New studies on scattering properties of four kinds of soot

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New polarization curves as a function of the scattering angle are studied with PROGRA2 instruments (Propriétés Optiques des Grains Astronomiques et Atmosphériques) for 4 different kinds of soot, levitating in a cloud and deposited on a surface. The smallest agglomerates of few µm produce higher polarization values than the full set of agglomerates. The small agglomerates and the packed particles on a surface have the higher polarization. These results need more studies. These curves will be used for detection of soot in the stratosphere by remote sensing measurements.

INTRODUCTION

It is now known that soot is present in stratosphere, and it can have a direct effect on the atmospheric chemistry and, through radiative transfer, on climate [1]. Reference scattering curves for soot are necessary to identify their types from remote sensing measurements in particular using balloon-born radiometer MicroRADIBAL [2]. Models using the Mie theory failed to give the optical properties of soot because of their fractal irregular shape [3]. An experimental database of the optical properties of soot made in a laboratory is then necessary. Preliminary studies on light scattering with some kinds of soot have already been done [1]. Here we present new measurements for the polarization produced by four types of soot obtained by the PROGRA2 experiment. New cameras used by PROGRA2 are more sensitive, so the optical properties of the smaller particles are easier to detect.

The studied samples have an unknown imaginary part of the index, complicating the explanation of the polarization degrees. Previous studies on polarization by deposited particles have been made by Shkuratov et al. 2006 [4], where the imaginary part of index was known.

MEASUREMENT SYSTEM WITH PROGRA2-VIS AND DATA ANALYSIS

PROGRA2 instruments are imaging polarimeters with two randomly polarized lasers at 632.8 nm and 543.5 nm. The laser beam carried by optical fibres illuminates the particles. Light scattered by samples at a given scattering angles is split between the parallel and perpendicular component by a beam splitter cube. The cube is followed by the detectors: two CCD cameras providing 25 images per second. PROGRA2-vis observes the levitating parti-

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cles lifted by a small injection of nitrogen in a vial. The incident laser beam and the vial rotate and the detection system is fixed. The size of the agglomerates having diameters greater than 20 µm can be determined by images recorded by the instrument. PROGRA2-surf performed measurement for deposited particles on a surface. More details of the PROGRA2 are presented in Renard et al. [5] and in Hadamcik et al. [6].

The degree of linear polarization P of the scattered light can be given

$$P = \frac{I_{per} - I_{par}}{I_{per} + I_{par}} , \qquad (1)$$

where I_{per} and I_{par} are the scattered intensities polarized perpendicular and parallel to the scattering plane, respectively.

The imaging system allows sorting out the polarization values per different grain size classes to estimate the effect of size on the polarization for a given sample.

SCATTERING CURVES FOR SOOT

Studied samples

Two of the studied samples are produced from an incomplete combustion of a liquid solvent: Toluene (C_7H_8). The two others are produced from a solid polymer; Polymethyl Methacrylate or PMMA ($C_5H_8O_2$). Table 1 shows information about the studied samples. The global equivalent ratio is defined as the ratio between the mass ratio of fuel to air during the experiment, and the mass ratio of fuel to air at stoichiometry [6].

Name	Ventilation flow rate	Global equiva-	Primary particle	Fractal dimen-	Soot density
	(m^3/b)	lence ratio	diameter (nm)	sion	(g/cm^3)
Toluene1	450	0.01	52	1.86	1.46
Toluene2	100	0.06	70	1.81	1.46
PMMA1	450	0.06	42	1.78	1.52
PMMA2	50	0.49	56	1.74	1.52

 Table 1. Detailed info about the four samples.

Levitating and deposited samples

Figure 1 presents the dependence of linear polarization on the scattering angle at $\lambda = 632.8$ nm for levitating agglomerates of soot. The amplitude of positive polarization branches are obtained with toluene1, followed by a similar behavior of the polarization produced by toluene2 and PMMA1. The lowest amplitude of the polarization is obtained with PMMA2. All the amplitudes of polarization are obtained for scattering angles between 80° and 90°.

The angular profile of linear polarization produced by the smallest agglomerates of a few µm in diameter can be retrieved by using images recorded after 30 seconds of the nitrogen injection. Fig. 2 shows the polarization curves of such particles. An obviously larger difference is noticed between the polarization amplitude obtained with the smallest agglomerates and those obtained with the full set of agglomerates. As we can see in the Fig. 2, the two samples issued with low flow ventilation rate PMMA1 and toluene1 produce the highest polarization curves and show similar behavior to each other. The two other samples, PMMA2 and toluene2, having lower flow ventilation rate than the others, produce lower polarization curves for the small agglomerates. When the ventilation flow rate decreases, the global equivalent ratio increases, and the fractal dimension decreases [7]. This could be an explanation for the impact of the ventilation flow rate on the polarization values.

It is interesting to check the dependence of polarization on the diameter of the agglomerate after averaging the data over size intervals of 50 μ m, following a power law fit. The imaging system allows identifying polarization produced by particles larger than 20 μ m in diameter. Fig. 3 shows an example given at 70° scattering angle, obtained with the sample Toluene1. The polarization increases with the diameter, and large fluffy absorbing particles produce higher polarization [6].



Figure 1. Polarization curves for the agglomerates for levitating samples.



Figure 3. Dependence of polarization on size after averaging the data over 50 µm size intervals.



Figure 2. Polarization curves for the small agglomerates for levitating samples.



Figure 4. Polarization curves for the PMMA1 sample measured in different conditions.

Figure 4 shows a comparison of amplitudes of positive polarization branches for the same sample, PMMA1, in different conditions. The highest amplitudes of polarization are

obtained with the two curves produced by the smallest agglomerates and with the packed particles on a surface. The amplitude for both of these curves is about 72 %. A negative polarization is noticed for the packed samples on a surface at large scattering angles. Deposited particles produce lower polarization curves than the packed ones and greater than the one obtained with lifted agglomerates. The lowest polarization amplitude is given by a dense cloud of particles; more studies are needed and are in progress to give an explanation for this result.

CONCLUSIONS

Smallest agglomerates in a range of few μ m in diameter present a higher amplitude of polarization than those obtained with the full set of agglomerates. The polarizations produced by small agglomerates mainly depend on the flow ventilation factor used during the incomplete combustion. Smallest agglomerates and packed particles on a surface give the highest amplitude of polarization. The lowest amplitudes are produced by a dense cloud of levitating agglomerates; further studies are needed to explain this result.

The new version of PROGRA2 using more sensitive cameras allows to detect agglomerates having low brightness. New measurements with other kinds of soot will be studied. Finally the data will be added to the PROGRA2 data base (http://www.icare.univlille1.fr/progra2/). In particular, they will be used to identify the type of the grains of soot detected in the stratosphere with MicroRADIBAL.

REFERENCES

- [1] J.-B. Renard et al. Optical proprieties of randomly distributed soot: improved polarimetric and intensity scattering functions. Appl Opt. **44**(4) (2005).
- [2] C. Brogniez et al. PSC microphysical properties measured by microRadibal instrument on January 25, 2000 above Esrange and modeling interpretation. J. Geophys. Res 108(D6) (2003).
- [3] S. Klusek et al. Compendium of scattering matrix element profiles for soot agglomerates. JQRST **79-80** (2003).
- [4] Yu. Shkuratov et al. Comparative studies of the reflectance and degree of linear polarization of particulate surfaces and independently scattering particles. J. Quant. Spectrosc. Radiat. Transfer 100 (2006).
- [5] J.-B. Renard et al. Light scattering by dust particles in microgravity: Polarization and brightness imaging with the new version of the PROGRA2 instrument. Appl. Opt. 41 (2002).
- [6] E. Hadamcik et al. Light scattering by agglomerates: Interconnecting size and absorption effects (PROGRA² experiment). JQSRT **110**(14–16) (2009).
- [7] F.-X. Ouf et al. Characterization of Soot Particles in the Plumes of Over-Ventilated Diffusion Flames. Combustion Science and Technolog. 180(4) (2008).