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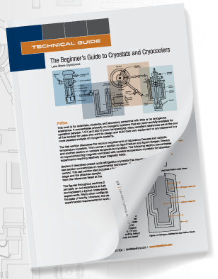

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How Important Is the Transition Zone Between Clouds and Aerosol?

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Abstract. Clouds are a macroscopic and easily observable natural phenomenon (a suspension of water droplets or ice crystals), which has been studied since the dawn of modern science. Clouds raise increasing interest in the scientific community, due to their fundamental role in the water cycle, energy balance, and climate. The atmosphere, however, contains other suspended particles that constitute the atmospheric aerosol, which also play an important role in the Earth's radiative balance. Moreover, they have a major impact on the formation, evolution, and characteristics of clouds. Study and quantification of clouds and aerosol often requires discriminating from each other. Although in general they differ in the proportion of water (liquid or solid), composition, size, shape, etc., there are situations that lie in the border between them (hydrated aerosols, haze, dissipating clouds, etc.) This communication reviews several studies that suggest that clouds and aerosol should be treated as a continuum or that an additional phase between them must be considered. Both observational works of this transition zone and studies that deal with the radiative effect of these transition situations are commented. We conclude that it is important to carry out additional research focused on suspensions of particles with characteristics between those of typical clouds and those of pure aerosol.

INTRODUCTION

Human observations of clouds (i.e., simple naked-eye records) were systematized two centuries ago in a classification into genera, species, and varieties, based on how they are perceived from the surface; this approach still applies (https://cloudatlas.wmo.int/en/home.html). In modern times, however, active and passive instruments have been developed to better characterize clouds and cloud fields (notably cameras, lidars, radars, other narrow-band radiometers), which may operate at wavelengths different from visible (infrared, microwaves, etc.) and from a variety of platforms (surface, aircraft, satellites). These instruments, however, do not necessarily detect the same as what a human observer would see, which complicates the use of the traditional classification (Boers *et al.*, 2010). An additional issue has to do with the spatial resolution of observations, which can carry errors in the estimate of cloud fraction (Di Girolamo and Davies, 1997). Regardless of the methods and instruments utilized for detecting the clouds (either visual observation or using sophisticated sensors), there are always conditions under which clouds are difficult to distinguish. Is it really cloudy? Is that patch a cloud? Where are the limits of this or that cloud?

Figure 1 shows some examples of all sky images. Despite the description given in the figure caption, all of them rise doubts about the nature of the suspension present in the air. Similar problems are found in other methods, either from the ground or from satellites, as in all cases the definition depends on algorithms and thresholds from which clouds are derived. These doubts arise because the separation between a cloud and the cloud-free air is not so clear. Or, in other words, because the definition of what is a cloud, from a macroscopic point of view, is not so precise (Spänkuch *et al.*, 2022), despite the separation of aerosol particles from cloud droplets is clearly defined from a microphysical perspective. Indeed, in cloud-free air there is always a certain amount of suspended particles, i.e., an

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aerosol. And these aerosol particles are often the condensation nuclei that initiate the formation of water droplets or ice crystals. A typical aerosol is made of particles smaller than 1 μm, with diverse composition depending on their origin, and mostly in solid phase. A cloud is constituted by water droplets and/or ice crystals (i.e., mostly water), typically greater than 5 μm. Visibility (or perceivability) of the suspension is a usual requirement to be considered a cloud, but this is also a diffuse term, as the threshold of visibility is uncertain and instrument dependent. It is quite plausible that there are suspensions of particles that present characteristics between these two typical situations. Thus, it has long been recognized that a continuum of conditions (Charlson *et al*., 2007) exists between cloud conditions and cloud-free –but containing aerosols– conditions. The intermediate condition has received different names, notably twilight zone (Koren *et al*., 2007) and transition zone (Calbó *et al*., 2017), but also haze, hydrated aerosol, etc.

This paper is a short review on the matter of the transition zone. The objective is to raise awareness and keep alive the interest in this particular kind of atmospheric suspensions, which are neither a typical aerosol nor a usual cloud. To do this, a few papers that deal with this matter are discussed in the next sections.

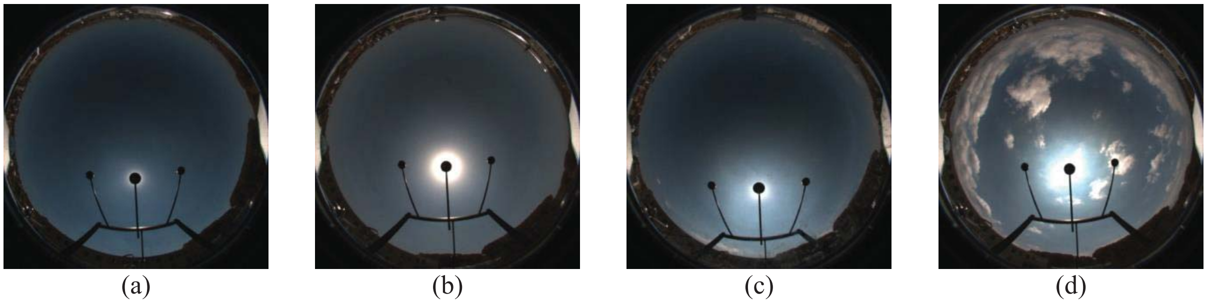

FIGURE 1. Four sample images from a sky-camera placed at the meteorological and radiometric station of the University of Girona, in Girona, Spain. (a) A cloud-free sky, with low aerosol load; (b) cloud-free sky, with high aerosol load; (c) apparently clear, but some thin clouds, or layers of aerosols, present; (d) scattered clouds.

THE TRANSITION ZONE

A first notable paper is that of Charlson *et al.* (2007), although these authors give credit to several earlier papers, some of them dating back to the 1980s. Charlson *et al.* (2007) advocate for a continuum of situations between the two main "modes" of the distribution: the clouds and the cloud-free air. They affirmed that the clear-cloudy distinction is ambiguous and aerosol dependent, and that partly cloudy atmosphere manifests a continuum of states between the end members 'clear' and 'cloud.' Therefore, the rationale for separating cloudy and clear skies in a wide variety of observational databases should be carefully reconsidered. Moreover, they found that cloud-clearing schemes employed by different observational methods are mutually inconsistent; this is relevant for any study that filters out cloudy situations or uses cloud masks. And all this has consequences: according to Charlson *et al*. (2007), the aerosol radiative forcing is inaccurately calculated as the average of clear and overcast conditions.

Also in 2007, another team coined the term "twilight zone" to refer to the conditions between the 'clear' and 'cloudy' (Koren et al., 2007). To support their arguments, their figure 1 is quite illustrative: by removing Rayleigh scattering, the image clearly shows that the influence of the cloud extends far beyond the "visible" (i.e., in the non-enhanced image) limits. They stated that despite it is customary to distinguish between 'cloudy' and 'cloud-free' areas and measure them separately, the shift between clouds to cloud-free atmosphere contains an additional component, a "Twilight Zone" or a gradual transition zone that depends on both the presence of nearby clouds and on the aerosol loading. The authors tried to quantify the extend of the area covered by the twilight zone, and as a result, they pointed out that for an average global cloud fraction of 0.51 the area of the 10 km border would cover 17% of the globe (34% of the cloud-free area) while a border of 30 km width would cover 30% of the globe (60% of the cloud-free area). Other papers followed, including that of Várnai and Marshak (2011) that estimated as 15 km the extension of the transition zone away from well-formed clouds.

The contribution of Schwarz *et al.* (2017) presents a quite different approach to the quantification of ubiquity of transition zone conditions. This research paper analyzed a series of MODIS images over the ocean, for the months of February and August, 2007–2011, and suggested that 'Lost' pixels, i.e., pixels that the algorithm is unable to classify as either cloud, clear, or aerosol, might be those containing transition conditions. Interestingly, they found that about 20% of all pixels are discarded by both MODIS aerosol and cloud retrievals and labeled as 'Lost' pixels. This must be an upper limit on the amount of transition conditions, as the class 'Lost A' is closer to cloud properties (and likely contain pixels with cloud fragments), while classes "Lost B" and "Lost C" are closer to aerosol properties. Specifically, the authors mention that 'Lost B' may involve hydrated aerosols in between aerosol loaded atmosphere and clouds, and 'Lost C' correspond predominantly to discarded –and in the first step of the algorithm classified as– aerosol pixels.

Also from 2017 is the study by Calbó *et al.* (2017), where the authors analyzed the sensitivity of three cloud-detection methods (pyranometers, Multifilter Rotating Shadowband Radiometer, and a Total Sky Imager) to the thresholds involved in these methods to separate cloud from cloud-free (aerosol) conditions. They found that there exist situations in which the distinction is far from obvious, and even when broken or scattered clouds are present in the sky, the borders between cloud/no-cloud are not always well defined. Results indicated that in more than 5% of the daytime hours the sky may be considered cloudless (but containing aerosols) or cloudy (with some kind of optically thin clouds) depending on the observing system and the thresholds applied. Similarly, the same paper (Calbó *et al.*, 2017) showed that at least 10% of the time the extension of scattered or broken clouds into clear areas is problematic to establish and depends on where the limit between cloud and aerosol is fixed.

Indeed, all the above papers, and many others (see references therein) cope with the definition of clouds. And precisely, "What is a cloud?" is the title of the paper by Spänkuch *et al.* (2022). The starting point is the different definitions between the American Meteorological Society (AMS) and the World Meteorological Organization (WMO): one definition involves the concept of visibility (AMS), while the other refers to perceivability (WMO); in addition, AMS includes all minute particles independent of their nature whereas WMO only considers such minute particles that consist of water and/or ice. In the end, the problem is the same: different instruments (including the human eye, but also a suite of ground based and satellite-borne instruments) involve different limits to distinguish clouds from clear (aerosol) conditions. Which is a situation that is "uncomfortable, confusing, and highly unsatisfactory" (Spanküch *et al.*, 2022). To address the issue, the authors suggest a new definition: "A meteorological cloud is an aggregate of minute particulate matter (solid, liquid, or mixed) in the atmosphere […] that becomes visible from ground at a line-of-sight optical depth of at least about 0.03 at day and 0.05 at night." This definition might seem more objective, involves a quantitative threshold, and includes as cloud many conditions that are currently considered aerosol. But it still has –at least– one problem: why would the same physical phenomenon, a given suspension of particles, be a cloud or not depending on the time of the day when it is observed?

RADIATIVE EFFECT OF THE TRANSITION ZONE

As the ubiquity of transition zone conditions in the atmosphere seems quite confirmed by the previous papers, it turns out that we should be able to take into account the corresponding radiative effects, which may affect both meteorological situations and climate, through the impact on the varying radiation fluxes and the average radiation balance respectively. In this sense, Jahani *et al.* (2019, 2020) explored how the description of the transition zone as either cloud or aerosol may involve important uncertainties in the estimation of the radiative effect of such a suspension of particles. These are theoretical studies, based upon the radiation parameterization included in the Weather Research and Forecasting (WRF) mesoscale model, which involve several simplifications (as for example plain-parallel approximation), but that anyway show important consequences.

Specifically, regarding the solar band (shortwave), Jahani *et al.* (2019) show that different treatments of the transition zone may lead to substantial uncertainties in simulation of direct, total, and diffuse irradiances and underline the importance of investigating the radiative effects of the transition zone, as the radiation field is of essential importance in meteorological and climate models. In addition, for the terrestrial band (infrared radiation), uncertainties related to the treatment as cloud or aerosol are also quantified: even at very small τ (optical depth) of 0.1, assuming a situation corresponding to the transition zone as cloud or aerosol may lead to a noticeable amount of uncertainty of the radiative effect at the top of the atmosphere ($\Delta\text{REtop}\uparrow$) of between 0.5–6.5 Wm^{-2} (values depending on layer height and season) and similarly at ground level ($\Delta\text{REbot}\downarrow$), 2.2–7.2 Wm^{-2} (Jahani *et al.*, 2020).

Besides the above sensitivity analyses, other recent studies involve actual observations of radiation fluxes in transition zone conditions (Eytan *et al.*, 2020; Jahani *et al.*, 2022). Both regard the radiative effect in the terrestrial band and combine satellite observations of top-of-the-atmosphere outgoing longwave radiation with other methods and techniques to estimate the fluxes in a cloud-free and aerosol-free atmosphere. The goal was to estimate in some way the radiative effect of layers of suspended particles that may represent the transition zone. In the first case (Eytan *et al.*, 2020), which is strictly for low-level layers over the ocean, the average longwave radiative effect of the twilight zone was found to be ~0.75 Wm^{-2}. The value seems low, but to put it in context, if it were a global average it would correspond to 75 additional ppm of CO_2 in the air. Therefore, the authors suggest that the twilight zone needs to be accounted for to accurately quantify cloud radiative effects and close the global energy budget. In the second case

(Jahani *et al.*, 2022), the radiative effect of the transition zone in the top of the atmosphere (REtrz) was on average equal to 8.0 Wm^{-2} (and cases of REtrz as large as 50 Wm^{-2} were found). The values are not so different from the previous study if the cases are restricted to transition zone suspensions in the lower levels of the atmosphere: the corresponding REtrz would be on average 0.8 Wm^{-2}. Given the uncertainties associated to the methodology, Jahani *et al.* (2022) pointed out that their results are probably upper bounds of the radiative effect of transition zone situations. Nevertheless, these values are not negligible in the context of the global energy budget.

CONCLUSION

After the brief review of a few papers about the matter, we may try to answer to some specific questions:

1. Is the no-cloud / no-aerosol suspension relevant? From the frequency of occurrence (or the spatial coverture) point of view, it appears that conditions between those of a well-formed cloud and a pure aerosol are relatively common and ubiquitous. Indeed, many studies show that time frequency and spatial extension of transition zone conditions are remarkable. In addition, from the point of view of the radiative impact of such suspensions, in turns out as well that associated uncertainties may be significant for radiative balance (climate forcing) estimation, according to several recent studies. Similarly, uncertainty in radiation fluxes (and the corresponding heating/cooling rates) may affect meteorological forecasts.
2. Is a new definition of cloud/aerosol needed? The authors of the current paper are not advocating such new definition. However, in studies devoted to 'pure clouds' or 'pure aerosol', the definition of these particle suspensions and the technique, thresholds, filters, and/or masks used to distinguish them should be clearly described and discussed, in order for the reader (or a follow-on researcher) to know exactly what a 'cloud' or an 'aerosol' refers to.
3. Should we simply talk about particle suspensions in the atmosphere? This could be a good option as would incorporate the idea of a continuum of conditions (which obviously includes the two extreme cases of cloud and aerosol). In particular, in radiation transfer models and parameterizations, considering this continuum of situations and characteristics (instead of a dichotomic separation) would probably improve results.

To summarize, the recommendation is to keep research on the transition (or twilight) conditions, in opposition to avoid including these conditions in cloud and aerosol studies. Other recent works (Marshak *et al.*, 2021) contribute to the debate and reach similar conclusions, for example by suggesting the term 'intercloud region" instead of the broadly used 'clear' sky.

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