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# Applicability of the effective-medium approximation to heterogeneous aerosol particles

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ABSTRACT

The effective-medium approximation (EMA) is based on the assumption that a heterogeneous particle can have a homogeneous counterpart possessing similar scattering and absorption properties. We analyze the numerical accuracy of the EMA by comparing superposition *T*-matrix computations for spherical aerosol particles filled with numerous randomly distributed small inclusions and Lorenz–Mie computations based on the Maxwell-Garnett mixing rule. We verify numerically that the EMA can indeed be realized for inclusion size parameters smaller than a threshold value. The threshold size parameter depends on the refractive-index contrast between the host and inclusion materials and quite often does not exceed several tenths, especially in calculations of the scattering matrix and the absorption cross section. As the inclusion size parameter approaches the threshold value, the scattering-matrix errors of the EMA start to grow with increasing the host size parameter and/or the number of inclusions. We confirm, in particular, the existence of the effective-medium regime in the important case of dust aerosols with hematite or air-bubble inclusions, but then the large refractive-index contrast necessitates inclusion size parameters of the order of a few tenths. Irrespective of the highly restricted conditions of applicability of the EMA, our results provide further evidence that the effective-medium regime must be a direct corollary of the macroscopic Maxwell equations under specific assumptions.

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## 1. Introduction

Owing to its unparalleled simplicity, the effective-medium approximation (EMA) has been widely used to model complex heterogeneous substances as being homogeneous and having a refractive index computed with one of the phenomenological mixing rules such as the Lorentz–Lorenz, Bruggeman, and Maxwell-Garnett formulas [1,2]. Applications of various mixing rules in remote sensing, atmospheric radiation, and climate modeling research have been so ubiquitous that it would hardly be possible to assemble a representative list of relevant publications as it would contain hundreds of entries.

The use of the EMA appears to be unavoidable in many cases given the extreme morphological complexity of the vast majority of aerosol particles (e.g., [3–12]). The main cause of this situation is the limited applicability of direct computer solvers of the Maxwell equations to representative ensembles of heterogeneous particles. It should be kept in mind however that, in the words of Chýlek et al. [2], mixing rules have always been heuristic shortcuts not derived explicitly from the Maxwell equations. As a consequence, the accuracy of such *ad hoc* effective refractive indices (ERIs) and the precise conditions for their permissible use have often been difficult to assess.

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Despite the obvious shortcomings of the EMA, its applications in remote sensing and climate research can be expected to be as widespread in the future as they have been in the past. For example, the use of the concept of an ERI has been and is expected to remain *implicit* in virtually all computations of electromagnetic scattering by dust-like aerosols since it has become the norm to ignore the internal heterogeneity of such particles. Therefore, the failure of this concept in application to dust-like aerosols may create an extremely problematic situation. This makes it imperative to perform an in-depth analysis of the range and conditions of practical applicability of the EMA.

This analysis has been initiated in several recent publications [13–18] by taking advantage of the latest improvements in first-principle modeling methodologies [19]. The overall outcome of these studies can hardly be characterized as optimistic. It should be recognized however that some of these initial results are based on a few rather artificial models of heterogeneous aerosols that do not necessarily offer the EMA a "fair chance". Indeed, it is well-known that the concept of bulk refractive index is a byproduct of deriving the macroscopic Maxwell equations from the microscopic Maxwell–Lorentz equations dealing with discrete elementary charges [20–30]. The main assumption in this derivation is that the microscopic electromagnetic field can be meaningfully homogenized over "physically infinitesimal" volume elements that are much smaller than the wavelength and yet contain vast numbers of molecules. It is obvious that the extrapolation of this approach to the case of a macroscopically heterogeneous material must also be based on the assumption that inclusions are quasi-uniformly distributed throughout the host medium, are sufficiently small, and are present in large numbers. Only then can one hope that the concept of the ERI might work.

We have already mentioned that a direct *analytical* derivation of the EMA from the Maxwell equations is still absent. However, the actual existence of the effective-medium regime has been demonstrated *numerically* by comparing Lorenz–Mie results for a homogeneous spherical particle with those obtained by applying the superposition *T*-matrix solver of the Maxwell equations to a spherical particle filled randomly with a large number of very small spherical inclusions [31]. The refractive indices of the host and of the inclusions were 1.33 and 1.55, respectively. The possibility to identify an ERI enabling the Lorenz–Mie theory to reproduce even the finest details of the angular profile of the scattering matrix demonstrated convincingly that the effective-medium concept must have physical validity. Yet the practical range of this validity may not necessarily be wide and may exclude many actual types of heterogeneous atmospheric particulates.

Given the great importance of mineral-dust aerosols in atmospheric radiative-transfer modeling and remote sensing, the main objective of this paper is to extend the analysis of [31] and demonstrate numerically the fundamental existence of the effective-medium regime in the case of two types of inclusions representing the largest refractive-index contrast with the mineral host, i.e., air bubbles and absorbing hematite grains. Furthermore, we trace and analyze the accumulation of errors of the EMA as the ideal conditions of the ERI regime are increasingly violated. A useful intermediate aspect of our study is a general analysis of the accuracy of the EMA as a function of the inclusion size parameter and of the refractive-index contrast between the host particle and the inclusions.

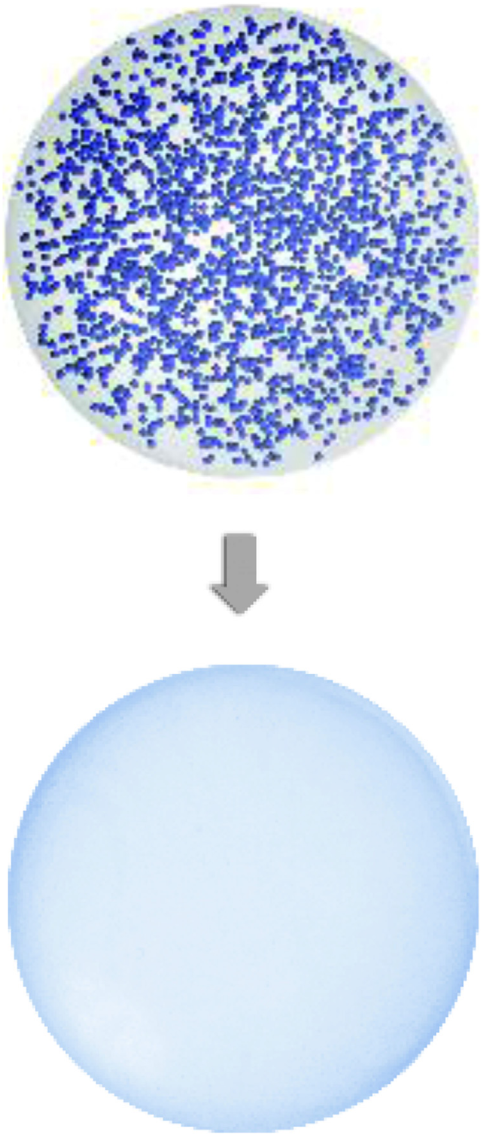

**Fig. 1.** The essence of the effective-medium approximation.

## 2. Modeling methodology

The gist of the EMA is illustrated in Fig. 1 wherein a particle randomly filled with numerous, quasi-uniformly distributed small inclusions is replaced by a homogeneous object of the same overall shape but with an artificial (effective) refractive index coinciding neither with that of the host nor with that of the inclusions. Given its artificial nature, the ERI carries no independent physical content and is useful only to the extent to which it can simplify the computation of relevant optical observables. In the case of atmospheric radiative-transfer and remote sensing research, all such observables can, in the final analysis, be expressed in terms of