

Importance of emissions-driven simulation of methane for representation of key atmospheric oxidants

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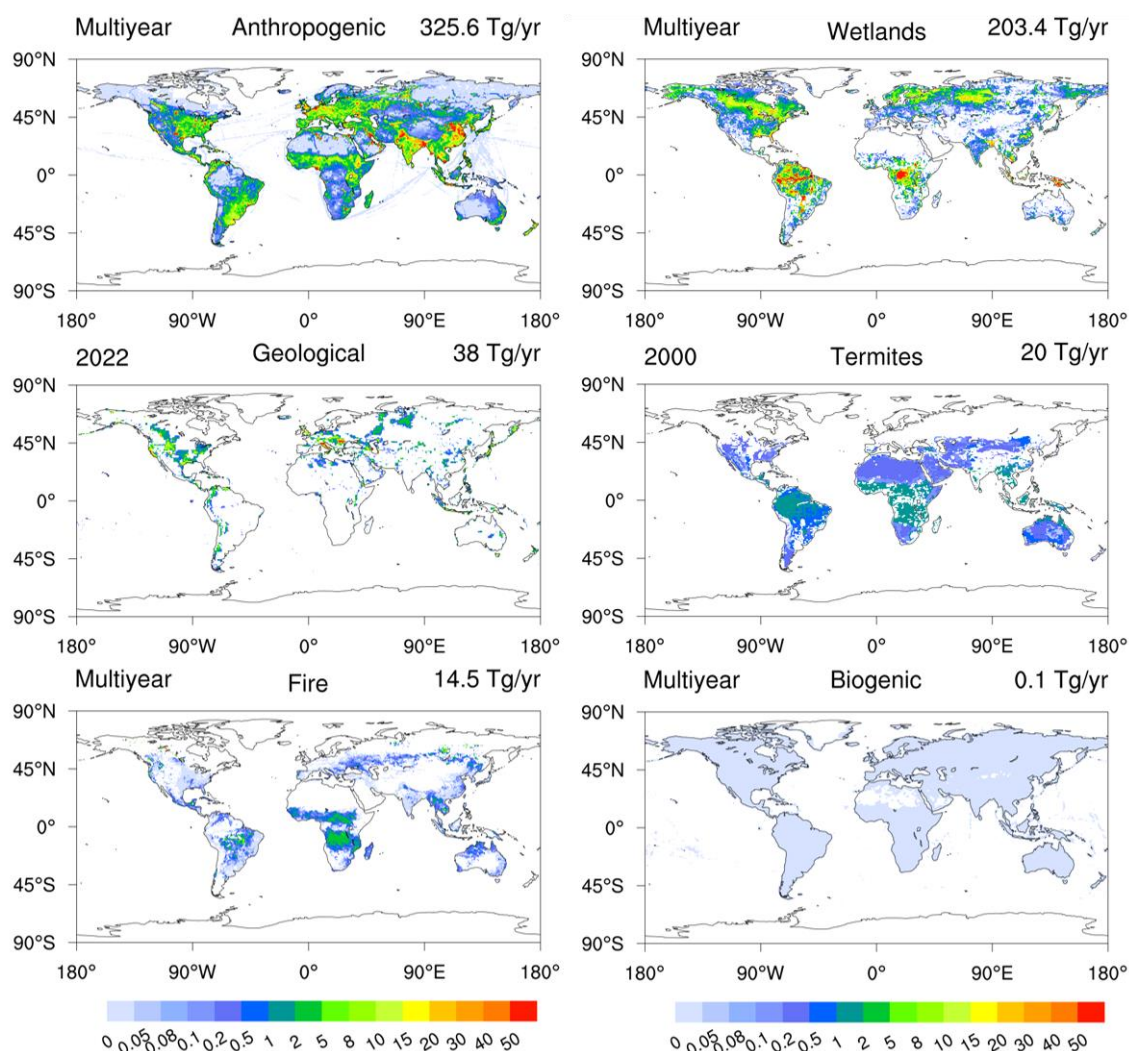


Figure S1. Distributions of CH₄ emissions (mg·m⁻²·d⁻¹) from anthropogenic, wetlands, geological, termite, fire and biogenic sources. Global totals (Tg·yr⁻¹) are shown in the top-right corner of each panel. The anthropogenic, wetlands, fire, and biogenic sources are multiyear averages from 2000 to 2017, The geological source is for 2022, and the termite source is for 2000.

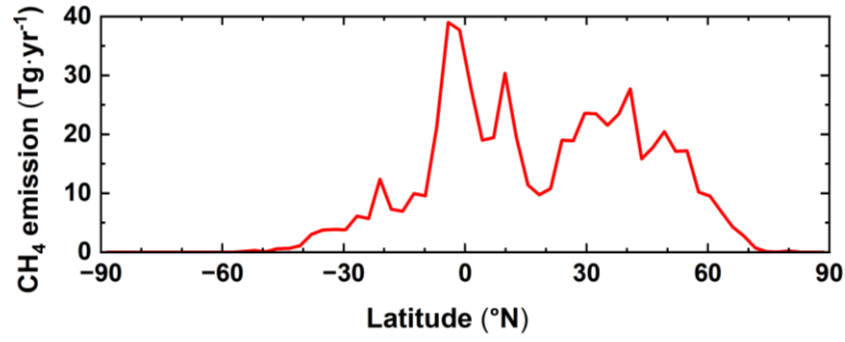


Figure S2. Latitudinal distribution of zonally integrated emissions for CH_4 ($\text{Tg}\cdot\text{yr}^{-1}$) average from 2000 to 2017.

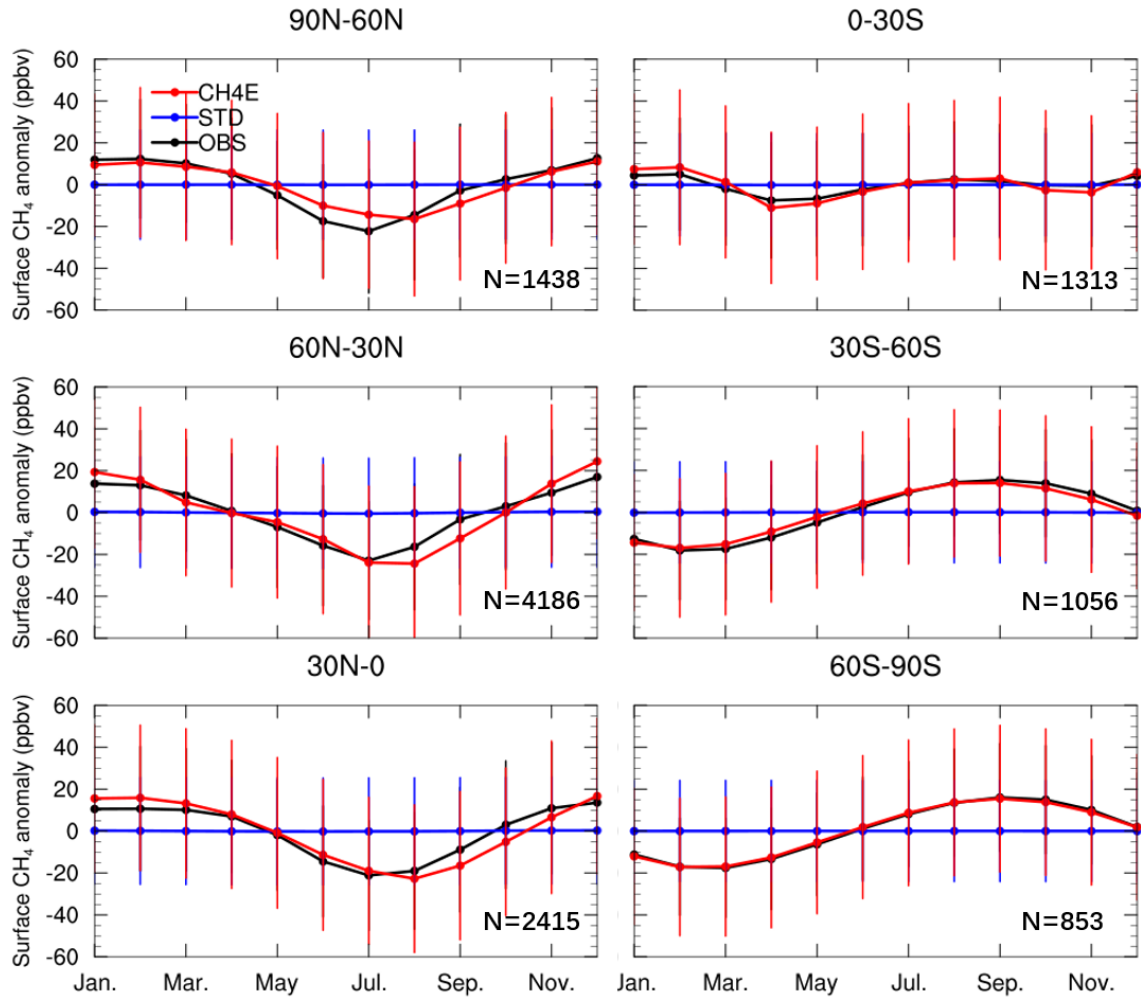


Figure S3. Multiyear mean seasonal cycle of surface CH_4 anomalies (ppbv) for observations (OBS; black), the prescribed surface concentration simulation (STD; blue), and the emission-driven simulation (CH4E; red) across six latitude bands during 2000–2017. For each dataset and latitude band, the anomaly for each calendar month is calculated as the corresponding multiyear monthly mean CH_4 mixing ratio minus the mean over all months during 2000–2017. Error bars show the standard deviation of CH_4 for each calendar month within the corresponding latitude band. N denotes the number of observational samples within each latitude band.

Table S1. Site details of surface CH₄

Station Name	Latitude/°N	Longitude/°E	Elevation/m	Dates	Country
Alert, Nunavut	82.45	-62.51	185	2000-2017	Canada
Ny-Alesund, Svalbard	78.91	11.89	474	2000-2017	Norway and Sweden
Summit	72.60	-38.42	3209.5	2000-2017	Greenland
Hydrometeorological Observatory of Tiksi	71.60	128.89	19	2011-2017	Russia
Barrow Atmospheric Baseline Observatory	71.32	-156.61	11	2000-2017	United States
Pallas-Sammaltunturi	67.97	24.12	565	2001-2017	Finland
Ocean Station M	66.00	2.00	0	2000-2009	Norway
Storhofdi, Vestmannaeyjar	63.40	-20.29	118	2000-2017	Iceland
Baltic Sea	55.35	17.22	3	2000-2011	Poland
Cold Bay, Alaska	55.21	-162.72	21.3	2000-2017	United States
Lac La Biche, Alberta	54.95	-112.47	540	2008-2013	Canada
Mace Head, County Galway	53.33	-9.90	5	2000-2017	Ireland
Shemya Island, Alaska	52.71	174.13	23	2000-2017	United States
Ochsenkopf	50.03	11.81	1022	2003-2017	Germany
Hohenpeissenberg	47.80	11.02	985	2006-2017	Germany
Hegyhatsal	46.96	16.65	248	2000-2017	Hungary
Ulaan Uul	44.45	111.10	1007	2000-2017	Mongolia
Black Sea, Constanta	44.18	28.67	0	2000-2011	Romania
Sary Taukum	44.08	76.87	595	2000-2009	Kazakhstan
Plateau Assy	43.25	77.88	2519	2000-2009	Kazakhstan
Centro de Investigacion de la Baja Atmosfera (CIBA)	41.81	-4.93	845	2009-2017	Spain
Trinidad Head, California	41.05	-124.15	107	2002-2017	United States
Shangdianzi	40.65	117.12	293	2009-2015	China
Niwot Ridge, Colorado	40.05	-105.59	3523	2000-2017	United States
Wendover, Utah	39.90	-113.72	1327	2000-2017	United States
Point Arena, California	38.96	-123.74	17	2000-2011	United States

Terceira Island, Azores	38.77	-27.38	19	2000-2017	Portugal
Tae-ahn Peninsula	36.74	126.13	16	2000-2017	Republic of Korea
Southern Great Plains, Oklahoma	36.61	-97.49	314	2002-2017	United States
Anmyeon-do	36.54	126.33	47	2013-2017	Republic of Korea
Mt. Waliguan	36.29	100.90	3810	2000-2017	Peoples Republic of China
Lampedusa	35.52	12.63	45	2006-2017	Italy
St. Davids Head, Bermuda	32.37	-64.65	12	2000-2010	United Kingdom
Tudor Hill, Bermuda	32.27	-64.88	30	2000-2017	United Kingdom
Moody, Texas	31.32	-97.33	251	2001-2010	United States
Weizmann Institute of Science at the Arava Institute, Ketura	29.97	35.06	151	2000-2017	Israel
Izana, Tenerife, Canary Islands	28.31	-16.50	2372.9	2000-2017	Spain
Sand Island, Midway	28.22	-177.37	4.6	2000-2017	United States
Key Biscayne, Florida	25.67	-80.16	1	2000-2017	United States
Lulin	23.47	120.87	2862	2006-2017	Taiwan
Assekrem	23.26	5.63	2710	2000-2017	Algeria
Dongsha Island	20.70	116.73	3	2010-2017	China Taiwan
Cape Kumukahi, Hawaii	19.56	-154.89	8	2000-2017	United States
Mauna Loa, Hawaii	19.54	-155.58	3397	2000-2017	United States
High Altitude Global Climate Observation Center	18.98	-97.31	4464	2009-2017	Mexico
Mariana Islands	13.39	144.66	0	2000-2017	Guam
Ragged Point	13.17	-59.43	15	2000-2017	Barbados
Christmas Island	1.70	-157.15	0	2000-2017	Republic of Kiribati
Mt. Kenya	-0.06	37.30	3644	2003-2017	Kenya
Bukit Kototabang	-0.20	100.32	845	2004-2017	Indonesia
Mahe Island	-4.68	55.53	2	2000-2017	Seychelles
Farol De Mae Luiza Lighthouse	-5.80	-35.19	50	2010-2017	Brazil
Ascension Island	-7.97	-14.40	85	2000-2017	United Kingdom

Arembepe, Bahia	-12.77	-38.17	1	2006-2010	Brazil
Tutuila	-14.25	-170.56	42	2000-2017	American Samoa
Gobabeb	-23.58	15.03	456	2000-2017	Namibia
Easter Island	-27.16	-109.43	47	2000-2017	Chile
Cape Point	-34.35	18.49	230	2010-2017	South Africa
Cape Grim, Tasmania	-40.68	144.69	94	2000-2017	Australia
Baring Head Station	-41.41	174.87	85	2000-2017	New Zealand
Crozet Island	-46.43	51.85	197	2000-2017	France
Ushuaia	-54.85	-68.31	12	2000-2017	Argentina
Drake Passage	-59.00	-64.69	0	2003-2017	N/A
Palmer Station, Antarctica	-64.77	-64.05	10	2000-2017	United States
Syowa Station, Antarctica	-69.01	39.59	14	2000-2017	Japan
Halley Station, Antarctica	-75.61	-26.21	30	2000-2017	United Kingdom
South Pole, Antarctica	-89.98	-24.80	2810	2000-2017	United States

Table S2. Site details of CH₄ vertical profile

Site ID	Latitude/°N	Longitude/°E	Elevation/m	Dates	Number of samples
acg	67.73	-155.94	15	2009-2017	22736
pfa	65.12	-147.83	264	1999-2017	65871
etl	54.34	-104.99	518	2005-2017	35923
esp	49.39	-126.54	7	2002-2017	43694
dnd	47.57	-98.87	462	2004-2016	34136
ulb	47.40	106.00	1360	2004-2009	5855
lef	45.97	-90.18	484	2000-2017	28175
cob	45.96	-90.97	440	2000	3328
fwi	44.66	-90.96	322	2004-2005	6838
nha	42.93	-70.56	0	2003-2017	42866
bgi	42.82	-94.41	348	2004-2005	6553
hfm	42.54	-72.17	360	2000-2007	26167
wbi	41.73	-91.36	239	2004-2017	44826
oil	41.28	-88.94	192	2004-2005	7674
thd	41.08	-124.24	0	2003-2017	36580
car	40.90	-104.80	1731	2000-2017	129393
bne	40.80	-97.18	464	2004-2011	17171
mci	40.31	-87.03	267	2007-2008	430
aoa	40.10	-88.56	211	2006-2009	26849
hil	40.07	-87.91	204	2004-2017	51334
bao	40.06	-105.01	1569.5	2008	149
cma	38.83	-74.32	0	2005-2017	37848
wgc	38.20	-121.50	19	2008-2009	339
sgp	36.61	-97.49	313	2000-2017	55807
mmp	33.34	-118.43	351	2004	262
sca	32.85	-79.55	0	2003-2017	51951
tgc	27.72	-96.73	0	2003-2017	42167
haa	21.23	-158.95	0	2000-2008	42246
hip	13.93	-154.91	0	2009-2011	28102
san	-2.85	-54.95	198	2000-2003	2610
ftl	-4.15	-38.28	50	2000-2003	1278
rta	-21.25	-159.83	0	2000-2017	42026
s2k	-24.26	28.50	880	2000	3322

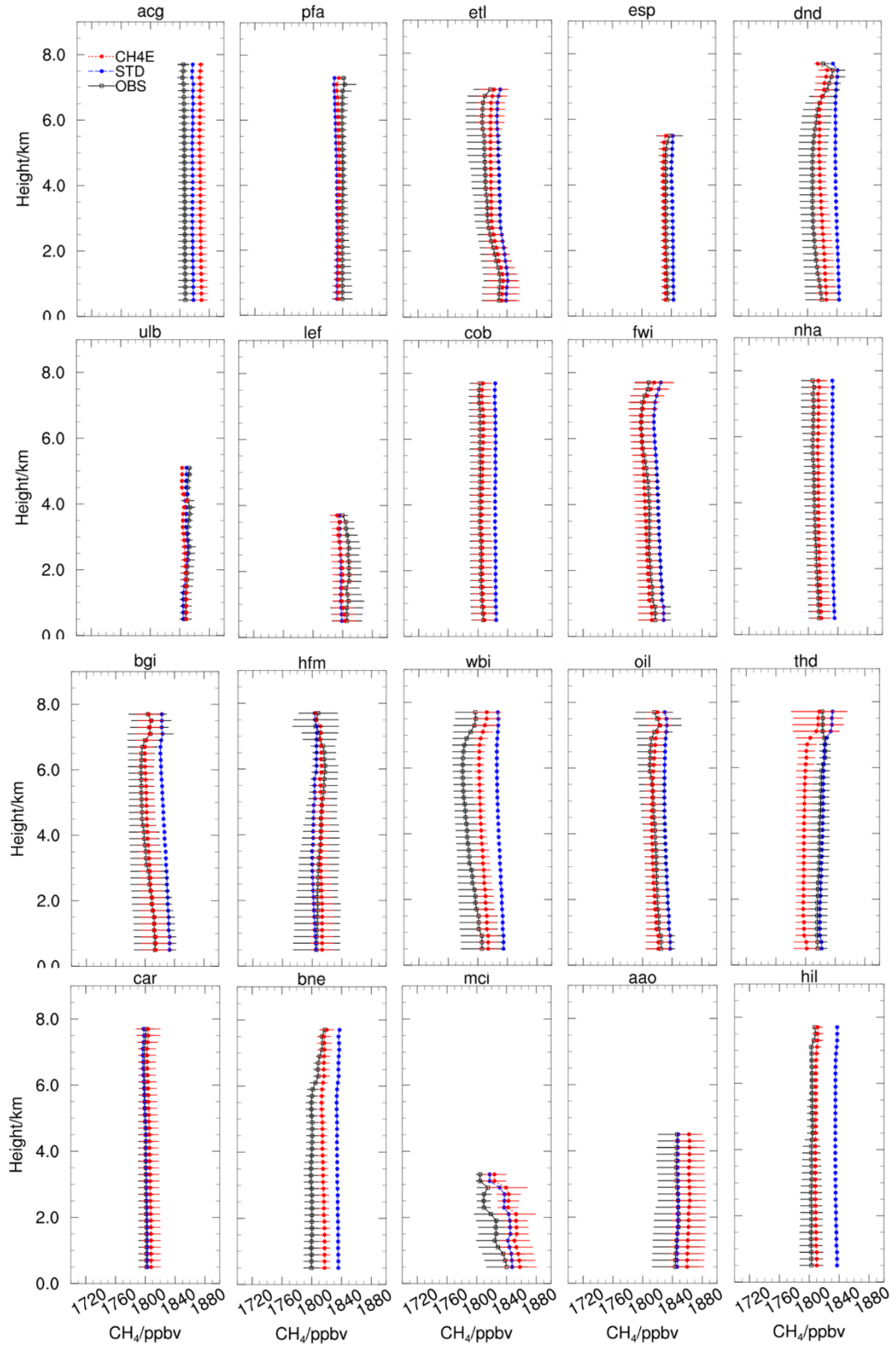


Figure S4. The vertical profile of CH₄ from aircraft observations (OBS), and from prescribed surface concentration (STD) and emission-driven (CH₄E) simulations (ppbv). Site data are shown in Table S2.

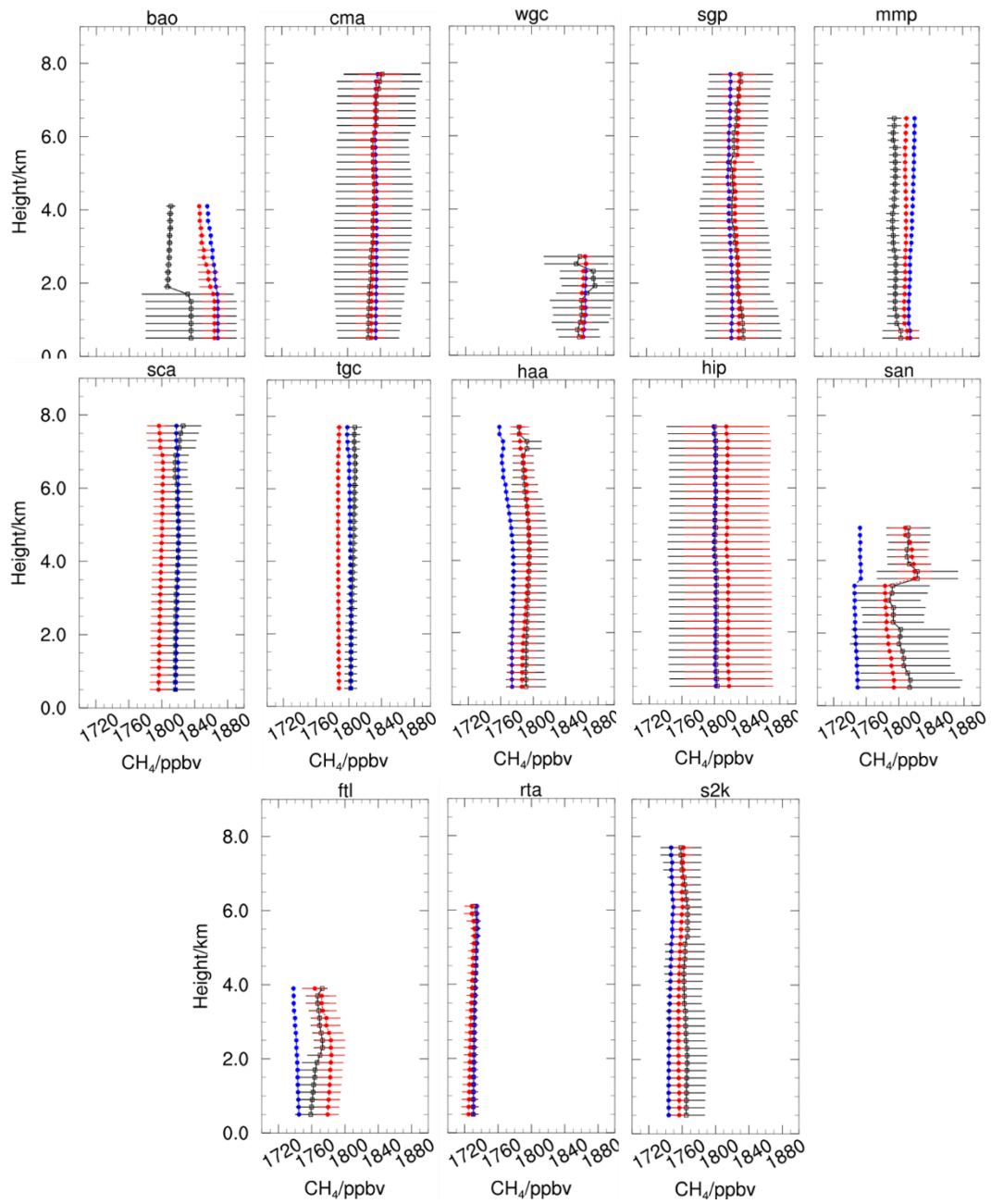


Figure S4. Continue.

Table S3. Key reactions leading to tropospheric production and loss of OH (Tmol year⁻¹) and their relative contribution (%) in the prescribed surface and emission-driven simulations.

	STD	CH4E
Production		
O(¹ D)+H ₂ O	91.5 (44.1%)	91.9 (44.1%)
HO ₂ +NO	68.3 (32.9%)	68.6 (32.9%)
HO ₂ +O ₃	26.3 (12.7%)	26.5 (12.7%)
H ₂ O ₂ +hv	17.1 (8.3%)	17.2 (8.3%)
VOCs,ROOH+hv	4.1 (2.0%)	4.1 (2.0%)
Other	0.4 (0.2%)	0.3 (0.2%)
Total	207.6 (100%)	208.6 (100%)
Loss		
OH+CO	-85.9 (41.4%)	-86.3 (41.4%)
OH+CH ₄	-31.8 (15.3%)	-32.1 (15.4%)
OH+VOCs, ROOH	-31.6 (15.2%)	-31.9 (15.3%)
OH+RCHO	-16.4 (7.9%)	-16.4 (7.9%)
OH+O ₃	-11.5 (5.5%)	-11.5 (5.5%)
OH+NO ₂	-2.3 (1.1%)	-2.3 (1.1%)
Other	-28.1 (13.6%)	-28.1 (13.5%)
Total	-207.5 (100 %)	-208.6 (100 %)