Supplement of: Effects of Strongly Enhanced Atmospheric Methane Concentrations in a Fully Coupled Chemistry-Climate Model

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Figure S1. Climatologies of annual mean sea surface temperatures (SSTs) of REF MLO (top left) and the climatology (Rayner et al., 2003) prescribed as lower boundary for REF QFLX and REF fSST (top right), and their difference (bottom middle).



Figure S2. Monthly climatologies of total sea ice area in the Northern Hemisphere (left) and in the Southern Hemisphere (right) for REF MLO (blue), S2 MLO (orange), S5 MLO (red) and the prescribed climatology (black). For the mixed layer ocean (MLO) simulations the interannual standard deviations are indicated by vertical lines.



Figure S3. Absolute differences of the residual mean streamfunction for season December, January, and February (DJF) of the sensitivity simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO and their respective reference in 10^9 kg s⁻¹. Non-stippled areas are significant on the 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S4. Absolute differences of the residual mean streamfunction for season March, April, and May (MAM) of the sensitivity simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO and their respective reference in 10^9 kg s⁻¹. Non-stippled areas are significant on the 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S5. Absolute differences of the residual mean streamfunction for season June, July, and August (JJA) of the sensitivity simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO and their respective reference in 10^9 kg s⁻¹. Non-stippled areas are significant on the 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S6. Absolute differences of the residual mean streamfunction for season September, October, and November (SON) of the sensitivity simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO and their respective reference in 10^9 kg s⁻¹. Non-stippled areas are significant on the 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S7. Relative differences between the annual zonal mean hydroxyl radical (OH) mixing ratio of the sensitivity simulations S2 MLO (left) and S5 MLO (right) and REF MLO in %. Non-stippled areas are significant on a 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S8. Relative differences between the annual zonal mean water vapour (H₂O) mixing ratio of the sensitivity simulations S2 MLO (left) and S5 MLO (right) and REF MLO in %. Non-stippled areas are significant on a 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S9. Cold point pressure in hPa (left) and difference of cold point temperature of the sensitivity simulations and their respective reference in K (right).



Figure S10. Relative differences between the annual zonal mean ozone (O_3) mixing ratio of the sensitivity simulations S2 MLO (left) and S5 MLO (right) and REF MLO in %. Non-stippled areas are significant on a 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological troppause height of REF MLO.



Figure S11. Stratospheric temperature adjustment radiatively induced by individual species changes in simulation S2 MLO (2×methane (CH₄)): (a) CH₄, H₂O and O₃ combined, (b) CH₄, (c) H₂O, (d) tropospheric H₂O only, (e) stratospheric H₂O only (SWV), (f) O₃, (g) tropospheric O_3 only and (h) stratospheric O_3 only. Note the different colour bars in panels (a), (b), (d) and (g).



Figure S12. Difference between stratospheric temperature adjustment in simulations S2 MLO and S2 fSST ($2 \times CH_4$) radiatively induced by individual species changes: (a) CH₄, H₂O and O₃ combined, (b) CH₄, (c) H₂O, (d) tropospheric H₂O only, (e) stratospheric H₂O only (SWV), (f) O₃, (g) tropospheric O₃ only and (h) stratospheric O₃ only. Note the different colour bars in panels (a), (b), (d) and (g).



Figure S13. Dynamical temperature response effect for season DJF of the simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO. The dynamical effect is calculated as the difference between the temperature response in the regular simulations (Δ T(SX-REF) with X either 2 or 5) and the sum of the individual contributions of CH₄, H₂O and O₃ to the adjusted stratospheric temperatures (Δ T_{addst}(SX*-REF*) with X either 2 or 5).



Figure S14. Dynamical temperature response effect for season MAM of the simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO. The dynamical effect is calculated as the difference between the temperature response in the regular simulations (Δ T(SX-REF) with X either 2 or 5) and the sum of the individual contributions of CH₄, H₂O and O₃ to the adjusted stratospheric temperatures (Δ T_{addst}(SX*-REF*) with X either 2 or 5).



Figure S15. Dynamical temperature response effect for season JJA of the simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO. The dynamical effect is calculated as the difference between the temperature response in the regular simulations (Δ T(SX-REF) with X either 2 or 5) and the sum of the individual contributions of CH₄, H₂O and O₃ to the adjusted stratospheric temperatures (Δ T_{addst}(SX*-REF*) with X either 2 or 5).



Figure S16. Dynamical temperature response effect for season SON of the simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO. The dynamical effect is calculated as the difference between the temperature response in the regular simulations (Δ T(SX-REF) with X either 2 or 5) and the sum of the individual contributions of CH₄, H₂O and O₃ to the adjusted stratospheric temperatures (Δ T_{addst}(SX*-REF*) with X either 2 or 5).



Figure S17. Absolute differences of zonal wind for season DJF of the sensitivity simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO and their respective reference in m s⁻¹. Non-stippled areas are significant on the 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S18. Absolute differences of zonal wind for season JJA of the sensitivity simulations (a) S2 fSST, (b) S5 fSST, (c) S2 MLO, (d) S5 MLO and their respective reference in m s⁻¹. Non-stippled areas are significant on the 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of REF MLO.



Figure S19. Relative differences of the annual zonal mean O_3 mixing ratio of REF MLO with respect to REF fSST in %. Non-stippled areas are significant on a 95 % confidence level according to a two sided Welch's test. The solid black line indicates the climatological tropopause height of the REF MLO.

Table S1. List of reactions, for which reaction coefficients changed from MESSy version 2.52 (fSST) to MESSy version 2.54.0 (MLO) in submodel Module Efficiently Calculating the Chemistry of the Atmosphere (MECCA; Sander et al. (2011a)).

Reaction	Reference for Reaction Rate Coefficient	
	fSST	MLO
$\mathrm{HO}_{2} + \mathrm{HO}_{2} \rightarrow \mathrm{H}_{2}\mathrm{O}_{2} + \mathrm{O}_{2}$	Christensen et al. (2002), Kircher	Burkholder et al. (2015)
	and Sander (1984)	
$NO_3 + NO_2 \rightarrow N_2O_5$	Sander et al. (2011b)	Burkholder et al. (2015)
$N_2O_5 \rightarrow NO_3 + NO_2$		
$NO_2 + HO_2 \rightarrow HNO_4$	Sander et al. (2011b)	Burkholder et al. (2015)
$\rm HNO_4 \rightarrow \rm NO_2 + \rm HO_2$		
$\text{ClO} + \text{ClO} \rightarrow \text{Cl}_2\text{O}_2$	Atkinson et al. (2007)	Burkholder et al. (2015)
$\rm Cl_2O_2 \rightarrow \rm ClO + \rm ClO$	Atkinson et al. (2007), Sander et al.	
	(2011b)	
$N_2O + O(^1D) \rightarrow 1.0 \text{ LossO3O} + 1. \text{ LossO3} + 2 \text{ NO}$	Sander et al. (2011b)	Burkholder et al. (2015)
$N_2O + O(^1D) \rightarrow 1.0 \ \text{LossO3O} + 1. \ \text{LossO3} + N_2 + O_2$		
$\label{eq:Clock} ClO + CH_3O_2 \rightarrow 1.0 \ LossO3Cl + 1. \ LossO3 + HO_2 + Cl +$	Sander et al. (2011b)	Burkholder et al. (2015)
НСНО		
$CH_3Cl + OH \rightarrow 1.0 \text{ ProdLCl} + LCARBON + H_2O + Cl$	Sander et al. (2011b)	Burkholder et al. (2015)
$CH_3CCl_3 + O(^1D) \rightarrow 3.0 \text{ ProdLCl} + 1.0 \text{ LossO3Cl} + 1.\text{LossO3}$	extrapolation from reactions	Burkholder et al. (2015)
+ 2 LCARBON + OH + 3 Cl	with CH_3CF_3 , CH_3CClF_2 and	
	CH ₃ CCl ₂ F from Sander et al.	
	(2006)	
$CH_3Br + OH \rightarrow 1.0 ProdLBr + LCARBON + H_2O + Br$	Sander et al. (2011b)	Burkholder et al. (2015)
$CHBr_3 + OH \rightarrow 3.0 \text{ ProdSBr} + LCARBON + H_2O + 3 \text{ Br}$	Sander et al. (2011b)	Burkholder et al. (2015)
$CH_2ClBr + OH \rightarrow 1.0 ProdSBr + LCARBON + LCHLORINE$	Sander et al. (2011b)	Burkholder et al. (2015)
+ H ₂ O + Br		
$HNO_3 + OH \rightarrow 1. ProdO3 + H_2O + NO_3$	Sander et al. (2011b)	Dulitz et al. (2018)

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