

Volatile and Intermediate-Volatility Organic Compounds in sub-urban Paris: variability, origin and importance for SOA formation: author's response to referee #2.

First, we would like to thank the reviewer for the valuable comments on the manuscript. We answer to the comments addressed by referee #2 and summarize the changes made to the revised manuscript in the following document.

Specific comments:

✓ *Page 4848, Line 26: I am unclear what the abbreviation m a.g.l. is? Please define. Also it is not clear what the time resolution of the adsorbent cartridge measurements were, this needs to be stated for both the IVOC and OVOCs. I think it would be really helpful to the reader to add the offline adsorbent cartridge measurements to Table 2, this table could have two parts one for the online measurements as already shown and another for the offline measurements.*

m a.g.l stands for “meter above ground level”. The sentence starting page 4848 Line 26 in the discussion paper has been modified in the revised manuscript and becomes:

“Gaseous compounds were sampled at approximately 4 m above ground level (a.g.l.)”

The time resolution of both adsorbent cartridges measurements (multi-sorbent cartridges and DNPH-coated cartridges) is 3 hours. This has been defined in the revised manuscript

As for the tables presenting the instruments, we do prefer to present separately the off-line and the on-line measurements. Some parameters differ from one type of measurements to another (e.g. the column sorbent and the analysis associated to each kind of cartridge). The reader could be confused if Table 1 (for the off-line measurements) and Table 2 (for the on-line measurements) are combined.

✓ *Page 4854, Line 15: Why are the OM measurements averaged to 3 hours, the AMS has much higher time resolution? Is this the time resolution of the adsorbent cartridge samples?*

3 hours is the time resolution of the adsorbent cartridge samples (it has been defined in the revised manuscript). This is indeed the reason why we averaged the OM measurements to 3 hours.

✓ Page 4852, Line 23: the authors describe the seasonal difference in the IVOCs (C_{12} - C_{16} n-alkanes) between winter and summer and say that the lower mixing ratios in winter may be due to gas-to-particle partitioning. I think that the difference between winter and summer should be explicitly defined as difference in the gas phase mixing ratios. How are the particles being removed during the gas phase adsorbent cartridge measurements? Could the use of different sampling modules (SASS in summer and ACROSS in winter) be the source of any of these differences? In the conclusions section, the authors state that the 'partitioning from the gas phase to the particulate phase dominates the variability and is enhanced in winter', in my opinion they have not shown this. Previous work (Williams, B. J.; Goldstein, A. H.; Kreisberg, N. M.; Hering, S. V., *In situ measurements of gas/particle-phase transitions for atmospheric semivolatile organic compounds. Proceedings of the National Academy of Sciences of the United States of America* 2010, 107, (15), 6676-6681.) showed that these n-alkanes are >90 % in the particle phase (from measurements and Pankow theory) so I expect enhanced partitioning will only be able to explain a small change in winter/summer concentrations. The authors have everything they need to calculate the gas to particle partitioning of these alkanes using Pankow partitioning theory (Pankow, J. F., 1994, *Atmospheric Environment*, 28, 185-188). This needs to be done to support the stated conclusions. Does it explain the observations?

Following the referee's advice, we calculated the gas-phase partitioning constant K_p , from Pankow theory (Pankow, 1994). We determined the summertime and wintertime K_p for the least volatile compounds we measured during the MEGAPOLI experiments, i.e. hexadecane.

The gas-phase partitioning constant of a compound i , $K_{p,i}$, is defined as (Pankow, 1994):

$$K_{p,i} = \frac{f_{om} 760 RT}{MW_{om} \zeta_i p_{L,i}^{\circ} 10^6}$$

With f_{om} the fraction of total aerosol mass that is organic matter, R the gas constant ($8.2 \times 10^{-5} \text{ m}^3 \text{ atm mol}^{-1} \text{ K}^{-1}$), T the ambient temperature (K), MW_{om} the average molecular weight of organic matter in the aerosol (g mol^{-1}), ζ_i the activity coefficient of the compound i , $p_{L,i}^{\circ}$ the liquid vapor pressure of compound i (Torr).

Table A presents the different parameters needed for the determination of $K_{p,\text{hexadecane}}$ from the SIRTA measurements. Concerning $p_{L,i}^{\circ}$, we determined it from the vapor pressure available in the Reaxys database (<http://www.elsevier.com/online-tools/reaxys>); we corrected it for temperature using Clausius-Clapeyron relationship (Williams et al., 2010). MW_{om} has not been determined for the MEGAPOLI experiments. Williams et al. (2010) have determined a value of 200 g mol^{-1} from ambient measurements of the particulate matter at the University of California, Riverside (USA). Their sampling site is located near an important highway (Docherty et al., 2011). Hence, the influence of

traffic and more generally of the local emission sources is certainly more important at Riverside than at SIRTa. Even though the composition of the organic matter might be different between the two sampling sites, we used the same MW_{om} value than Williams et al. (2010) (200 g mol^{-1}) in our calculations. Indeed, it appears to be a good estimate, especially since, in summer, f_{om} at Riverside (41%) (Williams et al., 2010) is similar than f_{om} at SIRTa (48.9%). As for $\zeta_{hexadecane}$ value, it is somewhere around 6: Chandramouli et al. (2003) have determined from models a mean activity coefficient of $6.5 (\pm 3.6)$ for heptadecane in particles emitted from catalyzed and uncatalyzed gasoline engine exhaust. For this reason, we have set $\zeta_{hexadecane} = 6$.

$K_{p,hexadecane}$ is found to be 2.16×10^{-4} and 8.61×10^{-4} in summer and in winter, respectively. Even though $K_{p,winter} < K_{p,summer}$, these results indicate that the fraction of hexadecane in the particulate phase is not significant in summer and in winter. Hence, hexadecane and the C_{12} - C_{15} n-alkanes IVOCs are mainly in the gas-phase during both MEGAPOLI campaigns. In consequence, we withdraw the conclusions stating that the seasonal variation in the IVOCs concentrations is due to an enhanced partitioning to the particulate phase in winter.

The referee suggests that the seasonal variation in the IVOCs concentration might be due to the different sampling instruments used during the MEGAPOLI campaigns. The two sampling instruments, both developed by TERA Environment, differ mainly by the number of cartridges that can be installed for the measurements. The sampling module is similar: they are both based on a pump coupled to a mass flow controller for air sampling. Detournay et al. (2011) carried on a complete characterization of one of the TERA Environment's sampling system, with identical sampling module than ACROSS and SASS, and they validated the C_{12} - C_{16} n-alkanes sampling by the instrument. At SIRTa, the flowrate was checked before, during and after the field campaigns, to ensure that there was no change in the flowrate. At last, in order to avoid the sampling of particles, we installed stainless-steel particle filters of $2 \mu\text{m}$ porosity (Swagelok®) between the sampling line and the cartridges. Considering all the precautions taken for the measurements, we do not believe that using different sampling systems caused the seasonal variation in the IVOCs concentrations.

Hence, the variation in these compounds concentrations between the two campaigns results more likely from seasonal modification(s) in the strength and/or type of the source emissions. We modified the conclusions in the revised manuscript as to:

“- the seasonal variation of C_{12} - C_{16} n-alkanes of intermediate volatility follows an opposite trend to traditional anthropogenic VOCs with lower concentrations in winter. The variation of these compounds mixing ratios rather results from a change in their emission sources (type, strength) than from the partition to the gas-phase to the particulate-phase of these lower volatility species.”

Table A. Parameters used for the determination of the gas-phase partitioning constant $K_{p,hexadecane}$.

	Summer experiment	Winter Experiment
f_{om} (%)	48.9	36.0
Mean T (K)	291.5	275.1
MW_{om} (g mol ⁻¹)	200	200
$p_{L,i}^{\circ}$ (Torr)	3.43×10^{-4}	5.97×10^{-5}
ζ_i	6	6

✓ In section 3.2, the authors describe the PMF analysis of the AMS data and describe the two factor solution, HOA and OOA. The HOA factor having been further split in other work into HOA-traffic and COA (which I assume is cooking organic aerosol because it is not explicitly defined here). In many previous papers OOA is further split into two factors called SV-OOA (semi-volatile OOA) and LV-OOA (low volatility OOA), I am curious as to whether the authors thought about doing this? Typically, SV-OOA represents fresher SOA which might be expected to form faster and be more associated with gas phase oxidation chemistry while LV-OOA is more aged. I would think that with the approach employed here that the authors would want to be comparing their results to SV-OOA as opposed to the total OOA.

For the summer campaign, two sets of PMF analysis have been performed on the AMS data. Freutel et al. (2013) identified two components (HOA and OOA), while Crippa et al. (2013) further split the components by identifying 5 factors. They split the OOA factor from Freutel et al.'s study (2013) into LV-OOA and SV-OOA. On average, SV-OOA and LV-OOA represent 56% and 44% of OOA, respectively. Our SOA estimation from the measured I/VOCs (46%) is consistent with the semi-volatile fraction of OOA. Hence, we added the following sentence in section 3.2:

“The OOA component has also been separated into two different factors from the PMF analyses of Crippa et al. (2013): semi-volatile OOA (SV-OOA) and low-volatile-OOA (LV-OOA). SV-OOA is considered to be the freshest SOA, i.e. freshly formed, contrary to the more aged LV-OOA (Jimenez et al., 2009). During the MEGAPOLI summer experiment, SV-OOA and LV-OOA represent on average 56% and 44% of OOA, respectively.”

And the following sentences in section 5.3.2.:

“When compared to the semi-volatile fraction of OOA, both Freney et al.’s SOA estimation (2013) and ours are consistent with SV-OOA, which represents 56% of OOA. We could both estimate the freshest OOA.”

Also, we defined COA (as Cooking Organic Aerosol) in the revised manuscript, following the referee’s comment.

✓ *The discussion of the use of an inert tracer for normalization (Page 4864) is very confusing and it is very unclear what was actually done. It seems like no normalization was done but I cannot be sure. This paragraph needs to be significantly reworded for clarity.*

The referee is right: no normalization was done in our study. In this paragraph, we discussed the reasons why we should normalize to an inert tracer and those why we excluded any normalization, especially using either CO or the VOC with the longest atmospheric lifetime. We added one sentence stating clearly that we did not normalize the VOC data, and we modified the two last sentences. Hence, the paragraph becomes:

“- The use of an inert tracer for the normalization: Eq. (7) coupled to Eq. (9) considers that the compounds are in a constant volume from the emission source until the receptor, neglecting the dilution. To overcome the effect of dilution, de Gouw et al (2009) had determined the VOC removal from the temporal evolution of the VOC-to-CO ratio; the removal relies then on the VOC emission ratio instead of a calculated initial concentration. However, here it was not possible to use CO as an inert tracer due to its levels close to background, estimated to be about 100 ppb at an European remote site (Balzani Lööv et al., 2008) (see Fig. 4). Besides, normalization to one of the C₄-C₅ alkanes, which exhibit the longest atmospheric lifetime among all the VOC_s measured during the MEGAPOLI summertime experiment (Table 3), was not feasible since they are emitted from diverse sources (Section 4.2). Finally, no normalization was applied at all.

Other simplifications are considered for the time-resolved approach: it is assumed that (i) no I/VOC is added to the plume between the emission site and the sampling site, which is described as an obvious simplification by de Gouw et al. (2009), (ii) SOA is produced instantaneously from the VOC reaction and (iii) is not subject to loss by deposition (Sjostedt et al., 2011).”

✓ *In Figure 5a, the datapoints are color coded by the O₃/CO ratio as a proxy for photochemical processing, why was this ratio used and not the photochemical age using m,p-xylenes and ethylbenzene that was determined early in the paper? What’s the justification for the use of this different photochemical aging metric used in this plot?*

Off-line measurements of I/VOCs were performed at a time resolution of 3 hours. This is much more than the time resolutions of OM, O₃ and CO measurements, which are 5 minutes, 1 minute and 15 minutes, respectively. By using the O₃/CO ratio, we could work at a lower time resolution than by using the photochemical age. Indeed, since [OH]×Δt is determined from the m,p-xylenes and ethylbenzene measurements performed at SIRTA, this product has also a time resolution of 3 hours. A lower time resolution implies more data and less uncertainty on the scatterplot of OM vs. CO. Hence, Figure 5a actually presents the scatterplots of OM vs. CO, with both OM, CO and O₃ averaged to 1 hour. Also, Figure 5b presents the scatterplots of HOA vs. CO, both were averaged to 1 hour too. In the revised manuscript, we defined the time resolution of the data used in both Figure 5a and Figure 5b in section 5.2.1 and in the captions of these two figures.

✓ *The Bahreini et al., (2012) citation is not correct for the statement made (Page 4867, Line 19). A more appropriate reference is Dallmann, T. R.; Kirchstetter, T. W.; DeMartini, S. J.; Harley, R. A., Quantifying on-road emissions from gasoline-powered motor vehicles: Accounting for the presence of medium and heavy-duty diesel trucks. Environmental Science and Technology 2013, 47, 13873-13881. Figure 3 in this paper clearly shows the emission differences for various pollutants including primary organic aerosol (POA). It should be stated in the paper that this factor of 14±5 is for heavy duty trucks and not passenger vehicles (they are very few diesel passenger vehicles in the US) which may differ in their emission characteristics.*

The Bahreini et al., (2012) citation has been removed and replaced by Dallmann et al. (2013). We also specified that the factor of 14±5 is for heavy-duty trucks. The following sentence, starting Page 4867, Line 19 in the discussion manuscript:

“The important use of diesel by light-duty cars in Europe, and more particularly in France, might explain the higher POA emission ratio determined at SIRTA: POA emission factor, diesel being known to emit 13 times more organic particles than gasoline (Bahreini et al., 2012).”

was modified and becomes:

“The important use of diesel by light-duty cars in Europe, and more particularly in France, might explain the higher POA emission ratio determined at SIRTA: not only diesel emits less CO than gasoline (Allan et al., 2010; Gentner et al., 2013), but POA emission factor is 14 times more important from diesel-fueled vehicles (heavy-duty) than from gasoline-fueled cars (light-duty) (Dallmann et al., 2013).”

✓ *In section 5.2.2, Line 27 should add that the 8 excluded datapoints are shown as grey symbols on Figure 9. During event 5 (“Atlantic polluted”) there is a substantial overestimation of the SOA mass that is not discussed. Can the authors elaborate on the likely cause of this?*

In section 5.2.2, Line 27 of the discussion manuscript, we added that the 8 excluded datapoints are shown as grey symbols on Figure 9.

As observed in Fig. 3, event 5 is characterized by high IVOCs mixing ratios, higher than 40 ppt and up to 120 ppt, while their mean mixing ratio is 21 ± 18 ppt (Table 3). They are 6 times more abundant during event 5 than the average, while the aromatics, which are on average much more abundant than the IVOCs, are only 1.6 times higher than their average mixing ratios (Fig. 3, Table 3). The C₁₂-C₁₆ n-alkanes IVOCs, and more particularly hexadecane which exhibits the highest SOA yield of all the IVOCs (Presto et al., 2010), govern the SOA estimation during event 5. The substantial and sudden increase in the IVOCs and other anthropogenic VOCs concentrations during event 5 might result from punctual emissions from local sources. Therefore, we might consider compounds that did not have enough time to react with OH during event 5. The following paragraph has been added in Section 5.2.2. in the revised manuscript.

“A noticeable overestimation of the SOA mass is observed during event 5, due to high IVOCs mixing ratios (Fig. 4). Hexadecane mixing ratio can reach up to 126 ppt, which is 6 times more than its mean mixing ratio (22 ± 19 ppt, Table 3) and only a third of the toluene mixing ratio in the same sample (344 ppt). Hence, since the SOA yield formation of hexadecane is at least equal to those of toluene (Ng et al., 2007a; Presto et al., 2010), the SOA estimation is mainly driven by the measured IVOCs, especially hexadecane, during event 5. And thus, contribution of the measured IVOCs can overtake the aromatics contribution to SOA formation.”

✓ *In Figure 9, the exclusions of the grey data points should be explained in the caption or at the very least the caption show direct the reader to the explanation in the text.*

Substantial modifications in the SOA formation lead us to remove Figure 9 from the revised manuscript.

✓ *Supporting Information Line 77 – there is a missing citation here, recently Gentner et al., 2013 reported emission factors for VOCs and IVOCs for light duty gasoline vehicles from tunnel measurements. [Citation: D. R. Gentner, D. R. Worton, G. Isaacman, L. Davis, T. R. Dallmann, E. Wood, S. Herndon, A. H. Goldstein and R. A. Harley (2013). Chemically speciated emissions of*

gas-phase organic carbon from motor vehicles and implications for ozone production. Environmental Science and Technology, 47, 11837-11848.]

We added Gentner et al. (2013) as citation in the supporting information.

✓ *Supporting information line 80, what is meant by American oil, does this refer to American gasoline? If so, are the differences between American and European gasoline known, where are they reported?*

We did refer to American fuels when we write “American oil”. The assumption of fuels of different compositions cannot be supported by studies, we have withdrawn this assumption. Hence, we modified the last paragraph of Supplementary material S2 (from line 76):

“As for the IVOCs, little information on the emission factors or emission ratios is available in the literature. Fraser et al. (1998), Schauer et al. (2002) and more recently Gentner et al. (2013) determined emission factor of various VOCs and IVOCs at the exhaust of gasoline light-duty cars. However, these results did not seem appropriate to our study since they have been determined under the Federal Test Procedure (FTP) urban driving cycle (Schauer et al., 2002) or real American driving conditions (Fraser et al., 1998; Gentner et al., 2013) and, then, are representative rather of the American fleet of vehicles than of the French one. Hence, ER determined from Fontaine’s EF (Fontaine, 2000) appear to be the best estimates for the C₁₂-C₁₆ alkanes.”

Technical corrections:

✓ *Page 4844, Line 10: Reword ‘If a high density of population characterizes the megacities, they are also remarkable by their...’ to read something like ‘If a high population density characterizes a megacity, they are also defined by their...’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4845, Line 13: Replace ‘qualified’ with ‘defined’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4845, Line 14: Replace ‘any’ with ‘either’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4848, Line 18: Add 'n-alkanes' after 'their C*,C₁₂-C₁₆'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4848, Line 19: Replace 'are' with 'were'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4850, Line 26: Replace 'great' with 'high'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4851, Line 8: Too many significant figures are shown considering the uncertainties, round these to read '(440 ± 220 m and 1340 ± 610)'*

Modification of the text has been made in the revised manuscript.

✓ *Page 4851, Line 13: Replace 'which the' with 'these is'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4853, Line 9: Suggest rewording 'suggesting their secondary origin' to read 'suggesting they were predominantly secondary in origin'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4854, Line 4: Add 'only a' before 'few sunny days. ...'*

Modification of the text has been made in the revised manuscript.

✓ *Page 4854, Line 4-5: Remove 'equal to' from the sentence 'average temperature was equal to 18.3 (± 3.8) m s⁻¹'.*

Modification of the text has been made in the revised manuscript.

- ✓ *Page 4854, Line 7: Remove 'equal to' from the sentence 'average wind speed was equal to 3.7 (\pm 1.5) m s⁻¹'.*

Modification of the text has been made in the revised manuscript.

- ✓ *Page 4854, Line 16: Replace 'which is equal to' with 'of'.*

Modification of the text has been made in the revised manuscript.

- ✓ *Page 4855, Line 1: Replace 'a' with 'one'.*

Modification of the text has been made in the revised manuscript.

- ✓ *Page 4855, Line 17: Add 'predominately' after 'CO is'.*

Modification of the text has been made in the revised manuscript.

- ✓ *Page 4855, Line 19: The sentence 'This may indicate that , during event 4, CO is of primary but also of secondary origin and/or issuing from long range transport, according to its long atmospheric lifetime which is about a month (Parrish et al., 1998). These assumptions are supported by...' does not make sense as written, this needs to be reworded for clarity.*

The sentence has been reworded as following:

“Hence, during event 4, CO might be of both primary and secondary origin. Besides, since it exhibits a substantial atmospheric lifetime of about a month (Parrish et al., 1998), long range transport of CO cannot be discarded either.”

- ✓ *Page 4856, Line 15: Remove 'the' before 'megacities'.*

Modification of the text has been made in the revised manuscript.

- ✓ *Page 4863, Line 20: Add 'the' after 'based on'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4863, Line 21: add 'of' after 'downwind'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4863, Line 22: Remove 'also' after 'Boynard et al., (2014)' and replace 'are' with 'were'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4863, Line 28: Replace 'change' with 'have changed'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4684, Line 24: Replace 'have' with 'had'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4865, Line 11: Replace 'any of the two' with 'both the'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4865, Line 11: Replace 'does not include the biogenic' with "did not include any biogenic".*

Modification of the text has been made in the revised manuscript.

✓ *Page 4865, Line 22: Reword 'until now, estimations have only considered the SOA formation in dry conditions' to read 'typically estimations only consider SOA formation under dry conditions'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4865, Lines 25-26: Reword 'Carlton et al., (2008) have improved the agreement between the observations and the prediction in the total mass concentrations and the variability', to read*

'Carlton et al., (2008) improved the agreement between observations and predictions of total mass concentrations and its variability,'.

Modification of the text has been made in the revised manuscript.

✓ *Page 4868, Line 1: Add 'likely' after 'also' and replace 'the' with 'an'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4866, Line 2: Remove 'the'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4866, Line 14: Replace 'consists in' with 'consists of'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4867, Line 4: Replace 'is equal to' with 'was'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4867, Line 9: Replace 'is equal to' with 'was'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4867, Line 19: replace 'Besides' with 'Also'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4868, Line 18: Rewrite 'It can be seen that light alkanes, which have the highest emissions, exhibit a low or no SOA yield formation' to read 'It can be seen that the light alkanes, which have the highest emissions, exhibit low to no SOA formation potential'.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4868, Line 19: Remove this ‘Indeed, their emission in the gaseous phase is enhanced by their high-volatility’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4868, Line 24: Replace ‘exceed’ with ‘exceeded’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4869, Line 4: Replace ‘by considering VOCs and the sum of VOCs and IVOCs, with ‘by using VOCs or IVOCs (sum of VOCs and IVOCs),’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4869, Line 1: Rewrite ‘the SOA estimated’ to read ‘the estimated SOA’.*

Modification of the text has been made in the revised manuscript.

✓ *Page 4869, line 21: Remove ‘a’ before ‘SOAformation’.*

Modification of the text has been made in the revised manuscript.

✓ *Figures 2 and 3 captions: Replace ‘The grey-shaded areas highlight the “Atlantic Polluted” wind regimes associated to stagnant conditions and corresponding to an OOA increase’ with ‘The grey-shaded areas highlight the “Atlantic Polluted” wind regimes associated with stagnant conditions and increased OOA’.*

Modification of the text has been made in the revised manuscript.

✓ *Figure 8 caption: This should include the definition of the grey bowes as previously defined in Figures 2 and 3. Add ‘The grey-shaded areas highlight the “Atlantic Polluted” wind regimes associated with stagnant conditions and increased OOA’.*

Modification of the text has been made in the revised manuscript.

✓ *Figure 9 caption: Replace ‘are’ with ‘were’.*

Modification of the text has been made in the revised manuscript.

✓ *Supporting Information, Line 56: Replace 'on' after represented with 'in'.*

Modification of the text has been made in the revised manuscript.

✓ *Supporting Information, Line 62: Change '... significantly over the last decade at $\pm 30\%$.' to read 'significantly ($\leq 30\%$) over the last decade'.*

Modification of the text has been made in the revised manuscript.

✓ *Supporting Information, Line 63: Replace 'leaded' with 'led'.*

Modification of the text has been made in the revised manuscript.

✓ *Supporting Information, Line 75: Replace 'few' with 'little'.*

Modification of the text has been made in the revised manuscript.

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