Modeling Studies in Tropospheric Ozone Changes:

The Effect of Natural and Anthropogenic Sources and Climate Change

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Our work

thus far

Previously LLNL studied 3D global O_3 with a tropo-spheric model:

- GRANTOUR
- Fully predictive troposphere; specified stratosphere
- Meteorology from GCM
- Chemistry included oxidation of:

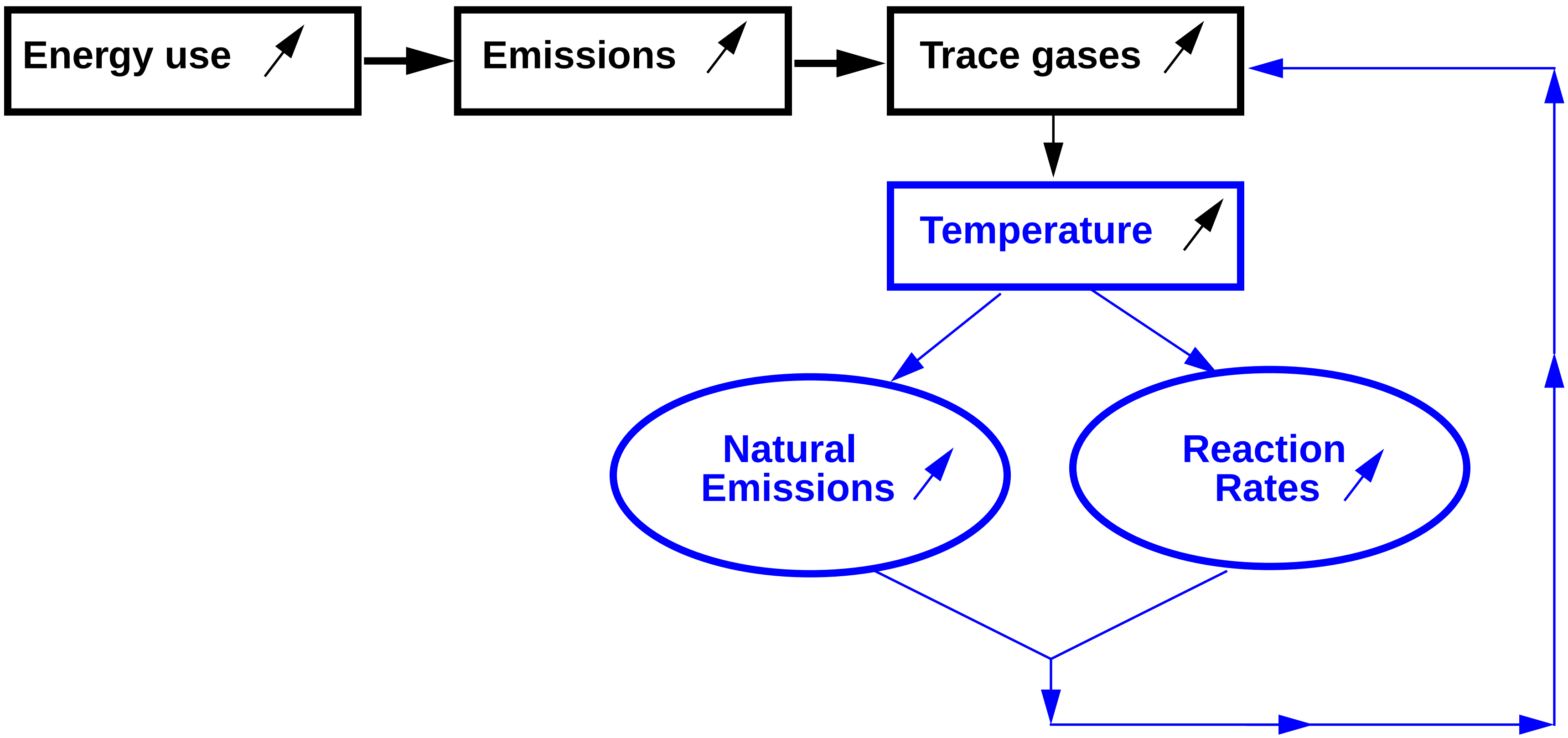
  CO, CH_4, isoprene, ethane, propane, butane, ethene, propene, butene, NO, NO_2, PAN, HNO_3,

  O_3, OH, H_2O_2, HCHO, etc.

- Regional to global issues

Recent results

Use Global and Box Models to understand:



IPCC 1995: Temp increases 1 - 4.5K by 2100:

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(Insert figure from IPCC here)

Annual mean temp. change (K) under Scenario IS92a, from 1795 to 2030, from CO_2 and aerosols (Mitchell et al., J. Climate, 10, 2365-2386, 1995)

Temperature increases will affect:

Reaction rates: $k = A \times \exp(-E_a/RT)$

Temperature dependent source emission rates:

Soil NO_x

Isoprene

Lightning NO_x

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Soil NO_x emissions depend strongly on temperature

For example, for grasslands:

$$\log_{10}(NO\ flux) = (0.0049 \pm 0.008)(^{\circ}C^{-1})T_A - (0.83 \pm 0.13)$$

[ ] = ng N/m^2-s

T_A = Air temperature

(Williams et al., **JGR 92**, 2,173 - 2,179, 1987)

ΔT = 2K $\Rightarrow$ NO flux increases 20%

ΔT = 4K $\Rightarrow$ NO flux increases 35 - 40%

Isoprene emissions increase exponentially with temperature

$$I(T,L) = I_S \cdot C_L \cdot C_T$$

$$C_L = \frac{\alpha c_{L1} L}{\sqrt{1+\alpha^2 L^2}} \qquad C_T = \frac{\exp[\frac{C_{T1}(T-T_S)}{RT_S T}]}{1+\exp[\frac{C_{T2}(T-T_M)}{RT_S T}]}$$

$\mathbf{I_S}$ = Emission rate at T_S and PAR = 1000 μmol/m2-s

L = Photosynthetically Active Radiation (PAR)

$\mathbf{T_{M,}}$, α, C_{L1}, C_{T1}, C_{T2} are specified from measurements

(Guenther et al., **JGR 98**, 12,609 -12,617, 1993)

ΔT = 2K $\Rightarrow$ I(T,L) increases 20%

ΔT = 4K $\Rightarrow$ I(T,L) increases ~40%

As the climate warms, lightning frequency increases:

$2 \times CO_2$ (~ 4.2°C)=> NO_x from lightning increases:

13% over oceans

70% over land

(Price and Rind, *JGR* **99**, 10,823-10,831, 1994)

Result - chemical implications of climate change:

Some natural emissions increase as temperature increases

Temperature change	Soil NO_x	Isoprene	Lightning
Δ T = 2K	+ 20%	+ 20%	-----
Δ T = 4K	+ 40%	+ 40%	+70% (land) +13% (ocean)

Method:

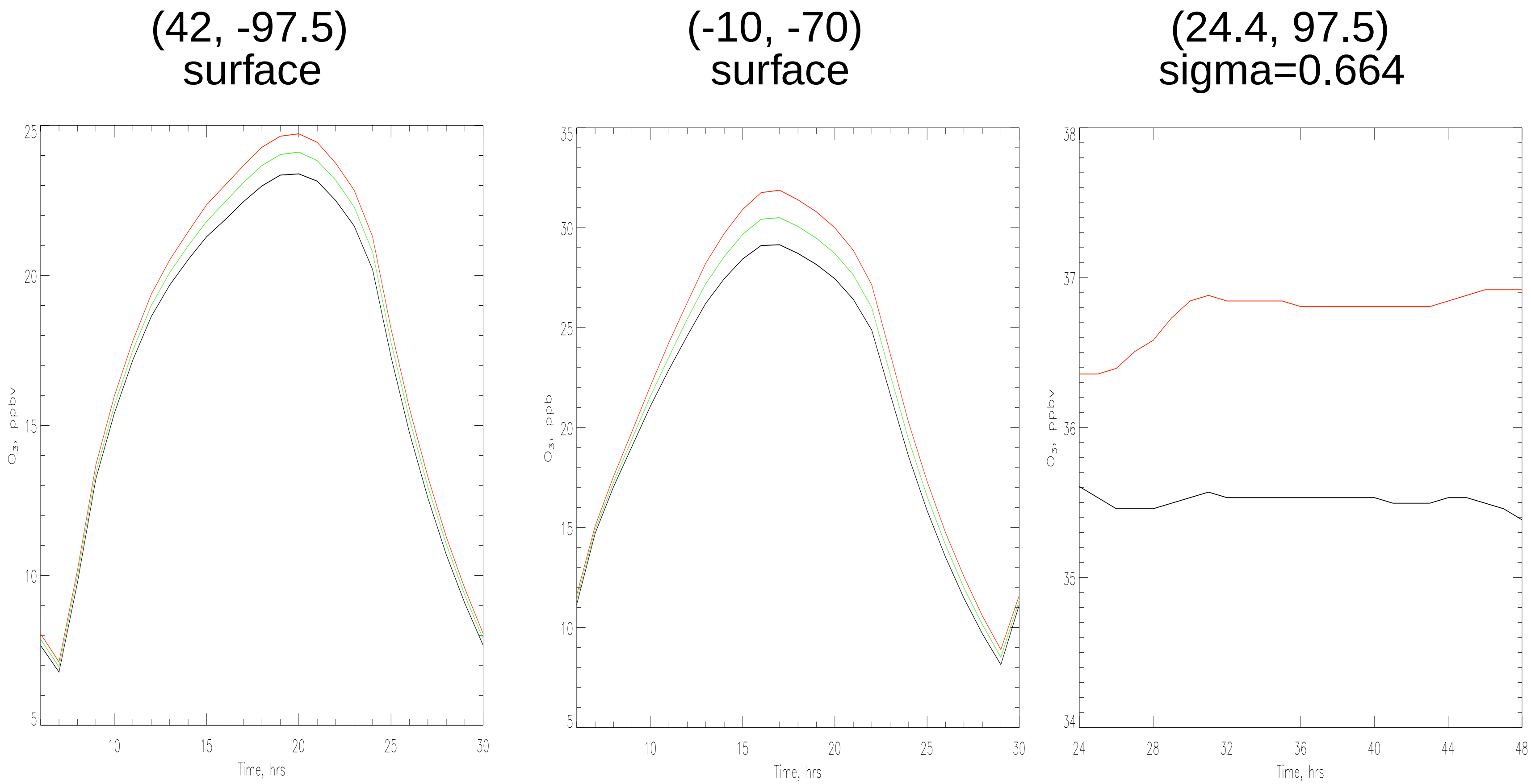
Conduct baseline present-day simulation with global model.

Extract concentrations and emissions for different regions.

Use box model to simulate regions for 3 scenarios.

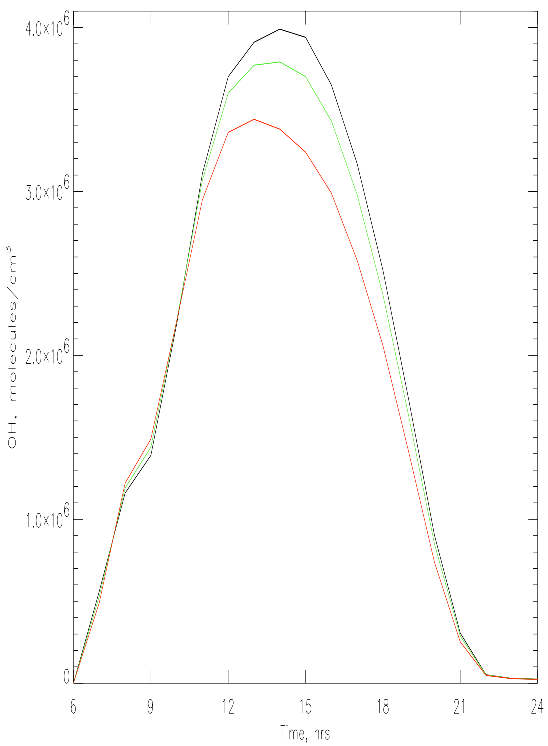
Scenario	Temperature	Soil NO_x	Isoprene	Lightning NO_x
Base	Base	Base	Base	Base
T+2K	$\Delta T = 2K$	+ 20%	+ 20%	-----
T+4K	$\Delta T = 4K$	+ 40%	+ 40%	+70% (land) +13% (ocean)

Changes in O_3 vary with location for Base, +2K, and +4K

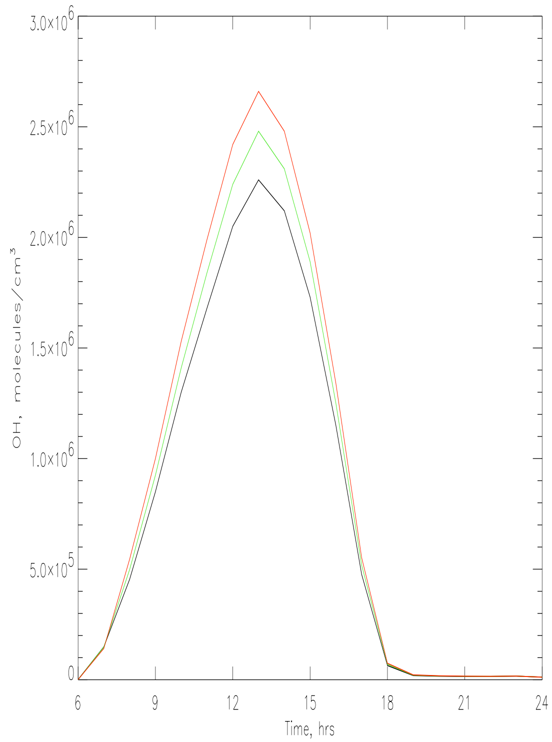


The OH may *increase* or *decrease* for Base, +2K, and +4K

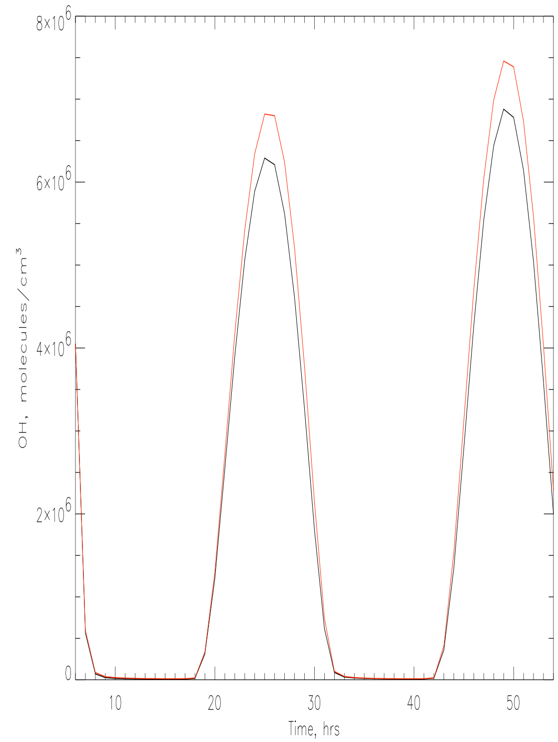
(42, -97.5)
surface



(-10, -70)
surface

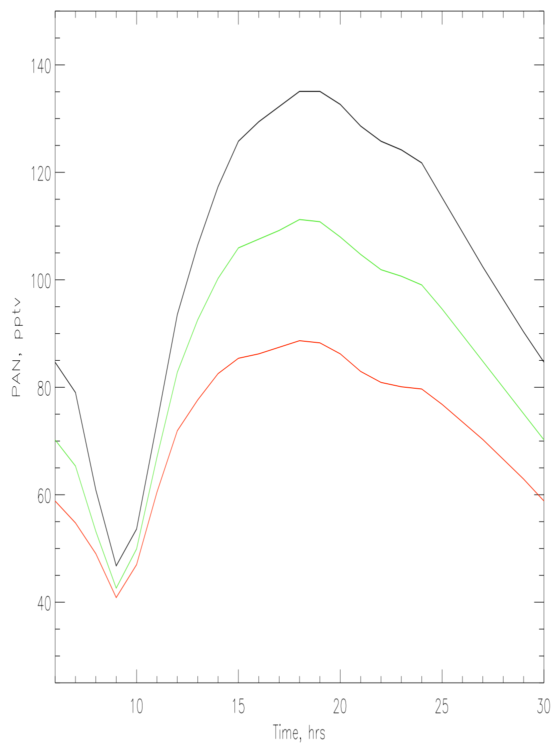


(24.4, 97.5)
sigma = 0.664

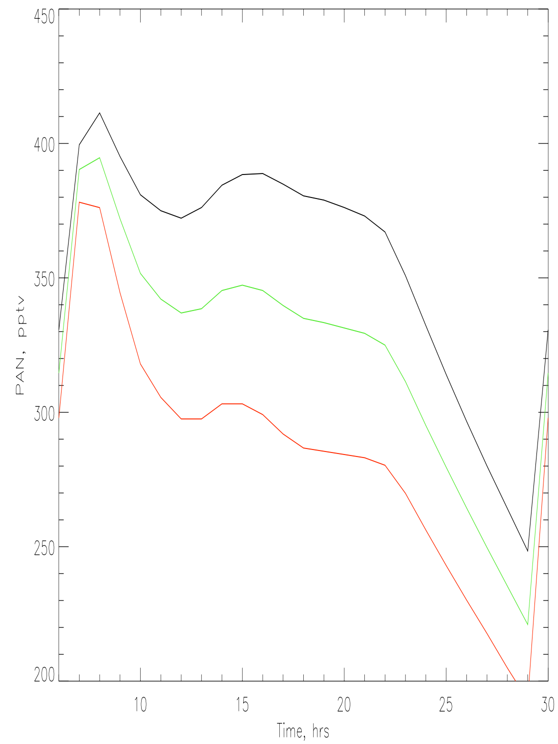


PAN concentrations change for Base, +2K, and +4K

(42, -97.5)
surface



(-10, -70)
surface



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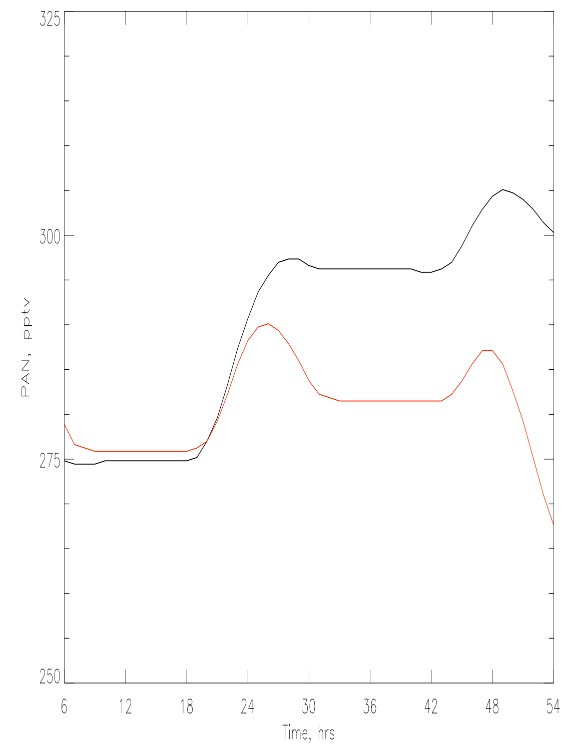

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Can we isolate the effects?

So far, have combined

Temp increase + isoprene + lightning NO_x + soil NO_x increases

For surface locations examine four separate scenarios:

(1) Base

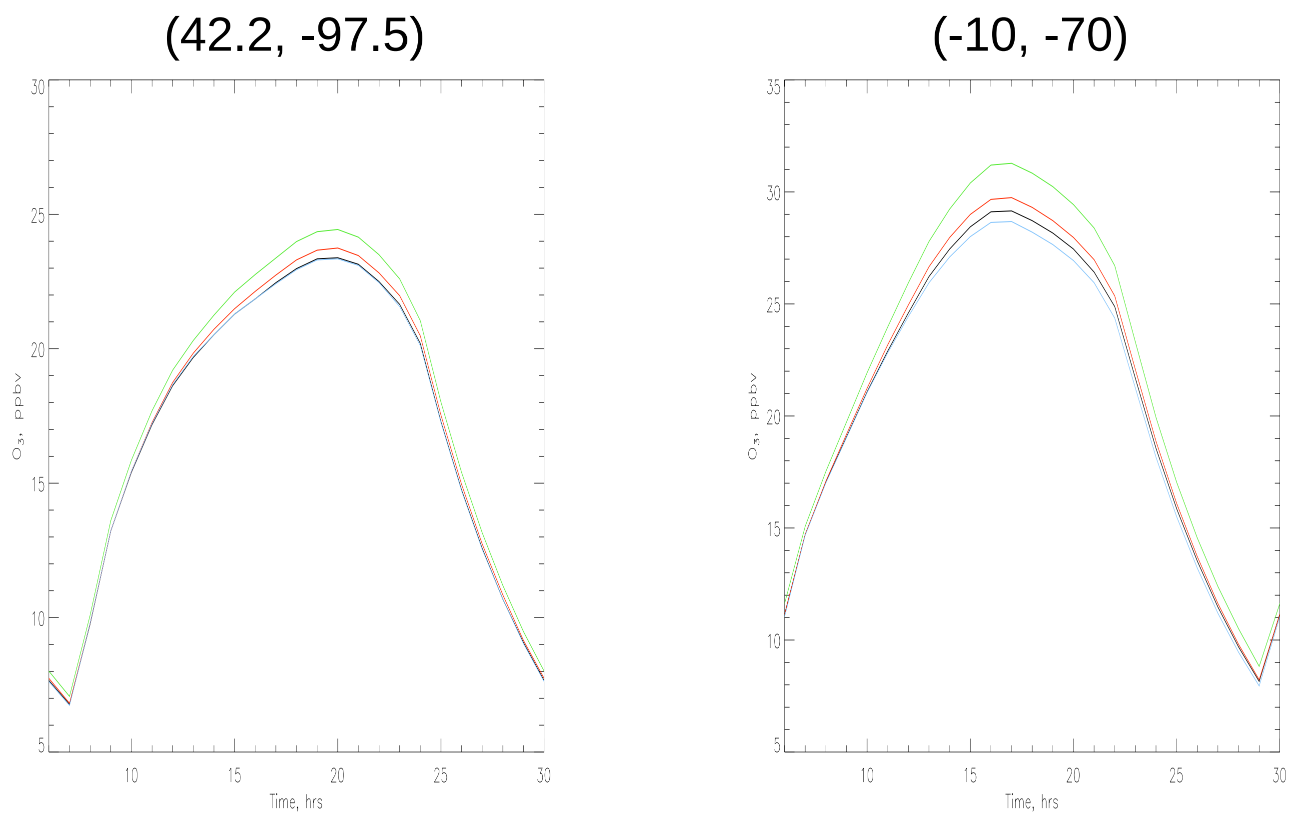
(2) Temperature + 4K only

(3) Soil NO_x + 40% only

(4) Isoprene + 40% only

What increase causes the greatest change in surface O_3?

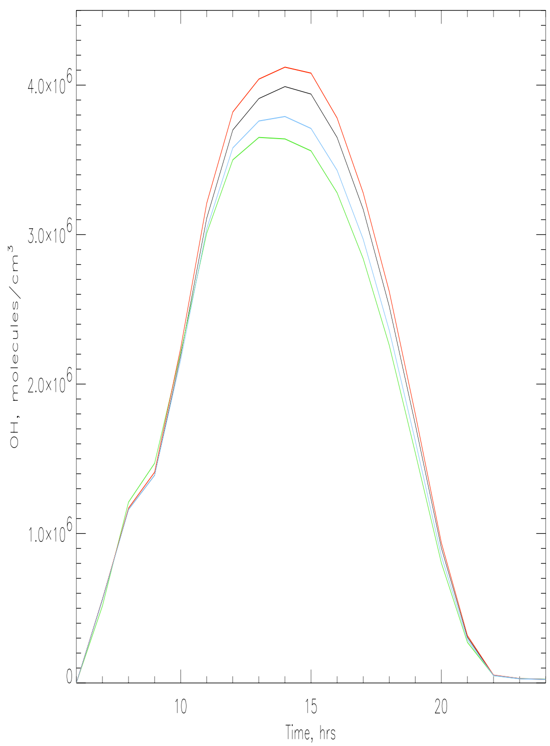
Base Temp+4K Soil NO_x only Isoprene only



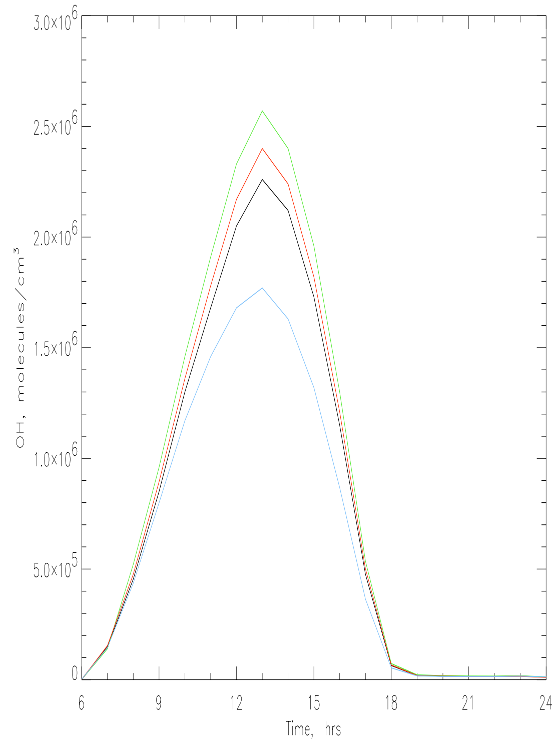
What increase causes the greatest change in surface OH?

Base   Temp+4K   Soil NO_x only   Isoprene only

(42,2, -97.5)



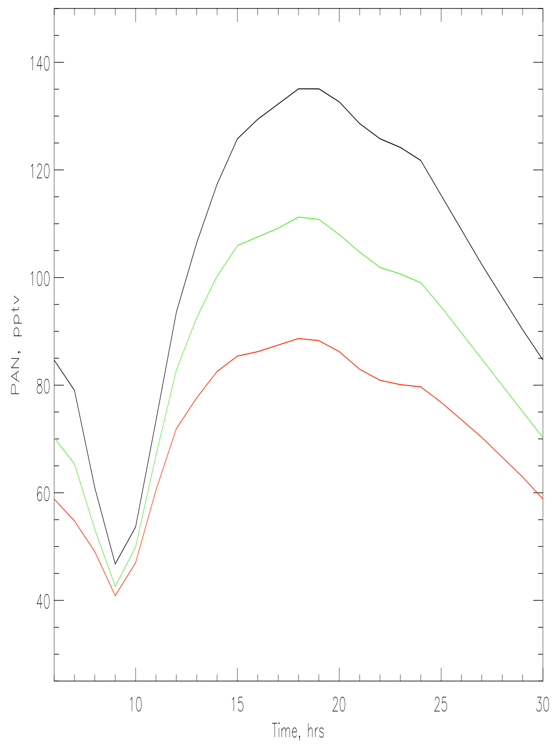
(-10, -70)



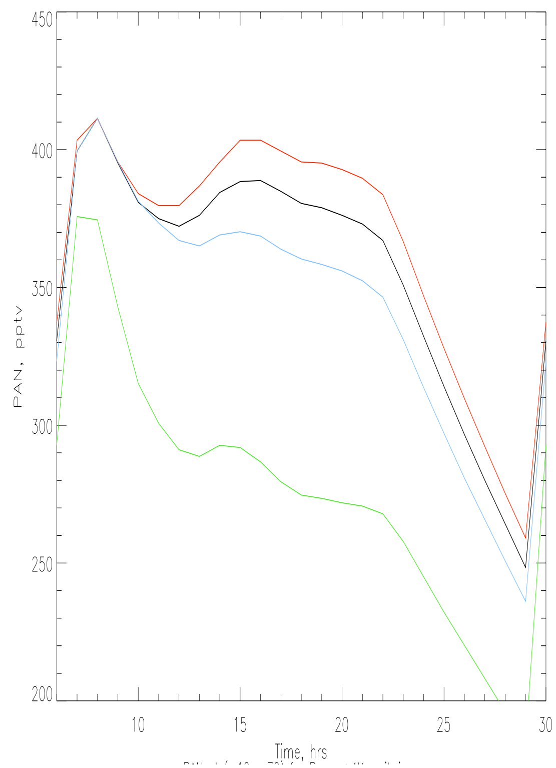
What increase causes the greatest change in surface PAN?

Base | Temp+4K | Soil NO_x only | Isoprene only

(42.2, -97.5)



(-10, -70)



Result - chemical implications of climate change:

Some natural emissions increase as temperature increases

Temperature change	Soil NO_x	Isoprene	Lightning
Δ T = 2K	+ 20%	+ 20%	-----
Δ T = 4K	+ 40%	+ 40%	+70% (land) +13% (ocean)

Climate/chemistry effects are regional:

(42, -98) surface

O_3	↑ 2 - 3 ppbv	
OH	↓ ~ 20%	
PAN	↓ 45 pptv	

(-10, -70) surface

O_3	↑ 3 - 4 ppbv	
OH	↑ ~ 20%	
PAN	↓ ~ 90 pptv	

(25, 98) elevated (sigma=0.664)

O_3	↑ > 2 ppbv	
OH	↑ ~ 10%	
PAN	↓ 30 pptv	

Conc = COMPLEX fcn(T, isoprene, soil NO_x, lightning No_x)

(Note that PAN changes will affect far-field distributions, also.)

Future work

LLNL is planning application studies with IMPACT:

- 3D, global
- Prognostic **troposphere** AND **stratosphere**
- High resolution meteorology from:

  Assimilated wind fields **OR** GCM

  2° latitude × 2.5°longitude

  46 vertical layers (to 65 km)

  Updated every 6 hours

- Contains fully prognostic stratospherere.

  Currently adding tropospheric chemistry

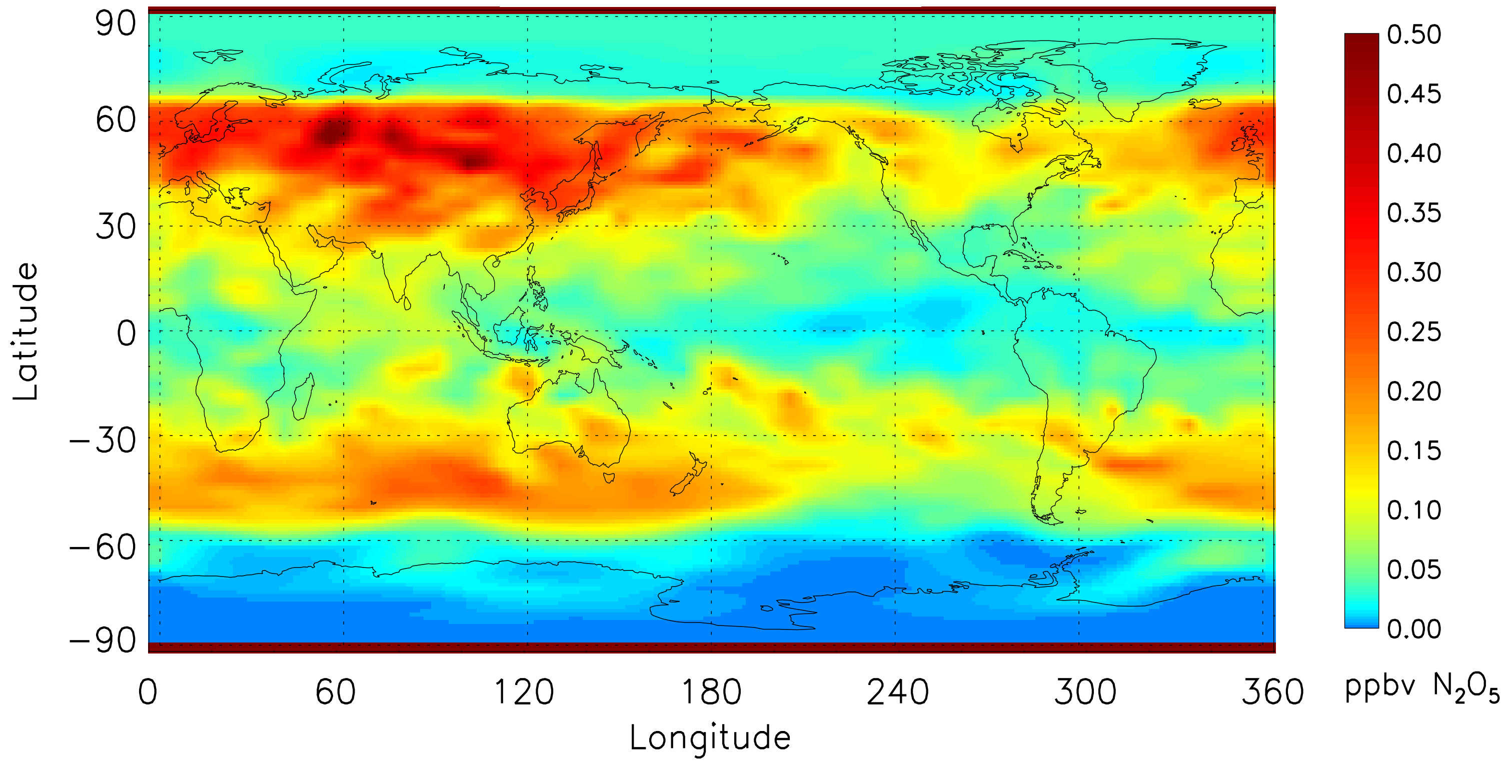
- Massively parallel (computer) code

Our proposed work includes:

- Adding heterogeneous reactions of NO_3 and N_2O_5 on aerosols
- Evaluating the role of natural versus anthropogenic sources of O_3
- Examining HC vs. NO_x limitations (regionally, seasonally)
- Preliminary assessment of effective mitigation strategies

----->Comparison with observations throughout<-----

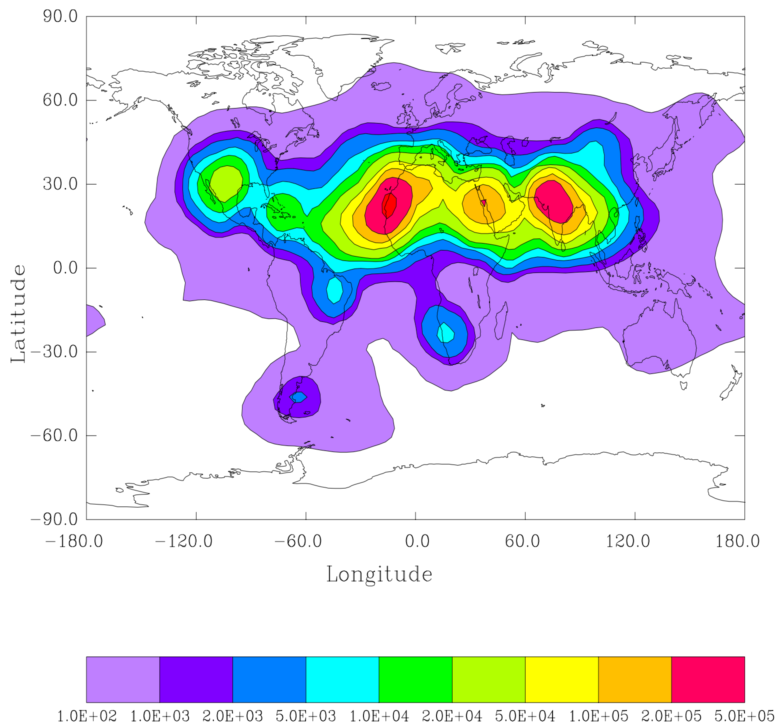
Below is the IMPACT simulated concentration of N_2O_5 at 55 hPa



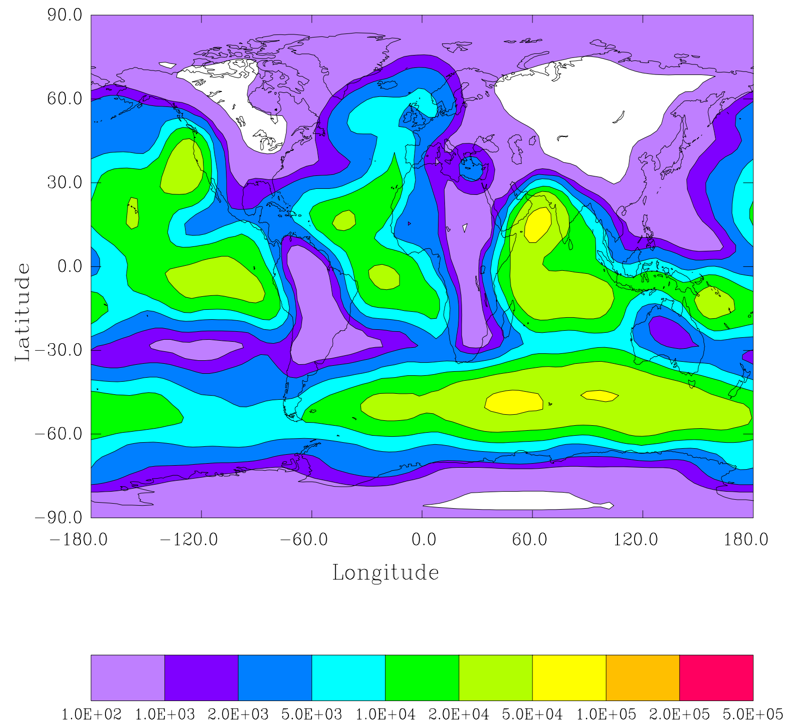
N_2O_5, ppbv, 3 August, midnight at Greenwich

Incorporating heterogeneous reactions of NO_3 and N_2O_5 on aerosols will affect chemical cycles (seasonally and regionally):

July mineral aerosol
(surface, ng/m^3)



July seasalt aerosol
(surface, ng/m^3)



Also: sulfate, carbonaceous aerosols

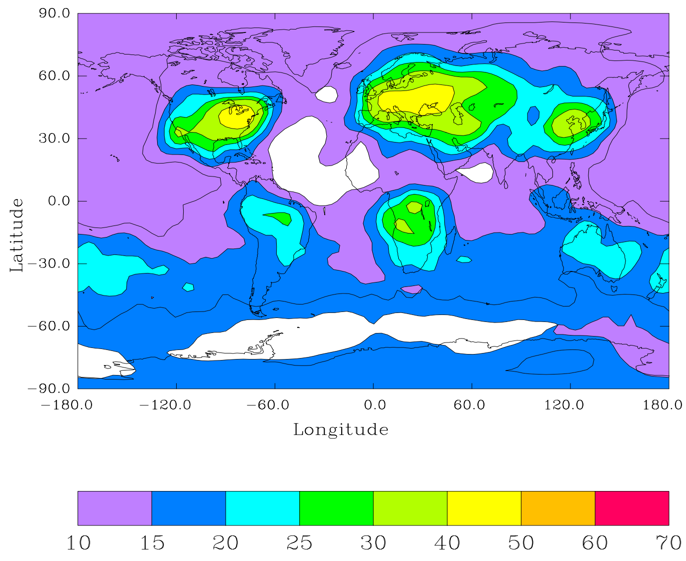
IMPACT will quantify natural vs. energy-related sources of O_3 (regional and seasonal variations):

Natural sources

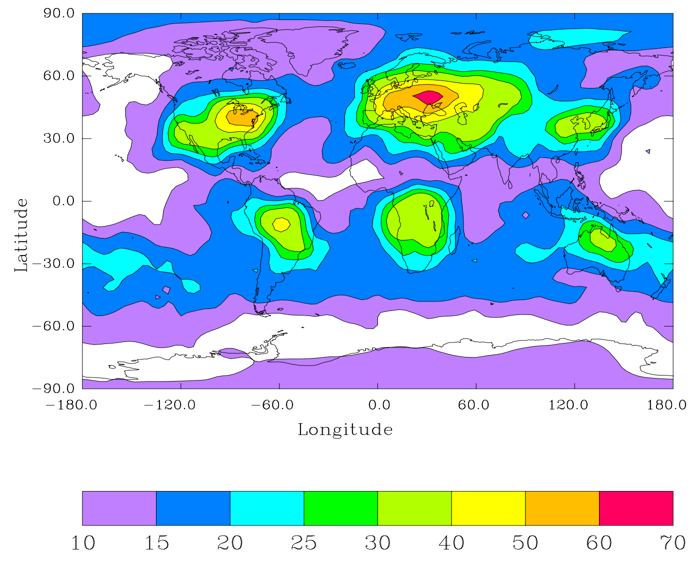
a) Natural HCs (isoprene, terpenes, CH_4, etc.) and CO

b) NO_x from lightning, soils

Preliminary GRANTOUR results - perpetual July - isoprene effect ONLY



CO, CH_4, All NO_x



Isoprene, CO, CH_4, All NO_x

IMPACT HC and/or NO_x sensitivity studies will address:

- Issues on a regional and seasonal basis
- Regions of HC and/or NO_x limitations
- Contribution of natural vs. energy-related emissions
- Effectiveness (O_3, etc.) of possible mitigation strategies

LLNL will further develop and apply global 3D models to help understand the role and importance of regional energy-related emissions on our atmospheric quality.

By understanding the role that both natural and anthropogenic emissions have, we can better develop energy-use and emission strategies to take us into the next century.