

Thank you to the thoughtful reviewers of this manuscript. Their input has helped improve the quality of the manuscript.

Author comments are in blue.

Review 1

General Comments I appreciate the work by the authors towards addressing the reviewer comments in this updated manuscript draft, and overall I am more satisfied with the quality of this study. Most of my specific concerns about measurement quality were commented on in the updated draft, and the relevance of this paper within the literature is established much more clearly. After addressing a handful more comments I am willing to consider this paper for publication.

Thank you for your careful consideration of this manuscript.

Major Comments 1. Figure 4: Thank you for updating the layout of this figure for clarity. As per my original comment, I additionally request the authors to update the color palette on this figure to something other than rainbow, as I find it difficult to read differences in adjacent points without a perceptually uniform palette. Depending on the programming language used to make this figure, I recommend a color map from the “cmocean” package (links for MATLAB, Python).

This figure has now been remade with a different colormap.

Minor and Technical Comments 1. For a paper focusing on novel observations of O₃, I would be in favor of moving the supplemental figures S1 and S2 into the main paper for easier reference, especially if they are referenced in the text anyways. Please consider moving them to Section 2. 2.

Both images have been added as Figure 2. The rest of the figures have been renumbered accordingly.

P8, L210: Thank you for adding this text with discussion on the iMet-XQ2 performance. There is a double reference for Kimball et al. at the beginning of this line, please update.

The second reference to Kimball has been deleted.

Review 2:

This paper has been rewritten to address most of the comments of the reviewers. It is certainly improved from the first version, and is now more limited by the experimental work rather than by the writing and organization. To reiterate from my previous review, the POM is an attractive choice for balloons (because it can be used on small balloons without obtaining special flight permissions) and UAS (for the same reason, and for its light weight and small size). **But is it really adequate for atmospheric measurements? Are the improvements and challenges needed to make it work mostly related to properly integrating it into the right UAS, or do improvements (or modifications by the user) need to be made to the sensor itself? The authors may not feel comfortable making these statements, but the answers to these questions would be useful to the community.** I would be happy with “suggestions for further improvements” being added to the Conclusions section, although the second-to-last paragraph on p. 15 (discussion of Li et al., 2020) may be a better spot, given that the authors have started to address this in that section. It is also OK

for the community to simply see what has been done and draw their own conclusions - I don't want to force something that is not fully supported by the work.

We agree that a section that summarizes what we learned from this experiment and what suggestions a reader should take home for implementation or further improvements is worthwhile to include. We added a suggestions section to Conclusions:

“In this study, the POM performance on UAS was improved by inlet positioning and slow flight parameters, top-mount placement on a robust UAS and increasing the rate of calibrations to pair each calibration with specific battery power source improved the precision and accuracy. However, added thermal insulation, as described by Li et al, appears another promising additional consideration for improved performance of the POM on UAS. The POM appears to be a robust enough instrument for course atmospheric measurements in the atmosphere (to 2 ppb precision) but integration onto a UAS should be carefully considered.

Specific comments (line #s from the “Track changes” version, not the final version):

P.2, l. 35 Certainly add “e.g.” before Kaser et al., perhaps elsewhere as well if appropriate.

Added to line 35

l. 43-45 Again, there is nothing in the Beekman et al., 1997 reference about tethered balloons over water. My confusion about this sentence is that, as written, it looks like it is one subject or thought, but actually I guess it is three different things. If you add “and” before “associating high ozone” it will make much more sense. If the large set of references at the end of the sentence are split up so some follow “profiles over water/urban”, some follow “ground to 1500 m”, and some follow “UT/tropopause folds” it will be helpful.

This sentence has been recrafted to specifically identify some aspects of each of the sources cited.

It now reads:

Tethered balloons have been used to study vertical ozone (Demuer et al., 1997; Peng et al., 2008; Knapp et al., 1998; Zhang et al., 2019; Greenberg et al., 2009), and meteorological conditions (Chandrasekar et al., 2003) gathering data at heights ranging from ground level to 1500 meters above ground level, which included evaluations of episodes of biomass burning (Xu et al., 2018) and mesoscale modeling of ozone in the upper troposphere (Peng et al., 2008).

l. 50 “lower free troposphere”?

now says “in the lower troposphere”

P.6, l. 145 “ozone concentrations” or “mixing ratios” or just “ozone” instead of “measurements”.

Deleted concentrations.

l. 172 “install” or “add” instead of “put”?

replaced “put” with “place”

l. 184-85 It seems that these two sentences could be combined into one, “The 2B Tech personal ozone monitor, POM, measures atmospheric ozone concentrations via UV absorption...” Also, I don’t think this paragraph is an accurate summary of Wilson and Birks, 2006, in that the artifact can affect both dual and single cell instruments. It would be best if the authors simply state how they addressed the issue of artifacts from (changing) humidity, either using something provided by 2B Technologies, or their own design, or if they did nothing. Fine to have a short, accurate sentence about what causes the artifacts, from Wilson and Birks and/or subsequent work.

The sentences have been rearranged to stress how the POM works following the reviewer’s suggestion.

P.9, l. 233 “both indistinguishable from zero”. In table 1, 3 out of 4 tower gradient measurements are statistically different from zero. Not sure what “both” means here. And I’m not sure why the gradient is more important than the actual values measured. They may look better in comparison, but they have twice the uncertainty. It does suggest whether the reason for disagreement is a zero offset, or something else.

Many boundary-layer parameterizations for mixing, flux-profile relationships, and so on are functions of gradients of scalars, momentum, or heat more so than absolute values. For some applications that may be more important, while for others the absolute magnitudes matter. We have updated the sentence to clarify meaning.

l.237 “larger differences” – larger than what? A simple rewrite of this sentence or section should be able to fix this.

Removed the word larger.

Comparing Table 1 to Figure S3, for July 16 the POM UAS gradient looks like it should be close to -20 ppb. Or is there a second blue square very close to the tower data?

There are overlying datapoints. The figure (Now S1) has been edited to only have one data point per altitude which was used to calculate the ozone gradient from the POM observations.

Actually, some of the ToF data in S3 are a little suspicious too, particularly where they go close to zero on July 11. There are a number of outliers in the tower data, and they (very nearly) all seem to be triangles, or ToF measurements. If the averaging for both tower instruments are done the same way, it suggests that there are some things to be cleaned up in the ToF data. This is beyond the scope of this paper, but affects how much one might trust the comparison. In this case, the results clearly indicate that some improvements needed to be made in the O3/UAS system, as discussed

next.

Table 2 –

We have updated Figure S1 (in previous version S3) to include the quality controlled TOF data that was posted to the CHEESEHEAD repository. These data do not include outliers that are above 75 ppb. The lower values from the TOF are considered indicative of reactive chemistry with biogenic VOCs or soil NO_x within the canopy (see Vermeuel et al 2021 GRL fluxes) or, less routinely, from reactions with NO_x emitted from a station power generator that was irregularly tested. It is not uncommon for O₃ values to reach <10 ppb at night in this region, as we have recorded values as low as 4 ppb at this same site in Fall 2020 (Vermeuel, et al. 2023) with the same Thermo 49i instrument used in this study. Further, the 1s-averaged limit of detection for this ToF is ~10 ppt (Novak et al., 2020), which allows for quantification at these low [O₃] levels.

Ozone deltas were recalculated for Table 1, based on the updated dataset from the CHEESEHEAD19 repository which did not include data from July 16, so that data point was omitted from the table, Table S1 was also updated.

Sources:

Vermeuel, M. P., Cleary, P. A., Desai, A. R., & Bertram, T. H. (2021). Simultaneous measurements of O₃ and HCOOH vertical fluxes indicate rapid in-canopy terpene chemistry enhances O₃ removal over mixed temperate forests. *Geophysical Research Letters*, 48(3), e2020GL090996.

Vermeuel, M. P., Novak, G. A., Kilgour, D. B., Claflin, M. S., Lerner, B. M., Trowbridge, A. M., ... & Bertram, T. H. (2023). Observations of biogenic volatile organic compounds over a mixed temperate forest during the summer to autumn transition. *Atmospheric Chemistry and Physics*, 23(7), 4123-4148.

Novak, G. A., Vermeuel, M. P., & Bertram, T. H. (2020). Simultaneous detection of ozone and nitrogen dioxide by oxygen anion chemical ionization mass spectrometry: a fast-time-response sensor suitable for eddy covariance measurements. *Atmospheric Measurement Techniques*, 13(4), 1887-1907."

For the measurements at Park Falls in 2020, why not use the DJI hexacopter?

We did not have access to the DJI hexacopter in September 2020 for the measurements in Park Falls as it was owned by Purdue University.

The table is fine for comparing the iMet measurements, but the UAS had already been upgraded.

I don't quite understand this comment. For all observations at the Tower site, the Yuneec Typhoon H UAS was used. For the Summer 2020 Chiwaukee Prairie observations, the DJI M300 was used in the collaborative project with Purdue University.

P.10, l. 260-61 I'm not sure why the word "dimensionality" is used. The atmosphere is inherently 3D, and any experiment or analysis needs to consider that. (Or it is 4-D including time, and the short duration of small UAS flights is not necessarily well-matched.) Maybe it is the small-scale vertical (and perhaps horizontal) structures that are well matched for UAS. Sorry to be so picky here, I think I know what you mean, but I also think this can be improved.

So edited to state

"The viability for UAS-mounted ozone observations to capture low-altitude features in ozone is well-matched to the **small-scale vertical structure** of marine layer ozone concentrations in a near-shore environment."

P.15, l. 352-370 Thank you for including this! Worthwhile to check wording in places though – for example, "did a heating method" on l. 360. Maybe something like "Li et al. state only that the regulatory monitor for comparison used a heating method for removing water vapor interference, instead of..." (It's hard to avoid overusing the verb "use", but "employ" is another option.)

So changed to:

Li et al. address only that the regulatory monitor they used for comparison which employed an in-line heating method for removing water vapor interference, instead of a dual-cell active subtraction in parallel as is typical for other regulatory monitors.

See sentence on l. 373 – besides "used" and "using", it has "uncrewed aerial systems" as a type of "UAS". Could change that to the type of system – hexacopter, etc.? Also, I'm not sure if there is a "water vapor absorption" or an interference; please check that carefully and other similar mentions in the paper.

Edited this section for clarity

l. 368 "constant ascents"?

Edited

P.16, l. 378 "Lake Michigan"

Edited

P.17 Figure 4 is much easier to read now, despite (or perhaps partially because of) the shorter horizontal scale for potential temperature.

P.18, l. 390 Can you make this first sentence of the Conclusions more related to this paper, instead of the general "has a proven utility", which could be determined from the existing literature?

l. 391 "including" instead of "included" or rewrite this sentence. (The content is fine.

So edited

l. 395 “towers”?

Not sure what this is in reference to

Line 33 on p2 is the only place where the word “towers” shows up.

References:

- Chandrasekar, A., Philbrick, C. R., Doddridge, B., Clark, R., and Georgopoulos, P.: A comparison study of RAMS simulations with aircraft, wind profiler, lidar, tethered balloon and RASS data over Philadelphia during a 1999 summer episode, *Atmospheric Environment*, 37, 4973-4984, 10.1016/j.atmosenv.2003.08.030, 2003.
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- Greenberg, J. R., Guenther, A. B., and Turnipseed, A.: Tethered balloon-based soundings of ozone, aerosols, and solar radiation near Mexico City during MIRAGE-MEX, *Atmospheric Environment*, 43, 2672-2677, 10.1016/j.atmosenv.2009.02.019, 2009.
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- Peng, Y. P., Chen, K. S., Lou, J. C., Hwang, S. W., Wang, W. C., Lai, C. H., and Tsai, M. Y.: Measurements and Mesoscale Modeling of Autumnal Vertical Ozone Profiles in Southern Taiwan, *Terrestrial Atmospheric and Oceanic Sciences*, 19, 505-514, 10.3319/tao.2008.19.5.505(a), 2008.
- Xu, Z. N., Huang, X., Nie, W., Shen, Y. C., Zheng, L. F., Xie, Y. N., Wang, T. Y., Ding, K., Liu, L. X., Zhou, D. R., Qi, X. M., and Ding, A. J.: Impact of Biomass Burning and Vertical Mixing of Residual-Layer Aged Plumes on Ozone in the Yangtze River Delta, China: A Tethered-Balloon Measurement and Modeling Study of a Multiday Ozone Episode, *Journal of Geophysical Research-Atmospheres*, 123, 11786-11803, 10.1029/2018jd028994, 2018.
- Zhang, K., Zhou, L., Fu, Q. Y., Yan, L., Bian, Q. G., Wang, D. F., and Xiu, G. L.: Vertical distribution of ozone over Shanghai during late spring: A balloon-borne observation, *Atmospheric Environment*, 208, 48-60, 10.1016/j.atmosenv.2019.03.011, 2019.