

Large historical changes of fossil-fuel black carbon aerosols

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[1] 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. *INDEX TERMS*: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Constituent sources and sinks; 0345 Pollution—urban and regional (0305). *Citation*: Novakov, T., V. Ramanathan, J. E. Hansen, T. W. Kirchstetter, M. Sato, J. E. Sinton, and J. A. Sathaye, Large historical changes of fossil-fuel black carbon aerosols, *Geophys. Res. Lett.*, 30(6), 1324, doi:10.1029/2002GL016345, 2003.

1. Introduction

[2] 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. Moreover, the climate effect of aerosols is complex, as some aerosols cause cooling while others are believed to cause warming.

[3] Over the past decade attention has focused on cooling 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 is relatively well known, because SO_2 emissions (the SO_4^{2-} precursor) depend mainly on the mass of fuel burned and its sulfur content.

[4] Black carbon (BC) is a product of incomplete combustion. Thus the BC amount depends critically on how efficiently the fuel is burned in addition to fuel amount. 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. Consequently there are strong regional and temporal variations in the proportion of SO_2 and BC emissions.

[5] The aerosol single scattering albedo (SSA, the scattering fraction of extinction) is a key parameter determining the influence of the aerosols on global and regional climate [Houghton *et al.*, 2001; Hansen *et al.*, 1997]. If $\text{SSA} > 0.95$, for example, aerosols will cause cooling under almost all conditions. If $\text{SSA} < 0.9$, 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; Krishnan and Ramanathan, 2002]. 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]. BC and sulfate aerosols, as strongly absorbing and perfectly reflective aerosols, have the greatest influence on the net SSA of anthropogenic aerosols.

[6] 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. We also combine the derived BC emissions with SO_2 emission data to estimate temporal trends of SSA of fossil fuel aerosols.

2. Results and Discussion

[7] 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. 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 global fossil-fuel use [Hansen and Sato, 2001].

[8] We include only fossil-fuels, i.e., we exclude bio-fuels as well as biomass burning. Although these are believed to be significant sources of BC, especially in

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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

^aEmission factors from *Cooke et al.* [1999].

^bFuel consumption data from *Gschwandner 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, India), and *China Statistical Yearbook* [2000] (China).

^cU, SD and D refer to undeveloped, semi-developed and developed technologies.

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].

[9] We use BC emission factors assembled by *Cooke et al.* [1999]. These are estimated for undeveloped, semi-developed and developed combustion technologies and are therefore particularly useful in considering past emission changes.

[10] Sources of fuel data and BC emission factors are summarized in Table 1 and its footnote. 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 coun-

tries in the previous half century. 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 [*Kirchstetter et al.*, 1999; *Christoforou et al.*, 1999]. We assume that the diesel BC emission factor in developing countries remained 10 g kg⁻¹ throughout the period considered.

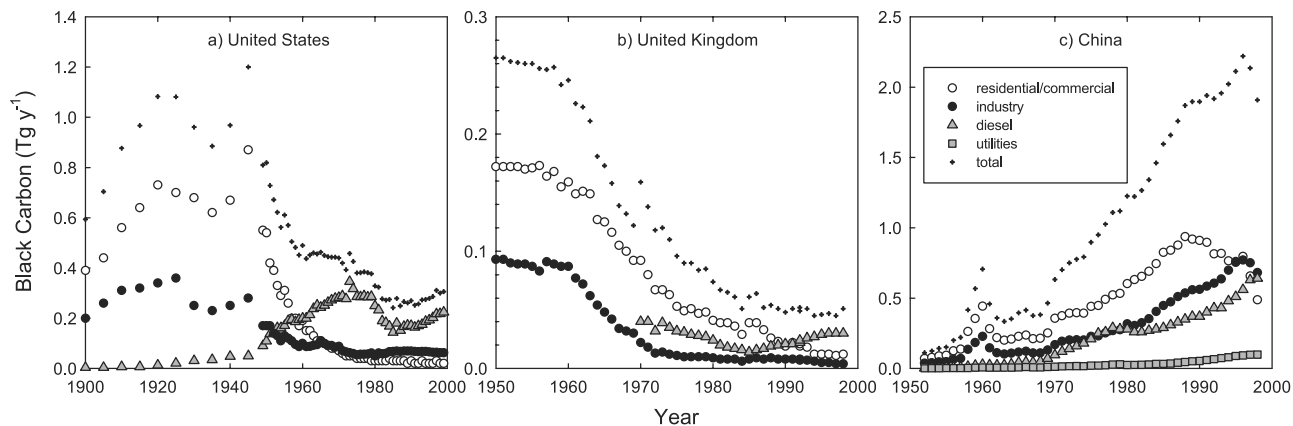
[11] Examples of time series for BC emissions for the US, UK, and China are shown in Figure 1. In developed countries, coal consumption decreased in the residential and commercial sectors, resulting in the reduction of BC emissions. BC emissions remained proportional to total coal consumption in regions where sectoral coal use fractions remained largely unchanged (e.g., China and India).

[12] Increased diesel fuel consumption accompanied the decrease of polluting coal use in western countries. Gradual improvements of diesel engine technology 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, assuming that no appreciable change of emission controls occurred.

[13] Our estimates for China and India are higher than those obtained by *Streets et al.* [2001] and *Dickerson et al.* [2002] who used much lower emission factors, especially for industrial coal and diesel fuel. However, *Dickerson et al.* [2002] found that BC emissions estimated from ambient measurements over the Indian Ocean could be 3 to 4 times higher than derived from emission factors and fuel inventories.

[14] 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, with correlation coefficient $R^2 = 0.91$.

[15] We calculate approximate time series of SSA for the different regions using our BC data, published SO₂ emissions [*Lefohn et al.*, 1999], and the following assumptions: 1) ambient concentrations are proportional to emissions, 2) the sum of BC, organic mass (OM) and

**Figure 1.** Examples of time series of total and sectoral black carbon emissions for three countries.

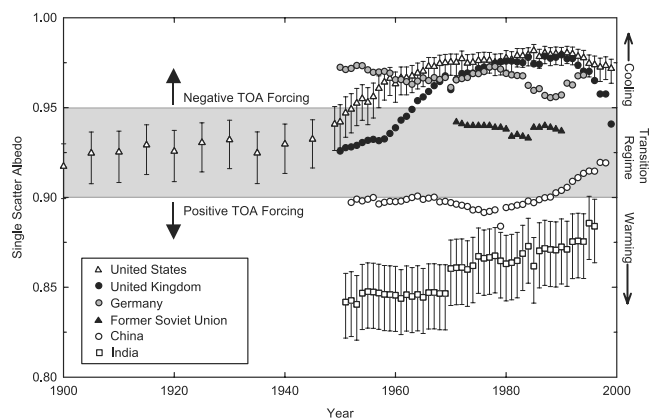


Figure 2. Single scatter albedo trend calculated for 40% conversion of SO_2 to SO_4^{2-} . Vertical bars indicate 50% and 30% conversion range.

sulfate in the form of ammonium bisulfate (NH_4HSO_4) accounts for most of the dry aerosol mass [Hegg *et al.*, 1997], 3) $\text{OM} \approx 1.3 \text{ OC}$, 4) $\text{OC} \approx 2 \text{ BC}$ for the fuels considered [Cooke *et al.*, 1999], 5) $40 \pm 10\%$ 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 $8 \text{ m}^2\text{g}^{-1}$, and scattering and absorption cross sections of BC are $4 \text{ m}^2\text{g}^{-1}$ and $10 \text{ m}^2\text{g}^{-1}$, respectively.

[16] Our calculated SSAs (Figure 2) 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. The higher calculated SSAs for the U.S. and Western Europe (Figure 2) are reasonably consistent with sunphotometer data [Dubovik *et al.*, 2002]. Instances with larger absorption [Hegg *et al.*, 1997] may be due in part to BC sources that we have not included such as forest fires and other biomass burning.

[17] Figure 2 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 ($1 - \text{SSA}$), 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 it probably changed little in other places (Figure 2). 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.

[18] Quantitative interpretation of global and regional climate fluctuations and trends requires use of global climate models. 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.

[19] Figure 3 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.

[20] Despite uncertainties in BC emissions, we believe that qualitative features in Figure 3 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 similar to ice core records in the Swiss Alps [Lavanchy *et al.*, 1999] were obtained, it may be possible to estimate BC forcing in global climate simulations with reasonable confidence.

[21] Climate forcing by BC aerosols may be of the order of $+0.5 \text{ W m}^{-2}$ [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]. Estimates of the current anthropogenic BC climate forcing are of the order of 1/3 to 1/2 of the current CO_2 forcing. The uncertainty in the change of climate forcing in the next few decades is probably larger for BC than for CO_2 . For example, IPCC [Houghton *et al.*, 2001] scenarios have BC increasing during the next 50 years, while others [Hansen *et al.*, 2000] argue that BC could decrease.

[22] 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 effect of achieving BC reductions in climate forcing.

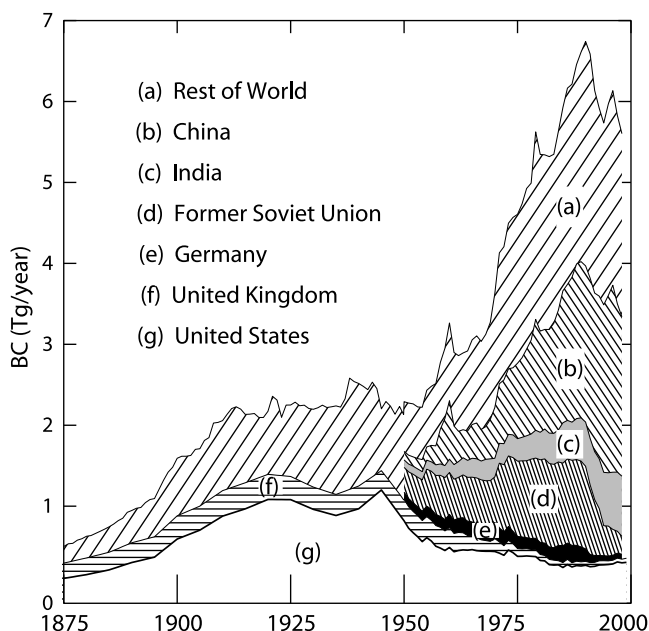


Figure 3. Estimated fossil-fuel BC emissions; see text, and Figure 1.

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References

- Charlson, R. J., et al., Perturbation of the northern hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols, *Tellus, Ser. A and B*, 43, 152–163, 1991.
- Christoforou, C. S., et al., Trends in fine particle concentration and chemical composition in Southern California, *J. Air Waste Manage. Assoc.*, 50, 43–53, 2000.
- Chuang, C. C., et al., An assessment of the radiative forcing of anthropogenic sulfate, *J. Geophys. Res.*, 102, 3761–3778, 1997.
- Cooke, W. F., et al., Construction of a $1^\circ \times 1^\circ$ fossil fuel emission data set for carbonaceous aerosol and implementation in the ECHAM4 model, *J. Geophys. Res.*, 104, 22,137–22,162, 1999.
- Dickerson, R. R., et al., Analysis of black carbon and carbon monoxide observed over the Indian Ocean: Implications for emissions and photochemistry, *J. Geophys. Res.*, 107, 8017, 10.1029/2001JD000501, 2002.
- Dubovik, O., et al., Variability of absorption and optical properties of key aerosol types observed in worldwide locations, *J. Atmos. Sci.*, 59, 590–608, 2002.
- Gschwandtner, G., et al., Historic emissions of sulfur and nitrogen oxides in the United States from 1900 to 1980, *J. Air Pollut. Assoc.*, 36, 139–149, 1986.
- Hansen, J. E., and M. Sato, Trends of measured climate forcing agents, *Proc. Natl. Acad. Sci. U. S. A.*, 98, 14,778–14,783, 2001.
- Hansen, J., et al., Radiative forcing and climate response, *J. Geophys. Res.*, 102, 6831–6864, 1997.
- Hansen, J., et al., Global warming in the twenty-first century: An alternative scenario, *Proc. Natl. Acad. Sci. U. S. A.*, 97, 9875–9880, 2000.
- Hansen, J., et al., A closer look at United States and global surface temperature change, *J. Geophys. Res.*, 106, 23,947–23,963, 2001.
- Hegg, D. A., et al., Chemical apportionment of aerosol column optical depth off the Mid-Atlantic coast of the United States, *J. Geophys. Res.*, 102, 25,293–25,303, 1997.
- Houghton, J. T., et al. (Eds.), *Climate Change: The Science of Climate Change, Contribution of Working Group I to the Second Assessment Report of the Intergovernmental Panel on Climate Change*, Cambridge Univ. Press, New York, 2001.
- International Energy Agency, *World Energy Statistics and Balances*, Int. Energy Agency, Paris, 2000.
- Jacobson, M. Z., Strong radiative heating due to the mixing state of black carbon in atmospheric aerosols, *Nature*, 409, 695–697, 2001.
- Kirchstetter, T. W., et al., On-road measurement of fine particle and nitrogen oxide emissions from light- and heavy-duty motor vehicles, *Atmos. Environ.*, 33, 2955–2968, 1999.
- Koch, D., et al., Tropospheric sulfur simulation and sulfate direct radiative forcing in the GISS general circulation model, *J. Geophys. Res.*, 104, 23,799–23,822, 1999.
- Krishnan, R., and V. Ramanathan, Evidence of surface cooling from absorbing aerosols, *Geophys. Res. Lett.*, 29, 1340, 10.1029/2002GL014687, 2002.
- Lefohn, A. S., J. D. Husar, and R. B. Husar, Estimating historical anthropogenic global sulfur emission patterns for the period 1850–1990, *Atmos. Environ.*, 33, 3435–3444, 1999.
- Menon, S., et al., Climate effects of black carbon aerosols in China and India, *Science*, 297, 2250–2253, 2002.
- Novakov, T., et al., Origin of carbonaceous aerosols over the tropical Indian Ocean: Biomass burning or fossil fuels?, *Geophys. Res. Lett.*, 27, 4061–4064, 2000.
- Ramanathan, V., et al., Aerosols, climate, and the hydrological cycle, *Science*, 294, 2119–2124, 2001a.
- Ramanathan, V., et al., Indian Ocean Experiment: An integrated analysis of the climate forcing and effects of the great Indo-Asian haze, *J. Geophys. Res.*, 106, 28,371–28,398, 2001b.
- Reiner, T., et al., Chemical characterization of pollution layers over the tropical Indian Ocean: Signatures of emissions from biomass and fossil fuel burning, *J. Geophys. Res.*, 106, 28,497–28,510, 2001.
- Satheesh, S. K., and V. Ramanathan, Large differences in tropical aerosol forcing at the top of the atmosphere and Earth's surface, *Nature*, 405, 60–63, 2000.
- Streets, D. G., et al., Black carbon emissions in China, *Atmos. Environ.*, 35, 4281–4296, 2001.
- United Nations, World Energy Supplies 1950–1974, *Stat. Pap. Ser. J No. 19*, United Nations, New York, 1976.
- Warren Spring Laboratory, *National Survey of Air Pollution 1962–71*, Dept. of Trade and Ind., London, 1972.

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