Review of manuscript by H. Petetin et al.
“Characterizing tropospheric O3 and CO around Frankfurt over the 1994-2012 period based on MOZAIC-IAGOS aircraft measurements"

The authors have extensively answered the concerns raised by the reviewers and the manuscript has changed substantially. In doing so, most critical issues were solved and many parts were improved. So far so good.

However, one major flaw I have, namely that the wording is largely not acceptable. It’s not so much the English spelling itself, but that the sentences and the argumentation are partially so awkward that I could decipher what is meant only after reading the relevant parts many times. Often the real information was hided and even sometimes not written. Under the line, I often didn’t catch the information of a section and at the end I couldn’t repeat what I have read, that is the take-home message was missing.

One main reason in my opinion is that you frequently try to condense an argumentation in one convoluted sentence, in which a native speaker would do it in three short sentences and in a logically straight-forward way. Second main reason is the lax and imprecise wording, see the many many examples listed below. Please, read each sentence carefully and think what you have read. Often the sense can only be understood with considerable creativity and own interpretation.

In addition, you steadily give twisted phrases, e.g. “the 1994-2012 period” (also in the title), “the Frankfurt and Munich airports”, … This is slang and inadequate for a scientific paper … and to my knowledge also in French not used. Below, I give more detailed suggestions in the abstract only, but thereafter also many … as improving the wording is not the task of a reviewer. Please work on it! Consider that the impact of a (linguisticly) badly written paper is poor.

I have to say that this was also the reason why the review took so long. After the first reading, I only said “oh gosh” and the fun to work on it was quite limited.

Minor remarks:

P.1, l.12. “vertical profiles of ozone (O3) and carbon monoxide (CO)” instead of “ozone and carbon monoxide vertical profiles”

P.1, l.12. Exchange “at several” with “in three”

P.1, l.12. Exchange “the Frankfurt and Munich airports” with “the German airports Frankfurt and Munich” and improve it later in the text

P.1, l.13. Exchange “the densest dataset in the world (about 96)” with “the worldwide densest vertical in-situ dataset of O3 and CO (with ~96)”

P.1, l.14. “mean vertical profile of ozone“ instead of “mean ozone vertical profile”

P.1, l.15. Skip “vertical”

P.1, l.15. Exchange “kilometre (deposition, titration by NO) during the whole year and close to the tropopause (stratosphere-troposphere exchanges)” with “kilometre (due to dry deposition at ground and titration by NO) during the whole year and close to the tropopause (due to stratosphere-to-troposphere in-mixing)”

P.1, l.17. “mean vertical profile of CO“

P.1, l.17. Skip “in the concentrations”

P.1, l.18. free troposphere

P.1, l.18. Exchange “In terms of seasonal variations, the mean O3 has a minimum in November-December in the whole troposphere, a broad spring/summer maximum …” with “O3 minimizes in November-December, shows a broad spring/summer maximum …”

P.1, l.21. Exchange “mean CO seasonal profile” with “seasonal variation (or course) of CO”

P.1, l.22. Exchange “surface, refined to September-October higher in the troposphere, while maximum concentrations occur” with “surface and in September-October higher in the troposphere, while the maximum occurs”

P.1, l.23. Exchange “the 1994-2012 period” with “the period 1994-2012” and later in the text

P.1, l.23. “O3 has changed insignificantly” instead of “the mean O3 trends are mostly insignificant”

P.1, l.27. all three tropospheric

P.1, l.28. “In contrast, for CO the mean” instead of “Conversely, the mean CO”

P.1, l.29. all three …

P.1, l.32. Exchange “The changes in the O3 seasonal cycle are also investigated, with a focus on the phase. Ozone maxima occur earlier and earlier with a shift around -12.1…” with ”The phase of the seasonal variation of O3 was found to change in the entire troposphere. The O3 maxima moves forward in time with a rate of -12.1…”

P.1, l.32ff. In my opinion, the abstract is too long. You can skip e.g. the first 2-3 lines of p.2

This were improvements for the abstract only.

P.2, l.20. The term “stratosphere-troposphere exchanges” doesn’t exist, “stratosphere-troposphere exchange (STE)” does. Change this everywhere in the text

P.2, l.25. Exchange “ozonesonde long-term observations” with “long-term ozonesonde observations”

P.3, l.8. Exchange “quantify all the terms of the ozone budget” with “quantify all factors influencing the budget of ozone”

P.3, l.11. The sentence "An alternative but more qualitative approach consists in taking benefit from the different seasonal patterns of the various ozone budget terms (e.g., precursors emissions, photochemical production, stratospheric intrusions, transport regimes) and aims at linking the evolution of the ozone seasonality to changes in the contribution of its various sources and sinks." is really awful and exemplary demonstrate my major concern. By the way "ozone budget terms" doesn't exist. Moreover this "alternative" will not work. You can't change and follow one single of the listed process (like in a laboratory) without influencing the other. Thus skip the sentence or change it (strongly).

P.3, l.14. Exchange "Mace Head coastal site" with "coastal site Mac Head"

P.3, l.19. „Derwent et al. (2013) noticed that the still increasing baseline ozone levels do not extend to the European ozone load…" I don't understand this sentence. Do you mean "the increasing baseline ozone levels do not impact the European load of O3? … which I also would not understand.

P.3, l.25. "Such a shift may reflect some changes in the contributions of the various ozone sources and sinks, e.g.,…" O3 is controlled by its sources and sinks. Thus "such a shift does reflect…", but the second half of the sentence then makes little sense.

P.3, l.27. After Parrish et al (2013) a line break. You should use much more line breaks!

P.3, l.32. Skip "measurements (close to precursors emissions and/or deposition sink)"

P.3, l.35. Change "long" to "moderate"

P.4, l.1. Change "interesting" to "powerful"; skip "useful"

P.4, l.4. Skip "used in the paper"

P.4, l.12. Change "since 1994 and 2002" to "since 1994 (O3) and 2002 (CO)"

P.4, l.18. … the precision, but more important what is the accuracy?

P.4, l.27. Skip "briefly"

P.4, l.29. Change "systems operations" with "instrumentation"

P.4, l.32. the airport Frankfurt

P.5, l.7. Change "As tropospheric air masses are subject to very different constraints depending on their altitude (e.g., distance from surface emissions or stratosphere)" with "As tropospheric ozone shows strongly varying sources, sinks and lifetimes with height"

P.5, l.7. Change "The UT is defined here as the 60 hPa-width layer below troppause plus 15 hPa" with "The UT is defined here as the layer having its top at the tropopause plus 15 hPa and spanning a pressure of 60 hPa, that is a layer ~1.6 km thick and starting/ending ~2.1/~0.5 km below the tropopause"

P.5, l.12. Change "Data collected in the 1-2 km layer" with "Data collected below"

P.5, l.12. Change “The p PV=2 parameter is used to determine the DT pressure at the top of the selected tropospheric vertical profile.” with “The pressure at the DT (pPV=2) minus 15 hPa defines the top of the UT applied here.”

P.6, l.4. “For instance, the ozone criteria may give a lower dynamical tropopause (DT) compared to the thermal method (Bethan et al., 1996).” In this sentence sticks some mistakes. First, O3 can define the chemical tropopause (CT), which is in the mid-latitudes mostly close to the DT, but it can’t define the DT”. Secondly, a thermal method doesn’t exist; you mean the thermal tropopause (TT, which is well defined). Thirdly, the CT (and DT) is only on average below the TT, in the mid-latitude over the UK by ~800m, but can also be above (at anticyclonic flow) or well below by up to 4-5 km (at cyclonic flow). Here, read the papers by V. Wirth (Thermal versus dynamical tropopause in upper tropospheric balanced flow anomalies. Quart. J. Roy. Met. Soc., 126, 299-317., and the ones thereafter).

P.7, l.10. … which indicates that the ECMWF-derived 2 PVU doesn’t work for defining the tropopause. ECMWF estimates it far too low. How often this appears. **Later you have to discuss the consequences of such an ill-defined tropopause.** The closer you get to the tropopause, the more questionable your O3 trends etc. get.

P.7, l.19. Skip “by the aircraft”

P.7, l.26. “sometimes referred as the potential emissions sensitivity (PES), that is the potential to catch up emissions from certain regions”

P.8, Figure caption. Can’t combine “Average residence time” with the units given in the picture. You show this figure, **but later it is (at least I believe) not once used, e.g. for interpreting the vertically differing trends etc. Do it!**

P.8, l.13. You show data between ground and 12 km, but not to the tropopause. Correct!

P.8, l.17. What is an O3 abundance? I would say an O3 density (in e.g. O3 molecules per cm^3) … which may decrease with height. Change e.g. to “mixing ratio”

P.8, l.18. You can’t simply write “(dry deposition …)”. “(due to dry deposition at the ground and enhanced titration by NO in the PBL)” would be possible. Please, be precise and clear. This lax wording is terrible. I think you are scientist.

P.9, l.1. No “tropopause”, but “12 km”

P.9, l.1.”(exchanges with the stratospheric reservoir)” … boah … change

P.9, l.3. “very low in winter and to a lesser extent in autumn” … likewise boah, you mean “very low in winter and even more very low in autumn”? … what is lower than very low. Once again, you are scientist! “low” is a poor description anyway, but lower then very low is already quite silly … but what means now “quite” … and thereafter you write “substantially enhanced” another time … boah

P.9, Figure caption. How the standard deviation is inferred, from the individual profiles or daily averaged? Change “overall climatological profile considering all seasons” to “annual mean profile”

P.9, l.10. Start with “To further characterize the variability of O3 and CO above Frankfurt/Munich, we…” In my opinion “daily variability” is a better word than “coefficient of variation”!

P.9, l.21. Change “close to the tropopause” to “In the UT”. Exchange “exchanges” with “in-mixing” … citations are missing

P.10, l.8. “high in altitude”? be precise!

P.10, l.8. “Over the entire tropospheric column, the mean CO mixing ratio reaches 117 ppb.” No clue what you want to say, e.g. “is 117 ppb”, “reaches to 117 ppb”, …?

P.10, l.10. Change “the CO mixing ratios in the first kilometre strongly increase as one moves closer to surface emissions (up to 243 ppb at the surface)” to “At the ground and close to surface emissions, CO maximizes with 243 ppb on the annual average”

P.10, l.13. You write so often “concentrations” which is “molecules cm^-3”, but you mean “mixing ratios” … be precise

P.10, l.15. Of course, CO clearly and significantly does change over the year. But the daily variability exceeds the seasonal amplitude. Change!

P.10, l.15. Also “The seasonal climatological profiles always remain at less than one sigma from the annual climatology” is not correct and the English … boah … this sentence has a (or no) sense like “in the night it is darker than outside” and the rest of the para is likewise not ok

P.10, l.21. “Concerning the CV (Fig. 4), one can see that CO is less variable than O3” … again, what a strange wording. Why not simply ”As shown in Fig. 4, the daily variability (DV) of CO is lower than the one of O3, in particular at altitude above 4 km…”

P.10, l.22. Also formulations like “The annual CV of CO shows values ranging from 44% …” are misleading and makes it so difficult to follow. Simply write “The annually averaged variability of CO ranges from 44%”, but don’t speak of “the annual CV (curriculum vitae) shows values …” In our research field “value” is basically an “item non grata” … it can be everything

P.11, to P.14, bottom. The entire chapter 3.2 is hard to digest. It is ‘only’ a description of ups and downs, fairly boring and at the end I had nothing in my mind, basically didn’t learn anything. I would shorten it and focus and the clearest features.

P.11, l.3. Change “annual and monthly” with “long-term”

P.11, l.9. Skip “previously” and write instead “As noted in section …” Change this at other locations, too!!!

P.11, l.11. Change “in the LT” to “in the entire tropospheric column”

P.11, l.13. Change “in May and August” to “between May and August”

P.12, l.8. “… is related to the 1997 El Niño that have enhanced stratospheric-tropospheric exchanges (Ordóñez et al., 2007)” … STE, but also the claim and cita-

tion are in my opinion not correct. May be in the meantime there are some first and vague speculations about the impact of ENSO on STE.

P.12, l.19. “variation” instead of “variability”

P.13, l.7. “which illustrates the high contribution of the CO background at the hemispheric scale” … makes no sense

P.13, l.9. At the tropopause O3 is longer lived than CO

P.13, l.22. “in section …“ instead of “previously”

P.13, l.23. I thought you skip the ground-based data. So “on tarmac, take-off/landing” should not count

P.13, l.34. “Considering the monthly time series, correlations are improved due to the seasonal variations (from 0.61 to 0.90).” doesn’t work

P.14, l.5. R = -0.21 means no correlation

P.15, Table 1. Add vertical lines after the 2. and 5. row to enhance the visibility. The sense and significance of the different time periods (1994-2012 and 2000-2012) don’t come clear to me. You short motivate this a bit. As written in the introduction, there was a positive O3 trend until ~2000 and thereafter no trend or at certain sites even a weak decrease. The logical way to go would be to analyze the trend between 1994 and ~2004 and between ~2002 and 2012, right? Why you didn’t do it? As written, here you have to motivate and explain your approach.

P.16, l.6. “in all three” instead of “in the three”

P.17, l.4. “Most of the few positive trends found here over the whole period are due to an increase of O3 in the 1990s.” makes no sense to me. Elevated values/trend at the beginning of the considered time period should cause decreasing O3 and a negative trend.

P.17, l.4. “Interestingly, at regional background sites in Europe over the 2-3 last decades, Parrish et al. (2012) highlighted that O3 trends, when they are expressed relatively to the concentration in 2000, are quite similar (around +1% O3,2000 yr -1) whatever the site and the season.” … quite awkward English.

P.17. The entire para / page, without any line break is very difficult to understand. For me there is no structure and even after 3 times reading, I do not see the take-home message.

P.17, l.35. “…This is likely due to the fact that only the troposphere is considered in this present study”. The variability in the lowermost stratosphere (LMS) is much higher and depends strongly on the actual flight route/statistics. You will thus need much more data in the LMS to infer significant trends.

P.18, eq. 1 and theta_month. If I enter theta = 0, I end up at a maximum on 15. March. Or if t=1 corresponds to 1.Jan, then 31.Dec is t=12.97. More logical would be, if t=1 corresponds to 15. Jan (middle of month) and t=12 to 15. Dec

Now 3 examples for the strange (and difficult to follow) type of formulation:

P.20, l.7. “Results show a decrease around 1-2 ppb of the amplitude” instead of simply “The amplitude decreases by 1-2 ppb”

P.20, l.10. “… This is consistent with the fact that O3 has increased significantly in the whole troposphere during the winter (the season of minimum O3) but not during spring/summer (the season of maximum O3)” instead of “Reason for the decreasing amplitude is the significantly increased yearly O3 minimum occurring in winter and to the same time constant O3 maximum occurring in spring/summer”

P.20, l.13. “Concerning the phase of the O3 seasonal cycle, results clearly highlight a shift toward earlier O3 maximum along the 1995-2012 period” instead of simply “The phase of the seasonal variation of O3 was found to shift forward during the period 1995-2012”

P.21, l.14. coastal site Mace Head

P.21, l.16. 3 last years studied

P.21, l.31. “much lower ones in the UT and to a lesser extent in the MT.” … boah

P.21, l.32. … worldwide densest

P.22, l.1. “shift extent” … boah

P.22, l.6. “an unambiguous explanation”. If so, you give some vague hints

P.22, l.11. (e.g., Thouret et al., 2006). Sorry, but there are hundreds of publications which are better suited for STE processes and the springtime maximum of downward stratospheric transport

P.22, l.14. which contradicts our observations

P.22, l.18. our period ?

P.22, l.31. “… based on the potential vorticity extracted from ECMWF meteorological data” No, only for defining the upper border of the UT