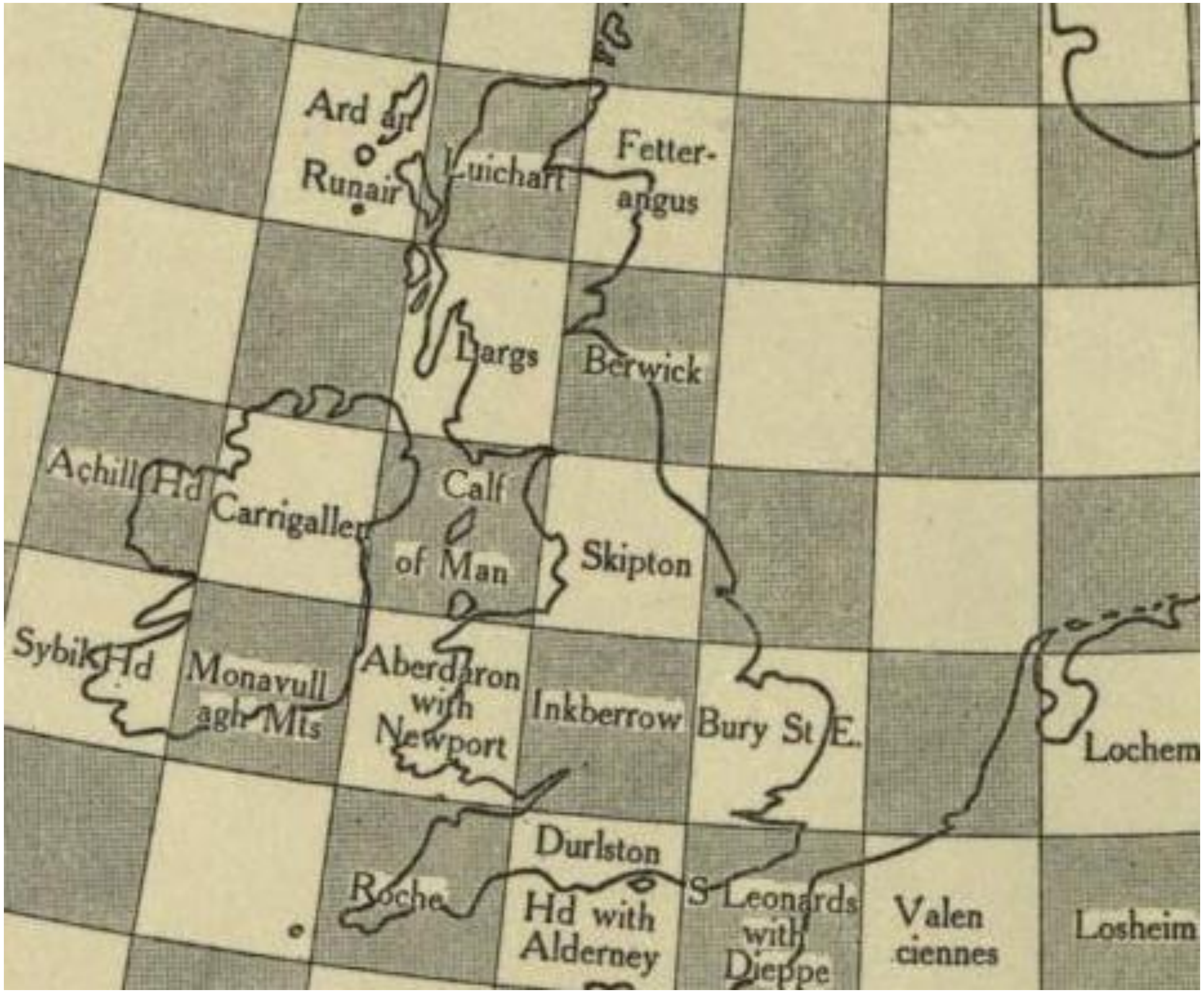
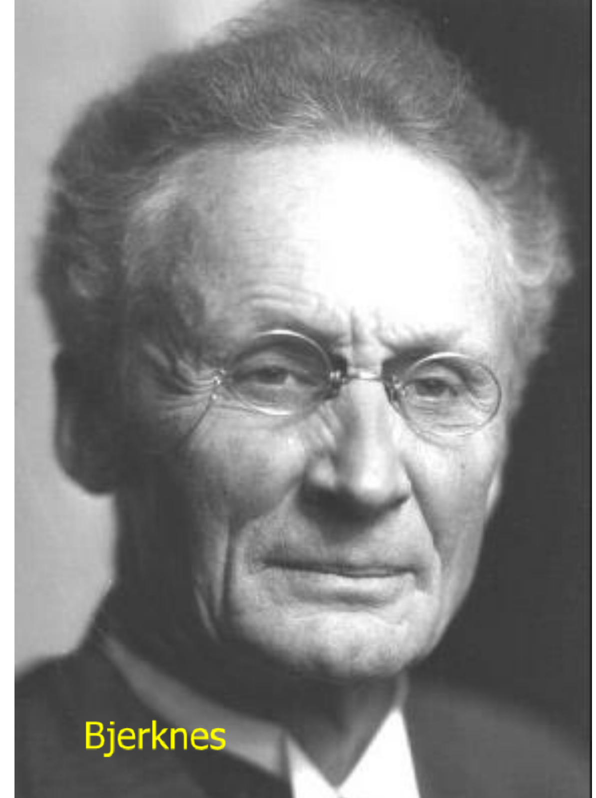
From Meteorology to Aeronomy and to Atmospheric Chemistry

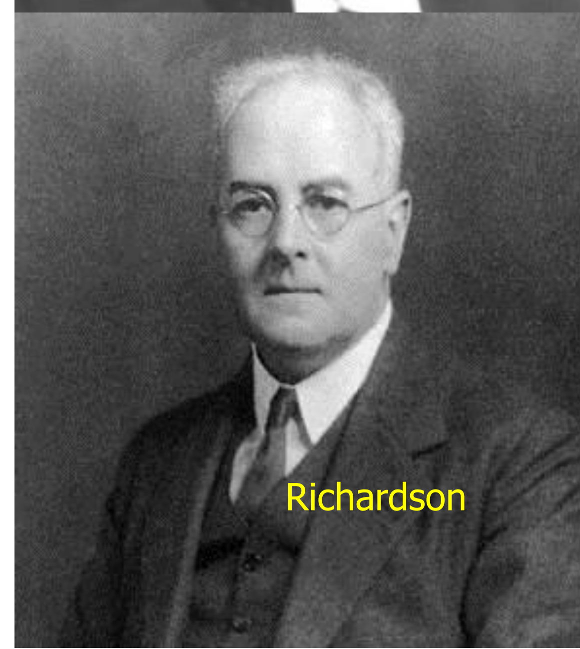
Guy P. Brasseur
Climate Service Center, HZG, Hamburg, Germany
and
National Center for Atmospheric Research
Boulder, CO, USA

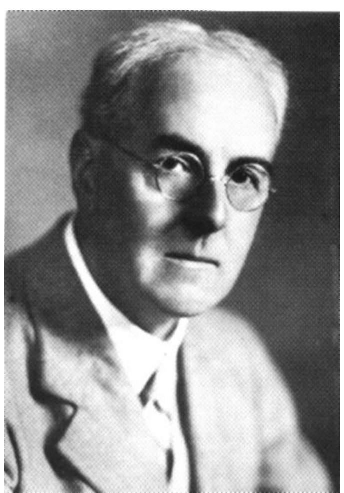
100 years ago….



failure.

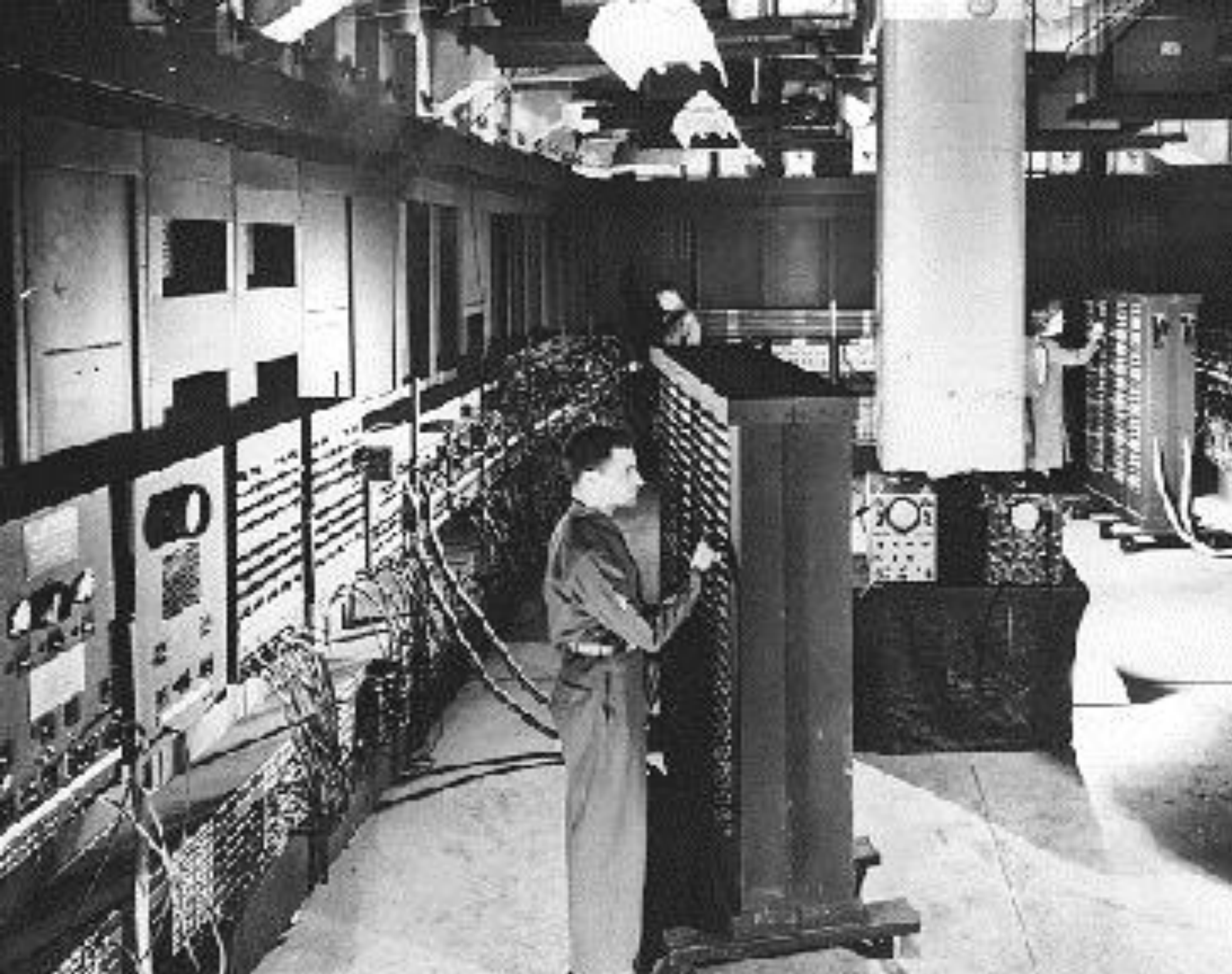






Before the Computer Age

In 1922, Lewis Fry Richardson suggests to assemble 64000 persons in a “numerical factory” to perform in parallel the tremendous number numerical operations required to solve the equations of meteorology. He had proposed the concept of parallel computing before the computer age.



With the ENIAC computer in the US, Jule Charney et John von Neuman successfully perform the first numerical weather forecast in 1950.

Ozone in the Atmosphere

- In 1840 Christian Friedrich **Schoenbein** at the University of Basel had discovered the presence of a a specific odor while performing the electrolyse of water. He called this property "ozon" (in German) from the Greek word "ozein".
- The presence of ozone in the atmosphere was reported in 1858 by Auguste **Houzeau** in Rouen, France.

Systematic observations of Ozone

- In the 1920’s the British scientist, **Gordon.M.B. Dobson** (Oxford University) develop a spectrograph and later a spectrophotometer that for many years remained the only accurate method to measure the ozone column abundance.
- These instruments were installed at different locations, which led Dobson to estimate the latitudinal and seasonal evolution of the ozone column.
- Dobson also discovered a strong influence of atmospheric dynamics on ozone.

The Dobson Ozone Photographic Spectrometer

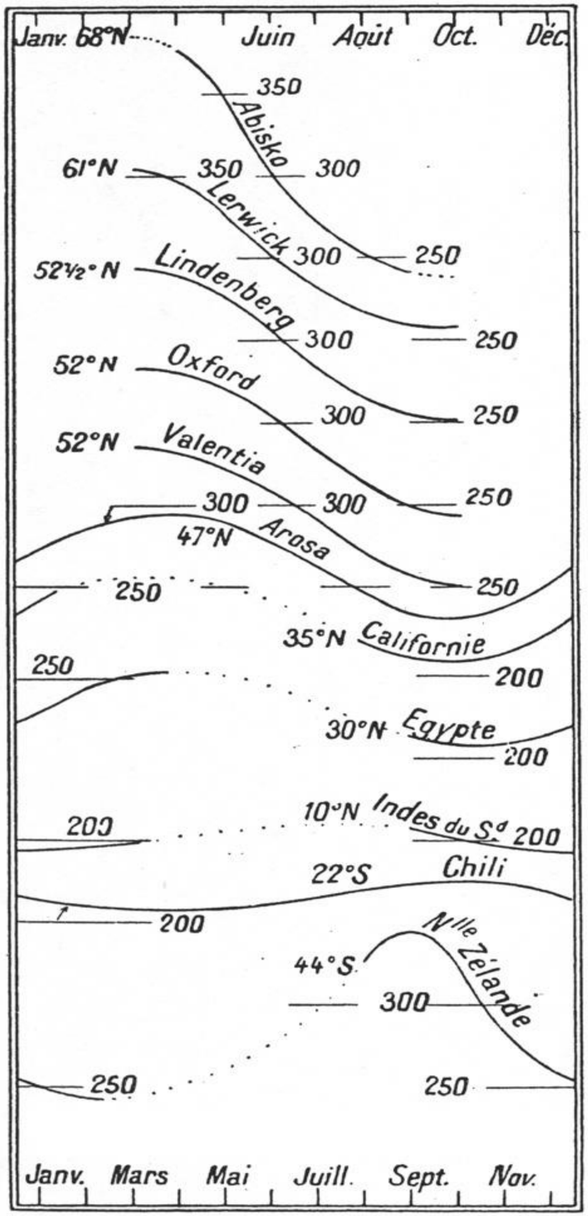
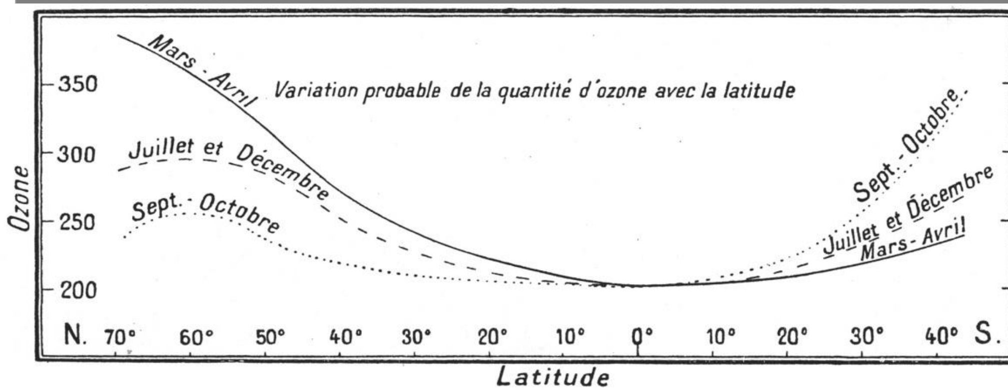
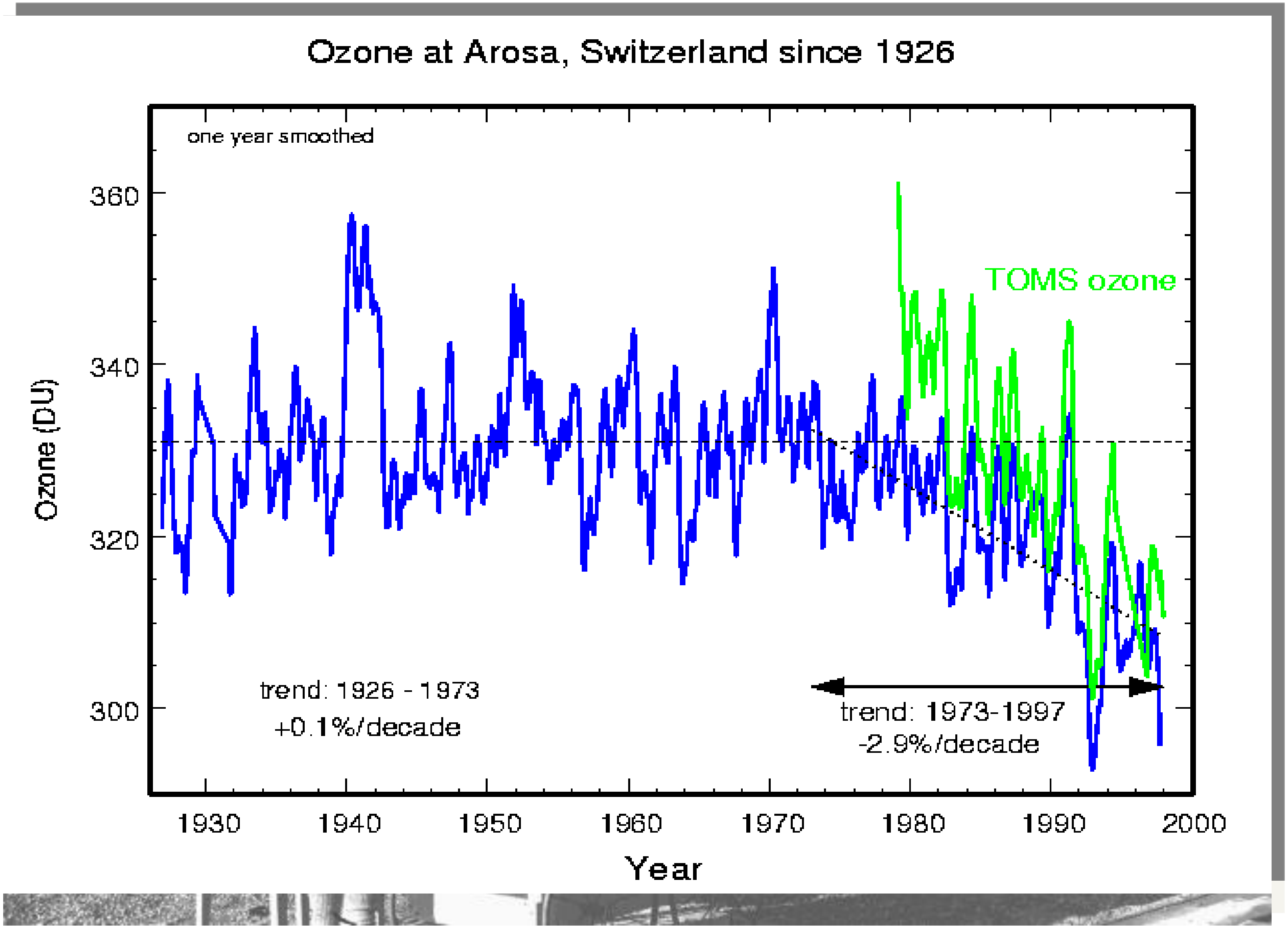


Fig. 3.



Ozone Observations at Arosa



The First Photochemical Theory (1929)

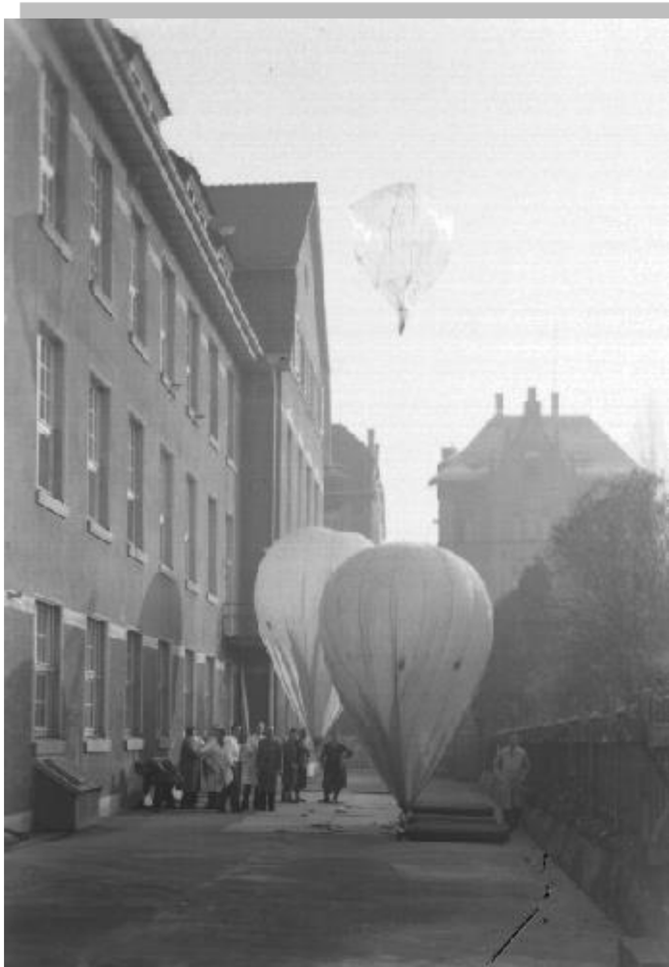
S. Chapman

- **Sydney Chapman** introduces the first scheme that describes the photochemistry of ozone by considering only 5 reactions:

$$O_2 + UV \rightarrow O + O$$
$$O + O + M \rightarrow O_2 + M$$
$$O + O_2 + M \rightarrow O_3 + M$$
$$O_3 + UV \rightarrow O + O_2$$
$$O + O_3 \rightarrow 2\,O_2$$

- He highlights the role of atomic oxygen

Height of the Ozone Layer

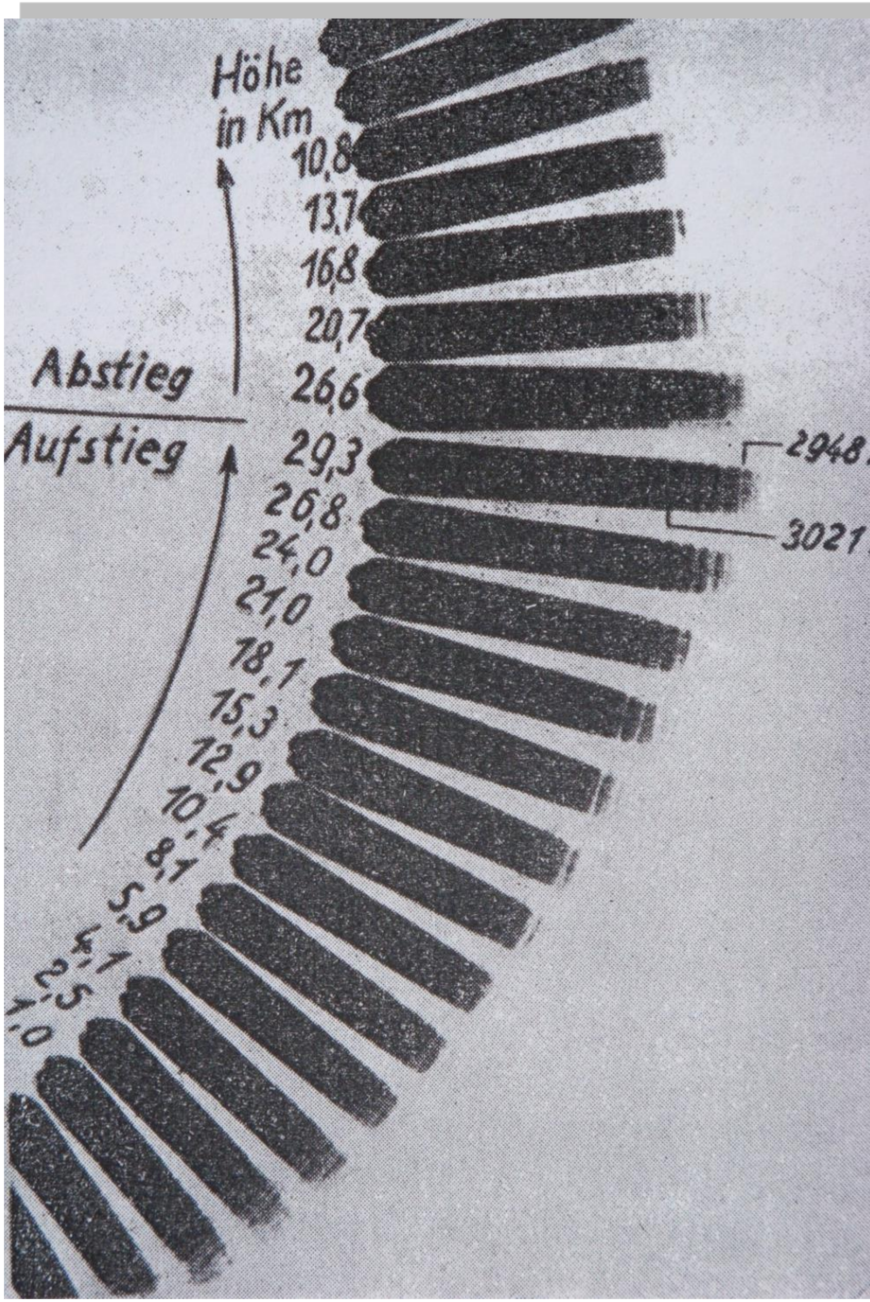


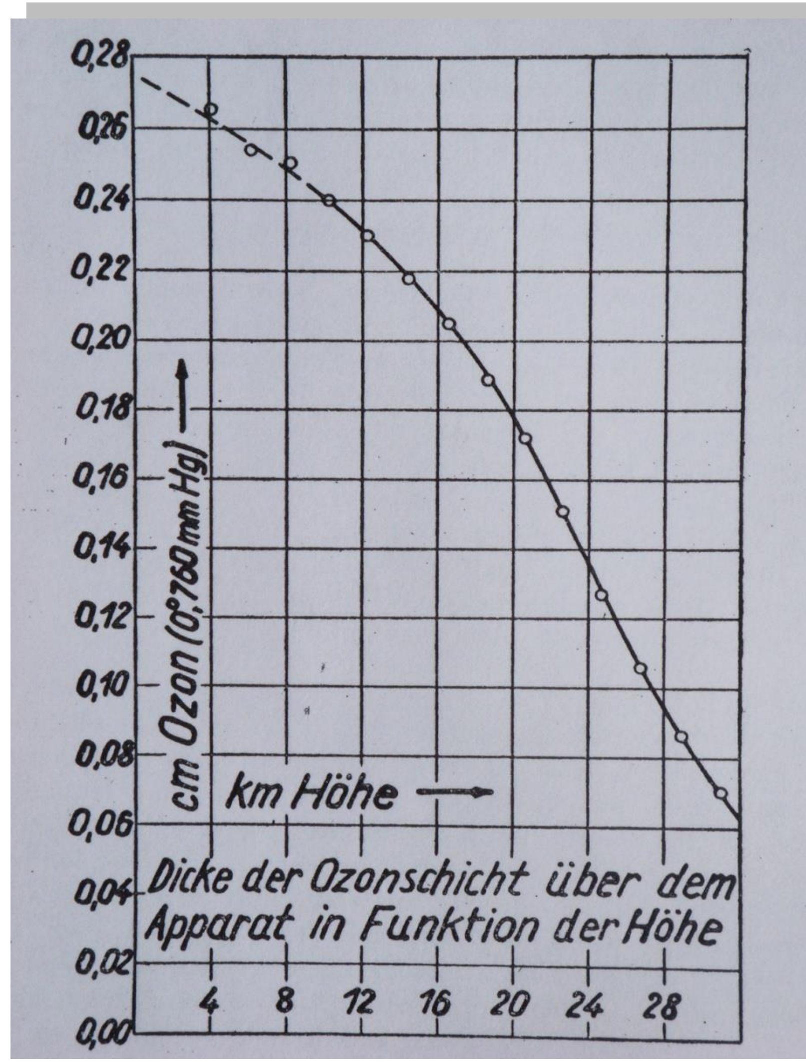
Erich Regener
1881-1955

Victor Regener
1913-2006

- **Erich Regener** and his son **Victor** who measure the solar ultraviolet absorption from a stratospheric balloon in 1934, show that the ozone maximum is located near 25 km.

Regener's Data





The RMI becomes a leading research center for meteorology

Jules Jaumotte, Director of RMI from 1919 to 1940, introducted in Belgium the concepts of frontology and the modern methods of dynamic and synoptic meteorology

The secretariat of the International Geophysical Year (1957-1958) is hosted at the RMI

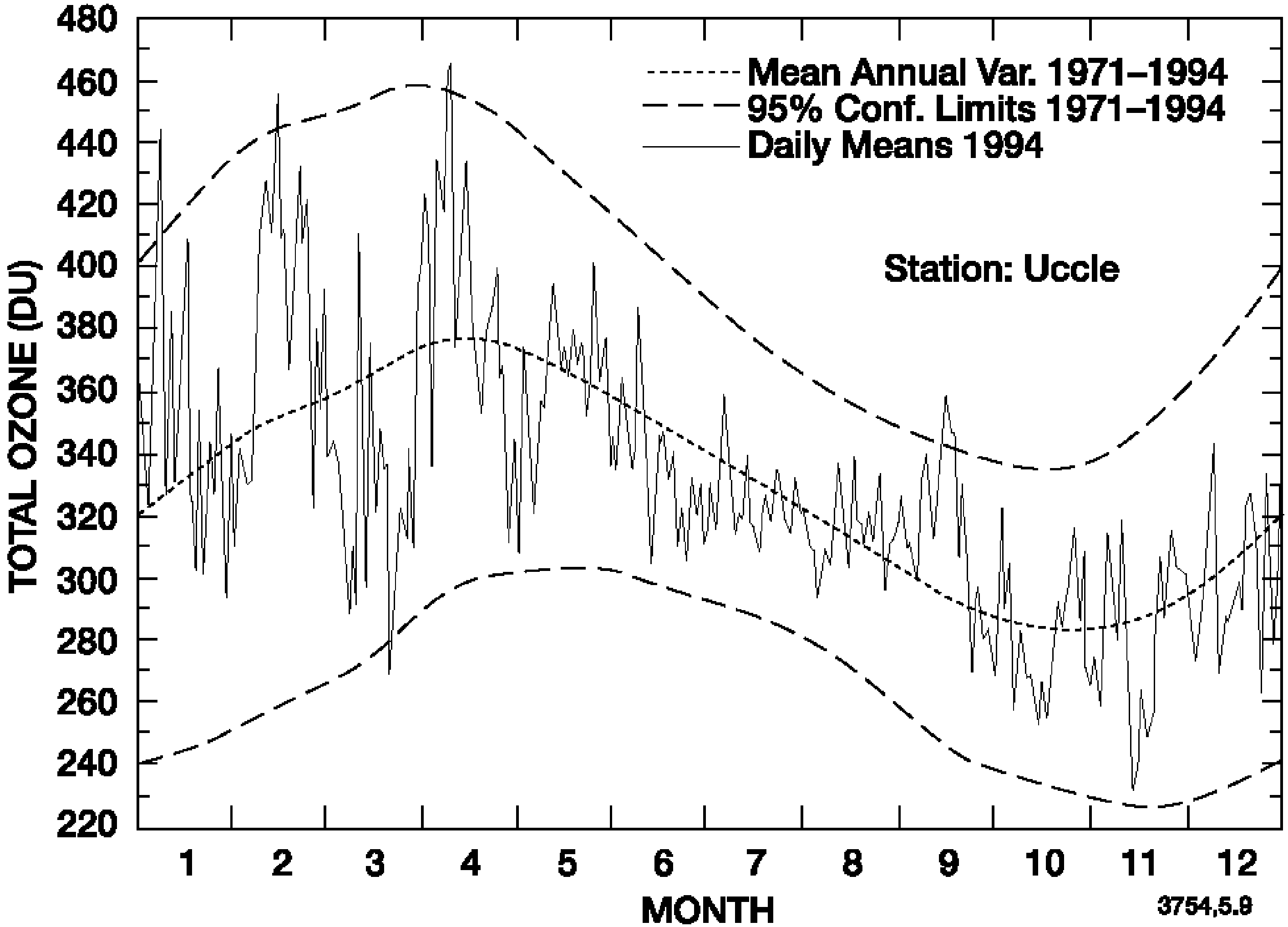
V.V. Belousov (USSR) , L. Berkner (USA), M. Nicolet (Belgium), J. Coulomb (France), and S. Chapman (Great Britain)

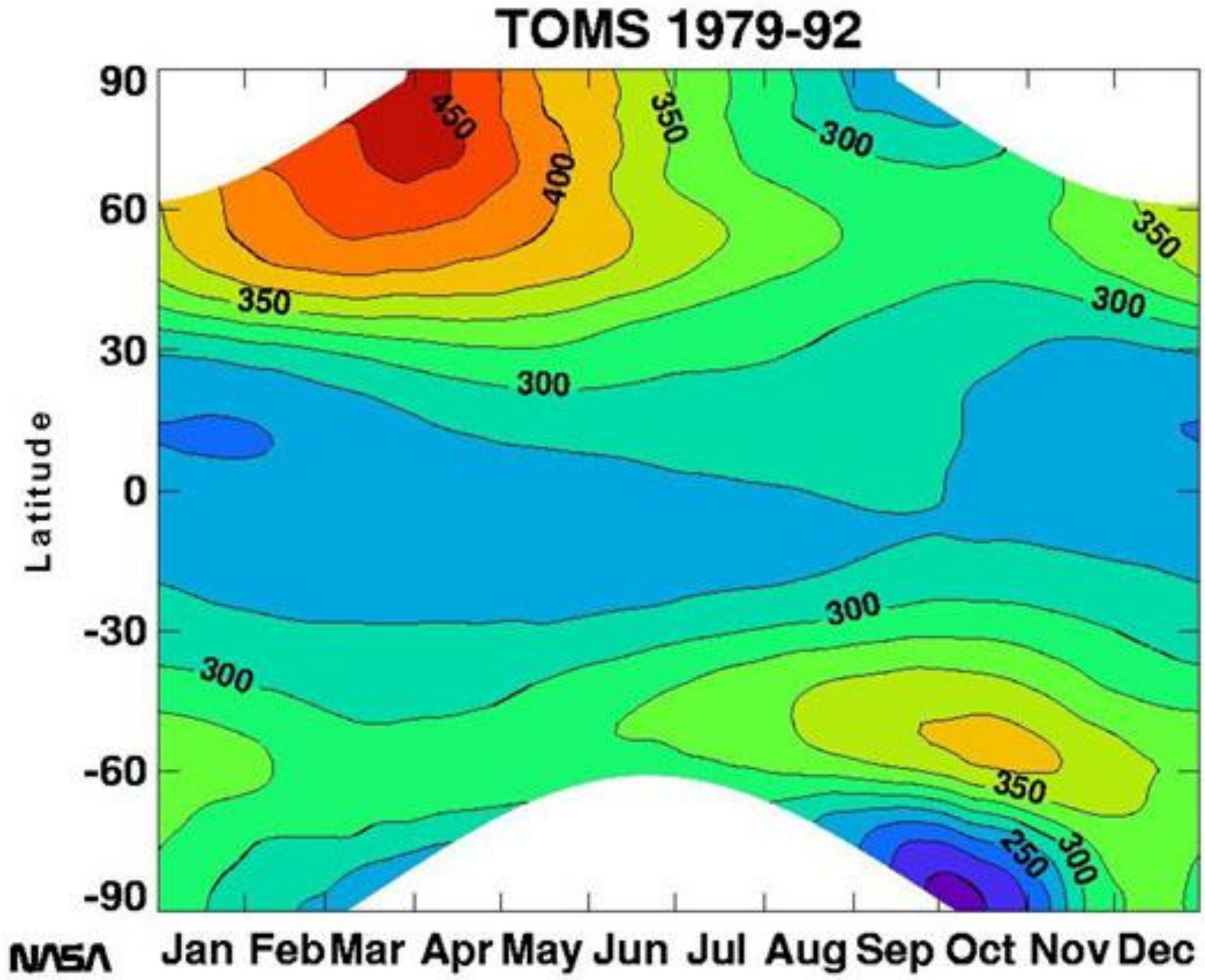

ANNÉE
GÉOPHYSIQUE
1957
Space Symposium

Considerable developments in dynamic meteorology, atmospheric energetics and atmospheric chemistry occurs in the 1950's and 1960's

Jacques van Mieghem, Director of the RMI from 1962 to 1970, creates a section on atmospheric chemistry and radioactivity and establishes regular ozone soundings in Uccle.

The RMI measures routinely the ozone column: a very important task





TOMS 1979-92
90
60
30
0
-30
-60
-90
Latitude
450
400
350
300
350
300
300
300
300
350
250
300
NASA
Jan
Feb
Mar
Apr
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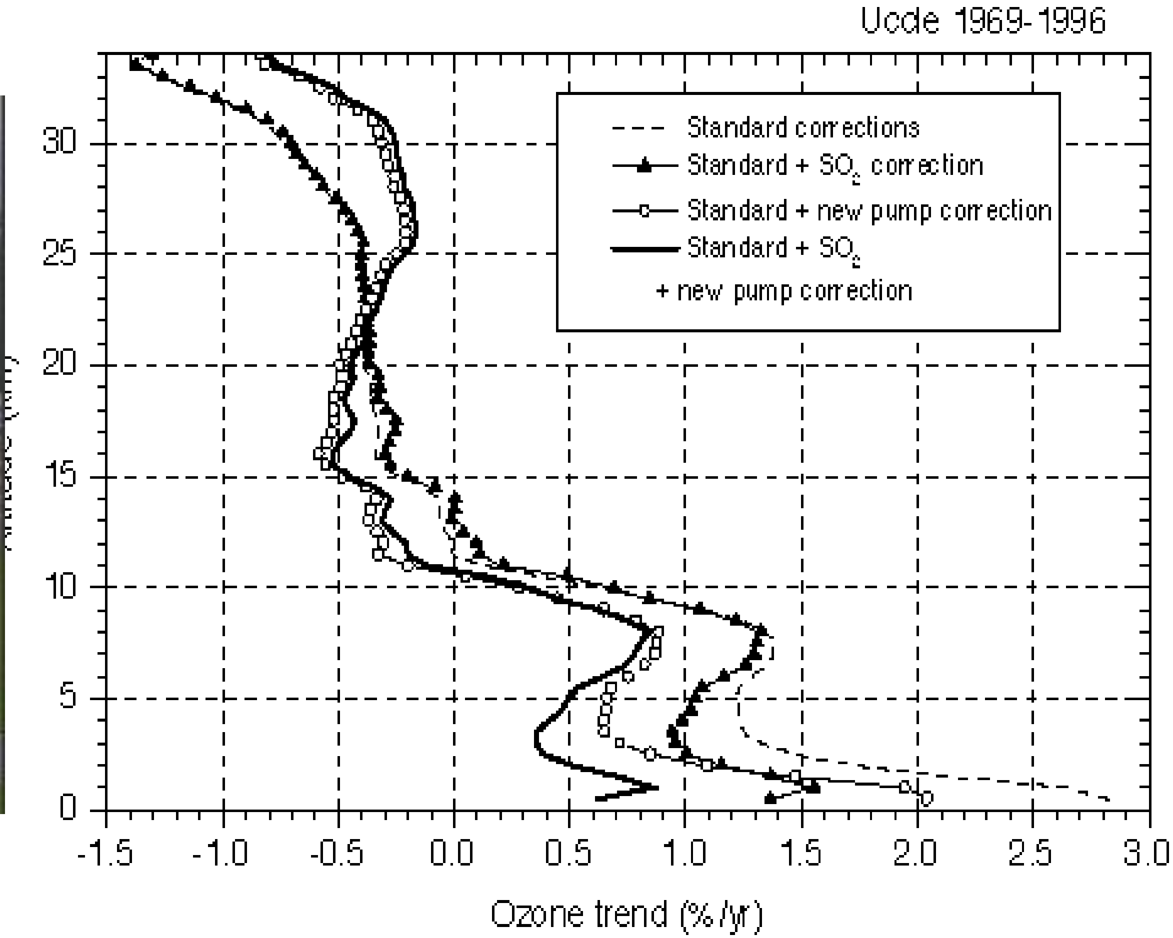
Oct
Nov
Dec

The RMI develops a systematic observation system for the ozone profile. Trends are observed



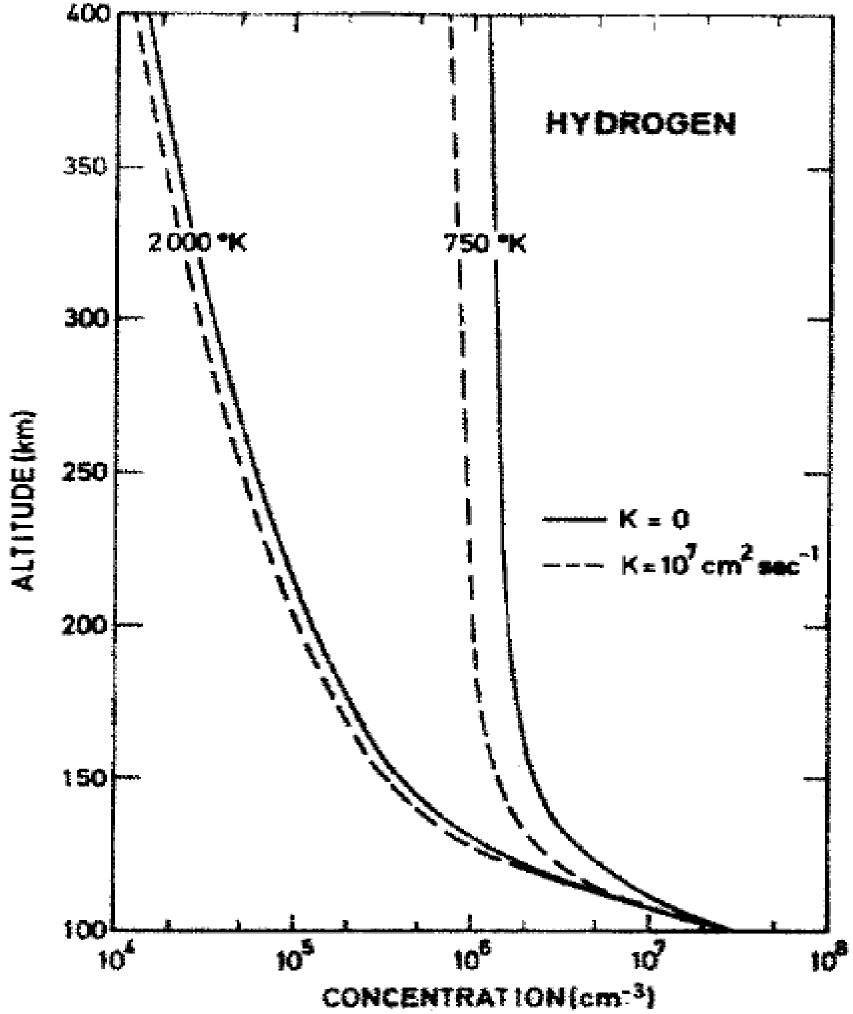
Space Aeronomy: The Creation of the National Center for Space Research at RMI.

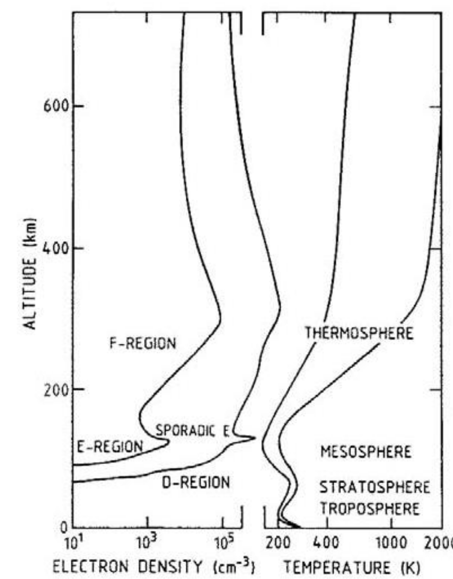
- The Royal Meteorological Institute creates in 1959 a National Center for Space Research.
- Established in 1964, the Belgian Institute for Space Aeronomy focuses on the physics and chemistry of the **atmosphere** of the Earth and other planets, and of outer space.

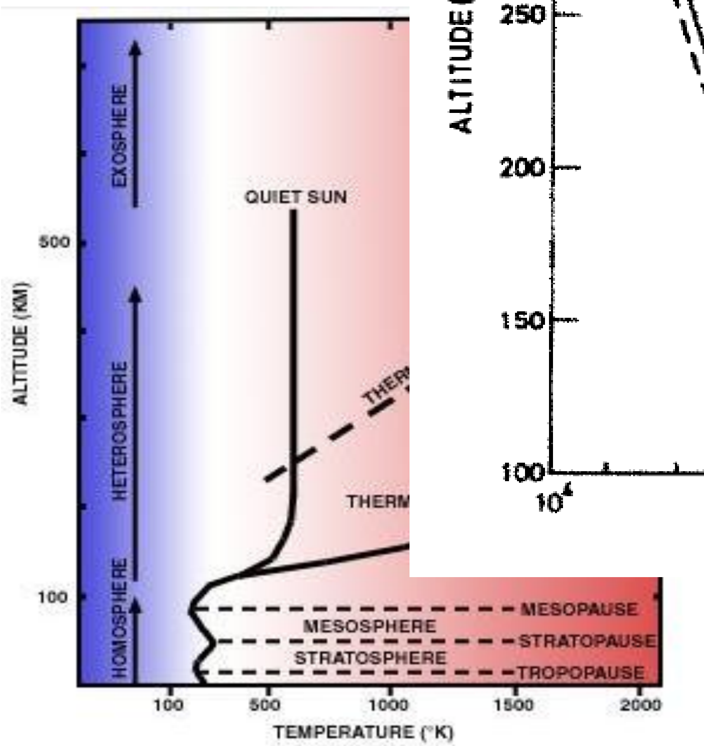
Active research is initiated at F
upper atmos
space age

k

Studies on the the
M. Nicolet and







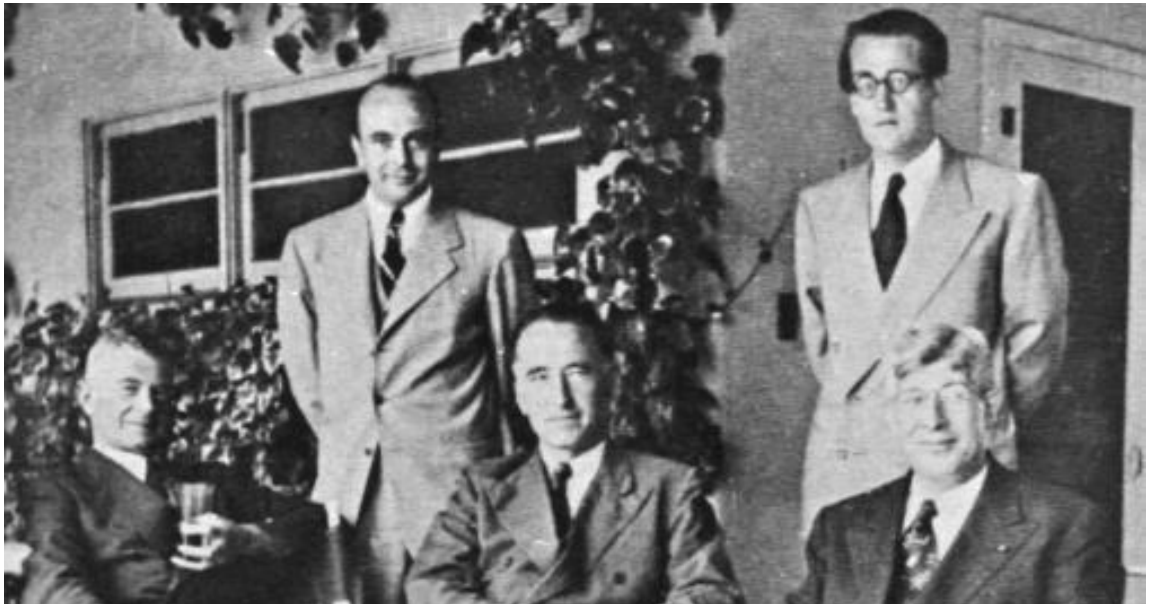
Back to Ozone

- In 1950, during a sabbatical at CalTech in Pasadena, Sir **David Bates** (Belfast) and Baron **Marcel Nicolet** (Brussels) suggest that hydrogen radicals (H, OH, HO_2) produced by photolysis of water vapor and methane provide a major ozone destruction mechanism in the ***mesosphere.***

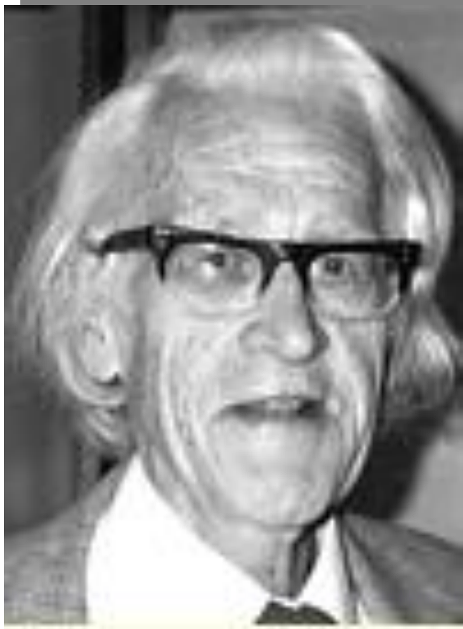
$OH + O_3 \rightarrow HO_2 + O_2$

$HO_2 + O \rightarrow OH + O_2$

Net: $O + O_3 \rightarrow 2O_2$



Bates



Nicolet

Crutzen

Ozone and Nitrogen

- **Paul Crutzen** shows that the major ozone loss in the stratosphere is provided by a catalytic cycle involving the presence of nitric oxide (NO).
- The solar proton event of 1972 confirms that ozone is depleted by NOx
- Nitric oxide is produced in the stratosphere by oxidation of nitrous oxide (N_2O). This gas is produced by bacteria in soils.
- The presence of nitrogen compounds in the stratosphere was first detected by **Rhine** et al. (7.6 10 E15 cm^{-2} above 18.8 km) and by **David Murcray** (U. of Denver).

$NO + O_3 \rightarrow NO_2 + O_2$

$NO_2 + O \rightarrow NO + O_2$

$O + O_3 \rightarrow 2O_2$

The detection of NO_2 and CH_4 from balloon-borne infrared instruments

Pure and Applied Geophysics (PAGEOPH)
Vol. 106–108 (1973/V–VII)
Birkhäuser Verlag, Basel

Stratospheric Methane and Nitrogen Dioxide from Infrared Spectra

By M. ACKERMAN and C. MULLER[1])

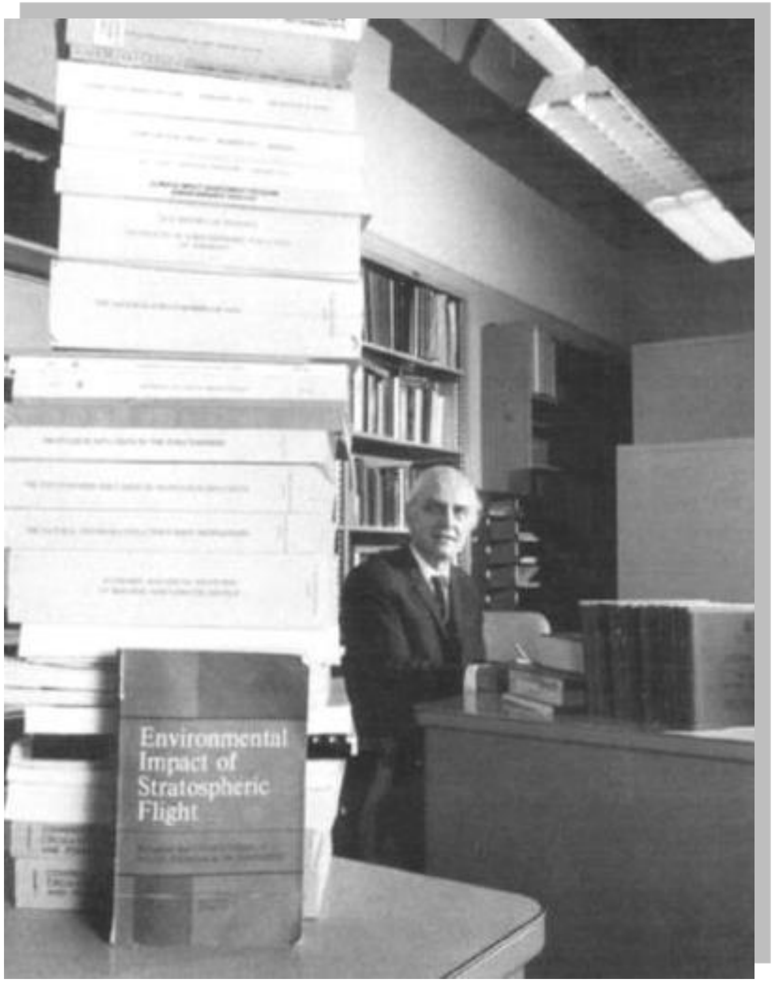
Abstract – Values of the mixing ratio by volume of stratospheric NO_2 and CH_4 deduced from infrared spectra taken by means of balloon-borne spectrometers are presented. Possible evidence for the presence of formaldehyde in the stratosphere is also given.

I. Introduction

Most of the information on minor stratospheric constituents has been deduced from infrared spectra. Until recently this technique has been applied to absorbers having volume-mixing ratios of the order of 10^{-7} or higher. The measurements based on absorption infrared spectrometry performed by means of balloon-borne spectrometers operating in the 30 km altitude range at solar zenith angles larger than 90° are allowed to reach concentrations lower by two orders of magnitude. This method offers several advantages: changes in absorption can be related to short-term variation of the optical thickness, ensuring the absence of contamination by the instrumentation; the absorption takes place mostly at constant temperature, absorber concentration and pressure, leading to an easier quantitative interpretation. Information on the vertical distribution of minor constituents can be obtained by this method since the optical depth is maximum

Baron Marcel Ackerman

The Impact of Aviation

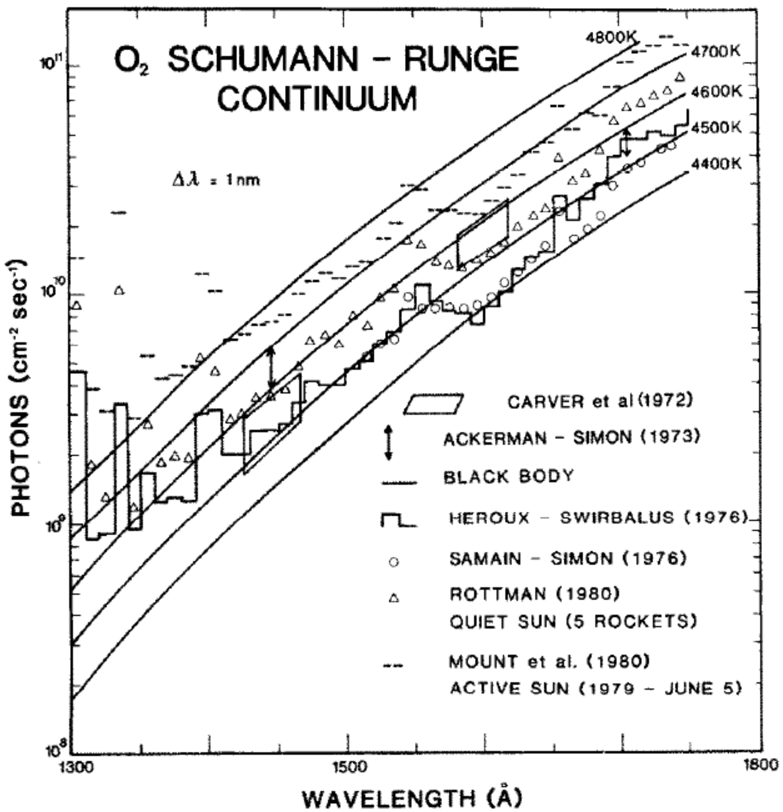


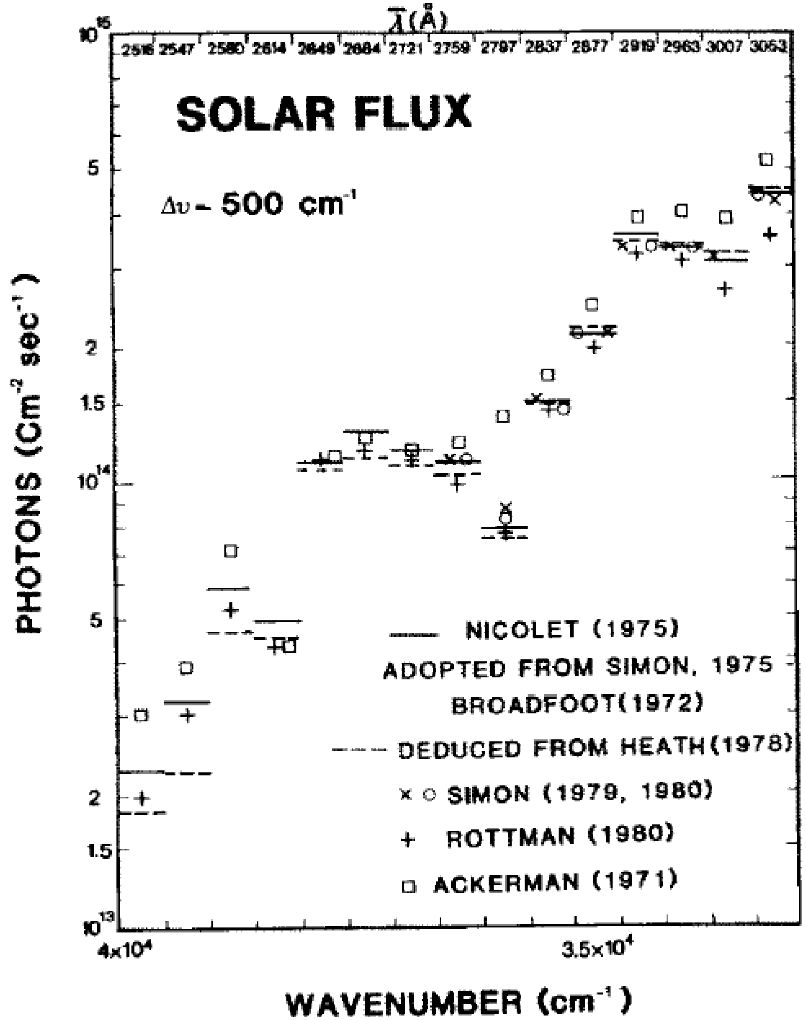
H. Johnston

- In 1971, **Harold Johnston** (University of California at Berkeley) suggests that the nitrogen oxides to be released by a projected fleet of **supersonic aircraft** could produce substantial ozone depletion.
- An intensive research program, the **Climatic Impact Assessment Program** (CIAP) is sponsored by the US Department of Transportation (1972-1974).
- This is the first **gold age** of stratospheric research.

The Role of Atmospheric Photochemistry: Observation of the UV Solar Spectrum

Paul Simon

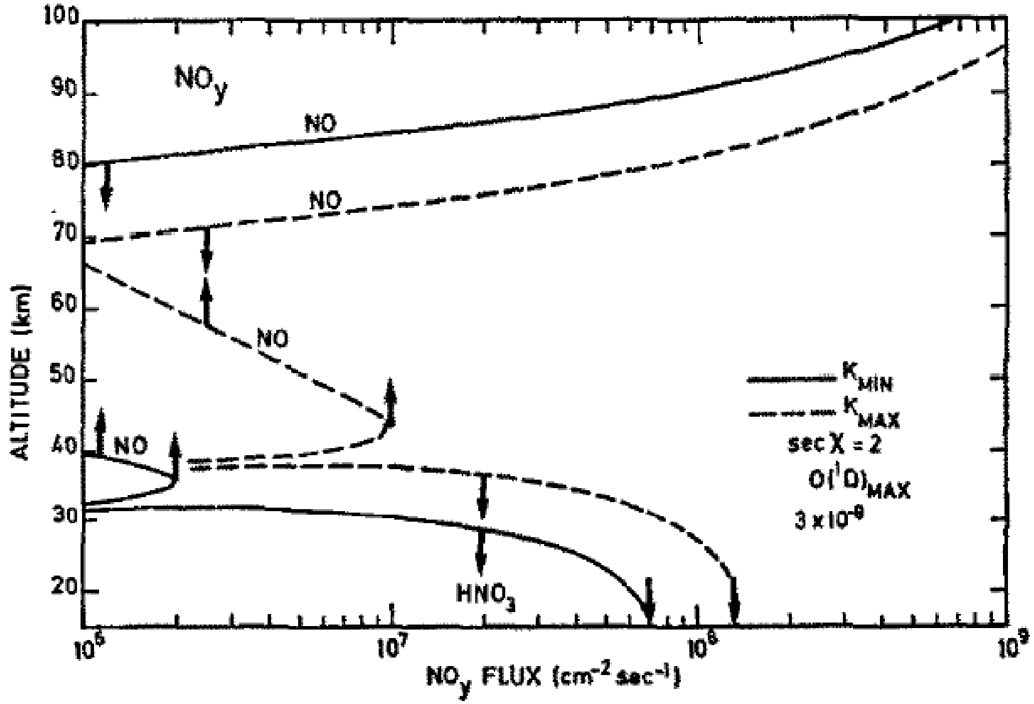


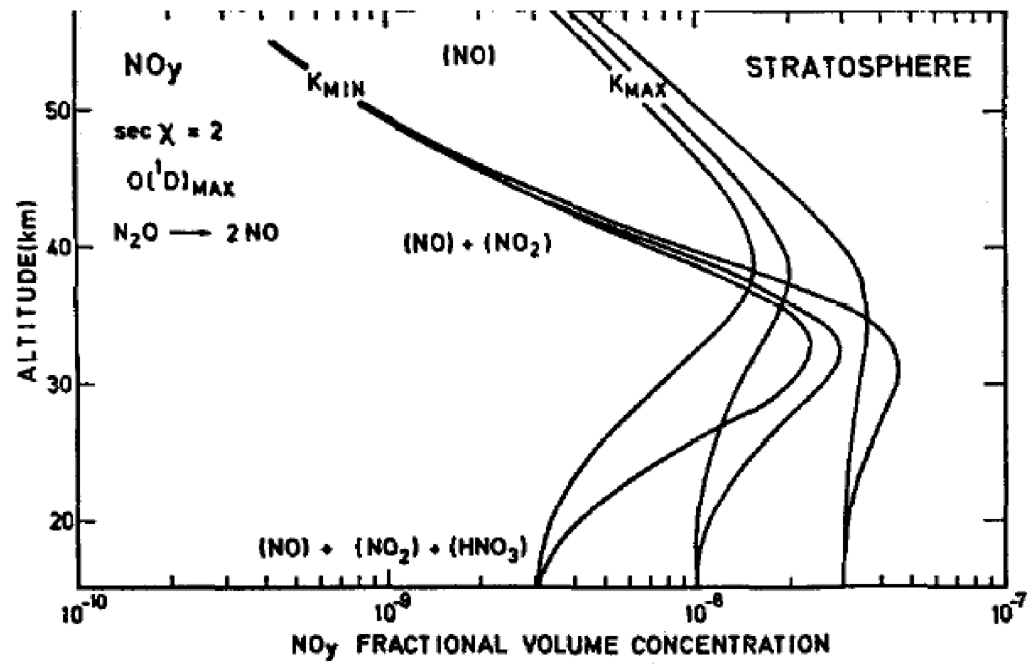


Nitrogen Oxides

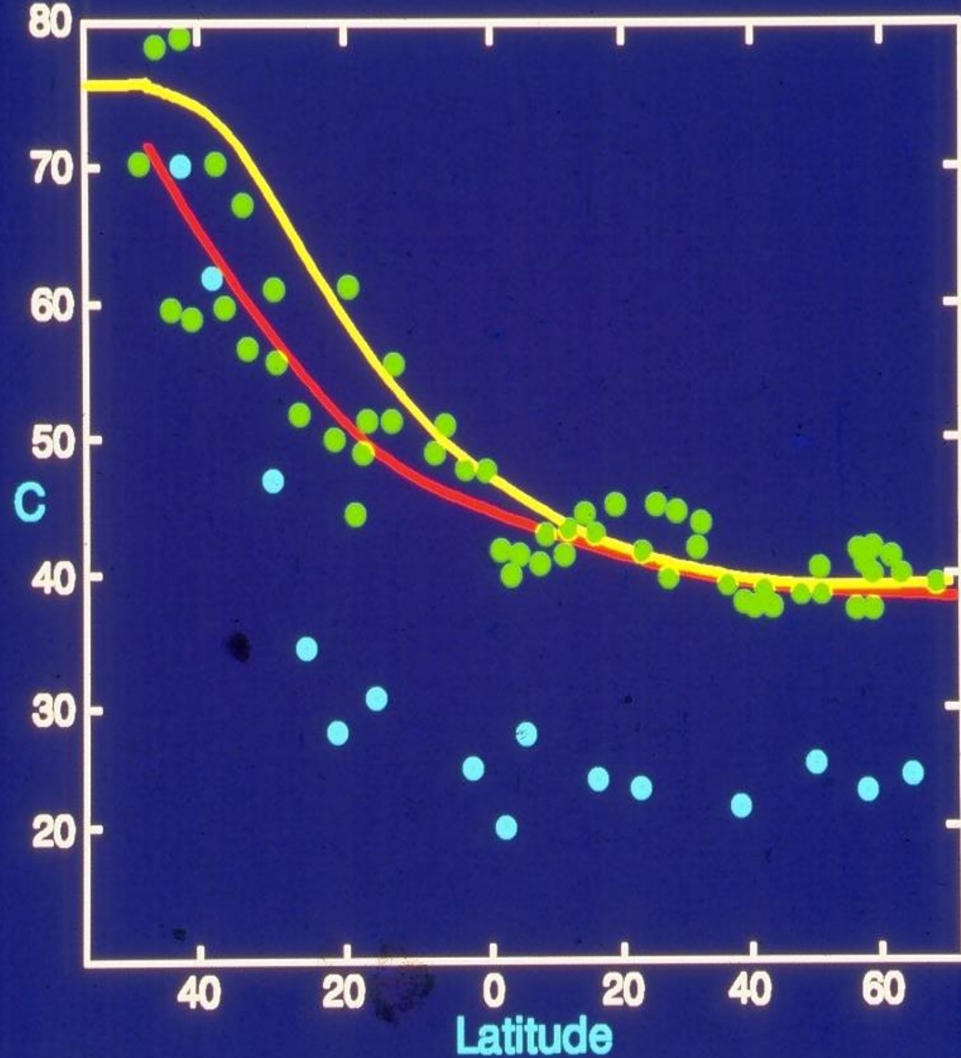
In Belgium, under the leadership of M. Nicolet, **numerical models of the middle atmosphere** are developed and used to calculate the vertical distribution of trace constituents in the stratosphere and mesosphere.

It is shown, for example (Brasseur and Nicolet, 1973), that NO produced above 100 km by ionospheric reactions **cannot be transported** to the stratosphere, and that the presence of NOx below 50 km altitude must be due to **local production.**





1971; James Lovelock measures CFC-11 in the Northern and Southern Hemispheres



Distribution of CCl_3F in and over the North and South Atlantic Ocean

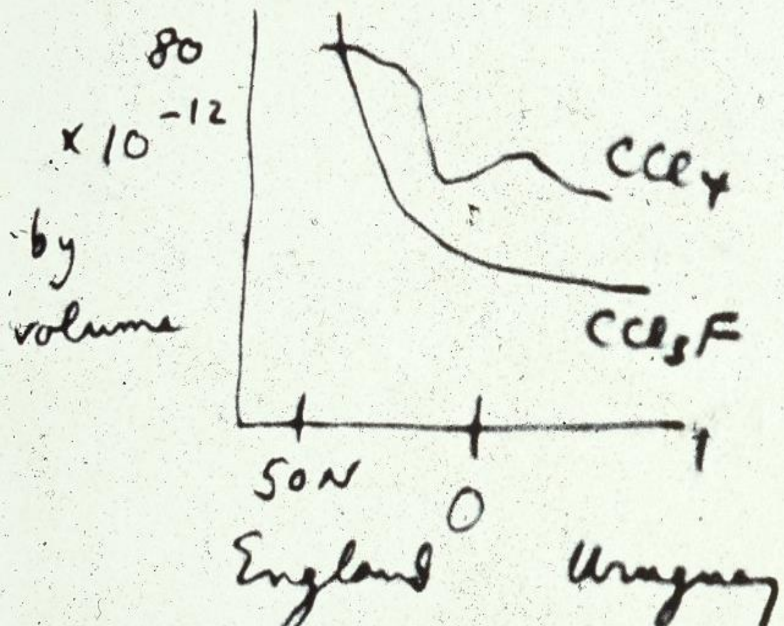
●, Aerial concentrations (x 10^{-12}) by volume

J. LOVELOCK ET AL. NATURE,19

Lovelock's data

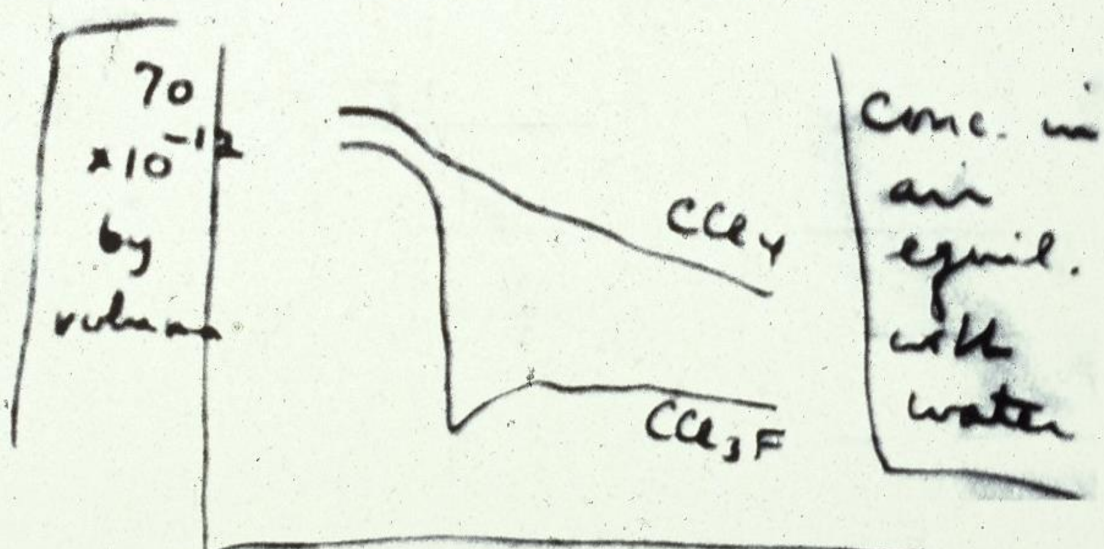
Lecture notes, F. S. Rowland, Fort Lauderdale, Florida, Feb. 1972

CCl_3F – Freon-11 inert gas in spray cans.



Dr. J. Lovelock

CCl_4 – appreciably different

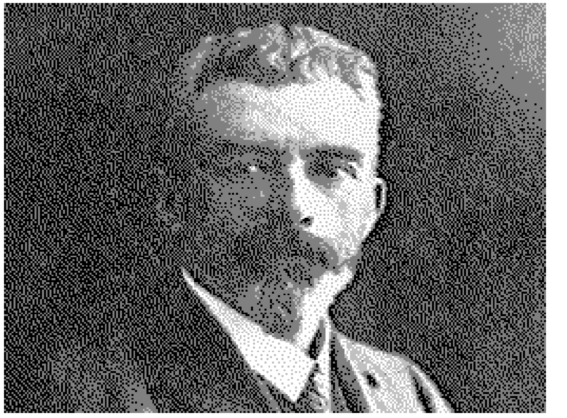


DuPont's estimates

most put out since 1960

Cruise of R. R. Shackelton
Nov. 1971

The First Synthesis of CFCs

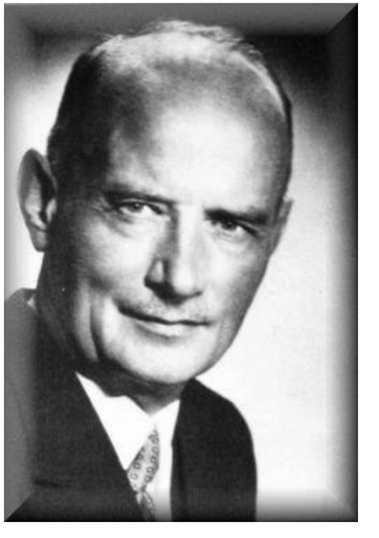


Swarts

- The first synthesis of chlorofluorocarbons was performed by a Belgian chemist **Frédéric Swarts** (Sept. 2, 1866 - Sept. 6, 1940).
- In 1892 he prepared $CFCl_3$ (CFC-11) by

$$SbF_3Br_2 + CCl_4 \dashrightarrow CFCl_3 + SbF_2Br_2Cl$$

Ruff



- Later Swarts and then **Otto Ruff** (Dec. 30, 1871 – Sept. 17, 1939) in Germany produced CF_2Cl_2 (CFC-12) by an entirely different process.
- The significance of these products was not recognized for about 30 years.

Chlorofluorocarbons as Refrigerants

Midgley

The pioneering work of Frederic Swarts prompted American scientists **Thomas J. Midgley Jr.** (1889-1944) and his assistants **Albert L.Henne** (1901-1967) and **Robert R. McNary** (1903-1988) at the Thomas and Hochwalt Laboratory in Dayton, Ohio to develop in 1928 chlorofulorocarbons as referigerants.

- Midgley demonstrated the **nontoxic** and **nonflammable** properties of the CFCs at a meeting of the American Chemical Society in April 1930 by inhaling CFC-12, then blowing it over a candle flame, extinguishing the flame.
- Midgley also invented the **leaded gasoline**.

Ozone and Chlorine

- In 1974, **Richard Stolarski** and **Ralph Cicerone**, then at the University of Michigan, suggested that **chlorine** could also catalytically destroy ozone in the stratosphere. They had been studying for NASA the possible impacts of the **Space Shuttle**. They also identified **volcanoes** as a source of atmospheric chlorine

$$Cl + O_3 \rightarrow ClO + O_2$$

$$ClO + O \rightarrow Cl + O_2$$

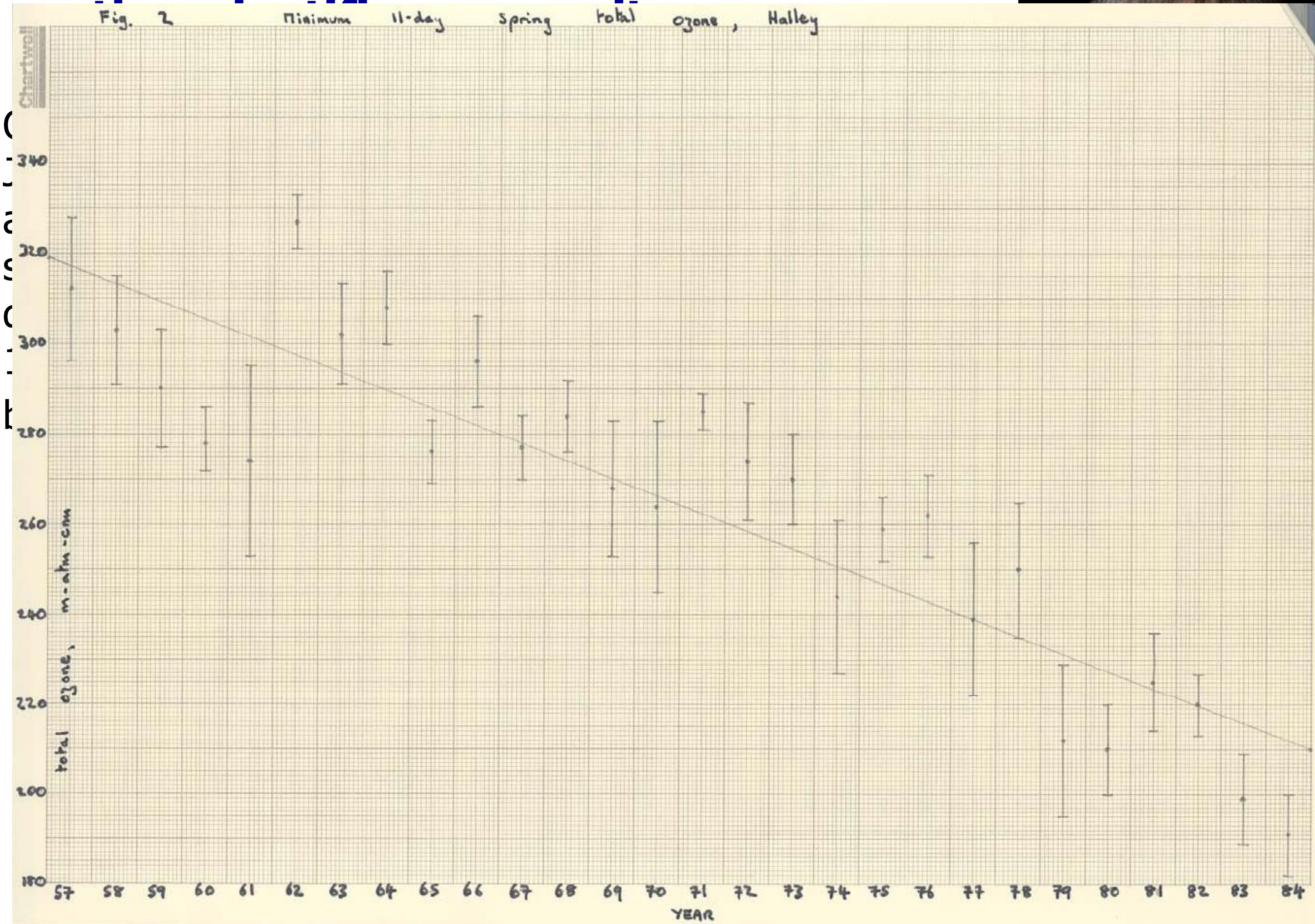
$$O + O_3 \rightarrow 2O_2$$

Stolarski

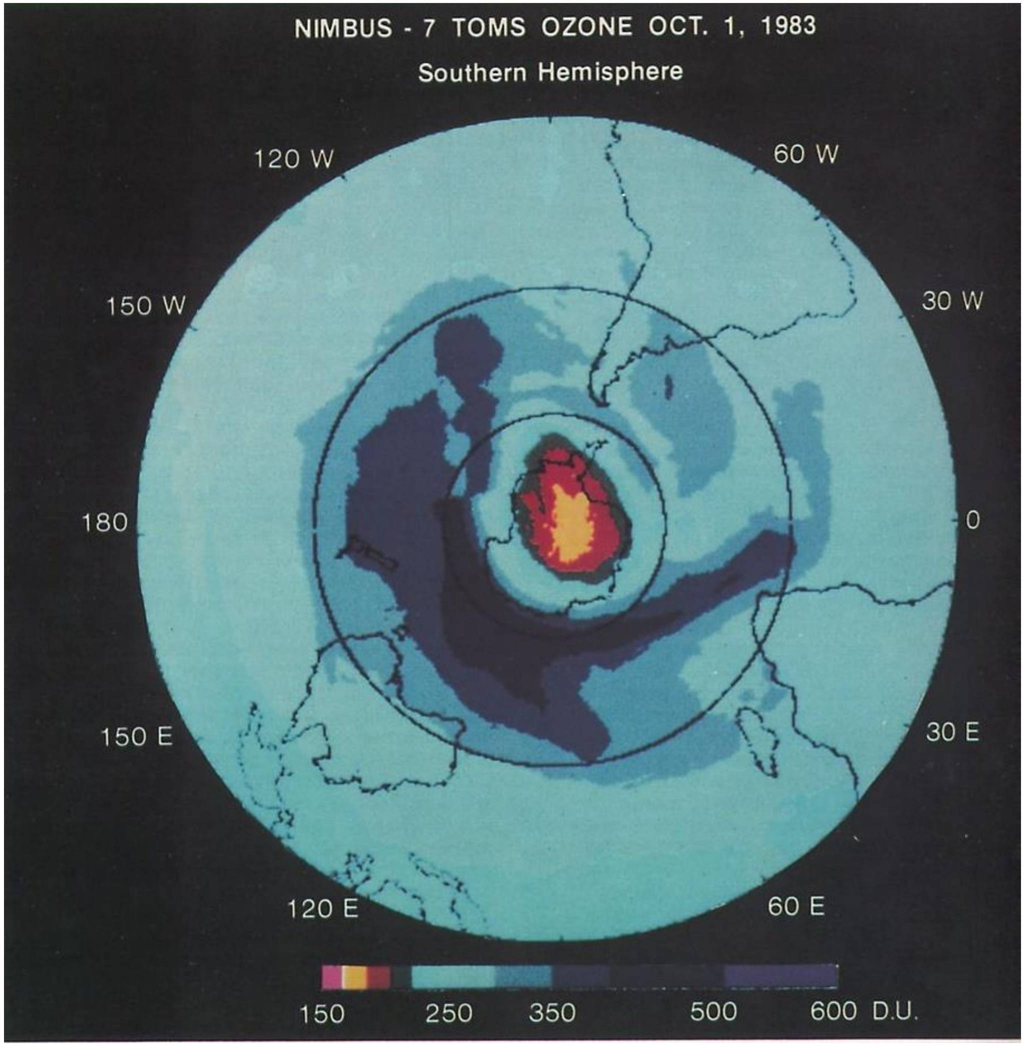
Cicerone

The Ozone Hole: A challenge for

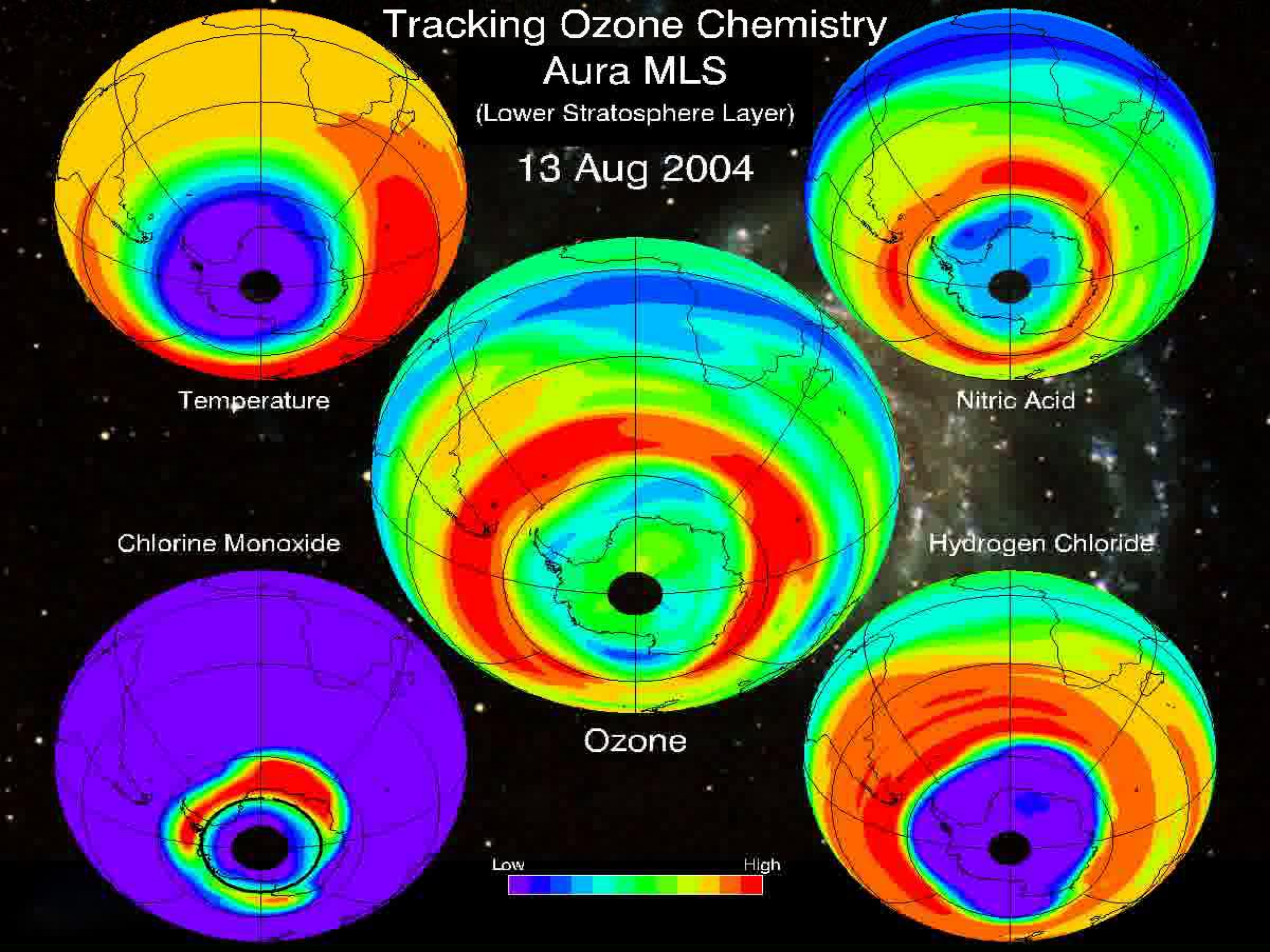
Chubachi



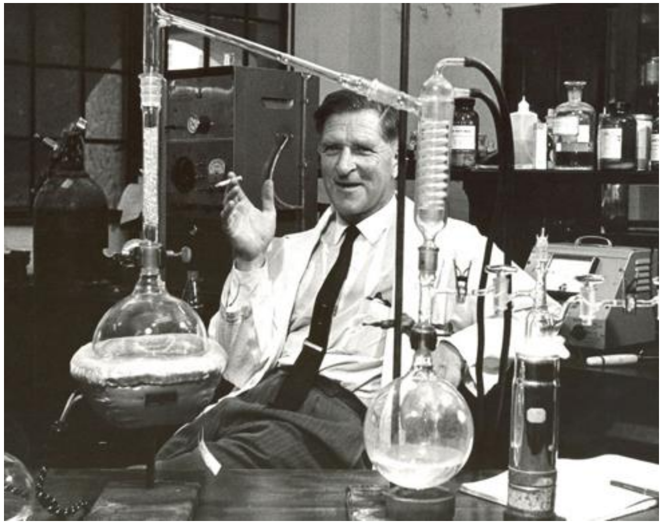
The First Satellite View of the Ozone Hole



- Total Ozone observed on October 1, 1983 by Nimbus-7 TOMS
- Bhartia et al., 1985.


Tracking Ozone Chemistry
Aura MLS
(Lower Stratosphere Layer)
13 Aug 2004
Temperature
Nitric Acid
Chlorine Monoxide
Hydrogen Chloride
Ozone
Low
High

The Photochemistry of Tropospheric Ozone in the Troposphere



Arie Haagen-Smith

- In the early 1950's, Dutch biogeochemist **Arie Haagen-Smith** suggests that the formation of urban ozone (Los Angeles smog) results from the action of sunlight on reactive hydrocarbons and nitrogen oxides released by oil refineries and automobiles.

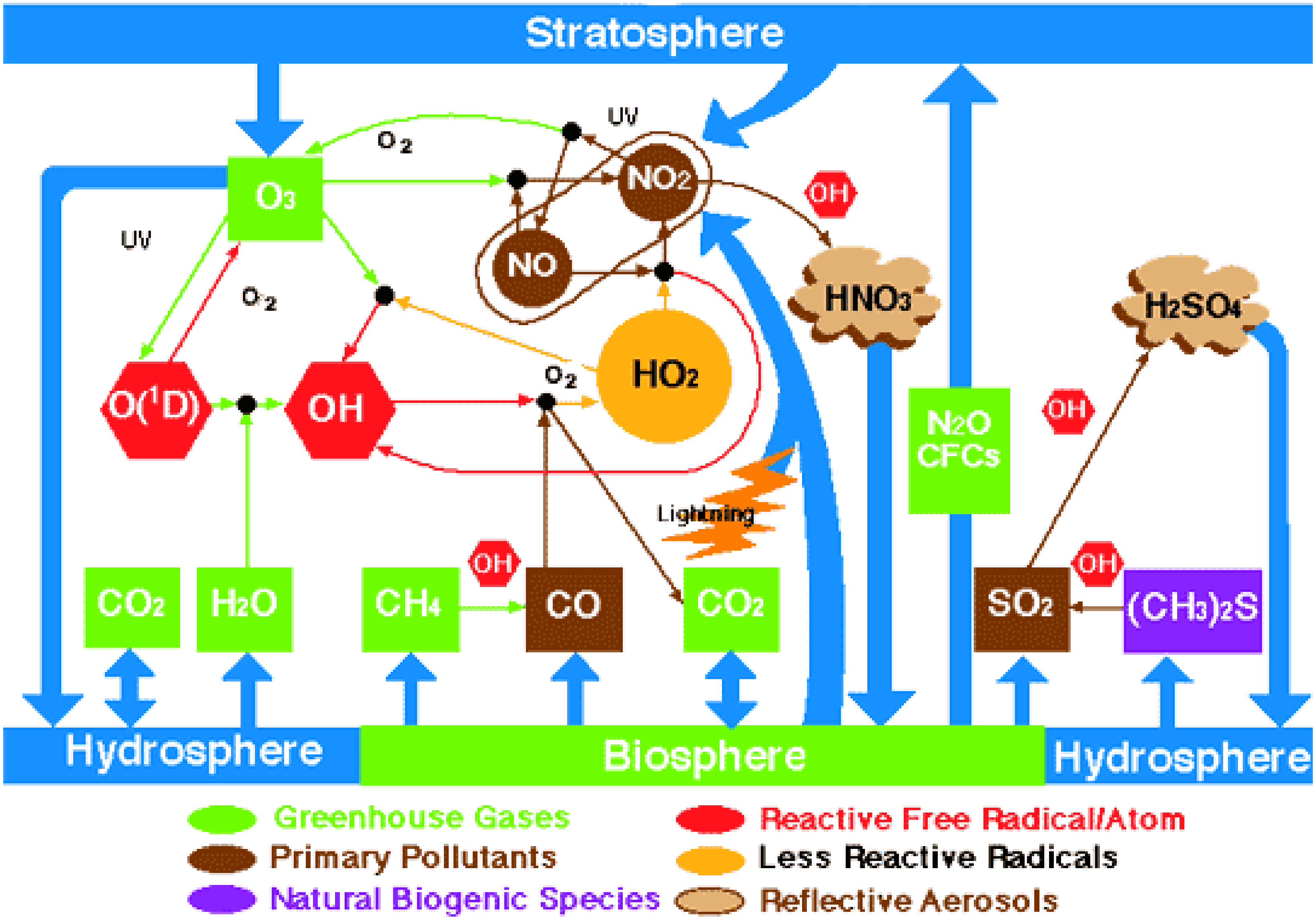
Tropospheric Ozone Photochemistry

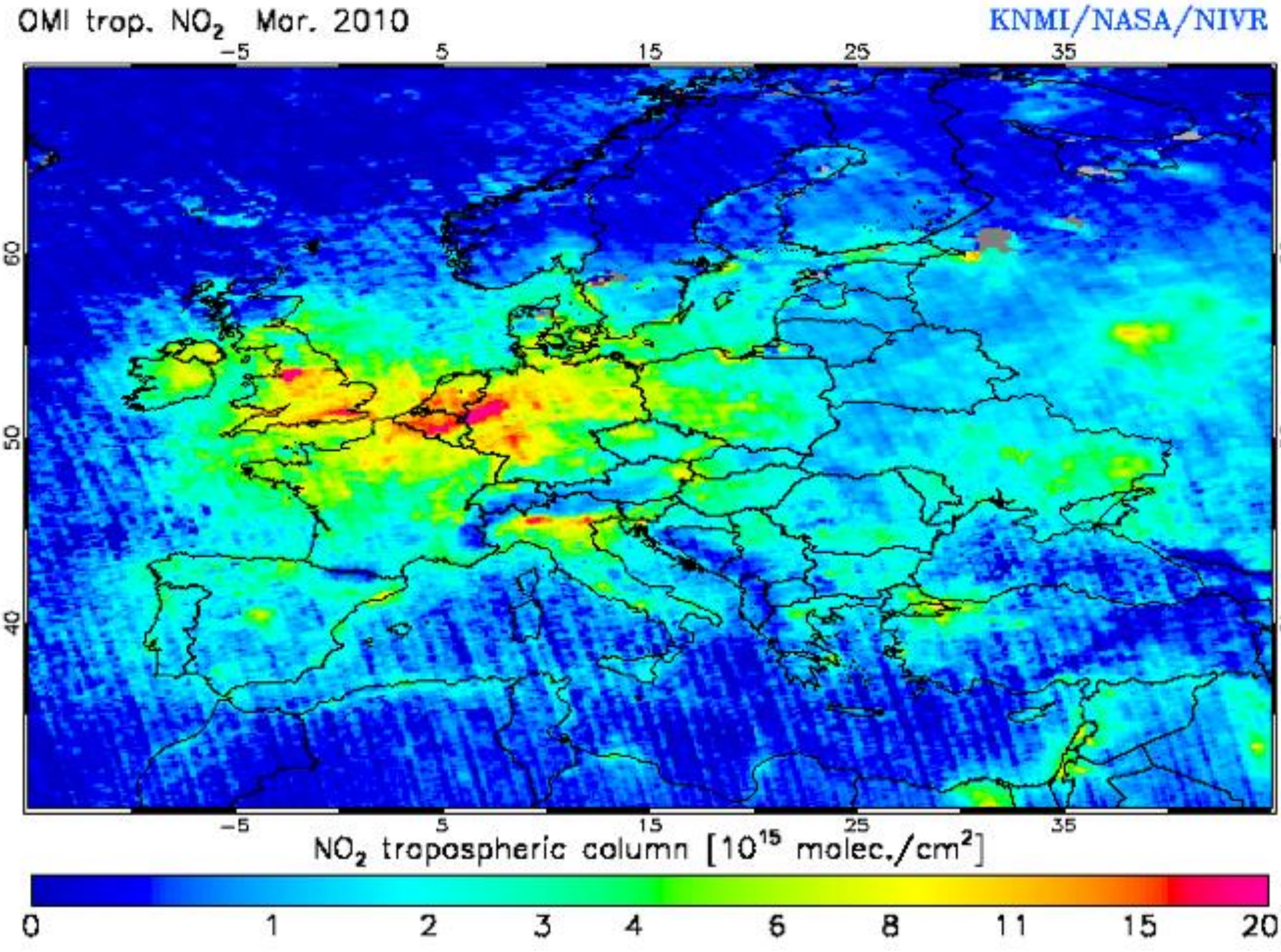
- In the early 1970, **Paul Crutzen** as well as **William Chameides** show that photochemical processes affect **tropospheric ozone** and the **hydroxyl radical** (OH) at the **global scale** and provide a mechanism to destroy pollutants in the atmosphere.

Crutzen

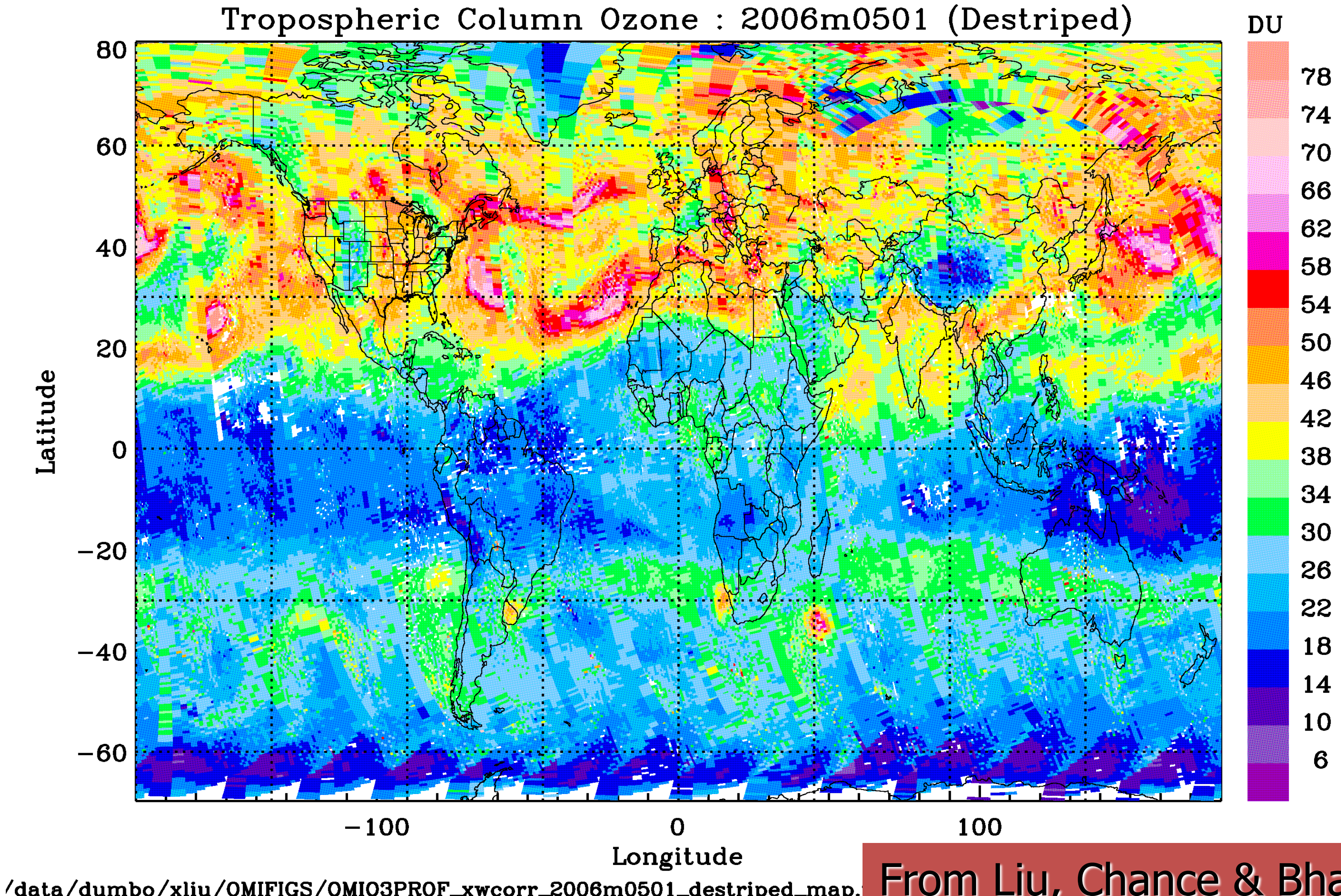
Chameides

Tropospheric Chemistry




OMI trop. NO_2 Mar. 2010
KNMI/NASA/NIVR
−5
5
15
25
35
60
50
40
NO_2 tropospheric column [10^{15} molec./cm^2]
0
1
2
3
4
6
8
11
15
20

Direct Retrieval of Trop Column O_3 by the Hyperspectral BUV technique

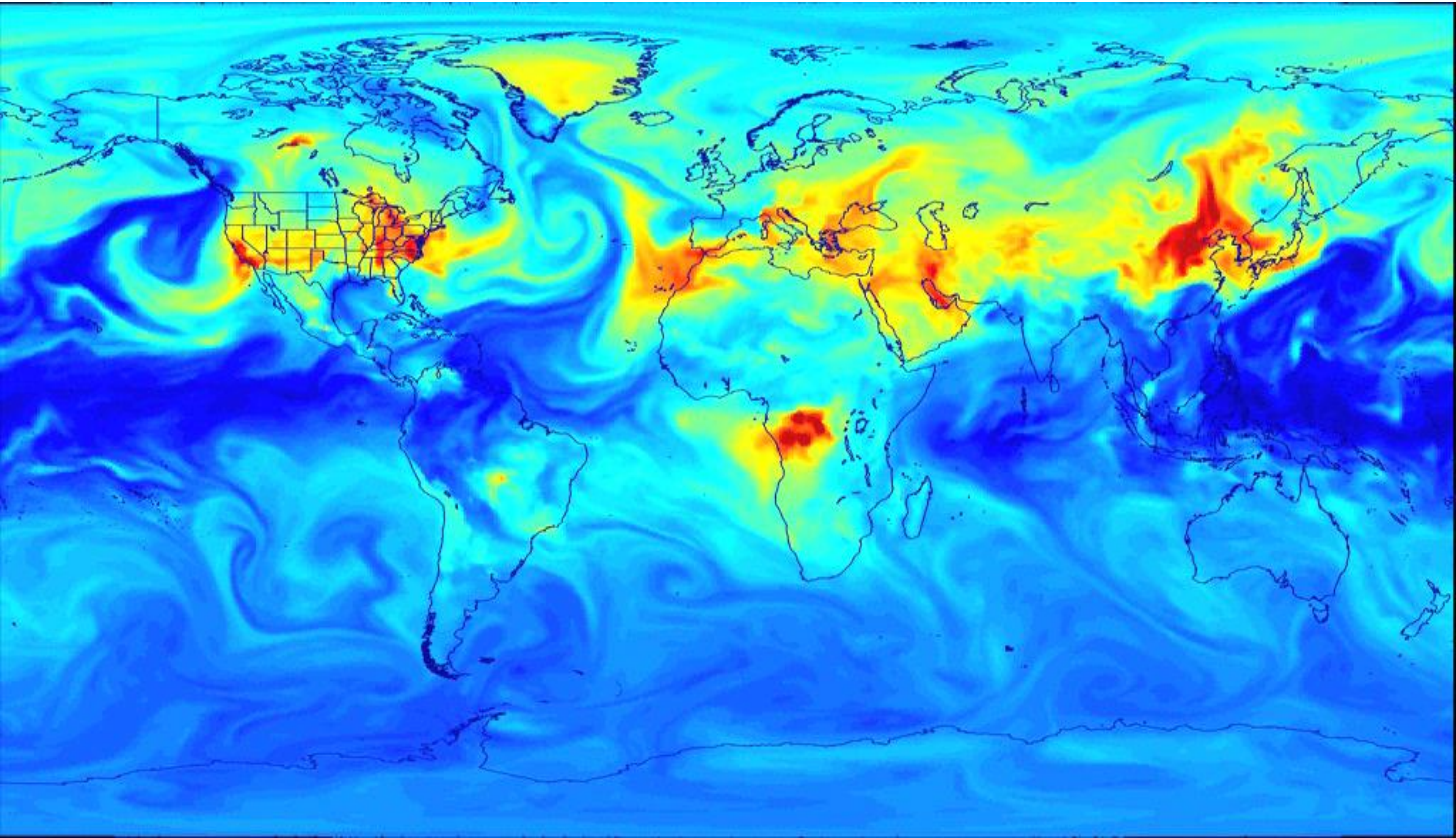


From Liu, Chance & Bhartia

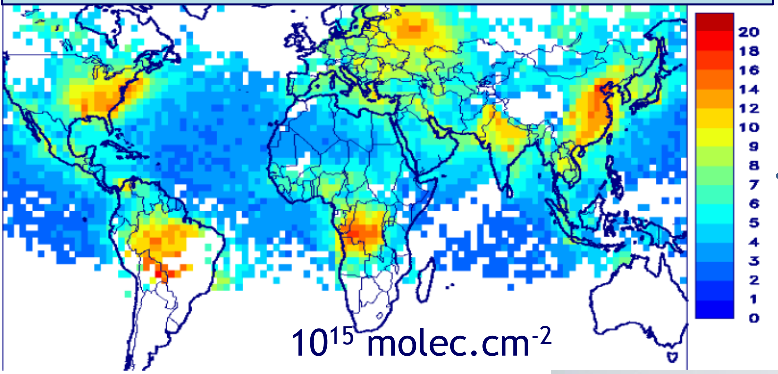
Global Modeling of surface ozone with a spatial resolution of 50 km.

Note the diurnal variation in the ozone mixing ratio

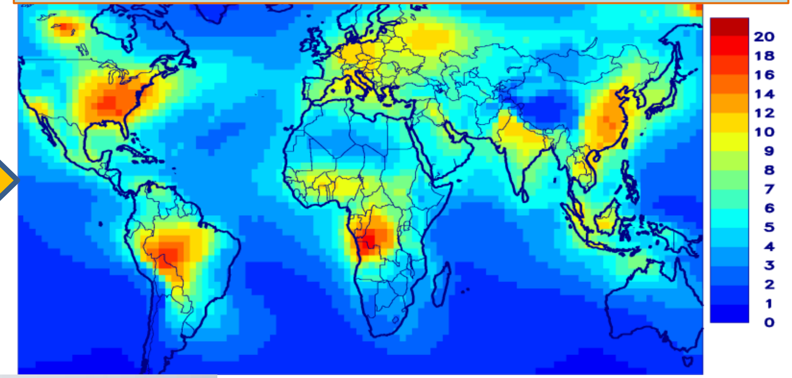
Louisa Emmons, NCAR



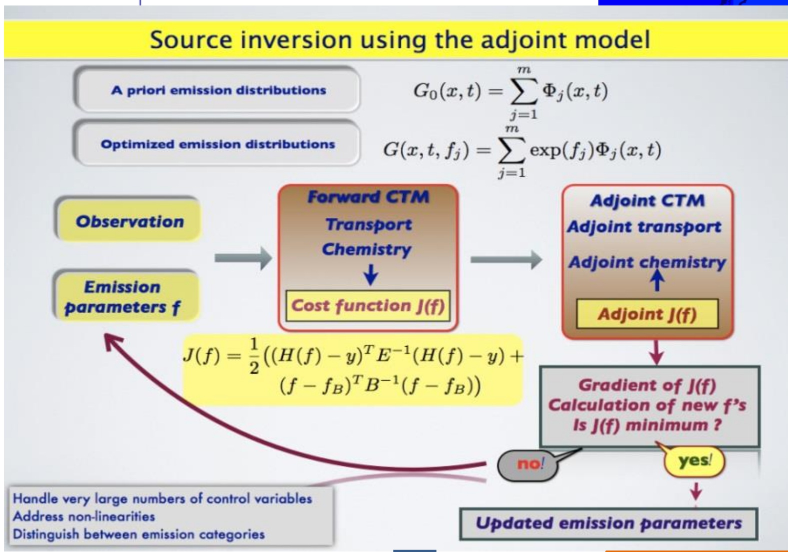
GOME-2 HCHO July 2010



IMAGESv2 HCHO columns

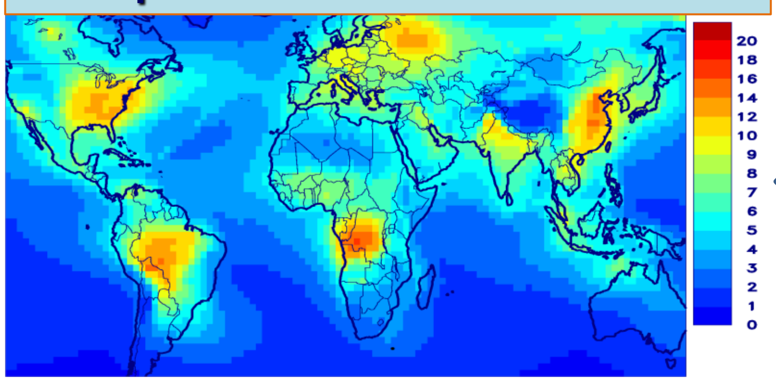


Deriving emissions of Isoprene from space observations of isoprene



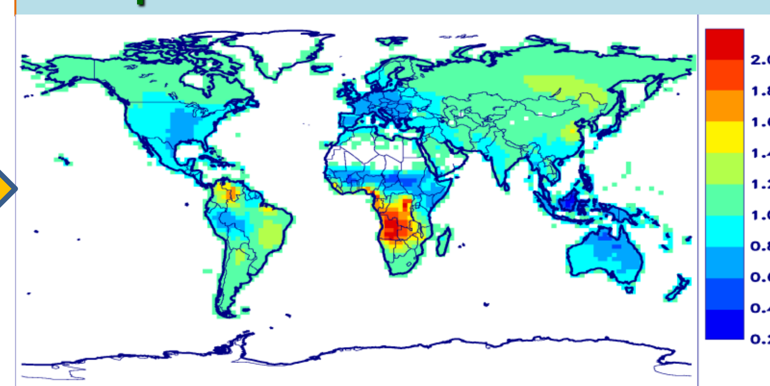
Combining chemical transport models with satellite data at BISA

Optimized HCHO columns

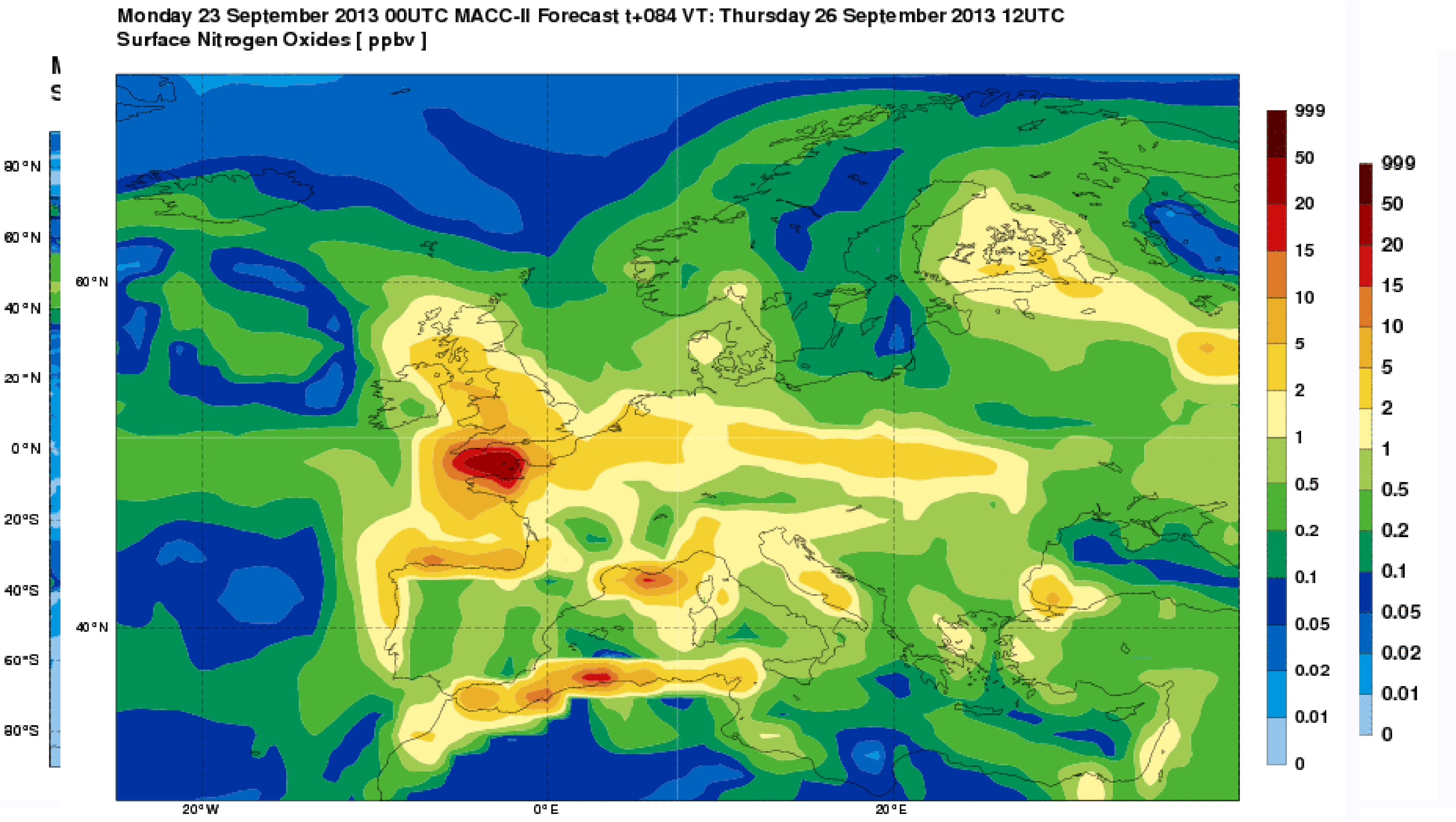


Stavrakou et al., Atmos. Chem. Phys., 2009

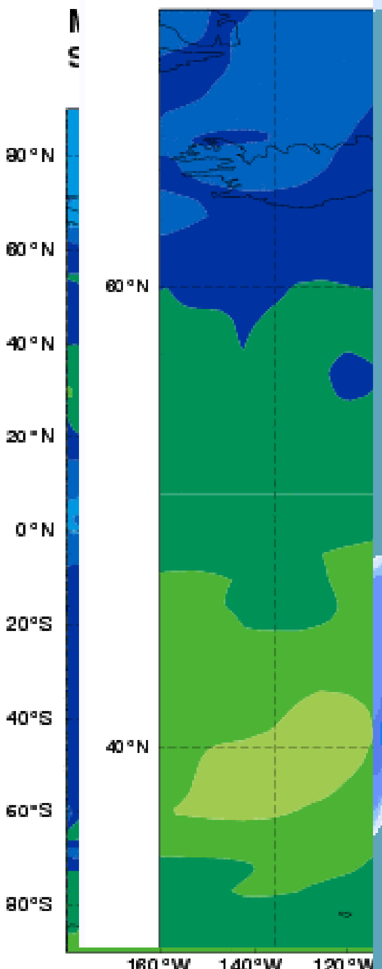
Isoprene emission increment



Predicting Nitrogen Oxides by the MACC II Project at ECMWF: Thursday 26 September 12:00 UT



Predict



80°N
60°N
40°N
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20°S
40°S
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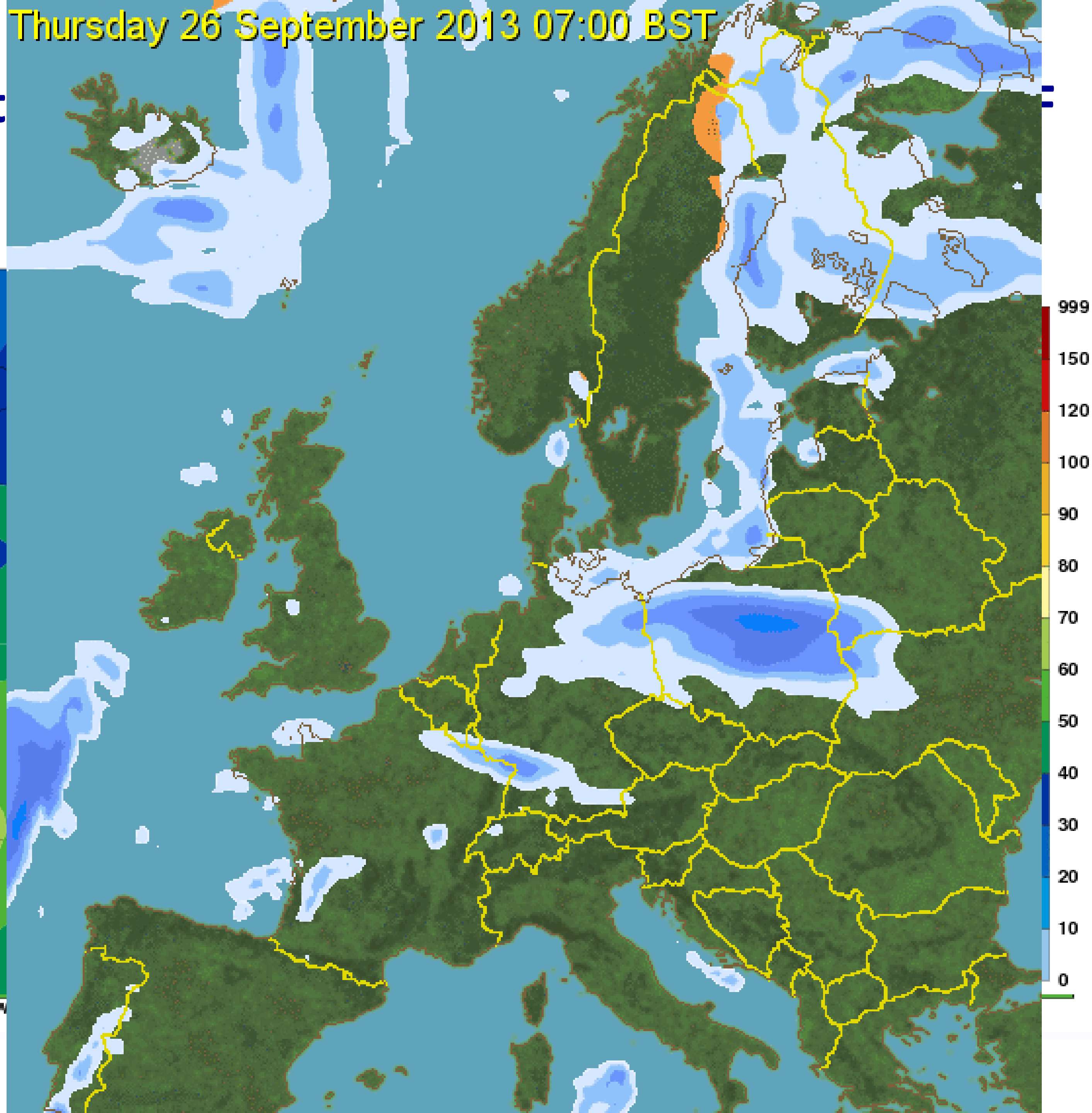
80°S
60°N
40°N
160°W
140°W
120°W



Thursday 26 September 2013 07:00 BST
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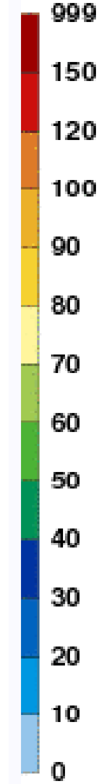

999
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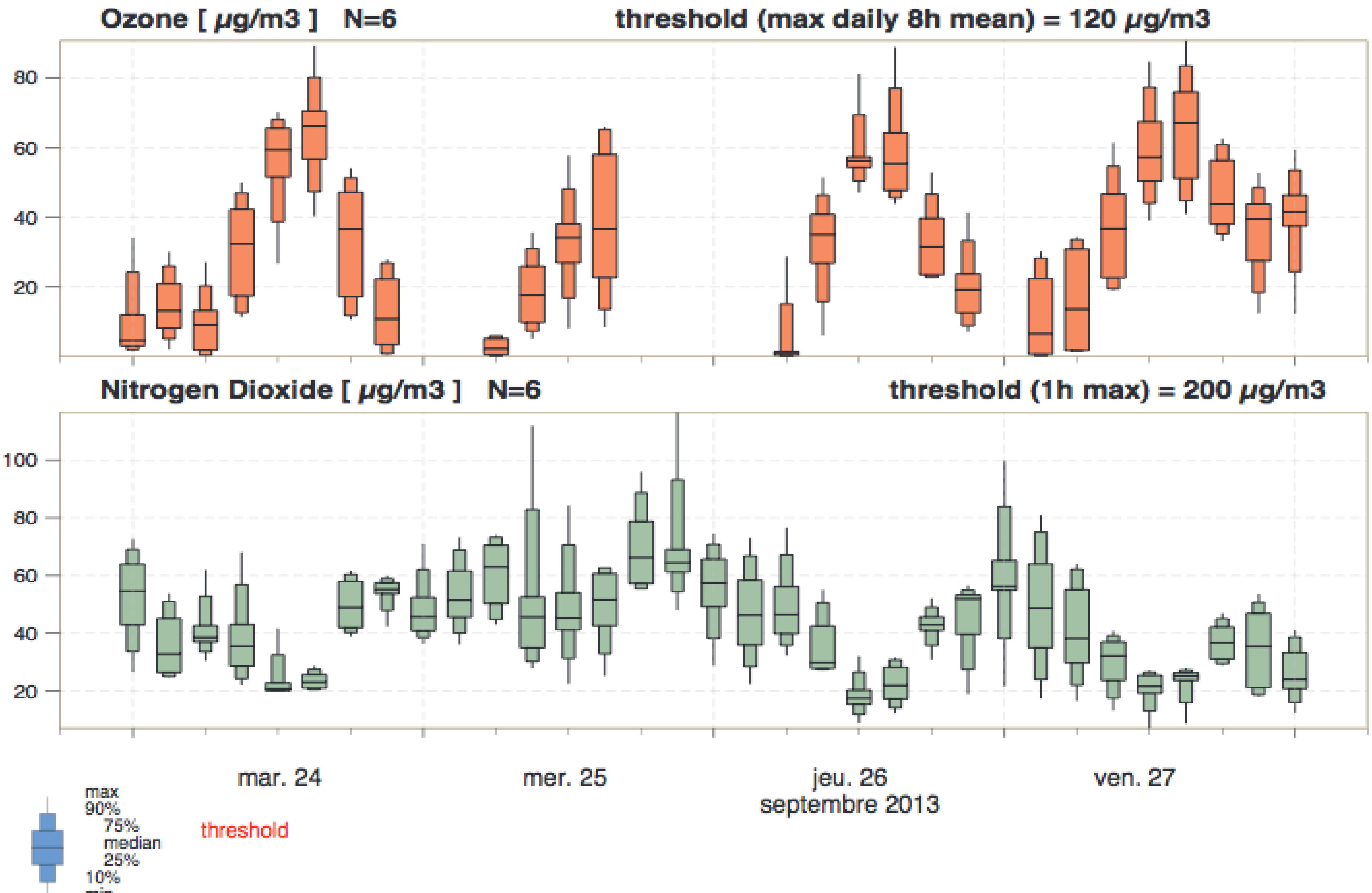
MACC RAQ EPSGRAM
Bruxelles(50.84°N, 4.37°E)
Forecast mardi 24 septembre 2013 00 UTC



A last word….

- Over the last 100 years, Belgium and specifically the Royal Meteorological Institute have played a key role in support of the international research in atmospheric sciences.
- The success must be attributed in large part to the support provided to world-class scientists.
- A world-class reputation requires a clear and long-term commitment to research, including fundamental aspects.

HAPPY BIRTHDAY, RMI !