Estimating surface NOx and CO emissions and lightning NOx sources by assimilating satellite observations of multiple chemical species

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We have developed a data assimilation system for the analysis of tropospheric chemical composition and emissions based on an ensemble Kalman filter (EnKF) approach. This system simultaneously optimizes multiple model parameters including the surface emissions of NOx and CO and the lightning sources of NOx together with the concentrations of various chemical species from assimilation of multiple satellite observations (OMI, TES, MOPITT, and MLS). At the workshop, I presented the following key results from the data assimilation:

1. With the multiple satellite datasets, an improved description of the chemical feedbacks can be obtained from the EnKF data assimilation, especially related to the NOx-CO-OH-O3 set of chemical reactions. In the simultaneous data assimilation system, improved atmospheric concentrations of chemically-related species have the potential to improve the emission inversion, while the improved emissions estimates will benefit the atmospheric concentration analysis through a reduction in the model forecast error. For instance, the emission optimization dominated the changes in the O3 profiles in the PBL in the tropics and at northern mid-latitudes, whereas the direct concentration adjustment was much more important in the free troposphere. This reveals the importance of the simultaneous adjustment of the emissions and concentrations for the tropospheric ozone budget and profile analyses (Miyazaki et al., 2012b).

2. The EnKF approach with the state augmentation method approach allows us to accumulate observational information with time and to reflect the non-direct relationship between the emissions and tropospheric columns because of the use of the background error covariance dynamically estimated from the ensemble of CTM forecasts. The assimilation of measurements for species other than NO2 provides additional constraints on the surface NOx emissions by adjusting the concentrations of the species affecting the NOx chemistry. The large influences highlight that uncertainties in the model chemistry impact the quality of the emission estimates. The multiple species assimilation improves the chemical consistency including the relation between concentrations and the estimated emissions (Miyazaki et al., 2012a; Miyazaki and Eskes, 2013).

3. The multiple species data assimilation provides comprehensive constraints on the global lightning NOx sources. This approach has the potential to reduce the influence of model errors on the LNOx source estimation by simultaneously optimizing various aspects of the chemical system, including the surface emissions of NOx and CO as well as the concentrations of 35 chemical species. Errors in these model fields other than the LNOx sources introduce additional model–observation mismatches into the inversion and de-grade the LNOx source estimation. The assimilation provides substantial adjustments to the NOx sources both at the surface and in the middle–upper troposphere, because of the use of multiple satellite data sets with different vertical sensitivities. The analysed LNOx sources

have important implications for improving LNOx parameterisations. For instance, the widely used C-shape assumption underestimates the source strength in the upper troposphere and overestimates the peak source height over land and the tropical oceans, especially along the ITCZ (Miyazaki et al., 2013).

References:

1. Miyazaki, K., Eskes, H. J., and Sudo, K.: Global NOx emission estimates derived from an assimilation of OMI tropospheric NO2 columns, Atmos. Chem. Phys., 12, 2263-2288, doi:10.5194/acp-12-2263-2012, 2012a.

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3. Miyazaki, K., and H. Eskes, Constraints on surface NOx emissions by assimilating satellite observations of multiple species, Geophys. Res. Lett., 40, 4745–4750, doi:10.1002/grl.50894, 2013.

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ECMWF workshop on parameter estimation, 22 October 2013

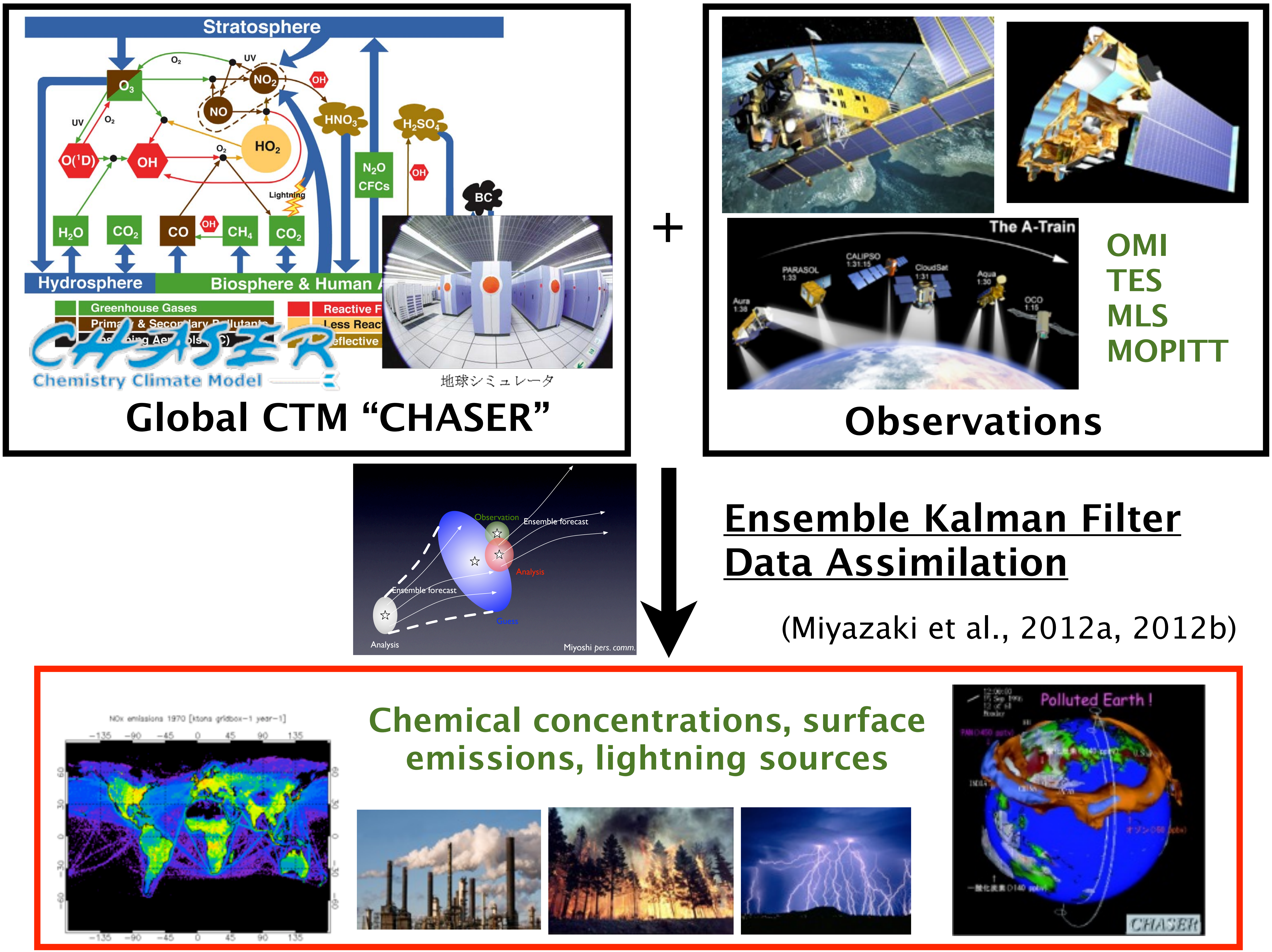
Tropospheric chemistry data assimilation

✓ The use of data assimilation for atmospheric chemistry, especially for short-lived chemical species, is still challenging (e.g., MACC).

✓ A large part of the chemical system is not sensitive to initial conditions, but is sensitive to the model parameters (e.g., reaction rates, emissions).

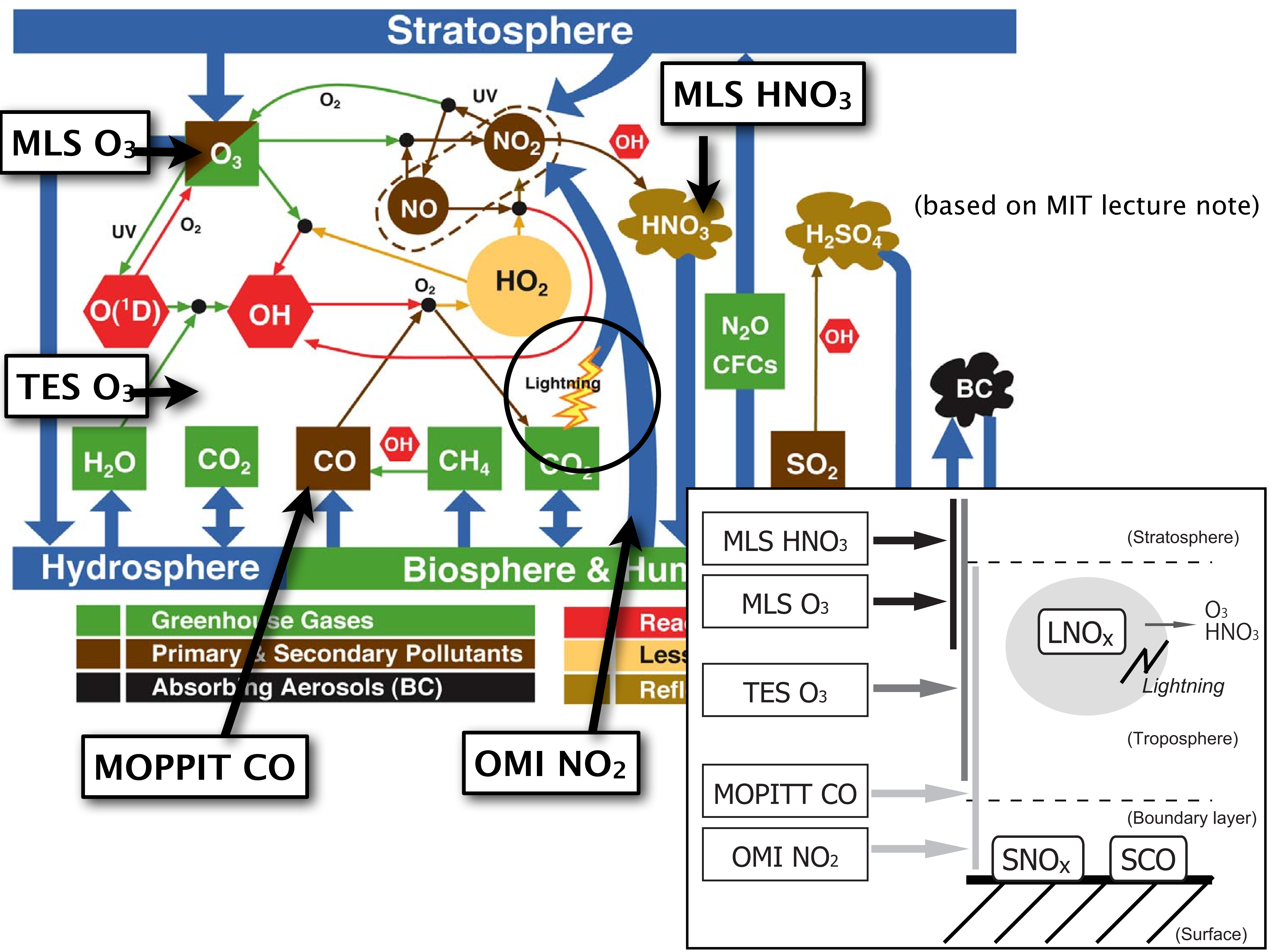
✓→ Simultaneous adjustment of model parameters and concentrations is a powerful framework.

✓ The advantage of Ensemble Kalman filter (EnKF) is its easy implementation for complicated systems and parameter estimations.


Stratosphere
Hydrosphere
Biosphere & Human A
Greenhouse Gases
Reactive F
Less Reac
Lightning
OH
BC
CHASER
Chemistry Climate Model
地球シミュレータ
Global CTM "CHASER"
+
The A-Train
PARASOL
CALIPSO
CloudSat
Aqua
Aura
OCO
OMI
TES
MLS
MOPITT
Observations
Observation
Ensemble forecast
Analysis
Guess
Miyoshi pers. comm.
Ensemble Kalman Filter
Data Assimilation
(Miyazaki et al., 2012a, 2012b)
NOx emissions 1970 [ktons gridbox−1 year−1]
Chemical concentrations, surface emissions, lightning sources
Polluted Earth !
CHASER

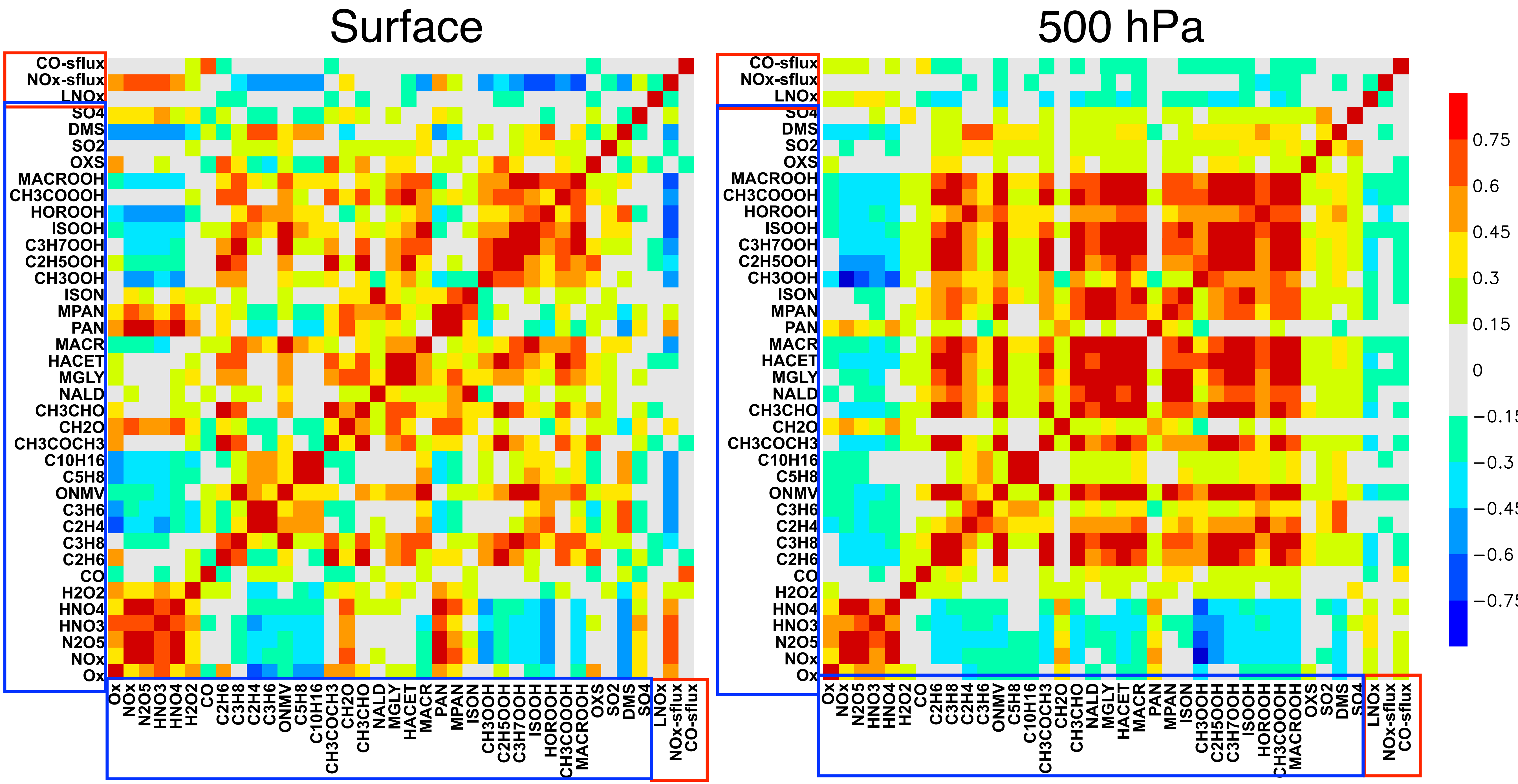
CHASER-DAS (Miyazaki et al., 2012a, 2012b, 2013a, 2013b)

Assimilation scheme	Localized EnKF (LETKF, Hunt et. al., 2007), 48 members
Forecast model	CHASER (Sudo et al., 2002), 47 species & 88 reactions, T42L32
A priori emissions	EDGAR4.2 + GFED3.1 + GEIA
State vector	NOx & CO emissions, lightning NOx, 35 chemical species
Obs operator	Averaging kernel and a priori information
Super Obs	applied for OMI NO_2 and MOPPIT CO data
Cycle	100 min.
Techniques	Spatial & variable covariance localization, covariance inflation
Assimilated data	OMI NO_2 (DOMINO2), TES O_3 (ver. 4), MOPITT CO (ver. 5), MLS O_3 & HNO_3 (ver. 3.3)
Validation data	SCIAMACHY NO_2, GOME-2 NO_2, TES CO, Ozonesonde, Aircraft (INTEX-B, HIPPO) etc



Stratosphere
MLS HNO3
MLS O3
TES O3
(based on MIT lecture note)
O2
UV
NO2
OH
NO
O3
HNO3
H2SO4
O(1D)
OH
HO2
N2O
CFCs
OH
Lightning
BC
H2O
CO2
CO
OH
CH4
CO2
SO2
Hydrosphere
Biosphere & Hun
Greenhouse Gases
Primary & Secondary Pollutants
Absorbing Aerosols (BC)
Rea
Less
Refl
MOPPIT CO
OMI NO2
MLS HNO3
MLS O3
TES O3
MOPITT CO
OMI NO2
(Stratosphere)
LNOx
O3
HNO3
Lightning
(Troposphere)
(Boundary layer)
SNOx
SCO
(Surface)

Background error covariance structure in EnKF



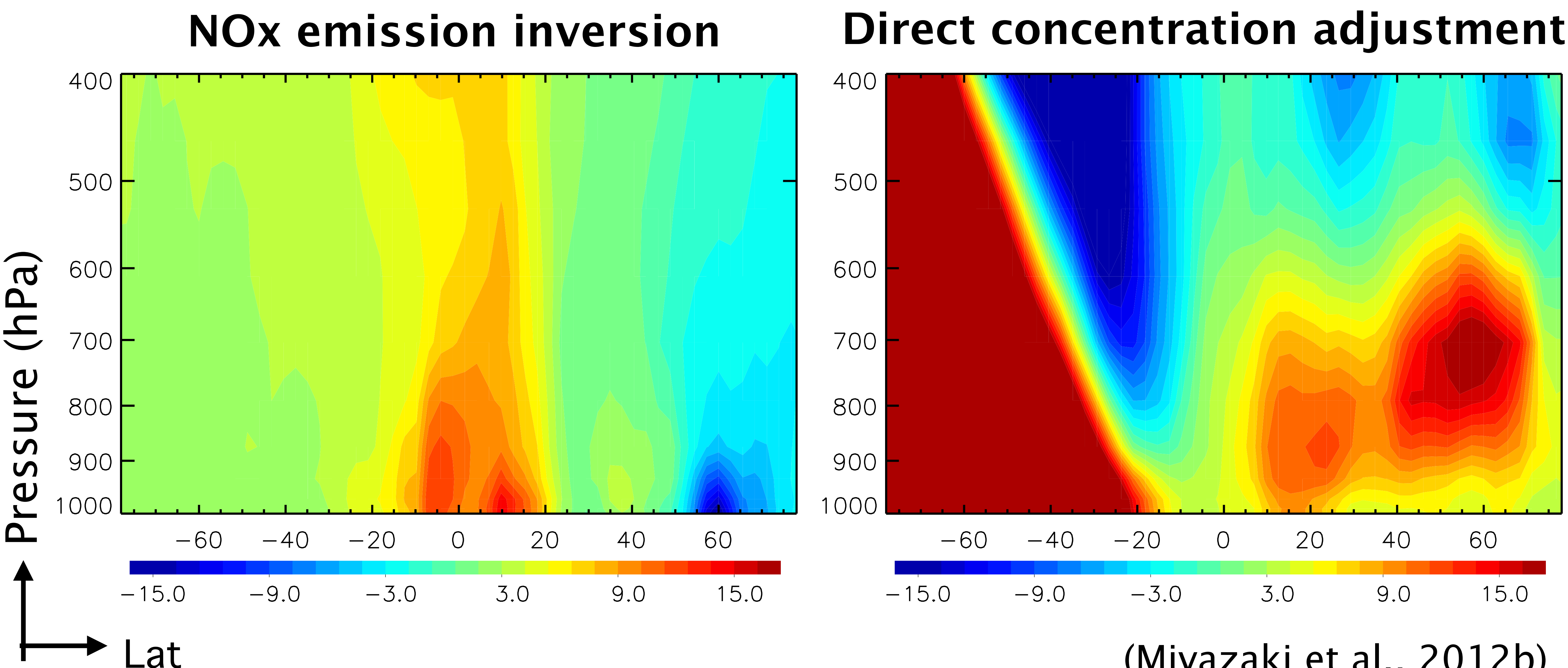
- Emission estimation based on state augmentation.
- Covariance among very weakly-related species is neglected (i.e., variable localization (Kang et al., 2011)).

$$\mathbf{x}_i^b = \begin{bmatrix} \mathbf{c}_i^b \\ \mathbf{e}(\mathrm{NO_x})_i^b \\ \mathbf{e}(\mathrm{CO})_i^b \\ \mathbf{e}(\mathrm{LNO_x})_i^b \end{bmatrix}$$

Concentration

Emissions

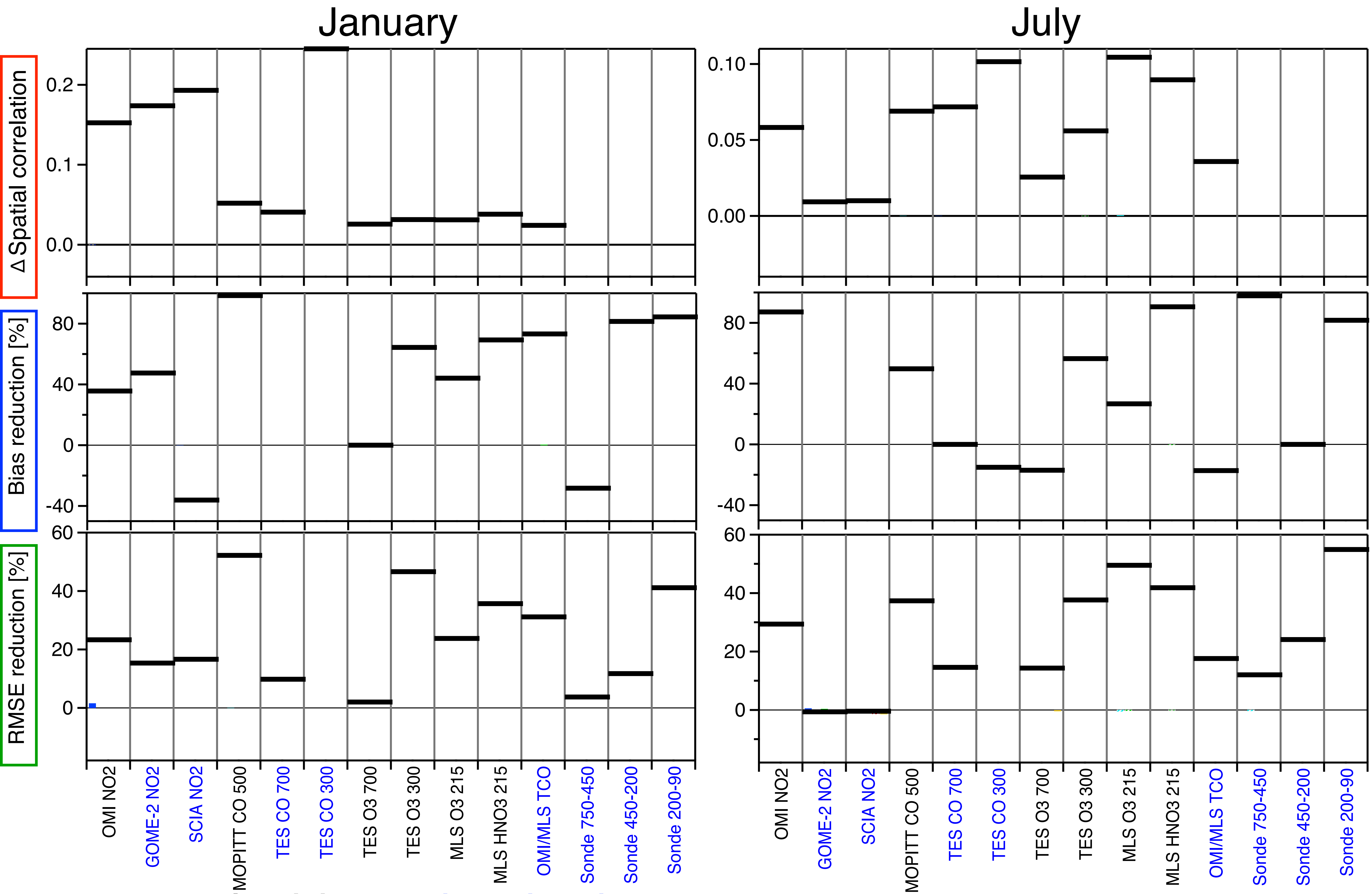
The relative impact (in %) of the NOx emission inversion (left) and the direct concentration adjustment (right) through assimilation on the vertical O_3 profile



(Miyazaki et al., 2012b)

The simultaneous adjustment of the emissions and the concentrations is a powerful approach to optimize the whole tropospheric profiles

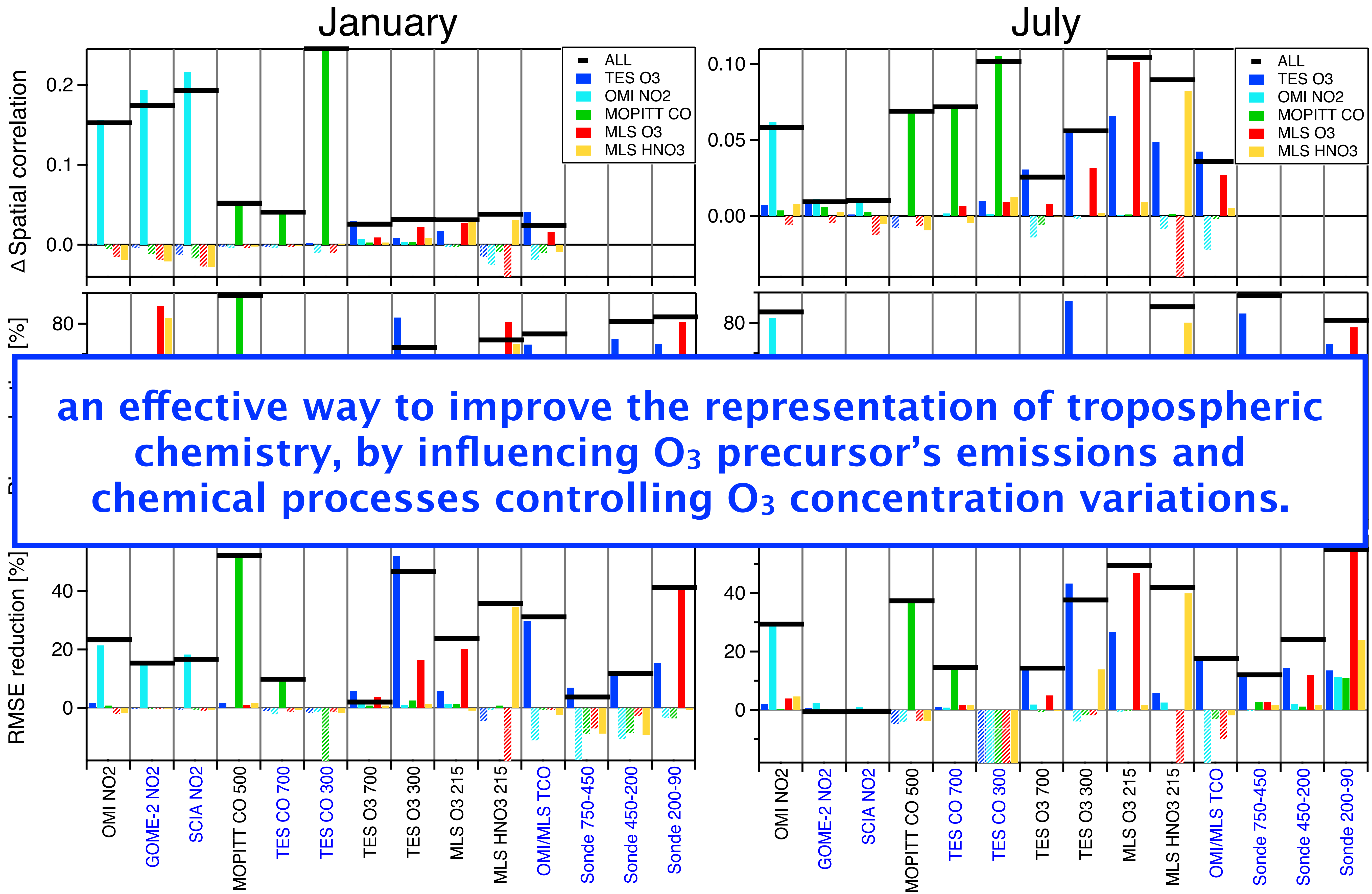
Spatial correlation increment BIAS reduction rate RMSE reduction rate



Assimilated data **Independent data**

(Miyazaki et al., 2012b)

Observing System Experiments (OSEs)



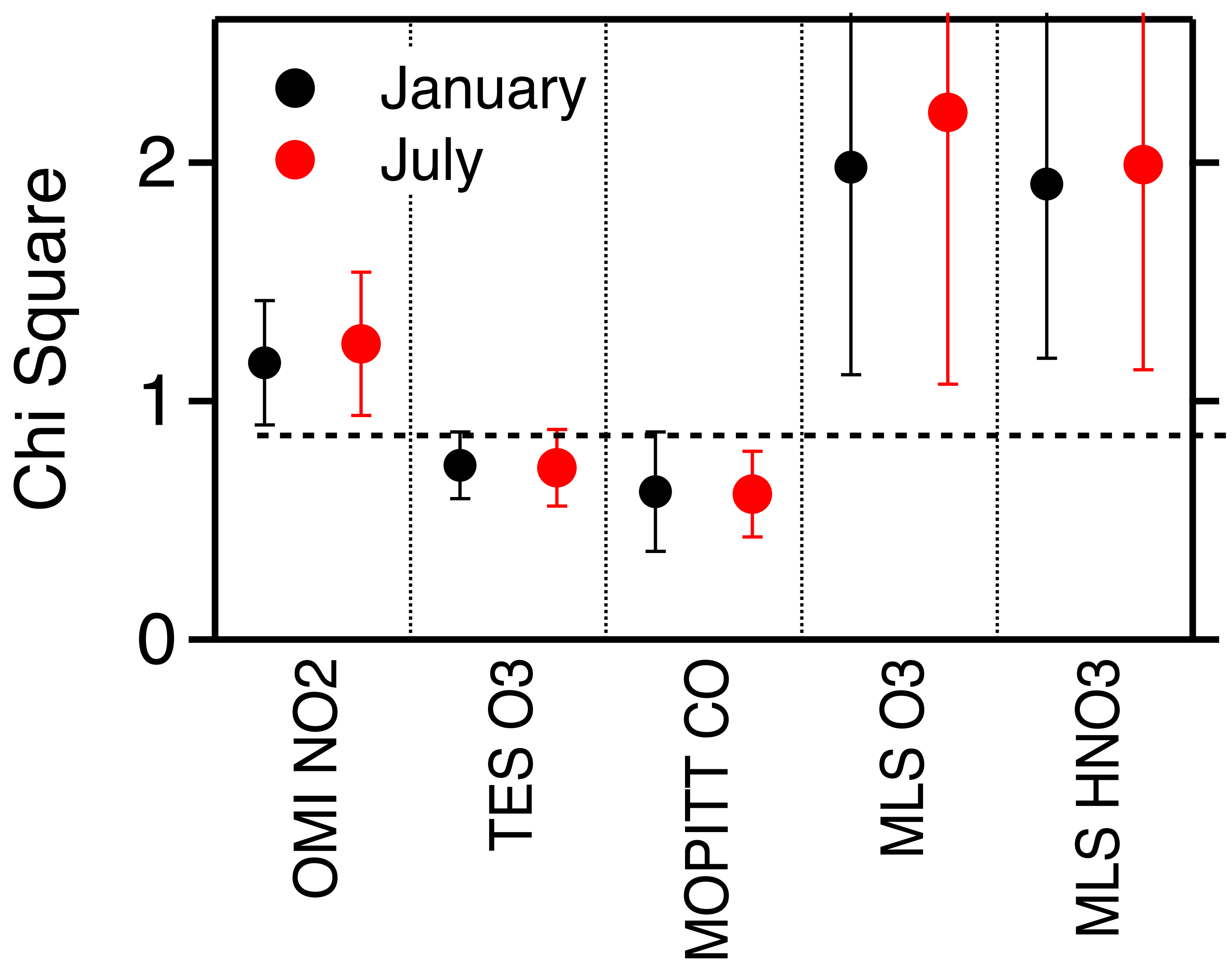
an effective way to improve the representation of tropospheric chemistry, by influencing O_3 precursor's emissions and chemical processes controlling O_3 concentration variations.

(Miyazaki et al., 2012b)

Self-consistency check: Chi-square test

An important test for the quality of data assimilation is whether the differences between the innovations are consistent with the covariance matrices for the model forecast and observations.

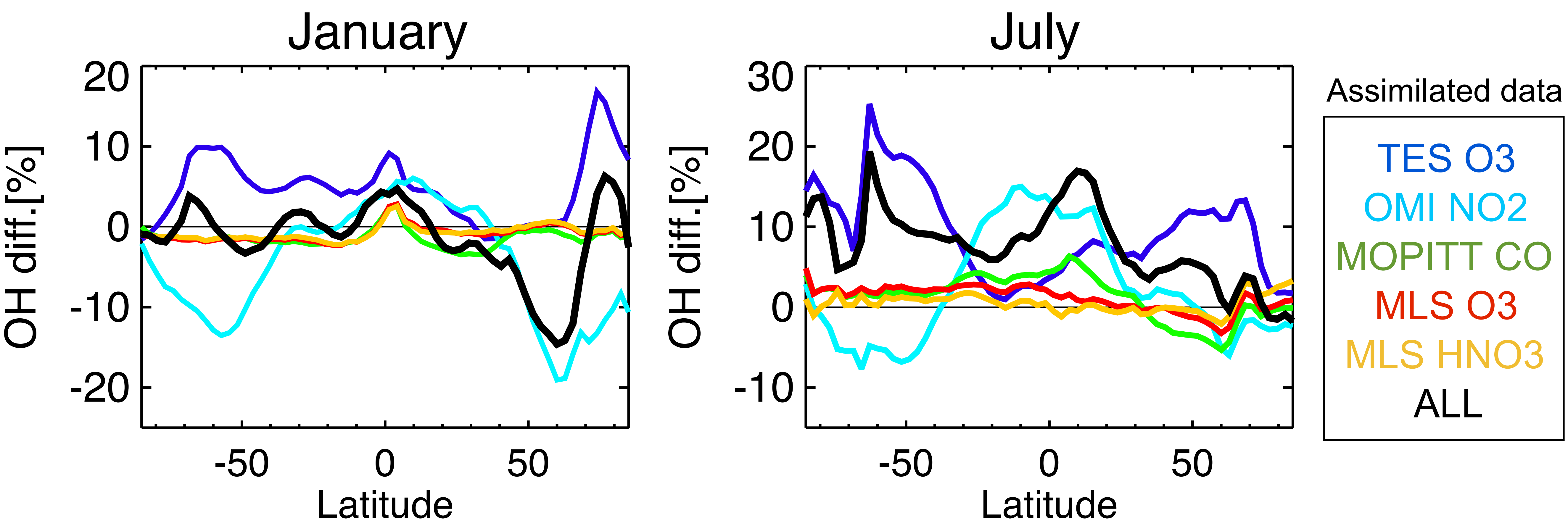
$$\mathbf{Y} = \frac{1}{\sqrt{m}} \left(\mathbf{H}\mathbf{P}^b\mathbf{H}^T + \mathbf{R}\right)^{-1/2} \left(\mathbf{y}^o - H\left(\mathbf{x}^b\right)\right). \qquad \chi^2 = \mathrm{trace}\,\mathbf{Y}\mathbf{Y}^T$$



(Miyazaki et al., 2012b)

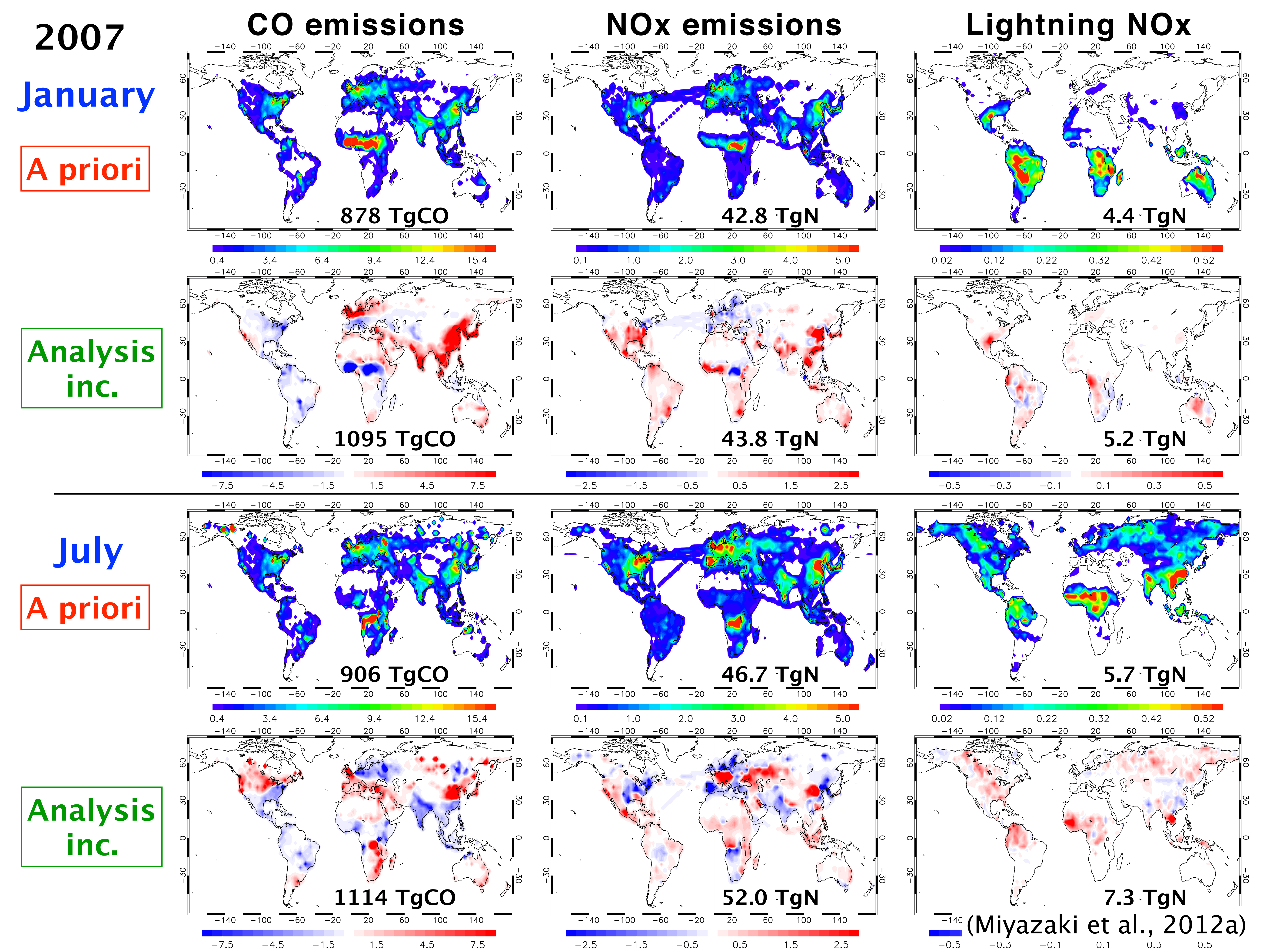
Influences on the oxidation capacity

- The OSEs confirm that the assimilation of each species data set has a strong influence on both assimilated and non-assimilated species.
- The inter-species influences are tightly associated with the changes in OH because of the chemical interactions in the CO-OH-Ox-NOx system.

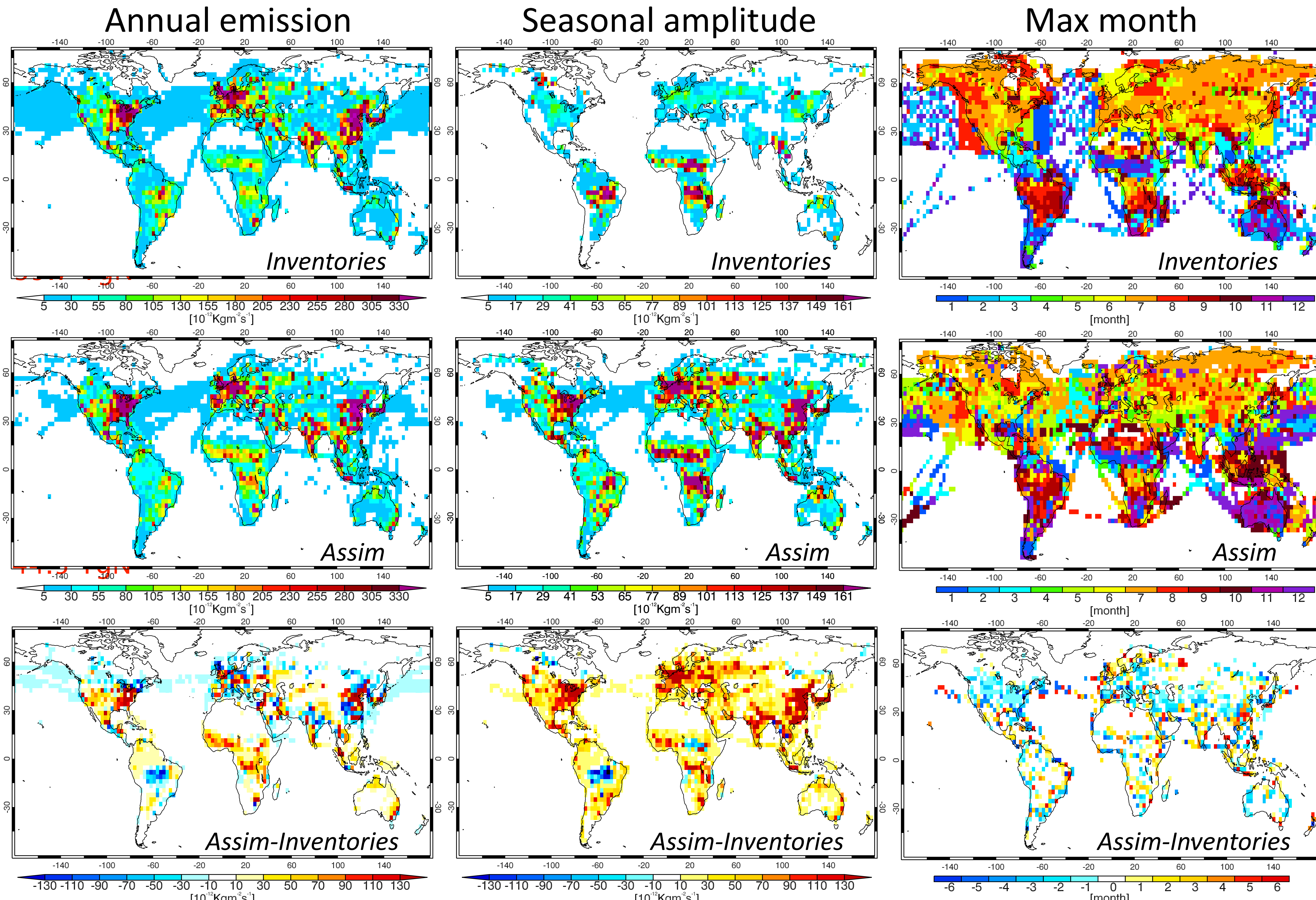


The obvious changes in the OH fields reveal the great potential of the multiple species assimilation to influence the NOx emission inversion etc.

(Miyazaki et al., 2012b)


2007
CO emissions
NOx emissions
Lightning NOx
January
A priori
878 TgCO
42.8 TgN
4.4 TgN
0.4 3.4 6.4 9.4 12.4 15.4
0.1 1.0 2.0 3.0 4.0 5.0
0.02 0.12 0.22 0.32 0.42 0.52
Analysis inc.
1095 TgCO
43.8 TgN
5.2 TgN
−7.5 −4.5 −1.5 1.5 4.5 7.5
−2.5 −1.5 −0.5 0.5 1.5 2.5
−0.5 −0.3 −0.1 0.1 0.3 0.5
July
A priori
906 TgCO
46.7 TgN
5.7 TgN
0.4 3.4 6.4 9.4 12.4 15.4
0.1 1.0 2.0 3.0 4.0 5.0
0.02 0.12 0.22 0.32 0.42 0.52
Analysis inc.
1114 TgCO
52.0 TgN
7.3 TgN
−7.5 −4.5 −1.5 1.5 4.5 7.5
−2.5 −1.5 −0.5 0.5 1.5 2.5
(Miyazaki et al., 2012a)

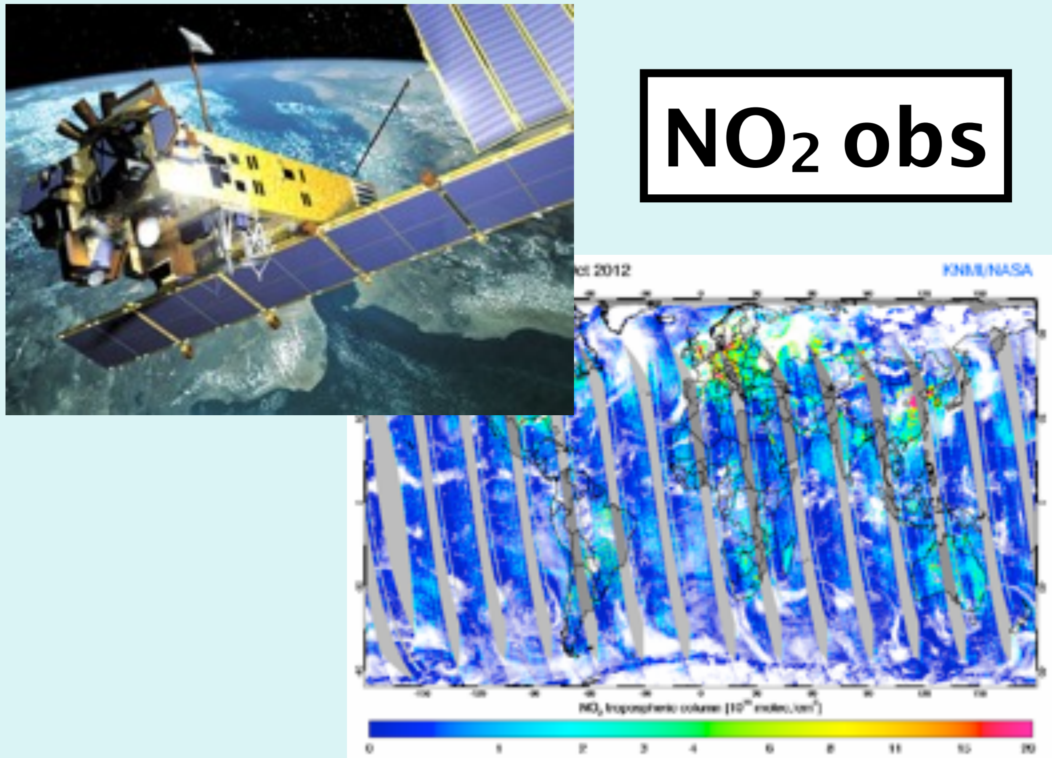
Surface NOx emissions in 2007



(Miyazaki and Eskes, 2013)

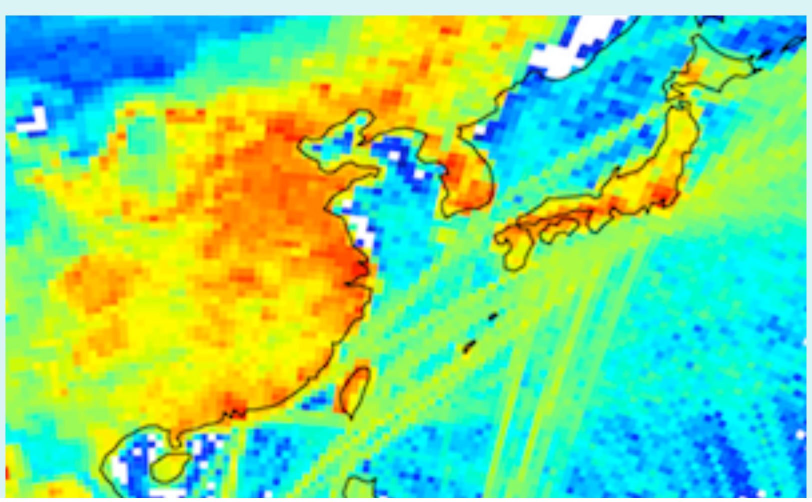
Top-down NOx emission estimates from satellite observations

Previous studies (only NO_2 obs used)



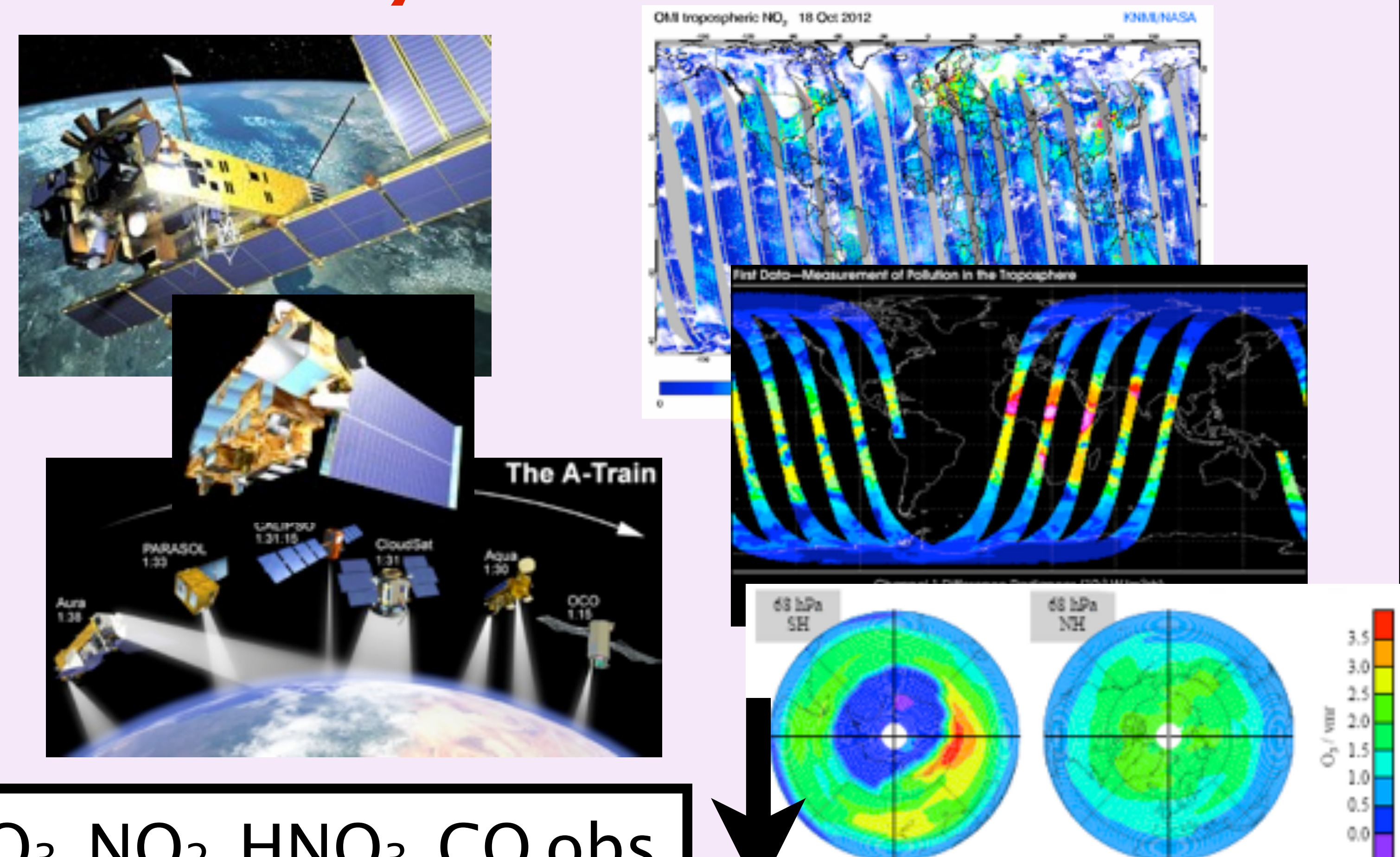
NO_2 obs

Obvious influences of model errors



(e.g., Martin et al., 2003; Jaeglé et al., 2005)

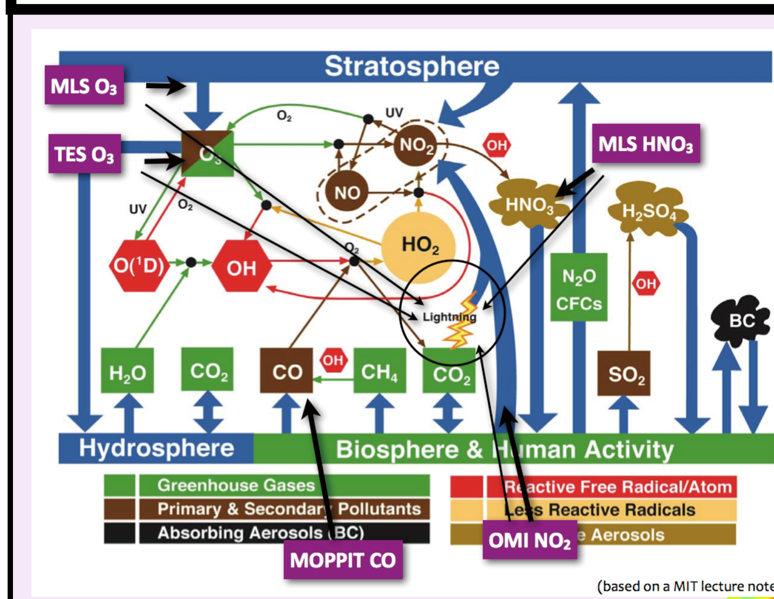
This study (uses chemically-related species obs)

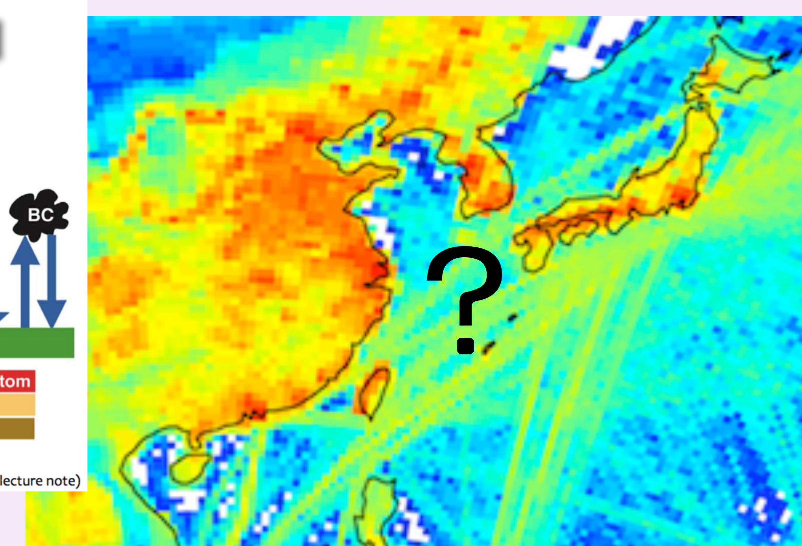


O_3, NO_2, HNO_3, CO obs

Constrain the chemical system

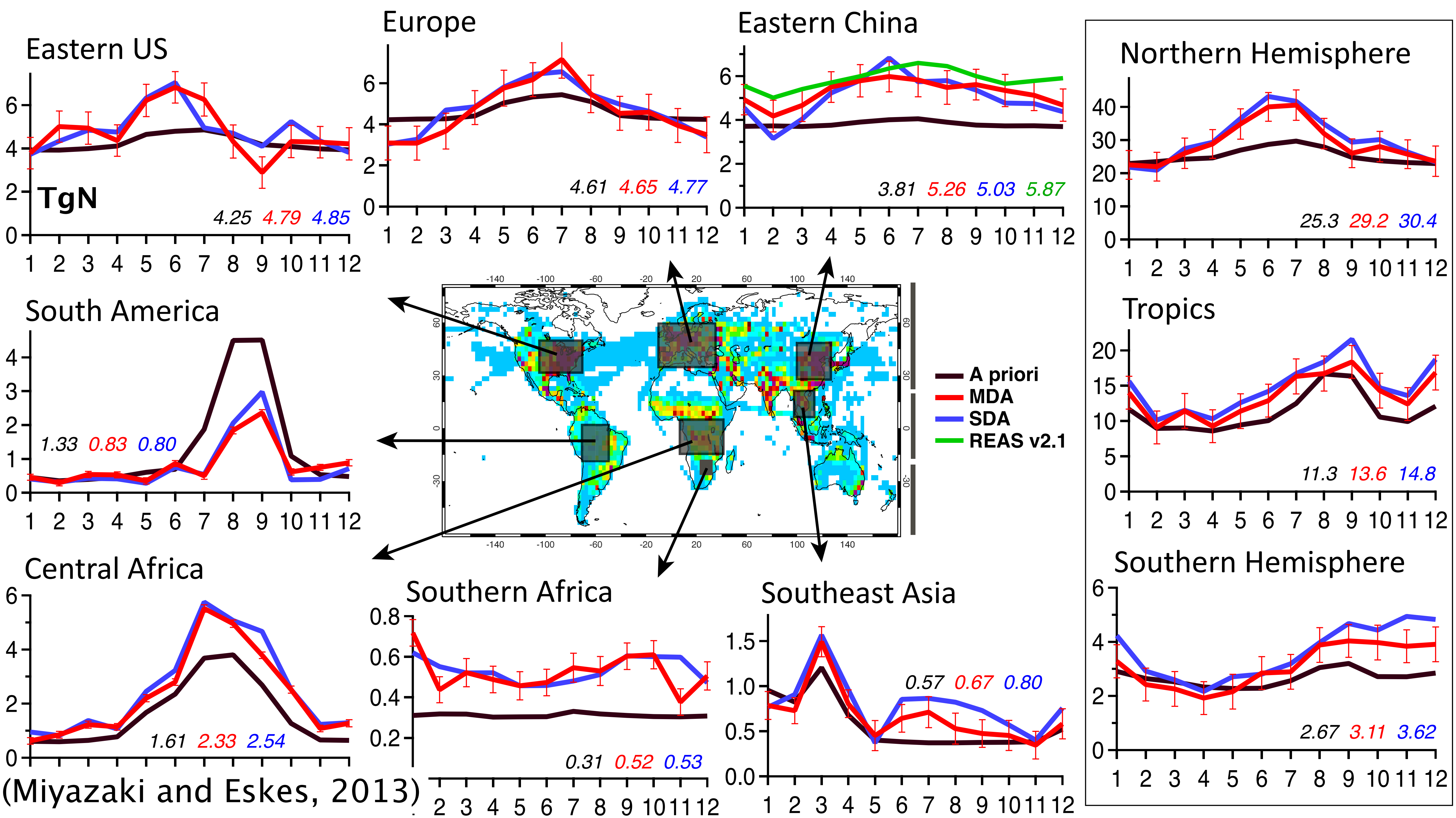
Reduce model errors and improve the emission analysis



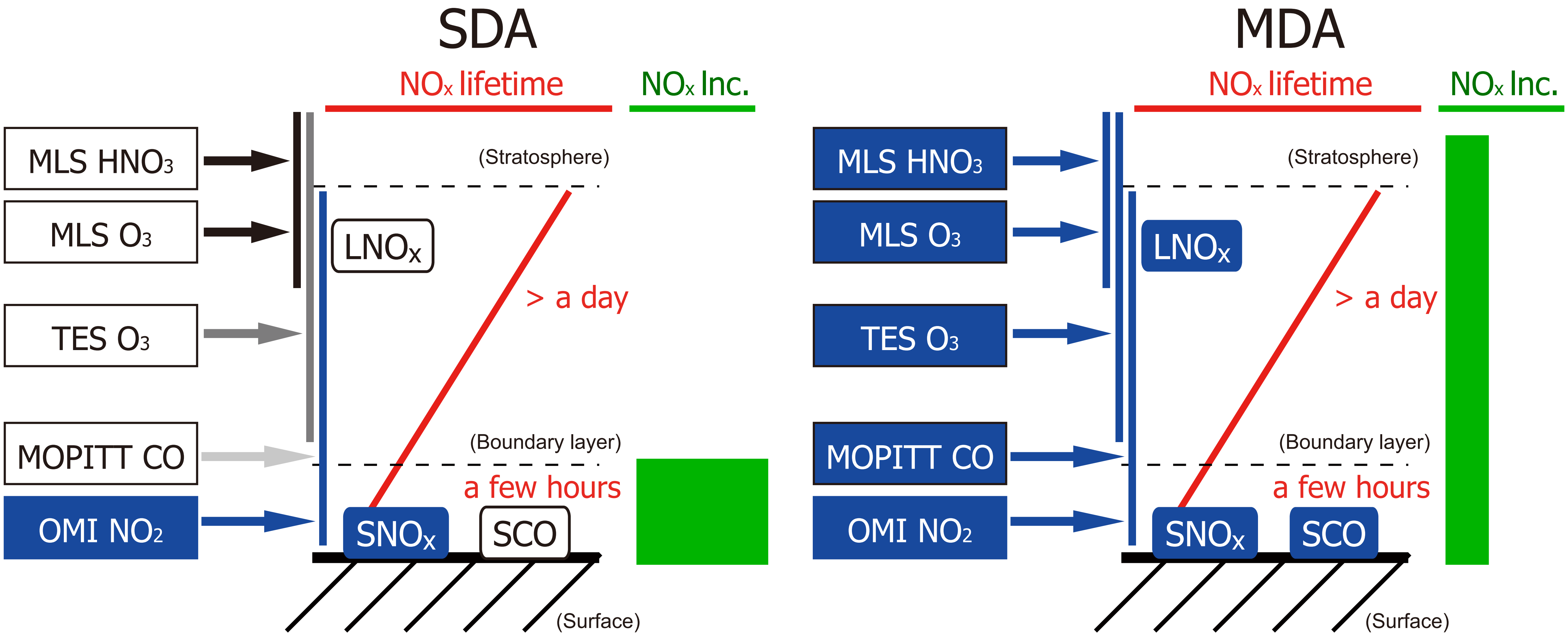


Multiple species constraints on surface NOx emissions

- The **multiple datasets assimilation (MDA)** provides additional constraints, as a consequence of the NO_2 profiles being modified by the non-NO_2 observations.
- The large influences of non-NO_2 data highlight the large uncertainty (by 58% on regional scale) in the NOx emissions inverted from **NO_2 observations only (SDA: single dataset assimilation)**.



(Miyazaki and Eskes, 2013)

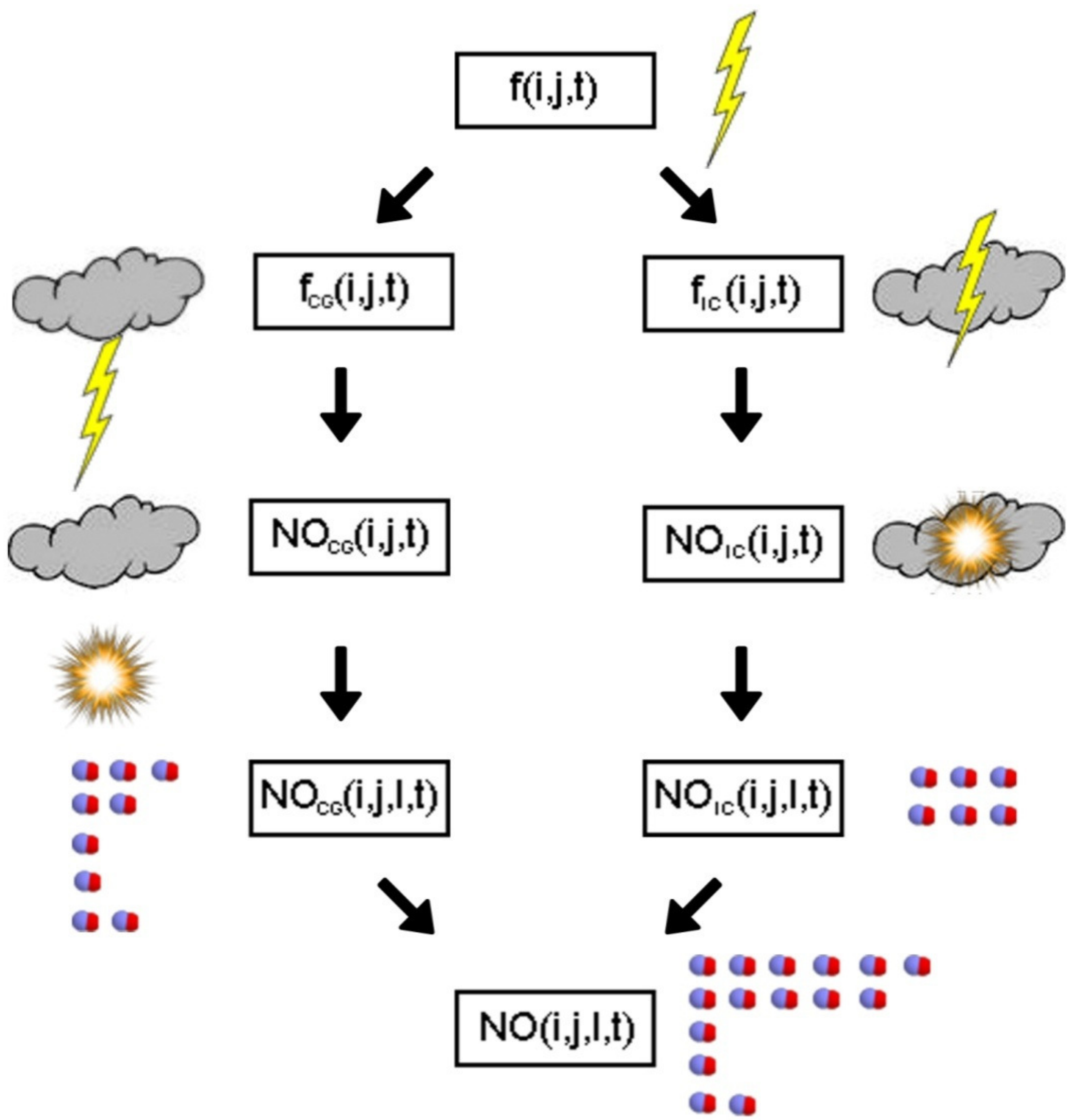


SDA
NOx lifetime
NOx Inc.
MLS HNO3
MLS O3
TES O3
MOPITT CO
OMI NO2
(Stratosphere)
LNOx
> a day
(Boundary layer)
a few hours
SNOx
SCO
(Surface)
MDA
NOx lifetime
NOx Inc.
MLS HNO3
MLS O3
TES O3
MOPITT CO
OMI NO2
(Stratosphere)
LNOx
> a day
(Boundary layer)
a few hours
SNOx
SCO
(Surface)

Accurate estimates of LNOx are important to understand variations in NOx, the oxidizing capacity, and several greenhouse gases (O_3, CH_4).

Larger uncertainly in the estimated total amount of NOx globally produced by lightning, i.e. ranging from 2 to 8 TgN/yr.

Bottom-up: The lightning and subsequent NOx formation are determined with the help of empirical parameterizations.



Spatial (2-D) and temporal distribution of lightning flashes

(Price and RInd, 1992)

Ratio of IC to CG flashes

NO production per IC & CG flash

Vertical NO distribution for IC & CG flashes

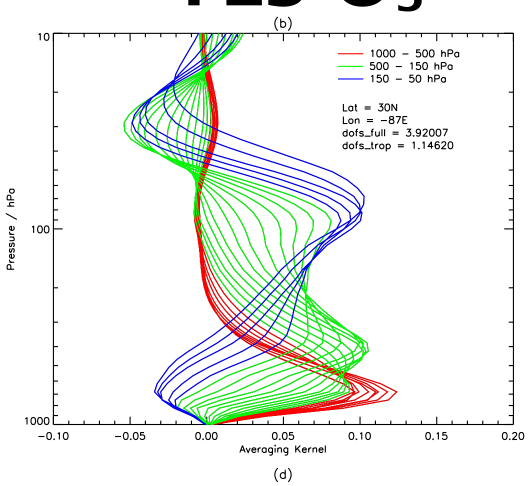
Total lightning NO profile

(From Folkert Boersma)

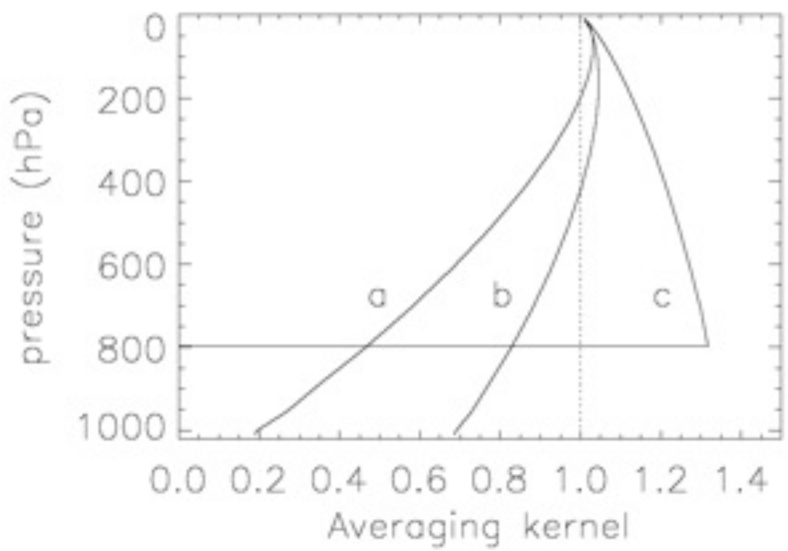
Top-down approach: Satellite data assimilation

Top-down approach uses satellite retrievals of chemical species to obtain optimal value of lightning NOx source in CTM simulation.

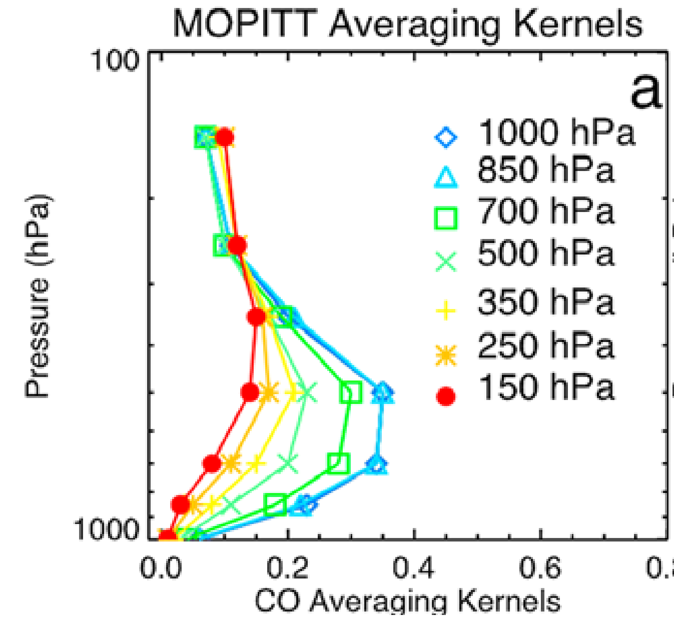
TES O_3



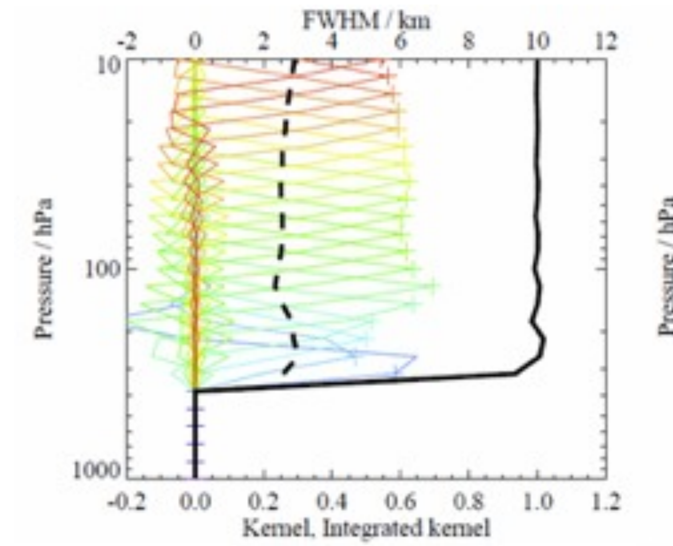
OMI NO_2



MOPITT CO



MLS O_3, HNO_3



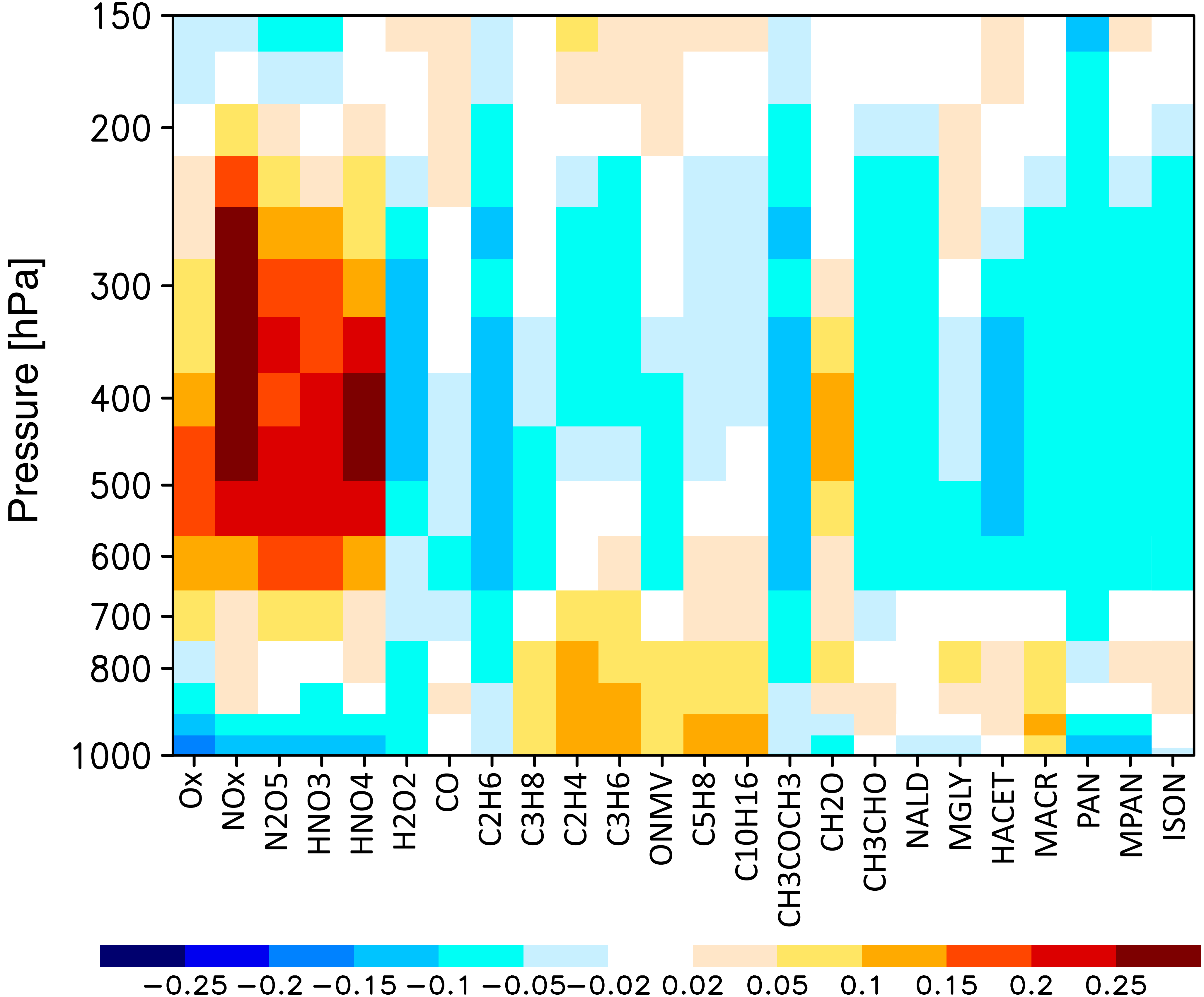
TES: DoF>1 for the middle/upper troposphere. Provides observations of ozone-enhanced layers downwind of convective events (valuable for estimating the LNOx profiles.)

OMI: The overpass time (13:30) is more suitable for LNOx estimation than the morning time observation (GOME-2, and SCIAMACHY). For the cloud-covered observations the AK shows a sharp drop roughly halfway the cloud, and very small sensitivities below.

MOPITT: (indirectly) affects the LNOx source estimation through their influence on the oxidation capacity and the NOx chemistry.

MLS: have a great potential to constrain the LNOx sources in the upper troposphere (i.e., the long lifetimes of NOx, HNO_3, and O_3 in the upper troposphere).

Background error covariance with LNOx



(Miyazaki et al., 2013)

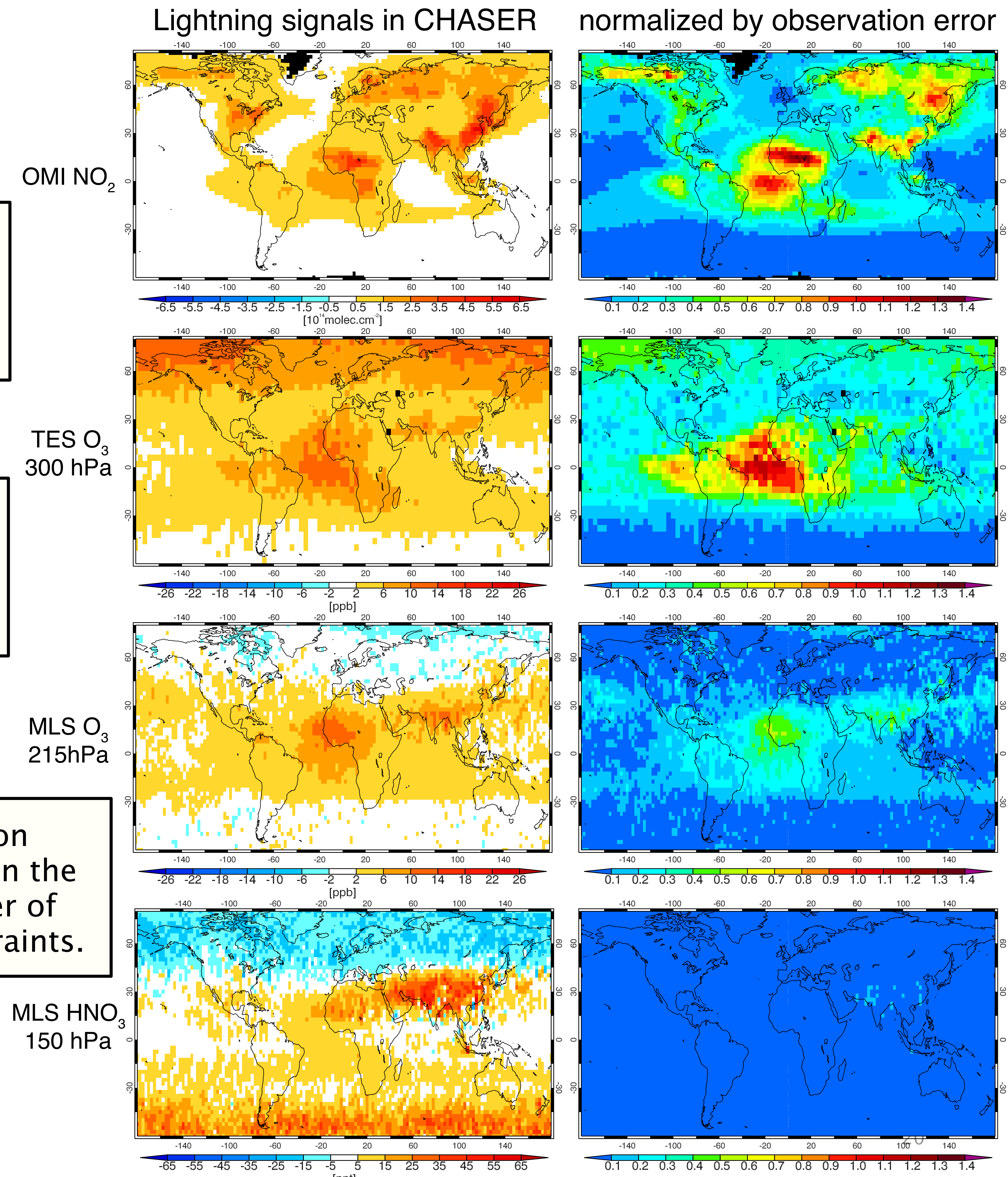
Lightning signal v.s. observation error

OMI NO_2: The lightning signals are large compared to the local super-observation error over the tropical Atlantic etc.

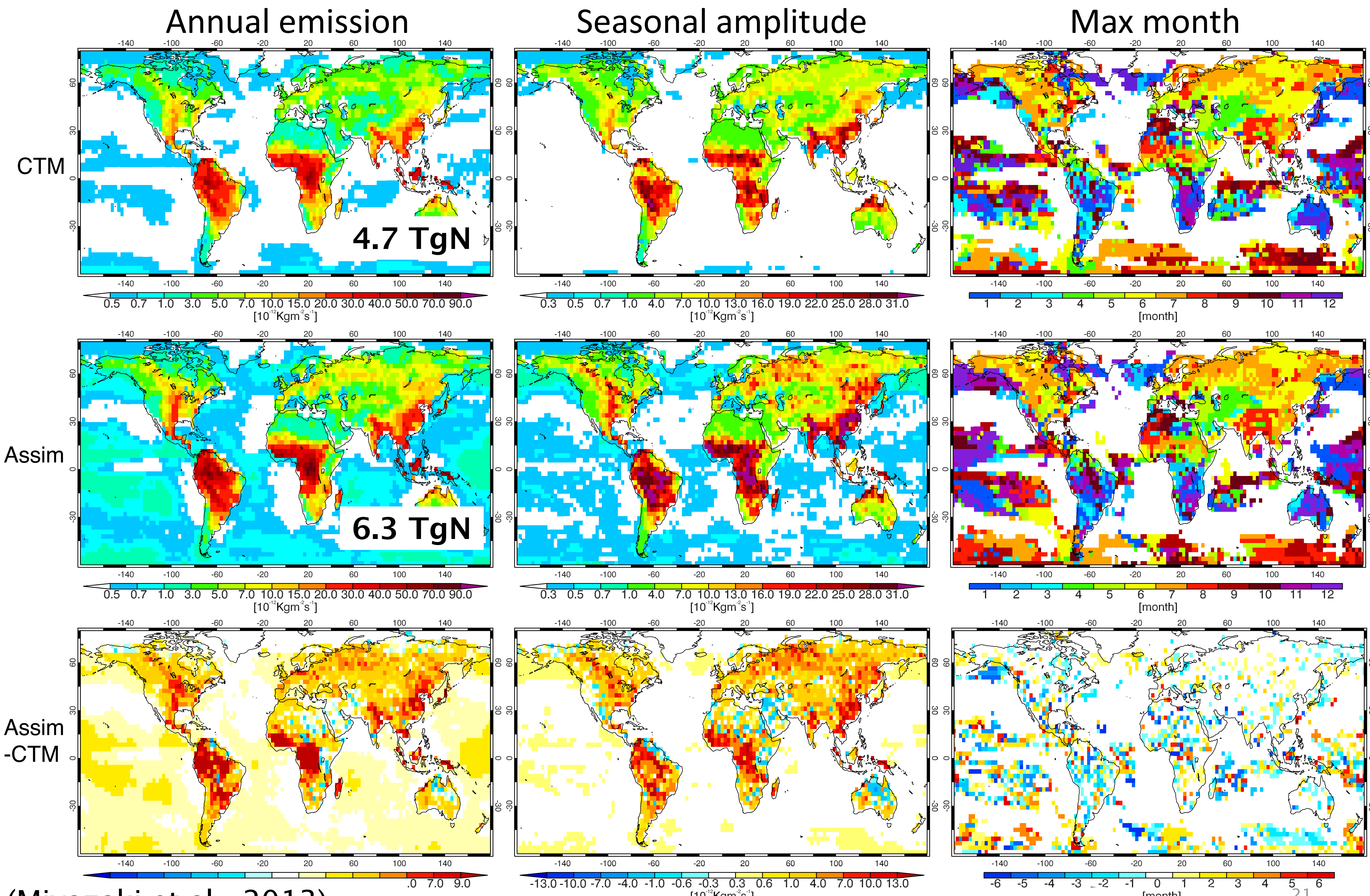
TES O_3: The large signals in the tropical upper troposphere (esp. over the Atlantic) are nearly equal to the mean observation error.

MLS O_3, HNO_3: The mean observation errors are generally much larger than the lightning signals, but a large number of observations can still provide constraints.

June-August 2007



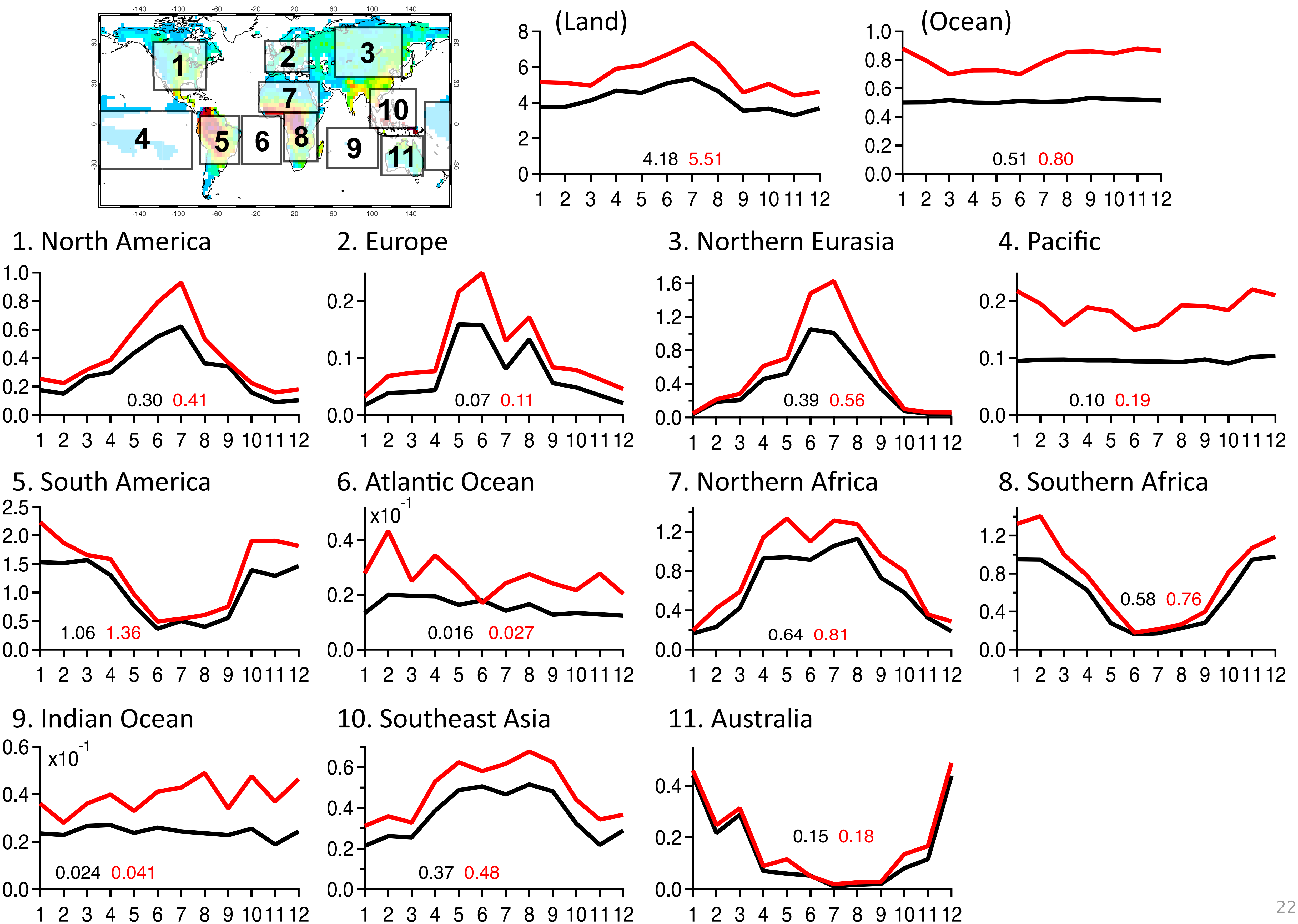
Lightning NOx sources in 2007



(Miyazaki et al., 2013)

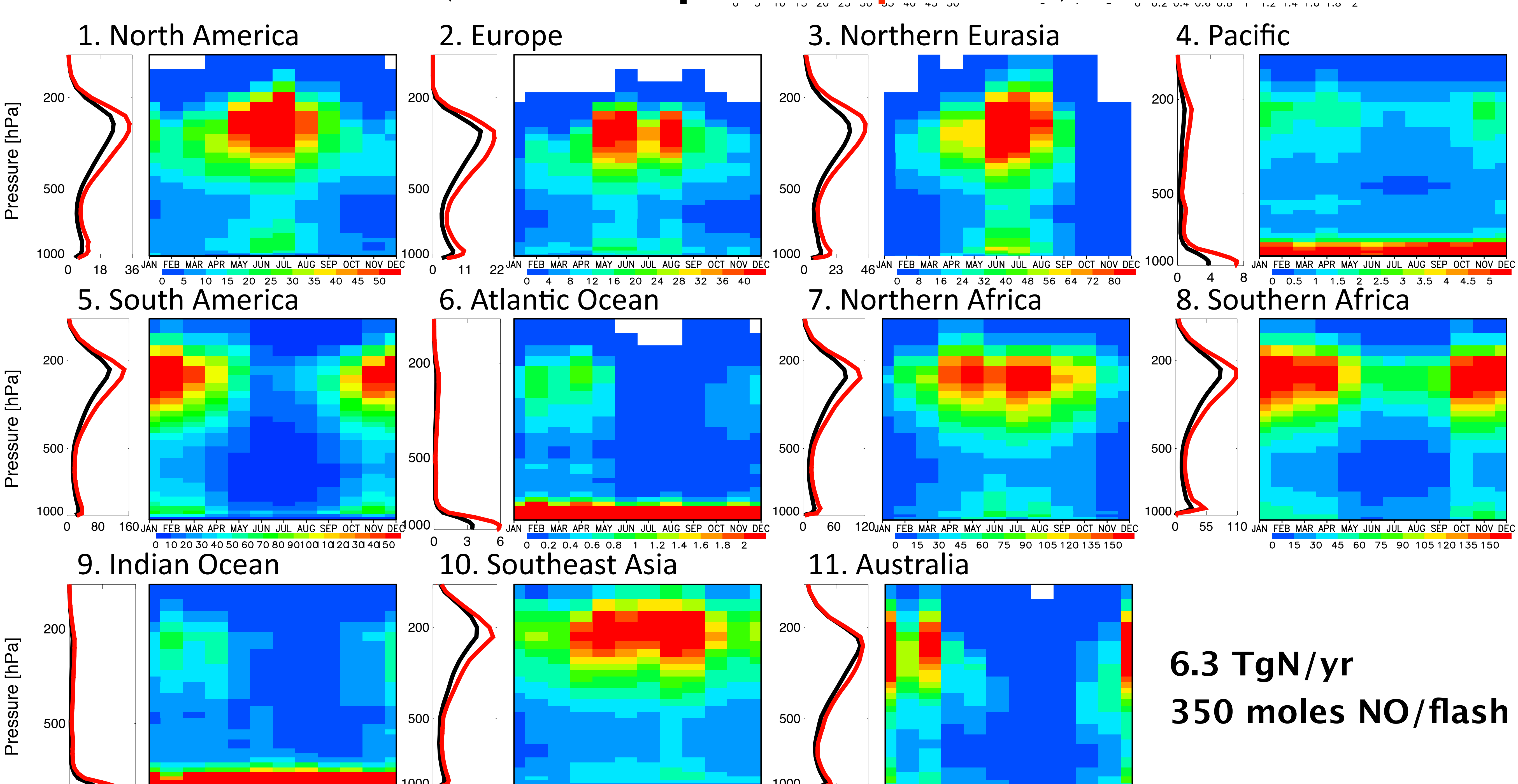
Seasonal variation of the LNOx sources

(**Bottom-up** and **Top-down**)



Seasonal variation of the vertical LNOx profiles

(**Bottom-up** and **Top-down**)



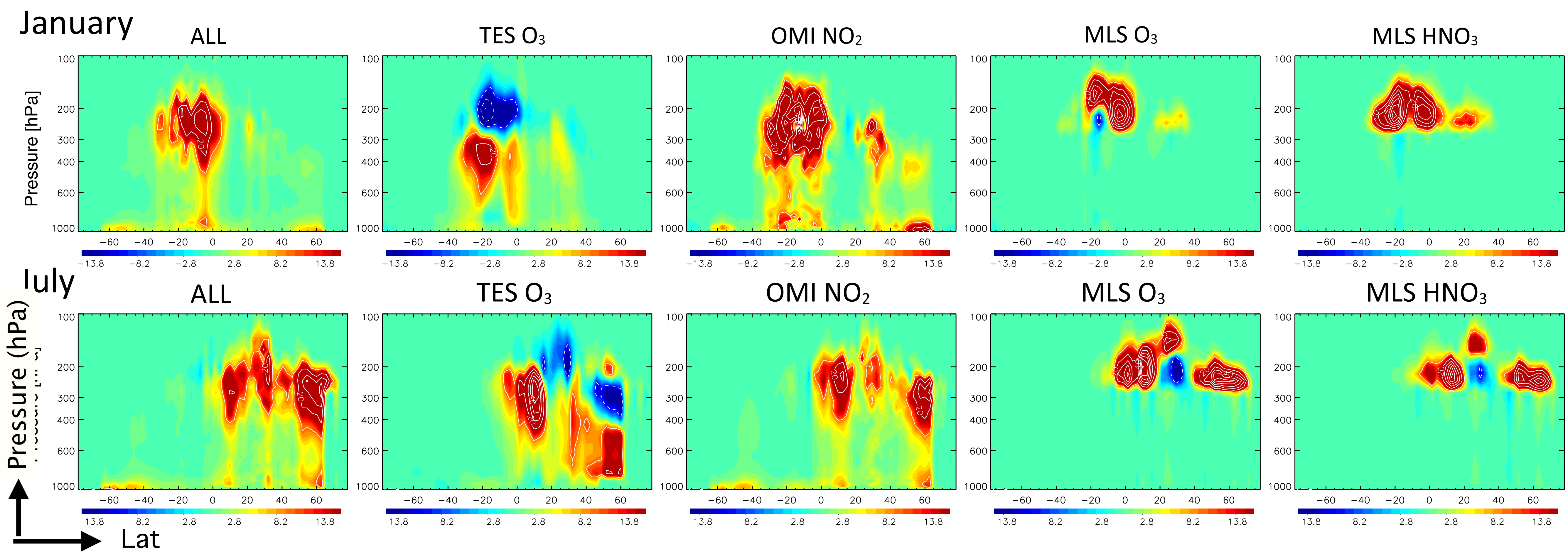
6.3 TgN/yr

350 moles NO/flash

The widely used lightning parameterisation based on the C-shape assumption underestimates the source amounts in the upper troposphere and overestimates the peak source height in the upper troposphere by up to 1 km over land.

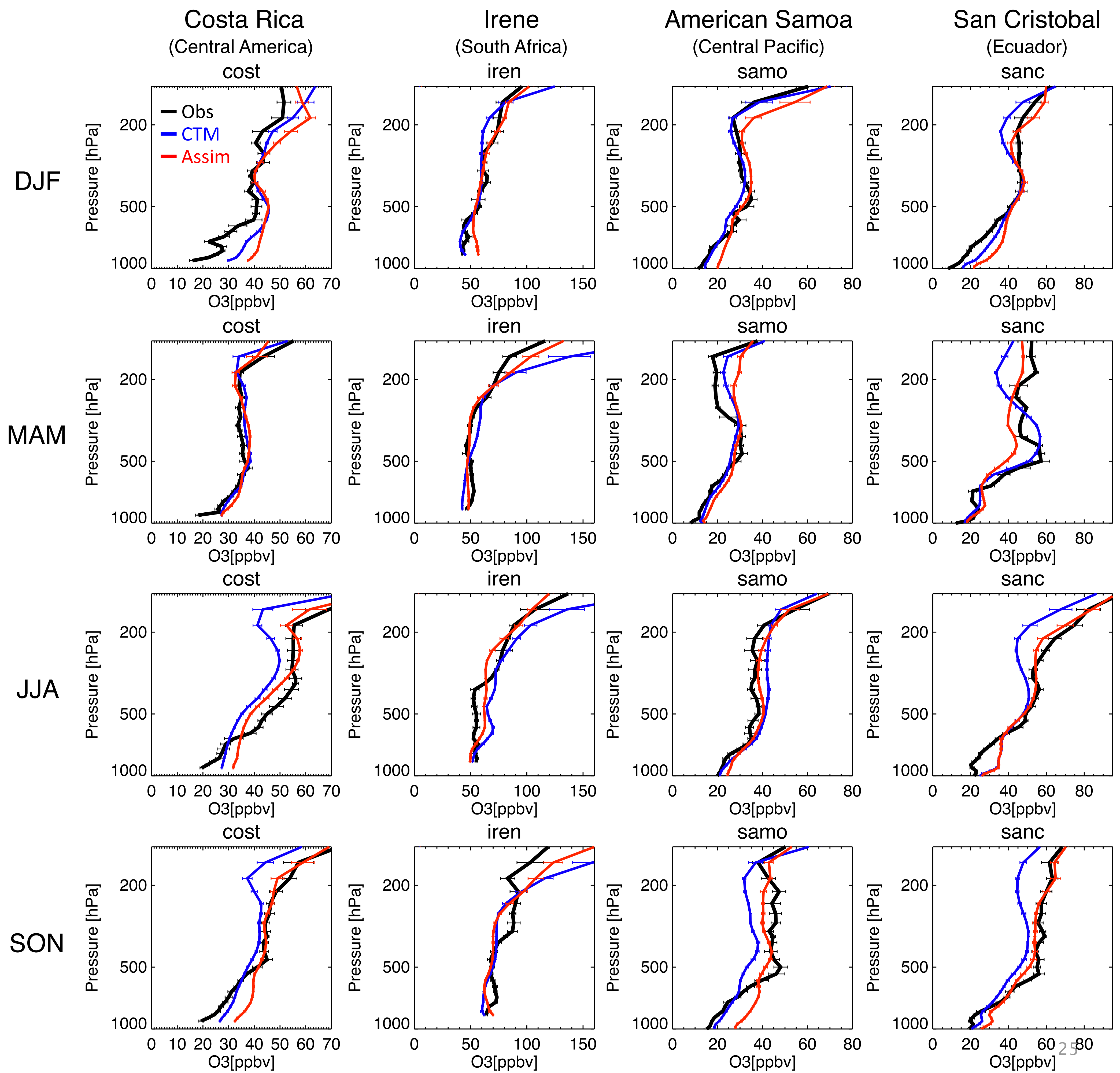
LNOx source increments from OSEs

90S–90N & 1000–100hPa cross-section



The combined use of the multiple datasets with different vertical sensitivities etc facilitates the estimation of the vertical LNOx profile and to distinguish between the surface NOx emissions and LNOx sources.

v.s. SHADOZ ozonesondes



Error estimation

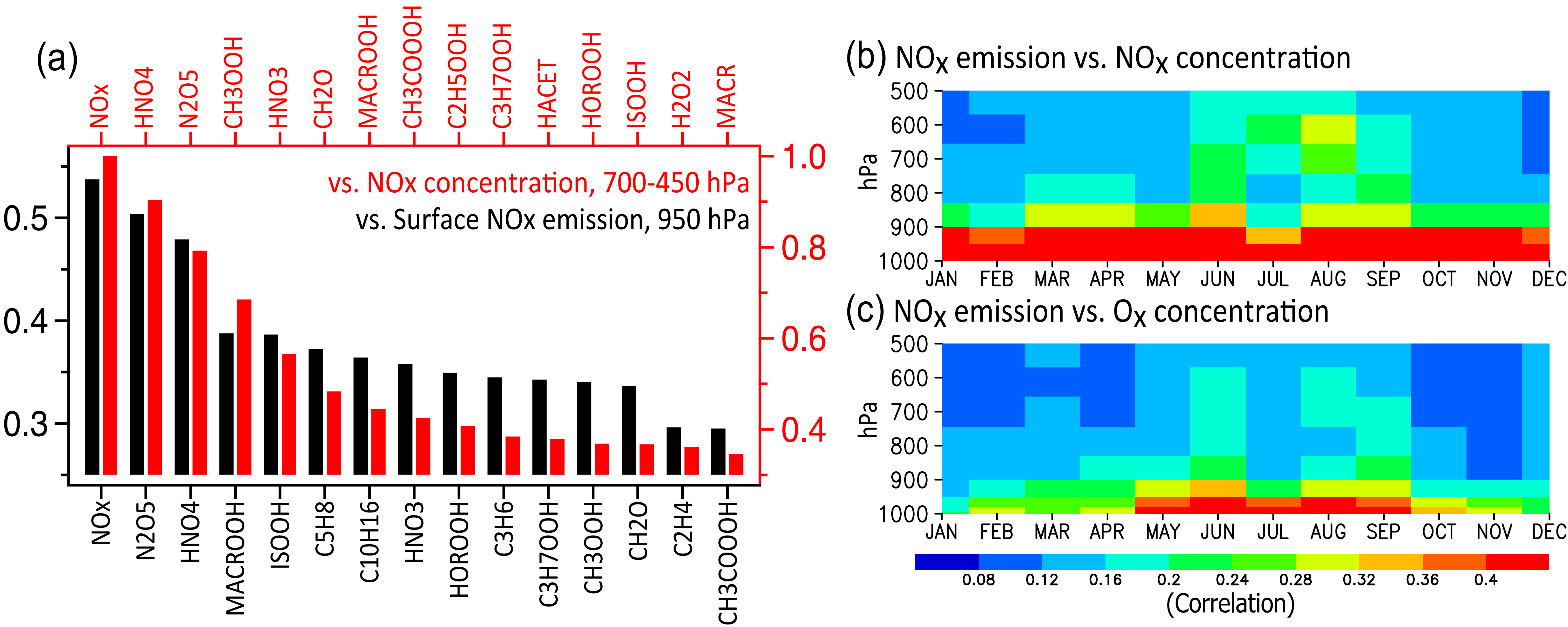
	January				July			
	NH	TR	SH	GL	NH	TR	SH	GL
Control	0.78	3.99	1.39	6.15	4.69	2.99	0.50	8.18
w/ OMI bias	0.87	3.97	1.46	6.31	4.61	3.08	0.50	8.18
TES bias corr.	0.68	3.79	1.36	5.83	4.19	2.74	0.29	7.21
w/o cloud OMI	0.76	4.04	1.31	6.09	4.13	2.89	0.29	7.33
year 1997 SST	0.76	3.89	1.37	6.03	4.71	3.06	0.51	8.26
+20% convection	0.80	3.76	1.37	5.89	4.27	2.99	0.50	8.09
+20% LNOx err.	0.83	3.75	1.32	5.90	4.59	2.93	0.51	8.03
+20% SNOx err.	0.81	3.77	1.27	5.85	4.58	2.83	0.50	7.90
+15% LNOx prior	0.83	4.10	1.48	6.41	5.29	3.16	0.57	9.02
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Total bias	0.16	0.47	0.20	0.66	1.06	0.38	0.31	1.58

(and more error sources in the chemical schemes etc (e.g., Stavrakou et al. 2013)

c.f. Schumann and Huntrieser (2007) have provided a best estimate of 5±3 TgN

Further developments in measurements and data assimilation will be important to reduce the uncertainty in the LNOx source estimation.

For further improvements in the emission estimates



OSSEs with a careful consideration of the complex chemical interactions and measurement characteristics for various species (incl. the seasonality) will support future instrumental design to improve the emission analysis.

(Miyazaki and Eskes, 2013)

Summary

- In the simultaneous DA framework, improved atmospheric concentrations of chemically-related species have the potential to improve the emission inversion, while the improved precursor's emission estimates benefit the concentration analysis through a reduction in the model forecast error.
- Assimilation of multiple datasets with different vertical sensitivities provides comprehensive constraints on the various emission sources. More datasets will be used to analyze further emission sources (e.g., VOCs) for improving the ozone analysis.
- Emissions from lightning etc (not only at the surface) considerably influence the predictability and the analysis quality of chemical compounds in the troposphere.
- Problems: very high computational cost, model errors etc.

Thanks to Henk Eskes (KNMI), Kengo Sudo (Nagoya Univ.), Takemasa Miyoshi (Riken), Folkert Boersma (Einthovohen Univ.), Michiel van Weele (KNMI)

Miyazaki et al., Global lightning NOx productions estimated from multiple satellite data assimilation, to be submitted.

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Nakamura, Miyazaki et al., A multi-model comparison of stratospheric ozone data assimilation based on an ensemble Kalman filter approach, JGR, 2013.

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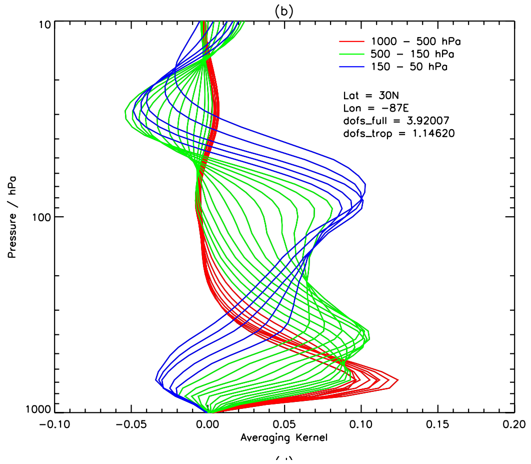
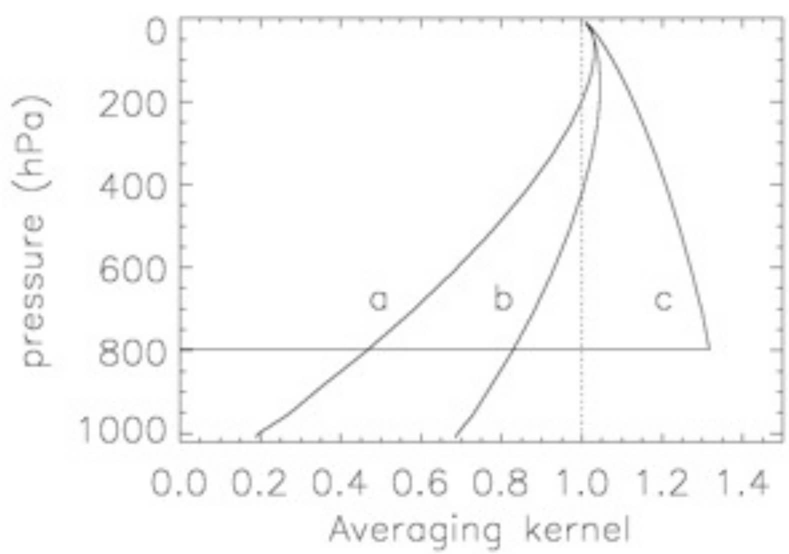
Observation operators

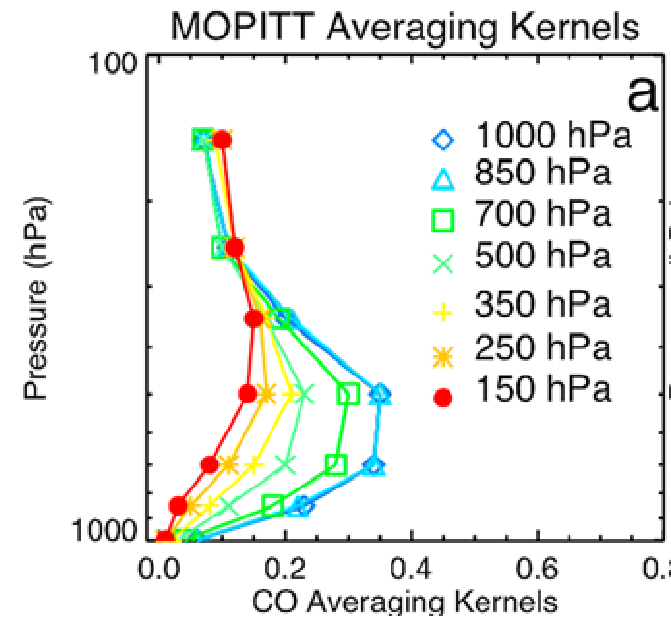
TES O3



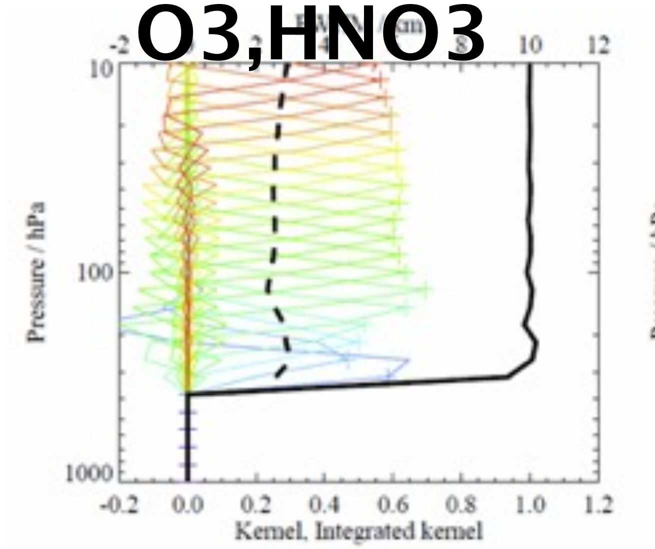
OMI NO2



MOPITT CO



MLS O3,HNO3



- The observation operator (H) converts the model profiles to the profile that would be retrieved from satellite measurements.

$$y^b = H(x) = x_a + \mathbf{A}(S(x) - x_a).$$

- The model-satellite difference (the innovation) is not biased by the a priori profile

$$y^o - y^b = \mathbf{A}(x_{true} - S(x)) + \epsilon,$$

(Rodgers, 2000; Eskes and Boersma, 2003)

- The observational error matrix (R) in each retrieval includes smoothing error, systematic error, measurement error, and representativeness error.