Response to reviewers

We thank both reviewers for their useful comments on our paper. Please note that all line numbers refer to the version with *tracked changes*. Many of the comments below reproduce our previously uploaded responses to the reviewers' comments. This document, however, provides more detailed information about how we have modified our manuscript.

Reviewer #1

"1. Why the latitudinal bands of 30-50 are left out? It covers a considerably large area, and may be more subjective to the horizontal mixing than the polar region. Even it may be messy and don't show an as good consistency among forcing agents and across models as the polar regions or the tropical region, it still worth reporting. Furthermore, the 50S-90S may not be a good representation of the Southern Hemisphere extratropics. This is because many models suffer a too strong southern polar vortex and hence the simulated southern polar stratosphere is too isolated. This can be hinted from Fig. 4a and Fig. S4, where a clear barrier is seen near 60S."

We first note that the response of ΔSWV_{slow} to surface temperature as a function of latitude is plotted in Fig. 4a of the submitted manuscript. We also reported the regression slope of ΔSWV_{fast} vs cold point temperature fast response in Fig. 4b of the submitted manuscript.

However, to more clearly answer the reviewer's question, we have included results at 200 hPa between 30°N and 50°N in the revised supplement. Figure S1 in the revised supplement shows the equilibrium ΔSWV_{slow} and ΔSWV_{fast} and their contribution to the total equilibrium ΔSWV for water vapor averaged at 200 hPa 30°N-50°N and 30°S-50°S. Figure S2 in the revised supplement shows the slope of ΔSWV_{slow} annual mean time series vs surface temperature time series for water vapor averaged at 200 hPa 30°N-50°N and 30°S-50°S.

In the revised manuscript, we mentioned these results in lines 180-184 and lines 222-223. Our major conclusions remain the same: The slow response plays a dominant role and contributes to close to 100% of the total response for most perturbations; The sensitivity shows general agreement across different perturbations.

"2. The regression method to get the equilibrium water vapor response seems to be unnecessarily complicate, especially the results are not too different from the simple average of the last 30 years. The authors first fit the radiative flux and water vapor time series with an exponential function, then regress the last 30 years of the fitted function. All these fitting and regression have potential introduce artificial biases and uncertainties. Recent studies also show that the ECS from the Gregory method may not be a good estimate of the true ECS (e.g. Winton et al. 2020). In addition, without a sufficiently long simulation, one can not validate whether the "equilibrium" from the regression is the true equilibrium. It makes more sense to me to simply use the average of the last 30 years while acknowledging that the models have not fully reached the equilibrium."

In an early draft of this manuscript, we approximated equilibrium ΔSWV using averages of the last 30 years of the runs. However, we analyzed one model that was run for 2600 years and found that the last 30 years of a 100- or 150-year run significantly underestimated the equilibrium. Thus, we developed the method that we presently use in the paper to better produce equilibrium estimates and validated it in the 2600-year model run, which is close to its equilibrium climate state. Details of this validation are described in lines 130-140 in the revised manuscript.

However, in response to this comment, we have listed slow response estimated by averaging over the last 30 years of the coupled simulation in Table S2 of the revised supplement.

"3. It may be worth pointing out how the PDRMIP model ensembles relate to the CMIP5 ensembles. From Fig. 2b, it seems that all of these models except HadGEM3 are on the weaker side of the CMIP5 ECS estimation range. I am also surprised to see that these models do not show an more distinct efficacy among different forcing agents (Hansen et al. 2005)."

We have added a statement to the revised manuscript comparing the PDRMIP models' ECS to that in the CMIP5 ensemble in lines 64-65.

As far as forcing efficacy goes, Hansenet al. (2005) also pointed out that efficacies depend on the method of which radiative forcing is defined. A more recent paper by Richardson et al. (2019) (which we already referenced in the submitted manuscript) using PDRMIP data showed that forcing efficacies calculated from effective radiative forcing have values close to one. Our results are in good agreement with Richardson et al. (2019) (Table S3 in the revised supplement).

"4. The authors relate the slow response to the surface temperature and relate the fast response to the cold point temperature. I believe the slow response would also be regulated by the cold point temperature. It may be interesting to show that if the relationship between the stratospheric water vapor and the cold point temperature holds from the fast adjustment to the slow response."

It certainly may be the case that the slow response is mediated by TTL temperatures, but by no means is that certain. Dessler et al. (2016) showed that, in two climate models, at least, a significant fraction of the long-term trend (and slow response) was due to increases in convective moistening, which bypasses the TTL cool trap.

We have done analyses testing whether the PDRMIP models and experiments show agreement for the relation between ΔSWV_{slow} vs the CPT slow response (Figure R1 below). Results from the models and experiments show good agreement. The slope is 0.72 ppmv/K, which is larger than the slope obtained from the fast response. Nevertheless, correlation does not prove causality and this result could arise from either TTL control or if convective moistening also correlates

with the CPT slow response, or some combination. We have added a sentence to the revised manuscript describing this analysis in lines 343-348.

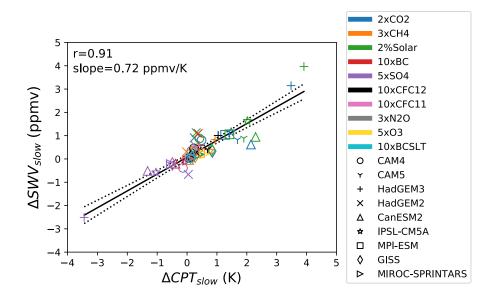


Figure R1: Same as Figure 6a of the submitted manuscript, but for TLS SWV slow response vs the CPT slow response.

"5. While Fig. 3 shows a consistent relationship between stratospheric water vapor and global mean surface temperature across various forcing, the temperature sensitivity does not seem to be so consistent in Fig. S4. Much more stratospheric moistening is seen in response to the solar forcing than others given the same surface temperature warming. This discrepancy needs to be resolved."

We list the regression slopes in the unit of ppmv/K in Table S4 and slopes in the unit of %/K in Table S5 in the revised supplement. The slope values in Table S4 are the same as we have shown in Fig. 3 of the submitted manuscript. It may not be clear in Fig. 3 of the submitted manuscript, but it is clear in Table S4 that the sensitivities are indeed larger in some experiments, such as the 2%Solar experiment. This is also the same for slopes in the unit of %/K in Table S5.

"Line 85-86: How does the averaged of fixed SST with baseline atmosphere compare to the average of the coupled baseline simulations."

For TLS SWV, the difference between fixed SST baseline simulation and coupled baseline simulation is on the order of 0.01 - 1 ppmv. For LMS SWV, the difference between fixed SST baseline simulation and coupled baseline simulation is on the order of 0.1 - 1 ppmv.

The results are averaged over the entire period of the baseline simulations for both fixed SST run and coupled run.

"Line 96: $y=c+ab^x -> y=c+ab^(-x)$ "

We have updates this (line 119).

Line 101: Fig. S1 was not showing what is stated here. It seems the intended Fig. S1 is missing.

Line 147: Fig. S2-4. -> Fig. S1-3

Line 167: Fig. S5 -> Fig. S4"

We have updated figures and figure numbers in the supplement.

"Line 191: Does the long wave effect of the tropospheric ozone also contribute?"

Yes, the tropospheric ozone has the long wave radiative effect. We have edited the text in the revised manuscript in lines 278-280.

Reviewer #2

General comments

1. "The paper largely focuses on interpreting the multi-model responses. While this is of course useful, it stops short of relating the new understanding to any real-world changes in SWV... How much does this work help in understanding past and possible future SWV changes?"

"In particular, note there has been some discussion of how the PDRMIP BC perturbations compare to observations (Allan et al, https://doi.org/10.1038/s41612-019-0073-9..."

We have added a new figure (Figure 7) and table (Table 2) and associated discussion to the paper (The "4. Historical changes in SWV" section). In this section (lines 366-400), we use our results to estimate observed changes in SWV and compare those to observations. Our estimate shows reasonable agreement with observed trend over 1980-2010.

2. "As noted in the specific comments, I feel that there is inadequate recognition that some of the results presented here are also presented, either explicitly or implicitly, in some earlier papers from the PDRMIP group – this is particularly so for the ERFs where no reference to, or comparison with, those earlier results, is given."

Thanks for pointing this out. We have added references to related results from earlier PDRMIP studies in the revised paper.

Please also see the responses to specific comments below related to previous PDRMIP studies:

"47-48: There is a slight overlap between this submitted paper and the paper published in ACP by the core PDRMIP team – Hodnebrog et al: https://doi.org/10.5194/acp-19-12887-2019, which is not cited here..."

Lines 53-55, 94-95, 374-375.

"129: I think it is necessary that a comparison of ERFs (and the associated feedback parameter) with Richardson et al. (including for the CFCs and N2O in their supplement) is presented both to confirm they are in reasonable agreement and also to make clear that the ERFs derived here are not original work with the PDRMIP output."

We have added a Table S3 in the revised supplement comparing our ERFs with "ERF_{sst}" from Richardson et al. (2019). The comparison shows good agreement. Texts mentioning this comparison are in lines 151-158 of the revised manuscript.

Specific comments

"14: This conclusion is specific to the TLS"

Yes, we agree with this, although the cold point temperature does have *some* influence in the lowermost SWV (Dessler et al., 1995). But the control is not as strong as that in the TLS (see Fig. 6b-c in submitted manuscript) and the lowermost SWV is controlled by multiple factors. We have edited the text to make this clearer (lines 13-14).

"16: "becomes weaker at higher altitudes and at higher latitudes below 150 hPa." This is a bit ambiguous. Does this means heights at pressures below 150 hPa or heights below the height of the 150 hPa surface. These would have opposite meanings."

It means altitudes below the 150 hPa surface. We have edited the text for clarity (line 16).

"57: Presumably the 3xCH4 experiments have no resulting change in SWV due to the oxidation of additional methane?"

Yes, indirect chemical effects are not included in the 3xCH₄ experiment. We have added a sentence saying this (line 77-78).

"90 and many other places: There are repeated statements that there is no surface temperature response in the fixed SST runs, but this is not correct, with implications for the definition of ERF."

Yes, the reviewer is correct that land surface temperatures can respond to the forcing. We have edited the text in the revised manuscript (lines 108-110).

"139: tend to be larger" Isn't it clearly larger?

Yes, the reviewer is correct. We have edited this text in the revised manuscript (line 175).

"*148 and throughout: Rather little is said about intermodel differences. For example, on HADGEM3, more discussion of its apparent outlier status on some plots seems necessary. The text says it is "likely connected" to the larger surface warming, but it seems the climate sensitivity is about double the multi-model average but the slow SW response is around a factor of 4 larger. Is that because the TTL temperature change is 4 times higher (per unit ERF)?"

To answer the question about the HadGEM3 model, Figure R2 below shows the equilibrium slow response of TTL temperature (ΔT_{slow}) per unit ERF. The HadGEM3 ΔT_{slow} /ERF is between 2.64-3.97 times the multi-model mean ΔT_{slow} /ERF for experiments 2xCO₂, 3xCH₄, 2%Solar, 10xBC, and 10xCFC-12. Since the surface warming in HadGEM3 is larger than all other models, its upper tropospheric warming is also largest. Longwave radiation emitted from the upper troposphere warms the TTL level (Lin et al., 2017), so the larger upper tropospheric warming in HadGEM3 also results in larger TTL heating than other models. The relationship between surface warming and TTL warming is not linear.

That said, we cannot conclusively identify a cause given the information archived. So we have removed the claim that the difference is likely connected to surface warming and we have added a sentence saying more work on the causes of these differences is warranted (lines 189-190).

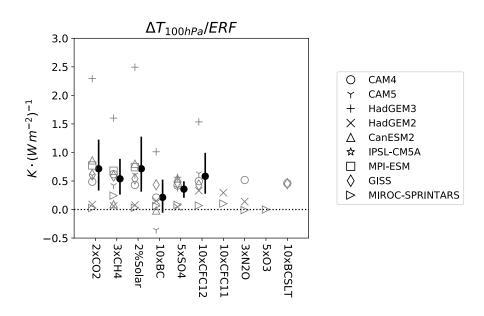


Figure R2: Equilibrium slow response of TTL temperature (100 hPa, averaged between 30°N-30°S) per ERF for all models and perturbations.

"Another example is that apparently half the models have a slow SW response to BC of the opposite sign (Fig 1a) to the multi-model mean. Is there any obvious reason why? As far as I can see BC causes a warming in all models."

Figure R4 below shows the vertical profile of tropical temperature slow (a) and fast (b) responses per unit ERF for the 10xBC experiment. The 10xBC does cause a warming at the surface and in the troposphere due to a positive TOA ERF in all models. In the TTL and lower stratosphere (LS), however, the heating is mainly caused by the fast adjustment (Fig. R3b below). The slow temperature response in the TTL is the residual of the total response minus the fast adjustment, which is negative or close to zero (Fig. R3a below).

It is therefore our contention that some of these negative values are artifacts of the method we use to estimate equilibrium response. Support for this comes from Fig. 3 of the paper. The values in this figure come from regressions of ΔSWV_{slow} vs. ΔTs in the BC runs. This method does not require differencing two large numbers, so we feel it is more robust. It shows that most models have a positive response of SWV due to BC-induced warming. For those models that produce negative slopes for ΔSWV_{slow} vs. ΔTs in the BC runs, there is large uncertainty in the regression, because the surface temperature change in those models are small.

We have noted this explanation in the revised manuscript (lines 191-196).

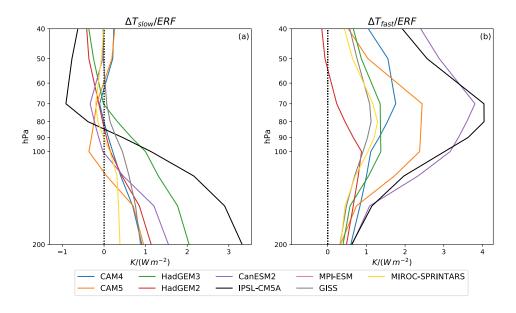


Figure R3: Profiles of equilibrium slow (a) and fast (b) temperature response for the 10xBC experiment, normalized by ERF (K·(Wm⁻²)⁻¹), and averaged over 30°N-30°S. The color coding indicates results from different models.

"One thing I miss from this study, and encourage the authors to look at if they have the resource, is the degree to which the model's background climatology of stratospheric water vapor or TTL temperature could explain some of the intermodel differences."

We have investigated the SWV in the fixed SST baseline simulations. Based on our analyses, the baseline climatology SWV does not explain the inter-model differences in the responses to forcing agents. As an example, Fig. R4 below shows the TLS SWV slow response (first row) and TTL temperature slow response (second row) vs. the baseline TLS SWV climatology and baseline TTL temperature climatology. We omitted $3xN_2O$, $5xO_3$, and 10xBCSLT, because fewer than three models performed these experiments. There is no correlation between the SWV and temperature slow responses and the baseline climatology. In particular, HadGEM3 produces extremely large slow responses for most experiments, however, in Fig. R4 below, its baseline SWV and temperature climatology is not the largest among the models.

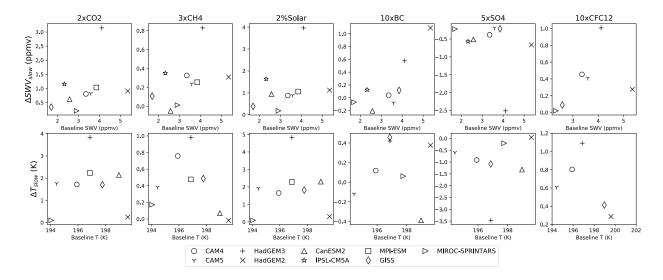


Figure R4: Top row: The TLS SWV slow response (ppmv) vs. the baseline TLS SWV climatology (ppmv). Bottom row: The TTL temperature slow response (K) (100 hPa, averaged over 30°N-30°S) vs. the baseline TTL temperature climatology (K) (100 hPa, averaged over 30°N-30°S). The baseline climatology is obtained from the fixed SST simulations averaged over the last 10 years.

"156: Is this linear regression done once across all simulations and all perturbations. If not, I am unclear which perturbations have been used for the regression."

We have added a sentence "We do this regression for each model and perturbation separately" to avoid confusion (lines 202-203).

"159: This is a relatively short paper and I wondered whether the supplementary figures could be brought into the main text?"

It remains our opinion that the key figures are included in the paper. Thus, in order to keep the take-home message concise, we have left the content of the supplement unchanged.

"171-172: This repeats a point already made at 141-142."

We'll have removed the repeated text.

"201: I am sorry if I miss it, but I see very little discussion of stratospheric temperature changes in the Jain et al paper. The role of CFCs on the vertical profile of temperature can be seen in many papers such as Forster et al. https://doi.org/10.1007/s003820050182 and Forster and Joshi 10.1007/s10584-005- 5955-7..."

In the submitted paper where we discussed the radiative heating in the UTLS by CFCs, we were referring to the text in Section 3.3 of (Jain et al., 2000), where they stated that "Halocarbons absorb predominantly in the window region (750-1250 cm⁻¹), in the linear line limit; therefore in the stratosphere they absorb the upwelling radiation from the troposphere and increase the heating rate of the stratosphere".

We agree with the reviewer that it is useful to reference papers that explicitly investigated vertical temperature profiles forced by CFCs. We have added these references in the revised manuscript (line 272).

"*204: This statement on shortwave radiation is strange. There may be a small shortwave effect from the reduced reflected flux from the troposphere, but there is a long history of simulations that clearly attribute the stratospheric cooling due to increased tropospheric ozone to the decreased upwelling thermal infrared radiation. E.g. Ramaswamy and Bowen https://doi.org/10.1029/94JD01310, Berntsen et al https://doi.org/10.1029/97JD02226 and the Forster et al. paper referred to above."

Thanks for pointing this out. We have edited the text to say, "Increases of tropospheric O₃ (5xO₃) reduce the upwelling longwave radiation, which cools the stratosphere. The longwave radiation absorbed heat the TTL region" (lines 278-280). References are also added in lines 278-280.

"206: "Tropospheric O3 is also transported". As I understand it, ozone is imposed in the models and not advected. I don't know what this sentence means."

This is correct: In the 5xO₃ experiment, the PDRMIP group used 5 times the tropospheric ozone distribution (TROP) in the paper by MacIntosh et al. (2016) (line 76). We have removed the text about transport.

"212: "larger than 50%". CAM5 and MPI-ESM look less than 50%?"

Yes, this was poorly worded. We have completely re-written the paragraph in lines 274-277.

"*248: Returning to General Point#1, the Summary feels a very mechanical repetition of the results in the paper without any discussion of the wider implications, remaining uncertainties, or possible future avenues/priorities for improving understanding."

We added our discussion on wider implications and remaining uncertainties to "4. Historical changes in SWV" section in the revised manuscript (lines 366-400).

"273: Strictly Fig 5 refers to TLS only"

We have edited the text (line 427).

"519-520: I think the markers are only reported when there are more than 3 contributing models?"

Yes, the multi-model mean and error bars are shown for perturbations that are performed by more than three models. We have added this caveat to the revised figure captions.

"46L "responses" -> "responds""

We have modified the text (line 53).

"Throughout: This may be common usage, but the paper refers throughout to the ensemble mean when other papers would refer to it as the multi-model mean (ensemble could refer to different runs from the same model with perturbed initial conditions or physics..."

Thanks for pointing this out. To avoid confusion, we have replaced the "ensemble mean" with multi-model mean in the revised manuscript.

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The response of stratospheric water vapor to climate change driven by different forcing agents

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Abstract. We investigate the response of stratospheric water vapor (SWV) to different forcing agents within the Precipitation Driver and Response Model Intercomparison Project (PDRMIP) framework. For each model and forcing agent, we break the SWV response into a slow response, which is coupled to surface temperature changes, and a fast response, which is the response to external forcing, but before the sea surface temperatures have responded. Our results show that, for most climate perturbations, the slow SWV response dominates the fast response. The slow SWV response exhibits a similar sensitivity to surface temperature across all climate perturbations. Specifically, the sensitivity is 0.35 ppmv K-1 in the tropical lower stratosphere (TLS), 2.1 ppmv K-1 in the northern hemispheric lowermost stratosphere (LMS), and 0.97 ppmv K-1 in the southern hemispheric LMS. In the TLS, the fast SWV response only dominates the slow SWV response when the forcing agent radiatively heats the cold point region — for example, black carbon, which directly heats the atmosphere by absorbing solar radiation. The fast SWV response in the TLS is primarily controlled by the fast adjustment of cold point temperature across all climate perturbations. This control becomes weaker at higher altitudes in the tropics and altitudes below 150 hPa in the LMS.

1 Introduction

Stratospheric water vapor plays an important role in global climate change. It is an important greenhouse gas (GHG), which affects the Earth's radiative budget (Forster and Shine, 2002; Solomon et al., 2010), and it also plays an important role in stratospheric ozone chemistry (Solomon et al., 1986; Dvortsov and Solomon, 2001).

SWV in the overworld (above 380-K isentropic surface) (e.g. Hoskins, 1991) and SWV in the extratropical lowermost stratosphere (LMS, between the extratropical tropopause and the 380-K isentropic surface) (e.g. Holton et al., 1995) are distinguished according to different mechanisms that control them. Overworld SWV is primarily controlled by the temperatures in the tropical tropopause layer (TTL) as air is transported through it (e.g. Mote et al., 1996; Fueglistaler et al., 2009) and by production from oxidation of methane (e.g. Brasseur and Solomon, 2005). The LMS SWV is controlled by three major sources, including the transport of overworld air by the downward branch of Brewer-Dobson circulation, adiabatic quasi-horizontal transport from the tropical upper troposphere, and diabatic cross-tropopause transport due to deep convection

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35 (Dessler et al., 1995; Holton et al., 1995; Plumb, 2002; Gettelman et al., 2011).

The response of SWV to climate change can be partitioned into two components: the fast response and slow response. The addition of a radiatively active constituent to the atmosphere can influence the atmosphere even before the surface temperature changes, leading to changes in SWV. This is often referred to as an "adjustment" to the forcing, and is generally considered part of the external forcing (e.g. Sherwood et al., 2015). We will refer to this as the "fast response" of SWV to the forcing. The slow response is the component in the SWV change that is coupled to changes of the surface temperature, which occurs on longer time scales. This slow response means that SWV could be an important positive feedback to global warming (Forster and Shine, 2002; Dessler et al., 2013; Huang et al., 2016; Banerjee et al., 2019). Banerjee et al. (2019) have shown that, when CO₂ is abruptly quadrupled, the change in SWV mainly consists of the slow response and that the fast response is less important.

- Previous studies have shown that climate models, which are able to accurately reproduce observed interannual variations in SWV (Dessler et al., 2013; Smalley et al., 2017), robustly project a positive long-term trend in overworld SWV at entry level with a warming climate due to increasing GHGs (Gettelman et al., 2010; Dessler et al., 2013; Smalley et al., 2017). This is mainly due to a warmer tropopause (Thuburn and Craig, 2002; Gettelman et al., 2010; Lin et al., 2017; Smalley et al., 2017; Xia et al., 2019), which is controlled, to some extent at least, by the warming surface (Gettelman et al., 2010; Shu et al., 2011;
- 50 Dessler et al., 2013; Huang et al., 2016; Revell et al., 2016; Lin et al., 2017; Smalley et al., 2017; Banerjee et al., 2019). Dessler et al. (2016) suggested that increases in convective injection into the stratosphere due to a warming climate may also be contributing to the trend in entry SWV. In the LMS, the climate models show larger increases in SWV (Dessler et al., 2013; Huang et al., 2016; Banerjee et al., 2019). It is not known how SWV responds to different forcing agents. Hodnebrog et al. (2019) investigated the response of global integrated water vapor to different forcing agents, but focused on the troposphere.
- 55 The goal of this study is to investigate the response of both overworld and LMS SWV to forcing agents with different physical properties. We will explicitly investigate the fast and slow responses in SWV and compare them. We will also investigate how SWV responds to surface temperature change when the climate is forced by different forcing agents.

2. Method

2.1 The PDRMIP set-up

60 In this paper, we analyze nine models from the Precipitation Driver and Response Model Intercomparison Project (PDRMIP)

(Samset et al., 2016; Myhre et al., 2017; Tang et al., 2018, 2019). These are Coupled Model Inter-comparison Project phase 5

(CMIP5) era models (Table 1) and each performed a baseline and multiple climate perturbation experiments (Table 1). This subset of the CMIP5 ensemble has a multi-model mean equilibrium climate system (ECS) of 3.6 K, close to the ensemble-

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average ECS of the entire CMIP5 ensemble (3.3 K) (Zelinka et al. 2020).

In the perturbation experiments, perturbations on a global scale are applied abruptly at the beginning of the model simulation. The five core experiments include a doubling of CO₂ concentration (2xCO₂), a tripling of CH₄ concentration (3xCH₄), a 2% increase in solar irradiance (2%Solar), an increase of present-day black carbon concentration or emission by factor of 10 (10xBC), and an increase of present-day SO₄ concentration or emission by factor of 5 (5xSO₄). In addition to the five core experiments, a subset of models also performed additional perturbation experiments: an increase in CFC-11 concentration from 535 ppt to 5 ppb (hereafter, 10xCFC-11), an increase in CFC-12 concentration from 653.45 ppt to 5 ppb (hereafter, 10xCFC-12), an increase in N₂O concentration from 316 ppb to 1 ppm (hereafter, 3xN₂O), an increase tropospheric O₃ concentration used in MacIntosh et al. (2016) by factor of 5 (5xO₃), and an increase of present-day black carbon with shorter lifetime by factor of 10 (10xBCSLT). We note that indirect chemical effects are not included in the 3xCH₄ experiment. Table 1 provides details about the models and the perturbations each one simulated.

The perturbations in GHGs and solar irradiance are relative to the models' baseline simulations, in which the concentration of the GHGs and solar irradiance are either at present-day levels or pre-industrial levels. The perturbations in the aerosols depend on whether it is possible to prescribe aerosol concentrations in the models. For models that are able to prescribe aerosol concentrations, the aerosol perturbations are based on a multi-model mean baseline aerosol concentration in 2000 obtained from the AeroCom Phase II initiative (Myhre et al., 2013). For those that are only able to produce aerosols through emissions, the perturbation is applied by increasing the emissions by the factors listed above. The 10xBCSLT experiment is performed only by models that are able to prescribe aerosol concentrations.

Each perturbation experiment is performed in two configurations: a fixed sea surface temperatures simulation ("fixed SST") and a fully coupled (slab ocean for CAM4 only) simulation. The fixed SST simulations use the SST climatology at either present-day level or pre-industrial level. The fixed SST simulations are at least 15 years and the coupled simulations are at least 100 years.

2.2 Fast response and slow response

- 90 When available, SWV mixing ratio is obtained directly from the specific humidity output by each model simulation. For the models that do not output specific humidity (CAM5, GISS-E2-R, and MIROC-SPRINTARS), we calculate specific humidity by multiplying the models' relative humidity by the saturation mixing ratio with respect to ice calculated using model temperature and pressure. Responses of specific humidity and relative humidity in the PDRMIP have been investigated by Hodnebrog et al. (2019), but they focused on water vapor in the troposphere.
- 95 We define ΔSWV, the change in SWV mixing ratio in response to a particular perturbation, to be the difference between SWV in the perturbed coupled run and that in the baseline coupled run. As discussed above, the ΔSWV can then be broken down

into the two components: the fast response (ΔSWV_{fast}) and slow response (ΔSWV_{slow}). We compute results in the tropical lower stratosphere (70 hPa, 30°N-30°S, hereafter, TLS), in the northern hemispheric (NH) lowermost stratosphere (50°N-90°N at 200 hPa, hereafter, NH LMS), and in the southern hemispheric (SH) lowermost stratosphere (50°S-90°S at 200 hPa, hereafter, SH LMS). Most previous studies have focused on response of water vapor in the TLS (e.g., Gettelman et al., 2010; Shu et al., 2011; Smalley et al., 2017). But recent studies report that the climate is most sensitive to changes in water vapor in the LMS (Solomon et al., 2010; Dessler et al., 2013; Banerjee et al., 2019), so we also investigate that region.

We use the fixed SST simulations to get ΔSWV_{fast} , the <u>rapid adjustment</u> in SWV before <u>sea</u> surface temperature changes. ΔSWV_{fast} is the difference between the SWV mixing ratio averaged over the last 10 years in the fixed SST run with the forcing perturbation and the SWV mixing ratio averaged over the last 10 years in the fixed SST <u>baseline simulation</u>. The fixed SST runs have some warming of the land-surface, meaning that our fast response includes a contribution from warming land-surface. We expect this will have a small impact on our results, but it remains one of the uncertainties in our analysis.

We calculate ΔSWV_{slow} as ΔSWV minus ΔSWV_{fast}. To estimate the time series of ΔSWV_{slow}, we use annual mean ΔSWV over the entire coupled run period (at least 100 years) minus the ten-year average ΔSWV_{fast}. To estimate equilibrium ΔSWV_{slow}, we use a regression method similar to the methodology introduced by Gregory et al. (2004). The basic concept is that we regress the annual mean global average net downward radiative flux (R) at the top of atmosphere (TOA) against the annual mean ΔSWV averaged at TLS, NH LMS, or SH LMS. The equilibrium ΔSWV is where the linear fit intercepts at R=0. Then we simply subtract ΔSWV_{fast} from the equilibrium ΔSWV to estimate equilibrium ΔSWV_{slow}.

These regressions can be very noisy and yield highly uncertain parameters, particularly for perturbations with relatively small amounts of radiative forcing and warming. To account for this, we first fit the R and Δ SWV time series using an exponential function $(y(t) = b + a1 \cdot e^{-t/\tau 1} + a2 \cdot e^{-t/\tau 2})$, and then do the regression using the fitted time series. For fully coupled models, we constrain $\tau 1$ to be within the range of 4 ± 2 years and $\tau 2$ to be within the range of 250 ± 70 years; for CAM4, in which the atmosphere is coupled to a slab ocean, we constrain $\tau 1$ to be within the range of 4 ± 2 years. We then compute the best fit of all parameters. The ranges for the time constants are based on previous estimations of climate system time scales (Geoffroy et al., 2013). We estimate the Δ SWV-intercept at R=0 by regressing the fitted R and Δ SWV data over the last 30 years, since the relation between R and Δ SWV is not necessarily linear over the entire 100-year period. The slow and fast responses of other variables, such as global average surface temperatures and cold point temperatures are computed using the same method.

125 We tested this method in a climate model that nearly reaches the equilibrium climate state. We analysed runs of the fully coupled Max Planck Institute Earth System Model version 1.1 (MPI-ESM1.1) (Maher et al., 2019), which has a transient climate response and an effective climate sensitivity near the middle of the CMIP5 ensemble range (Adams and Dessler, 2019);

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Dessler, 2020). It includes a 2000-year preindustrial control run and a 2614-year abruptly quadrupled CO₂ run. The values of ΔSWV averaged over the last 30 years of the 4xCO₂ run relative to the control run are 4.60 ppmv in the TLS, 22.40 ppmv in the NH LMS, and 9.69 ppmv in the SH LMS. We expect this to be close to equilibrium ΔSWV because the trend in global average surface temperature over the last 500 years of the 4xCO₂ run is 0.02 K per century. We use the regression method to estimate the equilibrium ΔSWV using MPI-ESM1.1 water vapor mixing ratio time series over the first 100 years and obtain estimates of 4.38 ppmv in the TLS, 20.01 ppmv in the NH LMS, and 9.07 ppmv in the SH LMS; these yield differences of 0.22 ppmv in the TLS, 2.39 ppmv in the NH LMS, and 0.62 ppmv in the SH LMS. Thus, our method underestimates the true equilibrium value by 5% in the TLS, 11% in the NH LMS, and 6% in the SH LMS.

140 Uncertainty for slow and fast responses of different quantities shown in this paper are obtained from Monte Carlo samples as follows: For each perturbation, we randomly sample with replacement 100,000 times for each model that performed that perturbation and from these samples compute the 2.5%-97.5% percentiles.

3. Results

3.1 The slow stratospheric water vapor response

145 We show equilibrium ΔSWV_{slow} and its percentage contribution to the total equilibrium ΔSWV in Figure 1. We show results in the TLS (Figs. 1a and 1d), in the NH LMS (Figs. 1b and 1e), and the SH LM (Figs. 1c and 1f). In evaluating the absolute magnitude of ΔSWV_{slow} in the first column of Fig. 1, we normalize the equilibrium ΔSWV_{slow} using effective radiative forcing (ERF), so that differences in the magnitude of the forcing do not confound our results.

ERF values used in construction of Fig. 1 are plotted in Fig. 2a; they are calculated as the difference in net radiation at the top

of atmosphere (TOA) averaged over the last 10 years between the fixed SST perturbed and baseline simulation. Previous studies have computed the ERF in the PDRMIP using various methods (Richardson et al., 2019; Tang et al., 2019). Our calculation uses the same method as Richardson et al. (2019) "ERF_{sst}" and a direct comparison with Richardson et al. (2019) showing good agreement can be found in the supplement (Table S3). The equilibrium global averaged surface temperature changes (ΔTs), estimated using the regression method described in Section 2.2 and normalized by ERF, are plotted in Fig. 2b.
 The multi-model mean ΔTs/ERF shows general agreement across different perturbations. This quantity is the inverse of the feedback parameter λ (e.g. Dessler and Zelinka, 2015), so Fig. 2b implies that the climate sensitivity to these different perturbations is similar, which also agrees with Richardson et al. (2019). We list the ERF and ΔTs quantities for each model and perturbation in Table S1.

In each region, the magnitude of <u>multi-model mean</u> $\Delta SWV_{slow}/ERF$ shows general agreement for different perturbations. The magnitudes of $\Delta SWV_{slow}/ERF$ in the LMS <u>are</u> larger than those in the TLS (Figs. 1b-c). This is consistent with previous studies,

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which showed that the long-term trend in SWV over the century in climate models is largest near the LMS tropopause (Dessler et al., 2013; Huang et al., 2016; Banerjee et al., 2019). This reflects different transport pathways into the LMS, including the downward transport by the Brewer-Dobson circulation, quasi-horizontal isentropic mixing from tropical troposphere, and convective influence (Dessler et al., 1995; Holton et al., 1995; Plumb, 2002; Gettelman et al., 2011).

In the LMS, the <u>multi-model mean ΔSWV_{slow}/ΔSWV</u> ratio is close to 100% for many perturbations (Figs. 1e-f). The latitude band (50°-90°) we choose is somewhat arbitrary, so in the supplement (Fig. S1), we also show ΔSWV_{slow}/ERF and ΔSWV_{slow}/ΔSWV ratio for water vapor averaged at 200 hPa between 30 and 50 degree latitudes in the NH and SH, respectively, which also show that the ΔSWV_{slow} plays a dominant role and contributes to close to 100% of the total ΔSWV for most perturbations. In the TLS, the <u>multi-model mean ΔSWV_{slow}/ΔSWV</u> ratio is generally above 50%, with a few exceptions. We will discuss this in detail in Section 3.3.

We note that inter-model variability in ΔSWV_{slow}/ERF and ΔSWV_{slow} is generally consistent for different perturbations. For example, HadGEM3 produces larger responses than the rest of the <u>models</u> for most perturbations (Figs. 1a-c, Table S1). GISS-E2-R and MIROC-SPRINTARS have ΔSWV_{slow}/ERF and ΔSWV_{slow} values generally below the rest of the <u>models</u> (Figs. 1a-c, Table S1). We have not further investigated the causes of these differences among models; this clearly warrants further investigation.

We also note that CAM5, CanESM2, and MIROC-SPRINTARS produce negative TLS ΔSWV_{slow}/ERF for 10xBC. These negative values are partly contributed by artifacts of the method we use to estimate equilibrium ΔSWV_{slow}, which is the residual of the total equilibrium ΔSWV minus ΔSWV_{fast}. When differencing two numbers with similar magnitudes, the residual may be quite uncertain. However, the negative values here do not necessarily mean that a BC-induced surface warming results in negative SWV slow response. The direct regression between ΔSWV_{slow} and surface temperature change described in the next section more accurately describe the relationship for these cases.

$3.2\ \mathrm{The\ slow\ stratospheric\ water\ vapor\ response}$ and the surface temperature change

Our results show that, in most climate perturbations analyzed in this study, the equilibrium response of water vapor in both the TLS and the LMS is dominated by ΔSWV_{slow} , which is the component mediated by <u>sea</u> surface temperature change. To directly quantify how SWV responds to surface temperature across a range of different climate change mechanisms, we linearly regress the time series of annual mean ΔSWV_{slow} over the entire period of the coupled simulations (at least 100 years) against the time series of annual mean global averaged surface temperature change (ΔTs). We do this regression for each model and perturbation separately. This is similar to the analysis of Banerjee et al. (2019), who did this for quadrupled CO₂ perturbation, but we do this for multiple perturbations.

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The scatter plot for each perturbation and model is shown in supplement (Figures $\underline{\$3-5}$). For most perturbations and models, the ΔSWV_{slow} time series in both the TLS and the LMS is positively correlated with the ΔTs time series, supporting the hypothesis that the surface temperature change contributes to the long-term trend in SWV for most cases.

Figure 3 shows the slopes of the regression for all perturbations and models. The corresponding slope values are listed in Table

84. We also list slopes in the unit of %/K in Table S5. The uncertainty of the slopes is obtained from Monte Carlo samples:

For each model and perturbation, we first randomly sample the slope 100,000 times, assuming a Gaussian distribution. Then, for each perturbation, we sample from the slope distributions with replacement 100,000 times for each model that performed that perturbation and from these samples compute the multi-model mean and 2.5%-97.5% percentiles.

In both the TLS and LMS, the slopes from different perturbations show general agreement (Fig. 3); this is also true for water vapor averaged at 200 hPa between 30 and 50 degree latitudes in the NH and SH (Fig. S2). In the TLS, the multi-model and multi-perturbation average slope is 0.35 ppmv K⁻¹ with a 95% confidence interval of 0.28-0.44 ppmv K⁻¹ (Fig. 3a). The LMS ΔSWV_{slow} time series has stronger correlations with the ΔTs time series (Figures 33-5) and produces larger sensitivities (Figs. 3b-c). Specifically, the multi-model and multi-perturbation mean slope is 2.1 ppmv K⁻¹ in the NH, and is 0.97 ppmv K⁻¹ in the SH, with 95% confidence intervals of 1.82-2.39 ppmv K⁻¹ and 0.79-1.15 ppmv K⁻¹, respectively. Our results are similar to those of Dessler et al. (2013) and Smalley et al. (2017) despite the fact that they used 500-hPa temperature as their regressor.

We show that the relation between ΔSWV_{slow} and ΔTs time series can be extended to the entire stratosphere (Figs. 4a). We regridded the zonal mean ΔSWV_{slow} from all models and perturbations onto the same pressure-latitude grid (10 hPa above 100 hPa and 50 hPa below 100 hPa, 4 degrees latitude) and regress the ΔSWV_{slow} time series at each grid point against global average ΔTs time series. The multi-model and multi-perturbation average slope of the linear fit at each grid point is shown in Fig. 4a (Figures for each individual perturbation are shown in Fig. 56). Since the vertical gradient of water vapor is large, we plot the percentage change of mixing ratio per degree K relative to the baseline. Lapse rate tropopause, the lowest level where the lapse rate decreases to 2 K km⁻¹, also plotted, is obtained using the atmospheric temperatures from the baseline coupled run and multi-model mean.

We clearly see the larger sensitivity of ΔSWV_{slow} to ΔTs in the LMS than in the overworld. In the LMS, the slope has a hemispheric asymmetry, with larger values in the NH. This is consistent with previous studies, which showed that isentropic transport brings more tropospheric water vapor to the NH than the SH (Pan et al., 1997, 2000; Dethof et al., 1999, 2000; Ploeger et al., 2013). In addition, convective moistening may be more important to the NH due to more land in the NH and, consequently, more convection (Dessler and Sherwood, 2004; Smith et al., 2017; Ueyama et al., 2018; Wang et al., 2019). We also see large responses in the tropical upper troposphere, which is the main part of the tropospheric water vapor feedback.

The sensitivity declines as one ascends through the TTL_{τ} Once above the TTL, the sensitivity in the overworld is relatively uniform with altitude.

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3.3 The fast stratospheric water vapor response

Figure 1 also shows the ΔSWV_{fast} normalized by the ERF (Figs. 1g-i) as well as its contribution to total equilibrium ΔSWV (Figs. 1j-l). As discussed previously, ΔSWV_{fast} is the <u>rapid adjustment</u> in SWV_w before the <u>sea</u> surface <u>temperatures respond</u>. For most perturbations, especially in the LMS, ΔSWV_{fast}/ERF is smaller than ΔSWV_{slow}/ERF, with a magnitude of a few tenths of a ppmv·(Wm⁻²)⁻¹.

For 2xCO₂, the near-zero TLS ΔSWVf_{ast}/ERF is the result of cancellation between cooling by a strengthening Brewer-Dobson circulation and increased local radiative heating (Lin et al., 2017). Some other GHG forcing agents, however, produce larger TLS ΔSWV_{fast}/ERF and contributions in the TLS. The multi-model mean ΔSWV_{fast} from 10xCFC-12 and 10xCFC-11 contribute about half of the total ΔSWV, respectively (Fig. 1j). This is a consequence of halocarbons producing more TTL warming per Wm⁻² by efficiently absorbing upwelling longwave radiation from the troposphere in the atmospheric window (Forster et al., 1997; Jain et al., 2000; Forster and Joshi, 2005). Fig. 5 shows the fast temperature response per unit ERF due to different perturbations and it shows heating in the TTL for both 10xCFC-12 and 10xCFC-11.

The 3xCH₄ also includes some models that produce large TLS ΔSWV_{fast}/ERF magnitudes. This is likely due to TTL heating (Fig. 5) by CH₄ shortwave absorption, which is explicitly treated in some models, including CAM5, CanESM2, MPI-ESM, and MIROC-SPRINTARS (Smith et al., 2018). These models are also the ones that produce the largest TLS ΔSWV_{fast} contributions (Figs. 1g and 1j).

Increases of tropospheric O₃ (in the 5xO₃ experiment) reduce the upwelling longwave radiation, which cools the stratosphere (Ramaswamy and Bowen, 1994; Berntsen et al., 1997; Forster et al., 1997). The longwave radiation absorbed heats the TTL region (Fig. 5), resulting in larger TLS ΔSWV_{fast}/ERF magnitude than ΔSWV_{slow}/ERF and larger contributions to total equilibrium ΔSWV (77%) (Figs. 1g and 1j). There is also heating in the LMS, resulting in larger LMS ΔSWV_{fast}/ERF magnitude than ΔSWV_{slow}/ERF (Figs. 1h-j). We note that our conclusion on 5xO₃ is based on only one model, MIROC-SPRINTARS.

ΔSWV_{fast} from 10xBC dominates total equilibrium ΔSWV in the TLS, with multi-model mean contribution of 84%. The magnitude of the multi-model mean ΔSWV_{fast}/ERF from 10xBC is also larger than any other perturbations in each region. This occurs because the 10xBC strongly absorbs shortwave radiation, causing large heating of the tropopause region in both the tropics and extra-tropics. Figure 5 shows the 10xBC gives by far the most warming per unit ERF, which is consistent with the vertical profile of fast temperature response shown in Stjern et al. (2017).

The $10xBC \Delta SWV_{fast}/ERF$ in the NH and SH LMS contributes to about 50% of the total equilibrium ΔSWV , with smaller magnitudes in the SH (Figs. 1h-i and 1k-l). This is because the total amount of black carbon is smaller in the SH (Myhre et al.,

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2017), since black carbon is a combustion product and is predominantly emitted over the NH continents (Ramanathan and Carmichael, 2008). The 10xBCSLT ΔSWV_{fast} also contributes about 50% of the total 10xBCSLT ΔSWV . The 10xBCSLT does not produce as strong a $\Delta SWV_{fast}/ERF$ as 10xBC, since the reduction in BC lifetime leads to less BC in the TTL and therefore less heating per unit ERF.

330 We quantify control of TLS ΔSWV_{fast} by the fast TTL temperature adjustments across a range of different climate perturbations by regressing the TLS ΔSWV_{fast} against the fast response of the cold point temperature (ΔTCP_{fast}). To estimate ΔTCP_{fast} in the models, we first find the minimum temperature in the profile at each grid point in the fixed SST runs (no interpolation is done, we simply find the minimum temperature on the output model levels). These minimum temperatures are then averaged between 30°N – 30°S to yield TCP_{fast} in each run. ΔTCP_{fast} is the difference between TCP_{fast} in the perturbed model run minus that in the baseline runs.

We find that TLS ΔSWV_{fast} is strongly correlated with ΔTCP_{fast} across all perturbations and models (Fig. 6a), with a slope of 0.52 ppmv K⁻¹ and a 95% confidence interval of 0.43 to 0.61 ppmv K⁻¹. Randel and Park (2019) pointed out that the slope from the Clausius-Clapeyron relationship evaluated near the tropical tropopause is close to this value, about 0.5 ppmv K⁻¹. We also tested the relationship between TLS ΔSWV_{slow} and slow response of the cold point temperature (ΔTCP_{slow}) across all perturbations and models, yielding a slope of 0.72 ppmv K⁻¹. However, for the slow response, correlation does not necessarily prove causality, since Dessler et al. (2016) showed that, in two climate models at least, a significant fraction of the long-term trend was due to increases in convective moistening, which bypasses the TTL cool trap. Therefore this relationship for the slow response could arise from either TCP control or a process that correlates with it, such as deep convective injection of ice, or some combination.

We also separately plot the slopes between ΔSWV_{fast} and ΔTCP_{fast} for each perturbation (Figs. 6d-f). For the perturbations that

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climate mechanisms and across multiple models.

have more than five participating models, including 2xCO₂, 3xCH₄, 2%Solar, 10xBC, 5xSO₄, and 10xCFC-12, we calculate the linear regression between ΔSWV_{fast} and ΔTCP_{fast} from the models and show the slopes and 95% confidence intervals. For the perturbations that have fewer participating models, including 10xCFC11, 3xN₂O, 5xO₃, and 10xBCSLT, we plot the ratio ΔSWV_{fast}/ΔTCP_{fast} and show only the <u>multi-model mean</u>. The slopes produced by different perturbations show general agreement (Fig. 6d). The larger uncertainty in the slopes produced by 2%Solar and 10xCFC-12 occurs because both the ΔTCP_{fast} and ΔSWV_{fast} produced by different models are similar and therefore the slope of the linear regression is uncertain. Overall, we find that the fast response of TTL temperature is a good predictor for the TLS ΔSWV_{fast} across a range of different

For the LMS ΔSWV_{fast}, the ΔTCP_{fast} does not show a control as strong as that in the TLS (Figs. 6b-c) due to the fact that TTL stemperatures are only one factor that influences the LMS. In addition, the regression between ΔSWV_{fast} and ΔTCP_{fast} across all perturbations at each grid point in the pressure-latitude domain shows that the slope (% K-1) follows the transport pattern

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of the BDC (Fig. 4b). The slope is large in the tropical overworld stratosphere and become weaker as one moves poleward and downward in the extra-tropics below 150 hPa. The value is lower in the LMS, again consistent with the fact that water vapor in the LMS is controlled by several processes, not just TTL cold-point temperature. Clearly, more work on this is warranted.

4. Historical changes in SWV

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Given the importance of SWV change, we now ask whether our results can help us understand historical variations in TLS ΔSWV over 1980-2010 (Figure 7). To do this, we estimate historical values of ΔSWV_{slow} and ΔSWV_{fast} based on the PDRMIP results, historical surface temperature change, and historical radiative forcing. For the slow component (blue in Fig. 7a), we multiply 0.35 ppmv K⁻¹, the multi-model multi-perturbation mean sensitivity of the PDRMIP TLS ΔSWV_{slow} to ΔTs, by the historical surface temperature change over 1980-2010. For the fast component (orange in Fig. 7a), we multiply the multi-model mean PDRMIP TLS ΔSWV_{fast}/ERF value for each perturbation by the corresponding historical radiative forcing and then sum it up. We also show the fast component of the historical ΔSWV contributed by each historical forcing agent in Fig. 7b. This is similar to the analysis done by Hodnebrog et al. (2019) in their Figure 6, where they used this method to estimate the historical water vapor lifetime change based on the PDRMIP results.

The historical surface temperature change and radiative forcing data used in this analysis are listed in Table 2. The historical radiative forcing we use here is defined as the change in net downward radiative flux at the tropopause, after adjustments in the stratospheric temperatures, while the surface and troposphere are held unperturbed (Myhre et al. 2013). This is different from the ERF we use in the PDRMIP calculations, which introduced uncertainties in the fast component of the historical ASWV we estimate based on PDRMIP.

Figure 7a shows our estimate that climate change over 1980-2010 has increased TLS SWV by 0.51±0.16 ppmv (Fig. 7a). 36% is due to the slow component, although this is probably an overestimate because our sensitivity value estimated using the PDRMIP results are for long-term. We find the rest of the ASWV, 64%, is due to the fast component, mainly from black carbon. We have also calculated the SWV sensitivity and SWV fast response over 35°N-45°N between 100-80 hPa to recompute the historical 1980-2010 ASWV using the same method, which is 0.65±0.20 ppmv. This value shows reasonable agreement with the SWV increase measured by Hurst et al. (2011) of 0.71±0.26 ppmv over Boulder between 16-18 km over 1980-2010.

Dessler et al. (2014) and Hegglin et al. (2014) argue that there is not a detectible trend over this period. Such a conclusion is not inconsistent with ours because any actual trend estimate has to contend with short-term interannual variability (i.e., like that from the QBO and Brewer-Dobson Circulation variability), which can mask a small trend. Our estimate of the trend is based on sensitivity estimated from 100 year-run and therefore short-term interannual variability has a small impact. Given continuous reliable long-term SWV observation record in the future, one will be able to better test the model-predicted values.

For the fast component of the estimated historical ASWV, radiative forcing by BC plays the dominant role (Fig. 7b).

390 Uncertainties exist in the historical BC radiative forcing we use in this analysis, which is shown in the IPCC AR5 (Myhre et al. 2013). In addition, Allen et al. (2019) pointed out that the radiative effect by BC in the PDRMIP is different from that shown in models using observationally constrained aerosol forcing, which may overestimate the heating in the UTLS region. However, Allen et al. (2019) also noted that uncertainties exist in their observationally constrained aerosol forcing. The uncertainties in the impact of BC forcing on SWV clearly merit more analysis in the future.

395 <u>5. Conclusions</u>

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It is of great interest for the climate community to understand how SWV changes when the climate changes since SWV plays an important role in the Earth's radiative budget and stratospheric ozone chemistry (Solomon et al., 1986, 2010; Dvortsov and Solomon, 2001; Forster and Shine, 2002). In this study, we investigate the response of stratospheric water vapor (SWV) to a range of different climate forcing mechanisms using a multi-model and multiple forcing agent framework. We use output from nine CMIP5 models participating the PDRMIP. Each model performs a baseline and up to 10 climate perturbation experiments, including 2xCO₂, 3xCH₄, 2%Solar, 10xBC, 5xSO₄, 10xCFC-11, 10xCFC-12, 3xN₂O, 5xO₃, and 10xBCSLT (Table 1). Each perturbation is performed in two configurations, including fixed SST simulations (at least 15 years) and fully coupled simulations (at least 100 years).

To better understand the SWV response (Δ SWV), we partition it into two parts: the slow response (Δ SWV_{slow}) and the fast response (Δ SWV_{fast}). The Δ SWV_{fast} is the change in response to a perturbation on short time scales, before the surface temperature has responded. Δ SWV_{slow} occurs on longer time scales and is coupled to the surface temperature change. Our results show that, for most perturbations, Δ SWV in the tropical lower stratosphere (TLS) and in the lowermost stratosphere (LMS) (200 hPa, 50°N-90°N and 50°S-90°S) is dominated by Δ SWV_{slow} (Fig. 1).

Analysis of ΔSWV_{slow} shows that a warming surface increases SWV (Figures §3.5). Furthermore, the response of SWV to the surface temperature change has a similar sensitivity across different climate perturbations in both the overworld stratosphere and the lowermost stratosphere (Figs. 3 and 4a). Specifically, the multi-model and multi-perturbation mean slope is 0.35 ppmv K⁻¹ in the TLS, 2.1 ppmv K⁻¹ in the northern hemispheric (NH) LMS, and 0.97 ppmv K⁻¹ in the southern hemispheric (SH) LMS (Fig. 3).

ΔSWV_{slow} in the LMS is more sensitive to ΔTs than the tropical overworld, reflecting different transport pathways into the LMS compared to the overworld (Dessler et al., 1995; Holton et al., 1995; Plumb, 2002; Gettelman et al., 2011). The ΔSWV_{slow} in the NH LMS is more sensitive than the SH LMS, consistent with hemispheric asymmetries in the isentropic transport and convective moistening reported by previous studies (Pan et al., 1997, 2000; Dethof et al., 1999, 2000; Dessler and Sherwood, 2004; Ploeger et al., 2013; Smith et al., 2017; Ueyama et al., 2018; Wang et al., 2019).

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The fast response of SWV from most perturbations are weak compared to the slow response and therefore plays a smaller role in Δ SWV (Fig. 1). In the TLS, for forcing agents that directly heat tropopause levels (Fig. 5), Δ SWV_{fast} makes a larger Deleted: However contribution to ΔSWV . In particular, when climate is perturbed by 10xBC, the ΔSWV_{fast} dominates the ΔSWV_{slow} and has larger magnitude than any other perturbed simulations. This occurs because black carbon absorbs shortwave radiation in the Deleted: in both the TLS and LMS atmosphere and directly heats the temperatures at tropopause levels. Other forcing agents also heat the tropopause levels and increase ΔSWV_{fast} through absorption of shortwave radiation or longwave radiation at the atmospheric window range (3xCH₄, 5xO₃, 10xBCSLT, 10xCFC-12, 10xCFC-11), but are not as strong as 10xBC. 430 The TLS \(\Delta SWV \) fast is controlled by the fast response of the cold point temperature across different climate change mechanisms (Fig. 6), with a slope of 0.52 ppmv K⁻¹, which is consistent with the Clausius-Clapeyron relationship evaluated near the tropical tropopause (Randel and Park, 2019). The control of cold point temperature fast response over ΔSWV_{fast} is stronger in the tropical overworld and becomes weaker at higher latitudes and altitudes below 150 hPa in the LMS (Fig. 4b). Deleted: in the LMS 435 Data availability: The PDRMIP data can be downloaded from this website: https://cicero.oslo.no/en/PDRMIP. Competing interests. The authors declare that they have no conflict of interest. Author contribution: Xun Wang performed analyses and wrote the paper. Andrew E. Dessler provided the conceptualization, Deleted: original draft. guidance, and editing. Acknowledgments: This work was supported by NASA grants 80NSSC18K0134 and 80NSSC19K0757. This work was also supported by the National Center for Atmospheric Research, which is a major facility sponsored by the National Science Foundation under Cooperative Agreement No. 1852977. Any opinions, findings and conclusions or recommendations expressed in this material do not necessarily reflect the views of the National Science Foundation. We would like to acknowledge the PDRMIP modelling groups and helpful discussions with Andrew Gettelman and William Randel. Deleted: . We acknowledge References. 445 Adams, B. K. and Dessler, A. E.: Estimating Transient Climate Response in a Large-Ensemble Global Climate Model Simulation, Geophys. Res. Lett., 46(1), 311–317, doi:10.1029/2018GL080714, 2019. Allen, R. J., Amiri-Farahani, A., Lamarque, J.-F., Smith, C., Shindell, D., Hassan, T. and Chung, C. E.: Observationally constrained aerosol-cloud semi-direct effects, npj Clim. Atmos. Sci., 2(1), 16, doi:10.1038/s41612-019-0073-9, 2019. Arora, V. K., Scinocca, J. F., Boer, G. J., Christian, J. R., Denman, K. L., Flato, G. M., Kharin, V. V., Lee, W. G. and

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Table 1: Columns 1-6: Description of PDRMIP models (Myhre et al., 2017). © American Meteorological Society. Used with permission. Column 7: List of perturbation experiments used in this study.

Model	Version	Resolution	Ocean setup	Aerosol setup	Key references	Perturbation
						experiments
Second Generation Canadian	2010	2.8°×2.8°,	Coupled	Emissions	(Arora et al., 2011)	2xCO ₂ ,
Earth System Model		35 levels	ocean			3xCH ₄ ,
(CanESM2)						2%Solar,
						10xBC,
						5xSO ₄
Community Earth System	1.0.3	2.5°×1.9°,	Slab	Fixed	(Neale et al., 2010;	2xCO ₂ ,
Model, version 1		26 levels	ocean	concentrations	Gent et al., 2011)	3xCH ₄ ,
(Community Atmosphere						2%Solar,
Model, version 4)						10xBC,
[CESM1(CAM4)]						5xSO ₄ ,
						10xCFC-12,
						3xN ₂ O,
						10xBCSLT
CESM1 CAM5	1.1.2	2.5°×1.9°,	Coupled	Emissions	(Hurrell et al., 2013;	2xCO ₂ ,
		30 levels	ocean		Kay et al., 2015; Otto-	3xCH ₄ ,
					Bliesner et al., 2016)	2%Solar,
						10xBC,
						5xSO ₄ ,
						10xCFC-12
Goddard Institute for Space	E2-R	2°×2.5°,	Coupled	Fixed	(Schmidt et al., 2014)	2xCO ₂ ,
Studies Model E2, coupled		40 levels	ocean	concentrations		3xCH ₄ ,
with the Russell ocean model						2%Solar,
(GISS-E2-R)						10xBC,
						5xSO ₄ ,
						10xCFC-12,
						10xBCSLT
Hadley Centre Global	6.6.3	1.875°×1.25°,	Coupled	Emissions	(Collins et al., 2011;	2xCO ₂ ,
Environment Model, version		38 levels	ocean		Martin et al., 2011)	3xCH ₄ ,
2-Earth System (includes						2%Solar,
Carbon Cycle configuration						10xBC,
with chemistry) (HadGEM2-						5xSO ₄ ,
ES)						10xCFC-12,

						10xCFC-11,
						$3xN_2O$
HadGEM3	Global	1.875°×1.25°,	Coupled	Fixed	(Bellouin et al., 2011;	2xCO ₂ ,
	Atmosp	85 levels	ocean	concentrations	Walters et al., 2014)	3xCH ₄ ,
	here 4.0					2%Solar,
						10xBC,
						5xSO ₄ ,
						10xCFC-12
L'Institut Pierre-Simon	CMIP5	3.75°	Coupled	Fixed	(Dufresne et al., 2013)	2xCO ₂ ,
Laplace Coupled Model,		×1.875°, 39	ocean	concentrations		3xCH ₄ ,
version 5A (IPSL-CM5A)		levels				2%Solar,
						10xBC,
						5xSO ₄
Max Planck Institute Earth	1.1.00p	T63, 47 levels	Coupled	Climatology,	(Giorgetta et al., 2013)	2xCO ₂ ,
System Model (MPI-ESM)	2		ocean	year 2000		3xCH ₄ ,
						2%Solar
Model for Interdisciplinary	5.9.0	T85 (approx.	Coupled	Hemispheric	(Takemura, 2005;	2xCO ₂ ,
Research on Climate-Spectral		1.4°×1.4°), 40	ocean	Transport Air	Takemura et al., 2009;	3xCH ₄ ,
Radiation-Transport Model		levels		Pollution, phase	Watanabe et al., 2010)	2%Solar,
for Aerosol Species (MIROC-				2 Emissions		10xBC,
SPRINTARS)						5xSO ₄ ,
						10xCFC-12,
						10xCFC-11,
			1	I .	l .	

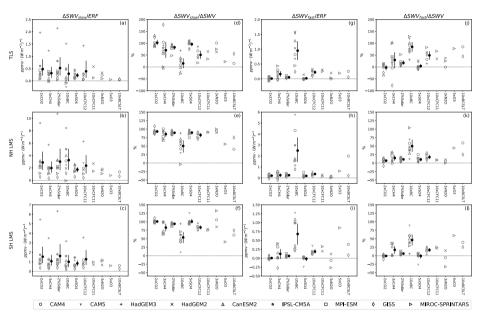


Figure 1. Panels (a)-(c): equilibrium ΔSWV_{slow} normalized by ERF (ppmv·(Wm²)-¹) in TLS (70 hPa, 30°N-30°S), NH LMS (200 hPa, 50°N-90°N), and SH LMS (200 hPa, 50°S-90°S). Panels (d)-(f): Contribution (%) of equilibrium ΔSWV_{slow} to total equilibrium ΔSWV . Panels (g)-(i): ΔSWV_{fast} normalized by ERF (ppmv·(Wm²)-¹). Panels (j)-(l): Contribution (%) of ΔSWV_{fast} to total equilibrium ΔSWV . The marker shapes indicate results from different models, For perturbations that are performed by more than three models, the solid circles and error bars for each perturbation plotted in weighted black are multi-model mean and 2.5%-97.5% percentiles of the model samples. Note that in the second and fourth columns, we took out models with extremely small ΔSWV magnitudes that yield extremely large $\Delta SWV_{slow}/\Delta SWV$ and $\Delta SWV_{fast}/\Delta SWV$ ratios.

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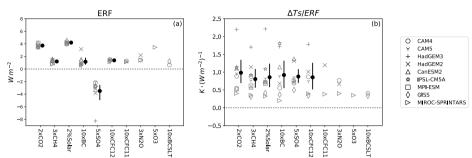


Figure 2. Panel (a): Global average ERF (W m^2) at the top of atmosphere. Panel (b): Global averaged surface temperature change per unit ERF (K-(W m^2)⁻¹). The marker shapes indicate results from different models. For perturbations that are performed by more than three models, the solid circles and error bars for each perturbation plotted in weighted black are nulti-model mean and 2.5%-97.5% percentiles of the model samples.

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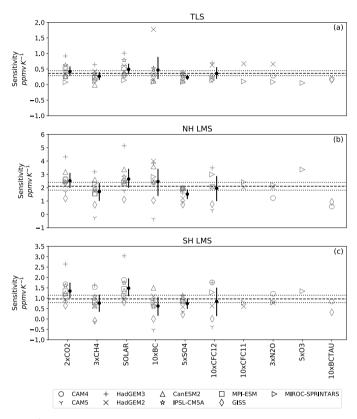


Figure 3. Slopes (ppmv K-1) from the linear regression between annual mean ΔSWV_{slow} time series and annual mean ΔTs time series.

The marker shapes indicate results from different models. For perturbations that are performed by more than three models, the solid circles and error bars for each perturbation plotted in weighted black are <u>multi-model mean</u> and 2.5%-97.5% <u>percentiles of the model samples</u>. The horizontal dashed line is the <u>multi-model mean</u> of all slopes, and the horizontal dotted lines are 2.5%-97.5% <u>percentiles of the model samples</u>.

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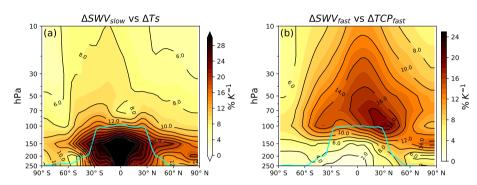
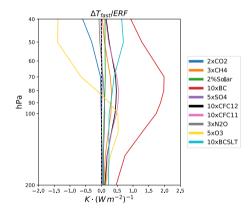


Figure 4. Panel (a): Multi-model and multi-perturbation mean slope (% K-1) from the regression between annual mean time series of ΔSWV_{slow} at each latitude grid point and pressure level and annual mean time series of global average ΔTs. Panel (b): Slope (% K-1) from the regression between ΔSWV_{fast} (ppmv) at each latitude grid point and pressure level and ΔTCP_{fast} (K). The solid cyan line is the multi-model mean lapse rate tropopause derived from the baseline simulations.

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760 Figure 5. Profiles of fast temperature response normalized by ERF (K·(Wm²)-¹) between 200 and 40 hPa, and averaged over 30°N-30°S. The color coding indicates results from different perturbations. Each profile is the <u>multi-model mean</u>.

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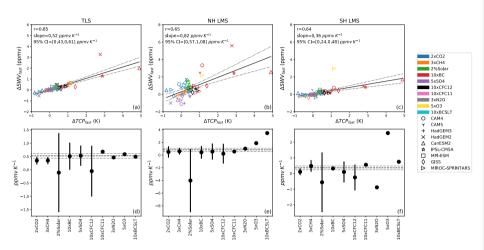


Figure 6. Panels (a)-(c): Linear regression between ΔSWV_{fast} (ppmv) and ΔTCP_{fast} (K) from all models and perturbations. The color coding indicates different perturbations, while the marker shapes indicate results from different models. The black solid line is the linear fit of the regression. The black dotted lines indicate the linear fits within the 95% confidence interval, estimated using a t-test. Panels (d)-(f): Slopes and their 95% confidence intervals (for perturbations that are performed by more than three models) obtained from linear regression between ΔSWV_{fast} (ppmv) and ΔTCP_{fast} (K) for each individual perturbation. The black dashed lines and dotted lines are the slopes and their 95% confidence intervals of regressions in (a)-(c).

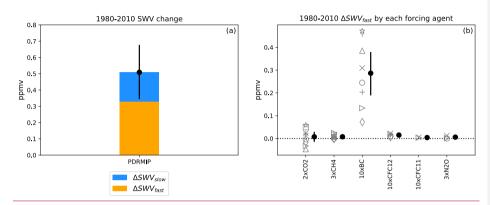


Figure 7. (a) TLS (30°S-30°N, 70 hPa) SWV change over 1980-2010 estimated using PDRMIP results. Blue indicates the component contributed by the slow response, while orange indicates the component contributed by the fast response. (b) The fast component of the PDRMIP-estimated 1980-2010 SWV change contributed by each historical forcing agent. The solid circles are multi-model mean. The error bars are 2.5%-97.5% percentiles of the model samples; in Panel (b) they are shown for perturbations that are performed by more than three models.

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Table 2: Historical global average surface temperature change and radiative forcing (RF) by Greenhouse gases (GHGs) and halocarbons over 1980-2010. The SWV change over 1980-2010 estimated using PDRMIP results is also listed, including the total SWV change, the slow component, and the fast component. For the fast component of SWV change contributed by each forcing agent, multi-model mean results are listed. The uncertainties are 2.5%-97.5% percentiles of the model samples.

GMST ^a (K)	0.506
Total ASWV (ppmv)	0.51±0.16
ΔSWV _{slow} (ppmv)	0.18±0.04
ΔSWV _{fast} (ppmv)	0.32±0.12

Forcing agents	RF (Wm ⁻²)	ΔSWV _{fast} by each forcing agent (ppmv)
CO2 ^b	0.715	0.007±0.022
<u>CH4</u> ^{<u>c</u>}	0.055	0.008±0.005
BC ^d	0.3	0.286±0.095
CFC-12e	0.068	0.015±0.004
CFC-11f	0.015	0.004
N_2O^g	0.042	0.005

a: We used NOAA Merged Land Ocean Global Surface Temperature Analysis V5 (Zhang et al. 2020) to compute the global surface temperature change. We use values averaged over 2005-2015 minus that averaged over 1975-1985.

b, c, e, f, and g: We compute the RFs using the formulae listed in Table 3 of Myhre et al. (1998). These formulae were also used to compute RFs of CO₂, CH₄, and N₂O in IPCC reports (Myhre et al. 2013).

b, c, and g: Concentrations of GHGs were used to compute RFs. CO₂ and CH₂ are samples collected in glass flasks at Cold Bay, Alaska, United States (CBA) from the ERSL GML website (Dlugokencky et al. 2020). N₂O is from the Combined Nitrous Oxide data from the NOAA/ESRL Global Monitoring Division. For CO₂, concentrations averaged over 2005-2015

and averaged over 1978-1985 are used. For CH₄, concentrations averaged over 2005-2015 and averaged over 1983-1985 are used. For N_2O , concentration averaged over 2005-2015 and averaged over 1977-1985 are used. e-f: Concentrations of CFC-12 and CFC-11 were used to compute RFs. We use CFC-12 and CFC-11 data from combined

e-f: Concentrations of CFC-12 and CFC-11 were used to compute RFs. We use CFC-12 and CFC-11 data from combined stations from the NOAA/ESRL Global Monitoring Division. Concentrations averaged over 2005-2015 and averaged over 1977-1985 are used.

d: We use 0.4 Wm², the BC RF between 1750-2011 reported in IPCC AR5, minus 0.1 Wm², the BC RF between 1750-1993 reported in 1995 IPCC report (See Table 8.4 of Myhre et al. 2013).