

Retrospective Mapping of the Deposition of Carbonaceous Particles from Fossil-fuel Combustion

Abstract

Carbonaceous particles from oil and coal burning are deposited in soils and sediments everywhere. These fly ash particles are indicators of deposition of airborne pollutants from fossil fuel combustion. Geographical surveys of the deposition during the last few years can be made by means of samples of surface sediments from lakes. In addition, carbonaceous particle records in sediment cores reveal temporal changes in deposition trends associated with industrial development. Soil samples can be used for large-scale and small-scale mapping of the total historical loading during the industrial time period.

Introduction

The burning of oil and coal produces carbonaceous particles together with other unwanted products such as sulphur dioxide. These pollutants are emitted into the atmosphere, transported over shorter or longer distances and finally deposited in the environment. Since the carbonaceous particles are very resistant, they accumulate in soils and sediments. Although sulphur also accumulates, it is more mobile and takes part in chemical processes that, for example, cause acidification of soils and surface water.

Some carbonaceous particles have a very characteristic appearance (Fig. 1) and can be used as specific indicators of deposition of airborne pollutants from fossil fuel combustion. The analyses of sediment and soil records allow the assessment of both recent geographical fall-out conditions and historical trends in atmospheric deposition. This is valuable since instrumented stations for monitoring deposition of airborne pollutants are rather few in most countries, and the available background measurements are generally sparse and limited to the most recent past (< 40 yrs). This report summarizes briefly our investigations of carbonaceous particles in sediments and soils in Sweden.

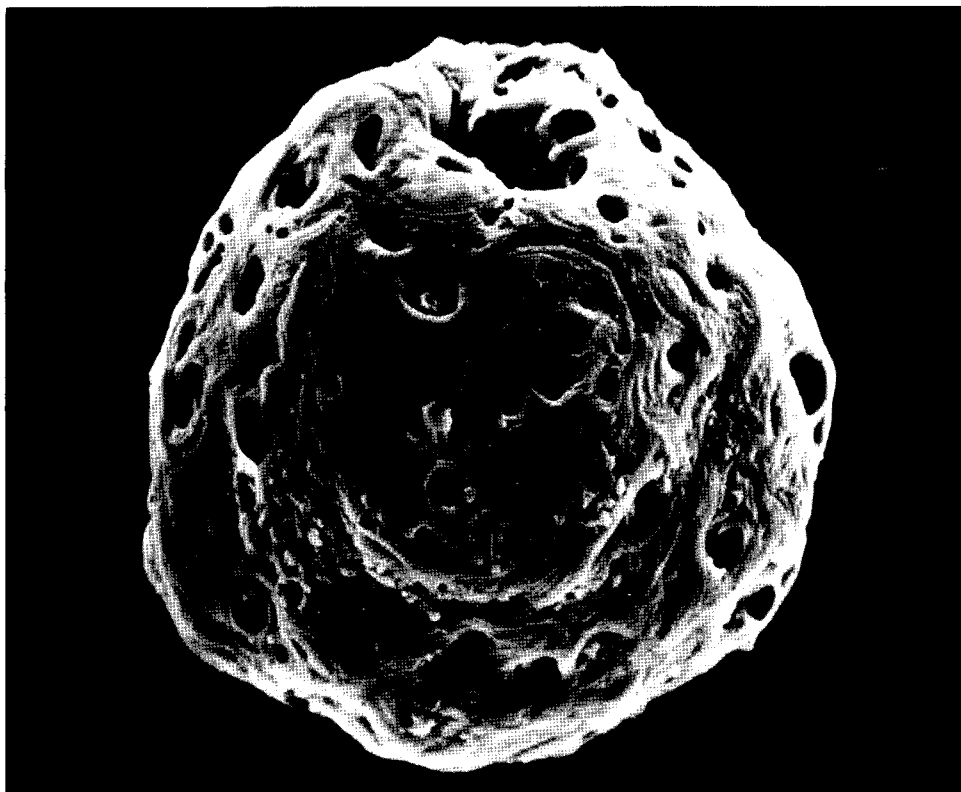


Fig. 1. SEM-photo of a spheroidal carbonaceous particle.

Methods

Griffin and Goldberg (1981, 1983) studied carbonaceous particles in sediments in N. America and used analytical methods based on IR-spectroscopy (Smith *et al.*, 1975). This method is, however, not specific for carbonaceous particles derived from fossil fuel combustion as it also includes charcoal from biomass burning. Sediments and particularly soils in Sweden often contain considerable amounts of charcoal. A method, specific for spheroidal carbonaceous particles, which are formed only by oil and coal burning, was therefore developed (Renberg and Wik, 1985b).

The method of preparing the samples and counting the spheroidal carbonaceous particles includes: oxidizing dry samples in 30 % hydrogen peroxide; washing, and preparation of a water suspension of the residue (carbonaceous particles, mineral grains, diatoms); pouring aliquots of this suspension into glass petri-dishes; evaporation of the water, and counting of the carbonaceous particles under a stereomicroscope (x50). The detectability limit is 5-10 μm , and most of the particles are below 40 μm in diameter.

Mapping of recent deposition by means of lake sediments

Surface sediment samples from lakes can be used for surveys of recent pollution conditions, i.e. of the fall-out during the last few years prior to sampling. Fig. 2 shows results from a study based on the analysis of spheroidal carbonaceous particles in surface sediments (0-1 cm) from 66 lakes around Göteborg in southwest Sweden (Renberg and Wik, 1985a). In lakes near the city more than 200 000 particles were found per gram organic matter (or more precisely per gram sediment matter solved by the peroxide treatment). The corresponding values 50 to 100 km away from the city were 10 000 to 20 000 particles. These are baseline values for the region, but are still at least ten times higher than values recorded in lakes in Lapland, a clean-air region in northern Sweden.

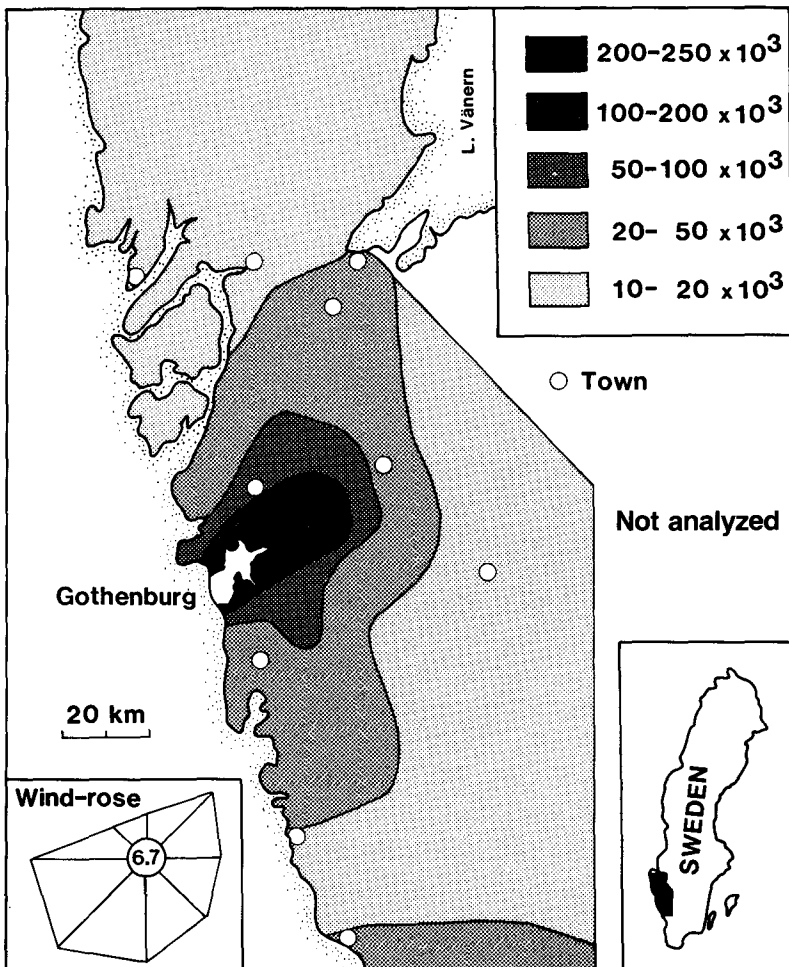


Fig. 2. Map showing spheroidal carbonaceous particle concentrations in surface sediments (0-1 cm) from lakes in southwest Sweden (no. g^{-1} organic matter). From Renberg & Wik 1985a.

In another similar study, comprising more than 100 lakes and covering the whole of Sweden, a correlation has been found between spheroidal carbonaceous particle concentrations in surface sediments and other kinds of data for the deposition of airborne pollutants from fossil fuel combustion and industrial processes (sulphur, lead), (Wik and Renberg, unpublished results).

Mapping the total historical loading with soil samples

Soil samples can be used for geographical mapping of the total historical loading of carbonaceous particles, at least in boreal forests where soil turn-over times are slow. This conclusion is based on a survey of a large area in northern Sweden, where we found that the geographical distribution of spheroidal carbonaceous particles was consistent with the regional air pollution conditions (Wik and Renberg, 1987). Values of more than 15 million particles m^{-2} ground surface were found in the A-horizon of the forest soil in industrialized coastal areas, particularly around the two towns of Skellefteå and Umeå (Fig. 3). Further inland, the numbers of carbonaceous particles decreased to 1 to 2 million m^{-2} and fell still further in the sparsely populated mountain region, although even there considerable numbers were recorded ($> 200\,000\ m^{-2}$). Around several small inland towns, the values were locally relatively higher (Wik and Renberg, 1987).

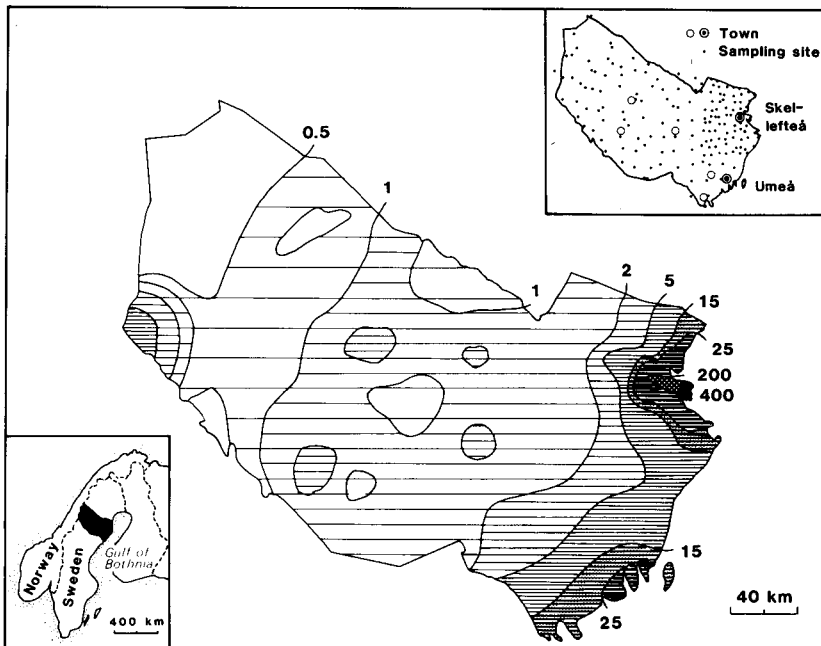


Fig. 3. Map showing the content of spheroidal carbonaceous particles in the A-horizon of pine forest soils in Västerbotten province, northern Sweden (no. $\times 10^6\ m^{-2}$ ground surface). From Wik and Renberg, 1987.

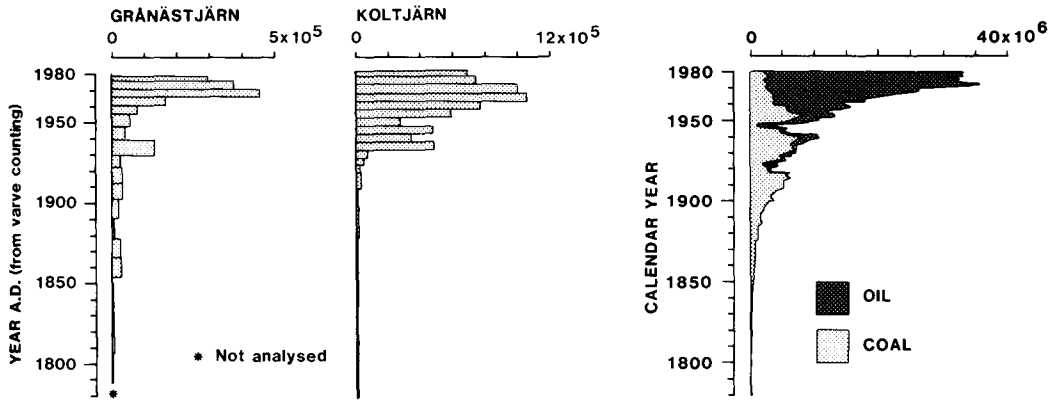
ACCUMULATION OF COARSE CARBONACEOUS SPHERES
IN VARVED LAKE SEDIMENTS : no. $m^{-2}yr^{-1}$ COAL AND OIL COMBUSTION
IN SWEDEN : tonnes

Fig. 4. Diagrams showing the net annual accumulation of spheroidal carbonaceous particles in varved sediments from two Swedish lakes, and to the right, coal and oil consumption trends in Sweden over the last two hundred years. From Wik *et al.*, 1986.

Studies of historical trends in deposition with sediment cores

Lake sediments accumulate continuously and incorporate carbonaceous particles together with other airborne pollutants deposited in the lakes. Lake sediments thus form records which can be used to study the history of atmospheric pollution.

With varved lake sediments it is possible to date the sediments accurately by simple varve-counting, and when quantitative sub-sampling of sediment cores is performed, reliable estimates of the net annual accumulation rate of carbonaceous particles in the sediments can easily be made. Investigations of varved sediments from northern Sweden (Fig. 4) show that the deposition trends of spheroidal carbonaceous particles match the historical trends of fossil fuel consumption. Deposition first increased in the mid-19th century, and a marked increase occurred after World War II. In Sweden there has been a considerable decrease in carbonaceous particle fall-out since the early 1970s. This trend is also recorded in sediments from several non-varved lakes in Sweden. In the U.K. and Norway concentration profiles in the sediments also parallel fuel consumption trends (Battarbee *et al.*, 1988 ; Wik and Natkanski, 1990).

In acidified lakes in northern Europe, there is a close stratigraphic correlation in the sediment records between carbonaceous particles and the changing composition of plant and animal remains that reflect lake acidification caused by acid precipitation (see papers in Battarbee *et al.*, 1990).

REFERENCES

- BATTARBEE, R.W., MASON, J., RENBERG, I. and TALLING, J.F. (eds), 1990, *Palaeolimnology and Lake Acidification*, The Royal Society, London.
- BATTARBEE, R.W., ANDERSON, N.J., APPLEBY, P.G., FLOWER, R.J., FRITZ, S.C., HAWORTH, E.Y., HIGGIT, S., JONES, V.J., KREISER, A., MUNRO, M.A.R., NATKAN-SKI, J., OLDFIELD, F., PATRICK, S.T., RICHARDSON, N.G., RIPPEY, B. and STEVENSON, A.C., 1988, *Lake Acidification in the United Kingdom 1800-1986 : Evidence from Analysis of Lake Sediments*, Ensis Publishing, London.
- GRIFFIN, J.J. and GOLDBERG, E.D., 1979, *Morphologies and Origin of Elemental Carbon in the Environment*, in *Science*, 206, p. 563-565.
- GRIFFIN, J.J. and GOLDBERG, E.D., 1981, *Sphericity as a Characteristic of Solids from Fossil Fuel Burning in a Lake Michigan Sediment*, in *Geochim. Cosmochim. Acta*, 45, p. 763-769.
- GRIFFIN, J.J. and GOLDBERG, E.D., 1983, *Impact of Fossil Fuel Combustion on Sediments of Lake Michigan : a Reprise*, in *Envir. Sci. Technol.*, 17, p. 244-245.
- RENBERG, I. and WIK, M., 1985a, *Carbonaceous Particles in Lake Sediments - Pollutants from Fossil Fuel Combustion*, in *Ambio*, 14, p. 161-163.
- RENBERG, I. and WIK, M., 1985b, *Soot Particle Counting in Recent Lake Sediments : An Indirect Dating Method*, in *Ecol. Bull.*, 37, p. 53-57.
- SMITH, D.M., GRIFFIN, J.J. and GOLDBERG, E.D., 1975, *Spectometric Methods for the Quantitative Determination of Elemental Carbon*, in *Anal. Chem.*, 47, p. 233-238.
- WIK, M. and NATKAN-SKI, J., 1990, *British and Scandinavian Lake Sediments Records of Carbonaceous Particles from Fossil Fuel Combustion*, in *Phil. Trans. R. Soc. Lond.*, B, 327, p. 319-323.
- WIK, M. and RENBERG, I., 1987, *Distribution in Forest Soils of Carbonaceous Particles from Fossil Fuel Combustion*, in *Water, Air and Soil Pollut.*, 33, p. 125-129.
- WIK, M., RENBERG, I. and NATKAN-SKI, J., 1986, *Sedimentary Records of Carbonaceous Particles from Fossil Fuel Combustion*, in *Hydrobiologia*, 143, p. 387-394.