Point by point response to the reviewers:

First of all, we would like to thank the reviewers for the positive and constructive comments. The line number below is indicated based on the clean version. The followings are response to the questions of reviewers.

Comments from the reviewers:

-Reviewer #1

General comments: The manuscript presents an analysis of 24 years of continuous in-situ ground-based methane observations at the remote Mount Waliguan station in China. Such records are very essential for monitoring atmospheric variability and trends of this important greenhouse gas. It is challenging to keep those observations up and running for decades. Thus, I fully support publication and release of these data. However, the analysis of the data is very descriptive and lacks conclusive statements. There is hardly any novel insight into the CH_4 cycle, which is currently drawn from the analysis. Several filters are applied to the data but it is difficult to follow how the filters are applied and which filtered dataset is used in which analysis. The paper may profit from merging chapter 3 (Results) and chapter 4 (Discussion). Moreover, the paper contains quite a number of linguistic flaws. Proofreading by an English native speaker is necessary. Some language mistakes are listed below. The paper is within the scope of 'Atmospheric Chemistry and Physics' but – to my mind – requires major revisions prior to its acceptance for ACP. See my specific comments below.

Response: Thank you very much for your good comments. Based on the comments, our paper has improved a lot. We revised the manuscript and answered the questions point by point.

Overall, i) according to your suggestion, we extended our records from 2017 to 2019, to increase the value and novelty of our study. We updated all our data and updated the respective part in our manuscript including the filtered time series (Table 1, Table 2, and Figure 2), the diurnal variations (Figure 3 and Table S1), the seasonal cycles (Figure 10, Figure 11, and Table S2), and the long-term trends

(Table 4, Figure 12, and Figure S6). Please refer to the revised manuscript for details.

ii) Different data filtering methods were further described and the descriptions of which dataset is used were added. For the meteorological approach, we revised the descriptions as below (Line 197-204, Line 283-285).

"In this study, the CH_4 records associated with local surface winds from selected sectors, i.e. NNE-…-ENE in spring, NE-…-SE in summer, NE-…-ESE in autumn, and NE-ENE in winter, were flagged as locally influenced (27.0%). Subsequently, we rejected portion of daytime records to minimize the effect of human activities (16.9%), e.g. 9:00-13:00 LT (local time) in summer, and 10:00-17:00 LT in winter. Finally, we filtered CH_4 data into locally influenced when the surface wind speeds were less than 1.5 m s^{-1} to minimize the very local accumulation (9.2%)."

"To precisely understand the characteristics of atmospheric CH_4, including seasonal cycles and long-term trends, it is vital to identify the CH_4 records in well-mixed air without local contaminations."

For the analysis between the city regions and the Tibetan plateau, the data filtering methods are described as below (Line 205-215).

"In order to investigate the influence of anthropogenic emissions from cities and remote area as Tibetan plateau, we divided the CH_4 data into two main influencing regions according to the analysis, including the geographical conditions, the effect of surface winds, the long-range transports, and the potential source distributions. The first region covers the northeast and southeast (NNE-…SE) of the WLG, which is denoted as City Regions (CR). The second region is located the south to west (S-…-W) of the station and is well known Tibetan (Qinghai-Xizang) Plateau (TP) (Fig. S1). Accordingly, the hourly CH_4 records when the surface winds coming from these sectors were divided into two subsets (i.e. TP and CR). The long-term variations between the two regions as well as the total regional time series were further compared and analyzed."

For the analysis of the long-range transport of emissions from cities, the data filtering methods are described as below (Line 665-674).

"As described above, the northeast and southeast city regions might have acted as strong regional sources influencing the atmospheric CH_4 at the WLG. Therefore, to analyze the effect of long-range transport of emissions from cities, the regionally representative data was further excluded by air mass transport, and the remaining regional records were denoted as 'TR'. First, the monthly cluster analysis was applied to hourly trajectories over 2005-2007, 2008-2012 and 2013-2017. Then, based on the cluster analysis, the clusters were divided into two groups, i.e. from city regions (red clusters in Fig. S8), and other (black clusters in Fig. S8). Finally, the regionally representative data were accordingly classified as two groups based on the cluster results (cities or other). The statistical results were presented in detail in Figure S8 and Table S3."

For other analysis, i.e. diurnal variation, local surface wind, air mass pathways, and potential source distributions, we used total hourly CH_4 data.

iii) We merged chapter 3 (Results) and chapter 4 (Discussion). The interpretation/conclusion was described in conjunction with results, and more discussions were added. Based on the long-term measurements, we concluded the main findings compared to the existing studies (Line 27-34). Generally, the characteristics of CH_4 varied in different observing periods: i) the diurnal cycle has been becoming apparent and the amplitudes of the diurnal or seasonal cycles increased over time, ii) the wind sectors with elevated CH_4 mole fractions switched from ENE-…-SSE sectors in early periods to NNE-…-E sectors in later years, iii) the area of source regions increased as the years progressed and strong sources shifted from northeast (city regions) to southwest (Northern India), iv) the annual growth rates in recent years (e.g. 2008-2019) were significantly larger than that in early periods (e.g. 1994-2007).

iv) We have asked the language company to help the English improvement.

Specific comments:

Abstract is rather long, you may consider shorten it.

Response: Thank you for your suggestion. We have shortened the abstract (Line

18-34) as below.

"A 26-year long-term record of atmospheric methane (CH_4) measured in-situ at the Mount Waliguan (WLG) station, the only WMO/GAW global station in inland Eurasia, is presented. Overall, a nearly continuous increase of atmospheric CH_4 was observed at the WLG with a yearly growth rate of 5.1 ± 0.1 ppb yr^{-1} during 1994-2019, except for some particular periods with near-zero or negative values, e.g. 1999-2000, and 2004-2006. The average CH_4 mole fraction was only 1799.0 ± 0.4 ppb in 1994 but increased about 133 ppb and reached a historic level of 1932.0 ± 0.1 ppb in 2019. The case study in the Tibetan Plateau showed that the atmospheric CH_4 increased rapidly. During some special period, it is even larger than that of city regions (e.g. 6.7 ± 0.2 ppb yr^{-1} in 2003-2007). Generally, the characteristics of CH_4 varied in different observing periods: i) the diurnal cycle has been becoming apparent and the amplitudes of the diurnal or seasonal cycles increased over time, ii) the wind sectors with elevated CH_4 mole fractions switched from ENE-…-SSE sectors in early periods to NNE-…-E sectors in later years, iii) the area of source regions increased as the years progressed and strong sources shifted from northeast (city regions) to southwest (Northern India), iv) the annual growth rates in recent years (e.g. 2008-2019) were significantly larger than that in early periods (e.g. 1994-2007)."

Lines 18-19: write "A 24-year long-term record of atmospheric methane (CH_4) measured in-situ at the Mt. Waliguan station, the only …, is presented."

Response: Revised the sentence (Line 18-19) as you suggested.

Line 20: "… 1994-2017 …": why don't you use more recent data (e.g. until end of 2019)?

Response: Thank you for your question. According to your suggestion, we extended our records from 2017 to 2019, to increase the value of our study. We updated all our data and updated the respective part in the manuscript including the filtered time series (Table 1, Table 2, and Figure 2), diurnal variations (Figure 3 and Table S1), seasonal cycles (Figure 10, Figure 11, and Table S2), and long-term trends (Table 4, Figure 12, and Figure S6). Please refer to the revised manuscript for details.

Line 20: "continuously" need to be "continuous"

Response: We have corrected it (Line 20).

Lines 20-22: "continuous increase" but "even negative growth trend … in particular periods" is a contradiction.

Response: Thank you for the correction. We rewrote it as “a nearly continuous increase” in the revised draft (Line 20).

Lines 24-25: "but unprecendeted elevated ~100 ppb" sounds awkward

Response: We updated the data to 2019 and changed it to “but increased about 133 ppb” (Line 24).

Line 24: "… historic high of 1903.8 ± 0.1 ppb in 2016…"; what about 2017?

Response: We have updated it to 2019. “… reached a historic level of 1932.0 ± 0.1 ppb in 2019” (Line 24).

Line 26: what does "$\triangle CO/\triangle CH_4$" mean? Why do you look at CO to CH_4 ratios?

Response: Thank you for your question. ΔCH_4 and ΔCO are the detrended time series of CH_4 and CO based on the method of Thoning et al. (1989). These are results of the original data minus the trend curve. $\Delta CO/\Delta CH_4$ is the ratio of the ΔCO and ΔCH_4. Because part of the CH_4 and CO in atmosphere are from the same anthropogenic sources (e.g. fossil fuel combustion), the long-term trend of $\Delta CO/\Delta CH_4$ is helpful to understand the variation of the sources/sinks. We added the descriptions about ΔCH_4 and ΔCO (Line 263-266, Line 464-467) as below.

“The detrended values are denoted as ΔCH_4 and ΔCO, which are the original data points minus the trend curve. To accurately obtain the correlation slopes of

ΔCO and ΔCH_4, i.e. $\Delta CO/\Delta CH_4$, a rolling linear regression was applied to the ΔCH_4 and ΔCO time series by the 'roll_lm' function in 'roll' package of R"

"Because parts of the CH_4 and CO in atmosphere were from the same anthropogenic sources (e.g. fossil fuel combustion), the long-term trend of $\Delta CO/\Delta CH_4$ was helpful to understand the variation in the sources/sinks in many studies (Buchholz et al., 2016; Niwa et al., 2014; Tohjima et al., 2014; Wada et al., 2011)."

Line 27: add "elevated" that is reads "… opposite to other elevated sites …"

Response: We have revised the abstract and deleted the sentence.

Line 30: "(the Northern India)" -> "(Northern India)"

Response: We revised it (Line 32).

Line 35: delete "What is interesting" I do not believe that you want to mention uninteresting things in the abstract of your manuscript.

Response: We deleted it (Line 27).

Line 47: "anomalously" -> "anomalous"

Response: We rewrote the abstract and deleted the related sentence.

Lines 47-49: If this statement is presented prominently in the abstract, it also needs to be discussed in the manuscript. Moreover, when looking at Fig. 11 (lower panel), I do not really see an increasing growth rate (but only a mostly positive growth rate).

Response: Thank you for your comment. Yes, the description is inappropriate here, it should be "larger growth rate" and this is rather a statement that fits to a synthesis analysis. We rewrote the abstract and deleted the related descriptions.

The mostly positive values will lead to larger average growth rate, but the increasing growth rate is not distinct in Fig. 11. We listed the periodic growth rates in Table 4 as below. The growth rates during 2008-2019 are larger than that during

1994-2002.

Table 4. Annual growth rates of atmospheric CH_4 in the City Regions (CR), the Tibetan Plateau (TP), and total regional records during 1994-2019 at the WLG station.

	1994-1997	1998-2002	2003-2007	2008-2012	2013-2019	1994-2019
CR	3.0 ± 0.1	3.6 ± 0.2	5.3 ± 0.2	7.0 ± 0.2	6.2 ± 0.1	5.2 ± 0.1
TP	3.4 ± 0.1	3.0 ± 0.2	6.7 ± 0.2	5.7 ± 0.2	5.7 ± 0.1	5.1 ± 0.1
Total	**4.9 ± 0.1**	**2.5 ± 0.2**	4.9 ± 0.1	**7.7 ± 0.1**	**5.5 ± 0.1**	5.1 ± 0.1

Line 60: write "CH_4 has an 8-12 years lifetime …"

Response: We have revised it (Line 45).

Line 64: "… CH_4 rapidly increased …"

Response: We have revised it (Line 49).

Lines 65-66: write "Results from ice core analyses in Antarctica showed …"

Response: We revised the sentence (Line 50-51).

Line 67: write "… has reached a level unprecedented over …"

Response: We have revised the sentence (Line 52).

Line 68: awkward English

Response: We have revised the sentence (Line 53-55) as below.

"At the beginning of the 1990s, the CH_4 mole fraction showed a decreasing trend in. Consequently, the reverse trend has been observed since 1998 due to the higher global mean temperature (Dlugokencky et al., 1998; Nisbet et al., 2014)."

Line 71: delete "special"

Response: We have deleted it.

Line 73: write "… (Nisbet, et al., 2019) followed by a renewed CH_4 increase since then".

Response: We revised the sentence (Line 58).

Line 78: don’t start a sentence with "And …"

Response: We have deleted it.

Lines 78-79: write "unexpected" instead of "not expected"; why the increase is "unexpected"?

Response: We have deleted the related sentence because it is inappropriate here (Line 62). Because the growth rate of CH_4 is originally very low and lasts a long time before 2007, but increases strongly and continuously from 2007 to now. Therefore, we used the ‘unexpected increase’.

Line 84: explain "C.E."

Response: We added “Common Era” (Line 66).

Line 85: write "Atmospheric CH_4 is mainly …"

Response: We have revised it (Line 68).

Lines 105-106: write "Systematic observations are a prerequisite to get an accurate understanding of spatial and temporal behavior of atmospheric CH_4 concentrations."

Response: We revised the sentence (Line 94-95).

Line 113: awkward English

Response: We revised the sentence (Line 101-107) as below.

“Recently, other stations have been installed for CH_4 observation, such as the Barrow (BRW), South Pole (SPO) (polar site), Cape Grim (CGO) and Minamitorishima (MNM) (coastal/island sites), Jungfraujoch (JFJ) and Mount Waliguan (continental mountain site). Hundreds of CH_4 observation stations worldwide are currently running under the framework of the WMO/GAW.”

Lines 114-118: There are many more stations continuously measuring CH_4 levels in the atmosphere; why did you select these ones?

Response: Thank you for your question. We select these stations because i) they just represent different types of global stations, ii) parts of the stations have similar altitude or latitude to Waliguan, e.g. MLO, JFJ, and MNM.

Lines 118-127: this is largely a repetition, merge with paragraph on the previous page (lines 85-104).

Response: We have moved and merged the sentences (Line 87-93).

Lines 146-147: write "… which is the longest record in China."

Response: We have revised it (Line 126).

Lines 157: write "WLG is the only …"

Response: We have revised it (Line 136).

Line 158: write "… and is run by the China Meteorological Administration …"

Response: We have revised it (Line 137).

Line 166: "Tibetan Plateau"

Response: We have revised it (Line 145).

Line 167: "dial variations … are influenced …"

Response: We have revised it (Line 146).

Line 221: write "… were flagged as locally influenced."; if the data are locally influenced they are poorly representative.

Response: Thank you for your comments. We have revised it (Line 200). Yes, the data are locally influenced means they are poorly representative. They represent the CH_4 that are strongly affected by local contaminations or not in well-mixed air. We excluded them to get the regionally representative data. Finally, based on the meteorological method, about 64% of the CH_4 data was classified as regionally representative over 1994-2019 at the WLG (Line 198-204, Table 1).

Lines 220-225: add percentages of rejection according to the individual filters.

Response: We have calculated and added the percentage of each filter (Line 198-204). Overall, about 27.0% by wind sectors, 16.9% from daytime records, and 9.2% by wind speed.

Line 224: "We filtered CH_4 data into local events …" sounds strange, how can you filter data into events?

Response: We have changed "local events" to "locally influenced". The locally influenced data cannot represent the CH_4 in well-mixed air.

Line 226: write "hourly CH_4 data was binned into 16 horizontal wind direction classes …"; is this done for all data or the regionally representative data only?

Response: We have revised the sentence (Line 216-217). This is done for all data.

Lines 235-236: elaborate on HYSPLIT: what's the spatial resolution of the model, what is the height above sea level of WLG in the model?

Response: The spatial resolution of the model is 0.5×0.5 degree and the height is 10 km a.s.l. We added the descriptions in the paper (Line 229-230).

"The spatial resolution of the model is 0.5×0.5 degree and the model height is 10 km a.s.l."

Line 239: write "The trajectories for January, April, …"

Response: We have revised the sentence (Line 231).

Line 285: "appropriately every five years a period" sounds strange.

Response: We have revised the sentence (Line 274-278) as bellow.

"The entire CH_4 time series were divided into five observing periods, i.e. 1994-

1997, 1998-2002, 2003-2007, 2008-2012, and 2013-2019, according to the significant stages or the critical time period of atmospheric CH_4 variations from previous studies."

Results and Discussion chapters (Chapters 3 and 4): I strongly encourage the authors to merge these two chapters. Results should be discussed in stronger conjunction with existing literature, preferably from Asian sites; I don't see the rationale why some findings are compared with conclusions from non-elevated sites in Europe.

Response: Thank you very much for your constructive suggestions. We have merged the section of 'Results' and 'Discussion'.

We added more discussions with existing long-term studies from Asian sites, e.g. Yonagunijima (YON) and Ryori (RYO) in Japan, Sinhagad (SNG) and Cape Rama station (CRI) over India, Ulaan Uul (UUM) in Mongolia, and Tae-ahn Peninsula (TAP) in Korea. Some added descriptions are as below.

"Tohjima et al. (2014) found an opposite variation at the Hateruma Island, which showed small slope values in the summer. Wada et al. (2011) analyzed more than 10-year seasonal variation of the $\Delta CO/\Delta CH_4$ ratios at three monitoring stations, i.e. MNM, Yonagunijima (YON), and Ryori (RYO) in Japan, which also showed an opposite trend to that of the WLG. It was because these sites were considerably affected by the Asian continental source regions, where had enhanced emissions of CH_4 in the summer (rice paddies) and CO in the winter (fuels combustion)." (Line 474-480)

"For other regional sites in the Asia, Guha et al. (2018) studied seasonal variability at the Sinhagad (SNG) and Cape Rama station (CRI) over India, which also showed an opposite trend to the WLG due to the strong impact of monsoon dynamics. Ahmed et al. (2015) found that the seasonal CH_4 showed a maximum in the winter and a minimum in the spring at two urban sites of Guro (GR) and Nowon (NW), in Seoul, Korea over 2004-2013. Kim et al. (2015) investigated the decadal variation (1991-2013) of CH_4 at the East Asian sites, e.g. Ulaan Uul (UUM) in Mongolia and Tae-ahn Peninsula (TAP) in Korea, which revealed again an opposite seasonal trend to that of the WLG." (Line 521-529)

"The seasonal amplitude at the WLG (~14 ppb) was significantly lower than many other sites in the Northern Hemisphere, by about 35-70 ppb. Such sites included MLO in America, BRW in North Pole, UUM in Mongolia, TAP in Korea, Ny-Ålesund in Norway, Bialystok in Poland, Ochsenkopf in Germany, and Beromunster in Switzerland (Dlugokencky et al., 1995; Kim et al., 2015; Morimoto et al., 2017; Thompson et al., 2009; Popa et al., 2010; Satar et al., 2016). MBL also showed a larger amplitude than WLG (Fig. 11). The study at the SNG and CRI over India showed a much larger amplitude close to 200 ppb (Guha et al., 2018)." (Line 546-553)

"Tohjima et al. (2002) found that the CH_4 levels at the Cape Ochi-ishi and Hateruma Island in1995-2000 respectively increased by 4.5 and 4.7 ppb yr^{-1}, which were also similar to that of the WLG. Tsutsumi et al. (2006) analyzed the trend of hourly CH_4 data from 1998 to 2004 on the YON, which showed a similar increase (~3.0 ppb yr^{-1}) to the WLG. The study at the GR and NW in Seoul, Korea, presented almost an identical trend of 2 ppb yr^{-1} between 2004 and 2013 (Ahmed et al., 2015), which was lower than that of the WLG in similar period." (Line 586-592)

"The growth rate of CH_4 observed at the Ny-Ålesund, Svalbard, increased from 0.3 ± 0.2 ppb yr^{-1} during 2000-2005 to 5.5 ± 0.2 ppb yr^{-1} during 2005-2014, which had a similar variation but with a little lower growth rates than that of the WLG (Morimoto et al., 2017). The study suggested that the temporal pause in 2000-2005 was ascribed to the reductions of CH_4 emissions from the microbial and fossil fuel sectors, while the increase in 2005-2014 was due to an increase in microbial release." (Line 598-604)

Additionally, we also compared the result with adjacent stations in China, e.g. Shangdianzi, Lin'an. For other sites with similar latitude or altitude in the Northern Hemisphere, e.g. Mauna Loa and Jungfraujoch, are also compared.

Lines 297-298: awkward English

Response: We have revised the sentence (Line 308-309) as bellow.

"In winter, a large increase of CH_4 was found during 9:00-17:00 LT, with the largest peak to trough amplitude of 7.1 ± 2.9 ppb."

End of Chapter 3.1: interpretation/conclusion missing.

Response: We merged the section of 'Results' and 'Discussion' and rewrote the Chapter 3.1 (Chapter 3.2 now). The interpretation/conclusion was described in conjunction with results (Line 310-333). Please refer to the revised manuscript.

Line 315: "As observed by the previous short-term observations"; I don't understand this statement.

Response: We have changed to "Similar to the previous studies…" (Line 319).

Line 326: Delete "What interesting is"

Response: We have deleted it.

Line 345: Write "It's obvious that CO showed …"

Response: We have revised it (Line 391).

Line 348: write "when percentages ranged from 0 to 40."; write "When data exceeded the 60% percentile, the high area probability areas …"

Response: We have revised the sentence (Line 394-395).

End of Chapter 3.2: interpretation/conclusion missing.

Response: We merged the section of 'Results' and 'Discussion' and rewrote the Chapter 3.2 (Chapter 3.3 now). The interpretation/conclusion was described in conjunction with results (Line 345-365, Line 374-375, Line 383-391, and Line 395-399). Please refer to the revised manuscript.

Line 365, 368-369: awkward English

Response: We have revised the sentence (Line 414-419) as below.

“Cluster 3 showed the highest CH_4 mole fraction, with an enhancement of ~4 ppb relative to the seasonal average. In winter, the air masses primarily came from northwest and southwest regions, e.g. cluster 3 (59%), and cluster 1 (34%) (Fig. 6d) and the cluster 1 brought the highest CH_4 mole fractions with an enhancement of ~7 ppb over the seasonal average.”

End of Chapter 3.3: interpretation/conclusion missing.

Response: We merged the section of ‘Results’ and ‘Discussion’, and rewrote the Chapter 3.3. The interpretation/conclusion was described in conjunction with results (Line 420-433, Line 451-461).

Line 389: write "regionally representative"

Response: We have revised all the descriptions in the manuscript.

Line 390: write ""locally influenced"

Response: We have revised all the descriptions in the manuscript.

Line 391: "data was … larger than … events"; awkward English

Response: We have revised the sentence (Line 288-289) as below.

“The average of the locally influenced data (1868.2 ± 0.3 ppb) was larger than that of the regionally representative records (Table 1).”

Line 394: Does it hold true for all data or regionally representative data?

Response: It hold true for both of them. The average of regionally representative data and all data both showed increasing trend over 1994-2019.

Line 396 (reference to Fig. 7): I suggest to move Fig. 7 above and make it to Fig. 2; show first the whole dataset before you analysis of the data

Response: Thank you very much for your suggestion. We changed Fig. 7 to Fig. 2 and moved Chapter 3.4 above as Chapter 3.1.

Line 402: what is deltaCO and deltaCH4? I assume it is excess CO and excess CH_4, i.e. data above baseline (looks like when looking at Fig. S1); if so, how was the background determined? Please elaborate; show also the CO time series.

Response: Thank you for your questions. Similar to the question you posed on L26, ΔCH_4 and ΔCO are the detrended time series of CH_4 and CO from 2004-2017 based on the method of Thoning et al. (1989).

The detrended value is the original data points minus the trend curve. The trend value is the polynomial part of the function plus the long-term filter of the residuals. We have added the descriptions in the manuscript (Line 261-266).

The ΔCH_4 and ΔCO time series are showed in Figure S4. The original hourly CO time series was also added in the paper (Figure S3).

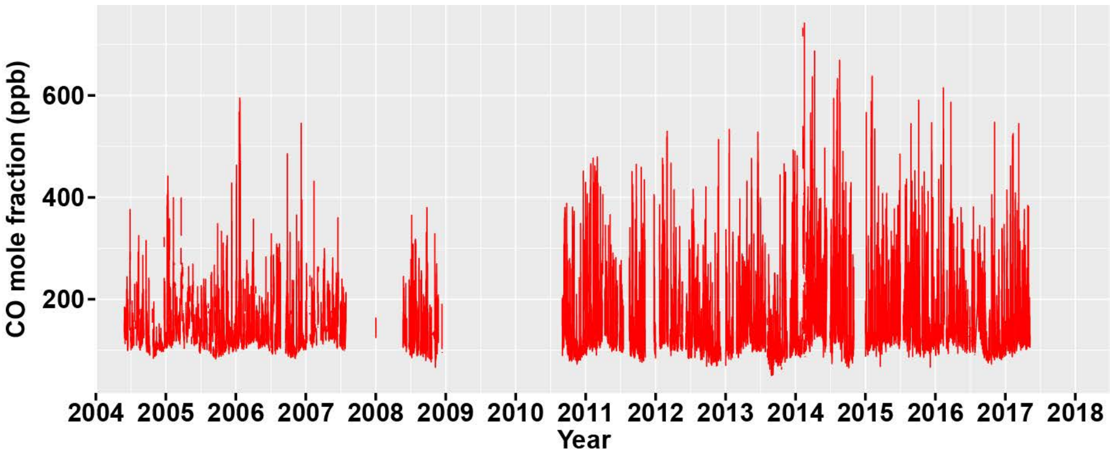

Figure. S3. The hourly CO data from 2004 to 2017 at the WLG station.

Line 411: incomplete sentence.

Response: We deleted the sentence because there is a repetition of above descriptions.

End of Chapter 3.5: what do we learn from the different deltaCO to deltaCH4 ratios?

Response: The strong seasonal variation of $\Delta CO/\Delta CH_4$ also revealed that the WLG was affected by different anthropogenic sources, e.g. sources from cities, and sources from the Tibetan Plateau, during a year, especially in the summer and winter. The long-term trend of the slopes implied that the source emission types (CO sources or CH_4 sources) around the WLG might have been changing with human activities, like straw burning, in the early years, or coal mining in recent years. We revised the Chapter 3.5 and added the descriptions (Line 498-504). Please refer to the revised manuscript.

Lines 415-419: this is a different filtering than the one described above, right? Move these lines to Chapter 2.3.

Response: Yes, this is a different data filtering method. We are using this method to further investigate the influence of anthropogenic emissions from cities and remote area as Tibetan plateau. We moved the paragraph to Chapter 2.3 (Line 205-215).

Line 426: write "… the seasonal averages of regionally representative CH_4 were …"; do these numbers relate to regionally representative data?

Response: We have revised it. Yes, the seasonal averages are calculated based on regionally representative data.

End of Chapter 3.6.1: interpretation/conclusion missing.

Response: We merged the section of 'Results' and 'Discussion' and rewrote the Chapter 3.6.1. The interpretation/conclusion was described in conjunction with results (see Chapter 3.6.1, Line 518-562).

Line 442: write "… growth rates were very small or even negative …"

Response: We have revised it (Line 567).

Lines 466.468: How is this filtering exactly done? Please explain. How many data (in %) are remaining?

Response: The regionally representative data are further filtered based on the hourly trajectories of cluster analysis.

First, the monthly cluster analysis was applied to hourly trajectories over 2005-2007, 2008-2012 and 2013-2017. Then, based on the cluster analysis, the clusters were divided into two groups, i.e. from city regions (red clusters in Fig. S8), and other (black clusters in Fig. S8). Finally, the regionally representative data were accordingly classified as two groups based on the cluster results (cities or other). We added the descriptions in the manuscript (Line 669-674).

Figure S8, Table S3, and Table 5 showed the filtered results in detail. Eventually, 67.1% of regionally representative data are remaining.

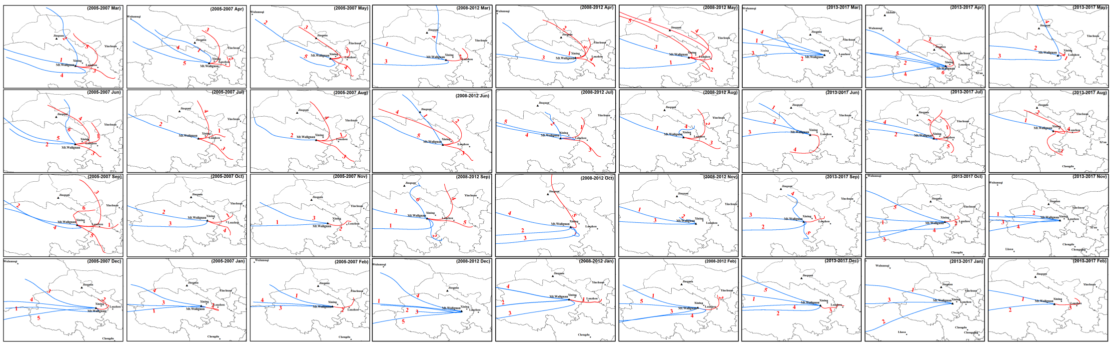

Figure S8. The monthly cluster analysis of hourly trajectories during 2005-2017 at the WLG station. The red clusters represent air masses transport from city regions.

Table S3. The statistics of the excluding trajectories over different observing periods during 2004-2017 at the 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

Table 5. The statistics of CH_4 data without air mass transport from city regions (TR) over different periods during 2005-2016 at WLG station.

	Transport regions	Hours	Percentage (%)	Average (ppb)	Updated growth rate (ppb yr-1)
2005-2007	TR	6922	77.2	1824.9 ± 0.2	2.7 ± 0.2
	City	2041	22.8	1835.9 ± 0.5	-

2008-2012	TR	7060	64.4	1853.7 ± 0.2	10.1 ± 0.1
	City	4254	35.6	1861.0 ± 0.3	-
2013-2016	TR	4152	61.6	1888.2 ± 0.3	6.3 ± 0.1
	City	2591	38.4	1888.5 ± 0.5	-
2005-2016	TR	18134	**67.1**	1850.6 ± 0.2	7.0 ± 0.1
	City	8886	32.9	1863.2 ± 0.3	-

Line 481: "no significant trend was found …" what does that mean? How do you interpret this finding?

Response: It means that the CH_4 from the Tibetan Plateau has similar growth rates to that from city regions. We revised the description (Line 581-582) as below. "Similar growth rates were found between the CR and the TP during 1994-2019 (Fig. S6)."

The finding implied that i) the atmospheric CH_4 at the WLG was not predominantly influenced by eastern cities in recent years and ii) large amounts of CH_4 were transported from the Tibetan Plateau to WLG in recent years (Line 699-702). Through the analysis of spatial distribution of emissions (Fig. 7, Fig. S10), the emission from southwest to south regions, i.e. Northern India, increased strongly after 2007, which may have contributed a lot to the high growth rates in the Tibetan Plateau in recent years.

Line 495: Write "WLG was increasingly affected by local sources …" which ones are these?

Response: We revised the sentence (Line 331-333).

"The increasing amplitude and CH_4 mole fractions suggested that the WLG was increasingly affected by local and regional anthropogenic sources, such as gas exploitation, and grazing."

Line 500: "… higher CH_4 … in past years"; which years, be more explicit.

Response: We revised the descriptions, "In 1994-2002, the CH_4 mole fractions …" (Line 310).

Line 505: this statement is trivial as Zhou et al. used the same data, right?

Response: Zhou et al. (2004) used the data from 1991 to 2002. However, we are using much longer observation records than them. The compared period is also not absolutely the same. Through the 26-year measurements, we found different results, which provided a broader view of changing characteristics of CH_4 to understand CH_4 cycle.

In this chapter 3.3, compared to Zhou et al. (2004), we found that wind sectors with elevated CH_4 mole fractions changed over time (Line 366-375). The elevated CH_4 was predominately from the ENE-..-SSE sectors in the early years (Fig. 4a-b) but evolved to the NNE-NE-ENE-E sectors in later years (Fig. 4c-e).

Line 509: "… which could emit large amount of CH_4 by human activities"; write "large amounts"; statement is very vague, can you be more specific?

Response: We have revised the sentence as below (Line 353-359).

"The two largest cities of Xining (with population of ~2.2 million) and Lanzhou (with population of ~4 million) are also situated in the northeast and east of the WLG, respectively. The heavy human activities from anthropogenic fossil combustion, landfills, and livestock, could have also emitted large amounts of CH_4. Based on the data from the Emissions Database for Global Atmospheric Research (EDGAR), the increase of CH_4 emission was 500 kg yr^{-1} in these two regions throughout 1994-2015 (Crippa et al., 2019)."

Line 512: write "… may also contribute to the high CH_4 …"; statement is very vague, can you be more specific?

Response: We have revised the sentence as below (Line 359-365).

"Also, the Yellow River Canyon industrial area (YRC), which is ~500 km northeast of the WLG, may also have contributed to the high CH_4 values (Zhou et al., 2003). With the rapid development of land-use, water utilization, and agriculture sources in the YRC, large CH_4 emissions could have easily transported to the WLG. Previous studies on black carbon (BC) and carbon monoxide (CO) also revealed that the high CH_4 values at the WLG in winter were a result of transport from the YRC (Tang et al., 1999; Zhou et al., 2003)."

Chapter 4.1 misses quantitative statements; what's new compared to existing literature?

Response: We merged Chapter 4.1 with the 'Results' and added more descriptions (see Chapter 3.2 and 3.3). The interpretation/conclusion was added in conjunction with results.

Compared to previous studies, we found that i) the diurnal variations were ambiguous before 2002, but significant diurnal cycles appeared afterward, ii) the highest CH_4 mole fraction was found in winter over 1994-2002, but in summer during the periods of 2003-2007, 2008-2012, and 2013-2019, iii) the wind sectors with high CH_4 mole fractions changed and concentrated on ESE-ENE sectors, and iv) the amplitude of enhancements was increasing.

Line 530: "It was possibly due to …"; very vague statement, can you be more conclusive?

Response: We have revised the statement (Line 424-433).

"The higher values seen in the northwest to southwest airflow were because the air masses had passed through the northwest of the Qinghai province and the central area of the Xinjiang Uygur Autonomous Region (XUAR). This is where the Ge'ermu urban area (the second largest city of Qinghai) was located and where there was with rapid industrial development, natural gas and petroleum resource exploitation, and residue burning of large crops, hence the CH_4 emissions were strong (Fang et al., 2013; Zhang et al., 2013). This result was also similar to the CPF percentile analysis (Fig. 5), as time went by, the southwest or northwest regions that were farther away from the site became the strongest source regions to the WLG."

Line 536: "the southwest or northwest region … may be also strong source regions"; very vague statement.

Response: We have revised it as “This result was also similar to the CPF percentile analysis (Fig. 5), as time went by, the southwest or northwest regions that were farther away from the site became the strongest source regions to the WLG.” (Line 431-433).

Line 540-542: awkward English; no conclusive statement.

Response: We have revised the sentence (Line 451-453) as below.

“More CH_4 sources appeared at the WLG along with the progression of time, which could have been attributed to the influence of human expansion.”

Lines 544-545: add reference.

Response: We added the reference (Fu et al, 2012) (Line 457).

Line 556: "It is of great possible …" awkward English

Response: We have deleted it.

Line 573: Hateruma Island is at sea level, why do you compare WLG with this station?

Response: We compared WLG with Hateruma Island (Tohjima et al, 2014), because the studies they reported about the long-term slope of $\Delta CO/\Delta CH_4$, and we used similar method by Tohjima et al. (2014) to calculate $\Delta CO/\Delta CH_4$. In addition, as an island station, it could well capture the CH_4 signal that is unaffected by human activities.

Line 582: awkward English

Response: We have merged Chapter 4.2 with ‘Results’, the statement is a repetition and we deleted it.

Chapter 4.2 is very descriptive and lacks conclusions

Response: We merged the section of ‘Results’ and ‘Discussion’ and added interpretation/conclusion in conjunction with result (see Chapter 3.4.1 and 3.4.2).

Lines 592-597: move in front of line 591.

Response: We have moved the sentence (Line 530-535).

Line 602: start station names with upper case letters; why do compare with these stations, which have different characteristics.

Response: We revised them (Line 546-550). The seasonal amplitude at WLG was lower than many other sites in the Northern Hemisphere. We compared these stations because they are typical stations with large seasonal amplitude. We also added more discussions to compare the study with Asian sites.

"The seasonal amplitude at the WLG (~14 ppb) was significantly lower than many other sites in the Northern Hemisphere, by about 35-70 ppb. Such sites included MLO in America, BRW in North Pole, UUM in Mongolia, TAP in Korea, Ny-Ålesund in Norway, Bialystok in Poland, Ochsenkopf in Germany, and Beromunster in Switzerland (Dlugokencky et al., 1995; Kim et al., 2015; Morimoto et al., 2017; Thompson et al., 2009; Popa et al., 2010; Satar et al., 2016). MBL also showed a larger amplitude than WLG (Fig. 11). The study at the SNG and CRI over India showed a much larger amplitude close to 200 ppb (Guha et al., 2018)."

In order to fully discuss the characteristic of CH_4 at WLG, based on the existing studies, we compared WLG with both adjacent stations around China and other stations with similar altitude or latitude in the Northern Hemisphere, e.g. Asian sites, European sites, and American site.

Line 611: why is the photochemical capacity weak? I would expect a high photochemical activity.

Response: Yes, the photochemical capacity is high in summer. We have revised the statement (Line 539-542). But as a plateau station with lower VOC and O_3, the oxidizing capacity at WLG is far below the lower altitude area.

Lines 630-631: numbers were already given in lines 454 ff.; repetition won't be needed if Results and Discussions are merged.

Response: Thank you for your good suggestion. We have merged ‘Results’ and ‘Discussion’.

Lines 644-645: why do you compare with a Swiss site? You may compare with composite numbers, e.g. from the WDCGG data summary report (most recent version is #43); downloadable on the WDCGG webpage.

Response: Thank you for your comments. We have compared the data from WDCGG sum43 as well as WMO GHG Bulletin, e.g. (Nisbet et al., 2016; 2019), (WMO, 2019; 2020) (Line 605-613). The comparisons of other sites with similar altitude or latitude are also listed, aimed to fully discuss the long-term CH_4 growth in the world. We also added more discussions about the Asian sites (Line 586-592 and 598-604).

Lines 656-657: are the conclusions drawn for the Swiss site also true for WLG?

Response: To our knowledge, until now, the reasons of the anomalous spikes or strong growth of atmospheric CH_4 have not yet been determined. The conclusion raised by the study at Beromünster, Swiss is also a possible reason. However, we are trying to provide more information to figure the cause for these spikes.

We analyzed the long-term variation between city regions and Tibetan Plateau, the CH_4 emission from different sectors in China, and source changes over different periods. We also discussed the potential reasons provided by other studies (Line 631-645). We concluded that i) the emission from solid fuel (e.g. coal) and rice cultivation may contribute to the anomalous increase (Line 649-653), ii) large emissions from Northern India in recent years may have contributed to the anomalous increase at the WLG (455-461), iii) the warming in the Tibetan Plateau is also an important factor (Line 693-695).

Lines 658-680: this paragraph may better fit into the introduction.

Response: Thank you for your comments. The paragraph describes the CH_4

emissions based on the data from EDGAR. It could fit into the introduction. But based on previous studies, we already generally described CH_4 sources using one paragraph in the introduction (69-93).

This paragraph particularly describes the CH_4 emissions from different sectors during 1995-2015 in China, which showed a similar period to our study. The data could be used to discuss the potential reasons for CH_4 growth at the WLG. Hence, we merged the paragraph with 'Results' (Chapter 3.6.2).

Lines 691-692: "suggested that there were possibly other strong CH_4 sources …": which ones? The following lines do not provide any answer.

Response: Based on our study, it's the strong sources from Northern India. India with abundant cattle as well as an extensive large-scale coal mining operation possibly contributed large amounts of CH_4 to move from northern India to the northeastern Tibetan Plateau (Fig. 7i & l). We have revised the sentence as below (Line 685-687).

"These results suggested that there were possibly other strong CH_4 sources at the WLG that were not from cities and the southwest region (Northern India) was the most likely contributor."

Line 702: "due to the emission from the two largest source regions": be more specific.

Response: We have revised the sentence as below (Line 697-699).

"This would be especially true with the scenario of quickly increasing CH_4 on the Qinghai-Tibetan Plateau due to the emissions from the two largest source regions of Northern India and Eastern China."

Lines 704-705: I don't understand what wants to be said here.

Response: We would like to show the importance of studying carbon cycle in the Tibetan Plateau. We merged the 'Results' and 'Discussion'. The sentence is not appropriate here. Hence, we deleted it and added the descriptions as below (Line 696-702).

“The rapid increase of CH_4 would probably make it difficult to meet the goals of carbon emission reduction in the future. This would be especially true with the scenario of quickly increasing CH_4 on the Qinghai-Tibetan Plateau due to the emissions from the two largest source regions of Northern India and Eastern China. The large growth rate of atmospheric CH_4 in the TP revealed that i) the atmospheric CH_4 at the WLG was not predominantly influenced by eastern cities in recent years and ii) large amounts of CH_4 were transported from the Tibetan Plateau to WLG in recent years.”

Line 705: "anomalously" -> "anomalous"

Response: We have revised it.

Lines 706-707: how about trends in other regions in the world? This statement is the same as in the abstract and I still don’t understand it. This is rather a statement that fits to a synthesis analysis as it is e.g. done in the annual WMO GHG bulletin.

Response: The trend at WLG is similar to other stations in the Northern Hemisphere, and also the global level reported by WMO and WDCGG sum43, especially after 2007, with high growth rate.

Thank you for your comment. Yes, the statements are not appropriate here, and they fit to a synthesis analysis better. We rewrote the abstract and conclusion and revised the related sentence (Line 715-719).

“Additionally, the Tibetan Plateau was intensively affected by strong sources over time, which showed a larger growth rate than that of the city regions in some periods. The anomalous variation and unprecedented growth rate of the atmospheric CH_4 in this region revealed that it was urgent to control CH_4 emissions.”

Lines 722-723: "… the long-term verification is extremely important to … understand CH_4 variations …"; did the understanding improve based on the present analysis? Which are the lessons-learnt?

Response: We have revised the conclusion and deleted the related sentence.

The 26-year measurements provided a broader view of changing characteristics of CH_4 at WLG and improved our understanding of the future trend, such as three developing stages of CH_4 (Fig. 12a), rather than a limited view on CH_4 growth, e.g. steady or negative growth in 1994-2006. We also discussed the potential reasons for the increasingly long-term trend (Chapter 3.6.2 and 3.7).

In this study, we found new characteristics of CH_4 at WLG. Generally, the characteristics of CH_4 varied in different observing periods: i) the diurnal cycle has been becoming apparent and the amplitudes of the diurnal or seasonal cycles increased over time, ii) the wind sectors with elevated CH_4 mole fractions switched from ENE-…-SSE sectors in early periods to NNE-…-E sectors in later years, iii) the area of source regions increased as the years progressed and strong sources shifted from northeast (city regions) to southwest (Northern India), iv) the annual growth rates in recent years (e.g. 2008-2019) were significantly larger than that in early periods (e.g. 1994-2007).

Lines 725-726: "Tibetan Plateau was with the highest average altitude …" awkward English.

Response: We have deleted the description.

Line 727: "anomalously" -> "anomalous"

Response: We have revised it (Line 717).

Data availability statement: very strangely, this paragraph only refers to the data from the other stations but nothing is said how to access WLG data. The data used in the analysis doesn't seem to be freely available since WDCGG only contains daily and monthly CH_4 averages from WLG; where can anybody access hourly CH_4 data from WLG?

Response: Thank you for your question. We will upload the data as supplementary material.

-Reviewer 2

General comments: In this manuscript, a 24-year long-term observation of atmospheric CH_4 at Waliguan WMO/GAW global station in the Tibetan Plateau was studied. This report is very meaningful and the analysis of the paper is very comprehensive. The CH_4 variations and its related potential causes during the long-term observation have been analyzed in details, which would help the scientific community to understand carbon cycle and formulate more informed carbon reduction policy. Given the importance and value of the long-term measurements of CH_4 in the Tibetan Plateau plus that this manuscript is well drafted, I would recommend accepting this paper after minor modifications listed as below.

Response: Thank you very much for your positive comments. We revised the manuscript and answered the questions point by point.

Specific comments:

Line 98: add 'About' before 90%.

Response: We have added it (Line 82).

Line 233-238: How frequent is the backward trajectory computed? Hourly? Please specify the numbers of trajectories are determined.

Response: Yes, it's hourly. We computed the 3-day back trajectories coincident with hourly CH_4 mole fractions. The number of trajectories was determined by the valid hourly CH_4 observations. We added the descriptions (Line 229-230). The related information, e.g. numbers, and average mole fractions, was listed in Table 3.

Some expression is not professional, e.g. "long-distance transport" (Lines 465), can be expressed by "long-range transport", and so on. Please polish the whole report.

Response: Thank you for your suggestion. We have revised all the descriptions in the manuscript.

There are several places in results and discussion where too many details are given, which make the text a little difficult to follow. Results and discussion are suggested to be merged.

Response: Thank you very much for your constructive suggestion. We merged 'Results' and 'Discussion' and rewrote the Chapter. The interpretation/conclusion was described in conjunction with results.

Lines 685-686: Suggest to give more discussions about the larger growth rates in the Tibetan Plateau than that of city region.

Response: We rewrote the Chapter 3.7. The interpretation/conclusion was described in conjunction with results. More discussions were added (Line 685-702).

"These results suggested that there were possibly other strong CH_4 sources at the WLG that were not from cities and the southwest region (Northern India) was the most likely contributor. The PSCF analysis also supported this result (Fig .7). At present, Northern India and Eastern China were the two largest sources of CH_4 at the WLG (Fig. S10) (Crippa et al., 2019). Since the Tibetan Plateau was coincidently trapped in the middle of them, the atmospheric CH_4 at WLG was very likely dominated by long-range transport from these two regions. Although CH_4 emissions increased slowly during 1994-2002, a negative trend appeared (Fig. S10), significantly increased emissions were found in both southeast and southwest Asia after 2007. Chen et al. (2013) illustrated that the warming (0.2 °C per decade) in the Tibetan Plateau resulted in substantial emissions of CH_4 due to the thawed permafrost and melted glaciers. The rapid increase of CH_4 would probably make it difficult to meet the goals of carbon emission reduction in the future. This would be especially true with the scenario of quickly increasing CH_4 on the Qinghai-Tibetan Plateau due to the emissions from the two largest source regions of Northern India and Eastern China. The large growth rate of atmospheric CH_4 in the TP revealed that i) the atmospheric CH_4 at the WLG was not predominantly influenced by eastern cities in recent years and ii) large amounts of CH_4 were transported from the Tibetan Plateau to WLG in recent years."

Line 1057: delete 'CO'.

Response: We are sorry for the mistake. The average CO mole fractions were added in Table 3.

Why are the points not on the line in figure 8 and 9?

Response: Thank you for your question. The lines are the smoothed curves of the points using the method of 'LOESS' Curve Fitting (Local Polynomial Regression), hence they do not just connect the points.

Changing characteristics of atmospheric CH_4 i~~o~~n the Tibetan Plateau:~~,~~ records from 1994 to ~~2017~~2019 at the Mount Waliguan station

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Abstract. A 26~~4~~-year long-term ~~observation~~ record of atmospheric methane (CH_4) ~~was presented~~measured in-situ ~~-~~at the Mount ~~t.~~Waliguan (WLG) station, the only WMO/GAW global station in inland ~~of~~ Eurasia, is presented. Overall, a ~~during 1994-2017,~~ nearly continuous~~ly~~ increase of atmospheric CH_4 was observed at the WLG with a yearly growth rate of 5.1 ± 0.1 ppb yr^{-1}~~,~~ during 1994-2019, ~~although near-zero and even negative growth appeared in~~ except for some particular periods with near-zero or negative values, ~~e.g.,~~e.g. 1999-2000, and 2004-2006. The average CH_4 mole fraction was only 1799~~805~~.0~~8~~ ± 0.4~~1~~ ppb in 1994~~5,~~ but ~~unprecedented elevated ~~increased about ~~100~~ 133 ppb and reached a historic ~~high~~ level of 19~~0~~32.0~~8~~ ± 0.1 ppb in ~~2016~~2019. ~~The seasonal averages of atmospheric CH_4 at WLG were ordered by summer, winter, autumn and spring, and the correlation slopes of ΔCO/ΔCH_4 showed a maximum in summer and minimum in winter, which was almost opposite to other sites in the northern hemisphere, e.g., Mauna Loa, Jungfraujoch, and was caused by regional transport.~~ The case study in the Tibetan Plateau showed that the atmospheric CH_4 increased rapidly. During some special period, it is even larger than that of city regions (e.g. 6.7 ± 0.2 ppb yr^{-1} in 2003-2007). Generally, the characteristics of CH_4 varied in different observing periods~~Strong potential sources at WLG were predominately identified in northeast (cities, e.g., Xining, Lanzhou) and southwest (the Northern India), and air masses from west and northwest regions were accompanied with higher CH_4 mole fractions than that from city regions.~~

~~What is interesting is that obviously changes appeared in different observing periods~~:~~. Generally,~~ i) the diurnal cycle has been becoming apparent and the amplitudes of the diurnal or seasonal cycles increased over time~~were continuously increasing over time~~, ii) the wind sectors with elevated CH_4 mole fractions switched~~moved~~ from ENE-…-SSE sectors in early periods to NNE-…-E sectors ~~(city regions)~~ in later years, iii) the area of source regions ~~was~~ increased~~ing~~ ~~along with~~as the years~~,~~ progressed and strong sources ~~gradually~~ shifted from northeast (city regions) ~~-~~to southwest (Northern India),

iv) the annual growth rates in recent years (~~e.g.,~~e.g. 2008~~13~~-~~2016~~2019) were significantly ~~significantly~~ larger than that in early periods (~~e.g.,~~e.g. ~~1998~~1994-~~2012~~2007). ~~We conclude that the site was more and more affected by regional sources along with the time. Northern India was possibly becoming the strongest source area to WLG rather than city regions before. The case study in the Tibetan Plateau showed that the atmospheric CH_4 observed in Qinghai-Tibetan Plateau changed not as expected, the annual growth rate was even larger than that in city regions in some period (e.g., 7.3 ± 0.1 ppb yr^{-1} in 2013-2016). It is unambiguous that the anomalously fluctuations of atmospheric CH_4 in this region are a warning to the world, its increasingly annual growth rate may be a dangerous signal to global climate change.~~

1 Introduction

Since the pre-industrial era, the emission~~s~~ of greenhouse gases (GHGs) has~~ve~~ increased continuously~~,~~ and large~~r~~ ~~absolute~~ increases ~~were~~ have been found in recent years, with the concentration now being higher than ever~~ now~~ (WMO, 2019). The GHGs~~ could~~ perturb the infrared radiation balance, which ~~-~~traps the heat in the atmosphere and~~,~~ ~~which~~ contributes to global warming, melting glaciers, extreme weather events and many other global climate changes (IPCC, 2014). The recent 30-~~-~~year span from 1983 to 2012 was~~ere~~ the warmest of the last 800 ~~-~~years in the Northern Hemisphere, and half of the rising surface temperatures were~~as~~ due to increased GHGs emissions (IPCC, 2014). As one of the most important GHGs, methane (CH_4) has a~~the~~ global warming effect that~~of~~ ~~methane (CH_4)~~ is just less than~~after~~ carbon dioxide (CO_2) (Etminan et al., 2016). CH_4~~It~~ has an 8-12 year~~s atmospheric~~ lifetime (Battle et al., 1996), with ~~the~~ a global warming potential~~ of~~ ~23 times greater than CO_2 over a 100-~~-~~year horizon (Weber et al., 2019). CH_4~~About~~ also contributed to about 17% of the radiative forcing caused by long-lived greenhouse gases was ~~contributed by~~from~~ CH_4 during~~ 1750 to ~~-~~2016 (Etminan et al., 2016). Since the beginning of the industrial era, the concentration of CH_4 ~~is~~ rapidly increased because of the influence of anthropogenic activities (Saunois et al., 2016). ~~The r~~Results from~~by the analyses of~~ ice cores analyses in Antarctica showed that the atmospheric concentration of CH_4~~ has~~ reached a level unprecedented ~~unprecedented~~ over the last 0.8 million years (IPCC, 2014).

At the beginning of the 1990s, the CH_4 mole fraction showed a decreasing trend. Consequently, the reverse trend has been observed since 1998 due to the higher global mean temperature~~In the beginning of 1990s, CH_4 concentration appeared a decreasing trend in global scale. However, high growth rates were found in 1998, which was possibly due to the higher global mean temperature~~ (Dlugokencky et al., 1998; Nisbet et al., 2014). However~~Subsequently~~, a low growth rate was sustained over 1999 to 2006, except for ~~special~~ years ~~(2002/2003)~~ with El Niño events (2002/2003) (Dlugokencky et al., 1998). The annual growth rate~~s~~ dropped from ~12 ppb yr^{-1} to near

0 from the late 1980s to ~~1999~~ 2006 ~~(Nisbet et al., 2019)~~, followed by a renewed CH_4 increase since then ~~. But thereafter, the atmospheric CH_4 concentration keeps rising from 2007~~(Nisbet et al., 2019). During 2007-2013, the annual growth rate of methane was 5.7 ± 1.2 ppb yr^{-1}. After 2013, the atmospheric CH_4 grew ~~even~~ at rates not observed ~~after 1980s~~, such as 12.7 ± 0.5 ppb yr^{-1} in 2014 and 10.1 ± 0.7 ppb yr^{-1} in 2015 (Nisbet et al., 2016, 2019). In the most recent decade, t~~T~~he ~~overall~~ global mean growth rate was 7.1 ppb yr^{-1} ~~in recent 10 years~~ (WMO, 2019). ~~And the not expected increase since 2007 would make it difficult to meet the targets of carbon emission reduction in the future.~~ The World Meteorological Organization/Global Atmospheric Watch P~~p~~rogramme (WMO/GAW) annual greenhouse gas bulletin revealed that the global~~ly~~ average~~d~~ ~~CH_4~~ mole fraction of CH_4 reached a new high with 1869 ± 2 ppb in 2018 (Rubino et al., 2019), which was ~259% of pre-industrial levels (~722 ppb around 1750 C.E. (Common Era)) (Etheridge et al., 1998; WMO, 2019).

~~The a~~Atmospheric CH_4 is mainly emitted from natural sources (e.g. about 40% from~~,~~ ~~e.g.,~~ ruminants and wetlands) and anthropogenic sources (e.g. about 60% from~~,~~ ~~e.g.,~~ paddies, cattle ranches, coal mines, fossil fuels, and biomass burning) (Hausmann et al., 2016; Saunois et al., 2016). Observations~~Studies~~ from the GAW ~~observations~~ indicated that the causes of the recent increase were likely attributed to anthropogenic emissions at mid-latitudes in the N~~n~~orthern H~~h~~emisphere and the wetlands in the tropics (WMO, 2019). The rapid development of population growth, economic expansion, and ~~countries~~ urbanization of countries has led to more and more fossil fuel production and consumption (~~e.g.~~e.g.~~,~~ the large-scale exploitation of natural gas, oil, and coal) and biomass burning.~~,~~ C~~c~~onsequently, large amounts of anthropogenic CH_4 were emitted around the world in recent years (Galloway, 1989; Streets and Waldhoff, 2000; Wang et al., 2002; Lin et al., 2014; Hausmann et al., 2016). The recent carbon isotope study revealed that biogenic emissions might also have driven CH_4 increases, including microbial sources ~~whether~~ from rice, paddies, ruminants, termites, enteric fermentation, or a combination~~all~~ of these (Nisbet et al., 2016; Schaefer et al., 2016; Wolf et al., 2017).

About 90% of the CH_4 destruction in the atmosphere is~~are~~ ~~mainly~~ from the reaction with hydroxyl radicals (·OH) (Vaghjiani and Ravishankara, 1991; Bousquet et al., 2011), an important oxidant in the troposphere (Logan et al., 1981). Therefore, the inter-annual ~~variability~~ variation of ·OH or the decline of the oxidative capacity of the atmosphere may also cause the recent~~ly~~ increase in the~~d~~ CH_4 growth rates (Rigby et al., 2017; Turner et al., 2017). T~~Even though, t~~he exact causes of significant~~ly~~ increase in the~~d~~ CH_4 emissions in past years ~~are still~~ remain~~ed~~ ~~unclear and~~ debated, especially for the anomalous periods with a suddenly large growth, due to the sparse time and space ~~sparsity of~~ measurements and the crude model~~ approaches~~s, which limited our understanding of the global variation of atmospheric CH_4 (Saunois et al., 2019; Weber et al., 2019). As ~~long as the reasons for rising CH_4 emissions contributed by natural sources (e.g. wetlands), anthropogenic sources (e.g. fossil fuels), or climate change feedbacks remain uncertainties~~a consequence, it will be impractical to predict CH_4 trends in the future, and then to develop realistic management (Nisbet et al., 2019). ~~Therefore, it is essential to establish typical observing regions and perform long observations.~~

Systematic observations are a prerequisite to~~To~~ get an accurate understanding of the spatial and temporal behaviors of atmospheric CH_4~~,~~ concentrations.~~a systematic observations network would perform the best. Hundreds of CH_4 observation stations worldwide are running under the framework of WMO/GAW.~~ Since 1978, systematic measurements of atmospheric CH_4 have been taken~~began~~ around the world (Blake et al., 1982; Rasmussen and Khalil, 1984; Dlugokencky et al., 1994). On the northern slope of the Mauna Loa volcano, Hawaii, there exists the first global station, Mauna Loa (MLO), which h~~w~~as performed at about 3397m above sea level (a.s.l.) and far away from local sources and sinks. It has the longest record~~s~~ of continuous atmospheric CH_4 observation (Keeling et al., 1976). Recently~~Later~~, ~~many~~ other ~~types of the sites~~stations have been ~~were~~ installed for CH_4 observation~~ system~~, such as~~such as~~ the Barrow (BRW),~~,~~ South Pole (SPO) (polar site~~, SPO~~) (Dlugokencky et al., 1995), Cape Grim

(CGO) and Minamitorishima (MNM) ~~in Australia~~ (coastal/island sites) (Pearman and Beardsmore, 1984; Wada et al., 2007)~~, Minamitorishima (MNM) in Japan (coastal/island sites) (Wada et al., 2007)~~, Jungfraujoch (JFJ) ~~in Switzerland~~ and Mount Waliguan ~~in China~~ (continental mountain site) (Zhou et al., 2004; Loov et al., 2008). Hundreds of CH_4 observation stations worldwide are currently running under the framework of the WMO/GAW. ~~Even though, the exact causes of significantly increased CH_4 emissions in past years are still remained unclear and debated, especially for the anomalous periods with suddenly large growth, due to the time and space sparsity of measurements and the crude model approaches, which limited our understanding of the global variation of atmospheric CH_4 (Saunois et al., 2019; Weber et al., 2019). As long as the reasons for rising CH_4 emissions contributed by natural sources (e.g. wetlands), anthropogenic sources (e.g. fossil fuels), or climate change feedbacks remain uncertainties, it will be impractical to predict CH_4 trends in the future, and then to develop realistic management (Nisbet et al., 2019). Therefore, it is essential to establish typical observing regions and perform long observations.~~

China has the largest anthropogenic CH_4 emissions in the world (Janssens-Maenhout et al., 2019). The~~.~~ Qinghai-Tibetan Plateau with~~has~~ an average altitude of over 4000m a.s.l.~~, which~~ has long been recognized as the roof of the world. By coincidence, the two largest CH_4 source regions in the world (~~i.e.~~i.e. Eastern China and Northern India) are trapped by the Tibetan Plateau in between~~the middle~~ (Zhang et al., 2011; Fu et al., 2012; Wilson and Smith, 2015). Under the characteristics of special geographical conditions, lower population density, rare~~ly~~ industrial activities, and high sensitivity to external disturbances, the Tibetan Plateau is ~~undoubtedly~~ one of the ideal regions to observe a continual CH_4 signal (Zhou et al., 2005; Fu et al., 2012; Zhang et al., 2013). Most of the previous studies reported on the short-term CH_4 variations in China and concluded that the ~~importance of~~ long-term observation ~~(Cai et al., 2000; Zou et al., 2005; Wang et al., 2009; Fang et al., 2013)~~~~, which~~ is of great value to enhance the understanding of the global carbon cycle (~~(~~Cai et al., 2000; Zou et al., 2005; Wang et al., 2009; Fang et al., 2013~~)~~; Yuan et al., 2019). As the rapid development of China and India continues, the ~~year to year difference as well as the sources and sinks~~characteristics of CH_4 on the Tibetan Plateau, might change significantly over time. Since 1994, in-situ measurements of atmospheric CH_4 have been launched at the Mt. Waliguan (WLG) station. To study the long-term variations of the atmospheric CH_4 ~~at the WLG~~ and get a new insight ~~of~~ on its characteristics in the inland of ~~the~~ Eurasia, ~~in this study, we evaluated~~ the performance of a ~~24~~ 26-~~year~~ year~~long-term~~ of in situ observations ~~of CH_4~~ at the Mt. Waliguan baseline observatory were evaluated, which is the longest ~~time observing~~ record~~s~~ in China. Temporal patterns, annual variations, long-term trends, air mass transports, and the spatial distribution of potential sources were analyzed. In addition, the case studies combining atmospheric carbon monoxide (CO) measurements and a separate analysis between the Tibetan Plateau and the city regions were performed to constrain the contribution of anthropogenic emissions.

2 Methodology

2. 1 Measurement site

The Mt. Waliguan (WLG, 36.28° N, 100.09° E, 3816m a.s.l.) station is situated at the edge of northeast~~ern of the~~ Tibetan (Qinghai-Xizang) Plateau, which ~~was~~ is in remote western China and isolated from populated and industrial regions (Fig. 1~~Fig. 1~~). WLG ~~was~~ is the only WMO/GAW global background station in Eurasia and is ~~running~~ run by the China Meteorological Administration (CMA). The surrounding ~~areas~~ of the site are pristine with sparse vegetation, naturally arid and semi-arid grasslands. Small farms with yak and sheep are in the valley. Two adjacent large cities Xining (~2.2 million populations) and Lanzhou, are located about 90 km northeast and 260 km east of the station, respectively. The Longyangxia hydroelectric station (~380 km^2) is located approximately 13 km south to southwest of the ~~the~~ WLG. The predominant winds at the WLG are mainly from southwest and east in winter and summer, respectively (Zhou et al., 2004; Zhang et al., 2011), which is controlled by Tibetan Plateau monsoon. Simultaneously, diurnal~~al~~ variations of vertical winds at the WLG ~~is~~ are influenced by mountain-valley breezes, where upslope flow brings heated air masses from the boundary layer to the site in daytime and downslope flow results in cool air masses transport from the mountain peak to the site. Under this unique location, the observation at the WLG ~~could~~ can obtain essential information on CH_4 sources and sinks from Eurasia (Zhou et al., 2005; Zhang et al., 2013).

2. 2 **Instrumental setup**

Atmospheric CH_4 has been measured quasi-continuously using a HP 5890 gas chromatograph (GC) equipped with a flame ionization detector (FID) since July 1994, and an Agilent 6890N GC equipped with a FID since June 2008. Both of the systems used the same sampling procedures. A Cavity Ring Down Spectroscopy system (Picarro G1301) began in January 2009 and the instrument was upgraded to Picarro G2401 in 2015. Ambient air is delivered to the above systems at about 5 L/min by a KNF Neuberger N2202 vacuum pump via a dedicated 0.95 cm o.d. sample line from an 80m intake line attached to an 89m steel triangular tower located approximately 15m from the main observatory. The residence time of the ambient air from the top of the tower to the instrument is 30 s. The ambient air is first passed through a 7 mm stainless steel membrane filter located upstream of the pump and then (after the pump) passed through a pressure relief valve set at 1 atm to release excess air and pressure. The ambient air is then dried to a dew point of approximately -60°C by passing it through a glass trap submerged in a -70°C methanol bath. All standard gases supplied to the instruments are from pressurized 37.5 L treated aluminum alloy cylinders fitted with high-purity, two-stage gas regulators. Stainless steel tubing (0.32 cm o.d., 0.22 cm i.d.) is used for the standard gas sample line and the ambient sample line after the cold trap. ~~The~~ An automated sampling module equipped with a VICI 8 ports valve is designed to sample from separate gas streams (standard tanks and ambient air). According to the comparability target of WMO/GAW program (WMO, 2019), methane mole fractions are referenced to a Working High standard (WH) and a Working Low standard (WL). Additionally, a calibrated cylinder filled with compressed ambient air is used as a Target gas (T) to check the precision and stability of the system routinely. Diagram of the observing system during different periods could be seen at Zhou et al. (2004) and Fang et al. (2013). Here, we focus on the longest continuous measurements of CH_4 from August 1994 to ~~May~~ December 2019~~7~~ at WLG. Data gaps in limited periods are because of the malfunction of instrument and the maintenance of the sampling system.

The records of CO in this study was initinially observed by an RGA-3 gas chromatograph (GC) equiped with an HgO reduction detector (Trace Analytical Inc.) since 1994. An automated sampling module was designed to sample from ambient air and a series 9 standards. Deailed diagram of the system was described by Zhang et al. (2011). Since 2010, the CO has been measured by the Cavity Ring Down Spectroscopy instrument (Picarro G1302 and G2401 since 2015). The scale for all of the CO measurement were further updated to WMO X2014A.

2. 3 **Data processing**

Most on-site CH_4 observations were unavoidably influenced by local sources and other complex conditions (~~e.g.,~~e.g. traffic transportation, various topography). As a result, the records cannot fully represent the regional atmospheric CH_4 in well-mixed conditions~~s~~ (Liu et al., 2019). To ~~precisely~~ get regionally representative ~~regional~~ records, we excluded the CH_4 ~~measurements~~ data influenced by local sources adjacent to the site (~~e.g.,~~e.g. agricultural fields, cities, traffic emissions). The hourly CH_4 data were classified as Locally/Regionally representative~~events~~ through the meteorological approach, which was based on essential meteorological information, similar to previous studies by Zhou et al. (2004) and Liu et al. (2019). In this study, the CH_4 records associated with local surface winds from selected sectors, ~~(i.e.~~i.e. NNE-…-ENE in spring, NE-…-SE in summer, NE-…-ESE in autumn, and NE-ENE in winter,~~)~~ were flagged as locally ~~representative~~influenced (27.0%). Subsequently, we ~~further~~ rejected portion of daytime records to minimize the effect of human activities (16.9%), ~~e.g.,~~e.g. ~~rush hours), including~~ 9:00-13:00 LT (local time) in ~~spring and~~ summer, ~~9:00-14:00 LT in autumn,~~ and 10:00-17:00 LT in winter. Finally, we filtered CH_4 data into locally ~~events~~ influenced when the surface wind speeds were~~as~~ less than 1.5 m s^{-1} to minimize the very local accumulation (9.2%).

In order to ~~further~~ investigate the influence of anthropogenic emissions from cities and remote area as Tibetan plateau, ~~investigate the characteristics of atmospheric CH_4,~~ we divided the CH_4 ~~observations~~data into two main influencing regions according to the ~~above~~ analysis, including the geographical conditions, the effect of surface winds, the long-range transports, and the potential source distributions. The first region ~~was~~ cover~~ed~~s the northeast and~~to~~ southeast (NNE-…SE) of the WLG, which ~~wa~~is denoted as City Regions (CR). The second region ~~wa~~is located the south to west (S-…-W) of the station and ~~wa~~is well known Tibetan (Qinghai-Xizang) Plateau (TP) (Fig. S~~2~~1). Accordingly, the hourly CH_4 records when the surface winds coming from these sectors were divided into two subsets (~~i.e.~~i.e. TP and CR). The long-term variations between the two regions as well as the total regional time series ~~(Total)~~ were further compared and analyzed ~~to explore new sight of atmospheric CH_4 variation at WLG.~~

To understand the influence of local surface wind, ~~the~~ hourly CH_4 data ~~was~~ was ~~calculated~~ binned ~~versus~~ into 16 horizontal wind direction classes (Fang et al., 2013). In this study, we used the ‘polarPlot’ function ~~located~~ in the ‘openair’ package of the statistical software R (R Core Team, 2020). It shows the bivariate (~~i.e.~~i.e. wind speed and wind direction) polar plot of CH_4 concentrations, and the concentrations are calculated as a continuous surface by modelling using smoothing techniques (Carslaw et al., 2006; Diederich, 2007). Also, conditional probability function (CPF) was used to ~~detect~~ investigate the probability of which wind directions are dominated by high CH_4 mole fractions (Uria-Tellaetxe and Carslaw, 2014). In order to study the pollution transport pathways of air masses at WLG, the cluster analysis of 3 days back trajectories was applied using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) dispersion model (Draxier and Hess, 1998; Rousseau et al., 2004) on the strength of gridded meteorological data (2004-2017) from the National Oceanographic and Atmospheric Administration's Air Resources Laboratory (NOAA-ARL). The spatial resolution of the model is 0.5×0.5 degree and the model height is 10 km a.s.l. We computed the back trajectories coincident with hourly CH_4 mole fractions. The trajectories ~~in four months, including~~for January, April, July and October, were calculated to represent the seasons of winter, spring, summer and autumn, respectively. The spatial source distributions of annual CH_4 were analyzed using the Potential Source Contribution Function (PSCF) approach, which computed the conditional probability of the residence times of air parcels with greater concentration than threshold transport to the exact~~ly~~ receptor site (Ashbaugh et al., 1985). In this study, PSCF value was calculated in 0.5×0.5-degree grid cell (i, j):

$$\mathrm{PSCF}_{ij} = m_{ij} / n_{ij} \qquad (1)$$

n_{ij} represents the number of endpoints that terminate in the ijth grid cell, while the number of trajectories with concentration exceed the threshold value was defined as m_{ij} (Polissar et al., 1999). In order to reduce the abnormal influence of small n_{ij} values in some grid cells, $PSCF_{ij}$ was further computed by an arbitrary weighting function W_{ij} as below.

$$W_{ij} = \begin{cases} 1.00 & 3n_{ave} < n_{ij} \\ 0.70 & 1.5n_{ave} < n_{ij} \le 3n_{ave} \\ 0.42 & n_{ave} < n_{ij} \le 1.5n_{ave} \\ 0.05 & n_{ij} \le n_{ave} \end{cases} \qquad (2)$$

W_{ij} represents the weight of cell (i, j), n_{ij} is the number of trajectory endpoints that fall in the ijth grid cell, while the n_{ave} shows the mean number of the endpoints in all grid cells.

In order to fill the data gaps so as to evaluate the long-term CH_4 trend, we applied the curve fitting approach by Thoning et al. (1989). We also calculated the trend curve that excluded the influence of seasonal variation, and then got the annual growth rates ~~of~~ by the average of the first derivative of the trend curve. The function consists of the polynomial part and the annual harmonics part:

$$f(t) = a_0 + a_1 t + a_2 t^2 + \cdots + a_{(k-1)} t^{(k-1)} + \sum_{n=1}^{nh} c_n [\sin(2n\,\pi\,t) + \varphi_n] \quad (3)$$

'k' represents the number of polynomial part. 'nh' is the number of harmonics part. We applied k = 3 polynomial terms (a quadratic) for multi-year trends and nh = 4 yearly harmonics for seasonal cycles in this study. The fast Fourier transform (FFT) was utilized to smooth the fitting residuals (Press et al., 1992).

The significant difference test was applied by the 'scheirerRayHare' function in the 'rcompanion' package of R software, which is a non-parametric test for two-way ANOVA analysis. ~~And t~~The multiple comparison was used by Wilcoxon rank sum test with R (R Core Team, 2020). For the correlation analysis between CH_4 and CO, we obtained the detrended time series of CH_4 and CO from 2004-2017 based on the method by Thoning et al. (1989). The detrended values are denoted as ΔCH_4 and ΔCO, which~~.~~ are the original data points minus the trend curve. ~~The detrended values are denoted as ΔCH_4 and ΔCO.~~ To accurately obtain the correlation slopes of ΔCO ~~–ΔCH_4~~ and ΔCH_4 ~~ΔCO~~, i.e. $\Delta CO/\Delta CH_4$ ~~accurately~~, a rolling linear regression was applied to the ΔCH_4 and ΔCO time series by the 'roll_lm' function in 'roll' package of R (R Core Team, 2020). We successively moved a 24-h time window by 1 h over the whole time series. Similar to the study by Tohjima et al. (2014), we set three~~3~~ criteria to achieve a better quality control of the slopes. When (i) the number of CH_4 record is less than five~~5~~ in 24 hours, (ii) the coefficient variation of the correlation slope is more than 15%, (iii) the absolute value of the correlation slope is less than 0.8 ($|R| < 0.8$), the correlation slopes were identified as statistically insignificant and inaccurately and were rejected. In order to understand the year to year variations, we ~~further~~ analyzed the CH_4 variation over different periods ~~over~~ in 1994-2019~~7~~. The entire CH_4 time series were divided into five observing periods, i.e. 1994-1997, 1998-2002, 2003-2007, 2008-2012, and 2013-2019, according to the significant stages or the critical time period of atmospheric CH_4

variations from –previous~~ly~~ studies (Zhou et al., 2004; Fang et al., 2013; Zhang et al., 2013; Nisbet et al., 2019; WMO, 2020)~~, appropriately every five years a period~~. Unless special notes, the average values in this study ~~were~~ are presented with 95% confidence intervals (CIs).

3 Results and discussion

3. 1 Extracting the regional atmospheric methane

To precisely understand the characteristics of atmospheric CH_4, including seasonal cycles and long-term trends, it is vital to identify the CH_4 records in well-mixed air without local contaminations (Liu et al., 2019). In this study, hourly CH_4 measurements between 1994 and 2019 were analyzed, resulting in 64.0% of the CH_4 data being classified as regionally representative, with an average CH_4 mole fraction of 1865.8 ± 0.4 ppb. The average of the locally influenced data (1868.2 ± 0.3 ppb) was larger than that of the regionally representative records (Table 1). The filtered regional/local time series was shown in Figure 2. It can be seen that the CH_4 mole fractions increased from 1994 to 2019. The atmospheric CH_4 showed a strong growth and displayed large fluctuation. In 1994, the average CH_4 mole fraction was only 1799.0 ± 0.4 ppb, however, the average value increased 133 ppb by the year 2019 (1932.0 ± 0.1 ppb) (Table 2). Compared with the global average mole fractions in recent years, i.e. 1853 ± 2 ppb in 2016, 1859 ± 2 ppb in 2017, and 1869 ± 2 ppb in 2018 (WMO, 2019; 2020), the atmospheric CH_4 mole fractions at the WLG were significantly higher. These results indicated that the WLG was affected by strong CH_4 sources in recent years, which were possibly due to the influence of the two largest source regions of Northern India and Eastern China (Fang et al., 2013; Zhou et al., 2004).

~~2. 4~~3. 2 Diurnal variations

~~Generally, d~~Distinct diurnal cycles were observed in four seasons during 1994-

20172019 at the WLG. The CH_4 mole fraction increased from early morning, and reached athe maximum at noon, and had a trough in the late afternoon (Fig. 3Fig. 2f). However, differences also existed infrom different four seasons. In spring and summer, the atmospheric CH_4 apparently increased from 9:00 to 13:00 LT at noon, with athe daily amplitude of 5.87 ±2.84 and 4.4 ± 3.4 ppb, respectively ppb(Table S1). In summer, the elevated CH_4 also appeared during 9:00-13:00 LT at noon, and the daily amplitude was 4.3 ± 2.6 ppb. In autumn, the diurnal variation showed anthe mean amplitude of 4.35 ± 32.14 ppb, significantly elevated CH_4 reached at 9:00-13:00 LT with one peak at noon. In winter, a largely increase of CH_4sing prwas found duringesented in the daytime at 9:00-17:00 LT, with the largest largest peak to trough amplitude of 7.1 ± 2.9 ppb of 6.2 ± 2.4 ppb among four seasons. For the diurnal variation over the whole monitoring period, the highest CH_4 mole fraction was observed in winter and the minimum value was found in spring (Fig. 2f).

Different patterns for diurnal CH_4 cycles were also found over different periods. In 1994-1997 and 1998-2002, the CH_4 mole fractions in the winter were apparently higher than the other seasons (Fig. 3Fig. 2a-b), which .was likely due to the manner of heating (e.g. large biomass burning) as well as the adverse diffusion conditions in cold weather (Fang et al., 2013). However,But its value was the highest value was found in summer during the periods of 2003-2007, 2008-2012, and 2013-20172019 (Fig. 3Fig. 2c-e),. which could be ascribed to the transport of anthropogenic emissions (e.g. southeast cities) by the meteorological factors (Zhang et al., 2013). Additionally, in summer, the intense herding or grazing activities around the WLG have enhanced the regional CH_4 emissions and hence contributed to the higher CH_4 mole fractions (Zhou et al., 2004).The atmospheric CH_4 values in winter were gradually falling behind the other seasons, and the gaps among different seasons were increasing, especially for summer.

Similar to the previous studies by Zhang et al. (2013) and Fang et al. (2013), Before 2002, diurnal cycles in four seasons were ambiguous before 2002 (Fig. 3Fig. 2a-b), but significant diurnal variations appeared afterwards (Fig. 3Fig. 2c-e)., which indicated that the local sources were weak at the WLG in the past. The apparent diurnal cycles after 2002 may be attributed to the intense activities by humans (e.g. grazing, burning fuel), which were enhanced in the daytime and weakened at nighttime (Fang et al., 2013). The peak to trough amplitude almost increased along with the time in almost all seasons. For example, in spring, the amplitude was 6.5 ± 3.01, 4.7 ± 2.11.8, 5.6 ± 2.76, 76.02 ± 32.1,4 and 6.98 ± 3.14 ppb over the periods of 1994-1997, 1998-2002, 2003-2007, 2008-2012, and 2013-20172019, respectively (Table S1Table S1). The meteorological conditions (e.g. diffusion and transport) could have also contributed to the increasing CH_4 amplitude. The WLG was remote from the populated center, therefore, the good diffusion conditions in the daytime may have brought high anthropogenic emitted CH_4 to the site. The increasing amplitude and CH_4 mole fractions suggested that the WLG was increasingly affected by local and regional anthropogenic sources, such as gas exploitation, and grazing (Zhou et al., 2004).

2. 53. 3 The impact of local surface winds

As observed bySimilar to the previous short-term studyvariations by Zhou et al. (2004), the atmospheric CH_4 at the WLG was significantly influenced by local surface winds from the northeast andto southeast sectors (Fig. 4Fig. 3f). Slight differences were also found among seasons. In the spring, when the wind was originating from the NNE-NE-ENE-E sectors, the atmospheric CH_4 was enhanced by 2.5-6.5 ppb as compared to the seasonal average (1839.7 ± 1.4 ppb) when the wind was originating from NNE-NE-ENE-E sectors. In the summer and autumn, the wind from the NNE-NE-ENE-E-ESE proinduced higher CH_4 mole fractions, with an enhancement of about 3-9.5 ppb and 4 to -18 ppb, respectively. In – winter, similar to that in spring, the CH_4 mole fractions significantly increaselevated from the from NNE-…-E sectorswind sectors that same as those found in spring, with a value of 7-21 ppb higher than the seasonal average (1854.5 ± 4.8 ppb). In summer, the prevailing winds were from the NE-…-ESE sectors

(~46%) (Fig. S2), and the CH_4 mole fractions were also higher in the related sectors. However, in autumn and winter, although the prevailing wind and the high wind speed were from SSW-…-W sectors (~ 40-50%) (Fig. S2), the high CH_4 mole fractions were from the opposite wind sectors of NNE-….-ESE (Fig. 4f), suggesting that strong local sources were distributed from northeast to southeast of the WLG (city regions), and even covered the emission of natural sources. Causes of the elevated CH_4 from these sectors could be attributed to the large plantations of highland barley as well as the high population density in those areas (Fang et al., 2013). The two largest cities of Xining (with population of ~2.2 million) and Lanzhou (with population of ~4 million) are also situated in the northeast and east of the WLG, respectively. The heavy human activities from anthropogenic fossil combustion, landfills, and livestock, could have also emitted large amounts of CH_4. Based on the data from the Emissions Database for Global Atmospheric Research (EDGAR), the increase of CH_4 emission was 500 kg yr^{-1} in these two regions throughout 1994-2015 (Crippa et al., 2019). Also, the Yellow River Canyon industrial area (YRC), which is ~500 km northeast of the WLG, may also have contributed to the high CH_4 values (Zhou et al., 2003). With the rapid development of land-use, water utilization, and agriculture sources in the YRC, large CH_4 emissions could have easily transported to the WLG. Previous studies on black carbon (BC) and carbon monoxide (CO) also revealed that the high CH_4 values at the WLG in winter were a result of transport from the YRC (Tang et al., 1999; Zhou et al., 2003).

The wind-rose distribution of CH_4 mole fractions indicated that the ~~Relatively, the amplitude of enhancements in winter and autumn were larger than those in spring and summer.~~

~~What interesting is that wind sectors~~ elevat~~ing~~ed CH_4 mole fractions var~~y~~ied in ~~different periods. The~~ early periods (~~i.e.~~i.e. 1994-1997 and 1998-2002) to~~were different from the~~ recent periods (2003-2007, 2008-2012, and 2013-2017). The elevated CH_4 was predominately from the~~about~~ ENE-~~E-ESE-SE.~~..-SSE sectors in the early years (Fig. 4~~Fig. 3~~a-b)~~,~~ but evolved to the NNE-NE-ENE-E sectors in later years (Fig. 4~~Fig. 3~~c-e).

Furthermore, the amplitude of enhancements was almost also increasing continuously along with the progression of the time. For example, in autumn, the maximum CH_4 mole fractionss were from the E in 1994-1997, ENE in 1998-2002, ENE again in 2003-2007, NE in 2008-2012, and finally ENE insectors in 1994-1997, 1998-2002, 2003-2007, 2008-2012 and 2013-2017, with the successively increasesing enhancements of 8.6, 12.1, 14.7, 16.8, and 19.7 ppb, respectively. Therefore, the local surface wind from the city regions had an increasing effect on the atmospheric CH_4 at the WLG.

We The CPF was applied the CPF to hourly CH_4 and CO data by considering intervals of entire data percentiles including 0-20th, 20th-40th, 40th-60th, 60th-80th, and 80th-100th 0-20, 20-40, 40-60, 60-80 and 80-100 to draw the CPF polar plot (Uria-Tellaetxe and Carslaw, 2014). It wais clear that the different sources only affected the CH_4 mole fractions ion different percentile ranges (Fig. 5), meaning that the. specific sources were prominent for specific percentile ranges. For example, for most wind speed-directions, the CPF probability of the CH_4 being greater than the 60^{thth} percentile was trending to zero (Fig. 4). And it is apparent that , and most sources contributed to the less than 60th percentiles of less than the 60th for the CH_4 mole fraction (e.g.e.g. 40th-60th 40-60) (Fig. 4). These results implied that most areas around the WLG had a small contribution to the CH_4 emissions. In addition, . The specific sources were prominent for specific percentile ranges. Tthe wind from the southwest and the southeast was important foron the cases of the the higher percentiles, resultinged in the highest CH_4 mole fractions of 1849-1872 ppb for the 60th-80th 60-80 percentiles and 1872-2031 ppb for 80-100the 80th-100th percentiles (Fig. 5Fig. 4), which revealed the existence of a southeastern and a southwestern strong source region. The anthropogenic emissions from cities (e.g. Lanzhou or Chengdu) were the only cause for high values in the southeast, and the southwest region that was farther away from the WLG was possibly due to sources from other countries, such as India.. IIt's's more obviousobvious that the CO sources the CO showed gradually shifted sources with the increase of the percentile ranges (Fig. 5). The areas where the CPF probabilities were higher is were to the NW-SW sectors when the percentages ranged from 0 to 40^{th}. Nevertheless, when the percentages were larger thandata exceeded the 60^{th} 60percentileth, the high probability areas completely moved to NE-SE sectors (Fig. 4). Unlike that of CH_4, the high CO mole fractions were consistent from the east regions (urbanized areas) (Fig. 5), indicating strong anthropogenic sources in the city regions (e.g. Xining and Lanzhou) and different source distributions between CH_4 and CO around the WLG (Zhang et al., 2011).

2. 63. 4 **Air mass pathways andLong-range transport and potential source distributions**

2. 6. 13. 4. 1 **Air mass transports**

Figure 5 6 illustrates the cluster analysis tof the 3-day back trajectories to WLG betweenduring 2004 and -2017. In the spring, the majority of the air masses were from the west and northwest regions, which accounted for about 24% (cluster 3) and 44% (cluster 5) of the total trajectories (Fig. 6Fig. 5a). These air masses were also accompanied with by higher CH_4 mole fractions than those from the east and northeast regions, such asi.e. cluster 1 (13.3% of total) and cluster 4 (11.69%) (Table 3Table 1). The largest enhancement was in cluster 3 at ~18 ppb (relative to the spring average) by cluster 3. In the summer, 45% of the air masses (cluster 1) were from eastern regions (cluster 1). However,But the highest CH_4 mole fractions were ion cluster 2 and cluster 5 from northwest and west regions, respectively., However,although low percentages were found in each of those clusters, i.e. 26% in were found (cluster 2 and 7%: 26%; in cluster 5: 7%) (Fig. 6Fig. 5b). The highest CH_4 mole fraction was associated with cluster 2, with ~9 ppb larger than the average in summer. In the autumn, a large proportion of air masses cluster 2 (49%) and cluster 3 (32%) was originatinged from the west and southwest stations, such asrespectively cluster 2 (49%) and cluster 3 (32%) (Fig. 6Fig. 5c). CThe highest CH_4 was from cluster 3 showed the highest CH_4 mole fraction, with anwith enhancement of ~4 ppb relative tothan the seasonal average. Similar to autumnIn winter, the air masses were primarily came from northwest and southwest regions in winter, including e.g. northwest cluster 3 (59%), and southwest cluster 1 (34%) regions (Fig. 6Fig. 5d). and tThe cluster 1 brought the highest CH_4 mole fractions was on cluster 1with anthe enhancement of ~7 ppb overthan the average valueseasonal average.

The air masses from east and northeast regions passed over the cities of Xining and Lanzhou (capital of Gansu province), which are the populated centers and industrial areas (Fig. 6). However, the higher CH_4 mole fractions were not observed when the air masses were from these sectors, the high values were frequently brought by air masses that came from the northwest to southwest (Table 3). The higher values seen in the northwest to southwest airflow were because the air masses had passed through the northwest of the Qinghai province and the central area of the Xinjiang Uygur Autonomous Region (XUAR). This is where the Ge'ermu urban area (the second largest city of Qinghai) was located and where there was with rapid industrial development, natural gas and petroleum resource exploitation, and residue burning of large crops, hence the CH_4 emissions were strong (Fang et al., 2013; Zhang et al., 2013). This result was also similar to the CPF percentile analysis (Fig. 5), as time went by, the southwest or northwest regions that were farther away from the site became the strongest source regions to the WLG.

2. 6. 23. 4. 2 Spatial distribution of potential source regions

In this study, tThe potential sources were analyzed over different periods, i.e.i.e. 2004-2007, 2008-2012, and 2013-2017 (Fig. 7). Generally, the strongest potential sources were located at the northeast orto southeast of the station, especially in summer,; but a large area of sources was identified from the southwest to the northwest regions, which also contained CH_4 emissions from the northwest Gansu province, the northwest

Qinghai province and the southeast of the XUAR. Additionally, t~~t~~he source regions differed in various seasons~~ as well as years (Fig. 6)~~. The~~ regions of~~ potential source regions in the spring (Fig. 7~~Fig. 6~~a-c) and winter (Fig. 7~~Fig. 6~~j-l) were~~as obviously~~ larger than those~~at~~ in the summer (Fig. 7~~Fig. 6~~d-f) and autumn (Fig. 7~~Fig. 6~~g-i). The seasonal difference~~. ~~ was due to the effect of westerlies or the southeast monsoons (Zhou et al., 2004).

There were also trends in~~for~~ the CH_4 source regions correlated~~along~~ with specific years: i) the area of the potential source regions was increasing ~~with the years~~over time, and ii) the location of strong ~~potential ~~sources changed along with the time. For example, in autumn and winter, the ~~strength of ~~CH_4 sources ~~were ~~were very strong in the southeast and~~to~~ northeast regions during 2004-2007 (Fig. 7~~Fig. 6~~g & i), and then weakened in 2008-2012 (Fig. 7~~Fig. 6~~h & k). Finally,~~,~~ from~~ and finally (i.e.~~ 2013-2017 the sources ~~),~~almost vanished in the eastern regions but moved to the southwest with a very large distribution area (Fig. 7~~Fig. 6~~i & l).

More CH_4 sources appeared at the WLG along with the progression of time, which could have been attributed to the influence of human expansion. The pattern of strong sources moving indicated that the southwest area relative to the WLG, e.g. Northern India, was gradually becoming a strong CH_4 contributor. India with abundant cattle as well as an extensive large-scale coal mining operation possibly contributed large amounts of CH_4 to move from northern India to the northeastern Tibetan Plateau (Fig. 7i & l) (Fu et al, 2012). The analysis of air mass transport (Fig. 6d) also supported the conclusions that the air masses from the southwest regions contributed the highest CH_4 mole fractions. The studies of atmospheric Hg at the WLG by Fu et al. (2012) also supported this phenomenon, which found the long-range transport of atmospheric Hg from India to the Northeastern Tibetan Plateau.

~~2.7 Extracting the well-mixed ambient methane~~

~~To precisely understand characteristics of atmospheric CH_4, e.g., seasonal cycle or~~

~~long-term trend, it is vital to identify the CH_4 records that were influenced by local sources and sinks. In this study, we analyzed hourly CH_4 measurements during 1994-2017, and 47.3% of CH_4 data were classified as regional representative, with the average CH_4 mole fraction of 1847.9 ± 0.3 ppb. The local representative data was obviously larger than regional events, with an average value of 1858.2 ± 0.4 ppb (Table 2). The proportion of regional events increased slightly before 2012, but significantly reduced in recent years (e.g., 2013-2017). The filtered regional/local time series was shown in Figure 7. It can be seen that the CH_4 mole fractions obviously increased from 1994 to 2017. The atmospheric CH_4 showed strong growth and displayed large fluctuation at WLG (Fig. 7). In 1995, the average CH_4 mole fraction was only 1805.8 ± 0.1 ppb, however, the average value increased 98 ppb by the year of 2016 (1903.8 ± 0.1 ppb) (Table 3).~~

~~2. 8~~3. 5 **Correlation analysis between CH_4 and CO**

Because parts of the CH_4 and CO in~~ ~~ atmosphere ~~are~~ were from the same anthropogenic sources (~~e.g.,~~e.g. fossil fuel combustion), the long-term trend of $\Delta CO/\Delta CH_4$ was helpful to understand the variation in the sources/sinks in many studies (Buchholz et al., 2016; Niwa et al., 2014; Tohjima et al., 2014; Wada et al., 2011). In this study, the hourly CO data from 2004 to 2017 was used to further analyze the long-term variations of CH_4 (Fig. S3). T~~we calculated t~~he regression slopes of $\Delta CO/\Delta CH_4$ from 2004 to 2017 were analyzed (Fig. S4~~Fig. S1~~). Figure ~~8~~ 8 presents the ~~average~~ seasonal cycles of the $\Delta CO/\Delta CH_4$ slopes. Generally, the slopes were larger ~~in~~in summer and small~~low~~er in winter during the observing period, except for 2004-2007 where~~with~~ the highest slope was in autumn~~.~~. This was primarily due to the effect of monsoons and air mass transport. Tohjima et al. (2014) found an opposite variation at the Hateruma Island, which showed small slope values in the summer. Wada et al. (2011) analyzed more than 10-year seasonal variation of the $\Delta CO/\Delta CH_4$ ratios at three monitoring stations, i.e. MNM, Yonagunijima (YON), and Ryori (RYO) in Japan, which also showed an opposite trend to that of the WLG. It was because these sites were considerably affected by the Asian continental source regions, where had enhanced emissions of CH_4 in the summer (rice paddies) and CO in the winter (fuels combustion). At the WLG, Regional polarization in the concentration ratios of CH_4 and CO were seen (Fig. S5), which implied different strong source distributions between CH_4 and CO. This result was also seen in the CPF analysis (Fig. 5). Furthermore, the cluster results (Fig. 6b & d) and the PSCF analysis (Fig. 7f & l) also supported this conclusion. In the summer, the source emissions were mainly from the east-southeast regions (cities) with large amounts of CO but relatively lower CH_4 (Table 3), leading to the largest $\Delta CO/\Delta CH_4$. In contrast, the sources mostly from southwest-west regions (Tibetan Plateau) emitted large amounts of CH_4 but with relatively lower CO in the winter. Hence, the opposite two air mass transports and source emissions led to a peak in $\Delta CO/\Delta CH_4$ in the summer and a trough in the winter (Fig. 8). These results revealed different local sources/sinks, the special topography conditions and source distributions around the WLG.

~~Additionally, the regression slopes increased along with the time, which showed the maximum in 2013-2017 and the minimum in 2004-2007. For the year to year variations, t~~The $\Delta CO/\Delta CH_4$ slopes showed large fluctuations from 2004 to 2017 at the WLG (Fig. 9~~Fig. 9~~). The slopes showed a decreasing trend during 2004-2007 but then increased from 2007-~~ to ~~2010, and ~~again ~~decreased again after 2010. ~~In spring and summer, increasing trends appeared again after 2014. The slopes in summer were almost the largest but the lowest in winter.~~

In 2007, a large increase in ΔCH_4 appeared, and from 2010 to 2013, the ΔCO decreased significantly (Fig. S4). Before 2010, large air masses and potential source regions were identified in eastern regions (cities) with the highest CO emission (Fig. 7 and Table 3). After 2010, the southwest regions had the highest CH_4 emissions but relatively low CO emissions. Therefore, the strong seasonal variation of $\Delta CO/\Delta CH_4$ also revealed that the WLG was affected by different anthropogenic sources, e.g.

sources from cities, and sources from the Tibetan Plateau, during a year, especially in the summer and winter. The long-term trend of the slopes implied that the source emission types (CO sources or CH_4 sources) around the WLG might have been changing with human activities, like straw burning, in the early years, or coal mining in recent years.

~~2. 9~~3. 6 Variation of long-term records

~~2. 9. 1~~3. 6. 1 Seasonal cycles

~~In order to further investigate the characteristics of atmospheric CH_4, we divided the CH_4 observations into two main regions according to the above analysis, including geographical conditions, the effect of surface winds, the long-range transports and the potential source distributions. The first region was covered the northeast to southeast (NNE-…SE) of WLG, which was denoted as City Regions (CR). The second region was located south to west (S-…-W) of the station and was well known Tibet (Qinghai-Xizang) Plateau (TP) (Fig. S2). Accordingly, the hourly CH_4 records when the surface wind coming from these sectors were divided into two subsets (i.e. TP and CR). The long-term variations between the two regions as well as the total regional time series (Total) were compared and analyzed to explore new sight of atmospheric CH_4 variation at WLG.~~

Overall, ~~at WLG,~~ the seasonal averages of regionally representative CH_4 at the WLG ~~CH_4~~ were almost in a cycle starting with~~ordered by~~ summer (~~1850~~1861.7~~.0~~ ± 0.4~~3~~ ppb), then winter (1856~~47~~.7~~4~~ ± 0.3 ppb), autumn (1855~~44~~.9~~4~~ ± 0.4~~3~~ ppb), and spring (1853~~41~~.1~~2~~ ± 0.3 ppb), except during 1994-1997 where there was a~~with the~~ maximum in the winter and a minimum in the autumn (Fig. 10~~Fig. 10~~). Seasonal averages in the CR were slightly different from that in the TP and even the total regional data. The seasonal averages in the TP were mostly higher than that in the CR from 1994 to 2019, except for in the winter (Table S2). The maximum mole fractions were mostly found in August and the minimum mole fraction appeared in April for the total regional time series, with a seasonal amplitude of 14.4 ppb. The peak to trough amplitude in the CR (~16.7 ppb) was higher than that in the TP (~15.1 ppb) during 1994-2019. The seasonal variations were consistent with the previous short-term studies from 2002-2006 at WLG (Zhang et al., 2013). However, these variations were almost opposite to observations at the adjacent stations of Lin'an, Shangdianzi, and Longfengshan in China (Fang et al., 2013, 2016). For other regional sites in the Asia, Guha et al. (2018) studied seasonal variability at the Sinhagad (SNG) and Cape Rama station (CRI) over India, which also showed an opposite trend to the WLG due to the strong impact of monsoon dynamics. Ahmed et al. (2015) found that the seasonal CH_4 showed a maximum in the winter and a minimum in the spring at two urban sites of Guro (GR) and Nowon (NW), in Seoul, Korea over 2004-2013. Kim et al. (2015) investigated the decadal variation (1991-2013) of CH_4 at the East Asian sites, e.g. Ulaan Uul (UUM) in Mongolia and Tae-ahn Peninsula (TAP) in Korea, which revealed again an opposite seasonal trend to that of the WLG.

The data were further compared with similar WMO/GAW global stations in the Northern Hemisphere, including MLO (19.54° N, -155.58° E, 3397m a.s.l.) (Dlugokencky et al., 1995, 2019a), JFJ (46.55° N, 7.99° E, 3580m a.s.l.) (Zellweger et al., 2016), MNM (24.29° N, 153.98° E, 7.1m a.s.l.) (Matsueda et al., 2004; Tsutsumi et al., 2006), as well as the marine boundary layer (MBL) from the NOAA/ESRL lab at a similar latitude (Dlugokencky et al., 2019b). It was seen that the stations in the Northern Hemisphere and the MBL showed an opposite trend to the WLG with the minimum being in summer and maximum in the winter or spring (Fig. 11). The peak in the summer at the WLG was attributed to larger grazing, human activities, ruminants, and easterly winds coming from neighboring areas over other seasons. The CH_4 emissions from yaks and other ruminants in the Tibetan Plateau (alpine pasture) were very strong in the summer, preceded only by paddy emissions (Fang et al., 2013; Zhang et al., 2013). Furthermore, the dynamic transport by airflow from the polluted northeast/southeast region was also strong in summer, which all induced high CH_4 mole fractions and consequently an opposite trend than other sites (Ma et al., 2002; Xiong et al., 2009).

The seasonal amplitude at the WLG (~14 ppb) was significantly lower than many other sites in the Northern Hemisphere, by about 35-70 ppb. Such sites included MLO in America, BRW in North Pole, UUM in Mongolia, TAP in Korea, Ny-Ålesund in Norway, Bialystok in Poland, Ochsenkopf in Germany, and Beromunster in Switzerland (Dlugokencky et al., 1995; Kim et al., 2015; Morimoto et al., 2017; Thompson et al., 2009; Popa et al., 2010; Satar et al., 2016). MBL also showed a larger amplitude than WLG (Fig. 11). The study at the SNG and CRI over India showed a much larger amplitude close to 200 ppb (Guha et al., 2018). The low amplitude at the WLG was because of the high elevation of the continental mountain sites, where there were relatively fewer effects from local influences than the coastal/island sites (Yuan et al., 2019). Additionally, the ~~Seasonal averages in CR were significantly different to that in TP and also the entire regional data (Total). The seasonal average in TP was mostly higher than that in CR, except for wintertime (Table S2). The atmospheric CH_4 in August was mostly the maximum and the April was the minimum for the total regional time series (Total), with the seasonal amplitude of 13.4 ppb. The peak to trough amplitude in CR (~15 ppb) was higher than that in TP (~13 ppb) during 1994-2016. Additionally, s~~seasonal amplitudes indicated different trends between the CR and TP. For the CR, the seasonal amplitude decreased at~~was~~ first~~ly~~ ~~–dropped~~ and then increased over~~along with~~ time, which ~~were~~ was similar to the variation of the total regional events~~ (Total)~~. But for the TP, the amplitude ha~~displaye~~d a continuously increasing trend, with values of~~ about~~ 15.1~~9~~, 18~~9~~.1~~3~~, 21.4~~6~~, 20~~3~~.4, and 23~~2~~.7~~4~~ ppb in 1994-1997, 1998-2002, 2003-2007, 2008-2012, and 2013-~~2016~~2019, respectively~~.~~

. This revealed that the Tibetan Plateau was intensively affected by strong regional sources (e.g. grazing or emissions from India) over time.

~~2. 9. 2~~3. 6. 2 Long-term trend

The fluctuating trend in atmospheric CH_4 during 1994-2019 at the WLG (Fig. 12) was similar to the global trend reported by many studies (Bergamaschi et al., 2013; Rigby et al., 2017; Nisbet et al., 2019). In the 1990s, the CH_4 growth rates were very ~~low~~ small ~~and~~ or even negative at WLG. Subsequently, ~~during 2002-2006,~~ a steady period ~~was found~~ with ~~a~~ near-zero growth rates was found during 2002-2006. However, ~~After 2007,~~ the atmospheric CH_4 increas~~was raise~~ed~~ed~~ significantly after 2007 (Fig. 12~~Fig. 11~~a). In ~~the year of~~ 1997/1998, 2000/2001, 2007/2008, and 2011/2012, a large~~r~~ ~~amplitude~~ fluctuation in~~of~~ the growth rates was found and a strong growth appeared (Fig. 12~~Fig. 11~~b). The growth rates fluctuated evenly with both positive and negative values before 2009. However,~~But~~ almost all of the growth rates showed a positive value after 2009. ~~From 1990s to 2010s, t~~Three ~~apparently~~ developing stages (~~i.e.~~i.e. highlighted green, blue, and red blocks) ~~were presented~~could be seen from the 1990s to 2010s.~~,~~ T~~t~~he CH_4 mole fraction slightly decreased during~~in~~ 1998-2000 (green color), and then ~~go~~ went through a relatively steady period during~~in~~ 2003-2006 (blue color), finally increased ~~steadily~~ rapidly after 2007 (red color) (Fig. 12~~Fig. 11~~).

The ~~overall~~ annual growth rate~~s~~ was~~ere~~ 5.1 ± 0.1 ppb yr^{-1} throughout~~over~~ 1994-~~2016~~2019 at the WLG (Table 4~~Table 4~~). ~~However, t~~The periodic annual growth rates were 4.9~~6~~ ± 0.1, 2.5~~6~~ ± 0.2, 4.9~~5.3~~ ± 0.1~~2~~, 7.7~~6~~ ± 0.1,~~2~~ and 5.5~~7~~ ± 0.1 ppb yr^{-1} during~~in~~ 1994-1997, 1998-2002, 2003-2007, 2008-2012, and 2013-~~2016~~2019, respectively. Similar growth rates were found between the CR and the TP during 1994-2019 (Fig. S6). In 1994-1997 and 2003-2007, the growth rates in the TP were even larger than that in the CR (Table 4). These results indicated that there were also strong CH_4 sources from the TP. The previous study by Zhou et al. (2004) showed the annual increase in CH_4 by 4.5 ppb yr^{-1} in 1992-2001, which was close to our study in 1994-1997 and 1998-2002. Tohjima et al. (2002) found that the CH_4 levels at the Cape Ochi-ishi and Hateruma Island in1995-2000 respectively increased by 4.5 and 4.7 ppb yr^{-1}, which were also similar to that of the WLG. Tsutsumi et al. (2006) analyzed the trend of hourly CH_4 data from 1998 to 2004 on the YON, which showed a similar increase (~3.0 ppb yr^{-1}) to the WLG. The study at the GR and NW in Seoul, Korea, presented almost an identical trend of 2 ppb yr^{-1} between 2004 and 2013 (Ahmed et al., 2015), which was lower than that of the WLG in similar period. In the early 1990s, the CH_4 growth rates at the WLG were very low and similar to the global level. The level of •OH radicals might control the decrease or increase of CH_4 in the atmosphere during this period (Dlugokencky et al., 1998; Rigby et al., 2017; Turner et al., 2017). However, the growth rates were high in 1998 (Fig. 12b), which may have been due to the high temperatures and a large amount of biomass burning (Cunnold et al., 2002; Lelieveld et al., 2004; Simmonds et al., 2005). The growth rate of CH_4 observed at the Ny-Ålesund, Svalbard, increased from 0.3 ± 0.2 ppb yr^{-1} during 2000-2005 to 5.5 ± 0.2 ppb yr^{-1} during 2005-2014, which had a similar variation but with a little lower growth rates than that of the WLG (Morimoto et al., 2017). The study suggested that the temporal pause in 2000-2005 was ascribed to the reductions of CH_4 emissions from the microbial and fossil fuel sectors, while the increase in 2005-2014 was due to an increase in microbial release.

The constantly larger CH_4 growth rate after 2007 at the WLG (Fig. 12) (Table 4) was similar to the recent studies by Nisbet et al. (2016) and (2019), which showed that the global CH_4 increased by 5.7 ± 1.2 ppb yr^{-1} in 2007-2014, and was much higher at 12.7 ± 0.5 ppb yr^{-1} in 2014, with 10.1 ± 0.7 ppb yr^{-1} in 2015, 7.0 ± 0.7 ppb yr^{-1} in 2016, and 7.7 ± 0.7 ppb yr^{-1} in 2017. The average growth rate in the Northern Hemisphere was 7.3 ± 1.3 ppb in 2007 and 8.1 ± 1.6 ppb in 2008 (Dlugokencky et al., 2009), which was also similar to the observation at the WLG (Table 4). After 2007, most sites in the Northern Hemisphere had large CH_4 growth rates. Also, the average global growth rate was similar to the WLG at 7.1 ppb yr^{-1} in the most recent ten years (WMO, 2019). Fang et al. (2013) showed that the annual growth rate of CH_4 was 9.4 ± 0.2 ppb yr^{-1} in 2009-2011 at the WLG, which was a little higher than this study in 2008-2012. The adjacent stations in China also revealed the high CH_4 growth rates of 8.0 ± 1.2 ppb yr^{-1} at Lin'an in 2009-2011, 7.9 ± 0.9 ppb yr^{-1} at Longfengshan in 2009-2011, and 10 ± 0.1 ppb yr^{-1} at Shangdianzi in 2009-2013 (Fang et al., 2013, 2016), which were all higher than the similar periods of 2008-2012 or 2013-2019 at the WLG (Table 4). The CH_4 measurements in other countries, such as the Beromünster tall tower station, also showed a high growth rate of 9.66 ppb yr^{-1} in 2012-2014 (Satar et al., 2016). The warm temperatures, biomass burning, and the climatic anomalies (El Niño or La Niña), likely enhanced the CH_4 emissions after 2007 (Dlugokencky et al., 2009). The anomalous years of increasing or decreasing (e.g. 2007/2008) might have a significant influence on the overall CH_4 trend (Fig. 12). These frequent anomalies also appeared in most long-term observation stations, e.g. MLO in the USA (Dlugokencky et al., 2009) and Mount Zugspitze in Germany (Yuan et al., 2019), due to climatic forces, such as those exceptions during the El Niño oscillation, forest fires, volcanic eruptions, and extreme weather events (Keeling et al., 1995; Dlugokencky et al., 2009; Keenan et al., 2016; Nisbet et al., 2019).

Many studies have investigated the potential reasons for the anomalous increasing. The study by Satar et al. (2016) at Beromünster, Switzerland explained that the short-term spikes were possibly related to emissions from agricultural activities, while the longer-lasting peaks were because of air mass transport and mixing. The isotopic evidence suggested that the significant increase of biogenic emissions was the dominant factor for the CH_4 rise. This was especially true in the tropical wetlands that had strong rainfall anomalies, and agricultural sources such as rice paddies and ruminants were a cause while fossil fuel emissions were not the main cause (Nisbet et al., 2016). The study from thaw ponds at Arctic regions revealed that there had very weak correlation between the amount of CH_4 released from ponds and environmental factors, e.g. air temperature and atmospheric pressure (Burke et al., 2019). Sweeney et al. (2016) using 29-year of measurements on North Slope of Alaska (BRW) to investigate the sensitivity of CH_4 emissions to the temperature change, which revealed that despite the short-term temperature sensitivity increases CH_4 emissions, it would have little impact in the long term. However, up to now, the specific causes of such distinct variability through the years, including the spikes or near-zero CH_4 growth rates, have not yet been determined. It is well established that human activities were mainly responsible for the recent rapid CH_4 growth rates and anomalies. The analysis from EDGAR showed the CH_4 emission per sector in China (Fig. S7) (Crippa et al., 2019). During the observing period, the waste, oil, and natural gas combustion, and open burning continuously emitted large amounts of CH_4 into the air. After 2000, the CH_4 emissions from solid fuel increased greatly in China. After 2003, the CH_4 emitted from rice cultivation also increased continuously (Fig. S7). The increased emissions from these sectors greatly contributed to the CH_4 increase at the WLG, as well as the other regions in China. In addition, studies revealed that China's coal sector dominated the positive trend in recent years, which contributed to the highest proportion of anthropogenic CH_4 emissions (~33%) (Janssens-Maenhout et al., 2019; Miller et al., 2019). In 2010-2015, China's coal production increased (from 3400 to 4000 million metric tons), but CH_4 emissions from rice cultivation, agriculture practices, ruminants, waste, and oil/gas consumption only increased slightly if at all (EIA, USA). Therefore, the control measures of coal mining reduction or limiting natural gas and petroleum exploitation may play an important role in slowing down CH_4 emissions in China.

The CH_4 growth rate in CR was significantly different from that in TP (Fig. S3). In 1994-1997, 2003-2007 and 2013-2016, the growth rates in TP were obviously larger than that in CR (Table 4). But in 2003-2007 and 2008-2012, the CR showed higher annual growth rates. In addition, for the entire observing period (i.e. 1994-2016), the growth rates in both TP (5.2 ± 0.1 ppb yr^{-1}) and CR (5.0 ± 0.1 ppb yr^{-1}) were similar to the overall annual growth rates (Fig. S3).

2. 103. 7 Case study for air mass transport

As described above, the northeast and southeast city regions might have acted as strong regional sources influencing the atmospheric CH_4 at the WLG. Therefore, to analyze the effect of long-~~distance~~ range transport of emissions from cities, ~~we further excluded~~ the regionally representative data ~~by air mass transport~~ was further excluded by air mass transport, and the ~~the rest of~~remaining regional records ~~were~~ were denoted as 'TR'. First, ~~We applied~~ the monthly cluster analysis was applied to hourly trajectories over 2005-2007, 2008-2012 and 2013-2017. Then, based on the cluster analysis, the clusters were divided into two groups, i.e. from city regions (red clusters in Fig. S8), and other (black clusters in Fig. S8). Finally, the regionally representative data were accordingly classified as two groups based on the cluster results (cities or other). The statistical results~~cluster results from city regions~~ were ~~presented~~ presented in detail in ~~Fig. S4~~Figure S8 and Table S3 ~~Table S3~~.

Consequently, t~~T~~he proportions of trajectories from cities were~~as~~ 40.3%, 32.5%, and 6.8% in 2005-2007, 2008-2012, and 2013-2017, respectively. A~~nd a~~bout 22.8%, 35.6%, and 38.4% of the regional records were associated with air masses ~~transport~~ from city regions in 2005-2007, 2008-2012, and 2013-2017, respectively (Table 5~~Table 5~~). The average CH_4 values for the~~when air masses transport from~~ city region air massess (1863.0 ± 0.3 ppb) were~~as obviously~~ higher than the other sectors (1850.6 ± 0.2 ppb) (Fig. S9~~Fig. S5~~). –The overall growth rates of the TR in the periods of 2005-2016 or 1994-2016 were similar to the original data series (Fig. S9). However, after excluding the CH_4 records for the~~when~~ air trajectory~~ies~~ transports from city regions, the growth rates of the TR in 2008-2012 (10.1 ± 0.1 ppb yr^{-1}) and 2013-2017 (6.3 ± 0.1 ppb yr^{-1}) (Table 5~~Table 5~~) were higher than the original regional data series (~~i.e.~~i.e. 7.6 ± 0.2 and 5.7 ± 0.1 ppb yr^{-1}) (Table 4~~Table 4~~). ~~And the overall growth rate for TR in 2005-2016 or 1994-2016 was still similar to original data series (Total), no significant difference was found (Fig. S5).~~

These results suggested that there were possibly other strong CH_4 sources at the WLG that were not from cities and the southwest region (Northern India) was the most likely contributor. The PSCF analysis also supported this result (Fig .7). At present, Northern India and Eastern China were the two largest sources of CH_4 at the WLG (Fig.

S10) (Crippa et al., 2019). Since the Tibetan Plateau was coincidently trapped in the middle of them, the atmospheric CH_4 at WLG was very likely dominated by long-range transport from these two regions. Although CH_4 emissions increased slowly during 1994-2002, a negative trend appeared (Fig. S10), significantly increased emissions were found in both southeast and southwest Asia after 2007. Chen et al. (2013) illustrated that the warming (0.2 °C per decade) in the Tibetan Plateau resulted in substantial emissions of CH_4 due to the thawed permafrost and melted glaciers. The rapid increase of CH_4 would probably make it difficult to meet the goals of carbon emission reduction in the future. This would be especially true with the scenario of quickly increasing CH_4 on the Qinghai-Tibetan Plateau due to the emissions from the two largest source regions of Northern India and Eastern China. The large growth rate of atmospheric CH_4 in the TP revealed that i) the atmospheric CH_4 at the WLG was not predominantly influenced by eastern cities in recent years and ii) large amounts of CH_4 were transported from the Tibetan Plateau to WLG in recent years.

Discussion

Anthropogenic emission on temporal patterns

In the early years, the daily cycles of atmospheric CH_4 at WLG were not distinct and the amplitude was small (Fig. 2a-b), which were similar to the previous studies by Zhang et al. (2013) and Fang et al. (2013). The ambiguous diurnal patterns indicated that the local sources were weak at WLG in the past. The apparent diurnal cycles after 2000 may be attributed to the intense activities by human (e.g., grazing, fuel burning), which was aggravated in daytime and weak in nighttime (Fang et al., 2013). The meteorological conditions (e.g., diffusion and transport) could contribute to the increasing CH_4 amplitudes. The WLG was remote from the populate center, the good diffusion condition in daytime may bring high CH_4 mole fractions to the site. The increasing amplitude (Table S1) and CH_4 mole fractions over time suggested that the WLG was affected by increasingly local sources (e.g., human activities) (Zhou et al., 2004). The maximum was found in summer in recent years (Fig. 2e) could also be ascribed to the transport of anthropogenic sources by the meteorological factors. In summer, the intensely herd or graze activities around WLG might enhance the regional CH_4 emissions and hence contribute to the higher CH_4 mole fractions. The higher CH_4 mole fraction in winter in past years (Fig. 2a) was probably because of the way of heating (e.g., large biomass burning) as well as the adverse diffusion conditions in cold weather.

The previous study by Zhou et al. (2004) found that higher CH_4 mole fractions appeared when the winds come from the ENE-E-ESE-SE sectors at WLG during 1991-2002, which was similar to our study in similar period (Fig. 3a-b). Causes of the elevated CH_4 from these wind sectors could be attributed to the large plantation of highland barley as well as high population density in those areas (Fang et al., 2013). Two largest cities Xining and Lanzhou are situated in northeast and east of WLG, respectively, which could emit large amount of CH_4 by human activities. Besides, previous studies on black carbon (BC) and carbon monoxide (CO) indicated that the emissions from the Yellow River Canyon industrial area, ~500km away from northeast of WLG may also donate to the high CH_4 values originating from ENE and NE sectors (Zhou et al., 2003). In summer, the prevailing wind directions were from NE … ESE sectors (~46%) (Fig. S6), and the CH_4 mole fractions were also higher in the related sectors. However, in the autumn and winter, although the prevailing wind and high wind speed were from SSW … W sectors (~ 40-50%) (Fig. S6), the high CH_4 mole fractions were from almost the opposite wind sectors of NNE …. ESE (Fig. 3f), which indicated that strong local sources were distributed from northeast to southeast (city regions), and even covered the emissions of natural sources. As time goes on, the wind sectors with high CH_4 mole fractions changed and concentrated on ESE to ENE sectors, and the amplitudes of enhancements were increasing, which further implied the effect of stronger emissions from anthropogenic sources in city regions in recent years.

Pollutant sources regions

Sources regions

The air masses from east and northeast regions passed over the cities of Xining and Lanzhou (capital of Gansu province), which is the populated center and industrial area (Fig. 5). However, the highest CH_4 values was not observed when air mass was from these sectors. Instead, high CH_4 mole fractions were frequently observed when air mass from the northwest to southwest (Table 1). It was possibly due to that the air masses from west and northwest had passed through the northwest of Qinghai province and the central area of Xinjiang Uygur Autonomous Region (XUAR), where located Ge'ermu urban area (the second largest city of Qinghai) with rapid industrial development, natural gas and petroleum resources exploitation and large crops residue burning (Zhang et al., 2013). Similar to the CPF percentile analysis (Fig. 4), the southwest or northwest region away from the site may be also strong source regions.

Most potential source identified in northwestern regions (Fig. 6) was possibly due to CH_4 emissions from the northwest Gansu province, the northwest Qinghai province and the southeast of XUAR. The different source distribution by seasons could be attributed to the effect of westerlies or the southeast monsoons (Zhou et al., 2004). The obviously increasing source region was clear evidence for the strong effect of the expansion of human activity. Moreover, the pattern of source region moved from the east to the southwest, especially in autumn and winter, indicated that the southwest away from the WLG, e.g., Northern India, were gradually becoming a strong CH_4

source region. India has abundant cattle as well as extensive large-scale coal mining, large amount of CH_4 emissions may transport from northern India to the northeastern Tibetan Plateau (Fig. 6i & l). The air mass transport result (Fig. 5d) also support the result that the southwest air masses (cluster 1) contributed the highest CH_4 mole fractions. The studies of atmospheric Hg at WLG by Fu et al. (2012) also revealed this phenomenon.

Different sources between CH_4 and CO

The percentile polar plot clearly showed the specific distribution of different CH_4 mole fractions (Fig. 4). The result revealed that most areas around WLG contributed to low CH_4 mole fractions, the southeast and southwest of the site exist two strong source regions. It is of great possible that the anthropogenic emission from cities (e.g., Lanzhou, Chengdu, etc.) was the only cause for high values in the southeast, and the southwest region away from WLG was possibly due to sources from other countries, such as India. Unlikely to the CH_4, the high CO mole fractions were consistent from east regions (urbanized area) (Fig. 4), indicating strong anthropogenic sources in city regions (i.e. Xining and Lanzhou) (Zhang et al., 2011).

The seasonal cycles of $\Delta CO/\Delta CH_4$ slopes (high in summer and low in winter) (Fig. 8) may primarily be due to the effect of monsoons and air mass transport. In summer, air masses arriving at WLG were predominantly transported from the northeast to east city regions (e.g., Xining, Lanzhou) with the largest CO mole fractions. In contrast, the air masses were mainly from the southwest in winter, which carried strong CH_4 emissions but few CO emissions (Zhang et al., 2011) (Table 1). Hence the opposite two air mass transport lead to a peak in summer and a trough in winter (Fig. 8). Moreover, we could see apparently regional polarization in the concentration ratio of CH_4 and CO (Fig. S7), implying the different strong source distribution between CH_4 and CO at WLG. The cluster results (Fig. 5b & d) and the potential sources analysis (Fig. 6f & l) also support this seasonal variation. Tohjima et al. (2014) found an opposite variation at Hateruma Island, which showed low slope values in summer. It could be attributed to different local sources and sinks, suggested the special topography condition and local source distribution around WLG. The large $\Delta CO/\Delta CH_4$ fluctuations (Fig. 9) over the study period was likely because of the anomaly years of different CH_4 or CO mole fractions as well as source regions. In 2007, large increase of ΔCH_4 appeared, and from 2010 to 2013, the ΔCO decreased significantly (Fig. S1). Before 2010, large air masses and potential source regions were identified in eastern regions (city regions) with the highest CO emission (Fig. 5; Fig. 6). After 2010, the southwest regions showed high contributions, with the highest CH_4 emission but relatively lower CO emission. Therefore, obviously variation of the slopes presented with almost an increase in 2007-2010 and a decrease after 2010 (Fig. 9).

Long-term variations

Seasonal cycles

The seasonal variations with maximum in summer and minimum in spring at WLG (Fig. 10) were consistent with the previously short-term studies, e.g., Zhang et al. (2013) in 2002-2006. However, it was almost opposite to observation in the adjacent stations such as Lin'an, Shangdianzi and Longfengshan in China (Fang et al., 2013, 2016), We further compared similar WMO/GAW global stations in the north hemisphere, including MLO (19.54° N, -155.58° E, 3397m a.s.l.) (Dlugokencky et al., 1995, 2019a), JFJ (46.55° N, 7.99° E, 3580m a.s.l.) (Zellweger et al., 2016), MNM (24.29° N, 153.98° E, 7.1m a.s.l.) (Matsueda et al., 2004; Tsutsumi et al., 2006), as well as the marine boundary layer (MBL) from NOAA/ESRL lab at similar latitude (Dlugokencky et al., 2019b).as well as most global stations in the northern hemisphere, e.g., MLO, BRW and JFJ (Dlugokencky et al., 1995; Loov et al., 2008). We further compared similar WMO/GAW global stations in the north hemisphere, including MLO (19.54° N, -155.58° E, 3397m a.s.l.) (Dlugokencky et al., 1995, 2019a), JFJ (46.55° N, 7.99° E, 3580m a.s.l.) (Zellweger et al., 2016), MNM (24.29° N, 153.98° E, 7.1m a.s.l.) (Matsueda et al., 2004; Tsutsumi et al., 2006), as well as the marine boundary layer (MBL) from NOAA/ESRL lab at similar latitude (Dlugokencky et al., 2019b). It can be seen that the stations in the northern hemisphere (i.e. MLO, JFJ and MNM) and the MBL showed an opposite trend to WLG with the minimum in summer and maximum in winter or spring (Fig. 12). And the seasonal amplitude at WLG (~14 ppb) was lower than many other sites in the northern hemisphere, about 35-70 ppb, e.g., MLO, BRW, bialystok in Poland, ochsenkopf in Germany and beromunster in switzerland (Dlugokencky et al., 1995; Thompson et al., 2009; Popa et al., 2010; Satar et al., 2016). MBL also showed larger amplitude than that in WLG (Fig. 12). The low amplitude at WLG was possibly because of high elevation of continental mountain sites (e.g., WLG, JFJ), where were relatively less affected by local influences than the coastal/island sites (Yuan et al., 2019). The peak in summer at WLG may be attributed to larger grazing and human activities than other seasons. The CH_4 emissions from yaks and other ruminants in Tibetan Plateau (alpine pasture) were very strong in summer, preceded only by paddy emission (Fang et al., 2013; Zhang et al., 2013). Furthermore, the photochemical capacities were very weak (high altitude) and the dynamic transport by air flow from polluted northeast/southeast region was also strong in summer at WLG, which all induced high CH_4 mole fractions in summer and consequently an opposite trend with other sites (Ma et al., 2002; Xiong et al., 2009).

Long-term trends in different observing periods

The entirely fluctuant trend of atmospheric CH_4 in 1994-2016 at WLG (Fig. 11) was similar to the global trend reported by quite a few studies (Bergamaschi et al., 2013; Rigby et al., 2017; Nisbet et al., 2019). The previous study by Zhou et al. (2004) showed the CH_4 annual increase of 4.5 ppb yr^{-1} in 1992-2001, which was similar to that in 1994-1997 (4.6 ± 0.1 ppb yr^{-1}) and 1997-2002 (2.6 ± 0.2 ppb yr^{-1}) in our study (Table 4). Tohjima et al. (2002) found similar growth rates that the CH_4 at Cape Ochi-ishi and Hateruma Island in1995-2000 was increased about 4.5 and 4.7 ppb yr^{-1}, respectively. In early 1990s, the CH_4 trend at WLG is very low, which was similar to the global growth rates (Fig. 11b). The levels of OH radicals may have controlled the decrease or increase of CH_4 in the atmosphere during this period (Dlugokencky et al., 1998; Rigby et al., 2017; Turner et al., 2017). The growth rates were high in 1998 (Fig. 11b), which may have been due to the high temperatures, large biomass burning and weak destruction (Cunnold et al., 2002; Lelieveld et al., 2004; Simmonds et al., 2005).

The continuously larger CH_4 growth rates after 2007 at WLG (Fig. 11), e.g., 7.6 ± 0.2 ppb yr^{-1} in 2008-2012, 5.7 ± 0.1 ppb yr^{-1} in 2013-2016 (Table 4), were similar to the recent study by Nisbet et al. (2016) and (2019), which showed the global CH_4 increased by 5.7 ± 1.2 ppb yr^{-1} in 2007-2014, and the much higher of 12.7 ± 0.5 ppb yr^{-1} in 2014, 10.1 ± 0.7 ppb yr^{-1} in 2015, 7.0 ± 0.7 ppb yr^{-1} in 2016 and 7.7 ± 0.7 in 2017 ppb yr^{-1}. The average growth rate in the northern hemisphere was 7.3 ± 1.3 ppb in 2007 and 8.1 ± 1.6 ppb in 2008 (Dlugokencky et al., 2009), which was also similar to the observation at WLG (Table 4). After 2007, most sites in the northern hemisphere displayed large CH_4 growth rates. Fang et al. (2013) showed that the annual growth rate of CH_4 was 9.4 ± 0.2 ppb yr^{-1} in 2009-2011 at WLG, which were a little higher than our study in similar period. The adjacent stations in China also revealed high CH_4 growth rates, e.g., 8.0 ± 1.2 ppb yr^{-1} at Lin'an in 2009-2011, 7.9 ± 0.9 ppb yr^{-1} at Longfengshan in 2009-2011, and 10 ± 0.1 ppb yr^{-1} at Shangdianzi in 2009-2013 (Fang et al., 2013, 2016), which was higher than the similar period in 2008-2012 or 2013-2016 at WLG (Table 4). The CH_4 measurements in other countries, e.g., Beromünster tall tower station also showed high growth rate of 9.66 ppb yr^{-1} in 2012-2014 (Satar et al., 2016). The very warm temperatures, large biomass burning and the climatic anomaly e.g., El Niño, La Niña event, were likely enhanced the CH_4 emissions after 2007 (Dlugokencky et al., 2009). Additionally, the anomalously sharply increasing or decreasing years (e.g., 2007/2008) may have a significant influence on the overall CH_4 trend (Fig. 11), and these frequent anomalies appeared in most long-term observation stations, e.g., MLO in USA (Dlugokencky et al., 2009), Mount Zugspitze in Germany (Yuan et al., 2019), which were also possibly attributed to climatic forces, such as the exception during the El Niño oscillation, forest fires, volcanic eruptions, and extreme weather events (Keeling et al., 1995; Dlugokencky et al., 2009; Keenan et al., 2016; Nisbet et al., 2019). The study by Satar et al. (2016) at Beromünster, Switzerland explained that the short-term spikes were possibly related to emissions from agricultural activities, while the longer lasting peaks were because of air mass transport and mixing.

It is well established that the human activities are mainly responsible for the recent rapid CH_4 growth rates and anomalies. The analysis from Emissions Database for Global Atmospheric Research (EDGAR) showed the CH_4 emission by per sector in China (Fig. S8) (Crippa et al., 2019). During the observing period, the waste, the oil and natural gas and open burning continuously emitted large amount of CH_4 into the air. After 2000, the CH_4 emission from solid fuel increased greatly in China. After 2003, the CH_4 emitted from rice cultivation also increased continuously (Fig. S8). The increased emissions from these sectors may greatly contribute to the CH_4 increase at

WLG, as well as the other regions in China. In addition, the recent studies reveled that China's coal sector may have dominated the clearly positive trend in recent years, which contributed the highest proportion of the anthropogenic CH_4 emissions (~33%) (Janssens-Maenhout et al., 2019; Miller et al., 2019). In 2010-2015, China's coal production increased obviously (from 3400 to 4000 million metric tons), but emissions trends of CH_4 by rice, agriculture, ruminants, waste, and oil/gas have grown slightly and even remained flat (EIA, USA). The isotopic evidence suggests that the significant increase of biogenic emissions was the dominant factor of CH_4 rise, especially in the tropical wetlands with strong rainfall anomalies, or the agricultural sources such as rice paddies and ruminants, fossil fuel emissions have not been the main cause (Nisbet et al., 2016). The study by Chen et al. (2013) illustrated that the warming (0.2 °C per decade) in the Tibetan Plateau resulted in substantial emission of CH_4 due to the permafrost thawing and glaciers melting. However, up to now, the specific causes of such distinct variability around the years, e.g., the spikes or near-zero CH_4 growth rates, are not yet determined.

Annual growth rate in Qinghai-Tibetan Plateau

Although similar annual growth rates were found among the City Regions (CR), the Tibet Plateau (TP) and total regional records (Total) in the entire observing period (1994-2016) (Fig. 11), significant differences were found in short-term periods (Fig. S3). In 2013-2016 (Table 4), the TP showed larger growth rate than that in CR, implying stronger CH_4 source in the Tibetan Plateau in recent years. The seasonal amplitude in the Tibetan Plateau was continuously increasing, which also revealed that the Tibetan Plateau was intensively affected by strong regional sources. Without air mass transport from the city regions, the significantly increased annual growth rate (TR) in 2008-2012 (10.1 ± 0.1 ppb yr^{-1}) and 2013-2016 (6.3 ± 0.1 ppb yr^{-1}) (Fig. S5 and Table 5) suggested that there were possibly other strong CH_4 sources around WLG not from cities. Northern India and eastern China were obviously the largest two source regions of CH_4 at WLG (Fig. S9) (Crippa et al., 2019). Since the Tibetan Plateau was coincidently trapped in the middle of the largest increased source areas, the atmospheric CH_4 at WLG was very likely dominated by long-distance transport from these two regions. Although CH_4 emissions increased slowly during 1994-2002, and even negative trend appeared in southeast China (Fig. S9), significantly increased emissions appeared in both southeast and southwest Asia after 2007. The rapidly increased CH_4 would

~~probably make it difficult to meet the goals of carbon emission reduction in the future. Especially on the scenario of quick increasing CH_4 on the Qinghai-Tibetan Plateau due to the emission from two largest source regions. In view of the integrated eco-environmental change processes and unique topography in the Qinghai-Tibetan Plateau, it may provide us one of the last precious regions to study global climate changes (Chen et al., 2013). The anomalously year to year fluctuations of atmospheric CH_4 in Tibetan Plateau were unquestionably a warning or alarm to the world, and the unprecedented annual growth rate might be a dangerous signal to global climate change.~~

~~3~~4 Conclusions

Three developing stages of atmospheric CH_4 at Mt. Waliguan from the 1990s to 2010s were found. The CH_4~~atmospheric CH_4 at Mt.Waliguan increased continuously during 1994-2017.~~ mole fractions slightly decreased during 1998-2000, and then went through a relatively steady period during 2003-2006 and finally increased rapidly after 2007. Although near-zero and even negative growth appeared in some ~~particular~~ periods, ~~e.g., 1999-2000, and 2004-2006,~~ the overall ~~trend of~~ CH_4 ~~was~~ increased rapidly, especially in recent ~~decade~~years. ~~Obvious diurnal cycle was found with the peak at noon and a trough at late afternoon. Due to the unique geophysical locations and transport pathway, the seasonal averages of CH_4 at WLG displayed an opposite trend with sites in the northern hemisphere, with summer maximum and spring minimum.~~ Although most areas around the WLG had small contributions to the CH_4 emissions, two strong source regions were found from the northeast and southwest of the site. ~~Large amount of air masses was from west and northwest regions of WLG, which accompanied with higher CH_4 mole fractions than that from city regions. The~~ Northern India possibly has bec~~o~~ame a stronger ~~source of CH_4~~contributor ~~to WLG rather~~ than city regions ~~before~~were in the past.

~~As time goes by, t~~The temporal patterns ~~(e.g., seasonal amplitude)~~, the annual variations, the long-term trends, or the ~~potential~~ source distribution of CH_4 at WLG we~~a~~re all ~~changing~~changed in recent years. We found that the WLG was increasingly affected by local sources such as human activities. ~~Thus, the long-term verification is extremely important to accurately understand CH_4 variations.~~ Additionally, the Tibetan Plateau was intensively affected by strong sources over time~~The case study in Qinghai-Tibetan Plateau revealed unprecedented annual growth rates of CH_4. In recent years~~, which~~the Tibetan Plateau~~ ~~even~~ showed a larger growth rate than that ~~in~~ of the city regions in some periods. –T~~Tibetan Plateau was with the highest average altitude and was almost impervious to strong human activities. There is no doubt that t~~he anomalous~~ly~~ variation and ~~the~~ unprecedented ~~annual~~ growth rate of the atmospheric CH_4 in this region ~~might be a dangerous signal to global climate change~~revealed that it was urgent to control CH_4 emissions. Reducing the emissions from strong source sectors like coal mining, natural gas or solid fuel exploitation, and rice cultivation may play an important role on CH_4 emissions reduction in China.

Data availability. The gridded meteorological data (2004-2017) from NOAA-ARL was available at ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/. The data from MLO, JFJ, MNM station was downloaded from World Data Centre for Greenhouse Gases (WDCGG) at https://gaw.kishou.go.jp/. The MBL data was available at ftp://aftp.cmdl.noaa.gov/data/trace_gases/CH4/flask/surface/. The geographical distribution of annual emission data by Emissions Database for Global Atmospheric Research (EDGAR) was from website https://edgar.jrc.ec.europa.eu/overview.php?v=50_GHG.

Author contributions. SL, SF and ZF designed the research. SL performed the data processing with assistance of SF and MG. The station were monitored, maintained ML and PL, and they collected, preprocessed, and provided the hourly observational dataset. SL and SF finished the manuscript with contributions from all the co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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Table 1. The statistics of the filtered CH_4 data series over different periods during 1994-2019 at the WLG station.

Year	Regionally representative			Locally influenced		
	Hours	Percentage (%)	Mean (ppb)	Hours	Percentage (%)	Mean (ppb)
1994-1997	16122	71.3	1801.7 ± 0.5	6481	28.7	1806.2 ± 0.3
1998-2002	26347	83.2	1832.6 ± 0.7	5336	16.8	1837.7 ± 0.3
2003-2007	28181	69.4	1832.3± 0.3	12443	30.6	1839.2 ± 0.2
2008-2012	19627	63.5	1856.2 ± 0.4	11287	36.5	1865.2 ± 0.3
2013-2019	21683	44.2	1906.8 ± 0.3	27329	55.8	1920.4 ± 0.4
1994-2019	111960	64.0	1865.8 ± 0.4	62876	36.0	1868.2 ± 0.3

Table 2. Yearly average CH_4 mole fractions at the WLG station.

Year	Mean (ppb)	year	Mean (ppb)
1994	1799.0 ± 0.4	2007	1837.2 ± 0.5
1995	18035.68 ± 0.1	20082006	1854.8 ± 0.11835.6 ± 0.2
1996	18084.86 ± 0.2	20092007	1847.2 ± 0.11839.9 ± 0.5
1997	181106.58 ± 0.2	20102008	1856.6 ± 0.21865.9 ± 0.3
1998	18267.80 ± 0.1	20112009	1867.4 ± 0.11849.1 ± 0.1
1999	181920.72 ± 0.1	20122010	1879.6 ± 0.21857.9 ± 0.2
2000	181920.70 ± 0.2	20132011	1895.7 ± 0.41872.6 ± 0.4
2001	18479.32 ± 0.4	20142012	1890.2 ± 0.21881.2 ± 0.3
2002	18335.75 ± 0.2	20152013	1913.0 ± 0.41896.3 ± 0.2
2003	18402.85 ± 0.23	20162014	1914.4 ± 0.21890.4 ± 0.1
2004	1836.17 ± 0.21	20172015	1911.6 ± 0.11905.6 ± 0.3
2005	18367.74 ± 0.1	20182016	1925.6 ± 0.31903.8 ± 0.1
2006	1834.7 ± 0.2	2019	1932.0 ± 0.1

Table ~~1~~3. The statistics for the cluster analysis result for both CH_4 and CO from 2004-2017 at the WLG station. The clusters from urban areas are highlighted ~~with face~~in bold.

	Cluster	Number	Average CH_4 mole fraction	Average CO mole fraction
Spring	**1**	**1243**	**1853.4 ± 2.7**	**185.4 ± 3.3**
	2	685	1852.6 ± 3.4	147.8 ± 3.6
	3	2231	1877.6 ± 2.5	175.6 ± 3.0
	4	**1093**	**1850.5 ± 2.2**	**196.0 ± 4.3**
	5	4108	1860.8 ± 1.5	137.8 ± 1.1
Summer	**1**	**3981**	**1869.9 ± 1.2**	**173.2 ± 2.4**
	2	2244	1878.5 ± 2.6	135.0 ± 2.6
	3	**1040**	**1866.3 ± 2.6**	**165.3 ± 5.2**
	4	916	1857.8 ± 2.4	152.2 ± 5.1
	5	578	1876.3 ± 5.1	146.5 ± 5.4
Autumn	**1**	**1133**	**1870.1 ± 3.7**	**159.1 ± 8.0**
	2	4235	1868.8 ± 1.3	110.3 ± 1.3
	3	2745	1873.6 ± 1.6	135.1 ± 2.6
	4	550	1865.2 ± 3.6	149.1 ± 6.6
Winter	1	3066	1879.8 ± 2.1	159.9 ± 3.6
	2	**601**	**1872.6 ± 3.4**	**282.2 ± 2.1**
	3	5261	1865.8 ± 1.0	129.6 ± 1.7

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Table ~~4~~4. Annual growth rates of atmospheric CH_4 in the City Regions (CR), the Tibetan Plateau (TP), and ~~original~~ total regional records ~~(Total) during~~ from 1994 to -2019~~6~~ at the WLG station.

	1994-1997	1998-2002	2003-2007	2008-2012	2013-~~2016~~2019	1994-~~2016~~2019
CR	3.0~~2.8~~ ± 0.1	3.6 ± 0.2	5.3~~6~~ ± 0.2	7.0~~1~~ ± 0.2	6.2~~5.5~~ ± 0.1	5.2~~0~~ ± 0.1
TP	3.4 ± 0.1	3.0 ± 0.2	6.7~~8~~ ± 0.2	5.7~~6~~ ± 0.2	5.7~~7.3~~ ± 0.1	5.1~~2~~ ± 0.1
Total	4.9~~6~~ ± 0.1	2.5~~6~~ ± 0.2	4.9~~5.3~~ ± 0.1~~2~~	7.7~~6~~ ± 0.1~~2~~	5.5~~7~~ ± 0.1	5.1 ± 0.1

Table ~~5~~5. The statistics of CH_4 data without air mass transport from city regions (TR) over different periods during 2005-2016 at the WLG station.

	Transport regions	Hours	Percentage (%)	Average (ppb)	Updated growth rate (ppb yr-1)
2005-2007	TR	6922	77.2	1824.9 ± 0.2	2.7 ± 0.2
	City	2041	22.8	1835.9 ± 0.5	-
2008-2012	TR	7060	64.4	1853.7 ± 0.2	10.1 ± 0.1
	City	4254	35.6	1861.0 ± 0.3	-
2013-2016	TR	4152	61.6	1888.2 ± 0.3	6.3 ± 0.1
	City	2591	38.4	1888.5 ± 0.5	-
2005-2016	TR	18134	67.1	1850.6 ± 0.2	7.0 ± 0.1
	City	8886	32.9	1863.2 ± 0.3	-

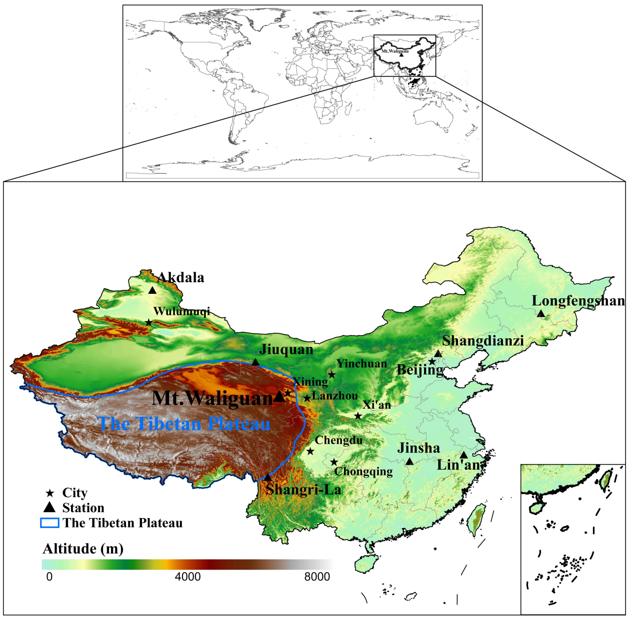


Figure 1. The l~~L~~ocation of Mt. Waliguan WMO/GAW global station as well as the other regional stations in China. The gradient color indicates altitude. The digital elevation model (DEM) was downloaded from Geospatial Data Cloud site, Computer Network Information Center, Chinese Academy of Sciences (http://www.gscloud.cn), and then processed by ArcGis software. The China map was derived from © National basic Geomatics Center of China (http://www.ngcc.cn/ngcc/). The world map was obtained from © OpenStreetMap (https://www.openstreetmap.org/). ~~And the other shpfile file data and entire map were created by ArcGis software.~~

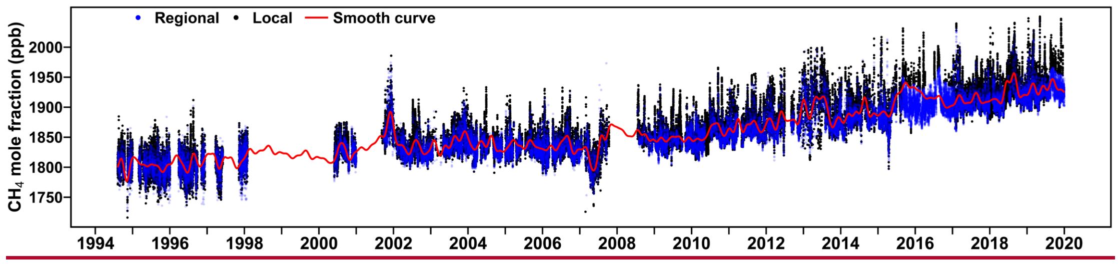
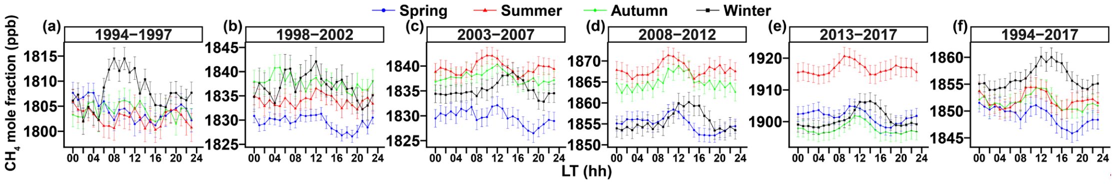


Figure 2. The filtered hourly CH_4 data series from 1994 to 2019 at the WLG station. The transparent blue points are regionally representative data. The black points are locally influenced data. The red lines are smooth values of the regional data obtained by the curve-fitting routine of Thoning et al. (1989).



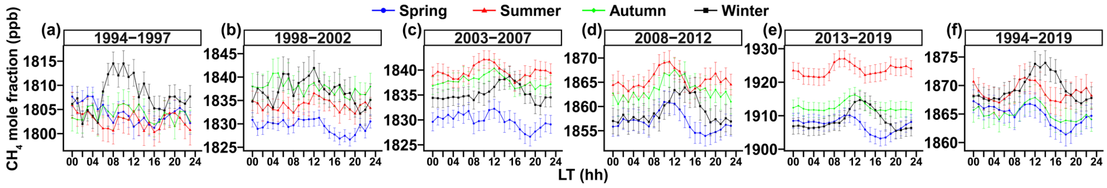


Figure 3. Diurnal CH_4 cycles in different periods from 1994 to ~~2017~~2019 at the WLG station. The lines with different colors represent various seasons. Error bars indicate the 95% confidence intervals.

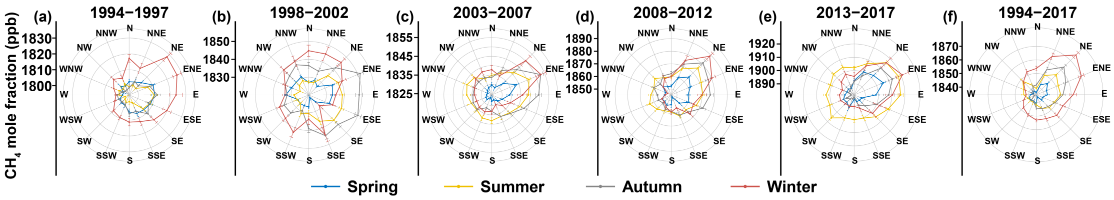


Figure 4. The wWind-rose distribution of the average hourly CH_4 records mole fractions from 16 horizontal wind directions over different periods duringin 1994-2017 at the WLG station. The different colors represent the CH_4 data in different seasons. Error bars in all directions indicate 95% confidence intervals.

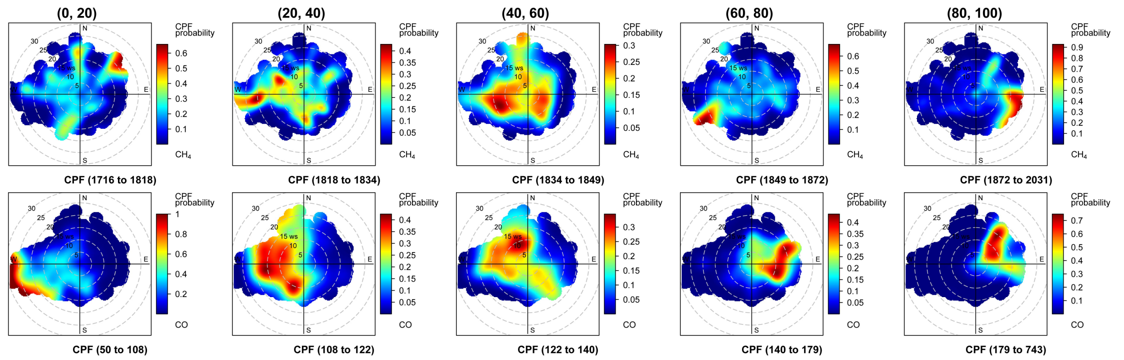


Figure 5. The polar plot of the distribution of the CH_4 and CO concentration ~~probability~~ probabilities in different percentile ranges at the WLG station. The analysis was based on ~~the~~ conditional probability functions (CPF) by Ashbaugh et al. (1985). The top plots show the ~~measurements~~ analysis of CH_4 from 1994 to 2017. The bottom plots show the CO measurements in 2004-2017. ‘ws’ ~~means~~ refers to the wind speed. The values ~~in~~ at the bottom of each panel show the range of concentrations in the relevant percentile range. Gradient colors represent the levels of CPF probability in different percentile ranges.

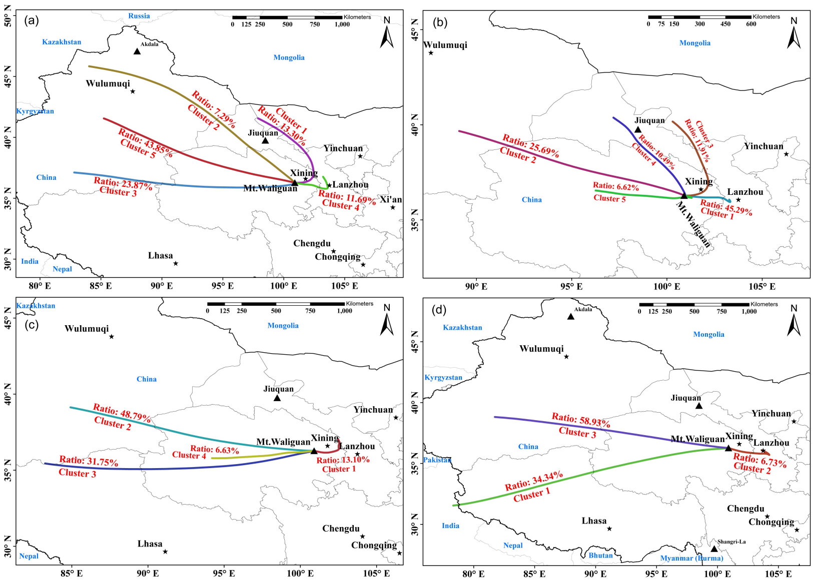


Figure 6. Cluster analysis ~~to~~ of the 72-h back trajectories in different seasons (spring (a), summer (b), autumn (c), and winter (d)) during 2004-2017 ~~ending~~ at WLG station. ~~The (a), (b), (c) and (d) represents spring, summer, autumn and winter, respectively.~~ The colored lines ~~with different colors~~ ~~denote~~represent different cluster ~~analysis results~~s. The proportion of trajectories ~~i~~on each cluster is also marked.

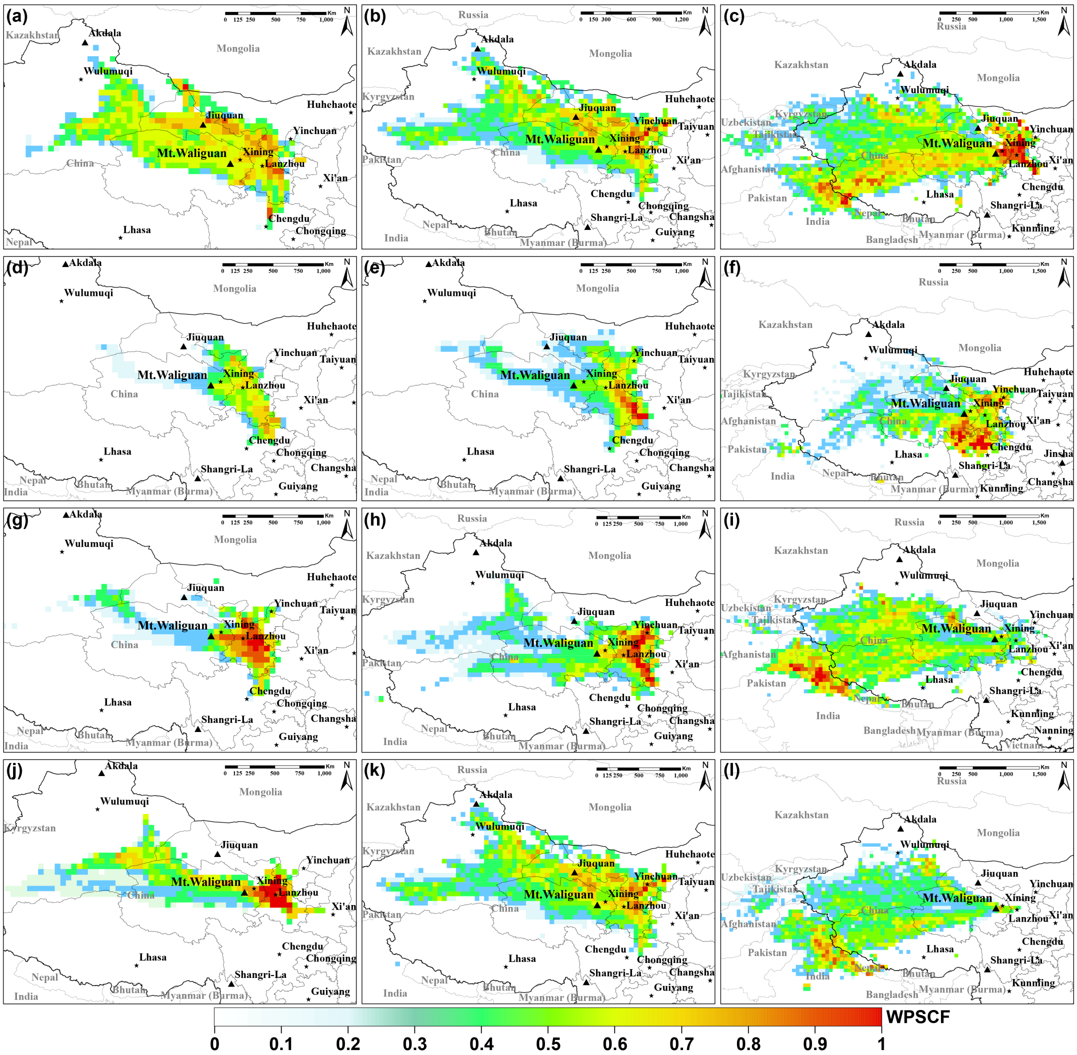


Figure 7. The g~~G~~eographical distribution of the weighted potential sources of CH_4 in different periods over 1994-2017 at the WLG station. The gradient color shows the strong levels of potential source regions in different seasons, ~~i.e.~~i.e. spring (a, b, c), summer (d, e, f), autumn (g, h, i), and winter (j, k, l), and different periods, ~~i.e.~~i.e. 2004-2007 (a, d, g, j), 2008-2012 (b, e, h, k), and 2013-2017 (c, f, i, l).

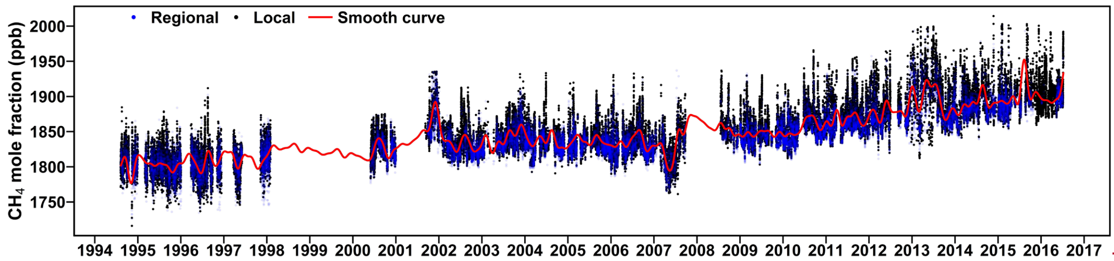


Figure 7. Filtered hourly CH_4 data series from 1994 to 2017 at WLG station. The blue points with transparency represent regional events. The black points are the filtered local events. The red lines are calculated smooth values to the regional data by curve-fitting routine of Thoning et al. (1989).

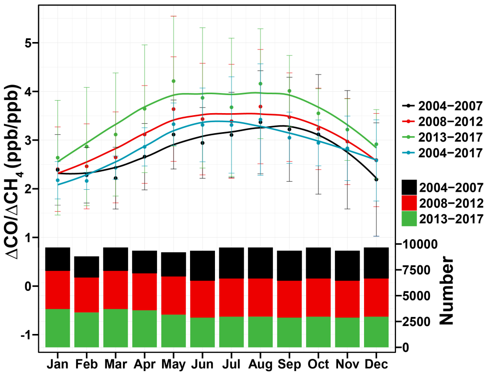


Figure 8. ~~A~~The average seasonal variation of the $\Delta CO/\Delta CH_4$ slopes in different periods ~~over~~ during 2004-2017 at the WLG station. The error bars show the standard deviation of the monthly averages. The vertical bars are the monthly numbers of data in different periods.

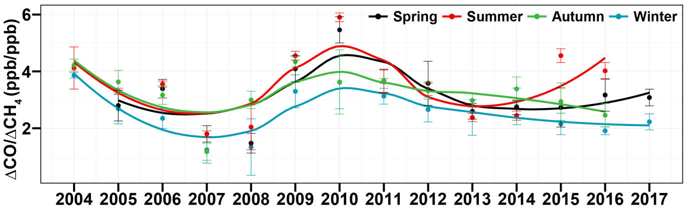
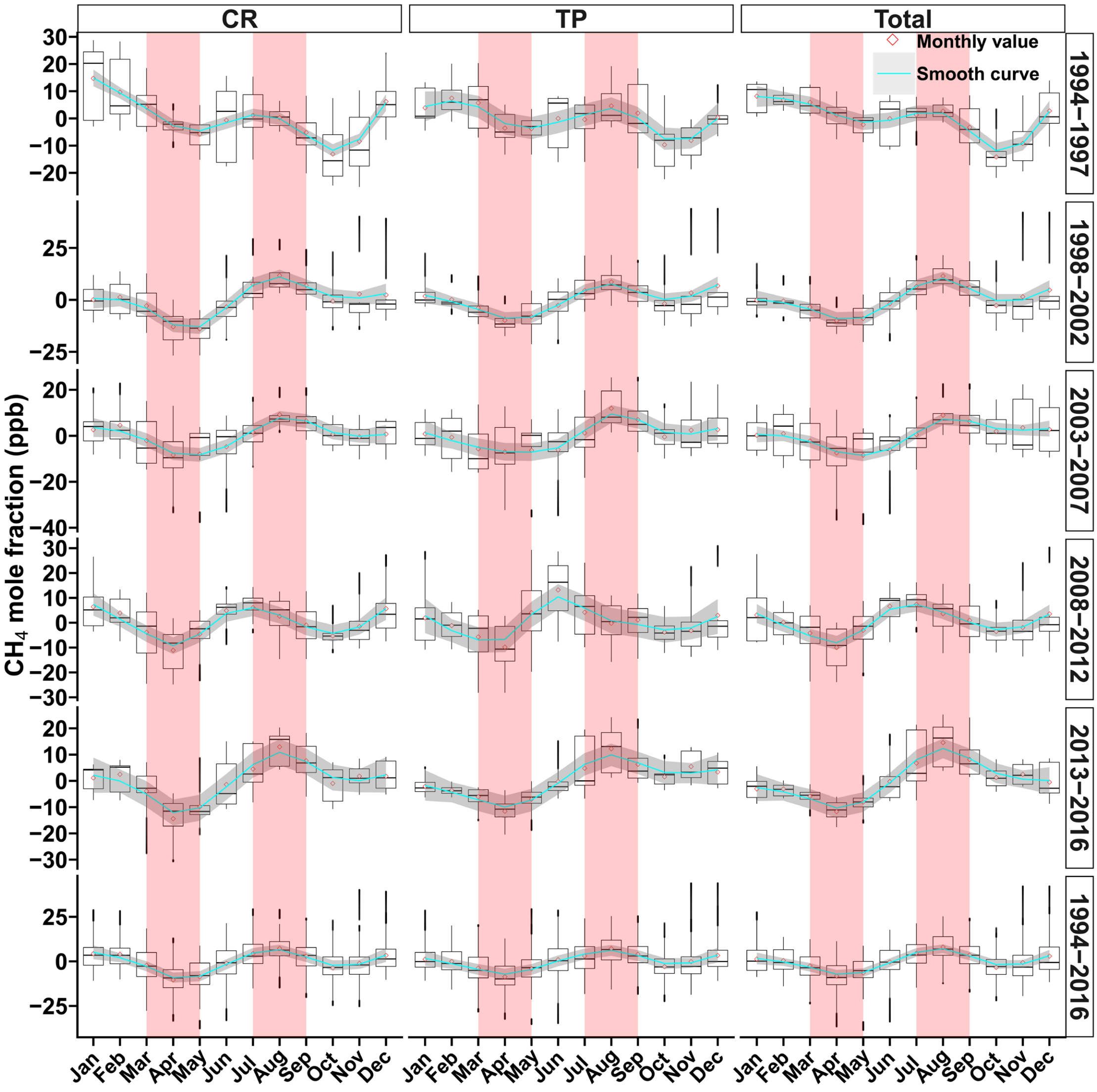


Figure 9. The l~~L~~ong-term trend of $\Delta CO/\Delta CH_4$ slopes over 2004-2017 at WLG station.


CR
TP
Total
Monthly value
Smooth curve
1994–1997
1998–2002
2003–2007
2008–2012
2013–2016
1994–2016
CH_4 mole fraction (ppb)
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec

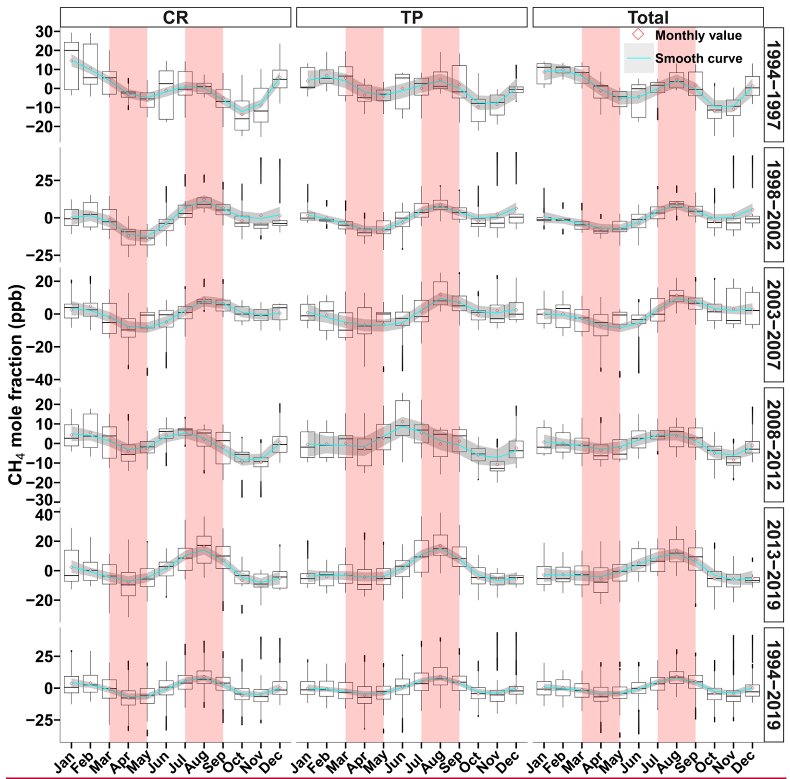


Figure 10. Monthly variations of regional CH_4 mole fractions from 1994 to 20162019 at the WLG station. The 'CR', 'TP' and 'Total' represents the measurements from the City Regions, the Tibetan Plateau and the original total regional records, respectively. The box respectively shows the 25th percentile, the median and the 75th percentile from bottom to top. The bottom and the top whiskers respectively reaches the minimum and 1.5 times the IQR (interquartile range). The black points are identified as outliers. The red squares are the averages. The cyan lines are the smoothed curve of the averages using the method of loess (Local Polynomial Regression Fitting). The gray bands are the 95% confidence interval of smoothed curve.

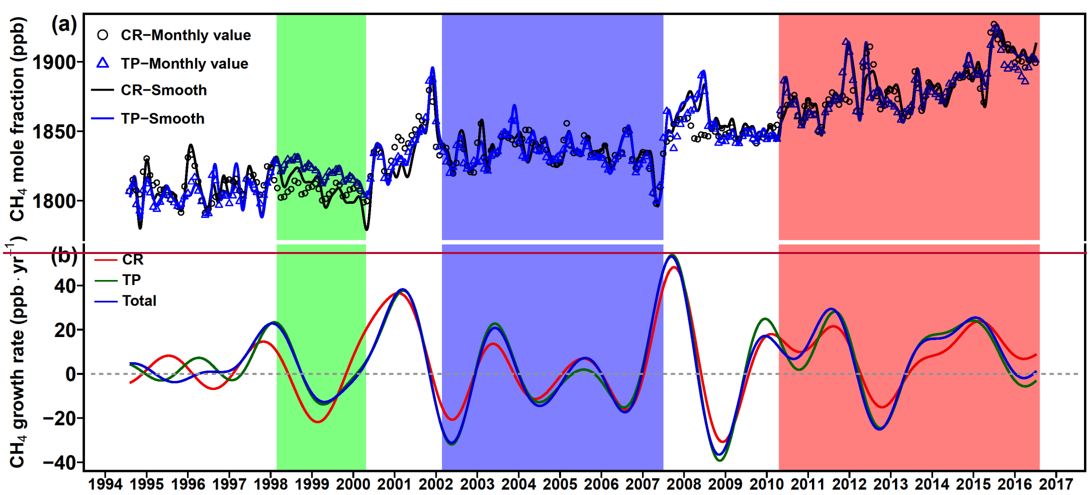
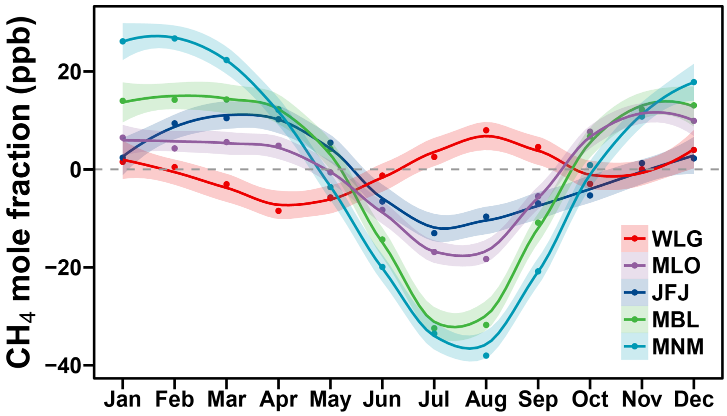


Figure 11. The top panel (a) shows the smooth curve and monthly values of CH_4 mole fraction in the City Regions (CR) and the Tibet Plateau (TP) during 1994-2016 at WLG station. The bottom panel (b) is the annual growth rates of atmospheric CH_4 records from CR, TP as well as the total regional time series (Total). The growth rates are calculated from the first derivative of trend curves. The smooth curve and the trend is calculated by the method of Thoning et al. (1989).



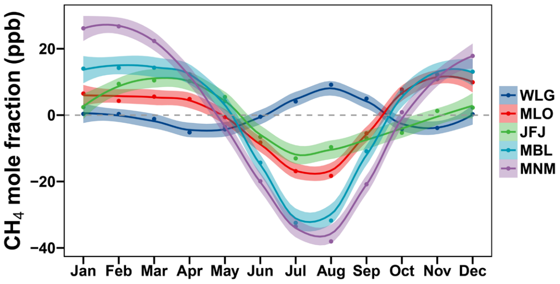


Figure 11 12. The seasonal cycles of atmospheric CH_4 observed over 1994-2017 at the WMO/GAW global stations of Mauna Loa (MLO, 1994-2018), Jungfraujoch (JFJ, 2005-2018), Minamitorishima (MNM, 1994-2019), and Mt. Waliguan (WLG, 1994-2019) in the Nnorthern Hhemisphere. The data of other sites (except WLG) weare from WDCGG. The data in the marine boundary layer (MBL, 1994-2019) weare from NOAA / ESRL lab at the a similar latitude to the WLG.

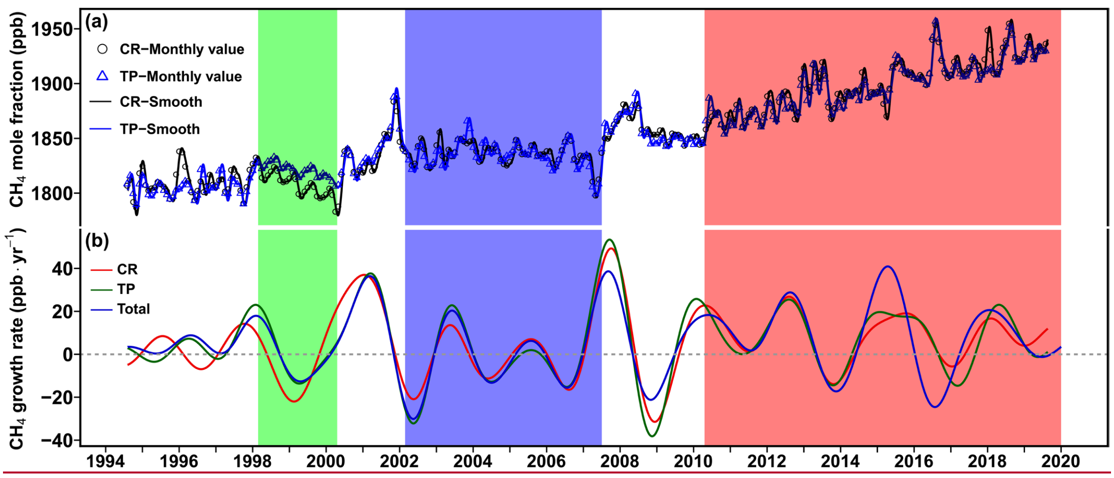


Figure 12. The top panel (a) shows the smoothed curves and monthly values of the CH_4 mole fractions in the City Regions (CR) and the Tibetan Plateau (TP) during 1994-2019 at the WLG station. The bottom panel (b) is the annual growth rates of the atmospheric CH_4 records from the CR and the TP as well as the total regional time series. The growth rates were calculated from the first derivative of the trend curves. The smoothed curves and the trends were calculated using the method of Thoning et al. (1989).