## **Reply to Zosia Staniaszek**

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We thank Zosia Staniaszek for her community comment. Below, we repeat each comment (in blue) and address it (in black). Changes of the manuscript are written in italics.

Thanks for a really interesting read and an important step in developing more models with methane emissions. The section
on tagging the ozone response is particularly novel in a methane emissions driven system and really illustrates the wide ranging impact of changes in methane (that are often excluded by using a lower boundary condition).
Thank you for the positive assessment of our study.

The increase in mixing ratio above the level of increase in emissions is as expected due to the chemical feedbacks in the
system and the lengthening of methane lifetime, albeit here the feedback factor is higher than most models (1.73 as a crude estimate using delta conc/delta emissions (Holmes et al 2018)). I would be interested in whether you could calculate a feedback factor for EMAC using these simulations. Also, a note that more recent feedback factors than those quoted here can be found in Sand et al 2023 Supplementary Table 2 (https://www.nature.com/articles/s43247-023-00857-8).

15 Thank you for pointing us to the study of Sand et al., 2023. We added the reference to the introduction. The respective sentence in line 56 reads now: *Estimates of f are in the range of 1.19 to 1.55 (Fiore et al., 2009; Voulgarakis et al., 2013; Stevenson et al., 2013; Thornhill et al., 2021b; Stevenson et al., 2020; Sand et al., 2023)* 

We calculated the feedback factor using Eq. 12 of Holmes (2018), and also using a curve fit of the spin-up of ERFCH<sub>4</sub> (see below). The feedback factor of 1.55 is most likely not representative for smaller methane (CH<sub>4</sub>) perturbations or smaller CH<sub>4</sub> burden, which might explain why it is at the larger end of previous estimates. We added the following to Sect. 3.2..

## Add in line 399:

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We derive the feedback factor f (see Eq. 1) from ERFCH<sub>4</sub> using two approaches. Firstly, it is calculated from Eq. 12 by Holmes (2018) as  $f = \frac{ln(m_1/m_0)}{ln(E_1/E_0)}$ . Secondly, it is derived from a curve fit of the function  $m(t) = m_0 * [2.75^f + (2.75 - 2.75^f) * (2.75^f + (2.75 - 2.75^f)) * (2.75^f + (2.75^f + (2.75^f + 2.75^f))) * (2.75^f + (2.75^f + 2.75^f + 2.75^f)) * (2.75^f + 2.75^f + 2.75^f + 2.75^f)) * (2.75^f + 2.75^f + 2.75^f + 2.75^f + 2.75^f + 2.75^f)) * (2.75^f + 2.75^f + 2$ 

25  $exp(-t/(f * \tau)]$  (Holmes, 2018) of the spin-up of the atmospheric mass of CH<sub>4</sub> using the yearly mean CH<sub>4</sub> lifetime with respect to OH oxidation for  $\tau$  (see Fig. S14). Both approaches suggest f = 1.55. However, the derived f is not expected to be representative for CH<sub>4</sub> perturbations of EMAC close to present-day conditions because f increases with increasing CH<sub>4</sub> burden (Holmes, 2018). This might also explain why our estimate of f is at the upper end of previous estimates (Fiore et al., 2009; Voulgarakis et al., 2013; Stevenson et al., 2013; Thornhill et al., 2021b; Stevenson et al., 2020; Sand et al., 2023).

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I would also be interested in the timescale of the change in methane mixing ratio, and the perturbation lifetime. In the methods section you mention a long spin up time. In UKESM-ems we found that with a large methane emissions decrease you get a much faster than expected change in mixing ratio due to the rapid increase in OH and decrease in methane lifetime, and I would expect the opposite effect in an increase such as done here.

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We added the following information about the spin-up of the simulation ERFCH<sub>4</sub>. In particular, the spin-up of the mass of CH<sub>4</sub> follows the exponential function of the form  $a - b \cdot exp(-t/c)$  closely. The fitted perturbation lifetime (parameter *c*) is 21.63 years. After 90 years the mass of CH<sub>4</sub> is 0.5% smaller than the expected equilibrium mass (parameter *a*). We even shortened the spin-up by initializing the atmospheric CH<sub>4</sub> mixing ratios with the reference values scaled by a factor of 2.75 (the increase factor of the emissions).

We additionally fitted f using the function  $m(t) = m_0 * [2.75^f + (2.75 - 2.75^f) * exp(-t/(f * \tau)]$  (Holmes, 2018) using the

yearly mean CH<sub>4</sub> lifetime with respect to OH oxidation for  $\tau$ , which suggest f=1.55. The perturbation lifetime corresponding to the last 5 years of the spin-up is 22.4 years. Both approaches do not account for the dependence of f on the CH<sub>4</sub> burden (Holmes, 2018), as a constant f is fitted.

The perturbation lifetime corresponding to our emission increase simulation is larger than that of the emission reduction by Staniaszek et al. (2022), to which we think you are referring. We think that it is plausible that the perturbation lifetime is shorter for a CH<sub>4</sub> emission reduction as *f* and *τ* depend on the CH<sub>4</sub> burden Holmes (2018). Therefore, also the perturbation lifetime *f* · *τ* depends on the CH<sub>4</sub> burden. In our emission increase simulation, the increase of the CH<sub>4</sub> lifetime is large, and therefore also the extension of the perturbation lifetime is large. It would be interesting to investigate this in targeted simulations, e.g.,
branching of both, a CH<sub>4</sub> emission reduction and emission increase of the same magnitude, from an identical reference.

## Add in line 168 (see also comment 2 by referee 2):

Time series of the global mean surface  $CH_4$ , the total atmospheric masses of  $CH_4$  and ozone  $(O_3)$ , the TOA radiation balance, and GSAT (for the MLO simulations) were monitored to decide whether an equilibrium is reached. In addition, we assessed the

- 55 spin-up of the mass of  $CH_4$  of the simulation  $ERFCH_4$  in more detail. A curve fit was applied to the spin-up period to derive the atmospheric mass of  $CH_4$  in equilibrium. The mass of  $CH_4$  follows the exponential function of the form  $a - b \cdot exp(-t/c)$ closely. The mass of  $CH_4$  in the last year of the spin-up, simulation year 90, is about 0.5% smaller than the derived equilibrium estimate (parameter a), and therefore spun-up sufficiently well (see Fig. S14). The derived perturbation lifetime (parameter c) is 21.6 years. We note that the perturbation lifetime is larger than that of the  $CH_4$  emission reduction experiment by Staniaszek
- 60 et al. (2022). As the perturbation lifetime increases with increasing  $CH_4$  burden (Holmes, 2018), this can be expected. In addition, model differences and the magnitude of the emission change might play a role.

## References

- Holmes, C. D.: Methane Feedback on Atmospheric Chemistry: Methods, Models, and Mechanisms, J. Adv. Model. Earth Syst., 10, 1087– 1099, https://doi.org/10.1002/2017MS001196, 2018.
- 65 Staniaszek, Z., Griffiths, P. T., Folberth, G. A., O'Connor, F. M., Abraham, N. L., and Archibald, A. T.: The role of future anthropogenic methane emissions in air quality and climate, npj Clim Atmos Sci, 5, https://doi.org/10.1038/s41612-022-00247-5, 2022.