Volatile and Intermediate-Volatility Organic Compounds in sub-urban Paris: variability, origin and importance for SOA formation: author’s response to referee #1.

First, we would like to thank the reviewer for the valuable comments on the manuscript. We answer to the comments addressed by referee #1 and summarize the changes made to the revised manuscript in the following document.

- ✓ *My first major concern is the paper’s framing of IVOCs – The manuscript only considers 5 IVOCs (all n-alkanes) but it seems to equate them with “IVOCs”. For example, the abstract states that “10% (of the SOA is) explained by only C_{12}-C_{16} IVOCs.” This statement is not correct. The only IVOCs that paper accounts for SOA formation from is C_{12} to C_{16} normal alkanes. That is a small subset of the IVOCs. There are likely many many more C_{12} to C_{16} IVOCs that were not measured that therefore not considered (branched, cyclic, PAH, etc.). This issue was mentioned in the discussion but the statement like that given above in the abstract will likely confuse many readers because the text implies in many places that they are estimating the SOA from IVOCs not 5 n-alkanes. By only measuring a few compounds, the paper is likely only exploring th so called tip of the iceberg when it comes to SOA formation from IVOCs. For example, Fraser et al. (Fraser, M. P.; Cass, G. R.; Simoneit, B. R. T.; Rasmussen, R. A., Air quality model evaluation for organics. 4. C2-C36 non-aromatic hydrocarbons. Environ. Sci. Technol. 1997, 31, (8), 2356-2367) demonstrates that the vast majority of IVOC mass in Los Angeles is not n-alkanes (or simple aromatics, etc.). The paper needs to use more precise language to not confuse the reader into thinking it is provides a comprehensive estimate of SOA from IVOCs.*

We do agree that our use of the “IVOC” term can be sometimes confusing, especially in the abstract. To avoid such confusion, we modified the text so the reader understands that our study focuses on 5 n-alkanes. Hence, every time it was needed, we specified “C_{12}-C_{16} n-alkanes” IVOCs. In the discussion manuscript:

P 4843, Line 26: “when the IVOCs are taken into account” becomes “when the C_{12}-C_{16} n-alkanes are taken into account”;

P 4868, Line 16: “by the IVOCs” becomes “by the C_{12}-C_{16} n-alkanes IVOCs”;

P 4868, line 25: “the IVOCs up to C_{16}” becomes “the n-alkanes IVOCs up to C_{16}”;

P 4869, Line 4: “VOCs and IVOCs” becomes “VOCs and C_{12}-C_{16} n-alkanes IVOCs”;

P 4869, Line 14: “and IVOCs” becomes “and C_{12}-C_{16} n-alkanes IVOCs”;

P 4870, Line 10: “the IVOCs” becomes “the C_{12}-C_{16} n-alkanes IVOCs”;

P 4871, Line 16: we added “(C_{12}-C_{16} n-alkanes)” after “some IVOCs were taken into account here” so it reads “some IVOCs were taken into account here (C_{12}-C_{16} n-alkanes)”;

P 4872, Line 15: “the seasonal variation of IVOCs” becomes “the seasonal variation of C_{12}-C_{16} n-alkanes of intermediate volatility”.

- ✓ *The second major point is that I have some problems with the SOA production estimates. The SOA yields for the IVOCs are taken from Lim and Ziemann. These experiments measured yields at very high concentrations (in excess of 500 ug/m3) in a smog chamber. These concentrations are substantially higher than the ambient concentration of 0.2~9 ug/m3. This completely biases the gas particle partitioning and will cause the yields to be overestimated. This point is mentioned in the text, but the authors do not do any analysis to try to quantify the potential bias. Simply applying the yields of Lim and Ziemann to the atmosphere is completely unrealistic and will greatly overestimate the amount of SOA from these compounds. The authors need to quantitatively examine this bias. One way would be estimate the partitioning bias in the Lim and Ziemann data using other n-alkane yield parameterizations. Another would be to simply use the high-NOx yields for n-alkanes which were measured at atmospherically relevant concentrations of Presto et al. (2010). The bottomline is the SOA mass yields of C9-C_{16} are substantially overestimated if the effect of OA concentrations on SOA yields was not taken into account.*

Following the referee’s comment we have changed the SOA yields used in our estimations. We investigated the SOA formation from the measured IVOCs degradation using the particulate yields determined by Presto et al. (2010). They have determined the SOA yields for the C_{12}-C_{16} n-alkanes under high-NOx conditions but at $0.1\ \mu g\ m^{-3} < C_{OA} < 50\ \mu g\ m^{-3}$. We used the SOA yields determined at $C_{OA}=2\ \mu g\ m^{-3}$, which is more atmospherically relevant considering the average OA ($1.8\ \mu g\ m^{-3}$) measured at SIRTA in July 2009. As for the aromatic compounds, we also used their more atmospherically relevant SOA yields, i.e. those determined during chamber experiments under low-NO_x conditions but with $C_{OA}=40\ \mu g\ m^{-3}$. Hence, in order to fairer compare the aromatics contribution with the IVOCs contribution to SOA formation, we also investigated the SOA formation using the C_{12}-C_{16} n-alkanes yields determined at $C_{OA}=40\ \mu g\ m^{-3}$,(high-NO_x) (Presto et al., 2010), and the aromatics yields determined under high-NOx conditions ($C_{OA}=40\ \mu g\ m^{-3}$) (Ng et al., 2007) following the integrated approach. As for the C_9-C_{11} n-alkanes, we used Lim and Ziemmann's SOA yields (2009) since they are the most recent SOA yields.

Hence we modified the SOA yields of the C_{12}-C_{16} n-alkanes and the aromatic compounds presented in Table 3. For these compounds, we show two set of values: the first is the one determined under the most relevant atmospheric conditions, i.e C_{OA}= 2 µg m^{-3} and high-NO_x for the IVOCs and C_{OA}= 40 µg m^{-3} and low-NO_x for the aromatics; the second is the one determined under similar conditions (C_{OA}= 40 µg m^{-3}.and high-NO_x) for both IVOCs and aromatics.

- ✓ *One consequence of using very high yields is that the few normal alkanes IVOC measured by this study contribute a surprising large fraction (10%) of the SOA. In diesel exhaust (likely the most important source of these normal alkanes) C_{12}-C_{16} n-alkanes contribute less than 10% of the IVOC emissions (Schauer et al., 1999 EST). As a result, the amount of SOA formed solely by IVOCs could be greater than the measured SOA (without taking into account other processes such as aqueous reactions). Estimation of SOA production from the C_{12}-C_{16} needs to be revisited.*

Following the referee's previous comment, we revisited the SOA production from the C_{12}-C_{16}. With the new SOA yields, under the environmental conditions encountered at SIRTA (C_{OA}= 2 µg m^{-3}), we estimated the SOA mass from these compounds to be 2% (integrated approach) and 8% (time-resolved approach).

There were a fair number of confusing statements with respect to SOA formation.

- ✓ *Page 4863 – In the same paragraph you say "SOA yields determined under low-NOx conditions were used when available (see Supplement, Sect. S3 for details)." But then, two sentences later you say "However, for more consistency, only the SOA yields determined under high-NOx conditions are used here." Which is it?*

Ideally, we would have used SOA yields determined under low-NOx conditions and low C_{OA}, since those experimental conditions are the closest to the ambient conditions encountered at SIRTA during the MEGAPOLI summertime campaign. SOA yields of the aromatic compounds were determined under such conditions, since C_{OA}~40µg m^{-3} and [NO_x] <1 ppb, on average (Odum et al., 1997; Ng et al., 2007). We used these SOA yields for the aromatic compounds. As for the IVOCs, dodecane is the only IVOC of interest in this study whose SOA yield has been determined under both low-NO_x and low C_{OA} conditions (Cappa et al., 2013; Loza et al., 2014). However, many studies have investigated the SOA yields of the C_{12}-C_{16} n-alkanes IVOCs under high-NO_x concentrations and/or high C_{OA} (Lim and Ziemann, 2005, 2009; Jordan et al., 2008; Presto et al., 2010). Instead of considering IVOCs SOA yields determined under very different experimental conditions, we rather used yields determine under similar conditions, i.e. high-NO_x and high C_{OA}, even though they are less representative of the atmospheric conditions at SIRTA in summer 2009. Thus, we could discuss about the contribution of the C_{12}-C_{16} n-alkanes IVOCs to SOA formation compared to those of the aromatic compounds. Finally, we used the Lim and Ziemann's SOA yields (2009), in the discussion manuscript.

We modified this paragraph (starting Page 4863 in the revised manuscript), taking into account the modification in the SOA estimation induced by the new IVOCs SOA yields (see the answer to the referee's previous comment). The paragraph becomes:

"- The SOA yields: The SOA yields used here (provided in Table 3) are the most recent ones determined in chamber experiments, and the most relevant to the MEGAPOLI campaign conditions. SOA yields are known to be highly influenced by the experimental conditions in the chamber, especially by the NO_x concentrations and the organic mass concentrations (C_{OA}) (Hildebrandt et al., 2009; Presto et al., 2010; Aumont et al., 2012; Tkacik et al., 2012). At SIRTA, on average, C_{OA}=1.8 µg m^{-3} and the NO_x concentration is 6.9±4.8 ppb. These environmental conditions correspond to low-C_{OA} and low-NO_x conditions, regarding chamber experiments. While chamber experiment conditions under which SOA yields are determined are quite variable, they never meet both the low-NO_x and low-C_{OA} criteria. SOA yields of the aromatic compounds were either determined at low-NO_x and high NO_x (Ng et al., 2007) conditions but always at high C_{OA}=40 µg m^{-3} (see supplementary material, Section S4 for details). On the opposite SOA yields of the volatile and C_{13}-C_{16} intermediate volatile n-alkanes were determined at high or low C_{OA} but always under high NO_x conditions (Lim and Ziemann, 2009; Presto et al., 2010). Therefore, the SOA formation will be studied using two sets of SOA yields for the C_{12}-C_{16} n-alkanes and the aromatics, from the integrated approach only. The first SOA yield values reported in Table 3 are those determined under the most relevant atmospheric conditions for this study, i.e low C_{OA}=2 µg m^{-3} (but high-NO_x) for the IVOCs and low-NO_x (but high-C_{OA}=40 µg m^{-3}) for the aromatics. The second SOA yield values are those obtained from chamber experiments performed under similar C_{OA} and NO_x conditions, i.e. high-NOx and high C_{OA}=40 µg m^{-3}.The second set of SOA yields will be used to compare the contribution to SOA formation of the C_{12}-C_{16} n-alkanes against the contribution of the whole VOC set. C_9-C_{11} n-alkanes SOA yields are those determined by Lim and Ziemann (Lim and Ziemann, 2009) under high-NO_x and high C_{OA} (400µg m^{-3}<C_{OA}<1 600 µg m^{-3}) conditions. High-NO_x SOA yields are much more important than low-NO_x SOA yields for the n-alkanes (Loza et al., 2014). Besides, the higher C_{OA} is, the higher the SOA yield is (Hildebrandt et al., 2009; Presto et al., 2010; Aumont et al., 2012; Tkacik et al., 2012).".

- ✓ *“C_{OA} in chamber experiments is ten to hundred times higher than the ambient organic mass concentrations.” This is true in some experiments (e.g. those of Lim and Ziemann) but that is not universally true.*

We agree with the referee; the statement is not universally true. We have withdrawn the sentence (see the answer to the referee’s previous comment).

- ✓ *Table 3 – I found the yields very confusing. I realize these are yield to CO, but you are forcing the readers to do unit conversions to compare with the original sources. The authors should report the yields in the standard (and much more interpretable) units of mass SOA/mass precursor reacted.*

The yields reported in table 3 are not yield to CO. The yields presented in Table 3 are already expressed in units of mass SOA (µg m^{-3})/mass precursor reacted (ppm); their unit is then µg m^{-3} ppm^{-1}. Hence we did not modify the SOA yields unit from Table 3.

- ✓ *CO as tracer of anthropogenic emissions, especially using VOC/CO ratios. CO (at least in the US) is dominated by emissions from gasoline vehicles. I am not sure in Europe. Gasoline vehicles are only one class of anthropogenic source. How robust is it to use these ratios, especially for IVOCs which are likely emitted by other (non-gasoline vehicle sources)?*

The determination of the emission ratios of IVOCs over CO are derived from the nonane-to-CO ratios from ambient observations in Paris and from the IVOC-to-nonane emission ratios derived from emission factors weighted by the proportion of diesel and gasoline motorizations in France (see section 4.1). Therefore these ratios take into account the relative importance of both classes of vehicles. So does the resulting IVOC-to-CO emission ratio. However one cannot exclude that this emission ratio would be different in the US. Indeed, in Europe as in the US, CO is dominated by emissions from gasoline vehicles and IVOCs are rather emitted from diesel-fueled engine (Schauer et al., 1999, 2002; Gentner et al., 2013). We estimated that the car fleet in Paris during the summertime MEGAPOLI experiment comprised 1.5 times more diesel cars than gasoline vehicles (Section 4.1 of the discussion manuscript). Thus, CO emissions from gasoline cars represent 60% of the total CO emissions in the Paris area, while diesel cars contribution to CO emissions is 40%. The importance of diesel vs gasoline emissions for CO and IVOCs in the US might influence the value of this ratio.

- ✓ *“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, diesel being known to emit 13 times* *more organic particles than gasoline" It is not only that diesel emit more POA than gasoline powered cars, but they also likely emit less CO so one needs to consider both components in the OA/CO ratio.*

We agree with the referee. Thus, we modified the sentence cited by the referee as following:

"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 the 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)."

- ✓ *Page 4868, line 13-14: "The I/VOC precursors explain 45% of the SOA measured at SIRTA". However, the SOA production from I/VOC was estimated using the integrated approach. This approach overestimates SOA production by assuming complete reaction of all precursors to form SOA. Since the time resolved approach was also used in this study and better predict the actual SOA production in the atmosphere, the contributions of I/VOC precursors to SOA using this approach should be included in Fig. 6.*

Following the referee's comment, we completed Fig. 6 by adding the results of the SOA estimation from the time-resolved approach.

- ✓ *2) Page 4852, line 23-29 (also in the abstract): The study reports a higher mixing ratio of* C_{12}*-*C_{16} *in the summer than the winter. They speculate that this was due to gas/particle partitioning. I am skeptical of this claim. These are very volatile species – the least volatile (*C_{16} *normal alkane) has* C^* *greater than 50,000 ug/m3. therefore it seems extremely unlikely that appreciable amounts of any of these species would partition into the condensed phase under any atmospheric conditions. Furthermore, the modest summer-winter swing in temperature will only modestly change the vapor pressure (this change can easily be estimated with measured temperature and clausius clapeyron). It seems much more likely that some other process (change in the emissions, change in boundary layer height) is driving the seasonal changes in IVOC concentrations. If the authors want to argue that it is partitioning then they need to provide some quantitative evidence for it. The authors did cite some studies that suggest seasonal partitioning effects – I looked up one of these references up (Bi et al. 2003). That study used filter followed by PUF. It is likely that the small amounts of IVOCs that they sampled in this carbon number range are simply sampling artifacts (adsorbed vapors on filters) as opposed to actual particle phase organics.*

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):

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

With f_{om} the fraction of total aerosol mass that is organic matter, R the gas constant (8.2×10^{-5} m^{-3} atm mol^{-1} 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, $p^{\circ}_{L,i}$ the liquid vapor pressure of compound i (Torr).

Table A presents the different parameters needed for the determination of $K_{p,hexadecane}$ from the SIRTA measurements. Concerning $p^{\circ}_{L,i}$, 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 (±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 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:

"- 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
$p^{\circ}_{L,i}$ (Torr)	3.43×10^{-4}	5.97×10^{-5}
ζ_i	6	6

- ✓ *Abstract "including for the first time C_{12}-C_{16} n-alkanes of intermediate volatility (IVOCs), suspected to be efficient precursors of secondary organic aerosol (SOA)." This statement is not true. In fact this paper cites some other studies that have measured n-alkanes. (Kadowaski, 1994; Bi et al., 2003; Guo et al., 2009). My favorite was not referenced – Fraser, M. P.; Cass, G. R.; Simoneit, B. R. T.; Rasmussen, R. A., Air quality model evaluation data for organics. 4. C2-C36 non-aromatic hydrocarbons. Environ. Sci. Technolo. 1997, 31, (8), 2356-2367. Previous models have also explicitluy accounted for the large n-alkanes on SOA formation (see e.g work of Havala Pye), chamber experiments to Jathar et al. (ACP 2012), etc.*

We have withdrawn "for the first time" from this sentence in the abstract. As for the reference the reviewer suggested to add, we did not add it in this paragraph, since we reported studies investigating the C_{12}-C_{16} n-alkanes concentrations during both summer and winter. Fraser et al. (1997) measured these compounds only in summer (September 8-9, 1993).

- ✓ *The paper performed comprehensive speciation analysis of organic gases. Both speciated VOCs and NMOC were measured, it would be great to show the fraction of speciated VOCs are NMOC.*

We agree with the referee: we did perform a comprehensive speciation of the organic gases which would be interesting to discuss. However, we are afraid that such discussion in this manuscript could be stepped aside by all the information already presented. Hence, we do not show the fraction of speciated VOCs and NMOC in this manuscript. And actually, we currently prepare another manuscript on the gaseous organic compounds measured at SIRTA during the MEGAPOLI experiments. We will then discuss on the prominence of the NMOC over the VOCs.

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