1 2	Trends in Downwelling Longwave Radiance over the Southern Great Plains
3	Lei Liu <sup>1</sup> , Yi Huang <sup>1</sup> , John R. Gyakum <sup>1</sup> , David D. Turner <sup>2</sup> , P. Jonathan Gero <sup>3</sup>
4	<sup>1</sup> Department of Atmospheric and Oceanic Sciences, McGill University, Quebec, Canada.
5	<sup>2</sup> NOAA/OAR/Global Systems Laboratory, Boulder, Colorado, USA.
6 7	<sup>3</sup> Space Science and Engineering Center, University of Wisconsin-Madison, Madison, Wisconsin, USA.
8	
9	Corresponding author: Lei Liu (lei.liu5@mail.mcgill.ca)
10	
11	Key Points:
12 13	• Long-term AERI measurements disclose distinctive DLR trends, demonstrating the advantages of the spectral data for climate monitoring.
14 15	• The changes in clear/cloudy sky fractions offset DLR changes caused by warming and increases in greenhouse gases.
16 17	• The radiance trend uncertainty mainly results from the natural variability, which emphasizes the need to continue the measurements.

## 18 Abstract

- 19 Downwelling longwave radiation (DLR) is an important part of the surface energy budget.
- 20 Spectral trends in the DLR provide insight into the radiative drivers of climate change. In this
- 21 research, we process and analyze a 23-year downwelling longwave radiance record measured
- 22 by the Atmospheric Emitted Radiance Interferometers (AERI) at the Southern Great Plains
- 23 (SGP) site of the Atmospheric Radiation Program. Two AERIs were deployed at SGP with
- an overlapping observation period of about 10 years, which allows us to examine the
- consistency and accuracy of the measurements and to characterize discrepancies between
- them due to undetected instrumentation errors. Using the 23-year record, we analyze the allsky radiance trends in DLR, which reflects the associated surface warming trend at SGP
- 28 during this same period and also the complex changes in meteorological conditions. For
- 29 instance, the observed radiance in the CO<sub>2</sub> absorption band follows closely the near-surface
- 30 air temperature variations. The changes in the sky fraction of clear-sky and thick cloudy-sky
- 31 scenes offset the radiance changes in the window band. Our analysis shows that the radiance
- 32 trend uncertainty in the DLR record to date mainly results from the climate internal
- 33 variability rather than the measurement error, which highlights the importance of continuing
- 34 the DLR spectral measurements to unambiguously detect and attribute climate change.

## 35 1 Introduction

36 Longwave radiation is a key component of the atmospheric energy budget that drives 37 climate change. At the top of the atmosphere (TOA), the outgoing longwave radiation (OLR), 38 as well as its spectrally resolved radiance, is monitored by satellites with global coverage and 39 long-term records (e.g., Liebmann & Smith, 1996; Stephens et al., 2012). This allows us to 40 study the changes in OLR and to test climate models (e.g., Harries et al., 2001; Huang & 41 Ramaswamy, 2009; Huang, Ramaswamy, Huang, et al., 2007; Huang, Ramaswamy, & 42 Soden, 2007; Wielicki et al., 2002). Despite the continuous spatiotemporal coverage of OLR 43 spectra, the compensating effects of greenhouse gas opacity and temperature warming make 44 it difficult to detect climate change (Huang & Ramaswamy, 2009).

45 Downwelling longwave radiation (DLR) emitted by the atmosphere is one key 46 component in the surface energy budget (Stephens et al., 2012; Trenberth et al., 2009). 47 Compared to the radiation budget at the TOA, the surface radiation budget is more uncertain 48 and DLR is a main contributor to the uncertainty (Trenberth et al., 2009; Wild et al., 2012). 49 This is largely due to the lack of global and long-term observations of DLR. DLR 50 observations, especially spectrally resolved radiance, have been limited to specific locations. 51 Despite the limited records, it has been demonstrated that DLR measurements are useful for 52 understanding the surface energy balance and testing the climate models. For example, Lubin 53 (1994) explained the super greenhouse effect using the observed DLR spectra over equatorial 54 oceans; Feldman et al. (2015) used the DLR spectra to measure CO<sub>2</sub> radiative forcing at the 55 Southern Great Plains (SGP) and the North Slope Alaska sites; Shupe and Intrieri (2004), 56 Kapsch et al. (2016), Huang et al. (2019), Sokolowsky et al. (2020) and a number of others 57 diagnosed the DLR variability in relation to sea ice, clouds and other climate changes in polar 58 regions.

59 Climate change is driven by changes in energy balance. This leads us to an 60 overarching question regarding the surface energy balance: can climate change be detected 61 and understood by monitoring the DLR spectrum? One advantage of the DLR, compared to 62 the OLR, is that the compensating effects mentioned earlier vanishes. In the DLR, the 63 greenhouse gas opacity and temperature warming effects reinforce each other to increase 64 DLR. This makes DLR a potentially advantageous means for monitoring climate change

- 65 (Huang, 2013). The signals from different meteorological variables such as temperature,
- 66 greenhouse gases and clouds imprint different spectral signatures. This allows for a spectral
- 67 fingerprinting of their changes (Huang et al., 2010). At the SGP site, the fifth generation
- 68 European Centre for Medium-Range Weather Forecasts atmospheric reanalysis dataset,
- ERA5 (Hersbach et al., 2020), shows that there has been a significant warming in surface air
- temperature with a magnitude of  $\sim 0.045$  K/year between 1996 and 2018 (Figure 1). Can this
- 71 warming be detected from the DLR spectral records?



72

Figure 1. Warming trend at SGP. Shown here is the ERA5 monthly mean 2-meter air
temperature time series at the SGP site (average of nine 0.25°x0.25° resolution grid boxes
centered at: 97.5° W and 36.5° N) between 1996 and 2018. The anomaly is defined with
respect to multi-year monthly mean of each calendar month.

77 We have two primary objectives in this paper. First, we are interested in constructing 78 a long-term monthly DLR spectral record based on the 23 years of measurements by the 79 Atmospheric Emitted Radiance Interferometers (AERIs) installed at the SGP site of the 80 Atmospheric Radiation Measurement (ARM) program of the U.S. Department of Energy. 81 Two AERI instruments have been deployed at this site and have rendered 10 years of 82 overlapping observations but with different sampling strategies (e.g., 3 min sky average every 83 8 minutes vs multiple 20-s sky average observations every 4 minutes). We will examine the 84 accuracy and consistency of the measurements and validate them against synthetic spectra 85 simulated from collocated atmospheric measurements using a benchmark radiation model. Second, we will show the analysis of the long-term DLR spectral trends measured by the two 86 87 AERIs for the period of 1996-2018. We are interested in whether the radiance trends are in 88 concert with the warming temperature trend (Figure 1). This work will also verify the trends 89 documented by Gero and Turner (2011) using the early years of the DLR record and analyze

90 the contributions from different sky conditions.

## 91 **2 Data and Methods**

92 2.1 AERI data processing

93 The AERI is a Fourier transform spectrometer that measures the DLR radiance 94 emitted from the atmosphere with good accuracy at high temporal and spectral resolution 95 (Knuteson et al., 2004a, 2004b). The measurements cover the spectral range between 520 and 96 3020 cm<sup>-1</sup> with a resolution of 0.5 cm<sup>-1</sup>. Two high-emissivity blackbodies, a hot blackbody 97 with a fixed temperature at around 60 degrees Celsius and another blackbody at ambient 98 temperature (Knuteson et al., 2004a), are used for radiometric calibration based on the 99 method of Revercomb et al. (1988). The long-term annual mean DLR spectra and the standard deviation of DLR spectra for different sky types (classification method explained 100 101 later) at SGP site are shown in Figure 2. The main difference in DLR between different sky 102 types is primarily in the window portion of the spectrum (between  $800 - 1200 \text{ cm}^{-1}$ ) shown in Figure 2a. The standard deviation of thick cloudy-sky DLR is found to be the smallest among 103 all the different sky types in the window band (Figure 2b), which indicates small variability 104 of the radiating temperature of the thick clouds. We focus on the mid-infrared spectral range 105 from 520 to  $1800 \text{ cm}^{-1}$  in this paper. 106



107

108Figure 2. (a) Annual mean AERI spectra for different sky types at SGP. (b) Standard109deviation of monthly mean AERI spectra for different sky types at SGP. (RU: Radiance110Units;  $1 \text{ RU} = 1 \text{ mW/(m}^2 \text{ sr cm}^{-1})$ )

111 The two AERIs deployed at SGP have different observational periods and different 112 sampling frequencies. AERI-01 operated from July 1995 to March 2014, while AERI-C1 has 113 operated from February 2004 to present. C1 is the current name of the Central Facility 114 location of SGP site which is used to be called E14, e.g. in Gero and Turner (2011). The two 115 AERIs were deployed virtually side-by-side (within 5 meters of each other). Given their field 116 of view (FOV) of 1.3 degrees, both instruments view essentially the same portion of the sky.

- 117 The overlapping observations make it possible to test the accuracy and consistency of the
- 118 measurements. However, the two instruments differ with respect to their sampling frequency.
- 119 AERI-01 measures one DLR spectrum approximately every 8 minutes; its measurement cycle
- 120 includes a 200-second sky-dwell period (Knuteson et al., 2004b) and the rest of the cycle is
- 121 used for viewing the blackbodies for calibration. AERI-C1 uses a rapid mode with a  $\sim 20$ -
- second sampling cycles (Turner et al., 2006). Such differences in the measurements necessitate appropriate procedures to homogenize the data from the two AERIs for inter-
- necessitate appropriate procedures to nomogenize the data from the two AERIS for inter-
- 124 comparisons and trend analyses.



125

- 126 **Figure 3.** Data processing flowchart. Yellow and purple squares represent AERI-01 and
- 127 AERI-C1 DLR data respectively. Blue squares represent important data processing steps.
- 128 Pink squares represent radiative transfer model simulations. (see texts for the details)
- 129 Figure 3 shows the flowchart of AERI data processing adopted in this paper. First,
- 130 rigorous quality control is performed on the data to retain reliable observations. During the
- 131 long history of observations at the SGP site, many factors have caused errors including: the
- 132 contamination of the scene mirror, malfunction of the interferometer, the failure of the
- 133 detector temperature sensor, and so on. We first discard all the erroneous data based on the
- 134 AERI quality control reports from the ARM program
- 135 (https://adc.arm.gov/discovery/#/results/instrument\_class\_code::aeri). In addition, similar to
- 136 the quality control method described in Turner and Gero (2011), the hatch status and the sky
- 137 view radiance variability are also implemented.

After the *Quality Control* step, we average the AERI-C1 spectra over 8-min intervals, to be consistent with the AERI-01 sampling period. Then, in the *Sky Classification* step, we apply a machine learning algorithm (detailed below) to classify the sky types (clear, thin and thick clouds) based on the 8-min mean radiance spectra. Next, we take hourly averages of the radiance data and verify proper diurnal sampling in each month to ensure no missing data for any 24-hour period. Then the monthly mean spectra are obtained by averaging the 24-hourly spectra of each day during the given month. Monthly means are discarded when the count of

145 hourly spectra is below  $400 (\sim 55\%)$ .

146 Some channels in the center of CO<sub>2</sub> absorption band (around 667 cm<sup>-1</sup>) and water 147 vapor absorption band  $(1300 - 1800 \text{ cm}^{-1})$  in which the near-surface atmosphere is so opaque that the channels are essentially uncalibrated are discarded based on the criterion that the 148 149 gaseous optical depth for a 1-meter layer of atmosphere at the surface is above 0.5 in the 150 Optical Depth Screening step. Finally, the monthly anomaly spectra are obtained by 151 subtracting the long-term monthly spectra of each calendar month (which effectively removes 152 the seasonal cycle). The results are illustrated in Figure 4. The long-term trends in the DLR 153 spectra are analyzed based on the monthly anomaly spectra with the help of synthetic clearsky DLR to differentiate the measurements of the two AERIs during the overlapping period 154 155 using the method detailed in Appendix A. 156



158 Figure 4. Monthly anomaly of AERI-observed DLR spectra and hourly spectra count in each159 month.

160 In most months, the number of hourly mean spectra is larger than 700 (Figure 4c),

161 which means the instruments operated for >95% of the time. The strongest monthly DLR

162 anomalies are seen in the window band  $(800 - 1200 \text{ cm}^{-1})$ . The pattern of the DLR radiance

anomalies in the overlapping observational period is very similar between AERI-01 andAERI-C1.

165 2.2 Sky classification

157

166 Clouds strongly influence the DLR flux and spectra, especially in the atmospheric 167 window  $(800 - 1200 \text{ cm}^{-1})$ . In order to identify the causes of the DLR trends, we separate the 168 clear-sky spectra from the cloudy ones and to examine their trends separately.

169 A sky classification model is developed using a machine learning method, the k-170 nearest neighbor (k-NN) algorithm(Cunningham & Delany, 2020). The 8-min AERI-01 and AERI-C1 data between March 1, 2011 to July 31, 2012 are used to train the k-NN model. We 171 172 use the same inputs and truth data from Raman Lidar as in Turner and Gero (2011). The k-NN classification achieves an accuracy of 94.8%. This algorithm determines the sky to be 173 174 clear or cloudy, while the cloudy sky is then further classified to be thin-cloud when the 70-175 minute averaged 985 cm<sup>-1</sup> brightness temperature is lower than 250K; otherwise, it is 176 classified to be *thick-cloud*. We also tried a classical backpropagation gradient-descent classification algorithm as used by Turner and Gero (2011), which achieves an accuracy of 177 178 90%. The resulting trends are not sensitive to the classification method chosen. The results 179 presented below are based on the k-NN algorithm.

Based on the classification of the *thin-cloud* and *thick-cloud*, the *thick-cloud* emitting temperature range is smaller than that for *thin-cloud* and *clear-sky*, primarily because *thickclouds* are opaque clouds relatively close to the surface while *thin-cloud* may be either partially cloudy scenes or clouds higher in the troposphere. This is why the *thick-cloud* classification has the smallest standard deviation of DLR among all three different sky conditions.

### 186 2.3 Homogenization

187 During the overlapping observational period, discrepancies larger than the documented AERI absolute calibration uncertainty (Knuteson et al., 2004a) were noticed 188 189 between the monthly mean spectra observed by AERI-01 and AERI-C1. Large radiance 190 discrepancies occur especially in the window band and are found to mainly come from clear-191 sky scenes (see Figure B1 and discussions in Appendix B). This suggests that the discrepancies likely result from calibration(Rowe, Neshyba, Cox, et al., 2011; Rowe, 192 193 Neshyba, & Walden, 2011) and other undetected errors. In order to avoid discarding 194 meaningful data in the trend analysis, we simulate the clear-sky DLR spectra using a 195 radiation model from the collocated atmospheric measurements and use the synthetic spectra 196 as a reference to assign proper weights in combining the data of AERI-01 and AERI-C1, 197 based on the findings of previous radiance closure studies (e.g., Turner et al., 2004) that 198 demonstrated high accuracy in such synthetic spectra.

199 The radiation model used here is the Line-by-Line Radiative Transfer Model 200 (LBLRTM v12.9) (Clough et al., 2005). To synthesize the clear sky DLR spectra at SGP, we use the temperature and water vapor profiles from the ARM Balloon-Borne Sounding System 201 202 (https://www.arm.gov/capabilities/instruments/sonde). The water vapor mixing ratio profiles 203 derived from radiosonde are scaled with a height-independent factor to match the precipitable 204 water vapor determined by the microwave radiometer at SGP site. This approach has been 205 used to compensate the dry-bias issue found in the radiosonde water vapor data (Holdridge, 206 2020; Revercomb et al., 2003; Turner et al., 2003; Wang et al., 2002). CO<sub>2</sub> and CH<sub>4</sub> 207 concentration profiles are obtained from the CarbonTracker website 208 (http://carbontracker.noaa.gov, Jacobson et al., 2020; Peters et al., 2007). O<sub>3</sub> concentration profiles are adjusted from NASA's Modern-Era Retrospective analysis for Research and 209 210 Applications, Version 2 (MERRA-2, Gelaro et al., 2017) ozone product to get a better radiative closure with AERI observed DLR (see more details in Appendix B). We use a 200-211 212 level input profile for the LBLRTM simulations. The first and second levels are at 0m and 213 10m above ground level respectively. The depth of each subsequent layer is increased by 2% 214 relative to the previous one.

215 As radiosonde observations of near-surface layers are essential to the DLR spectra,

216 the AERI data are selected to match the radiosonde launch time. We keep the spectra whose 217 observation time is within 10 minutes of the radiosonde launch time. For each month, about

observation time is within 10 minutes of the radiosonde launch time. For each month, about
 70 clear sky downwelling longwave spectra are simulated on average. The absolute radiance

biases ( $R_{bias}$ ) are determined as the monthly mean radiance differences between the synthetic and observed DLR spectra.

During the overlapping observational period, the monthly mean DLR is combined from AERI-01 and AERI-C1 observed DLR according to Equation (1) and Equation (2) by assigning a ratio r, which represents the proximity of the AERIs observed DLR spectra to the synthetic DLR spectra. In Equation (2),  $R_{AERI-01}$  and  $R_{AERI-C1}$  represent the observed AERI-

225 01 and AERI-C1 monthly mean DLR respectively.

226 
$$r = \frac{R_{bias(AERI-01-LBLRTM)}}{R_{bias(AERI-C1-LBLRTM)}}$$
(1)

227 
$$R = R_{AERI-01} \times \frac{1}{1+r} + R_{AERI-C1} \times \frac{r}{1+r}$$
(2)

228 2.4 Trend detection

A weighted linear regression method is applied to detecting the DLR radiance trends.
We develop our weighted linear regression model based on the regression model developed
by Tiao et al. (1990) and Weatherhead et al. (1998).

232 This model determines the radiance trend,  $\hat{\omega}$ , in each AERI channel as:

233 
$$\widehat{\omega} = \frac{\sum_{t=1}^{T} W_t (t - \overline{t}) y_t^{\star}}{\frac{1 - \phi}{12} \sum_{t=1}^{T} W_t (t - \overline{t})^2}$$
(3)

In Equation (3), *T* represents the total number of months.  $\phi$  is the autocorrelation in the noise of the time series considering a first-order autoregressive (AR1) process, and  $y_t^*$ represents the transformed radiance anomalies (see Figure A1) after removing the effect of the AR1 process (see details in Appendix A).  $W_t$  represents the weights which is determined as the intra-month variability of the all-sky observed DLR, shown in Equation (4):

$$W_t = \frac{N_t}{{\sigma_t}^2} \tag{4}$$

240 where  $N_t$  and  $\sigma_t^2$  represent the number and variance of hourly observations in each month.

Large variability of DLR radiance results in smaller weights. We use the same weights for allsky types.

Along with the magnitude of the trend it is also important to determine the associated uncertainty,  $\sigma_{\hat{\omega}}$ , which is shown in Equation (5). Here, we mainly account for two sources of uncertainty. First, there is the uncertainty arising from the internal climate variability. This is measured by the term in Equation (5) associated with  $\sigma_N$  and  $\phi$ . Second, there is the uncertainty arising from instrumentation errors measured by the term in Equation (5) associated with  $\sigma_e$ . We assume that two sources of uncertainty are independent of each other. The derivation of Equation (5) is given in Appendix A.

250 
$$\sigma_{\widehat{\omega}} = \frac{12\sqrt{\sum_{t=1}^{T} W_t^2 (t-\overline{t})^2}}{\sum_{t=1}^{T} W_t (t-\overline{t})^2} \sqrt{\sigma_N^2 \frac{1+\phi}{1-\phi} + \sigma_e^2}$$
(5)

251 The derived  $\sigma_{\hat{\omega}}$  in Equation (5) is referred to as the standard error of the trend 252 magnitude. It is used to test whether the trends deviate significantly from 0 at the 95% 253 significance level. A trend is considered to be significant at the 95% significance level if the 254 trend magnitude is roughly larger than  $2\sigma_{\hat{\omega}}$ . In following figures, the uncertainty envelope 255 corresponds to the 95% confidence interval.

## 256 3 Results

257 3.1 All-sky radiance trends

The homogenized downwelling longwave radiance records have been constructed, based on monthly averaged AERI-01 data from 1996 to 2013 and AERI-C1 data from 2004 to 2018. In total, we have 23 years of DLR data at SGP for analysis.

It can be inferred from the monthly anomalies shown in Figure 4 that the DLR radiance trends depend on the analysis period as the anomalies do not show monotonic changes over this 23-year period. It is noticeable that AERI-01 data (Figure 4a) show decreasing trends in window-band (800-1200 cm<sup>-1</sup>) DLR, which is consistent with the negative trends detected in Gero and Turner (2011). Including AERI-C1 data (Figure 4b)

- affords a longer DLR spectral record; the latest few years are especially characterized by
- warm anomalies.



268

Figure 5. The all-sky radiance trends. Each dot represents the trend at a different AERI channel and the trends in reds pass the 95% significance test while the grey ones do not. The shading in the figure is the 95% confidence interval.

The long-term all-sky radiance trends over the period of 1996-2018 are shown in Figure 5. The all-sky DLR trends have different features in different spectral bands. In the CO<sub>2</sub> absorption band centered around 667 cm<sup>-1</sup>, the trends are generally positive (increasing) and are statistically significant in the wings but not at the center. In the window band (800-1200 cm<sup>-1</sup>), there are no statistically significant trends. In the water vapor absorption band (1300-1800 cm<sup>-1</sup>), similar to the CO<sub>2</sub> absorption band, the radiance trends are generally positive and statically significant.

DLR radiance in different AERI channels are controlled by different meteorological variables. To illustrate this point, Figure 6a shows the correlation coefficients between the deseasonalized and detrended monthly anomalies in the radiance (brightness temperature) 282 spectra from the two AERIs and surface air temperature from ERA5. Note that AERI-01 and 283 AERI-C1 have different observational periods, which can cause the correlation coefficients 284 difference between AERI-01 and AERI-C1 especially in the window band. In the center of 285 the CO<sub>2</sub> absorption band (667 cm<sup>-1</sup>) and channels corresponding to strong H<sub>2</sub>O absorption lines, the correlation coefficient is close to one, indicating that the variance in the radiance in 286 these channels is primarily controlled by the surface air temperature. This is because the 287 288 atmospheric absorption is strongly saturated in these channels and thus they are less sensitive 289 to variations in the concentrations of the gases themselves. In comparison, at the wings of the 290 CO<sub>2</sub> band and the weaker H<sub>2</sub>O absorption lines, the atmospheric absorption is not saturated 291 so that variability in DLR radiance is subject to the variation in the temperature and gas 292 concentration, meaning that the trends both in temperature and gas concentrations drive the 293 radiance to increase, which explains the stronger and statistically more significant trend 294 signals in these channels, as seen in Figure 5.





Figure 6. (a)The correlation coefficient between the AERI observed brightness temperature
spectra and near surface air temperature from ERA5. (b-e) The time series of the
deseasonalized brightness temperature and near surface air temperature in four AERI
channels. In each title, the values in the brackets are the correlation coefficients between near
surface air temperature from ERA5 and observed brightness temperature by AERI-01 and
AERI-C1 respectively.

302 In Figure 6, the time series of the brightness temperature in four selected AERI channels: a CO<sub>2</sub> channel at 655.72 cm<sup>-1</sup>, a window channel at 887.63 cm<sup>-1</sup>, a O<sub>3</sub> channel at 303 1023.60 cm<sup>-1</sup>, and a H<sub>2</sub>O channel at 1447.89 cm<sup>-1</sup> (Figure 6b to 6e) are displayed. There is 304 305 good consistency between the AERI-01 and AERI-C1 observed brightness temperature in all 306 four channels. The all-sky brightness temperature at the CO<sub>2</sub> channel follows closely with the surface air temperature from ERA5 (Figure 6b). The near-surface warming of 0.045 K/year 307 (Figure 1) is equivalent to 0.071 RU/year at this channel, which is close to the observed all-308 sky radiance trend of ~0.072 RU/year (averaged trend between nearby 5 channels). In the 309 H<sub>2</sub>O channel, the brightness temperature measured by the AERIs also follows the near 310

- 311 surface air temperature (Figure 6e) but not as close as the CO<sub>2</sub> channel (Figure 6b). In
- 312 contrast, the brightness temperature anomalies in the window and O<sub>3</sub> channels have larger
- 313 fluctuations than that at the  $CO_2$  and  $H_2O$  channels and are evidently decoupled from the near
- 314 surface air temperature (Figure 6c and 6d).

315 That the radiance trend is reinforced by the warming and opacity effects in the weak

- 316 absorption channels leads to benefits of using these AERI measurements in climate change
- 317 detection. Assuming the trend magnitude and uncertainty determined from the 23-year
- 318 records hold unchanged into future, the years to detect a significant trend,  $n^*$ , at 90%
- 319 significance level is:

320

$$n^{\star} \approx \frac{3.3\sigma_{\widehat{\omega}}}{|\widehat{\omega}|} \times 23 \text{ years} \tag{6}$$

321 where  $\hat{\omega}$  is the 23-year trend determined by Equation (3) and  $\sigma_{\hat{\omega}}$  is the trend 322 uncertainty determined by Equation (5). Based on this equation, approximately 30 years are 323 needed to detect a significant trend in the 2-meter air temperature from the ERA5 data shown 324 in Figure 1. In comparison, Figure 7 shows earlier detectability of the radiance trends in weak 325 absorption channels, such as in the wings of the CO<sub>2</sub> band and in the weak absorption 326 channels in the H<sub>2</sub>O vibration-rotational band. We can conclude that it is advantageous to

327 monitor the DLR radiance in these channels for climate change detection.



328

Figure 7. Trend detectability. (a) Time to detect (T2D) radiance trends at 90% significance level in different AERI channels; in comparison, the T2D for the 2-meter temperature from the ERA5 reanalysis is about 30 years. (b) The T2D (color-coded), in relation to atmospheric absorption strength, measured by the optical depth of a 1-meter-thick atmospheric layer near the surface. The horizontal line marks optical depth of 0.5.

334 Trend detection in the radiance record is determined by comparing the trend signal to 335 the uncertainties arising from different causes. Here, based on Equation (5), we account for 336 uncertainties arising from climate internal variability ( $\sigma_N$ ) and also instrumentation error ( $\sigma_e$ ) 337 (Figure 5). The overall uncertainty is notably large in the window band for the all-sky 338 condition (Figure 5) or for different sky conditions (Figure 9), which impedes the detection of

any significant radiance trends in this especially variable spectral band. The analysis of the respective parameters in Appendix A (see Figure A2) indicates the climate internal variability dominates the instrumentation error when shaping the overall uncertainty envelope in Figure 5. It is found that the influence of the autoregressive process also does not strongly influence the trend uncertainty, as evident by the small value of  $\phi$ , especially in the window band. We conclude that the trend uncertainty mainly arises from the internal climate variability.

345 3.2 Trends in different cloud conditions

The results presented in the previous subsection demonstrate that the radiance trends in the window band are different from the greenhouse gas absorption bands; the window band also is prone to high levels of uncertainty due to the marked variability of the signal that ranges from small values in clear sky conditions to large values when opaque low-altitude clouds are overhead. Given the fact that clouds are a significant factor that influences this band (see Figure 2), we analyze the radiance trends under different cloud conditions in this subsection.

The fraction of time that each sky conditions occur in one month (referred as 'sky fractions') based on the hourly spectra are shown in Figure 8. First, there is a good agreement between AERI-01 and AERI-C1 in the sky fraction monthly time series, with correlation coefficients of 0.94, 0.89, and 0.94 for clear-sky, thin cloudy-sky, and thick cloudy-sky, respectively. The clear-sky fraction between June 1996 and May 2010 from our classification is around 42% which is comparable to what was found by Turner and Gero (2011).

The clear-sky fraction increases at a rate of  $0.17\pm0.09$  % per year, while the thick cloudy-sky fraction decreases at a rate of  $-0.18\pm0.09$  % per year. There is no significant trend for thin cloudy-sky fraction. The reason why the sky fraction trends for different sky conditions are different warrant further investigation in future work.



### 363

Figure 8. The monthly sky fractions of different sky conditions, categorized based on 8 minute mean spectra at SGP site. The overlapping observational period is between the two

366 vertical thick black lines.

Trends in DLR radiance for different sky types based on the k-NN classifier are shown in Figure 9. In the window band, the clear-sky and thin cloudy-sky trends are positive, while the thick cloudy-sky trends are negative; however, none of those trends is statistically significant from zero. In the spectral regions outside the window band, the trends for different sky types are generally positive and have the same features as the all-sky scene.



Figure 9. The trends in DLR radiance for different sky types at SGP site. Each dot represents
the trend at a different AERI channel and the trends in red pass the 95% significance test
while the grey ones do not. The shading in the figure is the 95% confidence interval.

The all-sky DLR trends is caused by changes in both sky fraction and radiance of each sky type. We use equation (7) to separate the contributions from these factors, in which  $R_{all}$  represents the all-sky radiance,  $f_i$  and  $R_i$  represent the sky fraction and mean radiance for different sky types.

372

$$\frac{dR_{all}}{dt} = \sum \frac{df_i}{dt} R_i + \sum \frac{dR_i}{dt} f_i + residual$$
(7)

The results of decomposed trends based on Equation (7) are shown in Figure 10. The 381 382 small residual term (purple line in Figure 10a), which comes from the nonlinear effects, suggests that the overall all-sky radiance trends can be well explained by Equation (7). In the 383 384 window band, the overall radiance trends are a result of the compensation between the sky 385 fraction change (orange line in Figure 10a) and the radiance change (yellow line in Figure 10a). While in the CO<sub>2</sub> absorption band (centered at 677 cm<sup>-1</sup>) and H<sub>2</sub>O absorption band 386  $(1300 - 1800 \text{ cm}^{-1})$ , the overall radiance trends are caused by radiance change which is due 387 almost entirely to the increases in the near-surface temperature because the atmosphere is 388 389 already too opaque to reflect any gas concentration changes.

The overall radiance trends caused by sky fraction changes (orange line in Figure 10a) are a result of the compensation between changes in the clear-sky (blue line in Figure 10b) and the thick cloudy-sky fraction (yellow line in Figure 10b). In the CO<sub>2</sub> absorption band (centered at 677 cm<sup>-1</sup>) and H<sub>2</sub>O absorption band (1300 – 1800 cm<sup>-1</sup>), there is a perfect

- 394 compensation resulting in almost no trends. In the window band, the negative trends are 395 mainly caused by the thick cloudy-sky fraction change.
- In the window band (800 -1200 cm<sup>-1</sup>), the overall radiance trends caused by radiance change (yellow line in Figure 10a) result from the compensation between positive clear-sky and thin cloudy-sky radiance change trends and negative thick-cloudy sky radiance change trends. While in the CO<sub>2</sub> absorption band (centered at 677 cm<sup>-1</sup>) and H<sub>2</sub>O absorption band (1300 – 1800 cm<sup>-1</sup>), the radiance changes for the three sky types all contribute similarly to the overall radiance trends caused by radiance change.



## 402

403 Figure 10. The all-sky DLR trends decomposed to the contributions from the sky fraction and radiance changes of different sky types. (a) The blue line represents the calculated all-404 405 sky DLR trends which is the same as that from Figure 5. Orange and yellow lines represent 406 the contribution from sky fraction change and radiance change determined using equation (7) respectively. The purple line is the residual term from Equation (7); (b) The all-sky DLR 407 408 trends caused by sky fraction change. The blue, orange, and yellow lines represent the 409 contributions from clear-sky, thin cloudy-sky, and thick cloudy-sky fraction changes 410 respectively; (c)The all-sky DLR trends caused by radiance change. The blue, orange, and yellow lines represent the contributions from clear-sky, thin cloudy-sky, and thick cloudy-sky 411 412 radiance changes respectively.

## 413 **4 Discussion and Conclusions**

414 In this study, a long-term record of DLR at SGP site have been constructed for 415 analyzing the DLR trends, based on a weighted linear regression method which considers 416 both natural climate variability and measurement error. Compared to previous studies, our 417 analysis is based on a longer DLR record combined from the two AERIs at the SGP site and 418 makes use of synthetic DLR data in validating and differentiating the AERI measurements 419 over their overlapping observational period. In addition, we quantitatively decompose the 420 overall radiance trends due to the contributions from sky fraction change and radiance 421 change.

422 The trends in DLR in different spectral ranges have different features. The trends are 423 generally positive in the CO<sub>2</sub> and H<sub>2</sub>O absorption bands, while no statistically significant trends are detected in the window band (Figure 5). We find that in the centers of the  $CO_2$  and 424 425 H<sub>2</sub>O absorption bands, the radiance are controlled by the near-surface air temperature (Figure 6) because of the strong saturated atmospheric absorption. The sensitivity of DLR to near-426 427 surface air temperature indicates the potential of DLR to monitor climate change. In the 428 wings of these absorption bands, both the near surface atmospheric warming and the increase 429 of the abundance of these trace gases contribute to the radiance trends (Feldman et al., 2015), which makes climate trend signal more readily detectable, as hypothesized by Huang (2013). 430 431 In the window band, the radiance are decoupled from the near surface air temperature (Figure 432 6) because of the impact of sky-fraction changes of different scenes (clear and cloudy-skies).

We find that the sky fraction change and the radiance change led to compensating effects on the DLR trends. This compensation results in weakly (statistically insignificant) negative radiance trends in the window band (Figure 10). In contrast, the radiance trends are dominated by the radiance change in the CO<sub>2</sub> and H<sub>2</sub>O absorption bands, which are similar in all three sky types.

The influences of both climate natural variability and measurement error are
considered when determining the uncertainty of the trend magnitude (Equation (5), Figure
A2). We find that for all the sky types, the majority of the uncertainty comes from the natural

441 variability. This underlines the necessity of continuous DLR measurements to ascertain the

442 DLR trends, especially in the mid-infrared window (Figure 5).

The two AERIs at the SGP site provide us an excellent opportunity to test the accuracy and consistency of the instruments. The discrepancies between the two AERIs in the overlapping periods may have come from calibration error and other undetected instrumentation errors. In this study, we use synthetic data to differentiate and combine the two AERIs' observations. Further investigation is required to understand the origin of the discrepancies and therefore to assure the measurement accuracy.

This paper has focused on the detection, as opposed to attribution, of the DLR trends. In the clear-sky case, atmospheric temperature and radiative gas concentration changes (primarily in water vapor) are likely the main contributors to the DLR radiance changes. As for the cloudy-sky case, changes in both the atmospheric states and cloud properties may contribute to the DLR radiance changes. Future work is warranted to understand and quantitatively attribute the DLR trends disclosed in this paper to different meteorological

- 455 variables.
- 456

## 457 Acknowledgments

458 This work is supported by grants from the Fonds de Recherché Nature et Technologies of

459 Quebec (PR-283823) and the Canadian Space Agency (19FAMCGB16). LL acknowledges

- 460 the support of a Milton Leong Graduate Fellowship of McGill University. The original AERI
- 461 data can be achieved from the ARM data repository (<u>http://www.arm.gov</u>). Our processed
- 462 monthly mean AERI spectra is available from Mendeley Data
- 463 (https://data.mendeley.com/datasets/hdwfm3zpd8/1). CarbonTracker CT2019B results
- 464 provided by NOAA ESRL, Boulder, Colorado, USA from the website at
- 465 http://carbontracker.noaa.gov. CarbonTracker-CH4 results provided by NOAA ESRL,
- 466 Boulder, Colorado, USA from the website at
- 467 <u>http://www.esrl.noaa.gov/gmd/ccgg/carbontracker-ch4/</u>.
- 468
- 469

#### 470 Appendix A: Trend Detection

We first summarize the linear trend model and trend estimation from Tiao et al.
(1990) and Weatherhead et al. (1998) in A.1 and A.2. We adopt the same notation in their
papers. Then we add the measurement error term to the trend detection in A.3 following Tiao
et al. (1990).

475 A.1 Basic linear trend modeling

476 In order to detect the linear trend, we first construct a simple model that describes the 477 monthly mean radiance  $Y_t$  as:

478  $Y_t = \mu + S_t + \omega X_t + N_t, t = 1, \cdots, T$ (A1)

479 where  $\mu$  is a constant term,  $S_t$  represents the seasonal component,  $\omega$  is the trend magnitude to 480 be determined,  $X_t = \frac{t}{12}$  represents time measured in the units of year,  $N_t$  represents the 481 unexplained portion of the data, i.e. the noise, and *T* represents the length of data set in 482 months.

483 The seasonal component  $S_t$  is determined by taking long-term average of each 484 calendar month. This component is subsequently removed from the monthly mean.

- 485  $y_t = Y_t S_t = \mu + \omega X_t + N_t, t = 1, \cdots, T$  (A2)
- 486 The noise  $N_t$  is assumed to be autoregressive of the order of 1 (AR1):

$$N_t = \phi N_{t-1} + \epsilon_t \tag{A3}$$

488 where  $\epsilon_t$  are the random white noise with zero mean and common variance  $\sigma_{\epsilon}^2$ ,

489  $\epsilon_t \sim W(0, \sigma_{\epsilon}^2)$ . The autocorrelations in the noise come from various natural factors.  $\phi$  is

490 determined as the autocorrelation coefficient of the AR1 process after removing from  $y_t$  a

491 linear trend component obtained by regressing  $y_t$  to time using a simple weighted linear least

492 squares method (i.e., neglecting the AR1). The all-sky  $\phi$  is shown in Figure A2a.

493 The variance of the noise  $N_t$  can also be determined from the detrended  $y_t$  time 494 series:

495  

$$\sigma_N^2 = Cov(N_t, N_t) = Cov(\phi N_{t-1} + \epsilon_t, \phi N_{t-1} + \epsilon_t)$$

$$= \phi^2 Cov(N_{t-1}, N_{t-1}) + Cov(\epsilon_t, \epsilon_t)$$

$$= \phi^2 \sigma_N^2 + \sigma_\epsilon^2$$
(A4)

496 Thus,

500

497 
$$\sigma_N^2 = \frac{\sigma_\epsilon^2}{1 - \phi^2} \tag{A5}$$

498 A.2 Trend estimation with weights

499 Given  $\phi$ , to obtain the trend estimation, we consider a transformed model:

$$y_{t}^{*} = y_{t} - \phi y_{t-1} \\ = \mu(1 - \phi) + \omega(X_{t} - \phi X_{t-1}) + \epsilon_{t} \\ = \mu(1 - \phi) + \omega \left[\frac{t - \phi(t - 1)}{12}\right] + \epsilon_{t}$$
(A6)  
$$= \mu(1 - \phi) + \frac{\omega \phi}{12} + \frac{\omega(1 - \phi)t}{12} + \epsilon_{t} \\ = \mu^{*} + \omega t^{*} + \epsilon_{t}$$

501 where  $\mu^* = \mu(1-\phi) + \frac{\omega\phi}{12}$  and  $t^* = \frac{(1-\phi)t}{12}$ . Thus, in the transformed model, there is no 502 more noise term  $N_t$ .

503 The transformed DLR radiance  $y_t^*$  is shown in Figure A1.





507 According to the weighted least square estimation:

504

508 
$$\widehat{\omega} = \frac{\sum_{t=1}^{T} W_t(t^* - \overline{t^*}) y_t^*}{\sum_{t=1}^{T} W_t(t^* - \overline{t^*})^2} = \frac{\sum_{t=1}^{T} W_t(t - \overline{t}) y_t^*}{\frac{1 - \phi}{12} \sum_{t=1}^{T} W_t(t - \overline{t})^2}$$
(A7)

509 where  $W_t$  represents the weights determined according to Equation (4),  $\overline{y_t}^* = \frac{\sum_{t=1}^T W_t y_t^*}{\sum_{t=1}^T W_t}$ ,  $\overline{t^*} = \frac{\sum_{t=1}^T W_t t^*}{\sum_{t=1}^T W_t}$  and  $\overline{t} = \frac{\sum_{t=1}^T W_t t^*}{\sum_{t=1}^T W_t t^*}$ 

510 
$$\frac{\Sigma_{t=1}W_t t}{\Sigma_{t=1}^T W_t}$$
, and  $\overline{t} = \frac{\Sigma_{t=1}W_t t}{\Sigma_{t=1}^T W_t}$ .

511 The variance of the estimated  $\omega$ :

512 
$$\sigma_{\widehat{\omega}}^{2} = Var(\widehat{\omega}) = Var\left[\frac{\sum_{t=1}^{T} W_{t}(t-\overline{t})y_{t}^{\star}}{\frac{1-\phi}{12}\sum_{t=1}^{T} W_{t}(t-\overline{t})^{2}}\right] = \frac{Var\left[\sum_{t=1}^{T} W_{t}(t-\overline{t})y_{t}^{\star}\right]}{(\frac{1-\phi}{12})^{2}\left[\sum_{t=1}^{T} W_{t}(t-\overline{t})^{2}\right]^{2}}$$

$$Var\left[\sum_{t=1}^{T} W_{t}(t-\overline{t})\epsilon_{t}\right] = \frac{\sum_{t=1}^{T} Var[W_{t}(t-\overline{t})e_{t}]}{\sum_{t=1}^{T} [Var[W_{t}(t-\overline{t})e_{t}]]}$$

513 
$$= \frac{\left[\sum_{t=1}^{T} W_t(t-\bar{t})^2\right]}{\left(\frac{1-\phi}{12}\right)^2 \left[\sum_{t=1}^{T} W_t(t-\bar{t})^2\right]^2} = \frac{\left[\sum_{t=1}^{T} W_t(t-\bar{t})^2\right]^2}{\left(\frac{1-\phi}{12}\right)^2 \left[\sum_{t=1}^{T} W_t(t-\bar{t})^2\right]^2}$$

514 
$$= \frac{Var(\epsilon_t) \sum_{t=1}^{T} W_t^2 (t-\bar{t})^2}{(\frac{1-\phi}{12})^2 \left[\sum_{t=1}^{T} W_t (t-\bar{t})^2\right]^2} = \frac{\sigma_{\epsilon}^2 \sum_{t=1}^{T} W_t^2 (t-\bar{t})^2}{(\frac{1-\phi}{12})^2 \left[\sum_{t=1}^{T} W_t (t-\bar{t})^2\right]^2} \quad (A8)$$

515 
$$\sigma_{\hat{\omega}} = \frac{\sigma_{\epsilon}}{\frac{1-\phi}{12}} \frac{\sqrt{\sum_{t=1}^{T} W_t^2 (t-\bar{t})^2}}{\sum_{t=1}^{T} W_t (t-\bar{t})^2} = \sigma_N g(T,\phi,W)$$
(A9)

516 
$$g(T,\phi,W) = \sqrt{\frac{1+\phi}{1-\phi}} \frac{12\sqrt{\sum_{t=1}^{T} W_t^2 (t-\bar{t})^2}}{\sum_{t=1}^{T} W_t (t-\bar{t})^2}$$
(A10)

517 Thus,

530

518 
$$\sigma_{\widehat{\omega}} = \sigma_N \sqrt{\frac{1+\phi}{1-\phi}} \frac{12\sqrt{\sum_{t=1}^T W_t^2 (t-\bar{t})^2}}{\sum_{t=1}^T W_t (t-\bar{t})^2}$$
(A11)

519 From equation (A11), we conclude that the trend uncertainty is affected by the 520 length of the available data, the natural variability in the data, the autocorrelation of the data 521 and the weights.

522 A.3 Effect of measurement error

523 When we consider the instrumentation errors  $e_t$  in the measurements, Equation A2 524 becomes:

525 
$$y_t = \mu + \omega X_t + N_t + e_t, t = 1, \cdots, T$$
 (A12)

526  $e_t$  is considered to be white noise with zero mean and common variance  $\sigma_e^2$ , 527  $e_t \sim W(0, \sigma_e^2)$ , and independent of  $N_t$  because  $N_t$  originates from unobserved or unsuspected 528 atmospheric factors, while  $e_t$  comes from the instrument itself.

529 In this case, the variance of noise comes from two parts:

$$\sigma^2 = \sigma_N^2 + \sigma_e^2 \tag{A13}$$

531 Similar to the derivation in Equation (A9), the variance of the estimated trend 532 magnitude is:

$$\sigma_{\widehat{\omega}}^{2} = \sigma_{N}^{2} g^{2}(T,\phi,W) + \sigma_{e}^{2} g^{2}(T,0,W)$$

$$= \sigma_{N}^{2} \frac{1+\phi}{1-\phi} \frac{144 \sum_{t=1}^{T} W_{t}^{2}(t-\overline{t})^{2}}{\left[\sum_{t=1}^{T} W_{t}(t-\overline{t})^{2}\right]^{2}} + \sigma_{e}^{2} \frac{144 \sum_{t=1}^{T} W_{t}^{2}(t-\overline{t})^{2}}{\left[\sum_{t=1}^{T} W_{t}(t-\overline{t})^{2}\right]^{2}} \qquad (A14)$$

$$= \left(\sigma_{N}^{2} \frac{1+\phi}{1-\phi} + \sigma_{e}^{2}\right) \frac{144 \sum_{t=1}^{T} W_{t}^{2}(t-\overline{t})^{2}}{\left[\sum_{t=1}^{T} W_{t}(t-\overline{t})^{2}\right]^{2}}$$

534 The uncertainty of the all-sky radiance trend magnitude caused by the natural 535 variability and the measurement error are shown in Figure A2b.



## 536

533

Figure A2. Parameters concerning the radiance trends. (a) The all-sky autocorrelation
coefficient considering AR1 process; (b) All-sky DLR trend uncertainty decomposition based
on Equation (A14). The blue line represents the total all-sky trend magnitude uncertainty.
The orange and yellow lines represent the all-sky trend magnitude uncertainty arising from
climate natural variability and measurement error respectively.

542 A.4 Time to detect the trend

543 The trend detection  $\omega$  is judged to be real or significantly different from zero at the 544 5% level if  $|\hat{\omega}| > 2\sigma_{\hat{\omega}}$ .  $\hat{\omega}$  is approximately normally distributed, so  $z = \frac{\hat{\omega} - \omega}{\sigma_{\hat{\omega}}}$  follows a 545 standard normal distribution.

$$Pr(|\widehat{\omega}| > 2\sigma_{\widehat{\omega}}) = Pr\left(z > 2 - \frac{\omega}{\sigma_{\widehat{\omega}}}\right)$$
 (A15)

547 To detect a real trend of specified magnitude  $|\omega|$ , with probability of 90%:  $2 - \frac{\omega}{\sigma_{\hat{\omega}}} <$ 548  $-1.3 \Rightarrow \omega > 3.3\sigma_{\hat{\omega}}$ .

549 Thus, the number of years  $n^*$  of data required to detect the trend  $\hat{\omega}$  which is 550 determined based on 23-year data: 551  $n^* \approx \frac{3.3\sigma_{\widehat{\omega}}}{|\widehat{\omega}|} \times 23 \text{ years}$  (A16)

552

## 553 Appendix B: Homogenization of the two AERI records

554

B.1 Comparison between the two AERIs

555 During the overlapping observation period, the all sky monthly mean radiance 556 difference between AERI-01 and AERI-C1 is shown in Figure B1. Since these two 557 instruments have different sampling frequency, the AERI-C1 spectra are averaged to match the sampling of AERI-01 spectra before the comparison. From Figure B1a, there are 558 559 noticeable discrepancies between the AERI-01 and AERI-C1 observations. Because of the 560 different sampling frequency, the two AERIs have random errors of different amplitudes (Turner et al., 2006). However, we find that removing the random errors using the principal 561 562 component analysis following Turner et al. (2006) has little impact on the discrepancies (not 563 shown). We find that in more than 20% of the AERI channels in the spectral range from 700 to 1300 cm<sup>-1</sup> and for more than 12% of the overlapping observational months, the radiance 564 difference between two AERIs is larger than the documented absolute calibration uncertainty 565 566 (Knuteson et al., 2004a).

567 For AERI-C1 data stream, multiple instruments were used. All these transitions can be seen in Figure B1a subtly or obviously. First, the transition from AERI-04 to AERI-05 568 happened in September 2009, which is subtly visible and is labelled by the green star in 569 570 Figure B1a. Next, in March 2010, the instrument changed from AERI-05 to AERI-06, which 571 is labelled by the green triangle in Figure B1a. Then, the transition from AERI-06 to AERI-572 106 happened in March 2011, which is very obviously visible and is labelled by the green square in Figure B1a. AERI-106 operated until July 2013 and was being replaced by AERI-573 574 108 until present. It's interesting that the radiance differences between all of these "AERI-575 C1" instruments and the AERI-01 have unique spectral signatures.



576

577 Figure B1. (a) The monthly mean DLR difference between AERI-C1 and AERI-01 (AERI-578 C1 – AERI-01). The green symbols mean the time of AERI-C1 instrument transition; (b)
579 Number of 8-min spectra for each month (the counts are identical after AERI-C1 spectra are resampled to match AERI-01).

581 When separating the measured spectra to different sky types, we find that the 582 prominent difference between the two AERIs in the window band mainly comes from 583 relatively clear sky conditions. Figure B2 shows the monthly mean radiance difference for 584 different sky types in October 2006 as an example. Here the DLR radiance at 985 cm<sup>-1</sup> is 585 used to classify the sky to be relatively clear (<40 RU) or relatively cloudy (>40 RU). We 586 chose 40 RU based on the threshold that Turner and Gero (2011) used to classify cloudy sky 587 to be thin or thick clouds scenes.



588

Figure B2. The monthly mean DLR difference between AERI-C1 and AERI-01 (AERI-C1 –
 AERI-01) for different sky conditions in October 2006

591 We examined various instrumental parameters recorded with AERI measurements, 592 including calibration blackbody temperatures, instrument responsivity, and so on, but found 593 that no instrumental parameter can indicate the radiance difference between the two AERIs.

594 B.2 Clear-sky LBLRTM simulations

595 Since the differences between two AERIs mainly come from relatively clear sky 596 scenes, we use clear sky synthetic spectra simulated from LBLRTM as a metric to distinguish 597 their relative accuracies. Here we use the classical backpropagation gradient-descent 598 classification algorithm mentioned in Subsection 2.2 to select clear-sky spectra. To make sure 599 the sky chosen is clear, we set the algorithm threshold to be 0.8, which means the possibility 600 of the sky to be clear is at least 0.8.

After matching all datasets including radiosonde dataset and gas concentrations datasets at SGP mentioned in the Method section to select atmospheric profiles, clear sky synthetic spectra are obtained during the overlapping observational period. For each month, about 70 downwelling longwave spectra are simulated on average. The LBLRTM simulation is validated based on the test in Feldman et al. (2015). We chose the same time slices selected in Feldman et al. (2015) to simulate the DLR spectrum and we can achieve similar radiative closures between observation and simulation.



608

609 Figure B3. The clear sky monthly mean difference between AERI observations and610 LBLRTM simulations in October 2006.

611 We originally used the ozone concentration profile from the Modern-Era 612 Retrospective analysis for Research and Applications Version 2 (MERRA-2, Gelaro et al., 613 2017) in simulating the synthetic spectra. A relatively poorer radiance closure between AERI 614 observations and LBLRTM simulations was found in the ozone absorption band near 1040 615 cm<sup>-1</sup>(not shown). By comparing to in situ measurements at SGP (available only at limited times), we find that this is due to poor representation of the local ozone concentration in the 616 617 MERRA-2 dataset. To address this issue, we vertically scale the ozone profile uniformly to 618 achieve an improved radiance closure in the ozone band as exemplified by Figure B3 (AERI-619 C1 line), although this has little impact on the all-sky radiance trend detected in Figure 5.

As exemplified in Figure B3, we find that AERI-C1 generally in better agreement
with LBLRTM simulations than AERI-01 especially in the window band. The radiance
difference in each channel is used to weight the spectra of AERI-01 and AERI-C1, according
to Equation (2), to form an integrated record of monthly mean DLR radiance spectra.

624

#### 625 References

- 626 Clough, S. A., Shephard, M. W., Mlawer, E. J., Delamere, J. S., Iacono, M. J., Cady-Pereira,
- 627 K., Boukabara, S., & Brown, P. D. (2005). Atmospheric radiative transfer modeling: a
- 628 summary of the AER codes. *Journal of Quantitative Spectroscopy and Radiative*
- 629 *Transfer*, 91(2), 233-244. <u>https://doi.org/10.1016/j.jqsrt.2004.05.058</u>
- 630

```
631 Cunningham, P., & Delany, S. J. (2020). k-Nearest neighbour classifiers: (with Python
632 examples). arXiv preprint arXiv:2004.04523.
```

- 633
- 634 Feldman, D. R., Collins, W. D., Gero, P. J., Torn, M. S., Mlawer, E. J., & Shippert, T. R.

635	(2015, Mar 19). Observational determination of surface radiative forcing by CO2
636	from 2000 to 2010. Nature, 519(7543), 339-343. https://doi.org/10.1038/nature14240
637	
638	Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A.,
639	Darmenov, A., Bosilovich, M. G., & Reichle, R. (2017). The modern-era retrospective
640	analysis for research and applications, version 2 (MERRA-2). Journal of Climate,
641	30(14), 5419-5454.
642	
643	Gero, P. J., & Turner, D. D. (2011). Long-Term Trends in Downwelling Spectral Infrared
644	Radiance over the U.S. Southern Great Plains. Journal of Climate, 24(18), 4831-4843.
645	https://doi.org/10.1175/2011jcli4210.1
646	
647	Harries, J. E., Brindley, H. E., Sagoo, P. J., & Bantges, R. J. (2001). Increases in greenhouse
648	forcing inferred from the outgoing longwave radiation spectra of the Earth in 1970
649	and 1997. Nature, 410(6826), 355-357.
650	
651	Hersbach, H., Bell, B., Berrisford, P., Hirahara, S., Horányi, A., Muñoz-Sabater, J., Nicolas,
652	J., Peubey, C., Radu, R., & Schepers, D. (2020). The ERA5 global reanalysis.
653	Quarterly Journal of the Royal Meteorological Society, 146(730), 1999-2049.
654	
655	Holdridge D. 2020. Balloon-Borne Sounding System (SONDE) Instrument Handbook. Ed.
656	by Robert Stafford, U.S. Department of Energy. DOE/SC-ARM/TR-029.
657	
658	Huang, Y. (2013). A simulated climatology of spectrally decomposed atmospheric infrared
659	radiation. Journal of Climate, 26(5), 1702-1715.
660	
661	Huang, Y., Chou, G., Xie, Y., & Soulard, N. (2019). Radiative control of the interannual
662	variability of arctic sea ice. Geophysical Research Letters, 46(16), 9899-9908.
663	
664	Huang, Y., Leroy, S. S., & Anderson, J. G. (2010). Determining longwave forcing and
665	feedback using infrared spectra and GNSS radio occultation. Journal of Climate,
666	23(22), 6027-6035.
667	

668	Huang, Y., & Ramaswamy, V. (2009). Evolution and Trend of the Outgoing Longwave
669	Radiation Spectrum. Journal of Climate, 22(17), 4637-4651.
670	https://doi.org/10.1175/2009jcli2874.1
671	
672	Huang, Y., Ramaswamy, V., Huang, X., Fu, Q., & Bardeen, C. (2007). A strict test in climate
673	modeling with spectrally resolved radiances: GCM simulation versus AIRS
674	observations. Geophysical Research Letters, 34(24).
675	https://doi.org/10.1029/2007g1031409
676	
677	Huang, Y., Ramaswamy, V., & Soden, B. (2007). An investigation of the sensitivity of the
678	clear-sky outgoing longwave radiation to atmospheric temperature and water vapor.
679	Journal of Geophysical Research, 112(D5). https://doi.org/10.1029/2005jd006906
680	
681	Jacobson, A. R., Schuldt, K. N., Miller, J. B., Oda, T., Tans, P., Arlyn, A., Mund, J., Ott, L.,
682	Collatz, G. J., Aalto, T., Afshar, S., Aikin, K., Aoki, S., Apadula, F., Baier, B.,
683	Bergamaschi, P., Beyersdorf, A., Biraud, S. C., Bollenbacher, A., Bowling, D.,
684	Brailsford, G., Abshire, J. B., Chen, G., Huilin, C., Lukasz, C., Sites, C., Colomb, A.,
685	Conil, S., Cox, A., Cristofanelli, P., Cuevas, E., Curcoll, R., Sloop, C. D., Davis, K.,
686	Wekker, S. D., Delmotte, M., DiGangi, J. P., Dlugokencky, E., Ehleringer, J., Elkins,
687	J. W., Emmenegger, L., Fischer, M. L., Forster, G., Frumau, A., Galkowski, M., Gatti,
688	L. V., Gloor, E., Griffis, T., Hammer, S., Haszpra, L., Hatakka, J., Heliasz, M.,
689	Hensen, A., Hermanssen, O., Hintsa, E., Holst, J., Jaffe, D., Karion, A., Kawa, S. R.,
690	Keeling, R., Keronen, P., Kolari, P., Kominkova, K., Kort, E., Krummel, P., Kubistin,
691	D., Labuschagne, C., Langenfelds, R., Laurent, O., Laurila, T., Lauvaux, T., Law, B.,
692	Lee, J., Lehner, I., Leuenberger, M., Levin, I., Levula, J., Lin, J., Lindauer, M., Loh,
693	Z., Lopez, M., Luijkx, I. T., Myhre, C. L., Machida, T., Mammarella, I., Manca, G.,
694	Manning, A., Manning, A., Marek, M. V., Marklund, P., Martin, M. Y., Matsueda, H.,
695	McKain, K., Meijer, H., Meinhardt, F., Miles, N., Miller, C. E., Mölder, M., Montzka,
696	S., Moore, F., Josep-Anton, M., Morimoto, S., Munger, B., Jaroslaw, N., Newman, S.,
697	Nichol, S., Niwa, Y., O'Doherty, S., Mikaell, OL., Paplawsky, B., Peischl, J., Peltola,
698	O., Jean-Marc, P., Piper, S., Plass-Dölmer, C., Ramonet, M., Reyes-Sanchez, E.,
699	Richardson, S., Riris, H., Ryerson, T., Saito, K., Sargent, M., Sasakawa, M., Sawa, Y.,
700	Say, D., Scheeren, B., Schmidt, M., Schmidt, A., Schumacher, M., Shepson, P.,

701	Shook, M., Stanley, K., Steinbacher, M., Stephens, B., Sweeney, C., Thoning, K.,
702	Torn, M., Turnbull, J., Tørseth, K., Bulk, P. V. D., Dinther, D. V., Vermeulen, A.,
703	Viner, B., Vitkova, G., Walker, S., Weyrauch, D., Wofsy, S., Worthy, D., Dickon, Y., &
704	Miroslaw, Z. (2020). CarbonTracker CT2019B.
705	https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/CT2019B/
706	
707	Kapsch, ML., Graversen, R. G., Tjernström, M., & Bintanja, R. (2016). The effect of
708	downwelling longwave and shortwave radiation on Arctic summer sea ice. Journal of
709	<i>Climate, 29</i> (3), 1143-1159.
710	
711	Knuteson, R., Revercomb, H., Best, F., Ciganovich, N., Dedecker, R., Dirkx, T., Ellington, S.,
712	Feltz, W., Garcia, R., & Howell, H. (2004a). Atmospheric emitted radiance
713	interferometer. Part I: Instrument design. Journal of Atmospheric and Oceanic
714	<i>Technology</i> , 21(12), 1763-1776.
715	
716	Knuteson, R., Revercomb, H., Best, F., Ciganovich, N., Dedecker, R., Dirkx, T., Ellington, S.,
717	Feltz, W., Garcia, R., & Howell, H. (2004b). Atmospheric emitted radiance
718	interferometer. Part II: Instrument performance. Journal of Atmospheric and Oceanic
719	<i>Technology</i> , 21(12), 1777-1789.
720	
721	Liebmann, B., & Smith, C. A. (1996). Description of a complete (interpolated) outgoing
722	longwave radiation dataset. Bulletin of the American Meteorological Society, 77(6),
723	1275-1277.
724	
725	Lubin, D. (1994). The role of the tropical super greenhouse effect in heating the ocean
726	surface. Science, 265(5169), 224-227.
727	
728	Peters, W., Jacobson, A. R., Sweeney, C., Andrews, A. E., Conway, T. J., Masarie, K., Miller,
729	J. B., Bruhwiler, L. M., Pétron, G., & Hirsch, A. I. (2007). An atmospheric
730	perspective on North American carbon dioxide exchange: CarbonTracker.
731	Proceedings of the National Academy of Sciences, 104(48), 18925-18930.
732	
733	Revercomb, H. E., Buijs, H., Howell, H. B., LaPorte, D. D., Smith, W. L., & Sromovsky, L.

734	(1988). Radiometric calibration of IR Fourier transform spectrometers: solution to a
735	problem with the High-Resolution Interferometer Sounder. Applied Optics, 27(15),
736	3210-3218.
737	
738	Revercomb, H. E., Turner, D. D., Tobin, D., Knuteson, R. O., Feltz, W., Barnard, J.,
739	Bösenberg, J., Clough, S., Cook, D., & Ferrare, R. (2003). The ARM program's water
740	vapor intensive observation periods: Overview, initial accomplishments, and future
741	challenges. Bulletin of the American Meteorological Society, 84(2), 217-236.
742	
743	Rowe, P. M., Neshyba, S. P., Cox, C. J., & Walden, V. P. (2011). A responsivity-based
744	criterion for accurate calibration of FTIR emission spectra: identification of in-band
745	low-responsivity wavenumbers. Optics express, 19(7), 5930-5941.
746	
747	Rowe, P. M., Neshyba, S. P., & Walden, V. P. (2011). Responsivity-based criterion for
748	accurate calibration of FTIR emission spectra: theoretical development and bandwidth
749	estimation. Optics express, 19(6), 5451-5463.
750	
751	Shupe, M. D., & Intrieri, J. M. (2004). Cloud Radiative Forcing of the Arctic Surface: The
752	Influence of Cloud Properties, Surface Albedo, and Solar Zenith Angle. Journal of
753	Climate, 17(3), 616-628. https://doi.org/10.1175/1520-
754	<u>0442(2004)017</u> <0616:Crfota>2.0.Co;2
755	
756	Sokolowsky, G. A., Clothiaux, E. E., Baggett, C. F., Lee, S., Feldstein, S. B., Eloranta, E. W.,
757	Cadeddu, M. P., Bharadwaj, N., & Johnson, K. L. (2020). Contributions to the Surface
758	Downwelling Longwave Irradiance during Arctic Winter at Utqiaġvik (Barrow),
759	Alaska. Journal of Climate, 33(11), 4555-4577. https://doi.org/10.1175/jcli-d-18-
760	<u>0876.1</u>
761	
762	Stephens, G. L., Li, J., Wild, M., Clayson, C. A., Loeb, N., Kato, S., L'Ecuyer, T.,
763	Stackhouse, P. W., Lebsock, M., & Andrews, T. (2012). An update on Earth's energy
764	balance in light of the latest global observations. Nature Geoscience, 5(10), 691-696.
765	https://doi.org/10.1038/ngeo1580
766	

767	Tiao, G. C., Reinsel, G. C., Xu, D., Pedrick, J., Zhu, X., Miller, A., DeLuisi, J., Mateer, C., &
768	Wuebbles, D. (1990). Effects of autocorrelation and temporal sampling schemes on
769	estimates of trend and spatial correlation. Journal of Geophysical Research:
770	Atmospheres, 95(D12), 20507-20517.
771	
772	Trenberth, K. E., Fasullo, J. T., & Kiehl, J. (2009). Earth's Global Energy Budget. Bulletin of
773	the American Meteorological Society, 90(3), 311-324.
774	https://doi.org/10.1175/2008bams2634.1
775	
776	Turner, D., Knuteson, R., Revercomb, H., Lo, C., & Dedecker, R. (2006). Noise reduction of
777	Atmospheric Emitted Radiance Interferometer (AERI) observations using principal
778	component analysis. Journal of Atmospheric and Oceanic Technology, 23(9), 1223-
779	1238.
780	
781	Turner, D. D., & Gero, P. J. (2011). Downwelling 10µm radiance temperature climatology for
782	the Atmospheric Radiation Measurement Southern Great Plains site. Journal of
783	Geophysical Research, 116(D8). https://doi.org/10.1029/2010jd015135
784	
785	Turner, D. D., Lesht, B. M., Clough, S. A., Liljegren, J. C., Revercomb, H. E., & Tobin, D.
786	(2003). Dry bias and variability in Vaisala RS80-H radiosondes: The ARM
787	experience. Journal of Atmospheric and Oceanic Technology, 20(1), 117-132.
788	
789	Turner, D. D., Tobin, D., Clough, S. A., Brown, P. D., Ellingson, R. G., Mlawer, E. J.,
790	Knuteson, R. O., Revercomb, H. E., Shippert, T. R., & Smith, W. L. (2004). The QME
791	AERI LBLRTM: A closure experiment for downwelling high spectral resolution
792	infrared radiance. Journal of the atmospheric sciences, 61(22), 2657-2675.
793	
794	Wang, J., Cole, H. L., Carlson, D. J., Miller, E. R., Beierle, K., Paukkunen, A., & Laine, T. K.
795	(2002). Corrections of humidity measurement errors from the Vaisala RS80
796	radiosonde—Application to TOGA COARE data. Journal of Atmospheric and
797	<i>Oceanic Technology, 19</i> (7), 981-1002.
798	
799	Weatherhead, E. C., Reinsel, G. C., Tiao, G. C., Meng, XL., Choi, D., Cheang, WK.,

800	Keller, T., DeLuisi, J., Wuebbles, D. J., Kerr, J. B., Miller, A. J., Oltmans, S. J., &
801	Frederick, J. E. (1998). Factors affecting the detection of trends: Statistical
802	considerations and applications to environmental data. Journal of Geophysical
803	Research: Atmospheres, 103(D14), 17149-17161. https://doi.org/10.1029/98jd00995
804	
805	Wielicki, B. A., Wong, T., Allan, R. P., Slingo, A., Kiehl, J. T., Soden, B. J., Gordon, C.,
806	Miller, A. J., Yang, SK., & Randall, D. A. (2002). Evidence for large decadal
807	variability in the tropical mean radiative energy budget. Science, 295(5556), 841-844.
808	
809	Wild, M., Folini, D., Schär, C., Loeb, N., Dutton, E. G., & König-Langlo, G. (2012). The
810	global energy balance from a surface perspective. Climate Dynamics, 40(11-12),
811	3107-3134. https://doi.org/10.1007/s00382-012-1569-8
812	
813	