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Supplement of

Three-dimensional methane distribution simulated with FLEXPART 8-CTM-1.1 constrained with observation data

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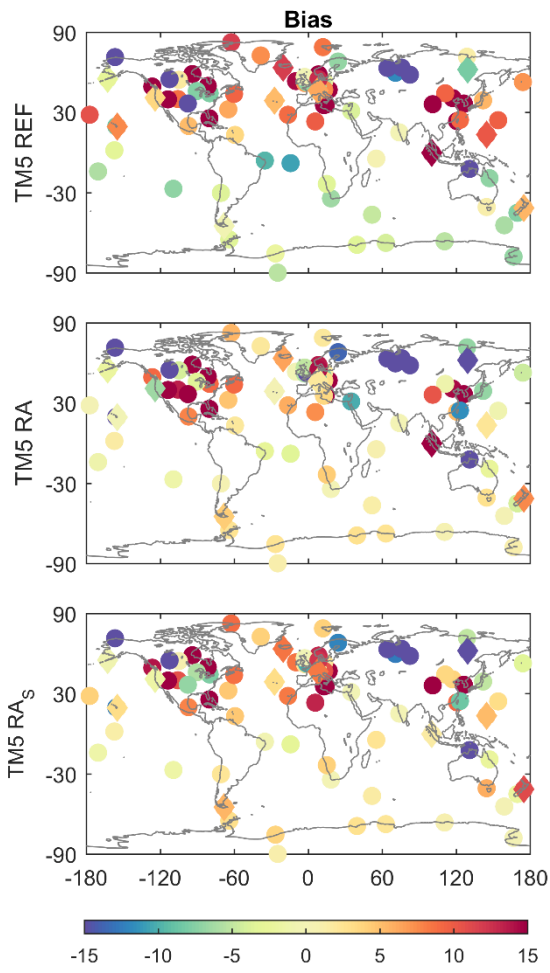
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Table S1 Names, coordinates and data providers of all sites that were included in this study as nudging or validation station.

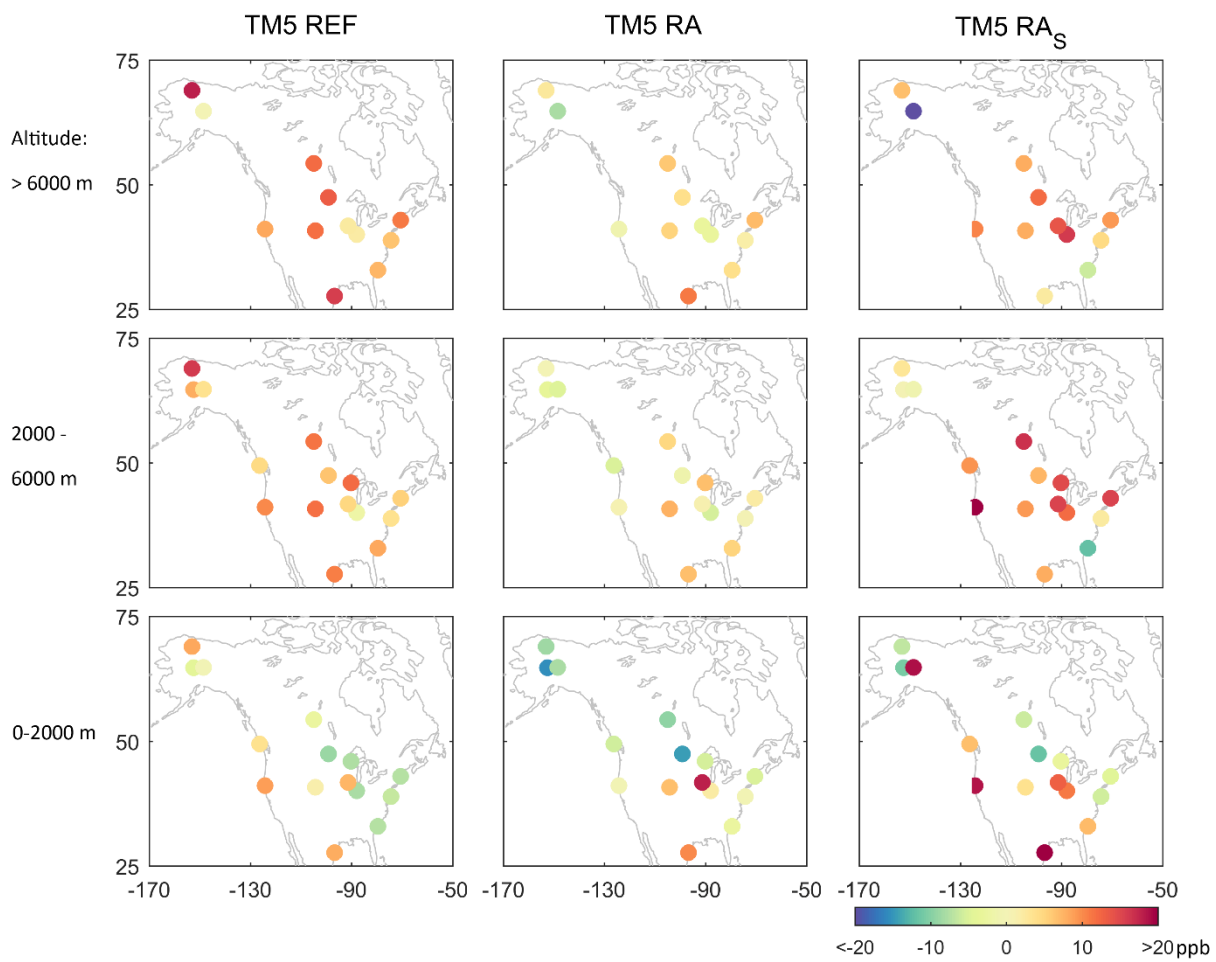
Site ID	Site name	Latitude (°N)	Longitude (°E)	Altitude (m)	Contributor(s)
ALT	Alert	82.45	-62.52	210	CSIRO/ECCC/NOAA_ESRL
AMY	Anmyeon-do	36.53	126.32	47	KMA
ARH	Arrival Heights	-77.80	166.67	184	NIWA
ASC	Ascension Island	-7.92	-14.42	54	NOAA_ESRL
ASK	Assekrem	23.27	5.63	2710	NOAA_ESRL
AZR	Terceira Island	38.77	-27.37	40	NOAA_ESRL
BHD	Baring Head	-41.41	174.87	85	NOAA_ESRL
BIR	Birkenes	58.39	8.25	219	NILU
BKT	Bukit Koto Tabang	-0.20	100.32	864.5	BMKG Empa/NOAA_ESRL
BMW	Tudor Hill	32.27	-64.87	30	NOAA_ESRL
BRW	Barrow	71.32	-156.60	11	NOAA_ESRL
CBA	Cold Bay	55.20	-162.72	25	NOAA_ESRL
CFA	Cape Ferguson	-19.28	147.05	2	CSIRO
CGO	Cape Grim	-40.68	144.68	94	AGAGE/CSIRO/NOAA_ESRL
CHL	Churchill	58.75	-94.07	35	ECCC
CHR	Christmas Island	1.70	-157.17	3	NOAA_ESRL
CMN	Monte Cimone	44.18	10.70	2165	UNIURB
CPT	Cape Point	-34.35	18.48	230	SAWS/NOAA_ESRL
CRI	Cape Rama	15.08	73.83	60	CSIRO
CRZ	Crozet	-46.45	51.85	120	NOAA_ESRL
CYA	Casey Station	-66.28	110.53	60	CSIRO
DEM	Demyanskoe	59.79	70.87	75	JR-STATION
EGB	Egbert	44.23	-79.78	253	ECCC
EIC	Easter Island	-27.13	-109.45	50	NOAA_ESRL
ESP	Estevan Point	49.38	-126.55	39	ECCC
ETL	East Trout Lake	54.35	-104.98	492	ECCC
FSD	Fraserdale	49.88	-81.57	210	ECCC
GLH	Giordan Lighthouse	36.07	14.22	160	Univ. Malta
GMI	Guam	13.43	144.78	2	NOAA_ESRL
GPA	Gunn Point	-12.25	131.05	25	CSIRO
HBA	Halley Bay	-75.57	-26.50	33	NOAA_ESRL
HPB	Hohenpeissenberg	47.80	11.02	985	NOAA_ESRL
HUN	Hegyhatsal	46.95	16.65	248	NOAA_ESRL
ICE	Heimaey	63.40	-20.28	100	NOAA_ESRL
IGR	Igrim	63.19	64.41	9	JR-STATION
IZO	Izaña (Tenerife)	28.30	-16.50	2367	AEMET/NOAA_ESRL
JFJ	Jungfraujoeh	46.55	7.99	3580	EMPA
KEY	Key Biscayne	25.67	-80.20	3	NOAA_ESRL
KRS	Karasevoe	58.25	82.42	76	JR-STATION
KUM	Cape Kumukahi	19.52	-154.82	3	NOAA_ESRL

LAU	Lauder	-45.03	169.67	370	NIWA
LEF	Park Falls	45.92	-90.27	868	NOAA_ESRL
LLB	Lac La Biche (Alberta)	54.95	-112.45	540	ECCC/NOAA_ESRL
LLN	Lulin	23.47	120.87	2867	NOAA_ESRL
LMP	Lampedusa	35.52	12.63	45	ENEA/NOAA_ESRL
MAA	Mawson	-67.62	62.87	32	CSIRO
MEX	High Altitude Global Climate Observation Center, Mexico	19.98	-97.17	4560	NOAA_ESRL
MHD	Mace Head	53.33	-9.90	8	AGAGE/NOAA_ESRL
MID	Sand Island	28.20	-177.37	7.7	NOAA_ESRL
MLO	Mauna Loa	19.54	-155.58	3397	CSIRO/NOAA_ESRL
MNM	Minamitorishima	24.28	153.98	8	JMA
MQA	Macquarie Island	-54.48	158.97	12	CSIRO
NAT	Natal	-6.00	-35.20	0	NOAA_ESRL
NGL	Neuglobsow	53.17	13.03	65	UBA
NMB	Gobabeb	-23.57	15.02	461	NOAA_ESRL
NOY	Noyabrsk	63.43	75.78	108	JR-STATION
NWR	Niwot Ridge (T-van)	40.05	-105.59	3523	NOAA_ESRL
OXK	Ochsenkopf	50.03	11.80	1185	NOAA_ESRL
PAL	Pallas-Sammaltunturi	67.97	24.12	560	NOAA_ESRL
PRS	Plateau Rosa	45.93	7.70	3480	RSE S.p.A.
PSA	Palmer Station	-64.92	-64.00	10	NOAA_ESRL
RGL	Ridge Hill	52.00	-2.54	204	UNIVBRIS
RPB	Ragged Point	13.17	-59.43	45	AGAGE/NOAA_ESRL
RYO	Ryori	39.03	141.82	260	JMA
SDZ	Shangdianzi	40.65	117.12	287	CMA NOAA_ESRL
SEY	Mahe Island	-4.67	55.17	7	NOAA_ESRL
SGP	Southern Great Plains	36.78	-97.50	314	NOAA_ESRL
SHM	Shemya Island	52.72	174.08	40	NOAA_ESRL
SMO	Tutuila (Cape Matatula)	-14.24	-170.57	42	AGAGE/NOAA_ESRL
SNB	Sonnblick	47.05	12.95	3106	UBA
SPO	South Pole	-89.98	-24.80	2810	CSIRO/NOAA_ESRL
SSL	Schauinsland	47.92	7.92	1205	UBA
SUM	Summit	72.58	-38.48	3238	INSTAAR/NOAA_ESRL
SYO	Syowa Station	-69.00	39.58	16	NOAA_ESRL
TAC	Tacolneston Tall Tower	52.52	1.14	56	UNIVBRIS/NOAA_ESRL
TAP	Tae-ahn Peninsula	36.72	126.12	20	NOAA_ESRL
THD	Trinidad Head	41.05	-124.15	120	AGAGE/NOAA_ESRL
TIK	Tiksi	71.59	128.92	8	NOAA_ESRL
TLL	El Tololo	-30.17	-70.80	2220	DMC Empa
TTA	Angus	56.56	-2.99	400+222	UNIVBRIS
USH	Ushuaia	-54.85	-68.31	18	NOAA_ESRL

UTA	Wendover	39.88	-113.72	1320	NOAA_ESRL
UUM	Ulaan Uul	44.45	111.08	914	NOAA_ESRL
WIS	Sede Boker	31.12	34.87	400	NOAA_ESRL
WLD	Westerland	54.93	8.31	12+0	UBA Germany
WLG	Mt. Waliguan	36.28	100.90	3810	CMA NOAA_ESRL
WSA	Sable Island	43.93	-60.02	5	ECCC
YAK	Yakutsk	62.09	129.36	264	JR-STATION
YON	Yonagunijima	24.47	123.02	30	JMA
ZEP	Zeppelifjellet (Ny-Ålesund)	78.90	11.88	475	NOAA_ESRL



5 **Figure S1 Methane bias (ppb) at surface stations for different simulations of TM5, with a priori fluxes (REF), optimized fluxes based on NOAA surface observations (RA) and optimized fluxes based on NOAA surface observations and GOSAT satellite retrievals (RA_S).**



10 **Figure S2 Bias of TM5 modelled methane profiles simulated with a priori fluxes (REF), optimized fluxes based on NOAA surface observations (RA) and optimized fluxes based on NOAA surface observations and GOSAT satellite retrievals (RA_S).**