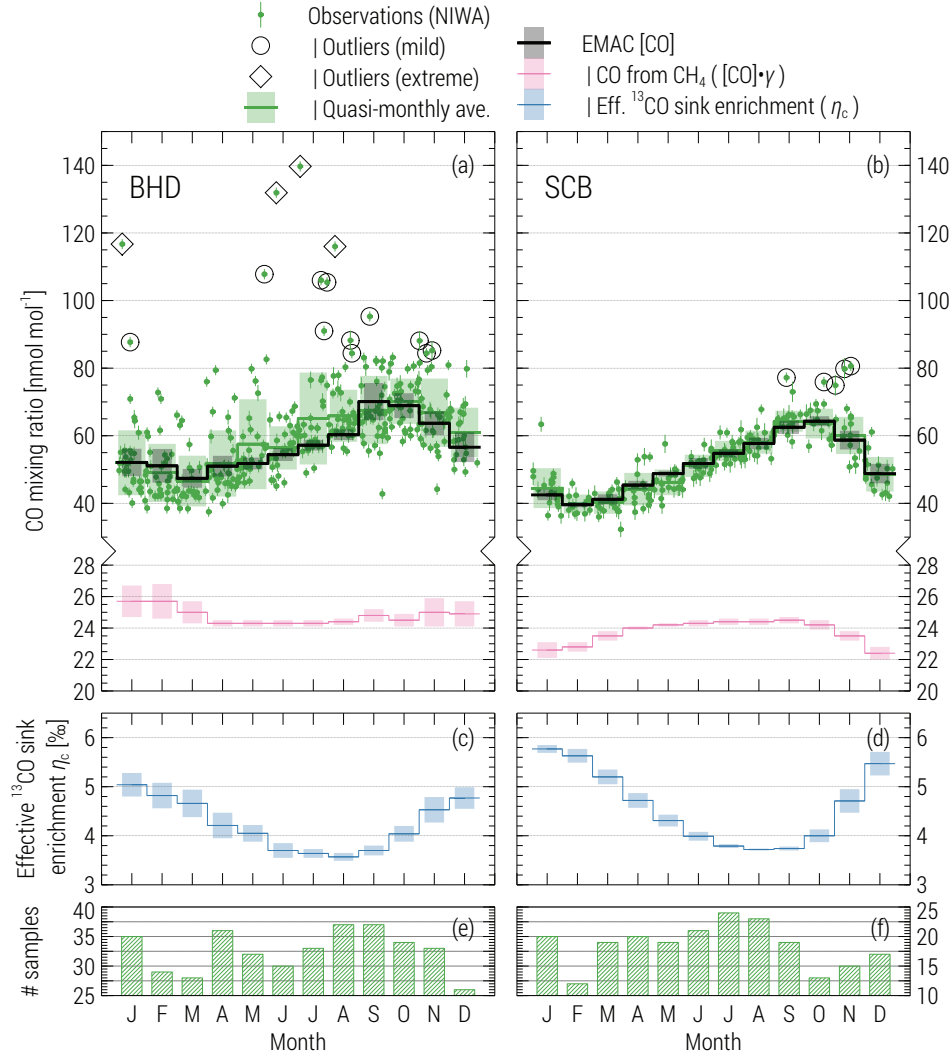


Fig. S3 Seasonal cycles CO mixing ratio at Baring Head (BHD, panel **a**) and Scott Base (SCB, panel **b**). Observations (entire data series plotted against day of year) are shown with symbols; circles and diamonds denote mild and extreme outliers (see Sect. 2.2 of the manuscript for details). Step lines refer to quasi-monthly averages derived from the observations (green) and from the EMAC model (1996–2005) for total CO (black) and its component derived from CH<sub>4</sub> oxidation (thin red line, lower scale). Panels **(c, d)** present the simulated effective sink  $^{13}\text{CO}$  enrichment, respectively. Vertical bars indicate  $\pm 1\sigma$  of the subsample used for quasi-monthly averages. Panels **(e, f)** show the number of samples in observational data. Mind the breaks and different scales of the ordinate axes.

Table S1 Annual average CO- and CH<sub>4</sub>-related integrals by domain simulated in EMAC for 1996–2005.

domain	zonal												
vertical	GLOB	ETNH	IT	ETSH	AR	AN	GLOB	ETNH	IT	ETSH	AR	AN	
	CO burden [Tg(C)]						Fraction of CO from CH <sub>4</sub> oxidation   γ [C/C]						
SRF	1.80	0.70	0.86	0.24	0.09	0.03	24.6%	17.1%	25.2%	44.8%	19.3%	46.0%	
BL	12.71	5.07	5.87	1.77	0.52	0.19	25.7%	17.5%	27.2%	44.5%	18.9%	45.8%	
MBL	6.39	1.67	3.31	1.40	0.20	0.05	32.5%	19.9%	33.5%	45.2%	19.0%	45.8%	
FT	135.71	46.82	68.13	20.75	7.45	2.42	29.5%	21.2%	31.0%	43.6%	20.1%	45.4%	
T	151.30	53.09	75.23	22.98	8.13	2.67	29.2%	20.7%	30.6%	43.7%	20.0%	45.5%	
TP	4.89	2.14	1.67	1.08	0.48	0.19	30.5%	23.9%	32.8%	40.3%	21.8%	43.6%	
LMS	15.64	6.71	4.25	4.68	1.45	0.84	40.7%	33.3%	44.7%	47.5%	30.5%	49.5%	
	Cl concentration [atoms cm <sup>-3</sup> ]						Yield of CO from CH <sub>4</sub> oxidation   λ [C/C]						
SRF	97	56	145	47	8	10	72%	72%	74%	62%	48%	51%	
BL	100	59	152	51	8	11	81%	83%	81%	73%	55%	62%	
MBL	110	59	165	49	8	9	47%	41%	45%	63%	103%	257%	
FT	269	163	352	152	36	42	96%	97%	95%	96%	98%	107%	
T	261	157	345	146	35	40	93%	95%	93%	93%	96%	104%	
TP	3151	1415	4945	1418	299	506	123%	119%	128%	121%	120%	120%	
LMS	16172	15892	15630	17260	15301	20021	102%	103%	101%	103%	103%	102%	
	CH <sub>4</sub> sink   S [Tg(C) yr <sup>-1</sup> ]						(via OH) [C/C]						
SRF	7.0	1.1	5.2	0.69	0.01	0.01	99.9%	99.9%	99.9%	99.8%	99.8%	99.8%	
BL	49.2	7.9	36.1	5.1	0.07	0.06	99.9%	99.9%	99.9%	99.8%	99.8%	99.8%	
MBL	32.7	2.9	25.9	3.9	0.03	0.01	99.9%	99.9%	99.9%	99.8%	99.8%	99.8%	
FT	313.8	49.5	227.8	36.5	2.22	1.01	99.7%	99.7%	99.7%	99.7%	99.7%	99.6%	
T	372.7	59.2	270.6	42.8	2.32	1.09	99.7%	99.7%	99.7%	99.7%	99.7%	99.6%	
TP	1.83	0.59	0.74	0.50	0.07	0.05	90.9%	94.2%	86.4%	93.7%	97.8%	96.5%	
LMS	20.2	5.5	9.1	5.59	0.67	0.67	53.4%	58.7%	47.9%	57.1%	64.7%	60.4%	
	(via O <sup>1</sup> D) [C/C]						(via Cl) [C/C]						
SRF	0.01%	0.01%	0.01%	0.01%	0.02%	0.04%	0.13%	0.11%	0.13%	0.14%	0.14%	0.19%	
BL	0.01%	0.02%	0.01%	0.01%	0.02%	0.05%	0.13%	0.12%	0.13%	0.15%	0.15%	0.19%	
MBL	0.01%	0.01%	0.01%	0.01%	0.02%	0.03%	0.14%	0.13%	0.14%	0.15%	0.15%	0.18%	
FT	0.07%	0.08%	0.07%	0.07%	0.06%	0.09%	0.25%	0.24%	0.25%	0.27%	0.22%	0.30%	
T	0.06%	0.07%	0.06%	0.06%	0.06%	0.09%	0.23%	0.22%	0.23%	0.25%	0.22%	0.29%	
TP	1.94%	1.13%	3.20%	1.05%	0.38%	0.40%	7.2%	4.7%	10.4%	5.2%	1.8%	3.1%	
LMS	33.0%	26.8%	40.3%	27.1%	18.6%	18.4%	13.7%	14.6%	11.8%	15.8%	16.7%	21.2%	
Domain abbreviations	zonal:	GLOB	Globe (90°S–90°N)				NH/SH	Northern/Southern Hemisphere					
		IT/ET	Intra/Extra-Tropics (separated at 23.4°N/°S)				AR/AN	Arctic/Antarctic (above 66°N/°S)					
	vertical:	SRF/TP	Surface (lowest model layer)				T	Troposphere (below the TP)					
		(M)BL	(Marine) Boundary Layer				FT	Free Troposphere (above the BL, below the TP)					
		TP	Tropopause				LMS	Lowermost Stratosphere (above the TP)					