Global burden of tropospheric ozone:

Effects of particulate nitrate photolysis and assimilation of satellite NO_2 measurements

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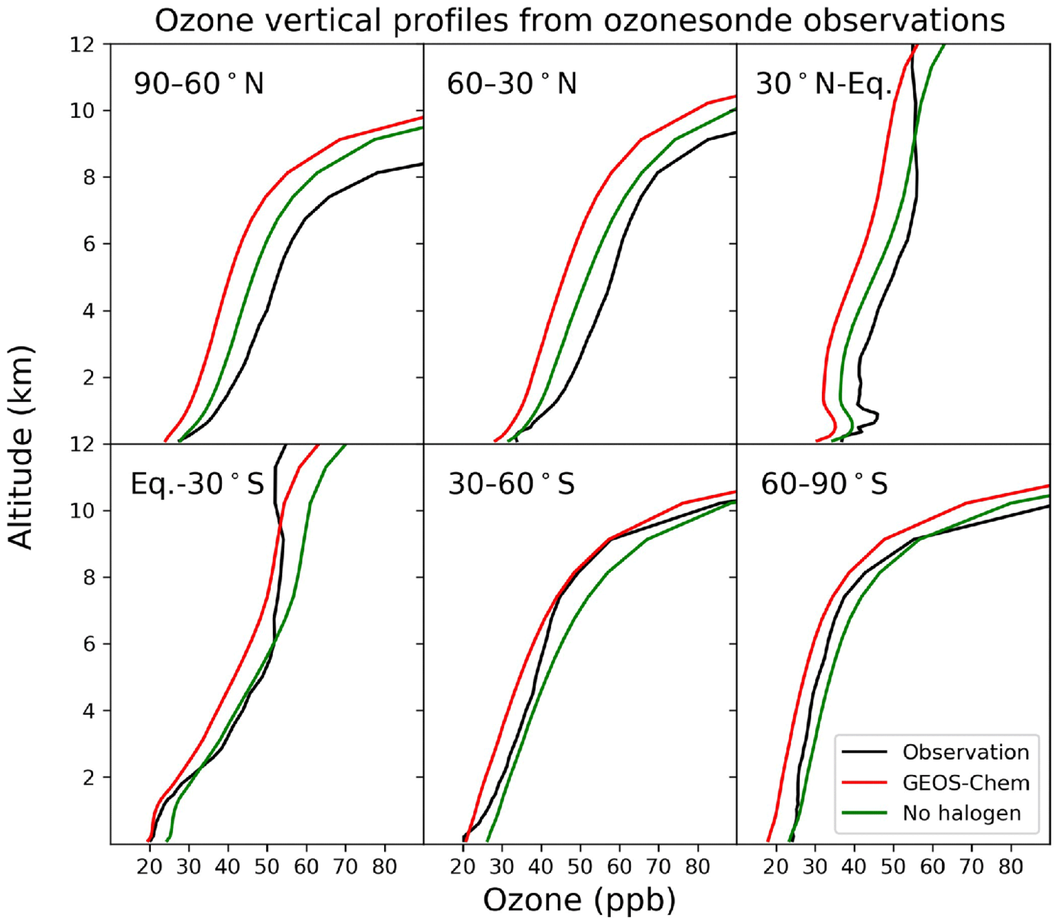
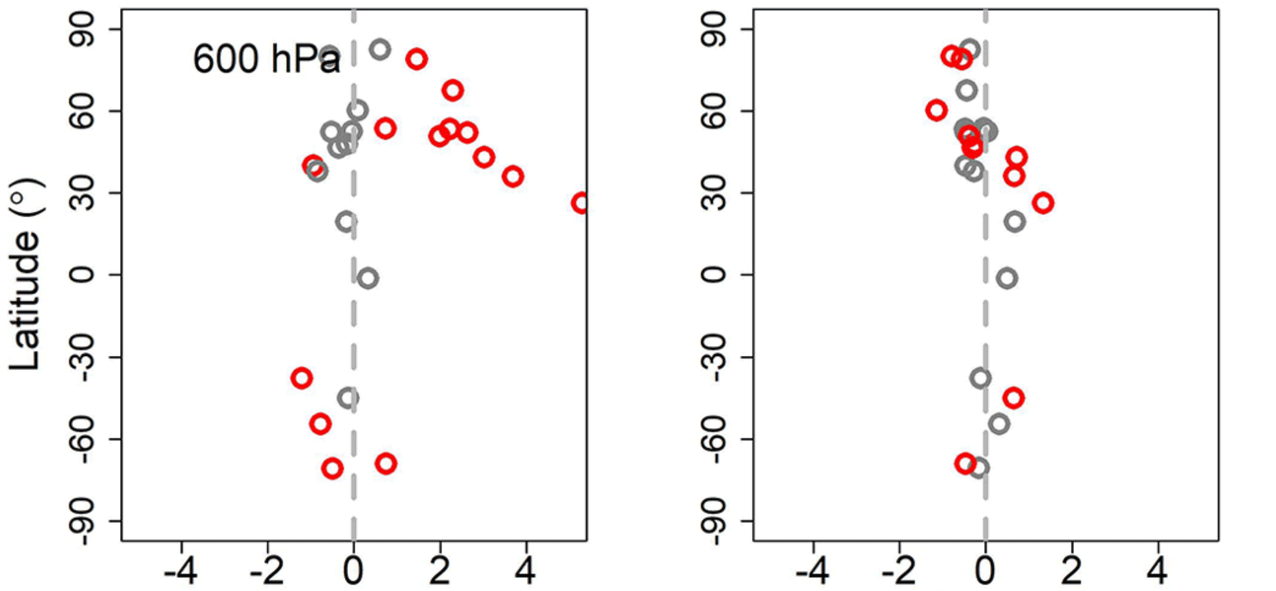
Tropospheric ozone in GEOS-Chem



(X. Wang et al. 2021)

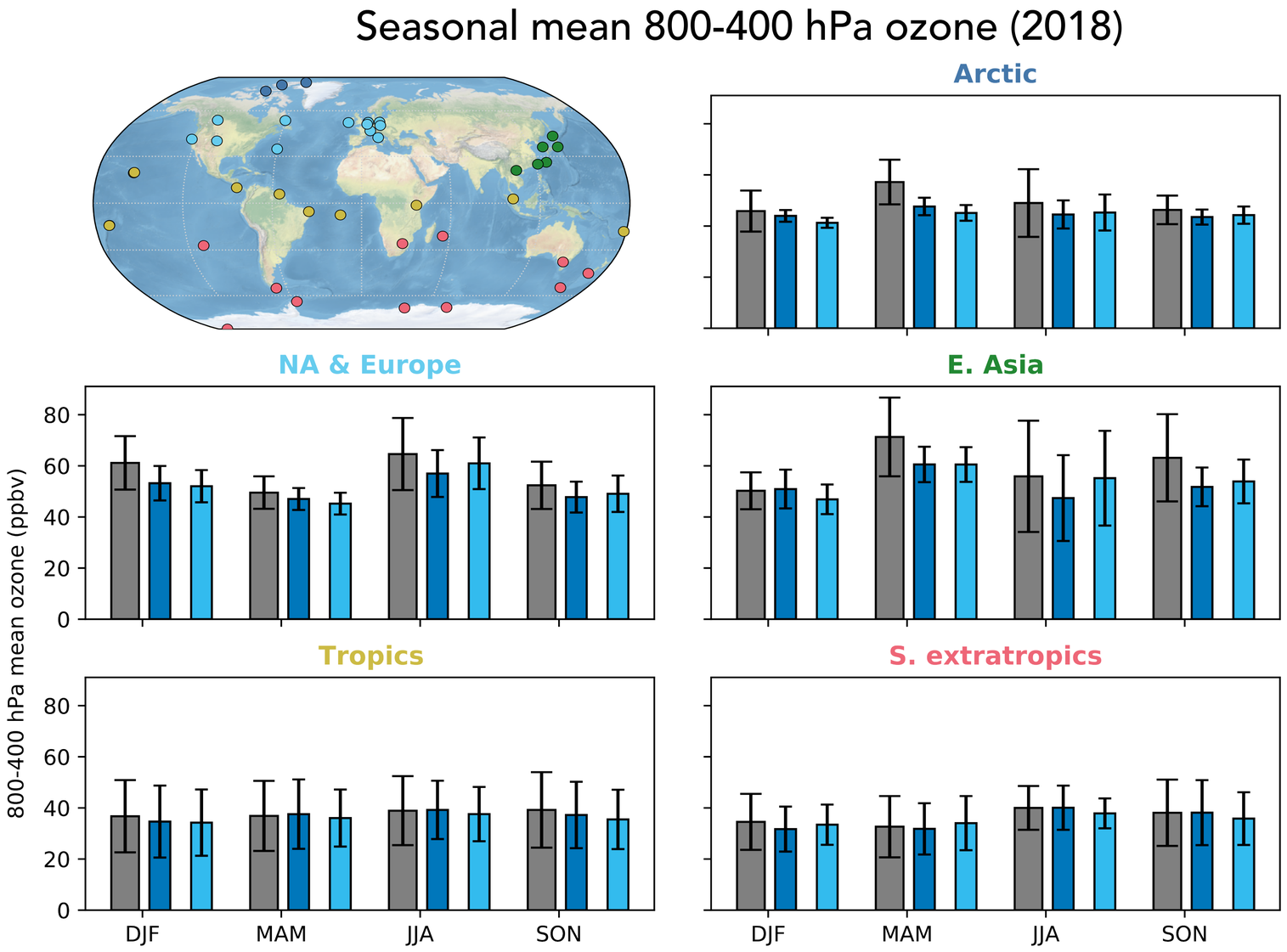
GEOS-Chem underestimates free tropospheric ozone concentrations and trends in the Northern midlatitudes

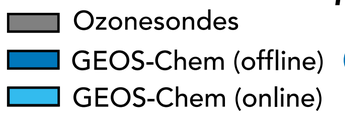
1990-2017 FT ozone trends
(ppbv/decade)



(Christiansen al. 2022)

Free tropospheric ozone in GEOS-Chem





GEOS-Chem version 14.0.0
No sea salt bromine source

CTM (4° x 5°)
Run within GEOS-5 (C90 or ~1°)

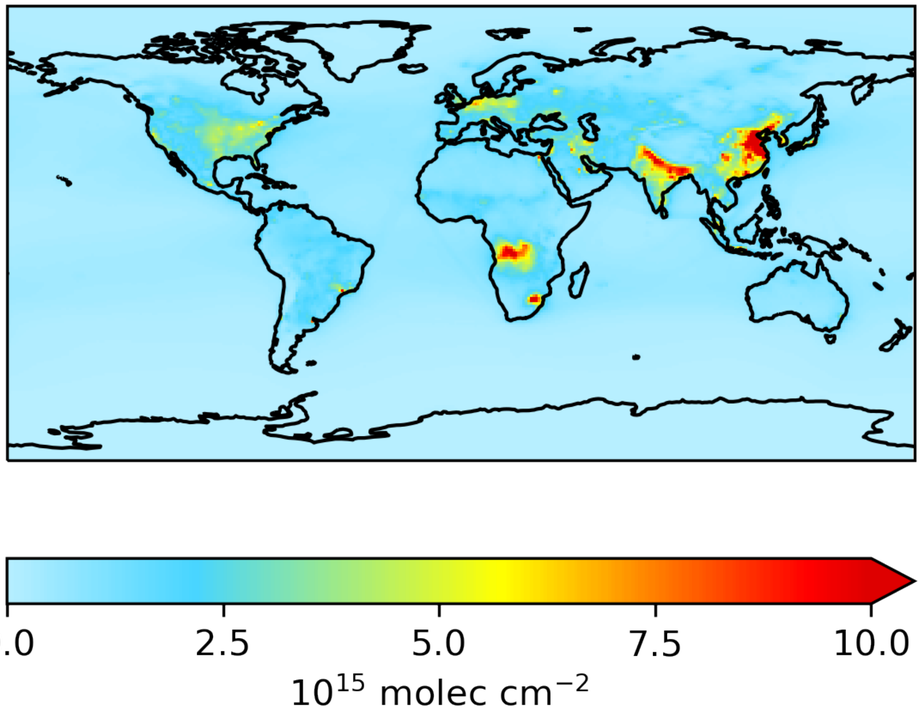
Offline version: ozone concentrations 5-10 ppbv too low over N. Amer, Europe & E. Asia

Improved summertime ozone in online simulation

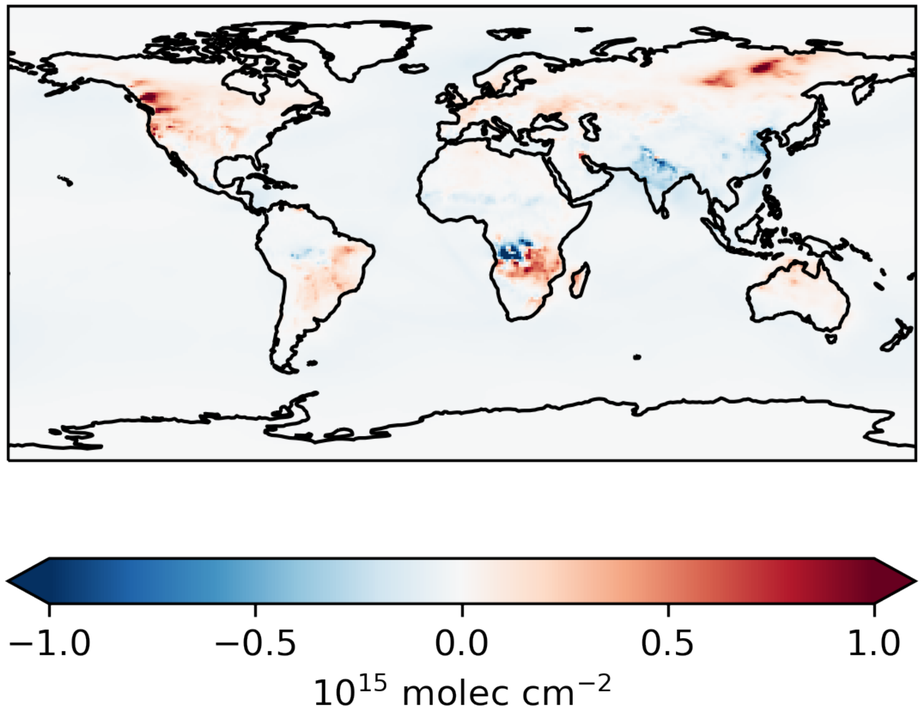
Effect of assimilating OMI NO_2 observations

3D-Var assimilation of OMI-retrieved NO_2 columns in the online GEOS-Chem model

JJA NO_2 tropospheric columns in online GEOS-Chem

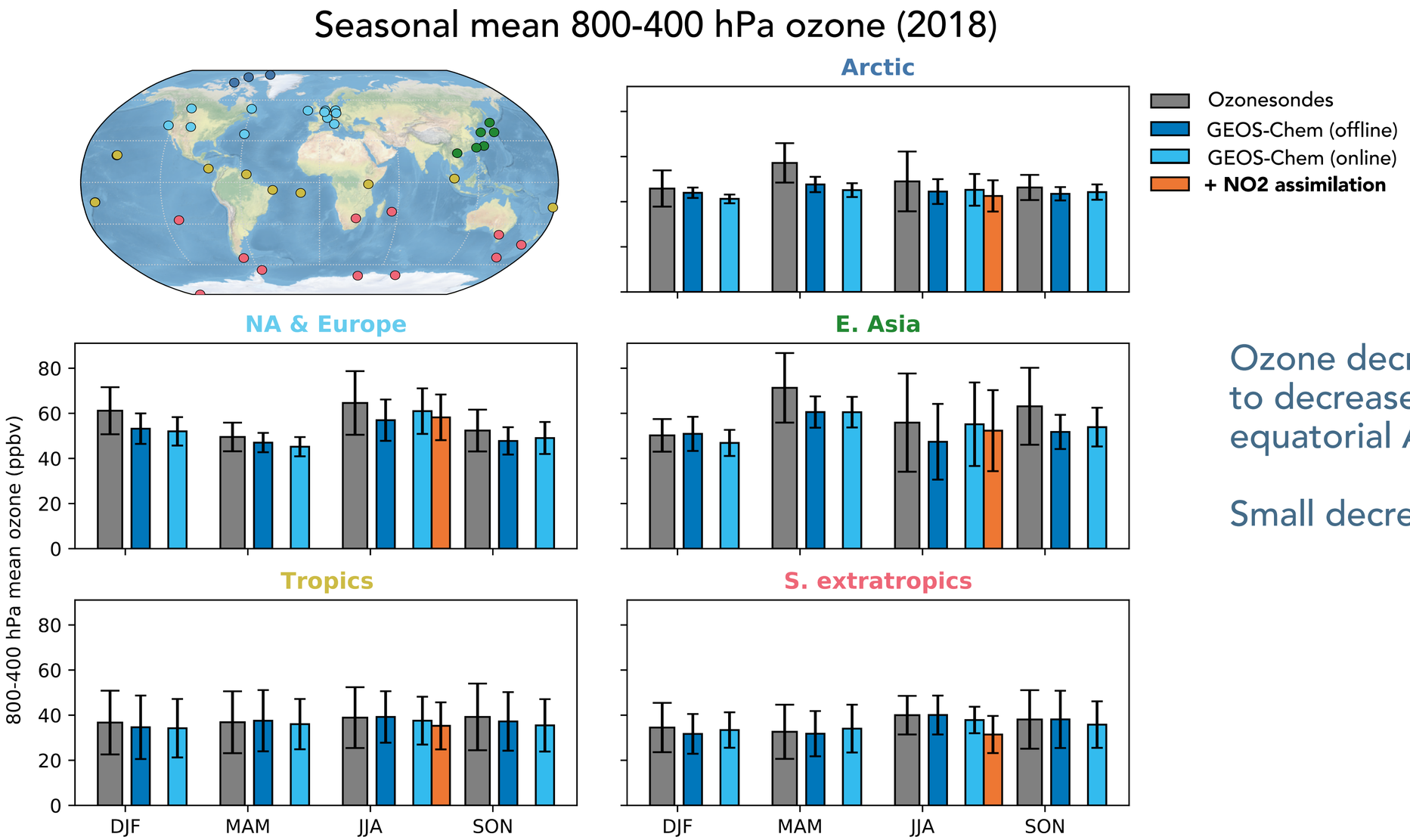


Assimilated NO_2 columns *minus* background NO_2 columns



Significant decreases in NO_2 over equatorial Africa; smaller changes over India and China

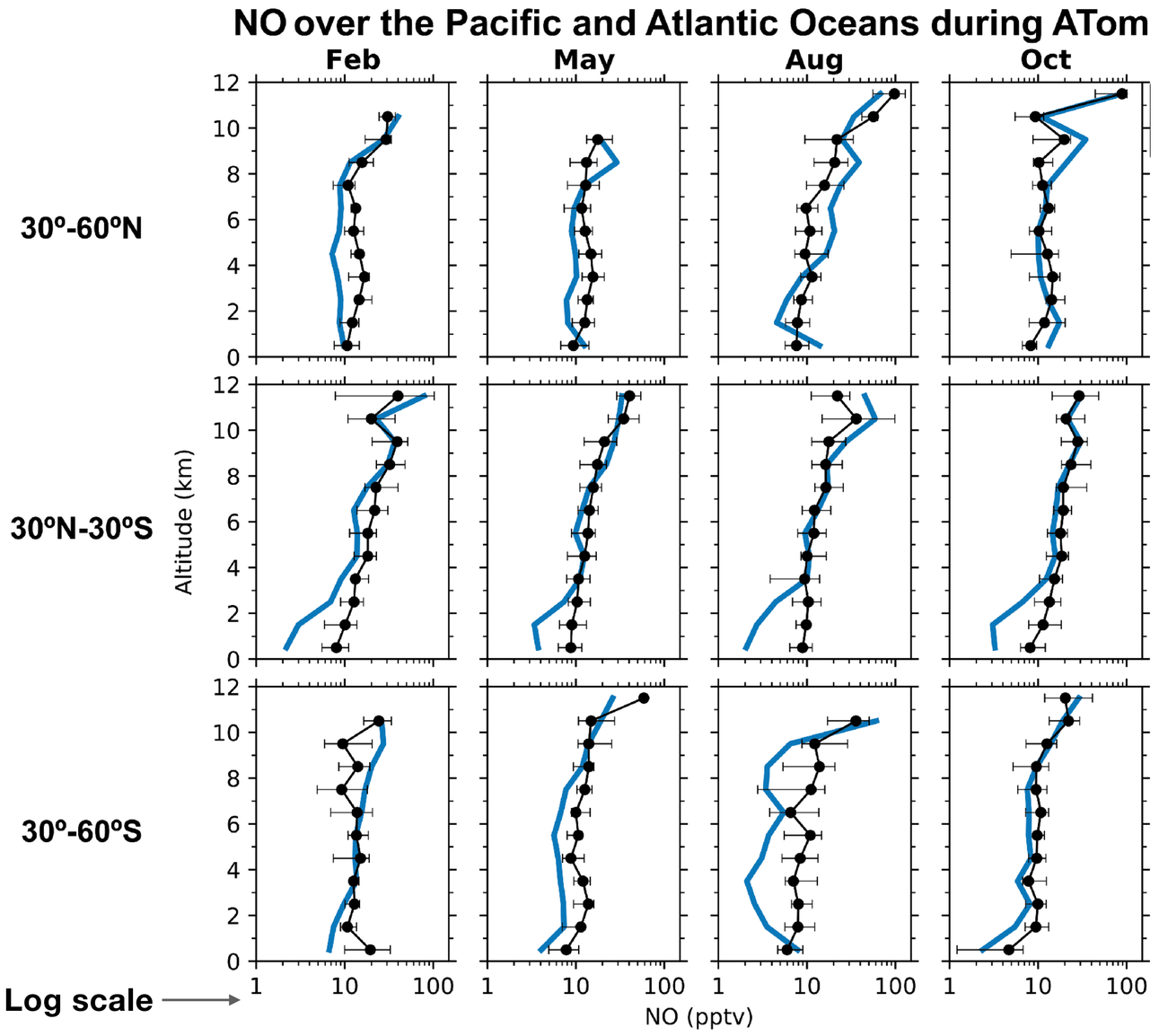
Effect of NO_2 assimilation on FT ozone



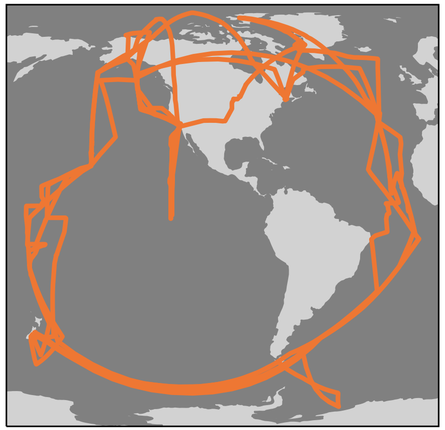
Ozone decreases significantly in SH due to decrease in ozone production over equatorial Africa

Small decreases elsewhere

GEOS-Chem underestimates NO in the remote troposphere



ATom flight tracks



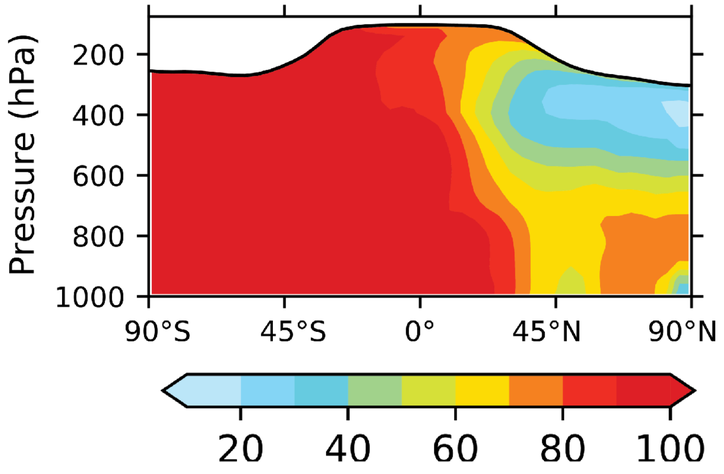
GEOS-Chem OK in the upper troposphere, lightning emissions OK

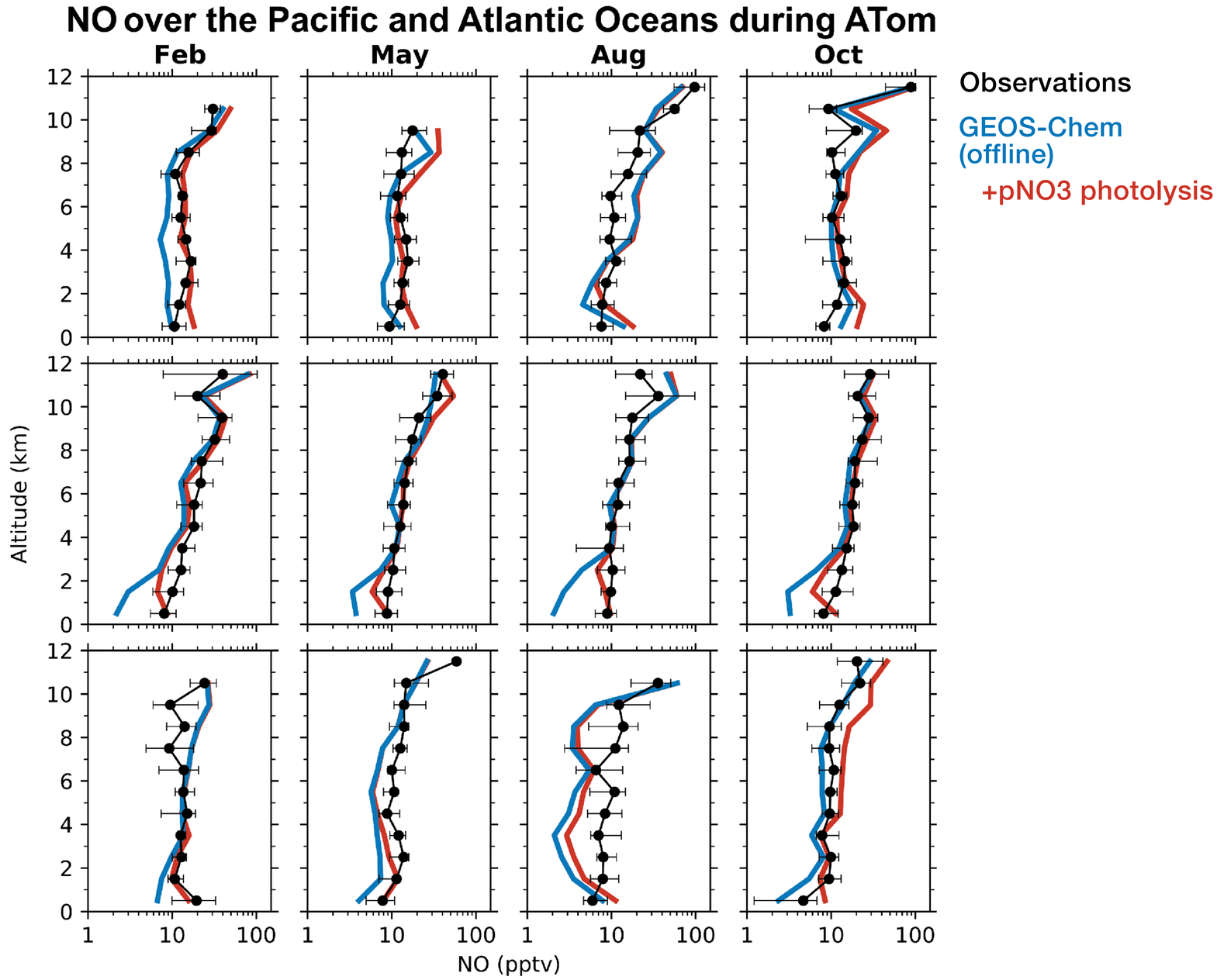
GEOS-Chem HNO_3 and PAN largely consistent with ATom observations

pNO_3 photolysis largely corrects the NO underestimate

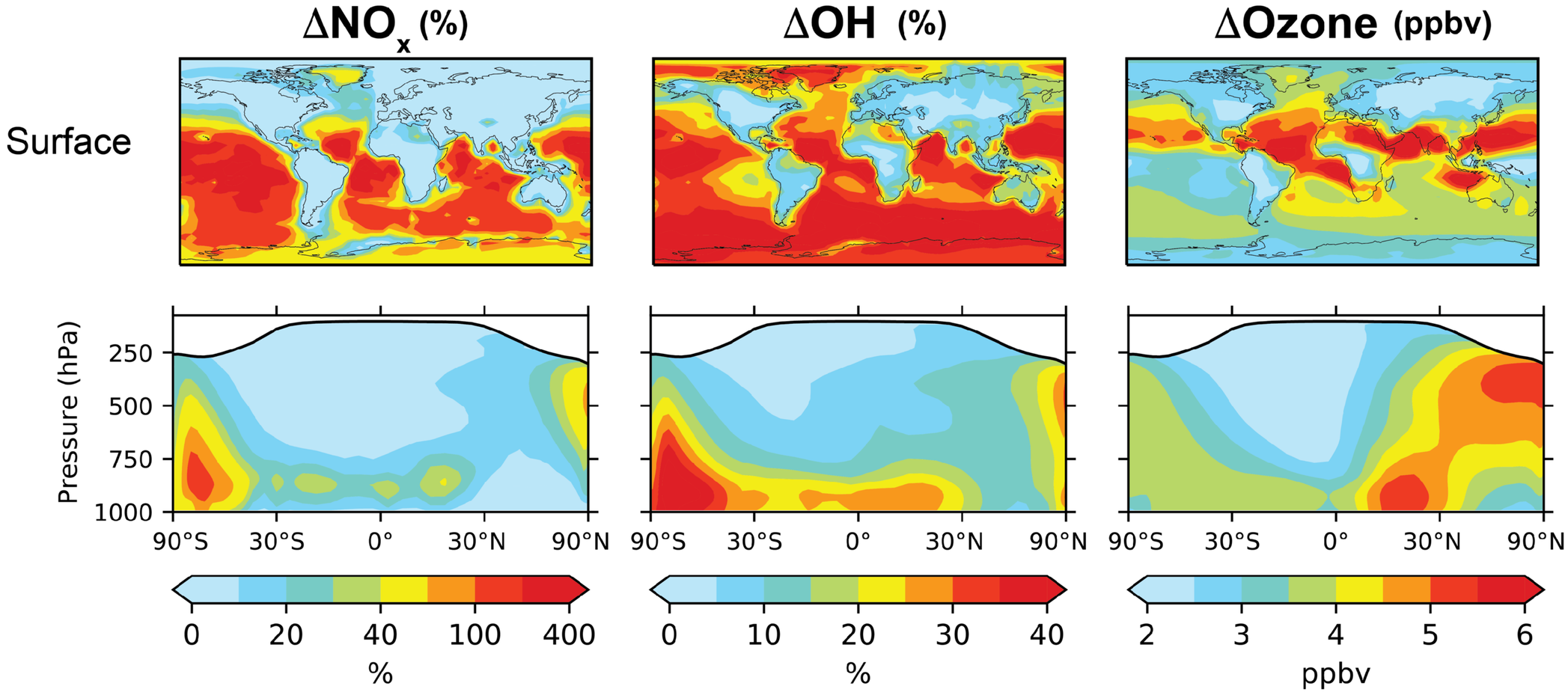
$$pNO_3^- + h\nu \rightarrow HONO + NO_2$$

EF: $j(pNO_3^-) / j(HNO_3)$





pNO_3 photolysis increases global OH and ozone

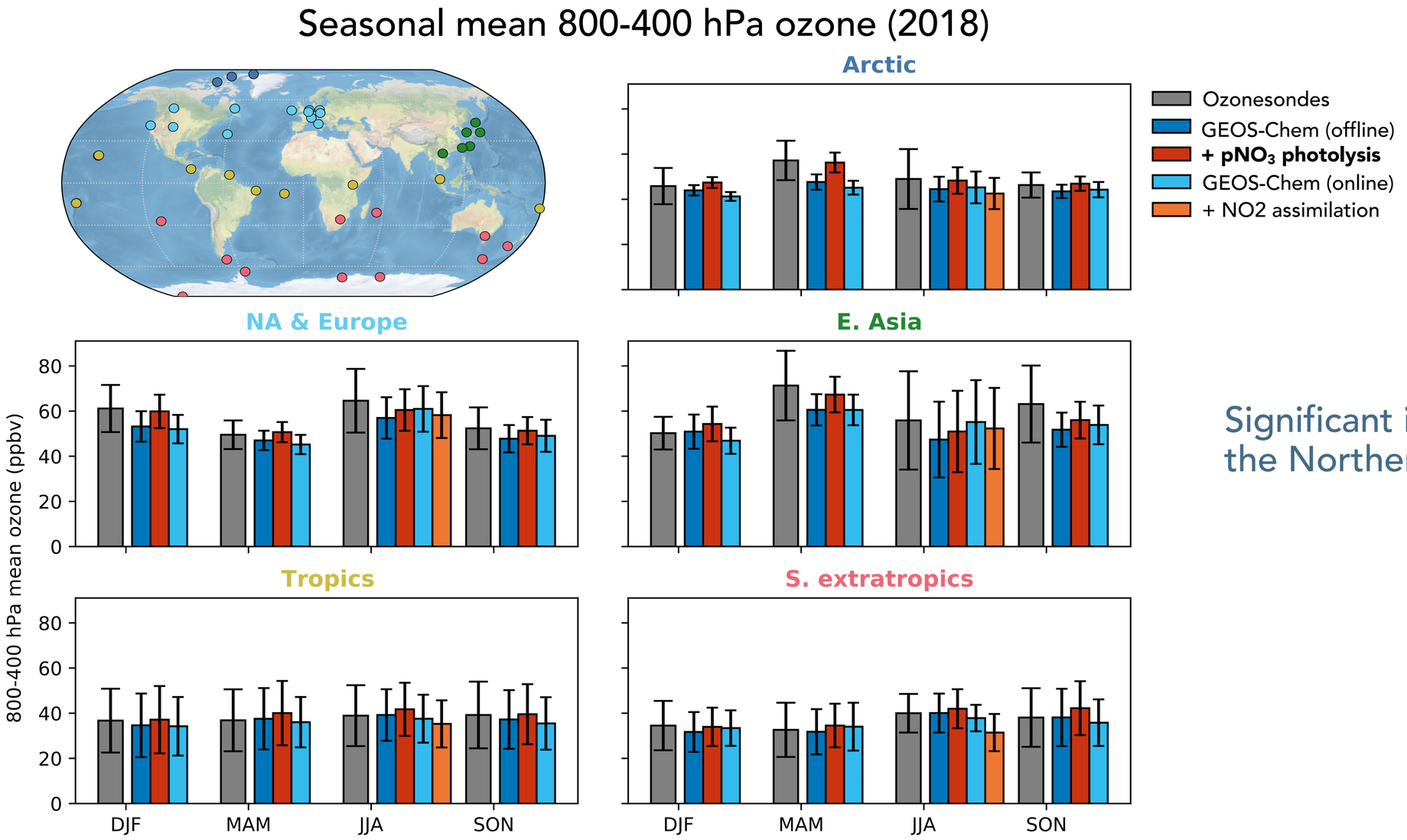


Global increase: 10%; mostly in MBL, smaller increase in northern midlats

Global increase: 19%; due to higher NOx, and more HONO production

Global increase: 10%; ~5ppb increase in FT in northern extratropics; improves simulation compared to obs

Including pNO_3 photolysis improves ozone simulation



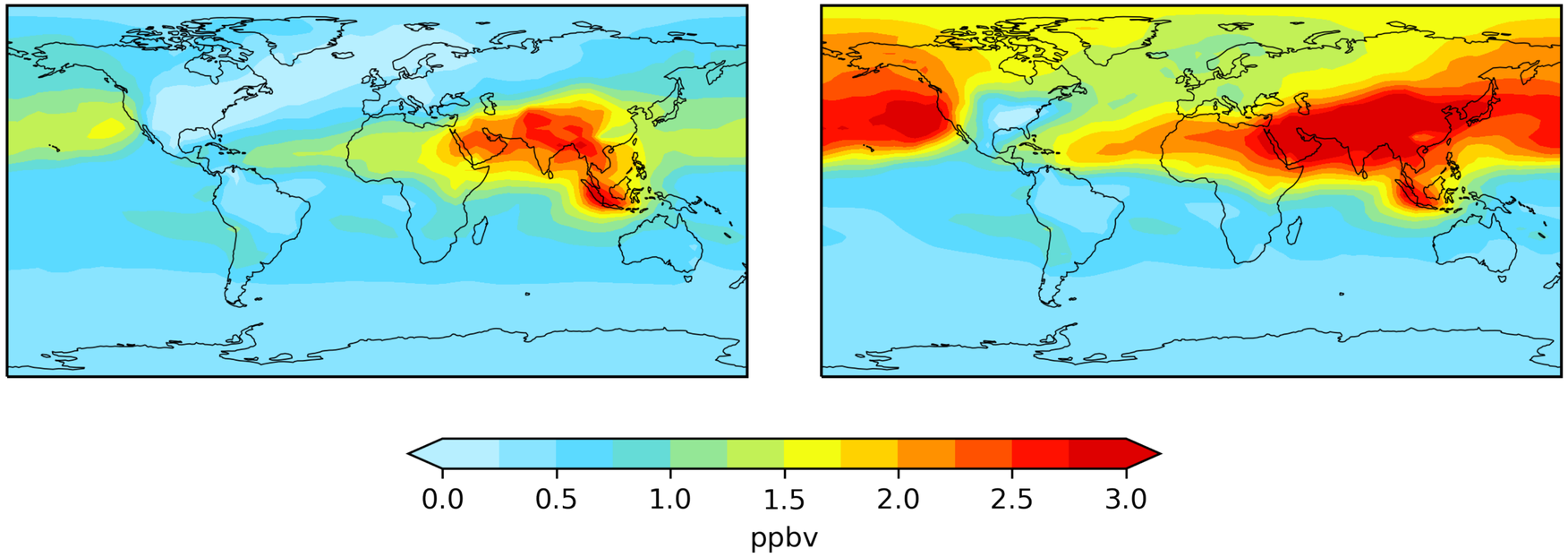
Significant improvement in ozone over the Northern Hemisphere

Including pNO_3 photolysis improves ozone simulation

Emission-driven change in 800-400 hPa ozone from 2008 to 2018

GEOS-Chem (offline)
without pNO_3 photolysis

GEOS-Chem (offline)
with pNO_3 photolysis



Higher sensitivity of FT ozone to emission changes in the NH;
Could explain the observed trends!

Conclusions

A. GEOS-Chem underestimates ozone in the free troposphere; because of an underestimate in free tropospheric NO_2

B. Including pNO_3 photolysis improves the simulation of free tropospheric NO_2

C. pNO_3 photolysis increases concentrations of free tropospheric ozone and its sensitivity to emission changes in the NH