S3. CLoud and Atmospheric Radiation Retrieval Algorithm (CLARRA)

S3.1 Resolution and Model Errors

Table S1 lists the spectral ranges for the microwindows used in this work. Microwindows were selected where contributions from strong lines was minimal, following Turner (2005).

Figures S1 – S6 show clear and cloud sky spectra and model errors in microwindows for resolutions of 0.1, 1, 2, 4, and 8 cm^{-1}.

S5. Results and Discussion

S5.1 Retrieval overview

The rational for using the log of the effective radius and for setting the maximum effective radius to ~50 μm is demonstrated through Figure S7 for spherical liquid drops. The figure shows $Q_{abs,liq}$ as a function of effective radius r_{liq} (note that the symbol names were simplified and that the x-axis is on a log scale) at several wavenumbers. $Q_{abs,liq}$ reaches a maximum value at each wavenumber, as indicated with asterisks. At these wavenumbers and values of r_{liq}, the kernels are zero for and changes in r_{liq} have a negligible effect on τ_a; thus the retrieval lacks sensitivity to r_{liq} at the given wavenumber. The value of $Q_{abs,liq}$ for which are kernels are <= 0 is indicated with a dashed vertical line; this represents a likely limit on the value of r_{liq} that can be retrieved.

The change in radiance with a change in r_{liq} in turn depends on the change in Q_{abs} with a change in r_{liq}. However, d$Q_{abs,liq}$ / dr_{liq} is not linear, as show in Fig S7b. Instead, $Q_{abs,liq}$ / dr_{liq} decreases as r_{liq} increases. This nonlinearity causes the inverse retrieval to overestimate the step size when r_{liq} is small, resulting in potentially overstepping the solution, and underestimating the step size when r_{liq} is large, resulting in decreased sensitivity (and increased likelihood of getting "stuck" at a large value of r_{liq}. The kernels can be made more linear by modifying the kernel to d$Q_{abs,liq}$ / d(log r_{liq}), shown in Fig. S7c.

S5.4 Cloud vertical inhomogeneity and Ice habit

Table S2 gives retrieval errors for combinations of imposed errors c: noise of 0.2 mW/(m^2 sr cm^{-1}), radiation bias of 0.15 mW/(m^2 sr cm^{-1}), and water vapor bias of 3%, with retrieved cloud heights. Overall, errors are smallest for inhomogeneous clouds and largest for dense and liquid topped clouds.

Table S1. Microwindows used in the microphysical cloud property retrieval. The top row gives the resolutions of the spectra while following rows give the spectral ranges (ν_1 - ν_2) of the microwindows used for those resolutions.

0.1 – 4 cm^{-1}		8 cm^{-1}	
ν_1	ν_2	ν_1	ν_2
494.9	499.0	495	499
530.0	533.7	530	538
558.0	562.0	558	562
770.9	774.8	771	780
785.9	790.7	785	793
809.5	813.5	810	819
817.0	823.5	817	827
828.6	834.6	829	838
843.1	848.1	843	853
860.1	864.0	860	870
872.5	877.5	872	883
891.9	895.8	890	898
898.2	904.8	898	908
929.6	939.7	929	939
958.0	964.3	958	966
985.0	991.5	985	993
1076.6	1084.8	1076	1083
1092.2	1098.1	1092	1100
1113.6	1116.6	1113	1120
1124.4	1132.6	1124	1131
1142.2	1148.0	1142	1150
1155.2	1163.4	1155	1162

Table S2. Errors in retrieved cloud microphysical properties for macroscopically varying clouds at a resolution of 0.5 cm^{-1} (where COD refers to cloud optical depth in the geometric limit, r_{liq} and r_{ice} are the effective radii of liquid and ice, and SD indicates standard deviation). For the upper set of cases, the following errors were imposed: noise of 0.2 mW/(m^2 sr cm^{-1}), radiation bias of 0.15 mW/(m^2 sr cm^{-1}), and water vapor bias of 3%; retrieved cloud heights were used.

Cloud type	Error	COD		Ice fraction		r_{liq} (μm)		r_{ice} (μm)	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
Dense	y	0.04	0.09	-0.07	0.12	0.1	1.9	5	9
Diffuse	y	0.06	0.08	-0.02	0.06	0.1	1.1	3	7
Inhomogeneous	y	0.03	0.08	-0.02	0.04	0.1	1.3	3	6
Liquid topped	y	0.07	0.09	-0.01	0.07	-0.3	1.6	6	8

Table S3. Errors cloud microphysical properties retrieved assuming a spherical ice habit, for ice clouds of varying habit (first column). COD refers to cloud optical depth in the geometric limit, r_{ice} is the effective radius of ice, and SD indicates standard deviation. The upper set of cases includes no other imposed errors and uses true cloud heights. The lower set of cases includes noise of 0.2 mW/(m^2 sr cm^{-1}), radiation bias of 0.15 mW/(m^2 sr cm^{-1}), and water vapor bias of -3%, and uses retrieved cloud heights. The final column shows the number of cases omitted due to having rms differences of greater than 1 RU between the measured and retrieved radiance.

Habit	Error	COD		Ice fraction		r_{liq} (μm)		Omit
		Mean	SD	Mean	SD	Mean	SD	
Sphere	n	0.008	0.019	0.00	0.01	-0.5	0.6	0
Hollow bullet rosette	n	0.6	0.4	0.05	0.07	-5	0.8	2
Smooth solid column	n	0.3	0.2	0.02	0.03	0	2	0
Rough solid column	n	0.3	0.2	0.02	0.03	-1	2	0
Smooth plate	n	0.5	0.3	0.00	0.06	-6	1.2	4
Rough plate	n	0.5	0.3	0.06	0.06	-5	1.5	2
Sphere	y	-0.01	0.06	0.06	0.15	-2	2	0
Hollow bullet rosette	y	0.6	0.4	0.11	0.14	-4	1	3
Smooth solid column	y	0.3	0.15	0.09	0.08	-2	1	0
Rough solid column	y	0.3	0.10	0.09	0.10	-1	3	0
Smooth plate	y	0.5	0.3	0.00	0.09	-4	1	4
Rough plate	y	0.6	0.3	0.11	0.13	-5	2	3

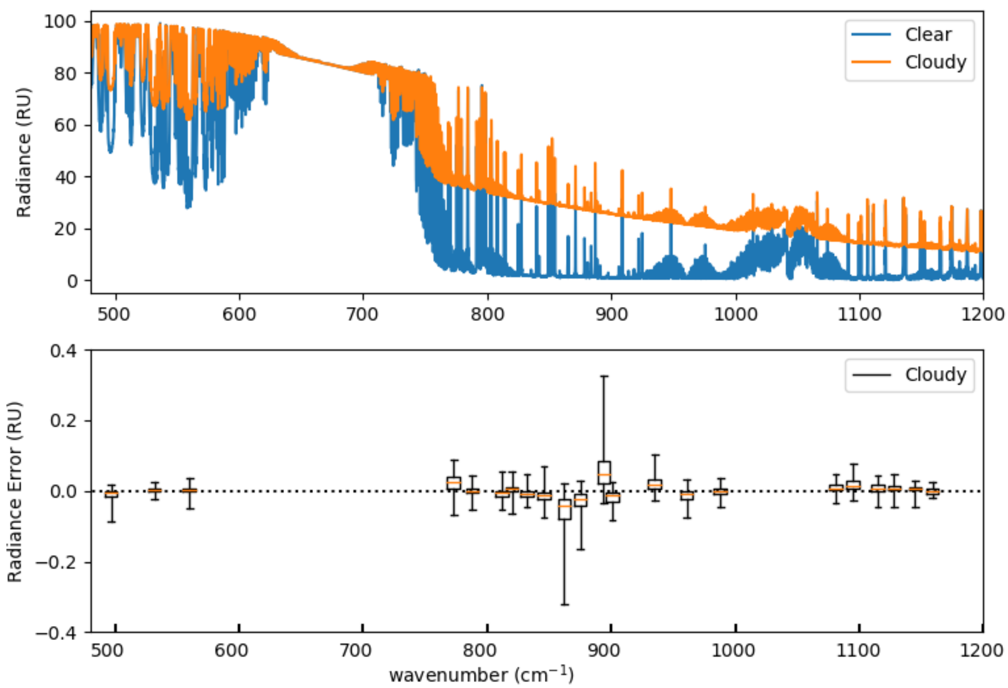

Figure S1. Clear and cloud-sky downwelling radiance (1 RU = mW / [m^2 sr cm^{-1}]) for typical polar atmosphere at a resolution of 0.1 cm^{-1} (top). Box and whiskers plots of model errors (model – true) for all radiances, averaged in microwindows (bottom; horizontal lines give the median, boxes give the 1st and 3rd quartiles, and whiskers give the range).

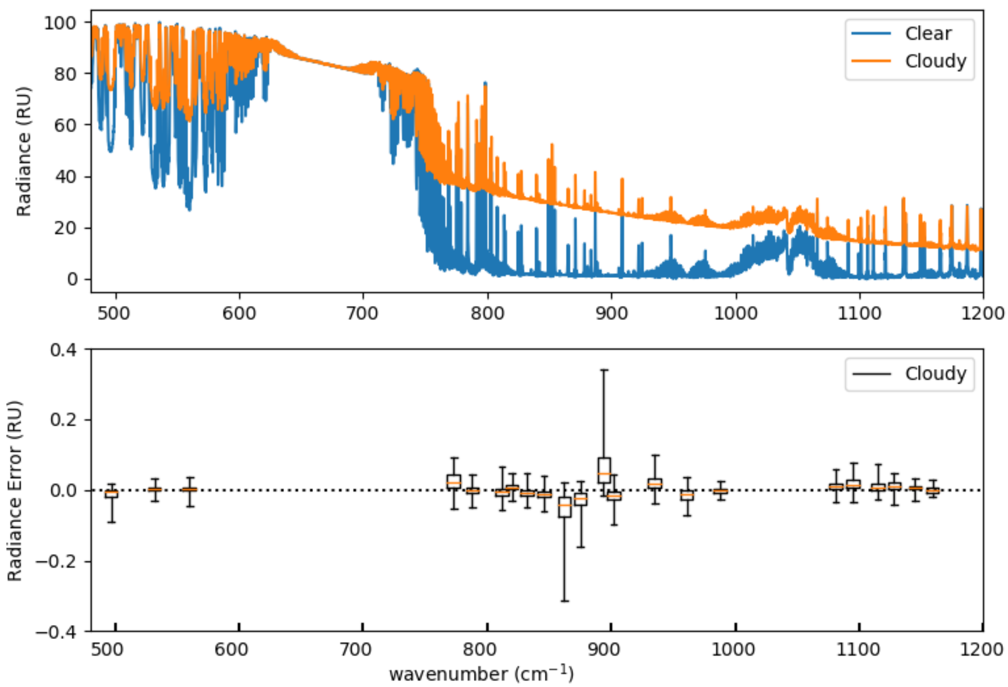

Figure S2. Clear and cloud-sky downwelling radiance (1 RU = mW / [m^2 sr cm^{-1}]) for typical polar atmosphere at a resolution of 0.2 cm^{-1} (top). Box and whiskers plots of model errors (model – true) for all radiances, averaged in microwindows (bottom; horizontal lines give the median, boxes give the 1st and 3rd quartiles, and whiskers give the range).

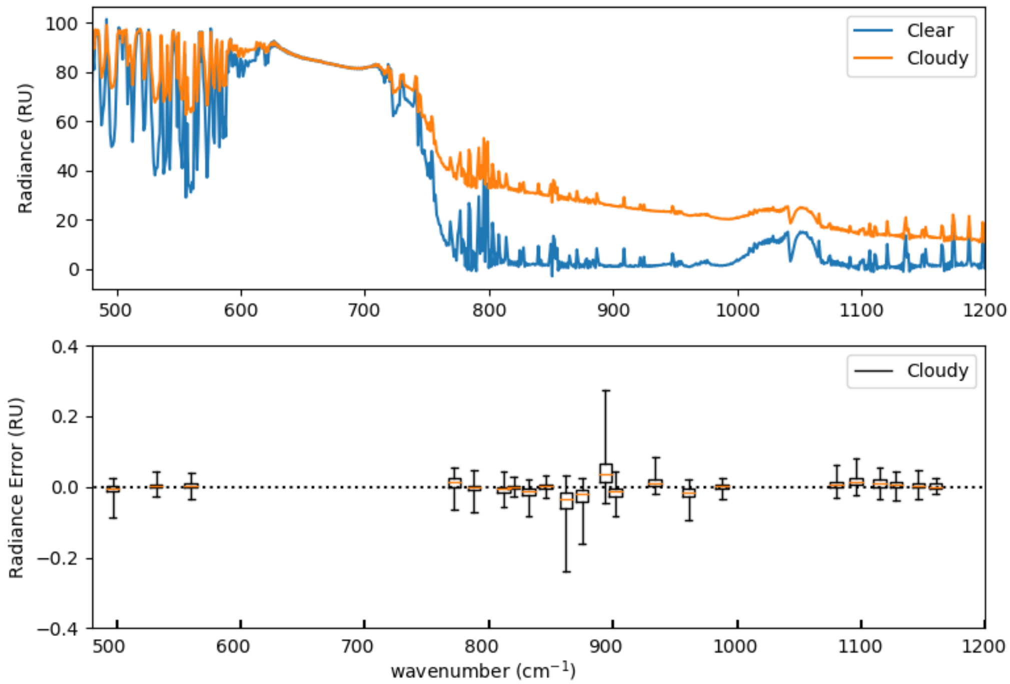

Figure S3. Clear and cloud-sky downwelling radiance (1 RU = mW / [m^2 sr cm^{-1}]) for typical polar atmosphere at a resolution of 1.0 cm^{-1} (top). Box and whiskers plots of model errors for all radiances, averaged in microwindows (bottom; horizontal lines give the median, boxes give the 1st and 3rd quartiles, and whiskers give the range).

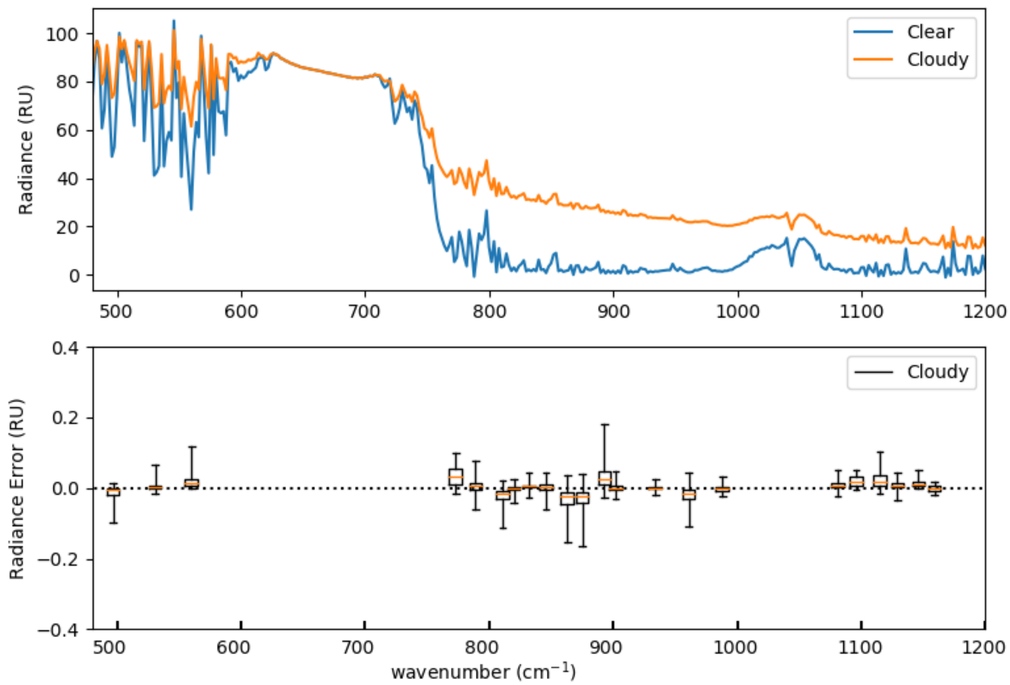

Figure S4. Clear and cloud-sky downwelling radiance (1 RU = mW / [m^2 sr cm^{-1}]) for typical polar atmosphere at a resolution of 2.0 cm^{-1} (top). Box and whiskers plots of model errors for all radiances, averaged in microwindows (bottom; horizontal lines give the median, boxes give the 1st and 3rd quartiles, and whiskers give the range).

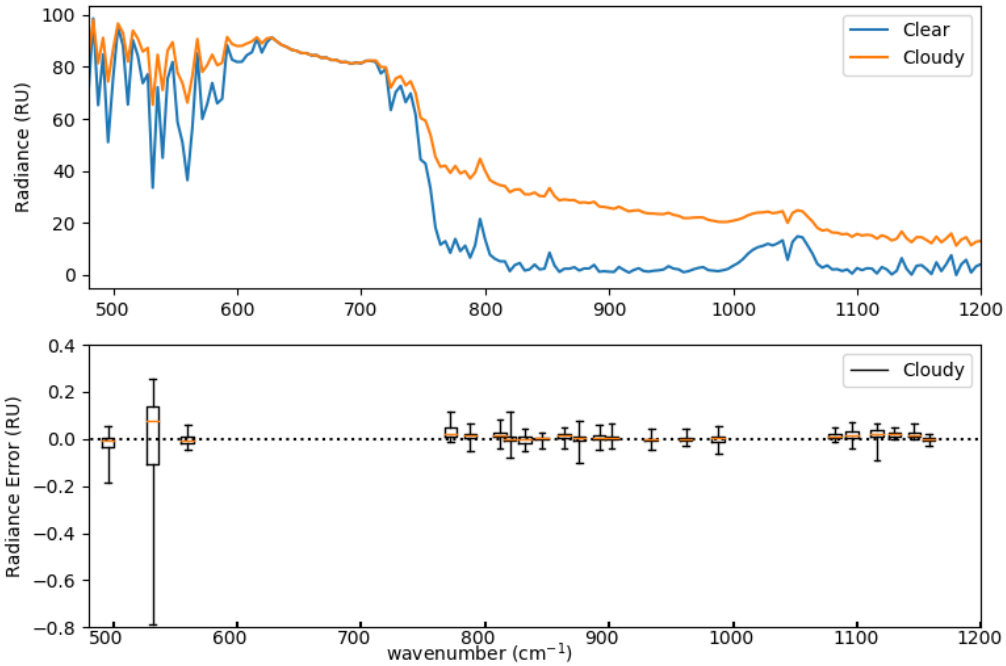

Figure S5. Clear and cloud-sky downwelling radiance (1 RU = mW / [m^2 sr cm^{-1}]) for typical polar atmosphere at a resolution of 4.0 cm^{-1} (top). Box and whiskers plots of model errors for all radiances, averaged in microwindows (bottom; horizontal lines give the median, boxes give the 1st and 3rd quartiles, and whiskers give the range).

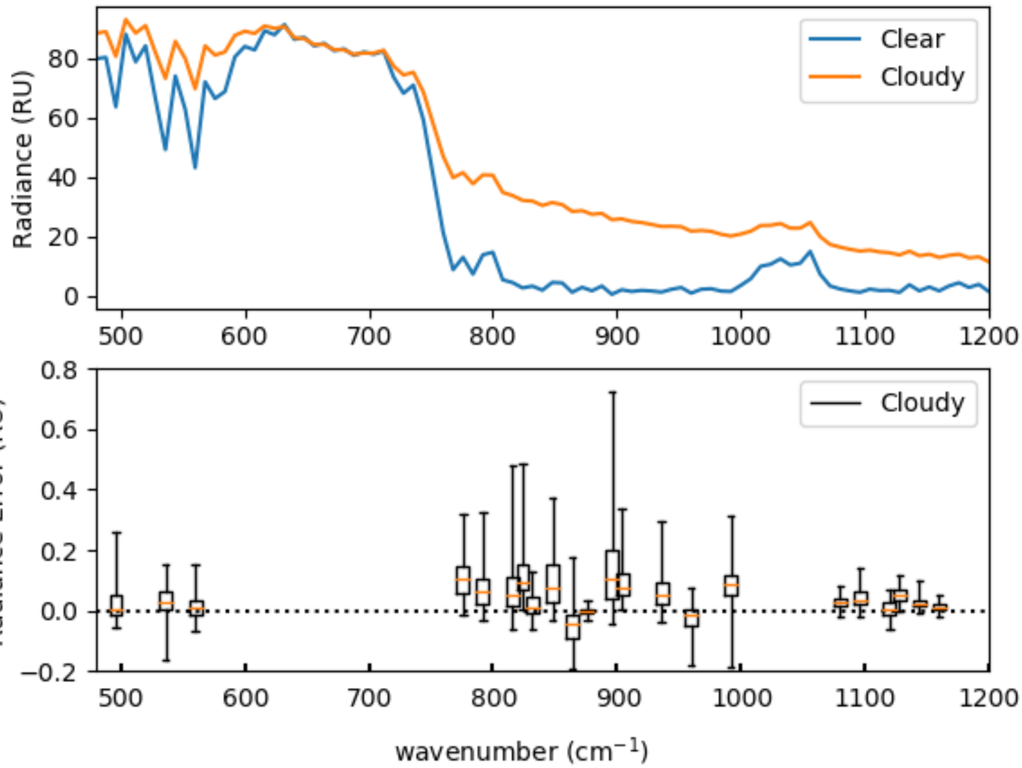

Figure S6. Clear and cloud-sky downwelling radiance (1 RU = mW / [m^2 sr cm^{-1}]) for typical polar atmosphere at a resolution of 8.0 cm^{-1} (top). Box and whiskers plots of model errors for all radiances, averaged in microwindows (bottom; horizontal lines give the median, boxes give the 1st and 3rd quartiles, and whiskers give the range).

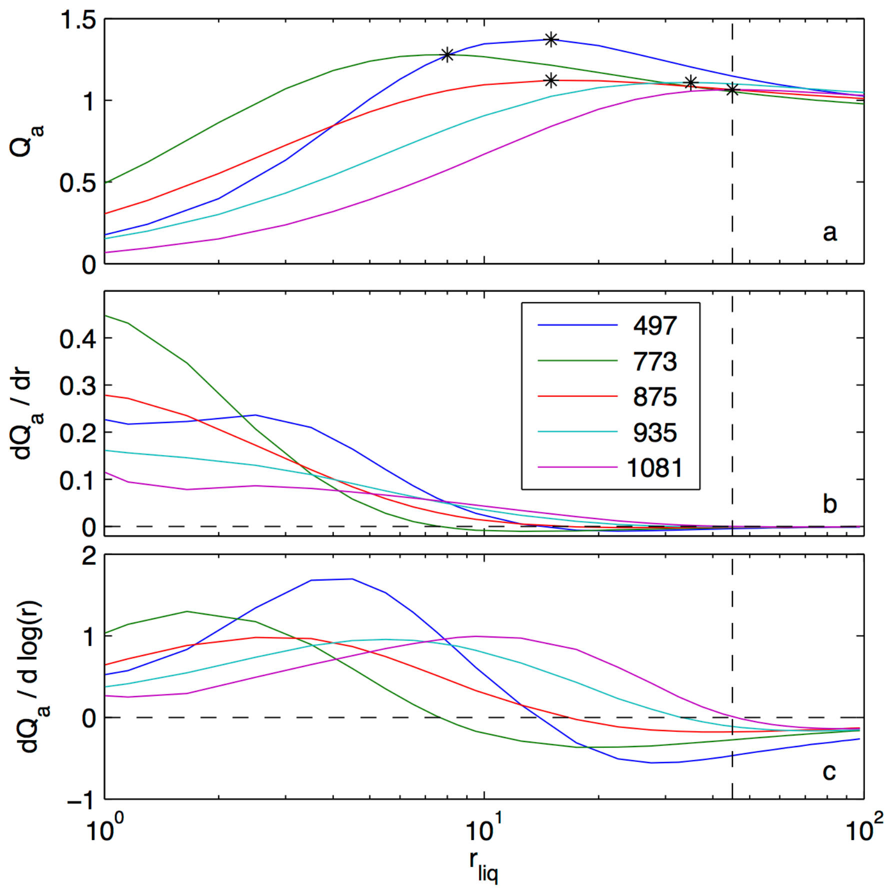

Figure S7. a) Absorption efficiency, Q_a, for spherical liquid water drops as a function of effective radius, r_{liq}. b) Change in Q_a with a change in effective radius of liquid. c) Change in Q_a with a change in log of the effective radius.