We thank the anonymous Referee #2 for his constructive comments, which helped us to significantly improve our manuscript with respect to the version published in AMTD.

Below, we provide point-by-point responses to his comments, which have been copied and pasted in the present document, where they appear in italics. Our responses – including details on how the manuscript has been changed – come between the Referee's lines.

General comments and changes

This paper presents a novel approach for performing Lagrangian measurements using constant volume balloons and ozonesonde instruments. The authors perform both ground-based (controlled) tests and a few limited comparisons with coincident data from three summer field campaigns in which the special balloons were deployed. Overall, the authors do a reasonably good job demonstrating the effectiveness of the approach in a paper that I found interesting and enjoyable to read. The approach described here could well be adopted by other measurement groups, so the work presented here is important to the sonde community. In fact, it may present the simplest way to sample an air parcel in a Lagrangian manner.

I had a number of questions as I read along, many of which the authors answered, but some of which remained unanswered. The approach of cycling the power on the ozone pump was at the center of my concerns/questions:

1. How is the pump efficiency affected by all of the power cycling?

2. How does the pump motor current (and indication of the pump efficiency and flow rate) vary with time?

3. What happens to the box temperature?

In Figure 6 of the revised manuscript (discussed in section 3.1.2 "Pump flow in warm-up regime"), a figure panel has been added, showing the volumetric flow rate evolution in absolute value during 7 work cycles of about 150 s (in addition to its relative variation, already shown and commented in the AMTD paper, Fig.7). Regarding the steadiness of the flow rate, it was found to vary by less than 3% over all the 7 cycles. This conclusion has been emphasized in the revised paper text.

During this experiment, several motor parameters were also recorded, including motor voltage, current intensity, rotation speed, and pump temperature (Figure i below). The motor voltage was found to be very well regulated and almost constant (relative variations of less than 0.01%). Consequently, the electrical power consumption was driven by the current intensity, that varied (generally decreased) by less than 3% during each work sequence. Despite this, the motor rotation speed remained quite steady (weak decrease by less than 0.1% in general – except -0.3% in one accidental case (black curves), when the current also increased).

Concerning the pump body temperature, it increased almost linearly by about 0.5°C during each work sequence, because of energy dissipation in the motor and the pump. During BLPB flights, the temperature value used to calculate ozone mole fraction was the temperature recorded at the end of each measurement sequence. The error on the mole fraction resulting from temperature variation within each sequence remains lower than 0.2%, which is negligible.

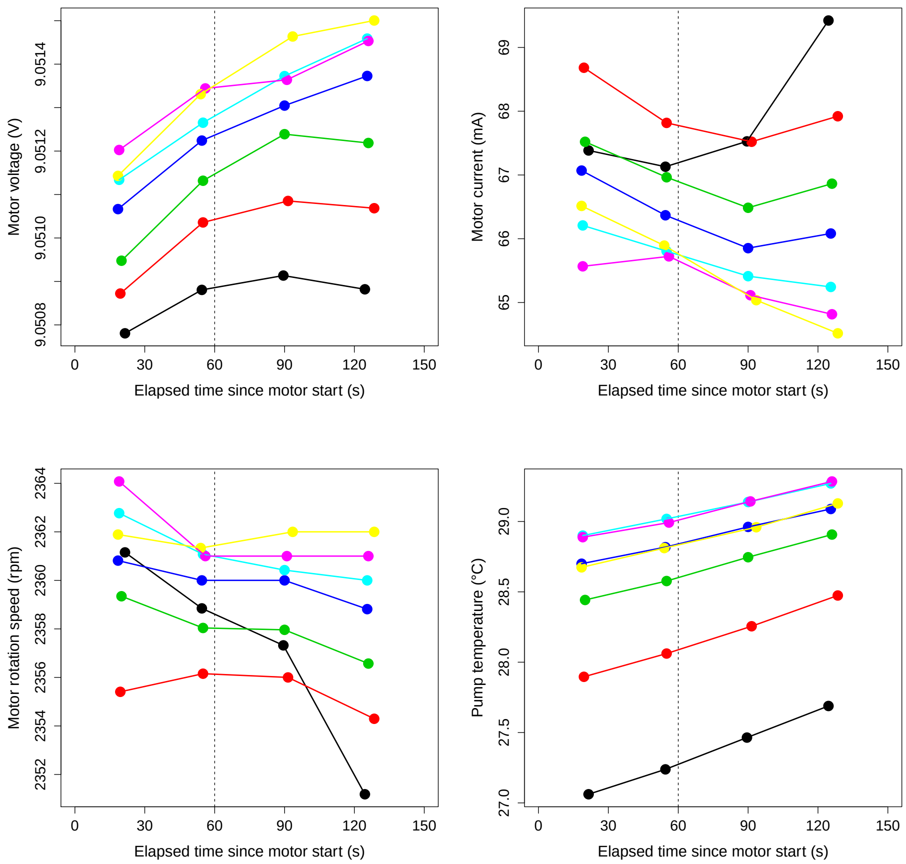

Figure i: Motor parameter evolution after sonde motor restart: (a) Voltage (V); (b) Current (mA); (c) Rotation speed (rpm); (d) Pump body temperature (°C).

Perhaps more informative but not mentionned in the AMTD paper, is the box temperature evolution over the course of a BLPB flight. For instance, during flight B55 (the longest flight presented in the paper), the box temperature varied from +36.8°C in the late afternoon on the launch area, to +14.9°C at ceiling (2400 m) during the night, then to +27.9°C during the next day and eventually to +15.1°C during the second night. Those long-term temperature variations were of course taken into account in ozone calculations.

We think that presenting Figure i and the comments above would make the paper too long, however we suggest all this could be presented as online supplementary material.

4. How is the apparent background current affected? There is some evidence that *the background current may not be constant – that it goes down as the instrument runs. Your data hints that might be the case as well. Did you take a look at that? That might be something that could be checked on a running instrument.*

The Referee is right: the background current is slowly drifting (decreasing) as long as ECC sondes run (e.g., Vömel and Diaz, AMT, 2010), which limits their ability to detect and quantify ozone trends in the troposphere. We did not specifically address the question of drifting background current with a dedicated experiment (e.g., intermittent sonde supplied with zero air over a long time). However, the material presented in the revised paper, especially Figure 8(b) (9b in the AMTD paper), is sufficient to give an upper limit to the effect of a background current drift. In Figure 8(b), a trend of +0.07 ppbv/h on measured ozone was evidenced from the difference between the sonde measurements and a UV-absorption analyzer (assumed to have negligible drift over the 5-day experiment). In the AMTD paper, we attributed this drift to solution evaporation only. This cannot be ruled out, but long-term background current drift is also a possible cause. A comment has thus been added in Section 3.2. But whatever the cause of drift in our experiment, we can conclude that ozone trends faster than ±0.1 ppbv/h in the atmosphere are detectable without ambiguity. Our conclusion remains unchanged on this point.

Here were my other larger concerns:

1. Your controlled tests looked at the response of the instrument with ozone varying over a pretty tight range (35 45 ppb), yet your actual data vary over the range 20–80 ppb. How confident are you that the responses over that larger range match your controlled tests in the tighter range? And if someone else wanted to use this approach, how confident would you be in your recommendations if the ambient ozone was closer to 120 ppb rather than 50 ppb? While the percent differences might track, the absolute differences would be quite different from one another, I think, and those absolute differences are meaningful to some in the audience that might like to employ this technique.

The Referee's concern is not perfectly clear to us. The experiment described in Figure 7 (revised manuscript) demonstrate that the sonde can cope with ozone variations in the range 0-60 ppbv. Ozone mole fractions up to 85 ppbv were also recorded during the balloon campaigns (e.g. flight B59 – but with no reference measurement for validation).

The Referee may also wonder whether the 60s warm-up phase would be long enough for ambient concentrations higher than 60 ppbv. In other words, would our approach and settings be recommended for measurements in very polluted plumes or in the stratosphere, with a satisfactory absolute accuracy (e.g., ±10 ppbv)? We are not able to answer this question from the experimental material presented in this paper. A supplementary experiment would be required, whereby an intermittent sonde would be exposed to ozone step changes of large amplitude (200 ppbv, for instance) by means of an ozone calibrated source. This is however beyond the scope of the present study. The question of using sonde intermittency in the stratosphere is very exciting but deserves further work.

2. HYPSLIT: never rely upon a single trajectory to help understand air mass his-

tory.

The HYSPLIT simulation has been recomputed using the ensemble option, resulting in 27 backtrajectories ending at the same point. In addition to the pathway already shown by the single trajectory in the AMTD paper, the ensemble reveals a second one from south-east continental Spain. This source region may thus influence the composition of the air mass tracked by the balloon flight B55. Figure 15(b) and the text of Section 4.4.2 have been revised consequently.

Recommendation: This paper should be published after some minor edits and with the answers to my questions.

Detailed comments and changes

In the abstract, how does the 1–2 ppb/hr compare with expectations?

A mention to the previous work by Bénech et al., 2008, has been added.

Below, the comments are listed by page number line number.

2-11: Do you mean to say the lower tropospheric ozone or do you mean what you said, the low tropospheric ozone concentration? Just want to be clear.

The sentence has been rephrased.

2-21: change mean to method

Done.

3-32: this info is probably relevant to my question about the abstract above.

Yes. The mention added in the abstract refers to Bénech et al. (2008) too.

4-17ff: I think most of this section could be replaced with a reference to Komhyr (1969) and subsequent papers. The info about the solution choice is important – the 0.5% half-buffered solution was the recommendation of JOSIE. One question I did have was whether you did the high ozone conditioning of the cathode cell or just the pump? Referring to Appendix B, it would seem you chose to bypass the cathode cell. Is that correct?

We find useful to keep Equation 1 and accompanying text, as it is the formula we actually used to retrieve ozone mole fractions, and also because it allows to recall and clarify which variables are needed, measured inflight or at the ground before the flight, etc. The information on the solution has also been kept.

Note that the appendices have been discarded and the whole section revised consequently, as recommended by the other referee. In turn, the sonde preparation (which mostly follows the GAW standard procedure) is no longer detailed in the paper. To answer the Referee's question, the cathode cell is conditioned with high ozone concentration only once, at the beginning of the advanced preparation. Then the cathode cell is bypassed during the flight-day preparation, when the pump body is again conditioned with concentrated ozone.

6-12: All of the 2Z En-Sci/DMT sondes I have used seem to have just one set of batteries – 2 9-volt batteries in series. I have not used the 1Z sondes – are they powered with two separate batteries?

Yes: one 9-V battery to power the electronics; one 12-V water-activable battery to power the pump motor. On our side, we have no experience with 2Z sondes, so we were not aware of this difference. To avoid confusion, the sonde model (Z) has been specified in the corresponding line.

6-15: . . .only a few percent.
Amended.

8-5ff: Im curious how you did this. Did you let the stopwatch run continuously and try to read it when the film crossed the 0 and 100 mL lines? How precise can those measurements be? What kinds of uncertainties are associated with your flow rate measurements. And since in Figure 7, you tie the y-axis data to the reading near 60 s, the precision of those measurements is going to affect that plot.

We used a sport timer that allows for logging up to 16 lap times. Only a button click is needed to log a lap time – the logged lap times being read afterwards. Therefore, the pump-time measurement precision is just the same as usual with a soap-film flowmeter. The GAW panel of experts report an accuracy below 1% for a volumetric flow rate in the range 200-230 ml/min. This is consistent with our own estimation, where the uncertainty is ± 0.2 s for a measured pump-time of about 30 s.

We acknowledge that an uncertainty of 1% on each flow-rate value is of the same order of magnitude as the observed decay (1-2%). Despite the measurement uncertainty, the decaying trend appears to be a common and robust feature of the 7 data series. Our conclusion (possible variation of the flow rate by 1-2% during each measurement sequence) remains very cautious and does not aim further than suggesting future technical developments. Thus, we do not feel that refining the experimental support is needed in this paper.

Our motivation to tie the y-axis to the reading near 60 s was to clearly show the evolution of the flow rate during the measurement phase, which starts after the 60-s spin-up. A figure panel has been added to show the absolute flow rate values (new Fig. 6a). Beyond the question of the uncertainty discussed above, the rescaling used here does not affect the conclusion.

8-12: . . .the motor was let to rest for at least. . .
This whole paragraph has been revised as recommended by Referee #1. Doing this, the sentence in question has been rephrased.

8-33: The 60s spin-up phase is equivalent to 2-3 reaction time constants. Knowing your 4.0 to 1.5 μA decay times would help identify the appropriate spin-up duration.
Unfortunately, we did not record this information, sorry.

9-1: I0 = 0.13 μA seems very high. Do you mean 0.013 μA? If not, our recommended operating procedure suggests not using sondes with background currents greater than 0.08 A. Change the solutions. Change the cells themselves if necessary.

No, it was really 0.13 μA. In our procedure, we had no absolute criterion on I_0, but instead (and in addition to the decay test) we checked if $(I - I_0) \times t_p$ matched within 5% the result obtained with a reference cell in the same experimental conditions (especially, same tuning of the ozonizer – $I \approx 5\mu$A). Anyway, Figure 7 (8 in the AMTD version) provides evidence that the sonde worked fine.

Note however, that from the data of this experiment, a better adjustment with the UV analyzer data could be obtained using ad-hoc values of 0.07 μA for I_0, and 33 s for the pump time – instead of the measured values of 0.13 μA and 32 s, respectively, as we used to retrieve the ozone data in Figure 7. The motivation of using trully measured instead of adjusted values was to investigate the question: what accuracy could be expected without any other ozone reference than the intermittent sonde operated following the standard procedure?

9-20: disequilibrium

We actually changed to "imbalance".

9-26: You say increase with time - but as Figure 9 shows, this is good - its getting closer to 0. It may well go past 0 and the agreement get worse again if time continues to pass. However, I recommend restating this observation, since the magnitude of the deviation is decreasing with time.

Referee #1 made a similar comment. The sentence has been rephrased.

11-16: Change uneasy to difficult.

Changed.

11-20: Change impossibility to inability.

Changed.

11-28-30: You suggest that condensation caused the problem. Do you have any evidence to that effect? Dew point temperatures versus temperature? Observations of clouds? Sondes do not seem to have trouble with most clouds. Is this a problem with a low cloud or fog that would affect the ozonesonde measurements differently than a typical cloud?

The balloon carried a humidity sonde at its north pole. During the period with spurious ozone data, the balloon altitude dropped by about 100 m (visible in Figure 11b), suggesting it was weighted. In the same time, the observed relative humidity rose above 80%. Then, humidity dropped below 50% after 06 UTC when ozone concentrations came again to more expected values. All those elements suggest that water condensation might cause the problem, but we have no definitive evidence of this. All this has been now specified in the text.

12-5: . . .Ersa surface station agrees fairly well by the end. . .

Amended.

12-15: I wouldnt use the word proved. At best, your observations demonstrate an ability to provide ambient ozone mole fractions. . .with an accuracy of about 10%.

The sentence has been rephrased.

12-21ff: You compare your observations with a campaign in the North Atlantic off New York City (Mao et al., 2006). Would you expect these to have similar results?

How do the currents, water temperatures, winds, convection, etc. compare between these two sites, and how do those factors influence the results? In other words, why should we expect the Mediterranean to look like the Atlantic?

The Referee is right, the differences between the explored environments should be more emphasized. Few sentences have been added to this paragraph.

13-4: Have you looked at potential temperature in addition to specific humidity?

During intervals with constant specific humidity, potential temperature and equivalent potential temperature were found to be fairly constant as well. A mention has been inserted.

13-27: . . .to conclude in situ ozone production.

Amended.

13-31-32: This is presumably the first time that ozone production is evidenced in the free troposphere from direct observation. See the paper from NASA-TC4 data by Mor- ris et al. (2010) on ozone production from a dissipating tropical convective cell in which they report 6 – 12 ppb/hr in the lower free troposphere. While the mechanism in that paper is lightning NOx production, it does provide a quasi-Langrangian analysis using an ozonesonde that oscillated between 2-5 km over a 90-minute period. Your study would be the first with an intentionally designed instrument to sample in a Lagrangian fashion.

We thank the Referee for his comment and this interesting reference. Morris et al. report a very nice and unique opportunity to estimate ozone enhancement in the free troposphere from ozonesonde in situ measurements in an air mass downwind of a storm cell (most likely in relation to lightning NOx production in this case). These authors do not actually claim the Lagrangian character of their observation, but in contrary, they investigate very carefully the question of gradient advection as contribution to the observed ozone changes (no contribution at all from advected gradients would be found in case of pure Lagrangian evolution). It turns out from their analysis that the sonde sampled the same horizontally-homogeneous air mass while crossing it vertically five times, although vertical wind shear and rotation make the sonde trajectory not strictly Lagrangian. They also show that advection of a vertical ozone gradient contributed up to 30% of the observed ozone change in the air mass.

Despite the non-Lagrangian nature of the sonde motion, this study is very nice and quite convincing on the fact that Lagrangian chemistry account at least partly for the observed ozone evolution. The sentence has been rephrased as suggested by the Referee, and a citation of the work by Morris et al. has been added in the text.

14-5: . . .allows us to clearly distinguish. . .

Amended.

14-12-14: This discussion about humidity and ozone-humidity relationships reads as though it is universally true. In particular, the comment that higher ozone concentrations are therefore expected in the free troposphere than in the boundary layer, *certainly relates to the local conditions for this flight and is not true generally. Just rephrase these statements to clarify your intended meaning.*

The discussion has been rephrased to sound more specific.

14-13: I would avoid the word global when you are talking about a general characteristic of your particular data set.

"Overall" has been used instead.

14-14: . . .section 1, and no chemical evolution. . .

Amended.

14-18 and 21: delete the word here

Done.

14-27: clarify that this is a positive temperature change: +4°C

Done.

15-3: . . .likely owing to water condensation weighting the balloon. The Morris et al. (2010) paper makes a case for this mechanism, too. But if you do, what is your evidence? Again, having the temperature and dew point temperature data should tell you whether this is indeed the case. If you cant demonstrate that explanation, I think you should just delete the sentence.

This statement was mostly speculative. As the cause for the balloon descent is of little importance in the discussion, the statement has been removed.

15-6: I would mark this discussion of the model results as a new section of Model results. One question I had at the end of this section, does the model predict/show the 4 different air regimes youve identified in Figure 14?

We acknowledge that the text in Section 4.4.1 (focused on balloon flight B62) is quite long. Unfortunately, a fourth level of sectionning is not allowed in AMT, and the presented model simulation is specific to flight B62. We thus prefer to keep all the discussion on B62 in a single section.

Beyond the fact that the model wind-field and the ozone (relative) evolution are mostly consistent with the balloon trajectory and ozone observation, we do not expect that such a relatively coarse model (horizontal resolution of about 20 km, and 40 m at best in the vertical) is able to capture such fine details as sharp boundary-layer/free troposphere interface, gap flows, hydraulic jumps, etc. For this reason, we do not go further in the model vs. observation comparison.

15-26: I think photochemical production is better.

Amended.

16-1ff: It is inadvisable to use HYSPLIT in a single parcel mode to evaluate air parcel histories. You should initialize a matrix around the starting lat/lon and run at several altitudes to verify that the trajectories hold together. The spread of the matrix will give you some indication of the reliability of the trajectory calculations and sensitivity to the initial conditions.

We performed a new HYSPLIT simulation with the ensemble option, generating a bunch of 27 backtrajectories. See our response above (in section "General comments and changes").

16-14: Delete in contrast.
Done.

16-19: I think photochemical production is better.
Amended.

17-17-18: The few other data sets available from. . .measurements) suitable for in-flight validation all show reasonable agreement. . .
Amended.

17-32: Same comment about the first observation of ozone photochemistry in the free troposphere – see Morris et al. (2010).
Rephrased. We now use the term "continous Lagrangian trajectory" to be more specific.

Appendices: While nice, I think most of Appendix A information could be subsumed into a reference (or series of references). The background current info is interesting, though.
This was among the major concerns of Referee #1. The appendices have been removed and the article main text revised. Please see details in our response to Referee #1.

22-22-23: The fact that the I0 value was adjusted so that the sonde matched the ozone analyzer might not have been the right thing to do. How different were these values, typically, before the adjustment?
The differences were generally within the range ± 5 ppbv before the adjustment (except +11 ppbv for one radiosounding concurrent with BLPB flight B53, see Fig.10a in the revised manuscript).

It is established that the uncertainty on I_0 value accounts for most of the uncertainty in the troposphere, which amounts to $\pm 10\%$. Even though it is not usual within the ozonesonde community, we actually see no fundamental objection to such adjustments (offseting data to adjust the zero value of an instrument is a common procedure in experimental sciences). Fig.10 shows comparisons of ozone vertical profiles from ECC sondes launched in the same time. The profiles match very well after adjustment but look parallel before, suggesting that using I_0 values measured during the final preparation (as recommended in the GAW procedure and usually done) would obviously bias our experimental results.

28-Table 3: I know that there are different recommendations that depend also on the solution type. Not sure about Z sondes. Just asking the authors to verify they are using the best correction factors for the sonde type and solution type. I realize that the solution type should have nothing to do with the pump efficiency, but the WMO group *noted that theres some offsetting of errors that happens, so these pump corrections do more than just correct the pump efficiency.*

The Referee is right (he certainly refers to the study by Johnson et al., JGR, 2002, doi:10.1029/2001JD000557). But actually, following the recommendation from the other Referee (#1), we actually got rid of the table specifying the used pump flow correction factors, as well as any discussion on this topic. The reason is that such a correction is important at high altitude (typically in the stratosphere) but negligible in the low troposphere as in the present work (less than 0.3% below the pressure level 650 hPa).

Figure 5: Difficult to see the reference sonde data. Change the scale? Plot differences between reference and experimental sonde vs. UV analyzer?

Referee #1 made a similar comment, and consequently, Figure 5 of the AMTD paper has been replaced by 3 panels (b-d) in Figure 4 in the revised manuscript. The new figure allows for clear comparisons between the three data sets.

Figure 7: Perhaps show the raw measurements, too, rather than % variation?

A new figure panel showing the volumetric flow rates as a function of time has been added in Figure 6 of the revised manuscript.

Figure 8: A -3.3 ppbv bias suggests to me that the background current used is too high. Thoughts?

Yes, the bias is certainly due in most part to the uncertainty on I_0. A short sentence has been added in the text. See also our response to the Referee's comment 9-1 above.

Figure 9: Looking at this figure got me thinking to ask how does the pump motor current change with time? How does that affect the flow rate and hence, the agreement of the measurement?

In the sonde electronic system developed for BLPB flights, the pump motor current was not among the measured variables. Nevetherles this variable was measured during the experiment presented in Section 3.1.2. See our response and the figure presented above in section "General comments and changes".

Figure 14: I think this comes across in the text, but looking at just this figure, region 1 is above the nocturnal boundary layer, region 2 is in the boundary layer, region 3 I sin the lower free troposphere, and region 4 is in the boundary layer again. Right?

Yes, all this comes across in the text. Note that region 1 is likely in a transition layer between the boundary layer and the free troposphere (as explained in the text).

Figure 15c: Might you also so this as a scatter plot of model versus measured ozone? That could be interesting to see how well the model picks up the variability. . .

We do not think that a scatter plot would provide more information than Figure 14c (15c in the AMTD version). As mentionned in our response to the Referee's comment 15-6, we prefer not to go beyond the conclusions presented in the text, and Figure 14c is sufficient to this goal.

Reference:

Morris, G. A., A. M. Thompson, K. E. Pickering, S. Chen, E. J. Bucsela, and P. A. Kucera (2010), Observations of ozone production in a dissipating tropical convective cell during TC4, Atmos. Chem. Phys., 10(22), 11189-11208, doi:10.5194/acp-10-11189-2010.

Franois Gheusi, on behalf of the coautors.