Responses to reviewers' comments on the paper of "Towards a satellite – in situ hybrid estimate for organic aerosol abundance" by Jin Liao et al.

We thank the reviewers for their comments on our paper. To guide the review process we have copied the reviewer comments in black text. Our responses are in regular blue font. We have responded to all the referee comments and made changes **in bold text**.

Anonymous Referee #2

R2.0. This paper attempts to develop an estimate of the surface OA concentrations using three variables: (1) satellite HCHO column, (2) conversion factor from satellite HCHO column to surface HCHO concentrations (from a model), and (3) relationship between surface OA and HCHO concentrations derived from airborne measurements. The authors examined the relationship between surface OA and HCHO concentrations from a number of airborne measurements. They find that this relationship varies greatly among different studies (Table 1 and Figure 2). They further examine the dependence of OA-HCHO relationship on ambient NOx concentrations, and they do find that the slope gets lower with increasing NOx (Figure 3). The GEOS-Chem model was able to reproduce this relationship in the absence of wildfires. Finally, the authors use the relationship established from SEAC4RS to estimate surface OA and it compares well with IMPROVE network. They found that adding NOx dependence or special treatment to urban cities do not change the correlation coefficient between OA estimate and IMPROVE OA. I have a few comments:

R2.1. It seems that this relationship is largely driven by the relative contribution of POA vs. SOA to the total OA burden. As HCHO is mostly secondarily produced, this OA-HCHO is expected to have high slopes when OA emission is dominated by POA (such as wildfires shown in Figure 2). For SEAC4RS or DC3 this slope is much lower because OA is likely dominated by SOA. For KORUS-AQ or CalNex, this slope is likely driven by a mixture of biogenic and anthropogenic emissions, which falls somewhere between. It seems important to understand the

contribution of POA vs. SOA to the total OA burden, before one uses this OA-HCHO to estimate OA. I am wondering if this can be investigated in their model.

We thank the reviewer for the comment. However, we believe that the situation is more complex, and that the OA-HCHO relationship is not just driven by the relative contribution of POA vs SOA to total OA.

For urban areas sampled from aircraft, recent analysis by Nault et al. (2018) indicates that the contribution of POA in KORUS-AQ is small (6-10%) and that the enhancement in OA is due to increased SOA production. This is consistent with past megacity studies (see references in that paper). Thus both OA and HCHO are dominantly secondary for the air masses sampled by the aircraft, and the slope is determined by their respective rates of production (see also Wood et al., 2010, which presents an analogous analysis for the OA vs Ox slope). At the relatively short photochemical ages sampled in KORUS-AQ, the difference in lifetimes for OA vs HCHO does not yet play a major role in changing the slope.

For biomass burning, POA is very large and the net formation of SOA is small (see e.g. Cubison et al., 2011). In this case the slope will be determined by the initial POA and HCHO emissions, together with SOA and especially HCHO production.

It is possible that in some cases the relative contributions of POA vs. SOA do play a controlling role on the OA/HCHO slope. This may become apparent as we analyze more cases.

Line 334-335 changed "The high OA air masses also had high acetonitrile during KORUS-AQ." to "During KORUS-AQ, the high OA/HCHO air masses tended to have high acetonitrile. By the time we sampled, most organic aerosols were secondary (Nault et al., 2018). This indicates that the formation rates of OA and HCHO from different emission sources contribute to the different slopes of OA-HCHO. This also indicates that emission sources with enhanced acetonitrile tend to form more OA relative to HCHO downwind."

The role of POA in impacting the slope of OA-HCHO in biomass burning case has been described in Line 339-343 "The slopes of OA vs. HCHO for BB air masses were higher than for anthropogenic and biogenic sources. This is consistent with high POA emission in BB conditions (Heald et al., 2008; Lamarque et al., 2010; Cubison et al., 2011), with low addition of mass due to

SOA formation (Cubison et al., 2011; Shrivastava et al., 2017)."

References:

Nault, B. A., Campuzano-Jost, P., Day, D. A., Schroder, J. C., Anderson, B., Beyersdorf, A. J., Blake, D. R., Brune, W. H., Choi, Y., Corr, C. A., de Gouw, J. A., Dibb, J., DiGangi, J. P., Diskin, G. S., Fried, A., Huey, L. G., Kim, M. J., Knote, C. J., Lamb, K. D., Lee, T., Park, T., Pusede, S. E., Scheuer, E., Thornhill, K. L., Woo, J.-H., and Jimenez, J. L.: Secondary Organic Aerosol Production from Local Emissions Dominates the Organic Aerosol Budget over Seoul, South Korea, during KORUS-AQ, Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2018-838, in review, 2018.

Wood, E. C., Canagaratna, M. R., Herndon, S. C., Onasch, T. B., Kolb, C. E., Worsnop, D. R., Kroll, J. H., Knighton, W. B., Seila, R., Zavala, M., Molina, L. T., DeCarlo, P. F., Jimenez, J. L., Weinheimer, A. J., Knapp, D. J., Jobson, B. T., Stutz, J., Kuster, W. C., and Williams, E. J.: Investigation of the correlation between odd oxygen and secondary organic aerosol in Mexico City and Houston, Atmos. Chem. Phys., 10, 8947-8968, https://doi.org/10.5194/acp-10-8947-2010, 2010.

Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D., Lechner, M. J., Brune, W. H., Apel, E., Diskin, G. S., Fisher, J. A., Fuelberg, H. E., Hecobian, A., Knapp, D. J., Mikoviny, T., Riemer, D., Sachse, G. W., Sessions, W., Weber, R. J., Weinheimer, A. J., Wisthaler, A., and Jimenez, J. L.: Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies, Atmos. Chem. Phys., 11, 12049-12064, https://doi.org/10.5194/acp-11-12049-2011, 2011.

R2.2. Time dependence of OA-HCHO relationship. If indeed OA is dominated by SOA,
one would expect a time-dependence of OA-HCHO relationship, as OA and HCHO are
produced at different rates. How would this possibly affect their results?

Thanks for the reviewer's comment. We understand that the OA-HCHO relationship could be time dependent, in principle, due to different lifetimes and different net chemical reaction rates. HCHO is close to being in steady-state with production rates roughly equal to loss rates while OA is not in steady-state with a

lifetime of a week. Therefore, OA can be accumulated relative to HCHO when air masses are aged. This is evident as shown in Fig S2, using altitudes as an approximation of air mass age. Line 630-648 discussed the impact of the time dependent OA-HCHO relationship. The time dependent OA-HCHO relationship does not largely affect our study for monthly average OA over continental US because our OA estimates show reasonably good agreement with ground sites IMPROVE OA measurements. This also indicates that enhanced SOA are near the source regions statistically. We will consider the impact of air mass age (or time dependence of OA-HCHO relationship) when using refined OA-HCHO relationship to get higher spatial and temporal resolutions of OA estimate in the future.

Line 634 added "HCHO is close to being in steady-state with production rates roughly equal to loss rates while OA is not in steady-state with a lifetime of a week. Therefore, OA can be accumulated relative to HCHO when air masses are aged."

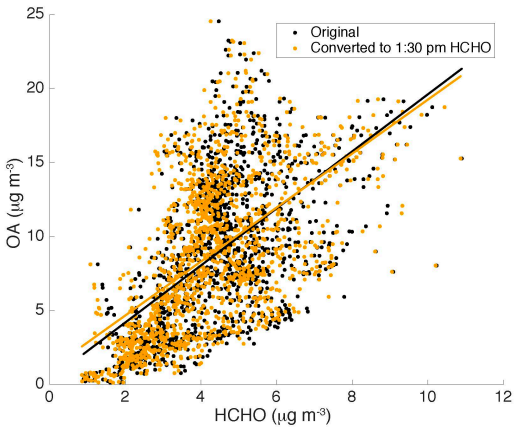
633-634 changed "OA vs. HCHO from SEAC^{4}RS and KORUS-AQ field campaigns, color-coded with altitude, are plotted in Fig. S2 (a) and (b), respectively." To "OA vs. HCHO from SEAC^{4}RS and KORUS-AQ field campaigns, color-coded with altitude as an indicator of air mass age, are plotted in Fig. S2 (a) and (b), respectively."

Line 648 added "Although OA-HCHO relationships depend on air mass age, it does not largely affect our study for monthly average surface OA over continental US because our OA estimates showed reasonably good agreement with ground sites IMPROVE OA measurements. This also indicates that SOA are enhanced near the source regions statistically. Nault et al. (2018) also showed the production of HCHO and SOA are similar and plateau around 0.5 - 1 photochemical day. So, in the near field of emissions and chemistry, the productions of these two species are similar; however, outside of near field of emissions and rapid chemistry, the long lifetime of OA vs the steady state of HCHO would start controlling the slopes and correlations."

R2.3. Conversion from mid-day to daily average. The authors use the relationship be-
tween HCHO and OA derived from airborne measurements, and satellite HCHO col-

umn (overpass time is 130pm local time), to derive a surface OA, which should be the
OA in local time 130pm. Then the authors compare that to IMPROVE monthly aver-
age data. It seems logical to add a correction factor from 1:30pm to daily or monthly
average. The argument in Line 494-495 “ground OA in 494 the Southeast US were
observed to have little diurnal variation” is not good enough.

Thanks for pointing this out. We agree with the reviewer that it is important to estimate the impact of the inconsistency in using mid-day satellite HCHO data and daytime aircraft OA-HCHO relationships because HCHO has a clear diurnal profile and OA doesn’t. Because the in situ flight data were collected in the daytime, the potential bias turns out to be small and we added as uncertainties in OA estimate. See the plot of the OA-HCHO relationships with and without HCHO corrected to 1: 30 pm according to the average HCHO diurnal profile. The text is modified as below.



Line 497 added “However, surface HCHO has evident diurnal profiles with the highest concentrations around the mid-day (Kaiser et al., 2016), which could add uncertainties to OA estimate when using inconsistent time ranges of satellite HCHO data measured in the mid-day and in situ airborne OA and HCHO relationships measured in the daytime. The SEAC^{4}RS HCHO concentrations were converted to 1:30 pm concentrations according to the average HCHO diurnal profile from the Southern Oxidant and Aerosol Study (SOAS) (Kaiser et al., 2016). The OA-HCHO relationship with HCHO converted to 1:30 pm yielded a slope of 5% lower than the original OA-HCHO relationship.”

Line 497-501 deleted. "The difference in mid-day and daytime HCHO concentrations is not prominent, depends on the location and may contribute to a small bias to the mid-day OA estimate (DiGangi et al., 2012). This is probably due to increased boundary layer height diluting the photochemical formation of HCHO in the mid-day."

R2.4. The authors show in Figure 7. that different cases make little difference on corre-
lation coefficients. Can the authors show how exactly the NOx dependence is imple-
mented? What kind of NOx concentrations did they for IMPROVE sites (130pm or daily
average)? Can the authors show how many large urban cities are treated differently
in case 3 and 4? So the reader can see how many IMPROVE sites are affected by
this and understand why the effect is so small. Also the authors need to show how OA
concentrations are affected as well.

Thanks for the comment. To be clearer, more detailed information is added.

Line 556 added " The details about how to implement chemical factors dependent OA estimates for the four cases are also provided in Table 2. Satellite OMI NO_2 data (at 1 : 30 pm) are used to represent NO_x levels and big cities are defined as $NO_2 > 4 \times 10^{15}$ molec cm^{-2} , the CalNex in situ OA-HCHO relationship is applied for big cities. It turns out that only 1 IMPROVE site (SAGA1) near LA was affected by high NO_2 and led to the insignificant change in case 3 compared to case 1. This is not unexpected because IMPROVE sites are in rural regions. The OA estimate in SAGA1 decreased from 1.88 ug m-3 from case 1 to 0.17 ug m-3 in case 3 while the measured OA in IMPROVE SAGA1 was 1.52 ug m-3. This may infer that CalNex is not very consistent with SEAC^{4}RS due to different sampling instruments, strategies and seasons. Lowing the NO_2 threshold when defining big cities did not help improve the agreement either."

Because case 3 only changed 1 site compared to case 1, case 4 was similar to case 2 as expected and did not improve the agreement between OA estimate and IMPROVE OA.

R2.5. Y-axis label of Figure 7 should be fixed.

Thanks for pointing this out. **It is changed to “Correlation coefficients of OA estimate v.s IMPROVE OA”.**