We report the measurements of the differential Raman backscattering cross sections for several carbonaceous ultrafine particles of environmental relevances. These were obtained by dispersing the target particles in liquid water which was used as the internal standard reference. The optical collection was performed in a configuration to ensure a detection as close as possible to the backward direction. These are the first cross sections on black carbon-type particles although Raman spectroscopy is widely used in Carbon science. The high values of the cross sections, few $10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$, reflect resonance effects that take advantages of the disordered polyaromatic structures. Because they were measured in conditions intended to mimic the aerosol phase, these measurements provide a crucial step to move toward quantitative Raman spectroscopy and enable development of dedicated teledetection of black carbon in the atmosphere and in combustion chambers.

Natural carbonaceous matter, found on Earth as well as in space, plays an effective bridge-building role between astrophysics and geophysics. Crystalline to amorphous forms of carbon find many technological applications, with a high potential for the new carbon nanomaterials. In addition, combustion of carbon-based fuels, including fossil fuels, biomass and biofuels, is still the main source of energy utilized by anthropic activities and dominates carbon-based materials production. Every year, about $10^7$ tons of black carbon (BC, atmospheric light absorbing particles that embrace soot) are emitted worldwide. As a consequence, BC occurred in 62% of the total mass of the particles in the polluted air in Mexico. Southeast Asia is one of the main hot spot of BC emission as it combines massive biomass burning events and a growing use of fossil fuels. BC are one of the most efficient climate radiative forcer. Secondary effects are also important, such as decreasing the albedo of snow and ice. The net impact of BC on climate depends on many factors including the compounds, the presence of co-emitted pollutants, atmospheric lifetime, altitude, aging and mixing processes in the atmosphere, in which the aging of particles plays a key role in the estimation of both the direct and indirect climate effects. In addition to climate effects, there are numerous epidemiological studies establishing associations between exposure to particulate pollution and increased morbidity and mortality for respiratory and cardiovascular diseases. It is thus of the uttermost importance to progress on the knowledge of their optical properties in order to better evaluate their climate impact. In addition, to help mitigating their emission, it is mandatory to continue the development of new tools to monitor the BC in the atmosphere or soot in combustion chamber.

Raman spectroscopy is a powerful tool for structural investigation of the carbon-based materials because it is sensitive not only to the crystalline structures but also to the molecular structures in amorphous matter and is already widely applied in environmental research or combustion. The main features in Raman spectra of polyaromatic carbonaceous materials are the so-called D and G peaks, highlighted by the pioneering work by Tuinstra and Koenig in 1970. The G peak corresponds to the bond stretching of all pairs of sp² sites in hexagonal aromatic rings. The D peak is due to the breathing modes of sp² sites in rings and its activation requires defects. However, despite the large amount of experimental data available in the literature concerning the Raman spectra of carbonaceous materials, most of the studies focused on the relative intensities and shapes of the Raman peaks but not on the absolute intensities. In fact, spontaneous Raman spectroscopy is rarely used as a quantitative spectroscopy in Carbon science because making the Raman spectroscopy quantitative for absorbing species remains a challenge.

The present work provides experimental measures of the differential Raman backscattering cross sections (DRCS) of several natural and artificial carbon-based fine particles, together with the development of a practical
method to quantify DRCS of other nanoparticles. The samples were chosen to provide a representative set of black carbon particles of atmospheric relevance.

Results

Raman spectra of black carbon particles in liquid water. The Raman spectra of the BC particles in liquid water are shown in Fig. 1. The superimposition of the typical G and D Raman bands at about 1600 and 1350 cm$^{-1}$, respectively,\textsuperscript{18,19,27}, with those of liquid water at about 1640 and 3500 cm$^{-1}$\textsuperscript{28,29} can be seen. The diesel soot spectrum exhibits a strong background but the Raman bands remain observable. The background is due to fluorescence, probably originating from the polycyclic aromatic hydrocarbons (PAHs) that are adsorbed on the soot particles. Additional bands can be seen which correspond to the Raman spectra of O$_2$ (at 1555 cm$^{-1}$) and N$_2$ (at 2331 cm$^{-1}$) from the atmospheric column along the line of sight. It should be noted that the absolute intensities of the N$_2$ and O$_2$ Raman bands are found almost constant in Fig. 1, only the BC and water Raman bands relative intensities are found varying.

In Fig. 2, the coal particles Raman spectrum in the form of powder is superimposed to that of the coal particles dispersed in liquid water. The overlap shows that the two spectra are very similar and confirms that the solvent
has no measurable effects on the Raman bands of the coal particles. The slight differences are due to the subtraction of the background and of the liquid water Raman band lying at 1640 cm$^{-1}$.

**Measurements of the Raman cross sections.** After systematic subtraction of the underlying background and of the contribution of the bending mode of water, all obtained Raman spectra of coal and water stretching bands of the mixtures at different coal concentrations are plotted in Fig. 3. Note that the O-H stretching band is much more intense than the D and G bands. It can be seen that the higher the concentration of the coal particles is, the higher the Raman signal of coal is. An opposite trend in the O-H stretching band is concomitantly observed because the probe volume decreases with the increase of the optical density (following the increase of coal concentration) of the solution. The D band intensity was taken by summing the photoelectron counts from 1000 to 1500 cm$^{-1}$ and for the G band intensity from 1500 to 1700 cm$^{-1}$. For the stretching mode of liquid water, no other feature contaminates its spectral range and integration was performed all over the band. The ratios between the observed Raman signals of the D and G bands and the O-H stretching band of the liquid water are shown in Fig. 4 as a function of coal particles concentrations. The experimental ratios are following linearly the coal concentrations (Fig. 4a) as expected for a pure dependence on the mass ratios even if the probe volume is changing due to the dependence of optical extinction of the solution with concentration. It confirms that the DRCS of the D and G bands can be evaluated using the liquid water band as an internal standard. The DRCS for the stretching mode of water is taken to be $\beta^w = 5.74 \times 10^{-36}$ cm$^2$.sr$^{-1}$.molecule$^{-1}$. From equation 5, the DRCS of D and G peaks are found to be $4.1 \pm 1.1 \times 10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$ and $2.7 \pm 0.6 \times 10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$,
respectively, as deduced from the average value of the DRCS deduced at each concentration (Fig. 4b). Note that the carbon content is 92% of the mass of coal and this value was taken into account.

The Raman spectra of CB Ensaco 350G dispersed in liquid water were more complicated to analyze than that of coal particles. While the D peak appeared clearly (Fig. 5) with no spectral contamination, the G peak was found to overlap with the atmospheric O$_2$ band and the bending mode of water. In addition subtraction of the bending mode contamination could not be performed using the pure water bending/stretching ratio: the measured liquid water bending mode signal was evidently weaker than expected. It traces an interaction of the solvent with the nanoparticles, their nanometer size shape ($\approx 30$ nm diameter$^{31}$) offering a much higher solvation surface than the coal microparticles (average diameter about 1 $\mu$m). A comparison of the shapes of the water stretching band at different concentrations shows that no spectral changes are observed. It underlines a weak effect for this mode. Therefore the data treatment assumed that the water stretching mode cross section was unchanged.

To fit the experimental spectra, we summed all components using the CB powder Raman spectrum and the O$_2$/N$_2$ Raman signals to determine the strength of the O$_2$ Raman band independently. The shape of the water bending mode was taken from the pure liquid water Raman data. The result of the fit is shown in Fig. 5. We carried out this analysis for several CB concentrations. The ratios of the observed Raman signals of the D, G and O-H stretching bands are plotted in Fig. 6a and exhibit the expected linear trend. The deduced DRCS for all concentrations are shown in Fig. 6b using equation 5. It should be noted that due to the higher optical density of these solutions, the range of concentration was reduced to 0.8–2 mg in 40 ml water, i.e. a smaller range as compared to

Figure 4. Panel (a) Intensity ratios of the Raman bands as a function of the coal particles concentrations. The slopes are found to be 0.0027 and 0.0016 for the D and G bands, respectively. Panel (b) DCRS for the G (red lines and points) and D (black line and points) bands of coal particles deduced at all concentrations, together with the average value.

Figure 5. Deconvolution of the Raman spectrum of Ensaco 350G carbon black nanoparticles dispersed in liquid water (about 2 mg in 40 ml of water). The bright blue line is the experimentally observed Raman spectrum; The blue solid line is the Raman spectrum of the CB in form of powder; The green solid line is the scaled Raman signal of O$_2$; The red solid line is the fitted bending Raman band of water; The red dot line is the fitted spectrum from the three components contributing to the experimental data.
that of the coal particles (Fig. 4). We obtained the average DRCS for the D band $5.1 \pm 1.2 \times 10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$ and for the G band $2.9 \pm 0.8 \times 10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$.

For the diesel soot, the measurements were done at two different concentrations: 9 mg and 5.4 mg of diesel soot in 40 ml water. The spectral contamination in the range of the G band is found similar to that the CB Ensaco 350G and the same fit was applied to extract the G band intensity (Fig. 7). In that case, the signal to noise ratio was even worse than that obtained for the CB Ensaco 350G, precluding measurements at lower concentrations. The average DRCS of the G and D bands are $8.8 \pm 3 \times 10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$ and $11 \pm 4 \times 10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$, respectively.

**Discussion**

The DCRS values are reported in the Table 1. There is no large difference between the DRCS of the three samples, which are on the order of few $10^{-28}$ cm$^2$.sr$^{-1}$.atom$^{-1}$. It should be noted that these are effective cross sections. Actually the measurements were scaled with respect to their mass (number of carbon atoms) although the granularity may have an influence on the scattering properties. For the coal particles, sizes were about 1 μm as revealed by optical microscopy on dried samples. The two other samples were composed of nanoparticles of about few tens of nm diameter, generally forming aggregates of up to few hundreds of nm size. It should be noted that the exact size distribution while being dispersed in liquid water under ultrasonication in the cleaning unit was not controlled. Thus, although the granularities were different for all samples, the linear dependence with concentration for the coal particles and the CB Ensaco 350G provide strong support for our values per carbon atoms.
The DRCS for Raman modes of various types of carbonaceous materials have been measured for only few carbonaceous materials that are graphite32,33, diamond34, C6035 and carbon nanotubes36. In the latter case, the values were not reported in Table 1 because it could not be given per carbon atom. Our study shows that the DRCS of our samples are approximately 100 times larger than that of the breathing mode of liquid benzene. It is also higher than that of diamond. The DRCS of coal, CB and diesel soot are found comparable to the graphite values but higher than those of the nano-graphites. For those bulky materials, the variations cannot involve the particle shapes scattering properties. The reason for the large cross sections is probably the appearance of resonance Raman effect in our samples leading to the enhancement of Raman signals37 and was shown to follow a polyaromatic unit size dependence33. The FWHMs of the D and G bands of our samples are much larger than those of the nano-graphites in Cançado et al.33. The FWHMs of their Raman bands range from 10 to 50 cm−1 while in our BC particles, they are about several tens to 200 cm−1. It shows that the polyaromatic unit size L a of our samples are much smaller than those of the nano-graphites, ranging from 20 to 65 nm33. Effectively diesel soot have L a of about 2 nm37, carbon black around 1–2 nm31 and the anthracite coal exhibits an average L a of about 2 nm as well (measured by high resolution transmission electron microscopy, not shown). It suggests that the resonance effects should be similar for the BC samples, as observed. In order to progress further and explain the high values, the DCRS would have to be measured at several wavelengths in order to probe the resonant effects from the near-infrared up to the UV.

An important outcome of this investigation is that it reveals the DRCS of BC are large. These are found close to that of graphite and nearly 3 orders of magnitude higher that of gaseous molecules38 (toluene, benzene . . .) or silicates39 commonly at the focus of atmospheric research. We believe our determination of the DCRS will contribute to the development of quantitative teledetection of BC emission in the atmosphere, for instances by extending Raman-Lidar capabilities recently developed for silicate aerosols39–41 or in combustion chambers by complementing the laser induced incandescence technique42,43 recently applied in atmospheric measurements44. The main drawback relies on the observed variations of the DCRS among the present set of reference BC up to a factor of 3. It is in line with the specificity of the BC family of aerosol whose optical properties are spanning a large range of values45. Additional work has to be performed in order to determine the best DCRS values to be used when monitoring complex mixtures of BC. It is noteworthy that the measure of the Raman G and D bands characteristics provide very valuable information for speciation as well. Overall, µg/m³ of BC concentration appear as a plausible target for Raman teledetection.

### Methods

**Calibration method for the Determination of the cross section.** The DCRS β (in cm².sr⁻¹.atom⁻¹) for a vibrational mode is defined as the number of inelastically scattered photons per unit time and per unit solid angle divided by the incident number of photons per unit time per unit area. It relates to the observed signal S (in photoelectron)38:

$$ S = (P_T \cdot \beta \cdot D \cdot K) \cdot (A_D \cdot O_D \cdot T \cdot Q) \cdot t $$  

(1)
where \( P \) (in photons s\(^{-1}\).cm\(^{-2}\)) is the photon irradiance, \( D \) (in molecules.cm\(^{-3}\)) the density of scatterers, \( K \) the constant of geometry (unit less), \( A \) (in cm\(^2\)) the sample area monitored by the spectrometer, \( O \) (in sr) the collection solid angle of the spectrometer, at the sample, \( T \) the transmission of the spectrometer and collection optics (unit less), \( Q \) the quantum efficiency of the detector (in e\(^-\)/photon), and \( t \) is the observation time (in seconds).

The first composition is the characteristic intensity defined as the total Raman scattering divided by the area of scattering volume. The second is a general instrumental component containing the collection optics and spectrophotograph. The DRCS in the equation 1 is difficult to use directly because it depends on a large number of variables. Thus we simplified the measurements by using internal calibration. It consists in normalizing the observed signal of the sample to that of the co-mixed standard reference (indicated by * in the following equations). If the sample and the standard substance, whose Raman cross section is known, are measured simultaneously, they have the same photon irradiance, the sample area monitored by the spectrometer, the same collection solid angle of the spectrometer and the same acquisition time. In this case, the ratio \( S/S^* \) is given by:

\[
\frac{S}{S^*} = \frac{(\beta D T) Q D T Q}{(\beta^* D^*) T (Q^*)}
\]

The observed signals in photoelectrons are proportional to the number of digital units “counts” \( I \) of CCD camera. So, the equation 2 is re-written:

\[
\frac{I}{I^*} = \frac{(\beta D T) Q D T Q}{(\beta^* D^*) T (Q^*)}
\]

The composition of the transmission \( T \) and the efficiency \( Q \) depends on the type of detector, grating, and optics used. Those parameters were measured with a calibration source. We used a blackbody calibration source BB4-A (Omega Engineering Inc.), whose source temperature is adjustable over a range of 100 to 982 Celsius degrees. The black body was installed at the sample position. The ratio between the Raman peaks of the samples and of the standard reference after being corrected with the spectral response then simplifies:

\[
\frac{I_{\text{corrected}}}{I_{\text{corrected}*}} = \frac{D}{D^*}
\]

From the equation 4, the DRCS of the sample, \( \beta \), is determined by:

\[
\beta = \frac{I_{\text{corrected}} D^*}{I_{\text{corrected}*} D} \beta^*
\]

This is the final equation used to deduce the DRCS of the samples. In our experiment, the dimension of \( D^* \) is water molecule.cm\(^{-3}\) while that of \( D \) is carbon atom.cm\(^{-3}\).

A crucial advantage of our method is that it permits the measurement of differential cross sections of particles using internal calibration. It allows to avoid the difficulty inherent to the case of absorbing material while the references are usually transparent.

**Samples preparation.** The samples were anthracite coal ultrafine particles from a coal sample originating from a mine in Vietnam and milled in an Agate mortar, an industrial carbon black (CB) Ensaco 350G (Timcal Graphite & Carbon) and the soot produced from a diesel engine (SRM2975, NIST) called diesel soot in this paper. The diesel particulate material used to prepare SRM 2975 was obtained from M.E. Wright of the Donaldson Company, Inc., Minneapolis, MN. The material was collected from a filtering system designed specifically for diesel-powered forklifts.

For the measurements, each sample was mixed in pure water at high concentrations. The beakers were installed in the water tank of an ultrasonic cleaning unit (Elasonic) operated at 37 kHz frequency and at 40W. After a half an hour, the majority of their masses were dissolved or suspended in the solvent, the rest was found floated on the surface and was removed and weighted for correction. Then dilution was applied to prepare solutions with targeted concentrations. Note that their concentrations were ensured transparent enough for transmission of the laser. During the acquisition, the samples were kept in the ultrasonic cleaning unit to prevent from sedimentation on the bottom of the beaker.

It should be noted that we tried using other solvents as internal standard: cyclohexane and ethanol. However, as most organic solvents, their Raman spectra exhibits several strong bands overlapping with the D and G bands.
of the BC samples and hiding them. Although the bending mode of water at 1640 cm⁻¹ is close to G peak, its DRCS is weak enough to enable subtraction. Thus water appeared as the best solvent when using ultrasonication to prepare the mixtures and during the measurements.

**Experimental set up.** In our experimental setup (Fig. 8), the excitation source was a continuous laser firing at 532 nm (Verdi, Coherent). The laser was illuminating the sample from the top using a broadband dielectric laser mirror to avoid fluorescence from the glass of the beaker. The scattered light was collected by a lens system positioned about 1.5 m from the target, filtered by a 532 nm Razor Edge filter before entering a round-to-linear fiber bundle. This filter blocks the radiations from 430 to 539 nm with an optical density (OD) greater than 6 and transmits up to 90% from 539 to 1200 nm. The fiber bundle contains 19 fibers arranged in a line configuration at one end and in a circular configuration at the other end. Each fiber has a core diameter of 200 μm. The linear end coupled with the slit of a spectrometer equipped with a CCD camera (model PIXIS 400, Princeton Inst.). The spectrometer was a 0.3 m focal length (Acton SP2300 spectrometer, Princeton Inst.) equipped with a 1200 lines grating blazed at 500 nm. A notch filter at 532 nm was inserted in front of the slit of the spectrometer to further reject the elastic scattering. This StopLine notch filter (Semrock) has OD greater than 6 blocking of wavelength at 532 nm and about 90% transmission from 551 to 1600 nm. The optical fibers are not polarization maintaining therefore all measurements were unpolarized. Typical laser power were about 0.2W and acquisition time for one spectrum was set to about 20 minutes. Each measurement was performed several times to ensure reproducibility and check that the concentrations were constant during the measurements.

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Author Contributions

T.P. and K.C.L. conceived the experiment. K.C.L. conducted the experiment with support from C.L. and T.P., K.C.L. and T.P. analyzed the results. All authors reviewed the manuscript.

Additional Information

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