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A study on the sensitivities of simulated aerosol optical properties to composition and size distribution using airborne measurements

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HIGHLIGHTS

- A framework to calculate aerosol optical properties at a given RH is presented.
- Calculations are made based on the aerosol composition and size distribution.
- FAAM BAe-146 aircraft data are used in a closure study for 2 different aerosol types.
- Uncertainties associated to the calculated aerosol optical properties are discussed.
- Sources of uncertainty are refractive indices, hygroscopicity and size distribution.

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ABSTRACT

We present a flexible framework to calculate the optical properties of atmospheric aerosols at a given relative humidity based on their composition and size distribution. The similarity of this framework to climate model parameterisations allows rapid and extensive sensitivity tests of the impact of uncertainties in data or of new measurements on climate relevant aerosol properties. The data collected by the FAAM BAe-146 aircraft during the EUCAARI-LONGREX and VOCALS-REx campaigns have been used in a closure study to analyse the agreement between calculated and measured aerosol optical properties for two very different aerosol types. The agreement achieved for the EUCAARI-LONGREX flights is within the measurement uncertainties for both scattering and absorption. However, there is poor agreement between the calculated and the measured scattering for the VOCALS-REx flights. The high concentration of sulphate, which is a scattering aerosol with no absorption in the visible spectrum, made the absorption measurements during VOCALS-REx unreliable, and thus no closure study was possible for the absorption. The calculated hygroscopic scattering growth factor overestimates the measured values during EUCAARI-LONGREX and VOCALS-REx by ~30% and ~20%, respectively. We have also tested the sensitivity of the calculated aerosol optical properties to the uncertainties in the refractive indices, the hygroscopic growth factors and the aerosol size distribution. The largest source of uncertainty in the calculated scattering is the aerosol size distribution (~35%), followed by the assumed hygroscopic growth factor for organic aerosol (~15%), while the predominant source of uncertainty in the calculated absorption is the refractive index of organic aerosol (28–60%), although we would expect the refractive index of black carbon to be important for aerosol with a higher black carbon fraction.

1. Introduction

Atmospheric aerosols affect the Earth's climate both directly, through the scattering and absorption of radiation (Charlson et al., 1992; Haywood and Shine, 1997), and indirectly, via changes to

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cloud microphysics and properties (Kaufman et al., 2005). Moreover, aerosols also affect visibility and air quality (Horvath, 1995), as well as human health (Lelieveld et al., 2002; Chu et al., 2003; Wang and Christopher, 2003).

In order to estimate the direct effect, climate models generally require aerosol optical properties such as the extinction coefficient, the single scattering albedo and the asymmetry parameter. For these, they need to quantify first the spectral refractive index, the size distribution, the hygroscopicity and the mixing state (internal or external) of atmospheric aerosols. Each of these properties is a complex function of aerosol size, composition, and chemical and physical processing (including impacts of humidity and clouds). Thus, due to this complexity of the atmospheric aerosols, we need to use models and measurements combined together in order to provide the information needed in climate models.

Closure between the measured aerosol scattering and absorption and that calculated with a scattering code using chemical composition and particle size information has been attempted before by several studies (Cai et al., 2011; Highwood et al., 2012; Liu et al., 2013; Quinn and Coffman, 1998; Sciare et al., 2005; Wex et al., 2002). However, recent additions to the instrumentation aboard the Facility for Airborne Atmospheric Measurements (FAAM) BAe-146 aircraft have made possible the measurement of aerosol scattering as a function of relative humidity and black carbon mass, allowing more accurate closure studies to be performed.

In this work we present a flexible framework for assessing parameterizations of optical properties and hygroscopic growth of aerosols. This framework is used to calculate the optical properties of atmospheric aerosols at a given relative humidity based on their composition and size distribution, which can then be compared with measured values of the same quantities. In our case, the FAAM BAe-146 aircraft provides measurements of the chemical composition, microphysical, optical and hygroscopic properties of the atmospheric aerosols (Johnson et al., 2000; Osborne et al., 2007; McMeeking et al., 2010; Morgan et al., 2010a), which allow us to explore here the agreement between models and measurements of the aerosol optical properties for two very different aerosol types. Section 2 of this paper describes the framework and the data from the FAAM BAe-146 aircraft used. Section 3 presents the closure study of the aerosol optical properties. Section 4 discusses the uncertainties associated to the calculated aerosol optical properties. The work's conclusions are presented in Section 5.

2. Methodology

2.1. Framework

We have developed a flexible framework to calculate the scattering and absorption by atmospheric aerosols at a given relative humidity based on the composition and size distribution. The framework can be used with different scattering codes and mixing states, but here we use Mie scattering for homogeneous internally mixed spheres. Although aerosols, and particularly black carbon (Hess et al., 1998), are not always spherical, this assumption is valid for well-mixed anthropogenic aerosols, especially in moderately humid environments (Highwood et al., 2012), and is frequently used for most anthropogenic aerosol types (Quinn and Coffman, 1998; Wex et al., 2002; Sciare et al., 2005; Cai et al., 2011; Costabile et al., 2013). The way in which the different components are distributed within the aerosol particles is referred to as mixing state, which ranges from external to homogeneous internal mixture. An external mixing state is an appropriate assumption for freshly emitted aerosols, which have not had time to undergo chemical reaction or coalescence. An internal mixture is a better assumption for older, well-mixed aerosol (Raes et al., 2000). Well-mixed anthropogenic aerosols can usefully be modelled as having a homogeneous internal mixing state, while a core and shell model would be more appropriate if a large mass of black carbon was present (Abel et al., 2003). Although our framework includes the possibility of choosing between this whole range of mixing states, since the cases considered here are of well-mixed anthropogenic aerosols with none or small amounts of black carbon, we will focus on the homogeneous internal mixing case.

In this framework, the mass concentration of the different aerosol components, as measured by an Aerosol Mass Spectrometer (AMS), and hygroscopicity values for each component taken from literature, are combined using the Zdanovskii–Stokes–Robinson (ZSR) volume mixing rule (Zdanovskii, 1948; Stokes and Robinson, 1966). This assumes that the components of the mixed aerosol do not interact, in order to calculate the hygroscopic growth factor of the internally mixed aerosol following the equation

$$HGF_{\mathrm{mix}} = \left(\sum_i \varepsilon_i HGF_i^3 \right)^{1/3} \quad (1)$$

where ε_i is the volume fraction of component i in the dry particle and HGF_i is the hygroscopic growth factor of the pure component i.

The ambient size distribution is then calculated by applying this mixed growth factor to the dry size distribution. Next, the mass of water taken up by the aerosol is calculated by comparing the average volume of the dry aerosol (based on the average radius from the dry size distribution) with that of the ambient aerosol (based on the average radius from the ambient size distribution). By including this water as an additional chemical component, it is then possible to calculate the refractive index of the internally mixed aerosol at a given relative humidity, and for a variety of wavelengths, by applying the ZSR volume mixing rule. The resultant ambient size distribution and refractive index are then passed in this case to the Mie scattering code of Wiscombe (1979) in order to calculate the aerosol optical properties.

Although other similar frameworks exist, including OPAC (Optical Properties of Aerosols and Cloud) by Hess et al. (1988) which is still widely used to specify aerosol for use in satellite retrievals, this framework is much more flexible, allowing the use of composition and size distributions directly. In addition, since it is closer to the parameterisations used in climate models, it allows convenient and rapid testing of the impact of uncertainties in data, or new measurements on climate relevant aerosol properties.

2.1.1. Refractive indices

The refractive indices of major aerosol components such as ammonium sulphate, ammonium nitrate, black carbon and organic aerosol assumed by the framework are based on a literature review of field observations and laboratory studies. The refractive index for sulphate, which is a scattering aerosol with no absorption in the visible spectrum, is taken from Toon et al. (1976). However, the refractive index for nitrate, another scattering aerosol with no absorption in the visible spectrum, is not well characterized although it is an important contributor to light scattering in the atmosphere (Diederen et al., 1985; Brink et al., 1996). In this framework, we use a single value with no absorption component from Weast (1985) below 0.7 μm, the values from Gosse et al. (1997) in the intermediate range and the values from Jarzembski et al. (2003) in the infrared. Due to technical issues in the measurement of the abundance and optical properties of black carbon, which is highly absorbing in the visible spectrum, there is considerable debate regarding the most appropriate value for its refractive index (Stier et al., 2007). We use here the more absorbing refractive indices from Bond and Bergstrom (2005). The refractive index of organic