Dear Michael Prather,
we thank you very much for your in-depth review of our manuscript egusphere-2024-324. Please find our replies to your comments below. Your original comments are repeated in italics and our replies in normal font.

The open review process under ACP is a great opportunity to have a fair and public discussion of the core element of this paper: the concept of tagging of chemical species like O3 that has been developed by Volker Grewe and his colleagues. First, in terms of review, this paper does an excellent job of calculating the global impacts of three different SSP scenarios with the MESSY model. That alone wit a little more documentation of the current model is publishable.

Reply: Thanks for the positive comment. According to the comments from referee #1 and referee #3 we added more documentation to the manuscript.

Where I have a problem is with the tagging methodology. I had to re-read the core Mertens 2020 paper (Atmos. Chem. Phys., 20, 7843–7873) to try again to understand why one would want tagging versus sensitivity studies. At the time, that 2020 paper had some difficulty in convincing the reviewers of the usefulness of tagging for a chemical system in which there are many feedbacks (as for CH4 and O3). The Mertens Table 1 helped explain the difference between sensitivity and tagging, but it did not give me confidence in the usefulness of the tagging "attribution" value. I am not sure that 100% of the O3 in the troposphere must be attributed to something.

Reply: First of all, we appreciate the positive feedback on our attempts to explain the differences in the methods in the respective Table 1 of Mertens et al. (2020). In addition to that, our point of view is as follows: all ozone in the atmosphere does have a source i.e., it is produced chemically by photolysis of oxygen and by photo-chemistry of other ozone precursors. That implies that 100 % of the calculated ozone has a specific source. The tagging approach decomposes ozone quantitatively, relating it to the shares of the different ozone precursor emission sectors and/or regions. Similar tagging approaches have been developed and used by various groups for a long time. Some examples are given here:

- Horowitz and Jacob (1999): NO_x Tagging
- Lelieveld and Dentener (2000): Labelling technique for NO_x and O_3.
- Emmons et al. (2010): Ozone tagging mechanism for MOZART
- Butler et al. (2018): TOAST 1.0 Ozone Tagging Mechanism

All of them are based on (partly) different assumptions (see for example detailed discussion by Butler et al., 2018, 2020), but all of them have the same goal: to explain 100 % of ozone for a given chemical state of the atmosphere. The latter is in contrast to the perturbation method, which targets on explaining **changes**

of ozone under perturbation of its precursors. These are two different, but complementary aspects.

If this is a misreading, please let me know. My point of view is that given the indistinguishable nature of O3 molecules – from whichever source – labeling such a molecule is simply not useful. The essence of any calculation for policy options should be simply what happens if a policy is invoked. For that purpose, I can understand how sensitivity runs give the correct answer, but tagging may or may not.

Reply: Here, we do not claim that the tagging method provides the only information for policy makers (see Mertens et al., 2020, Table 1), and we agree that this method is not primarily meant for this. However, there are also disadvantages of the perturbation method, since the timing of a sequence of policy measures might largely impact their environmental impact (Grewe et al., 2012). In addition, the perturbation method is not well suited to asses the share of one specific emission source to ozone by turning the emissions on/off because of the non-linerity of the ozone chemistryEmmons et al. (2010); Grewe et al. (2017); Mertens et al. (2018). Therefore, in our study we apply both methods, the source apportionment method to (scientifically) understand the share of different emission sectors in ozone for a given emission scenario. With the further sensitivity (perturbation) simulations, we assess if and how ozone and the ozone shares change, and what the implications for policy making are.

As a lesson, one can look at the idea of labeling/tagging CH4. If one emits a Tg of CH4 and colors it uniquely, we find it decays with the lifetime timescale (e.g., 9 yr), but if we model CH4 as a whole, we find that 99.5% of that Tg perturbation decays with the perturbation time scale (e.g., 12 yr). Well, our colored/tagged CH4 does decay in 9 yr because the perturbation to OH is small, but the remaining atmospheric methane responds to the added Tg and alters the abundance of the untagged CH4, so as to make the overall perturbation decay in 12 yr. Thus, the colored/tagged CH4 does not represent the system behavior, nor even the attributable response to the emission. This example is one of the fundamentals of atmospheric chemistry that we had to develop to "explain" the seemingly nonsensical behavior of a small CH4 perturbation, and it is why a linear attribution that sums to 100% is troublesome to me. For O3, the situation is the same, but in opposite sense. With the ATom data (Deconstruction of tropospheric chemical reactivity using aircraft measurements: the Atmospheric Tomography Mission (ATom) data, Earth Syst. Sci. Data, 15, 3299–3349, doi:10.5194/essd-15-3299-2023), we showed that increases in O3 result in significant drops in production (log sensitivity = -0.4) in addition to increased loss (log sensitivity 1). Thus, an O3 perturbation reduces net P-L for all tropospheric O3 and the perturbation decays much faster than expected (opposite to CH4). We recently showed that the impact of the stratosphere-troposphere exchange (STE) flux of O3 was much less than expected because of these chemical feedbacks (2024. Lifetimes and timescales of tropospheric ozone. Elem. Sci.

Anth., 12: 1. doi: 10.1525/elementa.2023.00112).

Reply: We happily agree to the sentence: "why a linear attribution that sums to 100% is troublesome to me". The point that bothers us is the thinking of a linear attribution. Here we use a highly non-linear technique, as also done e.g. in Emmons et al. (2010) and Butler et al. (2018), though differently. This non-linear decomposition is described in Grewe (2013). In Section 5 of that paper (Comparison of diagnostical methods) a simple non-linear differential equation is given. If we set alpha=1.5 in that equation, we can obtain a lifetime of 8 years and a perturbation lifetime of 12 years for that arbitrary species. Hence, any, perturbation decay then with a lifetime of 12 years, while the unperturbed lifetime is still 8 years. And we can analyze the transient lifetime of the perturbed situation analytically. We should not confuse these two approaches: An analysis of a current state and changes due to e.g. emission changes. We think the misunderstanding here arises from the thought of a linearisation that might be used in an attribution method, which is not the case here. We further think that in the comment the two approaches are indeed confused, which is indicated by the sentence *"... or even the attributable response to the emission .."*: We think that this might be the key to common misunderstandings: we never claimed that the "colored shares" provide any information about the systems "response characteristics"! Moreover, we do not apply any upscaling of perturbation results to 100%. And last, but not least, an "increase" *"... we showed that increases in O3 ..."* is the result of a "perturbation", but this is not what we do want to assess with the tagging method!

In particular, the use of tagged O3S tracers for attributing the role of STE in tropospheric O3 is found to be mistaken because the simple, linear loss does not include the reduced production for O3 of tropospheric 'origin'. The idea that an O3 molecule has an origin is flawed. The O3S tagged tracer is typically 30-40% of tropospheric O3, but the perturbation to tropospheric O3 caused by the total STE O3 flux is only about 8%.

Reply: Again, we do not claim that our ozone decomposition by tagging is any measure for ozone perturbations. With our tagging approach we do not tag processes or perturbations, but we attribute ozone shares to their precursor sources by taking into account all non-linearities (feedbacks and non-linear chemistry) between precursor emission and ozone production. Thus, O3S is not a measure for the perturbation that stratospheric ozone exerts on tropospheric ozone, but it is simply the share of ozone that has been produced originally in the stratosphere. The contribution of our tagged ozone from stratospheric origin (i.e., all ozone produced by photolysis of oxygen) is around 5 - 20 % in the lower and middle troposphere (see Grewe et al., 2017).

The authors are very worried about the non-linear O3-NOx relationship, but that is exaggerated since most of the results here are far from the pollution centers where the NOx-limited vs VOC-limited issues are fought. One of the

Mertens 2020 reviews notes that "the response of ozone to perturbation of precursor emissions in remote regions has been shown to be approximately linear," which I believe is true. Production of O3 is almost linear in NOx over the oceans as found in ATom. What we have globally for O3 and CH4 are chemical feedbacks caused by the non-linearity of chemistry – specifically, the reaction of two species always has 2nd-order Taylor expansion terms that produce a Jacobian with off-diagonal elements, which give us timescales that differ from lifetimes and indirect greenhouse gases (i.e., NOx and CO alter the CH4 timescale).

Reply: And just a side comment, we are not worried about non-linearities. It's the core of the fascinating atmospheric chemistry. Figure 10 clearly shows the non-linear behaviour, if emissions are reduced. See also the comment from referee#1.

My review is not intended to prevent publication of this manuscript in ACP, but I would like to be able to understand how tagging helps us understand how to alter emissions to produce a better result in such a coupled world.

Reply: This comment is very much appreciated and an excellent example of the online discussion forum. The short answer with which we think that we might be able to point at a misunderstanding is twofold:

1. The tagging mechanism is non-linear.
2. The tagging method does not provide information about the effect of altered (i.e., perturbed) emissions and we never claimed that it does.

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