Response to acp-2018-1192-RC2

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.

Specific comments:

Page 6, section 3.2: the author estimated the total national cancer risk of 33500 people and 439 cancer cases per year using OMPS HCHO observation. Please explain how to convert?

Responses: Thank you very much for this suggestion. The total number of people who may develop cancer due to outdoor HCHO exposure in China is the sum of average cancer risk multiplied by the population in each provincial level administrative region. The total cancer cases per year are calculated through dividing the total number of people by life expectancy. And detailed formula and parameters are added in the revised version.

Changes in manuscript: L8-13, P7, in the revised version; L15, P7, in the revised version: "(Eq. (5))"

Page 6, Line 10-11: HCHO concentrations from OMPS measurement were generally lower than those from FTS, and the underestimation from OMPS was attributed to errors of spectral fitting and AMF calculation. The authors should explain the detailed reasons of errors instead of simply attributing spectral fitting and AMF calculation.

Responses: Thank you very much for this suggestion. Errors from spectral fitting affect the accuracy of SCD, and the uncertainties from the scattering weights calculated by the radiative transfer model and HCHO vertical profiles modeled by WRF-Chem affect AMF calculation. Spatial average from satellite observations and errors from SCD fitting and AMF calculation may causing the underestimation.

Changes in manuscript: L13-15, P6, in the revised version: "The underestimation from OMPS was attributed to spatial average from satellite observations and errors from HCHO SCDs affected by spectral fitting and AMF calculation affected by scattering weights calculated by the radiative transfer model and HCHO vertical profiles modeled by WRF-Chem."

Page 10, section 3.3.3 line 28-30: The authors conclude that primary emission influenced the variation of ambient HCHO more significantly than secondary formation in winter. This conclusion should be supported by quantitative analysis.

Responses: Thank you very much for this suggestion. At PDNA site, the correlation coefficient between total HCHO and primary HCHO (R=0.61) is larger than that between total HCHO and secondary HCHO (R=-0.42). So primary emission influenced more significantly than secondary formation on variation of ambient HCHO in winter.

Changes in manuscript: L7-9, P11, in the revised version: "While at PDNA site, primary emission (R=0.61, Table 4) influenced more significantly than secondary formation (R=-0.42, Table 4) on variation of ambient HCHO in winter and became the most important source to ambient HCHO in spring and winter of 2016."

Page 11, section 4.1: The author discussed whether total HCHO can be regarded as the proxy for VOCs reactivity depending on the correlation between total HCHO with secondary HCHO. Why the relationship between them can be used to judge the representation of HCHO as the proxy for VOC reactivity?

Responses: Thank you very much for this suggestion. Secondary HCHO is an oxidation product of most VOCs and is produced with the formation of peroxy radicals (RO_2), positively correlating with RO_2 *(valin et al, 2015, ACP, wolfe et al, 2016, ACP)*. So only HCHO from secondary formation can be identified as the proxy for total VOCs reactivity. If total HCHO shows a good correlation with secondary HCHO, and then total HCHO will positively correlate with RO_2 and can be identified as the proxy for total VOCs reactivity.

Changes in manuscript: L22-23, P11, in the revised version: **"**The good correlation between total HCHO with secondary HCHO means a good correlation between total HCHO with RO_2, representing total HCHO as the proxy for VOCs reactivity."

Page 11, section 4.2: When discussing the HCHO control measures, it should be focused on HCHO pollution, i.e., HCHO concentrations beyond the air quality standard. When HCHO concentration is low, it is not necessary to discuss whether paying more attention to primary emission or secondary formation

Responses: Thank you very much for this suggestion. Here we define HCHO pollution with HCHO concentration reached the threshold value of 10 ppbv, the 95th percentile of all the observation data. HCHO pollution events occurred mostly in summer accounting for 81.0%, 81.3%, and 95.2% in Nanjing, Hangzhou and Shanghai. In summer, HCHO pollution occurred mostly during the periods from 24 to 27 July 2016 and 22 to 27 July 2017 in Nanjing, and from 23 to 27 July 2016 and from 20 to 27 July 2017 in Hangzhou. HCHO pollution in Shanghai lasts longer than Nanjing and Hangzhou, from 10 to 27 July of 2016 and 2017. Besides, HCHO pollution events were also observed during the period from 23 to 27 August 2015 and from 5 to 24 August 2017 in north of Shanghai center. In Shanghai suburb, three HCHO pollution periods were also observed, including: (1) from 19 June to 5 July 2015, (2) from 19 to 23 August 2016, (3) from 4 to 7 August 2017. During HCHO pollution events, the secondary formation contributed most to ambient HCHO and increased more largely than primary HCHO. Compared to the concentrations before pollution days, secondary HCHO and primary HCHO increased 0.90 ppbv and -0.12 ppbv in average, respectively, in Nanjing; increased 1.33 ppbv and 0.21 ppbv, respectively, in Hangzhou; and increased 1.26 ppbv and 0.14 ppbv, respectively, in Shanghai. So decreasing VOCs emissions, the source of secondary HCHO, has a more sensitive effect on controlling HCHO pollution in Nanjing, Hangzhou and Shanghai.

Changes in manuscript: L5-17, P12, in the revised version: "Here we define HCHO pollution with HCHO concentration reached the threshold value of 10 ppbv, the 95th percentile of all the observation data. HCHO pollution events occurred mostly in summer accounting for 81.0%, 81.3%, and 95.2% in Nanjing, Hangzhou and Shanghai. In summer, HCHO pollution occurred mostly during the periods from 24 to 27 July 2016 and 22 to 27 July 2017 in Nanjing, and from 23 to 27 July 2016 and from 20 to 27 July 2017 in Hangzhou. HCHO pollution in Shanghai lasts longer than Nanjing and Hangzhou, from 10 to 27 July of 2016 and 2017. Besides, HCHO pollution events were also observed during the period from 23 to 27 August 2015 and from 5 to 24 August 2017 in north of Shanghai center. In Shanghai suburb, three HCHO pollution periods were also observed, including: (1) from 19 June to 5 July 2015, (2) from 19 to 23 August 2016, (3) from 4 to 7 August 2017. During HCHO pollution events, the secondary formation contributed most to ambient HCHO and increased more largely than primary HCHO. Compared to the concentrations before pollution days, secondary HCHO and primary HCHO increased 0.90 ppbv and -0.12 ppbv in average, respectively, in Nanjing; increased 1.33 ppbv and 0.21 ppbv, respectively, in Hangzhou; and increased 1.26 ppbv and 0.14 ppbv, respectively, in Shanghai. So decreasing VOCs emissions, the source of secondary HCHO, has a more sensitive effect on controlling HCHO pollution in Nanjing, Hangzhou and Shanghai."

Technical corrections:

Page2, Line4: change "become increasing serious" to "become increasingly serious"
Responses: Thanks. It was corrected. Please see L4, P2 in the revised version.

Page5, Line20: change "Surface air pollutants monitored by CNEMC was" to "Surface air pollutants monitored by CNEMC were"
Responses: Thanks. It was corrected. Please see L25, P5 in the revised version.

Page5, Line27: change "one of the condition" to "one of the conditions"
Responses: Thanks. It was corrected. Please see L1, P6 in the revised version.

Page8, Line11: change "industrial zoon" to "industrial zone"
Responses: Thanks. It was corrected. Please see L22, P8 in the revised version.