

**Distribution and Properties of Carbonaceous aerosols:  
An Analysis of Existing Data  
Final Report to NASA's Global Aerosol Climatology Project**

Principal Investigator: T. Novakov

([tnovakov@lbl.gov](mailto:tnovakov@lbl.gov))

Environmental Energy Technologies Division  
Lawrence Berkeley National Laboratory, MS-73  
Berkeley, CA 94720

## **I. SUMMARY**

Direct and indirect aerosol climate forcings depend on the physical and chemical properties of the total aerosol including sulfates, other water-soluble inorganic compounds, carbonaceous material, sea salt, and mineral particles. Until recently sulfate aerosols have received most attention because its optical and nucleation properties, sources, and spatial distributions are relatively well known. Recent studies, however, have identified carbonaceous aerosols as one of the most important contributors to aerosol forcing. Carbon containing aerosols are composed of organic (OC) and black (BC) carbon. Both OC and BC influence direct aerosol climate forcing. OC scatters light. BC, in addition to being a light scatterer, is the most important light absorbing aerosol species. OC through its CCN (cloud condensation nucleus) activity contributes to indirect forcing.

In contrast to sulfate, the data on the concentrations, distributions, sources, and radiative properties of the carbonaceous components of aerosols are much scarcer. In this project we assembled, analyzed and systematized empirical data relevant to assessing the role of carbonaceous aerosols in climate forcing. This was to be achieved by a) compiling and analyzing the published data on mass concentrations of total (TC), organic (OC) and

black carbon (BC), as well as, the physical and chemical properties of these and other aerosol components, and b) examining the changes in past fossil fuel utilization practices and their effects on regional BC emissions. The main outcomes of this project are as follows:

*Derivation of historical BC aerosol emissions from fossil fuels.* Fossil fuel burning is the common source of BC and much of anthropogenic CO<sub>2</sub>, SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>. BC emissions, unlike CO<sub>2</sub> and SO<sub>2</sub>, critically depend on the combustion efficiency in addition to the mass of fuel consumed. Past fossil fuel BC emissions were vastly different in industrialized and in developing countries because of the differences in fuel utilization and combustion efficiency.

*Analysis of field data published in the past 20 years.* This analysis resulted in an empirical assessment of the relative contributions of sulfate and carbonaceous material to ambient aerosols, spatial distributions, temporal variations of mass concentrations, and BC/OC and BC mass fractions (BC/aerosol mass ratios).

*Utilization of regional of BC/OC ratios* to estimate the relative contribution of biomass (or biofuel) and fossil fuel combustion to carbonaceous aerosol over Indian Ocean.

## **II. CHANGES IN REGIONAL BC EMISSIONS FROM FOSSIL FUEL COMBUSTION DURING 20<sup>TH</sup> CENTURY**

[T.W. Kirchstetter, J.E. Sinton, and J.A. Sathaye contributed to the derivation fuel inventories]

Fossil fuel burning is the common source of BC and much of anthropogenic CO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>. Whereas CO<sub>2</sub> and uncontrolled SO<sub>2</sub> emissions depend only on the mass of fuel burned and its carbon and sulfur content, BC emissions critically depend on the combustion efficiency in addition to the mass of fuel consumed. Fuel utilization during the last century has radically changed from inefficient to more efficient in industrialized countries. However, in the developing regions no major changes in fuel utilization did not occur. Because inefficient combustion produces much more BC (per unit fuel mass) than

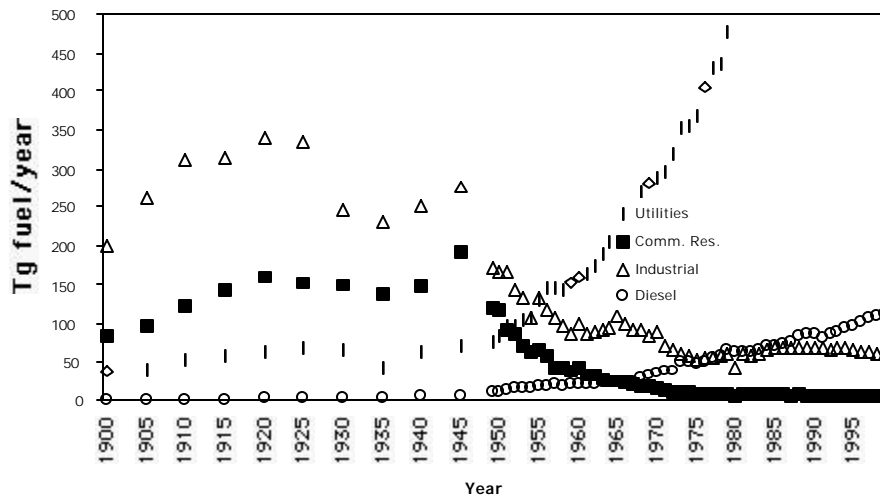
efficient burning it is obvious that BC emissions from industrialized and developing regions showed strikingly different temporal trends and dependence on total fossil fuel consumption.

Our approach involves assembling historical data on diesel fuel and coal consumption (assumed to be the principal fossil fuel BC sources) segregated into domestic, commercial, industrial and electric power generation sectors. BC emission factors are largest for coal burning by domestic/commercial sectors and lowest by coal burning in electric power generation plants. Time series of BC emissions (in  $\text{Tg y}^{-1}$ ) were estimated from sectoral fuel consumption data and sector-specific and emission factors (in g BC per kg of fuel) [Cooke *et al.*, 1999]. We have derived the time series of BC emissions for United States, United Kingdom, Germany (eastern and western parts), former Soviet Union, India and China.

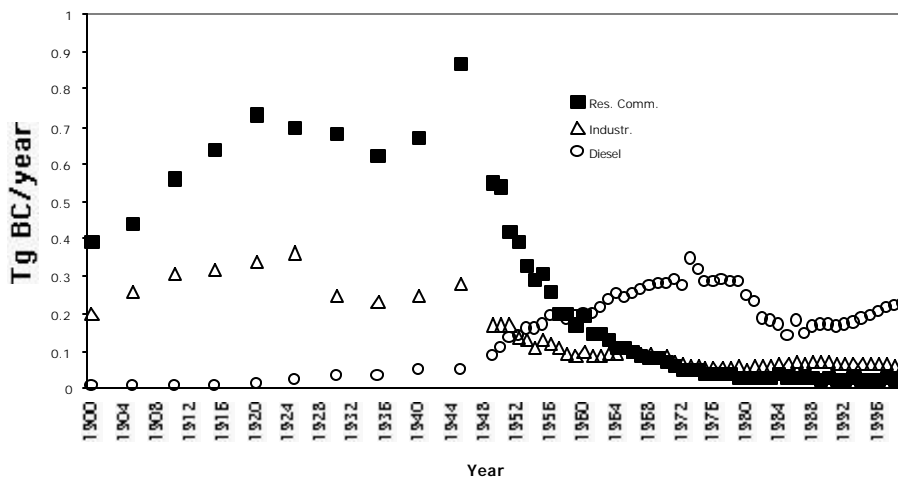
We illustrate our approach by considering two typical regions: United States and China. The former is an example of the industrialized West (or North), which produced more “absorbing” aerosols in the early part of the 20<sup>th</sup> century, and more “reflecting” aerosols during the later part. The latter is an example of the developing nations in the East (or South), which became a large source of light-absorbing BC aerosols during the second half of the century.

Fig. 1 shows the time series of the U.S. coal (in domestic/commercial, industrial and electric utility sectors) and diesel fuel consumption [Data from Gschwandtner *et al.*, 1986, U.S. DOE and EPA]. These data illustrate drastic changes in coal utilization from 1900 to present. During the first half of the century most coal was consumed by domestic/commercial and industrial sectors. During the second half, however, electric utilities consumed most of the coal. Because domestic stoves and industrial boilers are copious sources of BC and power generation is not, it is intuitively clear that the BC emissions from coal burning must have been largest in the first half of the century.

Time series of annual BC emissions estimated from different fuel utilization sectors are shown in Fig. 2. Large increase in BC emissions in the U.S. occurred in the first half of the century because of the prevailing coal use by the heavily polluting domestic and industrial sectors. The BC emissions reached a maximum in the 1945 —



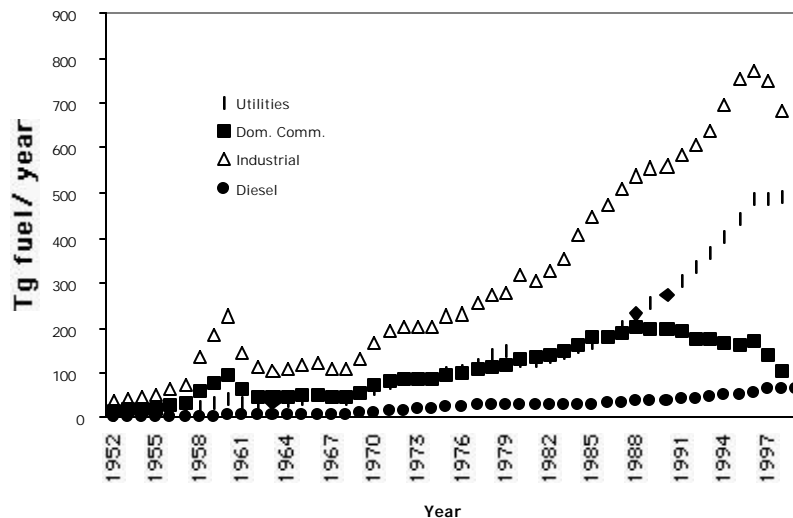
**Figure 1.** Coal consumption by sector and diesel fuel consumption for the United States in Tg per year.



**Figure 2.** BC emissions from coal and diesel in Tg per year for the United states.

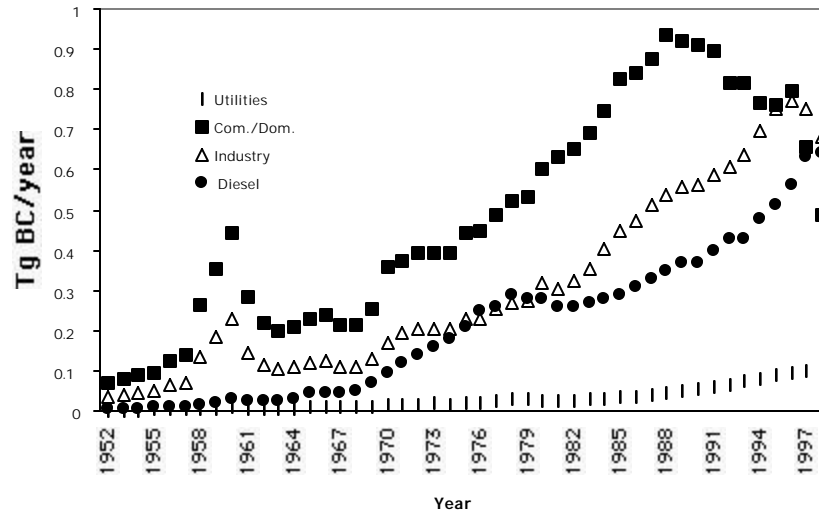
1950 period but steadily declining afterwards because of the shift in coal utilization to efficient coal combustion in electric utilities. Although the consumption of diesel fuel has rapidly increased from about 1950, BC emissions from diesel engines did not follow the trend in diesel fuel consumption because of the gradual improvements in engine technology that resulted in lowering BC emissions.

Fuel consumption by different sectors in China [China Statistical Yearbook, Beijing, 2000] is shown in Fig.3. Throughout the 1950 — 1998 period most coal in China was consumed by industrial sector, followed by utilities and domestic/commercial sectors. The consumption by the domestic/commercial sector decreased somewhat after 1990 but prior to that comprised an almost constant fraction of total coal consumption. Diesel consumption, although small in comparison with the U.S. shows a noticeable increase. The net result of such fuel utilization is that the BC emissions from China from all sectors monotonically increased in concert with fuel consumption (Fig. 4). This is in sharp contrast to the U.S. where the BC emissions have not followed total coal consumption and have drastically decreased in the same period.



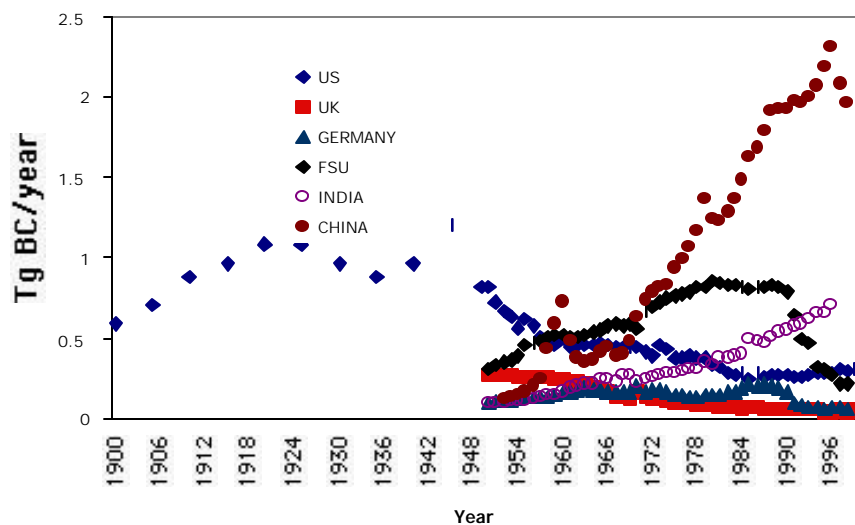
**Figure. 3.** Sector segregated coal and diesel fuel consumption for China.

Time series of BC emissions (from all fuel sectors) for the six countries considered (Fig. 5) show that India and especially China were the largest BC sources in

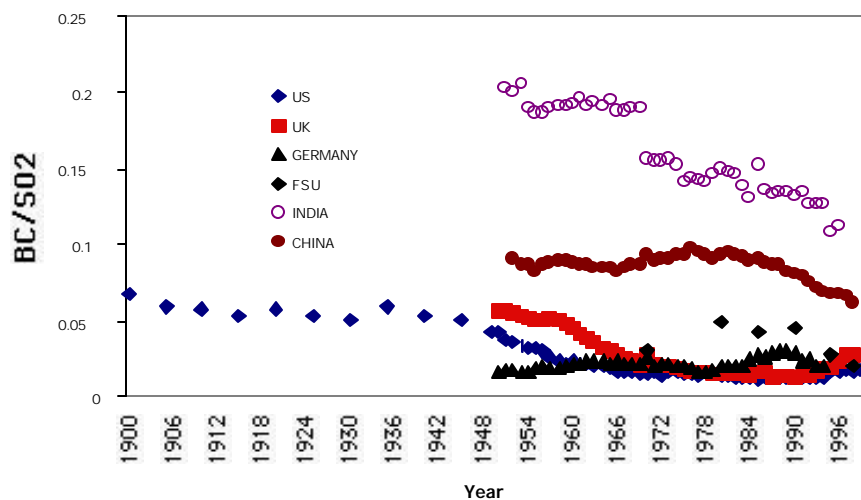


**Figure 4.** BC emissions from coal and diesel fuel in China.

the second half of the past century. (Sector segregated fuel consumption were obtained mostly from data published by UN and IEA.) During the first half BC emissions from the U.S. alone, however, were similar to those from China in about 1980. U.K. BC emissions after 1950 follow similar trend as the U.S. Coal consumption by the industrial and residential/commercial in the U.K. decreased after 1960's resulting in a significant decrease of BC emissions. Although data prior to 1950 are not available for countries other than the U.S., it is reasonable to assume that emissions from European industrial countries before 1950 were also high. Therefore, industrial western countries were the most likely BC sources in that period. BC emissions from Germany (east and west) abruptly decreased after the German reunification because of the closing of old polluting units and a reduction of soft coal use. Diesel fuel consumption, however, increased steadily until 1980 and stayed level afterwards. Because Germany is the largest consumer of diesel fuel in Western Europe the diesel BC emissions comprise a large fraction of total Germany's BC emissions. BC emissions from former Soviet Union steadily increased until the late 1980's and decreased about four-fold after its dissolution.



**Figure 5.** Total BC emissions from various countries in Tg per year.



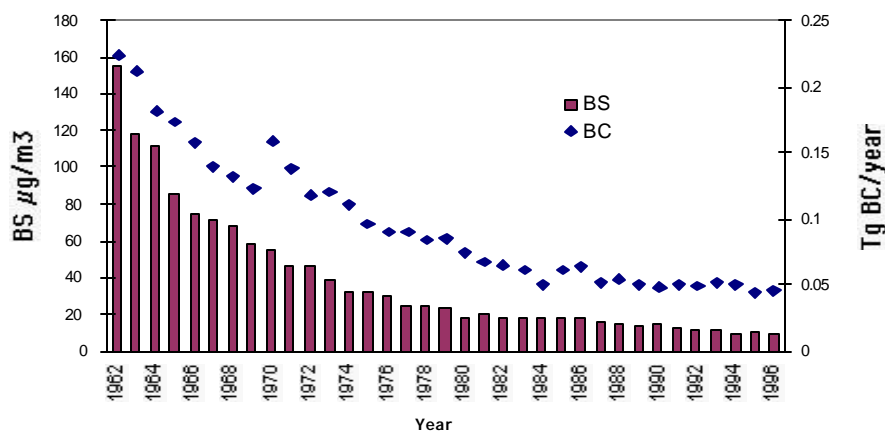
**Figure 6.** Time series of BC/SO<sub>2</sub> ratios for various countries.

Differences in temporal changes of BC and SO<sub>2</sub> emissions are evident from time series of BC/SO<sub>2</sub> ratios (Fig. 6). These ratios in the U.S. and the U.K. showed a significant decrease during the second half of the century BC/SO<sub>2</sub> emission ratios for China and, especially, India were systematically much higher than for the U.S. and European countries. As SO<sub>4</sub><sup>2-</sup> is the product of SO<sub>2</sub> oxidation, BC/SO<sub>2</sub> emission ratios may likely

be mirrored in the relative concentrations of aerosol BC and  $\text{SO}_4^{2-}$ . Furthermore high and low BC/ $\text{SO}_2$  ratios may be indicative of periods with low and high single scatter albedo.

The approximate validity of our BC time series for the U.S. is consistent with analyses of aerosol samples collected in major U.S. cities in 1950 and 1975. Annual concentrations of organic aerosol mass in 1975 [cited in *Katz*, 1961] were about four times lower than found in the same cities in 1950 [*Shah et al.*, 1984]. Though not measured in 1950, it is reasonable to expect that BC concentrations have also similarly decreased. These changes compare well with the 3.4-fold decrease in estimated BC emissions over the same period. Furthermore, data from Pittsburgh, Pennsylvania, typical of past pollution in U.S. cities, show that the number of dense smoke occurrences (visibility less than 0.4 miles) was largest in the 1940 – 1950 period and immensely decreasing thereafter [*Davidson*, 1979].

A more direct validation of estimated BC trends is supported by the continuous measurements (from 1962) of the "Black Smoke" (BS) in the U.K. [U.K. National Air Quality Archive]. (BS is a measure of the darkness of the filter deposit which is proportional to the BC concentration.) A comparison of estimated BC emissions and measured BS, averaged for the entire U.K., is shown in Fig. 7. As it is seen the trends of BC emissions and BS concentrations are remarkably similar.



**Figure 7.** Comparison of BC emissions and measured BS concentrations

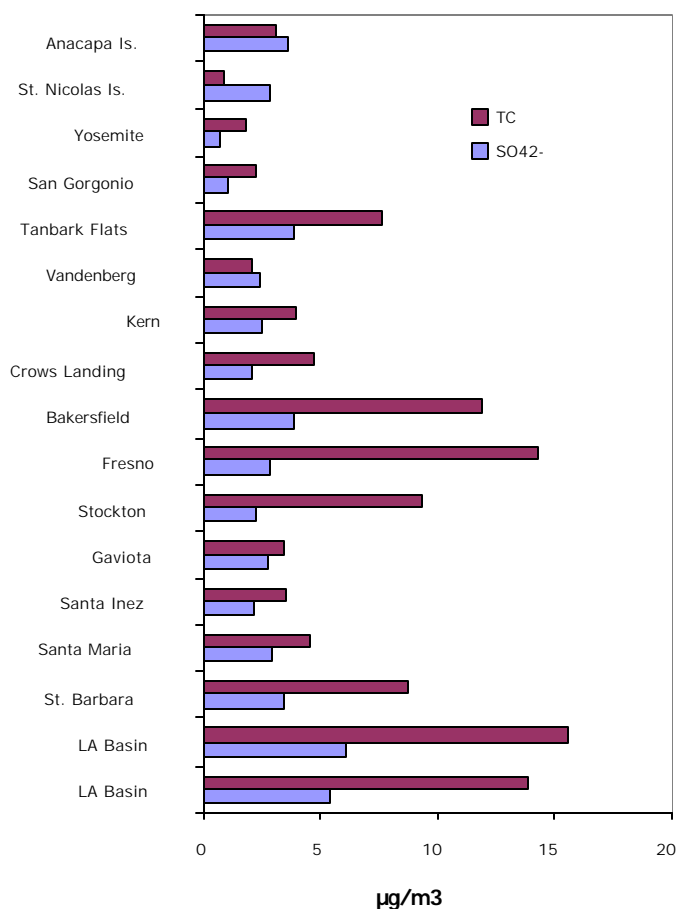


## II. SURVEY OF EXISTING DATA

The data used in this report are a sub-set of a larger data base assembled by examining over 100 journal publications covering approximately the 1980 - 2000 period. The data selected for detailed examination are limited to surface data and pertain to urban (i.e. source regions) and rural locations and that include OC, BC,  $\text{SO}_4^{2-}$  and aerosol mass concentrations. Data from studies were performed over sufficient time periods were emphasized. Furthermore, many data were obtained by different investigators using different methods. Very few, if any, of published data accounted for organic aerosol sampling induced artifacts. To minimize some of the potential problems with data quality we used mainly the data obtained by the same or similar sampling and analytical method.

### **Sulfate vs. carbonaceous aerosols.**

In view of the emphasis on "sulfate alone" by many of the past climate studies it is instructive to compare the mass concentrations of sulfate and carbonaceous aerosol actually measured at different regions. Here we present such comparisons for urban and rural sites in California, Japan and Korea. Annual mass concentrations of  $\text{SO}_4^{2-}$  and total aerosol carbon (TC) measured in California at a number of sites in the period from late 1980's to mid 1990's are shown in Fig. 8. (TC is defined as the sum of OC and BC with concentrations expressed as "C".) As these data show the  $\text{SO}_4^{2-}$  concentrations over most of the state are remarkably uniform. In contrast, the TC concentrations are highly variable being highest in urban locations. TC concentrations, on the average, exceed  $\text{SO}_4^{2-}$  mass concentrations at all urban and rural locations except at coastal locations where  $\text{SO}_4^{2-}$  exceeds TC.



**Figure 8.** Comparison of annual total carbon and sulfate concentrations across California. (Data from *Chow et al.*, 1993, '94, '95, '96; *Malm et al.*, 1994; *Solomon et al.*, 1989.)

The dominant contribution of carbonaceous aerosols at these sites is even more obvious when the organic aerosol mass (OM) is compared with sulfate concentrations. OM accounts for oxygen and hydrogen associated with aerosol organic mass and is commonly assumed to be 1.4 times higher than TC. Table 1 shows the average annual OM/SO<sub>4</sub><sup>2-</sup> and BC/SO<sub>4</sub><sup>2-</sup> ratios for California locations where both OC and BC were measured.

It is evident that OM substantially exceeds the SO<sub>4</sub><sup>2-</sup> concentrations in both urban and rural locations except at the marine site. BC concentrations, however, are about 50%

lower than sulfate concentrations at rural and urban locations except at the three Central Valley cities where BC significantly exceed the sulfate concentrations.

**Table 1.** Comparison of organic mass (OM), BC and sulfate concentrations at urban and rural locations in California

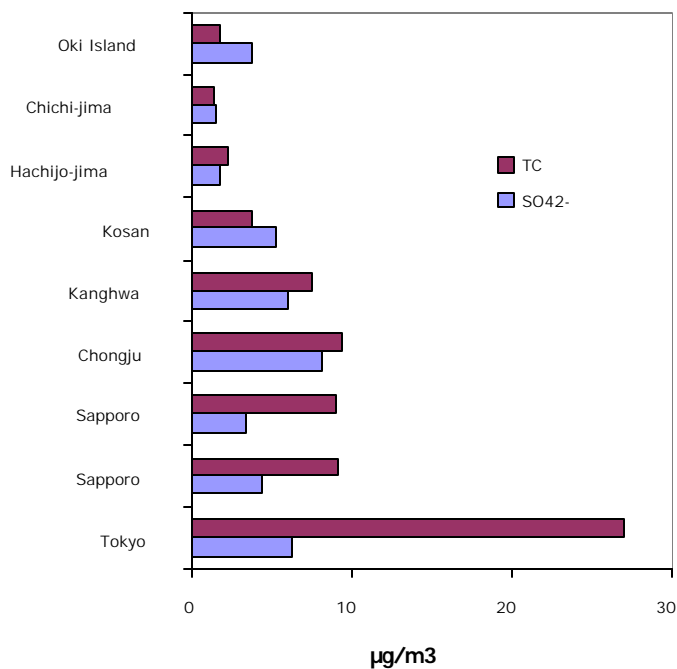
Location, year	OM/SO <sub>4</sub> <sup>2-</sup>	BC/SO <sub>4</sub> <sup>2-</sup>	Sites, (references.)
LA Basin, 1986	2.73	0.59	Urban, (1)
LA Basin, 1987	2.68	0.83	Urban, (2)
Stockton, 1988-89	3.30	1.69	Urban, (3)
Fresno, 1988-89	4.00	2.28	Urban, (3)
Bakersfield, 1988-89	2.33	1.39	Urban, (3)
Crows Landing, 1988-89	2.30	0.74	Rural, (3)
Kern, 1988-89	1.52	0.53	Rural, (3)
Tanbark Flats, 1986	2.30	0.31	Rural, (1)
San Geronio, 1988-91	2.50	0.50	Rural, (5)
Yosemite, 1988-91	3.0	0.43	Rural, (5)
St. Nicolas Is. 1987	0.39	0.04	Marine, (6)

(1) *Solomon et al.* 1989; (2) *Chow et al.*, 1994; (3) *Chow et al.*, 1993; (5) *Malm et al.*, 1994; (6) *Chow et al.* 1996.

Organic matter concentrations exceeded sulfate concentrations in the U.S. national parks in all regions except in the East [*Malm et al.*, 1994]. The OM/SO<sub>4</sub><sup>2-</sup> were about 2.2, 1.2, and 1.7 for the Northwest, Southwest, and Southern California respectively. For the Eastern region, however, the organic to sulfate ratio was about 0.5.

A similar analysis is also possible for a number of sites in Japan and Korea. Mass concentrations of SO<sub>4</sub><sup>2-</sup> and TC measured at those sites are higher than sulfate at all urban locations and lower, but still comparable, at the island sites (Fig. 9). OM/SO<sub>4</sub><sup>2-</sup> and BC/SO<sub>4</sub><sup>2-</sup> ratios obtained at locations where BC and OC have been measured are presented in Table 2. Average OM/SO<sub>4</sub><sup>2-</sup> ratio for all locations is 1.1. BC/SO<sub>4</sub><sup>2-</sup> ratios, however, exhibit a much larger variability than either TC/SO<sub>4</sub><sup>2-</sup> or OM/SO<sub>4</sub><sup>2-</sup>. The lack of

correspondence between  $OM/SO_4^{2-}$  (or  $TC/SO_4^{2-}$ ) and  $BC/SO_4^{2-}$  ratios is the consequence of differences in relative BC and OC concentrations (Table 3.)



**Figure 9.** Comparison of annual TC and  $SO_4^{2-}$  concentrations in Japan and Korea (data from *Ohta and Okita*, 1984, 1990; *Ohta et al.*, 1998; *Okamoto et al.*, 1990; *Lee and Kang*, 2001; *Lee et al.*, 2001; *Mukai et al.*, 1990)

**Table 2.** Comparison of organic mass (OM), BC and sulfate concentrations at urban and rural locations in Japan and Korea

Location, year	$OM/SO_4^{2-}$	$BC/SO_4^{2-}$	Site, (references)
Sapporo, 1982	1.37	1.17	Urban, (1)
Sapporo, 1992	1.18	1.25	Urban, (2)
Chongju, 1995-96	0.84	0.54	Urban, (3)
Kanghwa, 1996-97	1.50	0.16	Urban, (4)
Kosan, 1996-97	0.90	0.07	Island, (4)
Hachijo-jima, 1981	1.05	0.53	Island, (5)
Chichi-jima, 1981	0.64	0.40	Island, (5)

1) *Ohta and Okita*, 1990; (2) *Ohta et al.*, 1998; (3) *Lee and Kang*, 2001; (4) *Lee et al.*, 2001; (5) *Ohta and Okita*, 1984.

### Regional characteristics of OC/BC ratios and BC mass fractions

BC/OC ratios show a marked regional and temporal differences, indicative of the in the chemical composition of the carbonaceous aerosol material (Table 3). BC/OC 1 are observed in urban sites in Japan and Korea. These ratios, however, are about 0.4 in urban Europe. In California BC/OC ratios in Central Valley cities are about 2.5 times greater than in Los Angeles Basin. Generally, these ratios are much higher in source dominated urban than in rural regions. An exception is Japan where BC/OC in urban and island locations are similar.

**Table 3.** Average BC/OC and BC/Mass ratios characteristic of different regions and location types.

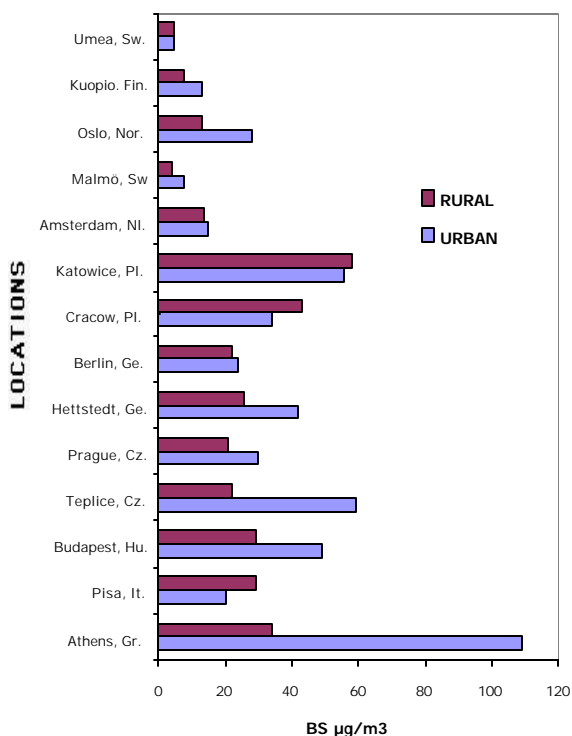
Region	Location	BC/OC	BC/Mass
JAPAN <sup>a</sup>	Urban	0.92	0.17
	Coastal (islands)	0.80	NA
KOREA <sup>b</sup>	Urban	0.82	0.1
	Rural	0.14	0.03
PORTUGAL <sup>c</sup>	Urban	0.43	NA
	Rural	0.25	NA
EUROPE, various locations <sup>d</sup>	Urban	0.43	0.14
	Rural	0.13	0.03
CALIFORNIA <sup>e</sup>	Urban, Central Valley	0.78	0.14
	Urban, L.A. Basin	0.30	0.06
	Rural	0.40	0.07
	National Parks	0.22	0.04

<sup>a</sup> Hayasaka *et al.*, 1992; Kaneyasu *et al.*, 1995; Kadowaki, 1990; references in Table 2. <sup>b</sup> references in Table 2. <sup>c</sup> Castro *et al.*, 1999; Pio *et al.*, 1996. <sup>d</sup> Kendall *et al.*, 2001; Müller, 1999; Castro *et al.*, 1999; Heinzenberg *et al.*, 1998; Cachier *et al.*, 1989; Valaoras *et al.*, 1988; Hitzenberger *et al.*, 1999; Zappoli *et al.*, 1999. <sup>e</sup> References in Table 1.

Besides regional differences, the BC/OC and BC/Mass ratios show prominent seasonal differences. For example, the Los Angeles Basin BC/OC ratio in winter and summer are 0.38 and 0.25 respectively. BC/Mass ratios in the same region are respectively 0.08 and 0.04 in winter and summer. Seasonal variations up to factor of two in the BC/OC and BC/Mass ratios are evident in the data from in Sapporo, Japan in the 1982 - 83 period [Ohta and Okita, 1990].

### Spatial distribution of BC over Europe

Figures 8 and 9 above illustrate the spatial distribution of TC in California and Japan. Spatial distribution of "Black Smoke" (BS), a measure of BC, across the North - South transect of Europe [Hoek *et al.*, 1997] is shown in Fig. 10. These measurements were performed almost simultaneously in both urban and non-urban locations during the 1993-94 winter. Non uniformity in BS concentrations are obvious.



**Figure 10.** Distribution of "Black Smoke" concentrations in urban and rural (non-urban) sites across Europe (data from Hoek *et al.*, 1997).

### **III. SOURCES OF CARBONACEOUS AEROSOLS OVER INDIAN OCEAN**

A complete account of this work is in *Novakov et al.*, 2000, the abstract of which is reproduced below.

Abstract. We present an analysis of the carbon, potassium and sulfate content of the extensive aerosol haze layer observed over the tropical Indian Ocean during the Indian Ocean Experiment (INDOEX). The black carbon (BC) content of the haze is as high as 17% of the total fine particle mass (the sum of carbonaceous and soluble ionic aerosol components) which results in significant solar absorption. The ratio of black carbon to organic carbon (OC) (over the Arabian Sea and equatorial Indian Ocean) was a factor of 5 to 10 times larger than expected for biomass burning. This ratio was closer to values measured downwind of industrialized regions in Japan and Western Europe. These results indicate that fossil fuel combustion is the major source of carbonaceous aerosols, including black carbon during the events considered. If the data set analyzed here is representative of the entire INDOEX study then fossil fuel emissions from South Asia must have similarly contributed to aerosols over the whole study region. The INDOEX ratios are substantially different from those reported for some source regions of South Asia, thus raising the possibility that changes in composition of carbonaceous aerosol may occur during transport.

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