VOCs draft

1. Introduction

(Note: is it needed to address 'Ozone?' maybe we can mention NMVOC more.) Troposhere ozone has been viewed as a critical component in air pollution for being a threat to human body. Recent years, due to active industrial activities, rapid urban construction, dense population, and many other factors, the coastal city clusters have not only become a catalyst for China's economic takeoff, but also brought serious air quality problems, and troposphere ozone pollution is a typical example. This demands for higher accuracy in air quality numerical model prediction. (Note: need much more polish …)

但是，因为源排放的不确定性，参数化方案，气象场与化学场的初始/边界条件误差等问题，许多空气质量模型都不能达到令人满意的预报精度 (warning: suspicious, need accurate cite)。数据同化在 air quality model 中已经有了广泛的应用。Filtering approach Kou et al. (2021), Variational (Elbern et al. 2007; Pagowski et al. 2010; Liu et al. 2011), hybird method (Schwartz et al. 2014). 源排放对空气质量模式的预报精度影响巨大 (Sandu and Chai 2011)，通过同时同化 chem ic and emission，可以更好地的改进模式的预报结果 (Elbern et al. 2007)。top-down 的源排放处理。

当视角转变到 Ozone 预报上，情况将更加复杂，因为控制模式 Ozone 预报的 factor 种类繁多，模式气相化学方案，输运过程，前体物源排放都会对 Ozone 造成影响，其中，源排放对于 Ozone 预报的影响格外明显 (Monks et al. 2015)。城市短期 Ozone 预报的一个重要的不确定性来源就是前体物 (mainly,NOx,CO,NMVOCs) 的源 (Tang et al. 2011; Xiao et al. 2010)。在 Ozone 的前体物中，NMVOCs 具有很大的不确定性，这种不确定性不仅体现在 emission 的总量上，也体现在 NMVOC speciation 上 (Meng Li et al. 2017)。具体来说，模式中不同的气相化学方案都对 VOCs 进行了不同的分类 (lumped together according to their similarities in chemical structure or reactivity), but these speciation almost always differ from inventories, which introduces extra uncertainty. This speciation uncertainty will greatly affect model performance, and many efforts have been devoted to construct a better mapping between inventory and model-ready emission(M. Li et al. 2014). But in data assimilation, uncertainty of NMVOCs speciation haven't got enough attention, previous studies usually treat NMVOCs as a unity and give them equal increments(eg. Tang et al. (2011)). (……) NMVOC is critical for ozone forecasting, and current assimilation method still remains large uncertainty.

目前还没有建立 NMVOCs 的常规业务化观测，并且据 (Koohkan et al. 2013)，VOCs 的观测由于其生命周期很短，限制了同化的空间半径，在使用空间分辨率较大的模式时，即使有稀疏的观测也难以达到理想的效果。这使得我们现阶段只能使用其他的常规观测污染物来尝试同化 VOCs。

2. Model configurations and data assimilation system

Version 3.6.1 of the WRF-Chem model was used, which is an 'online' model with fully-coupled chemical and meteorological fields(Grell et al. 2005). The EnSRF(Whitaker and Hamill 2002) with its expansion to chem component(Schwartz et al. 2014; Zhen Peng et al. 2017) is chosen to construct the data assimilation system. Section 2.1 and 2.2 will expand their settings more specificly.

2.1. WRF-Chem model setting

Table1 for wrf-chem paramenterization setting. See (Z. Peng et al. 2018) for an example. Table2 for RADM2 NMVOCs speciation in model. (14 VOCs we use).

Figure for domain setting and chem observation station distribution.

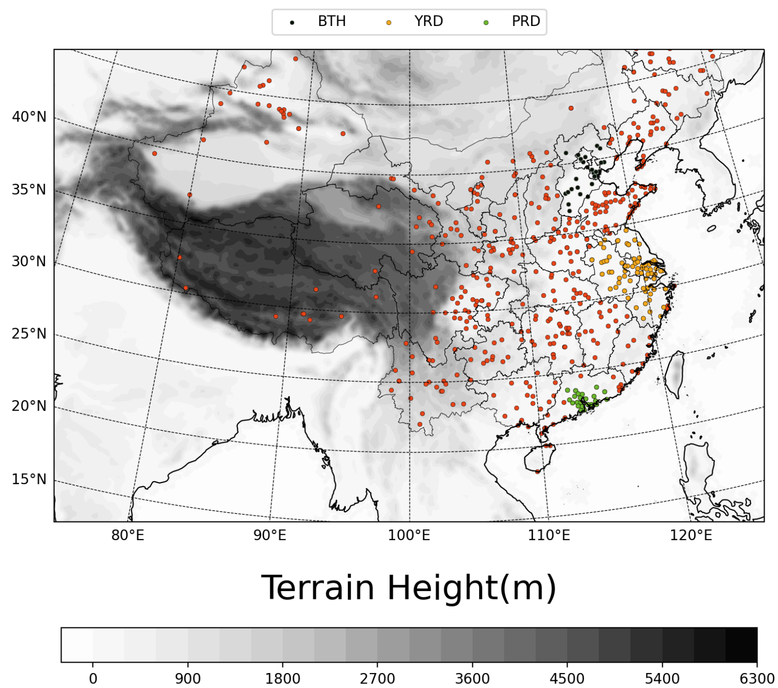

Figure 1: domain_station_fig

The update of source emissions through assimilation of chemical observations follows(Zhen Peng et al. 2017), detailed description will not expand here.

2.2. data assimilation setting

3. Observations

3.1. Meteo

3.2. Chem

3.3. Emission

Mainland China: The hourly prescribed anthropogenic emission is obtained from the Multi-resolution Emission Inventory for China (MEIC v1.3) (Meng Li et al. 2017; Zheng et al. 2018) in July 2017, which is the most updated dataset. In areas out of mainland china, we use EDGARv6.1 in July 2017.

4. Experiment Design

4.1. 30-day free forecast

4.2 VOC Factor

Define:

$$factor = corr * (norm(conc) - norm(emiss))$$

4.3. cycling and forecast setting

We set Meteo Observation assimilate Meteo state variable, six main air pollution's Observation assimilate their own model state variable and emission scaling factor as control experiment(CTRL), which has been proved to be capable of generating relatively good results in analysis and forecast(Z. Peng et al. 2020).

Previous works tend to use ozone observation to assimilate VOCs(Tang et al. 2011; Ma et al. 2019).

	VOC Species
CTRL	None
O3_all	⌢
O3_5pct	⌢
All_5pct	⌢

5. Results

6. Summary and discussion

一些可以用到的结论

1. Ozone 预报的一个重要的不确定性来源就是前体物的源。

   Precursor emissions have been pointed out as the important uncertainty sources of ozone forecast by many previous studies (Carmichael et al., 2008; Constantinescu et al., 2007b; Hanna et al., 1998)

2. 在城市的短期的 Ozone 预报中，前体物的源是除 Ozone IC 之外的最主要的来源 As reported by Tang et al. (2010b), precursor emissions are the most important uncertainty sources for short-term ozone forecast over urban areas beside ozone initial conditions, in contrast to the short-term ozone forecast over suburban areas where the precursor emissions show minor role

3. VOCs 的观测由于其生命周期很短，限制了同化的空间半径。(Koohkan 2013)it is shown that the use of in situ observations using a sparse monitoring network to estimate emissions of isoprene is inadequate because its short chemical lifetime significantly limits the spatial radius of influence of the monitoring data

4. 根据 Li 2014，MEIC 源根据不同的气相化学方案区分了不同的 NMVOCs 源。如果没有对 NMVOCs 进行更加细致的同化操作，实际上是浪费了这些信息。

5. Sandu 2011 Elbern2007 都在说 emission 是 control variable，这对 Ozone 也一样。

Elbern, H., A. Strunk, H. Schmidt, and O. Talagrand. 2007. "Emission Rate and Chemical State Estimation by 4-Dimensional Variational Inversion." Journal Article. *Atmospheric Chemistry and Physics* 7 (14): 3749–69. https://doi.org/DOI%2010.5194/acp-7-3749-2007.

Grell, G. A., S. E. Peckham, R. Schmitz, S. A. McKeen, G. Frost, W. C. Skamarock, and B. Eder. 2005. "Fully Coupled "online" Chemistry Within the WRF Model." Journal Article. *Atmospheric Environment* 39 (37): 6957–75. https://doi.org/10.1016/j.atmosenv.2005.04.027.

Koohkan, M. R., M. Bocquet, Y. Roustan, Y. Kim, and C. Seigneur. 2013. "Estimation of Volatile Organic Compound Emissions for Europe Using Data Assimilation." Journal Article. *Atmospheric Chemistry and Physics* 13 (12): 5887–5905. https://doi.org/10.5194/acp-13-5887-2013.

Kou, Xingxia, Zhen Peng, Meigen Zhang, Ning Zhang, Lili Lei, Xiujuan Zhao, Shiguang Miao, Ziming Li, and Qiuji Ding. 2021. "Assessment of the Meteorological Impact on Improved PM 2.5 Air Quality over North China During 2016–2019 Based on a Regional Joint Atmospheric Composition Reanalysis Data-set." Journal Article. *Journal of Geophysical Research: Atmospheres* 126 (11). https://doi.org/10.1029/2020jd034382.

Li, M., Q. Zhang, D. G. Streets, K. B. He, Y. F. Cheng, L. K. Emmons, H. Huo, et al. 2014. "Mapping Asian Anthropogenic Emissions of Non-Methane Volatile Organic Compounds to Multiple Chemical Mechanisms." Journal Article. *Atmospheric Chemistry and Physics* 14 (11): 5617–38. https://doi.org/10.5194/acp-14-5617-2014.

Li, Meng, Huan Liu, Guannan Geng, Chaopeng Hong, Fei Liu, Yu Song, Dan Tong, et al. 2017. "Anthropogenic Emission Inventories in China: A Review." Journal Article. *National Science Review* 4 (6): 834–66. https://doi.org/10.1093/nsr/nwx150.

Liu, Z. Q., Q. H. Liu, H. C. Lin, C. S. Schwartz, Y. H. Lee, and T. J. Wang. 2011. "Three-Dimensional Variational Assimilation of MODIS Aerosol Optical Depth: Implementation and Application to a Dust Storm over East Asia." Journal Article. *Journal of Geophysical Research-Atmospheres* 116 (D23): n/a–. https://doi.org/Artn%20D23206%2010.1029/2011jd016159.

Ma, C. Q., T. J. Wang, A. P. Mizzi, J. L. Anderson, B. L. Zhuang, M. Xie, and R. S. Wu. 2019. "Multiconstituent Data Assimilation with WRF-Chem/DART: Potential for Adjusting Anthropogenic Emissions and Improving Air Quality Forecasts over Eastern China." Journal Article. *Journal of Geophysical Research-Atmospheres* 124 (13): 7393–7412. https://doi.org/10.1029/2019jd030421.

Monks, P. S., A. T. Archibald, A. Colette, O. Cooper, M. Coyle, R. Derwent, D. Fowler, et al. 2015. "Tropospheric Ozone and Its Precursors from the Urban to the Global Scale from Air Quality to Short-Lived Climate Forcer." Journal Article. *Atmospheric Chemistry and Physics* 15 (15): 8889–8973. https://doi.org/10.5194/acp-15-8889-2015.

Pagowski, M., and G. A. Grell. 2012. "Experiments with the Assimilation of Fine Aerosols Using an Ensemble Kalman Filter." Journal Article. *Journal of Geophysical Research-Atmospheres* 117 (D21): n/a–. https://doi.org/Artn%20D21302%2010.1029/2012jd018333.

Pagowski, M., G. A. Grell, S. A. McKeen, S. E. Peckham, and D. Devenyi. 2010. "Three-Dimensional Variational Data Assimilation of Ozone and Fine Particulate Matter Observations: Some Results Using the Weather Research and Forecasting - Chemistry Model and Grid-Point Statistical Interpolation." Journal Article. *Quarterly Journal of the Royal Meteorological Society* 136 (653): 2013–24. https://doi.org/10.1002/qj.700.

Peng, Z., L. L. Lei, Z. Q. Liu, H. N. Liu, K. K. Chu, and X. X. Kou. 2020. "Impact of Assimilating Meteorological Observations on Source Emissions Estimate and Chemical Simulations." Journal Article. *Geophysical Research Letters* 47 (20). https://doi.org/ARTN%20e2020GL089030%2010.1029/2020GL089030.

Peng, Z., L. L. Lei, Z. Q. Liu, J. N. Su, A. J. Ding, J. M. Ban, D. Chen, X. X. Kou, and K. K. Chu. 2018. "The Impact of Multi-Species Surface Chemical Observation Assimilation on Air Quality Forecasts in China." Journal Article. *Atmospheric Chemistry and Physics* 18 (23): 17387–404. https://doi.org/10.5194/acp-18-17387-2018.

Peng, Zhen, Zhiquan Liu, Dan Chen, and Junmei Ban. 2017. "Improving PM_{2. 5} Forecast over China by the Joint Adjustment of Initial Conditions and Source Emissions with an Ensemble Kalman Filter."

Journal Article. *Atmospheric Chemistry and Physics* 17 (7): 4837–55. https://doi.org/10.5194/acp-17-4837-2017.

Sandu, A., and T. F. Chai. 2011. "Chemical Data Assimilation-an Overview." Journal Article. *Atmosphere* 2 (3): 426–63. https://doi.org/10.3390/atmos2030426.

Schwartz, C. S., Z. Q. Liu, H. C. Lin, and J. D. Cetola. 2014. "Assimilating Aerosol Observations with a "hybrid" Variational-Ensemble Data Assimilation System." Journal Article. *Journal of Geophysical Research-Atmospheres* 119 (7): 4043–69. https://doi.org/10.1002/2013jd020937.

Schwartz, C. S., Z. Q. Liu, H. C. Lin, and S. A. McKeen. 2012. "Simultaneous Three-Dimensional Variational Assimilation of Surface Fine Particulate Matter and MODIS Aerosol Optical Depth." Journal Article. *Journal of Geophysical Research-Atmospheres* 117 (D13): n/a–. https://doi.org/Artn%20D13202%2010.1029/2011jd017383.

Tang, X., J. Zhu, Z. F. Wang, and A. Gbaguidi. 2011. "Improvement of Ozone Forecast over Beijing Based on Ensemble Kalman Filter with Simultaneous Adjustment of Initial Conditions and Emissions." Journal Article. *Atmospheric Chemistry and Physics* 11 (24): 12901–16. https://doi.org/10.5194/acp-11-12901-2011.

Whitaker, J. S., and T. M. Hamill. 2002. "Ensemble Data Assimilation Without Perturbed Observations." Journal Article. *Monthly Weather Review* 130 (7): 1913–24. https://doi.org/Doi%2010.1175/1520-0493(2002)130%3C1913:Edawpo%3E2.0.Co;2.

Xiao, Tang, Wang Zifa, Z. H. U. Jiang, W. U. Qizhong, and Gbaguidi Alex. 2010. "Preliminary Application of Monte Carlo Uncertainty Analysis in O3 Simulation." Journal Article. *Climatic and Environmental Research* 15 (5): 541–50. qhhjen/article/abstract/20100502.

Zheng, Bo, Dan Tong, Meng Li, Fei Liu, Chaopeng Hong, Guannan Geng, Haiyan Li, et al. 2018. "Trends in China's Anthropogenic Emissions Since 2010 as the Consequence of Clean Air Actions." Journal Article. *Atmospheric Chemistry and Physics* 18 (19): 14095–111. https://doi.org/10.5194/acp-18-14095-2018.