

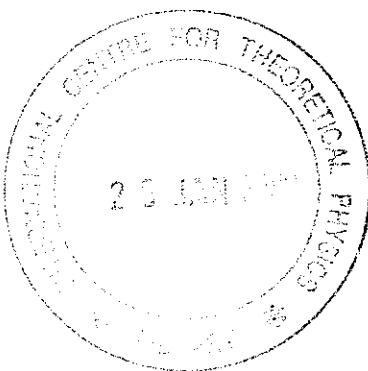
the  
**abdus salam**  
international centre for theoretical physics

**School on "Exploring the Atmosphere by  
Remote Sensing Techniques"  
18 October - 5 November 1999**

**1151-16**

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**"Ground-Based Measurements of Stratospheric Trace Gases in  
the Perspective of Climate & Environment"**



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Institut d'Aéronomie Spatiale de Belgique  
Brussels  
Belgium**



# **Ground-based measurements of stratospheric trace gases in the perspective of climate and environment**

- Ground-based remote sensing techniques, and O<sub>3</sub> sondes
  - ! Complementarity with space- and airborne techniques !
  - ! Complementarity with in-situ sampling methods !
- observed atmospheric changes: variabilities, trends
- assessments, related policies and perspectives

AA

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Alto Manica BIP VVW

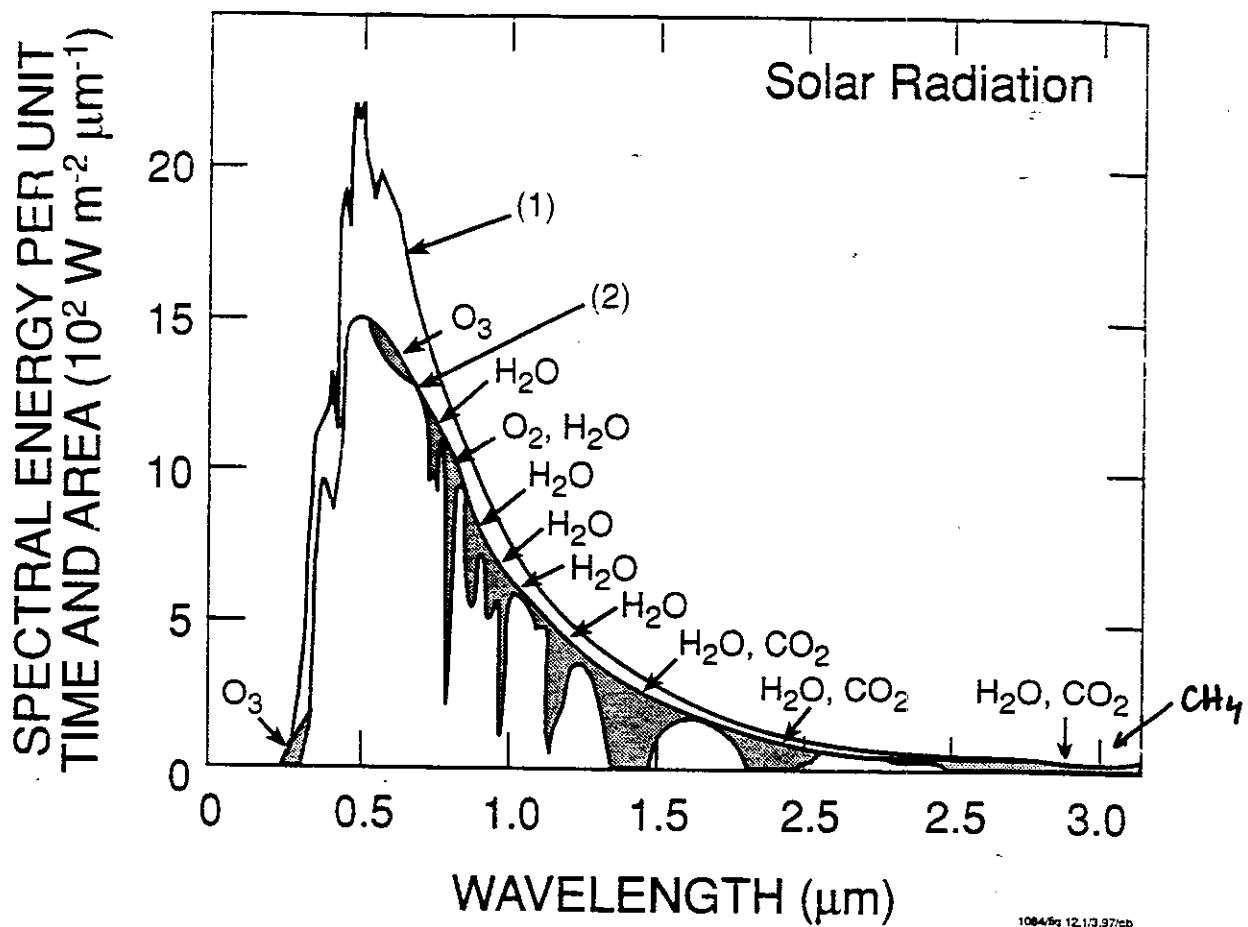
## Ground-based techniques (1/4)

- O<sub>3</sub> sonde
  - based on a chemical redox reaction in an electrochemical cell
  - balloon-borne, in-situ measurement of the O<sub>3</sub> vertical profile, from ground up to < 35 km

## Ground-based techniques (2/4)

- ☒ GB techniques are mostly based on extinction of light in the atmosphere
  - extinction  $\Rightarrow$  absorption and scattering by atmospheric constituents (gaseous, particles)
    - $\leftarrow$  laboratory data!
  - in various regions of the spectrum:
    - UV/Vis
      - DOAS
      - LIDAR (DIAL)
    - infrared
      - FTIR or Fourier-Transform Infrared Spectroscopy
    - microwave
  - remote sensing

Α



**Figure 15.1.** Spectrum of solar radiation (1) outside the Earth's atmosphere and (2) at sea level for clear sky conditions (from Gast, 1961). The shaded area represents the energy absorbed by various gases in a clear atmosphere.

# Ground-based techniques (3/4)

## Atmospheric data products

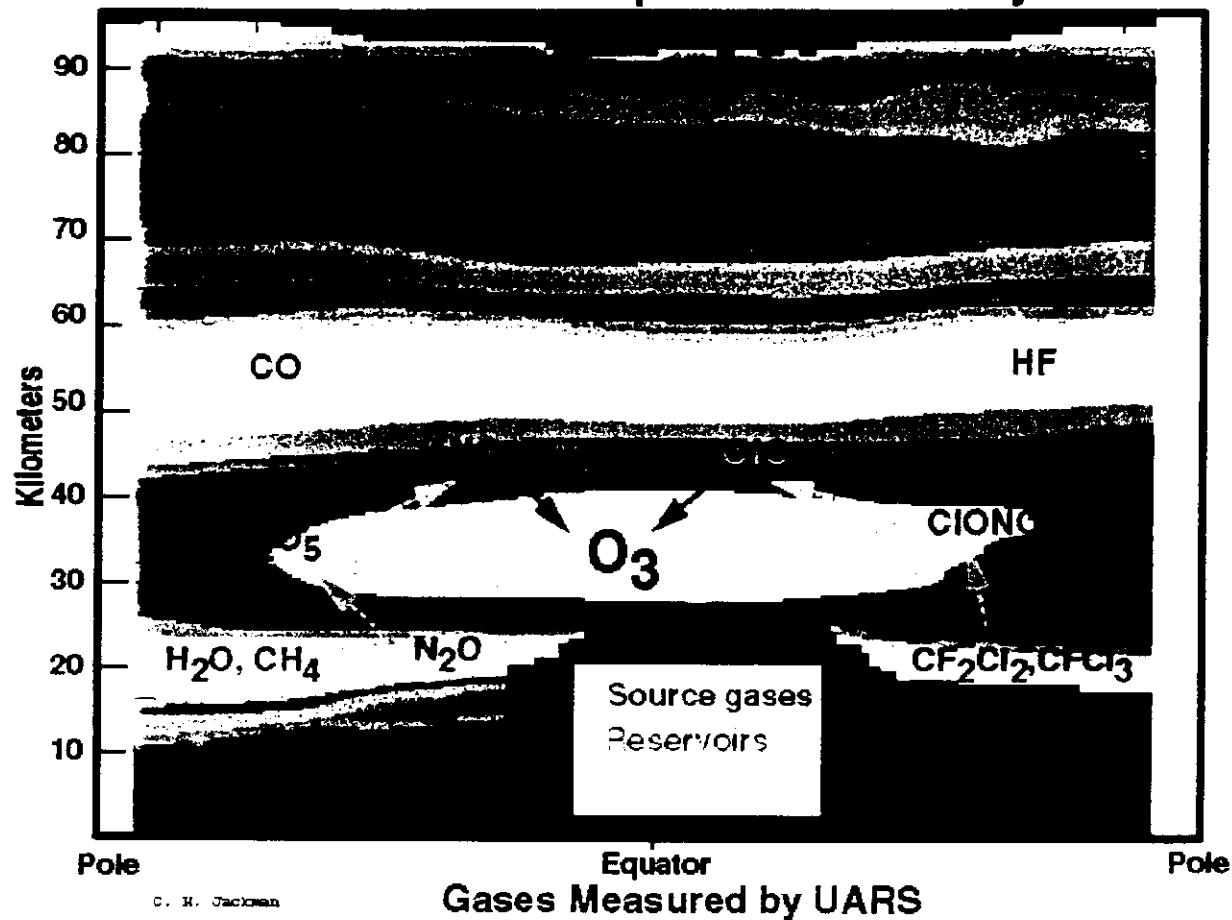
	GB	UV/Vis	1.11)ΔIR	1.71IR	Mwave
O <sub>3</sub>	C	P	C	C	stratP/C
NO <sub>2</sub>	C		C		
BrO	C				
OClO	C				
ClO			(C)		stratP/C
Cl <sub>y</sub>			C		
NO <sub>y</sub>			C		
H <sub>2</sub> O	(C)	P	C	C	stratP/C
tracers			C		
sources			C		
CO, CO <sub>2</sub>			C		

(Strat)P = (stratospheric) profile; C = column;  
 tracers = N<sub>2</sub>O, CH<sub>4</sub>, HF, O<sub>3</sub>, ... ; sources = N<sub>2</sub>O, CFC, HCFC, CO<sub>2</sub>, ...

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UV/VIS  
IR  
Mwave

## Middle Atmosphere Chemistry



# Ground-based techniques (4/4)

Atmospheric data products: interest ?

- ☒ Stratospheric O<sub>3</sub> depletion  $\Leftrightarrow$  increase of UV at Earth surface

➢ O<sub>3</sub>, Cl<sub>y</sub>, Br<sub>y</sub>, halocarbons (CFC, HCFC,...) NO<sub>y</sub>,  
aerosol/PSC, ...

- ☒ Changes in tropospheric and PBL O<sub>3</sub>  $\Leftrightarrow$  biological health

➢ H<sub>2</sub>O ( $\rightarrow$  OH, HO<sub>2</sub>), CH<sub>4</sub>, CO, RC, NO<sub>x</sub>, ...

- ☒ Climate and surface warming

➢ Greenhouse Gases: H<sub>2</sub>O, CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, O<sub>3</sub>,  
halocarbons, aerosol, clouds, ...

! Other factors are playing a role: dynamics, solar cycle, ...  
! Effects are coupled



## **Observed atmospheric changes: variabilities, trends**

- diurnal, seasonal cycles and daily variabilities
  - inter-annual variabilities
  - long-term trends ( $> 1$  decade)
    - possibly as a function of altitude
- ⇒ interest in long-term, regular monitoring,  
e.g., from ground-based monitoring networks.

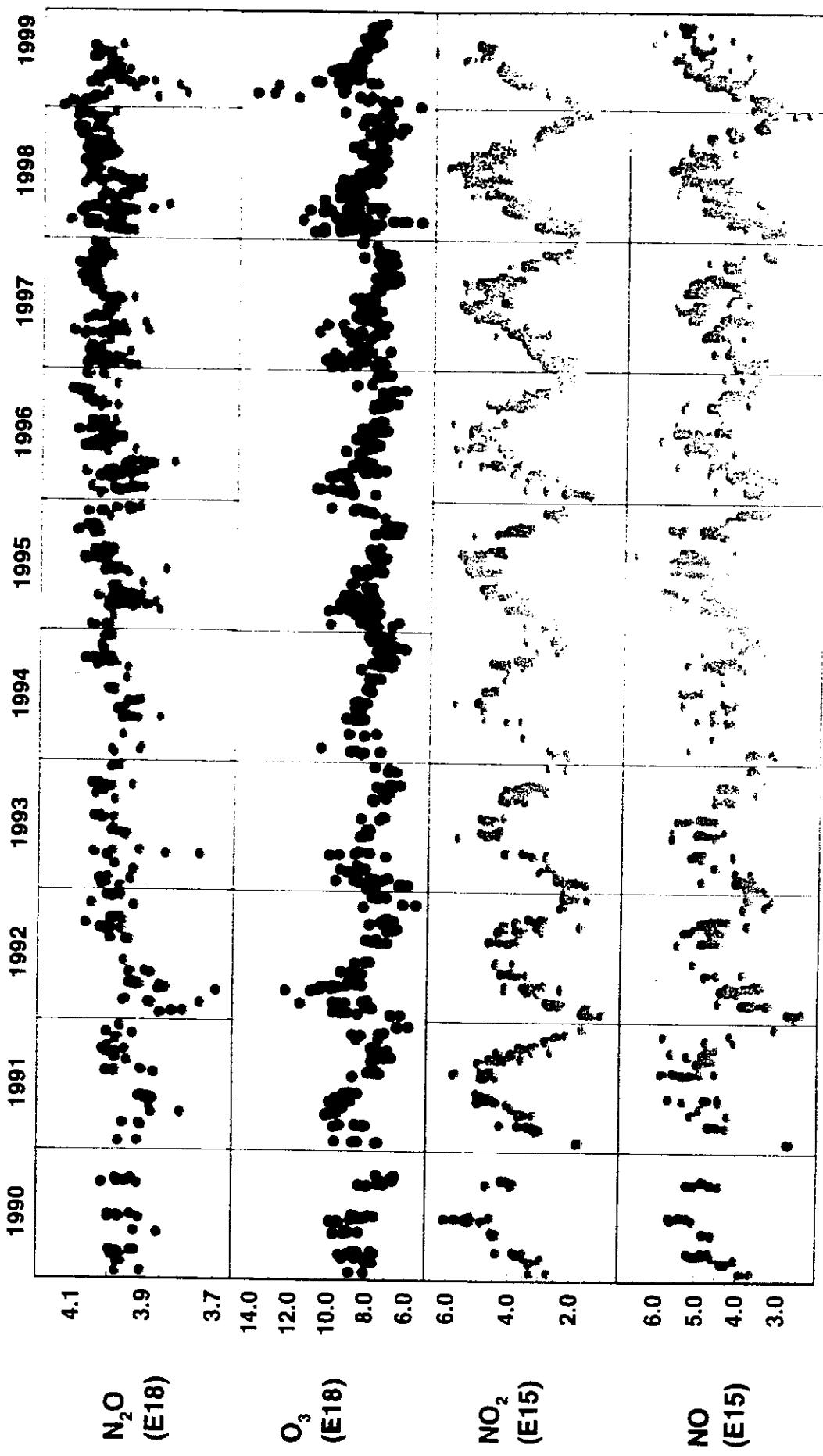
### *Example:*

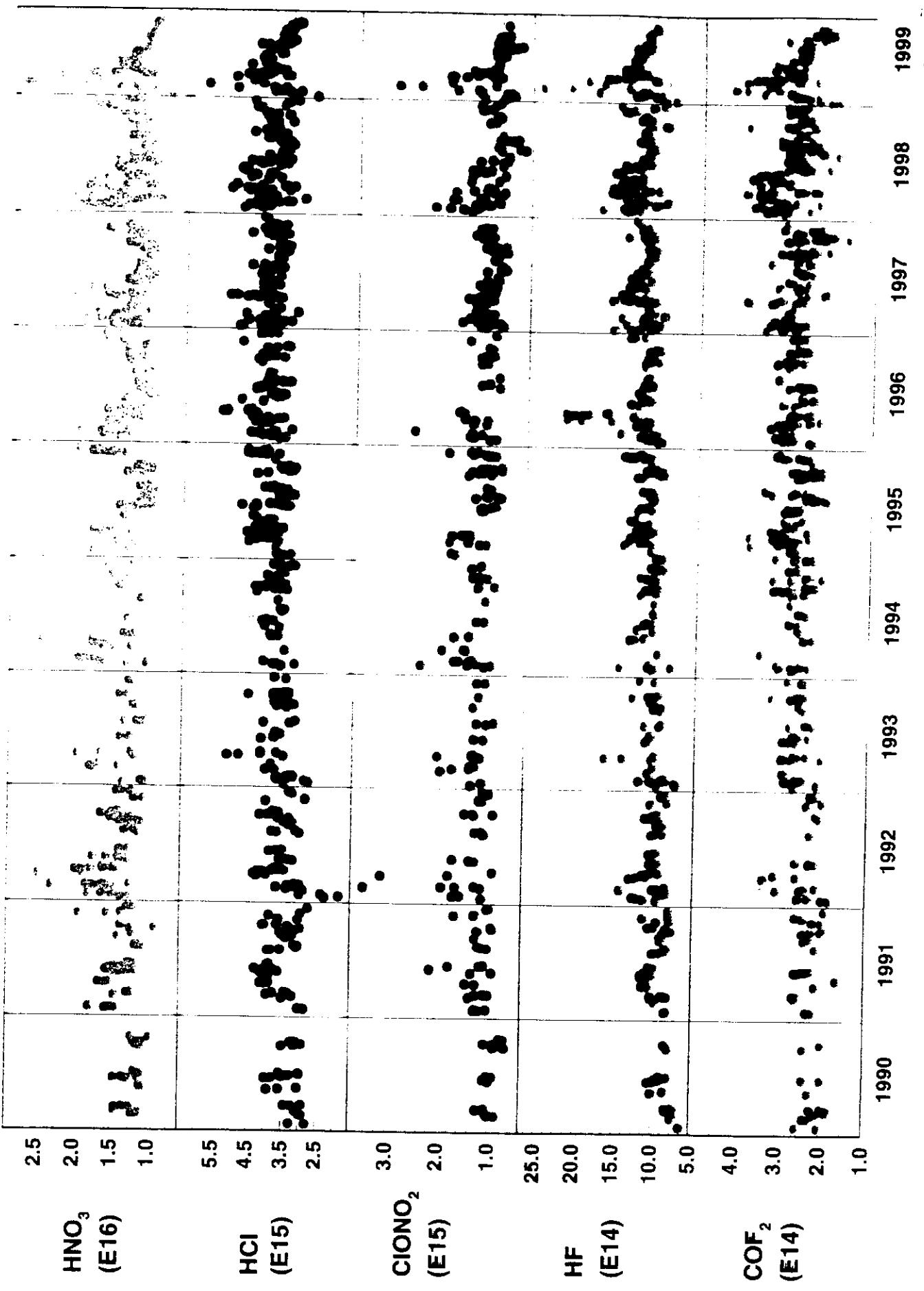
**Jungfraujoch Observatory in the Swiss Alps,**  
primary alpine NDSC station, at 3580 m asl,  $44.6^{\circ}\text{N}$ ,  $8.0^{\circ}\text{E}$ ,  
operating on a regular basis since 1985,  
some measurements already in the 50s





# Daily mean vertical column abundances of 8 stratospheric constituents above ISSJ





## Atmospheric variabilities:

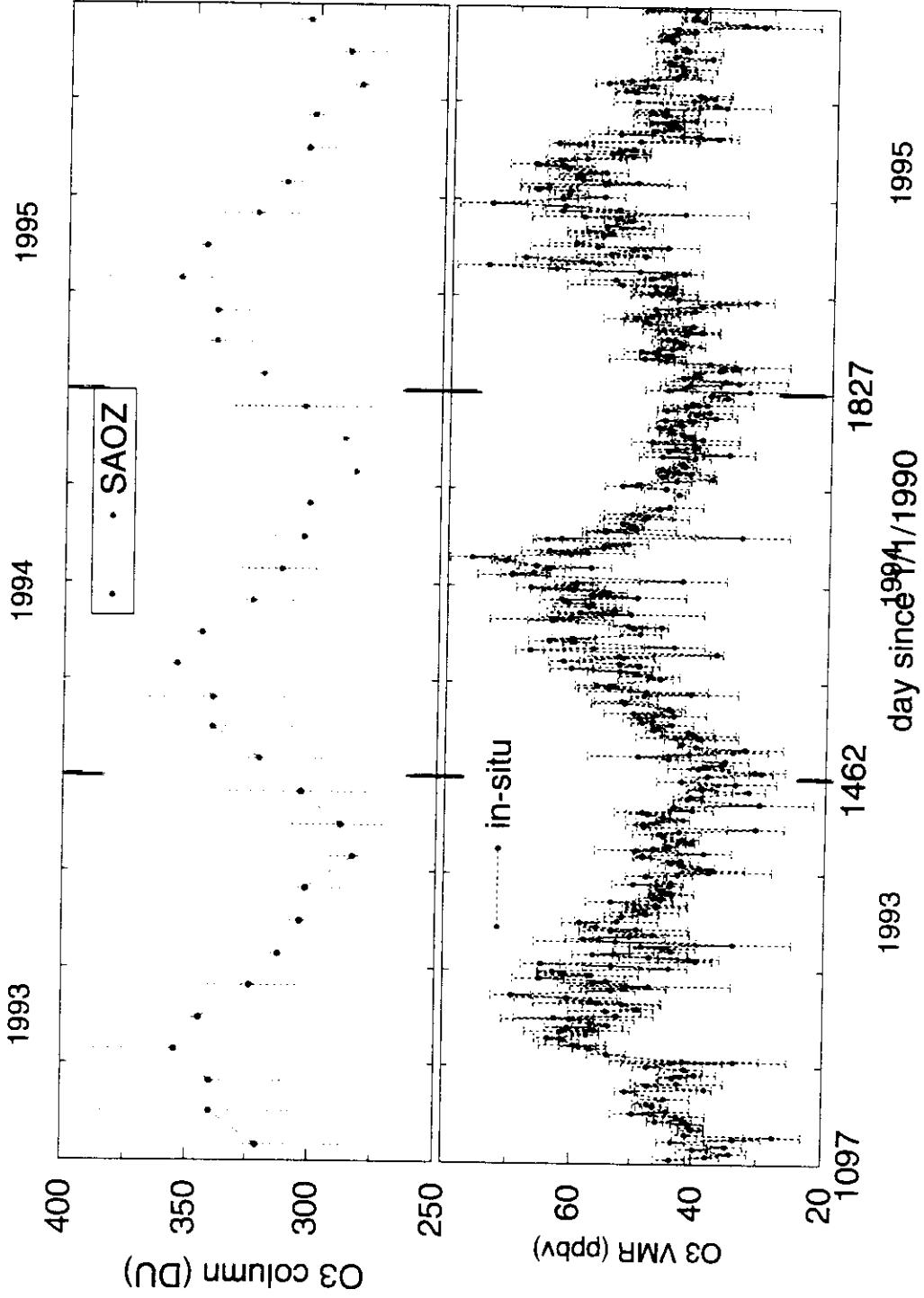
Examples of  $O_3$  + tracers, ClO & OCIO, and  $NO_2$

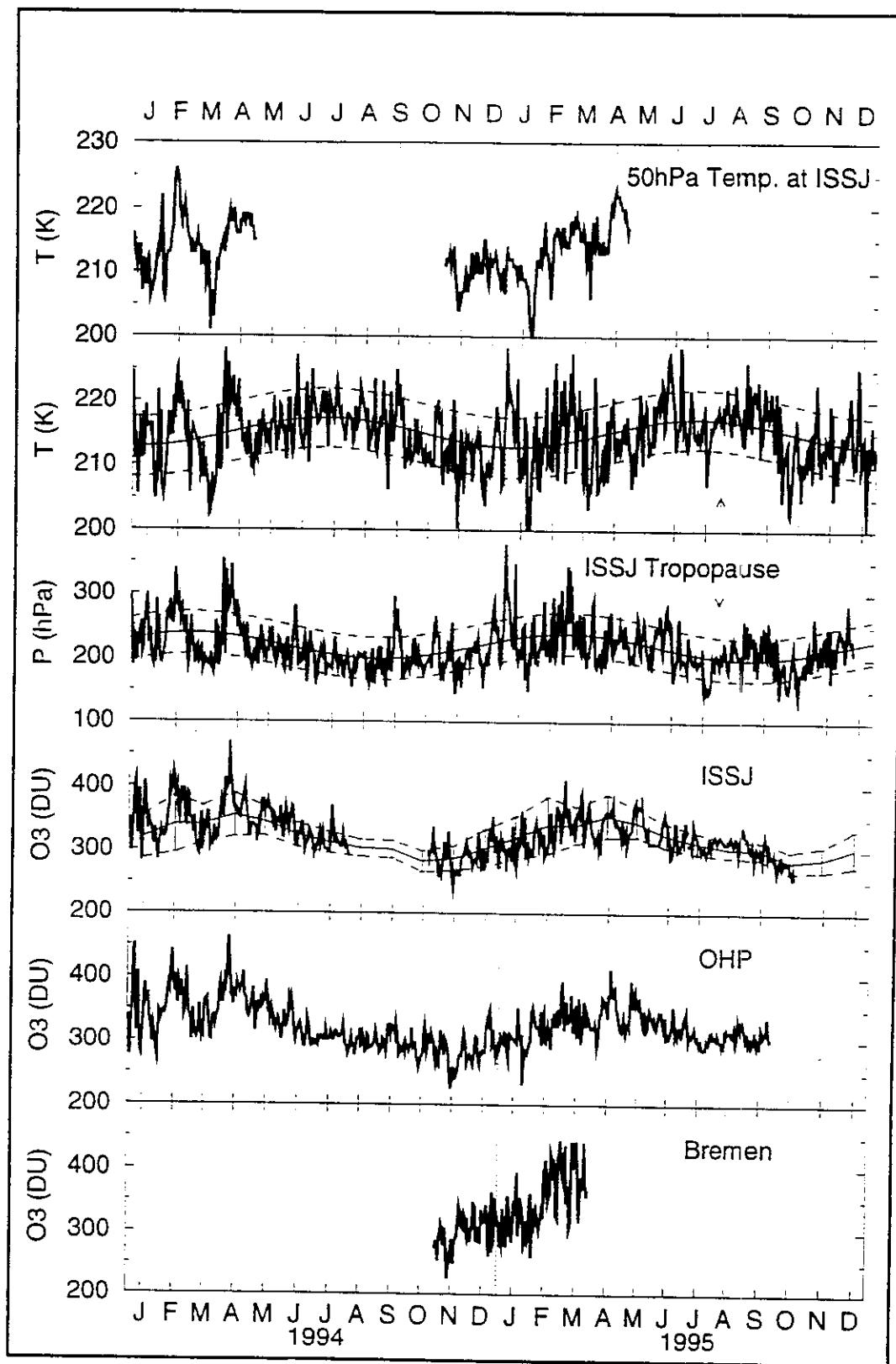
( $O_3$ )

- diurnal variation in the mesosphere, related to solar photolysis
- pronounced seasonal variation, altitude and site-dependent
  - difference as to dominant photochemical processes
- high day-to-day variation
  - to a large extent related to dynamics - cf. tropopause height variations
- $O_3$  destruction events
  - to a large extent related to ‘activation’ of catalytic  $O_3$  destruction catalysts (e.g., ClO, BrO, ...), T, hν

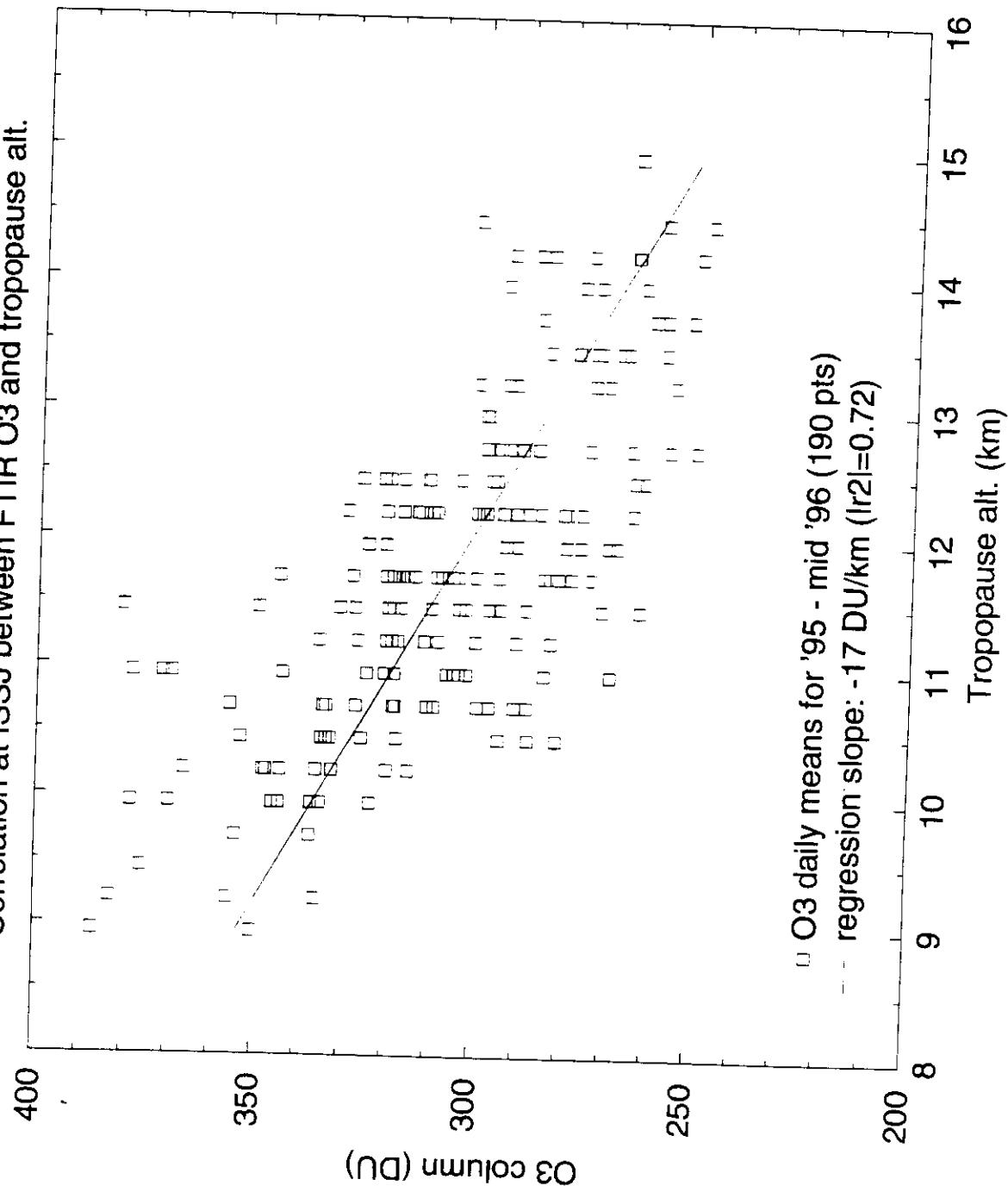
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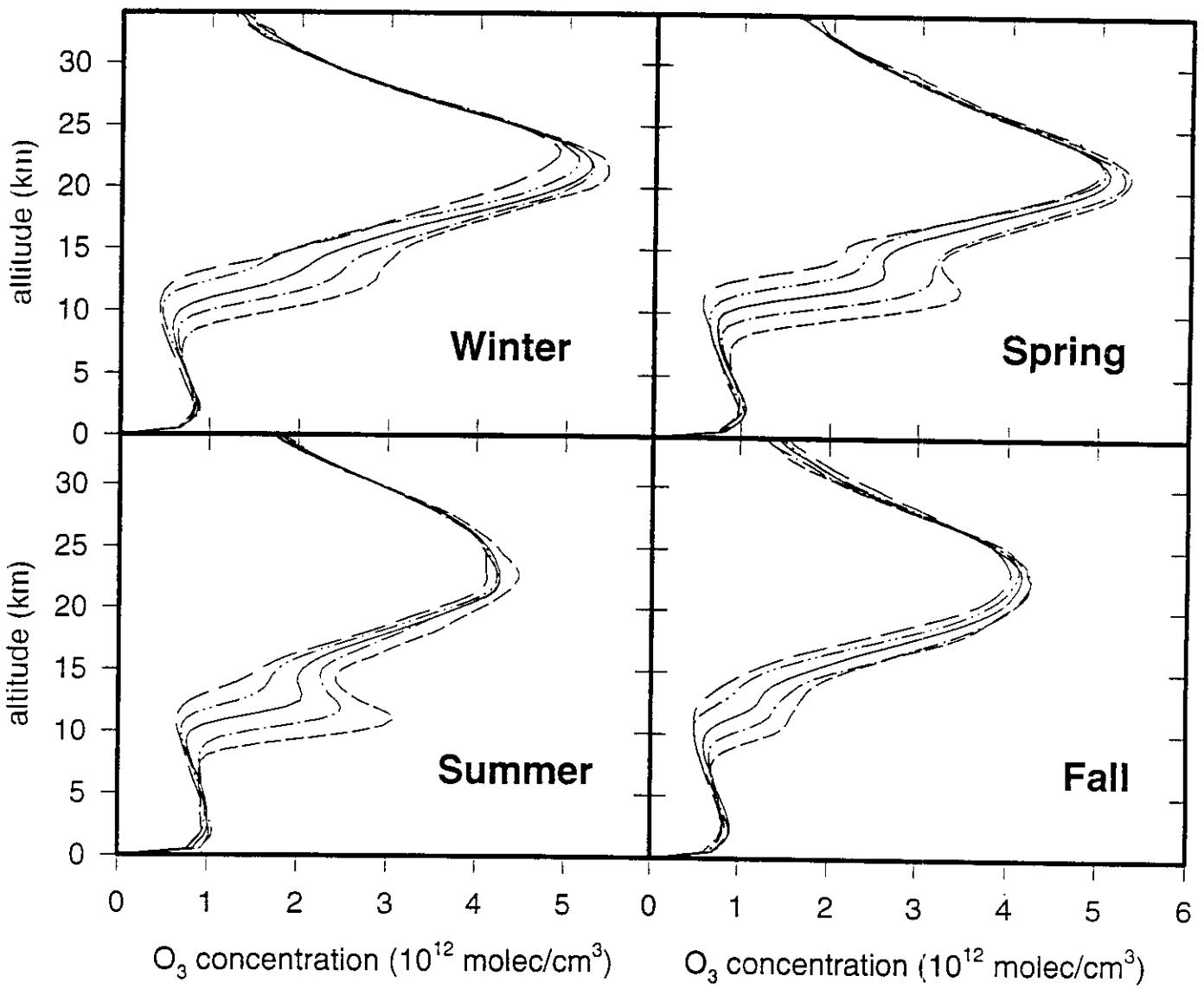
# Jungfraujoch



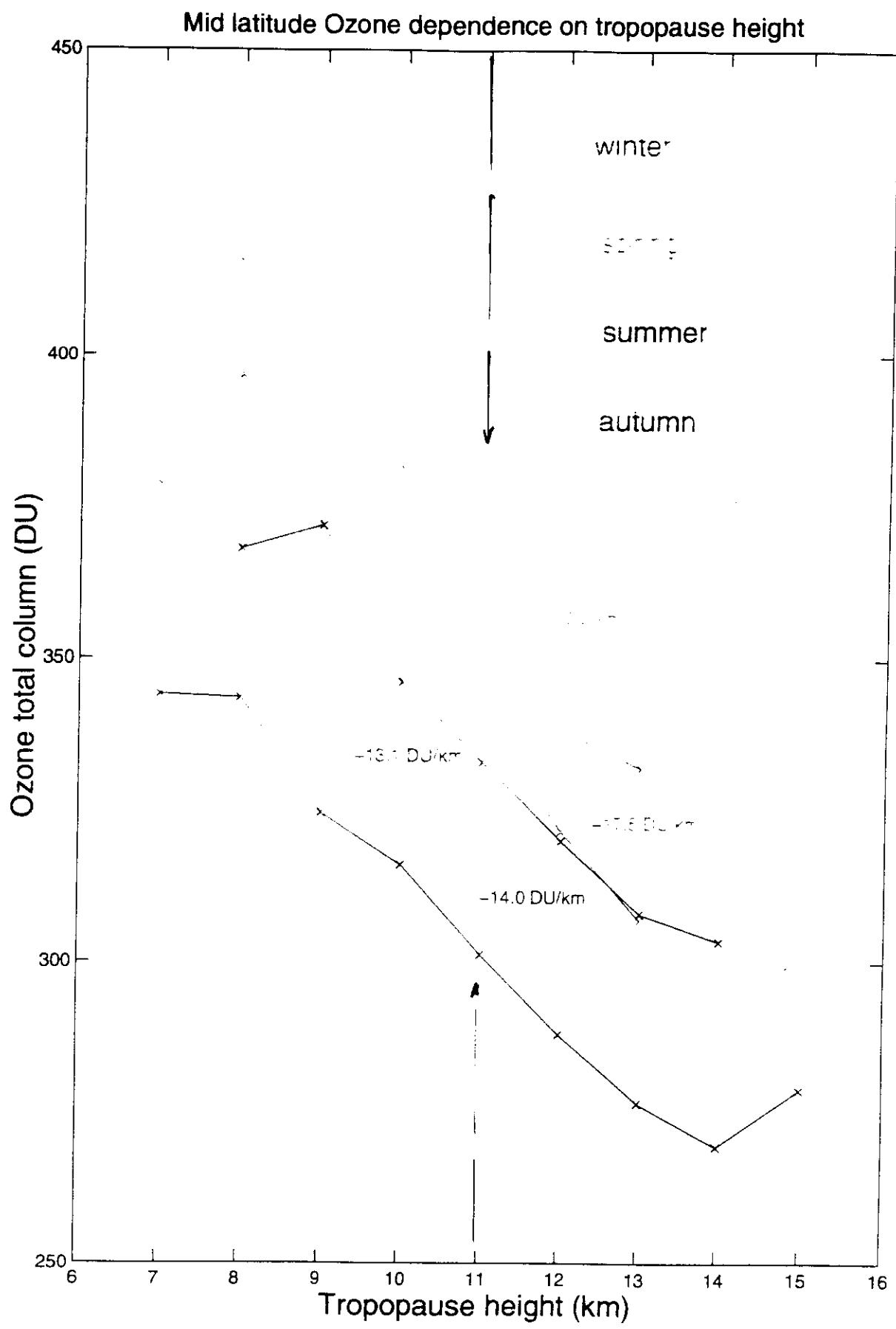


Correlation at ISSJ between FTIR O<sub>3</sub> and tropopause alt.





Tropopause height : 13 km → 9 km



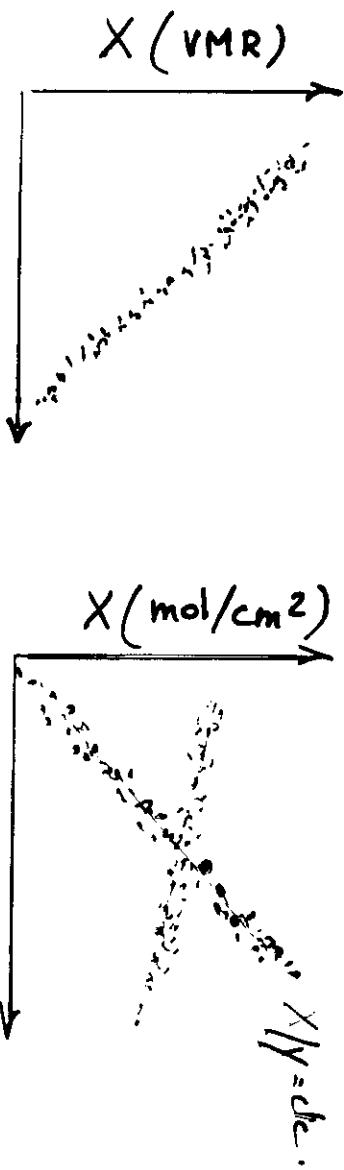
# Atmospheric variabilities:

Examples of O<sub>3</sub> + tracers, ClO & OCIO, and NO<sub>x</sub>

## Tracers of dynamics

e.g., O<sub>3</sub>, HCl, N<sub>2</sub>O, ...

- empirical correlation plot of a chemically active species versus an "inert" tracer represent situation dominated by transport

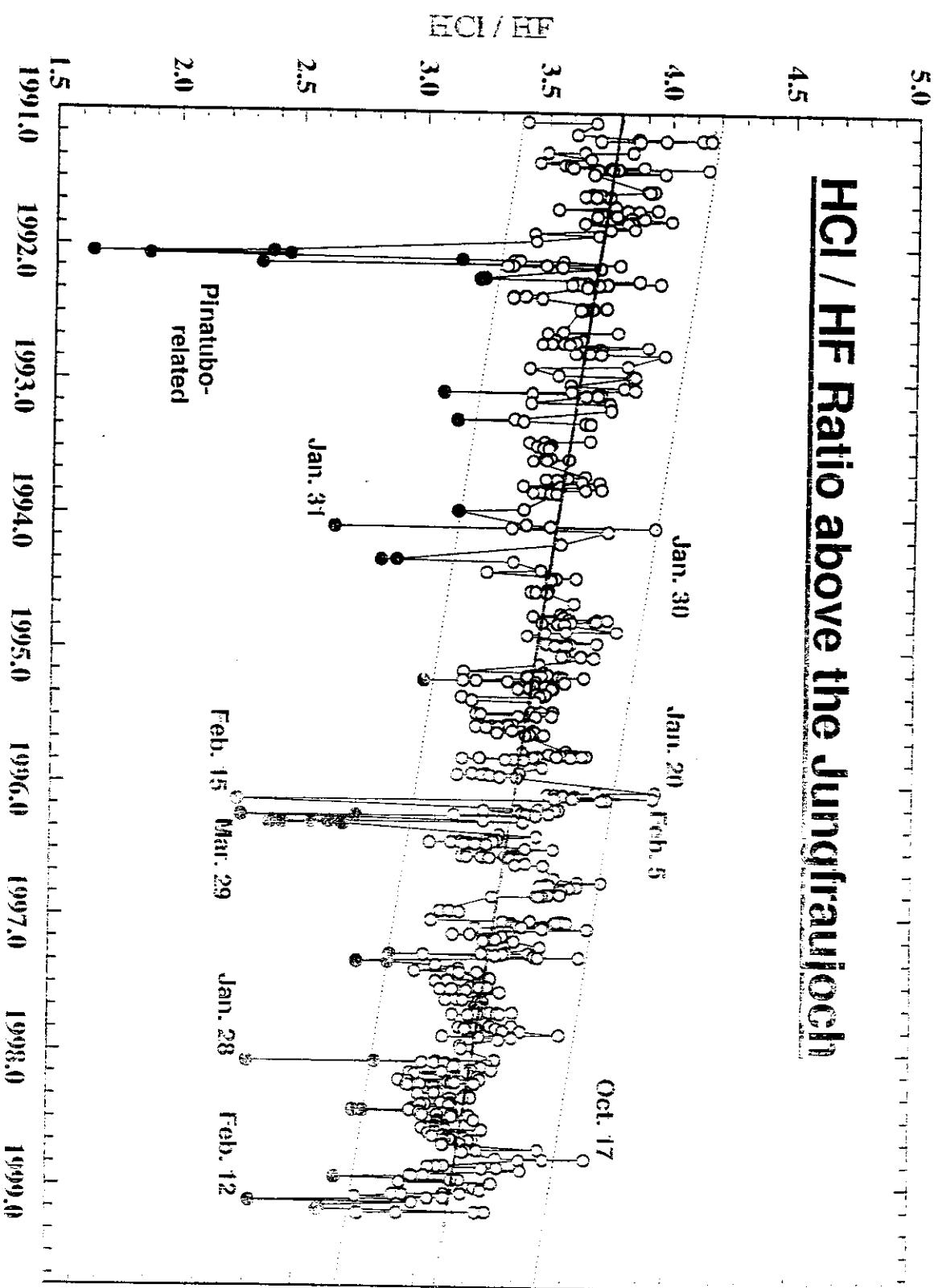


- deviations from normal correlation indicate chemical processing, e.g., HCl versus H, HNO<sub>3</sub> versus N<sub>2</sub>O

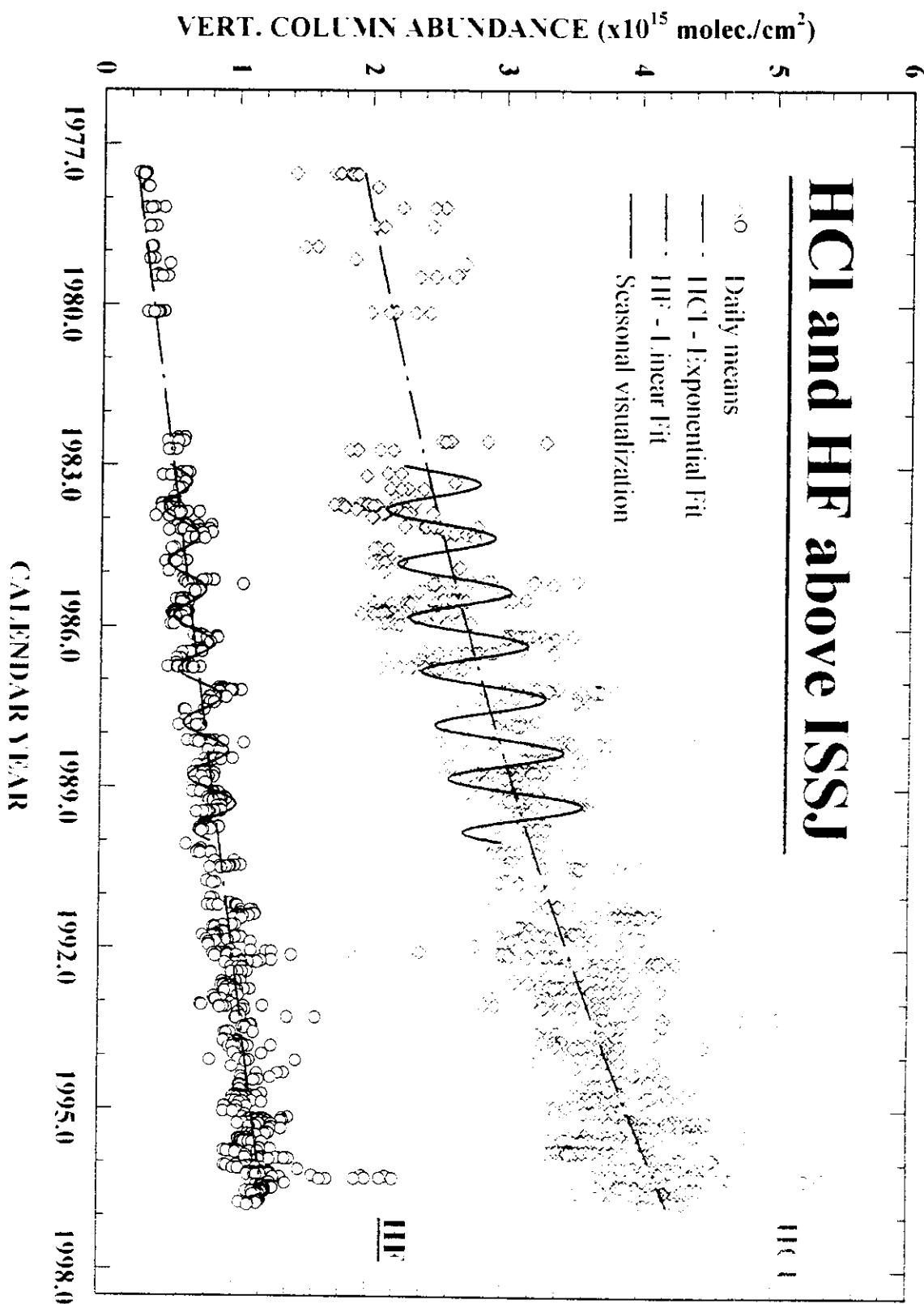
Atmospheric Variability



# HCl / HF Ratio above the Jungfraujoch



## HCl and HF above ISSJ



## Atmospheric variabilities:

Examples of  $O_3$  + tracers, ClO & OCIO, and NO<sub>x</sub>,

(HClO, HOCl)

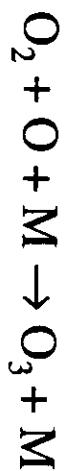
- ClO, BrO are catalysts for  $O_3$  destruction;
- chlorine activation, i.e., increased abundance of ClO, in case of low T, activation of heterogeneous chemistry
  - ⇒ typically in polar vortex conditions, in presence of PSC, or under high aerosol load
- OCIO short-lived component, formed in case of chlorine activation, i.e., high amounts of ClO, possibly with presence of BrO:
  - »  $ClOOCl + h\nu \rightarrow OCIO + Cl$
  - »  $ClO + BrO \rightarrow OCIO + Br$

⇒ Distinction of a long-term trend in activated chlorine ??



## Stratospheric O<sub>3</sub> production / loss

### Chapman atmosphere



Production

Loss

### Catalytic cycles (X = Cl, Br, NO, OH, H, ...)



# Polar Stratospheric Clouds

Type I PSC:

Nitric acid trihydrate ( $\text{HNO}_3 \cdot 3 \cdot \text{H}_2\text{O}$ )  
Ternary solution ( $\text{H}_2\text{O}, \text{H}_2\text{SO}_4, \text{HNO}_3$ )  
195 K  
 $1\mu\text{m}$   
10–24 km  
1km/30 days

Type II PSC:

Formation Temp:  
Particle diameter:  
Altitudes:  
Settling rates:

Water Ice  
188 K  
 $> 10\mu\text{m}$   
10–24 km  
 $> 1.5 \text{ km/day}$

PSC

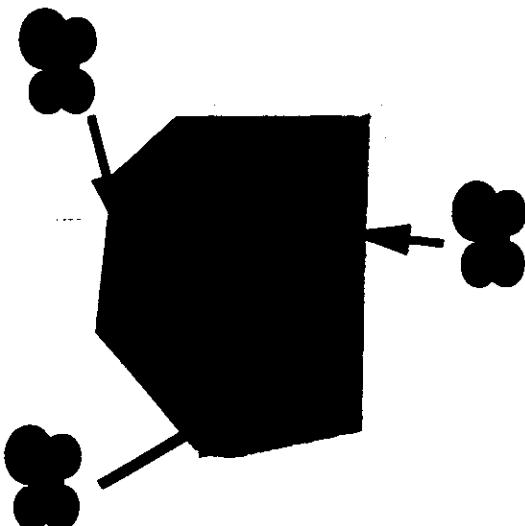


Heterogeneous reactions take place on PSCs, releasing chlorine from reservoir species ( $\text{HCl}$  and  $\text{ClONO}_2$ ) into reactive forms ( $\text{ClO}$ ) that can rapidly destroy ozone.

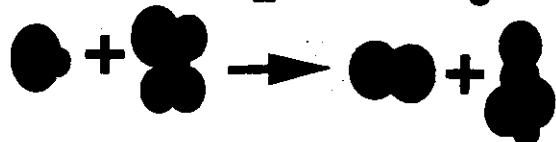
PSC over North Atlantic, January 1989, taken from the NASA DC-8 by O. B. Toon

# Polar Stratospheric Cloud Surface Reaction

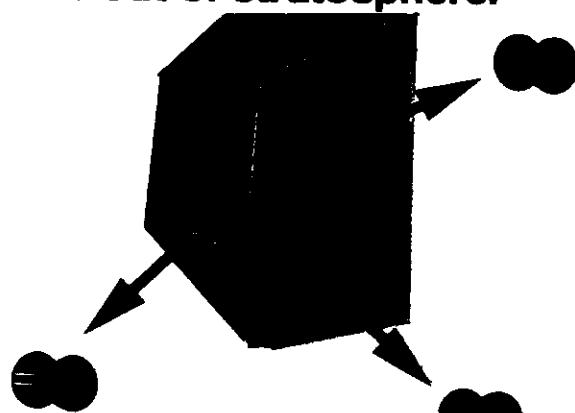
1. HCl and ClONO<sub>2</sub> collect on PSC



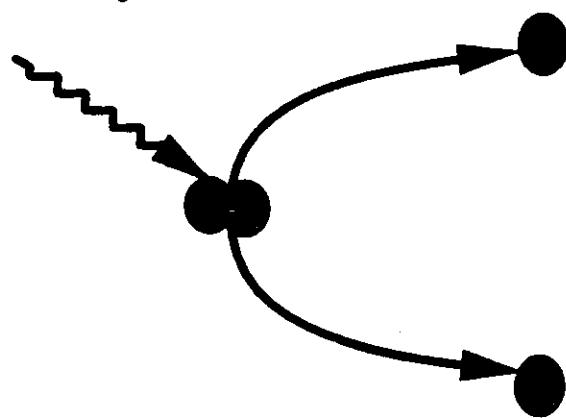
2. HCl and ClONO<sub>2</sub> react on PSC to form Cl<sub>2</sub> and HNO<sub>3</sub>



3. Cl<sub>2</sub> comes off PSC, while HNO<sub>3</sub> remains on PSC to settle out of stratosphere.

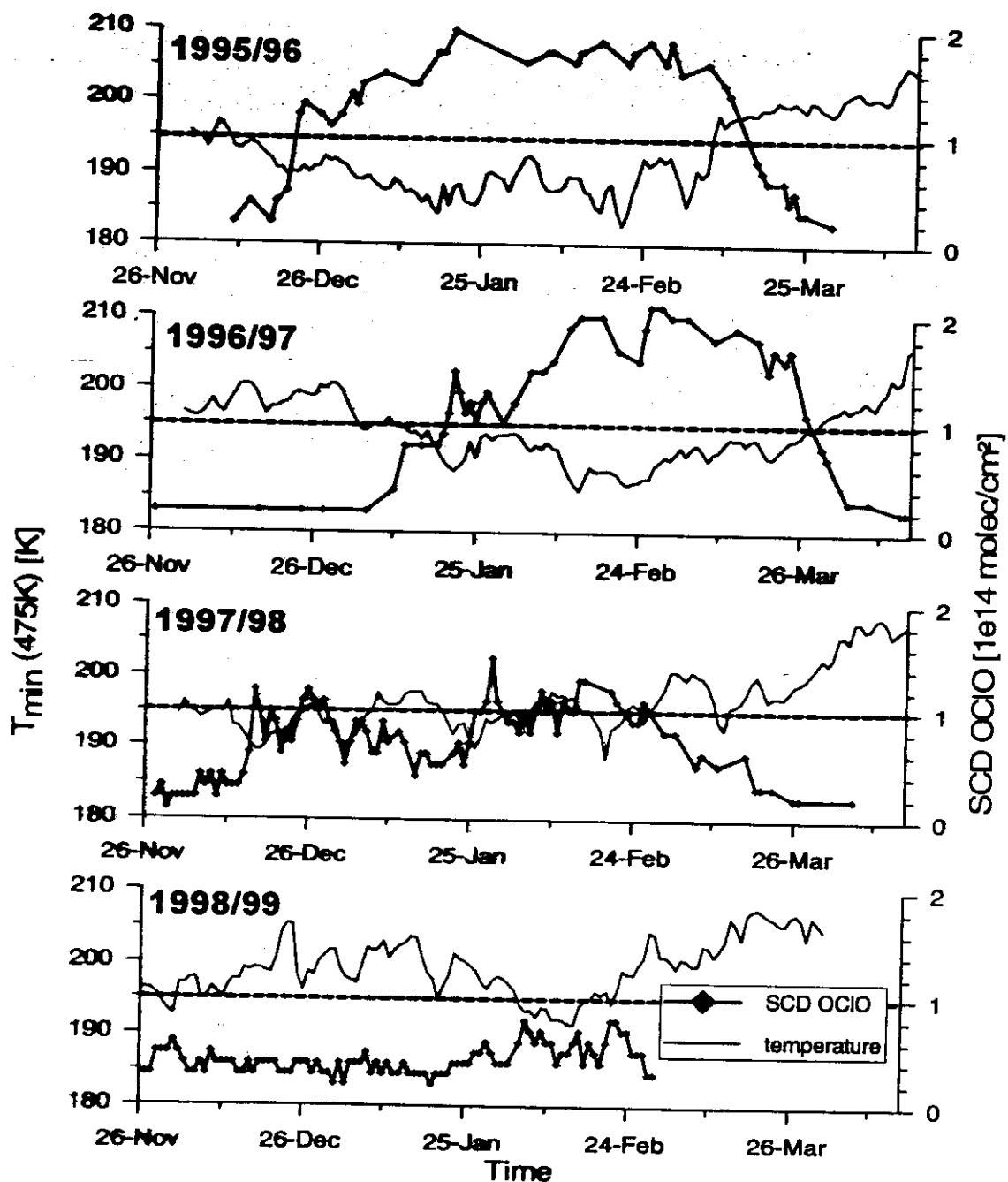


3. Cl<sub>2</sub> is photolyzed by visible wavelengths, and begins catalytic reaction.

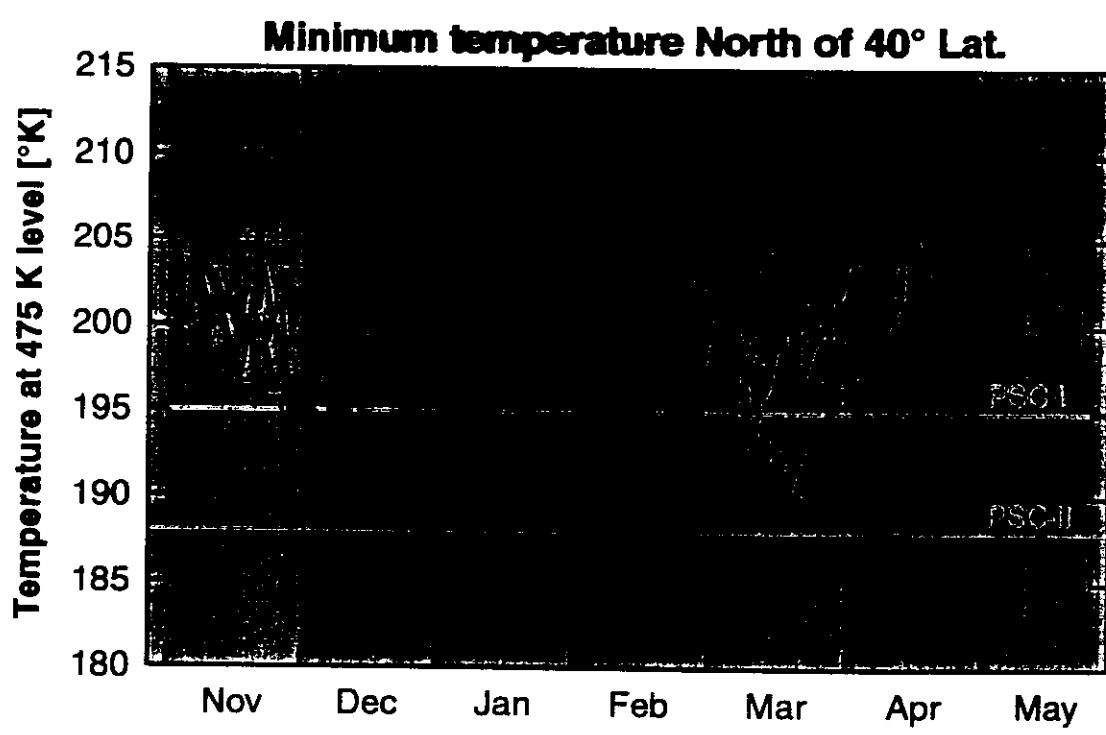
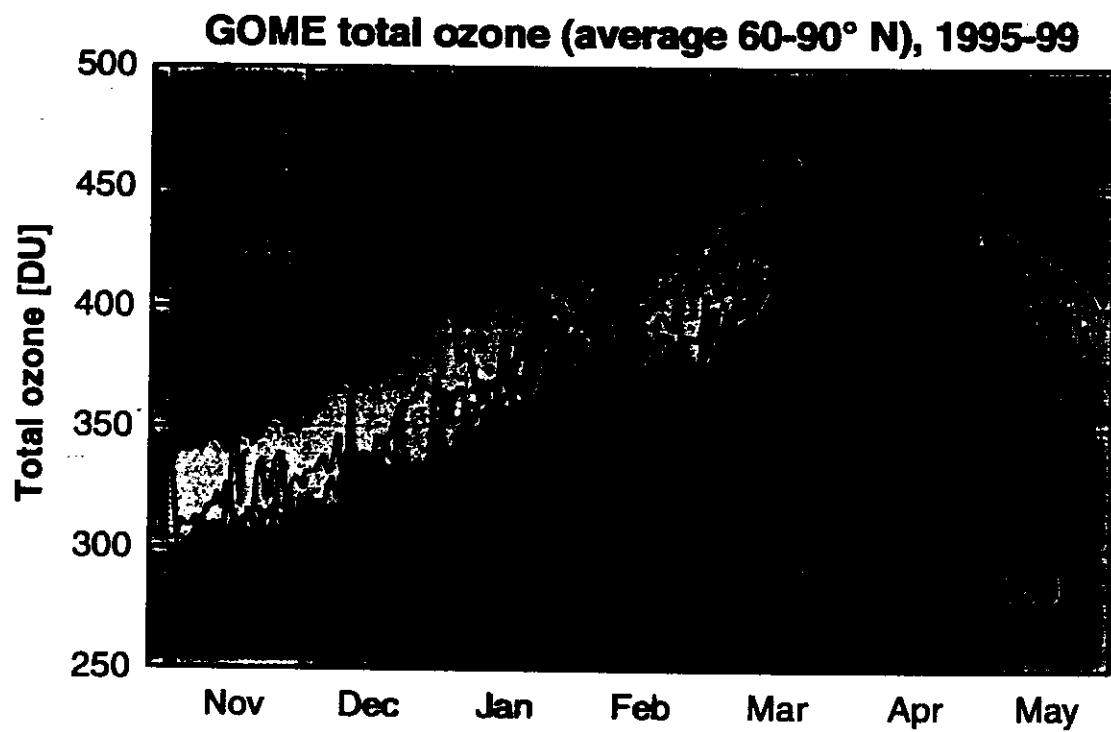




T. Wagner, University of Heidelberg

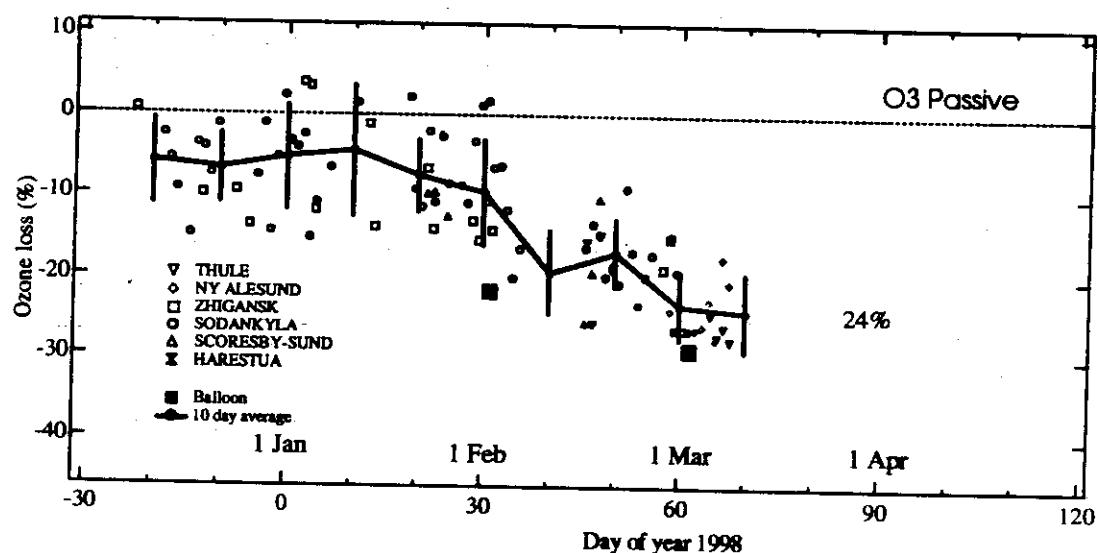


*Time series of the daily maximum OCIO SCDs (at a SZA of 90°) observed by GOME and the minimum stratospheric temperatures at the 475 K level (from ECMWF data) for the four Arctic winters after the launch in 1995. The formation threshold for PSCs is indicated by the dashed line.*

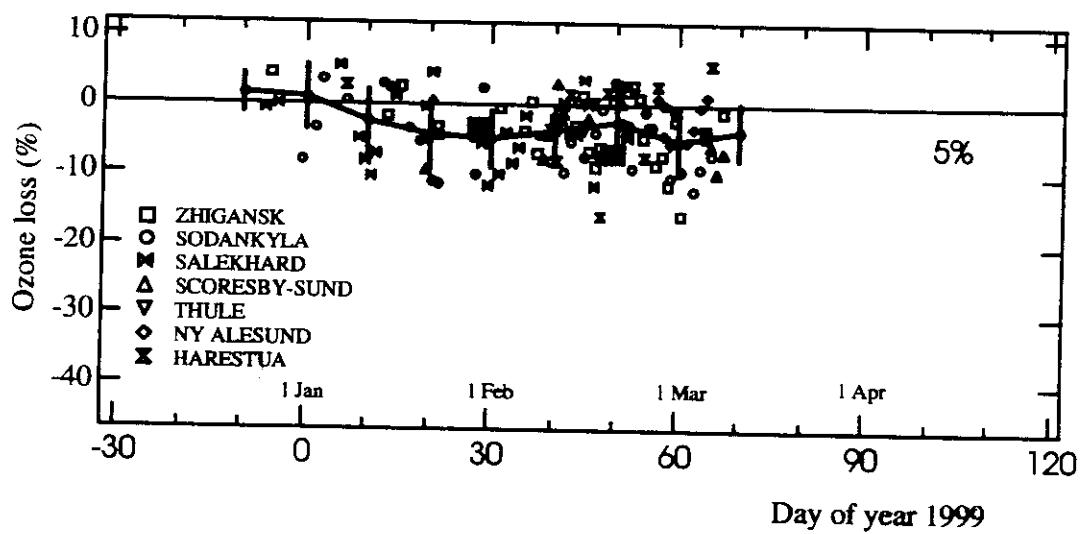


## Ozone loss studies

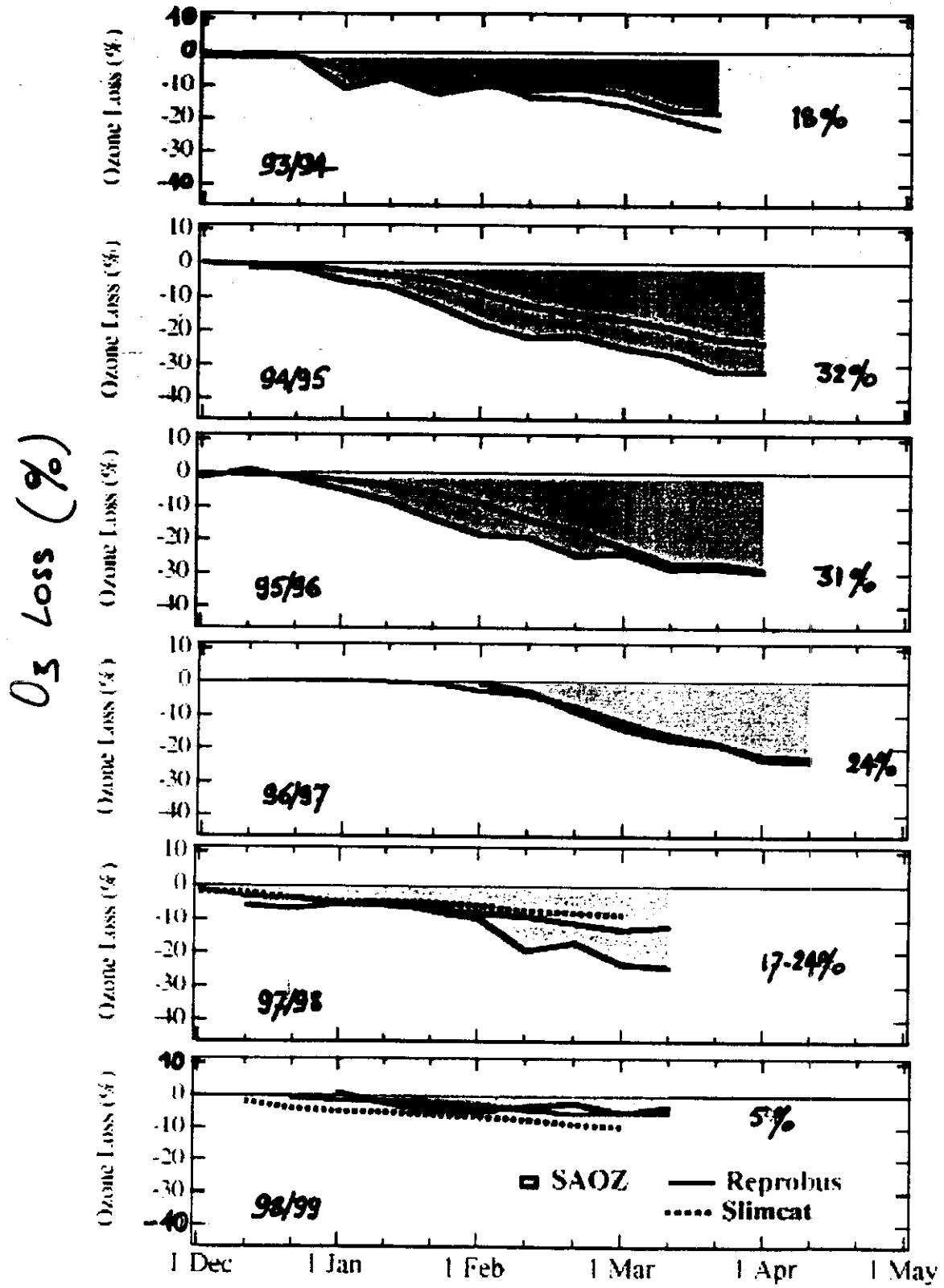
Winter 1997/98



Winter 1998/99



F. Goutail, et al.  
CNRS-SA



# Atmospheric variabilities:

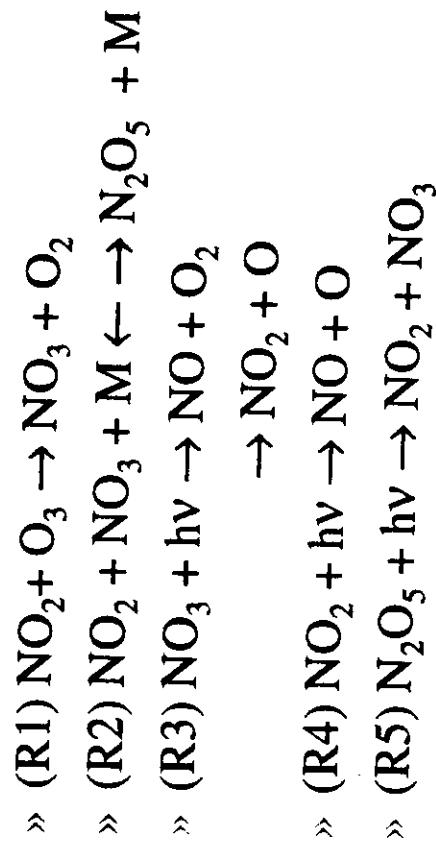
Examples of  $O_3$  + tracers, ClO & OCIO, and  $NO_2$



► pronounced diurnal variation

- related to day-night photochemistry

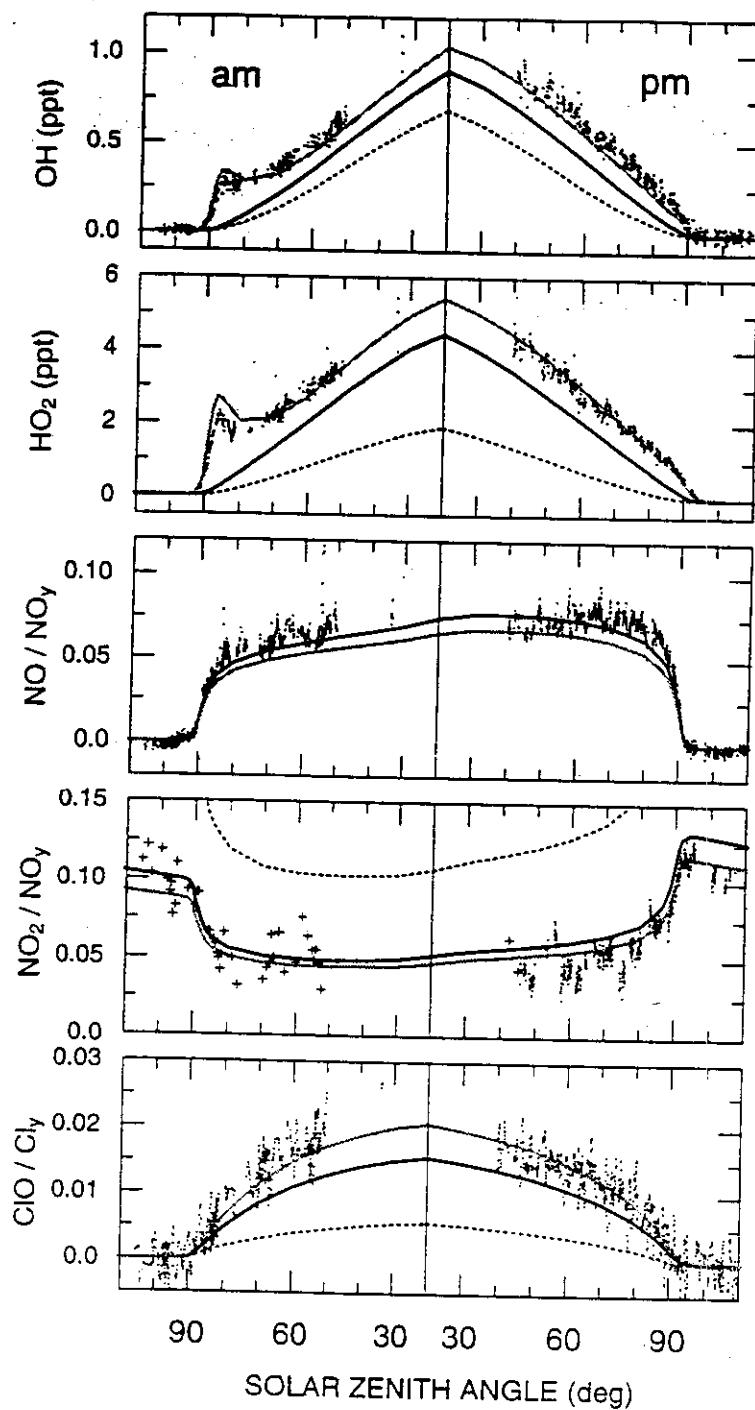
- simplified scheme:



$$\Rightarrow am/pm\ ratio(z) = \exp(-2k_l(z) [Q_2(z)] \Delta T_{night})$$

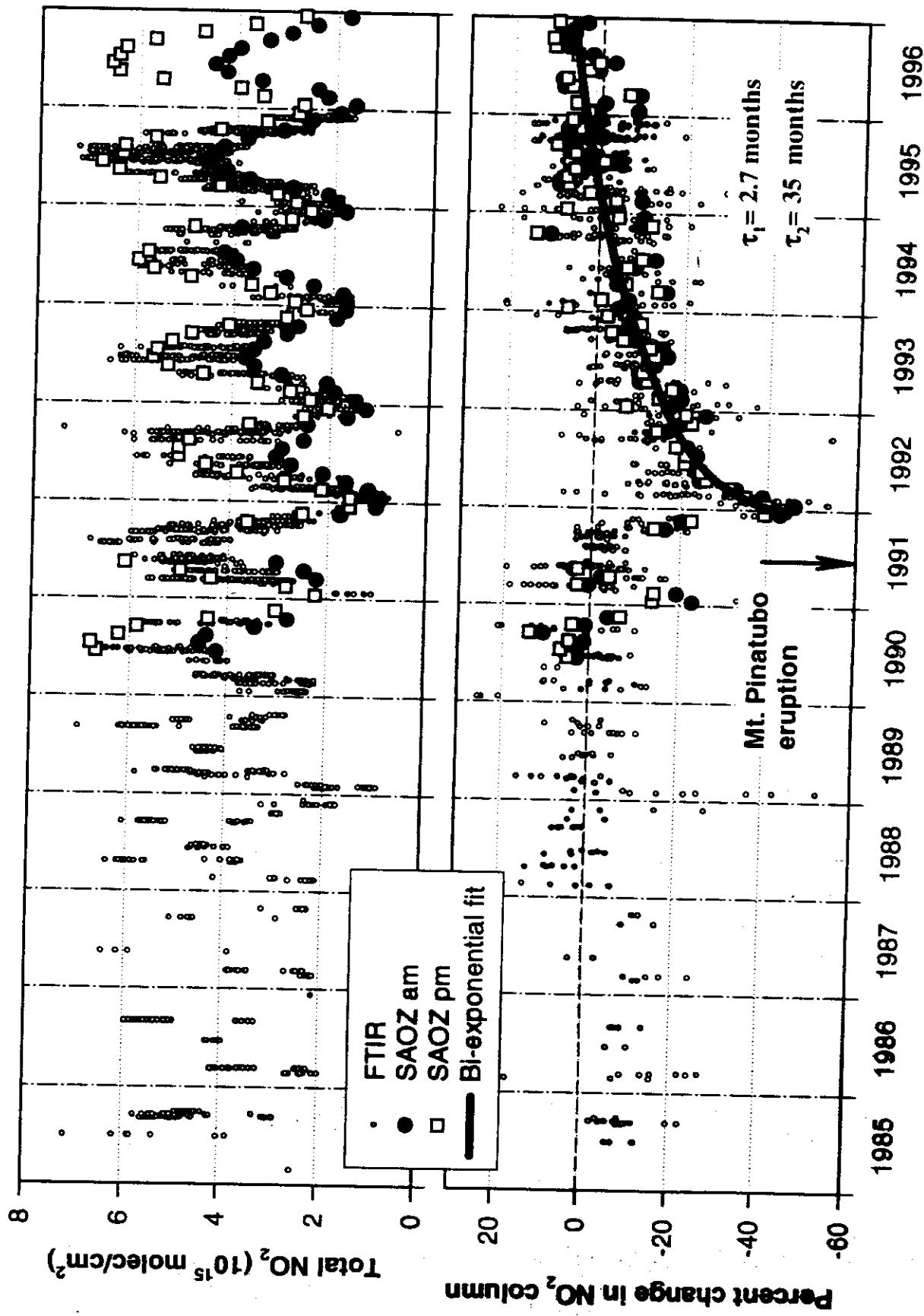
$$\text{with } k_l(z) = 1.2 * 10^{-13} \exp(-2450K/T(z))$$

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**Figure 4-5.** Measurements (dots) of the diurnal variations of stratospheric free radicals  $\text{NO}_2$ ,  $\text{NO}$ ,  $\text{HO}_2$ ,  $\text{OH}$  (crosses and dots represent data from the JPL and NOAA instruments, respectively), and  $\text{ClO}$  from two ER-2 flights of May 11 (sunrise) and May 12 (sunset), both near  $37^\circ\text{N}$  and  $63 \text{ hPa}$  and  $[\text{N}_2\text{O}]$  between 240 and 260 ppbv, plotted as a function of solar zenith angle. Also shown are results from a constrained data assimilation model (Salawitch *et al.*, 1994a). Three calculations are shown. Dark dotted curve: gas phase reactions only, using rate constants and cross sections of DeMore *et al.* (1992). Curve 1, dark solid line: as for above, except including also the heterogeneous hydrolysis of  $\text{N}_2\text{O}_5$  and  $\text{ClONO}_2$ . Curve 2, gray line: as for curve 1, except including the heterogeneous decomposition of  $\text{HNO}_4$  to form  $\text{HONO}$ , the  $\text{O}^{\text{(1D)}}$  quantum yield of Michelsen *et al.* (1994), and the temperature-dependent cross sections of  $\text{HNO}_3$  from Burkholder *et al.* (1993). (From Salawitch *et al.*, 1994a.)

# Total NO<sub>2</sub> reduction at the Jungfraujoch after Mt. Pinatubo eruption



**Atmospheric variabilities:**  
Examples of  $O_3$  + tracers, ClO & OClO, and  $NO_2$

$\text{NO}_2$  (cont'd)

## ► interannual variations

- example 1: impact of Mt. Pinatubo eruption

- dominant mechanism behind:



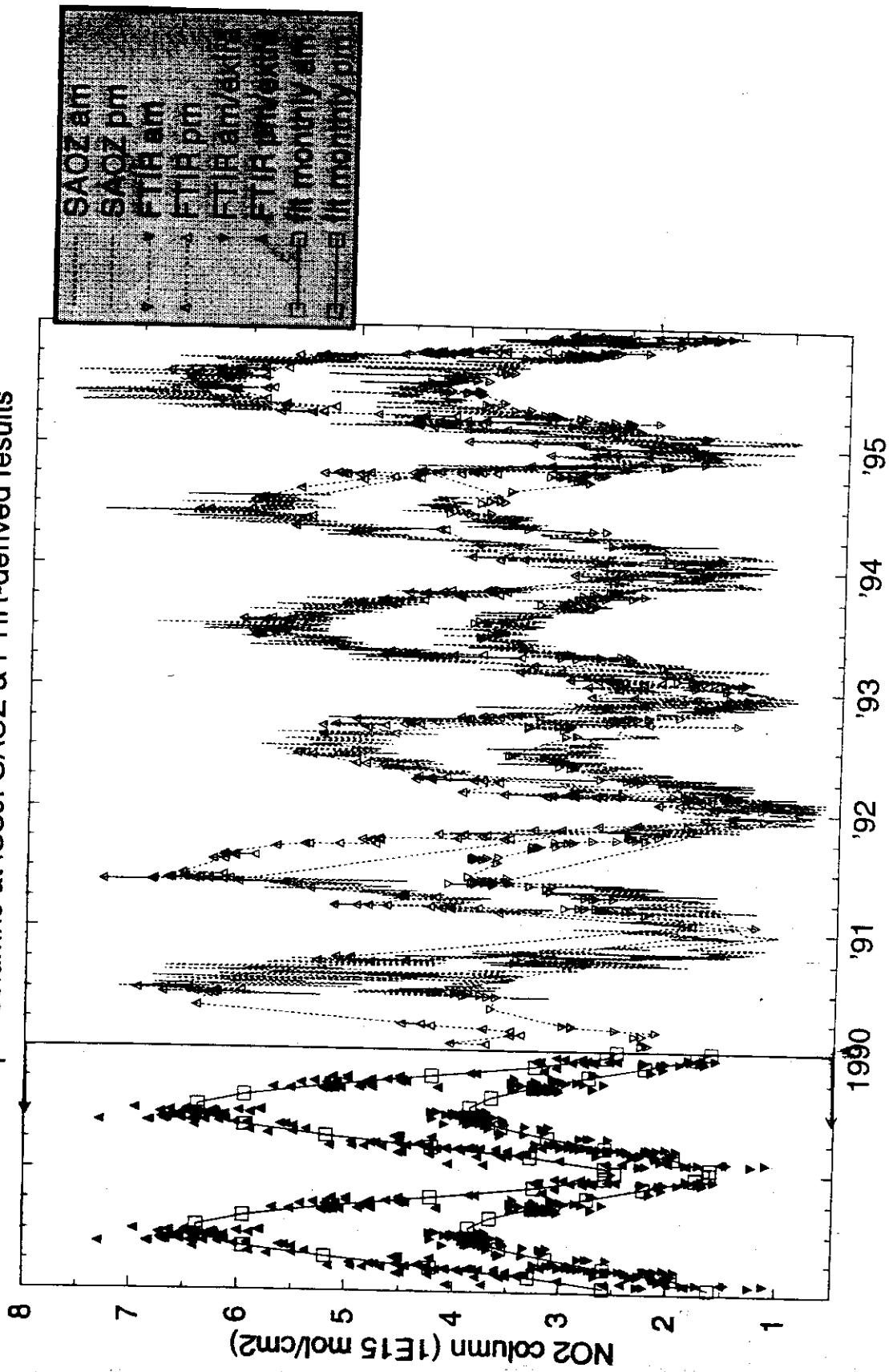
## on aerosol/PSC surface

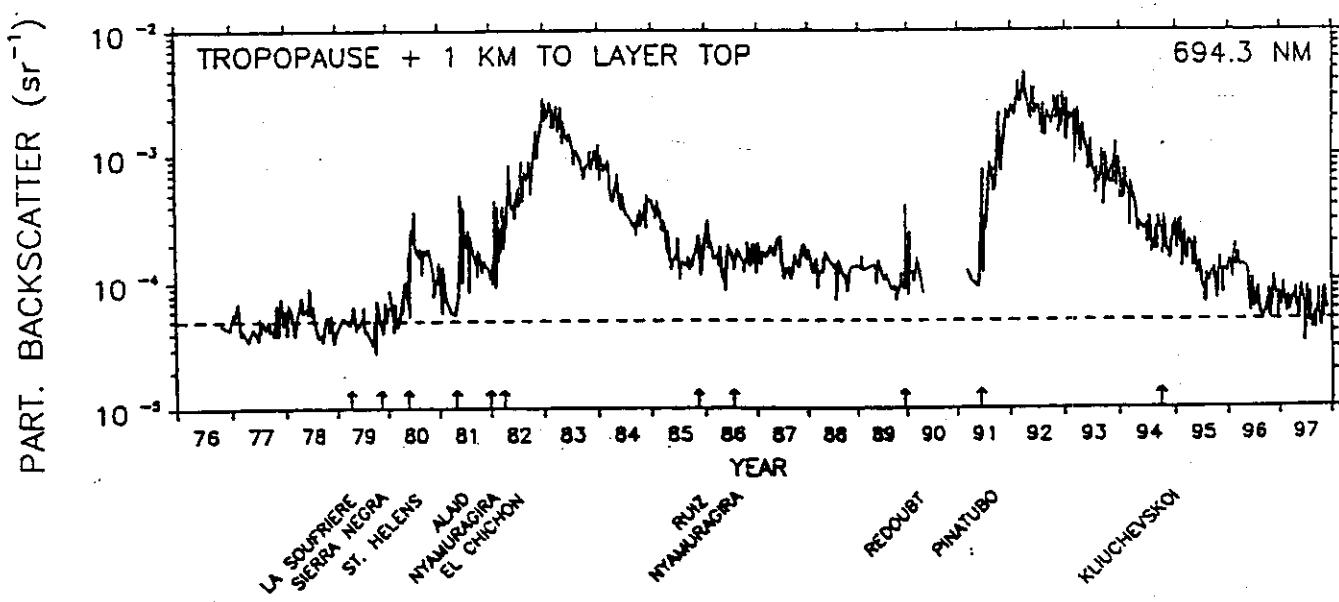
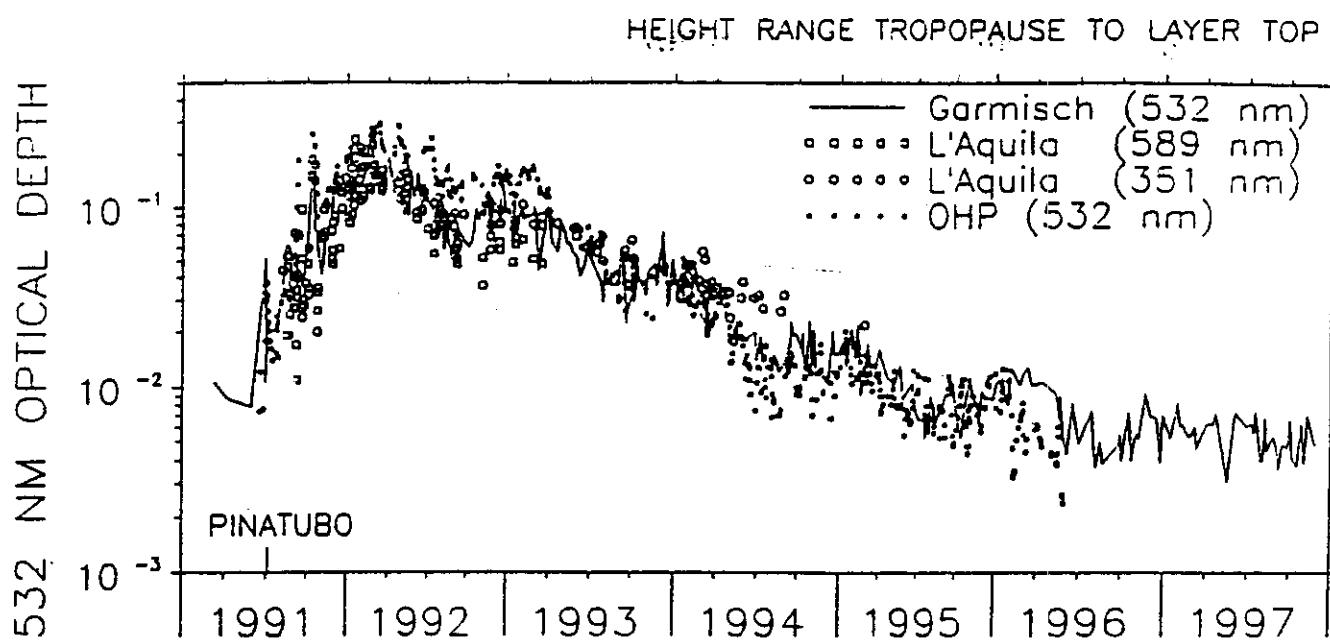


... (+ HCl)

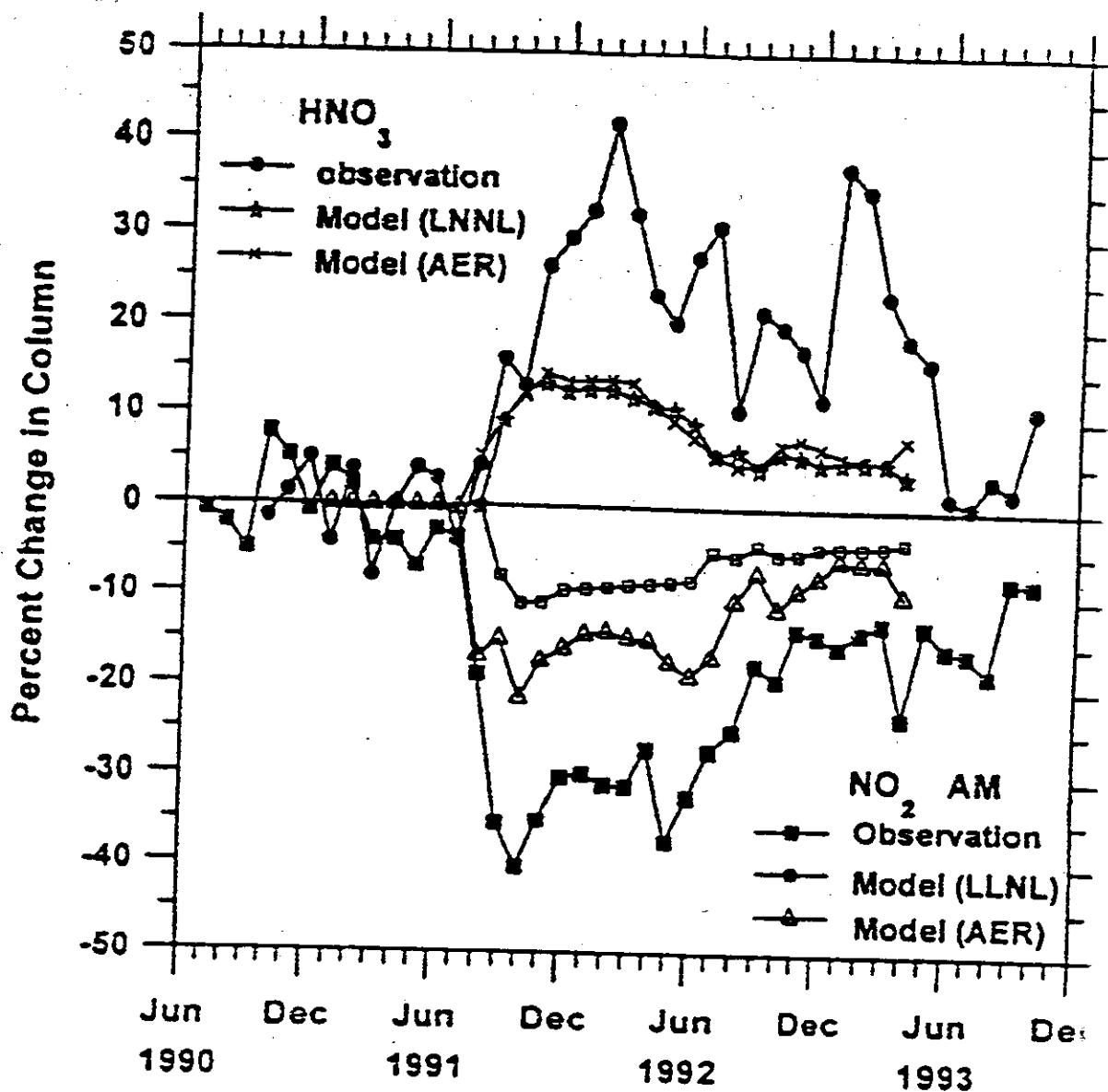
- example 2: denoxification / denitrification in winter
    - » similar reactions on PSC / settling out of  $\text{HNO}_3$

NO<sub>2</sub> am & pm columns at ISSJ: SAOZ & FTIR-derived results





H. Jaeger et al.



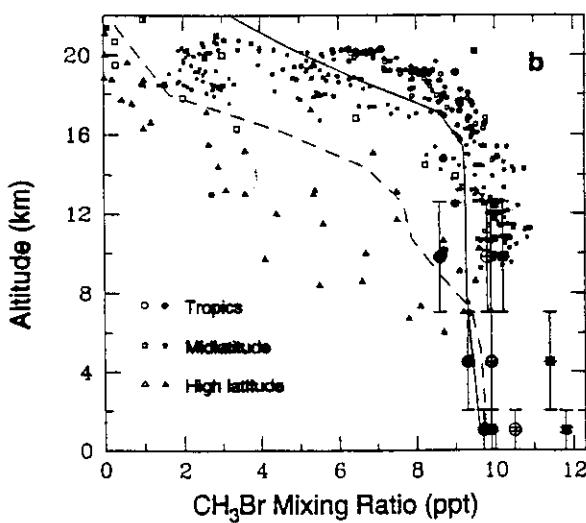
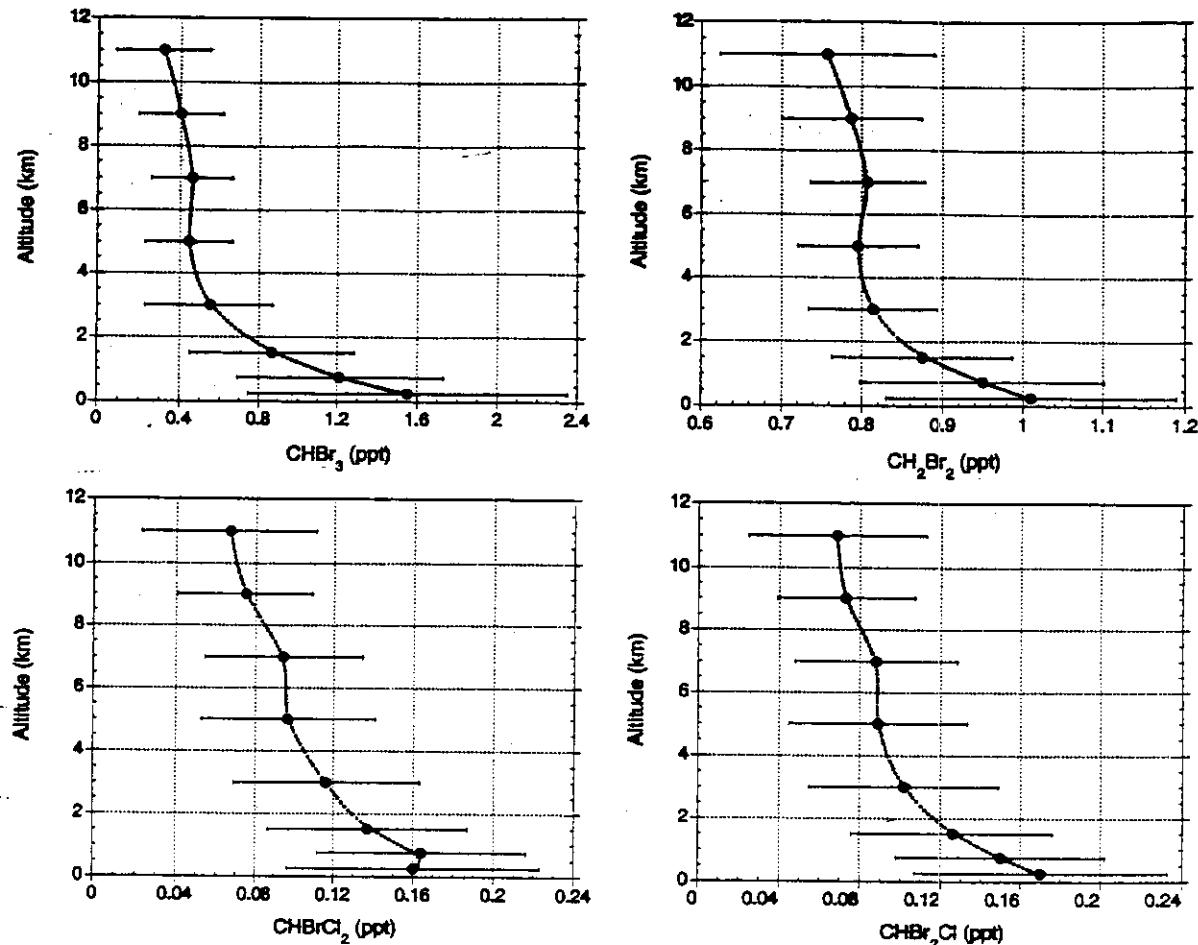
**Figure 4-9.** Percentage changes in  $\text{HNO}_3$  and  $\text{NO}_2$  column amounts above Lauder, New Zealand, ( $45^\circ\text{S}$ ) following the arrival of the Mt. Pinatubo aerosol. The Lawrence Livermore National Laboratory (LLNL) results are for  $42.5^\circ\text{S}$  and the Atmospheric Environmental Research, Inc. (AER) results are for  $47^\circ\text{S}$ . Heterogeneous chemistry is included in the calculations based on the observed aerosol field from SAGE II. (From Koike *et al.*, 1994.)

# Atmospheric long-term changes

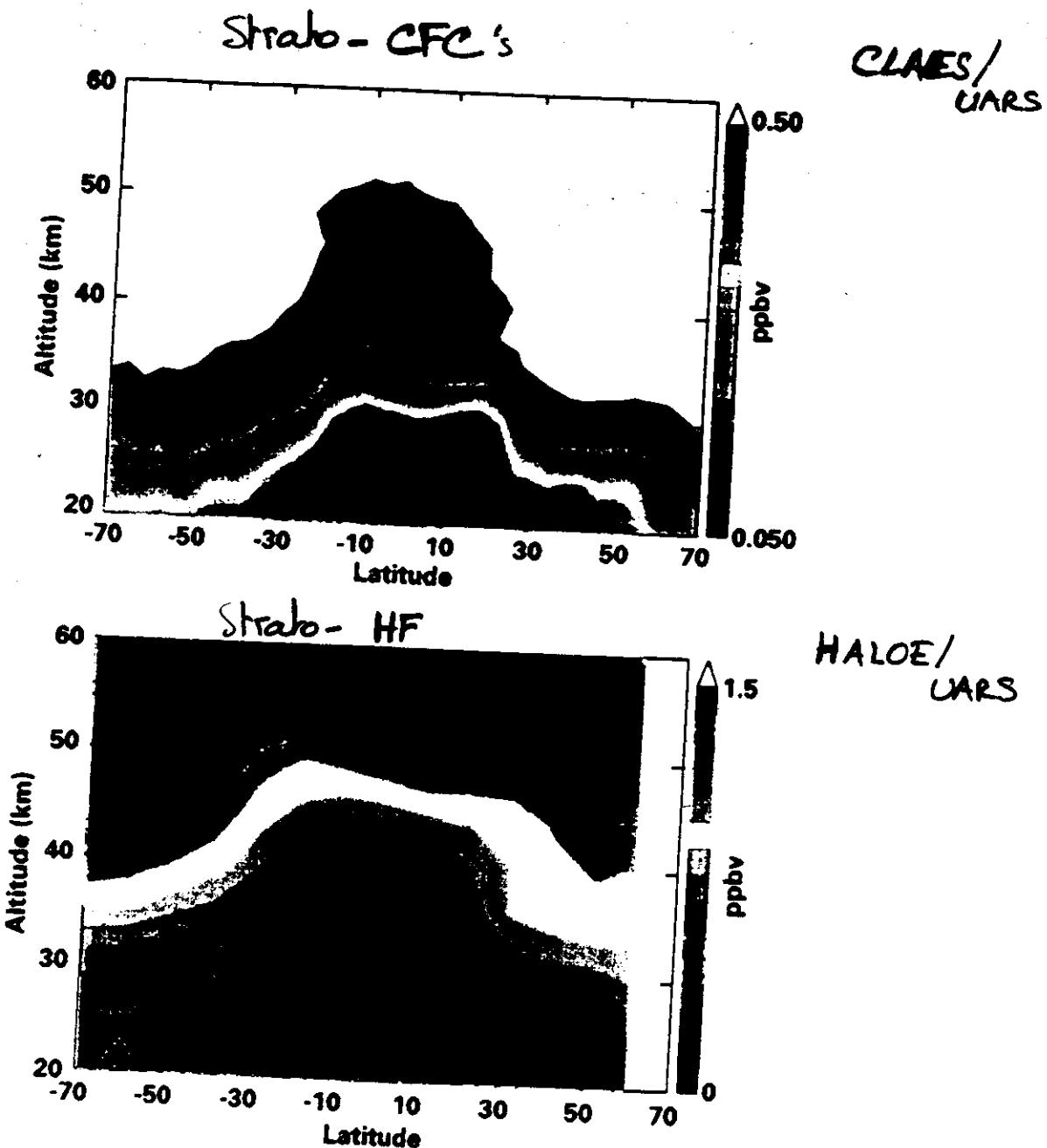
- halogen compounds:
  - short-lived (< 1 yr) and long-lived (> 1 yr) constituents
    - lifetime determines vertical distribution profile, in particular survival into the stratosphere
    - degradation, essentially by
      - photolysis
      - reaction with OH
  - sources = halocarbons, incl. HCFC
  - greenhouse gases (GHG)
  - temperature, dynamics, aerosol/PSC, clouds, ....
  - Ozone: tropo- and strato-

Industrial Name	Chemical Formula	Lifetime, WMO (1998) (years)	Actual Abundance / Source
Nitrous oxide	N <sub>2</sub> O	120	
CFC-11	CCl <sub>3</sub> F	45	③ ≈ 50% of [CH <sub>3</sub> Cl]
CFC-12	CCl <sub>2</sub> F <sub>2</sub>	100	② ≈ [CH <sub>3</sub> Cl]
CFC-113	CCl <sub>2</sub> FCClF <sub>2</sub>	85	
Carbon tetrachloride	CCl <sub>4</sub>	35	④ ≈ 20% of [CH <sub>3</sub> Cl]
H-1211	CBrClF <sub>2</sub>	11	
H-1301	CBrF <sub>3</sub>	65	
Methyl chloroform	CH <sub>3</sub> CCl <sub>3</sub>	4.8	
HCFC-22	CHClF <sub>2</sub>	11.8	
HCFC-141b	CH <sub>3</sub> CCl <sub>2</sub> F	9.2	
HCFC-142b	CH <sub>3</sub> CClF <sub>2</sub>	18.5	
HFC-134a	CH <sub>2</sub> FCF <sub>3</sub>	13.6	
HFC-23	CHF <sub>3</sub>	243	
Methane	CH <sub>4</sub>	8.9	
Methyl chloride	CH <sub>3</sub> Cl	~ 1.3	① Most abundant halocarbon <b>natural origin:</b> biomass burning, ocean, wood-rotting fungi
Methyl bromide	CH <sub>3</sub> Br	0.7	<b>natural origin:</b> oceans, fumigation, biomass burning; 50% anthropogenic (automobile)!
methylene chloride	CH <sub>2</sub> Cl <sub>2</sub>	5 to 6 months	
chloroform	CHCl <sub>3</sub>	~ 6 months	
trichloroethene	C <sub>2</sub> HCl <sub>3</sub>	~ 1 week	
tetrachloroethene	C <sub>2</sub> Cl <sub>4</sub>	3 to 4 months	
phosgene	COCl <sub>2</sub>	70 days	
methyl iodide	CH <sub>3</sub> I	4 days at surface 1.5 days (10 km)	
ethyl iodide	C <sub>2</sub> H <sub>5</sub> I	Similar to CH <sub>3</sub> I	
chloroiodomethane	CH <sub>2</sub> ClI	~ 100 minutes	
diiodomethane	CH <sub>2</sub> I <sub>2</sub>	2.5 minutes	
isopropyl iodide	CH <sub>3</sub> CHICH <sub>3</sub>	~ 16 hours	
1-iodopropane	CH <sub>3</sub> CH <sub>2</sub> CH <sub>2</sub> I	~ 1 day	
iodotrifluoromethane	CF <sub>3</sub> I	1 day	
dibromochloromethane	CHBr <sub>2</sub> Cl	≤ 120 days	
bromoform	CHBr <sub>3</sub>	≤ 36 days	
methylene bromide	CH <sub>2</sub> Br <sub>2</sub>	≤ 130 days	
bromodichloromethane	CHBrCl <sub>2</sub>	≤ 120 days	

**Figure 2-11.** Vertical profiles of individual reactive organic Br gases, total reactive organic Br, and fraction of reactive/total organic Br over the Pacific Ocean (adapted from Schauffler et al., 1998b). Mean and standard deviation are plotted.

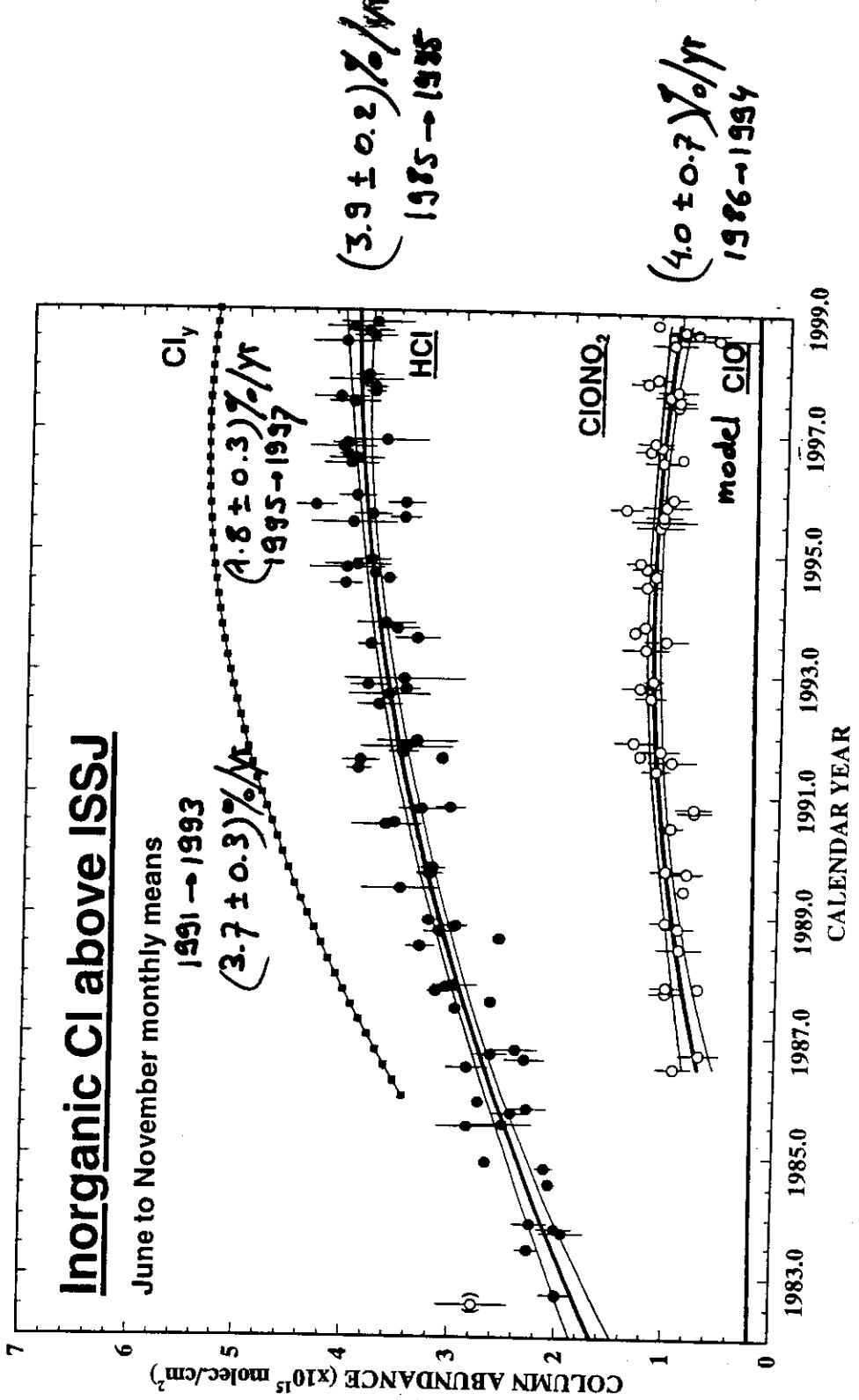


**Figure 2-10.** (a) Vertical profiles of CH<sub>3</sub>Br measured at various latitudes during winter (filled symbols) and summer (open symbols) using different techniques (big symbols - Blake et al., 1997; small symbols - Schauffler et al., 1998a; medium symbols - Lai et al., 1994, Kourtidis et al., 1998). Some of the data are normalized to 10 ppt at the tropospheric level. Measurements are compared with the NASA Goddard Space Flight Center 2-D model (Jackman et al., 1996) results; continuous line is for 15°N and dotted line is for 65°N. (b) Data shown on linear scale for greater clarity.



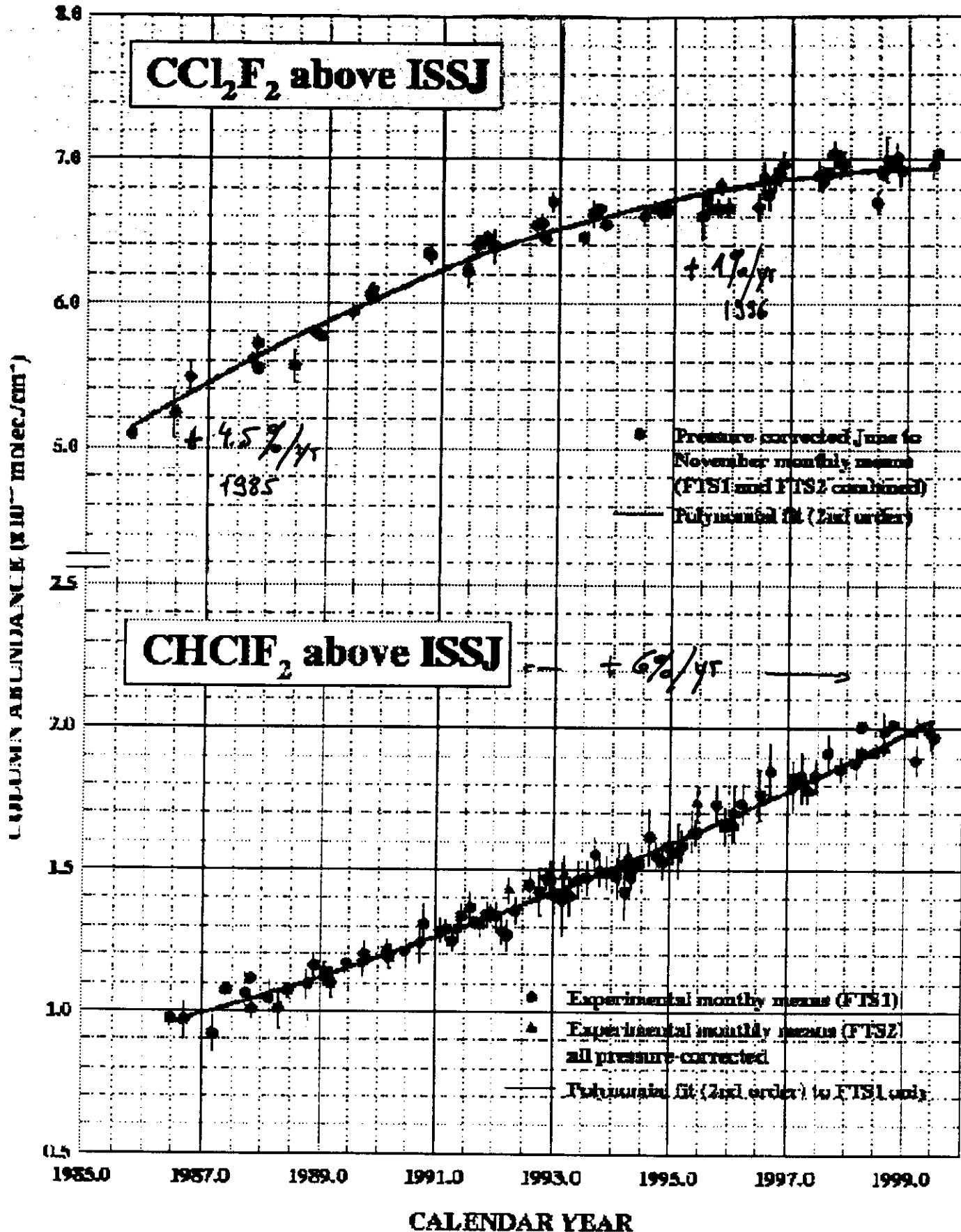
## Atmospheric long-term changes: halogens (2/2)

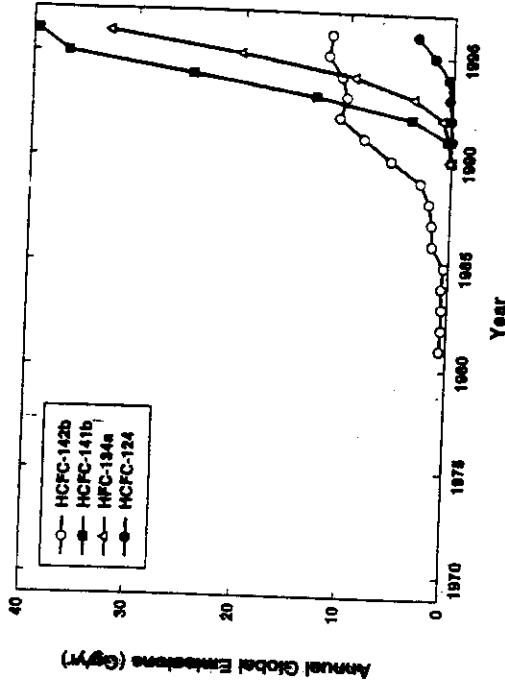
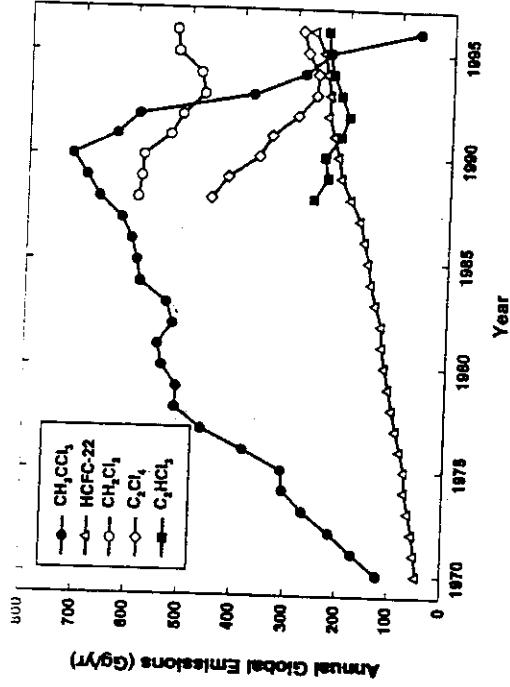
- Halogens: Cl, Br, F (I) -containing
- in the *stratosphere*:
  - 'aggressivity'/atom vis-à-vis O<sub>3</sub> or catalytic efficiency  $\alpha$ :  
 $\alpha_{\text{Br}} \cong 40\text{-}60 > \alpha_{\text{Cl}} \cong 1 > \alpha_{\text{F}} \cong 0$  ( $10^{-4}$ )  
cf. partitioning between reservoirs and active species depends on relative reaction rates;
    - Br mostly (>50%) in form of BrO
      - cf. HBr + OH  $\rightarrow$  BrO *fast*
    - Cl mostly in form of reservoirs: HCl, ClONO<sub>2</sub> (HOCl, Cl<sub>2</sub>O<sub>2</sub>)
      - cf. HCl + OH  $\rightarrow$  ClO *slow*
    - F almost only in form of HF
      - cf. HF + OH  $\rightarrow \dots$  *too slow*
  - but: Cl-abundance ( $\cong 3.5$  ppb)  $>>$  Br-abundance ( $\leq 20$  ppt); I has not been detected yet (estimate:  $0.2 \pm 0.3$  ppt)  

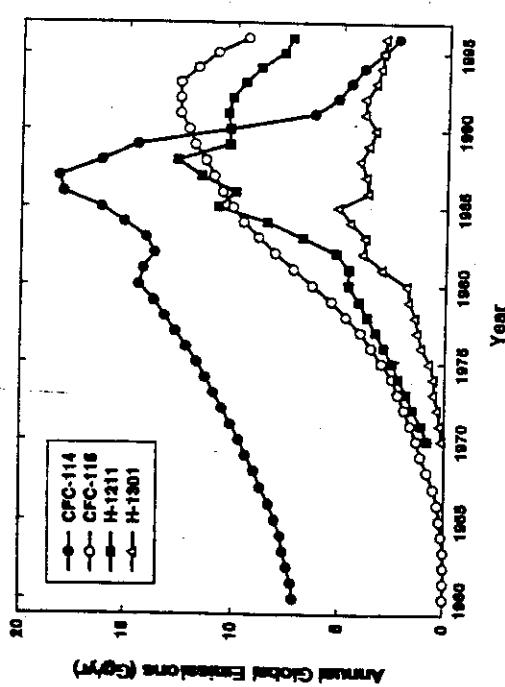
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Appl. Sci.: Chlorine in the stratosphere

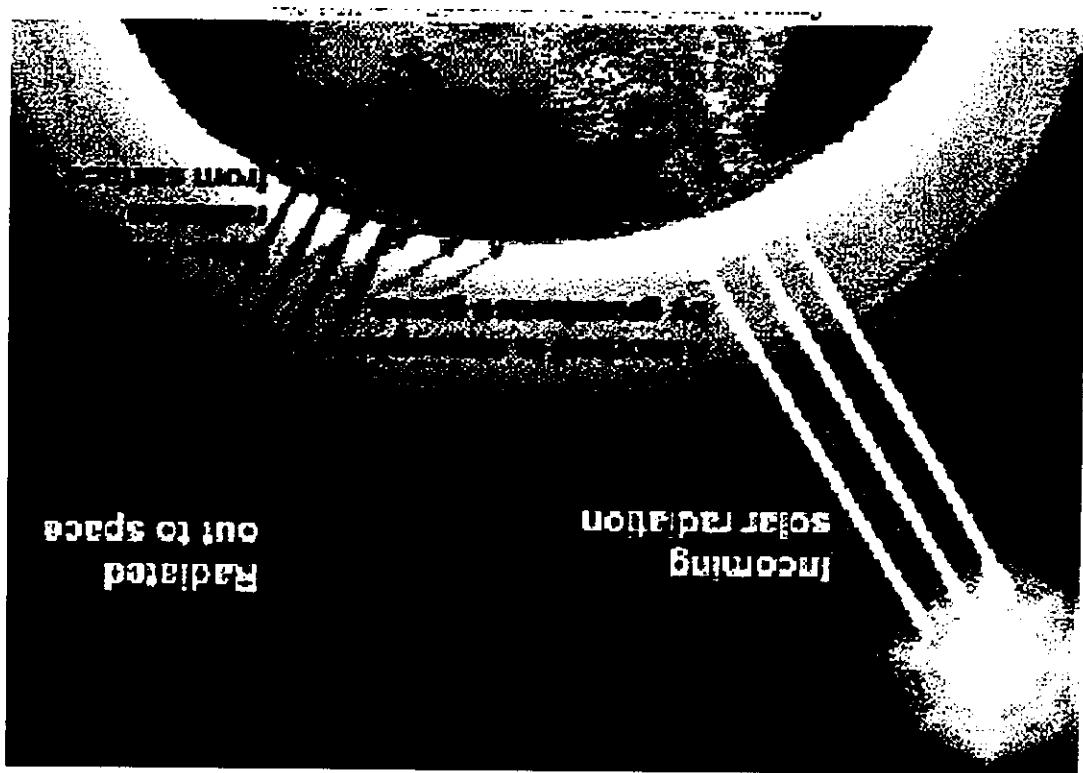




**Figure 2-1.** Annual global emissions of short-lived halocarbons ( $\text{kt yr}^{-1}$ ,  $\text{Gg yr}^{-1}$ ) estimated by industry from audited production, sales, and other data (AFEAS, 1998; Fisher *et al.*, 1994; Midgley and McCulloch, 1995; McCulloch and Midgley, 1996; and Midgley *et al.*, 1998). Note: There are no surveyed data for  $\text{CH}_2\text{Cl}_2$ ,  $\text{C}_2\text{Cl}_4$ , and  $\text{C}_2\text{HCl}_3$  prior to 1988.



**Figure 1-11.** Annual global emissions (in  $\text{Gg yr}^{-1}$ ) of long-lived halocarbons estimated by industry from audited production, sales, and other data (AFEAS, 1998; Fisher *et al.*, 1994; McCulloch, 1992; Fraser *et al.*, 1998; updated by P. Midgley, personal communication).



Atmospheric heating.

↑  
Greenhouse effect.

# Atmospheric long-term changes: 'Greenhouse' gases, climate change (1/5)

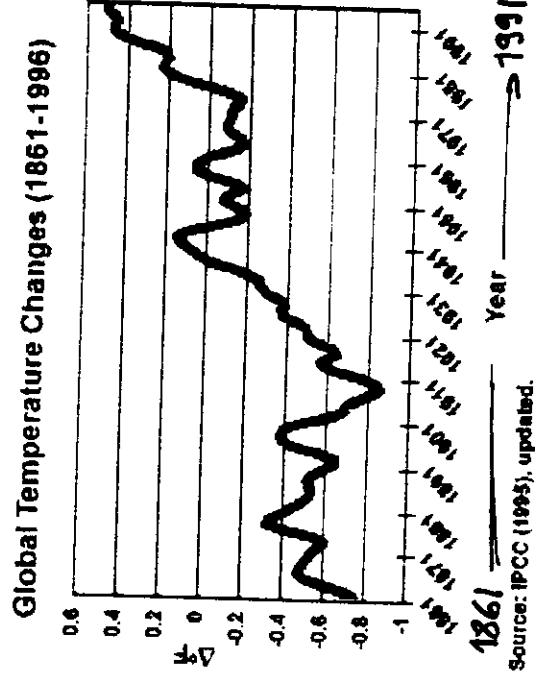
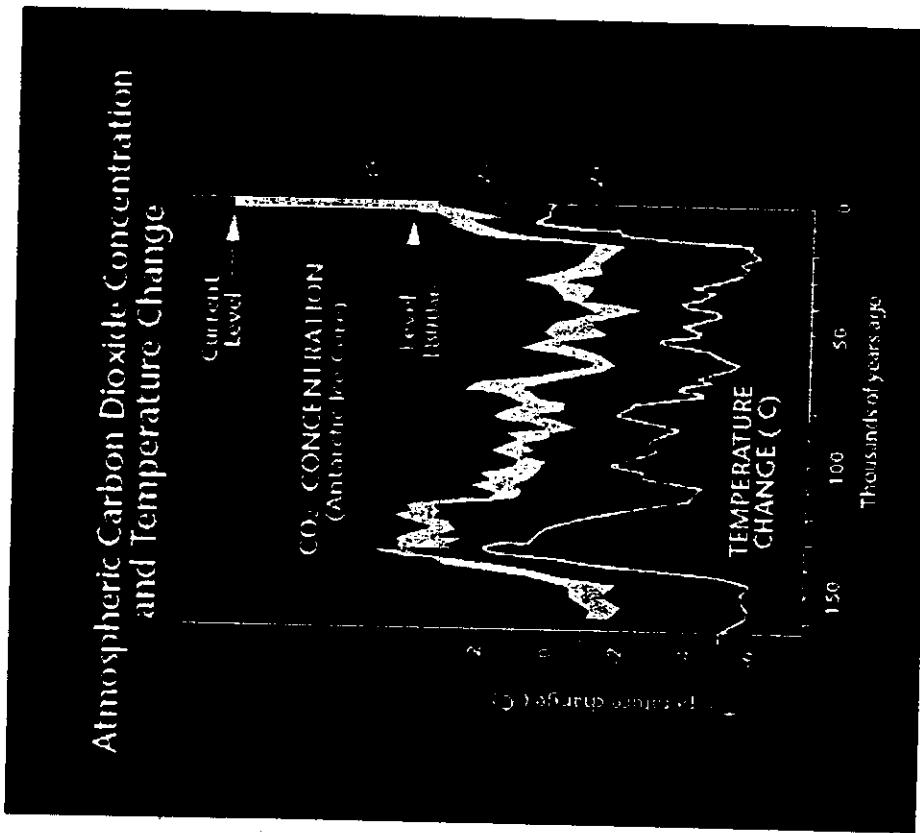


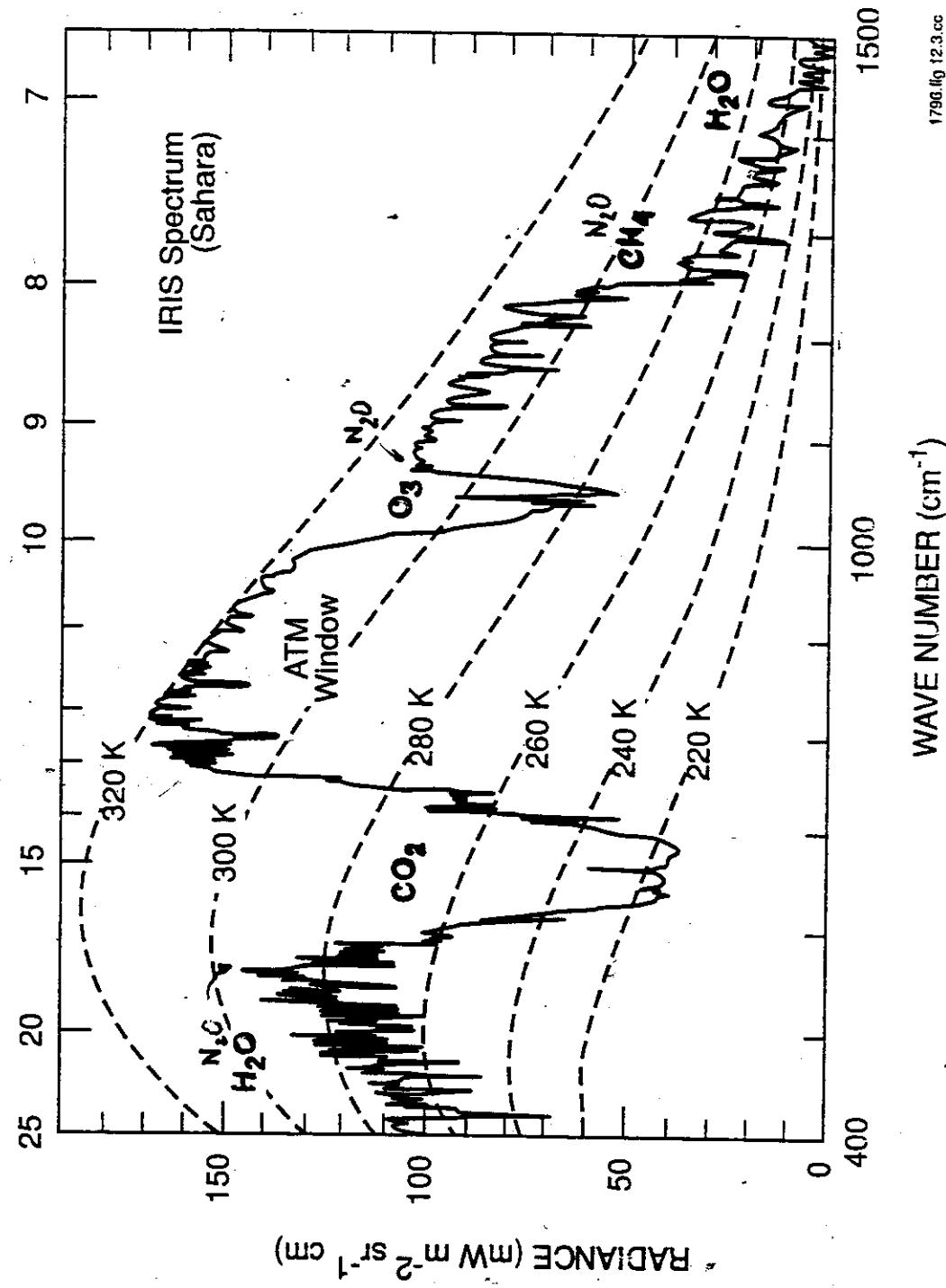
Figure 1: Trends in CO<sub>2</sub> Concentrations and Global Temperatures (per 100,000 years)  
PPMV = parts per million by volume

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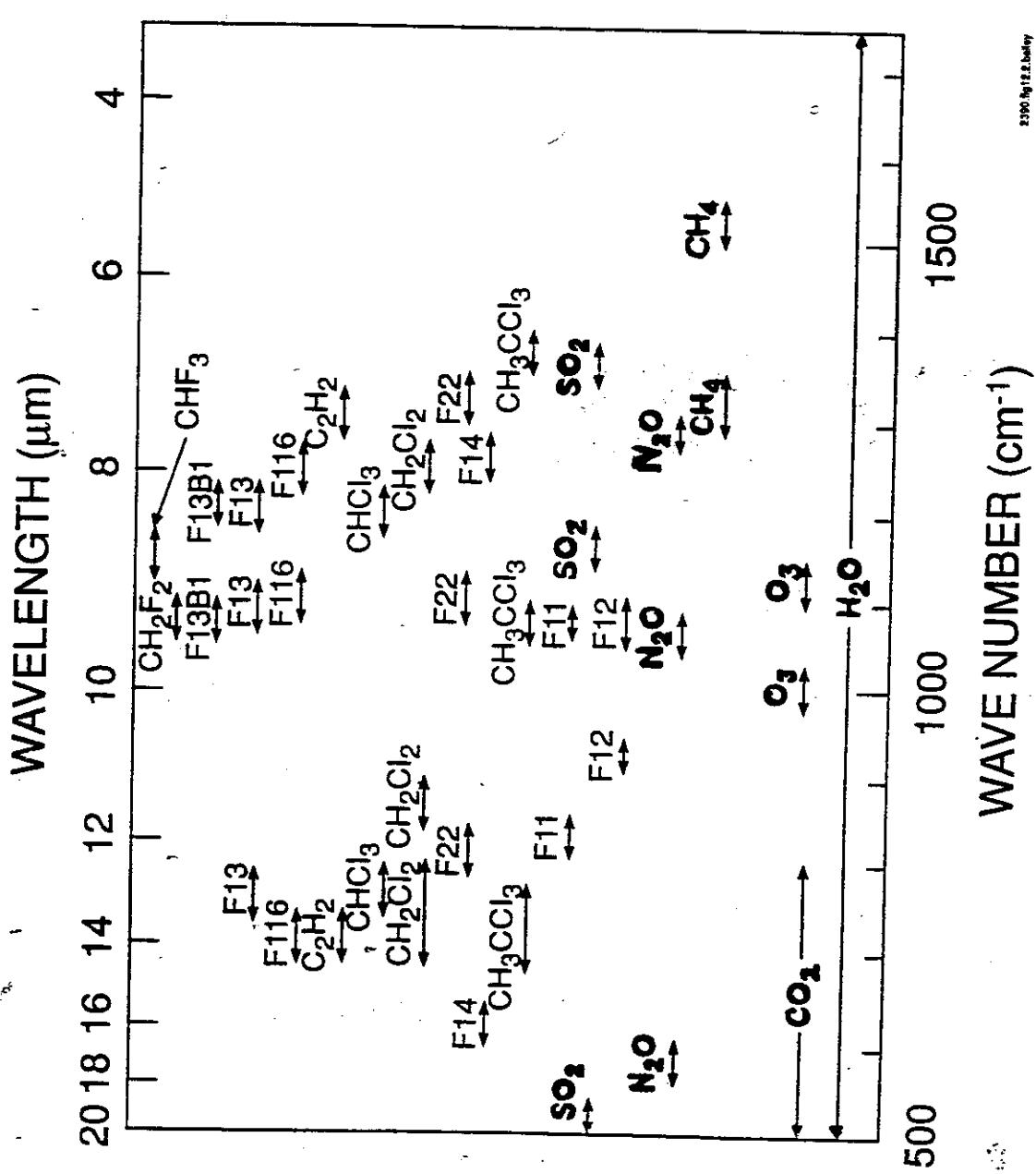
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**Figure 15.3.** Example of terrestrial radiation spectrum obtained by the Nimbus 3 IRIS instrument for clear sky conditions (from Hanel et al., 1972).

WAVELENGTH ( $\mu\text{m}$ )



**Figure 15.2.** Spectral locations of the absorption features of several atmospheric gases (WMO, 1986).

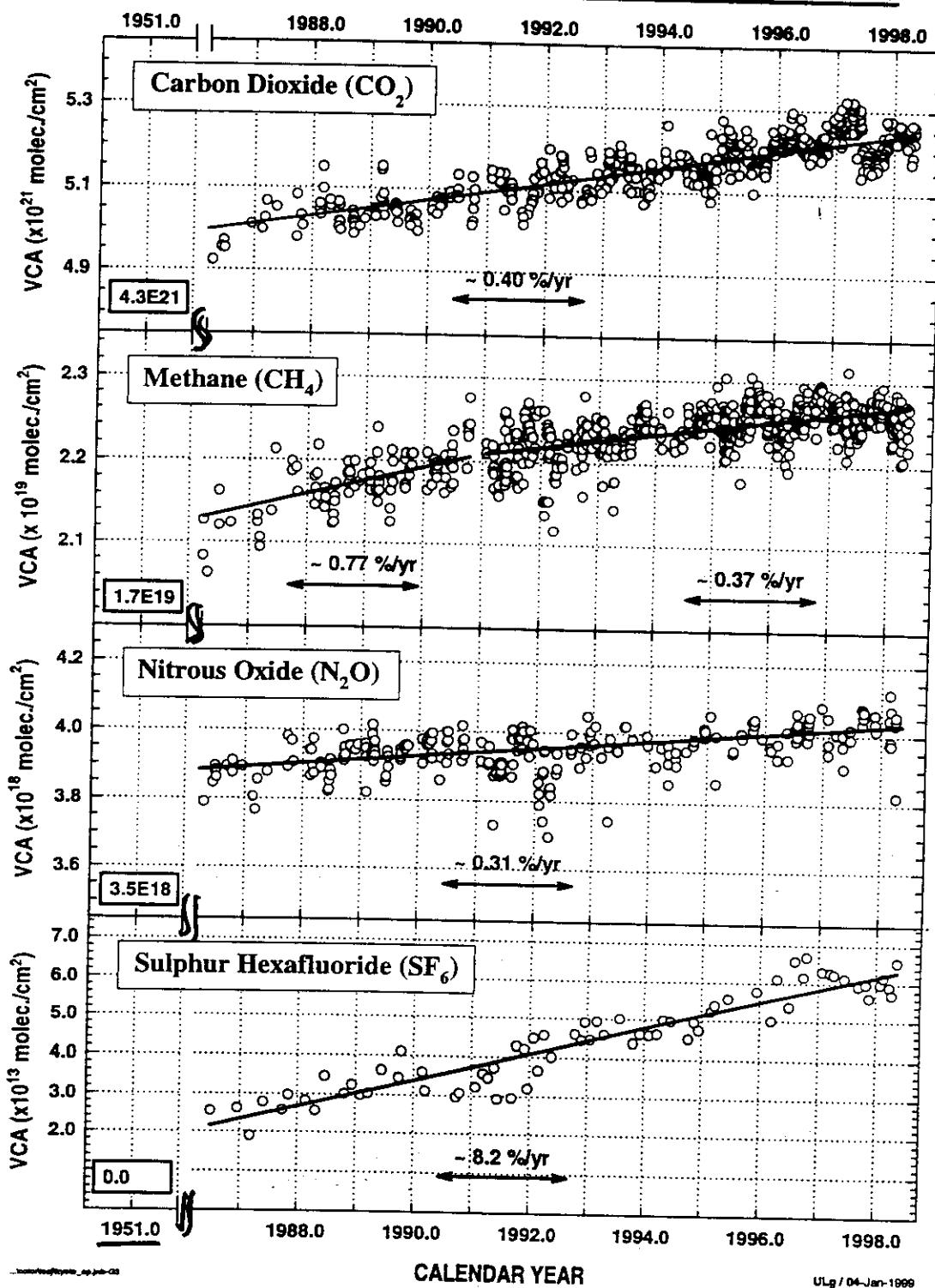


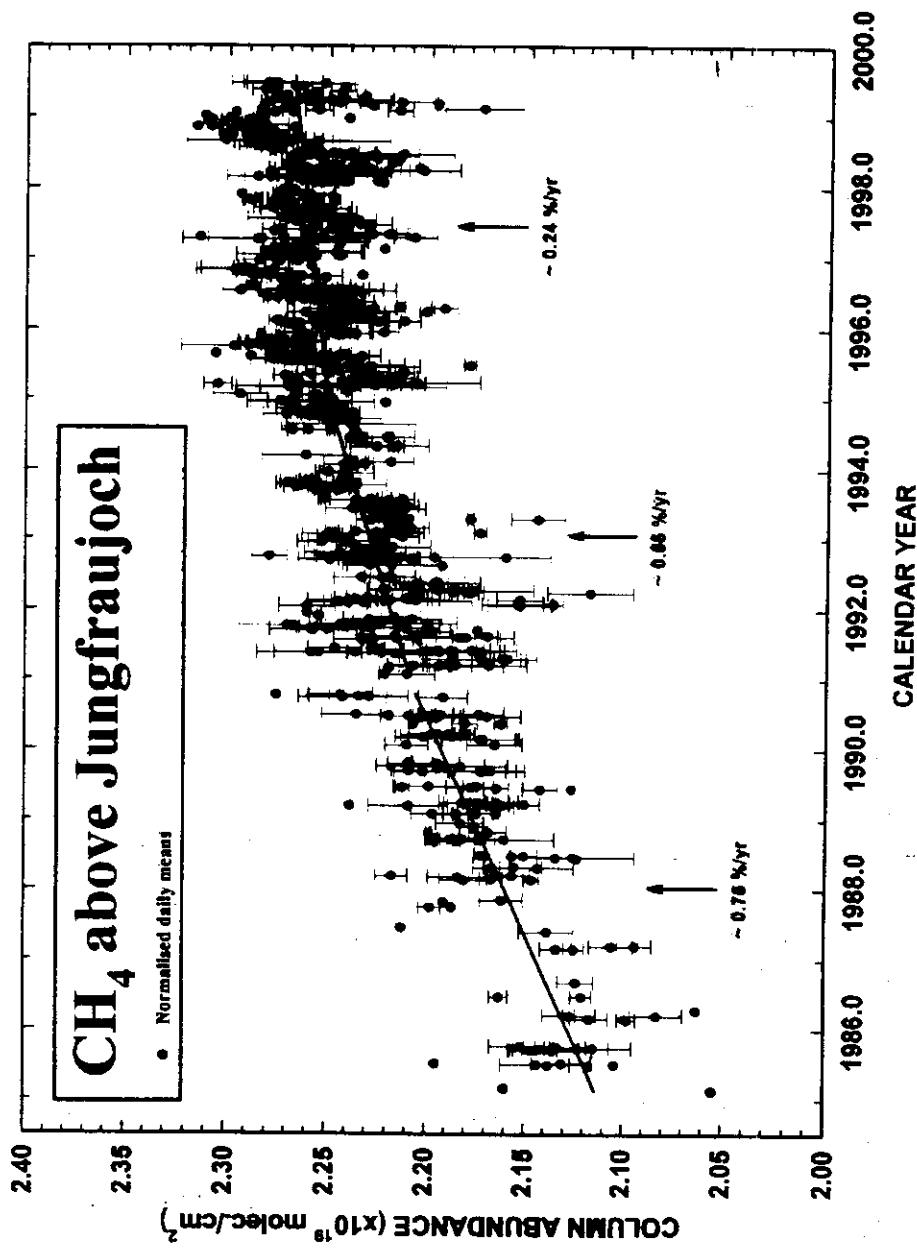
## **Atmospheric long-term changes: 'Greenhouse gases', climate change (2/5)**

- Direct contribution to greenhouse forcing (heat trapping in tropo-, UT/LS region):
  - H<sub>2</sub>O: largest forcing but little anthropogenic changes, *badly known!*
  - CO<sub>2</sub>: ± saturated, well-mixed in tropo, well-characterized
  - CH<sub>4</sub>: NRRF/mol≈ 25
  - N<sub>2</sub>O: NRRF/mol≈ 225, well-mixed in tropo
  - O<sub>3</sub>: tropo O<sub>3</sub> increase induces larger + forcing than - forcing from strato O<sub>3</sub> decrease
  - CFC, HCFC, HFC, ...: NRRF/mol ≈ 10000 - 20000
- Approximate contribution to enhanced heat trapping:  
60% CO<sub>2</sub>, 20% halocarbons, 15% CH<sub>4</sub>, 5% N<sub>2</sub>O  
(since mid-1800; IPCC, 1995)

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## Kyoto Protocol-related measurements above ISSJ

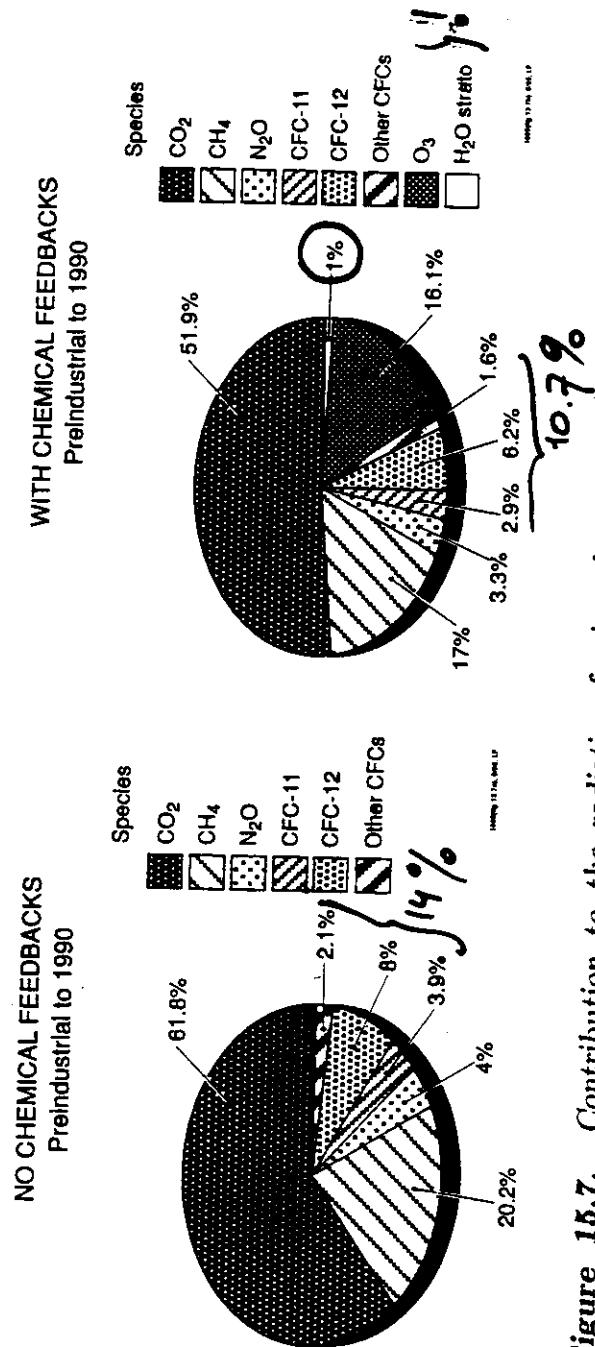




# Atmospheric long-term changes: 'Greenhouse gases', climate change (3/5)

- SF<sub>6</sub>: actually very small abundance ( $\approx 1$  ppt), but dramatic increase (8%/yr), very long lifetime (> 3000 yrs), NRRF/mol  $\approx 25000$
  - clouds ? Positive or negative forcing ?
  - negative radiative forcing impact on surface climate caused by *stratospheric O<sub>3</sub>* decrease & volcanic aerosol increases are smaller than positive forcing caused by increasing GHG (IPCC, 1996)
- Indirect forcing: through coupling with chemical effects
- Radiative cooling effect in the stratosphere ! But: *Stratospheric T changes are mainly due to strato-O3 changes.*

» LARGE UNCERTAINTIES, esp. as to H<sub>2</sub>O !



**Figure 15.7.** Contribution to the radiative forcing due to increases in greenhouse gas concentrations for the period 1900-1990. Note that these values, especially those including chemical feedbacks, are rather uncertain because of the strong nonlinearities in the coupled chemical and climate systems.

# Atmospheric long-term changes: ‘Greenhouse gases’, climate change (4/5)

## ☒ Climate Index (Hansen et al., 1998)

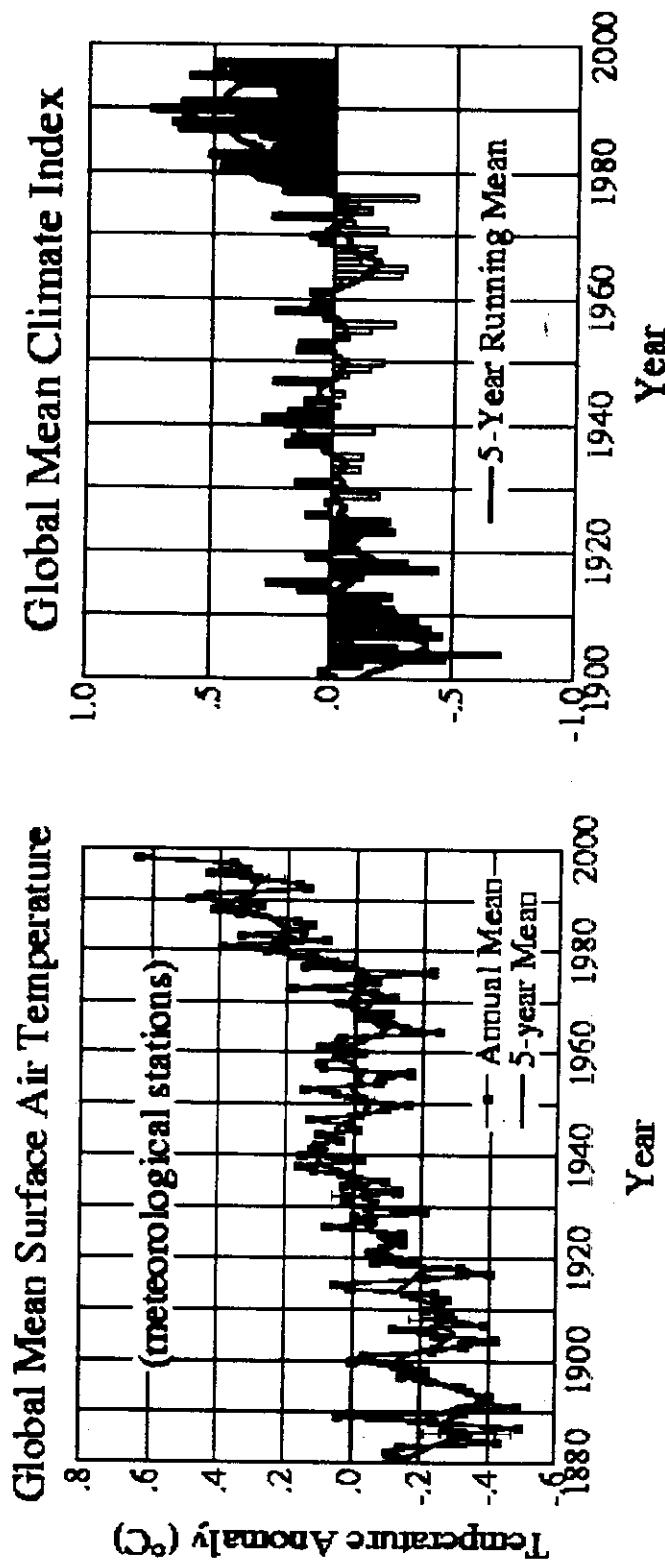
= mean of several climate change indicators, that tend to be noticed by people and have economic significance

➤ climate change indicators:

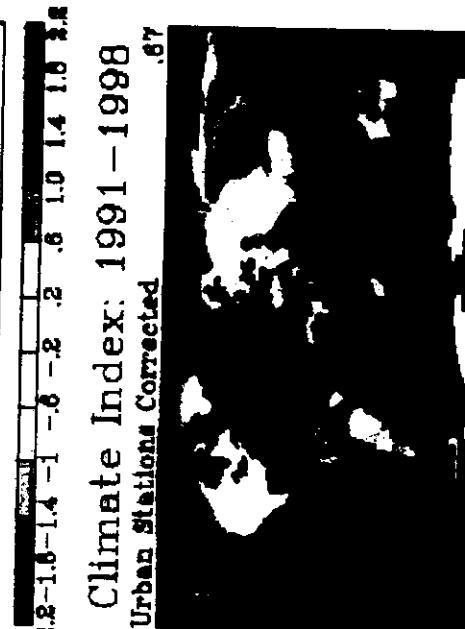
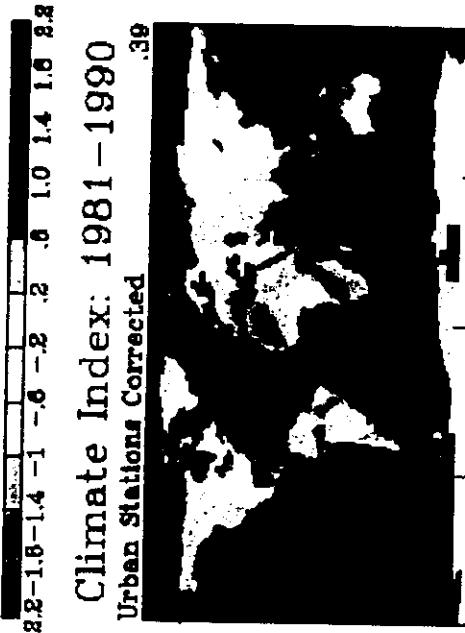
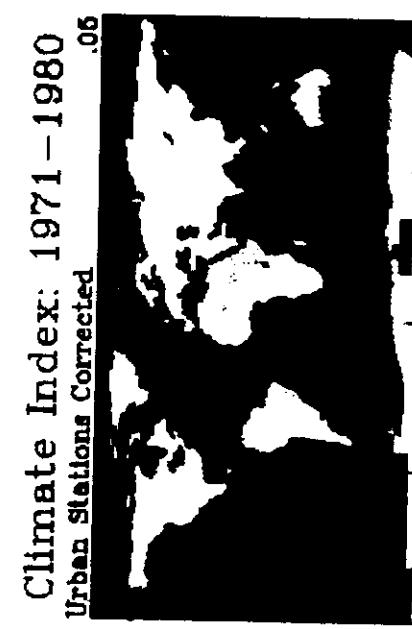
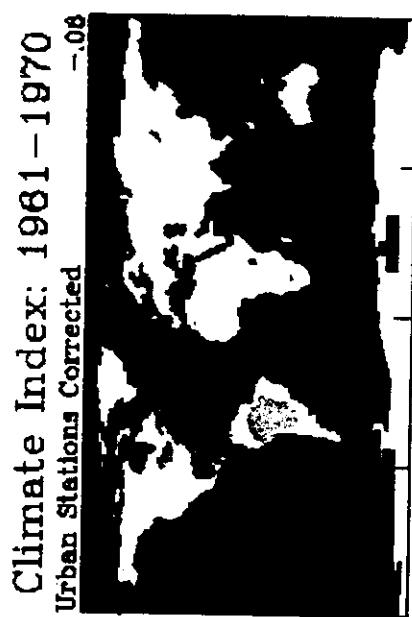
- T indicators
  - seasonal mean temperatures (4 seasons)
  - degree days (heating season, cooling season)
  - frequency of extreme temperatures (hot summer days, cold winter days)
  - record daily temperatures (records high, records low)
- moisture indicators
  - precipitation frequency,....

Scale:  $\pm 1 \Leftrightarrow \pm 1 \sigma$  (interannual standard deviation) during 1951-1980

# Atmospheric long-term changes: 'Greenhouse gases', climate change (5/5)



# Atmospheric long-term changes: 'Greenhouse gases', climate change (5/5)



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<u>Year</u>	<u>Policy Process</u>	<u>Scientific Assessment</u>
1981		<i>The Stratosphere 1981. Theory and Measurements.</i> WMO No. 11
1985	Vienna Convention	<i>Atmospheric Ozone 1985.</i> WMO No.16.
1987	Montreal Protocol	
1988		<i>International Ozone Trends Panel Report 1988.</i> Two volumes. MO No. 18
1989		<i>Scientific Assessment of Stratospheric Ozone: 1989.</i> Two volumes. WMO No. 20.
1990	London Adjustments and Amendment	
1990		<i>Climate Change, The IPCC first Scientific Assessment, Impacts Assessment and Response Strategies Reports</i>
1991		<i>Scientific Assessment of Ozone Depletion: 1991.</i> WMO No. 25.

1992

*Methyl Bromide: Its Atmospheric Science, Technology, and Economics (Assessment Supplement).* UNEP (1992).

1992 Copenhagen Adjustments and Amendment

1992 Rio de Janeiro Convention on Climate Change; Kyoto Protocol on Climate Change  
*IPCC Supplementary Report to the Scientific Assessment*

1994

- *Scientific Assessment of Ozone Depletion: 1994.* WMO No. 37  
- *Climate Change 1994. Radiative Forcing of Climate Change and an Evaluation of the IPCC IS92 Emission Scenarios*

1995 Vienna Adjustment

1995 *Climate Change 1995, The IPCC Second Scientific Assessment, Impacts Assessment .... Reports*

1997 - Montreal Adjustments and Amendment

• Kyoto Protocol (UNFCCC third session, Kyoto, Dec. 1997)

1998

*Scientific Assessment of Ozone Depletion: 1998.* WMO. No. 44

1999 11<sup>th</sup> Meeting of the Parties (China)

2000 *The IPCC Third Scientific Assessment, Impacts Assessment .... Reports*

## Total Chlorine loading of the mid latitude stratosphere

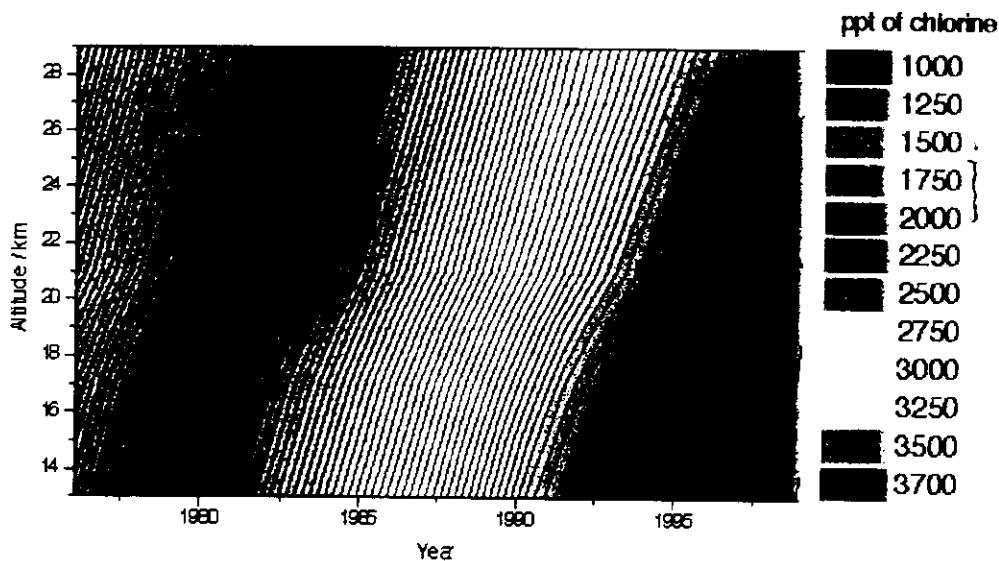


Figure 4. Reconstructed trend of  $Cl_{\text{total}}$ , derived from the tropospheric trend of the mixing ratios of the 7 most important chlorine source gases (see text for detail). The calculation of the vertical propagation is based on the age profile determined from  $SF_6$  measurements

## Predicted Total Chlorine loading of the mid latitude stratosphere

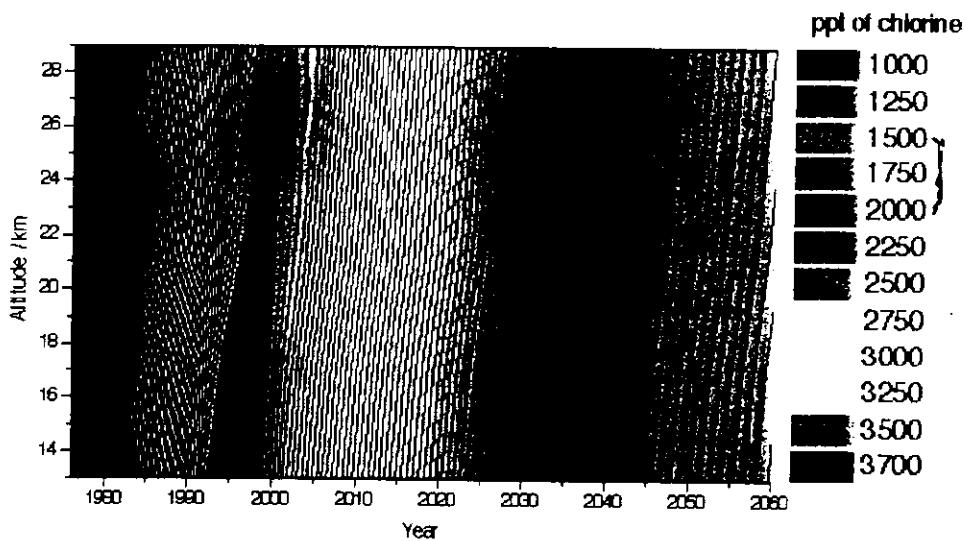


Figure 5. Same as Figure 4 but extended into the future based on the lifetimes of the chlorine compounds and assumptions about future emissions (see text). Pre-ozone hole values are expected to be reached again in about 50 years.

## **Atmospheric long-term changes: Ozone (1/2)**

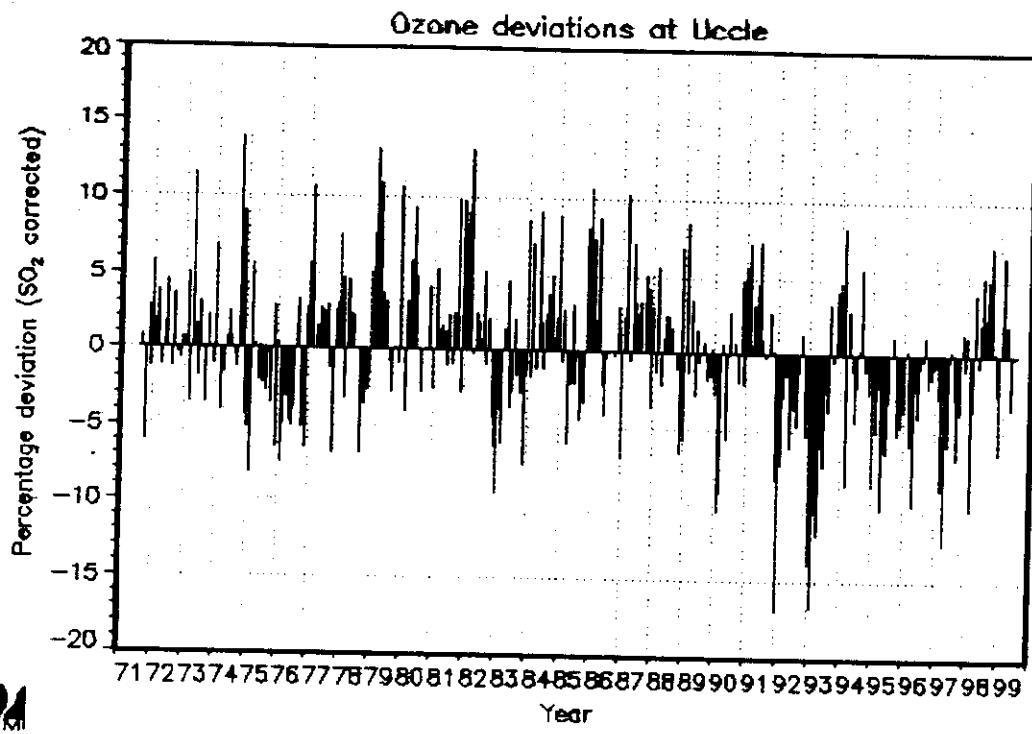
- ☒ The Montreal Protocol is working  
+ *Amendments*!  
but:
- ☒ The Ozone layer actually is in its most vulnerable state
  - we are approx. at the peak of chlorine (ODS) loading
  - the Antarctic O<sub>3</sub> hole is still at its highest level
  - Arctic O<sub>3</sub> loss has been observed in most years since 1990
  - mid-latitude O<sub>3</sub> changes are not as large as projected by early 1990
  - if any volcanic eruption ...?

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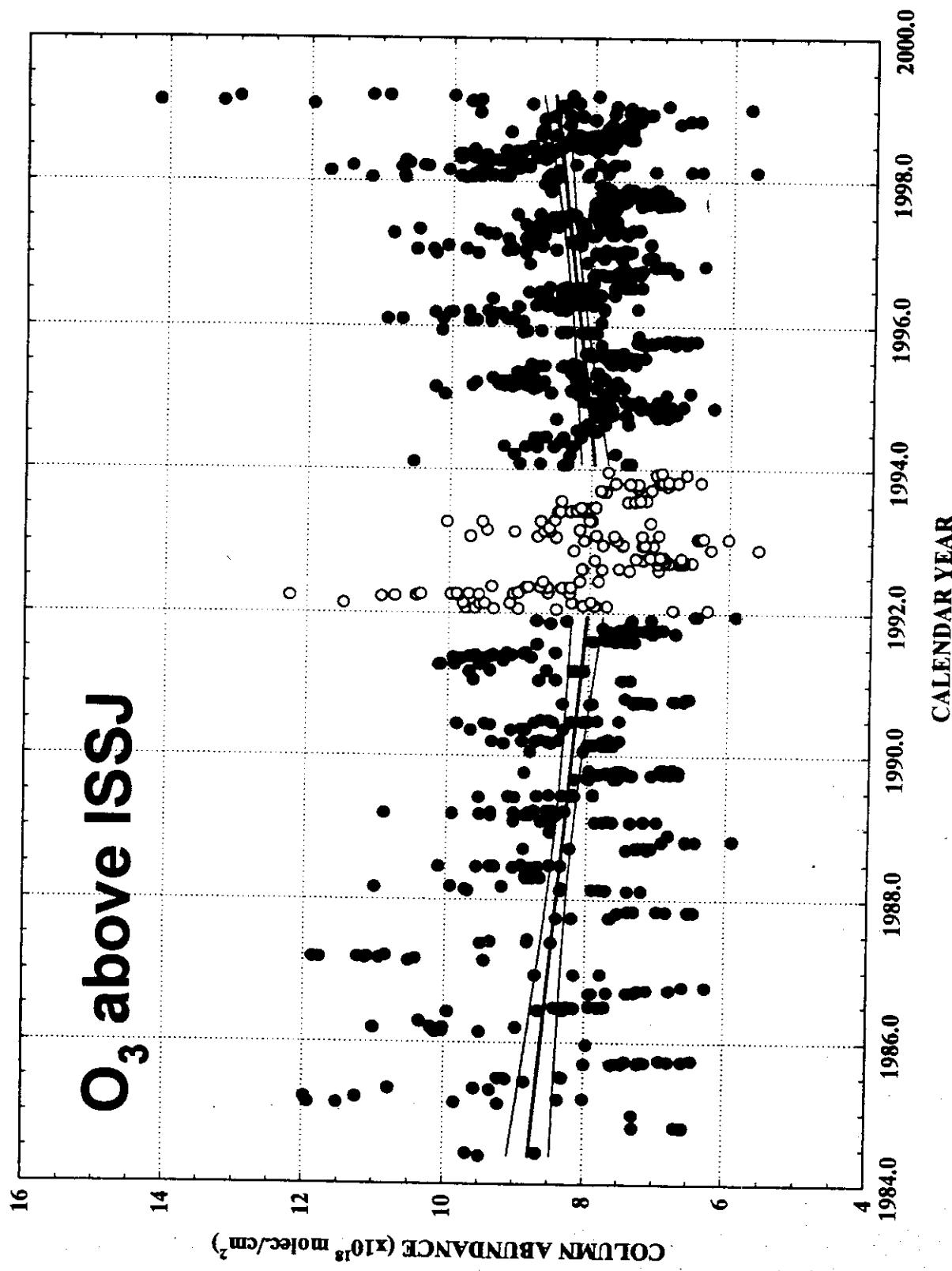
## Atmospheric long-term changes: Ozone (2/2)

- Current O<sub>3</sub> losses, since 1970,
  - about 6% at N midlatitudes in winter/spring
  - about 3% at N midlatitudes in summer/fall
  - about 5% at S midlatitudes all year round
  - about 50% in Antarctic spring
  - about 15% in the Arctic spring
    - *none in the tropics*
  - close to maximum !
- ☒ recovery is expected during next decades
  - return to pre-industrial levels of chlorine (2 ppbv) are expected by 2050
  - but O<sub>3</sub> recovery depends also on other changes: GHG, T, dynamics, etc...

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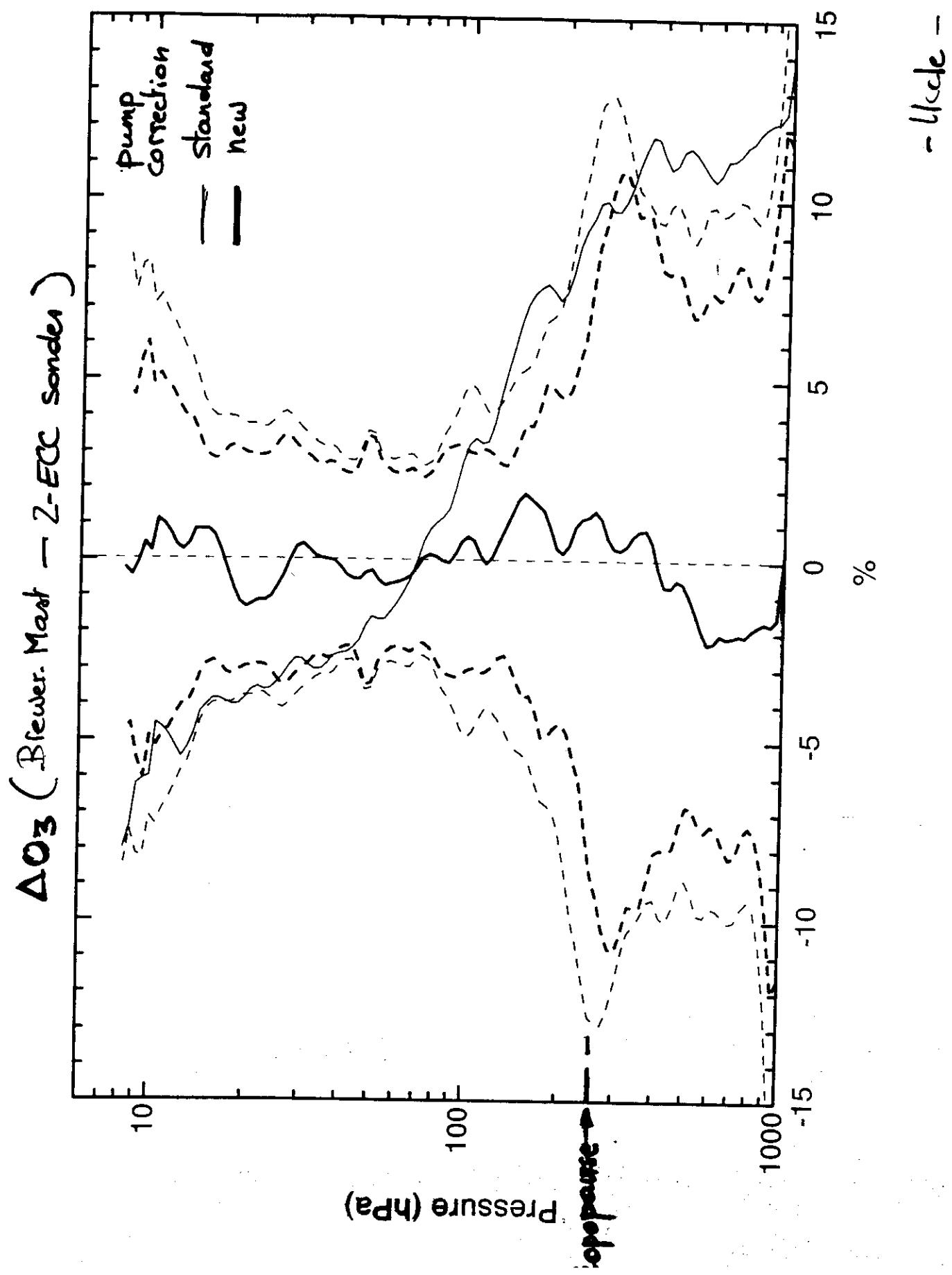


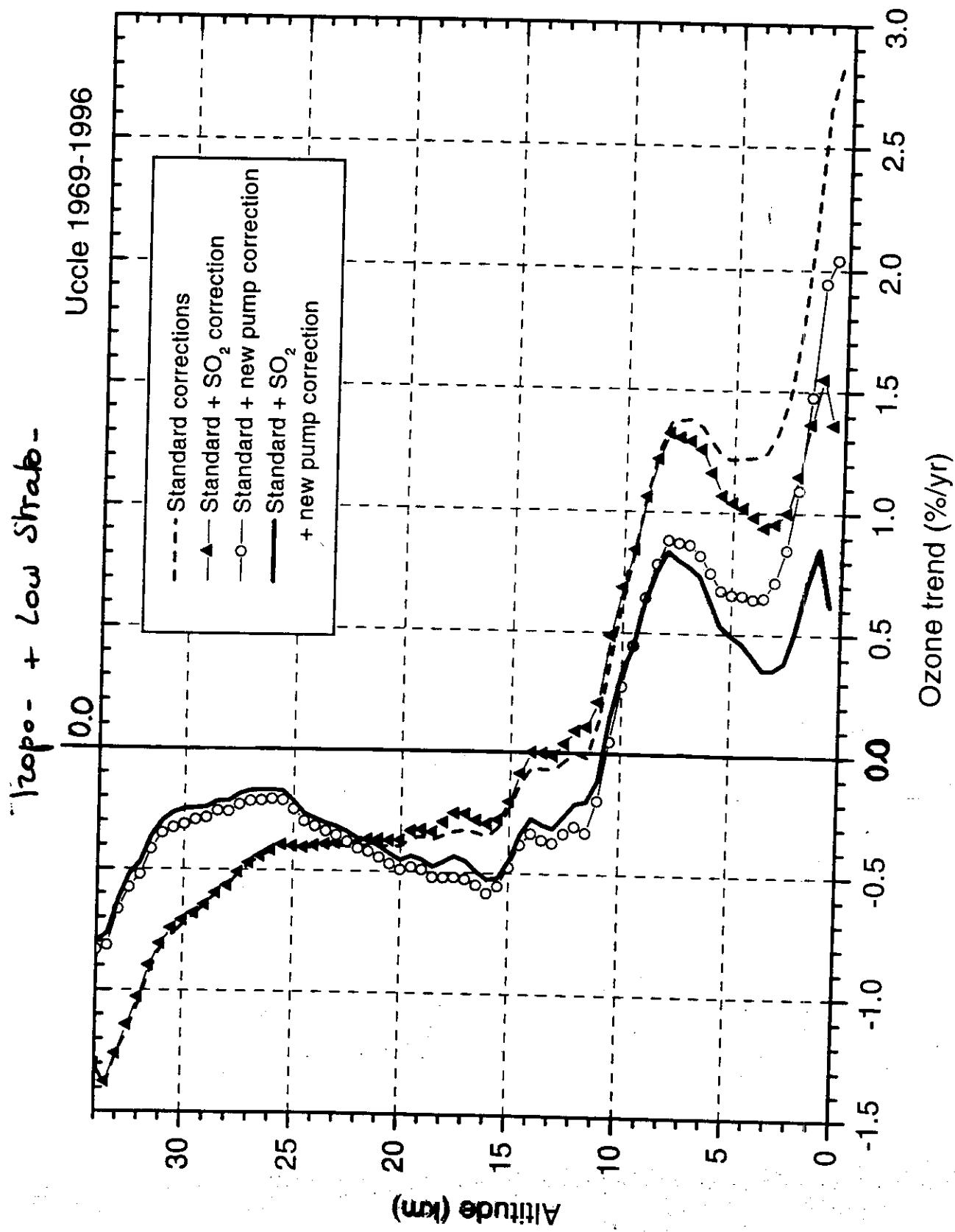
monthly means ; relative to long-term means

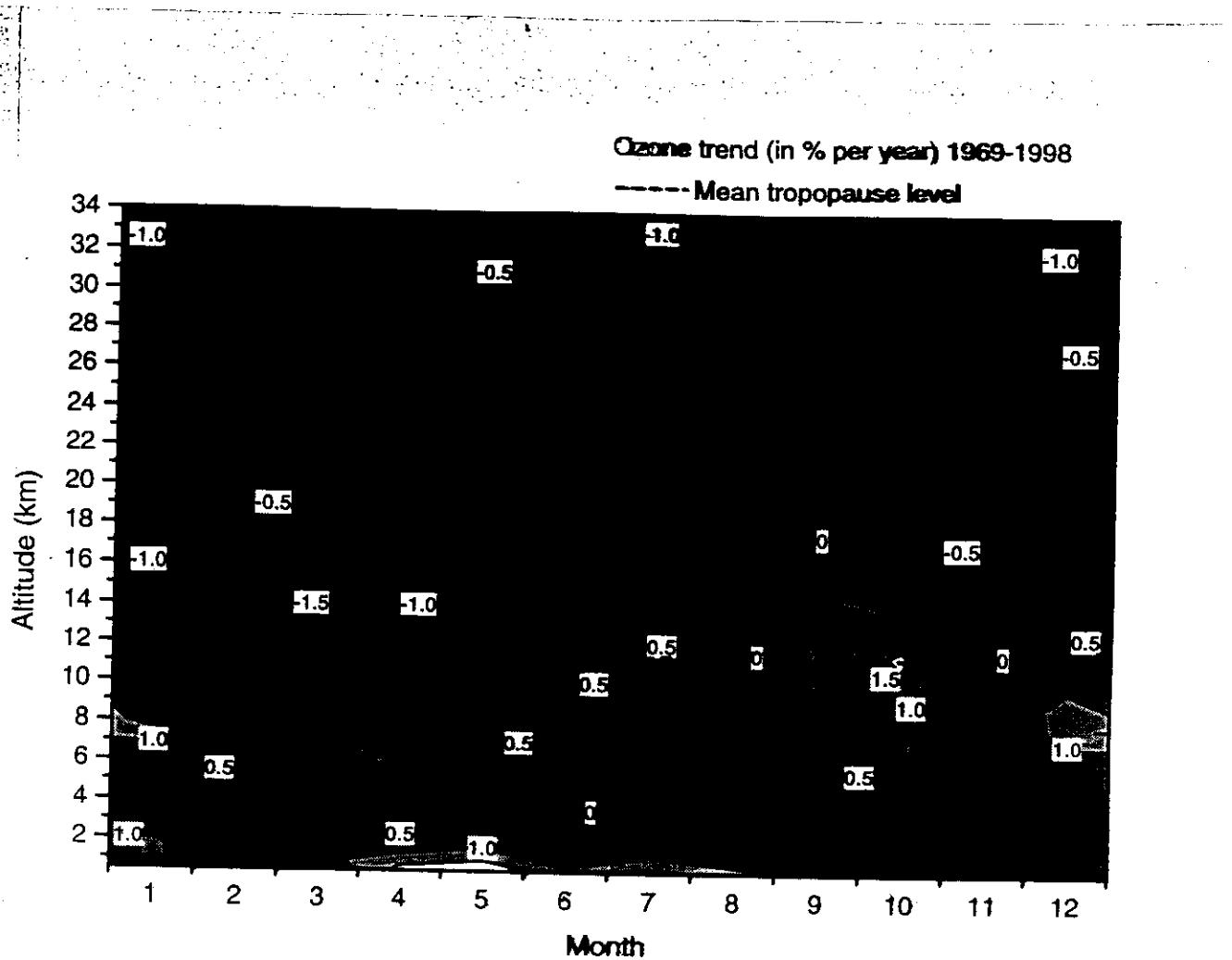


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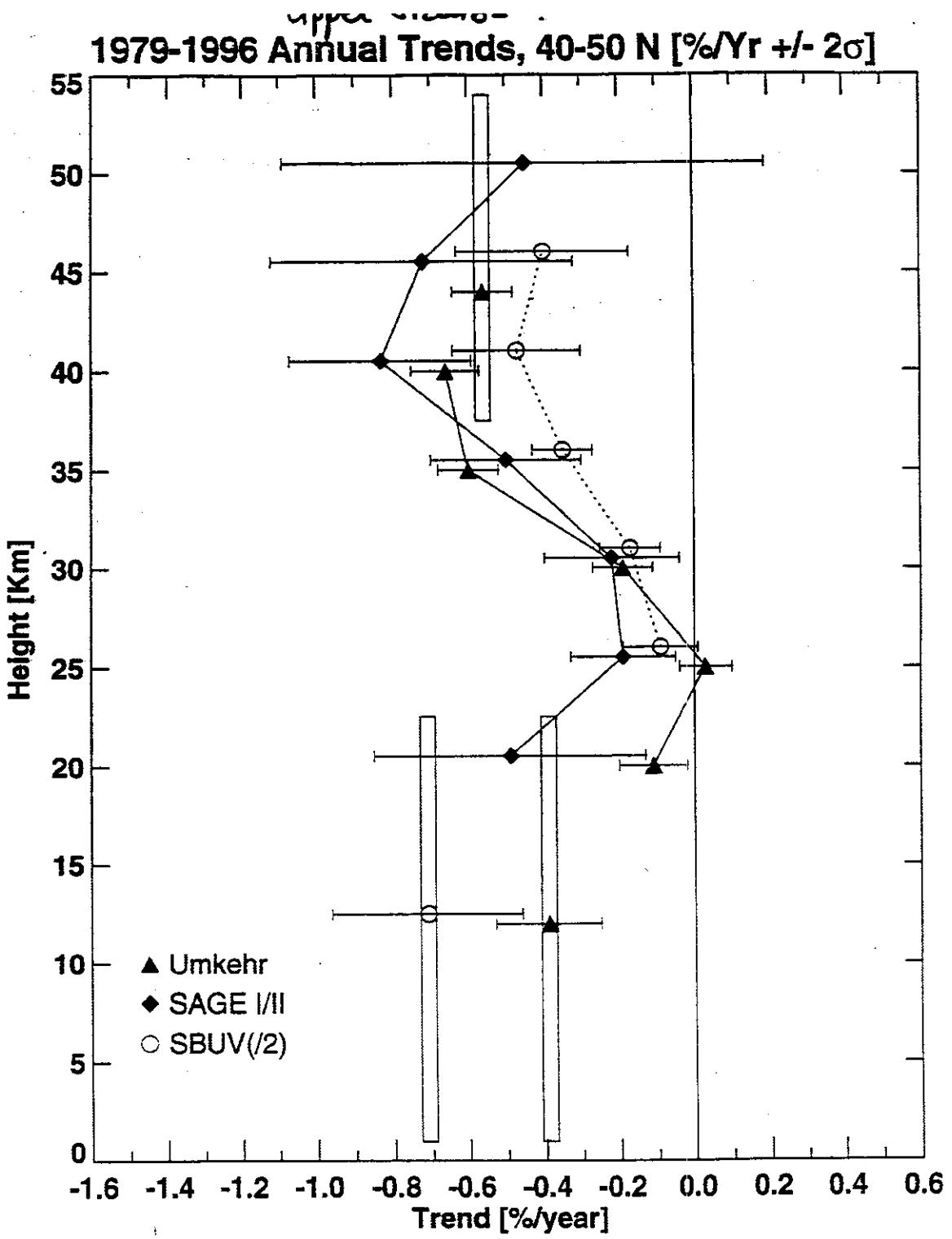
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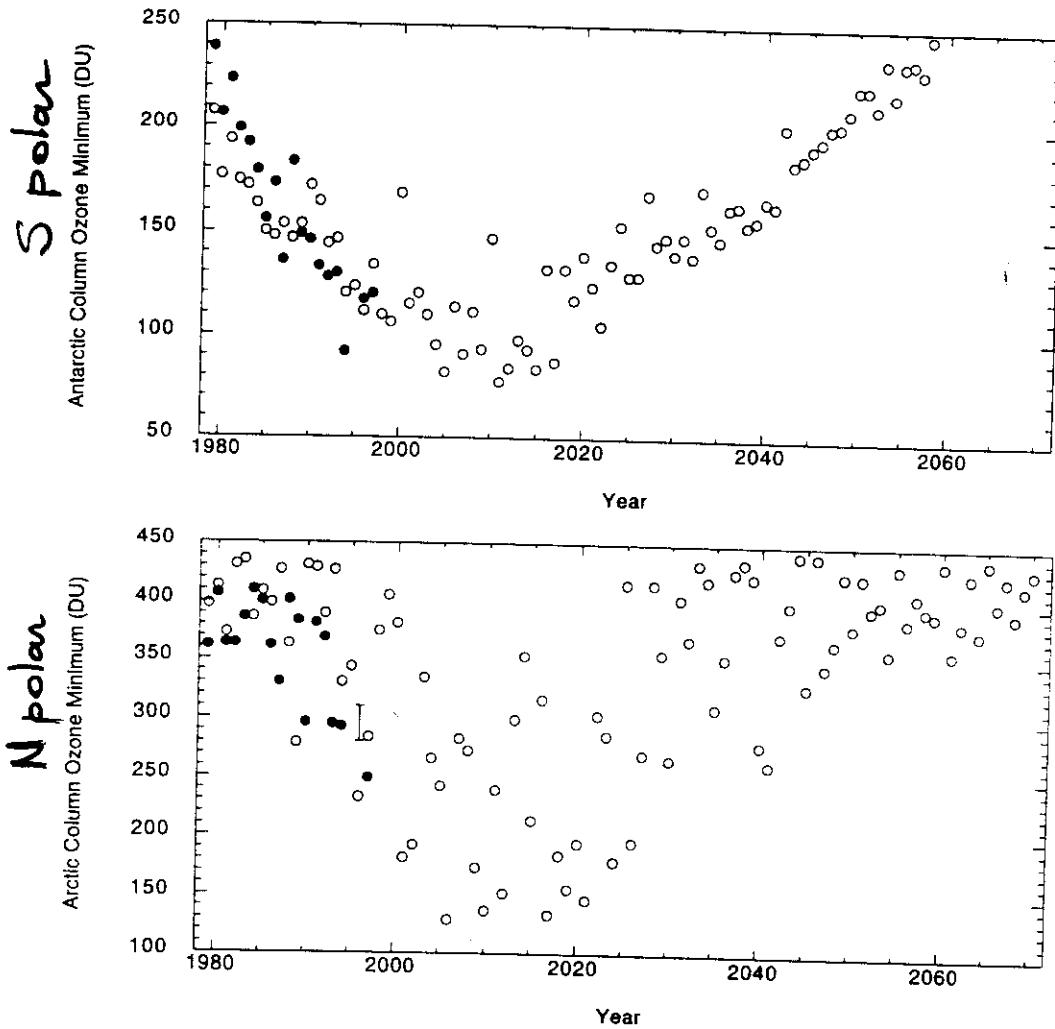
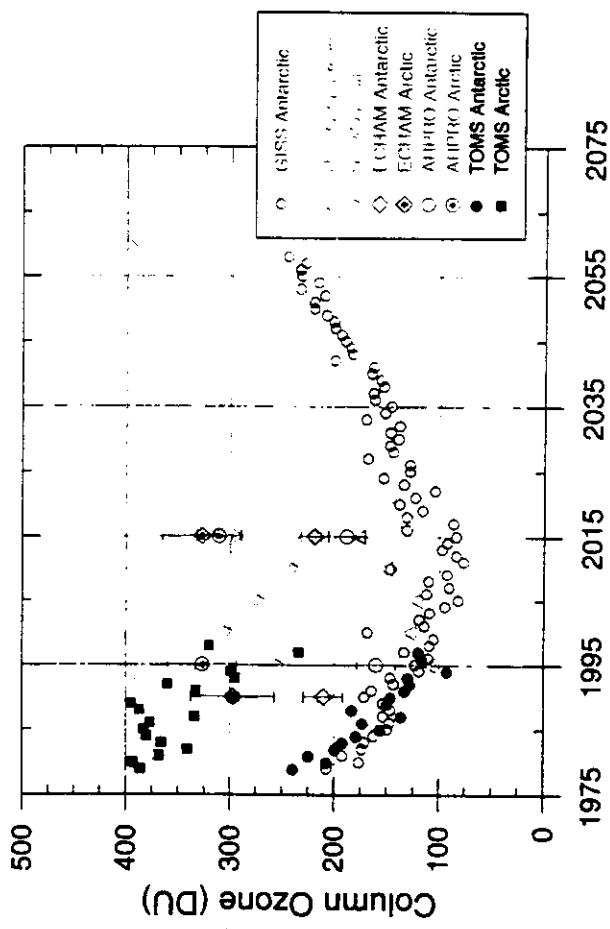


Figure 4



# **Ground-based measurements of stratospheric trace gases in the perspective of climate and environment**

## **Acknowledgements**

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(A)

