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Kristin S. Dooley
University of Central Arkansas, kdooley@uca.edu

Jessica DeYoung
University of Central Arkansas

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K.S. Dooley* and J. DeYoung

Department of Chemistry, University of Central Arkansas, Conway, AR 72035

*Correspondence: kdooley@uca.edu

Running Title: Refractive Indices of Particles Containing Inclusions

Abstract

Application of effective medium approximation (EMA) methods to two-component systems are presented. Systems studied are composed of water, sulfate, soot, and dust as these are commonly encountered atmospheric aerosol components. Atmospheric models often employ EMAs to include internally mixed aerosols without the computational burden of exact theory. In the current work, several types of mixing rules (Maxwell-Garnet, Bruggeman, and coherent potential approximation) have been applied to various two-component internally mixed particles at 550 nm using volume fractions of the minor component below 0.1. As expected, results show that the formulations tested produce very similar effective refractive indices indicating that electric field interactions between inclusions is negligible at the tested volume fractions. This indicates that the differences in component refractive index has only a minor effect on the validity of the EMA at the tested volume fractions. In all cases considered, the linear average of the refractive indices of the two components provides an upper limit for the EMAs.

Introduction

The optical properties of heterogeneous particles are of considerable interest in determining their role in climate change. Correctly modelling their effect on the global radiation energy budget hinges on the accurate computation of the particle's associated scattering and absorbing optical properties (Ackerman and Toon 1981; Lesins *et al.* 2002). A few scenarios where this type of internal mixing model is appropriate are when a hygroscopic component becomes solvated in a high humidity environment; cases where dust or soot particles contain numerous scattering inclusions due to adsorbed water or a mineral inclusion; or instances where particles are formed through various coagulation processes.

Mie scattering theory is used to computationally describe the scattering of radiation by particles which have a diameter similar to or larger than the wavelength of incident light. It can be used to retrieve information such as the single scattering albedo of the particle, when the particle's shape (in simple cases, particles are assumed to be spherical), size and refractive index of the particle are known (Bohren and Huffman 1983).

In more complex cases when particles are composed of multiple components in a random orientation, the particle does not have a homogeneous index of refraction and exact solutions of Maxwell's equations with appropriate boundary conditions are needed in order to solve the scattering problem. Solutions to this problem do exist in the literature (Chýlek and Videen 1998; Videen *et al.* 1995; Ioannidou and Chrissoulidis 2002). Unfortunately, the treatment is a multiple scattering problem resulting in solutions that are impractical for use in climate models due to their computational complexity. In atmospheric applications, the computational cost of exact solutions is unwarranted given that other data such as particle shape, and size distribution also contribute to the errors in the computed radiation budget. Consequently, prediction of the optical properties to within a few percent is often deemed sufficient for geophysical applications (Chýlek *et al.* 2000).

Therefore, it is appealing in the case of climate models to approximate the effective refractive index (n_{eff}) of a particle containing inclusions using various computational mixing formulations known as effective medium approximations (EMAs) (Chýlek *et al.* 2000). EMAs are based on *ad hoc* assumptions that lead to a simplified, more easily solvable model of the real particle. They allow for the computation of n_{eff} of the mixture based on the refractive indices of the individual components and their relative volume fractions as illustrated in Fig.1. Effective medium approximation formulations have been shown to be effective up to a volume fraction of inclusions (f) of 0.1 or smaller, although they are often used outside of this range

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(Bohren and Huffman 1983). These formulations are also valid only when the size of the inclusions is much smaller than the wavelength of the incident light.

Figure 1: The diagram illustrates how effective medium approximations (EMAs) simplify the treatment of a particle containing randomly placed inclusions. By inputting the permittivities of the components with their respective volume fractions, the EMA computes an effective permittivity, ϵ_{eff}, of the homogenized particle for use in Mie scattering calculations.

There are numerous EMA formulations in the scientific literature, with various and sometimes conflicting merits and limitations (Sihvola 1999). In the present work, the Maxwell-Garnet, Bruggeman, and coherent potential approximations are compared. These formulations all assume a dilute suspension of small inclusions (<0.01µm diameter) inside a single-component matrix. Although they have all been expanded to include ellipsoids and other types of mixtures, only the original formulations for spherical inclusions are considered here. The main difference in these formulations is the amount of coupling between the electric fields of the inclusions (Sihvola 1999).

For small volume fractions of inclusions, the three formulations should predict the same results for n_{eff}. The formulations differ at larger volume fractions of inclusions, but the point and degree of the difference is dependent on the permittivities of the components (Sihvola 1999). In the current work, the approximate formulations have been applied to mixtures composed of common atmospheric aerosol components (water, sulfate, soot, and dust).

Materials and Methods

All computational work was done using Mathcad 15 (PTC).

Computational parameters of mixtures

The effective refractive indices of two-component mixtures are computed at an incident wavelength of 550 nm as this is relevant to climate models. Because they are common materials in atmospheric aerosols and provide a range of relevant visible refractive indices,

water, sulfate, dust, and soot are chosen as the major components of the mixtures while one of the other four materials is chosen as the low volume fraction inclusions.

At 550 nm, the refractive indices of the components used are: $n_{water} = 1.333 + i1.96 \times 10^{-9}$, $n_{sulfate} = 1.470 + i1.00 \times 10^{-7}$, $n_{dust} = 1.530 + i5.50 \times 10^{-3}$, and $n_{soot} = 1.750 + i0.440$ (Hale and Querry 1973; Weast *et al.* 1986; Toon *et al.* 1976; d'Almeida *et al.* 1991). It should be pointed out that the refractive index of soot and dust are somewhat variable. The refractive index of soot will be dependent on the combustion conditions in which the particles are generated, and the specific mineral components will affect the refractive index of dust. As the point of the study is to explore the EMAs themselves, the use of these representative refractive indices is acceptable.

One significant advantage of using these parameters is that a small portion of the effective refractive indices computed in this study can be verified via data published previously (Erlick 2006). That work included only combinations of a non-absorbing particle with absorbing inclusions at low volume fractions corresponding to data reported in our Tables 5 and 6. The current values are in very good agreement with those previously reported. The current work expands that study to include all combinations of materials and volume fractions to provide a clearer understanding of the differences in the EMA formulations when used in instances where the indices of refraction of the major component and inclusion are quite similar and when they are relatively different.

As the incident wavelength is at least four times the size of the inclusions, it is more appropriate to apply the mixing rules to the dielectric constants of the materials rather than to the refractive indices themselves. The relationship between refractive index, n, and dielectric constant, ϵ, is $n = \epsilon^{1/2}$. The real and imaginary terms of the dielectric constant (corresponding to capacitance and loss, respectively) for a material are related to the real and imaginary terms of the index of refraction through $\epsilon_{real} = n_{real}^2$ and $\epsilon_{imag} = 2n_{real}n_{imag}$ (Bohren and Huffman 1983).

The effective dielectric constants (ϵ_{eff}) of the mixtures are computed using the mixing rules described in the following subsection. The effective dielectric constant may then be transformed to an effective refractive index (n_{eff}) by taking the complex square root of ϵ_{eff} (Bohren and Huffman 1983). These are given as:

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$$n_{real} = \left[\frac{\left(\epsilon_{real}^2 + \epsilon_{imag}^2\right)^{1/2} + \epsilon_{real}}{2} \right]^{1/2} \quad (1)$$

$$n_{imag} = \left[\frac{\left(\epsilon_{real}^2 + \epsilon_{imag}^2\right)^{1/2} - \epsilon_{real}}{2} \right]^{1/2} \quad (2)$$

Mean field formulations used to compute ϵ_{eff}

The Maxwell-Garnet, Bruggeman, and coherent potential approximations (CPA) are used for computation of ϵ_{eff}. In these equations, the permittivities of the inclusion and the major component are designated as ϵ_{incl} and ϵ_{major}, respectively. The value f refers to the volume fraction of the inclusion in the mixture. These results are also compared with a simple volume weighted linear average of the component refractive indices. A linear average has often been found to overestimate the imaginary term of the refractive index of a particle with inclusions; thus, it will provide an upper bound for this value (Sihvola 1996).

The most commonly used mixing rule is the Maxwell-Garnet formulation. This model depends only on the volume fraction (f) of the inclusion and assumes that second-order effects due to neighboring inclusions may be neglected (Bohren and Huffman 1983). The formulation for ϵ_{eff} is given in Equation 3.

$$\epsilon_{eff} = \epsilon_{major} + \frac{3f\epsilon_{major}(\epsilon_{incl} - \epsilon_{major})}{\epsilon_{incl} + 2\epsilon_{major} - f(\epsilon_{incl} - \epsilon_{major})} \quad (3)$$

The Bruggeman approximation makes many of the same assumptions as Maxwell-Garnett, but increases the influence of the inclusions to be symmetric to that of the background medium (Bohren and Huffman 1983). This formulation is shown in Equation 4:

$$(1-f)\frac{\epsilon_{major} - \epsilon_{eff}}{\epsilon_{major} + 2\epsilon_{eff}} + f\frac{\epsilon_{incl} - \epsilon_{eff}}{\epsilon_{incl} + 2\epsilon_{eff}} = 0 \quad (4)$$

The model known as the coherent potential approximation (CPA) assumes full coupling between neighboring inclusions. The formulation is shown as Equation 5:

$$\epsilon_{eff} = \epsilon_{major} + \frac{3f\epsilon_{eff}(\epsilon_{incl} - \epsilon_{major})}{3\epsilon_{eff} + (1-f)(\epsilon_{incl} - \epsilon_{major})} \quad (5)$$

These formulations can be combined into a unified mixing rule (Equation 6) where the formulation contains a unitless parameter v (Sihvola 1999).

$$\frac{\epsilon_{eff} - \epsilon_{major}}{\epsilon_{eff} + 2\epsilon_{major} - v(\epsilon_{eff} - \epsilon_{major})} = \frac{f(\epsilon_{incl} - \epsilon_{major})}{\epsilon_{incl} + 2\epsilon_{major} + v(\epsilon_{incl} - \epsilon_{major})} \quad (6)$$

The value of v produces the three previous formulations: $v = 1$ produces the Maxwell-Garnet formulation, $v = 2$ produces the Bruggeman formulation, and $v = 3$ gives the coherent potential approximation.

Results and Discussion

In the following subsections, the results of the calculations are organized by the optical properties of the components such that major and inclusion components with similar optical properties are viewed together. To this end, soot and dust are categorized as absorbing components while water and sulfate are categorized as non-absorbing. Refractive indices are computed for each two-component mixture using the volume fraction, f, of the inclusion equal to 0.0001, 0.01, 0.1. The formulations explored are all expected to be the same for f < 0.1. It is only for f > 0.1 that the formulations are expected to differ (Sihvola 1999). The point at which this difference occurs is important to note when choosing a model to use.

The computed effective refractive indices are reported here in terms of the real term which indicates stronger light scattering as it increases and the imaginary term which indicates more light absorption by the mixture as it increases. The terms of the refractive indices are split when recorded to facilitate the observation of trends within the formulations.

Non-absorbing materials with non-absorbing inclusions

There are two combinations of materials that are included in this category: water with sulfate inclusions, and sulfate with water inclusions. Both materials have very small imaginary refractive index terms at 550 nm which indicate that they do not efficiently absorb light at this wavelength. Table 1 includes the imaginary term for the refractive index, n_{eff}, for these mixtures, and Table 2 includes the real term.

The results of these formulations are not unexpected. In the case of water with sulfate inclusions, all of the formulations produce the same imaginary term for n_{eff}.

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For sulfate with water inclusions, the CPA produces a slightly larger imaginary term at f =0.1. That is also the only instance that an EMA formulation produced an imaginary term larger than that of the linear average.

Table 1: Imaginary term of n_{eff} computed for the linear average (Lin); Maxwell-Garnet (MG); Bruggeman (B); and coherent potential (CPA) formulations for listed volume fractions, f.

	Water with Sulfate Inclusions			
f =	Lin	MG	B	CPA
0.0001	1.97E-09	1.96E-09	1.96E-09	1.96E-09
0.01	2.94E-09	1.96E-09	1.96E-09	1.96E-09
0.1	1.18E-08	1.05E-08	1.05E-08	1.05E-08
	Sulfate with Water Inclusions			
f =	Lin	MG	B	CPA
0.0001	1.00E-07	1.00E-07	1.00E-07	1.00E-07
0.01	9.90E-08	9.88E-08	9.88E-08	9.88E-08
0.1	9.02E-08	8.94E-08	8.94E-08	9.06E-08

Table 2: Real term of n_{eff} computed for the linear average (Lin); Maxwell-Garnet (MG); Bruggeman (B); and coherent potential (CP) formulations for listed volume fractions, f.

	Water with Sulfate Inclusions			
f =	Lin	MG	B	CP
0.0001	1.33	1.33	1.33	1.33
0.01	1.33	1.33	1.33	1.33
0.1	1.35	1.35	1.35	1.35
	Sulfate with Water Inclusions			
f =	Lin	MG	B	CP
0.0001	1.47	1.47	1.47	1.47
0.01	1.47	1.47	1.47	1.47
0.1	1.46	1.46	1.46	1.46

The real terms of n_{eff} are all the same as the linear average. In both cases, you can begin to see the models differing only at f = 0.1 indicating that below this volume fraction, the interactions between inclusions is negligible.

Absorbing materials with absorbing inclusions

This category contains two combinations: soot with dust inclusions and dust with soot inclusions. The imaginary and real terms of the computed n_{eff} are given in Tables 3 and 4 respectively.

In these combinations it is notable that even at the low inclusion volume fractions, the imaginary term of the refractive index changes significantly in the dust mixture with soot inclusions indicating the large influence a small amount of a strong absorber can make on the overall material even when the major component is also somewhat absorbing. In the case of soot with dust inclusions, the linear average is only slightly larger than the EMA formulations. Where dust is the major component, this difference is larger. All EMA formulations produce identical imaginary terms at the volume fractions explored.

Table 3: Imaginary term of computed n_{eff}. Abbreviations are the same as in previous tables.

	Soot with Dust Inclusions			
f =	Lin	MG	B	CP
0.0001	4.40E-01	4.40E-01	4.40E-01	4.40E-01
0.01	4.36E-01	4.35E-01	4.35E-01	4.35E-01
0.1	3.97E-01	3.95E-01	3.95E-01	3.95E-01
	Dust with Soot Inclusions			
f =	Lin	MG	B	CP
0.0001	5.54E-03	5.54E-03	5.54E-03	5.54E-03
0.01	9.85E-03	9.63E-03	9.63E-03	9.63E-03
0.1	4.90E-02	4.70E-02	4.70E-02	4.70E-02

Table 4: Real term of computed n_{eff}. Abbreviations are the same as in previous tables.

	Soot with Dust Inclusions			
f =	Lin	MG	B	CP
0.0001	1.75	1.75	1.75	1.75
0.01	1.75	1.75	1.75	1.75
0.1	1.73	1.73	1.73	1.73
	Dust with Soot Inclusions			
f =	Lin	MG	B	CP
0.0001	1.53	1.53	1.53	1.53
0.01	1.53	1.53	1.53	1.53
0.1	1.55	1.55	1.55	1.55

As seen in Table 4, the real term of the computed n_{eff} is identical for all formulations and the linear average at these volume fractions.

Non-absorbing materials with absorbing inclusions

This category of particles includes either water or sulfate as the major component and soot or dust as the

inclusion. The imaginary and real terms of the computed n_{eff} are given in Tables 5 and 6 respectively. The values included here agree very well with data published previously (Erlick 2006). The few differences seem to be a result of the precision carried through the calculations and are differences of ± 1 in the last reported digit only. The values that differ from the published data are identified with an asterisk in Table 5.

Table 5: Imaginary term of computed n_{eff}. Abbreviations are the same as in previous tables. Values that differ from those reported previously are marked with an asterisk (Erlick 2006). It should be noted that the differences are insignificant and only occur in the last reported digit.

	Water with Dust Inclusions			
f =	Lin	MG	B	CP
0.0001	5.52E-07	5.18E-07*	5.18E-07*	5.18E-07*
0.01	5.50E-05	5.17E-05	5.17E-05	5.17E-05
0.1	5.50E-04	5.19E-04	5.21E-04	5.23E-04
	Water with Soot Inclusions			
f =	Lin	MG	B	CP
0.0001	4.40E-05	3.76E-05	3.76E-05	3.76E-05
0.01	4.40E-03	3.77E-03	3.78E-03*	3.78E-03
0.1	4.40E-02	3.80E-02*	3.87E-02	3.90E-02*
	Sulfate with Dust Inclusions			
f =	Lin	MG	B	CP
0.0001	6.50E-07	6.42E-07	6.42E-07	6.42E-07
0.01	5.51E-05	5.43E-05	5.43E-05	5.43E-05
0.1	5.50E-04	5.43E-04	5.43E-04	5.43E-04
	Sulfate with Soot Inclusions			
f =	Lin	MG	B	CP
0.0001	4.41E-05	4.08E-05	4.08E-05	4.08E-05
0.01	4.40E-03	4.07E-03	4.08E-03	4.08E-03
0.1	4.40E-02	4.10E-02	4.12E-02	4.12E-02

As expected, the addition of an absorbing inclusion causes a much larger increase in the imaginary term of n_{eff} as compared to that of the major component without inclusions. Similar to the previous mixtures studied, the linear average again provides the upper bound for the imaginary term when compared to the formulations at low volume fractions.

Also, the three EMA formulations are very similar for all volume fractions tested, although the mixtures

with water as the major component seem to already be beginning to produce differing estimations for the imaginary term at the $f = 0.1$ volume fraction. This may indicate that the models will not be equivalent at some moderately small volume fractions just over $f > 0.1$ and that care should be taken when choosing a formulation to use for particles modelled as a non-absorbing matrix with absorbing inclusions.

The real terms of the n_{eff} computed for non-absorbing materials with absorbing inclusions are provided in Table 6. In the case of a non-absorbing component with absorbing inclusions, the linear average again produced nearly the same value as all of the EMA formulations at all of the volume fractions tested.

Table 6: Real term of computed n_{eff}. Abbreviations are the same as in previous tables.

	Water with Dust Inclusions			
f =	Lin	MG	B	CP
0.0001	1.333	1.333	1.333	1.333
0.01	1.335	1.335	1.335	1.335
0.1	1.353	1.352	1.352	1.352
	Water with Soot Inclusions			
f =	Lin	MG	B	CP
0.0001	1.333	1.333	1.333	1.333
0.01	1.337	1.337	1.337	1.337
0.1	1.375	1.376	1.376	1.375
	Sulfate with Dust Inclusions			
f =	Lin	MG	B	CP
0.0001	1.470	1.470	1.470	1.470
0.01	1.471	1.471	1.471	1.471
0.1	1.476	1.476	1.476	1.476
	Sulfate with Soot Inclusions			
f =	Lin	MG	B	CP
0.0001	1.470	1.470	1.470	1.470
0.01	1.473	1.473	1.473	1.473
0.1	1.498	1.500	1.500	1.500

Absorbing materials with non-absorbing inclusions

The final category of mixtures considered is that of an absorbing material with non-absorbing inclusions. The real and imaginary terms of n_{eff} are provided in Tables 7 and 8. The main component of these particles is either soot or dust, and the inclusion is taken to be water or sulfate.

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As can be seen in Table 7, the imaginary term of the soot with water inclusions mixture decreases in this instance, and all formulations result in the same value as the linear average. It is only a very slight amount larger than the value from the other formulations.

It is also striking that the effect of either sulfate or water inclusions is virtually identical on the computed imaginary term in both the dust and soot cases.

Table 7: Imaginary term of computed n_{eff}. Abbreviations are the same as in previous tables.

	Soot with Water Inclusions			
f =	Lin	MG	B	CP
0.0001	4.40E-01	4.40E-01	4.40E-01	4.40E-01
0.01	4.36E-01	4.35E-01	4.35E-01	4.35E-01
0.1	3.96E-01	3.95E-01	3.94E-01	3.94E-01
	Soot with Sulfate Inclusions			
f =	Lin	MG	B	CP
0.0001	4.40E-01	4.40E-01	4.40E-01	4.40E-01
0.01	4.36E-01	4.35E-01	4.35E-01	4.35E-01
0.1	3.96E-01	3.94E-01	3.94E-01	3.94E-01
	Dust with Water Inclusions			
f =	Lin	MG	B	CP
0.0001	5.50E-03	5.50E-03	5.50E-03	5.50E-03
0.01	5.45E-03	5.44E-03	5.44E-03	5.44E-03
0.1	4.95E-03	4.94E-03	4.93E-03	4.93E-03
	Dust with Sulfate Inclusions			
f =	Lin	MG	B	CP
0.0001	5.50E-03	5.50E-03	5.50E-03	5.50E-03
0.01	5.44E-03	5.44E-03	5.44E-03	5.44E-03
0.1	4.95E-03	4.94E-03	4.94E-03	4.94E-03

As is the pattern for the other combinations of materials, the real term of n_{eff} seems to be predicted identically regardless of the EMA used. These are also identical to the linear average.

Conclusions

The effective refractive indices (n_{eff}) of various two-component materials have been calculated using various EMA formulations as well as a simple linear average. Effective refractive indices were computed at volume fractions (f) of inclusions in the matrix material equal to 0.0001, 0.01, and 0.1 which is within the region that the EMAs are thought to be valid. The main

difference between the formulations is the amount of interaction included between the inclusions themselves. At low volume fractions the formulations should produce identical results (Sihvola 1999). Comparison of results computed through these models generally support that conclusion.

The values of the real term computed for any of the two-component combinations show that no formulation produces a difference from the linear average. This is true at any volume fraction tested.

Table 8: Real term of computed n_{eff}. Abbreviations are the same as in previous tables.

	Soot with Water Inclusions			
f =	Lin	MG	B	CP
0.0001	1.750	1.750	1.750	1.750
0.01	1.746	1.746	1.746	1.746
0.1	1.708	1.707	1.707	1.707
	Soot with Sulfate Inclusions			
f =	Lin	MG	B	CP
0.0001	1.750	1.750	1.750	1.750
0.01	1.747	1.747	1.747	1.747
0.1	1.722	1.721	1.722	1.722
	Dust with Water Inclusions			
f =	Lin	MG	B	CP
0.0001	1.530	1.530	1.530	1.530
0.01	1.528	1.528	1.528	1.528
0.1	1.510	1.510	1.510	1.510
	Dust with Sulfate Inclusions			
f =	Lin	MG	B	CP
0.0001	1.530	1.530	1.530	1.530
0.01	1.529	1.529	1.529	1.529
0.1	1.518	1.524	1.524	1.524

In general, the data supports the understanding that the linear average serves as an upper bound on the imaginary term of the refractive index (Sihvola 1999), although in some cases this term is the same magnitude as the ones computed with the EMA formulations.

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