Response to acp-2018-1192-RC1

We really appreciate your constructive comments and suggestions on our manuscript. Your positive advice help improve our manuscript. We have considered every comment carefully, and responded on a point to point and marked every change in red in the revised version.

1. The authors use HCHO from satellite and primary tracer CO and secondary tracer O3 from ground in-situ measurements to explore the primary and secondary source of HCHO. HCHO data is column density while O_3 and CO are concentrations at the surface. How the authors link the two different data to determine the source of HCHO. Why not use O3 and CO data from satellite?

Responses: Thank you very much for this suggestion. HCHO column was converted to the mixing ratio at the surface using the formula (3) in the manuscript, and detailed information was described in sec 3.1. CO can also be observed by Atmospheric InfraRed Sounder (AIRS) on board satellite EOS-Aqua and Tropospheric Emission Spectrometer (TES) on board satellite EOS-Aura, close in time to OMPS observation. However, the global coverage of TES is significantly lower and the AIRS instrument is degrading (Worden et al., 2013). O_3 and CO concentrations at the surface were measured simultaneously from China National Environmental Monitoring Center (CNEMC) Network in long time series from 2014. So we use O3 and CO data at the surface from CNEMC Network instead of data from satellite to explore the sources of HCHO.

Changes in manuscript: L33-34, P3 to L1-3, P4 in the revised version: "Due to low global coverage of Tropospheric Emission Spectrometer (Aura-TES) (Luo et al., 2002; Luo et al., 2007) and increasing noise after 2004 of Atmospheric InfraRed Sounder (Aqua-AIRS) (Pagano et al., 2012) which measure tropospheric CO closely in time to OMPS observation, obtaining precise CO concentration in the YRD after 2013 from satellite was limited. Therefore, CO and O3 concentrations at the surface monitored by the China National Environmental Monitoring Center (CNEMC) Network simultaneously were used in this study."

2. Section 3.1: The authors compared the OMPS data with FTS data, but did not illustrate why to compare the two different dataset.

Responses: Thank you very much for this suggestion. The global Network for Detection of Atmospheric Composition Change (NDACC) provides high quality, consistent, and standardized measurements of trace gases using FTS instruments, and its retrieval results were widely used for satellite observation. The ground-based FTS instrument used in this study is a candidate site for NDACC, and long-term measurements of HCHO by FTS were performed. So HCHO concentration measured by FTS was used to validate OMPS HCHO observation.

Changes in manuscript: L6, P6 in the revised version: "In order to validate the OMPS observation with FTS measurement".

3. The authors discussed whether primary emission or secondary formation contributed significantly to the variations of HCHO, e.g. L15-20, P9, L5-10, P10, but did not tell the reader how they determine the contribution of primary and secondary sources to the variations

of HCHO. The authors need a standard to quantify the contribution to the variations of HCHO.

Responses: Thank you very much for this suggestion. Primary emission and secondary formation were fitted linearly with total HCHO, separately. And larger correlation coefficient means more contribution to the variations of HCHO.

Changes in manuscript: L27-28, P9, in the revised version: "and larger correlation coefficient means more contribution to the variations of HCHO".

4. The author discussed the primary and secondary sources of HCHO in the three megacity Shanghai, Nanjing and Hangzhou, e.g. the spatial and seasonal variations. They should also discuss the commonness and difference of HCHO in the three cities.

Responses: Thank you very much for this suggestion. The secondary source of HCHO in the three megacity have similar seasonal variation, ranking in the order of summer > autumn > spring > winter, similar to seasonal variation of HCHO concentration. And HCHO concentration form secondary formation in summer in Shanghai was larger than that in Nanjing and Hangzhou.

Changes in manuscript: L10-12, P11, in the revised version: "The secondary source of HCHO in the three megacity has similar seasonal variation, ranking in the order of summer > autumn > spring > winter, similar to seasonal variation of HCHO concentration. While HCHO concentration form secondary formation in summer in Shanghai was larger than that in Nanjing and Hangzhou."

5. Sec. 4.2: The discussion of HCHO control measures should use more results from this study instead of just citing the references.

Responses: Thank you very much for this suggestion.

Changes in manuscript: L18-20, P12, in the revised version: "In the Nanjing industrial zone, secondary source of HCHO was about four times larger than primary source in 2017. So at industrial sites, decreasing VOC emissions from related-chemical activities is an efficient way to control ambient HCHO concentration."; L21-27, P12, in the revised version: "Secondary HCHO in the eastern suburbs of Hangzhou was the largest from 2016 to 2017 (Table 1), corresponding to the previous study indicating that the largest amount of VOCs emissions were measured in the eastern suburbs of Hangzhou and VOCs emissions from furniture manufacturing is the largest sources (Lu et al., 2018a). Secondary HCHO contributed most to ambient HCHO, while was not controlled effectively from 2015 to 2017. Considering that industrial manufacturing is developing well and VOCs emissions occur in many types of industries i.e., textile industry, printing, Leather manufacturing and shoemaking (Liu et al., 2008; Khan and Malik, 2014), controlling VOCs emissions in Hangzhou will be a huge challenge."; L29-32, P12, in the revised version: "However, HCHO concentration from secondary source was not controlled effectively. Decreasing private cars on road and developing evaporative control regulations and improving traffic management suited for local conditions help reduce not only VOCs emissions but also HCHO emissions, especially in downtown (Wang and Zhao, 2008; Liu et al., 2015; Liu et al., 2017)."

6. In sec. 5 summary: The authors declare "only secondary HCHO can be used as the proxy

for VOCs reactivity", however, in sec. 4.1, the authors pointed out "Total HCHO can be regarded as the proxy for VOCs reactivity over a three-year study period". It seems paradoxical. The authors should make it explicit.
Responses: Thank you very much for this suggestion. It is corrected in the revised version.
Changes in manuscript: L23-26, P13, in the revised version: **"**Besides, the usability of total HCHO as the proxy of VOCs reactivity depends on time scale. Total HCHO can be regarded as the proxy for VOCs reactivity over a three-year study period, while only secondary HCHO can be used as the proxy for VOCs reactivity over a short study period, e.g. in winter."

7. So separating HCHO from different sources is full of great significance in sensitivity studies of O3 production.
Responses: Thanks. It was corrected.
Changes in manuscript: L26-27, P13, in the revised version: "So separating HCHO from different sources is of great significance insensitivity studies of O_3 production."

8. Line 12 in Page 2: "causing" should be "cause".
Responses: Thanks. It was corrected. Please see L12, P2 in the revised version.

9. Line 13 in Page 2: "favor to" should be "be in favor of".
Responses: Thanks. It was corrected. Please see L13, P2 in the revised version.

10. Line 21 in Page 5: "Surface air pollutants monitored by CNEMC was provided" should be "Surface air pollutants monitored by CNEMC were provided".
Responses: Thanks. It was corrected. Please see L25, P5 in the revised version.

11. Line 10 in Page 7: "the distribution of HCHO VCDs were homogenous "should be "the distribution of HCHO VCDs was homogenous ".
Responses: Thanks. It was corrected. Please see L18, P7 in the revised version.

12. Line 28 in Page 8: "The distribution of the three station selected to explore the primary and secondary sources of HCHO are shown" should be "The distribution of the three station selected to explore the primary and secondary sources of HCHO is shown".
Responses: Thanks. It was corrected. Please see L7, P9 in the revised version.

13. Line 4 in Page 11: The year "2015" should be "2005". The paper published in 2010 cannot research O3 pollution in 2015.
Responses: Thanks. It was corrected. Please see L16, P11 in the revised version.

14. Figure 3, the label of Yellow Sea and Bohai Sea goes against each other.
Responses: Thanks, we have fixed it in the revised version.

15. Figure 7: what are the P-value from the significance tests?
Responses: Thanks. The P-value from the significance tests were added in the Figure 7 captions.