Response to reviewer's comments:

Title: Secondary organic aerosol formation from smoldering and flaming combustion of biomass: a box model parametrization based on volatility basis set

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Dear Editor,

We thank the reviewers for their comments. Our detailed point-by-point responses to the reviewers' comments (in black regular typeset) are provided in blue regular typeset and the revised text (highlighted in the main text) is in *grey italic typeset*.

Reviewer 1:

Summary:

The authors burned wood in residential stoves under two burning conditions (flaming, smoldering) and with three different types of stoves. Emissions were sampled into a smog chamber and aged via OH-radical initiated oxidation. Several aging temperatures and VOC concentrations were explored. Resulting particles were measured with AMS and gas phase species with PTR-ToF. Gas-phase precursor species were sorted by chemical structure. SOA yields were measured. A box model was used to determine the volatility distribution of oxidation products. This was accomplished by iteratively adjusting the estimated concentrations of product species in various volatility bins, and the average enthalpy of vaporization, so that measured SOA concentrations best matched modeled concentrations. The relative contributions of different precursor VOC classes to SOA yield were determined using a genetic algorithm. The major results of the study are (1) smoldering and flaming conditions emit different primary VOCs, (2) cresols and phenols are important SOA precursors, (3) SOA yields increase at lower temperatures. However, the identification of which chemical classes of VOC are major SOA precursors was not consistent between different sets of experiments.

Overall I think that this work takes a promising approach to a difficult and important problem (SOA formation from biomass burning). The data set is interesting and the experimental techniques appropriate. However, there are some issues with disorganization in describing the analytical approach, the model implementation, and comparison of the effects of different variables (e.g. chemical composition, temperature, OH exposure) that make it difficult to follow the major conclusions and find information that could be useful for air quality modeling parameterization.

Major comments:

The analysis in this paper rests on the idea that there should be systematic differences in SOA yield between the six selected classes of SOA precursors. These classes are defined by structure: furans, PAH, unoxygenated aromatics, oxygenated aromatics, and two catch-all categories of other compounds with more and fewer than 6 carbon atoms. However, it is never clearly established why these six classes were chosen and so defined. Why not sort compounds by functionality (e.g. ketones, acids, diols, etc), O:C ratio, molecular weight, flaming vs. smoldering source, or some other characteristic? Why not handle each species separately? To summarize, the authors need to establish (1) why the lumping was

necessary, and (2) why this particular differentiation of chemical characteristics should be expected to explain variability in SOA formation. Secondly, it appears that the individual species included in each chemical class were not the same for each experiment. This is a significant weakness of the analysis. The authors should strongly consider a more consistent and chemical meaningful approach to grouping compounds.

The reviewer has raised two concerns in this comment. We have broken the comment into two specific points, addressing each in turn.

Point 1: the rationale behind the lumping approach:

As it can be seen from the relatively high uncertainties in the yields (up to factor of three, Figure 9), the size of our dataset does not allow us retrieving the volatility distribution for the single precursors, which would entail the determination of more than 80 free parameters. This is especially the case as the precursor time series, decaying with oxidation, are typically strongly correlated, which prevents resolving systematic differences between the yields of the different single precursors. Indeed, the current state-of-the-art VBS applications lump all SOA precursors in one single group (e.g. Ciarelli et al. 2017). Therefore, lumping is needed to decrease the model degree of freedom and currently, the model is solved for eight free parameters. The lumping approach is based on the two objectives of the study:

➔ Compare the SOA yields of specific precursors determined in complex emission experiments with those determined in single compound systems.

➔ Identify the main SOA precursors in flaming and smoldering wood emissions, at different temperatures.

To be able to compare to literature yields, we have lumped species that have similar yields in the same chemical class: e.g. at organic aerosol concentration of 10 µg m^{-3} the yields of PAHs is ~20% (first objective). In addition, we have classified the precursors based on the pathway by which they are emitted, which will allow us to determine which compounds dominate SOA formation in flaming and smoldering emissions (second objective). We differentiated between oxygenated aromatics, mainly emitted by lignin pyrolysis, furans emitted by cellulose pyrolysis and single-ring aromatics and PAHs generated from incomplete combustion, especially from flaming wood. The remaining SOA precursors are all oxygenated VOCs therefore we separated them according to their carbon number knowing that larger precursors will have higher yields than smaller precursors. Our ability to precisely extract yields specific to a precursor class heavily relies on differences in the oxidation rates or emission patterns of the precursors. Therefore, this classification approach adopted here, where classes have different contributions during different experiments, facilitates the extraction of yields of the different classes. Based on the reviewer comment we have added these clarifications in Section 3.1.4. (page 9), which now reads as follows:

"A common set of 263 ions were extracted from the PTR-ToF-MS. Among these ions, 86 showed a clear decay with time and were thus identified and selected as potential SOA precursors. Previous work based on Set2 experiments showed that the PTR-ToF-MS measures the most important SOA precursors, which explained the measured SOA mass within 40% uncertainty and without systematic bias (Bruns et al., 2016). Therefore, these compounds are expected to capture the dominant fraction of SOA mass, although we cannot rule out losses in the PTR-ToF-MS inlet or small contributions from other precursors such as alkanes. The compound identification was supported by previous publications (McDonald et al., 2000; Fine et al., 2001; Nolte et al., 2001; Schauer et al., 2001; Stockwell et al., 2015), including gas chromatography-mass spectrometry (GC-MS) analysis when available.

The size of our dataset does not allow us retrieving the volatility distribution for single precursors, which would entail the determination of more than 86 free parameters. This is especially the case as the time series of precursors, decaying with oxidation, are typically strongly correlated, which prevents resolving systematic differences between the yields of the different single precursors. Therefore,

lumping is needed to decrease the model degree of freedom. Accordingly, precursors are grouped in six chemical classes: furans, single-ring aromatic hydrocarbons (SAH), polycyclic aromatic hydrocarbons (PAH), oxygenated aromatics (OxyAH) and organic compounds containing more or less than 6 carbon atoms (OVOCc ≥6, OVOCc<6, respectively) (Table S1). This lumping approach is based on the two main objectives of the study:

1.   Compare the SOA yields of specific precursors determined in complex emission experiments with those determined in single compound systems.

2.   Identify the main SOA precursors in flaming and smoldering wood emissions, at different temperatures.

To be able to compare to literature yields, we lumped species that have similar yields in the same chemical class: e.g. at organic aerosol concentration of 10 µg m^{-3} the yields of PAHs is ~20% (objective 1). In addition, we classified the precursors based on the pathway by which they are emitted, which will allow us to determine which compounds dominate SOA formation in flaming and smoldering emissions (objective 2). We differentiated between oxygenated aromatics, mainly emitted through lignin pyrolysis, furans emitted through cellulose pyrolysis and single-ring aromatics and PAHs generated from incomplete combustion, especially from flaming wood. The remaining SOA precursors are all oxygenated VOCs therefore we separated them according to their carbon number knowing that larger precursors will have higher yields than smaller precursors. Our ability to precisely extract yields specific to a precursor class heavily relies on differences in the oxidation rates or emission patterns of the precursors. Therefore, the classification approach adopted here, where classes are expected to have different contributions during different experiments (Bhattu et al., 2019), facilitates the extraction of yields of the different classes."

Point 2: Selected precursors in Set1 and Set2:

The individual species included in each chemical class are the same for each experiment and for both the datasets. We are sorry for the misunderstanding and we add the following statement in the end of section 3.1.4.:

Page 9 line 16: "The selected precursors in each class are the same for each experiment and each dataset."

An additional point of concern is that multiple variables changed at same time: e.g. set 1 and set 2 experiments have different flaming/smoldering condition, different smog chamber temperatures, different OH exposure, and different VOC concentration. Is there a way to deconvolve these effects? The SOA yield of each experiment should best be compared at the same OH exposure.

Extracting class-specific yields essentially resides on the variability in the contribution of different classes to condensable species, between experiments (due to variability in the emissions) or within an experiment (due to differences in the precursors reaction rates). By nature, emissions are very complex and therefore hard to control and typically several parameters co-vary. It is important in fitting for the class specific yields in such a multivariate problem that the contribution of the different precursors to the condensable gases do not co-vary with other parameters (e.g. temperature, OH exposure), otherwise their respective effects cannot be distinguished and biases in the yields may arise. The main variability in the contribution of different classes to condensable species is due to variability in the emissions between Set1 and Set2. Most of the other parameters in Set1 and Set2 are consistent or have a significant overlap, allowing the retrieval of class specific yields and a direct comparison between the experiments. In the following, we compare Set1 and Set2 with respect to the different combustion and aging parameters:

➔ **Combustion conditions:** The combustion conditions between the two sets are comparable, in terms of the starting procedure and the type and the amount of wood burned. The main difference is related to the air input, which favored smoldering and flaming conditions for Set1 and Set2, respectively. This is directly reflected in the composition of the SOA precursors and such variability is key to deconvolve the class-specific yields. Despite this, the emission composition for Set2 is variable, with some experiments showing a similar composition as Set1 (see Fig. 2d), ensuring an overlap between Set1 and Set2 chemical composition when other parameters varied.

➔ **OH exposure:** The OH exposure is ~7 times higher for Set2 compared to Set1. This is because the OH production rate was similar between the two experiments, but its sink was significantly higher for Set1 due to the injection of higher emissions. However, we note that the OH exposure is not directly considered in the model. Instead, the integrated production of condensable species, inferred from the precursor decay, is used as inputs, which takes into account the differences in the OH exposures. Besides, we note that most of the SOA production occurs at OH exposures $< 8 \cdot 10^6$ molecules cm^{-3} h, which is reached in both experiments.

➔ **Emission loading:** On average, higher concentrations were injected during Set1 compared to Set2. This was not intentional, but occurred as smoldering emissions rates were much higher and therefore it was more challenging to control the amounts injected. That said it is important that the concentrations span a wide range (here 1.5 orders of magnitude) to be able to constrain the amounts of oxidation products in the different volatility bins. As mentioned in the manuscript, the volatility distributions are well constrained in the saturation concentrations between 10-1000 µg m^{-3}, corresponding to the range of OA concentrations studied here. We finally note that the concentration ranges in Set1 and Set2 have a significant overlap (for Set1, SOA = 50-800 µg m^{-3} vs. 20-140 µg m^{-3}), ensuring that the fitting is not biased due to difference in the loadings between the two sets.

➔ **Aging temperature:** conducting the experiments at different temperatures is essential for constraining the condensable species enthalpy of evaporation. Set1 experiments were conducted at 2°C, right in the middle of the two temperatures examined in Set2: -10°C and 15°C. Therefore, the temperature does not co-vary with the changes in the emission composition between Set1 and Set2 and the retrieval of the yields is not affected by the temperature.

Finally, we have assessed the sensitivity of the yield retrieval on the experiments included in the parameterization, through the bootstrap analysis. This influences the covariance between parameters and therefore this sensitivity analysis is a way to assess the biases resulting from such covariance. We present the resulting ranges of yield parameters from the sensitivity analysis.

Two papers using this dataset have been published previously (Bruns et al. 2016, 2017). In the introduction, the authors should more directly state the relationship of this work to the two previous papers and what new analysis is added.

Based on the reviewer comment, we have modified the introduction to clarify the links between the current study and Bruns et al. 2016 and 2017. The modified text reads as follows:

"Bruns et al. (2016) investigated the SOA formation from residential log wood combustion from a single type of stove under stable flaming conditions only. They reported that T-SOA precursors included in models account for only 3 to 27% of the measured SOA whereas 84 to 116% was from NT-SOA precursors including in total 22 individual compounds and two lumped compound classes, mainly consisting of polycyclic aromatic hydrocarbons from incomplete combustion (e.g. naphthalene) and cellulose and lignin pyrolysis products (e.g. furans and phenols, respectively). The estimated SOA

concentrations were based on the literature SOA yields of single precursors, obtained from smog chamber experiments, and a good agreement was observed between predicted and measured SOA. However, the method suffers from two drawbacks. First, the dependence of the yields on the organic aerosol loading and temperature was not considered. Second, although the relative contributions of different precursors to SOA were estimated, thermodynamic parameters for chemical transport models (CTMs) were not determined. Based on the same experiments, the lumped concentrations of the 22 non-traditional volatile organic compounds and 2 compound classes were constrained in a box model (Ciarelli et al., 2017a). Improved parameters were retrieved describing the volatility distributions and the production rates of oxidation products from the overall mixture of precursors present in biomass smoke. While this method is well suited for CTMs (Pandis et al., 2013; Ciarelli et al., 2017b), it does not provide any information about the contributions of the different chemical classes to the aerosol."

We have also explicitly defined the two sets of experiments in the method section:

"Experiments from Bertrand et al., 2017, 2018a will be referred to as Set1 and experiments from Bruns et al., 2016 and Ciarelli et al., 2017 as Set2."

The abstract could use some organizational editing, to clarify the major aims of the study, the analytical methods used to interpret the data, and the major results.

Based on the reviewer comment, we have reorganized the abstract and highlighted more clearly the major aims of the study. The modified abstract reads as follows:

"Residential wood combustion remains one of the most important sources of primary organic aerosols (POA) and secondary organic aerosol (SOA) precursors during winter. The overwhelming majority of these precursors have not been traditionally considered in regional models, and only recently, lignin pyrolysis products and polycyclic aromatics were identified as the principal SOA precursors from flaming wood combustion. The SOA yields of these components in the complex matrix of biomass smoke remain unknown and may not be inferred from smog chamber data based on single compound systems. Here, we studied the aging of emissions from flaming and smoldering-dominated wood fires in three different residential stoves, across a wide range of aging temperatures (-10°C, 2°C and 15°C) and emission loads. Volatile organic compounds (VOCs) acting as SOA precursors were monitored by a proton transfer reaction time-of-flight mass spectrometer (PTR-TOF-MS), while the evolution of the aerosol properties during aging in the smog chamber was monitored by a high resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). We developed a novel box model based on the volatility basis set (VBS) to determine the volatility distributions of the oxidation products from different precursor classes found in the emissions, grouped according to their emission pathways and SOA production rates. We show for the first time that SOA yields in complex emissions are consistent with those reported in literature from single compound systems. We identify the main SOA precursors in both flaming and smoldering wood combustion emissions at different temperatures. While single-ring and polycyclic aromatics are significant precursors in flaming emissions, furans generated from cellulose pyrolysis appear to be important for SOA production in the case of smoldering fires. This is especially the case at high loads and low temperatures, given the higher volatility of furan oxidation products predicted by the model. We show that the oxidation products of oxygenated aromatics from lignin pyrolysis are expected to dominate SOA formation, independent of the combustion or aging conditions, and therefore can be used as promising markers to trace aging of biomass smoke in the field. The model framework developed herein may be generalizable for other complex emissions sources, allowing determination of the contributions of different precursor classes to SOA, at a level of complexity suitable for implementation in regional air quality models."

Specific comments:

It is not clearly described how different "flaming" and "smoldering" conditions were achieved in the two sets of experiments, and how the relative levels of flaming and smoldering were determined. The effect of temperature is not noted in the abstract.

Ward and Hardy (1991) define the flaming and smoldering conditions according to the modified combustion efficiency, $MCE = CO_2/(CO+CO_2)$. Specifically, $MCE > 0.9$ is identified as flaming condition, while $MCE < 0.85$ is identified as smoldering condition. We have already reported these values in Table 1. Practically, we achieved the different burning conditions by varying the amount of air in the stoves, therefore changing the combustion temperature. For Set1, closing the air window decreased the flame temperature, resulting in a transition from a flaming to a smoldering fire. This could be visibly identified, together with the development of a thick white smoke from the chimney. We note that this conduct is very common in residential stoves, to keep the fire running for longer. Meanwhile, for set2, after lighting the fire, we kept a high air input to maintain a flaming fire. At the same time, we monitored the MCE in real time and only injected the emissions into the chamber when the MCE increased above 0.9. This information is now added to the manuscript, in Section 2.1:

"Two smog chamber campaigns were conducted to investigate SOA production from multiple domestic wood combustion appliances as a function of combustion phase, initial fuel load, and OH exposure. These experiments were previously described in detail (Bruns et al., 2016; Ciarelli et al., 2017; Bertrand et al., 2017, 2018a) and are summarized here. Experiments from Bertrand et al. (2017, 2018a) will be referred to as Set1 and experiments from Bruns et al. (2016) and Ciarelli et al. (2017) as Set2. The emissions were generated by three different logwood stoves for residential wood combustion: stove 1 manufactured before 2002 (Cheminées Gaudin Ecochauff 625), stove 2 fabricated in 2010 (Invicta Remilly) and stove 3 (Avant, 2009, Attika). For each stove three to four replicate experiments were performed with a loading of 2-3 kg of beech wood having a total moisture content ranging between 2 and 19%. The fire was ignited with 3 starters made of wood wax, wood shavings, paraffin and 5 natural resin. The starting phase was not studied. In total, 14 experiments were performed, consisting of two experiments at -10°C, seven experiments at 2°C and five experiments at 15°C. These experiments cover the typical range of European winter temperatures and are summarized in Table 1.

Ward and Hardy (1991) define the flaming and smoldering conditions according to the modified combustion efficiency, $MCE = CO_2/(CO+CO_2)$. Specifically, $MCE > 0.9$ is identified as flaming condition, while $MCE < 0.85$ is identified as smoldering condition. MCE values for the different experiments are reported in Table 1. According to this parameter, Set1 and Set2 experiments were dominated by smoldering and flaming, respectively. Practically, we achieved the different burning conditions by varying the amount of air in the stoves, therefore changing the combustion temperature. For Set1, closing the air window decreased the flame temperature, resulting in a transition from a flaming to a smoldering fire. This could be visibly identified, together with the development of a thick white smoke from the chimney. We note that this conduct is very common in residential stoves, to keep the fire running for longer. Meanwhile, for Set2, after lighting the fire, we kept a high air input to maintain a flaming fire. At the same time, we monitored the MCE in real time and only injected the emissions into the chamber when the MCE increased above 0.9."

Abstract: From the abstract it appears that one of the major conclusions is that flaming and smoldering conditions produce a different mix of VOCs, but this is not discussed in the paper.

Figure 2, 3 and 4 aim to describe the different mix of primary VOCs for the two datasets, their chemical differences among the two sets of experiments, their contribution to the total primary emissions and the amount decayed upon oxidation.

Page 4 Line 1: These values don't quite make sense to me; how is it possible that the contributions consistently add to more than 100%? Is this mass yield or carbon yield?
The percentage values (84-116%) reported in the text are not SOA yields, but refer to what extent the measured SOA could be explained by the measured precursors based on their SOA mass yields reported in literature. For more information we refer to Bruns et al. (2016).

Page 4 Line 9: Which parameters?
The model parameters refer to oxidation product production rates and volatility distribution. For clarity, we modified the text as follows:
"Based on the same experiments, the lumped concentrations of the 22 non-traditional volatile organic compounds and 2 lumped compound classes were constrained in a box model (Ciarelli et al., 2017a). Improved parameters were retrieved describing the volatility distributions and the production rates of the oxidation products from the overall mixture of precursors present in biomass smoke."

Page 5 line 11: If relative humidity was constant, than actual water vapor mixing ratios were quite different between the three temperature conditions. Is this expected to have an effect? Why was this particular humidity condition chosen?
For the particle phase, the particle water content, which affects the partitioning of semi-volatile products, is a function of RH and not of absolute humidity. Thus, we kept RH constant in the experiments in order to compare the different systems. Regarding the gas phase chemistry, water concentration would play a role if the main source of OH radicals were ozone. However, in our case, OH radical is produced from HONO photo dissociation. Therefore we do not expect absolute humidity to play a significant role under our conditions. All experiments were conducted at an RH of ~ 50%, typical of daytime values encountered in Europe during winter.

Page 5: What NOx levels and NOx:VOC ratios were present?
NOx concentrations at the beginning of the experiments are available just for Set1, ranging between 0.04 and 0.26 ppm before lights on. Therefore, the NOx/VOCs levels varied between 0.1 and 0.5 and are therefore representative of high NO_X conditions. These details were addressed in the previous publications.

Page 6 line 15: AMS collection efficiencies can be substantially less than 1, especially for very low volatility and highly oxidized particles. A collection efficiency correction could change the conclusions of this work. Can you support the assumption of CE=1?
The AMS measurements were thoroughly validated in the previous publications, reporting the results of the same experiments (Bruns et al., 2016; Bertrand et al., 2017; Kumar et al., 2018). Within the measurement uncertainties (20%), the sum of BC and non-refractory species is consistent with the

SMPS volume measurements when applying a CE of 1 and a density of 1.5, typical of biomass burning aerosols (Corbin et al., 2015).

Page 8 line 27: Was photolysis of VOCs considered?
Photolysis was not considered. We demonstrated that the decay of at least the major precursors is consistent with their loss by dilution and reaction with OH, indicating that other processes, including photolysis, are minor. For the other species, if photolysis occurs, it will have the same magnitude in both Set1 and Set2.

Page 9 Line 7: How were these 263 ions selected?
The 263 ions refer to all the species fitted using high-resolution analysis of the PTR data while the selected 86 compounds were selected as SOA precursors according to their clear decay upon oxidation in the smog chamber.

Eq. 4 and 5: Could the measured OA be simultaneously corrected for wall loss and dilution by dividing by the measured BC signal over time, normalized to 1 at t=0?
No, this is not possible. The loss of the particles onto the walls occurs through their deposition by diffusion and sedimentation and therefore affects only the particle phase. Accordingly, particle wall losses obey a first order process and therefore were corrected for using Eq. 5 and the wall loss rates determined from Eq. 4. In contrast, dilution does not only influence the physical removal of the particles, but also their partitioning between the gas and particle phase, which is dependent on the species volatility. Therefore, dilution cannot be simply corrected by normalizing to the BC concentrations. Instead, we have calculated the diluted concentrations of total condensable gases, which we use as input into the model to calculate the fractions that partition to the particle phase. The modelled particle phase after dilution is then compared with the measured wall loss corrected organic aerosol after dilution.

Page 11 Lines 30-Page 12 line 9: This needs to be better supported and more detailed. Each of the six main precursor classes was further subdivided into six volatility bins, and the average chemical properties were determined for each bin – is this correct? Was there a large range in #C, #H, #O within each bin? It is not entirely clear to me how the volatility bins were determined: was the volatility of each individual species determined, and species were then grouped into bins? Or were species first grouped using some other method, then the volatility of each group was determined?
Here, we believe that some clarifications are needed. The section refers to the precursor oxidation products, referred to as secondary surrogates, and not to the precursors themselves. The volatility of the precursor classes was not used in the model. The volatility basis set was used to distribute the oxidation products from the different precursor classes into logarithmically spaced volatility bins. These bins are preset; 6 bins were considered with a saturation concentration $C^* = \{10^{-1}; 10^0; 10^1; 10^2; 10^3; 10^4\}$ µg m^{-3} for the oxidation products of all precursor classes. This is described in the model inputs section 3.3.2 in lines 26-28. The box model using a genetic algorithm as a solver determines the fraction of the oxidation products in each of the bins, or what can be referred to as the volatility distribution of the oxidation products. The molecular composition of the secondary surrogates, which is used to calculate

their molecular weight and the bulk OA O:C ratios, is based on that of their precursors, as described on page 13 Lines 6 and 13.

Page 13 lines 1-7: Can you explain more clearly what mutation, crossover, and selection mean for the implementation of the genetic algorithm in this particular application? How does the algorithm actually identify the optimal set of parameters?

Based on the reviewer comment, we rephrased Section 3.3.4 (on Page 13 Lines 1-7) as follows:

"The model is optimized to determine the volatility distributions of the oxidation products from different precursor classes described by μ_j, σ and their temperature-dependence described by ΔH_{vap}, to best fit the observed OA concentrations. For the model optimization, we used a genetic algorithm (GA), a metaheuristic procedure inspired by the theory of natural selection in biology, including selection, crossover and mutation processes, to efficiently generate high-quality solutions to optimize problems (Goldberg et al., 2007; Mitchell, 1996). The GA is initiated with a population of randomly selected individual solutions. The performance of each of these solutions is evaluated by a fitness function, and the fitness values are used to select more optimized solutions, referred to as parents. The new generation of solutions (denoted children) are produced either by randomly changing a single parent (as mutation) or by combing the vector entries of a pair of parents (as crossover). The evolution process will be repeated until the termination criterion is reached, here maximum iteration time. In this study, a population of 50 different sets of model parameters (μ_j, σ and ΔH_{vap}) was considered for each GA generation. The sum of mean bias and RMSE between measured and modelled C_{OA} of the 14 experiments were used as fitness function to evaluate the solutions. We assume the termination criterion is reached if no improvement in the fitness occurs after 50 generations, with a maximum of 500 total iterations allowed. The GA calculations were performed using the package "GA" for R (Scrucca et al., 2017). A bootstrap method was then adopted to quantify the uncertainty in the constrained parameters."

Page 14 lines 21-23: It should be stated more clearly before this point that the individual species contributing to each of the 6 chemical classes were not consistent between different experiments. The inconsistency in chemical composition makes it extremely difficult to compare between different experiments or to draw conclusions about the SOA yield from different chemical classes.

As mentioned above, the individual species included in each chemical class are the same for each experiment and for both datasets.

Page 15 lines 4-6: Two groups of compounds could have substantially different distribution of OH reactivities and SOA formation potential, but similar average OH reactivity. The conclusion here is not well supported.

The aim of this analysis is to investigate whether the faster decay observed against the OH exposure in Set1 compared to Set2 is due to the presence of more reactive species in Set1 for the different chemical classes. Therefore, in Table 2, we calculated average OH reaction rate constants for each single experiment and for the different chemical classes. These were weighted by the abundance of the different compounds in their respective chemical class. We show that the reaction rates compare well between the two sets of experiments, which does not explain why some chemical classes decay faster in set1 compared to set2.

Technical corrections:

Abstract 8-10: Sentence is difficult to understand, please rephrase.
Abstract was adjusted and is reported above.

Page 5 line 49: should be sodium nitrite, NaNO2
Yes, we apologize for this mistake. This was corrected in the revised manuscript.

Eq. 1 Brackets for the sum term are missing.
This was adjusted in the modified text.

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