Integrated analysis of the impact of long-range transport of midlatitude pollution on ozone abundances in the Arctic troposphere

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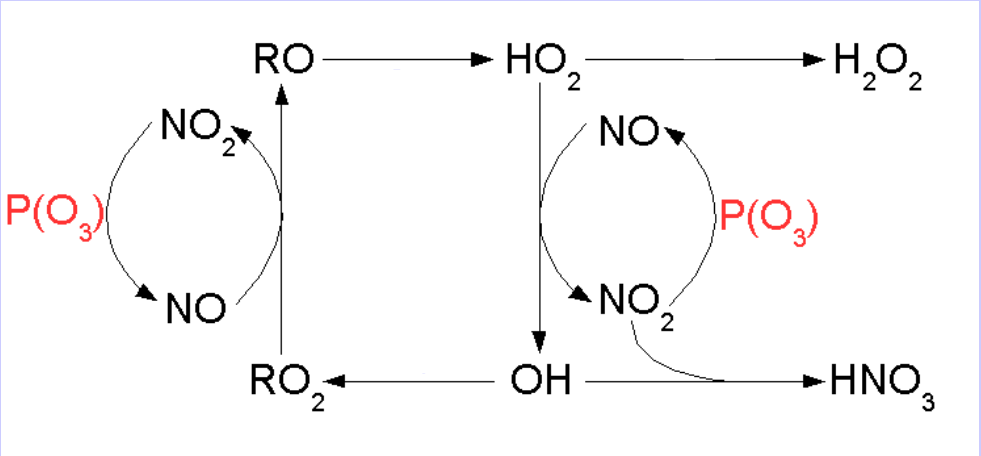
Tropospheric Ozone Chemistry



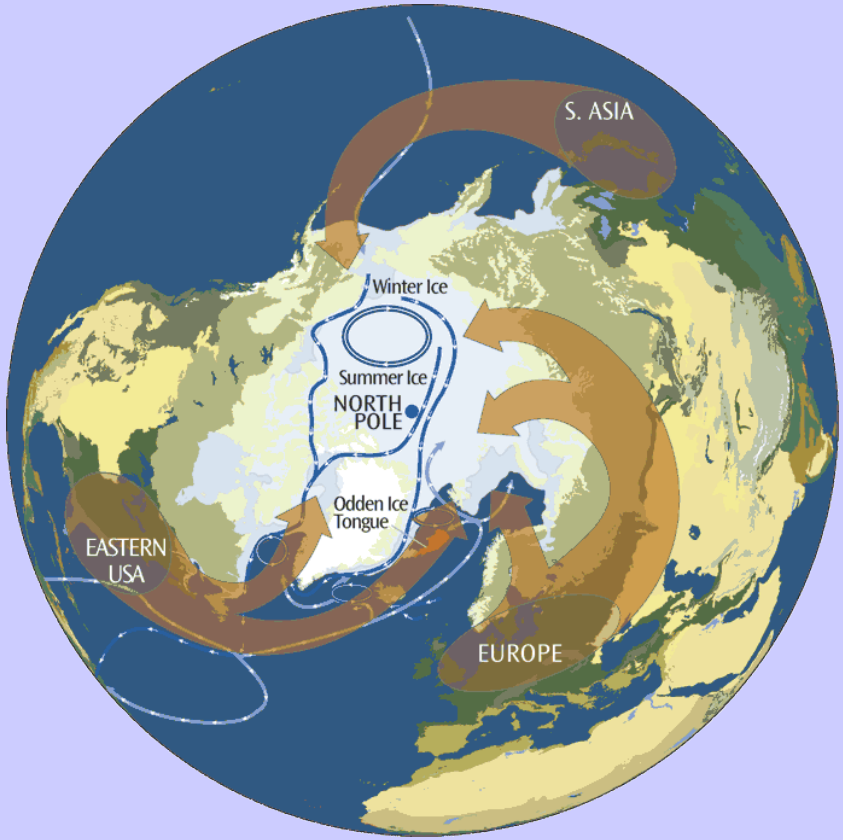
O_3 is not emitted directly

Produced through hydrocarbon oxidation in the presence of NO_x ($NO + NO_2$)

Atmospheric lifetime (O_3): ~3 weeks

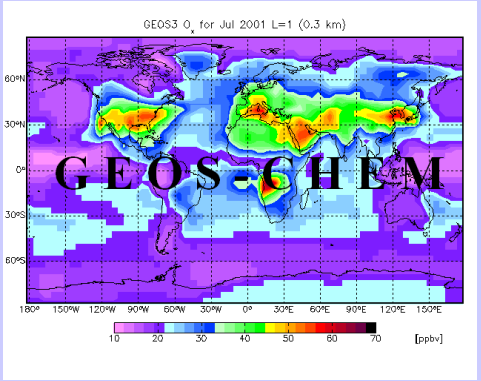
Atmospheric lifetime (NO_x): ~1-2 days

Ozone in the Arctic reflects either direct transport of ozone from lower latitudes and the stratosphere or in situ production (driven by NO_x transported into the region as PAN)



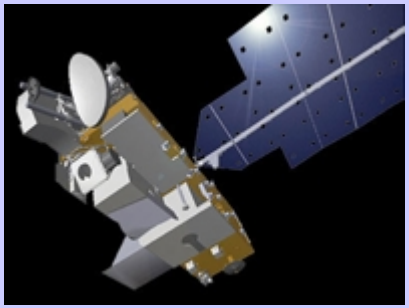
Model Description and Data Sources

GEOS-Chem: global chemical transport model



- NO_x-HO_x-VOC chemistry
- dynamics driven by GEOS meteorological data
- results shown here are from v7-02-04, resolution 4°x5° horizontal, 30 vertical levels
- tagged-O_x simulation permits regional source attribution
- adjoint permits detailed sensitivity analysis

Tropospheric Emission Spectrometer (TES):



- high resolution IR Fourier transform spectrometer
- 3.2 – 15.4 μm spectral range
- O_3 profiles assimilated into GEOS-Chem using suboptimal Kalman filter between 60°S - 60°N

Ozonesondes:

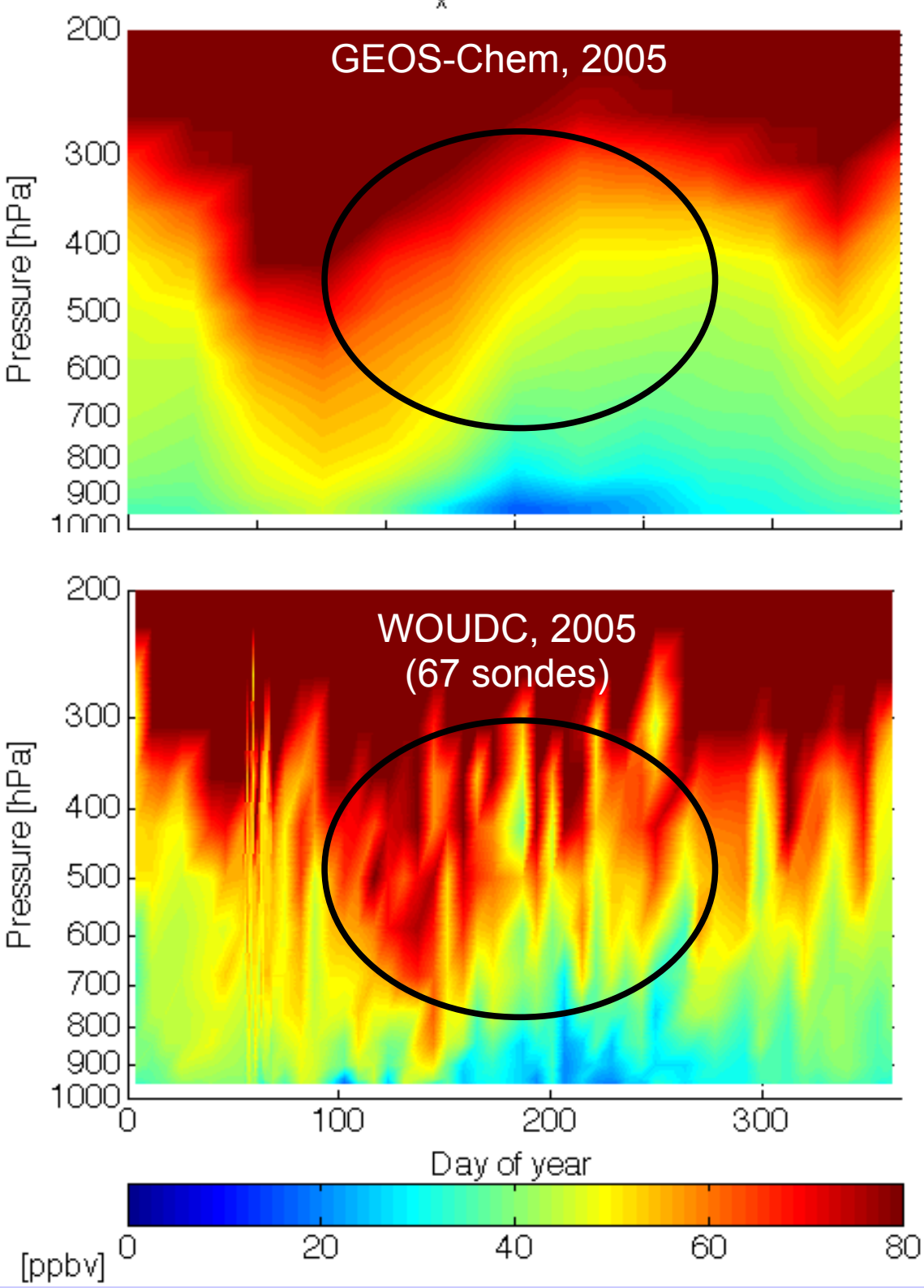
- vertical profiles of ozone, temperature, pressure
- launched roughly weekly, intense campaigns in spring

Surface measurements at Alert:

- hourly average O_3
- measurements of PAN every half hour

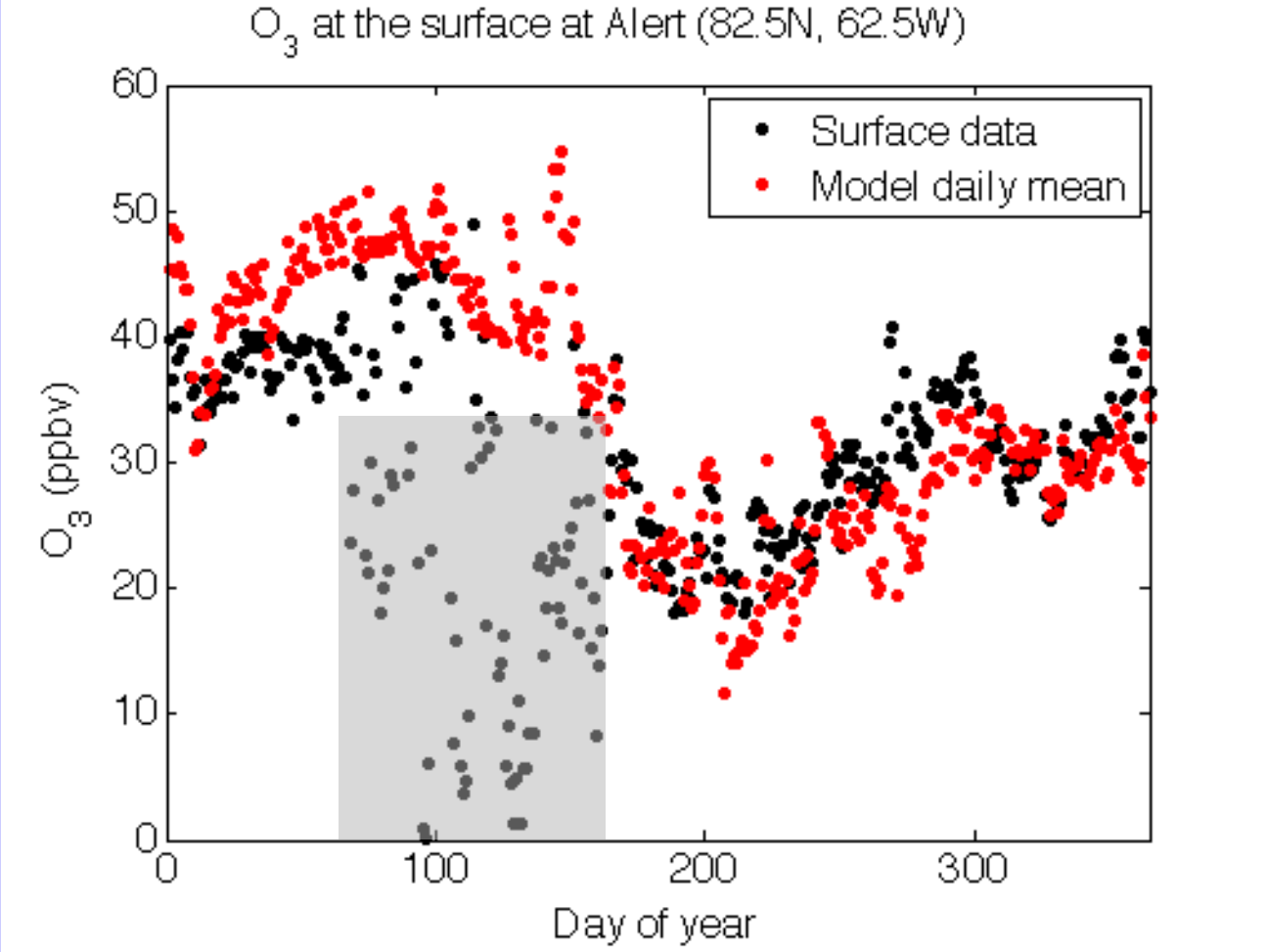
Monthly mean O_3 profiles above Eureka

- high ozone values in free troposphere (eg. ~500 hPa) appear in model earlier than observed
- the model significantly underestimates ozone abundances in the middle and upper troposphere in summer



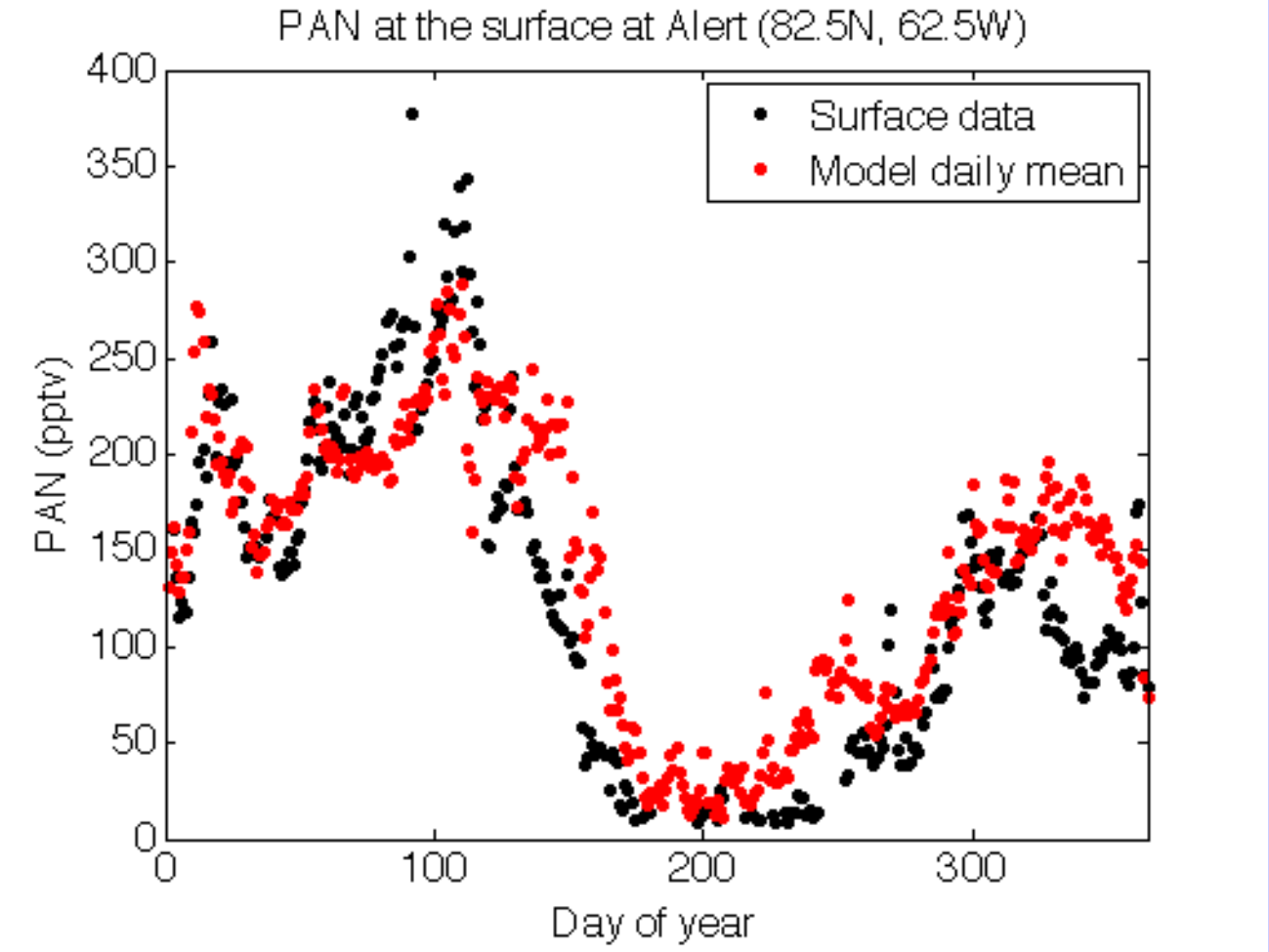
Model Simulation of Surface O_3: Agreement with Surface Data

- model reproduces seasonal cycle in surface ozone at Alert (2001)
- highest values in spring; lowest in summer
- slight overestimate in early spring; slight underestimate in fall
- depletion events due to halogen radical chemistry not represented in model



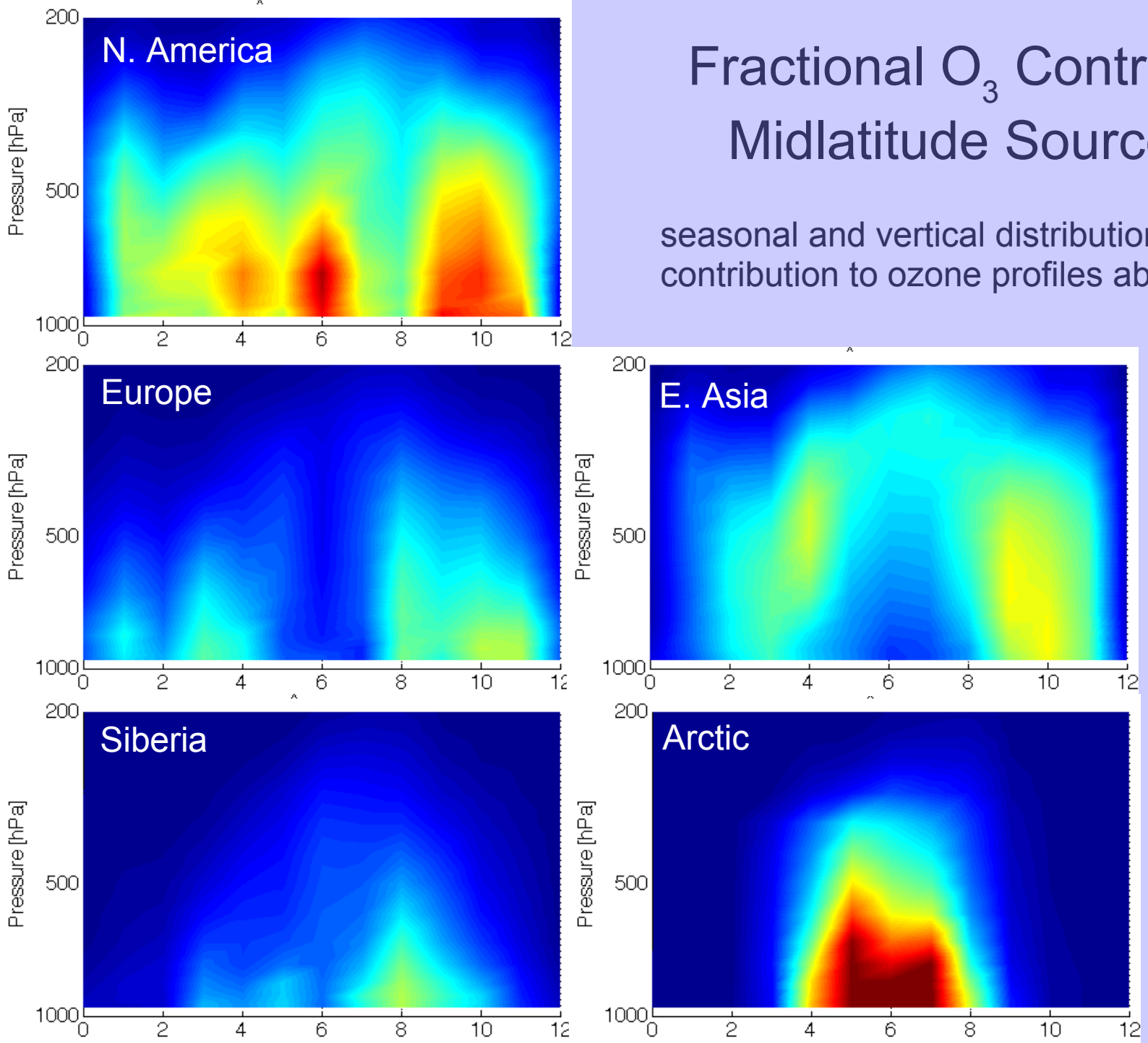
Model Simulation of Surface PAN: Agreement with Surface Data

- model reproduces seasonal cycle in surface PAN at Alert (2001)
- accumulation during winter leading to spring maximum; sharp drop to very low values in summer
- slight underestimate in spring
- seasonal cycle is apparent across the region, not just at Alert



Fractional O_3 Contribution from Midlatitude Source Regions

seasonal and vertical distributions of the fractional contribution to ozone profiles above Eureka (2005)

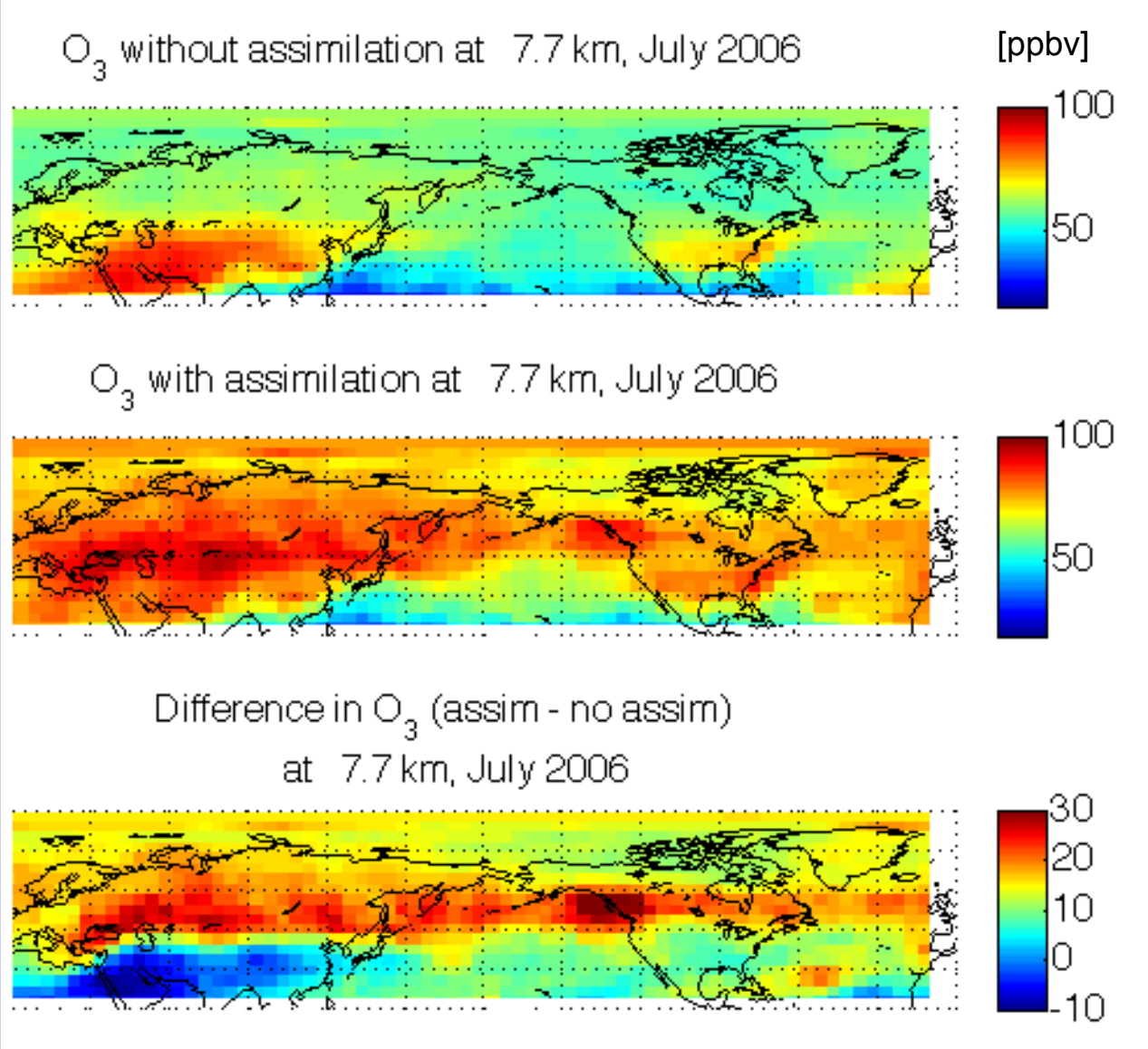


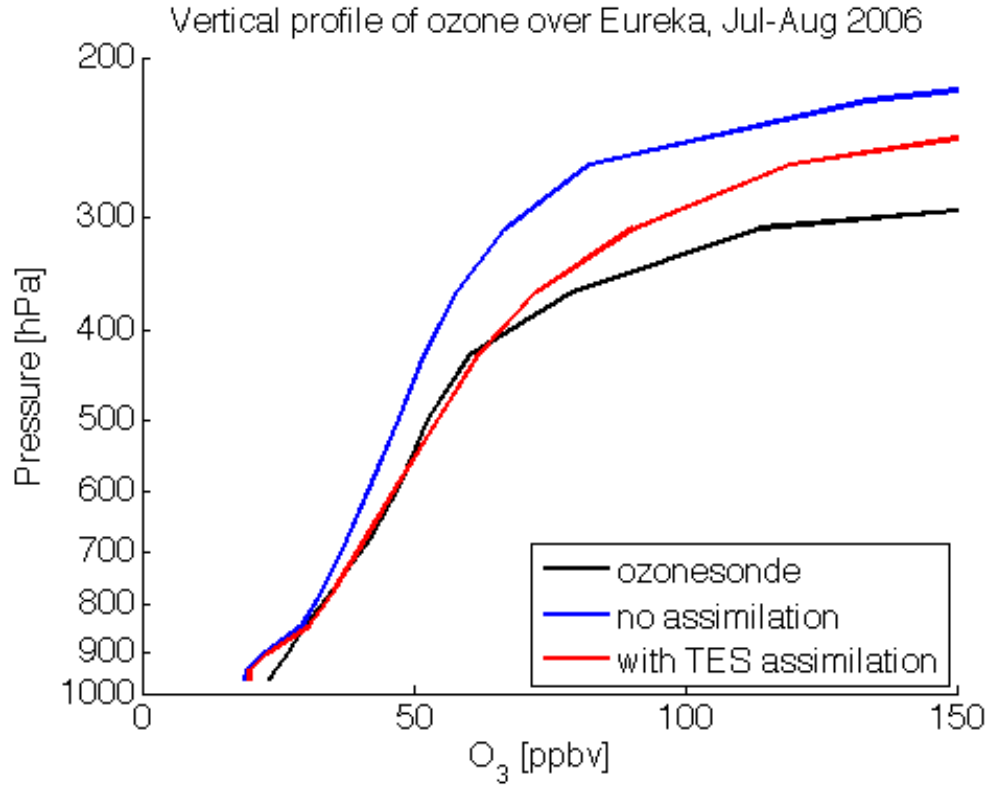
- in summer the dominant midlatitude contributions are from North America and Siberia

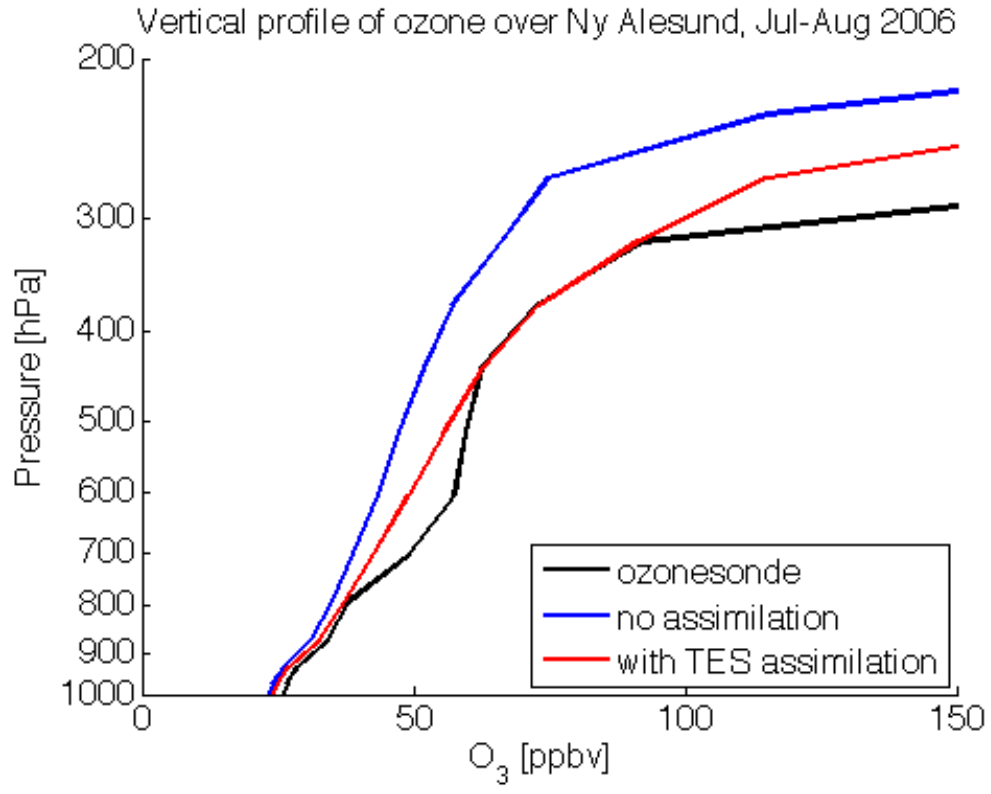
Satellite constraints on ozone transport into the Arctic

TES O_3 profiles are assimilated into GEOS-Chem equatorward of 60° between July 1 – Aug 31, 2006

- Parrington et al. [JGR, 2008] showed that over North America, the assimilation reduced the mean differences between the model and ozonesondes to less than 5% in the middle troposphere
- assimilation of TES O_3 increases concentrations across Northern mid-latitudes, with largest enhancements over Siberia and North America
- The assimilation provides an improved description of tropospheric O_3 at midlatitudes and thus an improved boundary condition for O_3 transport into the Arctic



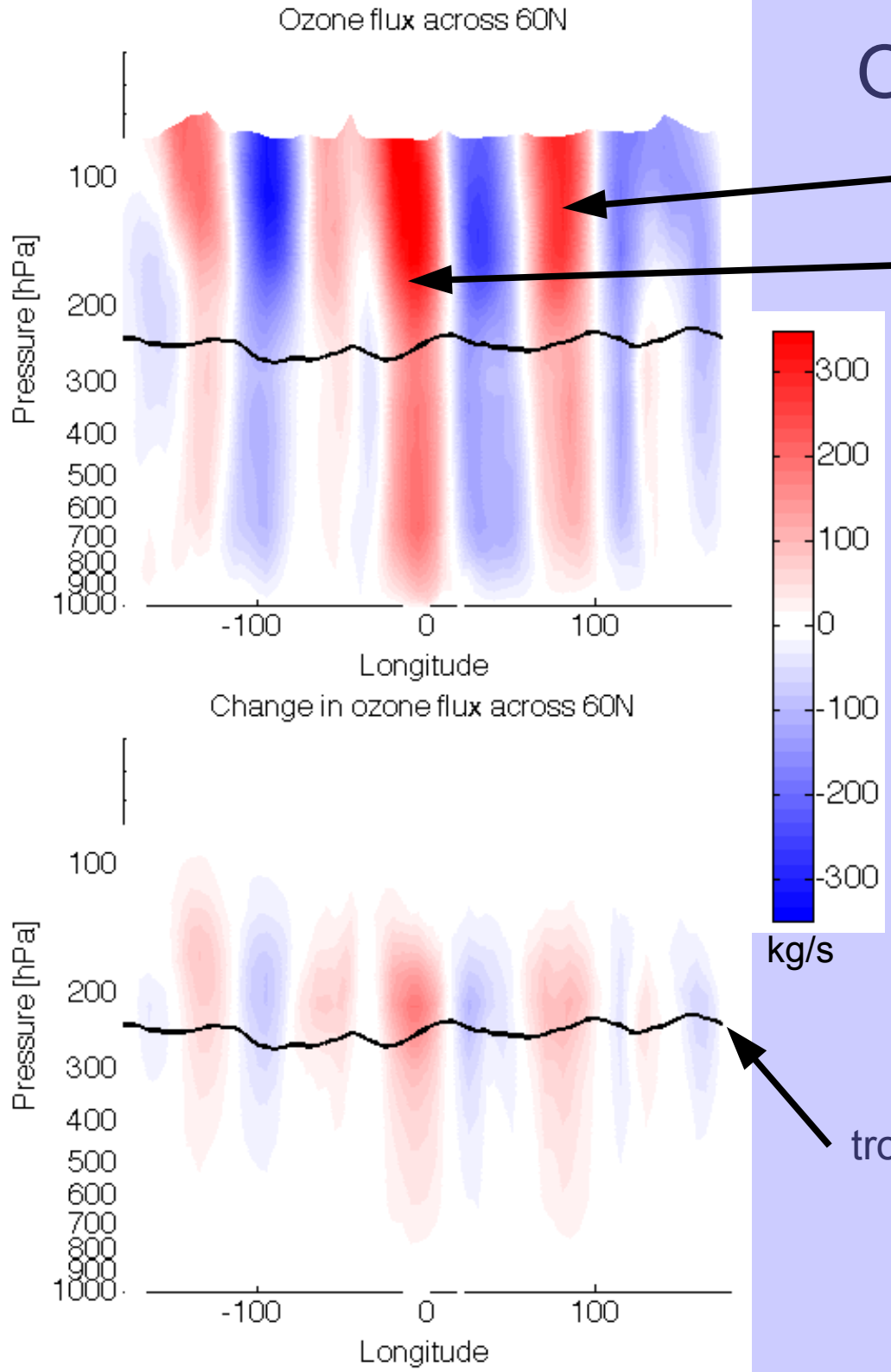




Assimilation of O_3 at midlatitudes corrects high latitude bias

- assimilation of TES O_3 results in significant correction to free troposphere ozone at high latitude sites
- mean bias between 400-500 hPa is reduced from +7.1 to -1.9 ppbv at Eureka, and from +11.5 to +1.9 ppbv at Ny Alesund
- little change to boundary layer

Ozone fluxes into the Arctic



above Siberia

above North Atlantic

tropopause

- monthly mean flux during July 2006
- change to flux due to assimilation is concentrated in the upper troposphere and lower stratosphere
- monthly mean net northward flux across 60°N changes from 1900 to 9700 kg/s with the assimilation
- net downward flux across 300 hPa north of 60°N changes from 320 to 760 kg/s with the assimilation (not shown)

Summary of Results

- model reproduces surface ozone and PAN seasonal cycles, but underestimates free tropospheric ozone in the summer
- midlatitude continental source regions do contribute fractions of simulated ozone at Eureka, North American influence being dominant
- assimilation of ozone data from TES into GEOS-Chem equatorward of 60^{o}N enhances midlatitude ozone abundances in the model, particularly over North America and Siberia, which reflects, in part, stratosphere-troposphere exchange captured by the TES measurements.
- improved midlatitude boundary conditions in the assimilation results in increase transport of ozone into the Arctic, mainly in the UTLS; the mean bias in ozone relative to ozonesondes decrease from +9.2 to -0.1 ppbv between 400-500 hPa over Eureka and Ny Alesund
- assimilation increases net horizontal flux across 60^{o}N, and increases net downward flux in the Arctic
  - what is the source of the increased flux? STE? tropospheric production?

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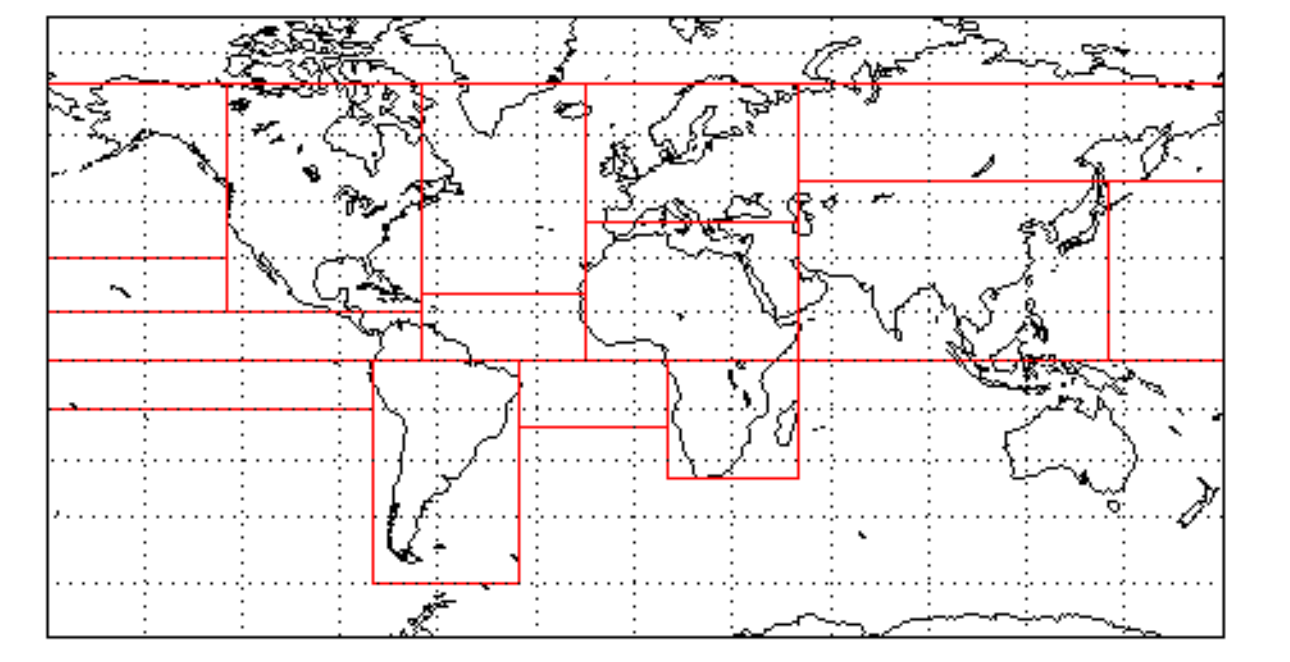
EXTRAS



Source regions for tagged O_x simulation

Why Tropospheric Ozone in the Arctic?

1. RADIATIVE IMPACT

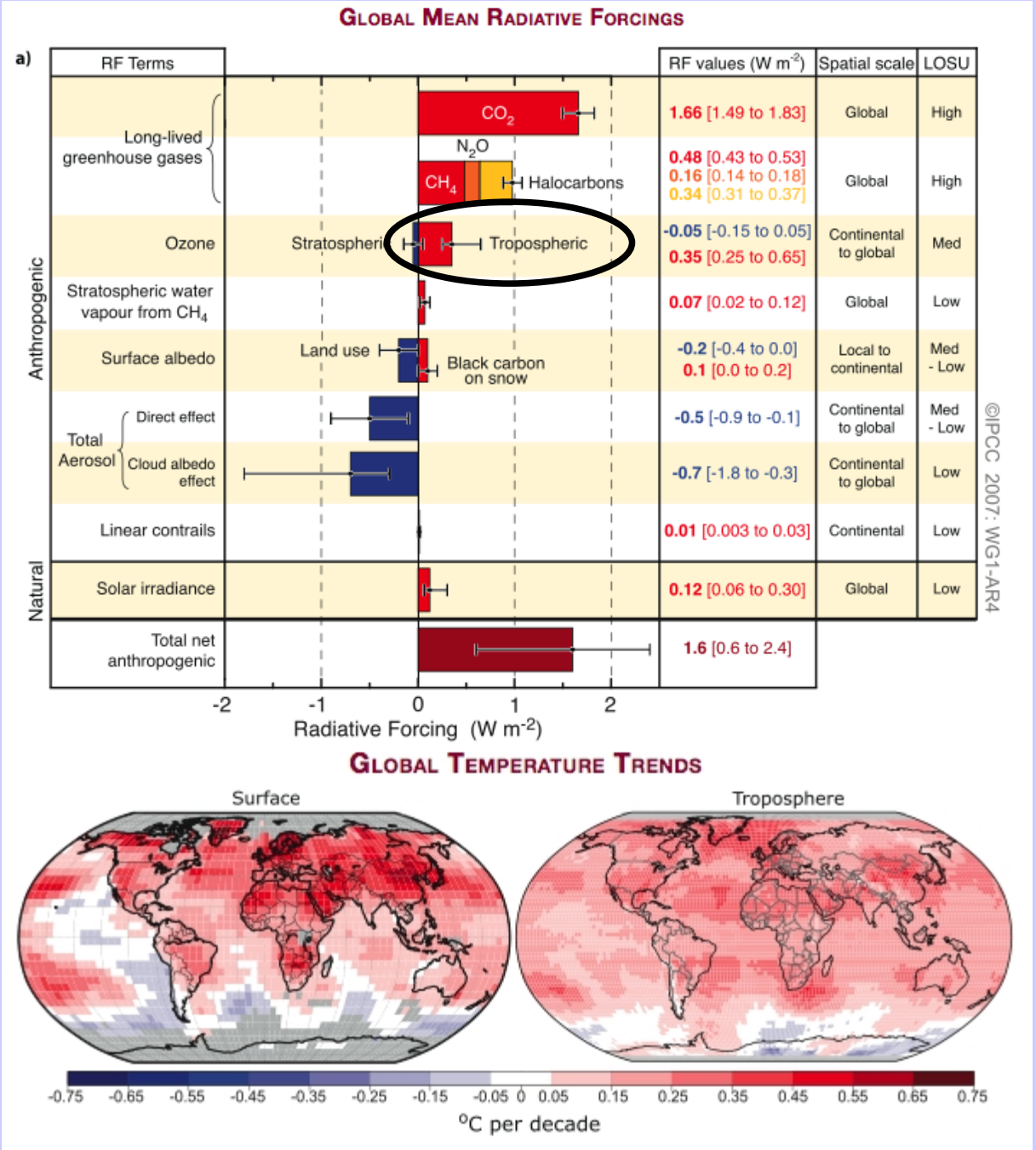
- direct greenhouse gas
- affects atmospheric lifetime of other greenhouse gases
- expected to increase

2. SENSITIVE ENVIRONMENT

- largest temperature trends are observed at high latitudes

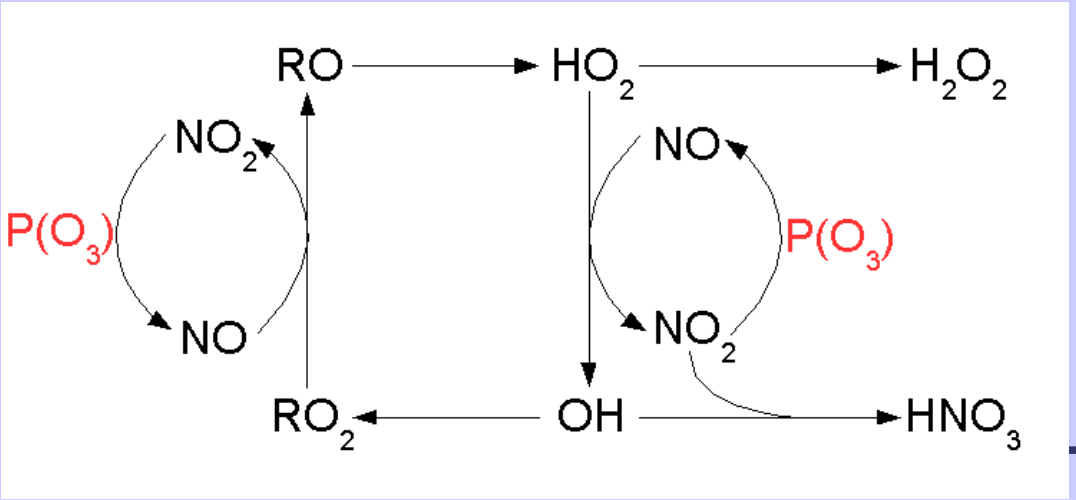
3. AIR QUALITY

- constituent of photochemical smog and of “Arctic haze”



from IPCC AR4, 2007

Ozone Production



$RH + OH \xrightarrow{(O2)} RO_2 + H_2O$ (R1)

$RO_2 + NO \rightarrow RO + NO_2$ (R2)

$RO + O_2 \rightarrow R'CHO + HO_2$ (R3)

$HO_2 + NO \rightarrow OH + NO_2$ (R4)

$2(NO_2 + hv \xrightarrow{(O2)} NO + O_3)$ (R5)

$RH + 4O_2 \rightarrow R'CHO + 2O_3 + H_2O$

hydrocarbon-limited case:

termination reaction

$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$ (R6)

yielding

$P(O_3) = 2k_4 \{P(HO_x)/k_6\}^{1/2} [NO]$

NO_x-limited case:

termination reaction

$HO_2 + NO + M \rightarrow HNO_3 + M$ (R7)

yielding

$$P(O_3) = \frac{2k_1 P(HO_x)[RH]}{k_7[NO_2][M]}$$

Adjoint model

Consider a linear expansion about a background set of inputs x_o:

$$y = M(x) = M(x_o) + \frac{\partial M}{\partial x}(x - x_o)$$

where $\frac{\partial M}{\partial x}$ is the tangent linear model (TLM).

Given a small perturbation Δx, the TLM can calculate the perturbation to the state Δy:

$$\Delta y = TLM(\Delta x)$$

Likewise, given the perturbed state, the adjoint model (transpose of TLM) calculates the required change in inputs

$$\Delta x = TLM^T(\Delta y)$$

Example: Δy could be the difference between simulated and observed ozone and Δx could be the required change in NO_x emissions to obtain that perturbed state

References

Parrington, M. et al. [2008], Estimating the summertime tropospheric ozone distribution over North America through assimilation of observations from the Tropospheric Emission Spectrometer. JGR, 113 (D18307), doi:10.1029/2007JD009341.