Dear Reviewer Two,

Thank-you for your review of our manuscript and targeted comments. We believe the changes we have made based on your perspective and comments improve the manuscript.

The manuscript describes a novel and accelerated method to determine the chemical composition of liquids for e-cigarettes before and after heating. The method uses an e-cigarette coil, submerged in the e-liquid to be analyzed. The liquid is filled into a small beaker, which is kept tilted at an angle of 45° so that aerosolized droplets would be collected on the beaker wall and run back into the liquid reservoir. Eventually, the remaining liquid in the reservoir is chemically analyzed by GC-MS. I have to admit that for me as an aerosol scientist, who is not very familiar with the characterization of e-liquids and e-cigarettes, the manuscript is difficult to follow.

• Thank-you for this feedback. We have considerably revised the manuscript for clarity and brevity and hope that this greatly improves the ability to follow the manuscript.

I am particularly struggling with understanding, why (according to the title) the paper is supposed to describe a quantitative method to analyze e-cigarette aerosols.

Thank-you for addressing this concern and for the opportunity to improve the manuscript by clarifying the aerosol detail as it was not conveyed clearly. The following description has been added now on line 104 of the manuscript to describe our premise more clearly behind the "accelerated method" and how it might represent the aerosol:

"Our premise for collection of e-cigarette aerosols in the liquid was as follows:

- 1. An e-cigarette is an evaporation condensation aerosol generator intended to modify the eliquid as little as possible during aerosolization, however, it does thermo-oxidise, hence the need for this research;
- 2. Our "accelerated" method of a heating the e-liquid via a submerged coil creates a "bubbling aerosol generator" (Vidamantas, 1997). Like an evaporation aerosol condensation generator, a bubbling generator will modify the e-liquid minimally, however, may allow more volatile compounds to preferentially aerosolise;
- 3. The creation of an aerosol via bubbling can allow aerosol capture either whilst bubbling through the bulk liquid (when cooling) or at the gas-liquid surface (Ghiaassiaan, 1997 and Koch, 2012);
- 4. Surface bubbles can generate aerosol either by jet or film droplets when they burst, and based on combinations of surface tension and bubble size, aerosol will recombine with the liquid the bubble arises from when it bursts (Koch, 2012, Mead-Hunter, 2018).
- 5. Thereby, a combination of these processes should ensure we retain a representative sample of the same material that is aerosolised, as well as possibly more of the thermos-oxidised (aged) material we are interested in."

The premise of the study was to produce an e-cigarette aerosol that would be immediately captured. Eventually, only liquids are analyzed and I miss a description why the analyzed liquids are supposed to be representative for the emitted aerosol (I assume that this is the goal). The authors should focus the description of the methodology more towards the aerosol characterization, bearing in mind that the journal is on "Aerosol Research".

• Thank-you for this observation. We hope that the above description and the related modification to the manuscript on line 104 greatly improves the description of how the analysed liquids are representative of the aerosol.

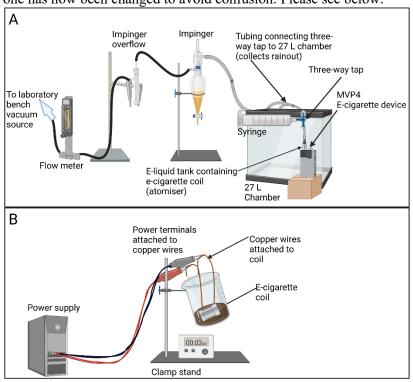
I also wonder how "quantitative" the method really is, considering that the fold-change comparison in Figure 3 reveals rather large discrepancies of the two methods and nothing is mentioned about the accuracy and recovery rate of the reference method. Shouldn't the method rather be termed "qualitative" or semi-quantitative"? For a quantitative method, I would expect to see more analyses on the method's uncertainty.

• Thank-you for this observation regarding the accuracy of the title wording "quantitative". The title has been changed to <u>semi-quantitative</u>.

Specific comments:

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- Line 91: "vacuum" should read "vacuum pump" (also applies to figure 1)
 - The image of a vacuum pump was a stand-in for the lab bench vacuum source. Figure one has now been changed to avoid confusion. Please see below:



- Line 91/92: model number of the flow meter is missing
 - our apologies for missing this detail it has now been added on line 90.
- Line 99: If the flow rate is 3 l/min and the total duration 4 h (240 minutes) then the total volume should be 720 l, not 7.2 l
 - Apologies it was not clear was the 7.2 L was referring to. This has been clarified in the revised manuscript (line 97) which shows that we refer to 7.2L of e-cigarette aerosol, not 7.2L of total air flowing through the system;

"This process was repeated every minute for two hours (with the atomizer tank refilled after \sim 60 minutes), for 120 puffs total. While we acknowledge that vaping topography is

extremely variable, 120 puffs over a 2 hour period (120 x 60 mL, puffs, therefore 7.2 L of inhaled air) was chosen to be representative of what a typical vaper might use (Etter, 2014).

- Line 110/111: What is meant with "aerosol (...) would recondense on the wall". As far as I understood, the goal is to collect only droplets and no (condensed) vapors. Do you mean that aerosolized droplet would collide with the wall and flow back into the reservoir? On the other hand, how certain is it that vapors do not recondense? And what is the efficiency for the collection of the droplets on the wall? I assume that this would be only based on impaction. Any information on the droplet size, velocity and Stokes numbers?
 - The authors were not able to measure this mode of collection with certainty and calculation of capture rates by impingement on the beaker wall would include many assumptions. This is recognised as a limitation of the study on line 225 to 230.
- Line 280/281: Here, the authors claim that the method potentially (!) allows for analyzing resultant aerosols.
 - Theoretically we were able to capture resultant aerosols as is now clarified on line 104 however some limitations of the method were identified insection 4.1, therefore the method only "potentially" captured aerosols, pending minor modifications to the described method. We have therefore changed the word potentially on line 284 to the phrase "with minor modifications", to more accurately represent what the authors were intending to convey.
- Figure 3: There is a typo in the caption of most axes: "Tirimasu" should read "Tiramisu"
 - Thank-you this has now been corrected.