Response to comments of Anonymous Referee#1

Dear Referee#1:

We are very pleased to receive your comments, which are quite helpful to improve this manuscript. The point-to-point replies are listed below.

(Annotation: YELLOW HIGHLIGHTs are additions or revise in the manuscript or in the appendix)

General Comments

This manuscript has systematically developed a carbonaceous aerosol emission inventory for five overlooked types of human activities (namely burning, sacrificial incenses, joss paper burning, fireworks, cooking, and barbecue) in China from 2000 to 2018, filling a gap in most existing emission inventories. It also contributes plenty of experimental emission factors of carbonaceous aerosols from these sources. The author analyzed the trends, characters, and drivers of these FMS emissions. The findings and datasets are unique and informative for the academic community, however, there are some essential issues requiring further clarification. And the released emission datasets need careful double-checking, especially their fluctuation trends, spatial and temporal distribution patterns. Besides, as a first-hand emission inventory and dataset, more comprehensive validations and uncertainty analyses should be conducted and discussed. Detailed comments are listed as followings.

Response:

Thanks for your positive comments and helpful suggestions on this manuscript. We have carefully revised the problems you raised. The data was re-uploaded and more validation works were done. Responses to the detailed comments are given below.

Specific Comments

1. In the methods section 2.3, the author adjusted the per capita consumption of sacrificial incenses, joss paper with statistical temple numbers, population, and forest

fires. I noticed the temple numbers and forest fires have the same weights in the adjustment (both 0.5, in equation 9). Is there any reference or principle for the adjustment methods, and what about the extent of the impact such adjustments may have on the activity data? Similar issues also apply to equation 10.

Response:

Thank you for this comment. Since this study is the first establishment of carbonaceous aerosol emission inventory for FMS, there are no additional coefficient values (like the above-mentioned 0.5) for reference, except for our previous work (Wu et al, 2021). Based on previous studies, we attempted to introduce more parameters (the ratio of sacrificial forest fires to all forest fires) to better describe the consumption of incense and paper money. Therefore, this is a subjective judgment according to current existing data, without reference. Before the adjustment, the average per capita joss paper consumption in each province was calculated of 1.21±1.65 kg. We believe that there existed high variation in per capita consumption for various provinces. After the adjustment, the average value was 1.07±0.81 kg. The differences among provinces have decreased. So we think that this processing can reduce errors.

Wu, J., Kong, S., Zeng, X., Cheng, Y., Yan, Q., Zheng, H., et al. (2021). First high-resolution emission inventory of levoglucosan for biomass burning and non-biomass burning sources in China. *Environmental Science & Technology*, *55*(3), 1497–1507. https://doi.org/10.1021/acs.est.0c06675

2. According to the author's analysis, firework plays an important role in FMS emissions, thus its activity trends should be carefully checked. In Figure3, there is an obvious peak in fireworks in 2014, and it also leads to a peak in BC/EC/ACSBrC emissions in 2014 (Figure 4). I did not find any interpretation of this peak throughout the manuscript. The author should check this peak value to figure out whether it makes sense. The author could also try to use other datasets, such as the variation of PM2.5 concentrations, and satellite-retrieved NO2, in the Chinese Spring Festival during 2013-2018 to offer some supporting validation.

Response:

Thanks for this suggestion. The surge in consumption was due to the huge sale volumes in statistic data, which might caused by the destocking after that the air pollution control plan was implemented. We attempted to collect the direct observation data. The Air Pollution Prevention and Control Action Plan was published on September 10, 2013, and the Chinese New Year's eve (CNE) and Chinese Spring Festival (CSF) were on February 9, and February 10. The CNE and CSF in 2014 were on January 30 and January 31. Therefore we did not collect the observation data of air quality for the CNE and CSF of 2013. Corresponding data for the year of 2014 were only available for about half of the cities. Thus, we adopted other datasets which have been cited for many times (DOI: 10.5281/zenodo.6398971; Wei et al., 2021; Wei et al., 2020) to verify our results. Considering that the firework burning was mainly concentrated in rural regions after 2013, we conducted a correlation analysis between the FMS **ACS_{BrC} emissions on New Year's Eve** and the **$PM_{2.5}$ concentration in non-urban regions on New Year's Eve** after 2013, and the result can be accepted (r=0.85, p<0.05). The relationship was also significant between 2000 and 2018 (r=0.59, p<0.01). The variation trends of the emission and $PM_{2.5}$ concentration were similar. Therefore, we believe that the emission peak in 2014 is possible. The relative discussion was added in **Text S6** and Line 354–355 (The surge in sales might have been caused by destocking after that the Air Pollution Prevention and Control Action Plan (APPCP) was implemented.)

Text S6: The surge in ACS_{BrC} emission in 2014 might be out of the ordinary. We attempted to use the $PM_{2.5}$ concentration dataset (Wei et al., 2020, 2021) to verify the accuracy of the inventory. The ACS_{BrC} emissions mainly came from fireworks burning (**Figure S8**), and most of the fireworks were burnt in rural regions during the Chinese New Year (**Section 3.3.3**). We conducted a correlation analysis between the FMS ACS_{BrC} emissions and $PM_{2.5}$ concentration in non-urban regions for the New Year's Eve. The results were shown in **Figure S16**. There was a positive correlation (r=0.59, p<0.01) between the ACS_{BrC} emissions and $PM_{2.5}$ concentration. The correlation (r=0.85,

p<0.05) was even higher if we focus only on the period after 2013. Thus, the emission surge in 2014 was possible. However, more accurate verification is still needed to be carried out by chemical transport models in the future.

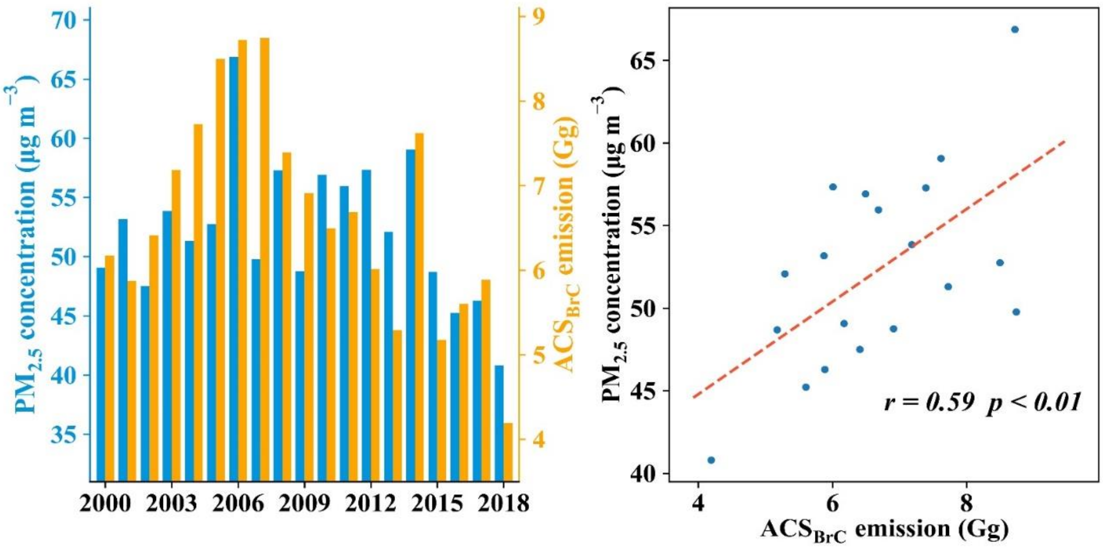

Figure S16 $PM_{2.5}$ concentration and ACS_{BrC} emission from FMS in rural China in 2000–2018 and the correlation between them.

Wei, J., Li, Z., Lyapustin, A., Sun, L., Peng, Y., Xue, W., Su, T., and Cribb, M. Reconstructing 1-km-resolution high-quality $PM_{2.5}$ data records from 2000 to 2018 in China: spatiotemporal variations and policy implications. *Remote Sensing of Environment*, 2021, 252, 112136. https://doi.org/10.1016/j.rse.2020.112136

Wei, J., Li, Z., Cribb, M., Huang, W., Xue, W., Sun, L., Guo, J., Peng, Y., Li, J., Lyapustin, A., Liu, L., Wu, H., and Song, Y. Improved 1 km resolution $PM_{2.5}$ estimates across China using enhanced space-time extremely randomized trees, *Atmospheric Chemistry and Physics*, 2020, 20(6), 3273-3289. https://doi.org/10.5194/acp-20-3273-2020

3. In the results section 3.3.1, the author analyzed the multi-year FMS emission variations. However, it is mostly the description of the emission results, rather than the driver analysis or explanation. For instance, there is no explanation of the emission trends of firework, which is the most important contributor to FMS emissions. On the other hand, the author focused on the analysis of RC emission trends in this section, however, RC contributes little to BC and EC emissions, thus the driver analysis of RC emissions might not be able to represent the driver of FMS. Besides, in Figure4, I

noticed the BC and OC emission of RC show different trajectories. Given the same emission source and similar control efficiency, what causes such differences?

Response:

Thanks for this helpful comment. We have added corresponding discussions of the driver analysis of FMS in Section 3.3.1 (now in Line 355–360: From 2000 to 2006, the resident's income raised by 76.5% due to the booming economy. The residents have more money to purchase fireworks. And only another 12 cities have forbidden fireworks burning in 2000–2006. It can be the reason for the increase in fireworks consumption amounts. From 2006 to 2018, although people's incomes continued to rise, while the urbanization rate increased by 16.0% and additional 201 cities have forbiden the fireworks burning, which lead to the decrease of fireworks consumption amount at this period). Furthermore, we have appropriately reduced the discussion of the impacts of RC on the total FMS emissions. For the error in Figure 4, the differences between BC and OC trajectories might be related with the y-axis view limits. Due to the image aesthetics, we set the y-axis view limits of the figure of OC at 3200–5499 to clearly show the emissions from sources other than RC. The y-axis view limits of the figure of BC were set as 1–357. If we set the y-axis view limits of the figure of OC and BC as 1–5499 and 1–50, the difference in the trajectories will be much smaller, but the emissions from other sources will be difficult to be seen (Figure 3-1, this is a draft image for the presentation only). Meanwhile, the image can not fully show the BC emission from FMS. In fact, since the same activity data was used and the emission factors were different, if we plot the OC and BC emissions from RC on the same graph, the temporal tendencies of their emissions are the same.

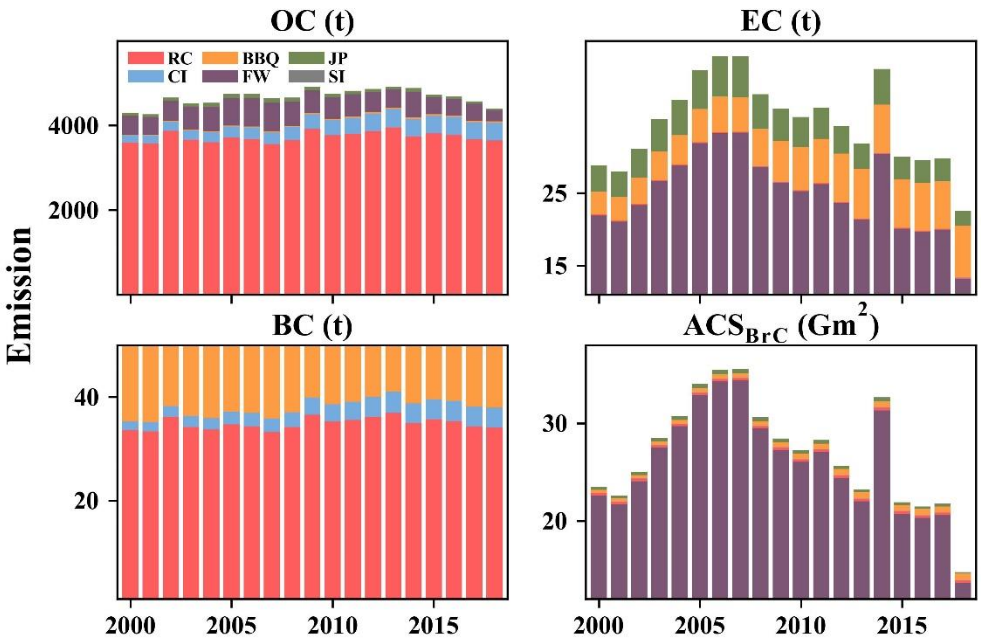

Figure 3-1 Total CA emissions from FMS in China from 2000 to 2018

4. The explanation of the spatial distribution methods is not clear or sufficient, and it makes me confused about some spatial variation analyses in section 3.3.2. First, whether the location of temple and restaurant points are used as the spatial allocation proxy? And the author pointed out that "High emission regions of sacrificial incense and joss paper overlapped with the areas with large numbers of temples" in Line378-379, however, I noticed several mismatches between sacrificial incense emissions (Figure S10) and the temple distributions (Figure S3), as showed in the following red labels. Besides, I suggest the author make some statistical analysis between different FMS emissions and their related activity data distributions, such as at the grid level or city level, to further evaluate their spatial correlations, as well as the spatial accuracy of the developed emission datasets.

Response:

Thanks very much for this suggestion. The POI data for temples was not directly used as the proxy, and it was used when calculating the per capita consumption of SI and JP in each city. The basic allocations of SI and JP were related to the population distribution. The POI data of restaurants were used as the proxy of CI because we believe that CI is a point source similar to industrial or power plant sources. The mismatch of the red label might be caused by the population. The labels are all located in densely populated regions. More than half of the population (59.6 million) of Northeast China is on the top-right red label (the rectangle of 123.5–130 °E and 41.5–48.5 °N). The population on the center red label (112.5–128.5 °E and 31.5–38 °N) is 338 million. The population is the basic proxy for the distribution of SI emissions, thus the large populations in the red labels might cause the mismatch between the spatial distributions of emissions and temple. We have calculated the correlation of the emissions and activity data at the city level, as shown in **Text S7** and **Table S3**. For example, the total emissions of SI were positively related ($r > 0.36$, $p < 0.01$) to the number of temples in each city. Thus the spatial distribution of emissions from SI coincides with that of temples to a certain extent. What's more, the population was the basic data for the emissions from all FMS, thus the correlations between population and emissions of each city were positive ($r > 0.47$, $p < 0.01$).

Text S7: As shown in **Table S3**, there existed a correlation between the activity data and the emissions. Population data was the basic data of the emissions for all FMS, thus the correlations between population and emissions of each city were positive ($r > 0.47$, $p < 0.01$). The other activity data for FMS were also correlated. For example, the total emissions of SI and JP in each city were positively related to the number of temples, which lead to their spatial distribution of emissions being coincidental with that of temples to a certain extent. This phenomenon also existed for other FMS.

Table S3 The correlation of OC emissions and activity data at a city level in 2018

Source	Activity data	r^{**}
SI	POI of temples	0.36
JP	POI of temples	0.44
FW	POI of firework shops	0.53
BBQ	POI of BBQ restaurants	0.68
CI	POI of restaurants	0.67
BBQ	meat consumptions	0.85
CI	meat consumptions	0.87
RC	meat consumptions	0.78

**: $p<0.01$

5. Following specific comment 5, another major confusion is about the spatial allocation of firework emissions. As the author pointed out, some cities, particularly the urban regions in Beijing city, have released a series of policies to forbade fireworks. However, I noticed the urban area in Beijing still shows high-level BrC emissions (which are majorly contributed by fireworks based on Figure S8) in 2018 (Figure 5). Given the spatial-temporal specialty of firework emission characters, as well as its domination of total FMS emissions, I think the firework forbidden policies should be fully considered in this dataset, both on emission calculation and spatial-temporal allocations. Besides, I simply downloaded the released dataset and compared it with the Figure 5 in the manuscript (BrC in 2018). I found some inconsistencies in the following red labels. I suggest the author double-check the released emission datasets.

Response:

Thanks for this comment. There are several reasons for the high ACS_{BrC} emissions in Beijing. First, Beijing is a highly urbanized city, over 86.5% of the population (about 18.63 million) live in the urban region. Second, Beijing starts to forbid fireworks burning in the last century, which means that most people in Beijing can not burn fireworks. At the same time, a large population number will also lead to abundant ACS_{BrC} emissions from cooking and BBQ, which lead to a relatively high-value area in the spatial distribution. In fact, cooking and BBQ in urban regions contributed 10.2% of ACS_{BrC} emissions in Bejing, and it is concentrated in the central urban regions. In addition, we have considered the prohibition on fireworks burning when allocated the spatial distribution of emissions, as shown in Figure 5-1a (ACS_{BrC} emission from all FMS) and Figure 5-1b (ACS_{BrC} emission from fireworks burning). Fireworks burning in non-urban regions contributed 86.6% of ACS_{BrC} in Beijing. Since there is no clear data on fireworks forbidden regions in each city, we consider the whole urban regions (divided by the grid data from Gong et al. 2019, 2020) as the forbidden regions when considering the impact of this policy on the spatial distribution of emissions. This is an alternative method as the lack of data currently. We think this approach is relatively reasonable but the subtle errors in detail are hard to fix by this method, which can be improved in the future.

We have also double-checked the datasets. When we processed the data and established the emission inventory, the raw data adopted the geographical coordinate system (WGS 84) with a resolution of 0.00833° (~1 km). There was a mistake when we converted the final calculated data to the projection coordinate system (1 km × 1 km). So the uploaded data was different from the data in the manuscript. We have re-uploaded the data at the resolution of 0.00833° and changed the description of the manuscript. We apologize for this error and thank you very much for comparing the data so carefully. We really appreciate your serious attitude on science.

The datasets can be found at https://doi.org/10.6084/m9.figshare.19999991.v2.

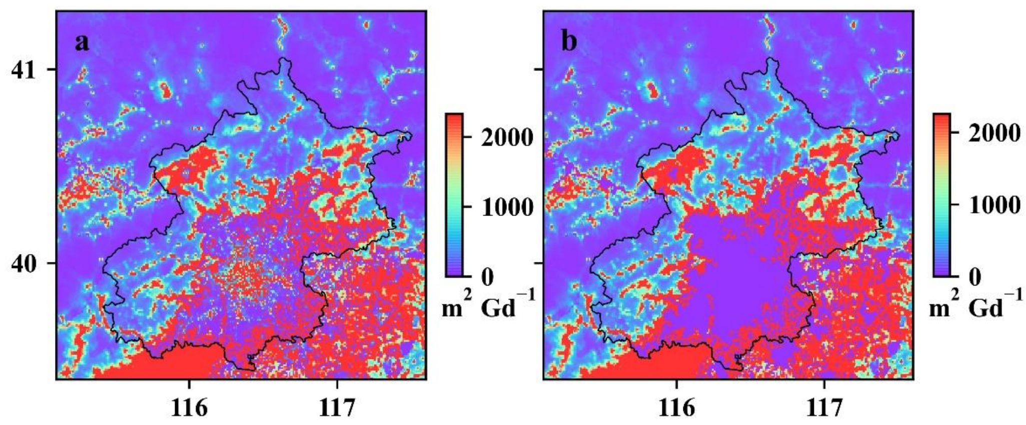

Figure 5-1 ACS_{BrC} emission from FMS (a) and fireworks (b) in Beijing

Gong, P., Li, X., & Zhang, W. (2019). 40-Year (1978–2017) human settlement changes in China reflected by impervious surfaces from satellite remote sensing. *Science Bulletin*, *64*(11), 756–763. https://doi.org/10.1016/j.scib.2019.04.024

Gong, P., Chen, B., Li, X., Liu, H., Wang, J., Bai, Y., et al. (2020). Mapping essential urban land use categories in China (EULUC-China): Preliminary results for 2018. *Science Bulletin*, *65*(3), 182–187. https://doi.org/10.1016/j.scib.2019.12.007

6. The author should carefully double-check the monthly variation of FMS CA emissions. In Figure 6, OC emissions in Jan/Feb are nearly high as twice as other months on average from 2000-2018; however, in Figure S13, OC emissions of Jan/Feb seem more than three times higher than other months in most years.

Response:

Thank you for this comment. Figure 6 shows the 19-year averaged monthly emissions, while Figure S13 shows each month's emission from 2000 to 2019. We have mentioned that 96.2% of fireworks were burned in CNE, CSF, and LF. In many years (2000, 2002, 2005, 2008, 2010, 2011, 2013, and 2016), all three festivals fell in the same month (Table 6-1). Furthermore, in 2000–2018, all CNE and CSF were in the same month. This means that most of the fireworks were set off in the same month, and it leads to the extremely high emission in Figure S13. While for Figure 6, the high values shown in Figure S13 are lost due to the average calculation. As a result, it appears that the value of Figure S13 is much higher than Figure 6. The data of monthly emissions have been uploaded. More discussions have been added in the manuscript (Line 408–409: all CNE and CSF are in the same month in 2000–2018, after the calculation of multi-year data, the results for January and February in **Figure 6** seem to be lower than those in **Figure S13**).

Table 6-1 Exact date of CNE, CSF, and LF in 2000–2018

Year	CNE	CSF	LF
2000	2000-02-04	2000-02-05	2000-02-19
2001	2001-01-23	2001-01-24	2001-02-07
2002	2002-02-11	2002-02-12	2022-02-26
2003	2003-01-31	2003-02-01	2003-02-15
2004	2004-01-21	2004-01-22	2004-02-05
2005	2005-02-08	2005-02-09	2005-02-23
2006	2006-01-28	2006-01-29	2006-02-12
2007	2007-02-17	2007-02-18	2007-03-04
2008	2008-02-06	2008-02-07	2008-02-21
2009	2009-01-25	2009-01-26	2009-02-09
2010	2010-02-13	2010-02-14	2010-02-28
2011	2011-02-02	2011-02-03	2011-02-17
2012	2012-01-22	2012-01-23	2012-02-06
2013	2013-02-09	2013-02-10	2013-02-24
2014	2014-01-30	2014-01-31	2014-02-14
2015	2015-02-18	2015-02-19	2015-03-05
2016	2016-02-07	2016-02-08	2016-02-22
2017	2017-01-27	2017-01-28	2017-02-11
2018	2018-02-15	2018-02-16	2018-03-02

7. In the summary and conclusions sections, the author suggests raising residents' income to reduce FMS emissions, such as fireworks. However, in section 3.3.4, only the relationship between residents' income and OC emissions is discussed. FMS-related OC emissions mainly come from residential cooking sources, with little from other sources (Figure S8). As the author figured out that various FMS emission sources have widely different characters, thus whether such a relationship also works for other FMS emission sources, especially the dominated firework sources?

Response:

Thanks for this query. We assessed the correlations between per capita fireworks OC emissions and income between 2000 and 2018. In general, a similar correlation exists (r^2=0.60, p<0.01). What's more, the ACS_{BrC} emission from FMS was also correlated (r^2=0.59, p<0.01) with income. Therefore, we believe that the conclusion that the residents' income impacted on the FMS emissions was reasonable. Corresponding discussion was added in the manuscript (Line 459–460: This correlation existed for ACS_{BrC} emissions dominated by fireworks burning, while the correlation was weaker ($r^2 = 0.59$, $p < 0.01$) than that of OC emissions dominated by cooking sources).

Other Comments

1. On Page3, Line70, 'Till,' means 'Till now'?

Response:

Yes. It has been changed to "Till now" (Line 70).

2. On Page7, Line166, the author pointed out they got the per capita consumption of various FMS activities of each province from household investigations; while in SI TextS2, the author said the survey covered only 27 provinces. Then where does the consumption data of other provinces come from? Any assumptions were applied?

Response:

Thanks for this query. As shown in Figure S9, China has seven geographical regions. We assume that the customs of the inner provinces of each geographical region are more similar than those of the outer provinces. Thus, the consumption data of other provinces (which were not in the 27 provinces) were the average of the geographical region where the province was located.

3. On Page17, Line 388-389, 'Emissions from urban regions were near zero, while emissions from suburbs and rural regions were much higher' It is hard to recognize in Figure 5. I suggest the author highlight or zoom in on some typical regions on the map.

Response:

Thanks for this suggestion. This sentence is more of a guide to Section 3.3.3, indicating the image of the fourth line of Figure 8. We have added the label of the image in parentheses (Line 395–396: (more details can be found in **Section 3.3.3**, **Figure 8**)). The zoom has been highlighted in Figure 8.

4. On Page20, Line 421-422, what does ‘to improve air pollution’ mean?

Response:

Thanks for your comments. Sorry for the unclear description. It has been corrected as “to improve the air quality”. We have made changes to this sentence in the manuscript. (Line 428–429: China set up 13 prevention and control regions (3 key regions and 10 city clusters, 3-10R) in 2013 to improve air quality).

5. On Page25, Line491-492, incomplete sentence.

Response:

Thanks for the comment. It has been revised as “As a result, OC, EC, BC, and ACS_{BrC} emissions from FMS have declined by 14.3–47.1%, 9.8–45.4%, 9.2–42.2%, and 10.4–48.2% in 2000–2018, respectively”. (Line 497–499)

6. On Page28, Line583, is the datasets of ‘the annual and monthly restaurant sales’ on a national level or a province level?

Response:

They are both on a province level.