

Editor comments

L. 58: It is not clear why you cite Buxton et al., 1997 here. This is a kinetic study and does not report on any products.

I. 93 – 95: I do not understand this new sentence. Either it seems that a word is (or several ones are) missing or the grammar is not correct. Please either clarify it or remove it.

I. 168: Remove 'greatly'

I. 192 – 196: What is the point you are trying to make here?

This sentence is way too convoluted and long. What is a direct result from your study? What is relevant to the discussion of your results? What is a logical continuation of the preceding sentence (Firstly, ..., Secondly...).

The meaning of the preceding sentence is clear: Low NO₂ leads to low NO which, in turn, weakens the titration effect of O₃.

In my opinion, you could end the next sentence after 'NO₃-': "Secondly, the reduction of NO₂ during the LCD could reduce the concentration of NO₃-".

The remainder of the sentence is unclear. Does it refer to your observations? Is all this information relevant for your discussion? – If you want to keep it, split this long sentence into at least three so it is clear how it relates to the discussion of your data.

I. 200: add 'the concentration': The concentration of OH radicals during the ...

I. 202: Add 'concentrations of': "... could lead to higher concentration of OH radicals..."

I. 222: I assume that you mean 'homologous', not 'homogenous' here.

I. 359: What do you mean by 'equilibrium concentrations' here? Why not simply 'concentrations'? Or can you say for sure that the system was in equilibrium.

I. 383: I did not mean that you should include my example in the paper. I only wrote in my comments to give you one possible example to illustrate the difference between causation and correlation.

There are likely many more possibilities why acidity and C₂ formation CORRELATE but are not CAUSED by each other.

As it is currently written it is not logical as two concepts (enhanced solubility vs acid catalysts) are mixed: You say that the acidity only affects the solubility of (M)Gly but then the reaction is acid-catalyzed.

For simplicity, I suggest removing the following text:

Such negative correlations were possibly because more SO₂ could lead to more SO₄²⁻ and lower pHs, which in turn enhanced the solubility of Gly and mGly and ultimately promoted C₂ formation. Therefore, an acid-catalyzed C₂ formation may occur in the aqueous phase under the present atmospheric conditions before the LCD.

I. 541 – 544: I do not understand the use of 'offset' in this sentence:

While previous studies focused on the importance of higher emission level and promoted secondary oxidation for producing more diacids and related compounds, this study strongly suggested that the

enhanced secondary formation of diacids and related species could offset the significant decline of organic precursors from anthropogenic pollutant emissions during the LCD.

Do you mean the following? (Please check the meaning – do not simply copy/paste this text!)
Previous studies focused on the correlation between higher emissions and enhanced production of diacids and related compounds. However, this study has shown that diacids are enhanced despite reduced emissions.

I. 549: This is overstating your findings and also the novelty of your study.

1) You did not measure OH in the aqueous phase. How can you state that you studied the effect of OH on C2 formation?

2) It is by far not the first study that inferred OH-driven C2 formation. There are many studies that – just like you – showed similar trends, e.g. (Crahan et al., 2004; Jiang et al., 2014; Mochizuki et al., 2017; Rinaldi et al., 2011; Sorooshian et al., 2006; Wonaschuetz et al., 2012; Zhu et al., 2020)

This list is by far not complete or exhaustive. They also did not measure OH directly. Some of the studies used models with measured data as input and this way could indeed quantify the role of OH pathways, based on constrained OH concentration, exceeding your approach.

I am not saying that you should add any of this to the paper. Instead, I suggest removing ““for the first time successfully””.

I. 552: Do your data really support any conclusions on the reactivity of ^{13}C ? You saw a trend that C2 was enriched in ^{13}C ...such a trend could be equally explained by slower loss processes of ^{13}C enriched oxalate. I suggest limiting your concluding section to facts that are supported by your results, e.g. “We found higher $\delta^{13}\text{C}$ values of C2 during the LCD than before the LCD (Fig. 10).”

References

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