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Solar absorption by Mie resonances in cloud droplets

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Abstract

Recent studies suggest that resonant absorption of sunlight by cloud droplets may constitute a significant and unaccounted-for solar energy sink in the atmosphere. We spectrally resolve, for the first time, all solar absorption, including sharp resonances, in typical liquid water clouds. Resolving all sharp resonances requires a resolution in size parameter $\chi = 2\pi r/\lambda$ (r—droplet radius, λ—incident wavelength) of about 10^{-7}. The canonical integration resolution $\Delta\chi \approx 10^{-1}$ produces absorption biases up to 70% over 10 nm spectral bands. Hence, neglecting Mie resonances may cause substantial biases in radiance-based retrievals from sensor channels where atmospheric absorption is particle dominated.

The canonical resolution produces broadband solar mean and RMS absorption coefficient biases of about 0.02% and 4%, respectively. Self-cancellation of the pseudo-randomly distributed biases explains why the mean bias is much smaller than the RMS bias. Exceeding 1% RMS accuracy in solar absorption requires $\Delta\chi < 10^{-5}$. Increased cloud heating due to resolving all resonant absorption is less than 0.1%, equivalent to about $0.01\,\mathrm{W\,m^{-2}}$ global annual mean heating. Overlap of droplet and water vapor absorption within clouds helps diminish the net enhanced absorption by sharp resonances. Hence, the heretofore unrepresented absorption is negligible for global climate, though very important for narrow spectral regions. These results apply to homogeneous liquid water clouds and aerosols.

Keywords: Mie resonances; Size parameter; Particle absorption; Water clouds; Aerosols

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1. Introduction

Atmospheric solar absorption is not directly observable on global scales. Hence the partitioning of known column solar heating between the atmosphere and the surface is poorly constrained and is model based. Global models estimate that clouds, aerosols, and trace gases absorb at least 20% of the solar radiation annually received by Earth [1,2]. Atmospheric models base their predictions of solar absorption within liquid water clouds on parameterizations (e.g., [3–5]) of Mie theory. However, these parameterizations are based on spectral or size resolution [6] insufficient to fully resolve the resonant absorption structure [7,8] that accounts for about 20% of solar absorption by cloud droplets [4,9]. Nussenzveig [9] concludes that results from optical calculations at canonical resolutions "cannot be trusted". Hence, the adequacy, accuracy, and reproducibility of most atmospheric model (including all climate model) predictions of solar absorption within clouds is dubious. Our objectives are two-fold: First, to determine the intrinsic bias in current model estimates of cloud absorption due to neglect of resonant absorption in liquid water droplets. Second, to document the tradeoff between accuracy and computational expense of increasing the canonical resolution in typical atmospheric applications such as cloud and aerosol radiative transfer.

The Mie theory for scattering and absorption by homogeneous spheres depends only on particle composition and the size parameter $\chi = 2\pi r/\lambda$ where r is the particle radius and λ the incident wavelength (e.g., [10]). Dave [6] established the canonical resolution $\Delta\chi \approx 10^{-1}$ to obtain "reliable results" for scattering by aerosol distributions. Resonances at much finer scales were long ago predicted [7,8] and measured (e.g., [11]). Chýlek et al. [8] found no resonance structure finer than $\Delta\chi = 10^{-7}$ in water droplets. Our study investigates the sensitivity of the bulk optical and radiative properties of a particle distribution to changing size parameter resolution from the minimal resolution (10^{-1}) to the finest resolution (10^{-7}) required to resolve resonances. In particular, we examine the absorption coefficient and bulk absorptance of typical water clouds interacting with solar radiation.

The solar absorption efficiency Q_a of liquid cloud droplets is a small fraction of the total extinction efficiency Q_e. Let $\langle Q_{a,r}\rangle$ be the mean absorption efficiency due to resonances across the approximate resonance period $\delta\chi = (\arctan\mu)/\mu$, where $\mu = \sqrt{n_r^2 - 1}$ and n_r is the real index of refraction [9,10]. Nussenzveig [9] provides two estimates of the fractional contribution of resonances to total particle absorption efficiency, $\langle Q_{a,r}\rangle/\langle Q_a\rangle$. He estimates $\langle Q_{a,r}\rangle/\langle Q_a\rangle \approx 29\%$ and 16%, based on complex angular momentum theory and the geometric optics approximation, respectively. Multiple scattering in optically thick clouds allows each photon numerous opportunities to undergo absorption by droplets. Hence, multiple scattering in clouds may compound small biases in Q_a due to unresolved resonances into larger biases in total droplet absorption.

Previous studies parameterized effective cloud droplet optical properties for use in climate models [3–5]. These parameterizations typically input a cloud droplet effective radius r_e and possibly the width σ_g of the droplet size distribution. The parameterizations output effective (i.e., size- and wavelength-integrated) microphysical optical properties necessary for broadband radiative calculations: the extinction and absorption coefficients and scattering phase function information. Due to the computational burden, these studies did not resolve the full resonance structure of liquid water droplets. Hence, their predicted specific absorption coefficient $\psi(\lambda)$ ($m^2\,kg^{-1}$) could underestimate cloud droplet absorption.

Section 2 describes our Mie theory and radiative transfer methods and models. Section 3 shows the resonance-induced absorption of a single water droplet, a typical droplet distribution, and an entire water cloud. Sections 4 and 5 summarize the processes that reduce resonant absorption biases, and the scale-dependent requirements for accurate representation of absorption in homogeneous spheres.

2. Methods

Our "brute force" method resolves the full resonance structure of liquid water droplet distributions and solar radiation by decreasing the element of integration $\Delta\lambda$ (rather than Δr). We define the effective size parameter χ_e for a size distribution over a broad spectral region in terms of the effective radius r_e and nominal wavelength λ_e as $\chi_e \equiv 2\pi r_e/\lambda_e$. For $\lambda_e = 1\,\mu m$, this implies $\Delta\chi \approx 2\pi r_e \Delta\lambda$. Our implementation of the Mie theory scattering solution for homogeneous spheres follows Wiscombe [12], and is parallelized over wavelength to accelerate solutions on shared-memory supercomputers.

The typical size distribution of marine stratus, the most pervasive liquid water cloud [13], may be represented as a lognormal size distribution with effective radius $r_e = 10\,\mu m$ [14] and geometric standard deviation $\sigma_g = 1.6$. The size distribution is discretized into 30 logarithmically spaced size bins covering $0.1 < r < 30\,\mu m$. Indices of refraction for liquid H_2O in the solar spectral region are from Segelstein [15] and Wieliczka et al. [16]. Optical properties were computed from 0.2–5.0 μm, and then integrated over the size distribution to a 10 nm spectral resolution. Size parameters considered span four orders of magnitude, $0.1 < \chi < 1000$. Simulations for $r_e = 5$ and 7 μm lead to similar conclusions as the $r_e = 10\,\mu m$ results shown below.

We use a high-resolution solar radiative transfer model to estimate the effect of these resonances on liquid cloud absorption at local and global scales. The Shortwave Narrow Band radiative transfer model SWNB2 [17] computes solar radiative fluxes at $10\,cm^{-1}$ spectral resolution from 0.2–5.0 μm. SWNB2 computes irradiance with a four-stream discrete ordinates radiative transfer algorithm [18]. SWNB2 accounts for H_2O, O_3, O_2, CO_2, NO_2, and $O_2 \cdot X$ absorption [19], and includes thermal emission. Constituent profiles and state variables are obtained from the standard Mid-Latitude Summer (MLS) atmosphere. We assume a Lambertian surface reflectance of 0.1, and neglect all aerosol.

3. Results

The location and width of Mie resonances may be efficiently predicted [20], unlike their amplitudes. Hence our "brute force" method appears to be the first to quantify the amplitudes of all resonances for solar radiation interacting with liquid water. The resonance of maximum amplitude (RMA) occurs at $\chi_{RMA} = 53.0259$ ($\lambda_{RMA} = 1.1849281\,\mu m$ for 10 μm droplets). Fig. 1 shows the resolved spectrum of the mass absorption coefficient in the RMA. The value of ψ_{RMA} depends on the imaginary index of refraction. The range of liquid water imaginary refractive index (n_i spans more than eight orders of magnitude ($1 \times 10^{-1} < n_i < 7 \times 10^{-10}$) in the wavelength range of solar radiation ($0.2 < \lambda < 5\,\mu m$) [15,16]. Fig. 1 shows the RMA for $n_i = 1 \times 10^{-6}$,

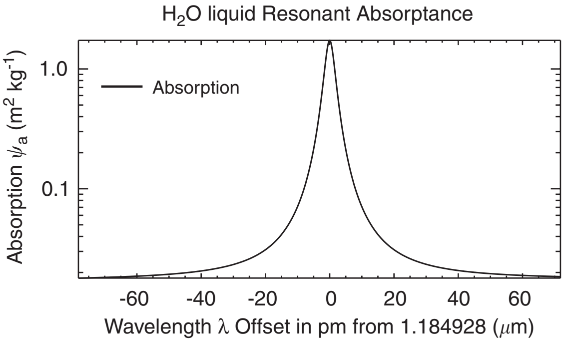

Fig. 1. The resonance of maximum amplitude for a sunlight and a 10 μm water droplet. Shown is the mass absorption coefficient ψ ($m^2\,kg^{-1}$).

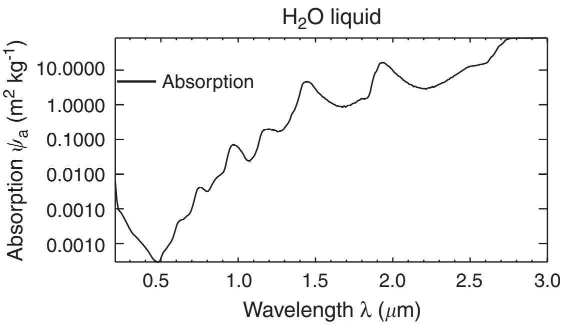

Fig. 2. Mass absorption coefficient $\psi(\lambda)$ ($m^2\,kg^{-1}$) of the cloud droplet size distribution $r_e = 10\,\mu m$, $\sigma_g = 1.6$ in the solar spectrum. Scale is logarithmic.

characteristic of liquid water for radiation near $\lambda = 1\,\mu m$. For this n_i, the maximum absorptance $\psi_{RMA} = 1.73\,m^2\,kg^{-1}$ is 70 times the local off-resonance absorptance.

Fig. 2 shows the mass absorption coefficient ψ ($m^2\,kg^{-1}$) of the cloud droplet size distribution $r_e = 10\,\mu m$, $\sigma_g = 1.6$ for the near solar spectrum. Integration over the size distribution smears out the narrow resonant absorption lines (e.g., Fig. 1). Absorption is less than $0.2\,m^2\,kg^{-1}$ for $\lambda < 1.3\,\mu m$, where incident solar flux is most intense. The top-of-atmosphere solar flux-weighted broadband mean specific absorption is $\bar{\psi}_{RMA} = 2.03\,m^2\,kg^{-1}$. Wavelengths $\lambda > 1.5\,\mu m$ dominate $\bar{\psi}_{RMA}$ since ψ increases with wavelength much faster than solar flux decreases.

To resolve all resonances and reach convergence, the quadrature point density per micron of spectrum, N, was increased from 10^2 to $10^8\,\mu m^{-1}$. This corresponds to reducing $\Delta\chi_e$ from $2\pi \times 10^{-1}$ to $2\pi \times 10^{-7}$, about six orders of magnitude finer than Dave [6]. Limited simulations with $\Delta\chi_e = 2\pi \times 10^{-8}$ yielded no new resonance features. Prior studies [8,21] also found that $\Delta\chi_e \approx 10^{-7}$ resolves all significant resonances.

Fig. 3 shows the percent change in mass absorption coefficient (Fig. 2) due to including all resonant absorption, i.e., increasing N from 10^2 to $10^8\,\mu m^{-1}$. These changes were averaged to 10 nm resolution for plotting. The maximum positive and negative changes, +13% and −74%,

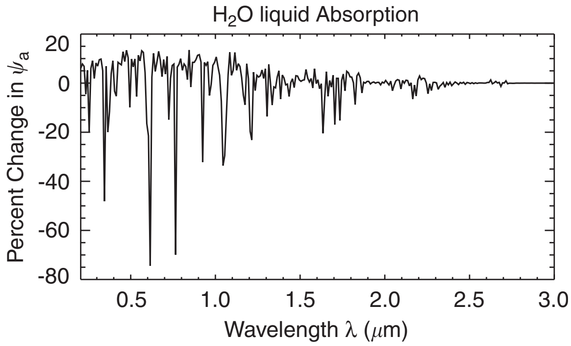

Fig. 3. Percent change in cloud droplet mass absorption coefficient $\psi(\lambda)$ (Fig. 2) due to including all resonant absorption. Changes were averaged to 10 nm resolution for plotting.

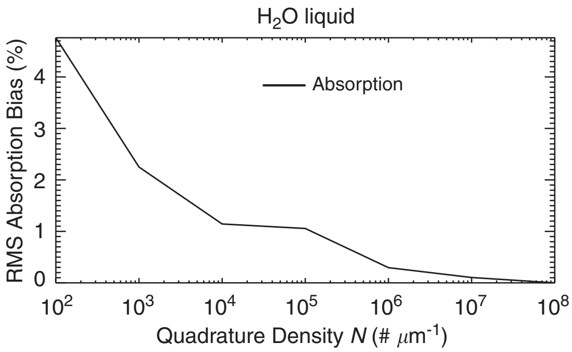

Fig. 4. RMS relative bias (%) in mass absorption coefficient of liquid water as function of spectral quadrature point density N (# μm^{-1}).

occur in the spectral bands $0.84<\lambda<0.85$ and $0.61<\lambda<0.62$, respectively. These results confirm the conclusion of Nussenzveig [9] that exceeding 10% accuracy in $\psi(\lambda)$ requires $\Delta\chi<0.1$. Moreover, we show that integrating over realistic size distributions does not significantly ameliorate the bias Nussenzveig [9] discusses for mono-disperse particle populations.

Resonances are positive definite and resolving them explains the positive changes in ψ. Quadrature points which coincide with resonances, but do not resolve them, weight resonances too heavily (causing positive biases). Optical properties computed at resolution finer than the resonances (e.g., Fig. 1) correct this undersampling, and therefore appear as negative excursions in Fig. 3. Not surprisingly, the envelope of the absolute percentage change grossly anti-correlates with the spectral mass absorption coefficient (Fig. 2).

We computed an RMS error metric to summarize the broadband average of the absolute magnitude of these spectrally distributed biases. This RMS error metric equally weights the relative error (relative to $N = 10^8\,\mu m^{-1}$) in all (280) 10 nm bands in $0.2<\lambda<3.0$ (Fig. 3, e.g., shows the relative error distribution for $N = 10^2$). Fig. 4 shows the RMS error metric (in %) for $10^2 \leqslant N \leqslant 10^8\,\mu m^{-1}$, corresponding to $2\pi \times 10^{-1} < \Delta\chi_e < 2\pi \times 10^{-7}$. The RMS error converges slowly and monotonically to zero as $\Delta\chi_e \to 0$. For the canonical $\Delta\chi_e = 0.1$, the RMS bias in ψ is

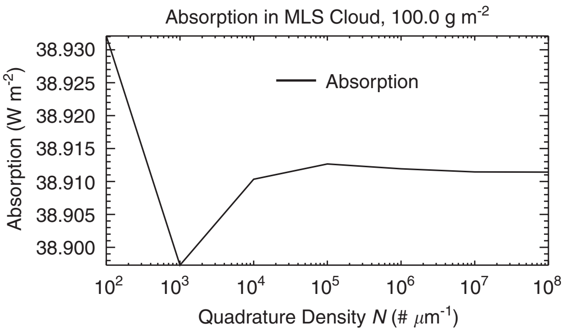

Fig. 5. Solar broadband radiative absorption ($W\,m^{-2}$) in a mid-latitude summer cloud with LWP $= 100\,g\,m^{-2}$.

about 4%. As $\Delta\chi_e \to 0$, the TOA flux-weighted broadband mean specific absorption $\bar{\psi}$ changes only about 0.02%, much less than the 4% change in the RMS bias (Fig. 4). Cancellation of positive and negative errors in different spectral regions (Fig. 3) explains the small size of the mean bias relative to the RMS bias.

Obtaining better than 1% RMS accuracy in solar absorption coefficients $\psi(\lambda)$ seems to be a reasonable target for radiative transfer applications which demand high accuracy. This 1%-RMS threshold requires $N > 10^5\,\mu m^{-1}$, equivalent to $\Delta\chi_e < 10^{-5}$ resolution.

Next we investigate whether the spectral biases in $\psi(\lambda)$ (Fig. 3) causes a significant broadband solar absorption bias in realistic clouds. Fig. 5 shows the solar broadband radiative absorption in a mid-latitude summer (MLS) cloud with liquid water path LWP $= 100\,g\,m^{-2}$ centered at 800 mb. The fractional change in cloud solar absorption due to resolving resonances is only $0.03\,W\,m^{-2}$, about 0.1% of solar cloud heating. The expected global mean, diurnal average change is approximately one half of this simulation at 60° zenith angle. Hence un-resolved resonances cause negligible broadband biases in cloud absorption.

4. Discussion

Light absorption in spheres may be orders of magnitude more efficient in resonances relative the surrounding spectral regions. Previous studies [8,21] found that size parameter resolution $\Delta\chi \sim 10^{-7}$ resolves the resonance structure in water droplets. Nussenzveig [9] showed that the much coarser resolutions $\Delta\chi \sim 10^{-1}$ typically used lead to absorption errors up to 30%, and so questioned the reliability of predictions which neglect narrow resonances. These studies were all conceptually based on the spectral distribution of resonances of a single particle. The net absorption due to all unresolved resonances from broadband radiation interacting with a particle distribution had not been quantified.

Defining an effective size parameter χ_e for a size distribution bathed in radiation near $\lambda = 1\,\mu m$, we found that resolving all sharp resonances requires effective size parameter resolution $\Delta\chi_e \sim 10^{-7}$. The canonical resolution $\Delta\chi \approx 10^{-1}$ produces absorption biases up to 70% over 10 nm spectral regions. In other words, narrow resonances from a size distribution do not overlap and blend into a smooth and easily integrable continuum. Satellites and aircraft carry moderate and high spectral resolution sensors with bandpasses $\Delta\lambda = 10\,\text{nm}$ and narrower. These sensors measure solar backscattered and thermal radiances which are often corrected for absorption by atmospheric particle absorption prior to retrieving the desired property (e.g., column O_3). Therefore neglecting Mie resonances could cause substantial biases in radiance-based retrievals in channels (e.g., 860 nm) where atmospheric absorption is particle-dominated. Better than 1% RMS accuracy in size distribution effective absorption coefficients on $\Delta\lambda = 10\,\text{nm}$ scales requires $\Delta\chi_e < 10^{-5}$. We recommend that remote sensing applications based on narrowband radiances, as well as benchmark forward radiative transfer codes, adopt this practice.

Effective resolution $\Delta\chi_e \sim 10^{-1}$ yields mean and $\Delta\lambda = 10\,\text{nm}$ RMS biases in absorption coefficient $\psi(\lambda)$ of about 0.02% and 4%, respectively. Fully accounting for this excess absorption in typical MLS clouds increases diurnal average solar absorption by less than 0.1%. Three reasons explain the small increase in broadband absorption. First, the typical practice of computing optical properties with (many orders of magnitude) too few quadrature points to resolve resonances leads to approximately correct broadband absorptances due to mutual cancellation of the relatively large positive and negative biases. Second, SWNB2 [17,19] estimates that cloud droplet absorption explains only 50–60% of solar absorption in liquid clouds. One-third to one-half of solar absorption in typical stratus clouds is due to interstitial water vapor. Overlap between water vapor absorption and random quadrature errors helps reduce errors due to neglecting droplet resonant absorption.

The third reason there is little increased broadband absorption is that potentially significant internally mixed absorbers such as soot [22] were not considered. Soot can significantly amplify resonant absorption in droplets [21]. Hence our results for homogeneous clouds are a lower bound on resonance effects in real clouds. Whether pollution significantly increases broadband solar absorption in clouds through resonance effects remains unanswered.

5. Conclusions

Excess absorption due to Mie resonances in pure liquid cloud droplets does not contribute significantly to broadband solar absorption. Fully resolving resonant absorption does increase net atmospheric absorption relative to standard model techniques. The global annual mean increase in atmospheric absorption due to resolving all resonant absorption is about $0.01\,\text{W}\,\text{m}^{-2}$. Hence, the heretofore unrepresented, and thus anomalous, absorption, is negligible. However, narrow resonances contribute very significantly to particle distribution absorption over moderate-to-fine spectral bands ($\Delta\lambda \lesssim 10\,\text{nm}$) used in remote sensing and benchmark radiative transfer applications. We expect all externally mixed homogeneous aerosols to behave qualitatively similarly to the water clouds studied here.

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