

**Responses to reviewer #1's comments on the revised paper of  
"Towards a satellite – in situ hybrid estimate for organic aerosol  
abundance" by Jin Liao et al.**

We thank the reviewer for his/her comments on our paper. 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**.

The manuscript is improved over the original submission, though some aspects of the analysis remain poorly described. I've provided suggestions below to address these. Given all the uncertainties and the simplistic application of highly-variable source-specific relationships (lines 364-366 encapsulate the fundamental problem that makes it hard to justify any sort of universal application of slopes), I don't think that the authors have made a convincing case that this approach is generally worth pursuing, but given the work that went into the analysis, I think that the paper meets the bar for publication (once they have addressed the remaining issues).

We agree that the simplistic application of highly variable source specific relationships induce uncertainties in estimating OA. However, we can extract the common factors that govern the different OA-HCHO relationships and refine the OA-HCHO relationships to reduce the uncertainties. For example, the source dependent OA-HCHO relationships (Fig. 2) showed higher OA-HCHO slope in biomass burning and anthropogenic sources with inefficient combustions (e.g. KORUS-AQ) compared to biogenic and clean anthropogenic sources. The in situ data indicate that inefficient combustions contribute to the high slopes of OA-HCHO, probably due to both enhanced primary OA and increased formation of SOA. Therefore, it is possible to have a universal application of sources and chemical factors dependent slopes but more analysis is needed and it is beyond the scope of this paper.

To be clear, line 381 added

**"Overall, the source dependent OA-HCHO relationships (Fig. 2) showed highest OA-HCHO slopes of BB and heavily polluted anthropogenic sources with inefficient combustion (e.g., KORUS-AQ) compared to biogenic and relatively clean anthropogenic sources. This indicated that inefficient combustions contribute to the high slopes of**

**OA-HCHO, probably due to both enhanced primary OA and increased formation of SOA. Enhanced pre-existing aerosols such as primary aerosols can provide more surfaces to increase VOCs condensation and SOA formation. VOCs co-emitted from heavily polluted anthropogenic sources can also form more SOA. It is possible to extract the factors that govern the different OA-HCHO relationships and potentially have a universal application of the slopes as a function of the factors (e.g., sources and combustion efficiencies).”**

**Line 358-381 is also re-organized to show that we can extract the factors (e.g. sources) that govern the OA-HCHO relationships.**

1. Lines 105-107: It is unclear what the reader is meant to take from this sentence. Does this approach from de Vries et al. work well for OA? Are there major flaws? Are you trying to provide an alternative approach? The approach from de Vries et al. characterized the aerosol types (e.g. OA) for AOD. Our approach is trying to quantify OA mass concentrations.

**Line 107 added**

**“Here we aim to provide a quantitative estimation of OA mass concentrations from satellite measurements.”**

2. Lines 127-128: Jimenez et al. (2009) do not show that OA is a major contributor to AOD (they do not discuss AOD) – an alternate reference is required for this statement.

**line 127-128 changed “This also suggests that OA, a major contributor to AOD in the above cases (Jimenez et al., 2009), and HCHO share common emission sources and photochemical processes.” To “This also suggests that OA share common emission sources and photochemical processes with HCHO and are an important contributor to AOD in the above cases.”**

3. Line 180: state in the text why you convert from STP units

**line 180 changed “The OA measurements are from 1 min merge data and converted from  $\mu\text{g sm}^{-3}$  (at 273 K and 1013 mbar) to  $\mu\text{g m}^{-3}$  under local T & P for each data point.” To “The OA measurements are from 1 min merge data and converted from  $\mu\text{g sm}^{-3}$  (at 273 K and 1013 mbar) to  $\mu\text{g m}^{-3}$  under local T & P for each data point, to be consistent with**

**HCHO concentrations in  $\mu\text{g m}^{-3}$  or molec  $\text{cm}^{-3}$  at local T & P.”**

4. Lines 182-189: give measurement details for NO<sub>2</sub> for DC3 and CalNex here (you discuss these measurements on lines 429-432). Also need to describe source of isoprene measurements for SEAC4RS.

**line 189 added “SEAC<sup>4</sup>RS isoprene measurements were from proton-transfer-reaction mass spectrometer (PTR-MS) (Wisthaler et al., 2002).**

**Reference:**

**Wisthaler A., Hansel A., Dickerson R. R., Crutzen P. J.: Organic trace gas measurements by PTR-MS during INDOEX 1999, J Geophys Res-Atmos, 107(D19), 8024, 2002.**

5. Lines 230-234: also state that using monthly product  
**line 230 changed “Here, we use collection 06 (MYD04\_L2, <https://ladsweb.nascom.nasa.gov>), retrieved using the Dark Target (DT) and Deep Blue (DB) algorithms (Levy et al., 2015).” to “Here, we used collection 06 (NASA MODIS AOD data archive), retrieved using the Dark Target (DT) and Deep Blue (DB) algorithms (Levy et al., 2015), monthly average data.**

**6. Lines 260-263: give global emissions of isoprene and NO<sub>x</sub> used in the model**

**line 260 changed “Global isoprene emissions are used to calculate an isoprene and NO<sub>2</sub> dependent OA estimate. Global isoprene emissions are from the Model of Emissions of Gases and Aerosols from Nature version 2.1 (Guenther et al., 2006) as implemented in GEOS-Chem and driven with MERRA (MEGAN-MERRA).” To “Global isoprene emissions from the Model of Emissions of Gases and Aerosols from Nature version 2.1 (Guenther et al., 2006) (MEGAN) and satellite NO<sub>2</sub> column data were used to calculate an isoprene and NO<sub>x</sub> dependent OA estimate (see Table 2). Global isoprene emissions from MEGAN were implemented in GEOS-Chem and driven with MERRA (MEGAN-MERRA).”**

**Also see line 654 added “OMI NO<sub>2</sub> column observations were used to represent surface NO<sub>2</sub> levels and surface isoprene emissions from**

**MEGAN were used to represent surface isoprene concentrations, assuming that NO<sub>2</sub> column observations reflect surface NO<sub>2</sub> distributions and isoprene emissions reflect the concentrations of isoprene due to its short lifetime (~1 hr). The detailed implementation is provided in the notes in Table 2. ”**

**line 227-229: changed “Satellite NO<sub>2</sub> column observations are also derived from NASA’s OMI level 3 data, archived at <https://disc.sci.gsfc.nasa.gov> as “OMI-Aura\_L3-OMNO2d” (Lamsal et al., 2014).” To “Satellite NO<sub>2</sub> column observations were also derived from NASA’s OMI level 3 data (Lamsal et al., 2014; NASA OMI NO<sub>2</sub> data archive). Satellite NO<sub>2</sub> observations were used to calculate NO<sub>x</sub> related chemical factor dependent OA estimate (see Table 2).”**

7. Line 271: mid-level of MERRA-2 surface layer is ~60m, not 140m  
Mid-level height of the surface layer should be used here.

Thanks for pointing out this.

**line271 changed “ $\eta(i)$  is the ratio of midday surface layer (~140 m)” to “ $\eta(i)$  is the ratio of midday surface layer (~60 m)”**

8. Line 302: concentrations at STP or under ambient conditions?

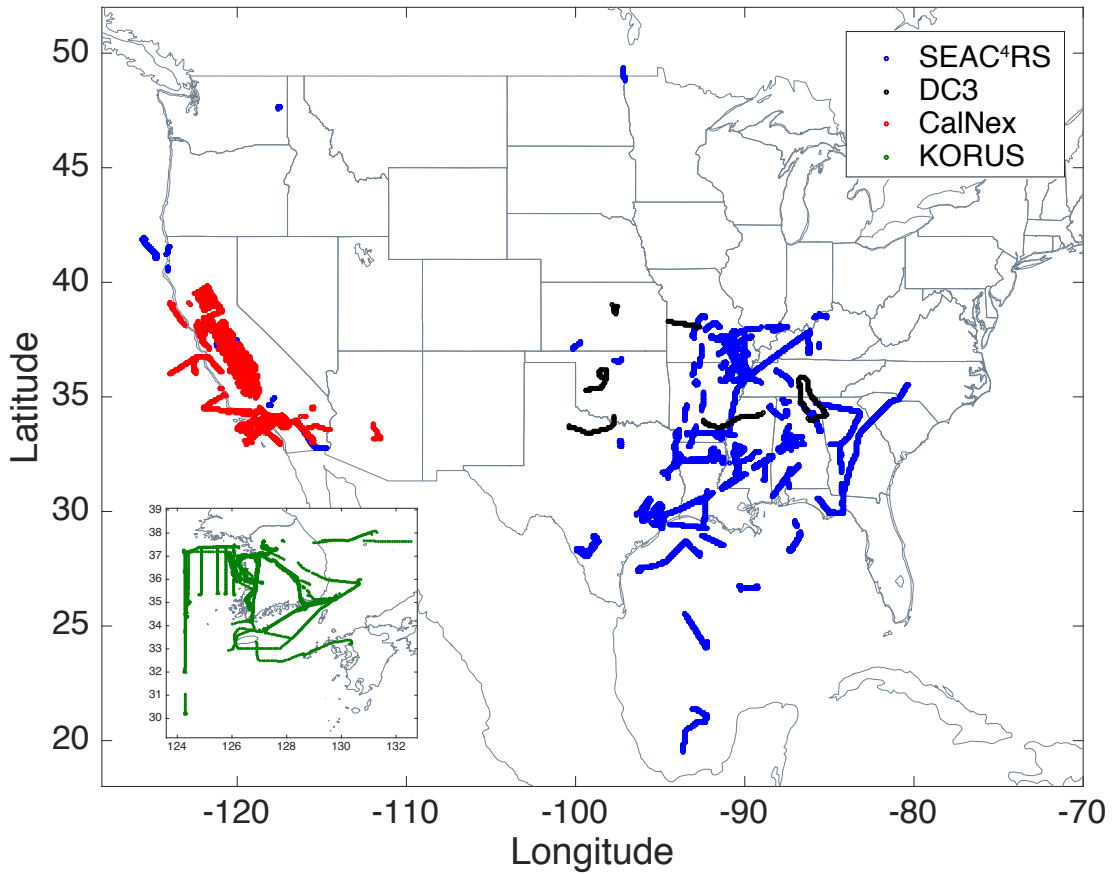
**line 302 changed “surface layer OA concentrations ( $\mu\text{g m}^{-3}$ ) to column OA concentrations ( $\mu\text{g m}^{-2}$ )” to “surface layer OA concentrations ( $\mu\text{g m}^{-3}$ , at ambient T & P) to column OA concentrations ( $\mu\text{g m}^{-2}$ )”.**

9. Lines 305: this is a false statement. Highest PM2.5 levels over the United States are generally in California and in urban regions (see “Our Nation’s Air” report here: <https://www.epa.gov/air-trends>). Please correct.

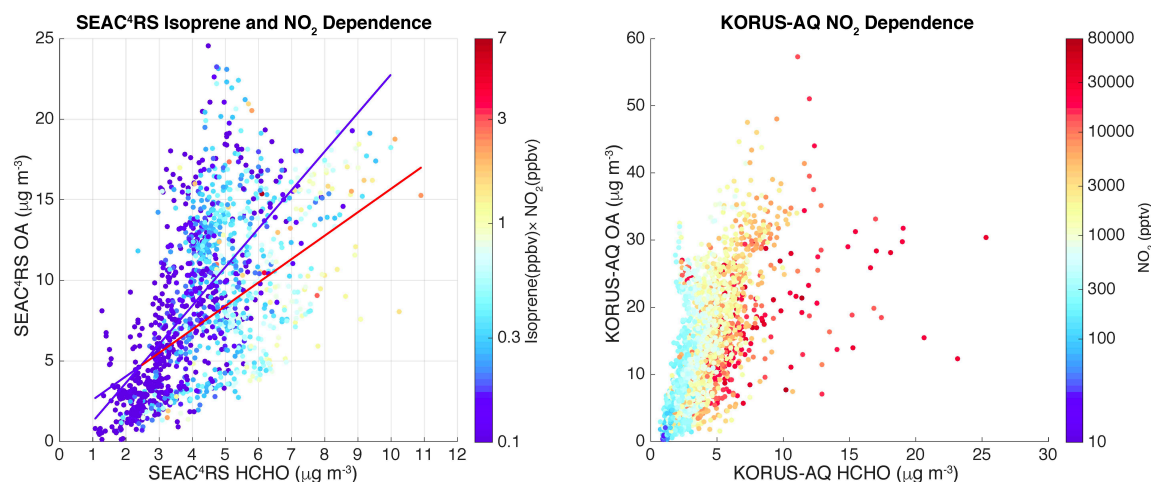
**line 305 changed “The shape of the average vertical profile of OA was very close to that of total aerosol mass over SE US (Wagner et al., 2015) where most of the enhanced aerosol concentrations over the US are located” to “The shape of the average vertical profile of OA (OA fraction: 0.54-0.7) was close to that of total aerosol mass over SE US (Wagner et al., 2015) where a large fraction of the enhanced non-BB aerosol concentrations in summer time over the US are located.”**

10. Figure 1: caption indicates that this shows the data used in this study, but the text indicates that only data < 1km was used. Please modify the figure to show only data below 1 km altitude.

Changed figure 1 as suggested



11. Figure 3a: a character is displaying incorrectly on the color bar label. Thanks for pointing this out. The character is corrected.



12. Section 3.1: The authors did not address my concern about the low number of points used to characterize the HCHO:OA from fires. To explicitly address this please add the number of points used for each category/campaign to Table 1 so that this is clear to the reader. Please also add the caveat “though data is limited” at the end of the sentence “...fires during SEAC4RS.” on line 376.

**line 375-376 changed “The slope of OA to HCHO was higher for wildfires than agricultural fires during SEAC<sup>4</sup>RS.” To “The slope of OA to HCHO was higher for wildfires than agricultural fires during SEAC<sup>4</sup>RS though data were limited (see Table 1).”**

	US (SEAC <sup>4</sup> RS)	US (DC3)	US (CalNex)	South Korea (KORUS-AQ)	Wild Fires (SEAC <sup>4</sup> RS)	Agricultural Fires (SEAC <sup>4</sup> RS)	SEAC <sup>4</sup> RS Low NO <sub>2</sub> and Isoprene	SEAC <sup>4</sup> RS high NO <sub>2</sub> and Isoprene
<b>In situ measurements OA v.s. HCHO</b>								
Slope <sup>a</sup>	1.93 ±0.07	1.30±0.10	1.34 ±0.02	2.75±0.05	25.08±0.30	3.22±0.37	2.39±0.09	1.45 ±0.19
Slope <sup>b</sup> (×10 <sup>-11</sup> )	9.61 ±0.34	6.49±0.49	6.66 ±0.09	13.7 ±0.25	1 ±1.25	0.05 ±1.85	11.9±0.43	7.25 ±0.96
Intercept <sup>c</sup>	0.34 ±0.32	1.10±0.30	-0.90 ±0.06	1.36±0.22	-6.85±2.80	10.41±5.82	-1.14 ±0.37	1.14 ±1.22
Correlation coefficient r	0.59	0.76	0.88	0.70	0.97	0.85	0.64	0.45
Number of points (1 min avg)	1506	134	1772	3425	515	32	1138	226
<b>GEOS- Chem model sampled along the flight track OA v.s. HCHO</b>								
Slope <sup>a</sup>	1.25 ±0.03			1.39±0.05	0.48±0.05			
Slope (×10 <sup>-11</sup> )	6.21 ±0.14			6.95±0.23	2.37±0.22			
Intercept	-1.32 ±0.11			1.88 ±0.07	0.12±0.03			
Correlation Coefficient r	0.76			0.43	0.53			

13. Line 432: you state that the NO<sub>x</sub> range during CalNex was large, but you do not indicate whether the HCHO:OA showed a relationship with NO<sub>x</sub> in this region. Please address.

**Line 432 changed “The tight OA and HCHO correlation during CalNex could be due to the combination of different VOCs sources and NO<sub>x</sub> levels.” To “The OA and HCHO correlation during CalNex was very tight and the slope of OA-HCHO did not show clear dependence on NO<sub>x</sub>, which could be due to the combination of different VOCs sources and NO<sub>x</sub> levels.”**

14. Section 6.1: why was this analysis performed on the monthly time scale? Would a daily analysis have any benefits? Please discuss.

**Line 524 added “The OA estimate was calculated on the monthly time scale, largely because OA estimate is based on OMI HCHO observations and uncertainty weighted average for a time scale of about one month (Gonzalo et al., 2015; Zhu et al., 2016) is needed to reduce the noise in daily OMI HCHO data. With improved satellite HCHO data from TROPOMI, higher time resolution (e.g. weekly average) HCHO data could be useful to estimate OA in the future.”**

15. Line 522: you used (NO<sub>x</sub>)(isoprene) to define the chemical conditions so this is what you should specify here, not “NO<sub>x</sub> levels” alone.

**Line 522: changed (e.g., NO<sub>x</sub> levels) to (e.g., NO<sub>x</sub> and isoprene levels)**

16. Line 527: specify in text what slope is used for Case 1

**Line 527 changed “The monthly average surface OA estimate over the US in August 2013 for case 1” to “The monthly average surface OA estimates over the US in August 2013 using SEAC<sup>4</sup>RS lump-sum slope and intercept”**

17. Line 569: remove “slightly”

**Removed.**

18. Line 604: Section 2.6 does not describe what fraction of AOD comes from OA; how was this determination made here?

**line 603-604: deleted “for the regions AOD is dominated by OA”**

19. Line 629-630: It should be noted in the text that the “GEOS-Chem simulation” and the “OA estimate” are not independent.

**line 629-630 changed “The GEOS-Chem simulation had a coarser resolution than satellite HCHO data.” To “Although HCHO vertical profiles from GEOS-Chem were used in OA estimate, the GEOS-Chem simulation had a coarser resolution than OA estimate.”**

20. Line 640: the authors should briefly explain the four cases in the text to start Section 6.5.

**line 640 added: “OA were estimated with different OA-HCHO relationships for 4 cases (Table 2). LUMP-SUM was using the non-BB SEAC<sup>4</sup>RS campaign lump-sum relationship, the same as shown in Fig. 6; ISOP-NO<sub>x</sub> was using non-BB SEAC<sup>4</sup>RS NO<sub>2</sub> and isoprene dependent relationship; URBAN was using CalNex for large urban cities and SEAC<sup>4</sup>RS lump-sum for other US regions; and COMBINE was using CalNex for large urban cities and NO<sub>2</sub> and isoprene dependent non-BB SEAC<sup>4</sup>RS for other US regions.”**

21. Lines 643-645: the authors indicate that they use NO<sub>2</sub> observations from OMI data. What is the source of the isoprene concentration data needed to apply the (NO<sub>x</sub>)(isoprene) chemical characterization in case 2 and 4? Is this from the GEOS-Chem model? If so, some comparison of how well GEOS-Chem reproduces the observed (NO<sub>x</sub>)(isoprene) during SEAC<sup>4</sup>RS should be added to the manuscript.

See the notes in Table 2 and line 260-263 about the representation of isoprene in chemical characterization in case 2 (ISOP-NO<sub>x</sub>) and case 4 (COMBINE). We aim to use spatiotemporal resolved global dataset (e.g. satellite data or emission inventories) to represent the chemical factor. Isoprene emissions instead of isoprene concentrations are used in chemical characterization. Because the lifetime of isoprene is sufficiently short (~ 1hr), isoprene emissions can generally reflect the isoprene concentrations. Isoprene emission inventory is also easier to access and used by global models to simulate isoprene concentrations. Similarly based on the short lifetime of isoprene, previous studies (e.g., Palmer et al., 2003) have used satellite column concentrations to derive isoprene emissions. Global



isoprene emissions are from MEGAN. See line 261-263: Global isoprene emissions are from the Model of Emissions of Gases and Aerosols from Nature version 2.1 (Guenther et al., 2006) as implemented in GEOS-Chem and driven with MERRA (MEGAN-MERRA).”

Reference:

Palmer P. I., Jacob D. J., Fiore A. M., and Martin R. V.: Mapping isoprene emissions over North America using formaldehyde column observations from space, J Geophys Res-Atmos, 108(D6), 4180, 2003.

**To be clear, line 654 added “OMI NO<sub>2</sub> column observations were used to represent surface NO<sub>2</sub> levels and surface isoprene emissions from MEGAN were used to represent surface isoprene concentrations, assuming that NO<sub>2</sub> column observations reflect surface NO<sub>2</sub> distributions and isoprene emissions reflect the concentrations of isoprene due to its short lifetime (~1 hr). The detailed implementation is provided in the notes in Table 2. ”**

22. Line 780-781: the authors should also indicate that the OA estimate is biased low.

**line 780-781: changed “The OA estimate over the continental US generally correlated well with EPA IMPROVE network OA measurements corrected for partial evaporation.” to “The OA estimate over the continental US generally correlated well with EPA IMPROVE network OA measurements corrected for partial evaporation, with a biased low slope of 0.62 or 0.60, mostly due to underestimation of HCHO concentrations from the OMI HCHO retrieval.”**

23. The manuscript includes many small grammatical errors and some awkward phrasing; it should be edited for language

Edited.