Sources of $PM_{2.5}$ during the 2013 winter haze in China constrained by surface observations

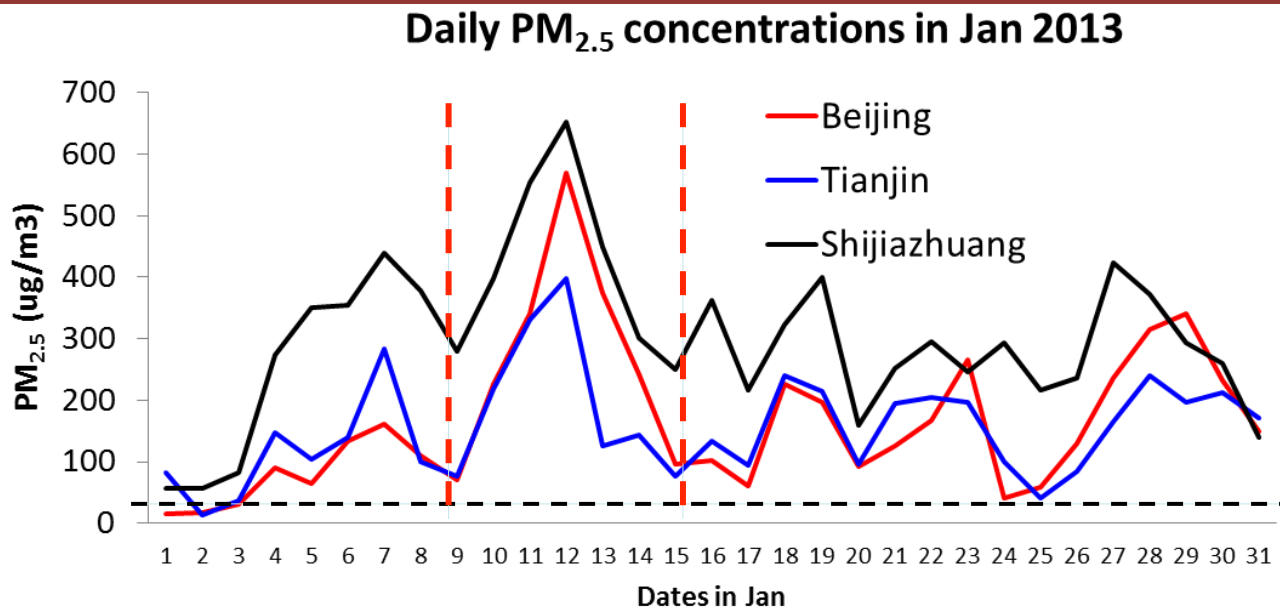
Yuxuan Wang[1,2], Qianqian Zhang[1], Jingkun Jiang[1], Wei Zhou[1], Buying Wang[1], Kebin He[1], Fengkui Duan[1], Qian Zhang[1], Sajeev Philip[3], Yuanyu Xie[1]

[1] Tsinghua University; [2] Texas A&M University; [3] Dalhousie University

Conclusions

- Current models (i.e., GEOS-Chem, CMAQ, WRF-Chem) simulate only up to 50% of observed surface $PM_{2.5}$ levels during winter haze in North China (p.s. observations ranging from 200 ~ 700 ug/m^3)
- Inorganic aerosols (sulfate in particular) are key drivers in observed $PM_{2.5}$ enhancements, but not captured by the GEOS-Chem model
  - SO_2 emissions over North China need to be doubled to match with surface SO_2 observations, while NOx emissions are OK
  - To reproduce sulfate enhancements in haze, heterogeneous uptake of SO_2 on deliquesced aerosols is required as an additional source of sulfate, with RH as a controlling factor
- OC and EC emissions are likely underestimated by a factor of 2 or more

Severe haze in Jan 2013: Surface observations and GEOS-Chem model predictions



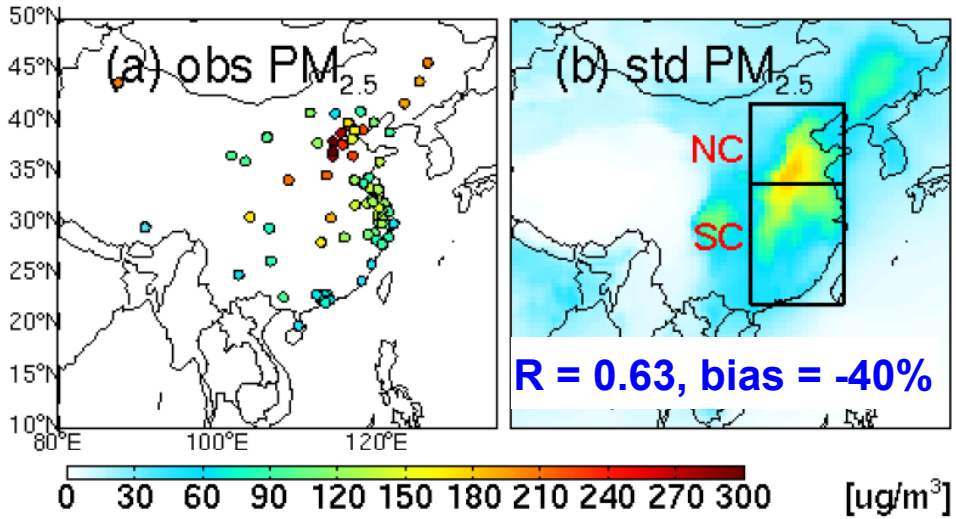
Maximum 24hr-mean was 711 ug/m^3 (13 Jan)

Monthly mean of 74 cities was 130 ug/m^3

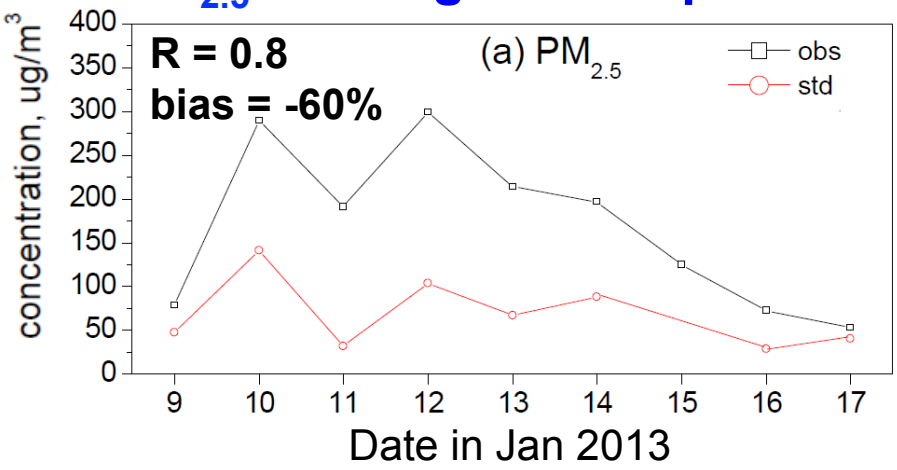
US EPA's 24-hr standard (35 ug/m^3)

Observations

GEOS-Chem model

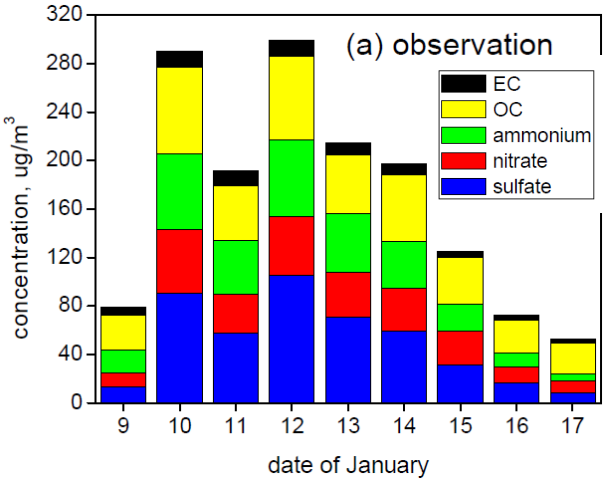


$PM_{2.5}$ at Tsinghua campus site

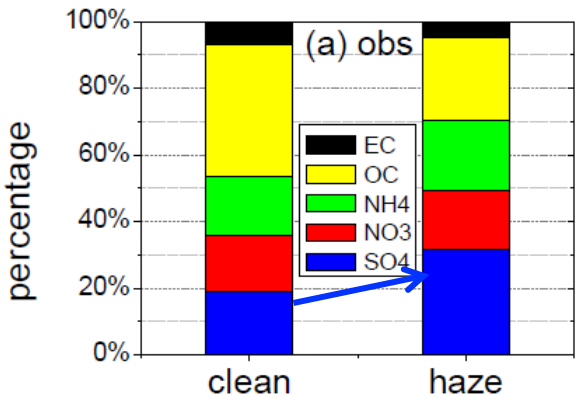


A doubling of SO_2 emissions are needed, but not sufficient to fully fix sulfate problem

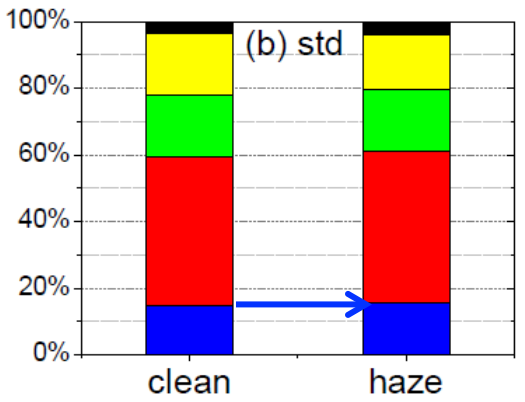
$PM_{2.5}$ chemical compositions at Tsinghua University campus site



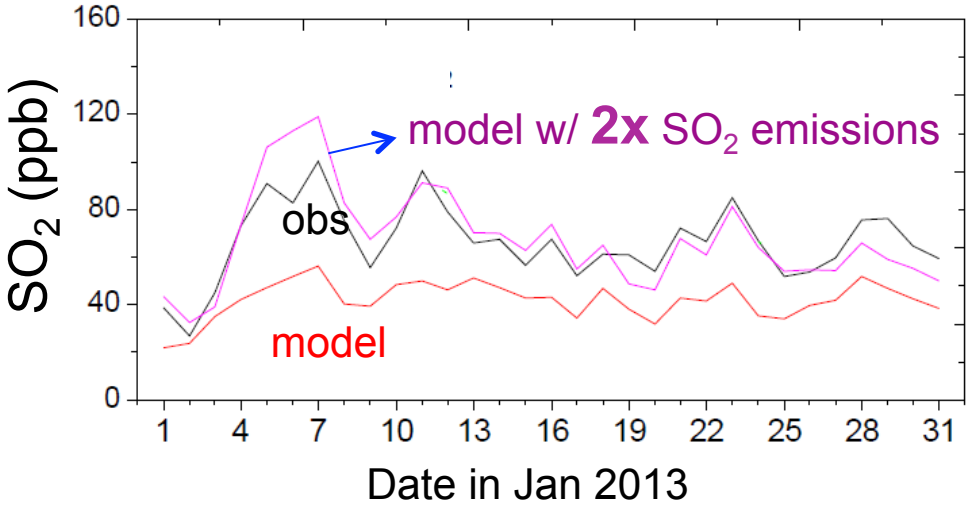
Observations



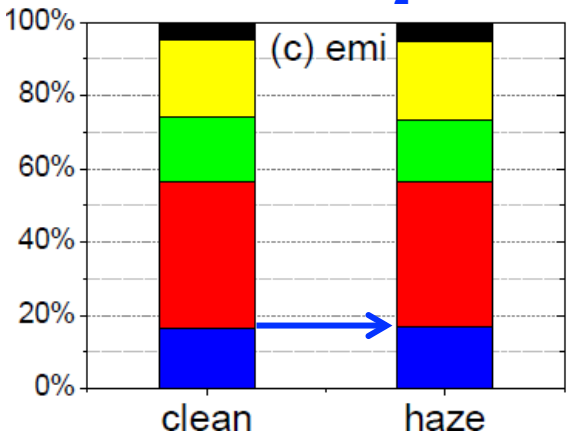
GEOS-Chem model (std)



Increase SO_2 emissions in North China by 100% on average to match with SO_2 observations → sulfate increases by 60%



Model w/ 2X SO_2 emissions



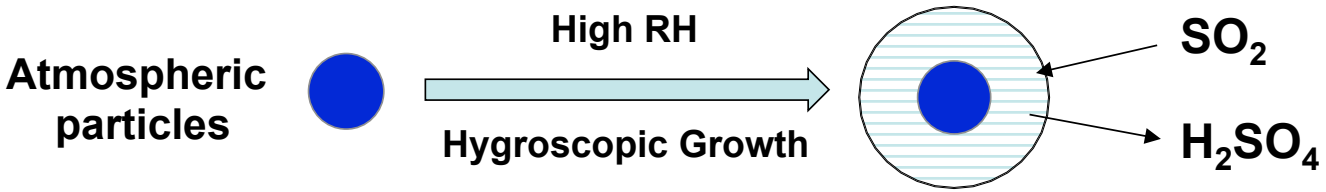
Need additional sulfate formation mechanism during the haze

Sulfate chemistry included in the model

- Gas phase oxidation (SO_2 oxidation by OH) (8%)
- In-cloud aqueous chemistry: SO_2 oxidation by H_2O_2 (71%) , and SO_2 oxidation by O_3 (21%)

Possible oxidation pathways (not included in the model)

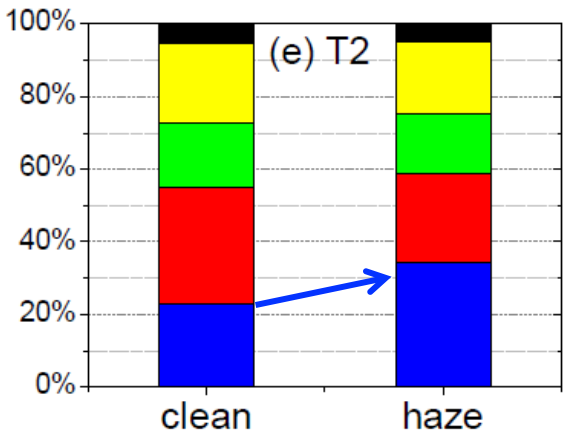
- In-cloud SO_2 oxidation by O_2 catalyzed by Fe(III) and Mn(II) (Cloud fractions or dust are not enhanced during haze)
- **Heterogeneous oxidation of SO_2 on wet aerosols**



$$\frac{d[SO_4^{2-}]}{dt} = (\tfrac{1}{4} A [SO_2] v_{SO_2}) \bullet \gamma$$

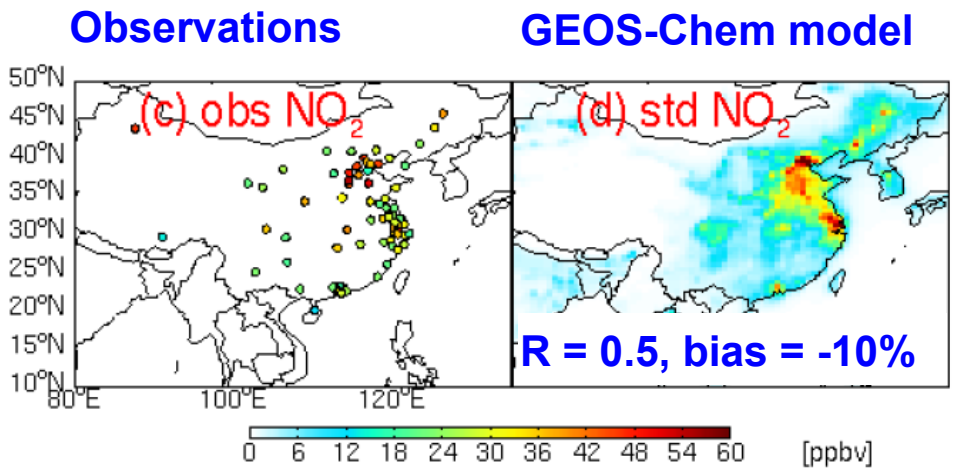
uptake coefficient

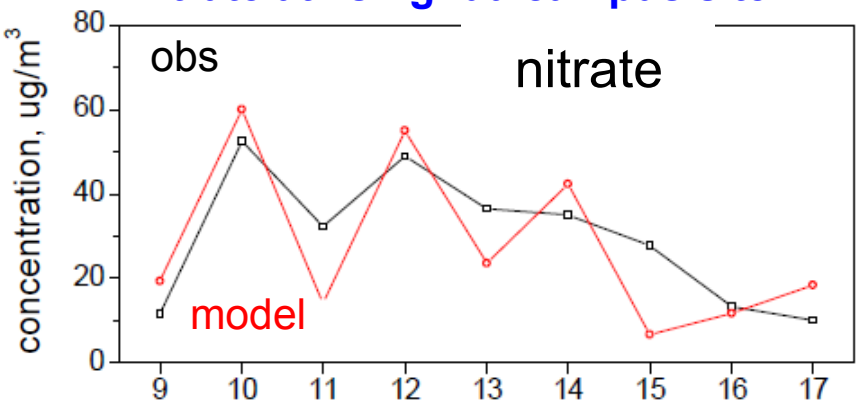
model w/ new oxidation



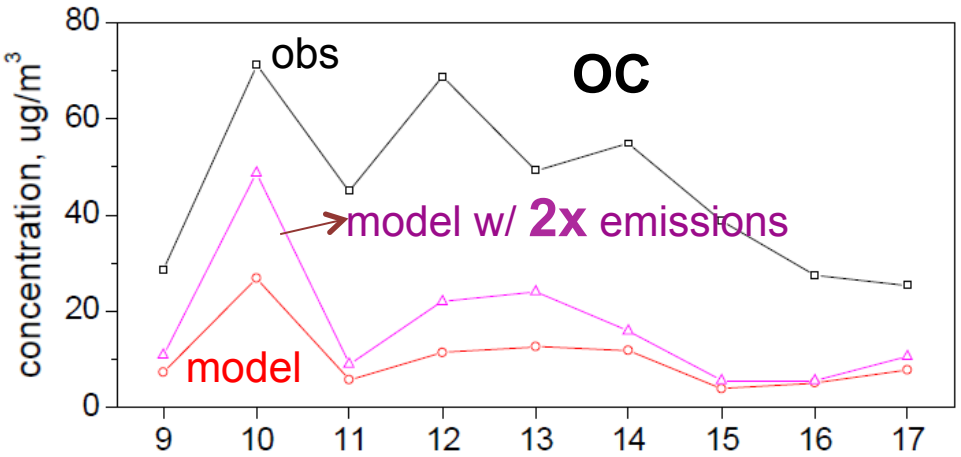
NOx emissions are OK, but OC and EC emissions too low by a factor of 2 or more

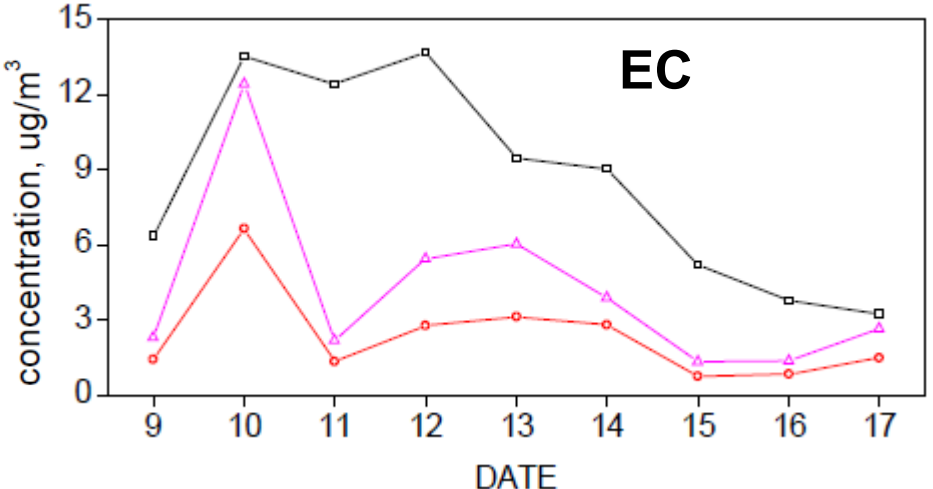
NO_x emissions in North China are consistent with surface observations





Doubling of OC and EC emissions in North China is not enough to reproduce observations at Tsinghua site, but we only have a single site





Conclusions

- GEOS-Chem simulates only up to 50% of observed surface $PM_{2.5}$ levels (130 ~ 700 ug/m^3) during winter haze in North China
- Inorganic aerosols (sulfate in particular) are key drivers in observed $PM_{2.5}$ enhancements, but not captured by the GEOS-Chem model
  - SO_2 emissions over North China need to be doubled to match with surface SO_2 observations, while NOx emissions are OK
  - To reproduce sulfate enhancements in haze, heterogeneous uptake of SO_2 on deliquesced aerosols is required as an additional source of sulfate, with RH as a controlling factor
- OC and EC emissions are likely underestimated by a factor of 2 or more

Reference:
Wang YX, QQ Zhang, JK Jiang, et al., Enhanced sulfate formation during China's severe winter haze episode in Jan 2013 missing from current models, *JGR, under review*