

Interactive comment on “In-cloud processes of methacrolein under simulated conditions – Part 2: Formation of Secondary Organic Aerosol” by I. El Haddad et al.

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The authors thank referee #1 for raising a number of important comments and interesting questions that will be considered in the corrected manuscript. Some comments/questions, however, exceed the strict framework of this study whose main objective was to demonstrate the feasibility of a process and to evaluate its potential atmospheric implications. We recognize that this work raises questions and requires further investigations to better understand the processes involved. This is a first exploratory step, which opens perspectives for future works. In the conclusions, a section will be dedicated to summarize the outstanding issues and to guide future works.

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Comment 1: As mentioned by referee #1, previous studies have investigated the contribution of cloud processes in the formation of low volatile products through laboratory experiments, atmospheric measurements or modelling. On the one hand, atmospheric measurements (Sorooshian et al., 2006 and 2007) have shown that cloud processes can act as aerosol source, but they could not demonstrate the implication of cloud photochemistry in this process. On the other hand, laboratory studies have proven the formation of high molecular weight products through OH-oxidation reactions (Altieri et al., 2006, 2008 and Carlton et al., 2006, 2007) or self-oligomerization upon water evaporation (Loeffler et al., 2006). However, these studies did not confirm the implication of these products in SOA formation, upon water evaporation. This paper provides strong experimental evidence that cloud processes can act through photo-oxidation reactions as important contributors to SOA formation. For this reason, the statement has been modified to: “This provides, for the first time to our knowledge, strong experimental evidence that cloud processes can act, **through photo-oxidation reactions**, as important contributors to SOA formation in the troposphere.”

Comment 2 and 7: Referee #1 suggests to examine the fate of methacrolein oxidation products through the mix of standards: 1) in the presence of H_2O_2 and separately in the presence of UV to determine whether any quantified products are affected by these during experiments intended to measure reactions with OH and 2) upon water evaporation (i.e. difference between the aerosol mass generated by the standard mix and high molecular weight products in the reaction vessel). These are interesting suggestions that will be taken into consideration when investigating thoroughly the fate of methacrolein oxidation products and their implication in the observed aerosol formation. However, in this study, the main objective was to determine if the tropospheric aqueous phase photochemistry of methacrolein was able to generate low volatile products and consequently SOA. These suggestions will be considered more accurately in future studies, and will be mentioned in the corrected manuscript.

Comment 3: Referee #1 questions the relevance of this process in the atmosphere

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considering the concentrations of methacrolein and OH used in this study. The concentrations used here are significantly higher than the one found in cloud water (Van Pinxteren et al., 2005) but are of the same order of magnitude as the ones used by Altieri et al., 2006 and 2008 during laboratory experiments, representative of aerosol water.

Comment 4: Occurrence of additional formation of high molecular weight products induced by the nebulisation/evaporation process is also an interesting question. Unfortunately, no simple answer can be given for now and especially without fine chemical analysis of the aerosol formed. In this study, the nebulisation of methacrolein did not reveal any SOA formation, which indicates that no high molecular weight products can be formed from methacrolein alone through the nebulisation/evaporation processes. In contrast, the formation of additional high molecular weight products by the nebulisation/evaporation processes through the accretion of methacrolein oxidation products, upon water evaporation (i.e. methylglyoxal, Loeffler et al., 2006), cannot be excluded here. This question will be considered in the revised version of the manuscript.

Comment 5: The authors agree with the referee #1 on the fact that the size distributions depend on how the atomizer is operated and that only the change in the size distribution, not the absolute size distribution is meaningful. For this reason in the manuscript an increase factor of 3.7 in the particle diameter was mentioned instead of absolute diameters of the obtained SOA: "Similarly, a significant increase of the aerosol diameter and number was observed: from 0 to 22 h, the aerosol number and diameter have increased by a factor of 254 and 3.7, respectively (Table 2)". The fact that the size distributions depend on how the atomizer is operated has also been mentioned more clearly in the corrected manuscript.

Comment 6: This point is particularly important and re-concentration process in the vessel during the nebulisation has been taken into considerations in our results. During the nebulisation of an aqueous solution from the reaction vessel, an increase of the aerosol mass by approximately a factor of 2 within 5 hours was observed. For the

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calculation of the size distributions and the mass of SOA obtained at each reaction time, an average was considered after 1 h of nebulisation and during approximately 2 h (increase of the SOA mass by a factor of 1.4). However this increase is significantly lower than the one observed during the reaction progress, as shown by the standard deviations on each experimental point in Figure 6.

Comment 7: Without chemical analyses of the generated aerosol, it is impossible to evaluate the fraction of the aerosol coming from high MW compounds. Again this is an interesting question, but the answer can be given only through further investigations.

Comment 8: The authors are aware that the yields obtained can depend on several experimental parameters such as the reactants (methacrolein and OH radical) concentrations in the reaction vessel. In addition, in this study the yield depends on the setup used to produce the SOA (nebulisation/evaporation). In the corrected manuscript, the remark of referee #1 will be taken into consideration and cautions that have to be considered when using these yields will be mentioned more clearly in the corrected manuscript.

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