

Table S1. List of NOAA-GMD marine boundary layer (MBL) sites

Code	Name	State	Country	Latitude	Longitude	Elevation (m)
ALT	Alert	Nunavut	Canada	82.451	-62.507	190
AMS	Amsterdam Island	N/A	France	-37.798	77.538	55
ASC	Ascension Island	N/A	United Kingdom	-7.967	-14.4	85
AVI	St. Croix	Virgin Islands	United States	17.75	-64.75	3
BME	St. Davids Head	Bermuda	United Kingdom	32.368	-64.648	12
BMW	Tudor Hill	Bermuda	United Kingdom	32.265	-64.879	30
BRW	Barrow	Alaska	United States	71.323	-156.611	11
CBA	Cold Bay	Alaska	United States	55.21	-162.72	21.3
CGO	Cape Grim	Tasmania	Australia	-40.683	144.69	94
CHR	Christmas Island	N/A	Republic of Kiribati	1.7	-157.152	0
CMO	Cape Meares	Oregon	United States	45.478	-123.969	30
CRZ	Crozet Island	N/A	France	-46.434	51.848	197
EIC	Easter Island	N/A	Chile	-27.16	-109.428	47
GMI	Mariana Islands	N/A	Guam	13.386	144.656	0
HBA	Halley Station	Antarctica	United Kingdom	-75.605	-26.21	30
ICE	Storhofdi	Vestmannaeyjar	Iceland	63.4	-20.288	118
KEY	Key Biscayne	Florida	United States	25.665	-80.158	1
KUM	Cape Kumukahi	Hawaii	United States	19.52	-154.82	3
MBC	Mould Bay	Northwest Territories	Canada	76.247	-119.353	30
MHD	Mace Head	County Galway	Ireland	53.326	-9.899	5
MID	Sand Island	Midway	United States	28.21	-177.38	11
POC000*	Pacific Ocean (0 N)	N/A	N/A	0	-155	10
POCN05*	Pacific Ocean (5 N)	N/A	N/A	5	-151	10
POCN10*	Pacific Ocean (10 N)	N/A	N/A	10	-149	10

POCN15*	Pacific Ocean (15 N)	N/A	N/A	15	-145	10
POCN20*	Pacific Ocean (20 N)	N/A	N/A	20	-141	10
POCN25*	Pacific Ocean (25 N)	N/A	N/A	25	-139	10
POCN30*	Pacific Ocean (30 N)	N/A	N/A	30	-135	10
POCS05*	Pacific Ocean (5 S)	N/A	N/A	-5	-159	10
POCS10*	Pacific Ocean (10 S)	N/A	N/A	-10	-161	10
POCS15*	Pacific Ocean (15 S)	N/A	N/A	-15	-164	10
POCS20*	Pacific Ocean (20 S)	N/A	N/A	-20	-167	10
POCS25*	Pacific Ocean (25 S)	N/A	N/A	-25	-171	10
POCS30*	Pacific Ocean (30 S)	N/A	N/A	-30	-176	10
POCS35*	Pacific Ocean (35 S)	N/A	N/A	-35	180	10
PSA	Palmer Station	Antarctica	United States	-64.92	-64	10
RPB	Ragged Point	N/A	Barbados	13.165	-59.432	15
SHM	Shemya Island	Alaska	United States	52.711	174.126	23
SMO	Tutuila	N/A	American Samoa	-14.247	-170.564	42
SPO	South Pole	Antarctica	United States	-89.98	-24.8	2810
STM	Ocean Station M	N/A	Norway	66	2	0
SYO	Syowa Station	Antarctica	Japan	-69.013	39.59	14
USH	Ushuaia	N/A	Argentina	-54.848	-68.311	12
ZEP	Ny-Alesund	Svalbard	Norway and Sweden	78.907	11.888	474

\* Latitude values given for the POCN and POCS sites are the centers of latitude bands of +/- 2.5 degrees, and observations can fall anywhere within those bands.

Table S2. List of NOAA-GMD sites with at least 20-year observations

Code	Name	State	Country	Latitude	Longitude	Elevation (meters)
ALT	Alert	Nunavut	Canada	82.451	-62.507	190
ASC	Ascension Island	N/A	United Kingdom	-7.967	-14.4	85
ASK	Assekrem	N/A	Algeria	23.262	5.632	2710
AZR	Terceira Island	Azores	Portugal	38.766	-27.375	19
BMW	Tudor Hill	Bermuda	United Kingdom	32.265	-64.879	30
BRW	Barrow	Alaska	United States	71.323	-156.611	11
CBA	Cold Bay	Alaska	United States	55.21	-162.72	21.3
CGO	Cape Grim	Tasmania	Australia	-40.683	144.69	94
CHR	Christmas Island	N/A	Republic of Kiribati	1.7	-157.152	0
CRZ	Crozet Island	N/A	France	-46.434	51.848	197
EIC	Easter Island	N/A	Chile	-27.16	-109.428	47
GMI	Mariana Islands	N/A	Guam	13.386	144.656	0
HBA	Halley Station	Antarctica	United Kingdom	-75.605	-26.21	30
HUN	Hegyhatsal	N/A	Hungary	46.95	16.65	248
ICE	Storhofdi	Vestmannaeyjar	Iceland	63.4	-20.288	118
IZO	Izana	Tenerife	Spain	28.309	-16.499	2372.9
KEY	Key Biscayne	Florida	United States	25.665	-80.158	1
KUM	Cape Kumukahi	Hawaii	United States	19.52	-154.82	3
LEF	Park Falls	Wisconsin	United States	45.945	-90.273	472
MHD	Mace Head	County Galway	Ireland	53.326	-9.899	5
MID	Sand Island	Midway	United States	28.21	-177.38	11
MLO	Mauna Loa	Hawaii	United States	19.536	-155.576	3397
NWR	Niwot Ridge	Colorado	United States	40.053	-105.586	3523
POCN00*	Pacific Ocean (0 N)	N/A	N/A	0	-155	10

POCN05*	Pacific Ocean (5 N)	N/A	N/A	5	-151	10
POCN10*	Pacific Ocean (10 N)	N/A	N/A	10	-149	10
POCN15*	Pacific Ocean (15 N)	N/A	N/A	15	-145	10
POCN20*	Pacific Ocean (20 N)	N/A	N/A	20	-141	10
POCN25*	Pacific Ocean (25 N)	N/A	N/A	25	-139	10
POCN30*	Pacific Ocean (30 N)	N/A	N/A	30	-135	10
POCS05*	Pacific Ocean (5 S)	N/A	N/A	-5	-159	10
POCS10*	Pacific Ocean (10 S)	N/A	N/A	-10	-161	10
POCS15*	Pacific Ocean (15 S)	N/A	N/A	-15	-164	10
POCS20*	Pacific Ocean (20 S)	N/A	N/A	-20	-167	10
POCS25*	Pacific Ocean (25 S)	N/A	N/A	-25	-171	10
POCS30*	Pacific Ocean (30 S)	N/A	N/A	-30	-176	10
PSA	Palmer Station	Antarctica	United States	-64.92	-64	10
RPB	Ragged Point	N/A	Barbados	13.165	-59.432	15
SEY	Mahe Island	N/A	Seychelles	-4.682	55.532	2
SHM	Shemya Island	Alaska	United States	52.711	174.126	23
SMO	Tutuila	N/A	American Samoa	-14.247	-170.564	42
SPO	South Pole	Antarctica	United States	-89.98	-24.8	2810
STM	Ocean Station M	N/A	Norway	66	2	0
SYO	Syowa Station	Antarctica	Japan	-69.013	39.59	14
TAP	Tae-ahn Peninsula	N/A	Republic of Korea	36.738	126.133	16

USH	Ushuaia	N/A	Argentina	-54.848	-68.311	12
UTA	Wendover	Utah	United States	39.902	-113.718	1327
UUM	Ulaan Uul	N/A	Mongolia	44.452	111.096	1007
WIS	Weizmann Institute of Science at the Arava Institute	Ketura	Israel	29.965	35.06	151
WLG	Mt. Waliguan	N/A	Peoples Republic of China	36.288	100.896	3810
ZEP	Ny-Alesund	Svalbard	Norway and Sweden	78.907	11.888	474

\* Latitude values given for the POCN and POCS sites are the centers of latitude bands of +/- 2.5 degrees, and observations can fall anywhere within those bands.

## GMD MBL Stations

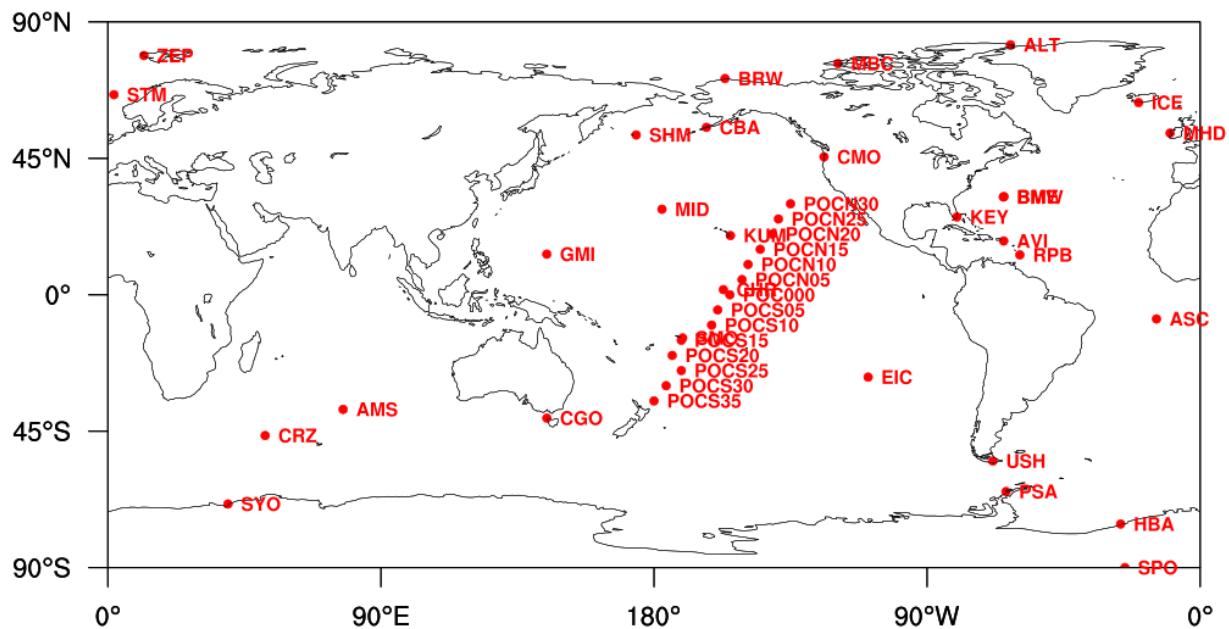


Figure S1. NOAA-GMD marine boundary layer (MBL) sites selected for background methane calculation.

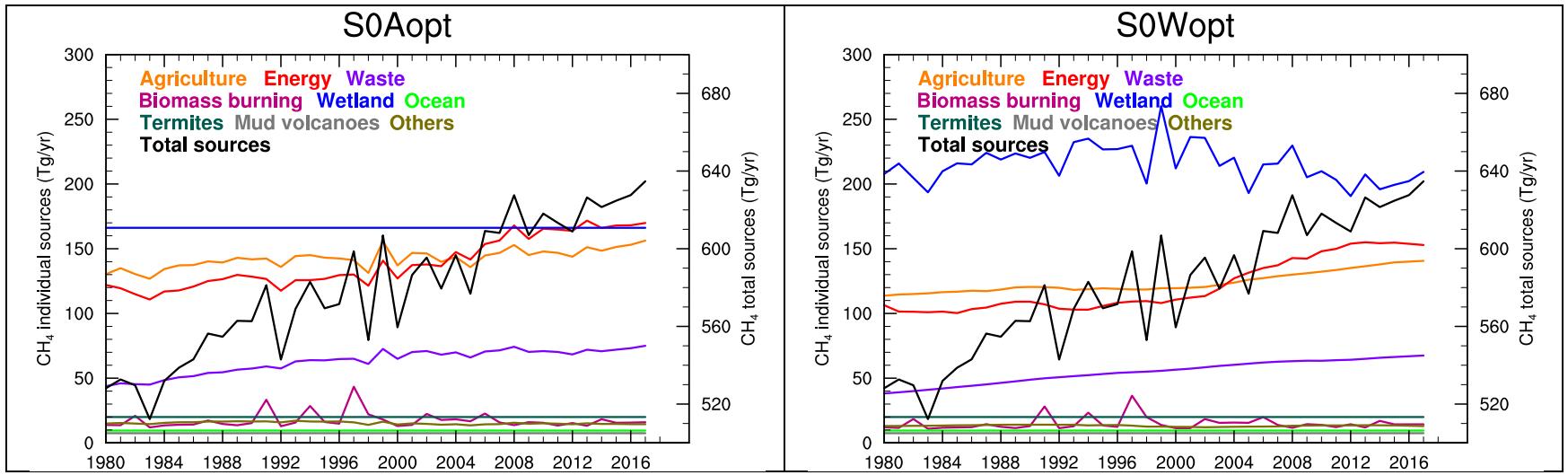
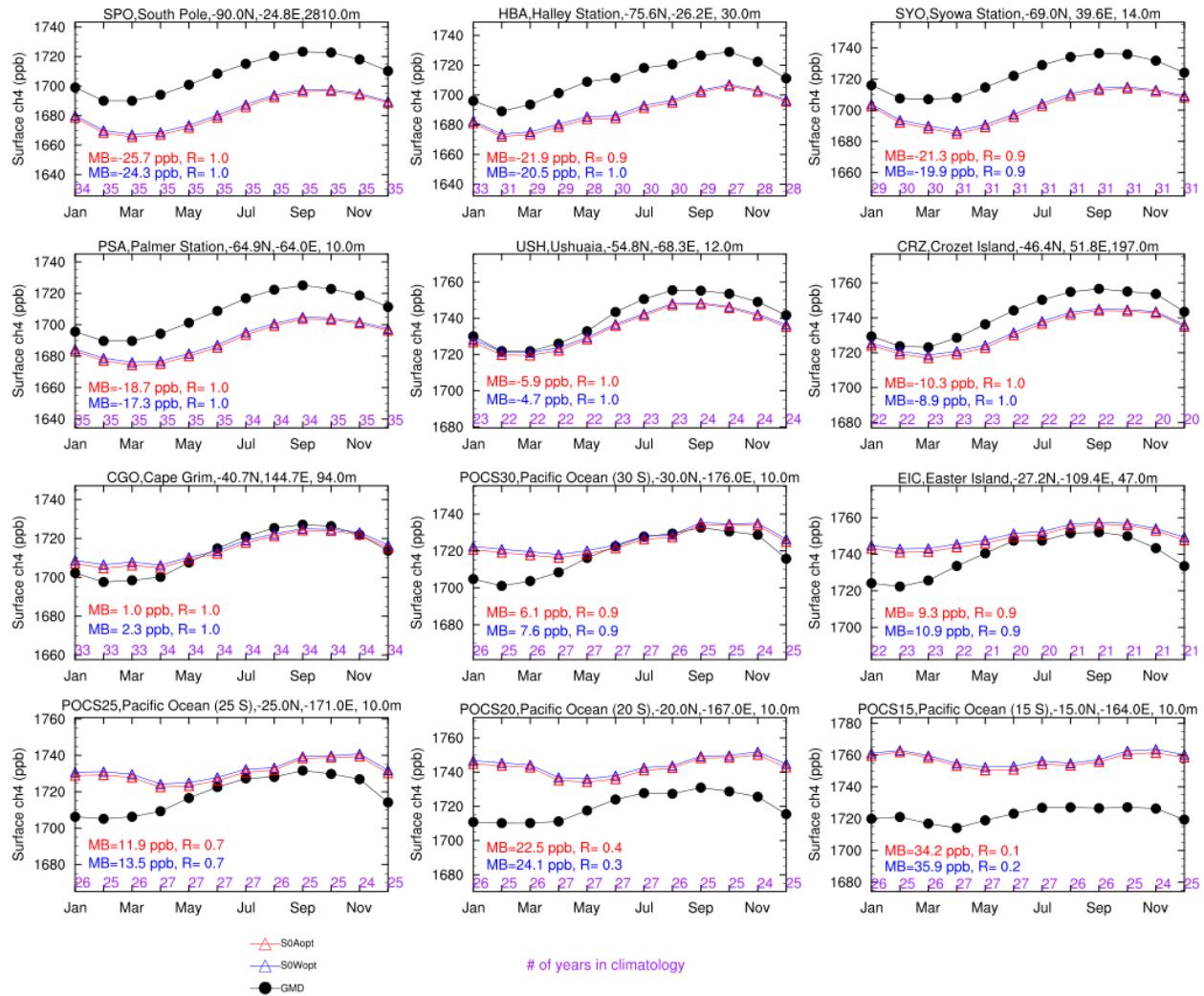
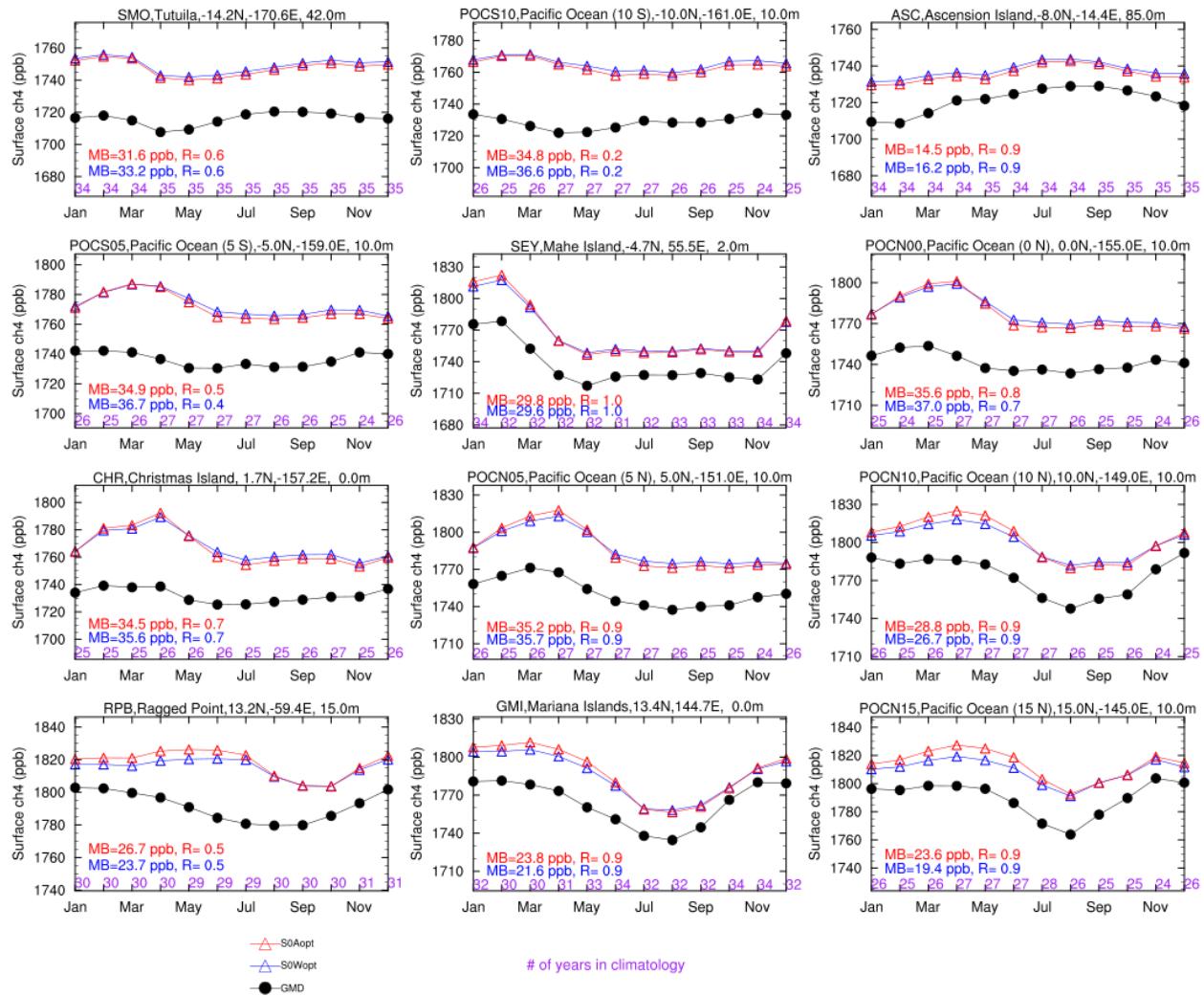
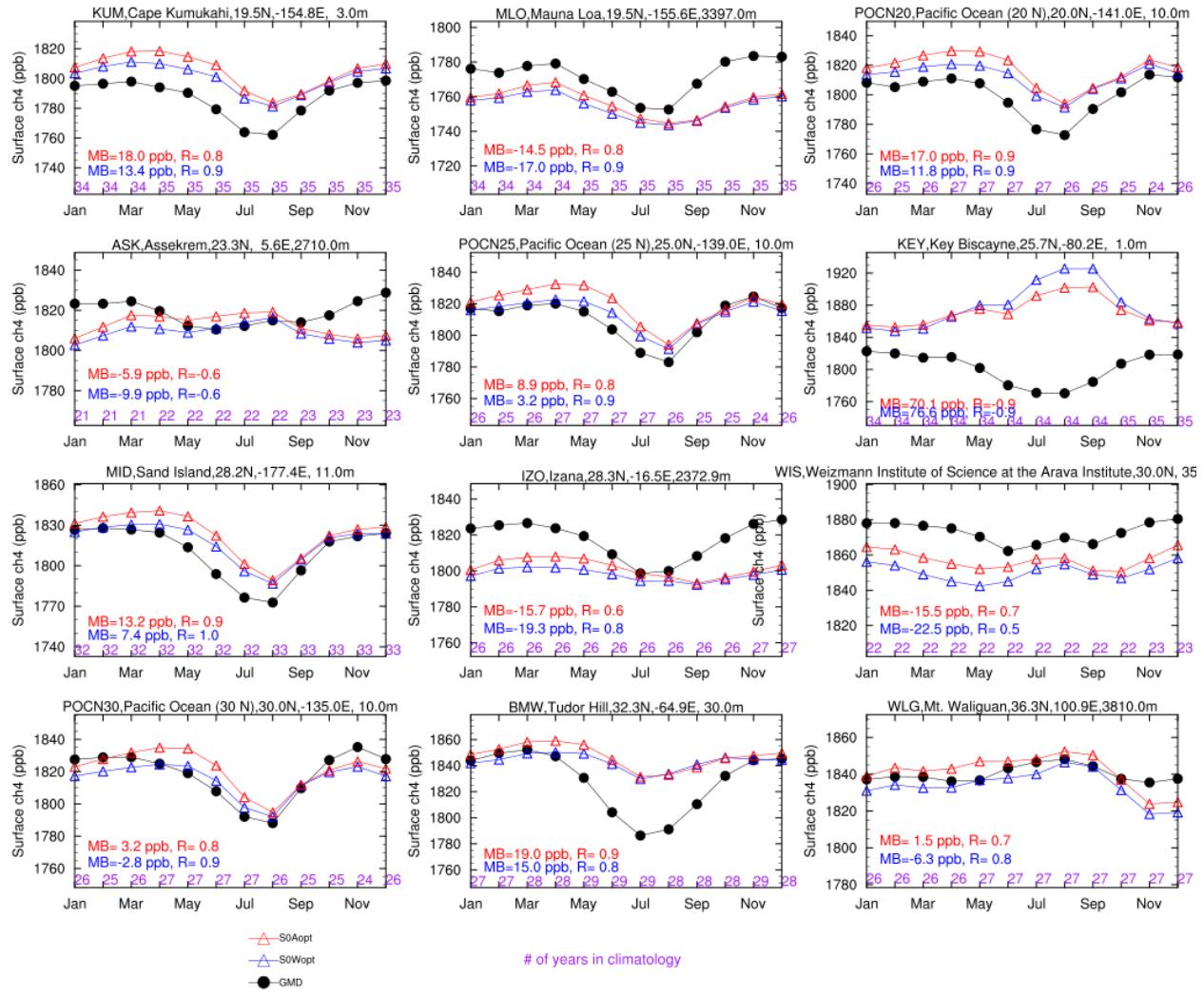
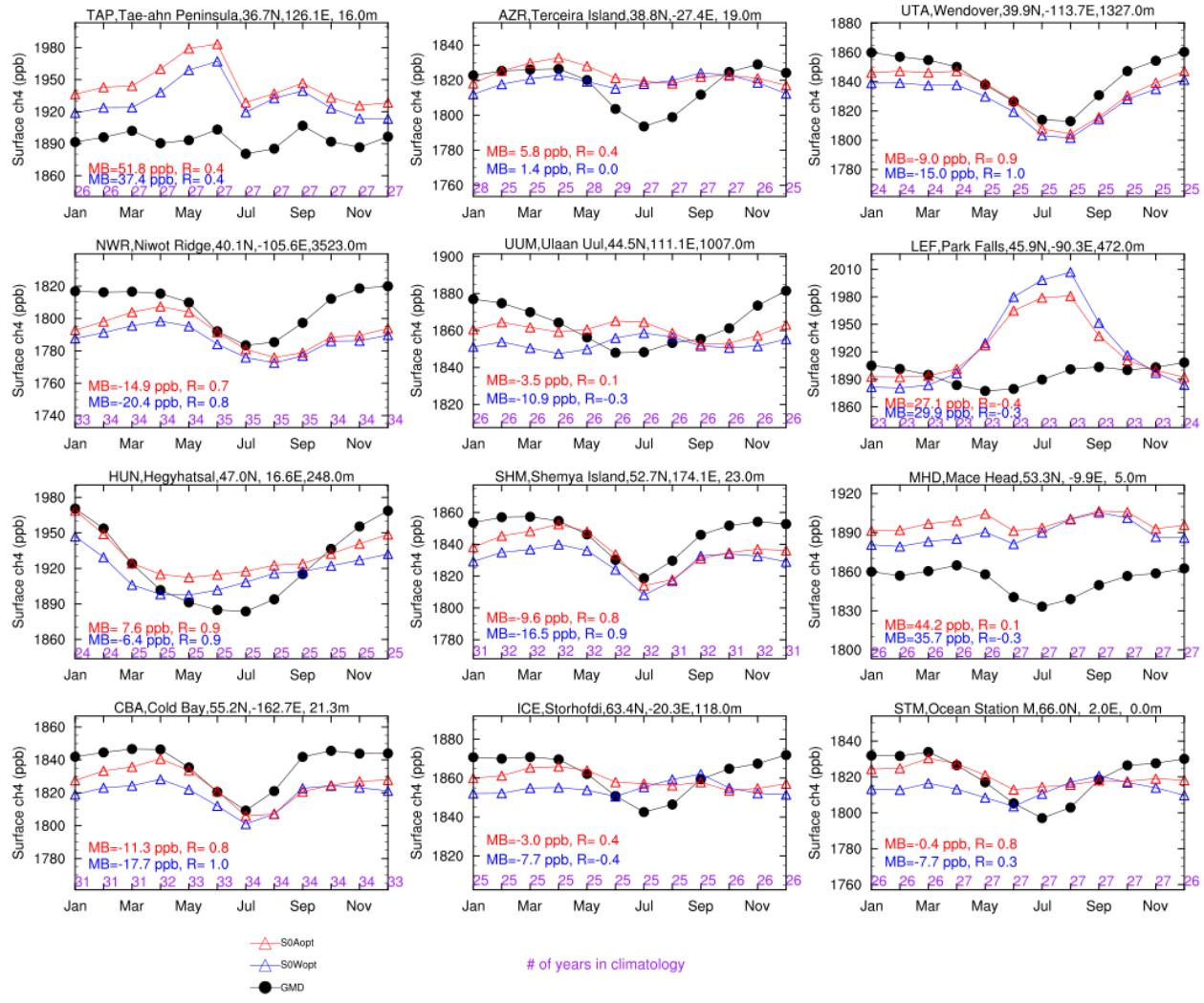


Figure S2. Time series of optimized methane emissions on anthropogenic sectors (S0Aopt, left) and wetland sector (S0Wopt, right) for the period of 1980-2017. The emissions for major sectors are shown on the left y axis, including agriculture sector, energy production sector, waste sector, biomass burning sector, wetland sector, ocean and near-shore fluxes, termites, mud volcanoes, and other sources (i.e., industrial processes, surface transportation, international shipping, residential, commercial, and others). The total methane emissions from the initial emission inventories (black line) are shown on the right y axis.









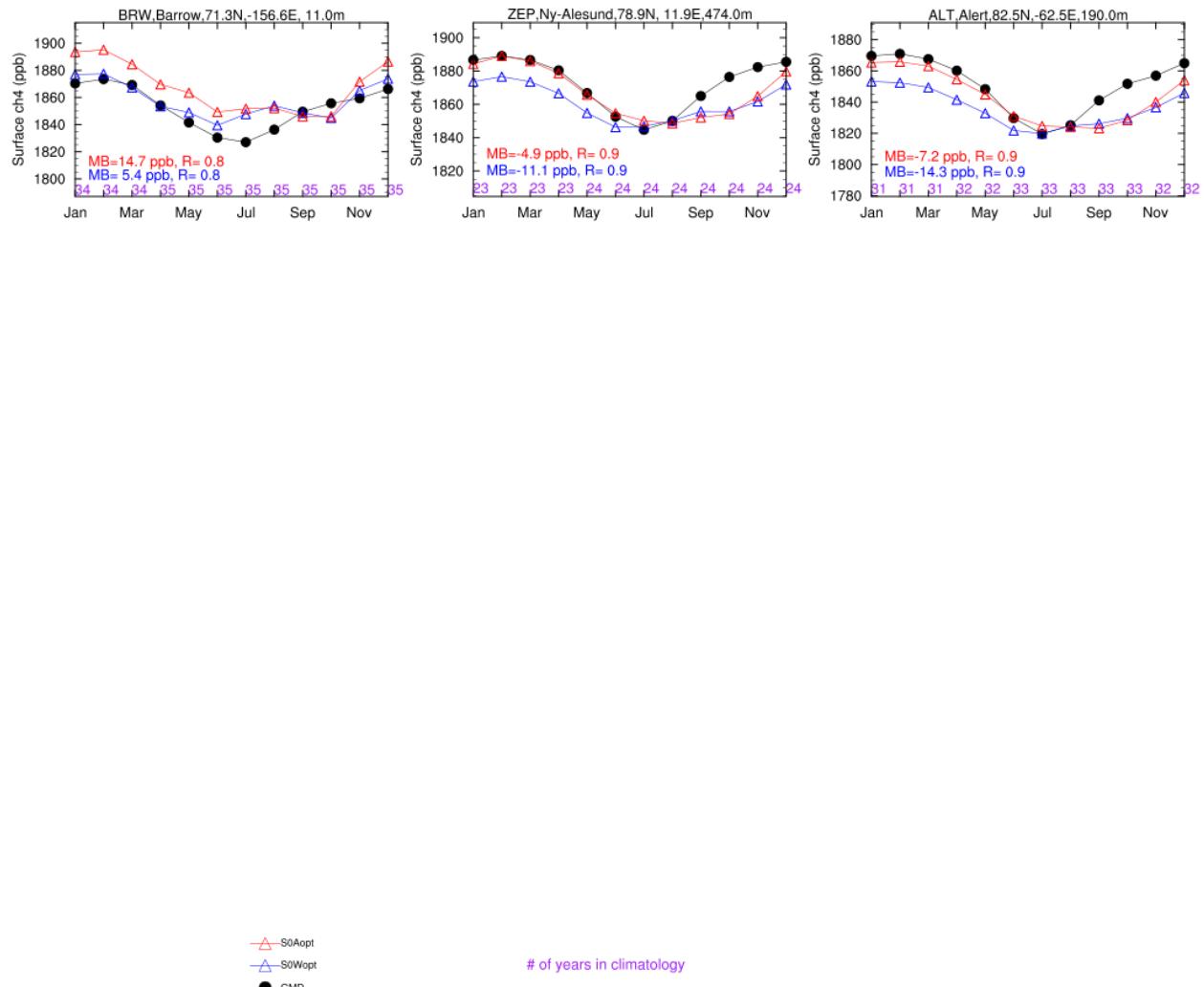
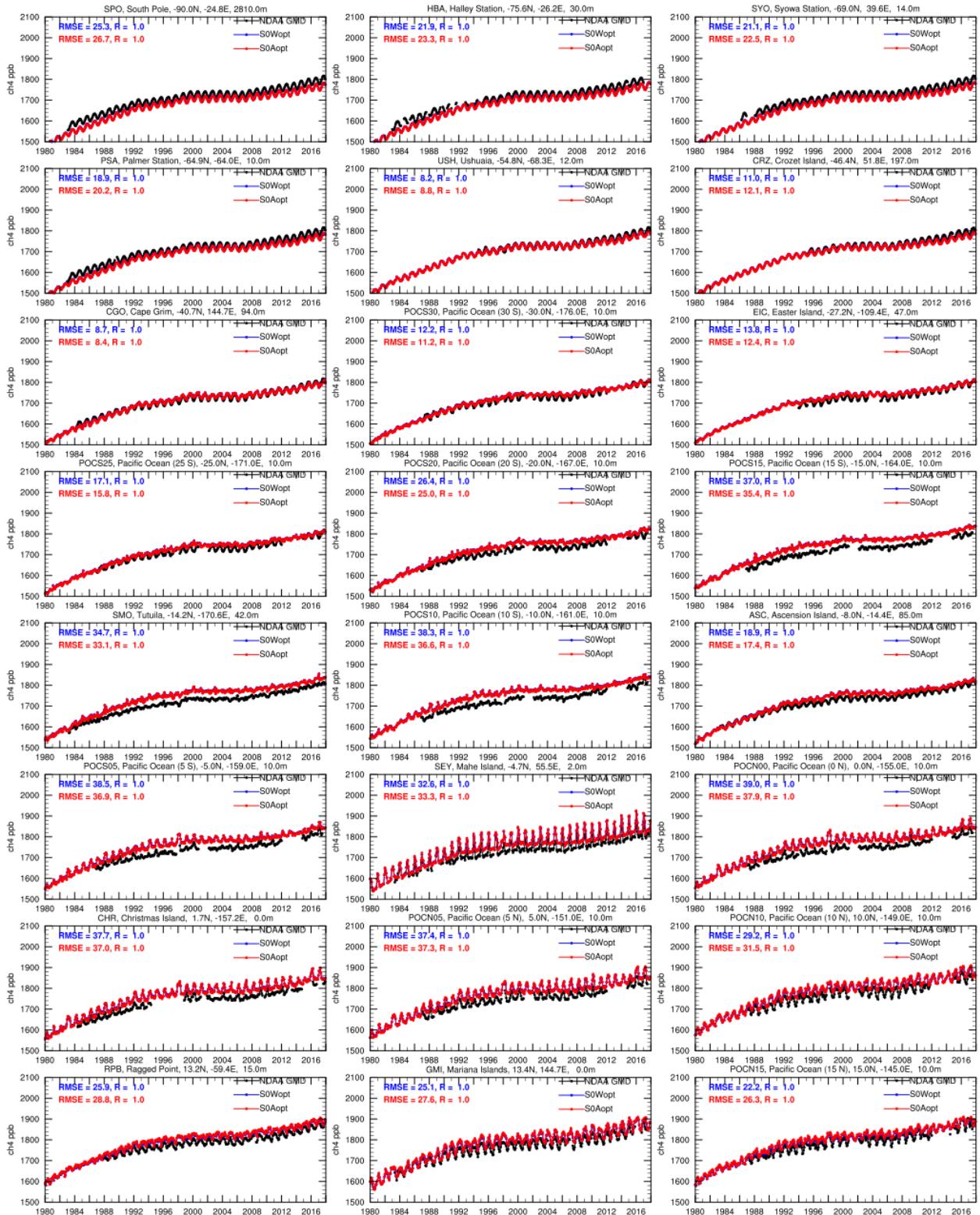
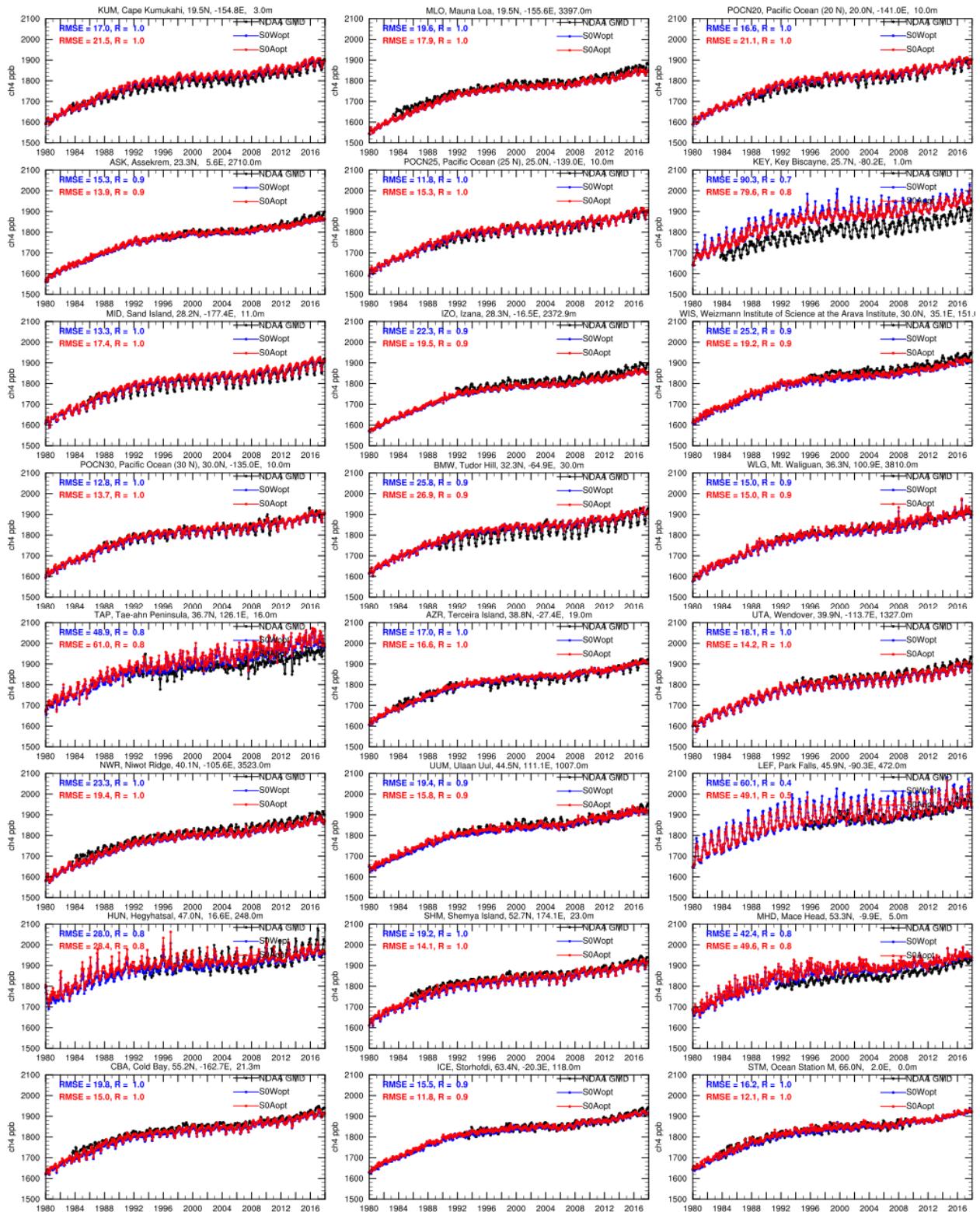


Figure S3. Comparisons of methane seasonal cycles against NOAA-GMD observations.





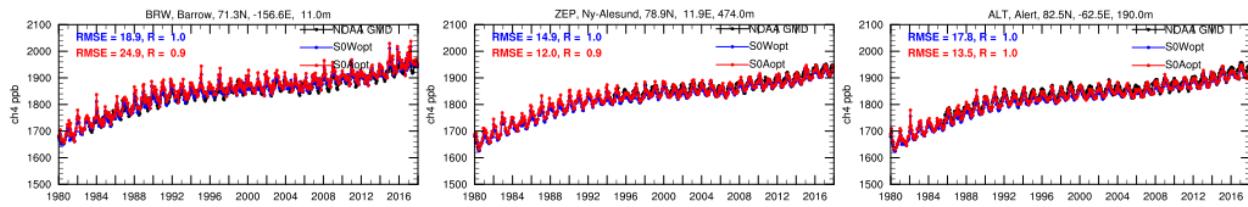


Figure S4. Comparisons of surface CH<sub>4</sub> dry-mole fractions to NOAA-GMD observations.

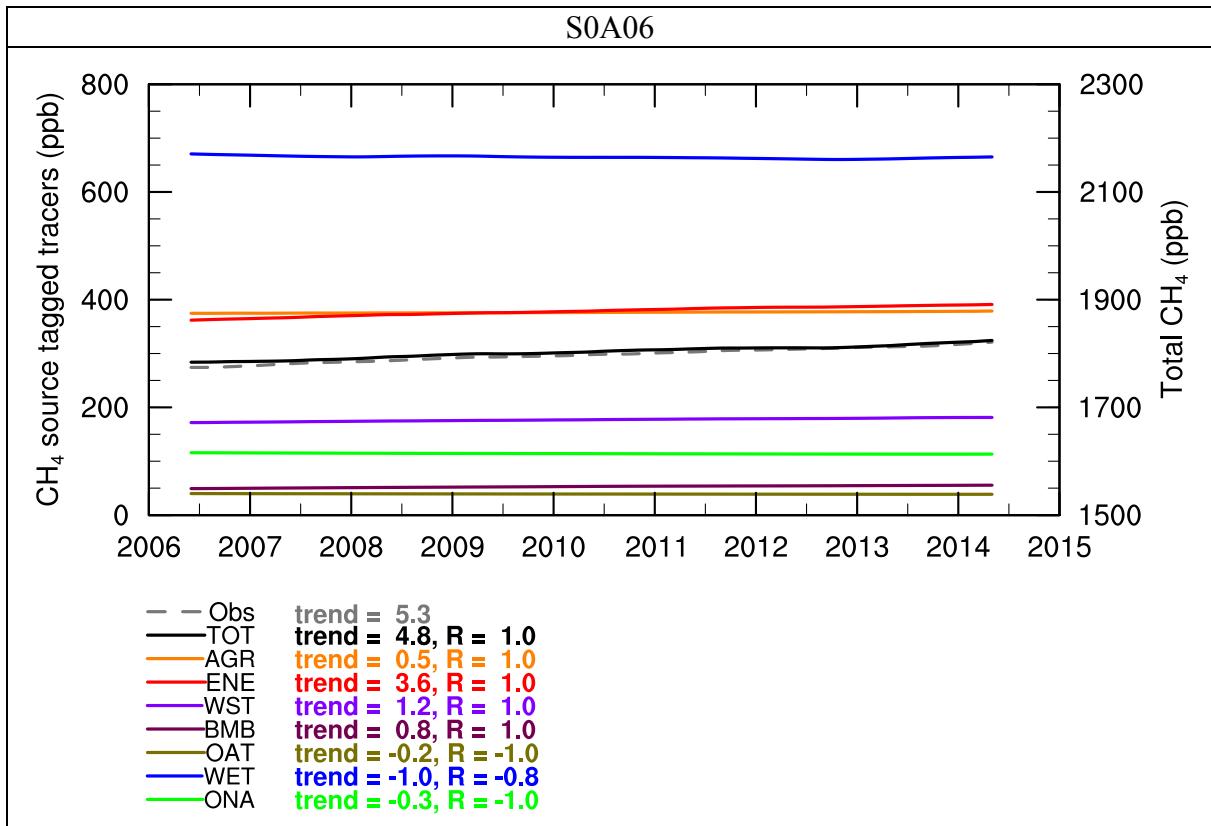


Figure S5. Global deseasonalized methane trends for source tagged tracers (left y axis) and total methane (TOT, right y axis) for sensitivity simulation “S0A06”. The source tagged tracers include tracer for agriculture sector (AGR), energy sector, (ENE), waste sector (WST), biomass burning sector (BMB), other anthropogenic sectors (OAT), wetland sector (WET), and other natural sectors (ONA). The gray dashed line represents total methane trend from NOAA-GMD observations. The trends (i.e., linear growth rates,  $\text{ppb yr}^{-1}$ ) for the period of 2007-2014 are shown below the figure. The correlation coefficient (i.e., R) for TOT is compared to observations and for other source tagged tracers is compared to total methane.

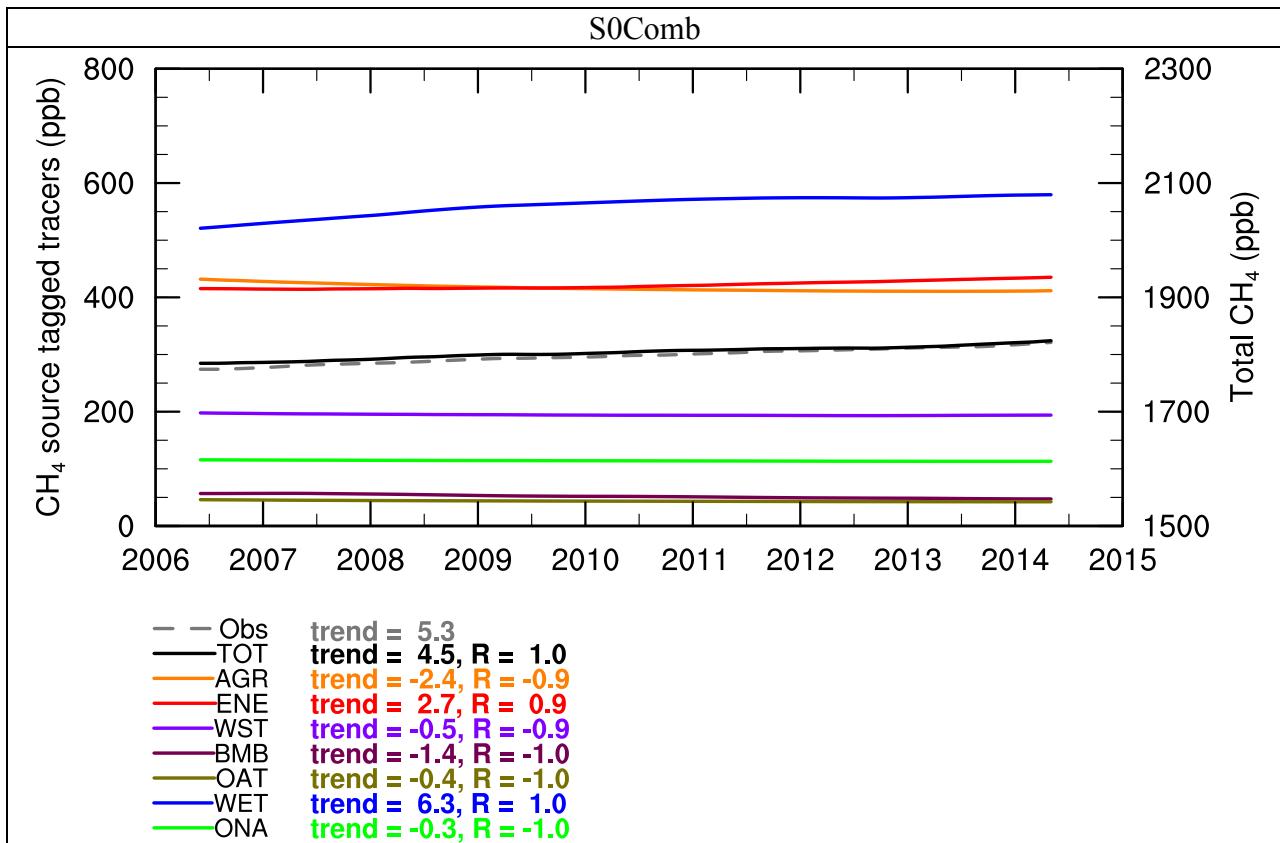


Figure S6. Global deseasonalized methane trends for source tagged tracers (left y axis) and total methane (TOT, right y axis) for sensitivity simulation “S0Comb”. The source tagged tracers include tracer for agriculture sector (AGR), energy sector, (ENE), waste sector (WST), biomass burning sector (BMB), other anthropogenic sectors (OAT), wetland sector (WET), and other natural sectors (ONA). The gray dashed line represents total methane trend from NOAA-GMD observations. The trends (i.e., linear growth rates, ppb yr<sup>-1</sup>) for the period of 2007-2014 are shown below the figure. The correlation coefficient (i.e., R) for TOT is compared to observations and for other source tagged tracers is compared to total methane.