

Referee 2:

This paper reports on new and exciting data from the BLUESKY experiment conducted during the early months of the COVID-19 pandemic. The paper is well written and the figures are clear and appropriate. In general I think that the aim of the paper is quite novel, and that the results could be useful for the research community. However, I have reservations about the analysis because the BLUESKY ozone data in 2020 are not lower than the ozone values in the earlier campaigns. In contrast, three new studies show ozone above Europe was anomalously low in 2020. This discrepancy needs to be reconciled before I can recommend the paper for publication.

We would like to thank the referee for the helpful feedback and the time to review our manuscript.

Major comments:

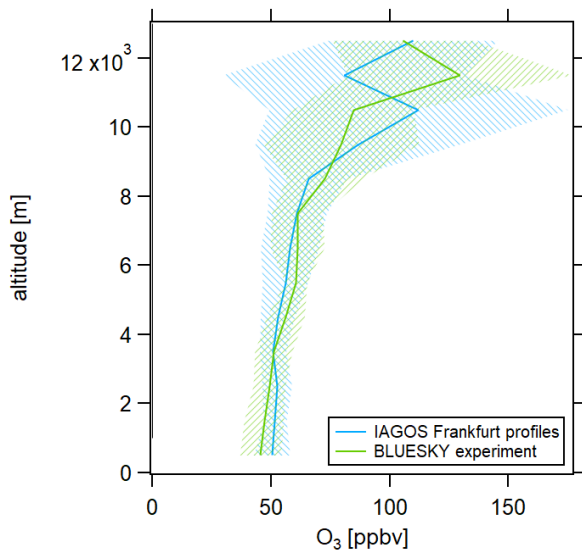
1) Three recent papers have shown a clear decrease of free tropospheric ozone above Europe during 2020, in association with the COVID-19 economic downturn. Two of these papers have been cited [Steinbrecht et al., 2021; Clark et al., 2021]. The third paper is by Chang et al., 2022, and it will appear any day as an accepted paper in AGU Advances (it will be posted here: <https://agupubs.onlinelibrary.wiley.com/toc/2576604x/0/ja>).

We have added this reference to our manuscript.

Lines 77 ff.: Steinbrecht et al. (2021), Chang et al. (2022) and Bouarar et al. (2021) reported decreases in O₃ concentrations in the free troposphere based on in situ observations and modeling studies in the northern hemisphere.

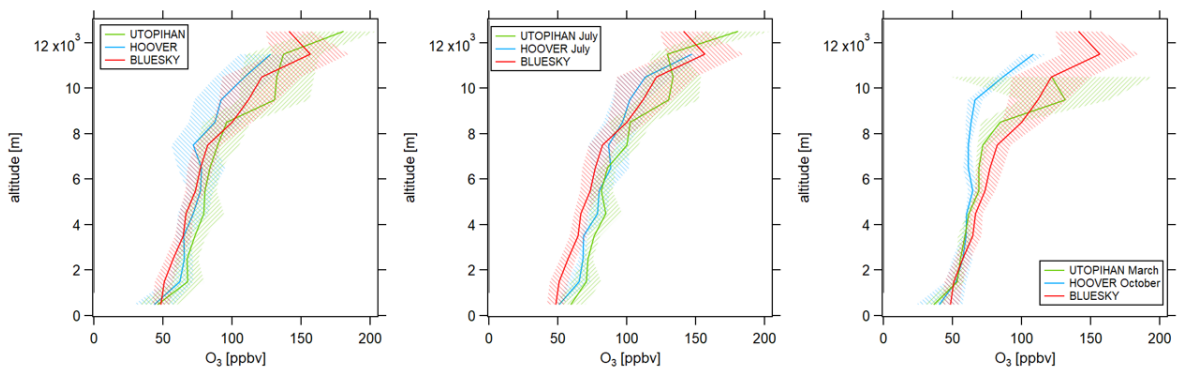
Given that 2020 was an anomalously low year for ozone, it is very puzzling as to why the BLUESKY ozone observations are not lower than the UTOPIHAN or HOOVER data. One possibility is that the sample size of these datasets is too low to provide an accurate estimate of monthly or seasonal mean ozone. Three papers have looked at the sample size necessary to quantify monthly mean ozone above Europe and determined that 12-20 profiles are necessary [Logan et al., 1999; Saunois et al., 2012; Chang et al., 2020]. Given that the IAGOS program has dozens of profiles per month from Frankfurt, you could compare your monthly mean profiles to those from IAGOS. The IAGOS monthly means will be accurate due to the high number of profiles and you can then determine if the aircraft campaign data are biased high or low.

Thank you for this helpful suggestion. We have analyzed the IAGOS ascending and descending profiles over Frankfurt during the BLUESKY campaign and found a very good agreement with our measured O₃. Please note that the measured O₃ data were filtered for the modeled tropopause pressure and the IAGOS data were filtered for 250ppbv O₃ to match the vertical profile above ~10km.

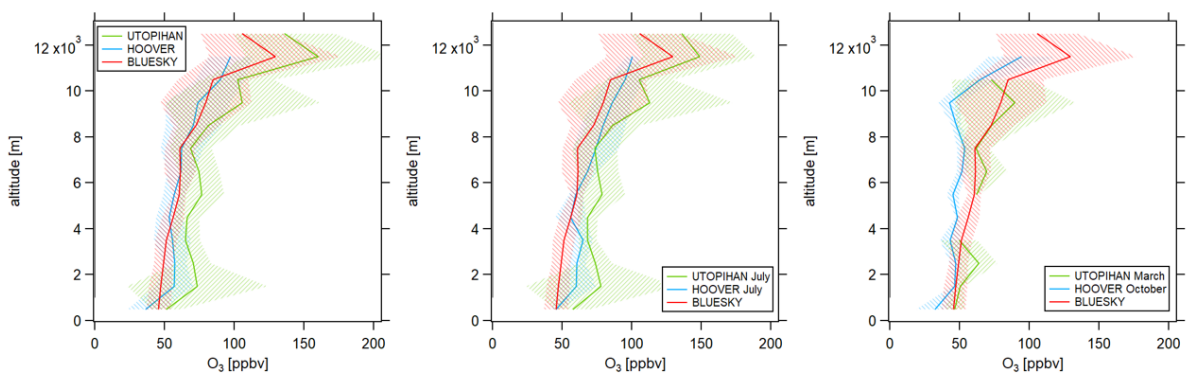


The BLUESKY campaign took place in May/June 2020. In comparison, the HOOVER campaign was partly carried out in October 2006 and in July 2007. Similarly, the UTOPIHAN campaign took place in June/July 2003 and March 2004. Due to the seasonality of ozone, being highest during summer months and lowest during winter months, the presented graphs do not show the lower O₃ values in 2020 described in the studies mentioned by the referee. The following graphs show the vertical profiles of modeled O₃ and measured O₃, separated into different seasons for the HOOVER and the UTOPIHAN campaign. When comparing just the summer data (center), we also observe lower O₃ values, both for the model and the measured data.

Modeled O₃



Measured O₃



We present the measured and modeled vertical O₃ profiles showing the seasonality in Figure S6 of the Supplement and have added text to the manuscript for explanation.

Lines 248 ff.: While ozone concentrations are dependent on various effects such as precursor levels (including NO_x and VOCs) or meteorology, seasonal variations with a maximum around summer time and a minimum during winter months are also of importance (Logan et al., 1985). The here shown campaigns include different seasons: the HOOVER campaigns took place in October and July and the UTOPIHAN campaigns include data from July and March. Figure S6 of the Supplement shows the vertical profiles of ozone separated into different seasons, for both modeled and measured data. Comparing late spring / early summer data of the three field campaigns reveals that O₃ levels during BLUESKY were lower compared to HOOVER and UTOPIHAN which is in line with findings from Clark et al. (2021), Chang et al. (2022), Bouarar et al. (2021) and Miyazaki et al. (2021).

2) Given that the three studies mentioned above report anomalously low ozone above Europe in 2020, we can conclude that net ozone production was below average in 2020, which matches the findings of Miyazaki et al., 2021. However, your conclusion seems to be that net ozone production was not unusual in 2020. How can you reconcile these different conclusions?

We would like to emphasize that net ozone production predominantly depends on the concentrations of O₃, NO₂ and NO, whereas ozone production occurs via NO₂ photolysis and ozone loss is mostly represented by the reaction of O₃ with NO. All three species were found to be lower in 2020 and therefore the impact on net ozone production depends on whether the decline in the production or the loss term prevails. Miyazaki et al., 2021 reports decreases in O₃ across all of the troposphere which is in line with our findings, but to our understanding does not present any observations regarding net ozone production rates.

The goal of our study is to investigate ozone production and the chemical regime during the COVID-19 lockdown, where the central tool is the comparison of the lockdown to a modeled no-lockdown / business-as-usual scenario. We conclude that while we observe both lower ozone production and lower ozone loss during the lockdown compared to a modeled no-lockdown scenario, the net production is not impacted (and only the O₃ cycling is slowed down). For drawing any conclusions on how the lockdown changed net ozone production, we prefer to avoid a direct comparison of the BLUESKY campaign with the earlier campaigns HOOVER and UTOPIHAN as many more factors such as meteorology, seasonality or year-to-year decline of pollutants due to legislation contribute to this development, which we cannot quantify.

3) Bouarar et al., 2021 concluded:

“Zonally averaged ozone in the free troposphere during Northern Hemisphere spring and summer is found to be 5%–15% lower than 19-yr climatological values, in good agreement with observations. About one third of this anomaly is attributed to the reduction scenario of air traffic during the pandemic”. As conclusion that the reduction of aircraft emissions impacted ozone in 2020 has already been published, you should specifically mention this finding in your paper. It would also be helpful to explain how aircraft emissions have strongly increased over the past 20 years [Lee et al., 2021].

We agree with the referee. We have included the references and some text to the manuscript for pointing out the findings of the mentioned studies.

Lines 77 ff.: Bouarar et al. (2021) found that reduced air traffic - a unique incidence after strongly increasing aircraft activities over the past decades as shown by Lee et al. (2021) - can explain around a third of the observed O₃ decrease in 2020, the remaining contributions coming from ground-level reductions and meteorological differences.

4) *I don't agree with this statement in the Conclusions:*

"We encourage future studies to investigate governing chemistry in the upper troposphere, a topic which has not received much attention in literature so far"

I know of many measurement and modelling studies of the chemistry of the upper troposphere, and a few that immediately come to mind are: Barth et al., 2021; Brunner et al., 1998,2001; Cooper et al., 2006; Huntrieser et al., 2002; Li et al., 2001,2005; Ridley et al., 1994.

If your comment is meant to refer to a specific chemical process in the upper troposphere, please make that point clear.

We regret that we were unclear with this statement. We meant to refer to the investigation of the dominating chemical regime in the upper troposphere, which to our knowledge has not been comprehensively explored. We have clarified this in the text.

Lines 364 ff.: We encourage future studies to investigate the dominating chemical regime in the upper troposphere, a topic which has not received much attention in literature so far (...).

Minor comments:

First line of the Abstract: lead should be led

Thank you, we have corrected that.

Lines 2 ff.: The COVID-19 (Coronavirus disease 2019) European lockdowns have led to a significant reduction in the emissions of primary pollutants such as NO (nitric oxide) and NO₂ (nitrogen dioxide).

Line 28-29

This line mentions ozone impacts on humans, animals and plants

"NO_x directly impacts the production of tropospheric ozone (O₃) which is a hazard to human, animal and plant health (Nuvolone et al., 2018)."

However, the reference only deals with impacts on humans. A good reference for the impact of ozone on plants is Mills et al., 2018. I do not know of any authoritative papers that report ozone impacts on animals. If the authors know of such a paper they need to cite it, otherwise, impacts on animals should not be stated as there seems to be no convincing evidence.

We agree with the referee. We have added the suggested literature on ozone effects on plant health and have deleted the statement on animal health.

Lines 28 ff.: NO_x directly impacts the production of tropospheric ozone (O₃) which is a hazard to human and plant health (Nuvolone et al., 2018, Mills et al., 2018).

Line 76

When reviewing studies that indicate ozone reduction in the free troposphere, you should also mention two recent studies that show ozone reductions at high elevation sites within the European boundary: Cristofanelli et al., 2021; WMO Air Quality and Climate Bulletin, 2021.

We have added these references and included them in the main text.

Lines 82 ff.: Cristofanelli et al. (2021) reported lower O₃ concentrations above the PBL in 2020 compared to the 1996 - 2019 average at Mount Cimone in Italy which is in line with findings by the World Meteorological Organization (2021), extended to include two mountain sites in Germany.

Line 166

“what” should be “which”

We have corrected this in the manuscript.

Lines 178 ff.: As a full set of in situ observations necessary for a regime analysis and calculating net ozone production rates, which includes (...)

Line 192

“trend” is not the right word as it refers to a change with time. Would “gradient” work better?

Yes, we agree with the referee. We have replaced the word ‘trend’ with ‘gradient’.

Lines 207: Figure 2f shows the vertical profiles of CH₄ which did not show any particular gradient with altitude.

References

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