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Article:

Nowack, PJ, Abraham, NL, Maycock, AC et al. (5 more authors) (2015) A large ozone-circulation feedback and its implications for global warming assessments. Nature Climate Change, 5 (1). pp. 41-45. ISSN 1758-678X

https://doi.org/10.1038/NCLIMATE2451

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A large ozone-circulation feedback and its implications for global warming assessments

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Supplementary Information

Figure S1 | **Gregory regressions for the SW components.** This is an extension of Fig. 2 in the main text, showing the regressions for the remaining (SW) components. Differences in the slopes of these regressions are less distinct than for the LW components.



Figure S2 | Changes in the residual mean vertical velocity. Shown are zonal mean values during Boreal winter at 70hPa averaged over the last 50 years of each experiment. The figure indicates an acceleration of the Brewer-Dobson circulation with increasing surface temperature from A to B to C1 at an altitude where major decreases in ozone in B occur (Fig. 3a in the main text). Shading marks ± 2 times the standard deviation for each experiment.



Figure S3 | **Relative water vapour changes.** Shown are annual and zonal mean percentage differences between experiments B and C1 relative to the climatology of C1 averaged over the last 50 years of each experiment. The relatively, and in terms of surface temperature change disproportionally, drier stratosphere in experiment B is a result of the lower tropical cold trap temperatures induced by changes in ozone which are not included in experiment C1 (Fig. 3 in the main text).



Figure S4 | **TOA effective forcing due to relative changes in stratospheric water vapour and ozone.** The radiative transfer calculations were carried out as described in Methods. The differences in ozone (Fig. 3a in the main text) affect both SW and LW radiative fluxes significantly. However, the impact on the LW flux is larger, in particular in 30°N-30°S.



Figure S5 | **Regional Gregory regressions for the CRE-LW component.** Displayed are regressions of regional CRE-LW TOA radiative imbalance changes in **a** 50°N-50°S, **b** 50°S-90°S and **c** 50°N-90°N against global mean surface temperature change, following the method proposed by Boer and Yu²³. Changes in $\alpha_{cre,lw}$ are mainly confined to 50°N-50°S where major cirrus cloud changes between B and C1 occur (Fig. 4 in the main text and Fig. S6).



Figure S6 | The mechanism behind the longwave cloud changes. Zonal and annual mean frozen cloud fraction per unit volume multiplied by factor 100 in 50°N-50°S where the deviations in $\alpha_{cre lw}$ are found. The calculations are based on the last 50 years of each experiment. The shading shows the cloud fraction differences between the experiments: C1 minus B in a and B minus A in b. Contour lines (interval 2.5) denote the climatology of B in a and of A in b. Non-significant differences using a two-tailed Student's t-test at the 95% confidence level and regions where the cloud abundance in both runs is smaller than 5‰ are hatched. The differences in cirrus clouds in the upper troposphere and lower stratosphere (UTLS) in a are of opposite sign to the expected trend under increasing surface temperature shown in **b** (where surface temperatures in experiment C1 are higher than in B, which itself is higher than in A). Therefore, the cloud changes in the UTLS do not follow a simple surface temperature correlation from A to B to C1, but show a clear dependence on the impact of the differences in ozone between experiments B and C1 on the UTLS temperature. Note that the changes in the cloud abundance also oppose the higher atmospheric water vapour content in experiment C1 than in B. meaning that more clouds are formed in B despite less water vapour being available to form clouds. This, in combination with the fact that the cirrus cloud changes conform with the sign of the differences in $\alpha_{cre,lw}$ (high-altitude cirrus clouds exert a positive LW feedback, resulting in a more positive $\alpha_{cre,lw}$), consolidates their significance relative to other factors^{25,26}.