

UC Irvine Developments re Chemistry-Climate WG

Michael Prather
Chemistry-Climate Working Group
26-27 Feb 2007 NCAR

- ▶ **Clouds-Aerosols-Photo & Heterogeneous Chem.**
- ▶ **Chemical model errors – can we quantify them ?**
- ▶ **Strat-trop interactions – what are we missing ?**
- ▶ **Why interactive chemistry-climate coupling ?**

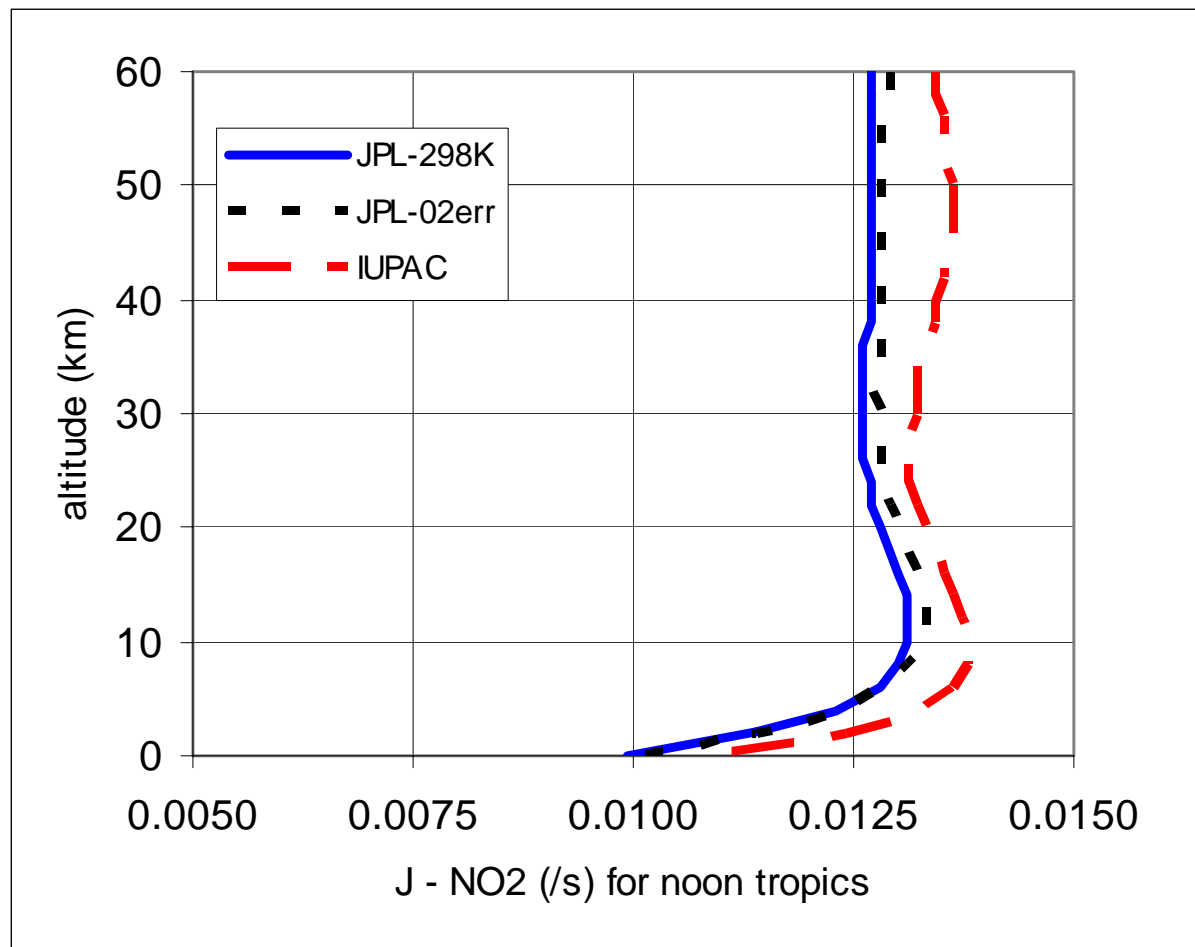
► Clouds-Aerosols-Photo & Heterogeneous Chemistry

With the approach to fractional cloud cover outlined by Jessica Neu, we believe that we can now introduce in-cloud chemistry in a CAM-CTM.

With fast-JX, we should consolidate the short-wave radiation schemes in CAM and CTM, since errors in both radiation and photolysis codes are likely (e.g., Collins analysis of the doubled-CO₂ RF in the AR4 models).

- demonstrate that fast-JX and fast-TUV are identical
- compare these schemes with CAM short-wave

J – NO₂: JPL-02 has T-dep error, IUPAC-05 is 8% larger



► Chemical model errors – can we quantify them ?

At UCI and FRSGC/U.Camb we have tried to evaluate model errors associated with resolution or numerical method.

Wild, O., and M. J. Prather (2006), Global tropospheric ozone modeling: Quantifying errors due to grid resolution, J. Geophys. Res., 111, D11305.

They appear to be of order 10 - 20%.



► **CTM transport errors & 2x-to-convergence**

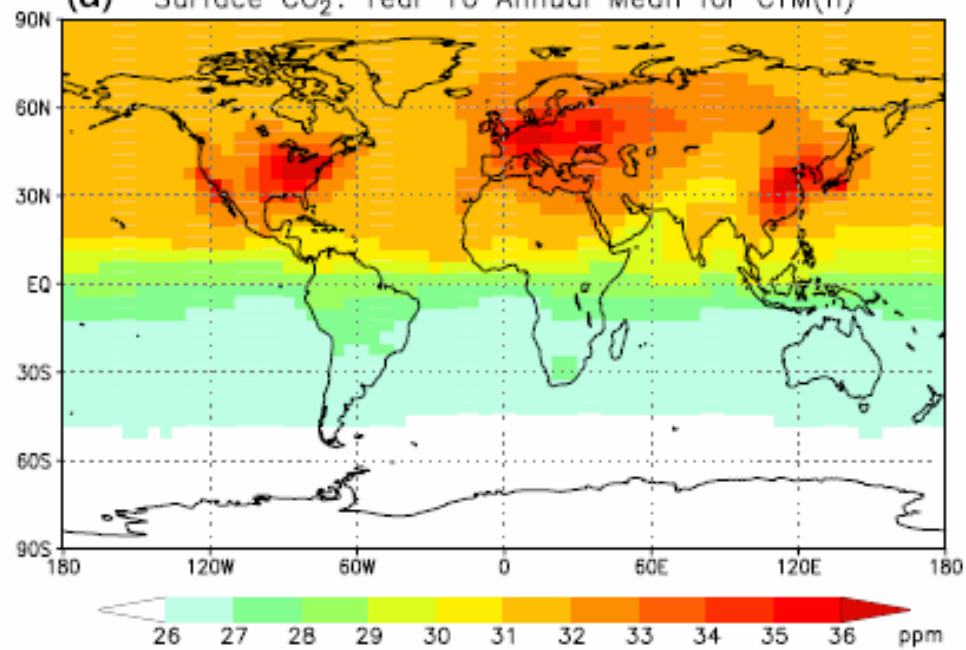
Global tropospheric ozone modeling: Quantifying errors due to grid resolution

Oliver Wild^{1,2} and Michael J. Prather³

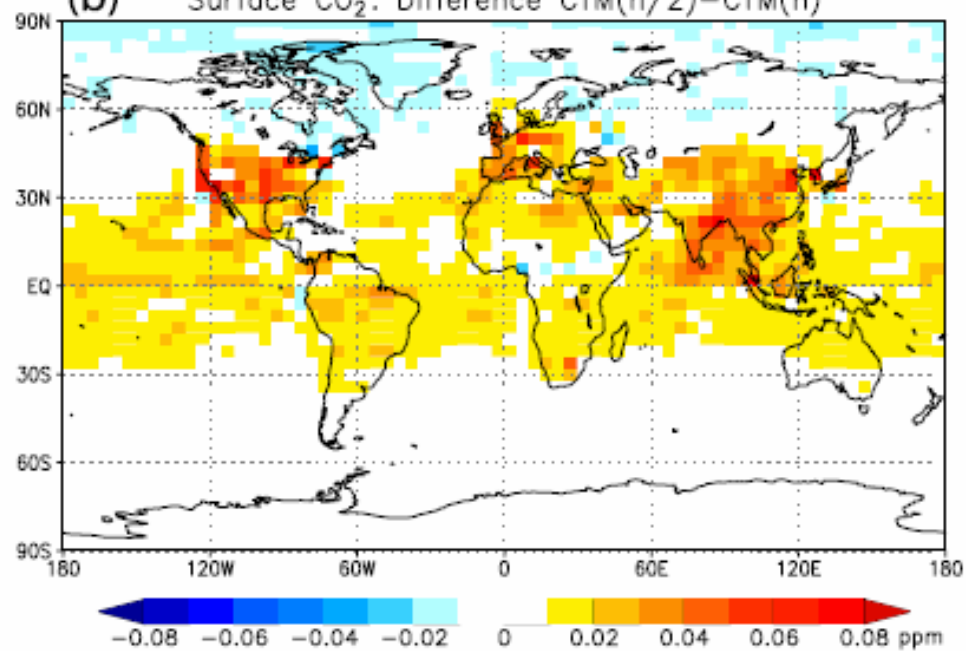
Received 22 August 2005; revised 2 December 2005; accepted 23 February 2006; published 9 June 2006.

[1] Ozone production in global chemical models is dependent on model resolution because ozone chemistry is inherently nonlinear, the timescales for chemical production are short, and precursors are artificially distributed over the spatial scale of the model grid. In this study we examine the sensitivity of ozone, its precursors, and its production to resolution by running a global chemical transport model at four different resolutions between T21 ($5.6^\circ \times 5.6^\circ$) and T106 ($1.1^\circ \times 1.1^\circ$) and by quantifying the errors in regional and global budgets. The sensitivity to vertical mixing through the

(a) Surface CO₂: Year 10 Annual Mean for CTM(h)



(b) Surface CO₂: Difference CTM(h/2)-CTM(h)



ratio of successive corrections being the convergence factor, k .

$$C(h_0) = C(h) + (C(h/2) - C(h)) + (C(h/4) - C(h/2)) + (C(h/8) - C(h/4)) + \dots$$

$$k = (C(h/4) - C(h/2)) / (C(h/2) - C(h)) = (C(h/8) - C(h/4)) / (C(h/4) - C(h/2)) > 1$$

$$C(h_0) = C(h) + (C(h/2) - C(h)) + (C(h/4) - C(h/2)) + \dots$$

[15] Thus all that is needed is an answer, and hence the method of doubling resolution simulation, particularly extensive on the value of k is a global geometrically converging related k 's.

$$k_{124} = (C(h/4) - C(h/2)) / (C(h/2) - C(h))$$

$$k_{248} = (C(h/8) - C(h/4)) / (C(h/4) - C(h/2))$$

[16] The CTM was recompiled of resolution. The original (G1), was doubled to 144 × 90 (G4), and 576 × 354 × 90 (G8).

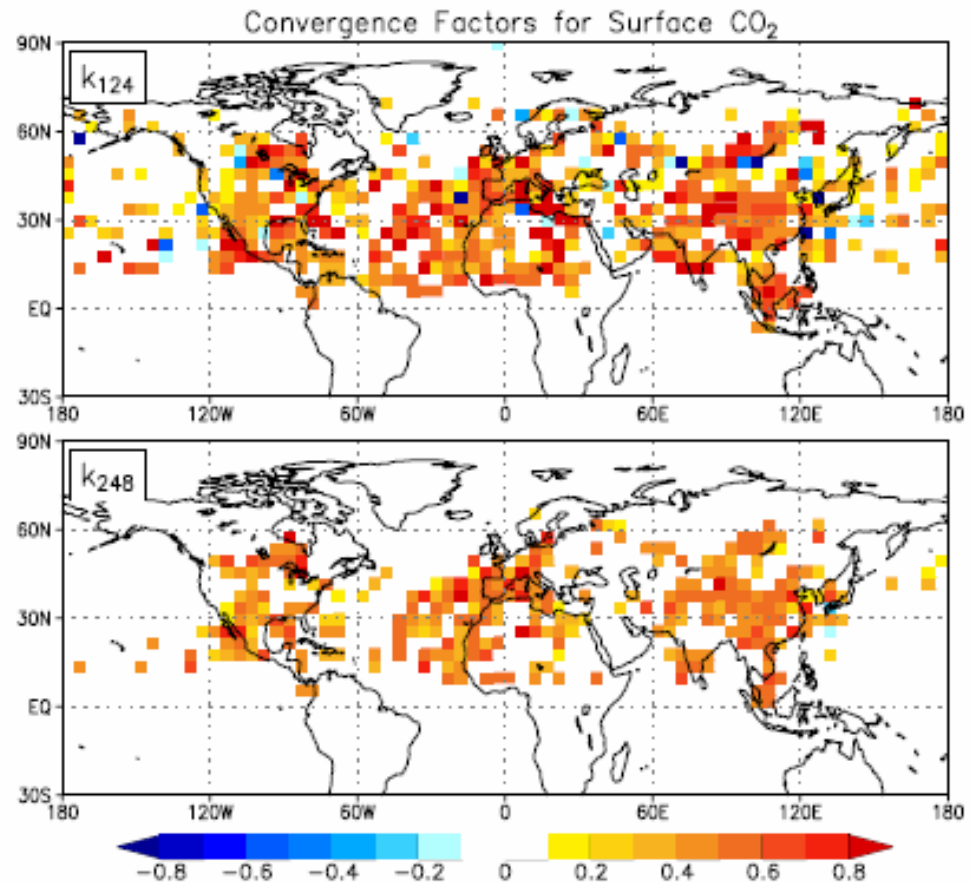


Figure 2. Convergence factors, k , for the 1–2–4 and 2–4–8 sequences of doubled resolution CTM

O₃ Production in PBL over East Asia in March/April 2001

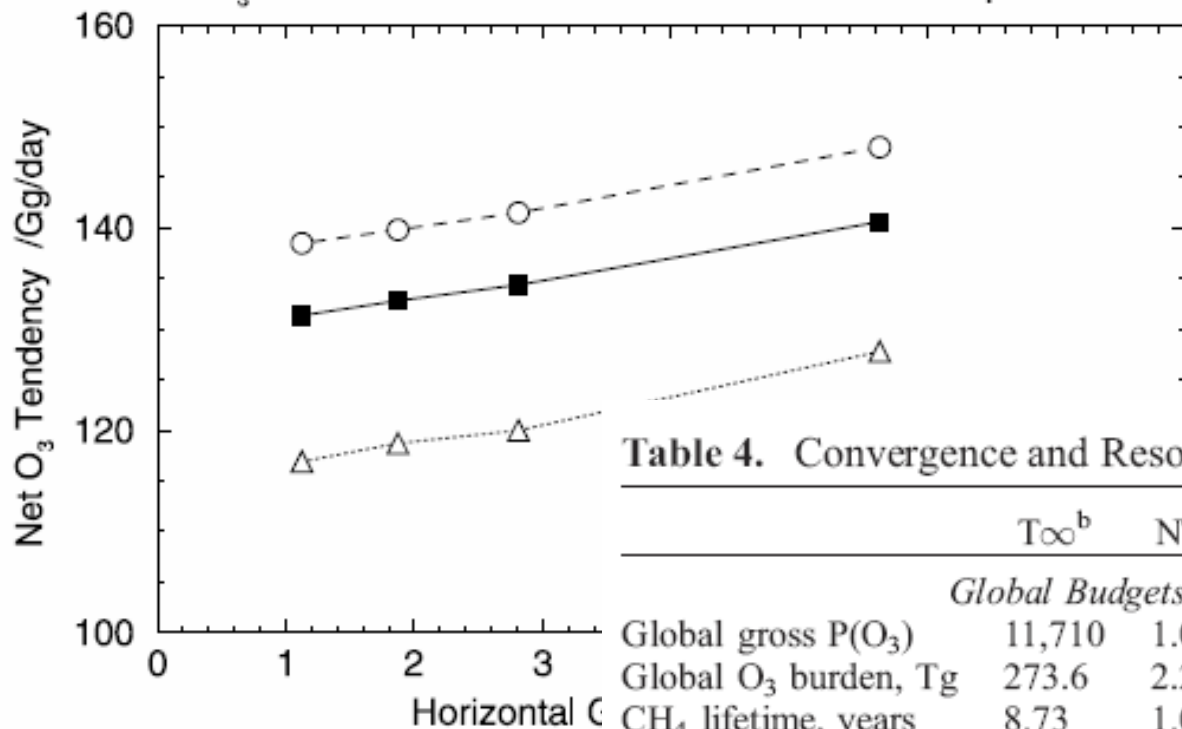


Table 4. Convergence and Resolution Errors in Oxidant Budgets^a

	T _∞ ^b	N ^c	ε ₂₁ ^d	ε ₄₂	ε ₆₃	ε ₁₀₆
<i>Global Budgets (From Table 2)</i>						
Global gross P(O ₃)	11,710	1.0	578	422	335	169
Global O ₃ burden, T _g	273.6	2.2	20.2	10.0	4.1	1.3
CH ₄ lifetime, years	8.73	1.0	-0.67	-0.41	-0.29	-0.16
<i>Impacts of East Asian Emissions (From Table 3)</i>						
Regional net P(O ₃)	43.0	1.0	15.8	6.2	4.3	2.5
Regional gross P(O ₃)	84.1	1.8	18.0	6.2	4.5	2.5
Regional O ₃ burden	20.8	2.9	6.6	1.4	0.3	0.1
Global gross P(O ₃)	210.4	2.3	20.6	12.8	5.0	1.5
Global O ₃ burden	49.6	2.1	6.6	3.4	1.4	0.5

^aBudgets in Gg/day and burdens in Gg unless stated.

^bCorrected budget based on T₄₂-T₆₃-T₁₀₆ convergence.

^cOrder N used in Richardson Extrapolation.

^dEstimated error at each resolution, ε₂₁ = T₂₁-T_∞.

10-yr runs for Trop. CO₂ (circa 2004)

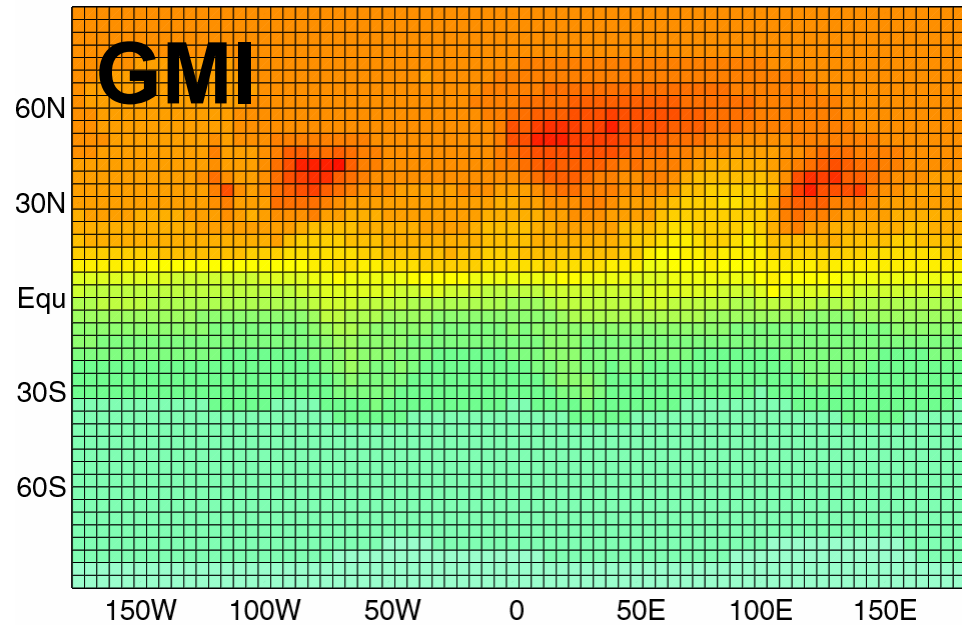
surface CO₂ mixing ratio

annual/monthly means from Yr 10

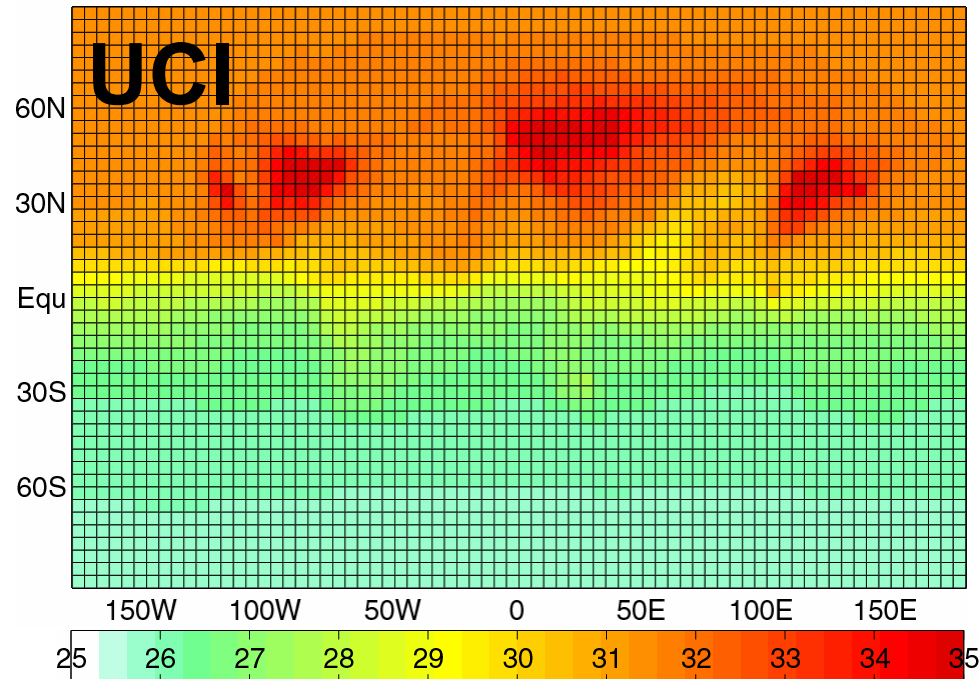
GMI appears more “diffusive”
UCI retains higher abundances
over source regions.

N.B. model 4x5 grid shown

surface CO₂ (ppm) - annual mean - GMI std

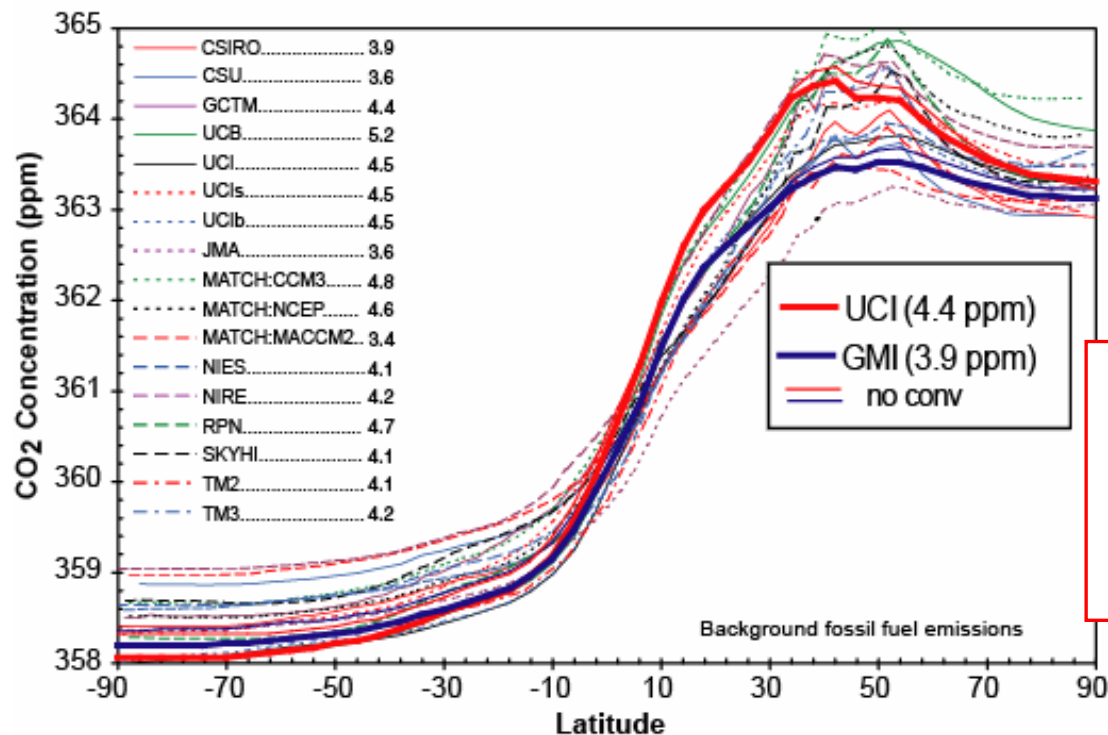


surface CO₂ (ppm) - annual mean - UCI std



TransCom 3 CO₂ inversion intercomparison: 1. Annual mean control results and sensitivity to transport and prior flux information

By KEVIN ROBERT GURNEY¹, RACHEL M. LAW², A. SCOTT DENNING¹, PETER J. RAYNER², DAVID BAKER³, PHILIPPE BOUSQUET⁴, LORI BRUHWILER⁵, YU-HAN CHEN⁶, PHILIPPE CIAIS⁴, SONGMIAO FAN⁷, INEZ Y. FUNG⁸, MANUEL GLOOR⁹, MARTIN HEIMANN⁹, KAZ HIGUCHI¹⁰, JASMIN JOHN⁸, EVA KOWALCZYK², TAKASHI MAKI¹¹, SHAMIL MAKSYUTOV¹², PHILIPPE PEYLIN⁴, MICHAEL PRATHER¹³, BERNARD C. PAK¹³, JORGE SARMIENTO⁷, SHOICHI TAGUCHI¹⁴, TARO TAKAHASHI¹⁵ and CHIU-WAI YUEN¹⁰



Numerical error in tracer transport is a large source of the uncertainty in inverse modeling.

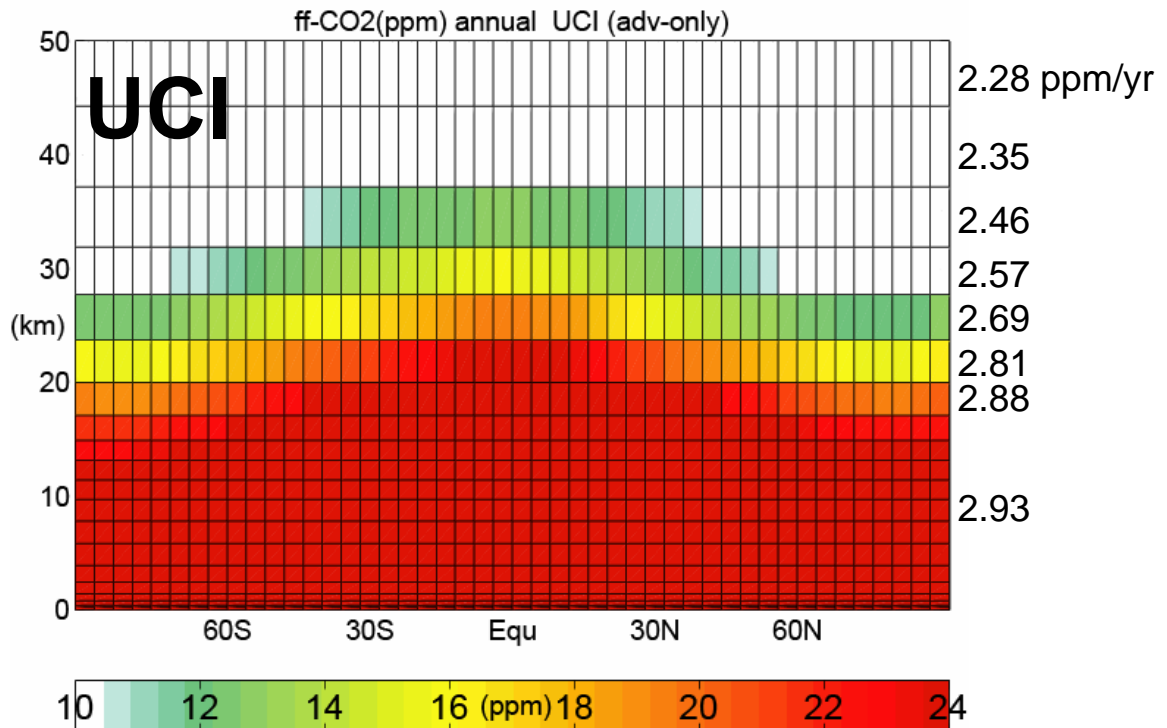
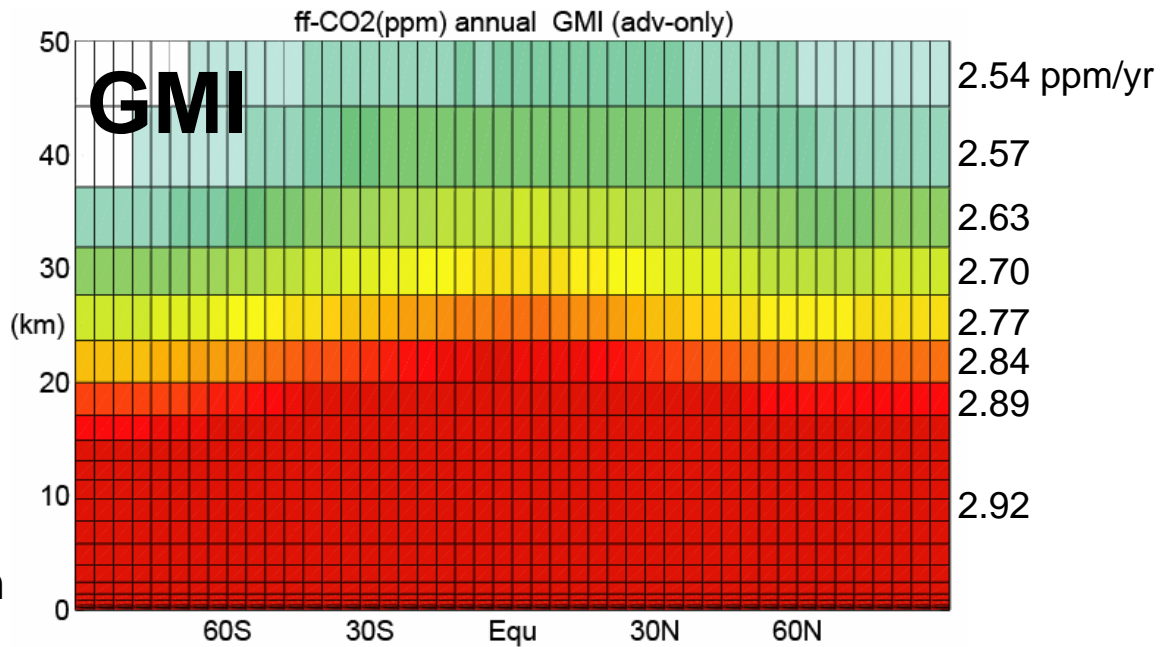
Fig. 3. Annual mean, zonal mean surface CO₂ concentration (ppm) resulting from the individual and combined (relative to a background concentration of 350 ppm) background fluxes for each of the models. The interhemispheric difference (ppm) for the background fossil and combined background CO₂ is listed in the key for each model. Note that the scale is different for each of the plots.

10-yr runs for Stratospheric CO₂ (circa 2004)

zonal mean CO₂ mixing ratio annual average of Year 10 actual model grid shown.

approach to steady-state shown as growth in Yr10

GMI vs. UCI (adv-only)
 GMI is more "diffusive"
 UCI is still 24% away from s-s



What is the Problem?

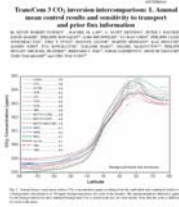
One expectation we have, when solving the chemistry-transport equations for atmospheric trace species, is that there should be a single, correct answer.

If we include the correct physics, refine our numerical methods, and increase the resolution, then the model will converge to this correct answer. With this optimism, the authors began a series of numerical experiments under the auspices of the NASA Global Modeling Initiative (GMI) to demonstrate that independent chemistry-transport models (CTMs) developed at UC Irvine and NASA Goddard could achieve this

After considerable effort to ensure that both CTMs simulated the same physical processes, we failed to produce two similar answers. We conclude that considerable uncertainty in the CTM simulation of trace species remains due to the choice of numerical methods, and we have not yet ruled out structural differences as the source of this error.

Why do we care?

Consider the TransCom3 (T3) effort to use the atmospheric variations in CO₂ abundance to deduce the pattern of fossil-fuel emissions. The differences among the dozen or so CTMs in simulating atmospheric CO₂ gradients from a prescribed fossil-fuel pattern became a major source of uncertainty in the inverse calculation.

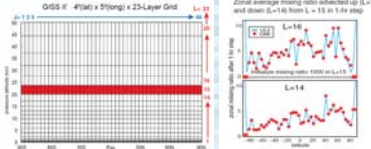


(K.R. Gurney et al., 2002, Nature 415, 626-630; 2003, Tellus 55B, 555-579).

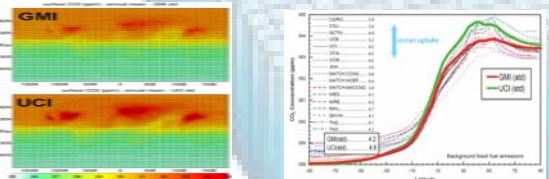
These T3 results were due to a wide range in meteorological fields and numerical methods. Using modern tracer-transport algorithms and the same met fields, we assumed that we could eliminate at least the errors in tracer transport. This optimism was short-lived. After correcting several minor "bugs" in both CTMs, ensuring that we interpreted the emissions and meteorological fields in the same manner, the differences were uncomfortably large: not significantly less than the original T3 model spread; comparable to the amplitude of the oceanic CO₂ sink

TEST:

GISS GCM II⁺ met fields, 4°lat x 5°long x 23 layers, 1-hr advection step with tracer only in L=15, excellent, except at poles.



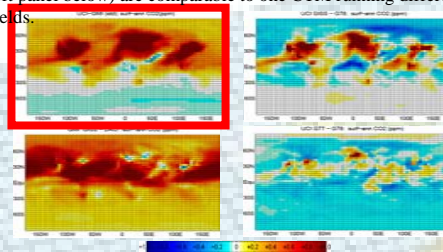
Surface CO₂, how bad can it be?



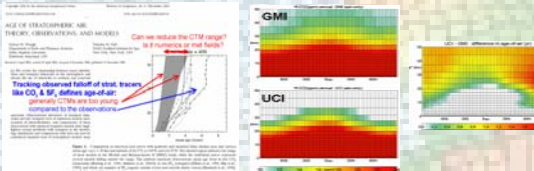
UCI-GMI Grades for Surface CO₂ mixed:

Tropics & S. Hemisphere: A
N. Hem. & Source Regions: C-

UCI - GMI differences in N. Hem, using the same met fields, (upper left panel below) are comparable to one CTM running different met fields.



Stratospheric age-of-air, how bad can it be?

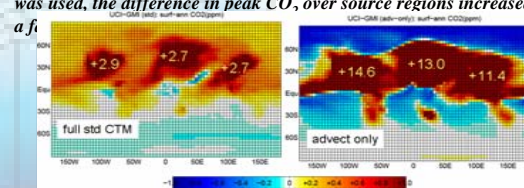


The age-of-air in the stratosphere (defined as the time since it was last in the troposphere) describes the stratospheric circulation and time-scales of chemical perturbations. It is measured directly from nearly inert trace gases that are increasing linearly in the troposphere (CO₂, SF₆). CTMs show a very wide range, as does the UCI - GMI comparison (figures above).

The year-10 zonal average of stratospheric CO₂ shows surprisingly large differences, corresponding to more than 1 yr in age. GMI is more diffusive,

the Advection Algorithm

and the problem is not obviously related to differing polar treatment. Multiple tests with both UCI and GMI CTMs showed that the age-of-air was barely influenced by wet convection or boundary layer mixing. Thus we continued with only advection of tracer by the winds resolved on the original 4x5 grid. The surface CO₂ differences were greatly reduced by convection/boundary-layer mixing: when only advection was used, the difference in peak CO₂ over source regions increased by a factor of 10.



Global rms differences (ppm): 1.6 (original CTMs), 0.6 (corrected CTMs), 2.8 (advection only CTMs)

Is there a Correct Answer?

Since tracer advection is represented physically (rather than parametrically), errors due to calculation on a finite grid and should disappear as the resolution increases.

With the UCI CTM we pursued a "doubling to convergence" approach: from F1 (original resolution Δ: 72x46x23) to F8 (Δ/8: 576x354x184). The series of calculated abundances (A) at any location and time obtained through Δ-halving converges (i.e. Aitken's):

$$A^{true} = A(\Delta) + A(\Delta/2) - A(\Delta) + A(\Delta/4) - A(\Delta/2) + A(\Delta/8) - A(\Delta/4) + \dots$$

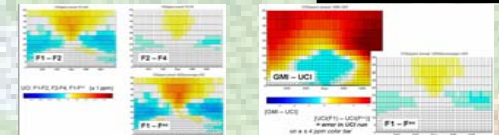
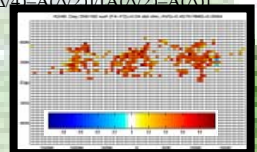
$$= A(\Delta) + A(\Delta/2) - A(\Delta) / (1 - k_{\Delta/2}),$$

where the convergence factor $k_{\Delta/2} = [A(\Delta/4) - A(\Delta/2)] / [A(\Delta/2) - A(\Delta)]$

Using a 2-month simulations with Δ/2, Δ/4, and Δ/8 and surface CO₂ abundances on day 62 (Sep 1), we calculate

$$k_{\Delta/2} = 0.46 \pm 0.17 \text{ and } k_{\Delta/4} = 0.46 \pm 0.06$$

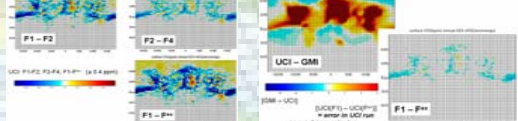
(see Wild & Prather, 2006, JGR D111, D11305).



Convergence of UCI CTM (F1=>F2=>F4) for stratospheric CO₂ in year 10 is shown as differences F1 - F2, F2 - F4, and extrapolated error F1 - F4.

The error is at most ±0.4 ppm, much less than UCI-GMI differences.

(Note the change in color scale from ±1 ppm to ±4 ppm.)

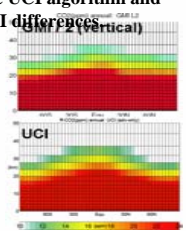


Surface CO₂ in year 10 shows similar convergence, and once again, the UCI errors are much smaller than the UCI-GMI differences.

Do both Numerical Methods get the same Answer?

Correcting the UCI CTM reduces the UCI-GMI differences, by at most 10%, suggesting that tracer advection errors using the GMI algorithm are much larger than those with the UCI algorithm and to first-order are represented by the GMI-UCI differences.

As of now, the GMI CTM has been able to complete only a single case with doubled vertical layers (23 to 46) and no change in horizontal resolution. The UCI-GMI differences are somewhat reduced (see figure), but closure on this topic awaits the full F2 and F4 GMI simulations.

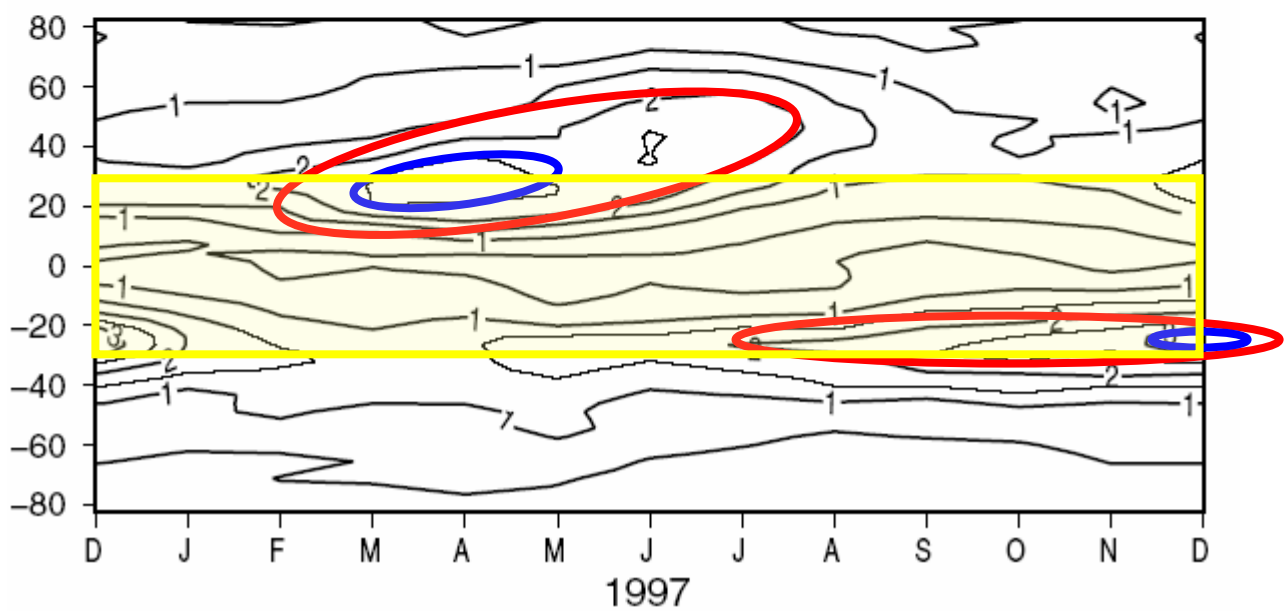
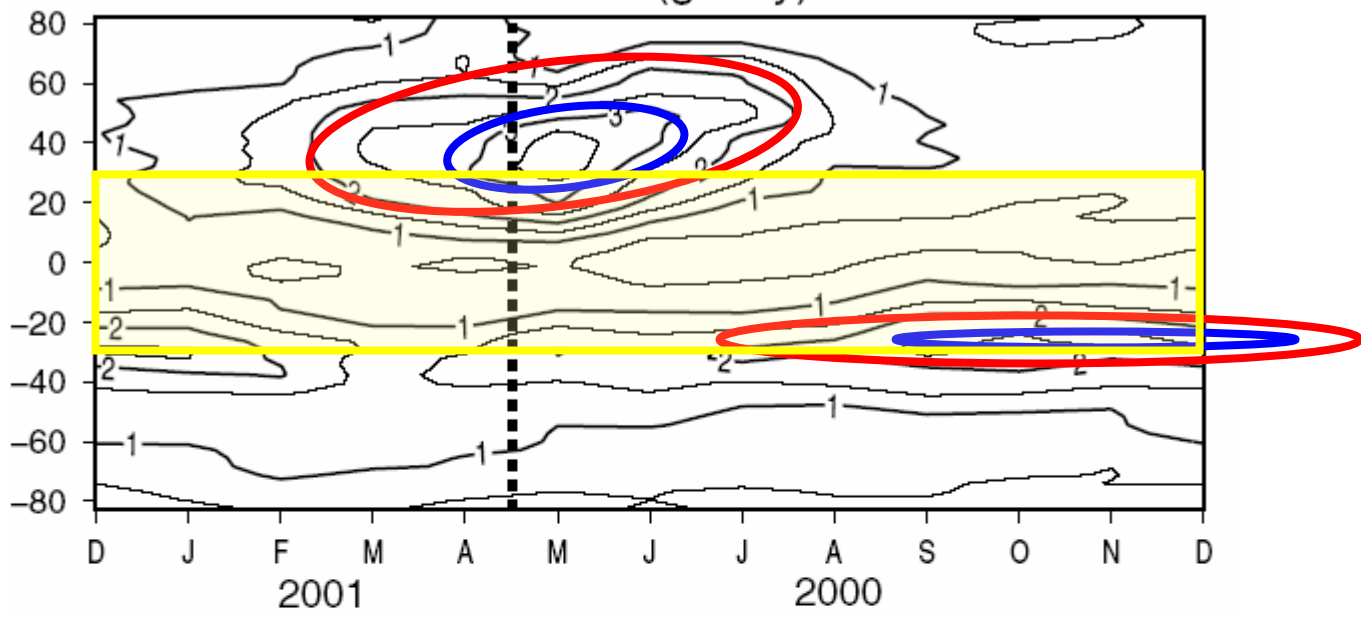


► Strat-trop interactions – what are we missing ?

Strat-Trop Exchange of Ozone is important for UT ozone, and is likely to shift with climate change.

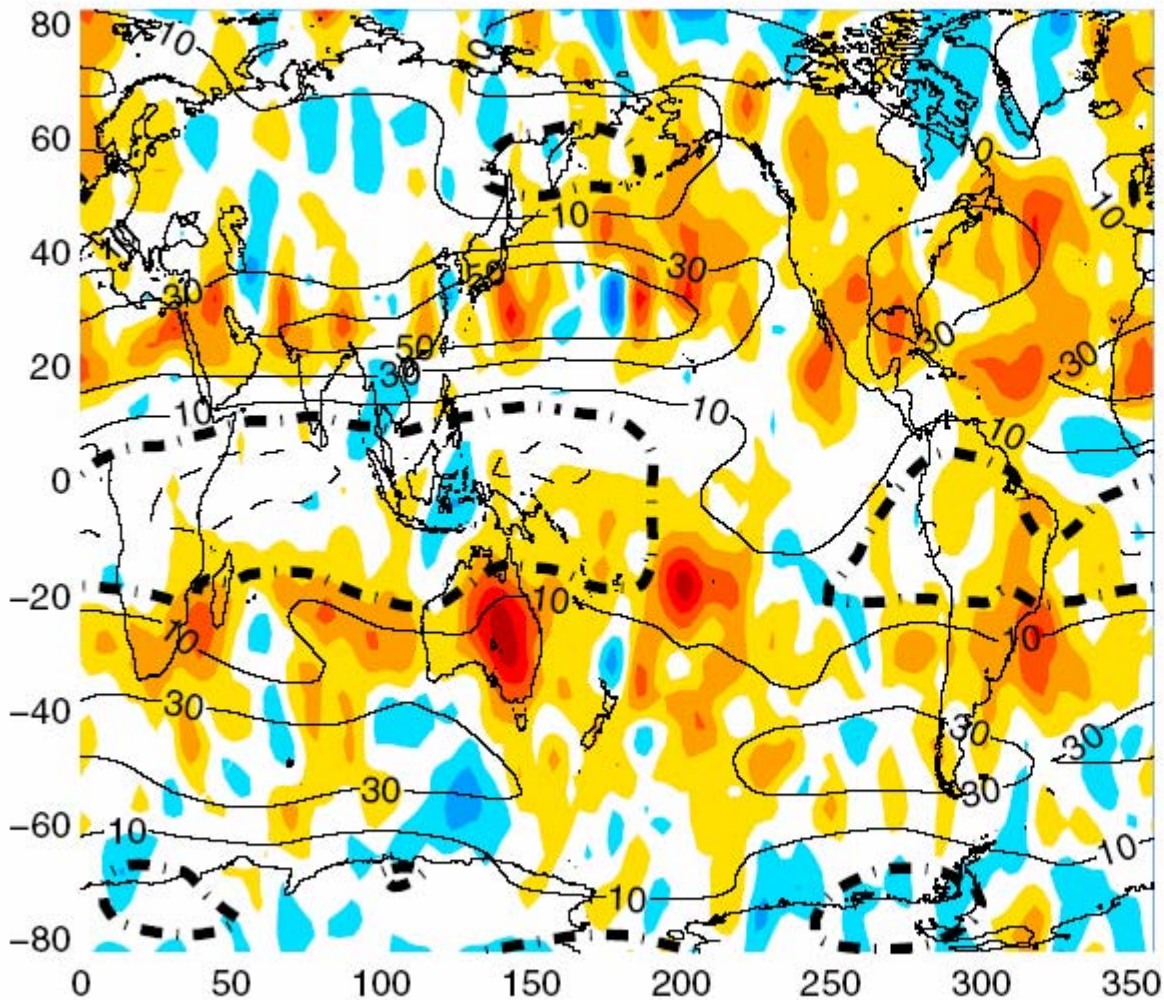
There is some interesting chemistry in the Upper Trop / Lower Stratosphere.

STE O₃ flux (g m⁻² y⁻¹): avg = 1.25; contours at 2 and 3

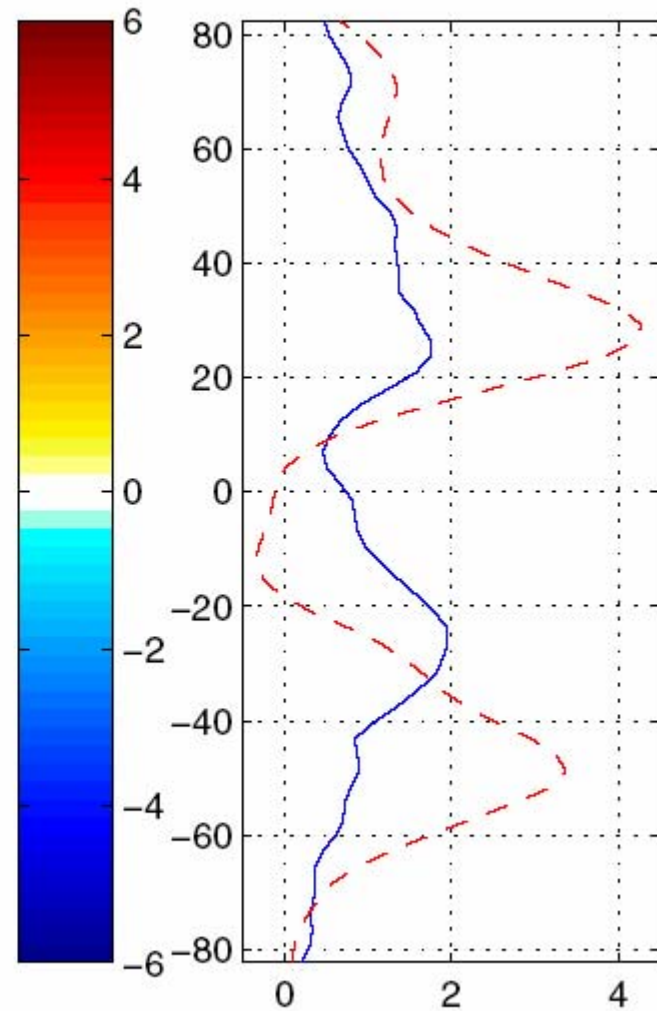


Monthly STE O_3 flux – N.B. blue regions are 'noise'

JAN 1997 ($g/m^2/year$)



STE O_3
zonal wind



Diagnosing the Stratosphere-to-troposphere Flux of Ozone in a Chemistry Transport Model

Juno Hsu,¹ Michael J. Prather,¹ Oliver Wild,²

Fast-JX application

CTM modeled STE O₃ flux (Tg)

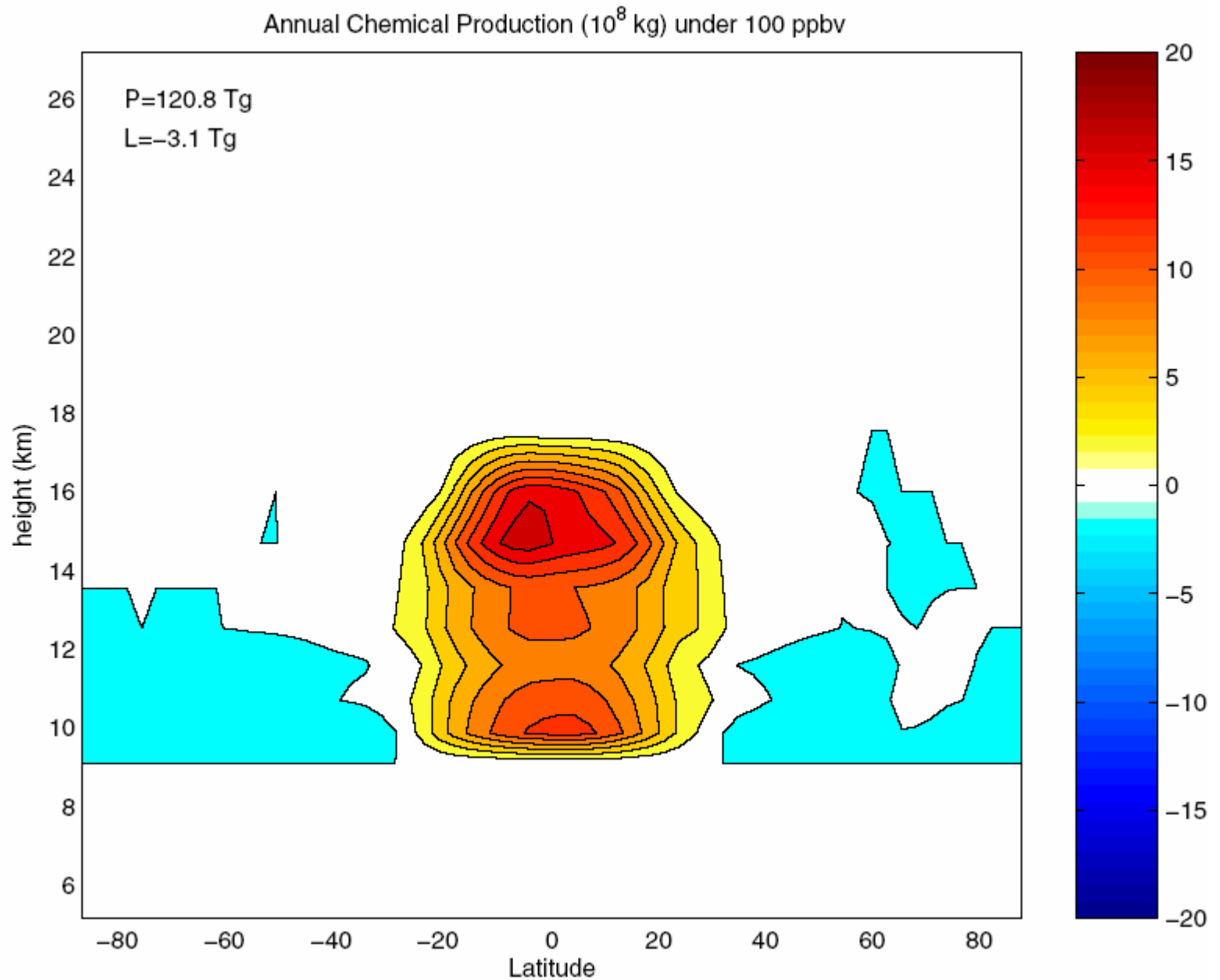
NH ~ SH 2000/2001

NH > SH 1997

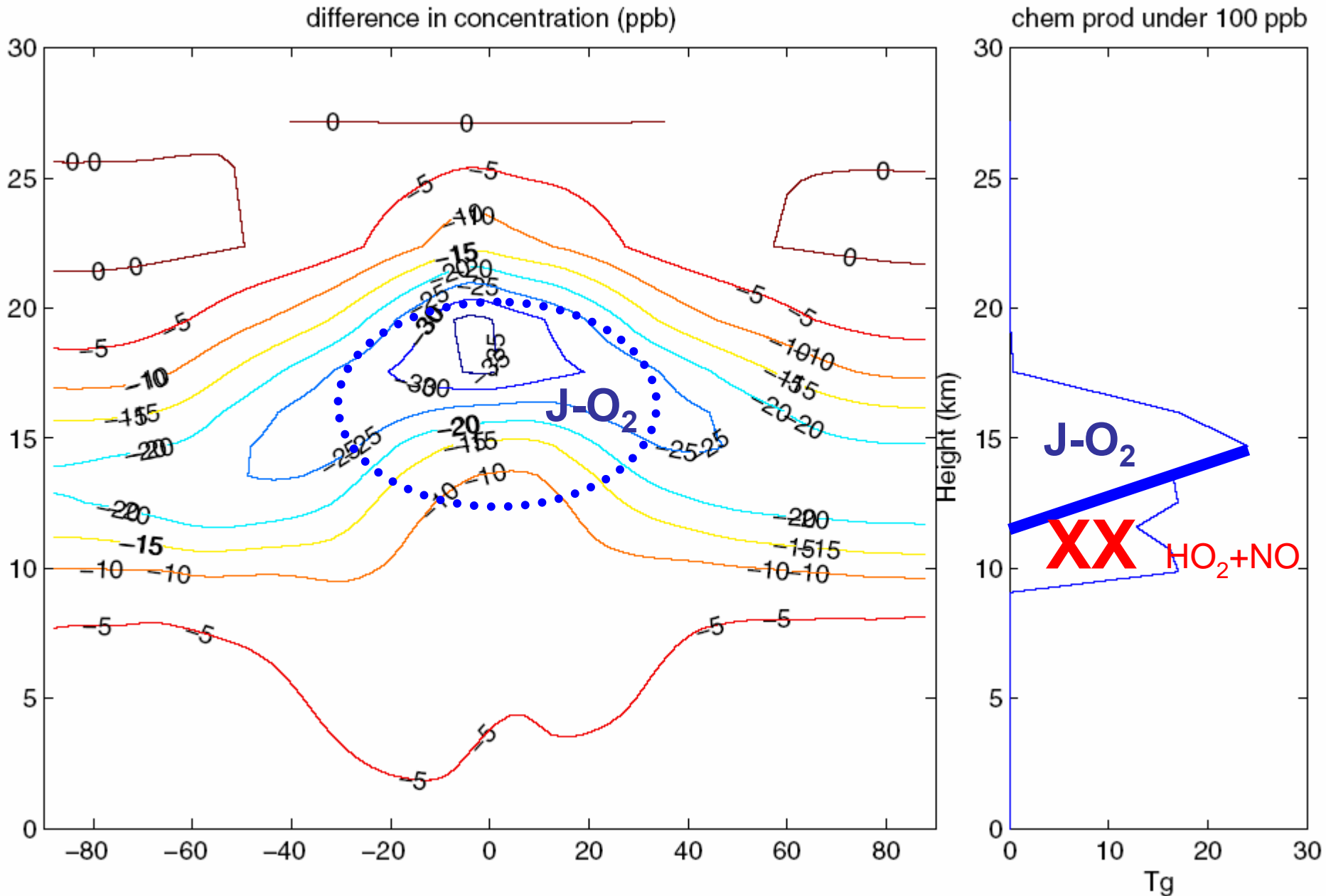
Total 530 ± 50? (due to lower/upper b.c.)

N.B. ~80 Tg from J-O₂ in tropical trop

There is considerable O₃ Production near and below the 100 ppb surface



N.B. Photolysis of O_2 is a significant source (>20 ppb) of trop O_3 in upper tropics



Diagnose O₃ flux into lower stratosphere:

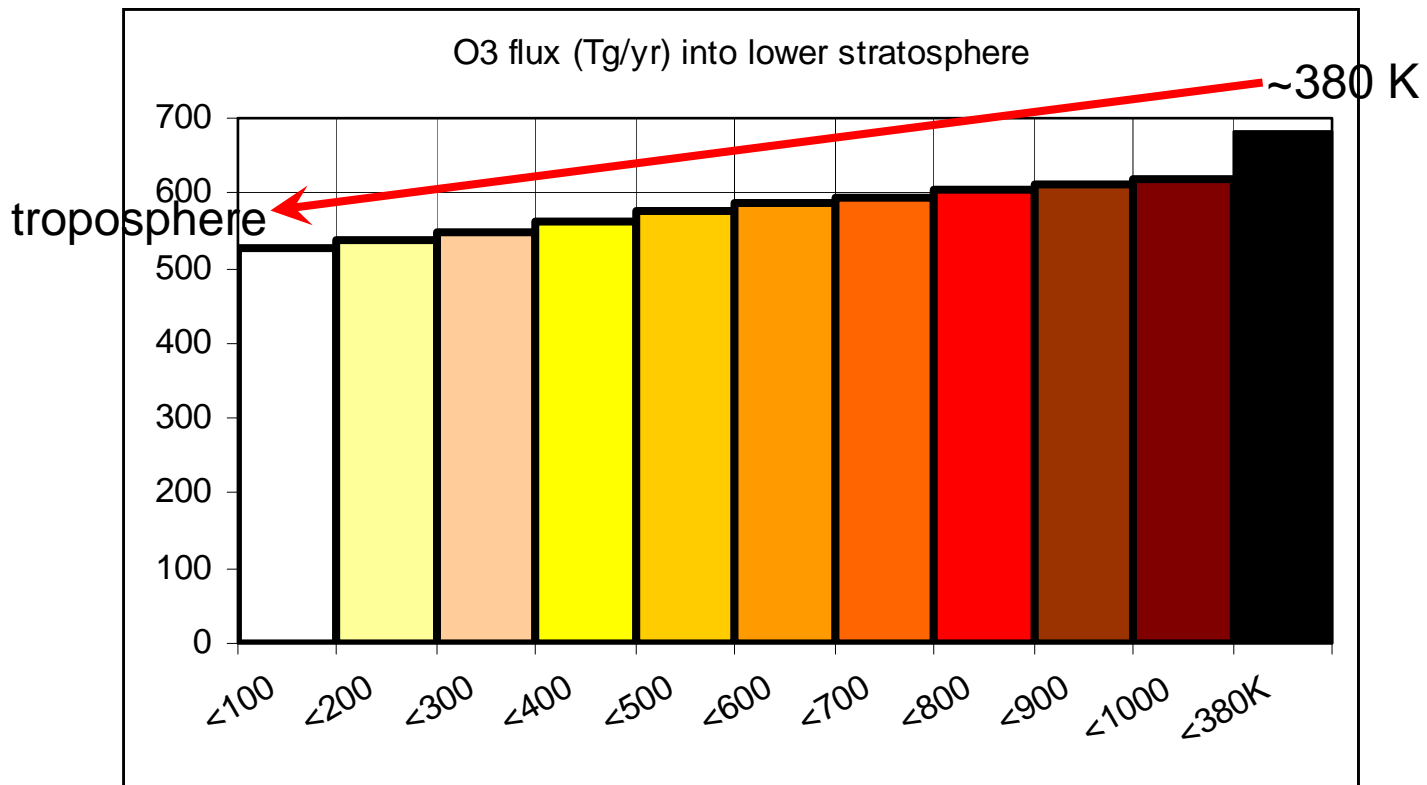
~675 Tg crosses L=33 (~?380K)

600 Tg crosses 1000 ppb surface

...

527 Tg crosses 100 ppb surface into troposphere!

? what observations can test this modeled 25% loss of the O₃ flux



Given Climate Change, what can we expect for changes in O₃ . . .

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 108, NO. D12, 8528, doi:10.1029/2002JD002617, 2003

Effect of stratosphere-troposphere exchange on the future tropospheric ozone trend

W. J. Collins, R. G. Derwent, B. Garnier, C. E. Johnson, and M. G. Sanderson
Met Office, Bracknell, UK

D. S. Stevenson

Department of Meteorology, University of Edinburgh, Edinburgh, UK

Received 25 June 2002; revised 22 October 2002; accepted 27 February 2003; published 15 May 2003.

[1] This paper investigates the impact of circulation changes in a changed climate on the exchange of ozone between the stratosphere and the troposphere. We have identified an increase in the net transport although a decreased ozone |

Table 1. Ozone Transport and Loss Fluxes Averaged Over 4 Years^a

	1991–1994	2091–2094	Difference
Flux across 200 hPa			
Downward	8057±48	9899±90	1842±102
Upward	6989±45	8433±82	1444±94
Net downward	1067±8	1465±13	398±15
Loss rate	4914±16	5304±14	390±21
Burden (below 200 hPa)	284±2	271±1	–14±2

^aFluxes are given in Tg yr⁻¹. Average ozone burden below 200 hPa (Tg). Quoted errors are due to interannual variability only.

Given Climate Change, what can we expect for changes in O₃ . . .

JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 109, D21104, doi:10.1029/2004JD005075, 2004

Identification of anthropogenic climate change using a second-generation reanalysis

Benjamin D. Santer,¹ Tom M. L. Wigley,² Adrian J. Simmons,³ Per W. Kållberg,³ Graeme A. Kelly,³ Sakari M. Uppala,³ Caspar Ammann,² James S. Boyle,¹ Wolfgang Brüggemann,⁴ Charles Doutriaux,¹ Mike Fiorino,¹ Carl Mears,⁵ Gerald A. Meehl,² Robert Sausen,⁶ Karl E. Taylor,¹ Warren M. Washington,² Michael F. Wehner,⁷ and Frank J. Wentz⁵

Received 31 May 2004; revised 2 August 2004; accepted 19 August 2004; published 4 November 2004.

[1] Changes in the height of the tropopause provide a sensitive indicator of human effects on climate. A previous attempt to identify human effects on tropopause height relied on information from 'first-generation' reanalyses of past weather observations. Climate data from these initial model-based reanalyses have well-documented deficiencies, raising concerns regarding the robustness of earlier detection work that employed these data. Here we address these concerns using information from the new second-generation ERA-40 reanalysis. Over 1979 to 2001, tropopause height increases by nearly 200 m in ERA-40, partly due to tropospheric warming. The spatial pattern of height increase is consistent with climate model predictions of the expected response to anthropogenic influences alone, significantly strengthening earlier detection results. Atmospheric temperature

► Why interactive chemistry-climate coupling ?

I would like to think that much of the chemistry → climate driving can be averaged except for

aerosol-cloud interactions

clearly this interaction changes the local dynamics

ozone in the upper trop / upper stratosphere

do the sharp gradients in O_3 heating affect the LS and STE dynamics?
how can we do this without some interactive chemistry in LS?

absorbing aerosols (and O_3 ?) in pollution plumes

ABC issues and the Monsoons, but also perhaps important in inter-hemispheric transport: differential heating in plumes is critical in maintaining them?