TRENDS IN ATMOSPHERIC ELEMENTAL CARBON CONCENTRATIONS, ~ 1835 to 2005

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Collaborators

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- Jianjun Li
- Tanveer Ahmed
- Kamal Swami
- A. Bari

Objectives:

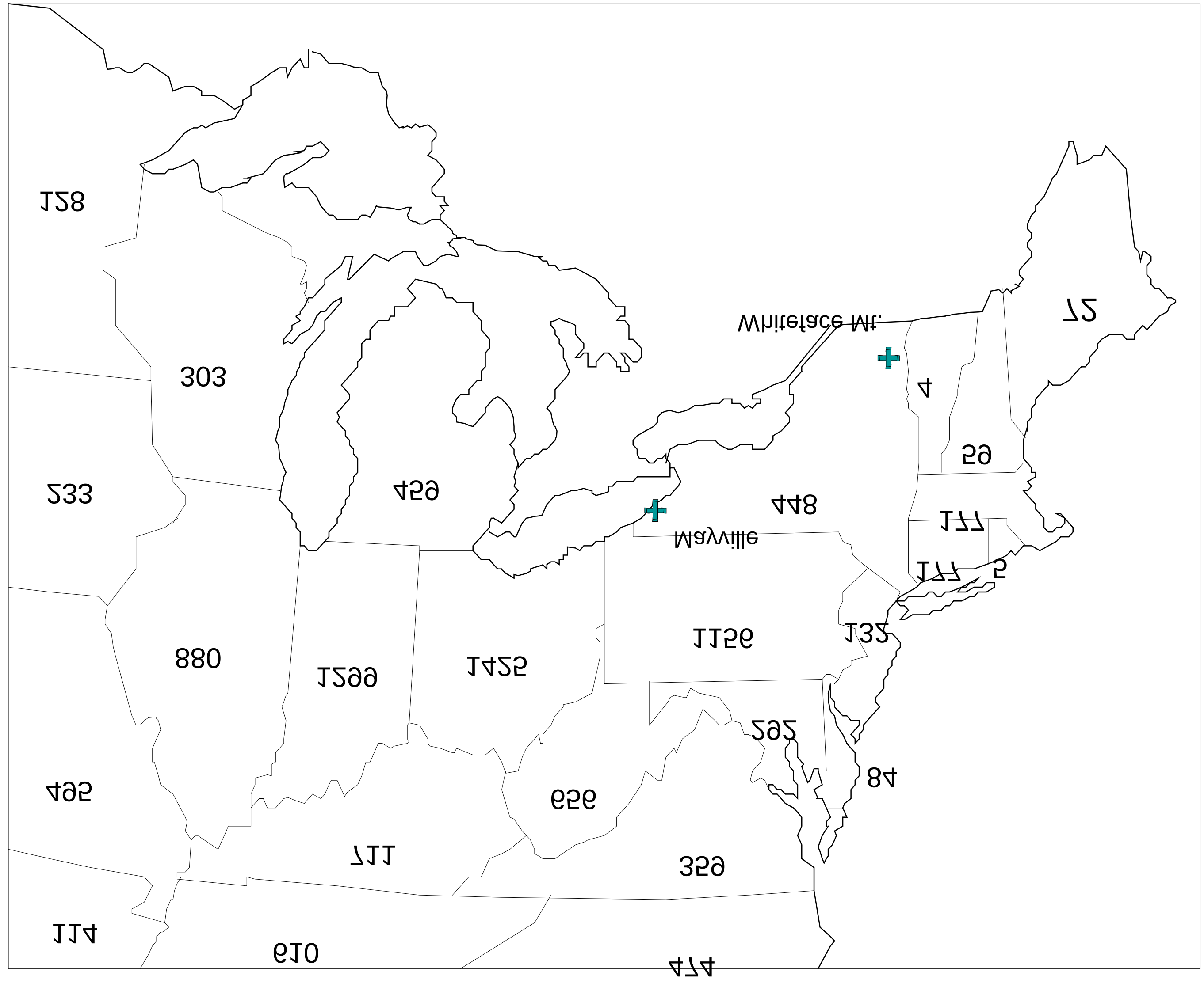
1. Determine EC concentrations in the atmosphere across the northeastern US from about 1850 to 2005;

2. Compare the EC data with historical emission records;

4. Use the data to estimate radiative forcing.

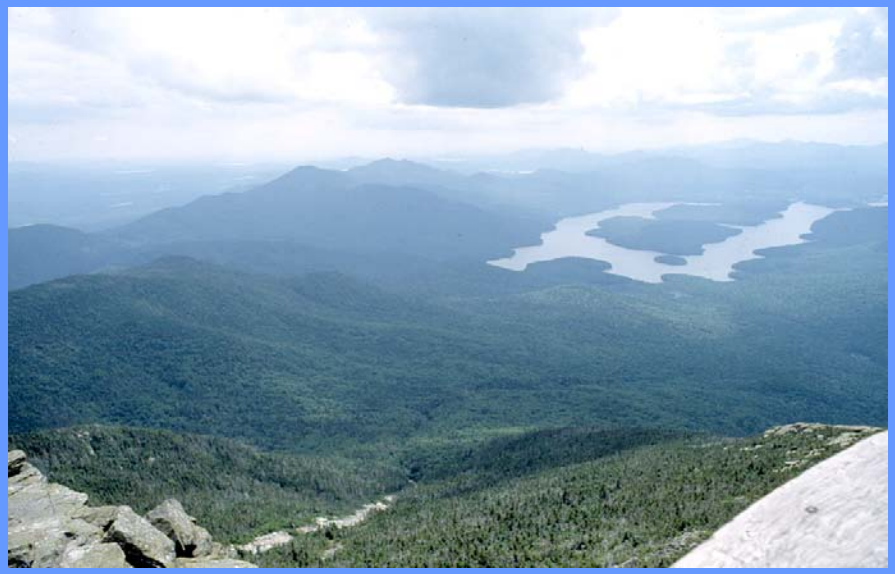
Approach:

1. Determine EC concentrations in monthly composites of archived filters at Whiteface Mountain, NY, from 1978 through 2007; and Mayville, NY from1983 to 2007.
2. Develop a new technique to determine EC concentrations using lake sediments from around 1978 to ~1850.



L. Husain, V.A. Dutkiewicz, and M. Das, *Geophysical Research Letters*, **25**, 7, 967- 970, 1998.

Whiteface Mountain, NY

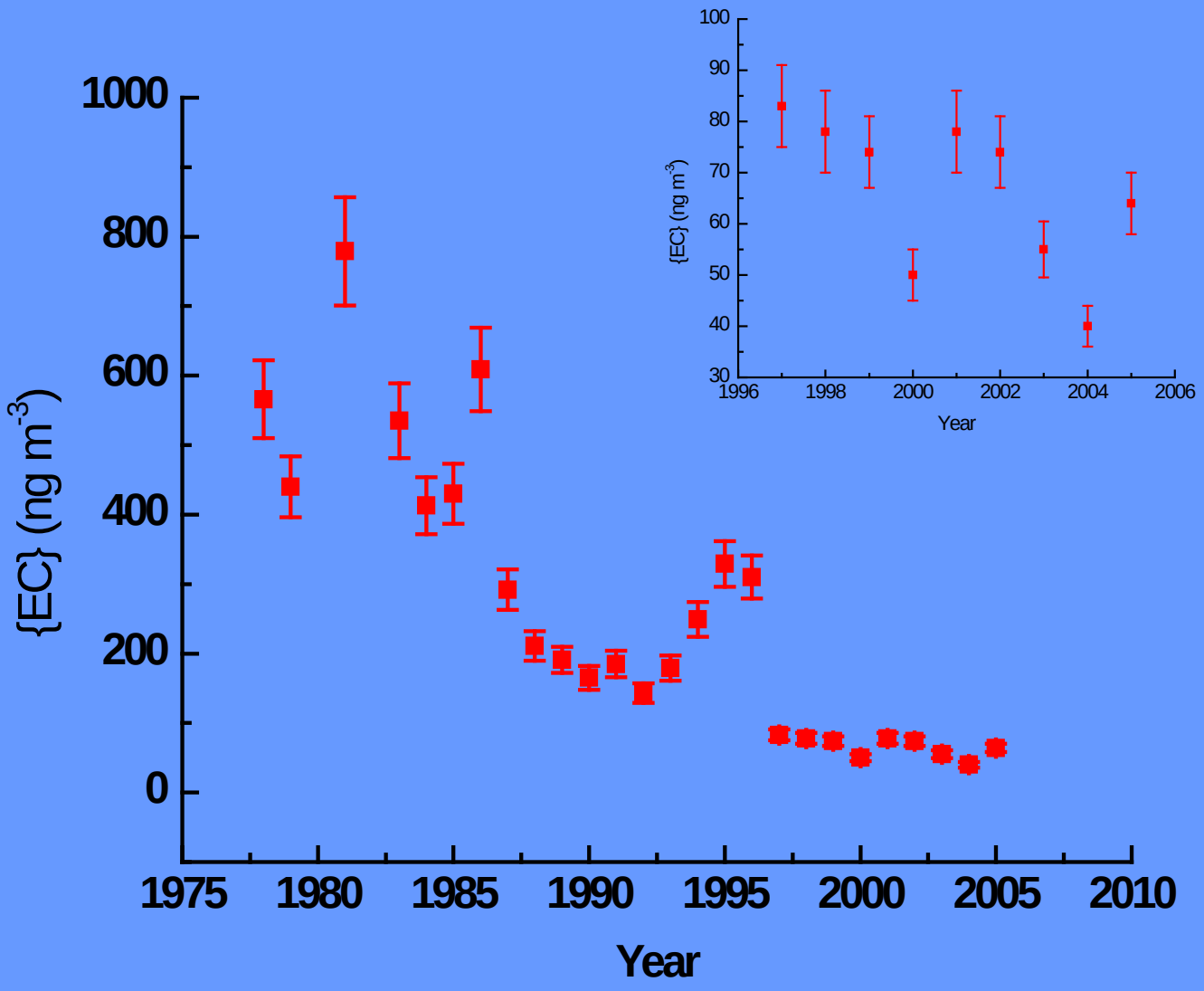


Methodology

- Whatman 41 filter does not permit a direct EC Measurement by Thermal Optical (TOT) method
- A pretreatment chemical technique was developed (Li et al., 2002) to separate EC from cellulose filters and transferred to Quartz filter
- Collection efficiency of Whatman 41 filter for EC aerosols was ~90%.
- EC collection efficiency of Whatman 41 filter were also compared with the Quartz filter and found to be 92% (R^2=0.82)

EC concentrations at WFM 1978-2005

Month	1	2	3	4	5	6	7	8	9	10	11	12	Annual Mean	SD
1978						0.554	0.730	0.638	0.257	0.582	0.633		**0.566**	**0.163**
1979	0.617	0.222	0.386		0.412	0.362	0.638	0.287	0.342	0.450	0.533	0.594	**0.440**	**0.139**
1980														
1981	1.063	1.101	0.686	0.757	0.655	0.473			0.560			1.494	**0.849**	**0.343**
1982														
1983	0.783	1.573	0.738	0.424	0.315	0.323	0.248	0.212	0.292	0.402	0.372	0.734	**0.535**	**0.383**
1984	0.336	0.398	0.303	0.559	0.365	0.323	0.337	0.435	0.271	0.627	0.540	0.461	**0.413**	**0.113**
1985	0.545	0.613	0.267	0.597	0.283	0.254	0.619	0.307	0.396	0.360	0.375	0.543	**0.430**	**0.144**
1986	1.041	0.342		0.599	0.918	0.787	0.396	0.398	0.609	0.213	0.792		**0.609**	**0.272**
1987	0.288	0.283	0.290	0.302			0.237	0.556		0.330	0.187	0.157	**0.292**	**0.114**
1988	0.207	0.152	0.180	0.153	0.216	0.287	0.381	0.185	0.300	0.179	0.160	0.131	**0.211**	**0.075**


{EC} (ng m-3)
1000
800
600
400
200
0
1975
1980
1985
1990
1995
2000
2005
2010
Year
{EC} (ng m-3)
100
90
80
70
60
50
40
30
1996
1998
2000
2002
2004
2006
Year

Recovery of Atmospheric EC Data Deposited in Lake Sediments for the Past ~150 Years

Dry and wet deposition in term of EC flux into the sediment can be expressed as [Seinfield and Pandis, 1998] :

For dry EC deposition:

$F_d = -V_d \{EC\}_{atm}$ …… (1)

where, F_d is the dry deposition flux, V_d is the dry deposition velocity, and $\{EC\}_{atm}$ is the concentration of EC.

For wet deposition,

Fw = {EC}precip (x,y,0,t) po ……(2)

- For EC in sediment

$$\{EC\}_{sed} = (K)\ \{EC\}_{atm}$$

- Where K depends upon
  - drainage or run-off
  - wet deposition of EC at lake surface
  - mixing and sedimentation processes

- Measurements be made on samples integrated over several years.
- Not affected by short-term meteorological factors.

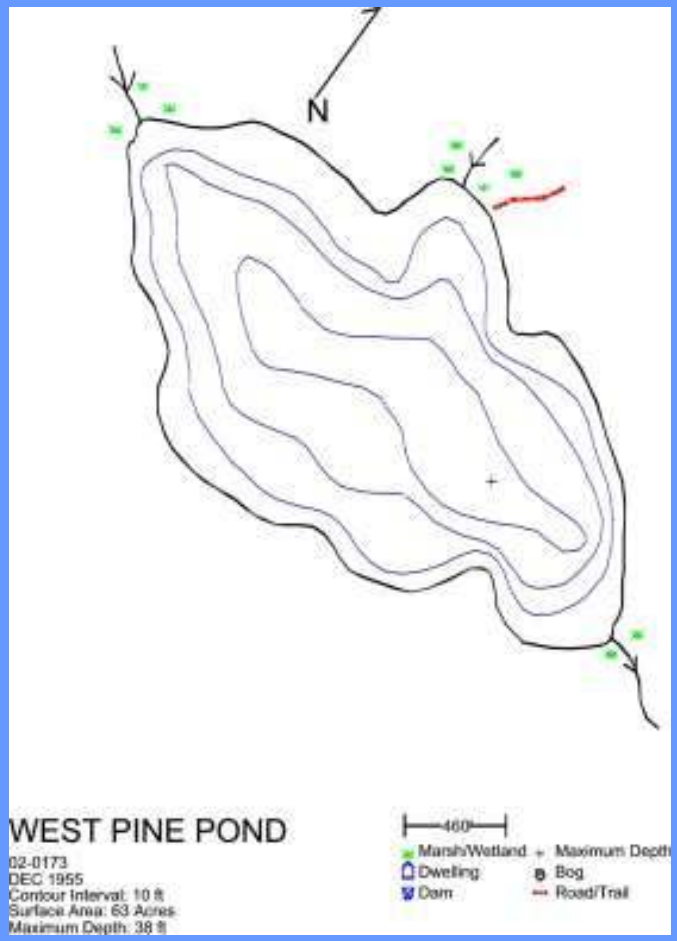
Lake Sediment Collection

Sediment cores were collected from West Pine Pond (44º20´N, 74º25´ W), within ~ 40 km of Whiteface Mountain.

No motor boats, no camping, no homes and away from roads.

West Pine Pond

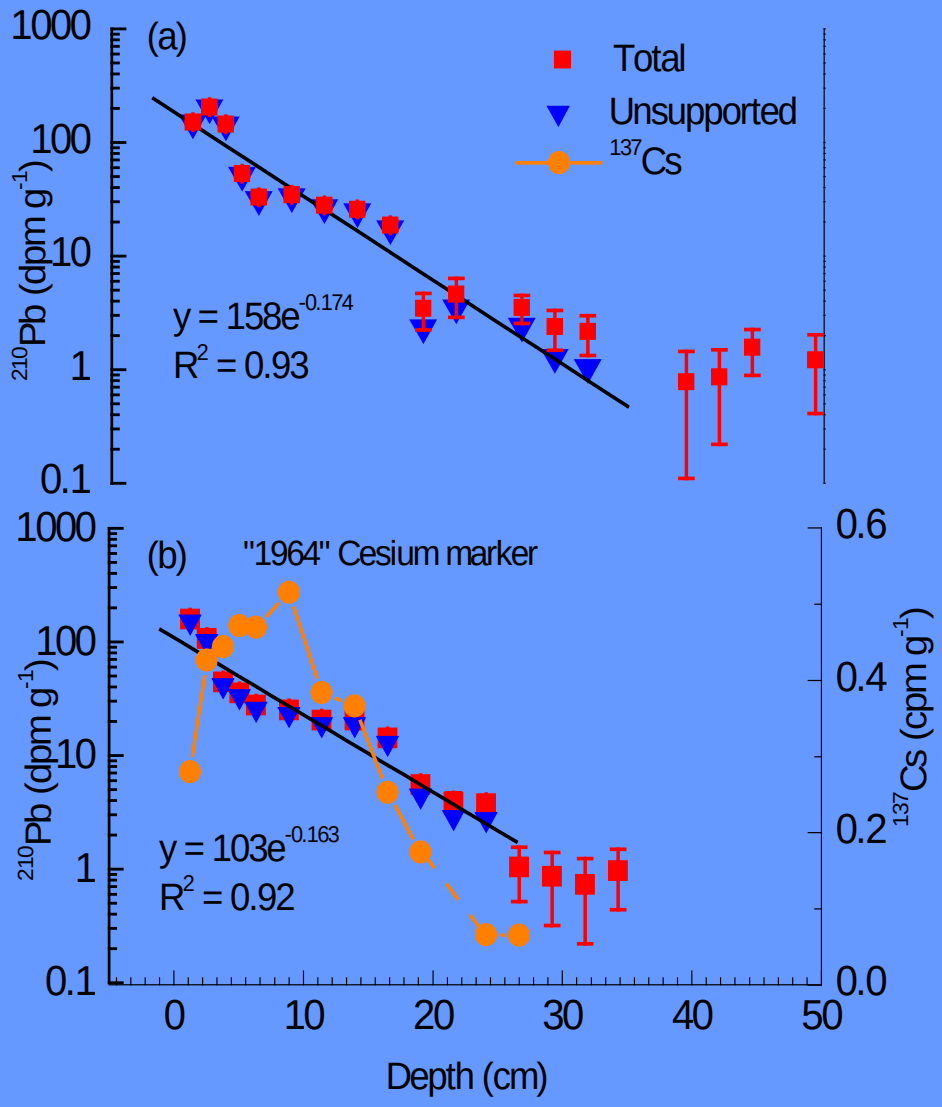
Latitude	44.334 N
Longitude	74.424 W
County	Franklin
Elevation	484 m
Surface Area	25.5 ha
Flushing rate	1.1 per year
Watershed	234.7 ha
WA/SA ratio	9.2
Maximum depth	~12 m
Mean depth	5.5 m
Sampling depth	~13 m



Adirondack Lakes Survey Corporation


upstate


upstate
technology


(a)
Total
Unsupported
137Cs
y = 158e-0.174
R2 = 0.93
210Pb (dpm g-1)
1000
100
10
1
0.1
(b)
"1964" Cesium marker
y = 103e-0.163
R2 = 0.92
137Cs (cpm g-1)
0.6
0.4
0.2
0.0
0
10
20
30
40
50
Depth (cm)

Separation of EC from Sediment

1) 3M HCl at 60 ^{o}C overnight to remove carbonates;

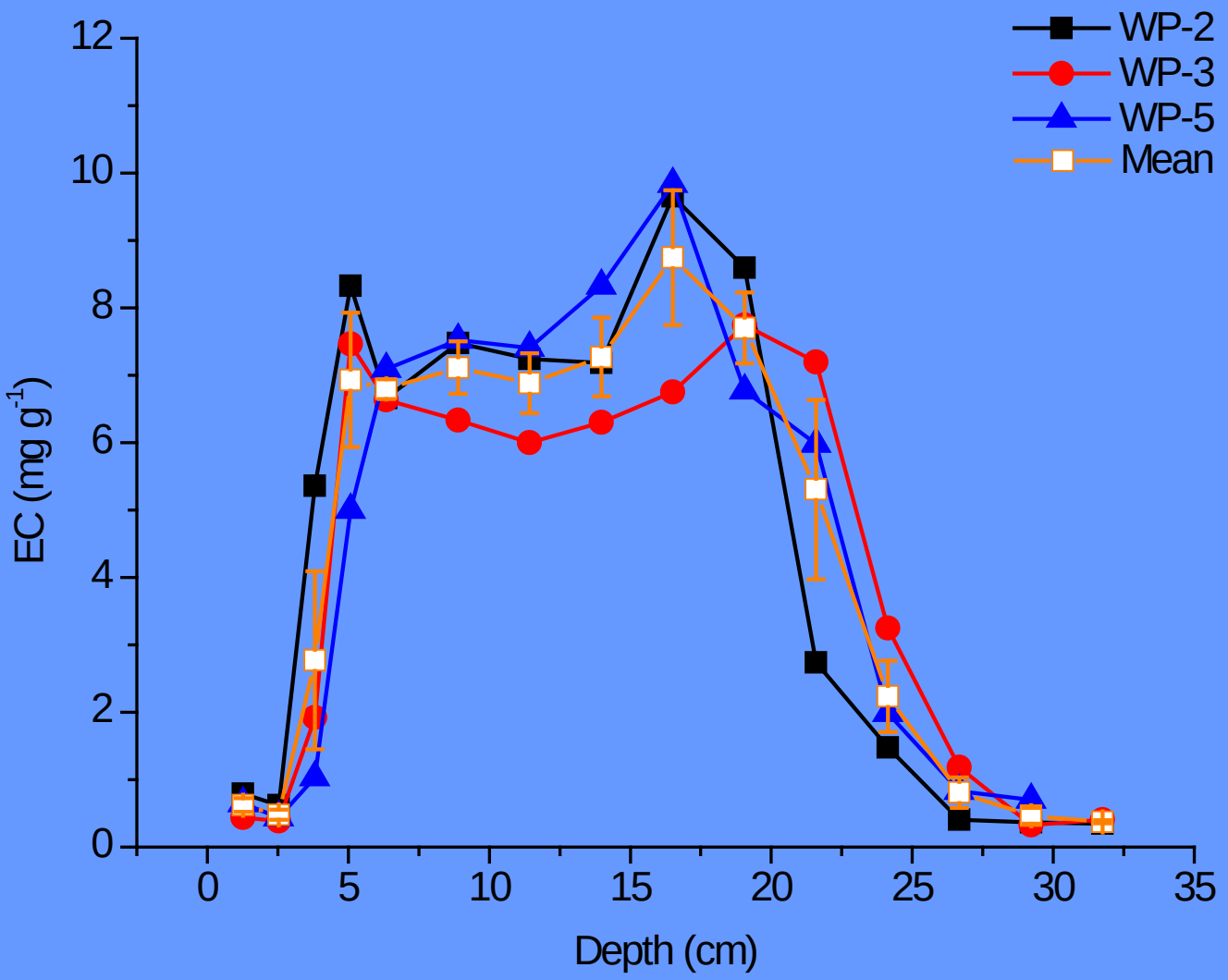
2) Heated with 22M HF and 6M HCl in an evaporating dish on steam bath until dryness to remove silicates;

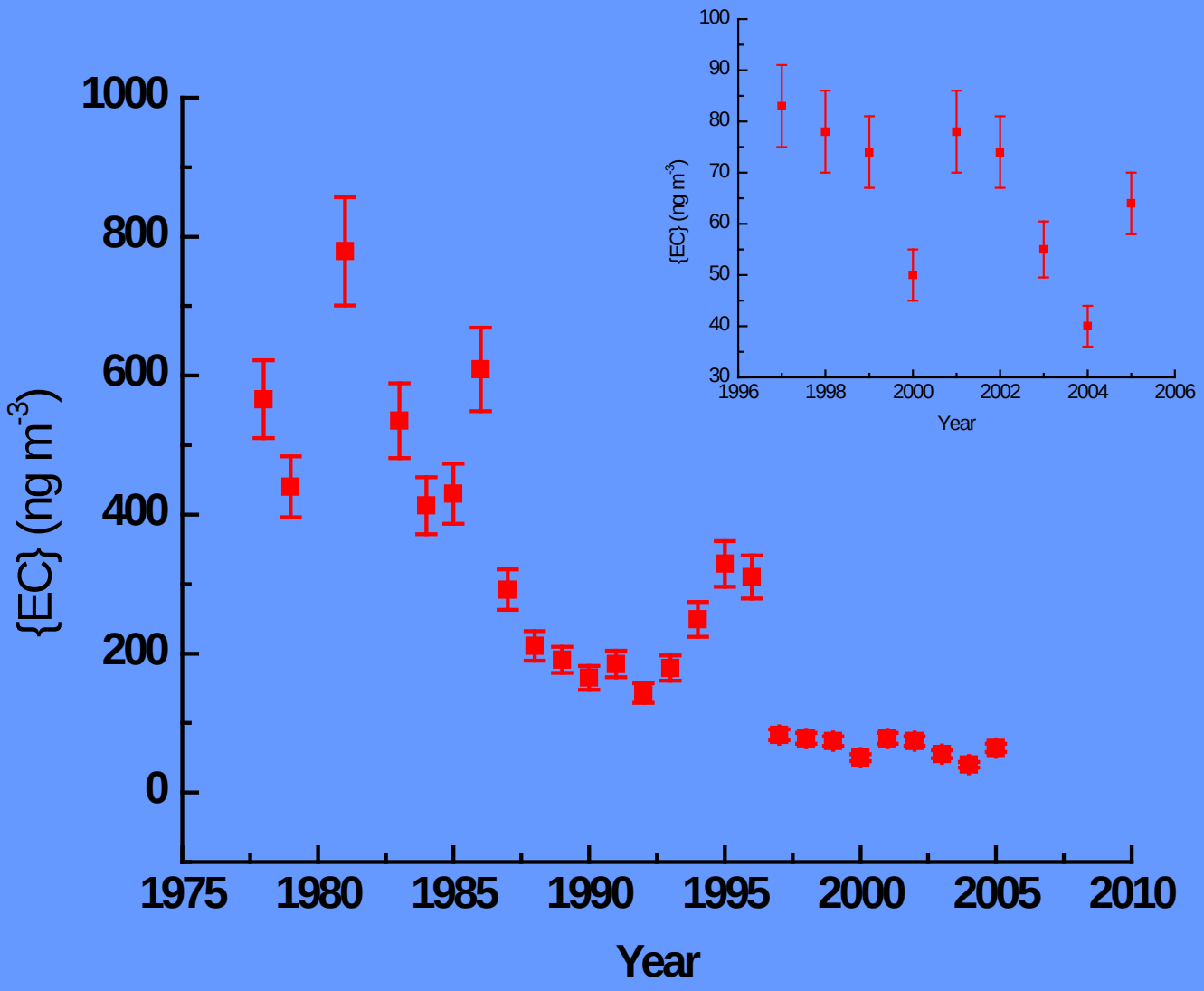
3) Heated at 60 ^{o}C overnight in 10 M HCl to remove any remaining carbonates;

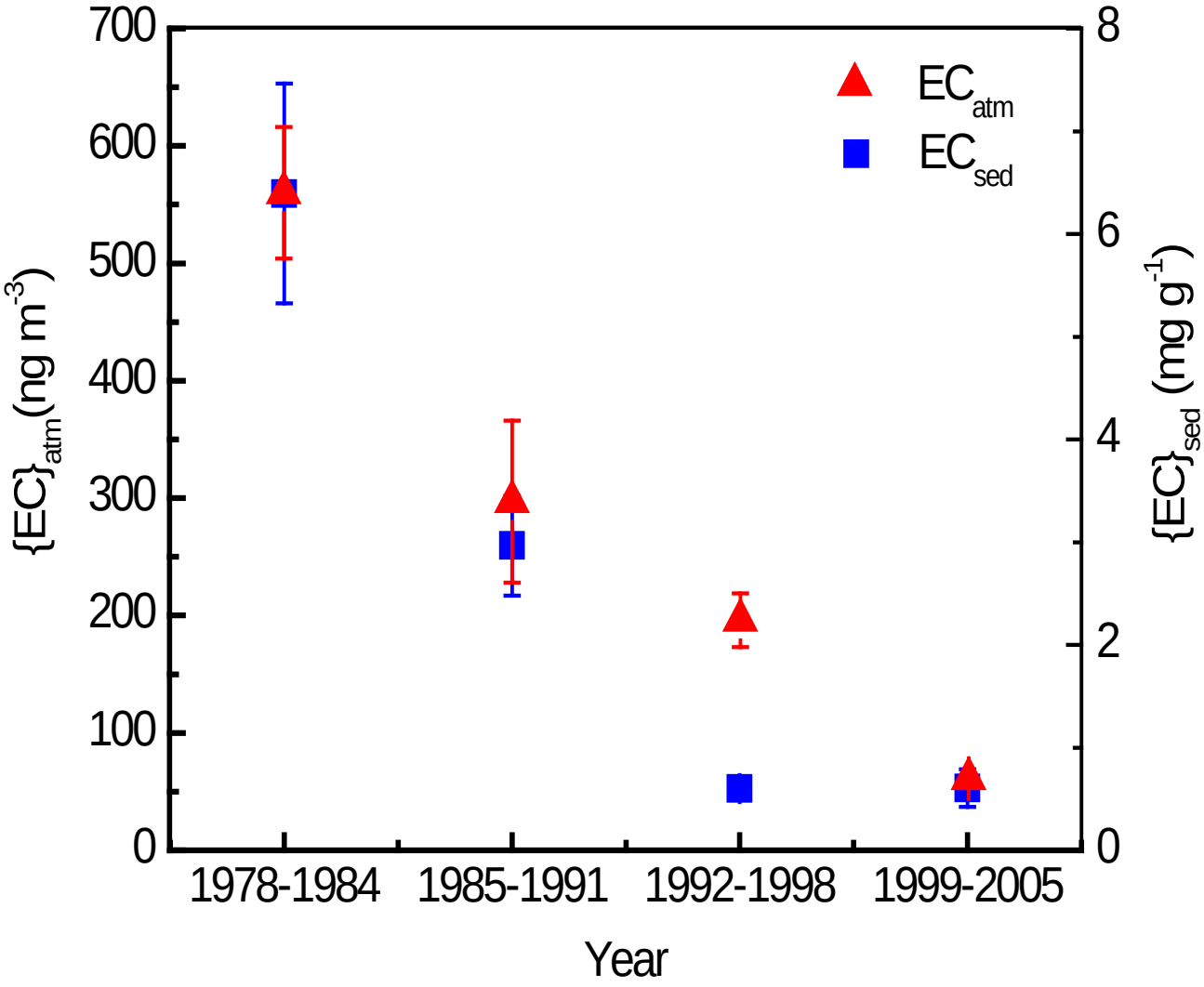
4) Heated at 60 ^{o}C overnight in 0.1M NaOH to remove humic acids;

5) Heated in 0.1M $K_2Cr_2O_7$ and 2M H_2SO_4 at 60 ^{o}C for 72 hours to remove organic carbon; and

6) Treated the residue with 70% $ZnCl_2$ and filtered the solution on quartz filter and determine EC.


WP-2
WP-3
WP-5
Mean
EC (mg g-1)
Depth (cm)
0
2
4
6
8
10
12
0
5
10
15
20
25
30
35


{EC} (ng m-3)
1000
800
600
400
200
0
1975
1980
1985
1990
1995
2000
2005
2010
Year
{EC} (ng m-3)
100
90
80
70
60
50
40
30
1996
1998
2000
2002
2004
2006
Year

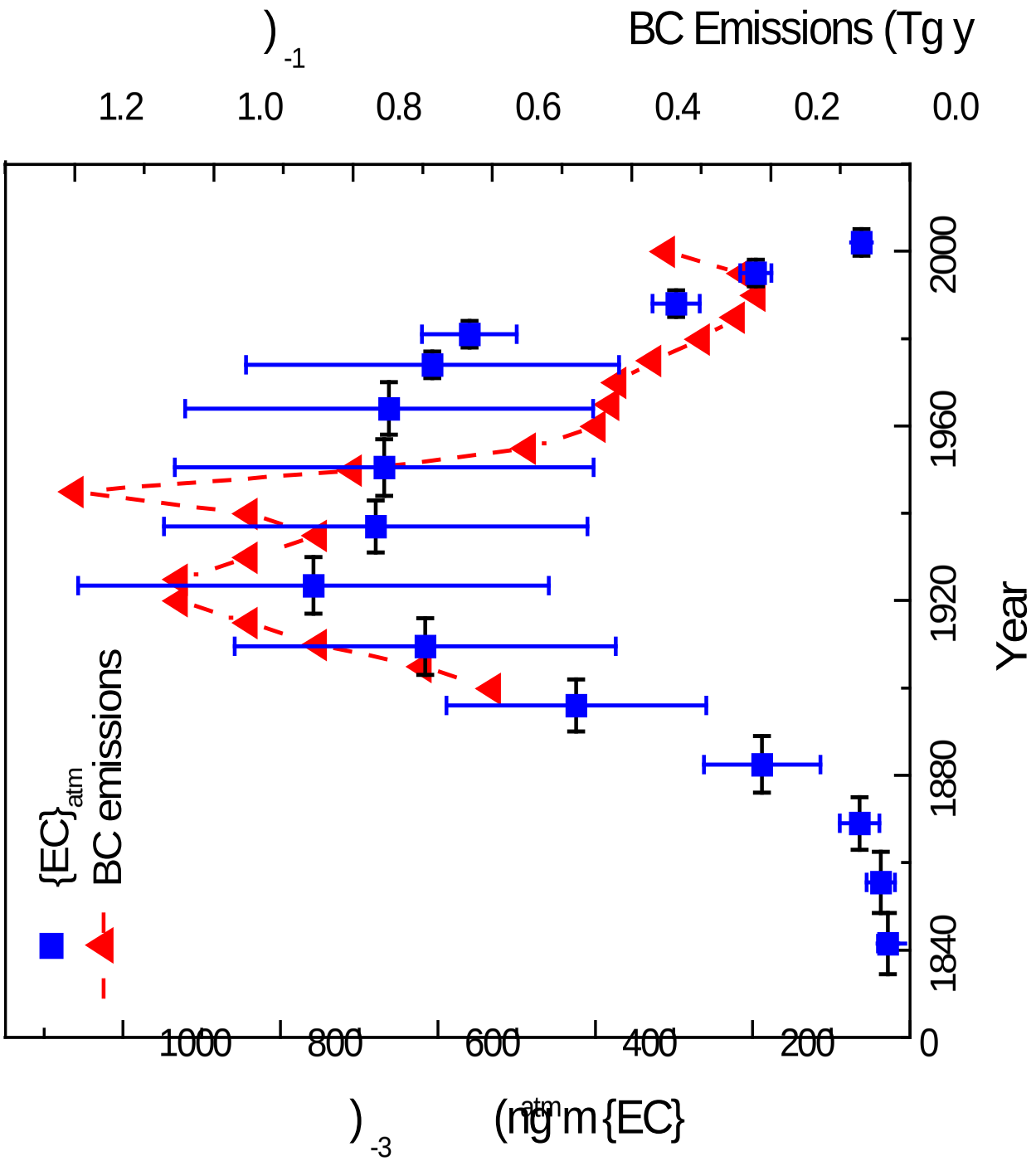

700
600
500
400
300
200
100
0
8
6
4
2
0
{EC}atm (ng m-3)
{EC}sed (mg g-1)
ECatm
ECsed
1978-1984
1985-1991
1992-1998
1999-2005
Year

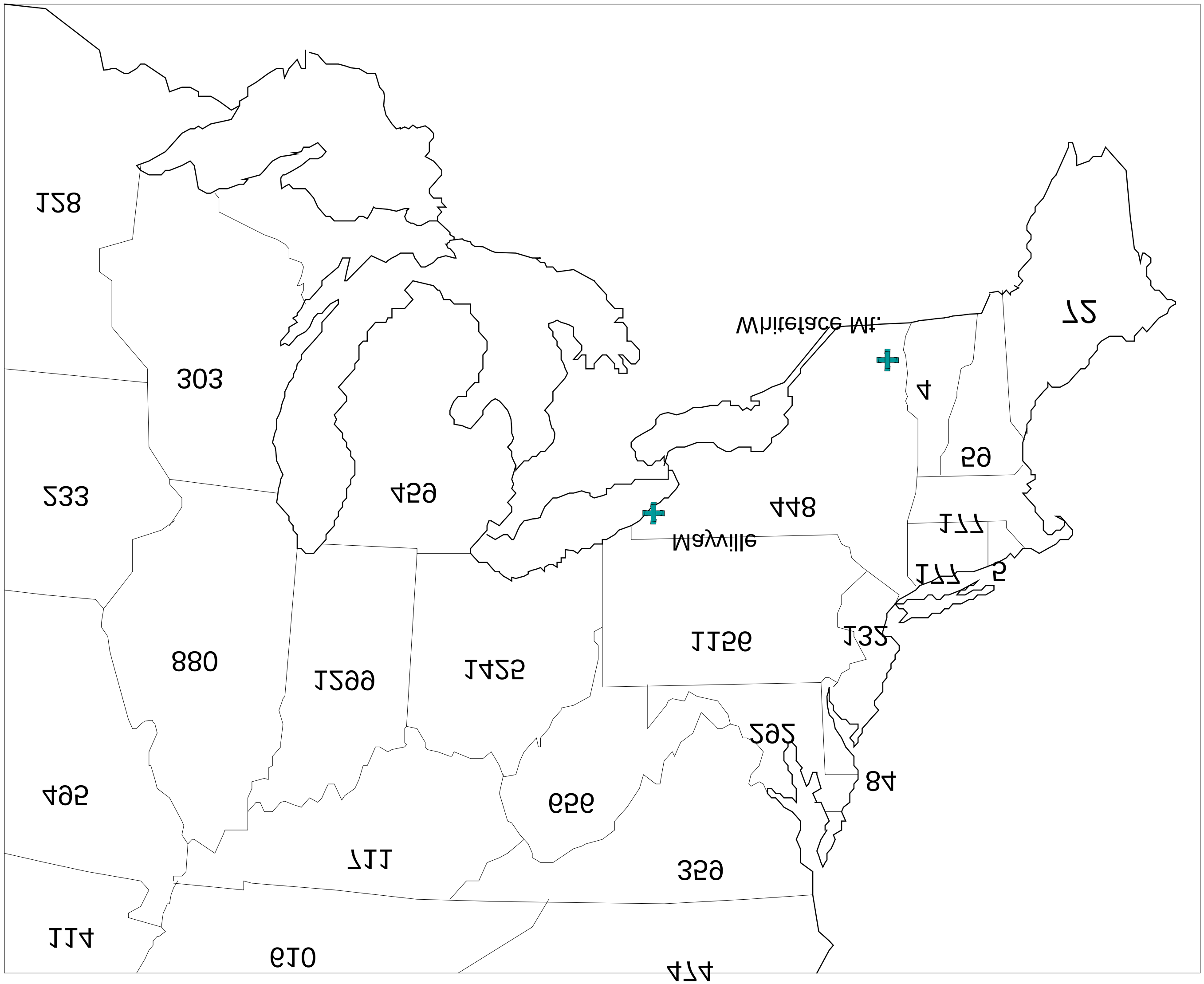
- For EC in sediment

$$\{EC\}_{sed} = (K)\ \{EC\}_{atm}$$

- Where K depends upon
  - drainage or run-off
  - wet deposition of EC at lake surface
  - mixing and sedimentation processes

- Measurements be made on samples integrated over several years.
- Not affected by short-term meteorological factors.


BC Emissions (Tg y
)
-1
1.2
1.0
0.8
0.6
0.4
0.2
0.0
2000
1960
1920
1880
1840
Year
{EC}atm
BC emissions
1000
800
600
400
200
0
(ng m
atm
{EC}
)
-3



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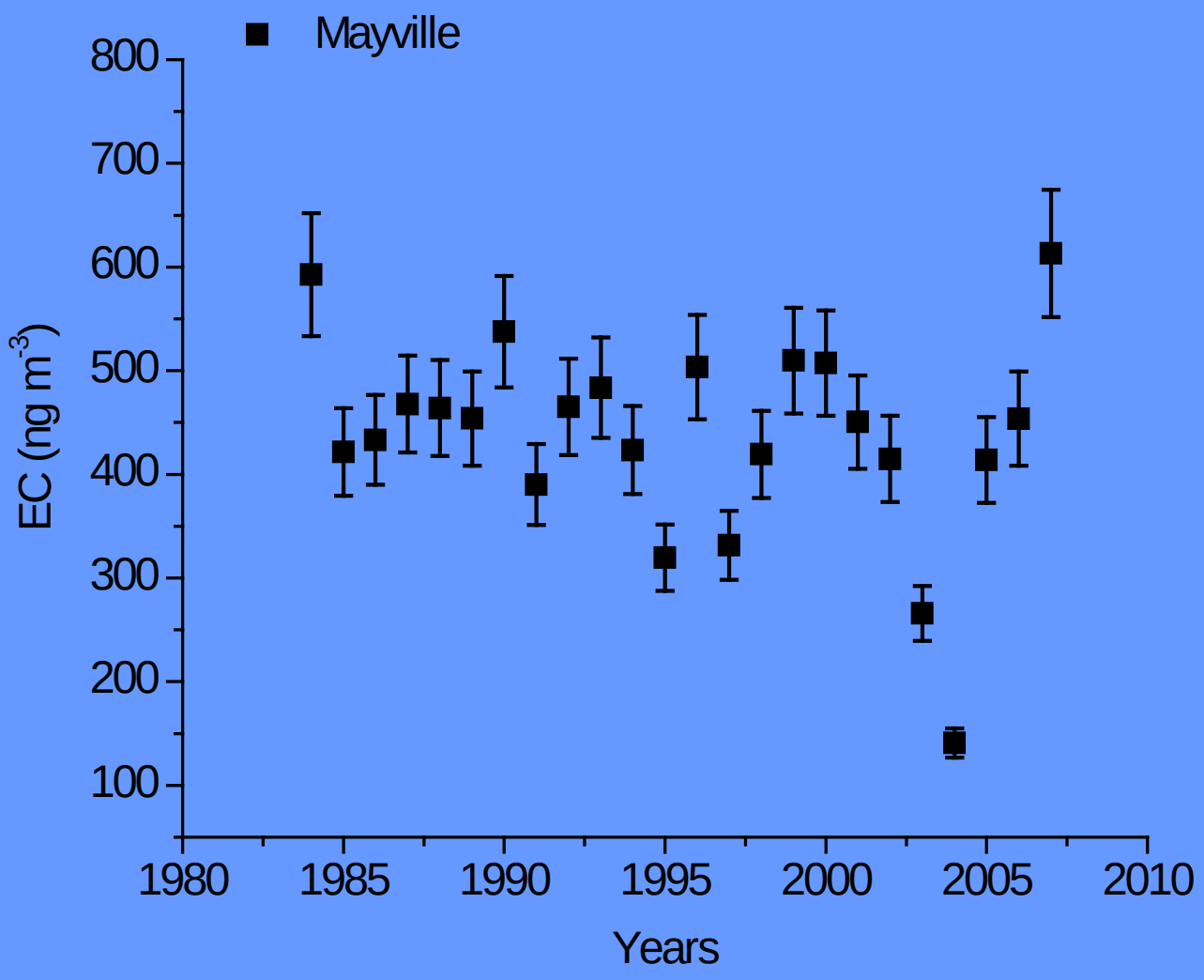
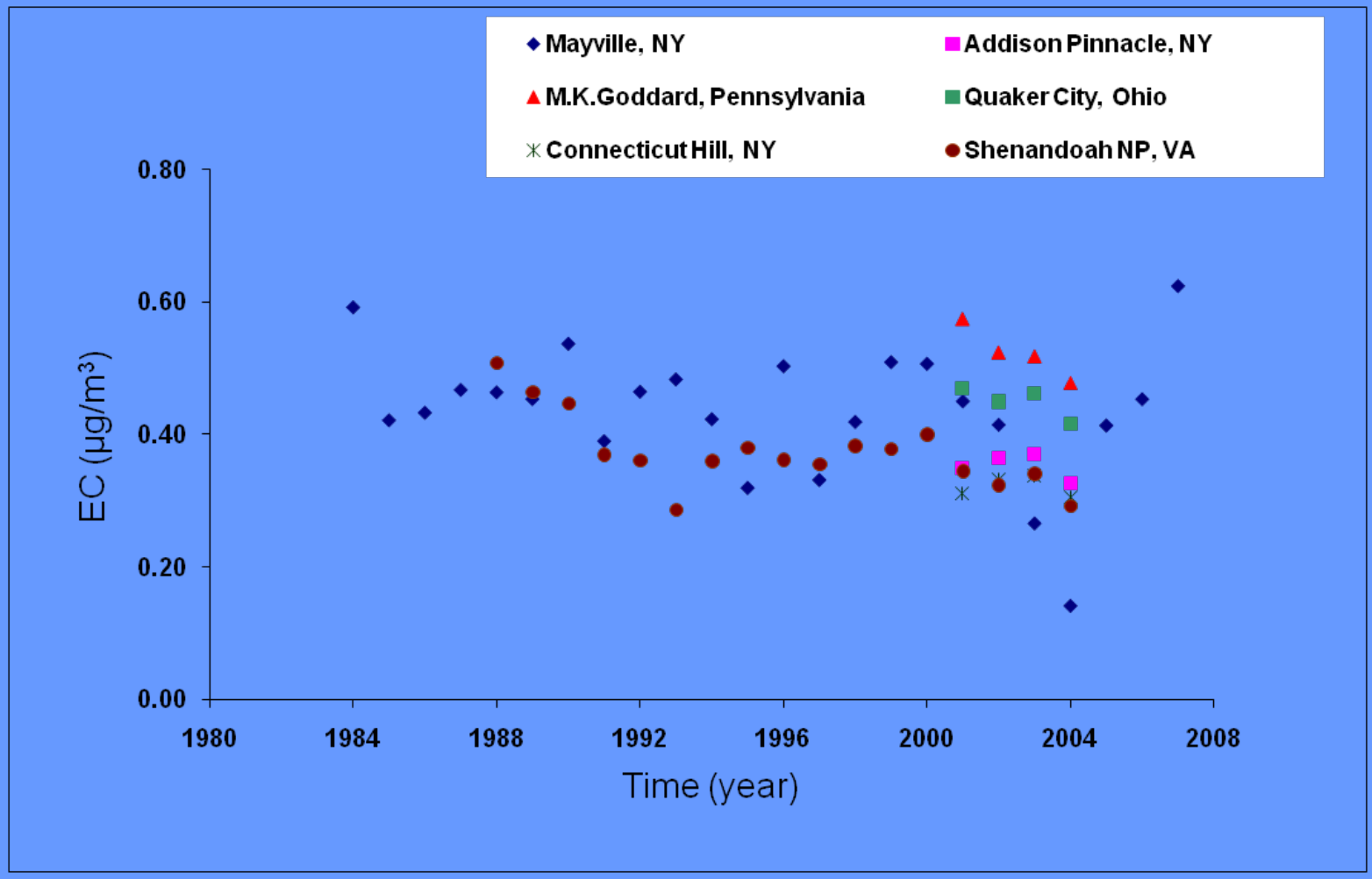

Mayville
800
700
600
500
400
300
200
100
EC (ng m-3)
1980
1985
1990
1995
2000
2005
2010
Years


Mayville, NY
Addison Pinnacle, NY
M.K.Goddard, Pennsylvania
Quaker City, Ohio
Connecticut Hill, NY
Shenandoah NP, VA
0.80
0.60
0.40
0.20
0.00
EC (µg/m³)
1980
1984
1988
1992
1996
2000
2004
2008
Time (year)

Conclusions

- At Whiteface Mountain, the annual mean {EC} were ~500 $ng.m^{-3}$ around 1980, decreased to about ~150 $ng.m^{-3}$ and are now ~ 50 $ng.m^{-3}$. The {EC} higher in summer, lowest in winter.

- The {EC} in the lake sediments mimicked the atmospheric concentrations.

- The {EC} in the aerosols can be used to calibrate EC deposition in the lakes.

conclusions contd….

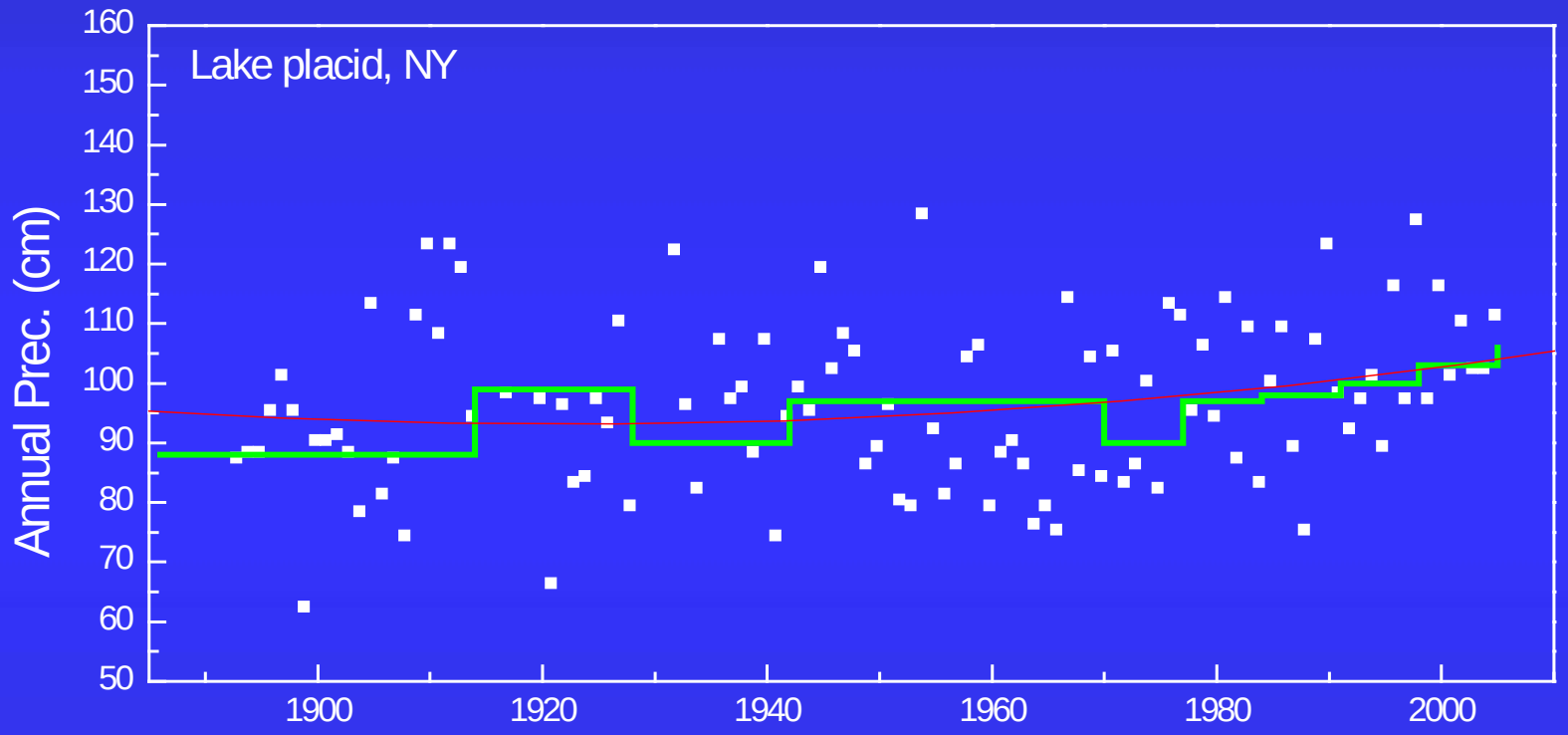
- The EC concentration pattern, as determined from the lake sediments, is quite similar to the pattern of BC emissions from ~1900 to about 1940 as calculated by Novakov's based on energy consumptions but differs significantly from ~1950 to the present.

- The {EC} showed no significant decrease at Mayville from 1983 to present.

- Thank you.

- You can contact me at:
- lhusain@albany.edu

Trend in Precipitation



Prec. data from National Climatic Data Center

^{210} Pb Dating

The unsupported $[^{210}Pb]_{un}$ activity (dpm/g) decreases exponentially with depth.

The age of each section of the core was determined from

$$([^{210}Pb]_{un})_t = ([^{210}Pb]_{un})_0 \, e^{-\lambda t}$$

The unsupported ^{210}Pb activity, (dpm/g), in a layer at depth z(cm), is expressed as:

$$([^{210}Pb]_{un})_z = ([^{210}Pb]_{un})_0 \, e^{-\lambda \, Z/\omega}$$

where ω and is the sedimentation rate (cm y^{-1}).

Method:

Sample Preparation:

1. The freeze-dried samples were weighed, ground to a fine powder, homogenized, and stored in 5-cm diameter plastic jars for gamma counting and EC analyses.

Separation of EC from Sediment

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3) Heated at 60 ^{o}C overnight in 10 M HCl to remove any remaining carbonates;

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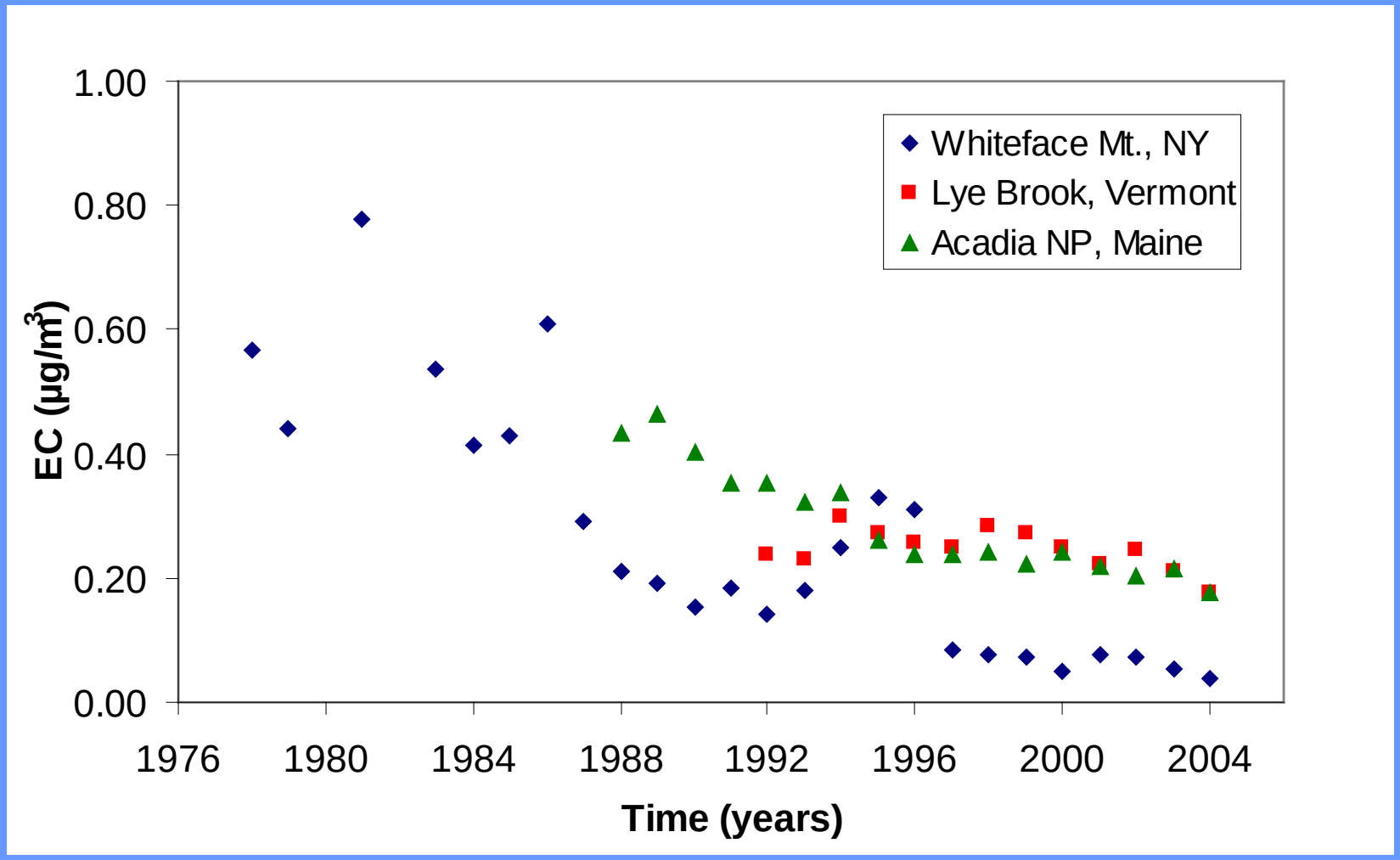
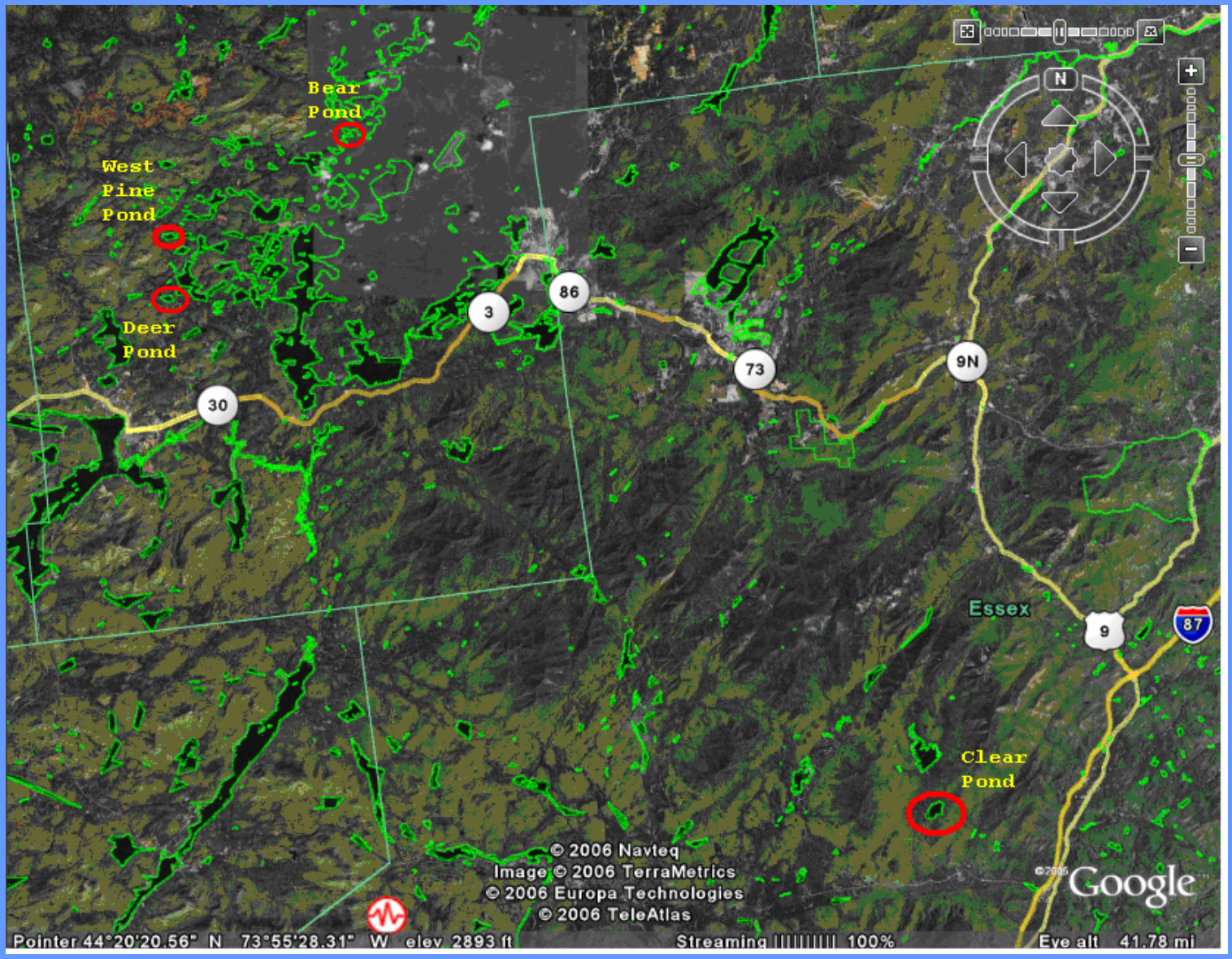
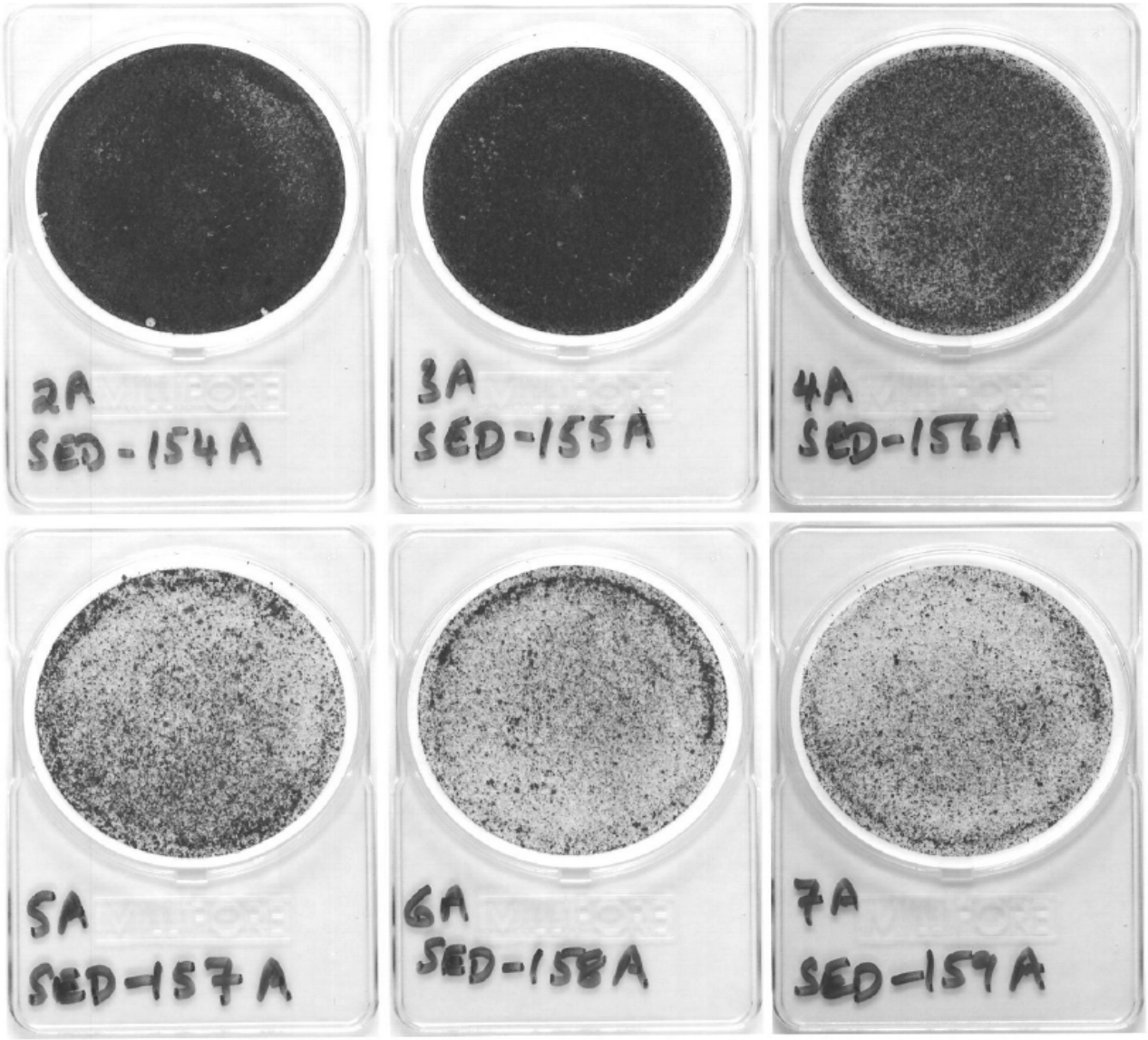
6) Treated the residue with 70% $ZnCl_2$ and filtered the solution on quartz filter and determine EC.

Month	1	2	3	4	5	6	7	8	9	10	11	12	Annual Mean	SD
1989	0.119	0.157	0.184	0.123	0.234	0.200	0.260	0.199	0.271	0.233	0.189	0.128	**0.191**	**0.052**
1990	0.103	0.093	0.133	0.162	0.149	0.265	0.192	0.234			0.160	0.130	**0.162**	**0.055**
1991	0.172	0.141	0.166	0.205	0.205	0.131	0.196		0.234	0.309	0.152	0.124	**0.185**	**0.054**
1992	0.131	0.147	0.137	0.154	0.210	0.199	0.146	0.224		0.091	0.064	0.073	**0.143**	**0.053**
1993	0.078	0.099	0.101	0.122	0.228	0.191	0.203	0.209	0.271	0.265	0.213	0.170	**0.179**	**0.065**
1994	0.194				0.284	0.276	0.256	0.196	0.195	0.189	0.300	0.355	**0.249**	**0.060**
1995	0.243	0.520	0.377	0.231	0.254	0.475	0.340	0.157	0.178	0.581		0.262	**0.329**	**0.143**
1996	0.348	0.235	0.372	0.230	0.582	0.658	0.403	0.376	0.297	0.116	0.042	0.062	**0.310**	**0.190**
1997	0.061	0.080	0.051	0.079	0.102	0.129	0.129	0.114	0.079	0.088	0.045	0.043	**0.083**	**0.030**
1998	0.039	0.037	0.084	0.080	0.117	0.060	0.096	0.122	0.127	0.064	0.030	0.075	**0.078**	**0.033**
1999	0.040	0.071	0.035	0.051	0.062	0.081	0.080	0.094	0.107	0.107	0.083	0.044	**0.071**	**0.025**
2000	0.031	0.048	0.036	0.033	0.061	0.077	0.058	0.050	0.072	0.084	0.018	0.037	**0.050**	**0.020**
2001	0.035	0.045	0.037	0.095	0.080	0.116	0.096	0.099	0.094	0.108	0.090	0.042	**0.078**	**0.030**
2002	0.040	0.075	0.078	0.058	0.068	0.096	0.105	0.123	0.105		0.042	0.025	**0.074**	**0.031**
2003	0.026	0.054	0.059	0.051	0.055	0.092	0.053	0.040	0.043	0.051	0.094	0.044	**0.055**	**0.020**
2004	0.025	0.030	0.050	0.044	0.050	0.050	0.045	0.068			0.028	0.012	**0.040**	**0.016**
2005	0.036	0.049	0.037	0.083	0.053	0.115						0.036	**0.058**	**0.030**


1.00
0.80
0.60
0.40
0.20
0.00
EC (µg/m3)
1976
1980
1984
1988
1992
1996
2000
2004
Time (years)
Whiteface Mt., NY
Lye Brook, Vermont
Acadia NP, Maine


Bear Pond
West Pine Pond
Deer Pond
86
3
73
9N
30
Essex
9
87
Clear Pond
© 2006 Navteq
Image © 2006 TerraMetrics
© 2006 Europa Technologies
© 2006 TeleAtlas
Google
Pointer 44°20'20.56" N 73°55'28.31" W elev 2893 ft
Streaming 100%
Eye alt 41.78 mi


2A
SED-154A
3A
SED-155A
4A
SED-156A
5A
SED-157A
6A
SED-158A
7A
SED-159A

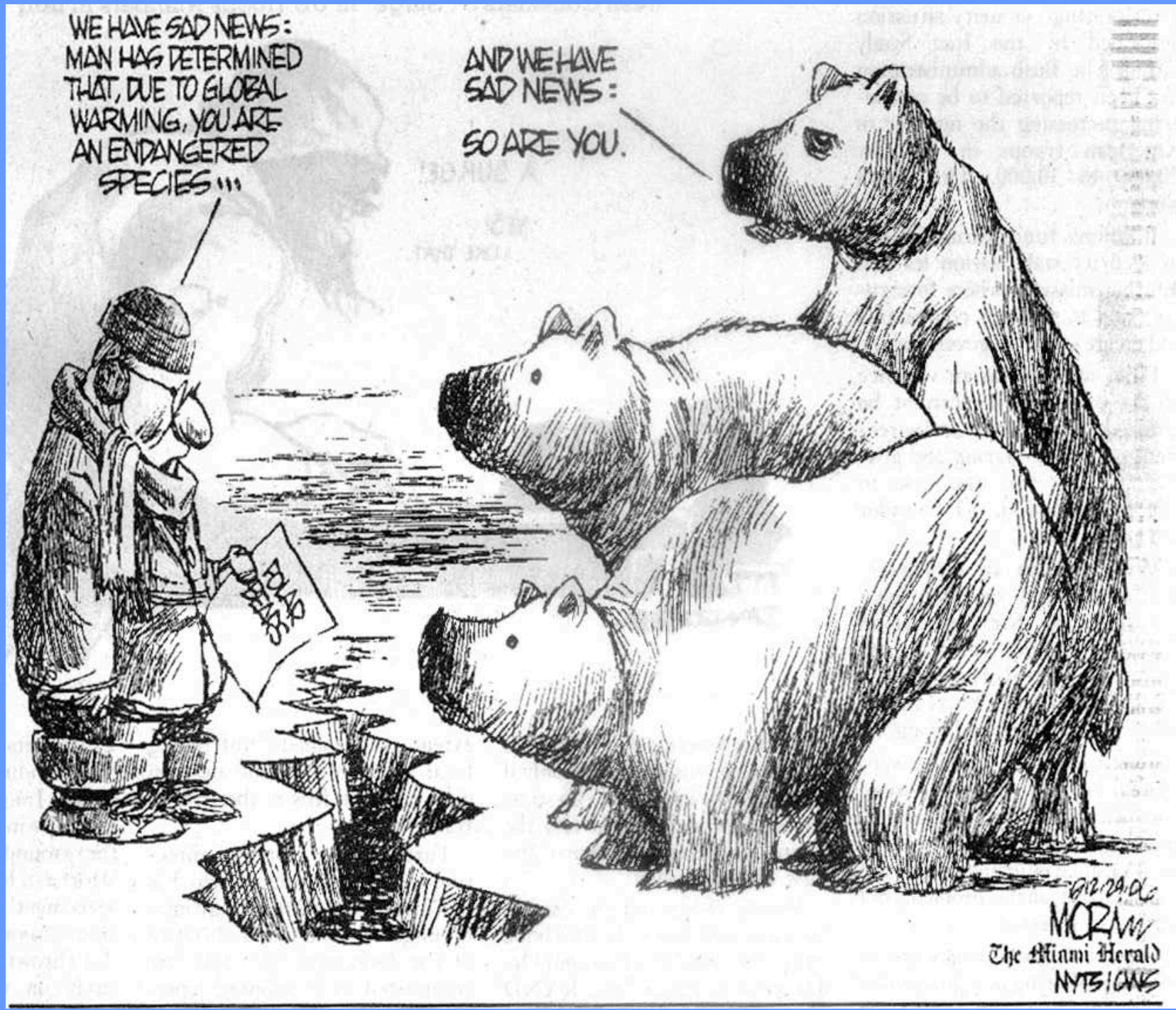
Terminology

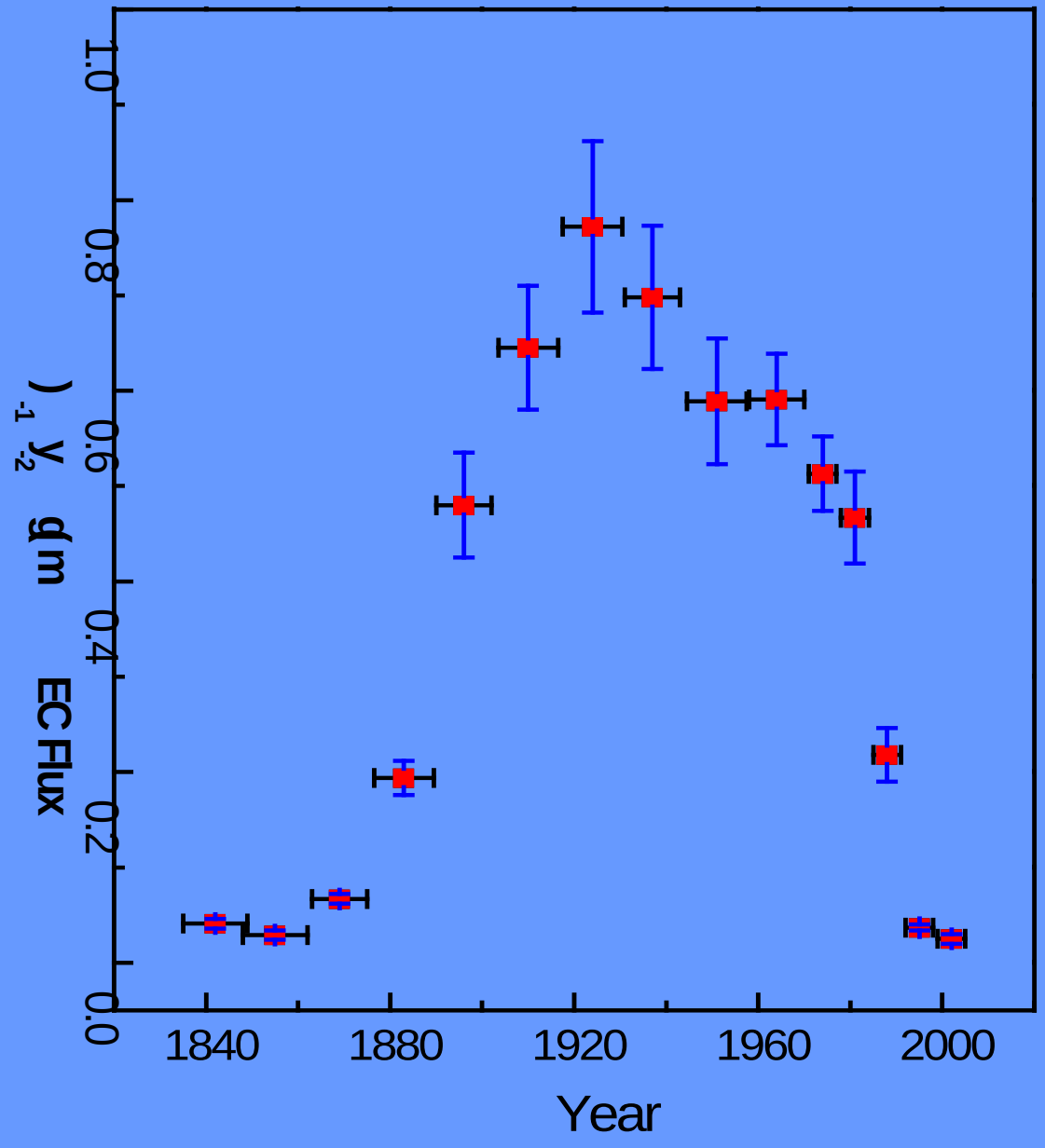
The term elemental carbon (EC), refers to carbon in aerosols with graphitic structure, insoluble in organic solvents and resistant to oxidation at temperatures below about 400°C.

The term soot, EC, and black carbon are often used interchangeably.

- **Radiative forcing calculations require data that represents regions.**

- **Do Mayville and Whiteface Mountain collect regionally representative EC aerosols ?**


WE HAVE SAD NEWS: MAN HAS DETERMINED THAT, DUE TO GLOBAL WARMING, YOU ARE AN ENDANGERED SPECIES!!!
AND WE HAVE SAD NEWS:
SO ARE YOU.
BOARD MEMBERS
MCRM
The Miami Herald
NYTSICNS


EC Flux
) -1 y -2 g m
0.0
0.2
0.4
0.6
0.8
1.0
1840
1880
1920
1960
2000
Year

Recovery of Atmospheric EC Data Deposited in Lake Sediments for the Past ~150 Years

Dry and wet deposition in term of EC flux into the sediment can be expressed as [Seinfield and Pandis, 1998] :

For dry EC deposition:

$F_d = -V_d \{EC\}_{atm}$ …… (1)

where, F_d is the dry deposition flux, V_d is the dry deposition velocity, and $\{EC\}_{atm}$ is the concentration of EC.

For wet deposition,

Fw = {EC}precip (x,y,0,t) po ……(2)

For wet deposition,

$F_w = \{EC\}_{precip}\ (x,y,0,t)\ p_o$ ……(2)

where $\{EC\}_{precip}$ (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).

The wash out ratio w_r relates EC concentrations in the atmosphere and in precipitation:

$w_r = \{EC\}_{precip}\ (x,y,0,t) / \{EC\}_{atm}\ (x,y,0,t)$.....(3)

Which can be substituted in (2)

$F_w = \{EC\}_{atm}\ (x,y,0,t)\ w_r\ p_o$ ……(4)

Equation (1) and (2) can be combined to define the total flux F_T :

$F_T = -V_d \{EC\}_{atm} + \{EC\}_{atm}(x,y,0,t) W_r P_o = [-v_d + w_r p_o] \{EC\}_{atm}$ .....(5)

$F_T = K_1 \{EC\}_{atm}$ ……(6)

For EC in sediment

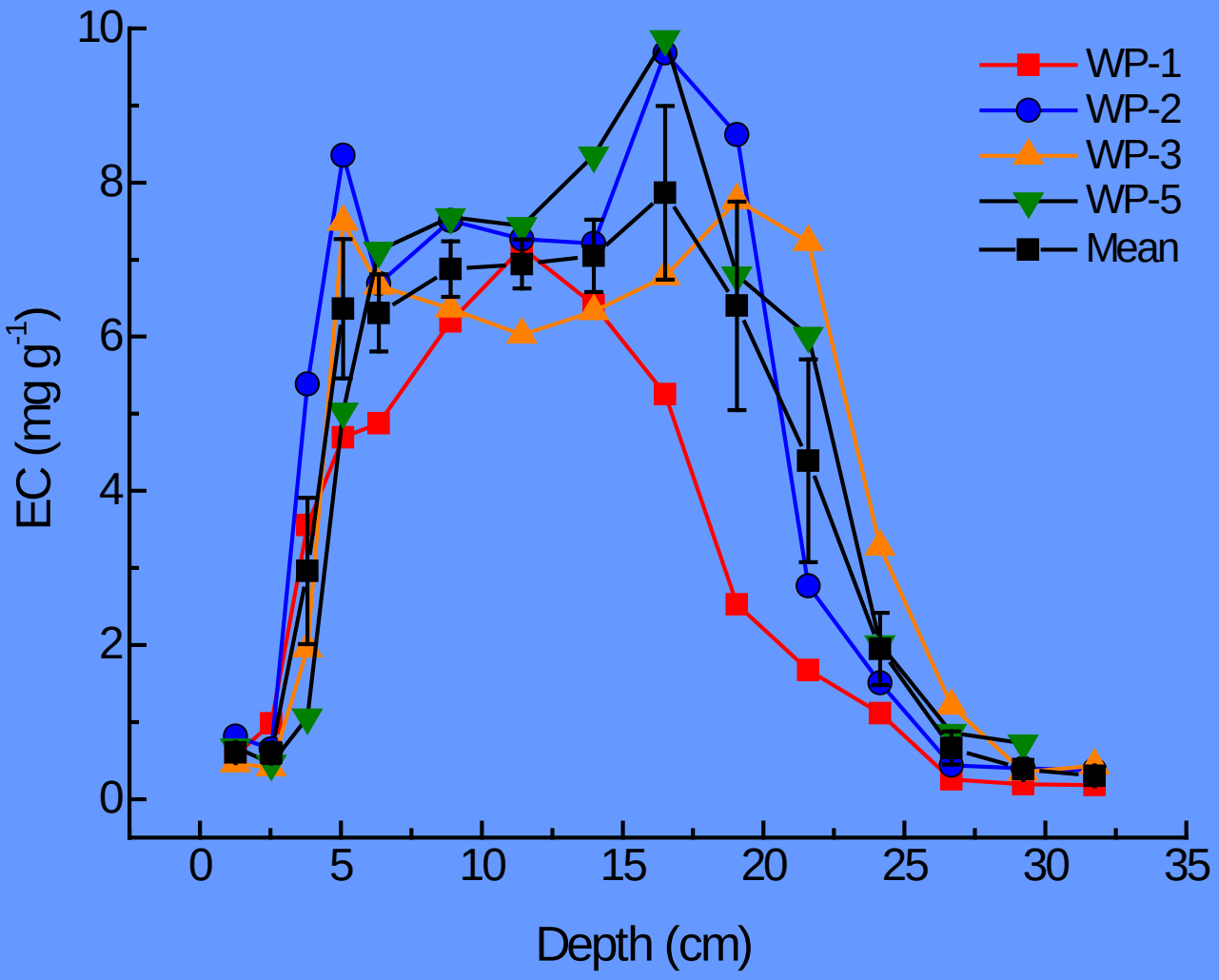
$\{EC\}_{sed} = (K_1+K_2) \{EC\}_{atm}$

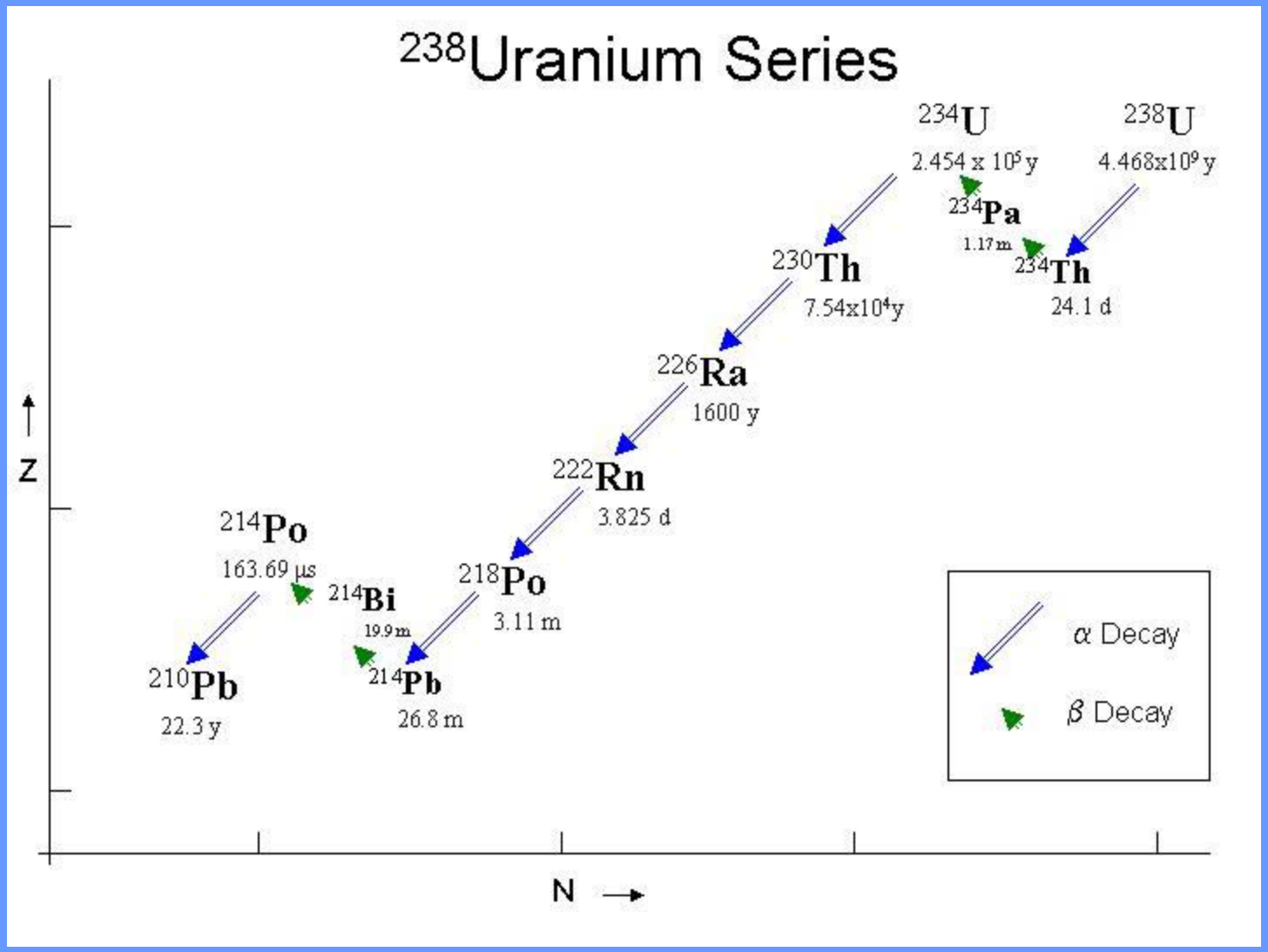
Where K_2 is another constant related to drainage or run-off deposition of EC and the Sedimentary processes including sediment focusing,

$$\mathbf{EC_{sed} = K\ EC_{atm},}$$

where K depends upon the dry and wet deposition of EC at lake surface, mixing and sedimentation processes within the lake.

The slow rate of sedimentation requires that measurements be made on samples integrated over several years. Hence, K is assumed to be a constant and not affected by short-term meteorological factors.


EC (mg g-1)
Depth (cm)
WP-1
WP-2
WP-3
WP-5
Mean
0
2
4
6
8
10
0
5
10
15
20
25
30
35


238Uranium Series
234U
2.454 x 10^5 y
238U
4.468x10^9 y
234Pa
1.17 m
234Th
24.1 d
230Th
7.54x10^4y
226Ra
1600 y
222Rn
3.825 d
218Po
3.11 m
214Po
163.69 µs
214Bi
19.9m
214Pb
26.8 m
210Pb
22.3 y
Z
N
α Decay
β Decay

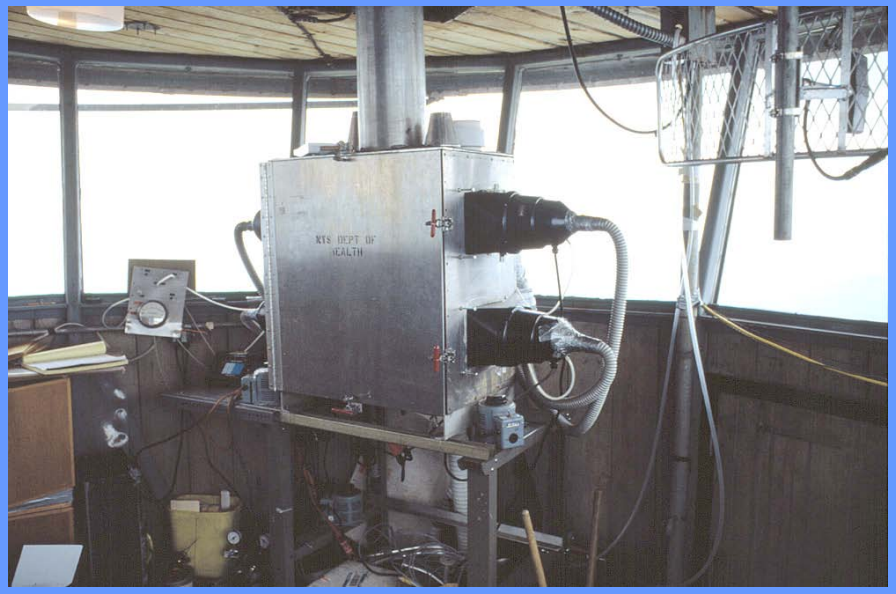
- For EC in sediment

$$\{EC\}_{sed} = (K)\ \{EC\}_{atm}$$

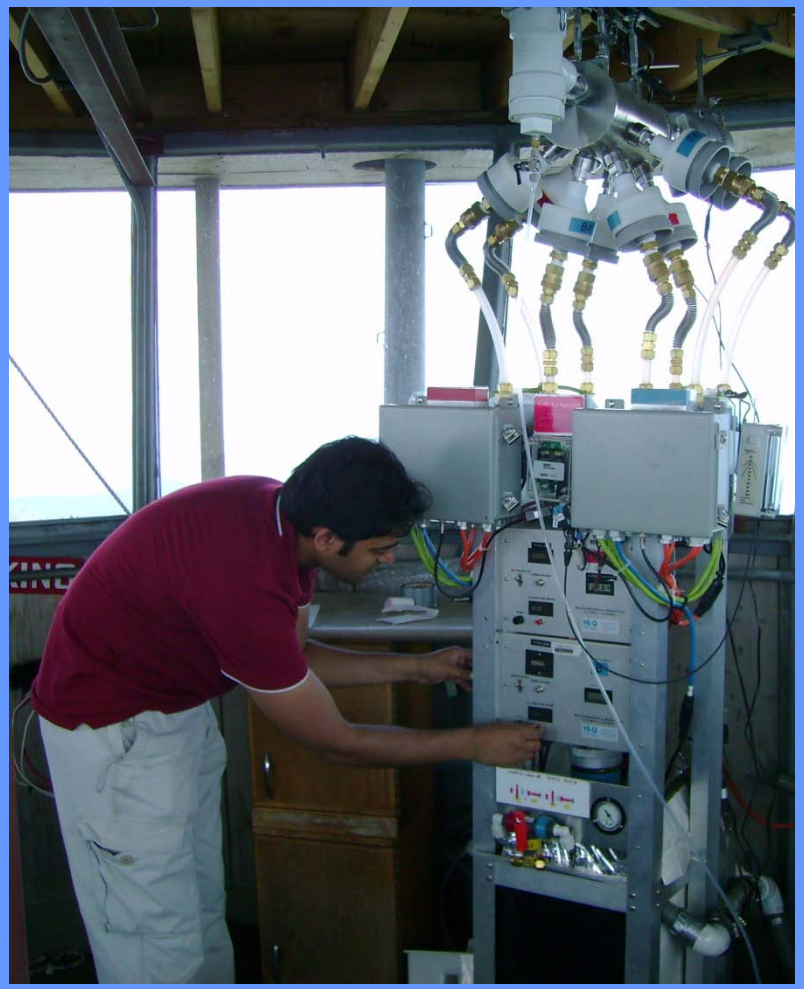
- Where K depends upon
  - drainage or run-off
  - wet deposition of EC at lake surface
  - mixing and sedimentation processes

- Measurements be made on samples integrated over several years.
- Not affected by short-term meteorological factors.

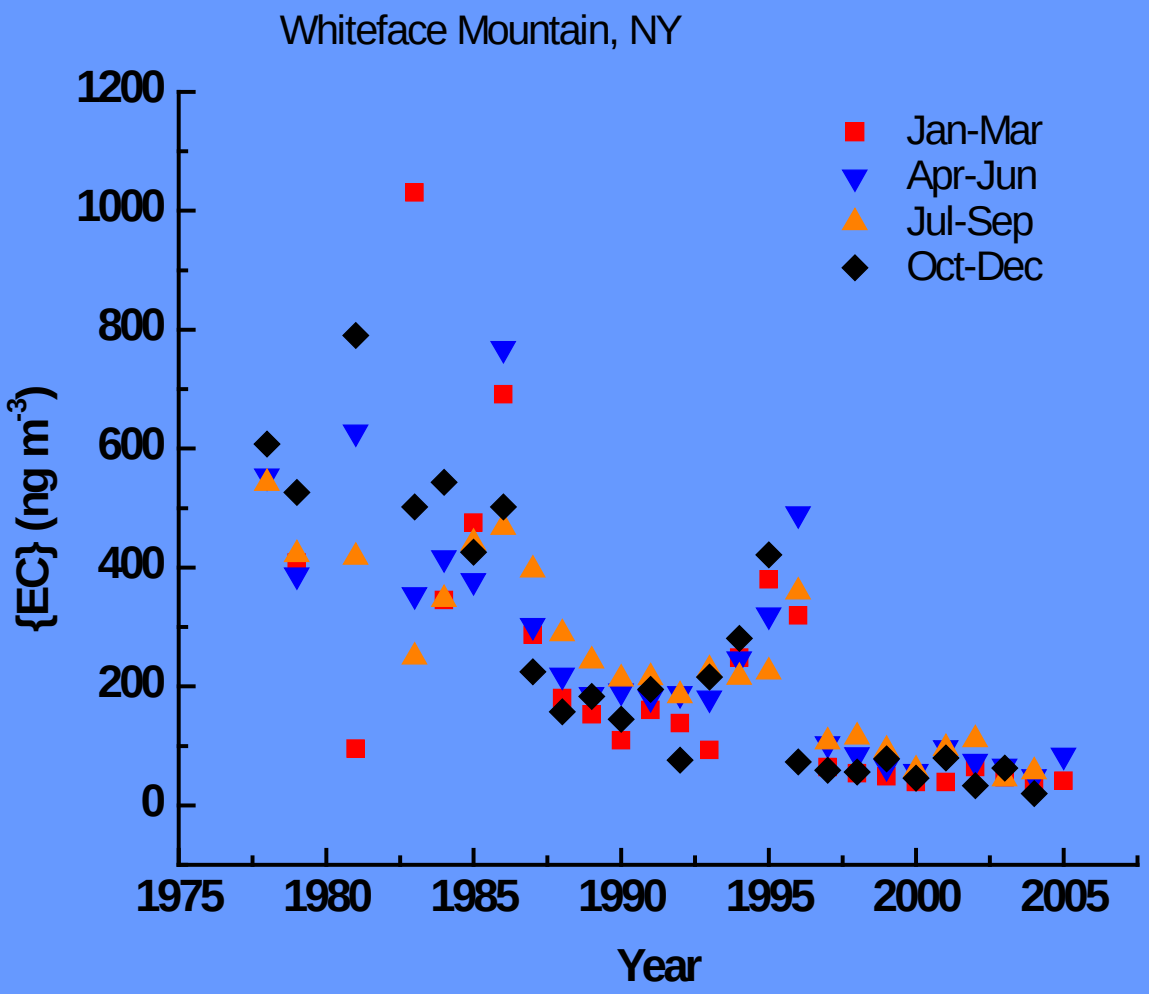
Aerosol Sampling Equipment



(1978 – 2005)



(2005 -)


Whiteface Mountain, NY
Jan-Mar
Apr-Jun
Jul-Sep
Oct-Dec
{EC} (ng m-3)
1200
1000
800
600
400
200
0
1975
1980
1985
1990
1995
2000
2005
Year