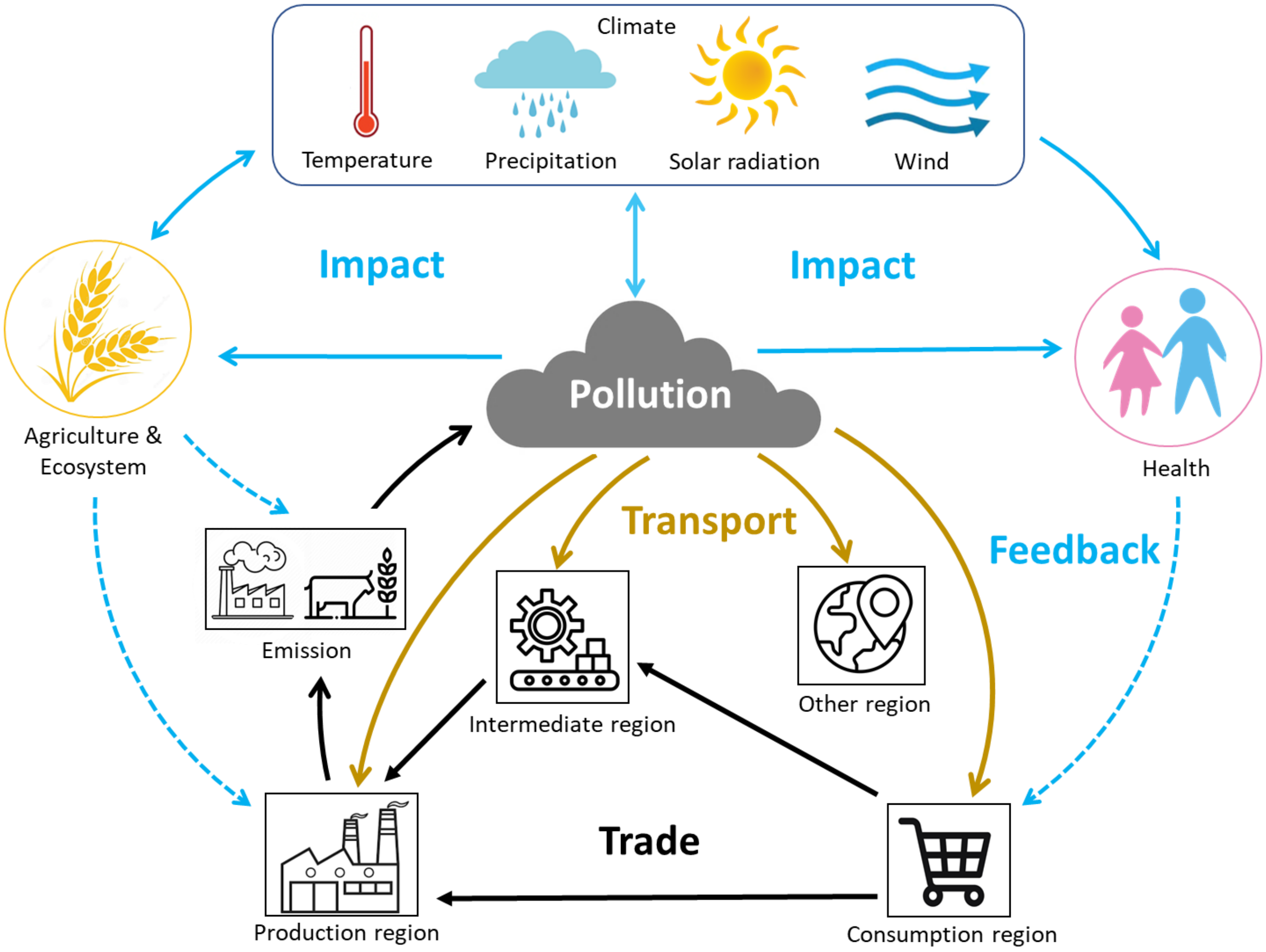
# Chapter 5

# Regional and Global Pollution Transport

# Quiz

1. Based on the collision theory, why does the reaction rate constant of bimolecular reactions tend to increase with T?
2. Can we consider $NO_2$ and ozone to be in the same chemical family (i.e., Ox)?
3. Potential human influences on recent tropospheric OH trends
4. How would NOx emissions affect the lifetime of OH?
5. Impacts of NOx on ozone at different cases: troposphere, boundary layer, urban, rural
6. Ozone production is normally VOC-limited in urban areas and NOx-limited in surrounding rural areas. To control urban ozone pollution, should we control NOx or VOC emissions?
7. How would changes in NOx affect the formation of nitrate and sulfate?
8. Why did China's $PM_{2.5}$, but not ozone, pollution decline with emission control over the past decade?
9. How can ozone and PM pollution affect each other?

# Globalizing Air Pollution

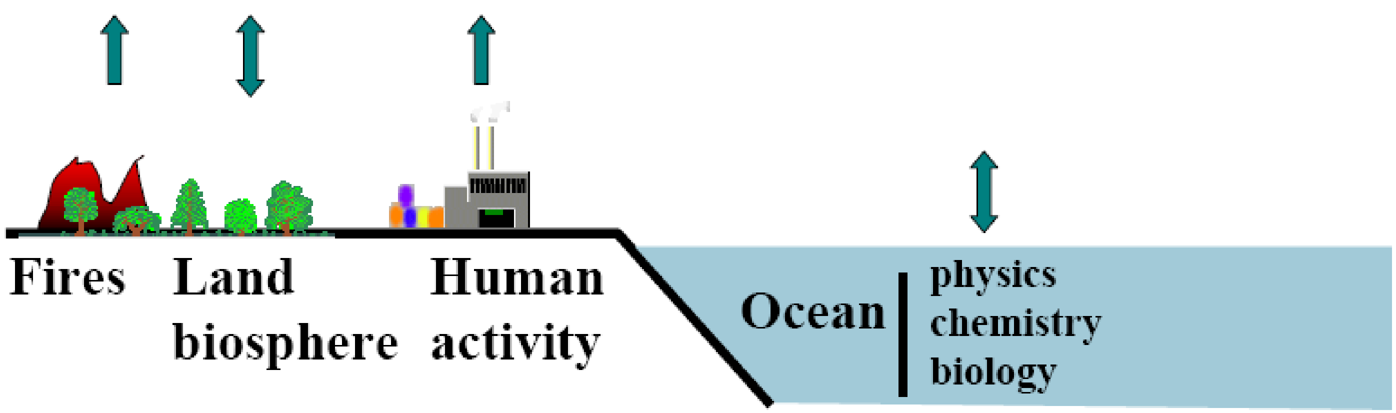



Lin et al., under review

# Budget of Air Pollutants

## AN ATMOSPHERIC CHEMIST'S VIEW OF THE WORLD


$$\frac{\partial C}{\partial t} = \overbrace{E}^{\text{Emis}} - \overbrace{D}^{\text{Dep}} - \overbrace{\nabla \cdot C\mathbf{V} - \nabla \cdot \overline{C'\mathbf{V}'}}^{\text{Transport \& Mixing}} + \overbrace{(P - L)}^{\text{Chemistry}}$$






Modified from Don Wuebbles' Lecture

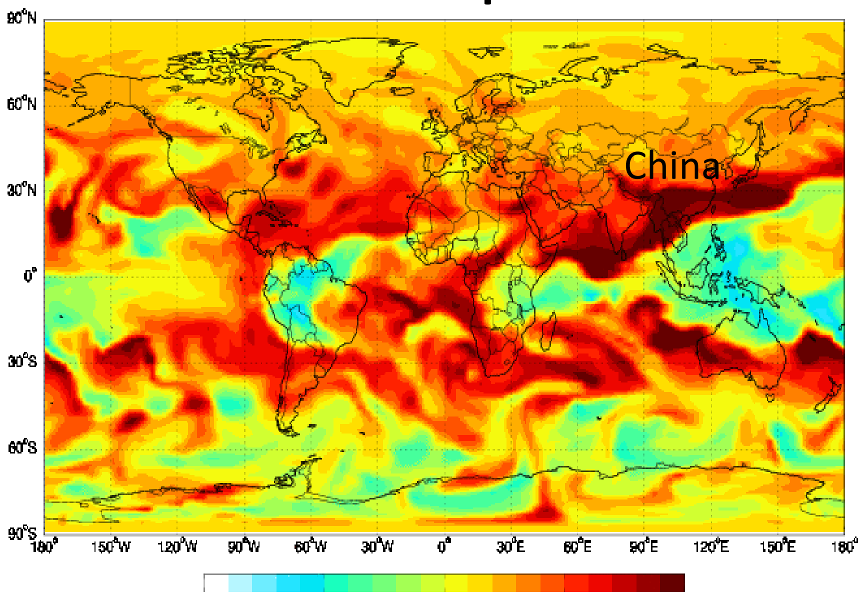
# Haze: Transported or Locally Formed ?

https://v.qq.com/x/page/f03620mzezq.html

# Globalizing Air Pollution:  Atmospheric Transport

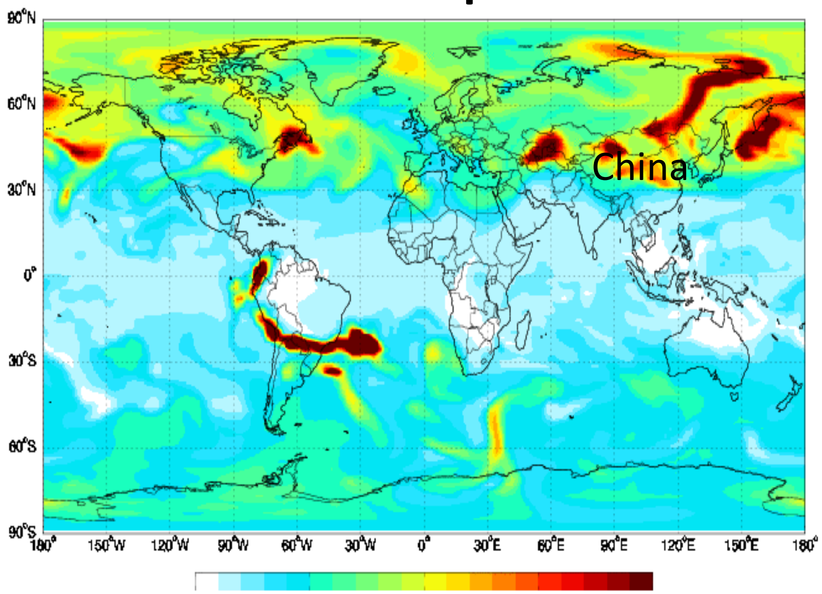
## Simulated by GEOS-Chem Chemical Transport Model

**Ozone in Mid-Trop. in Jan 2009**




0 20 40 60 80 ppb

**Sulfate in Mid-Trop. in Jan 2009**




0 5 10 15 20 x $10^{-2}$ ppb

- **Both local sources and transport of pollution are obvious**
- **The extent of transport depends on emissions, chemistry, etc.**
- **China is both a *source* and a *receptor* region**

Yan et al., 2014 ACP; 2016 ACP

# Sources of Air Pollution

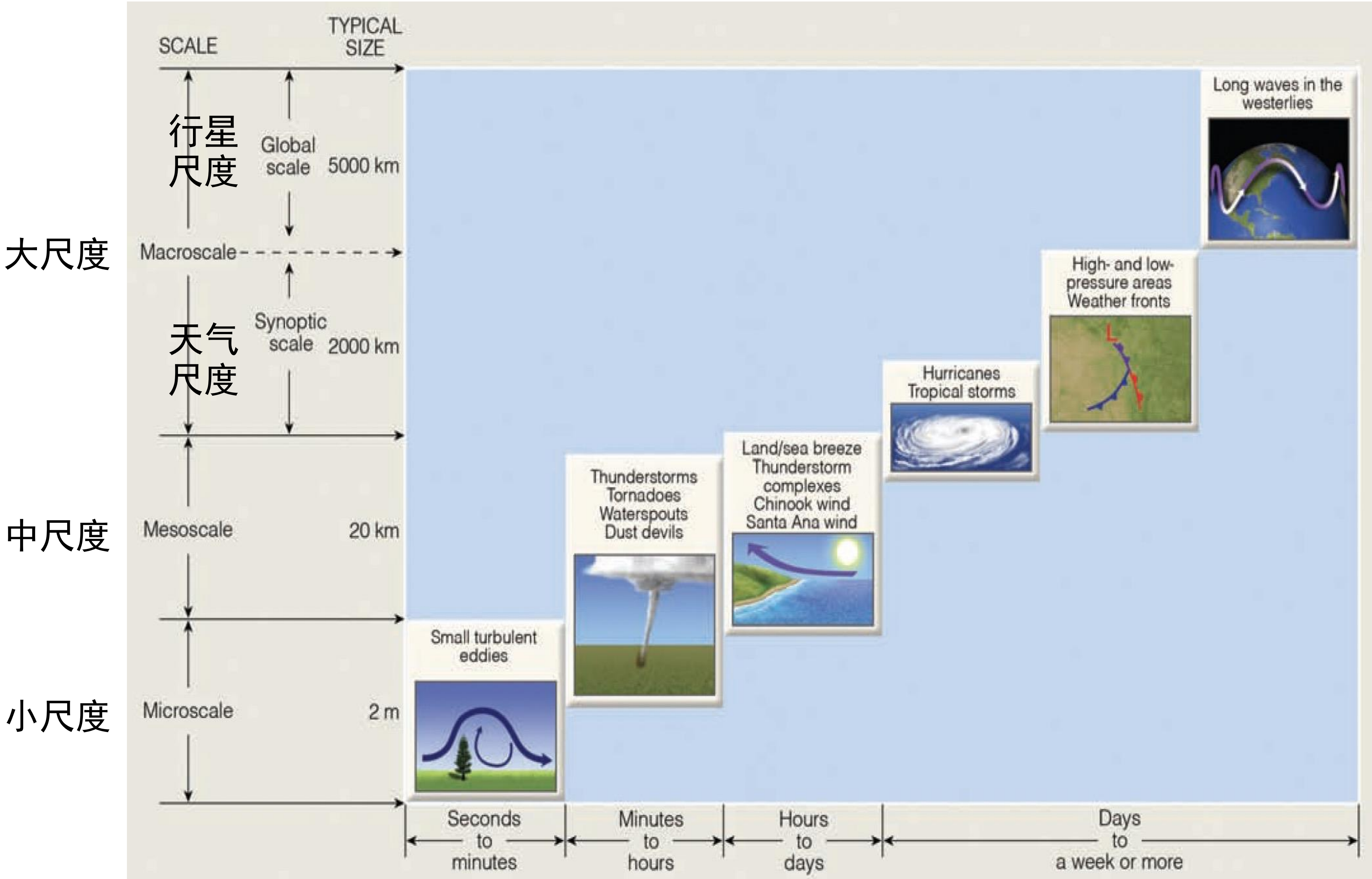
- Local emissions and/or production
- Inter-regional transport and transformation
- Global transport and transformation
- Stratosphere-troposphere exchange

❖ Natural versus anthropogenic sources

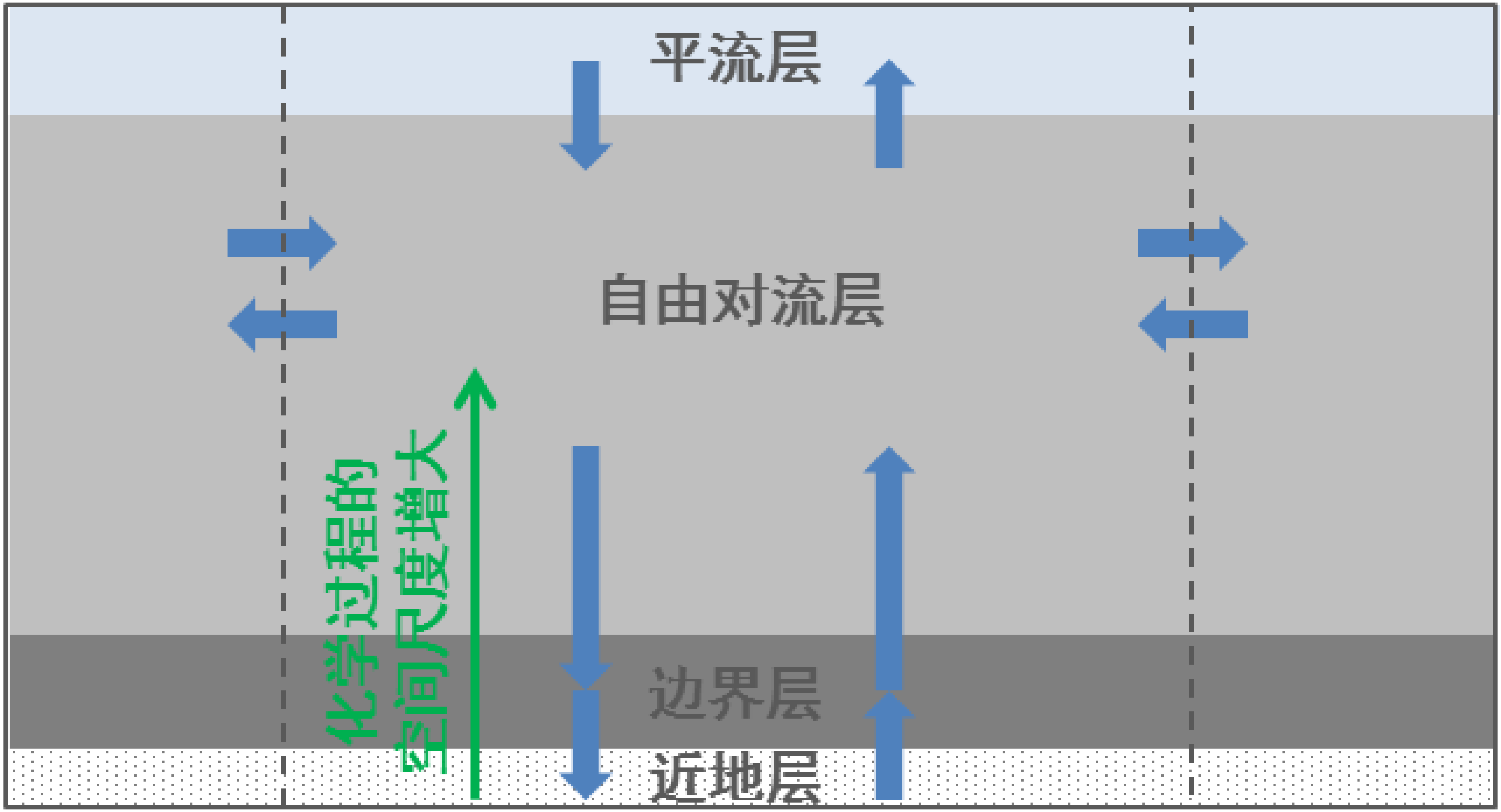
➢ Transport and transformation of air pollutants along the pathway

➢ Lifetime of pollutants is a key factor

# Spatiotemporal Scale of Atmospheric Motion (Transport)



# Local-Regional-Global Pollution Interconnection



# Characteristic Distance of Transport

- **Primary Pollutant:**

  $$D = U \times \tau = \text{Wind Speed} \times \text{Lifetime}$$

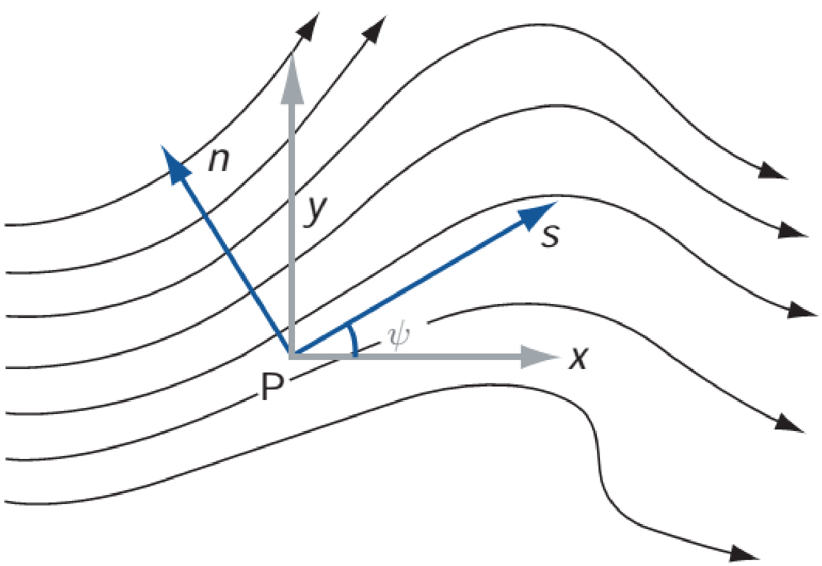
- **Secondary Pollutant:**

  $$D = U \times \tau^*, \text{ where } \tau_s < \tau^* < \tau_p + \tau_s$$

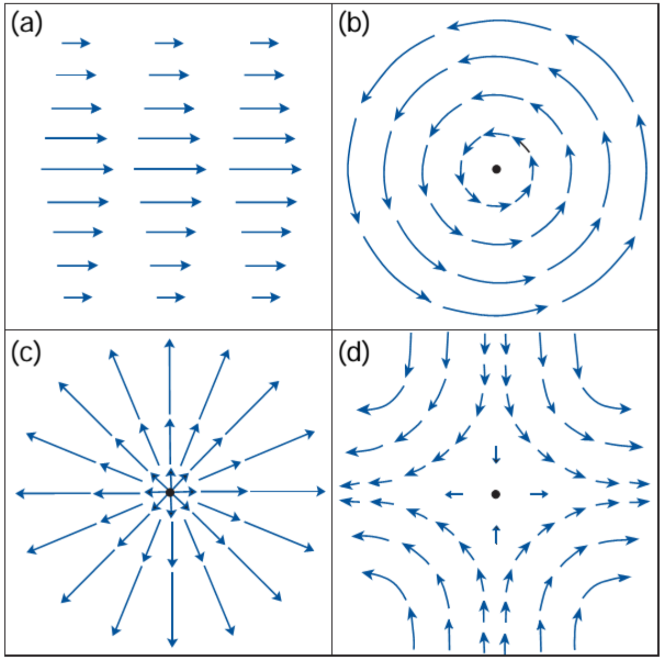
**$\tau^*$: Characteristic time**
**$\tau_p$: Lifetime of primary pollutants in conversion to secondary pollutants**
**$\tau_s$: Lifetime of secondary pollutants**

**e.g., Emission → [$SO_2$] → [$SO_4$] → deposition**
**Recall: NO emission → [NO] → [$NO_x$] → [$NO_z$] ?**

# Kinematics: Different Types of Flow Motions




| | | Vectorial | Natural coords. | Cartesian coords. |
|---|---|---|---|---|
| *Shear* | 切变 | | $-\dfrac{\partial V}{\partial n}$ | |
| *Curvature* | 曲率 | | $V\dfrac{\partial \psi}{\partial s}$ | |
| *Diffluence* | 分流 | | $V\dfrac{\partial \psi}{\partial n}$ | |
| *Stretching* | 拉伸 | | $\dfrac{\partial V}{\partial s}$ | |
| *Vorticity* $\zeta$ | 涡度 | $\mathbf{k} \cdot \nabla \times \mathbf{V}$ | $V\dfrac{\partial \psi}{\partial s} - \dfrac{\partial V}{\partial n}$ | $\dfrac{\partial v}{\partial x} - \dfrac{\partial u}{\partial y}$ |
| *Divergence* $\mathrm{Div}_H \mathbf{V}$ | 散度 | $\nabla \cdot \mathbf{V}$ | $V\dfrac{\partial \psi}{\partial n} + \dfrac{\partial V}{\partial s}$ | $\dfrac{\partial u}{\partial x} + \dfrac{\partial v}{\partial y}$ |
| *Deformation* tensor | 形变 | | | $\dfrac{\partial u}{\partial x} - \dfrac{\partial v}{\partial y};\ \dfrac{\partial v}{\partial x} + \dfrac{\partial u}{\partial y}$ |

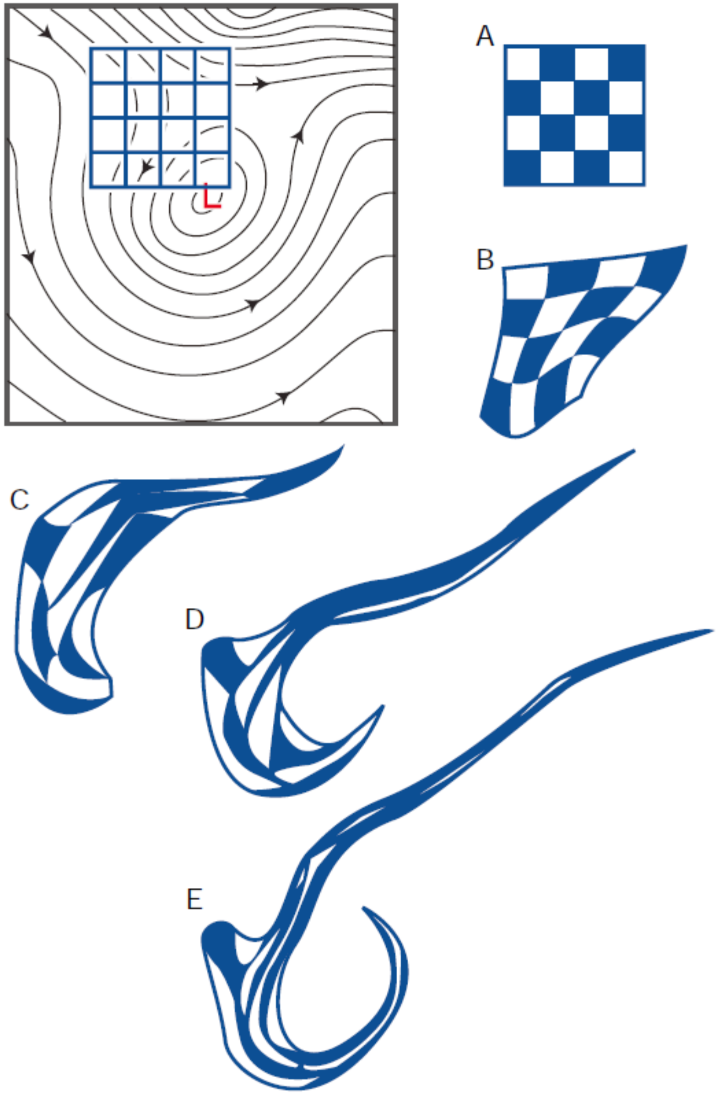



| | a | b | c | d |
|---|---|---|---|---|
| Shear | Y | Y | N | Y |
| Curvature | N | Y | N | Y |
| Diffluence | N | N | Y | Y |
| Stretching | N | N | Y | Y |
| Vorticity | Y | Y | N | N |
| Divergence | N | N | Y | N |
| Deformation | Y | Y | Y | Y |



Wallace and Hobbs, 2006

# Kinematics: Deformation




How a grid of air parcel is deformed by the flow as the tagged particles move downstream with time:

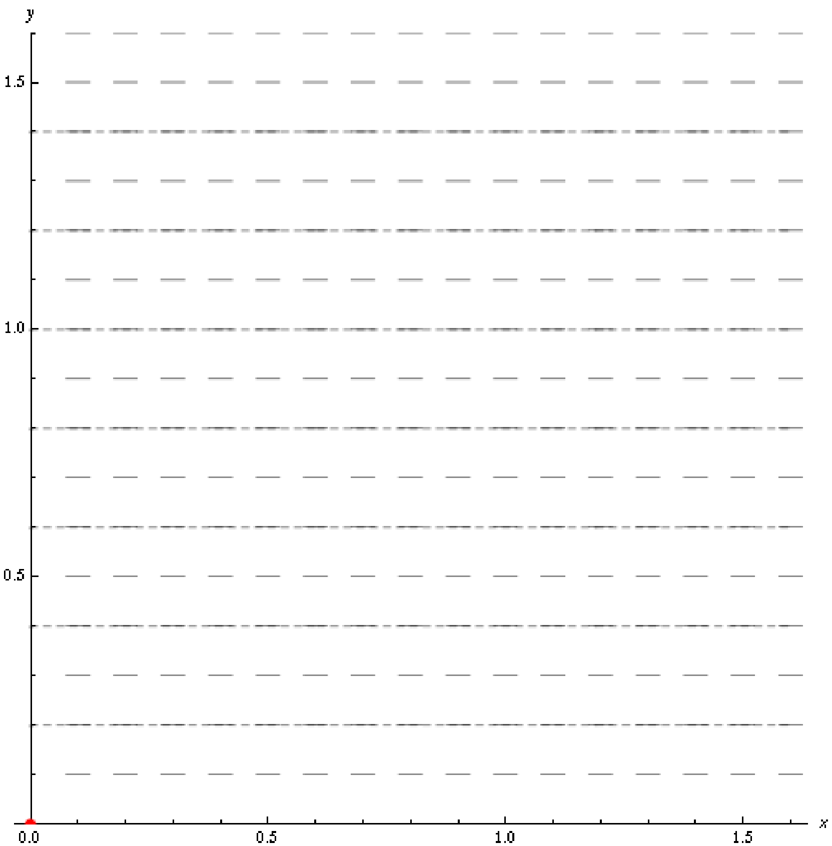
- ✓ Those in the upper right corner of the grid moving eastward.
- ✓ Those in the lower left corner moving southward and then eastward around the closed circulation.



Wallace and Hobbs, 2006

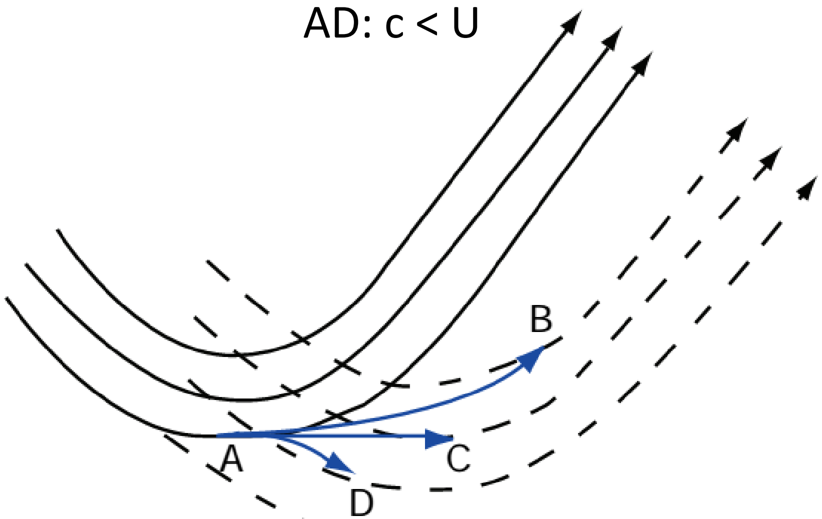
# Kinematics: Streamline versus Trajectory

**Pathline: 轨迹线**
**Streamline：流线（虚线）**
**Streakline：脉线**




https://en.wikipedia.org/wiki/File:Streaklines_and_pathlines_animation.gif

Initial streamlines: solid lines
Later streamlines: dashed lines
Phase speed of wave: c
Speed of background flow: U
AB: c > U
AC: c = U
AD: c < U




Wallace and Hobbs, 2006

# Real and Pseudo Forces in the Atmosphere

**Friction force** $\boldsymbol{F} = -\frac{1}{\rho}\frac{\partial \tau}{\partial z}$ $\boldsymbol{\tau} = -\rho C_D \boldsymbol{V} V$

$\tau$ = Shear Stress; $C_D$ = Drag coefficient

**Pressure gradient force** $\boldsymbol{P} = -\frac{1}{\rho}\nabla p$

**Gravity (Gravitation + Centrifugal)** $\boldsymbol{g} = \boldsymbol{g}^* + \Omega^2 \boldsymbol{R_A}$

**Coriolis force** $\boldsymbol{C} = -f\boldsymbol{k} \times \boldsymbol{V}$ $f = 2\omega_{earth} \sin\theta$

***Centrifugal force and Coriolis force are pseudo forces***



Wallace and Hobbs, 2006

# Horizontal Winds

$$\frac{d\boldsymbol{V}}{dt} = \boldsymbol{P} + \boldsymbol{C} + \boldsymbol{F}$$

$$= -\frac{1}{\rho}\nabla p - f\boldsymbol{k} \times \boldsymbol{V} + \mathbf{F}$$

*In hydrostatic balance:*

$$= -\nabla\phi - f\boldsymbol{k} \times \boldsymbol{V} + \mathbf{F}$$

$$\frac{du}{dt} = -\frac{\partial\phi}{\partial x} + fv + F_x$$

$$\frac{dv}{dt} = -\frac{\partial\phi}{\partial y} - fu + F_y$$

$\phi$ = geopotential $\quad d\phi = gdz$



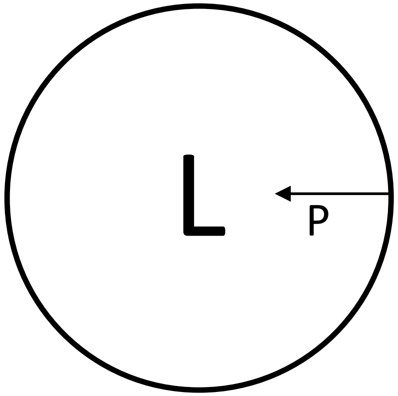
Wallace and Hobbs, 2006

# Rotational Wind (small-scale)

For small-scale motions, Coriolis force can be neglected

$$\frac{d\boldsymbol{V}}{dt} = \boldsymbol{n}\frac{V^2}{R_T} \approx -\nabla\phi$$

$$V_r = \sqrt[2]{(|\nabla\phi R_T|)}$$




The rotation could by cyclonic or anticyclonic

- ✓ $R_T > 0$ for cyclonic
- ✓ $R_T < 0$ for anticyclonic

For a typical midlatitude tornado, $R_T$ = 300 m, $V_r$ = 30 m s$^{-1}$

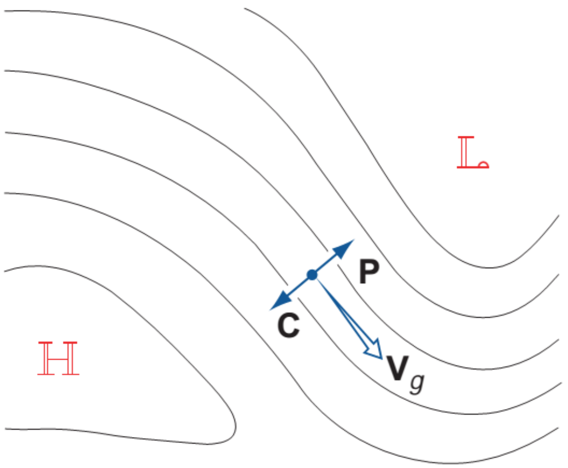
- ➢ The magnitude of Coriolis force ~ $10^{-3}$ m s$^{-2}$
- ➢ The magnitude of centripetal force ~ 3 m s$^{-2}$

# Geostrophic Wind (large-scale, without friction)

$$\frac{d\boldsymbol{V}}{dt} = \text{ } 10^{-4} \text{ m s}^{-2} \text{ in magnitude, about 10\% of Coriolis force}$$

Assume $\frac{d\boldsymbol{V}}{dt} \approx 0$

Pressure gradient force is in balance with Coriolis force, w/o friction



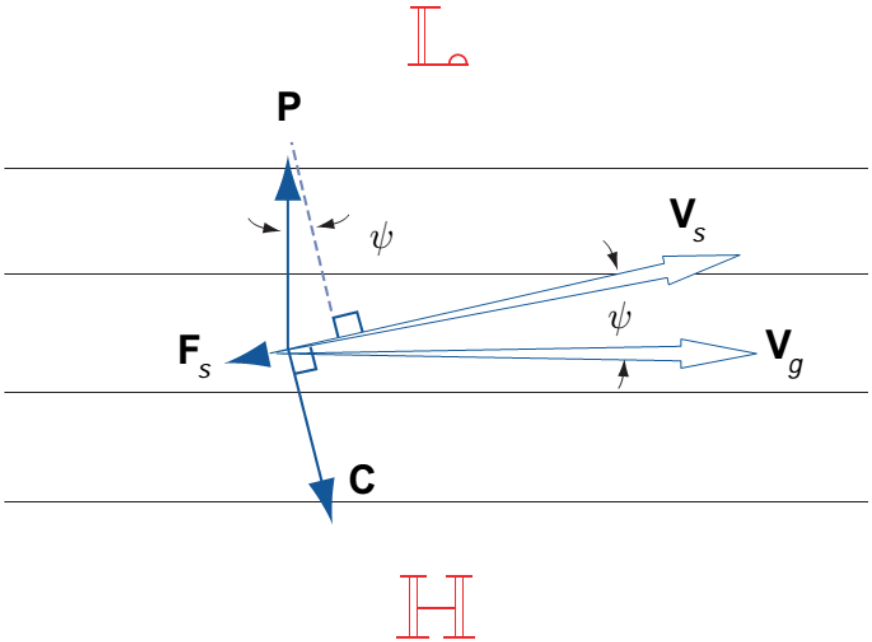

$$\nabla\phi = -f\boldsymbol{k} \times \boldsymbol{V}_g$$

$$\boldsymbol{V}_g = \frac{1}{f}\boldsymbol{k} \times \nabla\phi$$



Wallace and Hobbs, 2006

# Effect of Friction by Earth Surface




**P** = Pressure gradient force
**C** = Coriolis force
**Fs** = Friction force
**V**$_s$ = Wind
**V**$_g$ = geostrophic wind

$$fV_s = |\boldsymbol{P}|\cos\psi = |\nabla\phi|\cos\psi$$



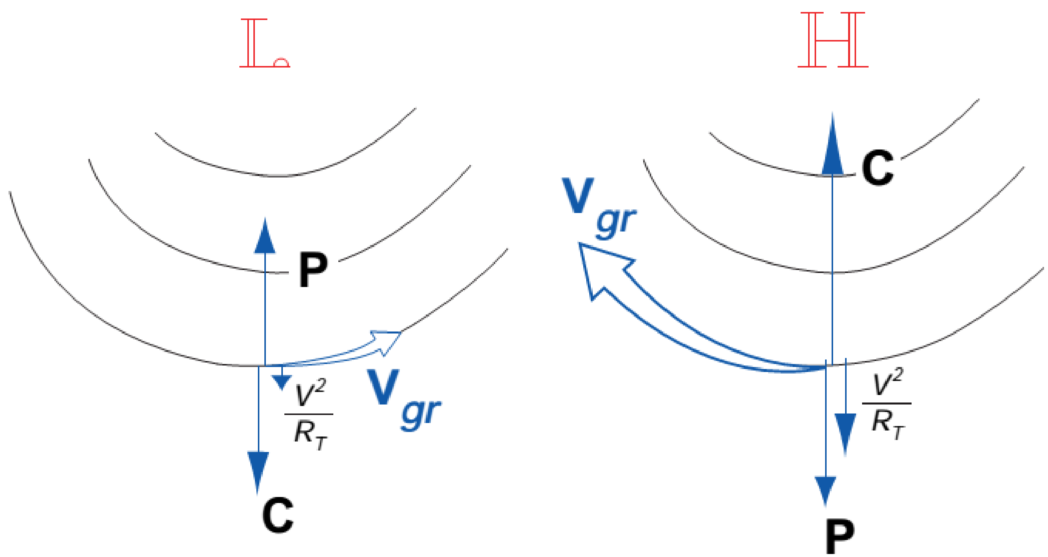
Wallace and Hobbs, 2006

# Gradient Wind (large-scale, without friction)

$$\frac{d\boldsymbol{V}}{dt} = \boldsymbol{n}\frac{V^2}{R_T} = -\nabla\phi - f\boldsymbol{k}\times\boldsymbol{V}$$

$$V_{gr} = \frac{1}{f}\left(|\nabla\phi| + \frac{V_{gr}^2}{R_T}\right)$$

$R_T > 0$ for cyclonic
$R_T < 0$ for anticyclonic




Subgeostrophic

Supergeostrophic



Wallace and Hobbs, 2006

# Thermal Wind (large-scale, without friction)

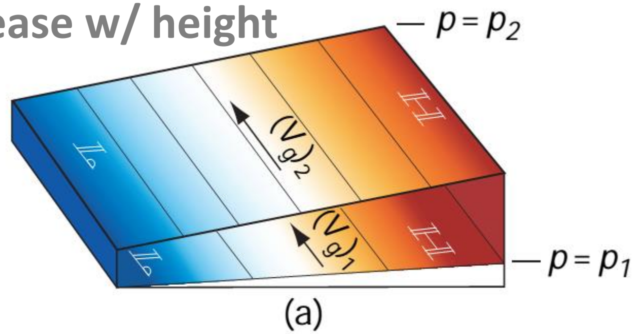
$$(\boldsymbol{V}_g)_2 - (\boldsymbol{V}_g)_1 = \frac{1}{f}\boldsymbol{k} \times \nabla(\phi_2 - \phi_1) \quad = \frac{g_0}{f}\boldsymbol{k} \times \nabla(Z_2 - Z_1)$$

$$= \frac{R}{f}\ln\frac{p_1}{p_2}\boldsymbol{k} \times \nabla\bar{T} \qquad \textit{Hydrostatic}$$

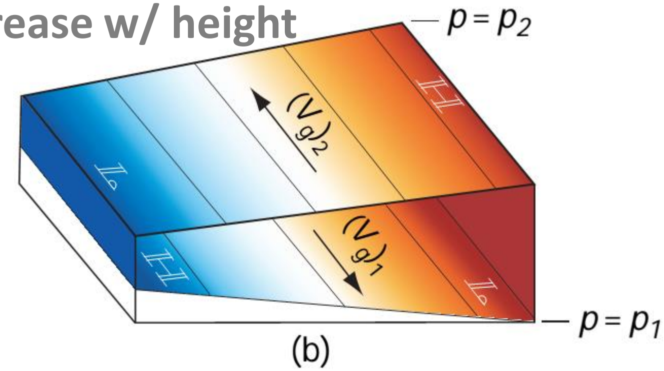
**Equivalent Barotropic**

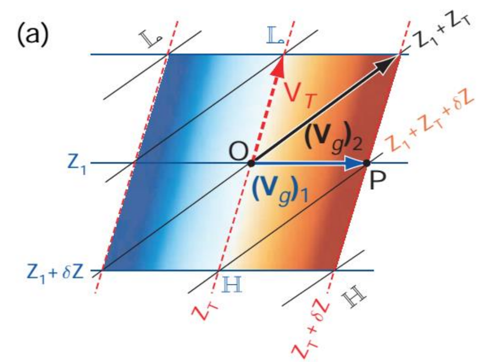
**Baroclinic: thermal advection**

**$V_g$ increase w/ height**

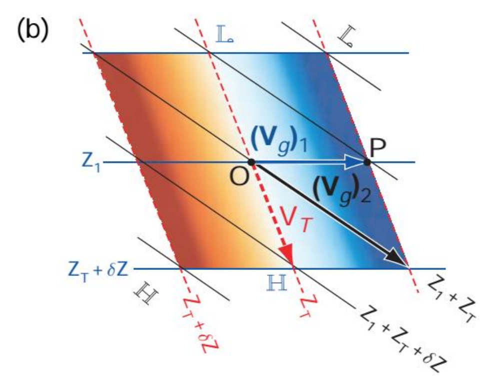



**$V_g$ decrease w/ height**




**Cold adv.**




**Warm adv.**

- ✓ L & H: Low & high geopotential heights
- ✓ Blue and orange colors: Cooler and warmer air



Wallace and Hobbs, 2006

# Primitive Equations for Large-scale Motions

Meteorological and climate models are based on these equations

**Hypsometric equation (hydrostatic)** $$\frac{\partial \Phi}{\partial p} = -\frac{RT}{p} \qquad d\Phi = g dz$$

**Horizontal equation of motion** $$\frac{d\boldsymbol{V}(u,v)}{dt} = -\nabla\Phi - f\boldsymbol{k} \times \boldsymbol{V} + \boldsymbol{F}$$

**Continuity equation** $$\frac{\partial \omega}{\partial p} = -\nabla \cdot \boldsymbol{V} \qquad \omega = \frac{dp}{dt}$$

**Thermodynamic energy equation** $$\frac{dT}{dt} = \frac{\kappa T}{p}\omega + \frac{J}{c_p} \qquad \kappa = R/c_p$$

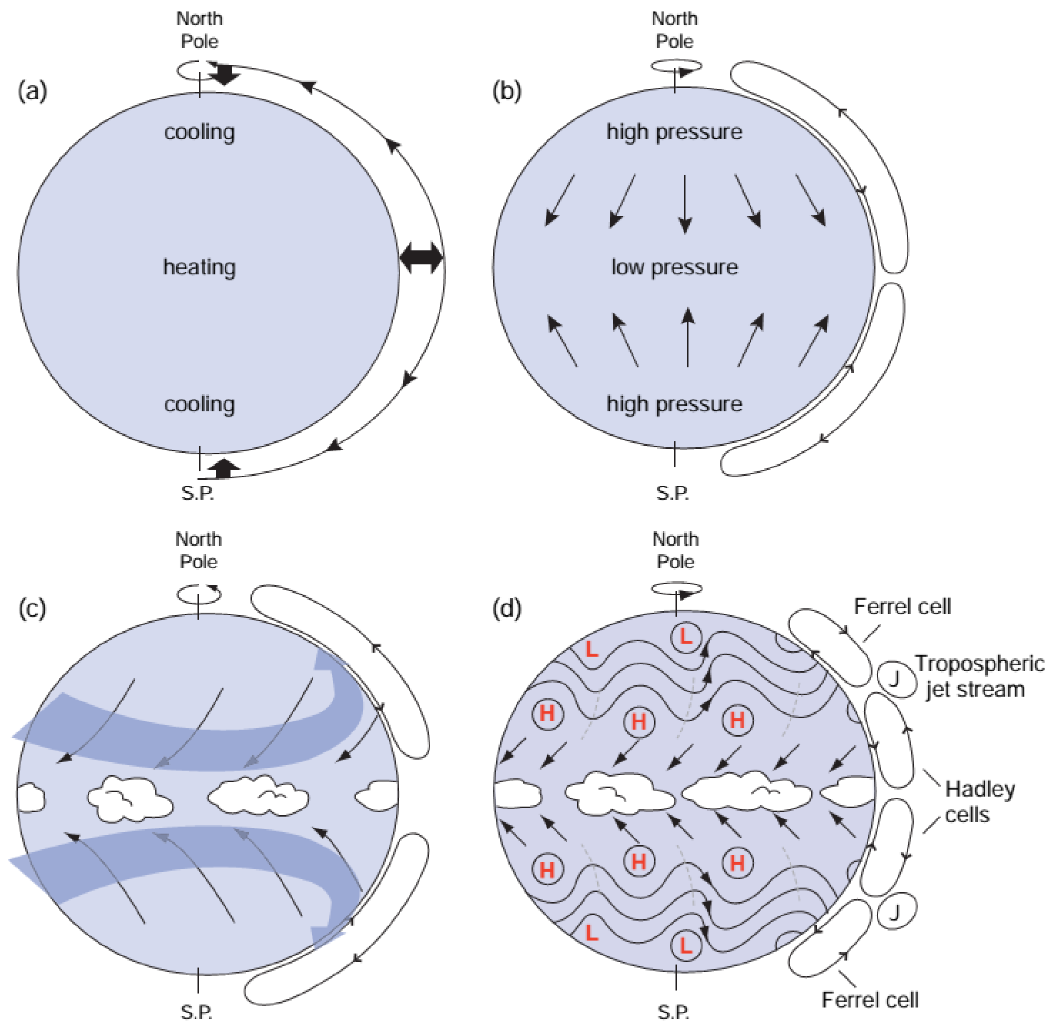
**Bottom boundary condition** $$\frac{\partial p_s}{\partial t} = -(\boldsymbol{V} \cdot \nabla p\,)_s - \left(w\frac{\partial p}{\partial z}\right)_s - \int_0^{p_s} (\nabla \cdot \boldsymbol{V}) dp$$

**Five unknowns** $$\mathbf{V}(u,v), \omega, \Phi, T$$

$J$ and $\boldsymbol{F}$ need to be parameterized

Wallace and Hobbs, 2006

# Primitive Equations and General Circulation

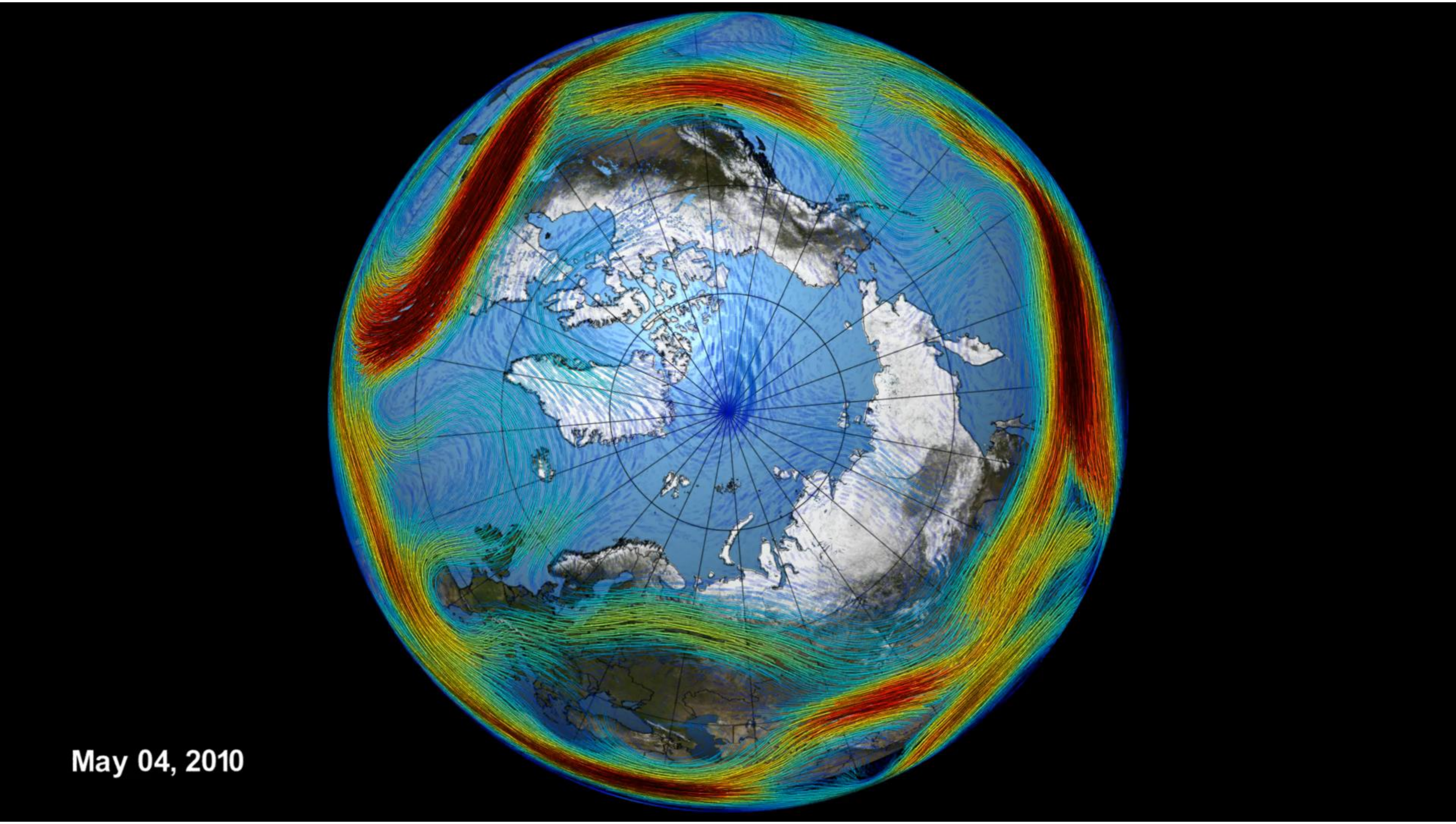



**Baroclinic instability and waves**



Wallace and Hobbs, 2006

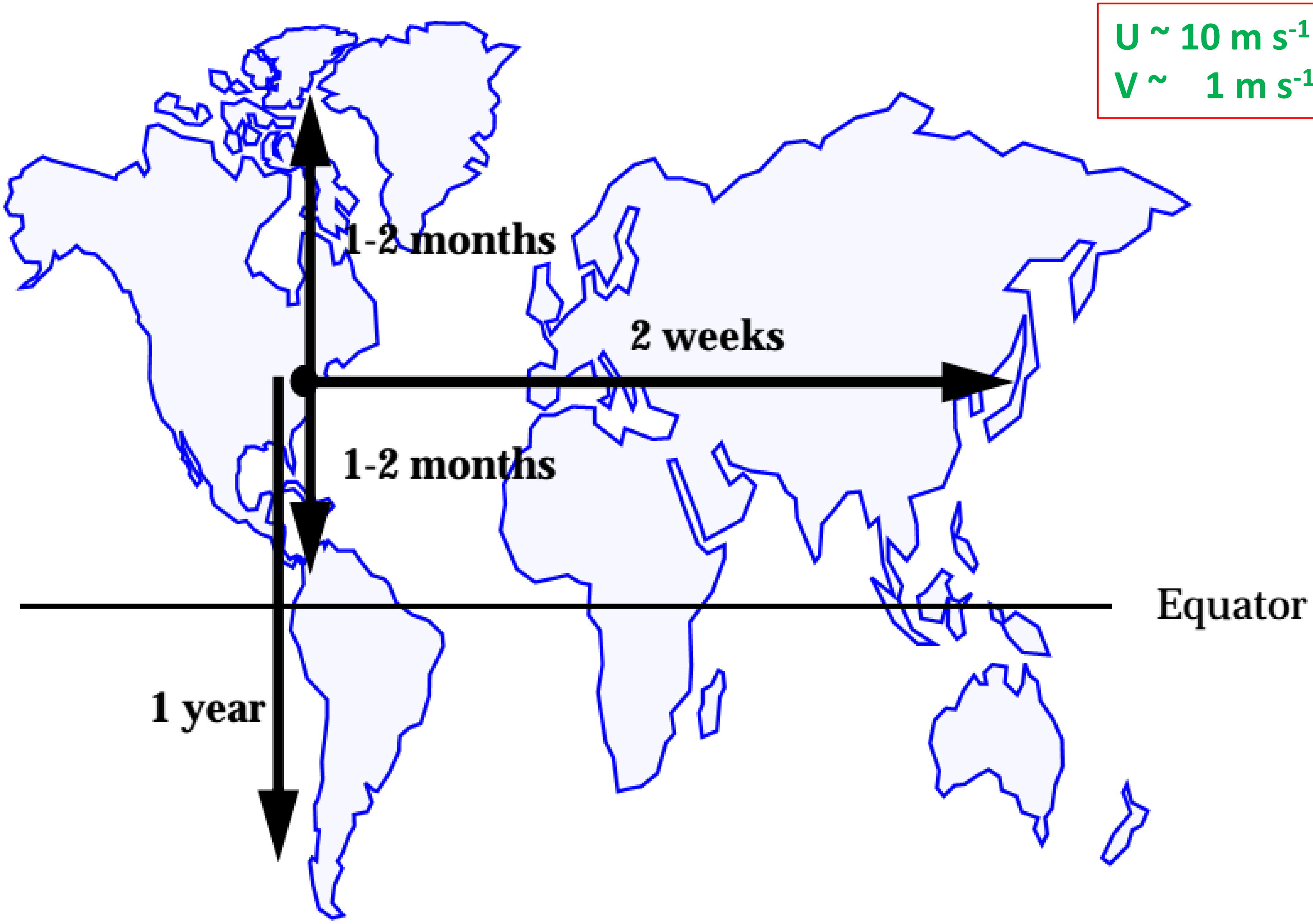
# Atmospheric Circulation




Source: https://svs.gsfc.nasa.gov/4148

# Characteristic Time Scales of Horizontal Transport

U ~ 10 m s$^{-1}$
V ~ 1 m s$^{-1}$




Jacob, 1999

# Vertical Transport Processes

**Large-scale vertical motion: w ~ 1 cm s$^{-1}$**

- **Time from surface to tropopause (10 km) ~ 11 days**

**Molecular Diffusion** ***Einstein's equation; Fick's Law***

$$F = -n_a D \frac{\partial C}{\partial z} \qquad \Delta t = \frac{(\Delta x)^2}{2D}$$

$$D = D_0 \frac{p_0}{p} \qquad D_0 = 0.2 \text{ cm}^2 \text{ s}^{-1}$$

F = diffusion flux
$n_a$ = number density of air
C = mixing ratio

- **At surface w/ $D_0$, it takes about 1 month to move 100 m !**
- **Only important above 100 km (0.01 hPa)**

Jacob, 1999

# Vertical Transport Processes

**Turbulence**

$$\bar{F} = -n_a K_z \frac{\partial \bar{C}}{\partial z} \qquad \Delta t = \frac{(\Delta z)^2}{2K_z}$$

| |
|---|
| $K_z$ = 2 x $10^5$ $cm^2$ $s^{-1}$ Trop. Mean |
| $10^2$ – $10^5$ $cm^2$ $s^{-1}$ if stable |
| $10^4$ – $10^6$ $cm^2$ $s^{-1}$ if neutral |
| $10^5$ – $10^7$ $cm^2$ $s^{-1}$ if unstable |

- **Static Instability ($\theta$, $\theta_e$ distribution)**
- **Mechanical Instability (wind shear)**

➢ **Boundary layer mixing: w ~ 1 m $s^{-1}$**
➢ **Convection (dry & moist): w ~ 1 m $s^{-1}$**

# Static Instability

## For Unsaturated Air

Stable: $\Gamma < \Gamma_d$ (i.e., θ increases with height)
Neutral: $\Gamma = \Gamma_d$ (i.e., θ increases with height)
Unstable: $\Gamma > \Gamma_d$ (i.e., θ increases with height)

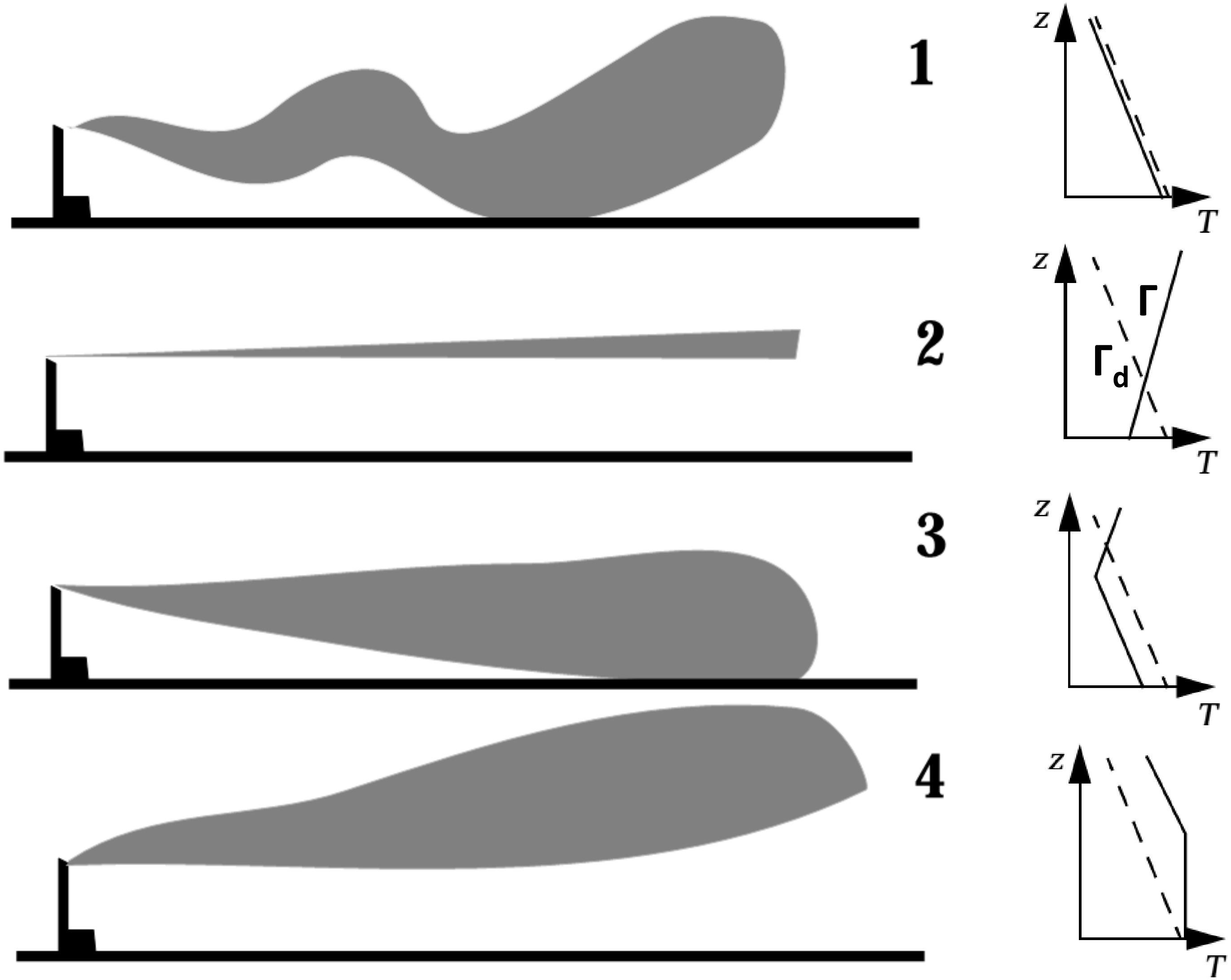
$$\Gamma_d = \frac{g}{c_p} \qquad \theta = T\left(\frac{p_0}{p}\right)^{R/c_p} \qquad \frac{1}{\theta}\frac{\partial \theta}{\partial z} = \frac{1}{T}(\Gamma_d - \Gamma)$$

## For saturated air

Stable: $\Gamma < \Gamma_s$
Neutral: $\Gamma = \Gamma_s$
Unstable: $\Gamma > \Gamma_s$

$$\Gamma_s \approx \frac{\Gamma_d}{1 + \frac{L_v}{c_p}\left(\frac{\partial w_s}{\partial T}\right)_p}$$

# Mixing in the Lower Troposphere

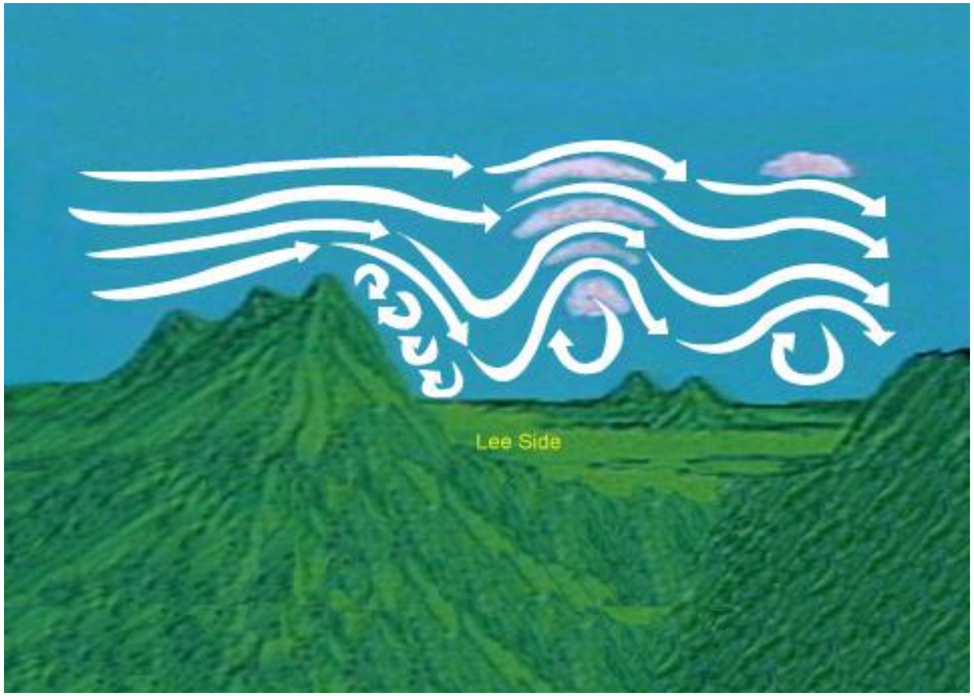



Jacob, 1999

# Mechanical Instability

Causes:

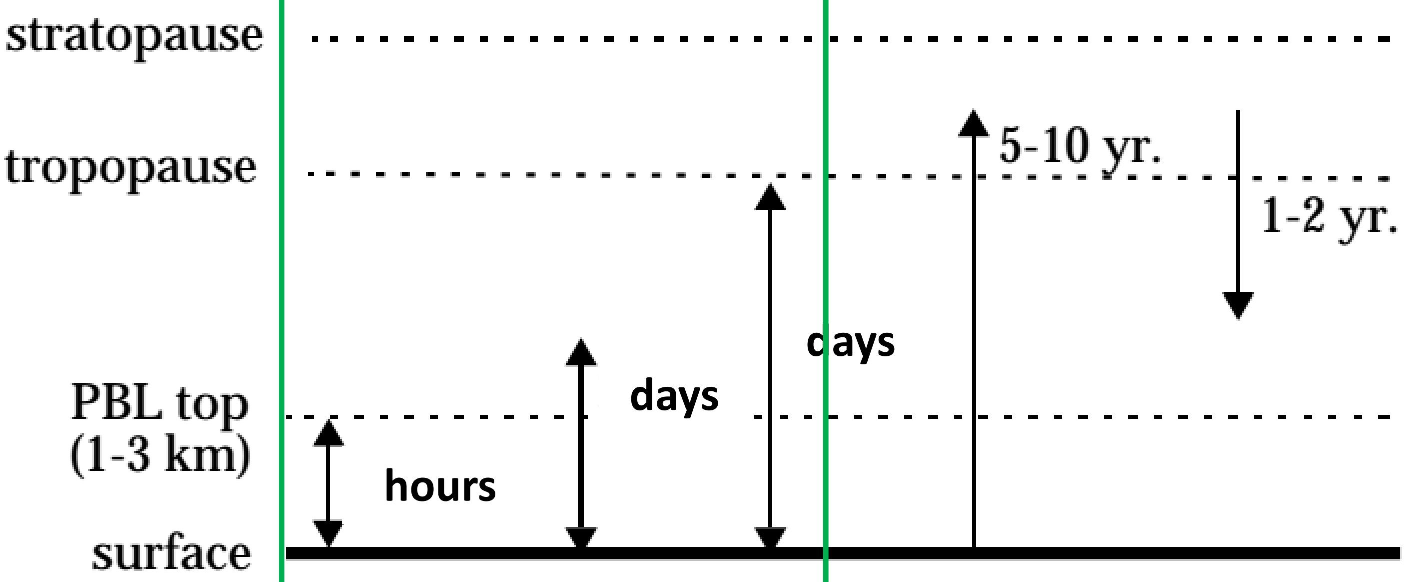
- Friction (e.g., by Earth surface)
- Barrier (e.g., by buildings, mountains)
- Wind shear (i.e., Kelvin–Helmholtz instability)




# Characteristic Time Scales of Vertical Transport

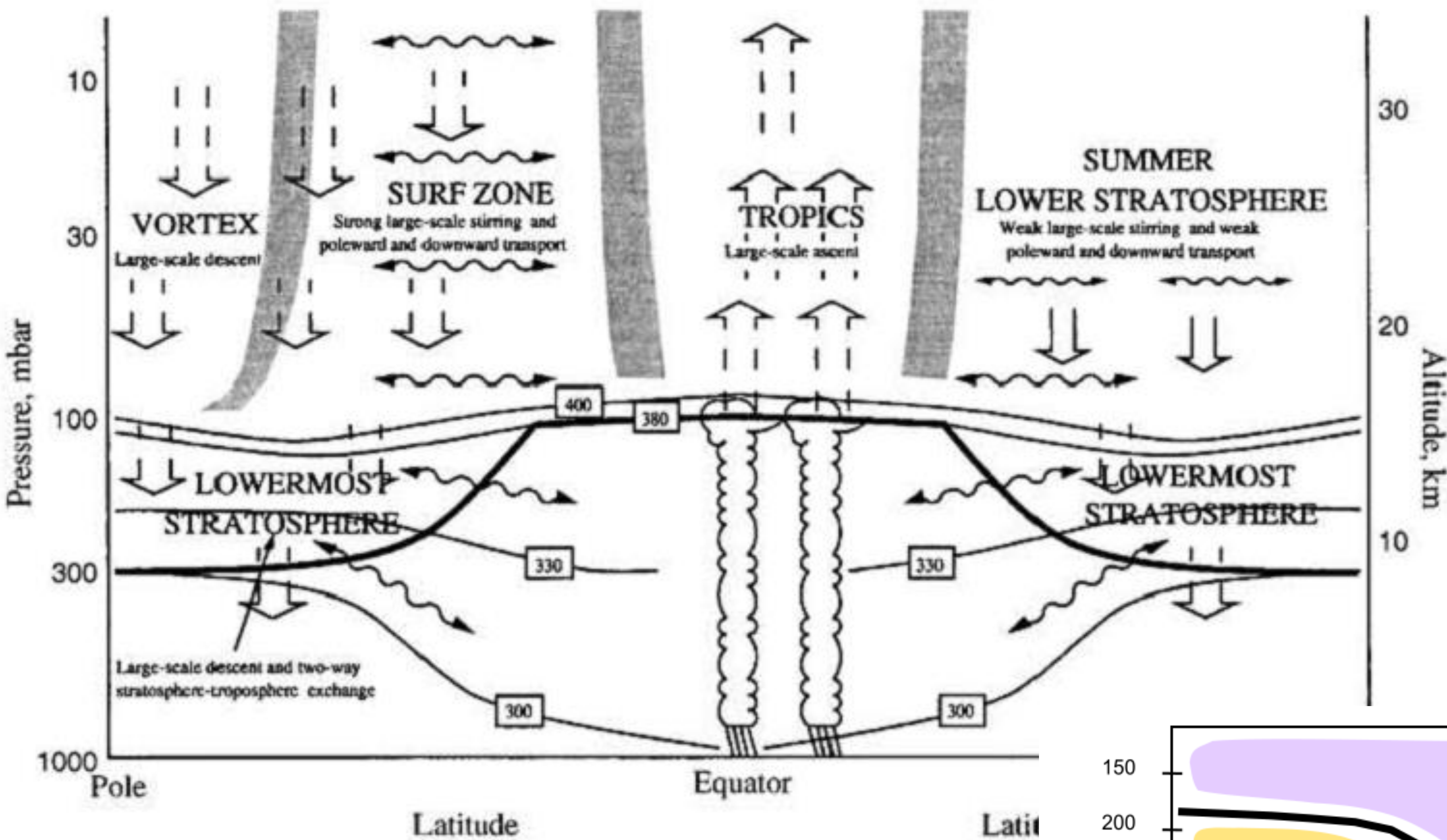
Estimate based on $K_Z$
Rough estimate
No "fast" processes

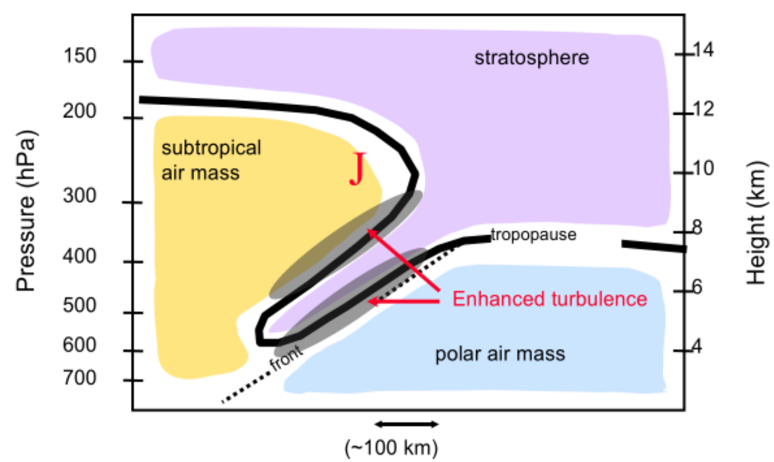
$K_Z = 2 \times 10^5\ cm^2\ s^{-1}$ Trop. Mean
$10^2 - 10^5\ cm^2\ s^{-1}$ if stable
$10^4 - 10^6\ cm^2\ s^{-1}$ if neutral
$10^5 - 10^7\ cm^2\ s^{-1}$ if unstable




Modified from Jacob (1999)

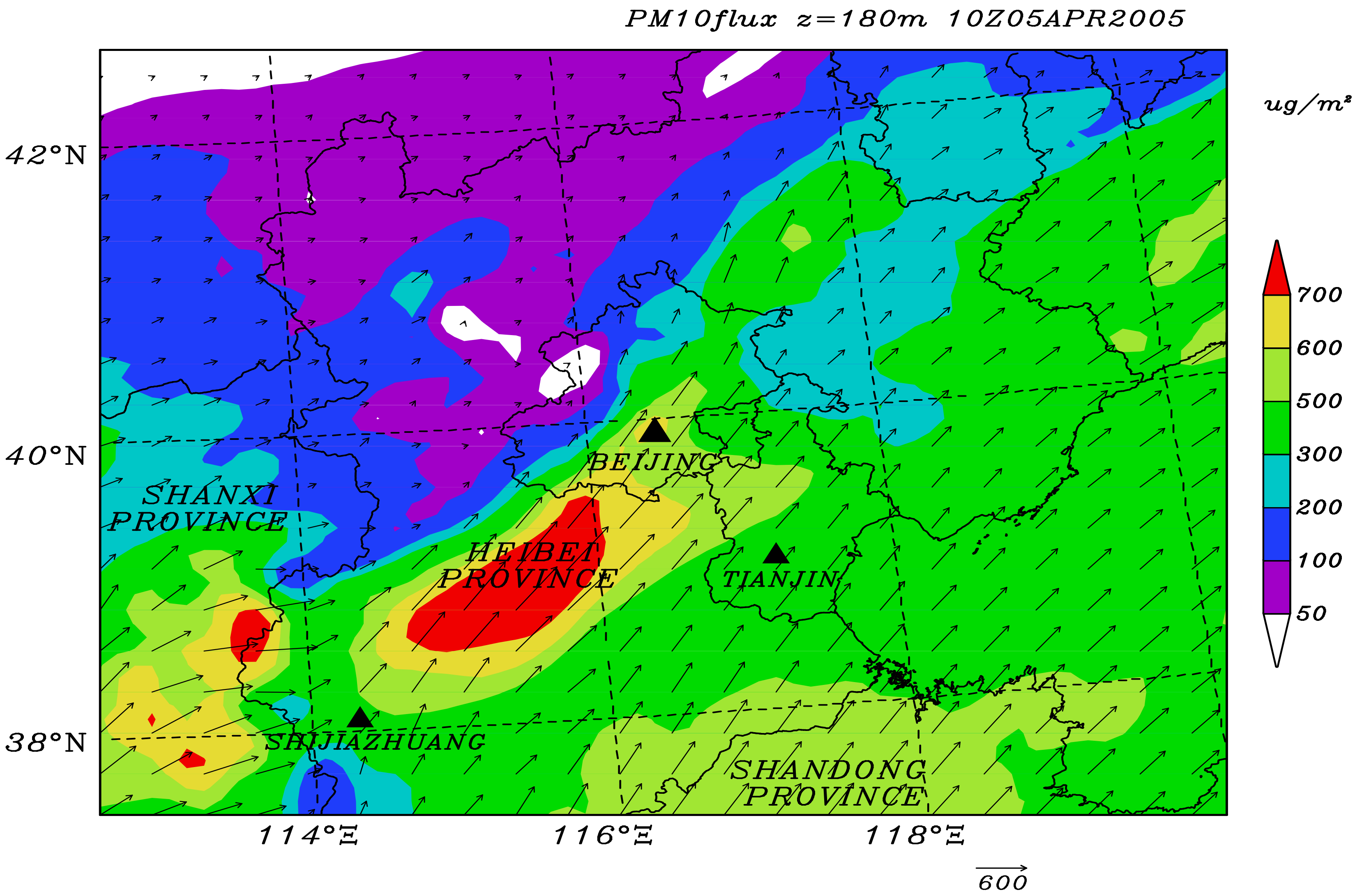
# Stratosphere-Troposphere Exchange



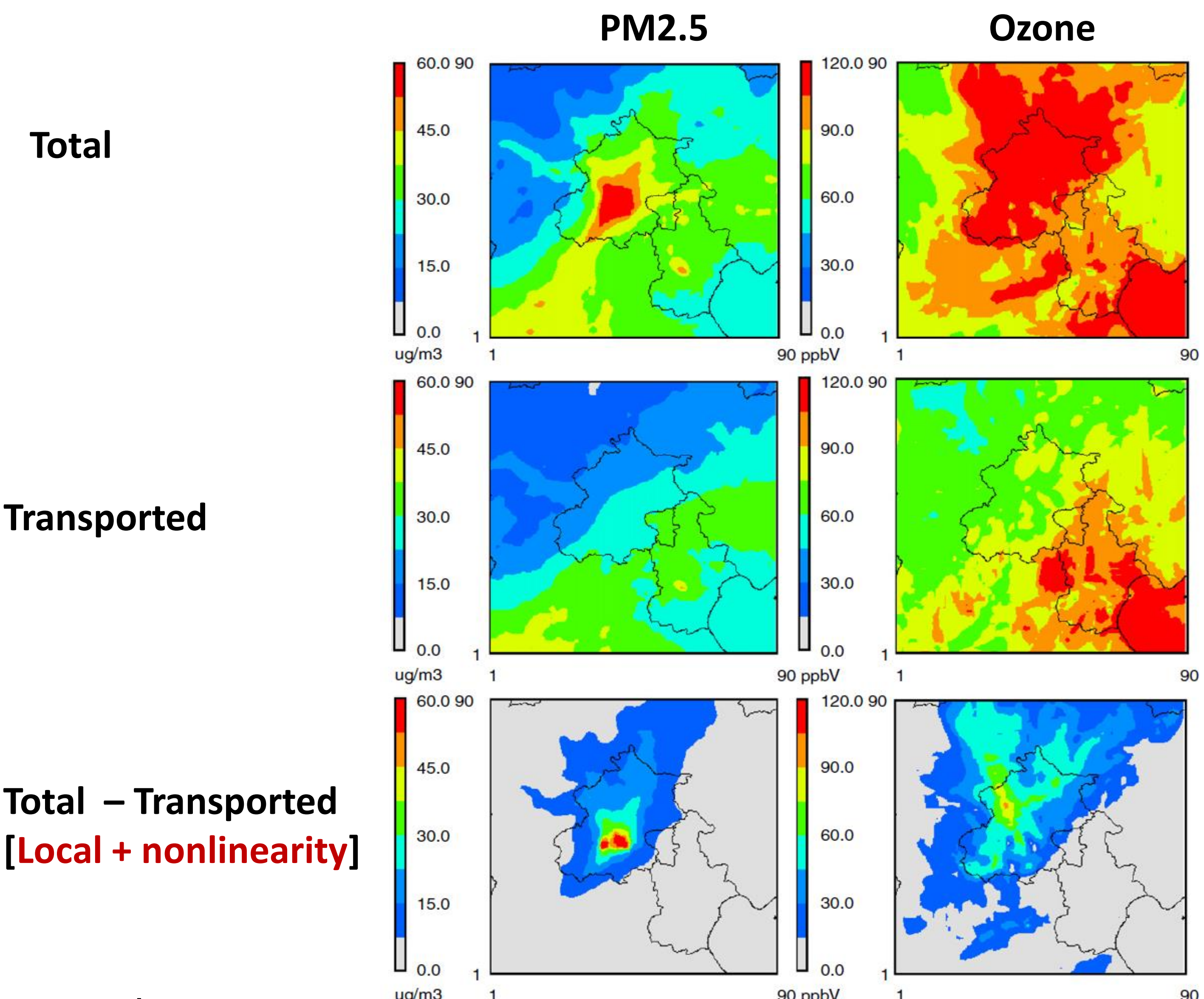

Seinfeld and Pandis, 2006

# Regional Transport and Transformation Affecting Beijing




An et al., ACP 2007

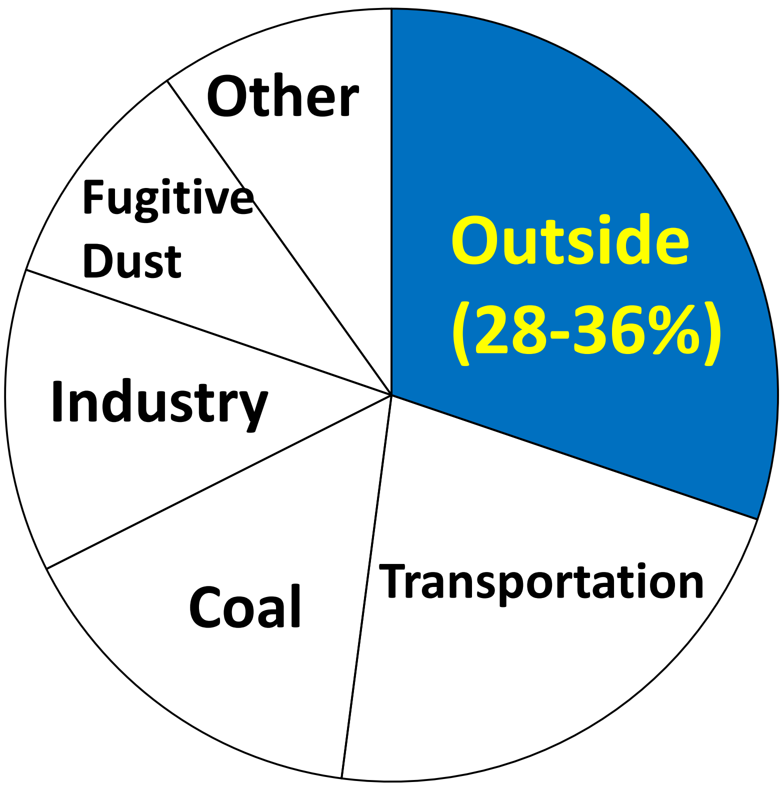
# Regional Transport and Transformation Affecting Beijing



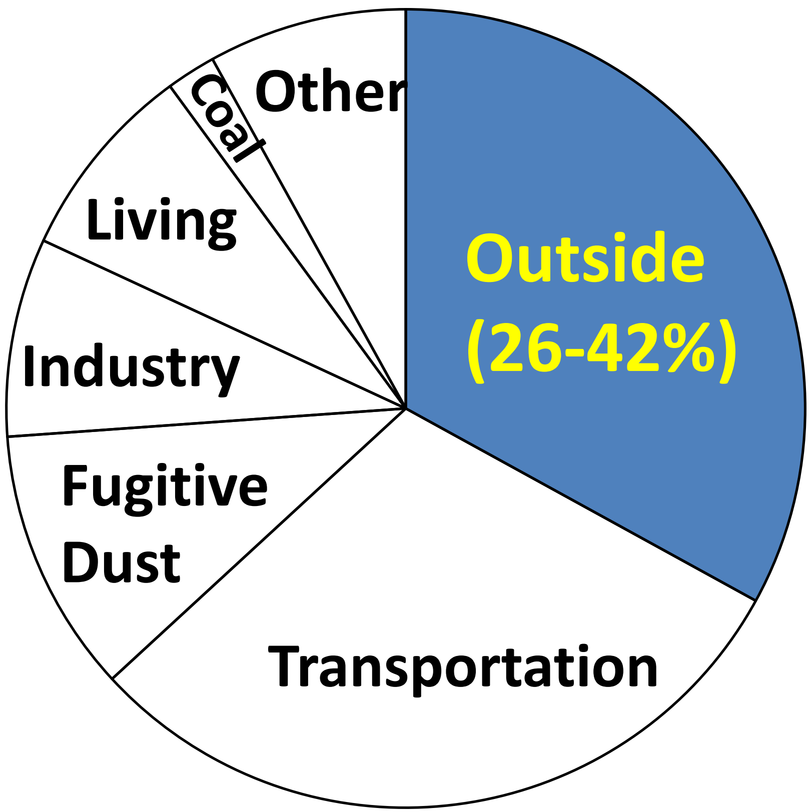

An et al., ACP 2007

# Atmospheric $PM_{2.5}$ Transport Affects Beijing

**Sources of Beijing's $PM_{2.5}$**
**(北京市环保局，2014)**



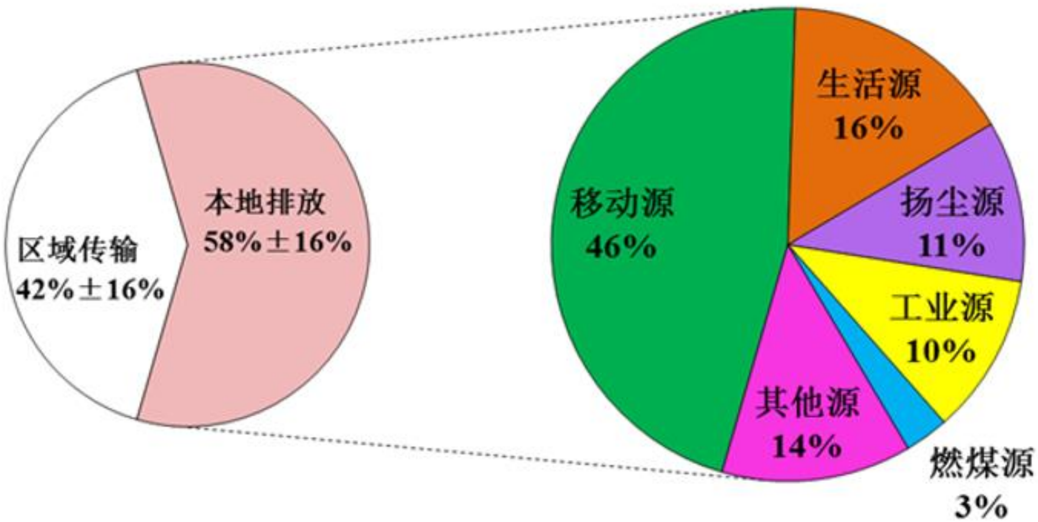

**Sources of Beijing's $PM_{2.5}$**
**(北京市环保局，2018)**



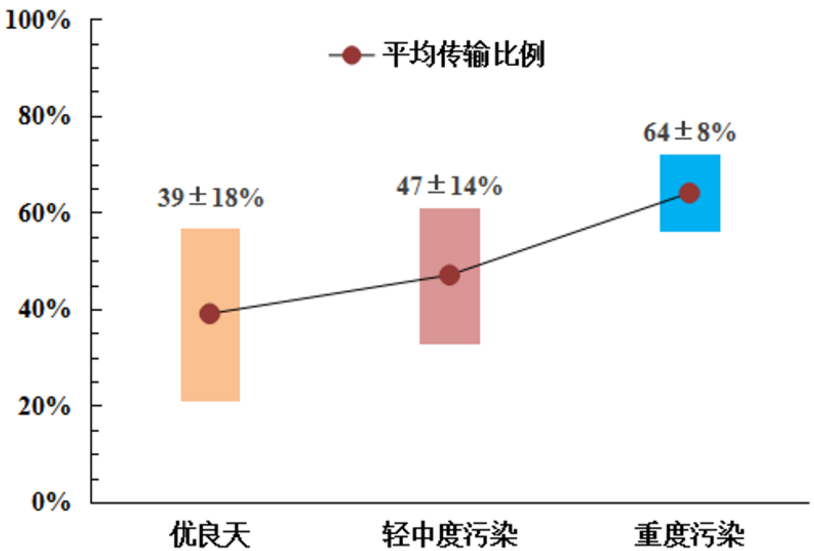

区域输送贡献：
- 年平均：26%-42%
- 中度污染（115-150 μg/m³）：34%-50%
- 重污染日（＞150 μg/m³）：55%-75%

# Increasing Role of Atmospheric Transport to Beijing's $PM_{2.5}$

**Sources of Beijing's $PM_{2.5}$ (北京市生态环境局，2021)**




- ✓ 32±4% in 2014 (第一轮)
- ✓ 34±8% in 2018 (第一轮)
- ✓ 42±16% in 2021 (第一轮)

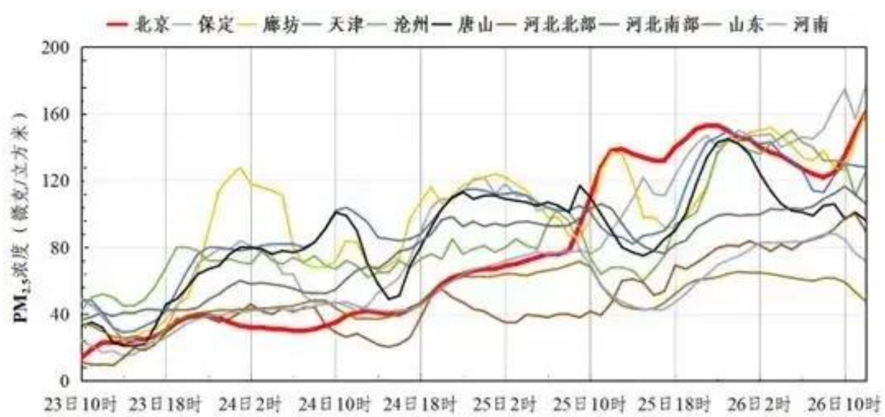


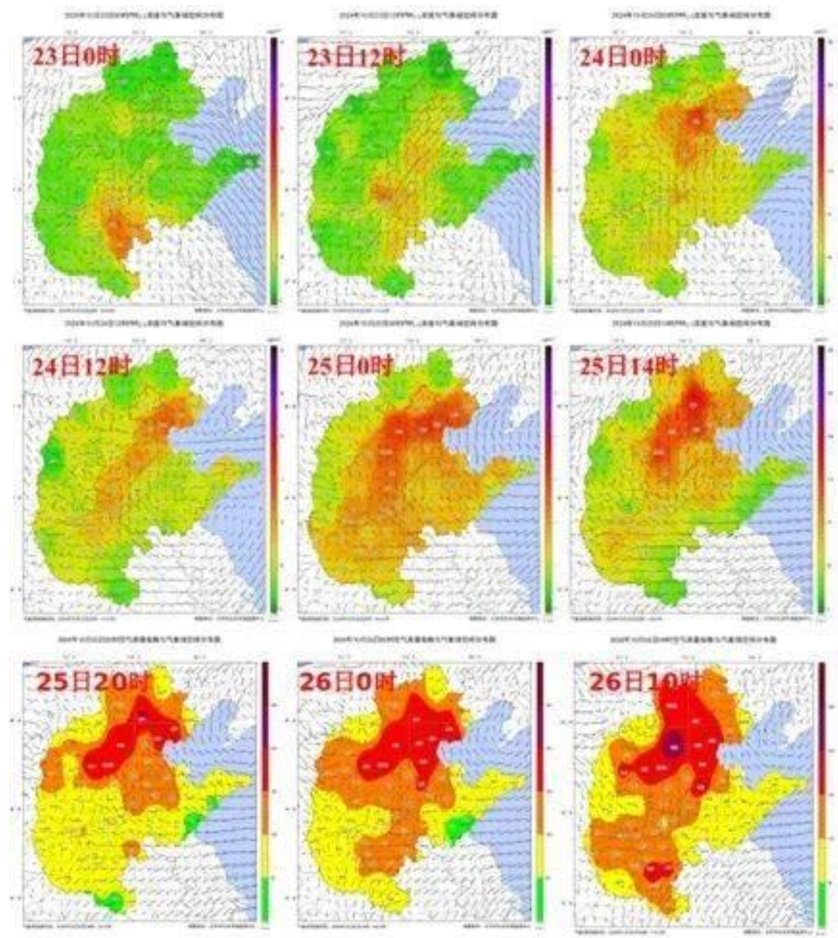
# Severe Regional PM Pollution Transport to Beijing

## Back-trajectory analysis of BJ's PM on 2014/10/10


# Severe Regional PM Pollution Transport to Beijing

## Transport-driven growth of BJ's PM on 2024/10/26

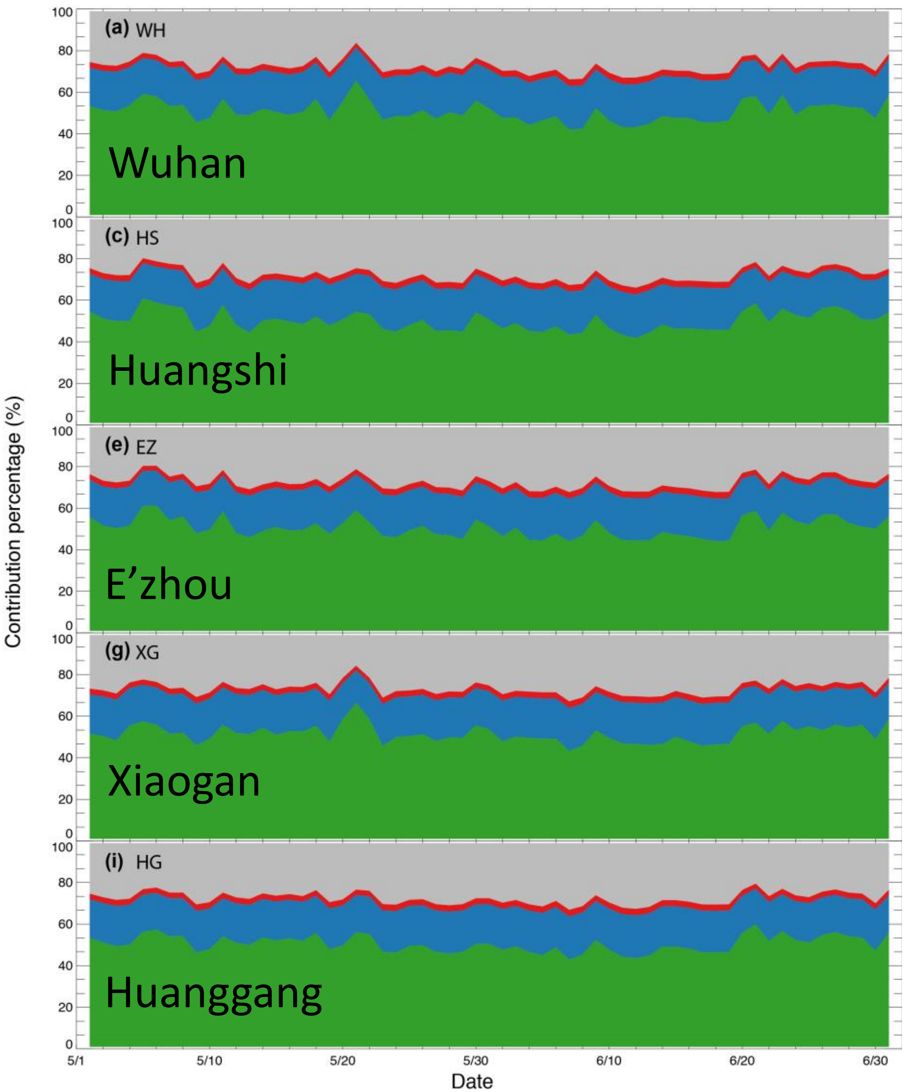



10月24日13时-26日14时，区域传输贡献77%，其中东南通道35%，西南通道18%，东南通道占据主导。

https://www.sohu.com/a/820625924_204474

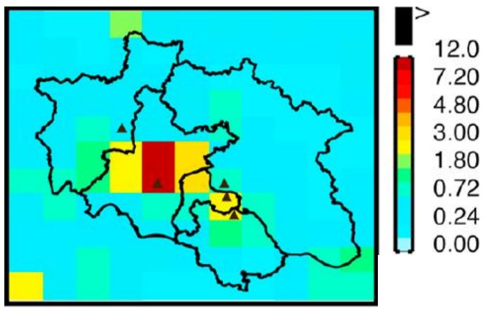
# Key Roles of Local Production and Atmospheric Transport for Ozone Pollution over Central China



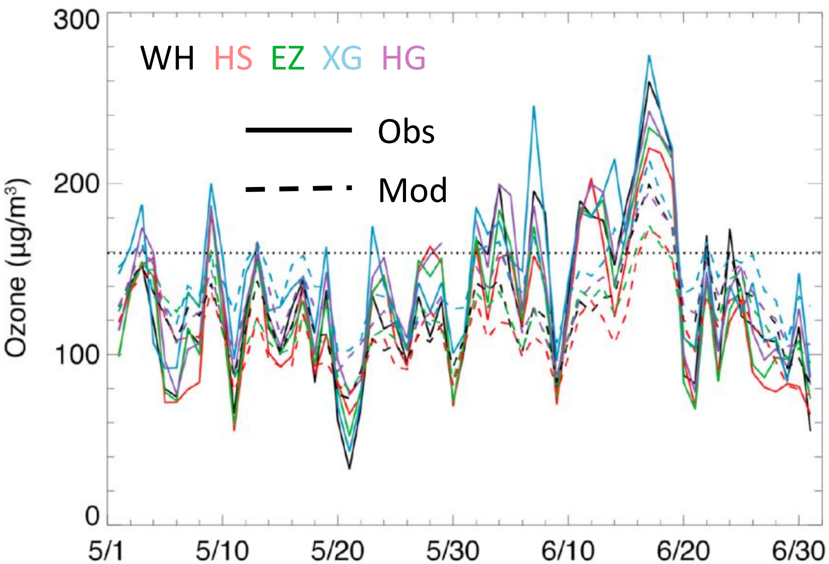

Yan YY et al., 2021, AE

Contributors:
- ✓ Local emis
- ✓ Cross-city
- ✓ Asian BG
- ✓ Global BG

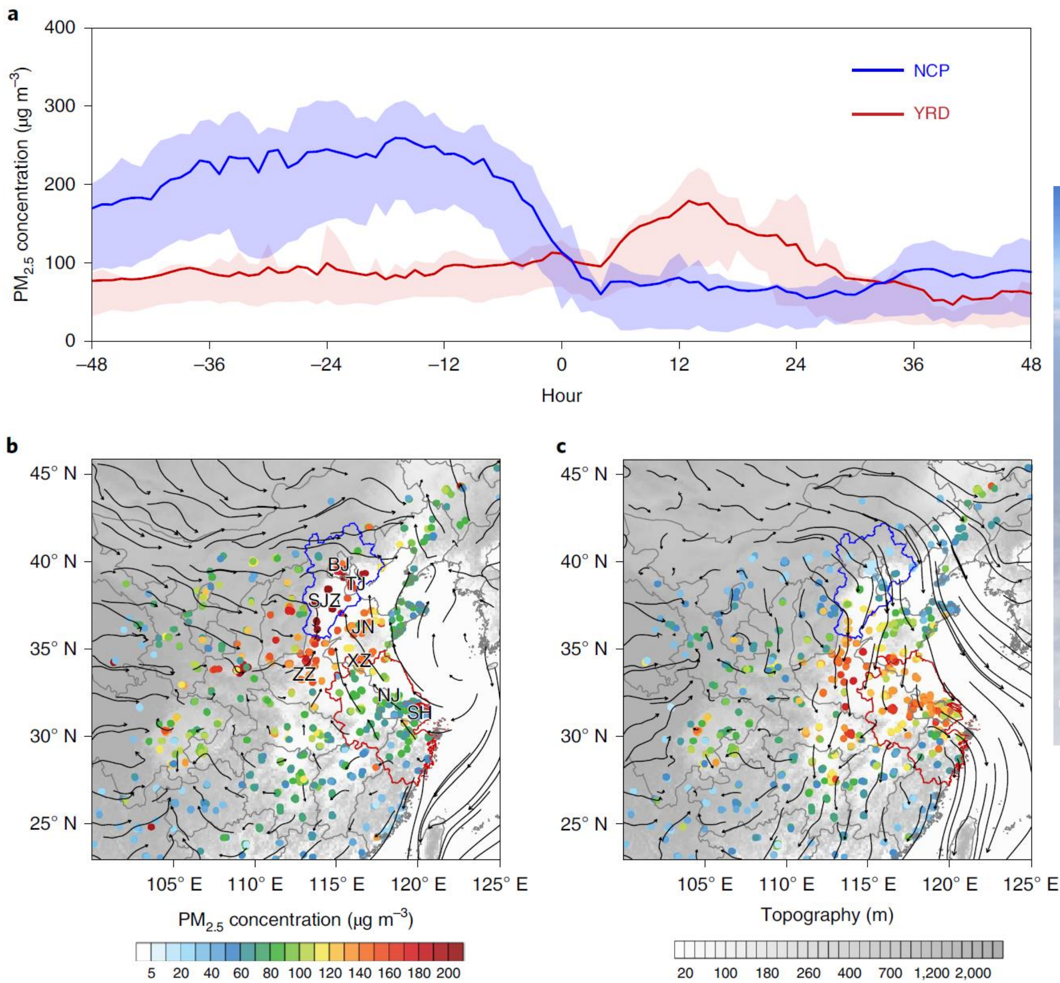
Anthro NOx emis ($g/m^2$)

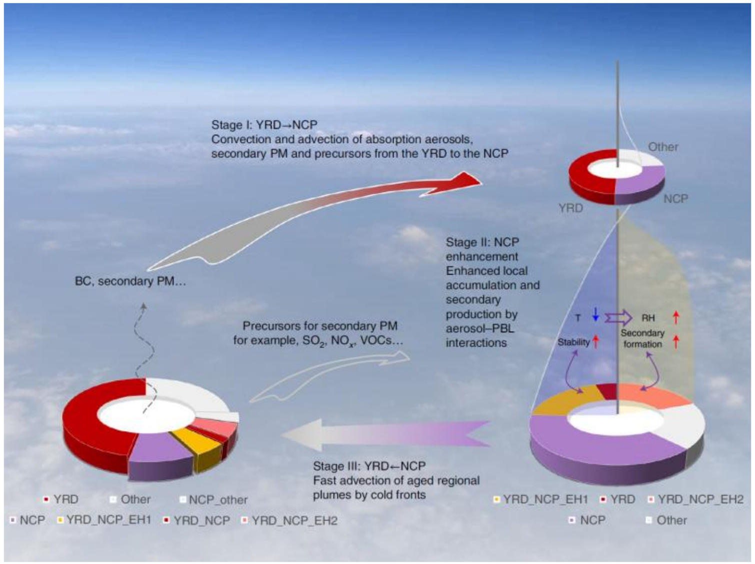



MDA8 ozone in May-June 2018



# Two-way Transport of $PM_{2.5}$ Between NCP and YRD

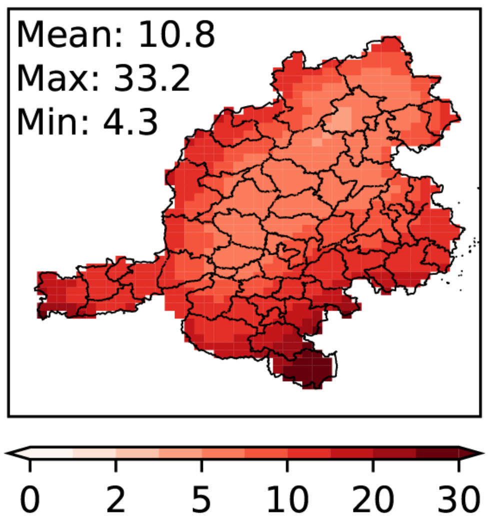


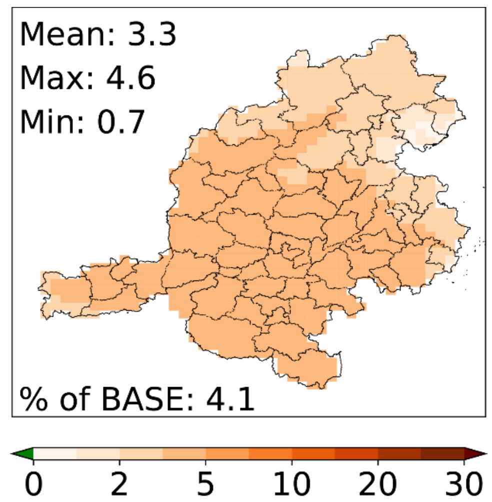

Huang et al., 2020, Nature Geoscience

# Large Impact of Transport to Summer Ozone over NCP + FW

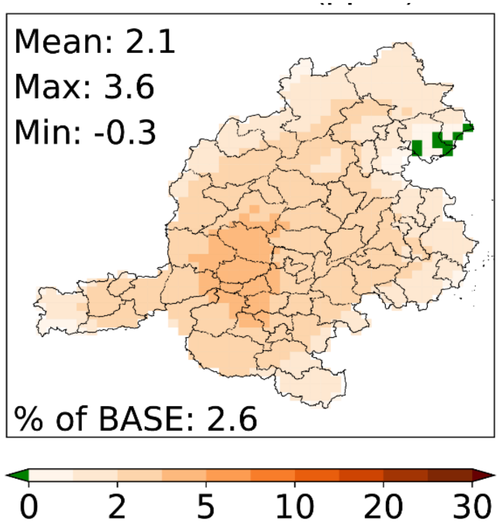
% contribution of transport to total JJA MDA8 O3 (%)



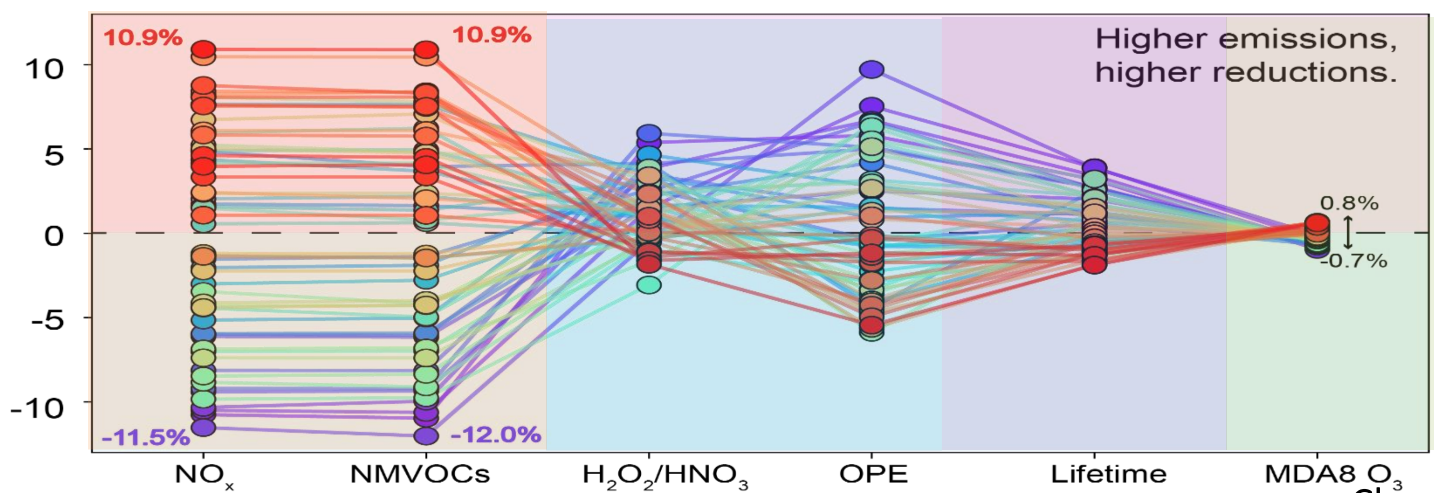

$\Delta O_3$ by 20% emis cut in China




$\Delta O_3$ by 20% emis cut in NCP+FW



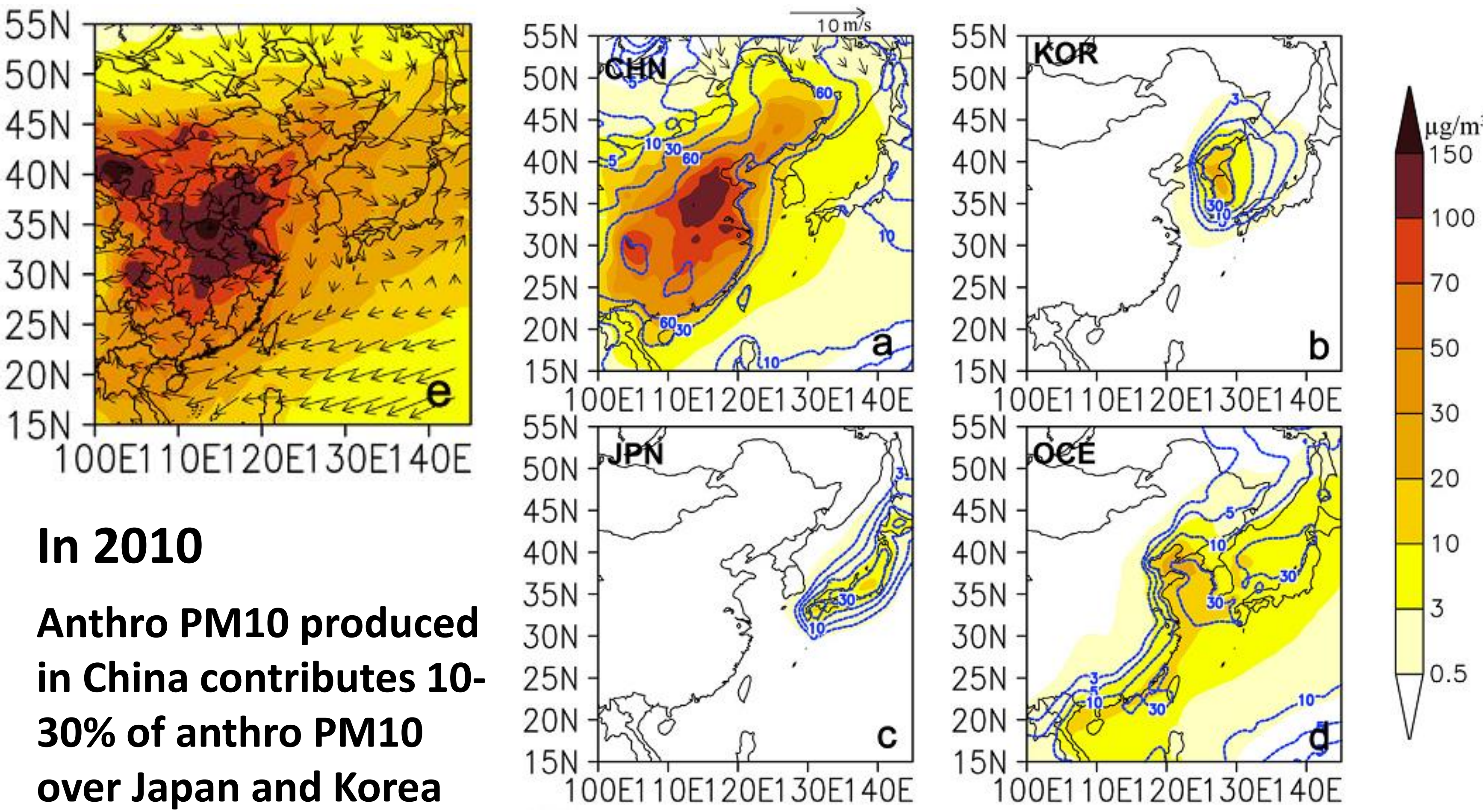

Relative to uniform 20% emis cut






Chen et al., ERL, 2025

# PM Transport Between Asian Countries



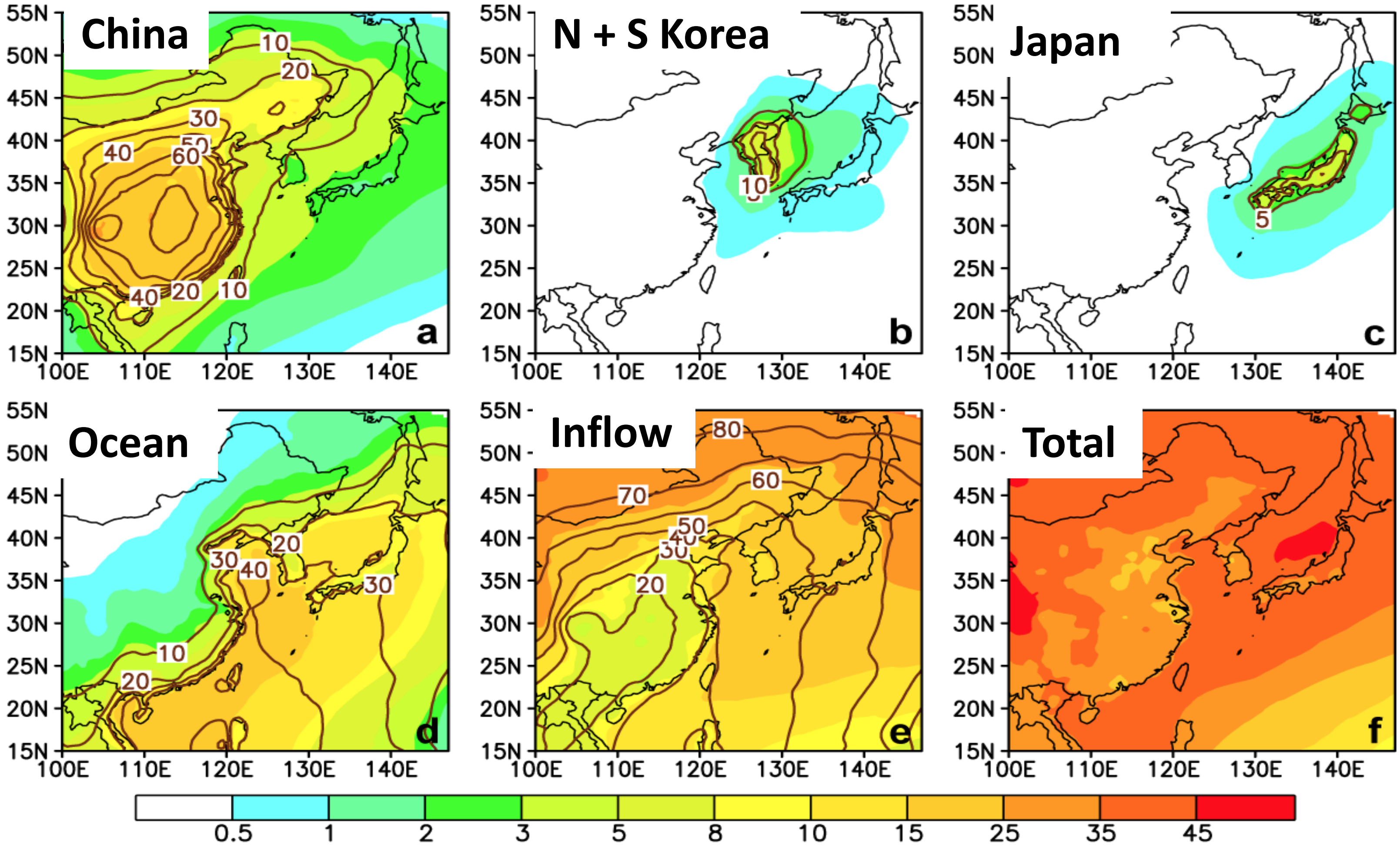

## In 2010

**Anthro PM10 produced in China contributes 10-30% of anthro PM10 over Japan and Korea**

**NAQPMS + tagging**

Li et al., 2014, AE

# Ozone Transport into and within East Asia



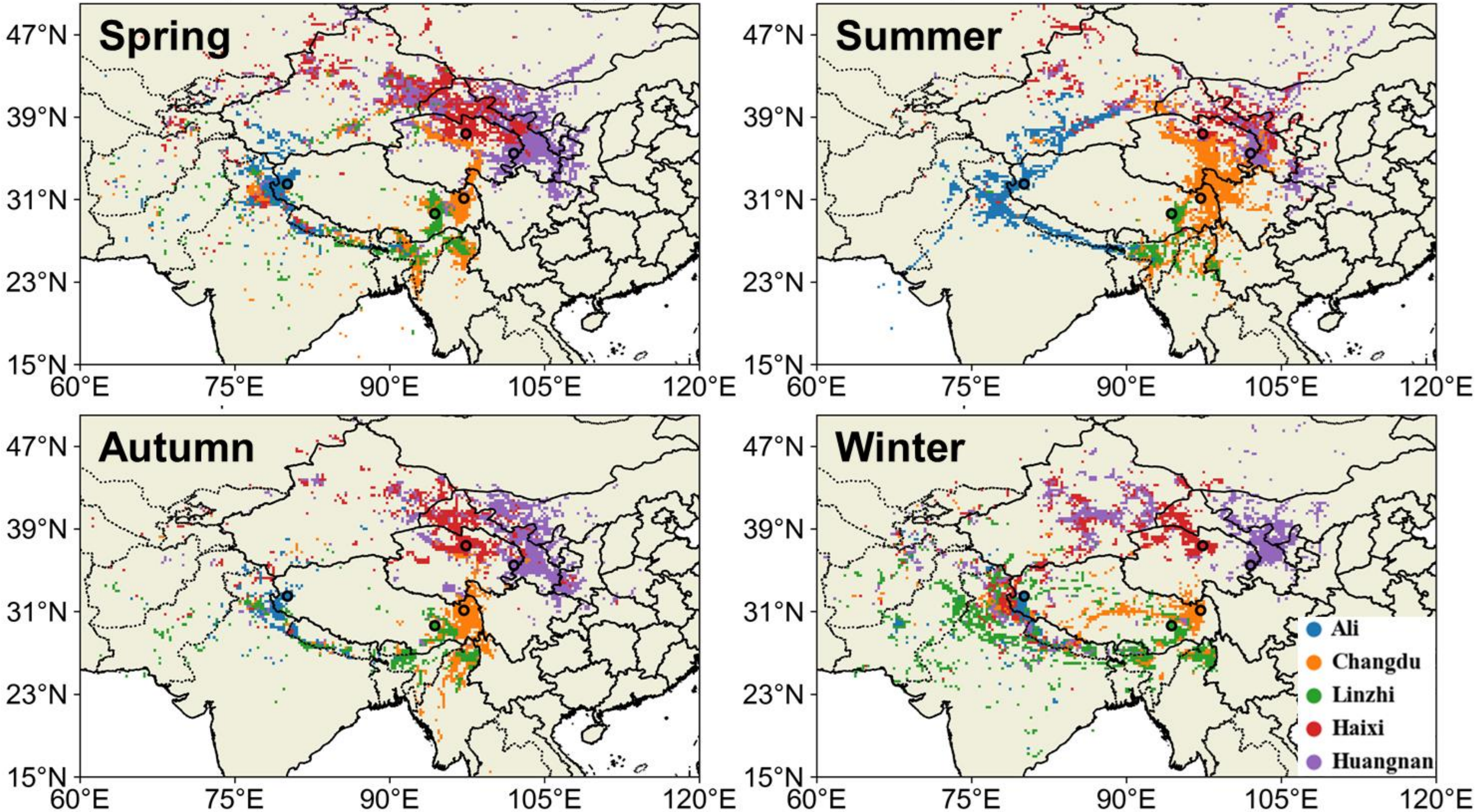

Color: ppbv; Contour: %

**2010 mean**

Li et al., 2016, AR

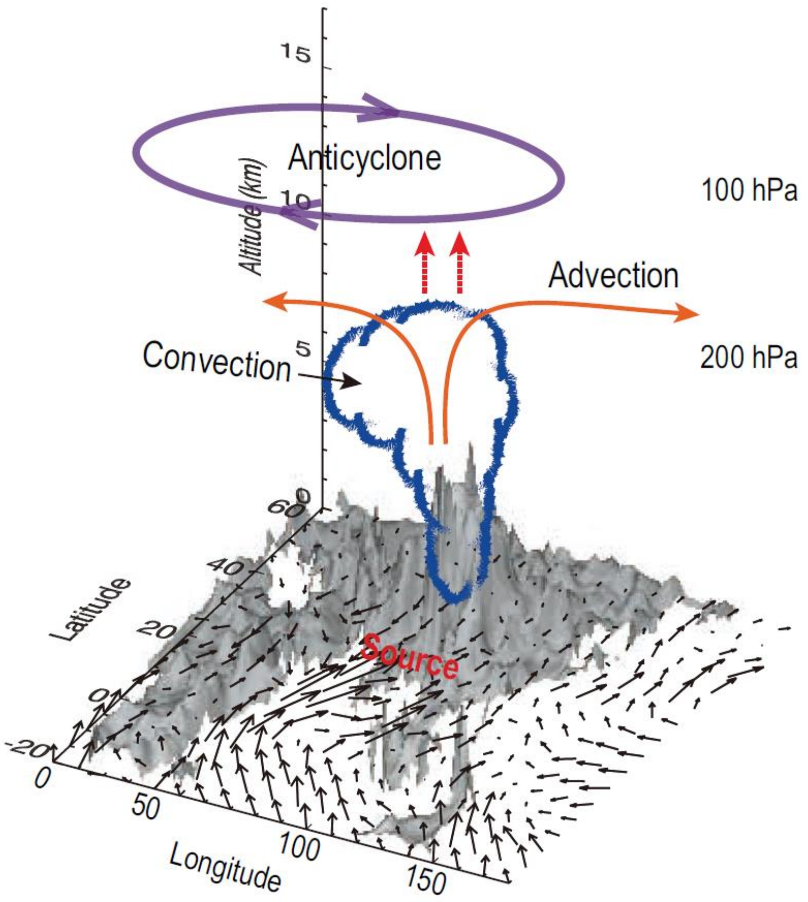
# Transboundary Sources of Ozone over Tibetan Plateau

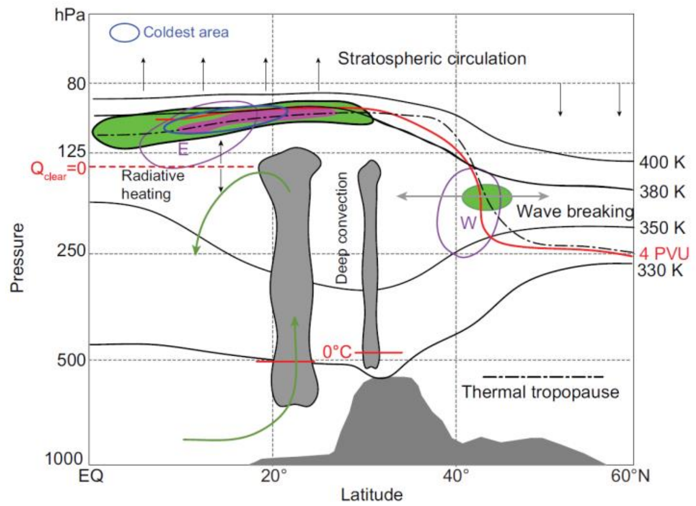
**Ozone sources in each season based on WPSCF**

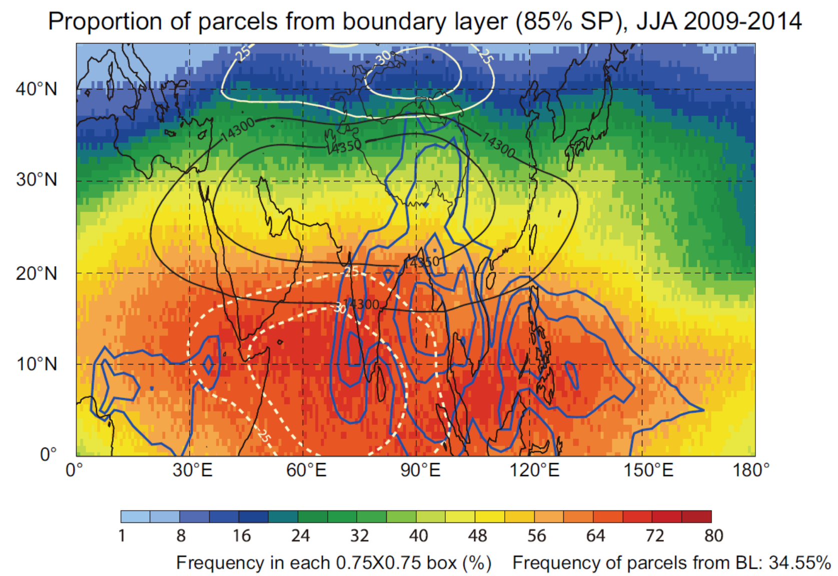



Xu et al., under review

# Transport into Asian Tropical UTLS




Fraction of parcels from BL (85% SP), JJA 2009-2014

# Pathways and Time of Transpacific Transport

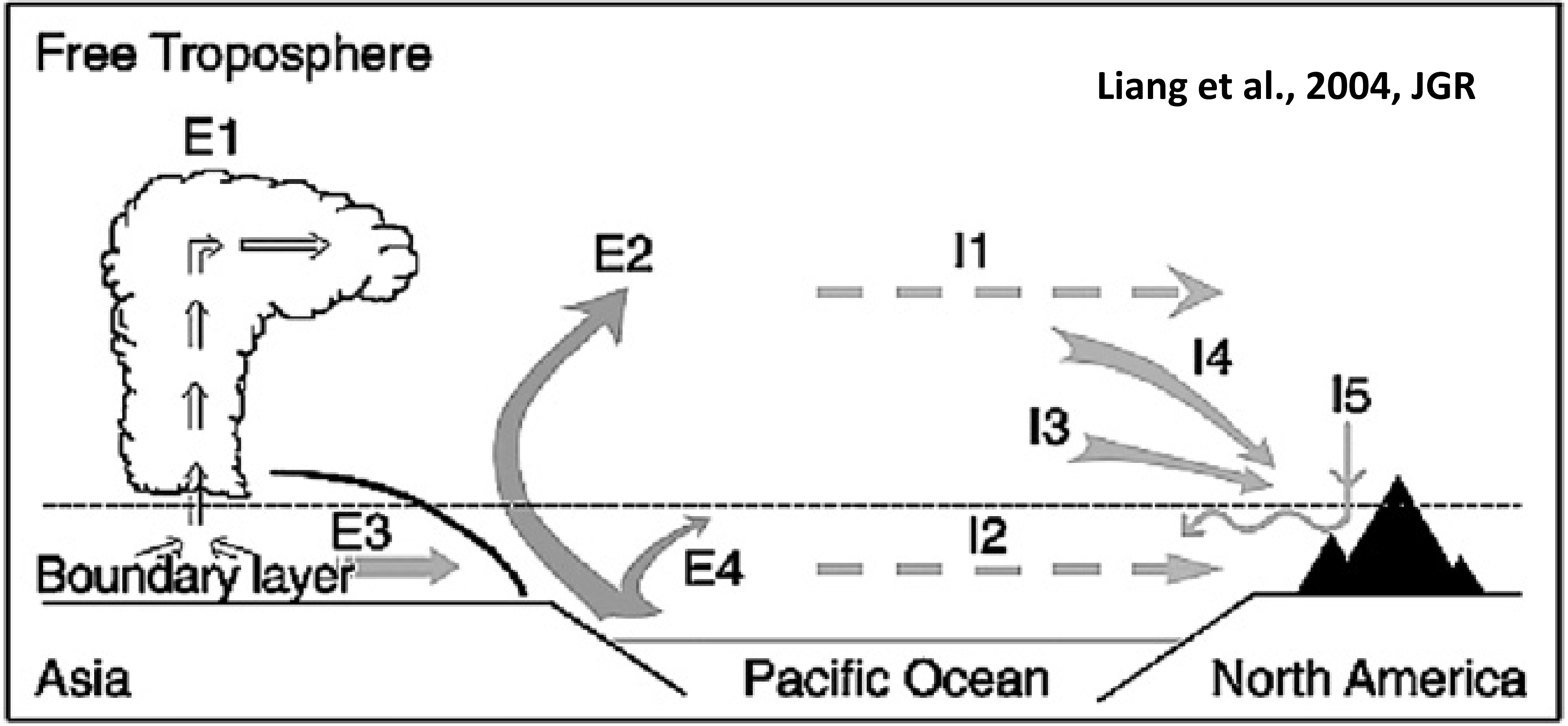
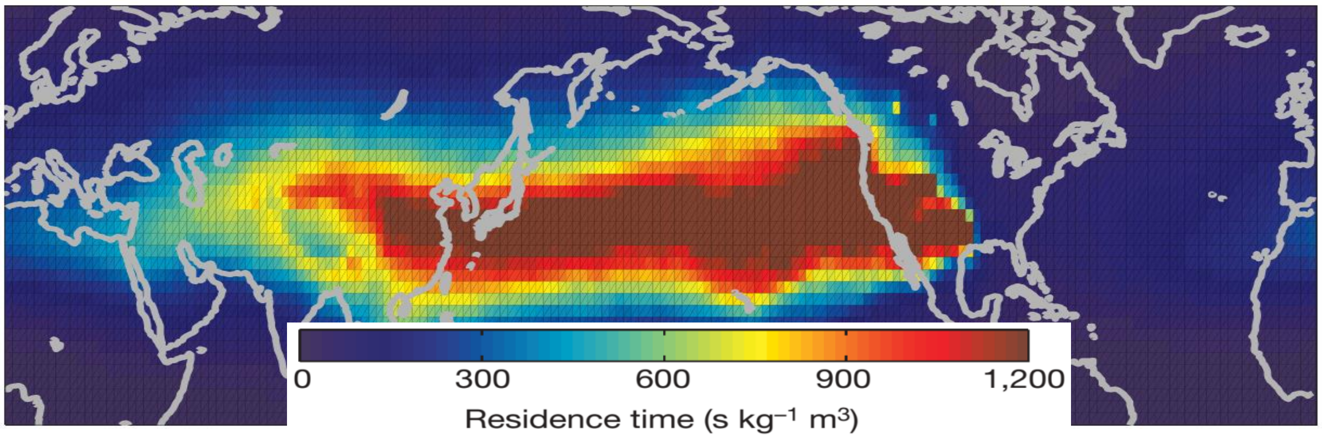


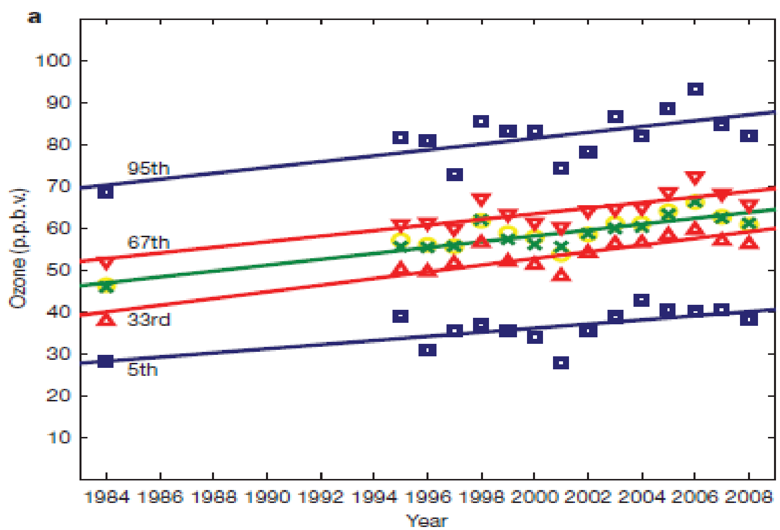

**Table 1.** 11-Year Average Inter-Continental Transport Times for Two Sets of Tracers in April (Unit: Weeks)

| Tracer Lifetime | EA->CPO | EU->Beijing | NA->Paris |
|---|---|---|---|
| 1–2 weeks | 2.5 | 2.0 | 2.0 |
| 4–8 weeks | 5.1 | 4.1 | 4.5 |

# Atmospheric $O_3$ Transport from China to U.S.

## Cooper et al., 2010, Nature



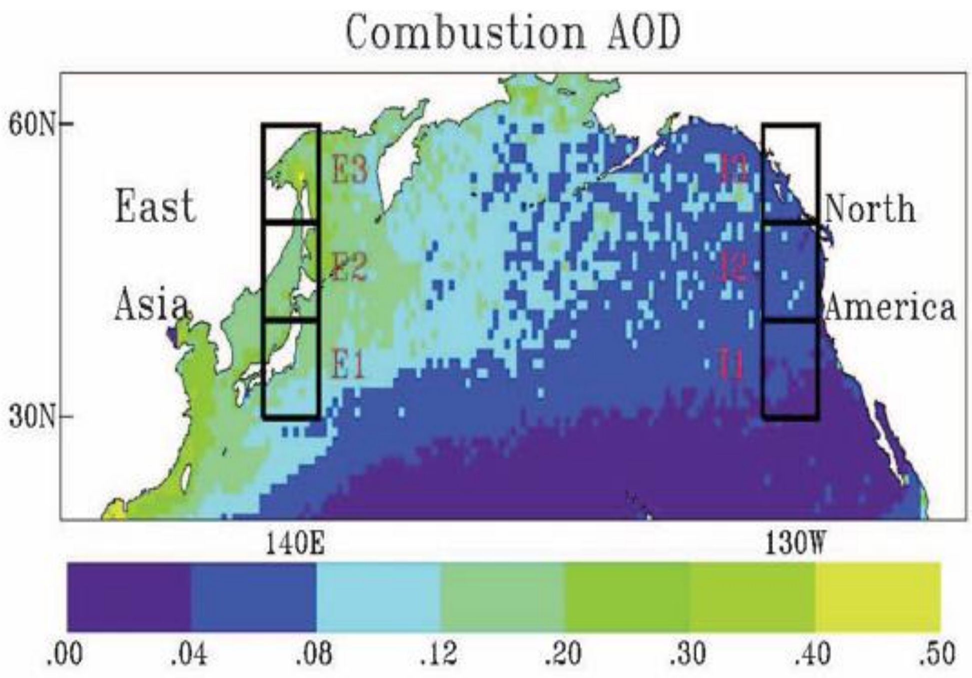

| Percentile | 1995–2008 increase (p.p.b.v. $yr^{-1}$) | *P* |
|---|---|---|
| 95th | 0.56 ± 0.71 | 0.11 |
| 67th | 0.62 ± 0.39 | 0.00 |
| 50th | 0.63 ± 0.34 | 0.00 |
| 33rd | 0.64 ± 0.35 | 0.00 |
| 5th | 0.48 ± 0.53 | 0.07 |

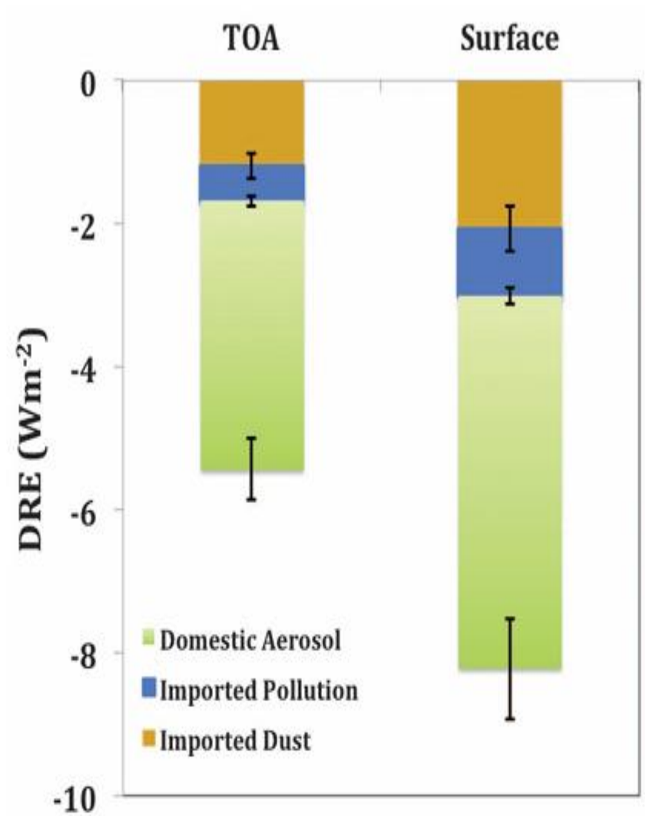
| Percentile | 1984–2008 increase (p.p.b.v. $yr^{-1}$) | *P* |
|---|---|---|
| 95th | 0.70 ± 0.45 | 0.01 |
| 67th | 0.67 ± 0.25 | 0.00 |
| 50th | 0.70 ± 0.22 | 0.00 |
| 33rd | 0.80 ± 0.24 | 0.00 |
| 5th | 0.49 ± 0.34 | 0.01 |

# Asian PM Transport Affects North America

**Yu et al., 2012, Science**

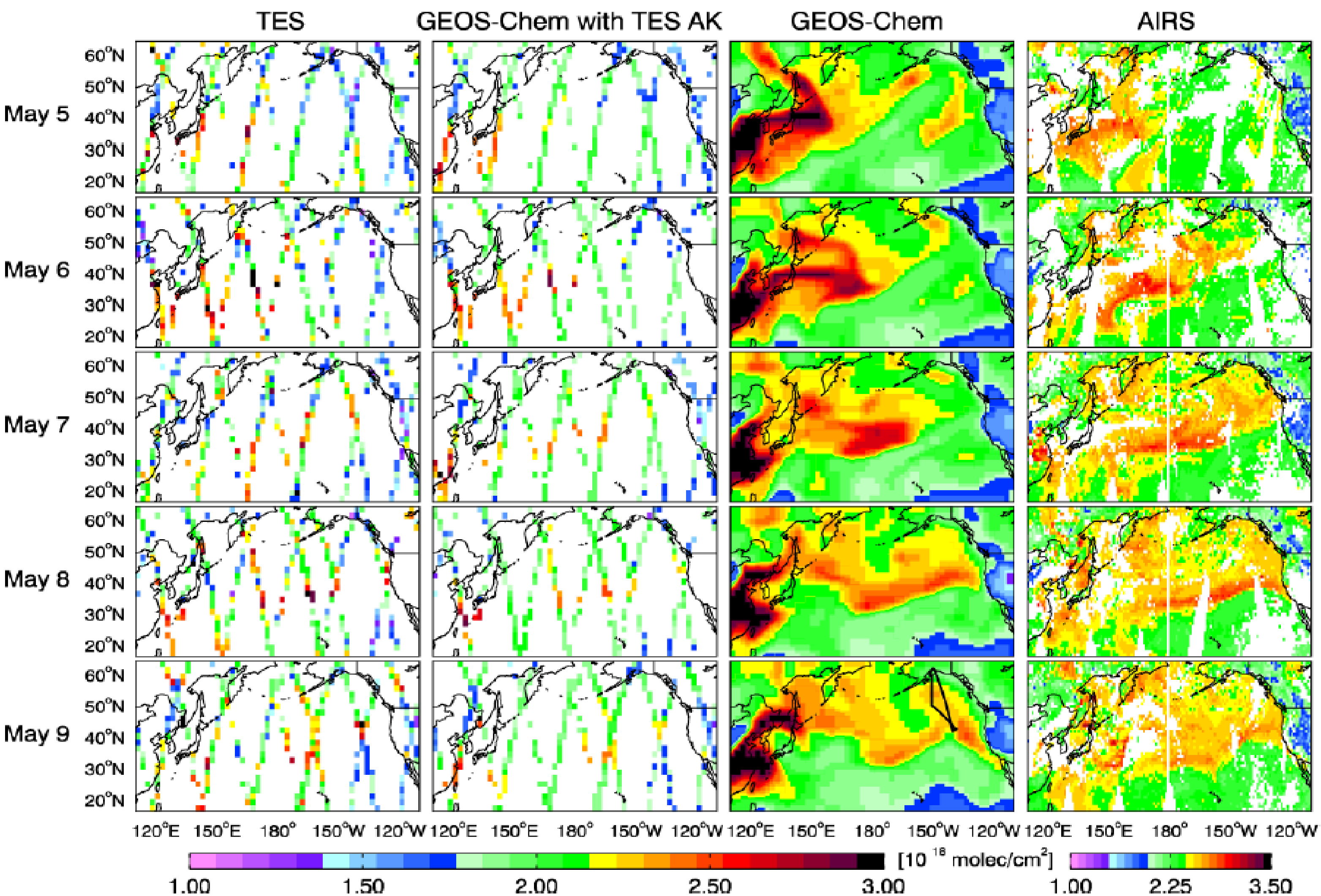
- East Asian PM pollution contributes 6% of N.A. DRE



# East Asian Influence: Satellite Obs. and CTMs

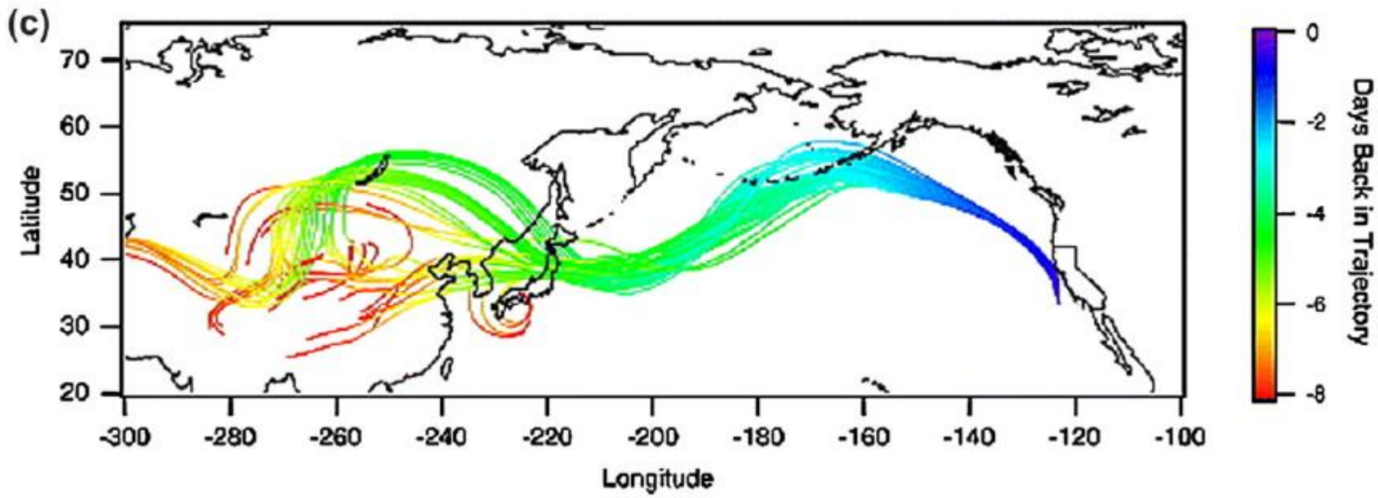
**Carbon Monoxide**

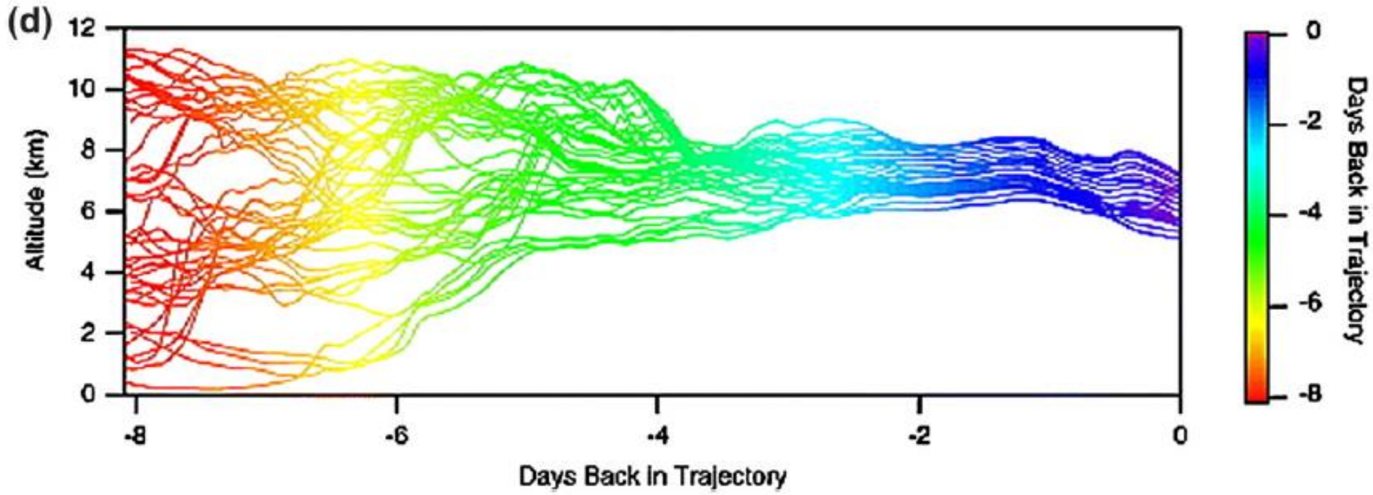



Zhang et al., 2008, ACP

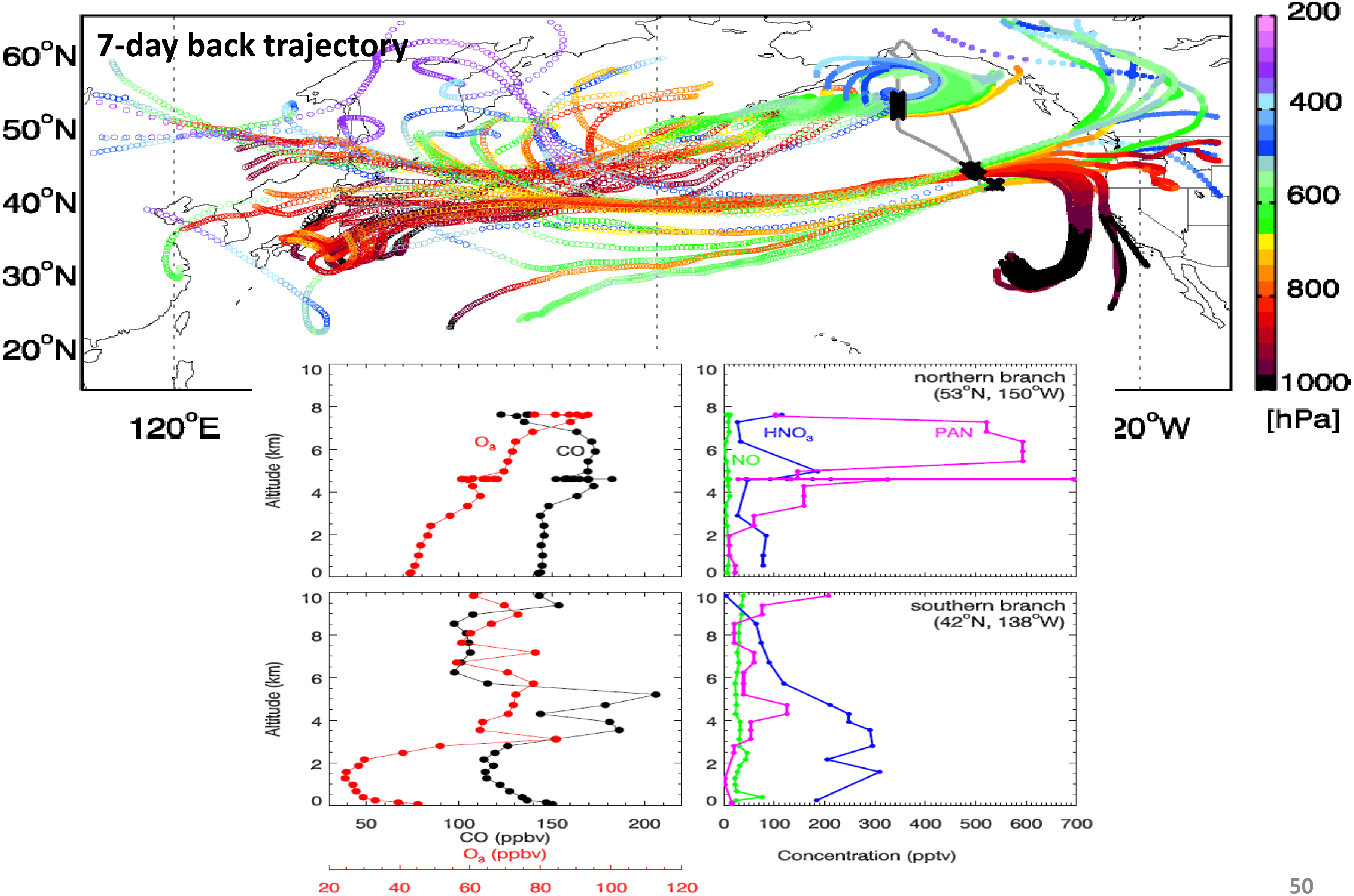
# Asian Influence: Back Trajectory Analysis

**Ozone**



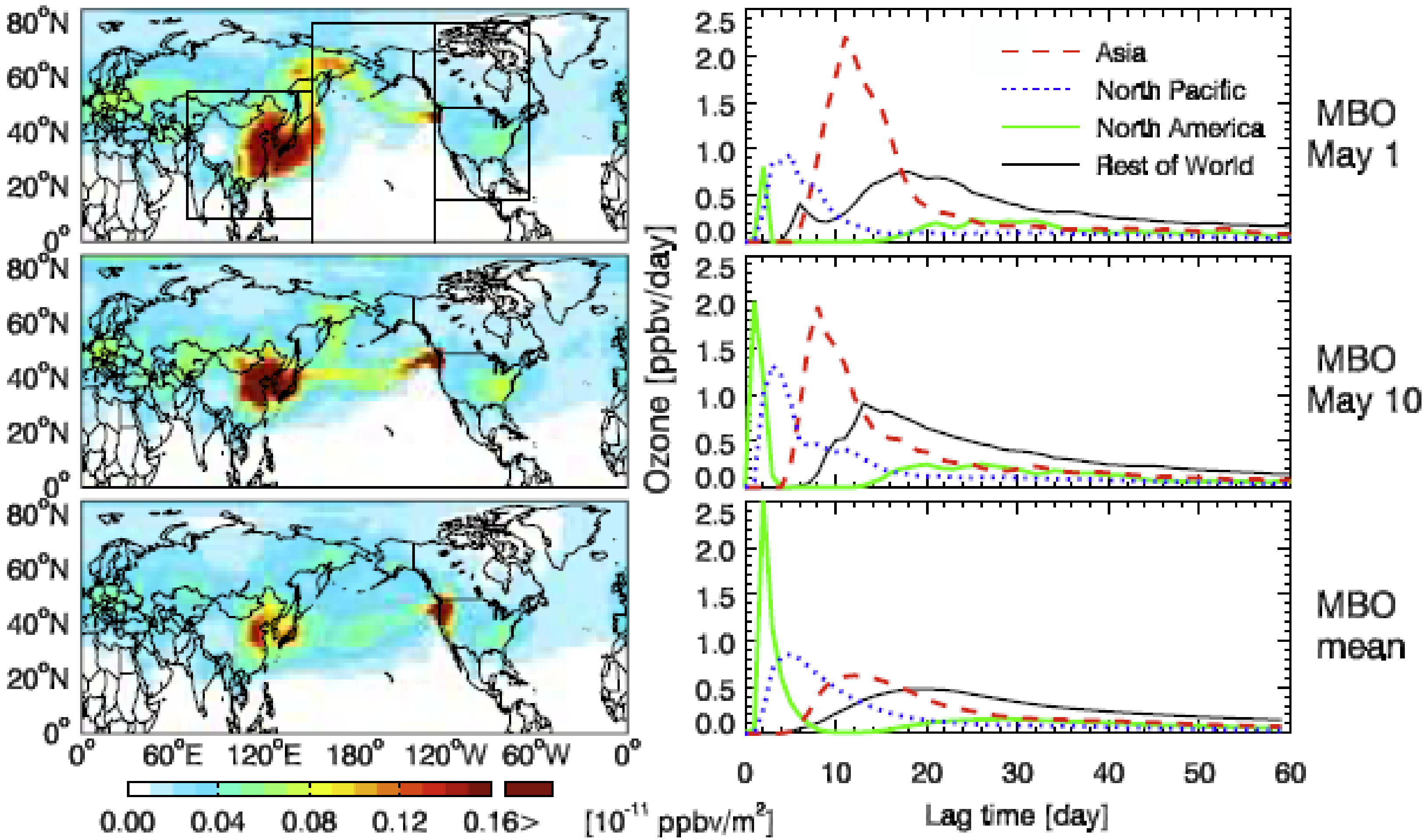
# Trans-Pacific Transport and Transformation




Zhang et al., 2008, ACP

# Adjoint Modeling for Intercontinental Transport

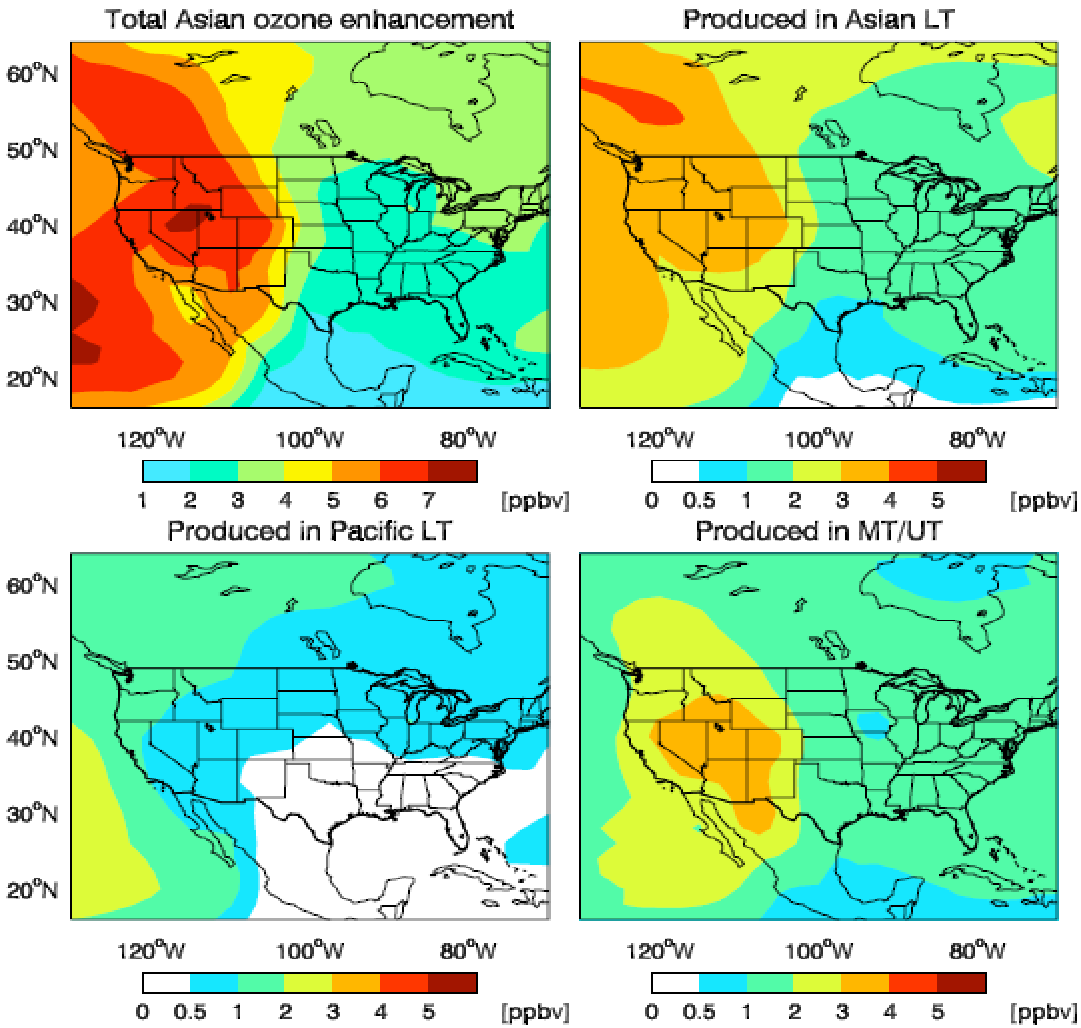
**Ozone**



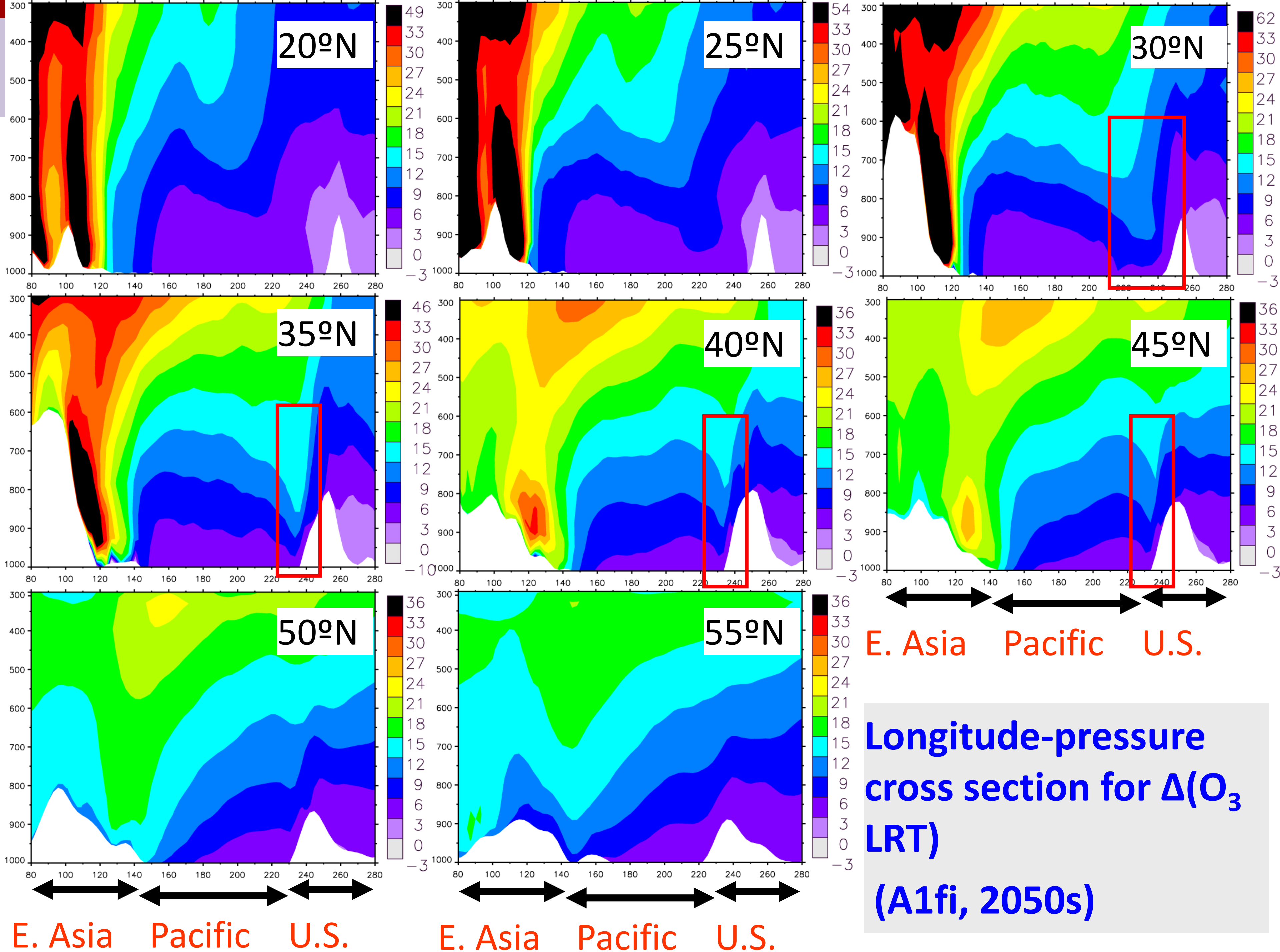

Zhang et al., 2010, GRL

An adjoint model is the transpose of a forward model; it is used for inversion studies

# Springtime U.S. $O_3$ Enhancement due to Transpacific Transport



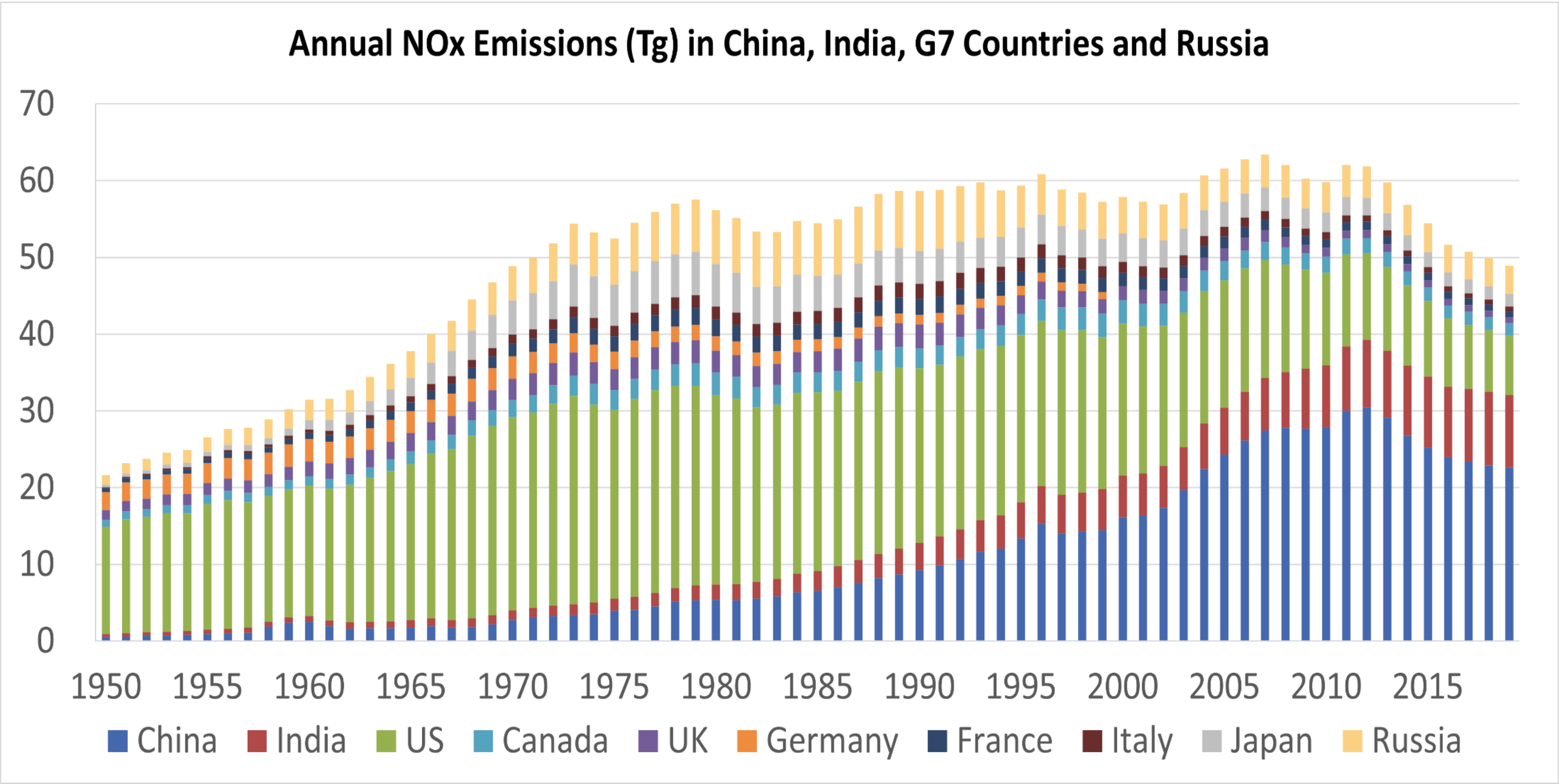

Zhang et al., 2008, ACP




**Longitude-pressure cross section for $\Delta(O_3$ LRT)**

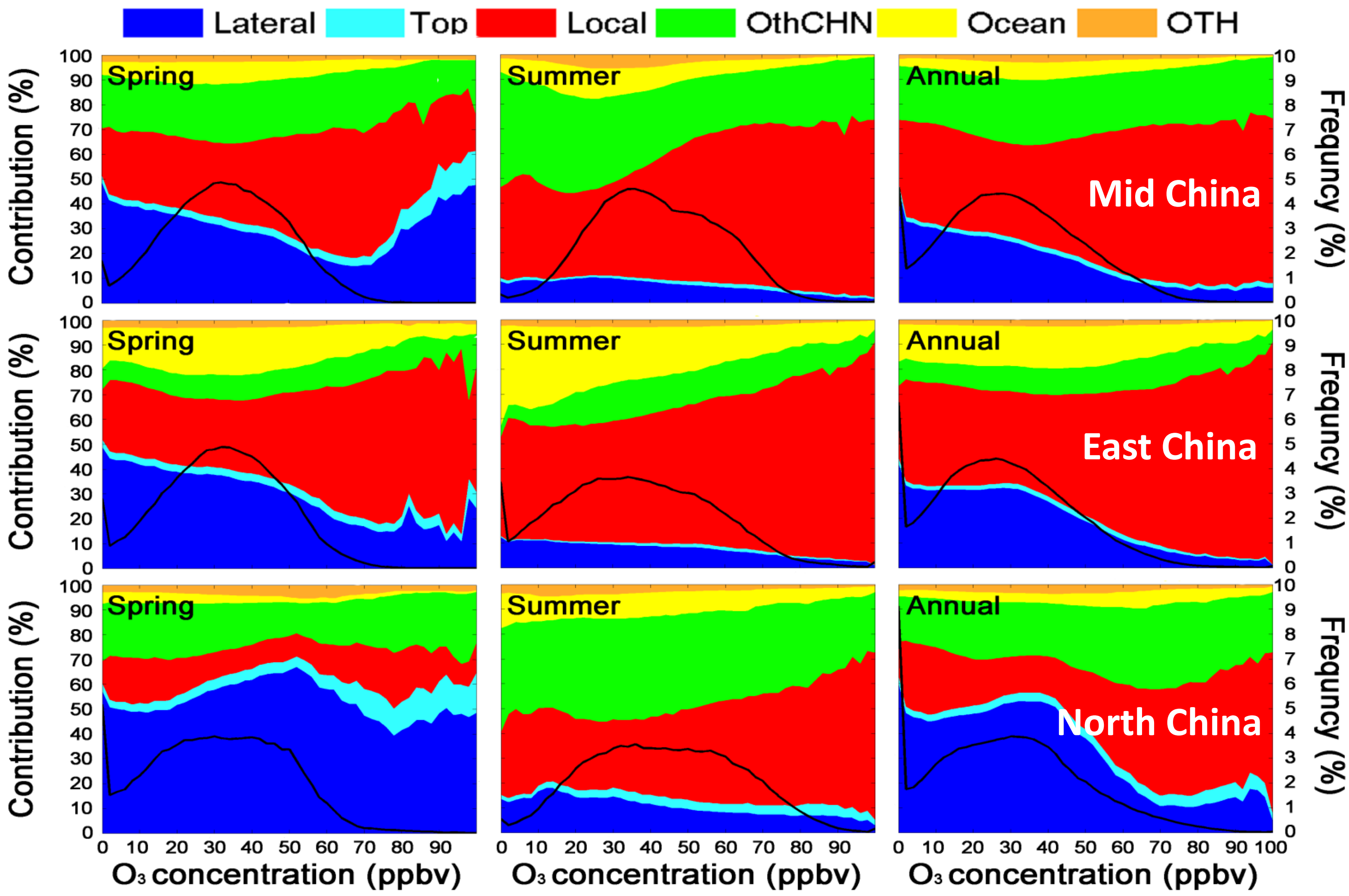
**(A1fi, 2050s)**

# Anthropogenic Emissions of NOx: 1950-2019




CEDS v2 inventory

# Strong Inflow of Ozone into Eastern China

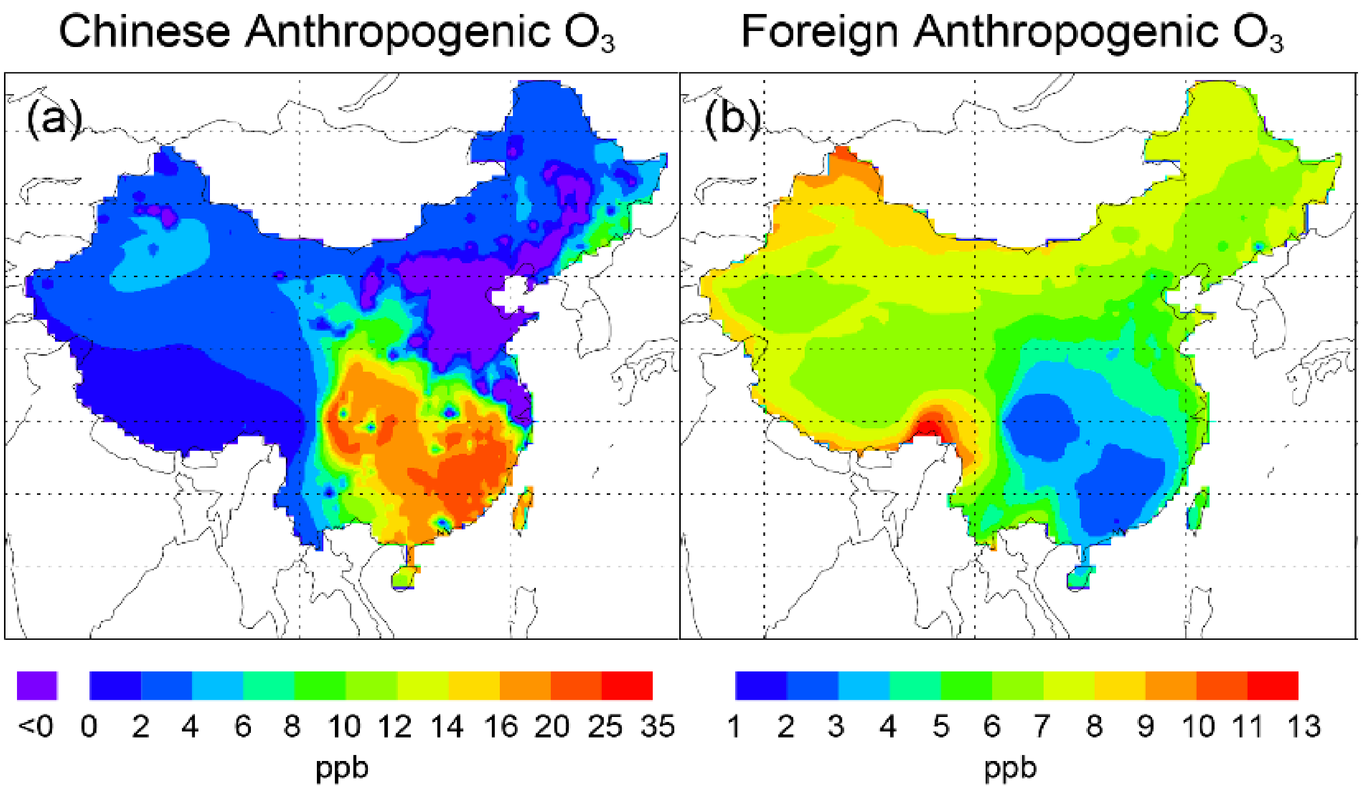



**For 2010**

Li et al., 2016, AR

# Foreign Pollution Greatly Affect China's $O_3$

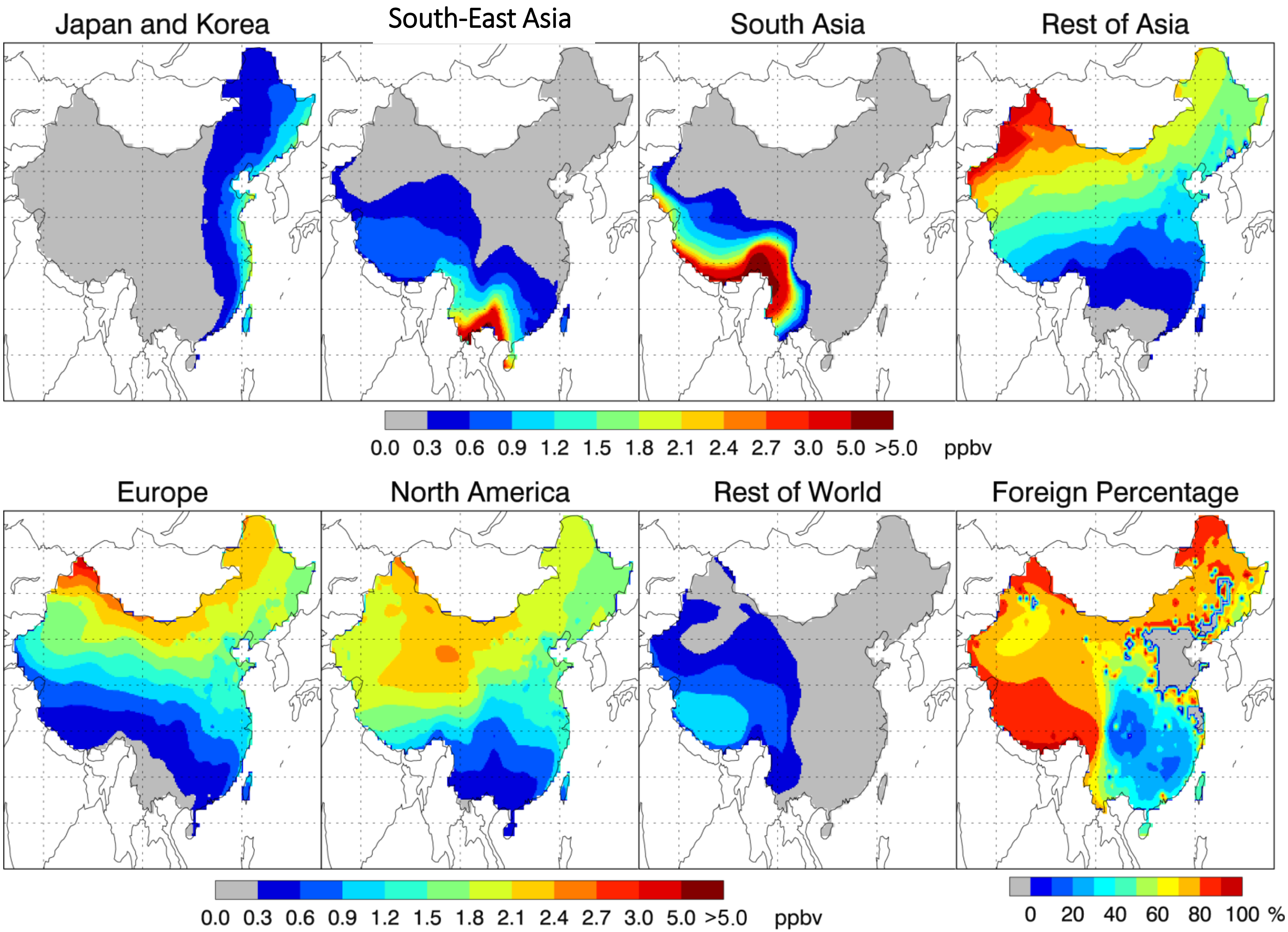
- **2–11 ppb of Surface $O_3$ over China in Spring 2008 are Foreign**




Method: *Zero-out* + *Tagged $O_3$* + *Linear weighting*

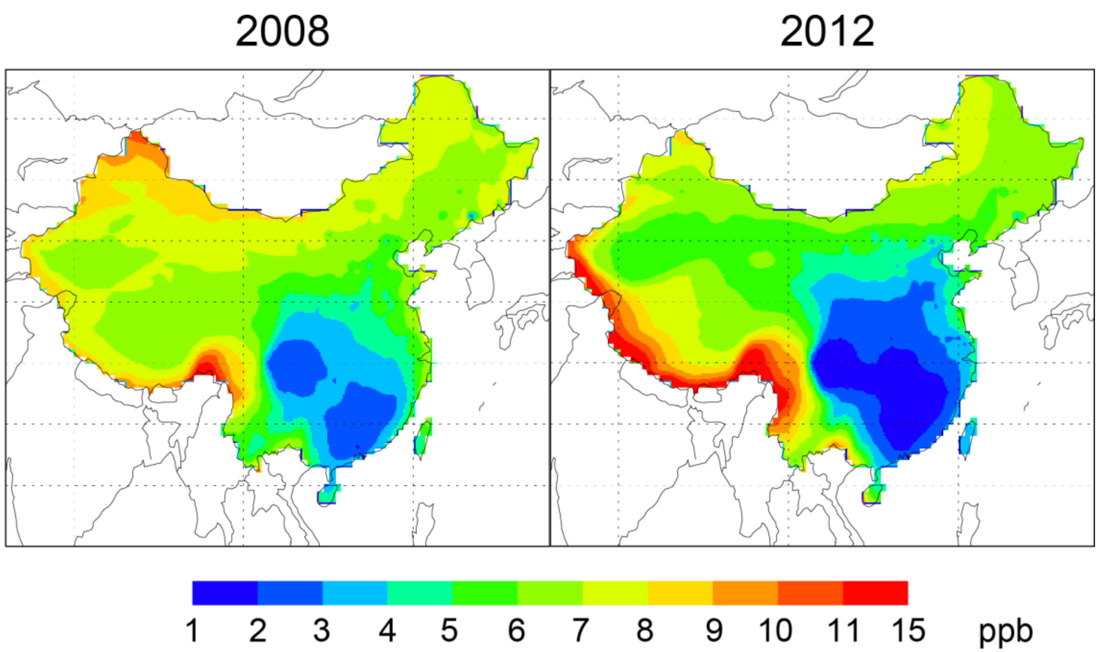
Ni et al., ACP, 2018

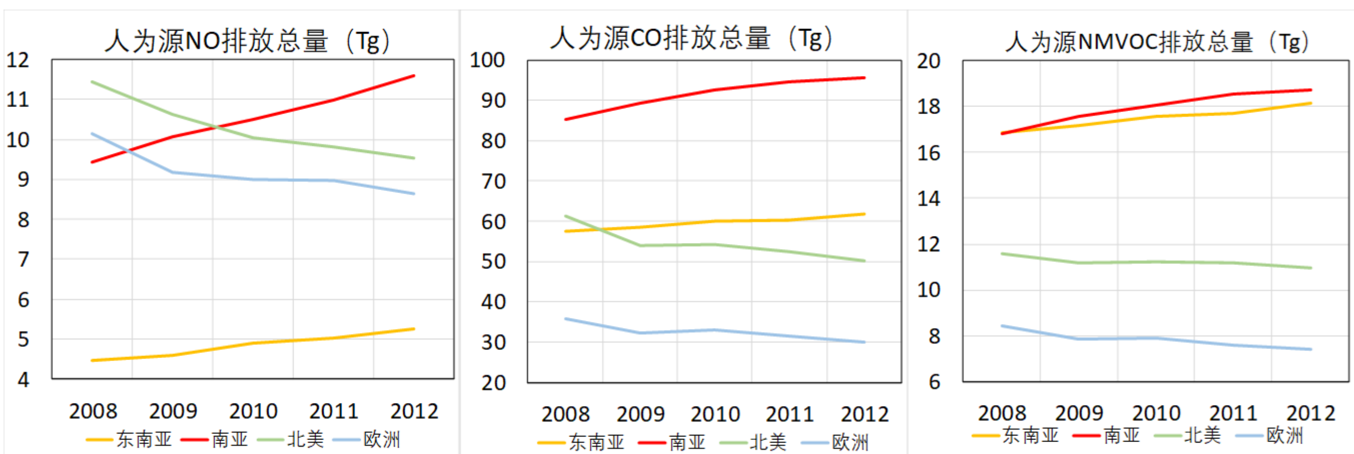
# Foreign Pollution Greatly Affect China's $O_3$: Spring 2008




Ni, R.-J. et al., 2018, ACP

# Changes in Springtime Foreign Anthropogenic Surface $O_3$

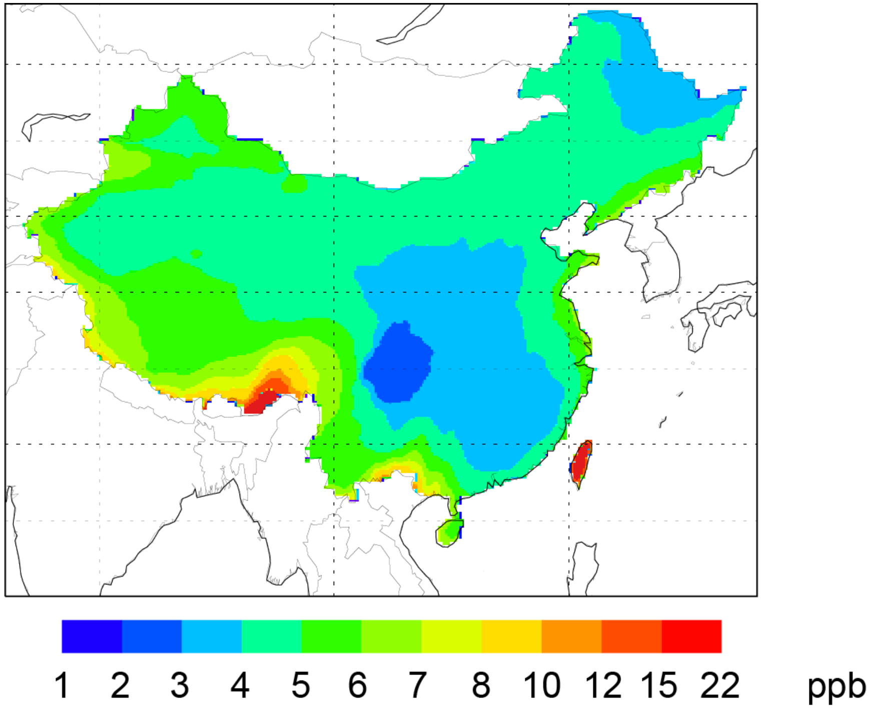


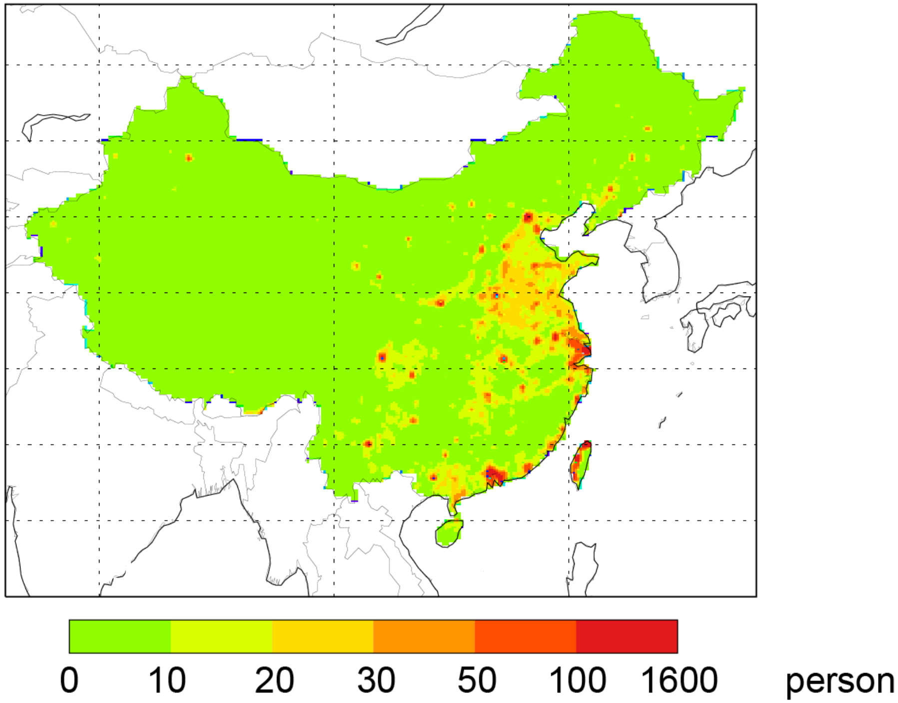

Ni et al., ACP, 2018

# Mortality in Mainland China Caused by Atmospheric Transboundary Pollution From Other Regions

**Transboundary MDA8 Ozone in 2015**



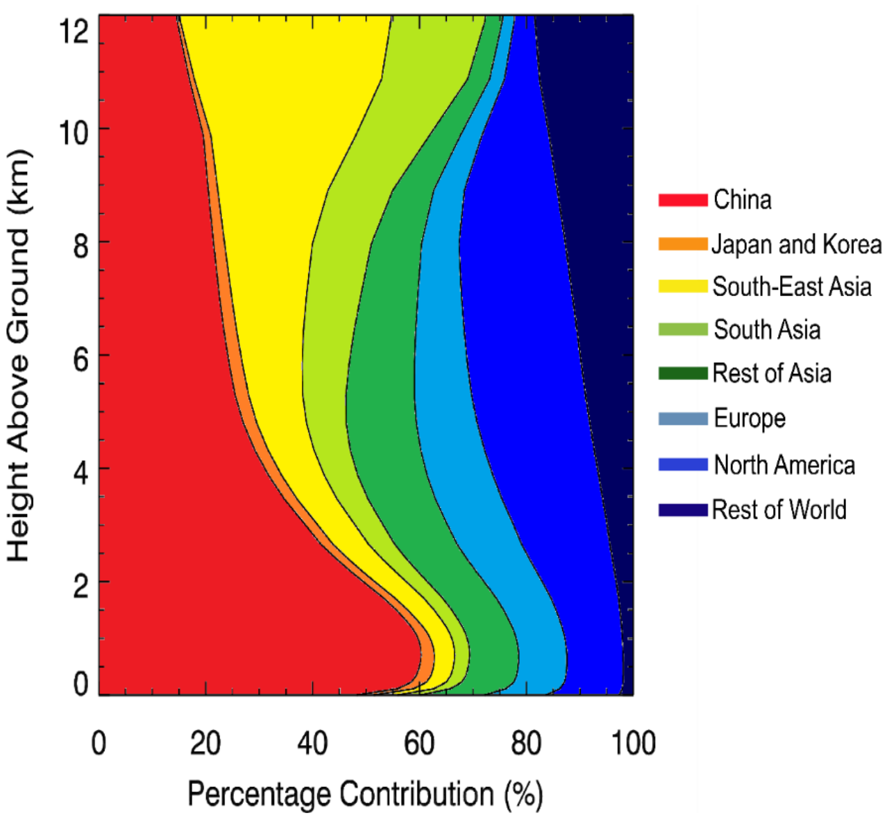

**Deaths wrt transboundary $O_3$**



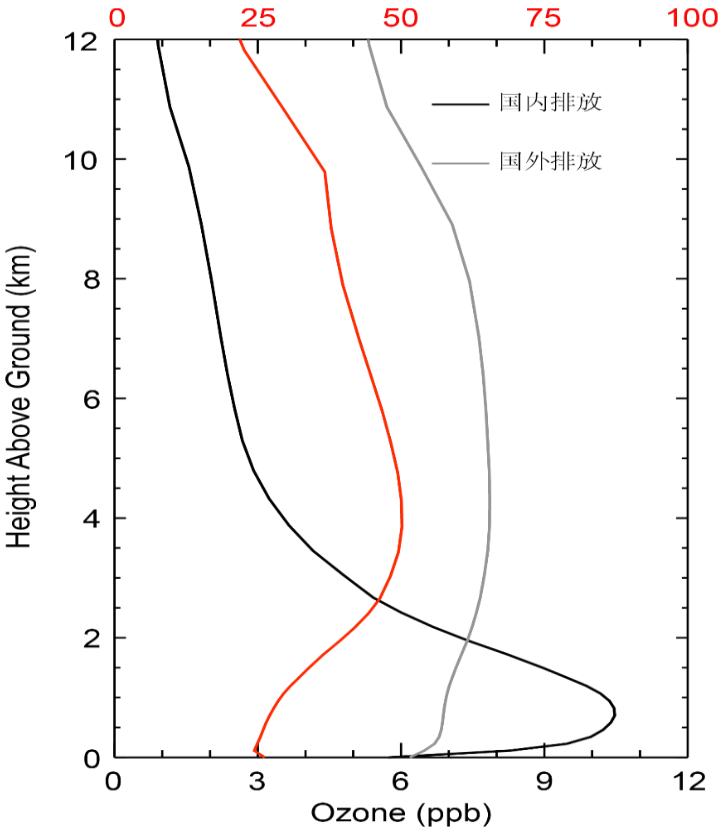

Ni et al., in prep

# Large Fractions of Tropospheric Anthropogenic $O_3$ over China in Spring 2008 are Foreign

**% of anthropogenic $O_3$ contributed by a region**




**% of $O_3$ produced within foreign source regions**



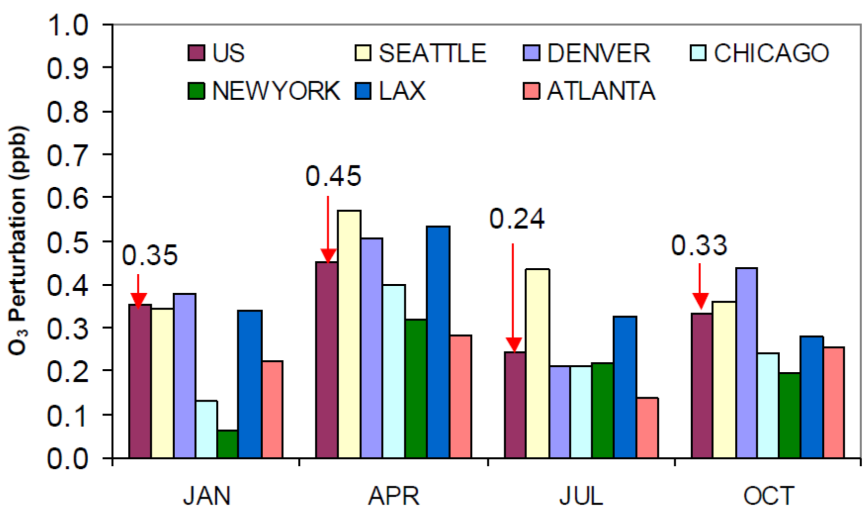

Method: *Zero-out* + *Tagged $O_3$* + *Linear weighting*

Ni et al., ACP, 2018

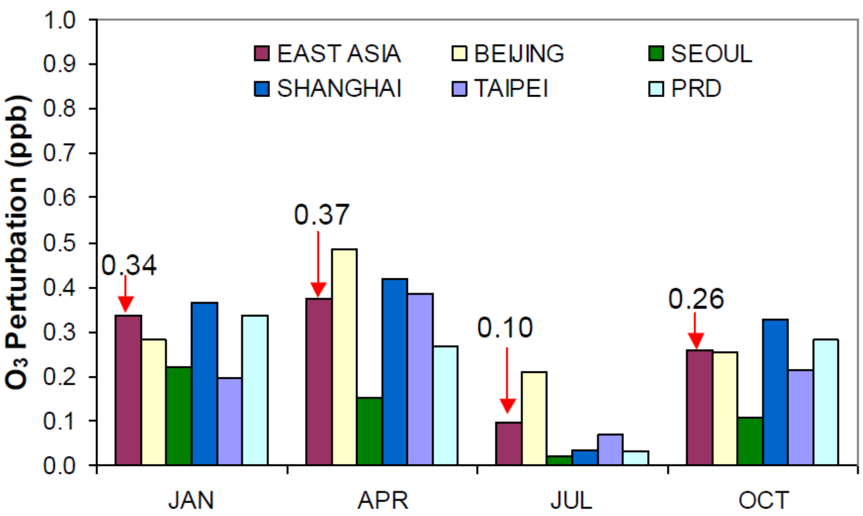
# Transboundary Ozone from E. Asia versus NA

WRF-Chem simulation at 36 x 36 km$^2$

**20% changes in anthro emis in E. Asia**



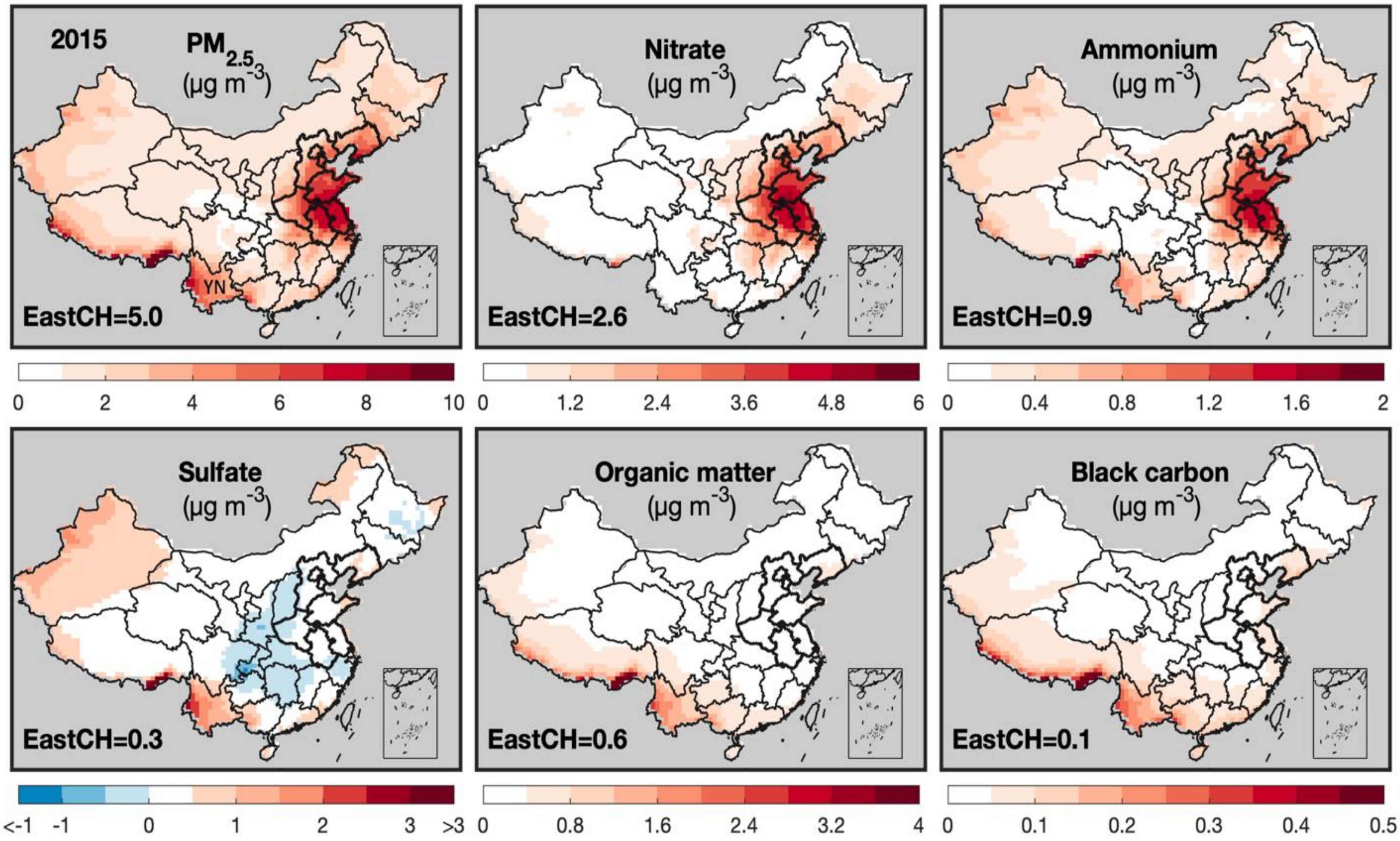

**20% changes in anthro emis in NA**




HTAP, 2010 (P206)

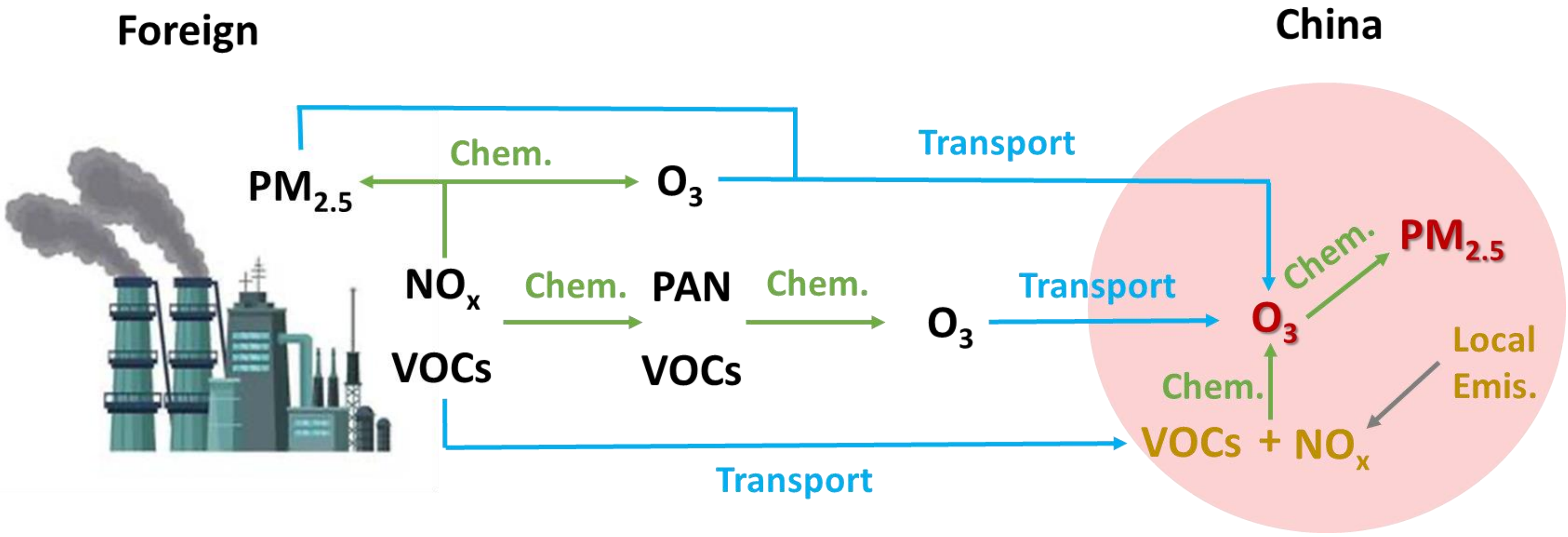
# Foreign Pollution Transport Worsens Chinese $PM_{2.5}$

## Foreign Contribution to E. China = Direct Transport (30%) + Chemistry (70%)




Xu et al., ACP, 2023, Highlight Paper

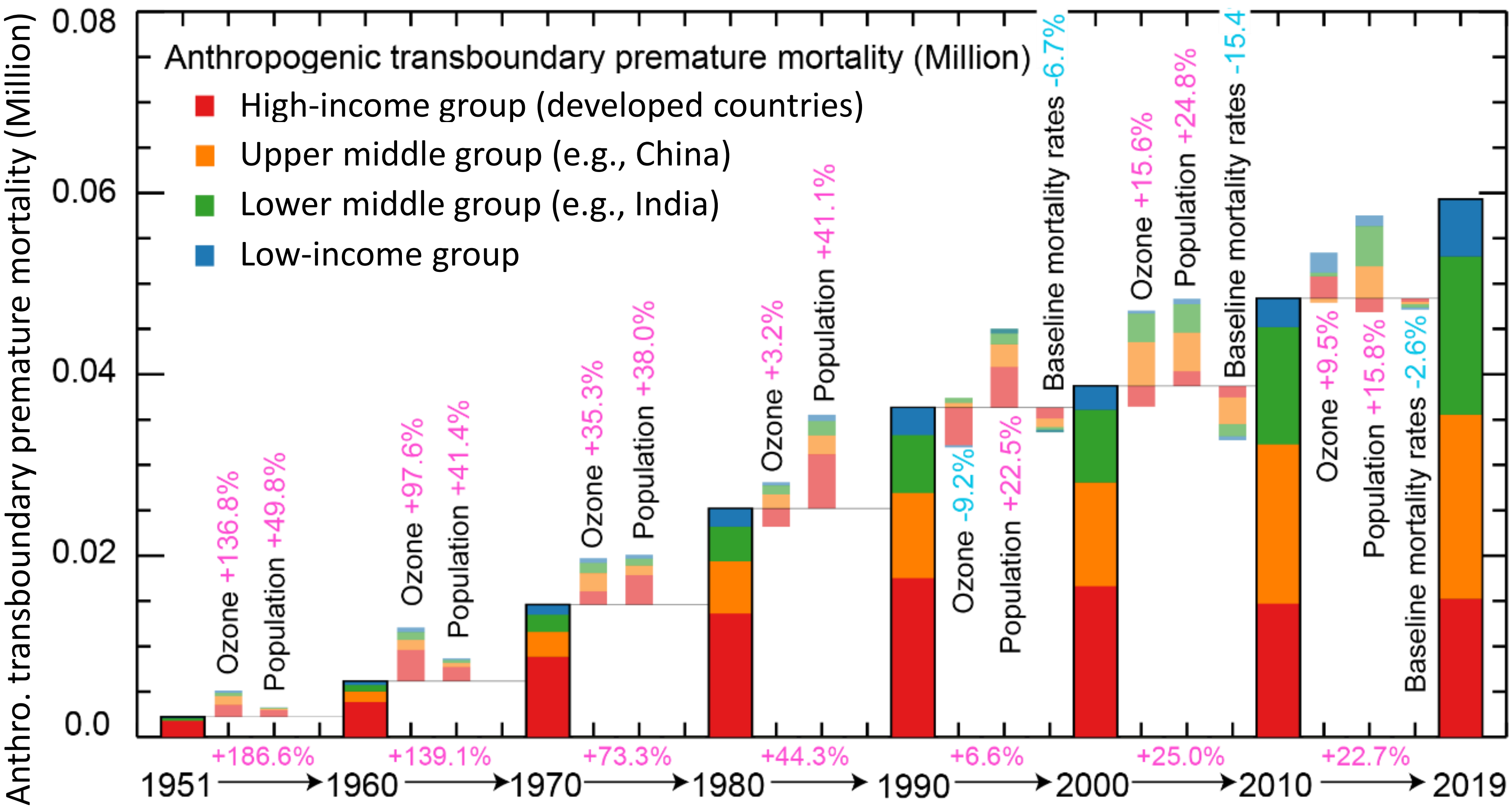
# Complex Chemical-Transport Mechanism



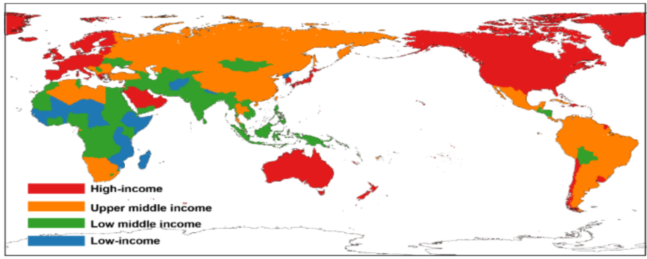

**Transboundary pollution mechanisms:**

1. **Emission or formation and then transport**
2. **Formation during transport**
3. **Transport and then interaction with pollution @ receptor**

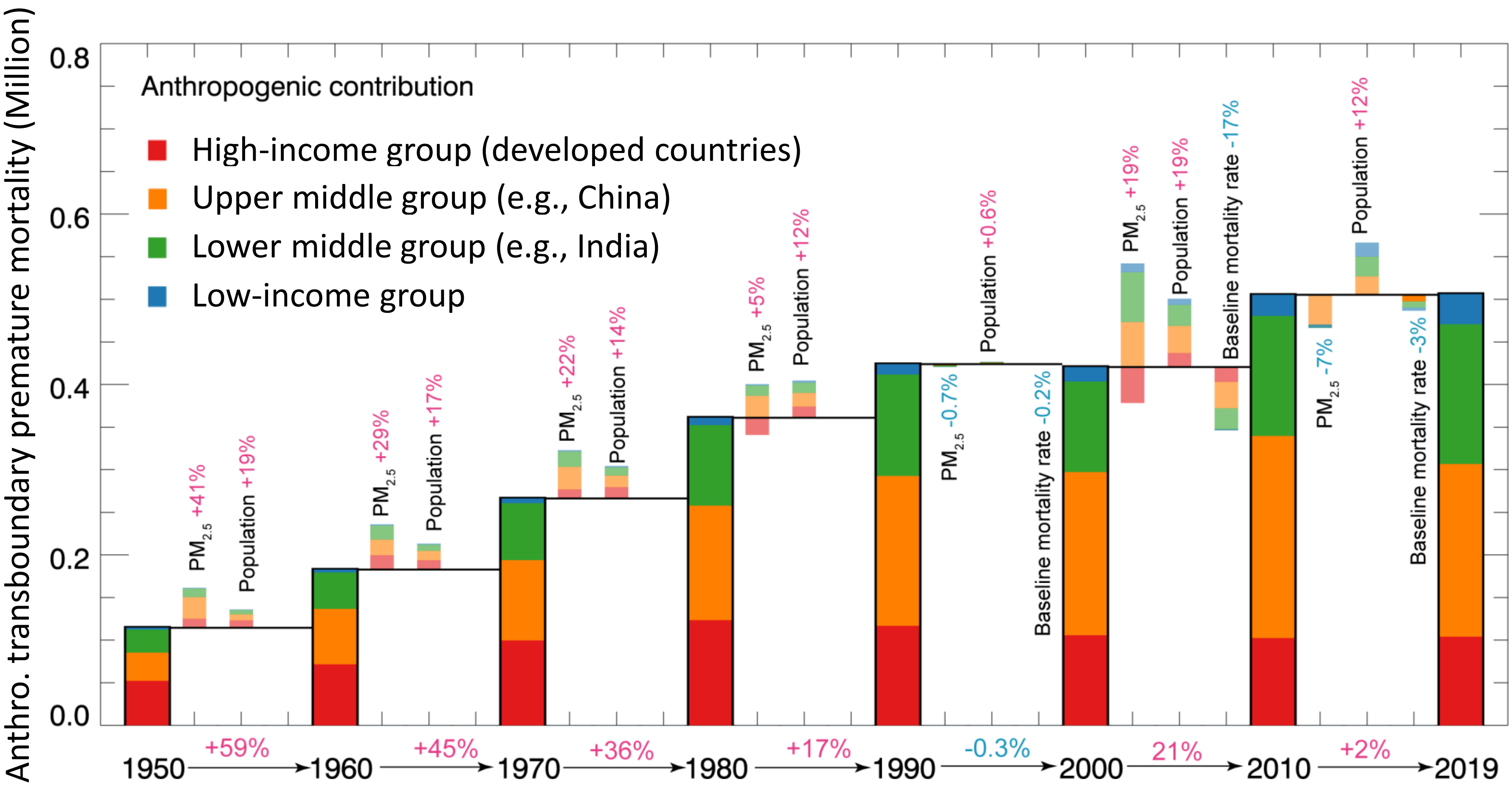
# Historical Transboundary Ozone Mortality via Atmospheric Transport



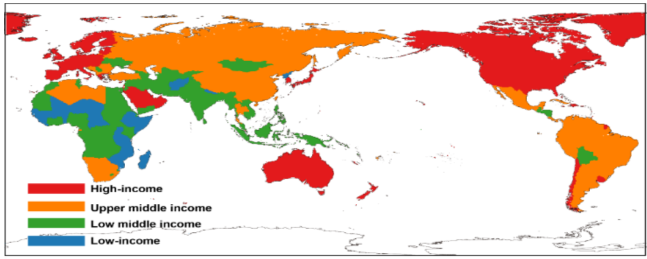

Chen et al., ERL, 2023



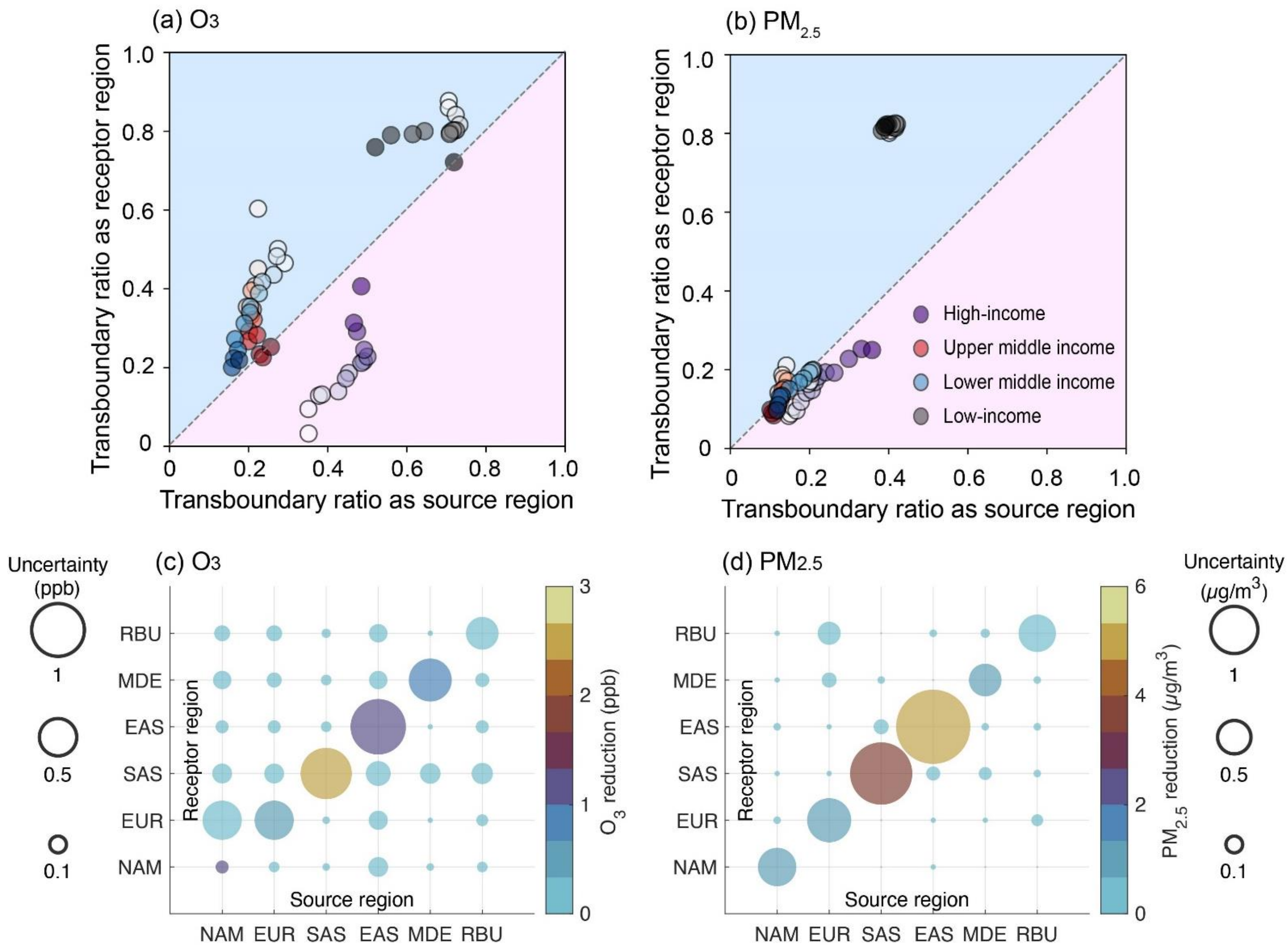
# Considerable Historical Global Deaths Due to Transported $PM_{2.5}$




Chen et al., Science Bulletin, 2022



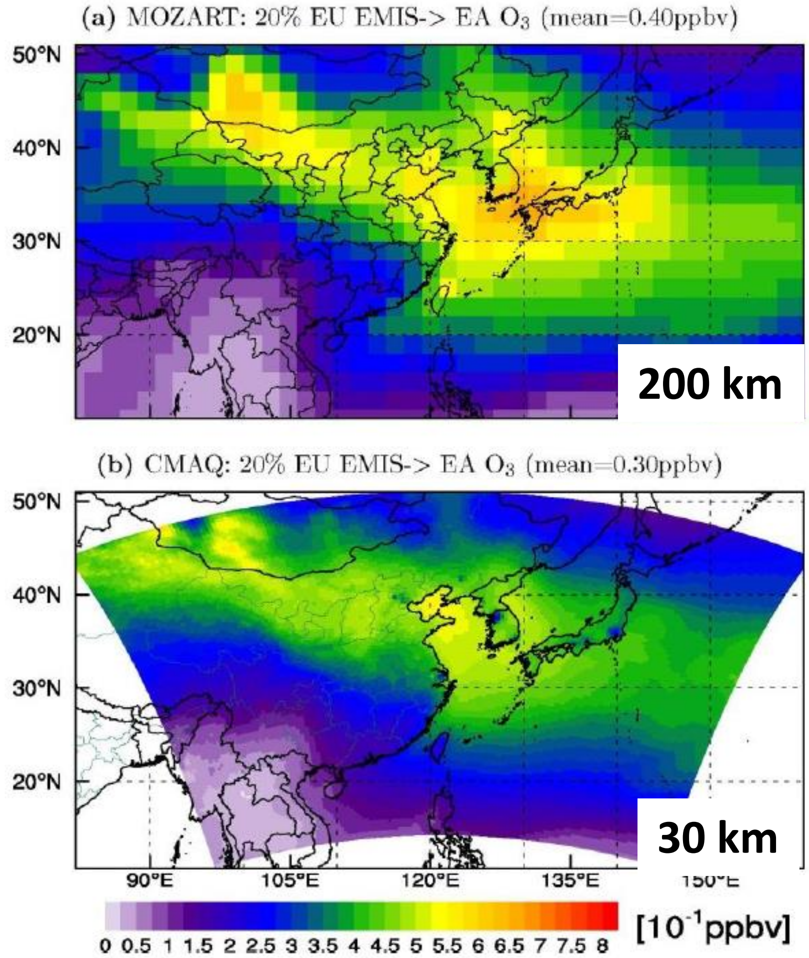
# Historical Transboundary Pollution via Atmospheric Transport




Lin et al., under review

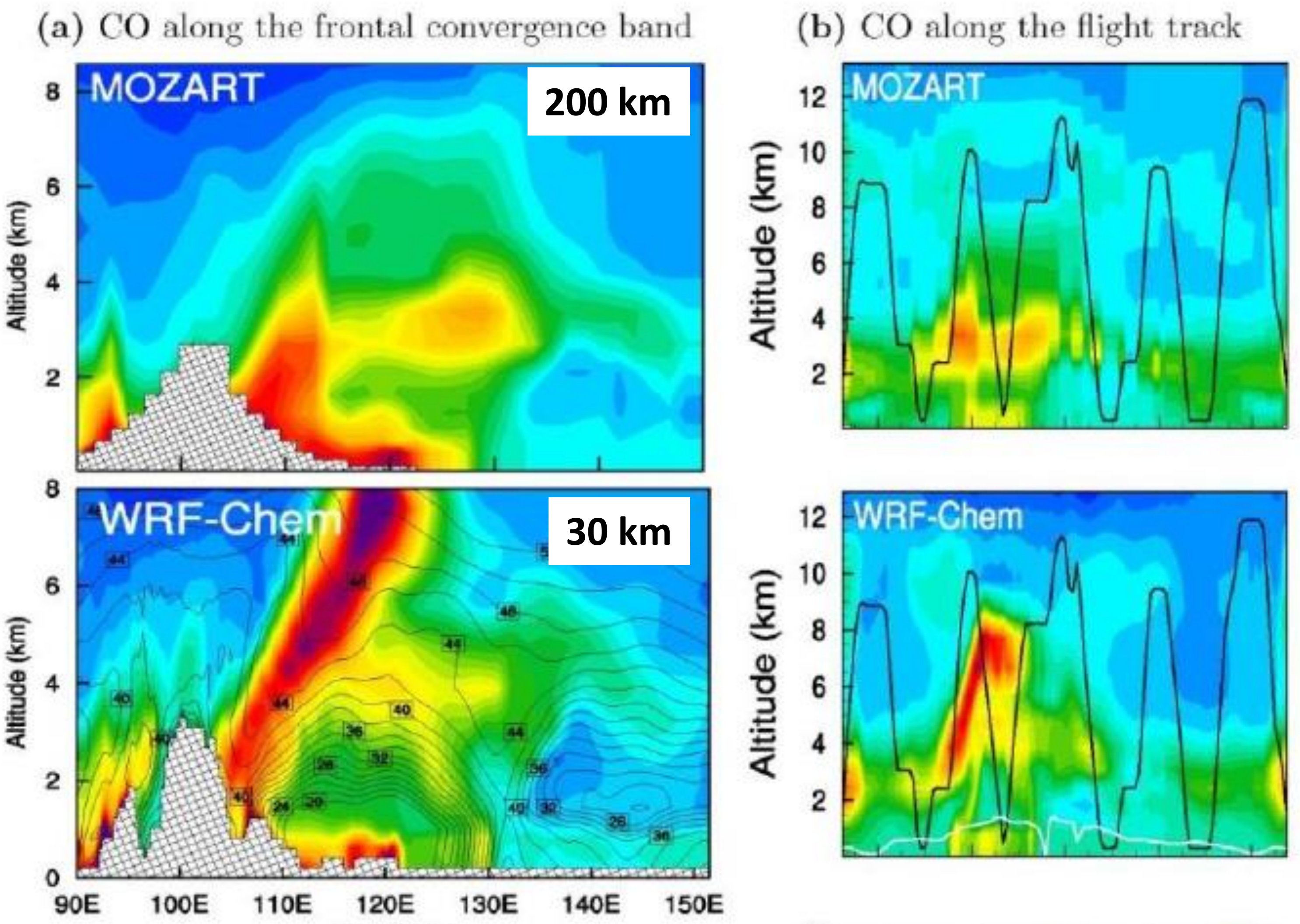
# Uncertainty in Import Due to Model Resolution

**Effects of reducing EU anthro emis by 20% on China' ozone in March 2001**




Lin et al., 2010, ACP

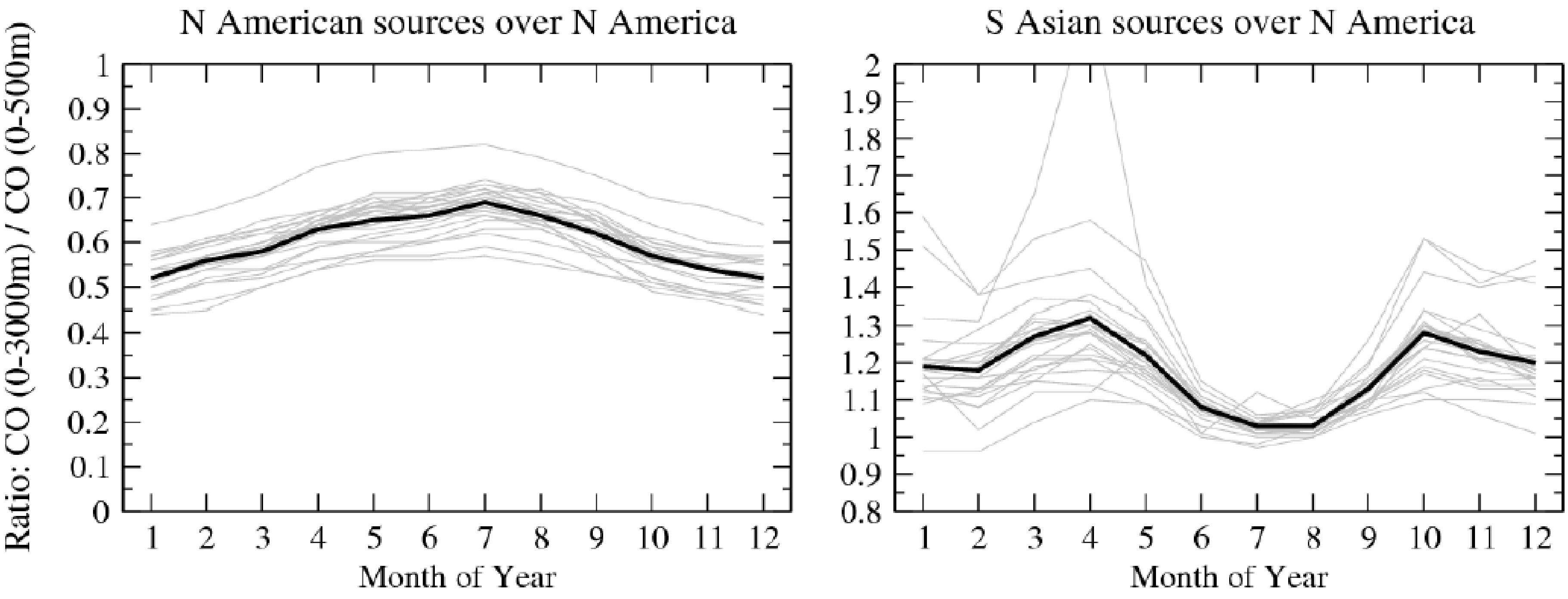
# Uncertainty in Export Due to Model Resolution




March 2001

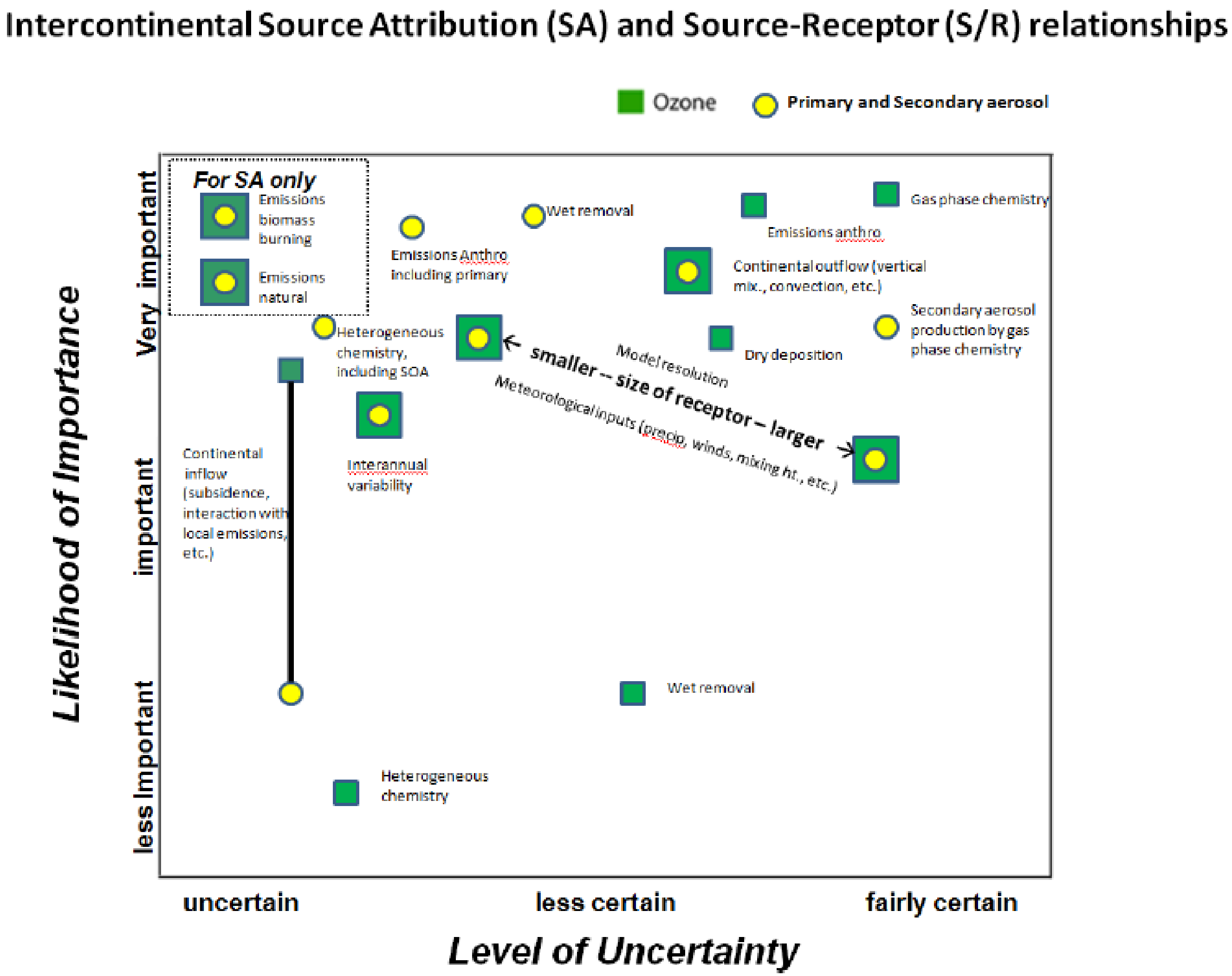
Lin et al., 2010, ACP

# Uncertainty Due to Model Transport Process




HTAP, 2010

# Uncertainties in Model Assessment



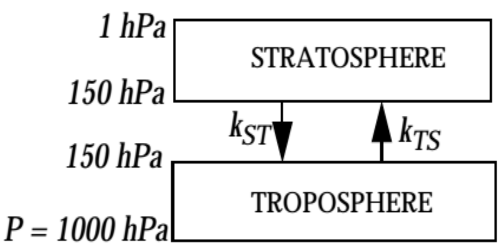

HTAP, 2010

# Quiz

- Role of dynamics versus chemistry in atmospheric pollution transport
- For a secondary pollutant such as nitrate aerosol, what are the factors affecting the distance it can be transported?
- For a randomly oriented horizontal wind fields at a speed of 5 km/hour with a temporally mean speed of zero and a directional change every hour, what kind of process can the horizontal movement be approximated as? Then, if the chemical lifetime of a species is 24 hours, estimate the characteristic distance of effective diffusion.
- Through what atmospheric mechanisms would the emissions from USA affect China?
- How to compare the pollution transported into versus out of China, in terms of concentrations and exposure?
- Compare the transport of PM and ozone
- How would climate change affect the transport of pollutants?

# Stratosphere-Troposphere Exchange

The rate of exchange of air between the troposphere and the stratosphere is critical for determining the potential of various pollutants emitted from the surface to reach the stratosphere and affect the stratospheric ozone layer. One of the first estimates of this rate was made in the 1960s using measurements of strontium-90 ($^{90}Sr$) in the stratosphere. Strontium-90 is a radioactive isotope (half-life 28 years) produced in nuclear explosions. It has no natural sources. Large amounts of $^{90}Sr$ were injected into the stratosphere in the 1950s by above-ground nuclear tests. These tests were banned by international treaty in 1962. Following the test ban the stratospheric concentrations of $^{90}Sr$ began to decrease as $^{90}Sr$ was transferred to the troposphere. In the troposphere, $^{90}Sr$ is removed by wet deposition with a lifetime of 10 days (by contrast there is no rain, and hence no wet deposition, in the stratosphere). An intensive stratospheric measurement network was operated in the 1960s to monitor the decay of $^{90}Sr$ in the stratosphere. We interpret these observations here using a 2-box model for stratosphere-troposphere exchange with transfer rate constants $k_{TS}$ and $k_{ST}$ ($yr^{-1}$) between the tropospheric and stratospheric reservoirs. The reservoirs are assumed to be individually well-mixed.




Let $m_S$ and $m_T$ represent the masses of $^{90}Sr$ in the stratosphere and in the troposphere respectively. Observations of the decrease in the stratospheric inventory for the period 1963-1967 can be fitted to an exponential $m_S(t) = m_S(0)\exp(-kt)$ where $k = 0.77\ yr^{-1}$.

1. Write mass balance equations for $m_S$ and $m_T$ in the 1963-1967 period.

2. Assuming that transfer of $^{90}Sr$ from the troposphere to the stratosphere is negligible (we will verify this assumption later), show that the residence time of air in the stratosphere is $\tau_S = 1/k_{ST} = 1.3$ years.

3. Let $m'_T$ and $m'_S$ represent the total masses of air in the troposphere and the stratosphere, respectively. Show that the residence time of air in the troposphere is $\tau_T = \tau_S\ (m'_T/m'_S) = 7.4$ years. Conclude as to the validity of your assumption in question 2.

4. Hydrochlorofluorocarbons (HCFCs) have been adopted as replacement products for the chlorofluorocarbons (CFCs), which were banned by the Montreal protocol because of their harmful effect on the ozone layer. In contrast to the CFCs, the HCFCs can be oxidized in the troposphere, and the oxidation products washed out by precipitation, so that most of the HCFCs do not penetrate into the stratosphere to destroy ozone. Two common HCFCs have trade names HCFC-123 and HCFC-124; their lifetimes against oxidation in the troposphere are 1.4 years and 5.9 years, respectively. There are no other sinks for these species in the troposphere. Using our 2-box model, determine what fractions of the emitted HCFC-123 and HCFC-124 penetrate the stratosphere.



Jacob, 1999

# Interhemispheric Exchange

In this problem we use observations of the radioactive gas $^{85}$Kr to determine the characteristic time for exchange of air between the northern and southern hemispheres. We consider a 2-box model where each hemisphere is represented by a well-mixed box, with a rate constant $k$ (yr$^{-1}$) for mass exchange between the two hemispheres. Our goal is to derive the residence time $\tau = 1/k$ of air in each hemisphere.

Krypton-85 is emitted to the atmosphere during the reprocessing of nuclear fuel. It is removed from the atmosphere solely by radioactive decay with a rate constant $k_c$ = 6.45x10$^{-2}$ yr$^{-1}$. The sources of $^{85}$Kr are solely in the northern hemisphere and their magnitudes are well known due to regulation of the nuclear industry. Atmospheric concentrations of $^{85}$Kr are fairly well known from ship observations. In 1983 the global $^{85}$Kr emission rate was $E$ = 15 kg yr$^{-1}$, the total atmospheric mass of $^{85}$Kr in the northern hemisphere was $m_N$ = 93 kg, and the total atmospheric mass of $^{85}$Kr in the southern hemisphere was $m_S$ = 86 kg.

1. Assume that the interhemispheric difference in the atmospheric mass of $^{85}$Kr is at steady state, that is, $d(m_N - m_S)/dt = 0$ (we will justify this assumption in the next question). Express $\tau$ as a function of $E$, $k_c$, $m_N$, $m_S$ and solve numerically using the 1983 values.

2. The global emission rate of $^{85}$Kr was increasing during the 1980s at the rate of 3% yr$^{-1}$. Justify the assumption $d(m_N - m_S)/dt = 0$. [Hint: use the mass balance equation for ($m_N$-$m_S$) to determine the time scale needed for ($m_N$-$m_S$) to adjust to steady state following a perturbation.]

Jacob, 1999

# Regional Outflow

We model the lower atmosphere over the United States as a well-mixed box extending horizontally 5,000 km in the west-east direction. The box is ventilated by a westerly wind of speed $U = 10$ m s$^{-1}$.

1. What is the residence time $\tau_{out}$ (in days) of air in the lower atmosphere over the United States?

2. Consider a pollutant emitted in the United States and having a lifetime $\tau_{chem}$ against chemical loss. Calculate the fraction $f$ of the pollutant exported out of the United States box as a function of the ratio $\tau_{out}/\tau_{chem}$. Plot your result. Comment on the potential for different pollutants emitted in the United States to affect the global atmosphere.

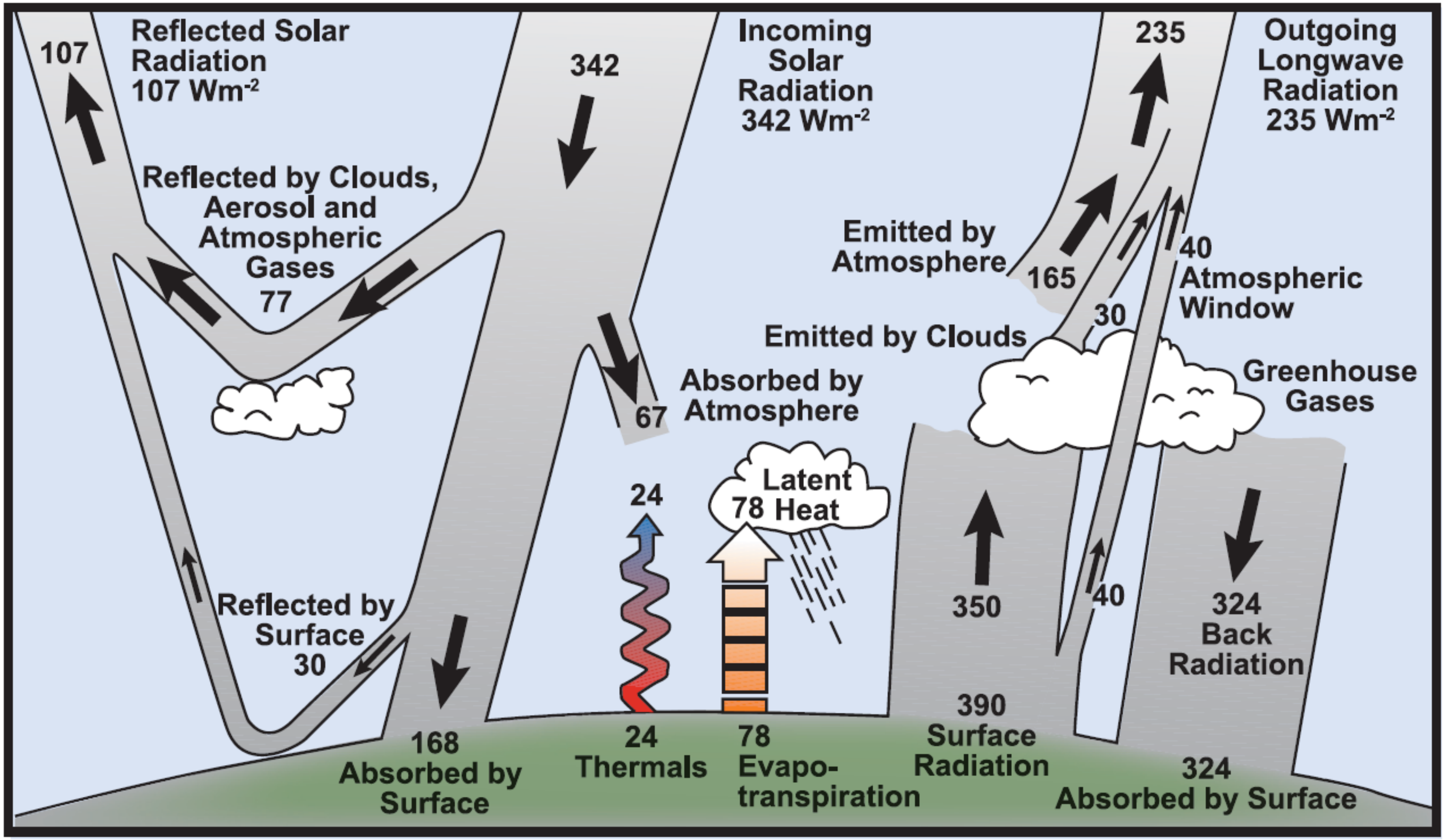
Jacob, 1999

# Local Transport and Transformation

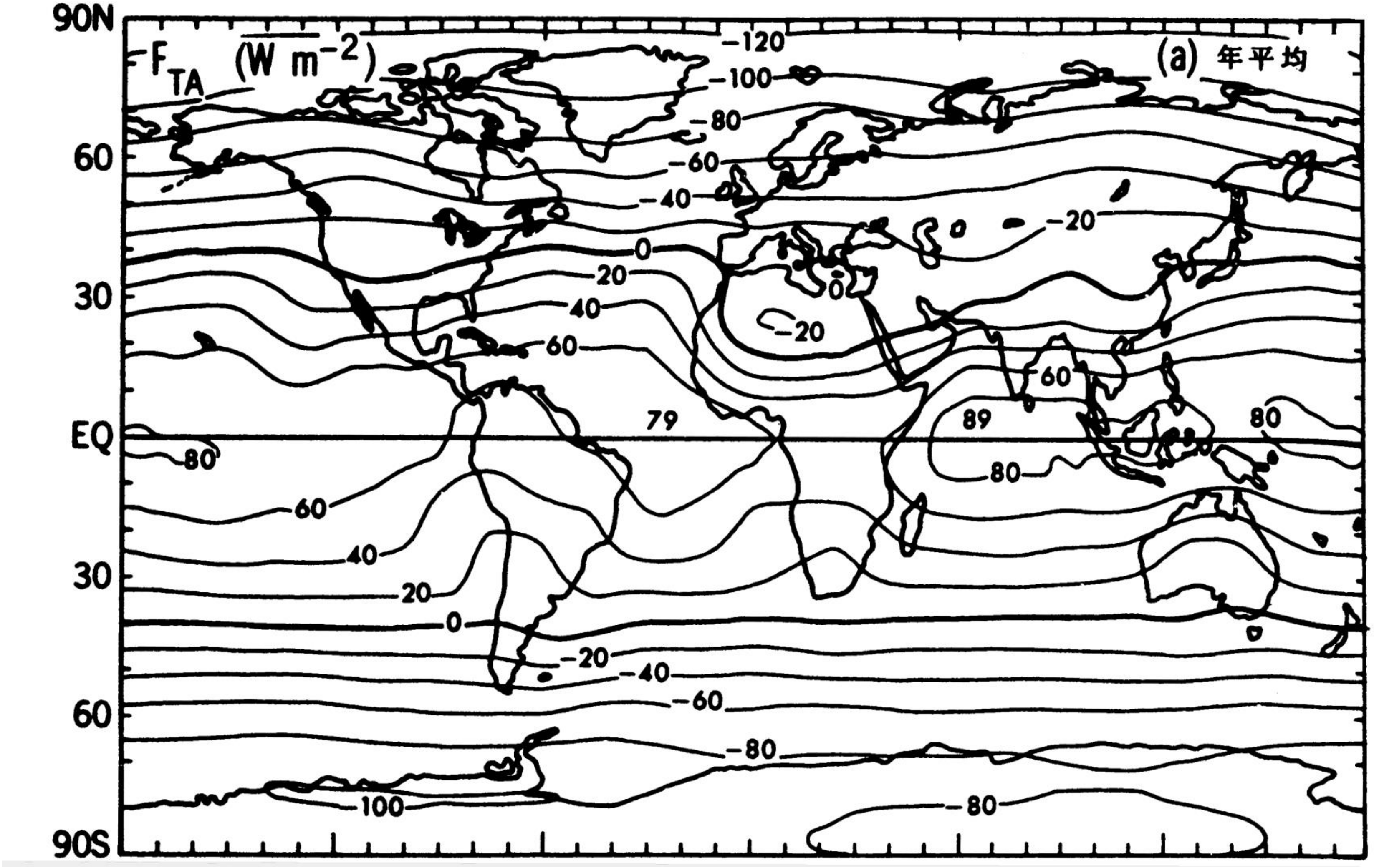
A cluster of coal-fired power plants in Ohio emits sulfur dioxide ($SO_2$) continuously to the atmosphere. The pollution plume is advected to the northeast with a constant wind speed $U = 5$ m s$^{-1}$. We assume no dilution of the plume during transport. Let $[SO_2]_o$ be the concentration of $SO_2$ in the fresh plume at the point of emission; $SO_2$ in the plume has a lifetime of 2 days against oxidation to sulfuric acid ($H_2SO_4$), and $H_2SO_4$ has a lifetime of 5 days against wet deposition. We view both of these sinks as first-order processes ($k_1 = 0.5$ day$^{-1}$, $k_2 = 0.2$ day$^{-1}$). Calculate and plot the concentrations of $SO_2$ and $H_2SO_4$ as a function of the distance $x$ downwind of the power plant cluster. At what distance downwind is the $H_2SO_4$ concentration highest? Look up a map and see where this acid rain is falling.

Jacob, 1999

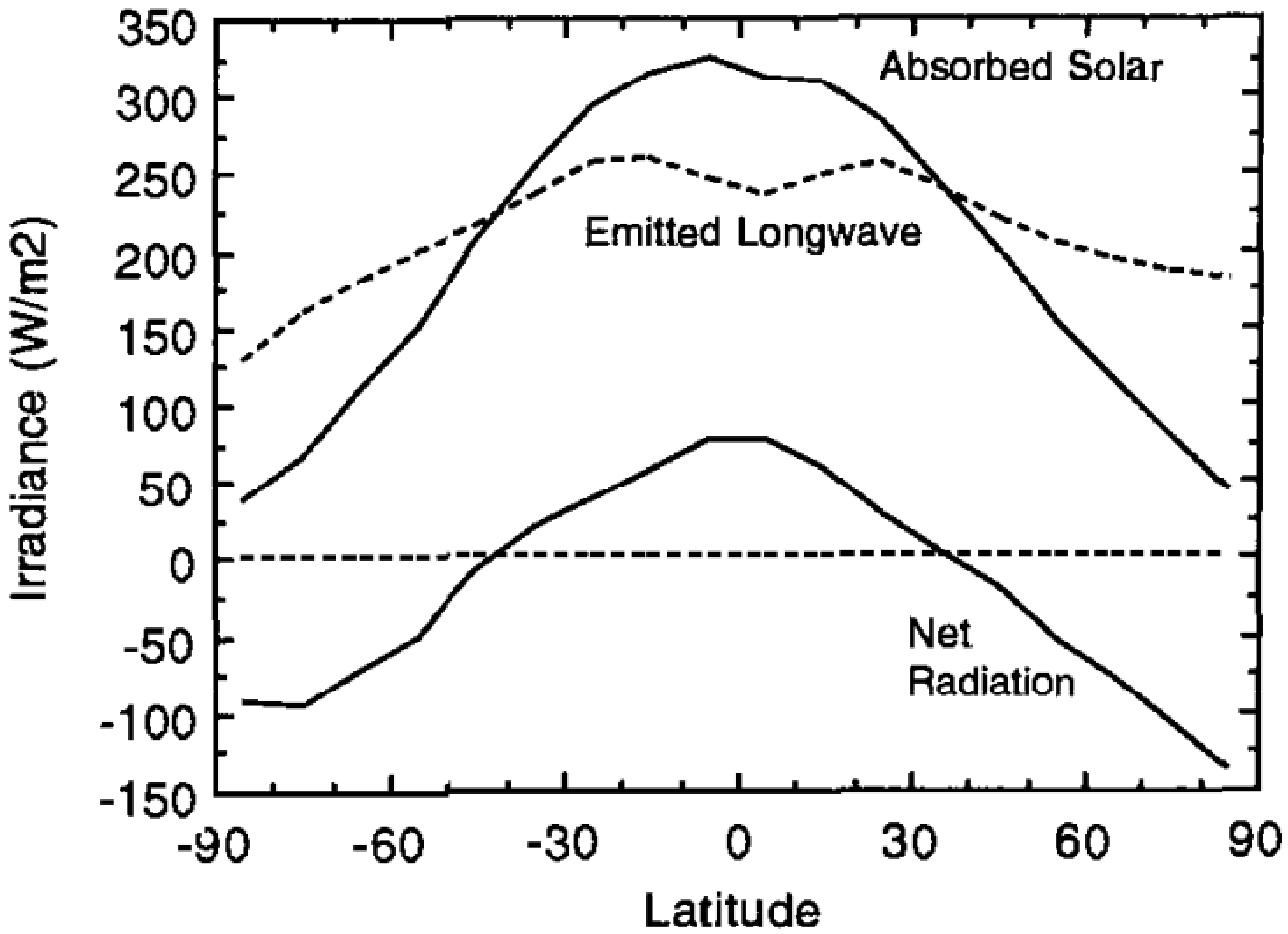
# Energy Budget of Earth Climate



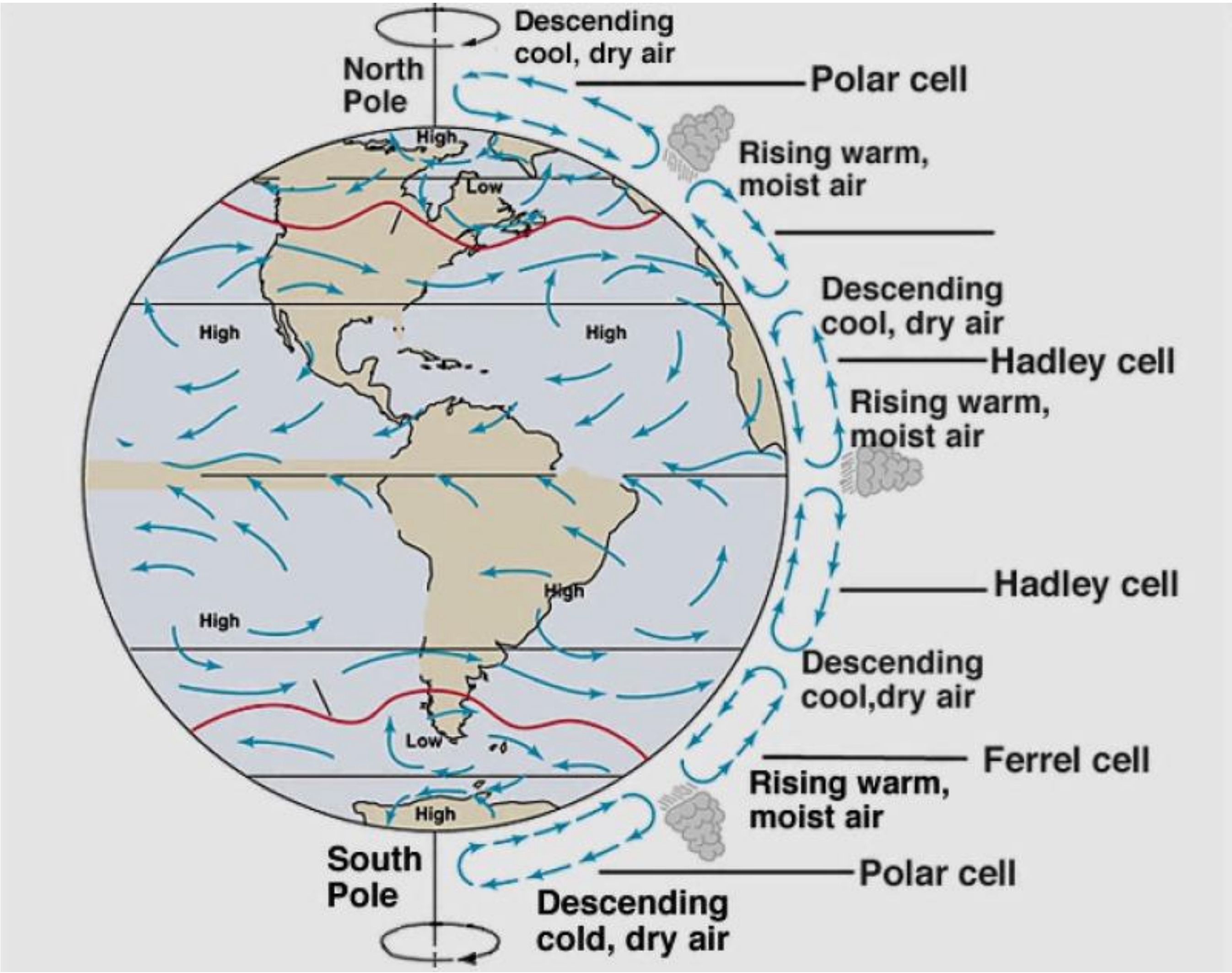
# Annual Mean TOA Net Radiation Flux (Solar + Thermal)



# Annual Mean TOA Net Radiation Flux (Solar + Thermal)



# General Circulation



# Kinematics

**From Stokes' Theorem:**

$$C \equiv \oint V_s ds = \iint \zeta dA$$

**From Gauss's Theorem:**

$$\oint V_n ds = \iint \mathrm{Div}_H \mathbf{V} dA$$

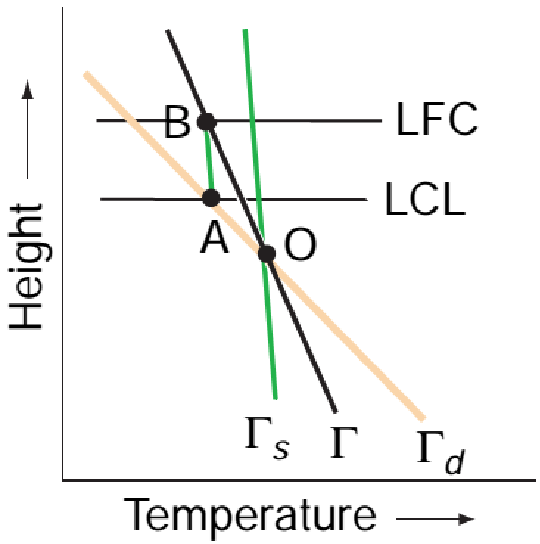


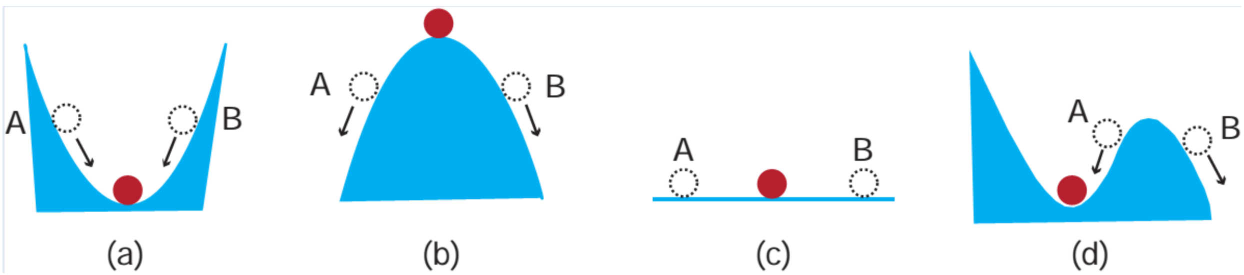
Wallace and Hobbs, 2006

# Conditional and Convective Instability

**Conditionally unstable:**

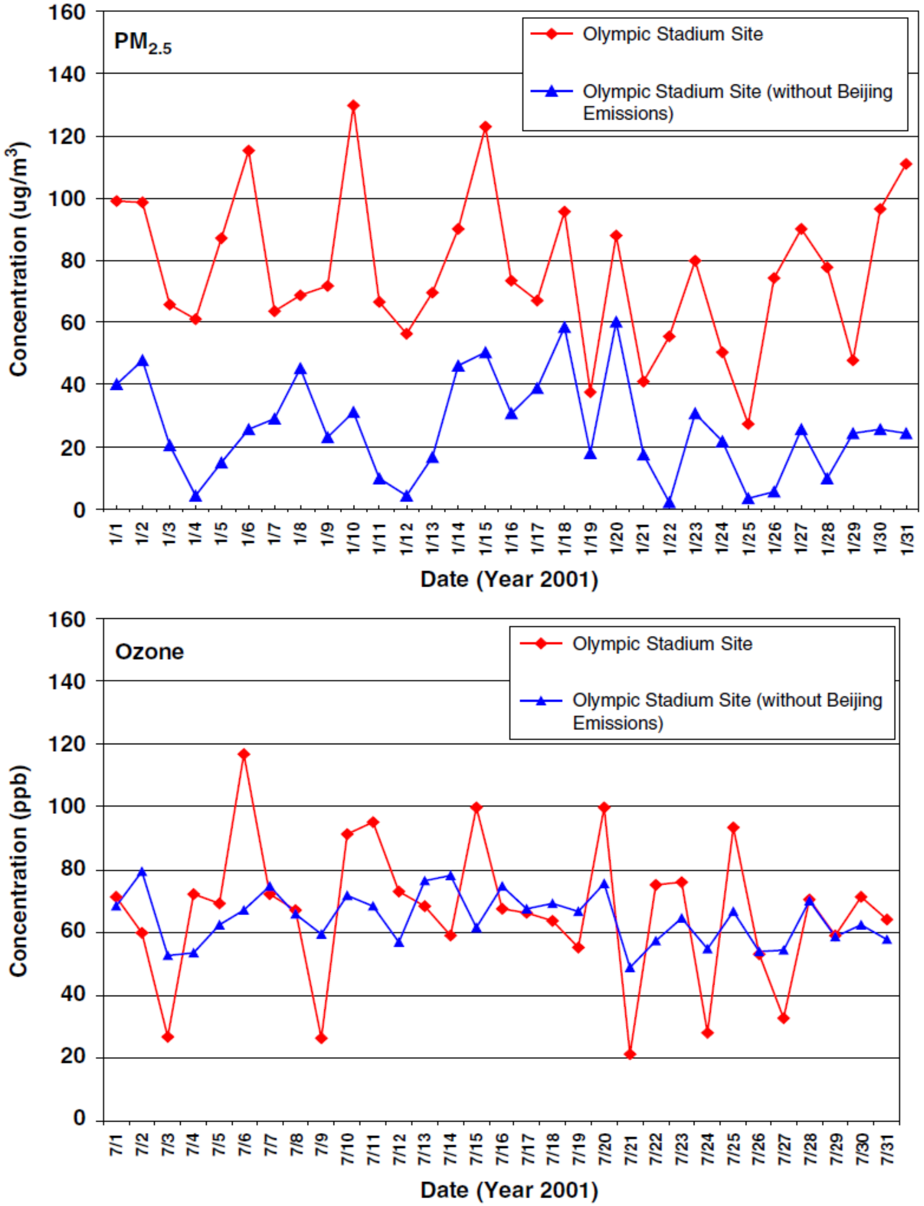
$\Gamma_s < \Gamma < \Gamma_d$



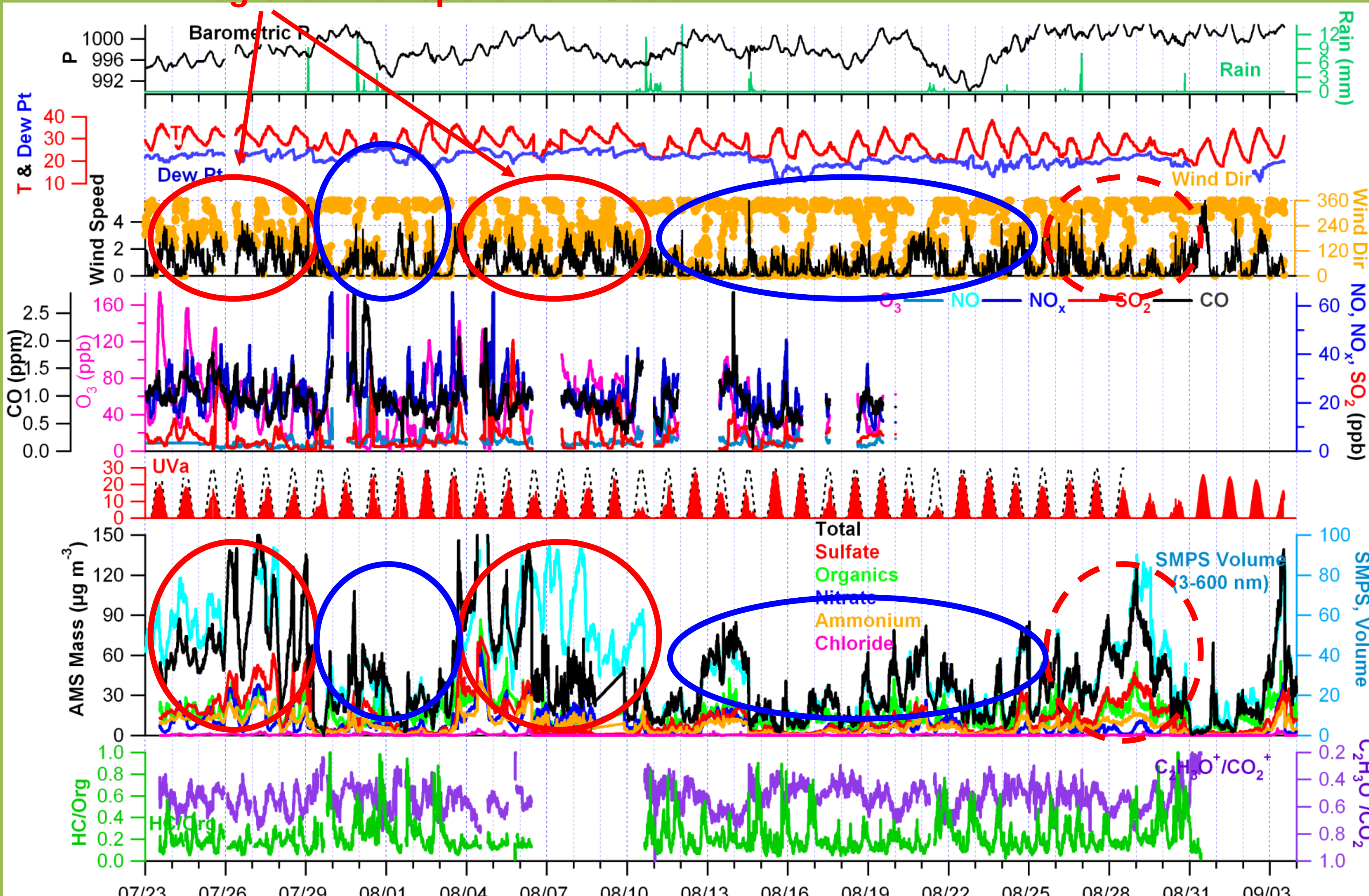




Stable Unstable Neutral Con. unstable

# Regional Transport and Transformation Affecting Beijing



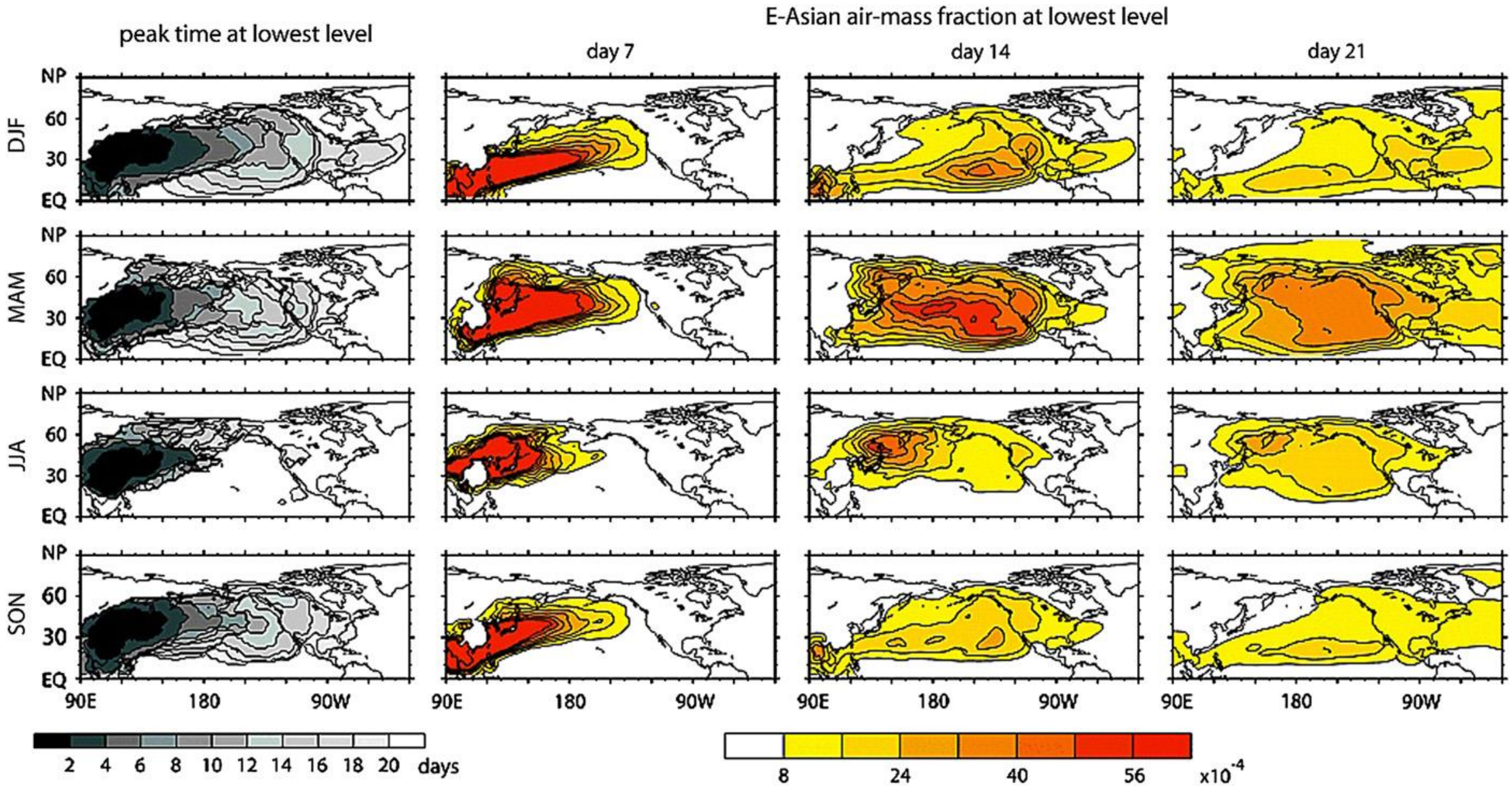
# Regional Transport from South




D. Worsnap, Q. Zhang, LY He

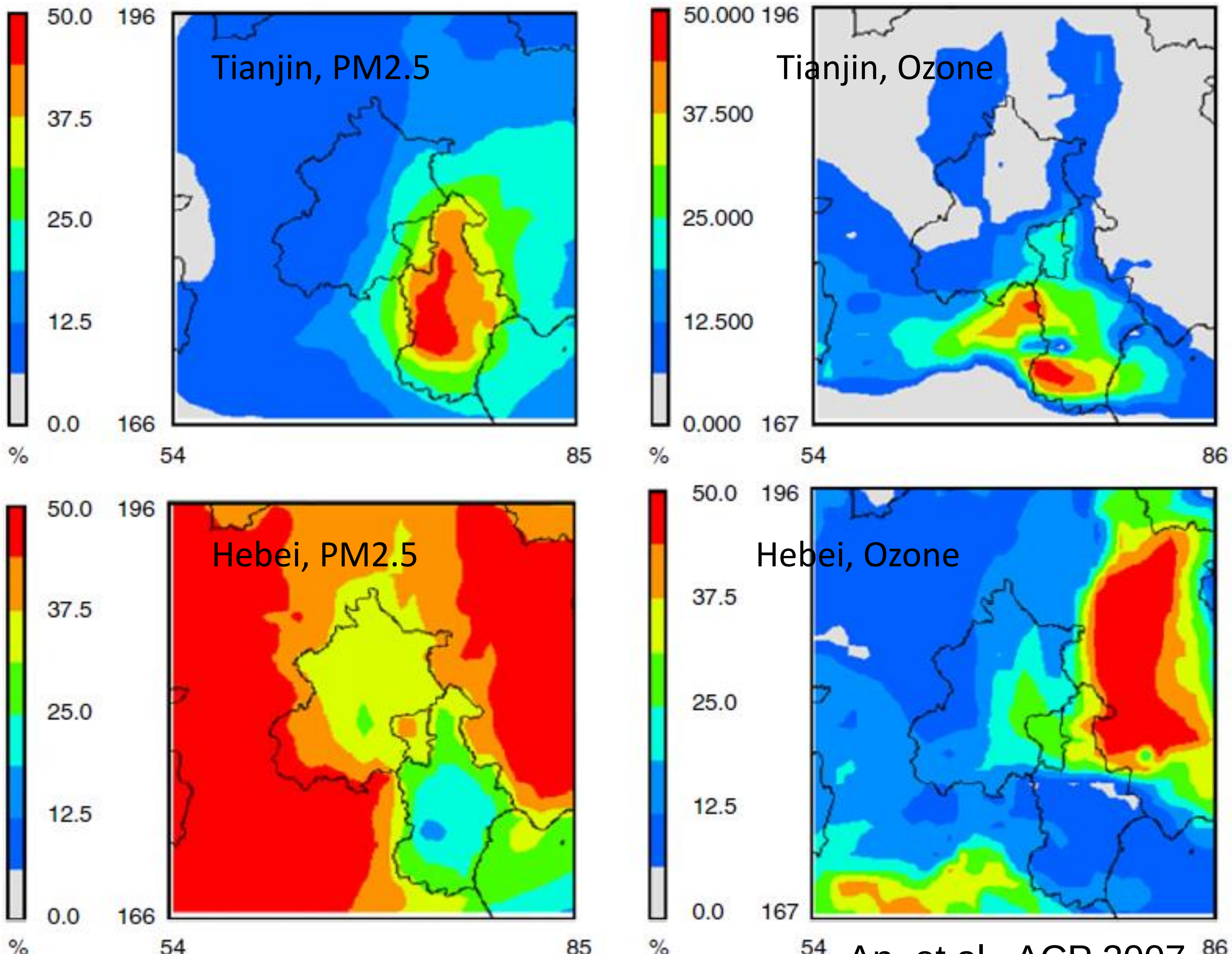
# East Asian Influence: Analysis by CTMs

Ozone




**Liu et al., 2009**

# Regional Transport and Transformation Affecting Beijing

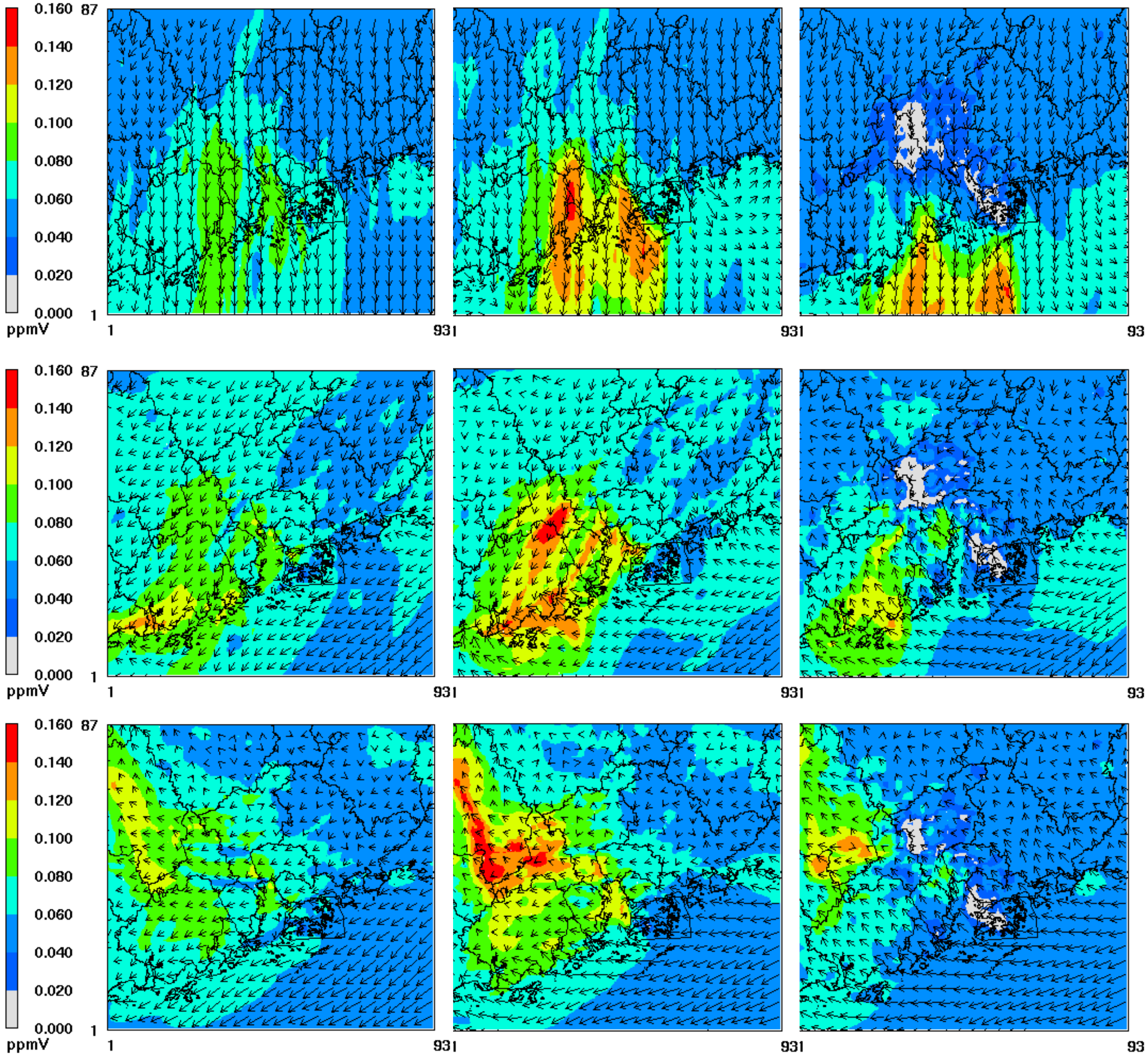



An et al., ACP 2007

# Three Evolution Patterns of Surface $O_3$ in PRD

**Influenced by synoptic weather conditions and sea-land breezes circulations, three evolution patterns of surface $O_3$ over PRD during Oct 4-31, 2004 are summarized.**

**12:00 15:00 19:00 (LST)**




## Oct 19, 2004

- ***Northerly/Northeasterly winds prevailing in the whole day***
- ***Elevated $O_3$ distributing over southern/southweatern PRD, then moved to southern water area***
- ***7 out of 28 days, 25%***

## Oct 16, 2004

- ***Weak northeasterly winds dominant in the daytime, southeasterly sea breeze developed along the coastal areas from afternoon to midnight***
- ***Elevated $O_3$ distributing over southweatern inland & coastal PRD***
- ***13 out of 28 days, ~50%***

## Oct 29, 2004

- ***Weak easterly winds dominant in daytime, southeasterly sea breeze developed from afternoon to midnight***
- ***Elevated $O_3$ distributing over western inland PRD& PRE***
- ***8 out of 28 days, ~25%***

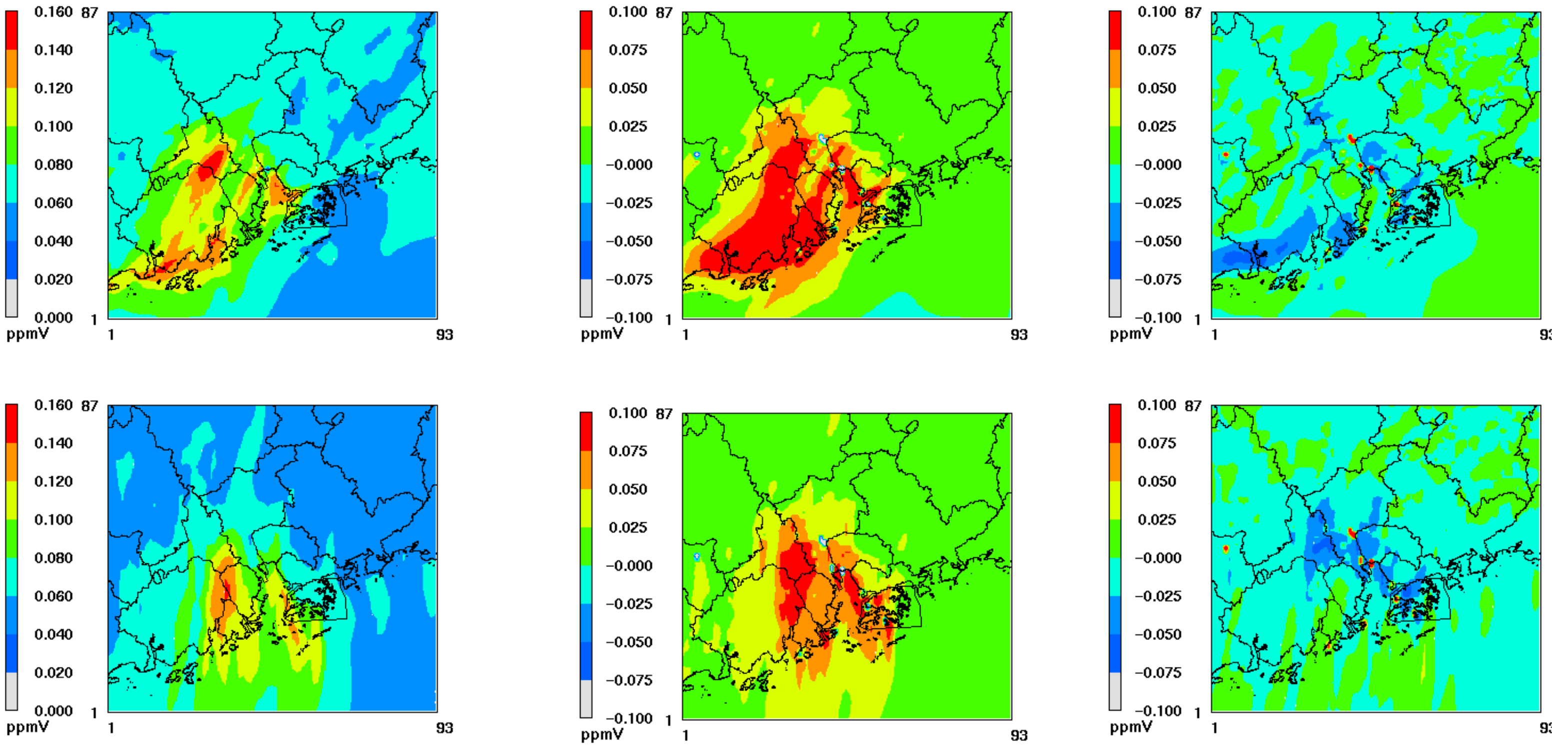
# Chemical Process Dominates the Daytime Build-up of Near-Surface (below 1km) $O_3$ over PRD

**Accumulated contributions to $O_3$ from 9:00 to 15:00 LST**

| Average $O_3$ concentration at 15:00 LST | Chemical production | Transport |
|---|---|---|

Oct 16

Oct 19

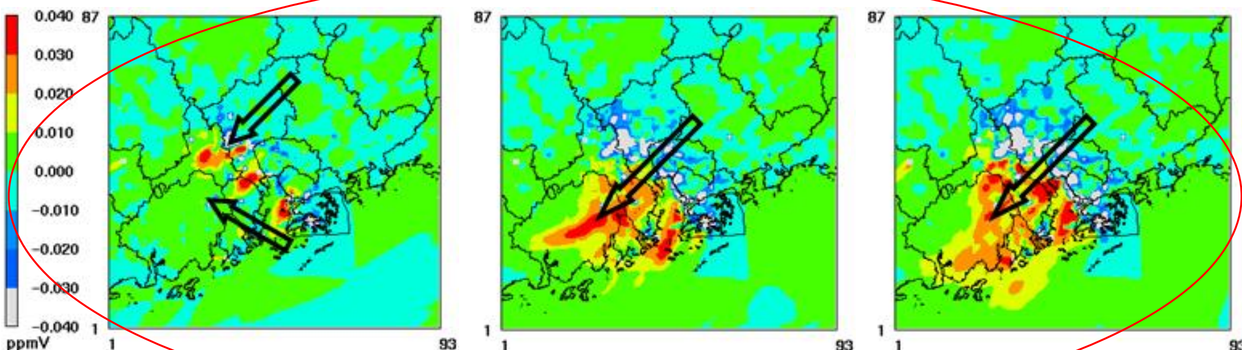



XS. Wang et al., Submitted to ACPD

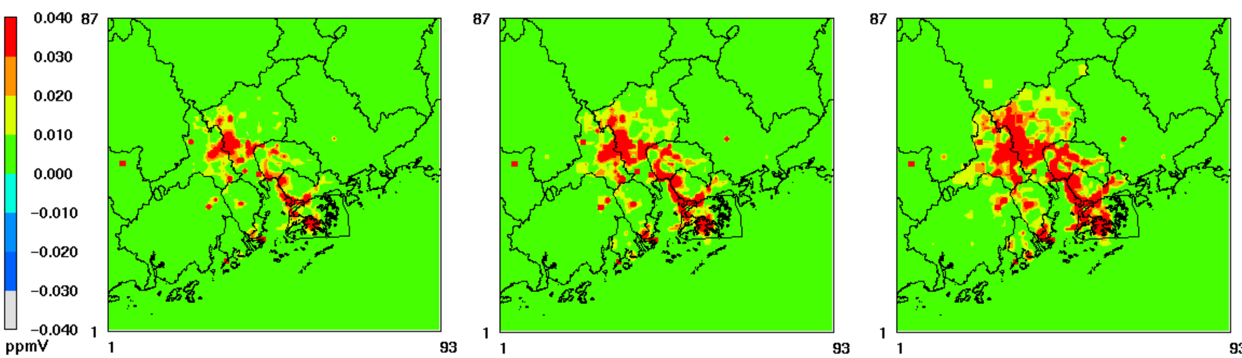
# Transport Process Redistributes Precursors from Night to Next Morning to Help Form Regional Daytime $O_3$

NOx from different source regions mixed and transported to downwind rural areas

| *from 17:00 Oct 15 to 00:00 Oct 16* | *from 17:00 Oct 15 to 09:00 Oct 16* | *from17:00 Oct 15 to 15:00 Oct 16* |
| --- | --- | --- |

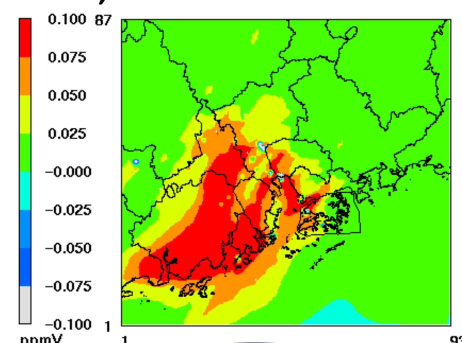



**Accumulated NOx transport over layer 1-7 during Oct 15-16**



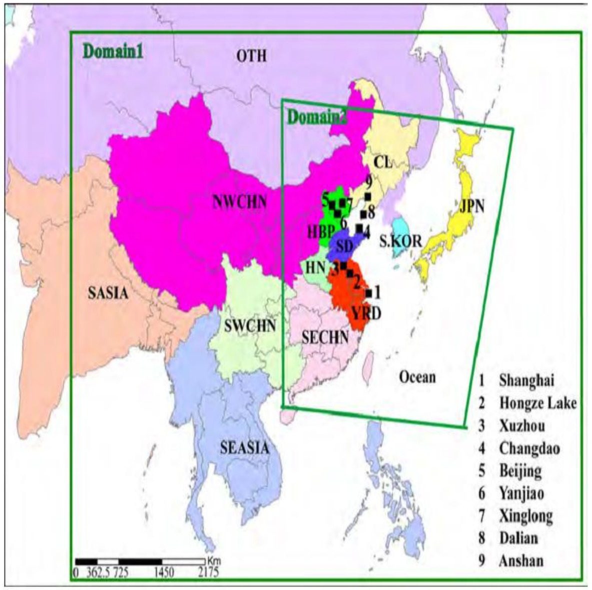

**Accumulated NOx emission over layer 1-7 during Oct 15-16**

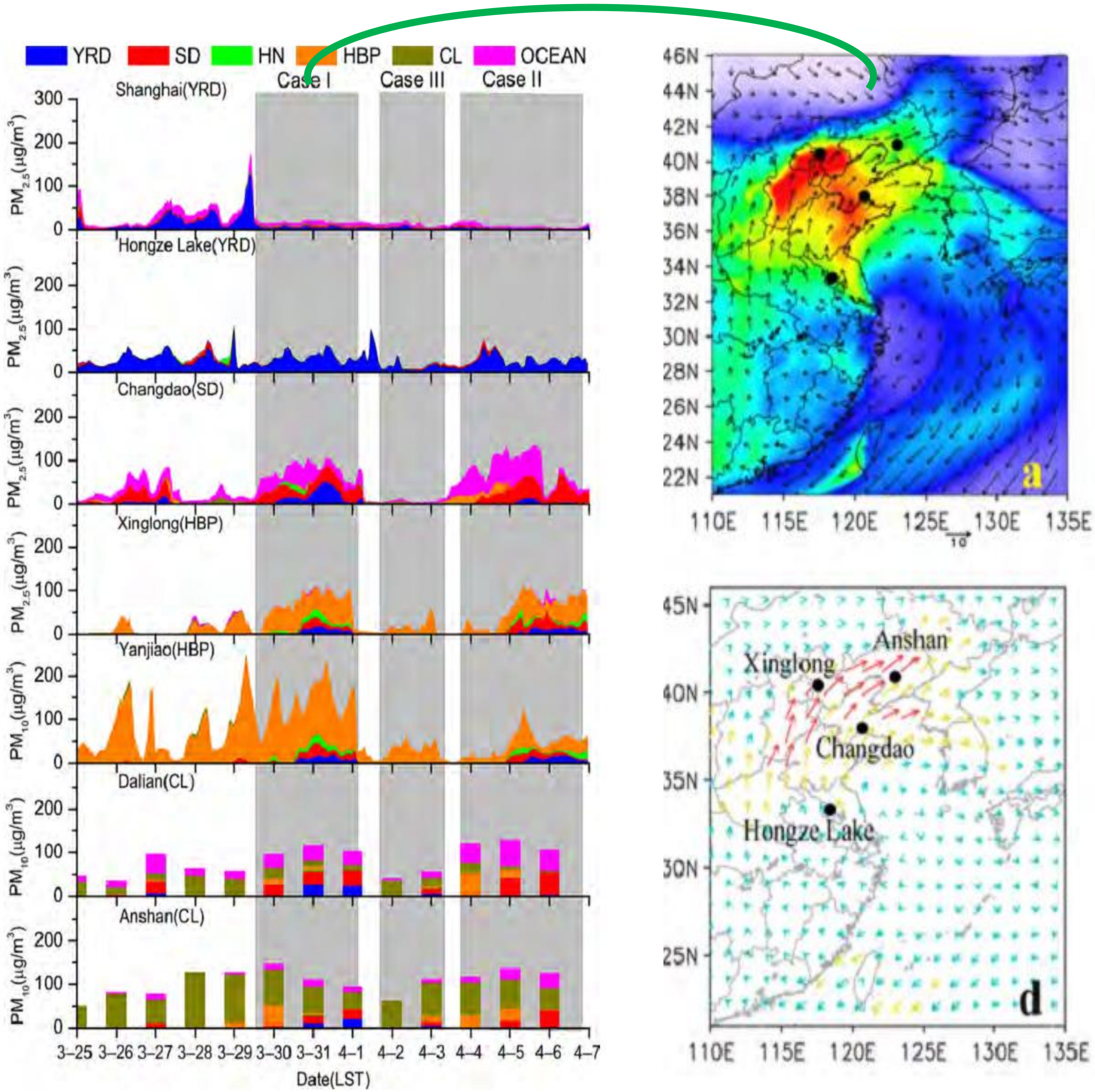
**Accumulated $O_3$ chemical production on 9:00-15:00 LST, Oct 16**




Redistributed NOx involved in the daytime $O_3$ chemical formation over south and west of PRD

# Transport of PM Between China's City Clusters



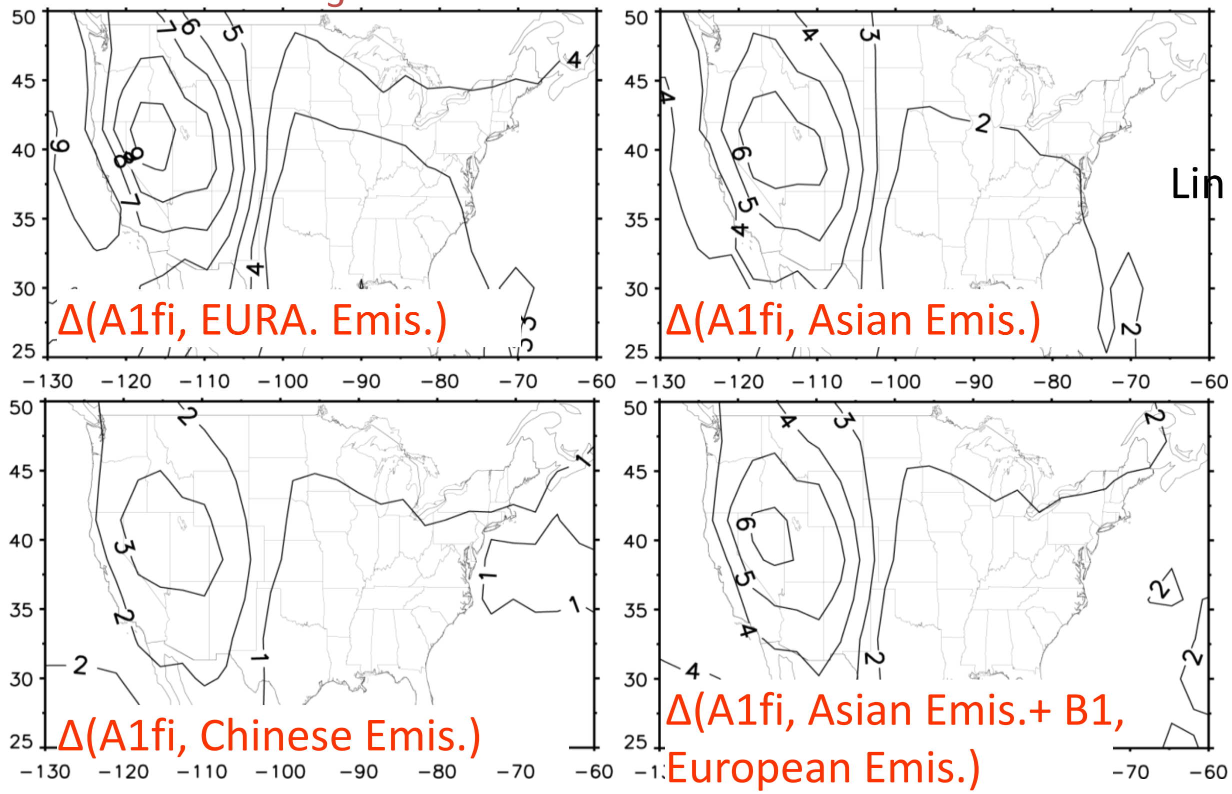

**Spring 2011**

**NAQPMS + tagging**

**Li et al., 2013, Tellus B**

# Effects of Δ(European/Asian Emissions)

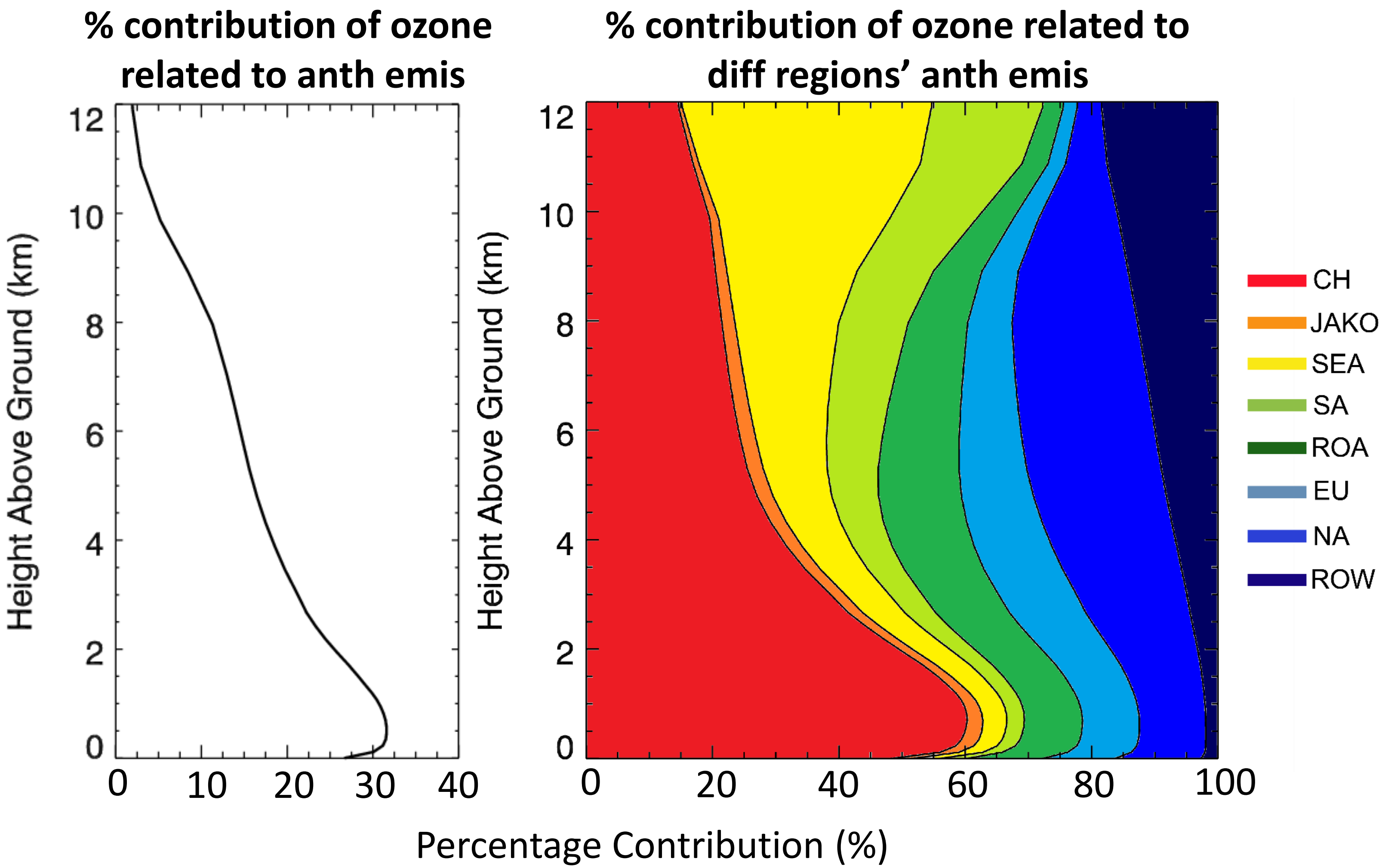
$\Delta O_3$ in JJA from 1999 to 2049




Lin et al., 2008, GRL

- ➢Changes in anthropogenic emissions over Europe and Asia, especially Asia, have large impacts on the long-range transport under A1fi. The effects are minor under B1.
- ➢Increases in biogenic emissions over Europe and Asia result in U.S. ozone enhancements of less than 2 ppb, with greater effects under A1fi than B1.
- ➢Climate warming reduces the long-range transport by < 1 ppb.

# Foreign Anthropogenic Emissions Contribute Significantly to Springtime Total Anthropogenic Ozone over China!

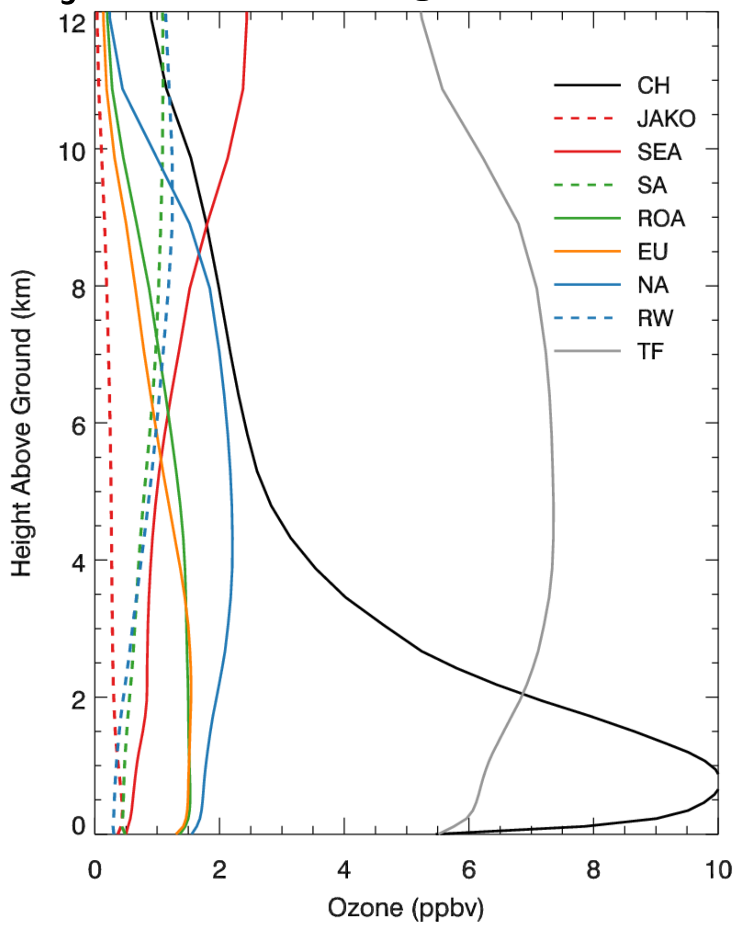



- Anthro emis contribute ~30% in PBL and the contribution decreases with height
- Foreign anthro emis contribute 40~80% to total anthro ozone and the contribution increases with height

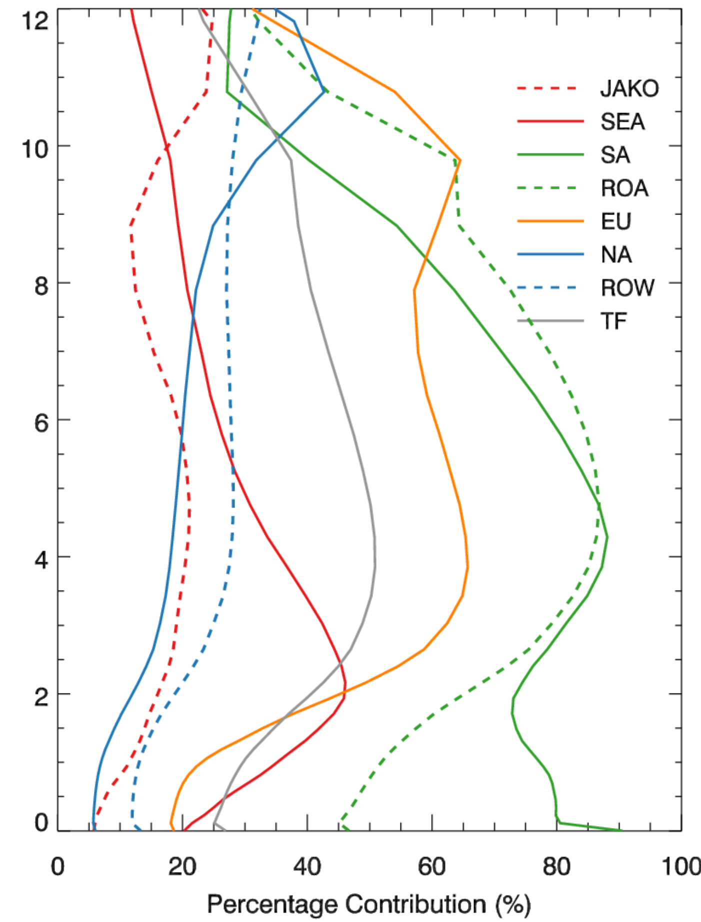
# Vertical Profile of $O_3$ Related to Anthro Emis over China

| EMIS | EU | NA |
|---|---|---|
| NOx (Tg N) | 1.24 | 1.27 |
| CO (Tg) | 12.5 | 17.8 |
| NMVOCs (Tg C) | 1.14 | 2.06 |

$O_3$ related to diff regions' anth emis



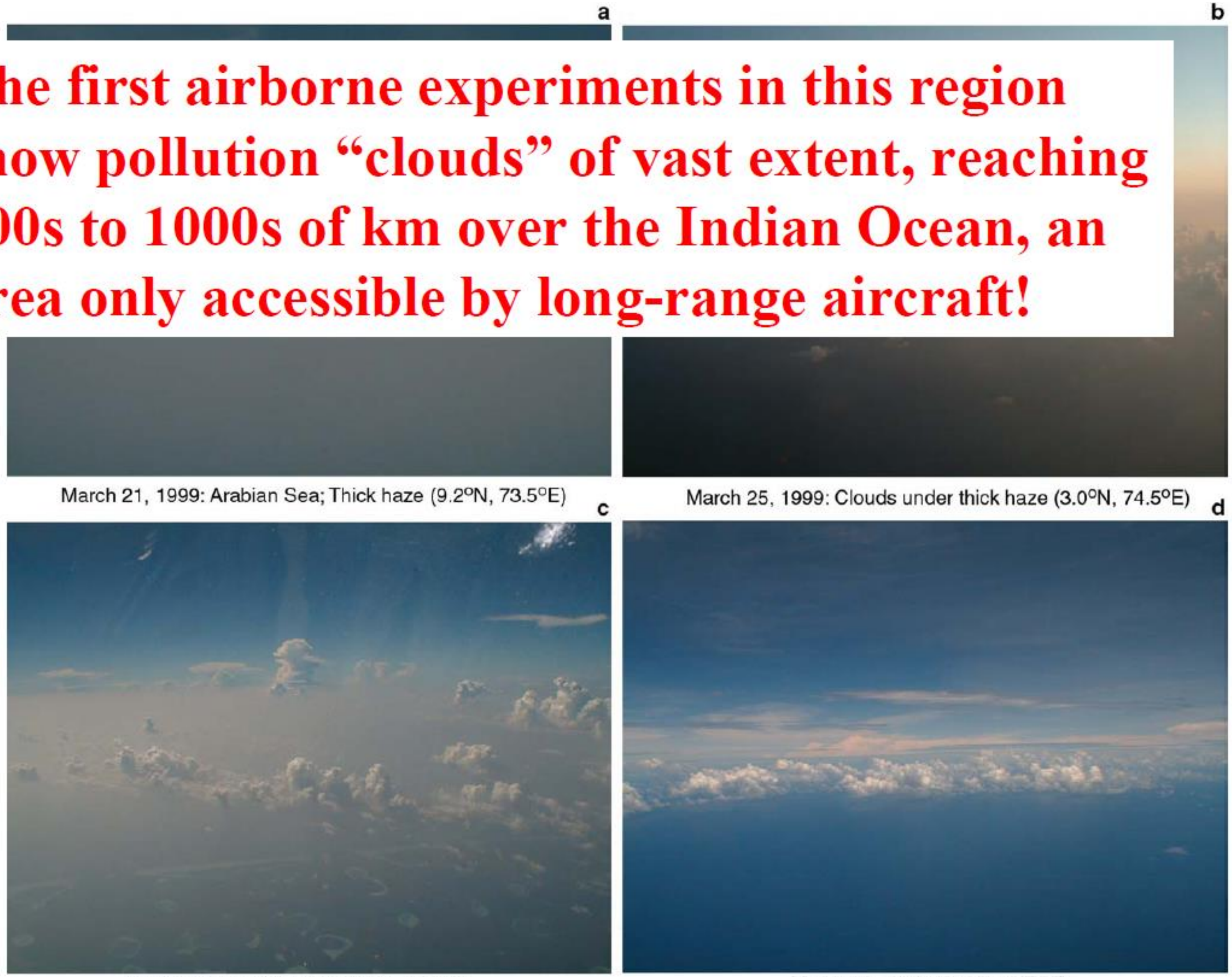

% contribution of ozone produced directly over source region




- Above 2km, foreign anth emis contributes more ozone than domestic emis
- Large portion of ozone is produced along the transport pathway, rather than produced over source regions

# Long-range Transport of Asian PM to the Tropics

**The first airborne experiments in this region show pollution "clouds" of vast extent, reaching 100s to 1000s of km over the Indian Ocean, an area only accessible by long-range aircraft!**




a: March 21, 1999: Arabian Sea; Thick haze (9.2ºN, 73.5ºE)

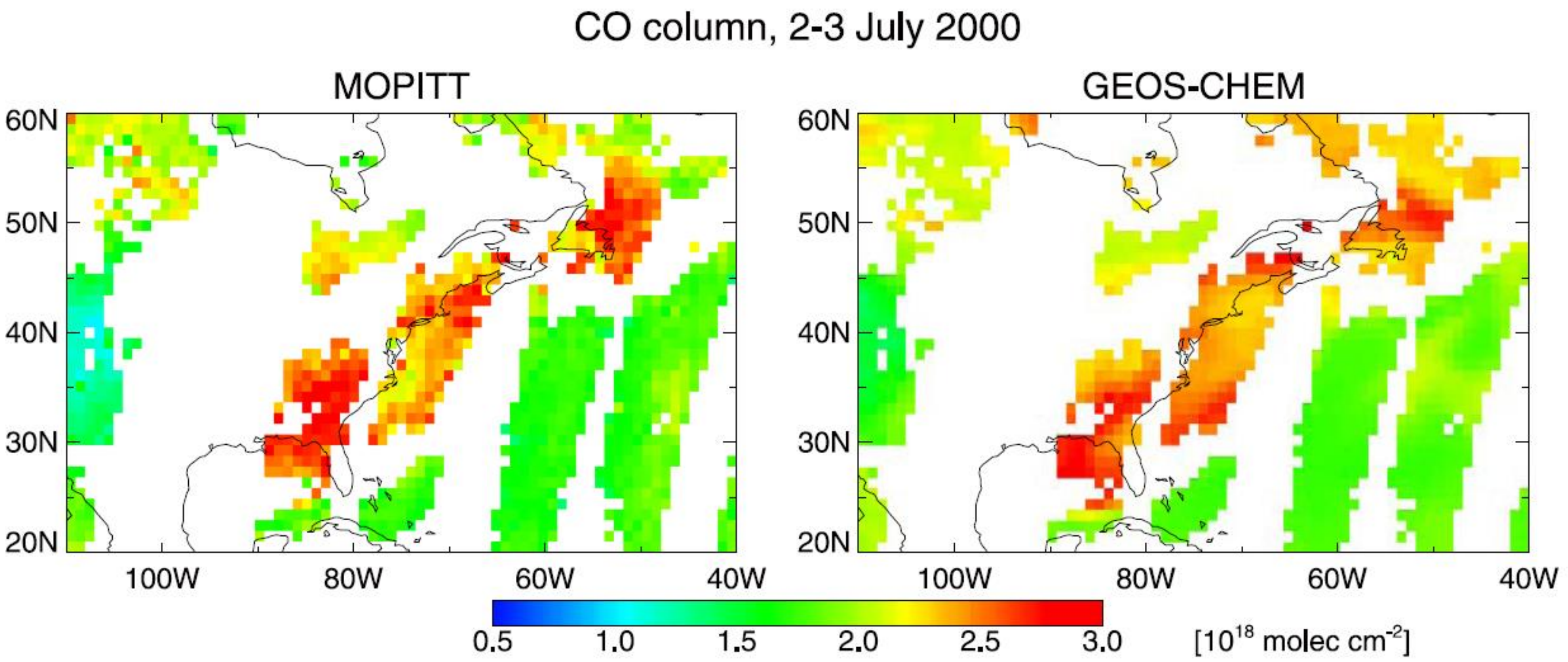
b: March 25, 1999: Clouds under thick haze (3.0ºN, 74.5ºE)

c: February 24, 1999: Just North of ITCZ; Haze extends up to top of Cu (0.5ºN, 73.3ºE)

d: March 24, 1999: South of ITCZ; Almost pristine clouds (7.5ºS, 73.5ºE)

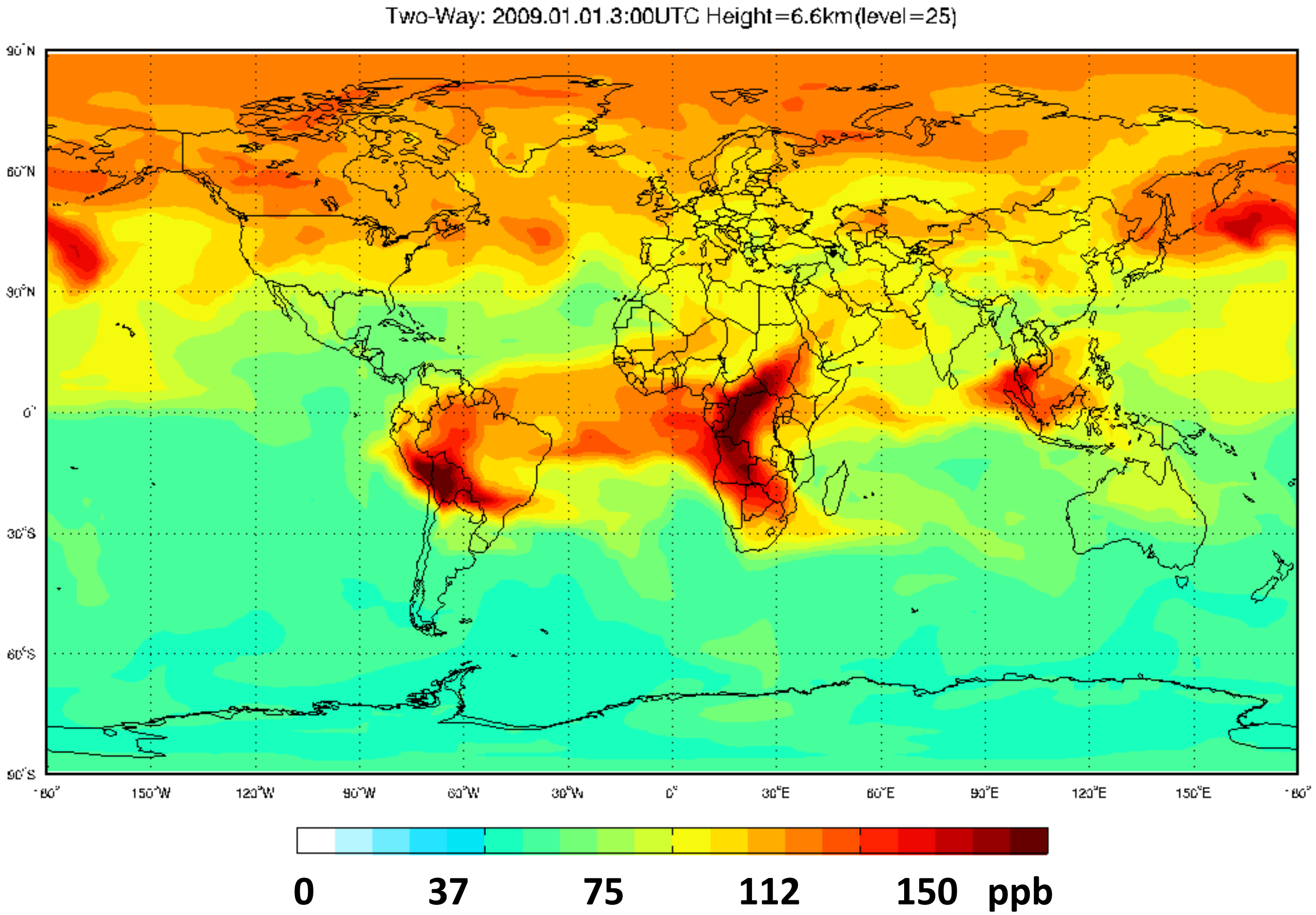
Photo credit: Center for Clouds, Chemistry and Climate; Scripps Institution of Oceanography; University of California, San Diego
Image available at: http://sio.ucsd.edu/supp_groups/siocomm/pressreleases/Indoexagu.html

# North American Outflow of CO



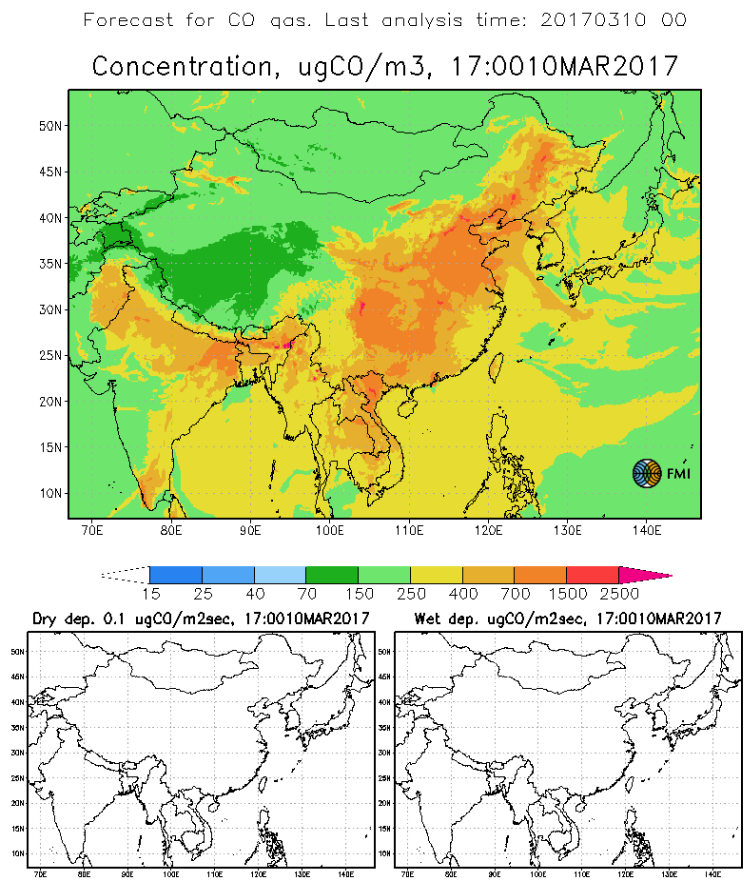
# Global Transport of Carbon Monoxide

## Mid-tropospheric carbon monoxide in Jan 2009




**Yan et al., ACP, 2014**

# Regional Transport of Carbon Monoxide




**SILAM Model Simulation**

# Characteristic Distance of Transport

- **Primary Pollutant:**

  $$D = U \times \tau = \text{Wind Speed} \times \text{Lifetime}$$

- **Secondary Pollutant:**

  $$D = U \times \tau^{*} \approx U \times [\, \tau_p * \tau_s \,] \,/\, [\, \tau_p - r * \tau_s \,]$$

  $$r = C_{primary} \,/\, C_{secondary}$$

  $$\text{when } C_{primary}/\tau_p < c_{secondary}/\tau_s$$

Characteristic Time

# Characteristic Distance of Transport

- **Primary Pollutant:**

  $$D = U \times \tau = \text{Wind Speed} \times \text{Lifetime}$$

- **Secondary Pollutant:**

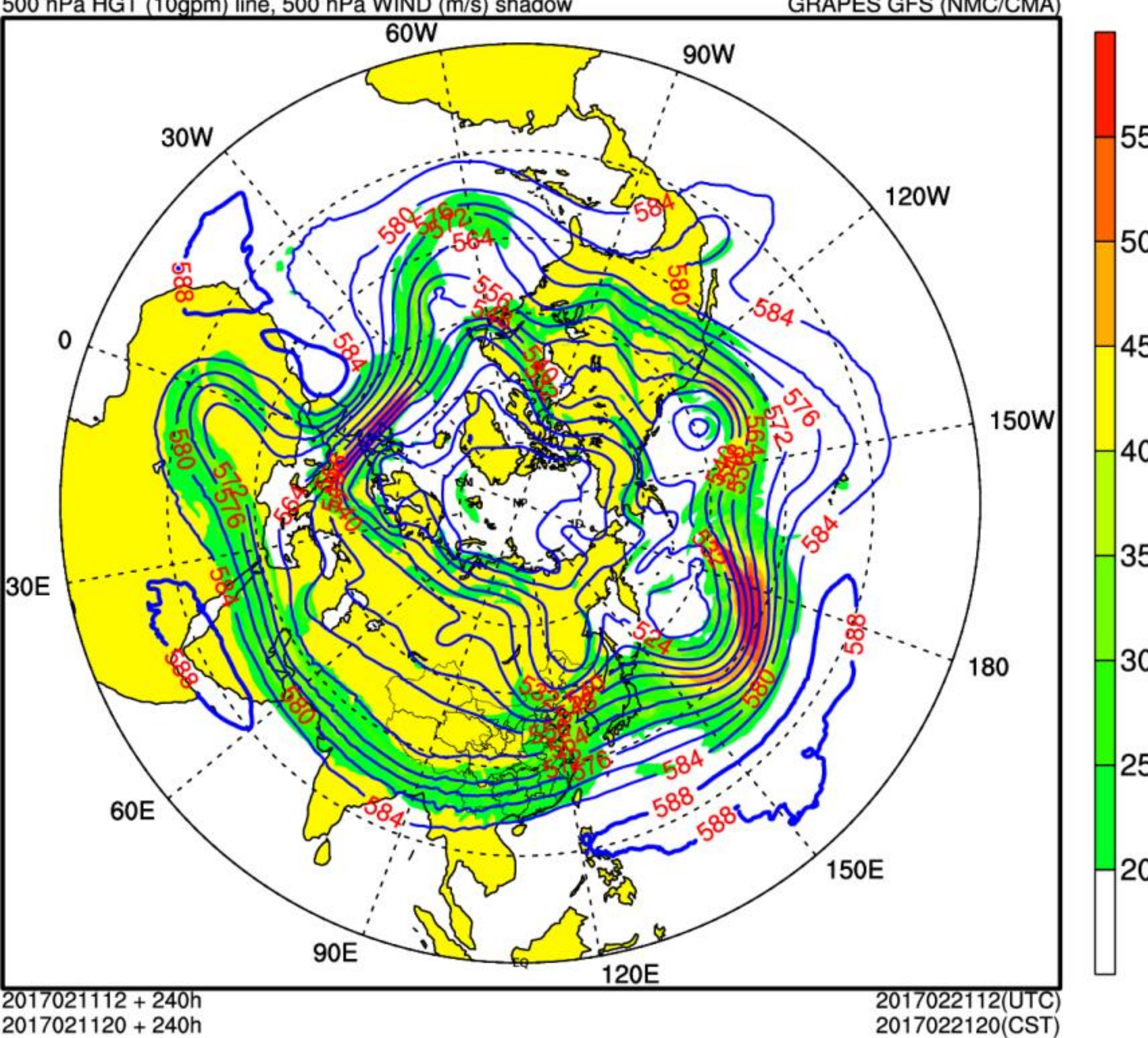
  $$D = U \times \tau^{*} = U \times \tau_s * [\,1 + r\,], \quad \text{where } r = C_p / C_s$$

  $$\approx U \times \tau_p \quad \text{when} \quad C_p / \tau_p << C_s / \tau_s$$

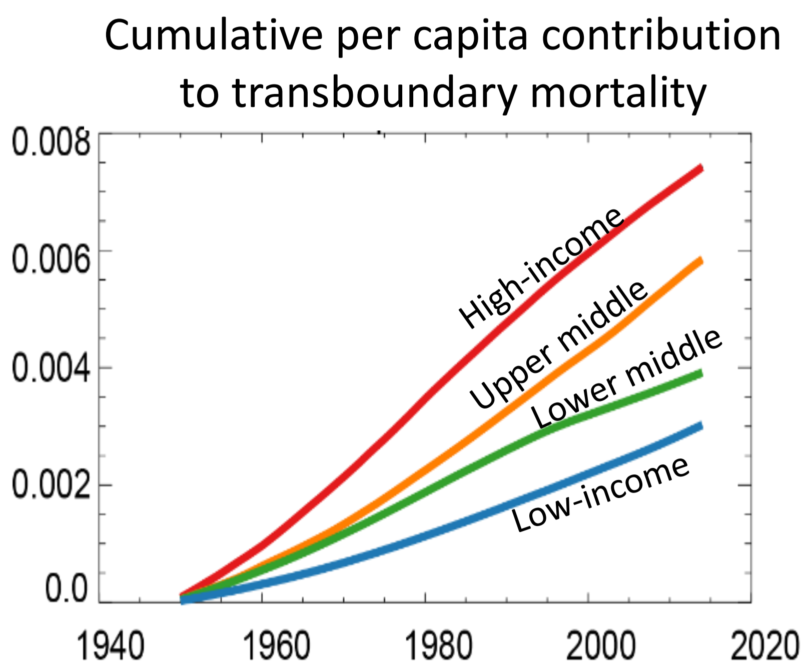
  $$= U \times [\,\tau_p + \tau_s\,] \quad \text{when} \quad C_p / \tau_p = C_s / \tau_s$$

  $$\approx U \times \tau_s \quad \text{when} \quad C_p / \tau_p >> C_s / \tau_s$$

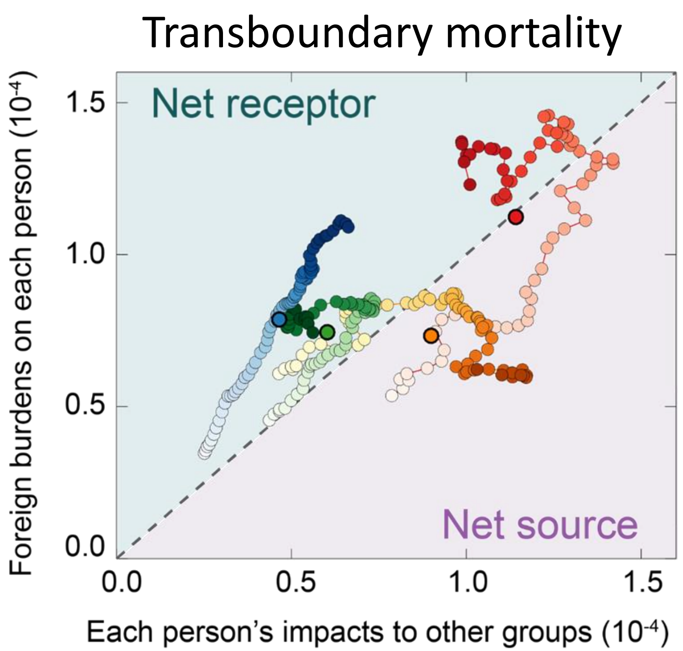
Characteristic Time


500 hPa HGT (10gpm) line, 500 hPa WIND (m/s) shadow
GRAPES GFS (NMC/CMA)
60W
90W
120W
150W
180
150E
120E
90E
60E
30E
0
30W
55
50
45
40
35
30
25
20
2017021112 + 240h
2017021120 + 240h
2017022112(UTC)
2017022120(CST)

# Transboundary $PM_{2.5}$ Mortality Via Atmospheric Transport



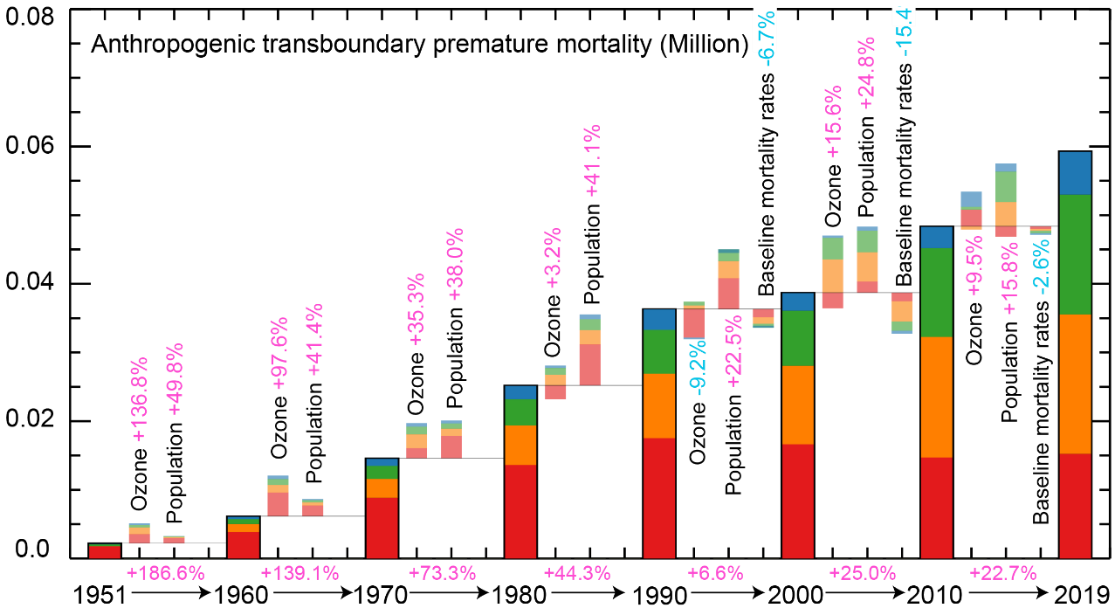

On a per capita contribution basis:
**Richer group exerts larger cumulative transboundary mortality**

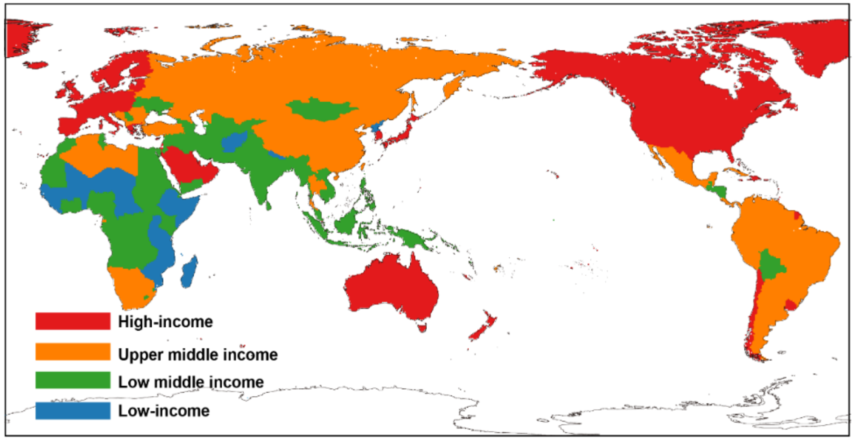



On a per capita *net effect* basis:
**Poorest suffers from heaviest *net* transboundary burden**

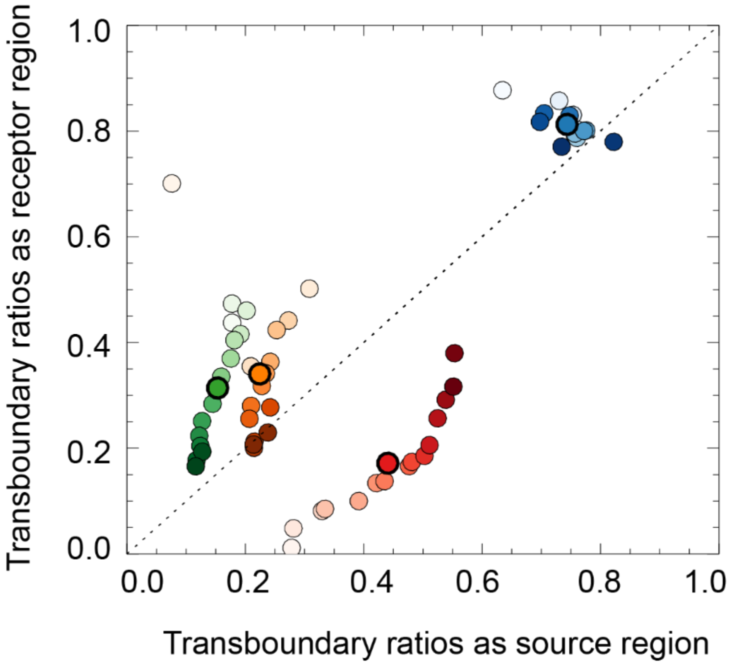
Chen et al., Science Bulletin, 2022

# Historical Transboundary Ozone Mortality via Atmospheric Transport

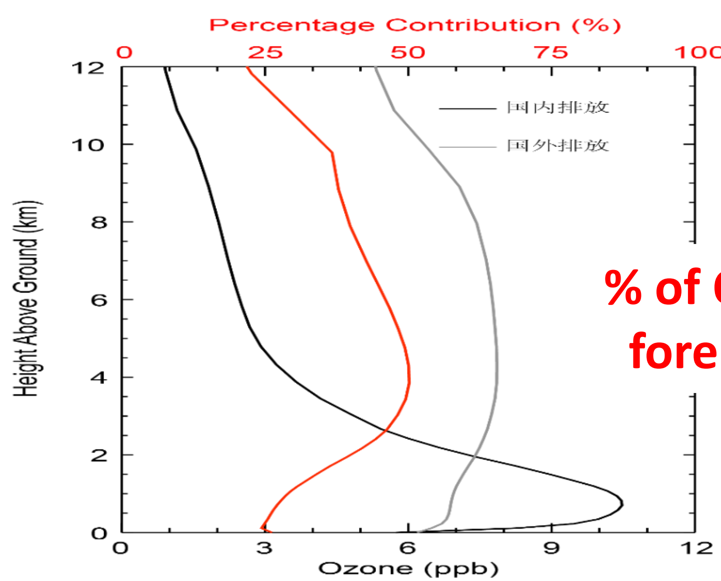



Chen et al., ERL, 2023



# Two Chemistry Mechanisms of Ozone Transport


% of $O_3$ produced within foreign source regions

Ni et al., ACP, 2018