This manuscript presents some rare airborne RO\textsubscript{2} measurements over much of Europe. These are useful measurements that should be shared, especially the vertical dependence of concentrations, but the analysis is not focused enough and requires significant revisions.

Major comments:

line 101 (in the intro) shows the photostationary state (PSS) equation for [HO\textsubscript{2} + RO\textsubscript{2}], and describes previous studies that compared this value to measured HO\textsubscript{2} + RO\textsubscript{2}. The introduction ends with "Consequently, this data set provides an excellent opportunity to gain a deeper insight into the source and sink reactions of RO\textsuperscript{2}* and the applicability of the PSS assumption for the different pollution regimes and related weather conditions in the free troposphere".

I was looking forward to seeing what insights the authors had to provide regarding the applicability of the PSS assumption.... but it does not show up at all later in the main text! The authors make use of the equation P(RO\textsuperscript{2}*) = L(RO\textsuperscript{2}*) quite a bit, but not the above NO-NO\textsubscript{2}-O\textsubscript{3}-RO\textsubscript{2} photostationary state assumption.

Line 119: in the abstract RO\textsuperscript{2}* is defined as RO2 + HO2. On line 119, it's defined as RO2 + HO2 + OH. Quantitatively there's little difference since [OH] is much smaller than the other two terms, but conceptually this is very important. Line 24 states "RO2* is primarily produced following the photolysis of ozone (O3), formaldehyde (HCHO), glyoxal (CHOCHO), and nitrous acid (HONO) in the airmasses investigated", which is true for
(RO₂+HO₂+OH) but not for (RO₂+HO₂). Please be consistent in terminology.

later in abstract: “The dominant removal processes of RO₂* in the airmasses measured up to 2000 m are the loss of OH and RO through the reaction with NOₓ during the radical interconversion”. This is very confusing – if a reaction is a radical interconversion reaction, then no radicals are lost. Moreover, reactions of RO with NOₓ are rare and not discussed at all later in the manuscript.

It appears that one of the main challenges the authors are facing when interpreting their dataset is that their instrument is much less sensitive to CH₃O₂ than HO₂, and so in the limit that all RO₂ are CH₃O₂, then RO₂* = HO₂ + 0.65 × RO₂. This relates to the high NO mixing ratios used in their instrument, as commented on by both reviewers for George et al. 2020. This limits how much can be gleaned from the comparison of calculated RO₂* with measured RO₂*, since a priori the CH₃O₂/RO₂* ratio is not known.

line 231: there is a major error in Equation 2. The penultimate term – the sum of OH + VOC reactions – should not be part of this equation as it reflects radical cycling rather than a primary radical source.

Figures 8-12 and accompanying text: These show variations of the same plot (RO₂ measured vs RO₂ calculated). I would recommend keeping just 2 of these and moving the rest to the SI as not much is revealed by the 3rd – 5th set of plots. This would help shorten the paper and improve the readability.

Additional comments:

Line 40 – remove comma

Line 90 – should say “R23 and R25 are two of the most…”

Section 3: the description of perceas is confusing. Nowhere does it even mention that the sampled air is mixed with NO and CO in amplification mode – the basics of PERCA operation. I do recognize that the instrument has been described in the referenced papers, but just a few more details would be helpful.

line 174: the relative sensitivity to CH₃O₂ vs. HO₂ (α) – was this just based on their
previous study, or was it experimentally determined again in between flights? Similarly, please provide more information on calibrations – how many were done? Were the eCL values stable (and their dependence on humidity) or were different values used for each flight?

That’s great that glyoxal and methyl glyoxal, in addition to the other OVOCs, were measured.

Line 202 “Typically, the highest RO2* mixing ratios were observed below 3000 m over Southern Europe. This is attributed to the higher insolation and temperatures favouring the rapid production of RO2* from the photochemical oxidations of CO and VOCs”

I question the inclusion of temperature in that sentence. If the authors are simply presenting a *correlation* between highest RO2 mixing ratios and temperature that is fine, but to *attribute* the high mixing ratios to elevated temperature requires some discussion. Are they inferring that the reaction rate constants are faster at higher temperatures? This is certainly not true for all of the reactions. Or are they referring to the increased emissions of biogenic VOC emissions at higher temperatures, leading to higher bVOC concentrations? This by itself won’t necessarily lead to higher RO2* mixing ratios.

For HONO photolysis, do the calculated numbers reflect the gross OH formation from HONO photolysis or the net amount (subtracting out the reverse reaction OH + NO)?

line 244 and 304: “calculated”, not “estimated”

Figure 4: I would have found it more useful to see a plot of altitude vs. P(RO2*) rather than [RO2*] colored by P(RO2*).

line 266- needs some re-wording. “...the high amount of H2O in the air masses probed results in the O3 photolysis and subsequent reaction of O1D with H2O (R1-R2a) and is the highest RO2* radical production rate”. The H2O itself does not cause O3 photolysis... rather the high H2O leads to the reaction O(1D) + H2O being the most important RO2* source.

line 305: The sentence “The [RO2*] < 0.5....” is awkward, change to something like “Measurements in which [RO2*] were less than xyz...”

Figure 7 and text: it’s good that someone has done this analysis! I think the correlations
observed in figure 8 are about as good as could be expected, though it’s interesting that the impact of NOx is not so clear.

line 338: “The second solution gives...” I don’t see any solutions.... please clarify.

line 340 – “the measured RO2* (RO2*m) mixing ratio” is confusing – what is RO2*(RO2*m)? Is this a product of RO2* and RO2*m? Or should it just be RO2*m?

line 341: “RO2*m and RO2*c are the measured and calculated RO2* respectively for d = 1, i.e. RO2* = HO2 and d = 0.5, i.e. HO2 = RO2.” Confusing. RO2*m is measured RO2, and RO2*c is calculated, but for which case – d = 1 or 0.5? This section should be prefaced with some text along the lines of “because not all peroxy radicals are detected equally by the instrument, the comparison of measured and calculated RO2* values is complicated. To investigate this, we ...”

eq 11: the first term on the right hand side of the equation refers to the RO2* loss reactions HO2 + HO2, RO2 + RO2, and HO2 + RO2. The 2nd term should represent the RO2* loss reactions RO2 + NO2 and HO2 + NO2 and OH + NO2, but not RO2 + NO or HO2 + NO as those are radical interconversion reactions. I recommend simply writing out the full equation as it is confusing to always deal with “HO2 + RO2” and “NOx” in these rate equations.

line 524: dependence, not dependency