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TRENDS IN ATMOSPHERIC ELEMENTAL CARBON CONCENTRATIONS, ~1850 – 2005

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Introduction

The atmospheric loading of carbonaceous aerosols which consists of elemental carbon (EC) and organic carbon (OC) has substantially increased since pre-industrial times (IPCC, 1995). EC is a product of incomplete combustion, characterized by high molecular weight, non-volatility, and a graphitic structure. Atmospheric EC has been found to vary greatly, from a background level of 1 ng/m^3 at South Pole, between 7,000 and 21,000ng/m^3 in Paris, France. Exceedingly high concentrations, daily averages of ~25,000 ng m^{-3}, are now commonly observed in urban areas of South Asia.

Elemental and other carbonaceous aerosols have been linked to cardiovascular and pulmonary disease in humans. EC aerosols strongly absorb solar radiation, and play a key role in the earth's temperature regulation, and after greenhouse gases may be the second biggest contributor to global warming. The magnitude of the forcing by EC is uncertain, estimated at ~ 0.5 W/m^2. Owing to their relatively short residence times, ~ 6 days, their concentrations are highest over regions with high industrial activities and/or biomass burning. Since field data are sparse the BC concentrations are estimated from models using fossil fuel consumptions. The models, however, have not been validated by field data. Hence, long-term data are needed to verify the models.

Our objective in this work is to *directly* determine the atmospheric {EC}, where {X} represents concentration of X, in the monthly composites of daily filters collected from 1978 to 2005 at Whiteface Mountain. We further propose to extend the {EC} measurements by about another 100 years by a novel approach using lake sediments. We show in this work that, at a given location, {EC} in atmosphere can be related to {EC} in sediments by $\{EC\}_{sed} = K\{EC\}_{atm}$ where K is a constant dependent on wet and dry deposition and sedimentation rate of EC for a given lake. We would determine K from the measured $\{EC\}_{atm}$ and $\{EC\}_{sed}$ for the period ~1978 to ~2005 and then apply it to the measurements of $\{EC\}_{sed}$ for the ~1980 to ~1850 period to obtain the historical atmospheric {EC}.

Theory of atmospheric EC determination from sediment EC

It can be shown that the total flux, F_T, from wet and dry deposition into lake sediment can be expressed as

$$F_T = -V_d \{EC\}_{atm} + \{EC\}_{atm}(x,y,0,t)\, W_r\, P_o = [-V_d + w_r\, p_o]\, \{EC\}_{atm} \quad .....(1)$$

where V_d is the dry deposition velocity in length per unit of time at a certain reference height, and $\{EC\}_{atm}$ is the concentration of EC at that height, $\{EC\}_{precip}$ (x,y,0,t) is the concentration of EC in precipitation at a location x,y and zero height at a given time and p_o

is the precipitation intensity (mm/h or m/y). The wash out ratio w_r relates EC concentrations in the atmosphere and in precipitation: $w_r = \{EC\}_{precip}(x,y,0,t)/\{EC\}_{atm}(x,y,0,t$

It can be shown that

$$\mathbf{\{EC_{sed}\} = K \{EC_{atm}\}} \quad \ldots\ldots (2)$$

where K depends upon the wet and dry deposition of EC bearing aerosols, mixing and sedimentation processes. With our current understanding of the atmospheric transport and deposition processes the value of K can only be determined empirically. From the measurements of $\{EC\}_{atm}$ in filters at Whiteface and $\{EC\}_{sed}$ we can determine K for 1978 to 2005. Assuming the value remains constant, as it should, it can be used to determine $\{EC\}_{atm}$ for an additional 100 years from the measurements in sediment..

Experimental Methods

Daily aerosol samples were collected on Whatman 41 filters using hi-vol samplers from 1978 to 2005 at Whiteface Mountain (44.37^0 N, 73.90^0W, 1.5 km above mean sea level). {EC} were determined in monthly composites prepared from aliquots of the daily samples by thermal-optical method [Khan et al., 2006, J. of Geophysical Research, 111, D04303, doi:10.1029/2005JD006505**].**

Sediment cores were collected from West Pine Pond (44º20´N, 74º25´ W) near Whiteface Mountain. Coring was done with a gravity – driven device. Sediment cores were sliced and freeze dried. The freeze-dried samples were ground to a fine powder, homogenized, and stored in 5-cm diameter plastic jars for analyses. The age or the time of deposition of each 3 to 5 mm section was determined by ^{210}Pb (half-life, 22.3 y) method [e.g., Appleby, 2001, Chronostratiographic techniques in recent sediments in Tracking Environmental Change Using Lake Sediments (editors W. M. Last, and J.P. Smol), Vol 1. 2001]. EC was chemically separated from other species (Lim and Cachier, 1996; Chemical Geology 131, 143-154) and then its concentration determined by the thermal optical method.

Results and Discussion

Figure 1 gives the plots of annual means of {EC} from 1978 to 2005 at Whiteface Mountain. In general, the data show a large decrease in {EC} with time. Further, the decrease in {EC} was observed in three modes i.e., 1978 to1986, 1987 to 1996, and 1997 to 2005 periods. Mean concentrations for the 1978- 1986, 1987- 1996, and 1997- 2005 periods were 549±152, 225±66, and 65±15 ng/m^3, respectively. A 59% decrease in mean {EC} was observed between 1978-1986 and 1987-1996 periods. A further 88% decrease in mean {EC} was observed between 1978- 986 and 1987-2005 periods.

Approximately 200 to 300 mg aliquot of dried sediment from each sliced layer was weighed and EC separated chemically. using the procedure cited earlier, and {EC} measured by the thermal-optical method. Using the $\{EC\}_{atm}$ and $\{EC\}_{sed}$, we calculated the constant K from equation 2 to be ~10500±2700 m^3/g. The value of K was then used to determine $\{EC\}_{atm}$ (Figure 2) and compared with the national BC emissions (Novakov et al., 2003, Atmospheric Environment 38, 4155-4163). EC_{atm} varied from 61 to 758ng/m^3. EC_{atm} concentration peaked between 1917 and 1930 when the coal was the major source of energy. From 1940 to 1970, residential wood consumption declined steadily as a result of abundant supply, low relative prices, and convenience of fossil fuels relative to wood for home heating, cooking, and heating water. A 28% percent decrease in coal BC

emissions from industrial sector was attributed to the improved coal combustion technology in North America [Bond et al., 2007, Global Biogeochemical Cycles, 21, GB2018, doi:10.1029/2006GB0028400] during 1950-2000. {EC} decreased substantially during 1940-1970. Energy consumption data for six adjacent states (IL, IN, MI, OH, PA and WI) which influence the EC at Whiteface Mountain, NY (Khan et al., 2006, JGR, 111, D04303, doi: 10.1029/2005JD006505] show that coal consumption in residential and industrial sectors decreased during 1960 and 2003 by several folds. This trend is evident from our data that EC varied little from 1930 and 1970 and then decreased by several folds till 2005. A large drop in $\{EC\}_{atm}$ from 56 to 30ng/m^3 (~46%) observed between 1978 and 1990 is consistent with the decrease in industrial emissions. Further decrease in EC_{atm} from 300 to 61ng/m^3 was mainly due to the improved diesel combustion technology in the transport sector. Further research is needed to validate the BC emissions for the northeastern US to use the EC values in climate change studies.

Conclusions

The annual $\{EC\}_{atm}$ at Whiteface Mountain (44.37^{0}N, 73.90^{0}W), NY from 1978 thru 2005 showed an unmistakable decreasing trend. The mean $\{EC\}_{atm}$ for 1978-1986, 1987-1996, and 1997-2005 periods were 549± 152, 225±66, and 65±15ng.m^{-3}, respectively. No significant seasonal variation in $\{EC\}_{atm}$ were observed.

Results show $\{EC\}_{atm}$ varied from 61 to 758ng.m^{-3} between 1870 and 2005. The $\{EC\}_{atm}$ peaked around 1920 and then decreased. The $\{EC\}_{atm}$ from ~1850 to the present was compared with the black carbon (BC) emissions based on consumption of fossil and bio-fuels and showed good correlation.

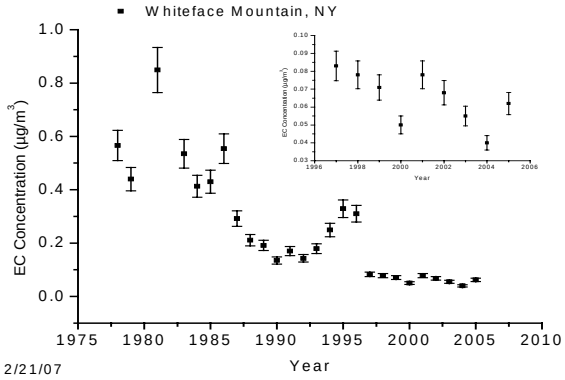

Figure 1: Annual mean EC concentrations at Whiteface Mountain, NY.

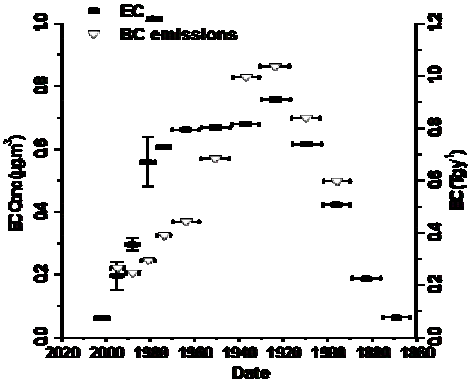

Figure 2. $\{EC\}_{atm}$ vs estimated BC based on fossil fuel consumption emissions.