



Supplement of

Simulating CH₄ and CO₂ over South and East Asia using the zoomed chemistry transport model LMDz-INCA

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1 **Supplementary materials**

2 **Table S1** The mean bias (\pm s.d.) and RMSE of the simulated annual gradient for different
 3 station groups: **(a)** CH₄ (unit: ppb), **(b)** CO₂ (ppm). Results from both ZAs and STs are
 4 presented. Statistics are given for stations outside and within the zoomed region, as well as
 5 for stations of different types within the zoomed region.

6 **(a)**

model version	statistics	outside Z	within Z	marine	mountain	coastal	continental
ST19_ED42	MB	-6.4 \pm 4.8	0.0 \pm 21.0	6.7 \pm 23.5	-3.2 \pm 10.2	-7.8 \pm 22.3	2.3 \pm 35.7
	RMSE	7.9	20.4	22.0	9.6	19.8	29.2
ZA19_ED42	MB	-0.6 \pm 8.0	3.8 \pm 16.5	15.0 \pm 18.0	-1.8 \pm 7.9	8.4 \pm 13.4	-10.4 \pm 19.3
	RMSE	7.7	16.4	22.0	7.2	13.8	18.9
ST39_ED42	MB	-6.8 \pm 4.4	0.1 \pm 22.3	7.1 \pm 24.6	-5.2 \pm 10.7	-7.3 \pm 25.5	4.9 \pm 36.6
	RMSE	8.0	21.6	23.1	10.9	22.1	30.3
ZA39_ED42	MB	-1.2 \pm 7.9	6.4 \pm 17.5	17.6 \pm 17.8	-1.9 \pm 7.8	11.9 \pm 14.5	-4.0 \pm 25.0
	RMSE	7.7	18.1	23.7	7.2	16.8	20.8

7 **(b)**

model version	statistics	outside Z	within Z	marine	mountain	coastal	continental
ST19_ED42	MB	-0.6 \pm 0.5	-0.1 \pm 2.5	-0.3 \pm 1.7	0.5 \pm 4.0	-1.5 \pm 1.8	0.2 \pm 1.9
	RMSE	0.8	2.4	1.5	3.6	2.1	1.7
ZA19_ED42	MB	-0.9 \pm 1.4	0.0 \pm 2.5	-0.3 \pm 2.2	0.2 \pm 3.9	-0.1 \pm 3.0	0.0 \pm 1.8
	RMSE	1.7	2.5	1.9	3.5	2.4	1.6
ST39_ED42	MB	-0.4 \pm 0.7	0.0 \pm 2.5	-0.1 \pm 1.7	0.5 \pm 4.0	-1.2 \pm 1.9	0.3 \pm 2.1
	RMSE	0.8	2.5	1.5	3.6	2.0	1.9
ZA39_ED42	MB	-0.9 \pm 1.4	0.1 \pm 2.6	-0.2 \pm 2.1	0.2 \pm 3.8	0.3 \pm 3.3	0.1 \pm 2.0
	RMSE	1.6	2.5	1.9	3.4	2.7	1.8

9 **Table S2** The observed and simulated mean annual gradient of **(a)** CH₄ and **(b)** CO₂ at
 10 stations mentioned in Section 3.1.1 and Section 3.1.2. The bias reduction rates (in percentage)
 11 of ZAs compared to STs are also given for both 19-layer and 39-layer simulations.

a)

CH₄	OBS (ppb)	ST19 (ppb)	ZA19 (ppb)	Bias reduction	ST39 (ppb)	ZA39 (ppb)	Bias reduction
PON	32.4±12.4	2.5±11.6	31.1±7.7	95.6%	0.4±11.9	34.1±7.8	94.7%
SDZ	90.0±15.4	125.1±18.8	86.8±16.0	91.0%	128.5±19.3	100.4±22.4	73.0%
TAP	64.9±10.7	79.5±8.1	88.6±8.4	n.a.	83.9±7.5	93.3±7.8	n.a.
UUM	38.6±5.6	46.1±9.7	42.8±13.3	44.1%	49.0±11.6	49.1±8.9	n.a.

b)

CO₂	OBS (ppm)	ST19 (ppm)	ZA19 (ppm)	Bias reduction	ST39 (ppm)	ZA39 (ppm)	Bias reduction
PON	2.7±1.6	1.3±0.3	1.8±0.5	35.2%	1.5±0.3	1.9±0.5	37.0%
SDZ	6.8±0.5	8.8±1.3	7.7±1.9	57.9%	9.3±1.5	8.1±2.3	48.1%
TAP	6.9±1.8	7.2±0.8	10.2±0.8	n.a.	7.5±1.0	10.8±1.1	n.a.

12

13 **Table S3** The correlation coefficients between the simulated and observed synoptic
 14 variability of CH₄ (**a**) and CO₂ (**b**) at PON over the period 2006–2013. The synoptic
 15 variability is calculated from residuals from the smoothed fitting curve.

16 **(a) CH₄**

Months	N. of Samples	ST19_ED42	ZA19_ED42	ST39_ED42	ZA39_ED42
Jan.–Mar.	132	0.40***	0.39***	0.42***	0.42***
Apr.–Jun.	81	0.46***	0.43***	0.49***	0.43***
Jul.–Sep.	123	0.48***	0.46***	0.48***	0.45***
Oct.–Dec.	88	0.36***	0.49***	0.39***	0.53***
All	424	0.40***	0.45***	0.42***	0.47***

17 **(b) CO₂**

Months	N. of Samples	ST19_ED42	ZA19_ED42	ST39_ED42	ZA39_ED42
Jan.–Mar.	124	-0.10	-0.24**	-0.08	-0.20*
Apr.–Jun.	69	-0.20	-0.23	-0.21	-0.24*
Jul.–Sep.	105	-0.20*	0.05	-0.22*	0.02
Oct.–Dec.	83	0.05	0.08	0.02	0.06
All	381	-0.11*	-0.11*	-0.11*	-0.11*

19 **Table S4** The statistics between the simulated and observed mean diurnal cycles of CH₄ for
 20 three exemplified stations GSN (**a**), PON (**b**) and BKT (**c**) over specific study periods. For
 21 BKT, results from outputs extracted at a lower model level (Level=2) are presented in (**d**).
 22 For each station, correlation coefficients and ratios of amplitudes are calculated from the
 23 simulated and observed diurnal cycles averaged over all the sampling days in a month with a
 24 complete 24-hour profile.

25 **(a) GSN**

Month	N. of Days	Amplitudes (ppb)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
200801	16	13.2	0.33	0.41	0.39	1.15	0.29	0.42	0.40	1.39
200802	16	13.8	0.54**	1.50	0.72***	0.95	0.49*	1.56	0.75***	0.93
200803	24	17.6	0.48*	0.11	0.51*	0.66	0.50*	0.14	0.45*	0.75
200804	13	27.1	0.64**	0.47	0.80***	0.75	0.69***	0.35	0.78***	0.81
200805	15	28.4	-0.81***	0.24	0.70***	0.52	-0.86***	0.18	0.63**	0.43
200806	19	45.6	0.78***	0.17	0.79***	0.56	0.78***	0.20	0.73***	0.58
200807	12	24.5	-0.05	0.75	0.00	0.43	-0.11	0.85	0.32	0.44
200808	14	58.4	0.83***	0.25	0.67***	0.72	0.86***	0.28	0.76***	0.74
200809	1	63.5	0.64**	0.57	0.08	0.35	0.59**	0.73	0.40	0.46
200810	12	28.2	0.48*	0.37	-0.29	0.85	0.47*	0.45	-0.35	0.80
200811	12	19.3	-0.30	0.69	0.28	0.86	-0.52**	1.08	-0.09	0.74
200812	17	17.0	0.09	0.53	0.35	0.99	0.43*	0.65	0.48*	1.03

26 * p < 0.05; ** p < 0.01; *** p < 0.001

27 **(b) PON**

Month	N. of Days	Amplitudes (ppb)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
201108	4	187.4	0.56**	0.04	0.79***	0.15	0.71***	0.06	0.76***	0.15
201109	14	163.1	0.96***	0.08	0.91***	0.40	0.93***	0.09	0.84***	0.37
201210	25	133.5	0.87***	0.13	0.96***	0.50	0.83***	0.18	0.95***	0.68
201211	26	229.6	0.95***	0.12	0.97***	0.33	0.95***	0.16	0.98***	0.40
201212	28	206.6	0.88***	0.06	0.98***	0.31	0.96***	0.07	1.00***	0.34
201301	27	309.0	0.89***	0.05	0.98***	0.22	0.94***	0.05	0.98***	0.26
201302	20	238.9	0.79***	0.08	0.97***	0.27	0.85***	0.08	0.97***	0.32
201303	29	146.9	0.85***	0.10	0.96***	0.48	0.91***	0.10	0.95***	0.60
201304	25	121.6	0.76***	0.09	0.94***	0.40	0.83***	0.10	0.90***	0.42
201305	15	78.9	0.93***	0.15	0.92***	0.41	0.90***	0.13	0.93***	0.36

28 * p < 0.05; ** p < 0.01; *** p < 0.001

29 **(c) BKT, at the station level**

Month	N. of Days	Amplitudes (ppb)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
201301	27	67.4	-0.09	0.19	0.80***	0.24	-0.49*	0.12	0.89***	0.34
201302	17	32.5	-0.12	0.33	0.08	0.28	-0.18	0.28	0.47*	0.26
201303	23	83.6	-0.22	0.14	0.70***	0.21	-0.61**	0.10	0.81***	0.26
201304	20	47.9	-0.19	0.21	-0.05	0.21	-0.28	0.19	-0.02	0.21

201305	17	33.7	-0.60**	0.31	-0.53**	0.26	-0.62**	0.31	-0.42*	0.31
201306	18	30.6	0.18	0.92	0.17	0.51	0.03	0.84	0.43*	0.51
201307	19	31.1	-0.24	0.46	-0.21	0.27	-0.37	0.47	-0.13	0.32
201308	21	50.2	-0.83***	0.14	-0.42*	0.13	-0.84***	0.13	-0.32	0.12
201309	8	22.6	0.51*	0.58	0.40	0.46	0.61**	0.57	0.34	0.52
201310	16	90.8	-0.27	0.09	0.20	0.07	-0.74***	0.07	0.27	0.09
201311	21	44.7	-0.11	0.24	-0.07	0.24	-0.12	0.19	-0.24	0.22
201312	22	58.3	0.50*	0.17	0.87***	0.34	0.57**	0.11	0.80***	0.43

30 * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$

31 **(d) BKT, at a lower model level (Level=2)**

Month	N. of Days	Amplitudes (ppb)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
201301	27	67.4	0.78***	0.51	0.94***	0.55	0.97***	0.87	0.98***	0.83
201302	17	32.5	0.78***	0.99	0.87***	0.71	0.83***	1.64	0.74***	1.21
201303	23	83.6	0.86***	0.38	0.91***	0.50	0.89***	0.67	0.84***	0.67
201304	20	47.9	0.72***	0.32	0.60**	0.22	0.72***	0.55	0.72***	0.38
201305	17	33.7	0.37	0.42	0.01	0.30	0.85***	0.64	0.88***	0.43
201306	18	30.6	0.56**	1.64	0.57**	0.84	0.55**	2.18	0.53**	1.17
201307	19	31.1	0.72***	0.73	0.46*	0.30	0.82***	1.13	0.70***	0.56
201308	21	50.2	0.65**	0.24	0.53**	0.16	0.83***	0.46	0.77***	0.38
201309	8	22.6	0.10	0.80	0.09	0.63	-0.22	1.24	-0.13	1.26
201310	16	90.8	0.81***	0.22	0.82***	0.13	0.95***	0.35	0.91***	0.27
201311	21	44.7	0.44*	0.43	0.38	0.29	0.70***	0.61	0.60**	0.29
201312	22	58.3	0.69***	0.55	0.81***	0.67	0.85***	1.07	0.87***	1.02

32 * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$

33

34 **Table S5** The statistics between the simulated and observed mean diurnal cycles of CO₂ for
 35 three exemplified stations GSN (**a**), PON (**b**) and BKT (**c**) over specific study periods. For
 36 BKT, results from outputs extracted at the surface model level (Level=1) are presented in (**d**).
 37 For each station, correlation coefficients and ratios of amplitudes are calculated from the
 38 simulated and observed diurnal cycles averaged over all the sampling days in a month with a
 39 complete 24-hour profile.

40 **(a) GSN**

Month	N. of Days	Amplitudes (ppm)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
200801	10	2.3	0.01	0.17	0.00	0.30	0.12	0.26	0.04	0.45
200802	13	4.0	0.59**	0.45	0.78***	0.27	0.56**	0.47	0.81***	0.26
200803	20	2.2	-0.52**	0.22	0.70***	0.49	-0.64**	0.19	0.53**	0.42
200804	17	3.2	0.79***	0.27	0.74***	0.32	0.82***	0.27	0.72***	0.37
200805	13	2.8	0.47*	0.12	-0.36	0.14	0.33	0.03	-0.43*	0.12
200806	12	3.5	-0.30	0.08	0.10	0.07	-0.42*	0.11	-0.26	0.08
200807	12	4.4	0.40	0.34	0.50*	0.18	0.54**	0.33	0.70***	0.21
200808	15	5.3	0.25	0.11	0.59**	0.40	0.30	0.14	0.66***	0.36
200809	9	5.8	-0.73***	0.16	0.00	0.30	-0.82***	0.15	-0.26	0.33
200810	13	4.0	-0.36	0.22	-0.74***	0.51	-0.34	0.27	-0.74***	0.52
200811	8	1.8	0.90***	0.55	0.90***	1.09	0.83***	0.23	0.85***	0.84
200812	20	1.7	-0.48**	0.35	0.09	0.80	-0.50**	0.35	0.25	0.65

41 * p < 0.05; ** p < 0.01; *** p < 0.001

42 **(b) PON**

Month	N. of Days	Amplitudes (ppm)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
201108	4	42.4	0.36	0.01	0.43*	0.03	0.41*	0.02	0.28	0.03
201109	14	30.4	0.90***	0.02	0.83***	0.10	0.86***	0.02	0.72***	0.11
201210	25	23.2	0.83***	0.04	0.94***	0.12	0.78***	0.06	0.95***	0.16
201211	26	35.7	0.86***	0.03	0.92***	0.05	0.85***	0.04	0.91***	0.06
201212	28	30.1	0.60**	0.01	0.96***	0.04	0.77***	0.01	0.95***	0.05
201301	27	38.2	0.81***	0.01	0.07	0.02	0.84***	0.01	0.02	0.02
201302	20	36.5	0.84***	0.02	0.34	0.02	0.81***	0.02	0.54**	0.02
201303	29	29.9	0.88***	0.05	0.95***	0.13	0.91***	0.04	0.97***	0.17
201304	25	19.4	0.77***	0.10	0.94***	0.35	0.80***	0.11	0.92***	0.37
201305	15	13.4	0.89***	0.11	0.56**	0.28	0.87***	0.11	0.51*	0.29

43 * p < 0.05; ** p < 0.01; *** p < 0.001

44 **(c) BKT, at the station level**

Month	N. of Days	Amplitudes (ppm)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
201301	27	23.1	0.04	0.07	0.52**	0.08	-0.07	0.05	0.52**	0.11
201302	17	20.3	0.09	0.07	0.54**	0.06	-0.25	0.04	0.66***	0.09
201303	23	25.3	-0.08	0.06	0.46*	0.08	-0.34	0.04	0.60**	0.10
201304	20	22.3	-0.17	0.06	0.26	0.06	-0.42*	0.04	0.48*	0.07

201305	17	19.5	-0.35	0.05	0.32	0.05	-0.64**	0.05	0.50*	0.08
201306	18	21.3	-0.53**	0.07	0.16	0.06	-0.83***	0.06	0.40	0.08
201307	20	19.7	-0.09	0.07	0.60**	0.06	-0.38	0.05	0.68***	0.07
201308	19	22.3	0.62**	0.06	0.96***	0.05	0.49*	0.05	0.95***	0.07
201309	8	14.2	0.82***	0.06	0.84***	0.11	0.71***	0.05	0.84***	0.14
201310	16	23.8	-0.09	0.06	0.40	0.06	-0.37	0.04	0.61**	0.07
201311	21	42.4	-0.29	0.03	0.11	0.03	-0.58	0.02	0.42*	0.04
201312	22	27.9	0.00	0.05	0.49*	0.06	-0.22	0.03	0.37	0.08

45 * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$

46 **(d) BKT, at the surface model level (Level=1)**

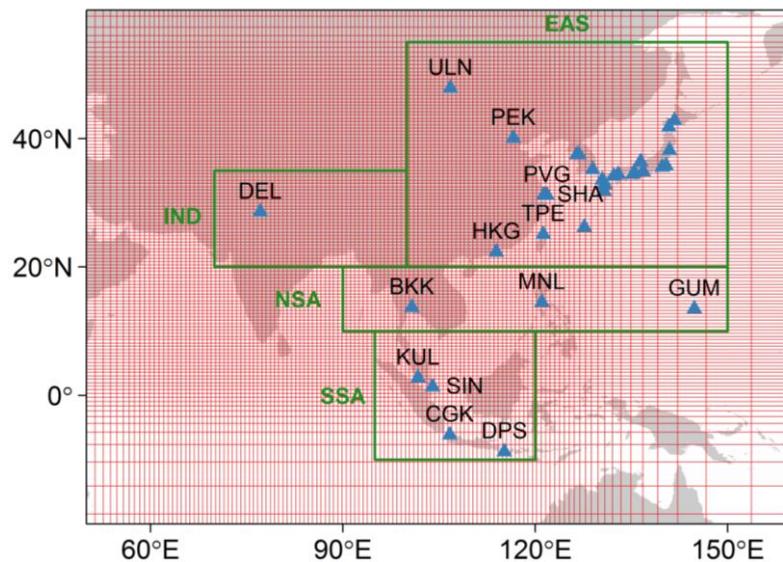
Month	N. of Days	Amplitudes (ppm)	ST19_ED42		ZA19_ED42		ST39_ED42		ZA39_ED42	
			R	A _m /A _o						
201301	27	23.1	0.95***	0.42	0.97***	0.48	0.96***	0.52	0.97***	0.57
201302	17	20.3	0.96***	0.44	0.97***	0.50	0.97***	0.57	0.97***	0.63
201303	23	25.3	0.97***	0.39	0.97***	0.46	0.96***	0.50	0.96***	0.54
201304	20	22.3	0.93***	0.40	0.96***	0.42	0.95***	0.51	0.96***	0.52
201305	17	19.5	0.93***	0.47	0.96***	0.43	0.94***	0.61	0.95***	0.59
201306	18	21.3	0.92***	0.48	0.94***	0.40	0.93***	0.55	0.94***	0.51
201307	20	19.7	0.94***	0.39	0.97***	0.37	0.96***	0.50	0.96***	0.49
201308	19	22.3	0.92***	0.17	0.95***	0.25	0.90***	0.25	0.94***	0.33
201309	8	14.2	0.93***	0.45	0.89***	0.33	0.95***	0.52	0.88***	0.41
201310	16	23.8	0.98***	0.42	0.97***	0.48	0.97***	0.55	0.95***	0.61
201311	21	42.4	0.96***	0.21	0.97***	0.24	0.96***	0.28	0.97***	0.28
201312	22	27.9	0.86***	0.36	0.90***	0.42	0.91***	0.45	0.92***	0.52

47 * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$

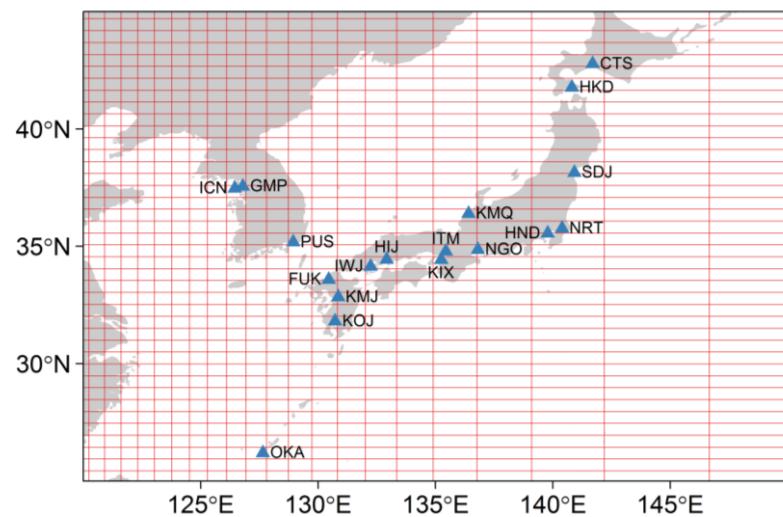
48

49 **Figure S1 (a)** Map of locations of airports in South and East Asia from the Comprehensive
 50 Observation Network for TRace gases by AIrLiner (CONTRAIL) project (Machida et al.,
 51 2008). **(b)** Close-up map for airports in Japan and Republic of Korea. The whole region is
 52 divided into four subregions, namely East Asia (EAS), the Indian sub-continent (IND),
 53 Northern Southeast Asia (NSA) and Southern Southeast Asia (SSA), and all the airports and
 54 vertical profiles are grouped into the four subregions accordingly. The zoomed grid of the
 55 LMDz-INCA model is also plotted as background.

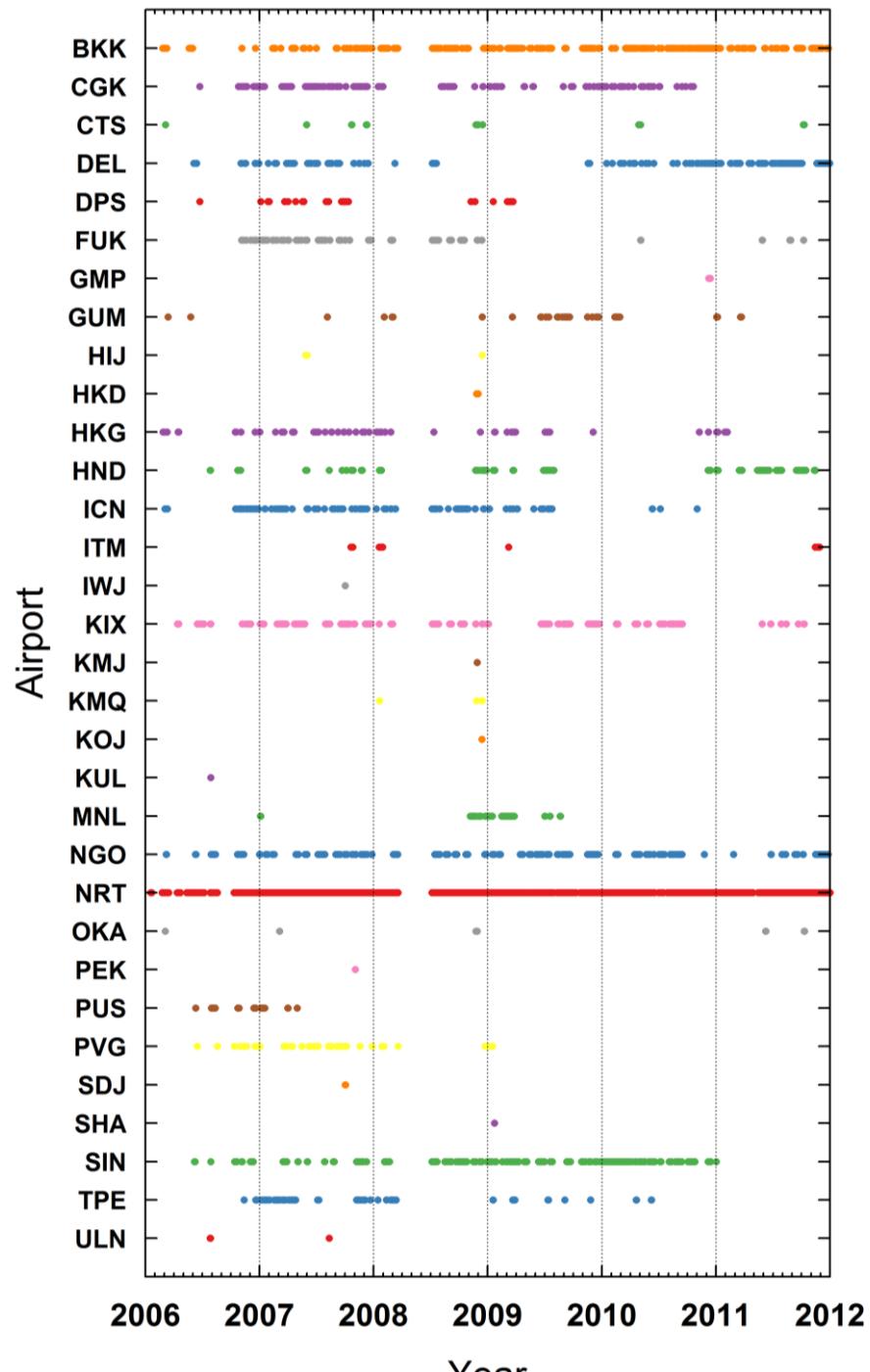
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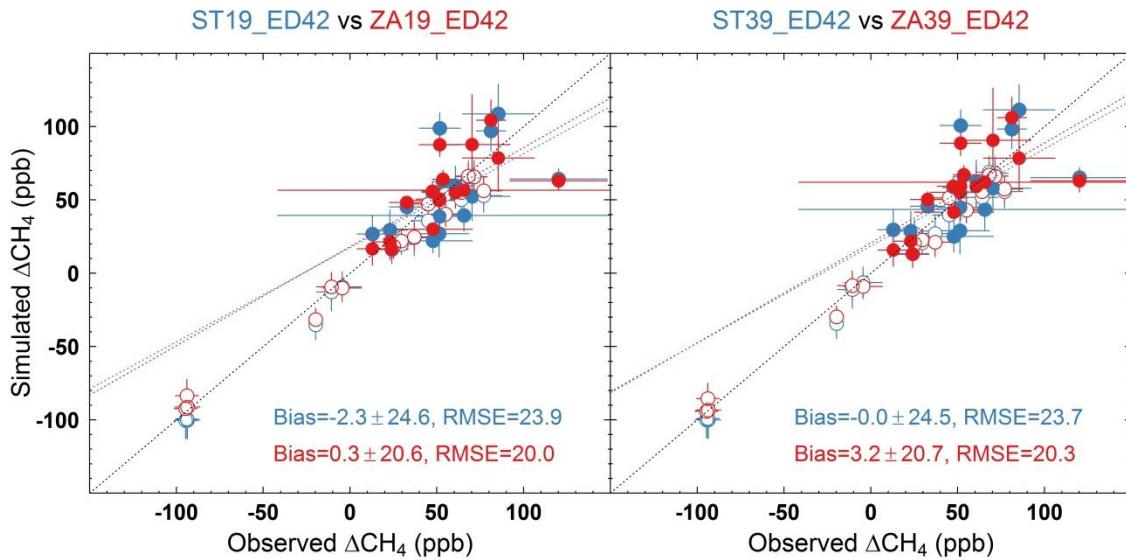


58 **Figure S2** Sampling dates of CO₂ measurements for airports in Figure S1. For each airport,
59 only sampling dates with vertical profiles available (i.e. measurements during ascending or
60 descending flights) are plotted.



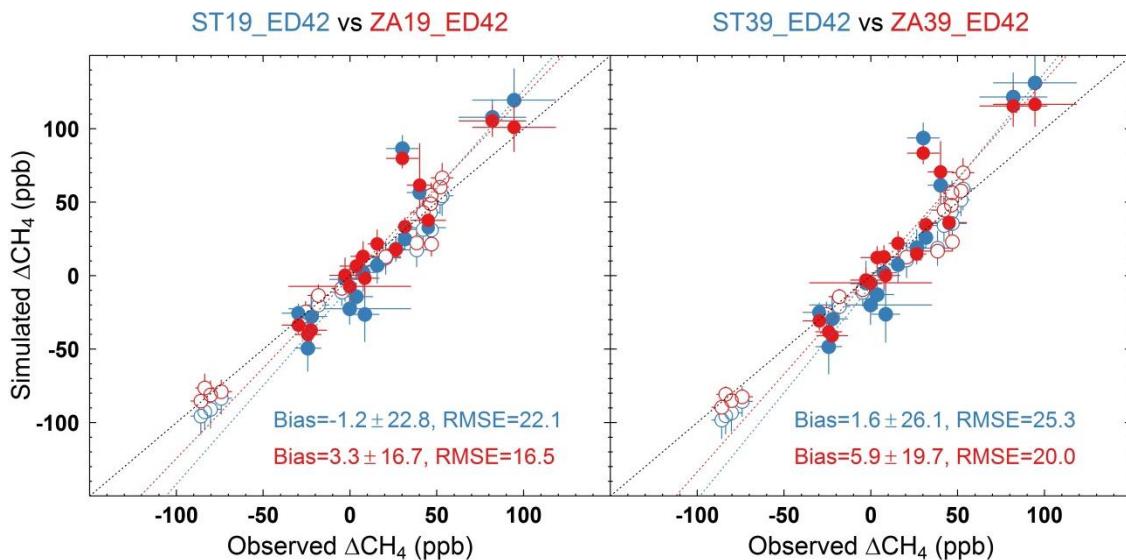
62 **Figure S3** Scatterplots of the simulated and observed CH₄ mean annual gradients between
 63 HLE and other stations for January–March (**a**), April–June (**b**), July–September (**c**), and
 64 October–December (**d**). In each panel, the simulated CH₄ gradients are based on simulations
 65 from the standard (blue circles) and zoom (red circles) versions, respectively. The black
 66 dotted line indicates the identity line, whereas the blue and red dotted lines indicate the
 67 corresponding linear fitted lines. The closed and open circles represent stations inside and
 68 outside the zoomed region.

69 **(a)** January–March



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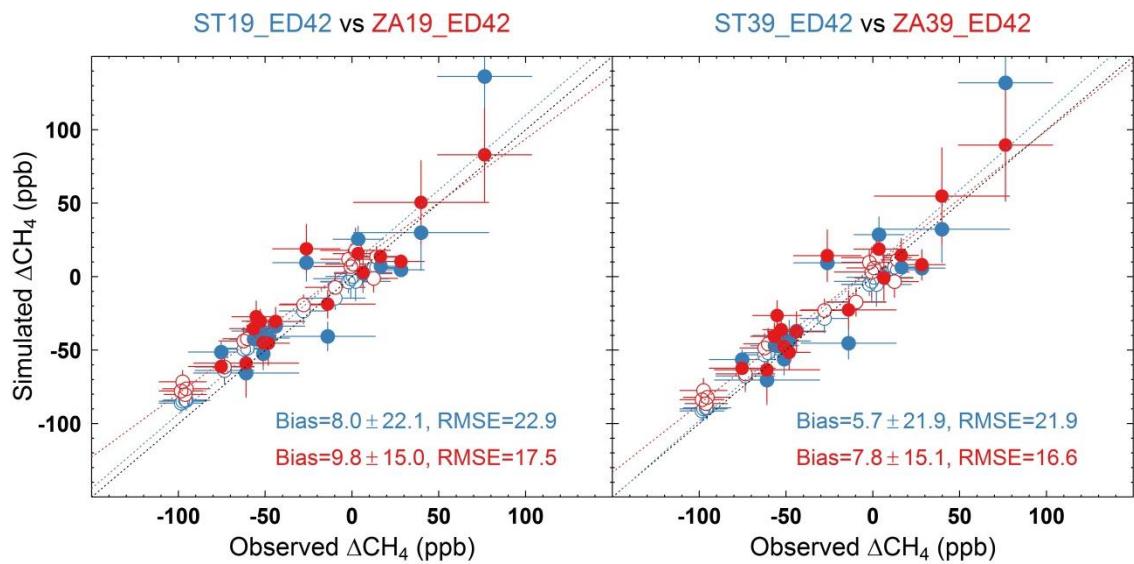
71 **(b)** April–June



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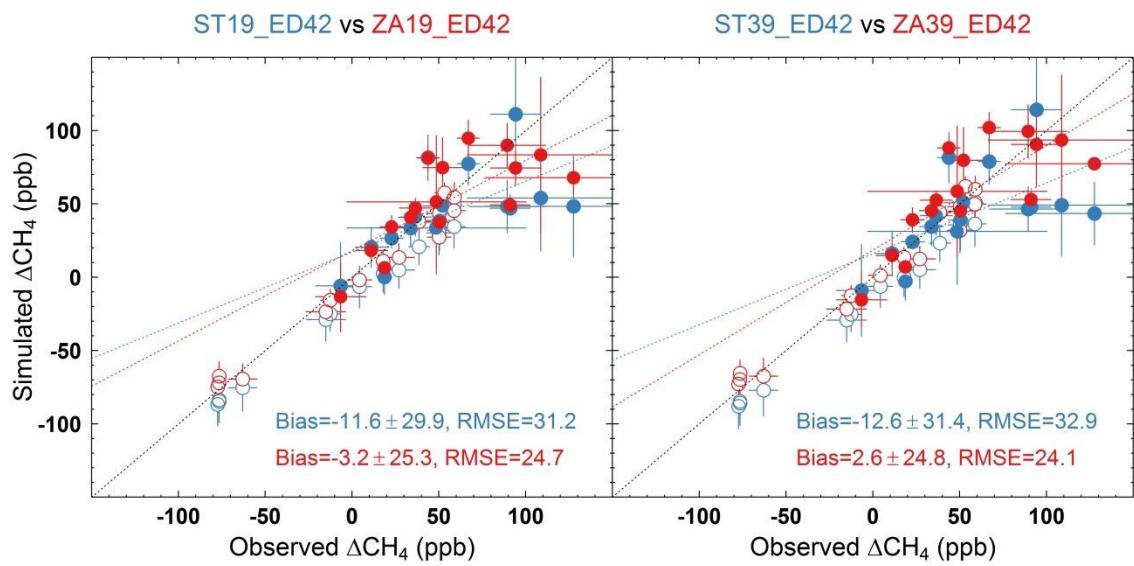
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74 (c) July–September



75

76 (d) October–December



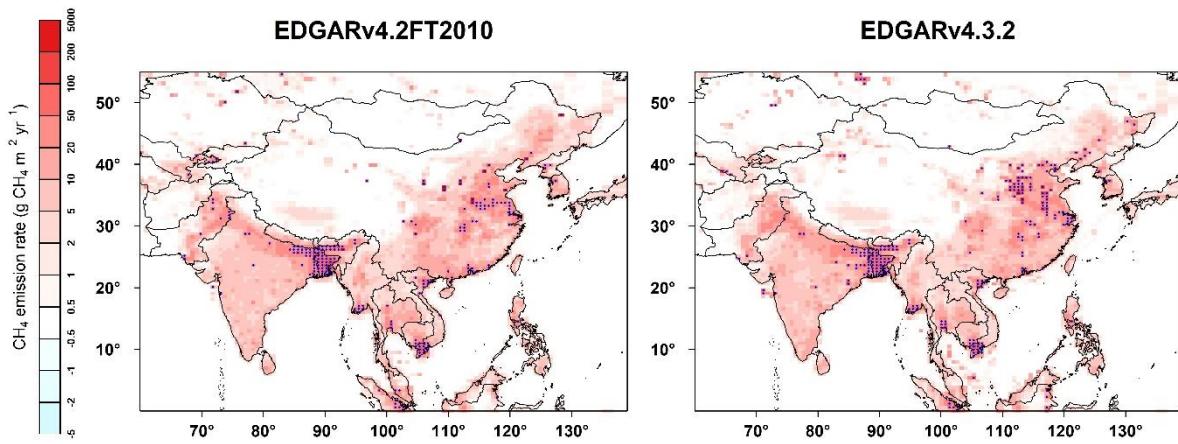
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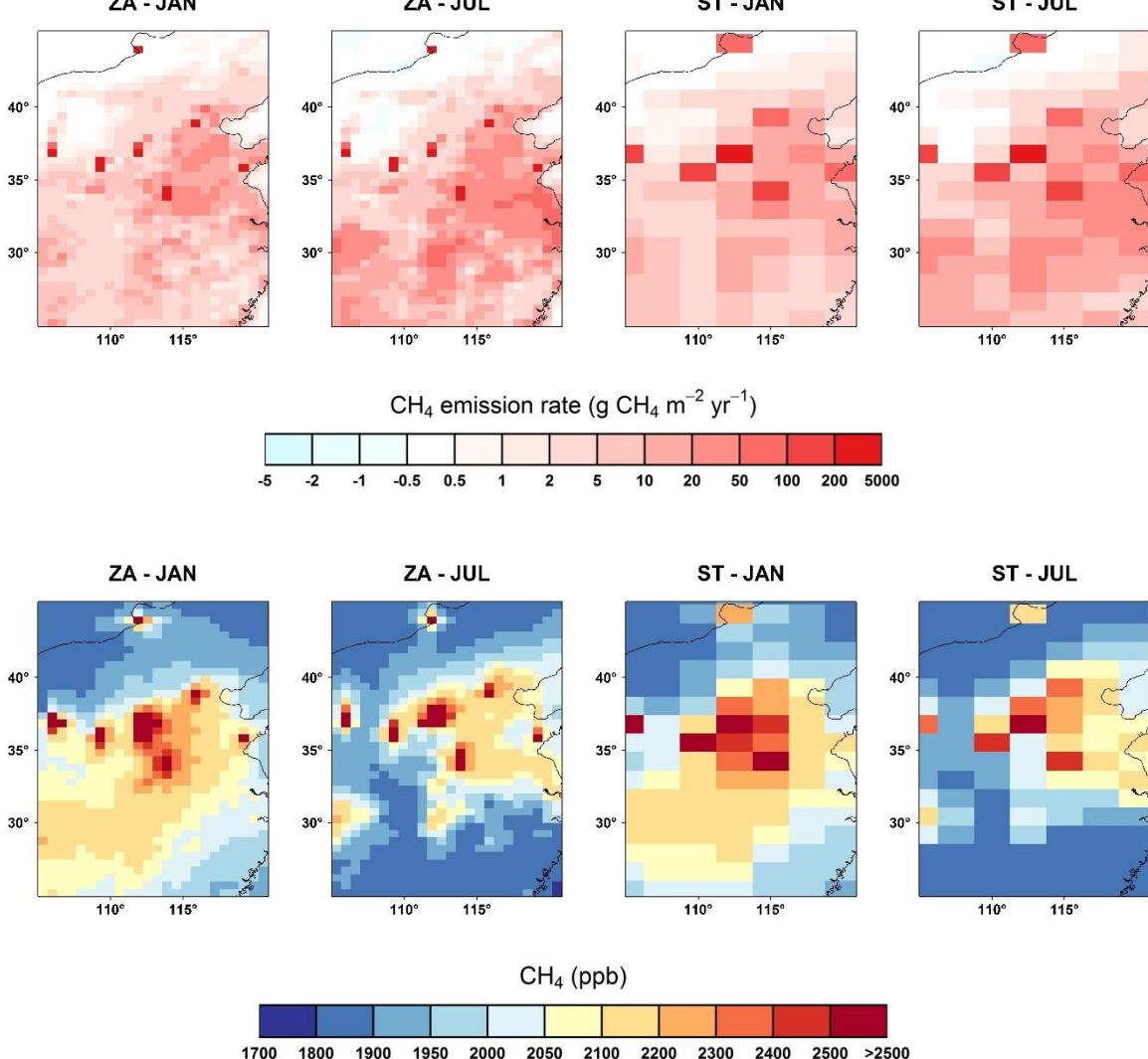
79 **Figure S4** CH₄ surface flux maps for South and East Asia (SEA), based on two different
80 inventories of anthropogenic emissions for the year 2010 from EDGARv4.2FT2010 and
81 EDGARv4.3.2 (<http://edgar.jrc.ec.europa.eu>). CH₄ hotspots, defined as the grids with
82 emission rates stronger than 1×10^{-9} kg CH₄ m⁻² s⁻¹ (≈ 0.8 Tg CH₄ yr⁻¹), are indicated by blue
83 dots. Both maps are generated in ZA grid meshes and with the same biogenic CH₄ fluxes as
84 given in Table 1.

85

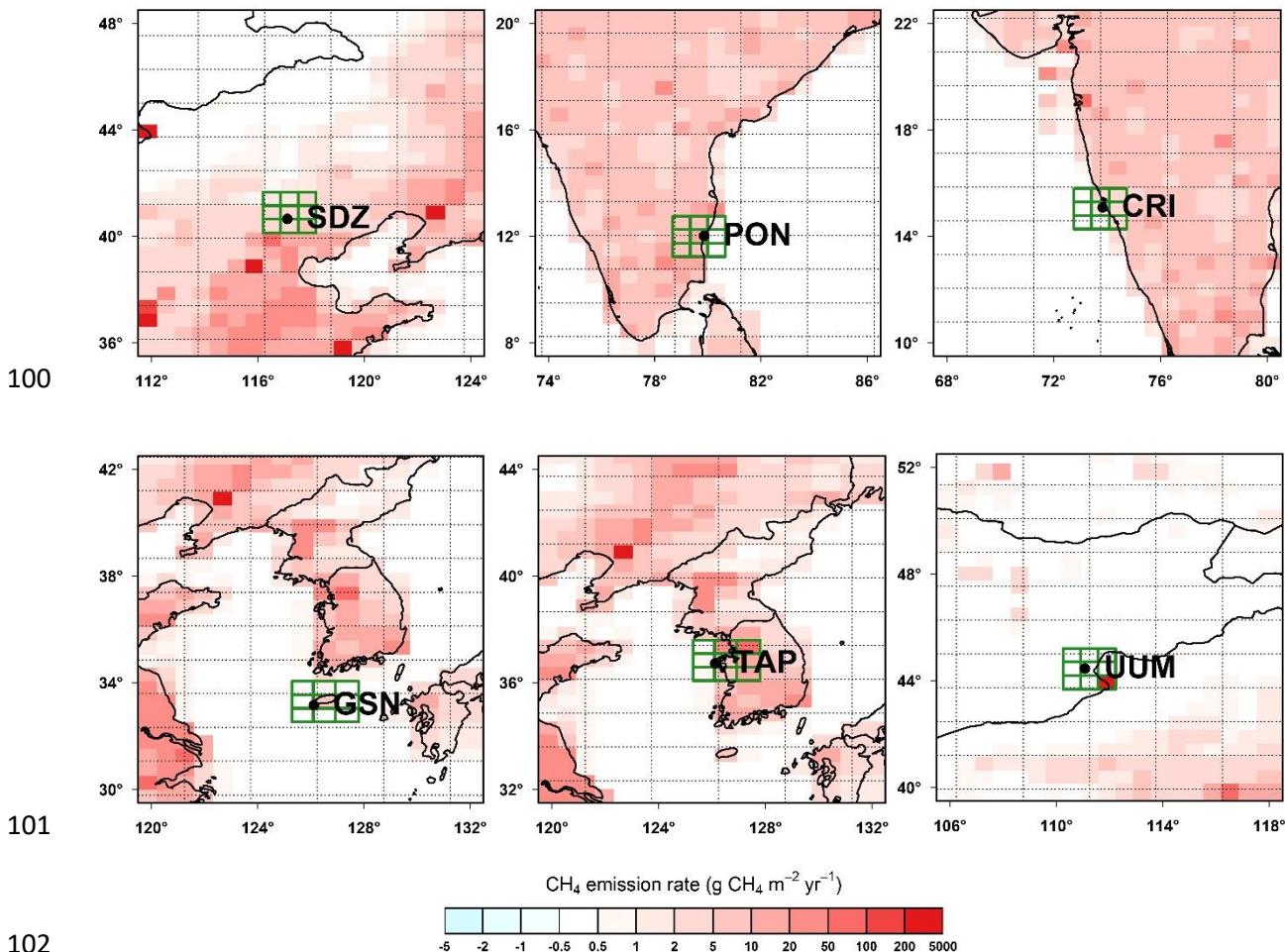
86



87 **Figure S5** Maps of CH₄ surface fluxes (upper panels) and CH₄ concentration fields at the
88 first model level (lower panels) for the year 2010. Results from both ZA and ST with 19
89 model layers
90 are presented for comparison.

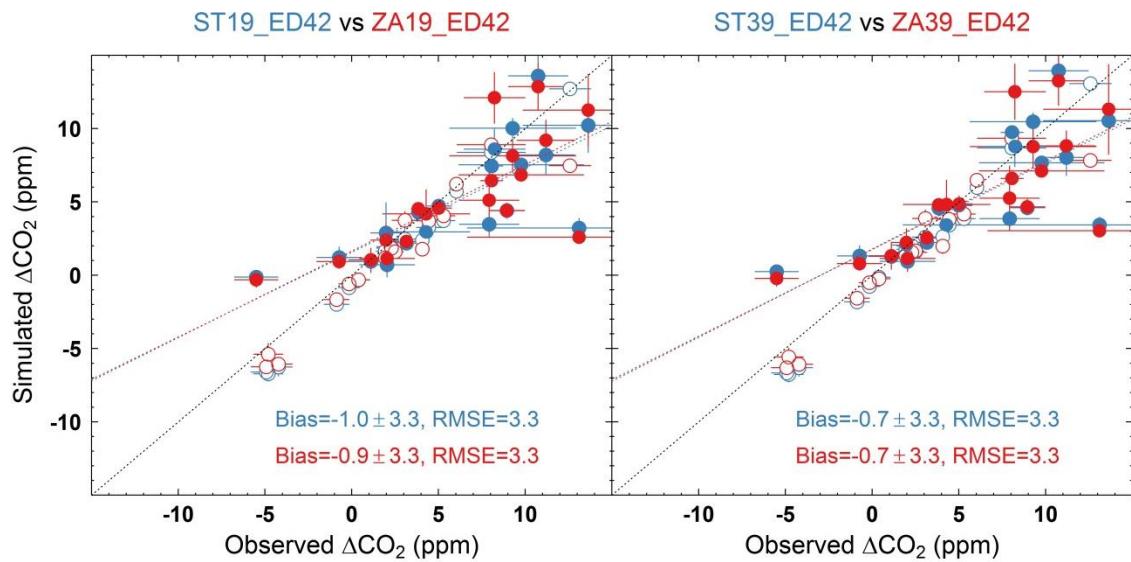


95 **Figure S6** The spatial distributions of mean annual CH₄ fluxes around the stations SDZ, PON,
 96 CRI, GSN, TAP and UUM for the year 2010 mapped with the ZAs model grids. The black
 97 meshes indicate the STs model grids. The black dot denotes the location of the station,
 98 whereas the 3×3 meshes colored in green indicate the grid where the station is located (the
 99 ‘center grid’) and its 8 neighbors.



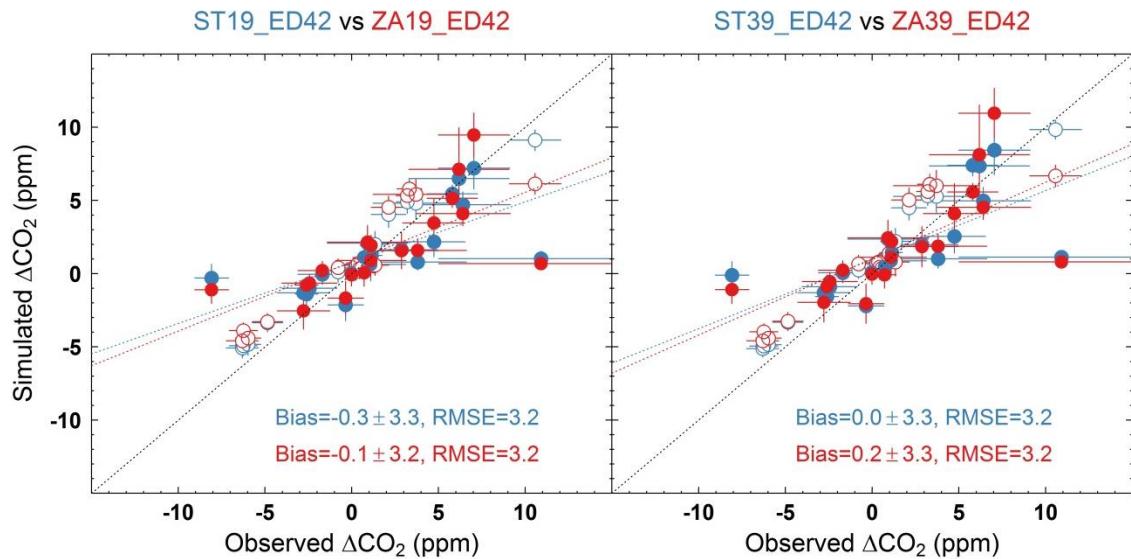
104 **Figure S7** Scatterplots of simulated and observed CO₂ mean annual gradients between HLE
 105 and other stations for January–March (**a**), April–June (**b**), July–September (**c**), and October–
 106 December (**d**). In each panel, the simulated CO₂ gradients are based on simulations from the
 107 standard (blue circles) and zoom (red circles) versions, respectively. The black dotted line
 108 indicates the identity line, whereas the blue and red dotted lines indicate the corresponding
 109 linear fitted lines. The closed and open circles represent stations inside and outside the
 110 zoomed region.

111 **(a)** January–March



112

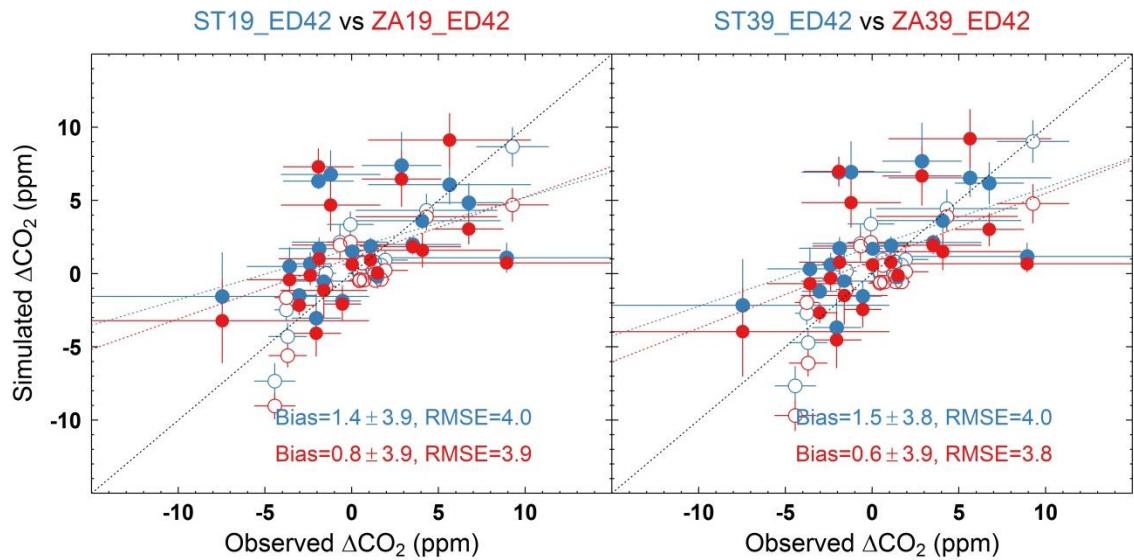
113 **(b)** April–June



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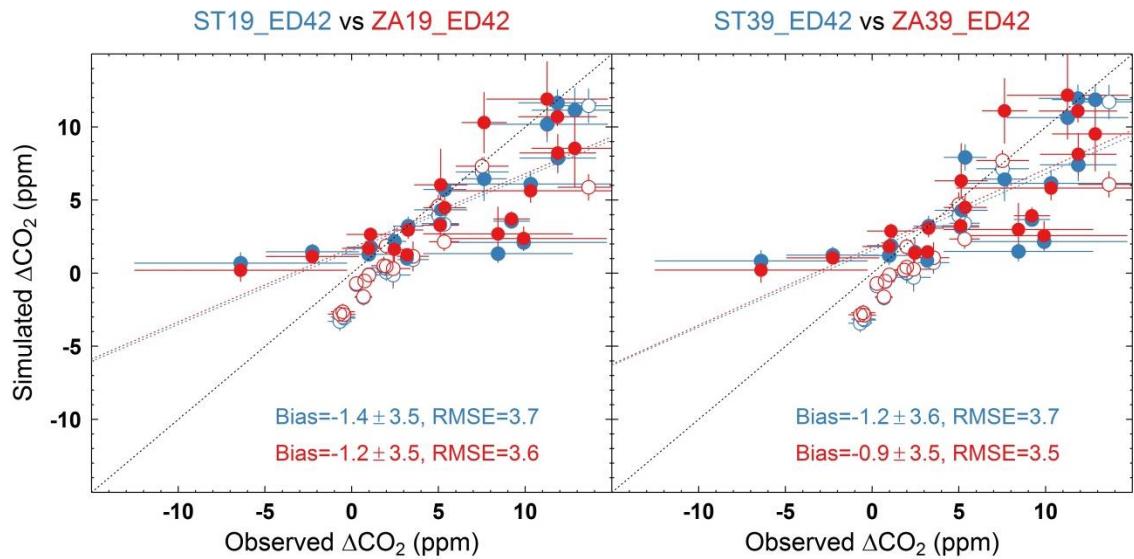
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116 (c) July–September



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118 (d) October–December

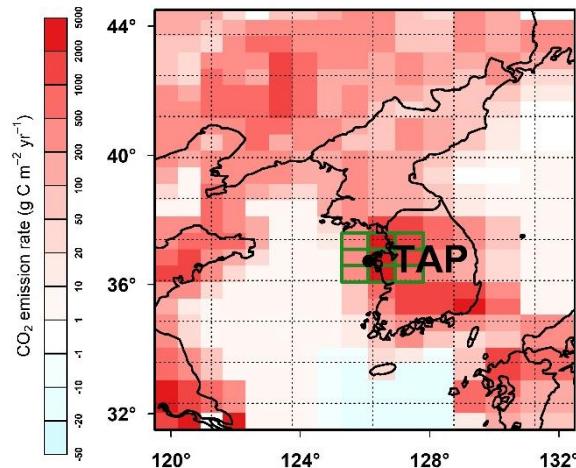


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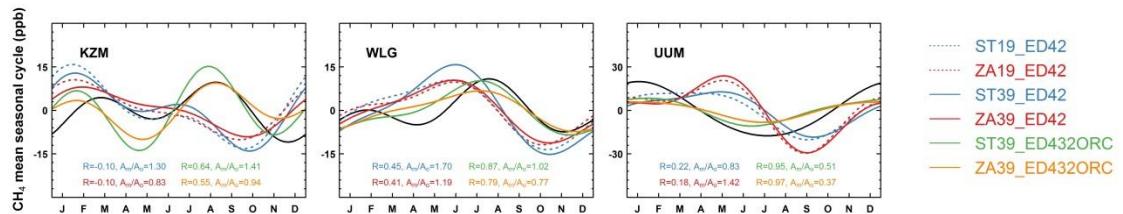
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122 **Figure S8** The spatial distributions of mean annual CO₂ fluxes around the station TAP for the
123 year 2010 mapped with the ZAs model grids. The black meshes indicate the STs model grids.
124 The black dot denotes the location of the station, whereas the 3×3 meshes colored in green
125 indicate the grid where the station is located (the ‘center grid’) and its 8 neighbors.



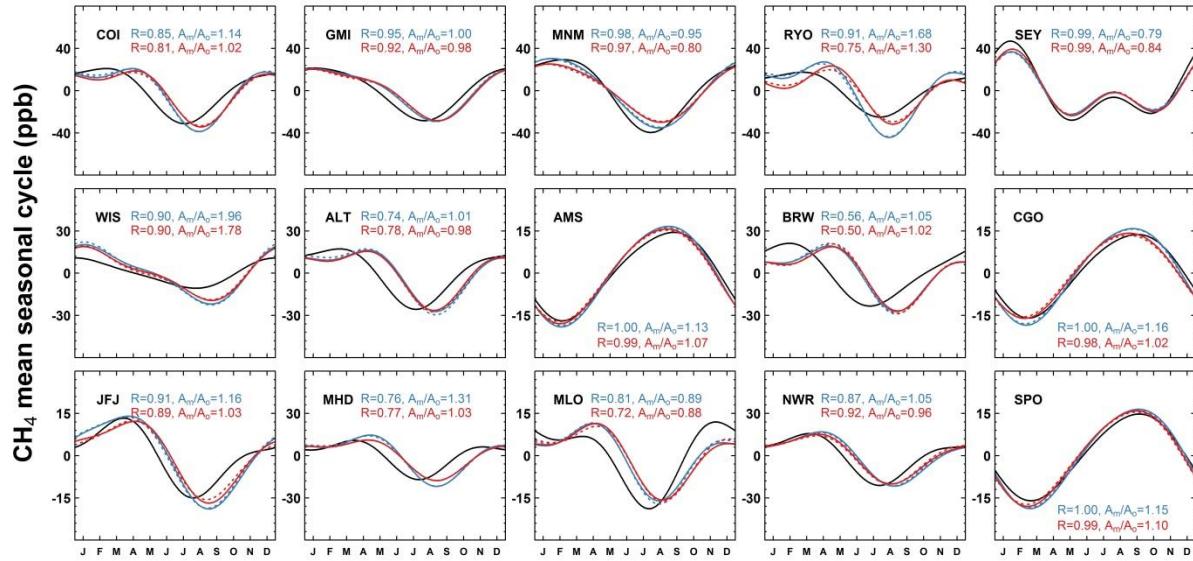
126

127 **Figure S9** The observed and simulated mean seasonal cycles of CH₄ for KZM, WLG and
 128 UUM. In each panel, the colors of lines are defined as Figure 3. In addition, we also show the
 129 mean seasonal cycles from sensitivity test simulations prescribed with wetland emissions
 130 from ORCHIDEE outputs (green and orange lines for the standard and zoom versions,
 131 respectively). The text shows statistics between the simulated and observed seasonal cycles
 132 for 39-layer models.



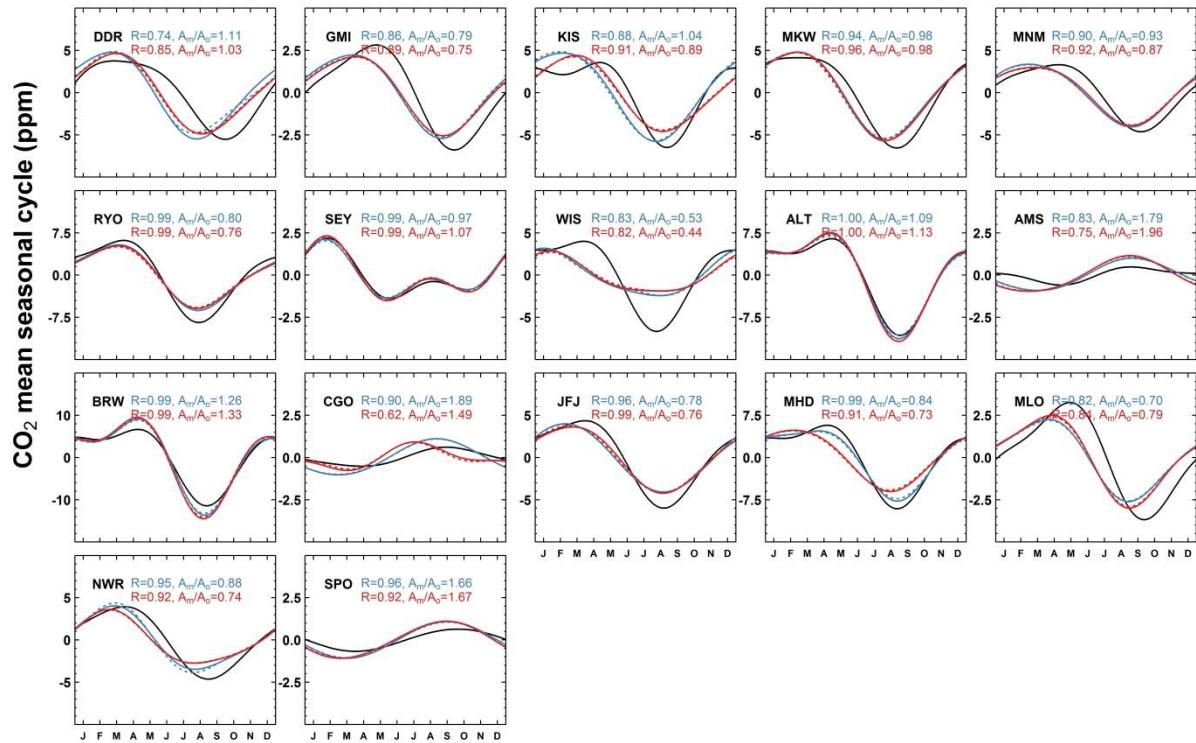
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134 **Figure S10** The observed and simulated mean seasonal cycles of CH₄ for stations outside the
 135 zoomed region. In each panel, the simulated mean seasonal cycles are based on simulations
 136 from the standard (blue lines) and zoom (red lines) versions, respectively. The text shows
 137 statistics between the simulated and observed seasonal cycles for 39-layer models.



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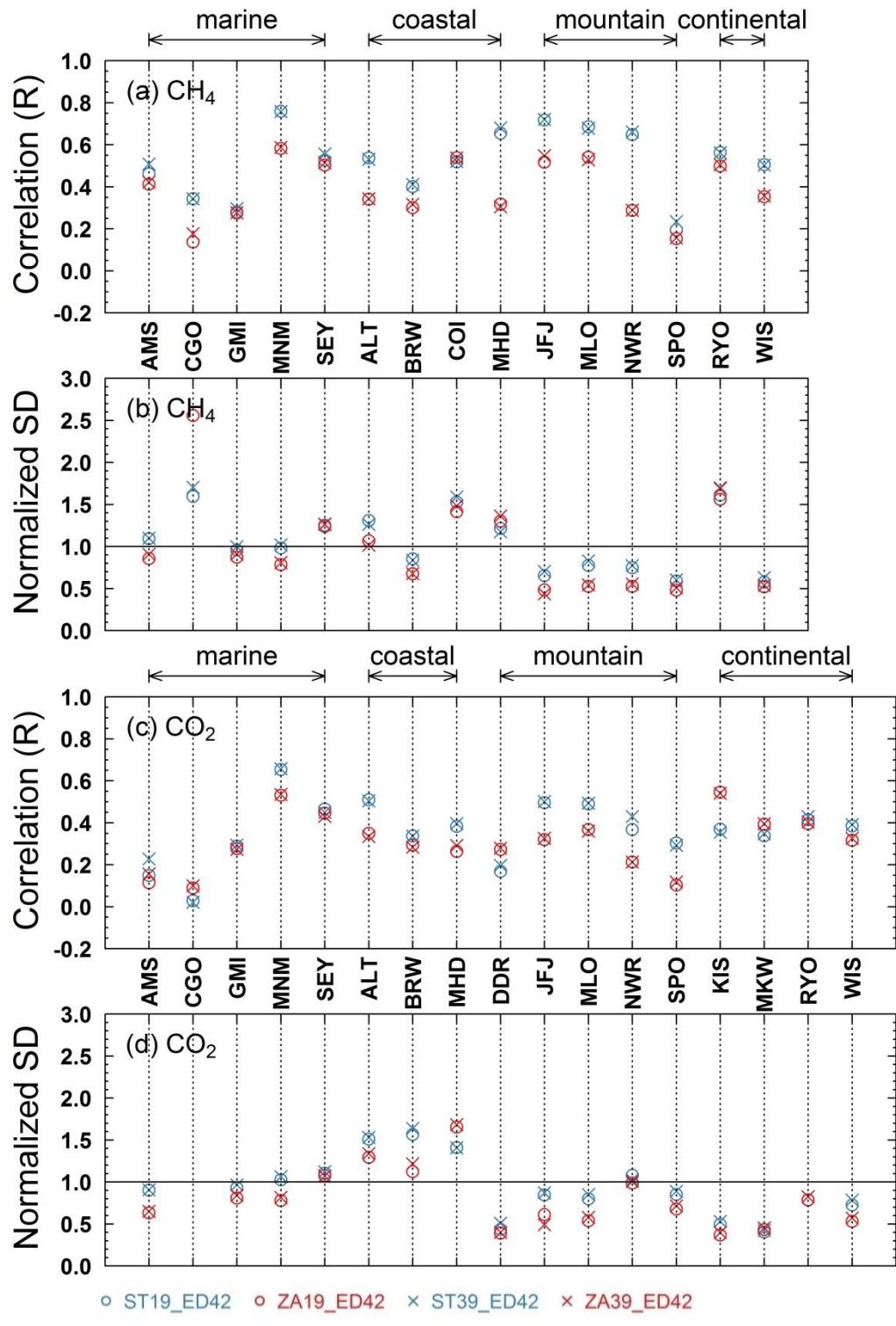
140 **Figure S11** The observed and simulated mean seasonal cycles of CO₂ for stations outside the
 141 zoomed region. In each panel, the simulated mean seasonal cycles are based on simulations
 142 from the standard (blue lines) and zoom (red lines) versions, respectively. The text shows
 143 statistics between the simulated and observed seasonal cycles for 39-layer models.



144

145

146 **Figure S12** The correlations and normalized standard deviations between the simulated and
 147 observed synoptic variability for CH₄ (**a,b**) and CO₂ (**c,d**) at stations outside the zoomed
 148 region. For each station, the synoptic variability is calculated from residuals from the
 149 smoothed fitting curve.



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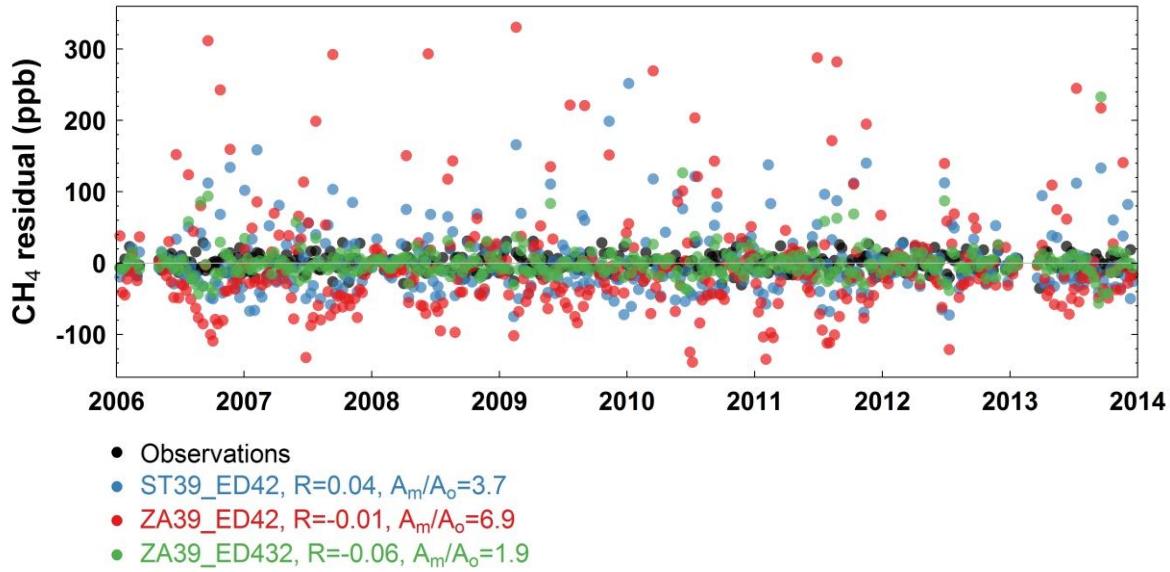
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154 **Figure S13** Time series of observed and simulated CH₄ synoptic variabilities at UUM over
155 the period 2006–2013. The synoptic variability is calculated from residuals from the
156 smoothed fitting curve.

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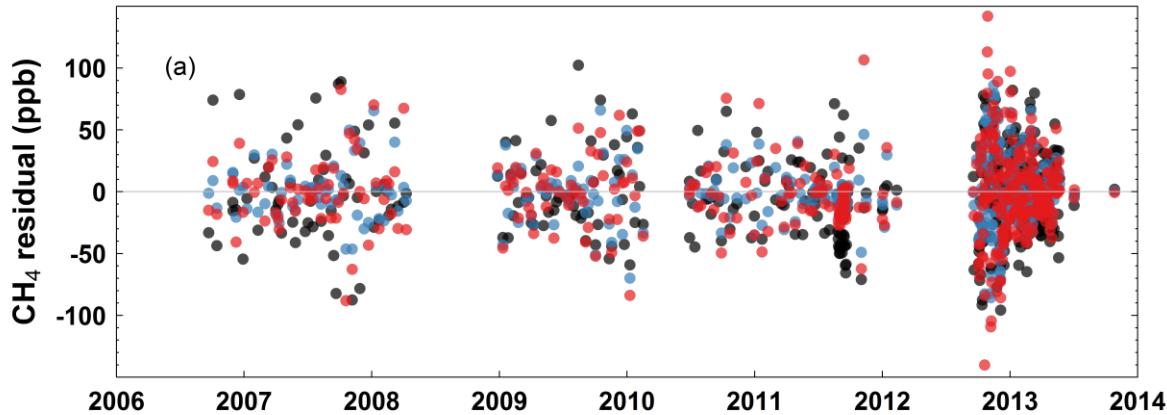
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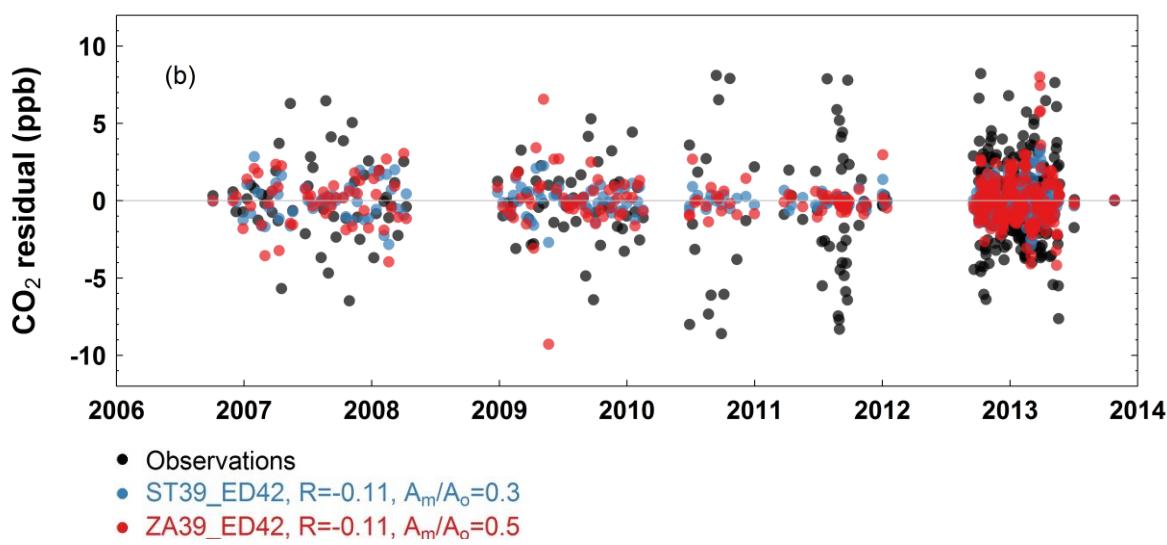


160 **Figure S14** Time series of observed and simulated CH₄ (**a**) and CO₂ (**b**) synoptic variabilities
161 at PON over the period 2006–2013. For each trace gas, the synoptic variability is calculated
162 from residuals from the smoothed fitting curve.

163



164



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168 **Reference**

169 Machida, T., Matsueda, H., Sawa, Y., Nakagawa, Y., Hirotani, K., Kondo, N., Goto, K.,
170 Nakazawa, T., Ishikawa, K. and Ogawa, T.: Worldwide Measurements of Atmospheric CO₂
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172 1744–1754, doi:10.1175/2008JTECHA1082.1, 2008.