

1 Apparent and Real Absorption of Solar Spectral Irradiance
2 in Heterogeneous Ice Clouds

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21 Abstract:
22 Coordinated flight legs of two aircraft above and below extended cirrus cloud scenes
23 played an important part in the Tropical Composition, Cloud and Climate Coupling (TC⁴)
24 Experiment (Costa Rica, 2007). The Solar Spectral Flux Radiometer (SSFR) measured
25 up- and downward irradiance on the high-altitude (ER-2) and the low-altitude (DC-8)
26 aircraft, which allowed deriving apparent absorption on a point-by-point basis along the
27 flight track. Apparent absorption is the vertical divergence of irradiance, calculated by the
28 difference of net flux at the top and bottom of a cloud. While this is the only practical
29 method of deriving absorption from aircraft radiation measurements, it differs from true
30 absorption when horizontal flux divergence is non-zero. Differences between true and
31 apparent absorption are inevitable in any inhomogeneous atmospheres, in particular
32 clouds. We show, for the first time, the spectral shape of measured apparent absorption
33 and compare with results from a three-dimensional radiative transfer model. The model
34 cloud field is created from optical thickness and effective crystal radius retrievals from
35 the MODIS (Moderate Resolution Imaging Spectroradiometer) Airborne Simulator
36 (MAS), and from reflectivity profiles from the Cloud Radar System (CRS), both onboard
37 the ER-2. We find correlations between apparent absorption and cloud optical thickness,
38 especially in the visible spectral range. They bring to bear a net horizontal photon
39 transport from local maxima to minima of optical thickness within a maximum horizontal
40 scale. Although the spectral shape is reproduced by the model calculations, the domain-
41 averaged apparent absorption in the visible spectral range is considerably higher than the
42 model results. This is possibly due to a net loss of photons into neighboring cirrus-free
43 areas that are not contained within the boundaries of the model domain.

44

45 1. Introduction

46

47 The issue of real versus apparent absorption of solar radiation within clouds was
48 discussed for decades after *Fritz and MacDonald* [1951] discovered cloud absorption
49 derived from measurements may far exceed that from model calculations. Despite its
50 significance for atmospheric energy budget assessments, cloud dynamics, and remote-
51 sensing, a conclusive explanation for this persistent bias is still lacking, and it re-emerges
52 regularly in literature. At best, a status quo was achieved where some authors argued that
53 the problem is ill-posed because the measurement (or model) errors are too large for a
54 final assessment of the bias, while others found model-measurement agreement within
55 the given uncertainties. *Stephens and Tsay* [1990] reviewed the observational evidence
56 for various manifestations of the effect and summarized explanations for the
57 discrepancies. Thereafter, a controversial discussion was initiated by new observations
58 [*Cess et al.*, 1995; *Ramanathan et al.*, 1995; *Pilewskie and Valero*, 1995]. Based on a
59 range of conditions, follow-up studies either rejected [*Hayasaka et al.*, 1995; *Arking*,
60 1996; *Stephens*, 1996; *Taylor et al.*, 1996; *Francis et al.*, 1997; *Ackerman et al.*, 2003] or
61 supported [*Pilewskie and Valero* 1996, *Valero et al.*, 1997; *Zhang et al.*, 1997; *Valero et*
62 *al.*, 2000; *O'Hirok et al.*, 2000; *O'Hirok and Gautier*, 2003] the existence of a bias. Many
63 studies favored horizontal photon transport in heterogeneous clouds as cause for the
64 discrepancies [*Newiger and Baehnke*, 1981; *Ackerman and Cox*, 1981; *Rawlins*, 1989;
65 *Titov*, 1998; *Marshak et al.*, 1997, 1998 and 1999] although other explanations were
66 suggested such as enhanced in-cloud water vapor absorption [e.g., *Francis et al.*, 1997;
67 *Arking*, 1999], large drop contributions [*Wiscombe et al.*, 1984; *Ackerman and Stevens*,

68 1987; *Knyazikhin et al.*, 2002], or in-cloud aerosols [*Newiger and Baehnke*, 1981; *Chylek*
69 *et al.*, 1984 and 1996, *Wendisch and Keil*, 1999].

70

71 Certainly, discrepancies may be caused by a combination of multiple effects, all of which
72 can be ascribed to inappropriate model assumptions or insufficient observations. When
73 attributing shortwave absorption biases to individual causes, adequate spectral resolution
74 in models and measurements to separate the roles of gas and condensed species is highly
75 valuable. For example, absorption by water vapor, aerosol, liquid water or ice can be
76 distinguished by spectral signatures, whether or not the absorption is enhanced due to
77 photon path lengthening in heterogeneous clouds. Most experiments, especially before
78 the late 1990s, did not make use of spectrally resolved measurements. Instruments with
79 full spectral coverage over the solar wavelength range were introduced by *Pilewskie et al.*
80 [2003] and *Wendisch et al.* [2001] for use in aircraft experiments. Despite this important
81 advancement in measurement technology, aspects of the absorption bias problem
82 lingered, for a number of reasons: In typical experiments, absorbed irradiance is derived
83 by differencing net irradiance (difference of downward and upward irradiance) at cloud
84 base and cloud top. This introduces large systematic errors in estimates of cloud
85 absorption because it is the (small) difference of four large quantities. If the
86 measurements above and below clouds are not coordinated in time and space, cloud
87 heterogeneities add further uncertainty. Even if they are coordinated, the measurements
88 may be affected by a net transport of photons through the sides of the sampling volume
89 (net horizontal photon outflux or influx). Horizontal irradiance divergence (convergence)
90 is balanced by the vertical flux divergence which can be misinterpreted as true

91 absorption. We refer to vertical flux divergence as apparent absorption, different from
92 true absorption by the magnitude of horizontal flux divergence.

93

94 Horizontal photon transport can be understood in the context of radiative smoothing.
95 Over some scale, contrasts in cloud optical thickness are smoothed out in the
96 corresponding reflectance and transmittance fields [*Marshak et al.*, 1995]. For cloud
97 fields with shadow effects (i.e. clouds with pronounced vertical structure) a roughening
98 can occur as well [*Marshak et al.*, 2006]. For the case of smoothing, the horizontal
99 displacement of a photon relative to its entrance into a cloud field is determined by the
100 number of scatterings which it undergoes. The photon transport is always directed from
101 higher to lower photon density. *Platnick* [2001] shows that this characteristic distance is a
102 function of wavelength (that is, single scattering albedo and asymmetry parameter). In
103 absence of shadows and sources, the horizontal re-distribution of photons in a smoothing
104 process as seen from space (reflectance) can be viewed as transport from optically thick
105 to optically thin regions within the characteristic smoothing scale. The photon path length
106 distribution associated with these processes can be fundamentally different for optically
107 thick regions (diffusion regime) and thin, sparsely populated areas [*Davis and Marshak*,
108 2000].

109

110 Various methods were proposed to correct for horizontal flux divergence in aircraft
111 measurements of absorption. *Ackerman and Cox* [1981] introduced a technique for
112 sampling radiation with a combination of broadband and filter radiometers. Absorption
113 measurements were corrected under the assumption that clouds do not absorb in the

114 visible wavelength range and that radiative smoothing affects non-absorbing and
115 absorbing wavelengths equally. *Marshak et al.* [1999] suggested various correction
116 schemes. One of these explicitly takes into account a pre-determined radiative smoothing
117 scale. Although this improves the Ackerman and Cox method considerably, it does not
118 entirely reproduce the true absorption. *Titov* [1998] (among others) suggested grand-
119 averaging of cloud absorption measurements and provided minimum domain sizes based
120 on typical boundary layer clouds.

121

122 In this paper, we pursue a different strategy. Since true absorption is difficult to derive
123 from measurements, we focus on apparent absorption, as obtained from two-aircraft
124 observations, and we reproduce measured apparent spectral absorption (vertical flux
125 divergence) on a pixel-by-pixel basis with 3D radiative transfer (RT) calculations. This
126 strategy is akin to that used by *O'Hirok and Gautier* [2003] who employed ground-based
127 cloud observations as input to 3D RT calculations. We used airborne measurements from
128 the NASA TC⁴ experiment (Tropical Composition Cloud and Climate Coupling, Costa
129 Rica, 2007; *Toon et al.*, 2009). An extensive set of instruments was deployed onboard
130 two aircraft, the NASA ER-2 and DC-8. The Solar Spectral Flux Radiometer (SSFR:
131 *Pilewskie et al.*, 2003) was flown on both platforms and measured spectrally-resolved
132 upward and downward solar irradiance. The ER-2 carried the MODIS (Moderate
133 Resolution Imaging Spectroradiometer) airborne simulator (MAS: *King et al.*, 2004), the
134 cloud radar system (CRS: *Li et al.*, 2004), and other remote-sensing instruments. It was
135 operated at 20 km altitude – well above cloud top level. The DC-8 was flown within and
136 below cloud layers and was equipped with instrumentation for cloud microphysical,

137 aerosol particle, and gas-phase measurements. On seven days, the ER-2 and DC-8 were
138 closely coordinated (in space and time) along several flight legs (typically about a 1/2
139 hour duration per leg) that were chosen in outflow regions near tropical cloud convective
140 cells. In this way, detailed cloud structure data were acquired along with simultaneous
141 above- and below-cloud measurements of solar spectral irradiance. Measurements of
142 cloud-reflected radiance were used for the retrieval of cloud thickness and particle size.

143

144 We determined point-by-point apparent spectral absorption for one case and compared
145 with model results. The calculated irradiance fields were obtained from 3D RT
146 calculations, using measurements from MAS and CRS to derive the input cloud field.
147 This paper is the third in a series of three radiation-related publications within the TC⁴
148 special issue (I: *Kindel et al.*: "Observations and modeling of cirrus shortwave spectral
149 albedo during the Tropical Composition, Cloud and Climate Coupling Experiment", II:
150 *Eichler et al.*: "Cirrus spatial heterogeneity and ice crystal shape: Effects on remote
151 sensing of cirrus optical thickness and effective crystal radius"). One aspect of part I is
152 the impact of cloud heterogeneities on radiance-irradiance conversion through cloud
153 retrievals. Part II focuses on the combined effect of scattering phase function and three-
154 dimensional cloud structure on remote-sensing products. This paper (part III) is dedicated
155 to the issue of apparent and real absorption. It starts with a brief description of the
156 instruments, measurement strategy, data processing, generation of the 3D cloud, and of
157 the 3D RT model (section 2). Results are presented in section 3. In the conclusions
158 (section 4), possible implications for remote-sensing and atmospheric energy budget are
159 discussed.

160

161 2. Instruments, data, and radiative transfer calculations

162

163 SSFR

164 The SSFR [Pilewskie *et al.*, 2003] measured spectral shortwave irradiance on the ER-2
165 (above clouds), and on the DC-8 below or within clouds. On both platforms, the up- and
166 down-looking optical inlets were fix-mounted on the aircraft fuselage and connected to
167 rack-mounted spectrometers though optical fibers. The spectral range (350-2150 nm) was
168 covered by using two spectrometers per optical inlet: a grating spectrometer with a
169 Silicon CCD array for near-ultraviolet (NUV), visible (VIS) and very-near-infrared (350-
170 1000 nm, 8 nm spectral resolution) and a spectrometer with Indium-Gallium-Arsenide
171 linear array detector for the shortwave infrared (900-2200 nm, 12 nm resolution)
172 wavelength range. Over the entire range, about 90% of the solar irradiance spectrum is
173 captured. The slit-functions and wavelength-response of the spectrometers were
174 measured in the laboratory prior to the field experiment. An absolute radiometric
175 calibration with a NIST-traceable light source (1000 W lamp) was performed in the
176 laboratory before and after the experiment, and the stability of the calibration was
177 monitored with field-calibrators regularly throughout the experiment. The absolute
178 radiometric accuracy was 3-5% (precision 0.1%). The data were corrected for the angular
179 response of the light collectors and for changes in downward irradiance due to aircraft
180 attitude. The attitude correction was necessary because the light collector reference plane
181 (SSFR horizon) deviated from horizontal alignment due to changes in aircraft pitch, roll,
182 and heading; no active stabilization as described by *Wendisch et al.* [2001] was available

183 for this experiment. In some cases, the attitude correction failed because of reflections
184 from nearby clouds that could not be accounted for by the correction algorithm.

185

186 Deriving cloud absorption from SSFR measurements

187 In aircraft measurements, cloud absorption is derived from the difference of net
188 irradiance, $F_{net} = F^\downarrow - F^\uparrow$, at the top and bottom of a layer: $\Delta F_V = F_{net, top} - F_{net, bot}$ where
189 ΔF_V denotes the vertical component of flux divergence (vertical difference of net
190 irradiance). It differs from true absorption ($F_{abs} = \Delta F = \Delta F_V + \Delta F_H$) when horizontal flux
191 divergence $\Delta F_H \neq 0$. Due to net horizontal photon transport, ΔF_H is non-zero for any
192 inhomogeneous distribution of atmospheric extinction, in particular in heterogeneous
193 clouds. It is only rarely measured directly. In absence of physical absorbers, $F_{abs} = 0$, and
194 ΔF_H is balanced by ΔF_V that is opposite in sign. The magnitude of ΔF_V is a measure for
195 net horizontal photon transport, and is called apparent absorption. For non-conservative
196 scattering, ΔF_V incorporates real absorption (F_{abs}) and net horizontal transport effects:
197 $\Delta F_V = F_{abs} - \Delta F_H$. For pronounced horizontal heterogeneity, ΔF_H may dominate ΔF_V ,
198 which makes it hard to estimate F_{abs} . The reason why we focus on ΔF_V is that no
199 assumptions about cloud heterogeneity are necessary to derive it, in contrast to F_{abs} .

200

201 Fractional absorption (or apparent layer absorptance) is obtained from ΔF_V by
202 normalizing with F^\downarrow_{top} . While error analysis is virtually impossible when estimating (F_{abs})
203 from ΔF_V , it is non-trivial to derive realistic error-estimates even for ΔF_V itself. A brute
204 force method would be combining the radiometric uncertainties (3-5%) with linear error
205 propagation: $e(\Delta F_V) \approx |e(F^\downarrow_{top})| + |e(F^\uparrow_{top})| + |e(F^\downarrow_{bot})| + |e(F^\uparrow_{bot})|$ where e denote systematic

206 absolute instrument uncertainties. However, since all spectrometers are calibrated with
207 the same light source, the errors are not independent. A more realistic uncertainty
208 estimate would be the stability of the spectrometer response functions throughout the
209 experiment (better than 1-2% during TC⁴). Another major contributor to total uncertainty
210 is horizontal misalignment of the sensors. Even after correcting for aircraft attitude, a
211 residual error remains. It can exceed radiometric uncertainty [*Wendisch et al.*, 2001] and
212 is hard to derive from theoretical considerations as it depends on the specific
213 measurement situation. We therefore used an empirical estimate of 7% for the maximum
214 total error in downward irradiance. This error subsumes contributions from radiometric
215 calibration, attitude correction, and angular response of the light collectors and was
216 determined by comparing downward modeled and measured irradiance above clouds and
217 in cloud-free areas for all wavelengths (excluding gas absorption bands). For the upward
218 irradiance, we used 5% as maximum error estimate. The net-irradiance error was
219 obtained from linear error propagation: $e(F_{\text{top}}) \approx |e(F_{\text{top}}^{\downarrow})| + |e(F_{\text{top}}^{\uparrow})|$ and $e(F_{\text{bot}}) \approx |e(F_{\text{bot}}^{\downarrow})|$
220 $+ |e(F_{\text{bot}}^{\uparrow})|$. The top-of-cloud and bottom-of-cloud errors were combined by Gaussian
221 error propagation: $e(\Delta F_{\text{V}}) \approx (e(F_{\text{top}})^2 + e(F_{\text{bot}})^2)^{1/2}$.

222

223 MAS

224 The horizontal cloud structure was inferred from the MODIS Airborne Simulator (MAS).
225 It provided fields of cloud top height, optical thickness (τ) and effective crystal radius
226 (r_{eff}) at a resolution between 50 and 100 m (depending on flight altitude and cloud top
227 height). For high clouds, the cloud top height retrieval was based on the CO₂ slicing
228 technique as used by MODIS [*Menzel et al.*, 2008]. The algorithm that normally uses

229 four CO₂ MODIS channels was adapted to use the three channels available on MAS. For
230 low clouds, the algorithm reverts to the IR window method. The retrieval of optical
231 thickness and crystal effective radius was based on *Nakajima and King* [1990]: For each
232 pixel, reflectance pairs in a visible (or very-near-infrared) channel and a near-infrared
233 channel were compared with one-dimensional forward model calculations. While the
234 shorter wavelength channel was chosen outside gas absorption bands and contains mainly
235 information on optical thickness, the longer wavelength near-infrared channel is affected
236 by liquid water or ice absorption and is sensitive to drop or crystal size. The closest
237 match of the observed reflectance with pre-calculated modeled values was used to infer
238 the optical thickness and effective radius pair. For the TC⁴ data processing, algorithms
239 similar to the ones used in MODIS collection 5 retrievals were used, where scattering
240 phase function and single scattering albedo for ice clouds rely on calculations by *Baum et*
241 *al.* [2005]. Liquid water cloud scattering phase functions were calculated from Mie
242 calculations based on gamma drop size distributions with an effective variance of 0.1
243 [*Platnick et al.*, 2003]. Detailed instrument information and a description of the retrieval
244 algorithm are given in King et al. [2004, 2009]. *Eichler et al.* [2009, this issue] discuss
245 the impact of crystal habit and 3D radiative effects on the retrievals. MAS data collected
246 during TC⁴ were compared with MODIS cloud retrievals [*King et al.*, 2009].

247

248 CRS

249 The vertical cloud structure below the ER-2 flight track was derived from the reflectivity
250 profiles measured by the cloud radar system (CRS: *Li et al.* [2004]) onboard the ER-2.
251 The resolution of the reflectivity field are 37.5 meters in the vertical and about 100

252 meters in the horizontal. The minimum detectable reflectivity is about -28 dBZ for CRS
253 at a distance of 15 km. The reflectivity from CRS has been compared with the reflectivity
254 from another radar at X- band on ER-2 near the cloud top. Near the cloud top, the
255 reflectivities at both radar frequencies are about the same, an indication that the ice
256 particles obey Rayleigh scattering [*Tian et al.*, 2009].

257

258 Case from 17th of July, 2007

259 We selected one of the well-coordinated flight legs from July 17th, 2007 (from 15:20 to
260 15:35 UTC). Figure 1 shows this flight leg in the larger scale context (Geostationary
261 Operational Environmental Satellite (GOES) infrared image from 15:28 UTC). It was
262 located 300 km south of Panama (around 5°N, 83°W), near the edge of a high-cloud
263 system. The concurrent GOES VIS image (not reproduced here) shows that the cirrus-
264 free area was partly covered by low-level clouds. The Sun azimuth was northeast, at a
265 zenith angle of approximately 35°.

266

267 Both aircraft were guided from the mission operation center at the airport in San José
268 using NASA's Real Time Mission Monitor tool (RTMM, <http://rtmm.nsstc.nasa.gov/>)
269 that allowed the mission manager to coordinate the aircraft within two minutes on the
270 same ground track. Despite the frequent occurrence of coordinated flight legs throughout
271 the experiment, only one case qualified for our study based upon stringent selection
272 criteria: In order to correctly quantify cloud absorption, the bulk of the cloud layer had to
273 be bracketed by the two aircraft. Due to logistical constraints the DC-8 was frequently
274 scheduled to fly only in-cloud; that is, no below-cloud legs were scheduled. Even for the

275 17th of July case studied here the DC-8 flew almost entirely within the cloud layer. Since
276 no vertical structure was available from MAS, the information from CRS onboard the
277 ER-2 was vital in order to account for the position of the DC-8 within the cloud. Without
278 this information, it would be impossible to match measured and modeled irradiance at the
279 position of the DC-8. A further, less stringent, requirement was that clouds be composed
280 entirely of ice crystals, determined by the pixel-by-pixel thermodynamic phase
281 information from MAS. Finally, only cases where the attitude correction could be applied
282 (pitch and roll angles within certain limits) were used. These three requirements limited
283 the amount of useable data considerably.

284

285 Figure 2 shows the MAS-retrieved cloud optical thickness (gridded to 500 m resolution),
286 CRS reflectivity, and the SSFR spectral albedo for the same ER-2 flight leg as in Figure
287 1. For the upper panel, blue colors correspond to low; red and black colors to high optical
288 thickness. Cloud gaps are represented by white. The length of the scene is 192 km, the
289 width (swath) 17.5 km. In Figure 2, the southeast to northwest flight track is aligned from
290 left to right. The green shaded areas in the CRS panel mark areas where no data were
291 available. The thick black line represents the MAS-derived cloud top height along the
292 ER-2 flight track which captures the cloud top structure rather well. The dotted line
293 indicates the approximate flight altitude of the DC-8, showing that during large portions
294 of the leg, the DC-8 was actually within cloud rather than below. On some sections of the
295 leg, the radar sensed low-level clouds between the surface and 4 km that were decoupled
296 from the high-level outflow of the cell northeast of the flight leg. The bottom panel shows
297 time series of spectral albedo, with the wavelength varying in the vertical. The SSFR

298 albedo is nearly saturated in the visible wavelength range (red values indicating an albedo
299 near unity) in the optically thick cloud regions. The albedo time series (horizontal lines in
300 the albedo panel) exhibits far less variability than the associated cloud optical thickness,
301 mainly due to the hemispherical (geometrical) averaging inherent to irradiance. Some of
302 the wavelengths show minima that correspond to gas absorption bands. Ice absorption
303 bands (for example around 1500 nm can also be distinguished.

304

305 Input Cloud Generation

306 The fields of optical thickness and effective radius from MAS and the reflectance data
307 from CRS were combined to provide the input to three-dimensional radiative transfer
308 calculations. The profile of radar reflectivity Z (in units of dBZ) was used to derive
309 approximate vertical profiles of ice water content ($IWC(z)$, in g m^{-3}) along the flight track
310 following *Liu and Illingworth* [2000]: $IWC = 0.137 * Z^{0.64}$. For each vertical profile along
311 the flight track, the column-integrated ice water path (IWP_{CRS}) was calculated. The IWP
312 was also retrieved from MAS: $IWP_{\text{MAS}} = 2/3 * \rho_{\text{ice}} * \tau * R_{\text{eff}}$, where ρ_{ice} is the density of ice
313 (approximately 0.925 g cm^{-3}). While the CRS profile was only measured along the center
314 (nadir) track, MAS-derived IWP was available across the entire swath for each point
315 along the track. In the model cloud, the IWC profiles were obtained through $IWC(z) =$
316 $IWC_{\text{CRS}} * IWP_{\text{MAS}} / IWP_{\text{CRS}}$. The entire profile was shifted in altitude corresponding to the
317 cloud top height as retrieved by MAS. Due to the lack of other information, the effective
318 radius was set to $R_{\text{eff}}(x,y,z) = R_{\text{eff,MAS}}(x,y)$, which is clearly a simplification because the
319 crystal size distribution in the lower regions of the cloud is fundamentally different from
320 that near the top. The MAS-derived effective radius is representative of the topmost layer

321 of the cloud [Platnick *et al.*, 2000] where ice crystals are often smaller than in lower
322 layers within the cirrus [Francis *et al.*, 1998; Gayet *et al.*, 2004]. However, a
323 considerable part of radiation is absorbed in the uppermost cloud layer. Therefore, the
324 cloud-top effective radius can be regarded as a valid representation for our study.

325

326 In summary, all cloud properties (*IWC*, optical thickness, effective radius, and cloud top
327 height) were tied to MAS measurements; the CRS profiles were used to distribute the
328 MAS-derived IWP vertically. Thereby, the CRS profiles (available only below the ER-2
329 flight track) were used across the entire MAS swath.

330

331 The generated 3D cloud was gridded to 500 m horizontal and 1000 m vertical resolution.
332 For testing, a version with 100 m horizontal resolutions was also generated. However, the
333 impact of spatial resolution is not the focus of this particular study, and the high-
334 resolution cloud was not used in the interest of saving CPU time.

335

336 Radiative Transfer Calculations

337 All calculations were done with the libRadtran radiative transfer package by Mayer and
338 Kylling [2005]. The generated cloud microphysical properties within the $384 \times 35 \times 20$
339 boxes ($n_x \times n_y \times$ number of layers) constitutes the main input for 3D RT calculations,
340 along with atmospheric profiles from dropsondes (launched from the DC-8), and from the
341 DC-8 and ER-2 meteorological data (pressure, relative humidity). For the spectral sea
342 surface albedo, data measured by SSFR during CRYSTAL-FACE (Cirrus Regional Study
343 of Tropical Anvils and Cirrus Layers – Florida Area Cirrus Experiment) was used

344 [*Schmidt et al.*, 2007b]. For the 3D RT calculations, we applied the forward version of
345 the Monte-Carlo code MYSTIC (Monte Carlo code for the physically correct tracing of
346 photons in cloudy atmospheres: *Mayer* [2009]; *Mayer* [1999]) which is embedded in
347 libRadtran (<http://www.libradtran.org>). The extraterrestrial spectrum by *Kurucz* [1992],
348 averaged over 1 nm bins, was used as top-of-the atmosphere incident solar irradiance
349 spectrum. For the sake of computational efficiency, the scattering phase functions were
350 represented by the Henyey-Greenstein parameterization based on the asymmetry
351 parameter g (the first moment of the phase function). As shown in *Schmidt et al.* [2007b],
352 this approximation is reasonably close to the exact representation of the cloud phase
353 function, at least for sun angles not too far from zenith position. Both asymmetry
354 parameter and single scattering albedo were taken from ray tracing calculations by *Yang*
355 *and Liou* [1998]. Calculations were performed for nine wavelengths: 400, 450, 500, 600,
356 700, 800, 850, 1200, and 1600 nm, using 10^9 photons each.

357

358 3. Results

359

360 As a first step, we compared the measured time series of upward and downward
361 irradiance above (that is, at ER-2 altitude) and below (or within) clouds (that is, at DC-8
362 altitude) with model results. To this end, the downward irradiance was rescaled such that
363 changes in solar zenith angle (ranging from $SZA = 34^\circ$ - 36° during the leg) were
364 compensated using $F^\downarrow(SZA_0) = F^\downarrow(SZA) \times (\cos(SZA_0)/\cos(SZA))$, where $SZA_0 = 35^\circ$ was
365 used in the model calculations as well. This correction is discussed in *Schmidt et al.*
366 [2007a]. For the ER-2, both upward and downward irradiance agreed within the expected

367 uncertainties for all nine wavelengths. Less agreement was achieved for the DC-8, which
368 was expected because firstly, the CRS profiles were only available directly underneath
369 the ER-2 and do not represent the vertical cloud structure across the entire MAS swath.
370 Secondly, any mismatches in model and measurement altitude translate directly into
371 discrepancies in irradiance values, due to the large vertical gradients of downward and
372 upward irradiance within cloud layers. The net irradiance is less sensitive; for
373 wavelengths outside gas and cloud absorption bands it is expected to be constant with
374 altitude.

375

376 The vertical difference of net irradiances on top and at the bottom of the cloud layer, that
377 is, vertical flux divergence (ΔF_V), is shown in Figure 3 for 500 nm. At this wavelength,
378 the clouds themselves do not absorb and atmospheric gas absorption is near zero. No
379 absorbing aerosol particles were present. Therefore, negligible values are expected for
380 F_{abs} . In absence of true absorption, positive values of ΔF_V (apparent absorption) indicate
381 that photons are lost through the sides of the cloud column ($\Delta F_H < 0$); negative values
382 (apparent emission) correspond to a net photon gain. The observations (black dots) are
383 shown with error bars that were estimated from the individual absolute uncertainties as
384 explained above. Throughout almost the entire leg, significant apparent absorption is
385 observed that is not balanced by negative values. On average, $0.17 \text{ W m}^{-2} \text{ nm}^{-1}$ are found.
386 In the modeled vertical flux divergence, in contrast, negative and positive ΔF_V values are
387 balanced and $\langle \Delta F_V \rangle = 0$ because $F_{\text{abs}} = 0$ and $\langle \Delta F_H \rangle = 0$. The domain-averaged horizontal
388 photon transport vanishes due to periodic boundary conditions. The bias between
389 observations and model varies between 0 and $0.2 \text{ W m}^{-2} \text{ nm}^{-1}$. In most areas, the modeled

390 values are within the uncertainty of the observations. In some places, the discrepancies
391 are larger than the error bars, for example at UTC=15.36 h and UTC=15.47 h.

392

393 The red line in Figure 3 shows the MAS optical thickness retrievals averaged within the
394 SSFR footprint. The SSFR footprint is defined as a circle from within which 50% of the
395 ER-2-measured upward irradiance originates. Usually, the SSFR footprint diameter is
396 contained within the MAS swath width. As can be seen in Figure 3, local maxima of
397 optical thickness are connected with high values of ΔF_V and thus a net horizontal outflow
398 of photons; local optical thickness minima are related to minima of ΔF_V .

399

400 A possible explanation for the discrepancy between observations and model results could
401 be the limited model domain size (given by the MAS swath width). While in the
402 calculations, photons are confined within the model boundaries, they are not restricted in
403 this way in the real world. If the measurement area is surrounded by regions of lower
404 optical depth or even clear sky, a net transport of photons into these regions can occur, in
405 the same way as between areas of different optical thickness *within* the domain.

406 However, there are theoretical limits for the horizontal displacement of photons. For
407 example, the mean horizontal distance traveled by transmitted photons is in the range of
408 cloud geometrical depth [Marshak *et al.*, 1995] (less for reflected photons). Although the
409 GOES IR image shows that there are indeed areas without high clouds southwest of the
410 flight track, they may be too far off to explain the observations.

411

412 In addition to the localized radiative smoothing, irradiance fields incur hemispherical
413 (cosine-weighted) averaging of the underlying radiance fields, which could also
414 contribute to the discrepancy. After all, only about 50% of the irradiance originates from
415 within the MAS swath, and the model results can be biased if the clouds outside the
416 domain are not properly represented by the model cloud. Both effects – photon loss into
417 neighboring areas and geometrical averaging – can only be examined by embedding the
418 MAS-based cloud within the larger context of GOES-derived cloud fields. This is beyond
419 the scope of this study. Until radiative transfer calculations in an extend domain prove the
420 explanations brought forward, one cannot rule out other causes for the discrepancies.

421

422 Figure 4 illustrates the relationship between cloud optical thickness and ΔF_V . The
423 observations are shown for 500 nm (black dots) and 1600 nm (red dots), as a function of
424 MAS-retrieved optical thickness (averaged over the SSFR footprint). The propagated
425 error from the measurement uncertainties is shown at maximum optical thickness. The
426 error is larger for 500 nm (only negative error bar is shown) than for 1600 nm because
427 500 nm is near the maximum of the solar spectrum, and ΔF_V is derived from the
428 difference of large (500 nm) as opposed to small (1600 nm) quantities. In this particular
429 case, the values of ΔF_V are comparable in magnitude for the two wavelengths although
430 the processes involved are fundamentally different: At 1600 nm, true absorption by ice
431 crystals prevails. The modeled dependence of F_{abs} on optical thickness is plotted as red
432 line. Observed (small) excursions from the modeled values can be explained by
433 horizontal photon transport: $\Delta F_V = F_{\text{abs}} - \Delta F_H$. At 500 nm, in contrast, the true absorption
434 is expected to be close to zero ($F_{\text{abs}} \approx 0$), and $\Delta F_V \approx -\Delta F_H$. As discussed before, almost all

435 the observations exhibit positive ΔF_V , whereas the values from the 3D model calculations
436 (blue dots) do not show such a bias.

437

438 There is some indication from the model and observations that net transport of radiation
439 occurs from optically thicker to thinner regions. Obviously no 1:1 relationship can be
440 established, partly because net photon transport takes place between *local* maxima and
441 minima of optical thickness. Net transport of radiation is not induced by optical thickness
442 contrasts if they are separated by scales larger than the mean horizontal photon
443 displacement. For example, radiative smoothing, and thus ΔF_H , at 1600 nm is suppressed
444 in comparison to 500 nm because the absorption has a shortening effect on photon
445 horizontal transport distances.

446 For small optical thickness (5-10 in Figure 4), positive and negative apparent absorption
447 occurs in the model calculations, uncorrelated with the optical thickness itself. These
448 excursions from zero absorption suggest that in areas with sparse or thin cloud cover,
449 other factors than large-scale horizontal cloud distribution dominate photon transport, for
450 example vertical heterogeneities (multiple layers) and interactions with the surface.

451

452 In Figure 5, we show the spectral shape of measurement-derived and simulated cloud
453 absorptance (defined as $\Delta F_V/F_{\text{top}}^{\downarrow} \times 100\%$) in two different areas of the cloud: at
454 UTC=15.38 h (local maximum of τ – red line) and at UTC=15.44 h (local minimum of τ
455 – blue line), both of which are marked in Figure 3. Results from the 3D model runs are
456 shown as red and blue symbols. The black line shows the absorptance spectrum obtained
457 when averaging observations over the entire leg from UTC=15.33 h – 15.53 h. In the VIS

458 and very-near-infrared, the averaged spectrum is close to the high optical thickness case,
459 the opposite is true the NIR wavelength range. Domain-averaged model results from the
460 independent pixel method are shown as dotted black line.

461

462 In all spectra, ice absorption effects are clearly visible in the NIR wavelength range (e.g.,
463 around 1500 nm and 2000 nm). Furthermore, the oxygen A-band around 762 nm, and
464 water vapor absorption at 940 nm, 1140 nm and 1350 nm can be distinguished. The water
465 vapor absorption is rather weak because at high altitudes, water vapor concentration is
466 low. In the cases presented here, considerable apparent absorption (up to 20%) is found
467 all across the visible wavelength range, with an upward slope at wavelengths short of 450
468 nm. For the higher optical thickness case, the spectral behavior is reproduced by the 3D
469 calculations (squares), although not equally across the spectrum (for example, 800 and
470 850 nm). This gives us some confidence that the observed effects are not measurement
471 artifacts. Above 500 nm, the range of uncertainty of absorptance excludes zero, and the
472 apparent absorption effect is statistically significant. For the low optical thickness case,
473 the 3D model predicts near-zero apparent absorption across most of the visible range. At
474 1200 nm, the model results fall only marginally within the range of uncertainty of the
475 observations. The domain-averaged independent pixel approximation (IPA) model results
476 do not reproduce the apparent absorption at VIS and very-near-infrared wavelengths.

477 This is expected because horizontal photon transport is ignored by IPA ($\Delta F_H=0$), thus
478 $\Delta F_V=F_{\text{abs}}=0$. In the NIR range, IPA over-predicts cloud absorption. A possible reason is
479 that on the domain-average, the MAS-derived effective radius is overestimated [*Eichler*
480 *et al.*, 2009] which implicates higher NIR absorption. The cross-over between under- and

481 overestimation of absorptance by IPA with respect to the observations is located at about
482 1400 nm.

483

484 The spectral shape in Figure 5 indicates that in high optical thickness areas, the
485 broadband-integrated value of ΔF_V consists of significant contributions from ΔF_H
486 (mainly from below 1400 nm) as well as F_{abs} (from above 1400 nm). In extreme cases,
487 ΔF_H can exceed F_{abs} . In our case, the observed broadband absorption, averaged over the
488 flight leg, amounts to 187 W m⁻², contrasted by only 81 W m⁻² modeled by IPA that
489 ignores horizontal photon transport.

490

491 The reason for the spectral slope at the shortest wavelengths is not entirely understood. It
492 is likely due to a combination of molecular scattering and wavelength-dependent
493 horizontal photon transport. *Marshak et al.* [2008] described a related effect for radiance,
494 the so-called "bluing of the atmosphere" around clouds. Since molecular scattering is
495 stronger at short wavelengths, enhanced reflected radiation near cloud edges gets
496 scattered more effectively at short ("blue") wavelengths and is redirected into satellite
497 sensors. This mechanism could also play a role in explaining the irradiance-based effect
498 that is described here. At very short wavelengths, the apparent absorption becomes
499 negative, leading to an apparent emission of irradiance in the near-UV to blue spectral
500 range.

501

502 The spectral signature of the apparent absorption may prove important for cloud and
503 aerosol remote sensing. If the reflectance at different wavelengths responds differently to

504 cloud heterogeneity effects, this has consequences for cloud retrievals. This spectral
505 aspect of cloud retrieval biases occurs in addition to various 3D effects that have been
506 discussed in the literature. Due to the different spatial scales, this additional effect may be
507 more important for cirrus than for boundary layer clouds. A further implication is that
508 any retrieval based on reflectance ratios in the near-UV and visible wavelength range,
509 such as the aerosol index, will be distorted in the presence of cirrus, or other clouds.
510 Regarding the correction technique by *Ackerman and Cox* [1981], the choice of a visible
511 wavelength able to correct for net horizontal photon transport in absorption
512 measurements appears to be problematic, since it is not constant even throughout the non-
513 absorbing part of the spectrum. Related techniques are not as dependent on the spectral
514 signature. However, they often rely on the assumption that horizontal photon transport is
515 comparable for non-absorbing and absorbing wavelengths, which is not the case.
516
517

518 4. Conclusions

519

520 In this paper, we studied measured and modeled solar spectral absorption, based on data
521 from the NASA TC⁴ experiment in Costa Rica (2007). Most previous studies sought to
522 infer true absorption F_{abs} from measurements of vertical flux divergence. This is
523 problematic in heterogeneous clouds where horizontal fluxes occur. Therefore, we
524 focused on apparent cloud absorption (vertical flux divergence ΔF_V), a quantity that
525 comprises net horizontal photon transport (horizontal flux divergence ΔF_H) as well as
526 true cloud absorption, F_{abs} : $\Delta F_V = F_{\text{abs}} - \Delta F_H$. We used SSFR measurements of upward
527 and downward spectral solar irradiance onboard the NASA ER-2 and DC-8 aircraft that
528 were flown in stacked formation above and below the outflow of a tropical convective
529 system on the 17th of July, 2007. NASA's aircraft-ground communication tool (RTMM)
530 allowed a close coordination of the two aircraft in time and space. In this way, the cloud
531 field was sampled over more than 192 km, and a time series of apparent absorption was
532 derived from the differences of above and below-cloud net irradiances. In addition,
533 simultaneous cloud remote-sensing data (MAS-derived horizontal distribution of cloud
534 optical thickness, crystal effective radius, and cloud top height, as well as CRS-derived
535 cloud extinction profiles) were available from the high-flying aircraft. This allowed
536 constructing a 3D model cloud that could be used as input to 3D radiative transfer
537 calculations to validate the measurements on a point-by-point basis along the entire flight
538 leg.

539

540 For the first time, we were able to determine the spectral shape of the vertical net flux
541 difference (apparent absorption), and to reproduce it with model calculations. We found
542 considerable positive apparent absorption in the visible wavelength range where clouds
543 do not absorb ($F_{\text{abs}}=0$) that could, at least in part, be explained by net horizontal photon
544 transport. Below 500 nm, the apparent absorption decreases with wavelength and can
545 become negative, thus entailing apparent emission of blue to near-UV radiation by
546 clouds. For absorbing wavelengths, no bias between model results and observations was
547 found. Adjustments of the effective radius like in *O'Hirok and Gautier* [2003] were not
548 required. For non-absorbing wavelengths, measured apparent absorption exceeded 3D
549 model calculations at various points along the leg and averaged over the entire leg. We
550 found correlations between local maxima (minima) of optical thickness on the one hand
551 and high (low) values of vertical flux divergence (ΔF_v), suggesting net horizontal photon
552 transport between those areas. The GOES-IR image indicates that the sampled cloud field
553 was surrounded by areas of lower optical thickness or cirrus-free sky which could give
554 rise to a net loss of photons from the sample area (unaccounted for by the model), thus
555 explaining the enhanced value of apparent absorption in the observations. It was,
556 however, beyond the scope of this study to explore if including the areas around the
557 sample cloud in the model calculations would support this hypothesis and thus fully
558 resolve the reasons for previously observed "absorption bias".

559

560 An open question is over which scales photons can effectively be transported within
561 clouds, or away from cloud systems into clear-sky areas. For boundary layer clouds,
562 theoretical limits exist for the maximum horizontal photon displacement [*Platnick, 2001*].

563 On average, the geometrical distance does not exceed the vertical extent of a cloud layer
564 [*Marshak et al.*, 1995]. When sampling clouds over areas that are larger than this
565 distance, the net horizontal photon flux is expected to be balanced ($\langle\Delta F_V\rangle=0$). Those
566 distances might be larger in high-cloud systems, especially when multiple layers are
567 involved. Moreover, the geometrical averaging inherent to irradiance introduces different
568 effects for boundary layer clouds and large-scale convection systems, just because of the
569 different dimensions. *Kindel et al.* [2009] (Figure 11) shows that irradiance-based
570 retrievals of cloud optical properties of anvils are biased low with respect to radiance-
571 based counterparts, because of the influence of clear-sky areas beyond the imager's
572 swath.

573

574 A different manifestation of net horizontal photon transport was observed by *Eichler et*
575 *al.* [2009], using the same model cloud as employed in this study. The net outflow of
576 photons from optically thick areas makes them appear darker and leads to an
577 underestimation of cloud optical thickness by the imager. The opposite effect in optically
578 thin areas does not fully compensate this bias and leads to a net effect of underestimation
579 of optical thickness. As shown above, this effect is not spectrally neutral. The apparent
580 absorption in optically thick areas is decreasingly effective at wavelengths shorter than
581 500 nm, which leads to a relative enhancement of shorter-wavelength irradiance.
582 Methods that rely on reflectance ratios at different wavelengths such as the aerosol index
583 will thus be heavily affected in the presence of clouds. A further implication of the
584 spectrally-dependent apparent absorption is that the method by *Ackerman and Cox* [1981]
585 for correcting absorption measurements is compromised: It assumes that horizontal

586 photon transport impacts all wavelengths equally and uses an arbitrary non-absorbing
587 wavelength to correct near-infrared measurements. However, the apparent absorption is
588 not equal across the non-absorbing wavelength range and radiative smoothing is
589 wavelength-dependent. Thus the choice of a visible wavelength for corrections becomes
590 somewhat arbitrary.

591

592 Forthcoming research in this area should focus on the following questions:

- 593 1. Explain the spectral shape of the near-UV and VIS apparent absorption on a
594 theoretical basis.
- 595 2. Determine over which scales horizontal photon transport occurs in high-cloud
596 systems and if embedding the MAS cloud scene in the larger context of GOES
597 can resolve the remaining positive bias between model and observation by
598 attributing it to net photon outflow into clear-sky areas.
- 599 3. Connect the spectral signature of heterogeneous clouds in the apparent absorption
600 with remote-sensing applications.

601

602

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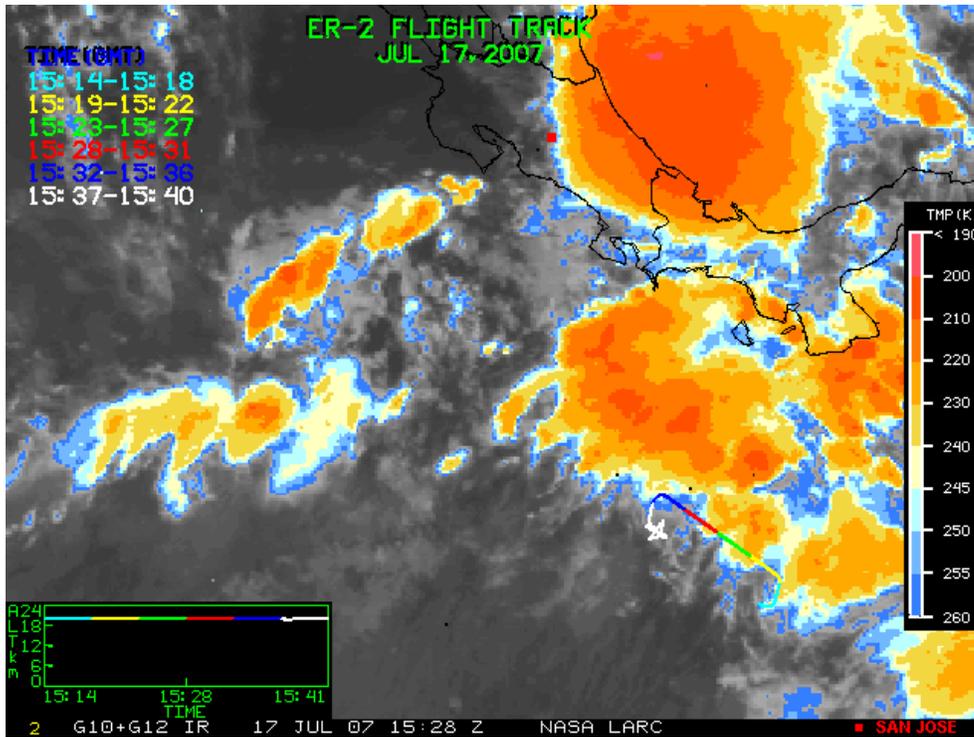
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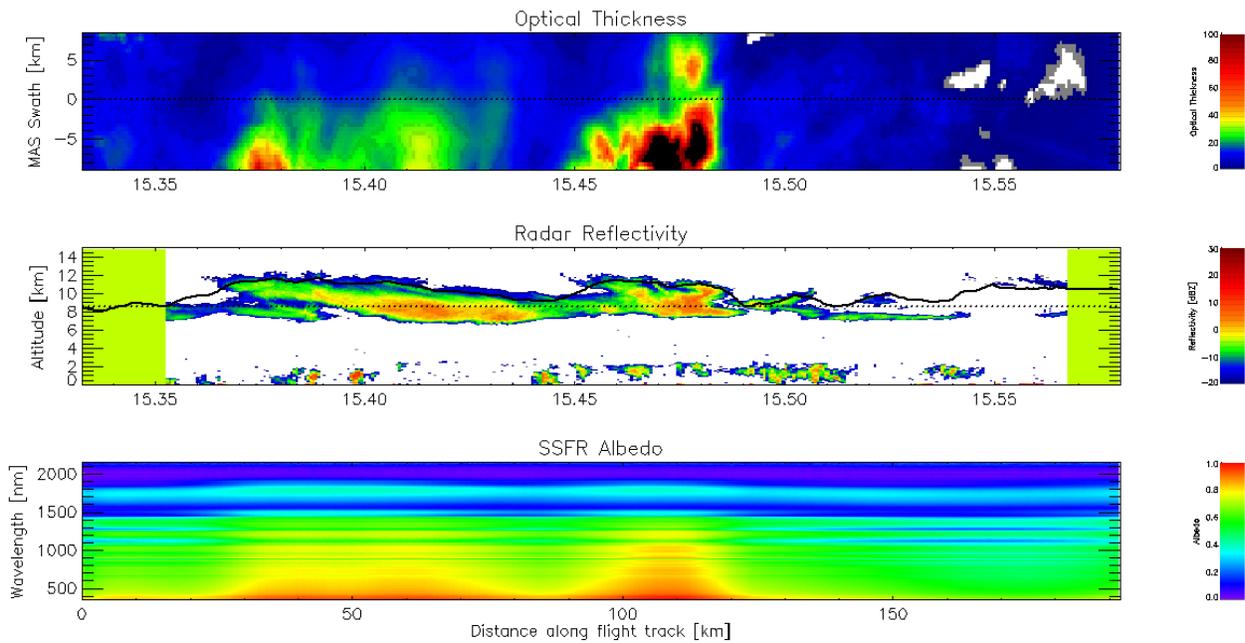


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849 **Figure 1** – ER-2 flight leg from 15:20 to 15:35 (straight part of the shown track), in the context of the
 850 GOES G10 and G12 IR image from 15:28 UTC. The DC-8 was flown directly underneath. Image courtesy
 851 NASA LARC (<http://www-angler.larc.nasa.gov/tc4/>)

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855 **Figure 2** – Data along the 17th of July flight track (15:20 to 15:35 UTC). Top panel: MAS-retrieved cloud

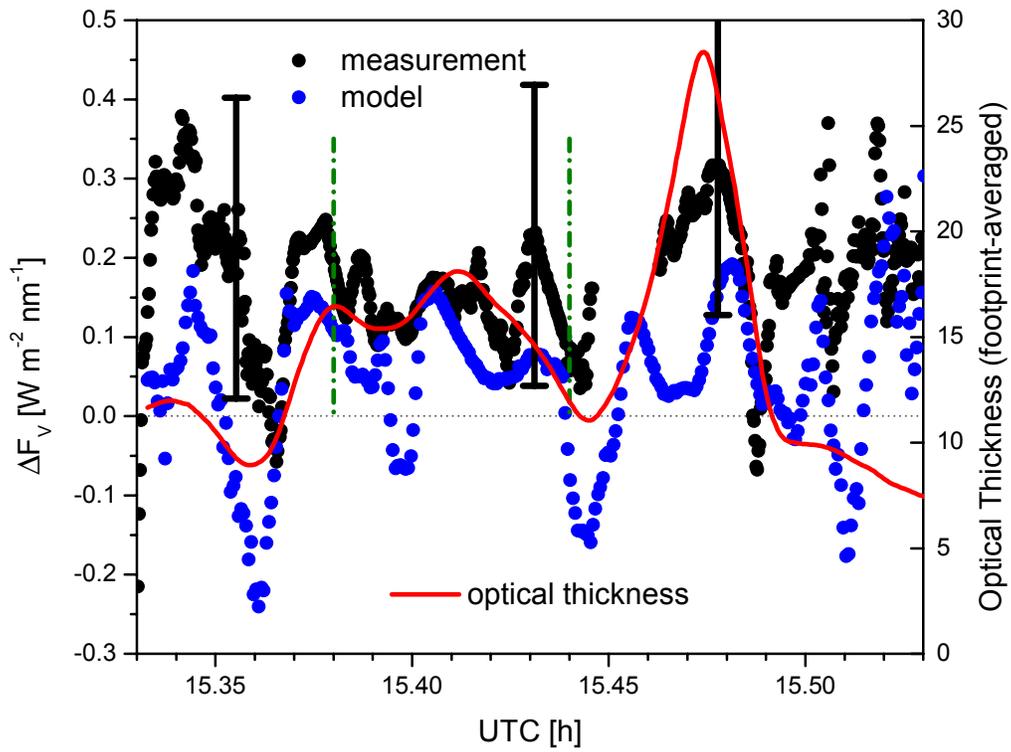
856 optical thickness (swath 17.5 km); below: radar reflectivity from CRS in dBZ. Regions with no data

857 available are marked in light green. Cloud top height from MAS along the ER-2 flight track is over-plotted

858 as thick black line; the dotted line indicates the approximate flight altitude of the DC-8. Bottom panel: ER-

859 2 albedo (wavelength vertical dimension) along the flight track.

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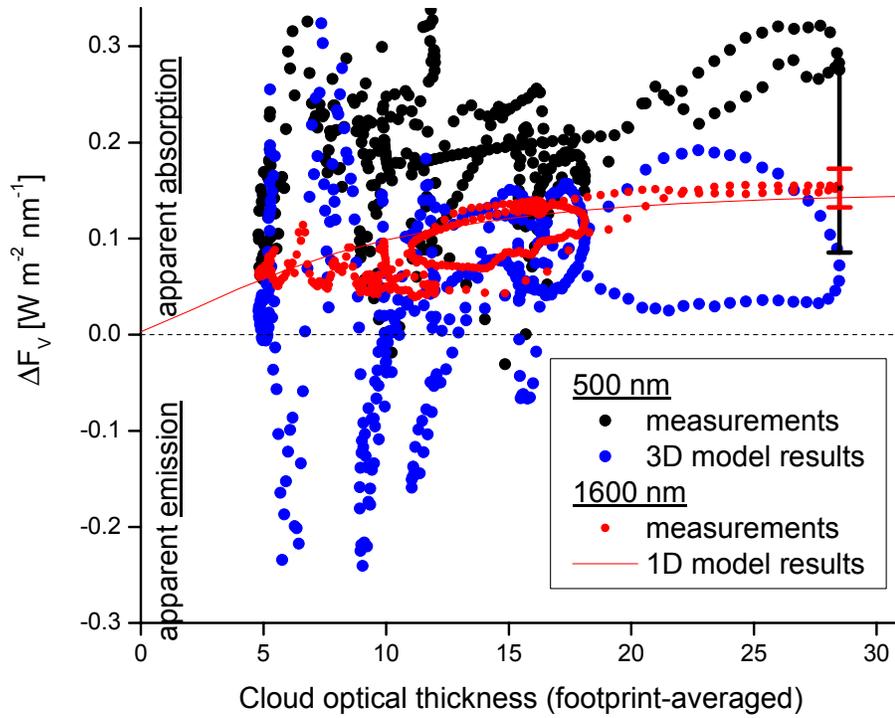


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862 **Figure 3** –Time series of measured (black dots) and modeled (blue dots) vertical difference of net
 863 irradiances at 500 nm, along with SSFR-footprint-averaged optical thickness (red line). The dash-dotted
 864 green lines at UTC=15.35 h and UTC=15.44 h mark where spectra of ΔF_V are shown in Figure 5.

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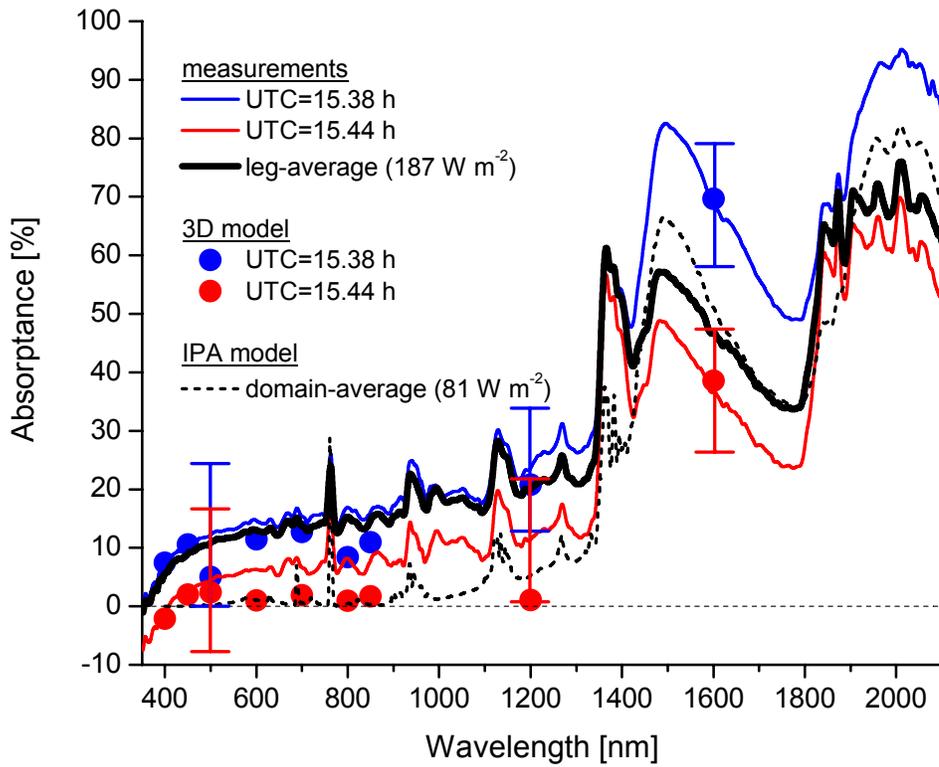
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868 **Figure 4** – Measured (black dots) and modeled (blue dots) vertical difference of net irradiances at 500 nm
 869 as a function of SSFR-footprint-averaged optical thickness. For comparison, the measurements at 1600 nm
 870 are shown (red dots) along with the 1D model true absorption (red line).

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874 **Figure 5** – Spectral absorptance (or fractional absorption) at two selected points along the flight track
 875 (UTC=15.44 h – optically thin region, and UTC=15.38 h – optically thick region). The lines show the
 876 measurements with ice absorption bands around, for example, 1500 nm. The symbols show model results.