Chapter 5

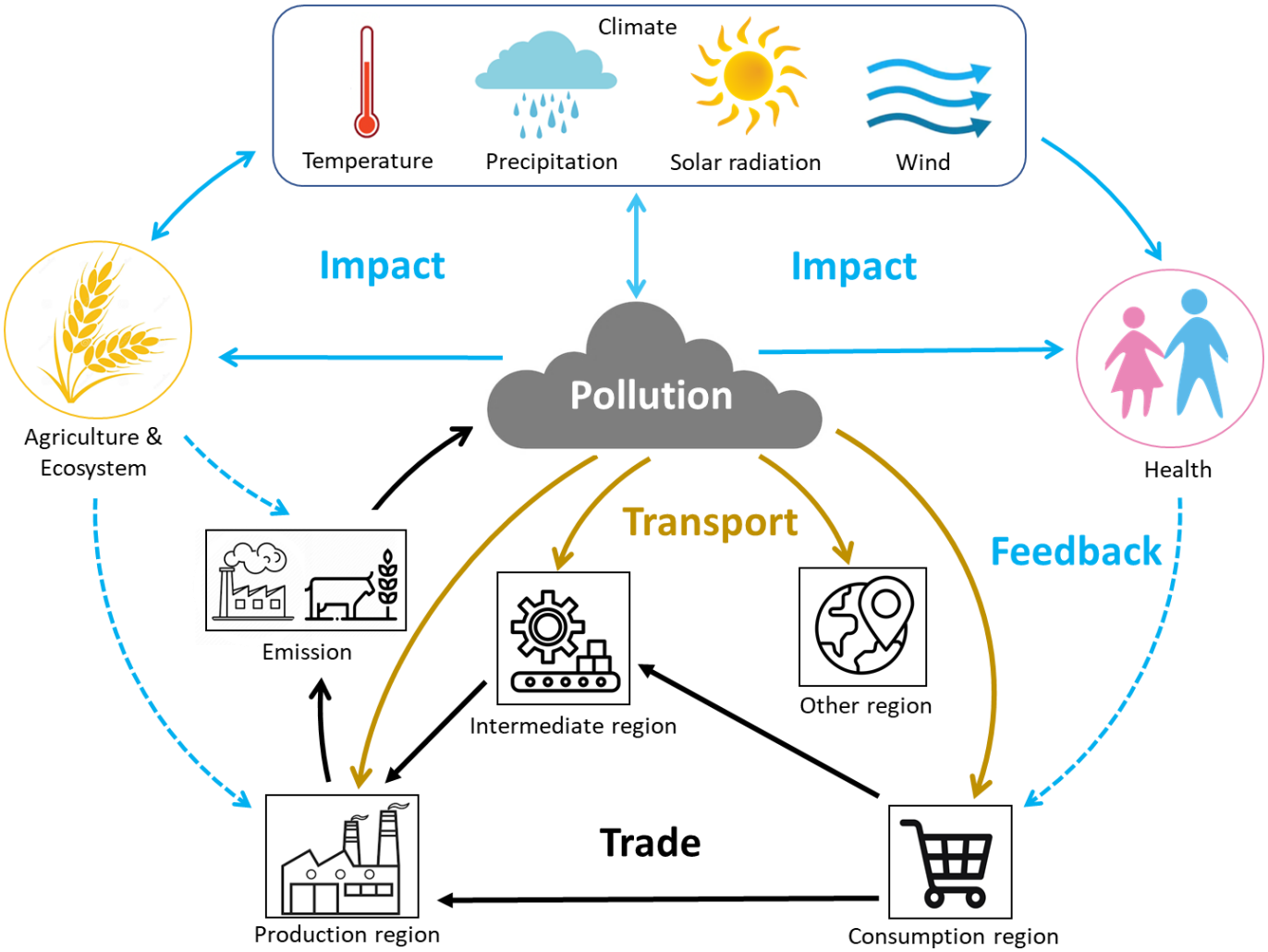
Regional and Global Pollution Transport

北京大学
PEKING UNIVERSITY

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



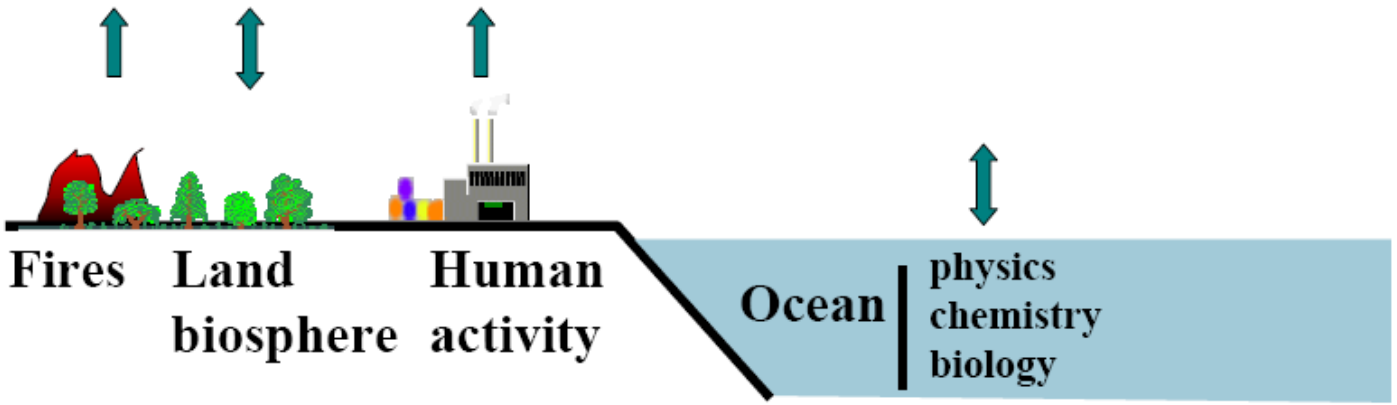
Lin et al., under review

Budget of Air Pollutants

AN ATMOSPHERIC CHEMIST'S VIEW OF THE WORLD



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



Modified from Don Wuebbles' Lecture

Haze: Transported or Locally Formed ?

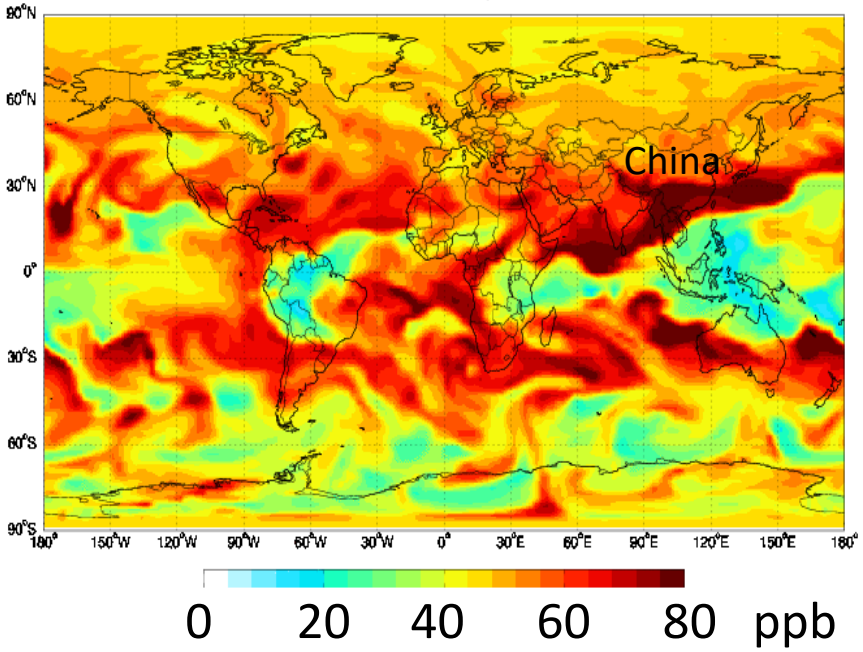


https://v.qq.com/x/page/f03620mzezq.html

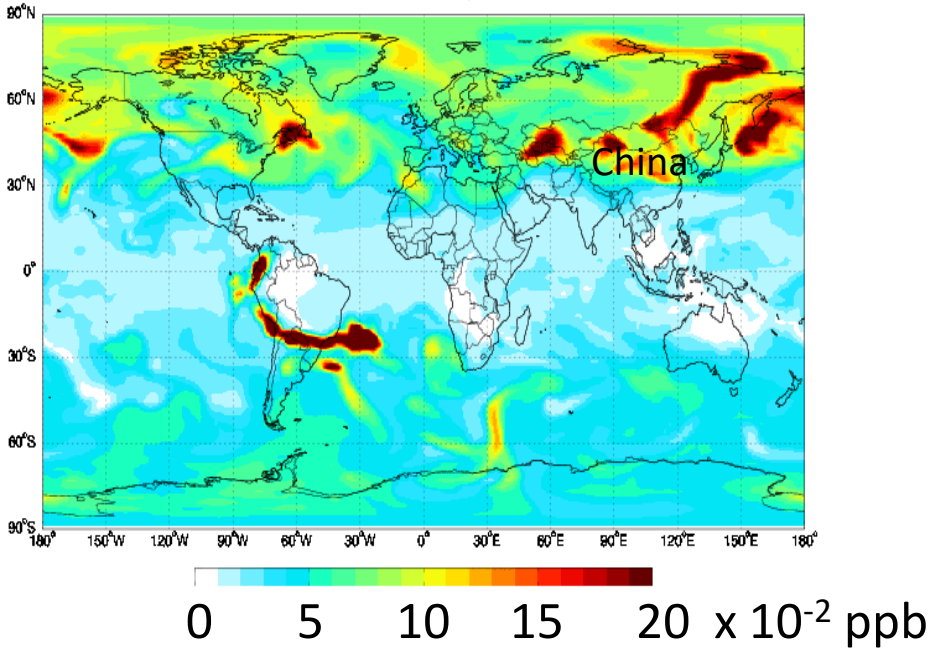
Globalizing Air Pollution: Atmospheric Transport

Simulated by GEOS-Chem Chemical Transport Model

Ozone in Mid-Trop. in Jan 2009



Sulfate in Mid-Trop. in Jan 2009



- **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

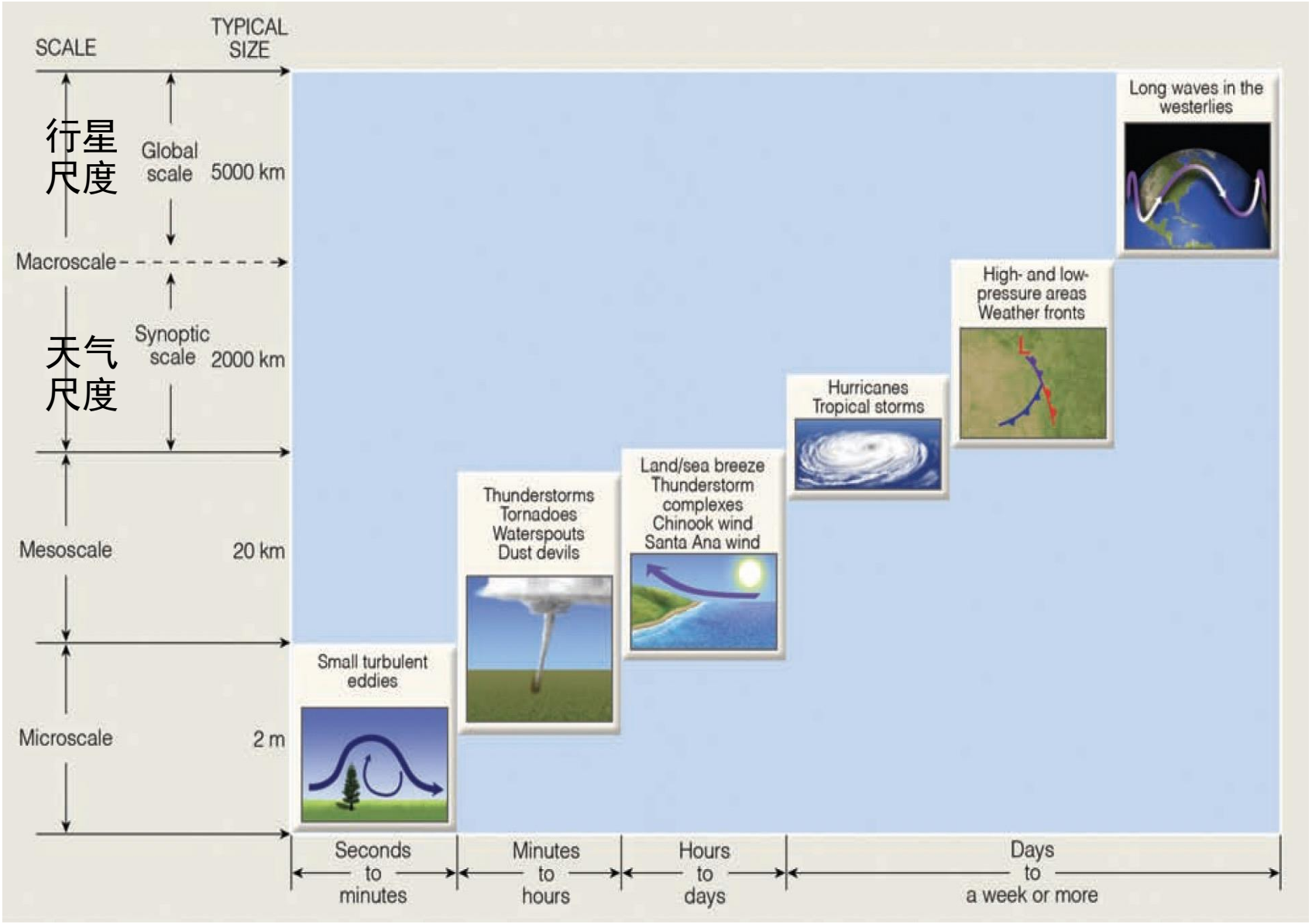
- Local emissions and/or production
- Inter-regional transport and transformation
- Global transport and transformation
- Stratosphere-troposphere exchange

❖ Natural versus anthropogenic sources

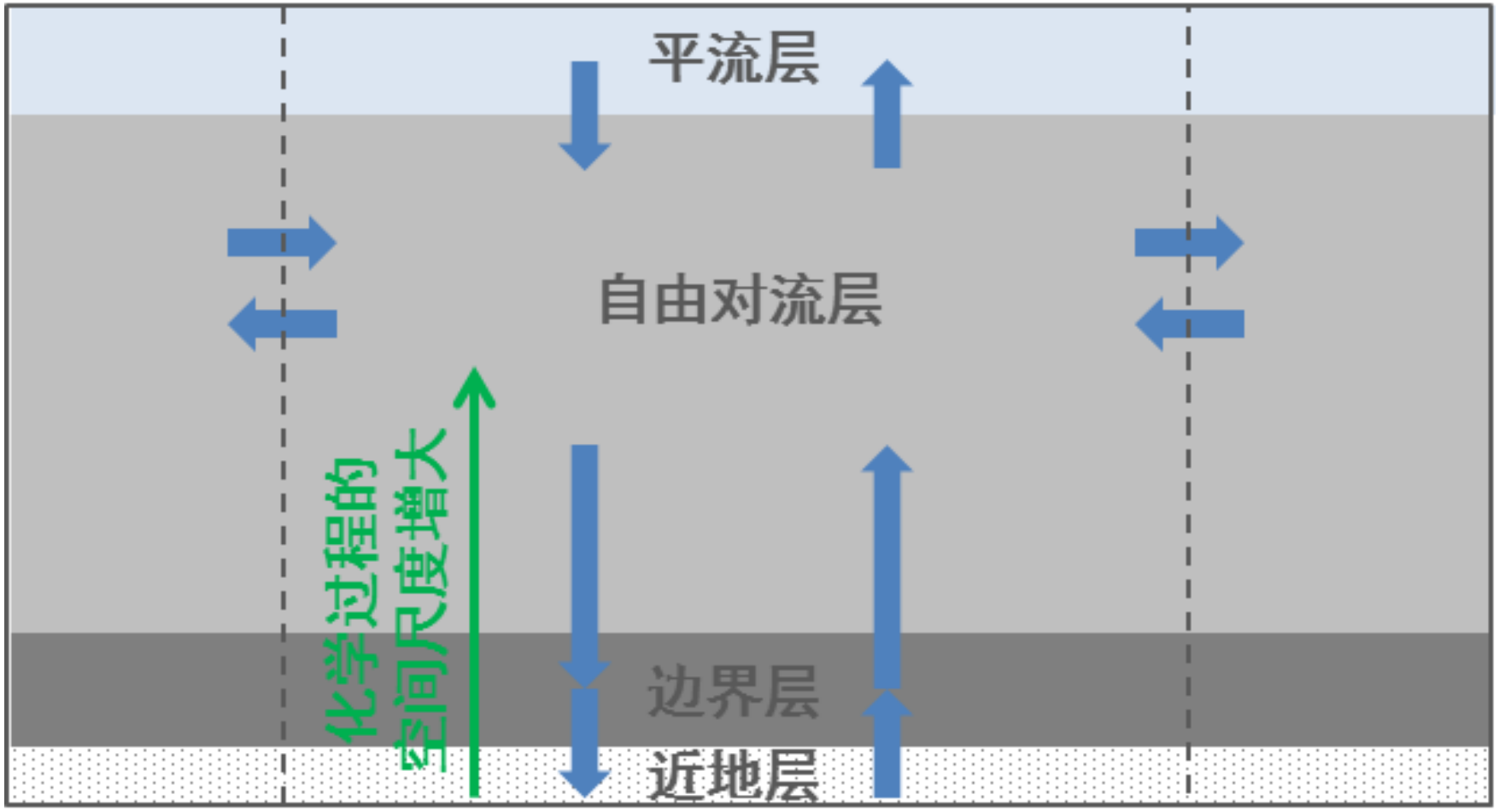
➢ Transport and transformation of air pollutants along the pathway

➢ Lifetime of pollutants is a key factor

Spatiotemporal Scale of Atmospheric Motion (Transport)



Local-Regional-Global Pollution Interconnection



Characteristic Distance of Transport

- **Primary Pollutant:**

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

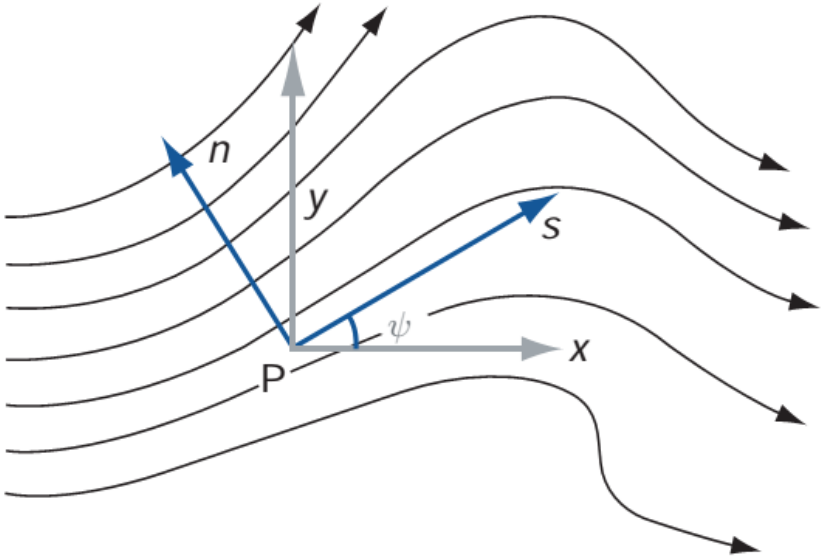
- **Secondary Pollutant:**

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

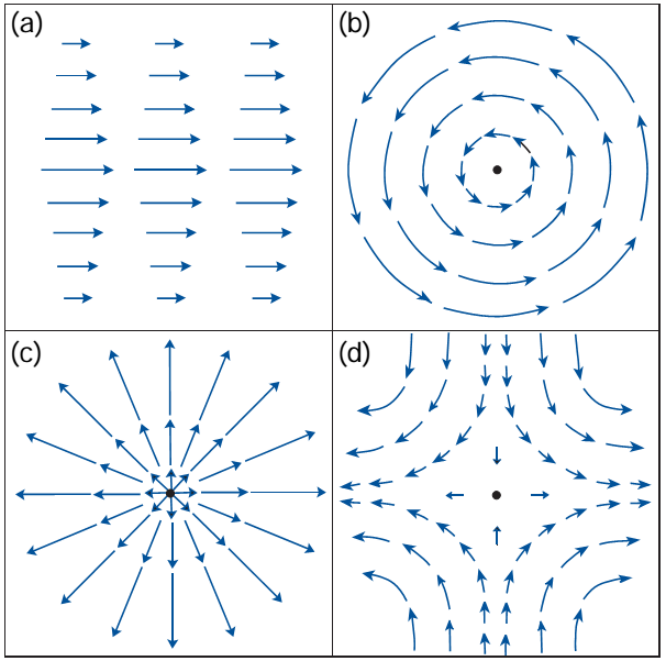
τ^*: Characteristic time
τ_p: Lifetime of primary pollutants in conversion to secondary pollutants
τ_s: Lifetime of secondary pollutants

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

Kinematics: Different Types of Flow Motions



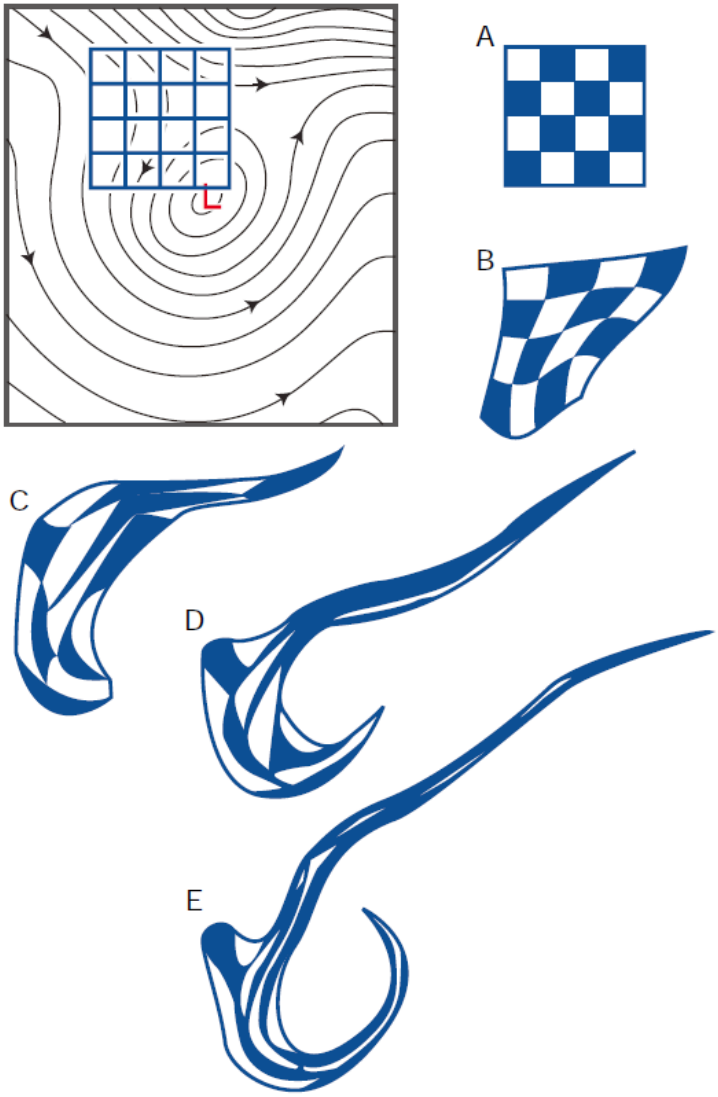
		Vectorial	Natural coords.	Cartesian coords.
Shear	切变		$-\frac{\partial V}{\partial n}$	
Curvature	曲率		$V\frac{\partial \psi}{\partial s}$	
Diffluence	分流		$V\frac{\partial \psi}{\partial n}$	
Stretching	拉伸		$\frac{\partial V}{\partial s}$	
Vorticity ζ	涡度	$\mathbf{k}\cdot\nabla\times\mathbf{v}$	$V\frac{\partial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



	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.

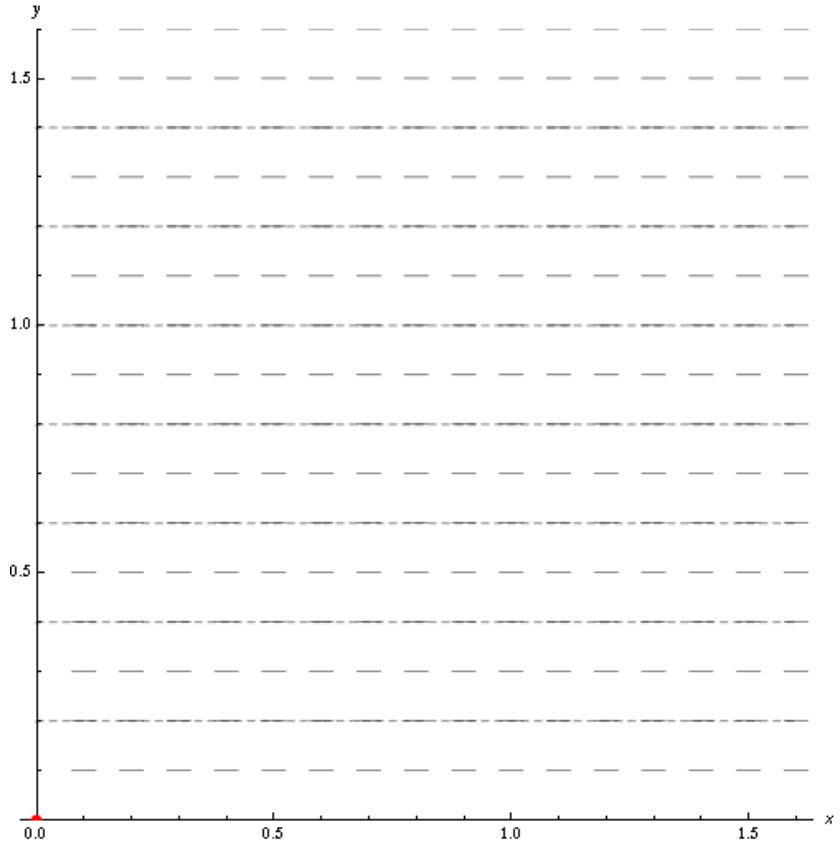
Wallace and Hobbs, 2006

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

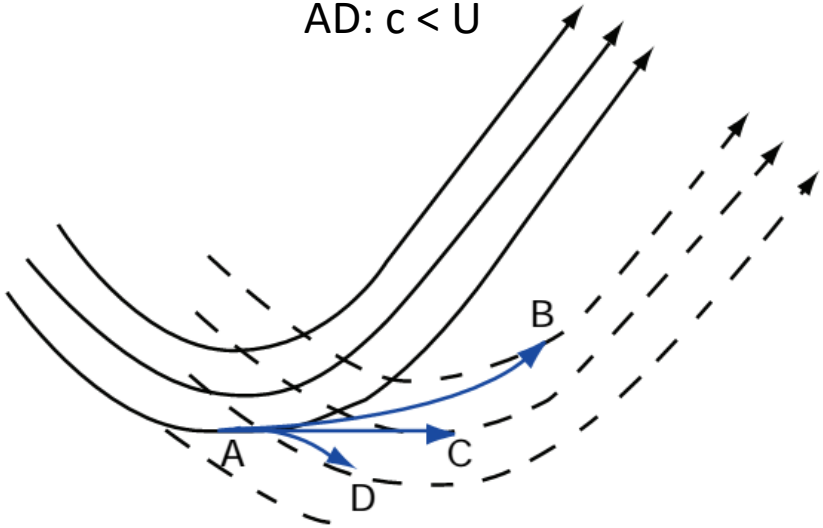
Phase speed of wave: c

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

τ = 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 $\boldsymbol{C} = -f\boldsymbol{k} \times \boldsymbol{V}$ $\qquad f = 2\omega_{earth} \sin\theta$

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:

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

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

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

ϕ = geopotential $\qquad d\phi = gdz$

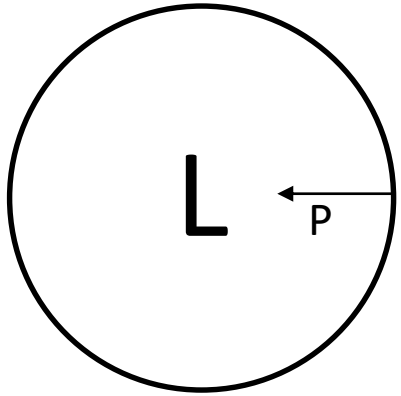
Wallace and Hobbs, 2006

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}

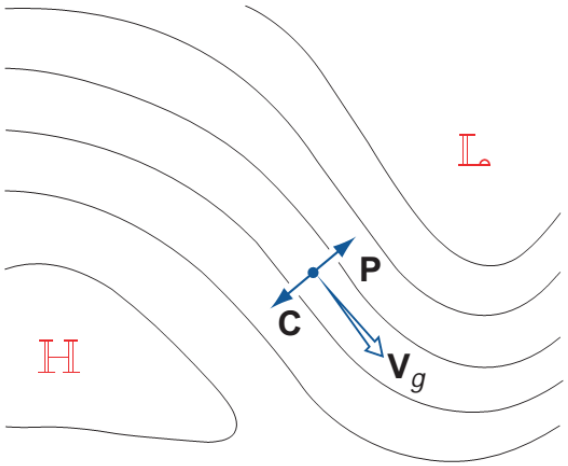
- ➢ The magnitude of Coriolis force ~ 10^{-3} m s^{-2}
- ➢ The magnitude of centripetal force ~ 3 m s^{-2}

Geostrophic Wind (large-scale, without friction)

$$\frac{d\boldsymbol{V}}{dt} =$$ 10^{-4} m s^{-2} 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

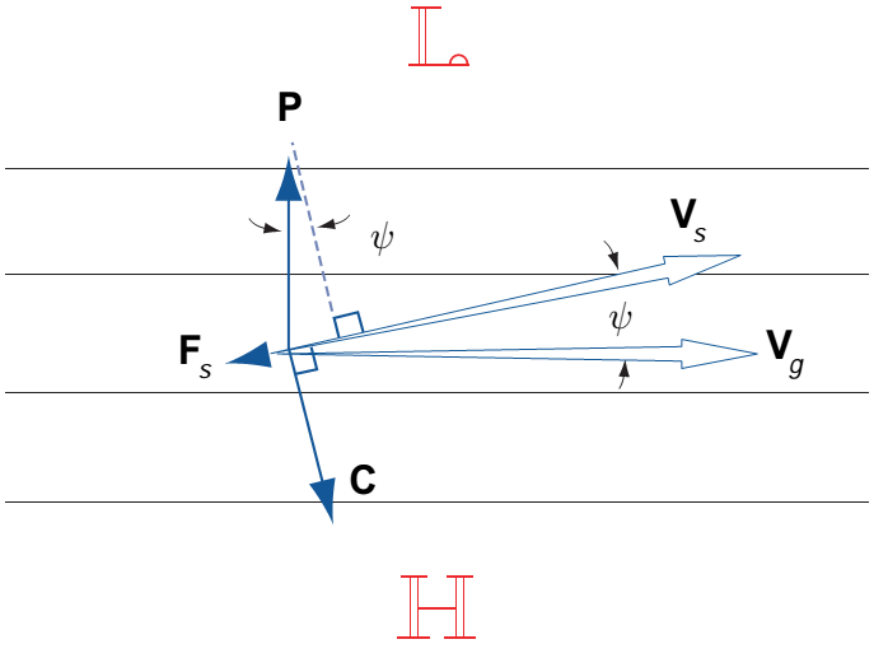


$$\nabla\phi = -f\boldsymbol{k} \times \boldsymbol{V}_g$$

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

Wallace and Hobbs, 2006

Effect of Friction by Earth Surface



P = Pressure gradient force
C = Coriolis force
Fs = Friction force
$\mathbf{V}_s$ = Wind
$\mathbf{V}_g$ = geostrophic wind

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

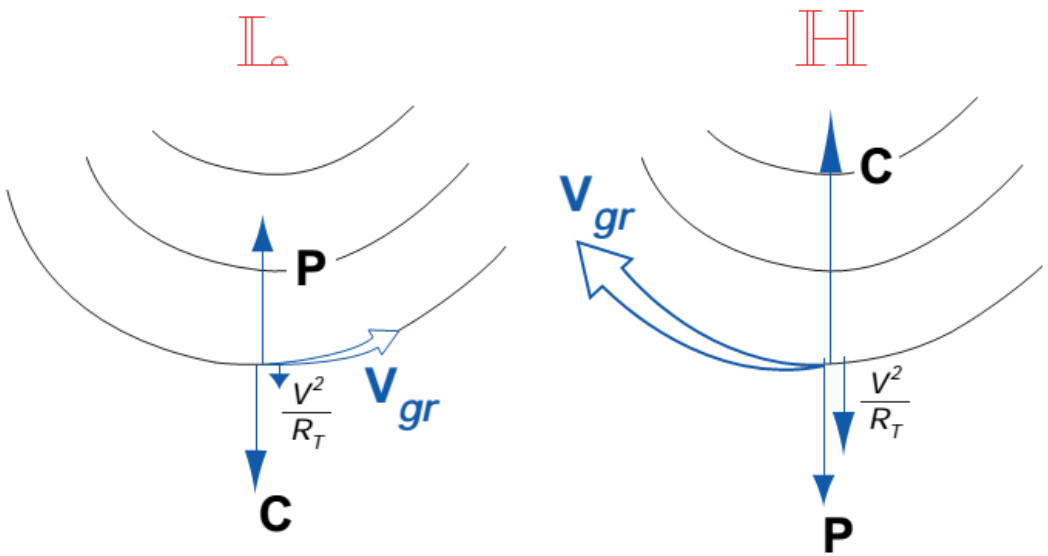
Wallace and Hobbs, 2006

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

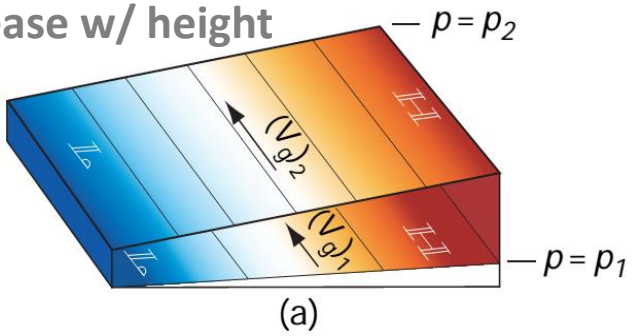
Thermal Wind (large-scale, without friction)

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

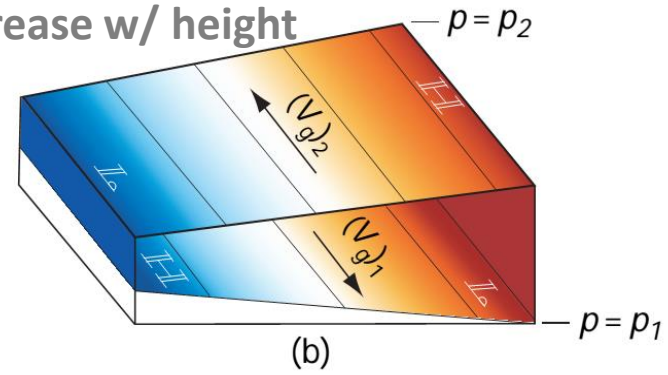
$$= \frac{R}{f}\ln\frac{p_1}{p_2}\boldsymbol{k} \times \nabla\bar{T} \qquad \textit{Hydrostatic}$$

Equivalent Barotropic

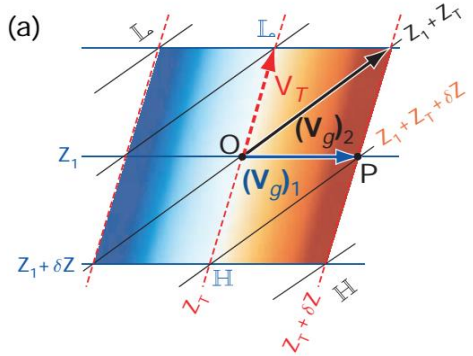
V_g increase w/ height



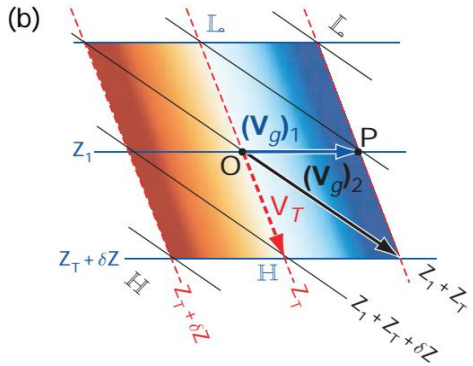
V_g decrease w/ height



Baroclinic: thermal advection



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)

$$\frac{\partial \Phi}{\partial p} = -\frac{RT}{p} \qquad d\Phi = gdz$$

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

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

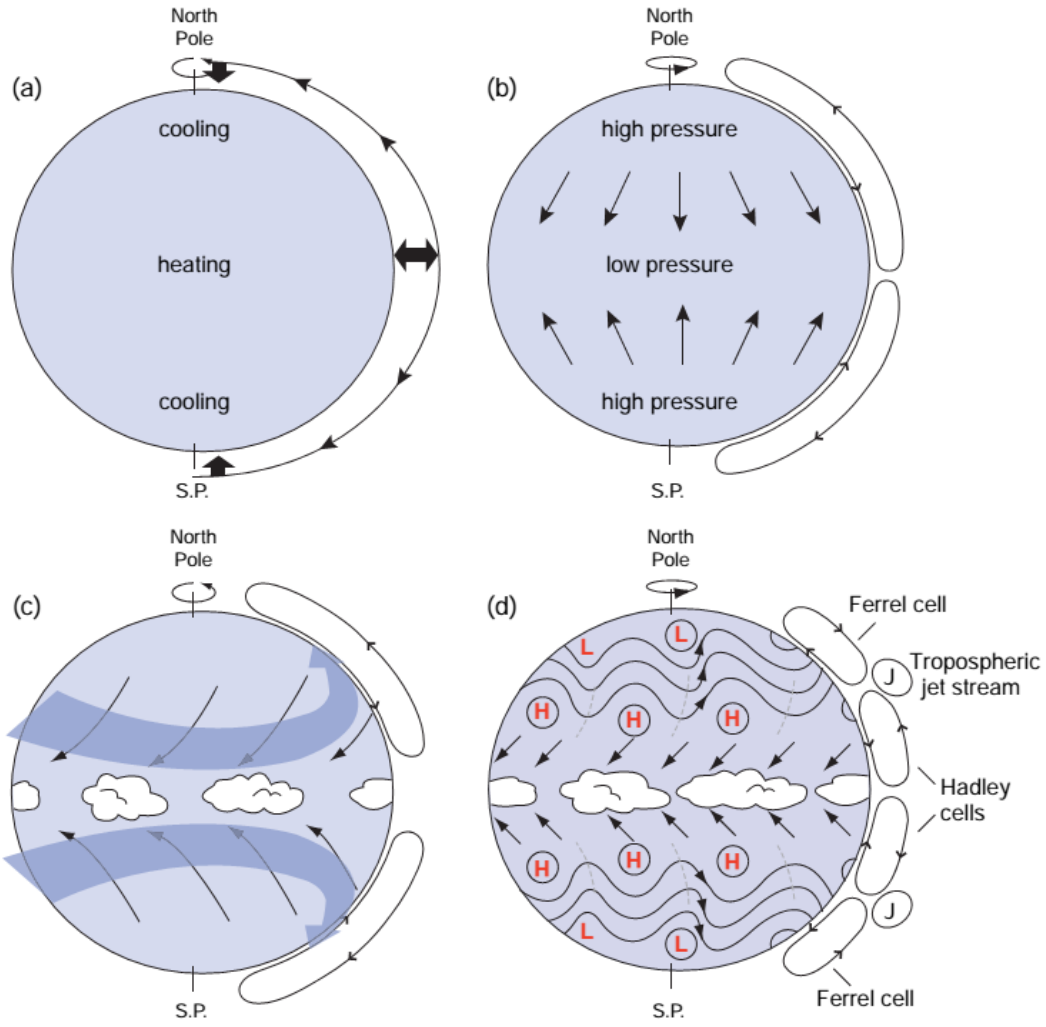
Five unknowns

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

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

Wallace and Hobbs, 2006

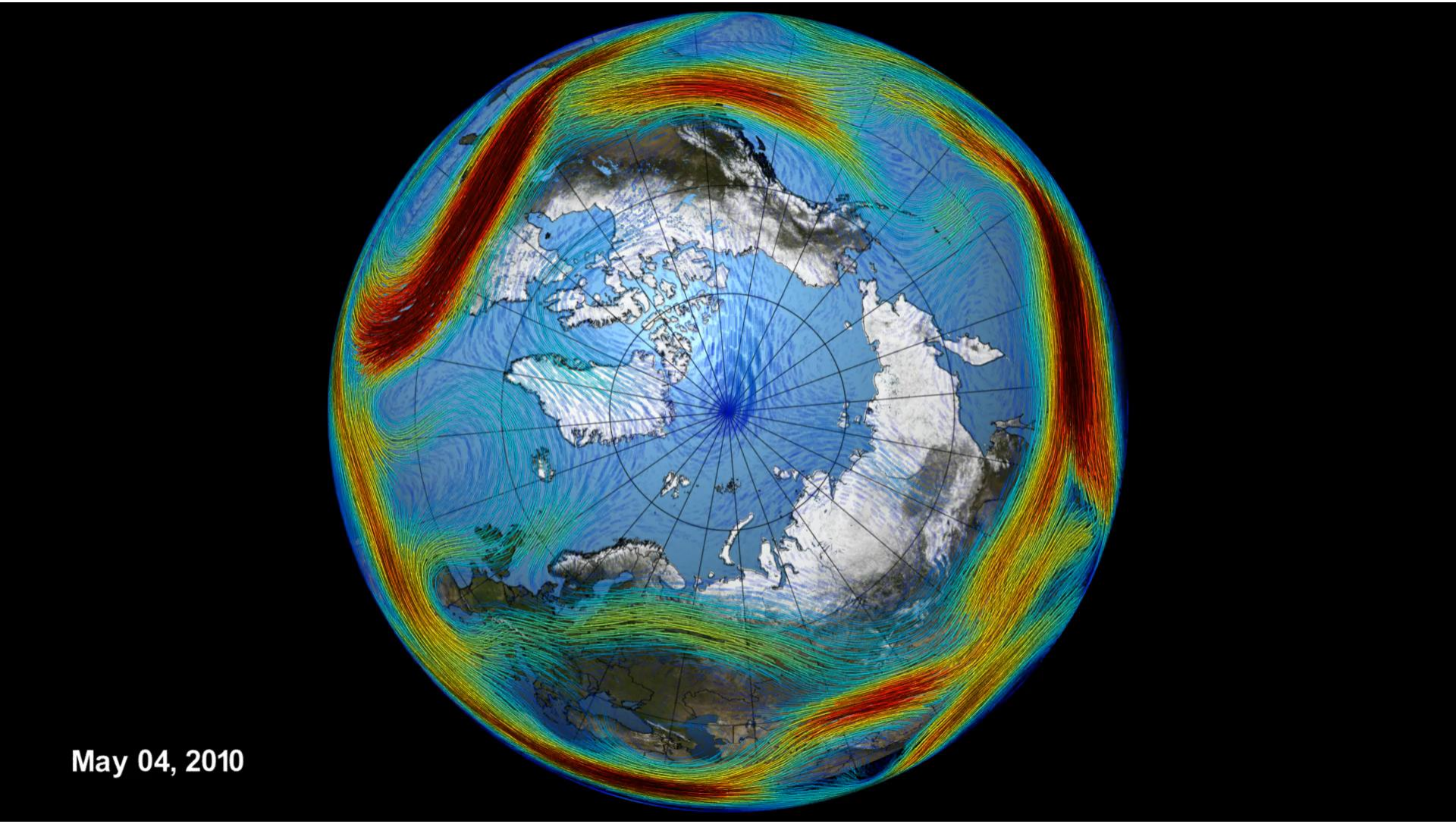
Primitive Equations and General Circulation



Baroclinic instability and waves

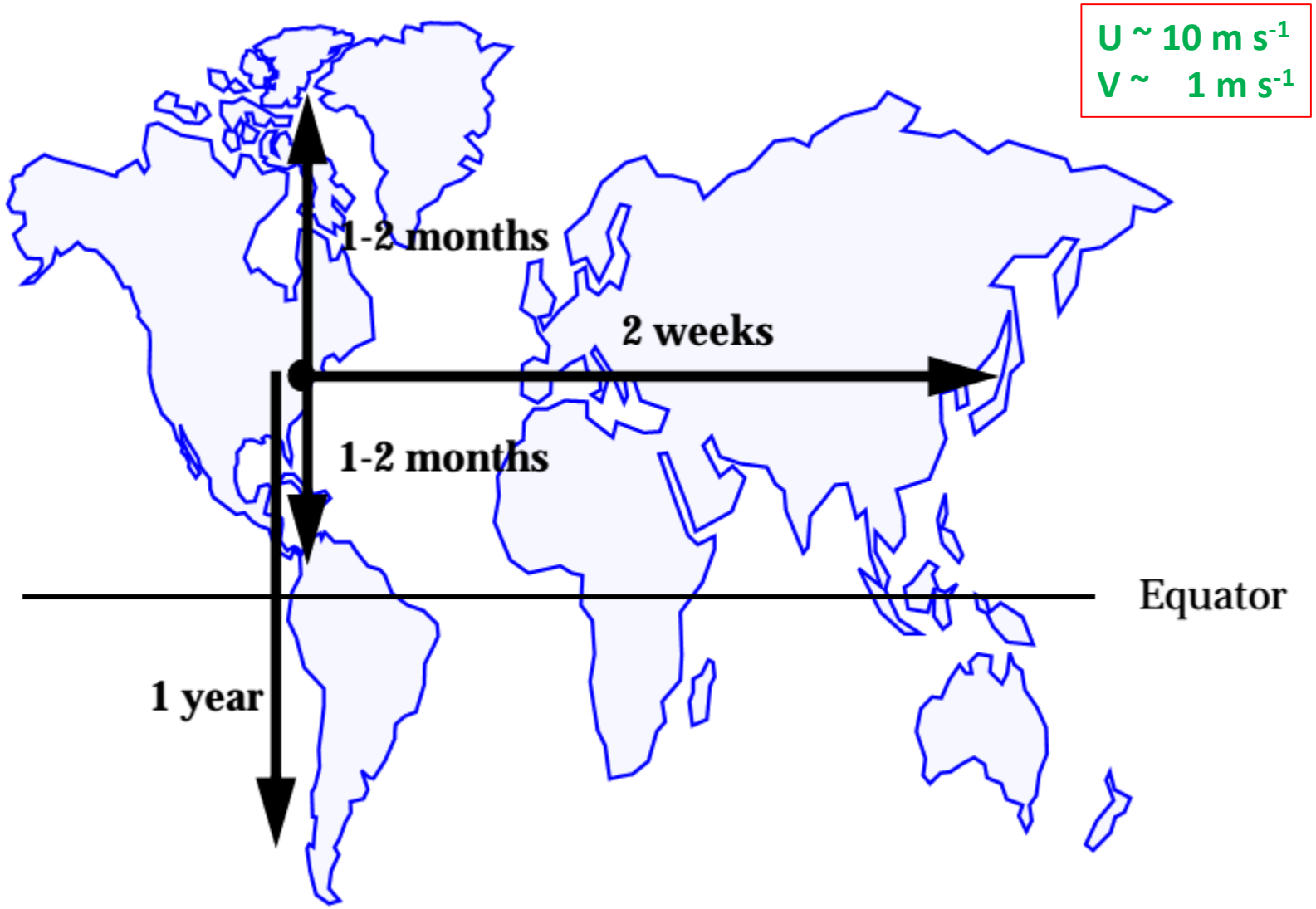
Wallace and Hobbs, 2006

Atmospheric Circulation



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

Characteristic Time Scales of Horizontal Transport



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 (θ, $θ_e$ 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)

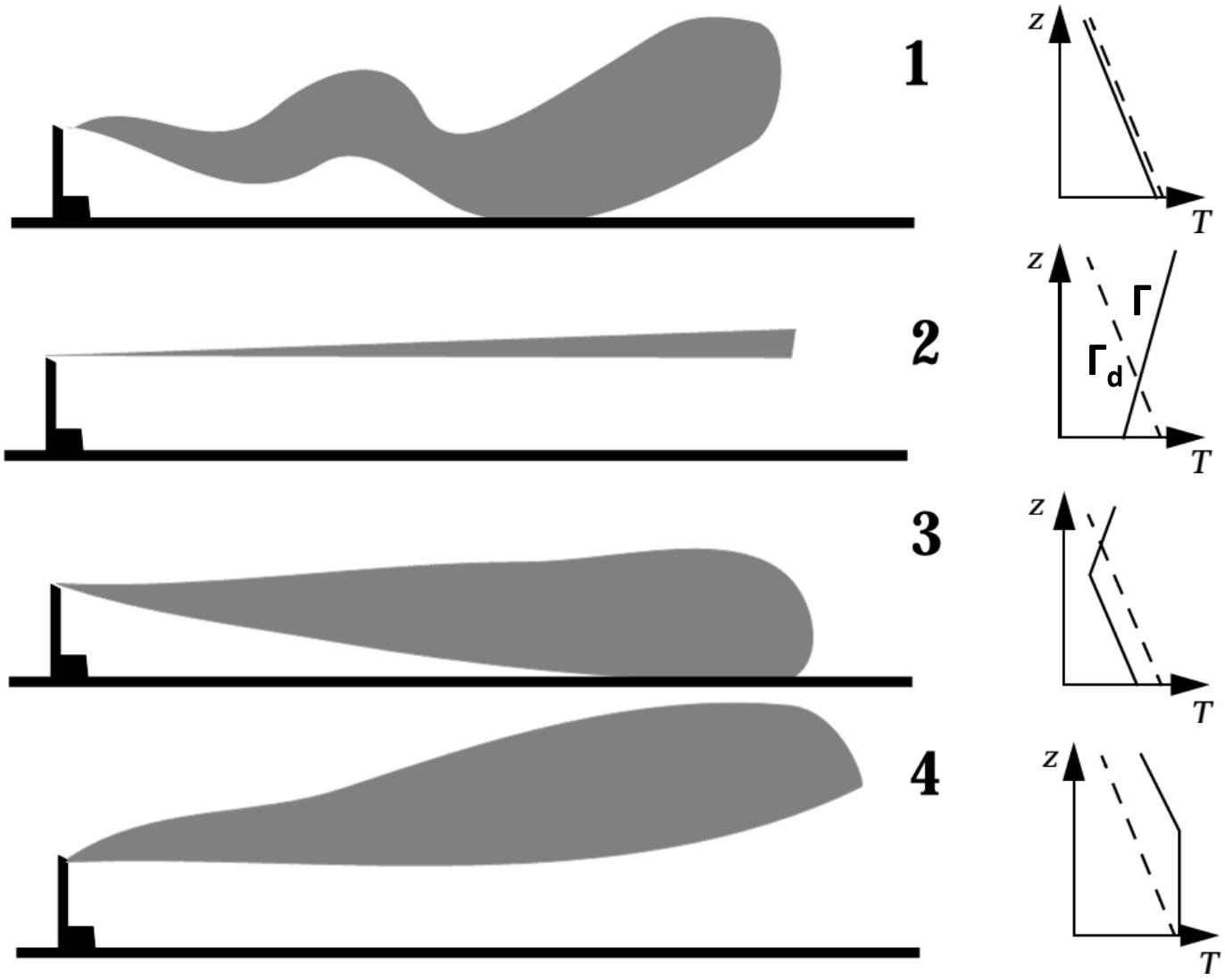
$$\Gamma_d = \frac{g}{c_p} \qquad \theta = T\left(\frac{p_0}{p}\right)^{R/c_p} \qquad \frac{1}{\theta}\frac{\partial\theta}{\partial z} = \frac{1}{T}(\Gamma_d - \Gamma)$$

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

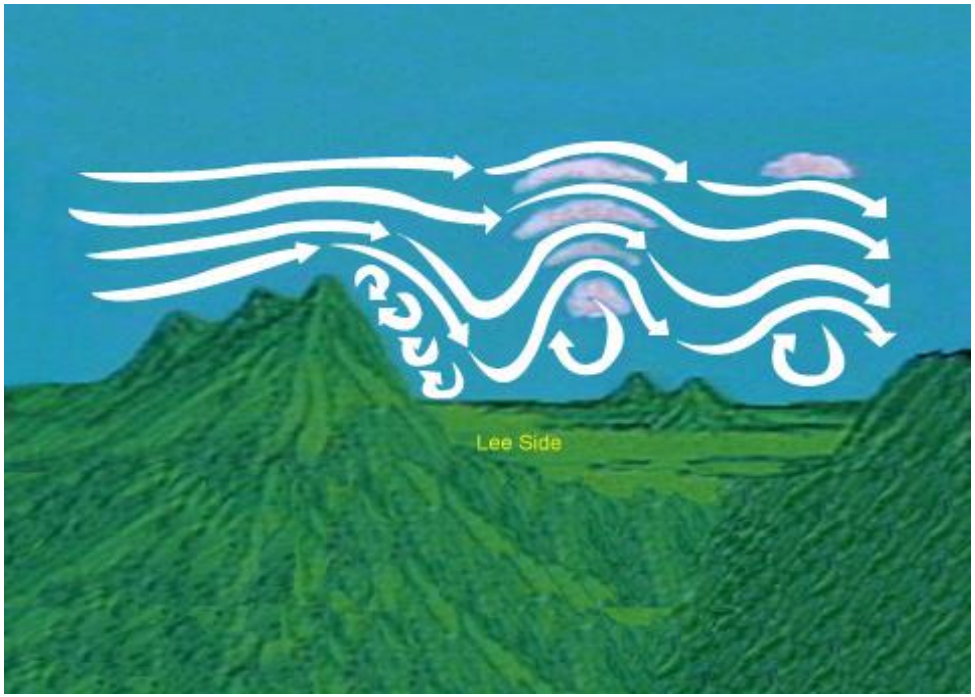


Jacob, 1999

Mechanical Instability

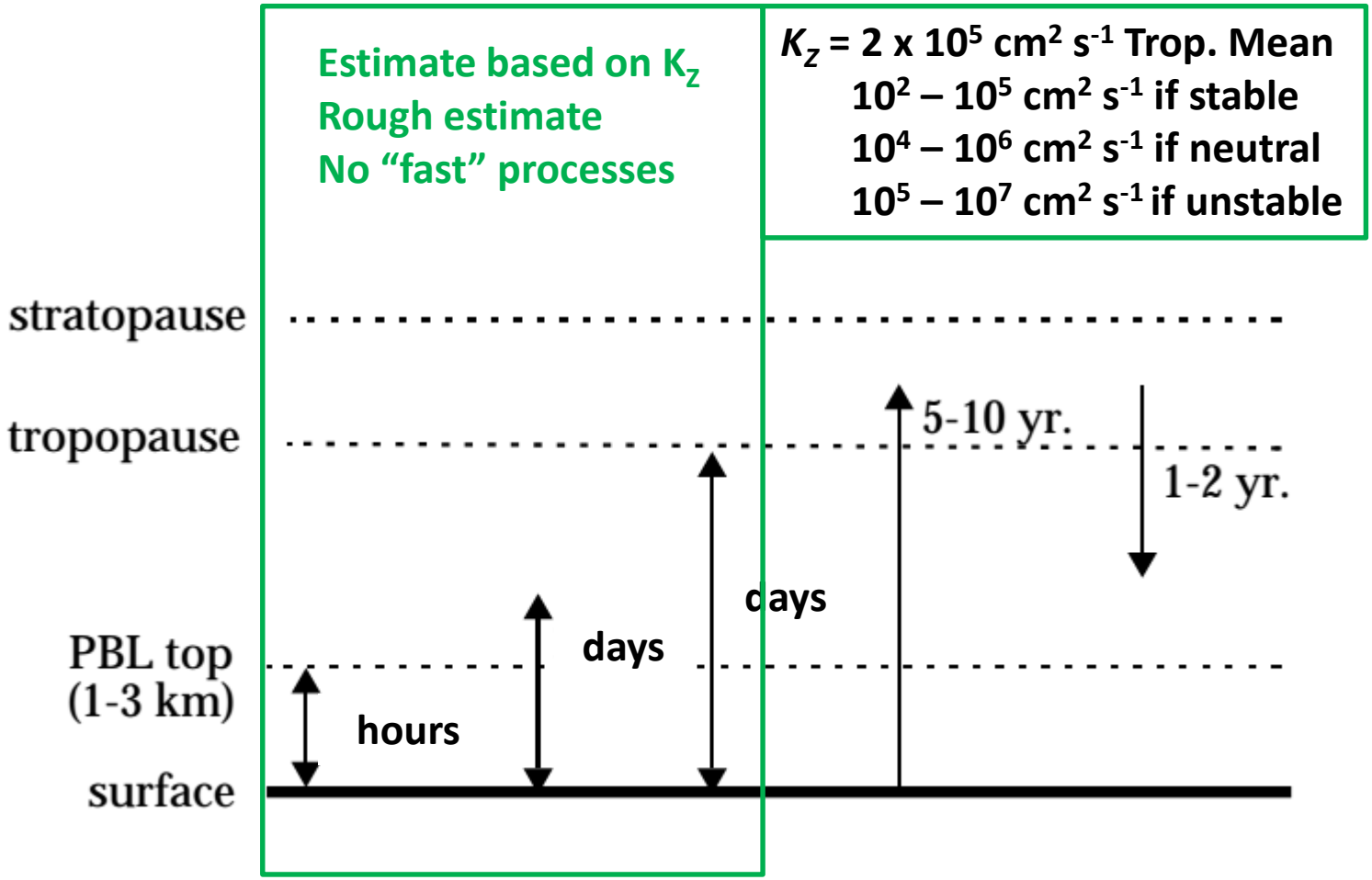
Causes:

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



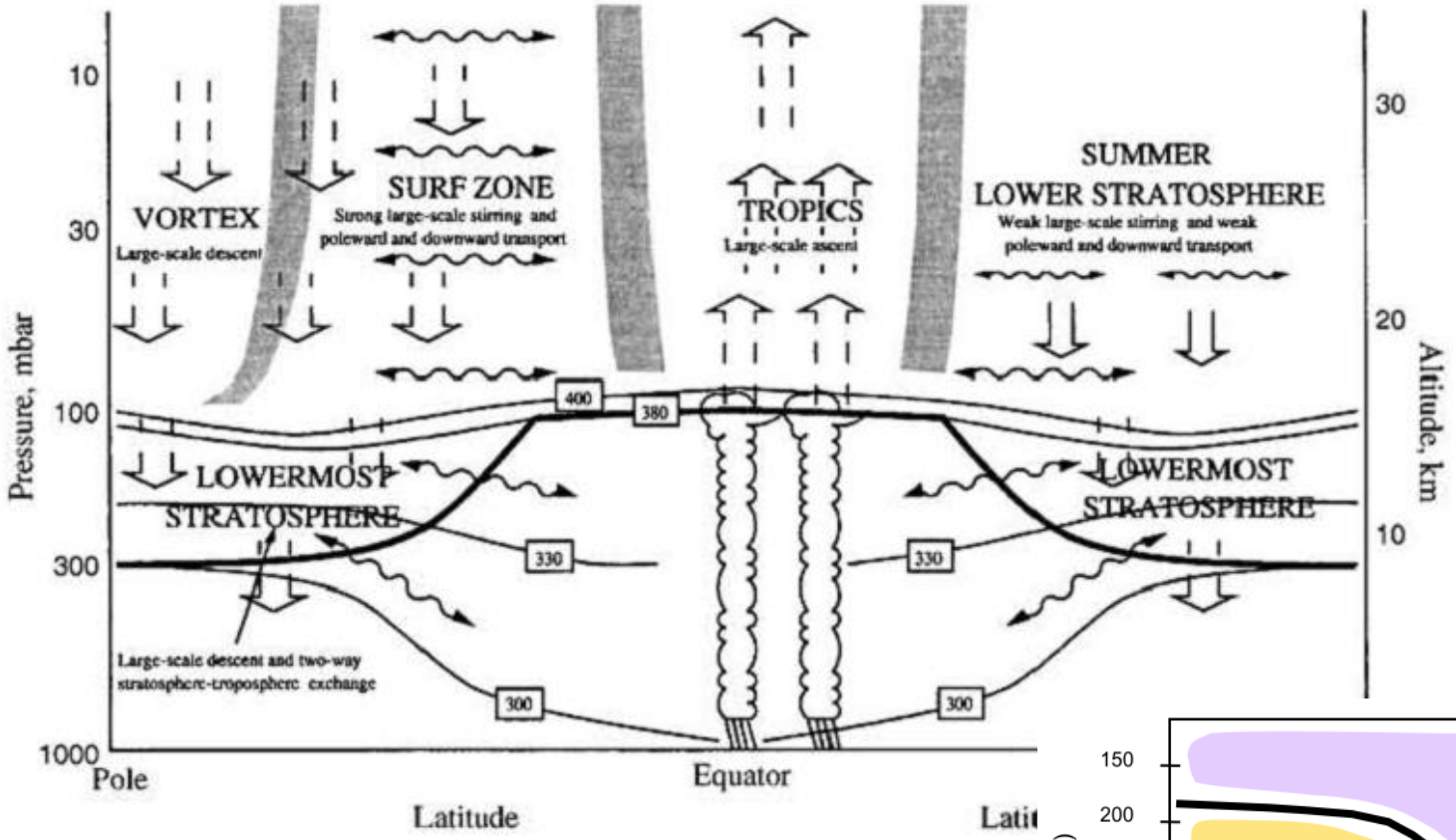


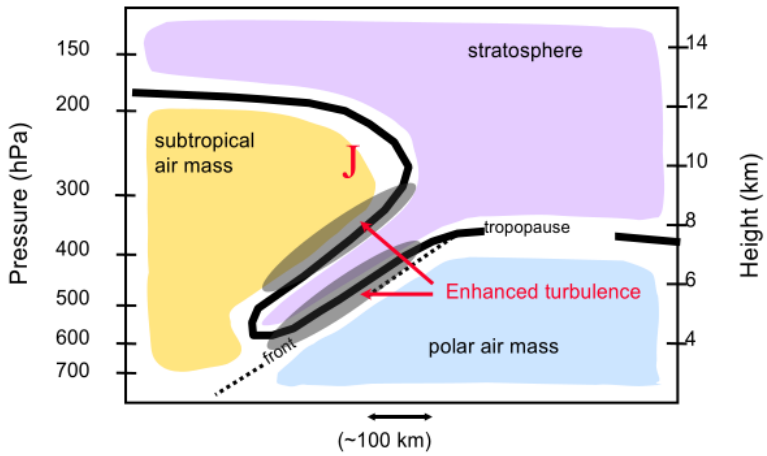
Characteristic Time Scales of Vertical Transport



Modified from Jacob (1999)

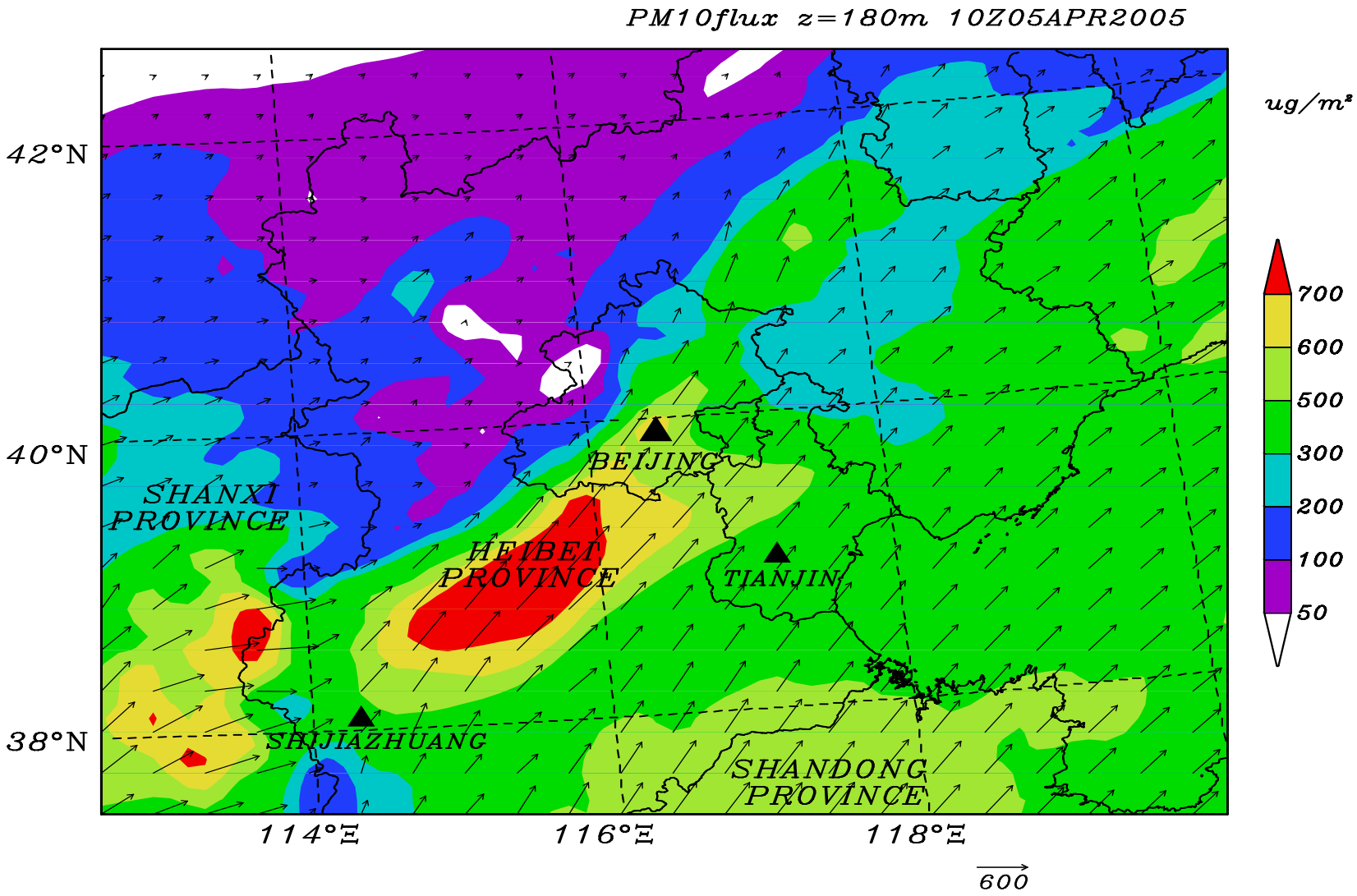
Stratosphere-Troposphere Exchange





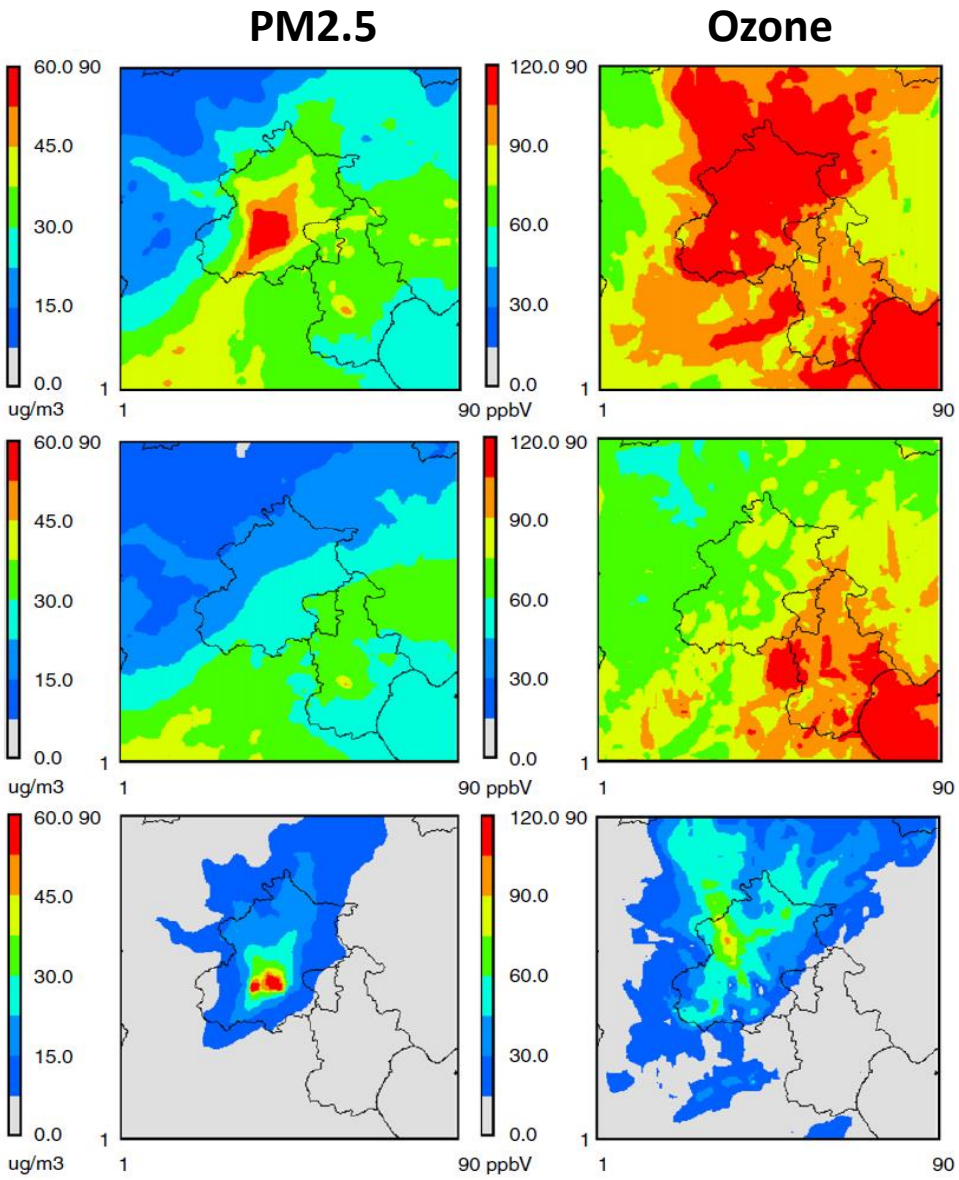
Seinfeld and Pandis, 2006

Regional Transport and Transformation Affecting Beijing



An et al., ACP 2007

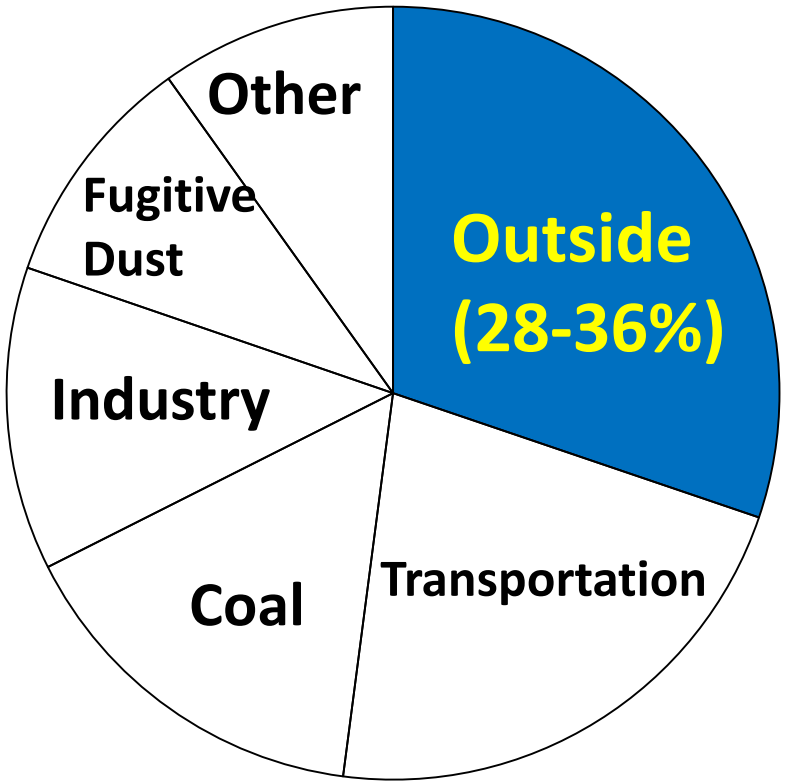
Regional Transport and Transformation Affecting Beijing



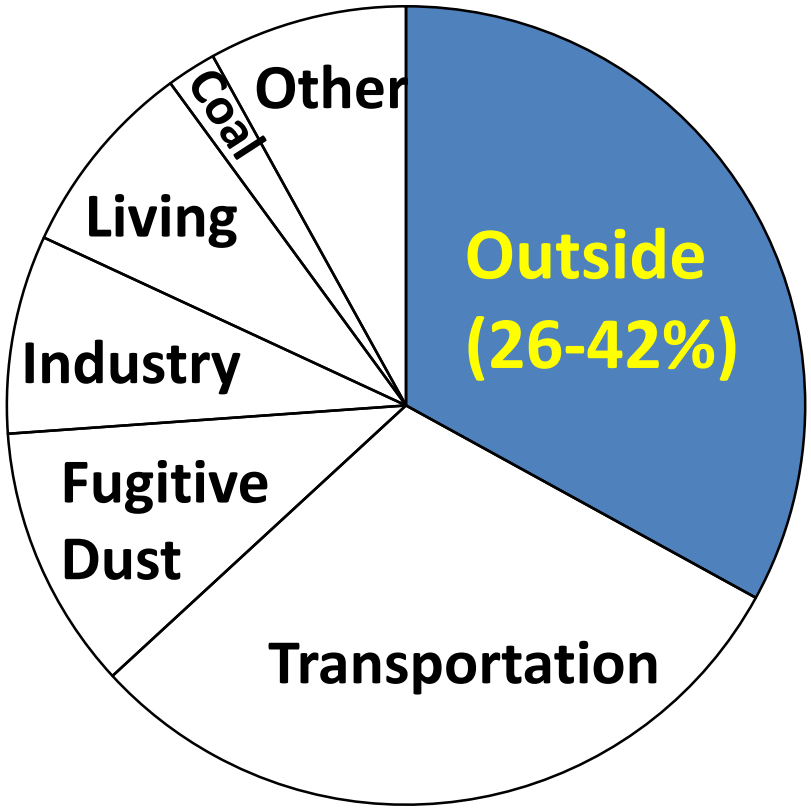
An et al., ACP 2007

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)

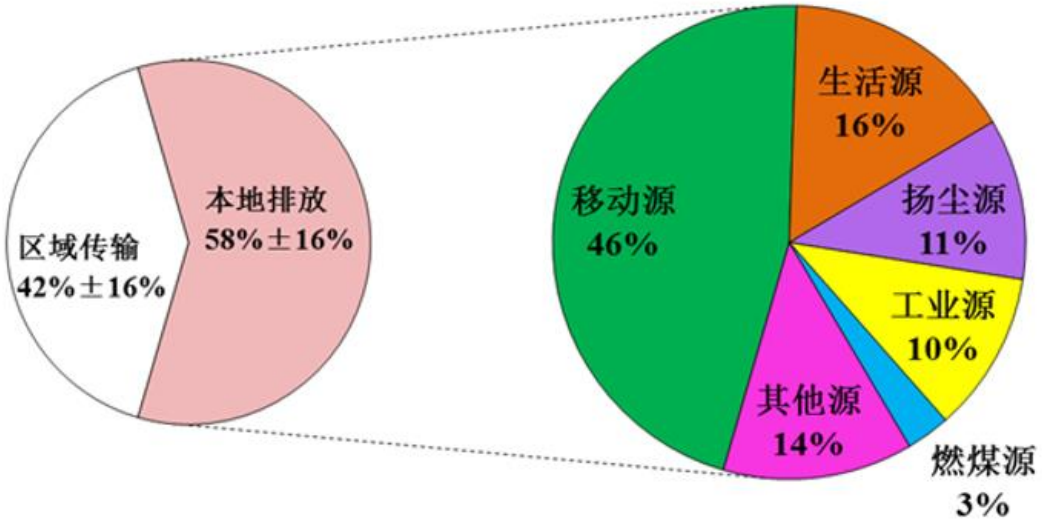


区域输送贡献：

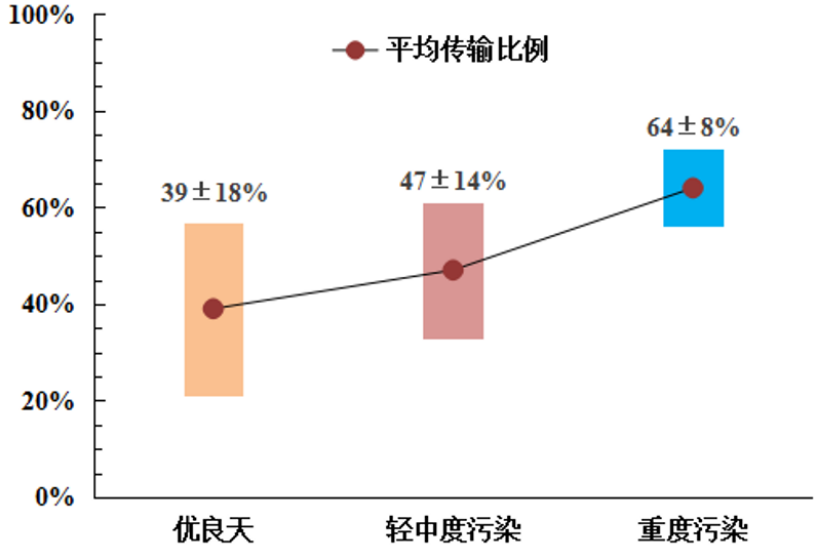
- 年平均：26%-42%
- 中度污染（115-150 μg/m³）：34%-50%
- 重污染日（＞150 μg/m³）：55%-75%

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

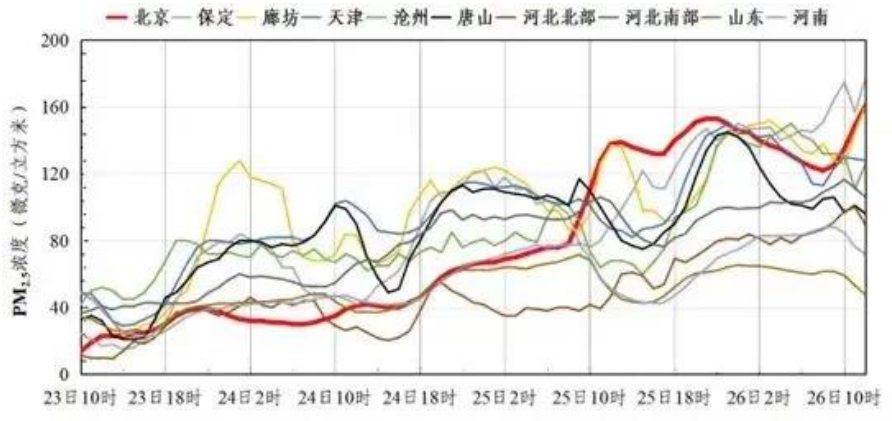
Back-trajectory analysis of BJ's PM on 2014/10/10

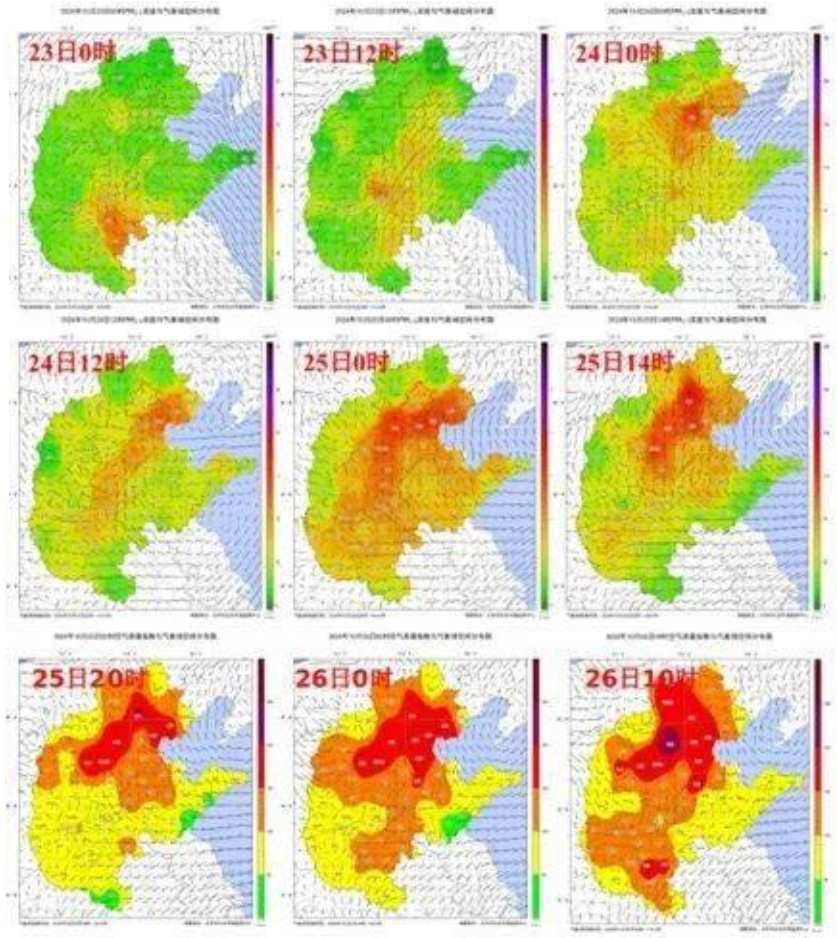




Severe Regional PM Pollution Transport to Beijing

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

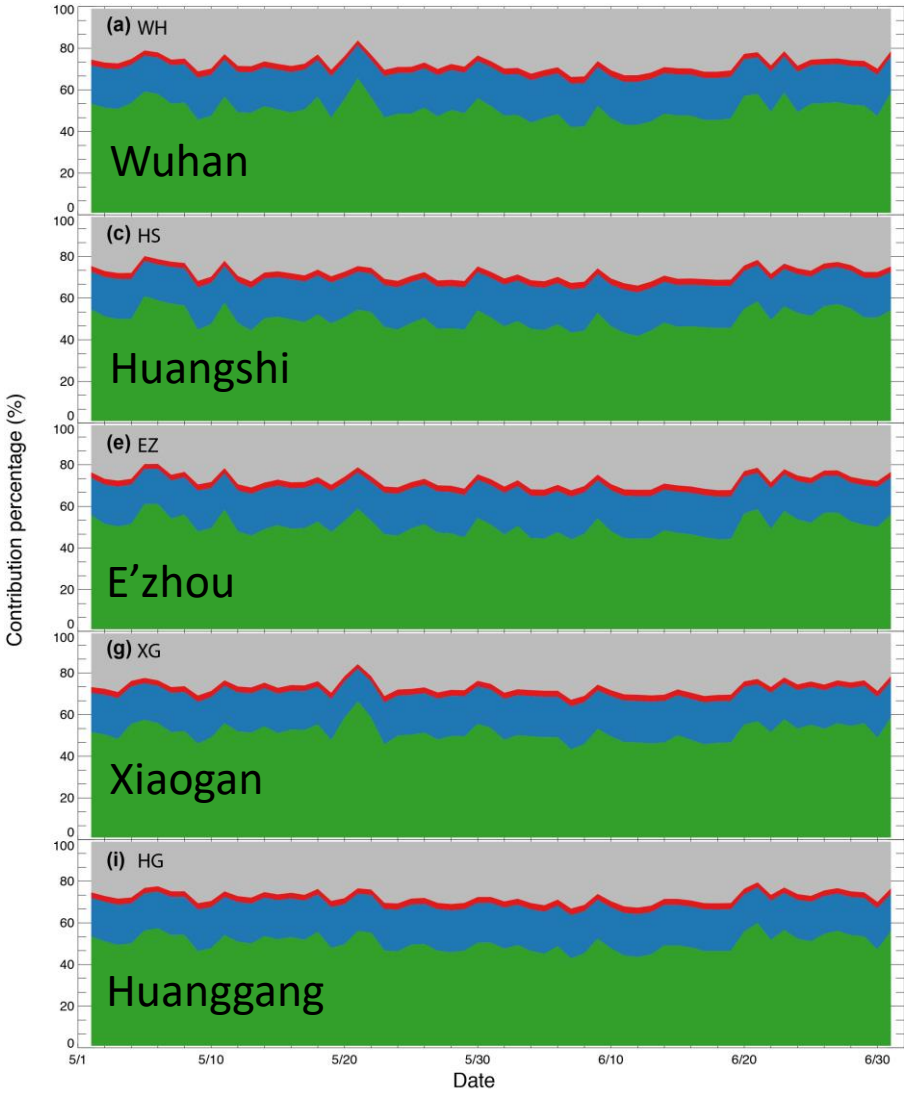




10月24日13时-26日14时，区域传输贡献77%，其中东南通道35%，西南通道18%，东南通道占据主导。

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

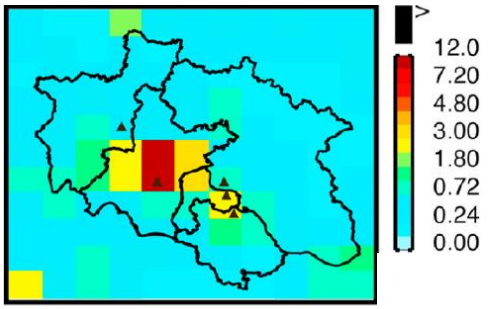
Key Roles of Local Production and Atmospheric Transport for Ozone Pollution over Central China



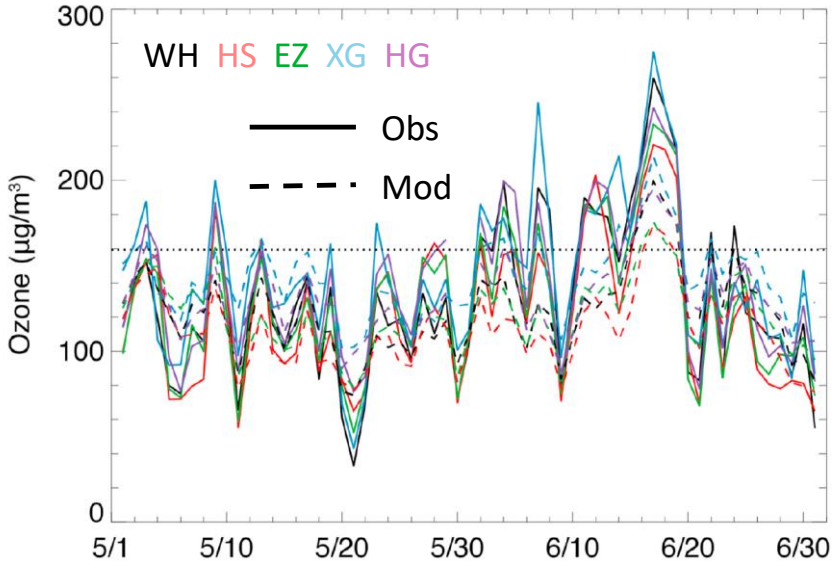
Contributors:

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

Anthro NOx emis (g/m^2)

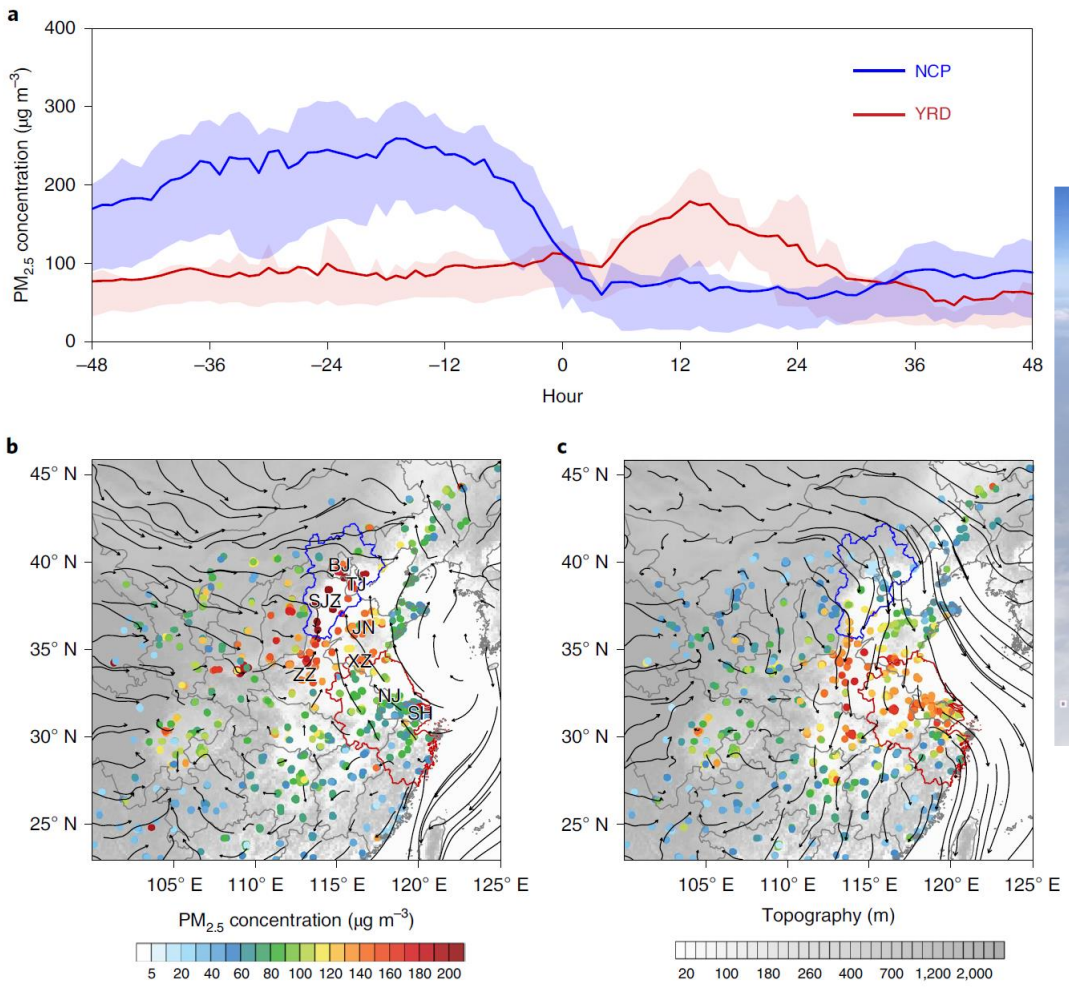


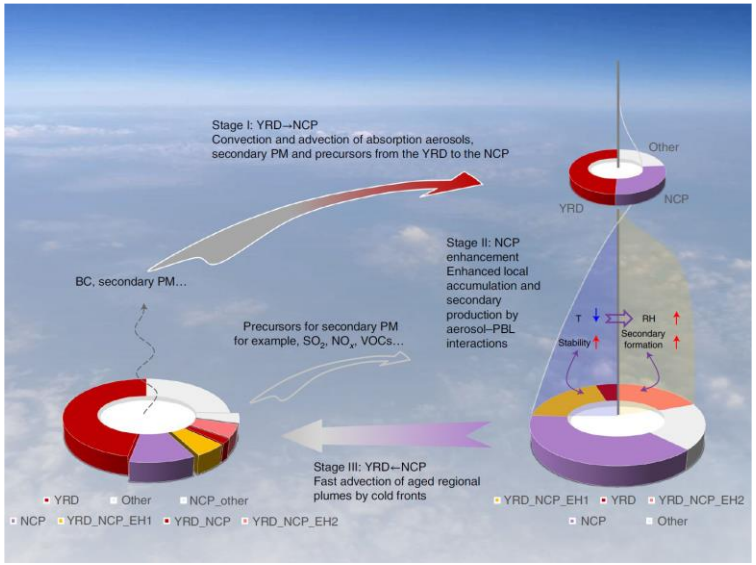
MDA8 ozone in May-June 2018



Yan YY et al., 2021, AE

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

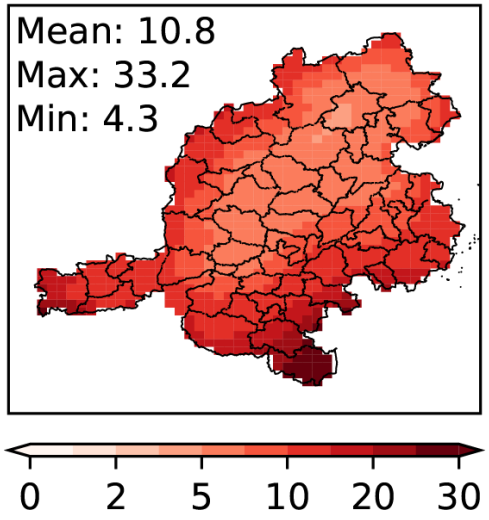




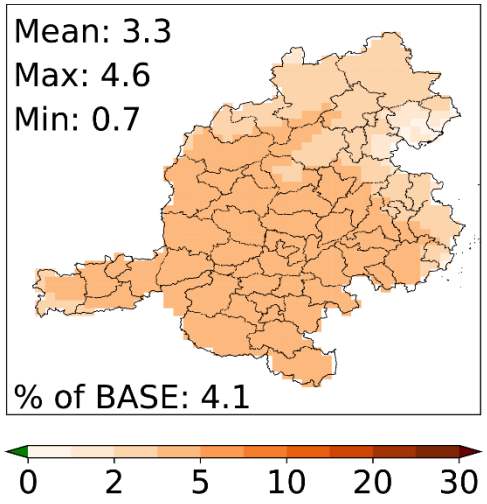
Huang et al., 2020, Nature Geoscience

Large Impact of Transport to Summer Ozone over NCP + FW

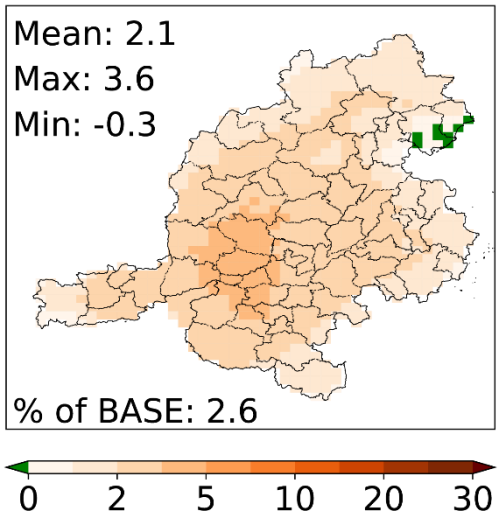
% contribution of transport to total JJA MDA8 O3 (%)



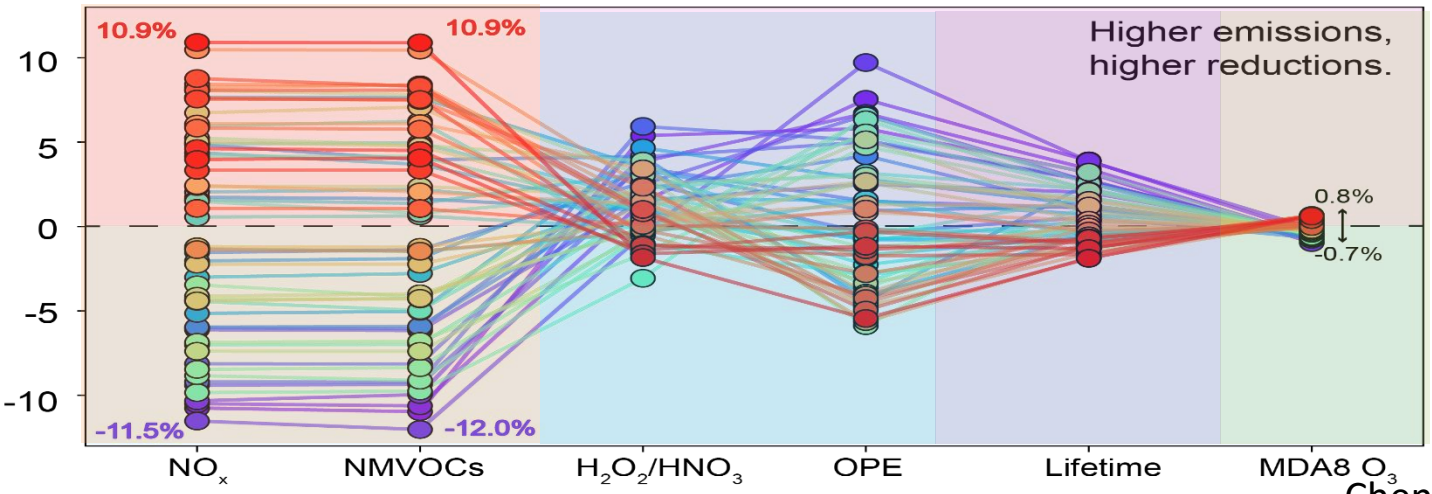
ΔO_3 by 20% emis cut in China



ΔO_3 by 20% emis cut in NCP+FW

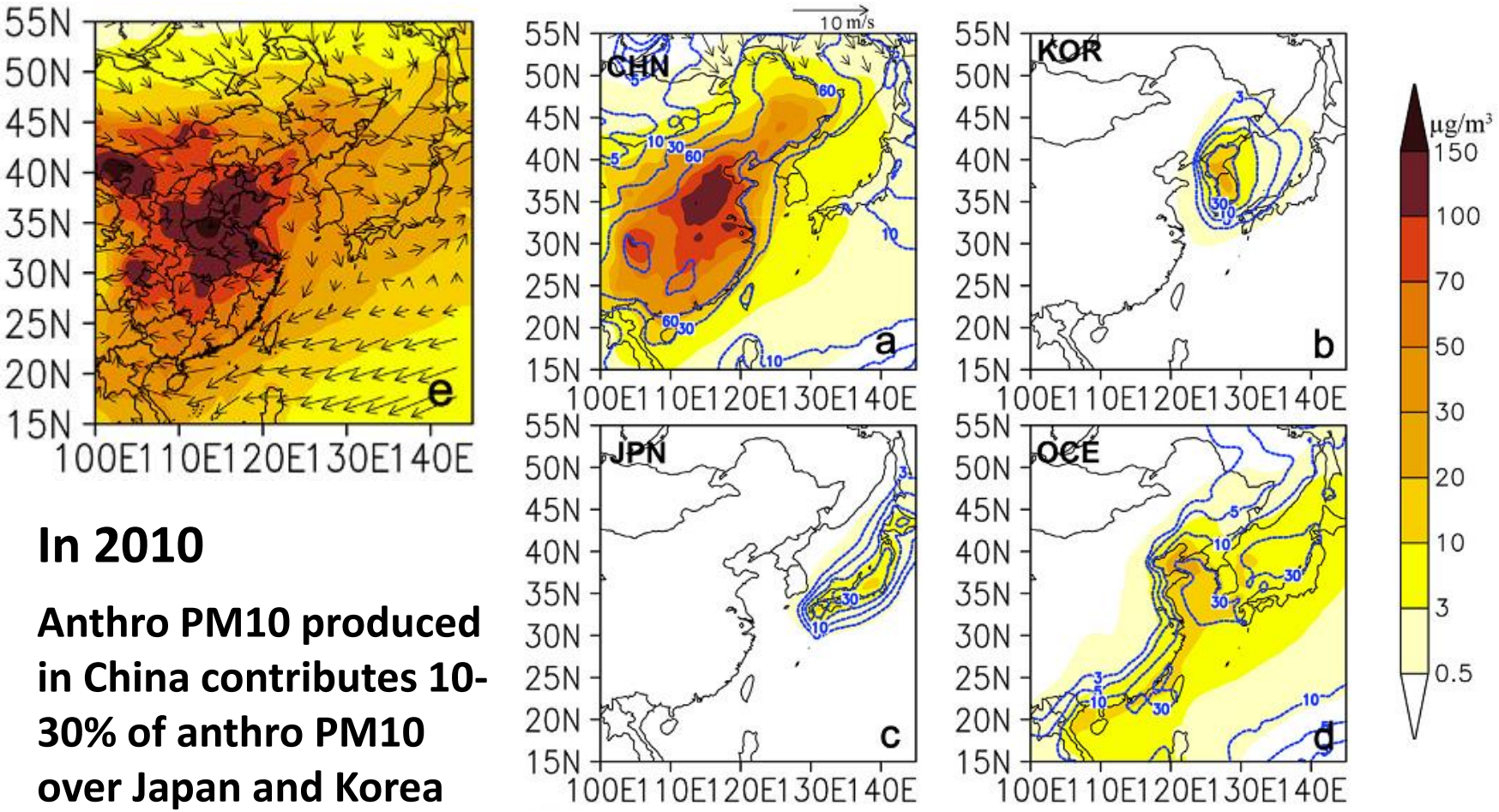


Relative to uniform 20% emis cut



Chen et al., ERL, 2025

PM Transport Between Asian Countries



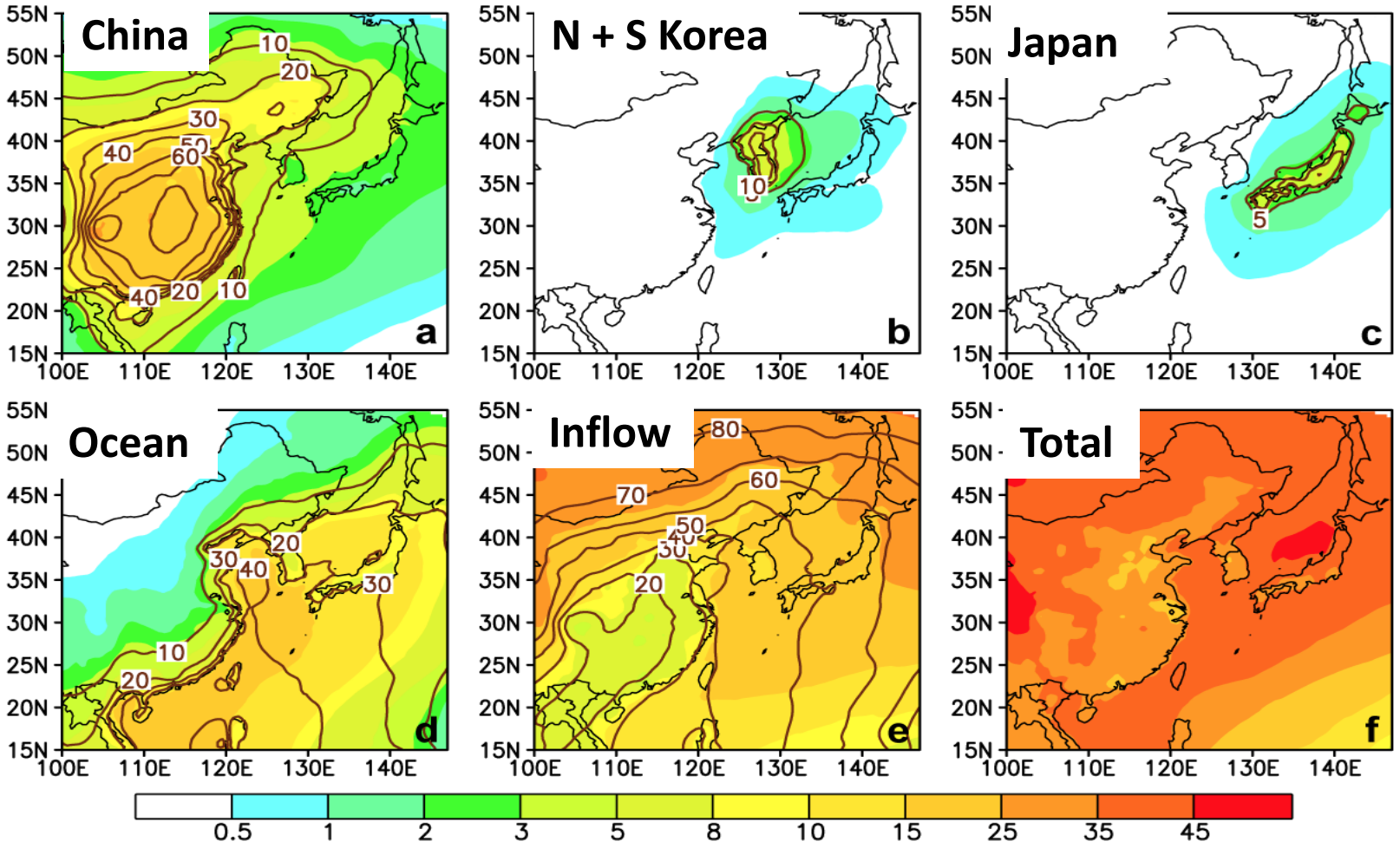
In 2010

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



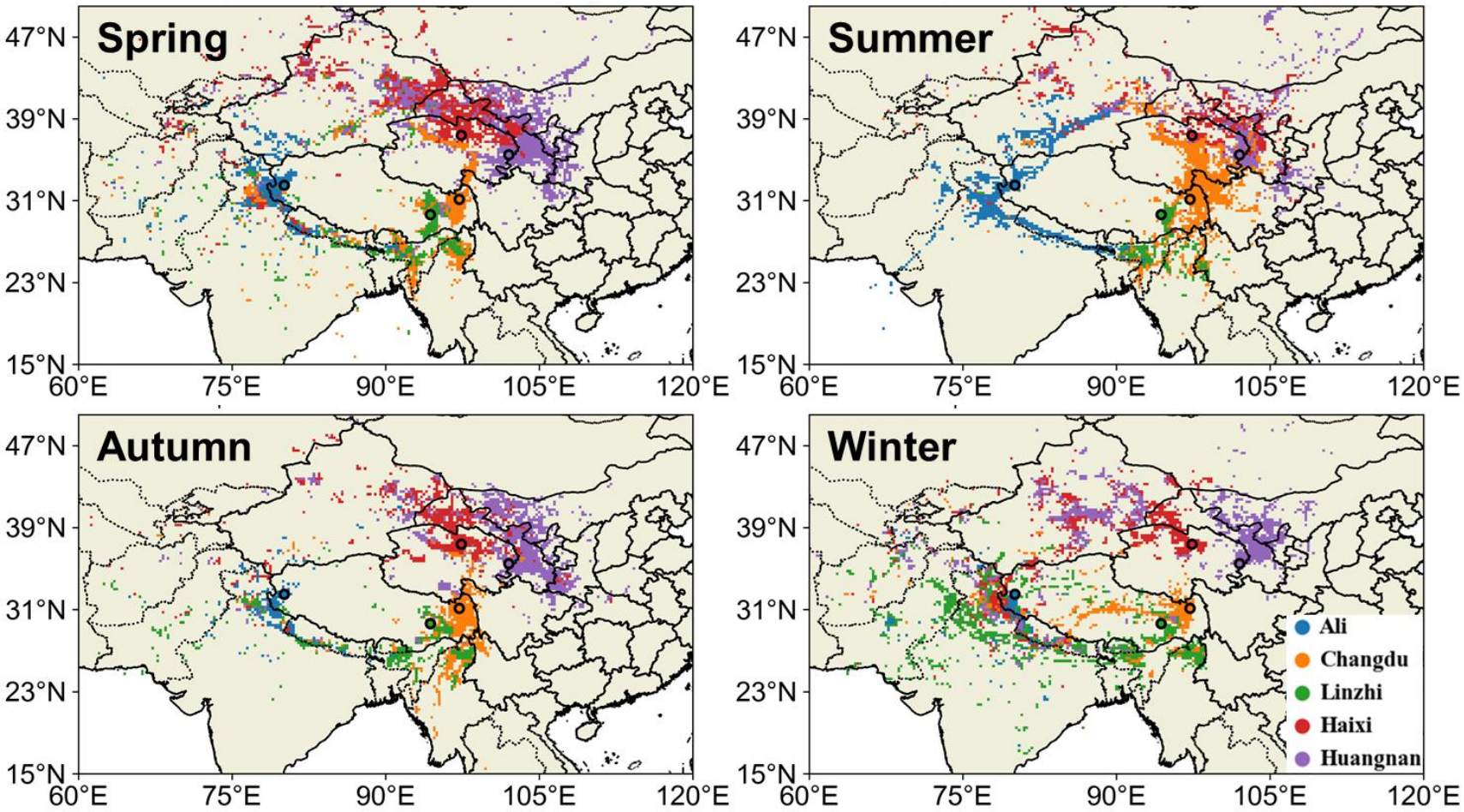
Color: ppbv; Contour: %

2010 mean

Li et al., 2016, AR

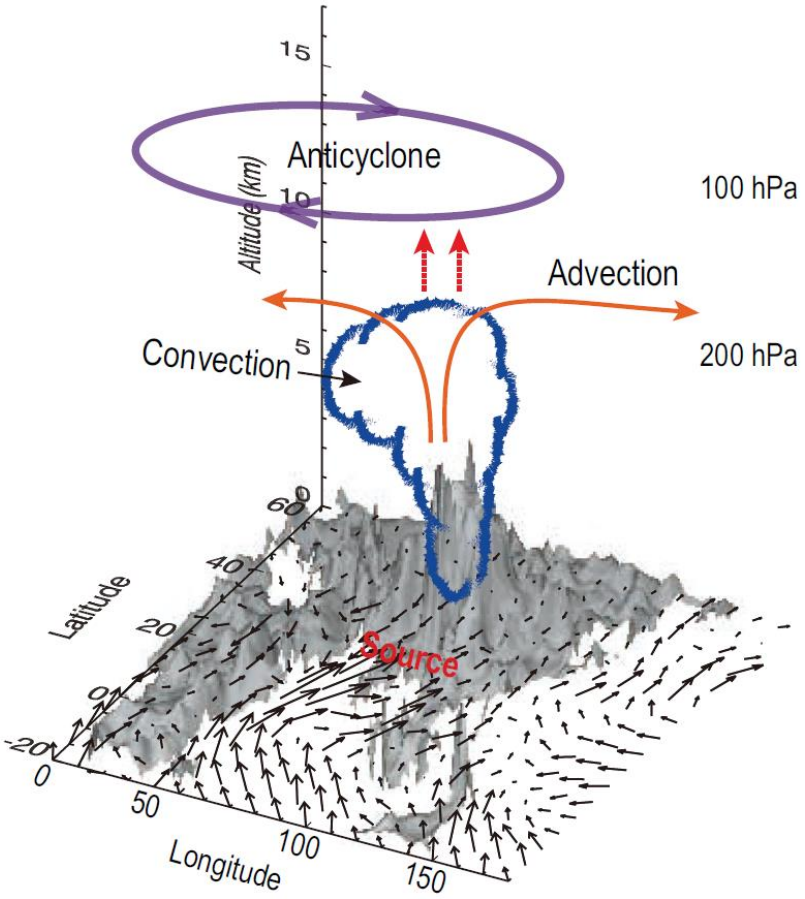
Transboundary Sources of Ozone over Tibetan Plateau

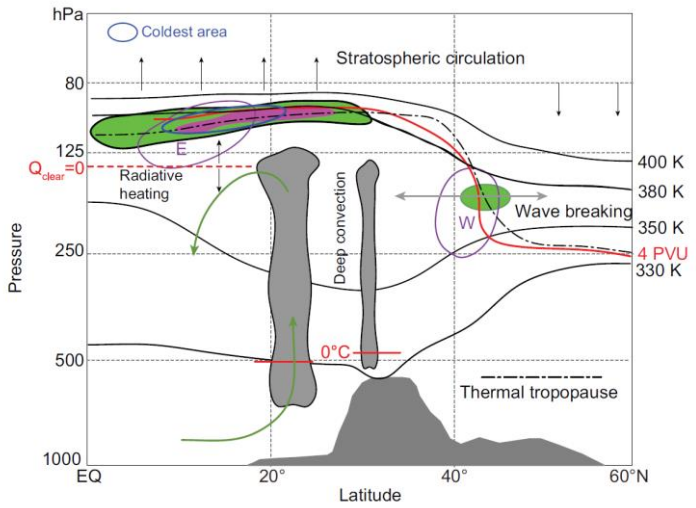
Ozone sources in each season based on WPSCF



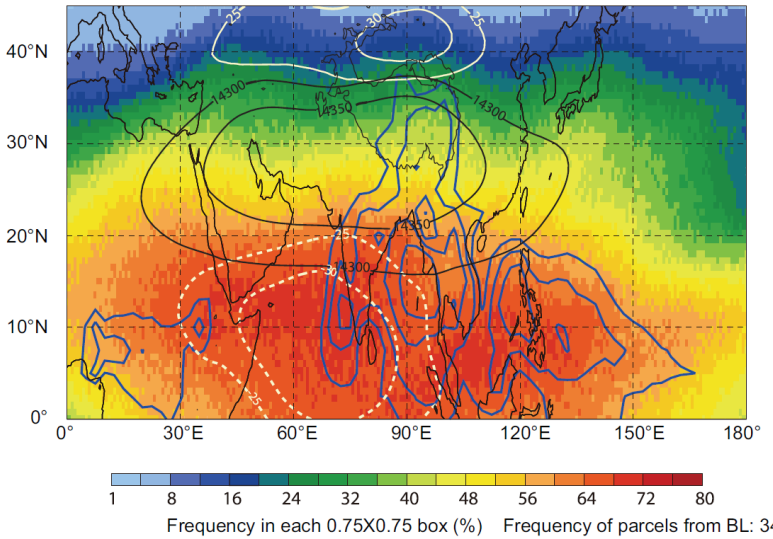
Xu et al., under review

Transport into Asian Tropical UTLS





Proportion of parcels from boundary layer (85% SP), JJA 2009-2014



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

Pathways and Time of Transpacific Transport

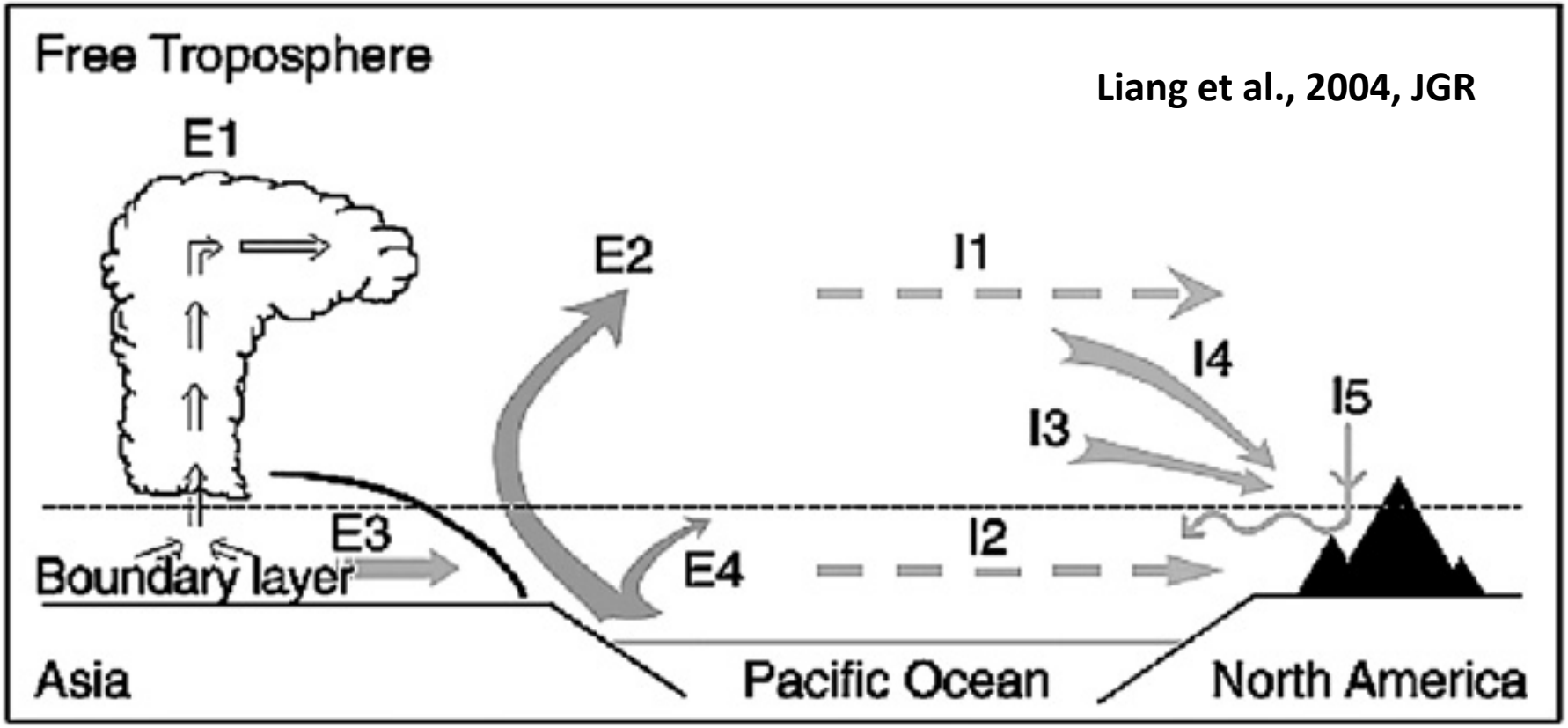
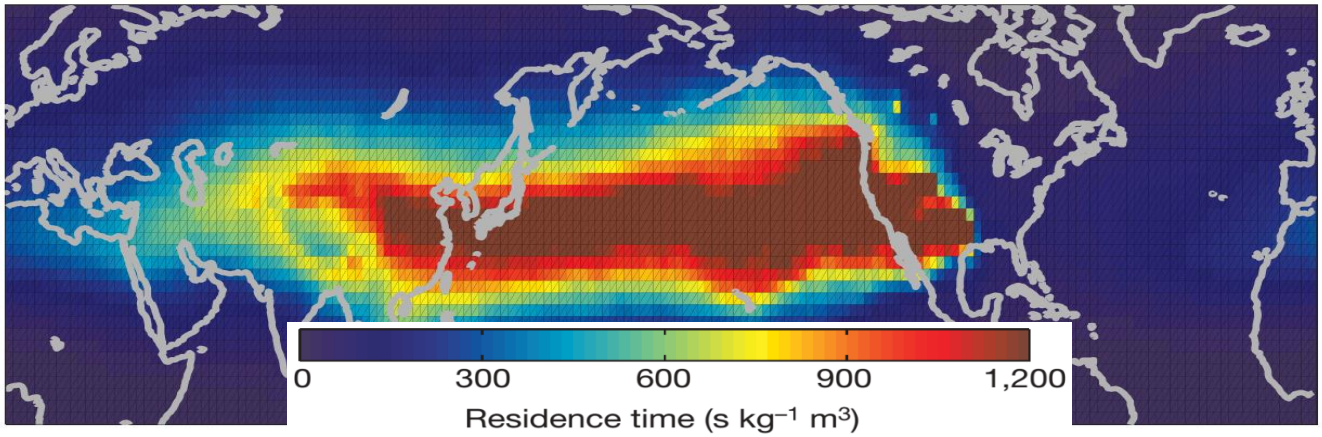


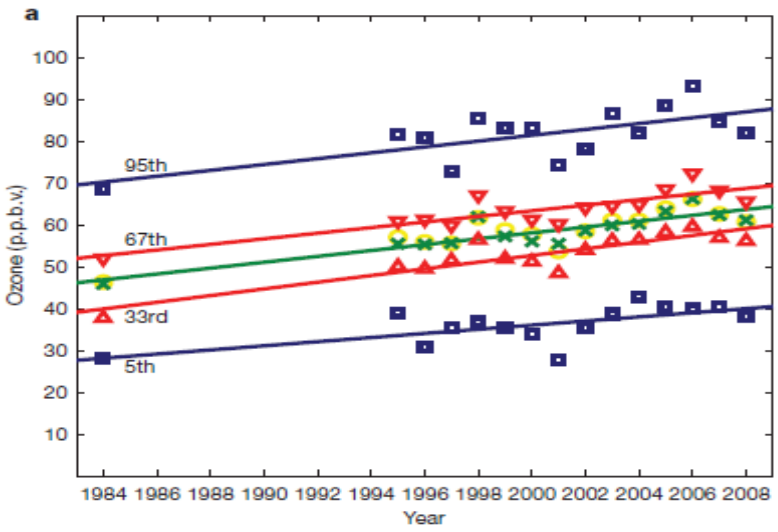
Table 1. 11-Year Average Inter-Continental Transport Times for Two Sets of Tracers in April (Unit: Weeks)

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





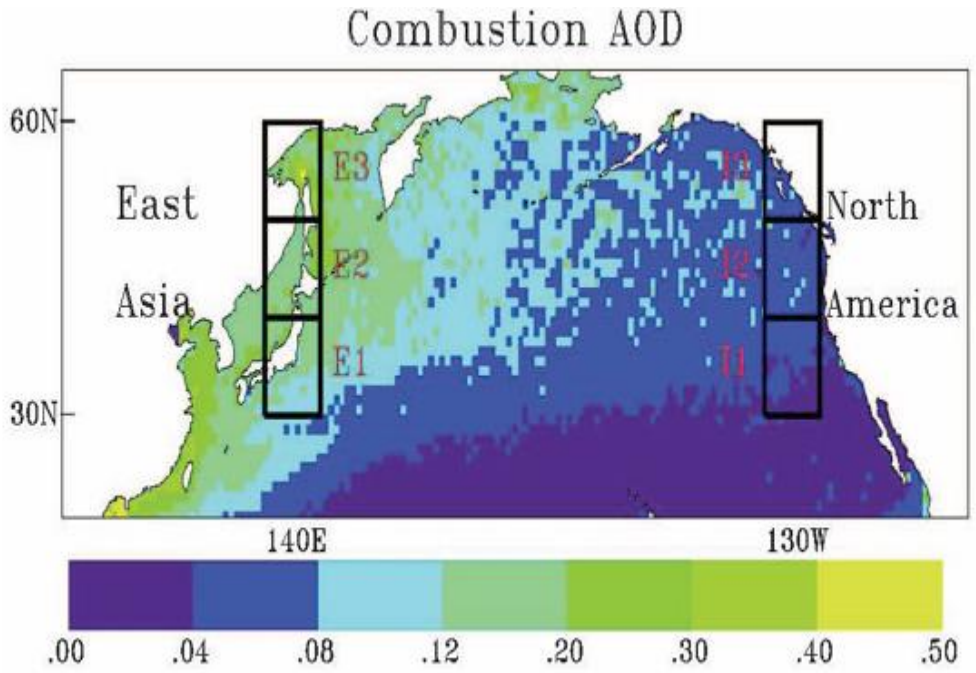
Percentile	1995–2008 increase (p.p.b.v. yr^{-1})	*P*
95th	0.56 ± 0.71	0.11
67th	0.62 ± 0.39	0.00
50th	0.63 ± 0.34	0.00
33rd	0.64 ± 0.35	0.00
5th	0.48 ± 0.53	0.07

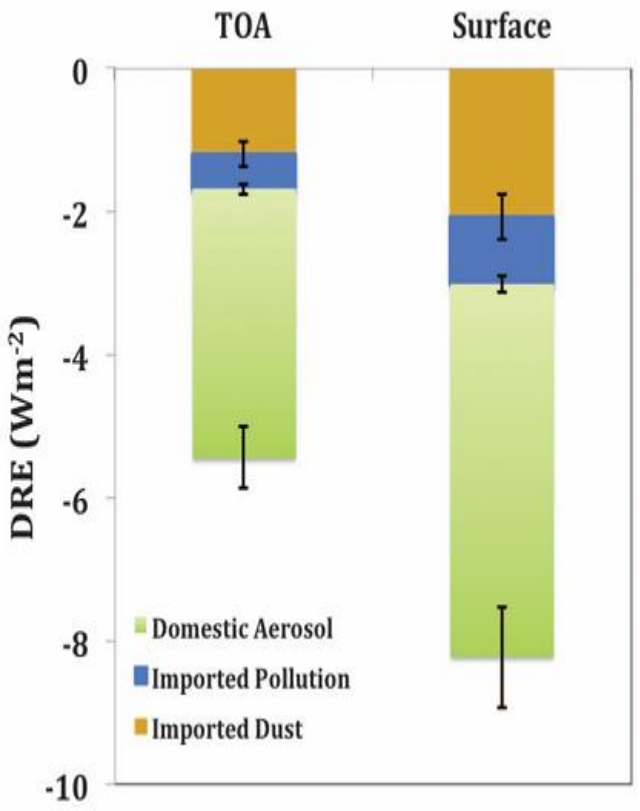
Percentile	1984–2008 increase (p.p.b.v. yr^{-1})	*P*
95th	0.70 ± 0.45	0.01
67th	0.67 ± 0.25	0.00
50th	0.70 ± 0.22	0.00
33rd	0.80 ± 0.24	0.00
5th	0.49 ± 0.34	0.01

Asian PM Transport Affects North America

Yu et al., 2012, Science

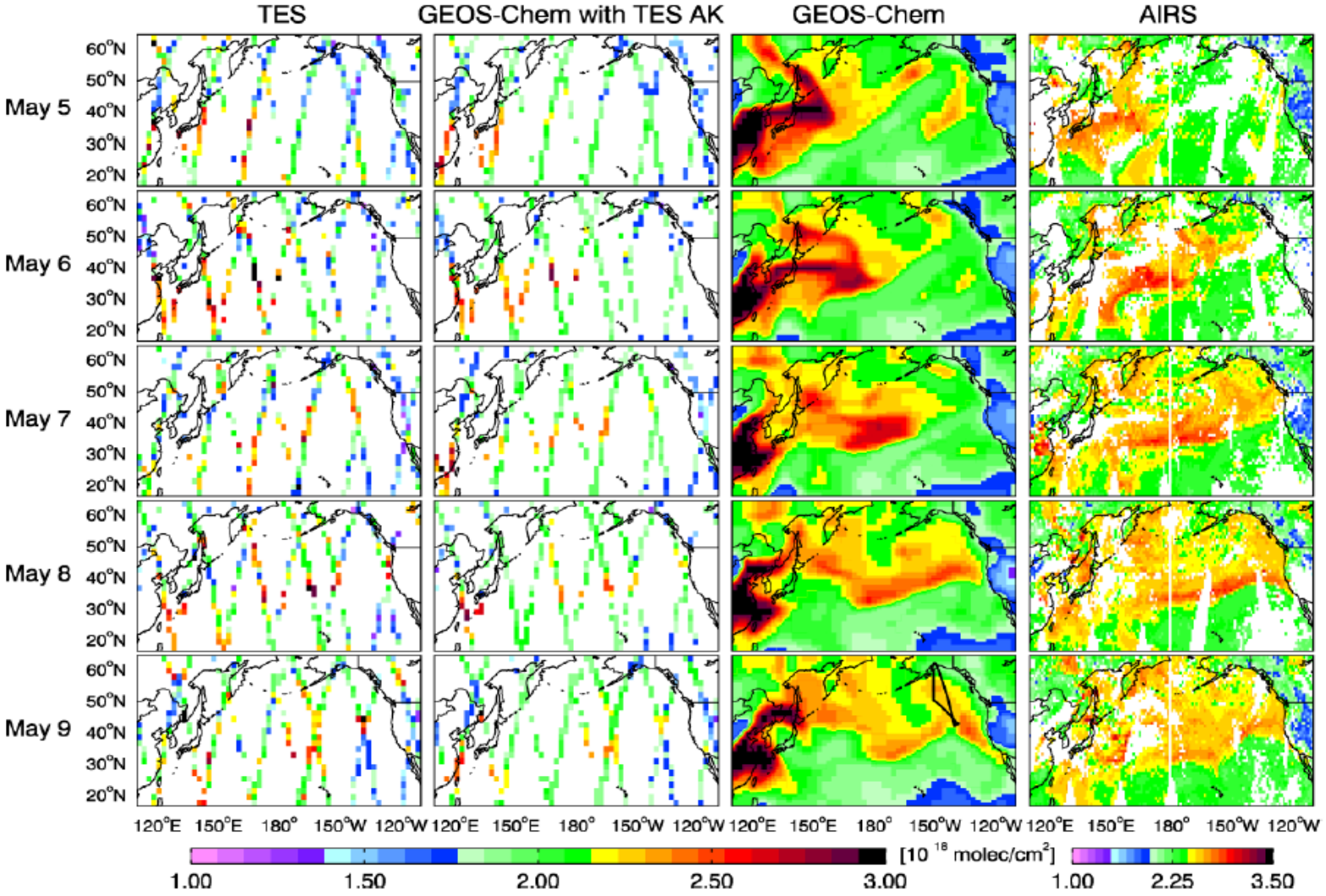
- **East Asian PM pollution contributes 6% of N.A. DRE**





East Asian Influence: Satellite Obs. and CTMs

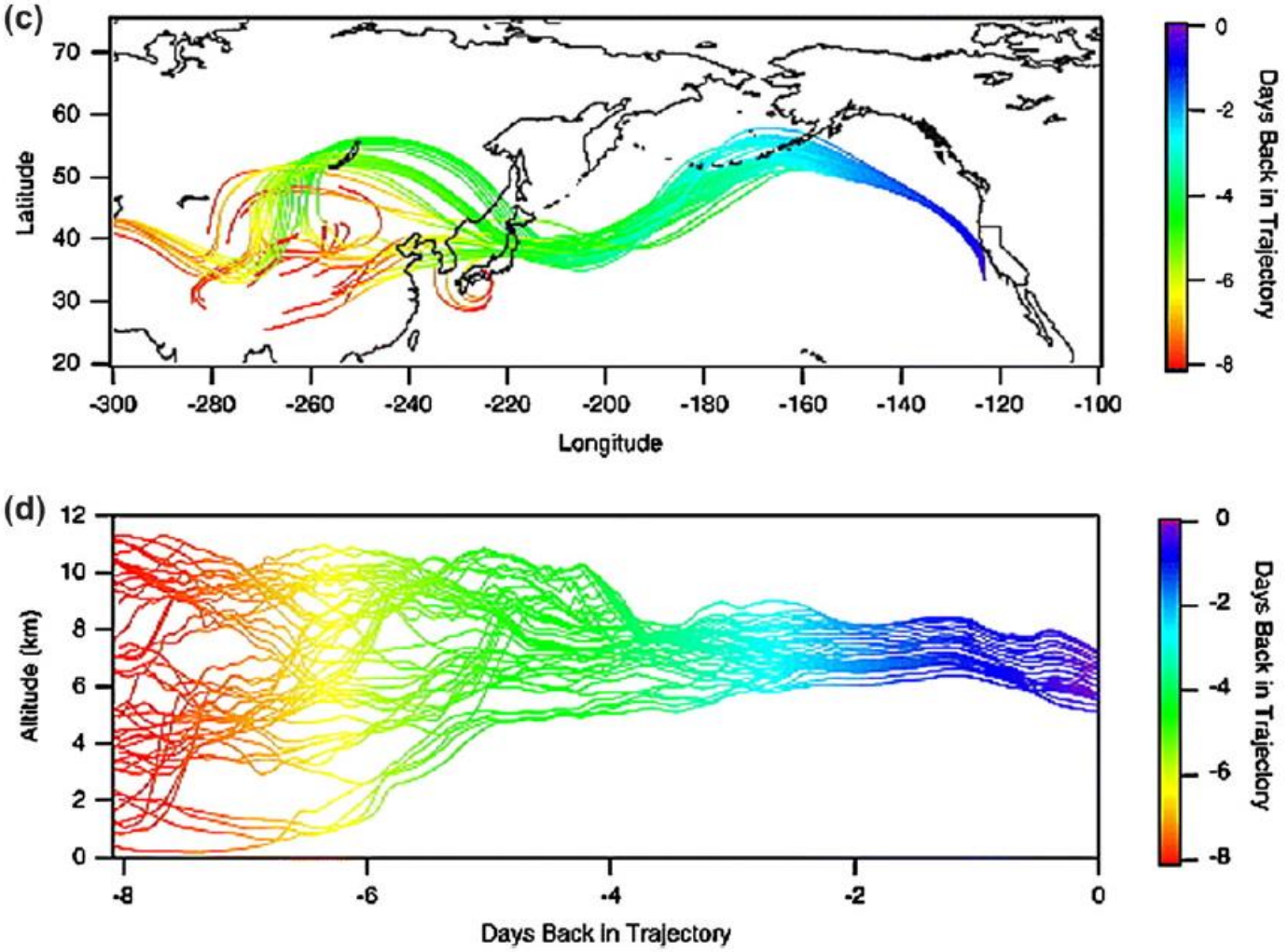
Carbon Monoxide



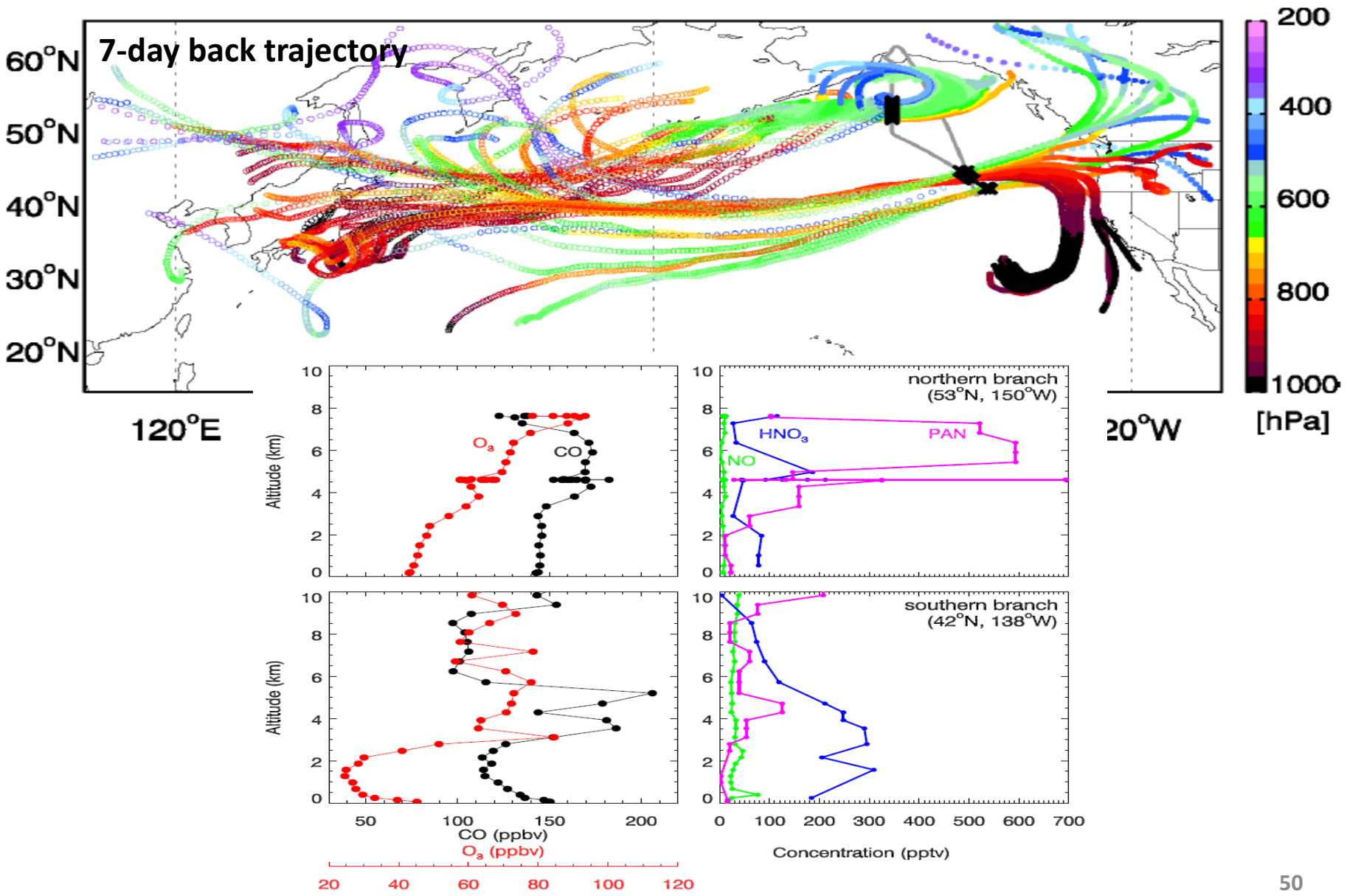
Zhang et al., 2008, ACP

Asian Influence: Back Trajectory Analysis

Ozone



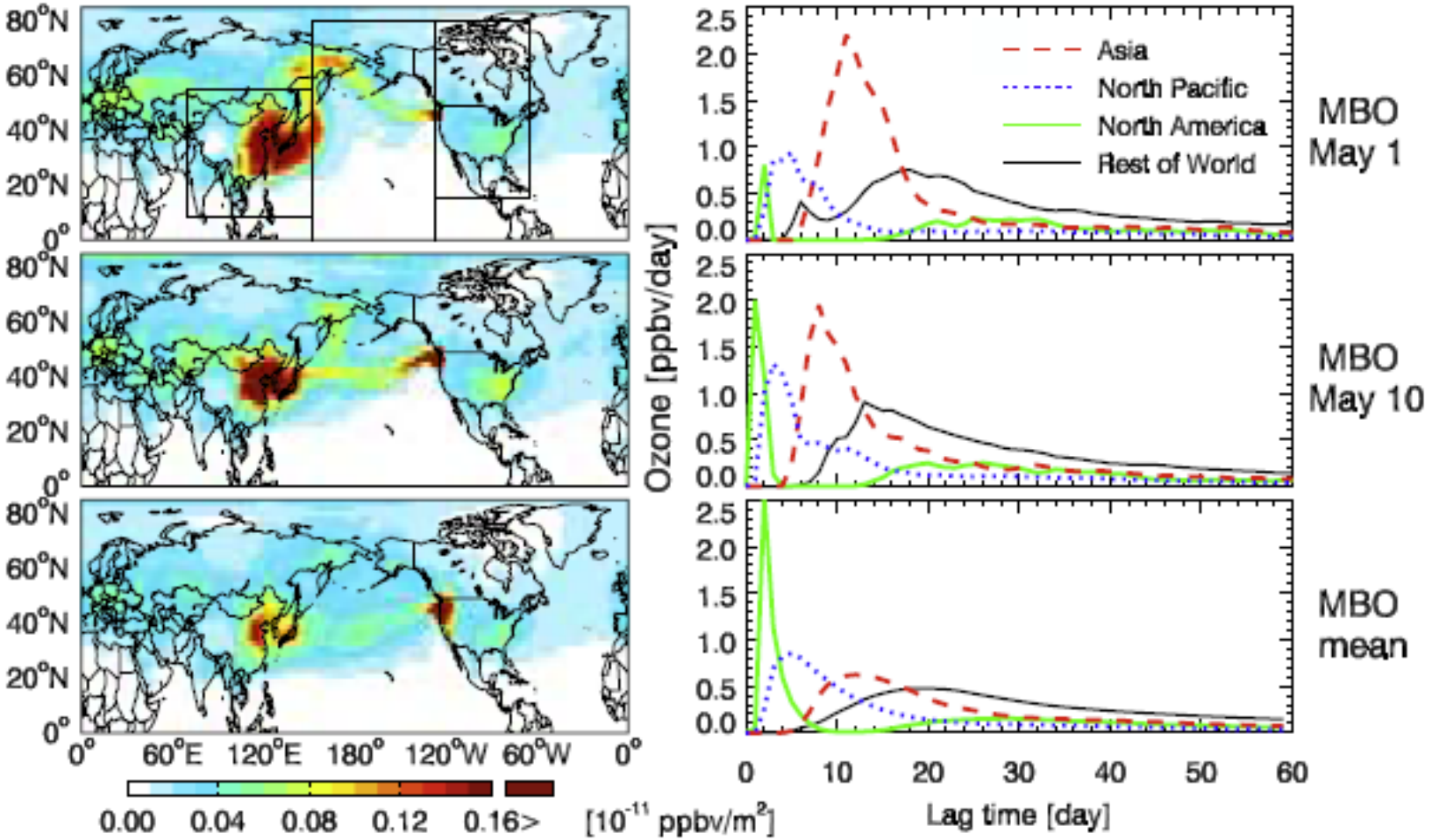
Trans-Pacific Transport and Transformation



Zhang et al., 2008, ACP

Adjoint Modeling for Intercontinental Transport

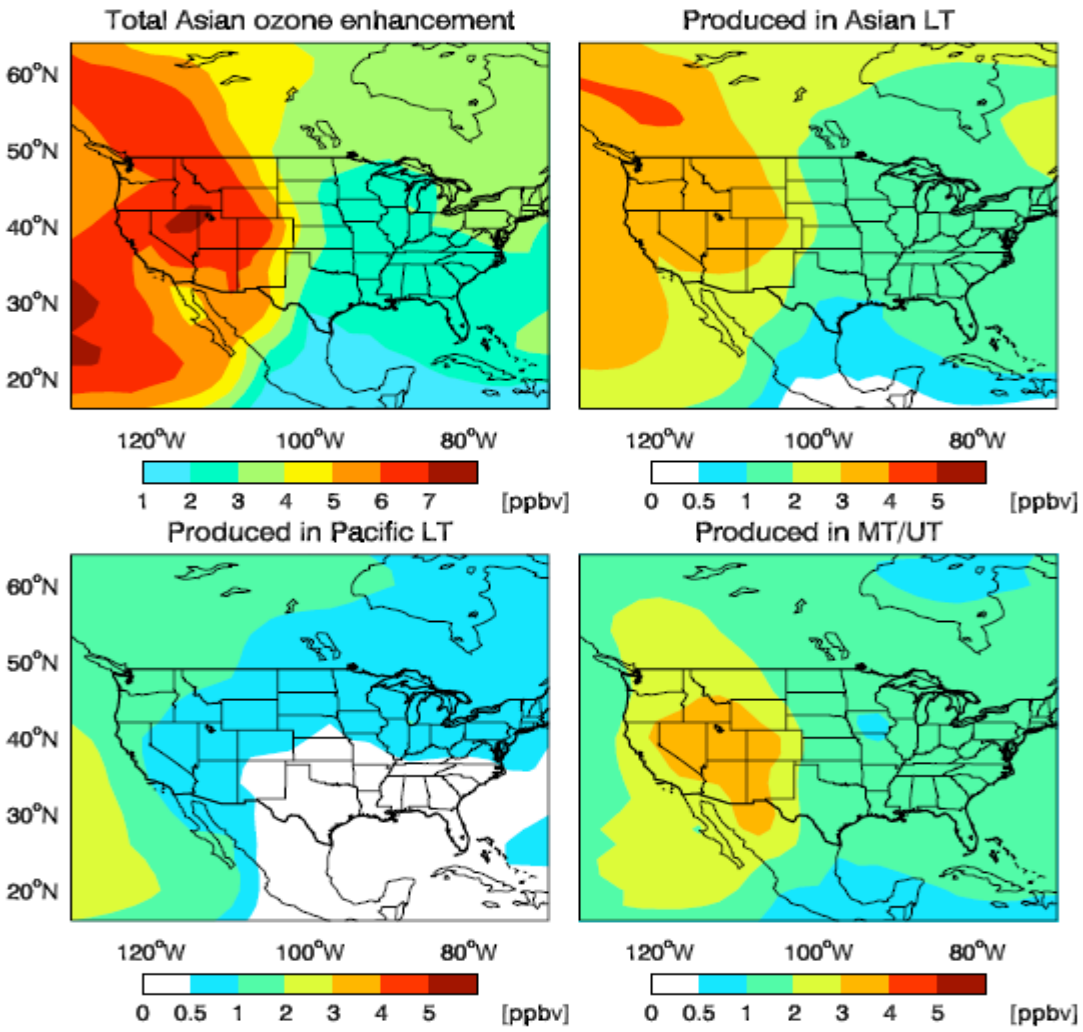
Ozone



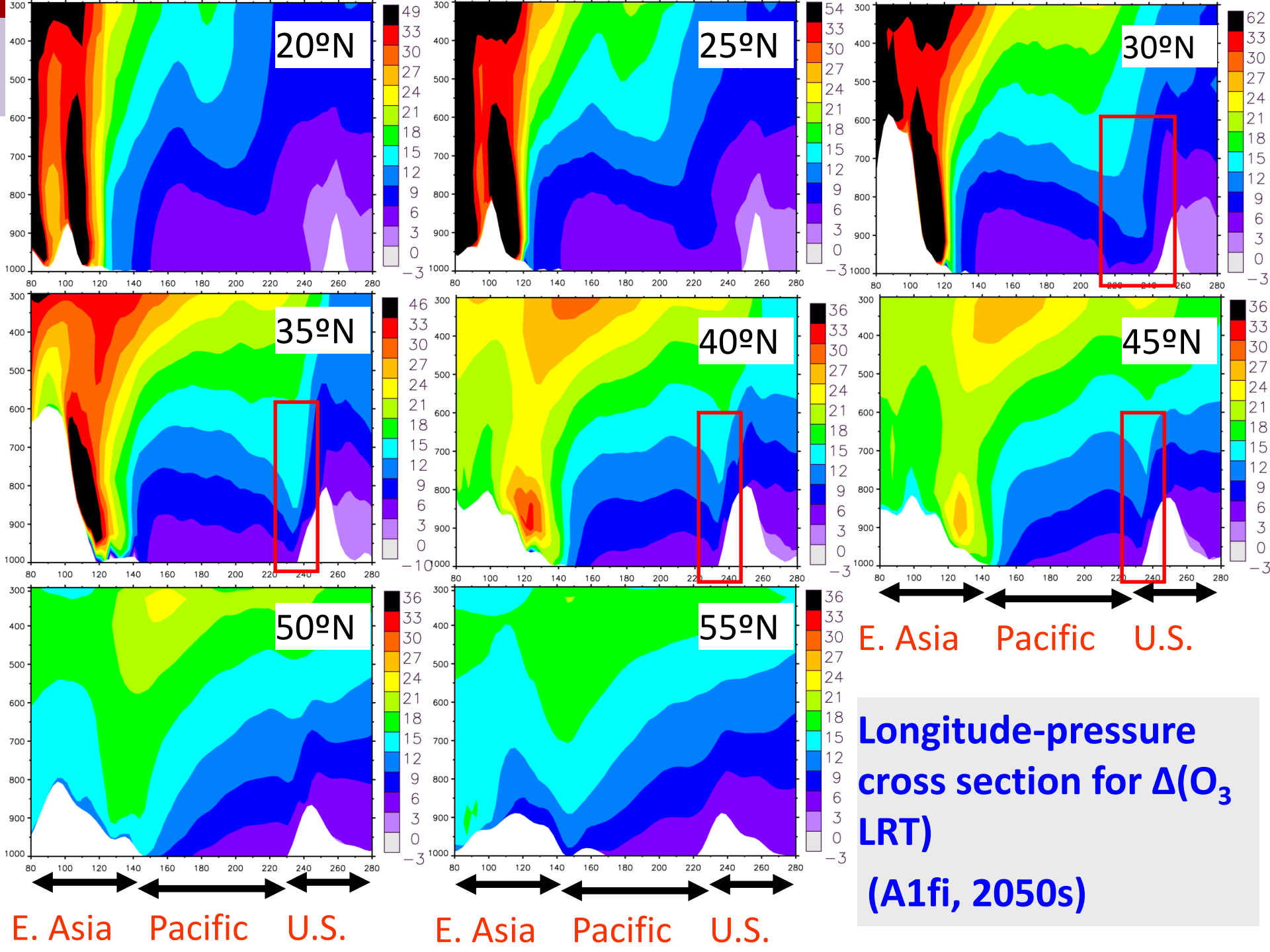
Zhang et al., 2010, GRL

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

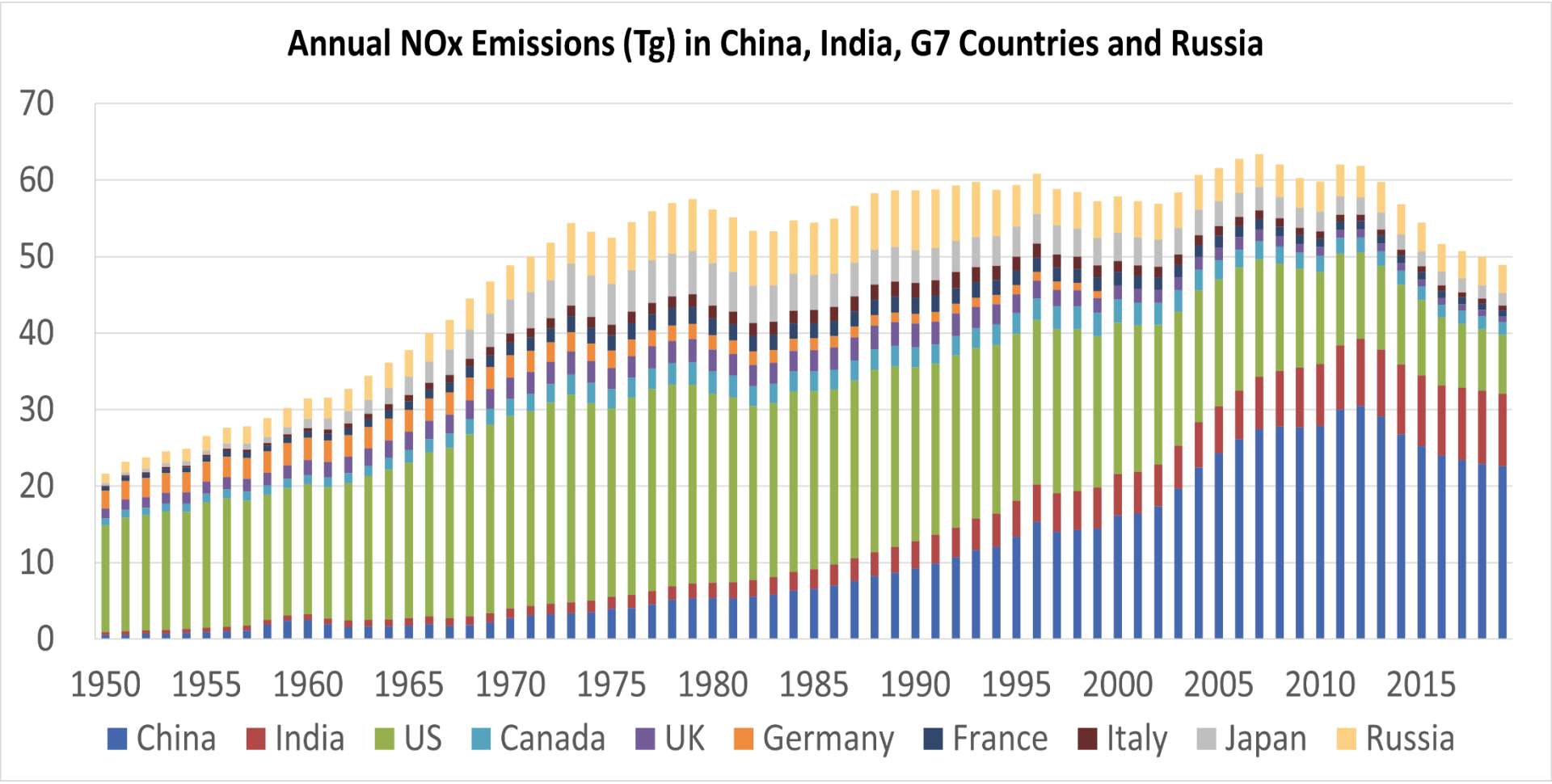
Springtime U.S. O_3 Enhancement due to Transpacific Transport



Zhang et al., 2008, ACP

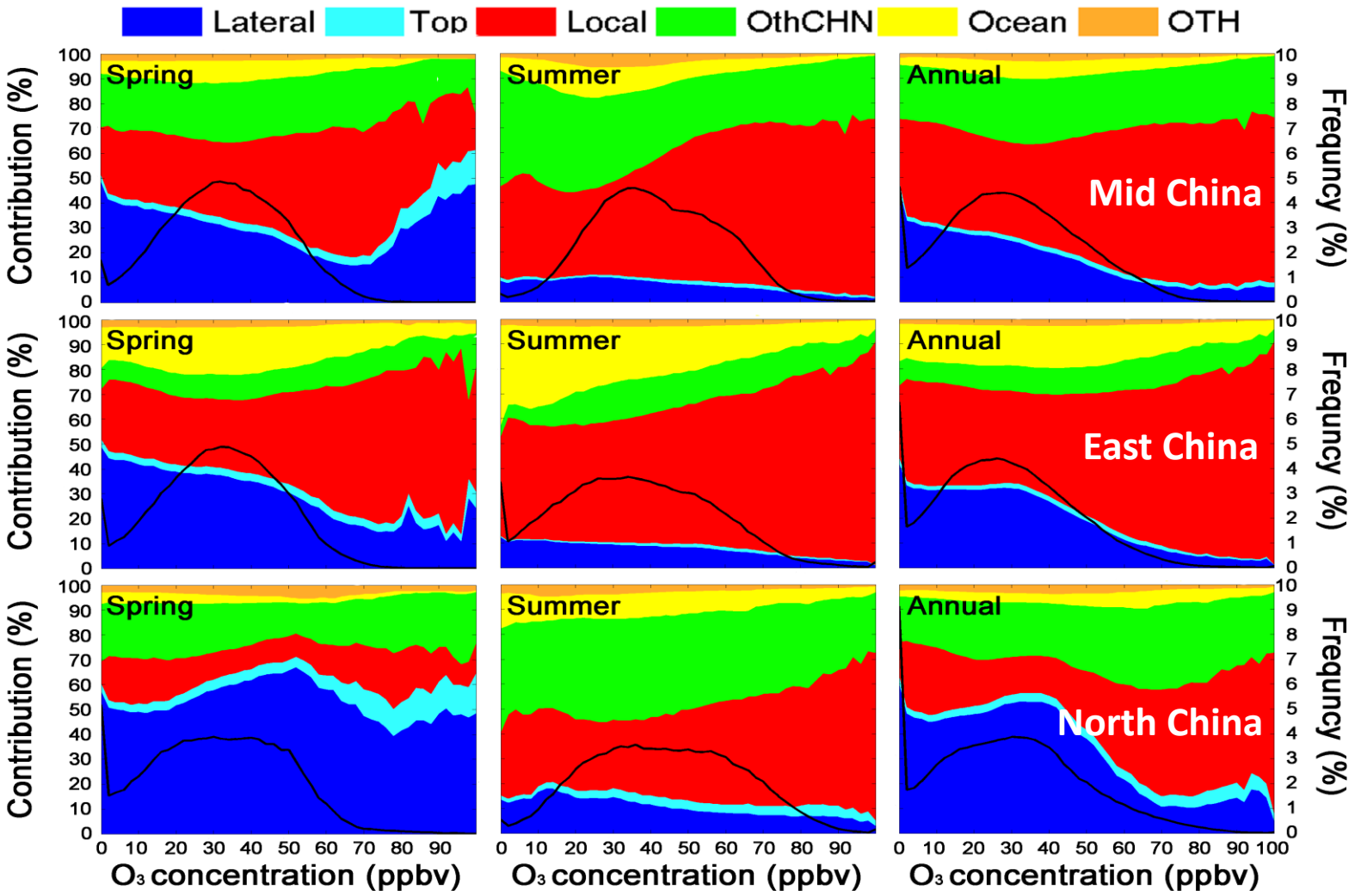

20ºN
25ºN
30ºN
35ºN
40ºN
45ºN
50ºN
55ºN
E. Asia
Pacific
U.S.
E. Asia
Pacific
U.S.
Longitude-pressure cross section for Δ(O3 LRT)
(A1fi, 2050s)

Anthropogenic Emissions of NOx: 1950-2019



CEDS v2 inventory

Strong Inflow of Ozone into Eastern China

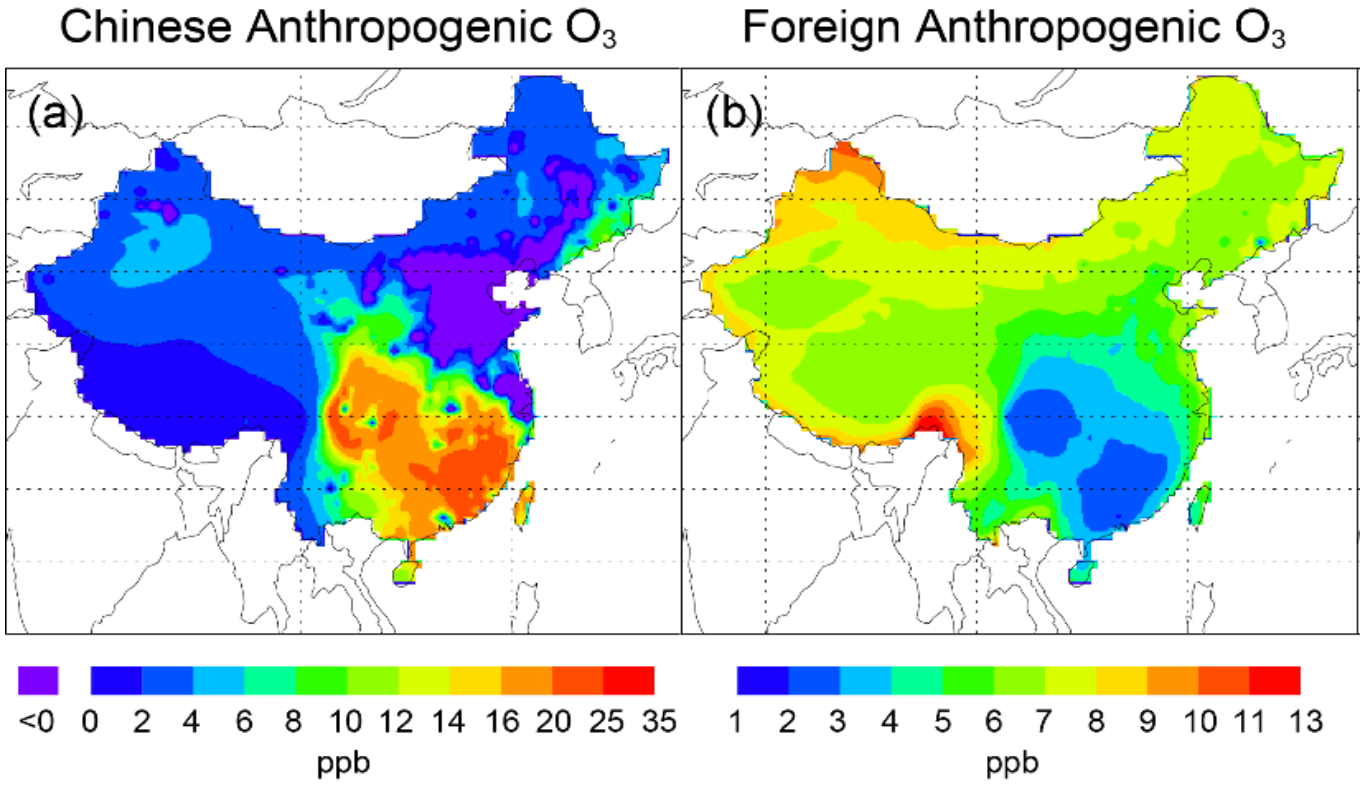


For 2010

Li et al., 2016, AR

Foreign Pollution Greatly Affect China's O_3

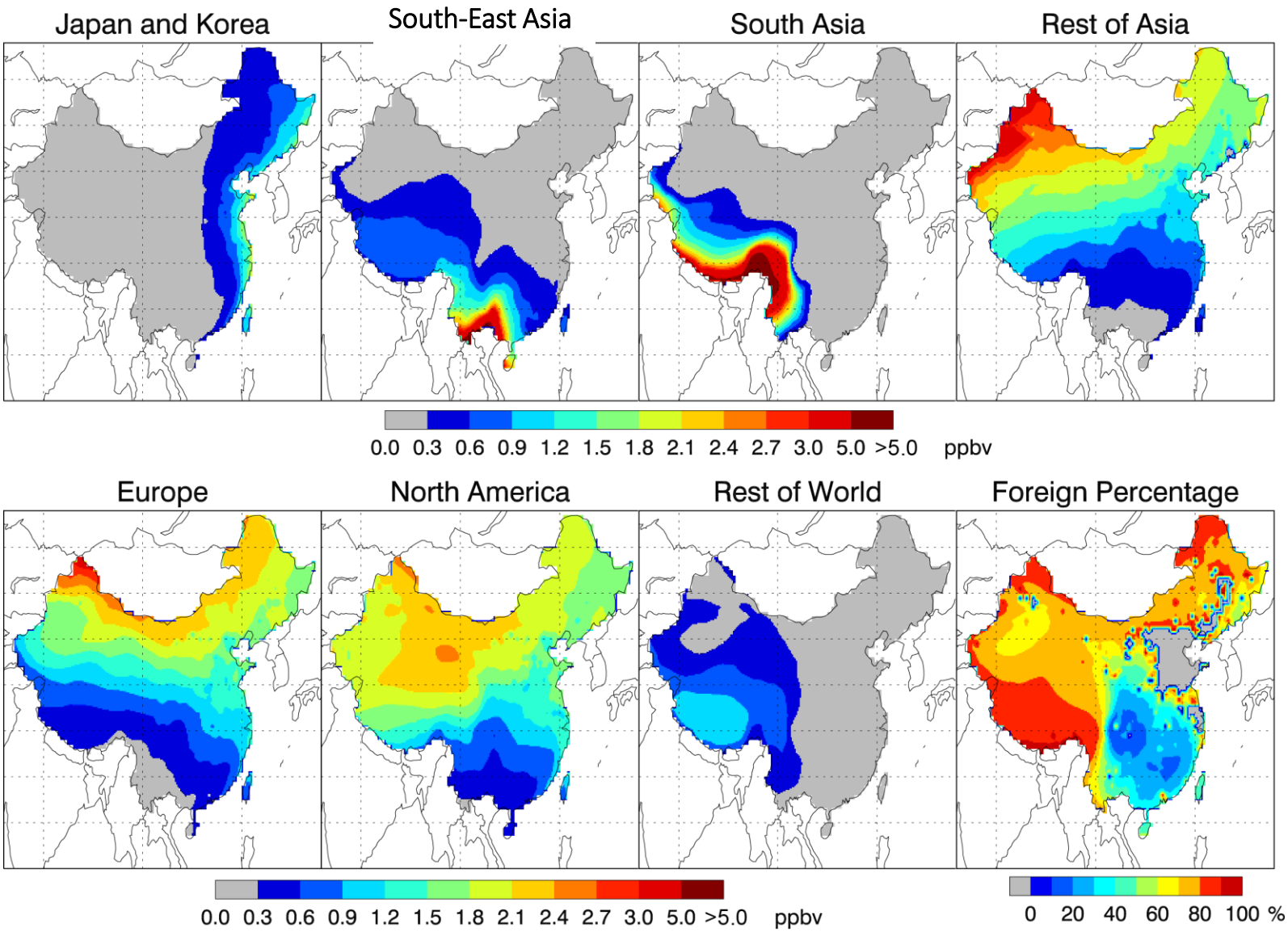
- **2–11 ppb of Surface O_3 over China in Spring 2008 are Foreign**



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

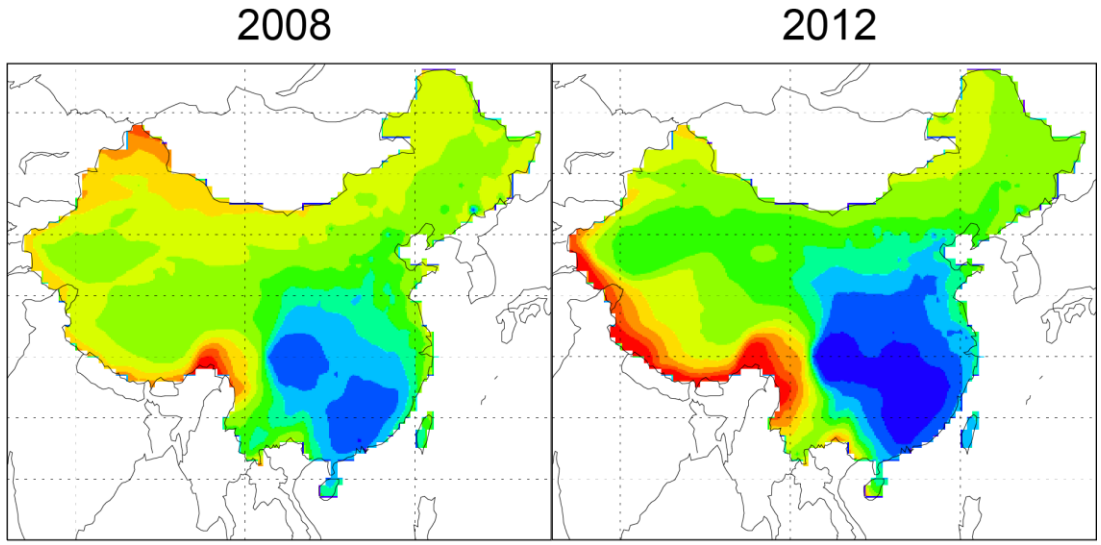
Ni et al., ACP, 2018

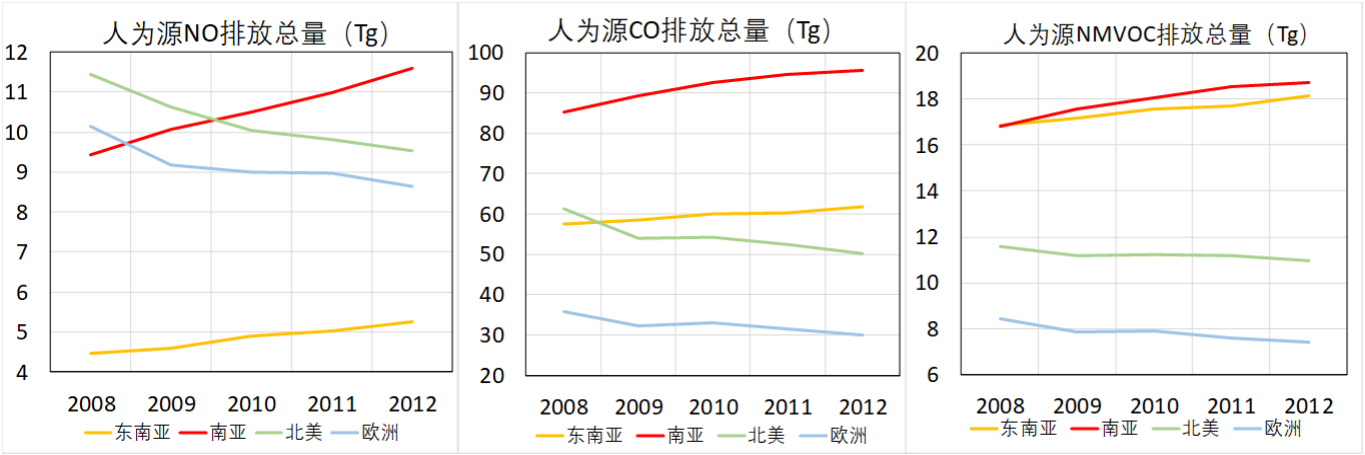
Foreign Pollution Greatly Affect China's O_3: Spring 2008



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

Changes in Springtime Foreign Anthropogenic Surface O_3

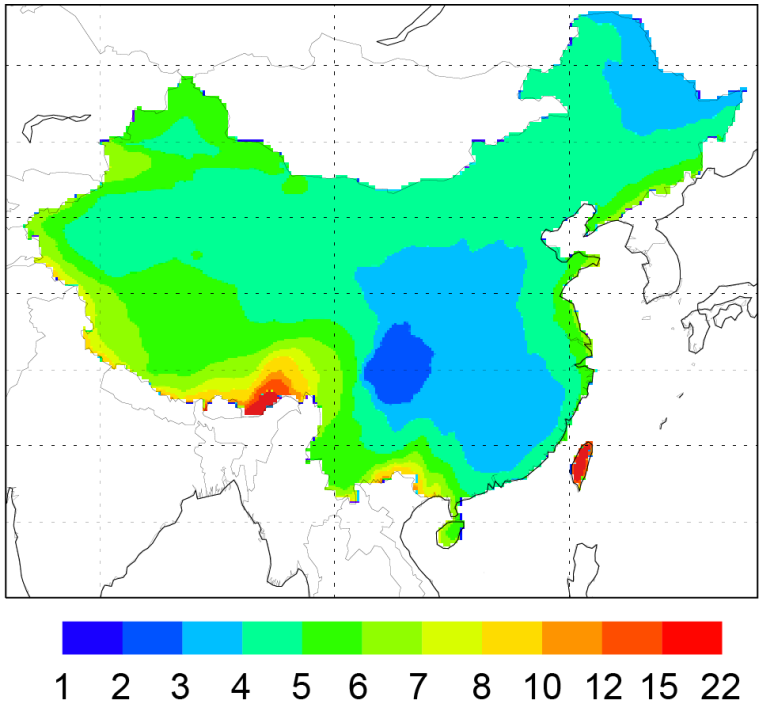




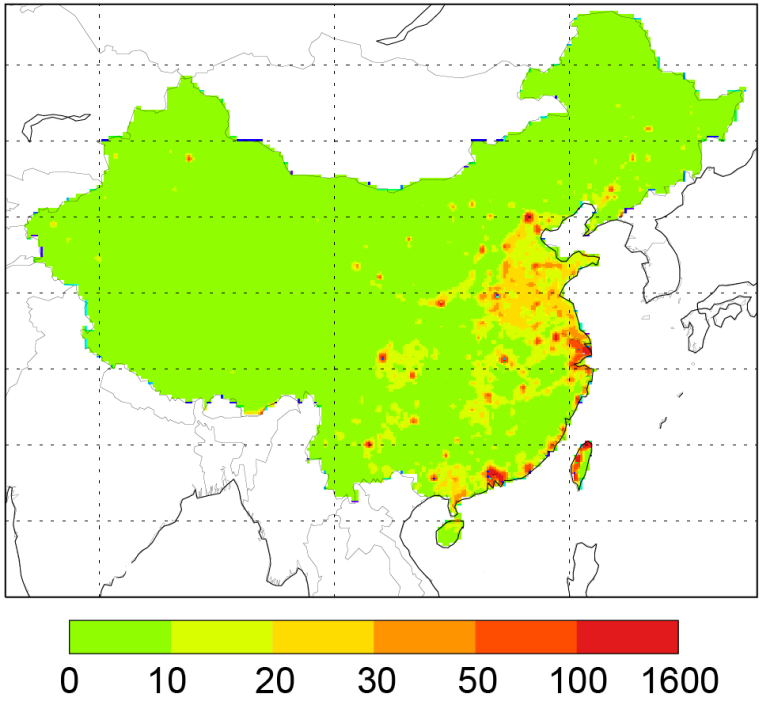
Ni et al., ACP, 2018

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

Transboundary MDA8 Ozone in 2015



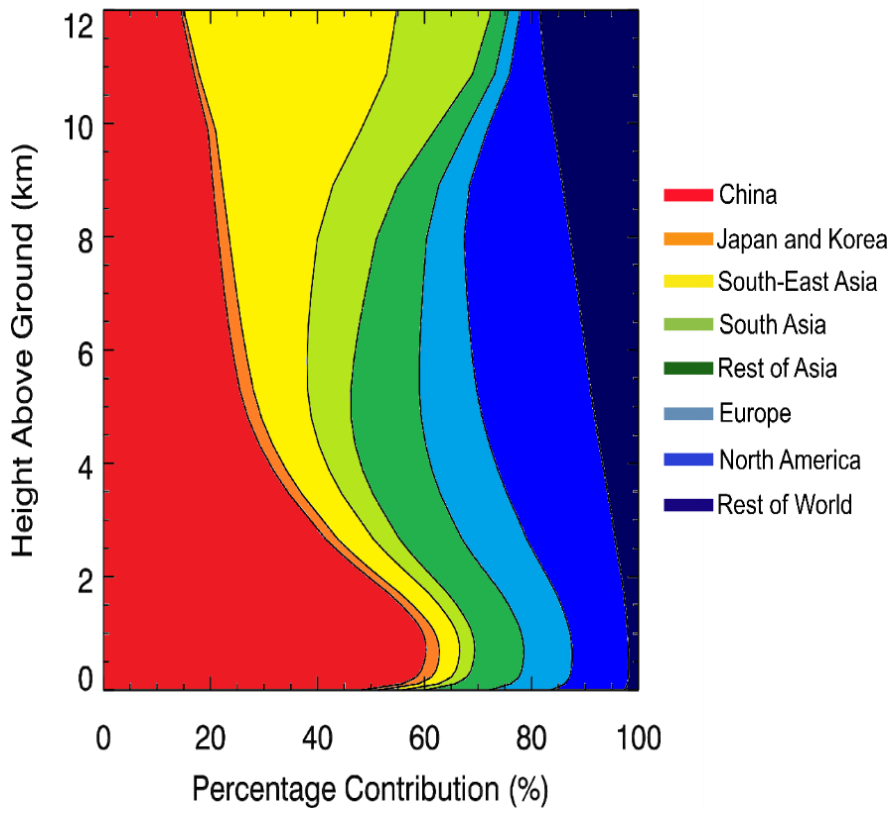
Deaths wrt transboundary O_3



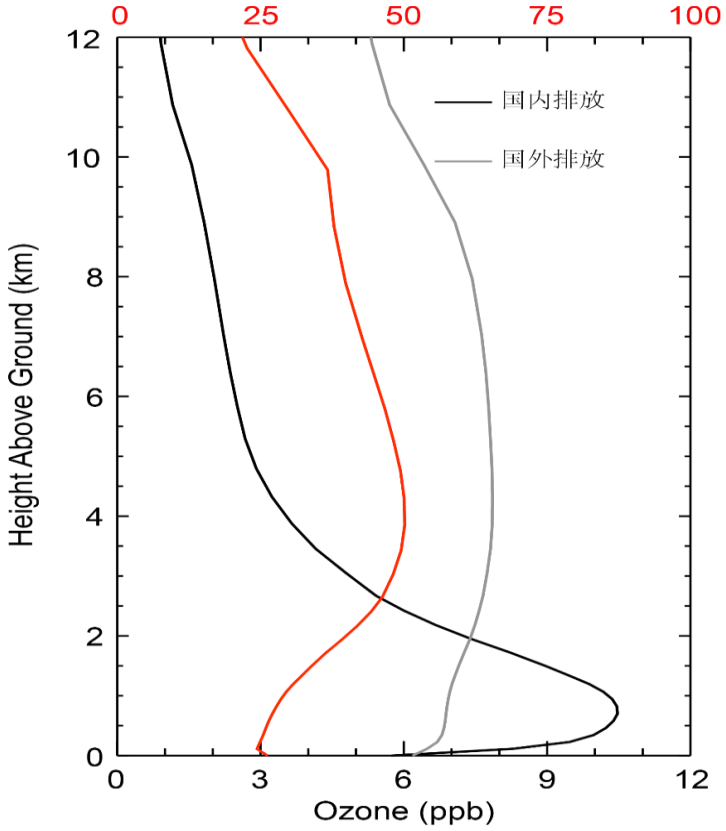
Ni et al., in prep

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



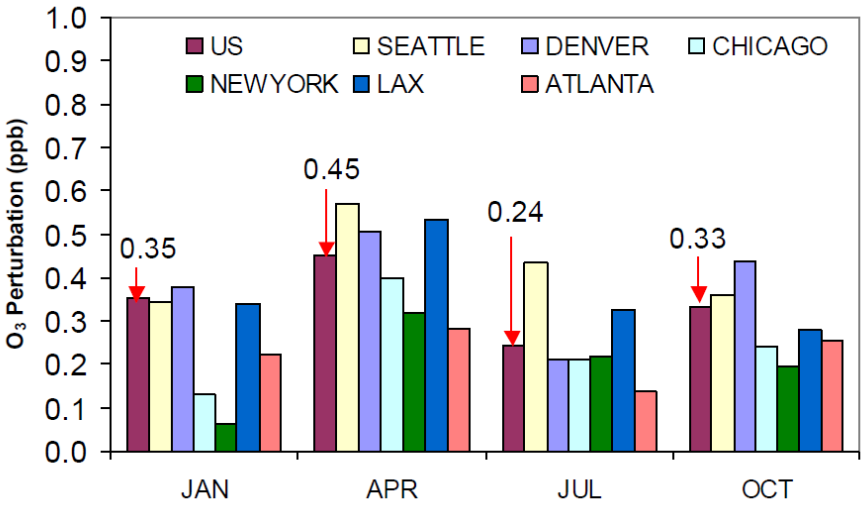
Method: *Zero-out* + *Tagged O_3* + *Linear weighting*

Ni et al., ACP, 2018

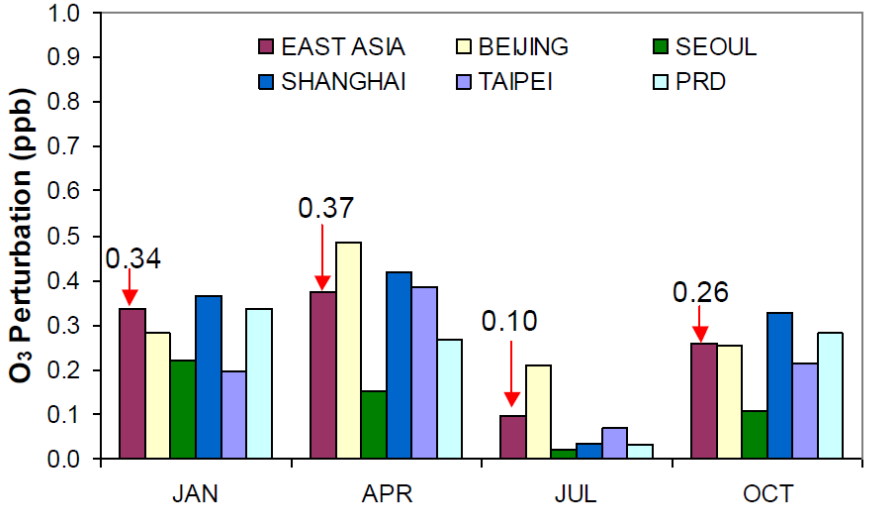
Transboundary Ozone from E. Asia versus NA

WRF-Chem simulation at 36 x 36 km^2

20% changes in anthro emis in E. Asia



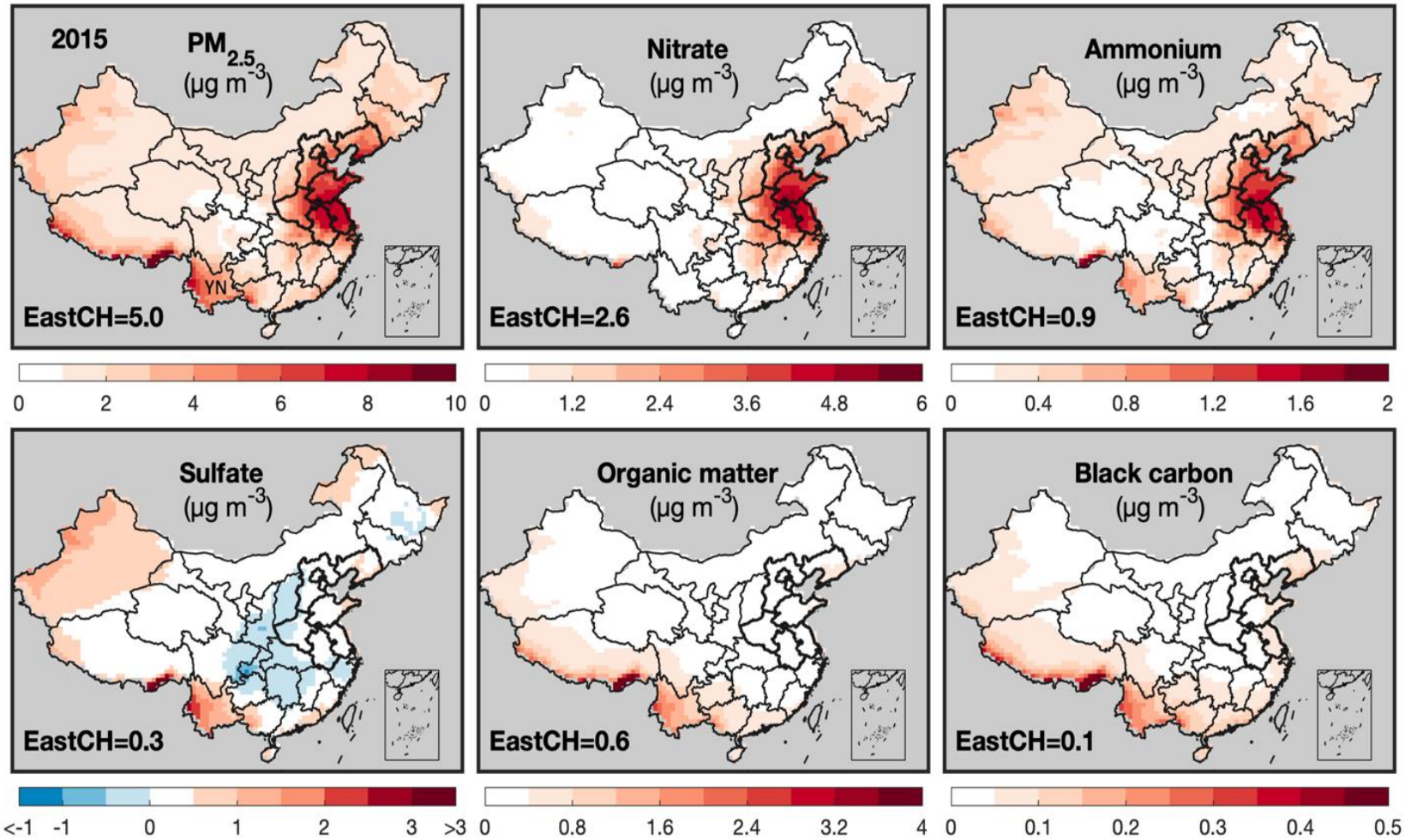
20% changes in anthro emis in NA



HTAP, 2010 (P206)

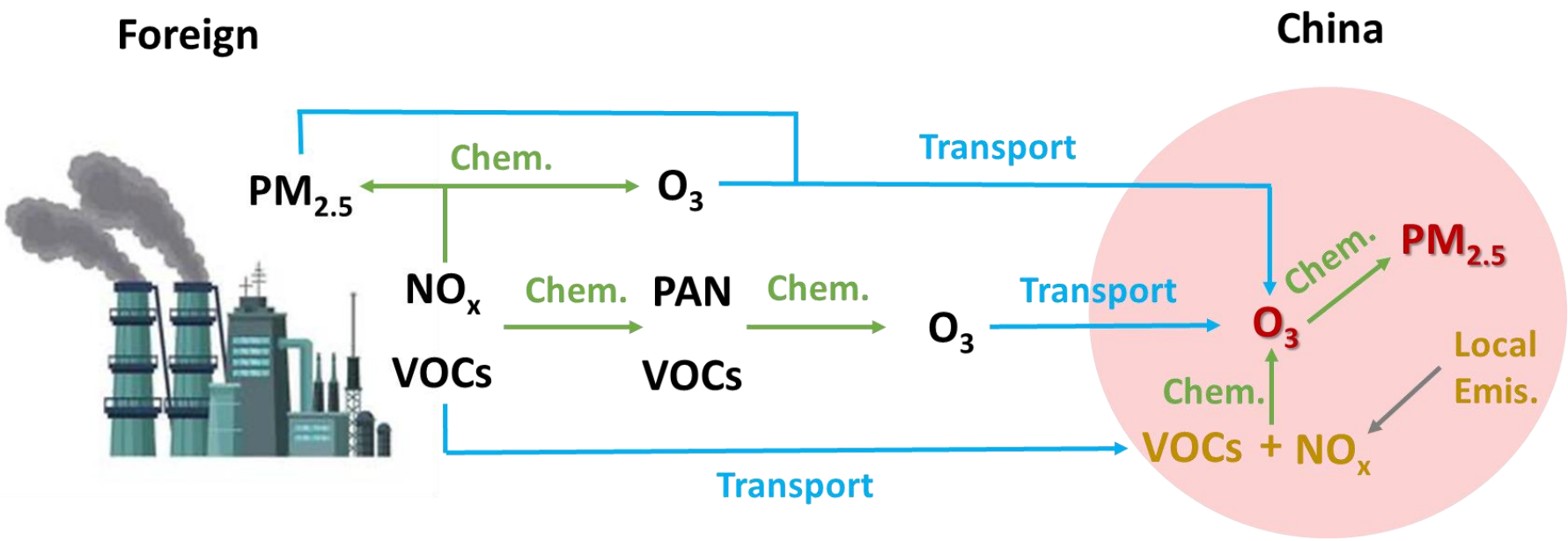
Foreign Pollution Transport Worsens Chinese $PM_{2.5}$

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



Xu et al., ACP, 2023, Highlight Paper

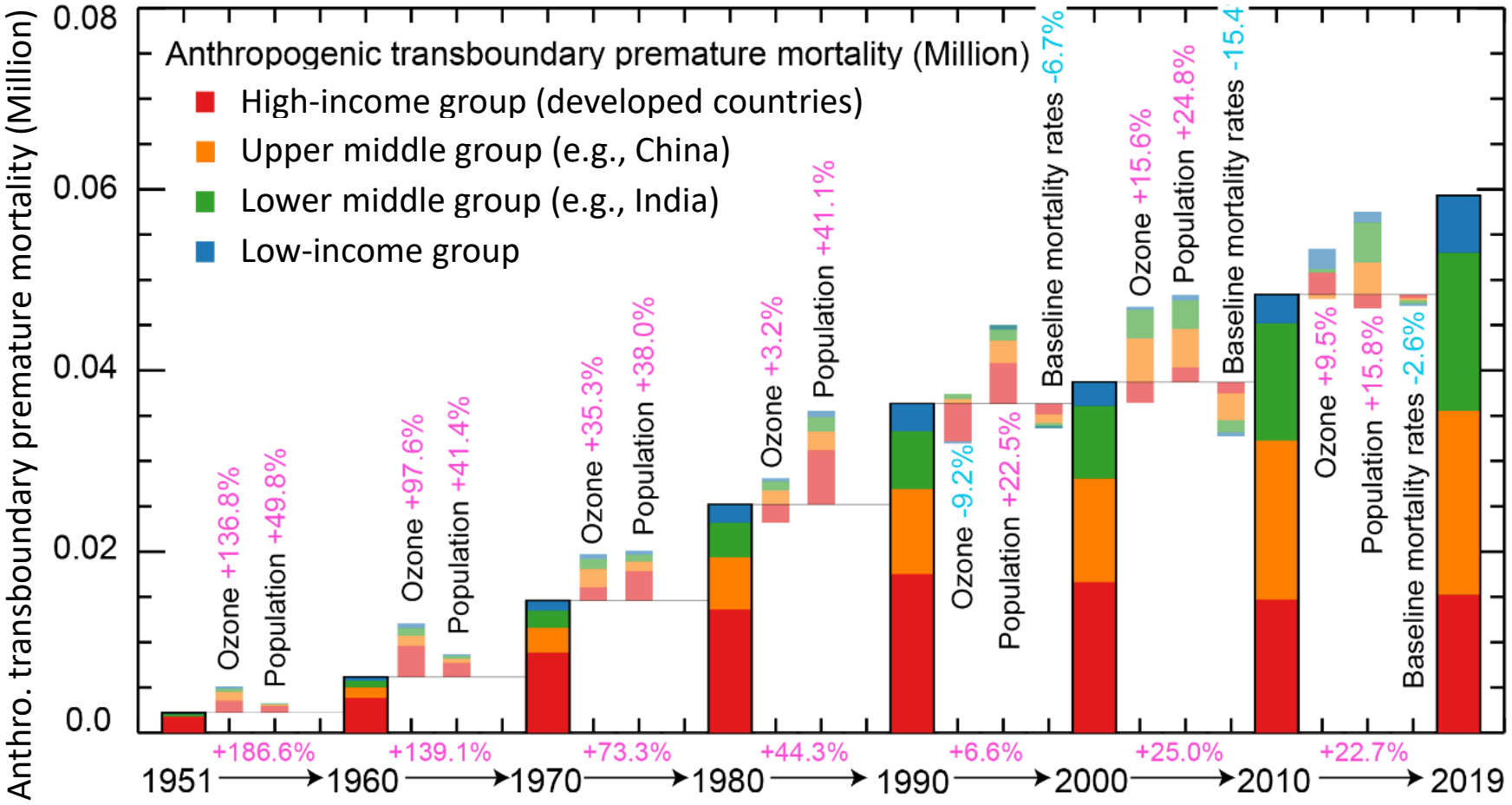
Complex Chemical-Transport Mechanism



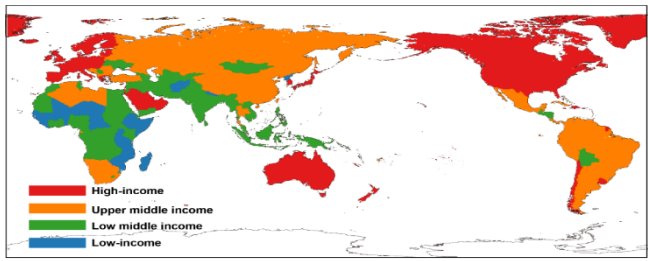
Transboundary pollution mechanisms:

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

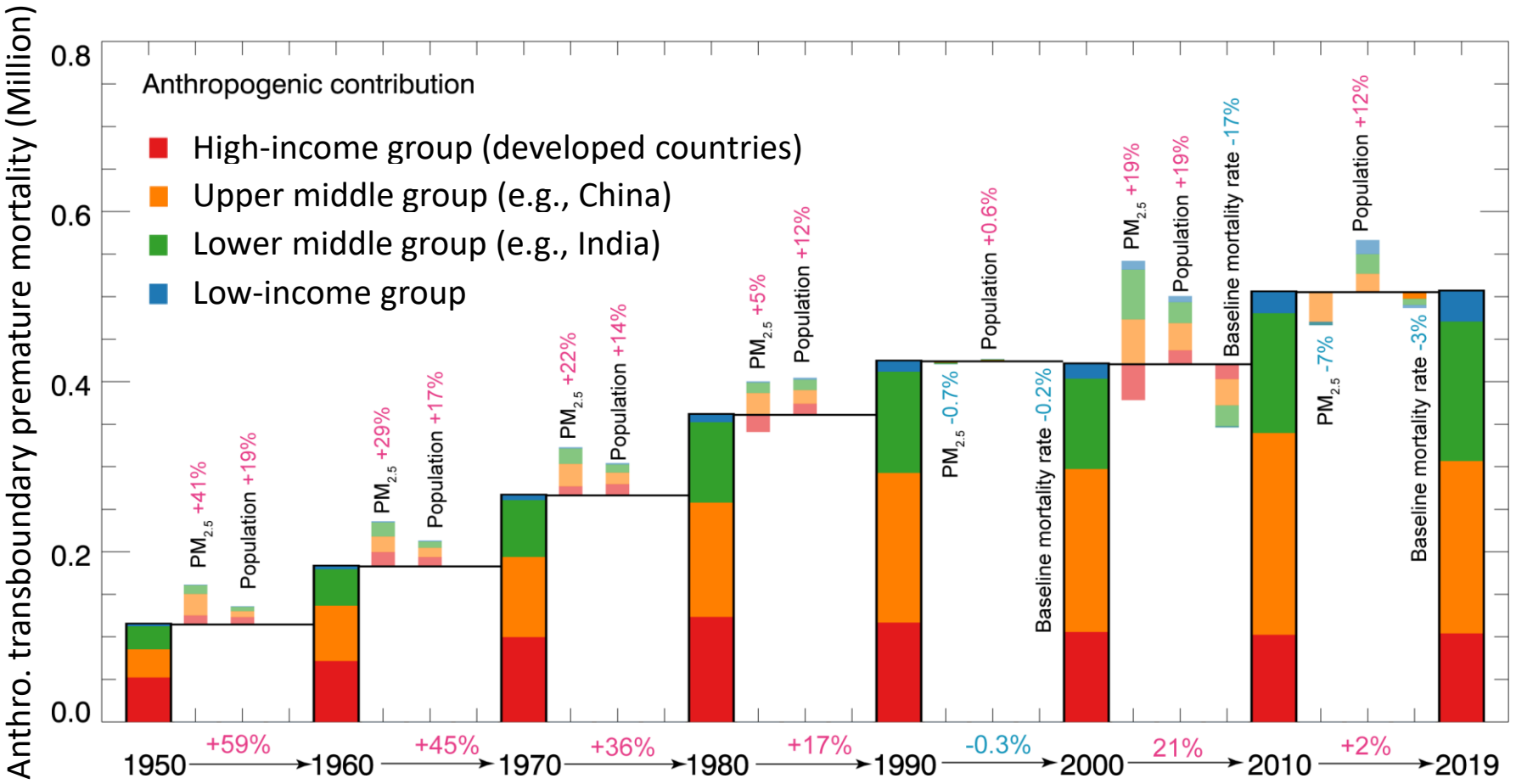
Historical Transboundary Ozone Mortality via Atmospheric Transport



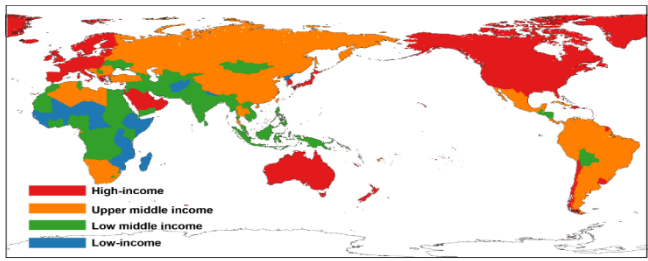
Chen et al., ERL, 2023



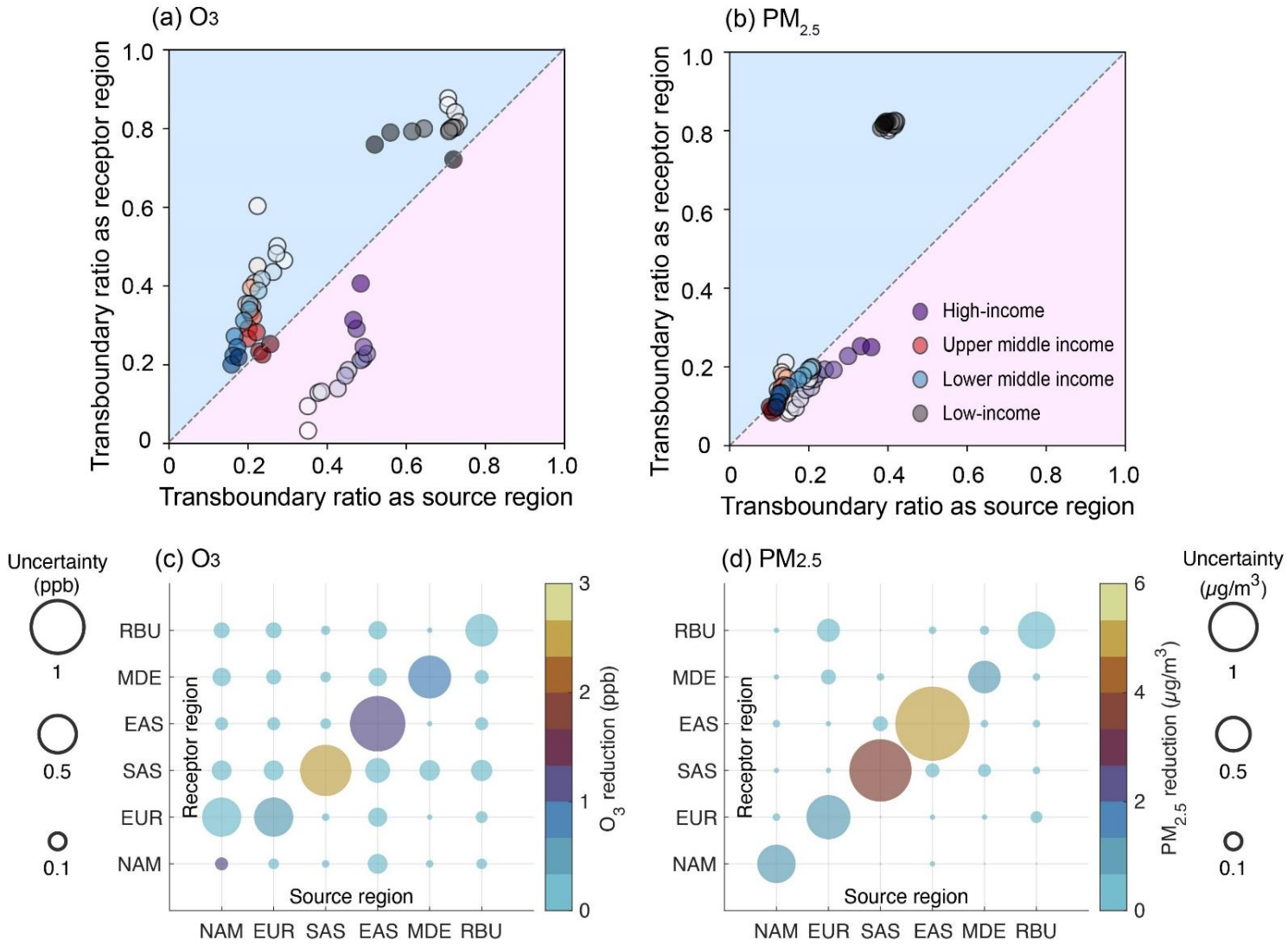
Considerable Historical Global Deaths Due to Transported $PM_{2.5}$



Chen et al., Science Bulletin, 2022



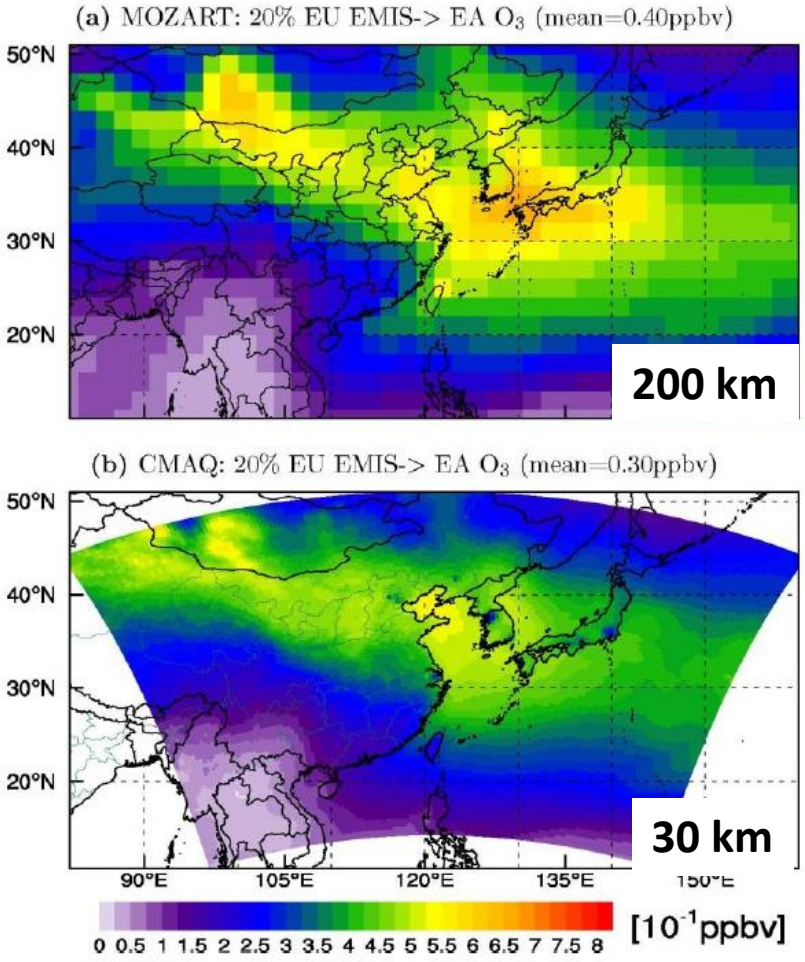
Historical Transboundary Pollution via Atmospheric Transport



Lin et al., under review

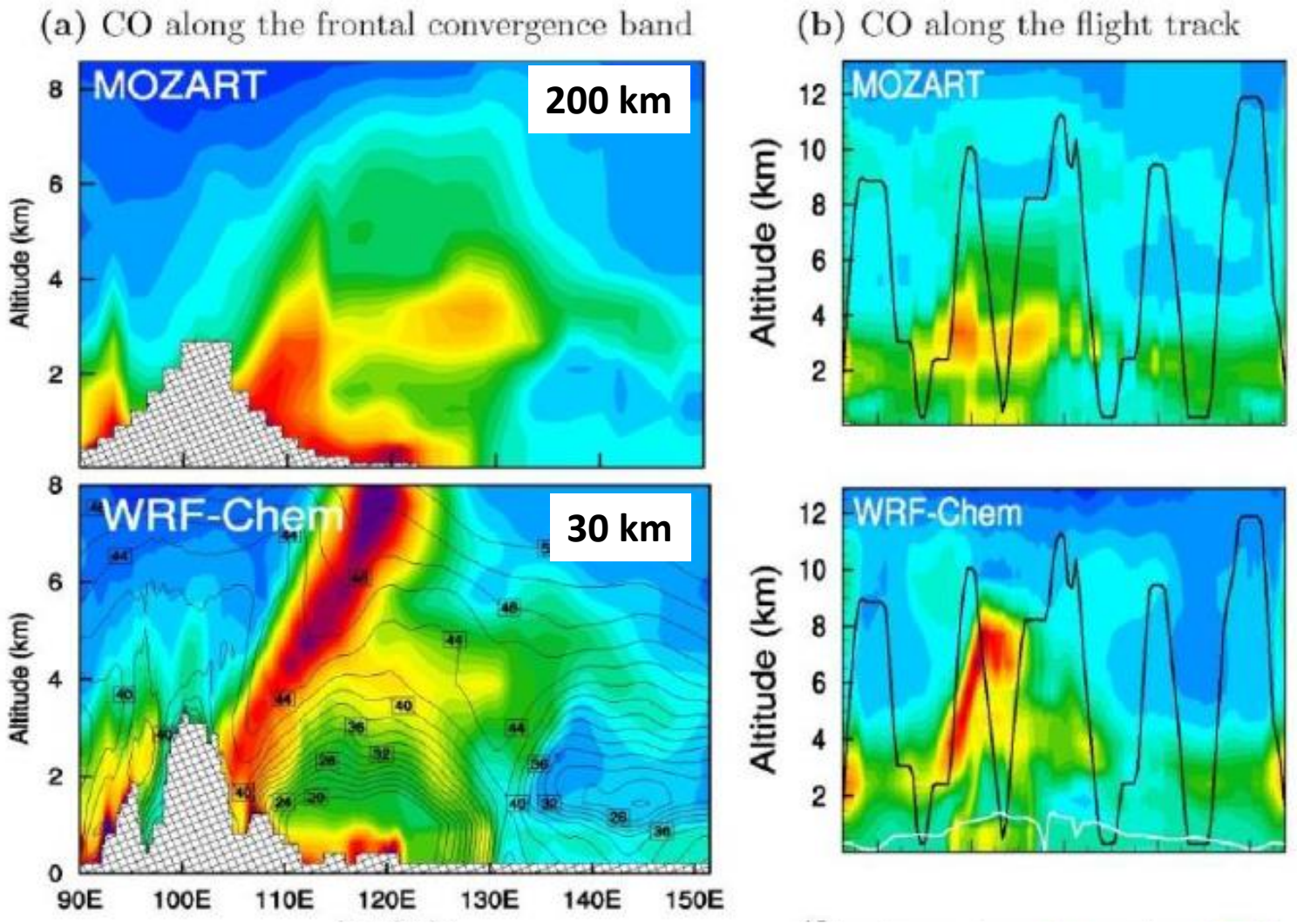
Uncertainty in Import Due to Model Resolution

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



Lin et al., 2010, ACP

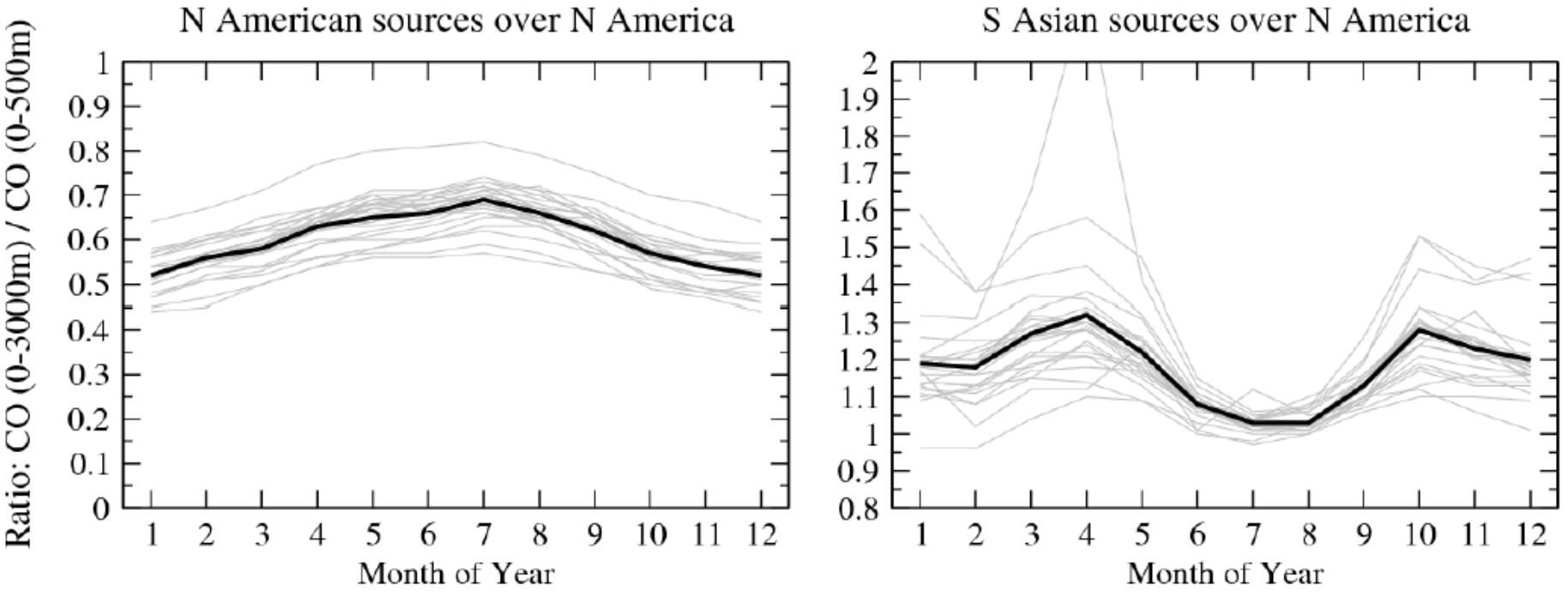
Uncertainty in Export Due to Model Resolution



March 2001

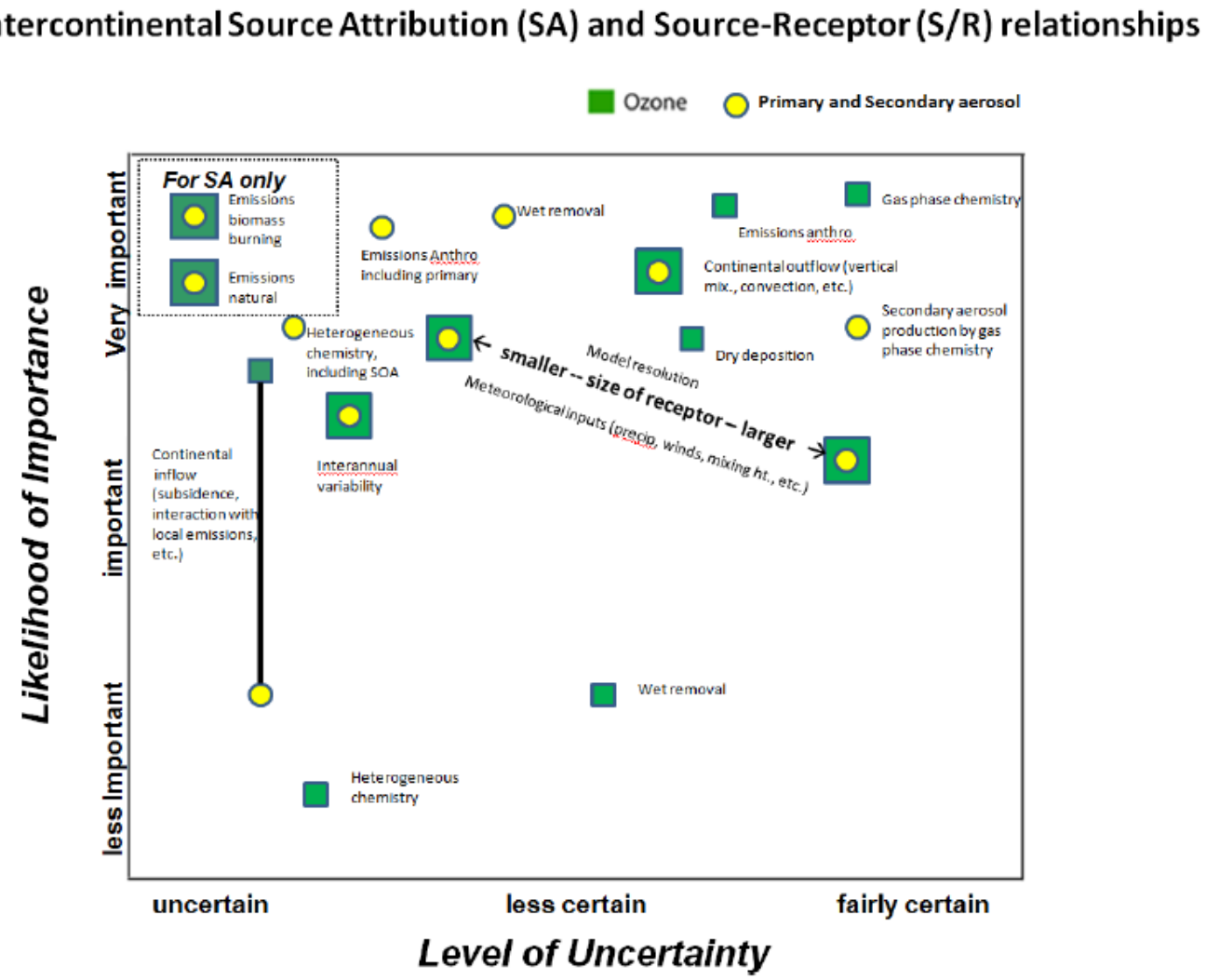
Lin et al., 2010, ACP

Uncertainty Due to Model Transport Process



HTAP, 2010

Uncertainties in Model Assessment



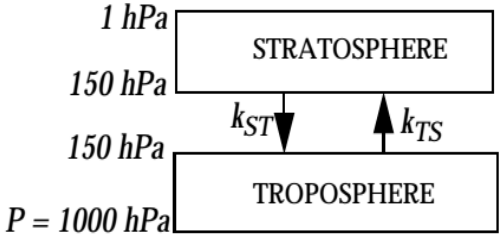
HTAP, 2010

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 $k_c = 6.45 \times 10^{-2}$ 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 τ 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 $U = 10\ \text{m s}^{-1}$.

1. What is the residence time τ_{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 τ_{chem} against chemical loss. Calculate the fraction f of the pollutant exported out of the United States box as a function of the ratio τ_{out}/τ_{chem}. Plot your result. Comment on the potential for different pollutants emitted in the United States to affect the global atmosphere.

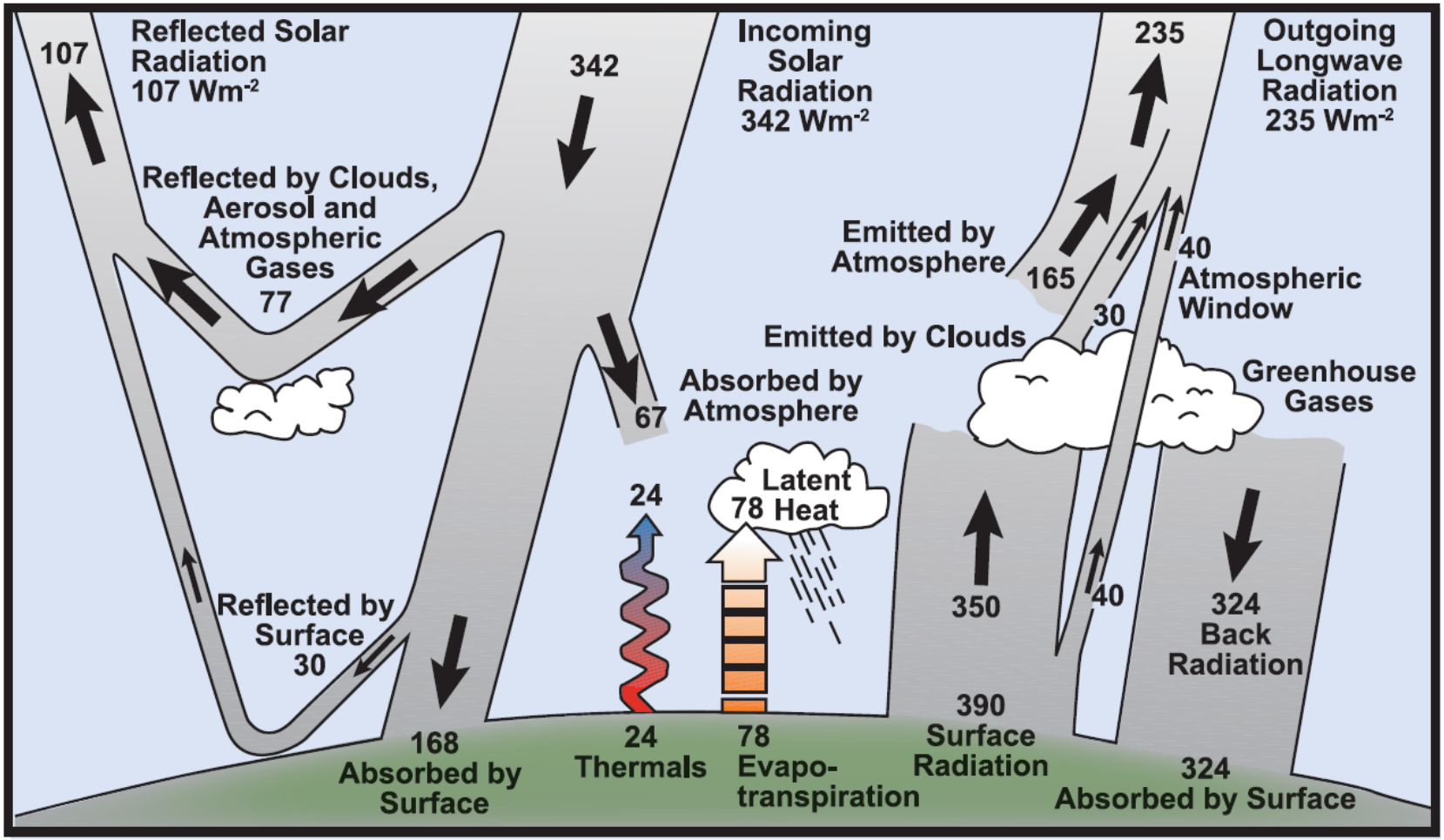
Jacob, 1999

Local Transport and Transformation

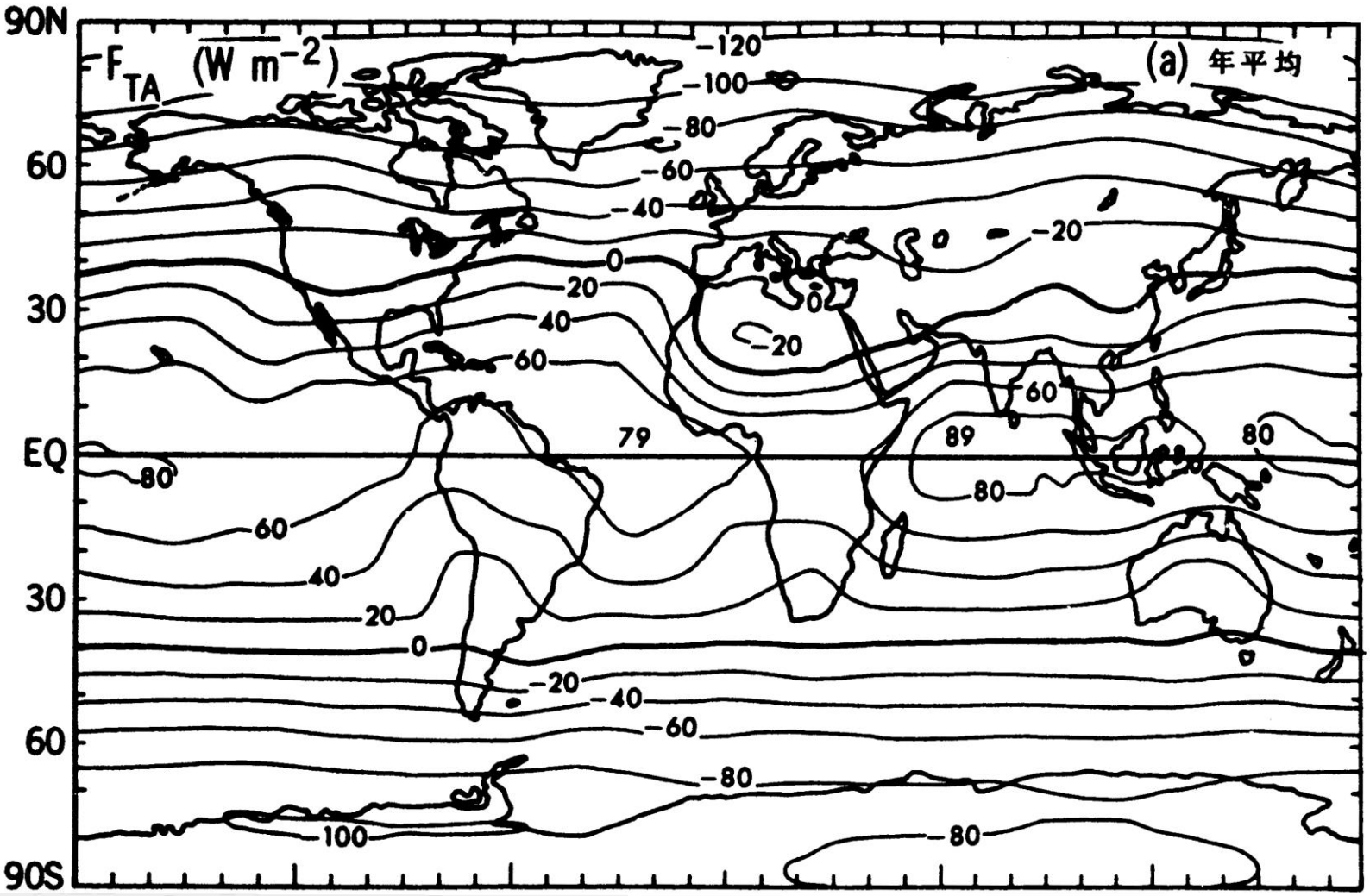
A cluster of coal-fired power plants in Ohio emits sulfur dioxide (SO_2) continuously to the atmosphere. The pollution plume is advected to the northeast with a constant wind speed $U = 5$ m s^{-1}. We assume no dilution of the plume during transport. Let $[SO_2]_o$ be the concentration of SO_2 in the 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

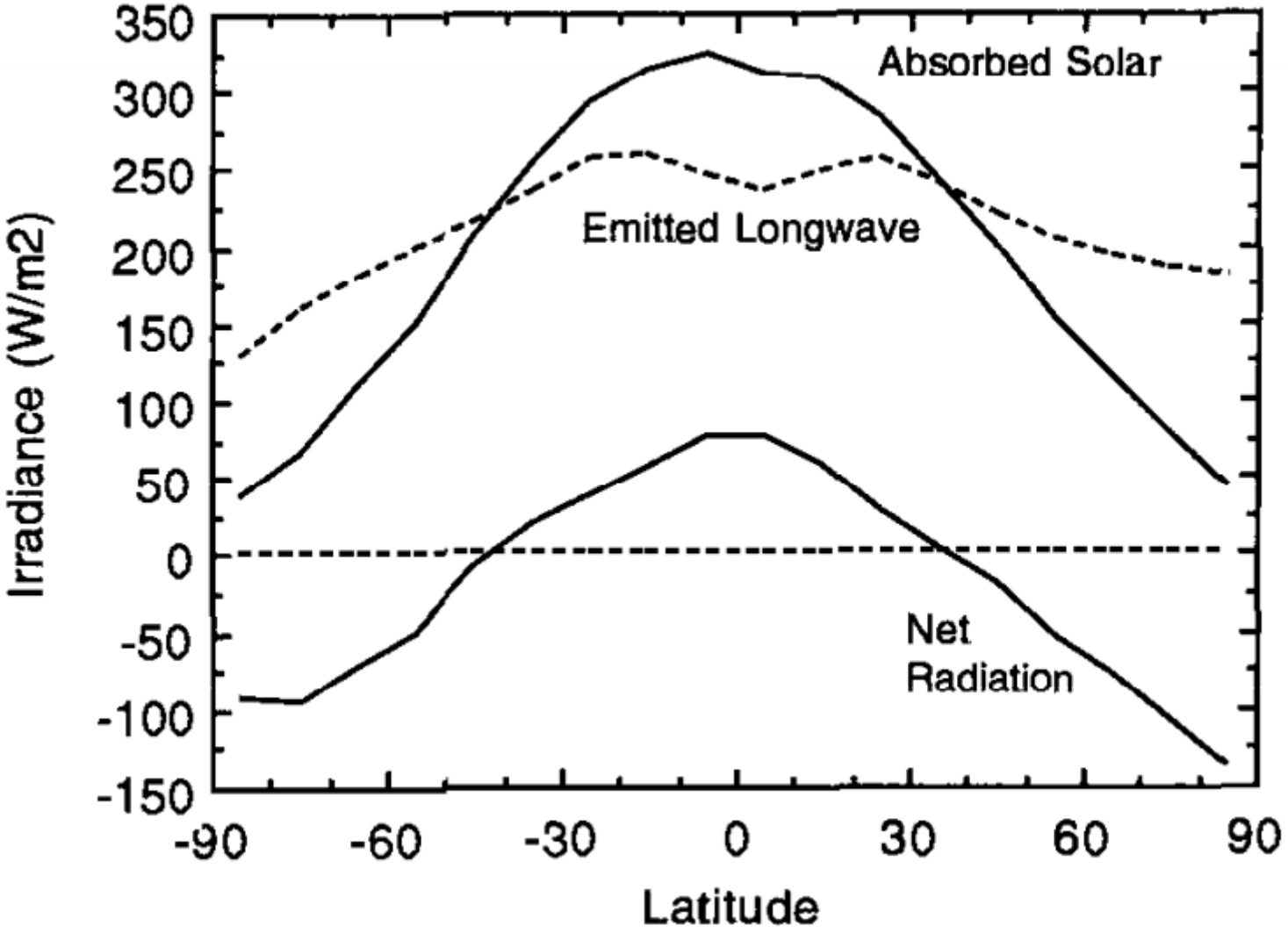
Energy Budget of Earth Climate



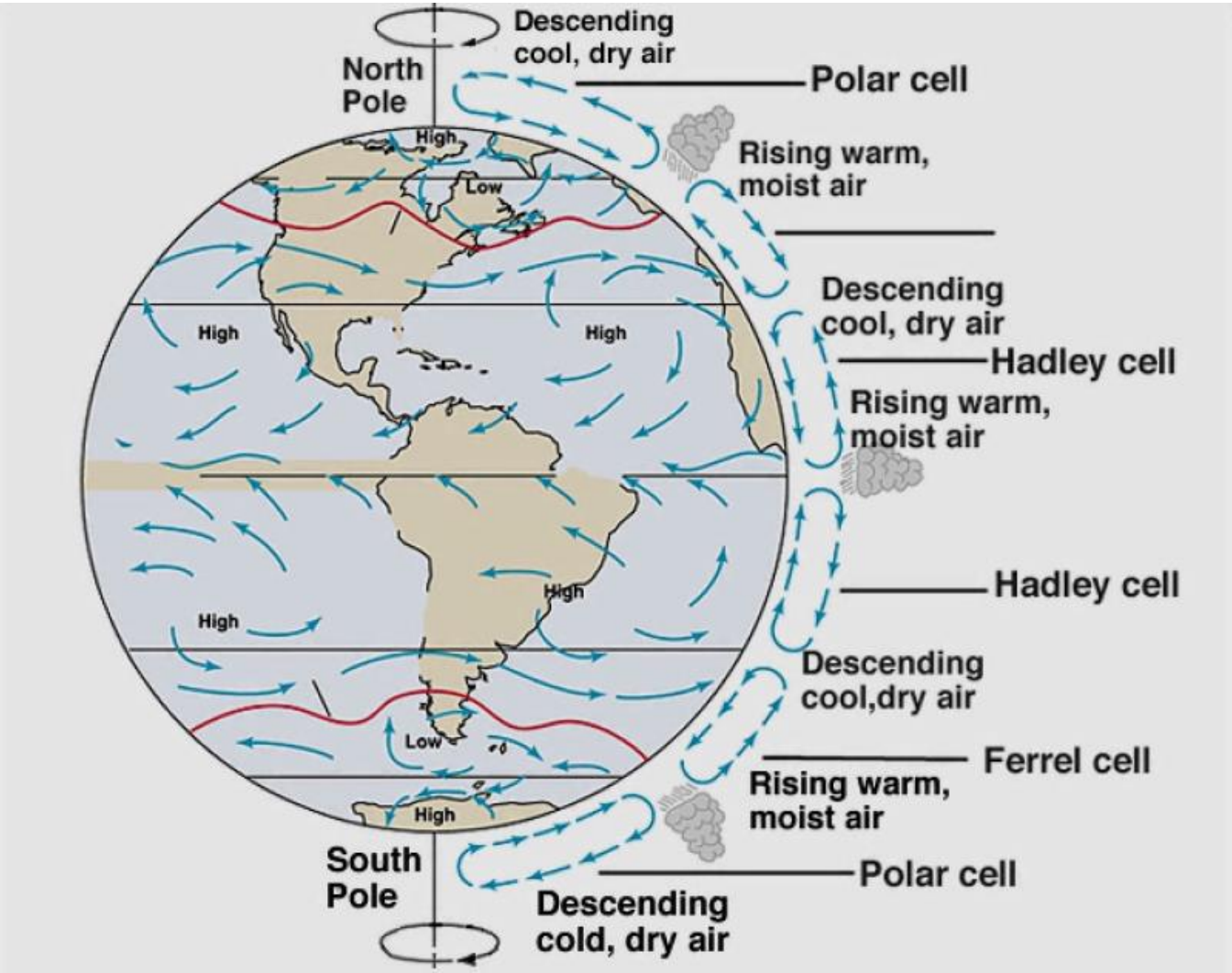
Annual Mean TOA Net Radiation Flux (Solar + Thermal)



Annual Mean TOA Net Radiation Flux (Solar + Thermal)



General Circulation



Kinematics

From Stokes' Theorem:

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

From Gauss's Theorem:

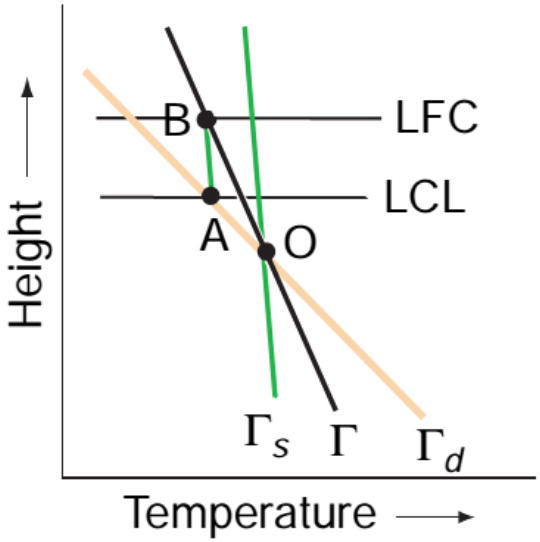
$$\oint V_n ds = \int\int \mathrm{Div}_H \, \mathbf{V} dA$$

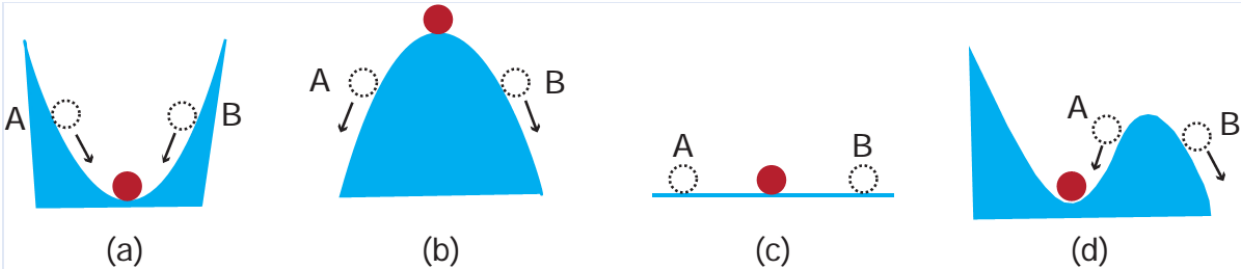
Wallace and Hobbs, 2006

Conditional and Convective Instability

Conditionally unstable:

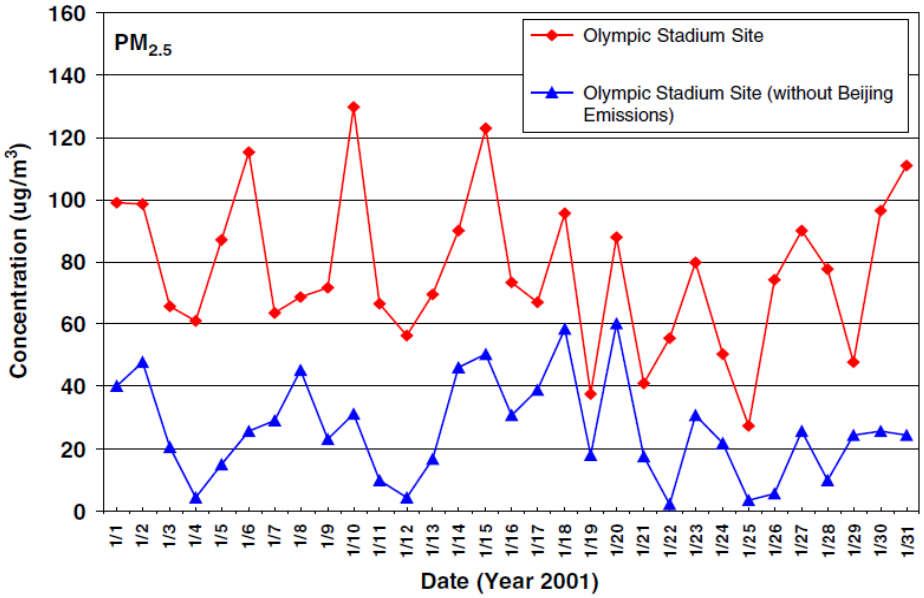
$\Gamma_s < \Gamma < \Gamma_d$

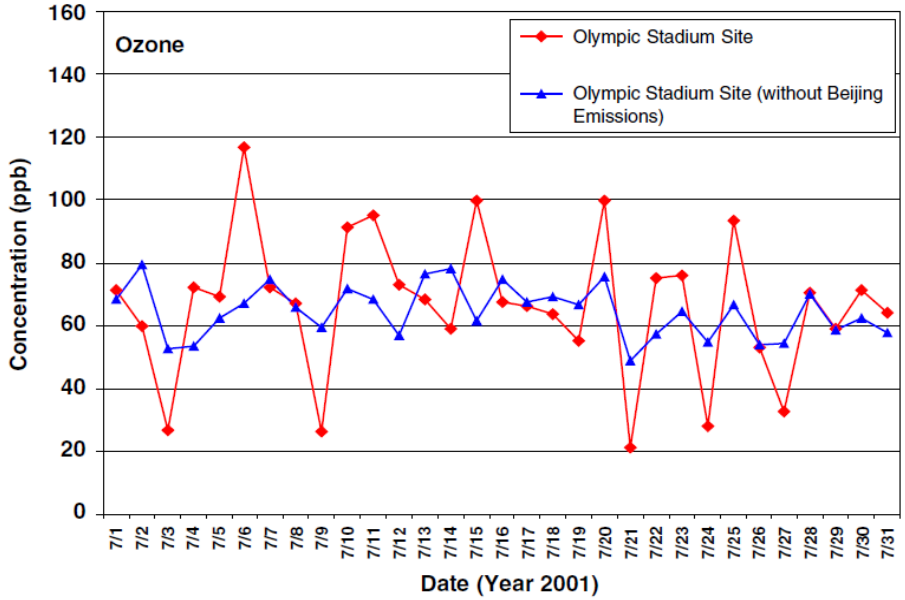




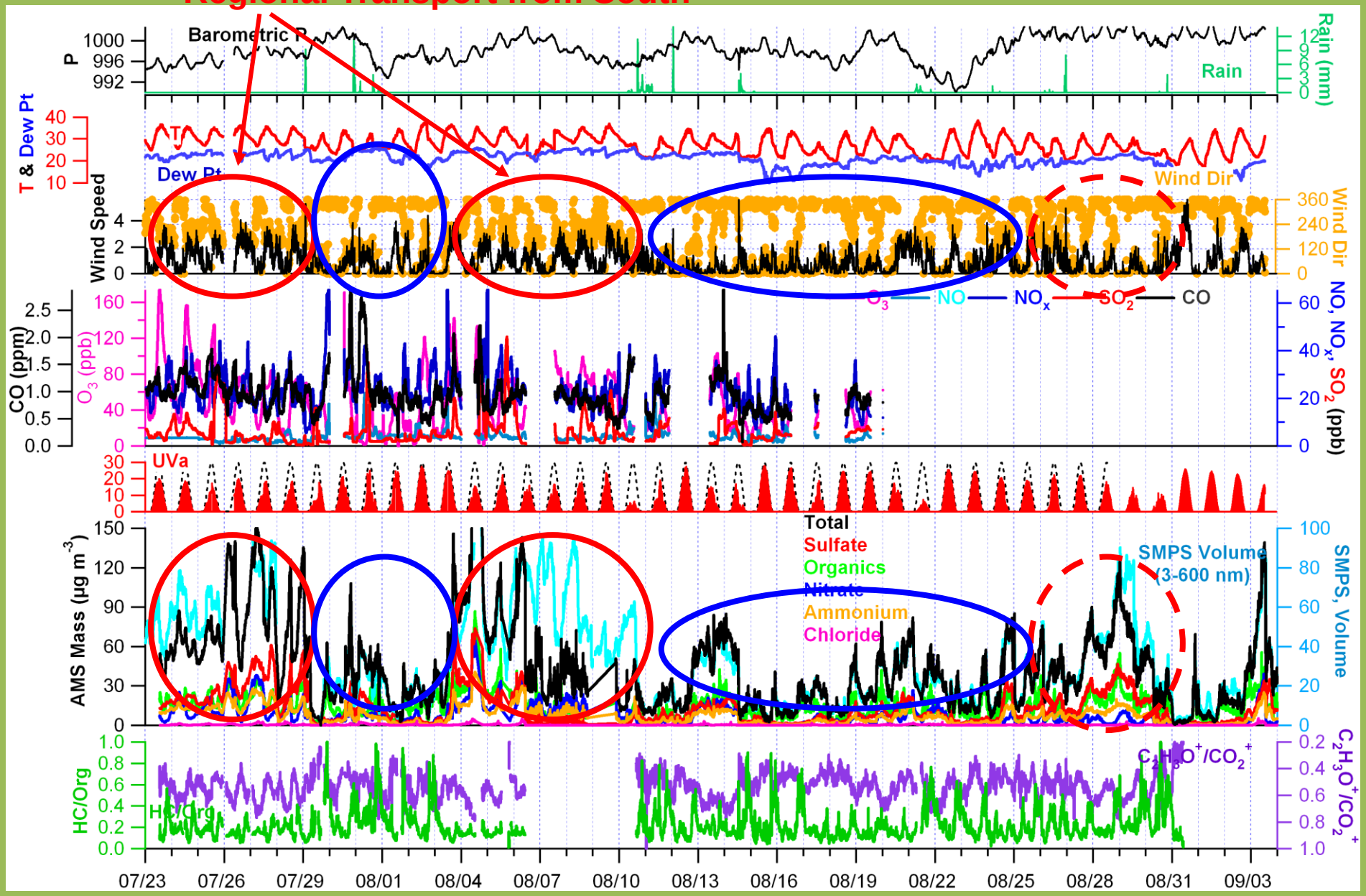
Stable Unstable Neutral Con. unstable

Regional Transport and Transformation Affecting Beijing





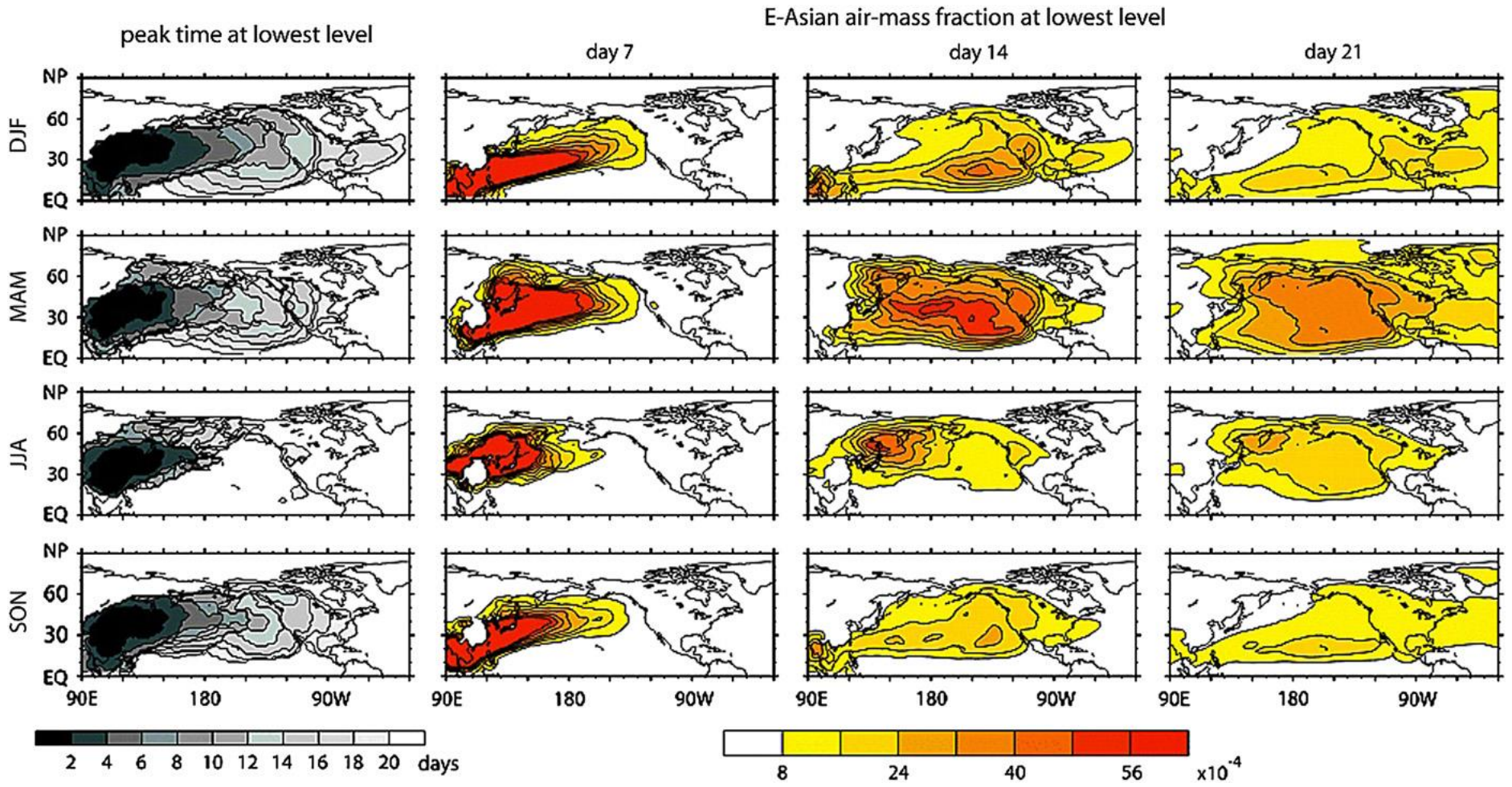
Regional Transport from South



D. Worsnap, Q. Zhang, LY He

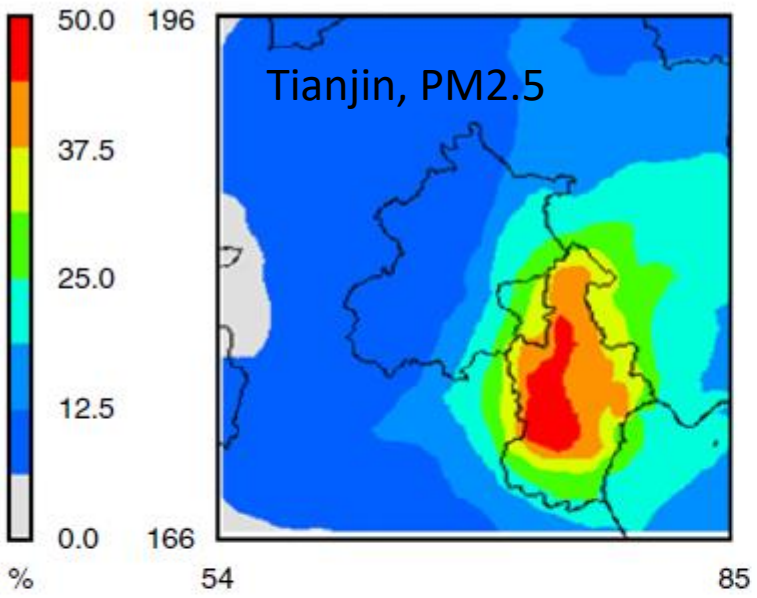
East Asian Influence: Analysis by CTMs

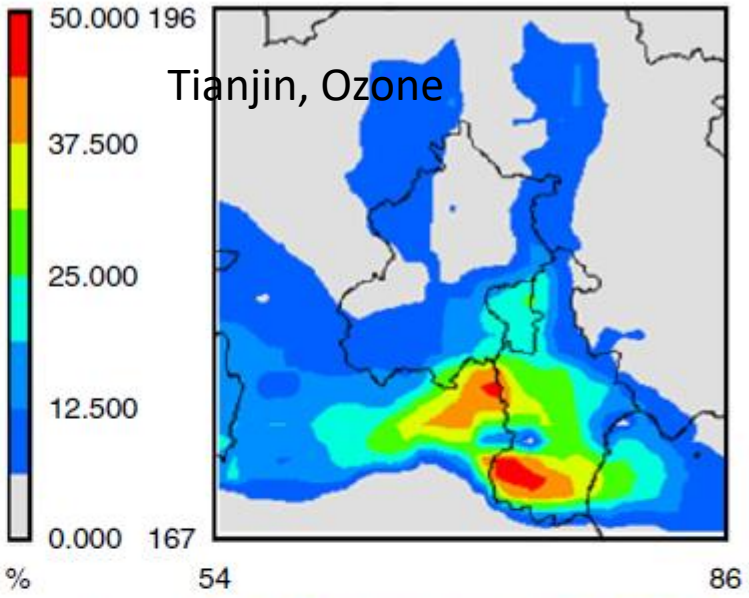
Ozone

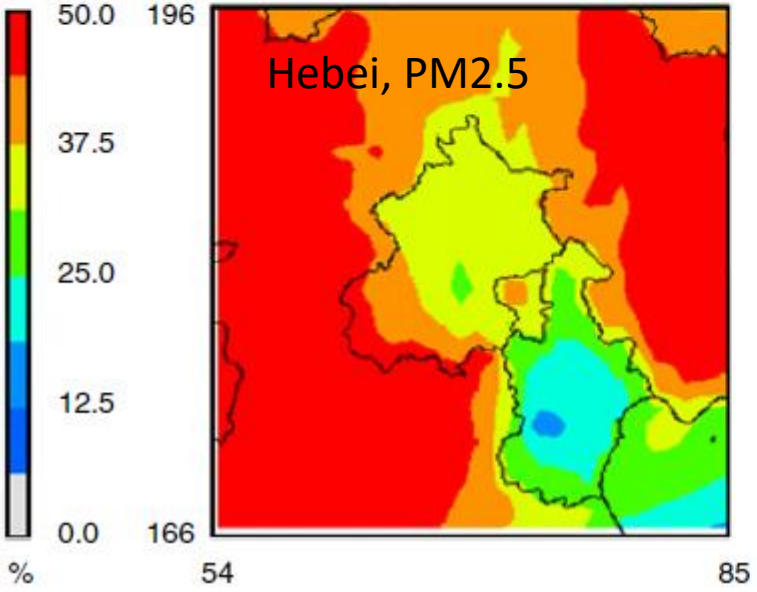


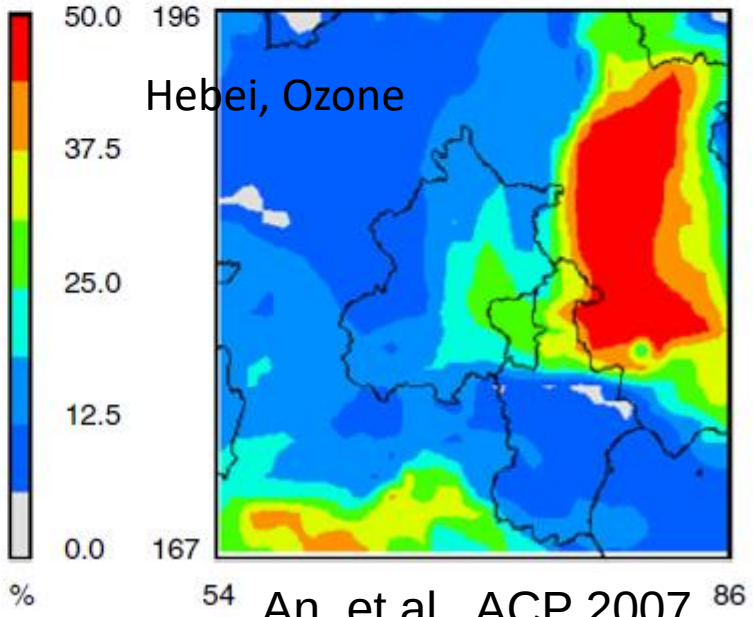
Liu et al., 2009

Regional Transport and Transformation Affecting Beijing





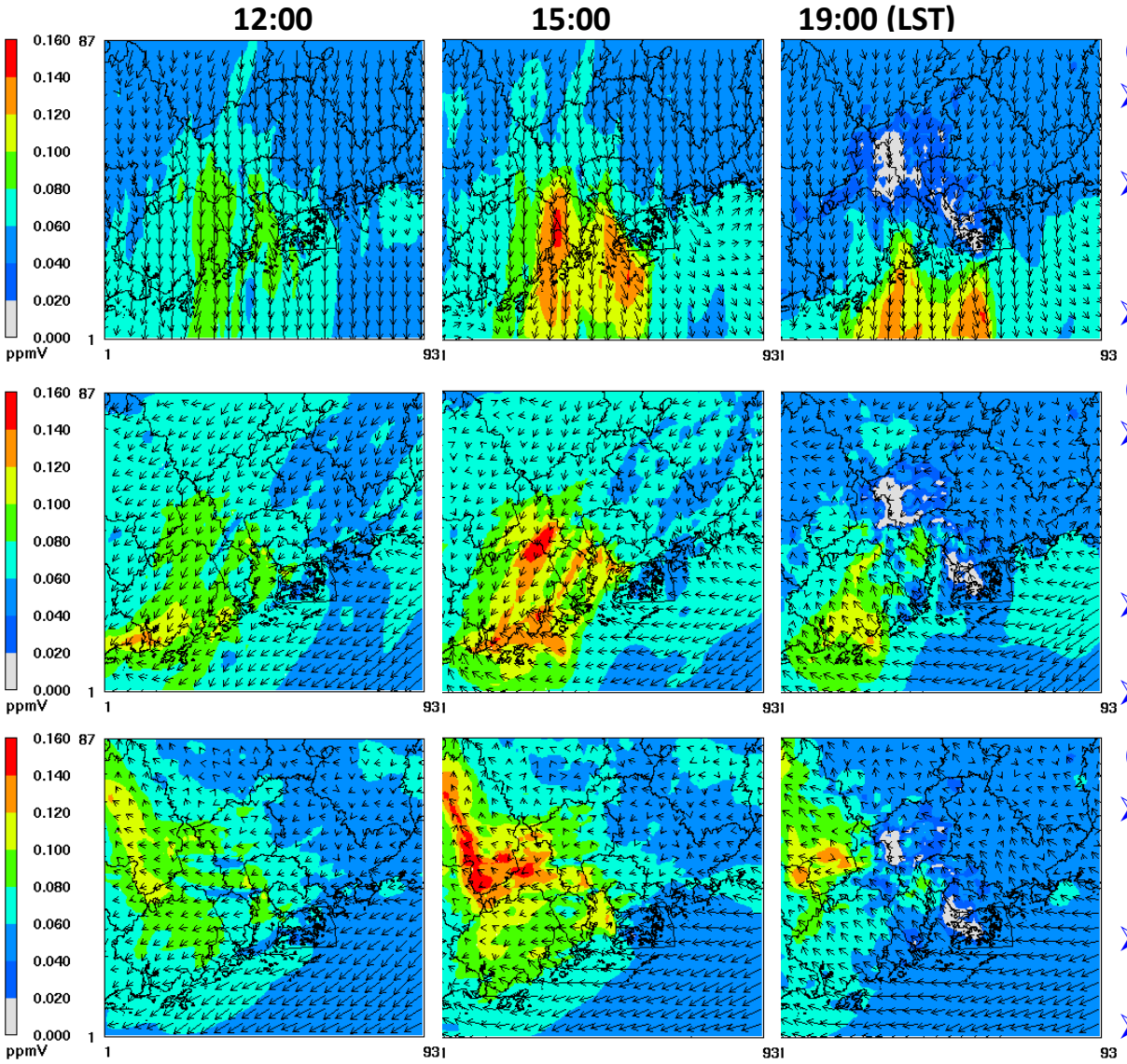




An et al., ACP 2007

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.



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

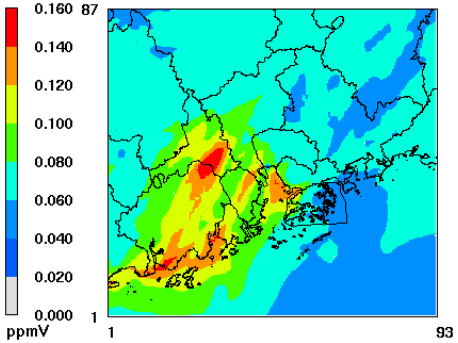
Average O_3 concentration at 15:00 LST

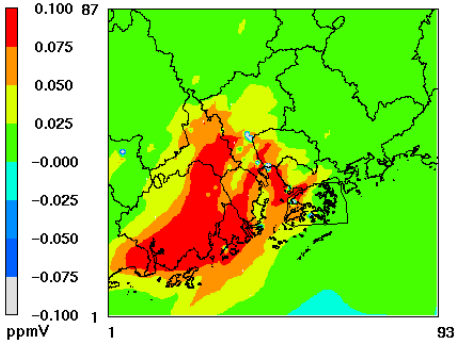
Accumulated contributions to O_3 from 9:00 to 15:00 LST

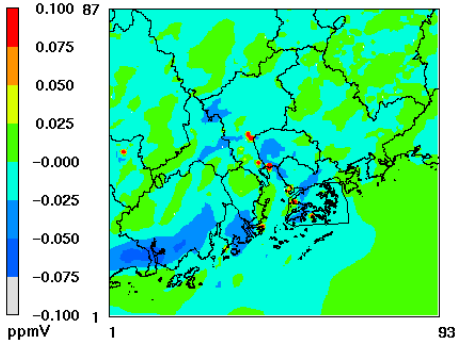
Chemical production

Transport

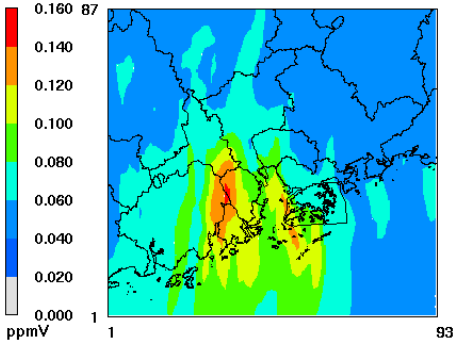
Oct 16

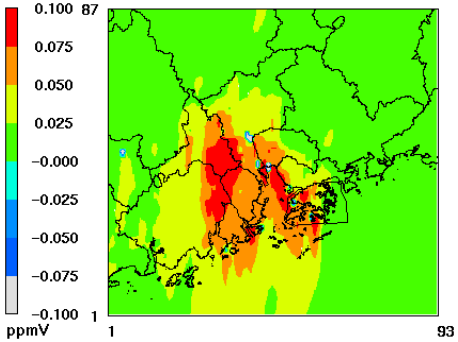


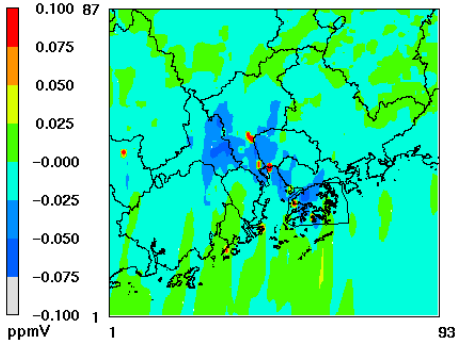




Oct 19







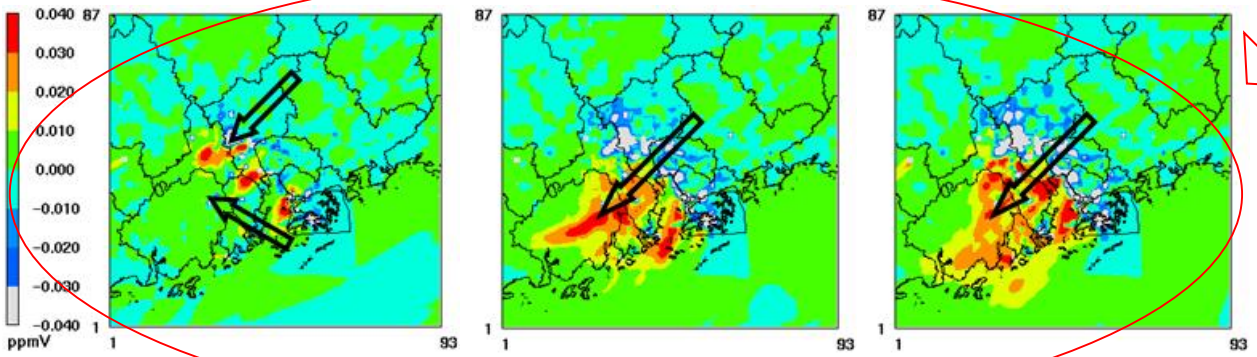
XS. Wang et al., Submitted to ACPD

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

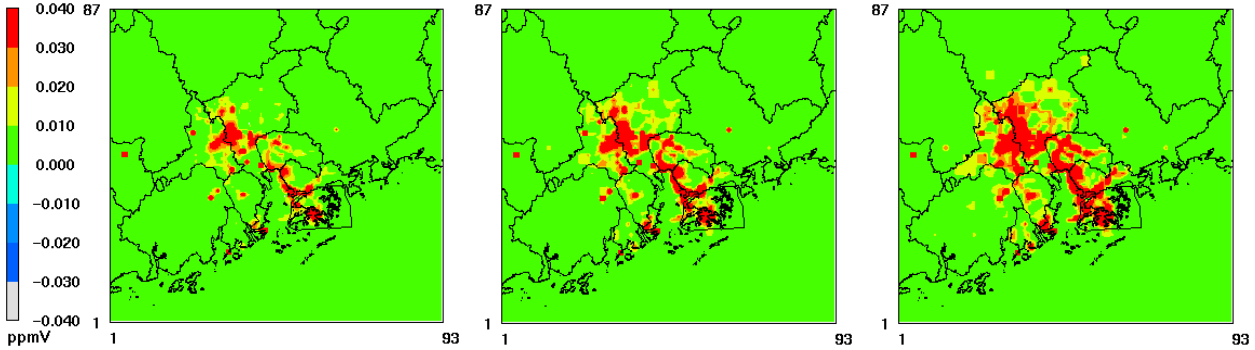
from 17:00 Oct 15 to 00:00 Oct 16

from 17:00 Oct 15 to 09:00 Oct 16

from17:00 Oct 15 to 15:00 Oct 16



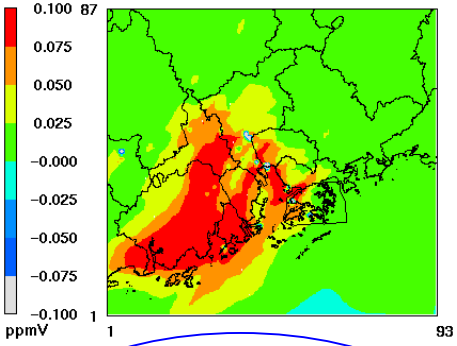
Accumulated NOx transport over layer 1-7 during Oct 15-16



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

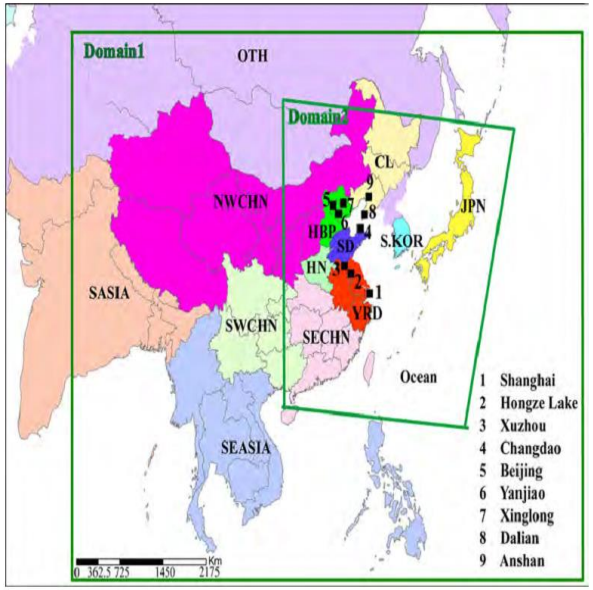
NOx from different source regions mixed and transported to downwind rural areas

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



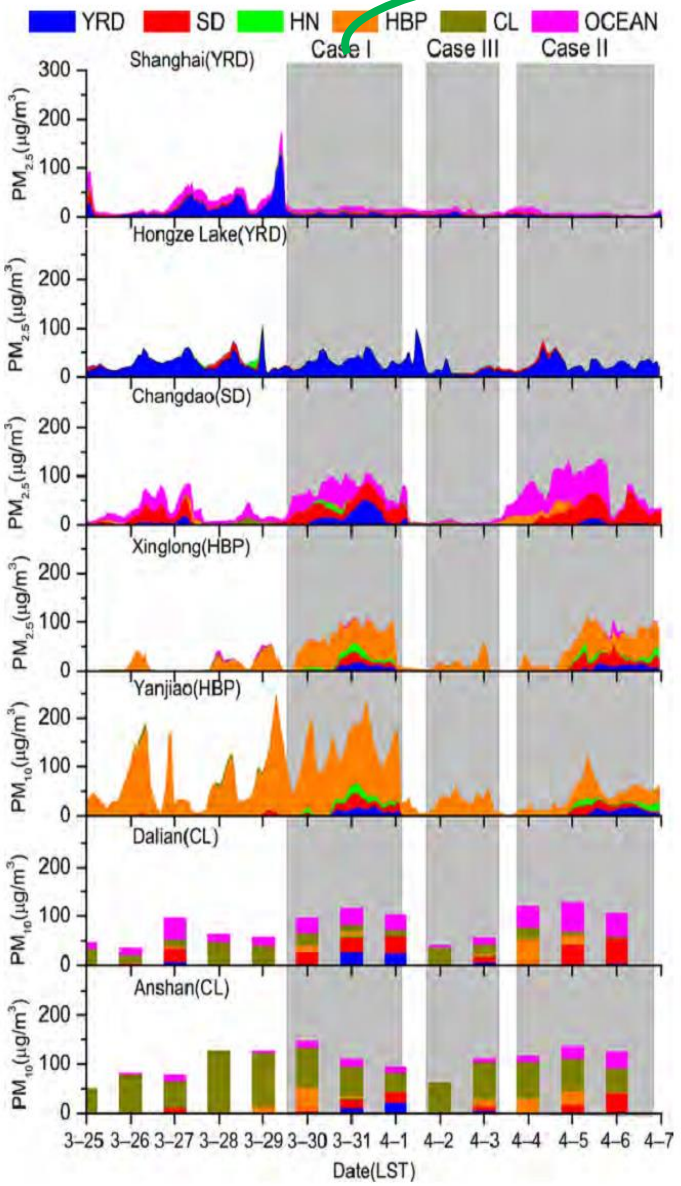
Redistributed NOx involved in the daytime O_3 chemical formation over south and west of PRD

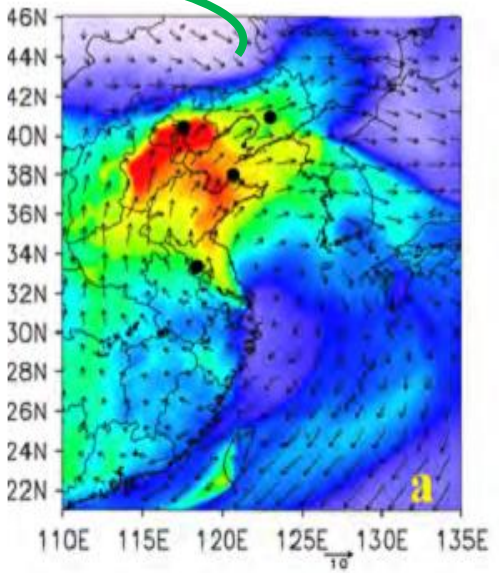
Transport of PM Between China's City Clusters

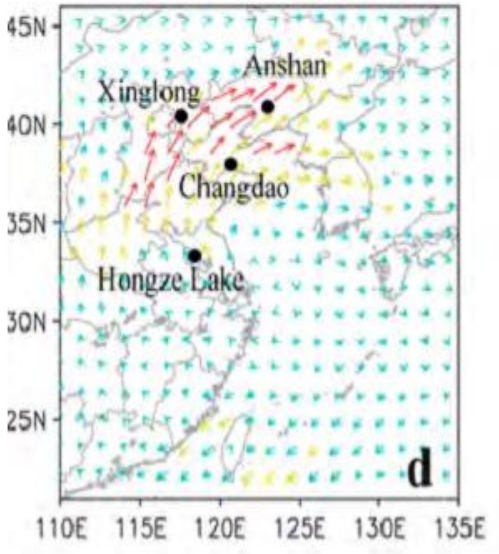


Spring 2011

NAQPMS + tagging



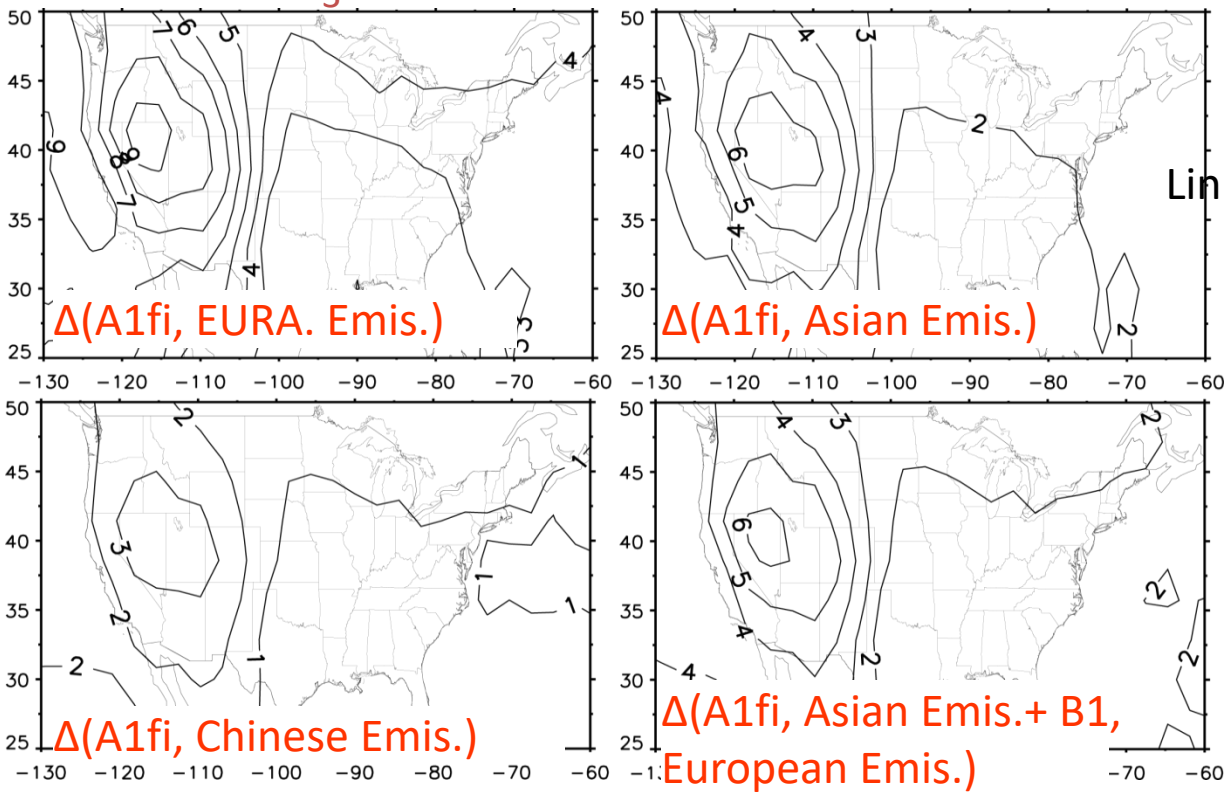




Li et al., 2013, Tellus B

Effects of Δ(European/Asian Emissions)

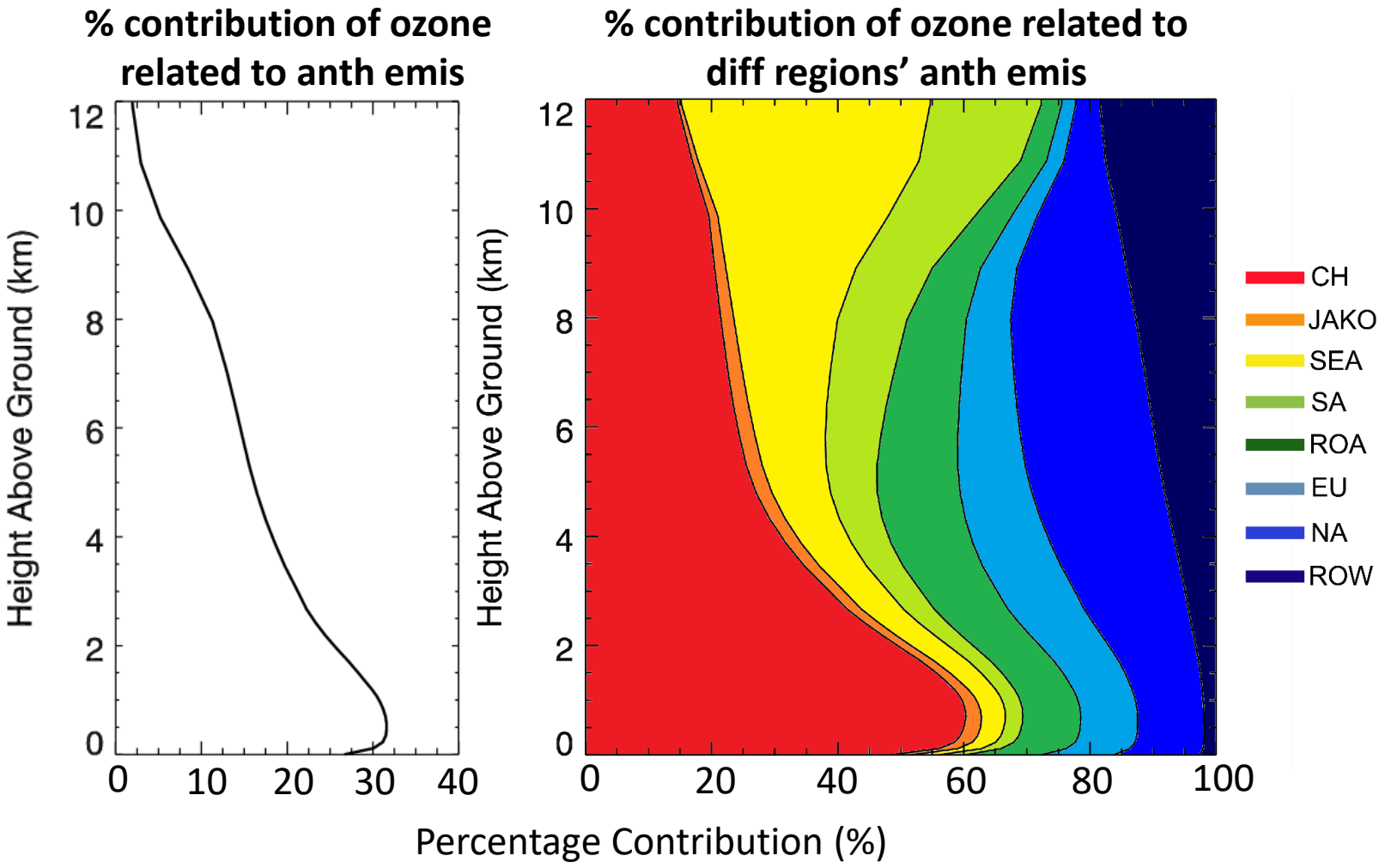
ΔO_3 in JJA from 1999 to 2049



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!

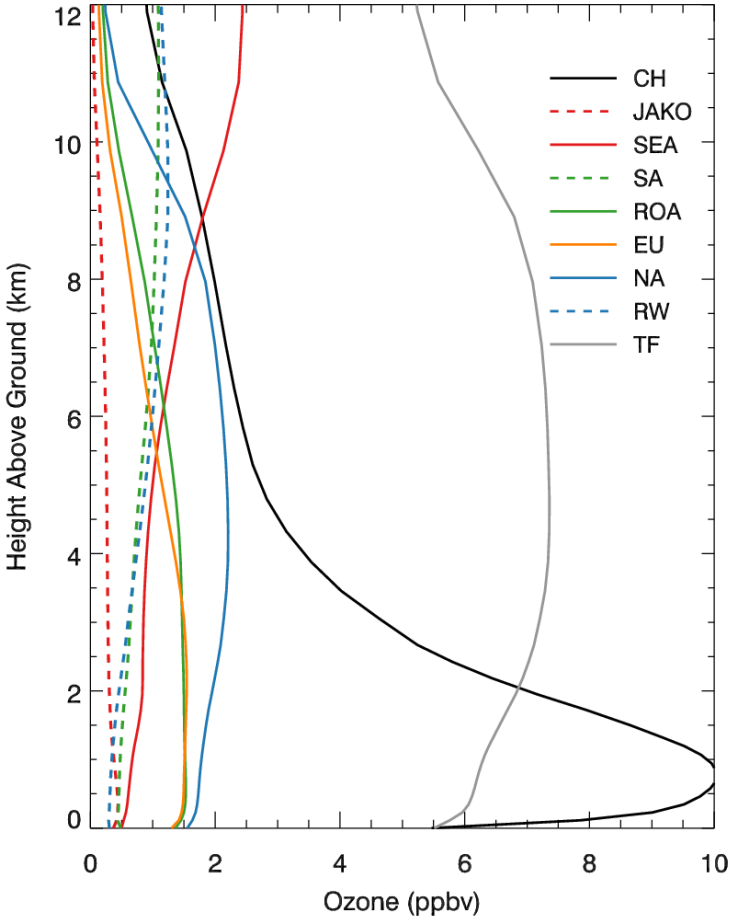


- Anthro emis contribute ~30% in PBL and the contribution decreases with height
- Foreign anthro emis contribute 40~80% to total anthro ozone and the contribution increases with height

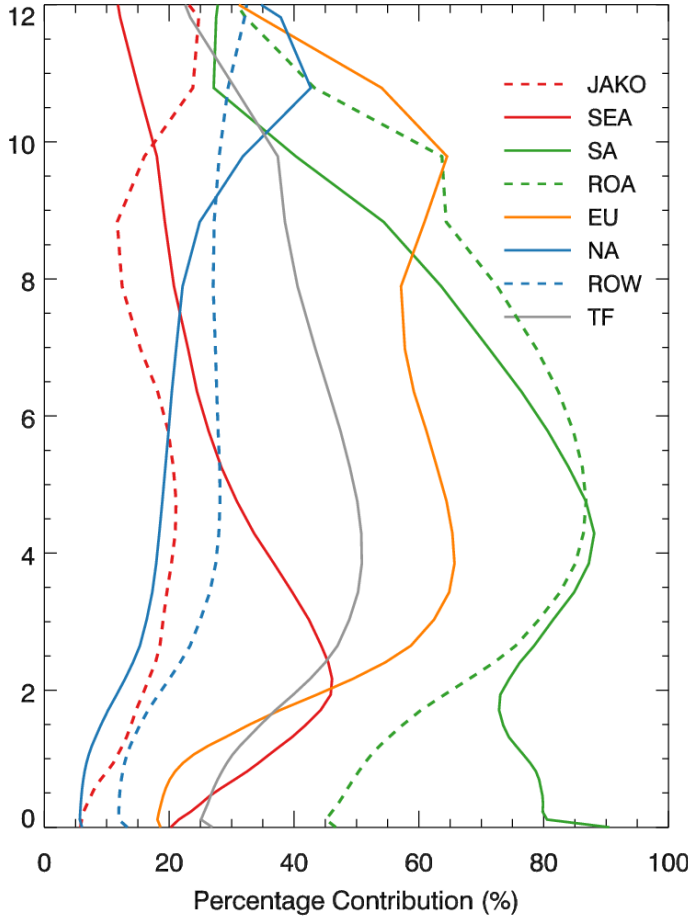
Vertical Profile of O_3 Related to Anthro Emis over China

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



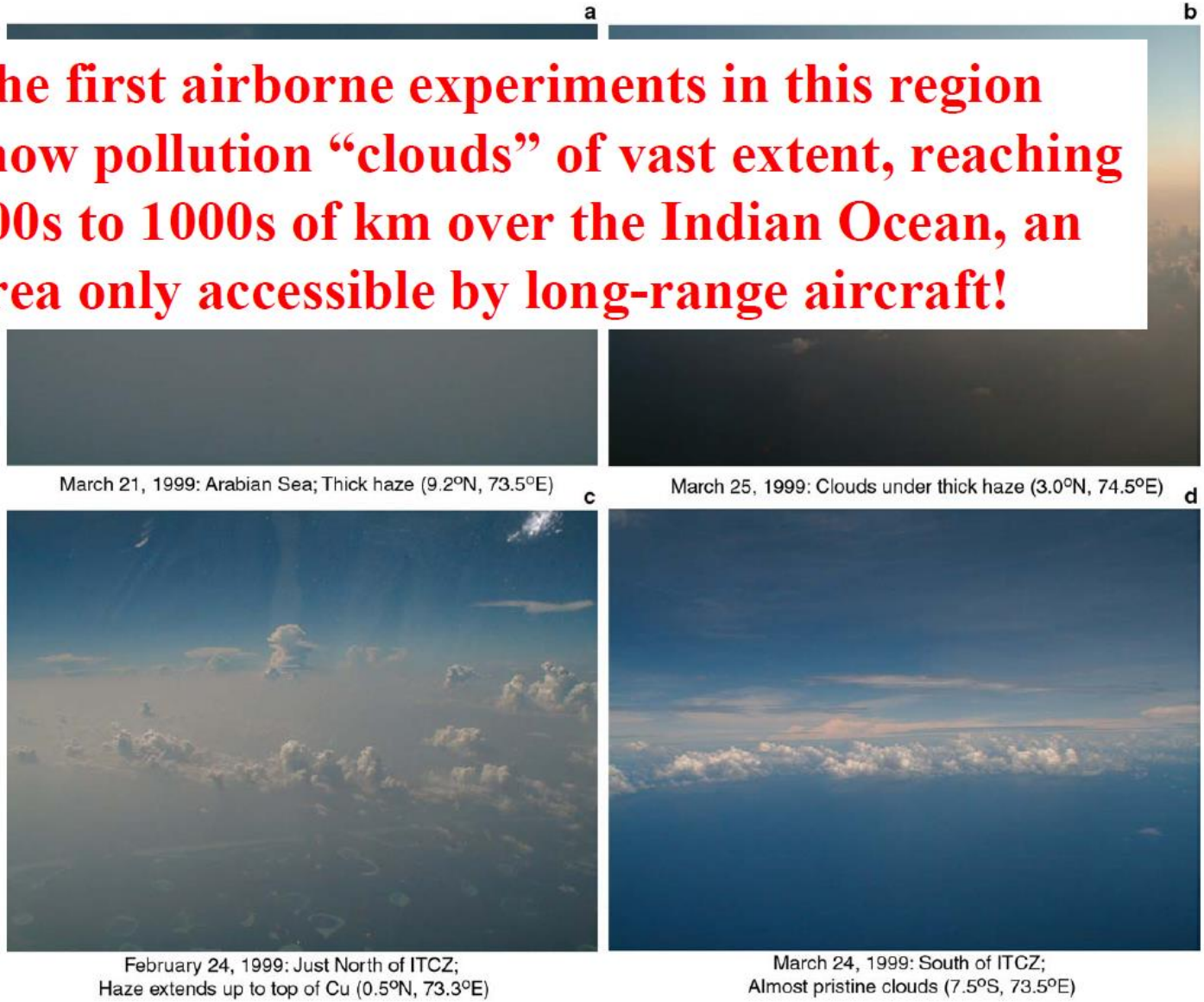
% contribution of ozone produced directly over source region



- 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!



March 21, 1999: Arabian Sea; Thick haze (9.2°N, 73.5°E)

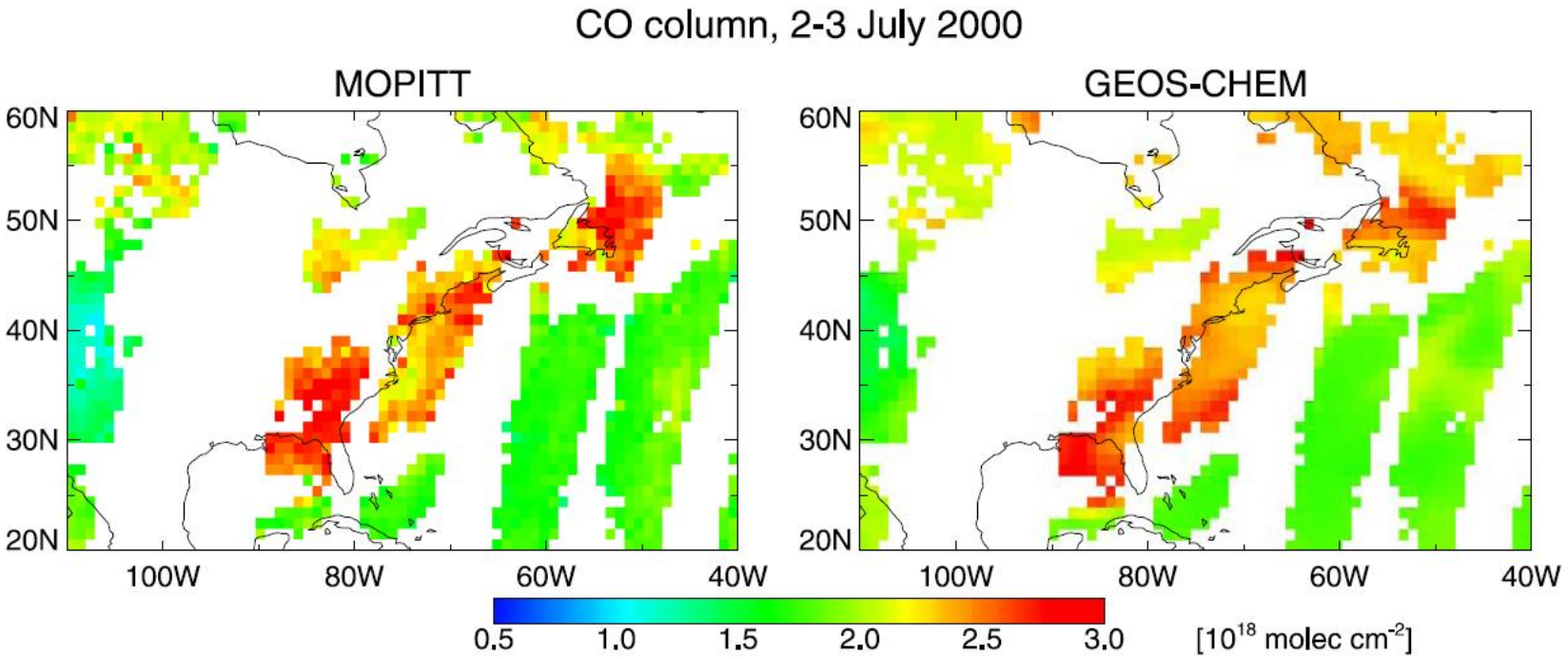
March 25, 1999: Clouds under thick haze (3.0°N, 74.5°E)

February 24, 1999: Just North of ITCZ;
Haze extends up to top of Cu (0.5°N, 73.3°E)

March 24, 1999: South of ITCZ;
Almost pristine clouds (7.5°S, 73.5°E)

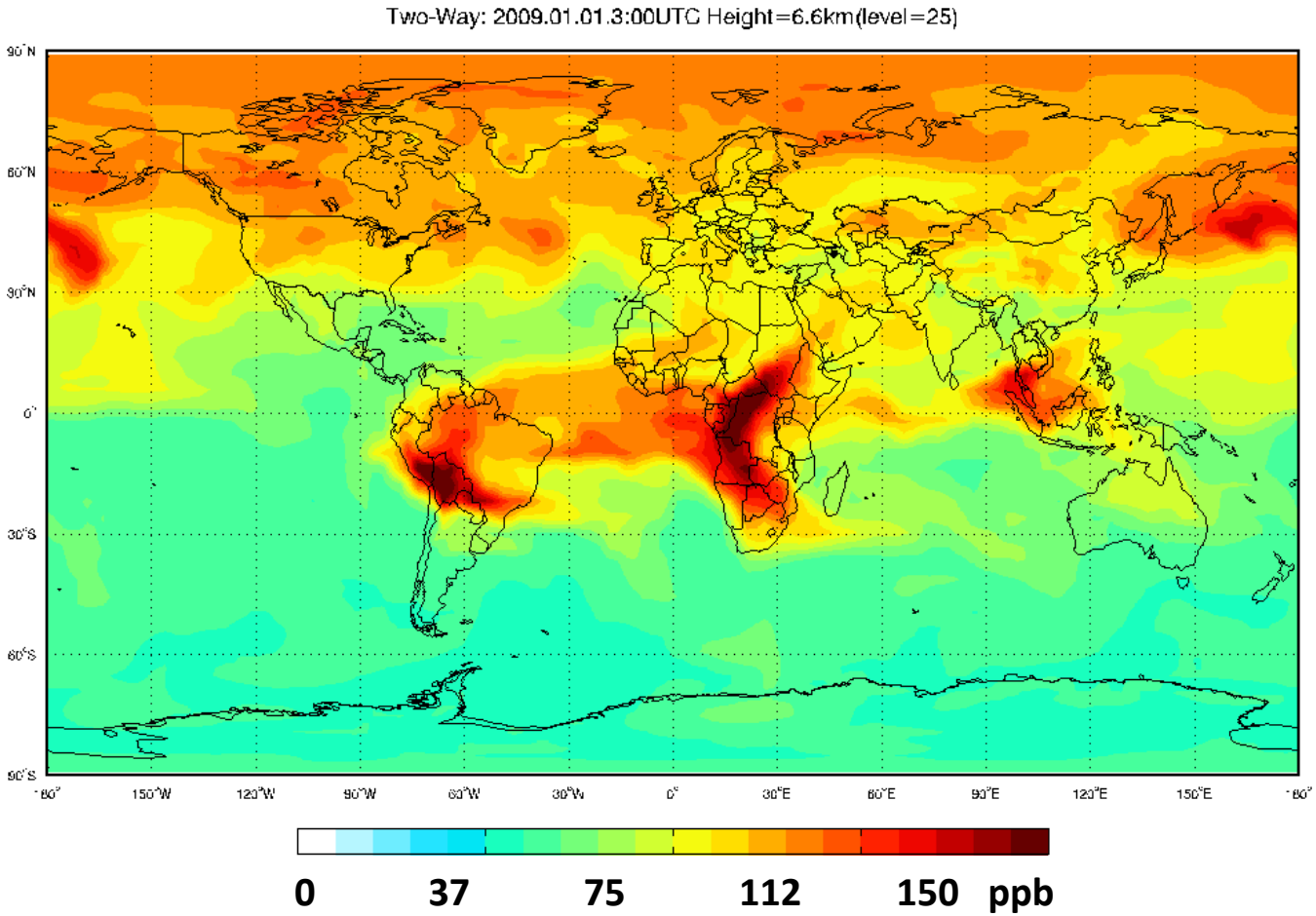
Photo credit: Center for Clouds, Chemistry and Climate; Scripps Institution of Oceanography; University of California, San Diego
Image available at: http://sio.ucsd.edu/supp_groups/siocomm/pressreleases/Indoexagu.html

North American Outflow of CO



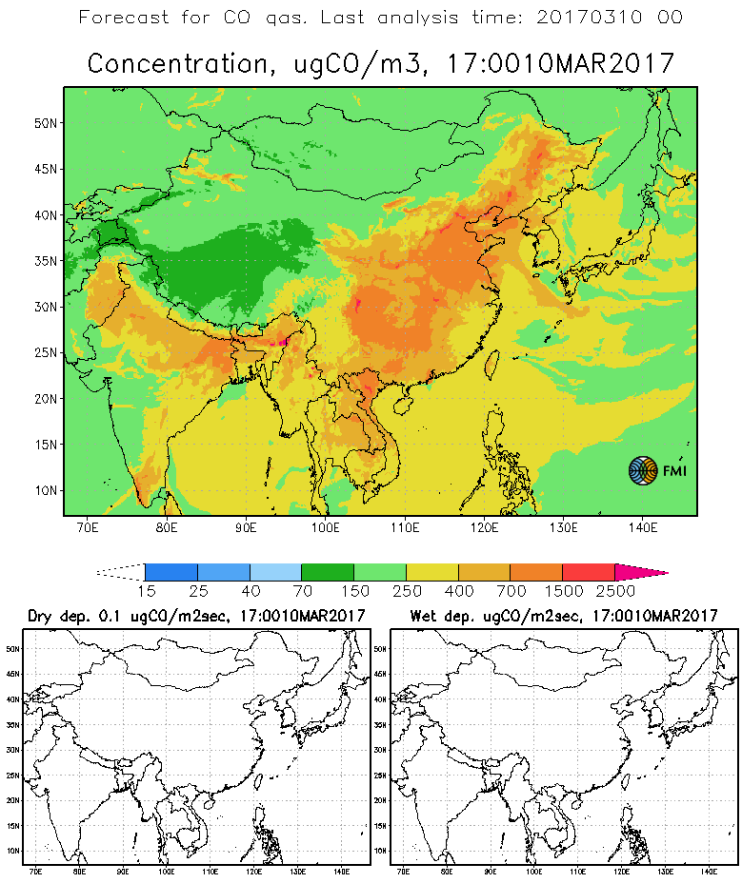
Global Transport of Carbon Monoxide

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:**

  $D = U \times \tau$ = Wind Speed x Lifetime

- **Secondary Pollutant:**

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

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

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

Characteristic Time

Characteristic Distance of Transport

- **Primary Pollutant:**

  $D = U \times \tau$ = Wind Speed x Lifetime

- **Secondary Pollutant:**

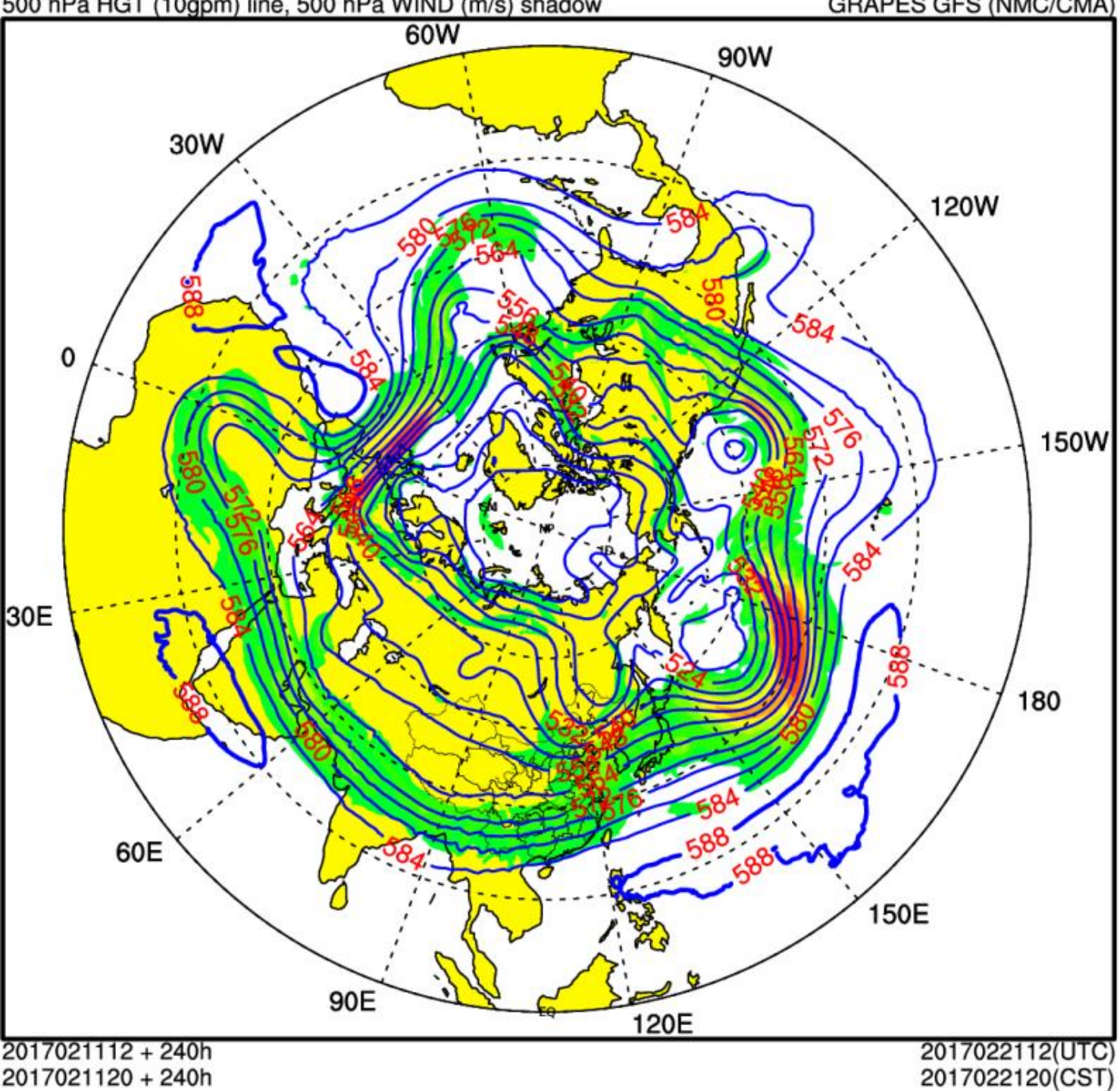
  $D = U \times \tau^* = U \times \tau_s * [\, 1 + r \,]$, where $r = C_p / C_s$

  $\approx U \times \tau_p$ when $C_p / \tau_p << C_s / \tau_s$

  $= U \times [\, \tau_p + \tau_s \,]$ when $C_p / \tau_p = C_s / \tau_s$

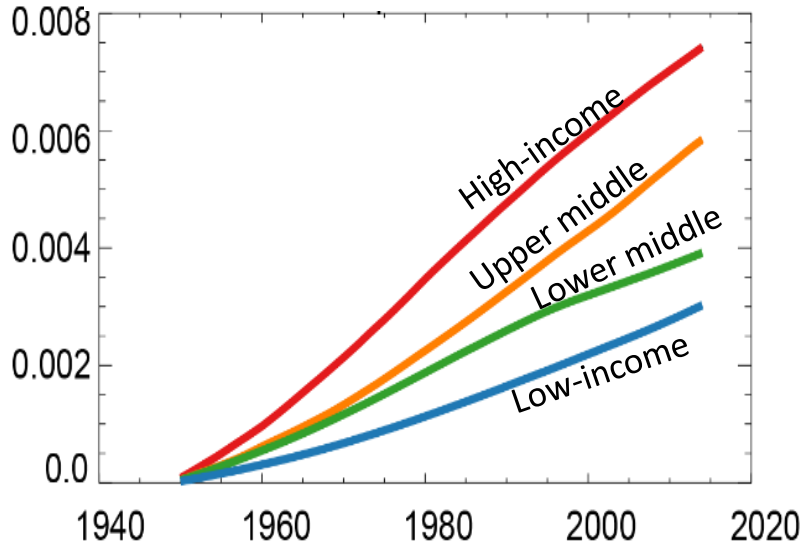
  $\approx U \times \tau_s$ when $C_p / \tau_p >> C_s / \tau_s$

Characteristic Time

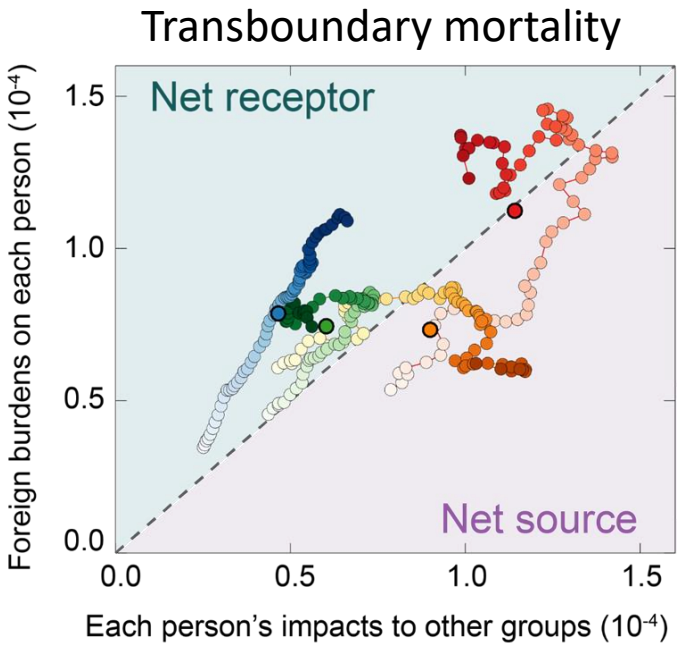

500 hPa HGT (10gpm) line, 500 hPa WIND (m/s) shadow
GRAPES GFS (NMC/CMA)
60W
90W
30W
120W
0
150W
30E
180
60E
150E
90E
120E
2017021112 + 240h
2017021120 + 240h
2017022112(UTC)
2017022120(CST)


55
50
45
40
35
30
25
20

Transboundary $PM_{2.5}$ Mortality Via Atmospheric Transport



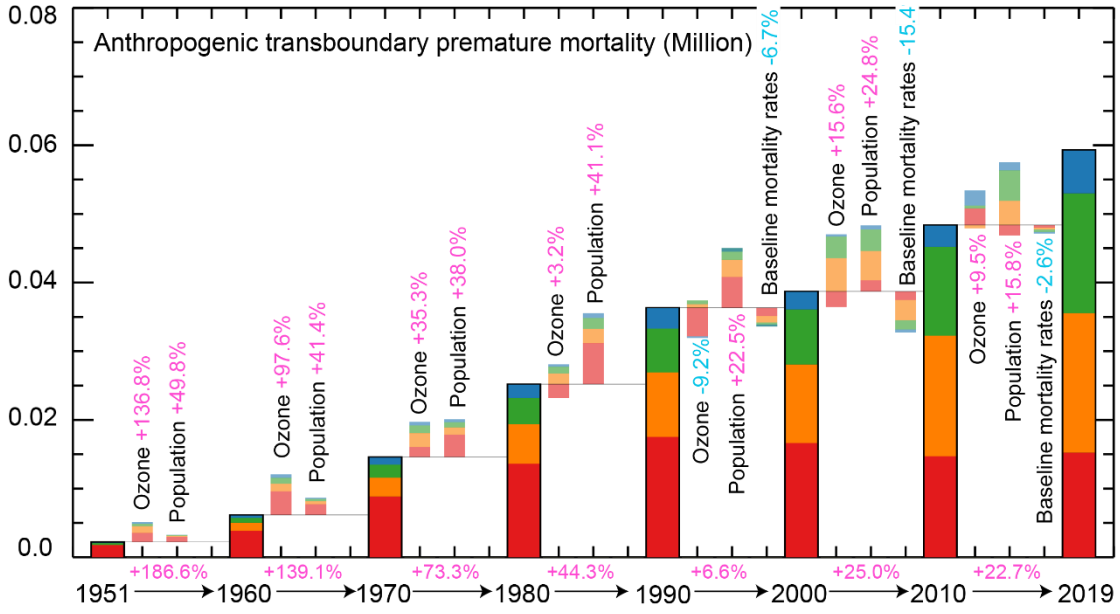
On a per capita contribution basis:
Richer group exerts larger cumulative transboundary mortality

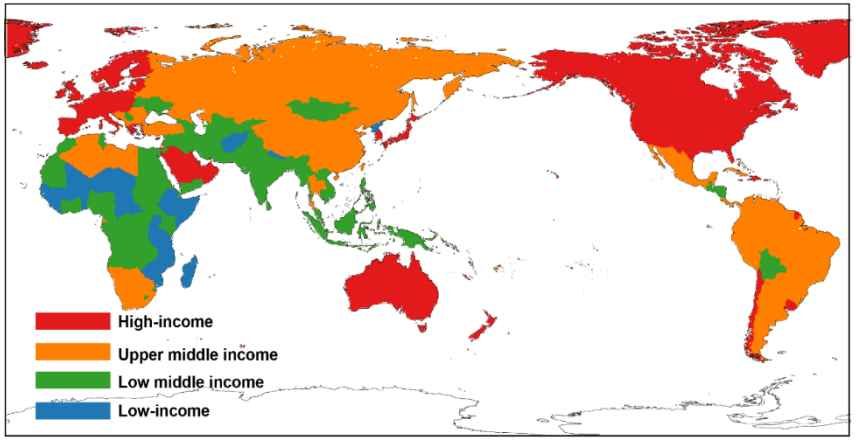


On a per capita *net effect* basis:
Poorest suffers from heaviest *net* transboundary burden

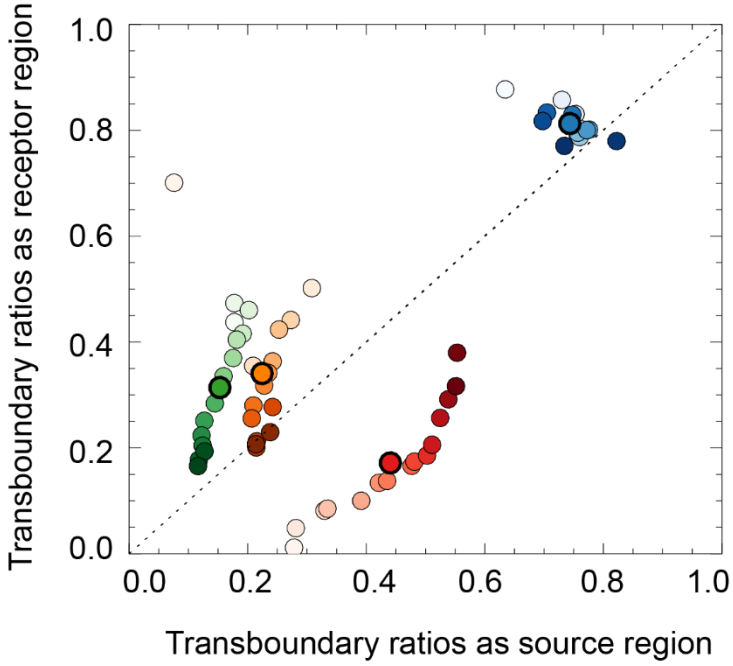
Chen et al., Science Bulletin, 2022

Historical Transboundary Ozone Mortality via Atmospheric Transport



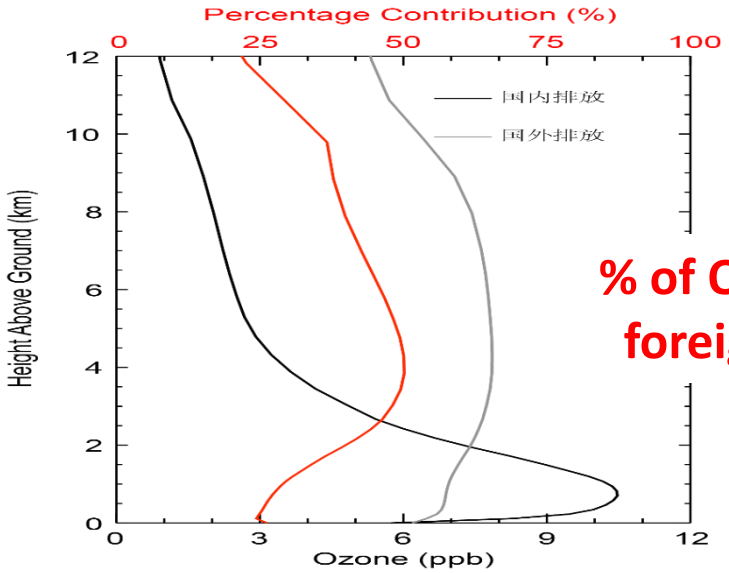


Chen et al., ERL, 2023



Two Chemistry Mechanisms of Ozone Transport





% of O_3 produced within foreign source regions

Ni et al., ACP, 2018