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

A.R. Esteve [a,b,*], E.J. Highwood [b], W.T. Morgan [c], G. Allen [c], H. Coe [c], R.G. Grainger [d], P. Brown [e], K. Szpek [e]

[a] *Department of Earth Physics and Thermodynamics, University of Valencia, Spain*
[b] *Department of Meteorology, University of Reading, UK*
[c] *Centre for Atmospheric Science, The University of Manchester, UK*
[d] *Atmospheric, Oceanic and Planetary Physics, University of Oxford, UK*
[e] *The Met Office, UK*

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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* Corresponding author. Departament de Física de la Terra i Termodinàmica, Universitat de València, C/Dr. Moliner 50, 46100 Burjassot, Valencia, Spain.
E-mail address: anna.esteve@uv.es (A.R. Esteve).

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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

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

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 aerosol is difficult to define because its properties vary according to source, location, combustion type and aerosol age. In this framework, we use a refractive index based on that of Swannee River Fulvic Acid (SRFA) at 532 nm (Dinar et al., 2008), with the wavelength dependence of Kirchstetter et al. (2004) in the imaginary part between 350 and 700 nm, and being wavelength independent in the real part between 400 and 700 nm. For wavelengths above 4 μm, the wavelength dependence for the water soluble type from Hess et al. (1998) is used. SRFA was assumed since it can be considered to be representative of aged organic aerosol (McFiggans et al., 2005), which was found in the measurement campaigns considered here. The refractive indices at 550 nm assumed by the framework for these major aerosol components are specified in Table 1.

2.1.2. Hygroscopic growth factors

The hygroscopic growth factors of major aerosol components such as ammonium nitrate, ammonium sulphate and black carbon have been well studied; that of organic aerosol is worse known. The hygroscopic growth factors of sulphate and nitrate depend strongly on the ambient relative humidity, and the values reported in the literature are either derived from or in agreement with the values reported in Tang (1996). However, while the growth factors of ammonium sulphate and ammonium nitrate depend on the initial size of the aerosol, data relating growth factor and initial aerosol size are very limited. Black carbon is a hydrophobic aerosol, and it is generally accepted that its growth factor is approximately 1, and independent of the ambient relative humidity or the initial aerosol size (Forster et al., 2007; McMeeking et al., 2011). The growth factor of organic aerosol is a complex function of its component organic compounds, the combustion processes which produced them, chemical processing in the atmosphere and mixing with ambient aerosol (Forster et al., 2007). Studies of individual organic compounds (Brooks et al., 2004; Prenni et al., 2003; Kanakidou et al., 2005), as well as various hygroscopic closure studies (Gysel et al., 2004, 2007; McFiggans et al., 2005), have generally found a modest growth factor for organic aerosol, and it is not thought to depend on the initial aerosol size (Brooks et al., 2004). The hygroscopic growth factors at a relative humidity of 80% assumed by the framework for these major aerosol components are specified in Table 1.

2.2. FAAM BAe-146 aircraft data

The instrumentation aboard the FAAM BAe-146 aircraft measures the chemical composition, microphysical, optical and hygroscopic properties of the atmospheric aerosols, and it has been described in detail in Johnson et al. (2000), Osborne et al. (2007), McMeeking et al. (2010) and Morgan et al. (2010a). In this study, we have used the data collected by the FAAM BAe-146 aircraft during the European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions Long Range Experiment (EUCAARI-LONGREX, Kulmala et al., 2009) and the VAMOS Ocean-Cloud-Atmosphere-Land Regional Experiment (VOCALS-REx, Wood et al., 2011). The EUCAARI-LONGREX campaign consisted of 15 flights over central Europe or off the UK coast (47–57°N and 12°W–22°E) during May 2008, and its meteorology and aerosol measurements have been fully discussed by McMeeking et al. (2010), Morgan et al. (2010a, 2010b), Hamburger et al. (2011) and Highwood et al. (2012). The VOCALS-REx campaign consisted of 10 flights over the South East Pacific region (20°S and 70–85°W) between October and November 2008, and the aerosol measurements made by the FAAM BAe-146 aircraft during this campaign have been described by Allen et al. (2011). Each flight for both campaigns consisted of a number of straight level runs (SLR) at different altitudes with varying time durations.

The motivation for using these two campaigns to explore the agreement between modelled and measured aerosol optical properties comes from the very different chemical composition of the atmospheric aerosols. Fig. 1 shows the mean mass concentration (in percentage) of the main aerosol components (nitrate, sulphate, organic matter and black carbon) for each of the flights during the two campaigns. During EUCAARI-LONGREX, aerosols were mainly composed of nitrate (26%), sulphate (27%) and organic matter (46%), with small concentrations of black carbon (1%) (Morgan et al., 2010a; McMeeking et al., 2011). However, during VOCALS-REx, the aerosol composition was clearly dominated by sulphate (85%) (Allen et al., 2011), thus potentially representing a

Table 1
Refractive indices and hygroscopic growth factors for aerosol components assumed by the framework.

Component	Refractive index at 550 nm	Hygroscopic growth factor at RH = 80%
Ammonium sulphate	1.53−0*i* (Toon et al., 1976)	1.50 (Tang, 1996)
Ammonium nitrate	1.611−0*i* (Weast, 1985; Gosse et al. 1997; Jarzembski et al. 2003)	1.50 (Tang, 1996)
Black carbon	1.95−0.79*i* (Bond and Bergstrom, 2005)	1.00 (Forster et al., 2007)
Organic aerosol	1.538−0.02*i* (Dinar et al., 2008; Kirchstetter et al., 2004)	1.07 (Brooks et al., 2004)

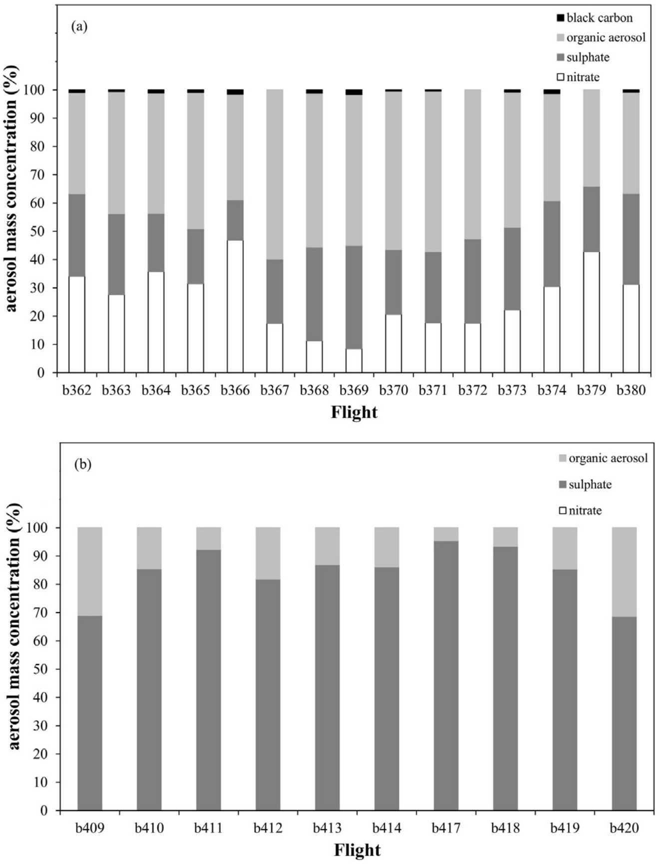

Fig. 1. Average mass concentration (in percentage) of main aerosol components for each flight of a) EUCAARI-LONGREX and b) VOCALS-REx (Morgan et al., 2010a; McMeeking et al., 2011; Allen et al., 2011). During VOCALS-REx, nitrate concentrations were not registered above the detection limit of the measurement instrument and black carbon concentrations were not measured.

“simpler” aerosol system, although there are subtle differences between composition data reported by the various VOCALS-REx studies (Hawkins et al., 2010; Lee et al., 2010; Allen et al., 2011; Kleinman et al., 2012; Twohy et al., 2013). Ammonium sulphate, found during EUCAARI-LONGREX (Morgan et al., 2010a), is assumed for VOCALS-REx, and the differences regarding the composition of the sulphate aerosol (e.g. sulphuric acid reported by Lee et al. (2010)) show little impact on our results. Nitrate is not reported for VOCALS-REx since its concentrations were not registered above the detection limit of the AMS. Additionally, the instrument aboard the FAAM BAe-146 aircraft to measure black carbon was not operational during the VOCALS-REx campaign. Furthermore, additional motivation for using these two campaigns is the different ambient relative humidity conditions, which would be expected to also have a significant impact on the aerosol properties. The ambient relative humidity for the SLR used in this study during the EUCAARI-LONGREX campaign was in the range 29–87%, with an average value of 52%, while during the VOCALS-REx campaign it was in the range 70–92%, with an average value of 85%.

3. Results

The aerosol scattering and absorption measured by the FAAM BAe-146 aircraft are averaged for each SLR of each flight of the EUCAARI-LONGREX and VOCALS-REx campaigns. These are assumed to be applicable to “dry” aerosol, and subsequently the scattering for “ambient” aerosol at a given relative humidity is derived from the dry and wet nephelometer system aboard the BAe-146 aircraft following the procedure described in Highwood et al. (2012). In this, a hygroscopicity curve is obtained for each flight of the campaign by plotting the hygroscopic scattering growth factor, $f(\mathrm{RH})$, which is the ratio of the scattering coefficient measured in the wet nephelometer to the scattering coefficient measured by the dry nephelometer, as a function of relative humidity, and fitting the data to Model 2 from Kotchenruther et al. (1999)

$$f(\mathrm{RH}) = \sigma_{s,d}\left(1 + a\left(\frac{\mathrm{RH}}{100}\right)^{b}\right) \qquad (2)$$

where $\sigma_{s,d}$, a, and b are fitting parameters to the data. Only data where RH < 30% were used to ensure that the measurements were as reasonable a representation of dry aerosol as possible. The scattering for “ambient” aerosol at a given relative humidity can then be derived by increasing the scattering from the nephelometer by the growth factor indicated by the hygroscopicity curve. These aerosol optical properties can also be calculated by the framework presented in Section 2 using the composition and size distribution measurements made by the FAAM BAe-146 aircraft at each SLR in every flight of both campaigns. Fig. 2 shows the comparison of the calculated values of the aerosol scattering at 550 nm with the measured values for “ambient” aerosol averaged for each SLR in every flight of EUCAARI-LONGREX and VOCALS-REx. There is very good agreement for all flights of EUCAARI-LONGREX, with the calculated scattering being within the 30% uncertainty of the measured scattering (assuming a measurement uncertainty of 20% and variability in the scattering during each SLR in the range 12%–35% (Highwood et al., 2012)), and slightly underestimating the measured values by ~11%. These results are in agreement with those obtained by Highwood et al. (2012) for dry aerosol during the EUCAARI-LONGREX campaign. However, the agreement for the flights of VOCALS-REx is very poor, with the calculated scattering clearly overestimating the measured values, and being outside of the 30% measurement uncertainty. The bias between the calculated and measured scattering could be due to the uncertainty in the choice of hygroscopic growth factors, especially those for organic aerosol and sulphate. Moreover, the different sign of this bias observed between EUCAARI-LONGREX and VOCALS-REx could be due to the different aerosol size distributions used to calculate the scattering and absorption for both campaigns. An aerosol size distribution with 15 bins ranging from 0.1 to 3 μm particle diameter recorded aboard the FAAM BAe-146 aircraft was used for each SLR of each flight of EUCAARI-LONGREX. However, the lognormal fit parameters to campaign-mean aerosol size distributions from Allen et al. (2011) were used for each SLR of each flight of VOCALS-REx. The sensitivity of our results to the different type of aerosol size distribution will be tested in Section 4.

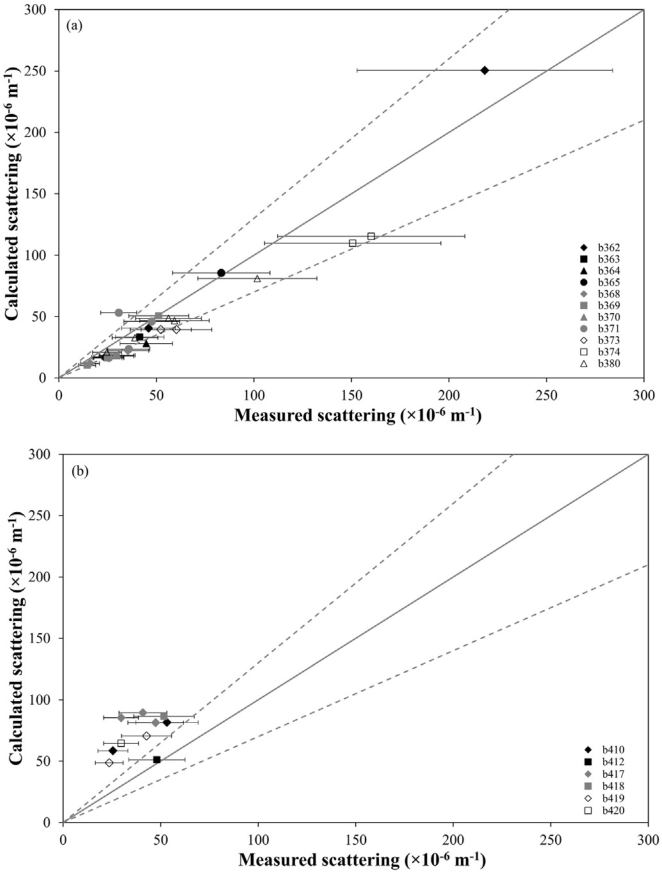

Fig. 2. Comparison of calculated with measured scattering at 550 nm for “ambient” aerosol averaged for each run in every flight from a) EUCAARI-LONGREX and b) VOCALS-REx. The 1:1 line is indicated (solid line) and also the ±30% tolerances (dashed lines). Horizontal error bars show the 30% uncertainty in measured scattering.

Fig. 3 shows the comparison of the calculated values of the aerosol absorption at 550 nm with the measured ones for “dry” aerosol averaged for each SLR in every flight of EUCAARI-LONGREX. The comparison for VOCALS-REx is not reported because the absorption registered for all flights of the campaign was below the detection limit of the Particle Soot Absorption Photometer (PSAP), in agreement with the fact that the aerosol composition during VOCALS-REx was clearly dominated by sulphate, which is a scattering aerosol with no absorption in the visible spectrum. The calculated absorption underestimates the measured values for most of the flights of EUCAARI-LONGREX, although the agreement between calculated and measured absorption is within the 50% uncertainty of the measurements (assuming a measurement uncertainty of 30% and variability in the absorption during each SLR in

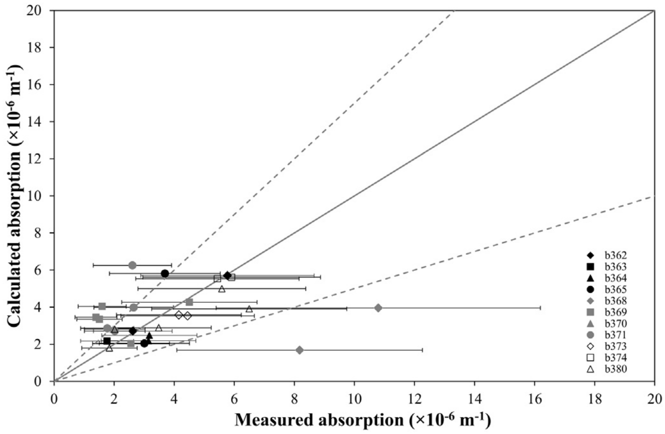

Fig. 3. Comparison of calculated with measured absorption at 550 nm for "dry" aerosol averaged for each run in every flight from EUCAARI-LONGREX. The 1:1 line is indicated (solid line) and also the ±50% tolerances (dashed lines). Horizontal error bars show the 50% uncertainty in measured absorption.

the range 16%–48% (Highwood et al., 2012)). These results are in agreement with those obtained by Highwood et al. (2012) for dry aerosol during the EUCAARI-LONGREX campaign. The bias between the calculated and measured absorption could be due to the uncertainty in the choice of refractive indices, especially those for black carbon and organic aerosol, although we expect the effect of the refractive index of black carbon to be weak here due to the small concentrations of black carbon registered during EUCAARI-LONGREX.

Fig. 4 shows the comparison of the calculated values of *f*(RH) (derived from the calculated scattering for "dry" and "ambient" aerosol) at 550 nm with the measured ones (derived from the measured scattering for "dry" and "ambient" aerosol) averaged for each SLR in every flight of EUCAARI-LONGREX and VOCALS-REx. There is poor agreement for all flights of EUCAARI-LONGREX, with the calculated *f*(RH) clearly overestimating the measured values by ∼30%. There is slightly better agreement for all flights of VOCALS-REx, with the calculated *f*(RH) overestimating the measured values by ∼20%. However, due to the large uncertainty of the measured *f*(RH) of 60%, the agreement between calculated and measured values is well within the uncertainty of the measurements. The bias between the calculated and measured hygroscopic scattering growth factor would be due to the same factors causing the bias between the calculated and measured scattering, i.e., the uncertainty in the choice of hygroscopic growth factors, especially those for organic aerosol and sulphate, and in the aerosol size distribution.

Although in Figs. 2–4 we have shown the uncertainty in the measurements but not in the calculations, uncertainty also exists in the calculated values mainly due to uncertainties in the refractive indices, the hygroscopic growth factors and the aerosol size distribution. Probably the uncertainty is higher for the EUCAARI-LONGREX campaign since it is chemically more complex than the VOCALS-REx campaign, the latter being dominated by sulphate which has been moderately well studied. In the following section we assess the sensitivity of the calculated scattering and absorption to these factors.

4. Discussion

To test the sensitivity of the calculated scattering and absorption to the choice of refractive indices, particularly those for black carbon and organic aerosol, we have repeated our calculations using different values of those refractive indices, and we have compared the new results with the old ones. There is still considerable debate regarding the most appropriate value for the refractive index of black carbon (Stier et al., 2007). Originally we used the "high absorbing" refractive index from Bond and Bergstrom (2005) (1.95–0.79*i*), but we could have used the value from Hess et al. (1998) (1.77–0.44*i*) or the "medium absorbing" refractive index suggested by Stier et al. (2007) (1.85–0.71*i*). Only the flights for the EUCAARI-LONGREX campaign have been used in this test. The sensitivity (in percentage) of the calculated scattering and absorption to assumptions in refractive indices of black carbon (RI_BC) and organic aerosol (RI_OC), hygroscopic growth factors of organic aerosol (GF_OC) and sulphate (GF_SU) and aerosol size distribution (SIZE_DISTR) is shown in a box diagram in Fig. 5. The dividing segment in the box is the median. The bottom/top box limits represent the 1st and 3rd quartiles. The box bars represent the minimum and maximum. Absorption is more sensitive than scattering to the refractive index for black carbon, but the effect here is very weak (∼2% on average and up to ∼5% for absorption) due to the small concentrations of black carbon present during EUCAARI-LONGREX (McMeeking et al., 2011). The refractive index for organic aerosol has therefore much more impact, especially in the absorption. All flights for both the EUCAARI-LONGREX and VOCALS-REx campaigns have been used in this test. Removing all absorption by organic aerosol as suggested in some previous

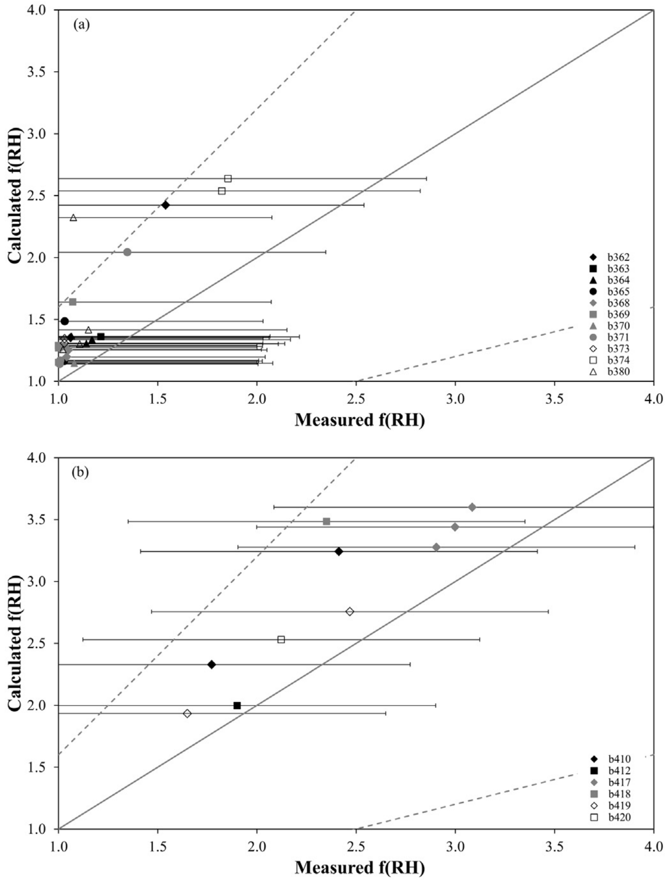

Fig. 4. Comparison of calculated with measured hygroscopic scattering growth factor at 550 nm averaged for each run in every flight from a) EUCAARI-LONGREX and b) VOCALS-REx. The 1:1 line is indicated (solid line) and also the ±60% tolerances (dashed lines). Horizontal error bars show the 60% uncertainty in measured *f*(RH). The hygroscopic scattering growth factor, which is the ratio of the scattering of the "ambient" aerosol to the scattering of the "dry" aerosol, must be always greater than 1.0, although the error bars showing the 60% uncertainty in the measured values extend below this value.

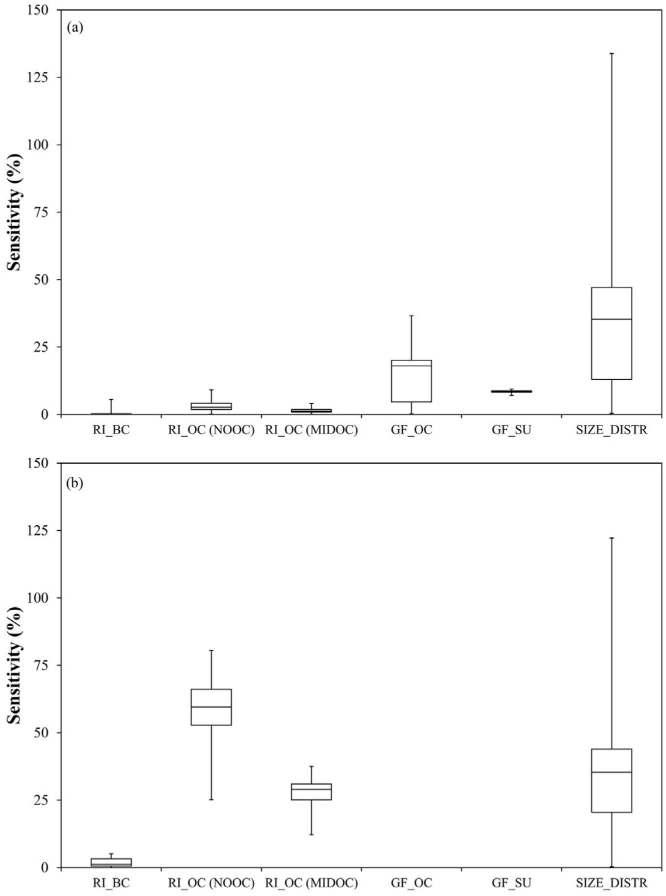

Fig. 5. Sensitivity (in percentage) of the calculated (a) scattering for “ambient” aerosol and (b) absorption for “dry” aerosol to assumptions in refractive indices of black carbon (RI_BC) and organic aerosol (RI_OC), hygroscopic growth factors of organic aerosol (GF_OC) and sulphate (GF_SU) and aerosol size distribution (SIZE_DISTR). The dividing segment in the box is the median. The bottom/top box limits represent the 1st and 3rd quartiles. The box bars represent the minimum and maximum.

studies ($1.538 - 0i$) produces changes in the calculated absorption of ~60% on average and up to ~81%, while the changes in the calculated scattering are of ~3% on average and up to ~9%. Having some weak absorption by organic aerosol ($1.538 - 0.01i$) reduces the changes in the calculated absorption and scattering to ~28% and ~1.4% on average, respectively. These observed changes worsen the agreement between the measured and calculated scattering and absorption. Following Highwood et al. (2012), RI_OC (NOOC) and RI_OC (MIDOC) in Fig. 5 refer to an assumption of no absorption and some weak absorption by organic carbon, respectively.

The sensitivity to the choice of hygroscopic growth factors, particularly those for organic aerosol and sulphate, is tested here for the calculated scattering. Most studies report hygroscopic growth factors for organic aerosol in the range of 1–1.65 (Topping et al., 2005a; Kanakidou et al., 2005; Varutbangkul et al., 2006), with a mean value of 1.20 (Gysel et al., 2007). We have repeated our scattering calculations for all flights of EUCAARI-LONGREX and VOCALS-REx using this mean value, which is independent of the ambient relative humidity, and we have compared them with the scattering calculated using the hygroscopic growth factor from Brooks et al. (2004). The observed change in the calculated scattering is of ~15% on average and up to ~37%, slightly improving the agreement between the measured and calculated scattering. The hygroscopic growth factors for sulphate depend on both the ambient relative humidity and the initial size of the aerosol. However, information on the latter is very limited. Originally we used the hygroscopic growth factors for sulphate from Tang (1996), who reported values in the range 1.20–1.75 with dependence on the ambient relative humidity, but not on the initial aerosol size. Topping et al. (2005b) reported values in the range 1.66–1.73 depending on the initial aerosol size for a fixed ambient relative humidity of 90%, which involve a variation of approximately ±4% in the hygroscopic growth factor for sulphate. We have repeated our scattering calculations for all flights of VOCALS-REx, which were clearly dominated by sulphate, using this variation of ±4% on the hygroscopic growth factor from Tang (1996), and we have compared them with the original calculations. The sensitivity of the calculated scattering to the hygroscopic growth factor of sulphate is small, of only ~8.5% on average and up to ~9.4%, although we would expect the hygroscopic growth factor of sulphate to be much more sensitive to the ambient relative humidity than to the initial aerosol size.

To test the sensitivity of the calculated scattering and absorption to the aerosol size distribution, we have repeated our calculations for EUCAARI-LONGREX using flight-mean size distributions instead of the measured ones for each SLR of each flight of the campaign, and we have compared both sets of results. The change in the calculated scattering and absorption is significant, ~35% on average for both the scattering and the absorption and up to 134% and 122% for the scattering and the absorption, respectively. Thus the use of a campaign mean as for VOCALS is likely to be a significant hindrance to obtaining a meaningful model of aerosol optical properties.

5. Conclusions

Climate models often require aerosol optical properties to be prescribed. We have presented a flexible framework for calculating aerosol optical properties from commonly made measurements of aerosol composition and size distribution. For two different aerosol types, a complex multicomponent aerosol dominated by organic aerosol and ammonium nitrate and a simpler sulphate dominated aerosol, we have assessed the degree to which we can achieve closure with measured aerosol optical properties. We have also identified and quantified the largest sensitivities of the optical properties calculated in this way.

Our framework can replicate ambient scattering to within the measurement uncertainty for the complex EUCAARI-LONGREX aerosol, although the agreement is less good for the simpler VOCALS-REx aerosol. However, we do not have access to detailed size distributions for individual SLR from VOCALS-REx and our sensitivity tests show that size distribution is a large source of uncertainty in scattering. The second largest source of uncertainty in scattering is the growth factor assumed for organic aerosol. Our framework can also replicate dry absorption to within measurement uncertainty for EUCAARI-LONGREX, however no closure study was possible during VOCALS-REx because of the unreliable absorption measurements below the detection limit of the measurement instrument. For absorption, the refractive index of the organic aerosol is the predominant source of uncertainty (although it should be noted that we expect the refractive index of black carbon to be important for aerosol with a higher black carbon fraction). The hygroscopic scattering growth factors, *f*(RH), predicted by the framework seem at odds with the relative agreement in scattering. The calculated hygroscopic scattering growth factors seem much larger than the measured values. The reason for this is unclear, but measurement uncertainty in *f*(RH) is large.

Our results indicate that improvements in the accuracy of the aerosol radiative impact would come from better representation of aerosol size distributions and measurements of growth factors at a variety of sizes and relative humidities for organic aerosol. Measurements of hygroscopicity of real atmospheric aerosol alongside optical properties and refractive indices measurements would be a significant advance.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.02.063.

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