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Physics and chemistry of the atmosphere

Wagner T.
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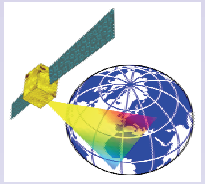

Atmospheric Chemistry and Physics
- brief overview on important effects –

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- Basic properties of the atmosphere
- Greenhouse effect
- Stratospheric chemistry: ozone layer
- Tropospheric chemistry:
    - winter smog
    - summer smog
    - oxidation capacity

Basic properties of the atmosphere: composition

Name	Symbol	Percent by Volume
Nitrogen	N_2	78.084 %
Oxygen	O_2	20.9476 %
Argon	Ar	0.934 %
Carbon Dioxide	CO_2	0.0314 %
Neon	Ne	0.001818 %
Methane	CH_4	0.0002 %
Helium	He	0.000524 %
Krypton	Kr	0.000114 %
Hydrogen	H_2	0.00005 %
Water vapor	H2O	typically near surface: 1-4% (average over full atmosphere: 0.4%)

Trace gases are important. They control:

-radiation: absorption/emission

-chemical reactions

-energy supply for organisms

-energy transport (latent heat)

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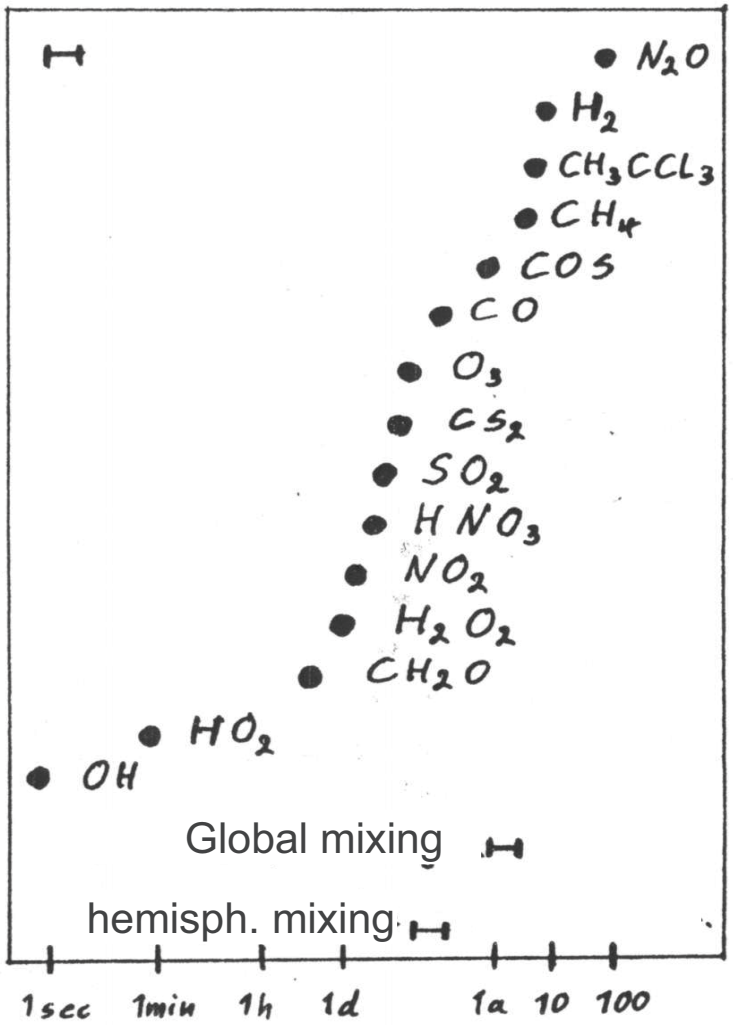
Residence time of various atmospheric trace gases



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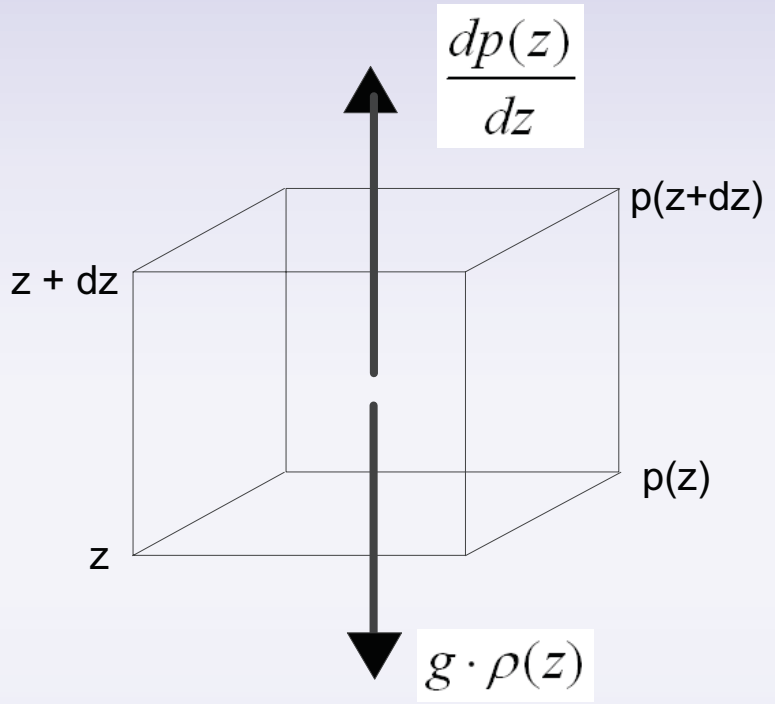
Basic properties of the atmosphere: pressure profile

$$-g \cdot \rho(z) = \frac{dp(z)}{dz}$$ Hydrostatic equation

$$p(z) = p(0) \cdot e^{-\frac{z}{z_0}}$$



Scale height $z_{0:}$ $\frac{RT}{Mg} = z_0 \approx 8\text{km}$

=> Scale height is different for different molecules!

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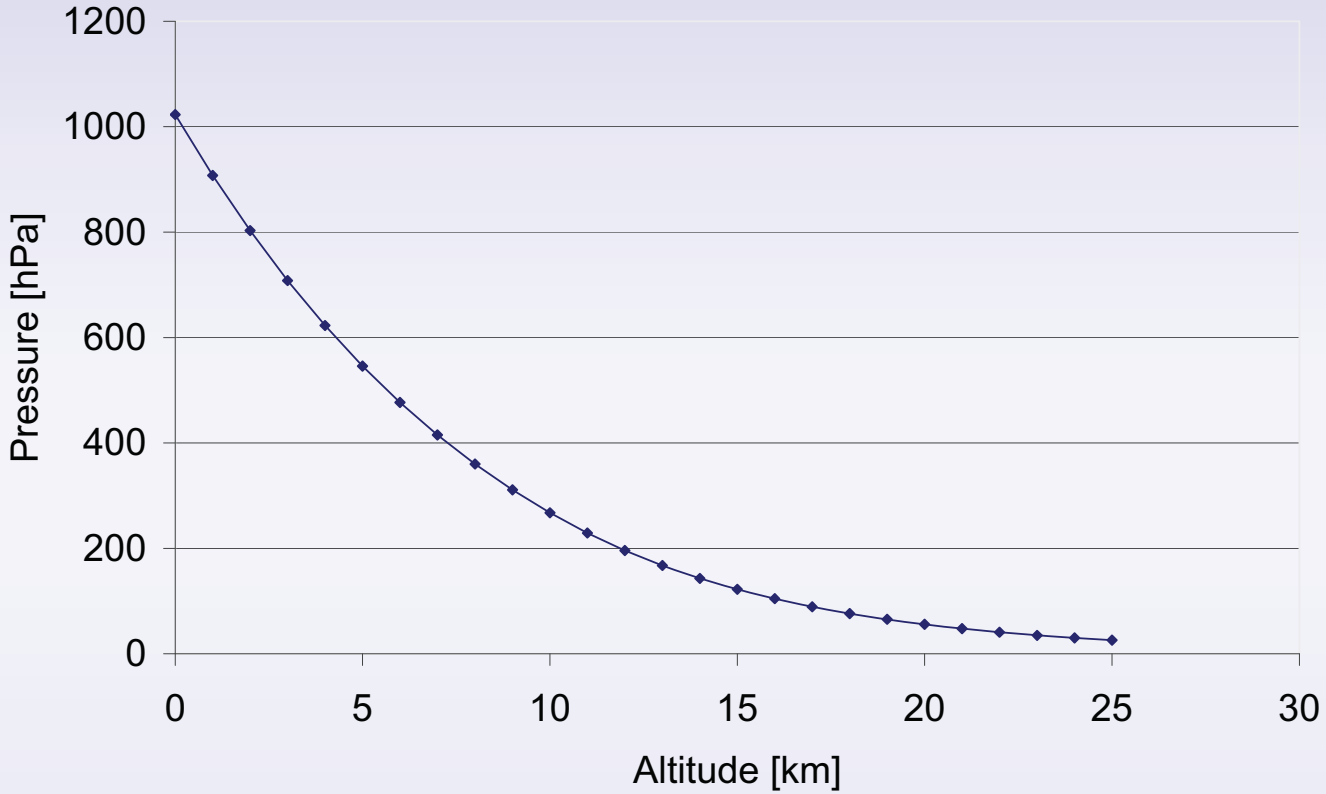
$$p(z) = p(0) \cdot e^{-\frac{z}{z_0}}$$

US std. atmosphere



% of total atmosperic mass	50	90	99	99.9	99.99
is below [km]	5.5	16.2	31.2	48.3	66.0

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Vertical temperature profile

-dry adiabatic temperature gradient: $\frac{dT}{dz} = -0.00981K/m \approx 1K/100m$

-moist adiabatic temperature gradient:

If air ascends and cools, it will eventually reach the temperature at which water vapor condenses. The condensation process releases latent heat which provides energy to the air parcel. Thus, the temperature decrease with height is smaller than for the dry adiabatic temperature gradient.

The typical temparture gradient in the lower atmospere (up to about 10km) is determined by the:

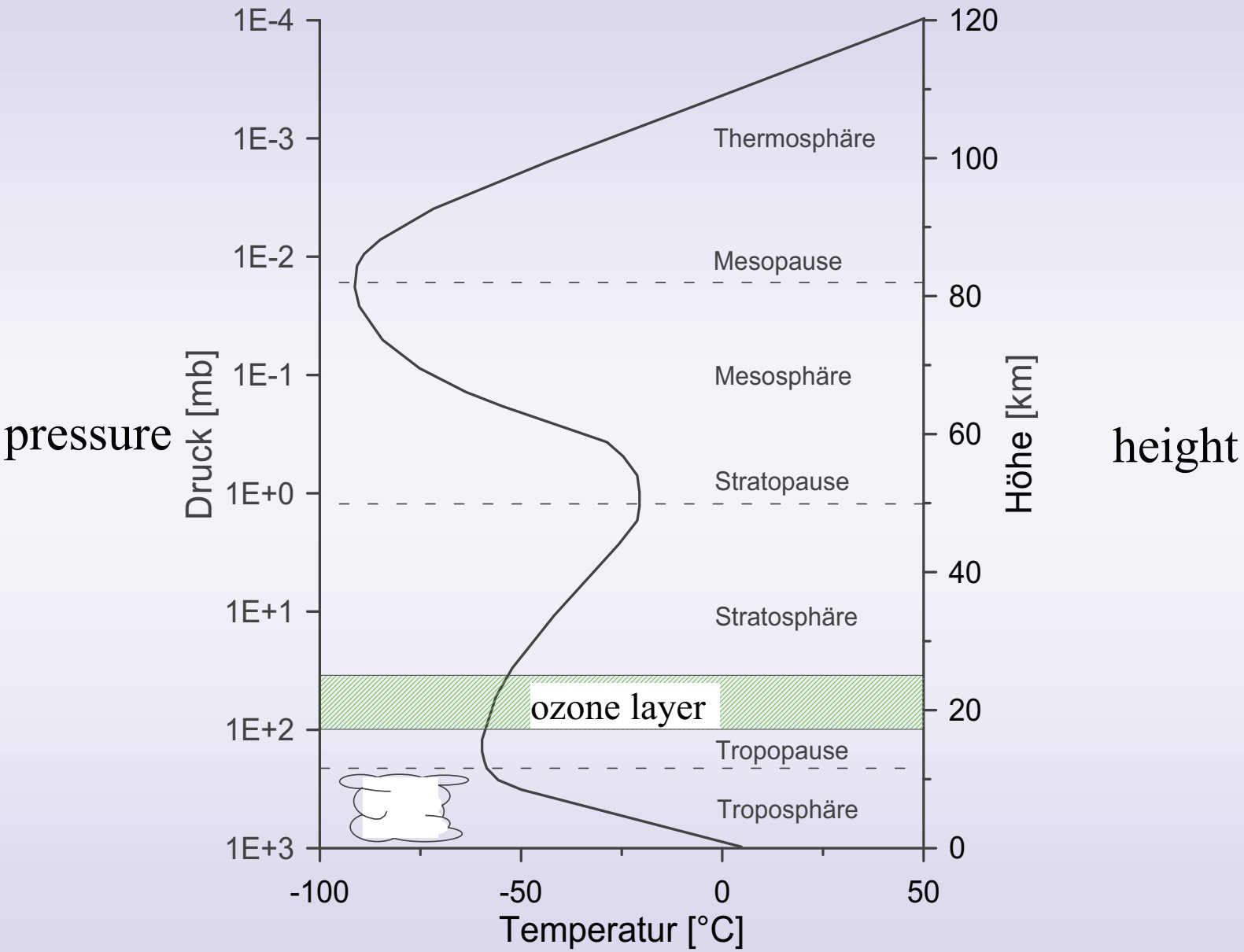
-moist adiabatic temperature gradient. Typical values are ≈0.6K/100m

Long time it was known (e.g. from mountain climbing) that the temperature decreases with height. It was also assumed that the temperature decrease would continue until the ‚upper edge' of the atmosphere.

About more than 100 years ago, several important observations were made, which showed that the actual temperature gradient differs from this expectation:

- 1880 - 1908: Discovery of the ozone absorption (especially in the UV)
- 1902: Balloon borne temperature measurements (Teisserenc de Bort and Richard Assmann) showed a temperature increase at about 11km.
- 1913: Balloon borne observations (Wigand) of the ozone absorption show no significant decrease of the UV intensity up to 10km
- 1926: Umkehr-Effekt (Paul Götz): Maximum of the ozone layer at about 25km

Basic properties of the atmosphere: temperature profile



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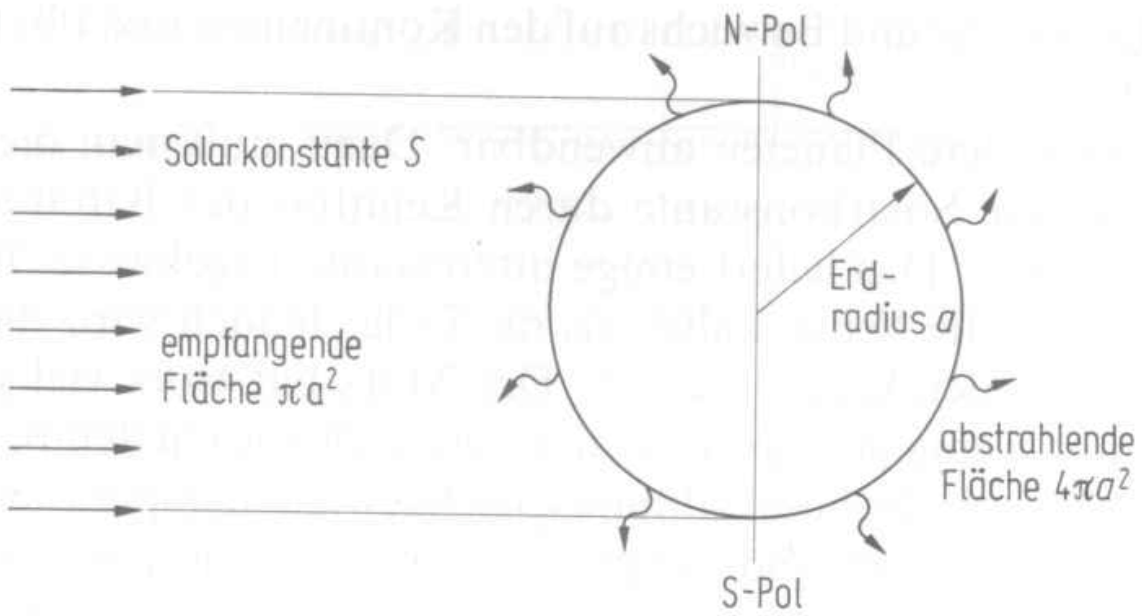
Greenhouse effect



The solar irradiation is S = 1368 W/m². The earth cuts a cross section of πr^2 off the solar beam. It reflects a fraction A (albedo) back into space. The absorbed power $(1-A)S\pi r^2$ has to be compensated by the terrestrial IR radiation to avoid a heating of the earth.

From the Stefan-Boltzmann-law it follows that the terrestrial radiation is $\sigma T^4 \cdot 4\pi r^2$

It follows: $T_{average} = -18°C$

but $T_{real} = +15°C$ Greenhouse effect

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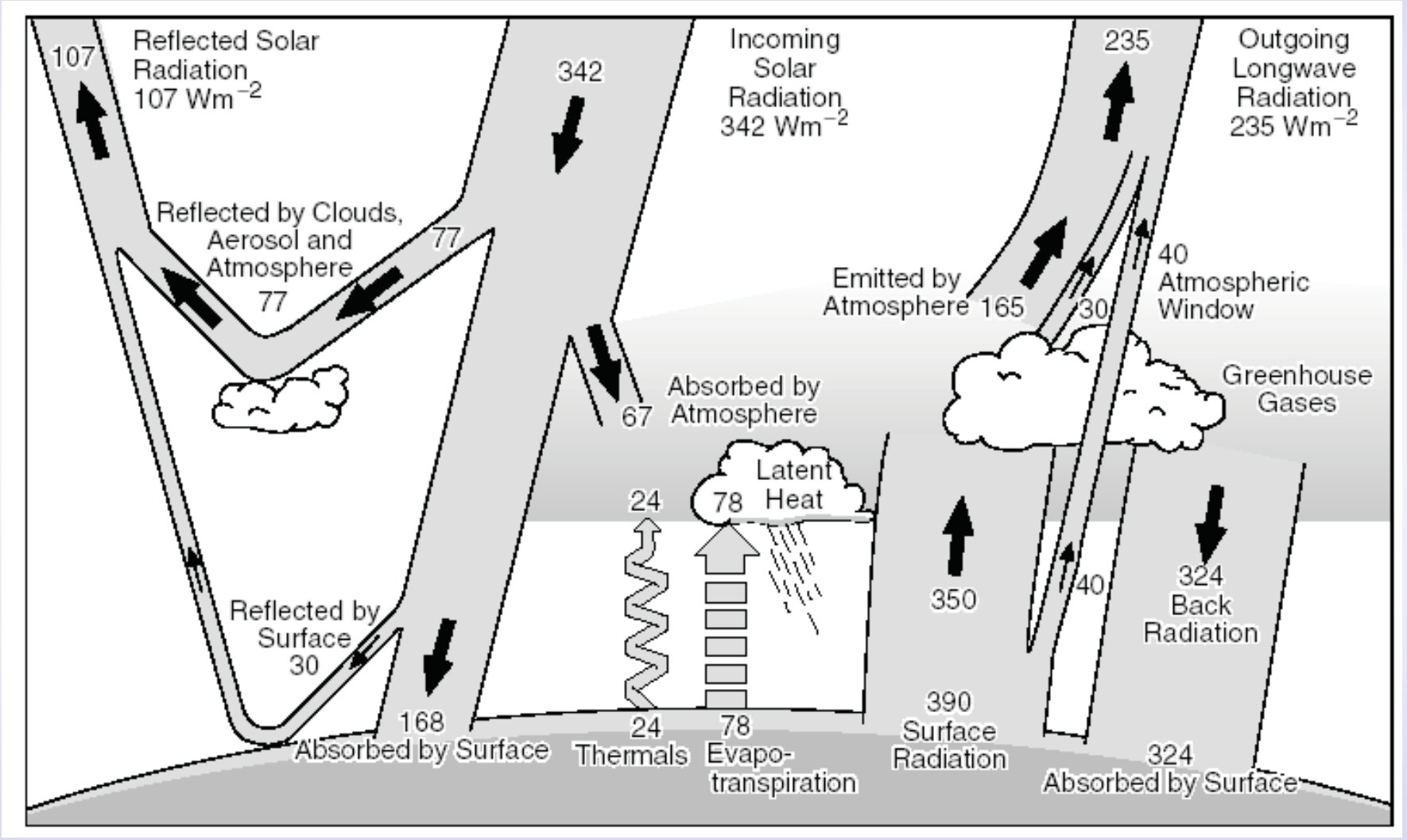
Solar irradiance = 1368 W/m² on average: 1368 W/m² / 4 =342 W/m²

Atmospheric radiation budget



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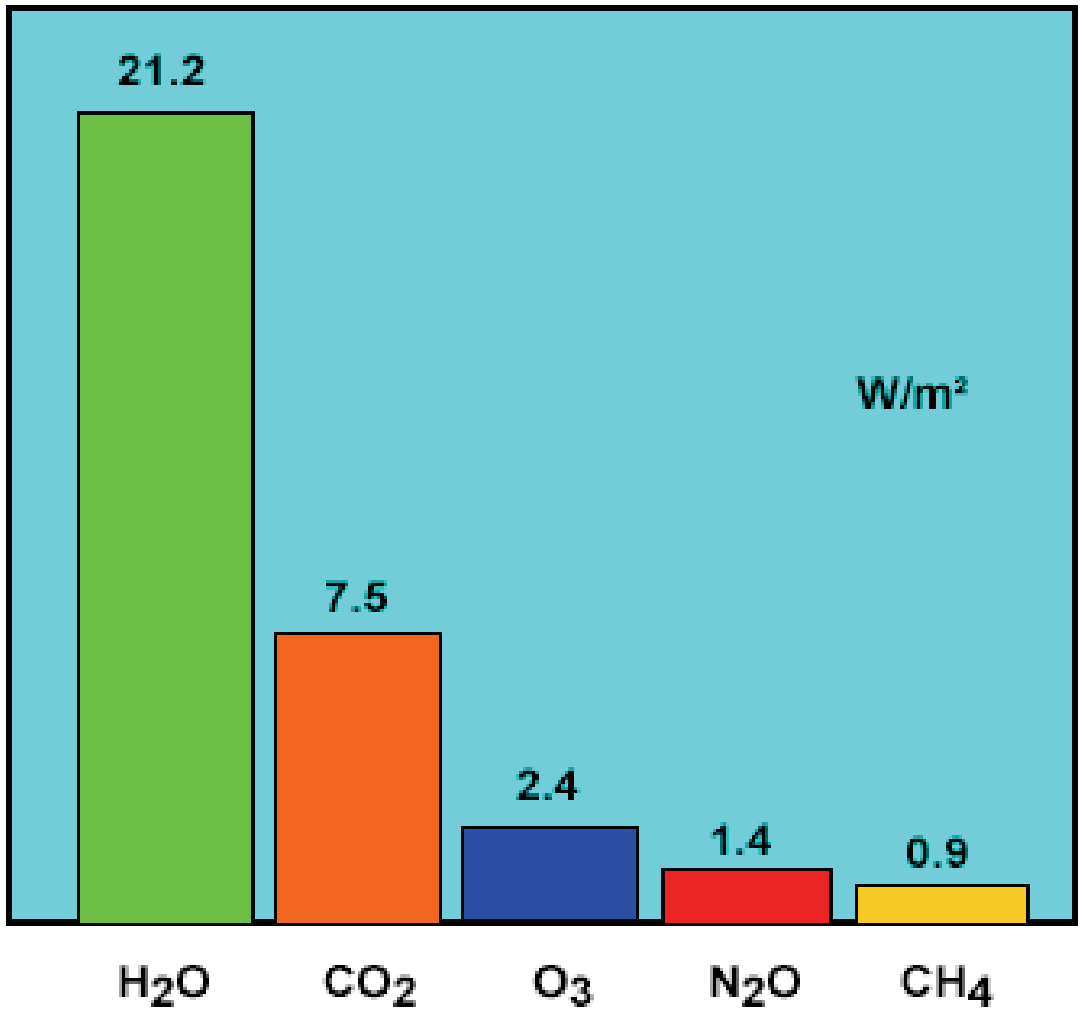
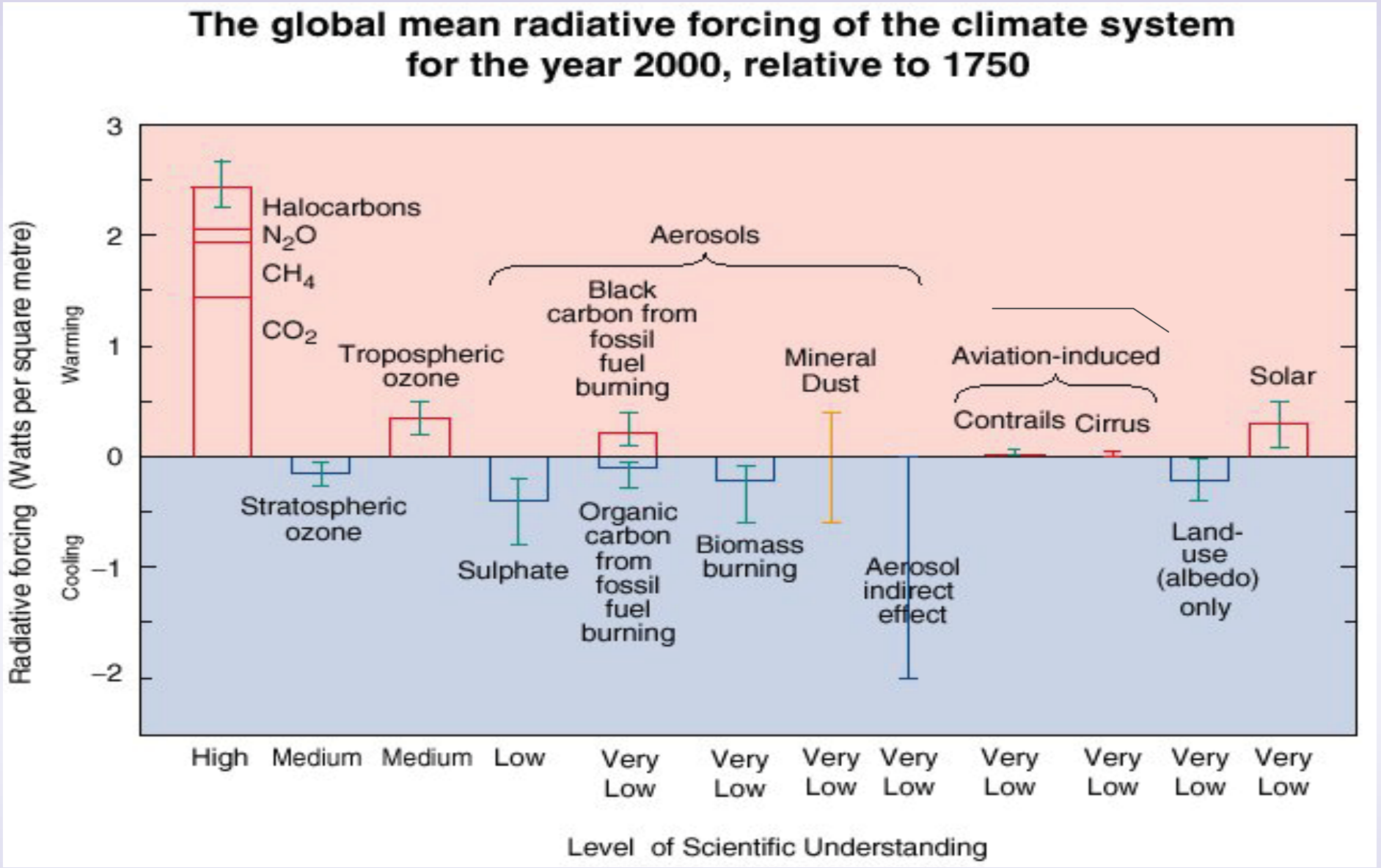


Contribution of different greenhouse gases to the natural greenhouse effect (IPCC, 1995)



(C) ICCP

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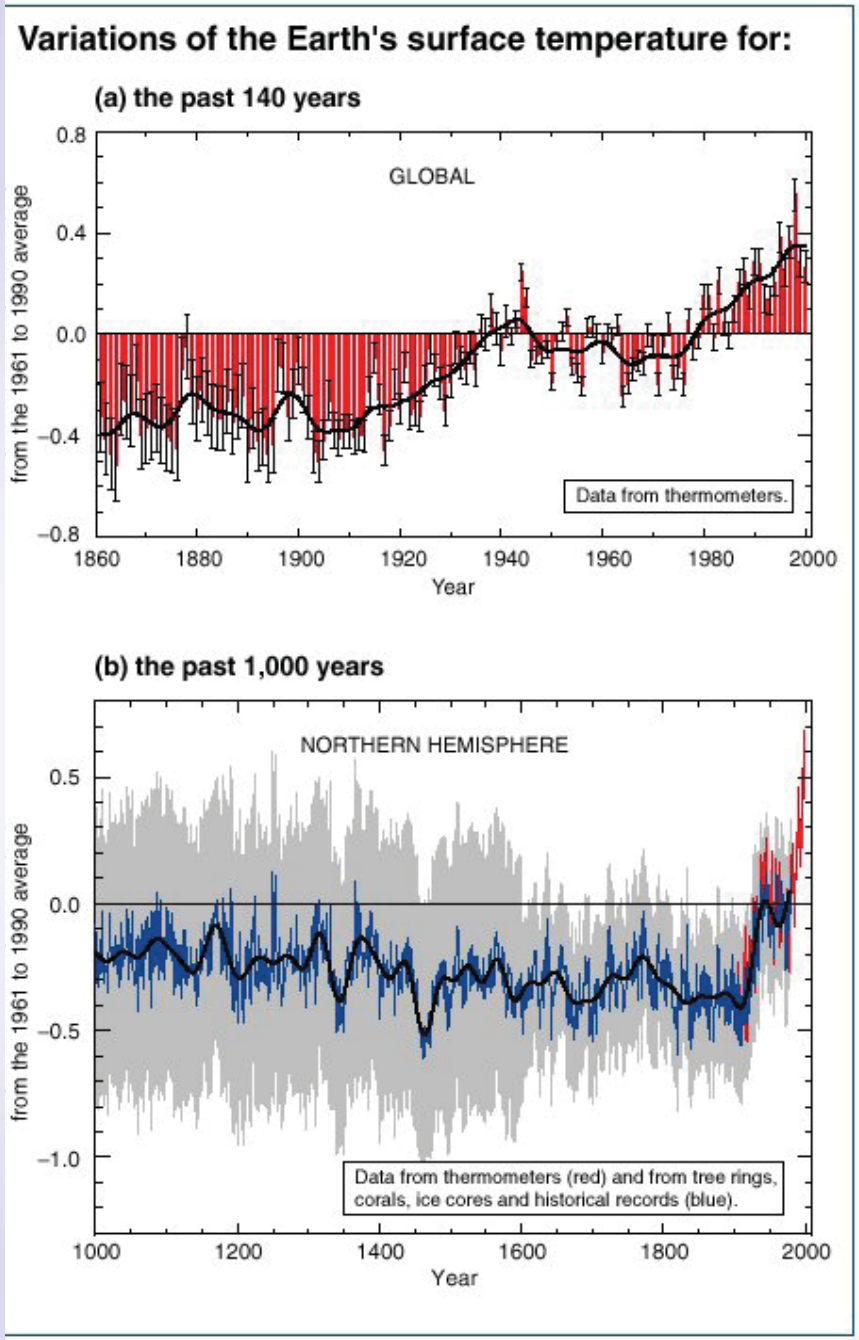

Figure 1: Variations of the Earth's surface temperature over the last 140 years and the last millennium.

(a) The Earth's surface temperature is shown year by year (red bars) and approximately decade by decade (black line, a filtered annual curve suppressing fluctuations below near decadal time-scales). There are uncertainties in the annual data (thin black whisker bars represent the 95% confidence range) due to data gaps, random instrumental errors and uncertainties, uncertainties in bias corrections in the ocean surface temperature data and also in adjustments for urbanisation over the land. Over both the last 140 years and 100 years, the best estimate is that the global average surface temperature has increased by 0.6 ± 0.2°C.

(b) Additionally, the year by year (blue curve) and 50 year average (black curve) variations of the average surface temperature of the Northern Hemisphere for the past 1000 years have been reconstructed from "proxy" data calibrated against thermometer data (see list of the main proxy data in the diagram). The 95% confidence range in the annual data is represented by the grey region. These uncertainties increase in more distant times and are always much larger than in the instrumental record due to the use of relatively sparse proxy data. Nevertheless the rate and duration of warming of the 20th century has been much greater than in any of the previous nine centuries. Similarly, it is likely[7] that the 1990s have been the warmest decade and 1998 the warmest year of the millennium.

[Based upon (a) Chapter 2, Figure 2.7c and (b) Chapter 2, Figure 2.20]

Report of the Intergovernmental Panel of Climate Change (IPCC), http://www.ipcc.ch/

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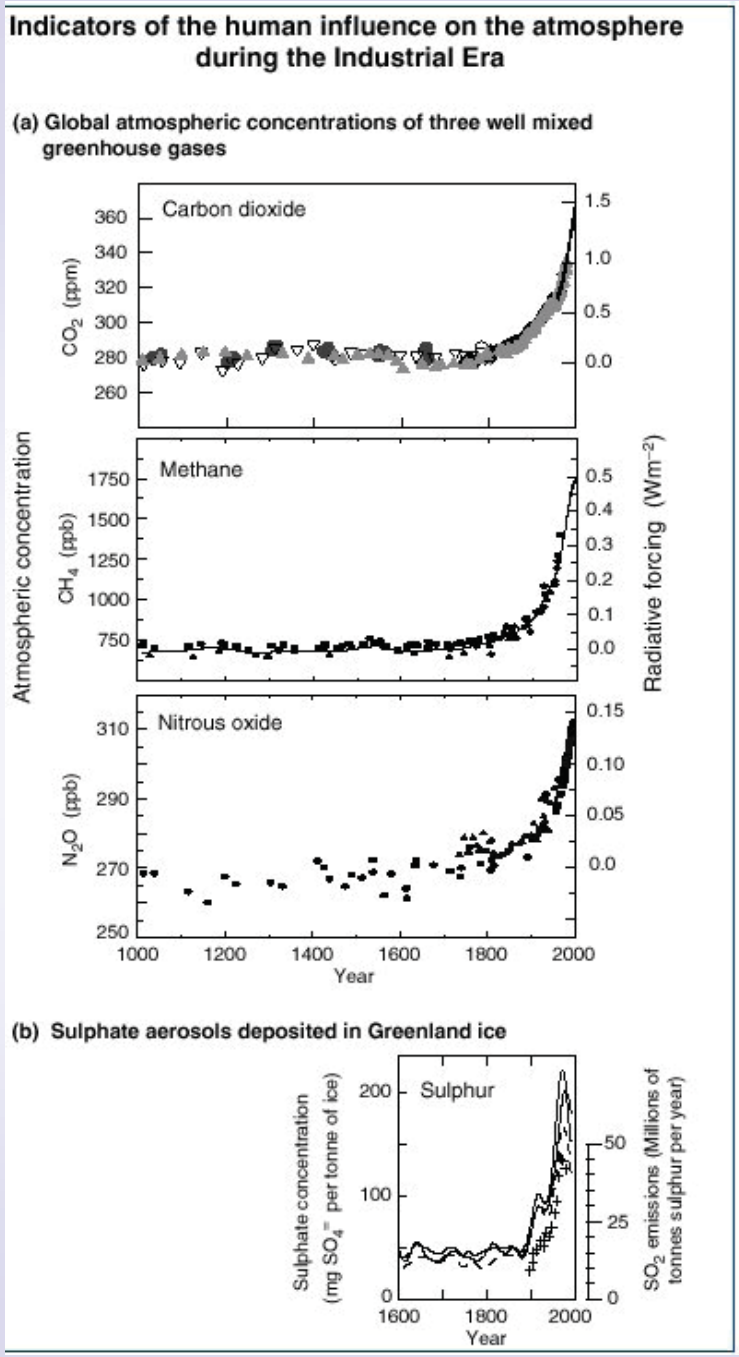
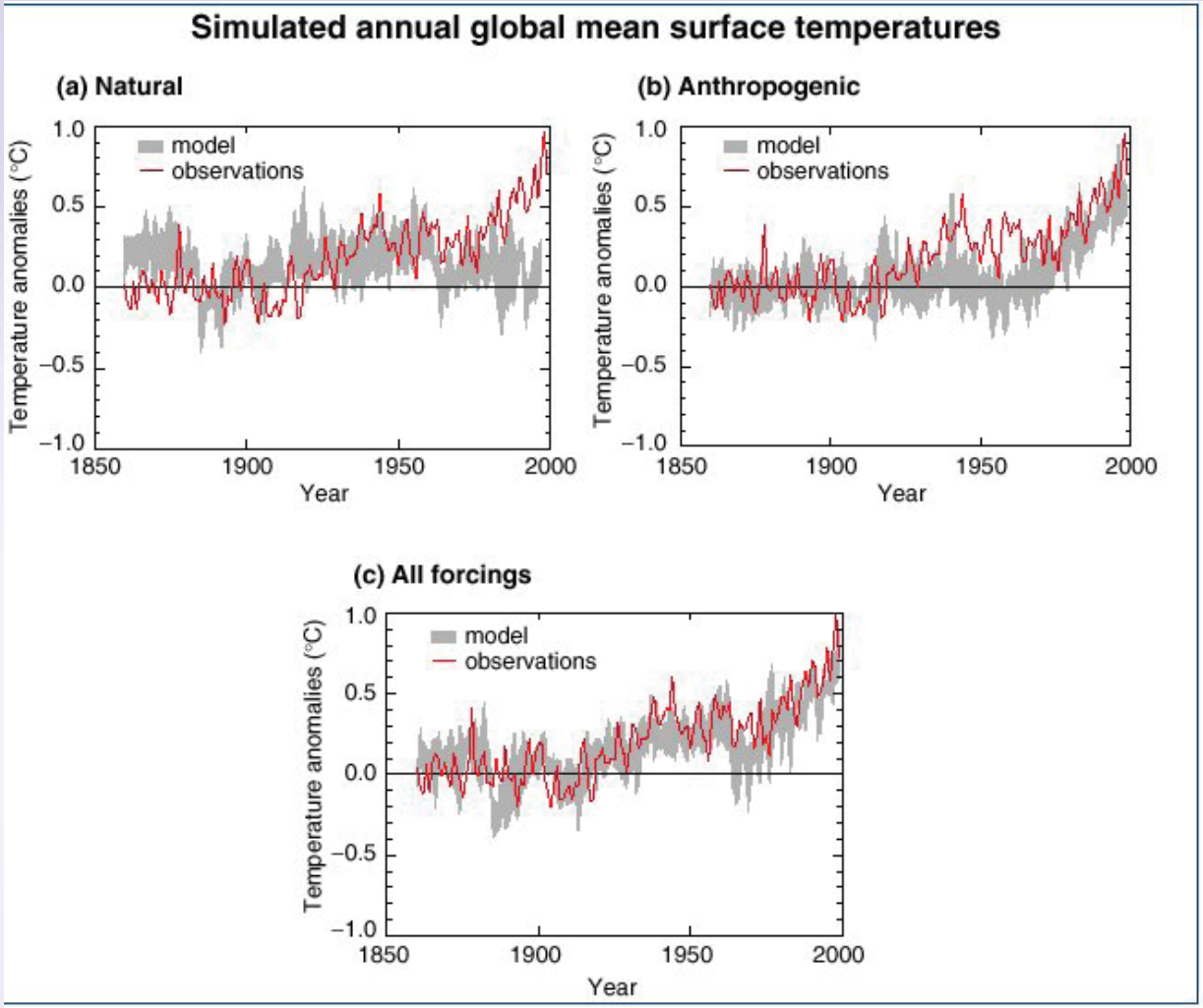


Figure 2: Long records of past changes in atmospheric composition provide the context for the influence of anthropogenic emissions.

(a) shows changes in the atmospheric concentrations of carbon dioxide (CO_2), methane (CH_4), and nitrous oxide (N_2O) over the past 1000 years. The ice core and fim data for several sites in Antarctica and Greenland (shown by different symbols) are supplemented with the data from direct atmospheric samples over the past few decades (shown by the line for CO_2 and incorporated in the curve representing the global average of CH_4). The estimated positive radiative forcing of the climate system from these gases is indicated on the right-hand scale. Since these gases have atmospheric lifetimes of a decade or more, they are well mixed, and their concentrations reflect emissions from sources throughout the globe. All three records show effects of the large and increasing growth in anthropogenic emissions during the Industrial Era.

(b) illustrates the influence of industrial emissions on atmospheric sulphate concentrations, which produce negative radiative forcing. Shown is the time history of the concentrations of sulphate, not in the atmosphere but in ice cores in Greenland (shown by lines; from which the episodic effects of volcanic eruptions have been removed). Such data indicate the local deposition of sulphate aerosols at the site, reflecting sulphur dioxide (SO_2) emissions at mid-latitudes in the Northern Hemisphere. This record, albeit more regional than that of the globally-mixed greenhouse gases, demonstrates the large growth in anthropogenic SO_2 emissions during the Industrial Era. The pluses denote the relevant regional estimated SO_2 emissions (right-hand scale).

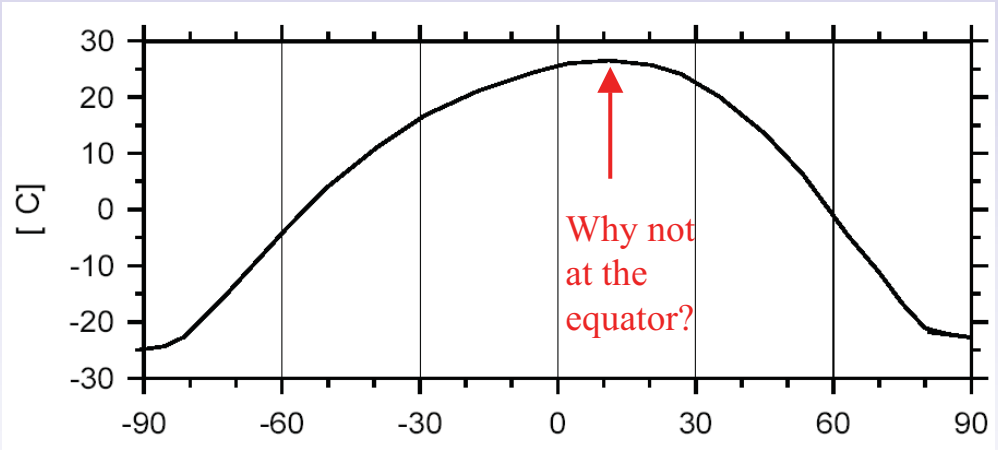
[Based upon (a) Chapter 3, Figure 3.2b (CO_2); Chapter 4, Figure 4.1a and b (CH_4) and Chapter 4, Figure 4.2 (N_2O) and (b) Chapter 5, Figure 5.4a]



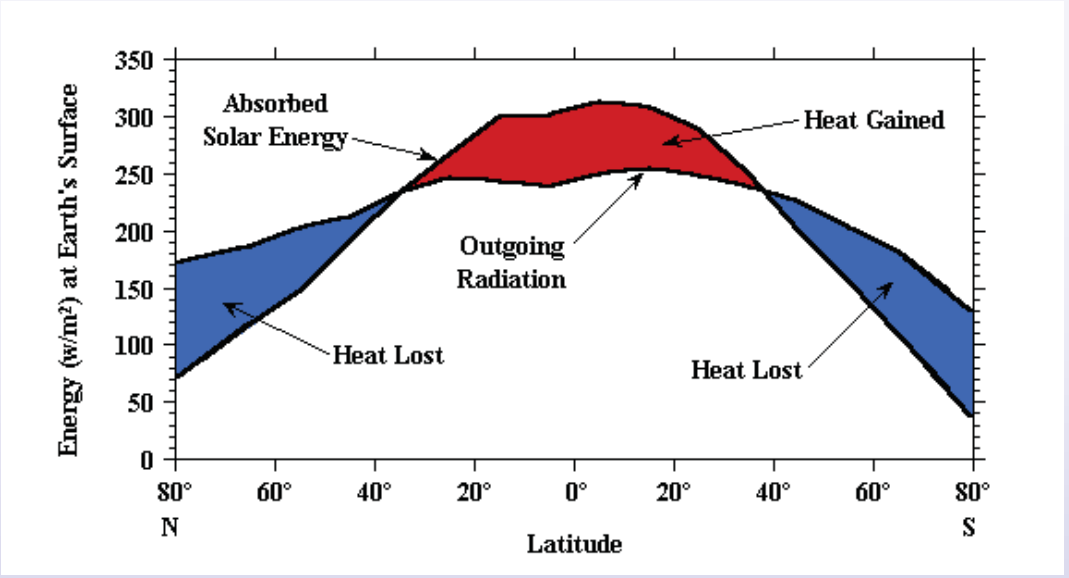
re 4: Simulating the Earth's temperature variations, and comparing the results to measured changes, can provide insight into the erlying causes of the major changes.

nate model can be used to simulate the temperature changes that occur both from natural and anthropogenic causes. The simulations sented by the band in (a) were done with only natural forcings: solar variation and volcanic activity. Those encompassed by the band in (b) were with anthropogenic forcings: greenhouse gases and an estimate of sulphate aerosols, and those encompassed by the band in (c) were done with natural and anthropogenic forcings included. From (b), it can be seen that inclusion of anthropogenic forcings provides a plausible explanation substantial part of the observed temperature changes over the past century, but the best match with observations is obtained in (c) when both ral and anthropogenic factors are included. These results show that the forcings included are sufficient to explain the observed changes, but do xclude the possibility that other forcings may also have contributed. The bands of model results presented here are for four runs from the same el. Similar results to those in (b) are obtained with other models with anthropogenic forcing. [Based upon Chapter 12, Figure 12.7]

Global circulation patterns redistribute energy



Average near surface temperature as function of latitude



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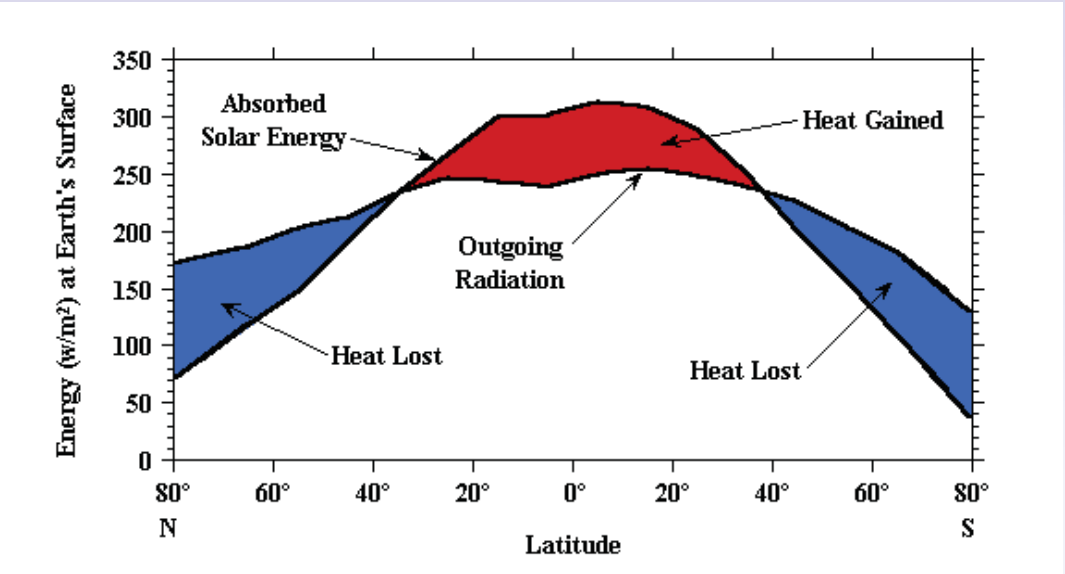
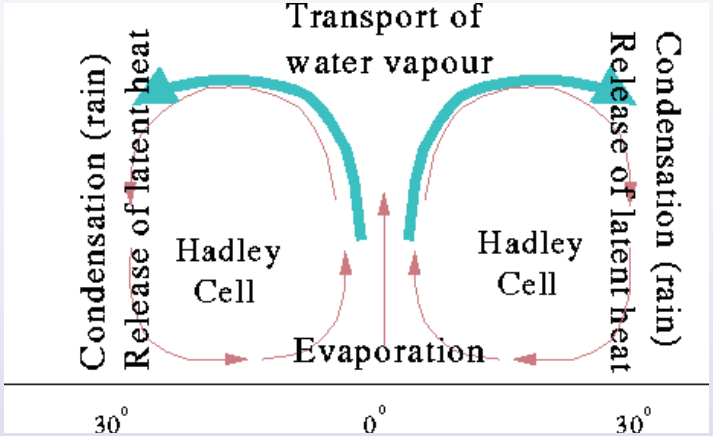
Global circulation patterns redistribute energy





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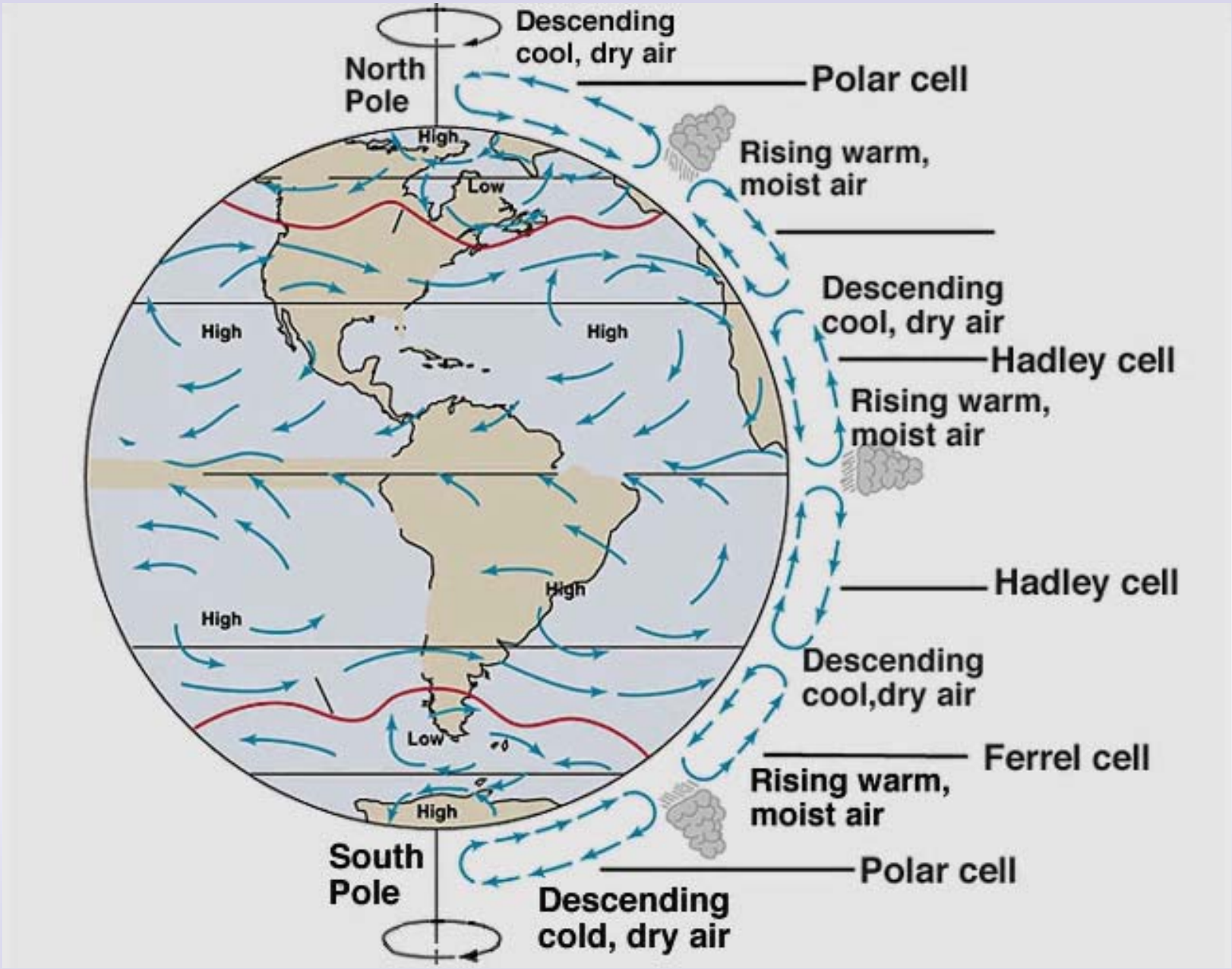

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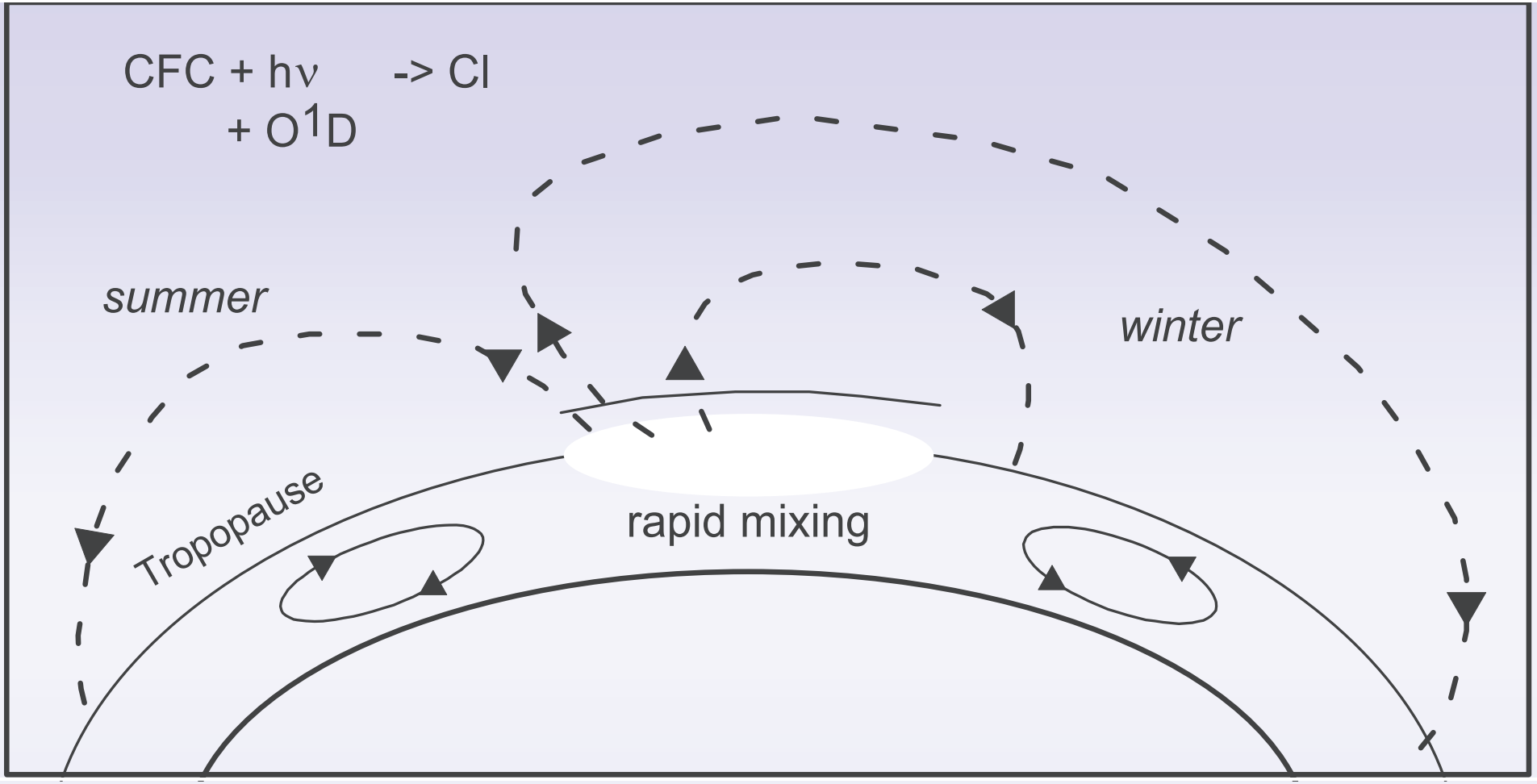

Schematic diagram of the Brewer-Dobson Circulation (adapted from Solomon et al. [1998]). Because the stratosphere contains only about 10% of the total atmosphere, the circulation must turn over many times to destroy all of the CFCs present, resulting in a long atmospheric live time of these species.

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Ozone in the stratosphere

Chapman Cycle [1930].

$O_2 + h\nu$	$\rightarrow$	$2O$	$(\lambda \leq 240nm)$	Odd oxygen is produced
$O + O_2 + M$	$\rightarrow$	$O_3 + M$		
$O_3 + h\nu$	$\rightarrow$	$O(^1D)$	$(\lambda \leq 320nm)$	Odd oxygen is conserved
$O(^1D) + M$	$\rightarrow$	$O + M$		
$O_3 + h\nu$	$\rightarrow$	$O + O_2$	$(\lambda \leq 1180nm)$	
$O + O + M$	$\rightarrow$	$O_2 + M$		Odd oxygen is destroyed
$O + O_3$	$\rightarrow$	$2O_2$		

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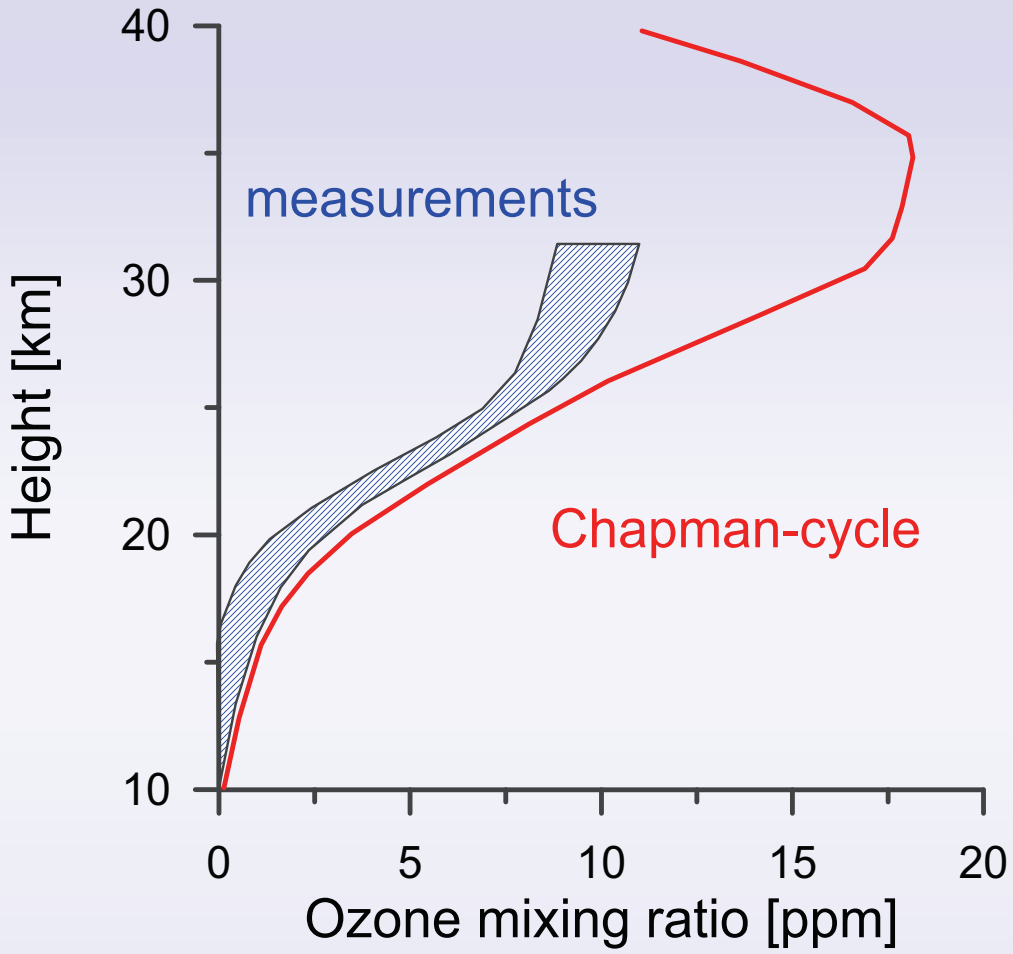

Comparison of measured ozone profiles and modelled ones taking into account only the reactions of the Chapman-cycle (adapted from Röth [1994]).

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Additional (catalytic) Ozone destruction mechanisms:

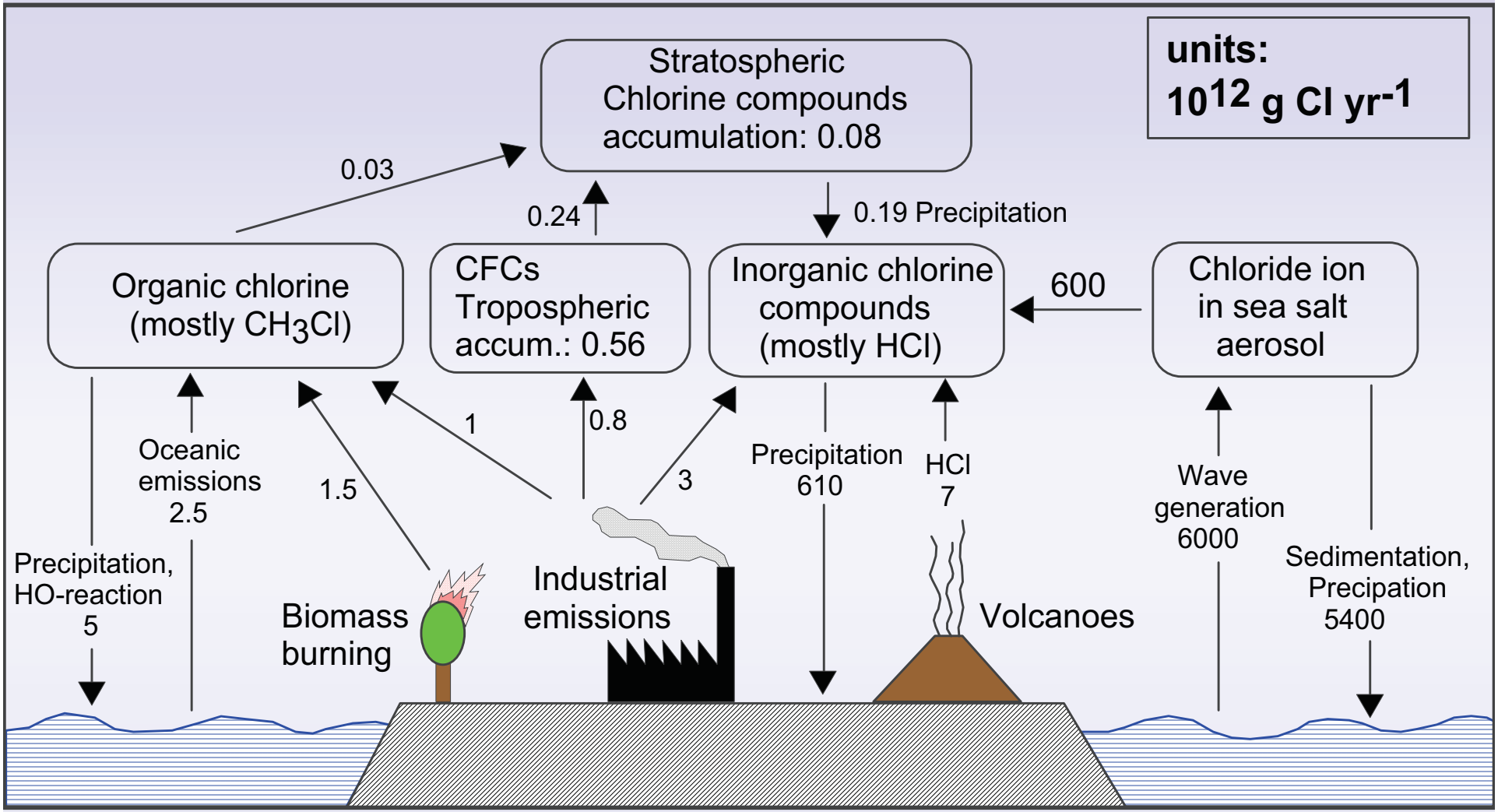
$$X\bullet + O_3 \rightarrow XO\bullet + O_2$$

$$XO\bullet + O \rightarrow X\bullet + O_2$$

Net: $O + O_3 \rightarrow 2O_2$

with:

X = OH, NO, Cl, Br



Global atmospheric chlorine cycle (adapted from Graedel and Crutzen [1993]).

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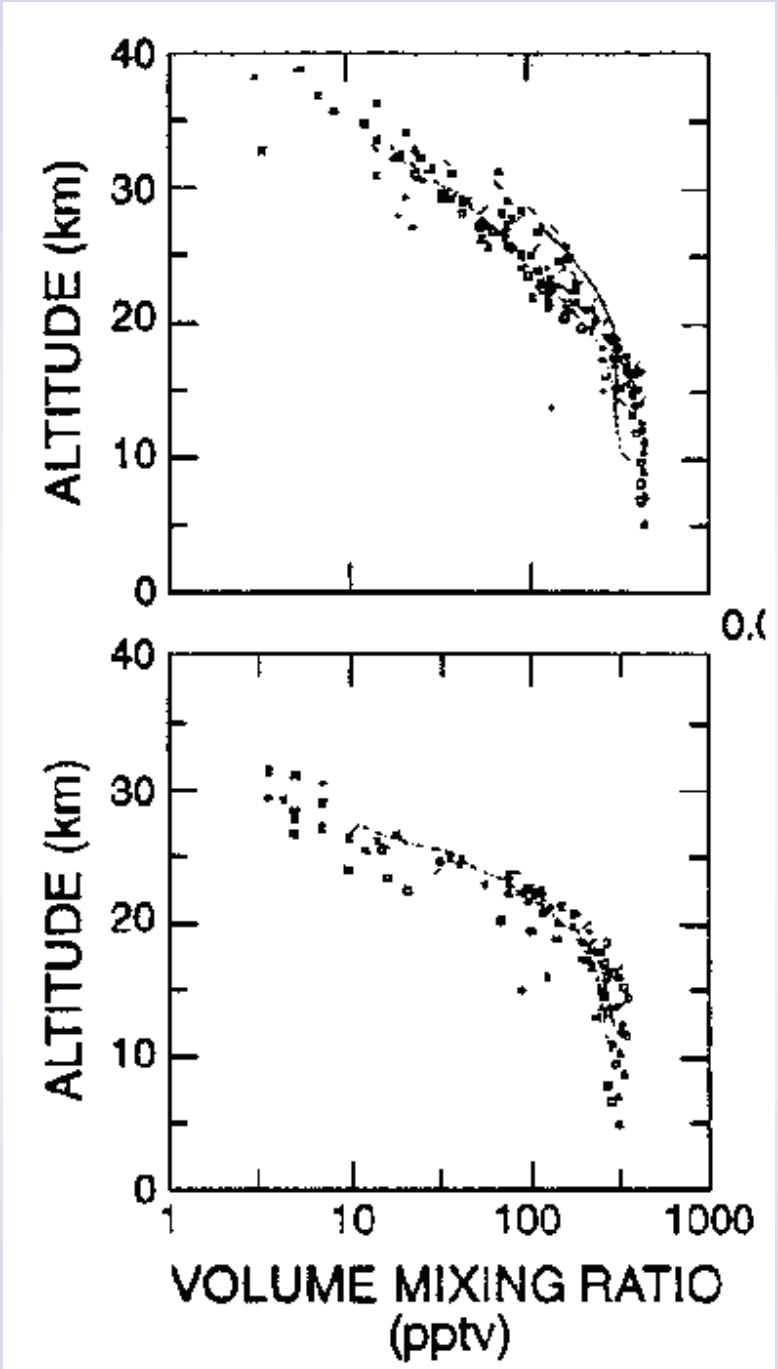

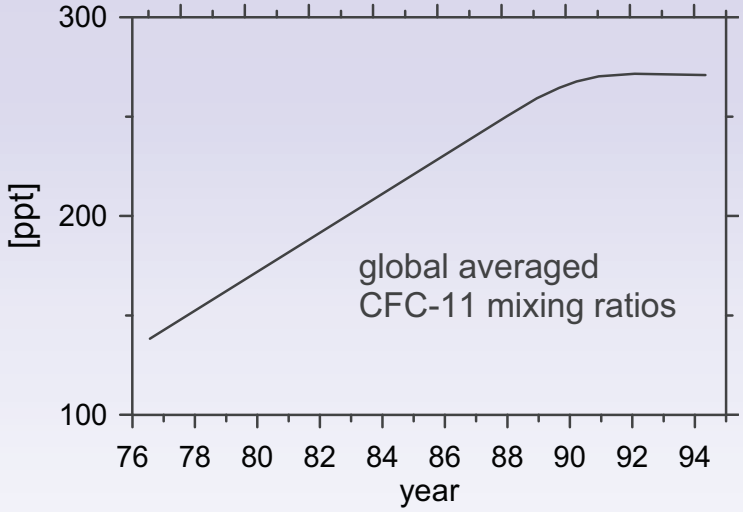

Global averaged increase of CFC-11 (adapted from IPCC [1996]).

Altitude profiles of CFC-11 (bottom) and CFC-12 (top) [NASA, 1994].

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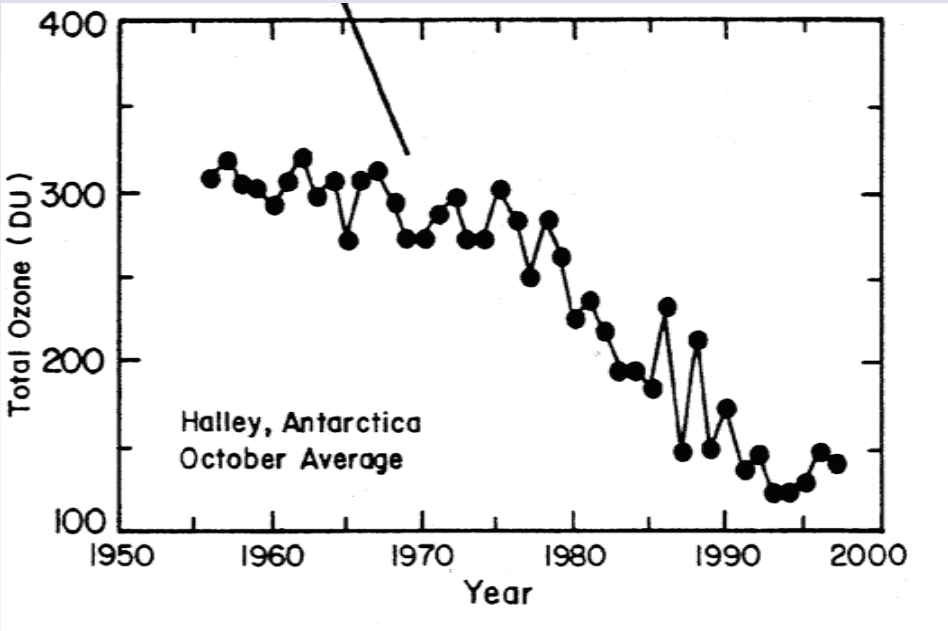
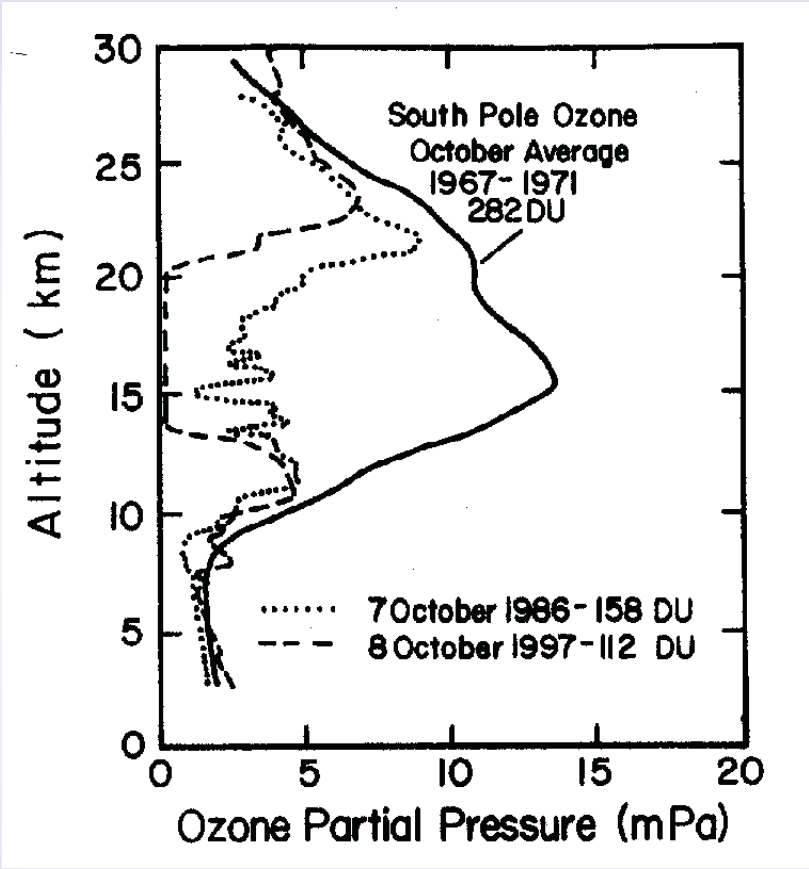
Ozone hole, Antarctica

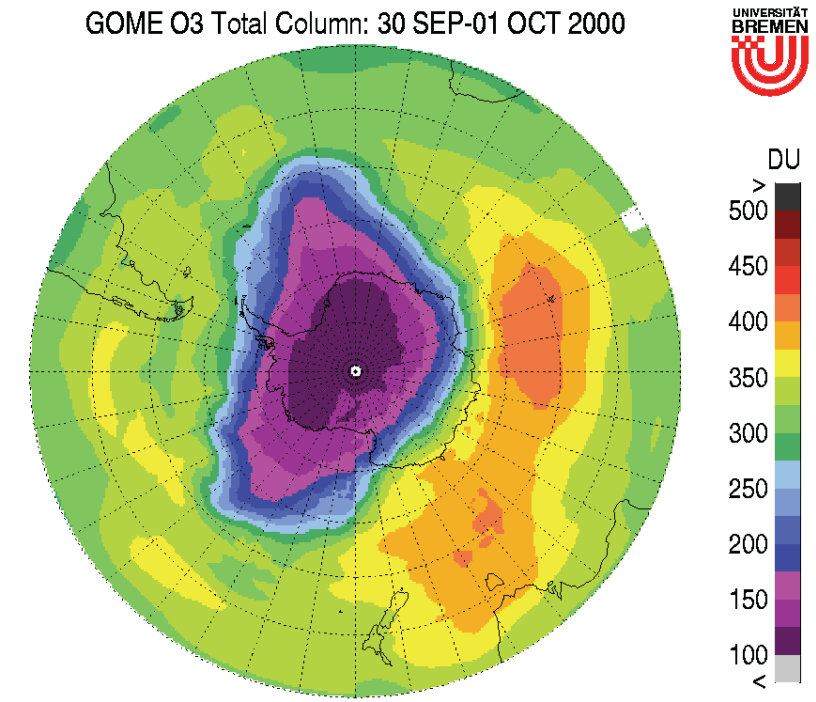


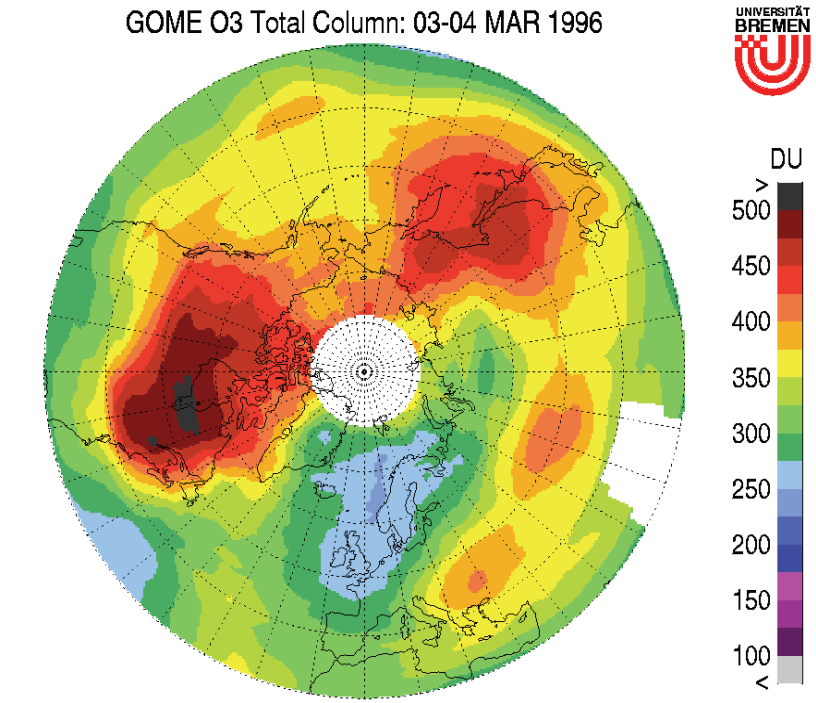


Ground based measurements,
Halley, Antarctis
Farman et al., 1986,
Jones and Shanklin, 1995

Over Antarctica, the ozone hole occurs regularly

Over the Arctic, in some years a ‚small' ozone hole occurs





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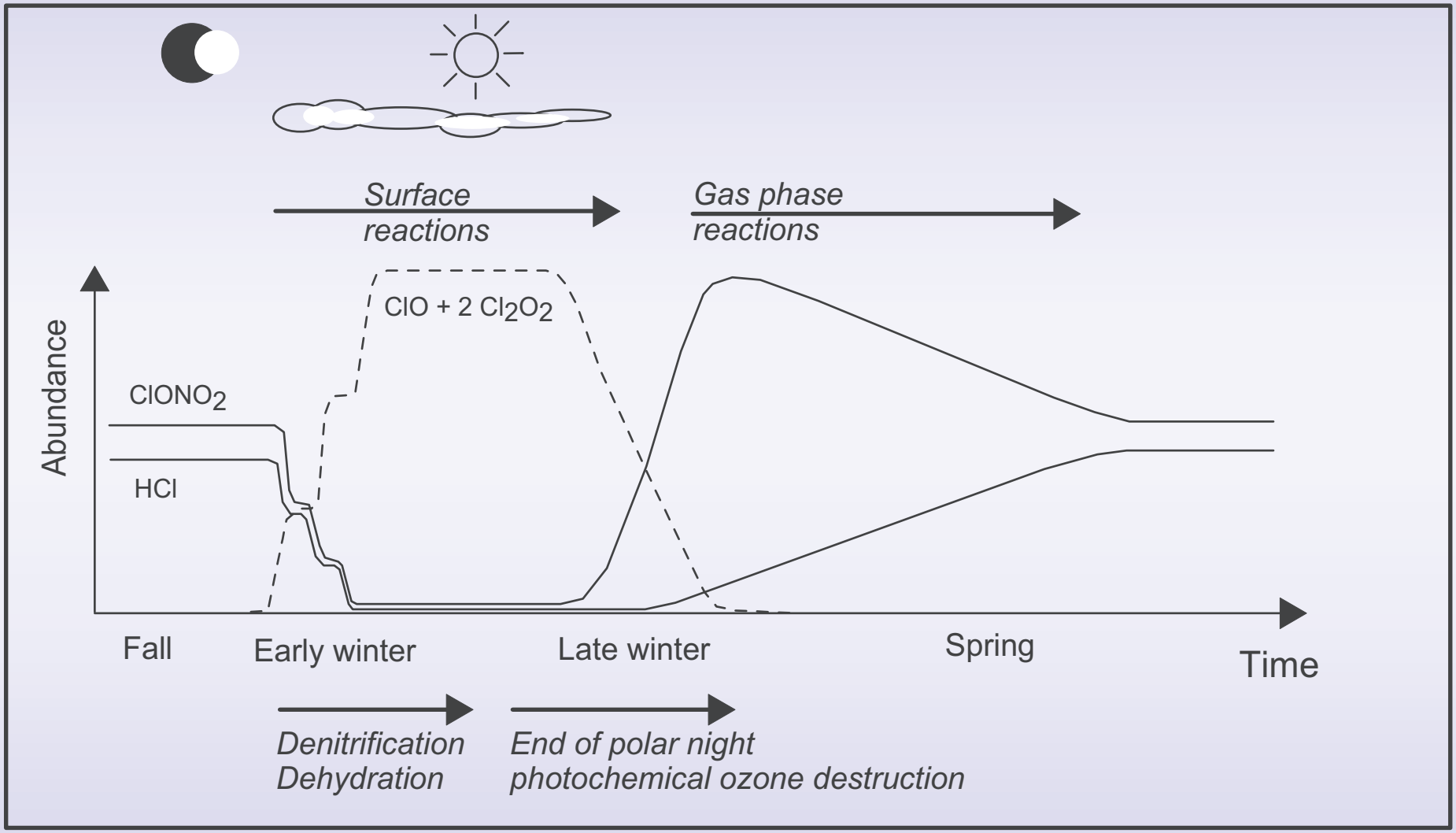
....what has been ignored so far...

Polar Stratospheric Clouds over Kiruna (Sweden), ©Carl-Fredrik Enell.

Dynamical and photochemical development in the stratosphere during polar winter



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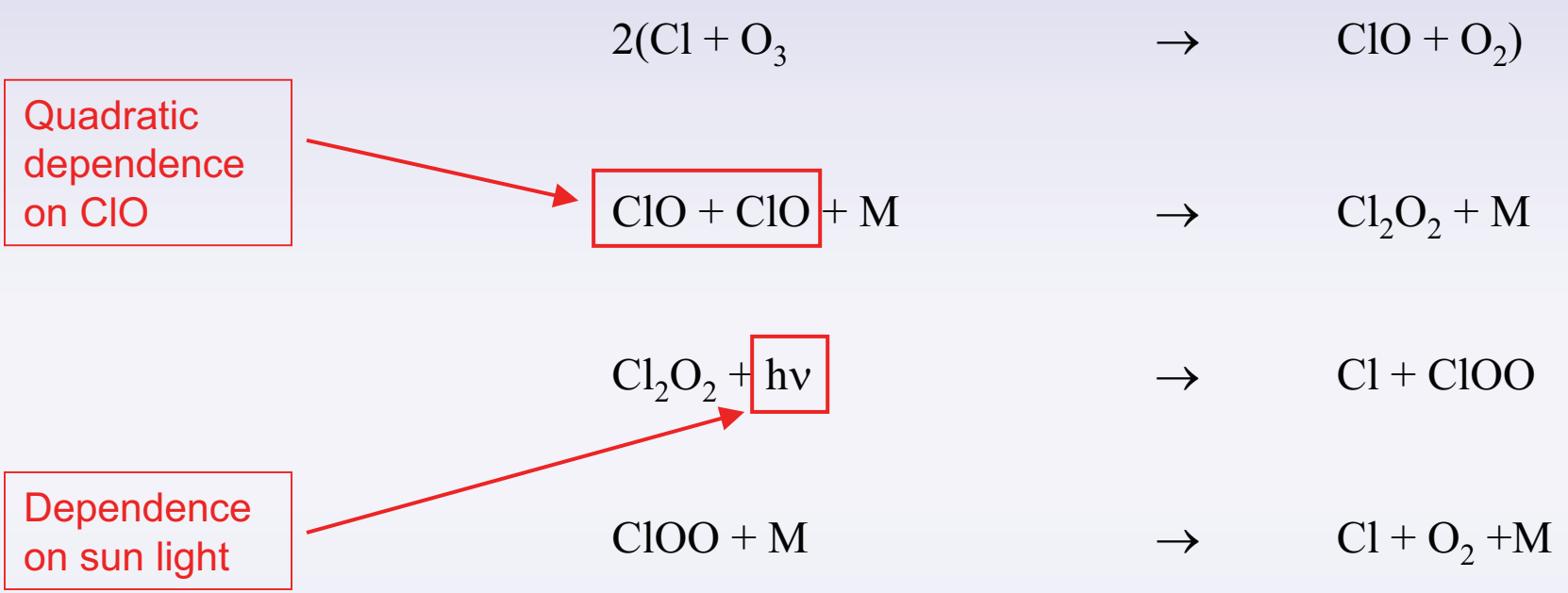
Catalytic ozone destruction cycle through chlorine



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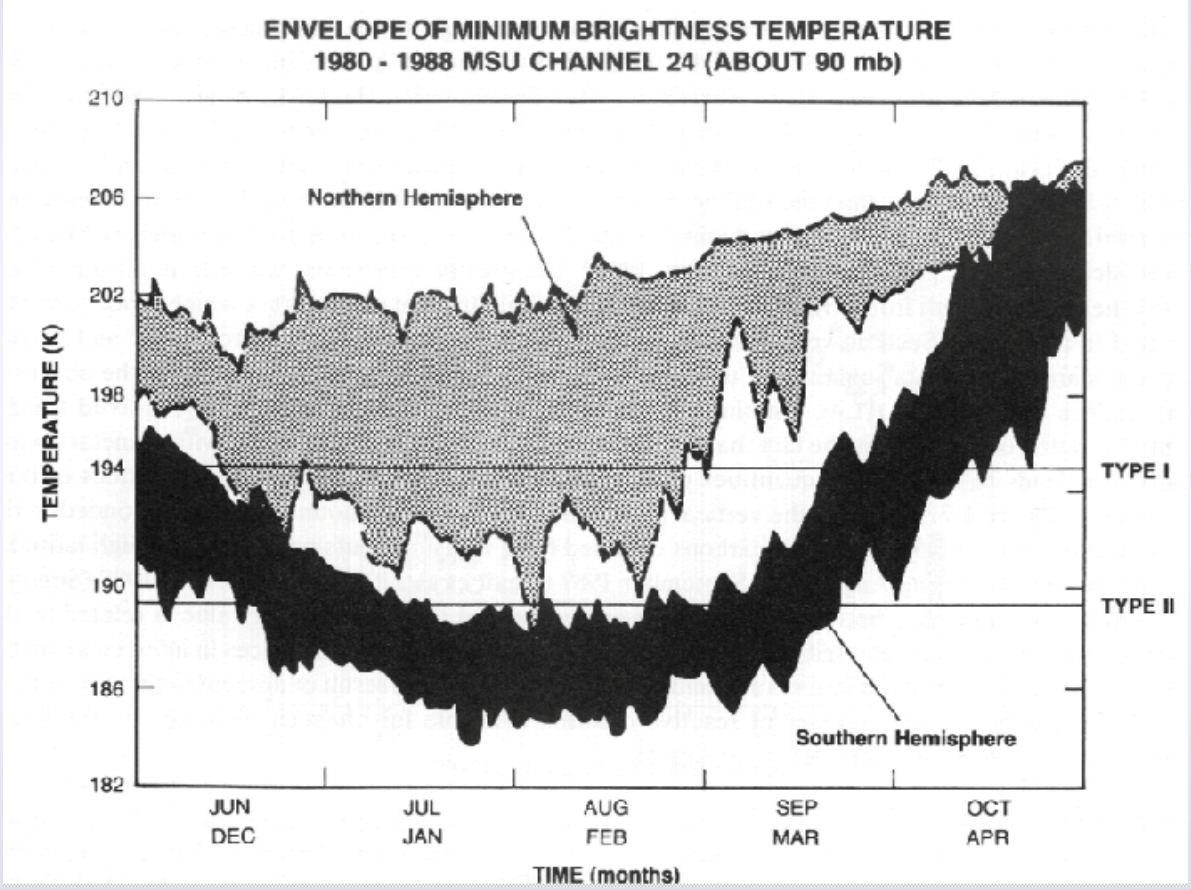
Temperature differences between both hemispheres



Envelope of minimum temperature 1980-1988 at about 90 mb from MSU measurements [WMO 1991].

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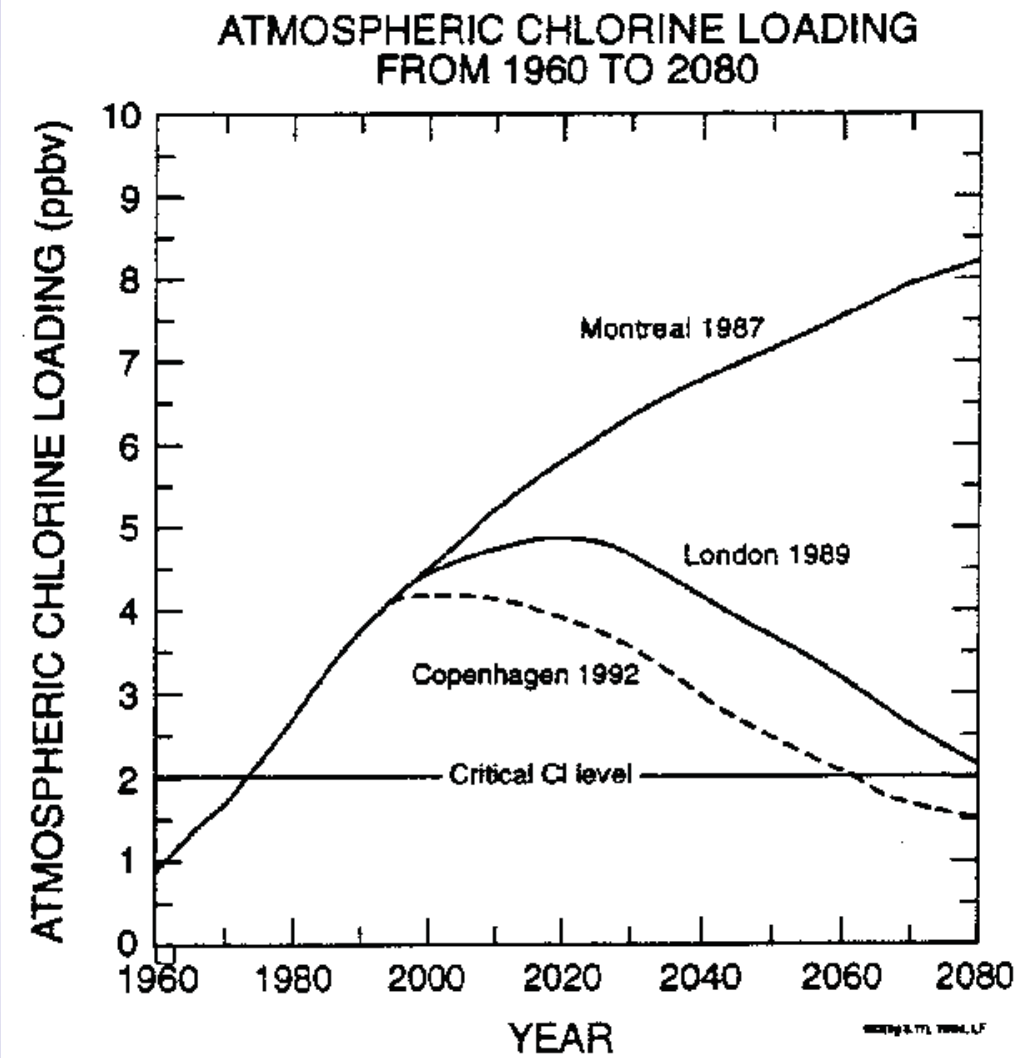

Predicted future atmospheric burden of chlorine (adapted from Brasseur [1995]).

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Questions:

- When will ozone hole close?
  (influence of climate change)
- How important are bromine compounds?
- Will there be an ozone hole over the Arctic?
- How strong does ozone change in mid-latiudes?

’London Smog’ (John Evelyn, Fumifugium, 17th century)

‘It is this horried smoake which obscures our church and makes our places look old, which fouls our cloth and corrupts the waters, so as the very rain, and refreshing dews which fall in the several seasons, precipitate to impure vapour, which, with its black and tenacious quality, spots and contaminates whatever is exposed to it.

But without the use of calculations it is evident to every one who looks on the yearly bill of mortality, that near half of the children that are born and bred in London die under two years of age. Some have attributed this amazing destruction to luxury and the abuse of spirituos liquors: these, no doubts, are powerful assistants; but the constant and unremitting poison is communicated by the foul air, which, as the town still grows larger, has made regular and steady advances in its fatal influence.’

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'London Smog' (also wintersmog, sulfur smog)

• In the 13th century coal (with high sulfur content) began to replace wood for domestic heating and industrial use in London

• Earliest massive human impact on the atmosphere

• The effects of smog on human health were evident, particularly when smog persisted for several days. Many people suffered respiratory problems and increased deaths were recorded, notably those relating to bronchial causes.

• The first smog-related deaths were recorded in London in 1873, when it killed 500 people. In 1880, the toll was 2000. London had one of its worst experiences with smog in December 1892. It lasted for three days and resulted in about 1000 deaths.

• London became quite notorious for its smog. By the end of the 19th century, many people visited London to see the fog.

• Despite gradual improvements in air quality during the 20th century, another major smog occurred in London in December 1952. The Great London Smog lasted for five days and resulted in about 4000 more deaths than usual.

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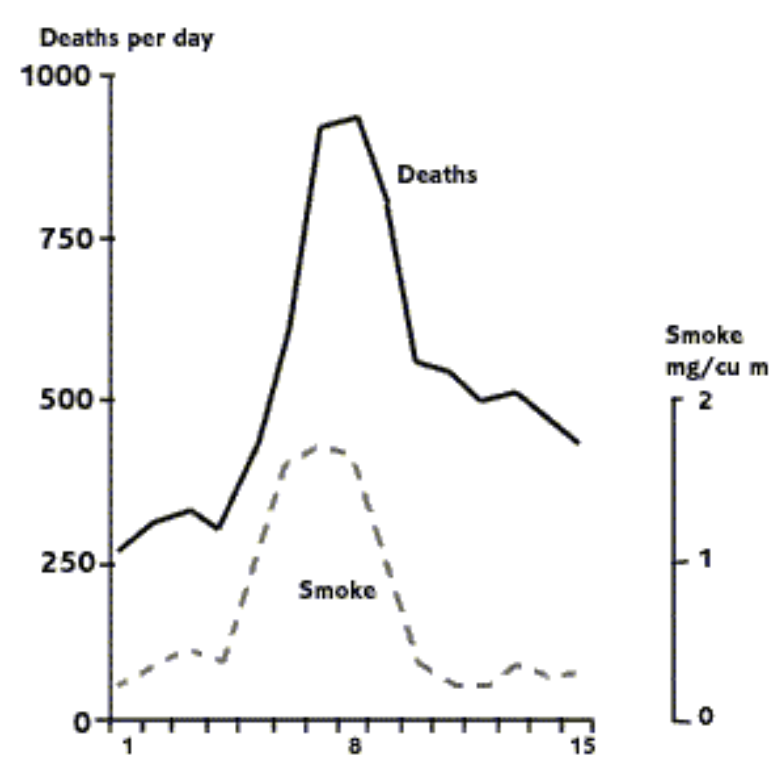

The London smog disaster of 1952. Death rate with concentrations of smoke

Hazardous driving conditions due to smog
(see also http://www.met-office.gov.uk/education/historic/smog.html)

Environmental damage due to sulfur emissions

- earliest massive human impact on the atmosphere
- Anthropogenic sulfur emissions were in particular responsible for the first non-local pollution
- At the end of the 1960s in Scandinavia a massive fish-dying occurred in inland waters. It was found out that the reason was an acidification of the soil. Finally it turned out that it was caused by strong industrial SO_2 emissions from Great Britain.
- The Waldsterben (dead of trees) was also mainly caused by SO_2 emissions
- Since the mid of the 1980s the SO_2 -emissions were strongly reduced in western countries due to effective filter techniques.

$SO_2 + OH \xrightarrow{M} HSO_3$ (1)

$HSO_3 + O_2 \rightarrow SO_3 + HO_2$ (2)

$SO_3 + H_2O_2 \xrightarrow{M} H_2SO_4$ (3)

In the dry stratosphere, particularly in the lower stratosphere where the concentration of OH is relatively small, the lifetime of SO_2 is longer than in the troposphere being of the order of several weeks.

Typical conditions for winter smog:

-primary pollution:	SO_2, soot particles
-secondary pollution :	H_2SO_4, Aerosols
-Temperature:	≤2°C
-Relative humidity:	high, typically foggy
-kind of inversion:	ground inversion
-time of maximum pollution:	early morning

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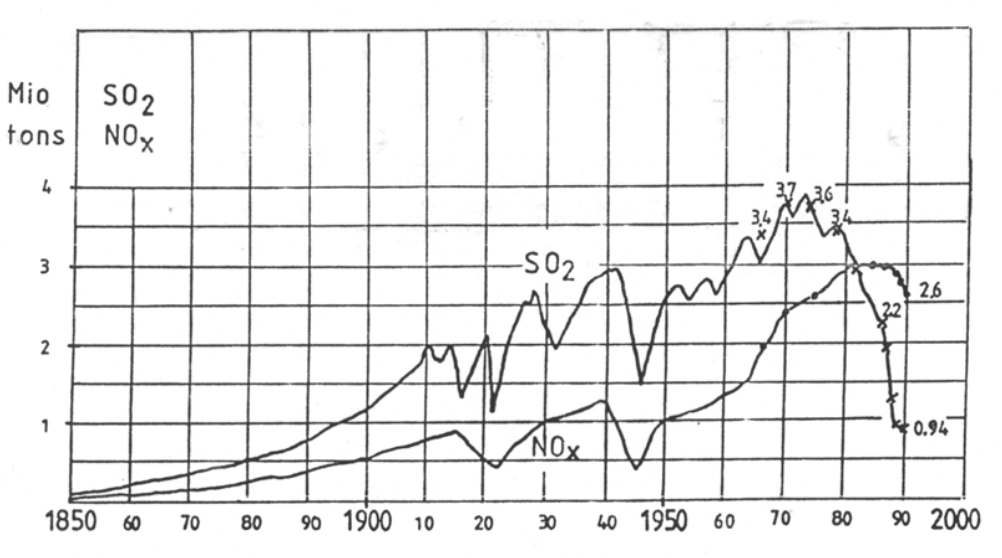
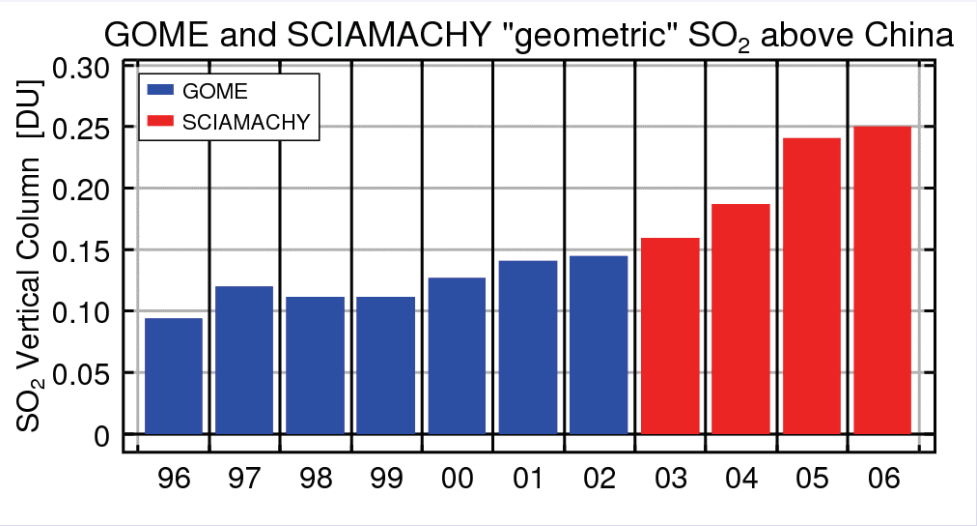
Long-term emission of SO_2 and NOx for the territory of the Federal Republic of Germany (without the former GDR)
Solid lines from 1850 - 1982: (ref.1)
Crosses and points: emission figures according to ref.2



Long-term time series of GOME (blue) and SCIAMACHY (red) SO_2-columns above the industrialised part of China (20°N, 100°E) – (40°N, 125°E). © Andreas Richter, Uni-Bremen, Germany



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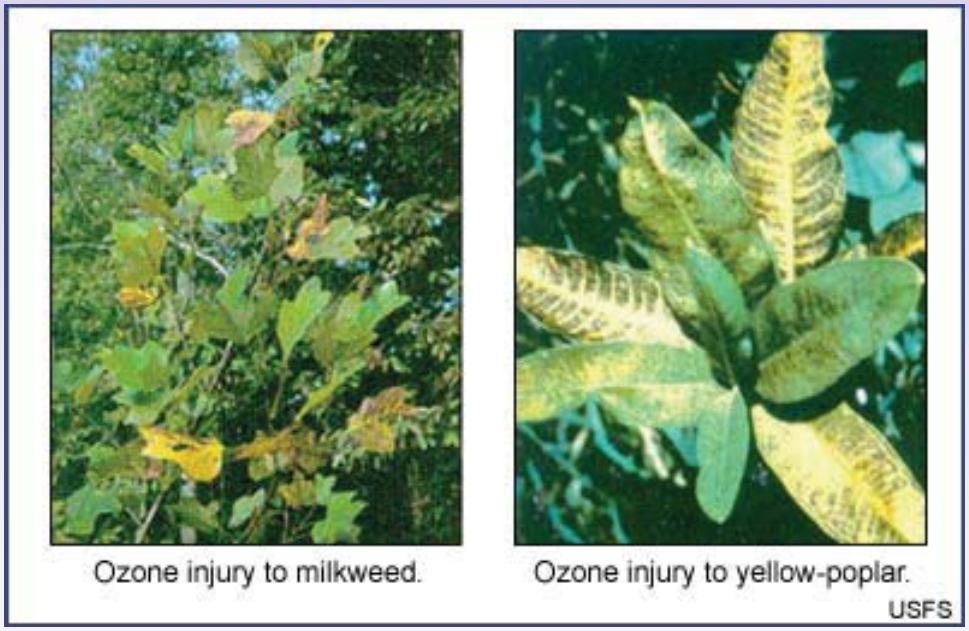
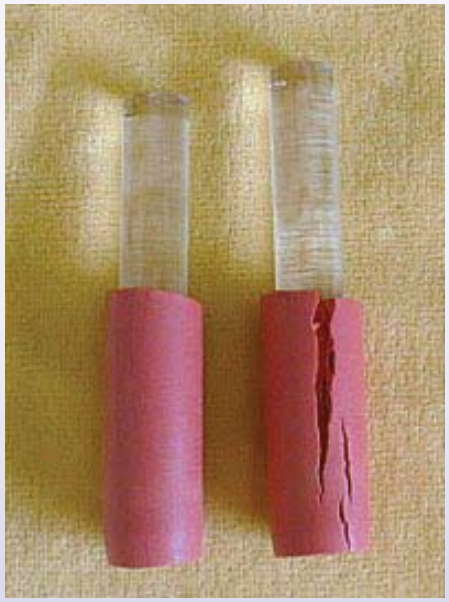
Los Angeles Smog

(Sommersmog, Ozonsmog)



- **Ozone affects health**
- **Ozone damages plants**
- **Ozone destroys material**
- **Ozone determines the oxidation capacity of the atmosphere**



Impact of Ozone on Rubber

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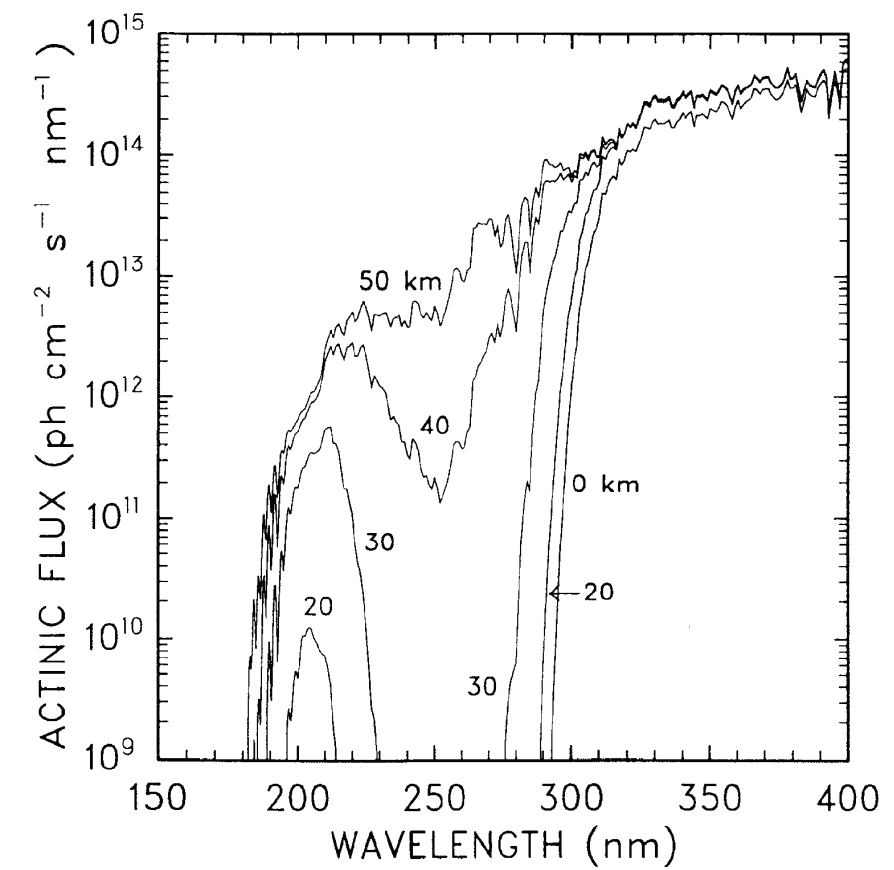
Ozone production in the troposphere, summer smog

-In the troposphere, not enough UV radiation is available for ozone formation through O_2 photolysis

-where does tropospheric O_3 originate from?

Penetration depth of UV radiation



DeMore et al., 1997

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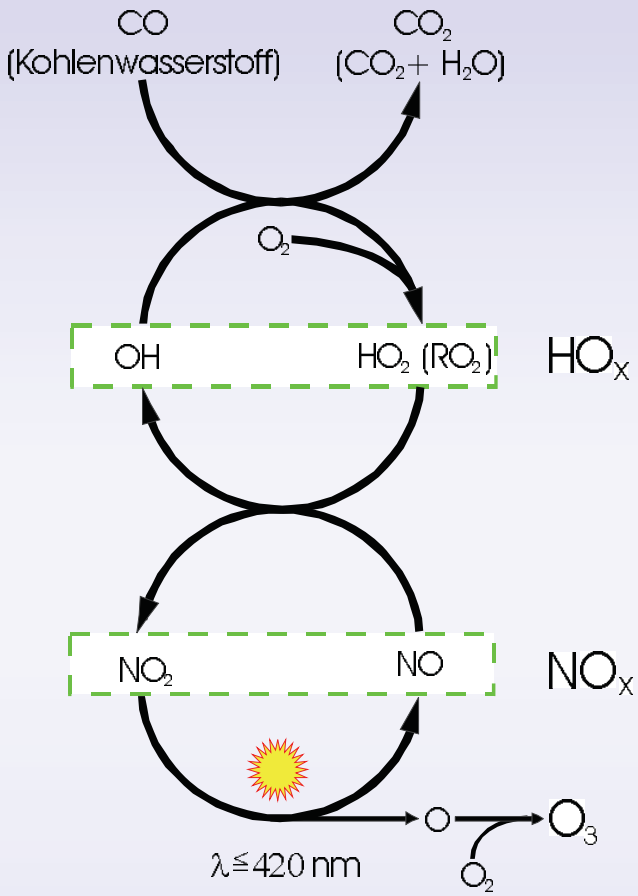
Ozonsmog
(Los Angeles, 1940s, Haagen-Smit, 1952)



Radical chemistry, Ozone production in the troposphere:

$$HO_2 + NO \rightarrow HO + NO_2$$

$$NO_2 + h\nu \rightarrow NO + O \quad (\lambda \leq 410nm)$$

$$O + O_2 + M \rightarrow O_3 + M$$

The occurance of ozone smog depends on the concentrations of volatile organic compounds (VOC) an nitrogen dioxid (NO_2)

Without pollution, ozone is produced by oxidation of CH_4 and CO => tropospheric background ozone

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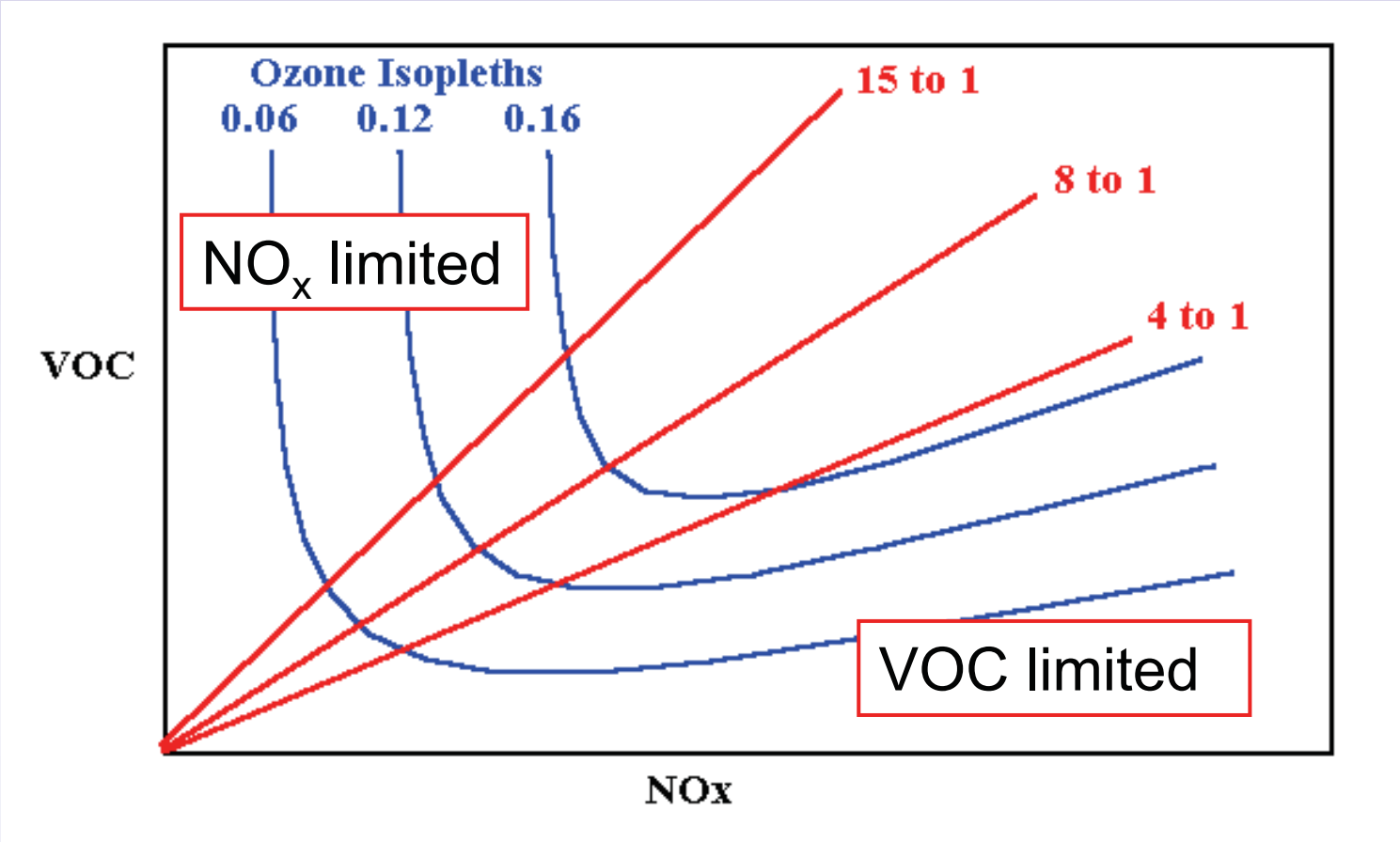

A VOC to NOx ratio of 8 to 1 is often cited as an approximate decision point for determining the relative benefits of NOx vs. VOC controls. At low VOC to NOx ratios (< about 4 to 1), an area is considered to be VOC-limited; VOC reductions will be most effective in reducing ozone, and NOx controls may lead to ozone increases. At high VOC to NOx ratios (>about 15 to 1), an area is considered NOx limited, and VOC controls may be ineffective. When VOC to NOx ratios are at intermediate levels (4 to 15), a combination of VOC and NOx reductions may be warranted.

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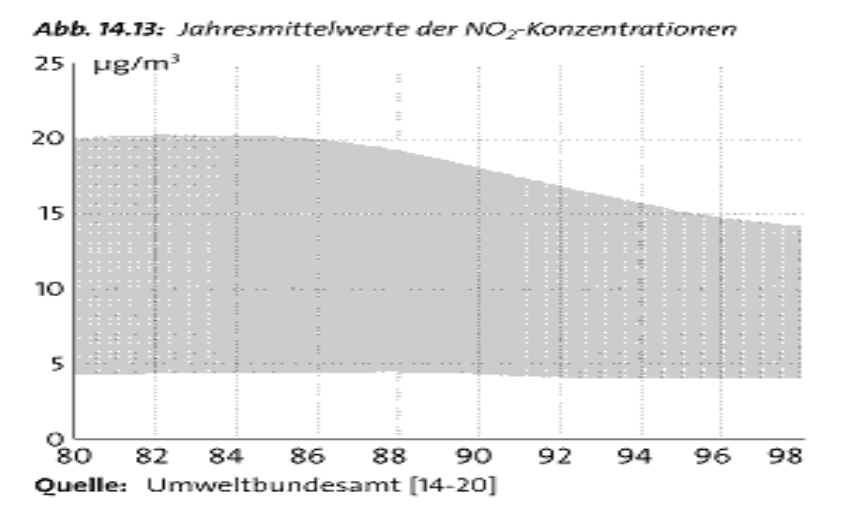
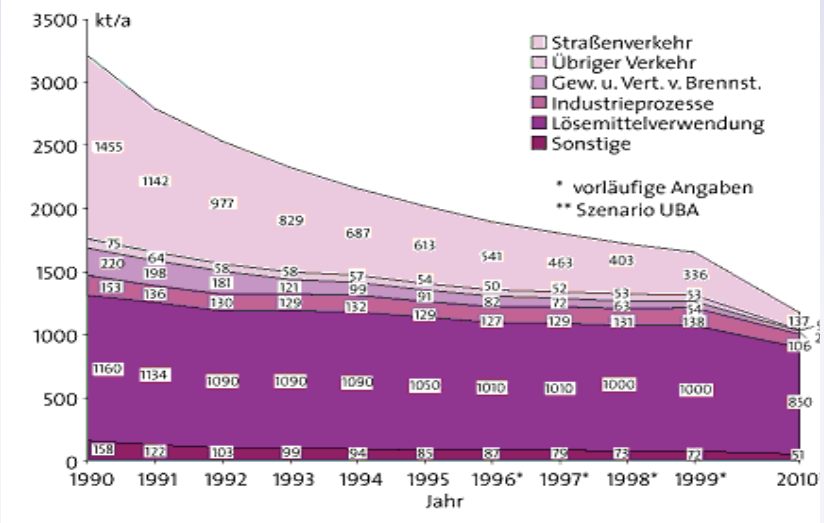
Importance of ‚local' effects?

Average concentrations of NO_2 in Germany



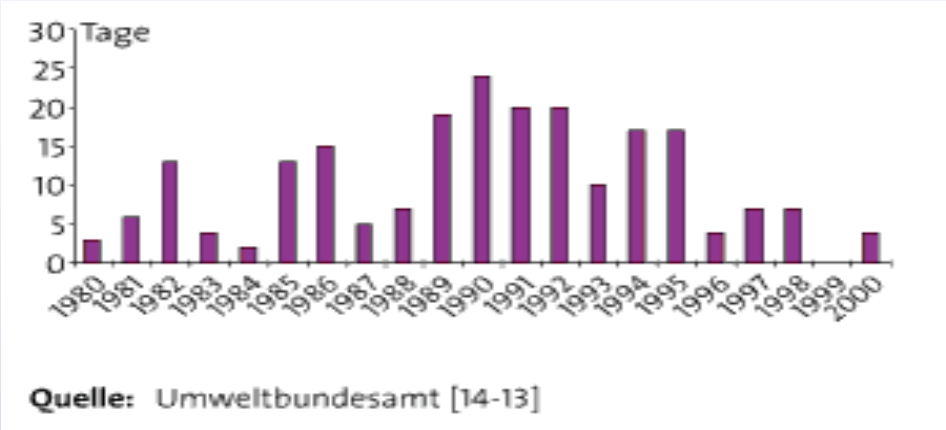
Emissions of NMHC in Germany



Days with

$[O_3] > 240$ ug/m³

in Germany



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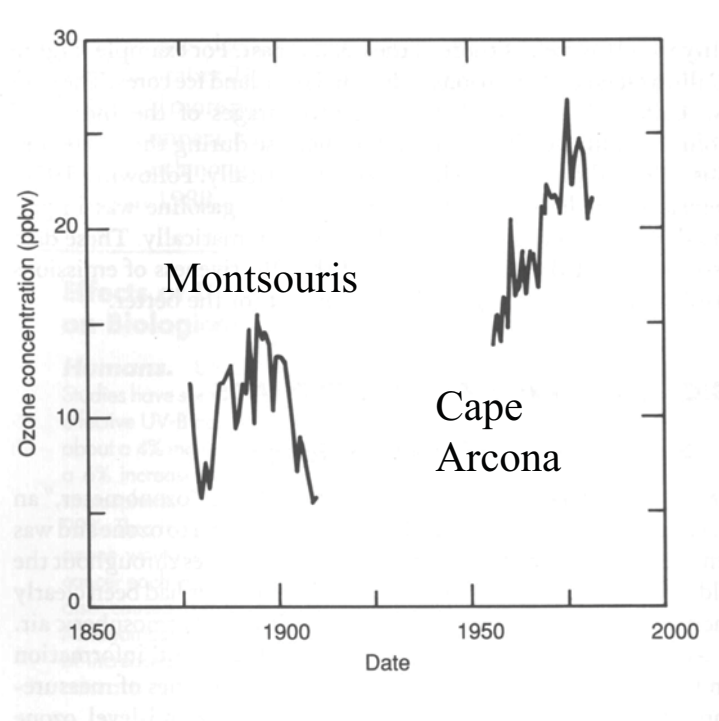
What about tropospheric background ozone concentrations?



Increase of background ozone in Europe

(Volz and Kley, 1988)

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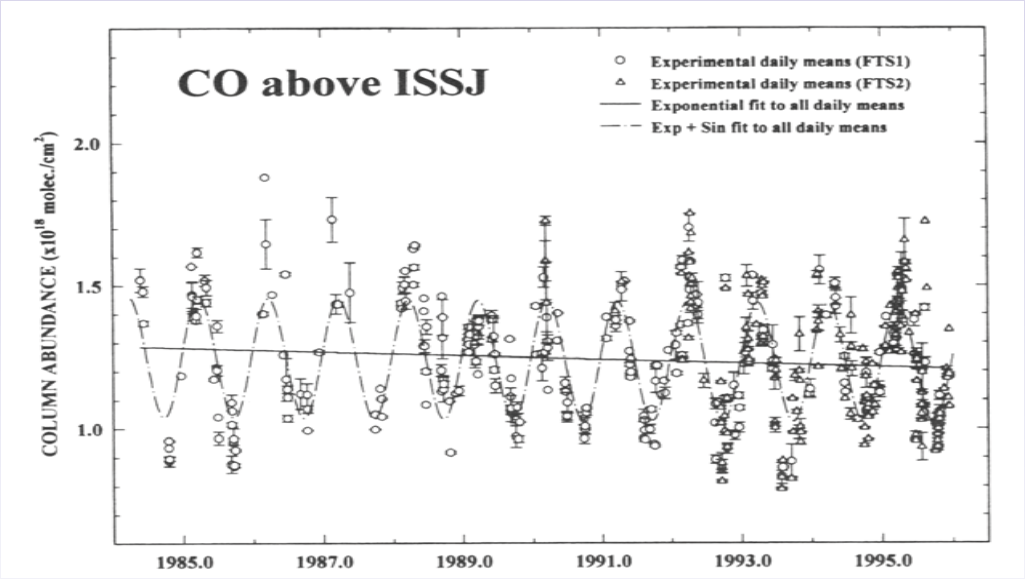
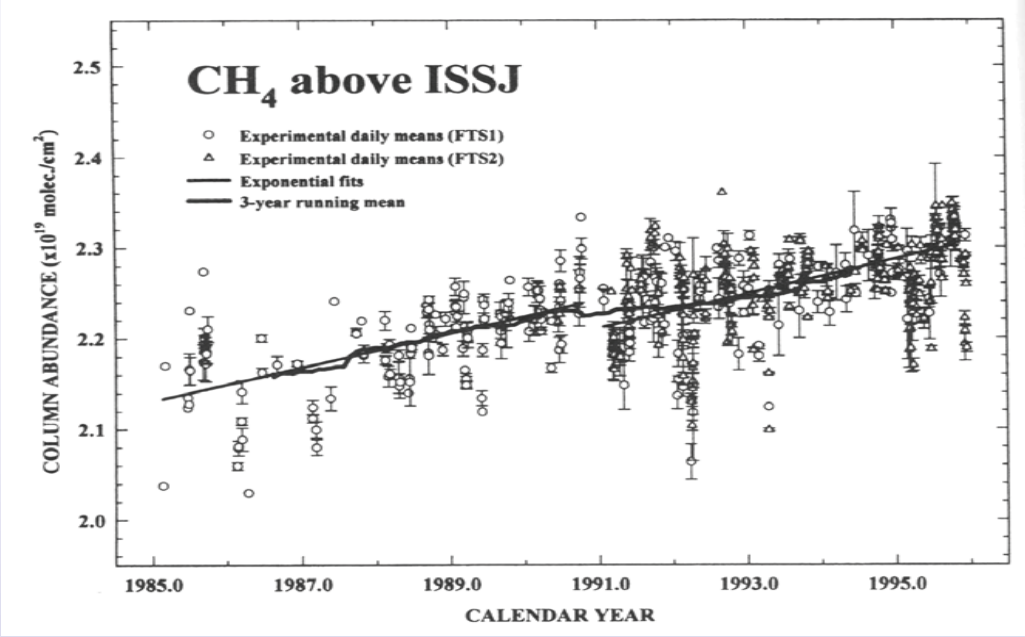
CO and CH_4 column above the Jungfraujoch station

1985 - 1996

Mahieu et al., 1997





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Discovery of the 'Cleansing agent' of the atmosphere (OH-radical, Levi, 1971)

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \qquad (\lambda \leq 310nm)$$

$$O(^1D) + H_2O \rightarrow 2OH\bullet$$

OH• ist a (free) radical. Radicals are molecule fragments with unpaired electrons. Thus their bound conditions are not required and they are very reactive. The production of radicals depends on the fission of molecules and requires high energy. Typically, this energy is supplied by photons. Radicals are the 'active players' of photochemistry.

OH reacts with almost all atmospheric trace gases which is the prerequisite for their destruction. This ability to destroy atmospheric trace gases is called oxidation capacity.

OH is very reactive (within seconds). Although its concentrations are very low, it is the most important atmospheric reactant.

OH exists only during day. Will concentrations of atmospheric pollutants increase during night?

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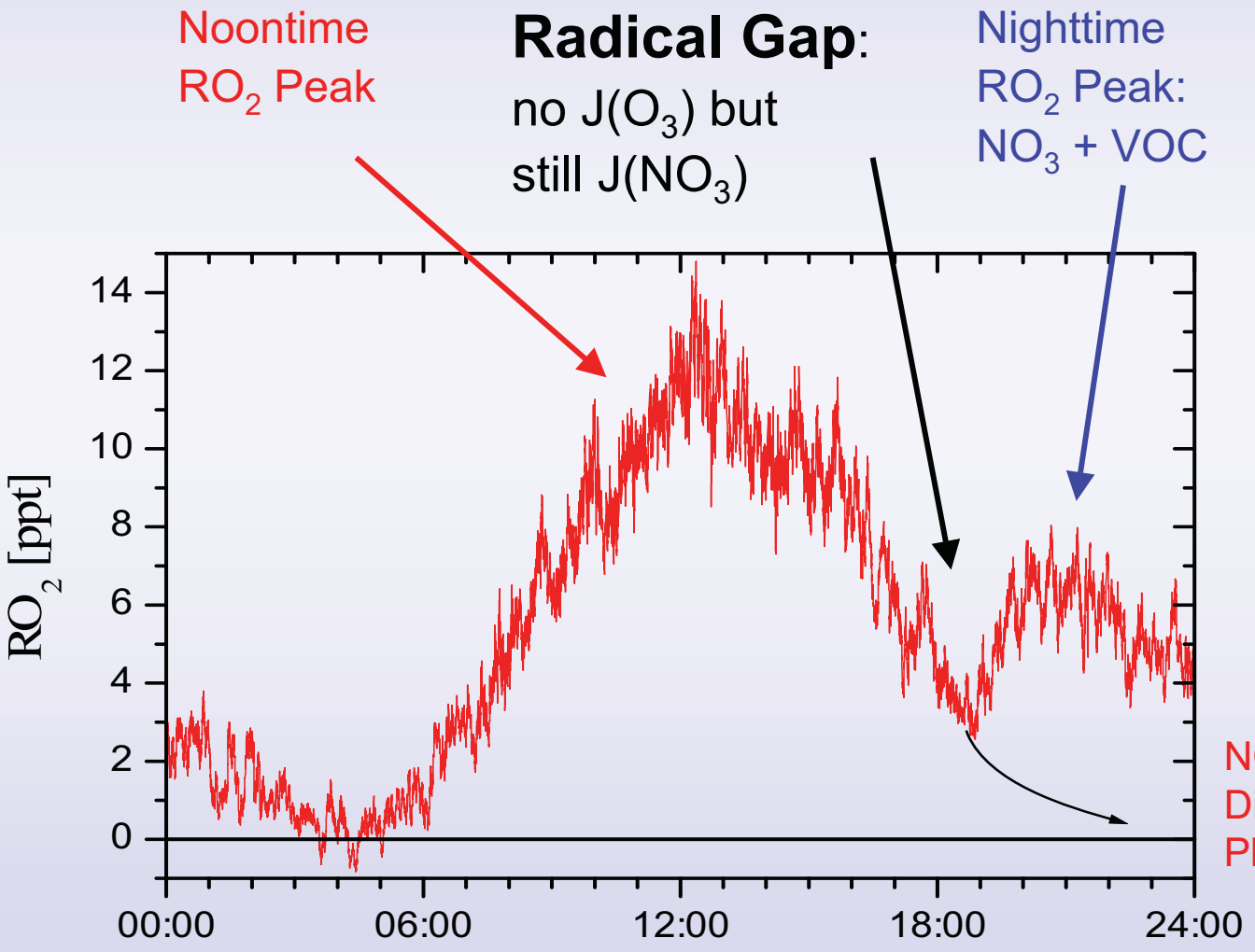
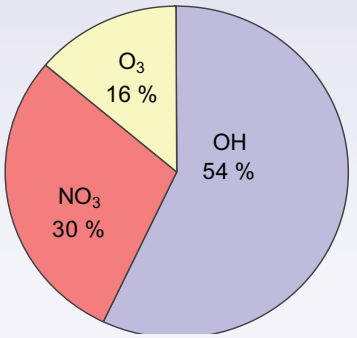
Average RO_X diurnal profile (Chemical Amplifier) during BERLIOZ

(July 14 to 15, 17 to 18, 24 to 26, Aug. 3, total of 8 days in 1998)





Contribution of NO_3 to the Atmospheric Oxidation Capacity in Pabstthum (BERLIOZ 1998)

NO_3 Data from Dieter Perner, in Platt et al. 2002

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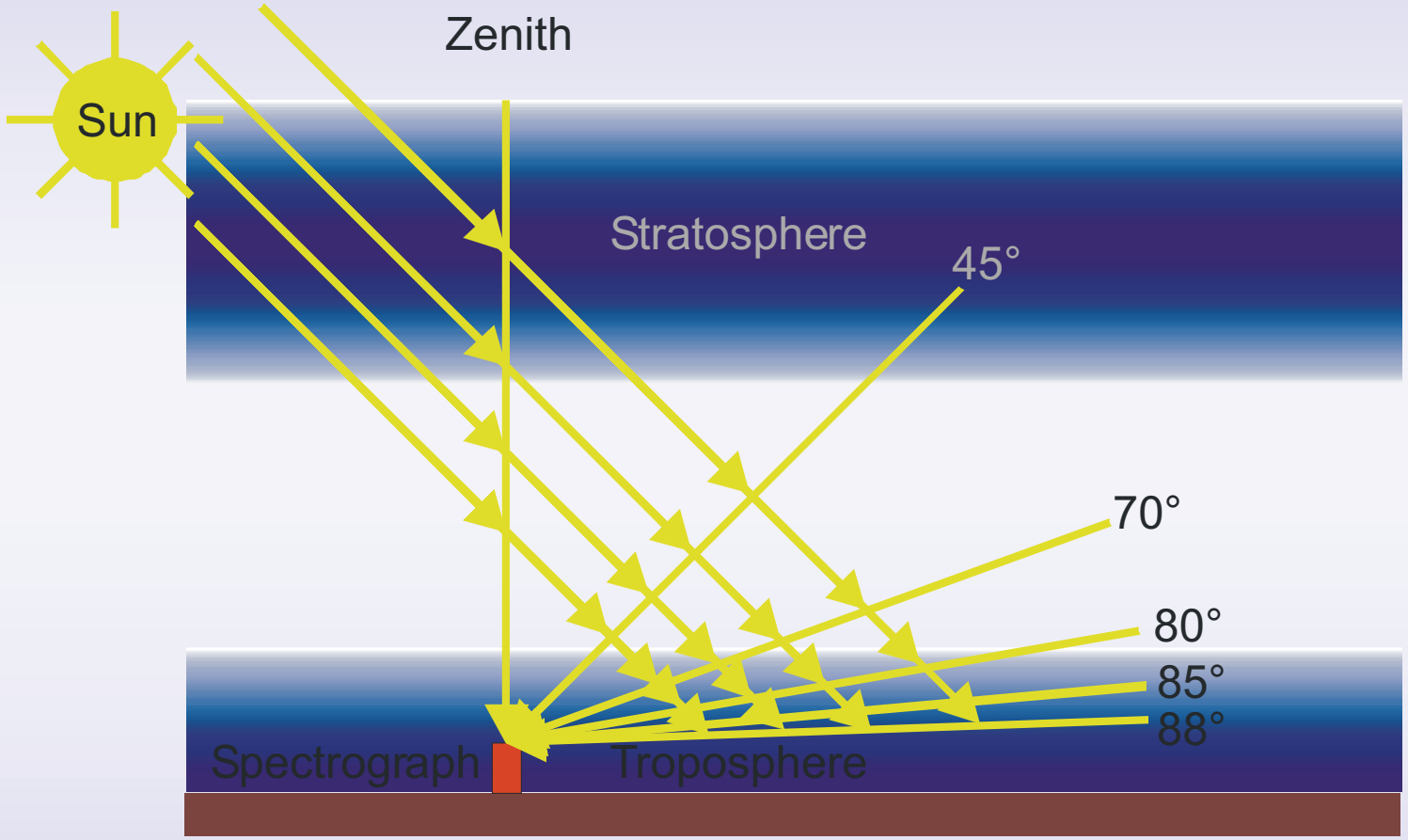
Multi-Axis- (MAX-) DOAS



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Multi-Axis- (MAX-) DOAS

Trieste, 2.2.2009 (lunch break)

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Atmos. Chem. Phys., 8, 7595–7601, 2008
www.atmos-chem-phys.net/8/7595/2008/

Atmospheric Chemistry and Physics

High spatial resolution measurements of NO_2 applying Topographic Target Light scattering-Differential Optical Absorption Spectroscopy (ToTaL-DOAS)

E. Frins[1], U. Platt[2], and T. Wagner[3]

[1]Instituto de Física, Facultad de Ingeniería, J. Herrera y Reissig 565, 11300 Montevideo, Uruguay
[2]Institut für Umweltphysik, University of Heidelberg, Im Neuenheimer Feld 229, 69120 Heidelberg, Germany
[3]Max-Planck-Institut für Chemie, Becherweg 27, 55128 Mainz, Germany

Montevideo,
November 6, 2007

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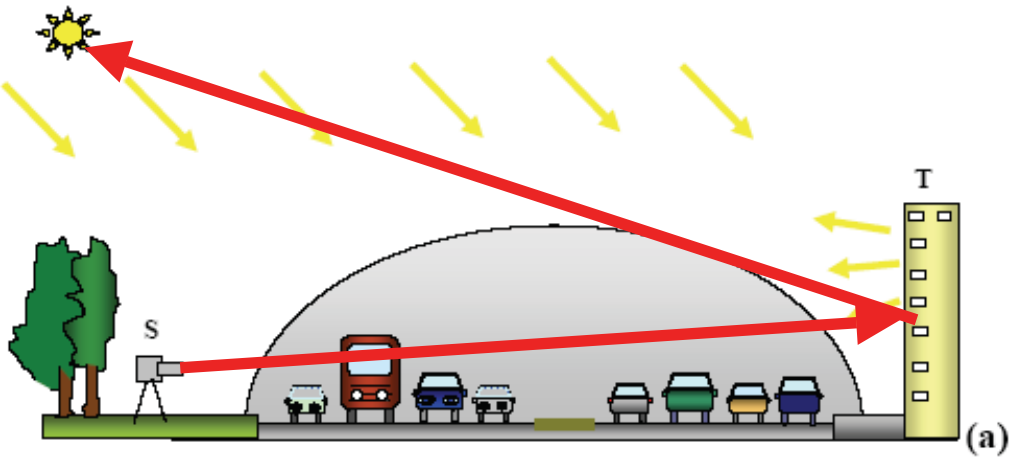
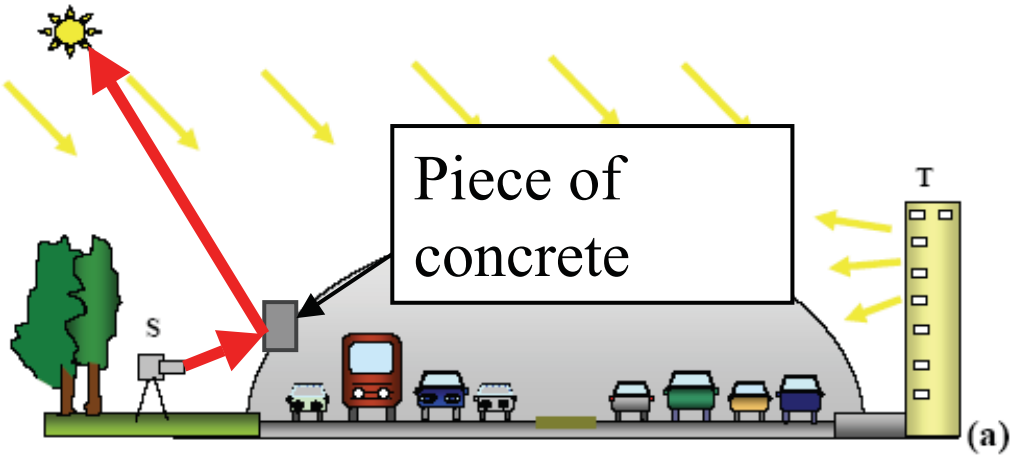
Image © 2007 DigitalGlobe
Google

(a)

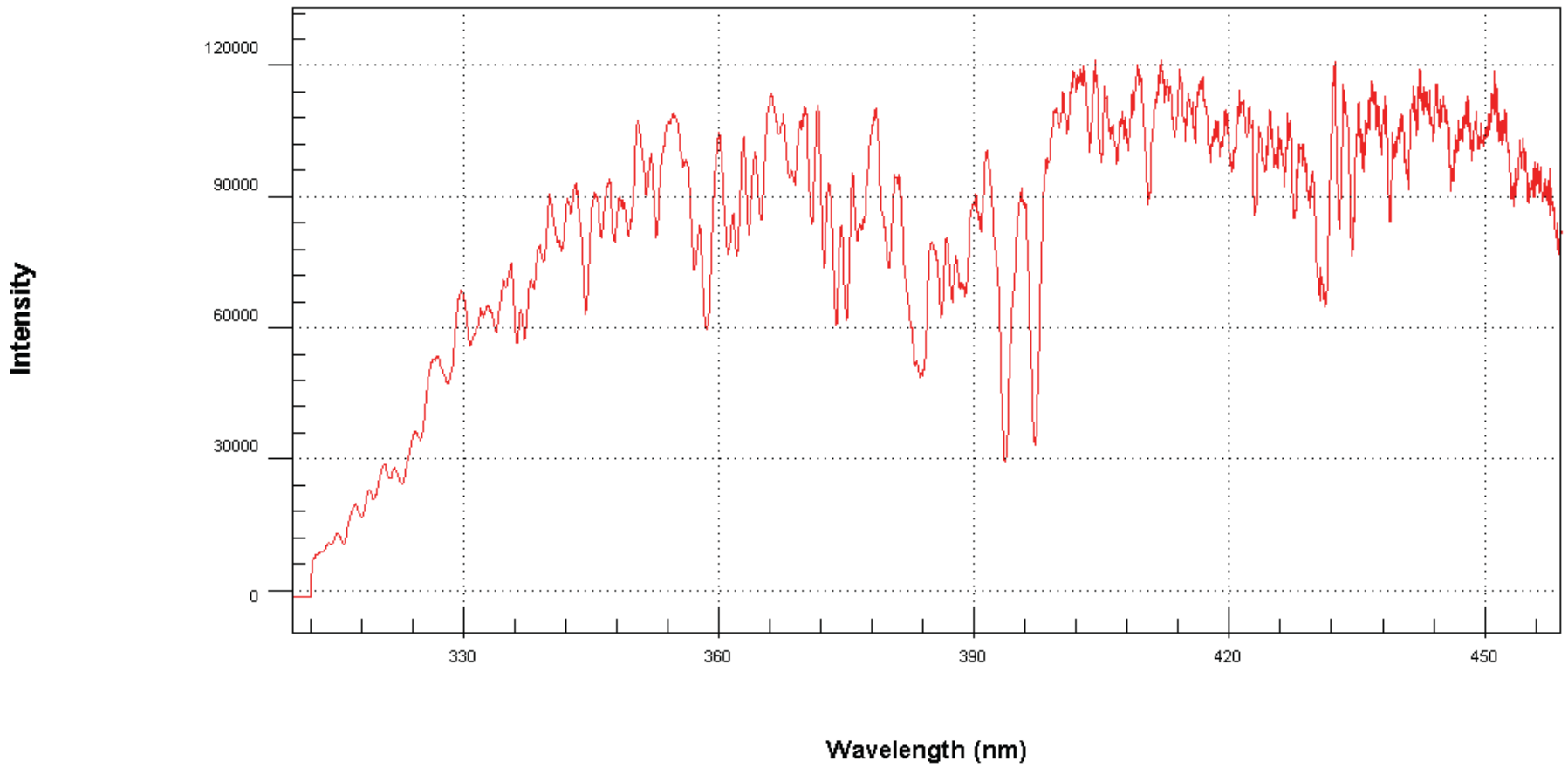

T
S

(a)


Viewing direction
to near building
Second and third building


Piece of
concrete
T
S

(a)



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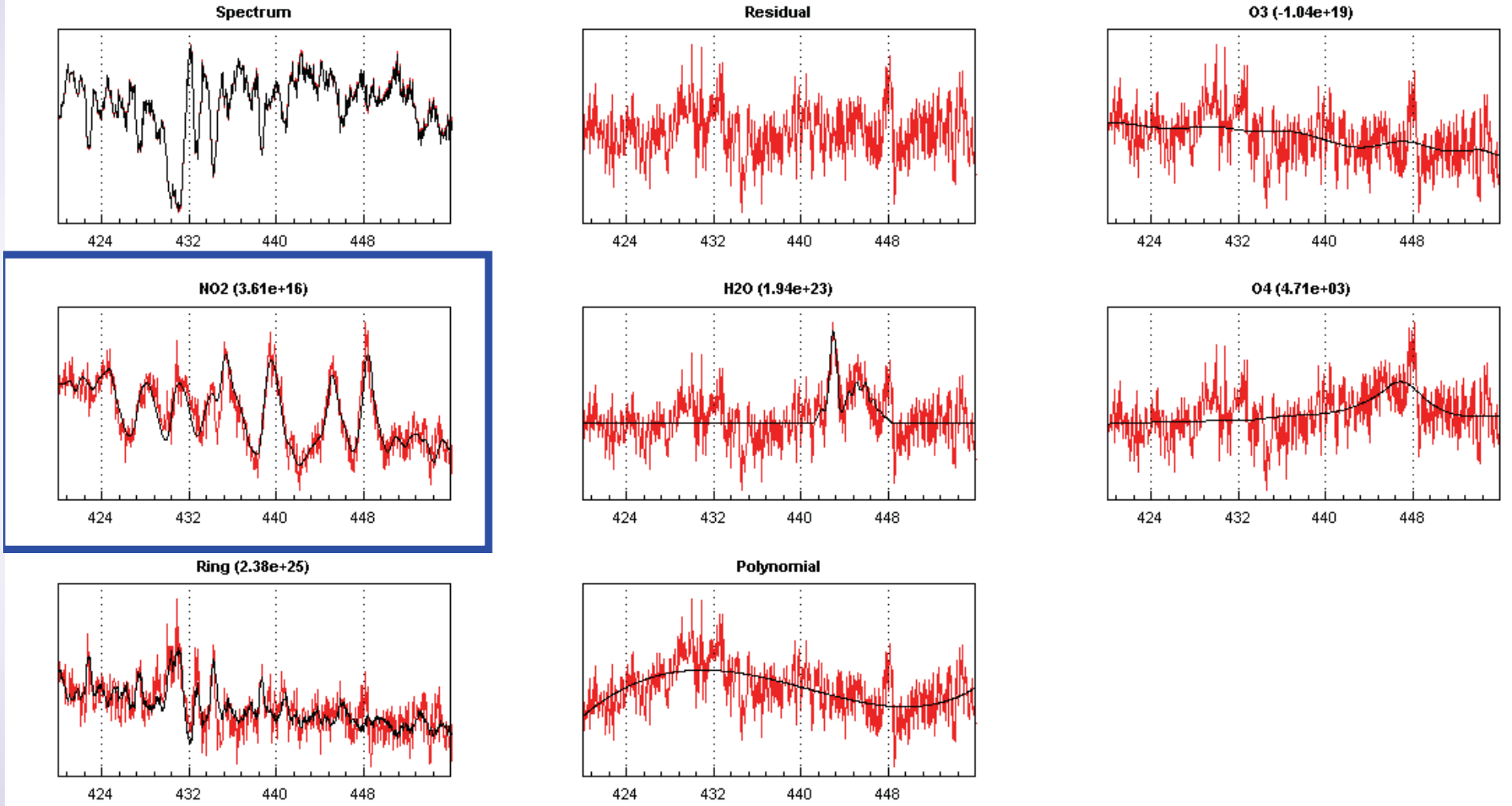
MAXDOAS measurements, Trieste, 2.2.2009 (lunch break)

3° elevation angle



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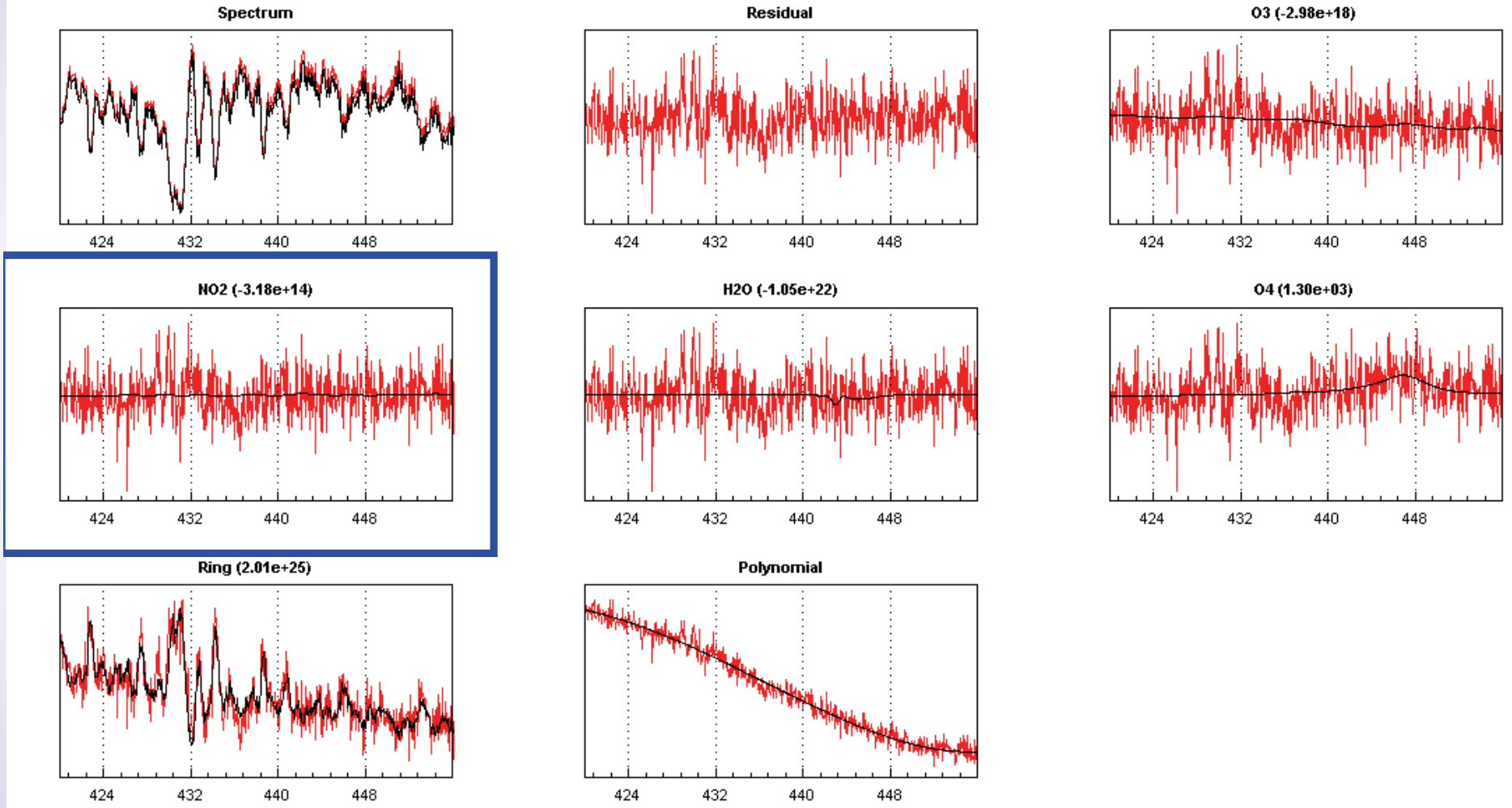
MAXDOAS measurements, Trieste, 2.2.2009 (lunch break)

50° elevation angle



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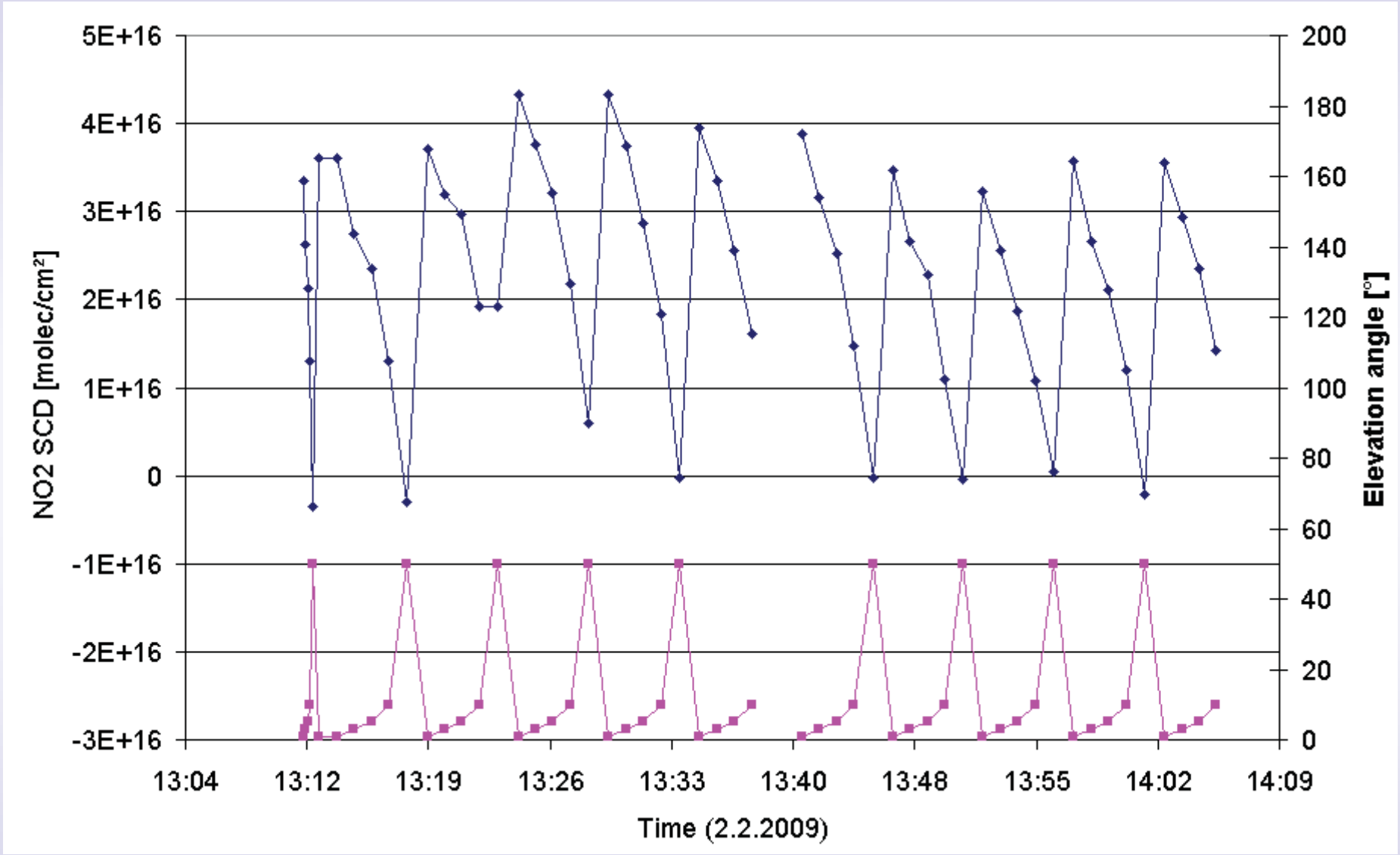
MAXDOAS measurements, Trieste, 2.2.2009 (lunch break)



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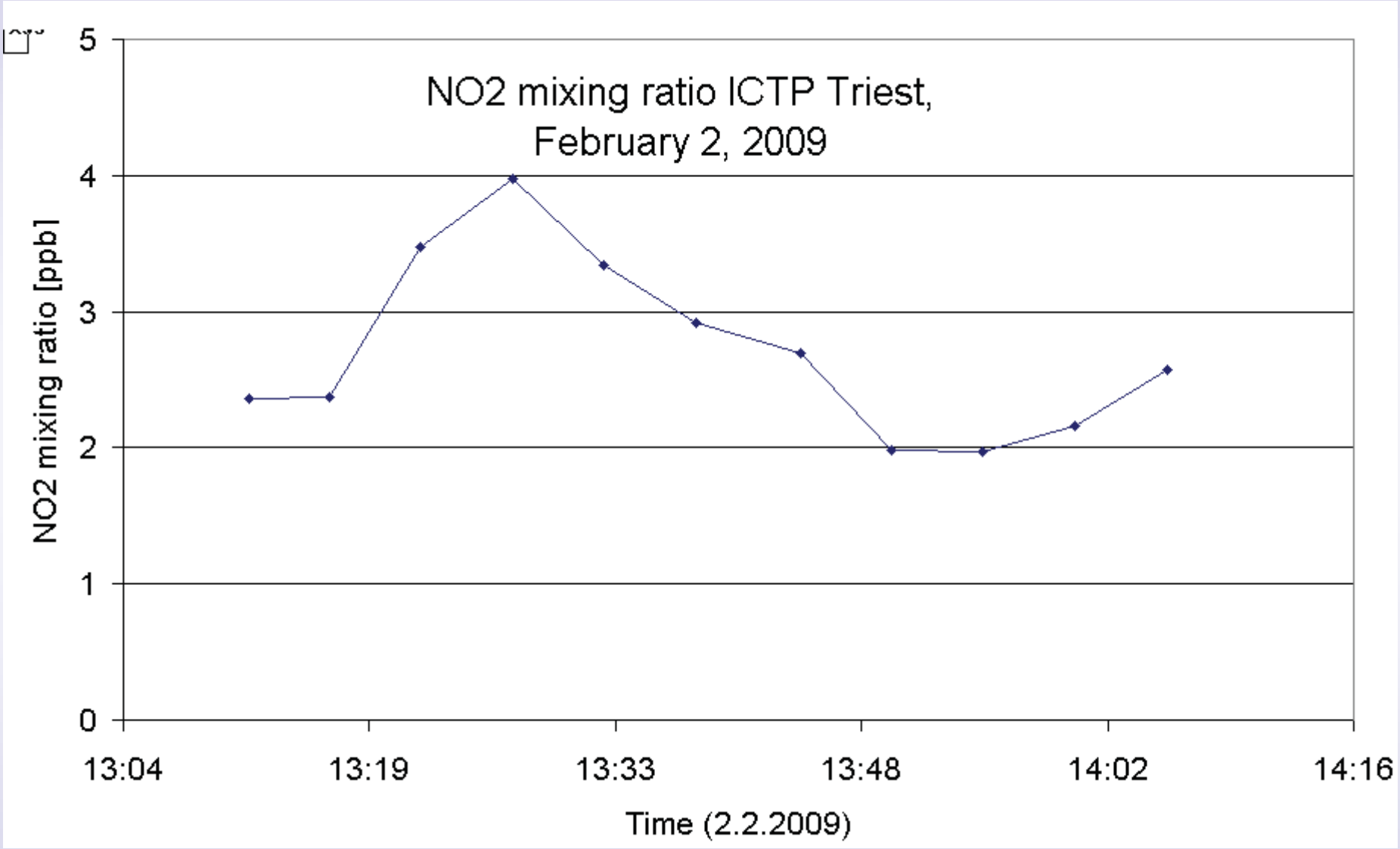

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