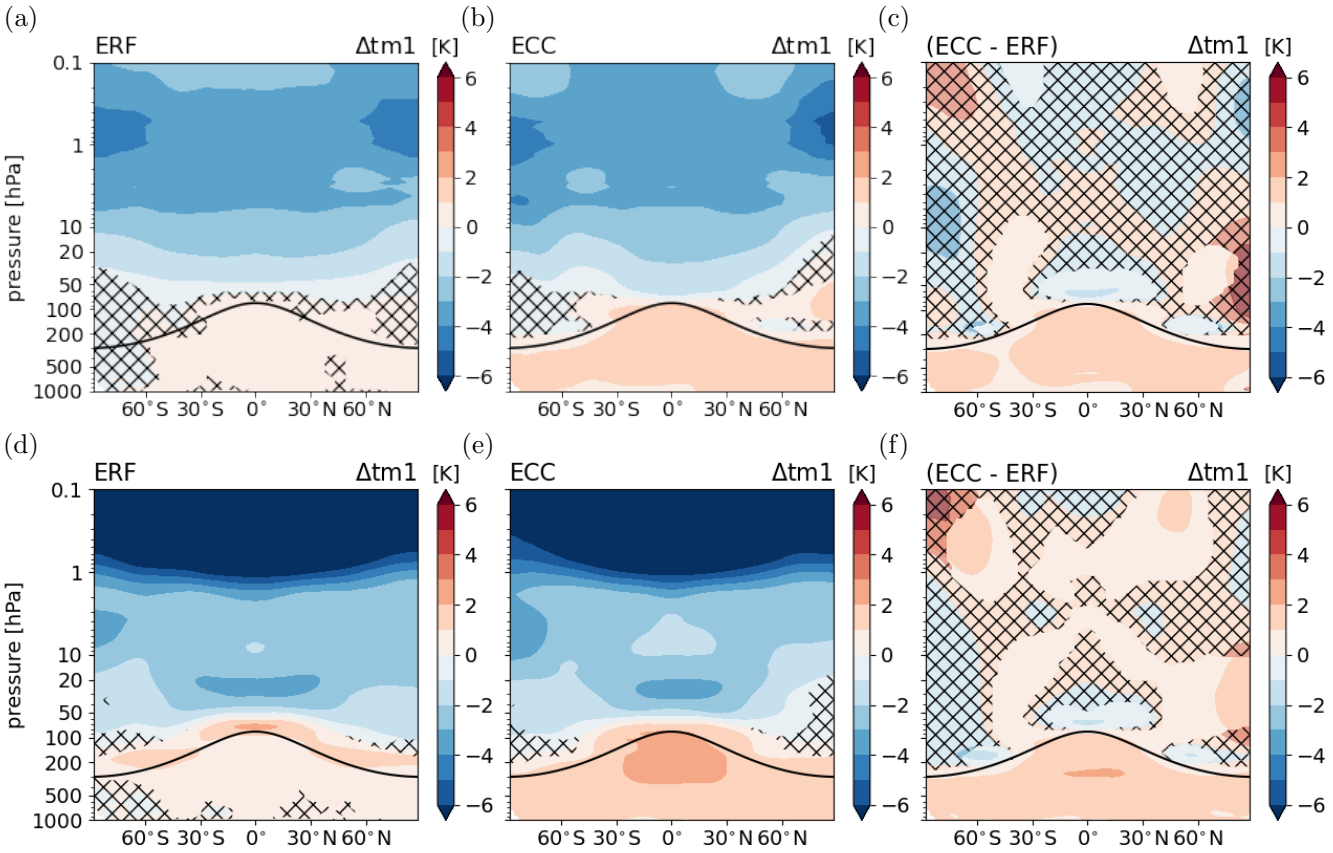

Figure S1. Annual zonal mean temperature response following 1.35×CO_2 (a,b,c) and 2.75×CH_4 (d,e,f) in [K]: Absolute temperature difference between the sensitivity simulations (a) ERFCO_2 (fast response) and (b) ECCCO_2 (full response) and their respective reference simulation. (c) Climate response as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

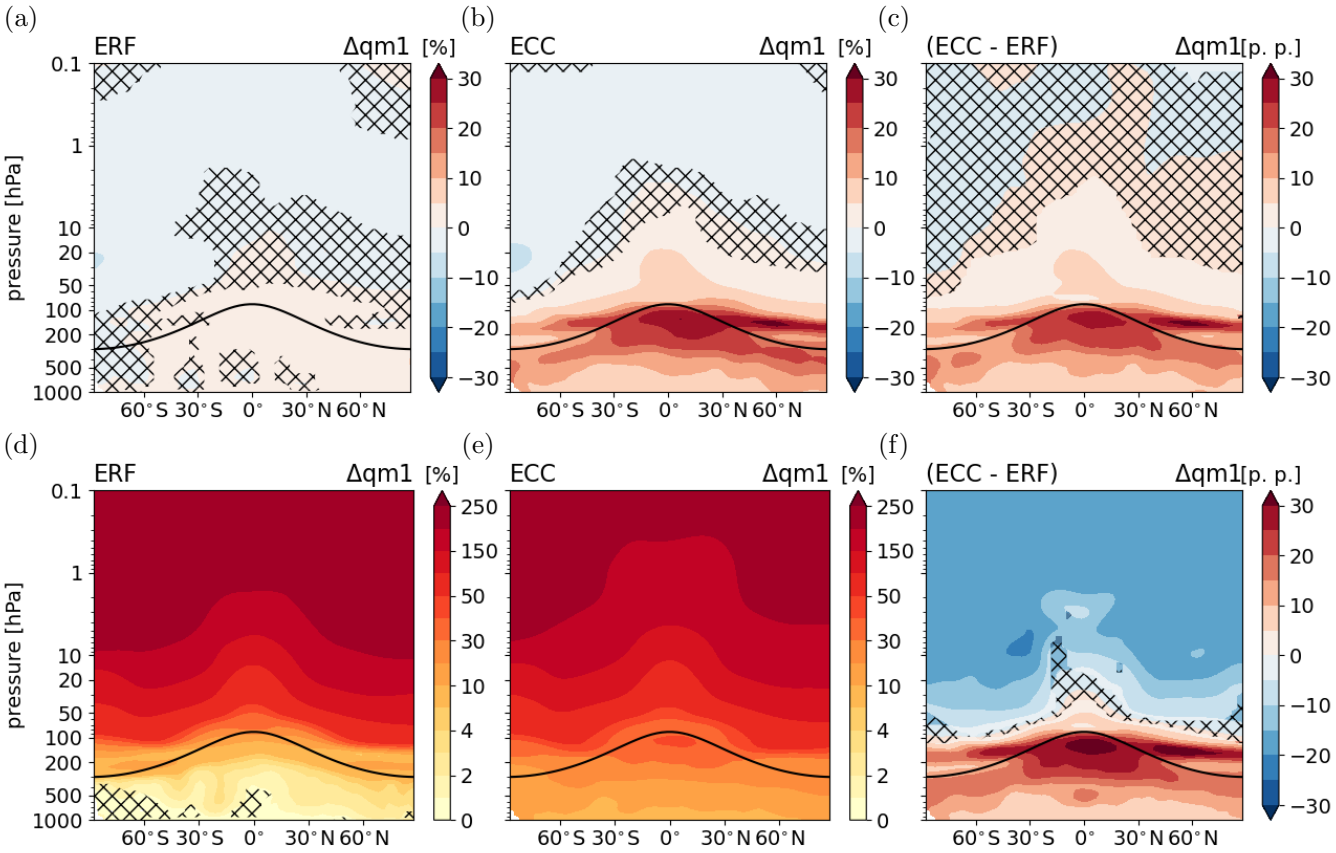

Figure S2. Annual zonal mean response of specific humidity following 1.35×CO_2 (a,b,c) and 2.75×CH_4 (d,e,f): Relative difference between specific humidity of sensitivity simulations (a) ERFCO_2 (fast response) and (b) ECCCO_2 (full response) and their respective reference simulation in [%]. (c) Climate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

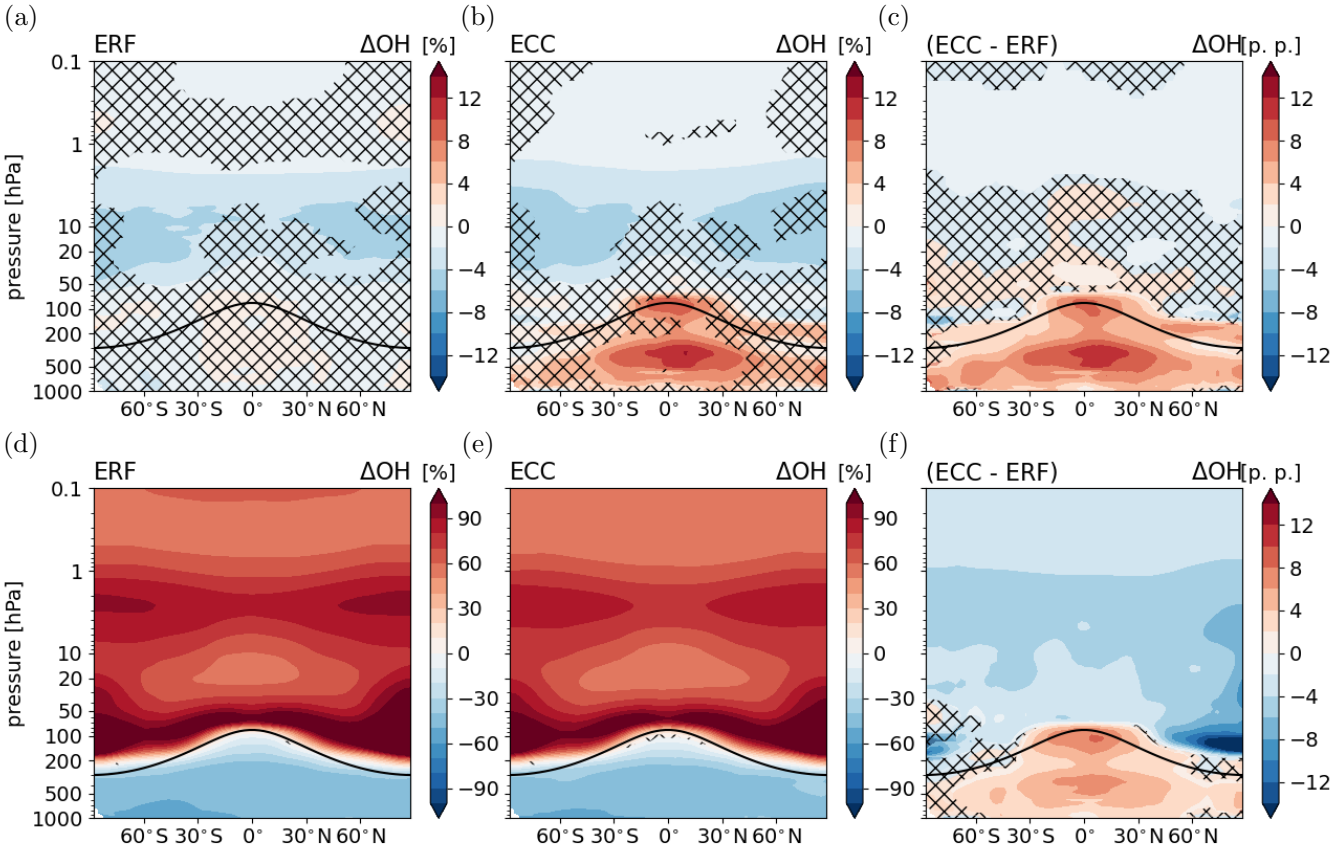

Figure S3. As Fig. S2 for the hydroxyl radical (OH).

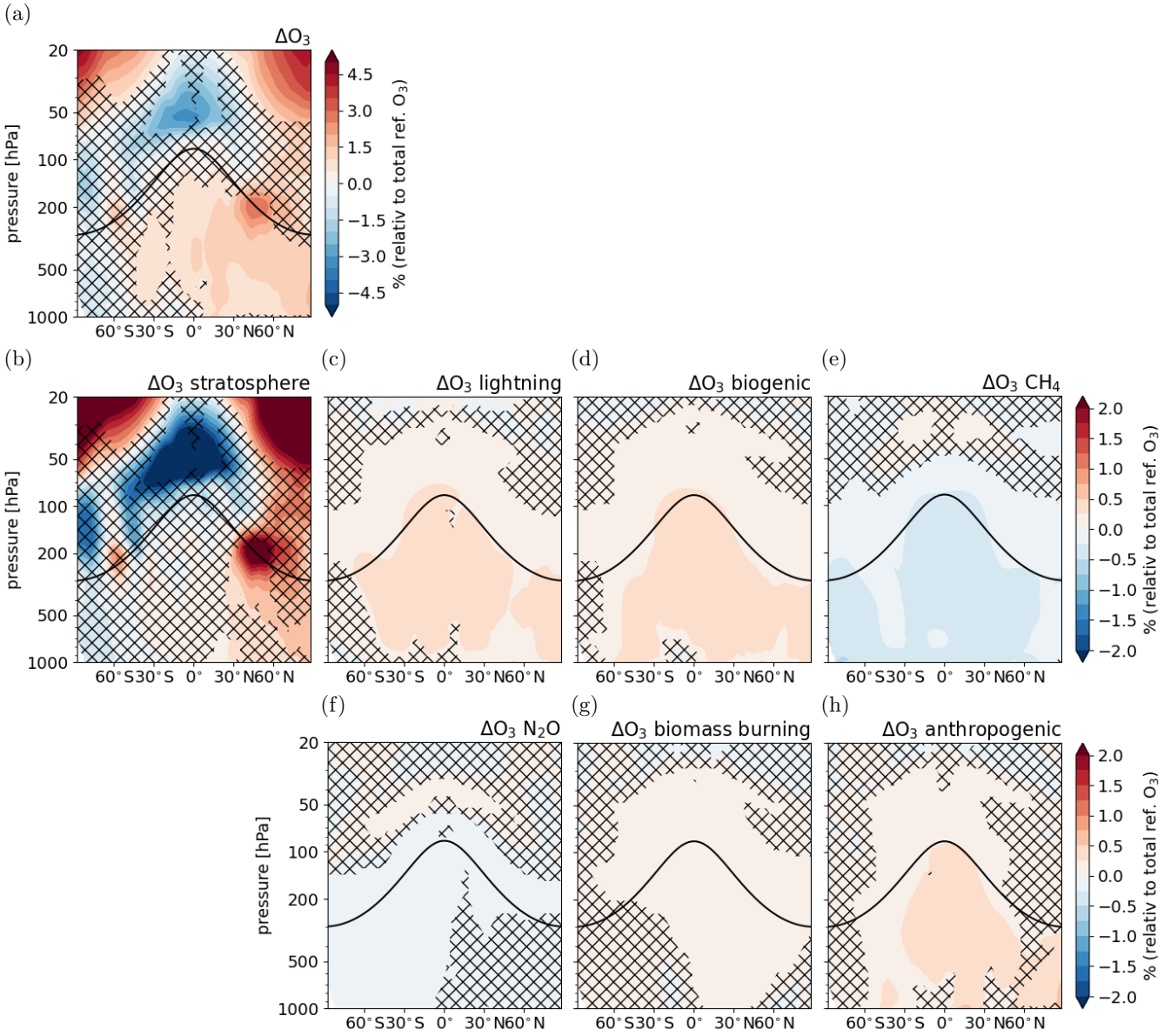

Figure S4. Fast response of tropospheric O_3 following the CO_2 perturbation: (a) response of total O_3 (same as Fig. 2 (a) in the main manuscript, but differently scaled colour levels to better compare with the response in the individual categories), (b) - (h) response of O_3 in individual source categories 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 on the 95% confidence level according to a Welch's test based on annual mean values. The solid black line indicates the location of the climatological tropopause.

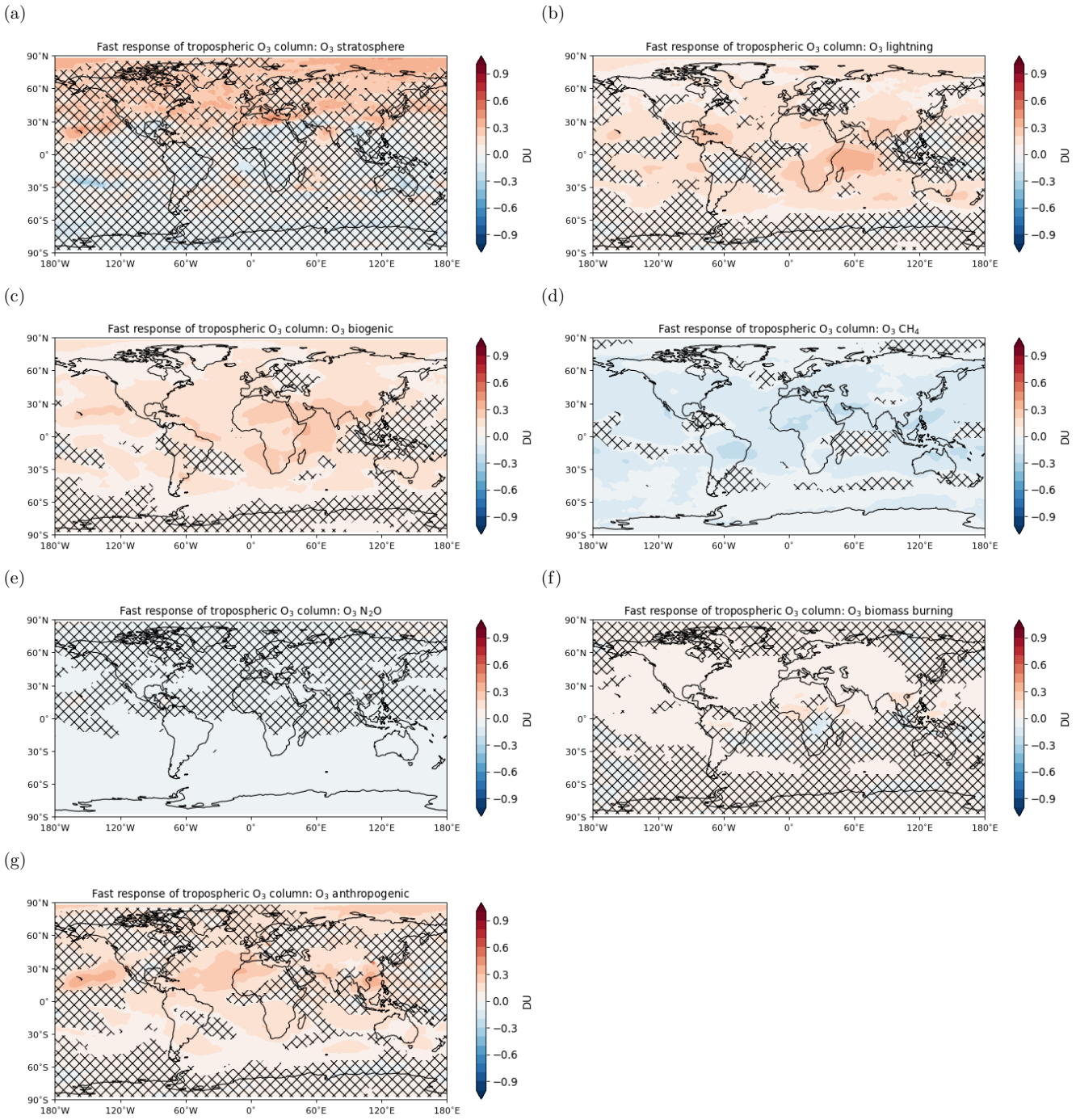

Figure S5. Fast response of tropospheric O_3 column following the CO_2 perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the simulation ERFCO$_2$ and REF-SSTfix on the 95% interval.

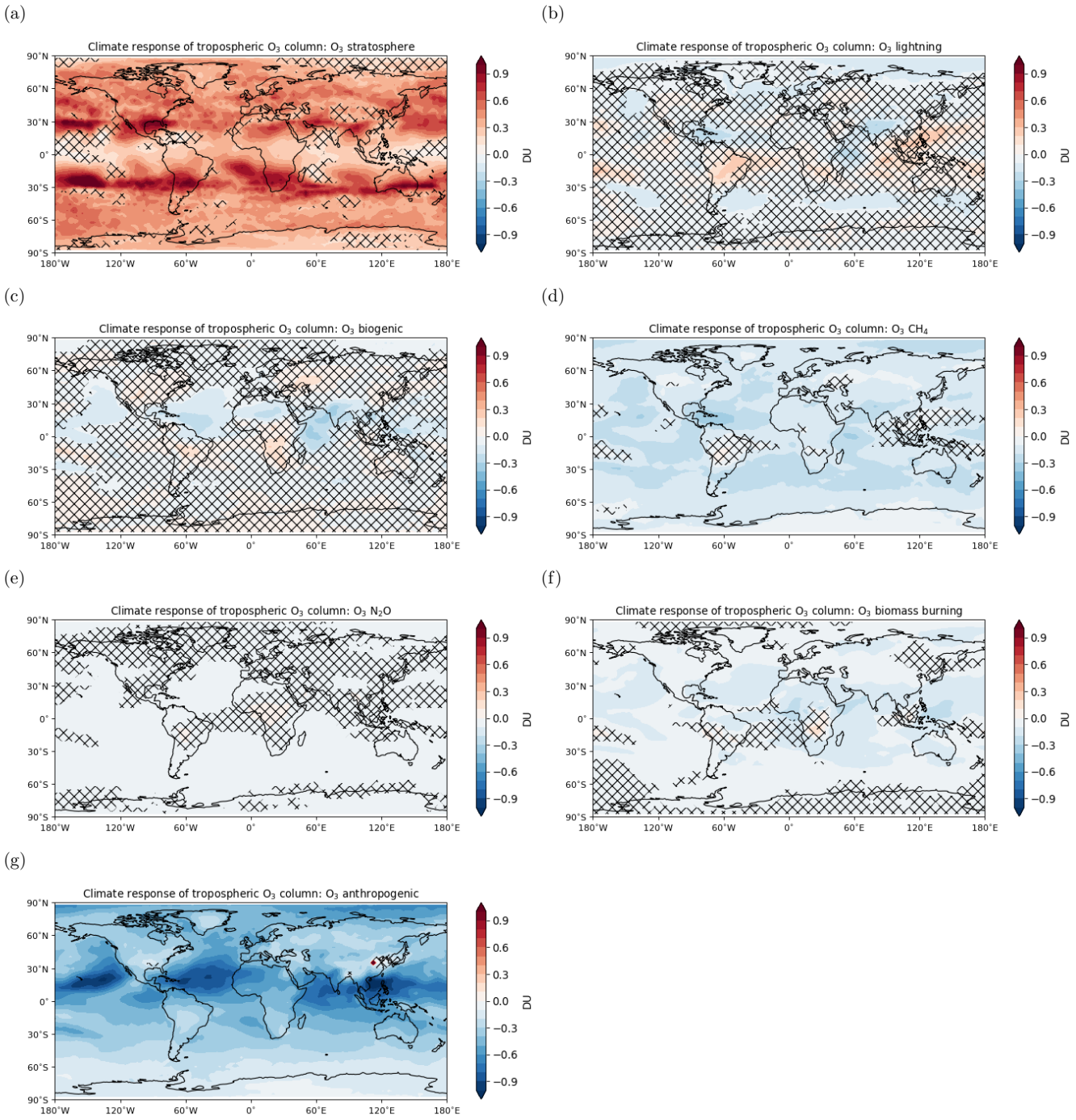

Figure S6. Climate response of tropospheric O_3 column following the CO_2 perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the fast and the full response.

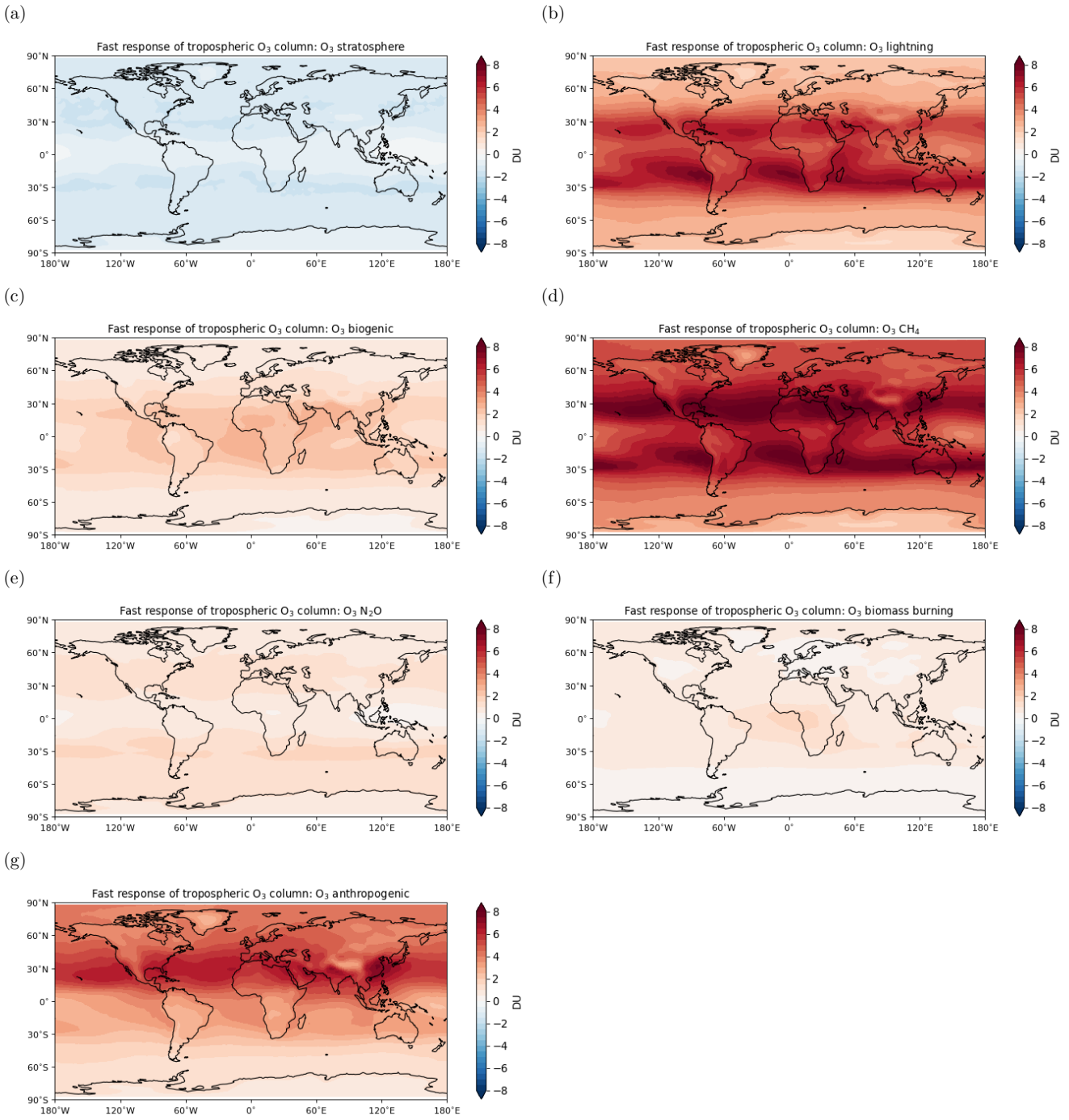

Figure S7. Fast response of tropospheric O_3 column following the CH_4 emission flux perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the simulation ERFCH$_4$ and REF-SSTfix on the 95% interval.

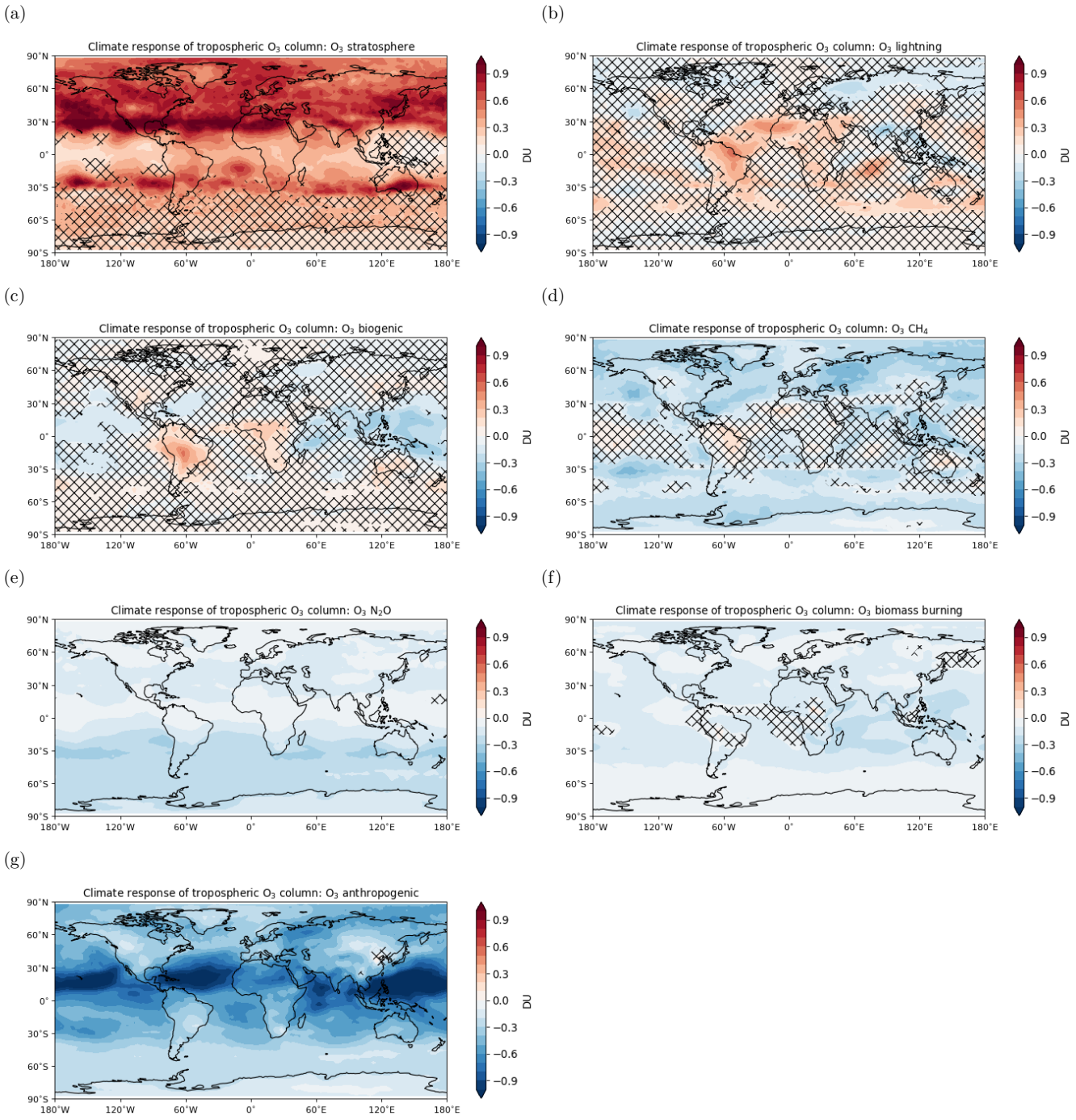

Figure S8. Climate response of tropospheric O_3 column following the CH_4 emission flux perturbation for individual source categories in DU. Non-hatched regions indicate significant differences between the fast and the full response.

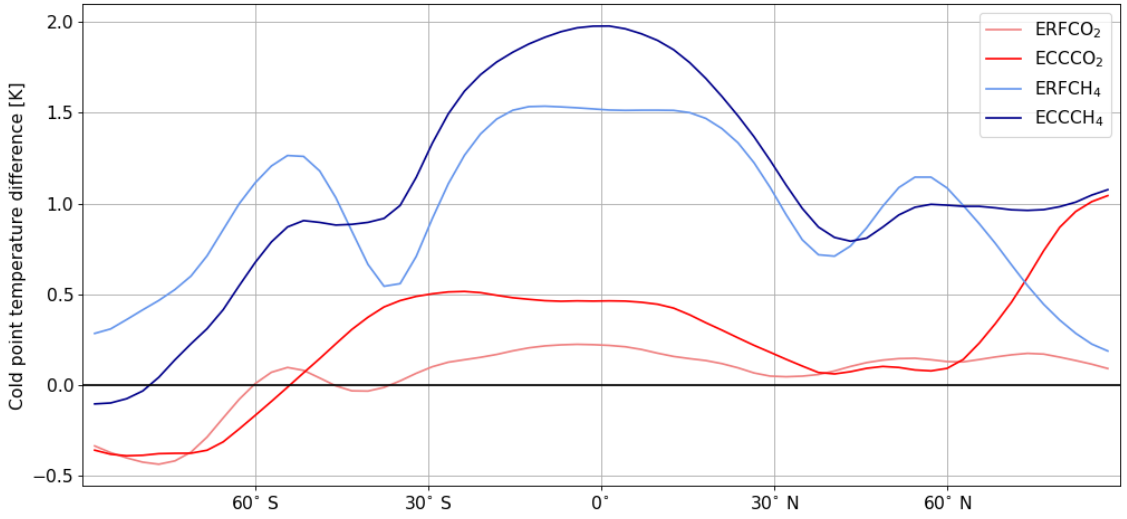

Figure S9. Zonal mean difference of cold point temperature between sensitivity simulations perturbed by 1.35×CO_2 mixing ratio (reddish colours) and 2.75×CH_4 emission flux increase (bluish colours) and the respective references in [K].

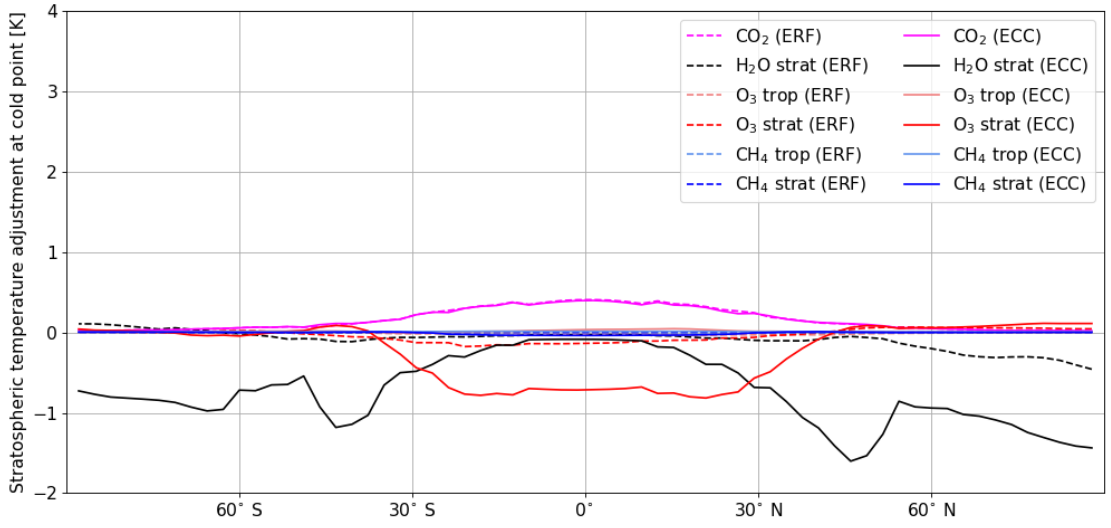

Figure S10. Stratospheric temperature adjustment corresponding to individual composition changes of CO_2, H_2O, O_3 and CH_4 in the simulations ERFCO_2 (ERF) or ECCCO_2 (ECC) evaluated at the cold point of the respective perturbation simulation in [K].

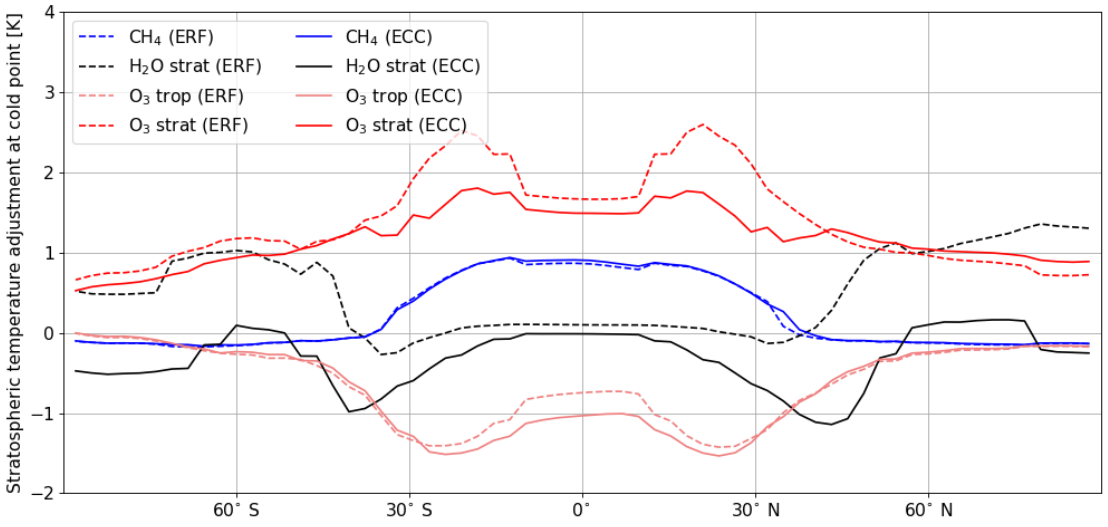

Figure S11. Stratospheric temperature adjustment corresponding to individual composition changes of CH_4, H_2O and O_3 in the simulations ERFCH_4 (ERF) or ECCCH_4 (ECC) evaluated at the cold point of the respective perturbation simulation in [K].