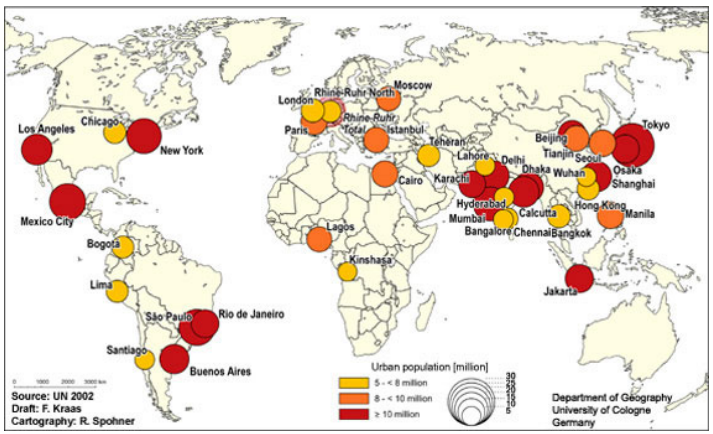
# GLOBAL CLIMATE CHANGE:

# AEROSOLS AND THEIR LINKAGES


## Jeffrey S. Gaffney

## Argonne National Laboratory

| Permanent Gases | |
|---|---|
| Nitrogen | 78.08% |
| Oxygen | 20.95% |
| Argon | 0.93% |
| Neon | 0.0018% |
| Helium | 0.0005% |
| Methane | 0.0001% |
| Hydrogen | 0.00005% |
| Xenon | 0.000009% |
| **Variable Gases** | |
| Water vapor | 0-4% |
| Carbon dioxide | 0.034% |
| Ozone | 0.000004% |
| Carbon monoxide | 0.00002% |
| Sulfur dioxide | 0.000001% |
| Nitrogen dioxide | 0.000001% |
| Dust, soot, etc. | 0.00001% |

# DOE Biological and Environmental Science
# Climate Change Research Division
# Atmospheric Science Program
# http://www.asp.bnl.gov/

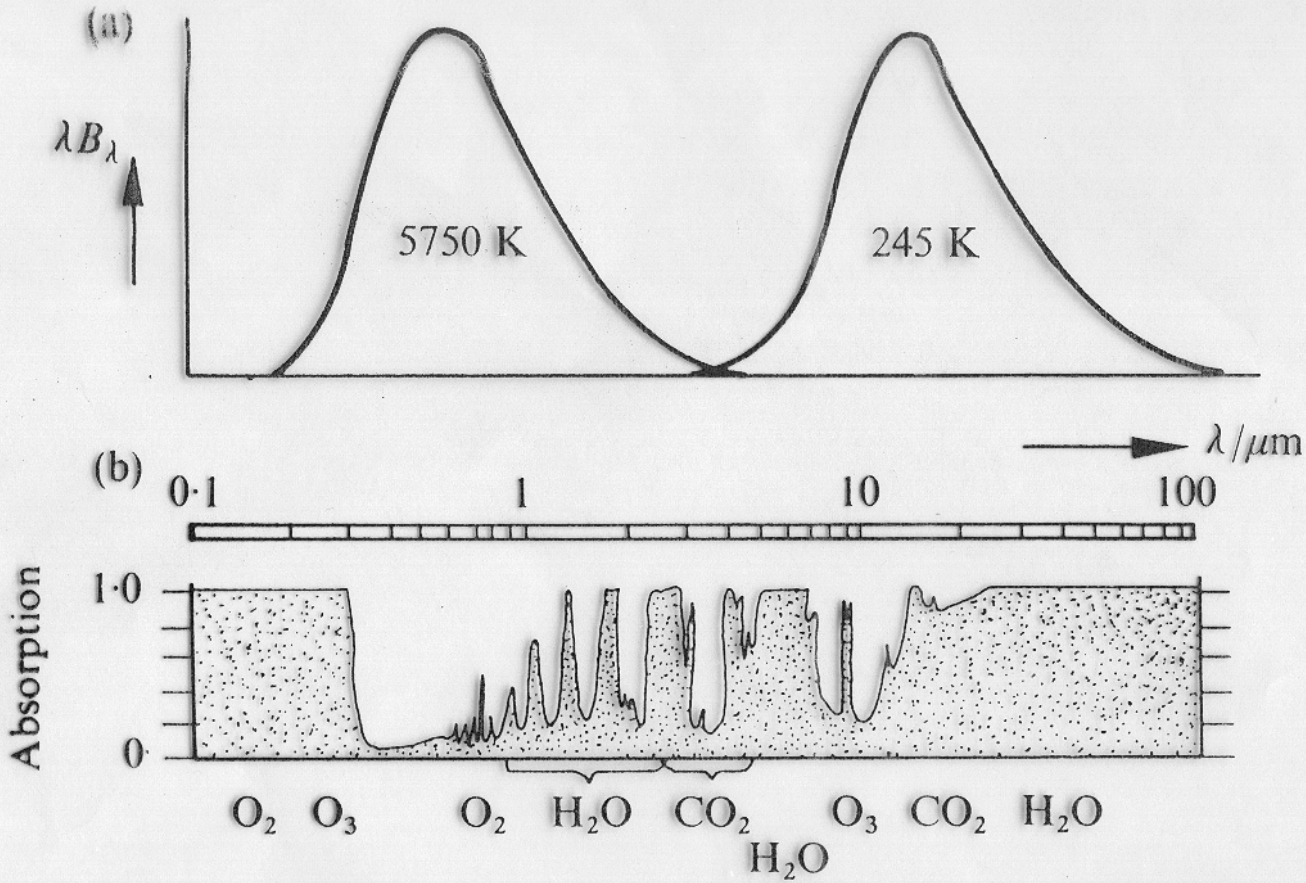



Fig. 2.1. (a) Curves of black-body energy $B_\lambda$ at wavelength $\lambda$ for 5750 K (approximating to the sun's temperature) and 245 K (approximating to the atmosphere's mean temperature). The curves have been drawn of equal areas since integrated over the earth's surface and all angles the solar and terrestrial fluxes are equal.
(b) Absorption by atmospheric gases for a clear vertical column of atmosphere. The positions of the absorption bands of the main constituents are marked.

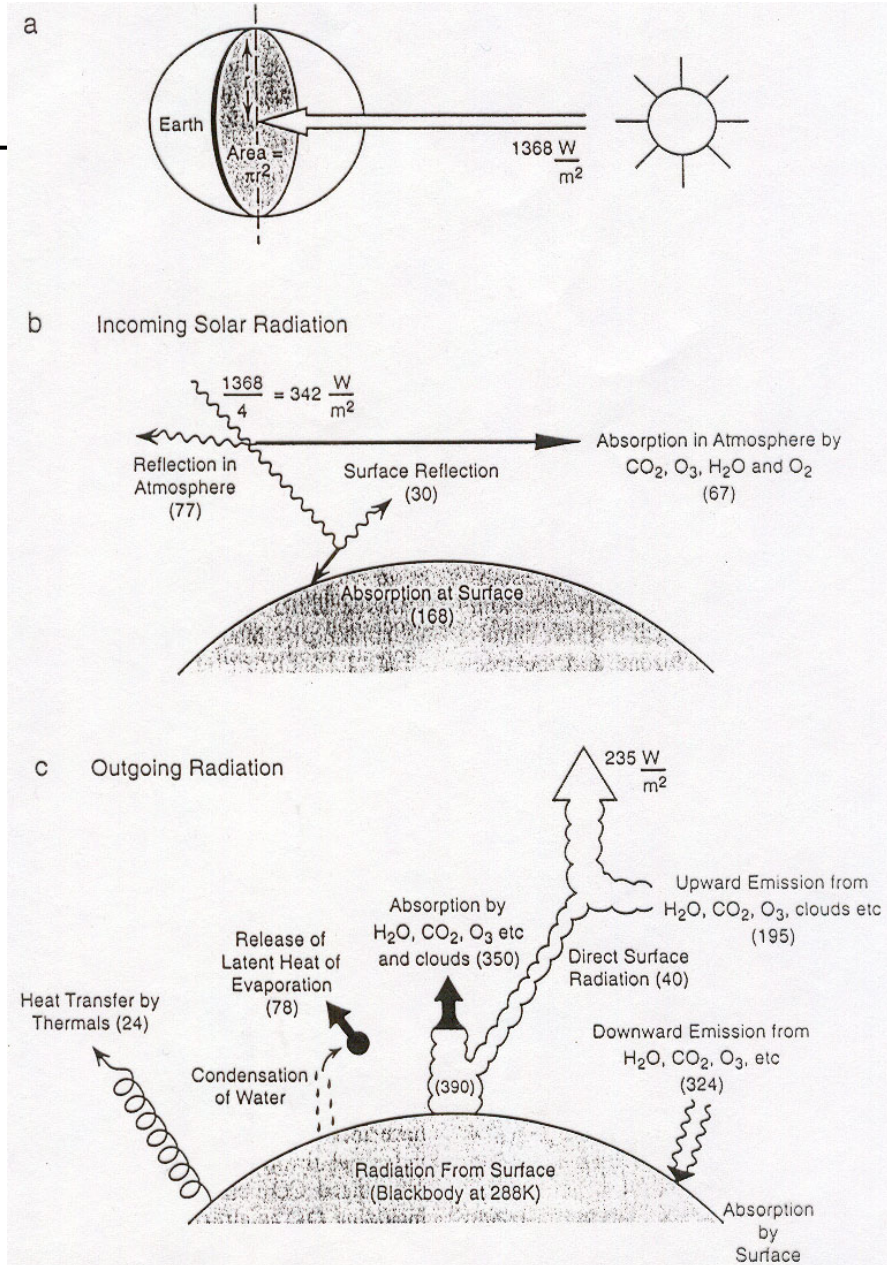
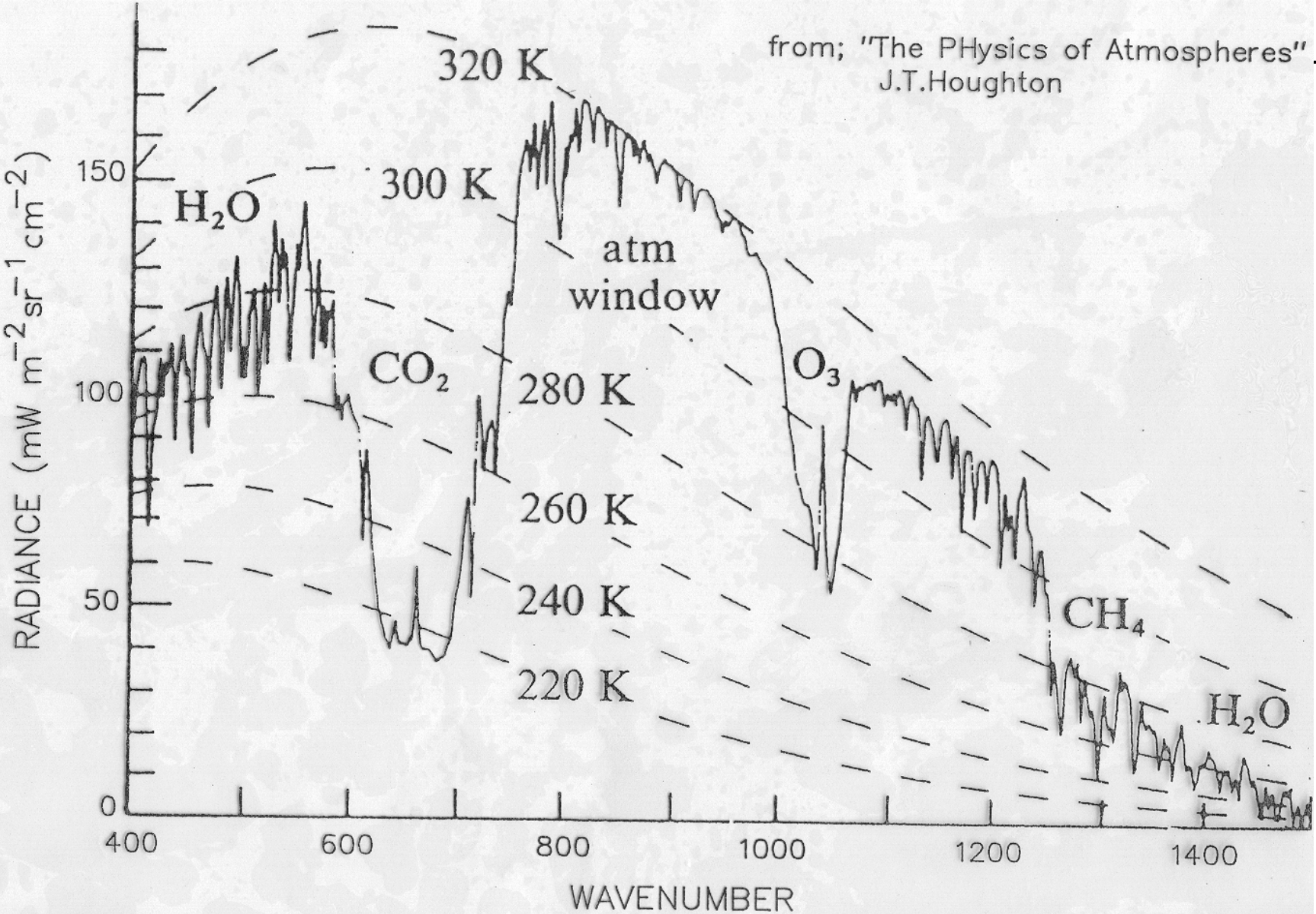
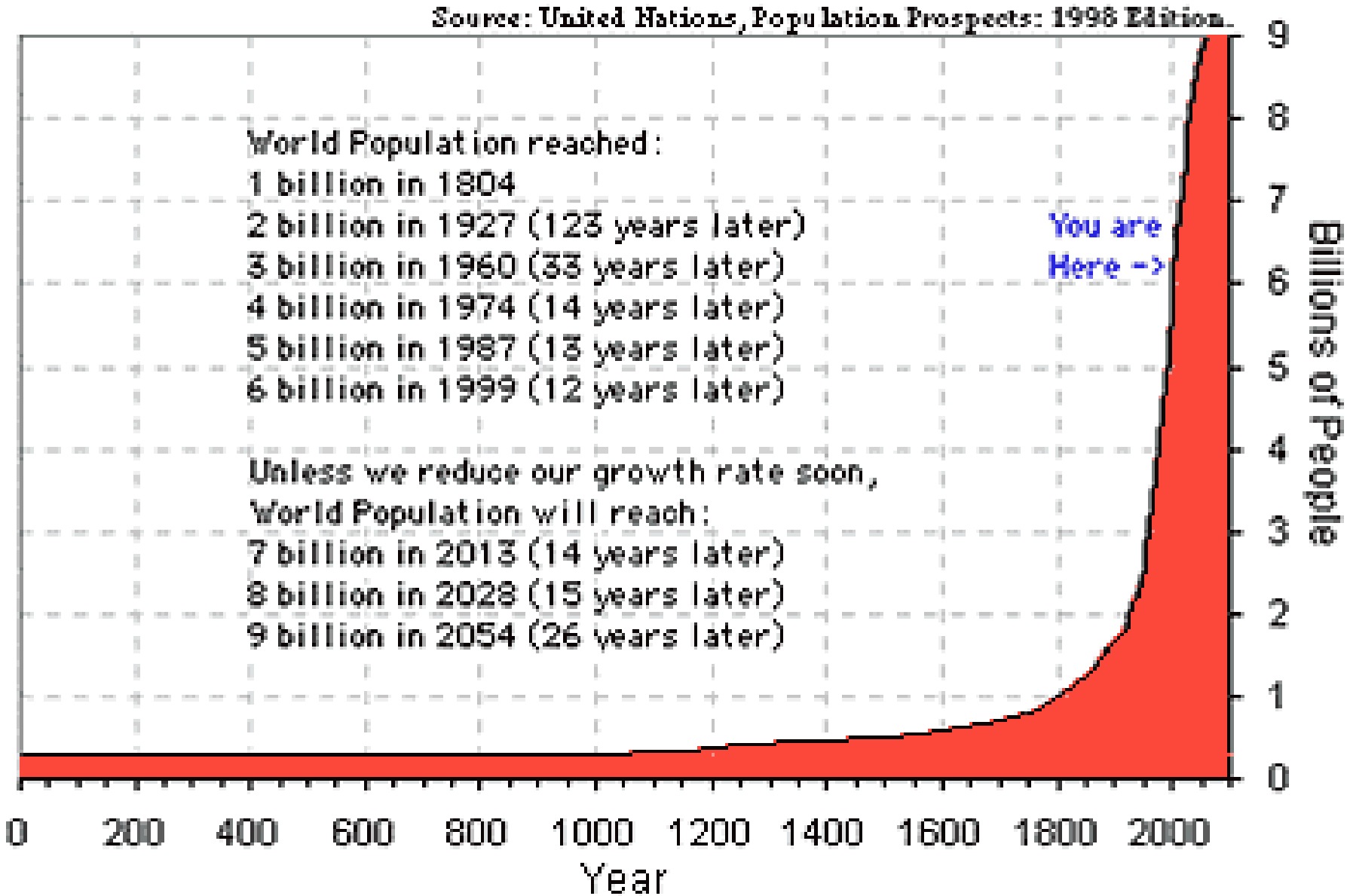



FIGURE 1.9 Global average mean radiation and energy balance per unit of earth's surface [adapted with permission from IPCC (1996) with numbers from Kiehl and Trenberth (1997)].

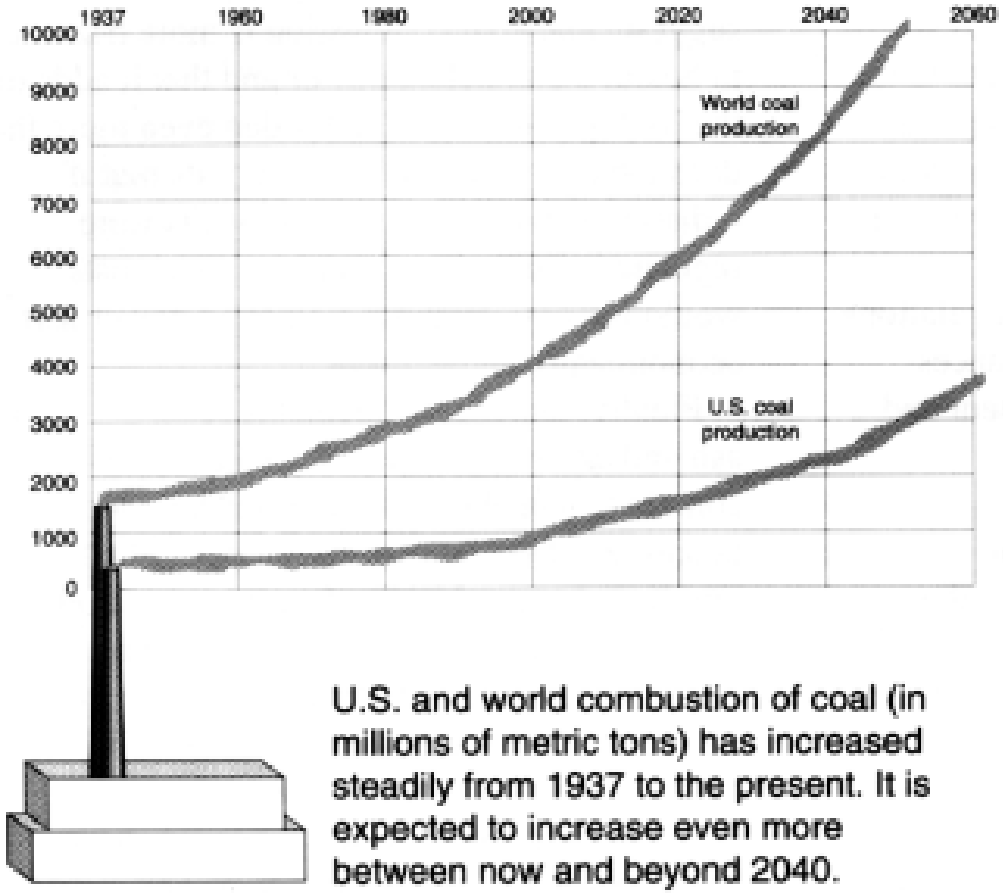
# THERMAL EMISSION FROM EARTH

Emitted vertically upwards over the Sahara.

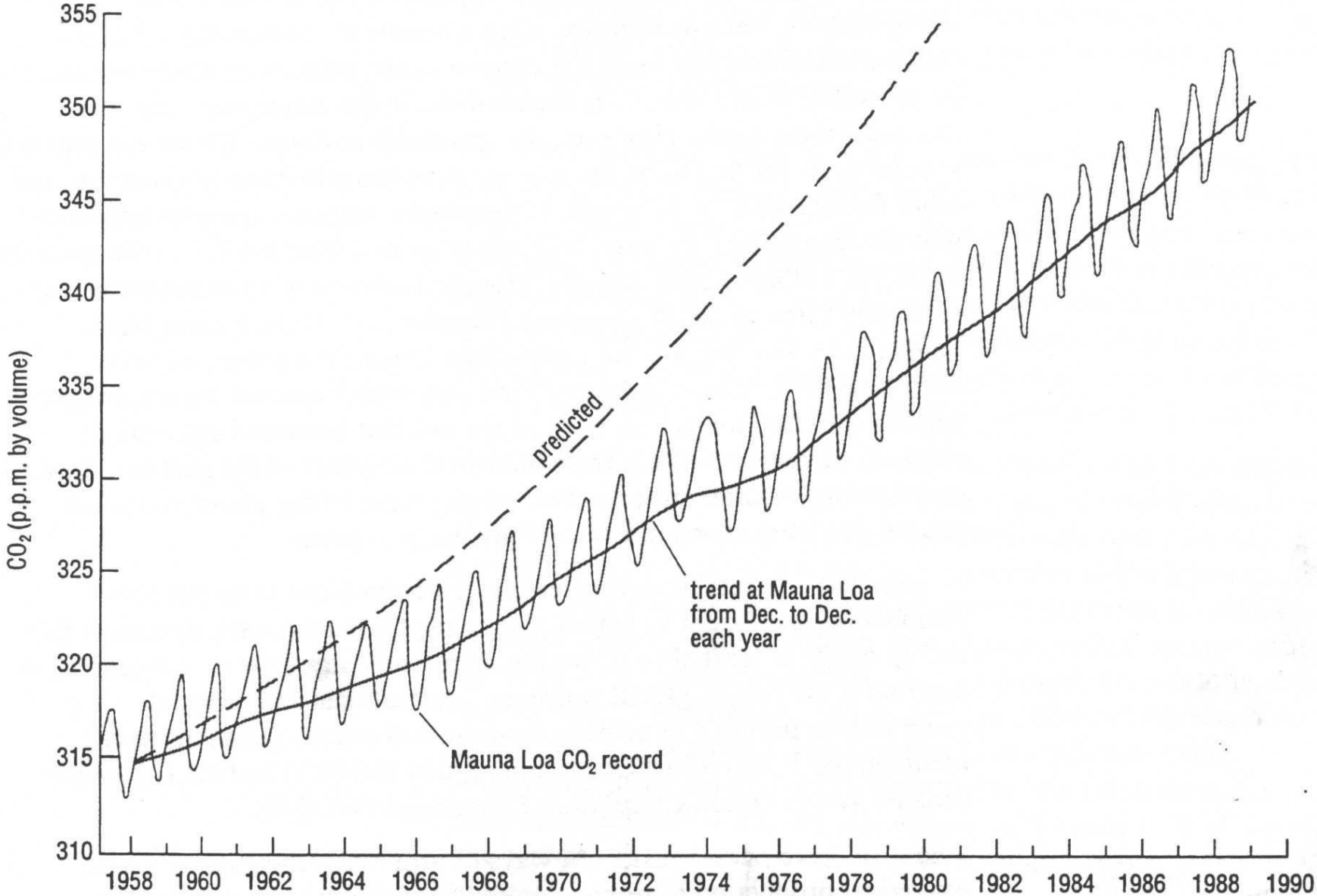



Source: United Nations, Population Prospects: 1998 Edition.
World Population reached:
1 billion in 1804
2 billion in 1927 (123 years later)
3 billion in 1960 (33 years later)
4 billion in 1974 (14 years later)
5 billion in 1987 (13 years later)
6 billion in 1999 (12 years later)
Unless we reduce our growth rate soon,
World Population will reach:
7 billion in 2013 (14 years later)
8 billion in 2028 (15 years later)
9 billion in 2054 (26 years later)
You are Here ->
Billions of people
0
1
2
3
4
5
6
7
8
9
0
200
400
600
800
1000
1200
1400
1600
1800
2000
Year

## U.S. AND WORLD COAL COMBUSTION (millions of tons)

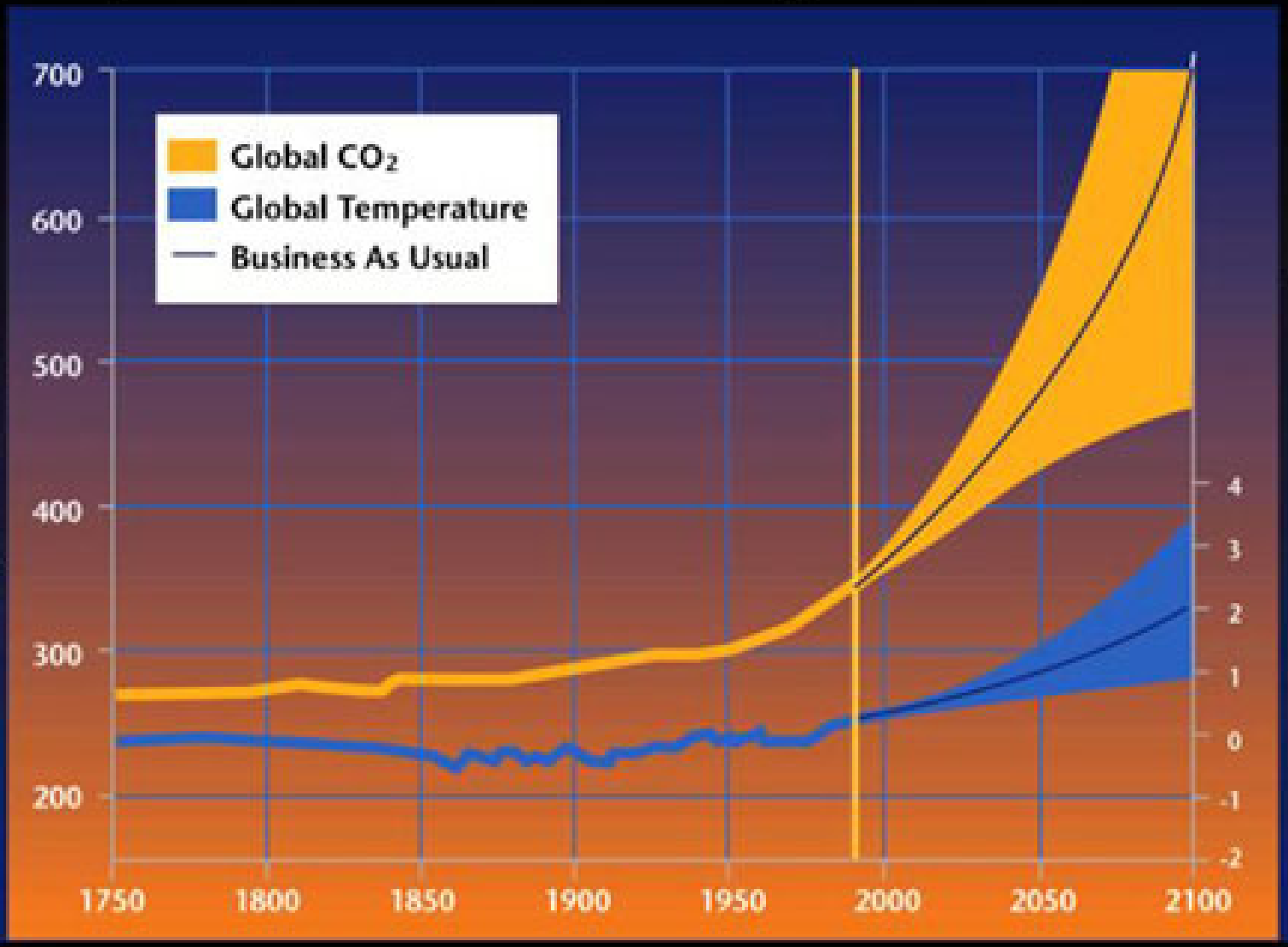



U.S. and world combustion of coal (in millions of metric tons) has increased steadily from 1937 to the present. It is expected to increase even more between now and beyond 2040.


355
350
345
340
335
330
325
320
315
310
$CO_2$ (p.p.m. by volume)
predicted
trend at Mauna Loa
from Dec. to Dec.
each year
Mauna Loa $CO_2$ record
1958
1960
1962
1964
1966
1968
1970
1972
1974
1976
1978
1980
1982
1984
1986
1988
1990

(b)

# Projection of $CO_2$ and Temperature to 2100



# NOT JUST CARBON DIOXIDE

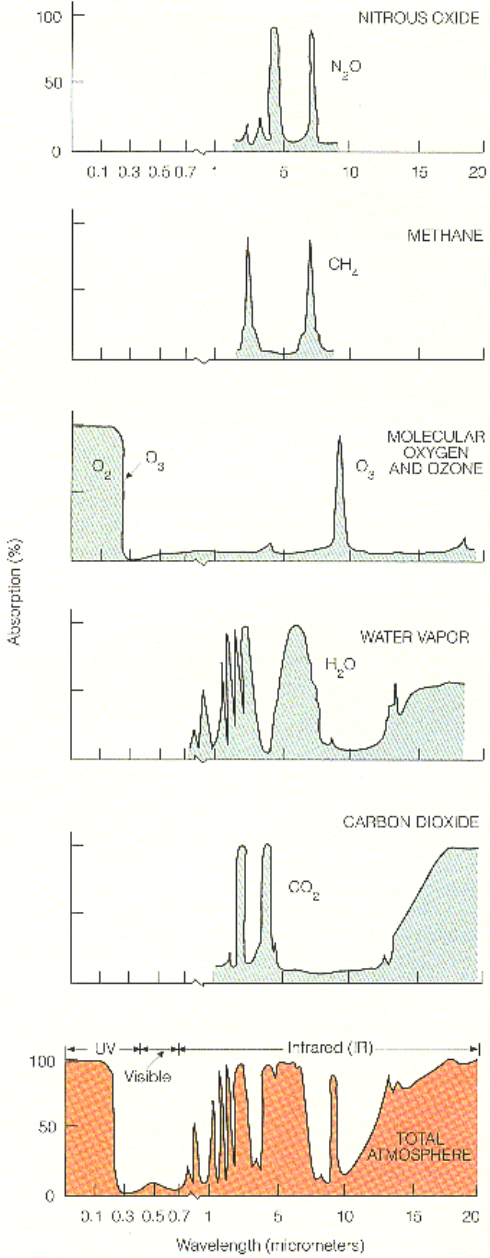
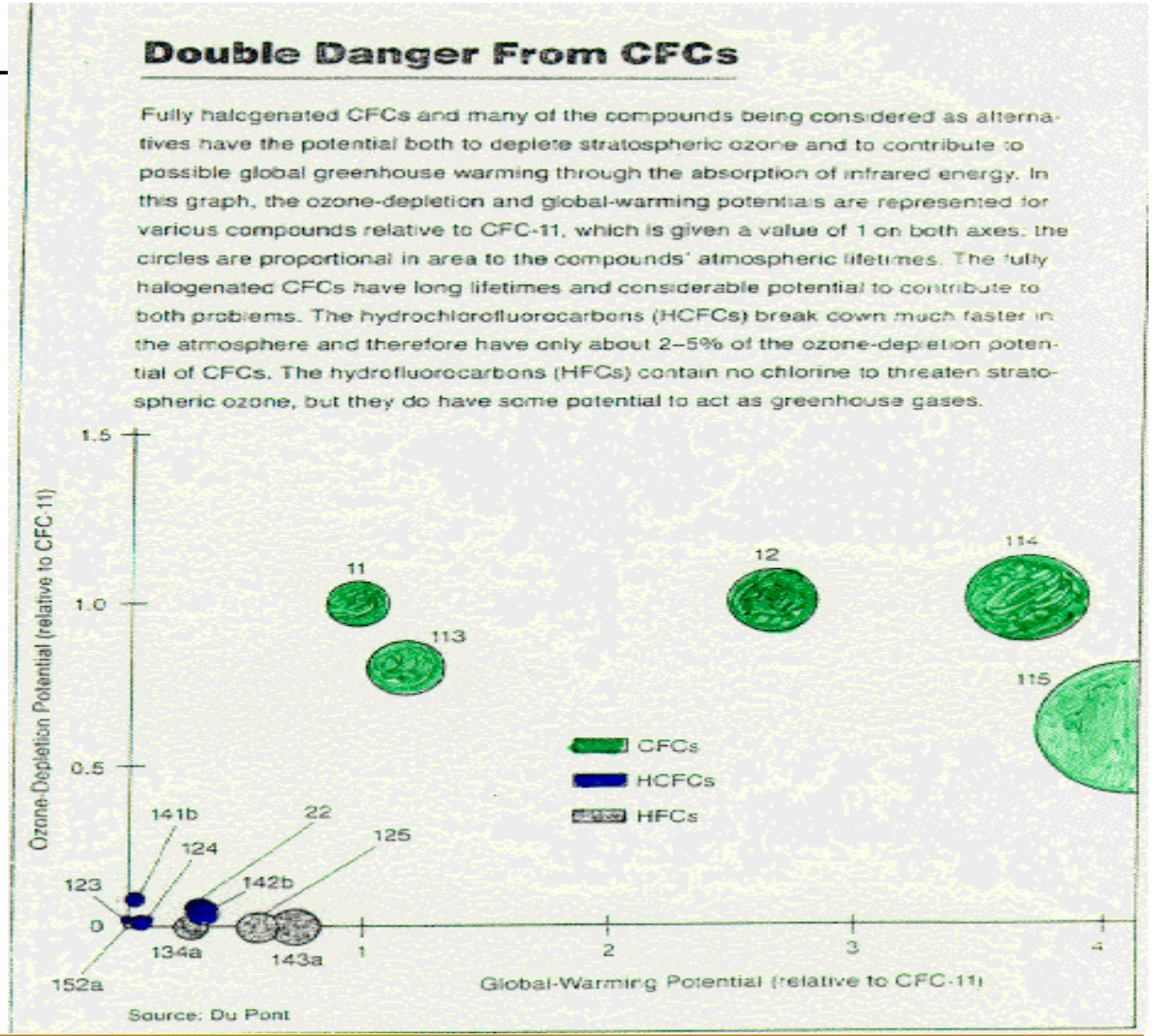
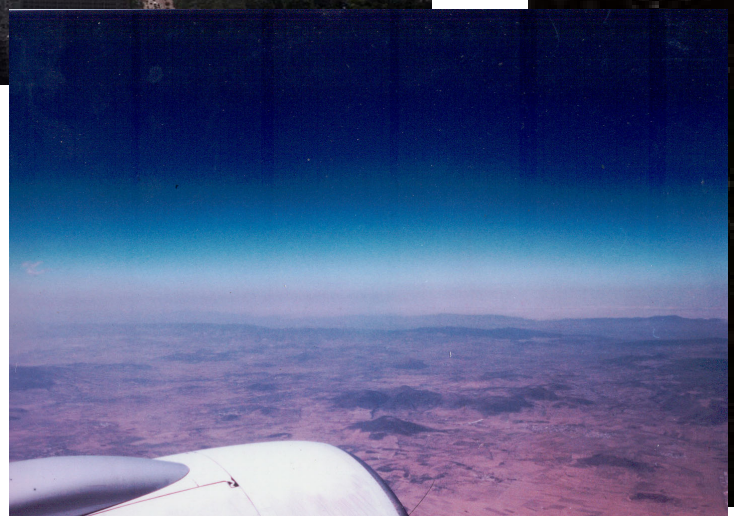



FIGURE 2.10
*Absorption of radiation by gases in the atmosphere.*

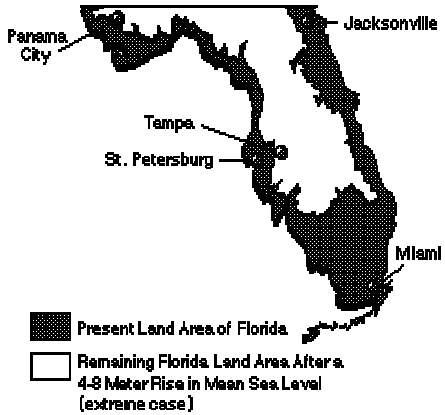


# POLLUTANT IMPACTS DEPENDS ON POSITION IN THE ATMOSPHERE AND THEIR LIFETIMES



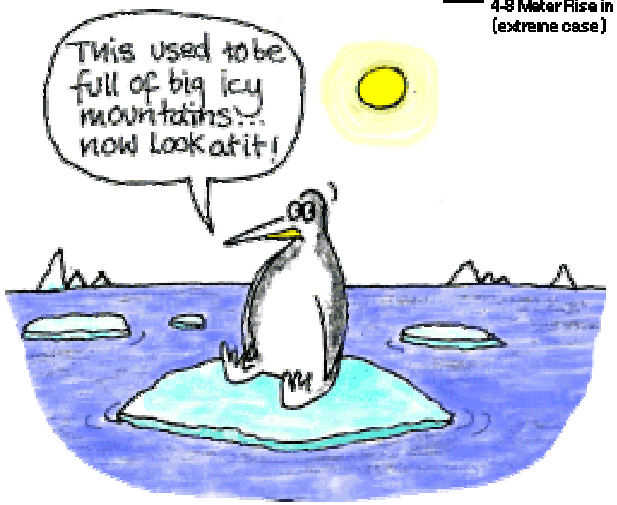
# Our Planet is not HOMOGENEOUS

## GLOBAL CHANGE – Global Climate Models (GCMs)




Wetter!

Drier!

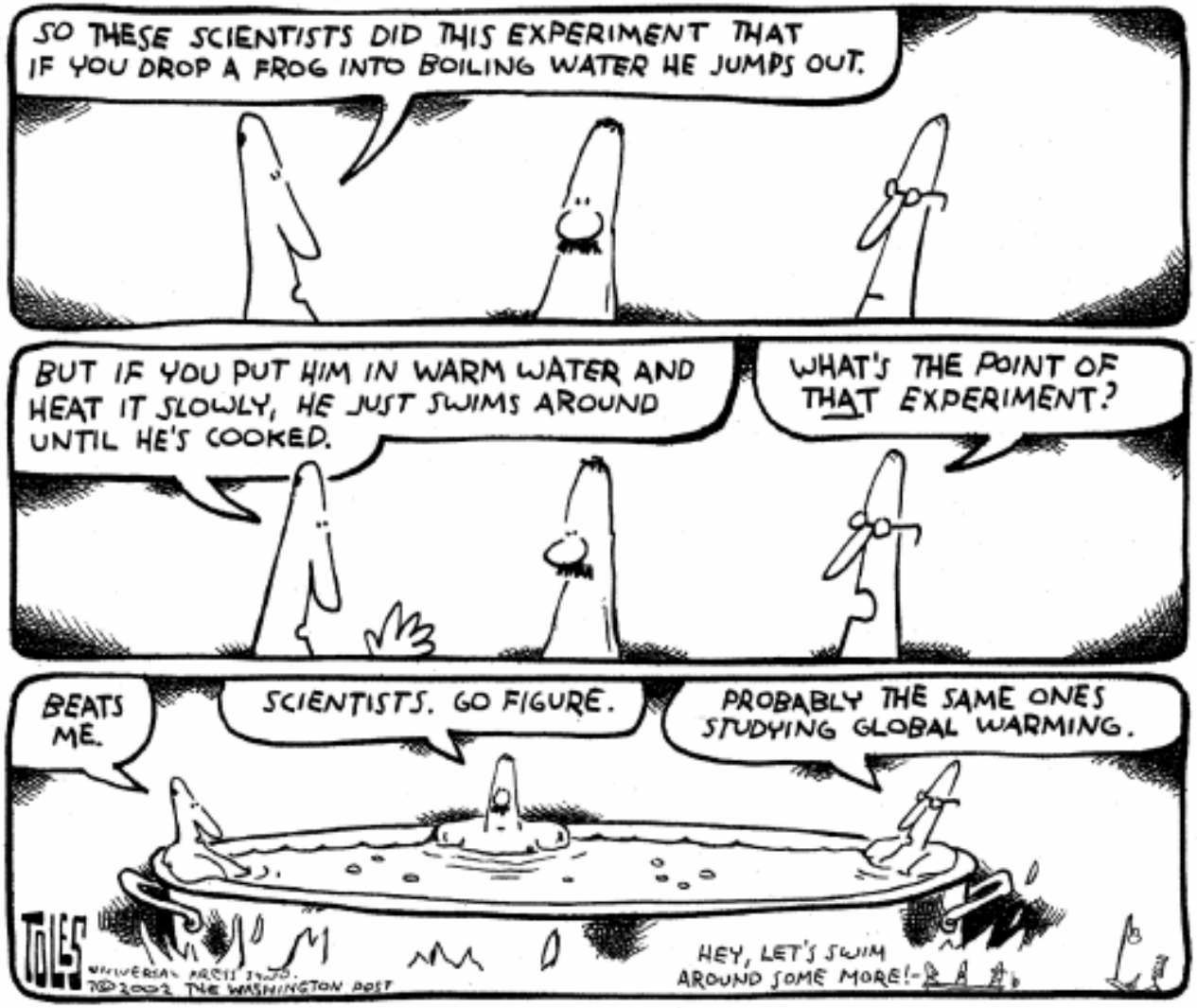



Hotter!

Colder!

# Increases in Severe Weather – EXTREMES!

# RATE OF CHANGE IMPORTANT – Timing!



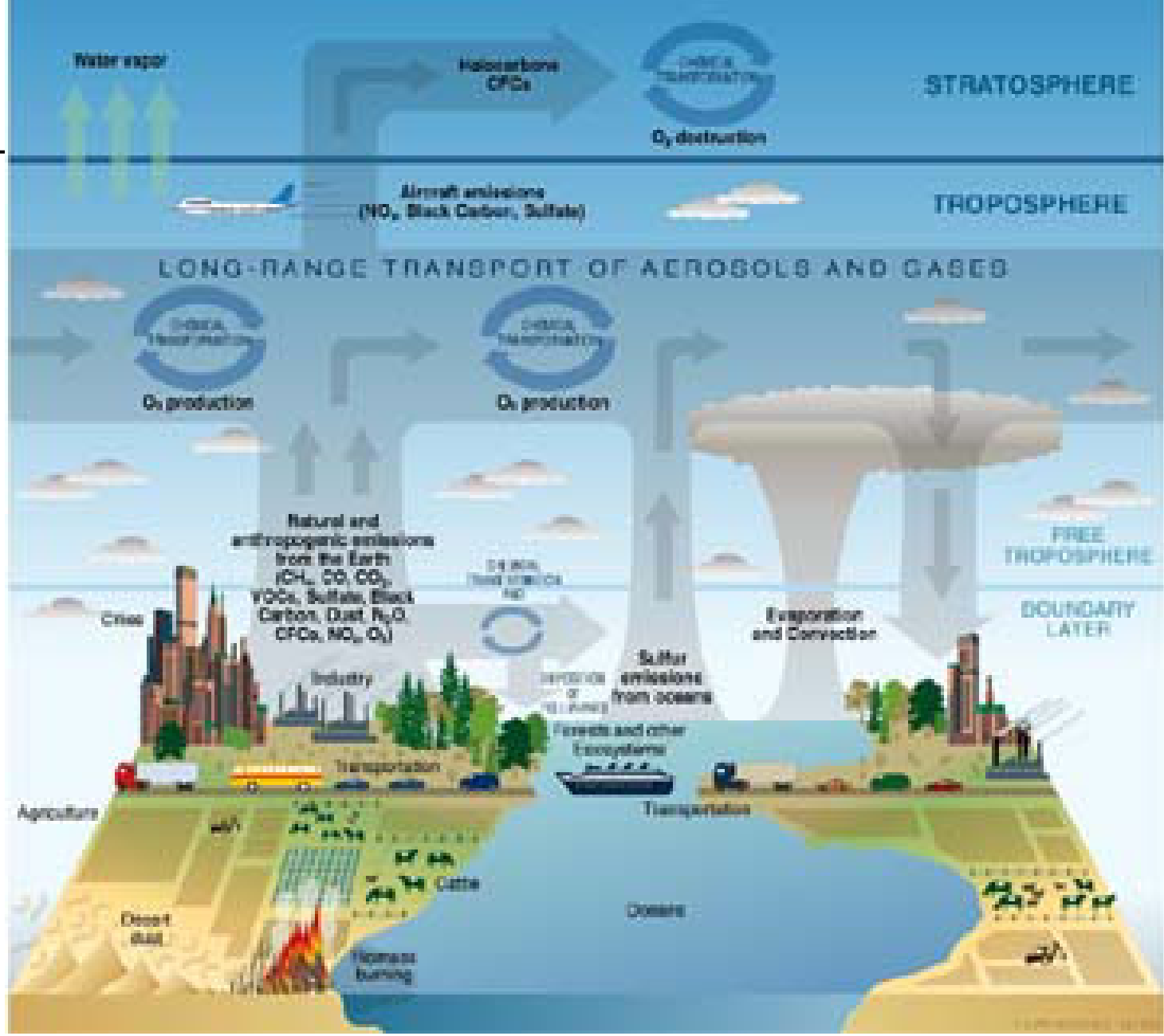

7-31-02

# Strategic Plan for the Climate Change Science Program

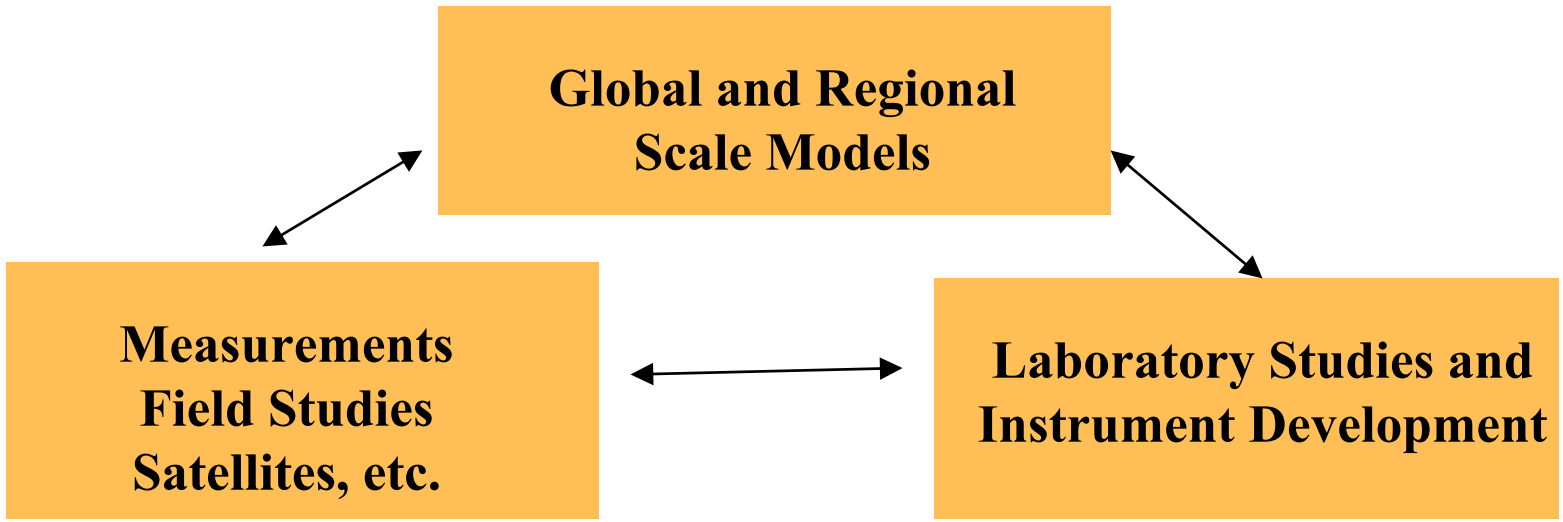
*Final Report*

*July 2003*

*Chapter 3. Atmospheric Composition*



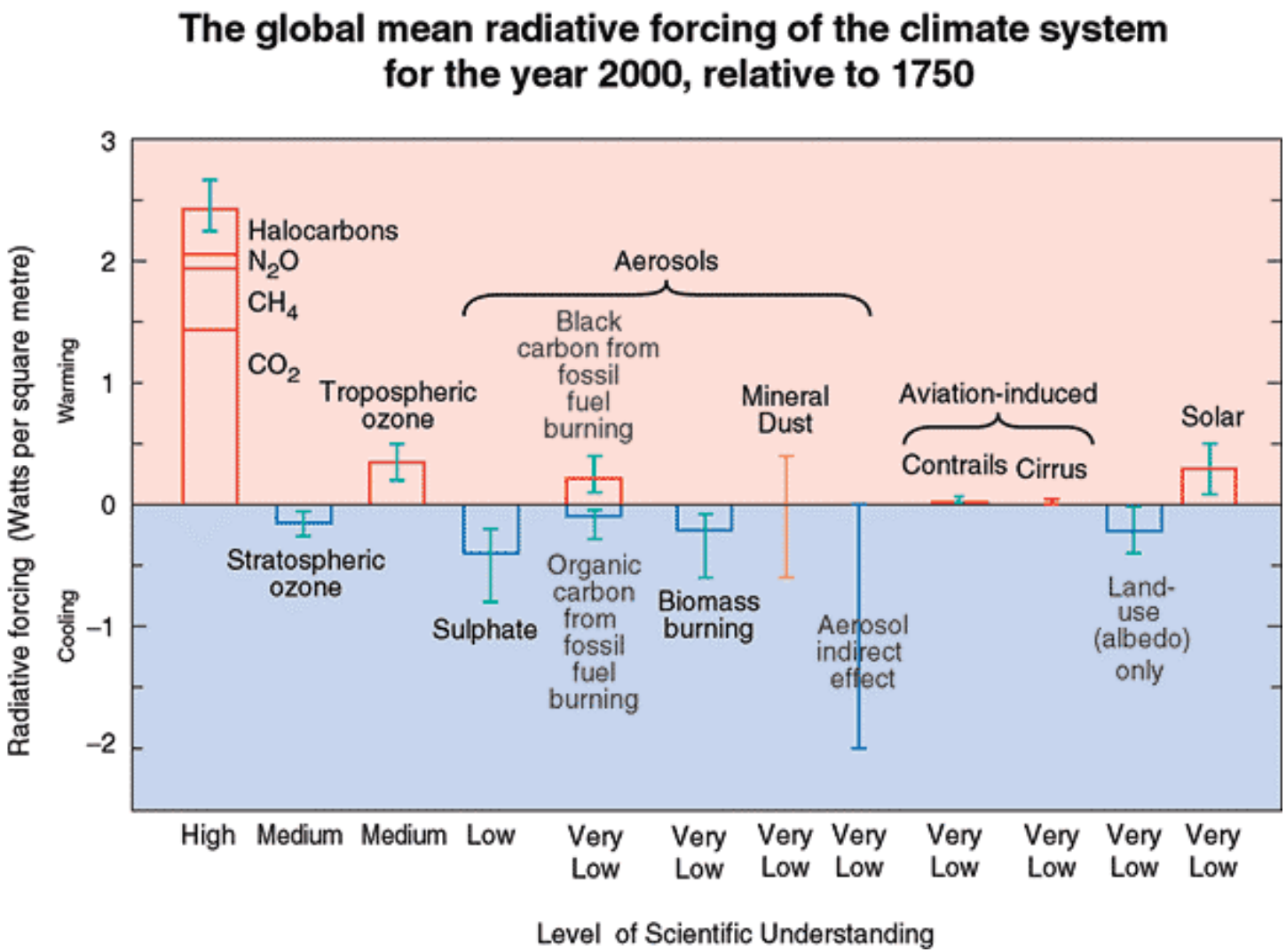
# Climate Model Improvements



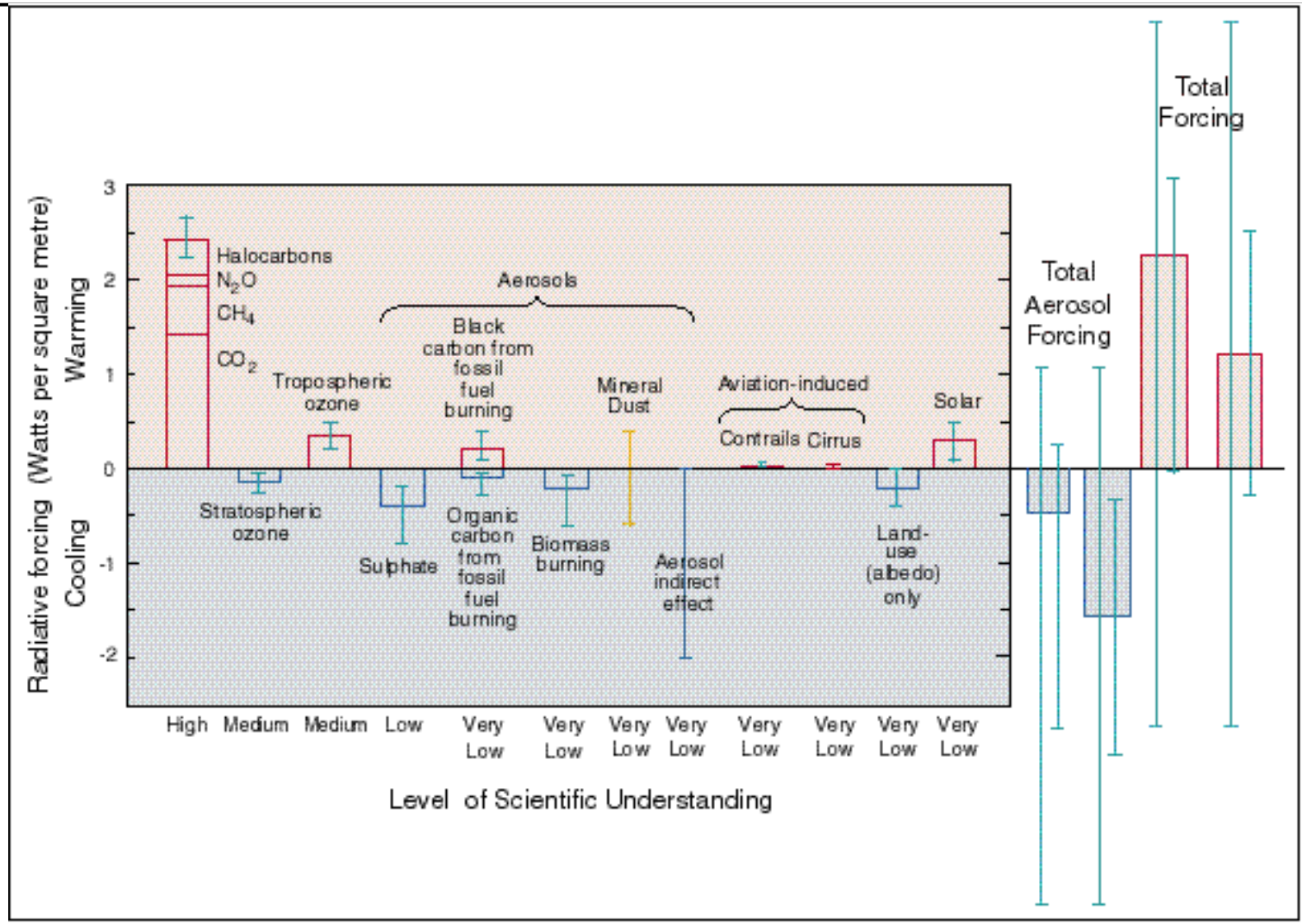

**Sources – Lifetimes – Radiative Properties**
**Feedbacks to Other Systems – Linkages**

**Processes- Oxidation – Aging – Secondary Aerosols/$H_20$!**
**All Greenhouse Gases and Aerosols are Not Created Equal!**

# AEROSOLS & CLOUDS – ARE NOW THE BIG UNCERTAINTY



# AEROSOL IMPACTS – NEW DOE/ASP FOCUS



US Climate Change Science Program
www.climatescience.gov

# Strategic Plan for the Climate Change Science Program

***Final Report, July 2003***

## CHAPTER 3. ATMOSPHERIC COMPOSITION

What are the climate-relevant chemical, microphysical, and optical properties, and spatial and temporal distributions, of human-caused and naturally occurring aerosols?

# CLIMATE EFFECTS OF FINE AEROSOLS

## Radiation and Precipitation Impacts!

**Light Scattering** – Cooling (short wave)

**Light Absorption** – Heating (long wave- all aerosols!)

**Indirect** – Cloud Condensation Nuclei – How Hygroscopic Are They?

Increased Clouds to HAZY SKIES –

Depending upon Concentration and Composition and Position.

# Direct Aerosol Radiative Forcing – You Need to Determine:

Optical Properties -(Size, Chemical Composition as Function of Size, Optical Constants, Etc.)

Sources - (Natural and Anthropogenic)

Lifetimes - (Size and Hygroscopicity)

Position in Atmosphere (Vertical & Horizontal Distributions)

Mexico City – MEGACITY MAJOR SOURCES

# AEROSOLS

**Black Carbon**

Wind Blown Dusts

Sea Salt

Sulfates

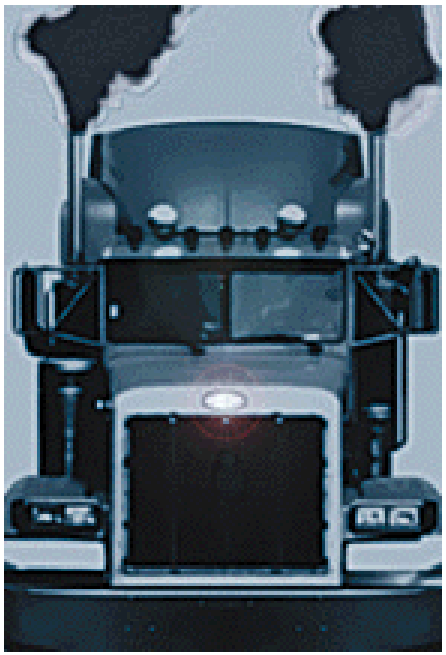
Nitrates

Secondary Organic Aerosols

# BLACK CARBON – PRIMARY AEROSOL

Strong Absorber of Short Wave And Long Wave Radiation.

DIESELS



BIOMASS BURNING

# BLACK CARBON

IS STRONG UV-VIS

ABSORBER AS WELL

AS LIGHT SCATTERER

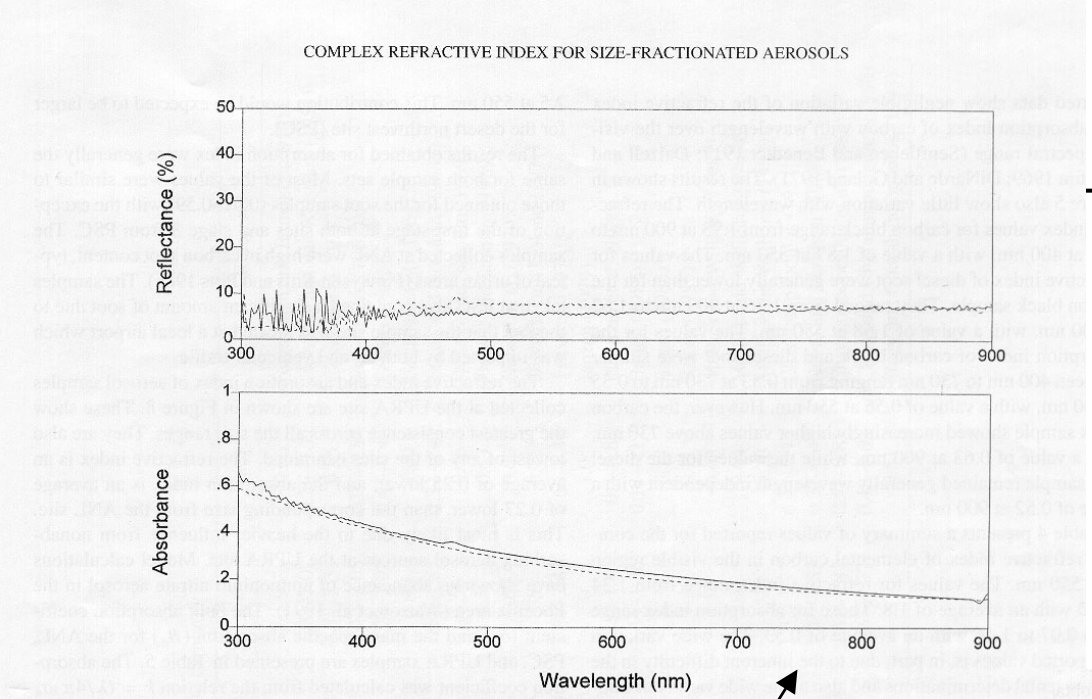



**Figure 4.** Total reflectance (top) obtained by using the 8° sample mount and absorption spectra (bottom) of carbon bla deposited on quartz collection slides and diesel soot (—) pressed into a KBr pellet.

**MEASUREMENTS**

**REAL AND IMAGINARY OPTICAL PROPERTIES FOR MODELS**

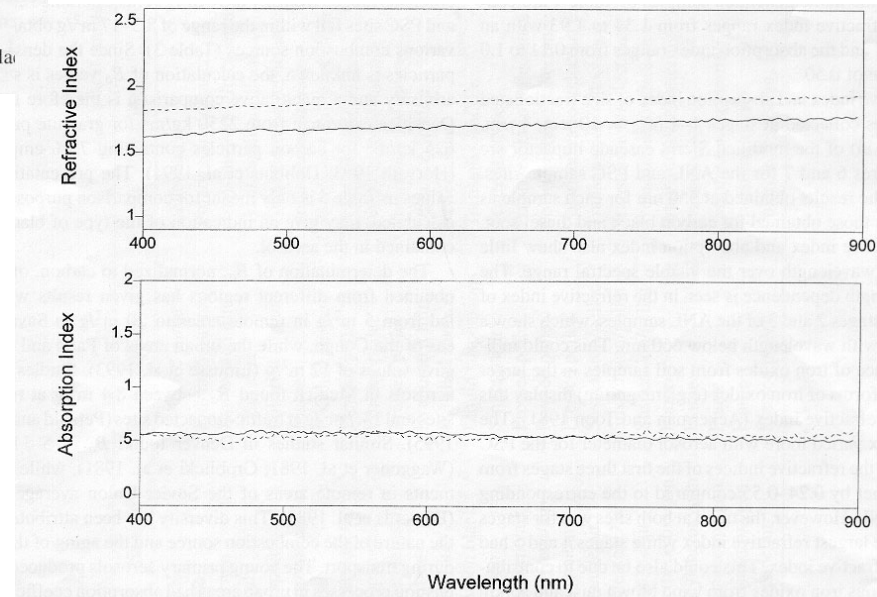
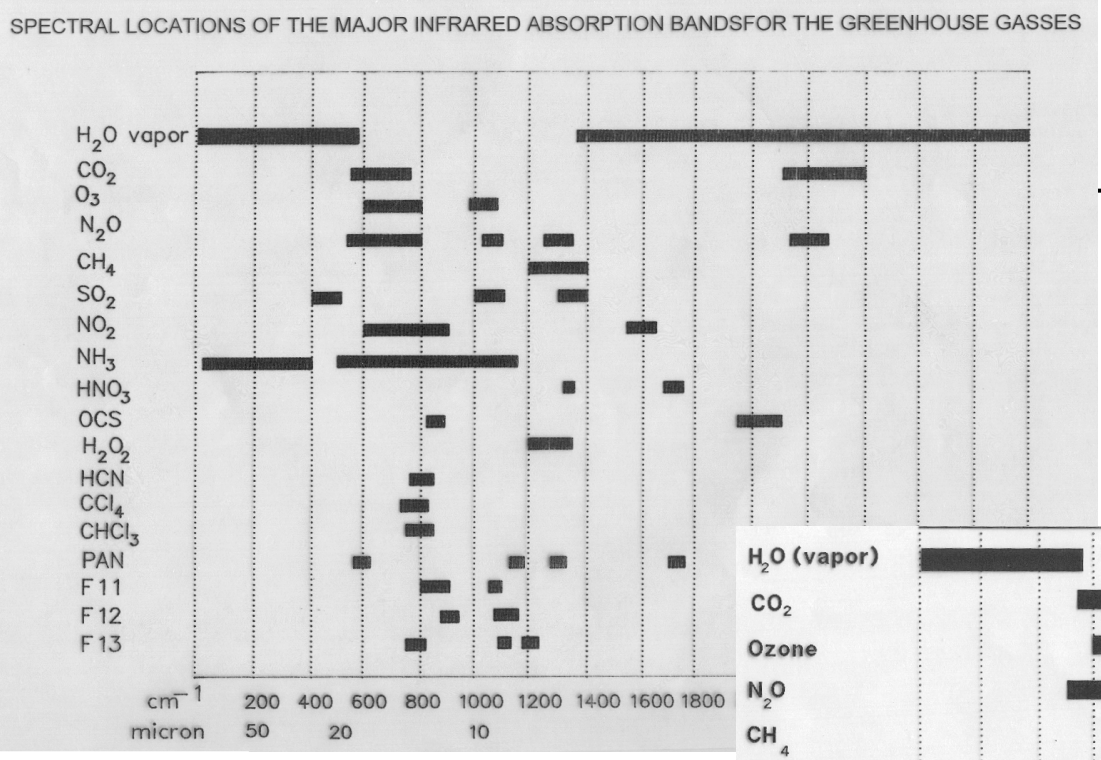
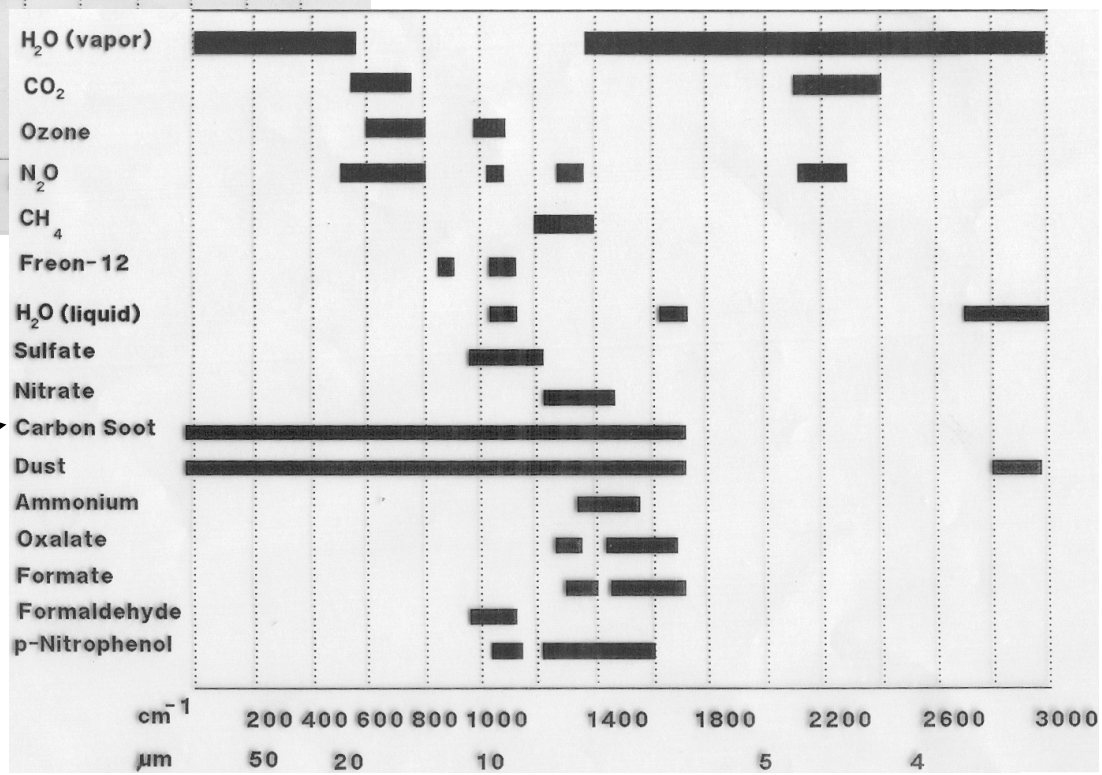



**Figure 5.** Refractive index (top) and absorption index (bottom) of carbon black (- - -) and diesel soot (–




# AEROSOLS ABSORB LONG WAVE RADIATION




**BLACK CARBON**

ALSO STRONG

Infra-Red ABSORBER

# MEGACITIES AND MINI-MEGACITIES MAJOR SOURCES OF OZONE, AEROSOLS, GREENHOUSE GASES

*THESE SOURCES WILL BE CHANGING OVER TIME AS THE CITIES DEVELOP AND THE TECHNOLOGIES EVOLVE*

**BLACK CARBON –** *DIESEL AND TWO-CYCLE ENGINES, ETC.*

**CARBONACEOUS AEROSOLS (ORGANIC & BLACK CARBON)**

**SULFATE, NITRATE –** *FOSSIL FUEL COMBUSTION*

**SECONDARY ORGANIC AEROSOLS –** *FOSSIL AND BIOGENIC*

**NEED TO BETTER CHARACTERIZE THE EMISSIONS**

**AND THEIR PROPERTIES (SIZE, ETC.) – CLIMATE IMPACTS!**

**HEALTH – VISIBILITY – LINKAGES…**

# MEGACITIES

➢ *10 Million*

1950 – 1 (NYC)

1995 – 14

2015 – 21

**Mini – MEGACITIES**

*5 Million – 10 Million*

1995 – 7

2015 – 37

**ASIA – AFRICA**

2/3 rural to ½ urban by 2025

# MOSES MAIMONIDES – 1135-1204

"Comparing the air of cities to the air of deserts and arid lands is like comparing waters that are befouled and turbid to waters that are fine and pure. In the city, because of the height of its buildings, the narrowness of its streets, and all that pours forth from its inhabitants and their superfluities… the air becomes stagnant, turbid, thick, misty, and foggy… If there is no choice in this matter, for we have grown up in the cities and have become accustomed to them, you should… select from the cities one of open horizons… endeavor at least to dwell at the outskirts of the city…If the air is altered ever so slightly, the state of the Psychic Spirit will be altered perceptibly. Therefore you find many men in whom you can notice defects in the actions of the psyche with the spoilage of the air, namely, that they develop dullness of understanding, failure of intelligence and defect of memory…."

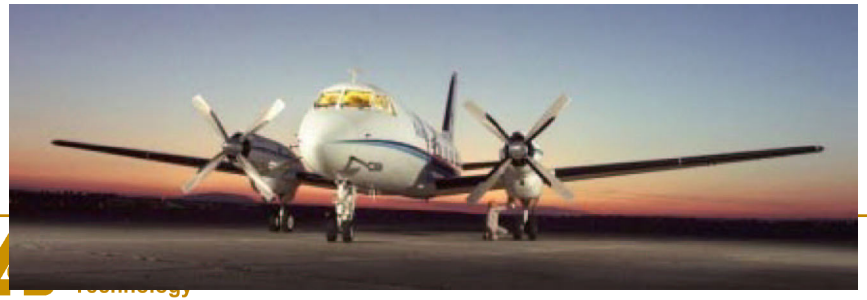
# MEGACITIES - TOP TEN

| | Population | |
|---|---|---|
| Megacity | 1991 | 2000 |
| Tokyo, Japan | 27,245,000 | 29,971,000 |
| **Mexico City, Mexico** | 20,899,000 | 27,872,000 |
| Sao Paulo, Brazil | 18,701,000 | 25,354,000 |
| Seoul, South Korea | 16,792,000 | 21,976,000 |
| New York, USA | 14,625,000 | 14,648,000 |
| Osaka, Japan | 13,872,000 | 14,287,000 |
| Bombay, India | 12,101,000 | 15,357,000 |
| Calcutta, India | 11,898,000 | 14,088,000 |
| Rio de Janeiro, Brazil | 11,688,000 | 14,169,000 |
| Buenos Aires, Argentina | 11,657,000 | 12,911,000 |

MEXICO CITY

# Megacity Aerosol eXperiment – Mexico City (MAX-Mex) 2006

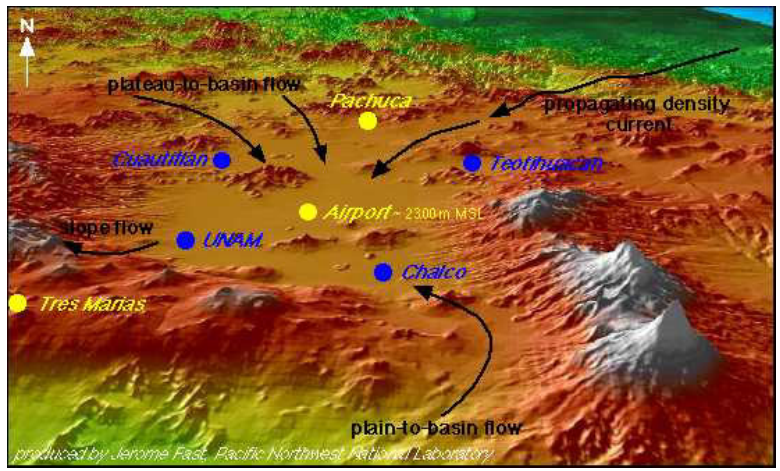
PLANNING INITIATED FOR JOINT AIRCRAFT
& GROUND BASED STUDY
COLLABORATION WITH
MCMA 2006 – MIT
MIRAGE 2006 – NSF
INTEX-B – NASA? & Mexican Scientists



# *TARGET OF OPPORTUNITY - MEXICO CITY*

## 2ND LARGEST MEGACITY

- LARGEST MEGACITY IN NORTH AMERICA
- BASIN METEOROLOGY - COMPLEX TERRAIN
- INFRASTRUCTURE CONNECTIONS!
- SIZE REASONABLE FOR AIRCRAFT AND GROUND STUDY
- PRELIMINARY GROUND FIELD STUDIES – **1997 & 2003**



# SOME FOCUS AREAS

• DETAILED CHARACTERIZATION OF A MEGACITY PLUME

*INTEGRATED AEROSOL SOURCE STRENGTHS FROM A MEGACITY – FOR MODEL COMPARISON*

***NSF and DOE Aircraft (NASA?)***

***ASP Infrastructure (RASS, SODARS, LIDARS)***

***Ground Based Measurements***

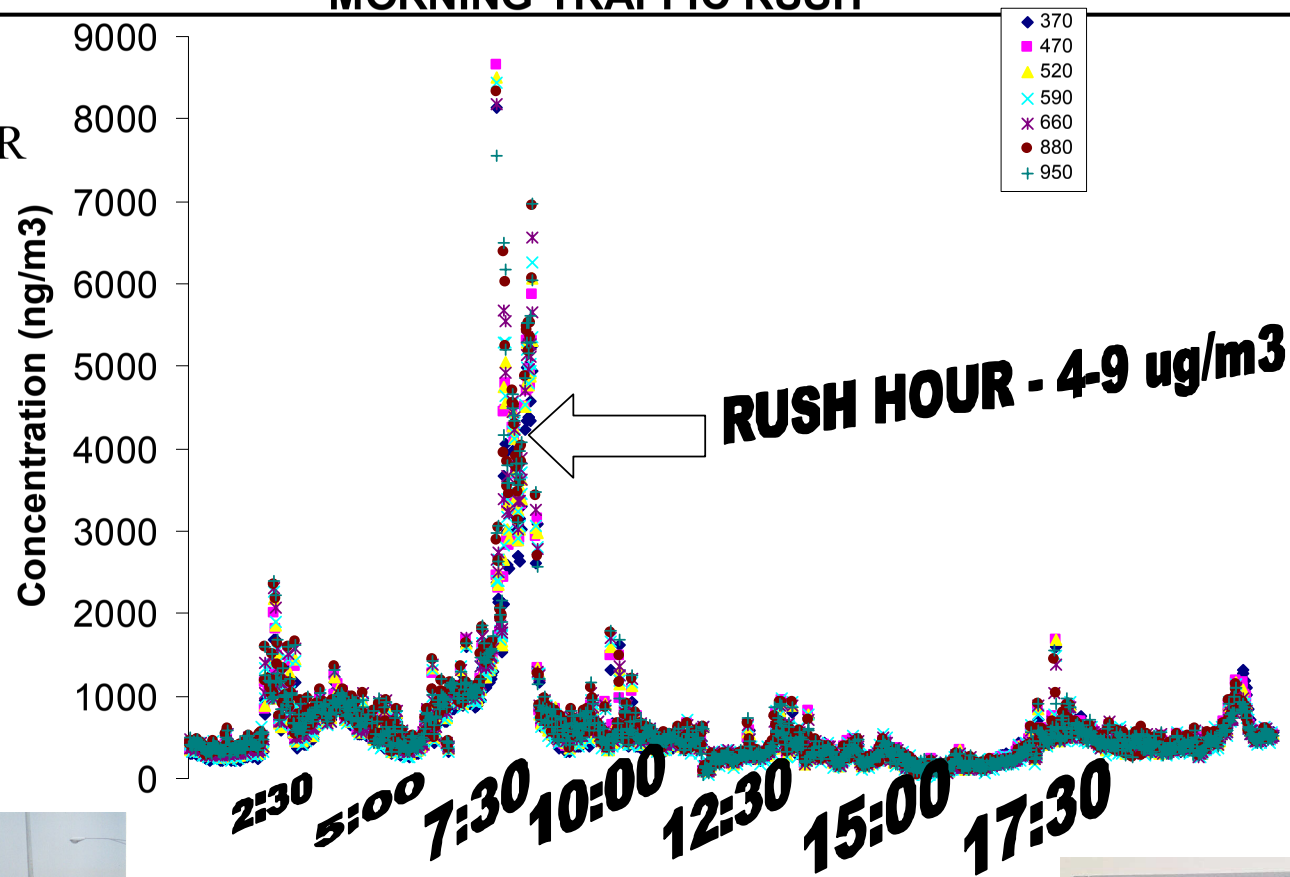
SECONDARY AEROSOL FORMATION & AGING – DOWNWIND PLUME MEASUREMENTS – BLACK CARBON, ETC.

NIGHTTIME RADIATIVE FORCING – "SMUDGE POT"EFFECTS (Jacobsen 2002) – GROUND BASED MEASUREMENTS.

# BLACK CARBON

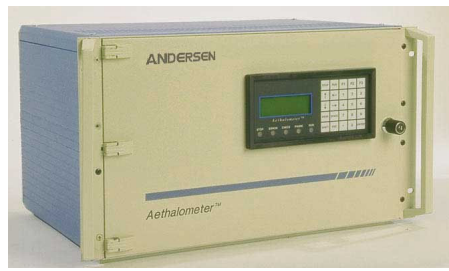
# BLACK CARBON - WED., AUG. 1, 2001, SHOWING MORNING TRAFFIC RUSH

AETHALOMETER – 7 CHANNEL

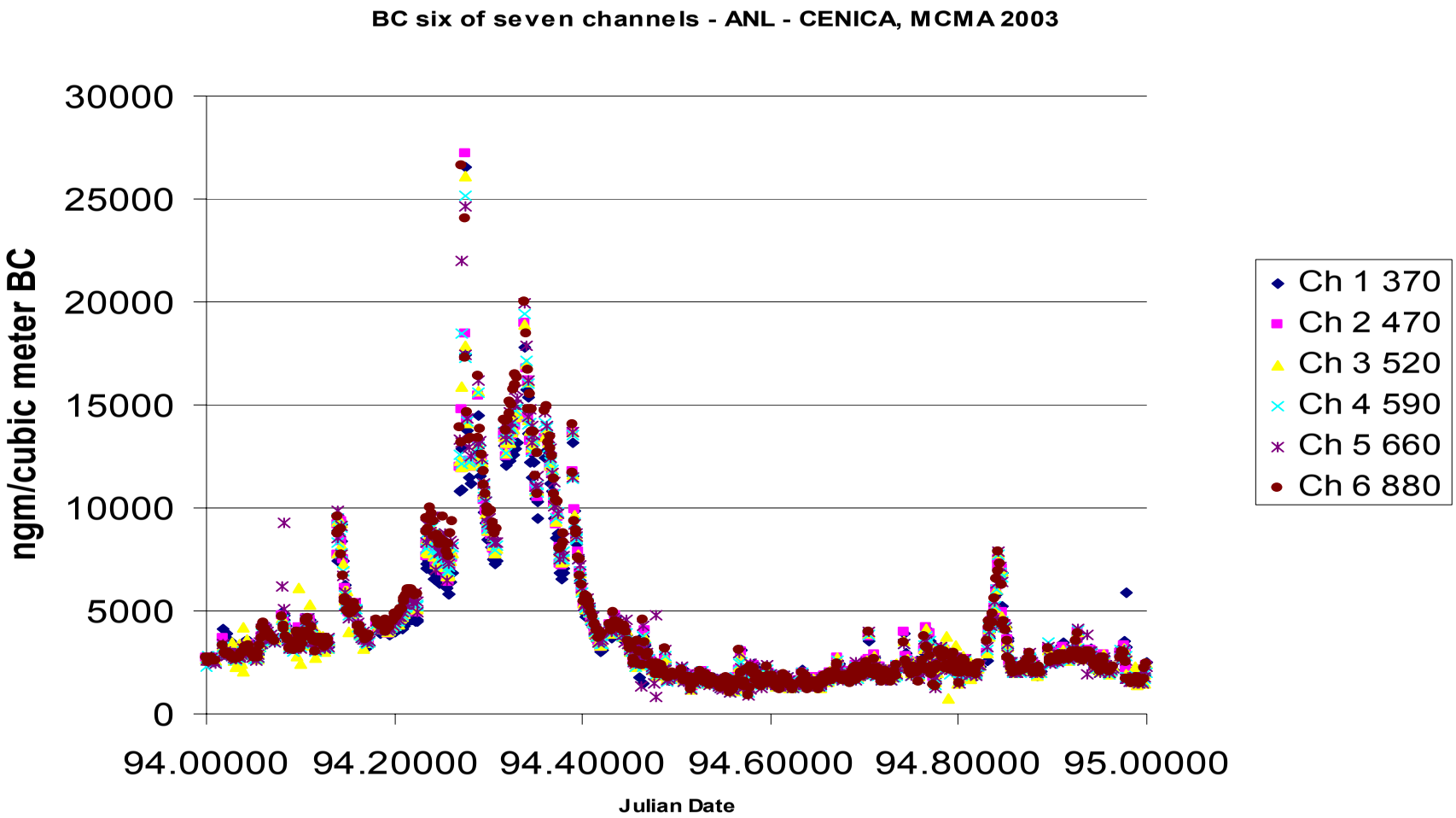


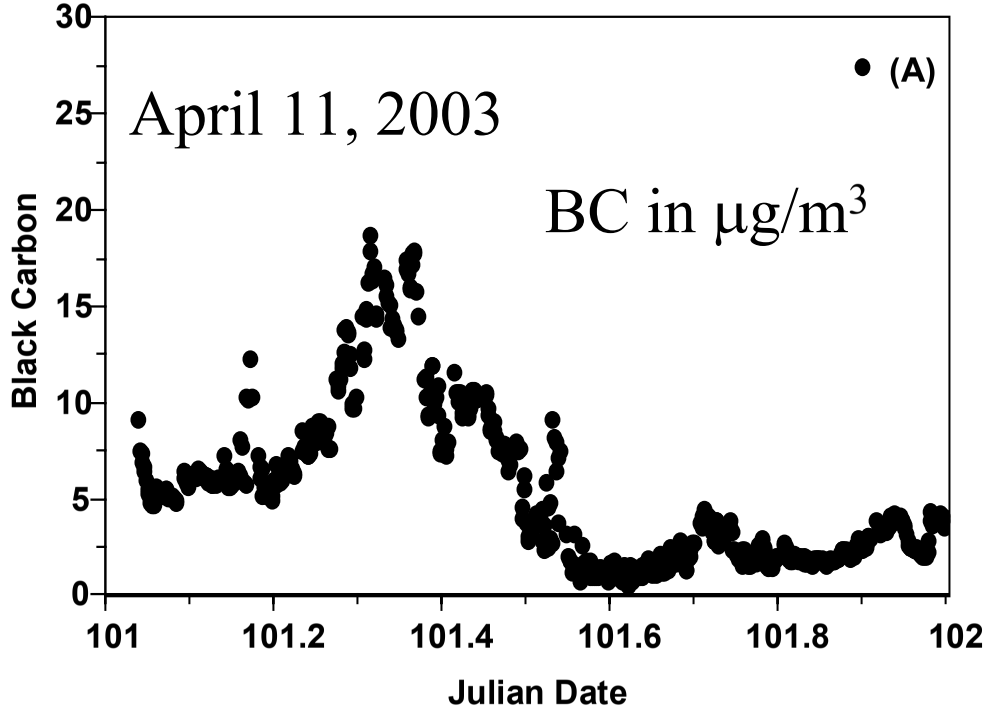
Center for Environmental Science U of C

Chicago, IL

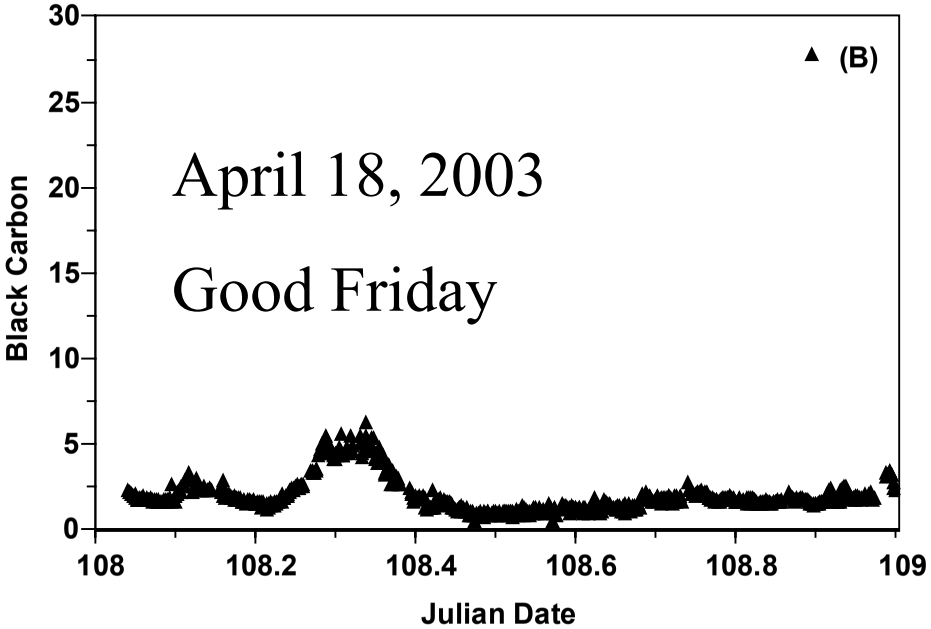


# APRIL 4, 2003 – MEXICO CITY




April 11, 2003
BC in μg/m3
(A)
Black Carbon
Julian Date
0
5
10
15
20
25
30
101
101.2
101.4
101.6
101.8
102

Soluciones en
EFECTIVO
AQUI


April 18, 2003
Good Friday
(B)
Black Carbon
Julian Date
0
5
10
15
20
25
30
108
108.2
108.4
108.6
108.8
109

BC in μg/m3

# *BC Data – 30 days – Aethalometer – Mexico City April 2003*

Daily Average - **5000 ng $m^{-3}$** (Std dev. 1500 ng $m^{-3}$)

Daily Maximum Ave **14300 ng $m^{-3}$** (Std dev. 6100 ng $m^{-3}$)

Daily Minimum Ave **1800 ng $m^{-3}$** (Std dev. 850 ng $m^{-3}$)

**PREVIOUS WORK** in 1997 Ave **PM-2.5 was 50 μg $m^{-3}$**

10 % EC estimated from thermal analyses compares very well with this work- i.e. ~ **5 μg $m^{-3}$**

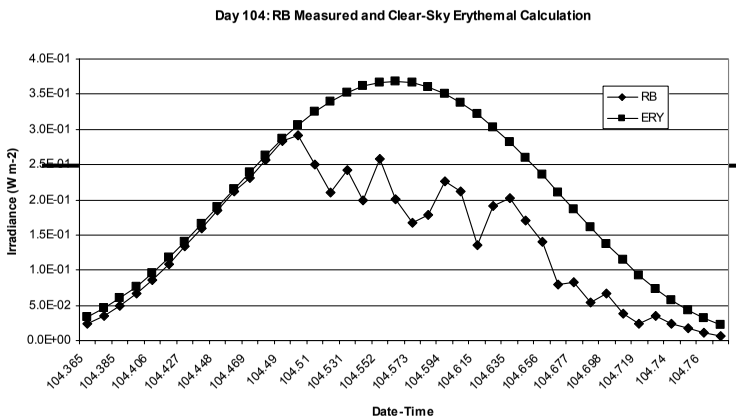
**DATA TAKEN DURING HOLY WEEK – SHOWS BC REDUCED**

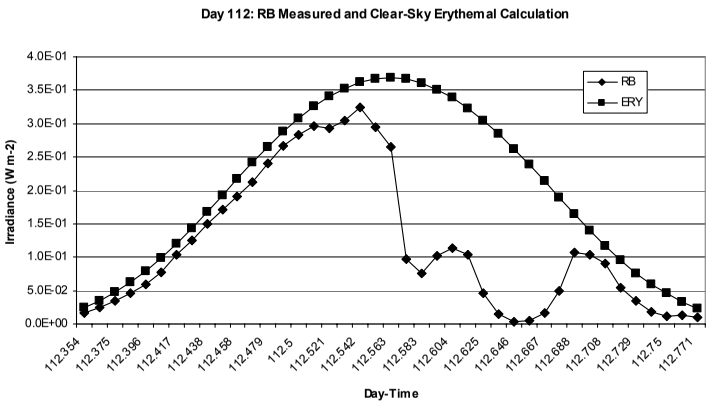
**NOT MUCH REDUCTION OVERALL IN 6 YRS IN BC!**

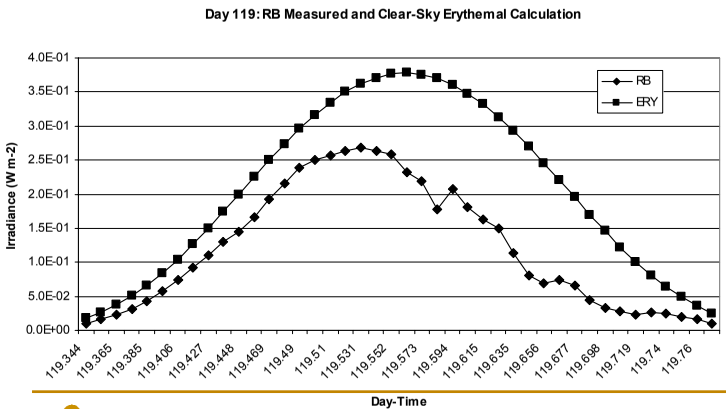
**CONSISTENT WITH CONTROLS… (i.e. NONE).**

**CHICAGO – 4-9 microgram per cubic meter max 1 ug $m^{-3}$ 24hr**

# PHOTOCHEMISTRY EFFECTS




**BC Absorbs Actinic Radiation**

Modeled UVB vs. Robertson-Berger Meter

Cloud Effects (Added Bonus!)

April 22 – Julian Date 112 – RAIN EVENT

UV-B at zero!

Early morning – Black Carbon

$CH_2O$, $NO_2$, etc. → Ozone

# IMPORTANCE OF BC PHOTOCHEMISTRY FEEDBACKS

**Less UV for PHOTOCHEMISTRY due to BC absorption**

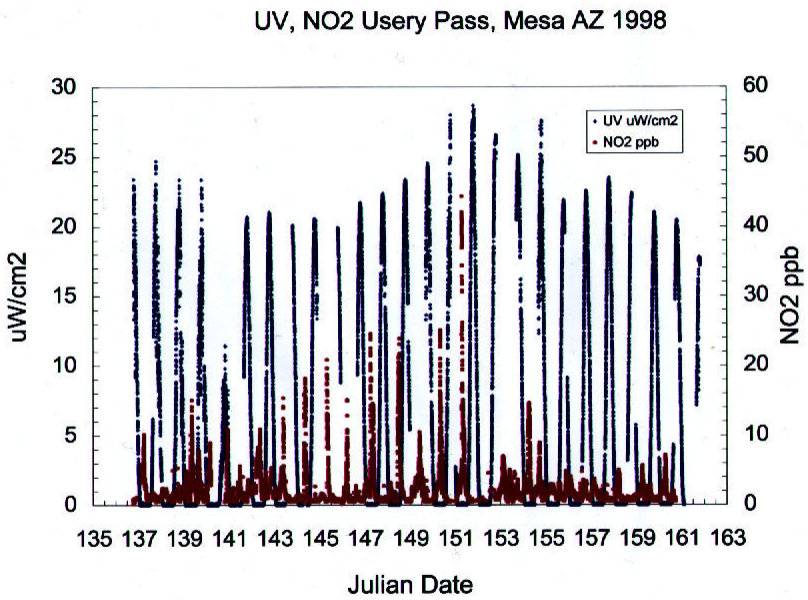
**Reduced Secondary Greenhouse Gas Formation (e.g. $O_3$)**

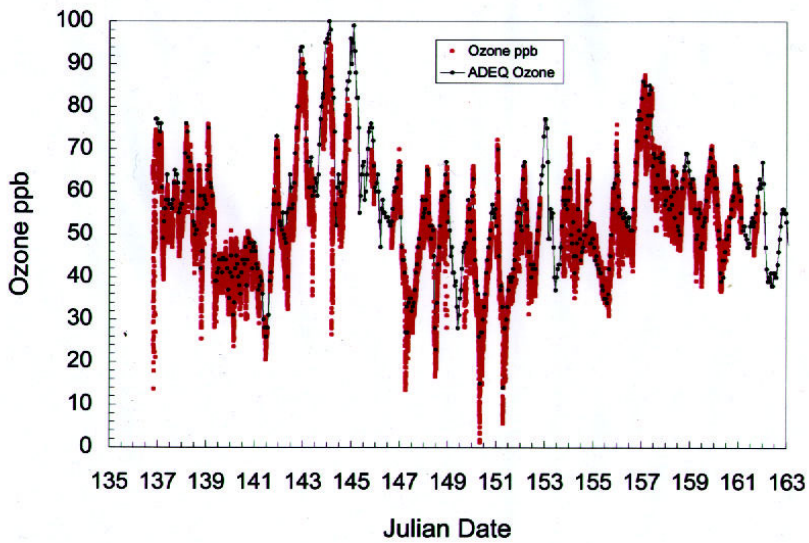
**Increased Lifetimes of Other Greenhouse Species (e.g. RH, CO)**

**Reduced Rates of Formation of Sulfates, Nitrates, Secondary Organic Aerosols**

**Biota Effects as Well – $CO_2$ Uptake – Evapotranspiration, etc.**

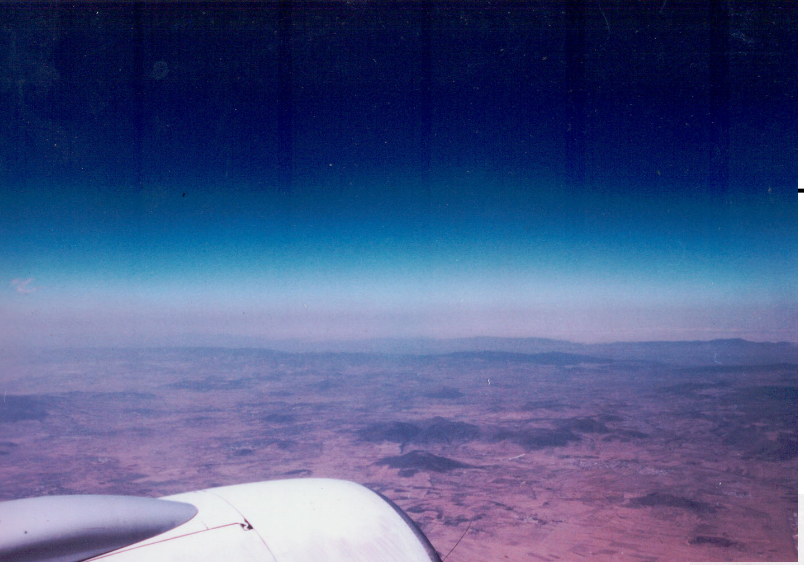
**Others?**




# BIOMASS BURNING SMOKE EVENT

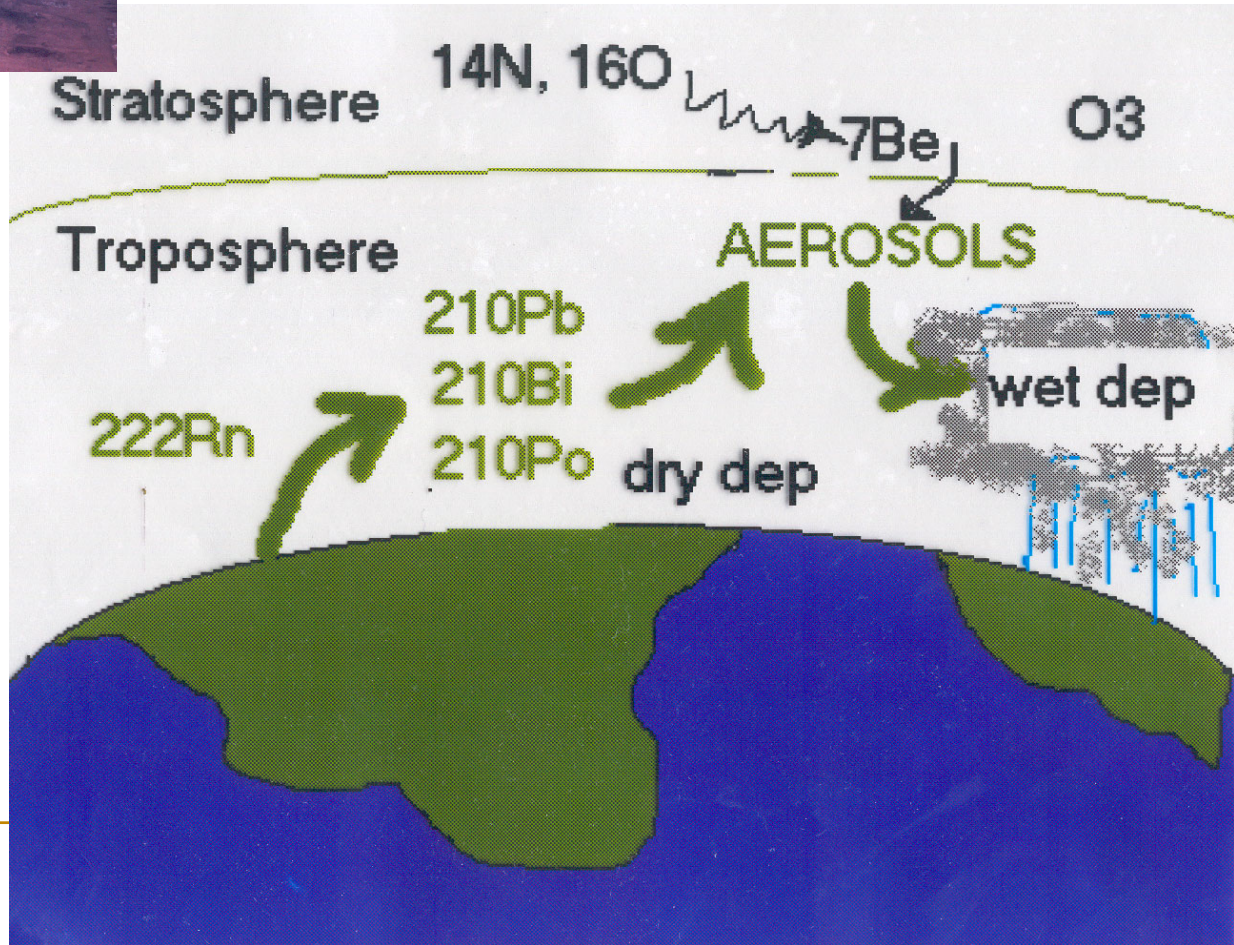
# BLACK CARBON IMPACT ON OZONE FORMATION



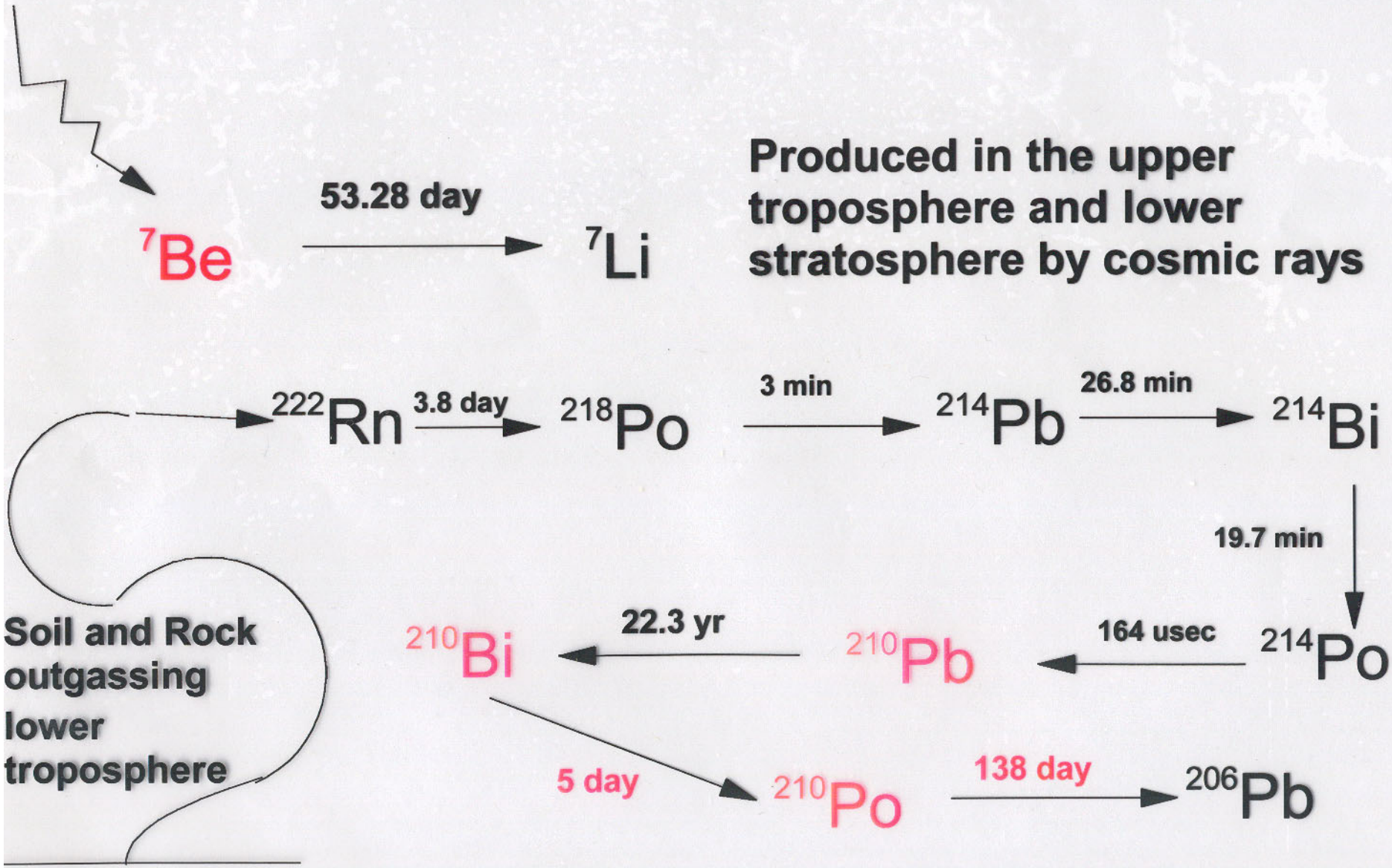
# LONG RANGE TRANSPORT DETERMINES REGIONAL AND GLOBAL IMPACTS

CAN Natural Radionuclides Help Determine

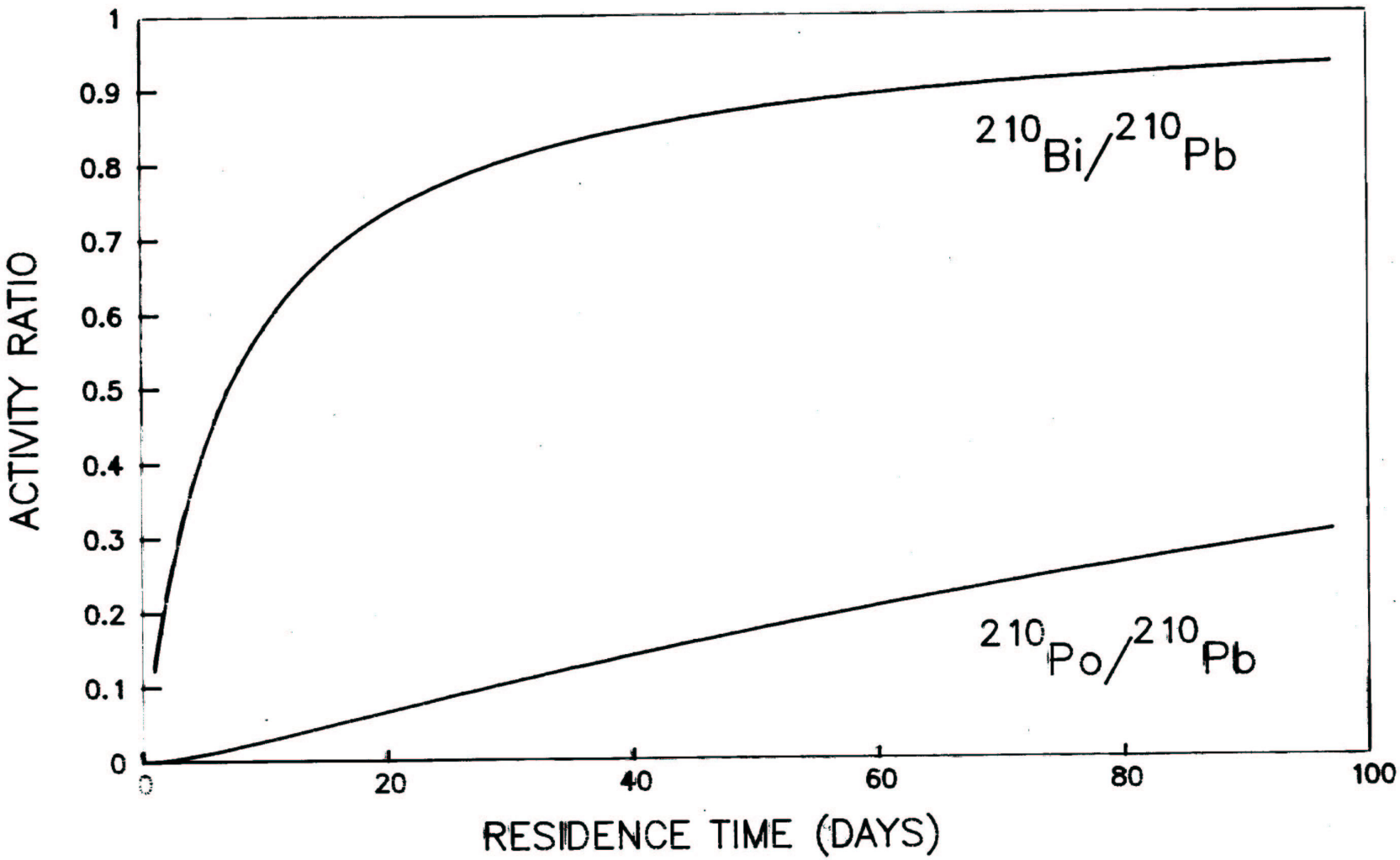
Lifetimes &Washout?



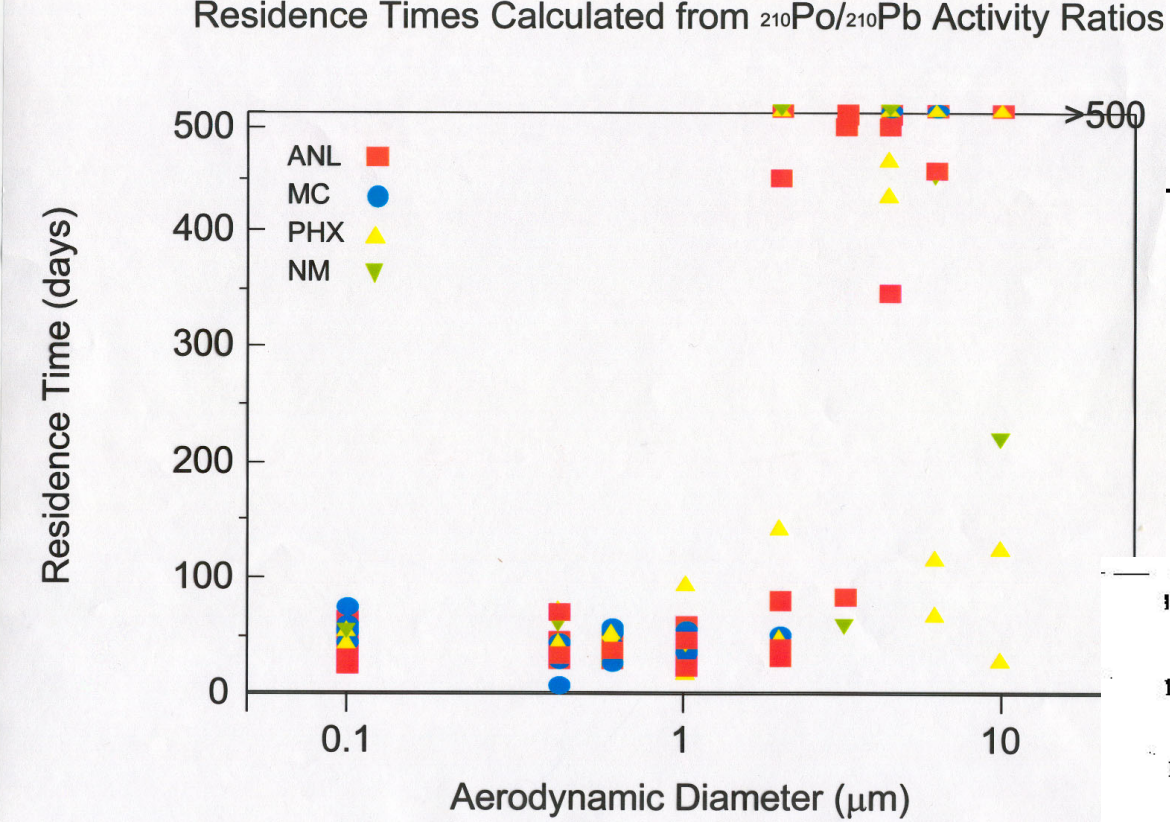
# Radioactive Decay of Natural Atmospheric Tracers

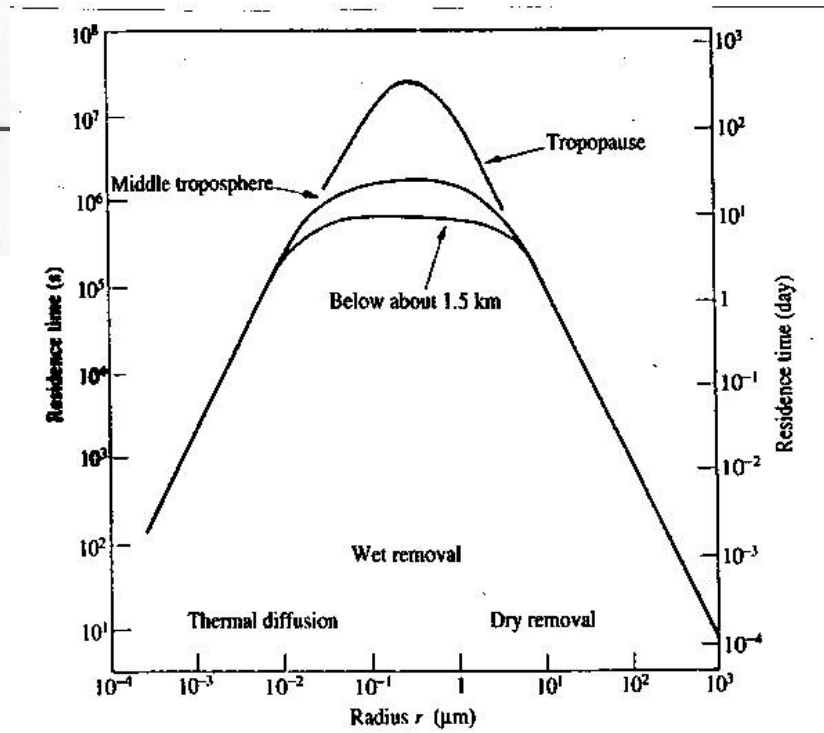


# Activity Ratio vs Residence Time for Aerosols Removed in Precipitation




(adapted from Nevissi, A.E. 1991, J. Radioanal. Nucl. Chem.)




MODELED AEROSOL

LIFETIMES

INCREASE WITH ALTITUDE

Jacobs Group – INDOEX results

RECENT RESULTS FOR PITTSBURGH (Gaffney, Nancy A. Marley, and Mary M. Cunningham, "Natural radionuclides in fine aerosols in the Pittsburgh area." *Atmospheric Environment*, 38, 3191-3200 (2004).)

**Apparent Ages for Aerosols – NETL**

| Sample | SIZE(μm) | CORR. AGE (days) |
|---|---|---|
| PA1 | <1 | 17 |
| PA2 | >1 | 11 |
| PA5 | <1 | 19 |
| PA6 | >1 | 20 |
| PA9 | <1 | 21 |
| PA10 | >1 | 12 |
| PA11 | <1 | 18 |
| PA12 | >1 | 15 |
| PA13 | <1 | 30 |
| PA14 | >1 | 27 |
| PA17 | <1 | 10 |
| PA18 | >1 | 30 |
| PA21 | <1 | 12 |
| PA22 | >1 | 25 |
| PA25 | <1 | 24 |
| PA26 | >1 | 32 |
| PA29 | <1 | 31 |
| PA30 | >1 | 46 |

OK SO WHAT IS CAUSING THIS?

WASHOUT – SULFATE, NITRATE - 10 Days

(Gaffney, et.al, "Measurement of $^{7}$Be and $^{210}$Pb in Rain, Snow, and Hail." *J. Applied Meteor*. **33** 869-873 (1994).)

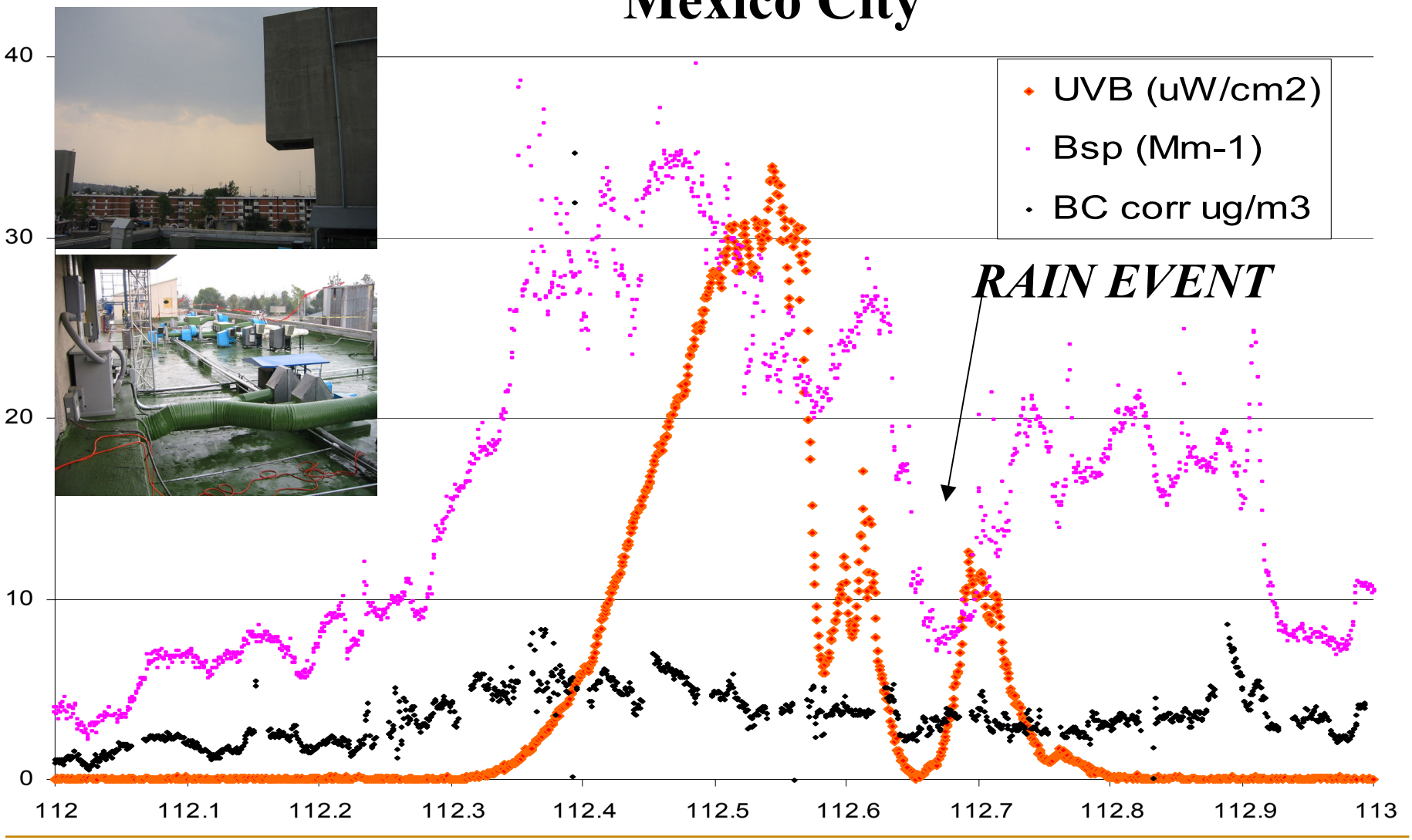
> 10 Days Aerosol Lifetimes?

**Something Hydrophobic**

**BLACK CARBON?**

# WASHOUT RATES VERY BC SLOWER TO WASHOUT

## Mexico City




April 22, 2003

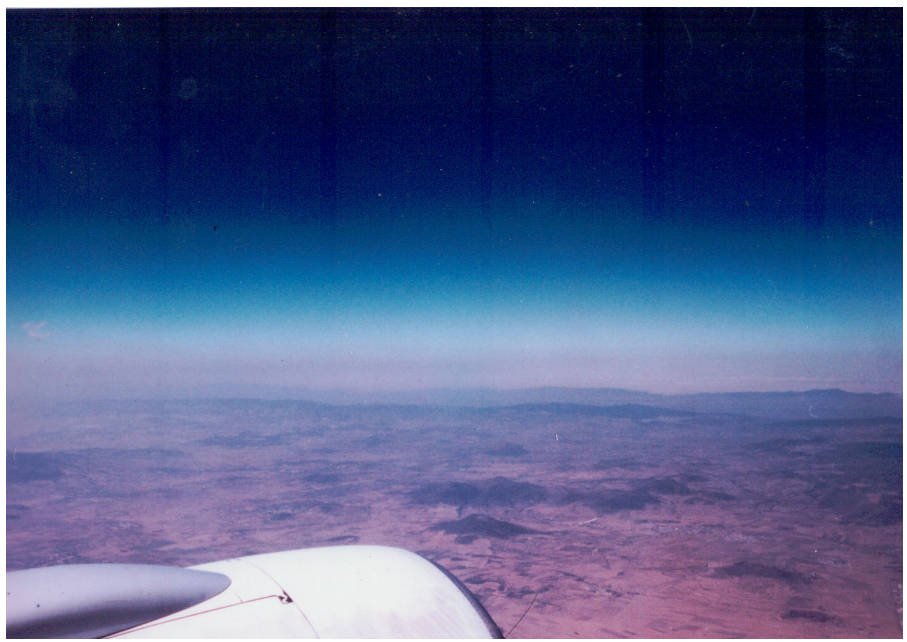
# Most Models – BC Treated Same as Hygroscopic Aerosols

## Lifetimes from 2-7 days

**This work and other data indicates BC has longer lifetime.**

**Models Typically Underestimate Black Carbon and Organic Carbon (Secondary Organic Aerosols) by Factors of 3-4 (or more) (Chung & Seinfeld 2002).**

***NEED TO ADDRESS***
***AGING OF BC***
***SOA Additions***
***MIXING STATES***



Mexico City Plume

# BETTER UNDERSTANDING OF BC – LINKS TO HEALTH
## Because Soot is small and hydrophobic it impacts our lungs!

**PM-2.5**

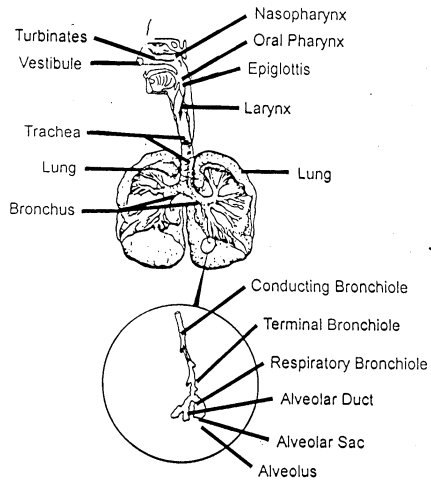
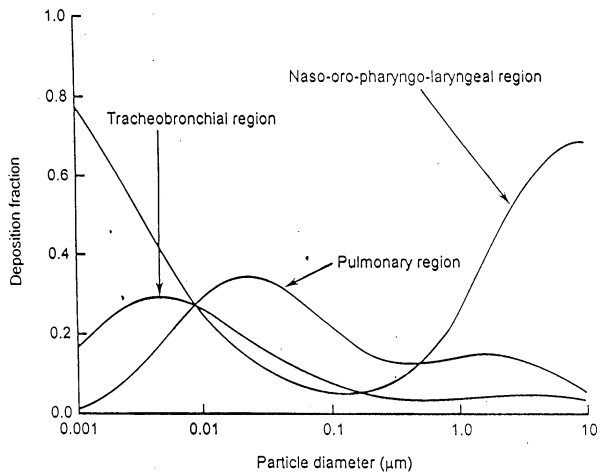
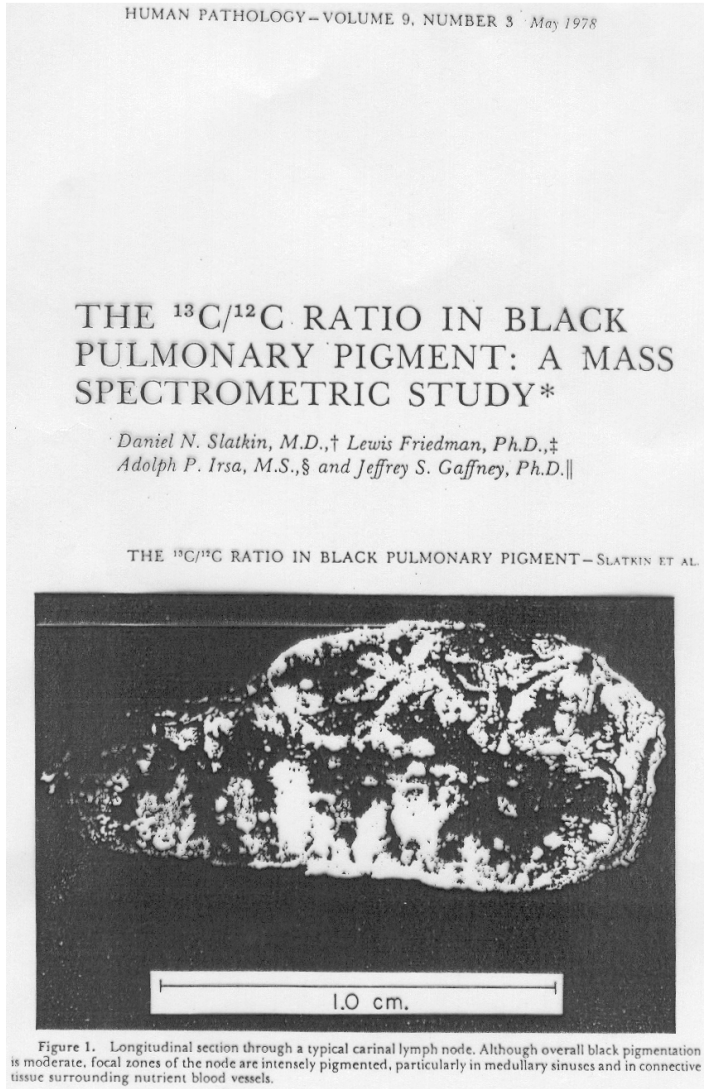



FIGURE 2.12 Schematic diagram of human respiratory tract. (From Hinds, W. C. *Aerosol Technology*. Copyright © 1982 John Wiley & Sons, Inc. Reprinted by permission of John Wiley & Sons, Inc.)



HUMAN PATHOLOGY – VOLUME 9, NUMBER 3 May 1978

THE $^{13}C/^{12}C$ RATIO IN BLACK PULMONARY PIGMENT: A MASS SPECTROMETRIC STUDY*

*Daniel N. Slatkin, M.D.,† Lewis Friedman, Ph.D.,‡ Adolph P. Irsa, M.S.,§ and Jeffrey S. Gaffney, Ph.D.‖*

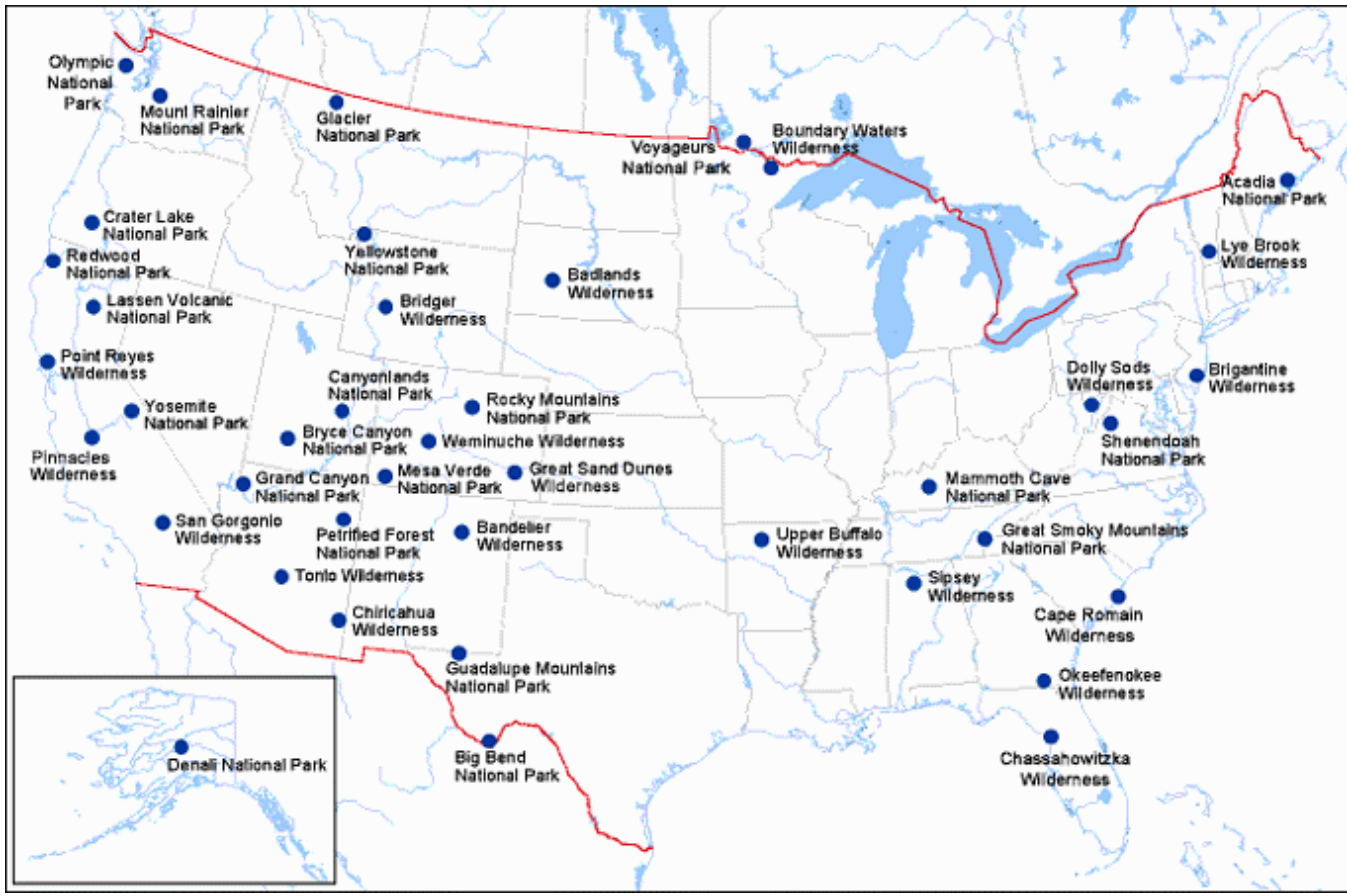
THE $^{13}C/^{12}C$ RATIO IN BLACK PULMONARY PIGMENT – SLATKIN ET AL.



Figure 1. Longitudinal section through a typical carinal lymph node. Although overall black pigmentation is moderate, focal zones of the node are intensely pigmented, particularly in medullary sinuses and in connective tissue surrounding nutrient blood vessels.

# DIRECT LINKAGE TO VISIBILITY REDUCTION – REGIONAL PROBLEM

## EPA's Regional Haze Program



# ESTIMATES FOR SOURCES OF SECONDARY AERSOLS

(In TG/y)

| | N. Hemisphere | S. Hemisphere | Global | Range |
|---|---|---|---|---|
| Sulfate | 145 | 55 | 200 | 107-374 |
| Man | 106 | 15 | 122 | 69-214 |
| Biosphere | 25 | 32 | 118 | 28-118 |
| Volcanic | 14 | 7 | 21 | 9- 48 |
| Nitrate | 14 | 4 | 18 | 12-27 |
| Man | 12 | 2 | 14 | 10-19 |
| Natural | 2 | 2 | 4 | 2-8 |

From Intergovernmental Panel on Climate Change
Climate Change 2001 Chapter 5. Table 5.4

# ESTIMATES FOR BC & SECONDARY ORGANIC AEROSOLS

NUMBERS VARY – A LOT

DEPENDS ON SOURCES –

BIOMASS BURNING VS 2 CYCLE ENGINE

SECONDARY CHEMISTRY

MONOTERPENES/ISOPRENE/SESQUITERPENES

HUMIC LIKE SUBSTANCES – HULIS

LOT OF WORK TO BE DONE HERE!

AGAIN – LINKS TO HEALTH AND VISIBILITY!

# *Emissions change with time ….*

- **As Global Tech Transfer Grows**
- **S controls….. Sulphate reductions**
- **NOx controls…. Nitrate reductions**
- **Diesel controls… BC reductions**
- **SOA? --- Carbon Dioxide Fertilization vs. Ozone Impacts**

- **Noting Transitions… Urban Controls Lead to Lower Reactivity Precursors – Populations Growing…. Transport and Downwind Production to Regional and Global Scales..**
- **LA Smog – Regional Smog – Global Smog**

# AEROSOLS – CLIMATE CHANGE – PRECIPITATION

## IMPACTED AREAS – BOREAL FORESTS
## MAJOR SOURCES OF ORGANICS
## FEEDBACKS ON ECOSYSTEMS AND AGRICULTURE?

## TROPICAL DISEASES

# CONCLUDING REMARKS

**OF ALL THE CURRENT CLIMATE IMPACTING ACTIVITIES CHANGING AEROSOL LOADINGS SHOULD HAVE MOST IMMEDIATE IMPACTS WHEN ONE CONSIDERS LIFETIMES OF THE SPECIES.**

**I.E. AEROSOLS LESS THAN ONE YEAR**
**GREENHOUSE GASES LONGER**
**METHANE – 10 YEARS…**

**QUESTION: WHAT SIGN IS THE FORCING? HOW MUCH ?**

**WE NEED TO BETTER UNDERSTAND AEROSOL BASICS!**

**WE WILL NEED TO HAVE INTERDISCIPLINARY EFFORTS!**

# ACKNOWLEDGMENTS

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