Large historical changes of fossil-fuel black carbon aerosols

T. Novakov,[1]* V. Ramanathan,[2] J.E. Hansen,[3] T.W. Kirchstetter,[1] M. Sato,[3] J. E. Sinton,[1] J.A. Sathaye[1]

[1]Environmental Energy Technologies Division, Lawrence Berkeley National Laboratory, Berkeley, CA, 94720, USA.

[2]Center for Atmospheric Sciences, Scripps Institution of Oceanography, University of California, San Diego, La Jolla, California 92093, USA

[3]NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA

*To whom correspondence should be addressed. E-mail: tnovakov@lbl.gov

Abstract. Anthropogenic emissions of fine black carbon (BC) particles, the principal light-absorbing atmospheric aerosol, have varied during the past century in response to changes of fossil-fuel utilization, technology developments, and emission controls. We estimate historical trends of fossil-fuel BC emissions in six regions that represent about two-thirds of present day emissions and extrapolate these to global emissions from 1875 onward. Qualitative features in these trends show rapid increase in the latter part of the 1800s, the leveling off in the first half of the 1900s, and the re-acceleration in the past 50 years as China and India developed. We find that historical changes of fuel utilization have caused large temporal change in aerosol absorption, and thus substantial change of aerosol single scatter albedo in some regions, which suggests that BC may have contributed to global temperature changes in the past century. This implies that the BC history needs to be represented realistically in climate change assessments. Records of BC deposition in ice, lake and swamp cores are needed to verify emission histories. Reducing global BC emissions could help to reduce the global warming "bounce" that is expected to accompany controls on sulfur emissions, while also benefiting human health. Our estimates suggest that global diesel emissions are growing and could become the dominant issue if uncontrolled household use of coal and biofuels in developing countries declines in the future.

I. Introduction

Anthropogenic atmospheric aerosols are believed to be a significant climate forcing agent, probably second only to greenhouse gases in their effect on global temperature in the past century [*Houghton et al.,* 2001]. However, the history of atmospheric aerosols is not nearly as well known as that of most gases because of the short atmospheric lifetime of aerosols and thus their very heterogeneous spatial distribution. Moreover, the climate effect of aerosols is complex, as some aerosols cause cooling while others are believed to cause warming.

Over the past decade attention has focused on sulfate (SO_4^{2-}) aerosols from fossil-fuel burning [*Houghton et al.,* 2001; *Charlson et al.,* 1991; *Chuang et al.,* 1997]. The history of anthropogenic sulfate aerosol amount can be estimated in a relatively straightforward way, because unscrubbed fossil fuel SO_2 emissions (the SO_4^{2-} precursor) depend only on the mass of fuel burned and its sulfur content. For recent years it is necessary to account for the fact that developed countries have begun to control SO_2 emissions, because of damaging effects of acid deposition (http://www.nnic.noaa.gov/CENR/NAPAP/), and there are signs that this trend may extend to rapidly industrializing countries [*Streets et al.,* 2001a].

Black carbon (BC), also known as elemental carbon, is a product of incomplete combustion and the principal light-absorbing component of "soot". Thus the BC amount depends critically on how the fuel is burned in addition to fuel amount. The BC emission factor for coal used in inefficient household stoves can be orders of magnitude greater than that for modern electric power plants. During the past century coal utilization in industrialized countries has changed from inefficient and highly polluting to more efficient and less polluting. In industrializing countries, however, similar changes in fuel utilization have not occurred or are just beginning. BC emissions from diesel engines depend on engine design, maintenance, and smoke reduction technology, and are, therefore, different in developed and developing regions. Consequently there are strong regional and temporal variations in the proportion of SO_2 and BC emissions, and this means that there are also variations in aerosol light absorption.

The aerosol single scattering albedo (SSA, the fraction of solar radiation striking the aerosol that is scattered, as opposed to being absorbed) is a key parameter determining the influence of the aerosols on global and regional climate [*Houghton et al.,* 2001; *Hansen et al.,* 1997]. If SSA > 0.95, for example, aerosols will cause cooling under almost all conditions. If

SSA < 0.9, aerosols heat the troposphere significantly and will tend to cause global warming of the surface, even though the surface under the region with aerosols may be cooled due to reduced penetration of sunlight [*Hansen et al.,* 1997; *Ramanathan et al.,* 2001a]. For example, the highly absorbing aerosols over the south Asian region had a strong winter time cooling effect in the Indian sub-continent [*Ramanathan et al.,* 2001a; *Krishnan and Ramanathan,* 2002] that was large enough to offset the warming effect of the greenhouse gases. However, such regional cooling by absorbing aerosols depends on the regional meteorology and may not be applicable elsewhere. In addition, there are indications that strongly absorbing aerosols, by heating the atmosphere, may have a large impact on regional climate and the hydrologic cycle [*Ramanathan et al.,* 2001a; *Menon et al.,* 2002*]*.

Although aerosol SSA has been measured recently, the history of the SSA of anthropogenic aerosols over the past century must be estimated because of the absence of direct measurements. BC and sulfate aerosols, as strongly absorbing and perfectly reflective aerosols, have the greatest influence on the net SSA of anthropogenic aerosols. Anthropogenic organic carbon (OC) and "desert" dust from anthropogenically disturbed soils are both scattering and only weakly absorbing [*Penner and Novakov,* 1996; *Dubovik et al.,* 2002], probably falling slightly on the side of causing global cooling.

In this paper we present estimates of past fossil-fuel BC emissions from the United States, United Kingdom, Germany, Soviet Union, India and China. These countries accounted in 1990 for about 70% and 60%, respectively, of the world consumption of coal and diesel fuel, which are the principal BC-producing fossil-fuels. These countries are representative of global regions with different degrees of industrialization and environmental controls. We also combine the derived BC emissions with SO_2 emission data to estimate temporal trends of SSA of fossil fuel aerosols.

II. Results and discussion

We estimate past BC emissions from annual consumption data for the principal BC-producing fossil-fuels and BC emission factors segregated by utilization sectors: industrial, residential/commercial, and electric power generation for coal, and transportation for diesel fuel. This approach is similar to that used for recent estimates of global fossil-fuel BC emissions for single years [*Penner et al.,* 1993; *Cooke et al.,* 1999]. We obtain data from all six regions for the

period since 1950. Data for the United States cover the 1900-1999 period. However, we note that in the first half of the century the United States was responsible for nearly 50% of fossil-fuel use [*Hansen and Sato*, 2001]. In addition, the United States did not have strong emission controls in that period, so the U.S. data may be reasonably representative of world conditions.

We include only fossil-fuels, i.e., we exclude biofuels as well as biomass burning. Although these are believed to be significant sources of BC, especially in India, we have no good basis for estimating the change of biomass burning over the past century or the history of biofuel use. Furthermore, recent studies have shown that most of the carbon monoxide (CO) in the Indian subcontinent outflow is from biomass and biofuels while most of the aerosols [*Reiner et al.,* 2001] including BC result from fossil fuel burning [*Ramanathan et al.,* 2001b; *Satheesh and Ramanathan,* 2000; *Novakov et al.,* 2000]. Our estimates should thus be recognized as applying only to fossil-fuel sources.

We use BC emission factors assembled by *Cooke et al.* [1999] believed to be uncertain by about a factor of two. These are estimated for undeveloped, semi-developed and developed combustion technologies and are therefore particularly useful in considering past emission changes. The appropriateness of using these emission factors is suggested by *Cooke et al.* [2002] who used these in a global general circulation model simulation and concluded that the mean measured concentrations exceed the simulated values but tend to be within about a factor of two at a majority of the sites.

Sources of fuel data and BC emission factors are summarized in Table 1 and its footnote. Because of the lack of measured values applicable to historical changes in fuel use and combustion technology we use the following approximations. The BC emission factor for utilities in the U.S., U.K., Germany and USSR is assumed to be negligible, and in developing countries it is assumed to be similar to the emission factor for industry in developed countries. Current emission factors for residential and commercial sources in developing countries are assumed equal to those in developed countries in the previous half century. In the U.S. and U.K. the consumption of coal in this sector has drastically declined. Therefore, the decrease in estimated BC emissions would be evident regardless of the adopted emission factor. Diesel BC emission factors range from 10 to 2 g kg^{-1}, respectively, for less developed and industrialized countries [*Köhler et al.,* 2001]. The diesel emission factor in the first half of the century in western countries was probably much higher than now. We assume that the diesel emission

factor in western countries decreased linearly from 10 g kg^{-1} in 1965 to 2 g kg^{-1} in 1985. We assume that the diesel BC emission factor in developing countries remained 10 g kg^{-1} throughout the period considered.

The resulting time series for BC emissions (Fig. 1) show that the United States and European industrialized countries were the largest sources of BC fossil fuel emissions in the early and middle 20th century, but in recent decades China and India have become the greatest sources of BC emissions. The reduction of BC emissions in developed countries was caused mainly by decreased coal use in the residential and commercial sectors. Estimated BC emissions remained proportional to total coal consumption in regions where sectoral coal use fractions remained largely unchanged (e.g., China and India), but not in countries where significant sectoral changes occurred. This implies that estimating BC emissions from total coal consumption [*Tegen et al.,* 2000] is not justified for some regions such as the U.S. or the U.K.

Increased diesel fuel consumption accompanied the decrease of polluting coal use in western countries. Gradual improvements of diesel engine technology [*Kirchstetter et al.,* 1999; *Christoforou et al.,* 1999] in the U.S. and Western Europe caused diesel emission factors to decrease, so their diesel BC emission trends do not mirror increased diesel fuel consumption. Diesel consumption in India and China increased after 1970, contributing markedly to their total BC emissions, under our assumption that no appreciable change of emission controls occurred in those countries.

BC emissions derived from fuel inventories depend on the accuracy of fuel inventories for the principal utilization sectors and, especially, on the chosen emission factors. To illustrate, we compare our estimates for China and India with those obtained by others. *Streets et al.* [2001b] estimated that China emitted 0.72 Tg of BC from fossil fuels in 1995, about 3 times less than our value of 2.1 Tg. One reason for the discrepancy is the use of much lower emission factors, especially for industrial coal and diesel fuel, which were assumed to be similar to the factors for the industrialized countries. As a case in-point we note that diesel vehicles in industrializing countries such as India, and probably China produce 17 times more particulate matter per mile than in the US [Kandli*kar and Ramachandran,* 2000]. In contrast to diesel and industry, our estimate for the residential sector BC of 0.76 Tg agrees well with 0.60 Tg derived by *Streets et al.* [2001b].

Dickerson et al. [2002] estimated BC emissions from India by using two approaches: from fuel use and emission factor of *Streets et al.* [2001b], and from ambient measurements CO and BC measurements over Indian Ocean. The former approach suggest that in 2000 fossil and bio-fuels contributed about 0.1Tg and 0.4 Tg respectively to BC emissions in India. However, estimates from ambient measurements indicate that BC emissions could be 3 to 4 times higher than derived from emission factors and fuel inventories. Our fossil fuel BC estimate for India (for 1995) is about 0.6 Tg, with diesel and residential/commercial sectors contributing about equally to the total. Although >80% of the vehicular fleet consists of gasoline powered vehicles, the total amount of diesel fuel consumed in India exceeds the usage of gasoline by close to a factor of five. Maintenance of vehicles is poor resulting in high emission factors. Although residential coal use is relatively small, fuel use by small-scale industry and commercial enterprises may be significant. Small scale industries are a significant part of the Indian economy and account for 40% of the industrial output [*Kandlikar and Ramachandran,* 2000].

One quantitative check on our BC emissions trends is provided by long-term measurements in the U.K. of ambient "black smoke" (BS) concentration, which is a surrogate for BC (www.aeat.co.uk/netcen/airqual/). Our estimated BC emissions and the observed BS concentration in the U.K. (mean of 165 stations) are in good agreement (Fig. 2), with correlation coefficient $R^2 = 0.91$. Observed improvement in visibility trends in the U.K. after 1973 [*Doyle and Dorling,* 2002], decreased BC concentrations in Southern California [*Christoforou et al.,* 2000], and decreasing aerosol [*Pitz et al.,* 2001] and BC concentrations [*Müller* , 1999] in eastern Germany are qualitatively consistent with our estimates.

We calculate approximate time series of SSA for the different regions using our BC data, published SO_2 emissions [*Lefohn et al.,* 2001], and the following simplifying assumptions: 1) ambient concentrations are proportional to emissions, 2) the sum of BC, organic mass (OM) and sulfate in the form of ammonium bisulfate (NH_4HSO_4) accounts for most of the dry aerosol mass, as commonly observed [*Hegg et al.,* 1997], 3) OM ≈ 1.3 OC, 4) OC ≈ 2 BC for the fuels considered [*Cooke et al.,* 1999]**,** 5) 40 ± 10% of SO_2 is converted to sulfate [*Koch et al.,* 1999], 5) scattering cross sections of SO_4^{2-} (as NH_4HSO_4) and OM are 8 m^2g^{-1}, and scattering and absorption cross sections of BC are 4 m^2g^{-1} and 10 m^2g^{-1}, respectively.

Our calculated SSAs (Fig. 3) agree reasonably well with recent observations. In the INDOEX region (characteristic of aerosols from South Asian countries) the observed SSA in

1998 and 1999 was about 0.85 to 0.9 [*Ramanathan et al.,* 2001b]; consistent with our calculation (Fig. 3). The higher calculated SSAs for the U.S. and Western Europe (Fig. 3) are reasonably consistent with sunphotometer data [*Dubovik et al.,* 2002], although greater absorption is sometimes measured [*Hegg et al.,* 1997]. Instances with larger absorption may be due in part to BC sources that we have not included such as forest fires and other biomass burning.

Figure 3 suggests that changes of fuel utilization, and thus of aerosol composition, have caused substantial temporal change of SSA in some regions. Our calculated aerosol SSA over the U.S. increased from about 0.925 prior to 1945 to about 0.975 after 1975, which is a change of the co-albedo (1 – SSA), a measure of aerosol absorption, by a factor of three. Such a change of SSA would alter the expected effect of the aerosols on temperature from being near neutral to a clear cooling effect. The SSA of fossil fuel aerosols also increased in some other countries (e.g., the United Kingdom), while probably changed little in other places (Fig.3). This adds to the suspicion that aerosols contributed to observed cooling in the period 1940-1965 [*Hansen et al.,* 2001]. Not only did global aerosol amount increase in that period, but the aerosol SSA also increased.

Quantitative interpretation of global and regional climate fluctuations and trends requires use of global climate models, which allow analysis of the roles of multiple climate forcings, feedbacks, and unforced variability. Such modeling of aerosol effects is beginning to occur on a large scale [*Houghton et al.,* 2001], but attention has been focused mainly on sulfates. We emphasize the possible importance of BC aerosols, and the fact that BC amount is not simply proportional to fossil fuel use. As BC is a product of incomplete combustion it is essential to take account of the "technology factor", which alters the magnitude of BC emissions as fuel use moves from one sector to another or as the technology in a given sector changes.

Figure 4 summarizes our estimates of fossil-fuel BC emission vs. time. We estimated emissions from the "rest of the world" by using global fossil-fuel CO_2 emissions data and assuming that the rest of the world has the same mean BC emission per unit CO_2 emission as in the portion of the world for which we have BC emission estimates. We extrapolated from 1900 to 1875 by assuming fossil fuel BC emissions were proportional to fossil fuel CO_2 emissions in that period. Many caveats must accompany these estimates. We may underestimate emission reductions due to improved technology. For example, although we reduced the diesel emission factor in developed countries to 2g kg^{-1}, current diesel emissions in the U.S. are probably in the

range 0.5-1g kg^{-1} [*Kirchstetter et al.,* 1999; *Christoforou et al.,* 2000]. Our estimates refer only to fossil-fuel BC. BC from biofuels and outdoor biomass burning may be of comparable magnitude.

Despite uncertainties in BC emissions, we believe that qualitative features in Fig. 4 are real, specifically the rapid increase in the latter part of the 1800s, the leveling off in the first half of the 1900s, and the re-acceleration in the past 50 years as China and India developed. Quantitative confirming evidence is needed at key locations around the world. If BC deposition histories were obtained from ice, bog and lake cores at several dozen sites around the world, similar to an available ice core record in the Swiss Alps [*Lavanchy et al.,* 1999], it may be possible to use estimated BC histories as a forcing in global climate simulations with reasonable confidence.

Climate forcing by BC aerosols may be of the order of +0.5 W m^{-2} [*Jacobson,* 2001; *Hansen and Sato*, 2001]. The forcing by sulfate aerosols, which is negative (cooling), is probably larger in magnitude [*Houghton et al.,* 2001; *Hansen and Sato,* 2001]. There is strong pressure in developed and developing countries to reduce sulfate emissions because of the adverse effects of acid deposition, and actions to achieve such reductions are already initiated or planned in many countries. The global warming "bounce" expected to accompany reduction of sulfur emissions might be partially alleviated if BC emissions were also significantly reduced [*Hansen and Sato,* 2001] Reduction of BC aerosols, or "soot", may also have significant health benefits [*Künzli et al.,* 2000].

Achievement of reduction of global BC probably requires finding ways to replace or reduce emissions from biofuel uses in developing countries, as well as reduction of BC emission from fossil fuel use in both developed and developing countries. Our estimates (Fig. 1) suggest that global diesel emissions are growing and could become the dominant issue if uncontrolled household use of coal and biofuels in developing countries declines in the future. Although technology can be developed to trap BC emissions of vehicles, a small fraction of nonconforming vehicles can dominate emissions. Improved knowledge of BC emissions and atmospheric amount as a function of time is needed to evaluate the role of BC in climate change and the potential for achieving BC reductions in climate forcing.

Acknowledgements

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Figure captions

Fig. 1. Time series of total and sectoral black carbon emissions for the six countries.

Fig. 2. Comparison of trends in estimated black carbon emissions and measured black smoke concentrations (www.aeat.co.uk/netcen/airqual/) averaged over the United Kingdom.

Fig. 3. Single scatter albedo trend calculated for 40% conversion of SO_2 to SO_4^{2-}. Vertical bars indicate 50% and 30% conversion range.

Fig. 4. Estimated fossil-fuel BC emissions; see text, and Fig. 1.

Table 1. Sector specific BC emission factors in g (BC) kg^{-1} fuel[a]

Fuel sectors[b]	U[c]	SD[c]	D[c]	U.S. and U.K	Germany	USSR	India and China
Hard coal							
Residential/ Commercial	4.6	4.6	2.8	4.6	4.6	4.6	4.6
Industry	1.0	0.3	0.2	1.0	1.0	1.0	1.0
Utilities				0	0	0	0.2
Soft coal							
Residential/ Commercial	8.2	8.2	5.9		8.2		
Industry	1.8	0.6	0.4		1.8		
Diesel	10	10	2	10→2	10→2	10	10

[a]Emission factors from *Cooke et al.,* [1999].

[b]Fuel consumption data from *Gschwandtner et al.,* [1986] and http://www.eia.doe.gov/fuelcoal.html (U.S.), *Warren Spring Laboratory* [1972] and www. aeat.co.uk/netcen/airqual/statbase/emissions/fueldata.html (U.K.), *International Energy Agency* [2000] and *United Nations* [1976] (Germany, USSR and India), and *China Statistical Yearbook* [2000] for China..

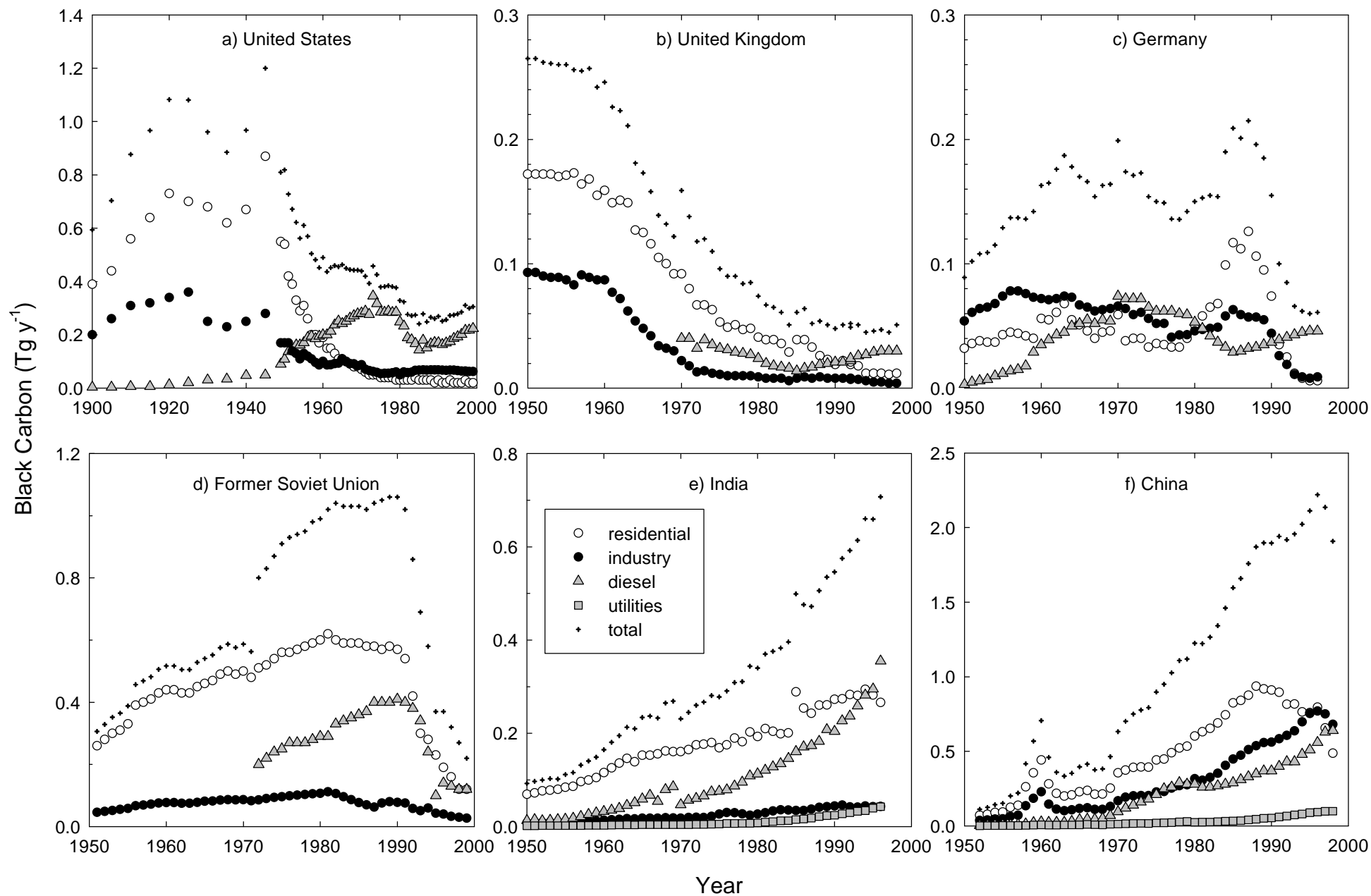[c]U, SD and D refer to undeveloped, semi-developed and developed technologies
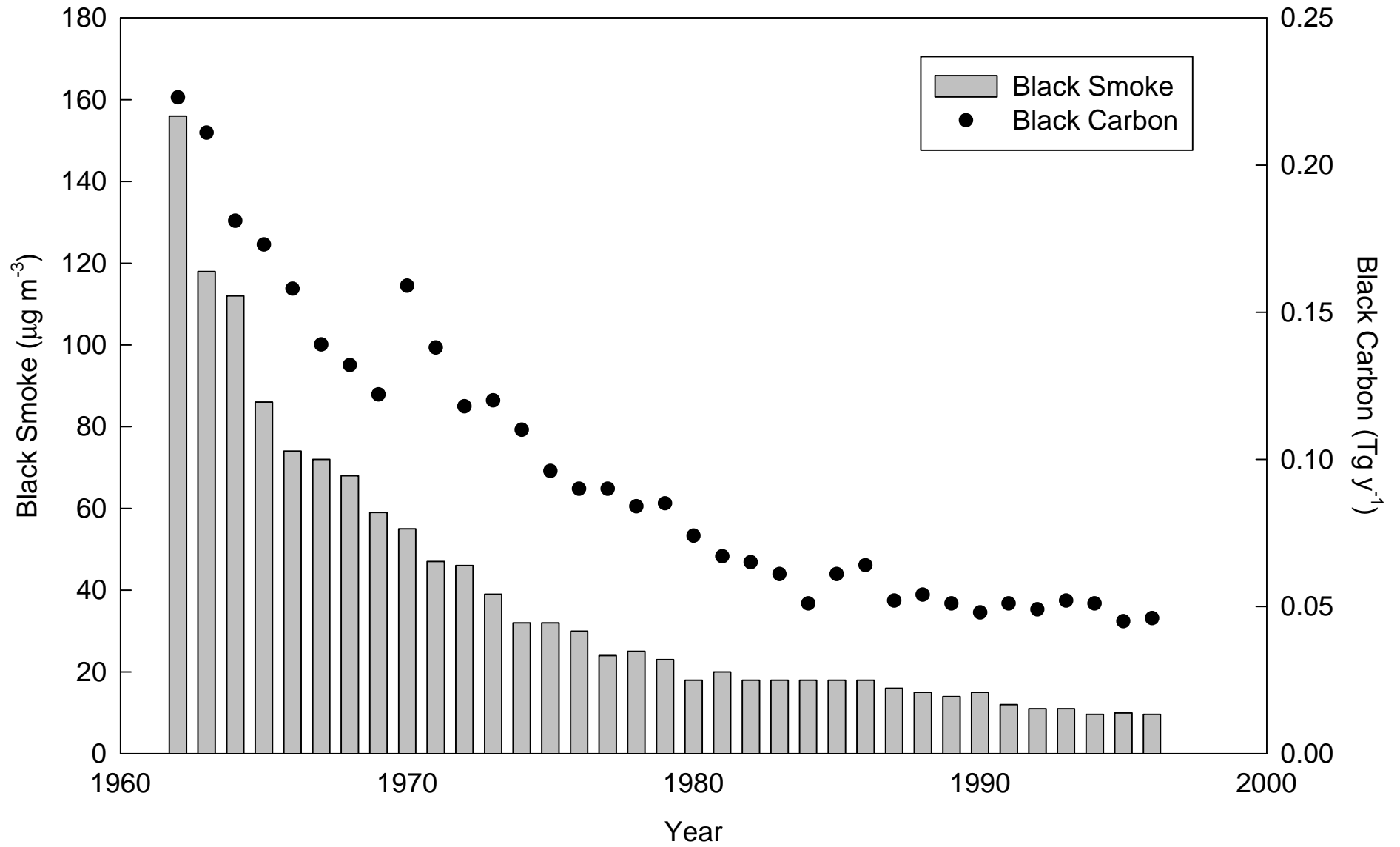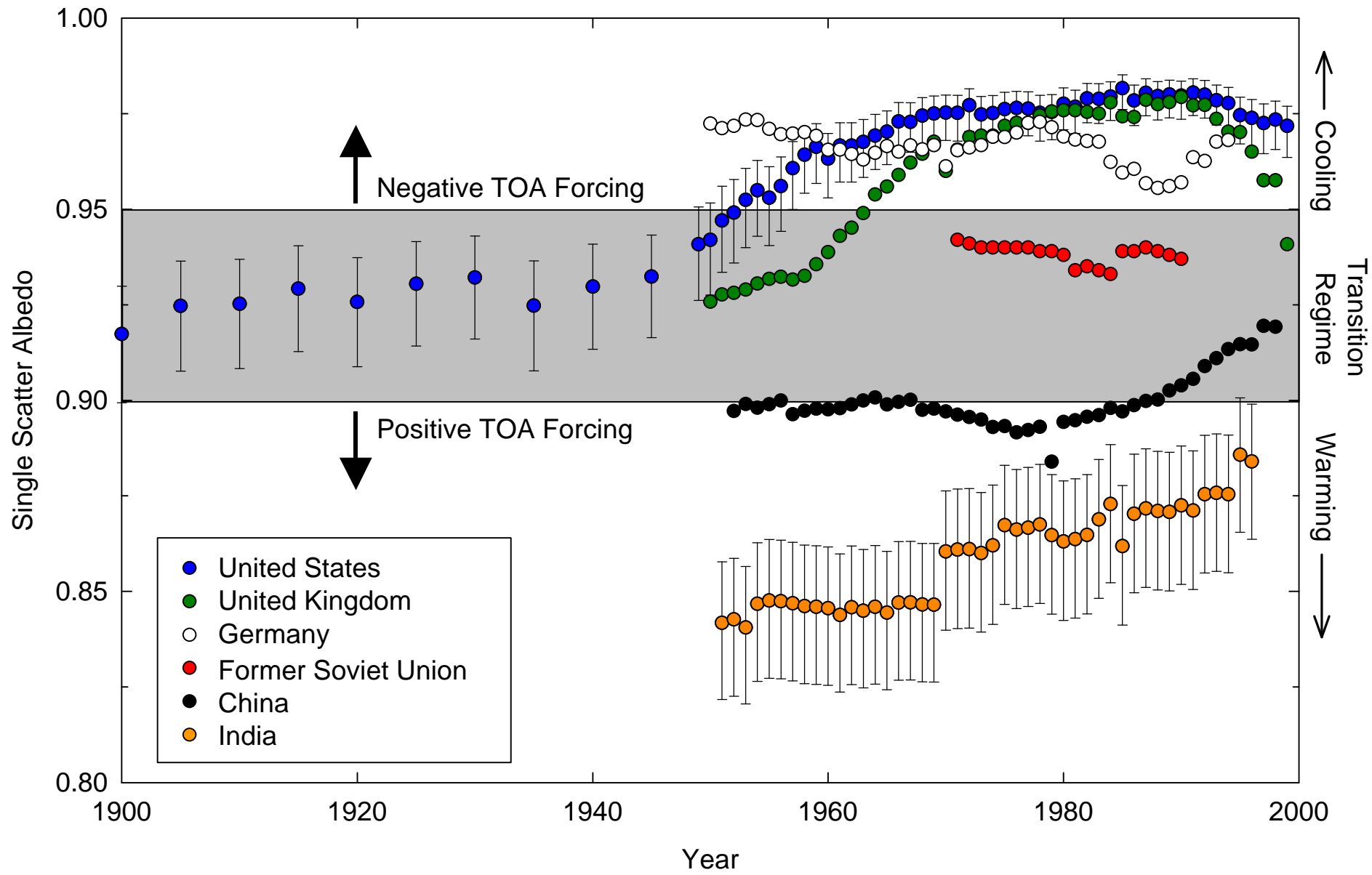
Figure 1

Figure 2

Figure 3

Fig. 4

Fossil-Fuel Black Carbon Emissions

Legend:
- Rest of World
- China
- India
- Former Soviet Union
- Germany
- United Kingdom
- United States

Y-axis: BC (Tg/year), 0 to 7
X-axis: 1875 to 2000