

# **Supplement of: Implication of extreme atmospheric methane concentration for chemistry-climate connections**

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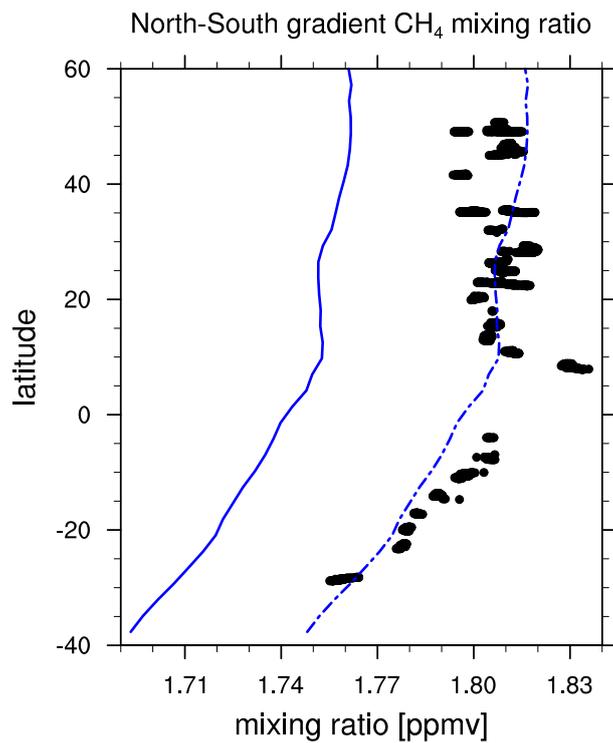
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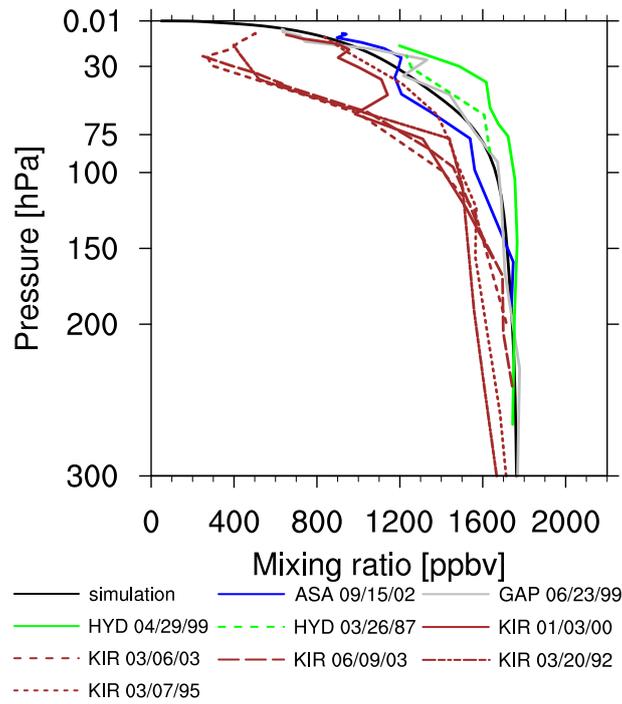
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## 1 Evaluation of reference with observations



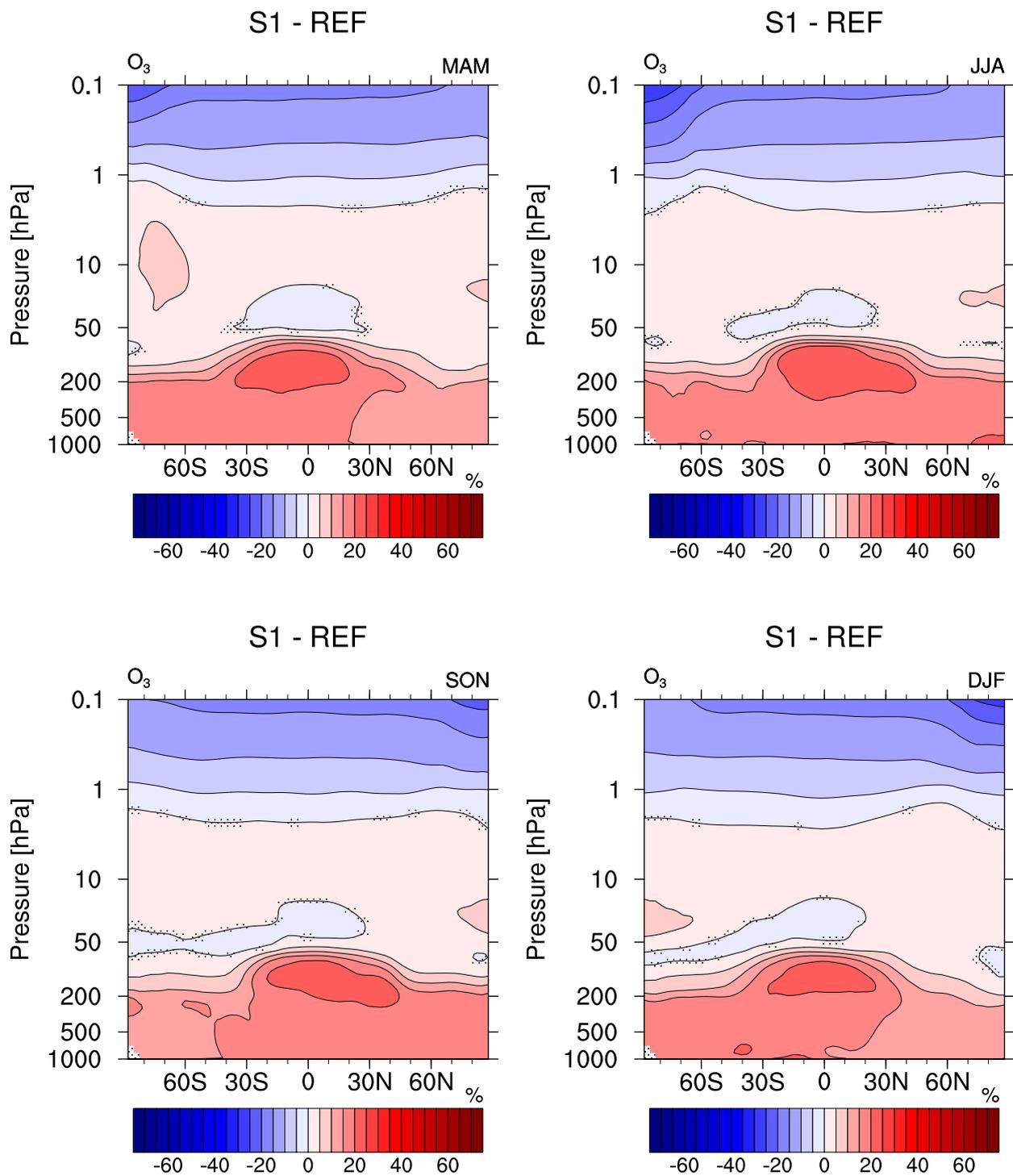
**Figure S1.** Comparison of observations on the research vessel *Polarstern* Klappenbach et al. (2015) (black) to annual zonal mean methane (CH<sub>4</sub>) columns of the reference simulation (blue) in [parts per million volume (ppmv)]. The solid line represents the original column derived from the reference simulations and the dash dotted line are the columns moved by +0.055ppmv (see text for explanation).



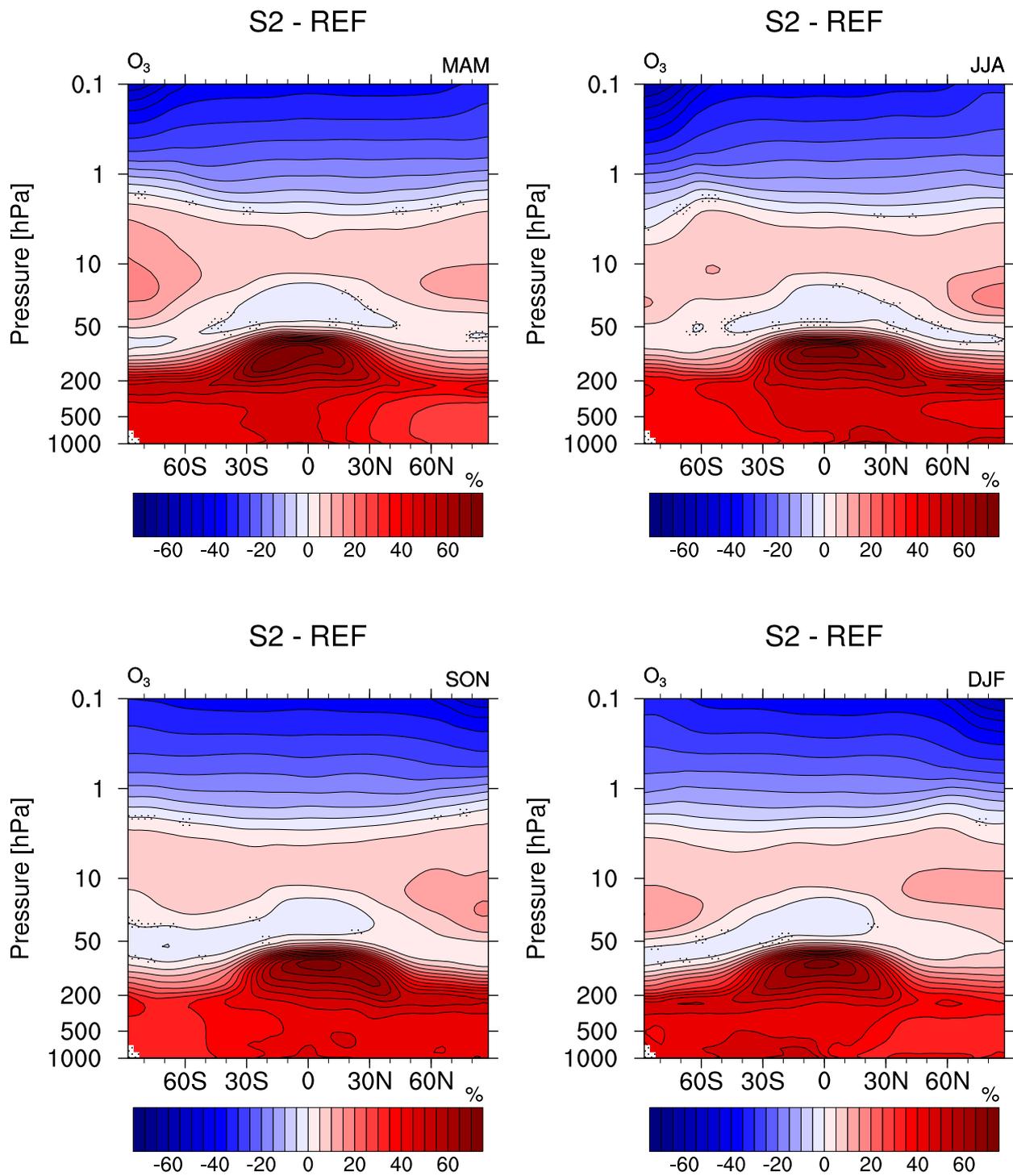
**Figure S2.** Comparison of vertical global mean CH<sub>4</sub> profile of simulation with balloon borne observations provided by Röckmann et al. (2011). The balloon launch sites are Hyderabad, India (HYD, 17.5° N, 78.60° E), Kiruna, Sweden (KIR, 67.9° N, 21.10° E), Aire sur l'Adour, France (ASA, 43.70° 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 O<sub>3</sub>



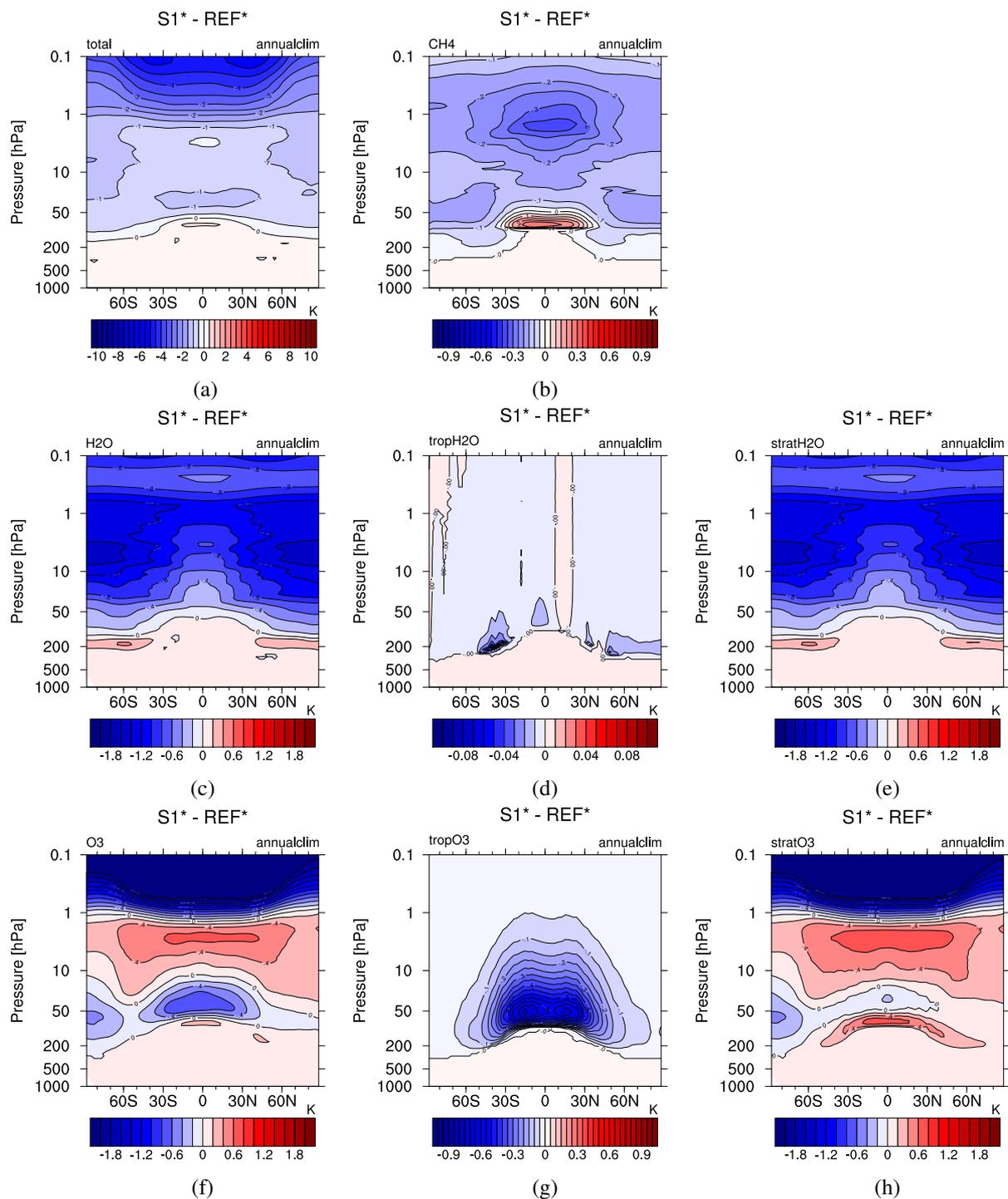
**Figure S3.** Seasonal differences in ozone (O<sub>3</sub>) between S1 and REF. Non-stippled areas are significant on a 95% confidence level according to a two sided Welch's test.



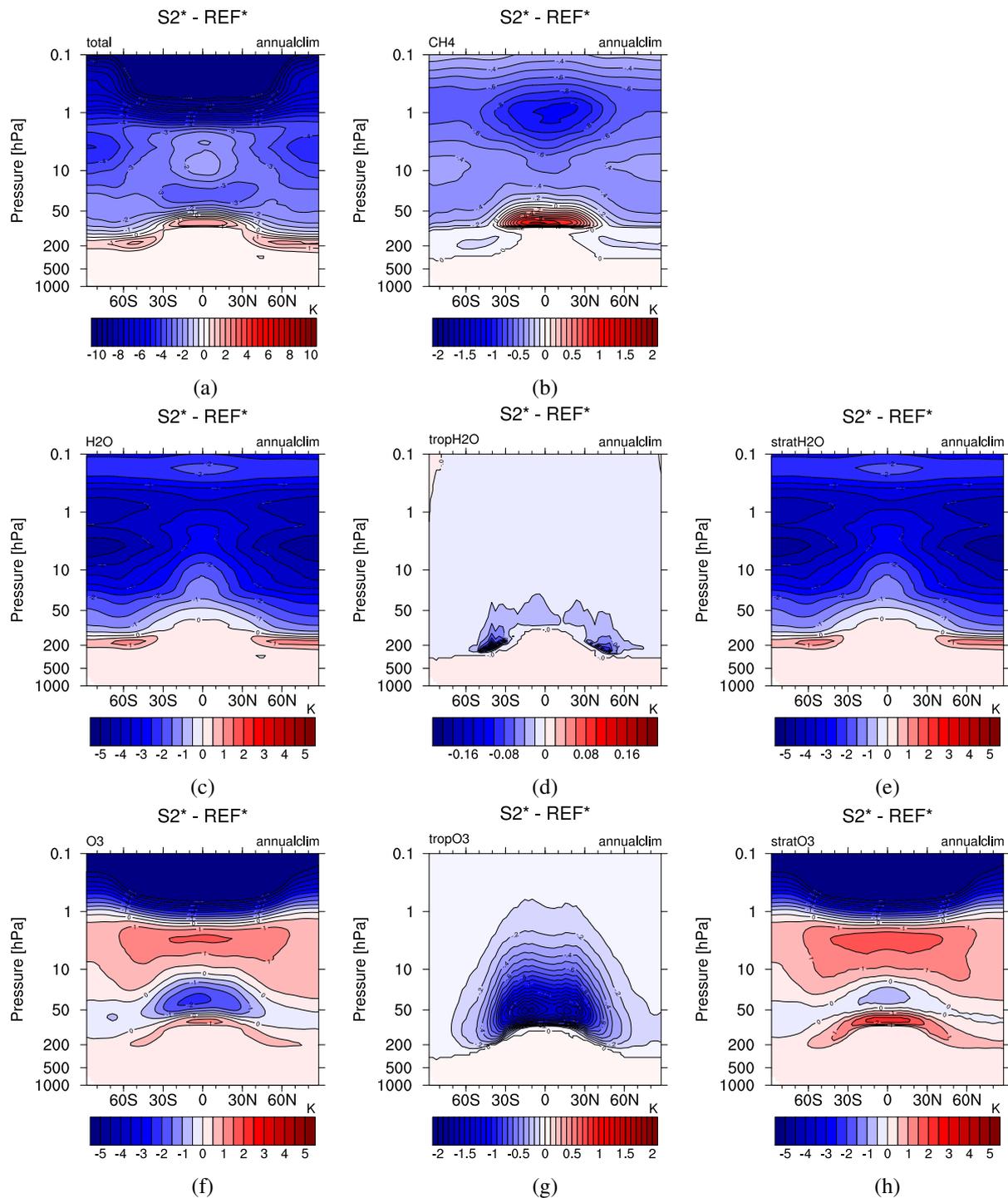
**Figure S4.** Seasonal differences in O<sub>3</sub> between S2 and REF. Non-stippled areas are significant on a 95% confidence level according to a two-sided Welch's test.



### 3 Adjusted temperature



**Figure S5.** Stratospheric adjusted temperature based on chemical changes in simulation S1\* (2xCH<sub>4</sub>) in (a) CH<sub>4</sub>, water vapour (H<sub>2</sub>O) and O<sub>3</sub> together, (b) CH<sub>4</sub>, (c) H<sub>2</sub>O, (d) tropospheric H<sub>2</sub>O only, (e) stratospheric H<sub>2</sub>O only, (f) O<sub>3</sub>, (g) tropospheric O<sub>3</sub> only, (h) stratospheric O<sub>3</sub> only. Note the different color bars in panels (a), (b), (d), and (g). **8**



**Figure S6.** Stratospheric adjusted temperature based on chemical changes in simulation S2\* (5xCH<sub>4</sub>) in (a) CH<sub>4</sub>, H<sub>2</sub>O and O<sub>3</sub> together, (b) CH<sub>4</sub>, (c) H<sub>2</sub>O, (d) tropospheric H<sub>2</sub>O only, (e) stratospheric H<sub>2</sub>O only, (f) O<sub>3</sub>, (g) tropospheric O<sub>3</sub> only, (h) stratospheric O<sub>3</sub> 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<sub>2</sub> and XCH<sub>4</sub> 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.