Point by point response to the reviewer #1:

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 reviewer #1.

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 "△CO/△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. ΔCO/ΔCH_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 ΔCO/ΔCH_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 deltaCH$_4$? 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).

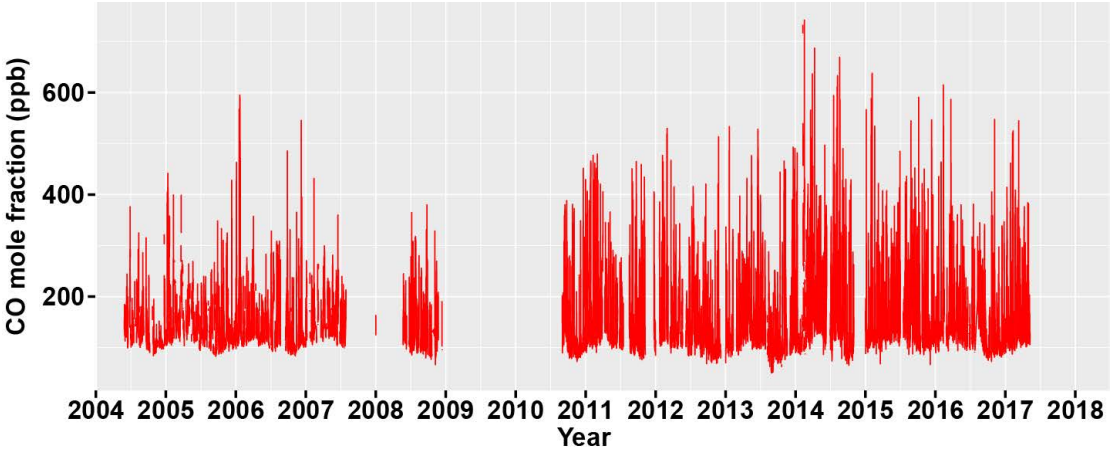

Figure. S3. The hourly CO data from 2004 to 2017 at the WLG station.

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 deltaCH$_4$ ratios?

Response: The strong seasonal variation of ΔCO/ΔCH$_4$ 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.

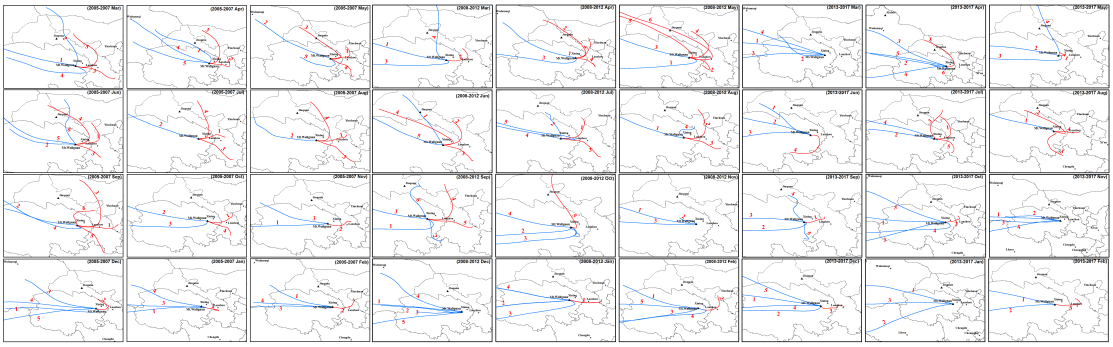

Figure S8. The monthly cluster analysis of hourly trajectories during 2005-2017 at the WLG station. The red clusters represent air masses transport from city regions.

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.