The Impact of Neglecting Ice Phase on Cloud Optical Depth Retrievals from AERONET Cloud Mode Observations

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Abstract. Clouds present many challenges to climate modelling. To develop and verify the parameterisations needed to allow climate models to represent cloud structure and processes, there is a need for high-quality observations of cloud optical depth

- 15 from locations around the world. Retrievals of cloud optical depth are obtainable from radiances measured by Aerosol Robotic Network (AERONET) radiometers in "cloud mode" using a two-wavelength retrieval method. However, the method is unable to detect cloud phase, hence assumes that all of the cloud in a profile is liquid. This assumption has the potential to introduce errors into long-term statistics of retrieved optical depth for clouds that also contain ice. Using a set of idealised cloud profiles we find that, for optical depths above 20, the fractional error in retrieved optical depth is a linear function of the fraction of the
- 20 optical depth that is due to the presence of ice cloud ("ice fraction"). Clouds that are entirely ice have positive errors with magnitudes of order 55% to 70%. We derive a simple linear equation that can be used as a correction at AERONET sites where ice fraction can be independently estimated.

Using this linear equation, we estimate the magnitude of the error for a set of cloud profiles from five sites of the Atmospheric

25 Radiation Measurement programme. The dataset contains separate retrievals of ice and liquid retrievals, hence ice fraction can be estimated. The magnitude of the error at each location was related to the relative frequencies of occurrence in thick frontal cloud at the mid-latitude sites and of deep convection at the tropical sites; that is, of deep cloud containing both ice and liquid particles. The long-term mean optical depth error at the five locations spans the range 2–4, which we show to be small enough to allow calculation of top-of-atmosphere flux to within 10%, and surface flux to about 15%.

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Commented [JS1]: B3. The opening sentence of the Abstract has been reworded so that the motivation reads better. Much of the Abstract has also been reordered and edited following many comments on the mark-up document from Reviewer B.

1 Introduction

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Clouds are a crucial part of the climate system, yet present many great challenges to climate science (Randall *et al*, 2007; Boucher *et al*, 2013). Despite recent progress, climate models struggle to represent the optical properties of clouds (Bender *et al*, 2006; Lauer and Hamilton, 2013; Klein *et al*, 2013; Calisto *et al*, 2014). Cloud optical depth is particularly important to represent reliably as it governs the effect of clouds on the Earth's radiation budget. The complex processes and interactions

- that describe the evolution of clouds occur on scales much smaller than a model grid box and hence require parameterisation (Pincus *et al*, 2003; Shonk and Hogan, 2010). To develop and validate these parameterisations, there is a need for global observations of cloud optical depth at high temporal and spatial resolution.
- 10 A common approach to measure cloud optical depth is to retrieve it remotely from measurements of reflectance, radiance or irradiance in multiple spectral bands. Various methods have been developed to retrieve cloud optical depth from satellite measurements (for example, Arking and Childs, 1985; Nakajima and King, 1990; Platnick *et al*, 2001; Cooper *et al*, 2007) and ground-based instruments (Marshak *et al*, 2000, 2004; Barker and Marshak, 2001; Chiu *et al*, 2006). The need for global observations is best met by satellites, which are capable of providing routine cloud optical depth retrievals all around the world.
- 15 However, on account of their large pixel size, they struggle to provide the high temporal and spatial resolution required to investigate cloud processes. The underlying surface adds to the complexity of variability in the optical properties, and broken clouds and subpixel clouds increase the chance of errors and biases (Stephens and Kummerow, 2007). Using ground-based observations eliminates many of these issues. The proximity of clouds to the ground (much closer than a satellite orbit) means that a radiometer can achieve much smaller pixel sizes for the same viewing angle, allowing much higher temporal and spatial 20 resolution, and reducing the incidences of cloud edge.

A disadvantage of using ground-based observations is the lack of global coverage. We are limited to the small number of locations around the world where routine cloud optical depth observations are made: until recently, sites of the Atmospheric Radiation Measurement (ARM) Programme (Stokes and Schwartz, 1994) and the sites of the Aerosols, Clouds and Trace

- 25 Gases Research Infrastructure (ACTRIS) network that were formerly part of Cloudnet (Illingworth et al, 2007). But Chiu et al (2010) noted that radiometers distributed throughout the world as part of the AERONET project (Holben et al, 1998) could provide a readily available source of cloud optical depth observations and hence provide greater global coverage. When the sun is not obscured by cloud, these radiometers are in "aerosol mode" and make regular measurements of aerosol properties. When the sun is obscured, however, aerosol measurements are not possible and the radiometer becomes idle. Marshak et al
- 30 (2004) proposed that the "down-time" when the aerosol measurements are not possible could be used to observe cloud properties ("cloud mode") via measurements of zenith radiance.

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Cloud optical depth retrievals are made using the method proposed by Chiu et al (2010). It is based on that of Marshak et al (2004), and uses zenith radiances measured at two wavelengths (440 nm and 870 nm; one visible, one infra-red) to retrieve cloud optical depth and cloud fraction. Above a green, vegetated surface, the radiative properties of the clouds are similar at these wavelengths, but there is a strong contrast in surface albedo. Retrieval is performed using a set of radiance look-up tables

5 calculated at the two wavelengths. The approach has been shown to be applicable for both overcast and broken cloud fields (Chiu et al, 2006), and performed well when applied to an artificial field of clouds whose optical depth was known (Marshak et al, 2004). A limitation to the method is that it does not perform well near cloud edge: clear-sky contamination of the field of view, and high radiances arising from direct solar illumination of cloud edge, can both generate unrealistic optical depths (Chiu et al, 2006). In AERONET, contamination problems are reduced by clustering retrievals into 1.5-minute intervals and 10 excluding extreme optical depth values (Chiu et al, 2010).

Using this method, AERONET "cloud mode" optical depth retrievals have now been made routinely at a number of sites around the world for several years. A requirement for a "cloud mode" site is that the surrounding area is generally green vegetation: suitable AERONET sites were selected using satellite-derived contrasts in albedo at the two wavelengths (Chiu et

- 15 al, 2010). Cloud mode retrievals from AERONET are beginning to appear in published studies. An evaluation of data from one AERONET site in Cuba was made by Barja et al (2012). Antón et al (2012) used cloud mode data in a study into the effects of cloud optical depth on the transmission of ultra-violet radiation; Li et al (2018) used it to investigate seasonal and spatial distributions of cloud optical depth across China alongside satellite optical depth retrievals from MODIS (the Moderate Resolution Imaging Spectroradiometer; Platnick et al, 2003). An AERONET radiometer was also taken aboard a ship to probe the properties of boundary layer cloud in the north-eastern tropical Pacific (Painemal et al, 2017). 20

An extension to the retrieval method by Chiu et al (2012) included a third wavelength (1640 nm), which allows a retrieval of cloud droplet effective radius to be obtained alongside cloud optical depth and cloud fraction. Effective radius retrievals tend to be very sensitive to uncertainty in surface albedo and radiance measurements, so Chiu et al (2012) suggested performing

- the retrieval 40 times with perturbations to surface albedo and the measured radiance, thereby providing mean values of the 25 retrieved values and an estimate of the uncertainty in these retrievals. This method was used in the study of Painemal et al (2017), although the standard retrievals available on the AERONET website use the two-wavelength method of Chiu et al (2010).
- 30 However, neither of these retrieval methods are capable of retrieving cloud phase, so an assumption is made. Given the tendency for the liquid component of a cloudy profile to be substantially optically thicker than the ice component, it is assumed that the entirety of the retrieved cloud optical depth value is due to the presence of liquid cloud. This "warm cloud assumption" has the potential, therefore, to introduce an error into cloud optical depth retrievals in any case where a cloudy profile contains ice cloud, which could cause problems in studies that analyse long-term statistics of cloud optical depth.

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The objectives of this study are to (1) investigate the magnitude and sign of the retrieval error due to the warm cloud assumption, (2) ascertain whether it is large enough to drastically affect the statistics of long-term optical depth retrievals and, if necessary, (3) discover whether a simple correction method could be used to account for the error. The next section of this

- 5 paper describes the Chiu *et al* (2010) retrieval method in more detail and provides a first estimate of the sign and magnitude of the error. In Section 3, we examine the relationship of the error with both total cloud optical depth and how the optical depth is partitioned between ice and liquid components by performing retrievals on a set of idealised cloud profiles. From these results, we propose a simple linear correction equation that could be employed in AERONET locations where ice fraction can be independently determined. In Section 4, we investigate the potential magnitude of the error in real clouds measured at five
- 10 ARM sites using retrieval methods described by Mace *et al* (2006). We then discuss the results in Section 5, then summarise the study in Section 6.

2 Two-channel retrieval method

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Retrievals throughout this study are performed using the two-channel method described by Chiu *et al* (2010). The method begins with a set of look-up tables, which contain the radiance that would be observed at the surface under a cloudy profile for a range of different cloud optical depths, solar zenith angles and values of droplet effective radius. Using the Discrete Ordinate Method for Radiative Transfer radiation code (DISORT; Stamnes *et al*, 1988), a set of tables is calculated for each of the two wavelength channels, 440 nm and 870 nm. The surface albedo in the two channels is set to 0.05 and 0.35 respectively (typical albedo values over a green vegetated surface as reported by Chiu *et al*, 2010). The scattering properties applied to DISORT

for all look-up table calculations are those of liquid water droplets. The look-up tables span the optical depth range 1 to 100,

A pair of measured radiances at the two wavelengths is fed into the retrieval algorithm along with an assumed liquid effective radius (taken to be 8 µm throughout this study) and the known solar zenith angle at that time. From the look-up tables, the algorithm then searches for values of optical depth and cloud fraction that produce the specified radiances at the two wavelengths. To estimate the uncertainty on the retrieval, we follow part of the method of Chiu *et al* (2012) and perform 40

25 calculations, each one with a random perturbation applied to both the surface albedos and the observed radiances to represent uncertainty in their measurement. The output retrieved optical depth and cloud fraction therefore consist of a mean value and an indication of uncertainty.

To make an initial estimate of the sign and magnitude of the "warm cloud error", we use DISORT to calculate a few look-up 30 tables using scattering properties of ice particles and compare them with the corresponding look-up tables calculated using the properties of liquid droplets. We use a set of ice crystal phase functions for a randomly aligned distribution of rough-surfaced ice crystals, consisting of a mixture of shapes (a "general habit mixture"), retrieved from www.ssec.wisc.edu/ice_models/. **Commented [JS7]: B7.** Clarification that the optical depth lookup tables go up to 100.

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These phase functions were calculated alongside other single-scattering properties from field campaign data by Baum *et al* (2011, 2014). Their calculated properties are designed for use with radiative transfer calculations that allow retrieval of optical properties from satellites, with a different set of properties for each satellite platform to allow consistent retrieval. Given the availability of phase functions near the two wavelengths used in AERONET cloud optical depth retrievals, we select the phase

5 functions designed for MODIS. Figure 1 shows the ice phase functions at wavelength 465 nm for particles with effective diameters of 25 μm and 100 μm (the range of effective diameters that we consider in this study). The corresponding phase functions at 855 nm are similar.



10 Figure 1. Ice phase functions used in this study, originally designed for use in cloud retrievals from MODIS. Phase functions are shown for the forward scattering direction at wavelength 465 nm, for two ice particle effective diameters (see legend).



Commented [JS9]: B5. The lines have been coloured to make the dashed line clearer.



Commented [JS12]: B6. Text updated to indicate that the radiance is normalised.

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cloud that is either purely ice or purely liquid, for a prescribed solar zenith angle of 30° and a top-of-atmosphere flux of unity (hence the radiances presented are normalised). For a given optical depth, the observed radiance for liquid clouds is always
more than that for an ice cloud of the same optical depth over the entire range of effective sizes used in this study. This is because liquid droplets have a greater tendency to forward scatter than ice crystals, resulting in a greater radiance at the surface

Figure 2. Normalised radiances extracted from the liquid (red) and ice (blue and green) look-up tables for a range of different optical

depths, all calculated with unit top-of-atmosphere flux, for a solar zenith angle of 30° and at the visible 440 nm wavelength over a

Figure 2 compares the radiances that would be observed at the surface at the respective visible wavelengths under a column of

surface of albedo 0.05. The numbers in the legend are values of liquid effective radius and ice effective diameter.

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for the same amount of extinction. For any profile whose true optical depth is in the branch of the curve on Figure 2 where the radiance is monotonically decreasing with increasing optical depth (that is, to the right of the maximum), the error in retrieved cloud optical depth will be positive. Consider an example: the observed normalised radiance is 0.4, and we assume that the cloud is liquid with an effective radius of 8 μ m and has an optical depth greater than 10. From Figure 2, we would retrieve an

5 optical depth of about 25. However, if all of the cloud is in fact rough ice crystals with an effective diameter between 25 μm and 100 μm, the actual optical depth might only be between 16 and 17, implying a positive error of between 47% and 56%.

3 Errors in idealised cloud profiles

For a better understanding of the retrieval error, we use the two-channel retrieval method to obtain cloud optical depth for a set of idealised cloud profiles where the cloud optical depth is known. Each profile includes two cloudy layers: the top layer

- 10 is filled with ice cloud and the bottom layer is filled with liquid cloud, both with a cloud fraction of one. The properties of these cloud layers are varied in two ways. First, the total combined optical depth of the two layers is varied. Second, the partitioning of this total column optical depth between the ice and liquid layer is varied. We define a variable called "ice fraction" this is the fraction of the total column optical depth that is due to the presence of ice cloud. For each combination of optical depth and ice fraction, a full radiative transfer calculation is performed using DISORT to obtain the zenith radiance
- 15 that would be detected at the surface by a vertically pointing radiometer, serving as the synthetic "observed" radiance. The appropriate scattering properties are used for the liquid and ice layers. We fix liquid effective radius at 8 μm, and perform radiance calculations for ice effective diameters of 25 μm, 35 μm, 55 μm and 100 μm and for solar zenith angles of 10°, 30°, 50° and 70°, in both the 440 nm and 870 nm channels. Aerosol concentrations are set to zero.
- 20 Retrievals of cloud optical depth are then made from the "observed" radiances under the assumption that all clouds are liquid. Figure 3 shows that the true optical depth is generally well matched by the retrieved optical depth for profiles that contain cloud that is entirely liquid (ice fraction equal to zero), while increasing ice fraction reduces the surface radiance for a given cloud optical depth and results in an increasingly positive error. Furthermore, at most optical depths shown here, the fractional error in retrieved optical depth is largely independent of the true optical depth and increases linearly with increasing ice
- 25 fraction. For clouds that are entirely ice (ice fraction equal to one), the fractional error reaches about 70% if the ice effective diameter is assumed to be 25 μm and about 55% if it is assumed to be 100 μm. The fractional error is also largely independent of solar zenith angle, remaining at about 70% when the ice effective diameter is fixed at 25 μm and the solar zenith angle is varied (Figure 4).

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Figure 3. Retrieved optical depth (τ_{ret} ; top row), and retrieved optical depth as a fraction of prescribed ("true") optical depth ($\Delta \tau_{ret}/\tau_{true}$; bottom row) as a function of the true optical depth for the idealised cloud columns. Retrievals are made from DISORT radiance calculations with a liquid effective radius of 8 µm, a solar zenith angle of 30°, and two values of ice effective diameter (see panel headers). The lines and markers are coloured according to the ice fraction (see legend). The uncertainty in the retrieval, depicted here as the standard deviation in the retrievals across the 40 samples, is indicated by the vertical bars. Note that the markers and bars for each ice fraction value are slightly horizontally offset for clarity. Black dashed lines indicate the one-to-one line on the top panels and the zero line on the bottom panels.

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At low optical depths (values below about 20), however, the relationship between fractional error and ice fraction becomes more complicated, with a dependence on both the true optical depth and the solar zenith angle. The range of low optical depths affected by this more complicated relationship is also dependent on solar zenith angle. A simple explanation for these two different "error regimes" arises from Figure 2, and how the shape of the curves change with changing solar zenith angle and ice fraction. At higher optical depths (the "linear regime"), the observed radiance decreases monotonically with increasing optical depth. Changes to the ice fraction or solar zenith angle may change the nature of the curve, but do not change this

15 monotonic behaviour. At lower optical depths (the "non-linear" regime), the change of shape does not just affect the gradients, but also the location of the maximum point of the curve, adding complicated non-linearity into the relationship. **Commented [JS16]: B7.** Added description of the one-toone/zero lines.



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Figure 4. Retrieved optical depth as a fraction of true optical depth $(\Delta \tau_{ret}/\tau_{true})$ as a function of the true optical depth in the idealised cloud columns. Retrievals are made from DISORT radiance calculations with a liquid effective radius of 8 µm, an ice effective diameter of 25 µm and four values of solar zenith angle (see panel headers). Lines and markers as described in Figure 3.

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Based on DISORT computations and the assumed ice cloud particle diameters above, the relationship between fractional error in retrieved optical depth $\Delta \tau / \tau_{true}$ and ice fraction *f* in the "linear regime" is quantified using a simple linear empirical equation of the form

$$\frac{\Delta \tau}{\tau_{\rm true}} = (a \pm \Delta a)f + (b \pm \Delta b) ,$$

where *a* and *b* are the regression coefficients, and Δa and Δb are the uncertainties in these coefficients. This regression is 10 demonstrated in Figure 5, and yields coefficients of *a* = 0.534, *b* = 0.067 and Δb = 0.052. (The value of Δa was found to be negligible and less than 0.001.) To ensure retrievals in the "non-linear regime" are excluded, this regression only includes profiles with a true optical depth of greater than 20. To include a measure of uncertainty in the size of the ice particles, we include retrievals for all four values of ice effective diameter. Given that the solar zenith angle is known for a retrieved profile, it is conceivable to calculate regressions for each solar zenith angle separately and then add a solar zenith angle dependence

15 to Equation 1. However, variations in the regression coefficients for different solar zenith angles were found to be small, so we include all four solar zenith angles in one single regression for simplicity.

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(1)





A simple linear equation of this form may be used to correct the warm cloud error in AERONET optical depth retrievals if an estimate of ice fraction is available at the AERONET site; for example, via separate retrievals of liquid and ice water paths from microwave radiometer and radar measurements respectively. For all clouds in the "linear regime" with true optical depths

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- of above 20, it can provide reliable correction in the range of solar zenith angles considered here. In the optical depth range 10 to 20, applying the correction equation could lead to errors in some instances of high sun or low sun, although these are likely to be small (see Figure 4). Below optical depths of 10, the "non-linear regime" dominates and the reliability of the correction equation becomes questionable, as the fractional errors start to become large. However, when the values of optical depth are low, the absolute magnitude of the errors will be small and hence not a substantial contribution to errors in long-term cloud
- statistics. For the purposes of this study, we retain the simple linear regression presented above and accept its limitations. But we recognise that, for applications where retrievals of low optical depth are important, a more complex correction equation may be needed to account for errors in the "non-linear regime". This is discussed further in Section 5.

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Commented [JS26]: B8. Reworded to emphasise that the small errors at low optical depth do not affect our study in the goal of biases in long-term statistics, yet may be of concern for some applications. Changes continue at the end of Section 4.

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4 Statistics from real cloud profiles

For optically thick clouds with a high ice fraction, the error in retrieved optical depth can be large following Equation 1 (for a cloud that is entirely ice and has an optical depth of 50, for example, the error is about 30). The question then follows as to how frequently such optically thick ice clouds occur at the location of the AERONET sites with "cloud mode" retrieval. The

- 5 assumption that the liquid component of a cloudy profile tends to be optically thicker than the ice component, stated in Section 1, suggests that optically thick ice clouds may not be a frequent occurrence and hence only provide a small contribution to long-term statistics of cloud optical depth. In this section, we address this question by examining the distribution of optical depth and ice fraction in real clouds.
- 10 We therefore require a dataset that can provide independent values of ice and liquid components of optical depth at sites that contain AERONET radiometers that operate in cloud mode. We hence use cloud data retrieved at five ARM sites, using algorithms described by Mace *et al* (2006) and hereafter referred to as "ARM Mace" data. The methods of Mace *et al* (2006) derive a wealth of properties of an atmospheric profile using a combination of ground-based remote sensing techniques and radiosonde soundings, and provide a series of cloud profiles averaged over 5-minute intervals with a vertical resolution of
- 15 90 m. Liquid water path is obtained from brightness temperatures measured in two wavelength channels by a microwave radiometer. Ice water content is determined from millimetre cloud radar measurements using two approaches, depending on whether the profile contains pure ice cloud or a combination of ice and liquid cloud (either in separate layers or mixed-phase). The former case uses one of a set of algorithms to determine a distribution of ice water content from radar reflectivity and either Doppler velocity or longwave radiance at the surface; the latter uses a specially developed parameterisation that also
- 20 uses radar reflectivity and Doppler velocity. Separate values of ice and liquid optical depth components are then calculated from the liquid water path and the vertically integrated ice water content, hence allowing an estimate of ice fraction.

We fetched all available ARM Mace data from 2005 onwards at the Southern Great Plains site (SGP) in Oklahoma, the three Tropical Western Pacific sites in Manus, Nauru and Darwin, and the North Slope of Alaska site (NSA) in Barrow. There are

- 25 at least three years of data at each site, although the range of available years varies (see top part of Table 1). From this ARM Mace data, we extracted profiles that are potentially observable by an AERONET radiometer in cloud mode. We first removed all night-time profiles, and any profiles measured during periods of rainfall. Rainy profiles are indicated by the "precipitation flag" that is contained within the ARM Mace dataset; night-time profiles are identified by instances where the solar zenith angle is greater than 90°. We also removed any profiles that contained a retrieved value of ice water content greater than
- 30 2 g m⁻³, as such values cannot be considered reliable according to the ARM Mace documentation.

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Table 1. A summary of cloud statistics across the five ARM sites discussed in this study. Profiles included in these statistics consist
only of those from the ARM Mace dataset at times when an AERONET cloud mode retrieval would have been possible (see third
and fourth paragraphs of Section 4 for criteria).

	SGP	NSA	Manus	Nauru	Darwin
Years of available data	2005-	2008-	2005-	2005-	2005-
	2009	2010	2007	2007	2008
Number of profiles	74,973	80,477	27,564	21,229	53,166
Percentage of profiles that contain					
Liquid cloud	26.5%	17.0%	16.9%	37.1%	29.3%
Ice and liquid cloud, $f < 0.5$	29.4%	62.2%	34.8%	28.0%	29.0%
Ice and liquid cloud, $f > 0.5$	10.8%	14.6%	14.4%	4.8%	4.1%
Ice and liquid cloud, all f	40.2%	76.8%	49.1%	32.7%	33.1%
Ice cloud	33.3%	6.2%	34.0%	30.2%	37.6%
Percentage of profiles with errors					
Greater than 5	18.3%	23.7%	20.2%	7.3%	13.4%
Greater than 10	9.2%	13.3%	9.0%	2.9%	5.9%
Greater than 20	3.1%	4.6%	2.9%	0.5%	1.8%
Mean error over all profiles	3.5	4.4	3.5	1.8	2.8

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5 Finally, we accounted for the upper limit of total optical depth that can be retrieved by the AERONET cloud mode algorithm by removing profiles that have a *retrieved* optical depth of greater than 100. Considering the ARM Mace optical depths to be the "*truth*", we used Equation 1 to simulate the AERONET cloud mode retrieval process, generating a set of "retrieved" optical depths. Any "retrieved" optical depths greater than 100 were excluded. The retrieval error for each profile was determined as the difference between the "true" and "retrieved" optical depth values.

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It should be noted that this sample does not exclude profiles where the cloud optical depth is low, yet an AERONET aerosol mode retrieval is possible. Such a profile would be rejected from the aerosol data set as cloud contaminated, but would also not count towards the cloud mode statistics. However, accounting for these low optical depth profiles would not be trivial. Aerosol mode retrievals can be made for aerosol optical depths of up to 5 to 7 (Giles *et al*, 2019), but there is no specific

15 corresponding threshold in cloud optical depth. In the interests of ensuring the profiles that are *potentially* observable by AERONET in cloud mode are included, we chose to retain all low cloud optical depth profiles in the analysis, recognising that the frequency of occurrence of such profiles is likely to be overestimated.

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Figure 6. Histograms of ice fraction for real clouds observed at five ARM sites. All available profiles in the period 2005 to 2010 are included for which an AERONET cloud mode retrieval would have been possible (see third and fourth paragraphs of Section 4 for conditions). The "liquid" and "ice" bars indicate the fraction of total profiles that contain purely liquid or ice; the <u>remaining bars</u> indicate all other profiles, separated into bins of ice fraction. Data from the Mace *et al* (2006) dataset ("ARM Mace").

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We begin by analysing profiles from SGP – a mid-latitude site whose cloud regimes consist of both frontal and convective clouds with an overall average cloud fraction of about 50% (Lazarus *et al*, 2000). Ice fraction for SGP profiles is shown as a histogram in Figure 6a. Of the profiles, 26.3% contain cloud that is purely liquid and 33.3% contain cloud that is purely ice. Of the remaining 40.2% that contain both liquid and ice cloud, profiles that are mostly liquid (f < 0.5) outnumber those that are mostly ice (f > 0.5) by about three to one.

Most of the profiles containing cloud that is either mostly or entirely ice have a low optical depth, and would therefore provide small contributions to long-term error statistics in a cloud optical depth climatology from AERONET (Figure 7a). Conversely, optical depth values for liquid or mostly liquid profiles tend to be greater, but the contributions to overall mean error are also

Commented [JS46]: B2. "Mixed-phase" removed from the caption and the axis label of Figure 6 as it was being used to refer to all types of cloud with both liquid and ice and not mixed-phase in the usual sense.

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likely to be small on account of low values of ice fraction. The contours on all panels of Figure 7 indicate the error in an AERONET retrieval as a function of optical depth and ice fraction following Equation 1. At SGP, just under one in ten of the profiles has a cloud optical depth retrieval error of greater than 10 (9.2%), while only 3.1% of the profiles lie in the region where the error is 20 or greater. The mean error across all profiles is **3.5**.

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At NSA, cloud fraction tends to be higher than SGP at about 75% (Dong *et al*, 2010), consisting of mostly stratiform cloud. There is a prevalence of thick, low-level mixed-phase cloud (Mülmenstädt *et al*, 2012), particularly in the summer when most NSA profiles occurred (note that NSA is inside the Arctic Circle, so no AERONET profiles are possible in the perpetual darkness of winter). Table 1 shows that there is a much greater frequency of cloudy profiles containing both liquid and ice at

10 NSA with respect to SGP, with much fewer profiles occurring that are either pure liquid or pure ice (Figure 6b). The result is a higher frequency of optically thicker clouds that are mostly ice, but a lower frequency of optically thicker profiles that are entirely ice (Figure 7b). The mean error in cloud optical depth as NSA is **4.4** – slightly higher than at SGP.

At the three tropical sites, the clouds tend to be much deeper and convective in nature, with a much greater occurrence of

15 upper-level ice clouds (Stubenrauch *et al*, 2010). Despite their relative proximity, however, the meteorological conditions at the three sites are quite different. Manus is situated in the western Pacific "warm pool", and experiences much more convective activity throughout the year (Jakob and Tselioudis, 2003), while Nauru is on the edge of the warm pool and experiences much less, although with a strong influence from the phase of the El Niño Southern Oscillation (Long *et al*, 2013). In contrast, Darwin experiences a strong seasonal cycle in its convective activity associated with the passage of the Australian Monsoon, with deep convective clouds occurring seasonally (Protat *et al*, 2011).

The prevalence of deep convection at the three sites reflects the differences in frequency of profiles with high ice fraction (Figures 6c, 6d and 6e). The total frequency of profiles that contain both ice and liquid and have an ice fraction greater than 0.5 is 14.4% at Manus, 4.8% at Darwin and 4.1% at Nauru. The greater frequency of convection at Manus appears as a higher fraction of profiles with high ice fractions (Figure 7c), resulting in the greatest overall error across the tropical sites (**3.5**). The

much lower frequency of convection at Nauru results in fewer profiles appearing in this area of the histogram (Figure 7e), and hence a much smaller overall error (1.8). With an intermediate amount of convection and a greater fraction of optically thick ice cloud, the mean error at Darwin lies between the values at Manus and Nauru (2.8).

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 Commented [JS50]: B4. "Would be" replaced.)

Commented [JS51]: B.

Commented [JS52]: B2. Mixed phase updated – the histogram cannot show whether this is mixed phase in the traditional sense, so we have replaced it.

Commented [JS53]: B2. Mixed-phase clarified again.

Commented [JS54]: B2. Mixed phase clarified.



Figure 7. Two-dimensional histograms of ice fraction and cloud optical depth at the five ARM sites for the same set of profiles as in Figure 6. The "liquid" and "ice" rows show the optical depth distribution of the profiles that contain purely ice or liquid; the rest of the plot separates the mixed-phase clouds by ice fraction as in Figure 6. The colour scale indicates the fraction of the total number of profiles in each two-dimensional bin. The blue lines show the absolute error in retrieved optical depth that would result from AERONET retrievals as a function of ice fraction and cloud optical depth, calculated from Equation 1.

5 Discussion

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The analysis above from the five ARM sites implies that, if an estimate of ice fraction is not available at a given AERONET site, using uncorrected retrieved optical depths will lead to a mean error of order 2–4 in long-term statistics. Assuming typical

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mean cloud effective radius values of $6-12 \mu m$, cloud optical depth errors of 2-4 are equivalent to errors in liquid water path of $8-32 \text{ g m}^{-2}$ (using Equation 2 in Chiu *et al*, 2012), which is of similar magnitude to retrieval uncertainty in liquid water path from microwave radiometer observations (Marchand *et al*, 2003; Crewell and Löhnert, 2003).

- 5 To compare these uncertainties to a relevant climate variable, let us set out to retrieve cloud optical depths to sufficient accuracy that both top-of-atmosphere and surface fluxes are correct to within 10%. According to Figure SB1 of Turner *et al* (2007), for a liquid cloud with a liquid water path of 100 g m⁻² and an effective radius of 8 μ m, a typical top-of-atmosphere shortwave flux would be 500 W m⁻², and the sensitivity of the top-of-atmosphere flux to the liquid water path about 1 W m⁻² (g m⁻²)⁻¹. In this case, reproducing the top-of-atmosphere flux to within 50 W m⁻² implies a need for retrieval with an error of less than
- 10 50 g m⁻², equivalent to a cloud optical depth error of about 10. The mean AERONET cloud mode error of 2–4 is within this limit. By a similar argument, the presence of the same liquid cloud would result in a surface flux of about 300 W m⁻² with a sensitivity of surface flux of about 2 W m⁻² (g m⁻²)⁻¹. To get the 10% accuracy in surface flux, the retrieval then would need to be accurate to less than about 15 g m⁻² in liquid water path, or 3 in optical depth. Our errors may be slightly higher than this limit in some locations, and could only reach ~15% accuracy in surface flux.
- 15

such estimates would be required.

At present, not all AERONET sites have the instrumentation to allow an ice fraction estimate to be made. A potential method to detect particle phase using AERONET radiometers that are polarimetrically sensitive could help with estimates of ice fraction, although further work is needed (Knobelspiesse *et al*, 2015). Estimates of ice fraction could be generated from other sources – for example, radiosonde soundings and satellite measurements. However, while these approaches may provide an
estimate of ice fraction over a given area or timescale, they would not be capable of providing the high temporal resolution of ice fraction needed to complement the frequency of AERONET cloud mode retrievals. Further work into the applicability of

Needless to say, if an independent estimate of ice fraction is available, we advocate the use of Equation 1 as a correction factor.

- 25 Given that it is specific to the retrieval algorithm, it will be globally applicable to radiance measurements from any AERONET radiometer under the assumption that the ice crystals in a cloud are rough, consist of a mixture of shapes and have effective diameters in the range 25 μm to 100 μm. The equation we have proposed here is applicable for all profiles with optical depths over 20 and performs satisfactorily on profiles with optical depths from 10 to 20. While the equation presented here does not perform well for profiles with optical depths below 10, it may easily be extended to provide better correction at low optical
- 30 depths via extra non-linear regressions. Alternatively, retrieval methods are being developed that allow the retrieval of low optical depths from surface radiometers: Guerrero-Rascado *et al* (2013) proposed a method to obtain cloud optical depth estimates using cloud-contaminated AERONET aerosol mode observations, which could provide an alternative source of data for low cloud optical depths. The method of Hirsch *et al* (2012) could also be used, although this would require the installation of specialised radiometers at AERONET sites.

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Commented [JS58]: A1. Comment on the possibility of including radiosonde information on retrievals.

Commented [JS59]: B8. Another reference to the fact that the equation could be redefined to fit errors at low optical depths. Commented [JS60]: B. Another possible extension to Equation 1 involves the treatment of mixed-phase clouds. We generated the equation using idealised profiles with separate layers of ice and liquid cloud, therefore working under the assumption that, generally, ice and liquid cloud is separate. This fails to account for layers of mixed-phase cloud, however, which consist of a mixture of ice and liquid particles. Following Sun and Shine (1994), the zenith radiance below a mixed phase cloud will be slightly lower than

5 liquid particles. Following Sun and Shine (1994), the zenith radiance below a mixed phase cloud will be slightly lower than that below the same cloud but with its ice and liquid particles separated into two layers. Quantifying the effect of this mixing on the correction equation would be a pertinent future step.

6 Summary and conclusions

- 10 The representation of cloud properties in climate models still presents a huge challenge to climate scientists. To make progress in our understanding of cloud processes, we need global observations of cloud optical depth at high spatial and temporal resolution. Ground-based measurements are best suited to provide such resolution, although global coverage is limited. Using the radiometers of the Aerosol Robotic Network (AERONET) increases the number of sites around the world by making routine "cloud-mode" measurements made during the "down time" when aerosol measurements are not possible. Retrievals are made using radiance at two wavelengths (440 nm and 870 nm) and a set of look-up tables. However, as the method is not
- able to retrieve cloud phase, the assumption is made that all of the retrieved optical depth is due to the presence of warm, liquid cloud hence, for any cloudy profile that contains an ice cloud component, there will be an error in the retrieval.

We began by investigating the sign and magnitude of this "warm cloud error". A set of idealised cloud profiles were generated 20 with varying total optical depth and "ice fraction" (the fraction of optical depth in the profile that is due to the presence of ice cloud). We calculated the radiances that would be observed by a radiometer at the surface underneath the cloud profiles, and then used these radiances to retrieve the cloud optical depth. Comparison of the retrieved optical depths with the true, prescribed optical depths revealed that, for profiles that are mostly or entirely ice, the fractional error in retrieved optical depth was between 55% and 70% for ice particle diameters between 25 µm and 100 µm. At optical depths above 20, the fractional

- 25 error was found to be a simple linear function of ice fraction and showed negligible dependence on optical depth or solar zenith angle. Using a simple linear regression, we were able to generate an empirical equation (Equation 1 in this paper) linking the fractional error to the ice fraction. This equation has the potential to be used as a correction factor for AERONET optical depth retrievals. However, independent estimates of ice fraction are needed, which is currently not possible at most AERONET sites.
- 30 We then estimated the error in retrieved optical depth for a range of profiles of real clouds. We used multiple years of cloud data from five sites of the Atmospheric Radiation Measurement (ARM) program, which were then sampled to include only profiles that could potentially be observed by an AERONET radiometer in cloud mode. Using Equation 1, an estimate of the

Commented [JS61]: B2. Recognising that mixed-phase clouds have not been accounted for in this study.

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retrieval error was generated for each profile. Clouds that were mostly ice tended to have lower optical depths, while optically thicker clouds tended be mostly or entirely liquid – both of these conditions lead to small errors. At each of the five sites, only \sim 15% of the profiles had an error in retrieved cloud optical depth of larger than 10. The magnitude of the mean error at each location was dominated by the frequency of occurrence of optically thick clouds that were mostly or entirely ice – that is, either

- 5 thick frontal cloud or deep convection. At the two sites located outside the tropics, where thick frontal cloud is the largest error contribution, the overall mean error was related to the frequency of occurrence of such optically thick clouds composed of both ice and liquid particles. In the tropics, the error at each location was related to the frequency of occurrence of deep convection, with much greater variety in the error statistics. This suggests that variations in convective cloud occurrence may have a greater influence on the overall error than variations in frontal cloud occurrence.
- 10

The mean value of optical depth retrieval error at the five ARM sites is typically in the range 2 to 4. We showed that errors of this magnitude are small enough to allow the calculation of top-of-atmosphere fluxes to within 10% accuracy, and surface fluxes to within about 15%. Furthermore, when expressed in terms of liquid water path, these errors are of comparable value to uncertainties in retrievals from microwave radiometers. These results alone suggest that AERONET cloud mode retrievals

15 provide a valuable source of cloud optical depth data from a large network of surface observation sites. A higher degree of accuracy may be possible, though, via the use of a correction equation if an independent estimate of ice fraction is obtainable at the AERONET site.

Data Availability

20 The ARM Mace data used in this study can be accessed from https://www.arm.gov/data/data-sources/atmcldradmace-19. AERONET cloud mode data can be accessed from https://aeronet.gsfc.nasa.gov/cgi-bin/type_piece_of_map_cloud.

Author Contribution

The experiment design and analysis was performed by JKPS and JCC. JKPS prepared the manuscript with contributions and advice from all co-authors.

25 Competing Interests

The authors declare that they have no conflict of interest.

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Acknowledgements

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5 calibrating and maintaining instrumentation and processing these data.

Commented [JS66]: More detail added on grants.

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Commented [JS67]: Reference added.

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Authors' response to reviewers comments on AMT-2019-169

We thank both reviewers for their suggestions and comments. Our responses are presented below in blue text. A marked-up version of the manuscript has been uploaded with changes highlighted.

Reviewer "A"

Kudos to the authors for this very well executed study and write-up. Implementation of the suggested linear correction factor for measured optical depths larger than 20 will provide a much needed improvement to current data bases of cloud optical depth and fraction.

 The only question that I have for the authors, who are free to address as they wish, is if local sounding information could be used to constrain estimates of cloud ice fraction when other types of measurements from radar or microwave radiometers are not available. Estimates of cloud base/top temperatures that can be derived from the 0 or 12Z soundings might be useful in this regard.

To apply the correction equation to AERONET cloud mode retrievals, we ideally need instantaneous estimates of ice fraction that could be aligned with an instantaneous AERONET retrieval of optical depth. However, approaches using radiosonde soundings (or also satellites) could be used to provide a general estimate of ice fraction over an area and a period of time. However, if the cloud regime changes, or the cloud formations are varying rapidly, this could be a source of extra error. Extra work would be needed on this. We have added sentences about this in the Discussion section (Section 5; page 15, lines 18–22).

Reviewer "B"

Summary: This paper reports the results of an algorithm that builds naturally on earlier ones developed by Marshak, Chiu, and others for retrieving cloud optical depth from two-channel (visible and near infrared) downwelling radiance measurements. Here, the goal of the algorithm enhancements is to reduce retrieval error when ice particles, as well as liquid particles, contribute to the downwelling radiances. The paper is easy to understand and all of the analyses make sense. I recommend its publication in AMT. My comments are minor in nature.

Comments:

1. The algorithm makes use of one-dimensional radiative transfer theory and differential surface reflectivity at the two wavelengths to compensate for it. After reading the Introduction it was not clear how accurate this approach is when applied to different cloud types. In particular, how good is it when applied to fields of horizontally small but vertically developed convective clouds in the tropics as a function of overall cloud fraction? Does significant side illumination and/or cloud side leakage of the convective elements cause problems? If Aeronet is located near a coastal site, do differences in the surface reflectivity of the land and the ocean cause problems? These types of questions are relevant to the paper because the algorithm is applied to tropical convective clouds at sites with mixtures of surface types in their environs. Strengthen the Introduction by making clear, with supporting references, how accurate the retrieval is expected to be for different cloud types and mixtures of different surface types. If this is not known, then say so and caveat your optical depth retrieval accuracies towards the end of the paper.

The performance of the method was examined by Marshak et al (2004) and Chiu et al. (2006), who showed that the method works well for both overcast and broken cloud fields.

The method will not work well when clouds do not fully cover the field of view (FOV) of the radiometer (the so-called "clear-sky contamination" issue; see Chiu et al., 2006). Therefore, unphysical cloud optical depth can happen near cloud edges. Such contamination is more frequent in small cumulus clouds, although convective clouds have sufficiently large horizontal extents that they can completely cover the narrow 1.2° FOV of the sun-photometers. Note that when a time series of retrievals is available (e.g., cases in Chiu et al., 2006), one can detect unphysical retrievals near cloud edges and remove them. For AERONET cloud

mode retrievals which are not made from a complete time series, it is more difficult to detect these unphysical retrievals. For this reason, AERONET reports a "cluster" average (see Chiu et al., 2010), excluding retrievals below the 25th and above the 50th percentile – a similar approach to that of Remer et al (2005), for aerosol retrievals.

Differences in surface reflectivity is important to consider for cloud mode retrievals. Surface albedo information is considered over a 4 × 4 km domain surrounding the AERONET site, and a combination of land and ocean surfaces surrounding a site is not ideal. For this reason, the sites included in the AERONET cloud mode dataset have been selected to ensure that the spectral contrast from surrounding vegetated surface is sufficient for the retrieval method (see Chiu et al, 2010).

We have added more detail addressing these questions into the fourth paragraph of the Introduction section and divided the paragraph into two (see page 3, lines 3—15).

A study that uses three-dimensional radiative transfer theory to pound on these types of uncertainties would be valuable if such a study has yet to be performed. If it has, do reference and discuss it within the Introduction.

The reviewer is right; such studies would be valuable. While a lot of studies have focussed on reflected radiance at the top of the atmosphere, there is actually no published paper for zenith radiance at the surface. We looked into 3D radiative effects on cloud mode retrievals a long time ago, but have not found time yet to wrap this up. We incorporate the reviewer's point in the manuscript and, hopefully, this could also motivate others to conduct thorough analyses on 3D radiative transfer.

2. Lines 23-25 on Page 5 and Lines 1-2 on Page 6 indicate that the radiative transfer is always performed with the ice in a top layer and the liquid in a bottom layer. So these calculations are done with ice on top and liquid underneath and not for what are traditionally called mixed-phase clouds. Does it make any difference if the layers are mixed up together to form what is generally called a mixed-phase cloud?

Mixing up the ice and liquid will affect the radiance (see Sun and Shine, 1994) – the zenith radiance will be slightly higher if the ice and liquid particles are fully mixed rather than in two separate layers as in our idealised calculations. More work would be required to understand and quantify the effect of this "mixing" on the correction equation. We have included this question in the future work paragraphs at the end of Section 5 (page 16, lines 1—7).

On Page 10, Line 4, the paper refers to mixed-phase cloud in the traditional sense. But on Page 12, Line 12, it is not clear what mixed phase means here. Just be sure to be clear everywhere exactly how the liquid and ice are being dealt with. It may not make any difference for the calculations, but it sure does make a difference for the retrievals: retrieving the properties of liquid only and ice only clouds is not easy but it sure is easier than retrieving the properties of ice and liquid particles all mixed together.

We have been quite inconsistent with what we mean by "mixed-phase" clouds in this study, especially in Section 4. We have now tidied it up in various places so that "mixed-phase" is only used to mean mixtures of ice and liquid particles, while general clouds containing both liquid and ice have been described as such. It should now be clearer in both places mentioned in the comment above. We have also cleared it up in Table 2, where the words "mixed-phase cloud" have been replaced with "ice and liquid cloud".

Error estimates for the retrieved liquid and ice particle properties would be more convincing if they were provided in the context of the types of clouds above. These context-based error estimates would be of value if percolated into uncertainties for the retrieved optical paper depths. For example, in convective clouds with mixtures of in-cloud rain, in-cloud ice precipitation, liquid cloud, and ice cloud, not clear at all as to what the actual errors in the retrievals might be.

We thank the reviewer for this suggestion. We agree that it would be good to provide errors in the context of cloud types, but we wish to leave it for future work because it requires substantial work and proper ancillary datasets to address this issue properly. Note that AERONET sun-photometers only operate in the absence of precipitation to keep lenses clean and dry. Therefore, we actually do not have many observations for convective clouds described above. If in-cloud rain evaporates before reaching the ground,

we have chances to sample such clouds and provide retrieval. However, we have found rain (or drizzle in most cases) do not affect zenith radiance significantly due to its small number of concentrations, based on work in Fielding et al. (2014, 2015). It would be important to tackle the properties of precipitating ice particles, but current retrievals and model simulations for ice microphysical and optical properties are quite uncertain. We are currently working on ice retrieval using polarimetric radar measurements. Hopefully, we can find good collocated datasets to address this.

Minor Details:

A marked-up manuscript is being returned to the authors; perhaps some of the mark up may be of value to them.

Thank you – this was a helpful inclusion. We have made a number of modifications following your suggestions on the mark-up version (not all of which are mentioned directly in a comment here).

 First sentence of the abstract: "Cloud optical depth remains a difficult variable to represent in climate models" might be true for a bunch of different reasons not related to "a need for high-quality observations of cloud optical depth from locations around the world". So, the first sentence of the abstract is not compelling.

We have reworked the first few sentences of both the Abstract and the Introduction to better link the challenges associated with modelling clouds to the need for cloud optical depth observations. Also, we have reworked and edited the Abstract following the many comments on Reviewer B's marked-up manuscript.

4. The words "could", "could be", "can be",..., are used a lot in the paper. These are weak words in a scientific context and replacing them all with well thought out stronger words would improve the paper.

All instances of these words (as highlighted in the reviewer's marked-up document) have been reviewed and, where appropriate, stronger words have been used.

5. Page 4, Figure 1: The dashed line is really hard to see close to 0.

The lines on Figure 1 are now coloured to make the contrast easier. For consistency, the colouring of the lines has been changed in Figure 2 to match.

6. Page 5, Line 13: Wrong units for radiance.

The units for radiance both here (now page 6, line 3) and in the y-axis label of Figure 2 should actually be dimensionless, as they correspond to radiances calculated for a unit flux at the top of the atmosphere. This has been corrected, and the normalised nature of the radiances explained in the caption in Figure 2. We have also clarified that the radiances presented here are normalised in the text (page 5, lines 18—19).

7. Page 6, Figure 3: Why not squares for the top row of figures with the same x- and y-axis range? A line along the diagonal would help too.

The square axes for the top panels of Figure 3 is not practical, as extending the range of true optical depths to 100 would result in the inclusion of many retrievals that exceed 100 (the maximum optical depth in the AERONET look-up tables; this limit has now been mentioned in Section 2). Hence we have only included true optical depths up to 50 as, in this range, none of the retrievals (however high their ice fraction) exceed 100. For clarity, however, we have added the one-to-one line on the top row of panels in Figure 3 as requested. For consistency, we have also added zero lines on the bottom row of Figure 3 and all of Figure 4.

8. Page 8, Lines 23-24: "hence far less of an issue ..." is a subjective statement and would depend upon the application. As such, it is not a correct statement for all situations.

Our paper centres on assessing whether the errors affect the long-term cloud optical depth statistics, but we do recognise that there may be instances where accurate retrievals are required at low optical depths. Greater accuracy at low optical depths could be achieved by generating an improved, more complex version of Equation 1. We have highlighted this in Section 3 (page 9, lines 14—18) and then again in a little

more detail in Section 5 (page 15, lines 28—30). This Discussion section is a new addition that brings together the various discussion points from the results sections in one place.

9. Page 10, Lines 14-19: Past tense would probably be better for describing what you did to execute the study.

The paragraphs describing the data and how we sampled it is all now in the past.

10. Page 13, Figure 7: Make sure all of the minor tick marks show up in the figure.

The grid on Figure 7, which previously was drawn behind the coloured boxes of the 2D histogram, is now replotted on top (but under the blue contours).

Please also note the supplement to this comment: <u>https://www.atmos-meas-tech-discuss.net/amt-2019-169/amt-2019-169-RC2- supplement.pdf</u>

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