Supplement of Atmos. Chem. Phys., 25, 5133–5158, 2025
https://doi.org/10.5194/acp-25-5133-2025-supplement

Atmospheric Chemistry and Physics
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EGU

Supplement of

Chemistry–climate feedback of atmospheric methane in a methane-emission-flux-driven chemistry–climate model

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Table S1. CH_4 loss, i.e. $\frac{1}{\tau}$, by reaction with OH, Cl and O(^{1}D) in the troposphere (trop.) and the stratosphere (strat.), separately, [a^{-1}].

	CH_4 [a^{-1}] loss by				
	trop. OH	trop. Cl	strat. OH	strat. Cl	strat. O(^{1}D)
REF-SSTfix	0.132	0.0003	0.018	0.008	0.014
REF-SSTvar	0.132	0.0003	0.018	0.008	0.014
$ERFCO_2$	0.132	0.0003	0.018	0.007	0.014
$ECCCO_2$	0.142	0.0003	0.018	0.008	0.014
$ERFCH_4$	0.069	0.00003	0.029	0.004	0.014
$ECCCH_4$	0.076	0.00004	0.029	0.004	0.014

The corresponding lifetime is calculated using either OH, Cl or O(^{1}D) as the reaction partner in Eq. 2. For the tropospheric (stratospheric) loss, all grid boxes below (above) the climatological tropopause (tp_{clim}= 300 hPa $-$ 215 hPa $\cdot$ $\cos^2(\phi)$) are taken into account. The corresponding reaction rate coefficients are calculated as in the applied kinetic equation system (submodel MECCA): $k_{CH_4+OH}(T) = 1.85 \times 10^{-20} \cdot T^{2.82} \cdot \exp(-\frac{987}{T})$, $k_{CH_4+Cl}(T) = 6.6 \times 10^{-12} \cdot \exp(-\frac{1240}{T})$, $k_{CH_4+O^1D} = 1.75 \times 10^{-10}$.

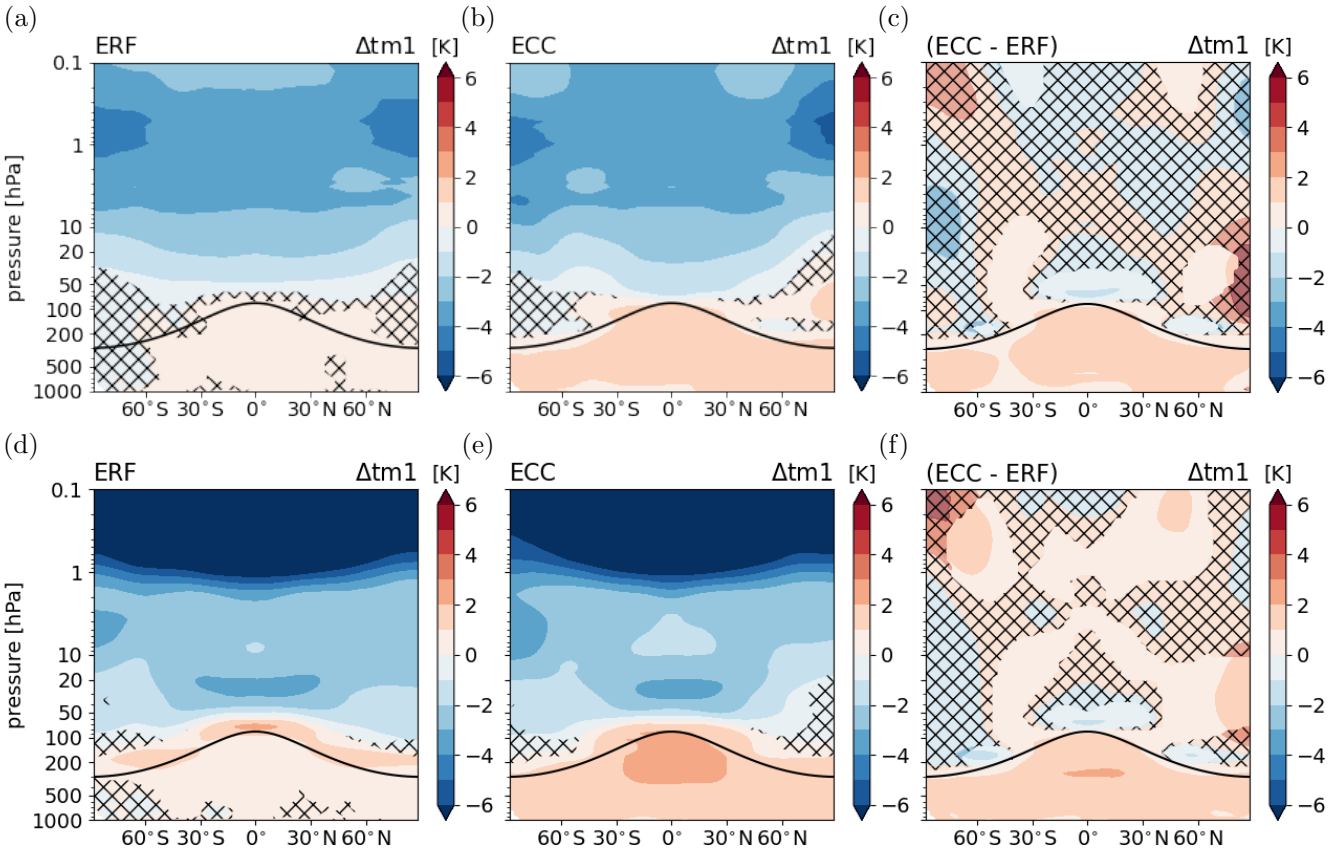

Figure S1. Annual zonal mean temperature response following 1.35×CO_2 (a,b,c) and 2.75×CH_4 (d,e,f) in [K]: Absolute temperature difference between the sensitivity simulations (a) ERFCO_2 (fast response) and (b) ECCCO_2 (full response) and their respective reference simulation. (c) Climate response as the difference between the temperature responses in panels (a) and (b). Absolute temperature difference between the sensitivity simulations (d) ERFCH_4 (fast response) and (e) ECCCH_4 (full response) and their respective reference simulation. (f) Climate response as the difference between the temperature responses in panels (d) and (e). Non-hatched areas are significant at the 95% confidence level according to a Welch test based on annual mean values. The solid black line indicates the location of the climatological tropopause.

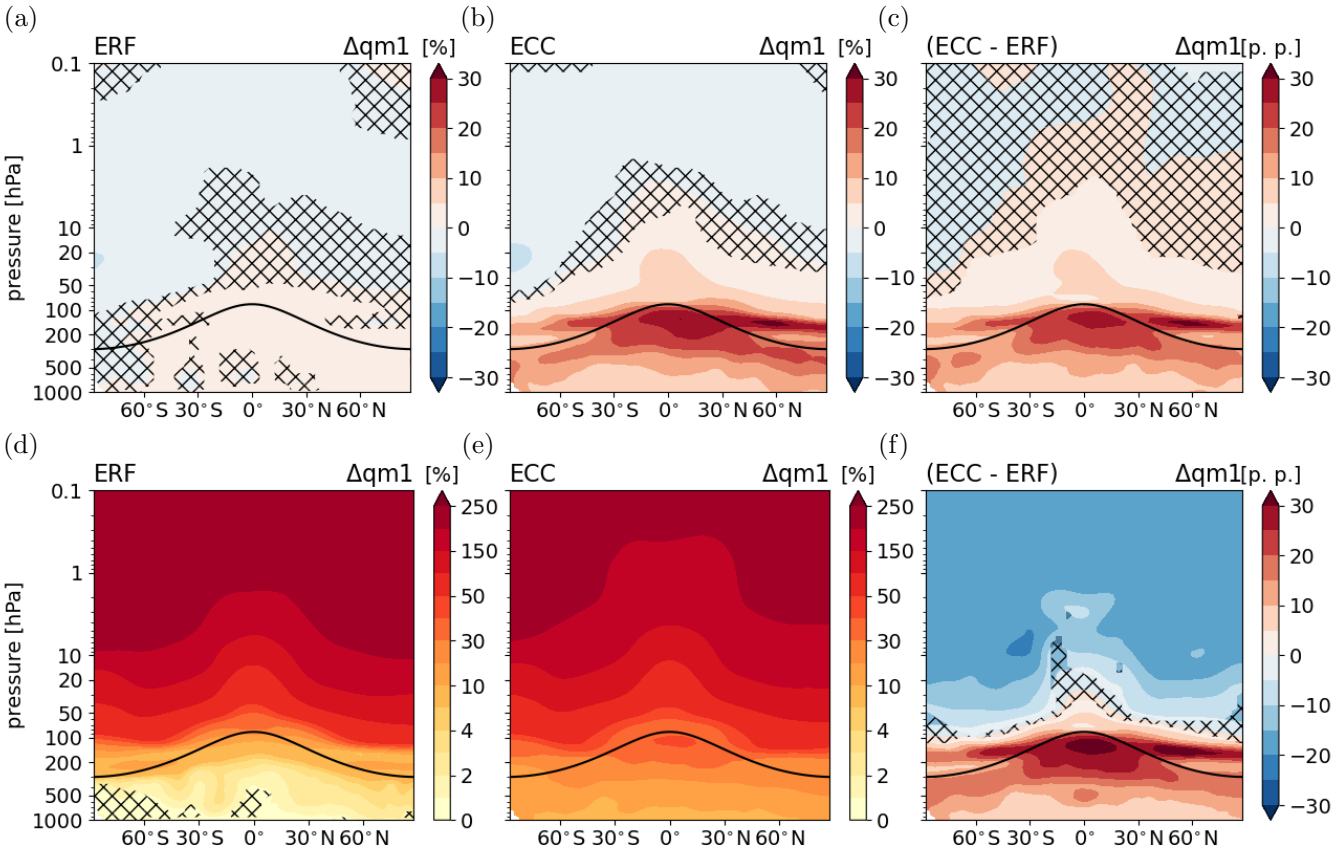

Figure S2. Annual zonal mean response of specific humidity following 1.35×CO_2 (a,b,c) and 2.75×CH_4 (d,e,f): Relative difference between specific humidity of sensitivity simulations (a) ERFCO_2 (fast response) and (b) ECCCO_2 (full response) and their respective reference simulation in [%]. (c) Climate response as the difference between the responses in panels (a) and (b) in percentage points [p.p.]. Relative difference between specific humidity of sensitivity simulations (d) ERFCH_4 (fast response) and (e) ECCCH_4 (full response) and their respective reference simulation in [%]. (f) Climate response as the difference between the responses in panels (d) and (e) in percentage points [p.p.]. Non-hatched areas are significant at the 95% confidence level according to a Welch test based on annual mean values. The solid black line indicates the location of the climatological tropopause.

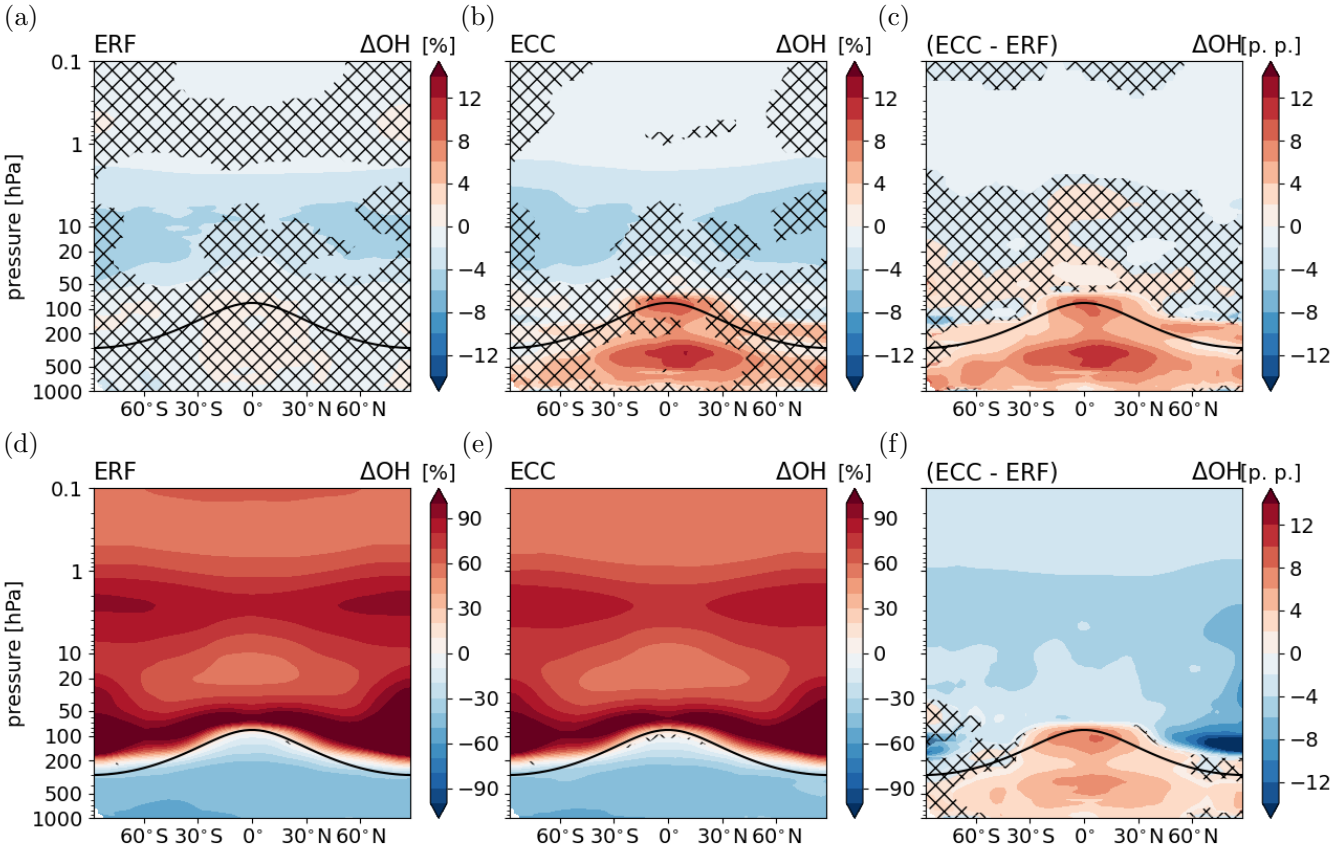

Figure S3. As Fig. S14 for the hydroxyl radical (OH).

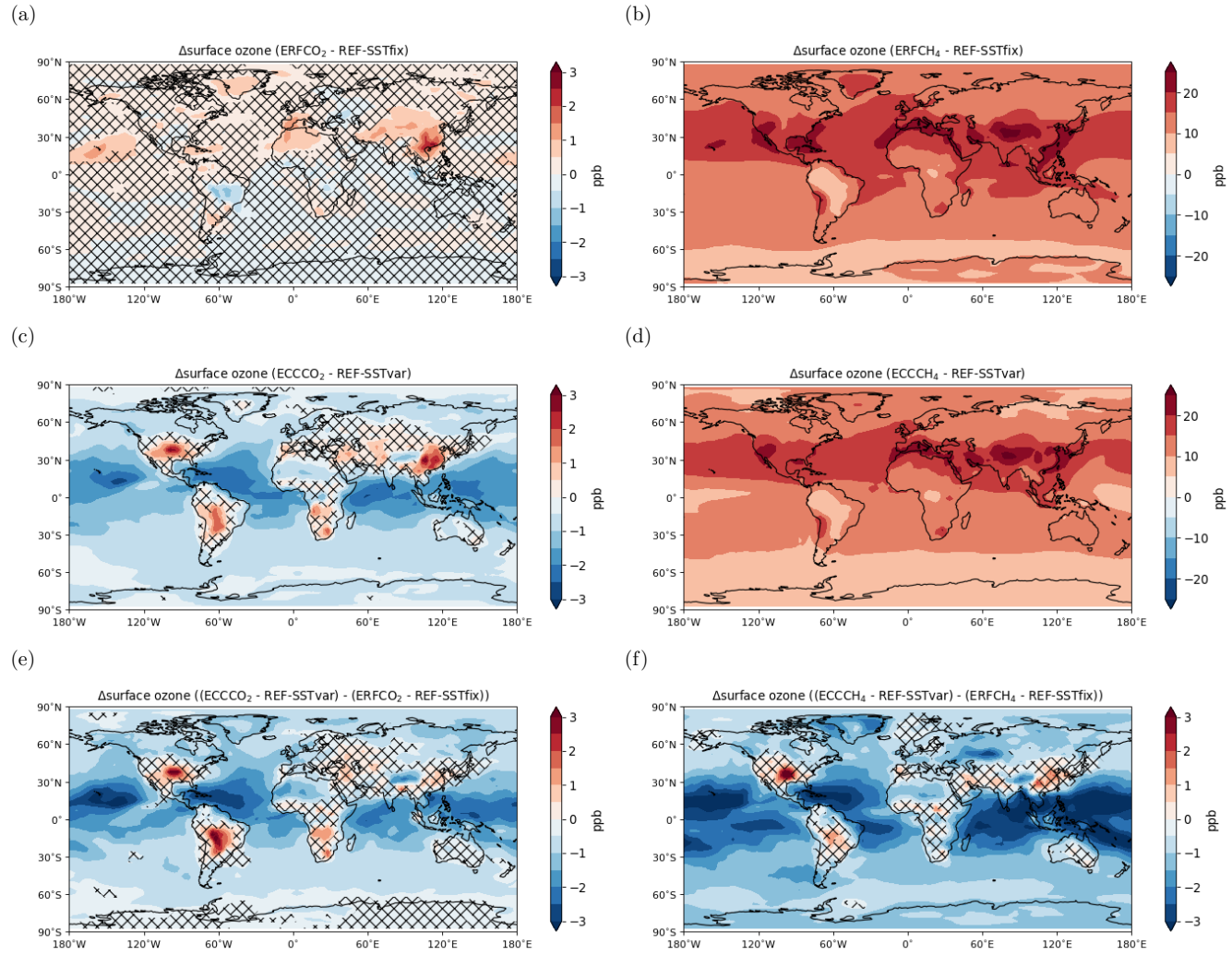

Figure S4. Annual mean surface O_3 response following $1.35\times CO_2$ (a,c,e) and $2.75\times CH_4$ (b,d,f) in [ppm]: Difference between the sensitivity simulations (a) $ERFCO_2$ (fast response) and (c) $ECCCO_2$ (full response) and their respective reference simulation. (e) Climate response as the difference between the surface O_3 responses in panels (a) and (c). Difference between the sensitivity simulations (b) $ERFCH_4$ (fast response) and (d) $ECCCH_4$ (full response) and their respective reference simulation. (f) Climate response as the difference between the surface O_3 responses in panels (b) and (d). Non-hatched areas are significant at the 95% confidence level according to a Welch test based on annual mean values.

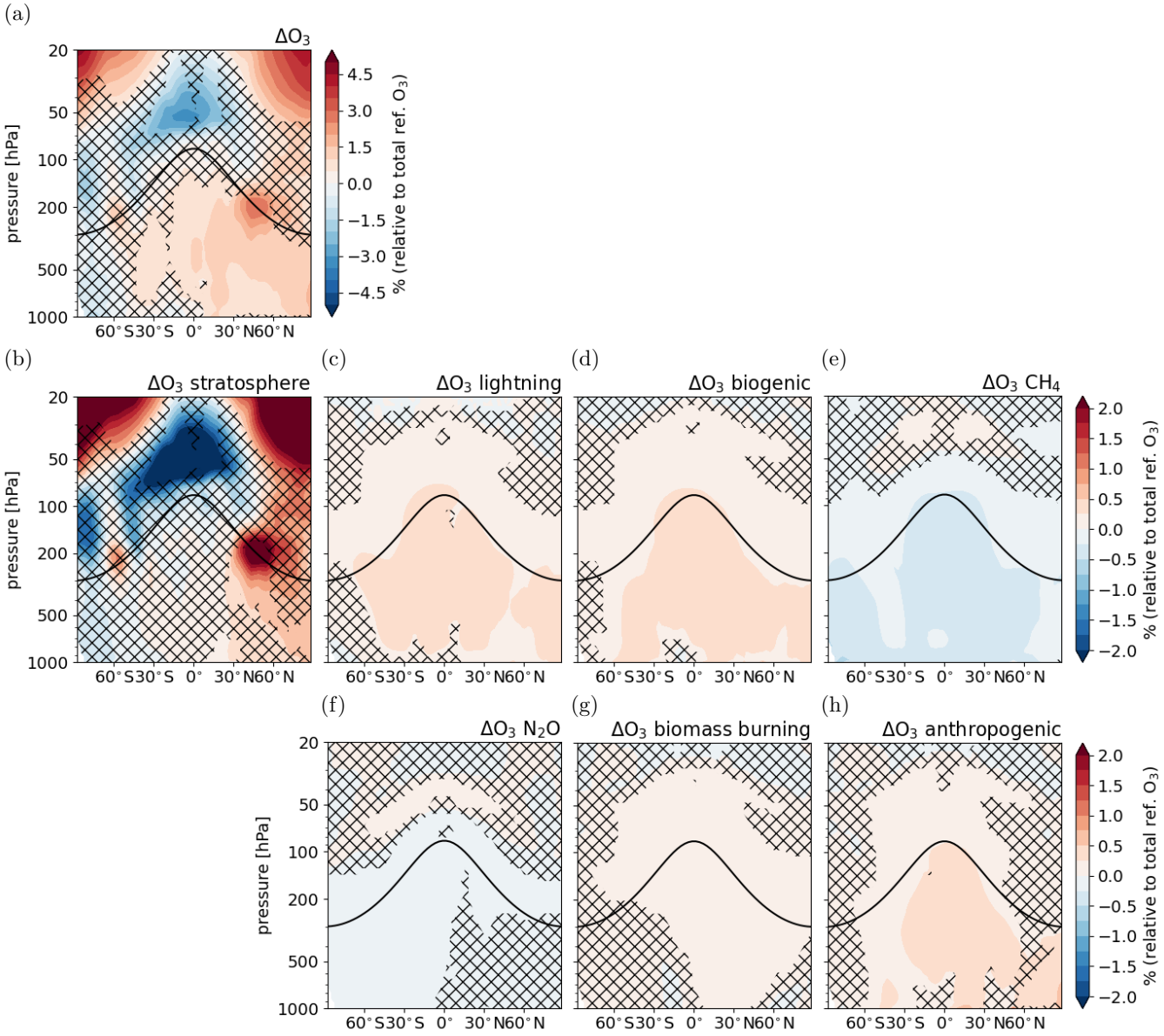

Figure S5. Fast response of tropospheric O_3 following the CO_2 perturbation: (a) response of total O_3 (same as Fig. 2 (a) in the main manuscript but with differently scaled colour levels to better compare with the response in the individual categories), (b-h) response of O_3 in individual source categories relative to total reference O_3 ($\Delta O_{3_{\text{cat}}} = \frac{O_{3_{\text{cat,ERF}}} - O_{3_{\text{cat,REF}}}}{O_{3_{\text{total,REF}}}}$). Non-hatched areas are significant at the 95% confidence level according to a Welch test based on annual mean values. The solid black line indicates the location of the climatological tropopause.

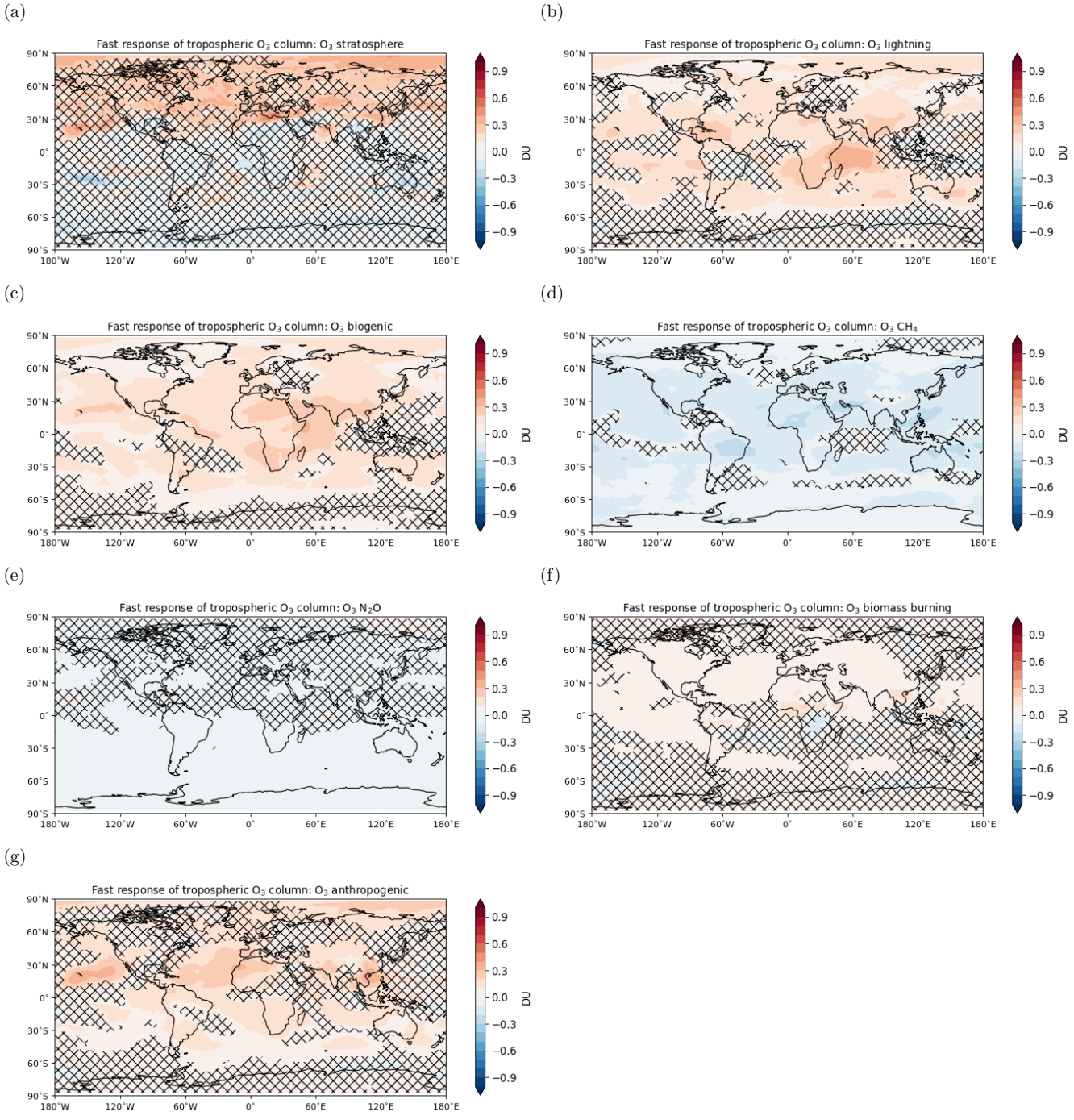

Figure S6. Fast response of tropospheric O_3 column following the CO_2 perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the CO_2 perturbation simulation with prescribed SSTs and SICs and interactive chemistry ($ERFCO_2$) and the reference simulation REF-SSTfix at the 95% interval.

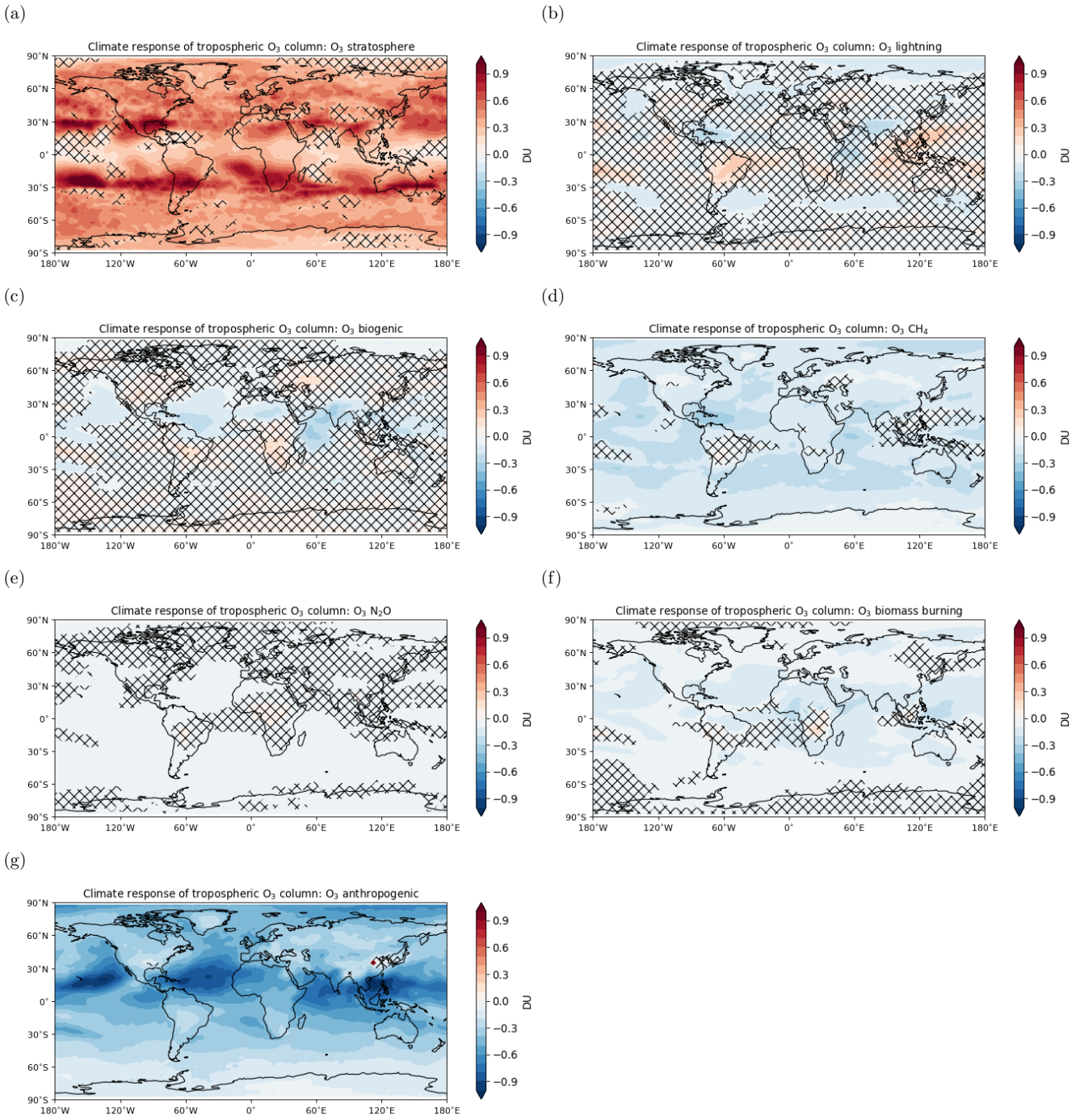

Figure S7. Climate response of tropospheric O_3 column following the CO_2 perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the fast and the full response at the 95% interval.

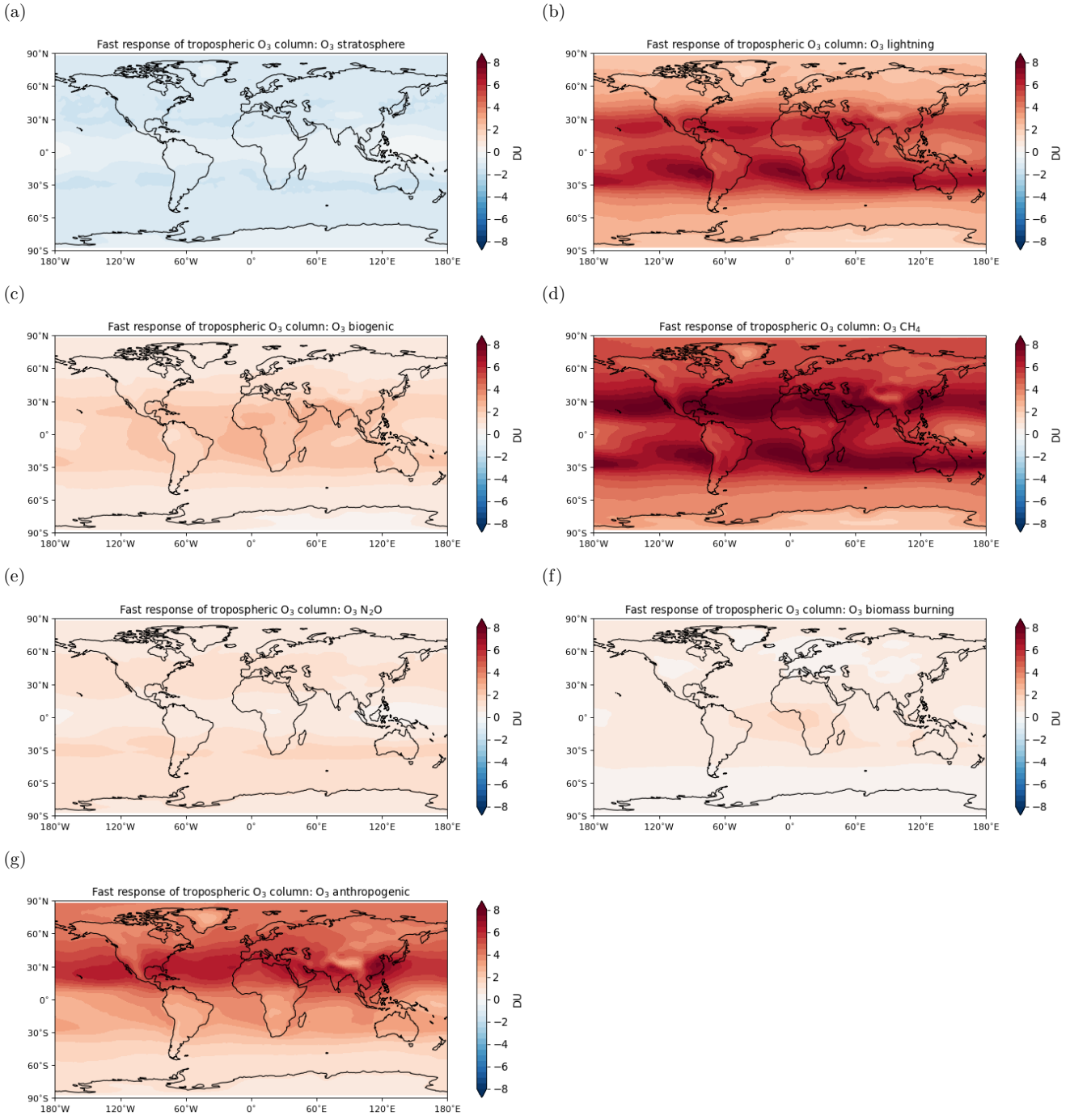

Figure S8. Fast response of tropospheric O_3 column following the CH_4 emission flux perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the CH_4 perturbation simulation with prescribed SSTs and SICs and interactive chemistry (ERFCH$_4$) and the reference simulation REF-SSTfix at the 95% interval.

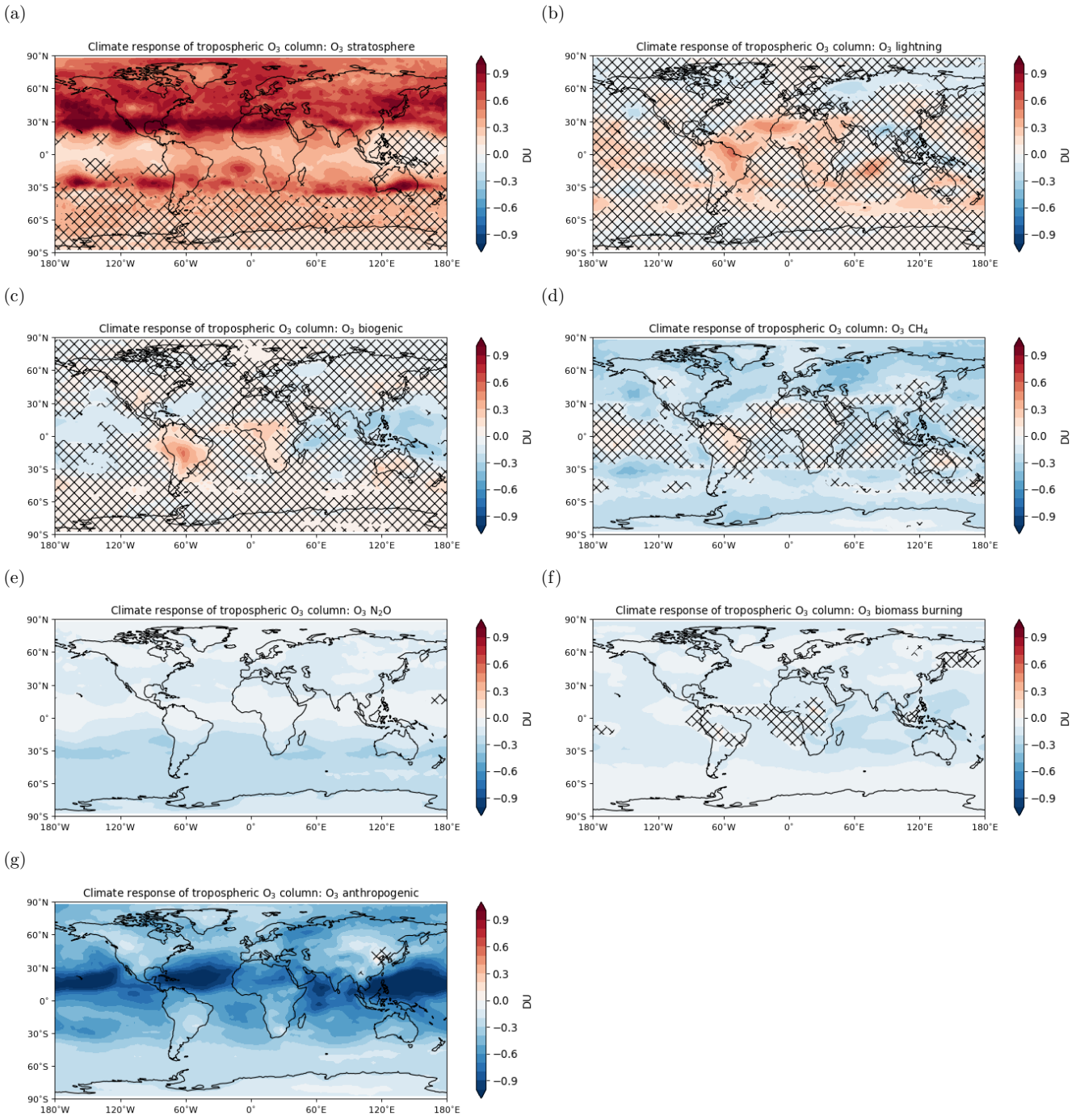

Figure S9. Climate response of tropospheric O_3 column following the CH_4 emission flux perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the fast and the full response at the 95% interval.

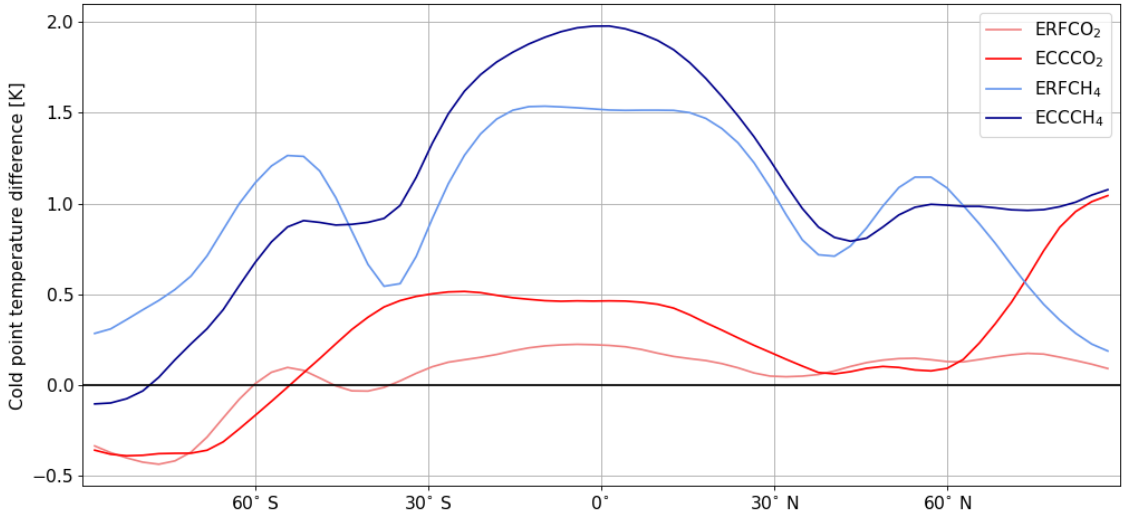

Figure S10. Zonal mean difference of cold point temperature between the sensitivity simulations perturbed by 1.35×CO_2 mixing ratio (reddish colours) or 2.75×CH_4 emission flux increase (bluish colours) and the respective references in [K].

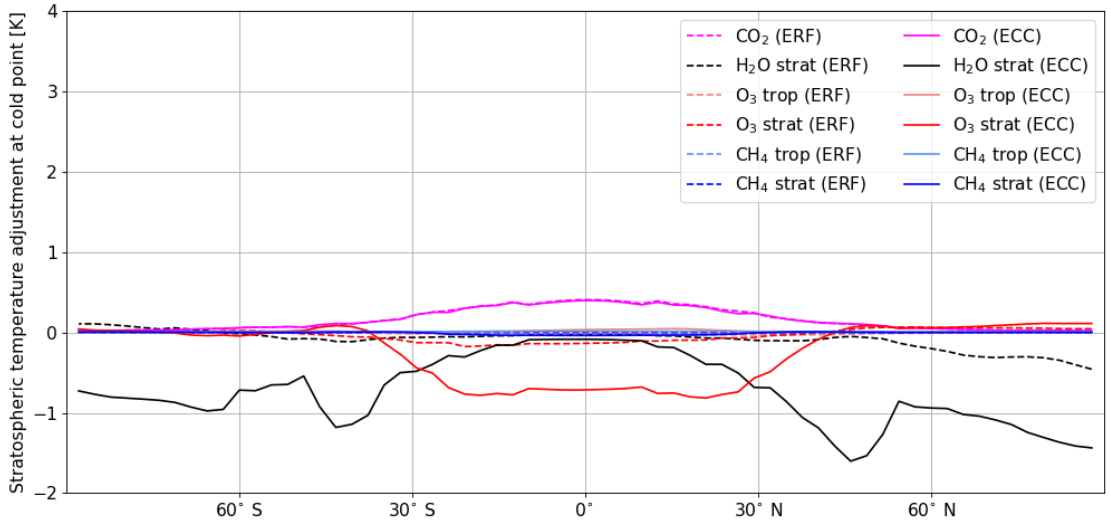

Figure S11. Stratospheric temperature adjustment corresponding to individual composition changes of CO_2, H_2O, O_3, and CH_4 in the simulations ERFCO_2 (ERF) or ECCCO_2 (ECC) evaluated at the cold point of the respective perturbation simulation in [K].

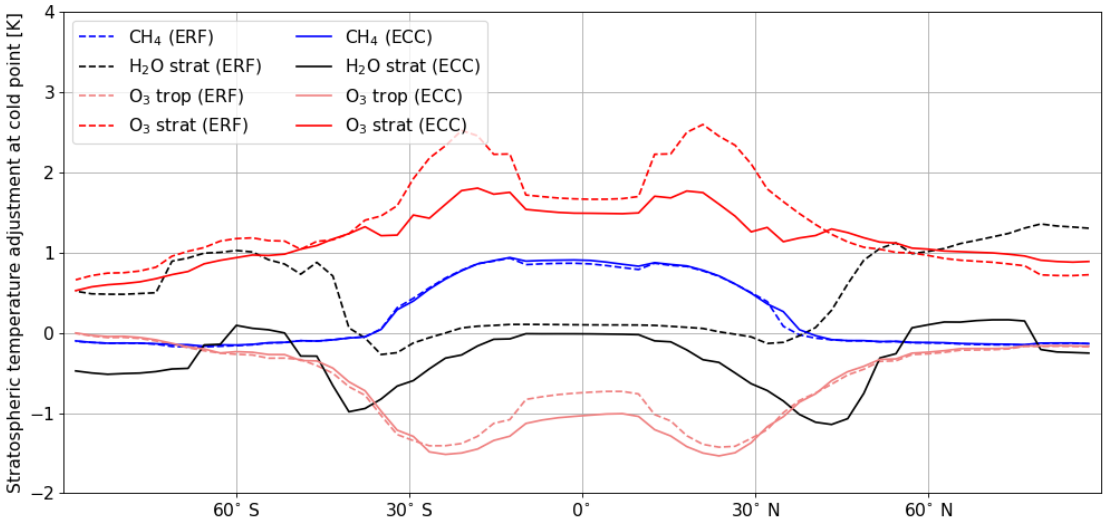

Figure S12. Stratospheric temperature adjustment corresponding to individual composition changes of CH_4, H_2O, and O_3 in the simulations ERFCH_4 (ERF) or ECCCH_4 (ECC) evaluated at the cold point of the respective perturbation simulation in [K].

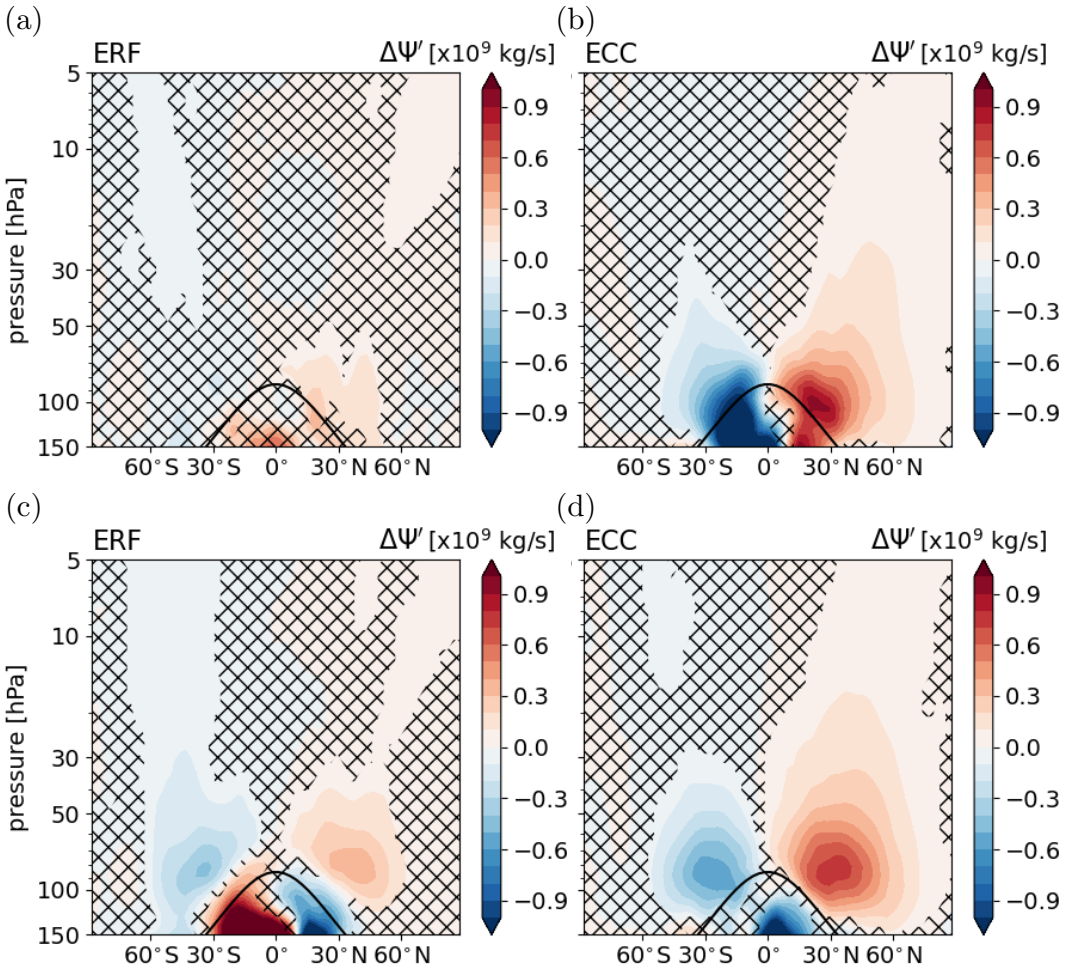

Figure S13. Annual zonal mean response of the residual streamfunction Ψ' following 1.35×CO_2 (a,b) and 2.75×CH_4 (c,d) in [10^9 kg s^{-1}]: Absolute differences between the sensitivity simulations (a) ERFCO_2 (fast response), (b) ECCCO_2 (full response), (c) ERFCH_4 (fast response), and (d) ECCCH_4 (full response) compared to their respective reference. Non-hatched areas are significant at the 95% confidence level according to a Welch test based on annual mean values. The solid black line indicates the location of the climatological tropopause.

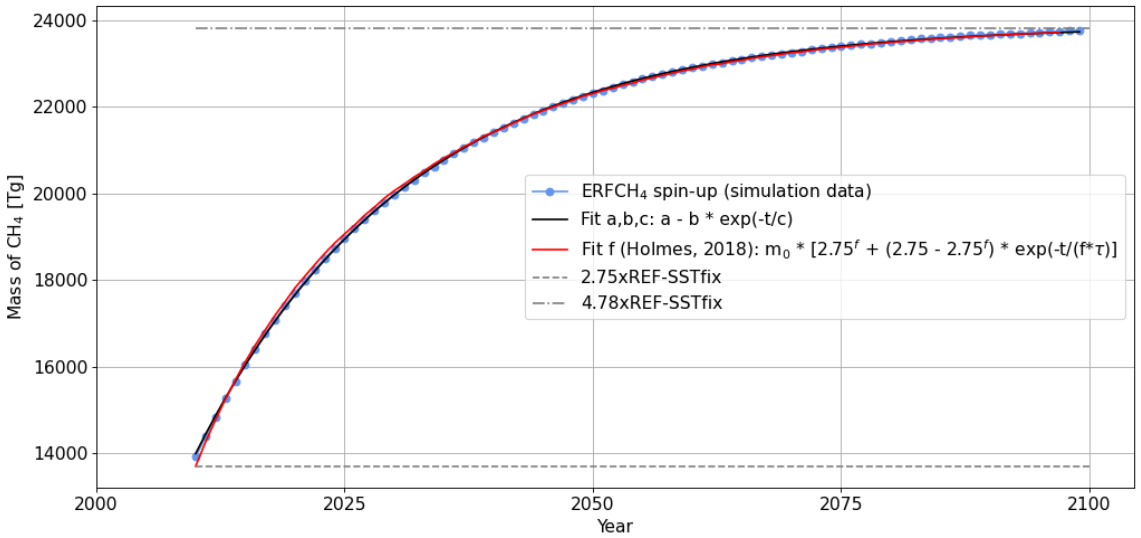

Figure S14. Yearly mean atmospheric mass of CH_4 of the spin-up period of the simulation ERFCH$_4$ (blue curve). In addition, the results of two curve fits are shown: A free parameter fit to the function $m(t) = a - b \cdot exp(-t/c)$ (black curve), as well as $m(t) = m_0 * [2.75^f + (2.75 - 2.75^f) * exp(-t/(f * \tau)]$ (Holmes, 2018) (red curve). The constant lines at 2.75×REF-SSTfix and 4.78×REF-SSTfix indicate the initial condition of the spin-up and the new equilibrium of ERFCH$_4$, respectively.

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.