Pollution Plume Transport of Ozone

1,000 to 10,000 km

Robert Chatfield
chatfield@clio.arc.nasa.gov

NASA
(Ames Res. Ctr, California, USA)
with Hong Guan, R. Esswein)
Continental plumes were extensions of urban plumes, which might merge and move out westward in the PBL. Mostly Northern Industrial-Urban Theory was an extension of industrial, urban

Fishman and Vukovich noted plumes could move forward by filling a deep continental PBL and then “override” a thin marine PBL, gaining velocity and isolated from some destruction.
(An early use of TOMS data for tropospheric interpretation.)
When Jack Fishman applied an early technique of subtracting stratospheric ozone,
... northern plumes were visible but the unexpected story was a prominent near-Equatorial ozone maximum, "Jack's Max"
New theory was required to explain this: even current models at 4x5 degree resolution have difficulties. Various explanations including stratospheric ozone came forth. I first favored an explanation involving lofted pollution

1) South America, cloud venting
   ... then
2) Africa, with cloud and PBL venting

Chatfield and Delany, 1990
Chatfield et al, 1996
Figure 2

Chatfield et al., 1996
NASA program 579-24-13-10
Components of a Photochemical Simulation

**Radiative Transfer (uv)**

- $hv, v > 295$ nm (UV-B wavelengths)
- $O_3 + hv \rightarrow O(\Delta) + O_2$

- Ozone absorption mainly in the stratosphere
- Scattering by aerosols + clouds in the troposphere,
  (% clouds??)
- Absorbing molecule spectrum, quantum yield
- Lower tropospheric turbulent mixing and venting to upper air

**Emissions Parameterization**

- Biogenic: isoprene, soil NO
- Anthropogenic: internal combustion
- Combined: biomass burning, CH4

**Meteorology**

- Competition of vertical-horizontal transport processes and transformation
- Frequency, timing, and correlation of most vigorous motions

**Surface Deposition**

- Rainout: e.g., acid rain
- Dry: e.g., $O_3$ ox. of plants

**Analysis ... understanding**

**Thermal and Photochemical Kinetics**

- $O(\Delta) + H_2O \rightarrow OH + OH$
- $OH + CO \rightarrow CO_2 + H \text{ and } H + O_2 \rightarrow HOO$
- $HOO + NO \rightarrow NO_2$
- $NO_2 + hv \rightarrow O + NO$
- $O + O_2 \rightarrow O_3$

**Liquid Phase Ion + Radical Kinetics**

- $O_3 + O_2^- \rightarrow OH + OH + 2O_2$

**Numerical Simulation in Grid Cells**

- Numerical stiffness
- Numerical diffusion

**Robert Chatfield** NASA / Ames R.C. Earth Science
Figure 2

Chatfield et al., 1996
NASA program 579-24-13-10
Make an idealized model of this flow in detail

CO concentrations, 60 to 140 ppb

- A simplified conceptual model useful to explain vertical mixing and transport

Chatfield et al., 1996
NASA program 579-24-13-10
The vertical expression of the tropospheric plume depends on the compound described:
- some compounds follow emission nearly molecule-for-molecule
- some compounds are removed by upward transport in raining clouds
- some compounds are preferentially produced or preserved in the upper troposphere

**Figure 7**

*CO tracer follows each molecule emitted; O₃ emphasizes upper troposphere*
local; downwind

lower stratosphere in upper troposphere

upwind → downwind
Origins of plumes in the Equatorial Atlantic west of Africa

Each weekly sonde has individual peaks, but there is a suggestion of an Atlantic “background” which also varies.

Tracking of peaks suggests lightning of biomass burning sources, … or both (much in accordance with photochemical theory)!
Background ozone also deserves explanation. Why is there an enhanced background? The two blue peaks (i) and (ii) and the red peak (iii) suggest an unexpected answer … … Asian pollution!

?-? (Bio...? (Biomass + Lightning?)

Global Atlantic Backgr. Enhanced Backgr.? Peak Max

Global Atlantic Backgr. Enhanced Backgr.? Peak Max
An analysis of the TTO ozone product along with the Lightning Imaging Sensor product could help explain suggested a W to E transport pattern could explain the highest ozone levels seen in the Ascension Sondes.
By a great stroke of fortune (or planning), there are 3 ozonesondes which illustrate the transport pattern. The sondes are not exactly along the trajectory, but are linked by common transport patterns.
Unexpected Biomass Burning Plumes at ~10000 km; discovered in 1996

Impact of Biomass Burning on the South Pacific

Ozone pollution expressed in the upper troposphere

DIAL Lidar from NASA DC-8

Browell, et al. NASA Langley
Other studies concur ... but it’s hard to distinguish stratosphere (high $O_3$, low CO) from burning influence (high $O_3$ and CO)

Fig. 3. Analysis of CO, $O_3$, and H2O for ER-2 profiles observed during ASHOE-MESA [Folkins et al., 1997]. While the $O_3$ vertical trend is similar to climatology, CO peaks indicate some upper tropospheric $O_3$ is clearly determined by vertical venting of burning emissions.
A simulation of CO indicated origins of the Southern Pacific plumes in large-scale lofting from Africa and South America.

Chatfield et al., 2002
A 3-d image of CO pollution being exported from Africa via mid-latitude convection and a subtropical warm front over S. Africa and teh Indian Ocean.
Origins of this megaplume were from a large synoptic front over South Africa and the Westernmost Indian Ocean.
Show Movie Here
Subtropical Global Plumes also occur in the Northern Hemisphere
PEM-Tropics B
What are the reasons for variability in the free-tropospheric ozone column, especially UT ozone? ... where ozone is an extremely variable, radiatively important trace species.

Upper Tropospheric Ozone is climatically important!

Gettleman et al, 2003 (submitted)
The radiation balance of the tropopause transition layer

See also Mickley et al., JGR, 2001

O₃ variation has strong effects on Upper-Troposphere and Tropopause-Transition Layer structure ... compared to other greenhouse gases and "standard" H₂O effects
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Summary:
Evolution of plume ideas.
- Boundary layer lofting
- Convective lofting
- Warm frontal lofting

Different species have different plumes, upper or lower troposphere.

Importance to global climate