Relative Contributions of Anthropogenic and Lightning Nitrogen Sources in the Upper Troposphere during the Asian Summer Monsoon

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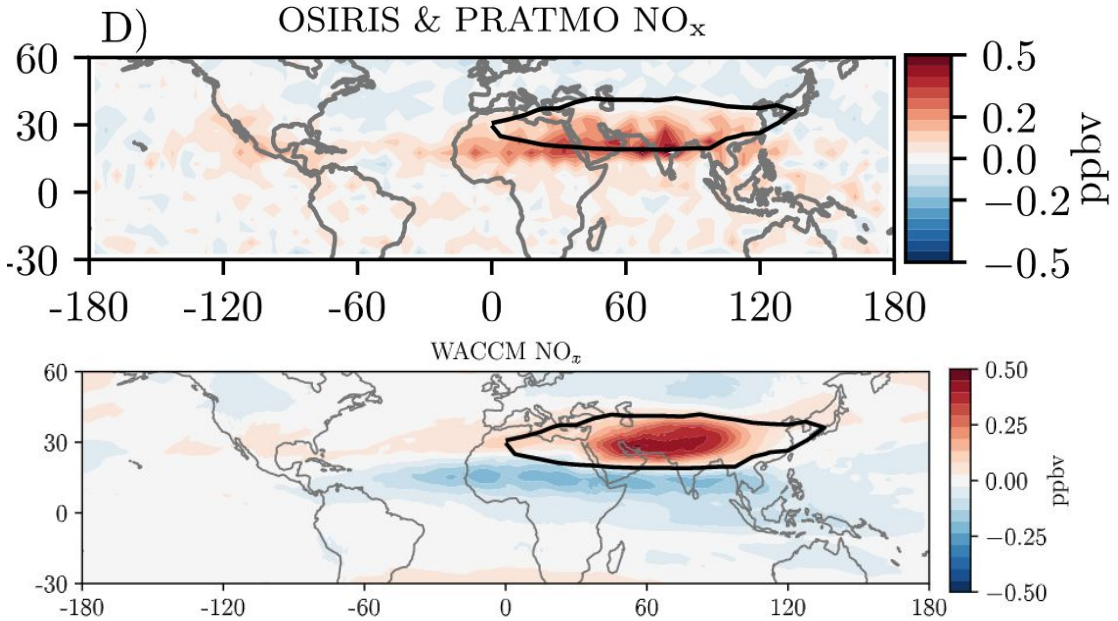
Motivation and significance of studying NOx in the upper troposphere

Previous study show NOx anomaly over the ASM region, but the source of this NOx anomaly is unclear.
Fadnavis et al. (2015) found that the transport of emissions to the UTLS is the largest factor affecting NOx in the ASM.
Lelieveld et al. (2018) found that lightning is still a key source of NOx in the upper tropospheric monsoon anticyclone.



Dubé et al., 2022

NO significance in the upper troposphere:

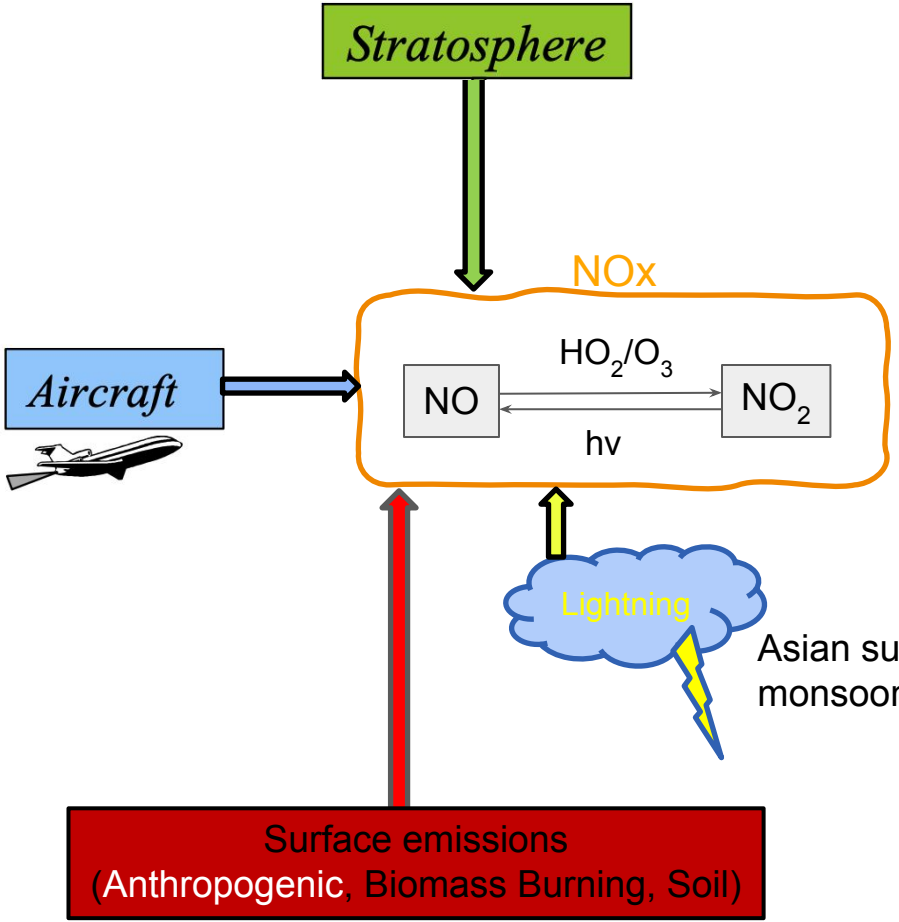
OH production:

$NO + HO_2 \Rightarrow OH + NO_2$

PAN formation:

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

NOx sources in the upper troposphere within ASM



Focus on NOx sources:

- surface anthropogenic;
- lightning

NOx tagging **anthropogenic and lightning** sources and regions

South Asia:

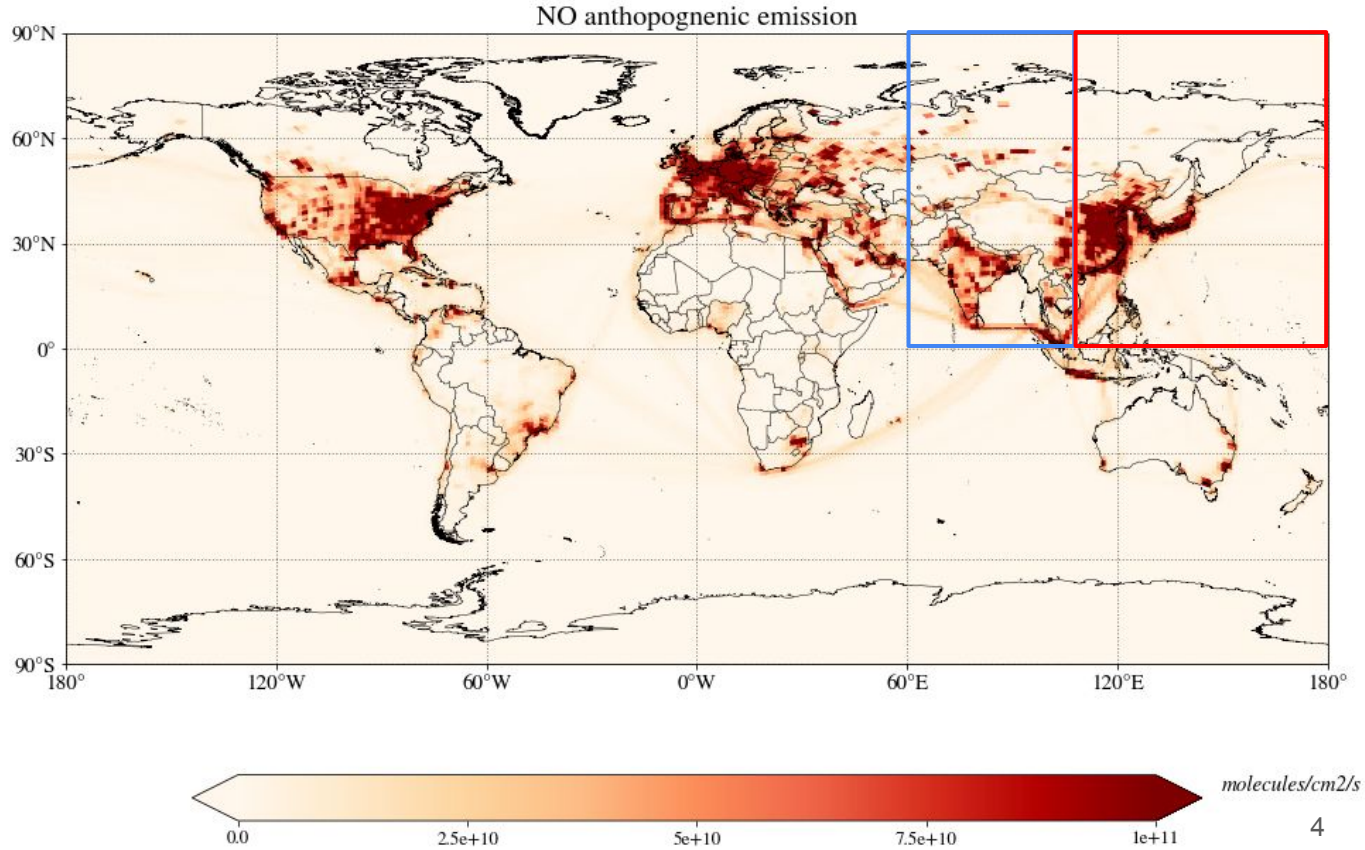
0-90 Lat; 60-100 Lon

East Asia:

0-90 Lat; 100-180 Lon

Other region:

all the regions outside South and East Asia



How we tag NOx?

E.g.,

NO + HO2 -> NO2 + OH

XNO + HO2 -> XNO2 + HO2

NO2 + NO3 + M -> N2O5 + M

NO2 + XNO3 + M -> XNO3NO2 + NO2 + M

XNO2 + NO3 + M -> XNO2NO3 + NO3 + M

XNO2 + XNO3 + M -> XNO2XNO3 + M

- 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.

Emmons et al., 2012

Model setup and simulations

- Whole Atmosphere Community Climate Model, version 6 (**WACCM6), 110L**, horizontal resolution of ~**1.0°** and a vertical resolution of ~**500m** in the UTLS.
- Full interactive tropospheric and stratospheric chemistry.
- **Specified dynamics** – nudged towards MERRA-2 reanalysis fields from the NASA Goddard Earth Observing System version 5 (GEOS5) (T, U, V)

Simulations	Tags
1. Anthropogenic SA	Only South Asia anthropogenic NOx
2. Anthropogenic EA	Only East Asia anthropogenic NOx
3. Lightning SA	Only South Asia Lightning NOx
4. Lightning EA	Only East Asia Lightning NOx
5. Other_trop	Anthropogenic and Lightning NOx in other region, biomass burning, soil, aircraft NOx
	Remaining is stratospheric NOx contribution

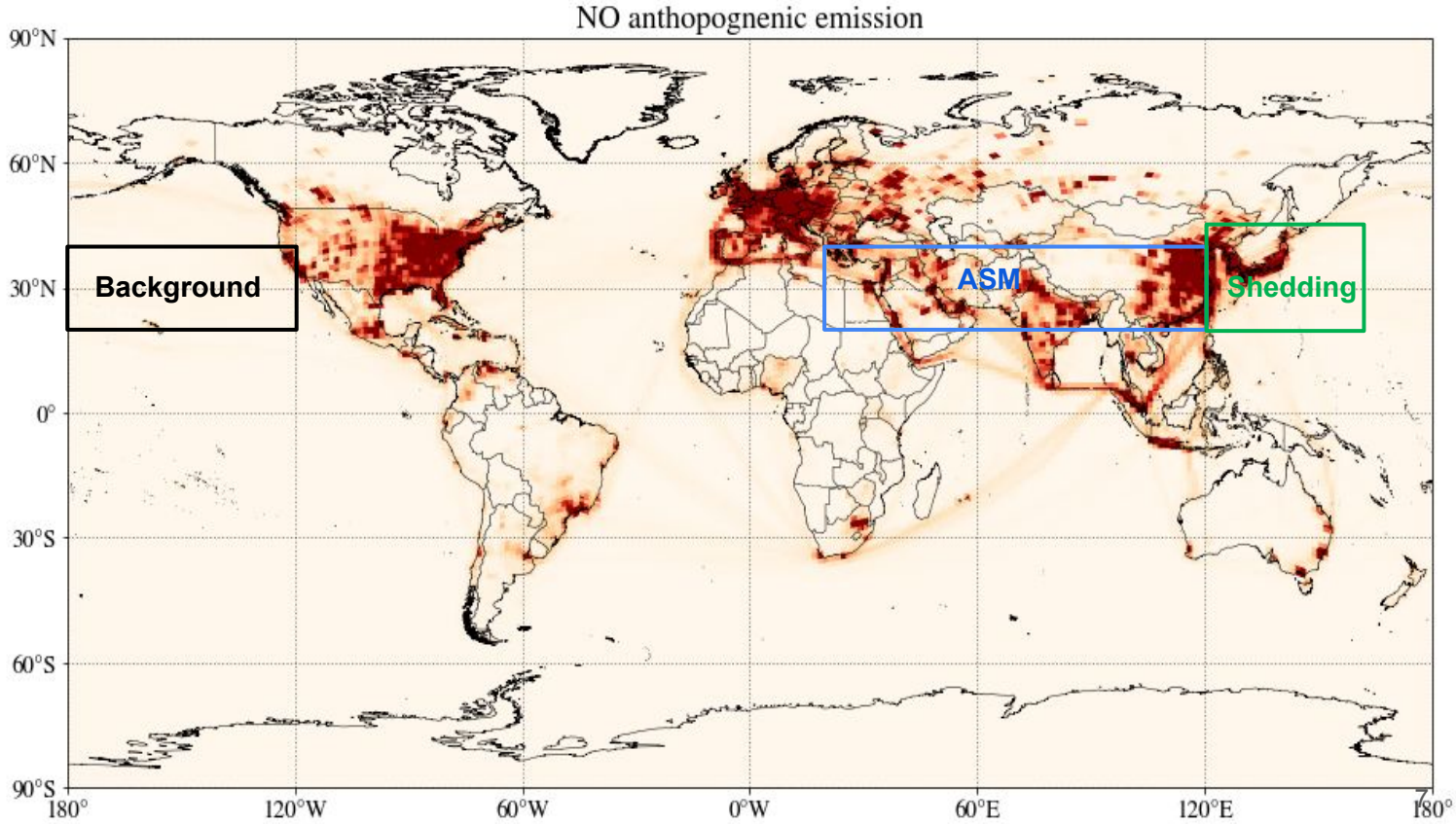
All tropospheric NOx contribution

Definition of Background, ASM anticyclone, east shedding region

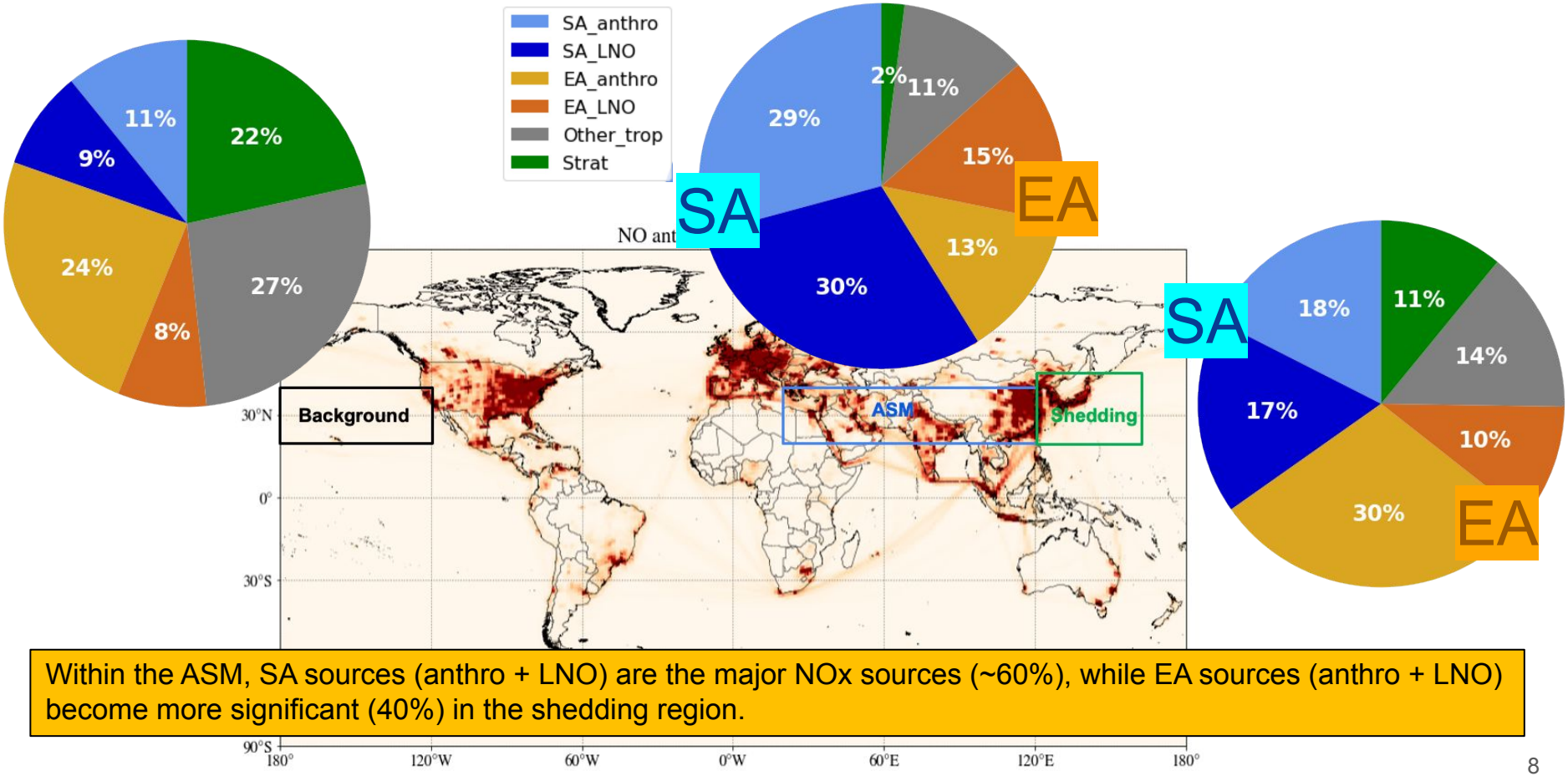
Background:
20°–40°N,
180°–120°W

ASM anticyclone:
20°–40°N,
20°–120°E

East shedding:
20°–45°N,
120°–160°E



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



Within the ASM, SA sources (anthro + LNO) are the major NOx sources (~60%), while EA sources (anthro + LNO) become more significant (40%) in the shedding region.

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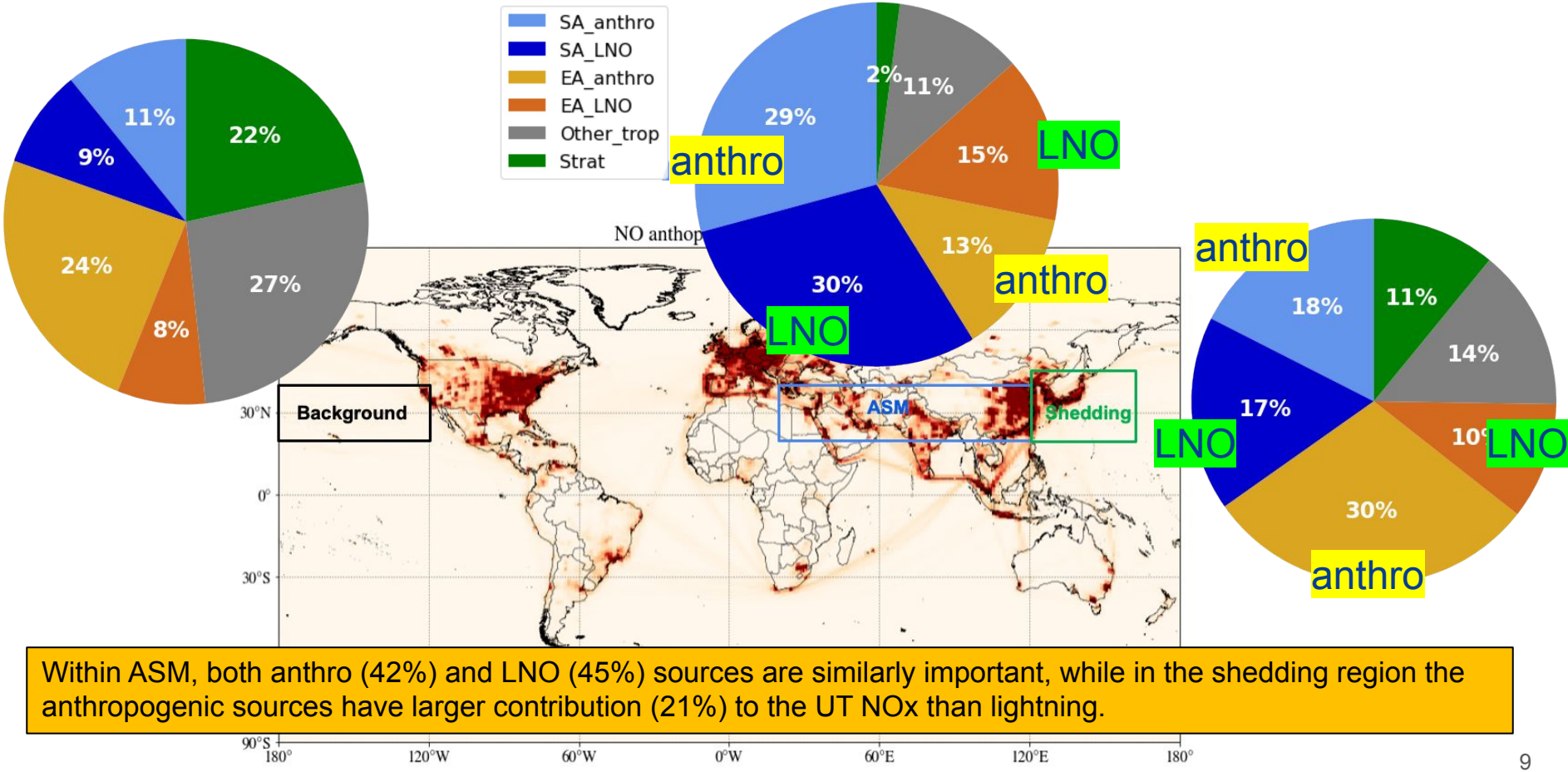
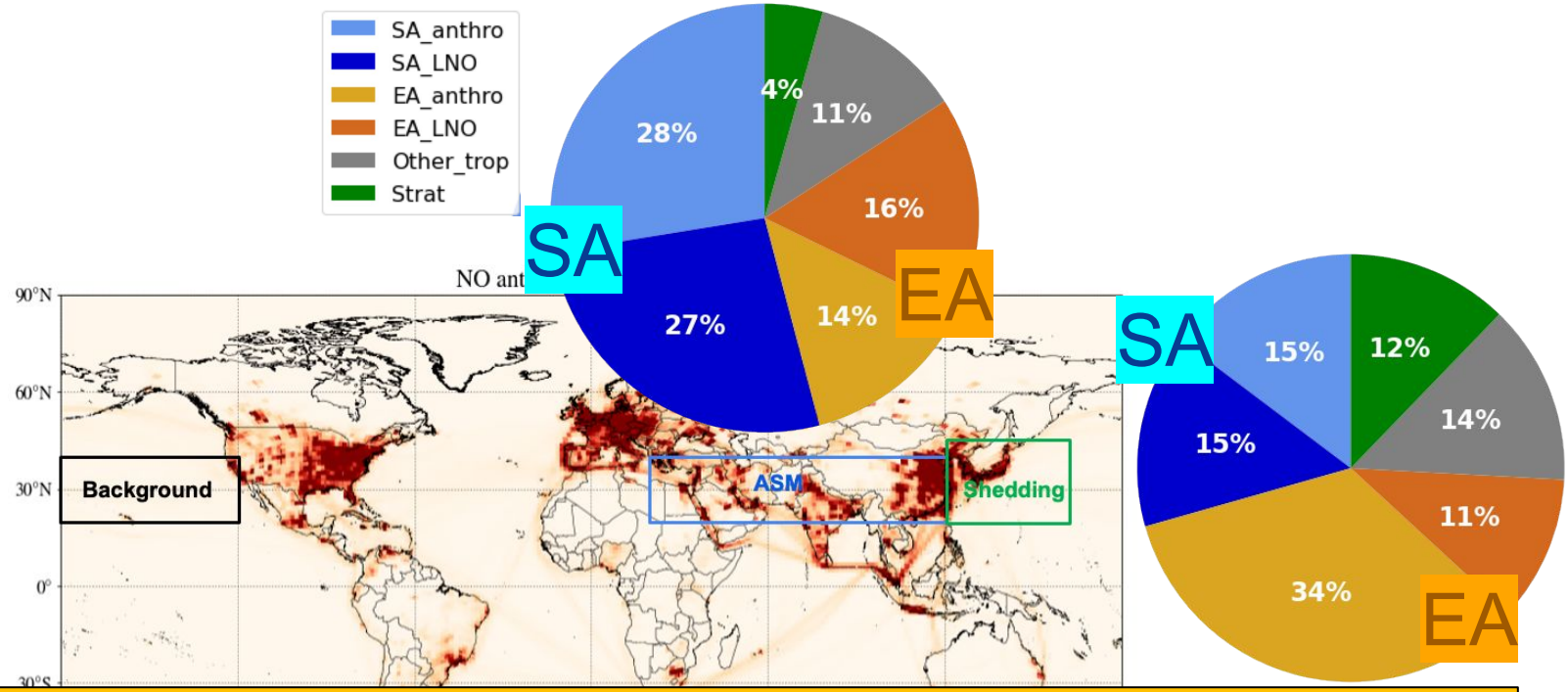


Within ASM, both anthro (42%) and LNO (45%) sources are similarly important, while in the shedding region the anthropogenic sources have larger contribution (21%) to the UT NOx than lightning.

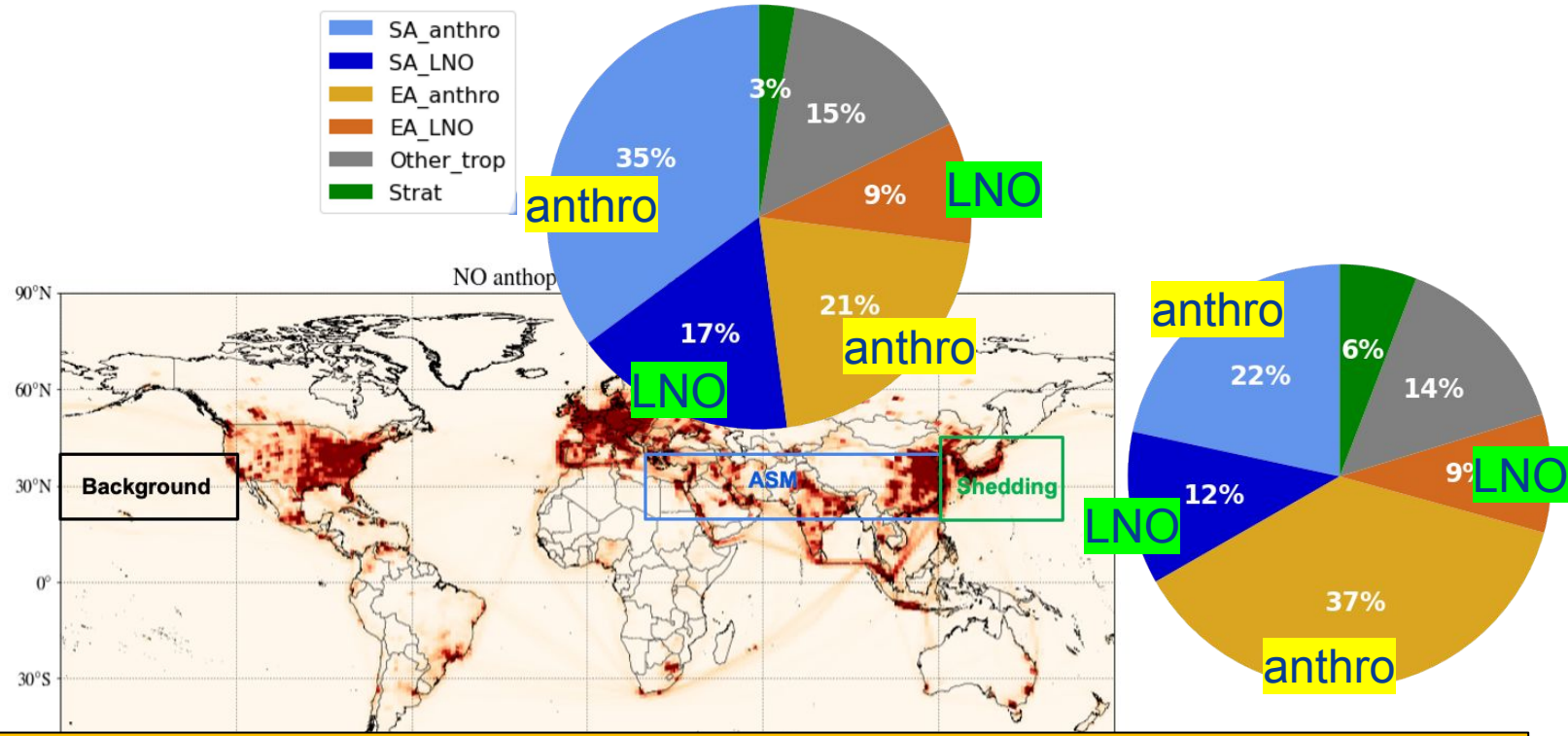
NO to OH production in the upper troposphere



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



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.

Summary

- Tagging mechanism works in this study.

- For NOx abundance and OH formation:
  - SA sources (anthro + LNO) are the major NOx sources (~60%) within the ASM, while EA sources (anthro + LNO) become more significant (40%) in the shedding region.

  - Within ASM, both anthro (42%) and LNO (45%) sources are similarly important, while in the shedding region the anthropogenic sources have larger contribution (21%) to the UT NOx than lightning.
- For PAN, anthropogenic NOx seem to have higher efficiency in PAN formation than lightning NOx.

- **Next step**: compare the model results to ACCLIP data.

Thank you for you attention. ☺

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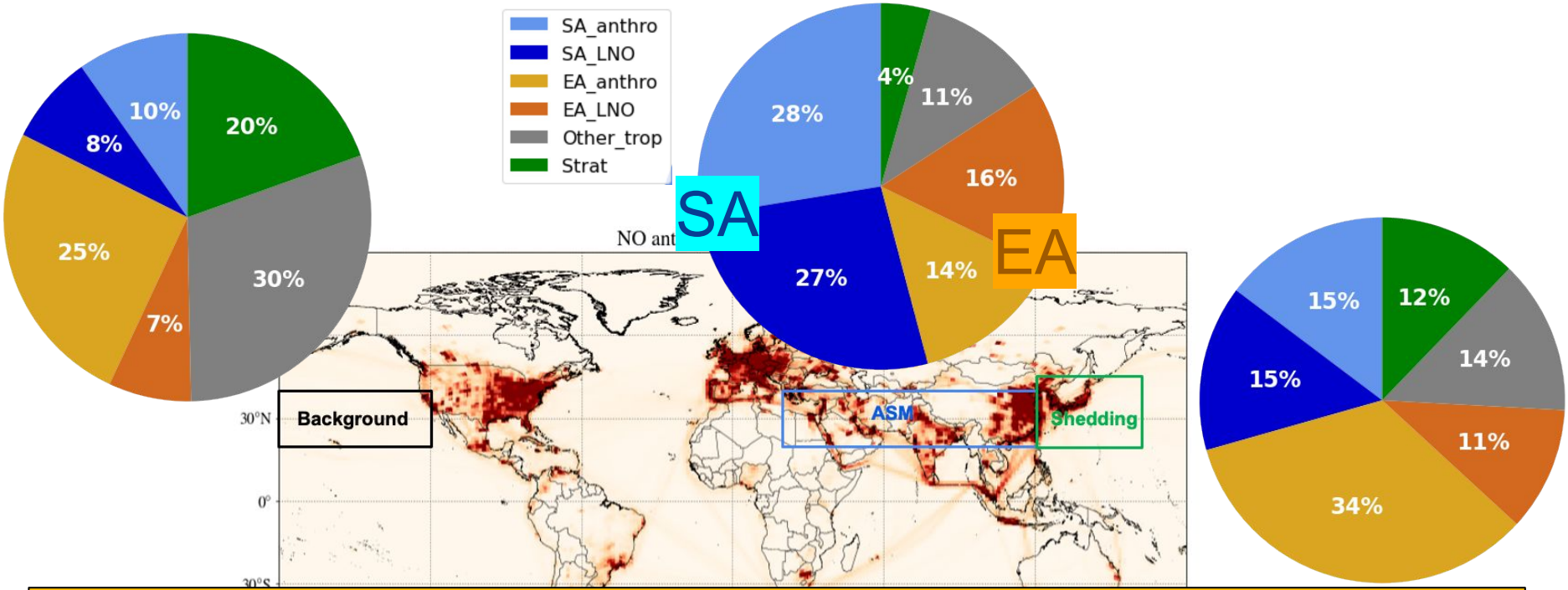

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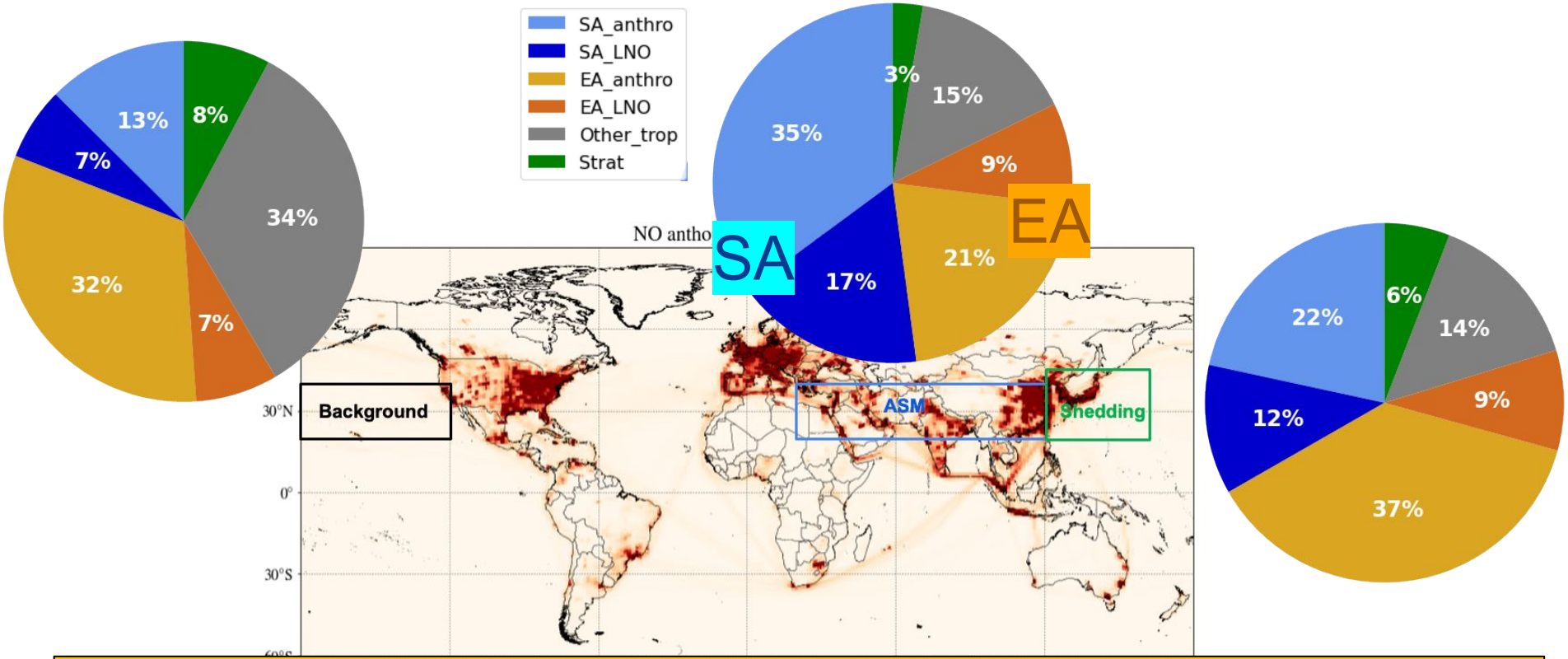

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