

## Pre-Industrial atmospheric background black carbon concentrations in North America.

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Black carbon (BC) aerosols in the atmosphere absorb solar radiation, and cause heating of the atmosphere, may alter Earth's cloud cover, and impact precipitation cycle. The best estimate for radiative forcing from BC with 90% certainty is + 1.1 Wm<sup>-2</sup> (+ 0.17 to 2.1 Wm<sup>-2</sup>), second only to that of CO<sub>2</sub>. A major uncertainty in the estimation is a serious lack of atmospheric BC measurements. Currently, models are used to estimate BC emissions into the atmosphere based on energy consumption and emission factors for variety of fuels and combustion methods used. Here we report long term measurements of BC in the atmosphere, over a period of several centuries using lake sediments, which contain records of atmospheric aerosols over thousands of years, if not longer. Retrieval of such records provides an invaluable source for understanding changes in the atmosphere with time. Several studies have been conducted with such an objective. Owing to a lack of knowledge of deposition rates of atmospheric aerosols into the lake sediments, a major shortcoming of these studies have been an inability to convert the measurements of chemical species in lake sediments into the atmosphere. We have developed a technique to overcome this shortcoming by measuring black carbon in atmospheric aerosols, and in lake sediment cores. The concentrations of BC were determined in daily filters collected at Whiteface Mountain, NY, from 1978 to 2005. Cores from two lakes around Whiteface Mountain, NY, were collected. Cores were (1) sectioned in thin slices, (2) freeze dried, (3) dated using the <sup>210</sup>Pb technique, (4) BC chemically separated, and (5) concentrations measured using the thermal-optical method. The deposition rate of BC from the atmosphere to the lakes was determined by comparing the BC concentration in air and the sediments for the 1978 -2005 period. The deposition rate so determined was used to convert the BC in the sediment into the atmosphere for the ~1978 to ~1100 period. The BC concentrations for the industrial period, ~1850 was low but rapidly increased from ~1900, peaked ~1925, decreased very slowly up to ~1980, followed by a sharp decrease, Fig 1.

Concentration has remained fairly steady since, ~70 ngm<sup>-3</sup>. A comparison of BC so determined is made with BC emissions estimated by NASA's global circulation model GISS GCM Model E (Koch et al, J. of Climate, 24, 2693-2714, 2011), and the European Atmospheric Chemistry Transport Model Oslo CTM2 (Skeie et al, ACP, 11, 6809-6836, 2011), using energy consumptions. Both model predictions agree reasonably well from ~1900 to about 1925, but under-predict by factors of 2 ~1950 and by a factor of 3 to 4 ~1980. These estimates are crucial for evaluating climate models against the *true* long-term measurements and will help seriously improve their predictive capability to simulate future warming trends. The 'natural' background of BC in the atmosphere for the ~1100 to ~1800 period was also fairly steady, with a notable spike ~1400, at 50ngm<sup>-3</sup>, only slightly lower than the current measurements at Whiteface Mountain, 70 ngm<sup>-3</sup>. The control measures in North America clearly have reduced atmospheric BC concentrations from ~700ngm<sup>-3</sup> ~ 1950s.

Figure 1. Comparison of atmospheric BC concentrations determined using lake sediments with model estimates.

Historical [EC]<sub>atm</sub> measurements reconstructed from lake sediments of WP. These were compared with the modeled derived atmospheric BC concentration BC<sub>22m</sub> data from Skeie et al. [2011].

