Sources and Regions Attributions to the Upper Troposphere Nitrogen Oxides during the Asian Summer Monsoon

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Motivation and significance of studying NOx in the upper troposphere

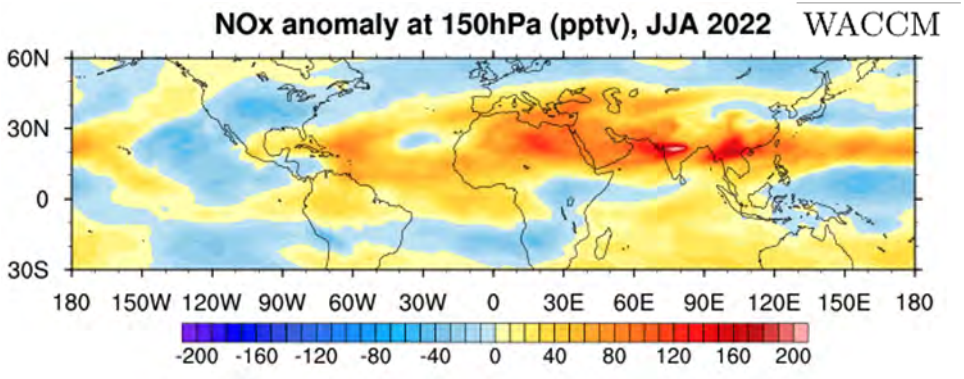
Nitrogen Oxides (NOx) significance in the upper troposphere:

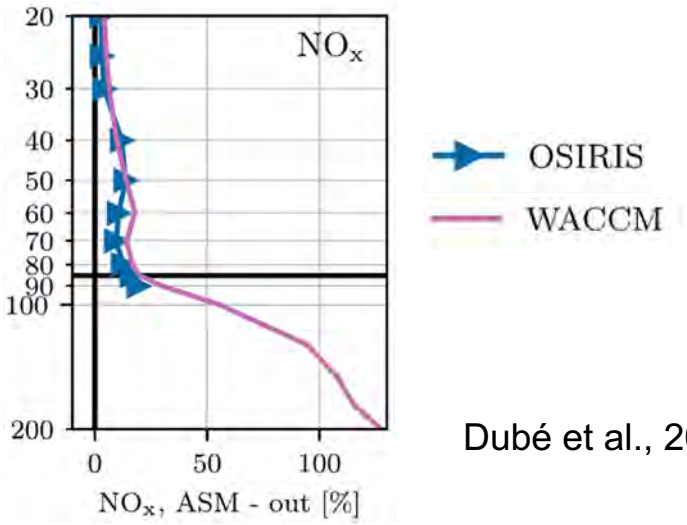
- OH production:
  $NO + HO_2 \Rightarrow OH + NO_2$
- PAN formation:
  $CH_3C(O)OO + NO_2 \Rightarrow PAN$

ASM NOx is an important source of UT NOx.

What is the source of this large positive NOx anomaly in the upper troposphere?

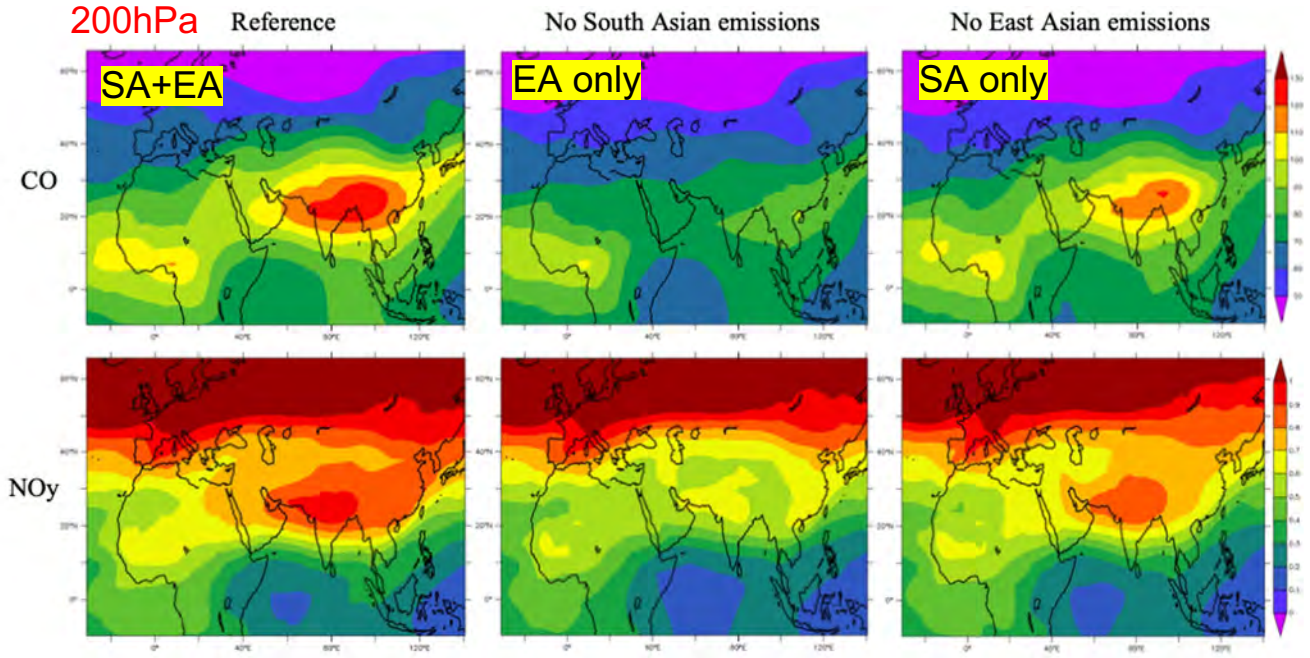
South Asia? East Asia?
Surface anthro.? Lightning? Others?





Dubé et al., 2022

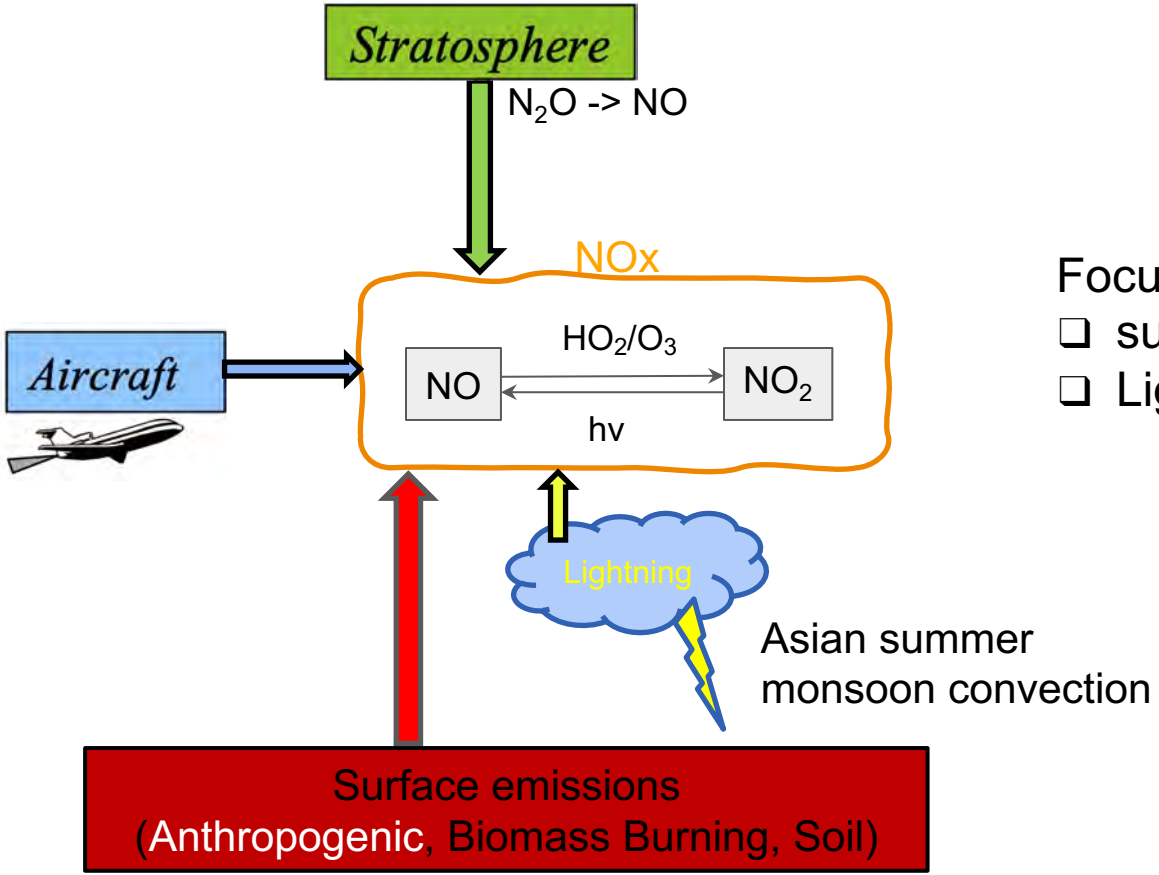
Motivation and significance of studying NOx in the upper troposphere



Lelieveld et al. (2018)

The goal of this study is to thoroughly examine and quantify the contribution of NOx from different regions and sources in the upper troposphere.

NOx sources in the upper troposphere within ASM



Focus on NOx sources:

- ❑ surface anthropogenic (**anthro**);
- ❑ Lightning (**LNO**)

How do we tag NOx?

E.g.,

Original reaction	$NO + HO_2 \rightarrow NO_2 + OH$
Tag reaction	$XNO + HO_2 \rightarrow XNO_2 + HO_2$

N_2O_5

Original reaction: $NO_2 + NO_3 + M \rightarrow NO_2NO_3 + M$

$XNO_2 + NO_3 + M \rightarrow XNO_2NO_3 + NO_3 + M$

$NO_2 + XNO_3 + M \rightarrow XNO_3NO_2 + NO_2 + M$

$XNO_2 + XNO_3 + M \rightarrow XNO_2XNO_3 + M$

- All the sources are considered simultaneously, but each nitrogen emitted is characterized by an artificial tracer XNOX.
- This allows us to follow the evolution of nitrogen from each source and region without affecting the overall chemical system of the atmosphere.
- Compared to the traditional commonly used technique perturbing NOx emissions in a region to determine its impact. This technique has the advantage of eliminating the nonlinearity induced to the chemistry.

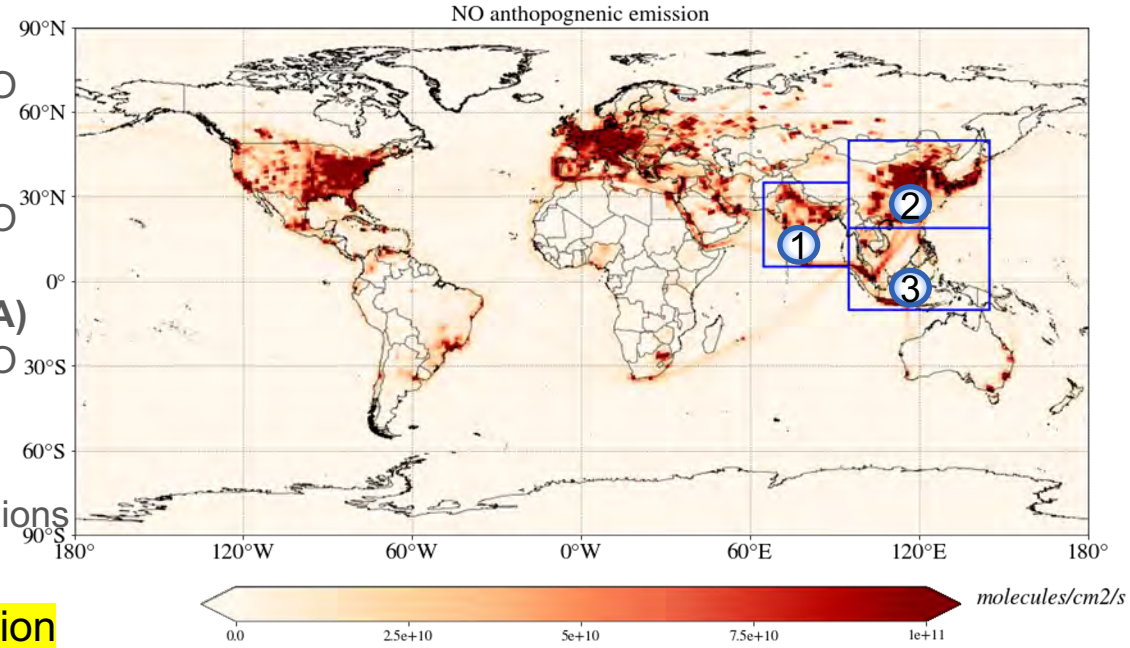
Emmons et al., 2012

NOx tagging in the model simulations

All tropospheric NOx contribution

1. **South Asia (SA)**
   2 sims: Anthro, LNO
2. **East Asia (EA)**
   2 sims: Anthro, LNO
3. **South East Asia (SEA)**
   2 sims: Anthro, LNO

Other region
all other NOx emissions in the troposphere

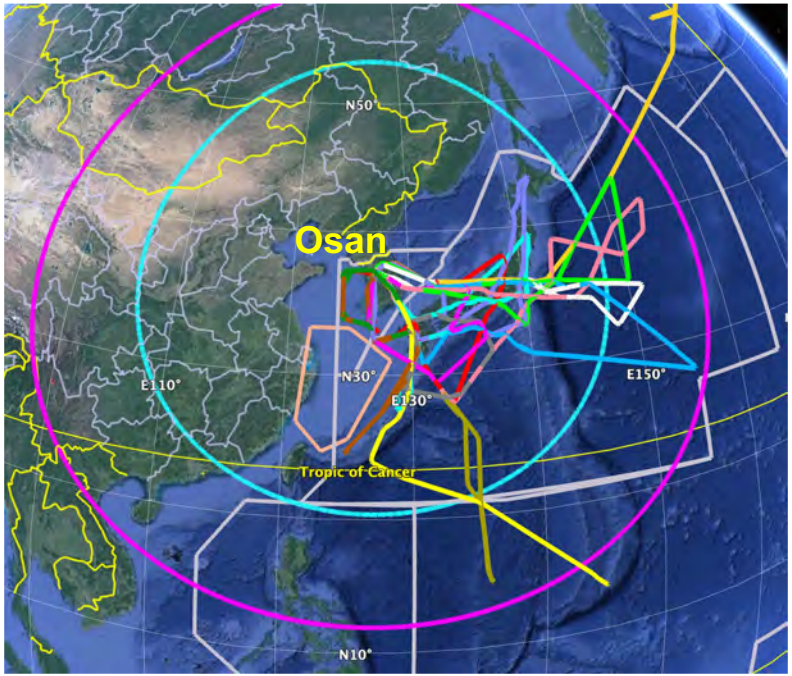


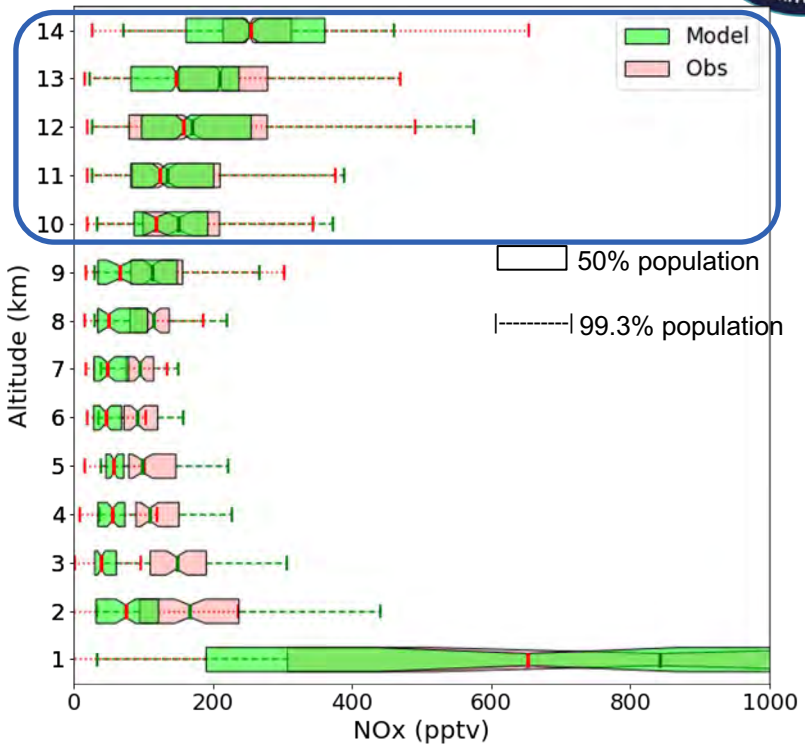
Remaining is stratospheric NOx contribution

- Whole Atmosphere Community Climate Model, version 6 (**WACCM6), 110L**, horizontal resolution of ~**1.0º** and a vertical resolution of ~**500m** in the UTLS.
- Full interactive tropospheric and stratospheric chemistry.
- **Specified dynamics** – nudged towards MERRA-2 reanalysis fields (T, U, V).

ACCLIP vs model NOx

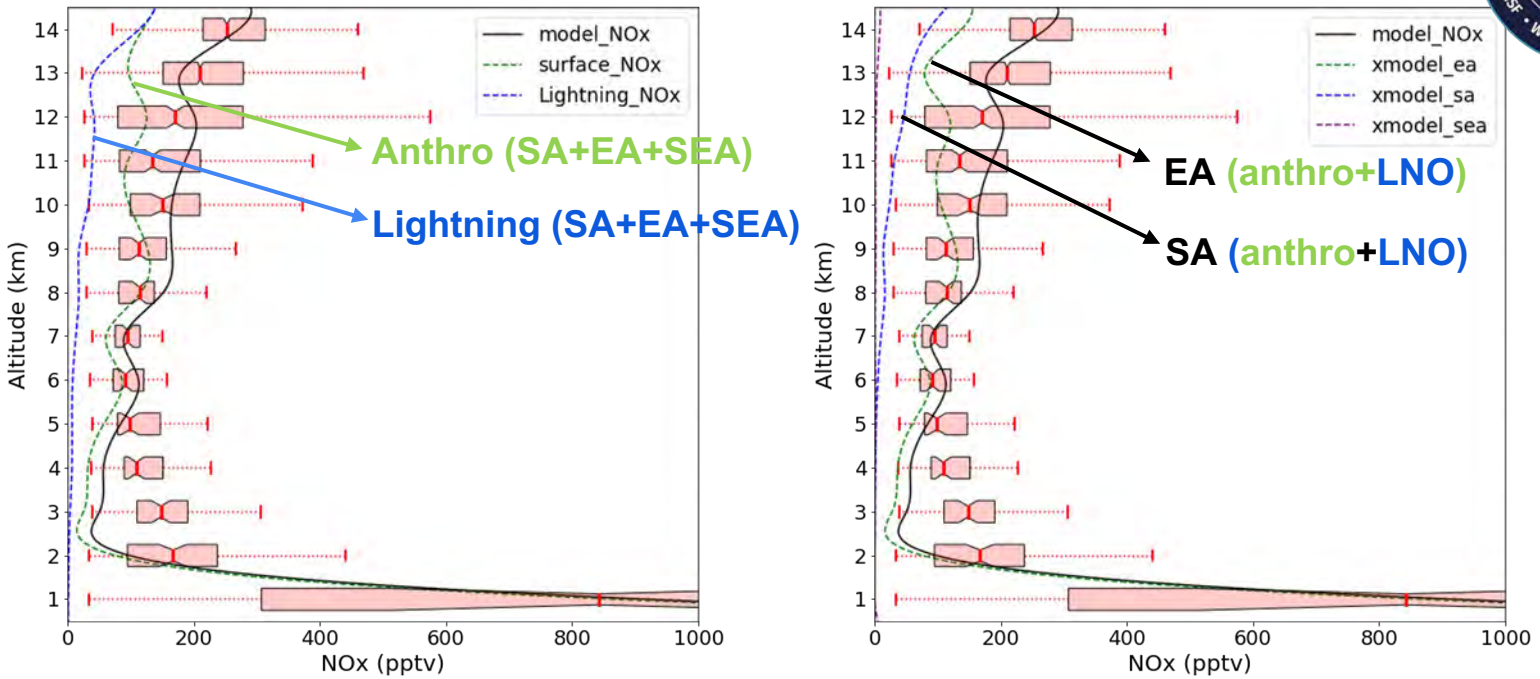
NSF/NCAR GV Flight Tracks
14 Research Flights, July 31 – August 31





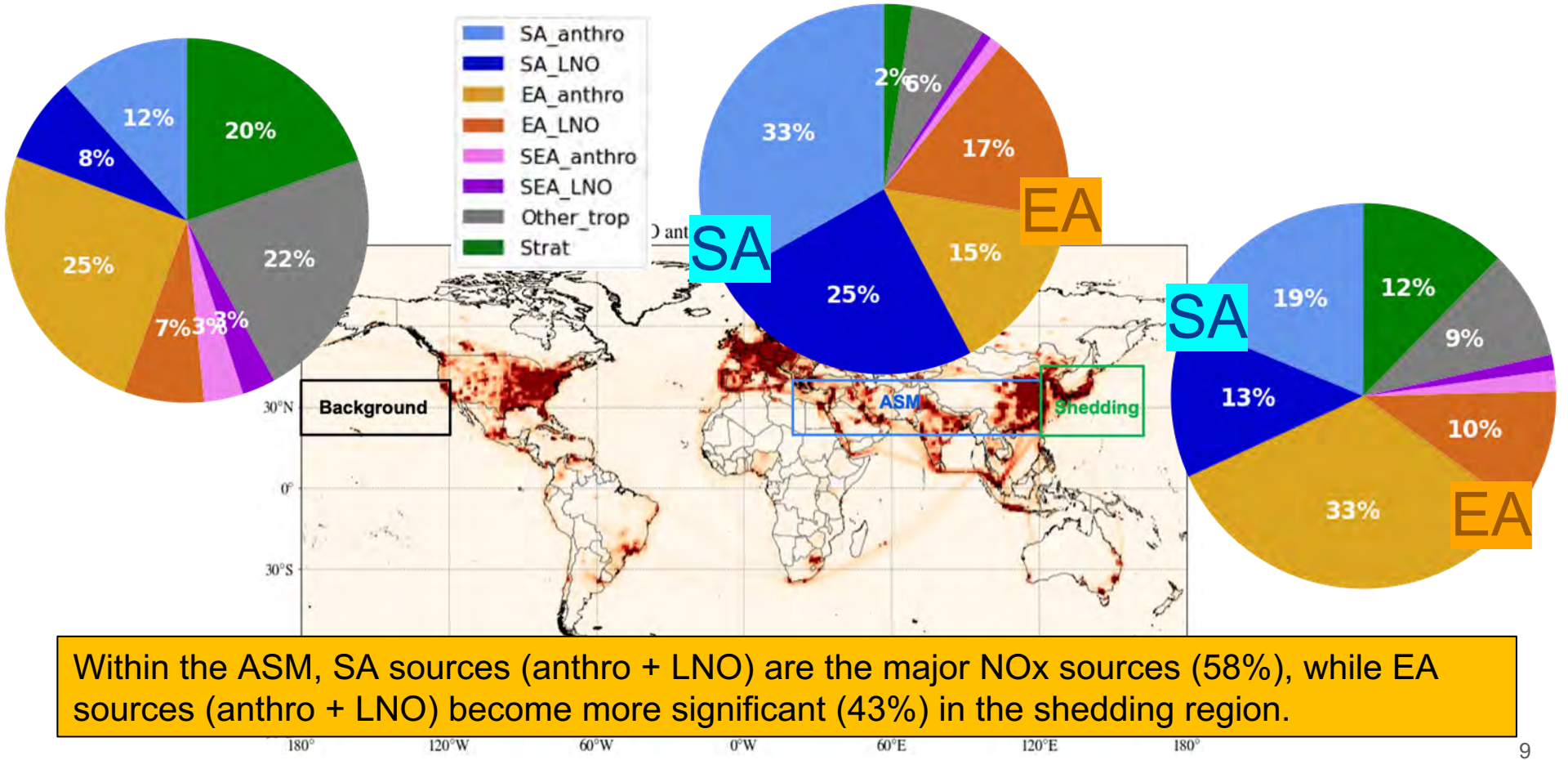
WACCM compares well against the ACCLIP GV NOx data, especially in the upper troposphere.

Source and region attribution of ACCLIP NOx from model



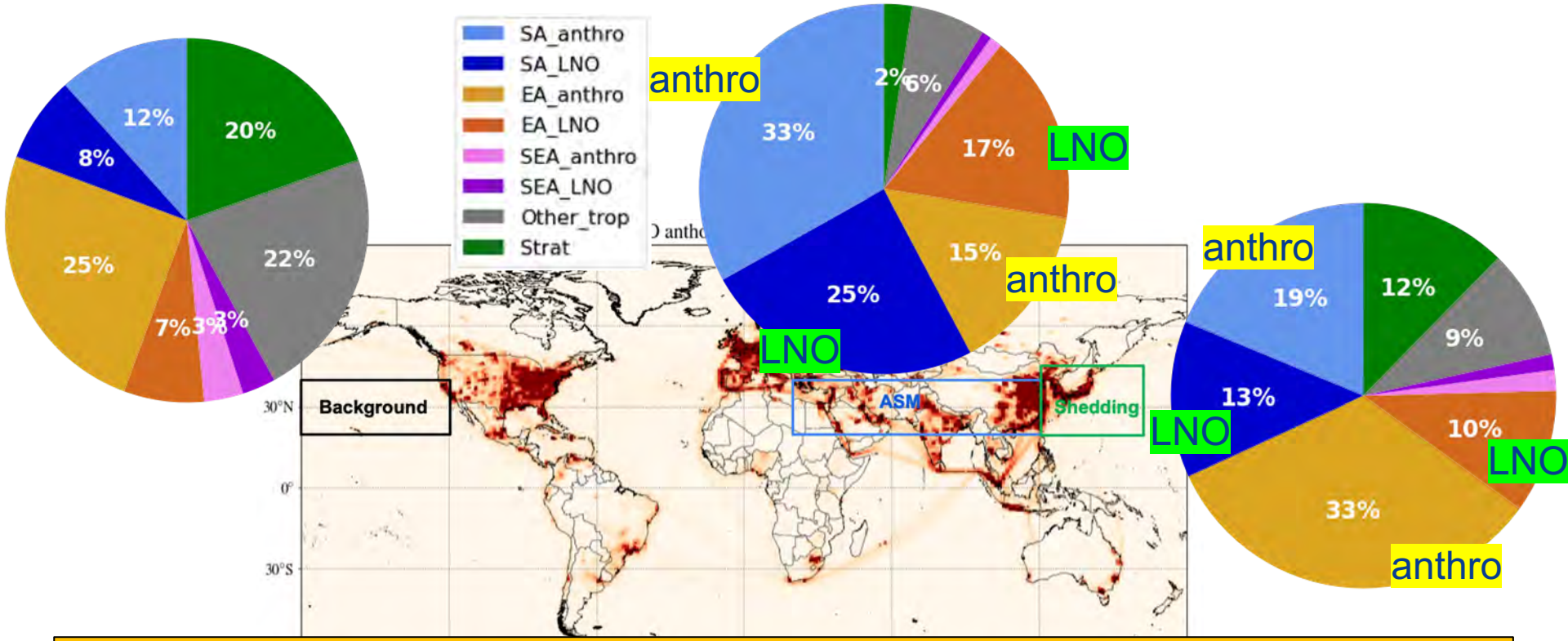
For ACCLIP NOx, surface anthropogenic NOx (SA+EA+SEA) has larger contribution than lightning NOx. NOx sources (anthro+LNO) from East Asia is more important to ACCLIP NOx than South Asia and South East Asia.

NOx sources in the upper troposphere (150 – 200 hPa)



Within the ASM, SA sources (anthro + LNO) are the major NOx sources (58%), while EA sources (anthro + LNO) become more significant (43%) in the shedding region.

NOx sources in the upper troposphere (150 – 200 hPa)

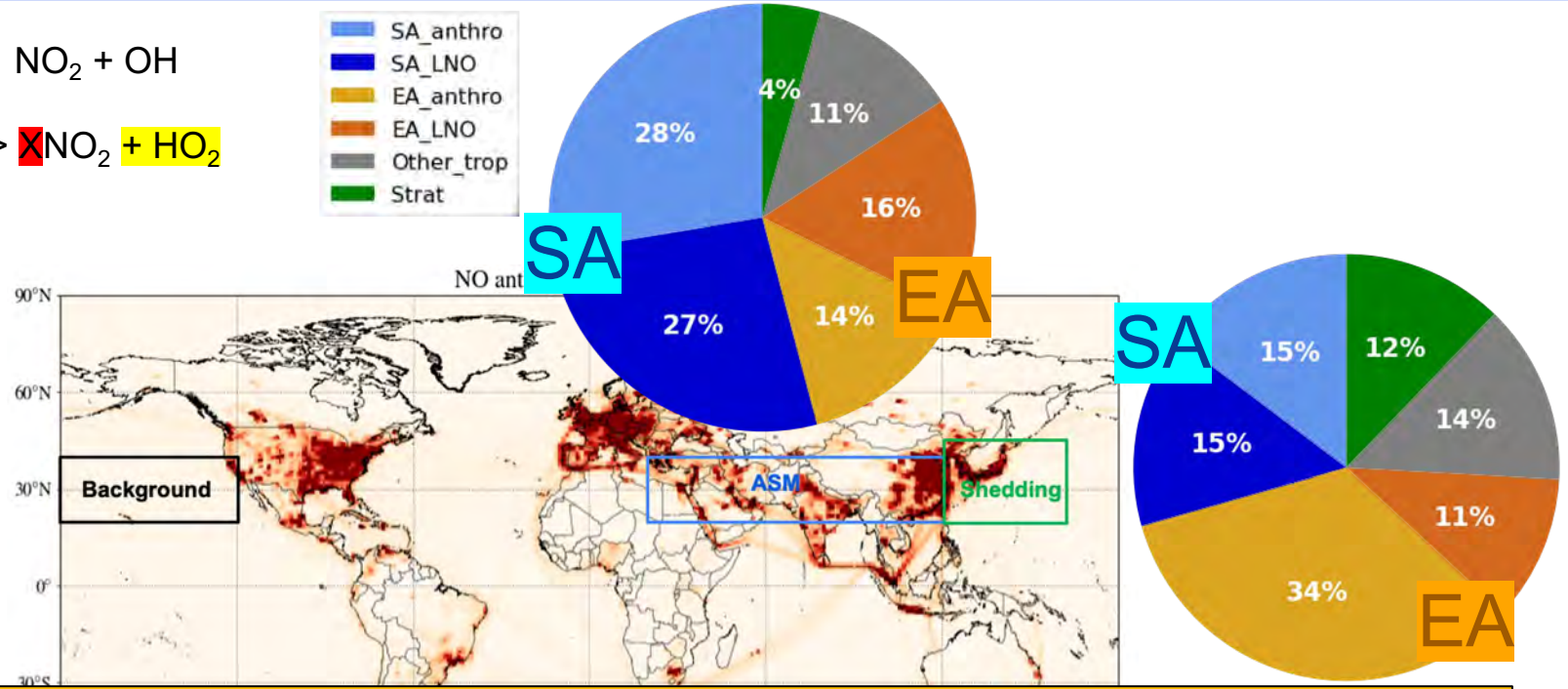


Within ASM, both anthro (48%) and LNO (42%) sources are similarly important, while in the shedding region the anthropogenic sources have larger contribution (~30%) to the UT NOx than lightning.

NO to OH production in the upper troposphere (150 – 200 hPa)

$NO + HO_2 \Rightarrow NO_2 + OH$

$XNO + HO_2 \Rightarrow XNO_2 + HO_2$



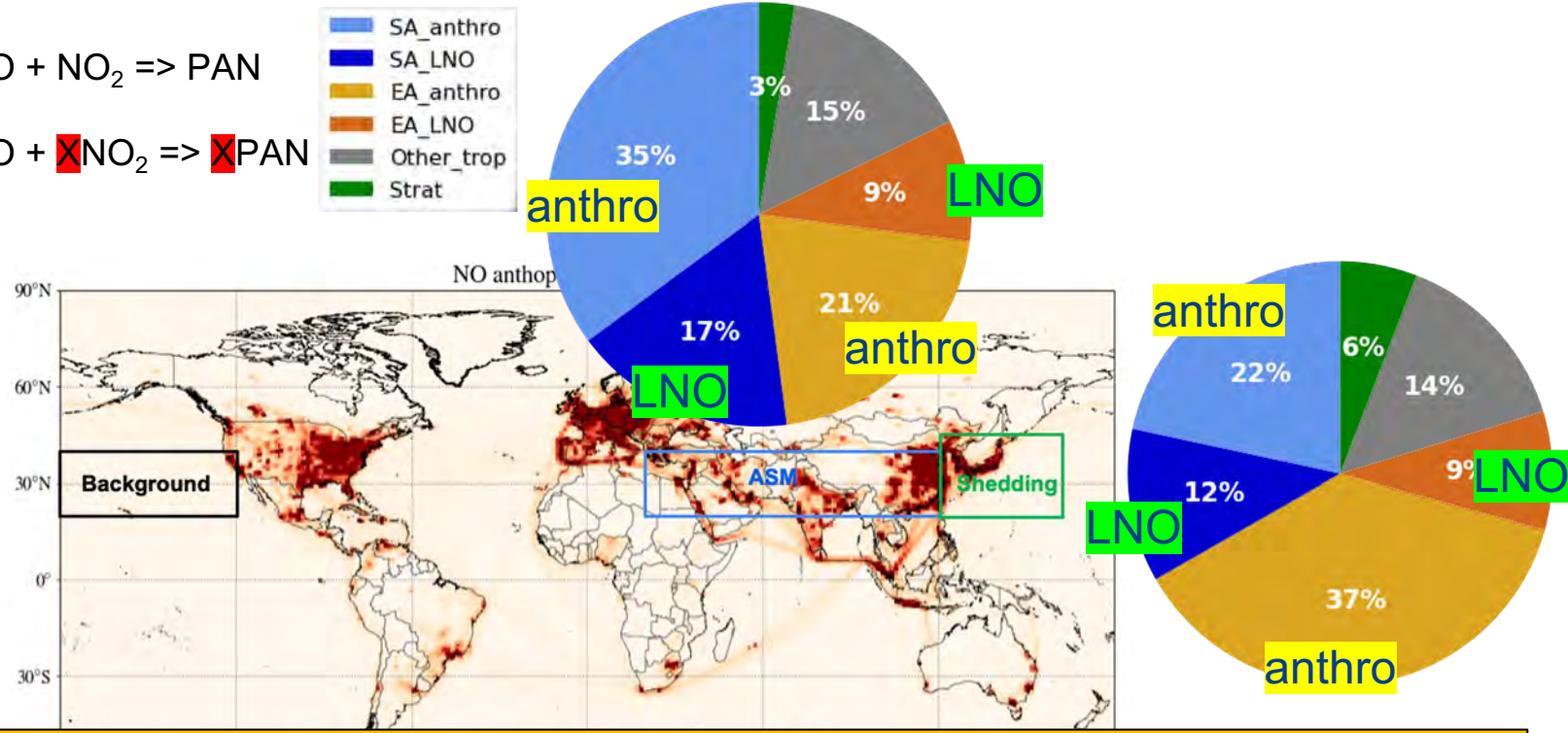
NO to OH production shows similar results as NOx.

SA sources (anthro + LNO) are the major NOx sources (55%) within the ASM, while EA sources become more significant (45%) in the shedding region.
Within ASM, both anthro and LNO sources are equally important, while in the shedding region the anthropogenic sources have larger contribution to the UT NOx than lightning.

PAN formation in the upper troposphere (150 – 200 hPa)

$CH_3C(O)OO + NO_2 \Rightarrow PAN$

$CH_3C(O)OO + XNO_2 \Rightarrow XPAN$

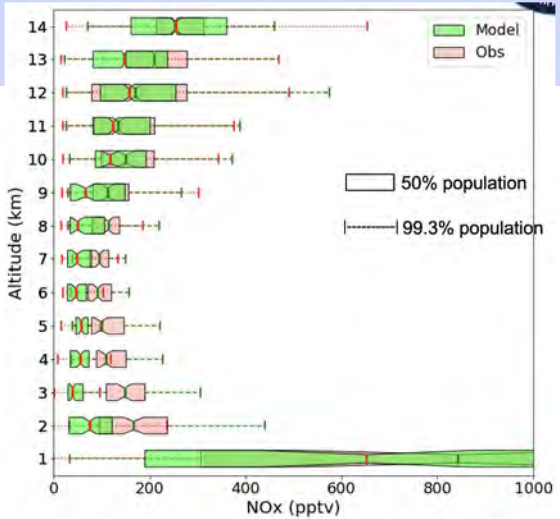


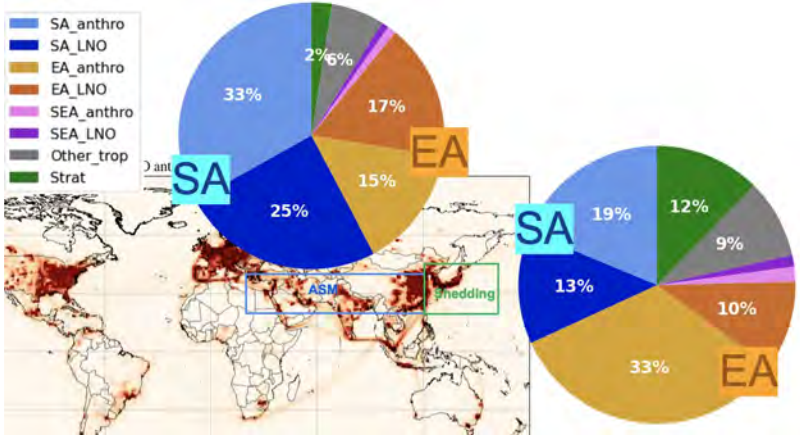
Within ASM, SA and EA anthropogenic NOx account for 56% of total PAN formation, which is more than doubling the contribution from lightning (26%).

Anthropogenic NOx seem to have higher efficiency in PAN formation than lightning NOx.

Conclusions

- WACCM 110L compares very well against the ACCLIP NOx data, especially in the upper troposphere, where most of ACCLIP data are measured.
- Within the ASM, SA sources (anthro + LNO) are the major NOx sources, while EA sources (anthro + LNO) become more significant in the shedding (ACCLIP) region.
- In the shedding (ACCLIP) region the anthropogenic source have larger contribution to the upper tropospheric NOx than lightning.





Thanks for your attention. ☺

Contact me with any questions and comments at jzhan166@ucar.edu

ACCLIP vs model Ozone