Anthropogenic and meteorological drivers of 2013-2017 variability in summer surface ozone in China

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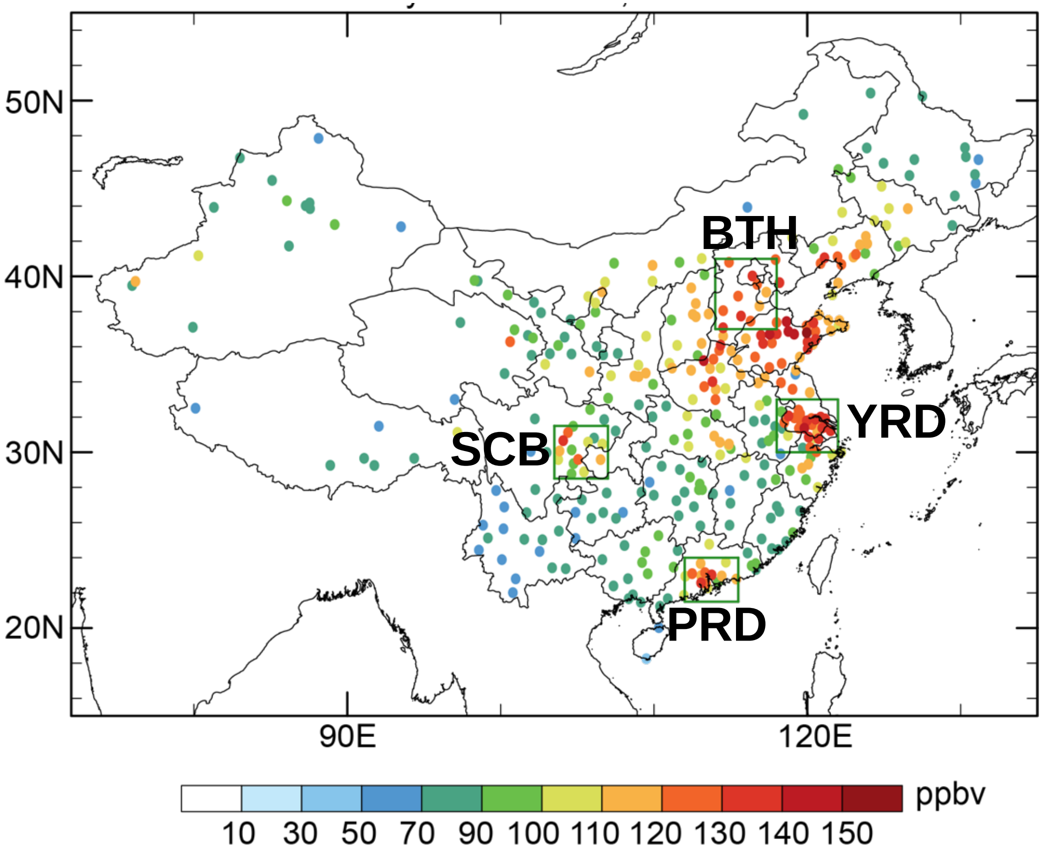
Motivations

Summer maximum daily MDA8 ozone, 2013-2017



Hourly surface O_3 observation network in urban areas in China started from 2013. The network had 450 monitoring stations in 2013 summer, growing to 1500 stations by 2017.

BTH: Beijing-Tianjin-Hebei

YRD: Yangtze River Delta

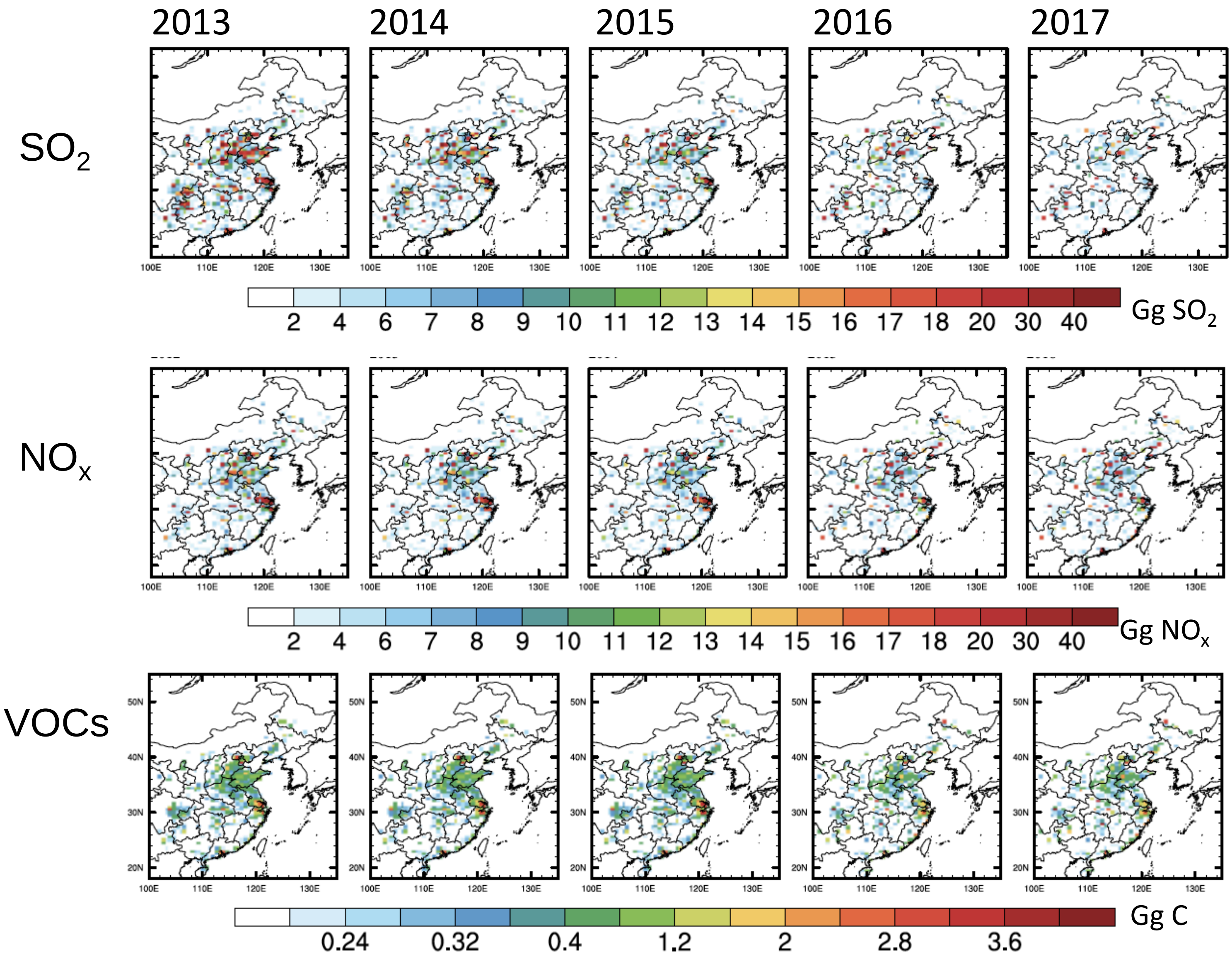
PRD: Pearl River Delta

SCB: Sichuan Basin

MDA8 O_3: maximum daily 8-h average O_3

Motivations

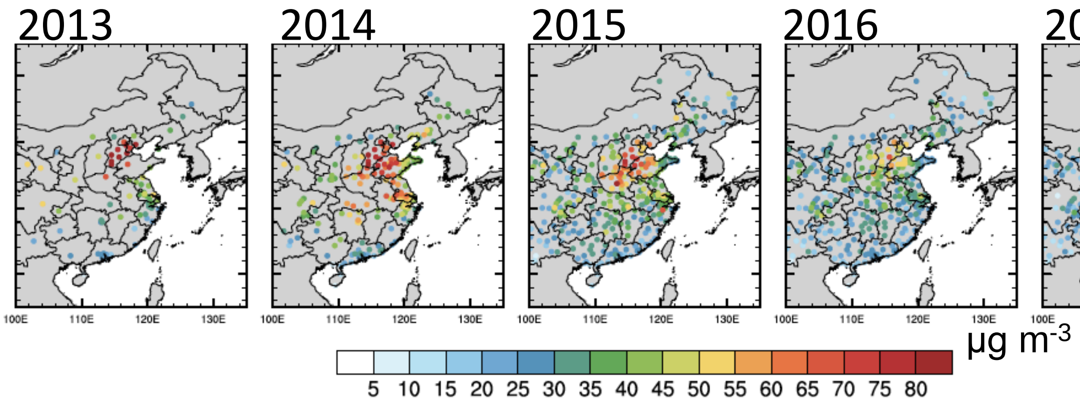
Anthropogenic emissions from MEIC



Zheng et al., 2018

Motivations

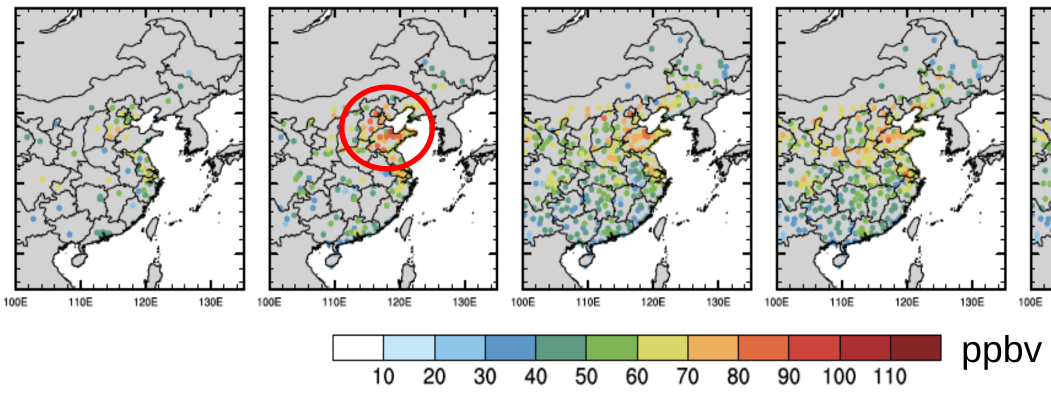
PM$_{2.5}$



Possible reasons:

- The effect of controls on NO$_x$ emissions, which decreased by more than 20% over 2013–2017 (Zheng et al., 2018), under VOC-limited condition
- Decreases in PM$_{2.5}$ could further affect O$_3$ through changes in heterogeneous reactions and photolysis rates (Lou et al., 2014; J. Li et al., 2018)

MDA8 O$_3$



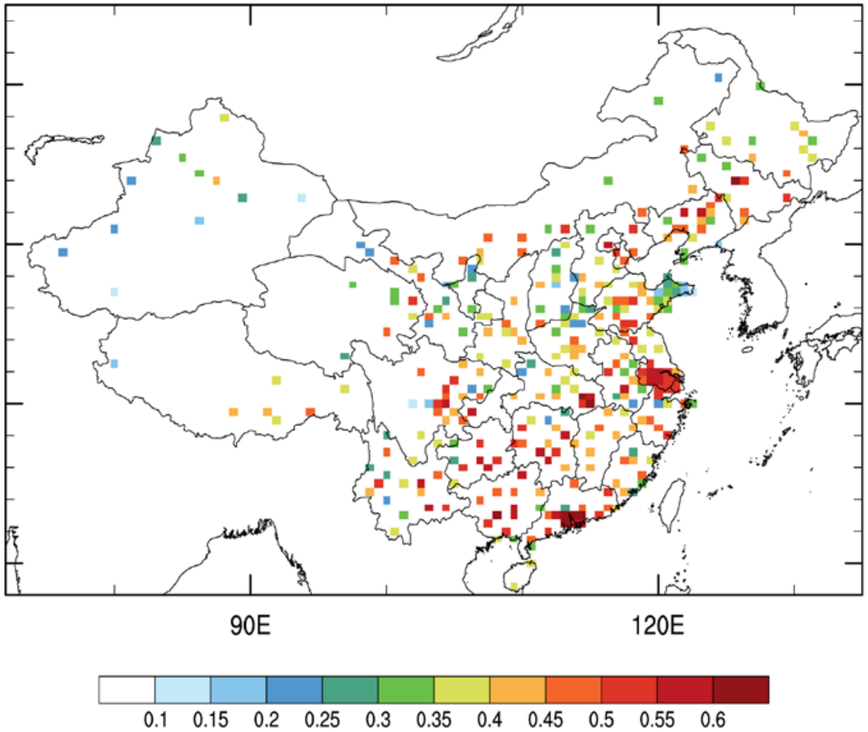
Key meteorological drivers of summer surface O_3

A stepwise multiple linear regression (MLR) model for each 0.5° × 0.625° grid cell:

$$y = \beta_0 + \sum_{k=1}^{9} \beta_k x_k + interaction\ term$$

MERRA2 variables	
Tmax	Daily maximum 2-m air temperature (K)
U10	10-m zonal wind (m s^{-1})
V10	10-m meridional wind (m s^{-1})
PBLH	Boundary layer height over daytime (m)
TCC	Total cloud area fraction over daytime (%)
Rainfall	Total precipitation (mm day^{-1})
SLP	Sea level pressure (Pa)
RH	Surface air relative humidity (100%)
V850	850 hPa meridional wind (m s^{-1})

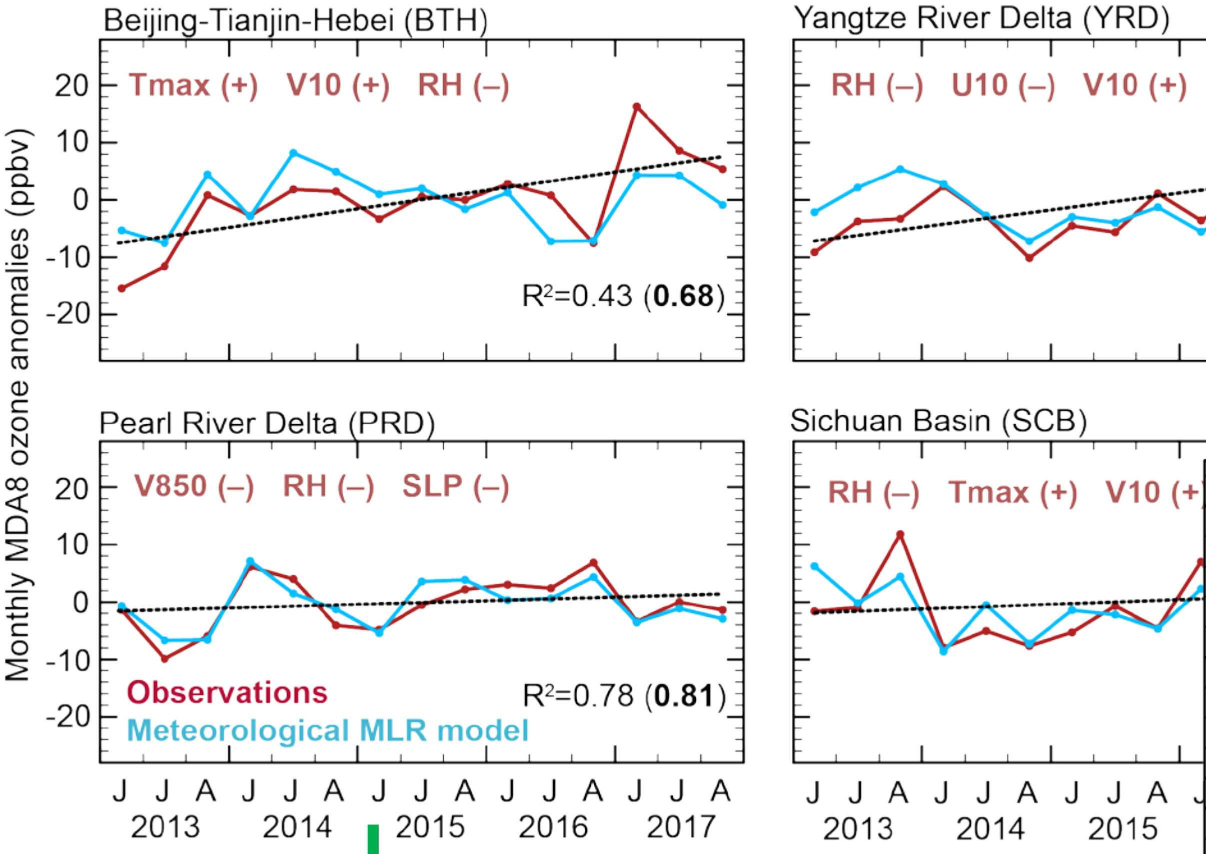
Adjusted coefficients of determination (R^2)



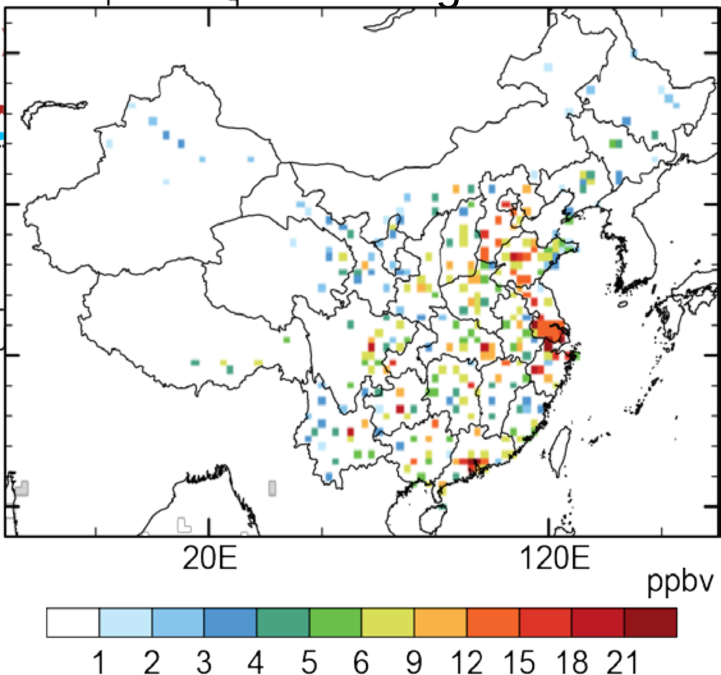
Over **50%** of MDA8 O_3 variability can be explained by meteorological variables in BTH, YRD, PRD, and SCB

Key meteorological drivers of summer surface O_3

Observed and fitted monthly MDA8 O_3 anomalies (ppbv)



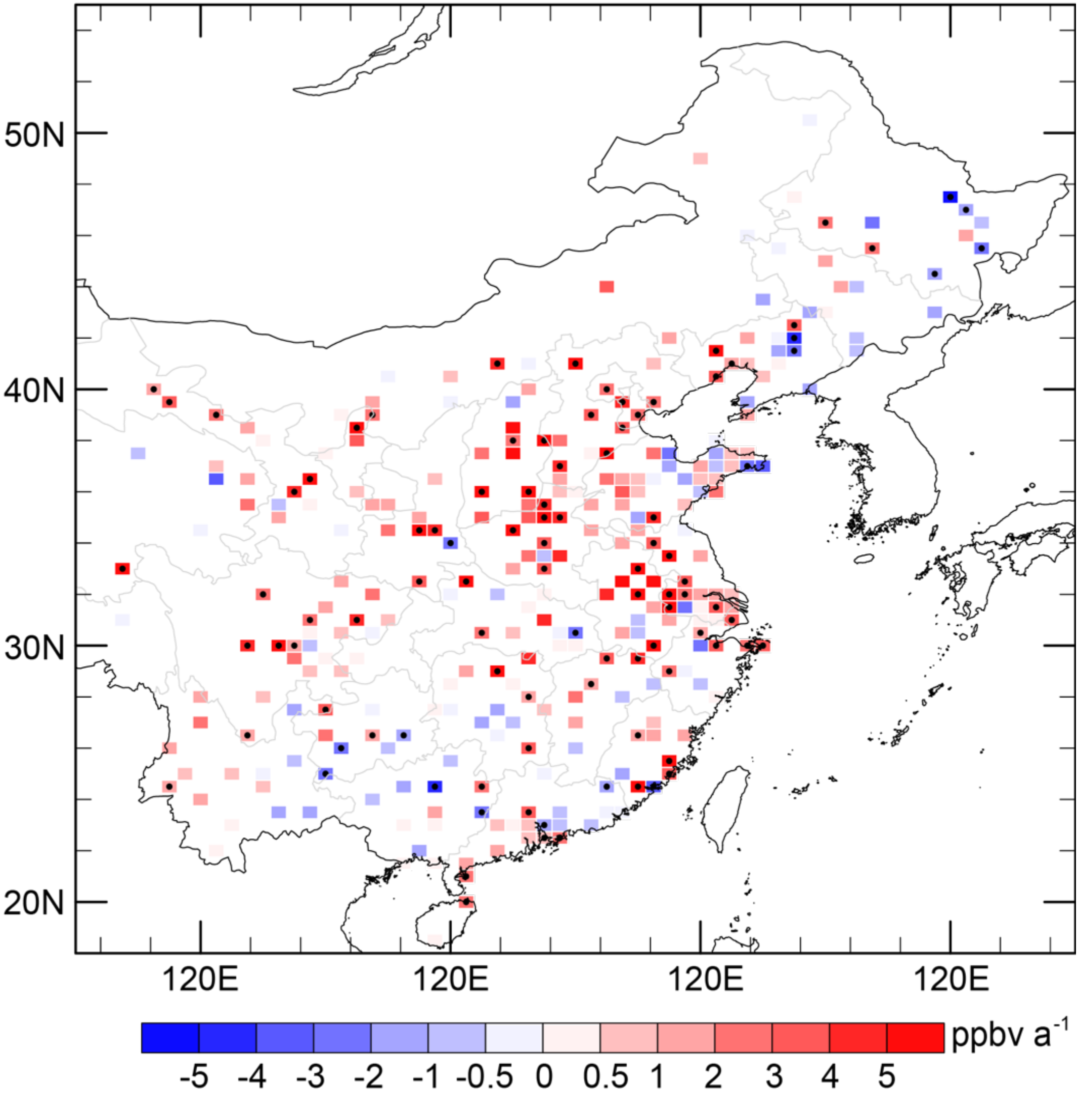
Temperature
surface winds
relative humidity
are the top predictors which can be viewed as general indicators of air stagnation.



O_3 in PRD is closely associated with transboundary transported O_3 driven by summer monsoon (Yang et al., 2014)

Anthropogenic drivers of recent O_3 increasing trend

Residual trend of JJA MDA8 O_3 during 2013-2017

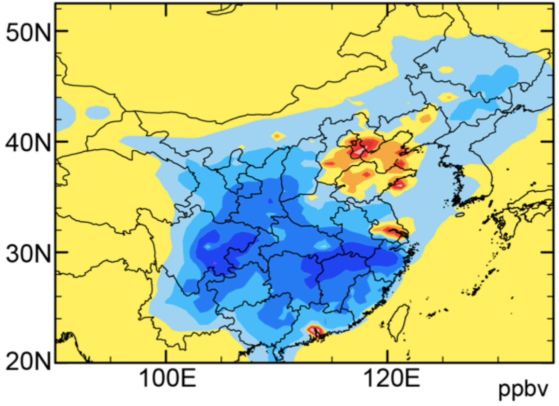


The increasing trend (ppbv a^{-1}) is estimated to be:
2.9 for BTH, **1.4** for YRD,
0.65 for PRD, **0.82** for SCB.
Here we used the median value of O_3 trend from all the sites for each region.

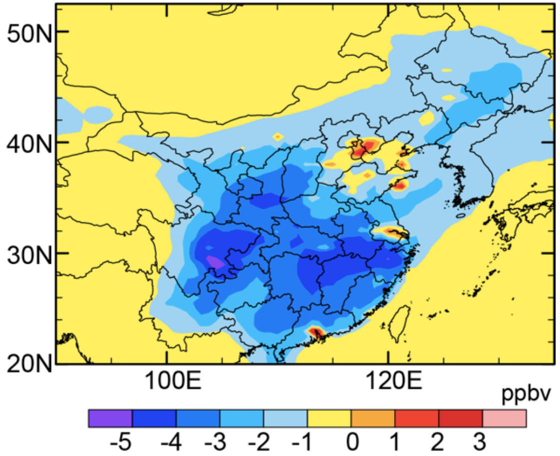
Trend in BTH is significantly larger than the earlier 2003–2015 trend of 1.1 ppbv a^{-1} (Ma et al., 2016)

Anthropogenic drivers of recent O_3 increasing trend

- **Baseline simulation: 2013 MEIC emissions and 2013 MERRA2, nested domain over Asia, v11-02d**
- **25% decrease of NOx emissions and 10% increase of VOCs emissions**
- **40% decrease of aerosol surface area for aerosol chemistry**
- **20% decrease of aerosol optical depth for photolysis rate**

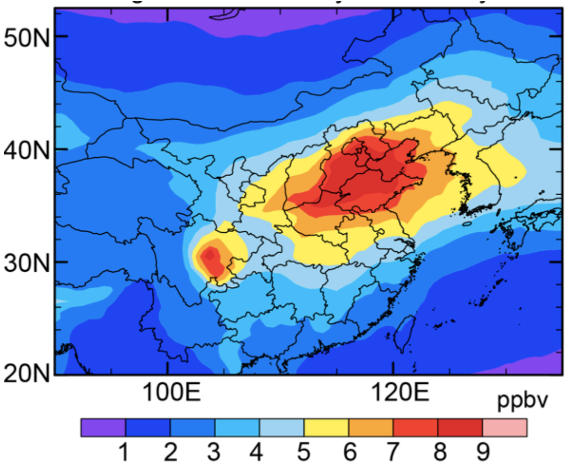


NO_x –25%; VOC +10%

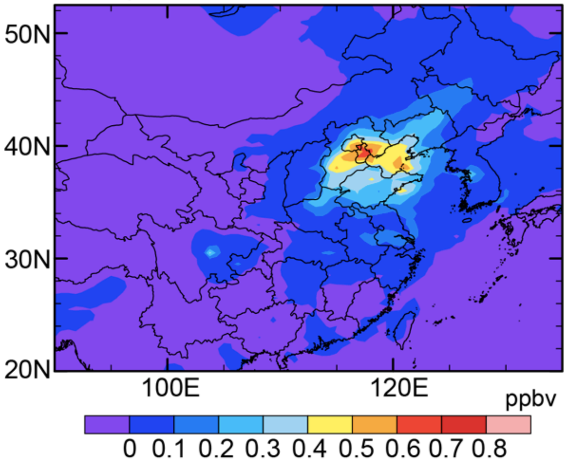


NO_x –25%

Surface O_3 will increase over BTH, YRD, and PRD at the condition of decreased NO_x and increased VOCs and it still happens if only NO_x emissions decrease



Aerosol surface area –40%; AOD –20% (photolysis only)

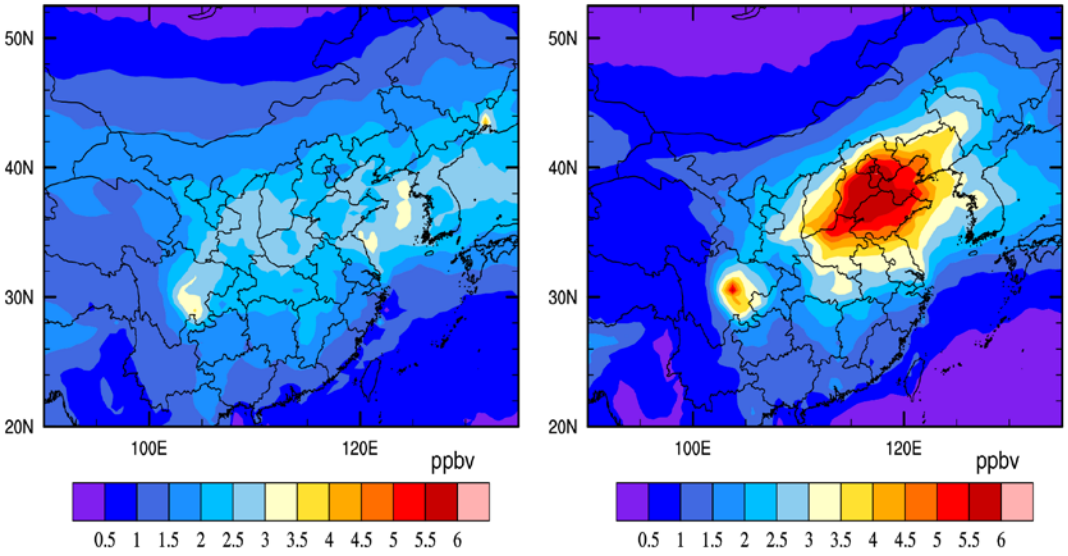


AOD –20% (photolysis only)

A remarkable model result is that changes in $PM_{2.5}$ are more important than changes in NO_x or VOC emissions

Anthropogenic drivers of recent O_3 increasing trend

Simulated effects of individual aerosol chemistry on MDA8 O_3



Uptake of N_2O_5 and NO_2/NO_3

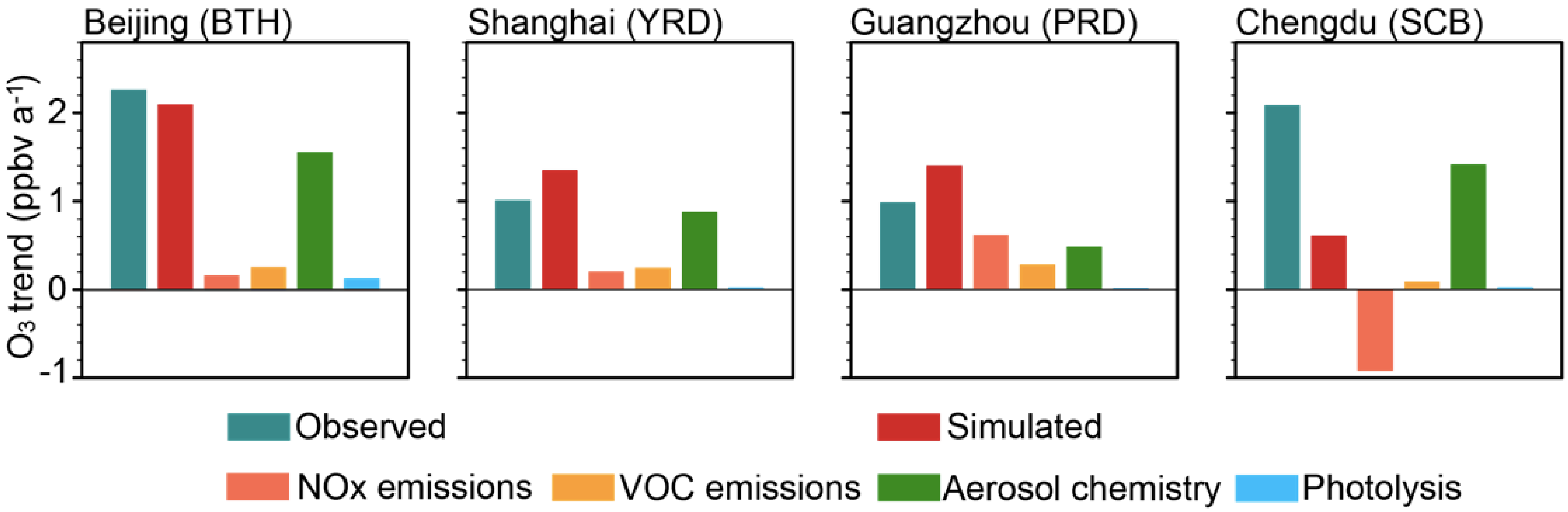
Uptake of HO_2

Uptake of HO_2 is generally the dominant effect

The main heterogeneous reactions responsible for decreasing O_3 in the model are reactive uptake of HO_2 with coefficient $\gamma = 0.2$ (Jacob, 2000) and conversion to H_2O (Mao et al., 2013), and reactive uptake of nitrogen oxides (NO_2, NO_3, N_2O_5) with conversion to HNO_3 (Jacob, 2000; Evans and Jacob, 2005).

Anthropogenic drivers of recent O_3 increasing trend

Observed MDA8 O_3 trend and estimated driving factors in four representative cities



Larger importance of effect of aerosol chemistry on O_3 in BTH and YRD than that in PRD is attributed to their more serious $PM_{2.5}$ pollution

Conclusions

- ❑ Analysis of variability and trends in the nationwide Chinese O_3 record for 2013-2017 shows a 1-3 ppb a^{-1} increase in urban areas across eastern China superimposed on meteorologically driven variability
- ❑ This increase is larger than would be expected from current NO_x emission decreases under VOC-limited conditions
- ❑ Decrease in $PM_{2.5}$ and of the associated sink for HO_x radicals may be more important in driving the O_3 trend
- ❑ There is a need to better understand the effect of aerosol chemistry on O_3 pollution

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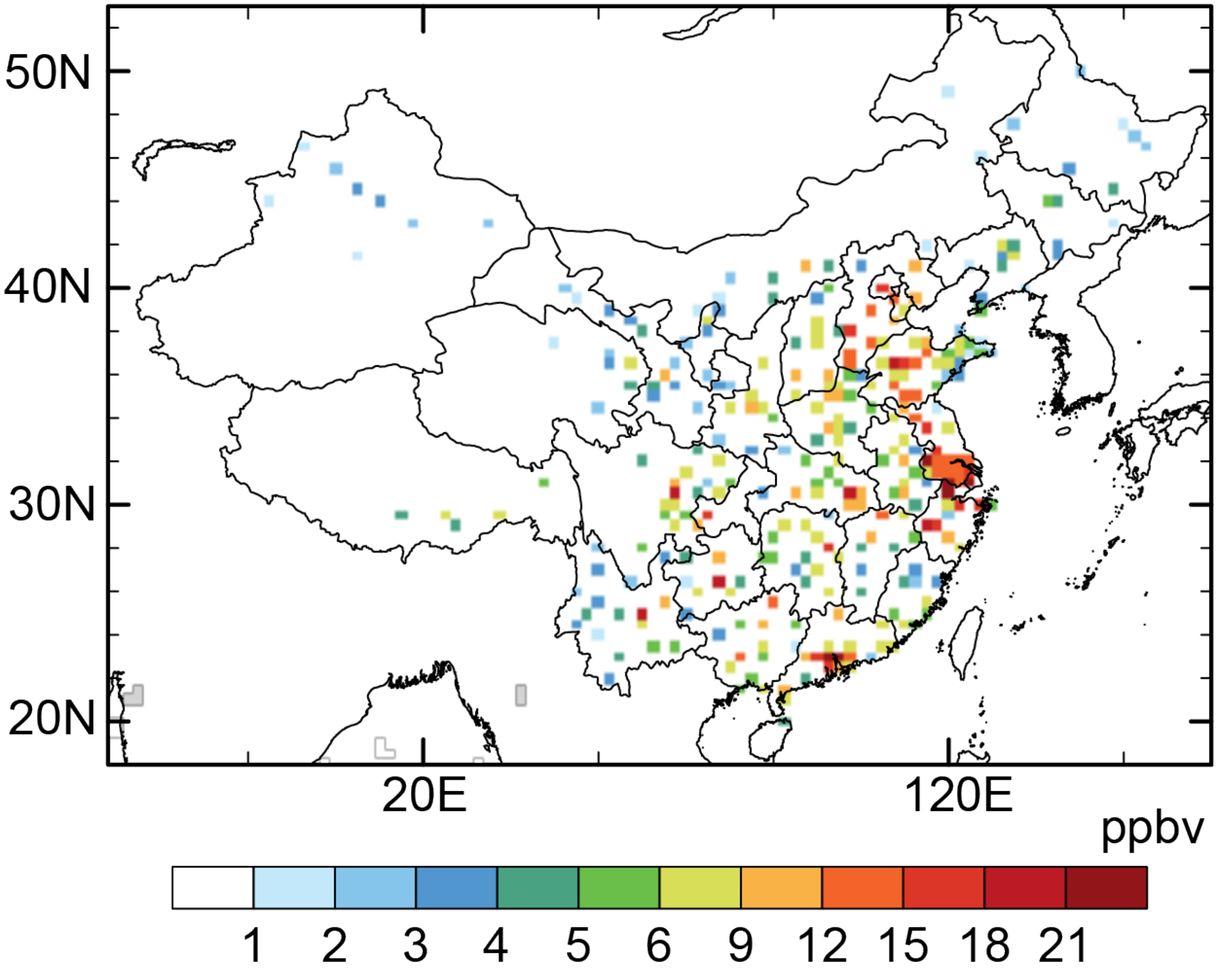
Thanks for your attention !

Sunset@Charles River

Composite MDA8 O_3 between stagnant and non-stagnant days during summer of 2013-2017



The air stagnation is calculated based on MERRA2 data and its definition follows Horton et al. (2014), i.e., a grid cell day is considered stagnant when daily-mean near-surface (10-m) wind speeds are <3.2 m s^{-1}, daily-mean mid-tropospheric (500 hPa) wind speeds are <13 m s^{-1}, and daily-mean precipitation accumulation is < 1mm.

Statistics for observed O_3 trend (ppbv a^{-1}) during 2013-2017

Region	Sample number	Median	Average	Maximum	Minimum
BTH	13	2.87	3.00	5.07	0.19
YRD	22	1.42	2.19	8.30	-2.72
PRD	12	0.65	0.52	3.87	-1.75
SCB	14	0.82	1.42	5.77	-0.56

Table S2. Statistics for observed O_3

-1

Experimental design and GEOS-Chem configuration

Experiments	**NO_x**	**NMVOCs**	**Heterogeneous reactions**	**Photolysis rates**
CTRL	2013	2013	YES	YES
Run_NO_VOC	decrease 25%	increase 10%	same with CTRL	same with CTRL
Run_NO	decrease 25%	same with CTRL	same with CTRL	same with CTRL
Run_Phot	same with CTRL	same with CTRL	same with CTRL	AOD decreases 20%
Run_Hete_Phot	same with CTRL	same with CTRL	aerosol surface area(ASA) decreases 40%	AOD decreases 20%
Run_Hete_N_2O_5	same with CTRL	same with CTRL	ASA decreases 40% only for N_2O_5	AOD decreases 20%
Run_Hete_NOx	same with CTRL	same with CTRL	ASA decreases 40% only for NO_2/NO_3	AOD decreases 20%
Run_Hete_HO_2	same with CTRL	same with CTRL	ASA decreases 40% only for HO_2	AOD decreases 20%

Baseline simulation used anthropogenic emissions in China from the MEIC emissions for 2013 (Zheng et al., 2018), and MIX for other Asian countries (Li et al., 2017) dirven by 2013 MERRA2 meteorology. CTRL and sensitive simulations were performed for the period of 1 June to 31 August of year 2013 after a one-month model spin up.

Observed changes (%) in $PM_{2.5}$ concentration during summer of 2013-2017

Region	**BTH**	**YRD**	**PRD**	**SCB**	**China**
$PM_{2.5}$ changes	51%	34%	15%	49%	50%