PAPILA Summer School 2020

Historical Introduction

Guy P. Brasseur

Air Pollution in Latin America

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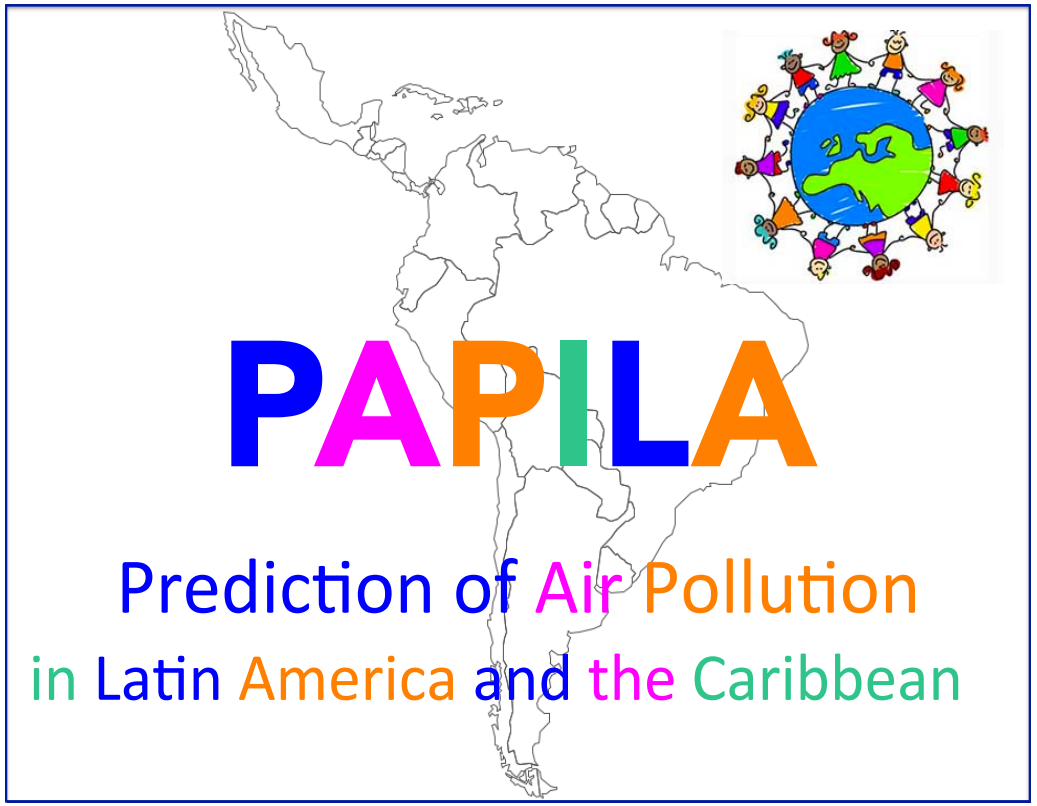
European Commission



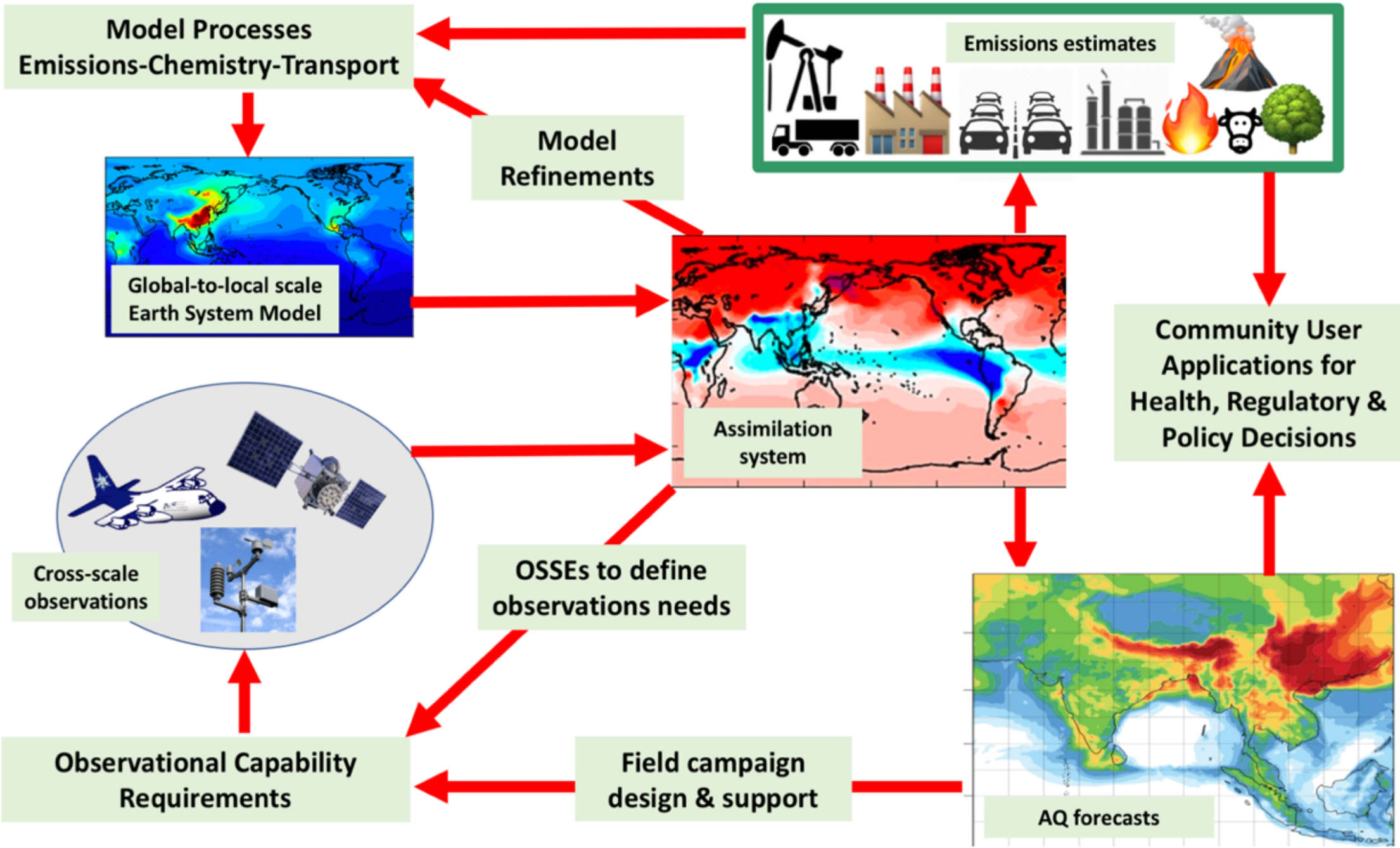
Prediction of Air Pollution in Latin America and the Caribbean

The **overall objective** of the PAPILA project is

- to establish a sustained network of partners with complementary expertise that develops and implements an **analysis and forecast system for air quality** with downscaling capability for Latin America and the Caribbean region;
- to assess the **impact of air pollution** on health and on the economy.

This system will

- help decision-makers improve air quality and public health, and
- avoid the occurrence of acute air pollution episodes, particularly in urban areas.


Model Processes
Emissions-Chemistry-Transport
Emissions estimates
Model
Refinements
Global-to-local scale
Earth System Model
Assimilation
system
Community User
Applications for
Health, Regulatory &
Policy Decisions
Cross-scale
observations
OSSEs to define
observations needs
Observational Capability
Requirements
Field campaign
design & support
AQ forecasts

Specific Objectives

- Real-time state-of-the-art multi-model quasi-operational air quality forecast system
- Real-time chemical data system that collects observations of air pollutants
- State-of-the-art data assimilation system
- Emission inventory (bottom-up and top-down)
- Dissemination platforms that will release daily “chemical weather” forecasts
- Transfer of data to national and international agencies

PAPILA
Prediction of Air Pollution
in Latin America and the Caribbean

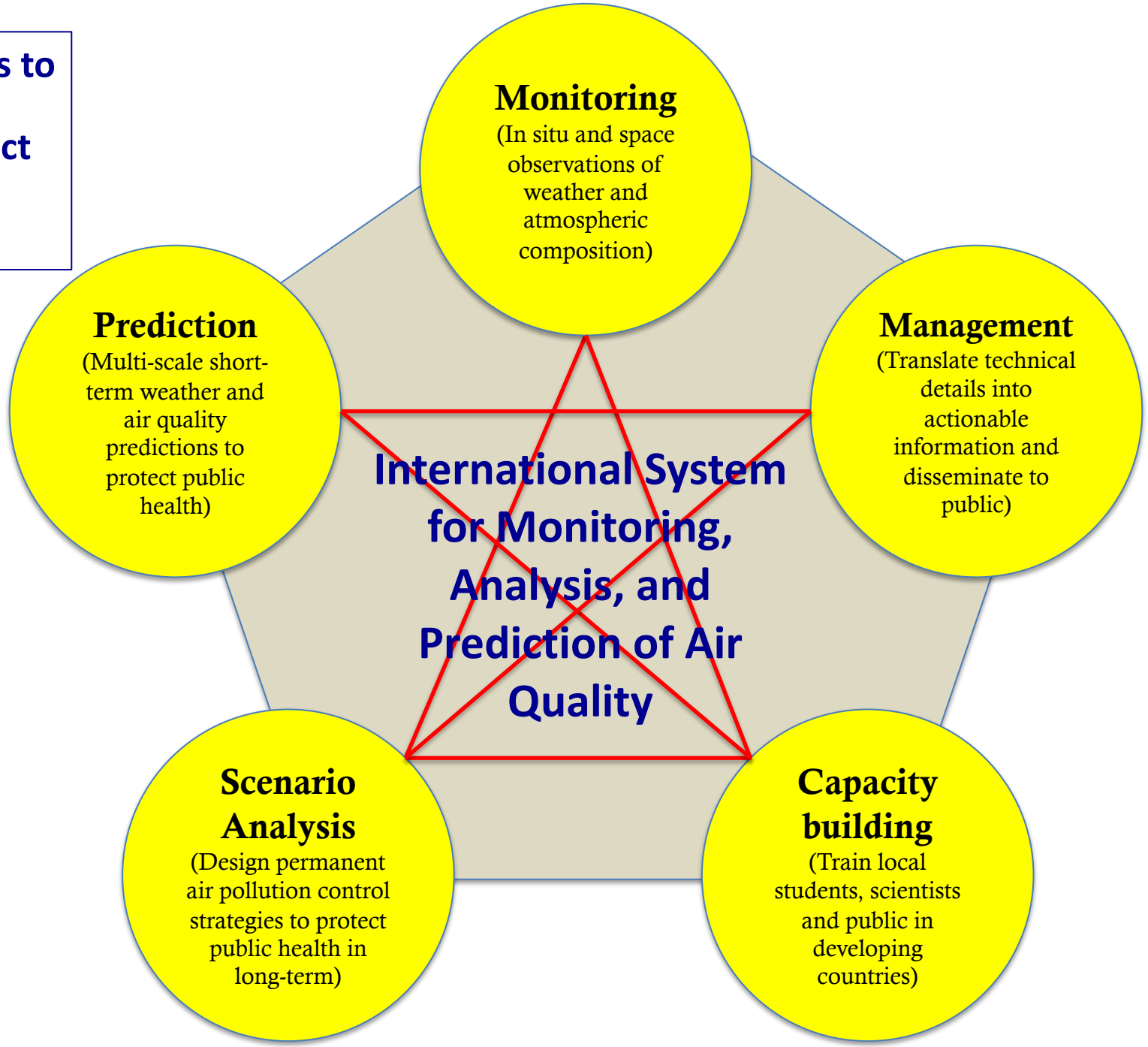
The Partners

MPG Germany	UCL Chile
CNRS France	USP Brazil
MET Norway	UNAL Colombia
FMI Finland	CNEA Argentina
INERIS France	UMSA Bolivia
KNMI Netherlands	USM Chile
SU Sweden	IVIC Venezuela
UGA France	UNAM Mexico
TROPOS Germany	UPR-RP Puerto Rico

The System will be used

- To provide daily ***deterministic and probabilistic air quality predictions*** and related air ***quality health indices*** in Latin America and the Caribbean region with spatial resolutions that will be increased in densely populated areas and other hotspots.
- To ***determine*** and ***release forecasts*** of regional distributions of primary chemical , and secondary species and health indices.

PAPILA contributes to the MAP-AQ Project of WMO and IGAC



MAP-AQ

Monitoring, Analysis and Prediction of Air Quality

What is a model?

A model: A simplified representation of a process or of a system.

A mathematical model: a representation of the essential aspects of an existing system.

A mathematical model: a description of a system using *mathematical* concepts and language to facilitate proper explanation of a system or to study the effects of different components and to make predictions on patterns of behavior.

Belgian surrealist painter René Magritte with just a few words probably best highlighted what a model is.

Mathematical Models

- Mathematical models are precise mathematical constructs developed to answer questions, to explore ideas, and to help gain insights about a system.
- Mathematical models allow us to access quantities that may not be directly observable, but nonetheless important to understand a problem.
- Models are the ultimate for of a quantification since all variables and parameters must be properly defined and quantified for the equations to make sense.

The 1920’s and 1930’s

The Dobson Ozone Photographic Spectrometer of Gordon Dobson at Oxford, UK.

Gordon Dobson

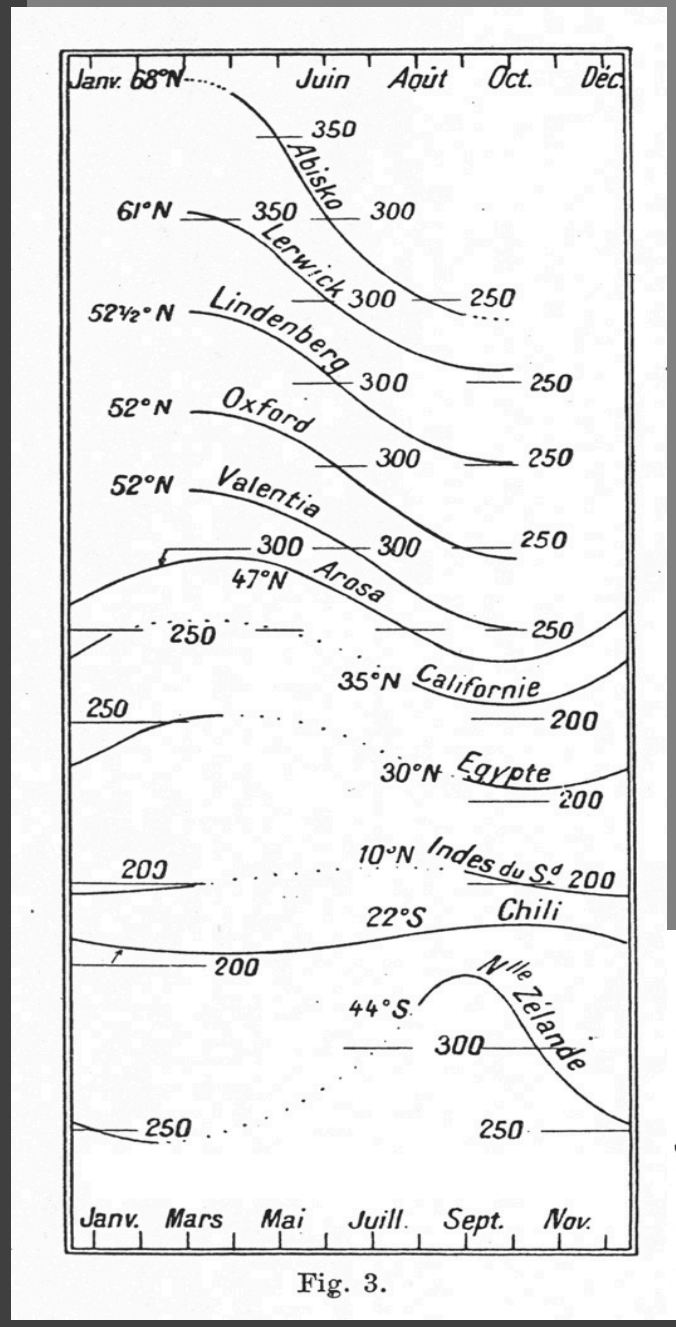
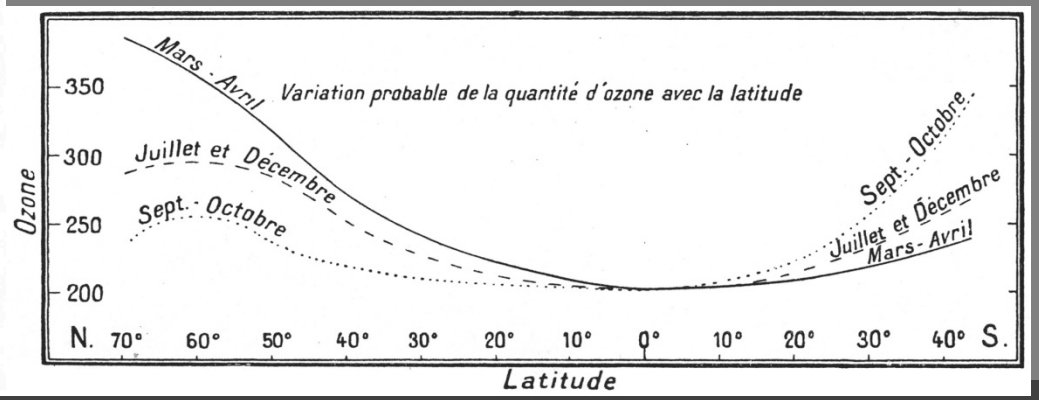
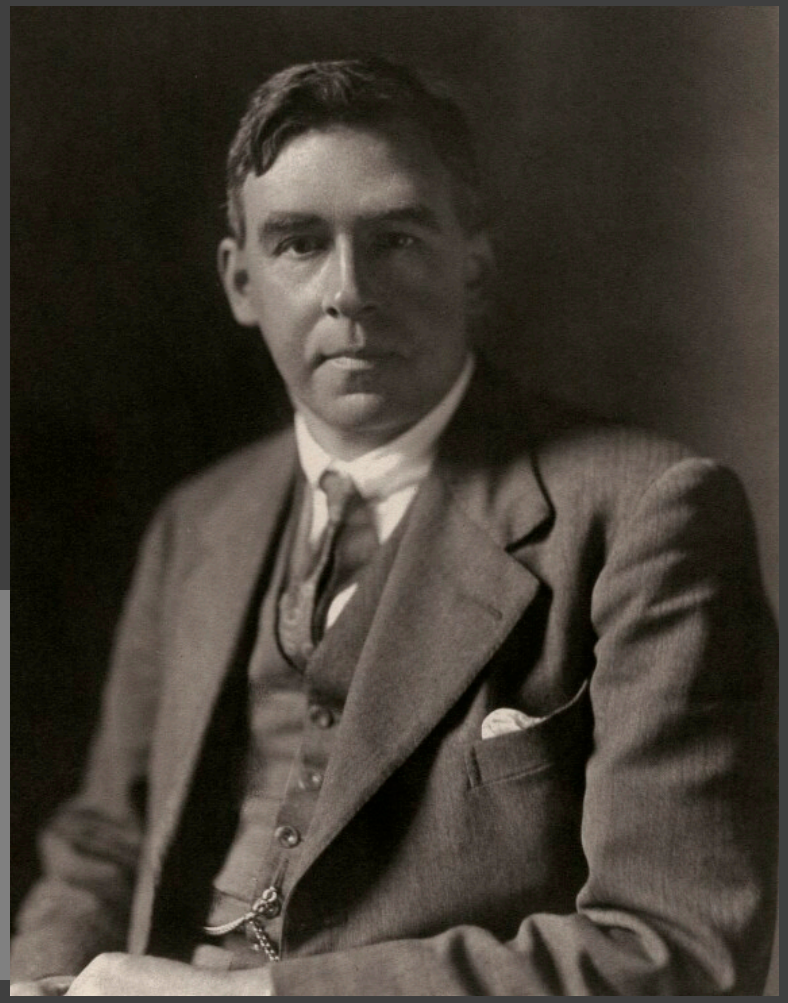


Fig. 3.





The First Ozone Conference in Paris (1929)

- At the first ozone conference in Paris, **Sydney Chapman,** a specialist of solar terrestrial relations, rather than invoking high-latitude auroral physics to explain Dobson observations, indicates that the formation of ozone probably results from the photolysis of molecular oxygen by solar ultraviolet radiation:

$$O_2 + UV \rightarrow O + O$$

- His theory does not explain the polar maximum in the ozone column.
- But his theory seems to confirm the early observations of Götz that the ozone layer is located near **40 km** altitude.

Sydney Chapman

The Chapman Scheme (pure oxygen atmosphere) constitutes the basis for the first photochemical models of the atmosphere

• (1) $O_2 + h\nu \rightarrow O + O$	Net production
• (2) $O + O + M \rightarrow O_2 + M$	Recombination of O at high levels
• (3) $O + O_2 + M \rightarrow O_3 + M$	Conversion of O into ozone
• (4) $O_3 + h\nu \rightarrow O_2 + O$	Conversion of ozone into O
• (5) $O_3 + O \rightarrow O_2 + O_2$	Net loss of O and ozone

1936: Wulf and Deming develop the first model of stratospheric ozone

THE THEORETICAL CALCULATION OF THE DISTRIBUTION OF PHOTOCHEMICALLY-FORMED OZONE IN THE ATMOSPHERE

By Oliver R. Wulf and Lola S. Deming

Absorption cross section of O_2 and O_3

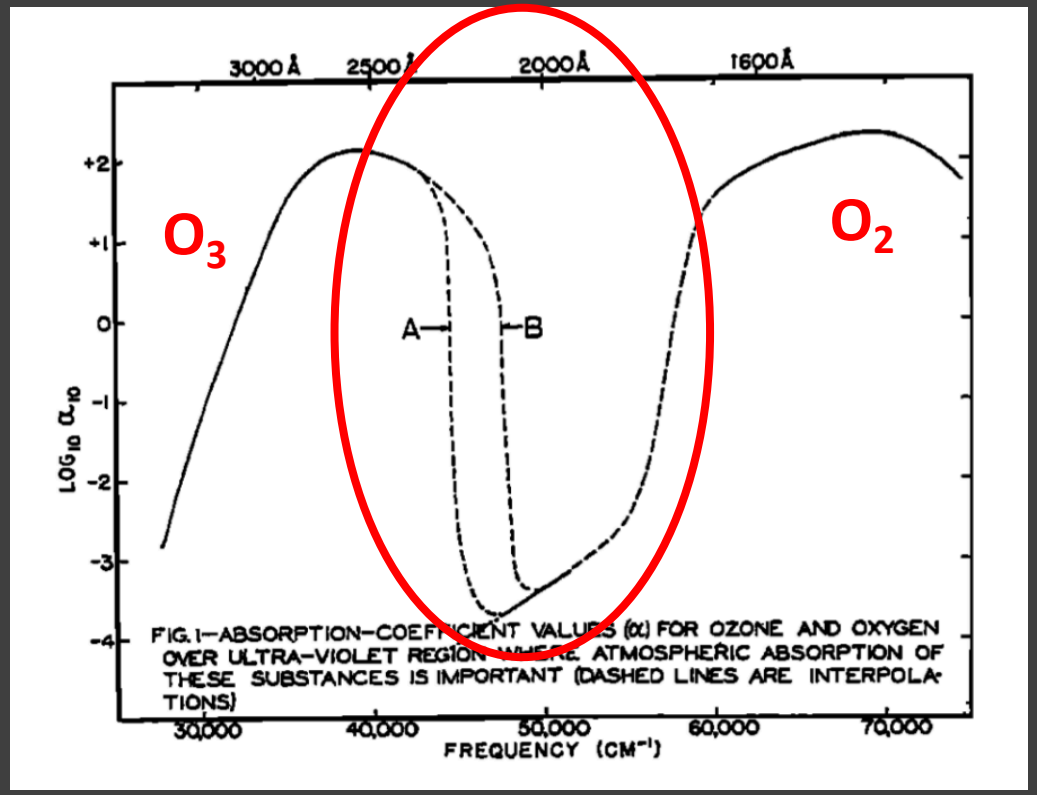
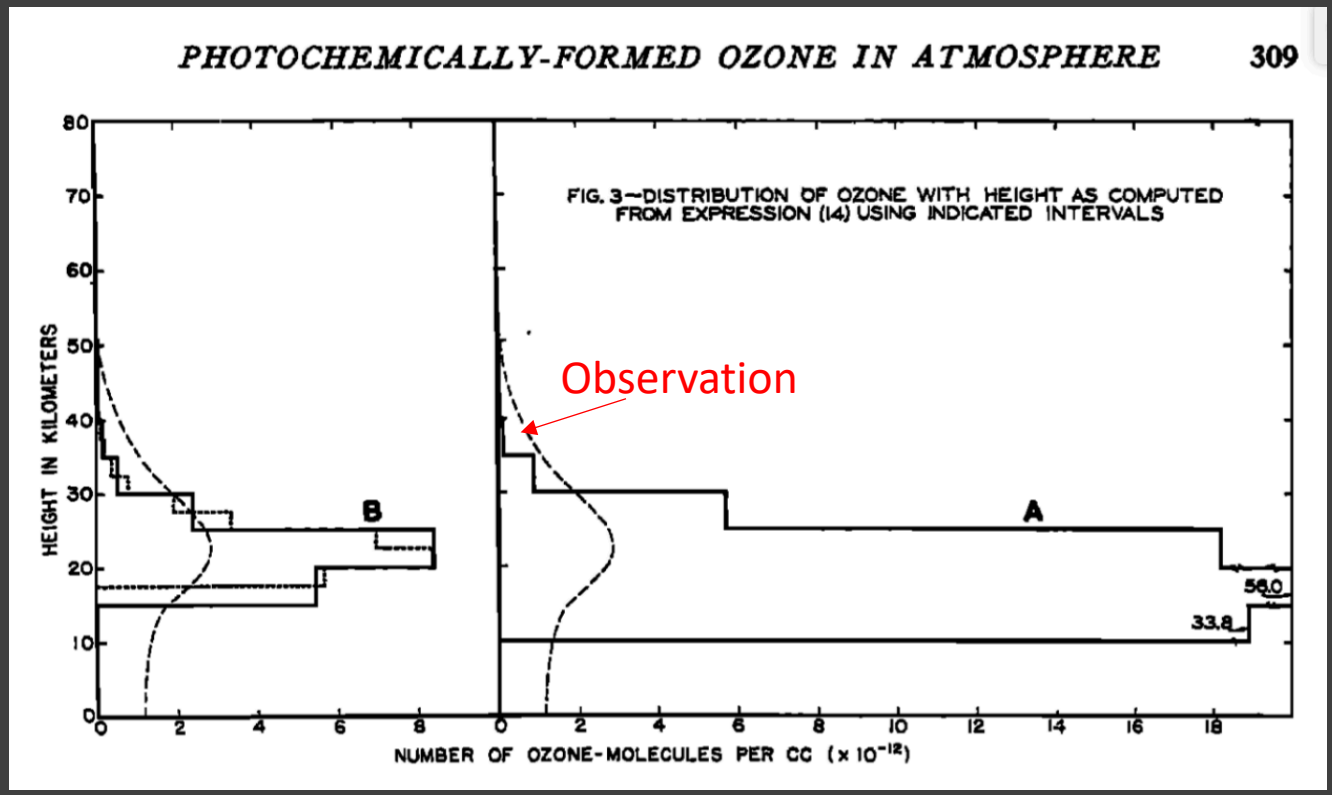


FIG. 1—ABSORPTION-COEFFICIENT VALUES (α) FOR OZONE AND OXYGEN OVER ULTRA-VIOLET REGION WHERE ATMOSPHERIC ABSORPTION OF THESE SUBSTANCES IS IMPORTANT (DASHED LINES ARE INTERPOLATIONS)

Calculated vertical distributions
The dashed curve refers to the observations



A Theory of Eddy Diffusion in the Atmosphere.

By O. G. Sutton, B.Sc.

(Communicated by G. C. Simpson, F.R.S.—Received October 19, 1931.)

1. *Introductory.*

The theory of eddy diffusion in the atmosphere put forward almost simultaneously by G. I. Taylor* and L. F. Richardson† in England and by W. Schmidt‡ in Austria is a direct generalisation of the classical theory of molecular diffusion. It is assumed that the mass effect of the eddies is entirely similar, except for a scale difference, to that of the molecules; thus we find an eddy-diffusivity of the order of 10^2 to 10^{11} cm.^{2}/sec. replacing a molecular diffusivity of the order of 10^{-1} cm.^{2}/sec. in entirely similar differential equations. Recent researches,§ however, have shown that the difference between the eddy structure of a turbulent fluid and the molecular structure of a fluid at rest is more than one of scale, and it is now clear that there is need of an extended theory to express this difference.

1931: The first step towards air pollution modeling:

Dispersion of pollutants can be represented by eddy diffusion.

It mimics molecular diffusion

1936: Industrialization is a source of air pollution:

The concept of eddy diffusion is used to simulate the dispersion of pollutants released by chimneys

THE SPREAD OF SMOKE AND GASES FROM CHIMNEYS.

By C. H. Bosanquet, M.A., and J. L. Pearson, B.A., Ph.D.

Received 11th March, 1936.

I. Introduction.

The authors have dealt elsewhere[1] with the following theoretical and practical aspects of the spread of chimney emissions :—

(*a*) a physico mathematical analysis of the phenomena involved in eddy diffusion ;

(*b*) space-concentration distribution and mass-over-area distribution of emission from point and from line sources ;

(*c*) the effects of chimney height on space-concentration and mass-over-area distributions.

Mass-over-area distribution of atmospheric pollution from chimney emissions is of practical importance, since it determines :—

(*a*) in rainy weather, the amount of acid and dust brought down by the rain,

(*b*) in fine weather, the extent of the curtailment of sunshine and ultra-violet rays.

[1] *J. Inst. Elec. Eng.*, July, 1935, and *J. Inst. Fuel*, April, 1935.

A Gaussian Plume Model

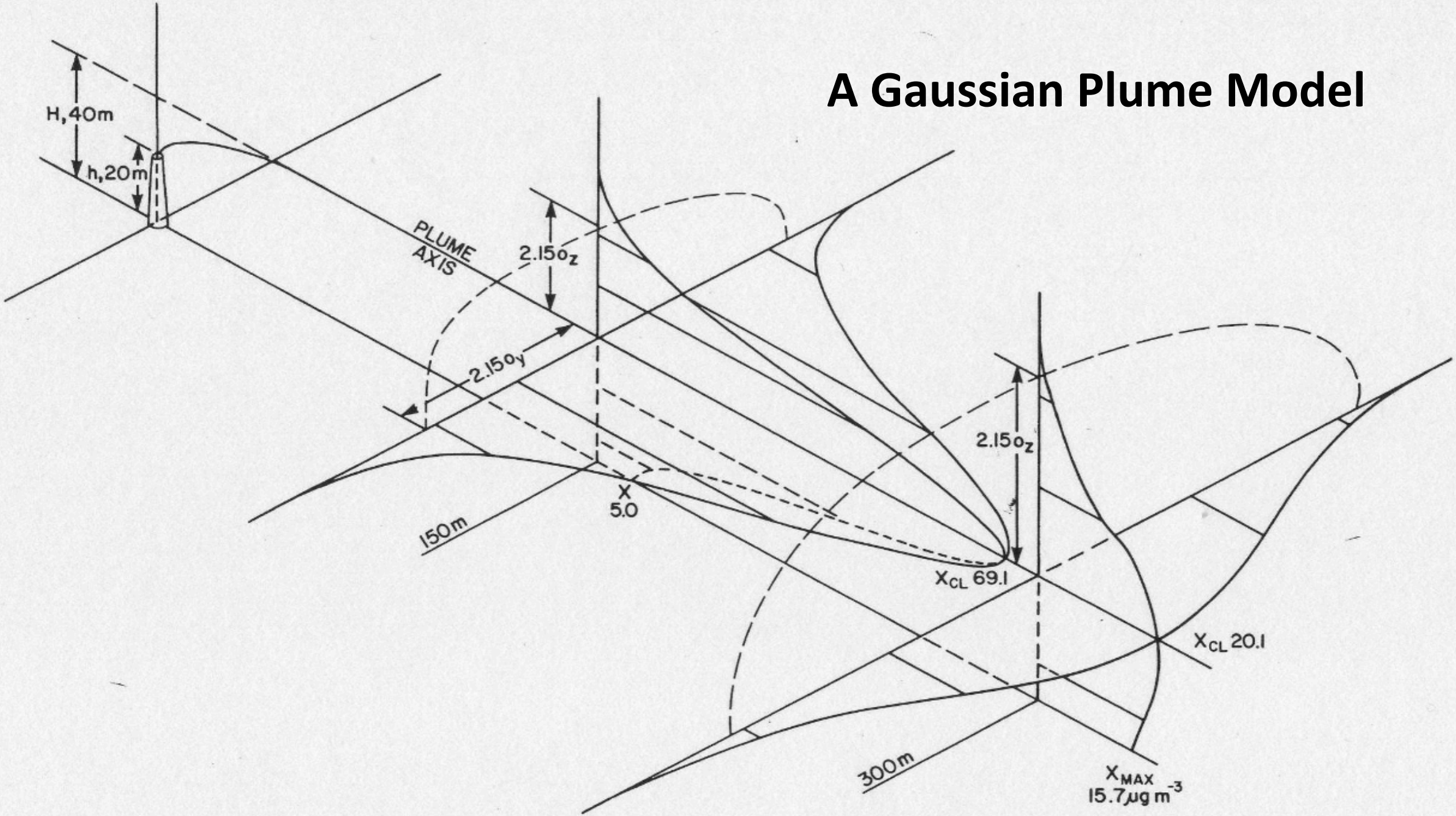


Fig. 19-5. Two cross sections through a Gaussian plume (total mass under curves conserved).

Today's Dispersion Models

AERMOD: http://www.epa.gov/scram001/dispersion_prefrec.htm#aermod

Steady-state Gaussian plume model. The wind field is derived from surface, upper-air, and onsite meteorological observations. Combines geophysical data such as terrain elevations and land use with the meteorological data to derive boundary layer parameters. •

CALPUFF: http://www.epa.gov/scram001/dispersion_prefrec.htm#calpuff

Non-steady state Lagrangian puff dispersion model. Realistically simulates the transport of substances in calm, stagnant conditions, complex terrain, and coastal regions with sea/land breezes.

The 1940’s and the 1950’s

1950: Catalytic Ozone Destruction by Hydrogen Radicals

- Sir **David Bates** (Belfast) and Baron **Marcel Nicolet** (Brussels), working together at Caltech in Pasadena, 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.*

- $O_3 + H \rightarrow OH + O_2$
- $O + OH \rightarrow H + O_2$

- $O + O_3 \rightarrow O_2 + O_2$ (net)

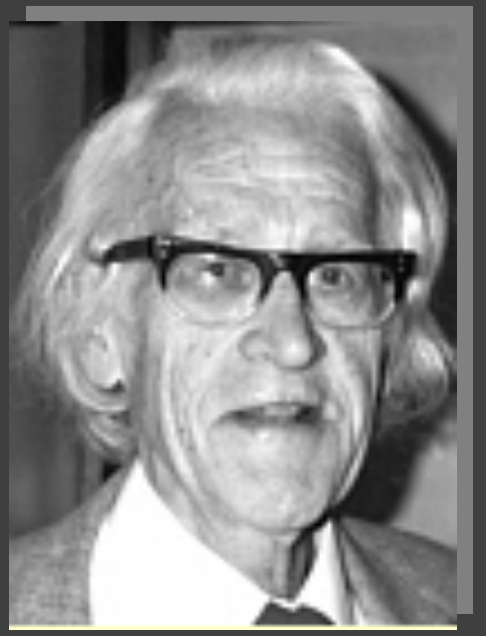
$OH + O \rightarrow H + O_2$

$H + O_2 + M \rightarrow HO_2 + M$

$HO_2 + O \rightarrow OH + O_2$

$O + O \rightarrow O_2$ (net)

Bates



Nicolet

The 1960’s

1960's:
The models based on the Chapman theory considerably overestimate ozone in the entire mesosphere and stratosphere.

The correction introduced by Bates and Nicolet (1950) did not reduce the bias in the stratosphere.

Barrie Hunt indicates that the reaction $O_3 + O(^1D)$ could be of importance in the atmosphere, but notes that the photochemical O_3 problem is left unresolved.

The Need for a Modified Photochemical Theory of the Ozonosphere

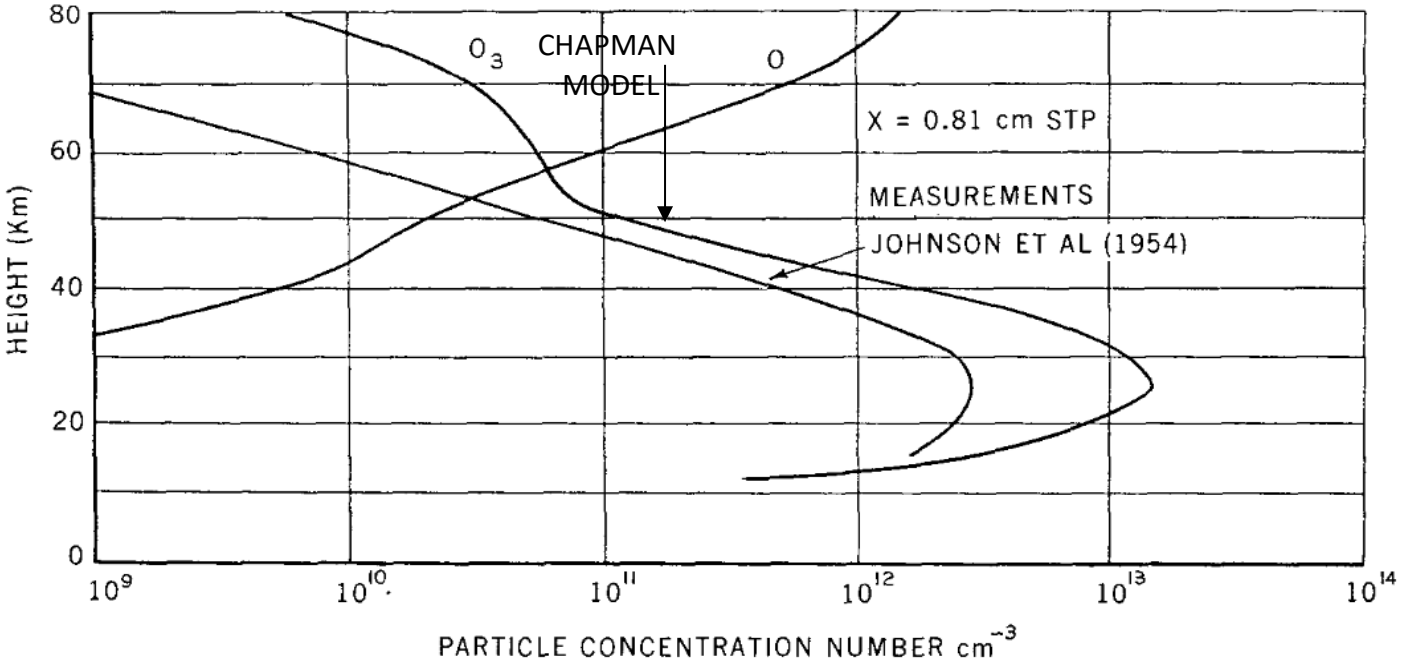
B. G. HUNT[1]

Weapons Research Establishment, South Australia

(Manuscript received 11 March 1965, in revised form 30 August 1965)

ABSTRACT

It is shown that when rate constants confirmed by recent laboratory studies are used in photochemical O_3 calculations unacceptably high O_3 concentrations and total O_3 amounts are obtained. In order to account for this disagreement, an investigation has been made to see whether reactions between O_3 and excited forms of molecular and atomic oxygen are of importance in the atmosphere, following recent laboratory work in this field. It was found that excited molecular oxygen may be neglected in the O_3 reaction scheme but that reactions between O_3 and $O(^1D)$ could be of importance in the stratosphere. The importance of this reaction depends very markedly on the rate of deactivation of $O(^1D)$ in the atmosphere, and a conflict of requirements exists between the O_3 and 6300 Å airglow values for this rate. Hence, in view of this conflict, the photochemical O_3 problem has been left unresolved.



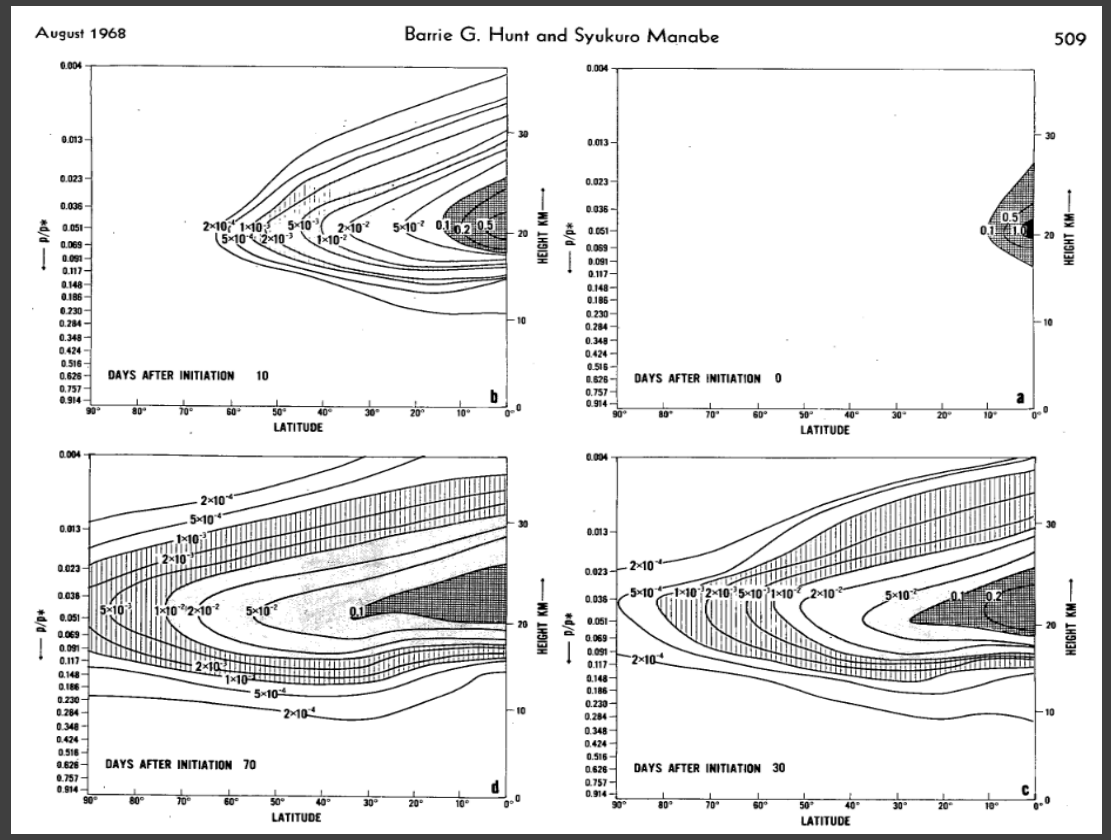
1969: First simulations of ozone in a General Circulation Model (oxygen-hydrogen atmosphere)

The "Ozone problem" is not solved

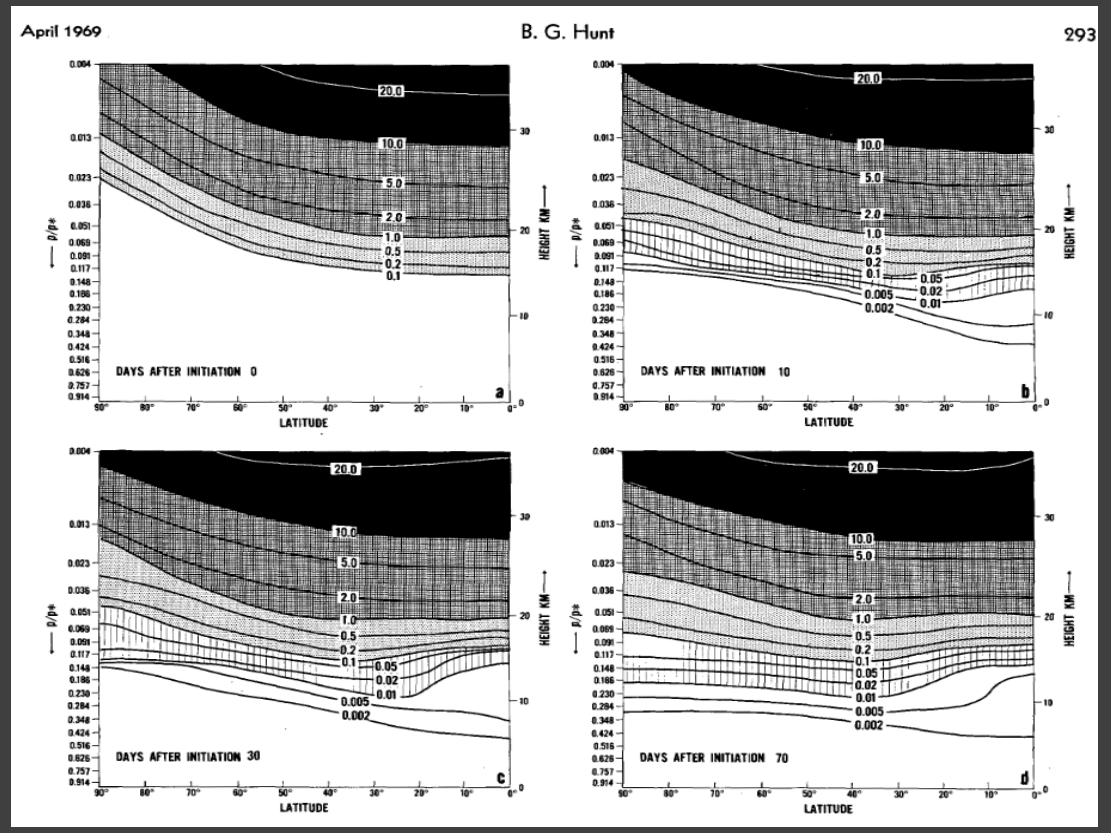
GFDL Model

G. E. Hunt

Inert Tracer mixing ratio



Ozone mixing ratio



The 1970’s and 1980’s

1970: Catalytic Ozone Destruction by Nitrogen Oxides

- **Paul Crutzen** shows that the major ozone loss in the stratosphere is provided by a catalytic cycle involving the presence of nitric oxide (NO)
- Nitric oxide is produced in the stratosphere by oxidation of nitrous oxide (N_2O)

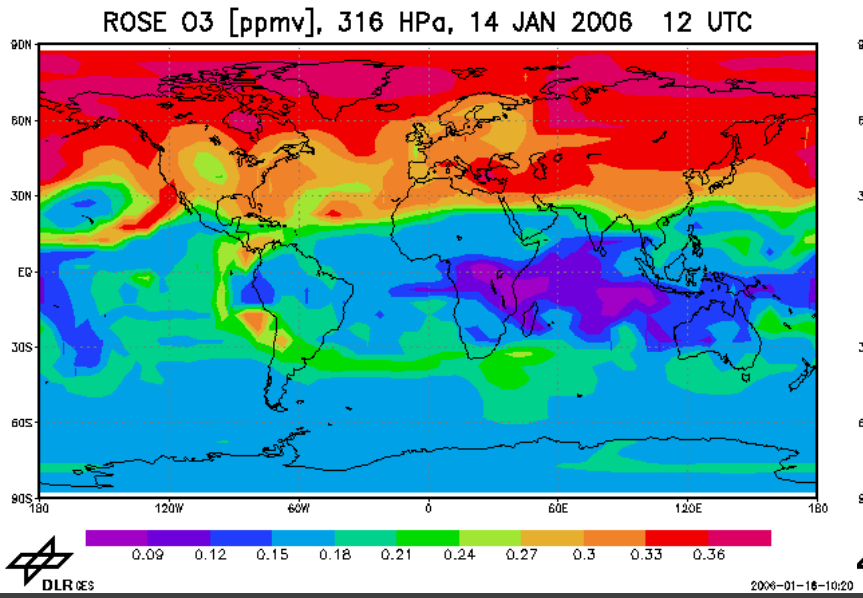
$$NO + O_3 \rightarrow NO_2 + O_2$$

$$NO_2 + O \rightarrow NO + O_2$$

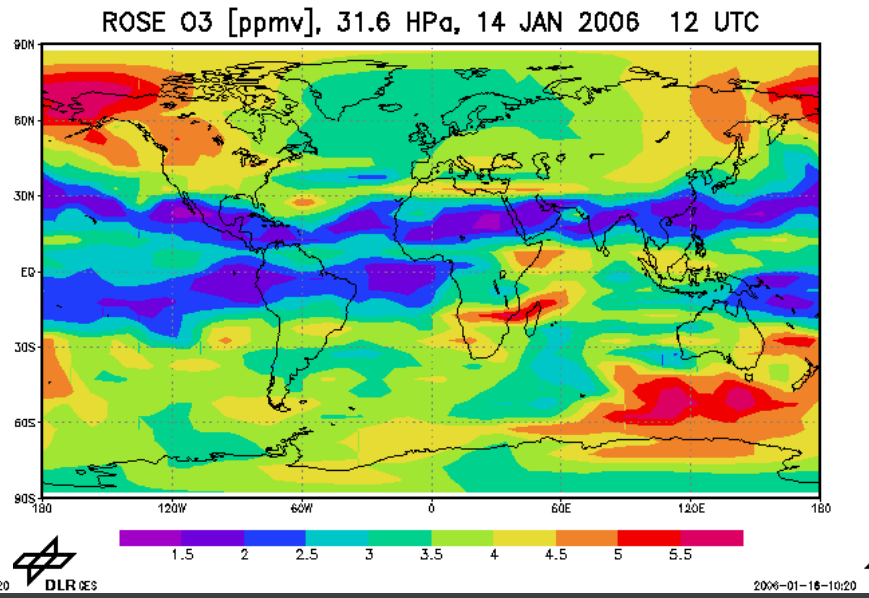
$$O_3 + O \rightarrow O_2 + O_2. \text{ (net effect)}$$

The ROSE Model developed in the 1980's

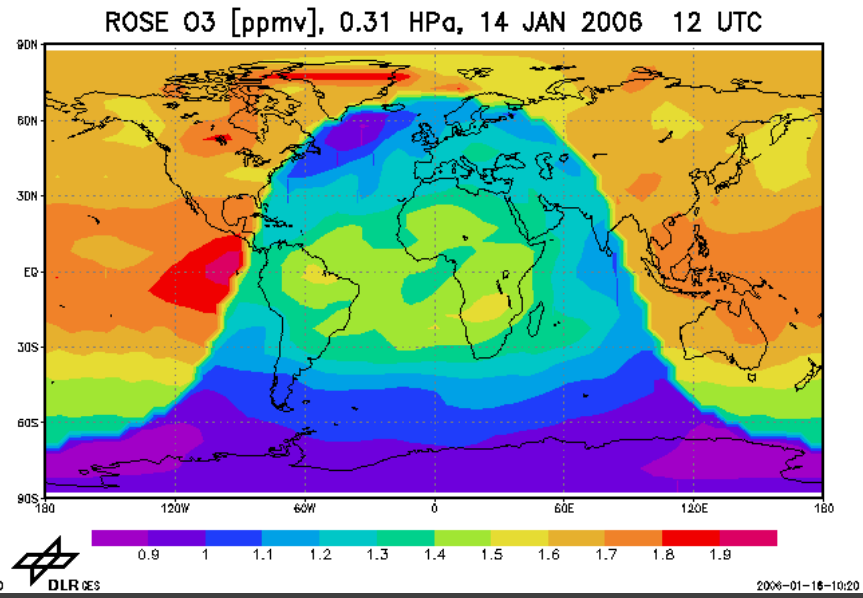
Simple three-dimensional model of ozone and related chemical species



316 hPa



31.6 hPa.



0.31 hPa

Air Pollution is more than a local problem

- In Europe: Shortly after 1970, scientists realized that air pollution is not only a local phenomenon. SO_2 and NOx emissions from tall stacks could lead to acidification at large distances from the sources. (acid rain)

- In the United States, scientists realized that ozone is a problem in urbanized and industrialized areas.

- These situations cannot be tackled by simple Gaussian-plume type modeling.

1973: Paul Crutzen and William Chameides introduce a photochemical theory for tropospheric ozone

Photochemical reactions initiated by and influencing ozone in unpolluted tropospheric air

By PAUL J. CRUTZEN,[1] *National Center for Atmospheric Research,*[2] *and University of Colorado, Boulder, Colorado 80302*

Observational and theoretical evidence in support of a significant in-situ photochemical source of tropospheric ozone

By JACK FISHMAN, *Department of Atmospheric Science, Colorado State University, Ft. Collins, Colorado 80523, U.S.A.*, SUSAN SOLOMON, *National Centre for Atmospheric Research,*[1] *P.O. Box 3000, Boulder, Colorado 80307, U.S.A.* and *Department of Chemistry, University of California, Berkeley, Berkeley, California 94720, U.S.A.*, and PAUL J. CRUTZEN, *National Centre for Atmospheric Research,*[1] *P.O. Box 3000, Boulder, Colorado 80307, U.S.A.*

A Photochemical Theory of Tropospheric Ozone

WILLIAM CHAMEIDES AND JAMES C. G. WALKER

Department of Geology and Geophysics, Yale University, New Haven, Connecticut 06520

$$n(O_3) = \frac{\{n(NO)[n(HO_2)k_{22} + n(CH_3O_2)k_6 + 2n(NO_3)k_{32}] + n(HNO_3)k_{25a}\}F}{f_2[n(H_2O)k_{16a} + n(H_2)k_{16b}] + n(NO)k_{29}(1 - F) + n(NO_2)k_{28}}$$

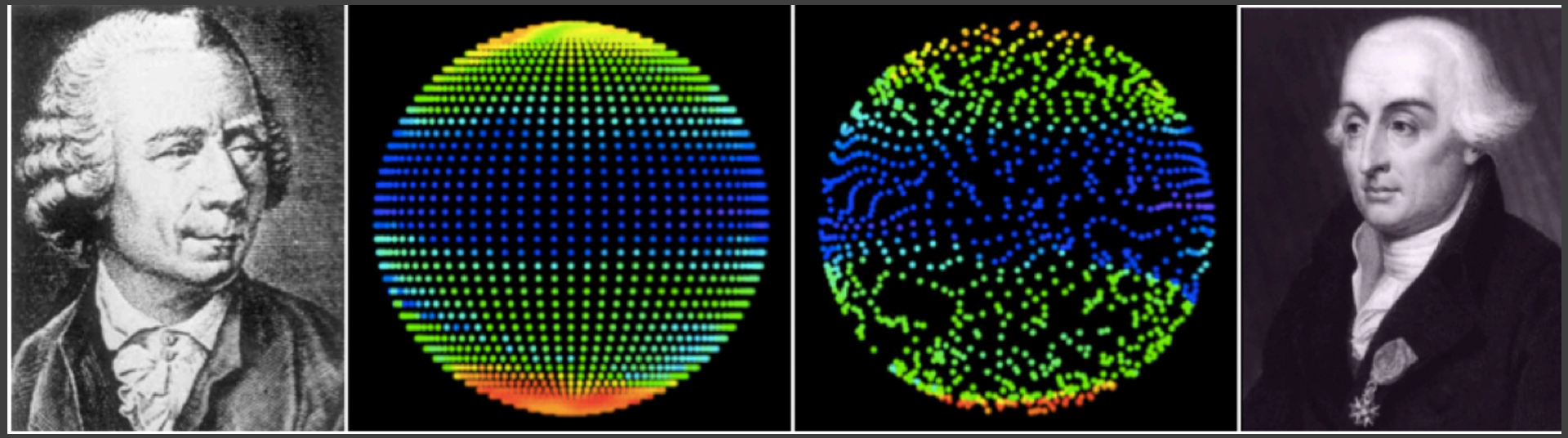
Atmospheric Chemistry Models

- Photochemical models simulate the changes of pollutant concentrations in the atmosphere using a set of mathematical equations characterizing the chemical and physical processes in the atmosphere.

- These models are applied at multiple spatial scales from local, regional, national, and global.

Two Different Modeling Approaches

- In Lagrangian modeling, an air parcel (or “puff”) is followed along a trajectory, and is assumed to keep its identity during its path.
- In Eulerian modeling, the area under investigation is divided into grid cells, both in vertical and horizontal directions



Eulerian or Lagrangian chemical transport models account for

--emissions and deposition

--chemical transformations

--advection

--sub-scale transport

Daniel Jacob, Harvard

Chemical transport model (CTM):

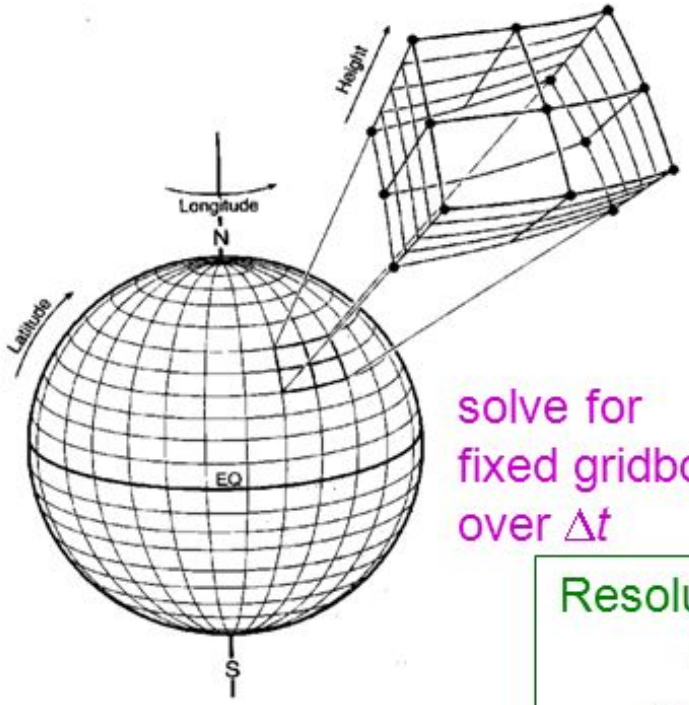
forward model for inverting emissions from atmospheric observations

Solve 3-D continuity equation for chemical concentrations in the atmosphere:

$$\partial C / \partial t = -\mathbf{u}\nabla C + \nabla K \nabla C + P - L + E - D$$

change in concentration with time	grid-resolved transport (advection)	subgrid transport (turbulence)	chemical production and loss	emission, deposition

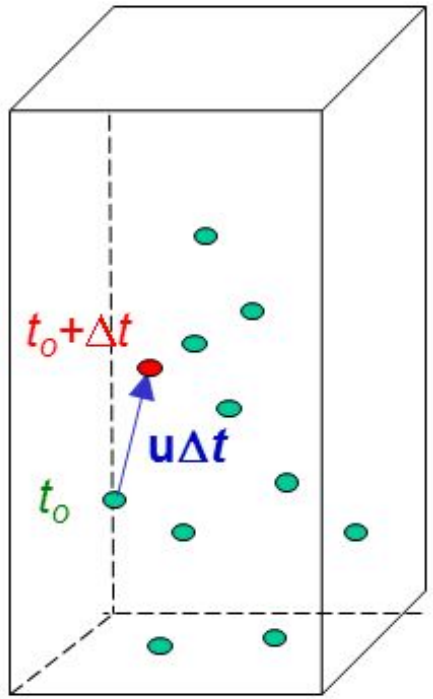
Eulerian framework



solve for fixed gridboxes over Δt

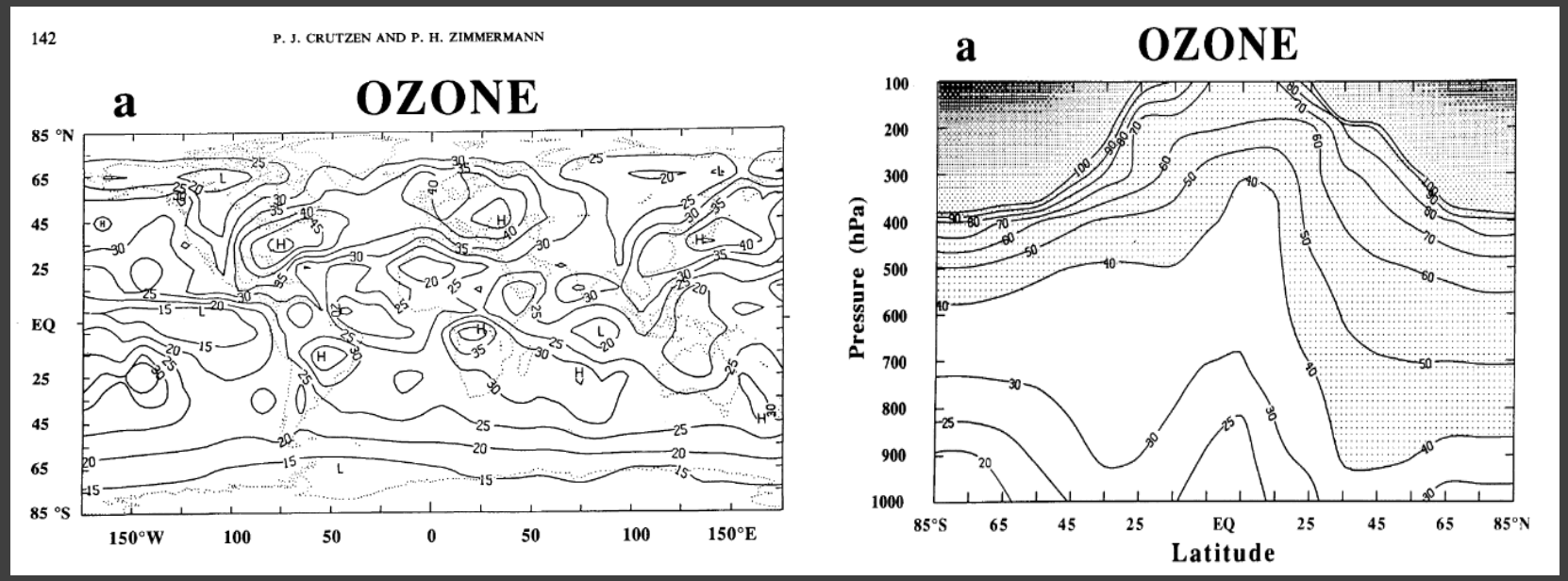
Lagrangian framework

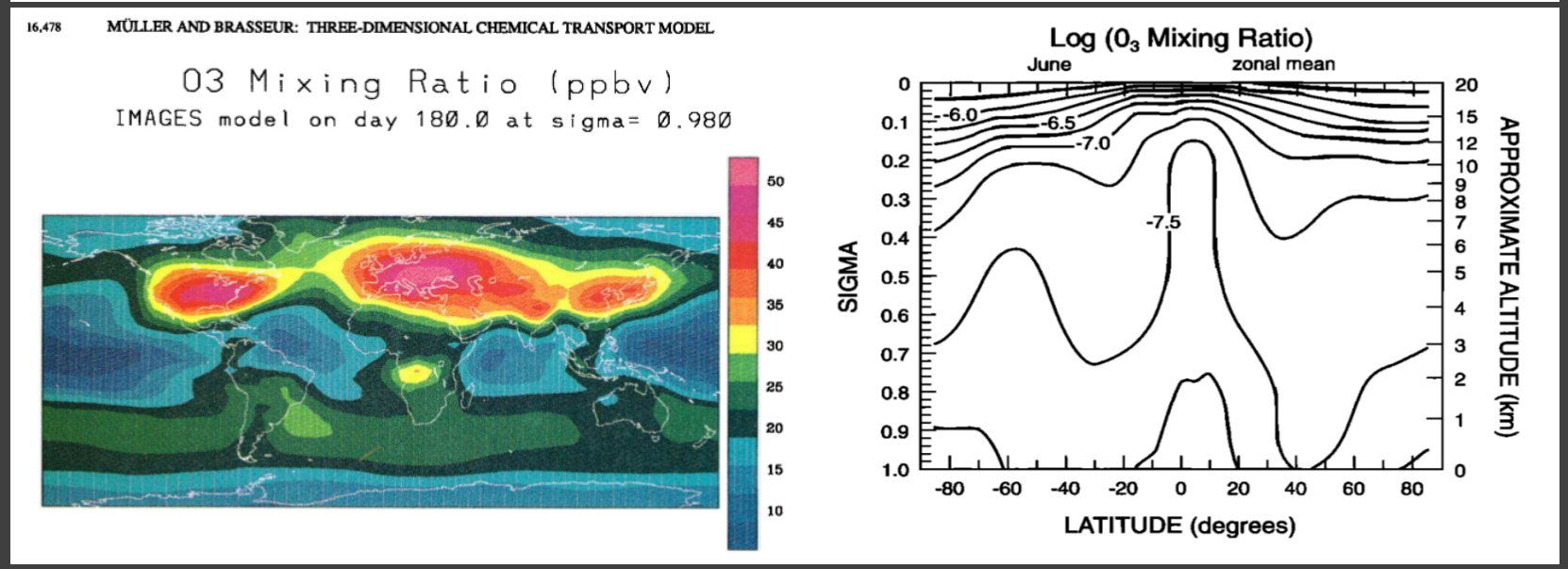
solve for point masses moving with the flow



Resolution of current models:
~100 km (global)
~ 10 km (continental)
~ 1 km (regional)

1990's: Simple 3-D global models of Tropospheric Chemistry



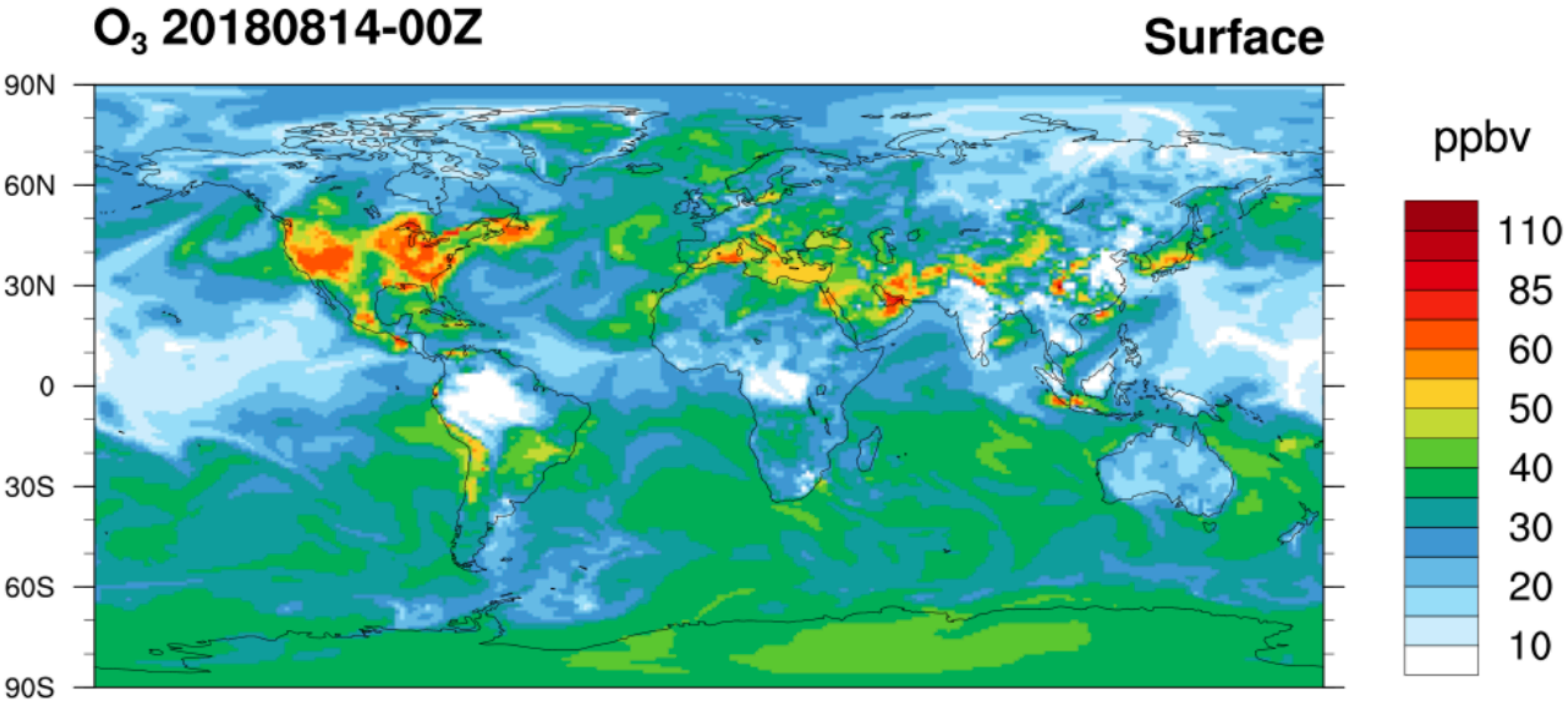


MONGUNTIA Model
MPI-Chemistry, Mainz
Peter Zimmermann

The first low resolution 3-D models use simple meteorology with mean winds and turbulent eddy diffusion, simple chemical schemes. Physical processes are highly parameterized.

IMAGES Model
Belgian Aeronomy Institute, Brussels
J.-F. Müller

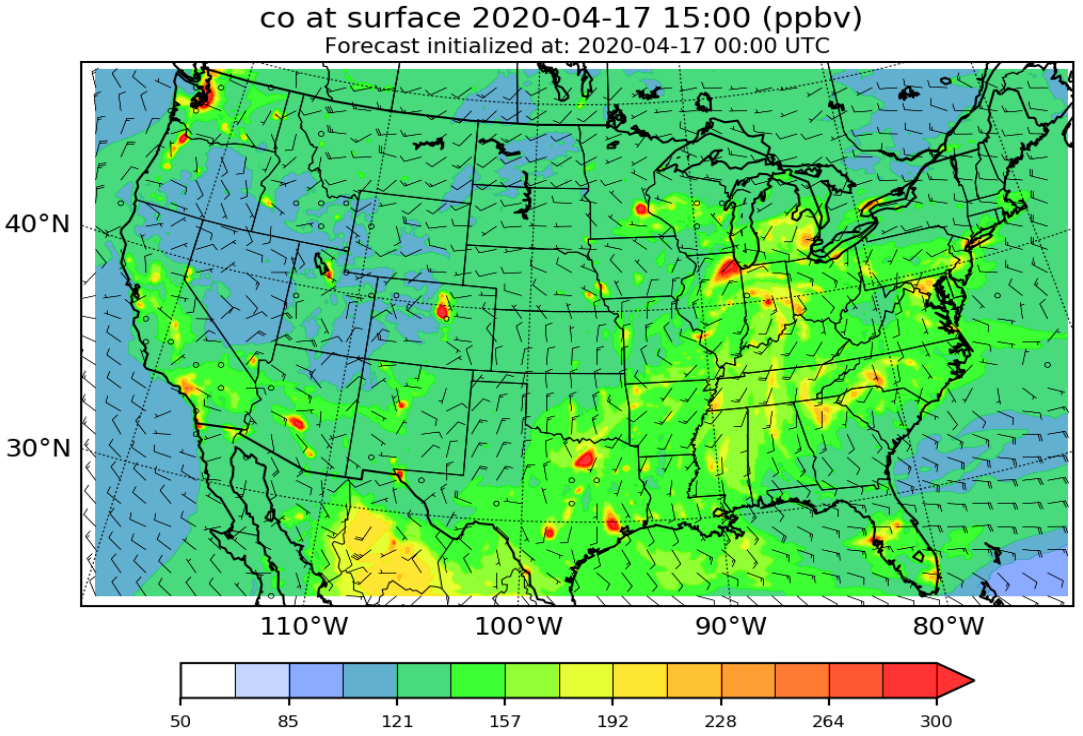
Prediction of surface ozone for 14 August 2018

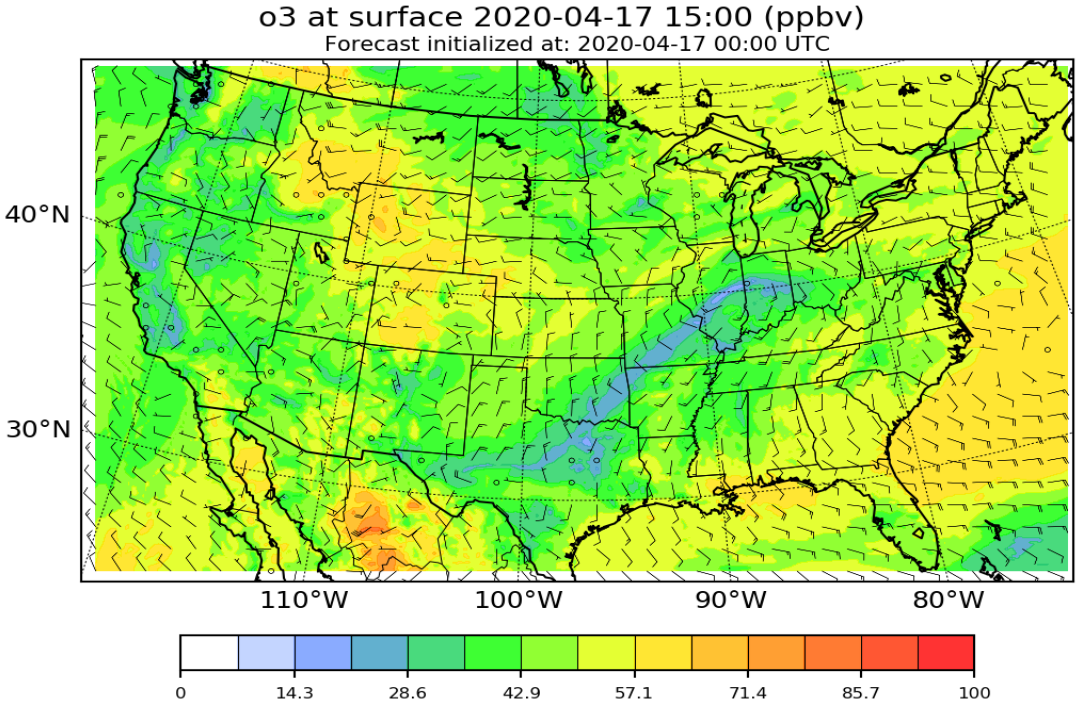


Regional Air Quality Prediction: WRF-Chem

Air pollution photochemical models provide predictions of air quality in different parts of the world.
They have become widely recognized and routinely utilized tools for regulatory analysis and attainment demonstrations by assessing the effectiveness of control strategies.

ACOM NCAR
https://www2.acom.ucar.edu/





Other Regional Air Quality Models

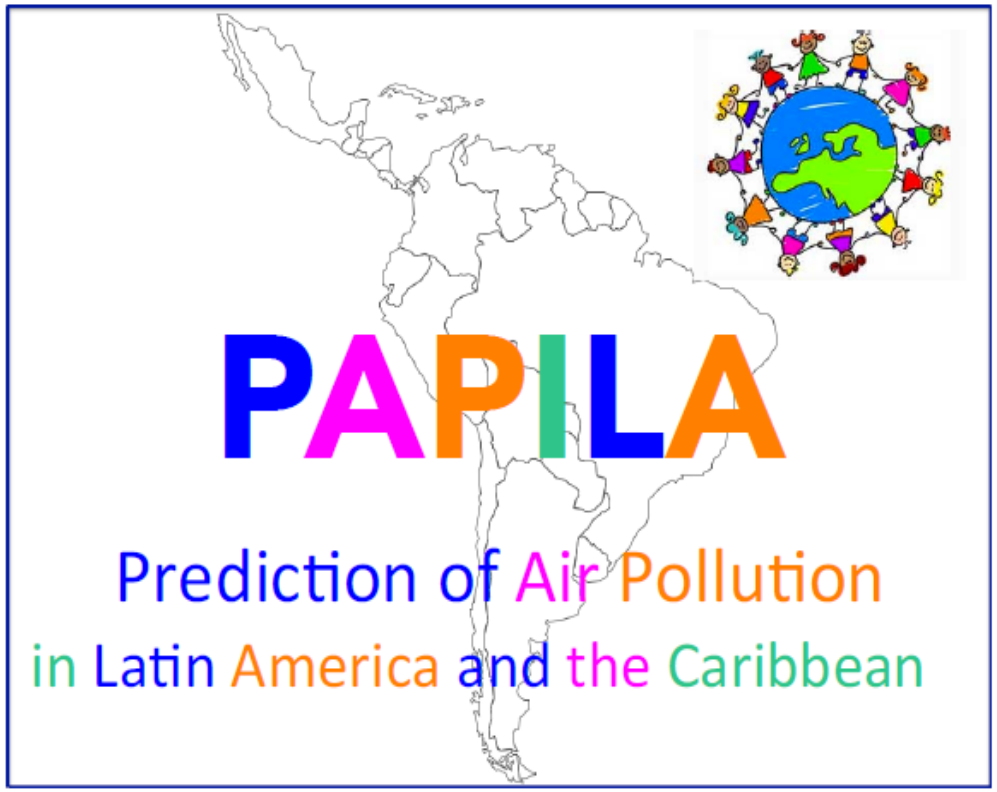
CMAQ: http://www.epa.gov/asmdnerl/CMAQ/index.html

The Community Multiscale Air Quality (CMAQ) modeling system improves

1) air quality management practices for multiple pollutants at multiple scales
2) the scientist's ability to better probe, understand, and simulate chemical and physical interactions in the atmosphere.

CAMX: http://www.camx.com/

The Comprehensive Air quality Model with extensions is a publicly available open-source computer modeling system for the integrated assessment of gaseous and particulate air pollution.



Thank You Very Much

Muchas Gracias

Muito(a) Obrigada