**Changing characteristics of atmospheric CH₄ on the Tibetan Plateau,
records from 1994 to 2017 at Mount Waliguan station**

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18 **Supplement**

19 **Table S1.** The seasonal peak to trough amplitude over different observing periods at WLG station.

	1994-1997	1998-2002	2003-2007	2008-2012	2013-2017	1994-2017
Spring	6.5 ± 3.0	4.7 ± 1.8	5.6 ± 2.6	6.2 ± 2.4	6.8 ± 3.4	5.7 ± 2.4
Summer	5.6 ± 3.5	4.4 ± 2.7	4.7 ± 2.3	5.7 ± 2.7	6.1 ± 3.9	4.3 ± 2.6
Autumn	4.7 ± 3.3	5.7 ± 3.3	4.2 ± 2.4	6.5 ± 2.9	5.8 ± 3.6	4.5 ± 2.4
Winter	12.5 ± 3.3	9.9 ± 4.0	5.9 ± 2.7	6.9 ± 3.2	8.3 ± 3.5	6.2 ± 2.4

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21 **Table S2.** Seasonal averages of atmospheric CH₄ in the City Regions (CR), the Tibet Plateau (TP)
 22 and the total regional records (Total) during 1994-2016 at WLG station. Different letters represent
 23 significant differences between regions and seasons in different observing periods.

		1994-1997	1998-2002	2003-2007	2008-2012	2013-2016	1994-2016
Spring	CR	1807.1 ± 0.2d	1812.4 ± 0.3i	1830.4 ± 0.2h	1859.6 ± 0.2h	1881.9 ± 0.3g	1839.1 ± 0.3g
	TP	1804.5 ± 0.2e	1821.2 ± 0.2h	1829.5 ± 0.3i	1861.7 ± 0.3g	1881.8 ± 0.4g	1841.0 ± 0.3f
	Total	1806.8 ± 0.1d	1822.2 ± 0.2g	1830.5 ± 0.3h	1859.6 ± 0.2h	1881.3 ± 0.4g	1841.2 ± 0.3f
Summer	CR	1809.4 ± 0.2c	1828.5 ± 0.3d	1840.0 ± 0.2de	1871.5 ± 0.2ab	1894.9 ± 0.4abc	1848.6 ± 0.3b
	TP	1807.0 ± 0.2d	1832.8 ± 0.2b	1840.1 ± 0.2de	1871.6 ± 0.3a	1894.7 ± 0.5bc	1849.3 ± 0.3b
	Total	1807.1 ± 0.1d	1835.8 ± 0.2a	1839.6 ± 0.2ef	1871.5 ± 0.3ab	1895.7 ± 0.5a	1850.0 ± 0.3a
Autumn	CR	1800.4 ± 0.2f	1826.9 ± 0.4e	1842.0 ± 0.2c	1864.5 ± 0.3e	1893.0 ± 0.4d	1843.1 ± 0.3e
	TP	1800.7 ± 0.2f	1831.6 ± 0.4c	1842.9 ± 0.3b	1863.6 ± 0.3f	1895.2 ± 0.4ab	1844.5 ± 0.3d
	Total	1797.6 ± 0.2g	1832.5 ± 0.4b	1844.5 ± 0.3a	1863.6 ± 0.3f	1894.2 ± 0.4c	1844.4 ± 0.3d
Winter	CR	1818.6 ± 0.3a	1823.7 ± 0.4f	1840.3 ± 0.2d	1870.8 ± 0.4b	1892.1 ± 0.2d	1848.8 ± 0.3b
	TP	1809.1 ± 0.1c	1831.4 ± 0.4c	1838.2 ± 0.2g	1865.9 ± 0.4d	1888.9 ± 0.3e	1847.0 ± 0.3c
	Total	1811.5 ± 0.1b	1831.0 ± 0.3c	1839.2 ± 0.2f	1866.7 ± 0.4c	1887.2 ± 0.3f	1847.4 ± 0.3c

25 **Table S3.** Statistics of excluding trajectories over different observing periods during 2004-2017 at

26 WLG station.

		Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Sum
2005-2007	Total	764	1193	938	1190	882	1139	921	1431	1427	843	1272	911	12911
	Reject	157	111	51	371	545	767	632	1325	911	181	133	19	5203
2008-2012	Total	3060	2081	1764	1811	1136	2250	1919	2826	2791	1527	1868	1079	24112
	Reject	41	228	194	467	675	1681	1428	1727	1298	90	0	0	7829
2013-2017	Total	2110	1738	2112	1882	1269	1259	461	1816	2186	1659	1844	1111	19447
	Reject	0	1	0	55	81	0	90	875	138	71	0	12	1323

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Figure S1. The time series of ΔCO , ΔCH_4 and $\Delta\text{CO}/\Delta\text{CH}_4$ slopes from 2004 to 2017 at WLG station.

Figure S2. The geographical distribution of obvious two CH₄ source regions, including the City Region covered from the northeast to southeast (NNE-...SE) of WLG and the Tibetan Plateau located south to west (S-...-W) of the station. The remote sensing image layer was downloaded from© Google Earth.

Figure S3. The annual growth rates of atmospheric CH₄ over different observing periods in 1994-2017 at WLG station. The ‘CR’, ‘TP’ and ‘Total’ represents the measurements from the City Regions, the Tibet Plateau and the total regional records, respectively. Different letters represent the significant difference between regions and periods.

Figure S4. The selected clusters (red color) with air mass transport from city regions, based on the monthly cluster analysis of trajectories during 2005-2017.

Figure S5. The annual growth rates of atmospheric CH₄ over different periods during 2005-2017 at WLG station. ‘TR’ represents the growth rate by regional data that without air mass transport from city regions. ‘Total’ means the growth rate by original regional data series. Different letters represent significant differences among the cases.

Figure S6. Wind-rose distribution of wind frequency and wind speed from 16 horizontal wind directions over different periods during 1994-2017 at WLG station. The different colors represent the CH₄ data in different seasons. Error bars of the wind speed indicate 95% confidence intervals.

Figure S7. The seasonal polar plot of the distribution of CH_4/CO ratio in 2004-2017 at WLG station.

The gradient colors represent the level of ratio values.

Figure S8. The CH₄ emission by per sector in China over 1994-2015 based on Emissions Database for Global Atmospheric Research (EDGAR).

Figure S9. The geographical distribution of the increase of total CH₄ emission over different periods in 1994-2015, based on the data from Emissions Database for Global Atmospheric Research (EDGAR).