Previous reviewer comments, amended by editor suggestions:

Reviewer 1, comment 1: Please be more specific how and where references were added and novel aspects are discussed.

Response. References were added as follows:

Section 3 (Model Description): The first sentence of this section refers to previous works in which the UNIPAR model has been demonstrated for other precursors. The sentence immediately following establishes that this study's novel aspect is the extension to linear alkanes.

Section 3.2 (Lumping and aging): The first sentence of this section establishes that the dynamic lumping array structure as well as the incorporation of aging was developed in previous studies. The sentence immediately following establishes that alkane products were lumped in this study according to that previous structure. The last sentence of this references a previous paper in which some of the information regarding the lumping structure and aging within this section was discussed in more detail.

Section 3.3: This section is entirely novel to this study and thus, is described with no references to previous development of the model.

Section 3.4 and 3.5: These sections outline equations determining the partitioning and particle-phase reactions present within the model. Each equation has appropriate references to previous works in which these equations were developed.

Section 3.6: The second sentences of this section explains that the method used in this section was developed in previous studies,

Reviewer 1, comment 2: (also Reviewer 2, comment 2): Both reviewers pointed out that the SOA burst in the morning needs more explanation. Referring to previous studies where this has been observed as well is not sufficient. I restate the reviewer's suggestion to do a sensitivity test and check whether your explanation in Section 4.3 may be valid that a high OH concentration may have caused this peak. Is the required OH concentration even realistic? What other factors could lead to such a peak? Were the conditions in the limonene study by Yu et al comparable to yours?

Response. In order to response to the comment from the reviewer and the editor, the additional chamber experiment has been conducted for C13 with the lower hydrocarbon (35ppb) and HONO (35ppb) concentrations that those in Table 1. The spike in SOA mass was significantly reduced and SOA mass was better predicted with the UNIPAR alkane model.

Similar to alkane of this study, the fast reaction of d-limonene with oxidants at low NOx levels can yield the rapid SOA growth with low volatility products. If d-limonene SOA forms with the low concentration of hydrocarbon, the spike in SOA mass will not occur.

The newly added sentence can be found in the end of Sect. 4.3 and reads now,

"When both the concentrations of HONO and the long-chain alkane are reduced, this spike can be suppressed as seen in Figure S3"

Reviewer 1, comment 4: I agree with the reviewer that it is very confusing to use identical symbols for different quantities. Please choose different symbols for different parameters.

Response: The symbol for polarizability has been changed to P_i rather than α_i.

Reviewer 2, comment 5(What are the levels of Hox and Nox in the chamber for the duration of each experiment? Both would undoubtedly affect RO2 fates and lifetimes, neither of which is discussed in the section of Nox impact on autoxidation. Authors go on to attribute the decrease in SOA yield with Nox to the partitioning of non-autox products, but that explanation is unclear and buried in a Table in the SI. A clearer demonstration with a graphic is needed.): How and where was this reviewer comment addressed? Just adding a figure is not sufficient as an explanation.

Response: Figure S4 was added to the SI as a clearer demonstration of the impact of alkane autoxidation products on yield differences between high and low NOx conditions, as well as different carbon number precursors, as demonstrated by a model sensitivity test. This replaced a previous table in which the fraction of autoxidation mass was reported for various high and low NOx experimental simulations, which was relatively difficult to interpret. The new Figure S4, however, is a straightforward figure which supports our assertion that "The fraction of autoxidation products to the total SOA mass generally increases with decreasing carbon number or decreasing NO_x level due to gas-particle partitioning of non-autoxidation lowly volatile products…". Additionally, figure S5 was added which shows the HOx and NOx concentrations from the sensitivity test, as well as the integrated reaction rates of selected RO2 species. This contributes to the added discussion of RO2 fates :

"Additionally, the impact of the autoxidation reactions on RO2 chemistry is examined using the Integrated Reaction Rates in Fig S6. Generally, the autoxidation reactions of selected RO2 species displayed (HO3C106O2, and HO3C126O2 from the MCM mechanisms of C10 and C12, respectively) were insignificant compared to reactions with NO. Additionally, the autoxidation reaction rate was slightly increased under low NOx conditions compared to high NOx conditions, which is consistent with Fig. S5."

Reviewer 2, comment 6: The reviewer asked for an optimization of model parameters. They also pointed out that your conclusions on the role of straight-chain alkanes may be misleading. Just adding another reference does not suffice to back up your model-based conclusions. Please address the reviewer comment "the authors conclude from this modeling exercise that straight chain alkanes are important for urban SOA - without showing much evidence. This is not backed up by the results shown" by justifying in detail your conclusions despite the extreme extrapolations that implies.

Response: The determination of the model parameters was performed by using the solver in the spreadsheet and are thus already optimized.

In order to response to the reviewer's comment, we modified our claim which now reads:

"Thus, we conclude that long-chain alkanes are an important source of SOA formation in air parcels originating from diesel fuel combustions."

Reviewer 2, comment 7(Line 153: Why 51 species? Where is this from?): Is this information added in the text?

Response: Yes, this information is in the text : "The α_i array consists of 6 different reactivity levels (very fast, fast, medium, slow, partitioning only, and multi-alcohol) and 8 different volatility levels based on vapor pressure which represent 48 species, along with 3 explicit species that are lumped separately (glyoxal, methylglyoxal, and epoxydiols)."

Additional editor comments:

Scientific:

l. 83: In the abstract, you write that in addition to NOX levels and seed conditions also temperature was controlled. In addition, referee 2 asked how light conditions were monitored. Please be consistent and clear which conditions where controlled and how.

Response: Temperature conditions were not controlled within the experiment. It is clear how this can be confusing when temperature is mentioned alongside NOx and seed conditions and thus, "temperature" has been removed from that line in the abstract. The information on how temperatures, RH, and light conditions were monitored can be found within the "Experimental section":

"A hygrometer (CR1000 measurement and control system, Campbell Scientific Inc., USA) was used to measure meteorological factors (temperature, relative humidity (RH) and an ultraviolet radiometer (TUVR, Eppley Laboratory Inc., USA) was used to measure sunlight intensity"

l. 121: It is not clear what you mean by 'The UNIPAR model has been demonstrated...'. What was demonstrated? E.g. good performance to predict SOA formation ? Or do you mean '... has been applied...'?

Response: This sentence has been updated: "The UNIPAR model's ability to accurately simulate SOA formation from various aromatic HCs (Im et al., 2014; Zhou et al., 2019; Han and Jang, 2022a), monoterpenes (Yu et al., 2021), and isoprene (Beardsley and Jang, 2016) has been previously demonstrated."

l. 130: What do you mean by 'mathematically' here? Can it be omitted? If not, please explain.

Response: The word "mathematically" has been removed.

l. 132: In the abstract, you state that there are three main pathways. Here, you refer to two pathways. Be consistent to avoid confusion.

Response: This sentence has been updated and now reads: "The SOA mass in the model forms via three pathways: OM produced via multiphase partitioning of organic products (OM_P),aerosol phase reactions of organic species to form OM_{AR} via oligomerization in the *org* phase, and reactions in the wet *inorg* phase which also form OM_{AR} (acid-catalyzed oligomerization and organosulfate (OS) formation)."

l. 139: What do you mean by 'The atmospheric process of alkanes? 'Atmospheric oxidation'?

Response: Yes. "atmospheric process" has been replaced with "atmospheric oxidation."

l. 150/151: Please revise this sentence. It is not clear what exactly is lumped.

Response: This sentence has been divided into two sentences to improve clarity and now reads:

"The lumping structure of the UNIPAR model, along with a dynamic α_i array which considers aging, has been developed in previous studies (Zhou et al., 2019; Han and Jang, 2020; Yu et al., 2021). According to this structure, the alkane oxidation products, originating from simulations at various NO_x levels (HC ppbC/NO_x ppb = 2~50) using the MCM and alkyl peroxy radical autoxidation mechanisms, were lumped ."

l. 158: What are 'photolytic products'? And why do you write 'or'? It seems that products could be more reactive and less volatile and originating from photolysis. Or did I misunderstand what you mean here?

Response: Photolytic products are products produced via photolysis by "photolysis products" may be clearer language. Generally atmospheric aging will result in products which are more reactive and less volatile. The general exception is when compounds undergo photolysis in the atmosphere which will create products which are more volatile, and possibly more reactive.

This sentence has been updated for clarity and now reads: "This atmospheric aging can augment the product distribution, forming more reactive and less volatile products via oxidation, or photolysis products which are more volatile but may be more reactive."

l. 176: Again, the use of 'mathematically' is not clear here. If it is necessary, please explain what you mean here.

Response: The word "mathematically" has been removed from this sentence.

Section 4.2: This section is very descriptive and brief though it is a key section of your paper. Please add more discussion on likely reasons of the better agreement in some but not in all cases.

Response: Additional discussion has been added to the end of this section:

"As discussed in sect. 3.3, Eq. 4 for the IVC was fitted to the product distributions of C10, C11, and C12 using unified coefficients a and b to account for the vapor pressure drop in products that results from the increase of the carbon number of the precursor. The vapor pressure drop is not linear as the addition of a carbon to a longer carbon chain causes a smaller decrease in volatility (i.e. the reduction of volatility from adding a carbon to C11 will be smaller compared to the reduction in volatility from adding a carbon to C10). Eq. (4) is non-linear but may not be the ideal equation to model the vapor pressure drop within the product array. Thus, as a consequence of the fitting process, the model performance of the C10 array may have been sacrificed to improve model performance in the case C11, and C12. Furthermore, the current explicit gas mechanism cannot include every possible product, which can be associated with the oxidation

on all carbons in each alkane chain. The products of this study are obtained from oxidation at a given carbon position, which is representative of the whole set of gas oxidation. This can cause some deviation of prediction from the true atmospheric oxidation process. Thus, the product distribution originating from these explicit mechanisms can also cause variation in prediction of SOA mass."

l. 369 – 371: Please be more quantitative here. How much higher are the SOA loadings in the chamber as compared to typical environmental conditions? With your model, you should be able to do a sensitivity study to explore how this might affect the OM(p) and OM(AR) ratio.

Response: The sensitivity test for the OM_p to OM_{ar} was performed at atmospherically relevant SOA loadings (OM_0 = 5 μg/m^3). The note that SOA loadings found in the chamber were much greater than those observed in the ambient environment was meant as a caveat that the validation of model performance was under relatively high SOA loadings. It is clear that our sentence that read : "Lower SOA loadings may change the relative distribution of OM_P and OM_{AR}." suggested that the sensitivity test itself was performed under conditions that were not atmospherically relevant and has thus been removed.

These sentences have been updated and now read:

"Fig. 7 also includes the contribution of OMP and OMAR to SOA in the absence of inorganic seed at atmospherically relevant conditions (OM0 = 5 μg/m3). Overall, the relative contribution of OMAR is higher with lower temperatures and higher carbon numbers. Notably, the levels of SOA loading found in the chamber experiments which were used to validate model performance are much greater than those observed in the ambient environment (up to 32 times higher than the annual PM air quality standard set by the EPA of 12 μg/m3 (Epa, 2012))."

Section 4.5:

1) The header of this section is very general, compared to the content. 'Model parameters' may also include temperature, vapor pressures, product distributions etc. However, in fact, you only varied two rate constants. Please clarify this in the section header.

Response: The heading of this section was changed to "Uncertainty of model rate constants."

2) What is the reasoning to change the rate constant by +/- 50%. This seems a very narrow range. Please add some references that show that such rate constants indeed only vary within this range.

Response: Previous studies have reported that calculated rate constants for autoxidation reactions have uncertainties of about a factor of 5. This uncertainty test has been updated such that this rate is increased/decreased by a factor of 5 and this information has been updated in section 4.5.

l. 381: Please add 'of SOA' in this sentence. ('source of SOA')

Response: This correction has been made.

l. 403-405: Is this a result form your study or by Gentner et al?

Response: This is a result from our study. This result is compared to the result from Gentner et al.'s study later on: "Notably, Gentner et al. (2012) found that the peak of SOA mass production from linear alkanes in diesel appears between C19 and C22 which is a slightly higher carbon number compared to our prediction."

Please add a section 'Summary and Conclusions'.

Response: The atmospheric implication section has been divided into two sections : "4.6: Application of IVC-base product distributions to SOA simulation from diesel-linear alkanes" and "4.7 Summary and Conclusions."

Technical:

l. 13: replace 'formation' by 'prediction' (hence you can omit ''better predict' at the end of the sentence).

Response: The sentence has been updated and now reads: "Autoxidation paths integrated with alkyl peroxy radicals were added to the Master Chemical Mechanismv3.3.1 to improve the prediction of low volatility products in the gas phase and SOA mass"

l. 14: It is not clear what 'lumping groups' are. Do you mean 'lumped into volatility-reactivity based groups'?

Response: Yes. This sentence has been updated and now reads: "The resulting gas products were then lumped into volatility-reactivity based groups that are linked to mass-based stoichiometric coefficients."

l. 47: replace 'have often' by 'has often'

Response: "have often" has been replaced by "has often"

l. 49/50: the addition of 'using the CHIMERE regional air quality model' seems at the wrong place. It should be inserted either at the very beginning of the sentence or after 'of SOA'.

Response: This sentence has been updated and now reads: "Using the CHIMERE regional air quality model, Hodzic et al. (2010) found that the inclusion of IVOCs led to a substantial improvement in predictions of SOA compared to data collected from the MILAGRO field experiment"

l. 53: replace 'predicted' by 'reproduced'

Response: "predicted" has been replaced with "reproduced"

l. 60: which 'current explicit mechanism' do you mean here? Are PRAM not included in any available mechanism or specific in MCM?

Response: This is referencing the MCM mechanism. "current explicit mechanism" has been changed to "current MCM mechanism".

l. 72: add 's' to precursor

Response: This correction has been made.

l. 73: add 'for' (account for...)

Response: This correction has been made.

l. 124: replace 'mechanisms' by 'mechanism'

Response: This correction has been made.

l. 175: MCM stands for 'Master Chemical Mechanism'; thus, MCM mechanisms seems redundant.

Response: "Mechanisms" has been removed from this phrase.

l. 177: replace 'increase' by 'increases'

Response: This correction has been made

l. 215: replace 'or' and 'in' by 'org' and 'inorg'. Please check carefully the full manuscript for other instances (eg.. Eq-6, 7, 10, 11 , l. 227, l. 233 etc).

Response: These corrections have been made.

l. 221: Is there anything missing or should the colon at the end of the sentence be replaced by a period?

Response: This colon has been replaced by a period.

l. 344: replace 'lowly volatile' by 'low volatility'

Response: This correction has been made.