

**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 #1

R1.0. This study aims to use the information in HCHO satellite observations, along with a model estimate of the vertical distribution, and in situ HCHO:OA relationships, to estimate surface OA concentrations over the United States. This is an interesting concept, though it is challenged with many uncertainties and assumptions and the manuscript is a bit unfocused. Here are the major issues that I have identified:

R1.1. The title is vague and somewhat inaccurate. The estimate involves satellite, in situ observations AND a model, the "hybrid" should be described as such. Given that the results presented are US only, "over the continental United States" should be specified in the title. And finally, I would recommend that the authors modify the title to include "formaldehyde" so that it is clear that this is not an estimate based off of satellite AOD.

We appreciate the reviewer's suggestion about the title.

To keep the title simple, "over the continental United States" is included in the abstract instead.

In the abstract we added "over the continental US" in Line 46 as:

"Near-surface OA over the continental US are estimated by combining observed in situ relationships with HCHO column retrievals from NASA's Ozone Monitoring Instrument (OMI)."

In the abstract (L 48), we added OA estimate with η from satellite a priori profiles, as:

"HCHO vertical profiles used in OA estimates are from climatology a-priori profiles in the OMI HCHO retrieval or output for specific time period from a newer version of GEOS-Chem."

Line 48 – 53, we have changed "We evaluate this OA estimate against OA observations from the US EPA IMPROVE network and simulated OA from the GEOS-Chem global chemical transport model. The OA estimate compares well

with IMPROVE data obtained over summer months (e.g. slope = 0.62, $r = 0.56$ for August 2013), comparable to intensively validated GEOS-Chem performance (e.g. slope = 0.57, $r = 0.56$) and superior to the correlation with satellite-derived total aerosol extinction ($r = 0.41$).”

To:

“We evaluate these OA estimates against OA observations from the US EPA IMPROVE network and simulated OA from the GEOS-Chem global chemical transport model. The OA estimates compare well with IMPROVE data obtained over summer months (e.g. slope = 0.60-0.62, $r = 0.56$ for August 2013), comparable to intensively validated GEOS-Chem performance (e.g. slope = 0.57, $r = 0.56$) and superior to the correlation with satellite-derived total aerosol extinction ($r = 0.41$). This also indicates that OA estimates are not very sensitive to HCHO vertical profiles and that a priori profiles from OMI HCHO retrieval are similar to that from the newer model version in estimating OA.”

Because satellite HCHO data containing the modeling components we need for the organic aerosol estimate, we still keep using “satellite – in situ hybrid” in the title.

As suggested by the reviewer, we included formaldehyde in the title.

We changed the title to be:

“Towards a satellite formaldehyde – in situ hybrid estimate for organic aerosol abundance”.

R1.2. What are the implications of the statement on lines 88-89 that “isoprene SOA is not the dominant source of SOA in summer” for the GEOS-Chem simulation in that region? The study refers several times to the GEOS-chem simulation for the SEUS being “extensively validated” (lines 52, 87, 239) but seems to refer to isoprene SOA. Does the simulation capture the non-isoprene components of SOA as well?

Thanks for pointing this out.

According to Figure 5 in Kim et al. (2015), this GEOS-Chem version (v9-02) reasonably well captured the non-isoprene components of SOA during SEAC⁴RS as well.

To avoid confusion, we deleted line 88-89 “but this is only one location and it is likely that isoprene SOA is not the dominant source of SOA in summer.”

R1.3. The fire analysis (inset Figure 2) is not very convincing given the small number of points. Why did the authors not include data influenced by fire from other regions in their

analysis? In particular, the agricultural fire analysis appears to rely on very few points and the discussion of those results in lines 324-326 and 344-346 should be tempered.

Thanks for the reviewer's comment. Now we use specific term to constrain the statements.

Lines 324-326: changed "OA exhibits a tight correlation with HCHO for both wildfires and agricultural fires". To "OA exhibits a tight correlation with HCHO for both wildfires and agricultural fires from SEAC⁴RS data."

Line 326 added: "SEAC⁴RS data are chosen because it sampled fires and had state-of-the-art, high quality measurements."

Lines 344-346: changed "The slope of OA to HCHO was higher for wildfires than agricultural fires. This may indicate that more OA is emitted in wildfires which often have higher intensity than agricultural fires (Liu et al., 2017; Forrister et al., 2015)."

To "The slope of OA to HCHO was higher for wildfires than agricultural fires during SEAC⁴RS. This is consistent with more OA emitted in wildfires than agricultural fires (Liu et al., 2017). The factors driving higher OA to HCHO with wildfires are not clear and may be related to burning conditions and fuels."

R1.4. The relationship between OA and HCHO seems to break down as you increase NO_x (it's not all that strong to begin with, see point 7). The correlation coefficient of 0.44 implies under high NO₂ less than 20% of the variability of OA can be explained by HCHO. An OA estimate based off of this weak relationship does not seem credible. Also: was NO₂ not measured during CalNex and DC3 (Lines 176-183)? Why are these campaigns not parsed for NO_x?

Thanks for pointing out the confusion. Ambient data are different from lab experiments that have controlled conditions or simulations that have known mechanisms. To be clearer to reviewer and readers, we modified the text as below.

Line 376 added "The correlation coefficient of 0.45 for high NO₂ and isoprene conditions during SEAC⁴RS is not very high but still shows significant dependence of the OA-HCHO relationship on the product of NO₂ and isoprene, considering that these are ambient data and other factors (e.g. different specific sources) also play a role in determining OA-HCHO relationships."

Line 383 changed “The OA / HCHO ratio clearly decreased as NO₂ levels increased during KORUS-AQ, suggesting that high NO conditions accelerated HCHO formation more than they did SOA production.” To “The OA / HCHO ratio clearly decreased as NO₂ levels increased during KORUS-AQ, suggesting that high NO conditions accelerated HCHO formation more than they did SOA production. OA-HCHO relationships do not have dependence on local time of the day (not shown). This further confirms that NO_x is an important factor that affects the OA/HCHO relationship.” This case is impressive for ambient data.

Line 560-562: changed “As the in situ data showed a NO₂–isoprene-dependent OA and HCHO relationship, we attributed this to the uncertainty of isoprene emissions from MEGAN or IMPROVE network measurements.” Into “As the in situ data showed a moderate NO₂–isoprene-dependent OA and HCHO relationship, we attributed this to the uncertainty in isoprene emissions from MEGAN, the locations of IMPROVE site at rural regions, the uncertainty in IMPROVE network measurements, or other factors (e.g. sources-dependent OA-HCHO) besides NO₂-isoprene that also need to be taken into account when determining the specific OA-HCHO relationship.”

Line 388 added “Because OA and HCHO were tightly correlated during CalNex and DC3, we did not parse for NO_x. The NO_x range during DC3 low altitude data was smaller than KORUS-AQ and SEAC⁴RS. DC3 OA and HCHO relationships only had slight dependence on NO₂ (not shown here), largely due to the limited dataset. The NO_x range during CalNex low altitude data was large. The tight OA and HCHO correlation during CalNex could be due to the combination of different VOCs sources and NO_x levels.”

R1.5. A key part of this analysis is the conversion of HCHO column to surface using the GEOS-Chem model. Are the AMFs used in the OMI product consistent with the eta’s used here? How does the model treat boundary layer mixing? Are the results sensitive to this? Why do the authors only show SEAC⁴RS profiles of HCHO? How does the model perform for DC3 and CalNex? Lines 455-456 discusses the potential for BB plumes to impact the vertical profile; this could be investigated with the DC3 data.

Thanks for pointing out this. The satellite a priori profiles are used now. The text is modified as below.

To evaluate if the OA estimates are sensitive to η from different model versions, we also estimated OA using η from a priori profiles used in OMI retrieval AMF calculations.

Line 267-269: changed “ η (i) is derived from GEOS-Chem (v9-02), which includes updated isoprene scheme for OA and is the next version of the model (v9-01-03) for a priori profiles used in SAO satellite HCHO retrievals.”

To “ η (i) is derived from the HCHO a priori profiles used in SAO OMI air mass factor (AMF) calculations (GEOS-Chem v9-01-03 climatology) or from GEOS-Chem v9-02, which includes updated isoprene scheme for OA and is the next version of the model (v9-01-03) for a priori profiles used in SAO satellite HCHO retrievals. HCHO a priori profiles are used to be consistent with satellite HCHO retrievals and also to show that OA estimate can be derived without running a global model separately. The newer version of GEOS-Chem is used to test the sensitivity of OA estimates to updated version of η . The newer version of GEOS-Chem also allows sampling through the flight tracks of a recent field campaign (SEAC⁴RS) flight tracks and examining the factors impacting η with both modeled and measured HCHO profiles. The detailed information about the impact of HCHO profiles on η is provided in Sect. 5.”

Line 490 added “ η in Fig. 6a OA estimate is from GEOS-Chem v9-02 output for the specific month August, 2013. η in Fig. 6b OA estimate is from the climatology HCHO profiles satellite data used as a priori profiles from GEOS-Chem v9-01-03. The correlations of OA estimates with IMPROVE OA indicate that OA estimates are not very sensitive to η from different model versions”

Line 445 added “Because the sensitivity of OA estimates to η is investigated with η from different GEOS-Chem versions (Sect. 6.2), we don’t compare HCHO vertical profiles from model and measurements for a comprehensive set of field campaigns. We chose SEAC⁴RS to investigate the main factors impact the η over US because SEAC⁴RS has a larger spatial coverage than DC3 and CalNex.”

Line 455-456: changed “High concentrations of HCHO (e.g., in BB plumes) lofted by convection can also impact the vertical profile (Barth et al., 2015).” To “High concentrations of HCHO (e.g., in BB plumes) lofted by convection can also impact the vertical profile (Barth et al., 2015), **which is not further investigated because OA estimates with BB influences over US are excluded in current study.**”

R1.6. The South Korea analysis is a null result and adds little to the paper. I suggest that the authors eliminate Section 6.7 and earlier analysis of KORUS-AQ data. They could re-cap in the Conclusions that a similar analysis was attempted for South Korea but was precluded by low HCHO from OMI.

Thanks for the reviewer's suggestion. We still want to keep the KORUS-AQ data analysis because the analysis provides useful information to indicate that this method enables estimating OA beyond continental US. To state the purpose of the South Korea analysis, we added the following text.

Line 602, added “Although an OA estimate for South Korea could not be retrieved in the current study, the consistency in the dependence of OA-HCHO relationships on chemical factors (e.g. emission sources, NO_x, and altitudes) provides important information for potential application of chemical factors dependent OA-HCHO relationships to the geographical domain beyond the continental US, especially with improved satellite HCHO data from Tropospheric Monitoring Instrument (TROPOMI).”

R1.7. Need to develop a more detailed and quantitative discussion of uncertainties. Several potentially large uncertainties are discussed on lines 501-508 and Section 7. The authors should strengthen and consolidate this discussion to outline all the assumptions and potential impact on their analysis. In particular, I note that the correlations between HCHO and OA in the in situ observations are actually not all that high to begin with (in the case of SEAC4RS HCHO only explains 35% of the variability in OA).

Thanks for the comment. It is challenging to quantify the uncertainties of the OA estimates. For example, one of the uncertainty sources comes from satellite HCHO data. Zhu et al. (2016) had a detailed study about the comparison of different satellite HCHO retrievals with model results and in situ measurements. It is challenging to have an apple to apple comparison between satellite data with in situ measurements. The correlation coefficient between SAO HCHO retrieval and in situ measurements was only 0.24. To have a more detailed and quantitative discussion of uncertainties, we modified the text as below.

Line 488 changed “The good correlation (correlation coefficient $r = 0.56$) between the OA estimate and corrected IMPROVE network measurements (Fig. 6e) indicates that the OA estimate can generally capture the variation of OA loading over the US.” To

“Considering the uncertainties in satellite HCHO measurements, in using a campaign lump-sum OA-HCHO relationship to represent spatial resolved OA, in HCHO vertical profiles, and in ground IMPROVE network measurements, the correlation (correlation coefficient $r = 0.56$) between the OA estimate and corrected IMPROVE network measurements (Fig. 6f and 6g) is reasonably good

and indicates that the OA estimate can generally capture the variation of OA loading over the US.”

Line 490 added “The correlation coefficient between HCHO SAO retrievals and in situ measurements during SEAC⁴RS was not high ($r = 0.24$) but this may be partly because they were not sampled at the same time. The uncertainty in HCHO SAO data was likely less than 76%. The uncertainty in applying a campaign lump-sum OA-HCHO relationship to individual spatial resolved satellite HCHO data to estimate OA induced an uncertainty of 41% according to the correlation coefficient of OA-HCHO in the field campaign. η in the Fig. 6a OA estimate is from a GEOS-Chem v9-02 output for the specific month August, 2013. η in the Fig. 6b OA estimate is the climatology from GEOS-Chem v9-01-03, the same as satellite data a priori profiles. The good correlations of OA estimates with IMPROVE OA indicate that OA estimates are not very sensitive to η from different model versions. The largest difference between the two OA estimates is their concentrations over East Texas. There are no IMPROVE OA measurements in the East Texas to evaluate which works better. The uncertainties in IMPROVE OA measurements, such as using a constant correction factor to correct the partial evaporation across all southeast US sites, and the spatially dependent OA/OC ratio (Tsigaridis et al., 2014), may also have contributed to the discrepancies between the OA estimate and EPA IMPROVE sites OA. Therefore, higher quality of satellite HCHO data and refining OA-HCHO relationships will help improve our OA estimate products. This combined with a spatially resolved IMPROVE OA correction factor and OA/OC ratios will help improve the correlation coefficients between OA estimates and IMPROVE OA.”

Line 501-503 changed “Instead, the potential underestimation of HCHO from satellite retrieval (by 37%) (Zhu et al., 2016) compared to SEAC⁴RS may cause the low slope between the OA estimate and IMPROVE OA according to Eq. (1).” To “Instead, the potential underestimation of HCHO from satellite retrieval (by -37%) (Zhu et al., 2016) compared to SEAC⁴RS may be one of the most important reasons that cause the systematic difference (low slope) between the OA estimate and IMPROVE OA according to Eq. (1). Satellite HCHO data corrected by the low bias (by -37%) (Zhu et al., 2016) will increase our slopes of 0.60-0.62 to be close to the unity.”

Line 501 added “The potential uncertainty (30%) in OA/OC ratio could also contribute to the systematic difference because we used OA/OC of 2.1 and studies (e.g. Pye et al., 2017; Canagaratna et al., 2015) showed that the OA/OC can range from 1.4 to 2.8.”

R1.8. Also many assumptions are based solely on the SEUS:

a. That OA accounts for a large fraction of submicron aerosol (lines 277-278). What about in other regions of the US?

Generally, OA is a major submicron aerosol component over the US.

Line 277-278: added “and is one of the major submicron aerosol components over the US generally (Jimenez et al., 2009).”

b. That the vertical profile of OA matches the total aerosol (line 284) – this has not been shown outside of SEUS and the potential for dust, nitrate, smoke plumes to alter this should be discussed

More information about the potential impact of dust, nitrate, and smoke plumes is added.

Line 284: changed “The shape of average vertical profile of OA was very close to that of total aerosol mass (Wagner et al., 2015)” to “The shape of the average vertical profile of OA was very close to that of total aerosol mass for Southeast US (Wagner et al., 2015) where most of the enhanced aerosol concentrations over the US are located. Data with smoke plumes interferences are excluded in the following analysis. The potential contribution of dust and nitrate could alter the shape of the vertical profiles and introduce uncertainties when using OA vertical profiles for other parts of the US. Similar vertical profile shapes of OA and submicron particles were also observed in a campaign outside the US over South Korea (Nault et al., 2018). Though OA accounted for ~40% of the total submicron particles, the shape of OA and total submicron particles vertical profiles were nearly identical.”

Line 536 added “The high surface aerosol extinctions ($> 150 \text{ Mm}^{-1}$) (outliners in the scatter plot) are located in the Southeast US and therefore are not due to potential contribution of dust and nitrate altering the shape of vertical profiles outside of the SE US.”

c. Applying the OA:HCHO relationship from SEAC4RS for the entire US. The authors tested using data from the LA Basin as well, but how would OA (and HCHO) differ in the Midwest and NEUS? In Section 6.2 the authors discuss the ability of the OA estimate to

capture IMPROVE observations – how good is the estimate outside of the SEUS, where observations were used to construct the HCHO:OA relationship?

Thanks for the comment. The following discussions about Midwest and NEUS are added.

Line 563 added “SEAC⁴RS and DC3 only had a few low altitude data in the Midwest and did not cover the Northeast US. The measured OA-HCHO relationship in the Midwest did not show significant difference from the SE US. The scatter plots (Fig. 6f and 6g) of OA estimates and IMPROVE OA do not show outliers for the Northeast and Midwest. This indicates that using the SEAC⁴RS lump sum OA-HCHO relationship can reasonably capture regions outside of the SEUS.”

MINOR

R1.9. The abstract is missing a few key descriptors: that the analysis is performed for summertime only, that the estimate of near-surface OA is for the United States, and that that estimate also relies on the GEOS-Chem vertical profile of HCHO.

Thanks for the comment. The abstract is modified to include the information.
Abstract line 46 changed “An estimate of near-surface OA is derived by combining observed in situ relationships with HCHO column retrievals from NASA’s Ozone Monitoring Instrument (OMI)” to “An estimate of summer time near-surface OA over US is derived by combining observed in situ relationships with HCHO column retrievals from NASA’s Ozone Monitoring Instrument (OMI). HCHO vertical profiles used in OA estimates are from climatology a-priori profiles in the OMI HCHO retrieval or output of specific period from newer version of GEOS-Chem.”.

R1.10. Lines 155-166: how do ISAF and DFGAS measurements compare during SEAC⁴RS?

As mentioned in the text, DFGAS measurements were available during DC3 but not SEAC⁴RS. **Line 162 added “HCHO measurements from ISAF also had a good agreement with DFGAS, with a correlation coefficient of 0.98 and a slope of 1.07.”.**

R1.11. Line 174: specify what temperature was assumed for STP (273 and 298K are both used)

Line 174 added “(at 273 K and 1013 mbar)”.

R1.12. Line 180: correct “for daytime NO”

Corrected

R1.13. Section 2.3: The spatial resolution of the products and details of gridding and averaging are missing.

Line 212 changed “Here we use the OMI HCHO version 2.0 (collection 3) retrieval” to “Here we use the OMI HCHO version 2.0 (collection 3) gridded (0.25 × 0.25) retrieval data”. The averaging information is provided in Line 218-219 “The monthly average HCHO columns were also weighted by the column uncertainties of the pixels.”

R1.14. Lines 273-277: This sentence is awkwardly phrased (“can estimate the fraction of OA” and then the sentence says it can’t do this...). Also why is MISR discussed here and later (lines 525-531) when it is not used in this study?

Changed “The MISR satellite instrument can estimate the fraction of OA relative to total AOD, due to constraints on size range, shape and absorbing properties,” to “The MISR satellite instrument can estimate a subset of AOD, due to constraints on size range, shape and absorbing properties”

Line 277 added “Because MISR estimates a subset of AOD, it is discussed here to verify we are not neglecting a satellite dataset that has already captured OA AOD.”

R1.15. Line 280: need to define A_{ext} in the text

Line 281: added “ A_{ext} is the calculated aerosol extinction (Mm^{-1}),”

R1.16. Line 294: insert “near-surface in situ”

Done.

R1.17. Line 298: define “BB” in text

BB was defined in previous paragraph (Line 106)

R1.18. Line 402/Section 2.4: The emissions are not described in the model description section.

Line 241 added “This model version uses the fourth-generation global fire emissions database (GFED4) (Giglio et al., 2013) as BB emission inventory.”

R1.19. Section 6.1: Are daily HCHO columns converted to surface concentrations using daily etas and then averaged to monthly values, or is the analysis performed using monthly means for everything?

This analysis is performed using monthly average satellite HCHO data.

Line 463 changed “Satellite HCHO column” to “monthly average satellite HCHO column”

Line 463 changed “Satellite HCHO columns” to “Satellite monthly average HCHO column data.”

Line 464 added “either from climatology a priori profiles or monthly average HCHO profiles”

R1.20. Line 488-9: A correlation coefficient of 0.56 is pretty modest. When only 31% of the variation is captured it is not accurate to state that the estimate “generally captured the variation” of the observations.

Line 488-489:

Changed “The good correlation between the OA estimate and corrected IMPROVE network measurements (Fig. 6(e)) indicates that the OA estimate generally captured the variation of OA loading over the US.” To “Considering the uncertainties in satellite HCHO measurements, in using the campaign lump-sum OA-HCHO relationship to represent spatial resolved OA, in HCHO vertical profiles, and in ground IMPROVE network measurements, the correlation (correlation coefficient $r = 0.56$) between the OA estimate and corrected IMPROVE network measurements (Fig. 6(e)) is reasonably good and indicates that the OA estimate can generally capture the variation of OA loading over the US.”

R1.21. Lines 537-538: The authors state their goal in using the extinction data here at the end of the Section. I suggest that the authors explain more clearly at the start of Section 6.3 why they are exploring extinction.

Line 519-520: changed “To further evaluate the method of using satellite HCHO to derive an OA surface estimate, satellite measurements of AOD were converted to extinction for comparison.” To “To further evaluate the method of using satellite

HCHO to derive an OA surface estimate, satellite aerosol measurements are used to approximate surface OA extinction for comparison. Satellite measurements of AOD were converted to surface extinction for the regions AOD is dominated by OA.”