Decadal changes in global NOx, CO, and SO_2 emissions derived from multi-model multi-constituent satellite data assimilation

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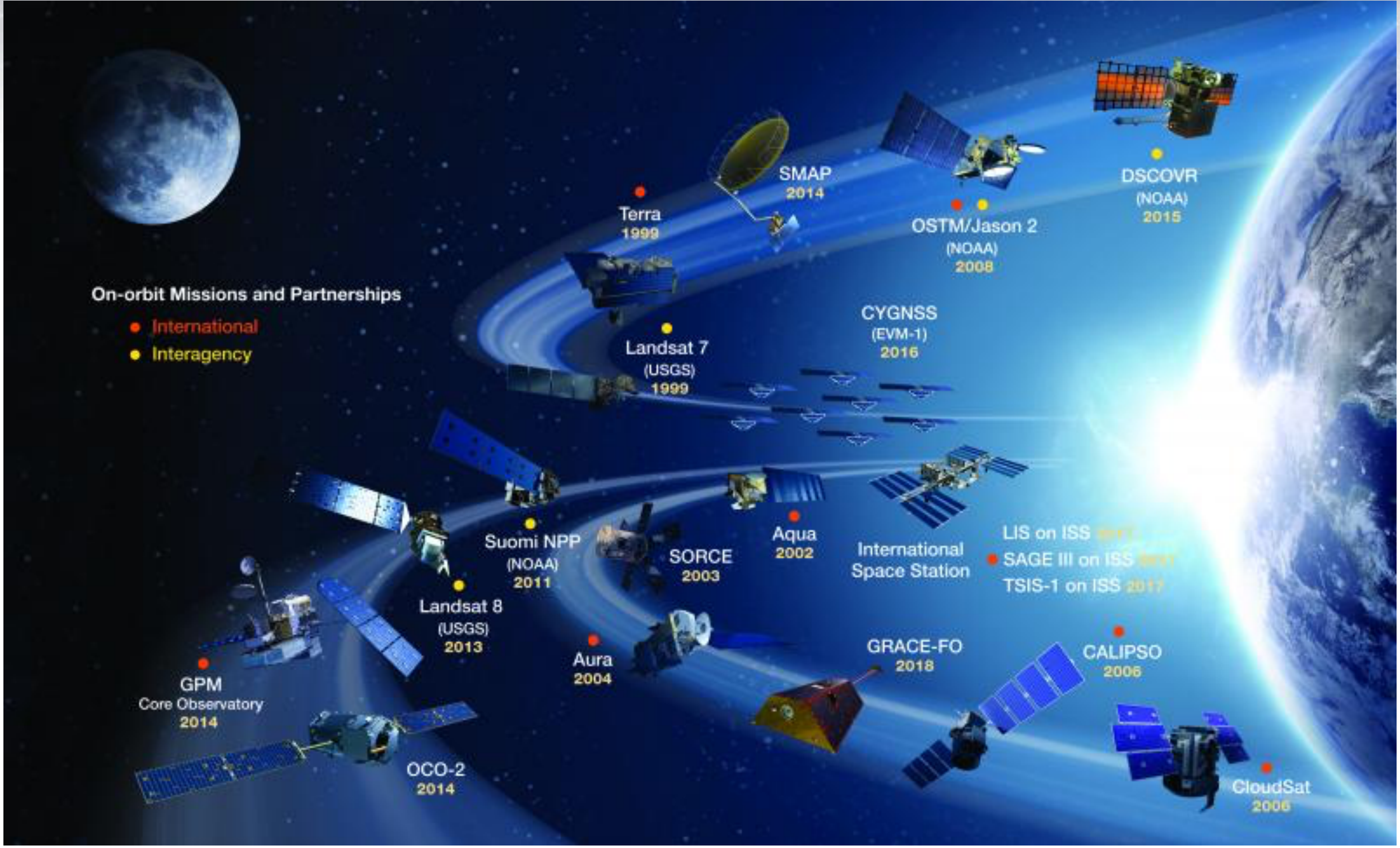
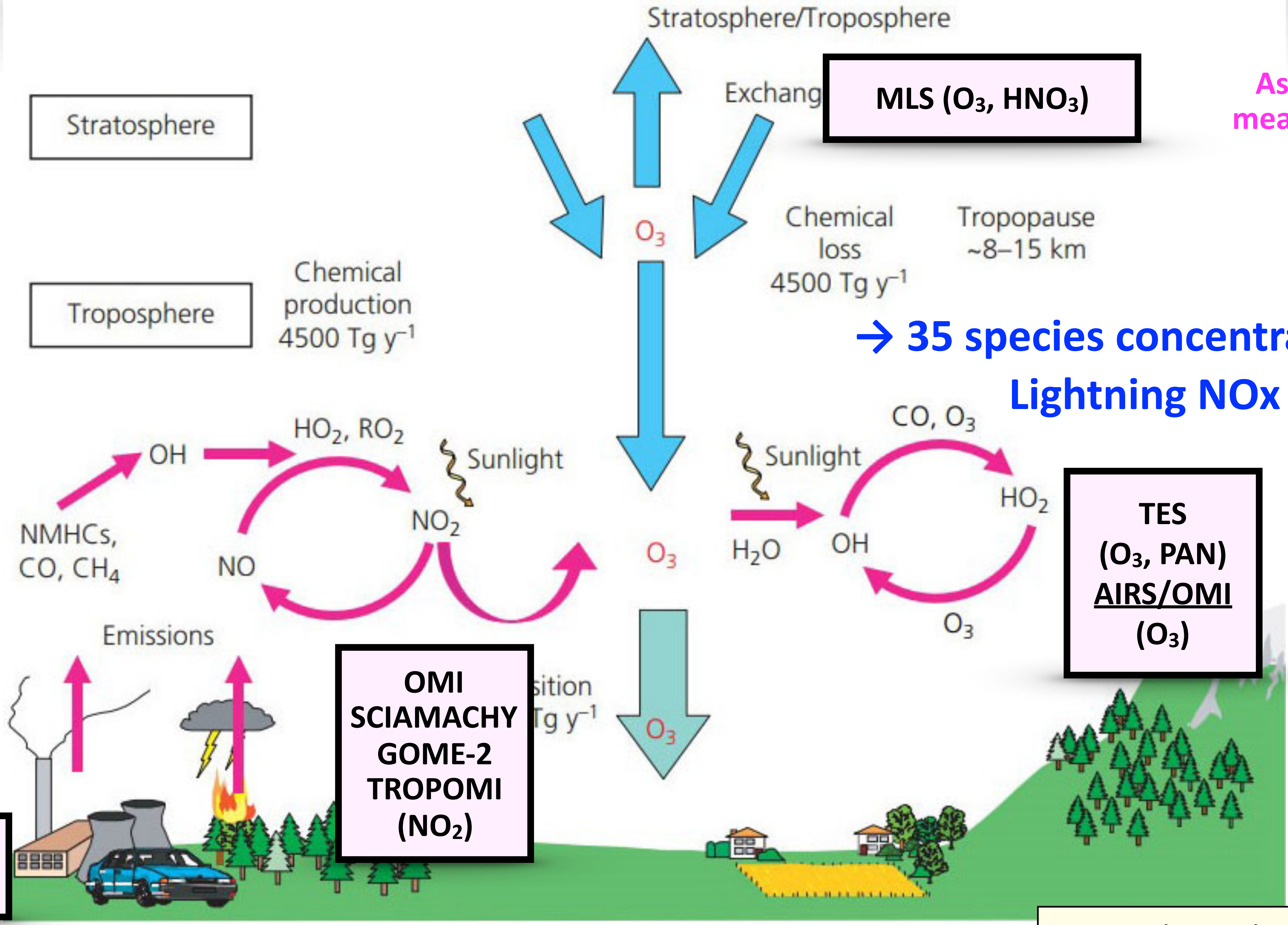
Multi-constituent chemical data assimilation



- make best use of all available data, from heterogeneous sensors
- extend analysis on non-observed space/species & emissions
- produce consistent long-term integrated datasets

Multi-constituent chemical data assimilation

EnKF data assimilation to integrate a suite of measurements from multiple satellite sensors



Assimilated measurements

MLS (O_3, HNO_3)

→ **35 species concentrations + Lightning NOx**

TES
(O_3, PAN)
AIRS/OMI
(O_3)

OMI
SCIAMACHY
GOME-2
TROPOMI
(NO_2)

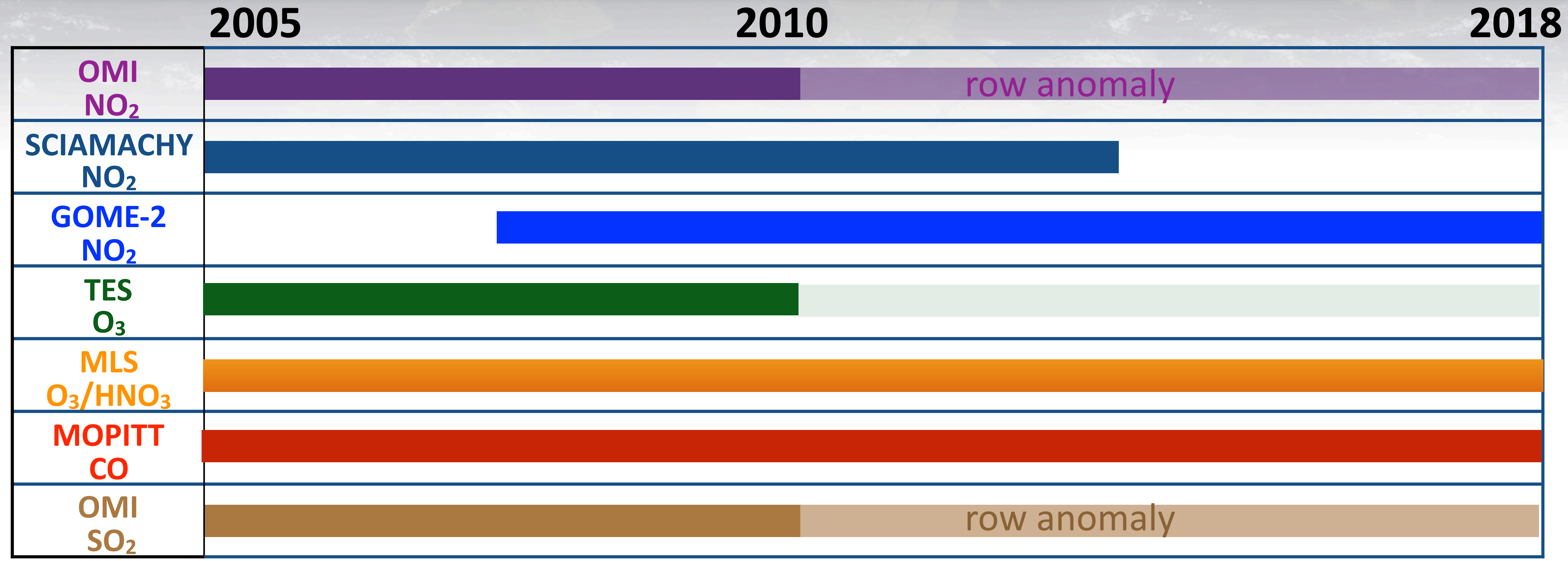
MOPITT
(CO)

OMI (SO_2)

→ **NOx, CO, SO_2 emissions**

Miyazaki et al., 2012, 2013, 2014, 2017, 2019

Tropospheric chemistry reanalysis (TCR-2)

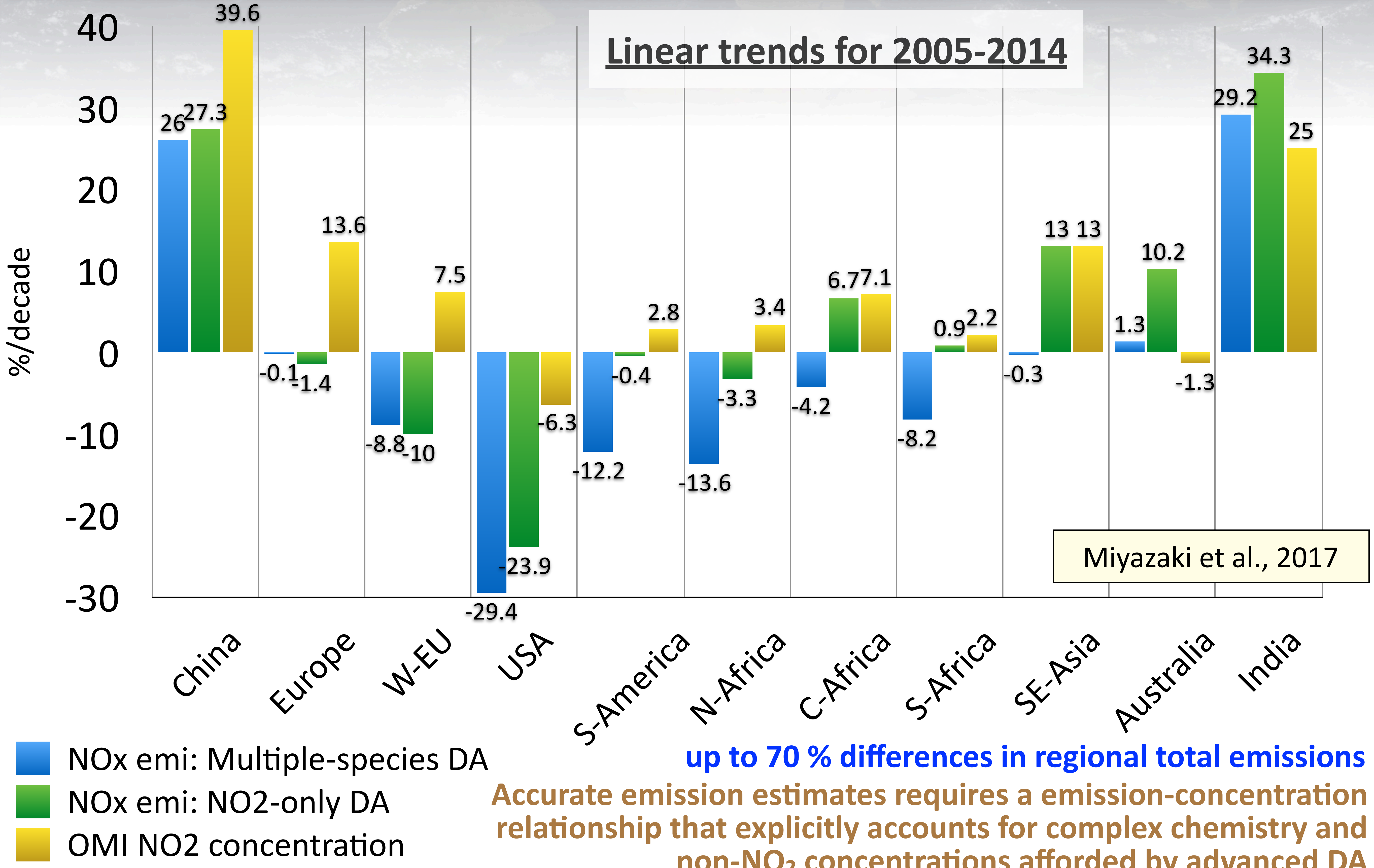


Two-hourly, 1.1°x1.1° resolution, up to 70 hPa level

(1) understand the processes controlling the atmospheric environment
(2) provide initial/boundary conditions for climate/chemical simulations
(3) evaluate climate models and bottom-up emission inventories
(4) suggest developments of models/observations (e.g., satellite concepts)

O_3 (AIRS/OMI)
NO_2 (QA4ECV)
SO_2 (NASA PCA)
NO_2 (QA4ECV)
NO_2 (QA4ECV)
O_3 (v6)
CO (v7J)
O_3,HNO_3 (v4.2)

Multi-constituent constraints on NOx emissions



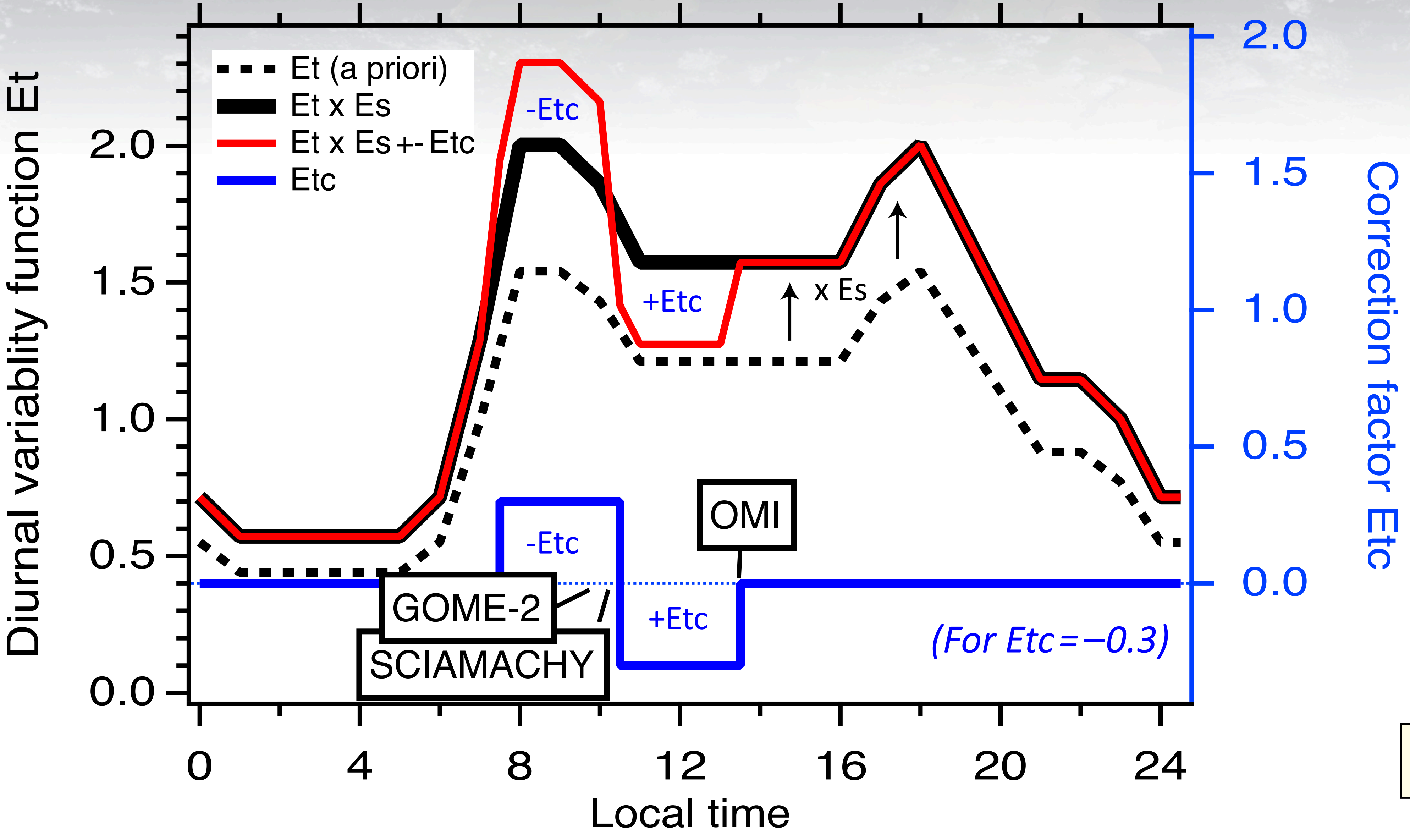
Linear trends for 2005-2014

Miyazaki et al., 2017

up to 70 % differences in regional total emissions

Accurate emission estimates requires a emission-concentration relationship that explicitly accounts for complex chemistry and non-NO_2 concentrations afforded by advanced DA

Multi-sensor constraints on diurnal NOx emissions

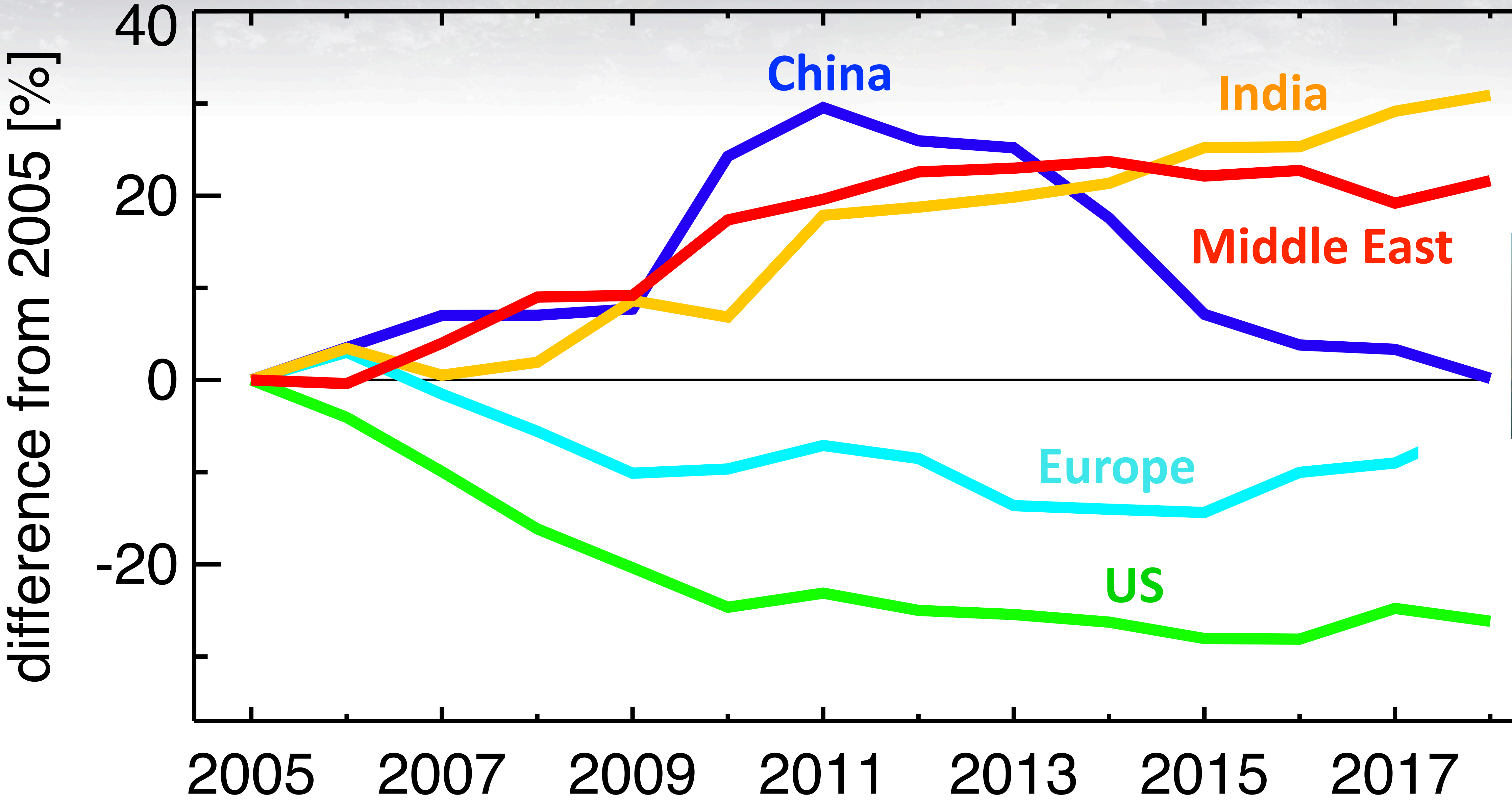


GOME-2 NO_2
SCIAMACHY NO_2
➡ 07:30-10:30
OMI NO_2
➡ 10:30-13:30

Miyazaki et al., 2017

- A correction scheme is applied to modify the shape of the diurnal emission variability using multiple NO_2 measurements obtained at different overpass time.
- **Etc is mostly negative** **->** A larger negative bias in simulated NO_2 in the morning. Larger underestimations in emissions (e.g., morning traffic rush) and/or larger model errors in chemical lifetime.

Global NOx emission trends (2005-2018)



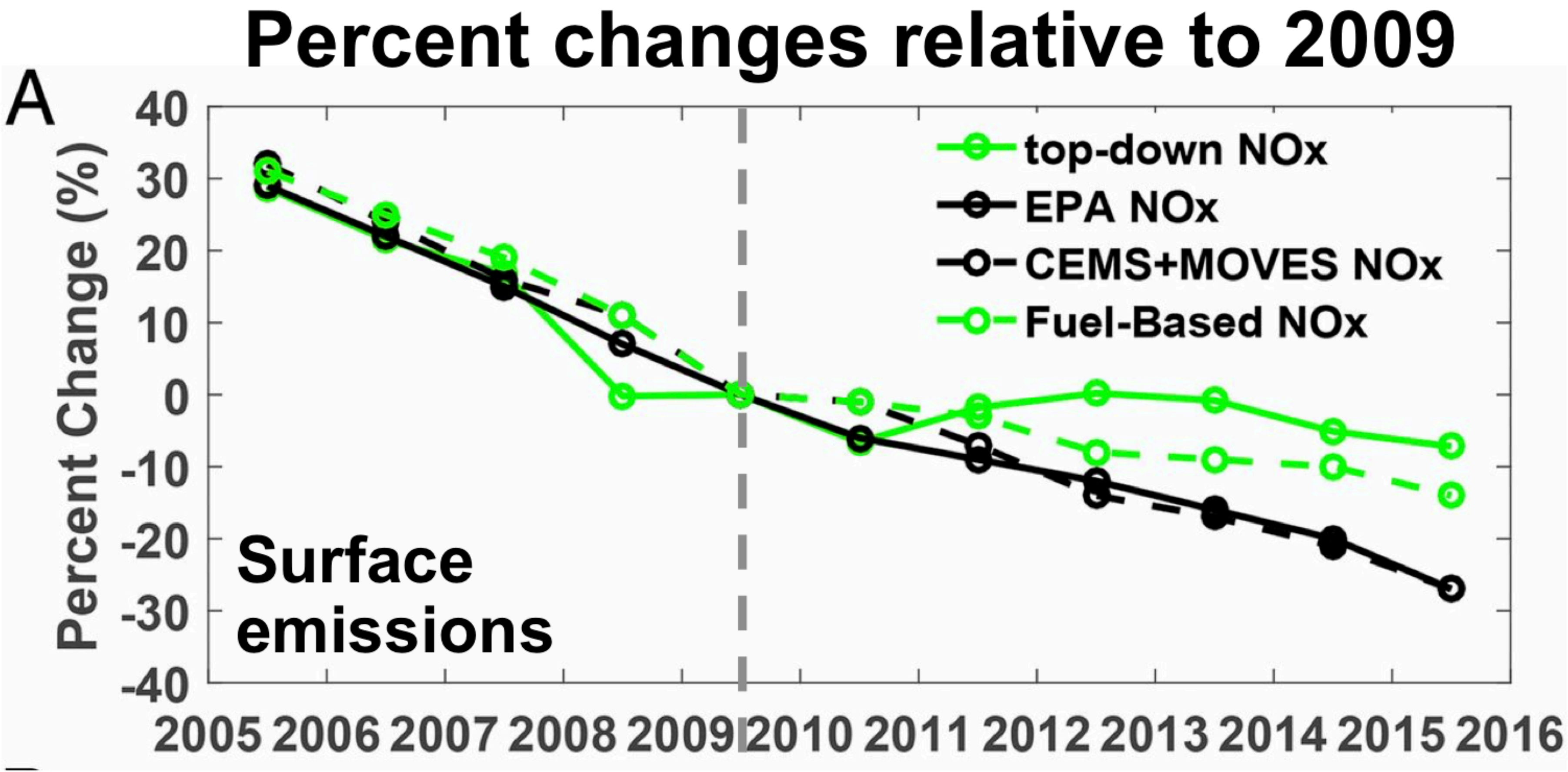
Miyazaki et al.,
2017 & in prep

- **The global total soil emissions are estimated at 7.9 TgN (5.4 TgN in GEIA).** Increased over Australia, the central Eurasian continent, the Sahel, and SW US.

- **Ships emissions are decreased from HTAP v2 by 20-40%**

Unexpected slowdown of US NOx emission reduction

Jiang et al., PNAS, 2018



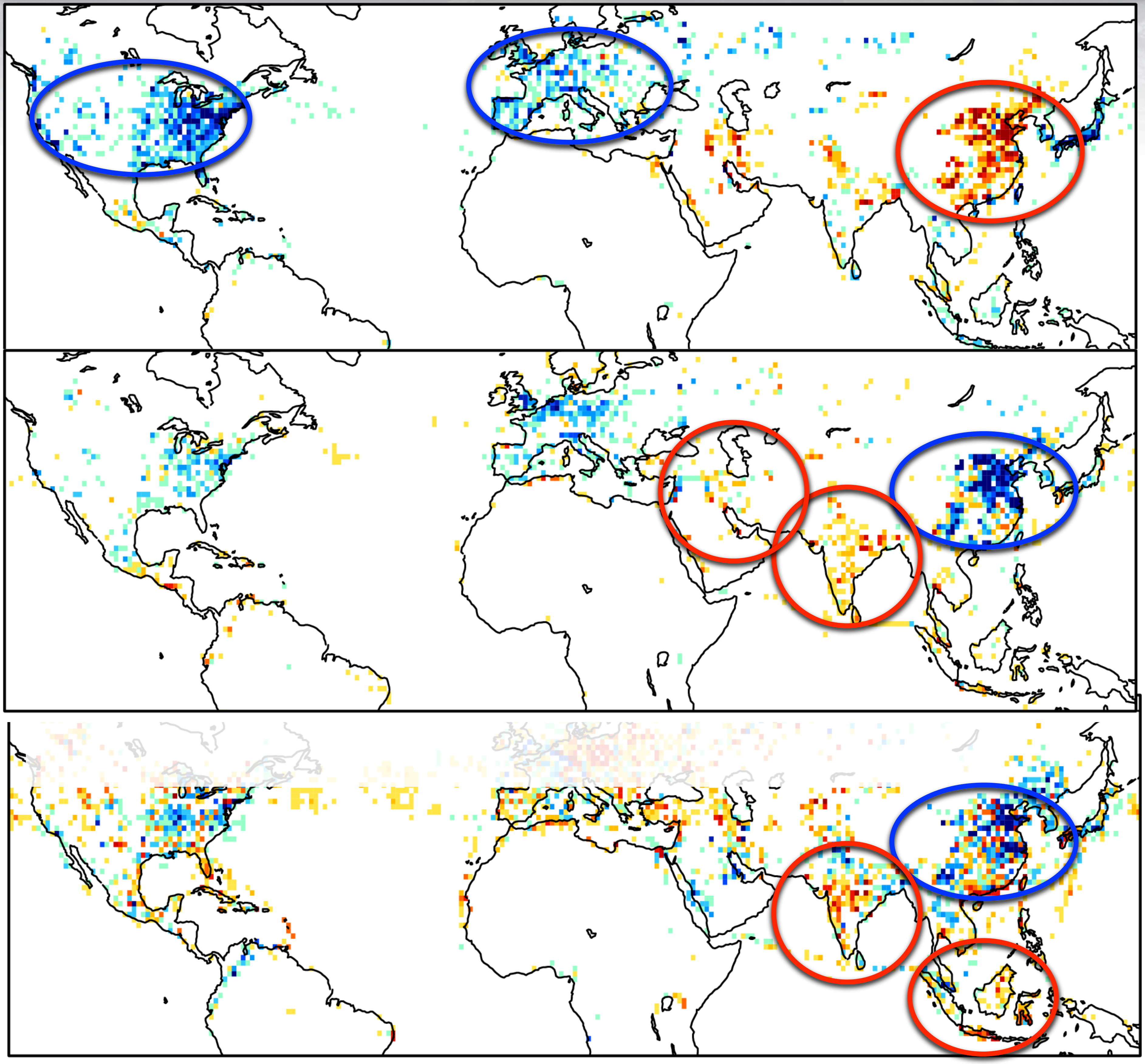
	2005–2009	2009–2016
NO_x top down	$= -7.0 \pm 1.4\%\ a^{-1}$	$= -1.7 \pm 1.4\%\ a^{-1}$
EPA NO_x	$= -6.4\%\ a^{-1}$	$= -5.3\%\ a^{-1}$
Fuel based	$= -6.7\%\ a^{-1}$	$= -2.9\%\ a^{-1}$

Fuel-based NOx bottom up estimates show some flattening in the trend.

Main contributions to fuel-based and NEI trend differences:

1. Off-road vehicles and area sources (industrial & residential) ⬆
2. On-road diesel emissions not decreasing as expected ⬆
3. On-road gasoline vehicles contributing fractionally less and maybe reaching diminishing returns ⬇

Global NOx emission trends (2005-2018)



2005-2010

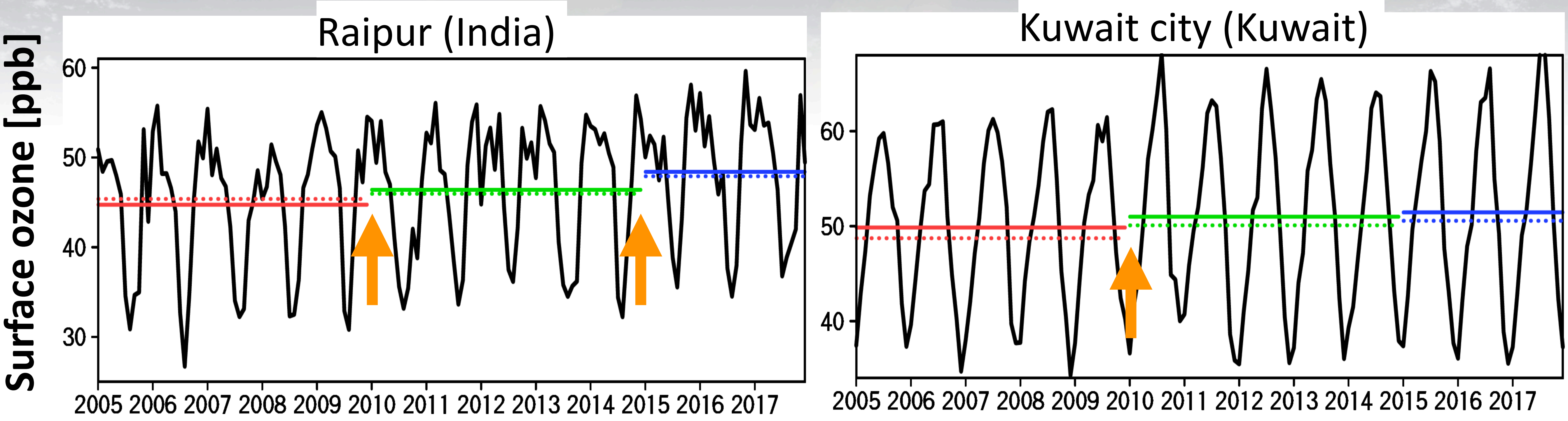
while keeping almost constant global total emissions…

2010-2015

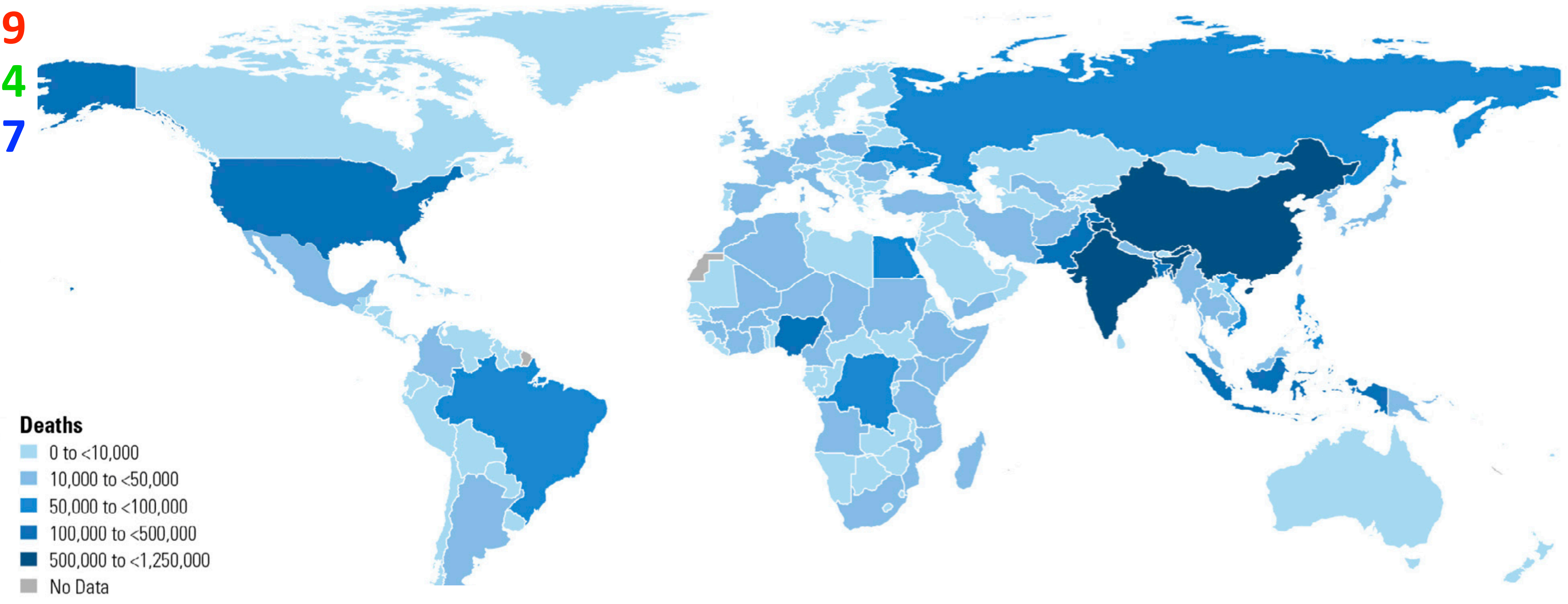
2015-2018

Miyazaki et al., in prep

Implications for air quality and human health

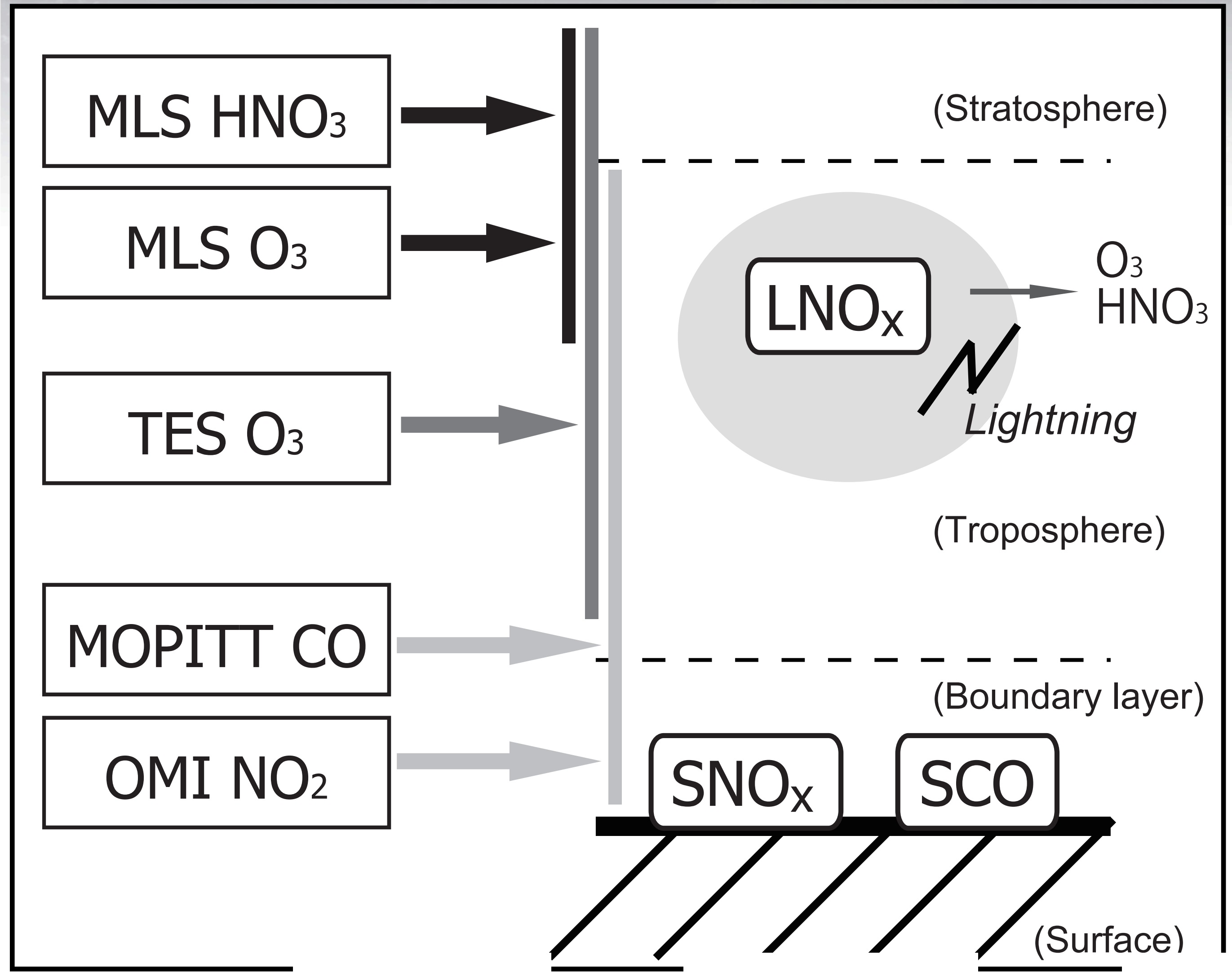


2005-2009
2010-2014
2015-2017

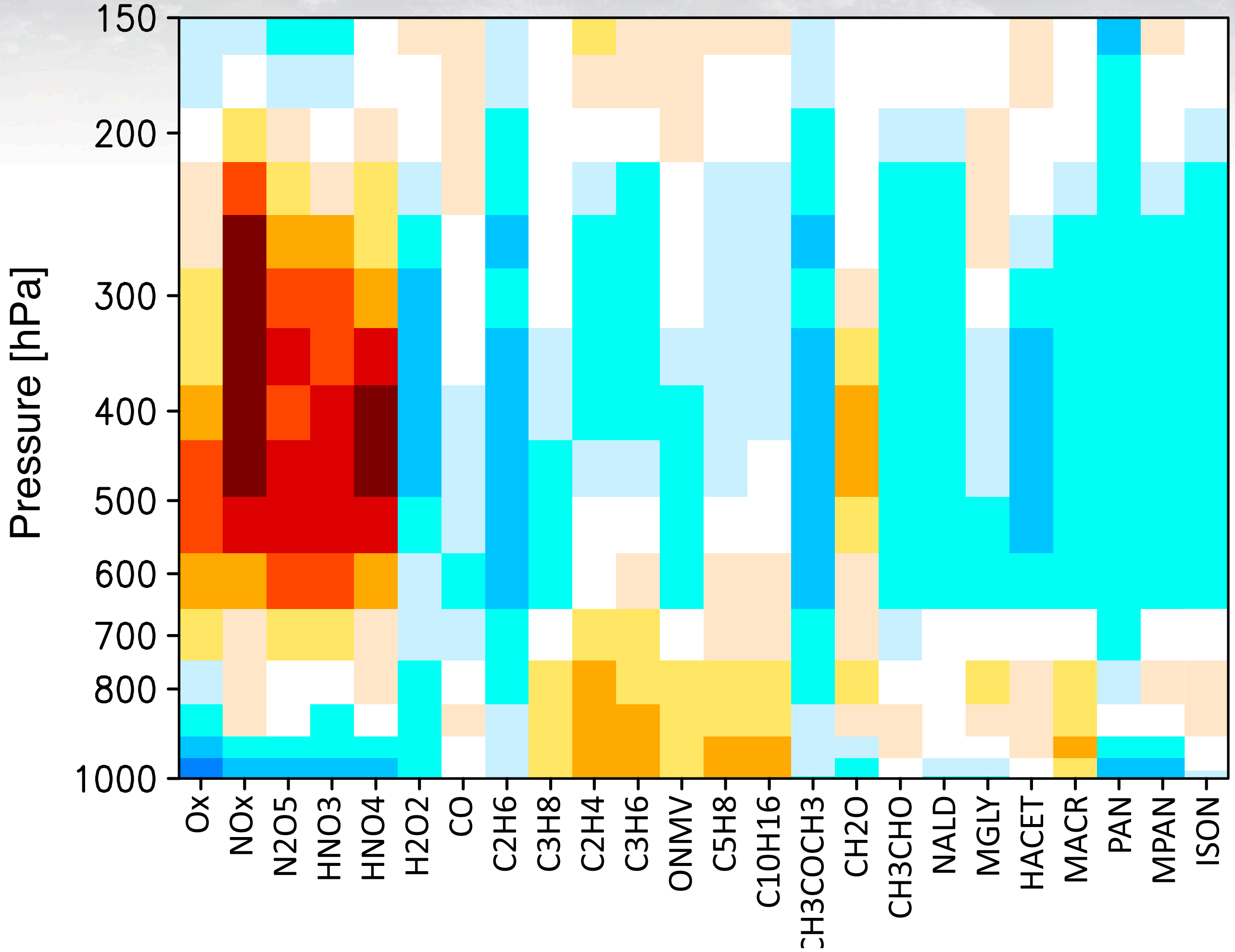


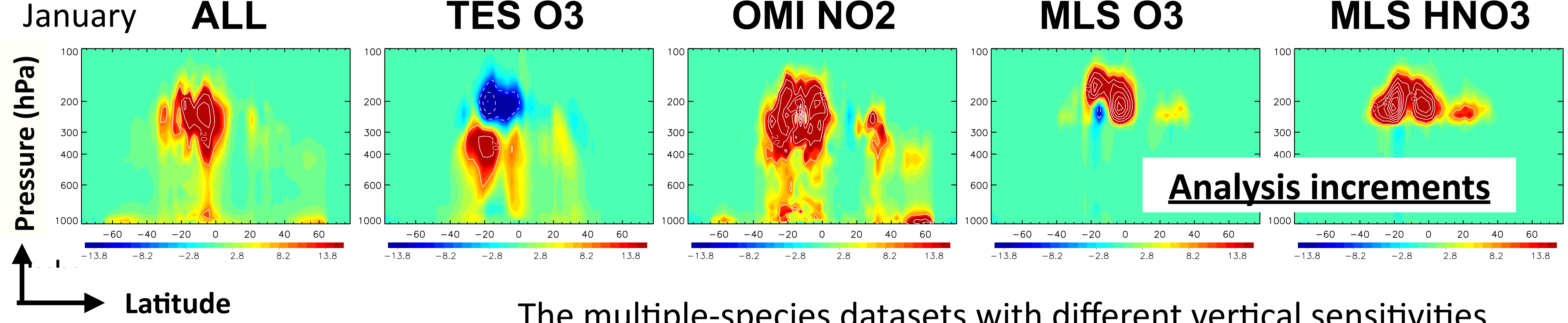
Numbers of deaths attributable to air pollution in 2017 (State of global air 2019)

Lightning NOx sources (2005-2018)



Background error covariance with LNOx



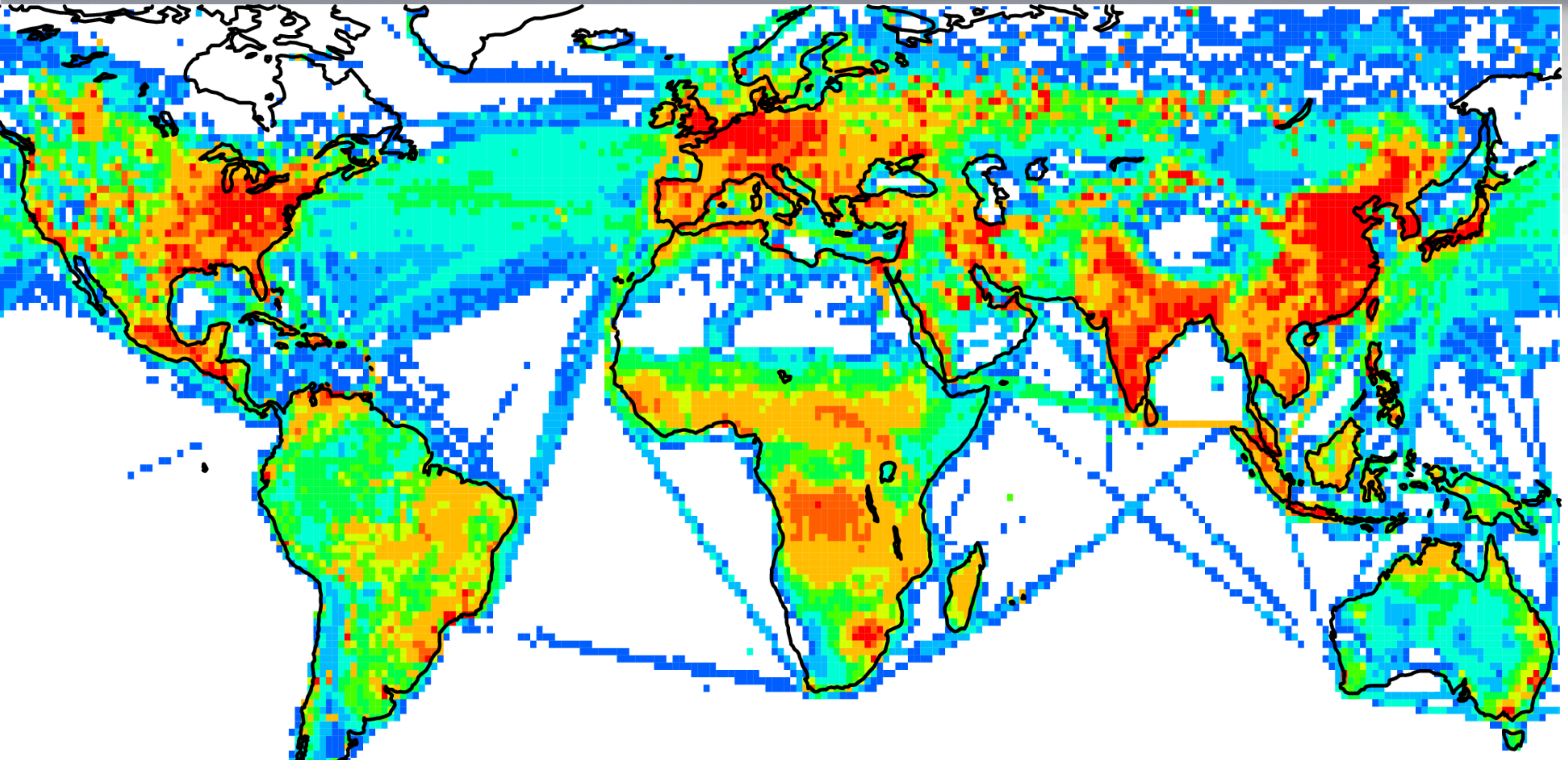


Analysis increments

Miyazaki et al., 2014

The multiple-species datasets with different vertical sensitivities benefits the optimization of the vertical LNOx profile.

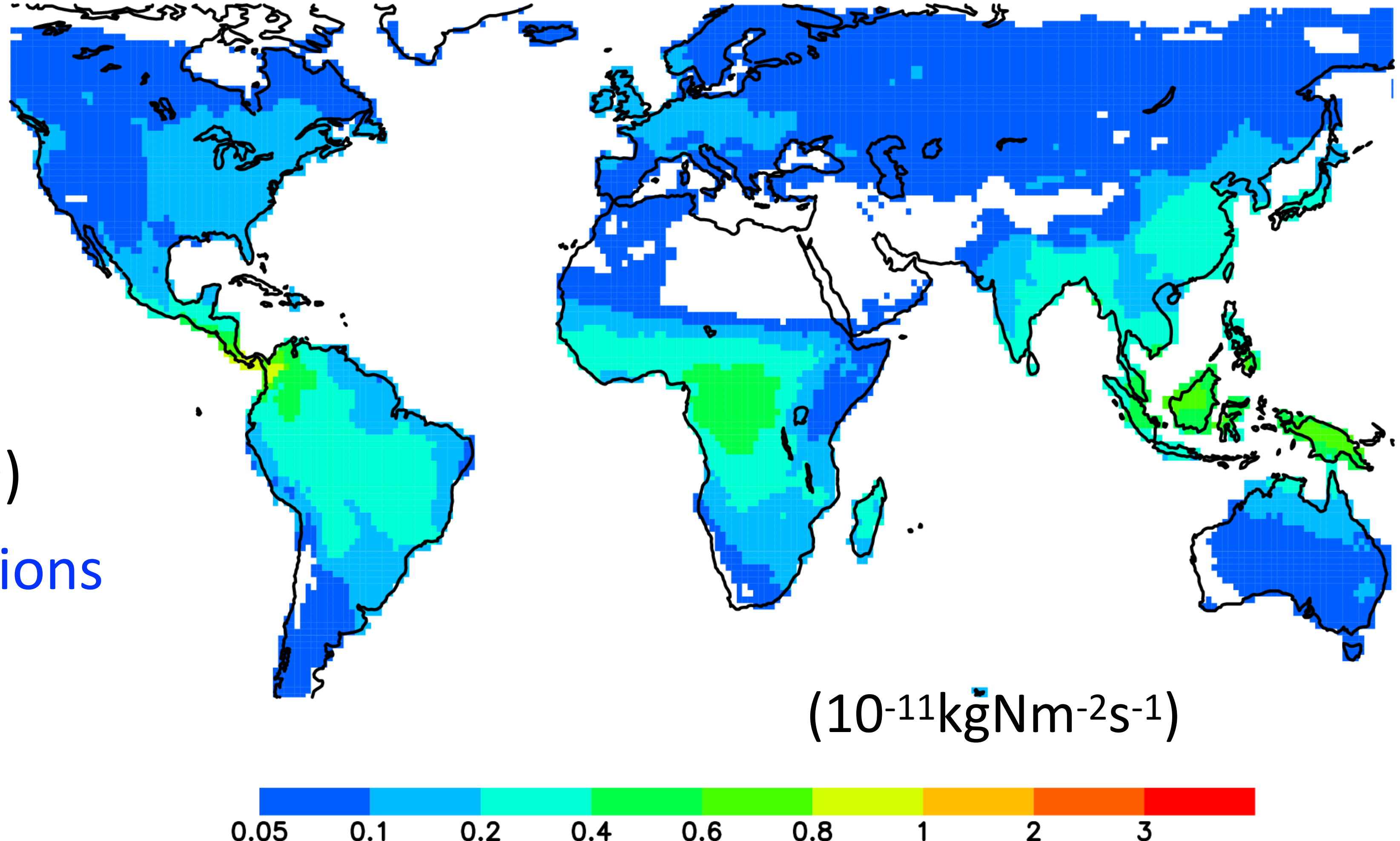
Lightning NOx sources (2005-2018)



Miyazaki et al., in prep

Surface NOx emissions

47.4 TgN (2009) to 50.6 TgN (2015)

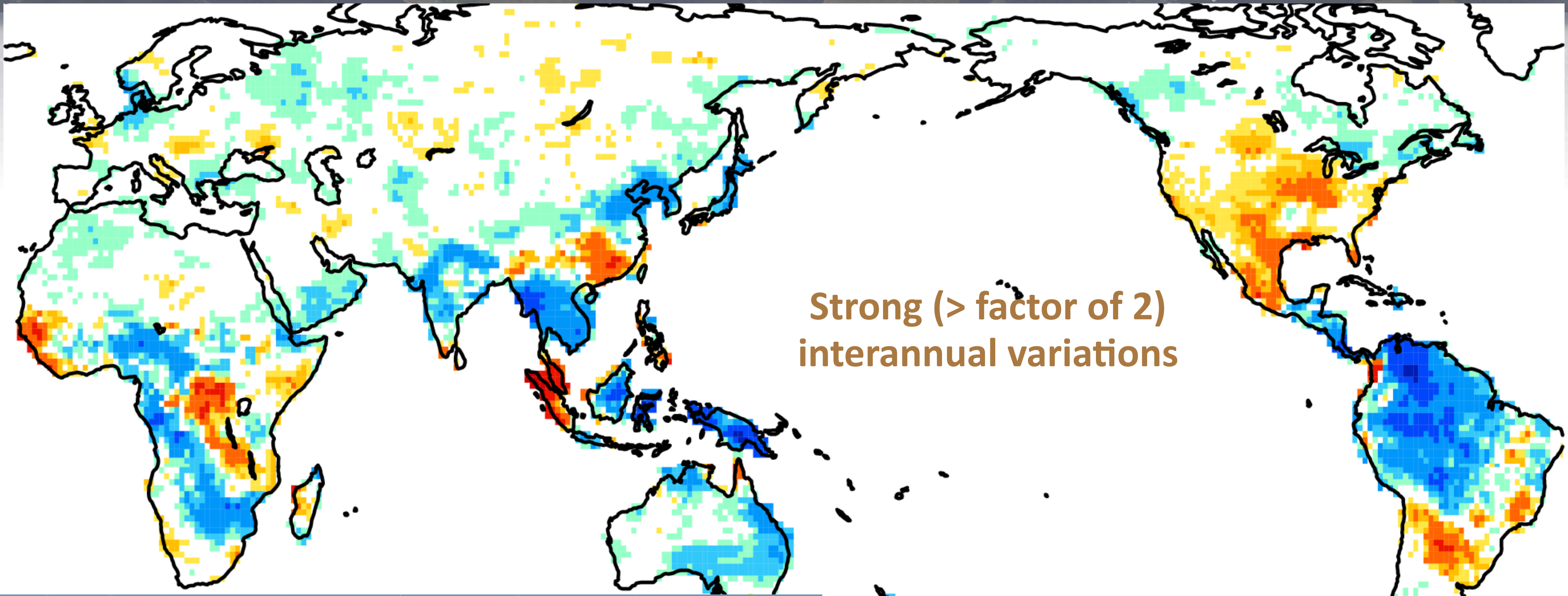


Lightning NOx sources:

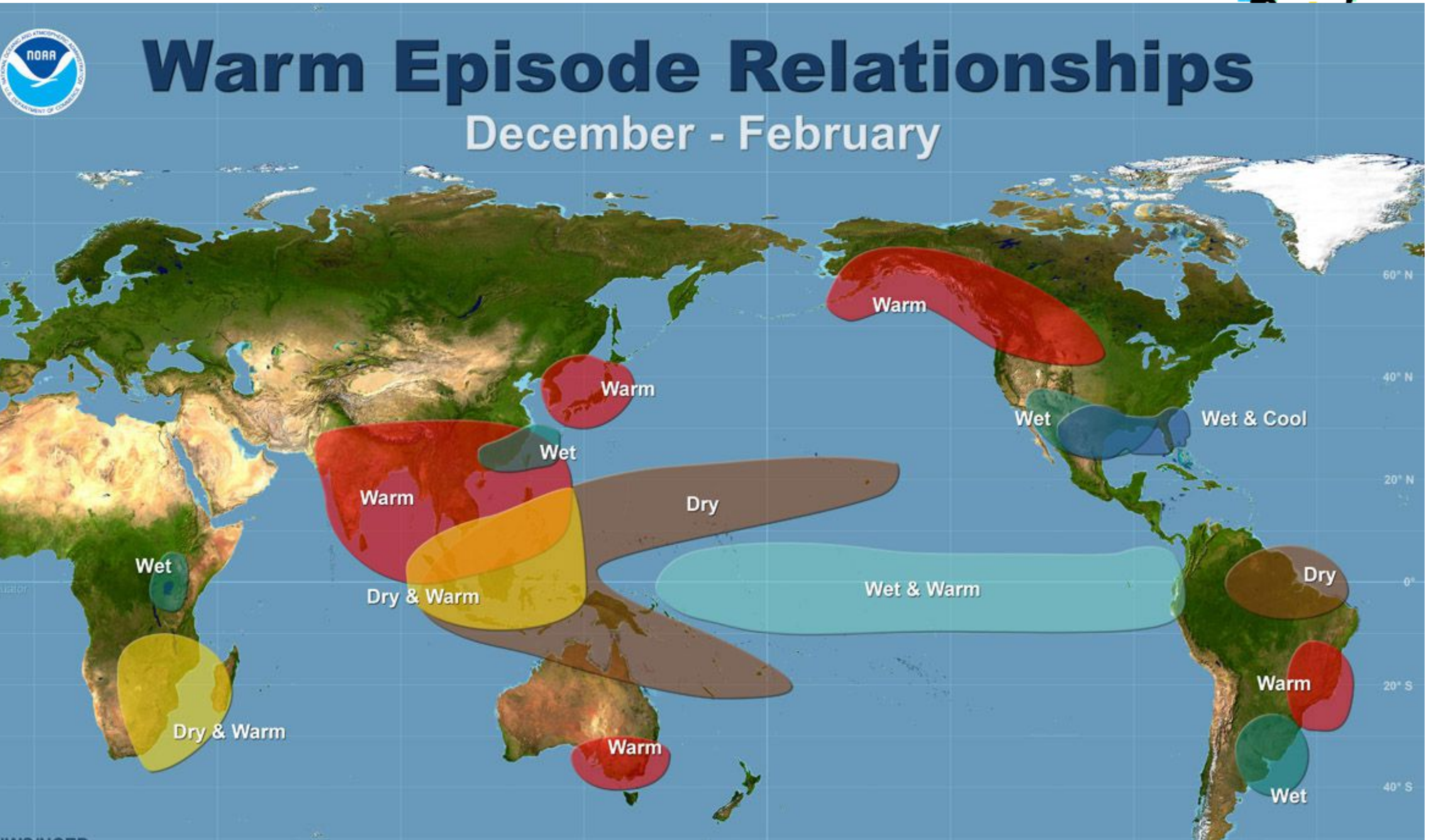
7.1 TgN (2015) to 7.6 TgN (2009)

About 15 % of the total NOx emissions

Lightning NOx: 2015 El Niño anomaly



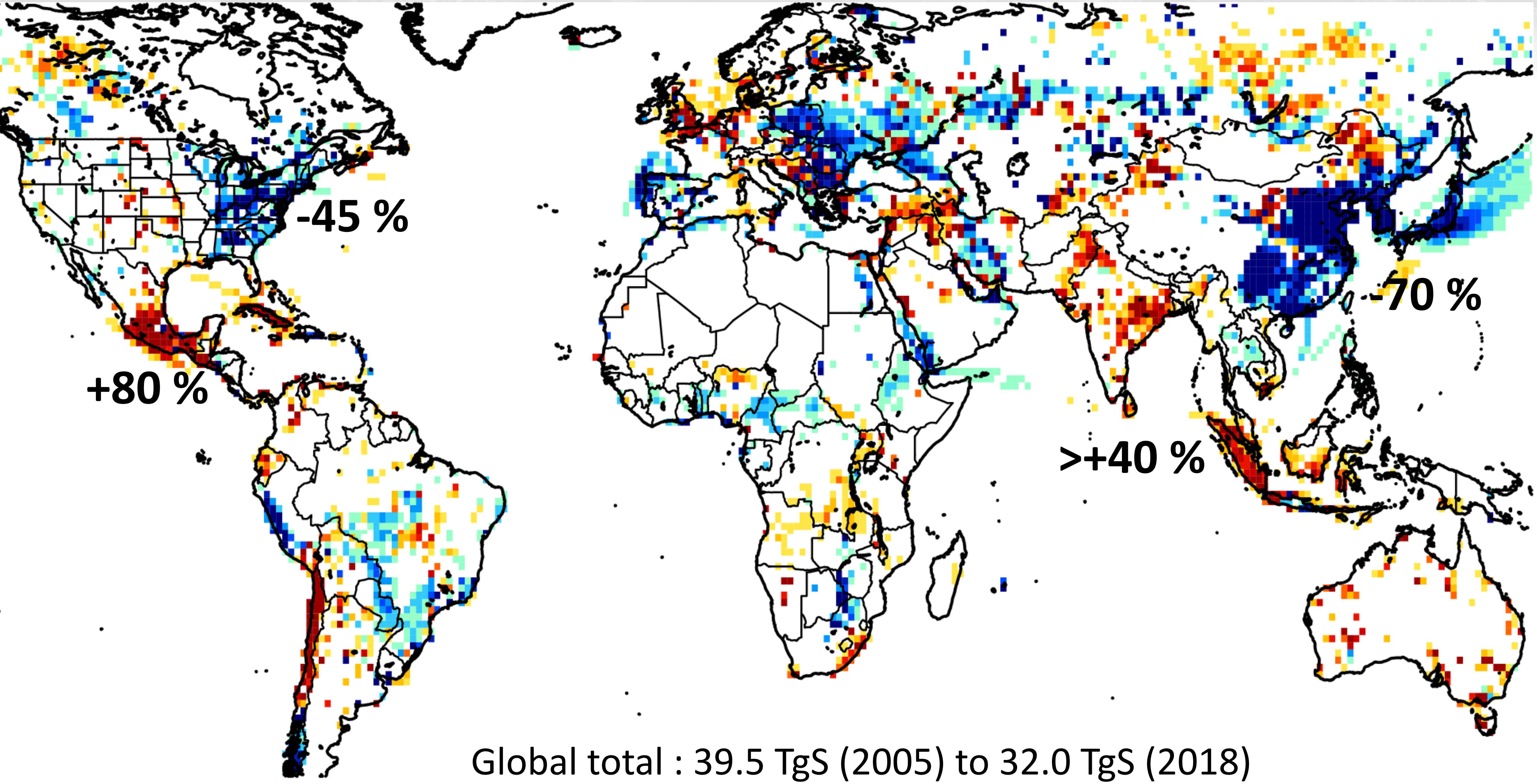
Strong (> factor of 2) interannual variations



Substantial lightning NOx variations need to be considered for accurate estimates of surface NOx emissions using tropospheric NO_2 columns

Miyazaki et al., in prep

Global SO_2 emission trends (2005-2018)



Global total : 39.5 TgS (2005) to 32.0 TgS (2018)

Miyazaki et al., in prep

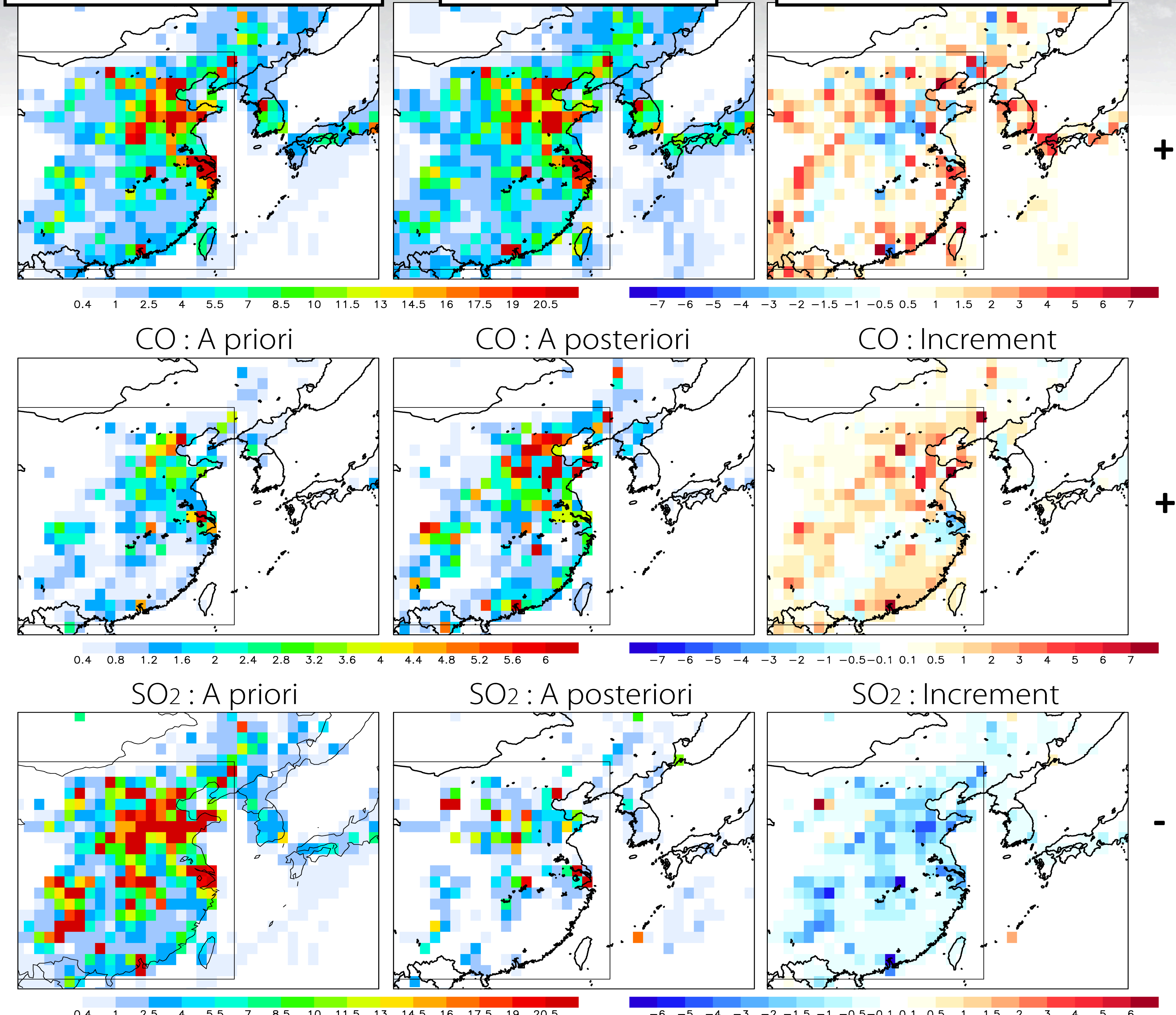
→ *Aerosols, climate, human health*

KORUS-AQ aircraft campaign (May 2016)

A priori (HTAP v2)

A posteriori

Increments



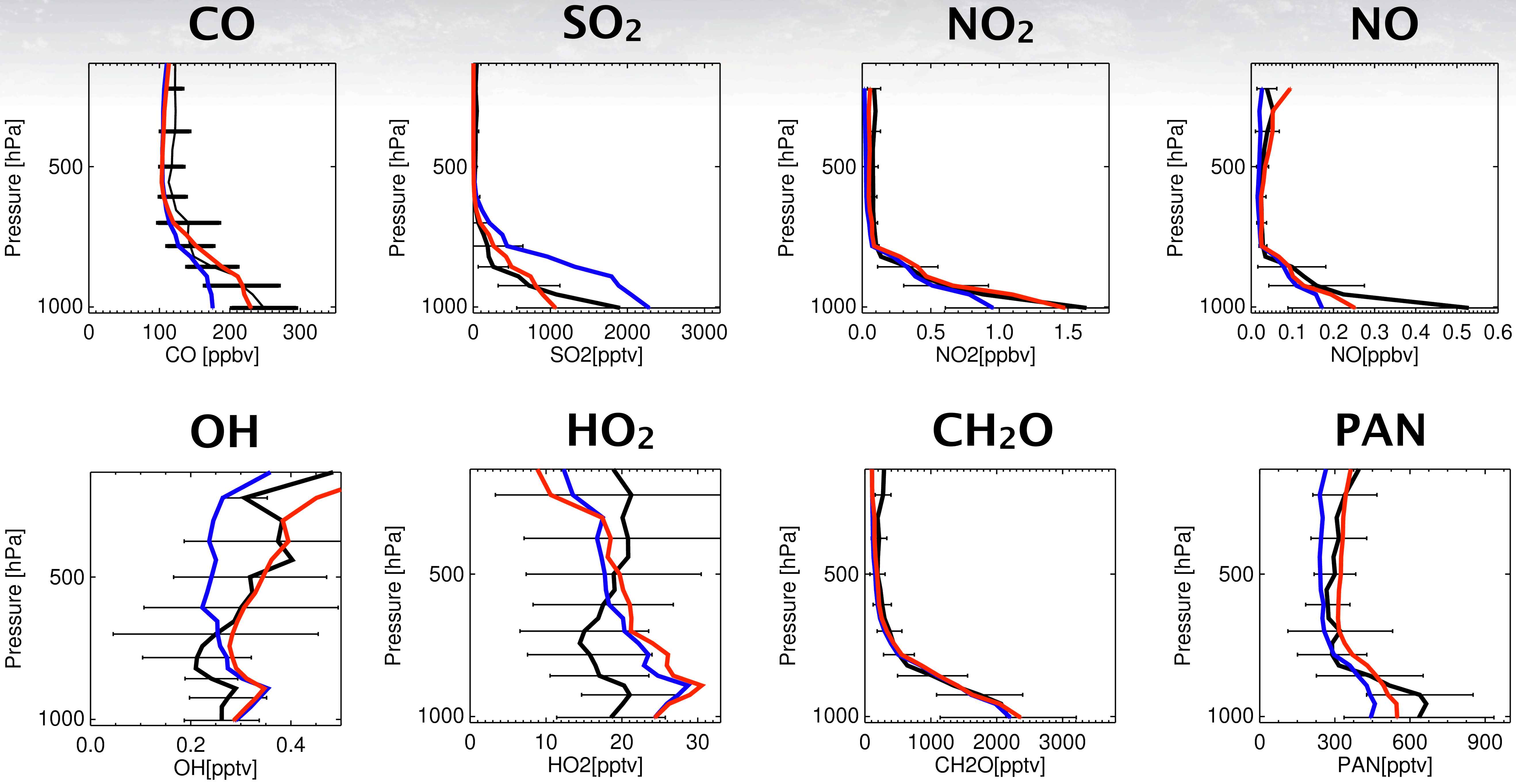
NOx **+75%–94%**

CO **+70%–140%**

SO_2 **- 63%–83%**

Miyazaki et al., 2019a

KORUS-AQ aircraft campaign (May 2016)



CO

SO$_2$

NO$_2$

NO

OH

HO$_2$

CH$_2$O

PAN

outside SMA

DC-8 observation

Model

Reanalysis

Miyazaki et al., 2019a

KORUS-AQ aircraft campaign (May 2016)

	$TgNyr^{-1}$	South Korea			Eastern China		
		NOx	CO	SO_2	NOx	CO	SO_2
Bottom-up	HTAP-v2 2010	0.30	0.6	0.12	7.6	194.6	12.8
	EDGAR 4.2 2008	0.43	2.6	0.8	8.2	107.5	29.8
	KORUS v2	0.30	0.9	0.26	-		
Top-down	This study	0.42	1.1	0.07	8.3	231.3	4.5
	GlobEmission	0.37			6.2		20.9

South Korea

- **NOx** : 40 % higher
- **CO** : 22–80 higher

→ **increased PBL O_3 by 7.5 ppb**

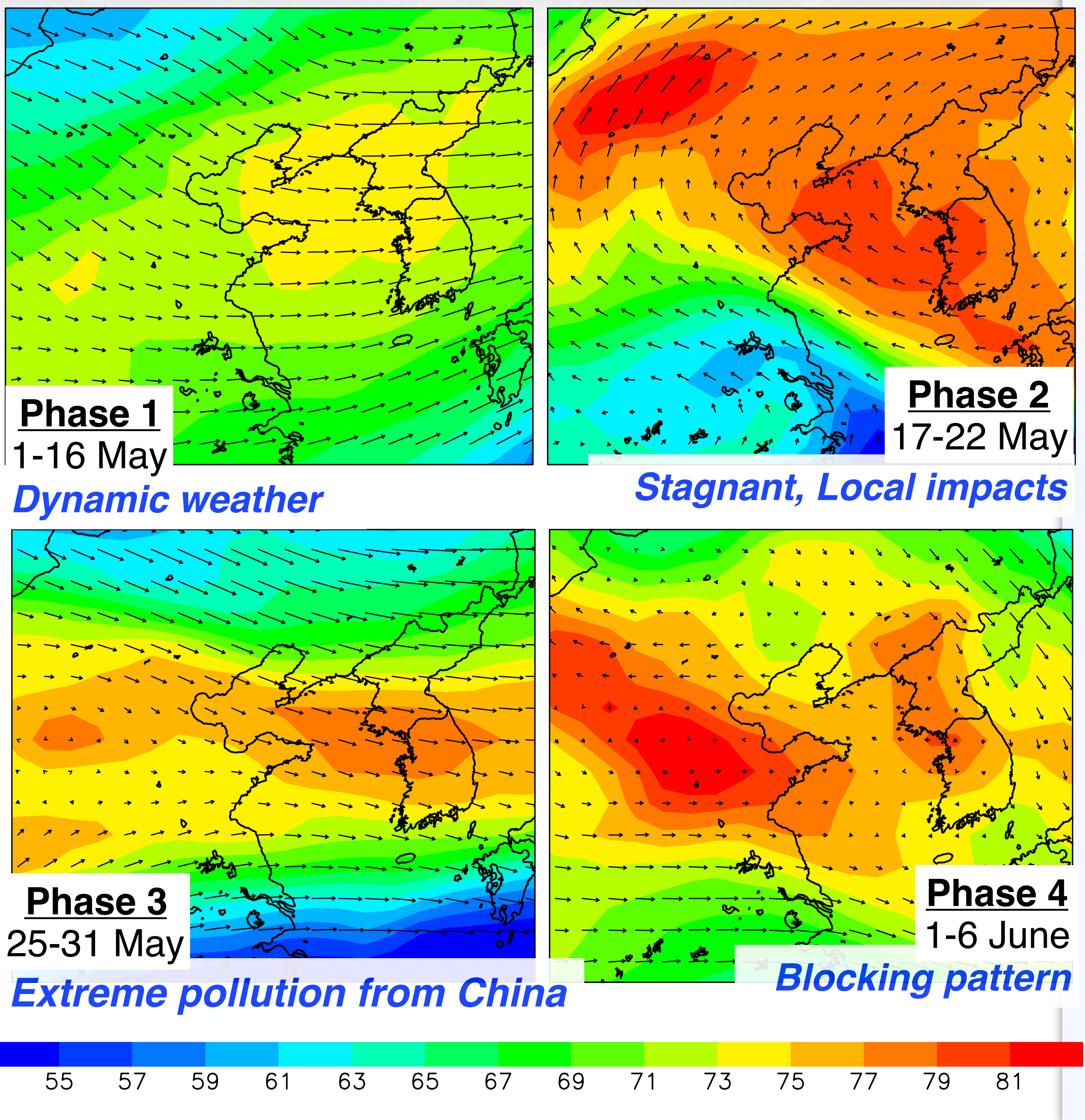
- **SO_2** : 70–83% lower

for an accurate estimate of
the source-receptor relationships
local pollution or long-range transport?

Miyazaki et al., 2019a

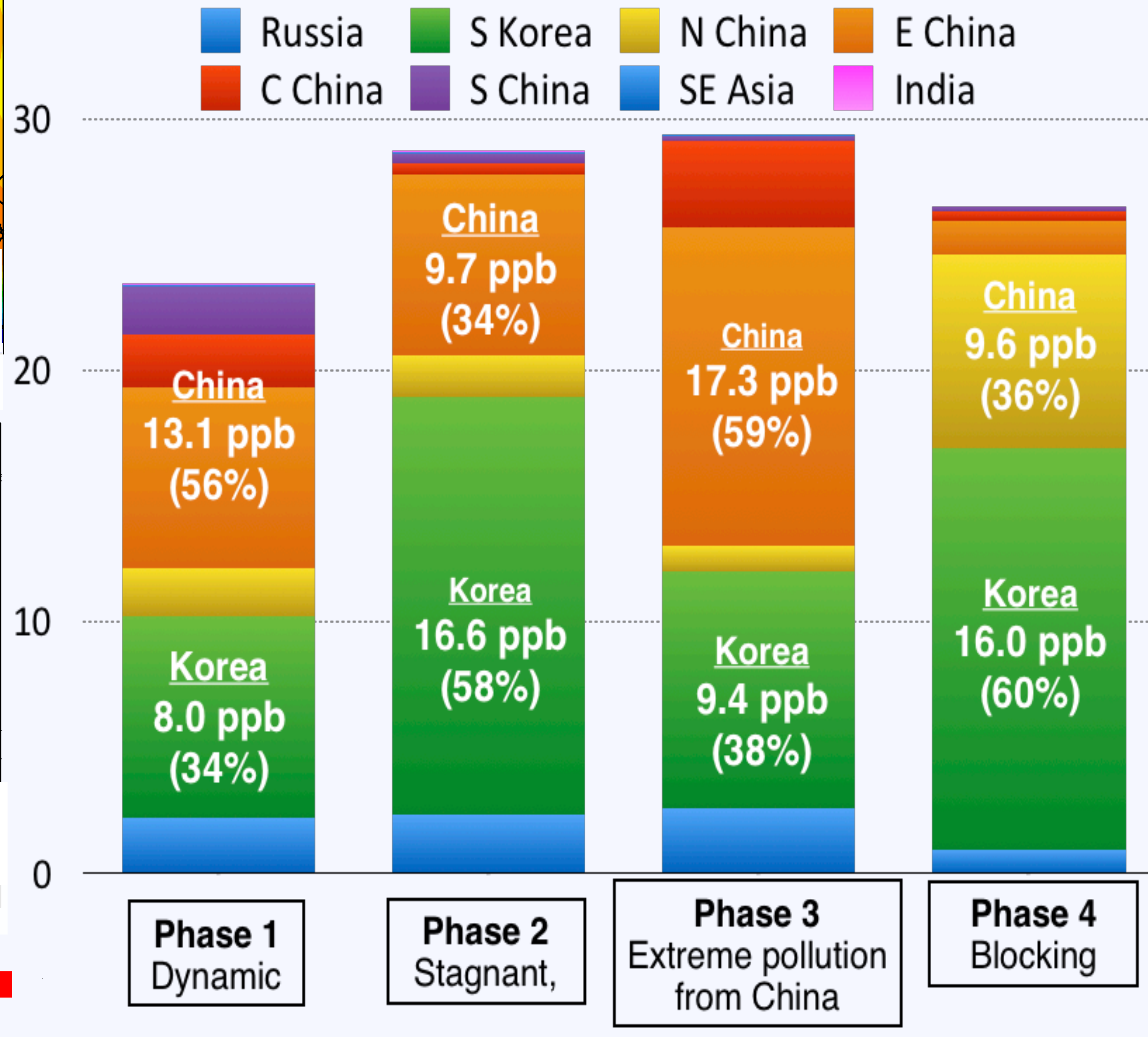
KORUS-AQ aircraft campaign (May 2016)

Reanalysis ozone at 700 hPa



Source-receptor analysis

from NOx emissions to O_3 over Seoul at 900 hPa



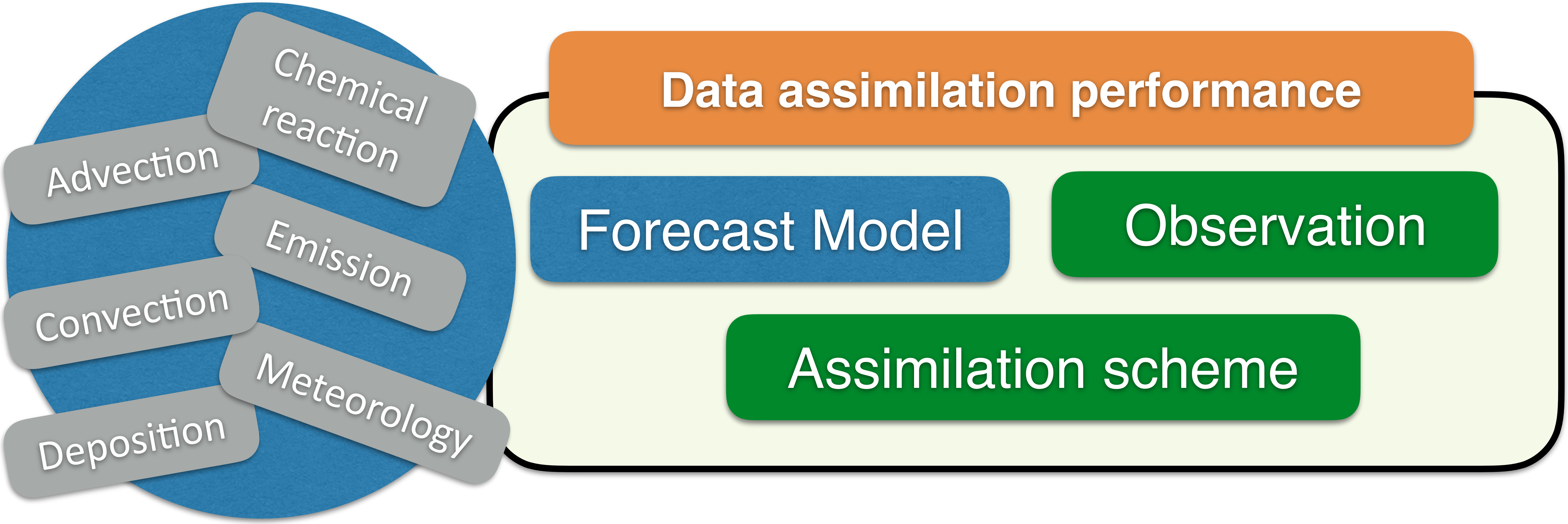
persistently higher over Seoul (76.0±7.9ppbv) than over the broader domain (70.9±9.4ppbv)

Miyazaki et al., 2019a

Multi-model data assimilation integration

Multi-mOdel multi-cOnstituent CHEMical data assimilation (MOMO-Chem)

- investigate the importance of forecast model performance
- provide multi-model integrated data assimilation analysis & possible error ranges in the current top-down emission estimates



Chemical reaction

Advection

Emission

Convection

Meteorology

Deposition

Data assimilation performance

Forecast Model

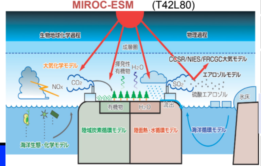
Observation

Assimilation scheme

Miyazaki et al., submitted

GEOS-Chem

CHASER
Chemistry Climate Model

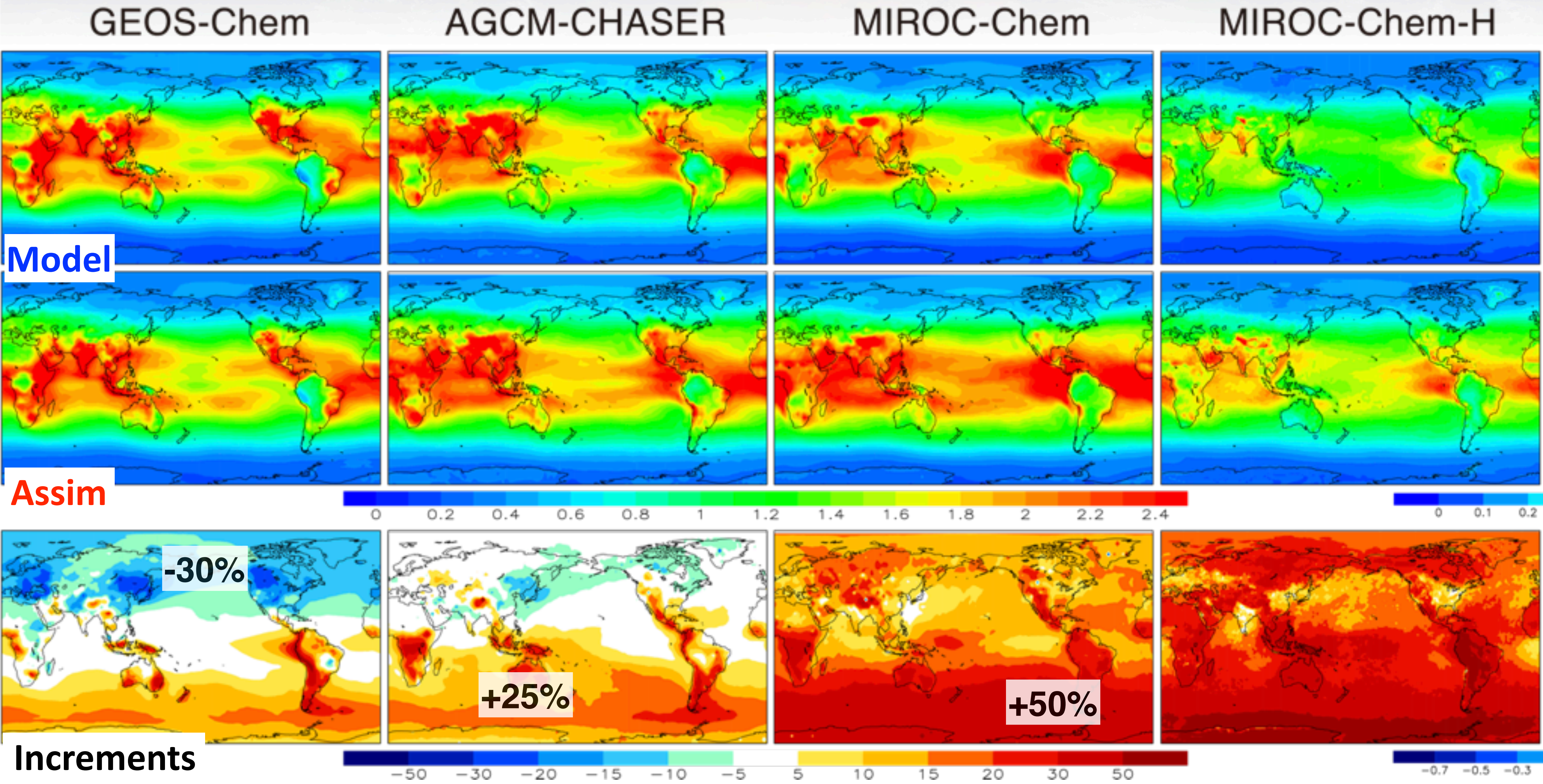


TCR

	1. GEOS-Chem	2. AGCM-CHASER (TCR-1)	3. MIROC-Chem	4. MIROC-Chem-H (TCR-2)
Horizontal resolution	2°x2.5°	2.8°x2.8°	2.8°x2.8°	1.1°x1.1°
Vertical resolution	47 layers to 0.1 hPa (hybrid)	32 layers to 4 hPa (sigma)	32 layers to 4 hPa (hybrid)	32 layers to 4 hPa (hybrid)
Forecast model	GEOS-Chem v9 (adjoint v35)	CCSR/NIES/FRCGC AGCM-CHASER	MIROC-Chem	MIROC-Chem
Chemistry	43 species, 318 reactions	47 species, 88 reactions	92 species, 262 reactions	92 species, 262 reactions
Met data	GEOS-5	Nudged to NCEP-2	Nudged to ERA-Interim	Nudged to ERA-Interim
A priori emissions	EDGAR, NEI2008, RETRO, GFED2,	EDGAR 4.2, GFED 3.1, GEIA	EDGAR 4.2, GFED 3.1, GEIA	HTAP2, GFED4, GEIA
Assimilated measurements	OMI, SCIAMACHY (DOMINO2), TES (v5), MOPITT (v6 NIR), MLS (3.3)	OMI, SCIAMACHY (DOMINO2), TES (v5), MOPITT (v6 NIR), MLS (3.3)	OMI, SCIAMACHY (DOMINO2), TES (v5), MOPITT (v6 NIR), MLS (3.3)	OMI (QA4ECV, PCA), SCIAMACHY (QA4ECV), TES (v6), MOPITT (v7J), MLS (v4.2)
Assimilated species	O_3, CO, NO_2, HNO_3	O_3, CO, NO_2, HNO_3	O_3, CO, NO_2, HNO_3	O_3, CO, NO_2, SO_2, HNO_3
State vector	Concentrations of 43 species + emissions (NOx, CO, LNOx)	35 species + emissions (NOx, diurnal variability, CO, LNOx)	35 species + emissions (NOx, diurnal variability, CO, LNOx)	35 species + emissions (NOx ,diurnal variability, CO, SO_2, LNOx)
Model reference	Henze et al. (2007)	Sudo et al. (2002)	Watanabe et al. (2011)	Sekiya et al. (2018)
DA reference	Miyazaki et al., in review	Miyazaki et al. 2012a,b 2013, 2014, 2015	Miyazaki et al. 2017	Miyazaki et al. 2019a

Multi-model multi-constituent data assimilation

Tropospheric OH: Annual mean

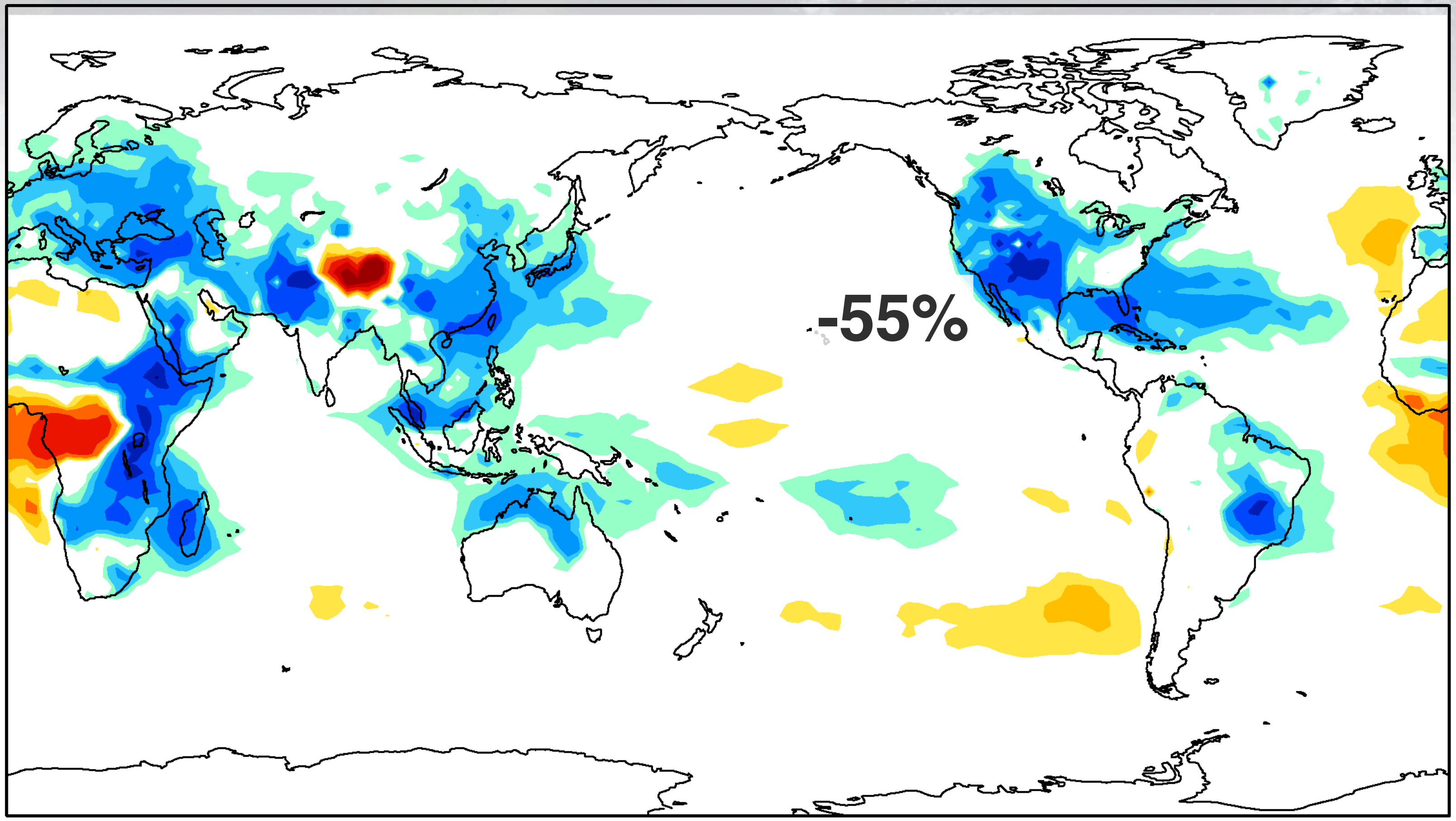


Data assimilation modified global OH distributions,
associated with corrections made to ozone, CO, and NOx.

Miyazaki et al.,
submitted

Multi-model multi-constituent data assimilation

Relative changes [%]
in multi-model OH spreads



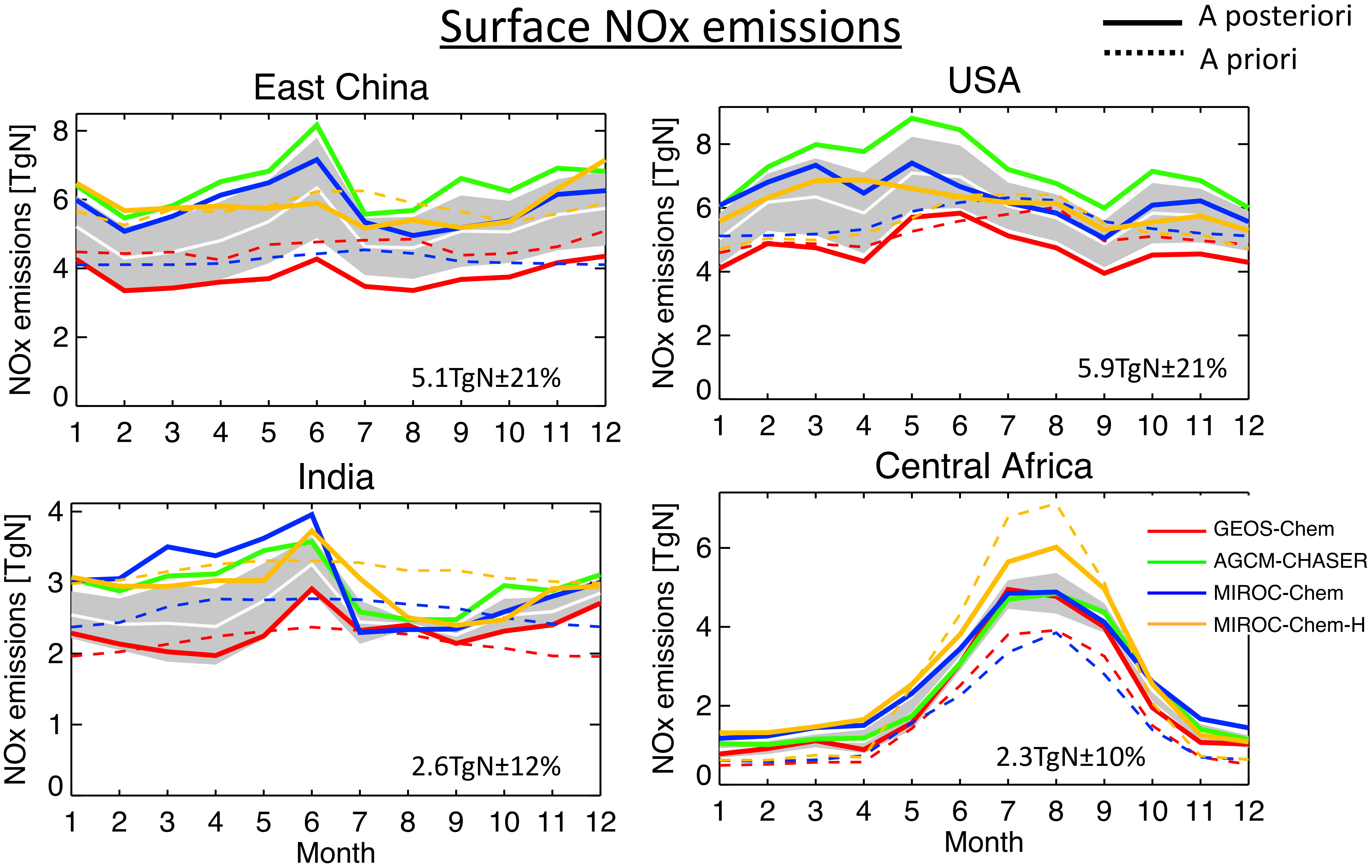
NH-SH OH ratio

	GEOS-Chem	AGCM-CHASER	MIROC-Chem	MIROC-Chem-H	Multi-model
Model	**1.30**	**1.36**	**1.29**	**1.31**	**1.29±0.03**
Assim	**1.16**	**1.23**	**1.18**	**1.21**	**1.18±0.03**

The significant changes in OH are important in modulating the chemical lifetimes
→ *Emission source estimates*

Miyazaki et al., submitted

Surface NOx emissions



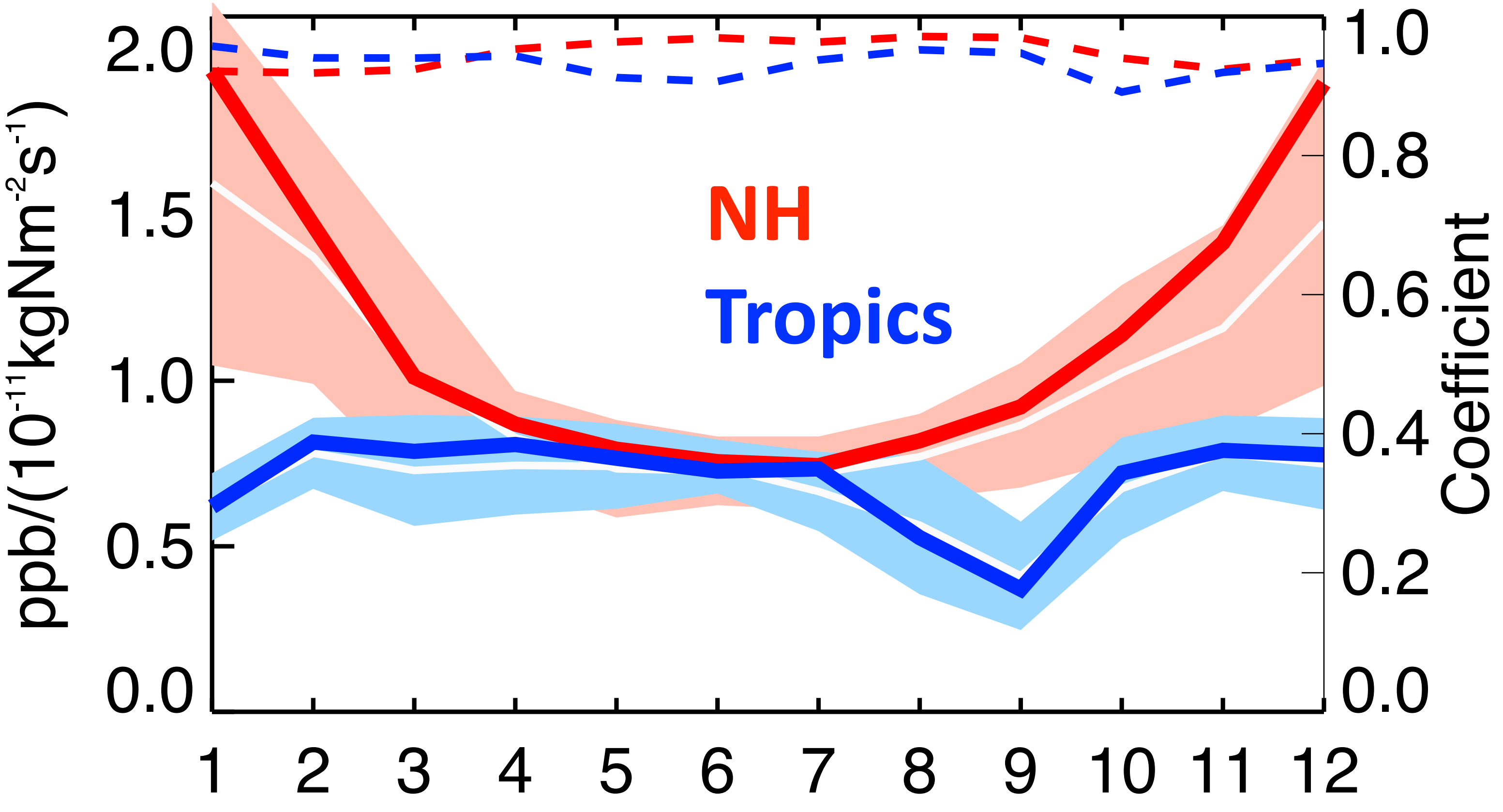
Multi-model standard deviations: 4–31% for regional emissions

= uncertainty ranges due to model errors

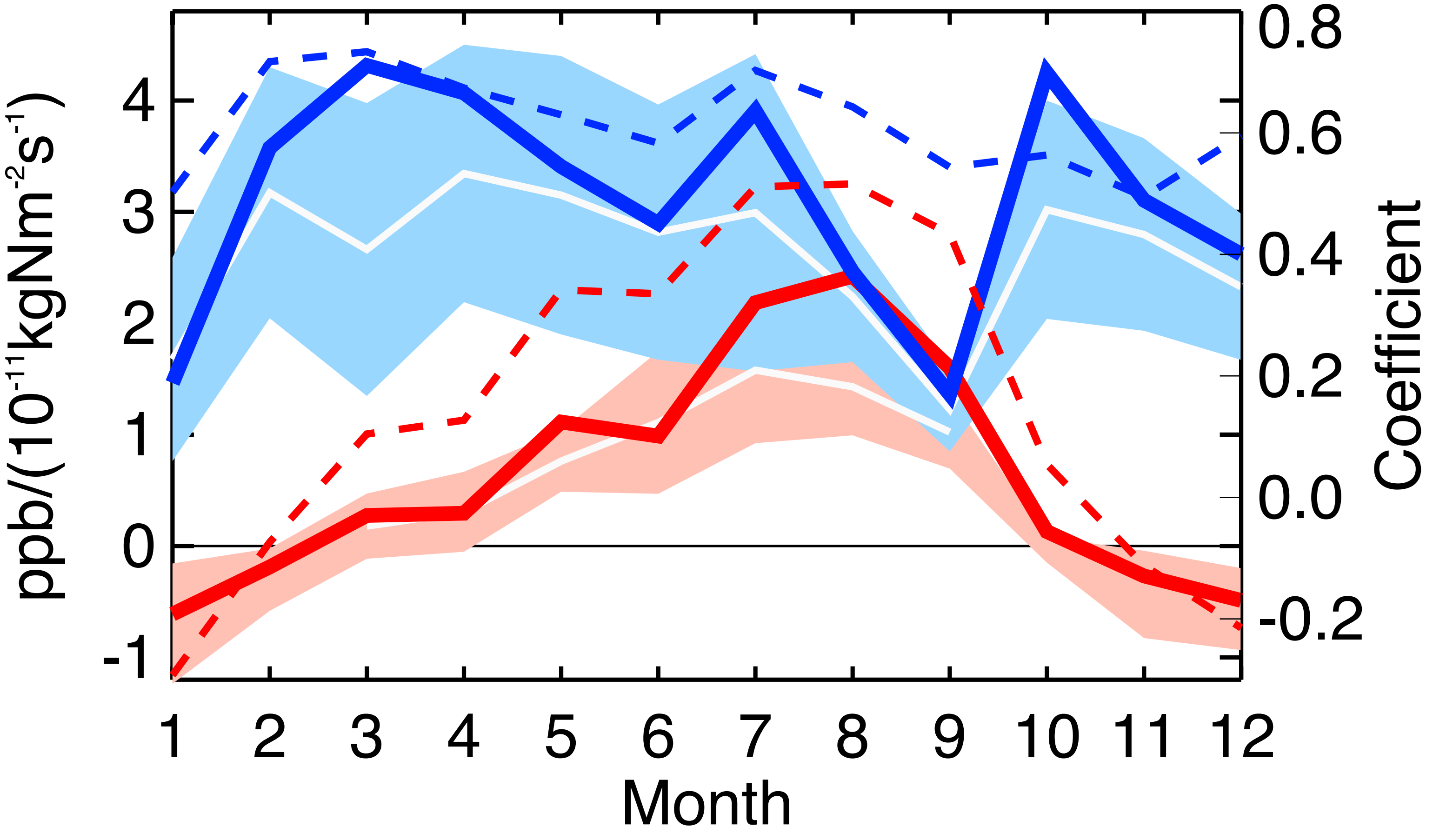
Commonly suggests potential problems in the bottom-up inventories

summertime soil (too low), open BB in over India (missing), wildfire BB biases

Miyazaki et al.,
submitted



NO$_2$ response to NOx emissions



Ozone response to NOx emissions

The ozone/emission analysis increment information can be used as a diagnostic to quantify model sensitivities.

The sensitivity varied by a factor of 2 for end-member models revealing fundamental differences in the fast model processes.

A systematic investigation of model ozone response and analysis increment in MOMO-Chem could benefit evaluation of prediction of chemistry-climate system as a hierarchical emergent constraint (Bowman et al., 2018) and for making effective ozone control strategies.

Miyazaki et al., submitted

Conclusion

- A 14-year chemistry reanalysis has been conducted using multi-constituent multi-sensor satellite DA, in order to provide comprehensive information on atmospheric composition and emissions variability.

- The multi-constituent DA plays an important role in reducing model-observation mismatches and led to up to 70 % differences in the emissions.

Global total 2005-2018 mean: 49.6 TgN (NOx), 7.2 TgN (LNOx), 1096 TgCO (CO), 34.2 TgS (SO_2)

- The multi-model DA provides integrated unique information: e.g., uncertainty ranges in the top-down estimates (4–31% for NOx and 13–35 % for CO).

- Assimilating datasets from a new constellation of LEO sounders and GEO satellites will provide more detailed knowledge of precursors’s emissions, for air quality, human heath, policy and climate applications.