Short communication

Color-related differences in the chemical composition of aerosol-laden filters

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Abstract

We examined color-related differences in the chemical composition of ~ 6000 aerosol filters collected from 1989 to 1997 at three sampling sites in the North Atlantic Ocean (Barbados, Bermuda, and Izaña) operated for the Atmosphere/Ocean Chemistry Experiment (AEROCE). The concentrations of the trace elements (Al, Br, Ca, Cl, Fe, Na, Sb, Sc, Se, V) and two ions, NO_3^- and SO_4^{2-}; the activities of ^{210}Pb and ^7Be; and the mixing ratios of ozone were compared and contrasted for groups of samples and data matched to the three basic colors of aerosol filters (brown, gray or white). Chemical composition is related to filter color, that is, the brown and gray samples correspond to mineral dust and absorbing aerosol from various pollution sources, respectively, although these substances often are mixed in varying proportions. The white filters are best regarded as indicating the absence or low concentrations of dust or other absorbing aerosols, and they are often indicative of clean marine air. Relationships among atmospheric substances differ significantly in the color-stratified data subsets, and grouping the samples by color provides unique insight into the relationships among mineral dust, pollution aerosol, and other substances over the North Atlantic. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

The fundamental problem of understanding the chemical components of aerosol samples has been addressed by a variety of approaches, including factor analysis (e.g. Henry, 1997); single-particle analysis (Anderson et al., 1996); trajectory analysis (Moody and Galloway, 1988); and graphical methods (Rahn, 1999). Many years of handling and analyzing samples for the Atmosphere/Ocean Chemistry Experiment (AEROCE) has led us to try a simple approach, that is investigating the extent to which the brown, gray and white filter samples correspond to three main types of aerosol (crustal, pollution, marine) observed over the semitropical North Atlantic

Ocean. The seasonal differences in color of air filters from Miami (red-brown in summer and gray in winter) have been previously described by Prospero et al. (1987), but a more comprehensive study of color-related differences in chemistry of aerosols has become possible only after detailed analyses of the AEROCE filters were completed.

The basic strategy for this study was to (1) group by color ~ 6000 aerosol filters collected from 1989 to 1997 at three island sites in the North Atlantic, and (2) examine the compositional differences and relationships among various atmospheric constituents in data matched to the color stratified aerosol samples.

2. Materials and methods

The aerosol samples for trace element analyses were collected at three sampling sites operated for AEROCE:

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Ragged Point, Barbados (13.18°N, 59.43°W); Tudor Hill, Bermuda (32.24°N, 64.87°W); and the Izaña Observatory, Tenerife (28.30°N, 16.48°W, 2360 m above sea level) (for details of the sampling procedures see Arimoto et al., 1995). The average volume sampled during the 24-h sampling period at Barbados and Bermuda was $\sim 1000\,m^3$. At Izaña, the average volume was $\sim 500\,m^3$ because the aerosol samplers operated only during the downslope flow at night to sample free-tropospheric air. There were no major differences between average volumes of air for the samples of different colors. Elemental concentrations for these samples were determined by instrumental neutron-activation analyses (INAA, Arimoto et al., 1992).

The color of the exposed filters was determined by eye in a clean room while the filters were being prepared for the analyses. At Bermuda, the colors of the filters were also determined in the field by a site operator. Out of 400 filters viewed by two independent observers, about 25% were classified differently, especially when they were the lightest shades of gray or brown. Differences in perception of color among persons and in the type of lighting used for viewing the filters in the clean room and in the field were probably responsible for these differences in classification. Another complication is the fact that brown desert dust, gray pollutants, and gray biomass burning products are often mixed en route to the sampling sites (Savoie et al., 1989).

Aerosol samples for the radionuclide studies were collected from 1989 to 1993 without wind sector controllers using a dedicated system with quartz-fiber filters (Graustein and Turekian, 1996). The two nuclides of interest are ^{210}Pb, which is supplied to the atmosphere by the radioactive decay of ^{222}Rn which emanates from soils, and ^7Be, which is produced in the upper troposphere/lower stratosphere by spallation reactions. Data for these two naturally occurring radionuclides were generated by a gamma-counting technique (Graustein and Turekian, 1996). Ozone data were provided by Oltmans (CMDL, National Oceanic and Atmospheric Administration, Boulder, CO80303), and the concentrations of Na and the major ions Cl^-, NO_3^-, and SO_4^{2-}, were provided by Savoie (University of Miami, Miami, FL 33149). Non-sea salt sulfate (nss SO_4^{2-}) was calculated as total sulfate minus the sulfate from sea salt, which was determined using Na as a reference element (Savoie et al., 1989).

We grouped the data for 5913 sample filters by color and compared the chemical characteristics of the three groups for each of the three sites. This large data set was broken down into about 2000 samples of each color with several hundred of each color at each site. Half of the samples at Izaña were white, whereas at Bermuda and Barbados the dominant colors were gray (54%) and brown (44%), respectively. We used multivariate analyses of variance (MANOVAs, Walpole and Myers, 1978)

to determine whether the differences in the concentrations and ratios of trace elements, ions, and radionuclides in the color groups from the three sites were significant. The ozone data matched to the collection periods for the aerosol samples were analyzed in the same way.

3. Results and discussion

The most pronounced differences in the color-stratified samples from Barbados were for the crustal elements (Al, Fe, Sc) whose concentrations were highest in the brown samples (Table 1); this was also true at Izaña and Bermuda. The marine elements (Na, Cl, Br) did not exhibit strong color-related differences, presumably because sea-salt particles are effectively colorless. The concentrations of Sb, a pollution indicator, and Se, which has mixed anthropogenic and marine sources, differed much less among color groups than did the crustal elements. In the brown samples from Izaña and Barbados, the Sb/Al and Se/Al ratios closely resembled those in dust from the Sahara (Schütz and Rahn, 1982) even though these elements are commonly enriched in the atmosphere over crustal values (Arimoto et al., 1995). Therefore while dust and other types of light-absorbing aerosols often are transported in the same air masses, it is not uncommon for dust over the North Atlantic to be transported in air that does not contain high concentrations of trace element pollutants. At Barbados and Izaña, the concentrations of sulfate, nitrate, and ^{210}Pb were consistently high in brown samples while the mixing ratios of ozone and the $^7Be/^{210}Pb$ ratios peaked in data matched to the white samples. The highest concentrations of Se, Sb, sulfate, nitrate, O_3, ^7Be, and Sb/Al ratios occurred in gray samples from Bermuda, and this is consistent with the known substantial levels of pollution at this site (e.g. Arimoto et al., 1995).

At Barbados, the majority of the color-related differences in the concentrations and ratios of selected species were statistically significant (Table 2) with the exception of Cl, Na, Ca in white and gray samples. For the other two sites, the highest numbers of statistically significant differences were found between brown and white samples at Izaña (18/19) and brown and gray samples at Bermuda (16/19). Interestingly, the concentrations of sulfate, nitrate, ^{210}Pb, and ^7Be at Bermuda differed significantly between the white and gray samples even though the trace elements did not. At Izaña, the gray and white samples differed only in Sb, ^{210}Pb, and ^7Be and the $^7Be/^{210}Pb$ ratios. Color-related differences in Fe/Al and other X/Al ratios (where X is any element of interest) for several crustal elements (Co, Sc, Th) not shown in Table 2 were not statistically significant, presumably due to dominating influence of Saharan dust.

The concentrations of $nssSO_4^{2-}$ over the North Atlantic previously have been shown to be correlated

Table 1
Arithmetic mean concentrations of selected substances and ratios in aerosol samples of different color from Barbados (1991–1997)

Variable	Color[a]	n[b]	Mean	Std. error	Variable	Color	n	Mean	Std. error
Al	b	1027	2.33E + 00	8.00E − 02	Fe/Al	b	590	5.95E − 01	1.80E − 02
	g	804	3.95E − 01	1.90E − 02		g	236	5.63E − 01	1.50E − 02
	w	334	1.47E − 01	2.20E − 02		w	365	6.38E − 01	1.70E − 02
Br	b	736	2.30E − 02	2.00E − 03	Sb/Al	b	519	2.10E − 05	1.02E − 06
	g	716	2.00E − 02	1.00E − 03		g	154	9.59E − 05	1.50E − 05
	w	461	2.00E − 02	1.00E − 03		w	95	1.00E − 03	1.54E − 04
Ca	b	718	8.74E − 01	2.90E − 02	Se/Al	b	612	3.25E − 05	6.00E − 06
	g	535	2.55E − 01	9.30E − 02		g	343	2.08E − 03	2.20E − 04
	w	303	2.78E − 01	2.50E − 02		w	318	2.26E − 02	2.24E − 03
Cl	b	1005	9.88E + 00	2.42E − 01	V/Al	b	1017	1.00E − 03	1.29E − 05
	g	823	9.15E + 00	2.99E − 01		g	677	4.00E − 03	2.47E − 03
	w	496	1.00E + 01	5.58E − 01		w	220	1.40E − 02	2.00E − 03
Fe	b	602	1.25E + 00	4.30E − 02	O$_3$	b	254	1.74E + 01	2.09E − 01
	g	244	2.86E − 01	2.30E − 02		g	107	2.07E + 01	4.32E − 01
	w	282	7.40E − 02	1.20E − 02		w	149	2.32E + 01	4.86E − 01
Na	b	1013	5.87E + 00	1.39E − 01	NO$_3^-$	b	627	7.14E − 01	1.40E − 02
	g	832	5.46E + 00	1.70E − 01		g	259	6.60E − 01	3.40E − 02
	w	500	5.89E + 00	3.33E − 01		w	381	2.80E − 01	1.00E − 02
Sb	b	527	3.81E − 05	1.07E − 06	nssSO$_4^{2-}$	b	624	1.07E + 00	2.30E − 02
	g	158	2.07E − 05	1.52E − 06		g	251	7.08E − 01	3.00E − 02
	w	128	1.28E − 05	9.80E − 07		w	367	3.78E − 01	1.50E − 02
Sc	b	600	4.50E − 04	1.54E − 05	^{7}Be	b	308	3.40E + 00	7.20E − 02
	g	245	9.51E − 05	7.87E − 06		g	140	3.57E + 00	1.22E − 01
	w	312	2.29E − 05	3.73E − 06		w	192	2.81E + 00	8.60E − 02
Se	b	600	4.36E − 04	6.80E − 06	^{210}Pb	b	308	5.60E − 01	1.42E − 02
	g	246	3.54E − 04	1.02E − 05		g	140	4.15E − 01	1.80E − 02
	w	358	2.38E − 04	5.33E − 06		w	190	1.61E − 01	8.00E − 03
V	b	1017	3.00E − 03	9.28E − 05	^{7}Be/^{210}Pb	b	308	6.86E + 00	1.90E − 01
	g	689	1.00E − 03	2.93E − 05		g	140	1.01E + 01	4.39E − 01
	w	270	4.13E − 04	4.35E − 05		w	189	2.69E + 01	2.47E + 00

[a] Color key: b = brown, g = gray, w = white.
[b] n stands for number of samples.

with those of atmospheric dust (e.g. Li-Jones and Prospero, 1998). Mineral dust carried in the same air-masses as non-sea-salt sulfate provides potential reaction surfaces, such as for the reaction of SO$_2$ on calcium-rich mineral particles (Dentener et al., 1996). Whether such reactions actually occur can only be determined through further studies, but the analyses that follow demonstrate that nssSO$_4^{2-}$ and mineral dust share common pathways in the atmosphere over the North Atlantic and that the relationships between these substances vary in relation to the colors of the aerosols collected on sample filters.

Using our color criteria, we found functional differences in the relationships between Al and nssSO$_4^{2-}$ in filters of different colors. As shown in a scatterplot of dust (as represented by Al) versus nssSO$_4^{2-}$ at Barbados (Fig. 1) and Izaña (not shown), these two substances display a clear positive relationship in brown and gray samples but not in white ones. The percentage of the variance in nssSO$_4^{2-}$ explained by its relationship with Al was 43, 15 and 8% for the brown, gray and white groups,

respectively. For all groups combined, the dust-associated nssSO$_4^{2-}$ (estimated from the explained variance from the Al versus nssSO$_4^{2-}$ correlations) is 66%, quite comparable to the 60% estimated by Savoie et al. (1989).

At Izaña, the seasonally averaged concentrations of nssSO$_4^{2-}$ in brown samples (but not in gray or white) tracked those of Al (correlation coefficient, $r = 0.51$; number of samples, $n = 22$; probability for chance occurrence, $p = 0.015$, Fig. 2) and Ca ($r = 0.75$; $n = 21$; $p = 0.0001$, not shown). Using the approach described above, we calculate that at Izaña 51% of the nss sulfate is associated with dust, explaining 37, 0.17, and 14% of the respective variance in the brown, gray, and white groups.

Seasonal patterns in ozone, ^{7}Be, and ^{210}Pb in samples of different colors revealed further differences between white and brown samples from Izaña. Summertime ozone at Izaña previously was shown to be anticorrelated with aerosol pollutants (NO$_3^-$, nssSO$_4^{2-}$) and ^{210}Pb and positively correlated with ^{7}Be, suggesting potential influences from upper tropospheric/lower stratospheric

Table 2
Multivariate analysis of variance (MANOVA) testing for statistically significant differences between sample groups of different colors at Barbados, Bermuda, and Izaña (pairs with a probability for chance occurrence < 0.05 are shown in boldface)

| Variable	Barbados			Bermuda			Izaña		
	Brown vs. white	Brown vs. gray	White vs. gray	Brown vs. white	Brown vs. gray	White vs. gray	Brown vs. white	Brown vs. gray	White vs. gray
Al	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.246	**< 0.001**	**< 0.001**	0.487
Ca	**< 0.001**	**< 0.001**	0.635	**< 0.001**	**< 0.001**	0.531	**< 0.001**	**< 0.001**	0.811
Cl	**0.003**	**0.032**	0.178	0.056	0.055	0.924	0.842	0.490	0.719
Fe	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.820	**< 0.001**	**< 0.001**	0.492
Na	**0.004**	**0.034**	0.195	0.122	**0.005**	0.116	**0.008**	**0.003**	0.609
Sb	**< 0.001**	**< 0.001**	**< 0.001**	0.869	**< 0.001**	**< 0.001**	**< 0.001**	0.063	**0.048**
Sc	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.886	**< 0.001**	**< 0.001**	0.539
Se	**< 0.001**	**< 0.001**	**< 0.001**	0.134	**< 0.001**	**< 0.001**	**0.025**	0.601	0.087
V	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.173	**< 0.001**	**< 0.001**	0.202
Fe/Al	0.083	0.257	**0.028**	0.404	0.887	0.087	**< 0.001**	0.487	0.157
Sb/Al	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.527	**< 0.001**	**< 0.001**	0.119
Se/Al	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**0.022**	**< 0.001**	**< 0.001**	0.085
V/Al	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.132	**< 0.001**	**0.009**	0.947
O_3	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.645
NO_3^-	**< 0.001**	0.127	**< 0.001**	0.570	**< 0.001**	**< 0.001**	**0.001**	0.534	0.147
$nssSo_4^{2-}$	**< 0.001**	**< 0.001**	**< 0.001**	0.522	**0.001**	**< 0.001**	**0.009**	0.551	0.057
^{7}Be	**< 0.001**	0.617	**< 0.001**	0.233	**< 0.001**	**< 0.001**	**< 0.001**	0.752	**0.013**
^{210}Pb	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	0.983	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**
^{7}Be/^{210}Pb	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**< 0.001**	**0.003**
No. significant/total	18/19	16/19	16/19	11/19	16/19	9/19	18/19	12/19	4/19
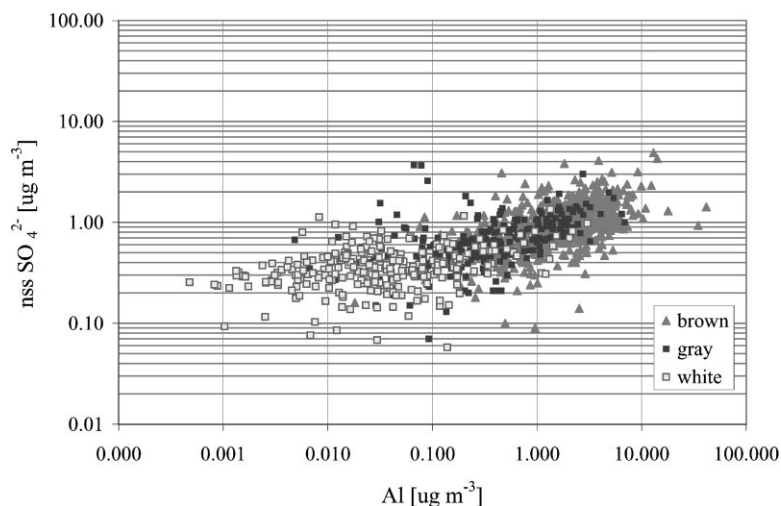
Fig. 1. Non-sea salt sulfate ($nssSO_4^{2-}$) versus Al in brown, gray, and white samples from Barbados (1991–1994).

sources (Prospero et al., 1995). This is consistent with the anticorrelation evident in a scatterplot of ozone versus ^{210}Pb (Fig. 3, $r = -0.48$; $n = 176$; $p = 0.0001$) and a positive correlation between O_3 and ^{7}Be ($r = 0.35$; $n = 176$; $p = 0.0001$) for O_3, ^{210}Pb, and ^{7}Be matched to the brown samples, which are most common in summer.

In contrast, O_3 and ^{210}Pb matched to the white samples, which are least frequent in summer, were weakly but significantly correlated ($r = 0.16$; $n = 586$; $p = 0.0001$, Fig. 3), and ^{7}Be was correlated with ^{210}Pb in data matched to white ($r = 0.37$; $n = 627$; $p = 0.0001$) but not brown samples ($r = 0.006$; $n = 183$; $p = 0.92$, Fig. 4).
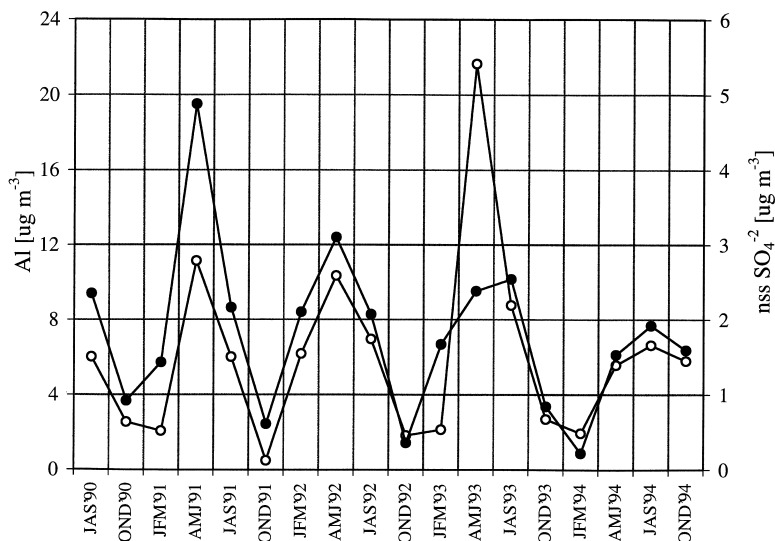
Fig. 2. Seasonal cycles of Al (open circles), nssSO$_4^{2-}$ (filled circles) in brown samples from Izaña.
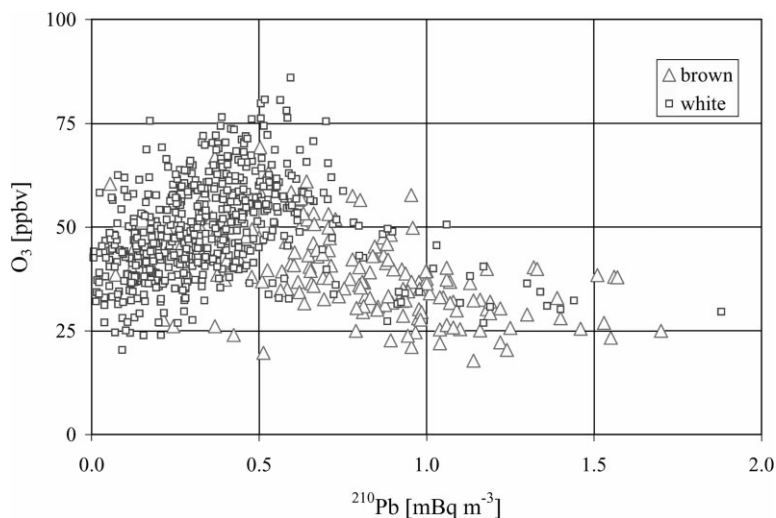

Fig. 3. Ozone versus ^{210}Pb in samples matched to brown and white filters from Izaña (1989–1993).

The seasonal cycle for ozone matched to white samples closely corresponds to that of ^{7}Be (Fig. 5), and the correlation between O$_3$ mixing ratios and ^{7}Be activities matched to the white filters was stronger ($r = 0.59$; $n = 585$; $p = 0.0001$) than for the brown samples. These results suggest that the coupling between O$_3$ and ^{7}Be, possibly through upper tropospheric/lower stratospheric influences, is stronger when conditions are not conducive to dust transport.

One possible explanation for the observed relationships for data matched to the white filters is that ^{210}Pb evidently can be produced in the upper atmosphere from ^{222}Rn transported aloft in wet convective updrafts

(Balkanski et al., 1993). Gaseous ^{222}Rn escapes scavenging while the dust and pollution aerosols that would cause the filters to be colored are scavenged by precipitation associated with the updrafts. Another possibility is that the correlation between ^{7}Be and ^{210}Pb is driven by mixing, that is, that air transported from the high latitudes (and high altitudes) mixes with air from the low latitudes containing lower concentrations of these nuclides (Savoie et al., 1992).

In summary, this first attempt to relate observed differences in the colors of air filters to aerosol composition and the relationships among atmospheric substances presents only a few basic findings due to the limited scope
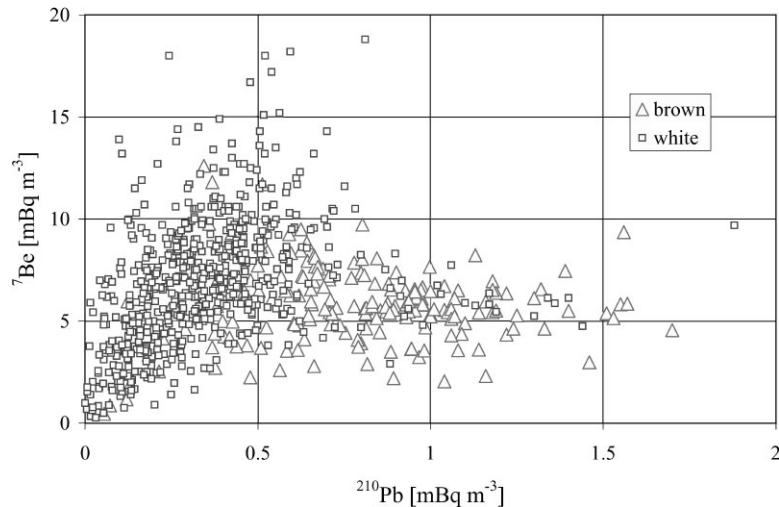
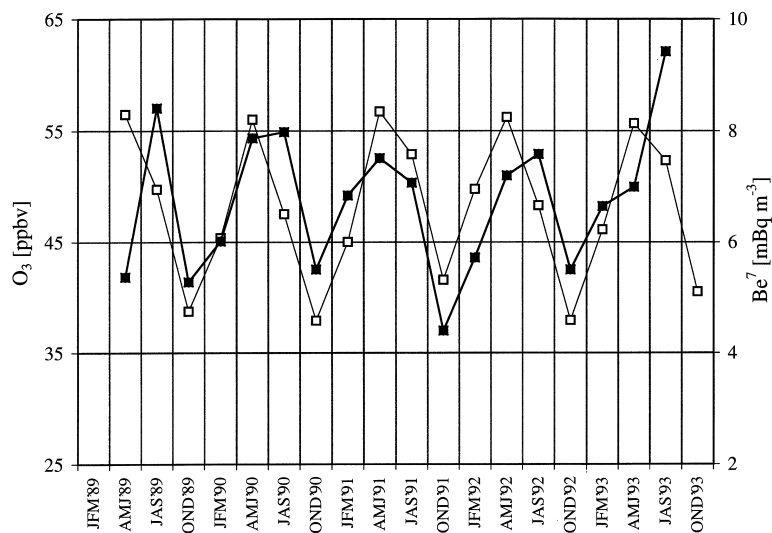Fig. 4. ^{7}Be versus ^{210}Pb in samples matched to brown and white filters from Izaña (1989–1993).

Fig. 5. Seasonal cycles in ozone (open squares) and ^{7}Be (filled squares) in samples matched to white filters from Izaña.

of the paper. We establish that differences in chemical composition in the brown, gray and white aerosol deposits in samples collected from over the North Atlantic are statistically significant. The brown samples are characterized by the highest concentrations of crustal elements (Al, Fe, Sc), and, especially at Barbados, the lowest X/Al ratios of pollution elements (Se, Sb) closely resemble those in dust from the Sahara. The gray samples with much lower concentrations of crustal elements are highly enriched in trace element pollutants (Sb, Se). The gray samples differ in composition between three sites, most likely reflecting the fact that the gray color can result from both biomass burning or pollution emissions whose

influences differ among the sampling sites. The white samples are probably best viewed as indicating the absence of high concentrations of dust or other absorbing aerosols.

One of the unique advantages of grouping the data by filter color is that it allows investigators to compare and contrast relationships between chemical constituents of atmospheric aerosols and their temporal patterns in subgroups of large data sets. The previously unreported correlation between ^{7}Be and ^{210}Pb in white filters and the strong correspondence in the annual cycles of O_3 and ^{7}Be for data matched to the white filters from Izaña are just two examples of furthering our understanding of

atmospheric processes by the color approach. In spite of obvious limitations, including the mixing of aerosol types and the non-specific nature of the gray-colored filters, stratifying the data by color provides a useful starting point for more in-depth analyses. Furthermore, with the development of digitalimaging technology and other instrumental methods, the subjective element of the filter-color determinations will be greatly reduced, if not eliminated completely.

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