Anonymous Referee #1

General Description of manuscript:

The authors developed an emission inventory of anthropogenic non-methane volatile organic compounds for Beijing-Tianjin-Hebei (BTH) region of China for 2015 using emission factor approach. Their estimate of total anthropogenic VOCs emissions over BTH in 2015 is 3277.66 Gg. The authors reported that their emission inventory shows significant consistence with both ambient measurements and satellite-derived emission inventory. PMF analysis of online measurements and their emission inventory show that vehicle emissions dominate the anthropogenic VOCs in Beijing. This study is interesting, and within the general scope of ACP. However, there are some weaknesses in current version. For example, many key statements come without citation; some references are inappropriate; some conclusions are not fully supported by their figures and numbers; some discussions are not quantitative. Therefore, I think this manuscript needs a major revision before it become suitable for publication.

Response: Thanks a lot for your dedicated work. We really appreciate the careful reading and the useful suggestions, which help to improve the manuscript considerably. We have fully considered the comments and made revisions to our manuscript. The response and changes are listed below. The responses are in black, and revised portions are marked blue in the letter.

General Comments:

1. Activity data is quite important in EF-based emission inventory. Where did the activity data come from? Can you please provide a table of activity data for each source and for each category?

Response: Accepted. Thank you for your valuable suggestion. In the revised manuscript, a table containing activity data for each source and for each category and the corresponding reference was provided in the Supplementary Information (Table S2).

2. What is the monthly variability of the EF-based emission inventory? And what's the difference between your monthly emissions and the satellite-derived monthly emissions?

Response: Accepted. Thank you for your comment. The monthly VOC emissions of EF-based emission inventories can be calculated by monthly profile for each source, which usually developed based on monthly statistics (Li et al., 2017b). According to the method of Li et al. (2017b), Wu et al. (2018), and Zhang et al. (2009), we provided monthly profiles for every source in the Table S3 of the Supplementary Information.

In the revised manuscript, we provided the monthly variability of the EF-based emission inventory and discussed the difference between our monthly emissions and the satellite-derived monthly emissions. A table of monthly profiles used in this study was provided in the Supplementary Information (Table S3). Following sentences were added in section 2.1.2 to describe the method of temporal distribution, "The monthly variability of this VOC emission inventory was calculated based on the monthly profiles (Table S3). In summary, monthly profiles for industrial emissions were developed based on the monthly output of industrial products (NBS, 2015). Power plant monthly profile was derived from monthly statistics of power generation (NBS, 2015). Monthly profiles of

residential fossil fuel combustion were estimated based on household survey results (Guo et al., 2015;Zheng et al., 2014). Monthly profile of on-road vehicle emissions was derived from Li et al. (2017b). For field crop residue burning, the monthly profile was estimated based on the MODIS fire counts in croplands (Li et al., 2016). We assumed that there was no monthly variation for the emissions from the other sources (Wu and Xie, 2018)."

The monthly variability of the EF-based emission inventory obtained by this study was shown in Figure 12, which didn't exhibit obvious seasonal variations. The EF-based VOC emission inventories developed by the other studies (Li et al., 2017b; Wu and Xie, 2018) also didn't exhibit obvious seasonal variations. The discrepancies among seasons were very tiny because of little monthly variation in emissions from industrial processes, transportation, and solvent utilization (Wu and Xie, 2018). Monthly variations of the satellite-derived VOC emissions exhibit obvious seasonal characteristics, with the maximum in winter and minimum in summer, which are consistent with the seasonal characteristics of the ambient VOC mixing ratios. Thus, the satellite-derived emission inventories may better reflect the monthly characteristics of VOC emissions.

In the revised manuscript, Figure 12 was revised and comparison between the monthly variations of the satellite derived emission inventory and EF-based emission inventory was added in section 3.4 as follows, "The temporal resolution of the satellite-derived emission inventory is one month. As shown in Fig. 12, monthly variations of VOC emissions exhibit obvious seasonal characteristics, with the maximum in winter and minimum in summer, which are consistent with the seasonal characteristics of the ambient VOC mixing ratios (Fig. 5). However, monthly profiles for the EF-based emission inventory, which developed based on monthly statistics, didn't exhibit seasonal variations. EF-based VOC emission inventories developed by the other studies (Li et al., 2017; Wu and Xie, 2018) also didn't exhibit obvious seasonal variations because of little monthly variation in emissions from transportation, industrial processes, and solvent utilization (Wu and Xie, 2018). The satellite-derived emission inventories may better reflect the monthly characteristics of VOC emissions and be used to allocate monthly emissions". In addition, the subtitle of this part was changed to "Verification of spatial and temporal distributions."

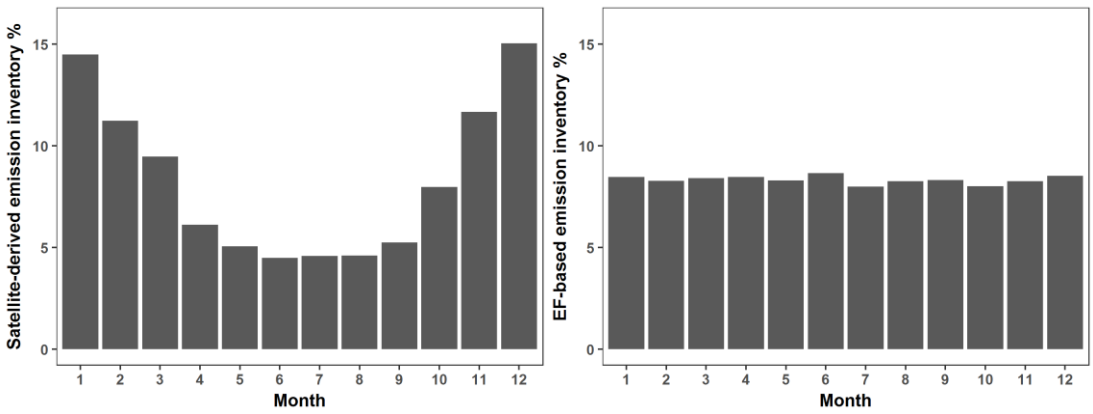


Figure 12. Monthly variability of VOC emissions from the satellite-derived and EF-based emission inventories.

References:

Guo, J., Huang, Y., and Wei, C.: North–South debate on district heating: Evidence from a household survey, Energ. Policy, 86, 295-302, 10.1016/j.enpol.2015.07.017, 2015.

Li, J., Li, Y., Bo, Y., and Xie, S.: High-resolution historical emission inventories of crop residue burning in fields in China for the period 1990–2013, Atmos. Environ., 138, 152-161, 10.1016/j.atmosenv.2016.05.002, 2016.

Li, M., Zhang, Q., Kurokawa, J.-i., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos. Chem. Phys., 17, 935-963, 10.5194/acp-17-935-2017, 2017b.

NBS: China Economic Statistics Express, 2015.

Wu, R., and Xie, S.: Spatial Distribution of Secondary Organic Aerosol Formation Potential in China Derived from Speciated Anthropogenic Volatile Organic Compound Emissions, Environ. Sci. Technol., 52, 8146-8156, 10.1021/acs.est.8b01269, 2018.

Zheng, J., Zhang, L., Che, W., Zheng, Z., and Yin, S.: A highly resolved temporal and spatial air pollutant emission inventory for the Pearl River Delta region, China and its uncertainty assessment, Atmos. Environ., 43, 5112-5122, 10.1016/j.atmosenv.2009.04.060, 2009.

Zheng, X., Wei, C., Qin, P., Guo, J., Yu, Y., Song, F., and Chen, Z.: Characteristics of residential energy consumption in China: Findings from a household survey, Energ. Policy, 75, 126-135, 10.1016/j.enpol.2014.07.016, 2014.

3. Can you please add comparison between source structure from PMF analysis and that from your emission inventory for each season? As there might be a seasonal variability in source structure of your emission inventory.

Response: Accepted. Thank you for your suggestion. As we described in the response to your General Comment No.2, monthly profiles for EF-based emission inventories usually didn't exhibit seasonal variations.

In the revised manuscript, we added comparison between source structure from PMF analysis and that from our emission inventory for each season in Figure 9, and the following sentences were added in section 3.3.2 "Compared with the seasonal PMF results, the emissions from industrial processes, transportation, and solvent utilization of the emission inventory didn't exhibit obvious seasonal variations (Fig. 9). It is because the monthly profiles of these sources, which developed on monthly statistics, have little monthly variations (Wu and Xie, 2018). The emissions from fuel combustion of the emission inventory exhibit similar seasonal variations with the PMF results with much higher emissions in winter than the other seasons. However, the relative contribution of fuel combustion for each season in the emission inventory was significantly lower than the contribution in the PMF results, especially for winter. On the basis of the above comparisons of the VOC source structure, we inferred that: (1) the annual contributions of the vehicles, solvent utilization, and industrial processes from the emission inventory and the PMF results were similar, but the monthly profiles of these sources cannot replicate the temporal variations; and (2) the fuel combustion in the emission inventory showed a considerably lower relative contribution than the value from the PMF analysis, especially in winter, the central heating season in Beijing."

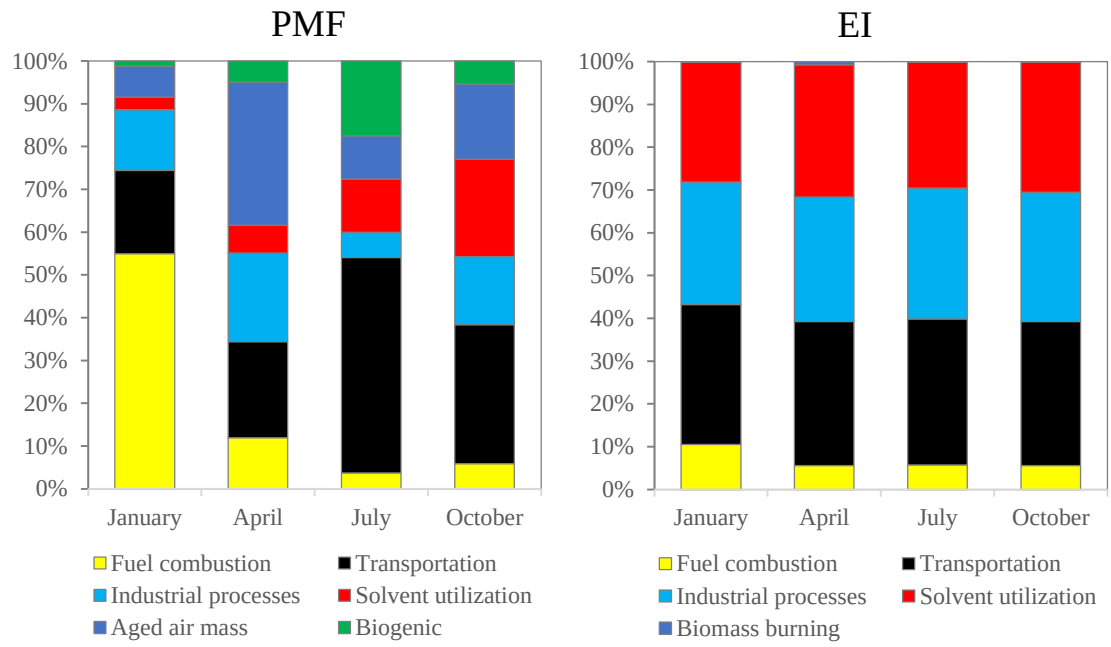


Figure 9.Monthly VOC source structure identified by the PMF analysis (left) and the emission inventory (right).

Specific Comments:

1. Line 33: "Their direct emission sources include biogenic and anthropogenic sources". Forest fire emissions are also worth to mention here. In addition, please include some appropriate citations here.

Response: Accepted. Thank you for your suggestion. Forest fire emissions were mentioned and some appropriate citations were added here. In the revised manuscript, this sentence was revised to "Their direct emission sources include biogenic sources, forest fires, and anthropogenic sources (Guenther et al., 2006; Kansal et al., 2009; Simpson et al., 2011).

References:

Guenther, A., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181-3210, 2006.

Kansal, A.: Sources and reactivity of NMHCs and VOCs in the atmosphere: a review, J. Hazard. Mater., 166, 17-26, 10.1016/j.jhazmat.2008.11.048, 2009.

Simpson, I. J., Akagi, S. K., Barletta, B., Blake, N. J., Choi, Y., Diskin, G. S., Fried, A., Fuelberg, H. E., Meinardi, S., Rowland, F. S., Vay, S. A., Weinheimer, A. J., Wennberg, P. O., Wiebring, P., Wisthaler, A., Yang, M., Yokelson, R. J., and Blake, D. R.: Boreal forest fire emissions in fresh Canadian smoke plumes: C1-C10 volatile organic compounds (VOCs), CO2, CO, NO2, NO, HCN and CH3CN, Atmos. Chem. Phys., 11, 6445-6463, 10.5194/acp-11-6445-2011, 2011.

2. Line 42-46: Zhang et al. (2009) and Li et al. (2014) have done a lot work in compiling anthropogenic VOCs emissions over China. They are also worth to cite here.

Response: Accepted. These studies were cited here.

3. Line 63-66: Both Karplus et al. (2018) and Henne et al. (2016) are not appropriate references here, because Karplus et al. (2018) talks about SO2 and Henne et al. (2016) talks about methane. Please include some NMVOC-related references here.

Response: Accepted. We are sorry for the inappropriate references. Fu et al. (2007) constrained the NMVOC emissions from multiple sources over East and South Asia by analyzing the spatiotemporal variability in the observed formaldehyde columns. Cao et al. (2018) used satellite retrievals along with a chemical transport model to constrain NMVOC emissions from China.

In the revised manuscript, the sentence was revised to "Since the satellite data possess the advantage of reflecting the spatial characteristics of VOCs (Fu et al., 2007), satellite-derived anthropogenic VOC emission estimations obtained from the chemical transport model can be utilized to evaluate the spatial distribution of the EF based emission inventories (Cao et al., 2018)".

References:

Cao, H., Fu, T.-M., Zhang, L., Henze, D. K., Miller, C. C., Lerot, C., Abad, G. G., De Smedt, I., Zhang, Q., van Roozendael, M., Hendrick, F., Chance, K., Li, J., Zheng, J., and Zhao, Y.: Adjoint inversion of Chinese non-methane volatile organic compound emissions using space-based observations of formaldehyde and glyoxal, Atmos. Chem. Phys., 18, 15017-15046, 10.5194/acp-18-15017-2018, 2018.

Fu, T.-M., Jacob, D. J., Palmer, P. I., Chance, K., Wang, Y. X., Barletta, B., Blake, D. R., Stanton, J. C., and Pilling, M. J.: Space-based formaldehyde measurements as constraints on volatile organic compound emissions in east and south Asia and implications for ozone, J. Geophys. Res., 112, 10.1029/2006jd007853, 2007.

4. Line 67-69: Please include some references here to denote "Earlier studies" and "most studies".

Response: Accepted. Gaimoz et al. (2011) evaluated the source structure of VOC emission inventories by source appointment with one month ambient measurements; Borbon et al. (2013) and Wang et al. (2014) evaluated the species-specific emissions by the emission ratios to an inert tracer; Cao et al. (2018) evaluate the spatial distribution by satellite retrievals. Gaimoz et al. (2011), Borbon et al. (2013) and Wang et al (2014) were based on the data from one, one, and two-month ambient measurements, respectively. In the revised manuscript, the references to denote "Earlier studies" and "most studies" were added as follows, "Earlier studies by various research groups applied only one of these methods to evaluate either the source structure or species-specific emissions of VOC emission inventories (Gaimoz et al.,2011; Borbon et al., 2013; Wang et al. 2014; Cao et al., 2018). Moreover, most studies have been based on the data from one or two-month ambient measurements, which cannot accurately represent the annual emissions (Gaimoz et al., 2011; Borbon et al., 2013; Wang et al. 2014)".

References:

Borbon, A., Gilman, J. B., Kuster, W. C., Grand, N., Chevaillier, S., Colomb, A., Dolgorouky, C., Gros, V., Lopez, M., Sarda-Esteve, R., Holloway, J., Stutz, J., Petetin, H., McKeen, S., Beekmann, M., Warneke, C., Parrish, D. D., and de Gouw, J. A.: Emission ratios of anthropogenic volatile organic compounds in northern mid-latitude megacities: Observations versus emission inventories in Los Angeles and Paris, J. Geophys. Res.-Atmos., 118, 2041-2057, 10.1002/jgrd.50059, 2013.

Cao, H., Fu, T.-M., Zhang, L., Henze, D. K., Miller, C. C., Lerot, C., Abad, G. G., De Smedt, I., Zhang, Q., van Roozendael, M., Hendrick, F., Chance, K., Li, J., Zheng, J., and Zhao, Y.: Adjoint inversion of Chinese non-methane volatile organic compound emissions using space-based observations of formaldehyde and glyoxal, Atmos. Chem. Phys., 18, 15017-15046, 10.5194/acp-18-15017-2018, 2018.

Gaimoz, C., Sauvage, S., Gros, V., Herrmann, F., Williams, J., Locoge, N., Perrussel, O., Bonsang, B., d'Argouges, O., Sarda-Estève, R., and Sciare, J.: Volatile organic compounds sources in Paris in spring 2007. Part II: source apportionment using positive matrix factorisation, Environ. Chem., 8, 91, 10.1071/en10067, 2011.

Wang, M., Shao, M., Chen, W., Yuan, B., Lu, S., Zhang, Q., Zeng, L., and Wang, Q.: A temporally and spatially resolved validation of emission inventories by measurements of ambient volatile organic compounds in Beijing, China, Atmos. Chem. Phys., 14, 5871-5891, 10.5194/acp-14-5871-2014, 2014.

5. Line 105: what is "COPERT 4" short for? Please give a full name of this software when you first mention it.

Response: Accepted. "COPERT" is short for "COmputer Programme to calculate Emissions from Road Transport". In the revised manuscript, the full name of the software was added as "EFs were calculated by COmputer Programme to calculate Emissions from Road Transport version 4 (COPERT 4)".

6. Line 122-123: Where did "county-level", "city-level", and "provincial-level" data come from? Please include corresponding references here.

Response: Accepted. We are sorry for the unclear descriptions. In the revised manuscript, a table containing activity data and corresponding reference for each source and for each category was added in the supplementary information (Table S2).

7. Line 132: Please give the source profile of Wu and Xie (2017), you can put it in the supplementary information.

Response: Accepted. The source profile used in this study was added in the supplementary information (Table S4). In the revised manuscript, "The source profile database used in this study was listed in Table S4." was added.

8. Line 137: Please also give the height of roof site.

Response: Accepted. In the revised manuscript, the height of the roof site was added as follow: "The roof of the Technical Physics Building at Peking University, with a height of approximately 15 m above the ground (PKU, 39.99 °N, 116.33 °E, Fig. 1) was selected as the sampling site".

9. Line 169-170: Please give the emission ratios of VOC species relative to CO you obtained from the linear fit model.

Response: Accepted. The emission ratios of VOC species relative to CO obtained from the linear fit model were listed in Table S5.

10. Line 171-172: Please include references to support "(1) CO has similar sources as that of anthropogenic VOC and (2) CO emissions show lower uncertainty compared with VOC emissions".

Response: Accepted. Thank you for your suggestion. A lot of studies have chosen CO as the reference compound to study the emission ratios for many cities in the world (Warneke et al., 2007; Coll et al., 2010; Borbon et al., 2013; Wang et al., 2014). Wang et al. (2014) calculated the emission ratios for Beijing and described that CO has similar sources as that of anthropogenic VOC in Beijing and the uncertainty of CO emissions are lower than that of VOC emissions in detailed. In the revised manuscript, references were added as follows "In this study, we selected CO as a reference compound considering that: (1) CO has similar sources as that of anthropogenic VOC and (2) CO emissions show lower uncertainty compared with VOC emissions (Warneke et al., 2007; Wang et al., 2014). Thus, CO was a suitable reference compound (Coll et al., 2010;Borbon et al., 2013;Wang et al., 2014)".

References:

Borbon, A., Gilman, J. B., Kuster, W. C., Grand, N., Chevaillier, S., Colomb, A., Dolgorouky, C., Gros, V., Lopez, M., Sarda-Esteve, R., Holloway, J., Stutz, J., Petetin, H., McKeen, S., Beekmann, M., Warneke, C., Parrish, D. D., and de Gouw, J. A.: Emission ratios of anthropogenic volatile organic compounds in northern mid-latitude megacities: Observations versus emission inventories in Los Angeles and Paris, J. Geophys. Res.-Atmos., 118, 2041-2057, 10.1002/jgrd.50059, 2013.

Wang, M., Shao, M., Chen, W., Yuan, B., Lu, S., Zhang, Q., Zeng, L., and Wang, Q.: A temporally and spatially resolved validation of emission inventories by measurements of ambient volatile organic compounds in Beijing, China, Atmos. Chem. Phys., 14, 5871-5891, 10.5194/acp-14-5871-2014, 2014.

Coll, I., Rousseau, C., Barletta, B., Meinardi, S., and Blake, D. R.: Evaluation of an urban NMHC emission inventory by measurements and impact on CTM results, Atmos. Environ., 44, 3843-3855, 10.1016/j.atmosenv.2010.05.042, 2010.

Warneke, C., McKeen, S. A., de Gouw, J. A., Goldan, P. D., Kuster, W. C., Holloway, J. S., Williams, E. J., Lerner, B. M., Parrish, D. D., Trainer, M., Fehsenfeld, F. C., Kato, S., Atlas, E. L., Baker, A., and Blake, D. R.: Determination of urban volatile organic compound emission ratios and comparison with an emissions database, J. Geophys. Res., 112, 10.1029/2006jd007930, 2007.

11. Line 177: Please include appropriate references on validation of CO emission inventory of MarcoPolo Project.

Response: Accepted. Thank you for your suggestion. MarcoPolo project is a collaborative research project by Chinese and European partners to study the emission sources using state-of-the-art techniques and to provide the latest air up-to-data emission inventories for China. For the CO emission inventory, MarcoPolo project copied the Multi-resolution Emission Inventory for China (MEIC) emissions (Hooyberghs et al., 2016). The MEIC is a uniform emission model framework developed by Tsinghua University to estimate anthropogenic emissions over China (http://www.meicmodel.org/index.html). The CO emission inventory of MEIC has been validated by the chemical transport model (Hu et al., 2017), satellite observations (Yumimoto et al. 2014), and comparison with other studies (Li et al., 2017a).

In the revised manuscript, appropriate references was added as follows: "The annual emission value of CO was obtained from the CO emission inventory of the MarcoPolo Project (http://www.marcopolo-panda.eu, Hooyberghs et al., 2016), which was copied from the Multi-resolution Emission Inventory for China (MEIC) emissions (http://www.meicmodel.org/index.html). This emission inventory has been validated by the chemical transport model (Hu et al., 2017), satellite observations (Yumimoto et al. 2014), and comparison with other studies (Li et al., 2017a)".

References:

Hooyberghs, H., Veldeman, N., and Maiheu, B.: Marco Polo Emission Inventory for East-China: Basic Description, 2016.

Hu, J., Li, X., Huang, L., Ying, Q., Zhang, Q., Zhao, B., Wang, S., and Zhang, H.: Ensemble prediction of air quality using the WRF/CMAQ model system for health effect studies in China, Atmos. Chem. Phys., 17, 13103-13118, 10.5194/acp-17-13103-2017, 2017.

Li, M., Liu, H., Geng, G., Hong, C., Liu, F., Song, Y., Tong, D., Zheng, B., Cui, H., Man, H., Zhang, Q., and He, K.: Anthropogenic emission inventories in China: a review, National Science Review, 4, 834-866, 10.1093/nsr/nwx150, 2017a.

Yumimoto, K., Uno, I., and Itahashi, S.: Long-term inverse modeling of Chinese CO emission from satellite observations, Environ. Pollut., 195, 308-318, 10.1016/j.envpol.2014.07.026, 2014.

12. Line 186-187: Can you please give a brief description (or formulas) on how your sampled VOCs uncertainties were calculated?

Response: Accepted. The observed uncertainty file was set following the method proposed by Polissar et al. (1998), which was recommended by the user guide of the PMF model. In the revised manuscript, the description was added as follows: "The observed uncertainty file was set following the method proposed by Polissar et al. (1998), which was recommended by the user guide of the PMF model. The uncertainty is calculated by Eq. (3), if the mixing ratio is equal to or less than the MDL; the uncertainty is calculated using Eq. (4), if mixing ratio if larger than the MDL (USEPA, 2014a).

$$\text{Uncertainty} = \frac{5}{6} \times MDL \qquad Eq.(3)$$

$$\text{Uncertainty} = \sqrt{(Error\ Fraction\ \times mixing\ ratio)^2 + (0.5\ \times MDL)^2} \qquad Eq.(4)$$

References:

Polissar, A. V., Hopke, P. K., Paatero, P., Malm, W. C., and Sisler, J. F.: Atmospheric aerosol over Alaska: 2. Elemental composition and sources, J. Geophys. Res.-Atmos., 103, 19045-19057, 10.1029/98jd01212, 1998.

USEPA: Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide, US Environmental Protection Agency, Office of Research and Development, Washington, D.C, 2014a.

13. Line 198: ". . .limited. . ." should be ". . .constrained. . ."
Response: Accepted. "limited" was changed to "constrained".

14. Line 199-200: "HCHO is a high-yield product of VOCs species oxidation" should be "HCHO is a high-yield product of many VOCs species oxidation". Also, please include some appropriate references to support this sentence. Such as Millet et al. (2006), Stavrakou et al. (2015).
Response: Accepted. "HCHO is a high-yield product of VOCs species oxidation" was revised to "HCHO is a high-yield product of many VOCs species oxidation". And some appropriate references such as Millet et al. (2006), Stavrakou et al. (2015) were added.

15. Line 200: ". . .relative to. . ." should be ". . .against. . ."
Response: Accepted. "relative to" was changed to "against".

16. Line 235: ". . .in January, with an average value of 62.26 ppbv"? But figure 5 shows that the January average was less than 60 ppbv.
Response: Accepted. Thank you for your comment. Figure 5 does not show the distribution of the data well. The following box-plot shows the distributions of the VOC mixing ratios in January, April, July, and October 2015. As shown in this figure, the January average was larger than 60 ppbv (red dot). In the revised manuscript, the box-plot was added in the Supplementary Information as Fig. S4.

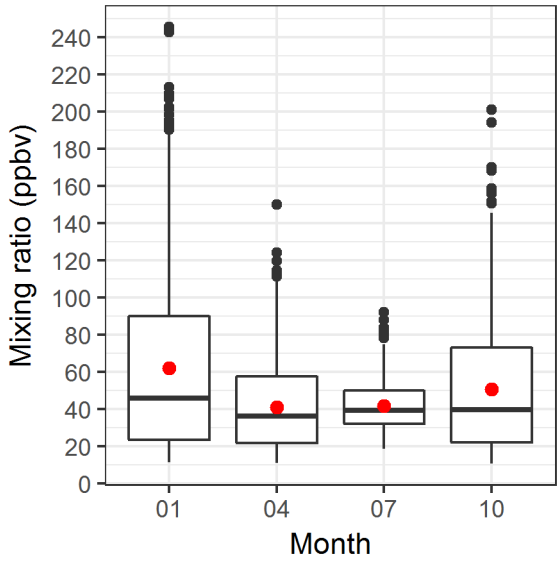


Figure S4. Box-plot of VOC mixing ratios in in January, April, July, and October 2015.

17. Line 237: "In October, the average mixing ratio of VOCs was 50.64 ppbv". Again, the October average in figure 5 seems to be less than 50 ppbv.

Response: Accepted. Please refer to the figure shown in the response to the **specific comment #16**. The October average was larger than 50 ppbv (red dot).

18. Line 246: "The VOCs accumulated in early October decreased sharply in the middle of the month, then began to accumulate again with the change of the diffusion condition". Can you please provide some meteorological analysis or some references to support this sentence?

Response: Accepted. Thank you for your valuable comment. Time series of wind speed and VOC mixing ratios in October 2015 was shown in the following figure, which can support this sentence. In early October, the VOCs accumulated when the wind speed was low. Then VOCs decreased sharply when the wind speed became higher. And the VOC began to accumulate again with change of the wind speed. According to the comment of the other review, this paragraph is meaningless in this manuscript. The point of using the observations is to validate the inventory and this section should be concise.

Combined with reviewers' comments, in the revised manuscript, the second paragraph of section 3.2.1 and Figure 6 was moved to the Supplementary Information. The paragraph was revised to "Figure S2 presents the time series of VOC mixing ratios. The mixing ratios of VOCs in January were variable, with maximum value of 245.54 ppbv. There were lots of periods with high VOC mixing ratios in January. In April, the average VOC mixing ratio was not as high as in January but the mixing ratios of VOCs change a lot, a maximum value of 150.24 ppbv. The mixing ratios of VOCs in July were stable, with the highest level of 92.28 ppbv. The highest VOC mixing ratio in October was 201.10 ppbv. In early October, the VOCs accumulated when the wind speed was low (Fig. S3). Then VOCs decreased sharply when the wind speed became higher. And the VOC began to accumulate again with change of the wind speed"

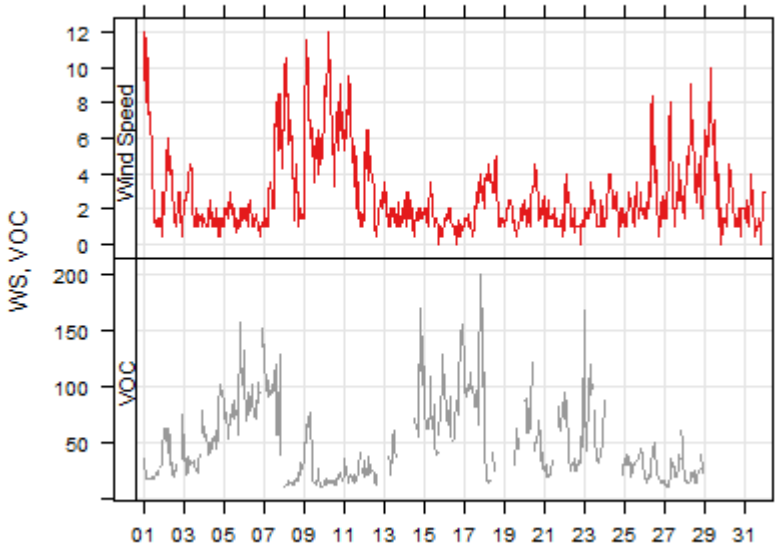


Figure S3. Time series of wind speed and VOC mixing ratios in October 2015.

19. Line 265: ". . .a few different emission sources from CO" should be ". . .a few emission sources different from CO sources"

Response: Accepted. "a few different emission sources from CO" was changed to "a few emission sources different from CO sources".

20. Line 275-277: "After the comparison with results obtained from measurements, the emissions for a majority of the non-methane hydrocarbon (NMHC) species were agreed within ±100% in the emission inventory". Can you please quantify the "a majority"?

Response: Accepted. There are 56 NMHC species measured by the GCMS/FID system in this study. The emissions for 51 NMHC species were agreed within ±100% in the emission inventory. In the revised manuscript, this sentence was revised to "the emissions for 51 of 56 kinds of non-methane hydrocarbon (NMHC) species were agreed within ±100% in the emission inventory, 15 species agreed within ±50%, and 10 species agreed within ±25%".

21. Line 277: "The emissions for acetonitrile came from the two methods were similar" doesn't seem to be supported by the acetonitrile emissions values (0.21 Ton yr-1 vs 16.52 Ton yr-1) listed in Table S2.

Response: Accepted. We are sorry for the mistake in Table S2. 0.21 was the ER value for acetonitrile instead of the emissions determined by the measurements (ER method). As shown in Fig. 6 and Fig.7, the acetonitrile emission value determined by the measurements be 106.7 Ton yr^{-1}. The acetonitrile emissions determined by the measurements (106.7 Ton yr^{-1}) is around 5 times the acetonitrile emissions determined by the emission inventory (16.52 Ton yr-1).

In the revised manuscript, Table S2 is corrected. In addition, this sentence was revised to "The acetonitrile emissions determined by the measurements is around 5 times the acetonitrile emissions determined by the emission inventory".

22. Line 282-283: "The annual emissions for alkanes were in agreement between the two methods". Can you please quantify the "agreement"? As the detailed emission values of many alkanes in Table S2 have large difference between these two methods.

Response: Accepted. It is not appropriate to use "agreement" here. There are 27 alkane species measured by the GCMS/FID system in this study. The emissions for 25 alkane species were agreed within ±100% in the emission inventory, 8 species were agreed within ±50%, and 6 species were agreed within ±25%.

In the revised manuscript, this sentence was revised to "the annual emissions for alkanes were agreed within ±100% between the two methods, except ethane and propane, which are important tracers of natural gas and LPG".

23. Line 287: "The annual emissions for the alkenes, except ethene, correlated well". Again, please quantitatively state the "correlate well".

Response: Accepted. It is not appropriate to use "correlated well" here.In the revised manuscript, this sentence was revised to "The annual emissions for the alkenes, except ethene, were agreed within ±100%".

24. Line 288-289: "Ethene and acetylene are mainly emitted through an incomplete combustion process". Please include appropriate references for this sentence.

Response: Accepted. We are sorry for the lack of references. Liu et al. (2008) experimentally determined the profiles of major VOC sources in China, and Mo et al. (2016) complied a database of VOC profiles for 75 species in China by measuring and available literature. Both of them indicated that ethene and acetylene are mainly emitted through an incomplete combustion process. In the revised manuscript, the references were added.

References:

Liu, Y., Shao, M., Fu, L. L., Lu, S. H., Zeng, L. M., and Tang, D. G.: Source profiles of volatile organic compounds (VOCs) measured in China: Part I, Atmos. Environ., 42, 6247-6260, 10.1016/j.atmosenv.2008.01.070, 2008.

Mo, Z., Shao, M., and Lu, S.: Compilation of a source profile database for hydrocarbon and OVOC emissions in China, Atmos. Environ., 143, 209-217, 10.1016/j.atmosenv.2016.08.025, 2016.

25. Line 293-297: What's the local VOC emission standards? Can you please give a reference?

Response: Accepted. We are sorry for the lack of references. Beijing had issued local VOC emission standards for many solvent utilization sources since 2015. These emission standards included "Emission standard of volatile organic compounds for printing industry (DB11/1201-2015)","Emission standard of air pollutants for wood furniture (DB11/1202-2015)", "Emission standard of air pollutants for industrial surface coating (DB11/1226-2015)", "Emission standard of air pollutants for painting process of automobile manufacturing industry (DB11/1227-2015)", and "Emission standard of air pollutants for vehicle maintenance and repair industry (BD11/1228-2015)". In the revised manuscript, the code names for these emission standards were added "The common character of these sources is that Beijing had issued local VOC emission standards for the above sources since 2015 (DB11/1201-2015; DB11/1202-2015; DB11/1226-2015; DB11/1227-2015;

BD11/1228-2015)".

26. Line 303: ". . .reaction. . ." should be "reactivity".
Response: Accepted. "reaction" was changed to "reactivity".

27. Line 316: "appointment" should be "apportionment"?
Response: Accepted. "appointment" was changed to "apportionment".

28. Line 323-335: Can you please add some comparison between your PMF analysis with PMF analysis from other studies during these seasons?
Response: Accepted. Thank you for your suggestion. We compared the PMF result in this study with PMF analysis from other studies conducted in Beijing during these seasons.

In the revised manuscript, Comparison of the relative contributions of sources of VOC emissions in Beijing calculated by the PMF model in this study and results from the other studies was added in Table S7 of the Supplementary Information. Following descriptions were added in section 3.3.1 "Comparison of the relative contributions of VOC emission sources in Beijing calculated by the PMF model of this study and results from the other studies during these seasons was listed in Table S7. Results of this and other studies have shown that the fuel combustion was the largest VOC contributor in winter. The contribution proportions of fuel combustion in winter were ranged from 45% - 55% (Li et al., 2015, Yang et al., 2018). Results of this and other studies have shown that vehicle-related source was the largest VOC contributor in summer and winter, with the contribution ranged from 50% - 57%, and 33% - 42%, respectively. The contribution proportion of summer biogenic emission in this study was larger than that in the other studies".

Table S7. Comparison of the relative contributions of sources of VOC emissions in Beijing calculated by the PMF model in this study and results from the other studies.

Reference	Sampling period			Site type	Source categories					
	Year	Period	Season		Vehicle	Industrial	Solvent	Fuel	Biogenic	Others [a]
This study	2015	April	Spring	Urban	22%	21%	7%	12%	5%	33%
This study	2015	July	Summer	Urban	50%	6%	12%	4%	18%	10%
Song et al.,2007	2005	1–26 August	Summer	Urban	55%	20%	5%	5%	2%	11%
Li et al.,2016	2015	11–19 August	Summer	Urban	57%	4%	14%	10%	1%	14%
Yuan et al., 2009	2006	15 August to 10 September	Summer to Autumn	Urban	62%		16%	6%	3%	13%
Yuan et al., 2009	2006	15 August to 10 September	Summer to Autumn	Rural	39%		14%	3%	8%	37%
This study	2015	October	Autumn	Urban	33%	16%	23%	6%	5%	18%
Wu et al., 2016	2014	1–15 October	Autumn	Urban	49%	11%	9%	22%		9%
Li et al.,2015	2014	18–31 October	Autumn	Urban	43%	22%	12%	9%		14%
Yang et al.,2018	2014	25 October–2 November	Autumn	suburban	42%	14%	22%	22%		0%
This study	2015	January	Winter	Urban	19%	14%	3%	55%	1%	7%
Li et al.,2015	2014	13–22 November	Winter	Urban	20%	14%	11%	45%		10%
Yang et al.,2018	2014	13 November–13 December	Winter	suburban	17%	25%	13%	45%		0%

[a] The source categories of different PMF studies are different. Except for some comment categories (vehicle, industrial processes, solvent utilization, fuel combustion, and biogenic), there are some other categories such as aged air mass, long-lived species, biomass burning,

background, secondary formation, LPG, NG, which were defined as others in Table S7.

References:

Li, J., Wu, R., Li, Y., Hao, Y., Xie, S., and Zeng, L.: Effects of rigorous emission controls on reducing ambient volatile organic compounds in Beijing, China, Sci. Total. Environ., 557-558, 531-541, 10.1016/j.scitotenv.2016.03.140, 2016.

Li, J., Xie, S. D., Zeng, L. M., Li, L. Y., Li, Y. Q., and Wu, R. R.: Characterization of ambient volatile organic compounds and their sources in Beijing, before, during, and after Asia-Pacific Economic Cooperation China 2014, Atmos. Chem. Phys., 15, 7945-7959, 10.5194/acp-15-7945-2015, 2015.

Song, Y., Shao, M., Liu, Y., Lu, S. H., Kuster, W., Goldan, P., and Xie, S. D.: Source apportionment of ambient volatile organic compounds in Beijing, Environ. Sci. Technol., 41, 4348-4353, 10.1021/es0625982, 2007.

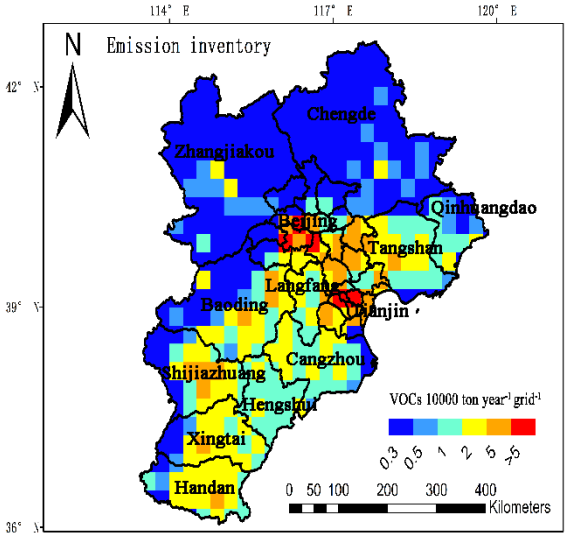
Wu, R., Li, J., Hao, Y., Li, Y., Zeng, L., and Xie, S.: Evolution process and sources of ambient volatile organic compounds during a severe haze event in Beijing, China, Sci. Total. Environ., 560-561, 62-72, 10.1016/j.scitotenv.2016.04.030, 2016.

Yang, W., Zhang, Y., Wang, X., Li, S., Zhu, M., Yu, Q., Li, G., Huang, Z., Zhang, H., Wu, Z., Song, W., Tan, J., and Shao, M.: Volatile organic compounds at a rural site in Beijing: influence of temporary emission control and wintertime heating, Atmos. Chem. Phys., 18, 12663-12682, 10.5194/acp-18-12663-2018, 2018.

Yuan, Z., Lau, A. K. H., Shao, M., Louie, P. K. K., Liu, S. C., and Zhu, T.: Source analysis of volatile organic compounds by positive matrix factorization in urban and rural environments in Beijing, J. Geophys. Res., 114, 10.1029/2008jd011190, 2009.

29. Line 384: Can you please make markers of Xingtai and Handan on the maps of figure 13?

Response: Accepted. The name of each city was added in figure 13 as follows:



30. Line 386-391: The subtitle of this part is "Verification of spatial distribution", but here you are discussing the monthly variabilities of satellite-derived emissions and another bottom-up emission inventory (Li et al., 2017) at length. It seems a little odd. If you want to discuss monthly variation, can you please provide the comparison between your monthly VOCs emissions and satellite-derived monthly emissions? And also, please keep the subtitle consistent with your text.

Response: Accepted. Thank you for your suggestion. Because EF-based VOC emission inventories usually didn't exhibit obvious seasonal variations for little monthly variation in monthly statistics (Li et al., 2017; Wu and Xie, 2018). So we discussing the monthly variabilities of satellite-derived emissions and another bottom-up emission inventory (Li et al., 2017). I agreed it seems odd.

In the revised manuscript, we compared the monthly profiles between the satellite-derived emission inventory and our EF-based emission inventory in Figure 12. And the subtitle of this part was changed to "Verification of spatial and temporal distributions". For the detailed method and comparison about the monthly distribution please refer to the response to the **General Comment #2**.

31. Line 392-393: Again, Geng et al. (2017) talks about NO2 and NOx emissions. Can you please include some appropriate references on top-down VOCs emissions and OVOC satellite observations? Such as Palmer et al. (2006), Fu et al. (2007), Marais et al. (2014), Stavrakou et al. (2015), Bauwens et al. (2016).

Response: Accepted. Thanks a lot for providing these appropriate references. In the revised manuscript, these references were added as follows: "The satellite-derived emission inventory possesses the advantage of efficiently reflecting the spatial and monthly characteristics of the VOC emissions (Palmer et al., 2006; Fu et al., 2007;Marais et al., 2014;Stavrakou et al., 2015;Bauwens et al., 2016).".

32. Line 407-408: Again, can you please quantify the consistence between the NMHCs emissions derived from online measurements and those from your emission inventory?

Response: Accepted. In the revised manuscript, it was revised to "the annual emissions of 91% NMHCs derived from the measurements were agreed within ±100% with the results of the emission inventory".

33. Table 1.: What are "1,1,2,2-" and "1,2-" in the last 3 rows?

Response: Accepted. We are sorry for the wrong format of Table 1. In the revised manuscript, Table 1 is corrected.

34. Figure 2.: The unit "Ton/grid" should be "Ton year^{-1} grid^{-1}".

Response: Accepted. The unit "Ton/grid" was revised to "Ton year^{-1} grid^{-1}".

35. Figure 4.: What's the difference between "Other VOCs" and "Others" in the bar plot? What's "Other thenes"? Why is "Others" the largest contributor in bar plot, while it is the smallest one in the pie plot?

Response: Accepted. We are sorry for the misleading descriptions. VOCs consist thousands of compounds. We determined a unified species list for source profiles (Table S4). These species were measurable, abundant, or highly reactive in the atmosphere (Wu et al., 2017), which can be group into alkanes, alkenes, alkynes, aromatics, halocarbons, OVOCs, nitriles, and others.

The pie plot in Fig.4 showed the contributions of each VOC groups. Because the measured VOC species from identical sources typically varied among different studies, there are some VOC species didn't belong to the VOC groups (alkanes, alkenes, alkynes, aromatics, halocarbons, OVOCs, and nitriles). These species were group into **"Others" in the pie plot**. The name of species in the bar plot is consistent with the source profile database (Table S4). Because the space is limit, we only displayed the top 35 VOC species with the largest VOC emissions. The emission of all the other VOC (except the top 35 VOC species) added together was defined as emission from **"Others" in the bar plot**. Thus,

the “Others” is the largest contributor in bar plot, while it is the smallest one in the pie plot.

The name of top 35 VOC species in the bar plot is consistent with the species name in the source profile database. The source profile database consisted of 152 individual species in total. In the database, there are 16 abundant alkenes, and the other alkenes except the 16 abundant alkenes were defined as **“Other alkenes”**. We tried our best to determine VOC species according to available source profiles, however, there are some VOC species were defined as “unknown” or “other VOC” in some source profiles. So we defined the “unknown” or “other VOC” as **“Other VOCs”** in the database. In the revised manuscript, we changed the “Other VOCs” in the database to “Unspecific VOCs”.

In order to make it clearer, Figure 4 and related description were revised as follows: “Figure 4 illustrates the chemical compositions of VOC emissions in the BTH region. Emissions of a total of 152 VOC species (Table S4) bellowing to alkanes, alkenes, alkynes, aromatics, halocarbons, OVOCs, nitriles, and others were calculated in this study. The emissions of aromatics, alkanes, OVOCs, and alkenes accounted for 34%, 32%, 17%, and 11% of total anthropogenic VOC emissions, respectively.”

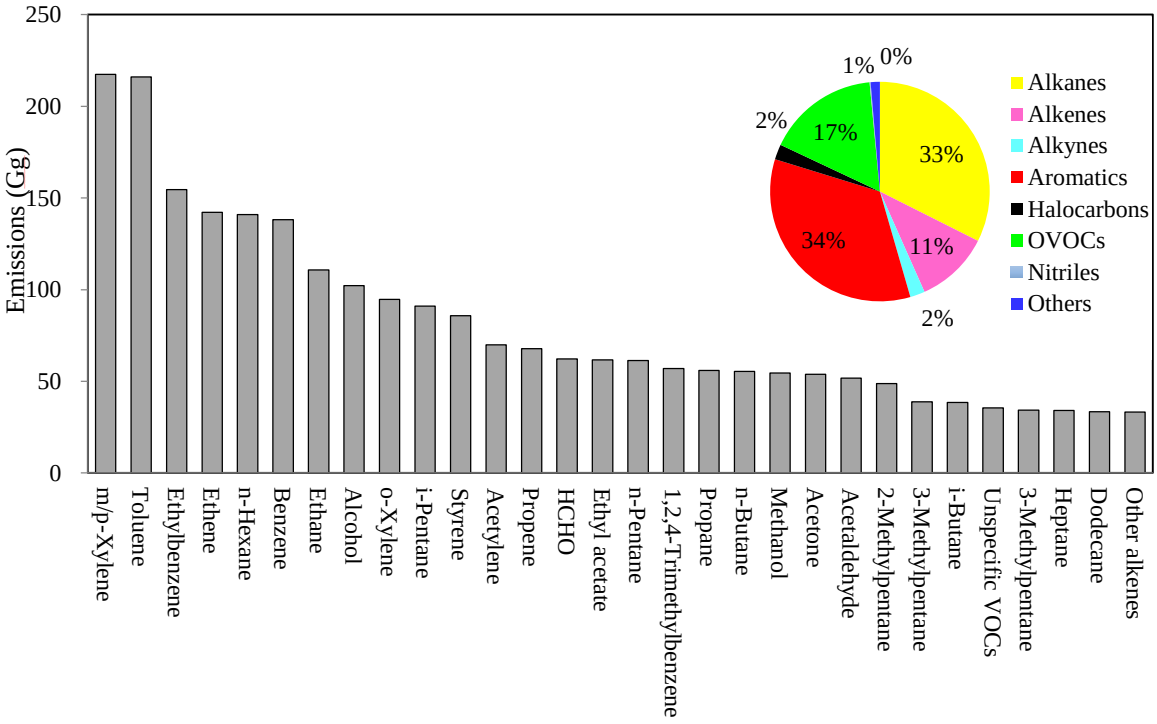


Figure 4. The top 30 VOC species with the highest emissions (bar plot) and the contributions of VOC groups to the total emsssions (pie plot) in the BTH region, China.

References:

Wu, R., and Xie, S.: Spatial Distribution of Ozone Formation in China Derived from Emissions of Speciated Volatile Organic Compounds, Environ. Sci. Technol., 51, 2574-2583, 10.1021/acs.est.6b03634, 2017.

36. Line 580-586: “. . .Sci Total Environ . . .” in line 582, and “. . .Journal Of Geophysical Research-Atmospheres. . .” in line 585. The formats of references are not unified. Please unify the reference format throughout the whole references.

Response: Accepted. The reference format throughout the whole references was checked and corrected.

Review References:

Bauwens, M., Stavrakou, T., Müller, J.-F., De Smedt, I., Van Roozendael, M., van der Werf, G. R., Wiedinmyer, C., Kaiser, J. W., Sindelarova, K., and Guenther, A.: Nine years of global hydrocarbon emissions based on source inversion of OMI formaldehyde observations, Atmos. Chem. Phys., 16, 10133-10158, https://doi.org/10.5194/acp-16- 10133-2016, 2016.

Fu, T.-M., Jacob, D. J., Palmer, P. I., Chance, K., Wang, Y. X., Barletta, B., Blake, D. R., Stanton, J. C., and Pilling, M. J.: Space-based formaldehyde measurements as constraints on volatile organic compound emissions in east and south Asia and implications for ozone, J. Geophys. Res., 112, D06312,https://doi.org/10.1029/2006jd007853, 2007.

Li, M., Zhang, Q., Streets, D. G., He, K. B., Cheng, Y. F., Emmons, L. K., Huo, H., Kang, S. C., Lu, Z., Shao, M., Su, H., Yu, X., and Zhang, Y.: Mapping Asian anthropogenic emis- sions of non-methane volatile organic compounds to multiple chemical mechanisms, Atmos. Chem. Phys., 14, 5617–5638, https://doi.org/10.5194/acp-14-5617-2014, 2014.

Marais, E. A., Jacob, D. J., Guenther, A., Chance, K., Kurosu, T. P., Murphy, J. G., Reeves, C. E., and Pye, H. O. T.: Improved model of isoprene emissions in Africa using Ozone Monitoring Instrument (OMI) satellite observations of formaldehyde: implications for oxidants and particulate matter, Atmos. Chem. Phys., 14, 7693-7703, https://doi.org/10.5194/acp-14-7693-2014, 2014

Millet, D. B., Jacob, D. J., Turquety, S., Hudman, R. C., Wu, S., Fried, A., Walega, J., Heikes, B. G., Blake, D. R., Singh, H. B., Anderson, B. E., and Clarke, A. D.: Formaldehyde distribution over North America: Implications for satellite retrievals of formaldehyde columns and isoprene emission, J. Geophys. Res., 111, D24S02,https://doi.org/10.1029/2005jd006853, 2006.

Palmer, P. I., Abbot, D. S., Fu, T.-M., Jacob, D. J., Chance, K., Kurosu, T. P., Guenther, A., Wiedinmyer, C., Stanton, J. C., Pilling, M. J., Pressley, S. N., Lamb, B., and Sumner, A. L.: Quantifying the seasonal and interannual variability of North American isoprene emissions using satellite observations of the formaldehyde column, J. Geophys. Res., 111, D12315, https://doi.org/10.1029/2005jd006689, 2006.

Stavrakou, T., Müller, J.-F., Bauwens, M., De Smedt, I., Van Roozendael, M., De Mazière, M., Vigouroux, C., Hendrick, F., George, M., Clerbaux, C., Coheur, P.-F., and Guenther, A.: How consistent are top-down hydrocarbon emissions based on formaldehyde observations from GOME-2 and OMI?, Atmos. Chem. Phys., 15, 11861–11884, https://doi.org/10.5194/acp-15- 11861-2015, 2015.

Zhang, Q., Streets, D. G., Carmichael, G. R., He, K. B., Huo, H., Kannari, A., Klimont, Z., Park, I. S., Reddy, S., Fu, J. S., Chen, D., Duan, L., Lei, Y., Wang, L. T., and Yao, Z. L.: Asian emis- sions in 2006 for the NASA INTEX-B mission, Atmos. Chem. Phys., 9, 5131–5153, https://doi.org/10.5194/acp-9-5131-2009, 2009.