1	Supporting Information for					
2	Titan's Global Radiant Energy Budget During the Cassini Epoch (2004-2017)					
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19 S1. Theoretical Methodology

20 To determine the radiant energy budget for a planet or a moon, we have to measure the 21 absorbed solar energy and the emitted thermal energy (Conrath et al., 1989). We have estimated 22 Titan's emitted thermal energy with the observations recorded by the Cassini Composite Infrared 23 Spectrometer (Creecy et al., 2019). In this study, we focus on measuring Titan's absorbed solar 24 energy with the observations obtained by the Cassini Imaging Science Sub-system and Visual and 25 Infrared Mapping Spectrometer. The methodology of computing the absorbed solar energy is provided in some previous studies (Conrath et al., 1989), and discussed in detail in one of our 26 27 previous studies (Li et al., 2018). Here, we briefly introduce the main idea of the methodology. 28 Generally, we measure the reflected solar energy and then subtract the reflected solar energy from 29 the total solar energy to get the absorbed solar energy. The key parameter of computing the

30 reflected solar energy is the Bond albedo A. The Bond albedo at each wavelength is called the 31 monochromatic Bond albedo (A_m) . It is defined as $A_m = A_0 q$, where A_0 and q are the 32 monochromatic geometric albedo and the monochromatic phase integral, respectively. The 33 monochromatic geometric albedo A_0 is defined as the full-disk albedo at 0° phase angle for a 34 given wavelength. The phase integral q is the integral of the reflected solar irradiance over phase 35 angle (Li et al., 2018). Once the monochromatic Bond albedo A_m at each wavelength is measured, 36 we can compute the Bond albedo A for a planet or a moon by weighting the monochromatic Bond

albedo with the solar spectral irradiance (see sections as below).

38

39 S2. Summary of Observational Data Sets

Based on the methodology we discussed in "Methods", we know the monochromatic Bond 40 albedo (A_m) is computed by the product of the monochromatic geometric albedo (A_0) and the 41 monochromatic phase integral (q). Both qualities are related to the ratios between the reflected 42 43 solar irradiance and the incoming solar irradiance at each wavelength. Therefore, the incoming 44 solar irradiance at each wavelength, which is also named as the solar spectral irradiance (SSI), 45 provides the reference for computing the monochromatic Bond albedo. In this study, we also 46 investigate the temporal variations of Titan's Bond albedo and the radiant energy budget during 47 the Cassini epoch (2004-2017). So the first data set is the time-varying SSI for the Cassini epoch. 48 To compute the total reflected solar irradiance, we need integrate the reflected solar irradiance 49 over the phase angle from 0° to 180° (i.e., phase function or phase integral). The observations of 50 phase function and the reflected spectra are mainly provided by the Cassini Imaging Science 51 Subsystem (ISS) (14) and the Visual and Infrared Mapping Spectrometer (VIMS) (15). The Cassini 52 ISS and VIMS do not cover the complete wavelength range from 0 nm to 6000 nm, so we need 53 observations of the reflected spectra at these wavelengths outside the Cassini wavelengths. These 54 observations not only help fill the observational gaps in wavelength but also help validate the 55 Cassini ISS and VIMS observations. The observations of the reflected spectra, which come from 56 the Cassini ISS and VIMS observations and the other observations, are defined as the spectral 57 observations. The albedo spectra in the wavelength range 150-190 nm come from the Cassini 58 Ultraviolet Imaging Spectrograph (UVIS) observations (Esposito et al., 2004; Ajello et al., 2008). 59 The Faint Object Spectrograph (FOS) of the Hubble Space Telescope (HST) and the European 60 Southern Observatory (ESO) also provide albedo spectra of Titan (Karkoschka, 1994, 1998; 61 McGrath et al., 1998).

For the phase function of Titan's albedo, the Imaging Photopolarimeter (IPP) on the spacecraft Pioneer 11 conducted the first observations (Tomasko and Smith, 1982). The coverage of phase angle is better for the Cassini ISS/VIMS observations than for Pioneer 11 observations, but the latter can help validate the Cassini observations. Table S1 in the Supporting Information summarizes the data sets for the SSI, the phase integral, and the spectral observations. These data sets are discussed in detail in following sections.

68

69 S3. Solar Spectral Irradiance Data

The Solar Spectral Irradiance (SSI) provides a base for computing Titan's albedo and hence the reflected/absorbed solar power at each wavelength. The SSI from 0 to 6000 nm contributes to ~ 99.7% of the total solar power, so we construct the SSI in the wavelength range 0-6000 nm. The SEE, SORCE, and ASTM are three data sets used for the solar spectral irradiance (SSI). They are the Solar EUV Experiment (SEE) (http://lasp.colorado.edu/lisird/data/timed_see_ssi_l3a/), the 75 Solar Radiation Climate Experiment and (SORCE) 76 (http://lasp.colorado.edu/lisird/data/sorce_ssi_13/), and the American Society for Testing and 77 Materials (ASTM) (https://www.nrel.gov/grid/solar-resource/spectra-am1.5.html), respectively. 78 We do not find trustable data sets for the SSI in the wavelength range 4000-6000 nm, so we 79 compute it by assuming the blackbody spectra for the Sun with a temperature 5778 K. 80 It should be mentioned that the longest wavelength of VIMS is 5131 nm, so our analyses of

81 Titan's albedo is mainly in the wavelength range 0-5131 nm, which contains more than 99.5% of 82 the total solar power. The total solar power varies $\sim 0.1\%$ on the time scale of decades (Willson 83 and Mordvinov, 2003; Lean and Rind, 2009) (also see Fig. S1), but the SSI at some wavelengths 84 can vary with much larger magnitudes. Therefore, it is better to resolve the temporal variations of 85 the SSI. We construct the SSI in the wavelength range 0-6000 nm for the Cassini epoch (2004-86 2017) from different data sets (see Table S1) listed in Fig. S1. Figure S1 shows both the SSI (panel 87 A) and the total solar power at Earth (i.e., solar constant) (panel B) by integrating the SSI over 88 wavelength.

Based on the constructed SSI at Earth (Fig. S1), we can compute the SSI at Titan (panel A of Fig. S2) with the distance between the Sun and Titan (panel B of Fig. S2). The distance between the Sun and Titan during the period of 2004-2017 (panel B) comes from the NASA/JPL solar system dynamics – the Horizons Web-interface (<u>https://ssd.jpl.nasa.gov/horizons.cgi</u>). In addition, the total solar power at Titan (i.e., solar flux), which is computed by integrating the SSI (panel A) over wavelength, is shown in panel C. The SSI at Titan is used for the computation of Titan's albedo.

96

97 S4. Supplementary Observations and Data

98 As shown in Table S1, there are observations different from the Cassini ISS and VIMS 99 observations (i.e., Supplementary Observations), which can help to validate the Cassini ISS and 100 VIMS measurements and fill the observational gaps in wavelengths. There are many observations 101 of Titan's albedo spectra in different wavelengths and times. Here, we select the best available 102 observations. The first observations come from the ESO (Karkoschka, 1994, 1998), one of the best 103 observations of Titan's albedo spectra cover the wavelengths from ~ 305 nm to 1050 nm with a 104 very high spectral resolution (~ 0.4 nm). Two data sets from the ESO are used: one is the albedo spectra at a phase angle 2.7° in 1993 and the other is the albedo spectra at a phase 5.7° in 1995. 105 106 The two spectra are shown in Fig. S3.

107 The shortest effective wavelength of the ESO observations is about 305 nm, so we also 108 searched for albedo spectra with wavelengths less than 305 nm. Two data sets are used: one comes 109 from the Cassini UVIS (Esposito et al., 2004) and the other comes from HST/FOS (McGrath et 110 al., 1998). Based on the observations from the Cassini UVIS and the HST/FOS, the studies by 111 Ajello et al. (2008) and McGrath et al. (1998) provide the albedo spectra in the wavelength ranges 112 of 150-190 nm and 190-305 nm, respectively. We are unable to locate good data sets for the albedo 113 spectra in the wavelength range 0-150 nm, so we assume that the albedo spectra in this wavelength 114 range are the same as that at 150 nm.

115 There are very limited observations for the phase function of Titan's albedo mainly because 116 the Earth-based observations cover a very narrow range of phase angle. To the best of our 117 knowledge, the only good measurements of the phase function of Titan's albedo aside from Cassini 118 were conducted by the IPP on Pioneer 11 (Tomasko and Smith, 1982). Compared to the Cassini 119 observations, the coverage of phase angle by the Pioneer 11 observations are quite limited, but 120 they are the best observations for the phase function of Titan's albedo before the Cassini epoch. 121 The IPP on the spacecraft Pioneer 11 conducted measurements of Titan's albedo in two 122 wavelengths (452 nm (blue) and 648 (red)) with phase angles varying from 21.9 ° to 95.9 °. The 123 Pioneer 11/IPP measurements, which have been described in a previous study (Tomasko and 124 Smith, 1982), are shown in Fig. S5.

125

126 S5. Cassini ISS/VIMS Data and Data Processing

127 The Cassini spacecraft conducted on-orbit observations of the Saturn system from October 128 2004 to September 2017. During this period, there are many observations of Titan with multiple flybys. In this study, we mainly analyze the observations of Titan's reflected solar irradiance from 129 130 two Cassini instruments: ISS and VIMS. The observations recorded by the Cassini ISS and VIMS 131 have many improvements than the previous observations, which have been discussed in our 132 previous studies (Li et al., 2010, 2011, 2018; Creecy et al., 2019). Therefore, our analyses of 133 Titan's Bond albedo, which are based on the Cassini observations, represent the best existing 134 measurements of Titan's Bond albedo. The measurements of Titan's Bond albedo are combined 135 with our previous measurements of Titan's emitted power (Creecy et al., 2019) with the 136 observations from the Cassini Composite Infrared Spectrometer (CIRS) (Flasar et al., 2004) to 137 provide the first precise measurements of Titan's global radiant energy budget. More importantly, 138 the long-term Cassini observations from 2004 to 2017 make it possible to investigate the temporal 139 variations of Titan's radiant energy budget and examine the possible energy imbalance for the first 140 time.

141 Here, we briefly introduce the two Cassini instruments (i.e., ISS and VIMS), which are used 142 to measure Titan's Bond albedo for the Cassini epoch. As the imaging system of the Cassini 143 spacecraft, the ISS is a Charged-Coupled Device with two cameras (one is the narrow-angle 144 camera and the other is the wide-angle camera) (Porco et al., 2004). The characteristics of the ISS 145 instrument and the related data processing (e.g., calibrating and navigating) are described in 146 previous studies (Porco et al., 2004; Li et al., 2018). The ISS has multiple filters (i.e., wavelengths) 147 ranging from the ultraviolet to the near infrared. Here we mainly use the 12 filters with a 148 wavelength range from 264 nm to 939 nm, which were summarized in our previous study of 149 Jupiter's radiant energy budget and internal heat (Li et al., 2018), to compute the full-disk albedo 150 of Titan. The basic characteristics of 12 filters (three ultraviolet filters "UV1", "UV2", and "UV3"; 151 three methane-absorption filters "MT1", "MT2", and "MT3"; the three continuum filters "CB1", "CB2", and "CB3"; and the three color filters "BL1", "GRN", and "RED") are introduced in the 152 153 ISS introductory paper (Porco et al., 2004) and summarized in our previous study of Jupiter (Li et 154 al., 2018).

To compute Titan's full-disk albedo, we searched for the global images of Titan from the complete ISS data set (<u>https://pds-imaging.jpl.nasa.gov/volumes/iss.html</u>). We selected the ISS global images with spatial resolutions from ~ 6 km/pixel to ~ 200 km/pixel, which resolve Titan well, to conduct the measurements. The corresponding phase angles vary from ~ 0° to ~167°, which are the best among all available observations. There are observational gaps in phase angle even with the Cassini ISS observations, so the least-squares method is used to fill the observational gaps in phase angle (see section "Filling Observational Gaps in Phase Angle").

After collecting the raw ISS global images, we calibrated the recorded digital number of brightness to radiance using the latest version of the Cassini ISS CALibration software (<u>https://pds-</u> <u>imaging.jpl.nasa.gov/data/cassini/cassini_orbiter/coiss_0011_v4.3/</u>), which was developed by the ISS team (West et al., 2010; Knowles et al., 2020). An example of such calibration is presented in Fig. S6. The radiance at each pixel of the global images is multiplied by the area of the pixel, and summed over all pixels in Titan's disk with an effective radius (see section "Effective Radii of Titan's Atmosphere") to get the observed full-disk reflected solar irradiance. At the same time, the reference SSI (Fig S2) is multiplied by the total area of Titan's disk with the effective radius to get the reference full-disk solar irradiance. Then the ratio between the observed full-disk solar irradiance and the reference full-disk solar irradiance is taken as the full-disk albedo (i.e., I/F).

172 The Cassini ISS observations have the best coverage of phase angle, but they focus on limited 173 wavelengths only. On the other hand, the Cassini VIMS instrument is a spectral camera that takes 174 images in 352 wavelengths between ~350 nm and ~5130 nm with varying spectral resolutions 175 from ~ 4 nm to ~ 25 nm (Brown et al., 2004). The VIMS observations can help extend the spectral 176 coverage of the ISS observations. The raw global VIMS images, which are available on the PDS system (https://pds-imaging.jpl.nasa.gov/volumes/vims.html), are well calibrated by the VIMS 177 178 Operations Team (Brown et al., 2004; McCord et al., 2004; Filacchione et al., 2007; Buratti et al., 179 2010; Pitman et al., 2010).

180 For the wavelengths covered by the Cassini ISS observations (~ 264-939 nm), the reflected 181 solar irradiance is dominant over the emitted thermal radiance in the recorded radiance, so the 182 emitted thermal radiance from Titan can be neglected in the ISS recorded radiance. When the 183 wavelength increases, the emitted thermal radiance becomes stronger, even though it is still much 184 smaller than the reflected solar irradiance (even at the longest VIMS wavelength 5131 nm). 185 However, it is more accurate to consider the emitted thermal radiance when computing Titan's 186 albedo in the relatively long infrared wavelengths covered by the VIMS observations. Based on 187 the incident solar angle, we divide each VIMS global image into the day-side and night-side parts 188 (see examples in Fig. S7). The radiance recorded by the night-side part is mainly from the thermal 189 emission, and the radiance recorded by the day-side part includes both the reflected solar irradiance 190 and the thermal emission. In order to precisely measure the reflected solar irradiance and hence 191 full-disk albedo, we subtract the night-side thermal emission from the day-side radiance to get the 192 reflected solar irradiance.

193 The spatial resolutions are generally much lower for the VIMS images than for the ISS 194 images. Here, we select the VIMS global images with spatial resolutions better than 210 km/pixel. 195 The coverage of phase angle is also much more sparse for the VIMS observations than for the ISS 196 observations. We searched the complete data set of the VIMS observations and found high-quality global observations at ~11 phase angles only (see section "Filling Observational Gaps in Phase 197 198 Angle" and Fig. S23). The selected VIMS observations are used to address the phase functions of 199 Titan's albedo in the wavelength range 350-5131 nm. But these VIMS observations are distributed 200 in different years of the Cassini era, so they cannot resolve temporal variations of the phase 201 function of Titan's albedo. The temporal variations of the phase function observed at the ISS 202 wavelengths were extrapolated to these VIMS wavelengths outside of the ISS wavelengths (936-203 5131 nm) (see section "Filling Observational Gaps in Wavelength and Time").

204 The Cassini spacecraft has one more instrument observing the reflected solar irradiance of 205 Titan - the UVIS (39). The UVIS was used to observe Titan at the wavelengths (56 - 190 nm), 206 which are shorter than the wavelengths covered by the ISS and VIMS. The UVIS team has already 207 generated Titan's full-disk geometric albedo in the wavelength range 150-190 nm (Ajello et al., 208 2008), which is used in this study (see Fig. S4 in the Supplementary Information 4). There are 209 some difficulties in retrieving Titan's albedo in the wavelength range 56-150 nm with the UVIS 210 observations (Ajello et al., 2008). In addition, the SSI in the wavelength range 56-150 nm occupies 211 only ~ 0.006% of the total solar irradiance. So the UVIS-retrieved albedo in the wavelength range 212 56-150 nm are not analyzed in this study. Instead, we simply assume that Titan's albedo in the

213 wavelength range 0-150 nm is the same as that at 150 nm (see section "Filling Observational Gaps

- 214 in Wavelength and Time").
- 215

216 S6. Effective Radii of Titan's Atmosphere

217 Titan's thick atmosphere extends to a few hundred kilometers, which is not negligible 218 compared to its solid body radius ~ 2575 km (Zebker et al., 2009). Therefore, Titan's optical radius 219 in which the solar irradiance is effectively absorbed and reflected (i.e., effective radius) is generally 220 larger than its solid radius. In this section, we discuss the effective radius, which varies with 221 wavelength. We follow the method from a previous study (Smith, 1980), in which the edge of the 222 optical disk corresponds to the maximal radiance contrast. We therefore compute the gradient of 223 the calibrated radiance around the boundary of Titan's optical disk and search for the maximal and 224 minimal radiance gradients. The pixel positions with the maximal and minimal radiance gradients 225 are used to locate the left and right edges of the optical disk, respectively.

226 We first test this idea with Cassini observations of Enceladus. For Enceladus, its effective 227 radius should be equal to its solid surface because this moon does not have a visible atmosphere. 228 We first select two lines around the boundaries of the optical disk (i.e., two horizontal solid lines 229 in panel A of Fig. S8), which are along the equator of Enceladus. The calibrated radiances for the 230 pixels in the two lines are plotted in panel B of Fig. S8. The gradients of the radiance along the 231 two solid lines are presented in panel C. Then we can determine the pixel location of the maximal 232 gradient of the line around the left boundary (i.e., the location of the left edge of the optical disk) 233 and the pixel location of the minimal gradient of the line around the right boundary (i.e., the 234 location of the left edge of the optical disk). The left and right edges are shown by the two vertical 235 dashed lines respectively in panel A of Fig. S8. The distance between the two edges (i.e., the 236 product of the pixel number between the two edges and the spatial resolution) is used to determine 237 the diameter of the optical disk, and half of the diameter is the effective radius.

For the estimate of the error bar in determining the effective radius by the radiant gradient, we simply assume there is one-pixel uncertainty in such a method. So we can use the size corresponding to one pixel (i.e., spatial resolution) to estimate the error bar of the effective radius. Based on the analysis shown in Fig. S8, we have the effective radius of Enceladus is 253.8 ± 2.6 km, which is consistent with the solid radius of Eaceladus 252.1 ± 0.2 km (Thomas, 2010). This test validates the method of the maximal/minimal radiance gradient works for determining the effective radius of moons.

245 After the validation, we apply the method to the global images recorded by the ISS 12 246 filters. We try the global ISS images at both low and high phase angles. The inter-comparison of 247 the comparisons between the low and high phase angles can be used to double check the results. 248 Examples of determining the effective radius are demonstrated with ISS images recorded by the 249 red filter. The analyses of the observations at the low and high phase angles are shown in Fig. S9 250 and Fig. S10, respectively. The analyses based on the observations at a (Fig. S9) generate an 251 effective radius 2849.0±10.8 km, which is basically consistent with the effective radius 252 2857.9±12.4 km from the analyses from the observations at a high phase angle (Fig. S10).

We also test the temporal variations of the effective radius. Among the global images recorded by the ISS 12 filters, the largest number of images were recorded by the CB3 filter. Therefore we searched for the highest-quality global images of Titan recorded by the CB3 filter in different years to examine the temporal variations of its effective radius. Panels A, B, and C in Fig. S11 show the global images recorded by the CB3 filter in 2004, 2009, and 2016, respectively. The effective radii in the three years are displayed in panel D of Fig. S11. We can see the effective radius at the CB3 wavelength is basically constant with time.

Titan's effective radii at the wavelengths covered by the ISS 12 filters are shown in Fig. S12. Panel A shows that the differences of effective radius between the low and high phase angles are smaller than the error bars, which suggests that the results based on the low and high phase angles are consistent. The analyses at both low and high phase angles show that the effective radius decreases with wavelength except for small oscillations at some wavelengths. The effective radii averaged over the analyses at the low and high phase angles are shown in panel B of Fig. S12.

266 We also validate the effective radii measured from the Cassini/ISS observations with some 267 previous studies. Based on the Pioneer observations, Smith (1980) measured Titan's effective radii at the blue (440 nm) and red (640 nm) wavelengths. There are also some theoretical investigations 268 269 of Titan's effective radii (55-57). With a model study, Toon et al. (1992) suggested a formula to 270 estimate the effective radii at visible wavelengths. In panel A of Fig. S13, we compare the Cassini 271 results with the two previous studies. First, the Cassini ISS results are basically consistent with the 272 Pioneer IPP results (i.e., the differences between them are smaller than the error bars of 273 measurements). The trend of decreasing effective radius with wavelength, which is shown by the 274 ISS observations, is also consistent with the results from the model study (Toon et al., 1992). In 275 panel B of Fig. S13, we further compute the ratio of the difference between the ISS results and the 276 previous results over the ISS results. We find that the ratio is smaller than 3%, which also suggests 277 that the Cassini ISS results are approximately consistent with the previous results.

278 The Cassini ISS multi-filter observations cover limited wavelengths only, so we have to 279 use the VIMS observations to determine Titan's effective radii in more wavelengths. Generally, 280 the spatial resolution in the radial direction is much better for the VIMS solar-occultation observations (Maltagliati et al., 2015; Cours et al., 2020) than for the VIMS direct observations of 281 282 Titan at low and high phase angles. The glow of Titan's atmosphere in some wavelengths (e.g., 283 3200-3500 nm) can reach ~ 700 km in the VIMS images of Titan (Baines et al., 2005), which 284 makes the method of determining the effective radius by the radiance gradient based on the VIMS 285 images of Titan at low and high phase angles does not work. Therefore, the VIMS solar-occultation 286 observations are used to determine the effective radii in the longer wavelengths.

287 The solar-occultation observations were performed by the infrared part of the VIMS instrument (~ 884-5000 nm) only, so there are no data acquired in the visible wavelengths during 288 289 the solar-occultation observations. The solar-occultation can help us determine the cross section 290 of Titan's atmospheric extinction (Maltogliati et al., 2015; Cours et al., 2020), and the cross section 291 is used to determine the effective radius. Four solar-occultation observations are used in our 292 measurements of the effective radii in the infrared wavelengths, and we average the four 293 measurements to get the effective radii in the VIMS infrared wavelengths (see Fig. S14). In the 294 radial direction, the spatial resolutions change from 7 km to 15 km for the four solar-occultation 295 observations (Maltagliati et al., 2015). Such spatial resolutions are comparable to the standard 296 deviation of the four measurements of effective radius (~ 5-20 km). We combine the spatial 297 resolution of observations and the standard deviation of measurements to represent the 298 uncertainties of the VIMS measurements of effective radii. It should be mentioned that the standard 299 deviation of the VIMS measurements of Titan's effective radius is probably related to the spatio-300 temporal variations of Titan's atmospheric processes (e.g., haze) (West et al., 2018, Seignovert et 301 al., 2021). The uncertainties of the VIMS measurements of Titan's effective radius, which includes 302 the possible spatio-temporal variations, are accounted in the analysis of Titan's Bond albedo and 303 hence the absorbed solar energy. Figure S15 further compares the effective radii between the ISS

and VIMS measurements. The differences between the ISS and VIMS measurements are smaller
 than the error bars of the VIMS measurements, which suggests that they are basically consistent.

307 S7. Validation of Cassini ISS and VIMS Results

After determining Titan's effective radii by the ISS and VIMS observations (Figs S12 and S14), we can calculate Titan's full-disk albedo by integrating the calibrated radiance over the disks with the effective radii in these wavelengths covered by the ISS and VIMS (see section "Cassini ISS/VIMS Data and Data Processing"). To validate the full-disk albedo computed from the ISS and VIMS observations, we first made an inter-comparison between the ISS and VIMS results. Then we compared the ISS and VIMS results with other studies.

314 Figure S16 shows the comparison of Titan's full-disk albedo between the ISS and VIMS 315 observations. Only 10 ISS filters are displayed in this figure because the wavelengths of the UV1 316 and UV2 filters are outside of the VIMS wavelength range. The VIMS observations dispersed in 317 different years of the Cassini epoch, and it is difficult to find a simultaneous ISS observation for 318 each VIMS observation. Titan's full albedo, based on all available ISS and VIMS observations 319 during the Cassini epoch, is displayed in Fig. S16. Both the ISS and VIMS observations show that 320 Titan's albedo decreases from phase angle 0° to ~ 100-140° then increases from phase angle ~ 321 $100-140^{\circ}$ to ~ 160° . The increase of albedo at the high phase angles is due to the efficient forward 322 scattering of sunlight by Titan's thick atmosphere (Garcia Munoz et al., 2017). Figure S16 323 demonstrates that the ISS and VIMS measurements of Titan's full-disk albedo are consistent.

324 The spacecraft Pioneer 11 conducted observations of Titan's full-disk at the blue (440 nm) 325 and red wavelengths with phase angle varying from 21.9 ° to 95.9 °. The range of phase angle from the Pioneer 11 observations are less than that of the Cassini ISS observations (0-167°), but the 326 327 Pioneer 11 observations are the best in coverage of phase angle before the Cassini epoch. Here, 328 we compare the Pioneer 11 observations at blue and red wavelengths⁴⁶ with the Cassini ISS and 329 VIMS observations at the corresponding wavelengths (Fig. S17). The Cassini blue (459 nm) and 330 red (648 nm) filters have slightly different wavelengths from the blue and red wavelengths of the 331 Pioneer 11. Figure S17 shows that the differences between the Cassini and Pioneer measurements 332 are smaller than the uncertainties in the Pioneer measurements, which suggests that the Cassini 333 measurements are consistent with the Pioneer measurements.

334 The Pioneer observations have the best coverage of phase angle among the observations 335 before the Cassini epoch, but the Pioneer observations are limited to two wavelengths (blue and 336 red). The Earth-based ESO provide high-spectral-resolution (~ 0.4 nm) measurements of Titan's 337 full-disk albedo in a relatively wide wavelength range (305-1050 nm) (Karkoschka, 1994, 1998). 338 The geometry of Earth-Sun-Titan makes the phase angles of the Earth-based observations vary in 339 a very narrow range. For the measurements conducted in 1993 and 1995 (Karkoschka, 1994, 1998), the phase angles are 2.7° and 5.7°, respectively. Here we use the ESO observations in 1995 340 341 (Karkoschka, 1998) because the observational time is closer to the Cassini epoch. The ISS UV1 342 filter (~ 264 nm) has the wavelength shorter than the low limit the ESO wavelength range (~305 343 nm). To validate the ISS UV1 measurements, the observations recorded by the FOS on the HST, 344 which cover the wavelength 264 nm, are used. As shown in Fig. S18, the comparisons between 345 the Cassini ISS measurements and the other measurements suggest that they are basically 346 consistent.

347 The Cassini ISS observations cover limited wavelengths, but the Cassini VIMS 348 observations have much better coverage of wavelength. So we also compare the spectral lines 349 between the VIMS observations and the ESO observations. The VIMS observations do not have high-quality global images at phase angle less than 11.7°. Here, we compare the ESO observations

with the VIMS observations at a phase angle 11.7° , which is the closest to the phase angle 5.7° for the ESO abservations in 1005. Figure 510 shares the true shares in 1005 and 1005.

the ESO observations in 1995. Figure S19 shows the two spectra, which suggests that the ESO spectra and the VIMS spectra have the same spectral structures. However, there are significant

differences between the two spectra, which can be explained by the different phase angles between

them (5.7° for the ESO spectra and 11.7° for the VIMS spectra). In addition, the spectral resolution

- is much worse in the VIMS observations ($\sim 4-25$ nm) than in the ESO observations (~ 0.4 nm).
- 357 Therefore, the VIMS observations cannot resolve some fine spectral structures, which are revealed
- 358 by the ESO high-spectral-resolution observations.
- 359

360 S8. Filling Observational Gaps in Phase Angle

361 In this study, we want to examine the temporal variations of Titan's Bond albedo and hence 362 the reflected/absorbed solar power during the Cassini epoch. Such examinations are further 363 combined with our previous measurements of the temporal variations of Titan's emitted power 364 (Creecy et al., 2019) to determine the temporal variations of Titan's radiant energy budget. We 365 first organize the ISS observations by time. In addition, the computation of Bond albedo requires 366 the observations at different phase angles (Li et al., 2018). Therefore, the computed full-disk albedo 367 is organized over time and phase angle. Figure S20 displays the organized full-disk albedo from 368 the ISS observations in the two-dimensional domain of time and phase angle.

369 Figure S20 suggests that there are observational gaps in phase angle and such observational 370 gaps vary from year to year. We use the least-squares fitting (Bevington and Robinson, 2003) to 371 fill the observational gaps in phase angle. In our study of Jupiter's Bond albedo, we tried different 372 functions (Li et al., 2018) to fit the phase function of Jupiter's full-disk albedo and we concluded 373 that the polynomial functions of phase angle work well for fitting the phase function. Here, we try 374 different polynomial functions to fit Titan's data, and we find a six-order polynomial function $P(f) = c_1 f^6 + c_2 f^5 + c_3 f^4 + c_4 f^3 + c_5 f^2 + c_6 f + c_7$ (where f is phase angle and the parameters c_1 , 375 376 c_2, c_3, \ldots, c_7 are fitting coefficients to match the observations with least-squares method) has the 377 smallest fitting residual. An example of such fitting is shown in Fig. S21. For comparison, a physically-based function (i.e., the double Henyey-Greenstein (H-G) function) is also included in 378 379 Fig. S21. The double H-G function (Henyey & Greenstein 1941; Hapke, 2002) is defined as $P(A_{HG}, g_1, g_2, f, f) = A_{HG} \cdot (fP_{HG}(g_1, f) + (1 - f)P_{HG}(g_2, f))$, where A_{HG} is the coefficient to match 380 the amplitude of the observed phase function. The term $P_{HG}(g, f)$ represents both forward (with a 381 factor g_1 and $g_1 \hat{i}$ [0,1]) and backward (with a factor g_2 and $g_2 \hat{i}$ [-1,0]) scattering lobes, 382 respectively. The factor $f(f \mid [0,1])$ stands for the fraction of the forward versus backward 383 scattering. The term $P_{HG}(g, f)$ (g can be g_1 or g_2 and f is phase angle) has a form as 384 $P_{HG}(g,f) = (1-g^2)/(1+g^2+2g \cdot \cos f)^{3/2}$. Figure S21 shows the polynomial-function fitting is 385 386 better than the double H-G fitting. In addition, the double H-G fitting is smaller than the 387 polynomial-function fitting at the highest phase angles (165-180°). Therefore, our following 388 estimate of the fitting uncertainty at the highest phase angles by assuming the uncertainty can reach 389 the fitted values of the polynomial-function fitting (see B.2 in section S11. Analyses of the 390 uncertainties in determining Titan's radiant energy budget) is good enough. Note that Titan's albedo is larger at very high phase angles (i.e., $> 170^\circ$) than at 0° phase angle, which is caused by 391 392 Titan's thick atmosphere and efficient forward scattering of sunlight (Garcia Munoz et al., 2017).

Then the six-order polynomial functions from the least-squares technique are used to fit the phase functions and fill the observational gaps in phase angle for the ISS-derived albedo in the 12 filters during the Cassini epoch. Note that the direct fitting does not work for the data in some years with very poor coverage of phase angle. For these years, we increase the coverage of phase angle by linear interpolation/extrapolation in time from the neighboring years before doing the fitting. The fitting results for the ISS-derived albedo during the Cassini epoch are presented in Fig. S22.

The fitting residuals, which are the differences between the fitted results (Fig. S22) and the data (Fig. S20), are used to evaluate how well the fitting preforms. The ratios between the fitting residuals and the raw data are shown in Fig. S23. Most of the ratios are less than 5% and a few ratios are larger than 5% but smaller than 10%. Generally, the six-order polynomial functions work well in fitting the ISS full-disk albedo and hence filling the observational gaps in phase angle.

There are relatively few high-quality global images from the VIMS observations, so we only find 10 VIMS observations with the phase angle varying from 11.7° to 159.1°. The 10 observations disperse in different years of the Cassini epoch, so the VIMS observations themselves cannot resolve the temporal variations of the phase function of Titan's full-disk albedo. The temporal variations retrieved from the ISS observations are extrapolated to the VIMS wavelength range to address the temporal variations of the phase functions of Titan's Bond albedo in that range (see section "Filling Observational Gaps in Wavelength and Time").

412 The available VIMS high-quality spectra are displayed in Fig. S24. The 10 VIMS 413 observations do not cover the complete range of phase angle, so we have to fill the observational 414 gaps in phase angle. We first try the six-order polynomial function for the VIMS data. The 415 comparison of fitting in the overlap wavelengths between the ISS and VIMS suggests that the six-416 order polynomial function works well for the range of phase angle for the VIMS observations 417 (11.7°-159.1°), but it does not work well for fitting the VIMS data in the ranges of low and high phase angles ($< 11.7^{\circ}$ and $> 159.1^{\circ}$) because the VIMS observations are lacking in these ranges. 418 419 The ISS observations have better coverage than that of the VIMS observations, especially in the 420 range of the low and high phase angles. So we first use the ISS observations in low and high phase 421 angles to fill the VIMS observational gaps. Then we use the six-order polynomial function to fit 422 the data, and such a fitting works well for filling the VIMS observations gaps.

423 The wavelength range of the VIMS observations is larger than the wavelength range of the 424 ISS observations. For the VIMS wavelengths covered by the ISS 12 filters, we follow the method 425 discussed above to do the fitting and then fill the observational gaps. For the VIMS wavelengths 426 not covered by the ISS filters, we interpolate/extrapolate the ISS observations from the ISS 427 wavelengths to the VIMS wavelengths to fill the VIMS observational gaps at the low and high 428 phase angle first. Then we apply the six-order polynomial function to fit the VIMS data and then 429 fill the observational gaps. After filling the VIMS observational gaps in phase angle, we have the 430 full-disk albedo in the two-dimensional domain of wavelength and phase angle for the VIMS 431 wavelength range (350-5131 nm), which is shown in Fig. S25.

Figure S26 displays the ratio between the fitting residual (i.e., fitting results – observational results) and the observational results for these phase angles where the VIMS observations exist. Most of the fittings have ratios less than 10%, but there are fittings with residual ratios larger than 10% or even 20%. We further examine these fittings with the large residual ratios. We find that the residual ratios generally get bigger with longer wavelengths (especially in these wavelengths longer than 2000 nm). The observational data (i.e., full-disk albedo) are extremely small when wavelengths longer than 2000 nm, so even the small fitting residuals can make the residual ratiospretty large.

These fittings with large residual ratios are mainly in the wavelengths longer than 2000 nm. In these longer wavelengths, solar spectral irradiance contributes a very small fraction (< 6%) to the total solar power. So the uncertainties in these wavelengths do not significantly affect our measurements of Titan's Bond albedo and hence the reflected/absorbed solar power. However, the uncertainty related to such fitting residuals is considered in our analyses of the measurement uncertainties (see section "Analyses of the Measurement Uncertainties").

446

447 S9. Filling Observational Gaps in Wavelength and Time

448 The Cassini observations of Titan's full-disk albedo have observational gaps in not only 449 phase angle but also wavelength. After filling the observational gaps in phase angle, which is 450 discussed in the previous section, we work on the observational gaps in wavelength. Both the ISS 451 and VIMS data have observational gaps in wavelength. The ISS 12 filters record Titan's albedo in 452 separated and limited wavelengths from ultraviolet (~ 264 nm) to near infrared (~ 939 nm) and the 453 VIMS observations do not cover the wavelengths shorter than 350 nm. In order to compute Titan's 454 full-disk albedo, we need measurements of Titan's albedo in the wavelength range of 0-5131 nm. 455 The SSI in this wavelength range contributes to more than 99.5% of the total solar power.

456 We first fill the observational gaps in wavelength for the Cassini ISS observations. The 457 ground-based observations (see Fig. S3) and the Cassini VIMS observations both suggest that the 458 magnitude of Titan's albedo spectra change with phase angle but the spectral structure and shape 459 basically stay constant. It means that the phase functions are correlated among different 460 wavelengths. With the least-squares fitting, we have derived Titan's full-disk albedo over the 461 whole range of phase angle for these wavelengths covered by the Cassini ISS 12 filters (see Fig. 462 S22). Please note that the ISS UV1/2 observations do not cover all years during the Cassini epoch 463 (see Fig. S20), and we use interpolation and extrapolation to fill the observations gaps in time for 464 the UV1/2 results. Then, we use the complete phase functions (i.e., distribution over phase angles 465 0-180°) at the Cassini ISS 12 wavelengths to derive the phase functions at all wavelengths from 466 264 nm (i.e., the shortest wavelength of the ISS observations) to 939 nm (i.e., the longest 467 wavelength of the ISS observations) by referring to the spectral shape at the phase angle 5.7° which 468 is from the high-spectral-resolution measurements (see Fig. S3). Figure S27 displays examples of 469 the ISS-derived albedo in the two-dimensional domain of wavelength and phase angle for these 470 years during the Cassini epoch.

471 To validate the ISS-derived albedo in the two-dimensional domain of wavelength and 472 phase angle, we compare the derived results between the ISS and VIMS observations. We first 473 average the ISS-derived two-dimensional albedo over the Cassini epoch. Then we compare the 474 time-mean ISS-derived albedo with the VIMS-derived albedo in the overlap wavelengths (350-475 939 nm). Figure S28 shows that the results are basically consistent between ISS (panel A) and 476 VIMS (panel B) results. Figure S29 further provides the differences and differences ratio between 477 the ISS-derived and the VIMS-derived results. Panel A shows that large differences are mainly 478 concentrated in the low and high phase angles. However, the difference ratios suggest that the 479 values are less than 15% even for these large differences in the low and high phase angles.

To address the temporal variations of Titan's Bond albedo and hence the reflected/absorbed solar power during the Cassini epoch, we require the time series of Titan's full-disk albedo in the two-dimensional domain of wavelength and phase angle with the complete coverage of phase angle (0-180°) and wavelength (0-5131 nm). The above ISS-derived albedos (Fig. S27) with the

484 complete coverage of phase angle are used for the wavelength range 264-939 nm. It should be 485 mentioned that we get Titan's albedo in the two-dimensional domain from both the ISS 486 observations (Fig. S27) and the VIMS observations (Fig. S25) for the overlap wavelengths (i.e., 487 350-939 nm) between the ISS and VIMS observations. In this study, we use the ISS-derived results 488 for the overlap wavelengths (i.e., 350-939 nm) because: (1) the ISS observations resolved the 489 temporal variations of the phase functions for Titan's full-disk albedos; (2) the coverage of phase 490 angle is much better for the ISS observations than for the VIMS observations so that the fitted 491 phase functions are more precise for the ISS observations than for the VIMS observations; and (3) the spectra from the Earth-based observations⁴², which are used to derive the ISS albedo (Fig. 492 493 S27), have a very high spectra resolution ~ 0.4 nm. Such a spectral resolution is much better than 494 the spectral resolution of the VIMS observations (~ 4-24 nm), so that some fine spectral structures 495 can be better resolved.

496 The low and high limits of the wavelengths for the ISS observations are 264 nm (UV1) and 497 939 nm (CB3), respectively. For the temporal variations of Titan's full-disk albedo in the 498 wavelengths shorter than 264 nm (i.e., 0-264 nm), we refer to the ISS measurements at the UV1 499 filter (264 nm). We first interpolate and extrapolate the UV1 phase functions in 2009 and 2015 500 (Fig. S22) to the whole Cassini epoch (2004-2017). We then extrapolate the available geometric 501 albedo spectra from the Cassini UVIS (150-190 nm) and the HST/FOS (190-305 nm) (Fig. S4) to 502 the wavelength range 0-150 nm to get the geometric albedo spectra in the wavelength range of 0-503 264 nm. Finally, the extrapolated phase function in each wavelength of the wavelength range 0-504 264 nm is combined with the available albedo at the same wavelength to derive Titan's albedo 505 over the complete range of phase angle. For each year of the Cassini epoch, we build the albedo 506 in the two-dimensional domain of wavelength and phase angle for the wavelength range 0-264 507 nm.

508 For the temporal variations of Titan's albedo at wavelengths longer than 939 nm (i.e., 939-509 5131 nm), we combine the ISS and VIMS observations together. The basic idea is that we 510 extrapolate the temporal variations at 939 nm, which are based on the ISS CB3 observations, to 511 the wavelength range 939-5131 nm. The albedos in the wavelength range 939-5131 nm (Fig. S25), 512 which are derived from the VIMS observations, are assumed to be the time-mean albedo. During 513 the process of extrapolating the temporal variations of the ISS observations to the VIMS 514 wavelengths 939-5131 nm, the time-mean two-dimensional albedos (Fig. S25) are used as a 515 reference.

After addressing the temporal variations of Titan's albedo in the wavelengths outside of the ISS coverage (i.e., 0-264 nm and 939-5131 nm), we have the two-dimensional albedo in the complete coverage of phase angle (0-180°) and wavelength (0-5131 nm) for each year during the Cassini epoch (2004-2017). Some examples of such two-dimensional albedo are shown in Fig. S30.

521

522 S10. Computing Titan's Bond Albedo and the Related Qualities

Based on Titan's full-disk albedo in the complete domain of wavelength and phase angle for each year of the Cassini epoch (Fig. S30), we can compute Titan's monochromatic Bond albedo and the related qualities at each wavelength for each year of the Cassini epoch. The monochromatic geometric albedo, which is defined as the albedo at phase angle 0°, is automatically found from Fig. S30. The phase integral, which is the integral of phase function of Titan's albedo, can be computed from the distribution of Titan's albedo with phase angle at each wavelength. Finally, we can derive the monochromatic Bond albedo from the product of the monochromatic geometric 530 albedo and monochromatic phase integral (see section "Method of Computing the Bond Albedo"). 531 Figure S31 displays the monochromic geometric albedo, phase integral, and Bond albedo 532 in the two-dimensional domain of time (2004-05-2017) and wavelength (0-5131 nm). For the 533 distribution with wavelength, the geometric albedo has roughly the same spectral structures as 534 those shown in the Earth-based observations (Fig. S3) and the VIMS observations (Fig. S24) at 535 high phase angles. The phase integral largely increases with wavelength. In the visible part (300-536 800 nm), the phase integral has the smallest values ~1.6-2.3. In some infrared parts (e.g., 3900-537 4500 nm), the phase integral can reach the largest values ~5-5.5. In the direction of wavelength, 538 Panel E shows that the Bond albedo has similar distributions as those of the geometric albedo. The 539 Bond albedo reaches maxima (~ 0.4-0.5) around the visible wavelengths between 600 nm and 800 540 nm, and it becomes very small (< 0.2) at wavelengths longer than 2200 nm. The large Bond albedo 541 between 600 nm and 800 nm is mainly due to the large geometric albedo in the same wavelengths. 542 The small monochromatic Bond albedo at wavelengths longer than 2200 nm is because the small 543 geometric albedo is dominant over the large phase integral at these wavelengths. The anomalies 544 (i.e., difference from time-mean) suggest that the temporal variations of the geometrical albedo 545 (panel B of Fig. S31) and Bond albedo (panel F of Fig. S31) are mainly concentrated in the 546 wavelength range 400-1000 nm. The anomaly of the phase integral (panel D of Fig. S31) displays 547 the temporal variations basically in all wavelengths.

548 Based on the distribution of the monochromatic Bond albedo (Panel E of Fig. S31), we can 549 compute the wavelength-average Bond albedo (i.e. Titan's Bond albedo) by weighting the 550 monochromatic Bond albedo with the SSI (Fig. S2), as we did in our study of measuring Jupiter's 551 Bond albedo (Li et al., 2018). Titan's Bond albedo during the Cassini epoch is displayed in Fig. 1 552 in the main text.

553 The product between the SSI at Titan (panel A of Fig. S2) and the monochromatic Bond 554 albedo (panel E of Fig. S31) generates the reflected SSI. Subtracting the reflected SSI from the 555 SSI at Titan, we have the absorbed SSI at Titan. Multiplying the SSI, the reflected SSI, and the 556 absorbed SSI by the effective radius at each wavelength, we have the total SSI, the total reflected 557 SSI, and the total absorbed SSI over Titan's optical disk at each wavelength. Integrating the total 558 SSI, the total reflected SSI, and the total absorbed SSI at each wavelength over the complete 559 wavelength range 0-5131 nm, we have the total solar power, the total reflected solar power, and 560 the total absorbed solar power at Titan. Their temporal variations during the Cassini epoch are 561 presented in Fig. 2 in the main text.

562

563 S11. Analyses of the uncertainties in determining Titan's radiant energy budget

In this section, we discuss the uncertainties in computing Titan's radiant energy budget. Titan's radiant energy budget is determined by the emitted thermal energy and the absorbed solar energy of Titan. Therefore, we mainly discuss the uncertainties in the measurements of Titan's emitted power and absorbed power. In addition, other possible energy sources (e.g., emission from Saturn and Titan's internal heat), which possibly affect the radiant energy budget, are also discussed.

570 A. Uncertainties in the measurements of Titan's emitted thermal power

We first discuss the uncertainties in computing Titan's emitted power with the Cassini/CIRS observations. Such uncertainties were briefly discussed in our previous studies of Titan's emitted power (Li et al., 2011; Creecy et al., 2019), which are based on the methodology we developed in our investigation of Saturn's emitted power (Li et al., 2010). Here, we provide more discussions on the uncertainty in computing Titan's emitted power. There are two dominant 576 uncertainty sources in computing Titan's emitted power with the Cassini/CIRS observations: (1)

577 the uncertainty related to the CIRS data calibration; and (2) the uncertainty related to filling the 578 observational gaps of the CIRS data.

579 A.1. Uncertainty in the CIRS data calibration

580 The basic approach for calibrating the CIRS measurements to radiance of targets (e.g., 581 Titan) was introduced in the introductory CIRS paper (Flasar et al., 2004) and discussed in our 582 previous study of Saturn's emitted power (Li et al., 2010). The main process of the CIRS 583 calibration is to remove the radiance of the background (i.e., the instrument itself) from the 584 radiance of targets. To estimate the background radiance, the CIRS routinely viewed deep space 585 (i.e., zero radiance from target) and recorded the spectra of the deep space. Generally, the spectra 586 of deep space are much smaller than the spectra of targets. Figure S32 displays the comparison 587 between the spectra of Titan and the typical spectra of deep space, which were recorded by the 588 three focal planes (FP1, FP3, and FP4) of the CIRS. Figure S32 suggests that calibration 589 uncertainty mainly comes from the FP1, which is dominant among the three focal planes. So we 590 focus on the FP1 data in the following discussion.

591 The absolute radiometric calibration of the FP1 was operated at 170 K instrument 592 temperature. The radiance from deep space is the main error source for the CIRS calibration. 593 Figure S33 shows the temporal variations of the FP1 spectra of deep space, which suggests a good 594 stability. The uncertainty related to the background noise in the FP1 spectra is estimated as follows: 595 we randomly choose ~ 100,000 spectra of deep space for each year of 2004–2017. These spectra 596 are first integrated in wavenumber because Titan's spectra are integrated when calculating the 597 emitted power. The mean value and standard deviation of the 100,000 wavenumber-integrated 598 spectra are used as an estimator for the uncertainty due to the background radiance in each year. 599 The mean values and the standard deviations of some years were presented in one of our previous 600 studies (Li et al., 2010). We add the absolute mean value to the standard deviation to estimate the 601 absolute calibration uncertainty of Titan's emitted power for each Earth year.

602 It should be mentioned that the thermal control of the CIRS instrument possibly affects the 603 data calibration. The entire FP1 interferometer and detector were operated at the 170 K instrument 604 temperature. The sensors were calibrated before flight and didn't show any evidence of drift during 605 the Cassini observational period. The temperature control was about 0.1 K, so an absolute accuracy 606 of 0.2 K overall is a good estimate for the upper limit. Panel A of Fig. 34 shows the blackbody 607 spectra of the CIRS instrument at 170 K. In contrast, we also plot the blackbody spectra at 170.2 608 K by adding the temperature control (0.2 K) to the designed temperature of the CIRS instrument 609 (170 K). Panel B shows the difference between the two spectra shown in panel A, which is used 610 to estimate the spectral noise related to the temperature control of the CIRS instrument. Panel C is 611 the comparison between the spectral noise and typical spectra of deep space, which suggests that 612 the spectral noise related to the temperature control is much smaller than the spectra of deep space.

613 Therefore, we only consider the spectra of deep space for the CIRS data calibration.

614 A.2. Uncertainty related to filling CIRS observational gaps

615 The other uncertainty of computing Titan's emitted power is related to filling observational 616 gaps of the CIRS data in emission angle. Based on the uncertainty analysis in our study of Saturn's 617 emitted power (Li, et al., 2010), we have the sum of unknown radiance in the observational gaps 618 along a single latitude ($P_{emit}(N)$) as

619
$$P_{emit}(N) = 2\rho \sum_{k=1}^{N} I(d_k) \cos d_k \sin d_k \mathsf{D} d = \sum_{k=1}^{N} c_k I_k$$
(1)

620 where N the number of the radiance at the unknown emission angles and $I(\mathcal{O}_k)$ is the radiance at

- 621 the unknown emission angles (d'_k). In the above equation, we also simplify the equation with a
- 622 coefficient c_k , which is represented by $c_k = 2\rho \cos d_k \sin d_k Dd$. Then the total difference between

623 the fitted radiance and the real radiance for all observational gaps along the latitude $(P_{emit}(N)^{l})$ 624 can be expressed as

$$P_{emit}\left(N\right)^{\complement} = \mathop{\bigotimes}\limits_{k=1}^{N} c_k I_k^{\complement}$$
⁽²⁾

626 where I_k^{l} is the radiance difference between the fitted value and the real value at unknown emission 627 angles.

628 The variance of $P_{emit}(N)^{\complement}$ is used to estimate the uncertainty of emitted power related to 629 filling the observational gaps. The variance of the sum of multiple variables can be determined by 630 the following equation (23)

631
$$S^{2} \overset{\mathfrak{A}}{\underset{e}{\Diamond}} P_{emit} \left(N \right)^{\overset{\circ}{\underset{i}{\leftrightarrow}}} = \overset{N}{\underset{k=1}{\overset{o}{a}}} c_{k}^{2} S_{k}^{2} + 2 \overset{N}{\underset{i=2}{\overset{i-1}{a}}} \overset{i-1}{\underset{j=1}{\overset{o}{a}}} c_{i} c_{j} S_{ij}^{2}$$
(3)

where S_k^2 is the variance of the radiance difference I_k^{l} and S_{ij}^2 is the covariance of the radiance 632 difference at two different unknown emission angles with the two corresponding coefficients C_i 633 and c_j . Our previous analyses (19, 20) show that least-squares fitting does a good job in fitting the 634 recorded radiance. Figure S35 shows an example of the fitting and the fitting residual. The fitting 635 residual is used to estimate the variance of the radiance difference O_k^2 at the observational gaps. 636 The covariance d_{ij}^2 will disappear if we assume that the radiances at different unknown points are 637 independent from each other. Then Eq. (3) can be used to compute the uncertainty related to filling 638 639 the observational gaps.

640 A.3. Total uncertainty of emitted power

625

With the analysis in the above section, we have the uncertainty related to filling the observational gaps at each latitude. In addition, we assume that the calibration uncertainty is constant with latitude. As an example, the meridional distribution of calibration uncertainty and filling uncertainty in 2009 are shown in Fig. S36. It suggests that the fitting uncertainty is larger than the calibration uncertainty by roughly one order of magnitude.

646 We combine the two uncertainties as the total uncertainty of emitted power by 647 $S_{all} = \sqrt{S_{calibration}^2 + S_{fitting}^2}$. Now, we discuss the uncertainty of the global-average emitted power. 648 The global-average emitted power can be written as (Li et al., 2010)

649
$$\overline{P} = \frac{1}{2(1 - 2e/3)} \sum_{i=1}^{N} (1 - 2e\sin^2 f_i) P(f_i) \cos f_i Df = \sum_{i=1}^{N} c_i P(f_i)$$
(4)

where e is the oblateness of Titan (~(2574.91-2574.34)/2574.91 ~ 0.00022), f is latitude, and $P(f_i)$ is the emitted power at the latitude f_i . The index N is the number of latitude bands from pole to pole. The coefficient c_i is represented by $c_i = \left[\left(1 - 2e\sin^2 f_i \right) \cos f_i Df \right] / \left[2 \left(1 - 2e/3 \right) \right]$. With the coefficient, the global-average emitted power is the sum of multiple variables. Again, we can
use the analysis of the variance of the sum of multiple variables (Bevington and Robinson, 2003)
to estimate the uncertainty of the global-average emitted power.

656 Using the uncertainty of emitted power at each latitude $(S^2(P_i))$ (Fig. S36) and assuming 657 the emitted power is independent at different latitudes, we have the uncertainty of the global-658 average emitted power as

659 $S^{2}\left(\overline{P}\right) = \bigotimes_{i=1}^{N} c_{i}^{2} S^{2}\left(P_{i}\right)$ (5)

Based on eq. (5), we can compute the uncertainty of global-average emitted power in each Earth year during the Cassini time period of 2004–2017. Figure S37 shows the time series of different uncertainties for Titan's emitted power. Panel B of Fig. S37 suggests that the ratios between the total uncertain and Titan's emitted power are about 0.3 percent for the Cassini/CIRS measurements. Such ratios are much smaller than the ratios in the meridional direction (Fig. S36), because averaging uncertainties over all latitudes smoothens the uncertainties into a much smaller value, as Eq. (5) suggests.

The effective radius is different between Titan's emitted thermal emission and absorbed solar irradiance, so we integrate the uncertainty over the effective radius of Titan's thermal emission (i.e., 2575+500 km) (Li et al., 2011) to get the uncertainty of Titan's sphere-integrated emitted power (Fig. S38). Such uncertainty is used in the comparison between the sphereintegrated emitted thermal power and the disk-integrated absorbed solar power (Fig. 3 in the main text), which helps to determine if Titan's global radiant energy budget is balanced.

673 B. Uncertainties in the measurements of Titan's absorbed solar power

The absorbed solar power is determined by Titan's Bond albedo with known solar flux at Titan (Fig. S2). Therefore, the uncertainties in the absorbed solar energy mainly come from the uncertainties in computing Titan's Bond albedo with the Cassini observations. At each wavelength, the monochromatic Bond albedo (A_1) can be expressed as below (Li et al., 2018)

678
$$A_{I} = \frac{2}{S/D^{2}} \sum_{f_{i}=0}^{f_{i}=180^{\circ}} I_{I}(f_{i}) \sin f_{i} \mathsf{D} f$$
(6)

679 where / is wavelength, ρS is the solar constant at Earth, *D* is the distance of the planet from the 680 Sun in astronomical units (1 AU = 149.6×10⁹ m), and $I_{I}(f_{i})$ is the reflected solar irradiance at

681 phase angle f_i .

We organize the uncertainty sources in the measurements of monochromatic Bond albedo in three categories: (1) the uncertainty in calibrating the Cassini ISS and VIMS images; (2) the uncertainty related to filling observational gaps with the least-squares fitting; and (3) other uncertainties.

686 B.1. Uncertainty in the ISS and VIMS data calibration

We first discuss the uncertainty in the calibration of the ISS and VIMS data. For the ISS images, we use the latest version of the Cassini ISS CALibration software (Knowles et al., 2020), to calibrate the data. The calibrated ISS images are generated with a unit of radiance, as shown in Fig. S39. In Fig. S39, the domain outside Titan's disk corresponds to deep space. It should be mentioned the calibrated radiance in the domain of deep space, which is used to estimate the absolute calibration of the CIRS spectra, cannot be used to estimate the absolute calibration uncertainty of the ISS images. In the ISS images, the domain of deep space contains light from Titan's disk spreading out by the point spread function, plus stray light from scattering of Titanlight off the structures in the telescope (West et al., 2010; Knowles et al., 2020).

696 In the ISS calibration papers (West et al., 2010; Knowles et al., 2020) and the Cassini 697 calibration manual (https://pds-rings.seti.org/viewmaster/volumes/COISS_0xxx/COISS_0011/), 698 the calibration uncertainties, which include many error sources (e.g., uneven bit-weighting, 2-Hz 699 noise, dark current in the ISS cameras, bright/dark pixel pair artifacts from anti-blooming mode, 700 flat-field artifacts), are discussed in detail. Most of the calibration uncertainty sources vary with 701 filter, viewing geometry, and observing object. But the combined effect of these uncertainty 702 sources typically results to \sim a few percent of the calibrated radiance (2-6%) (Knowles et al., 2020). 703 Here, we assume that the total calibration uncertainty is 5% for the absolute calibrated radiance of 704 Titan's images. It should be emphasized that the 5% calibration uncertainty is not systematic and 705 it is a random error (Knowles et al., 2020). In other words, it overestimates the real radiance at 706 some wavelengths & phase angles but underestimates the radiance at other wavelengths and phase 707 angles (Knowles et al., 2020). When we integrate the recorded radiance over wavelength and phase 708 angle for computing the Bond albedo, the calibration uncertainties at different wavelengths and 709 phase angles cancel each other so that its effect on the Bond albedo becomes very small (<1%). It 710 should be mentioned that there is a systematic uncertainty related to the ISS instrumental 711 effect. For the ISS images in which Titan occupies almost all of the image frames, some of Titan's 712 reflected flux can be outside of the ISS image frames. This effect systematically underestimates 713 the real radiance. Fortunately, such images are relatively few (< 10% of the total global images 714 used in our analysis). The tests by the ISS calibration team suggest that such a systematic bias is 715 $\sim 0.5\%$ of the total scattered flux from Titan, which is accounted in our computation of Titan's 716 Bond albedo.

717 For Cassini VIMS observations, the calibration has already been discussed in a few 718 previous studies (15, 50-53). Work described by McCord et al. (2004) and Filacchione et al. (2007) 719 conducted the basic calibration processes (e.g., correcting flat-field artifacts, subtracting the noise 720 from the radiation of Cassini's power generators, and removing cosmic rays). The calibration of 721 the VIMS data of satellites of Saturn (Pitman et al., 2010) suggests that the calibration uncertainty 722 is on the order of 5% of the calibrated radiance. Based on these previous analyses, the calibration 723 uncertainty for the VIMS data is 5%. Such an estimate is the same as that of the ISS data even 724 though the two instruments did independent calibrations. As discussed with respect to the ISS 725 calibration uncertainty, the VIMS 5% calibration uncertainty is not systematic either. The random 726 calibration errors at different wavelengths and phase angles cancel each other when integrating 727 over wavelength and phase angle for Titan's Bond albedo.

The comparison of full-disk albedo between the ISS and VIMS (Fig. S16 in section "Validation of Cassini ISS and VIMS results"), which are based on the calibrated data from the two instruments, also suggests that the calibrations from the two instruments are consistent. Finally, the validation of the Cassini ISS/VIMS data by the other observations (Fig. S17-19 in section "Validation of Cassini ISS and VIMS results") also suggests that the Cassini ISS/VIMS data are calibrated well.

734 B.2. Uncertainty related to filling ISS/VIMS observational gaps

From the equation of monochromic Bond albedo (Eq. (6)), we know that the Bond albedo is determined by the reflected solar irradiance at phase angles varying from 0° to 180°. But there are observational gaps in phase angle for the Cassini ISS/VIMS observations (see Figs. S20 and S24). In the equation of monochromic Bond albedo, the part for the observational gaps can be expressed as

740
$$A'_{I} = \frac{2}{S/D^2} \sum_{i=1}^{N} I_{I}(f_{i}) \sin f_{i} \mathsf{D} f = \sum_{i=1}^{N} c_{i} I_{I}(f_{i})$$
(8)

where N is the number of unobserved points in phase angle and the coefficient c_i is expressed as $c_i = 2D^2 \sin f_i D f/S$. Then the Bond albedo for the observational gaps can be represented as the sum of multiple variables. Likewise, we use the variance of the sum of multiple variables (Bevington and Robinson, 2003) to estimate the uncertainty of the Bond albedo from filling the observational gaps as below

746 $S^{2}(A \varsigma) = \bigotimes_{i=1}^{N} c_{i}^{2} S(I_{I}(f_{i}))$

747 Now we discuss how to estimate the variance of the reflected radiance at the unobserved 748 phase angles. The observational gaps in phase angle are filled by least-squares fitting for the 749 Cassini ISS/VIMS observations of Titan's reflected solar irradiance. Figure S40 shows a fitting 750 example for the full-disk albedo recorded by the ISS observations, which suggests that the least-751 squares fitting does a good job in fitting the Cassini observations. Panel C of Fig. S39 further 752 suggests that most of the residual ratios are less than 3%. The fitting residual is used in the estimate 753 of the uncertainty in the CIRS measurements of Titan's emitted power by least-squares fitting. 754 Such a method works for the fitting for the ISS/VIMS observational gaps at the relatively s, but it 755 does not work for the ISS/VIMS observational gaps at very high phase angles.

756 The smallest phase angles of the ISS observations change from ~ 0.5° to ~ 4.4° for most 757 filters except for the filters UV1, UV2, and CB1 (see Fig. S20). The smallest phase angles for the ISS images recorded by UV1, UV2, and CB3 are between ~ 8.4° and ~ 9.8°. The smallest phase 758 759 angle for the high-quality VIMS observations is ~ 9.8° (see Fig. S24). The Cassini ISS/VIMS observations at the s are consistent with other observations at s including 0° phase angle, which 760 761 were recorded by the HST and ESO (see Fig. S18). So we think least-squares fitting works well 762 for extrapolating the ISS/VIMS observations at the s including 0° phase angle. Therefore, we use 763 the fitting residuals to estimate the variances of the ISS/VIMS observational gaps at the relatively 764 s.

765 The estimate of the variances of the ISS/VIMS observational gaps at very high phase angles 766 is a different story. In our discussion of the uncertainty in the measurements of Titan's emitted 767 power with the Cassini CIRS observations, the fitting residuals are used to estimate the variance 768 for these observational gaps at both low and high emission angles. But there is one difference 769 between the CIRS observational gaps in emission angle and the ISS/VIMS observational gaps in 770 phase angle. For the CIRS observations, we have the data around the lowest and highest emission 771 angles (0° and 90° respectively) for most latitudes (see Fig. S35), so we know the basic distribution 772 of emitted radiance along emission angle. But for the ISS/VIMS observations, we have the 773 observations around the lowest phase angle (0°) but not around the highest phase angle (180°) , as 774 shown in Fig. S20. There are no high-quality ISS/VIMS observations at the phase angles larger 775 than $\sim 165^{\circ}$ because the solar irradiance comes into the instrument and could damage the Cassini 776 detectors if the phase angle is even higher. In addition, the ground-based telescopes and Earth-777 orbiting observatories can conduct observations of Titan with s only ($< 6.5^{\circ}$) due to the orbit 778 geometry of Earth and Titan. The largest phase angle of the Pioneer 11 observations is $\sim 96^{\circ}$.

779 In summary, there are no high-quality observations of Titan at phase angles larger than 780 $\sim 165^{\circ}$. Lacking observations makes it difficult to estimate the uncertainty in filling the

(9)

observational gap at the phase angles larger than 165°. Using the fitting residuals at the lower phase angle probably underestimates the uncertainties at high phase angles larger than 165°. In this study, we use the fitting residuals at these available points (Fig. S40) to estimate the variance of the observational gaps with phase angles smaller than 165°. For the observational gaps with phase angles larger than 165°, we set larger uncertainty and assume that the uncertainty can reach the fitted values.

787 **B.3.** Other uncertainties in the measurements of Titan's Bond albedo

788 In addition to the uncertainties from the Cassini data calibration and filling the 789 observational gaps in phase angle, there are other uncertainty sources. The first one is the high-790 altitude detached haze. Some haze features have altitudes higher than 500 km, which are beyond 791 the effective radii of Titan's reflected solar irradiance. Figure S41 shows that the detached high-792 altitude haze shows in the ISS images at some filters but not in other filters. Even for the ISS 793 images including strong-reflection haze (e.g., panel A of Fig. S41), our calculation suggests that 794 the reflected solar irradiance from the detached haze is $\sim 0.2\%$ of the total reflected solar irradiance 795 from the full disk of Titan. Therefore, the high-altitude haze does not significantly contribute to 796 the uncertainty of the Bond albedo compared to the calibration and filling uncertainties. However, 797 we include it in the uncertainty analysis.

The errors in estimating the effective radii of Titan's reflected solar irradiance also introduce uncertainty in computing Titan's Bond albedo. Based on the average effective radius (~ 2884.9 km) retrieved from the ISS observations and the corresponding average error (~ 9.7 km), we simply estimate the uncertainty as $((2884.9+9.7)^2-2884.9^2)/2884.9^2 \sim 0.7\%$.

802 Titan has a thick atmosphere, which is not uniform in latitude and longitude. In addition, 803 atmospheric processes (e.g., clouds) vary with time. Finally, the solar irradiance can be reflected 804 from Titan's surface at some wavelengths (e.g., the ISS CB3 filter). The optical characteristics of 805 Titan's surface vary spatially. The heterogeneous nature of the atmosphere and surface and their 806 possible temporal variations can introduce more uncertainty in measuring the full-disk albedo of 807 Titan. Figure S42 shows full-disk images of Titan recorded by the ISS CB3 filter at different times 808 but with the same phase angle (~ 14°). The CB3 images record both the atmosphere and surface 809 of Titan, so the CB3 images at different times can be used to address the heterogeneous property 810 of Titan's atmosphere and surface and their temporal variations. The first two global images are separated by ~ 3 days, which are shorter than the orbital period of Titan around Saturn (~ 16 Earth 811 812 days). The two images cover different longitudes (panel A mainly covers longitudes ~ 200-360° 813 and 0-20° and panel B mainly covers longitudes ~150-330°). So the comparison between panels 814 A and B can help us to address not only the heterogeneous property but also the diurnal variation 815 of Titan's atmosphere and surface. However, we find that the full-disk albedo only changes $\sim 0.4\%$ 816 from 0.1911 in the first image to 0.1903 in the second image in Fig. S42.

817 We average the observations in each year of the Cassini period (2004-2017) to get yearly 818 albedo of Titan. So the temporal variations with time scales longer than one Titan day (~ 16 Earth 819 days) but shorter than one Earth year are not resolved. Panel C of Fig. S42 shows an image of 820 Titan recorded ~ 2 Earth months after the second image (panel B). But the two images have the 821 roughly same latitude/longitudinal coverage. Therefore, the comparison between panel B and 822 panel C can help us examine the temporal variations of Titan's full-disk albedo at the time scales 823 longer than one Titan day but shorter than one Earth year. Titan's full-disk albedo changes $\sim 0.7\%$ 824 from 0.1903 in panel B to 0.1890 in panel C. Therefore, Titan's heterogeneous property and 825 temporal variations at the time scales shorter than one Earth year is probably smaller than 1% and 826 we assume 1% for this uncertainty.

827 The cloud bands also develop on Titan sometimes. An example of such cloud bands is 828 shown in Fig. S43. Cloud bands are generally composed by very bright clouds aligned in the 829 longitudinal direction. Our estimates shows that the cloud bands shown in Fig. S43 increase the 830 original albedo by $\sim 20\%$. The ratio between the area of the cloud bands and the full-disk area is 831 $\sim 2\%$, so the cloud bands increase the full-disk albedo by 0.4%. It is hard to examine the whole 832 lifetime (i.e., from birth to death) of the cloud bands. But it is probable that the cloud bands have 833 lifetimes less than one half of an Earth year. So the cloud bands shown in Fig. S43 increase the 834 annual-mean albedo with an upper limit $0.4\% \times 1/2 = 0.2\%$.

Combining the uncertainties from the high-altitude haze, the error in determining Titan's effective radius, the spatio-temporal variability of Titan's atmosphere and surface, and the effects of cloud bands, we have the combined uncertainty as $\sqrt{0.002^2 + 0.007^2 + 0.01^2 + 0.002^2} = 0.012 =$ 1.3%.

839 **B.4.** Total uncertainty of Bond albedo

We combine the calibration, fitting, and other uncertainties into the total uncertainty of Titan's monochromic Bond albedo $(\mathcal{O}(A_1))$. The total uncertainty can expressed as

842 $\mathcal{O}(A_{f}) = \sqrt{\mathcal{O}_{cal}^{p}(f) + \mathcal{O}_{fit}^{p}(f) + \mathcal{O}_{other}^{p}(f)}$ (10)

where $\mathcal{O}_{cal}(I)$, $\mathcal{O}_{fit}(I)$, and $\mathcal{O}_{other}(I)$ are uncertainties related to data calibration, fitting, and 843 other error sources, respectively. Titan's Bond albedo is computed by weighting the monochromic 844 845 Bond albedos over the whole wavelength range (0-6000 nm) by the solar spectral irradiance. But 846 the Cassini ISS/VIMS observations do not cover the whole wavelength range, and there are 847 observational gaps in wavelength. First, we interpolate/extrapolate the uncertainty in these 848 wavelengths recorded by the Cassini ISS/VIMS to other wavelengths in the wavelength range of 849 0-6000 nm. Figure S43 shows the spectral distribution of the uncertainties in the measurements of 850 monochromic Bond albedo during the Cassini epoch.

851 Figure S44 suggests that the uncertainty related to filling the observational gaps in phase 852 angle is dominant in the total uncertainty. This figure also shows that the temporal variations of 853 the uncertainties are not very strong. That is because the ISS and VIMS data calibrations and the 854 observational gaps do not vary significantly with time. Figure S45 further shows the ratio between 855 the total uncertainty and the corresponding monochromic Bond albedo. First, we can see that the 856 spectral distribution of the total uncertainty is basically the same as that of the measured Bond 857 albedo, which means that the uncertainties of the large Bond albedo are also large. Second, the 858 ratio can reach 35% in some wavelengths (panel C of Fig. S45). These large ratios appear in the 859 wavelengths longer than 1000 nm. The relatively small monochromic Bond albedos at these 860 wavelengths are the main reason why there are large ratios. It should be mentioned that the large 861 ratios in these wavelengths do not significantly contribute to Titan's Bond albedo and the corresponding uncertainty because the SSI in the wavelengths longer than 1000 nm are relatively 862 863 small compared to the SSI in the short wavelengths.

864 Based on the spectral distribution of the uncertainties in the monochromatic Bond albedo 865 (Fig. S44), we can estimate the uncertainties of Titan's Bond albedo. The Bond albedo (A) is 866 computed by weighting the monochromic Bond albedos with the SSI as below (Li et al., 2018)

867
$$A = \frac{1}{SSI_{sum}} \mathop{\overset{}}_{j=0}^{I=5131nm} SSI_{j}A_{j} = \mathop{\overset{}}_{j=0}^{S131nm} c_{j}A_{j}$$
(11)

where $SSI_{/}$ and $SSI_{/}$ are the SSI at different wavelengths and the sum of SSI over wavelength, respectively. The coefficient $c_{/}$ is defined as $c_{/} = SSI_{/}/SSI_{sum}$. Eq. (11) suggests that the process of computing Bond albedo is like the sum of the monochromatic Bond albedos at different wavelengths with weighting factors. Therefore, the uncertainty of Titan's Bond albedo can be estimated from the uncertainties of the monochromatic Bond albedo by applying the rule of error propagation of addition (Bevington and Robinson, 2003) as below

874 $S^{2}(A) = \stackrel{\circ}{a}c_{I}^{2}S^{2}(A_{I})$ (12)

where $d^2(A)$ and $d^2(A_1)$ are variances of Titan's Bond albedo and monochromatic Bond albedo, respectively. Combining the spectral distribution of the monochromatic Bond albedo (Fig. S44) and Eq. (12), we have uncertainty of Titan's Bond albedo shown in Fig. S46. Compared with the uncertainties in the monochromic Bond albedo (Fig. S44), the uncertainties in Titan's Bond albedo (Fig. S46) are much smaller because the uncertainties of monochromic Bond albedo can cancel each other when they are averaged over wavelength.

881 When investigating Titan's radiant energy budget, we need to determine the emitted 882 thermal power and the absorbed solar power. Considering that the effective radius is different 883 between Titan's emitted thermal emission (Creecy et al., 2019) and absorbed solar irradiance (this 884 study), we need to compute the sphere-integrated emitted power and absorbed power. The 885 uncertainties in the sphere-integrated emitted thermal power are discussed in previous section. 886 Here, we discuss the uncertainty of disk-integrated absorbed solar power, which are related to the 887 uncertainties of disk-integrated solar flux and reflected solar power.

888 Titan's disk-integrated solar flux is computed by production of the SSI at Titan (Fig. S2) 889 and Titan's disk areas based on the effective radii (Figs. S8-S15). The solar flux at Titan is based 890 on the measured solar constant at Earth (Fig. S1), which have negligible uncertainties. So the 891 uncertainty in the disk-integrated solar flux is mainly determined by the uncertainty in the 892 measurements of effective radii. Because the uncertainty in the measurements of effective radius 893 is very small and such small uncertainty becomes even smaller when averaging over wavelength 894 (see Eq. (12)). Then the uncertainty in the disk-integrated solar flux can be used to compute the 895 uncertainty in the disk-integrated reflected solar power. The disk-integrated reflected solar power can be computed by $P_{reflect} = P_{solar} \land A$ (where P_{solar} and $P_{reflect}$ are disk-integrated solar flux and the 896 897 reflected solar power respectively). Based on the error propagation, we have the uncertainties of 898 disk-integrated reflected solar power as

899

$$\frac{\mathcal{O}^{2}\left(P_{reflect}\right)}{P_{reflect}^{2}} = \frac{\mathcal{O}^{2}\left(P_{solar}\right)}{P_{solar}^{2}} + \frac{\mathcal{O}^{2}\left(A\right)}{A^{2}}$$
(13)

where $\mathcal{O}^{2}(P_{reflect})$ and $\mathcal{O}^{2}(P_{solar})$ are variances of the disk-integrated solar flux and reflected solar power, respectively. The disk-integrated absorbed solar power (P_{absorb}) can be computed by $P_{absorb} = P_{solar} - P_{reflect}$. Then the uncertainty in the disk-integrated absorbed solar power ($\mathcal{O}(P_{absorb})$)) can expressed as

904

$$\mathcal{O}^{2}\left(P_{absorb}\right) = \mathcal{O}^{2}\left(P_{solar}\right) + \mathcal{O}^{2}\left(P_{reflect}\right) \tag{14}$$

where $d^2(P_{absorb})$ is the variance of the disk-integrated absorbed solar power. The uncertainties of the disk-integrated solar flux, reflected solar power, and absorbed solar power are shown in Fig. S47, which are furthered used in Figs. 2 and 3 in the main text.

908 C. Other uncertainties affecting the radiant energy budget of Titan

909 When investigating the global radiant energy budget of Titan, we take the atmosphere and 910 surface as a system. For such a system, there are other energy sources, which should be considered. The first one is the emitted thermal radiance from Saturn. Saturn's emitted power is ~ 4.95 Wm⁻² 911 in the Cassini epoch (Li et al., 2010). Such a power drops to ~ 0.012 Wm⁻² at the distance of Titan. 912 During the Cassini epoch, Titan's average solar constant (Fig. S2) and Bond albedo (Fig. 1 in the 913 main text) are ~ 15.04 Wm⁻² and ~ 0.26, respectively. So the absorbed solar power is ~ $15.04 \times (1-$ 914 $(0.26) = 11.13 \text{ Wm}^{-2}$. Assuming that the albedo for the Saturn's thermal radiance is the same as 915 Titan's Bond albedo (~ 0.26), we have the absorbed power from Saturn's thermal emission is ~ 916 $0.012 \times (1-0.26) = 0.009 \text{ Wm}^{-2}$, which is about 0.1% of the absorbed solar power (0.009/11.13 ~ 917 918 0.1%).

919 There are two other energy sources: (1) internal energy released from the surface (Sohl et al., 1995; Tobie et al., 2006); (2) tidal heat (Tobie et al., 2006). Both have values of order 10^{-3} 920 Wm⁻², which is comparable to the thermal radiant power from Saturn's emission. The two powers 921 are also comparable to the uncertainty in the measurements of Titan's emitted thermal power (~ 922 0.006 Wm⁻²) (see Fig. S37) but are much smaller than the uncertainty in the measurements of 923 924 Titan's absorbed solar power. The measurements of Titan's Bond albedo and hence the absorbed 925 solar power have large uncertainty because there are no observations of reflected solar irradiance at the highest phase angles. Based on the uncertainty of Titan's Bond albedo (~ 0.0026, see Fig. 926 S45) and the solar constant at Titan ($\sim 15.04 \text{ Wm}^{-2}$, see Fig. S2), we have the uncertainty in the 927 measurements of Titan's absorbed solar power as $15.04 \times 0.0026 = 0.039$ Wm⁻². Such an 928 929 uncertainty is much larger the uncertainties from internal energy and tidal heat. Therefore, the 930 powers from Saturn's thermal emission, internal energy, and tidal heat are not considered in our 931 discussion of the global radiant energy budget of Titan.

932 In summary, the uncertainty related to filling observational gaps is dominant in the 933 measurements of Titan's emitted power (see Figs. S36 and S37). For the uncertainty of the 934 measurements of Titan's Bond albedo and hence absorbed solar power, the large observational 935 gaps at the highest phase angles ($\sim 165-180^{\circ}$) significantly contribute to the uncertainty (see Fig. S44). The large observational gaps in measuring Titan's Bond albedo also make the uncertainty is 936 much larger in the measurements of Titan's absorbed solar power (~ 0.039 Wm^{-2}) than in the 937 measurements of Titan's emitted thermal power (~ 0.006 Wm^{-2}), as we discussed above. 938 939 Considering that the emitted power goes off from the whole sphere and the absorbed solar power 940 acts on the cross section of Titan, we have that uncertainties of the total power are not that different between the disk-integrated absorbed power (~ 10×10^{11} W, see Fig. S47) and the sphere-integrated 941 emitted power (~ 7×10^{11} W, see Fig. S38). 942

943 Extending the Cassini analysis to a complete Titan's year

944 The Cassini epoch (2004-2017) occupies slightly less than one half of Titan's orbital period 945 around the Sun (i.e., a Titan year ~ 29.4 years). In order to examine Titan's radiant energy budget 946 during a complete Titan year, we investigate the possible seasonal cycles of the absorbed solar 947 power and the emitted power. The temporal variations of the absorbed solar power follow the 948 seasonal cycle of the solar irradiance (Fig. 2). Earth's global emitted power (Jacobowitz et al., 949 1979; Yang et al., 1999) clearly displays a seasonal cycle, so we assume that Titan's emitted power 950 also has a seasonal cycle (Creecy et al., 2019). We use a sine function with a period of 29.4 years 951 to fit the observed absorbed power and emitted power (Fig. 3) and estimate their seasonal cycles,

which are displayed in Fig. S48. The uncertainty of fitting is estimated by the fitting residuals andextrapolating the fitting residuals from the Cassini epoch to a complete Titan year.

Integrating the fitted functions over a complete Titan year (Fig. 3), we find that the total absorbed solar energy, $(2.676\pm0.005)\times10^{23}$ J, and the total emitted thermal energy, $(2.549\pm0.053)\times10^{23}$ J, have an even bigger energy imbalance of $(0.127\pm0.053)\times10^{23}$ J for the complete Titan year. Such an energy imbalance is $5.0\pm2.1\%$ of the total emitted energy for a complete Titan year. The extrapolated energy imbalance over a complete Titan year is larger than the energy imbalance during the Cassini epoch because the relatively large energy imbalance mainly happened in the time period before the Cassini epoch (see Fig. S48).

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Figure S1. The Solar Spectral Irradiance (SSI) at Earth from 2004 to 2017. (A) Earth's SSI. (B) Earth's solar power (i.e., solar constant). The solar power is computed by integrating the SSI over wavelength. The time-varying SSI from 2004 to 2017 in the wavelength range 0-200 nm and 200-2400 nm comes from the Solar EUV Experiment (SEE) and the Solar Radiation and Climate Experiment (SORCE), respectively. The climatological SSI in the wavelength range 2400-4000 nm comes from American Society for Testing and Materials (ASTM). The SSI in the range 4000-6000 nm is computed by assuming the blackbody spectra with a temperature 5778 K (which matches best the observed SSI over the wavelength range 0-4000 nm), in which the temporal variations of SSI are not considered (see Table S1 for more details).



Figure S2. The SSI, Sun-Titan distance, and solar flux at Titan from 2004 to 2017. (A) The SSI at the distance of Titan. (B) The distance between the Sun and Titan. (C) The solar flux at the distance of Titan. The SSI at the distance of Titan (panel A) is computed by dividing the SSI at Earth (panel A of Fig. S1) by the square of the distance between the Sun and Titan (panel B). The solar flux at 1150 Titan is computed by integrating the SSI (panel A) over wavelength.



Figure S3. Titan's albedo spectra (305-1050 nm) recorded by the European Southern Observatory
(ESO). The two spectra were recorded in 1993 and 1995 with phase angles 2.7° and 5.7°
respectively. Note that the spectra generated by Karkoschka were derived with the solid radius of
Titan ~ 2575 km (see section "Supplementary Observations and Data" in Materials and Methods).
Titan's wavelength-dependent effective radii (see section "Effective Radii of Titan's
Atmosphere") are considered in the spectra.



Figure S4. Titan's albedo spectra (150-305 nm) from the Cassini UVIS and the HST/FOS. The geometric-albedo spectra in the wavelength range 150-190 nm come from the Cassini UVIS and the geometric-albedo spectra in the wavelength range 190-305 nm come from the Faint Object Spectrograph (FOS) of the Hubble Space Telescope (HST) (also see Supplementary Information Table S1). Note that Titan's wavelength-dependent effective radii (see section "Effective Radii of Titan's Atmosphere") are considered in the spectra.





Figure S5. Phase function of Titan's albedo from Pioneer 11 observations. The variations of Titan's albedo as a function of phase angle at blue (452 nm) and red (648 nm) wavelengths come from a previous study based on the observations recorded by the Imaging Photopolarimeter (IPP) on the Pioneer 11 spacecraft. Note that Titan's wavelength-dependent effective radii (see section "Effective Radii of Titan's Atmosphere") are considered. Vertical lines represent error-bars of measurements.





Figure S6. An example of the ISS raw and calibrated images. (A) The ISS raw image. (B) The
calibrated image with a unit of radiance. The raw image was taken by the ISS CB3 filter on August
27, 2009 with a phase angle ~ 0.75° and a spatial resolution ~ 8.6 km/pixel. The ISS raw images
are calibrated by the Cassini ISS CALibration (CISSCAL) software (see section "Cassini
ISS/VIMS Data and Data Processing").





Figure S7. Examples of the VIMS calibrated images. (A) Full-disk image. (B) Day-side image.
(C) Night-side image. The corresponding VIMS raw image was taken by the VIMS on July 1,
2006 with a phase angle ~ 60.7° and a spatial resolution ~ 135.5 km/pixel. The VIMS took images
from ~ 350 nm to ~ 5131 nm. The example shown here has a wavelength ~ 2000 nm.



Figure S8. Test of the effective radius of Enceladus. (A) A calibrated image of Enceladus. The corresponding raw image in panel A was taken by the Cassini ISS at RED filter with a phase angle of $\sim 1.36^{\circ}$ and a spatial resolution ~ 2.58 km/pixel. The two horizontal solid white lines in panel A, which are located in the left and right boundaries of Enceladus respectively, are used to conduct the analyses in panels B and C. The two vertical dashed white lines in panel A show the locations of the two boundaries with Enceladus' optical disk, which are determined by the analyses in panels B and C. (B) The calibrated radiance along the two horizontal solid white lines shown in panel A. (C) The gradient of radiance along the horizontal direction for the two boundary lines shown in panel B. The locations of the vertical dashed white lines in panel A, which suggest the left and right boundaries of Enceladus's optical disk, are determined by the pixel locations with the maximal gradient of the line crossing the left boundary and the minimal gradient of the line crossing the right boundary, respectively. The product between the pixel number between the two vertical dashed lines and the spatial resolution is used to estimate the effective diameter, and half of the effective diameter is the effective radius (also see section "Effective Radii of Titan's Atmosphere").



Figure S9. Titan's effective radius at the RED wavelength with a high phase angle. This figure is the same as Fig. S8 except for the image of Titan. The raw image corresponding to the calibrated image in panel A was taken by the RED filter of the Cassini ISS on April 13, 2013 with a phase angle ~4.26° and a spatial resolution ~ 10.8 km/pixel.



Figure S10. Titan's effective radius at the RED wavelength with a high phase angle. This figure is the same as Fig. S9 except that the image was taken at a high phase angle. The raw image corresponding to the calibrated image in panel A was taken by the RED filter of the Cassini ISS on June 29, 2007 with a phase angle ~166.56° and a spatial resolution ~ 12.4 km/pixel.


Figure S11. Temporal variations of Titan's effective radius at the CB3 wavelength. Panels A-C are the ISS calibrated images recorded by the CB3 filter in three different years. (A) A calibrated image in 2004. The corresponding raw image was taken by the CB3 filter of the Cassini ISS on December 13, 2004 with a phase angle ~ 161.21° and a spatial resolution ~ 21.8 km/pixel. (B) A calibrated image in 2009. The corresponding raw image was taken by the CB3 filter on July 22, 2009 with a phase angle ~ 161.30° and a spatial resolution ~ 9.7 km/pixel. (C) A calibrated image in 2016. The corresponding raw image was taken by the CB3 filter on November 10, 2016 with a phase angle ~ 164.06° and a spatial resolution ~ 10.6 km/pixel. (D) Titan's effective radius at the CB3 wavelength in different years. Please see the caption of Fig. S8 for explanations of the solid and dashed white lines in panels A-C. Vertical lines in panel D represent error-bars of measurements.



Figure S12. Titan's effective radii measured by the Cassini ISS 12 filters. (A) Effective radii at the wavelengths of the ISS 12 filters with low and high phase angles. Note: the CB1 is composed by two sub-filters (CB1a and CB1b). It is hard to differentiate the CB1 observations between CB1a and CB1b. We use the CB1 observations to get the effective radii first. Then we use the slope of effective radii at the neighboring filters (GRN and RED) to interpolate the CB1 results to the wavelengths of CB1a and CB1b. (B) Averaged effective radii at the wavelengths of the ISS 12 filters. We average over the analyses at the low and high phase angles shown in panel A to get the average radii. Vertical lines in the two panels represent the error-bars of the measurements (see section "Effective Radii of Titan's Atmosphere").





 $\begin{array}{c} 1432\\ 1433 \end{array}$

1434 Figure S13. Comparison of Titan's effective radii between the Cassini ISS measurements and 1435 other studies. (A) Comparison among the Cassini results, the Pioneer results, and the results from 1436 a model study. (B) The ratio of the difference between the Cassini results and other results over the Cassini results. Please note the wavelengths of the red (640 nm) and blue (440 nm) filters from 1437 1438 the Pioneer observations are slightly different from the wavelengths of the RED and BL1 filters 1439 from the Cassini observations. We linearly interpolate the Cassini results to the Pioneer red and blue wavelengths and then compute the difference between the Cassini results and the Pioneer 1440 1441 results (please see section "Effective Radii of Titan's Atmosphere" for more details and 1442 references).

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Figure S14. Titan's effective radii measured by the Cassini VIMS observations. The measurements are based on four solar-occultation observations. The four measurements are averaged to get the mean effective radii in the infrared wavelengths of the VIMS instrument (thick black line). The vertical dashed lines represent the uncertainties of the effective radii (see section "Effective Radii of Titan's Atmosphere").





1475 Figure S15. Comparison of effective radii between the Cassini ISS and VIMS measurements. The 1476 vertical solid and dashed lines are for error-bars of the measurements of ISS and VIMS, 1477 respectively. As we discussed in text, the solar-occultation observations were conducted in the 1478 infrared part of the VIMS instrument (840-5000 nm). The VIMS infrared wavelengths are 1479 overlapped with two ISS filters/wavelengths only (i.e., MT3-889 nm and CB3-938 nm).





Figure S16. Comparison of Titan's full-disk albedo between the Cassini ISS and VIMS observations. Among the ISS 12 filters, the three violet filters (UV1 ~ 264 nm, UV2 ~ 306 nm, and UV3 ~ 343 nm) are out of the wavelength range of the VIMS (350-5131 nm). The wavelength of the UV3 filter (343 nm) is close to the low limit of the VIMS wavelength (350 nm), so we keep the ISS UV3 results and compare them with the VIMS measurements at 350 nm (panel A).





Figure S17. Comparison of Titan's full-disk albedo between the Cassini measurements (ISS and VIMS) and the Pioneer measurements. The Pioneer measurements at the blue (440 nm) and red (640 nm) wavelengths were conducted by the IPP on Pioneer 11 (please see Fig. S5 for more details). The VIMS measurements at 440 nm and 640 nm are plotted for comparison. The measurements based on the ISS BL1 (459 nm) and RED (649 nm) filters, which are close to the Pioneer blue and red wavelengths, are also plotted for comparison (see section "Validation of Cassini ISS and VIMS Results").



Figure S18. Comparison of Titan's full-disk albedo between the Cassini ISS observations and Earth-based observations. The ground-based ESO measurements in 1995 are used (see Fig. S3). Please note that the ESO measurements are based on the observations over Titan's disk with a solid radius (Fig. S3). We scale the results by the ratio between the ISS measurements with the solid radius and the ISS measurements with the effective radii. The ESO observations were conducted in the effective wavelength range 305-1050 nm with a phase angle ~ 5.7° , so we use the ISS observations with a phase angle 5.7° for comparison. The ISS UV1 (264 nm) filter is outside of the ESO wavelength range (305-1050 nm). So we use the observations from the HST/FOS (also see Fig. S4). The full-disk albedo from the HST was corrected to zero phase angle, so we use the ISS UV1 measurements at zero phase angle too.





Figure S19. Comparison of Titan's full-disk albedo between the Cassini VIMS observations and Earth-based observations. Same as Fig. S18 except for the Cassini VIMS observations are compared with the Earth-based ESO observations in 1995 (40). Note the VIMS observations and the ESO observations have different phase angles (11.7° for VIMS and 5.7° for ESO) and spectral resolutions (~ 4-25 nm for VIMS and ~ 0.4 nm for ESO) (see section "Validation of Cassini ISS and VIMS Results" for more details).





Figure S20. Titan's full-disk albedo from the observations recorded by the Cassini ISS 12 filters during the Cassini period of 2004-2017. (A) BL1 filter (459 nm); (B) GRN filter (569 nm); (C) RED filter (648 nm); (D) UV1 filter (264 nm); (E) UV2 filter (306 nm); (F) UV3 filter (343 nm); (G) CB1 fitler (CB1a ~ 635 nm and CB1b ~ 603 nm); (H) CB2 filter (751 nm); (I) CB3 fitler (939 nm); (J) MT1 fitler (619 nm); (K) MT2 filter (728 nm); (L) MT3 fitler (890 nm). The full-disk albedo functioning as phase angle is displayed in each year from 2004-05 to 2017. There are only three-month (October-December) high-quality observations in 2004, so the 2004 observations are combined with the 2005 observations. The blank areas are observational gaps.

- 0.28 **ISS data** polynomial-function fitting 0.25 double Henyey-Greenstein fitting 0.22 full-disk albedo 0.19 0.16 0.13 0.1 0.07 phase angle (degree)

Figure S21. An example of fitting the phase function of Titan's full-disk albedo recorded by the
Cassini ISS observations. The raw ISS data were recorded by the CB3 filter of the Cassini/ISS in
2009. A six-order polynomial function (red line) and the double H-G function (blue line) are used
for fitting the ISS data (see section "Filling Observational Gaps in Phase Angle" for more details).



Figure S22. Fitting the phase functions of Titan's full-disk albedo recorded by the Cassini ISS 12 filters. (A) BL1 filter (459 nm); (B) GRN filter (569 nm); (C) RED filter (648 nm); (D) UV1 filter (264 nm); (E) UV2 filter (306 nm); (F) UV3 filter (343 nm); (G) CB1 filter (CB1a ~ 635 nm and CB1b = (022 nm); (II) CB2 filter (751 nm)) (II) CB2 filter (020 nm); (II) MT1 filter (C10 nm)) (IV)

1654 CB1b ~ 603 nm); (H) CB2 filter (751 nm); (I) CB3 fitler (939 nm); (J) MT1 fitler (619 nm); (K)
1655 MT2 filter (728 nm); (L) MT3 fitler (890 nm).



Figure S23. Ratios between fitting residuals and observed albedos for the ISS 12 filters. The fitting residual is defined as the difference between fitted albedo (Fig. S22) and the observed albedo (Fig. S20). (A) BL1 filter (459 nm); (B) GRN filter (569 nm); (C) RED filter (648 nm); (D) UV1 filter (264 nm); (E) UV2 filter (306 nm); (F) UV3 filter (343 nm); (G) CB1 filter (CB1a ~ 635 nm and CB1b ~ 603 nm); (H) CB2 filter (751 nm); (I) CB3 filter (939 nm); (J) MT1 filter (619 nm); (K) MT2 filter (728 nm); (L) MT3 filter (890 nm).







Figure S24. Titan's full-disk albedo observed by the Cassini VIMS. The times (spatial resolutions) for the VIMS observations with phase angles 11.7°, 18.1°, 32.6°, 49.9°, 60.7°, 74.4°, 89.2°, 115.8°, and 144.7° are 2007 (206 km/pixel), 2006 (191 km/pixel), 2007 (147 km/pixel), 2007 (178 km/pixel), 2006 (139 km/pixel), 2008 (147 km/pixel), 2008 (161 km/pixel), 2006 (206 km/pixel), and 2007 (129 km/pixel), respectively. The largest phase angles of the high-quality VIMS observations are 159.1° and 156.1° for the visible part (~350-1046 nm) and the infrared part (~1046-5131 nm), respectively. The observations with the phase angle 159.1° (visible part) were recorded in 2007 with a spatial resolution ~ 151 km/pixel. The observations with the phase angle 156.1° (infrared part) were recorded in 2012 with a spatial resolution ~ 126 km/pixel. We combine the visible spectra at 159.1° and the infrared spectra at 156.1° together (red line).



Figure S25. Fitting the phase function of Titan's full-disk albedo by the VIMS observations (3505131 nm). The VIMS observational gaps with phase angles smaller than 11.7° and larger than
159.1° are filled by the ISS data before the least-squares fitting (see section "Filling Observational
Gaps in Phase Angle").



Figure S26. Ratio between fitting residual and observed albedo for the VIMS observations. The
fitting residual is defined as the difference between fitted albedo (Fig. S25) and the observed
albedo (Fig. S24). Please note that the phase angle 159.1° in y axis is for the VIMS visible part
(350-1046 nm) only. The VIMS infrared part (1046-5131 nm) has a phase angle 156.1° (see Fig.
S24).



Figure S27. ISS-derived albedo in the two-dimensional domain of phase angle (0-180°) and wavelength (264-939 nm) during the Cassini epoch. The derived albedo is based on the ISS fitted phase functions in Fig. S22 and reference albedo spectra with a high spectral resolution shown in Fig. S3 (40) (see section "Filling Observational Gaps in Wavelength and Time"). Titan's twodimensional albedo in each year of the Cassini period of 2004-2017 is derived. Here, only four years (2004-05, 2009, 2013, and 2017) are shown as examples.



Figure S28. Comparison of Titan's albedo between the ISS derived results and the VIMS fitted results for the overlap wavelengths (~ 350-939 nm). (A) The ISS derived albedo. Panel A shows the time-mean albedo, which is averaged over the Cassini epoch (2004-2017) (see Fig. S27). (B) The VIMS fitted albedo. The VIMS results come from Fig. S25.



Figure S29. Difference of Titan's albedo between the ISS derived results and the VIMS fitted results. (A) Difference of Titan's albedo between the ISS results (panel A of Fig. S28) and the VIMS results (panel B of Fig. S28). (B) Difference ratio. The difference ratio is defined as the ratio between the difference (panel A) and the mean value.



Figure S30. Titan's albedo in the complete wavelength range 0-5131 nm during the Cassini epoch. The time-varying albedo in the wavelength range 264-939 nm comes from Fig. S27. The time-varying albedo in the wavelength range 939-5131 nm (outside of the ISS wavelength range but in the VIMS wavelength range) is based on the VIMS fitted results (Fig. S25) and assume the VIMS results have the same temporal variations of those of the ISS CB3 results at 939 nm. The time-varying albedo in the wavelength range 0-264 nm (uncovered by the Cassini ISS & VIMS observations) is based on the albedo spectra from Cassini/UVIS (150-190 nm) the HST/FOS (190-264 nm) and assume that the albedo spectra in the wavelength range 0-150 nm has the same albedo as that at 150 nm. In addition, the phase functions and the corresponding temporal variations from the ISS UV1 (264 nm) observations are used to approximate the phase functions and their temporal variations in the wavelength range 0-264 nm (see section "Filling Observational Gaps in Wavelength and Time").







Figure S31. Titan's monochromatic geometric albedo, phase integral, and Bond albedo in the wavelength range 0-5131 nm during the Cassini epoch. Panels (A) and (B) are monochromatic geometric albedo and anomaly (i.e., difference from time-mean), respectively. Panels (C) and (D) are monochromatic phase integral and anomaly, respectively. Panels (E) and (F) are monochromatic Bond albedo and anomaly, respectively.







Figure S32. Comparison between typical spectra of Titan (solid line) and typical spectra of deep
 space (dashed line). Both spectra of Titan and deep space were recorded by the three focal planes
 of the Cassini/CIRS in 2011 with a spectral resolution 5 cm⁻¹.



1932 Figure S33. Spectra of deep space in different years. For each year, 100 spectra of deep space1933 were plotted.





Figure S34. Comparison between the spectral noise related to the temporal control of the CIRS instrument and the spectra of deep space. (A) Spectra of the designed temperature of the CIRS instrument (170 K) and the possible temperature of the CIRS instrument with the temperature control (170.2 K). (B) Difference of the two spectra shown in panel A, which is defined as the spectra noise related to the temporal control of the CIRS instrument. (C) Comparison between spectral noise and typical spectra of deep space.



Figure S35. Examples of fitting CIRS data. (A) Raw data. (B) Least squares fitting. (C) Fitting
residual. Fitting residual is defined as the difference between the fitted values and raw data.



Figure S36. Examples of uncertainties in the measurements of Titan's emitted power. The uncertainty analysis for the CIRS measurements of Titan's emitted power in 2009 is shown. (A) Uncertainties. (B) Titan's emitted power. (C) Ratio between uncertainties (panel A) and the corresponding emitted power (panel B).



Figure S37. Uncertainties in the measurements of Titan's emitted power during the Cassini epoch.
(A) The absolute uncertainties in the measurements of Titan's emitted power. (B) The ratio
between the uncertainties (panel A) and Titan's emitted power.





Figure S38. Uncertainties in the measurements of Titan's sphere-integrated emitted power during the Cassini epoch. The uncertainties in the emitted power over a unit area (Fig. S37) are integrated over a sphere with a effective radius 2757+500 km (where 2757 is the radius of solid Titan and 500 km is the effective height to capture all important thermal emission sources).

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Figure S39. Example of ISS calibrated images with a unit of radiance. The raw image was taken
by the ISS CB3 filter on December 11, 2004 with a phase angle ~ 18.15° and a spatial resolution
~ 6.5 km/pixel. The ISS raw images are calibrated by the latest version of the Cassini ISS
calibration software.

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Figure S40. Examples of ISS data fitting and fitting residual. (A) Data fitting shown in Fig. S21.
(B) Fitting residuals, which are defined as the difference between the fitting values and raw data.
(C) Ratios between fitting residuals and raw data.





Figure S41. High-altitude haze shown in some filters of the Cassini ISS images. The raw images recorded by the ISS MT3 (panel A), RED (panel C), and UV1 (panel D) filters were taken on April 13, 2013 with a spatial resolution ~ 10.77 km/pixel and a phase angle ~ 4.27°. The image recorded by CB3 filter (panel B) was taken on August 27, 2009 with a spatial resolution ~ 8.62 km/pixel and a phase angle ~ 0.75° . The high-altitude haze is pointed by arrows in panels A and C.





radiance (10¹¹ photons/s/cm²/nm/ster)

- Figure S42. Titan's reflectivity and time variations seen in ISS images at different longitudes and
 times. All three images in panels A, B, and C were recorded by the ISS CB3 filter with the same
 phase angle ~ 14°. The spatial resolutions for the three images are 8.84 km/pixel, 17.56 km/pixel,
- and 18.64 km/pixel, respectively.



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Figure S43. An example of cloud bands on Titan. The raw image was taken by the ISS CB3 filter on May 6, 2017 with a phase angle ~ 14.3° and a spatial resolution ~ 3.1 km/pixel. The ISS raw images are calibrated by CISSCAL software.



Figure S44. Spectral distribution of the uncertainties in the measurements of monochromic Bond albedo during the Cassini epoch (2004-2017). (A) Uncertainty related to the Cassini data calibration. (B) Uncertainty related to filling observational gaps in phase angle. (C) Uncertainty from other error sources. (D) Total uncertainty by combining the uncertainties in panels A, B, and C.







Figure S45. Ratio between the total uncertainty and the corresponding monochromatic Bond
albedo during the Cassini epoch (2004-2017). (A) Total uncertainty. (B) Bond albedo. (C) Ratio
between the total uncertainty (panel A) and the corresponding monochromatic Bond albedo (panel
B). Panels A and B are the same as panel D of Fig. S43 and panel E of Fig. S31, respectively.



Figure S46. Uncertainties in the measurements of Titan's Bond albedo during the Cassini epoch.
(A) Different uncertainties. (B) Uncertainty ratio. The uncertainty ratio is defined as the ratio
between the uncertainties shown in panel A and Titan's Bond albedo (Fig. 1 in main text).


Figure S47. Uncertainties in the disk-integrated solar flux, reflected solar power, and absorbed solar power.





Figure S48. Fitted Titan's absorbed power and emitted power. The observed absorbed power and emitted power are fitted by sine functions with a fixed period – one Titan year (29.424 years). The red and blue horizontal lines represent the annual-mean absorbed power and emitted power, respectively.

Table S1. Observational Data Sets For Measuring Titan's Bond Albedo. The numbers in the parentheses are observational wavelengths and times. The full names of the abbreviations in the table are introduced as below. The ISS, VIMS, and UVIS are three instruments on the Cassini spacecraft. They are Imaging Science Sub-system (ISS), the Visual and Infrared Mapping Spectrometers (VIMS), and Ultraviolet Imaging Spectrograph (UVIS), respectively. The SEE, SORCE, and ASTM are three data sets for the solar spectral irradiance (SSI). They are the Solar EUV Experiment (SEE), the Solar Radiation and Climate Experiment (SORCE), and the American Society for Testing and Materials (ASTM), respectively. The Pioneer 11/IPP represents the Imaging Photopolarimeter (IPP) on the spacecraft Pioneer 11, whose observations were used to measure the phase function of Titan's albedo in a previous study. The HST/FOS and ESO stand for the Faint Object Spectrograph (FOS) of the Hubble Space Telescope (HST) and the European Southern Observatory (ESO), respectively. Their observations also provide the albedo spectra of Titan (see sections "Summary of Observational Data Sets" and "Supplementary Observations and

- 2346 Data" for more details and references).

Variable	Cassini Observations	Other Observations
solar spectral		SEE (0-200nm, 2004-2017),
irradiance (SSI)		SORCE (200-2400 nm, 2004-2017),
		ASTM (2400-4000 nm,
		climatology), and blackbody
		spectrum (4000-6000 nm)
phase function	ISS (264-939 nm, 2004-2017) and	Pioneer 11/IPP (452 nm (blue) and
-	VIMS (350-5131 nm, 2004-2017)	648 nm (red), 1979).
spectral	ISS (264-939 nm, 2004-2017),	HST/FOS (190-305 nm, 1991 and
observations	VIMS (350-5131 nm, 2004-2017),	1992) and ESO (305-1050 nm, 1993
	and UVIS (150-190 nm, 2004).	and 1995)