



Supplement of

Implication of strongly increased atmospheric methane concentrations for chemistry–climate connections

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Contents

1	Evaluation of reference with observations	2
2	Difference in the annual cycle of O₃	5
3	Climatological annual cycle of O₃ total column change	7
5	4 Adjusted temperature	9

1 Evaluation of reference with observations

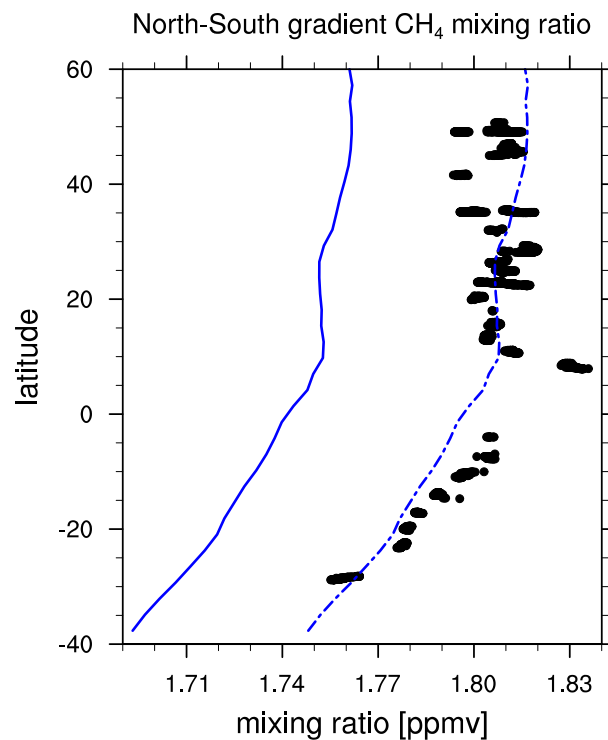


Figure S1. Comparison of observations on the research vessel *Polarstern* Klappenbach et al. (2015) (black) to annual zonal mean methane (CH₄) columns of the reference simulation (blue) in [parts per million volume (ppmv)]. The solid line represents the original column derived from the reference simulations and the dash dotted line are the columns moved by +0.055ppmv (see text for explanation).

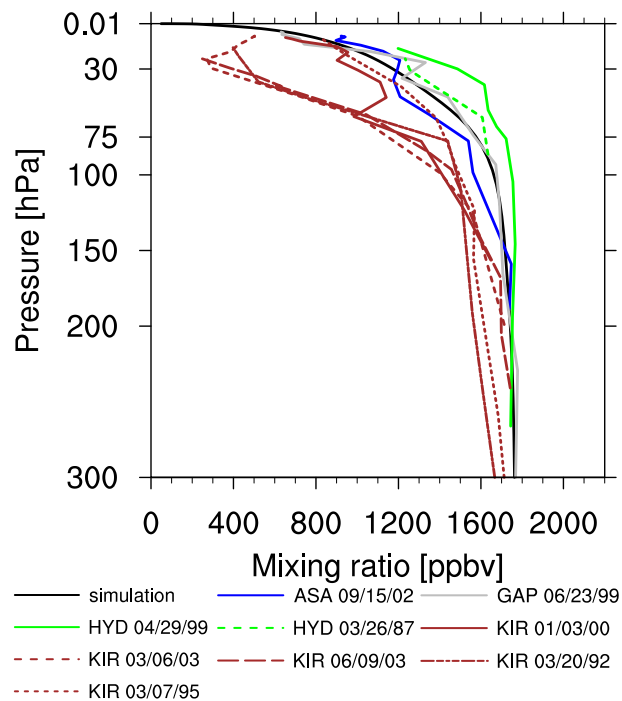


Figure S2. Comparison of vertical global mean CH₄ profile of simulation with balloon borne observations provided by Röckmann et al. (2011). The balloon launch sites are Hyderabad, India (HYD, 17.5° N, 78.60° E), Kiruna, Sweden (KIR, 67.9° N, 21.10° E), Aire sur l'Adour, France (ASA, 43.70° N, -0.30° E) and Gap, France (GAP, 44.44° N, 56.14° E) (see text for explanation).

2 Difference in the annual cycle of O₃

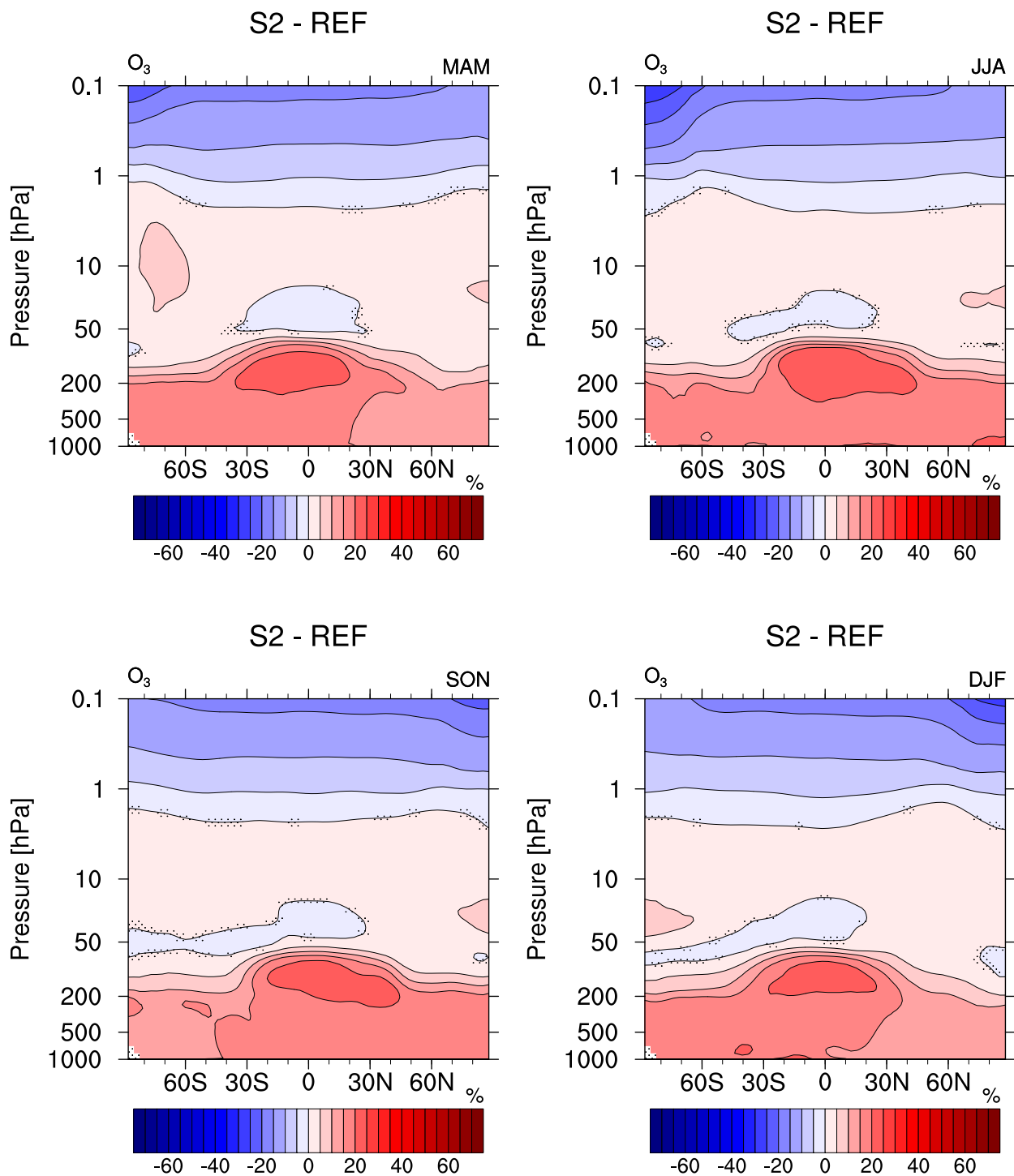


Figure S3. Seasonal differences in ozone (O₃) between S2 and REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

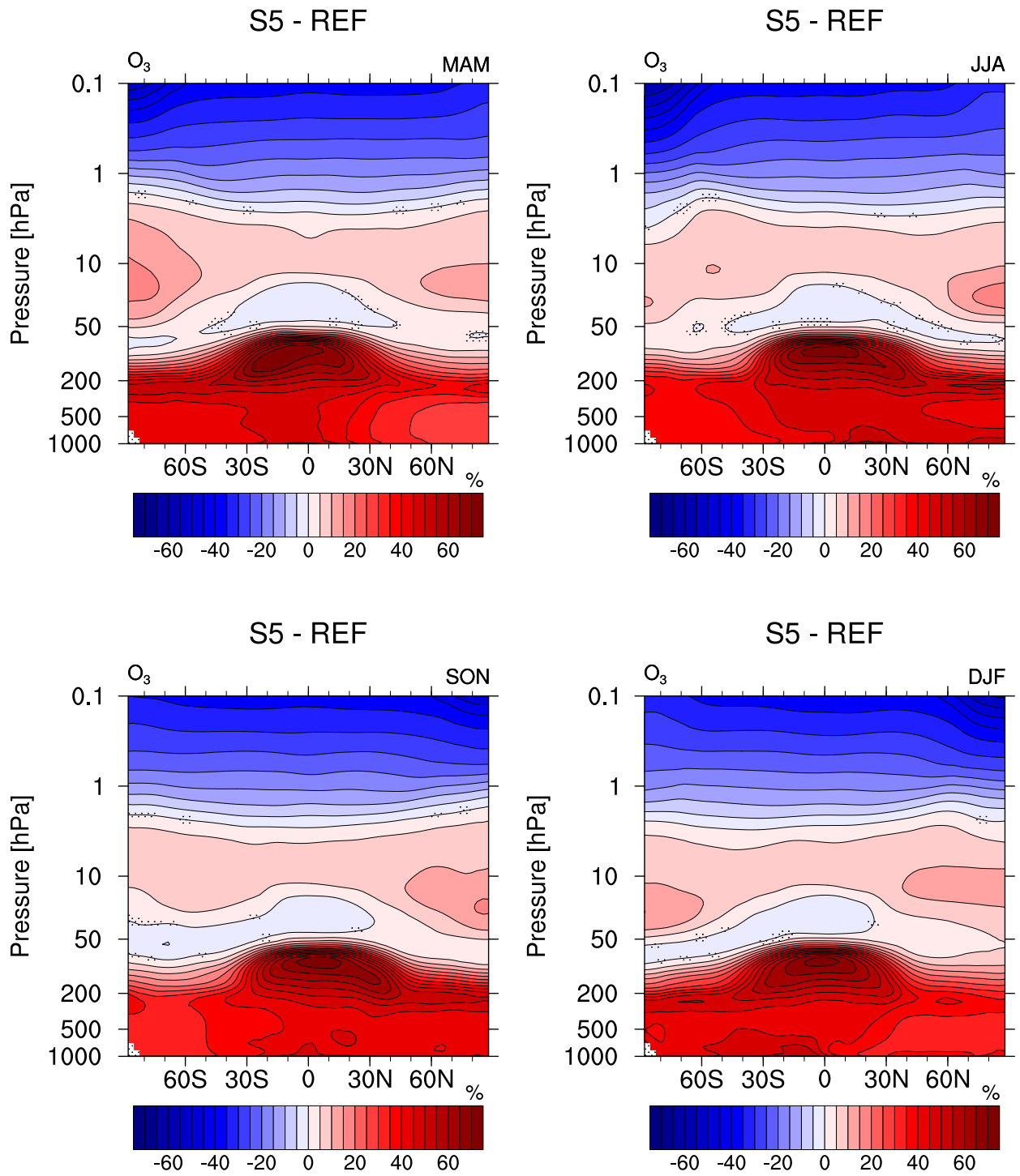


Figure S4. Seasonal differences in O₃ between S5 and REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.

3 Climatological annual cycle of O₃ total column change

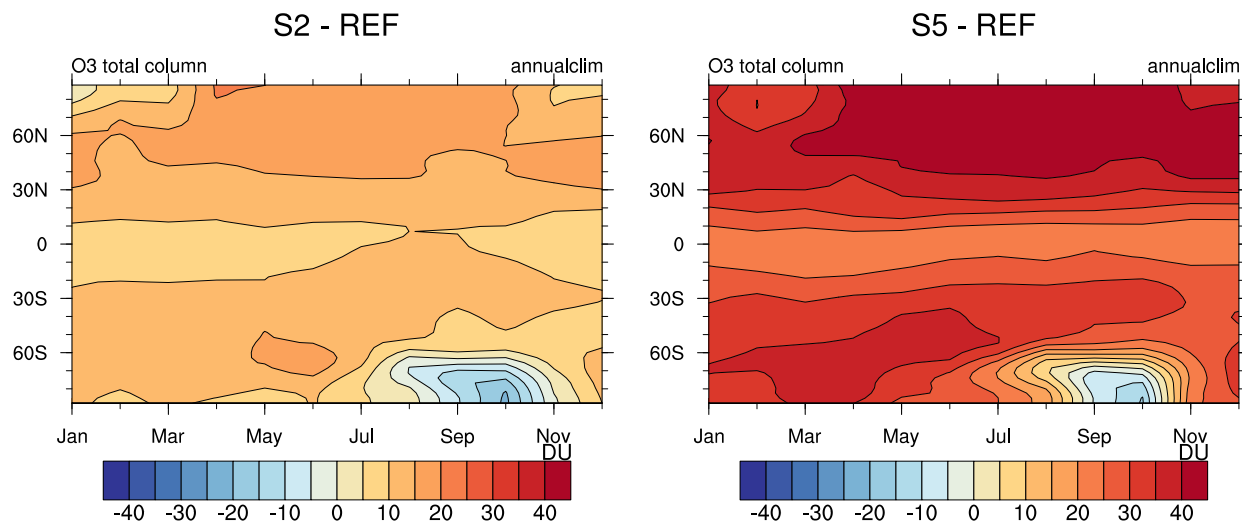


Figure S5. Climatological annual cycle of the total O₃ column change in Dobson Units (DU).

4 Adjusted temperature

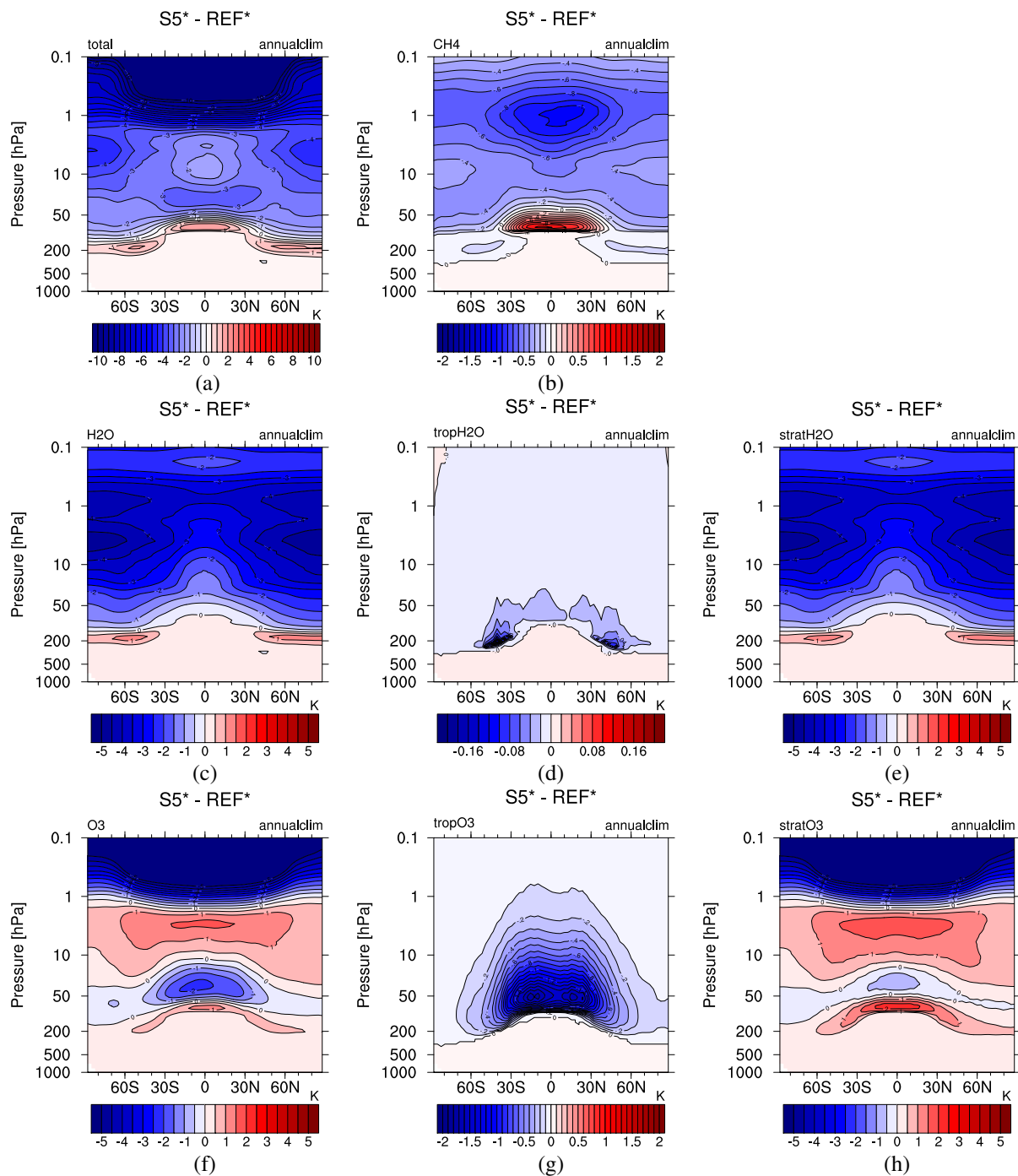


Figure S6. Stratospheric adjusted temperature based on chemical changes in simulation S5* (5xCH₄) in (a) CH₄, water vapour (H₂O) and O₃ together, (b) CH₄, (c) H₂O, (d) tropospheric H₂O only, (e) stratospheric H₂O only, (f) O₃, (g) tropospheric O₃ only, (h) stratospheric O₃ only. Note the different color bars in panels (a), (b), (d), and (g).

References

- Klappenbach, F., Bertleff, M., Kostinek, J., Hase, F., Blumenstock, T., Agusti-Panareda, A., Razinger, M., and Butz, A.: Accurate mobile remote sensing of XCO_2 and XCH_4 latitudinal transects from aboard a research vessel, *Atmos. Meas. Tech.*, 8, 5023–5038, <https://doi.org/10.5194/amt-8-5023-2015>, <https://www.atmos-meas-tech.net/8/5023/2015/>, 2015.
- 5 Röckmann, T., Brass, M., Borchers, R., and Engel, A.: The isotopic composition of methane in the stratosphere: high-altitude balloon sample measurements, *Atmos. Chem. Phys.*, 11, 13 287–13 304, <https://doi.org/10.5194/acp-11-13287-2011>, 2011.