

Greenhouse gas forcing and climate feedback signatures identified in hyperspectral infrared satellite observations

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Introduction

The supporting text below expands on the radiative transfer simulations and general circulation model simulations conducted for this study. The supporting tables provide quantitative estimates of radiative forcing. The supporting figures provide supplemental information to the main text.

Text S1.

The LBL-ERA5 calculations use a GPU-optimized LBL code called GFDL-GRTCODE.

GRTCODE has been benchmarked as part of the RFMIP-IRF CMIP6 radiation code intercomparison (Pincus et al., 2020). It uses HITRAN 2016 and the MTCKD2.5 water vapor continuum. A gaussian-weighted non-scattering solver based upon Clough et al. (1992) is used and emissivity is set to 1.0. While both assumptions could lead to inaccuracies in absolute fluxes, our expectation is that their influence on the zonal and global-mean trends presented in this paper would be minimal. The code is driven with monthly mean output from ERA5 reanalysis of humidity, O₃, temperature and cloud data at a 90x144 lat-lon resolution and global WMGHG (including CFCs) concentrations from the NOAA GML observational record (<https://gml.noaa.gov/ccgg/trends/>). Cloud fraction, cloud liquid, and cloud ice from ERA5 are used as input to GRTCODE. Cloud optical properties (optical depth, single scatter albedo, and asymmetry factor) at each grid point layer is calculated. To represent sub-monthly variability, 10 stochastically generated columns per grid box are used (Pincus et al., 2006; Tompkins, 2002).

For the "LBL-GCM" calculations, we take annual mean output of WMGHG (excluding CFCs), humidity, O₃, and temperature at 90x144 lat-lon resolution from idealized radiative forcing and global warming GCM runs. These conditions are then used in the RFM (Dudhia, 2017) LBL code coupled with the GRTCODE solver to estimate the spectral features associated with climate change (see Table 1 for details). This code is also benchmarked in Pincus et al., 2020 but with the DISORT solver (GFDL-RFM-DISORT). Here we bring in the GRTCODE solver for computational efficiency, but this has minor impact on results in the absence of scattering. These "LBL-GCM" calculations are just performed for clear-sky conditions. Finally, GRTCODE spectra are sampled and solved at 1cm⁻¹ resolution and GFDL-RFM spectra are sampled and

solved at 0.01 cm^{-1} from $1\text{-}1500 \text{ cm}^{-1}$ and at 0.1 cm^{-1} for higher wavenumber. After experimentation, it was found that 1 cm^{-1} sampling was sufficient resolution to capture the trends observed by AIRS.

Experiment 8. Band-AM4/MERRA-Total: quantify narrowband ΔOLR

Although Experiment 1. provides all-sky ΔOLR_v at high spectral resolution, monthly-mean inputs to the radiative transfer model may cause biases in the simulation, especially with respect to cloud fields. To bridge this gap, we conduct radiative transfer calculations at 3-hourly timescales in GFDL Atmospheric Model 4 (AM4; Zhao et al., 2018) to produce clear-sky and all-sky OLR at 9 bands that follow the Schwarzkopf and Ramaswamy (1999) longwave radiation parameterization. We prescribed observed sea surface temperatures (SST) and sea-ice in GFDL AM4 during 2003-2021 (Atmospheric Modeling Intercomparison Project (AMIP) conditions) and nudged the model toward Modern-era retrospective analysis for research and applications (MERRA; Rienecker et al., 2011) reanalysis temperature and wind fields to produce a simulation that resembles observations better.

Finally, although the spectral signature of the surface temperature response identified in this study will be in the infrared alone, we note that the observed surface temperature response arises from all forcing agents in the infrared and solar spectra.

Name	Wavenumber range (cm ⁻¹)
Rotational band of H ₂ O	160-560
CO ₂	560-630, 630-700, 700-800
Window	800-900, 900-990, 990-1070, 1070-1200
O ₃	990-1070
CH ₄ , N ₂ O	1200-1400
Rotational-vibrational band of H ₂ O	1400-2000

Supplementary Table S1: Wavenumber ranges for gas species based on GFDL AM4 bands. Note that CERES Window is defined as 8-12 μm , i.e., 833-1250 cm^{-1} . To be consistent with AM4 bands, we treat the window as 800-1200 cm^{-1} (Row 3).

Band (cm ⁻¹)/Gas	Trend ($\text{Wm}^{-2}\text{decade}^{-1}$)
1. 560-800/CO ₂ main	-0.15
2. 800-1200/CFCs and CO ₂ weak	-0.02
a) 830-860, 870-930, 1080-1120, 1140-1160/CFCs	+0.01
b) CO ₂ weak (2.-2a.)	-0.03
3. 1200-1400/CH ₄ , N ₂ O	-0.04
Total	-0.21

Supplementary Table S2: LBL-ERA5-IRF breakdown by bands/gases.

Dataset/Study	Trend Estimate ($Wm^{-2}decade^{-1}$)	Model	Includes Land Warming?	SARF or ERF?
LBL-ERA5-IRF + LBL-GCM-Adj. (Strat.)	-0.30 ± 0.02	Line-by-line radiative transfer	No	SARF
IPCC AR6 Forster et al., 2021/Smith et al., 2021, Meinshausen et al., 2020, Etminan et al., 2016	-0.34 ± 0.03	Simplified expressions	No	SARF
GFDL AM4	-0.26 ± 0.03	Parameterized radiative transfer	Yes	ERF

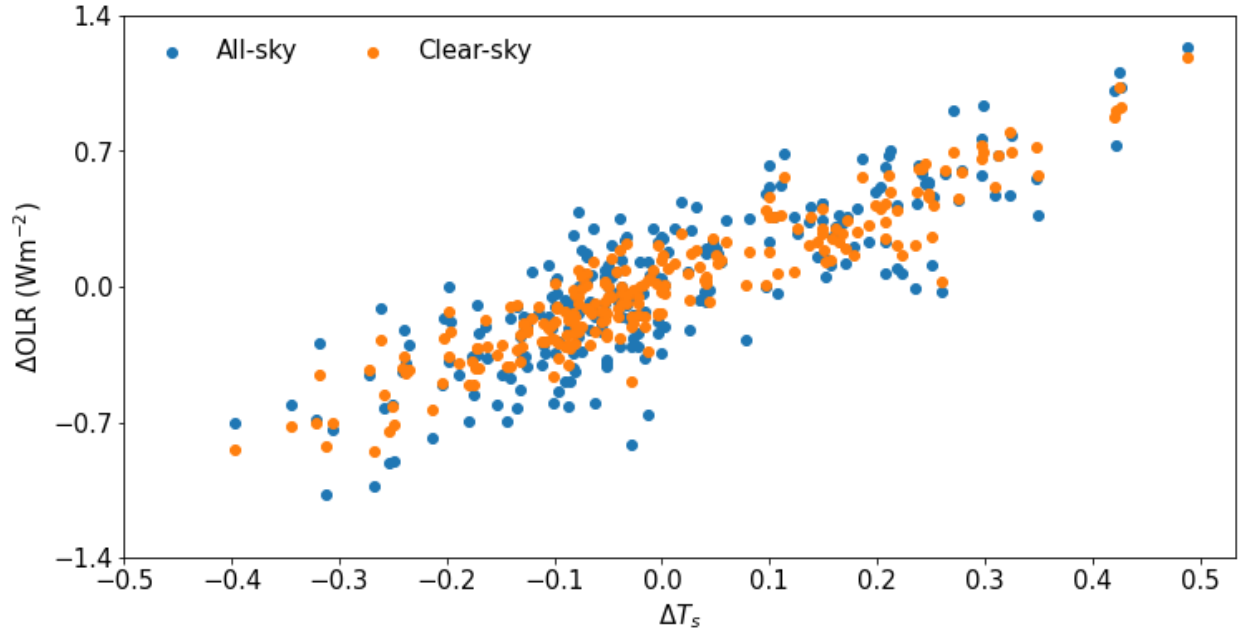
Supplementary Table S3: All-sky, LW, broadband, global-mean, trends in radiative forcing due to WMGHGs (CO₂, CH₄, N₂O, CFC-11, CFC-12, CFC-113, HFC-22) during 2003-2021.

Row 1's estimate comes from: SARF = LBL-ERA5-IRF (all-sky) + LBL-GCM-Adj. (Strat.) (clear-sky). As there are little-to-no clouds in the stratosphere, this equation provides an estimate of all-sky SARF. Note that SARF in Row 1 provides stratospheric adjustments only due to quadrupling CO₂ (but scaled), not due to any of the other WMGHGs. Uncertainties given by 95% CI ($1.96 \times S.E.$, where S.E. is the standard error associated with linear fit of the time series).

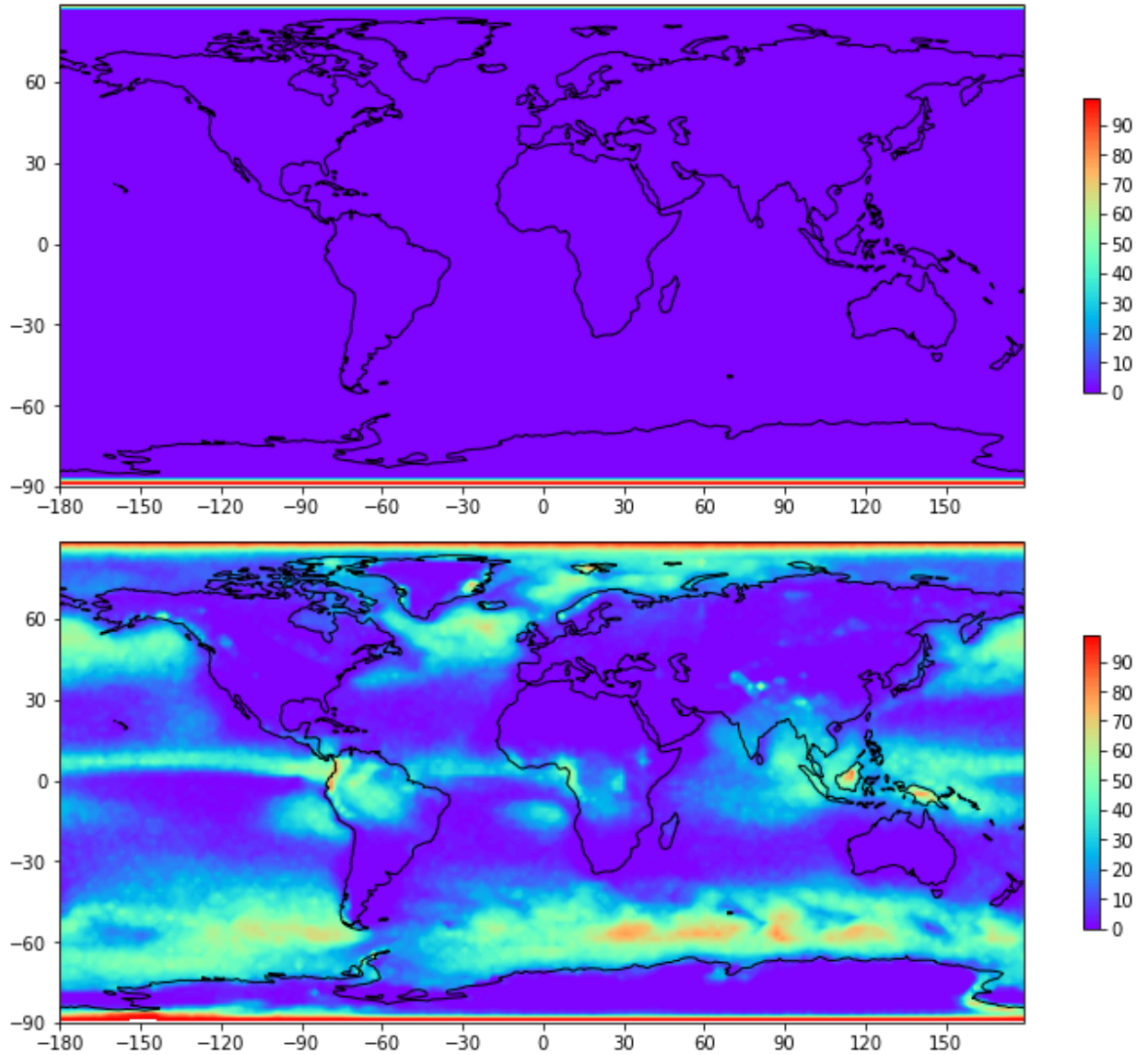
Row 2's estimate using IPCC AR6/Meinshausen et al., 2020 expressions agree with Etminan et al., 2016 expressions to $< 0.003 Wm^{-2}decade^{-1}$ for the sum of CO₂, CH₄, and N₂O forcing. Given this agreement, the estimate listed above is from the IPCC AR6 expressions but corrects for SW contributions using Etminan et al., 2016's SI Table 1: SW CO₂ = -4% of Net (their Row 25), SW CH₄ = +5% of Net (their Row 7), and N₂O = -2% of Net (their Row 3)). Row 2's uncertainty is derived by applying the $\pm 8.3\%$ uncertainty of the ERF best-estimate during 1750-2019 (their Table 7.5 and 7.SM.1.3.2) to the 2003-2021 trend ($\pm 8.3\% \times 0.34 = \pm 0.03$). This provides a 90% CI which we then scale to a 95% CI, which is also a $\pm 0.03 Wm^{-2}decade^{-1}$ uncertainty.

Row 3's estimate comes from the RFMIP piClim-histghg experiment (Pincus et al., 2016) in GFDL AM4.

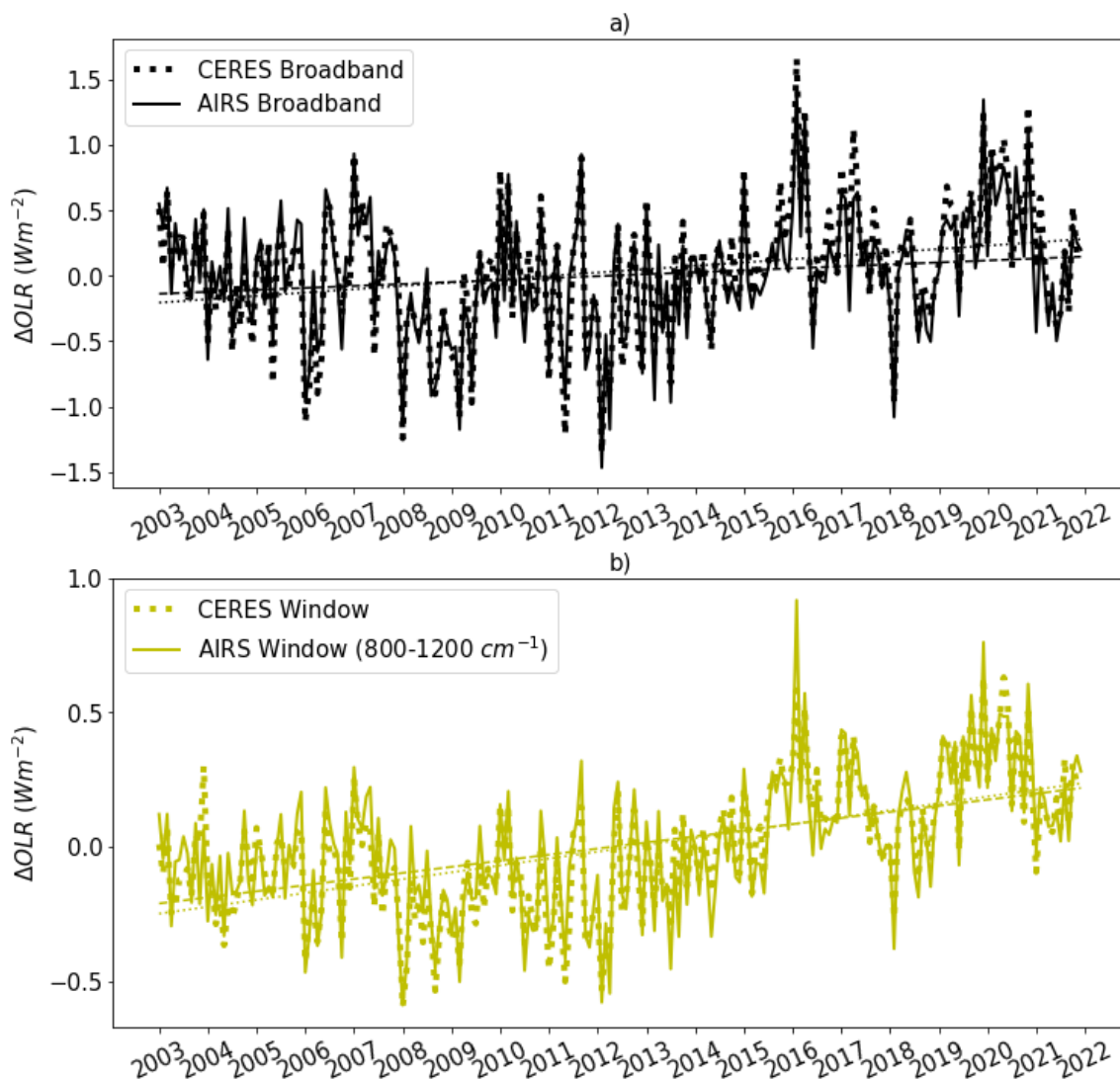
SARF = stratospheric-temperature-adjusted radiative forcing. ERF = Effective radiative forcing. WMGHG = well-mixed greenhouse gases. Negative values imply less outgoing longwave radiation, i.e., heating Earth.



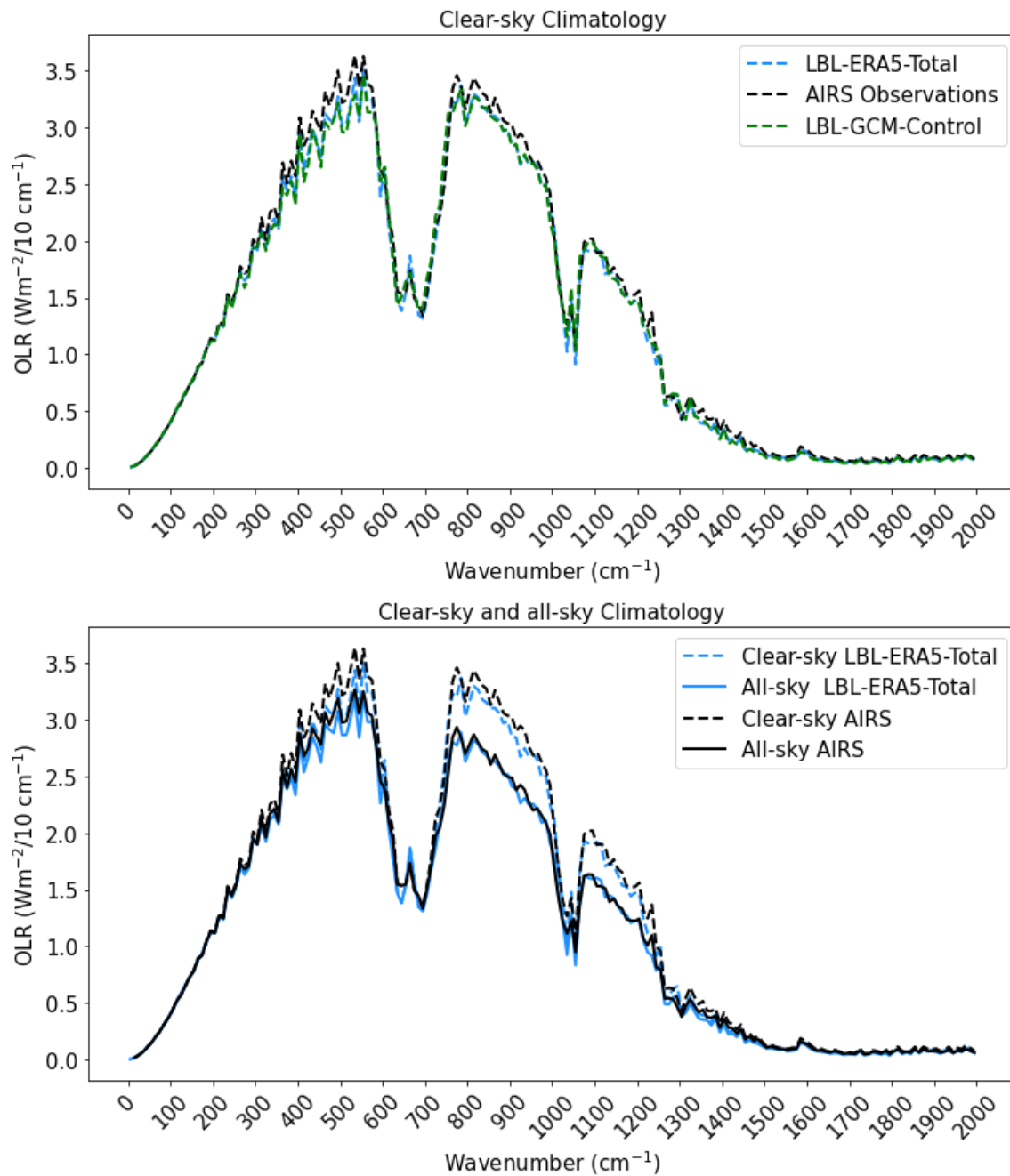
Supplementary Figure S1: Monthly-mean anomalies in OLR vs. surface temperature during 2001-2020. Data taken from GFDL AM4 PSST experiment which isolates the radiative response (Raghuraman et al., 2021). Ensemble-mean of 20 realizations plotted. $r(\Delta T_s, \Delta OLR) = 0.83, 0.93$ for all-sky and clear-sky respectively.



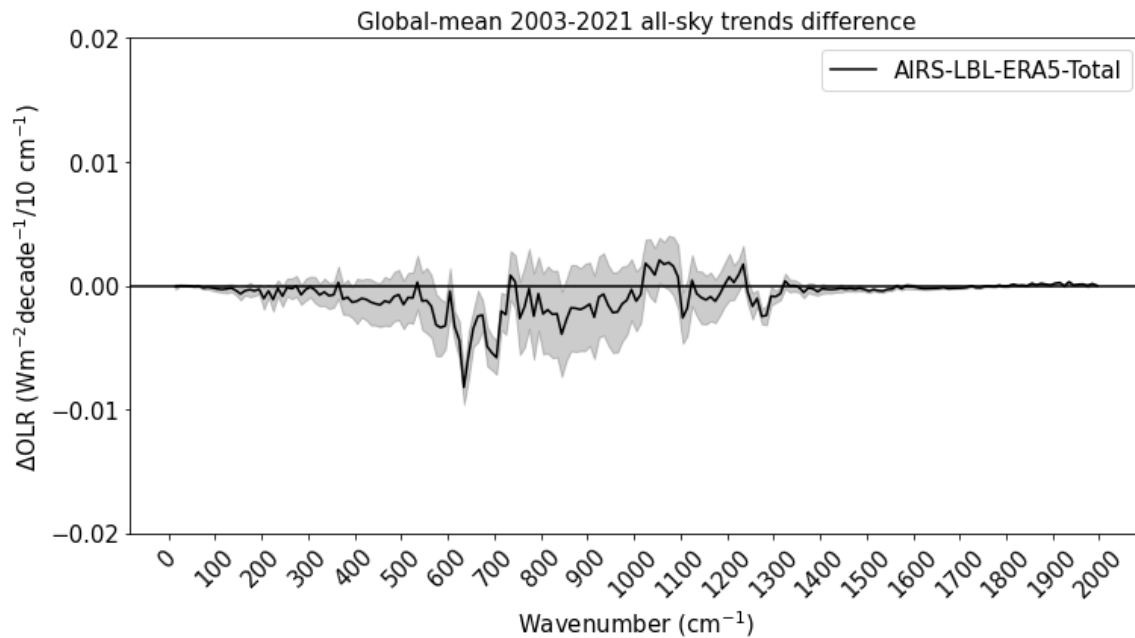
Supplementary Figure S2: a) Percentage of months without a measurement for AIRS all-sky OLR at each grid point during September 2002-December 2021. b) Same as a) but for clear-sky OLR. As a result of missing data at poles we consider global-mean = 85°S-85°N for both all-sky and clear-sky OLR. Clear-sky OLR zonal-mean trends are not considered due to poor spatial coverage.



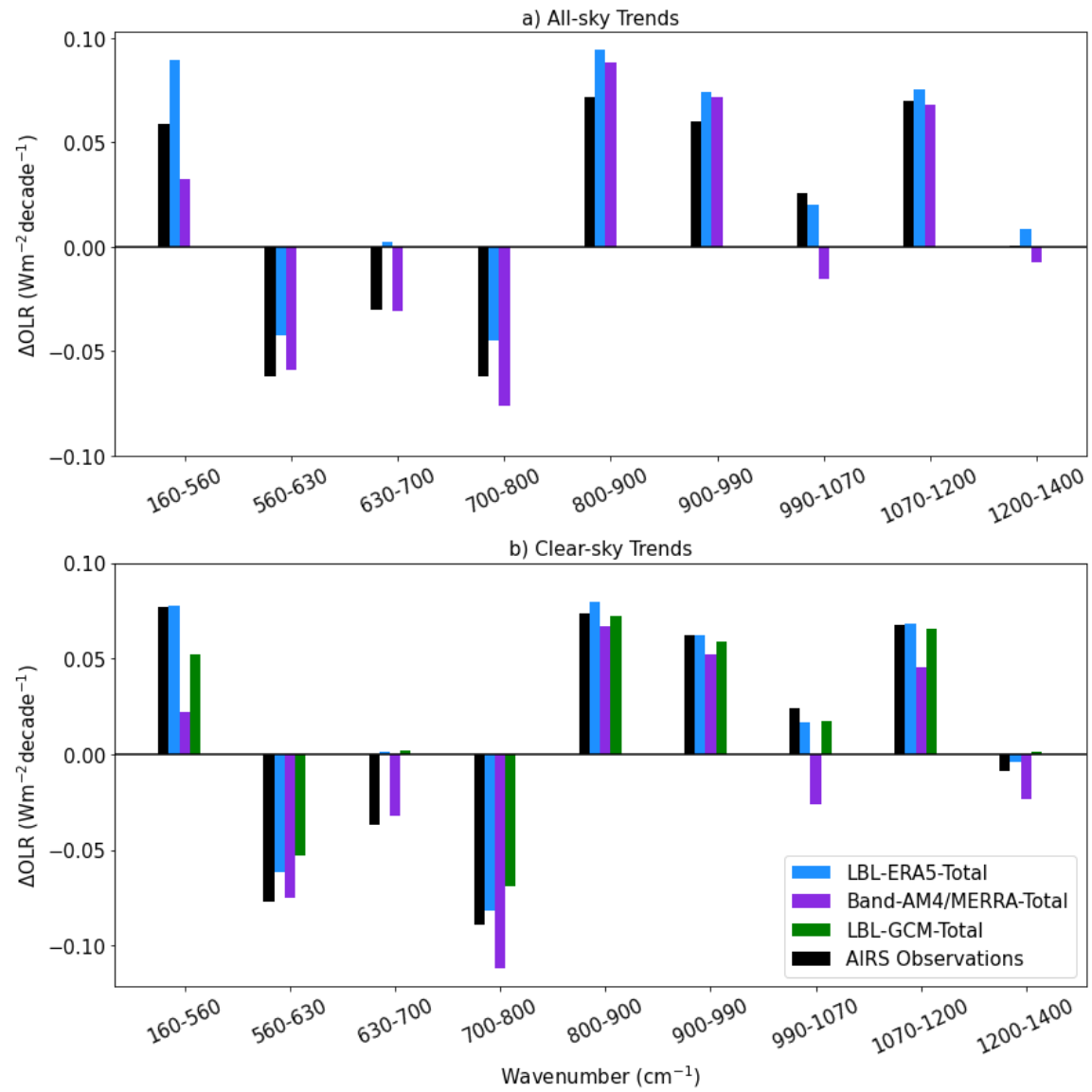
Supplementary Figure S3: Comparison between all-sky CERES and AIRS. a) Broadband fluxes. The timeseries are highly consistent with each other ($r=0.94$). CERES and AIRS trends are $0.26 Wm^{-2}decade^{-1}$ and $0.15 Wm^{-2}decade^{-1}$, respectively. b) Window fluxes. CERES and AIRS trends are $0.26 Wm^{-2}decade^{-1}$ and $0.23 Wm^{-2}decade^{-1}$, respectively.



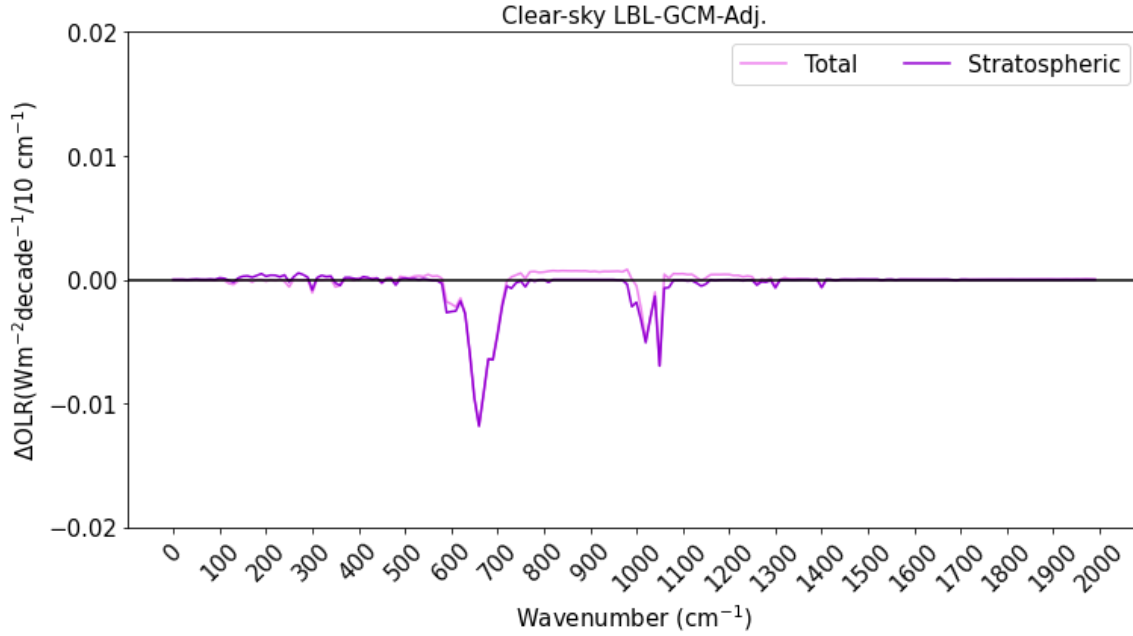
Supplementary Figure S4: Global-mean spectrally-resolved climatologies. a) Clear-sky only. b) clear-sky and all-sky comparison.



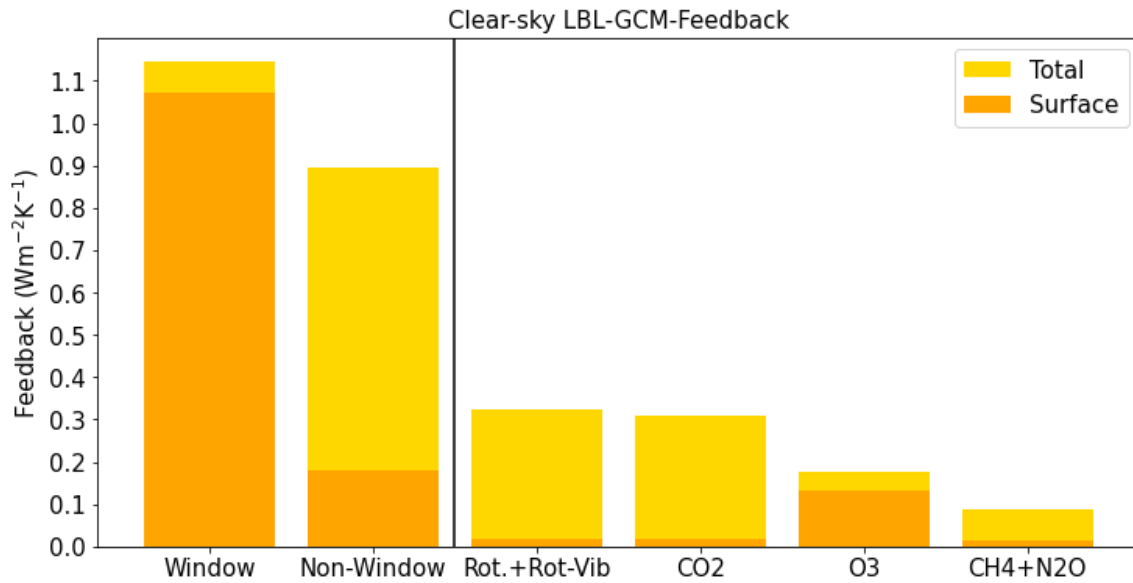
Supplementary Figure S5: Difference between AIRS and LBL-ERA5-Total for global-mean all-sky OLR trends, 2003-2021.



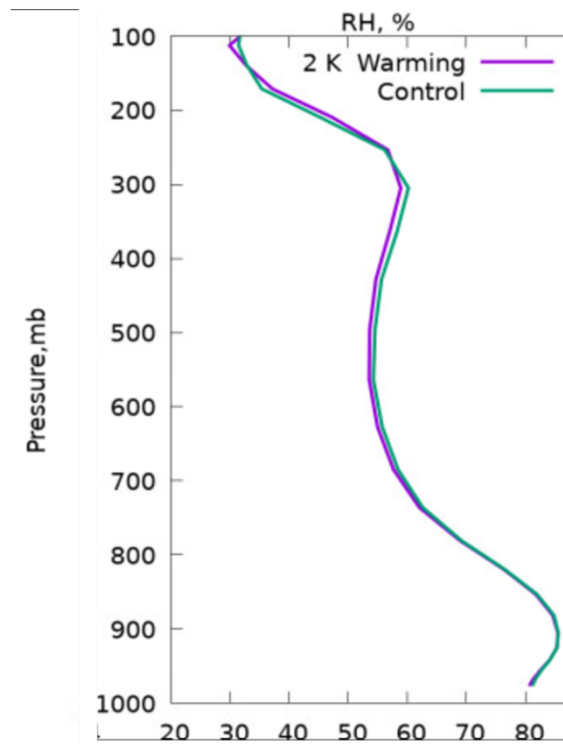
Supplementary Figure S6: Global-mean narrowband trends. a) All-sky. b) Clear-sky.



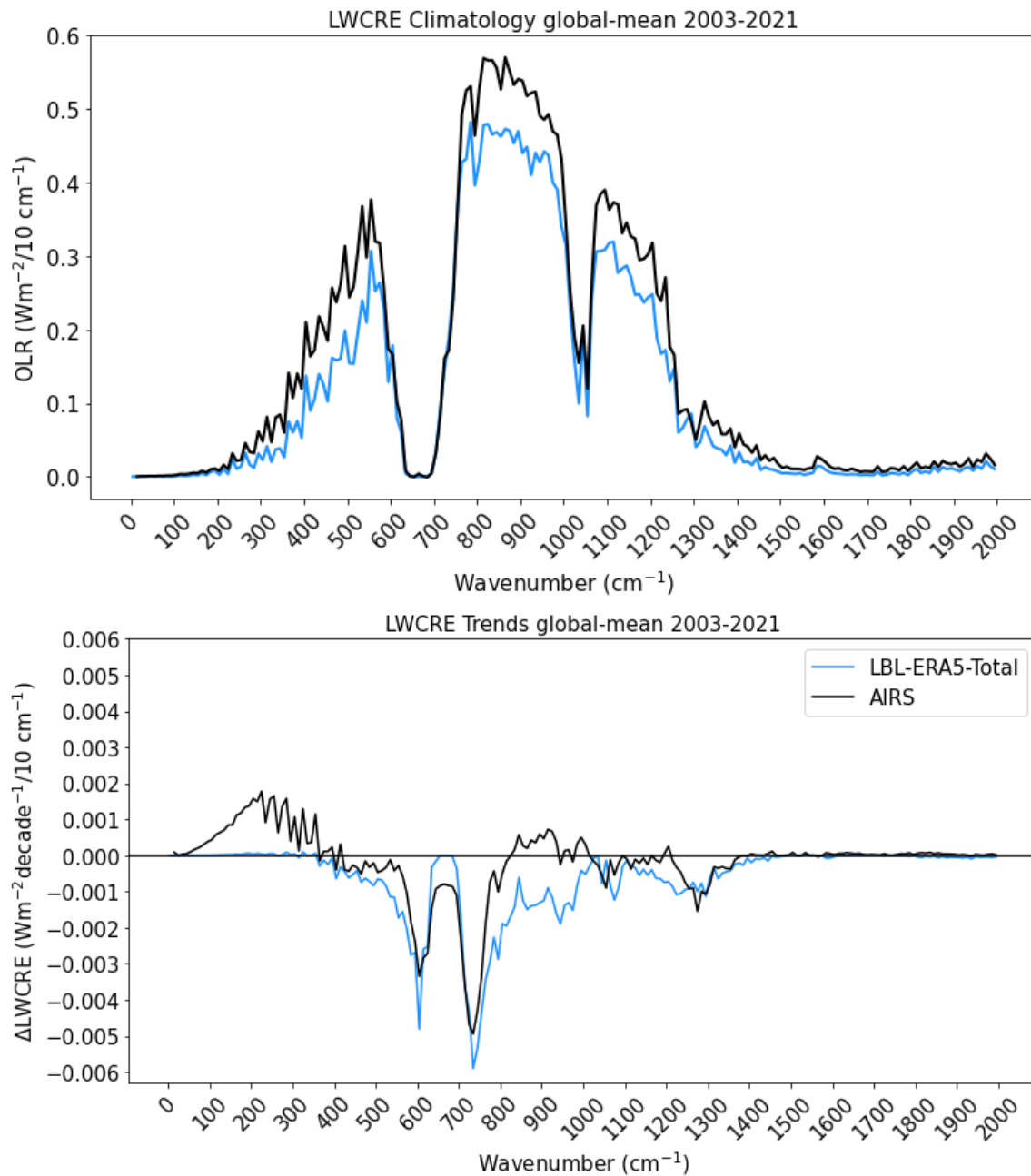
Supplementary Figure S7: Global-mean clear-sky adjustments breakdown. LBL-GCM-Adj. = Stratospheric adjustments + Tropospheric adjustments + Land warming. LBL-GCM-Adj. (Strat.) = Stratospheric adjustments. The difference between these two simulations ($\sim 0.03 \text{ Wm}^{-2} \text{ decade}^{-1}$, summed over all wavenumbers) is thus tropospheric adjustments + land warming. Clear-sky tropospheric adjustments are near-zero because this figure shows a small increase in OLR but that is from land surface warming because it manifests in the window.



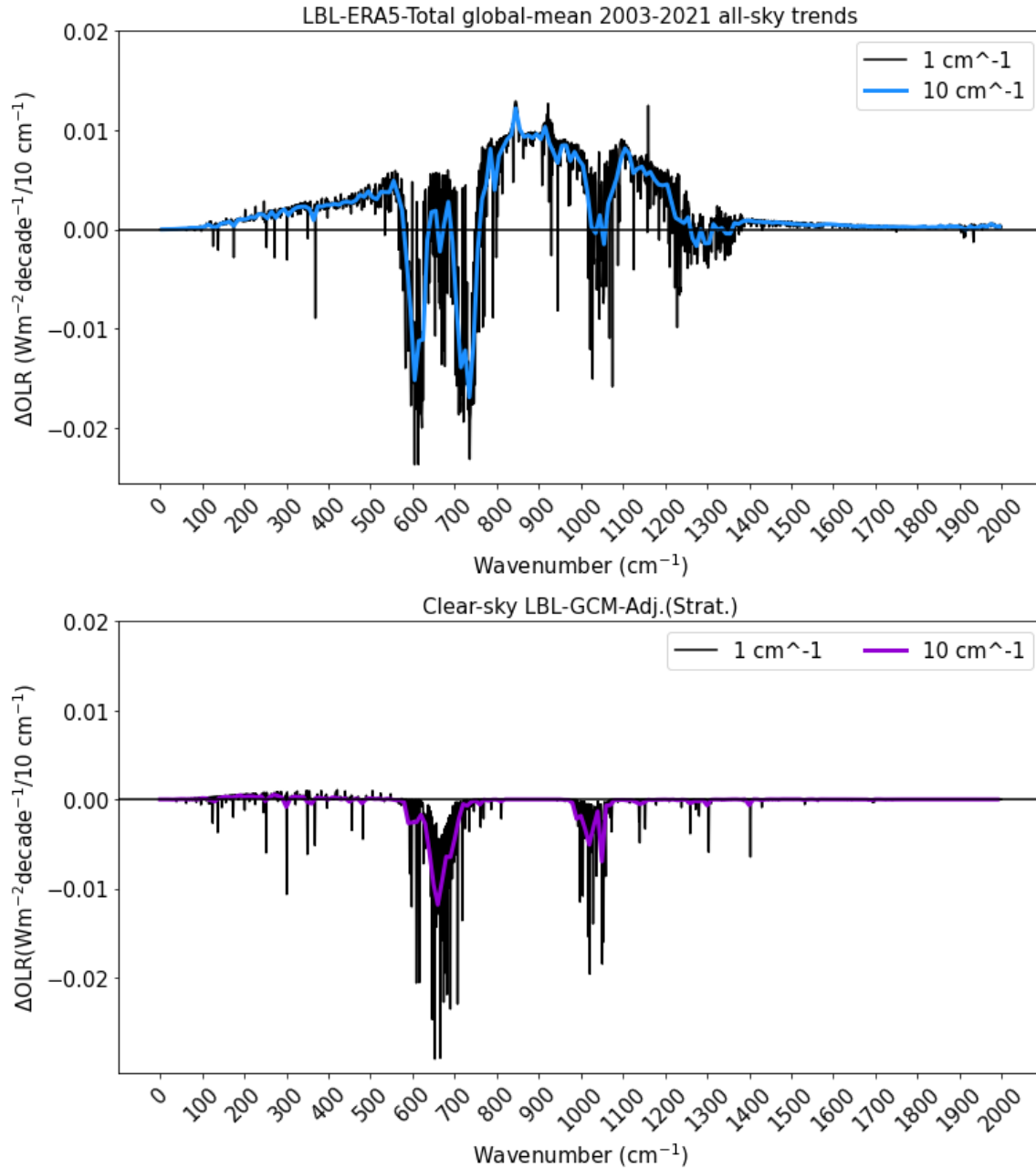
Supplementary Figure S8: Summed over wavenumber ranges in Figure 2c.



Supplementary Figure S9: RH profiles (units %) in GFDL CM3 at Control and TCR states.



Supplementary Figure S10: Global-mean climatology and trends in LWCRE in LBL-ERA5 and AIRS. a) Climatology. b) Trends.



Supplementary Figure S11: a) All-sky, global-mean, spectral OLR trends during 2003-2021 in LBL-ERA5-Total at 1 cm⁻¹ and 10 cm⁻¹ resolution. b) Clear-sky, global-mean, spectral OLR trends during 2003-2021 in LBL-GCM-Adj. (Strat) at 1 cm⁻¹ and 10 cm⁻¹ resolution.

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