**Supplementary materials**

**For**

**Refractive index of engine-emitted black carbon and the influence of organic coatings on optical properties**

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**S1. Error analysis of the derived RI.**

The purpose of this error analysis was to give an indication of the level of confidence of the retrieved refractive indexes based on the measurement errors. Generally, there are two types of error in measurement or physical constraint that need to be considers; precision errors, which represent random datum-to-datum variation, and accuracy errors, which are systematic deviations brought on by biases or calibration errors. Precision errors can be evaluated through statistical analysis of the raw data, whereas accuracy had to be estimated based on knowledge of the instruments functions and prior evaluations of variations between calibrations. Because precision errors diminish with averaging and these experiments took place over sustained periods of time, it was found that when analysing all sources of error, the estimates of accuracy dominated over precision and thus are taken to be the main sources of error in the derived products.

Because the derivation of refractive indexes involves an optimisation step, it is not possible to describe the relationship between independent and dependent variables analytically, thus an explicit error propagation cannot be performed. The most rigorous method of analysing errors would be to perform a Monte Carlo based analysis of derived refractive indexes based on a modelled error function, however this is procedurally complex and computationally expensive. Given that the accuracy estimates in themselves contain an element of estimation based on expert judgement and are therefore not absolutely accuracy in themselves, the benefit of a comprehensive treatment was deemed questionable. Instead, the approach adopted here is to inspect individual dependencies of refractive index errors on measurement errors through numerically calculated derivatives and scaling accordingly. Where individual sources of error were found to dominate, the propagation of these errors were taken to be a sufficient estimate of the overall error.

The estimation of the partial errors of the derived RI in this study due to errors in the extinction and absorption measurements are given below:

(S1)

(S2)

Where and is the standard error of the extinction coefficient and absorption coefficient, and is the differential of real and imaginary part of RI with the extinction coefficient, and is the differential of real and imaginary part of RI with the absorption coefficient. These differentials are calculated by running the Mie model over a range of real and imaginary refractive indexes around the derived value and calculating absorption and extinction, then taking the reciprocals of the numerically-calculated partial derivatives , , etc.

Here, the 1.5% uncertainty associated with the accuracy in the extinction coefficient, and 5% uncertainty associated with the accuracy in the absorption coefficient is applied in the calculation (*Cotterell et al.*, 2020).

Another source of error was the the CPC-derived number concentrations. Again, due to the averaging process, the error was dominated by accuracy rather than precision. These instruments were serviced and calibrated by the manufacturer prior to the experiment and cross-calibrated in the laboratory, however an absolute accuracy of 1% would seem reasonable. This error would be manifested in the same manner as the overall absorption and extinction, so this can be applied in the same way as these measurements.

Another potentially significant source of error is the error in the sizing accuracy of the SMPS used as the primary sizing reference throughout the experiment. The size integrating method diminishes precision issues, and while this was calibrated using NIST traceable PSL spheres, there is a possibility of drift during the experiment. A 1% uncertainty was assumed to reflect this. Because the relationship between size and derived refractive index involves an optimisation step, deriving a partial derivative is not straightforward. However, a function of extinction or absorption dependent on diameter can be easily derived using the Mie model and in turn numerically differentiated to derive and . By multiplying these with the assumed diameter errors, the equivalent errors in extinction (5%) and absorption (4%) can be derived. After compared with the other errors, 5% was used as the basis for the overall error.

**S2. The optical models used to estimate the optical properties of coated BC particles.**

For the external mixing, the core and coating components contribute independently to the total scattering and absorption. As the BC core size () is determined by the SP2, The effective diameter of the coating components () can be calculated by equation S3 with the coating ratio (CR) value.

(S3)

For internally mixed black carbons particles, the effective refractive index () of the mixture is calculated using the equation S4.

(S4)

Where denotes the electric permittivity.

The Maxwell-Garnett mixing rule assumes that an inclusion with a permittivity and volume fraction () is embedded in a host matrix of permittivity , The resulting effective permittivity is obtained from the equation S5.

(S5)

The Bruggemann mixing rule assumes the two materials are embedded in a host medium with an can be calculated by equation S6.

0 (S6)

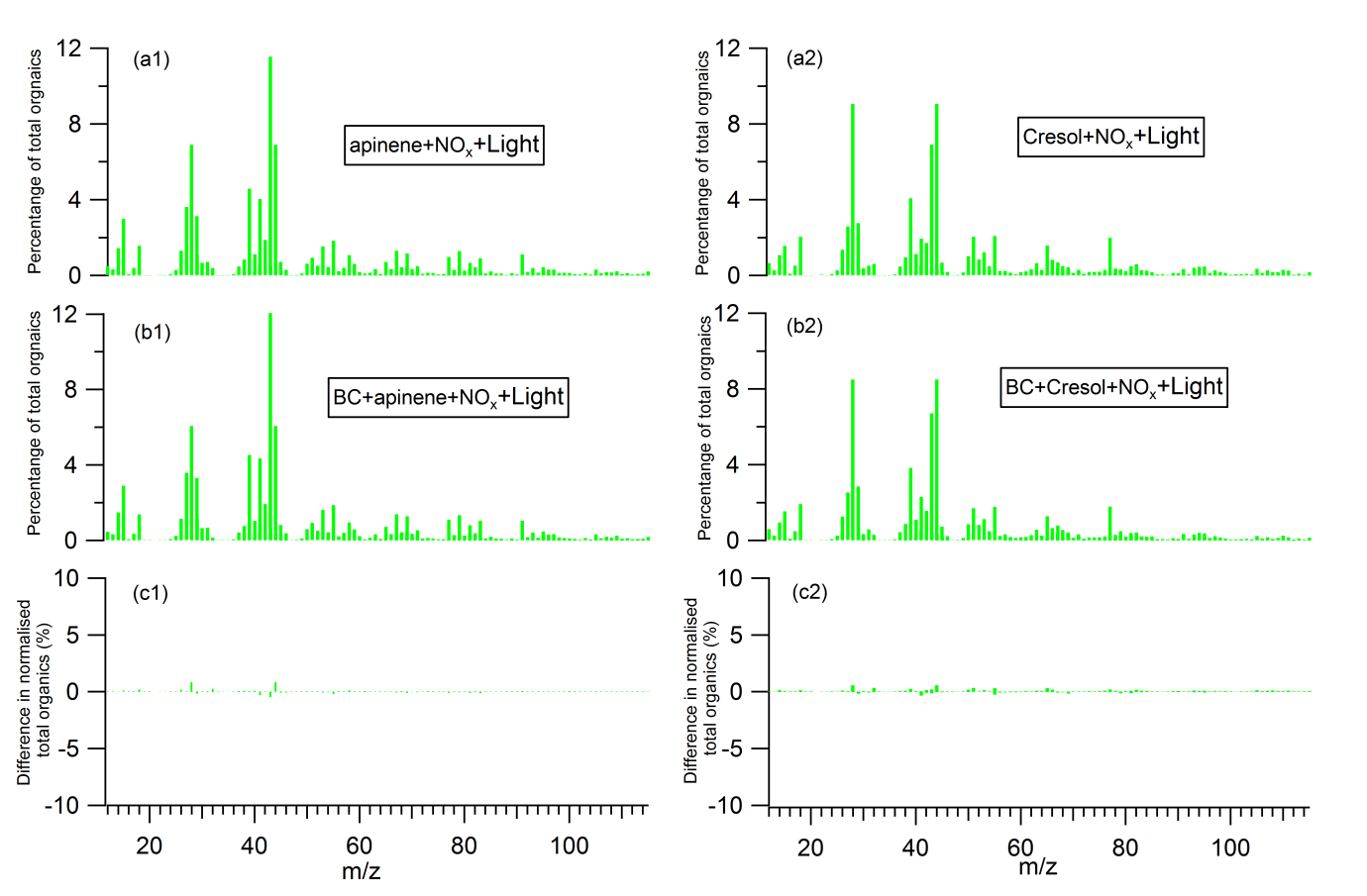
For the volume mixing rule, the of the coated particles is calculated assuming a volume weighted average based on the RI of each component.

(S7)

The measured and calculated values for the input of each model are summarized in Table S1 and S2.

**S3. Comparing the chemical compositions of SOA generated with and without Soot Particles.**

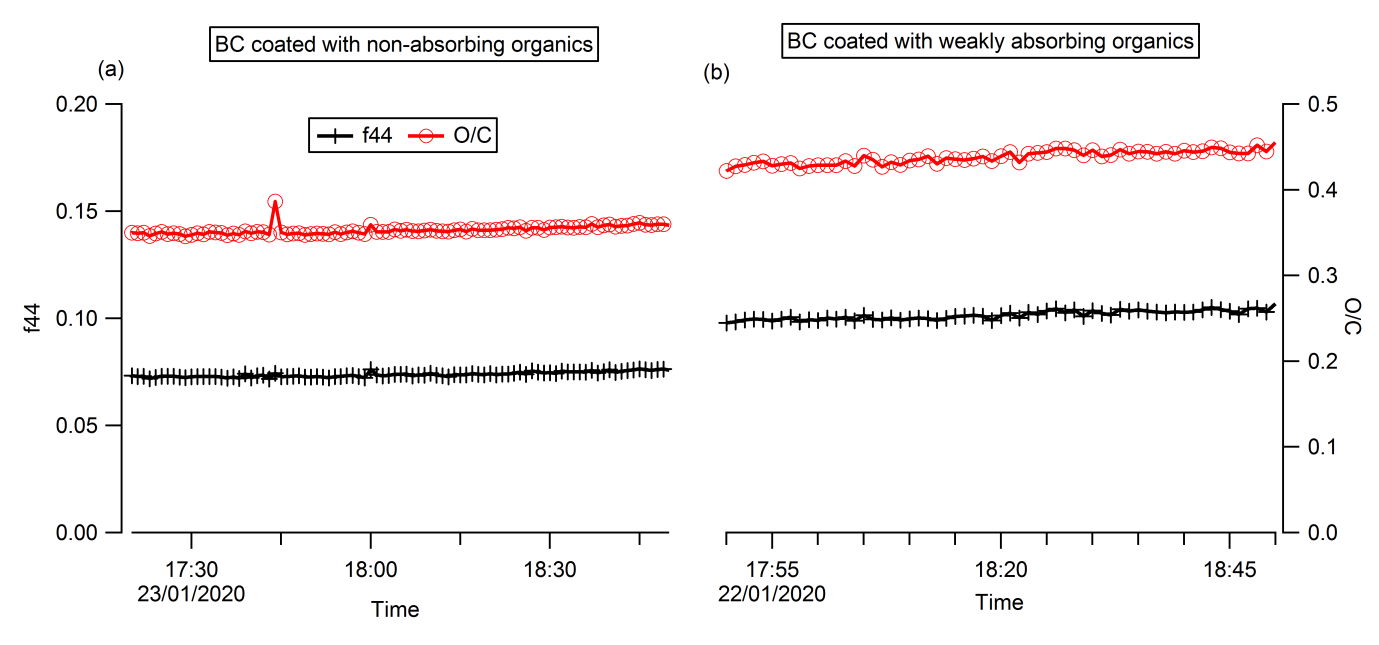
In this study, the AMS was utilized to measure the bulk aerosols throughout the experiment. Fig. S1 presents the normalized average mass spectra of pure SOAs during the period used to derive the RI values (represented as (a1) and (a2)), the normalized average mass spectra of the coating organics of the coated BC particles during the investigated period ((b1) and (b2)), as well as the differences between them ((c1) and (c2)). Notably, there is minimal variation observed in the chemical composition of the SOAs generated in the presence and absence of BC particles.



**Fig. S1** Normalized average mass spectra of pure SOAs during the period used to derive the RI values (represented as (a1) and (a2)), normalized average mass spectra of the coating organics of the coated BC particles during the investigated period ((b1) and (b2)), as well as the differences between them ((c1) and (c2)).

**S4. Stability of chemical compositions of organics during the investigated coating ratio period.**

The time series analysis of O/C and f44 values of bulk aerosols during the investigated coating ratio period (T1-T6/T7) is shown in Figure S2. The figure clearly illustrates that there is negligible variation in the O/C and f44 values throughout this period. These results provide robust evidence supporting the use of a single refractive index (RI) derived from pure SOAs to effectively represent the coating organics of the coated BC particles.



**Fig. S2** Time series of O/C and f44 values of bulk aerosols during the investigated coating ratio period: (a) BC coated with non-absorbing organics, and (b) BC coated with weakly absorbing organics.

**Table S1.** Input values for simulating the optical properties of BC particles coated with non-absorbing organics.

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  |  | | **CRSMPS** | | | | | | | | **CRSP2** | | | | | | |
|  |  | | **T1** | | **T2** | **T3** | **T4** | **T5** | **T6** | **T7** | **T1** | **T2** | **T3** | **T4** | **T5** | **T6** | **T7** |
| **Core-Shell** | | Dc | | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 |
| CR | | 2.56 | 2.85 | 2.80 | 2.84 | 2.87 | 2.80 | 2.91 | 2.37 | 2.46 | 2.55 | 2.64 | 2.67 | 2.71 | 2.74 |
| RIBC | | 1.816 + 0.624i | | | | | | | 1.816 + 0.624i | | | | | | |
| RICoating | | 1.584 | | | | | | | 1.584 | | | | | | |
| **External Mixing** | | Dc | | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 |
| DpCoating | | 213.08 | 236.73 | 246.36 | 249.53 | 251.82 | 245.55 | 254.74 | 196.65 | 202.54 | 223.42 | 230.77 | 233.57 | 236.50 | 239.07 |
| RIBC | | 1.816 + 0.624i | | | | | | | 1.816 + 0.624i | | | | | | |
| RICoating | | 1.584 | | | | | | | 1.584 | | | | | | |
| **Maxwell-Garnett** | | Dc | | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 |
| CR | | 2.56 | 2.85 | 2.80 | 2.84 | 2.87 | 2.80 | 2.91 | 2.37 | 2.46 | 2.55 | 2.64 | 2.67 | 2.71 | 2.74 |
| VFBC (%) | | 5.97 | 4.34 | 4.55 | 4.37 | 4.24 | 4.53 | 4.07 | 5.97 | 4.34 | 4.55 | 4.37 | 4.24 | 4.53 | 4.07 |
| VFCoating (%) | | 94.03 | 95.66 | 95.45 | 95.63 | 95.76 | 95.47 | 95.93 | 94.03 | 95.66 | 95.45 | 95.63 | 95.76 | 95.47 | 95.93 |
| RIMixing | | 1.6012+0.0357i | 1.5965+  0.0260i | 1.5971+  0.0272i | 1.5966+  0.0261i | 1.5963+  0.0254i | 1.5971+  0.0271i | 1.5958+  0.0243i | 1.6055+  0.0447i | 1.6035+  0.0405i | 1.6013+  0.0360i | 1.5998+  0.0327i | 1.5992+  0.0315i | 1.5986+  0.0302i | 1.5981+  0.0292i |
| **Bruggemann** | | Dc | | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 |
| CR | | 2.56 | 2.85 | 2.80 | 2.84 | 2.87 | 2.80 | 2.91 | 2.37 | 2.46 | 2.55 | 2.64 | 2.67 | 2.71 | 2.74 |
| VFBC (%) | | 5.97 | 4.34 | 4.55 | 4.37 | 4.24 | 4.53 | 4.07 | 5.97 | 4.34 | 4.55 | 4.37 | 4.24 | 4.53 | 4.07 |
| VFCoating (%) | | 94.03 | 95.66 | 95.45 | 95.63 | 95.76 | 95.47 | 95.93 | 94.03 | 95.66 | 95.45 | 95.63 | 95.76 | 95.47 | 95.93 |
| RIMixing | | 1.6009+0.0357i | 1.5964+  0.0259i | 1.5970+  0.0272i | 1.5964+  0.0261i | 1.5961+  0.0253i | 1.5969+  0.0271i | 1.5956+  0.0243i | 1.6050+  0.0446i | 1.6031+  0.0404i | 1.6010+  0.0359i | 1.5995+  0.0326i | 1.5989+  0.0314i | 1.5983+  0.0301i | 1.5979+  0.0292i |
| **Volume mixing** | | Dc | | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 | 85.00 | 84.44 | 89.36 | 89.18 | 89.08 | 88.91 | 88.86 |
| CR | | 2.56 | 2.85 | 2.80 | 2.84 | 2.87 | 2.80 | 2.91 | 2.37 | 2.46 | 2.55 | 2.64 | 2.67 | 2.71 | 2.74 |
| VFBC (%) | | 5.97 | 4.34 | 4.55 | 4.37 | 4.24 | 4.53 | 4.07 | 5.97 | 4.34 | 4.55 | 4.37 | 4.24 | 4.53 | 4.07 |
| VFCoating (%) | | 94.03 | 95.66 | 95.45 | 95.63 | 95.76 | 95.47 | 95.93 | 94.03 | 95.66 | 95.45 | 95.63 | 95.76 | 95.47 | 95.93 |
| RIMixing | | 1.5979+0.0373i | 1.5941+  0.0271i | 1.5946+  0.0284i | 1.5941+  0.0272i | 1.5938+  0.0265i | 1.5945+  0.0283i | 1.5935+  0.0254i | 1.6013+  0.0466i | 1.5997+  0.0422i | 1.5980+  0.0375i | 1.5967+  0.0341i | 1.5962+  0.0328i | 1.5957+  0.0315i | 1.5953+  0.0305i |

**Table S2.** Input values for simulating the optical properties of BC particles coated with weakly-absorbing organics.

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  |  | | **CRSMPS** | | | | | | | **CRSP2** | | | | | |
|  |  | | **T1** | | **T2** | **T3** | **T4** | **T5** | **T6** | **T1** | **T2** | **T3** | **T4** | **T5** | **T6** |
| **Core-Shell** | | Dc | | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 |
| CR | | 2.49 | 2.52 | 2.63 | 2.65 | 2.65 | 2.61 | 2.42 | 2.50 | 2.54 | 2.59 | 2.64 | 2.64 |
| RIBC | | 1.923 + 0.656i | | | | | | 1.923 + 0.656i | | | | | |
| RICoating | | 1.738 + 0.0316i | | | | | | 1.738 + 0.0316i | | | | | |
| **External Mixing** | | Dc | | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 |
| DpCoating | | 206.68 | 207.59 | 217.76 | 218.55 | 218.58 | 215.53 | 199.99 | 205.42 | 209.72 | 212.91 | 217.35 | 218.27 |
| RIBC | | 1.923 + 0.656i | | | | | | 1.923 + 0.656i | | | | | |
| RICoating | | 1.738 + 0.0316i | | | | | | 1.738 + 0.0316i | | | | | |
| **Maxwell-Garnett** | | Dc | | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 |
| CR | | 2.49 | 2.52 | 2.63 | 2.65 | 2.65 | 2.61 | 2.42 | 2.50 | 2.54 | 2.59 | 2.64 | 2.64 |
| VFBC (%) | | 6.45 | 6.25 | 5.51 | 5.36 | 5.38 | 5.61 | 7.07 | 6.44 | 6.12 | 5.77 | 5.47 | 5.42 |
| VFCoating (%) | | 93.55 | 93.75 | 94.49 | 94.64 | 94.62 | 94.39 | 92.93 | 93.56 | 93.88 | 94.23 | 94.53 | 94.58 |
| RIMixing | | 1.7531+  0.0709i | 1.7527+  0.0697i | 1.7509+  0.0652i | 1.7506+  0.0642i | 1.7506+  0.0644i | 1.7512+  0.0658i | 1.7546+  0.0747i | 1.7531+  0.0708i | 1.7523+  0.0689i | 1.7515+  0.0668i | 1.7508+  0.0649i | 1.7507+  0.0646i |
| **Bruggemann** | | Dc | | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 |
| CR | | 2.49 | 2.52 | 2.63 | 2.65 | 2.65 | 2.61 | 2.42 | 2.50 | 2.54 | 2.59 | 2.64 | 2.64 |
| VFBC (%) | | 6.45 | 6.25 | 5.51 | 5.36 | 5.38 | 5.61 | 7.07 | 6.44 | 6.12 | 5.77 | 5.47 | 5.42 |
| VFCoating (%) | | 93.55 | 93.75 | 94.49 | 94.64 | 94.62 | 94.39 | 92.93 | 93.56 | 93.88 | 94.23 | 94.53 | 94.58 |
| RIMixing | | 1.7528+  0.0708i | 1.7524+  0.0696i | 1.7507+  0.0651i | 1.7504+  0.0642i | 1.7504+  0.0643i | 1.7509+  0.0657i | 1.7542+  0.0745i | 1.7528+  0.0707i | 1.7521+  0.0688i | 1.7513+  0.0666i | 1.7506+  0.0648i | 1.7505+  0.0645i |
| **Volume mixing** | | Dc | | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 | 84.74 | 84.17 | 84.42 | 83.91 | 84.04 | 84.13 |
| CR | | 2.49 | 2.52 | 2.63 | 2.65 | 2.65 | 2.61 | 2.42 | 2.50 | 2.54 | 2.59 | 2.64 | 2.64 |
| VFBC (%) | | 6.45 | 6.25 | 5.51 | 5.36 | 5.38 | 5.61 | 7.07 | 6.44 | 6.12 | 5.77 | 5.47 | 5.42 |
| VFCoating (%) | | 93.55 | 93.75 | 94.49 | 94.64 | 94.62 | 94.39 | 92.93 | 93.56 | 93.88 | 94.23 | 94.53 | 94.58 |
| RIMixing | | 1.7499+  0.0719i | 1.7496+  0.0706i | 1.7482+  0.0660i | 1.7479+  0.0651i | 1.7480+  0.0652i | 1.7484+  0.0667i | 1.7511+  0.0757i | 1.7499+  0.0718i | 1.7493+  0.0698i | 1.7487+  0.0676i | 1.7481+  0.0657i | 1.7480+  0.0654i |

**References**

Cotterell, M. I., Szpek, K., Haywood, J. M., & Langridge, J. M. (2020), Sensitivity and accuracy of refractive index retrievals from measured extinction and absorption cross sections for mobility-selected internally mixed light absorbing aerosols, *Aerosol Sci. Technol.*, *54*(9), 1034-1057, doi:10.1080/02786826.2020.1757034.