

Particulate Carbon Content in Rain at Various Temperate and Tropical Locations

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Abstract. Particulate organic and black carbon concentrations in rain were determined in various source or remote regions, in order to gain information on the incorporation of atmospheric carbonaceous particles in hydrometeors. The analyses of rainwaters indicate that all the samples contained particles derived from combustion. Data obtained on a sample basis, show an important areal and temporal variability of the composition of rain carbonaceous particles, a variability which is reported to that of the black carbon to total carbon ratio, Cb/Ct, ranging from 10 to 72%. In addition to the fluctuations of the aerosol atmospheric burden, these variations may be related to alterations of the organic fraction of the particles or their involvement in in-cloud nucleation processes during atmospheric transport. Also, a comparison of the mean relative abundance of black carbon in aerosols and in rainwaters, gives evidence of a partial disappearance of the organic particles, a phenomenon which could be due to their dissolution when incorporated in the hydrometeors. Precipitation scavenging ratio values of black carbon particles, which range from 100 to 370, are similar to those found for sulphate anthropogenic aerosols. Due to their hygroscopic properties and mean size, black carbon aerosols could possibly trace the physico-chemical processes involved in the incorporation of fine combustion particles into hydrometeors. It is also suggested that smoke particles may act as cloud condensation nuclei (CCN). Consequently, emissions of particulates derived from combustion in some tropical or industrial regions could result locally in alteration of cloud albedo and precipitation regimes.

Key words. Black carbon, particulate organic carbon, rain, scavenging ratio, wet deposition.

1. Introduction

Carbonaceous particles are produced in large amounts by various anthropogenic activities, mainly industrial combustions or biomass burning processes. On the basis of their chemical, optical and thermal properties, two fractions may be distinguished in aerosols: organic carbon (Co), which is always predominant, 50 to 90% (Brémond *et al.*, 1989; Cachier *et al.*, 1989a), and a black carbon component (Cb). Evidence for the dispersion of combustion aerosols in the global troposphere has been provided by the presence of black carbon in remote areas (Rosen *et al.*, 1981; Andreae, 1983; Heintzenberg, 1985; Hansen *et al.*, 1988; Cachier *et al.*, 1990). It is now recognized that increasing concentrations of carbonaceous aerosols in the atmosphere could have a local and even a regional climatic impact, which was the case with oil fires in Kuwait (Hahn, 1991), primarily due to the radiative properties of the particles (Clarke, 1989). In addition to the high light-absorption of black carbon aerosols, an important effect of all the carbonaceous particles could lie in their scattering of incoming radiation as well as the possibility

of them being involved in nucleation processes. It has been hypothesized that these radiative effects have resulted globally in a cooling of the atmosphere and, until now, have masked the aftermaths of the anthropogenic greenhouse effect (Penner *et al.*, 1991). Despite the potential importance of the presence of combustion aerosols in the atmosphere, little attention has been paid to the main tropospheric sink for such particles, i.e. their scavenging by wet deposition processes (Ogren and Charlson, 1984; Ogren *et al.*, 1984). Indeed, up to now, data on particulate carbon concentrations in precipitations have been sparse in comparison with those obtained for the aerosol phase, and mostly restricted to measurements of the black carbon content of snows (Cadle and Dasch, 1988; Noone and Clarke, 1988; Chylek *et al.*, 1987). Thus, the wet removal efficiency for carbonaceous particles remains difficult to assess. As for other aerosols, the removal of particulate carbon is likely to occur via two main scavenging processes: the in-cloud process whereby particles are directly incorporated into cloud droplets; and the below-cloud process, where particles are washed out by the precipitation itself. One of the major unknowns resides in the physico-chemical atmospheric processes which would transform young combustion particles, expected to be hydrophobic, into a water-soluble aerosol phase. The atmospheric behaviour of the carbonaceous particles is likely to be dictated by the chemical nature of their surfaces. If the surface is hydrophobic, the particle remains inactivated. On the other hand, if it is coated with hygroscopic substances, it may be activated enough to be incorporated in water droplets (Ogren and Charlson, 1983). If so, for the in-cloud processes, the role of such particles as cloud condensation nuclei (CCN) could be important (Crutzen and Andreae, 1990; Cachier *et al.*, 1990).

As a first step in evaluating the scavenging of carbonaceous aerosol by precipitation, particulate carbon concentrations were determined in rainwater samples collected over long periods of time at various locations, in source regions or more remote areas. To our knowledge, this is the first set of data for long temporal series. We also show that preliminary insights into the scavenging processes of the carbonaceous particles may be gained from comparison of the incorporation of black carbon, organic carbon and water-soluble aerosols.

2. Experimental

2.1. Collection Sites – Sampling

Rainwater was always collected on an event basis. Samples were obtained in the Paris region during two years, on the CNRS Campus, at Gif-sur-Yvette, and were assumed to characterize the wet deposition in semi-urban source areas. The site of Lamto, in the wooded savanna of the Ivory Coast, was chosen for the study of wet deposition in tropical source regions. Rain was also sampled during one year, in the primary forest of the Northern Congo (Enyele), a region almost unaffected by local human activities, for an assessment of the transport of pollutants derived from

tropical biomass burning. Finally, during a short field experiment, rains were also sampled at the maritime site of Mace-Head (Ireland).

The duration of the different sampling campaigns was long enough to allow the data to be considered as representative of a given atmospheric environment. However, at Lamto (the savanna site), the rainwaters were collected only during the wet season.

2.2. Analysis

Precipitation was sampled using automatic stainless steel rain collectors equipped with 1 litre-glass-bottle. These were covered with a 75 μm mesh sieve to prevent any contamination by vegetation and insect debris. The collector was fitted with a rain detector which releases the opening mechanism as the first drops fall. After each collection, the collector was carefully washed. Rainwater was then rapidly ultrasonicated and filtered on pre-cleaned glass-fibre filters (GF/F, Whatman) to minimize any dissolution of the particles or loss on the walls of the glass-bottle. Rainwater samples which could not be filtered shortly after the collection were protected against biological growth by cooling or adjunction of mercuric chloride. Filter samples were kept frozen until analysis. Prior to the analytical protocol, the particulate samples were decarbonated with 0.2N HCl. After this step, the remaining carbon is referred to as particulate total carbon (Ct), which includes both the organic (Co) and the black carbon (Cb or soot) components. The filters were then analyzed by a thermal method previously optimized for aerosols in our laboratory (Cachier *et al.*, 1989b), and adapted for this study. Black carbon determinations were performed after the thermal removal of the organic component. Both total carbon and black carbon contents were determined by coulometric titration with a Ströhlein Coulomat 702 C. The precision of the analyzer ($C < 1 \mu\text{gC}$) and the reproducibility of blank filter samples allowed the determination of carbon contents down to 3 μg per sample, which is the detection limit of the instrument. The precision of the measurements was, on average, better than 5%. It could, however, reach up to 10% for lightly loaded samples ($C < 20 \mu\text{gC}$). Duplicate analyses have shown that the heterogeneity of the collection surfaces could be the major uncertainty factor. Consequently, in most cases, two samples were collected simultaneously, and devoted to separate Ct and Cb determinations. The mean volume required for optimized filtrations of the rainwaters was 100 ml in source regions and 300 ml elsewhere.

2.3. Validation of the Sampling Protocol of Particles

Two major artefacts may alter the sampling of particles: the attachment of particles on the walls of the rain collector during the collection step and the filter efficiency during the separation of the particulate phase of rainwater (Ogren *et al.*, 1983). In various attempts to quantify these losses, we could notice that any assessment with

pure graphitic calibration standards was not relevant. Indeed, there are important differences in the behaviour of atmospheric particles and graphitic particles, the latter always showing a marked hydrophobic character. Therefore, the quantification of artefacts was conducted on natural rain samples collected in our regions. Ten rainwater samples were analysed for this purpose.

(a) GF/F glass-fibre filters were chosen for the collection of the carbonaceous particles. Due to their unique fibrous matrix, these filters which are universally used for the analysis of particles in waters have a high retention for small particles (98% for particles with $d \geq 0.7 \mu\text{m}$ according to the manufacturer), which is much better than that obtained with quartz-fibre filters. The filtration efficiency was determined by passing the filtrate through a preweighted $0.1 \mu\text{m}$ pore diameter polycarbonate membrane filter. Losses during the filtration were estimated by comparing the total carbon load of the fibre filters with the total aerosol load of the back up nuclepore membrane. These loads show a mean ratio of $3.3 \pm 2.3\%$. As carbon is not the only component of the particulate phase in rainwaters, this average loss of fine particles of the order of 4% may be considered as a maximum, and the filtration efficiency higher than 96%.

(b) Losses on the walls of the sampling device were evaluated only for the collection bottles, where rainwaters may stay a few hours. For this purpose, we could assess the amount of black carbon which had been retained on the walls of the bottle, by rinsing and ultrasounding the container with a diluted detergent solution. This washing solution was then filtered on a GF/F filter, and the blackness of the deposit analyzed optically in transmission with an aethalometer. It must be pointed out that in spite of several rinsing steps, due to the impregnation of the filter by the detergent, only the optical analysis of the deposit was possible. This optical procedure had necessitated to conduct several analyses of detergent-impregnated blank filters. The evaluation of wall losses was then achieved by comparing the optical attenuation of the first filter sample with that of the 'cleaning' solution filter.

This processing has indicated that $7.8 \pm 1.5\%$ of black carbon is attached on the walls of the glass container. It must be pointed out that this result was obtained in the worst conditions, rainwaters being stored during more than one day. In average conditions, losses should be minored.

Thus, following our experimental protocol: a thorough washing of the sampling device prior to the rain collection, ultrasounding the rainwater glass-bottles and collecting the particles on GF/F filters, losses of carbonaceous particles may be estimated to be lower than 10% in mass.

3. Results

3.1. *Particulate Carbon in Rain*

One of the most striking features of our results is the occurrence of noticeable concentrations of black carbon particles in each sample wherever the site. Although

black carbon concentrations were lower at the remote locations, the levels remained always measurable. As black carbon is produced only by combustion processes, it is therefore a suitable tracer of this phenomenon. Consequently, the ubiquitous presence of these particles in our rainwater samples gives evidence of the significant influence of combustion inputs in rains, and confirms the widespread dissemination of combustion aerosols in the global troposphere (Andreae, 1983; Clarke, 1989; Hansen *et al.*, 1988; Cachier *et al.*, 1990).

It must be noticed in Figure 1 that the particulate carbonaceous phase in rain is for the most part of organic nature, this fraction accounting for about 60 to 80% of the total carbonaceous concentration, whatever the sampling site. The predominance of the organic mode in the combustion emissions has been found in previous studies on carbonaceous aerosols (Brémond *et al.*, 1989; Cachier *et al.*, 1989a). Moreover, it must be also underlined that at a given site, the temporal variations of Cb and Ct are similar and that concentrations peak simultaneously. Such a covariation has been already observed for the carbonaceous aerosol (Grosjean, 1984; Gray *et al.*, 1984; Shah *et al.*, 1986; Brémond *et al.*, 1989; Cachier *et al.*, 1989a). This could point to a common origin and common meteorological processes affecting both components (Cb and Co). The linear regression analysis of organic carbon against black carbon concentrations performed for each site, shows that, the Lamto site expected, these two fractions are well correlated (statistically significant at 1% level). As combustions are the only sources of black carbon this strongly suggests that the major part of the organic particulate content of rainwater

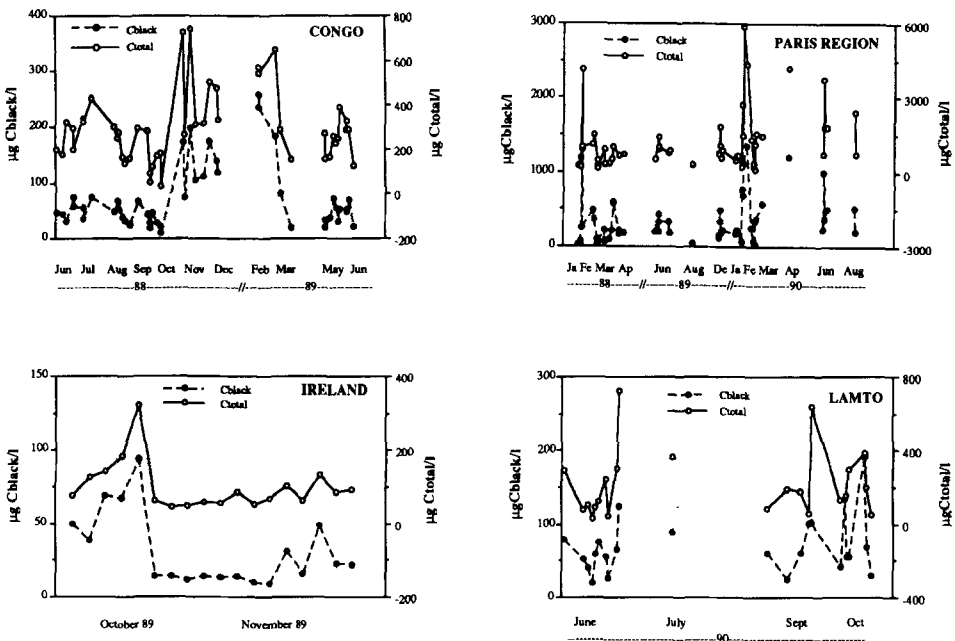


Fig. 1. Black and total carbon contents in rainwaters from different environments.

originates from combustion processes too, fossil-fuel or biomass burning combustions.

Mean values obtained for black and total carbon contents in precipitation are given in Table I. A general decrease of the concentrations is observed from source regions to more remote areas, which is consistent with the dilution of the combustion plumes in the tropospheric air masses and with the removal of these combustion particles from the atmosphere during their transport. The presence of high concentrations in rain in urban areas or in savanna source regions may directly reflect the intensity of the combustion emissions. The case of the equatorial samples (Enyele) is noteworthy, as black and total carbon concentration values measured in these rains compare with those found in source areas, although the sampling site is 100 km from the savanna regions. In a previous work (Cachier and Ducret, 1991), we have shown that due to a favourable conjunction of factors, rains falling over this site are influenced during a large part of the year by major savanna fires raging in Africa. This indicates that pollutants derived from biomass burning may affect rains even at a great distance from the source regions. Rains collected in the wooded savanna of the Ivory Coast during the wet season, when bush fires are almost nonexistent, are also significantly and permanently loaded with combustion particles. This data set confirms the existence of an important background of burning activities (agricultural burns, domestic use of charcoal and wood, clearing practices, etc.), which take place all year long and affect rainwater chemistry in these regions (Crutzen and Andreae, 1990; Delmas *et al.*, 1991).

At each site, both total and black carbon contents are highly variable, as shown in Figure 1. Temporal variations in concentrations may reach two orders of magnitude in source regions. In other areas, the variability appears less important. A comparison with our background aerosol concentration data for temperate and tropical source regions indicates that the variability of the carbonaceous particulate concentrations could be more important in rainwaters than in the aerosols. Fluctuations of the emission sources are not sufficient to explain the variations of particulate carbon contents in the rainwaters. Such variations for both black and total carbon contents are likely to be related to the amount of rainfall and the height and composition of the atmospheric column which is washed by the falling water droplets. It might also reflect processes inside the cloud which would affect concentrations in cloud water, depending on the extent of the hydrodynamic collection of water by the available particles (Mitchell and Lamb, 1989).

3.2. Rain Particulate Composition

Other information can be derived by considering the relative abundance of black carbon (related as the C_b/C_t ratio) in the particulate carbonaceous phase of rainwaters. Despite the apparent covariation of the total and the black carbon contents, the particulate composition of the liquid phase is highly variable. Such C_b/C_t fluctuations were previously reported for the aerosol phase (Brémond *et al.*, 1989;

Table I. Comparison of the arithmetic mean values for particulate carbon in rains sampled in different regions (n : number of samples)

	Concentration ($\mu\text{g C/l}$)			
	C _{black}		C _{total}	
	Range	Mean	Range	Mean
<i>Tropical regions</i>				
(biomass burning processes)				
Savannah (Lamto, Ivory Coast)				
nonfire season: Jun–Oct ($n = 21$)	20–192	69	33–727	226
Equatorial Forest (Enyele, Congo)				
Nov–Mar ($n = 12$)	75–258	155	265–745	477
May–Oct ($n = 33$)	11–75	45	34–430	236
<i>Temperate regions</i>				
(industrial aerosols)				
Paris region (France) ($n = 58$)	27–1348	333	69–5868	1184
Mace Head (Ireland) ($n = 18$)	9–94	31	47–323	100
Seattle (U.S.A.) ($n = 5$) ^a	28–130	60		
Sweden ($n = 60$) ^a	20–600	100		

^a Data from Ogren *et al.* (1984).

Cachier *et al.*, 1989a), and were shown to be related to the variety of combustion processes, mainly governed by the nature of the various combustion fuels and differences in the physical parameters of the burns. The data also indicate temporal changes in source-term apportionment. Thus, the black carbon content variability in rains could result primarily from changes in source apportionment. However, this variability appears to be more pronounced in rainwaters than in the atmospheric aerosols. In addition to the source variability, it is possible that the heterogeneous processes do not identically affect the various carbonaceous particles during their transport in the air masses, which would lead to further variability of the particulate phase composition. The phenomena involved here could be primarily the chemical alteration of the organic aerosol and the incorporation of the particles into the cloud droplets. In relation with the clear areal partition of combustion sources (industrial combustions at temperate latitudes, biomass burning activities mainly in tropical areas), it may be noticed from the results in Table II, that there are significant differences in the mean C_b/C_t ratios. The Gif and Mace Head rain particulate samples both display the same C_b/C_t value of 29%. Given that, at the latitude of both sites, atmospheric carbonaceous particles originate primarily from fossil-fuel combustions, such a C_b/C_t ratio value could be representative of 'industrial' particles after their incorporation in hydrometeors. Furthermore, the similarity of particulate carbonaceous composition in the rains sampled in these two regions could indicate that such aerosols do not undergo noticeable chemical transformations during their atmospheric transport. On the

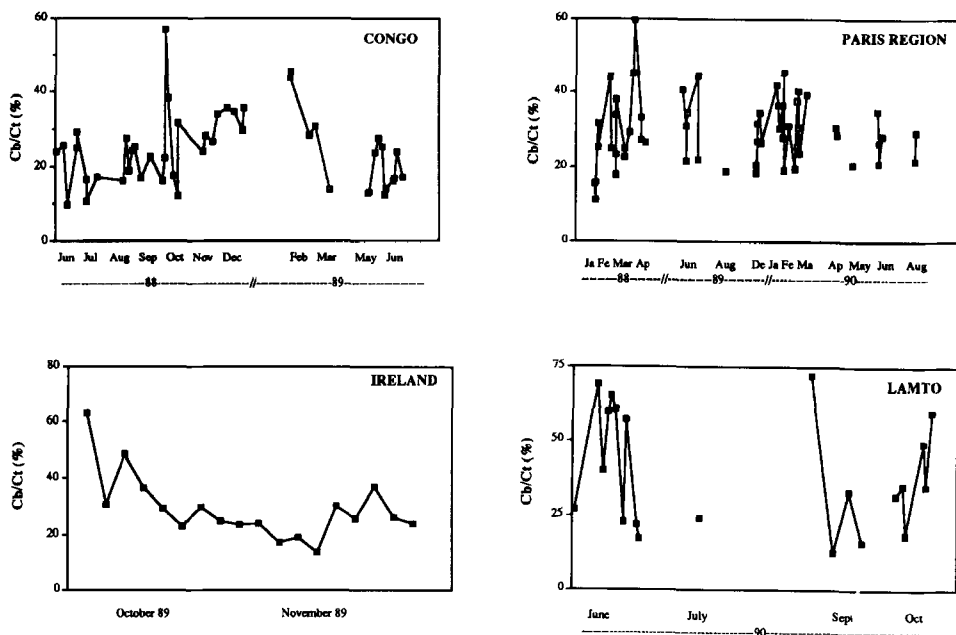


Fig. 2. Relative abundance of the black carbon component in the particulate phase of rains at various locations.

other hand, a simple characterization of the biomass burning particles by their Cb/Ct ratio in rain is not so obvious, since rains sampled at Lamto and at Enyele display significant differences in their composition. Reasons for this discrepancy may be found in the diversity of the combustion processes, as already assessed for the tropical aerosol. This diversity is also reflected in the notable temporal variability at the tropical sites, as shown by the standard deviation of the Cb/Ct values. Also, it must be underlined that such a characterization for tropical biomass burning particles has up to now been difficult to assess, because rain samples were collected only during the rainy season.

Also, from Table II, the comparison of the Cb/Ct ratio in rain and aerosol particles indicates that black carbon is more abundant in rainwaters than in the aerosols. This trend is found whatever the environmental location. As the production of black carbon in the atmosphere is highly unlikely, the relative enhancement of this component could be due to the partial disappearance of the organic fraction from the particulate phase, presumably by dissolution in the hydrometeors. This systematic relative increase in black carbon in rains is more pronounced for biomass burning aerosols than for industrial ones, as suggested by the Lamto tropical source data. This phenomenon gives additional evidence of the differences of origin of the combustion processes which have generated the aerosols. On the average, vegetation burning occurs at a lower temperature than industrial combustion. As the aerosols produced by the former combustion sources are enriched in organic components (Muhlbaier and Williams, 1982; Crutzen *et al.*, 1984; Cachier

Table II. Comparison of the relative abundance of the black carbon fraction in the particulate phase of rain and in the aerosols

	Cb/Ct (%)	
	In rain	In aerosol
Lamto (Ivory Coast)	39 (\pm 19)	12 (\pm 6)
Gif-sur-Yvette (France)	29 (\pm 10)	22 (\pm 4)
Mace Head (Ireland)	29 (\pm 11)	24 (\pm 5)
Enyele (Congo)	24 (\pm 10)	

et al., 1989a), it may be hypothesized that they could undergo chemical transformations more readily, and particularly the organic fraction which could dissolve in the liquid phase. On the other hand, the organic carbonaceous aerosols emitted by industrial combustions, are sufficiently refractory to resist further major chemical alteration during their transport in the atmosphere. The same Cb/Ct value obtained for particles in rain in Ireland and in France is consistent with this hypothesis.

3.3. Scavenging Ratio

The partial dissolution of the organic fraction of the combustion aerosols clearly shows that an accurate assessment of the scavenging of carbonaceous particles from the atmosphere must focus on the particulate black carbon, which apparently does not undergo alterations during these heterogeneous atmospheric processes. The study may be achieved using a parameter frequently reported for wet deposition: the scavenging ratio, a dimensionless number noted S (Slinn, 1984; Barrie, 1985), for which one way of expressing could be

$$S = \frac{[X]_{\text{rain}}}{[X]_{\text{air}}}$$

where $[X]_{\text{rain}}$ is the concentration of a species in rain (in $\mu\text{g/l}$) and $[X]_{\text{air}}$ its concentration in air (in $\mu\text{g/m}^3$). This parameter gives information on the efficiency by which the species is removed from the atmosphere. However, the different S values must be used and interpreted carefully as they are estimated from near-surface air concentrations, which may be quite different from those at the precipitating cloud altitude. Although the scavenging ratio does not account for the physical properties of the incorporation processes, it does provide a way to estimate wet removal rates. The main factors which are known to affect the scavenging ratio (S) include

the size of the particles being scavenged, their physical and chemical form and the cloud properties (Slinn, 1983; Buat-Ménard and Duce, 1986; Jaffrezo and Colin, 1988).

Based on our data set and on data from Table III, the average S value for black carbon particles at all sites, has been calculated to be 200 ± 115 , and the arithmetic mean value from site to site ranges from 100 to 400. These data compare with the few data available in the literature. Moreover, the S values found for black carbon particles are of the order of those reported for sulphate (Cadle and Dash, 1988; Jaffrezo and Colin, 1988), a soluble aerosol, and other particulate pollution aerosol also primarily attached to fine particles (Buat-Ménard *et al.*, 1983; GESAMP report, 1989). This could suggest that on the average black carbon behaves like fine aerosol particles, whose removal from the atmosphere is primarily via wet deposition. This is consistent with the picture of a pollutant aerosol phase, where the particles either form an internal mixture or are rapidly coated with similar water-soluble gaseous and liquid species, leading to a hydrophilic behaviour of the aerosol, whatever the chemical nature of the core (Ogren *et al.*, 1984; Lioussé *et al.*, 1992). Furthermore, the particle size, relative to the age of the aerosol phase, does not seem to be a determinant parameter for the incorporation processes. Although samples were collected at very different sites, the S values do not exhibit an important scatter. This low variability appears somewhat contradictory with results obtained by other authors for pollution aerosols as lead (Patterson and Settle, 1987) or soluble aerosols as sulphate (Cadle and Dash, 1988). If confirmed, our results could indicate that black carbon particles of different sizes behave similarly in the atmosphere.

It is very difficult to determine the causes of the S fluctuations because of large uncertainties generated by differences in the vertical distribution of concentrations in S calculation. It is possible, however, to explain the higher value of 370 found for the Irish rain samples. It could be hypothesized that mechanisms for incorporating black carbon into precipitation may have been more effective in Ireland than in the industrial temperate source region; this phenomenon would be consistent with the notion of a fresh initially-hydrophobic aerosol, thought to be the case in urban source regions, being coated during aging by hygroscopic substances, such as sulphates or organics. Such an explanation was proposed by Ogren *et al.* (1984) for atmospheric particles in Sweden. However, it is also possible that during the period of our field experiment at Mace Head, the important washout of the particles in the atmosphere led to a more uniform vertical distribution of the particles, whereas in source areas the aerosol concentration found at the surface was not satisfactorily representative of the cloud air concentration, leading to an overestimate of the aerosol concentration in the S calculations.

4. Conclusion

The presence of black carbon in each rain sample highlights, as previously found for the aerosol, the large dispersion of combustion inputs in the atmosphere, their

Table III. Average scavenging ratios of black carbon at various locations, calculated from particulate black carbon in rain, and in aerosol at ground level

Location	Mean black carbon concentrations		Scavenging ratio
	In aerosol ($\mu\text{g}/\text{m}^3$)	In rain ($\mu\text{g}/\text{l}$)	
Urban site (Gif-sur-Yvette, France)	2.2 ± 1.3 (Brémond <i>et al.</i> , 1989)	333 ± 321 (this work)	150
Savannah (Lamto, Ivory Coast) nonfire season: Jun–Oct	0.36 ± 0.07 (Cachier <i>et al.</i> , 1989a)	69 ± 40 (this work)	170
Equatorial Forest (Congo) February	2.3 ± 0.65 (Cachier, unpublished data)	247 ± 15 (this work)	100
Marine site (Mace Head, Ireland)	0.084 ± 0.065 (Liousse, 1990)	31 ± 25 (this work)	370
Urban site (Detroit, U.S.A.)			250 (Dash and Cadle, 1986)
Rural site (Michigan, U.S.A.)			160 (Cadle and Dash, 1988)

transport in the troposphere and their ubiquitous influence in rainwater. Our scavenging ratio results give striking evidence that carbonaceous particles incorporate the hydrometeors with an efficiency similar to that of watersoluble aerosols such as sulphates, suggesting that these aerosols gain rapidly a hydrophilic nature. This behaviour is found whatever the combustion source (biomass burning or industrial) which had generated the aerosol. In source regions, considerable amounts of particulate carbon are found in rainwaters. It may be suggested that due to the importance of the local combustion inputs, and accordingly to the high atmospheric burdens of particles, scavenging by below-cloud mechanisms may constitute an important part of the removal processes in these regions.

Our Cb/Ct ratio data for carbonaceous particles in rain and aerosol indicate that a significant part of the organic particulate fraction dissolves in the aqueous phase. Consequently, black carbon only may be used as a tracer of the incorporation of combustion particles into hydrometeors. As the scavenging ratios of black carbon particles compare with those found for water-soluble aerosols, this species could be used satisfactorily as a tracer of these atmospheric incorporation processes.

A problem of considerable concern is the increase of concentrations of combustion particles in the atmosphere, in relation with their climatic impact and their influence on the microphysical and optical properties of clouds. These particles are potential cloud condensation nuclei (CCN), and thus could be responsible for

changes in cloud coverage, perturbation of the hydrological cycle (Crutzen and Andreae, 1990) and counteracting of the greenhouse effect warming (Penner *et al.*, 1991). The size distribution of combustion aerosols displays an important sub-micron mode (Heintzenberg, 1982), which is compatible with such behaviour. Also, the partial dissolution of the particulate organic fraction hypothesized in this study could have happened during the incorporation of the carbonaceous particles in the hydrometeors, suggesting that these particles could serve as CCN. The importance of this phenomenon deserves further investigations and should be completed by dissolved organic carbon (DOC) measurements in precipitations and by simulated leachings of combustion aerosols.

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References

- Andreae, M. O., 1983, Soot carbon and excess fine potassium: Long-range-transport of combustion-derived aerosols, *Science* **220**, 1148–1151.
- Barrie, L. A., 1985, Scavenging ratios, wet deposition and in-cloud oxidation: an application to the oxides of sulphur and nitrogen, *J. Geophys. Res.* **90**, 5789–5799.
- Brémond, M. P., Cachier, H., and Buat-Ménard, P., 1989, Particulate carbon in the Paris region atmosphere, *Environ. Techn. Lett.* **10**, 339–346.
- Buat-Ménard, P., Ezat, U., and Gaudichet, A., 1983, Size distribution and mineralogy of aluminosilicate dust particles in tropical Pacific air and rain, in H. R. Pruppacher *et al.* (eds.), *Precipitation Scavenging, Dry Deposition and Resuspension*, Elsevier, New York, pp. 1259–1269.
- Buat-Ménard, P. and Duce, R., 1986, Precipitation scavenging of aerosol particles over remote marine regions, *Nature* **321**, 508–510.
- Cachier, H., Brémond, M. P., and Buat-Ménard, P., 1989a, Carbonaceous aerosols from different tropical biomass burning sources, *Nature* **340**, 371–373.
- Cachier, H., Brémond, M. P., and Buat-Ménard, P., 1989b, Determination of atmospheric soot carbon with a simple thermal method, *Tellus* **41B**, 379–390.
- Cachier, H., Brémond, M. P., and Buat-Ménard, P., 1990, Organic and black carbon aerosols over marine regions of the Northern Hemisphere, in Newman and Kiang (eds.), *Proceedings of the International Conference on Global Atmospheric Chemistry*, Beijing, 1989, Brookhaven National Lab. Press, pp. 241–261.
- Cachier, H. and Ducret, J., 1991, Particulate carbon traces biomass burning influence in rains of Equatorial Africa, *Nature* **352**, 228–230.
- Cadle, S. H. and Dash, J. M., 1988, Wintertime concentrations and sinks of atmospheric particulate carbon at a rural location in northern Michigan, *Atmos. Environ.* **22**, 1373–1381.
- Chylek, P., Srivastava, V., Cahenzli, L., Pinnick, R. G., Dod, R. L., Novakov, T., Cook, T. L. and Hinds, B. D., 1987, Aerosol and graphitic carbon content of snow, *J. Geophys. Res.* **92**, 9801–9809.
- Clarke, A. D., 1989, Aerosol light absorption by soot in remote environments, *Aer. Sci. Techn.* **10**, 161–171.

- Crutzen, P. J., Galbally, I. E., and Bruhl, C., 1984, Atmospheric effects from post-nuclear fires, *Climatic Change* **6**, 323–364.
- Crutzen, P. J. and Andreae, M. O., 1990, Biomass burning in the Tropics: Impact on atmospheric chemistry and biological cycles, *Science* **250**, 1669–1678.
- Dash, J. M. and Cadle, S. H., 1986, Atmospheric carbon particles in an urban area: Wintertime sources and sinks, General Motors Res. Publ. GMR-5778.
- Delmas, R. A., Loudjani, P., Podaire, A., and Menaut, J. P., 1991, Biomass burning in Africa, in Levine J. S. (ed.), *Global Biomass Burning*, MIT Press, Cambridge, Mass., pp. 126–132.
- GESAMP (Joint Group of Experts on Scientific Aspects of Marine Pollution), 1989, *The Atmospheric Input of Trace Species to the World Ocean*, Reports and Studies GESAMP 38, World Meteorological Organization.
- Gray, H. A., Cass, G. R., Huntzicker, J. J., Heyerdahl, E. K., and Rau, J. A., 1984, Elemental and organic particle concentrations: a long-term perspective, *Sci. Total Environ.* **36**, 17–25.
- Grosjean, D., 1984, Particulate carbon in Los Angeles air, *Sci. Total Environ.* **32**, 133–145.
- Hahn, J., 1991, Environmental effects of the Kuwaiti oil fires, *Environ. Sci. Technol.* **25**, 1530–1532.
- Hansen, A. D. A., Bodhaine, B. A., Dutton, E. G., and Schnell, R. C., 1988, Aerosol black carbon measurements at the South Pole: initial results, 1986–87, *Geophys. Res. Lett.* **15**, 1193–1196.
- Heintzenberg, J., 1982, Size-segregated measurements of particulate elemental carbon and aerosol light absorption at remote Arctic locations, *Atmos. Environ.* **16**, 2461–2469.
- Heintzenberg, J., 1985, What can we learn from aerosol measurements at baseline stations? *J. Atmos. Chem.* **3**, 153–169.
- Jaffrezo, J.-L. and Colin, J.-L., 1988, Rain-aerosol coupling in urban area: Scavenging-ratio measurement and identification of some transfer processes, *Atmos. Environ.* **22**, 929–935.
- Lioussé, C., 1990, Optical properties of black carbon. (in French) DEA, University of Paris VII.
- Lioussé, C., Cachier, H., and Jennings, S. J., 1992, Optical and thermal measurements of black carbon aerosol content in different environments: variation of the specific attenuation cross section, σ , *Atmos. Environ.* in press.
- Mitchell, D. L. and Lamb, D., 1989, Influence of riming on the chemical composition of snow in winter orographic storms, *J. Geophys. Res.* **94**, 14831–14840.
- Muhlbaier, J. L. and Williams, R. L., 1982, Fireplaces, furnaces and vehicles as emissions sources of particulate carbon. In G. T. Wolff and R. L. Klimisch (eds.), *Particulate Carbon: Atmospheric Life Cycle*, Plenum Press, New York, pp. 185–199.
- Noone, K. J. and Clarke, A. D., 1988, Soot scavenging measurements in Arctic snowfall, *Atmos. Environ.* **22**, 2773–2778.
- Ogren, J. A. and Charlson, R. J., 1983, Elemental carbon in the atmosphere: Cycle and lifetime, *Tellus* **35B**, 241–254.
- Ogren, J. A., Charlson, R. J., and Groblicki, P. J., 1983, Determination of elemental carbon in rain-water, *Anal. Chem.* **55**, 1569–1572.
- Ogren, J. A. and Charlson, R. J., 1984, Wet deposition of elemental carbon and sulfate in Sweden, *Tellus* **36B**, 262–271.
- Ogren, J. A., Groblicki, P. J., and Charlson, R. J., 1984, Measurement of the removal rate of elemental carbon from the atmosphere, *Sci. Total Environ.* **36**, 329–338.
- Patterson, C. C. and Settle, D. M., 1987, Review of data on eolian fluxes of industrial and natural lead to the lands and seas in remote regions on a global scale, *Mar. Chem.* **22**, 137–162.
- Penner, J. E., Ghan, S. J., and Walton, J. J., 1991, The role of biomass burning in the budget and cycle of carbonaceous soot aerosols and their climate impact, in J. S. Levine (ed.), *Global Biomass Burning*, MIT Press, Cambridge, Mass., pp. 387–393.
- Rosen, H., Novakov, T., and Bodhaine, B. A., 1981, Soot in the Arctic, *Atmos. Environ.* **15**, 1371–1374.
- Shah, J. J., Johnson, R. L., Heyerdahl, E. K., and Huntzicker, J. J., 1986, Carbonaceous aerosol at urban and rural sites in the United States, *J. Air Pollut. Contr. Assoc.* **36**, 254–257.
- Slinn, W. G. N., 1983, Air-to-sea transfer of particles, in P. S. Liss and W. G. N. Slinn (eds.), *Air-Sea Exchange of Gases and Particles*, D. Reidel, Dordrecht, pp. 299–405.
- Slinn, W. G. N., 1984, Precipitation scavenging, in D. Randerson (ed.), *Atmospheric Sciences and Power Production*, Oak Ridge, pp. 466–532.