Summary:

This study describes the characterisation and operational use of an ambient gas sampling system consisting of sorbent tubes and glass vials installed on a UAV with MTOW < 25 kg. Target gases include CO2, CH4 (and their C-13 fractionation), and a range of VOCs, all measured offline (in the lab) by GC-C-IRMS etc. Performance is described with reference to certified gas and C-13 isotope standards and interpreted for mass recovery. Finally, some interesting field measurements are reported for these gases by UAV sampling near to the surface over both a landfill and forest site.

This review is limited to ambient atmospheric sampling, UAV operations, C-13 isotope measurements and GHG concentrations in general, as I am not a specialist in offline GC-IRMS methods or sorbent tube sampling.

In terms of context, this paper is most welcome as it continues to push the boundaries of what UAVs can offer as an atmospheric sampling platform. A system that can use UAVs to take whole air samples for later high-precision laboratory analysis is an interesting and novel contribution to a rapidly growing measurement field. Measurements such as those described are likely to have high scientific value to others in the field of GHG and VOC emission research.

Overall, the paper seems to have excellent scientific value and originality and high standards of academic rigour. However, the presentation of the paper does not do the work justice. It is currently very hard to read and absorb and contains many grammar mistakes and sentences that make very little sense. However, this could be fixed with a thorough proof-read and edit by an experienced technical writer. Some technical aspects are not described as completely as might be necessary for this to be an excellent resources for others to follow. It would be a much better paper if it can be written a little better.

I have a technical question about the nature of the WAS system and how this may impact the accuracy of ambient measurements of GHGs and their isotopic fractionation (see below). My main comment is that the paper needs revision to make it more accessible - a relatively minor correction, and some attention to detail on figure captions etc.

We thank you for your helpful review and can present you with a completely revised version. The content of the revised manuscript was corrected by a scientific colleague with the same topic and who is native English speaker in order to hopefully be able to present you with a comprehensible manuscript

Specific comments:

1/ Line 11 - In the abstract it is stated that the instruments can be mounted to UAVs (and reads/implies that any UAV can carry them). However, not every UAV can carry them (some UAVs are extremely small and can carry no payload). I would suggest that this sentence is better qualified by saying "...can be mounted to a UAV with a payload capability greater than X kg..." and insert what x should be. Moreover, this sentence overall does not make sense grammatically – insert a full stop before "for all compounds...".

Revised

2/ Line 20 – what are "replicates" in this context? I've not heard this terminology before. Also, what is meant by "triplicate" CO2 measurements – does this mean that the same sample was measured three times to arrive at the stated precision, or that three measurements of different samples were used to calculate the precisions? I'm struggling to understand what was done to arrive at the precisions stated in the abstract here as the terminology is very confusion.

The abstract was modified to avoid such awkward wording and provide the reader with a clearer overview. The meaning of "replicates" and "triplicates" is now explained more clearly in the methods and results.

3/ Introduction. The literature review may be well served by citing and discussing a recent review paper of UAV methane measurements by Shaw et al., 2021 - <u>https://royalsocietypublishing.org/doi/full/10.1098/rsta.2020.0450</u>

The paper of Shaw was added to the introduction, because it provides a helpful summary of available sampling and measurement setups including payloads and measurement precision.

4/ Line 70 – the studies cited here are examples of such studies and not an exhaustive list as the list of citations implies. If you only wish to cite these three examples, please add "e.g.". Otherwise, a more complete list of UAV GHG measurement studies up to 2021 is described in Shaw et al., 2021 review paper (link in the comment above).

Revised.

5/ Section 2.2.2. The WAS system appears to consist of 12 evacuated (flushed and pretreated) glass vials. Injection of ambient air appears to be via a side-port needle. Are these vials evacuated to 0.5 Pa as suggested on line 151? If not under high vacuum, the injected (ambient) air would be mixed with whatever residual air was in the vial, potentially biasing the concentration and C-13 fractionation measured offline (by high precision instrumentation in the lab) toward the concentration and fractionation of the residual air inside the vial. The final sentence of this section states that "The dead-volume of the transfer line (100 μ L) and the residual flush-gas volume inside the evacuated glass vial did not show any significant influence to follow-up measurement setup." This is an example of a sentence that doesn't make complete sense to me (i.e. what is "follow-up measurement setup?). Please could you provide more information/data on why you are confident that there was no significant influence, and what you define "significant influence" to mean (quantitatively)? I am concerned that this effect could greatly bias the GHG concentration and isotope measurements made from the filled vials. GHG concentration measurements are required to have a very high precision to be acceptable under WMO reporting standards. The text reads as though it is only the initial negative pressure that fills the tube to an equilibrium pressure with ambient pressure. If the vials are indeed evacuated to 0.5 Pa (high vacuum), and there is only 100 ul residual in the transfer line, I would agree any bias would be very small, but it may be useful to quantify the maximum expected bias this may introduce for each quantity measured. Please could you explain and clarify this in the response and in the manuscript.

Revised. Vials were evacuated using a rotary vane pump, which can achieve 0.5 Pa. Beforehand, vials were flushed with synthetic air to ensure that the residual gas volume after the evacuation does not contain residual air constituents. The description of the vial conditioning now also incorporates the results of conditioned vial, which were then filled with synthetic air to reveal measurement blanks.

6/ Figure 2 – the caption does not describe what the error bars represent – what do these represent?

Revised

7/ I'm pleased to see the honest discussion about the potential effect of propeller downwash, especially for near-surface measurements where disturbed air may be sampled, making it very hard to assign a measurement height to sampled air. Others have tackled this by only recording measurements on rapid descent profiles (or rapid ascent profiles where vertical profiles are required), or by taking measurements forward in the direction of motion horizontally (moving fast enough such that downwash does not have time to mis/disturb the air ahead of the UAV). It may be useful to discuss this as a potential mitigation to these sampling problems.

Revised. References in concern of flight and sampling paths were added.

Technical comments:

Line 15: Change to "...sampling or collected and sampled..."

Revised

Line 23: "The results emphasized the functionality

of the sampling and measurement setup described, demonstrating that it a viable tool for monitoring atmospheric trace gas inventories and identifying emission sources." - This is a bit superfluous and confusing – recommend changing text to read: "The results demonstrate that the UAV sampling system here represents a viable tool for monitoring atmospheric trace gas inventories and identifying emission sources". This is just one example of many sentences that are rather verbose and partially meaningless. It may also be useful to emphasise the isotopic capability of this system versus other in situ measurements of trace gas cocentrations, and how the two may complement each other (a combination of such systems could be very powerful).

Revised

There seems to be an inappropriate use of commas in many places throughout the manuscript, e.g. the sentence beginning on Line 45 does not need a comma. Please proof read and check correct use of commas throughout.

Revised

Line 50: This sentence makes no sense. I think a full stop may be missing between "precision" and "sample"?

Revised

Line 391: What does "chilled" mean in this sentence? Was the sampling entrance point chilled?

<mark>Revised</mark>

Line 395: "...of height 13,..." please add "m" after "13".

<mark>Revised</mark>

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