The thin border between cloud and aerosol: sensitivity of several ground based observation techniques.

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11 Abstract

Cloud and aerosol are two manifestations of what it is essentially the same physical 12 phenomenon: a suspension of particles in the air. The differences between the two come from 13 the different composition (e.g., much higher amount of condensed water in particles 14 15 constituting a cloud) and/or particle size, and also from the different number of such particles (10-10,000 particles per cubic centimeter depending on conditions). However, there exist 16 situations in which the distinction is far from obvious, and even when broken or scattered 17 18 clouds are present in the sky, the borders between cloud/not cloud are not always well defined, a transition area that has been coined as the "twilight zone". The current paper 19 presents a discussion on the definition of cloud and aerosol, the need for distinguishing or for 20 considering the continuum between the two, and suggests a quantification of the importance 21 and frequency of such ambiguous situations, founded on several ground-based observing 22 techniques. Specifically, sensitivity analyses are applied on sky camera images and 23 broadband and spectral radiometric measurements taken at Girona (Spain) and Boulder (Co, 24 USA). Results indicate that, at these sites, in more than 5% of the daytime hours the sky may 25 26 be considered cloudless (but containing aerosols) or cloudy (with some kind of optically thin 27 clouds) depending on the observing system and the thresholds applied. Similarly, at least 10% of the time the extension of scattered or broken clouds into clear areas is problematic to 28 establish, and depends on where the limit is put between cloud and aerosol. These findings 29 30 are relevant to both technical approaches for cloud screening and sky cover categorization 31 algorithms and radiative transfer studies, given the different effect of clouds and aerosols (and the different treatment in models) on the Earth's radiation balance. 32

33 **1. Introduction**

The Earth's atmosphere contains suspended particles, i.e. particles that because of their size have terminal fall velocities of the order of centimeters per second at most, so they have atmospheric residence times on the order of hours, days, or much longer in some cases. These particles vary in their chemical composition, have concentrations that vary in space and time, are present in both the solid and liquid phases and have sizes ranging over several orders of

39 magnitude. In gross aggregate, the suspension of particles receives two names: either cloud or aerosol. Simplifying, a cloud is an aggregate of a number of particles formed mainly of water, 40 in liquid or solid state (i.e., hydrometeors) of sizes between a few microns to some 41 millimeters and in sufficient concentration to be perceived by human vision from the Earth's 42 43 surface. Any other aggregate of particles is called, generically, atmospheric aerosol, and generally contains less liquid water than clouds. This includes wind-borne dust, sea spray 44 particles of salt, sulfate and organic particles, or ash and soot arising from combustion. 45 Precipitating particles such as rain, snow or hail (which have terminal fall velocity of the 46 order of meters per second) are excluded from this discussion. 47

Despite the above differences in origin and composition, clouds and aerosol could be 48 considered two manifestations of the same phenomenon. However, their description, 49 characteristics, and -in particular- interactions with solar and terrestrial radiation have 50 historically been studied separately. Indeed, the study of clouds extends back to ancient times 51 whereas the study of atmospheric aerosol is much more recent. In fact, the term was proposed 52 53 in the early 20th century, and has become popular within the atmospherics science community only after the 1960s or so, as previously unspecific names (dust, smoke, etc.) or 54 more technical designations (lithometeor, etc.) were used. The interactions between clouds 55 and aerosols are known, although their climatic significance is far from being fully quantified 56 (see the reviews of Heintzenberg 2012; Rosenfeld et al. 2014; Seinfeld et al. 2016). The 57 58 presence of different types or concentrations of aerosols has impacts on clouds, as some particulate matter (cloud condensation nuclei or ice nuclei) are more amenable for water 59 vapor to condense into droplets or crystals to form clouds. These effects, especially in the 60 field of energy balance, have been known as aerosol indirect effects (Albrecht, 1989; 61 62 Twomey, 1974) to distinguish from the direct (purely radiative by absorption and scattering) effect that aerosols have on the radiative energy transfer in the atmosphere. 63

Broadly speaking, there are two features that distinguish a cloud from other suspension of 64 particles in the air: i) the content of water in droplets and/or ice crystals, and ii) the visibility, 65 i.e., the appearance of a more or less clearly delimited form of (usually) white/grey color, 66 which is possible to see evolve (it should be noted that some aerosol suspensions are also 67 clearly visible, for example, a smoke plume). Both features allow quantification, i.e. one can 68 propose a threshold for the concentration of droplets or ice crystals (or for the amount of 69 condensed water), and also for the optical effect (the optical thickness at a certain 70 71 wavelength). Dupont et al. (2008) showed that solar irradiance and sky imagery retrievals 72 tuned to reflect human observations allow up to a visible optical depth of 0.15 to 0.2 of 73 primarily high ice haze to be traditionally classified as "cloud free" sky. But historically the decision on whether a volume of air is cloud or not (leaving no room for intermediate cases) 74 has been based on the judgment of a human observer on the ground. This does not seem very 75 76 scientific, since it can happen that the same volume of air containing aqueous particles are labeled as cloud or not depending on the contextual conditions in which the observation is 77 made, subject to the judgment and perception of the observer. Similar difficulties arise when 78 79 clouds are observed from satellites (Koren et al., 2008).

80 Consequently, fundamental questions remain: What is the limit of visibility from which a suspension of droplets must be considered cloud? Should this limit be set for an "average" 81 human eye, or can it be objectively established for some instrument as in Dupont et al. 82 (2008)? Or is it even reasonable to consider such a limit given that the aerosol/cloud particle 83 suspension could be considered as a continuum and not a dichotomic phenomenon. How does 84 85 one define visibility when observations are performed in a wavelength outside the visible range of the human eye? Droplets form on soluble hydrophilic particles whereas many ice 86 particles form on insoluble hydrophobic particles so how does one decide if the suspended 87 particles are aerosol particles or hydrometeors? When observation is performed by automated 88 instruments, trying to reduce to a three level classification (cloud / aerosol / clear sky) is even 89 more difficult (Tapakis and Charalambides, 2013). Subsequently, this classification has 90 consequences for climate studies (Charlson et al., 2007), including trend analysis, as derived 91 trends may depend on the instrument and/or methodology used to infer cloud amounts (Wu et 92 93 al., 2014).

94 A good example of the difficulties of defining cloud and aerosol is found regarding sky 95 images taken by "all-sky" cameras (they "see" an entire 180° sky view from a particular point at the surface). The digital images are analyzed to obtain information on the state of the sky, 96 in particular cloud cover and cloud type (Calbó and Sabburg, 2008; Heinle et al., 2010; 97 98 Kazantzidis et al., 2012; Long et al., 2006b). The problem is what thresholds to set to 99 distinguish between the "clear" and "cloudy" pixels. Even if more complex approaches are adopted (Li et al., 2011; Saito and Iwabuchi, 2016), they rely on the initial human decision 100 taken on the training images. In fact, sky cameras have also been proposed as devices to 101 observe and characterize the atmospheric aerosol (Cazorla et al., 2008). 102

103 This is not unique for cloud observations by ground-based imaging in the visible. For example, the difficulties in trying to distinguish clouds and aerosol in sunshine duration 104 records have been pointed out elsewhere (Sanchez-Romero et al., 2014). In addition, many 105 works focus on removing cloud "contamination" from aerosol observations performed with 106 sunphotometers or shadowband radiometers (Alexandrov et al., 2004; Kassianov et al., 2013; 107 Michalsky et al., 2010). The problem further expands when considering other views 108 (satellite) or other wavelengths (ceilometers in the infrared, microwave radiometers, weather 109 radars). All these difficulties have consequences in both meteorological and climatological 110 studies (e.g. Boers et al., 2010; Várnai and Marshak, 2011; Sanchez-Lorenzo et al., 2009; Wu 111 et al., 2014). 112

In general the distinction between a cloudy and a cloudless sky, and the separation between 113 cloud and aerosol, is appropriate for attribution studies and modeling radiative effects of 114 different climate forcing mechanisms, but imposing this classification may be unnecessary 115 (or inconvenient) in relation to new and advanced methods of observation and measurement. 116 If so, the distinction could also be unnecessary in radiative transfer models, or in future 117 parameterizations included in weather and climate models. This approach of a continuous 118 119 treatment of aggregates of particles in the atmosphere is relatively new, although some previous works have already pointed in this direction. 120

For example, Charlson et al. (2007) highlighted the importance that has been given to the 121 separation between the "cloud" and "clear" regimes in various fields of study including the 122 radiative forcing by clouds and the quantification of direct effects and indirect radiative 123 forcing by aerosols. The paper questioned the separation between the two regimes, and 124 suggested the desirability of treating the phenomenon as a continuum. Similarly, Koren et al. 125 (2007) described a transition zone ("twilight" zone) around the cloud in which the optical 126 properties are close to those of the cloud itself. The authors estimated that an appreciable 127 fraction (between 30 and 60%) of the part of the globe at any given time considered free of 128 clouds could correspond to that area of transition, a fact that could have important climate 129 implications. The question of the climatic importance of clouds that are considered "small" in 130 size was addressed by Koren et al. (2008), as well as the effect of the aerosol in the regions 131 between clouds (Koren et al., 2009). Also, Bar-Or et al. (2010) introduced the concept of 132 cloud field as an area that includes detectable clouds and twilight zone, and found that the 133 cloud field fraction could be as large as 97% in an area where the detectable cloud fraction is 134 53%. In the cited works, several methodologies were used: spectral radiometry from the 135 surface in the visible and near infrared, satellite measurements, and modeling. Also long-136 wave spectral radiometry is being used for the purpose of studying the properties of thin 137 138 clouds and the transition region (Hirsch et al., 2014, 2012).

139 Other researchers have studied radiative effects occurring in the vicinity of the clouds. Thus, 140 the question of increased reflectivity and the "bluish" aerosol in the vicinity of the visible clouds has been attributed to the Rayleigh scattering of the radiation reflected by the cloud; 141 that is ultimately a three-dimensional effect (Eck et al., 2014; Kassianov et al., 2010; 142 Marshak et al., 2008; Várnai and Marshak, 2009; Wen et al., 2008). They have also studied 143 144 the transition region from satellite measurements and stated its important radiative effects (Várnai and Marshak, 2011) and have explored the combination of data from two different 145 146 satellites with the goal of obtaining detailed information on aerosols near clouds (Várnai and Marshak, 2012). Recently, Ten Hoeve and Augustine (2016) confirmed from ground-based 147 and satellite measurements that the aerosol optical depth increases in the vicinity of a cloud; 148 Jeong and Li (2010) had previously found that aerosol humidification effects could explain 149 one fourth of a reported correlation between cloud cover and aerosol optical depth. Moreover, 150 Chiu et al. (2009) and Marshak et al. (2009) addressed the description of the continuum from 151 152 measurements of zenith spectral radiance in the visible and near infrared, with the high temporal resolution that the rapid transitions between cloud and clear sky require. Chiu et al. 153 (2010) successfully replicated these results by radiative modeling. 154

The goal of the current paper is to quantify the importance and frequency of situations where 155 ambiguity between clouds and aerosol occur; in other words, situations where the suspension 156 of particles depend on subjective definition to be classified as either cloud or aerosol. These 157 transition situations populate the continuum between what is clearly a cloud and what is to be 158 called undoubtedly an aerosol. We realize that such quantification depends both on the 159 instrument or technique used for observing the sky, and on the climate and geographical 160 conditions of the site. Therefore, several ground-based, passive observing techniques are 161 considered: specifically, sensitivity analyses are applied on sky camera images, broadband 162

radiation measurements, and spectral measurements. Two sites are considered: Girona(Spain), and Boulder (Co, USA).

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2. Data, measurements, and observations

166 The University of Girona has maintained a radiometric and meteorological station since the early 1990s. Instruments are placed on the roof of a university building (41°58'N, 2°50'E, 110 167 m asl). The site is located in the northeast of the Iberian Peninsula, some 30 km from the 168 Mediterranean Sea and 40 km from the Pyrenees mountain range; the climate is 169 Mediterranean, meaning mild winters and hot summers, and relatively dry, with more rain in 170 equinoctial seasons. Characteristics of the site and of instruments of this station can be found 171 in Calbó et al. (2016); here we will only give some details of the relevant instruments used in 172 the current research. First, the site holds a set of instruments measuring downwelling 173 shortwave (solar) and longwave (atmospheric) radiation, including the three components for 174 the solar irradiance (global, direct, and diffuse). These instruments strive to adhere to the 175 specifications of the Baseline Surface Radiation Network (BSRN) for calibration, daily 176 routine supervision, and temporal resolution of sampling and recording data. Second, 177 operation of a Multifilter Rotating Shadowband Radiometer (MFRSR) began at the site in 178 2012. This instrument is oriented towards characterizing the atmospheric aerosol, specifically 179 its optical depth (AOD) at several visible and near infrared wavelengths and is described 180 elsewhere (Harrison et al., 1994; Sanchez-Romero et al., 2016). The MFRSR measures both 181 global and diffuse solar radiation in one broadband and six narrow bands of the solar 182 spectrum (specifically the narrow band filters are centered nominally at 400, 500, 615, 670, 183 870, and 940 nm). The MFRSR at Girona sampled at 1 minute time step until September 184 2014; since October 2014 it has sampled at 15 second intervals. Third, a whole sky camera is 185 186 used to take images of the sky during daylight hours, at 1 minute time steps. The camera is a 187 conventional digital CCD camera, provided with a fish-eye (i.e. >180° field-of-view) lens and mounted on a sun tracker, in such a way that a black sphere projects its shadow on the lens, 188 blocking the direct sun from entering the camera. In the current research, one year (2014) of 189 data and observations from each of these instruments will be analyzed. 190

The Surface Radiation Budget Network (SURFRAD) was established in 1993 through the 191 support of the National Oceanic and Atmospheric Administration (NOAA) Office of Global 192 193 Programs. Its primary objective is to support climate research with accurate, continuous, long-term measurements of the surface radiation budget over the United States. Thus, 194 currently seven SURFRAD stations are operating across the US (Augustine et al., 2005, 195 2000). Here we will use data and observations from one site, which is the NOAA-Earth 196 System Research Laboratory Test Facility at Table Mountain, located 13 km north of Boulder 197 (40°7'N, 105°14'W, 1689 m asl). The instruments used here are part of a larger set 198 maintained at this location and used for annual intercomparisons and other research. 199 Radiation measurements at SURFRAD stations cover the range of the electromagnetic 200 spectrum that affects the earth/atmosphere system. Like in Girona, total downwelling (global) 201 202 solar radiation is measured by an upward looking broadband pyranometer, the direct component is monitored with a normal incidence pyrheliometer mounted on an automatic sun 203 tracker, the diffuse component is measured by a shaded pyranometer that rides on the same 204

tracker, and an upward looking pyrgeometer measures longwave (thermal infrared) radiation
emitted downward by clouds and other atmospheric constituents. In addition, a third
pyranometer and another pyrgeometer are mounted facing downward, on cross arms atop of a
10-meter tower to measure solar radiation reflected from the surface and upwelling long
wave radiation respectively. Similar to Girona, the SURFRAD suite of instruments includes
an MFRSR. A sky camera of the model TSI (Total Sky Imager) from Yankee Environmental
Systems (YES) takes sky images at 1-minute time steps.

The two locations are middle-latitude, Northern Hemisphere sites. However, they hold some 212 geographic and climatic differences that make pertinent the use of data from both sites in the 213 current research. First, Girona is at low altitude and close to the sea, while Boulder is at high 214 215 altitude and thousands of km away from the closest coast. Therefore, climate in Boulder is much more continental, in the sense that warmer summers and colder winters are likely; more 216 important here is that the atmosphere above Boulder is in general drier and cleaner, so 217 different cloudiness regimes and lower aerosol load are expected. Dominant aerosol types at 218 219 the two sites are likely different too, with maritime aerosol and Saharan dust relatively usual at Girona, and continental dust and wildfire smoke more common at Boulder." 220

3. Methods

222 Raw measurements and observations from the above instruments need to be processed in order to obtain quantitative or qualitative information about the sky condition, clouds and/or 223 aerosol. In all processing and algorithms used (and explained below) decisions must be taken 224 to distinguish between clear sky (either clean or with a certain aerosol load) and clouds, or 225 226 between clouds and aerosol. These decisions usually take the form of thresholds, which are somewhat subjectively selected after some tuning procedure. Sometimes, the human 227 intervention is obvious, for example when deciding which sky images are considered as 228 cloudless references. In the next paragraphs, we will explain the standard methods applied to 229 230 raw data, and will describe the sensitivity analyses that we have performed on them to reach 231 our goal.

Broadband solar radiation measurements at high temporal resolution (< 5 minutes) can be 232 used to infer the sky conditions. In this regard, after some initial attempts (Calbó et al., 2001; 233 Duchon and O'Malley, 1999) a Radiative Flux Analysis (RadFlux) technique was developed 234 235 to provide quantitative sky cover characteristics (Long et al., 2006a; Long and Ackerman, 2000) and is currently quite broadly used. Updated versions of this methodology also make 236 use of longwave irradiance measurements (Long and Turner, 2008) and of other 237 meteorological variables; here we will focus, however, on the use of solar radiation 238 measurements. In summary, the method comprises two steps. The first step (Long and 239 Ackerman, 2000) consists in identifying clear (i.e., totally cloudless) instances within a time 240 series of solar radiation data. Several conditions must be met: primarily global irradiance 241 must be between certain limits, diffuse irradiance must be less than an imposed threshold, 242 global irradiance must show low variability, and the ratio of diffuse to global solar irradiance 243 must also have low variability. These conditions are applied to "normalized" values (i.e., the 244 cosine and the longer path effects due to changing Sun's position are removed). On the basis 245

of the identified clear sky periods within the time series, empirically adjusted clear-sky estimated values of global, direct, and diffuse irradiances are computed for the whole time series, regardless of sky condition. In the second step (Long et al., 2006a) the daylight fractional sky cover (*fsc*) is estimated based upon the "diffuse cloud effect," i.e., the ratio of the difference between measured diffuse irradiance and the estimated clear-sky diffuse irradiance normalized by the estimated clear-sky global irradiance. It should be noted that other conditions are also applied to identify totally overcast and totally cloudless instances.

Although several empirically or subjectively given parameters are used within these 253 algorithms, the most relevant one regarding the differentiation between thin clouds, aerosols, 254 and a clean sky is the maximum diffuse irradiance that is admissible for clear skies. The 255 diffuse irradiance is produced by scattering, thus this setting limits how much scattering is 256 allowed for skies to be classified as "clear." Indeed if the Max_Diff parameter is set to a very 257 low value, such as that for Rayleigh (molecular) scattering in the atmosphere, it is likely that 258 very few instances would be identified as clear. Similarly, when estimating cloud cover, cases 259 260 with heavy aerosol loading that produces large amounts of diffuse irradiance will be considered as cloudy situations. If it is fixed to a too high value, cases with thin clouds may 261 be considered clear. In the original paper, Long and Ackerman (2000) suggested a value 262 around 120-150 W m⁻². Below we analyze the effect of changing this threshold on clear sky 263 determination and fractional sky cover estimation. 264

As its name indicates, the MFRSR uses a rotating shadowband to consecutively shade and 265 unshade the detector. In this way, direct (beam) radiation on the horizontal surface of the 266 detector is estimated by subtracting the shaded diffuse measurement from the corresponding 267 unshaded global measurement. These inferred beam measurements, if their value is greater 268 269 than zero, are then processed for aerosol optical depth. As explained in many papers (Harrison and Michalsky, 1994; Michalsky et al., 2010; Sanchez-Romero et al., 2016), this 270 process has several steps, including continuous Langley calibration of the spectral sensors, 271 evaluation of total optical depth, subtraction of Rayleigh and ozone extinction, and -more 272 importantly here- cloud screening. Thus, after the first three steps, an optical depth (OD, that 273 might be due to water droplets or ice crystals, or aerosol particles, or both) for each 274 wavelength but the longest (940 nm), which is affected by water vapor absorption, is 275 computed. Using two of these values (OD at 500 and 870 nm) the Ångström exponent, that 276 accounts for the wavelength variation of OD and varies inversely with the particle size, is 277 278 also estimated. These values then must go through scrutiny to distinguish those that actually 279 correspond to the effect of aerosols in the atmosphere from those which are affected by 280 (necessarily) thin clouds. Obviously, a number of cases correspond to situations that cannot be clearly identified as either clouds or aerosol, but in general, the cloud screening procedures 281 routinely implemented are conservative in the sense that they tend to guarantee that the 282 filtered cases are free of "cloud contamination." 283

There are in the literature several suggestions for cloud screening the OD data from MFRSR. The SURFRAD network applies the technique described in Augustine et al. (2008) that is a hybrid of the cloud screening methods of Michalsky et al. (2010) and Alexandrov et al. (2004). As in the case of identifying clear skies from solar radiation data, the variability of 288 the measurement is the basis of all methods: the underlying assumption is that clouds make solar radiation (either broadband or spectral) more variable in time than aerosols. Here we 289 will use the methodology as presented by Michalsky et al. (2010), which consists of two 290 filters applied consecutively on a moving time window of a given width (10 minutes in the 291 original paper). The first, coarser filter takes the difference between each adjacent 292 measurement, and also calculates the maximum minus the minimum OD in the window. If all 293 differences are less than a given threshold, and if the range of measured OD within the time 294 window is less than another threshold, then the points pass the first filter. The second, more 295 stringent filter scales the allowed variability according to the magnitude of the OD, which is 296 estimated by applying a low-pass filter on the series. Thus, the absolute value of the largest 297 difference between adjacent data must be less than a given fraction of the estimated OD at the 298 midpoint of the sample window, and the range must be less than another fraction of the same 299 estimate. The values of the four thresholds were 0.02 and 0.03 (absolute differences of OD at 300 550 nm) and 10 and 20% respectively in the original paper. In the present study, we will 301 change these four values, and also the time window where differences and ranges are 302 calculated, to assess their effect regarding "transition" cases. The final result of the MFRSR 303 cloud screening is every sample tagged as "good" or "bad," meaning that can be 304 representative of aerosols or not. In the current paper, we will assume that samples labeled as 305 "bad" correspond to the presence of some kind of clouds. 306

307 As mentioned above, images of the whole sky are becoming more ubiquitous both in atmospheric research and in solar energy management applications. Automatically captured 308 sky images allow a continuous (many such cameras take images every minute or even more 309 often) visible record of the sky. Images can then be visually scrutinized to identify clouds, 310 aerosols, rain, and any other meteors. Usually, however, an automatic digital image analysis 311 is set to obtain an estimation of the fractional sky cover and (sometimes) other sky or cloud 312 313 characteristics. In the present research, one year of 1-minute sky images taken at Girona have been visually inspected and tagged according the presence of situations where the distinction 314 between clouds and aerosols is unclear. In addition, one year of 1-minute images taken by the 315 TSI at Table Mountain have undergone digital processing to account for the fractions of sky 316 that is free of cloudiness, or covered by optically thin and thick clouds. The basis of the 317 distinction is the ratio between the red and the blue intensities (R/B) in the RGB image: a 318 319 clear (blue) pixel has a low value of R/B, while a cloud (white) pixel has a greater value. In this case, the process uses mainly two R/B thresholds referenced to the baseline clear-sky 320 threshold set by the manufacturer to make these distinctions: one to distinguish between clear 321 sky and thin cloud, a second one to separate between thin and thick clouds. As explained 322 elsewhere (Long, 2010; Long et al., 2006b), a post-image-processing algorithm addresses 323 issues in regions of particular difficulty (a circle around the Sun, the region close the horizon 324 in the direction of the Sun), which occur due to forward scattering of visible light by aerosols 325 and haze, and the intensity range limitations of the detectors of the cameras used to record the 326 sky images. But in our analysis, we have focused mainly on the threshold that affects the 327 distinction between clear sky (with more or less aerosol content) and thin clouds. 328

4. Results

4.1 Clear sky detection and cloud cover estimation from radiation flux analysis

RadFlux analysis results for Girona, 2014 are presented below. The analysis was applied 332 twice, with values of Max_Diff equal to 100 and 200 W m⁻². We also checked the value of 333 300 W m⁻², but those results were nearly the same as those for the 200 W m⁻² setting, 334 indicating that when such high values of diffuse radiation are in principle set to correspond to 335 clear sky, other tests for clear-sky detection filter out these cases anyway. It should be noted 336 that even with the lower threshold, the diffuse irradiance allowed as "clear sky" is well above 337 the Rayleigh limit, i.e., a certain amount of scattering particles larger than molecular is 338 always allowed. A summary of results is presented in Table 1. There are almost 11,000 339 minutes identified as clear when the higher threshold is used but labeled as not clear when the 340 lower threshold is applied. This means that almost 5% of the daylight hours (specifically, of 341 342 the time when the Sun is more than 10° above the horizon, a condition for the RadFlux algorithm to identify clear skies), may be considered clear or not depending on where we set 343 the maximum allowed diffuse limit. These situations often correspond to what can be 344 generically called "haze," that is conditions that can hardly be classified as either cloud or dry 345 aerosol. For Boulder, results are similar. More than 15,000 minutes (7.5%) are considered 346 clear or not depending on the Max_Diff value (here the two tested values were 100 and 180 347 $W m^{-2}$). 348

The difference in mean *fsc* when data are processed with one or the other threshold is 0.023 349 (0.022) in Girona (Boulder). This difference might not seem very large, but, as we will show 350 351 below, it is produced by larger differences for some particular conditions. Thus, differences between the mean *fsc* when one or the other clear sky reference is used (i.e. based on clear 352 sky periods identified by using one or the other Max_Diff value) increase significantly when 353 cloudless and overcast conditions are not considered. Indeed (see Table 1), for scattered to 354 broken cloud conditions, the average difference is 0.044 (0.046), which is more than 10% of 355 the average *fsc* of about 0.4 at both sites. Logically, since RadFlux uses the difference 356 between measured and estimated clear-sky diffuse as the basis for *fsc* estimation, estimated 357 fsc tends to be lower when Max_Diff is greater. In absolute value, differences tend to be 358 greater for lower *fsc* (see Figure 1 corresponding to Girona data). Table 2 summarizes the 359 number of records with significant differences in *fsc*. It is worth noting that for about 15,000 360 (17,000) minutes the difference is greater than 0.10. This means that in about 14% (17%) of 361 362 non-cloudless and non-overcast cases, the average difference of *fsc* is a relatively high value of 0.16 (0.15). 363

We will next present the evolution of solar radiation and results of RadFlux (and also from MFRSR) for some particular days, as examples of the behavior of the algorithm with regard to the objectives of the present research. The mean values of *fsc* for these days when using the two Max_Diff values are shown in Fig. 1. Figure 2 presents in detail measurements and processed results for two days that, based on visual inspection of the images (see Figure 3) and the RadFlux with the greater threshold, are considered to be clear. March 6, 2014, is an example of a cloudless and very clean atmosphere day. Measured diffuse and direct

(projected onto a horizontal surface) irradiances match almost exactly their clear sky 371 estimates. It should be noted that for this day, no significant difference exists between the 372 estimate of clear sky diffuse irradiance after using one or the other threshold of Max_Diff. As 373 a consequence, for this day, the estimate of *fsc* is zero, whatever the threshold used. 374 Consistently, the MFRSR cloud screening algorithm flagged almost all minutes as "good" 375 376 (i.e., not contaminated by clouds), and the OD at 500 nm is less than 0.1 for the whole day, while the Ångström exponent (AE) evolves between 1 and 2, indicating relatively small 377 particles. For June 21, 2014, both diffuse and direct irradiances follow a relatively smooth 378 evolution that matches the clear sky estimates quite well. For diffuse, this is true if the clear 379 sky estimate corresponds to the higher threshold. Therefore, if the higher Max_Diff is used, 380 the RadFlux produces an almost cloudless day (*fsc* is very close to zero, with a daily average 381 of 0.02). In contrast, when a Max_Diff = 100 W m^{-2} is used, not a single minute is considered 382 clear, and the *fsc* is estimated to be between 0.2 and 0.4 (daily mean, 0.27). As mentioned 383 above, however, there are no "visible" clouds in the sky, and in fact, the MFRSR cloud 384 screening algorithm labels all records of this day as "good." Nevertheless, the AOD for this 385 day is relatively high (0.15-0.30) and, more significantly, the AE is low (0.4-0.9) indicating, 386 as suspected, larger (and/or hydrated) particles. 387

Two particular cases of days with variable cloudiness are presented in Figure 4: one presents almost exactly equal *fsc* estimations, while the other presents notable *fsc* differences between the two estimations. As can be seen in the corresponding sky images (Figure 5), the first day (April 24) shows middle and high clouds, and low aerosol load, which is confirmed by the MFRSR OD data: for the times that pass the cloud screening, OD is around 0.15 and AE is around 1.5. But the second day (June 23) has low level clouds in a more "whitish" sky, which results from a greater OD (0.25) and lower AE (< 1).

395 4.2 MFRSR estimations of OD and AE, and cloud screening

Figure 6 shows the behavior of the Ångström exponent versus the optical depth at 500 nm, 396 for all instantaneous measurements that are processed by the MFRSR algorithm, that is for all 397 instances when there is some direct irradiance and the solar zenith angle is less than 80°. 398 These are measurements from Girona for the year 2014. The whole unscreened dataset of 399 measurements are represented by black circles. It should be noted that plotted is the optical 400 depth of whatever is in the atmosphere (other than well-mixed gases and ozone) as we don't 401 know, in principle, if this optical depth is due to dry particles, water droplets or ice crystals. 402 There are many points with negative values of AE, which must be the result of the 403 uncertainties of the method to estimate AE. As shown by Sanchez-Romero et al. (2016), the 404 405 uncertainty attached to AE computation is of the order of 0.5 for OD values of about 0.1, and greater for lower OD values. Therefore, when the true value of AE should be close to 0, the 406 random error associated with this uncertainty may result in negative values. A deeper analysis 407 of the effect of OD measurement errors on the computation of AE that explains how negative 408 AE values may be obtained, was developed by Wagner and Silva (2008). Their results 409 410 discourage the computation of AE when OD is less than 0.15, given the typical uncertainties of the current instrumentation. That same study shows that the error in the AE estimation may 411 412 be skewed depending on the relative errors of each of the monochromatic ODs used.

The data points highlighted in red in Fig. 6 are all those that have passed the cloud screening 413 filter (i.e., labeled as "good"). Initially (Fig. 6a) we used the same time window and threshold 414 values for cloud screening as suggested in Michalsky et al. (2010). Subsequently, we changed 415 these values as summarized in Table 3. Changing the time window means that the filter 416 would take into consideration the variability of the computed OD over a shorter or a longer 417 period. Changing the other thresholds would mean that the filter accepts aerosol conditions 418 that show more or less variability. With the default cloud screening, 42% of points are 419 considered representative of atmospheric aerosol. Most of these points have OD < 0.5 and 420 AE between 0 and 2. The sensitivity analysis showed that shortening the time window has an 421 important effect of allowing more points to be considered aerosol. In contrast, lowering the 422 thresholds corresponding to relative values of differences and range substantially reduces the 423 number of points that pass the filter. We combined a shorter time window and higher values 424 of the thresholds to apply a "relaxed" cloud screening or conversely a longer time window 425 and lower thresholds to produce a more "strict" filter. When the former is applied, almost 426 58% of points are considered aerosol (Fig. 5b), but when the latter is used, less than 19% of 427 the points pass the filter (Fig. 5c). With very few exceptions, all points with OD > 1.0 and 428 429 most points with negative AE (and OD > 0.1) do not pass the cloud screening even with the relaxed filter. 430

431 The numbers in Table 3 allow an estimation of the frequency of transition cases between 432 cloud and "pure" aerosol. We start with about 420,000 instantaneous measurements for Girona performed by the MFRSR with solar zenith angle less than 80° (it should be noted 433 that this number is a combination of 1-min measurements during 9 months plus 15-sec 434 measurements during 3 months). From these, about 212,000 (50.5%) are not processed by the 435 436 MFRSR, due to the presence of clouds thick enough to occult the Sun and to preclude obtaining valid optical depth results. From the rest of the samples (208,000) about 88,000 437 438 additional points must be considered clouds, as even the most relaxed cloud screening labels them as "bad." Therefore, in about 70% of instances, there are clouds (either optically thick 439 440 or thin) in front of the Sun. This "cloud occurrence" might be eventually an estimation of the mean cloudiness, but it should be noted that only clouds that are in front of the Sun from the 441 perspective of the MFRSR are considered by the cloud screening algorithm, so it is not a 442 hemispheric view of the sky. In other words, whereas the clear sky identification from the 443 444 RadFlux refers to the whole hemispheric sky view, the cloud screening for the MFRSR refers only to the direct sun, not the whole sky. From the rest of the samples, we can almost assure 445 (according to the result of applying the "strict" cloud screening) that about 39,000 data points 446 correspond to clear sky (in the direction of the Sun beam) affected by a certain amount of 447 aerosols. This means that about 81,000 measurements (19% of the initial number) are 448 affected by particles in the atmosphere that are problematic in being classified as aerosol or 449 as cloud, at least based on the cloud screening applied which is built upon the assumption that 450 clouds induce higher variability than aerosols in the measured irradiances. 451

The discussion above concerns results from Girona. When the same analysis is applied to measurements from Boulder, the numbers obviously change, but not the main result of a large percentage of cases in the transition zone. We started with 610,000 instantaneous

measurements (note that 20-sec resolution was used in Boulder for the whole year) from 455 which about 158,000 (25%) were not processed by the MFRSR, due to thick clouds occulting 456 the Sun. Then we applied the three cloud screenings (default, relaxed, strict) to the rest of the 457 samples (452,000, see Table 3). About 242,000 additional points were labeled as "bad" (i.e. 458 clouds) by the relaxed cloud screening, so in a total of 66% of instances there are clouds 459 before the Sun (note the important difference between the number of thick and thin clouds as 460 compared with Girona). From the remaining data, about 37,000 data points correspond to a 461 cloudless sky affected by aerosols. This means that about 173,000 measurements (28% of the 462 initial total) correspond to the kind of particle suspension that represents the transition region 463 between aerosols and clouds, at least regarding the observations by the MFRSR, which are 464 mostly affected by what is present in the direction of the Sun. 465

Again for Girona and for the four days analyzed in Figs. 2 and 4, Figure 7 presents the 466 periods that passed the MFRSR cloud screening (i.e., the calculated OD is considered to be 467 due to aerosols) for the three different settings (default, strict, relaxed). The first two days are 468 469 considered cloudless by the default screening (and obviously, by the relaxed one), and also mostly cloudless by the strict screening. Somewhat surprisingly, a period of the first day 470 (10:00-11:30 approximately) is filtered out by the strict screening, owing to a relatively high 471 variability of the signal despite that sky images and the RadFlux analysis indicate that this is 472 a totally cloud free, clean day. Conversely, the second day raises some doubts about what it is 473 474 in the air according to images and RadFlux, but is considered almost totally cloudless even by the strict MFRSR screening. 475

With all this in mind, we find that the variability of the optical depth (which is intended to 476 reflect the variability in the atmosphere, although we cannot rule out some contribution from 477 478 the instrument's noise when the signal is very low) is not sufficient to discriminate between cloud and aerosol. The optical depth itself and its wavelength dependence (AE) do not help 479 very much in this distinction (despite that Kassianov et al. 2013 suggested a method that 480 worked well for their Arctic site). Figure 8 shows the histograms of the populations of points 481 screened out as clouds (this is for Girona, so 88,000 points), points that passed the strict filter 482 so they are considered aerosol (39,000), and points that belong to the transitional zone 483 (81,000), as a function of OD and AE. It is obvious that the filter does a good job as the 484 dominant OD and AE values for clouds and aerosols respectively are quite different among 485 them (despite some data with large AE is labeled as cloud, and other minor inconsistencies 486 are found as well). But it is also apparent that the third group of points presents values of OD 487 and AE that cover a very broad range. Even if we assume that all points with OD < 0.032 or 488 AE > 1.6 must be considered aerosols, and all points with OD > 0.32 or AE < 0.15 must be 489 considered clouds (these values derived from the percentiles 1 and 99 of the distributions of 490 cloud and aerosol points), there are still about 58,900 points within the range of OD [0.032-491 0.32] and AE [0.15 - 1.6] for which their corresponding variability is not either high enough 492 to be considered clouds nor low enough to be considered aerosols without some doubts. This 493 number represents 14% of the initial measurements and 28% of the instances processed by 494 the MFRSR. The corresponding relative values for Boulder data are 10.7% and 14.5%. It 495 should be noted that the previous thresholds for OD and AE, based upon the statistical 496

distributions of our data, must not be taken for general application to all atmospheric
situations. For example, Antón et al. (2012) found a very large OD (0.8-1.5) and very low AE
(0.1-0.25) under a strong Saharan dust event.

500 *4.3 Images from sky cameras*

For Girona, there are about 200,000 total sky images corresponding to 341 days during the 501 year 2014. The most significant gap in the series of sky images corresponds to the first 502 fourteen days of the year (there are also some missing days in April and June). Since visually 503 inspecting each and every one of these images is unfeasible, we built a movie for each day by 504 displaying all images successively. Watching these movies is much more practical, and 505 allows identifying the most important features in the images. Once some particularly 506 interesting days or periods within a day were selected, the individual images were visually 507 inspected in more detail. Some examples of these images have already been used to discuss 508 previous results (see Fig. 3 and Fig. 5). Overall, there is a high percentage of days when we 509 have detected some period (that may last less than one hour or may last the whole daylight 510 time) of challenging distinction between clouds and aerosols (40% of days). Indeed, most of 511 the other days (those without doubtful periods) correspond to mostly overcast days, or, 512 exceptionally, to unusually clear days. Regarding the challenging periods, there are a few 513 different situations. First are relatively clean and apparently cloudless skies where a kind of a 514 thin "veil" can be seen in the images, without an analyst being able to subjectively discern 515 whether it is made up of dry aerosols or extremely thin clouds (either low or high). Second 516 517 are hazy or foggy skies with corresponding difficulties in affirming what kind of suspension is causing the haziness. Third are skies with scattered or broken clouds (typically, cumulus) 518 that have poorly defined boundaries or are continuously forming and vanishing, which makes 519 520 it difficult to decide whether a portion of the sky is cloudless or not.

We have been able to perform a more quantitative analysis on TSI images from the Boulder 521 site. In 2014, there are 217 days with images every 1 minute, which translates to 145,000 522 523 images being processed. The missing periods correspond to most of January, April, and July, with some additional missing days randomly distributed across the other months. Main 524 reasons for missing images are a malfunctioning camera or rotating mirror, but sometimes the 525 outages are due to inclement weather. The original processing of the images produced an 526 average cloud fraction of 0.62 of which 0.13 is considered thin cloud (again, in average for 527 all images). These numbers are the result of using the SURFRAD operator settings in the TSI 528 529 image processing for the Table Mountain site, i.e. a nominal R/B threshold between clear sky and thin clouds of 0.30 and an effective threshold between thin and thick clouds of 0.57. To 530 test the sensitivity of the cloud fraction results to the first threshold, we first lowered it to 531 0.20, and then raised it to 0.40. This produced, respectively, an increase in average fsc to 532 0.68, and a decrease to 0.57, mainly due to the change of the amount of thin clouds (since we 533 kept the thin to thick cloud threshold the same). It should be noted that the range of total 534 cloud fraction from TSI estimates (0.57-0.68) is relatively close to the estimate from the 535 536 Rad Flux (0.54-0.56) and to the cloud occurrence found by the MFRSR (0.66), with the difference likely mostly related to the circumsolar and near horizon issues described in Long 537

(2010) and Long et al. (2006b), besides different field of view (for MFRSR) and slightlydifferent periods (due to missing data or images).

More important than these aggregate numbers is to look at the particular cases with large (or 540 small) cloud fraction changes when the threshold is changed. In this sense, Table 4 shows the 541 differences in thin cloud fraction between the original processing and new processing using 542 two modified thresholds. For the case of the reduced threshold, the thin cloud fraction 543 estimate in almost 80% of images increases by less than 0.10. This of course includes those 544 images that presented a very high cloud fraction (overcast, or almost overcast), but there are 545 also a large number of images for which the change is very small independently of their 546 cloudiness. This illustrates the robustness of the digital image processing of TSI images, i.e., 547 it shows that even if the "correct" threshold is unknown, the cloud fraction estimate is, in 548 general, quite consistent. Figure 9a presents an example of these cases: a totally cloudless 549 image, which is considered as such even by the lower threshold used. Similarly, Fig. 9b 550 shows a broken cloud sky, where again changing the threshold does not affect the result as 551 552 the clouds have very well defined limits and the sky is very clean (blue). On the other extreme, more than 20% of images exhibit a change in the estimate of the thin cloud fraction 553 greater than 0.10 with the changing thresholds. It should be noted that this increase is not 554 always reflected in an equivalent increase of the total cloud fraction, since the sophisticated 555 algorithm that sets the particular thresholds that are used in each region of each image may 556 result in slight changes of the opaque cloud fraction too. Fig. 9c shows a sample of an image 557 where the change in the threshold produces a large change in the thin cloud fraction, because 558 a large part of that image is made up of what seems very thin clouds. In the example of Fig. 559 9d, the large differences seem to be related to a relatively high atmospheric aerosol load. 560

For the case of the increased threshold, the thin cloud fraction estimate in a little more than 561 562 80% of images decreases by less than 0.10 (Table 4). This includes a) some situations of cloudless skies, b) situations with scattered to broken cloudiness but with a low amount of 563 thin clouds (in these two cases, the increase of the threshold of course makes it impossible to 564 get lower cloud fractions), and c) situations of overcast skies with thick clouds, that present 565 much higher values of the red to blue ratio (or that are set as cloudy because of very low light 566 intensity, i.e., very thick clouds). Again, this result confirms that the method is quite robust, 567 and also that almost 20% of time a change in the threshold produces a change in the thin 568 cloud fraction of more than 0.1. In Figures 9a-f the result of the cloud identification with the 569 higher threshold is also displayed, and we can see the moderate effect on cases of Fig. 9c and 570 9d, corresponding to clouds (or aerosols) with not well defined limits and that are mainly 571 visible when they are in front of the Sun due to their forward scattering characteristics. The 572 greatest effect of changing the threshold is found in situations such as those presented in Fig. 573 9e and 9f, where the hazy atmosphere (involving cumulus clouds formation) is too 574 problematic to be classified as "clear" or "cloudy" with thin or even opaque clouds by the 575 method applied to TSI raw images. It should be noted that in the cases where the effect of 576 changing the threshold is small (Fig. 9a and 9b), the optical depth as measured by the 577 MFRSR was also very low (around 0.02), while for the rest of cases (Fig. 9c-f) the optical 578 depth (in the nearest periods when is available) is always greater than 0.1 and in fact, close to 579

580 0.2, which is the value that Dupont et al. (2008) found as the limit related to the more 581 common differentiation between cloud and not cloud.

582

5. Discussion, summary and conclusion

583 We have presented observations from three ground based, passive systems, that are intended to detect clouds and aerosols in the atmosphere. Indeed the three systems share one 584 characteristic, which is that they are sensitive to the solar radiation flux once it has been 585 modified (affected) by the presence of suspended particles in the air (of course, solar 586 radiation flux is also affected by atmospheric gases). Thus, sky cameras "map" radiation 587 coming from the whole sky dome and record this radiation in three color channels (red, 588 green, blue). The presence in the intervening atmosphere of particles (whether in the form of 589 clouds or aerosol) modifies the aspect of the sky, that is the partitioning of light 590 corresponding to each color. Broadband radiometers perceive the effect of clouds and 591 aerosols as these modify the total amount of solar radiation reaching the ground, and/or the 592 partitioning of radiation between the direct and diffuse components. Finally, the MFRSR 593 determines the effect on the direct solar beam so it can estimate the optical depth resulting 594 from the absorption and scattering (i.e. attenuation) phenomena associated with the 595 suspended particles. Since the latter is a spectral instrument, it can be used to evaluate optical 596 depths for several wavelengths and therefore, the variation of the optical depth with 597 wavelength, which is related with the size and characteristics of the particles. 598

The general conclusion of the analyses performed here is that the number of challenging 599 cases where distinction between aerosol and cloud is nebulous (in fact, only subjectively 600 601 resolved) is not negligible, for at least the two mid-latitude sites analyzed and for the instruments used. A very rough estimate of the frequency of instances when the mentioned 602 cloud-aerosol distinction is challenging is a figure not less than 10%. This number includes 603 those cases that are cloudless in principle, but with a very thin veil in the sky that is hardly 604 visible but affects the partitioning of the solar radiation in the diffuse and direct components 605 (5 and 7.5% of time according to RadFlux in Girona and Boulder respectively, Section 4.1). 606 This gross estimate also includes cases with scattered and broken clouds, when the cloud 607 limits are not very definite, or when there is a suspension that somewhat attenuates the direct 608 beam in the "clear" patches between clouds. According with the MFRSR datasets and applied 609 analyses, the lowest estimates of these situations at Boulder and Girona respectively are 10.7 610 and 14% (Section 4.2). In all these cases the spectral signature of solar radiation is also 611 affected, which is reflected in the computation of the Ångström exponent. These numbers are 612 also in agreement with what we find from the sky images. The sensitivity analysis performed 613 showed that changing the threshold that distinguishes clear sky and thin cloud, which is 614 essentially how "whitish" the cloud-free sky (includes cases with relatively high aerosol) is 615 allowed to be, produces large differences (in the "thin" cloud fraction) a significant amount 616 of time. Specifically, we find a change in the thin cloud fraction of more than 0.1 in about 617 618 20% of images (Section 4.3). This latter result is for Boulder, but for Girona, a visual 619 inspection of images suggests similar results.

620 These values are quite in agreement with the few previous numbers given in the literature, although they are not directly comparable. Indeed, Koren et al. (2007) estimated that between 621 30 and 60% of the part of the globe at any given time considered to be free of clouds could 622 correspond to the challenging characteristics transitioning between clouds and aerosols 623 (which they called "twilight zone"). Similarly, for a particular area of the Atlantic Ocean, 624 625 Bar-Or et al. (2010) found a value of about 35-45%. In these latter studies, a spatial approach was considered, i.e., they accounted for the extension of this zone in a snapshot of the sky. 626 Our study, however, combines this approach (for sky camera images and partially for 627 broadband hemispheric solar radiation measurements) with a temporal approach, that is 628 accounting how often the atmosphere presents a state that cannot be distinctly categorized as 629 cloud or as aerosol (for broadband hemispheric radiation measurements and also for MFRSR, 630 Sun pointing, measurements). Therefore, our numbers correspond mainly to temporal 631 frequencies, are limited to two particular sites, and are quite conservative, but if we discard 632 the overcast conditions, the relative frequency of the transition cases increases to more than 633 15% of the remaining cases (this number is estimated by dividing the above overall value of 634 10% by the frequency of non-overcast cases, which is about 70% at the two involved sites). 635

Our results support the argument that clouds and aerosol are two extreme manifestations of 636 the same physical phenomenon, which is a suspension of particles in the atmosphere. This 637 reasoning was already suggested by other authors (e.g., Charlson et al. 2007). This of course 638 could be questioned, as the origin and the many processes involved differ quite a lot. On the 639 one hand, processes involved in producing different types of aerosols such as sea salt spray, 640 secondary pollutant particles, forest fire smoke, and desert dust, may be quite different. Also 641 different are processes leading to the formation of different types of clouds: for example 642 643 maritime thin stratus, cumulonimbus, or high ice clouds. Therefore, we can conclude that a suspension of particles in the air may have many different origins, compositions, properties, 644 645 etc., but, despite their differences of origin and composition, their radiative effect as seen from the ground does not easily distinguish the type of suspension. In particular, at times we 646 647 cannot classify the suspension in either of the two most usual cases: clouds (relatively large condensed water particles) and aerosols (relatively small dry particles). Difficulties in 648 separating these cases may result from the fact that the instruments have a hemispheric view 649 of the sky (pyranometers, cameras) which may integrate radiation fluxes (radiances) that, 650 separately, could be distinctly affected by clouds or aerosols. Or even in the case of 651 directional measurements (such as the MFRSR measurements), the solar beam attenuation 652 that is obtained is the result of the light crossing through different layers of the atmosphere 653 which may contain a cloud at some height and suspended aerosols at other heights. 654

Many cases may correspond to a situation where the particles in the suspension show characteristics (size and composition) that are somewhere in between what is typically considered cloud and aerosol. In these situations, it might not make sense trying to classify the suspension as a cloud or aerosol. Rather, it might be more convenient to treat the phenomena as a continuum, of which the extremes would be the entities corresponding to the usual definitions of cloud and aerosol, but with a non-negligible number of intermediate conditions (Charlson et al., 2007). The two most common situations with the characteristics in the "middle" of the continuum are those with clear skies with a thin veil showing some sort
of structure, and those with partly cloudy skies with poorly defined cloud boundaries (Koren
et al., 2008, 2009).

Although other active cloud observation instruments (such as ceilometers, radars, microwave 665 radiometers) do provide more detailed information on cloud structure and particles forming 666 667 the clouds, they in fact also support the case of a continuum of properties, and of the difficulty in setting precise limits on what must be labeled as a cloud (Boers et al., 2010). 668 Similarly, the use of radiance measurements in several directions of the sky dome, as for 669 example those performed by CE-318 sunphotometers (CIMEL Electronique, France), can 670 map the presence of clouds in selected planes so helping in the detection of clouds and the 671 672 distinction from pure aerosol. The quantification of the transition situations as seen from these other instruments could be a matter of future research. 673

As for the consequences of the above conclusions and suggestions, we think that considering 674 the intermediate situations between cloud and aerosol may have a significant impact in 675 energy balance studies, either local or global, and must be taken into account when 676 parameterizing radiation transfer within weather and climate models. The point here is that 677 cloud-screening algorithms for aerosol products and cloud detection algorithms for cloud 678 products both tend to be conservative, which bias both sets of products in a way that the 679 transition area is maximally omitted from products. For example, when considering scattered 680 cloud conditions, assuming that there are, or are not, condensed particles in the patches 681 682 between clouds will produce different downward and upward radiation fluxes (both in the solar and in the thermal infrared bands), that would be different also from what is actually 683 observed by radiometers on the ground. The properties of the suspended particles that are 684 685 around or between the clouds are also relevant where radiation transfer is concerned (Schmidt 686 et al., 2009). High spatial resolution observations of cloud boundaries, such as in Schwartz et al. (2017) where cloud structure is examined on scales 3 to 5 orders of magnitude finer than 687 satellite products, or high temporal sampling of radiation spectra at cloud boundaries by using 688 array spectrometers (González et al., 2017), open new paths for examination the cloud-689 aerosol conundrum. Even if a homogeneous stratified atmosphere is assumed in a given 690 region, considering that there is a layer of a very thin cloud or a layer of aerosol particles 691 having differentiated radiative properties may also result in different estimates of the 692 radiation fluxes. If possible, it would be more realistic and accurate to treat the phenomenon 693 694 as a continuum in the radiative transfer calculations. Despite the existence of several studies that already address some of these concerns (Charlson et al. 2007; Koren et al. 2008, 2009; 695 696 Hirsch et al. 2012, 2014; Yang et al. 2016), all these subjects must be the object of further 697 research.

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Table 1. Summary of radiation flux analysis results for Girona and Boulder (Table Mountain), year 2014, when using two different thresholds for Max_Diff.

	Gir	ona	Table M	Iountain
Max_Diff	100 W m^{-2}	200 W m^{-2}	100 W m^{-2}	180 W m^{-2}
Minutes analyzed	220	,658	214,	,616
Clear-sky minutes	25,998	36,988	33,813	49,587
Average fractional cloud cover, <i>fsc</i>	0.555	0.532	0.560	0.538
Average <i>fsc</i> for non-cloudless and non-overcast cases	0.430	0.386	0.441	0.395

Table 2. Average differences between the estimated fractional sky cover ($\Delta_f sc = 891 \quad fsc_100-fsc_200$) when using the two different Max_Diff thresholds for clear sky identification,

892	only for the non-cloudless and non-overcast cases.
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	Giro	na	Table Mo	ountain
	Number of minutes	$<\Delta_fsc>$	Number of minutes	$<\Delta_fsc>$
$\Delta_fsc > 0.05$	35,906	0.11	36,626	0.11
$\Delta_fsc > 0.10$	15,158	0.16	16,905	0.15
$\Delta_fsc > 0.20$	2,765	0.29	1,574	0.24
$\Delta_fsc > 0.30$	757	0.41	106	0.36
Total with $fsc_200 \le 0.95$ and $fsc_100 \ge 0.05$	107,281		102,737	

Table 3. Sensitivity analysis of the cloud screening procedure applied to MFRSR measurements. See
Michalsky et al. (2010) for details on the method. Total number of points scrutinized, 208,259

896 (Girona), 451,793	(Table Mountain).
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	Thresholds applied				Number of cases		
	Time	Maximum	Maximum	Maximum	Maximum	passing th	ne screening
	window (min)	difference	range	relative	relative	Girona	Table
	(11111)			unierence	Tange		Mountain
Default	10	0.02	0.03	10%	20%	86,701	125,084
1	5	0.02	0.03	10%	20%	103,615	
2	15	0.02	0.03	10%	20%	75,143	
3	10	0.01	0.02	10%	20%	78,604	
4	10	0.03	0.05	10%	20%	90,782	
5	10	0.02	0.03	5%	10%	52,801	
6	10	0.02	0.03	20%	40%	98,146	
Relaxed	5	0.03	0.05	20%	40%	120,221	209,847
Strict	15	0.01	0.02	5%	10%	38,971	37,184

Range of differences	Threshold applied		
	0.20	0.40	
≤ -0.30	0.0%	1.0%	
(-0.30, -0.20]	0.0%	2.8%	
(-0.20, -0.10]	0.0%	14.6%	
(-0.10, -0.05]	0.0%	29.3%	
(-0.05, 0)	0.0%	38.7%	
= 0	6.2%	13.6%	
(0, 0.05]	36.3%	0.0%	
[0.05, 0.10)	35.7%	0.0%	
[0.10, 0.20)	16.9%	0.0%	
[0.20, 0.30)	4.3%	0.0%	
≥ 0.30	0.7%	0.0%	

Table 4. Frequency of differences in the thin cloud fraction when two different thresholds are appliedin the TSI image processing (instead of the default value of 0.30) for several values of the differences.



- Figure 1. Box-plot of the *fsc* estimated when using Max_Diff = 200 W m⁻² organized by bins of the *fsc* estimated when using Max_Diff = 100 W m⁻². Boxes indicate median and 1^{st} and
- 3rd quartiles, whiskers indicate percentiles 5 and 95. Data is from Girona. Blue circles
- indicate the mean values for days presented in Figures 2-5.





Figure 2. Two examples of "clear" days at Girona. Left, March 6, 2014; right, June 21, 2014. Top 908 panels, diffuse and direct irradiances; middle panels, estimation of fractional sky cover; bottom 909 panels, outputs from the MFRSR: OD, AE, cloud flag. 910

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Figure 3. Two sky images for each of the two days represented in Figure 2.



Figure 4. Two examples of variable cloudiness days at Girona. Left, April 24, 2014; right, June 23,
2014. Top panels, diffuse and direct irradiances; middle panels, estimation of fractional sky cover;
bottom panels, outputs from the MFRSR: OD, AE, cloud flag.





Figure 5. Two sky images for each of the two days represented in Figure 4.



Figure 6. The aerosol Ångström exponent (AE) versus the optical depth (OD) at 500 nm as result of
processing the MFRSR spectral measurements of global and diffuse solar radiation. Note the
logarithmic scale of the OD axis. Black circles, all data; red dots, data that pass the cloud screening
filter. a) Default values in the cloud screening; b) "Relaxed" values; c) More "strict" values. It should
be noted that there are points that do not pass the filter which are hidden by the red dots.



Figure 7. For the four days presented in Figures 1 and 3, periods that are considered "aerosols" by the
"default" (blue), "strict" (red), and "relaxed" (green) MFRSR cloud screening.



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Figure 8. Distribution of the points considered aerosols (38,971 points, orange bars), clouds (88,038 930 points, blue bars), and "transition" (81,250 points, purple bars) across the range of OD values (left) 931

and AE values (right), after applying the MFRSR cloud screening algorithm with different thresholds. 932

It should be noted that "clouds" refers to instances that have not passed the screening by the MFRSR. 933

934 Dashed lines indicate the (approximate) percentiles 1 and 99 of the "clouds" and "aerosols"

935 distributions.



- Figure 9. Original and processed TSI images, with different thresholds for the distinction between 936 937 clear sky and thin clouds. a) March 8, 2014, 11:00. An example of very clean day, when changing the 938 threshold does not affect the cloud fraction estimation. b) August 23, 2014, 15:00. Very white clouds in a very clean (blue) sky. Again, changing the threshold has a very minor effect on the estimation. c) 939 940 May 16, 2014, 08:14. An example with large circumsolar radiation, where lowering the threshold greatly increases the thin cloud fraction estimation. d) May 19, 2014, 09:00. A similar case, but with 941 an apparently large aerosol load. e) June 10, 2014, 10:30. Small cumulus forming in a somewhat hazy 942 943 atmosphere. The effect of changing the threshold is huge. f) August 10, 2014, 11:00. A similar case,
- showing again the large effect of changing the threshold.