

AA8: The aerosol refractive index was calculated using the Bruggeman mixing rule (or effective medium approximation), which is not a simple coated sphere assumption. The Bruggeman mixing rule is a part of more general mixing rule formulation resumed in Aspnes et al. (1982) that is as follows:

$$\frac{\epsilon_{eff} - \epsilon_h}{\epsilon_{eff} + 2\epsilon_h} = \sum_{i=1}^n f_i \frac{\epsilon_i - \epsilon_h}{\epsilon_i + 2\epsilon_h}$$

where ϵ_{eff} is the complex effective dielectric constant of the mixture ($m_{eff} \approx \sqrt{\epsilon_{eff}}$), ϵ_h represents the dielectric function of the host medium, ϵ_i and f_i are the complex dielectric constant, and the volume fraction, of the i -th component respectively.

Depending on the choice of host medium, we may obtain three different mixing rules: 1) Maxwell-Garnett (MG) if the host medium is one of the components ($\epsilon_i = \epsilon_h$) (Stier et al., 2007; Schuster et al., 2005; Bohren and Huffman, 1983; Aspnes 1982; Heller, 1965) and in this case it is possible to refer to the Maxwell-Garnett as coated sphere assumption; 2) Lorentz-Lorenz (LL) if the host medium is the vacuum ($\epsilon_h = 1$) (Liu and Daum, 2008; Aspnes 1982; Heller, 1965); 3) Bruggeman (BR) if no choice of host medium is made, and inclusions are considered embedded in the effective medium itself (Stier et al., 2007; Aspnes 1982; Heller, 1965, Bruggeman, 1935). Stier et al. (2007) and Aspnes (1982) point out that the BR mixing rule overcomes the dilemma of the choice of host medium among the various aerosol components. From this point of view, the BR mixing rule considers all possible positions of each component (BC, dust, water soluble materials...) in an aerosol particle, respect to the other components. Thus, it allows simulating the real complexity of aerosols and making the BR mixing rule suitable for use in calculating the aerosol m_{eff} . For this reason, the BR mixing doesn't consider a simple coated sphere assumption and implies that the left part of the aforementioned equation vanishes giving the equation 8 reported in the paper at page 553, line 24, section 2.3.1.

Finally, the Bruggeman mixing rule avoids the risk of overestimating the imaginary part (k) of m , thus reducing the uncertainties, as instead happens using both the linear volume-average and the linear mass-average mixing rules in the presence of highly absorbing inclusions (i.e. BC) in a non-absorbing medium (Stier et al., 2007; Lesins et al., 2002; Chýlek et al., 1995).

We would also underline that all the values (density, refractive indexes) used as input in the refractive index mixing rule were carefully chosen from the literature (especially for BC) considering only state-of-the-art values (i.e. see values in Bond and Bengtstrom, 2006) as reported in section 2.3.1 (see also AA2). Moreover, the E-AIM (used in the hygroscopic growth of the aerosol) was previously validated through measurements conducted in an Aerosol Chamber (results reported in Ferrero et al., 2014).