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Journal

Atmospheric Environment, 38(25)

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Publication Date

2004-04-22

LBNL-54109

**Black Carbon Emissions in the United Kingdom during the past four decades:
An empirical analysis**

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Abstract – We use data from a unique 40-year record of 150 urban and rural stations in the “Black Smoke and SO₂ Network” in Great Britain to infer information about sources of atmospheric black carbon (BC). The data show a rapid decline of ambient atmospheric BC between 1962 and the early 1990s that exceeds the decline in official estimates of BC emissions based only on amount of fuel use and mostly fixed emission factors. This provides empirical confirmation of the existence and large impact of a time-dependent “technology factor” that must multiply the rate of fossil fuel use. Current ambient BC amounts in Great Britain comparable to those in western and central Europe, with diesel engines being the principal present source. From comparison of BC and SO₂ data we infer that current BC emission inventories understate true emissions in the U.K. by about a factor of two. The results imply that there is the potential for improved technology to achieve large reduction of global ambient BC. There is a need for comparable monitoring of BC in other countries.

Keywords: Black carbon, black smoke, emission trends, sources, technology factor,

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1. Introduction

Soot is among the most important of air pollutants. Soot is a particle-phase product of incomplete combustion of carbon containing fuels. Its main components are black carbon (BC) and organic carbon (OC). Most studies of the human health effects of air pollution have not been composition specific, but there is evidence that tiny soot particles, which usually include toxic organic carbon and metals, are carcinogenic and among the most harmful pollutants (Künzli et al., 2000).

Black carbon (BC), the principal light-absorbing component of soot, arguably rivals methane for being the second largest contributor to global warming, causing a total forcing, including its indirect effects on snow and cloud albedos, of $0.8 \pm 0.4 \text{ W/m}^2$ (Hansen and Nazarenko, 2003). BC may affect regional climate; for example, it has been suggested that the heavy concentration of soot over China and India may be responsible for a trend toward increased flooding in the south and drought in the north (Menon et al., 2002). Soot reduces atmospheric transparency and visibility, by enough in India and China to reduce agricultural productivity an estimated 10-20% (Chameides et al., 1999) with additional productivity loss from soot deposited on plant leaves (Bergin et al., 2001). Soot is also esthetically displeasing as it is responsible for the brown appearance of urban hazes and soiling of buildings.

Soot in general and BC in particular have not received sufficient attention in measurements and analyses to define well their role in global climate change. The amount of BC in the atmosphere today is very uncertain. Sato et al. (2003) argued, from analysis of sunphotometer measurements at about 100 sites around the world, that global aerosol models understate black carbon absorption by a factor of 2-4, although they suggest that part of the observed absorption is due to enhancement from internal mixing (Chylek, 1988; Jacobson, 2000), rather than from underestimate of BC mass. Knowing the variation of BC during the past century is needed to interpret climate change. Thus data that shed light on the amount of BC in the air today and historical changes of BC amount would be valuable.

BC is a product of incomplete combustion. Unlike SO_2 emissions, which principally depend on the mass of fuel burned and its sulfur content, BC emissions are governed by both the fuel consumed and combustion technology. For example, the BC emission factor (emitted BC mass per unit mass of fuel) for coal combustion in inefficient household stoves can be orders of

magnitude greater than the emission factor for efficient coal burning in large power plants. Consequently, the aerosol optical properties (light absorption and scattering) depend on the relative contributions of emissions from inefficient and efficient fuel utilization sectors.

During the past century coal utilization in industrialized countries has shifted from residential, commercial and industrial sectors to the electric power generation sector. Since BC emissions from the former are higher than from the latter, BC emissions in these countries decreased as coal use by the non-power generation sectors decreased. In most industrializing countries, however, fossil fuel emissions increased more nearly in concert with fuel consumption because the proportion of fuel use by different sectors did not change appreciably. Novakov et al. (2003) estimated effects of changes in fuel use on BC emissions during the past century using sector-segregated fossil fuel consumption data and published emission factors.

Here we present empirical evidence for the effect of changing fuel utilization on measured BC concentrations, compare these results with expectations based on emission inventories, and discuss potential ramifications. Our analysis employs data generated by the "Black Smoke and SO₂ Network" that has been operating continuously since 1962 at more than 150 urban, peri-urban and rural monitoring sites throughout much of the U.K. (Loader et al., 2003). These data, consisting of both measured concentration and estimated BC emissions, are notable in the sense that they cover four decades characterized by large changes in fossil fuel utilization.

Section 2 specifies the sources of the data that we employ. Section 3 presents the data with a qualitative discussion of its temporal variations. In Section 4 we obtain more quantitative inferences from the relative amounts and changes of BC and SO₂. We discuss implications of the analyses in Section 5.

2. Data sources

Measurement sites in the Black Smoke (BS) and SO₂ Network reporting in 2001 are shown in Fig. 1, which illustrates the mean SO₂ measured for that annual period (Loader et al. 2003). The majority of sites are urban or peri-urban, although several are in rural areas. Most of our analysis refers to annual BS and SO₂ concentrations averaged over all monitoring sites, and to emission inventories of these species for the entire U.K. (sources of data are summarized in Table 1). Note that we used two BS data sets. An older one covering the 1962-1970 period and a recent one for 1970-1999. The annual averaging reduces variability associated with weather

fluctuations and noise in the data. The national average further reduces noise, but we present examples of local data for an urban site and a rural site to show the effect of distance from major sources.

BS is determined by an optical reflectance method that measures the darkness of stain on white filter paper through which the particle-laden air is drawn (Bailey and Clayton, 1982). The measured concentrations of light-absorbing aerosols in units of $\mu\text{g (BS) m}^{-3}$. In the following we convert these units to more commonly used units of $\mu\text{g (BC) m}^{-3}$ via the relationship $\text{BC} = 0.23 \text{ BS}$ obtained by (Erdman et al., 1993). We note that a different relationship, $\text{BC} = 0.13 \text{ BS}$, was obtained in an older study (Edwards et al., 1983). Our conclusions, however, would not change if only BS was used.

We express SO_2 concentrations and emissions in $\mu\text{g (S) m}^{-3}$ and Tg (S) yr^{-1} , respectively. We denote SO_2 emissions as S , ambient gaseous SO_2 as S_g , and total ambient sulfur S_t as the sum of S_g and particle phase (sulfate) S_p . BC values are derived from BS data as discussed above.

3. Interpretation of BC historical trends

Our analysis is based on the expectation that average long-term ambient mass concentrations of BC and SO_2 , in a source-dominated region such as the U.K., should be proportional to the source strengths of these species, if the monitoring sites are affected by all UK sources. This assertion is supported by Handley and Tuomi (2001) who have shown that the gradients in SO_2 concentration trends do not differ greatly at site located at different distances from the main U.K. sources. Furthermore, network averaged annual BC concentrations are highly correlated with BC concentrations at Didcot ($R^2 = 0.851$) and Manchester ($R^2 = 0.957$). Also the trends in BC gradients at the two sites are quite similar to the trend in network averaged BC. The temporal and spatial variations of emissions and ambient concentrations, discussed below, are consistent with this expectation. Although our interest is in BC, we present SO_2 measurements in this section as well, because we use the SO_2 amounts in section 4 to constrain estimates of the BC source strength.

Time series of BC and S annual emissions in the U.K., as reported by the data sources in Table 1, are shown in Fig. 2a. The annual means of the measured BC and S_g ambient concentrations, averaged over the Black Smoke and SO_2 Network sites, are shown in Fig. 2b. The emissions and ambient concentrations of both species decrease almost monotonically

throughout the period of record. BC emission estimates in Fig. 2 (from U.K. National Atmospheric Emission Inventory – NAEI) are based on fuel use statistics with the assumption that BC emission factors (per unit mass of fuel) are constant throughout the period for each fuel use sector (power plant, industry, residential and diesel). Diesel emission factors in the period 1992-1999 assumed to decrease because of improving engine technologies [<http://www.aeat.co.uk/netcen/airqual/statbase/emissions/bs-99.html>].

Emissions and ambient amounts of both BC and S_g declined dramatically over the past four decades in the U.K. Note that the fractional decline of BC was more rapid early in the period, while the fractional decline of S_g was more rapid late in the period. This is consistent with the emphasis on soot reduction that followed devastating "London smog" problems of the 1950s and the emphasis on reducing acid rain in recent decades.

Fig. 3a shows the BC ambient amount as a function of BC emissions, while Fig. 3b is the analogous graph for S_g . In both graphs the plotted points go from the earliest date (1962) in the upper right to the most recent date in the lower left. We note that S emissions are about 10 times greater than BC emissions, according to the emission inventories (Fig. 2a). Yet Fig. 2b suggests that measured atmospheric amounts of gaseous sulfur (S_g) are only about three times larger than measured ambient BC. This inconsistency suggests that BC emissions may be underestimated. We explore this quantitatively in Section 4, where we consider other factors that may affect interpretation of the measurements

In Fig. 3 it is apparent that both BC and S_g decrease more rapidly than linearly with emissions. Reduction of S_g is expected because of decreasing coal use and possibly reduced sulfur content of fuels. The reduction of BC emissions and concentrations is caused principally by decreased use of coal. Although BC emissions from coal were steadily declining, leveling off of BC concentrations in recent years has been caused by increasing use of diesel fuel, as illustrated below.

Interpretation of changing BC ambient amount is aided by examination of the temporal change of emissions as a function of sector and the geographical variation of emissions and ambient amounts. Fig. 4 shows fuel consumption in the U.K. in Tg of fuel per year for four sectors, "power plant", "total industry" and "residential" for coal and diesel fuel, according to the International Energy Agency (IEA, 2000). Sector-segregated BC emissions for the U.K. show that the "industry" and "residential" coal sectors together with diesel use account for almost 90%

of British BC emissions (<http://www.aeat.co.uk/netcen/airqual/statbase/emissions/bs-99.html>). This is because BC emission from power plants is assumed to be negligible, while emission factors for industrial and residential coal use are estimated as 0.23 g/kg and 9.2 g/kg, respectively, and as 1.9 g/kg for diesel (Table 2). Diesel fuel has a moderate estimated emission factor and contributed relatively little to BC emissions until the past two decades, when use rose while coal consumption continued to decline. The primary source of BC through most of the period of record is thus residential and industrial coal burning. The decline in BC ambient amounts in Fig. 3a, more rapid than the decline of fuel amount, must reflect technology change in the dominant residential sector, which is consistent with a known trend from use of individual stoves to less polluting central heating. We note that the primary source of atmospheric BC, or soot, in Great Britain today is diesel fuel, which is very unlike the situation in prior decades when coal burning was the primary source of soot. The long range transport from the European continent, however, although undoubtedly present could be ignored for the purpose of this paper. For example Derwent et al.(2001), concluded that transported mean BC concentrations transported from Europe during 1995-1998 are $0.69 \mu\text{g m}^{-3}$. This should be compared with a network average BC concentration of $2.2 \mu\text{g m}^{-3}$ the 1995-1998 period (or 3 times higher).

Fig. 5 compares ambient BC concentrations measured at urban (Manchester) and rural (Didcot) sites, as a function of national BC emissions (data source in Table 1). The urban BC amount is not only larger, it also decreases more rapidly with time. The urban BC concentration decreases more steeply than linearly with estimated emissions, when, as in Fig. 3, there is essentially no “technology factor” correction included in the emissions estimate. The ambient BC amount in Manchester, dominated by local sources, is especially large in winter, i.e., during the heating season. These facts support the conclusion that BC emissions have decreased because of the decrease in coal consumption and because the technology factor in residential sector caused a decrease of the BC emission factor over time. In the rural Didcot location, distant from large sources, the BC amount is not only smaller, but it also decreases more slowly as a function of reported national emissions. It is possible that the rural BC amount is influenced more by the diesel source, which has increased over time, or other sources that have not decreased as rapidly as residential and industrial coal burning.

Ambient annual BC concentrations in the 1998-2000 period averaged about $1.8 \mu\text{g m}^{-3}$. This value is comparable to the range $0.6 \mu\text{g m}^{-3}$ to $1.6 \mu\text{g m}^{-3}$ recently measured in rural

Hungary and Portugal (Zappoli et al., 1999; Castro et al., 1999). (Note that the U.K. concentrations are derived using $BC = 0.23 BS$; using the factor 0.13 reported by Edwards et al. (1983) would lower the ambient BC to about $1 \mu\text{g m}^{-3}$.) Obviously, the tendency of ambient BC amount to level out in recent years should not be interpreted as an indication that BC amounts in Great Britain are approaching a clean atmospheric “background” level. Even at the rural (Didcot) site the BC amount is $\sim 1000 \text{ ng/m}^3$, which is typical of moderate to heavily polluted regions in middle latitudes of the Northern Hemisphere and about twice as great as the typical BC amount in the well-studied Arctic Haze episodes in the 1980s (Hansen and Novakov, 1989). Although the “black smoke” record is not the most precise measurement of BC, the measured BC amount clearly falls in the category of moderately polluted continental air.

The BC data by themselves illustrate that the large decrease in ambient BC amounts, which has occurred especially in urban areas, is a product of both changing fuel use and combustion technology. We suggest below that more quantitative statements can be made by comparison of the BC and SO_2 records, somewhat analogous to the comparison of BC and CO data by Dickerson et al. (2002). Specifically, in the following section we use the ratio of BC to SO_2 to evaluate the emission factor for BC, which is much more poorly known than the emission factor for SO_2 .

4. Evaluation of BC emission factor

BC emissions to the atmosphere are highly uncertain. BC emissions are estimated from fuel consumption data and assumed sector-specific emission factors, but emission factors are not well measured, often being uncertain by a factor of two or even more (Bond et al., submitted to *J. Geophys. Res.*, 2003; Cooke et al., 1999). It has been argued that global BC distributions derived from typical emission factors yield much less absorption of sunlight than observed by a widespread network of sun photometers (Sato et al., 2003), suggesting that either BC emission factors are underestimated or there are additional BC sources not included in current aerosol models. However, Bond et al. (submitted to *J. Geophys. Res.*, 2003) argue that reexamination of emission factors suggests that the emission factors are generally overestimated, not underestimated. Thus there is a need for an alternative evaluation of BC emission factors.

The Black Smoke and SO_2 Network, with its simultaneously measured BC and SO_2 measurements, provides an opportunity for an indirect assessment of the mean BC emission

factor in Great Britain. This assessment depends on the assumption that uncontrolled SO₂ emissions are relatively well known, because they depend principally on amount of fuel consumed and the sulfur content of fuels. A complication is caused by the fact that the Black Smoke and SO₂ Network measures only SO₂ gas (S_g), while the sulfur produced in burning of fossil fuels resides in both S_g and S_p (sulfate particles). We handle this uncertainty by considering an appropriate range for the uncertain sulfate fraction of total sulfur.

Fig. 6a plots BC emissions vs. S emissions, with both of these based on the estimates from the sources specified in Table 1. Fig. 6b similarly compares the measured ambient amounts of BC and S_g averaged over the Black Smoke and SO₂ Network. The BC and S_g ambient amounts are well correlated, with both of them decreasing in Great Britain over the entire period, but their relationship changed in about 1975. Between 1962 and 1975 BC decreased by a factor 6.5 while S_g decreased by a factor 3.1. Between 1975 and 2000 BC decreased by a factor 2.3 and S_g decreased by a factor 3.8. This change in BC vs. SO₂ coincides with the change in the fraction of non-power coal use (Fig. 4). For example, in 1963 coal consumed by the residential sector accounted for 65% of coal used by the electric power sector. By 1975 this fraction had decreased to 14%, and it decreased further through the remainder of the period considered.

The rapid BC reduction prior to 1975 is related to the UK clean air act of 1958 with consequent emphasis on reducing coal smoke emissions from domestic and industrial sectors. Since about 1975 there has been more emphasis on reducing acid rain and thus on the sulfur content of fuels. BC and S_g both decline during both periods as most actions to reduce one constituent also reduce the other, but there seems to have been a detectable change of emphasis in the late 1970s.

Fig. 7a shows the ratio of emissions, BC/S, in the U.K. National Atmospheric Emission Inventory (NAEI), and the ratio of ambient atmospheric amounts, BC/ S_g , measured by the Black Smoke and SO₂ Network (Fig. 7b) as a function of time. The ratio for the ambient atmospheric amounts is about three times larger than the ratio of estimated emissions. The larger ratio in the atmosphere could be due in part to the fact that a fraction of the emitted sulfur has been oxidized into sulfate particles (S_p). S_p/S_g has been measured at several urban (Nicholson and Davies, 1990) and rural (Irvin et al., 2002) sites in the U.K. as 0.14 ± 0.03 and 0.39 ± 0.18 , respectively. The greater degree of conversion to particles at larger distances from the principal sources is expected.

Fig. 7b shows the range of BC/S with S corrected to include sulfur in particle form for the measured range of S_p/S_g . Most of the stations in the Black Smoke and SO₂ Network are urban and peri-urban, so the 0.14 correction is probably the more appropriate. However, even at the extreme $S_p/S_g = 0.39$ the measured atmospheric BC/S ratio is about double the ratio for estimated emissions. We believe this indicates that BC emissions are underestimated. However, let us consider alternative explanations. One possibility is that the ratio S_p/S_g is even larger than measured at rural sites. However, the required S_p/S_g ratio of 2 or larger is implausible for a small source-dominated region such as Great Britain. Another possibility is that the S emissions escape measurement by being emitted at high altitudes by tall stacks and then wafted out of the country. This explanation, however, would require that escape of S is largest at the beginning of the period of record, when tall stacks were least common.

We conclude that the most likely explanation is that the BC emission inventory underestimates actual BC emissions. This seems to be a feasible explanation given the broad spread in published BC emission factors. Table 2 gives examples of emission factors that have been used in estimating BC emissions in the U.K. Apparently there is a good deal of uncertainty in BC emission factors, as has been emphasized by Cooke et al. (1999) and Bond et al. (submitted to *J. Geophys. Res.*, 2003). For example, it seems possible that the NAEI emission factor for industrial coal use could be substantially too small. There could be other BC sources that are not included in the inventories. However, the fact that BC ambient amounts declined rapidly as the known sources declined implies that the identified sources are the main sources, and it thus suggests that the emission factors have been underestimated.

5. Summary and implications

Aerosol filter samples obtained by the Black Smoke and SO₂ Network and the optical reflectance measurements on the filters yield four-decades-long consistent record on BC and SO₂ concentrations. The measurement record is supplemented by the BC and SO₂ emission inventories covering the same period as the measurements. This combined record contains a valuable history of BC change in the United Kingdom over the past four decades.

The data show that the ambient atmospheric amount of BC in the United Kingdom declined steadily during the past 40 years, although possibly leveling off during the past several years. The decline was more rapid than the decrease in BC-producing fuel use, even with the

assumption that fossil fuel power plants emit negligible BC. This confirms the existence a time-dependent “technology factor” that reduces BC emissions for a given fuel amount. There is still substantial use of coal in the United Kingdom, but BC emissions have been reduced.

We infer from the Black Smoke and SO₂ data that current BC emissions in Great Britain are underestimated by about a factor of two. The underestimation at earlier times was even larger, about a factor of three or more in the 1960s. These values are based on an assumption that the average ratio of S in sulfate particles to S in gaseous form is about 0.25 for the network of measuring stations, which we suggest yields a conservative estimate for the amount of BC. As black smoke measurements are continued, it would be valuable to have them supplemented with high-precision composition-specific analytical studies. This would make the unique long records of the Black Smoke and SO₂ Network even more valuable.

We estimate the current BC amount for the Black Smoke and SO₂ Network locations as approximately 1-2 $\mu\text{g m}^{-3}$. Thus despite a substantial decline over the past four decades, the air in Britain still has moderate to heavy BC pollution. The measured BC has almost leveled off in the past several years, possibly because increasing diesel fuel emissions have compensated for declining coal emissions.

Thus there are two ways to view this record. From one perspective, the large decrease of BC over the first three decades of the record shows the potential for technology to reduce BC from coal emissions. On the other hand, the data also tell a cautionary tale, as they suggest that ambient BC amounts are leveling off and diesel emissions are increasing. Diesel soot emissions include organic carbon and metals (Wang et al., 2003) and have been explicitly implicated in health effects (Künzli et al., 2000). Future trends of BC could go either direction, as there is increasing use of diesel fuel for transportation vehicles, but also the potential for technological improvements that reduce emissions. The actual trend will have implications for climate and other effects, as well as human health.

Despite its practical importance, the amount of BC in the global atmosphere is very uncertain. This gross ignorance needs to be rectified for the sake of understanding the practical effects of soot today as well as for monitoring future changes in atmospheric amounts. We have found that even simple inexpensive measurements, such as the filter samples of the Black Smoke and SO₂ Network in the United Kingdom, yield a valuable record, but modern analytical

techniques could add important composition information. Long-term monitoring with standardized procedures is needed on a more global basis. It is well justified for the sake of providing information that will be needed by decision-makers as the multiple effects of soot are understood better and as it becomes desirable to document progress in reducing atmospheric amounts.

Acknowledgments. This was supported by the Director, Office of Science, Office of Biological and Environmental Research, U.S. Department of Energy and National Aeronautical and Space Administration. We thank A. Loader for information regarding SO₂ measurement methods employed by the network.

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Table 1. Types and sources of data

Data	Period	Reference
SO ₂ and BC* concentration	1962 – 2001	http://www.environment-agency.gov.uk/commodata/103608/fig3_so2_data.txt http://www.airquality.co.uk/archive/index.php
SO ₂ emission	1962-1990	Lefohn et al. (1999)
SO ₂ emission	1990-2000	http://www.defra.gov.uk/environment/statistics/des/index.htm
BC* emissions	1962-1970	Warren Spring Laboratory (1972)
BC* emissions	1970-1999	http://www.aeat.co.uk/netcen/airqual/statbase/emissions/bs-99.html

* BC values derived from BS data (See text).

Table 2.

Comparison of BC emission factors (g/kg) ranges used for U.K. with other compilations.

Sector	NAEI*	Cooke et al. (1999)	Streets et al. (2001)**
Coal, Domestic	9.2 - 5.2	4.6 - 2.8	3.7 - 20
Coal, Industry	0.23 - 0.13	1.0 - 0.2	0.003 - 0.02
Diesel	1.9 - 1.1	10 - 2.0	1.1 - 2.5

* BC values derived from BS data reported by U.K. National Atmospheric Emission Inventory (See text).

** Central and high values

Figure captions

Fig. 1. Measurement sites of the UK Smoke and SO₂ Network in 2001. The concentrations shown correspond to mean net acidity expressed as SO₂ equivalent. From Loader et al., (2003), reproduced with permission.

Fig. 2. (a) BC and S_g emissions for the U.K., (b) BC and S_g mean ambient amounts for the network..

Fig. 3. BC and S_g measured ambient amounts, averaged over the network, as a function of national emissions.

Fig. 4. Fuel consumption in Britain by sector, for sectors [IEA, 2000] that together contribute about 90% of estimated BC emissions.

Fig. 5. Ambient BC amounts measured at urban (Manchester) and rural (Didcot) locations, as a function of national emissions.

Fig. 6. (a) BC emissions vs. S emissions, and (b) BC ambient concentration vs. S_g ambient concentration.

Fig. 7. (a) BC/S ratio in estimated emissions and in ambient atmospheric concentrations measured at the Black Smoke and SO₂ Network, (b) the same as Fig. 7a, but with the ambient S corrected to include sulfur in particle form, for the measured range S_p/S_g = 0.14 to 0.39.

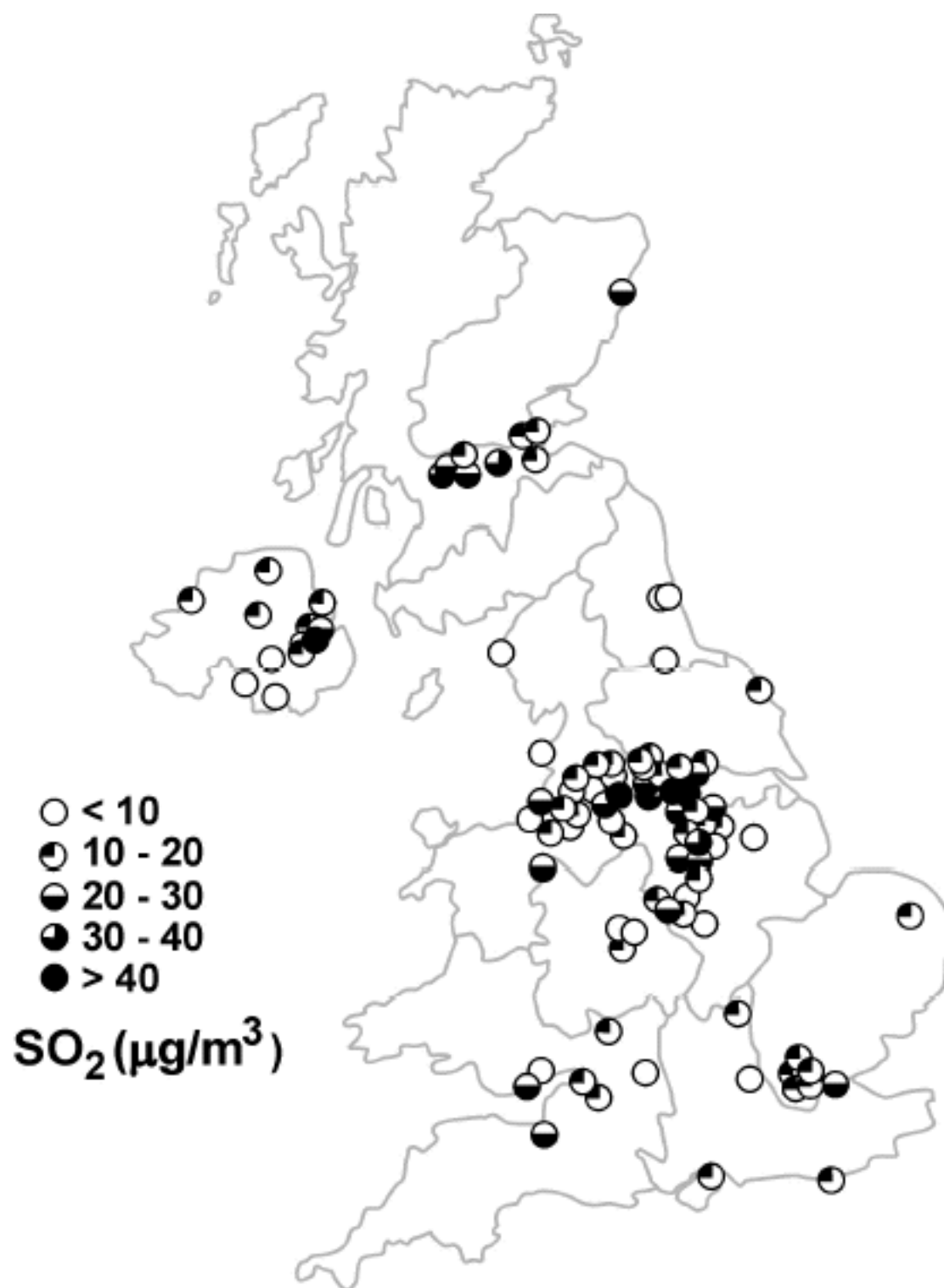


Fig. 1

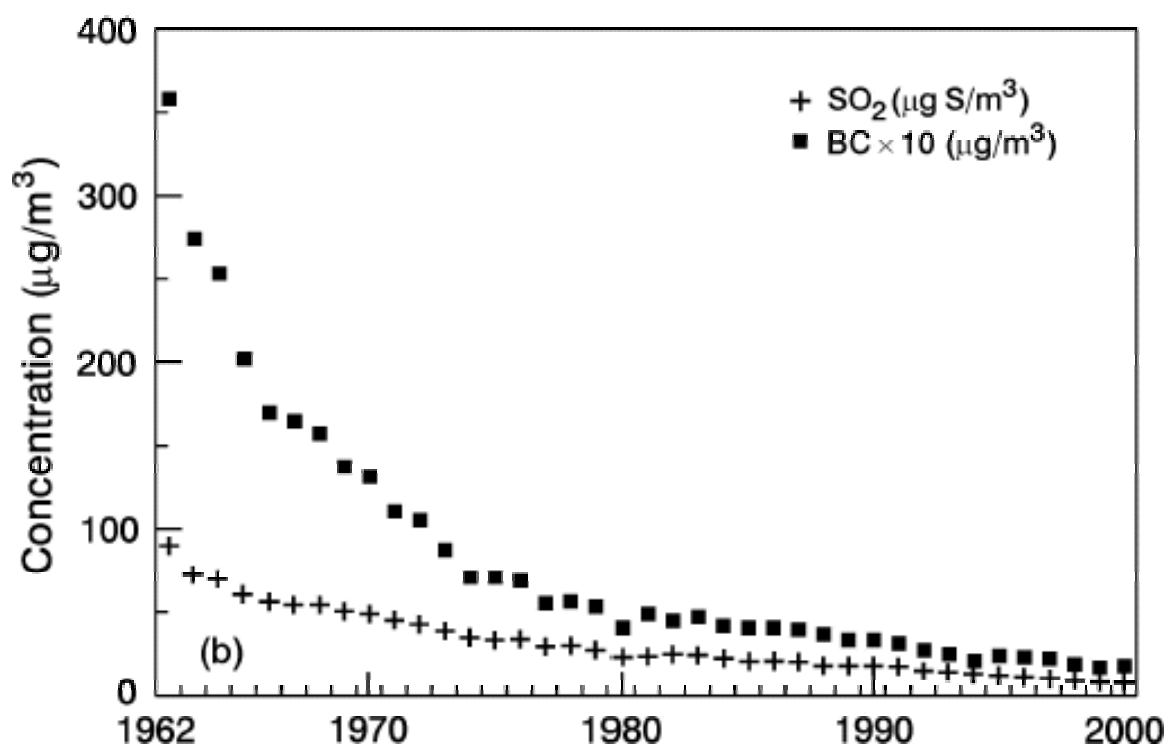
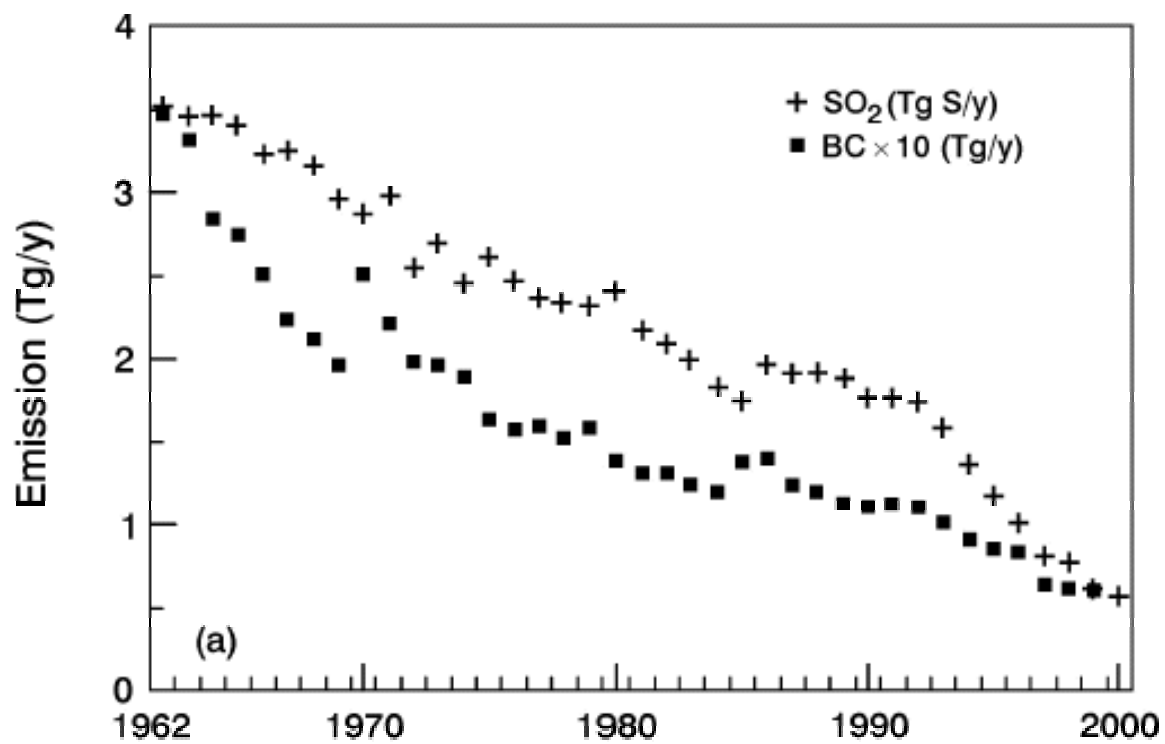


Fig. 2

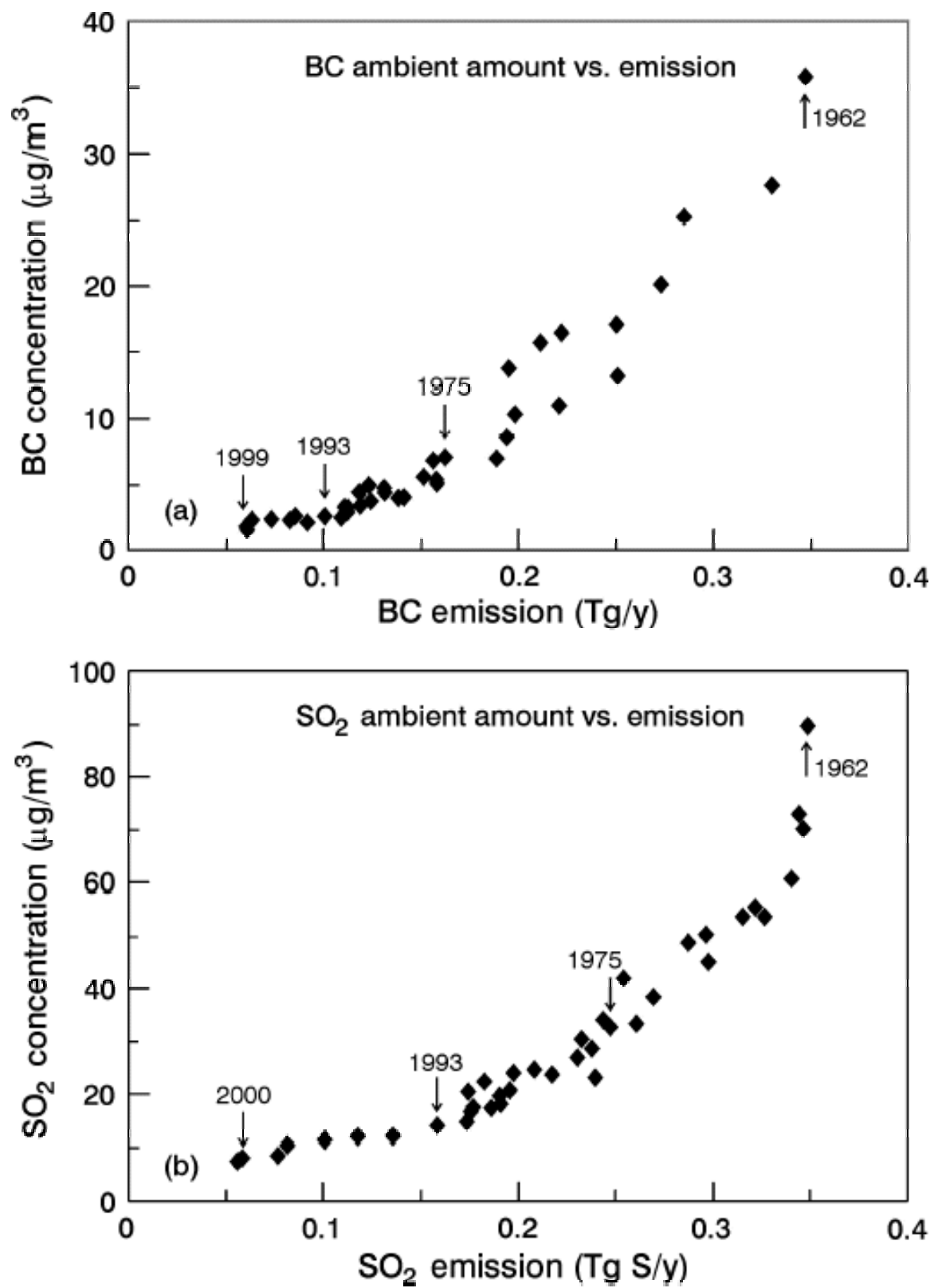


Fig. 3

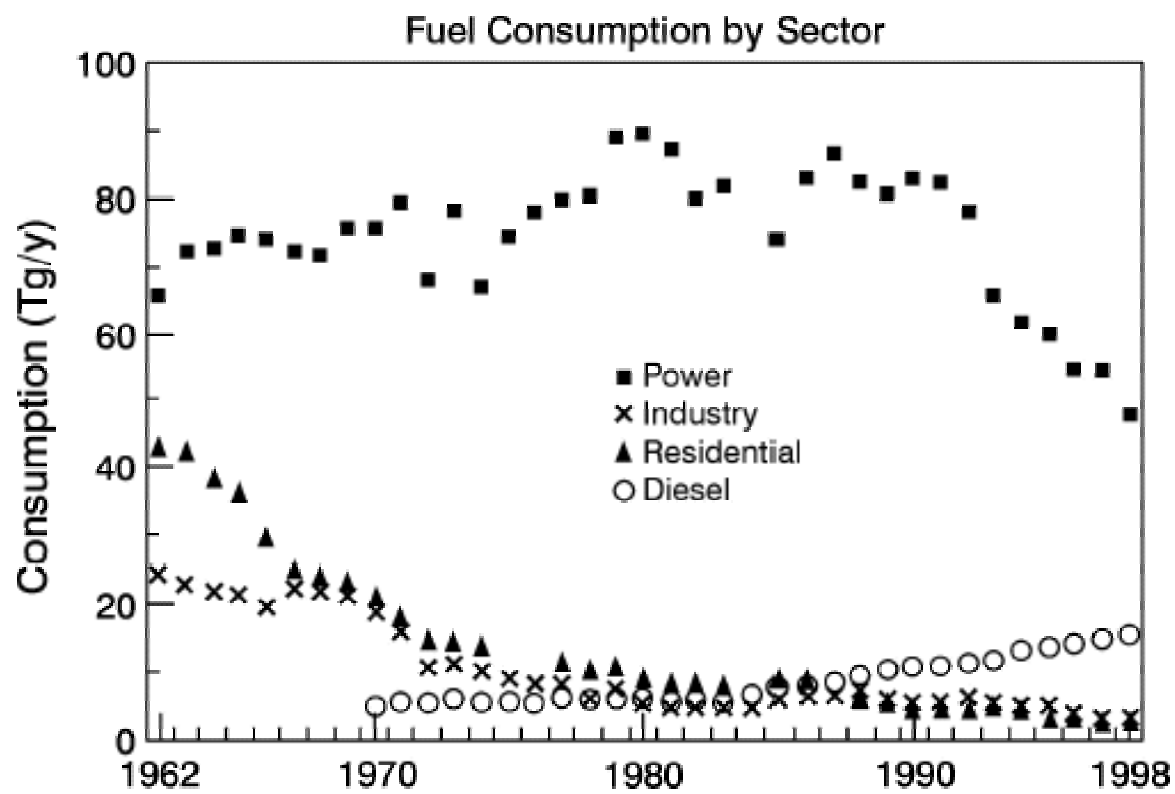
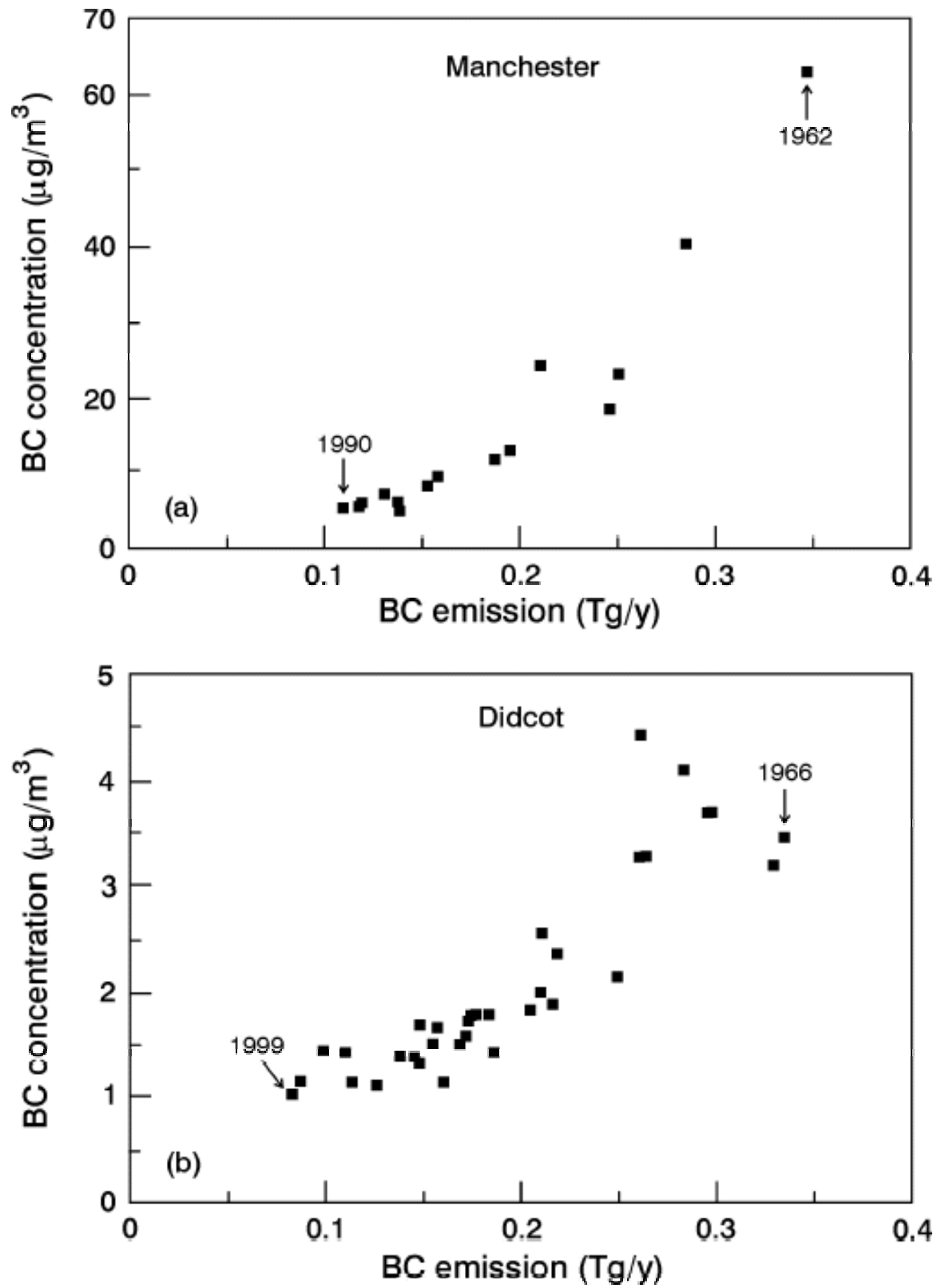


Fig. 4



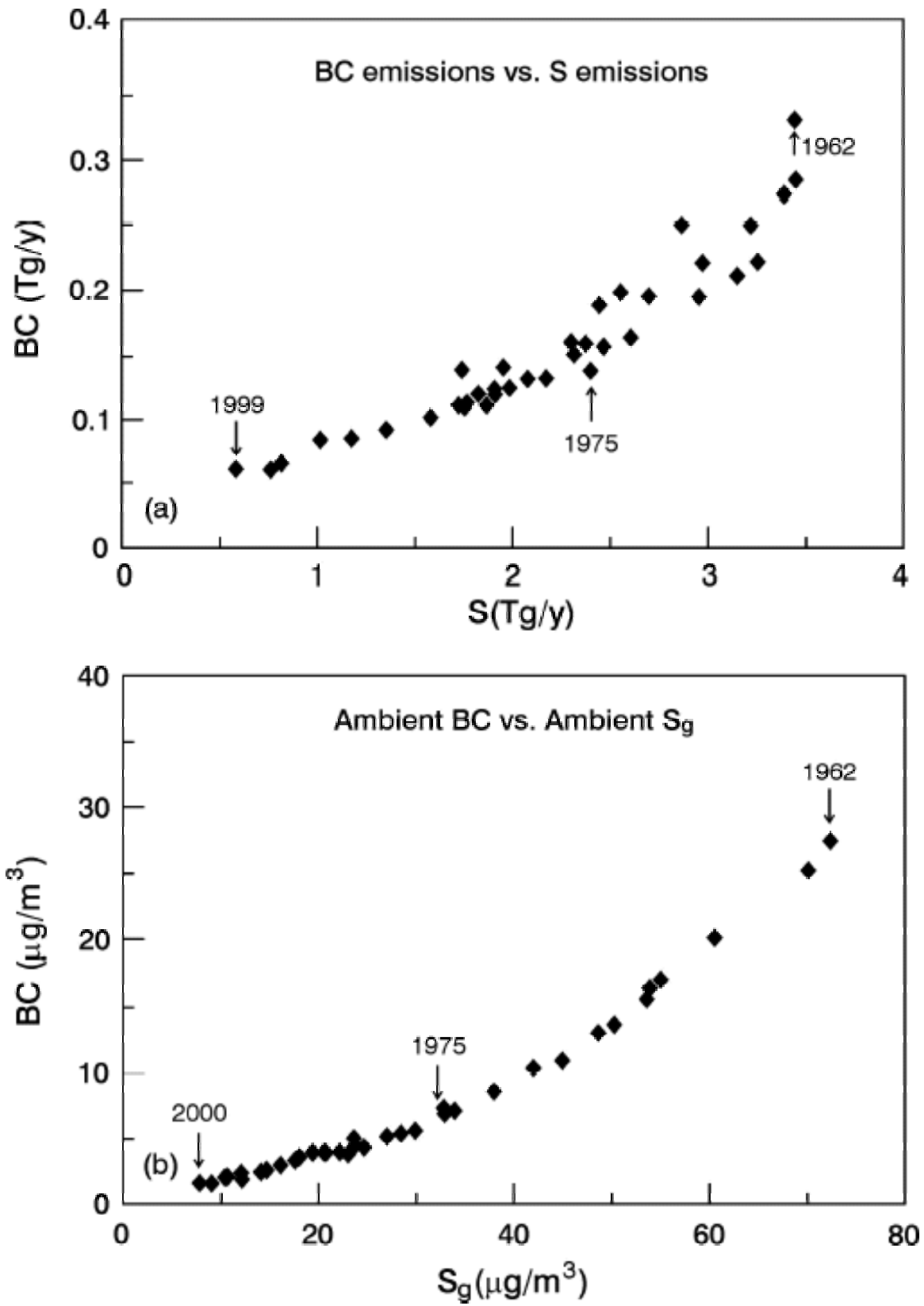


Fig. 6

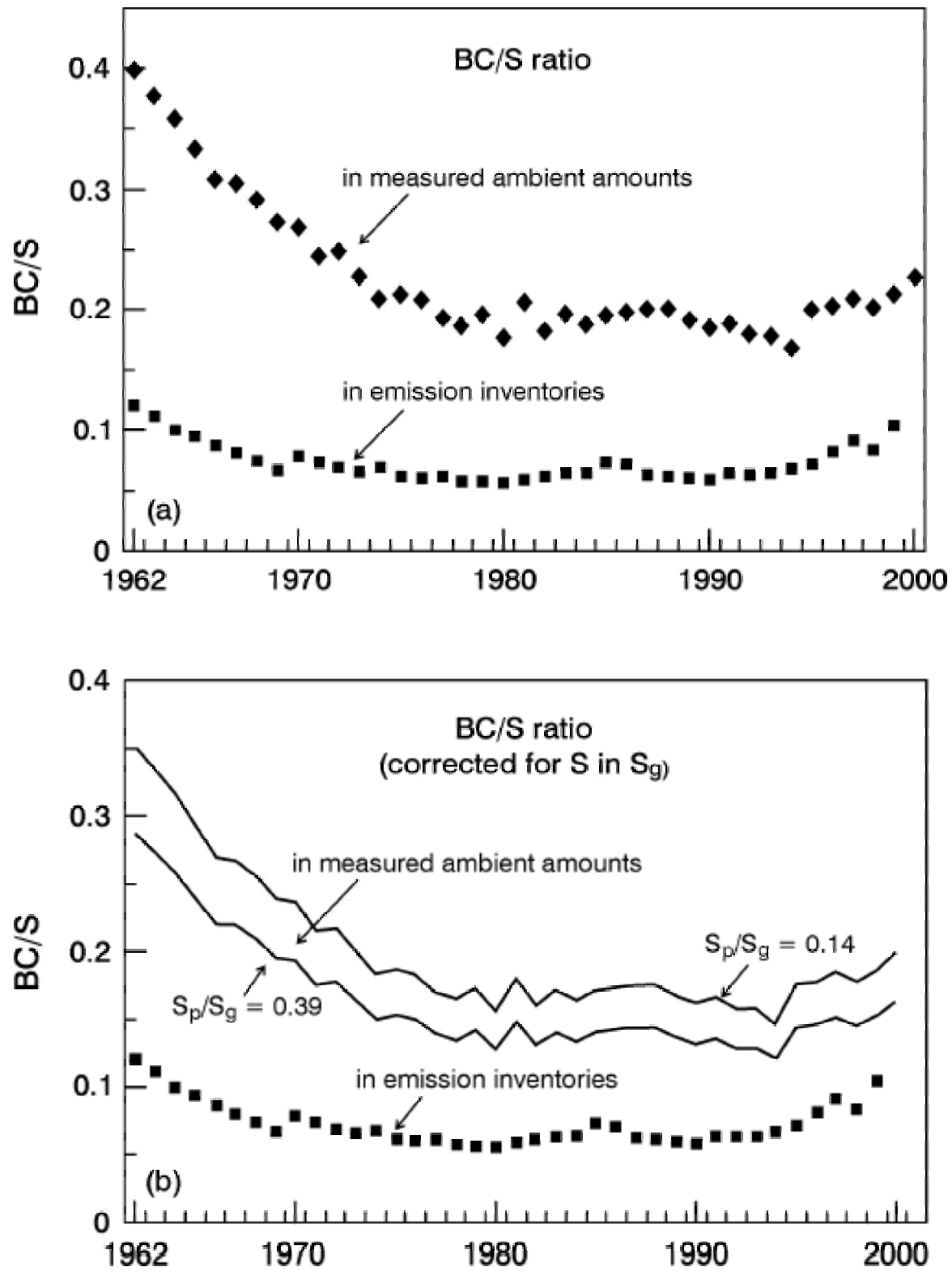


Fig. 7