

# Anthropogenic Emissions of NO<sub>x</sub> and Seasonal Variation in China — Multi-satellite Constraint with NO<sub>2</sub> Retrievals from GOME-2 and OMI

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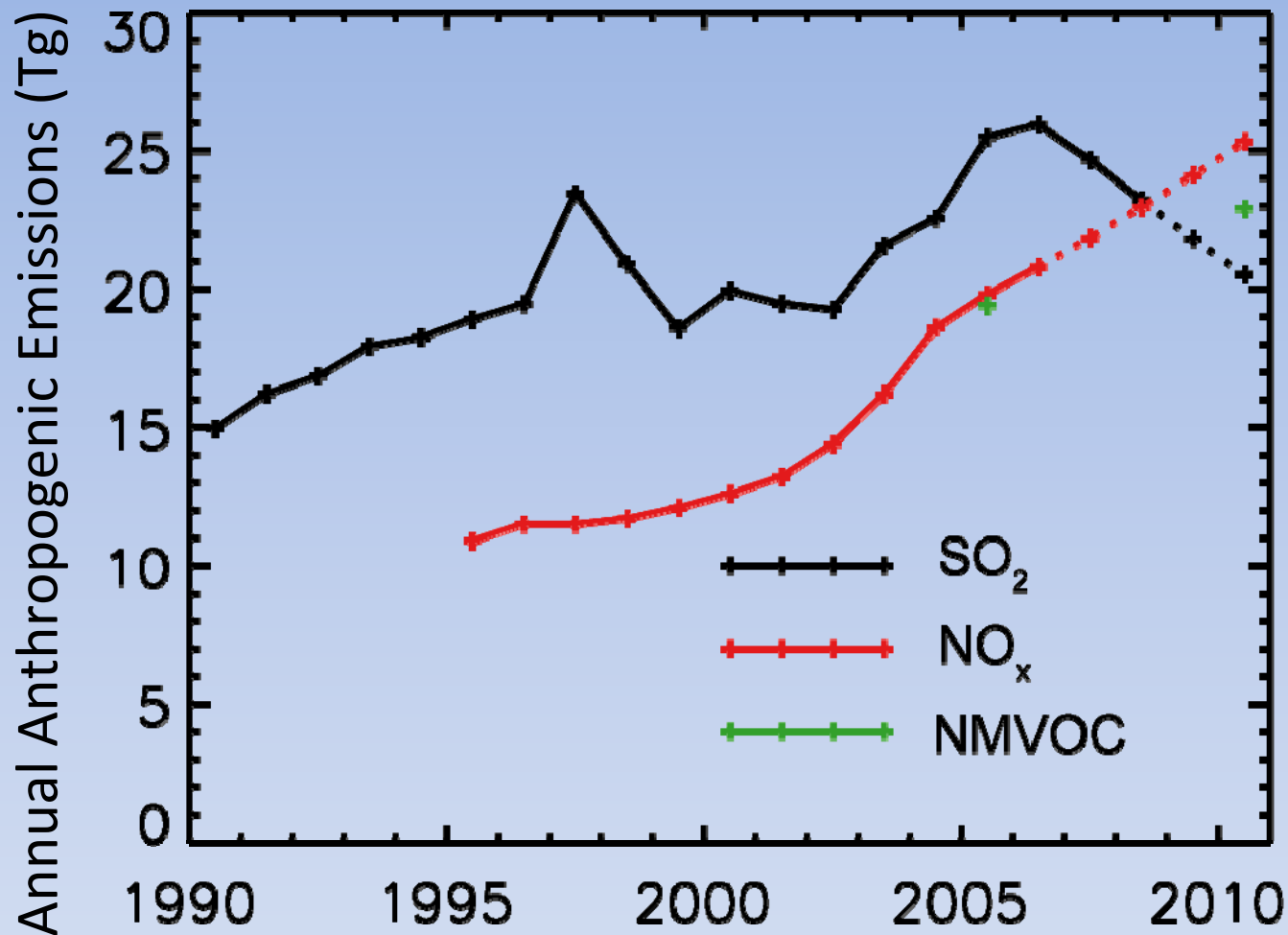
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<http://people.seas.harvard.edu/~jlin5/index.html>

Lin et al., 2009 ACP; Lin and McElroy, 2009 AE

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Zhang et al. (2009): Effects of SO<sub>2</sub> emission reduction from 2005 to 2010 on soil acidification in China may be cancelled out by increases of nitrogen emissions.

Zhang et al., 2007  
Cao et al., 2009  
Wei, 2009  
<http://www.stats.gov.cn/tjsj/>

## Bottom-up anthropogenic emissions (TgN/yr)

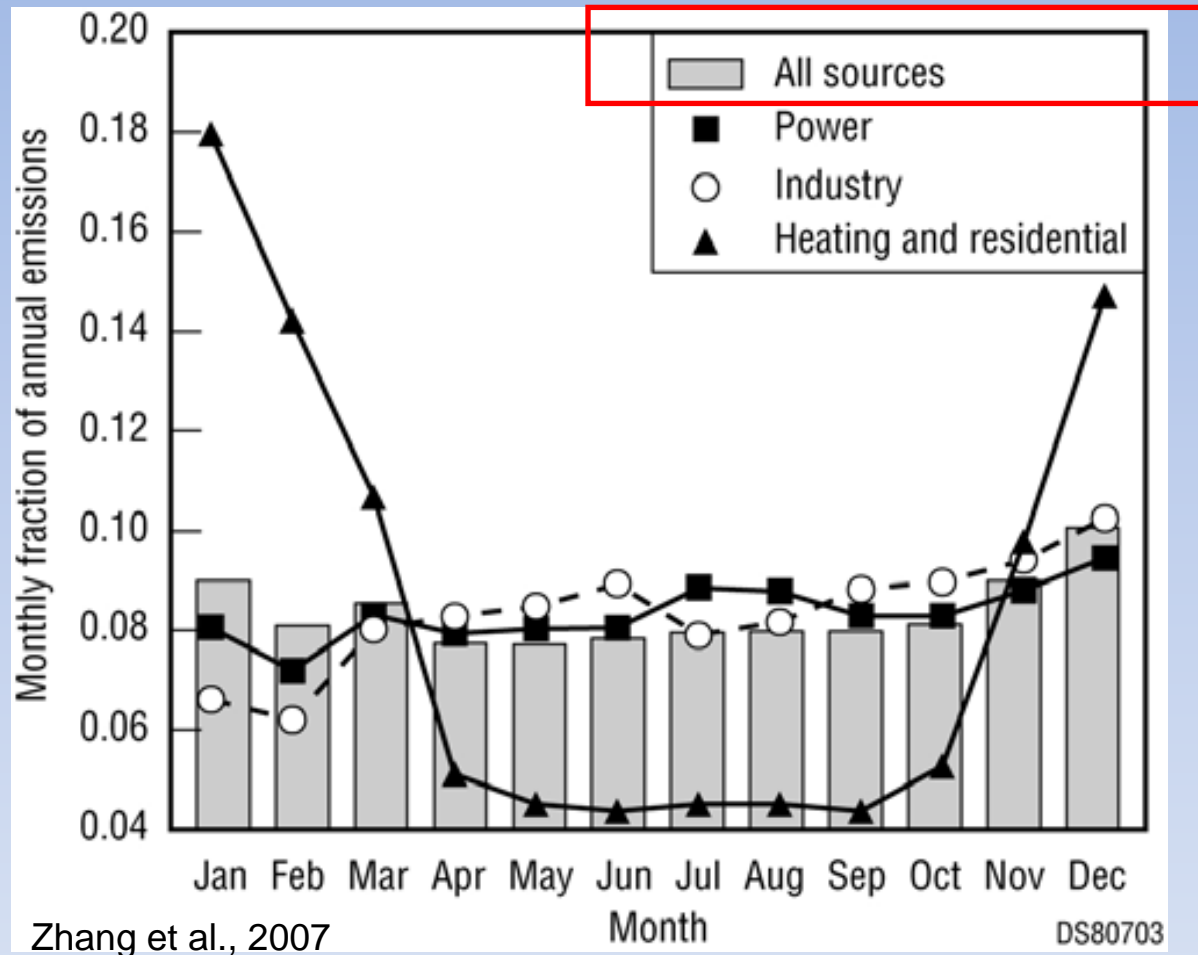
Authors	1995	96	97	98	99	2000	01-06	2008
Many	3.2-4.0	3.7	3.6-3.8	3.4		3.2-4.2		
Zhang	3.3	3.5	3.5	3.6	3.7	3.8	4.0-6.6	
Lin et al. 2009								6.2 (E. China)

## Top-down emissions using Martin et al. method (TgN/yr)

Authors (Instruments)	Sources	Region	Year	Emis.
Martin et al. 2003 (GOME)	all	China+Korea	96-97	4.7
Jaegle et al. 2005 (GOME)	FF+BF	China+Korea	2000	5.3
Martin et al. 2006 (SCIAMACHY)	all	East Asia	04-05	12.7
Wang et al. 2007 (GOME)	all	East China	97-00	5.1
Lin et al. 2009 (GOME-2; OMI)	FF+BF	East China	2008	7.5-14.8

Martin et al. method: Proportioning daily mean NO<sub>x</sub> emissions to NO<sub>2</sub> column at a particular time of day; sensitive to existing systematic retrieval errors

# Seasonality for Anthropogenic Emissions of NO<sub>x</sub>



- Bottom-up: winter/summer < 120% (Streets et al.; Zhang et al.)
- Top-down by Martin et al. method: winter/summer = ~ 140% (Jaegle et al.; Wang et al.)
- Differences between top-down and bottom-up are largest in winter

# New Methodology

$$\partial \Omega / \partial t = E - \Omega / \tau \quad (\text{neglecting horizontal transport})$$

Solving  $\text{NO}_x$  column  
at  $(i+1)^{\text{th}}$  hour

$$\Rightarrow \Omega_{\text{NO}_x} |_{i+1} = E_i * \tau_i * (1 - e^{-\Delta t / \tau_i}) \\ + \Omega_{\text{NO}_x} |_i * e^{-\Delta t / \tau_i}$$

Solving  $\text{NO}_x$   
column at  $n^{\text{th}}$  hour

$$\Rightarrow \Omega |_n = \sum_{i=0}^{n-1} (E_i * \Lambda_i) + \Omega |_0 * K$$

$\Omega |_n$  —  $\text{NO}_x$  column at  $n^{\text{th}}$  hour

$\Omega |_0$  — Initial  $\text{NO}_x$  column

$\Lambda_i$  — Function of  $\text{NO}_x$  lifetime tau

$K$  — Function of  $\text{NO}_x$  lifetime tau

$E_i$  — Top-down emissions **to be determined**

*Detailed derivation is presented by Lin et al., 2009 ACP*

# New Methodology

$$\partial \Omega / \partial t = E - \Omega / \tau$$

$$\Rightarrow \Omega_{NO_x} |_{i+1} = E_i * \tau_i * (1 - e^{-\Delta t / \tau_i})$$

$$+ \Omega_{NO_x} |_i * e^{-\Delta t / \tau_i}$$

$$\Rightarrow \Omega |_n = \sum_{i=0}^{n-1} (E_i * \Lambda_i) + \Omega |_0 * K$$

$\Omega |_n$  — NO<sub>x</sub> column at n<sup>th</sup> hour OMI + GEOS-Chem (2:00pm)

$\Omega |_0$  — Initial NO<sub>x</sub> column GOME-2 + GEOS-Chem (10:00am)

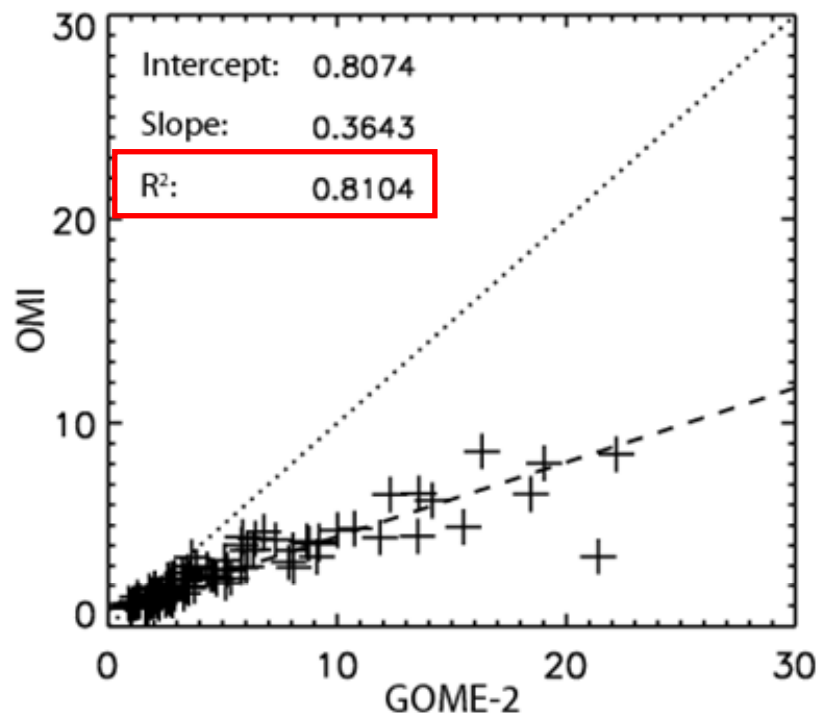
$\Lambda_i$  — Function of NO<sub>x</sub> lifetime tau GEOS-Chem

$K$  — Function of NO<sub>x</sub> lifetime tau GEOS-Chem

$E_i$  — Top-down emissions **to be determined**

# GOME-2 versus OMI

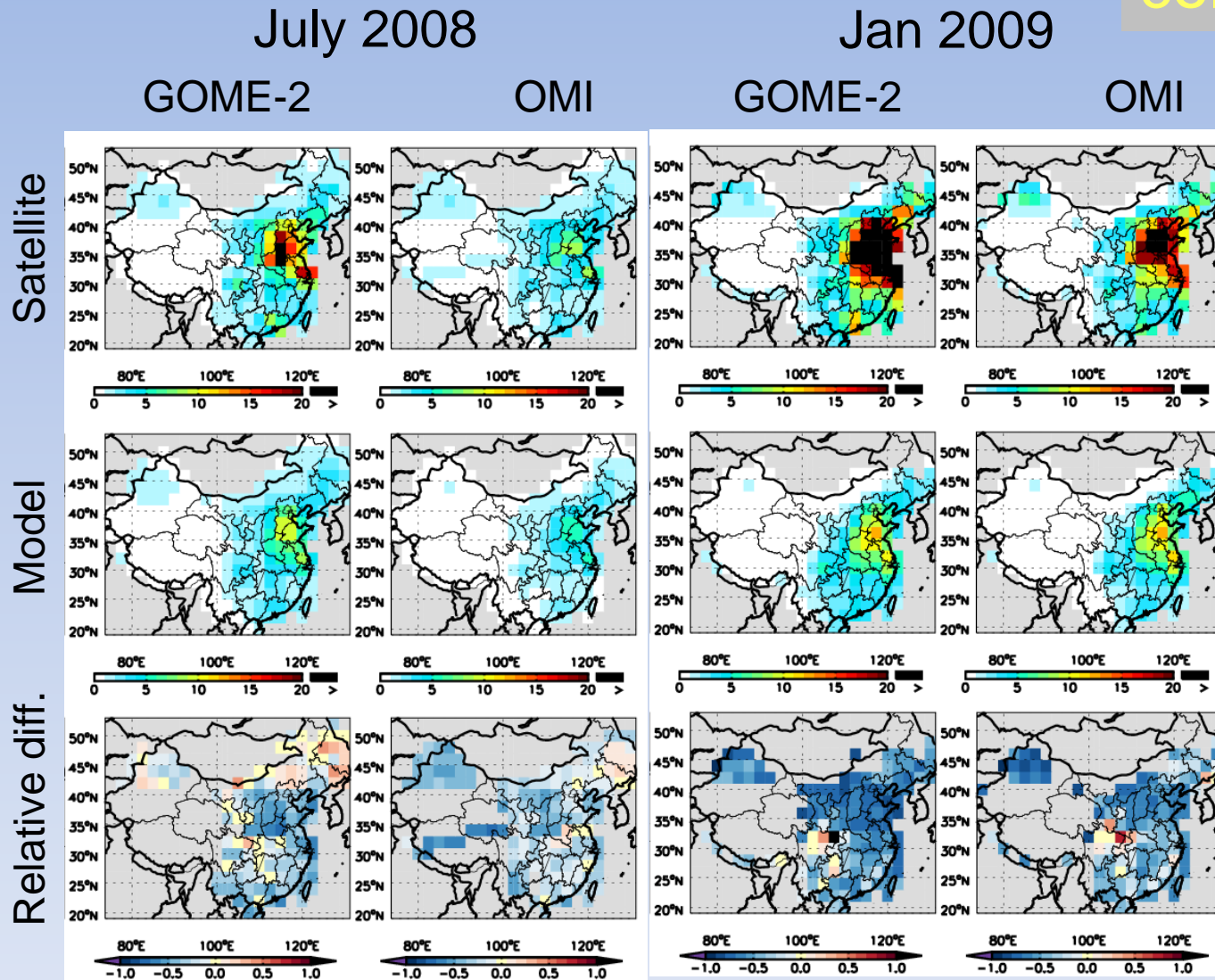
Instrument	Overpass time over China	Nadir view resolution	Global coverage	Spectral window	Cloud scheme	Surface reflectivity
GOME-2	~10:00am	40x80 km <sup>2</sup>	~1 day	425-450 nm	FRESCO+	TOMS + GOME
OMI	~2:00pm	13x24 km <sup>2</sup>	1 day	405-465 nm	O <sub>2</sub> -O <sub>2</sub>	TOMS + GOME



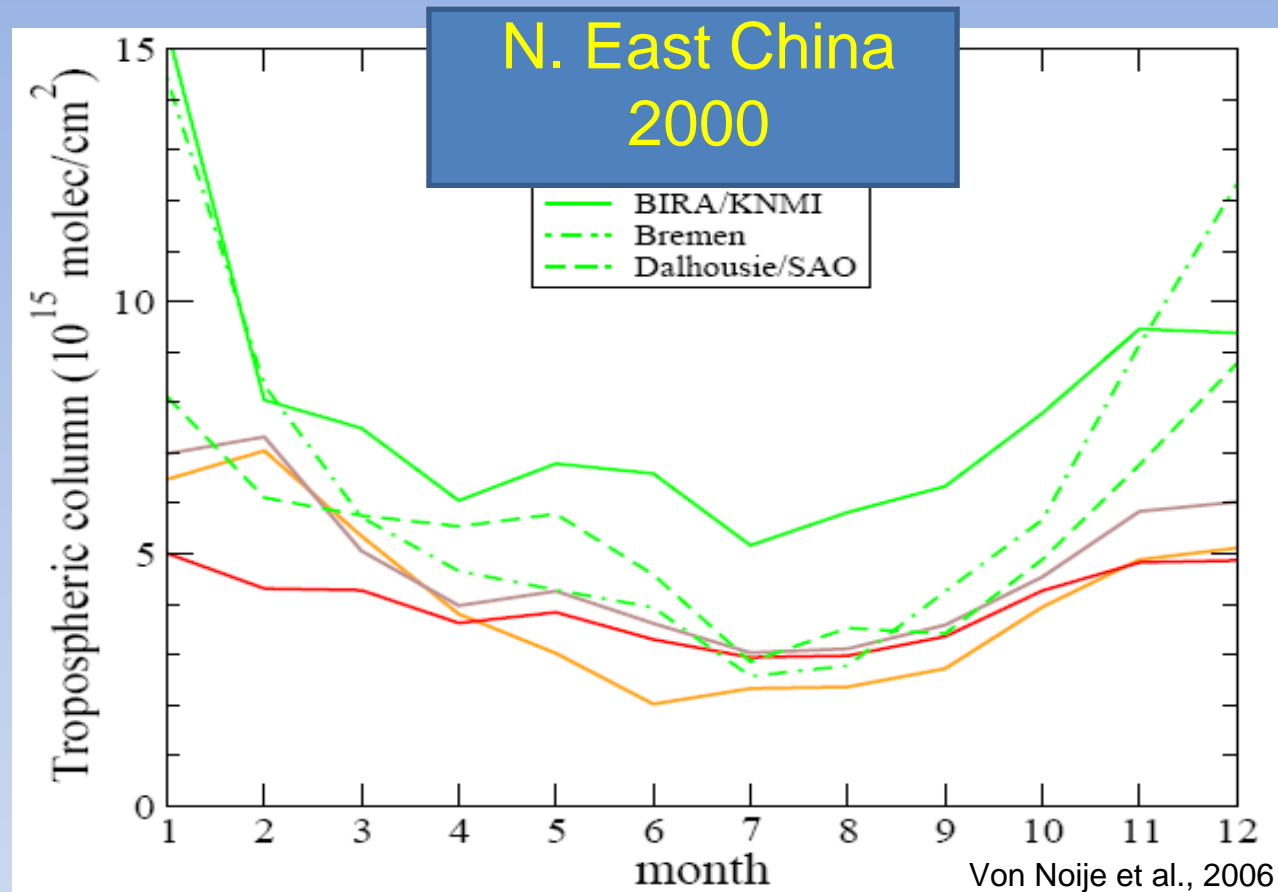
$$VCD = (SCD - SCD_{str}) / AMF$$

- Tropospheric NO<sub>2</sub> column densities retrieved from both instruments use similar approaches by KNMI
- As a result, systematic biases are consistent in both products

# Tropospheric NO<sub>2</sub> column density

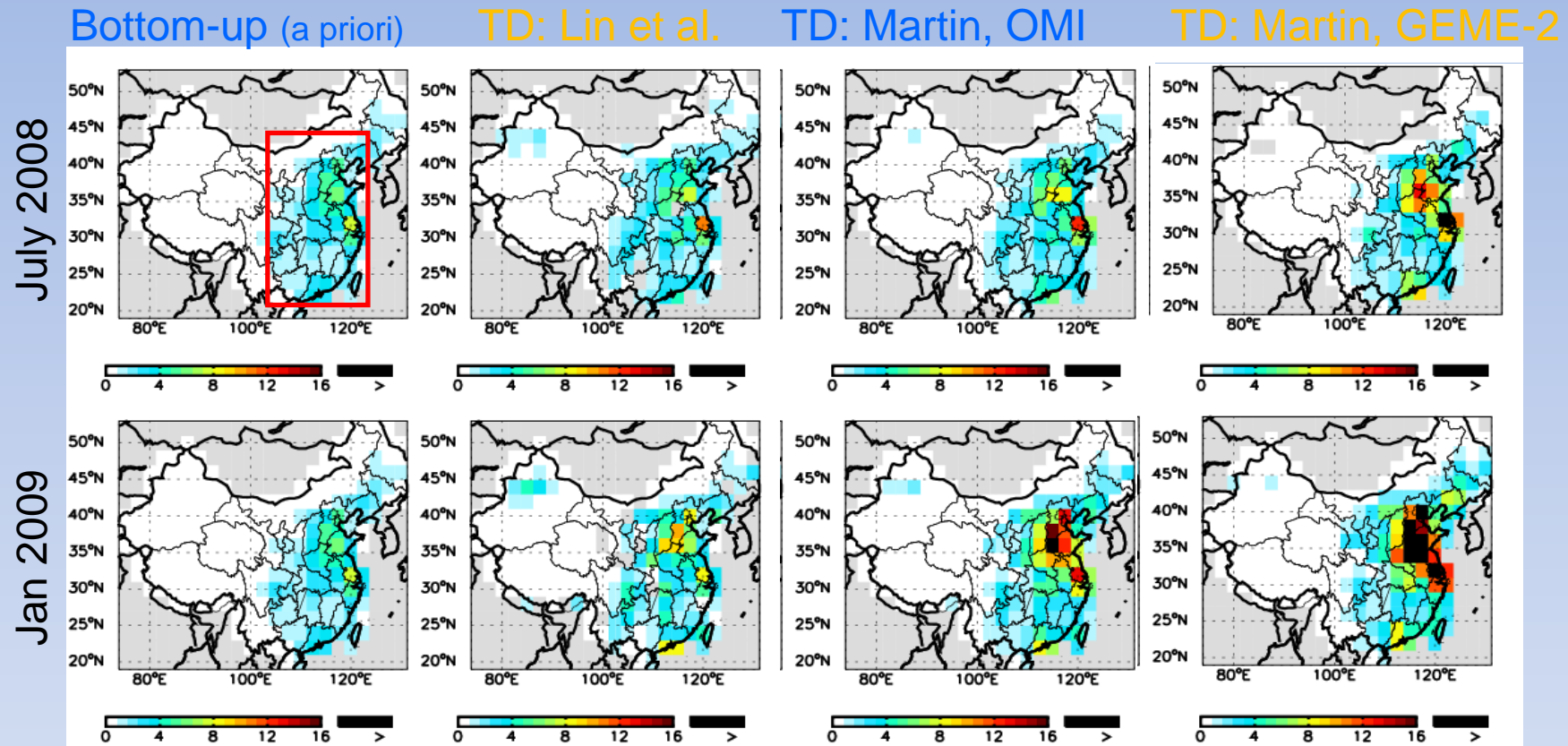


- Retrieved NO<sub>2</sub> columns are much larger than modeled
  - Differences are larger in winter
- Spatial patterns are similar



- Assuming the mean of the three retrievals above represents the true VCD, the KNMI retrieval for GOME would overestimate by 32% in July and 16% in January
- In addition, OMI VCD by KNMI is positively biased by 0-30% irrespective of season (Boersma et al., 2009, etc.)
- Systematic errors in KNMI retrievals are consistent due to similar retrieval approaches (Lin et al., 2009, etc.)

# Top-down versus Bottom-up Emissions



- Top-down emissions with different approaches result in spatial distributions similar to bottom-up
- The new approach results in emissions closer to the bottom-up, particularly for Jan 2009

## Top-down versus Bottom-up Emissions

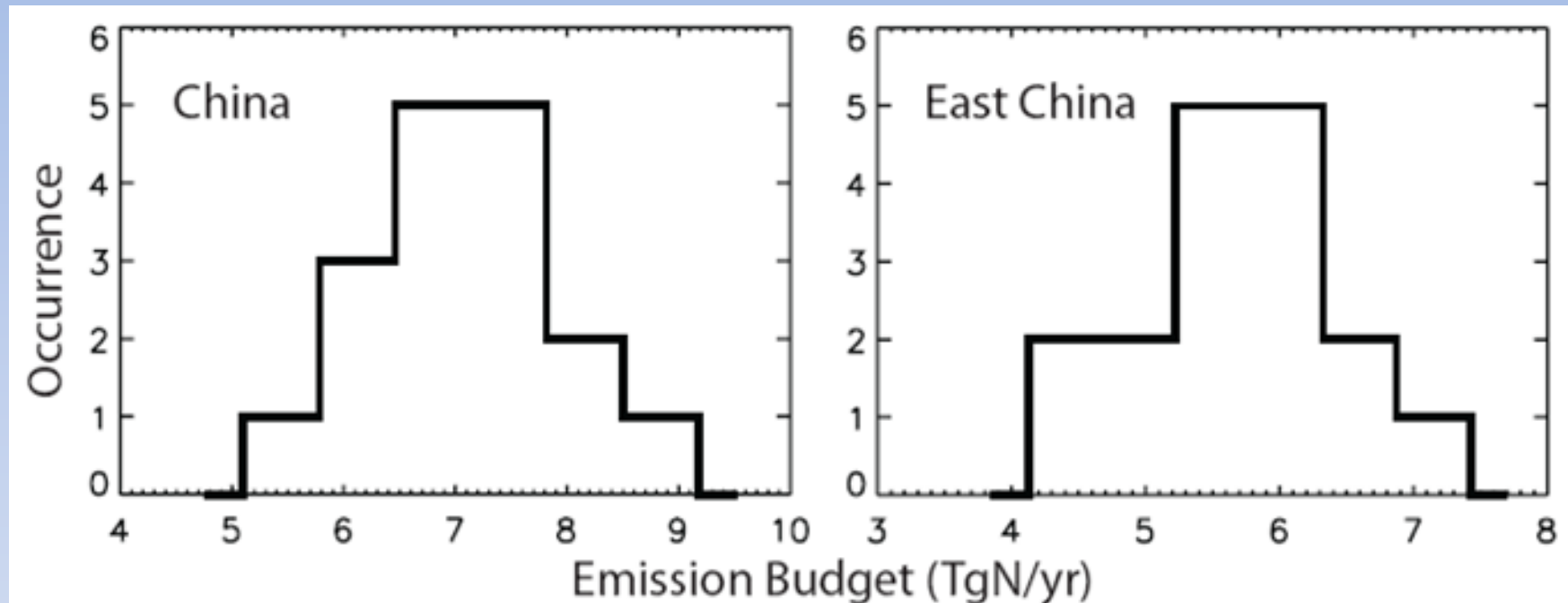
Month	Bottom-up	Top-down for East China		
		OMI (M)	GOME-2 (M)	GOME-2 + OMI (L)
July 2008 (TgN/yr)	5.9	7.5	9.5	6.6
January 2009 (TgN/yr)	6.8	11.3	14.8	8.0
January to July ratio	<u>116%</u>	<u>152%</u>	<u>157%</u>	<u>122%</u>

- As compared to Martin et al. method, the new approach leads to:
- Top-down emission budgets closer to bottom-up
  - Seasonal variation similar to bottom-up

## Sensitivity Analysis for East China

Case	Description	Emis.
(1) <i>a priori</i>	<i>a priori</i>	5.7
(2) TD: best estimate	best estimate	5.5
(4) TD: upper limit	no emission diurnal variations	6.6
(5) TD: lower limit	emission variations use the U.S. EPA 1989 estimation	4.9
(6) TD: adj_GOME-2	GOME-2 scaled down by 113%	6.4
(7) TD: adj_GOME-2 & OMI	GOME-2 reduced by 32% and OMI reduced by 15%	5.8
(8) TD: SCIA+OMI	SCIAMACHY in place of GOME-2	6.0
(9) TD: fullmix	assume full mixing PBL	5.7
(10) TD: 2xltg	lightning NOx emissions doubled	4.6
(11) TD: 1.5xHC	Anthro. emissions of CO and VOCs increased by 50%	5.3
(12) TD: dv_emis	best estimate emis. diurnal variations applied to the <i>a priori</i>	6.0
(13) TD: 1.5xpow	budget uncertainty for power plants increased by 50%	5.5
(14) TD: 1xsoil	similar to (2), but with soil emissions unchanged	6.0
(15) TD: 3xsoil	similar to (2), but with soil emissions tripled	5.0

## Uncertainties in Top-down Emissions (July 2008)



- Most deviations are within 15% of the best estimate
- Some errors may not be random/independent, however, for purposes of estimating top-down uncertainties based simply on these values

# Conclusions

- The new method for constraining surface anthropogenic emissions of NO<sub>x</sub> using satellite remote sensing data is an important improvement over the widely used Martin et al. method:
  - Incorporating information from *multiple instruments*
  - *Less susceptible* to systematic retrieval errors
  - Distinguishing *individual sources* simultaneously
  - Explicit treatment for diurnal variations of NO<sub>x</sub> emissions, column and lifetime; and no nighttime evolution involved
- Consequently, the new method results in top-down emissions of NO<sub>x</sub> and seasonal variation *in better agreement* with the current understanding from the bottom-up perspective.
- Individual errors in retrievals, model, and methodology typically result in uncertainties of top-down estimate by less than 15%.