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Direct Observational Evidence from Space of the Effect of CO_2 Increase on Longwave Spectral Radiances: The Unique Role of High Spectral Resolution Measurements

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Abstract. We present a direct measurement of the impact of increased atmospheric CO_2 on the spectra of Earth's longwave radiation obtained from space. The goal of this study is to experimentally confirm that the direct effects of CO_2 increase on the Earth's outgoing longwave spectra follow theoretical estimates, by developing a methodology that allows for a direct and more precise comparison between theory and observations. In this methodology, a search is performed to find selected ensembles of observed atmospheric vertical profiles of temperature and water vapor that are as close as possible to each other in terms of their values. By analysing the spectral radiances measured from space by the Atmospheric Infrared Sounder (AIRS), corresponding to the selected ensembles of profiles, the effects of increased CO_2 on the spectra can be isolated from the temperature and water vapor effects. The results illustrate the impact of the increase of CO_2 on the longwave spectra and compare well with theoretical estimates. As far as the authors are aware, this is the first time that the spectral signature of the increase of CO_2 (isolated from temperature and water vapor changes) has been directly observed from space.

1 Introduction

As is clearly discussed in the excellent historical compilation of Archer and Pierrehumbert (2011), and the essential references therein, it has been known for decades that an increase of atmospheric greenhouse gases such as CO_2 can lead to global warming essentially by changing the longwave radiative fluxes of energy at the top of the atmosphere. While remarkable progress has been made over the last few decades in laboratory, theoretical, modelling and prediction studies of the physics of climate change (e.g., Plass, 1956; Manabe and Wetherald, 1967, 1975; Hansen et al., 1984; Ramanathan, 1988; Ramaswamy et al., 2018), the experimental confirmation from space of the direct effects of CO_2 (independent from temperature and water vapor changes) on the Earth's outgoing longwave spectra has been elusive. The goal of the present study is to experimentally confirm that the direct effects of CO_2 increase on the Earth's outgoing longwave spectra follow theoretical estimates, by developing a methodology that allows for a direct and more precise comparison between theory and observations.

Kiehl (1983) discussed the possibility of utilizing spectrally resolved satellite measurements of longwave radiation to detect and characterize the impact of climate change on the longwave spectra, and simulated the changes in clear-sky spectra due to

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increases in CO_2 and temperature. This pioneering modelling study has been followed by other modelling studies focused on the spectral signature of climate change (e.g., Mlynczak et al., 2016; Brindley and Bantges, 2016).

The fact that measurements from space of the direct effects of increased CO_2 on the longwave spectra have been notoriously difficult to obtain is associated with the sparsity of high spectral resolution observations of longwave radiances before the early 21st century and with the challenge of disentangling the effects of CO_2, temperature, and water vapor on the spectral radiances. While measurements of the spectral effects of the combined changes in CO_2, temperature, water vapor and other gases have been published (e.g., Harries et al., 2001; Brindley and Bantges, 2016; Strow and DeSouza-Machado, 2020; Whitburn et al., 2021; Huang et al., 2022), the direct effects of CO_2 alone have been difficult to depict accurately.

For example, in a pioneering study, Harries et al. (2001) calculated the spectral differences between two infrared instruments, one launched in the 1970s and the other in the 1990s. Despite the difficulties of accurately estimating spectral differences between two different instruments, Harries et al. (2001) are able to discern and potentially assign, using model simulations, some of the spectral differences to changes in greenhouse gases such as CO_2. However, they do not attempt to disentangle the effects due purely to CO_2, from temperature and water vapor changes, in the observational data.

The recent studies of Strow and DeSouza-Machado (2020) and Huang et al. (2022) investigate in detail the AIRS instrument radiance trends over the last several years and highlight the remarkable stability of the AIRS radiance record, while also discussing the role of temperature, water vapor, CO_2, and other gases in framing the nature of the AIRS radiances. However, they only isolate these effects using modelling/theoretical approaches. These studies do not attempt to disentangle the impact of CO_2 (or other gases) from temperature and water vapor in the observational data.

In the present study, a new methodology is proposed for a more direct measurement of the effect of CO_2 increase on longwave spectral radiances in such a way as to provide a direct and more precise comparison to theoretically derived radiances. The goal of this approach is to isolate the effects of CO_2 from the effects of temperature and water vapor. This is achieved by searching for atmospheric profiles of temperature and water vapor that are as close to each other as possible (referred to as analogues), but that have CO_2 concentrations that are significantly different. Measuring from space the spectral radiances that correspond to these analogues allows to detect, given the right circumstances, the unique impact of CO_2 on the radiances with enough precision and accuracy in key spectral regions. Given the steady increase of global CO_2, the best way to make sure that analogues with significantly different values of CO_2 are selected, is to search for these analogues over several years.

Two specific approaches are explored. In experiment A, a single set of individual temperature and water vapor profiles is selected (the reference profiles), and a search is performed for analogues that fit within a specific uncertainty range. In experiment B, to increase the number of samples, this approach is extended to 100 sets of reference profiles, followed by a search for analogues that fit these reference profiles to within a slightly less strict uncertainty range.

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2 Observational Data

The spectral longwave radiances are measured by the Atmospheric Infrared Sounder (AIRS), and the profiles of atmospheric temperature and water vapor, as well as cloud properties, are from the AIRS and AMSU (Advanced Microwave Sounding Unit) suite of instruments on Aqua (e.g., Chahine et al., 2006) jointly with other datasets – see appendix for details.

3 Experiment A: Single Set of Reference Profiles

For experiment A, a search is performed, from 2005 to 2015, for temperature and water vapor profiles within specific Root-Mean-Square (RMS) difference thresholds, with respect to a set of reference profiles. 148 analogues are selected for a specific set of reference profiles (see appendix for details), corresponding to thresholds of 1.2 K for temperature and SST, and 1.2 gkg^{-1} for water vapor (with cloud cover less than 10 %). For each of these analogue profiles, the corresponding cloud-cleared (e.g., Susskind et al., 2003) AIRS spectral radiances are selected. From the ensemble of 148 analogues, to stay as close to nadir as possible, only spectra measured with scan-angles between -5° and 5° from nadir are selected, which corresponds to six 2005 analogues to estimate the 2005 mean and 18 (unevenly distributed across different years) analogues for all the years between 2006 and 2015.

To estimate the impact of CO_2 increase on the radiances: (1) the spectral radiance mean of the 2005 analogues is calculated, and (2) for spectral radiances for each analogue after 2005, the differences between each of these analogues and the 2005 mean are calculated and used to estimate the average annual difference. These differences between the spectral radiances - that are measured at different years and as such reflect different amounts of CO_2 - are compared with theoretical estimates of the effect of CO_2.

To estimate the theoretical values, the spectral radiances corresponding to the reference 2005 temperature and water vapor profiles are simulated with different values of CO_2 that reflect its increase from 2005 to 2015. The kCARTA forward model (Strow et al., 1998; DeSouza-Machado et al., 2020) is used to simulate the spectral radiances and is convolved with the AIRS spectral response functions to get theoretical AIRS radiances.

The theoretical spectral radiances are constructed using the corresponding (observed) monthly mean CO_2 and standard deviation as described in the appendix. For each theoretical analogue (including the ones corresponding to 2005) a value of CO_2 is drawn from a normal probability density function (PDF) defined by its observed mean and standard deviation, to represent CO_2 uncertainty. To match how the observed differences are calculated, the 2005 mean radiance spectrum is subtracted from each theoretical spectra (from 2006 to 2015) to calculate the theoretical annual mean spectral differences.

Figure 1 shows the annual mean differences (between years 2006 to 2015 and the mean 2005 values) in terms of spectral radiances for the AIRS observations (mean and standard deviation of the mean) and the expected theoretical values (mean and standard deviation) given CO_2 uncertainties as discussed. Note that the figure shows the corresponding theoretical curves using the methodology described for 100 realizations – i.e., showing the averages of 100 means and standard deviations of theoretical spectral radiances.

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This figure is focused on the 680 to 780 cm^{-1} spectral range, which represents a wing of the 15 μm CO_2 band and is a spectral region where the CO_2 signal is particularly significant. In this spectral region, the enhanced absorption within the troposphere, where temperature decreases with height, leads to a reduction of the outgoing radiation. From a broader climate perspective, the reduction of outgoing radiation is behind the increase of global surface temperature that is necessary for the overall climate system to re-establish energy balance at the top of the atmosphere, and as such is a critical component of global warming. Note that, during this period, the measured monthly mean CO_2 mole fraction at Mauna Loa increased on average by approximately 2 ppm per year (see appendix).

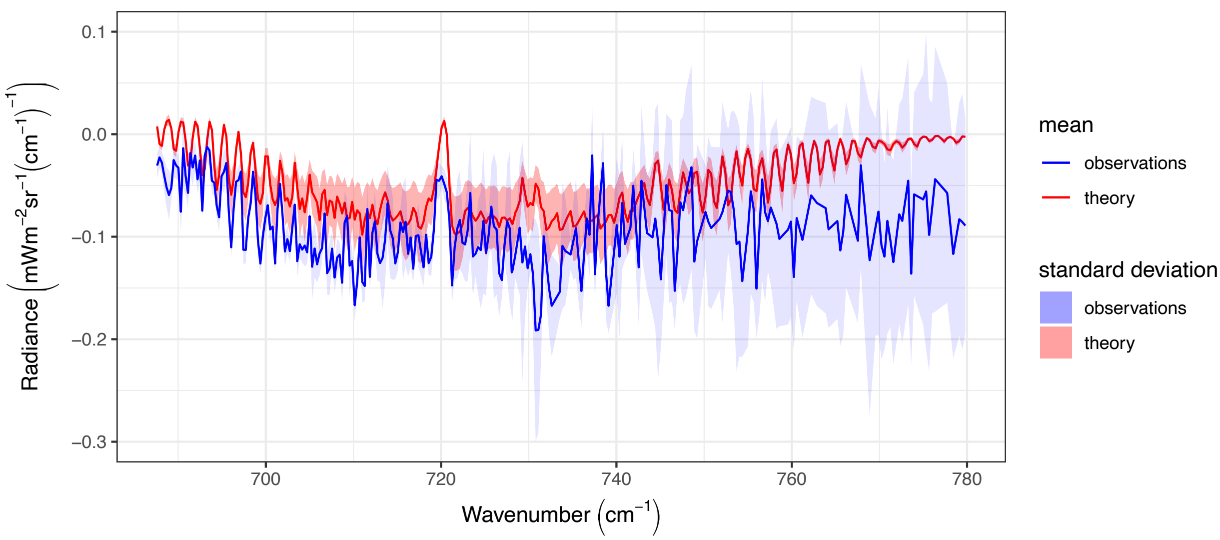

Figure 1: Annual mean radiance differences (in mWm^{-2} sr^{-1} (cm^{-1})$^{-1}$), from the AIRS observations (blue line) and from theory (red line), and standard deviations (blue and red shading), between years 2006-2015 and the mean 2005 values, following the methodology described in the text (experiment A) and illustrating the direct impact of CO_2 increase on the spectral radiances during this period (see text for details). Based on observations with scan-angle between -5º and 5º.

Figure 1 illustrates that this methodology leads to observed radiance differences that, although often displaying a negative bias and a non-negligible amount of noise, are close to the theory given the uncertainty estimates. This is particularly significant given the small sample of analogues. These results also illustrate how CO_2 uncertainty impacts the theoretical radiance estimates. While the results illustrate the potential of this approach, the observations show a significant bias and uncertainty in the 760-780 cm^{-1} region due to the large role of water vapor in this region.

4 Experiment B: Multiple Sets of Reference Profiles

To increase the sample size, 100 sets of temperature and water vapor reference profiles, from June-July-August (JJA) 2003 and within an SST range of 298 K to 302 K, are selected. For each of these 100 reference profiles a search is performed to

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find analogue profiles that are within RMS thresholds of 1.4 K for temperature and 1.4 gkg^{-1} for water vapor at any vertical level (with respect to each of the reference profiles). These thresholds are less strict than for experiment A. The search spans a 10 year period from 2003 to 2012, but only includes the JJA period for each year. As in experiment A, these profiles occur in the tropical/subtropical oceans, in (almost) clear sky (cloud cover less than 10 %) and that the SST values are within 1.4 K. For each of these analogue profiles, the corresponding cloud-cleared (e.g., Susskind et al., 2003) AIRS spectral radiances are selected.

Following experiment A, only spectra measured within scan-angles between -5° and 5° from nadir are selected (see appendix) and to estimate the impact of CO_2 increase on the spectral radiances, the following procedure is executed: For each set of analogues corresponding to each of the reference profiles (1) the spectral radiance mean of the 2003 reference state and analogues is calculated, and (2) for spectral radiances for each analogue after 2003, the differences between each of these analogues and the corresponding 2003 mean are calculated and used to estimate the average annual difference. These annual difference means for each reference state, are combined to estimate a single annual difference mean in terms of spectral radiances for all reference states. These differences between the spectral radiances are compared in figure 2 with theoretical estimates of the effect of CO_2. In experiment B, a key assumption is that the annual mean spectral radiance differences corresponding to each reference state are (to first order) not sensitive to the reference state itself for these selected reference profiles.

The theoretical spectra are determined using kCARTA simulations of the radiances for each of the 100 temperature and water vapor reference profiles for two values of CO_2: mean JJA for 2003 and 2012 as measured by the NOAA Mauna Loa station (see appendix).

Figure 2 is similar to figure 1 in that it shows annual mean radiance differences, from the AIRS observations (blue line) and from theory (red line) for the 680-780 cm^{-1} spectral region, but for a larger sample. Specifically, the focus is on approximately 200 of the closest analogues to the mean reference states, which represents a sample that is about one order of magnitude larger than in figure 1. The theoretical annual mean differences are calculated based on the reference states. This allows to estimate not only the theoretical annual mean difference but also the associated standard deviation, which is shown as red shading. Note that the standard deviation is so small that it is almost imperceptible in the figure. This apparent lack of theoretical sensitivity to the reference states supports the key assumption, mentioned above, that is made in experiment B.

Despite some noise and a small negative bias in some spectral regions, there is good agreement between theory and observations in figure 2, with the observations following closely the theoretical impact of increased CO_2. It is noticeable that in the 760 to 780 cm^{-1} spectral region - where water vapor plays a large role, and the observations show a clear negative bias in figure 1 - the observations in figure 2 match the theory to a good level of accuracy. The theoretical uncertainty associated with the lack of accurate knowledge of the CO_2 values for the reference states and analogues is not shown in figure 2. This is done to highlight the theoretical uncertainty associated with the sensitivity to the reference states (which is small). But the impact of CO_2 uncertainty is still present in the data analysed for figure 2, and the observations, in some regions where there

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is a negative bias, still seem to be within the standard deviation range shown in figure 1. This suggests that the slightly negative observational biases are at least partly related to uncertainties about the CO_2 values of the analogues.

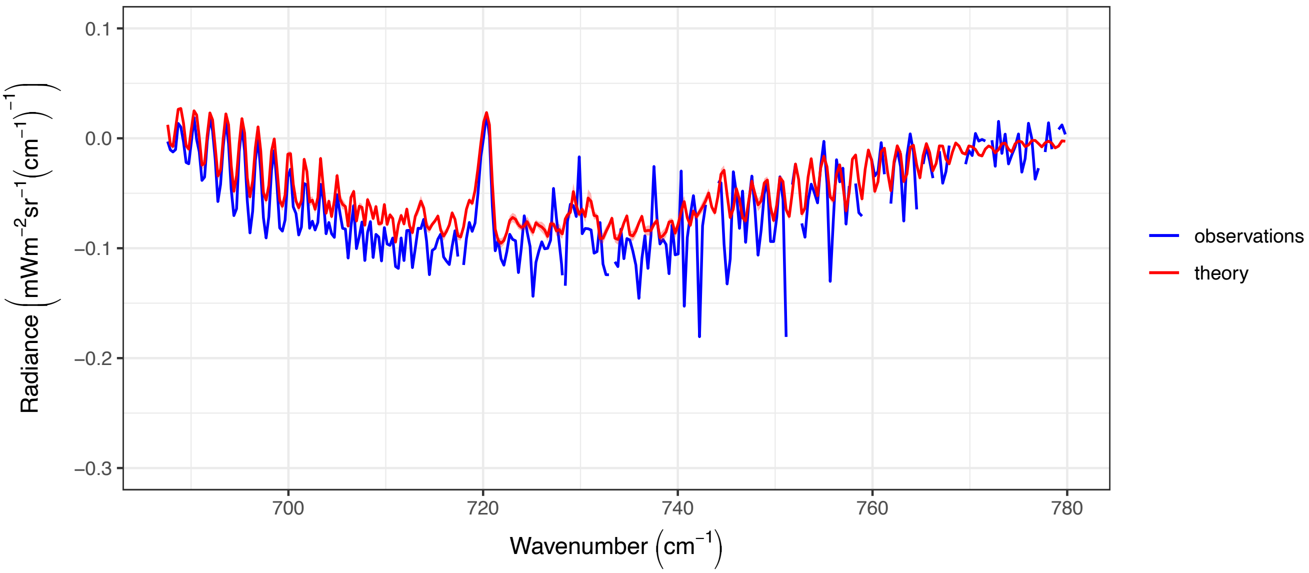

Figure 2: Annual mean radiance differences (in mWm^{-2} sr^{-1} (cm^{-1})$^{-1}$), from the AIRS observations (blue line) and from theory (red line for the mean and red shading for the standard deviation), following the methodology described in the text for experiment B, and illustrating the direct impact of CO_2 increase on the spectral radiances during the 2003-2012 period (see text for details). Based on observations with scan-angle between -5° and 5°.

5 Conclusions

We present a direct measurement of the impact of increased atmospheric CO_2 on the spectra of Earth's longwave radiation obtained from space. The approach involves a new methodology to disentangle the impact of CO_2 on the observed longwave spectral radiances, from the effects of temperature and water vapor, in such a way as to provide a direct and more precise comparison with theoretical estimates of the radiance impact of CO_2. The observations obtained using this methodology compare well with theoretical estimates of the direct CO_2 radiative impact on the Earth's longwave spectra. In the future, variants of this methodology could be used to isolate the observational radiative impact of different physical and chemical properties of the climate system and as such provide a better observational depiction of the Earth's radiative forcing and of climate feedbacks.

The stability of the AIRS instrument has been determined to be about one order of magnitude smaller (better) than the climate temporal signal for this spectral region (Strow and DeSouza-Machado, 2020) providing a high level of confidence for the results presented. This work also illustrates the unique and critical role of accurate and stable hyperspectral infrared observations from space in addressing fundamental climate physics questions.

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As far as the authors are aware, this study represents the first attempt to establish a more precise experimental confirmation from space of the direct effects of CO_2 on longwave spectral radiances. The results (solely based on observations) confirm that the effects of the recent atmospheric CO_2 increase on longwave spectral radiances follow theoretical estimates. As such, these results confirm a critical foundation of the science of global warming.

Appendix: Data and Methods

In this work, the focus is on sets of individual (instantaneous and localized) temperature and water vapor profiles (and the corresponding spectral radiances) that belong to a common physical regime associated with clear sky (or negligible cloud amounts) atmospheric thermodynamics over the tropical and subtropical oceans (from 30º S to 30º N).

AIRS is a hyperspectral sounder on the Aqua spacecraft (Parkinson, 2003) covering the 3.7-15.4 µm infrared spectral region with 2378 channels and spatial resolution of 13.5 km at nadir. AIRS was launched into a 705 km altitude orbit on May 4, 2002, and has been in routine data gathering mode essentially uninterrupted since September 2002. The 1:30 PM ascending node and orbital altitude of the Aqua spacecraft orbit have been accurately maintained (until 2022) and daily (nearly) global coverage is essentially achieved from the ascending and descending orbits. The AIRS absolute radiometric calibration accuracy is around 0.2 K in brightness temperature (e.g., Aumann et al., 2006). AIRS radiances are routinely assimilated in all major global weather prediction systems and are used to retrieve vertical profiles of atmospheric temperature, water vapor, and key atmospheric constituents as well as cloud and surface parameters (e.g., Susskind et al., 2003; Smith and Barnet, 2020).

Recently, Strow and DeSouza-Machado (2020) confirmed that the AIRS instrument stability for about 400 channels is within 2.10^{-3} $Kyear^{-1}$ in brightness temperature, which is about one order of magnitude smaller than the climate temporal signal in brightness temperature for the spectral region that is investigated in this study. Note that, according to Huang et al. (2022), the trend due to the AIRS instrument spectral shift is also within 2.10^{-3} $Kyear^{-1}$.

The Advanced Microwave Sounding Unit (AMSU) on Aqua is a 15-channel microwave (MW) instrument with 12 temperature sounding channels in the 50-58 GHz oxygen absorption band, that are used to produce an AMSU MW-only retrieved temperature profile dataset (Rosencranz 2001) and are also part of the AIRS/AMSU retrieval.

The atmospheric profiles of water vapor used in this study are from the combined AIRS/AMSU retrieval products that are based on level 1 data from AIRS and AMSU, as well as on a neural network retrieval first-guess (Milstein and Blackwell, 2016) that uses the European Centre for Medium-range Weather Forecasts (ECMWF) analyses as the training dataset. It is fair to state that the AIRS/AMSU retrieved profiles of atmospheric temperature and water vapor depend directly on AIRS and AMSU data, and indirectly on the ECMWF data-assimilation system as well as on a variety of observational datasets that are assimilated (e.g., radiosondes, Global Navigation Satellite System (GNSS) Radio Occultation (RO), other infrared (IR) and MW sounders) via a neural network retrieval algorithm. Specifically, version 6 of the AIRS and AMSU retrieved

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products is used. The standard pressure levels for the retrieved temperature and water vapor profiles are described in: https://docserver.gesdisc.eosdis.nasa.gov/public/project/AIRS/V7_L2_Standard_Pressure_Levels.pdf.

For the temperature profiles, both the AIRS/AMSU as well as the AMSU MW-only products are analysed. Although the AIRS/AMSU retrievals utilize a large variety of different sources of information about the atmosphere (as described above), the AMSU MW-only retrievals (that are based on the oxygen band and have no dependency on CO_2) are also used as an additional (somewhat) independent temperature profile dataset.

The fact that the analogues have similar characteristics in their temperature profiles in both the AIRS/AMSU (that use a variety of sources of information) and the AMSU MW-only datasets (that are based on the oxygen band) suggests that the analogues are in practice independent of specific CO_2 assumptions in the retrieval algorithms.

Note that the temperature and water vapor profile retrieved products mentioned above are only used to find analogues and, in this context, other observational estimates of temperature and water vapor could be used for this purpose: e.g., from other IR sounders such as the Infrared Atmospheric Sounding Interferometer (IASI) or the Cross-track Infrared Sounder (CrIS), from other microwave sounders, or from analysis and re-analysis products. The key reason why AIRS/AMSU and AMSU temperature and water vapor profiles are used is the inherent collocation with the AIRS radiance spectra which are directly used in this study.

For experiment A, a few sets of reference profiles of temperature and water vapor were tested. For RMS thresholds of 1 K for temperature and 1 gkg^{-1} for water vapor (mixing ratio) at every vertical level, 7 analogue profiles for a particular set of reference profiles are found (the largest number of analogue profiles attained with this criteria), which is a sample that is too small for a meaningful analysis. Relaxing the thresholds to 1.2 K and 1.2 gkg^{-1}, leads to a much larger number of analogues (148) for that set of reference profiles. These profiles occur in (almost) clear sky (cloud cover less than 10 %) and the sea surface temperature (SST) values are within 1.2 K. This is the set of analogues that is analysed in experiment A. All 148 analogue profiles are from June, July, or August.

For figure 1, the theoretical spectral radiances are calculated for three different values of CO_2 that correspond to the mean July CO_2 values for 2005, 2010 and 2015, as measured by the National Oceanic and Atmospheric Administration (NOAA) at Mauna Loa (e.g., Thoning et al., 1989). A function is fitted to these three sets of spectral radiances to be able to estimate spectral radiances for any other value of CO_2. Since the actual CO_2 values are not accurately known for each individual analogue profile, the monthly mean observed CO_2 values (at Mauna Loa) of the corresponding month and year are used.

The uncertainty associated with the lack of accurate knowledge of CO_2 values for each analogue profile can lead to noteworthy uncertainties in the theoretical spectral radiances. The CO_2 uncertainty for each theoretical analogue (due to CO_2 variability in space and time) is estimated in the following way: from the Orbiting Carbon Observatory-2 (OCO-2) data (Crisp et al., 2004), a value of 1.7 ppm is obtained as an approximate estimate of the spatial standard deviation, and to this value an additional 0.5 ppm is added as a rough estimate of the temporal variability within the June-July-August period.

For perspective, when analysing figure 1, note that changes in temperature and water vapor of the order of the ones experienced by the atmosphere during the first 20 years of the 21st century lead to positive theoretical changes in brightness

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temperature that are fairly constant in this spectral interval, and that correspond to approximately 25% (in absolute value) of the CO_2 theoretical changes in the 720-740 cm^{-1} region, increasing (in percentage) to much higher values toward both the 680 cm^{-1} and the 780 cm^{-1} extremes of the figure (e.g., Huang et al 2022).

As mentioned in the main text, only the AIRS observed radiances measured within a scan angle of -5° to 5° are used. To examine the sensitivity to scan-angle selection, results with scan-angles between -10° and 10° from nadir are also analysed for experiment A (not shown), which correspond to 7 analogues for the 2005 mean and 27 analogues for years between 2006 and 2015. Overall, the results with scan-angles between -10° and 10° are fairly similar to figure 1, with a worse negative bias (compared to theory) in the region from 700 to 740 cm^{-1}, and a slightly less negative bias in the region from 760 to 780 cm^{-1}.

For experiment B (as in experiment A) to stay as close to nadir as possible, only spectra measured with scan-angles between -5° and 5° from nadir are selected, which leads to a reduction from 100 to 55 reference profiles (states). This is because for 45 of these reference states not a single analogue, for years after 2003, is found within scan-angles of -5° to 5°.

For figure 2, to remove outliers and to select analogues that are as close as possible to the reference states, analogues that have absolute radiance differences, as compared to the reference states, that are larger than 0.5 $mWm^{-2}sr^{-1}(cm^{-1})^{-1}$ are filtered out. Results with a larger filter value of 1 $mWm^{-2}sr^{-1}(cm^{-1})^{-1}$ are similar to the ones with 0.5 $mWm^{-2}sr^{-1}(cm^{-1})^{-1}$ from 680 to 740 cm^{-1} but slightly noisier above 740 cm^{-1} where water vapor plays a larger role. Note that the theoretical radiance differences are about 50 times smaller than the filter value in the spectral region 770-780 cm^{-1} where the observational noise is more significant when compared to theory.

These results provide evidence that (i) AIRS has the stability required to address in an accurate and precise manner, climate change questions of the nature described here, and that (ii) space based spectral measurements are becoming of comparable quality to prior spectroscopic estimates. Note that Strow and DeSouza-Machado (2020) estimate that the trend uncertainty due to CO_2 spectroscopy uncertainties is of the order of their estimate for the stability of the AIRS instrument in these channels at around 2.10^{-3} $Kyear^{-1}$. The similarity between the observations and theory in figure 2 further supports this point.

Code availability: The kCARTA model is available at (SRCv1.18 was used): https://github.com/sergio66/kcarta_gen.

Data availability: The AIRS/AMSU version 6 and AMSU MW-only L2 standard products are used for the temperature and water vapor profiles: https://doi.org/10.5067/Aqua/AIRS/DATA201. For AIRS L1B infrared radiances (version 5): https://doi.org/10.5067/YZEXEVN4JGGJ.

Author contributions: JT developed the methodology, performed the analysis, and wrote the manuscript. RCW and HTT implemented the methodology and contributed to the analysis and the manuscript.

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