

First, we would like to thank our two referees, and Dr Golam Sarwar and Dr Luc Vereecken for their critical but fruitful comments to improve the quality of our manuscript 'Oxidation of SO₂ by stabilized Criegee Intermediate (sCI) radicals as a crucial source for atmospheric sulphuric acid concentrations'.

The aim of our manuscript was to investigate the very recent improvements of a few sulphuric acid producing reactions to that which is stated in the Master Chemical Mechanism (MCM), which is used widely in different models. We are observing the formation of sulphuric acid using the MCM and using an alteration of MCM by these new reaction rate coefficients. Our aim is to investigate what is the predicted sulphuric acid concentration in field conditions when using what researches normally use (so MCM) or when using MCM with the improved reaction rate coefficients. Now, as in the paper, we do also acknowledge that there are very large uncertainties connected to the Criegee Intermediate chemistry. The aim of our paper is not to investigate, or test, all possible uncertainties because this would be out of the scope for one manuscript. However, we will extend the written paragraph on the uncertainties according to the discussion by both the reviewers and the open comments.

In the following we will first answer the comments from the two referees and then the comments from Dr Sarwar and Dr Vereecken.

Anonymous Reviewer Nr. 1

In this paper the authors present direct impact of new kinetic measurements of the reaction of C.I.s with SO₂. The reaction rate constants are employed in a simple model to quantify the impact of faster CI rates of reaction on the H₂SO₄ budgets. However, the estimation on the importance of the CI-SO₂ reaction and their modelling work may be inappropriately extrapolated.

*1. The paper assumes that SO₃ is the product of the reaction of C.I. + SO₂. What actual experimental evidence is there for this hypothesis? In essence their work is an extrapolation of the indirect methodology of Cox and Penkett (Nature **230**, 321-322, 1971). It has been pointed out by the interactive comment there is very little detail given of the model set up and much more information is needed.*

In chapters 3.1 and 3.2 we provide short descriptions of the two models (MALTE and SOSA) used in this study with references linking to detailed descriptions of the two models. In chapter 3.3 - the most important part of this work - the chemistry and the assumed scenarios are presented. As mentioned in the text we have used the chemistry based on the Master Chemical Mechanism of Leeds University (Version 3.2) throughout our studies with the exceptions of different reaction rates for the reactions of sulphur dioxide with certain selected Criegee Intermediates. All changes concerning the reaction rates assumed for our studies are provided in detail in Table 1. It would be possible to show as an appendix the whole chemistry code, however because this is in different formats available at the MCM-website the authors assumed that it will not be necessary.

If the authors have assumed a 100% SO₃ formation this needs to be stated and more importantly the experimental evidence (or otherwise) that they use to support their hypothesis.

We acknowledge that there is an ongoing discussion on the production yield of SO₃ from CI + SO₂. We will include this in the paper together with references to Vereecken et al. 2012 and Carlsson et al. 2012. As written above, as a general responds to all the comments to this ACPD version, our aim is not to test all the uncertainties connected to the CI chemistry. We use MCM and MCM assumes a 100 % yield.

2. The authors suggest that the **direct** kinetic measurements of Welz et al. are too fast. They suggest that their modelling results and observations imply a much slower rate coefficient for the reaction of Cl with SO₂. However, a rate constant of 10-11 cm³ molecule⁻¹ s⁻¹ is supported by both experimental and theoretical work. Welz et al., reported a direct measurement for the reaction of CH₂OO with SO₂. The theoretical results of Kurtén et al. (*J. Phys. Chem. A*, **2011**, 115 (31), 8669–8681) show that the SO₂ reaction with acetone oxide, which has no alpha-hydrogen, has a barrier less entrance channel and a low-lying transition state for SO₃ + carbonyl formation, similar to that for CH₂OO + SO₂. Jiang et al. (*J. Phys. Chem. A*, **2010**, 114, 12452–12461) theoretically characterized CH₂OO + SO₂ and the Criegee + SO₂ reactions for the Cl's from limonene ozonolysis. In a similar vein to Kurtén et al, they do not report barriers on the entrance channel for either reaction. In light of these findings, it would suggest that the rate coefficient with Cl with SO₂ would be faster than the 10-13 cm³ molecule⁻¹ s⁻¹ implied in the manuscript. Indeed, in more recent work Vereecken et al., (*PCCP*, 14, 14682-14695, 2012) have also reported a theoretical rate coefficient of the order of 10-11 which has been further supported by recent experimental work (Carlsson et al., *PCCP*, 14, 15637-15640, 2012). A much more balanced view of the reported rate coefficients needs to be reported, indeed, specifically it should be addressed why their data is different from direct measurements, indirect measurements and theoretical work.

We are not implying that the measurements by Welz et al. are incorrect. We are stating that including the measured reaction rate coefficient by Welz et al. in the MCM, we are not able to reproduce the measured H₂SO₄ concentration in the field. This can be of course in relation with an underestimation of the reaction rates of the sCl with H₂O which will be discussed below. In general there is currently a quite enthusiastic discussion in the scientific community about how Criegee Intermediates can react with different compounds. In our study we used the results from Mauldin et al. and Welz et al. to investigate to which extent the new published rate coefficients from their papers could explain the missing observed sulphuric acid concentrations under high and low concentrations of monoterpenes in the field. The paper has not the aim to present a sensitivity study on all available reactions rates with sCl published until now.

3. The impact of the decomposition rate of the Cl will have a large impact on the order of magnitude that can be used to fit the field data. Again little information is given, for instance if a larger decomposition rate was used the rate coefficient for Cl with SO₂ would have to be increased for the observational data. Indeed it has been shown that the decomposition rate can vary from 0.3 – 250 s⁻¹ (*J. D. Fenske, A. S. Hasson, A. W. Ho, S. E. Paulson, J. Phys. Chem. A* 104, 9921, 2000). Given that the decomposition rate can vary by orders of magnitude this would imply that the rate coefficient for Cl + SO₂ could be varied by orders of magnitude to fit the observational data. A discussion of this needs to be included in the manuscript and more importantly a quantification of the fit as a function of chosen Cl + SO₂ rate coefficient for each scenario provided in a table, i.e. a sensitivity study needs to be carried out.

As already mentioned above we used the MCM-chemistry without changing the decomposition rates of the Cl in any of the runs. We agree that this issue could also bring a certain uncertainty in the overall outcome but until now the values published in the literature are still so unsecure that we will not change this parameter in the MCM-chemistry used. However, we will add the discussion about the decomposition rate in chapter 5 'Uncertainties' and acknowledge helpful discussions with anonymous referees and G. Sarwar and L. Vereecken.

Concerning the recommended table of sensitivity studies on the chosen Cl+SO₂ reaction rates, the authors see no reason for it. As discussed already in detail in chapter 5 there are still a lot of different uncertainties in the context of the oxidation capacity of Criegee Intermediates in the atmosphere. Our manuscript is aimed to show under which conditions in the field these new reaction rates contribute to the observed sulphuric acid concentrations. Although there might be

smaller or even stronger new findings on the rate coefficients between CI with SO₂, NO₂, H₂O or other compounds, in the decomposition of CI or in the yield of SO₃, the main result of our paper is that the gap between modelled and measured H₂SO₄ concentrations could be explained by the oxidation of the CI with SO₂ at low and high loadings of monoterpenes with a high agreement.

4. As pointed out by the open comment the reaction of CI will also have a large impact on the impact of CI + SO₂. Again a summary of the sensitivity of the retrieved fit as a function of CI + H₂O is needed. Indeed, Angalada et al., (PCCP, 13, 13034-13045) using CCSD(T), CASSCF and CASPT2 ab initio methods in conjunction with transition state theory suggest that syn conformer of the CI reacts fast than the anti conformer, how has this been taken into account? Regardless, a quantification of the fit as a function of chosen CI + H₂O rate coefficient for each scenario provided in a table, i.e. a sensitivity study needs to be carried out.

In this point we agree with two referees and the Dr Golam Sarwar that the reaction rate of CI with H₂O vapour as discussed in other publications already can have a strong influence. We will prepare sensitivity studies by changing the reaction rate of CI+H₂O from MCM-chemistry of around 1E-17 to higher values and include this in the final version of the manuscript.

5. What is the impact of the pressure dependence on the rate coefficient? In particular, Vreecken et al., (PCCP, 14, 14682-14695, 2012) have suggest that the product of the reaction of SO₂ + CI at higher pressures would not be SO₃, however it could be a sulphur bearing secondary ozonide. What would be the impact of this? How was this taken into account in the model and again a quantification of the fit as a function of chosen CI + SO₂ branching ratio for each scenario is required.

The pressure impact has not taken into account at all. Currently we are still discussing in the community about the magnitude of the rate coefficient for the different CI with other compounds. In our opinion the influence of other parameters like pressure or temperature is of course possible but needs much more theoretical and experimental clarification before we can add these into atmospheric chemistry models.

6. The paper states that that rate coefficient reported by Welz can not be applied to the atmosphere as they were in the low pressure regime and therefore are not applicable to tropospheric conditions. Under what circumstances is it not applicable to tropospheric conditions? If the reaction is pressure independent then the low pressure rate coefficient is directly relevant to atmospheric conditions. If the reaction is pressure dependent then the rate coefficient is a lower limit, if the reaction is pressure dependent but has reached its high pressure limit by 4 Torr then the rate coefficient is directly relevant to tropospheric conditions, but the products may change with pressure. In all cases, the rate coefficient is an important guide to tropospheric conditions.

The paper states that using the rate coefficient from Welz in the MCM-chemistry for field data will lead to much higher sulphuric acid concentration as observed. However, we will change the sentence that because of the low pressure regime the new findings by Welz and co-authors are not applicable to the atmosphere based on the explanations of referee 1. And we will add another scenario including the recommended reaction rate of CI's with H₂O by Welz et al. to show the strong effect this reaction will have on the lifetime and possibility to oxidize SO₂. This is one major point which was not included in the first version of the manuscript and the authors will clearly point out in the final version that the reaction rate from Welz et al. are atmospheric realistic by including an updated rate for the water channel.

7. In the paper it is stated that if the reaction with Cl intermediates are all orders of magnitude faster the amount Cl intermediate available for reaction with SO₂ will decrease significantly, however this statement does not seem to make sense. The steady state concentration is given by the ratio of the total production rate / total loss rate. Given that the total loss rate $\sim k(\text{decomposition}) + k_{\text{water}}[\text{H}_2\text{O}] + k_{\text{SO}_2}[\text{SO}_2] + k_X[X]$ etc., where X is another reactant. If it is assumed that the $k(\text{decomposition})$ is 200 s⁻¹ (Fenske et al., J. Phys. Chem. A 104, 9921, 2000) this means that any other species must be ~ 1 ppm if its rate coefficient is $\sim 1 \times 10^{-11}$ cm³ molecule⁻¹ s⁻¹ for example to have an impact of steady state [Cl], so apart from water, the effect on [Cl] by any other reactant is tiny.

We agree with the referee that the used formulation in the manuscript for this issue is not correct formulated and we will change it in the final version.

Anonymous Reviewer Nr. 2

The paper uses ambient observations and modelling analysis in an attempt to quantify the contribution of stabilised Criegee Intermediates (sCI) as an oxidant of sulphur dioxide and a source of sulphuric acid. The paper concludes that sulphuric acid concentrations are under predicted by the model unless the reaction between sCI and SO₂ is accounted for. The paper presents a useful analysis on an important topic that will be of interest to the community. I recommend publication after the following comments have been accounted for.

Major comments

1) *It would be useful to include an analysis to demonstrate that the simulated H₂SO₄ with the standard model is statistically outside the observations given the uncertainty in the observations.*

We will perform sensitivity runs by taking the uncertainty of the measured OH-concentrations (+/- 30 %) into account and show this in the final version.

2) *The rate of reaction between sCI and H₂O needs to be discussed and accounted for. A wide variety of rate constants are available in the literature and the sensitivity of the results to this uncertainty should be explored. See also the open comments on this issue by G. Sarwar.*

Will be done, see also comments to referee 1 point 4.

Minor comments

P27695, L22. Include citations to earlier papers that also demonstrated this (e.g., Spracklen et al., 2008; Makkonen et al., 2009, Pierce and Adams, 2009, Wang and Penner, 2009).

Ok will be done

P27700, L13. No need to spell out acronym for MALTE again here.

Ok will be done

P27700, L1. Here it is not clear whether you use the models to simulate both Hyytiälä and Hohenpeissenberg or just Hyytiälä. Please clarify.

We will add in this sentence that MALTE was used for both stations and SOSA only for Hyytiälä

P27700, L24. What about data from Hohenpeissenberg?

We agree and will add an explanation which radiation data were used for Hohenpeissenberg.

P22701, L7. Same comment as above.

As mentioned above we will explain that SOSA was only used for Hyytiälä.

P27701, L20. Would be useful to say how thick the lower model layers are and how many model layers are typically within the forest canopy.

As stated in the paper, the thickness of the layers increases logarithmically from the bottom to the top of the model (3000 m). Since we used in these simulations 100 layers, it means that the thickness of the lowest layer is 0.0842 m. The thickness of the top layer is 234 m. There are 37 layers inside the canopy and 21 layers inside the crown of the canopy. We will add this more specific clarification in the final version.

P27702, L2. What do you assume for forest environment at Hohenpeissenberg?

We do not use any emission module for the MALTE-box runs at Hohenpeissenberg, so there is no need to specify the forest at this station.

P27703, L26. Only total monoterpenes have been measured at Hyytiälä. How do you know that beta-pinene concentrations are not high here?

We have GC-MS measurements for Hyytiälä from the simulated time period together with other time periods as well. For the simulated period the beta-pinene concentration was up to around 100 ppt (with occasionally higher peaks). The alpha-pinene concentration was, for the same period, at least twice as large (see also references given in the paper).

P27707, L16. Behaviour might be a better word than trend here.

Ok will be changed

P27707, L18-19. Replace the word behaviour with variability.

Ok will be changed

P27708, L22. Clarify what these percentages refer to.

We will change the sentence: ... unrealistic high sulphuric acid concentrations with 320% and 165% for Hyytiälä ... into ... unrealistic high sulphuric acid concentrations with 320% and 165% compared to the measured values at Hyytiälä ...

P27708, L5. Please include the equation for the calculation of CV(RMSE).

Will be added

P27714, L24. Inclusion of these mechanisms in a global model has now been implemented by Pierce et al. (2012). References

Will be added

References given by referee 2:

Makkonen, R., Asmi, a., Korhonen, H., Kokkola, H., Järvenoja, S., Räisänen, P., Lehtinen, K. E. J., Laaksonen, a., Kerminen, V.-M., Järvinen, H., Lohmann, U., et al.: Sensitivity of aerosol concentrations and cloud properties to nucleation and secondary organic distribution in ECHAM5-HAM global circulation model, *Atmospheric Chemistry and Physics*, 9(5), 1747–1766, 2009.

Pierce, J. R. and Adams, P. J.: Uncertainty in global CCN concentrations from uncertain aerosol nucleation and primary emission rates, *Atmospheric Chemistry and Physics*, 9(4), 1339–1356, doi:10.5194/acp-9-1339-2009, 2009.

Pierce, J. R., Evans, M. J., Scott, C. E., D'Andrea, S. D., Farmer, D. K., Swietlicki, E., and Spracklen, D. V.: Weak sensitivity of cloud condensation nuclei and the aerosol indirect effect to Criegee + SO₂ chemistry, *Atmos. Chem. Phys. Discuss.*, 12, 33127– 33163, doi:10.5194/acpd-12-33127-2012, 2012.

Spracklen, D. V., Carslaw, K. S., Kulmala, M., Kerminen, V. M., Sihto, S. L., Riipinen, I., Merikanto, J., Mann, G. W., Chipperfield, M. P., Wiedensohler, A., Birmili, W., et al.: Contribution of particle formation to global cloud condensation nuclei concentrations, *Geophysical Research Letters*, 35(6), - 06808, 2008.

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, *Atmospheric Chemistry and Physics*, 9(1), 239–260, doi:10.5194/acp-9- 239-2009, 2009.

Comment by Golam Sarwar

The authors should be applauded for their research on an emerging topic of sCl radical chemistry. To better inform interested readers about the impact of the sCl radical chemistry on sulfuric acid, I recommend the authors to consider the following suggestions:

(1) The impacts of the sCl chemistry on sulfuric acid depend on the rate constant of the sCl + SO₂ reaction as well as sCl + H₂O reaction. Hatakeyama and Akimoto (1994) reported that the rate constant for the sCl + H₂O reaction vary widely (2.0x10⁻¹⁹ to 1.0x10⁻¹⁵). The reported rate constants for the sCl + SO₂ reaction also vary substantially (Welz et al., 2012 reported 3.9x10⁻¹¹, Mauldin et al., 2012 reported 6.0x10⁻¹³, previously reported values are much lower). Mauldin et al. (2012) did not report any rate constant for the sCl + H₂O reaction. We conducted simulations using a box model and found that the sCl chemistry only enhances sulfuric acid when a lower rate constant for sCl + H₂O reaction is used. We used the rate constant reported by Welz et al. (2012) for the sCl + SO₂ reaction. Here, the authors have not stated the rate constant that they used for the sCl +H₂O reaction; I suggest that it be explicitly reported in the article.

As mentioned under referee 1 we used the MCM-chemistry and the rate constants for the reaction between sCl and H₂O as given in there. This therefore also includes the sCl + H₂O reaction rate coefficients. (According to MCM version 3.2 the reaction rate coefficient for the reactions: sCl from alpha-pinene, limonene, and isoprene + H₂O is 1.6E-17 and for the reaction: sCl from beta-pinene + H₂O is 6.0E-18). However, as pointed out under referee 1 under point 4 we will add some sensitivity studies on this reaction rates (sCl+SO₂) in the final version of the manuscript.

(2) The authors are perhaps using the lower limit of the reported rate constant for the reaction of sCI + H₂O. Hatakeyama and Akimoto (1994) suggested an upper limit of 1.0×10^{-15} , Welz et al. (2012) reported an upper limit of 4.0×10^{-15} , Leather et al. (12, 469-479, 20120, ACP) reported an upper limit of 1.0×10^{-12} . I suggest the authors also report the results of their model using the upper value of the rate constant reported in the literature. This will provide a lower and an upper range of the impacts of the sCI chemistry on sulfuric acid and the readers will be better informed of the impact of the chemistry on sulfuric acid.

Same under referee 1 point 6!

(3) The authors report that the use of the rate constant for the reaction of sCI + SO₂ reported by Welz et al. (2012) overestimates the sulfuric acid concentrations by 100%. Again, the results depend not only on the sCI + SO₂ reaction but also on the sCI + H₂O reaction. If the authors use the rate constant reported by Welz et al, 2012 for the reaction of sCI + SO₂ and the upper limit of the reported rate constant for reaction of sCI + H₂O, model will not over-estimate the sulfuric acid predictions by 100%. On the other hand, if the authors use the rate constant reported by Mauldin et al. (2012) for the reaction of sCI + SO₂ and the upper limit of the reported rate constant for reaction of sCI + H₂O, the model predicted sulfuric acid will be substantially under-estimated.

The reaction rates published by the two manuscripts from Welz et al. and Mauldin et al. have one crucial difference. The rates from Welz are based on direct measurements whereas Mauldin and co-workers used an indirect method. In the second case the 'overall' reaction rate includes also loss of the CI by the reaction with water and this may be the reason for the much lower reaction rate published by Mauldin compared to Welz (also taking the different size of the CI into account). We will perform sensitivity runs for an increased reaction rate for CI plus water for the monoterpene products and use the recommended reaction rate from Welz for the water channel of CH₂OO.

(4) The authors suggest that the new oxidation mechanism is crucial in regional and global models. We recently implemented it in a regional model and found that the impact depends on the selected rate constants for sCI + SO₂ and sCI + H₂O (Sarwar et al., 2012, potential impacts of two SO₂ oxidation pathways on regional sulphate concentrations: aqueous-phase oxidation by NO₂ and gas-phase oxidation by Stabilized Criegee Intermediates, accepted for publication by the Atmospheric Environment). When we use the rate constant reported by Welz et al. (2012) for sCI + SO₂ and a value of 2.4×10^{-15} (lower than the reported upper limit) for sCI + H₂O, the model does not enhance sulfuric acid. When we lower the rate constant of sCI + H₂O to 1.0×10^{-16} , the model enhances sulfuric acid. Since the reported rate constants for both reactions vary substantially, it will be instructive to the interested readers if the authors provide recommendation on the rate constants that should be used in such models.

In our study we used the published rate constants from Welz and Mauldin and showed that in the lowest level of the PBL the new findings from Mauldin could have a significant impact. However, concerning the many uncertainties in the production, life time and reaction path of the CI as discussed in chapter 5 we will not give any final recommendation to the readers beside what is already stated in the paper.

(5) The regional and global models do not constraint OH concentrations by observed OH values. In this study, the authors have constrained their box model with measured OH concentrations. I suggest the authors also report their box model results without constraining OH concentrations.

We chose to constrain the OH concentration in order to minimise uncertainties, and we are not in the belief that an unconstrained OH concentration will improve the certainty of our results. If you

wish to see the outcome with unconstrained OH concentration, we refer to the vertical simulations, where no constrain of the OH concentration was performed.

Comment by Luc Vereecken

The authors make an interesting analysis of SCI as a potential oxidant for SO₂, combining field observations with a model analysis to quantify the expected enhancement of H₂SO₄ formation. We have recently published a paper (Vereecken, Harder, Novelli, PCCP 14, 14682-14695, 2012; referred to as VHN below) that may shed some light on some aspects of this analysis; that paper might not have been available yet to Boy et al. when submitting their current paper.

Key parameters for the reaction of SCI + SO₂ are its rate coefficient, and the (effective) yield of SO₃ or, indirectly, H₂SO₄. Boy et al. propose (p. 27696) that the rate coefficients derived experimentally by Welz et al, and later Percival et al. (CH₃CHO + SO₂, presented at the Int. Symp. Gas Kinet. 2012) are not applicable to the atmosphere as they were obtained at lower pressure. This assertion, however, is incompatible with the potential energy surface as examined in 4 theory-based papers (Aplincourt and Ruiz-Lopez 2000, Jiang et al. 2010, Kurten et al. 2011, VHN 2012); this PES shows a barrierless addition reaction with a deep energy well, followed by several exit transition states well below the energy of the reactants. Such reactions typically have very little re-dissociation to the reactants at room temperature, and hence a total rate coefficient that is nearly independent of pressure. If anything, a higher pressure should lead to an increased rate coefficient, as already indicated by Welz et al. Pressure dependence of k(T) is therefore unlikely to be the reason for the difference between the experimentally measured SCI+SO₂ reaction rate (Welz et al., $\sim 10^{-11}$ cm³ s⁻¹), and the experimental observations (Mauldin et al, $\sim 10^{-13}$ cm³ s⁻¹). On the other hand, as discussed and quantified by VHN, the product distribution (never quantified experimentally) is expected to be sensitive to pressure. At atmospheric pressure, it was found that only a few percent of the SCI+SO₂ adduct form SO₃ directly for large SCI, with the remainder mostly forming a cyclic compound. A yield of only a few percent of SO₃ would lead to an effective rate coefficient for SO₃ formation two orders below the overall rate coefficient, compatible with Welz et al. versus Mauldin et al. This is only a tentative explanation, as the ultimate fate of the cyclic compound is as yet undetermined.

Firstly, we should probably emphasize that the reaction rate coefficient by Welz et al. is not necessarily atmospherically applicable due to large uncertainties on the pressure dependency of CI reactions. However, as you point out, there is a large indication that it actually is. We will reformulate this statement and add your mentioned references. Also, in the paragraph listing the uncertainties connected to the CI chemistry, we will mention the possibility that the formation of SO₃ from CI+SO₂ does not necessarily give a 100% yield, including a reference to VHN. VHN was, as you mention yourself, published the same time as this ACPD paper, why we have assume a 100 % yield as according to MCM.

A second point touched by VHN is the difficulty at this time to estimate the steady state concentrations of SCI in the atmosphere. The sources of SCI are ill characterized, particularly their most effective sources such as the sesquiterpenes (fast reaction with O₃, high stabilization yield). Likewise, the sinks are barely known: the unimolecular decay rate of SCI is uncertain by many orders of magnitude, the rate coefficients of its bimolecular reactions are uncertain by similar magnitudes, and the concentrations of many of the coreactants (e.g. oxygenates) are also not accurately determined. While these uncertainties are acknowledged explicitly by Boy et al., and variations on the SCI+SO₂ rate coefficient were incorporated in the analysis, it would be instructive to perform a more complete error analysis, by also examining the modeled H₂SO₄ formation using a different set of SCI rate coefficients. An example set could be the rate coefficients proposed by VHN, estimated based on the available theoretical data as recently reviewed by Vereecken and Francisco (Chem. Soc.

Rev. 2012), and found in some cases to be quite different from what is used in the MCM. In a similar vein, it might be worthwhile to explicitly consider the very large differences in reactivity/lifetime/atmospheric fate between different SCI, as highlighted by VHN, particularly with respect to their reaction with water.

We agree that more sensitivity studies are needed (for the water channel) as responded to the other reviewers and open comments. However, this manuscript aims to investigate the impact of the new published reaction rates by Mauldin et al. in the field. It would be a good idea to make a comprehensive study on all published reaction rates between the stabilized Criegee Intermediates with several compounds including the different published values for the decomposition and the yield of the SO₃ formation, however this would be wide over the scope from this manuscript.