CUMULUS-PARAMETERIZED CONVECTIVE TRANSPORT AND WET SCAVENGING OF TRACE GASES

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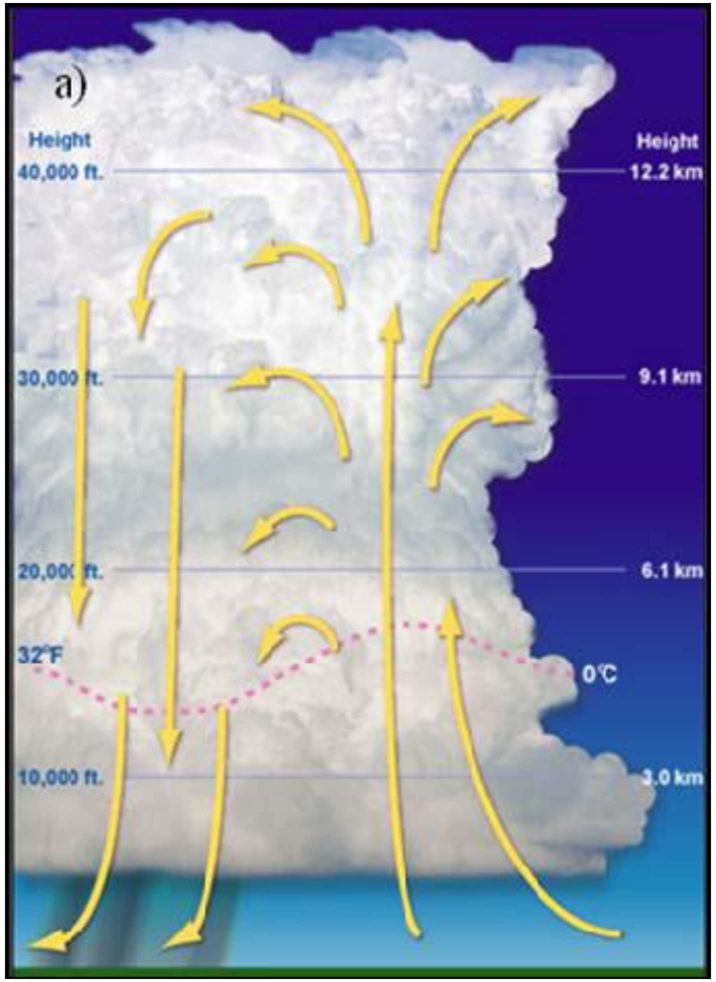
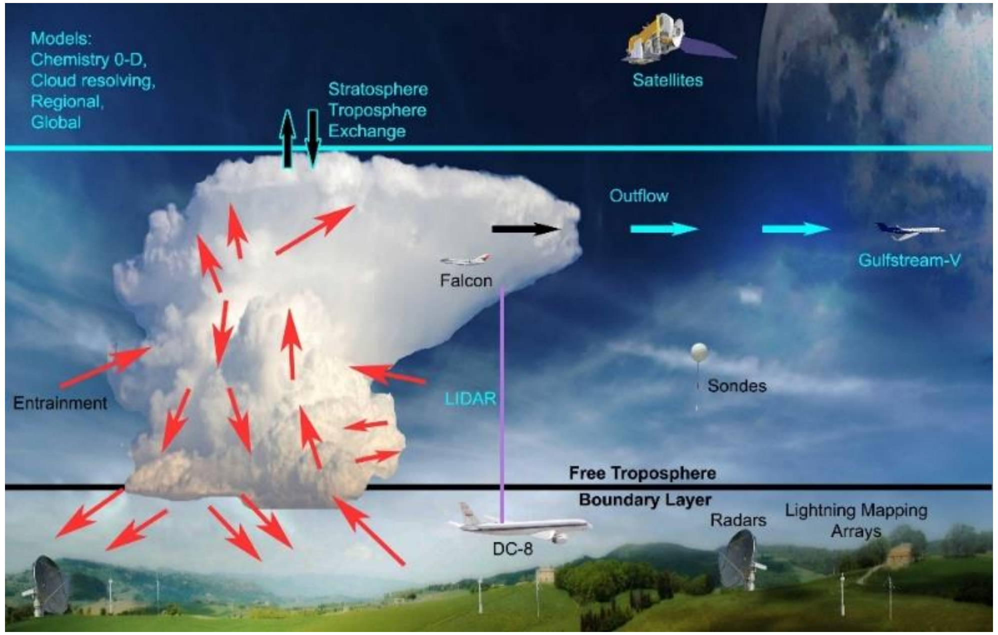
DEEP CONVECTIVE TRANSPORT OF TRACE GASES

- Deep convection can transport **surface moisture and pollution** from PBL to the upper troposphere (UT) **within a few minutes.**
- Vertical transport of local air pollutants
  - Transforms local air pollution problems into regional or global atmospheric chemistry problems.
- Vertical transport of O_3 and O_3 precursors, surface moisture, and aerosol
  - Affects the Earth's radiation budget and climate.
- Needs to be parameterized in regional and global models



DC3 FIELD CAMPAIGN

- Storms discussed in this study were observed during the 2012 Deep Convective Clouds and Chemistry (DC3) field campaign (May15 - June 30, 2012).
- Location:
  - Northeastern Colorado
  - Central Oklahoma to west Texas
  - Northern Alabama
- It made use of various types of measurements
  - Aircraft Measurements: DC-8, GV and Falcon aircraft
  - Doppler radar
  - Upper air soundings
  - Lightning mapping arrays (LMA)
  - Satellite data

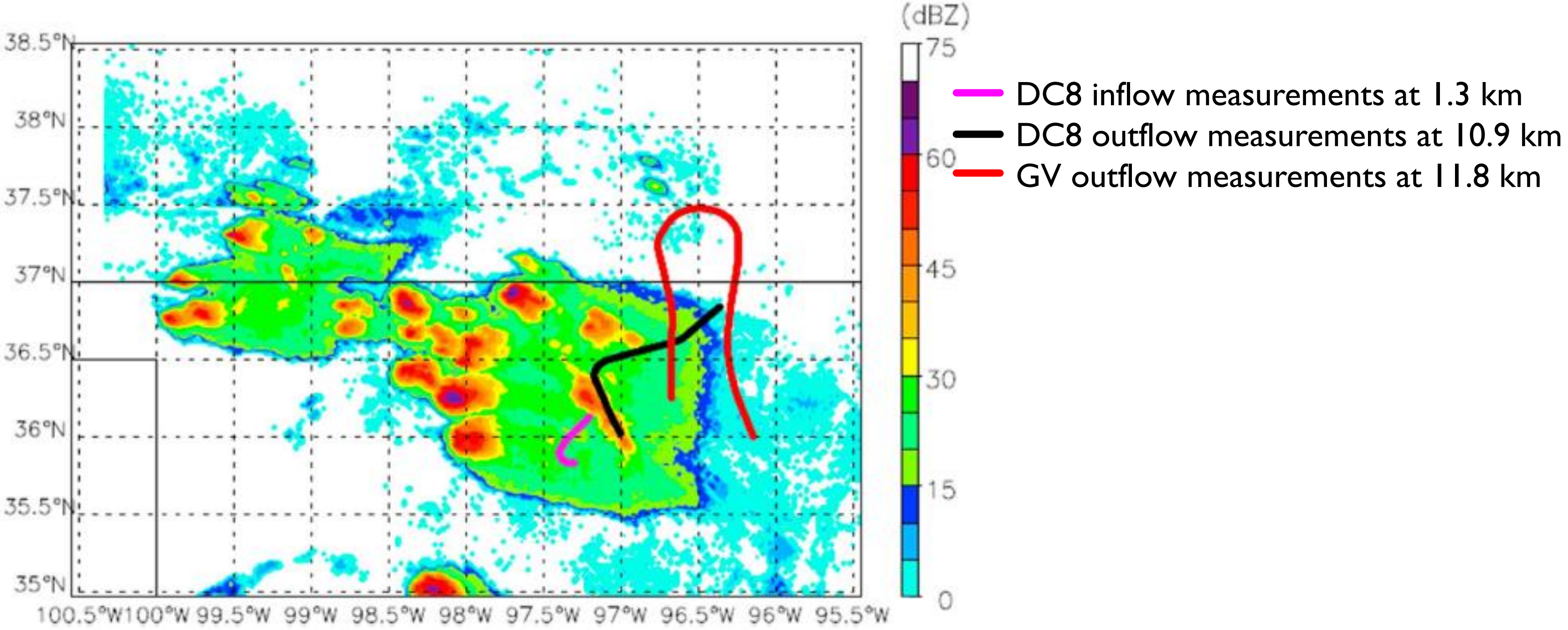


Barth et al., 2015, *BAMS*

- Primary goal:
  - **In the first few hours of active convection**: quantify the convective transport of emissions and water to the upper troposphere, investigate storm dynamics and physics, lightning and its production of nitrogen oxides, wet scavenging and chemistry in the anvil.
  - **12-48 hours after active convection**: quantify the downwind changes in chemistry and composition in the upper troposphere.

MAY 29 OKLAHOMA SEVERE STORM

- The thunderstorm system developed around 21Z on 29 May on the Kansas/Oklahoma border and continued past sunset. Developed into a complex of severe cells.
- The aircraft (DC8, GV) took measurements of the storm from ~20Z on 29 May to ~01Z on 30 May. The DC8 focused on storm inflow and outflow, while the GV concentrated on outflow.



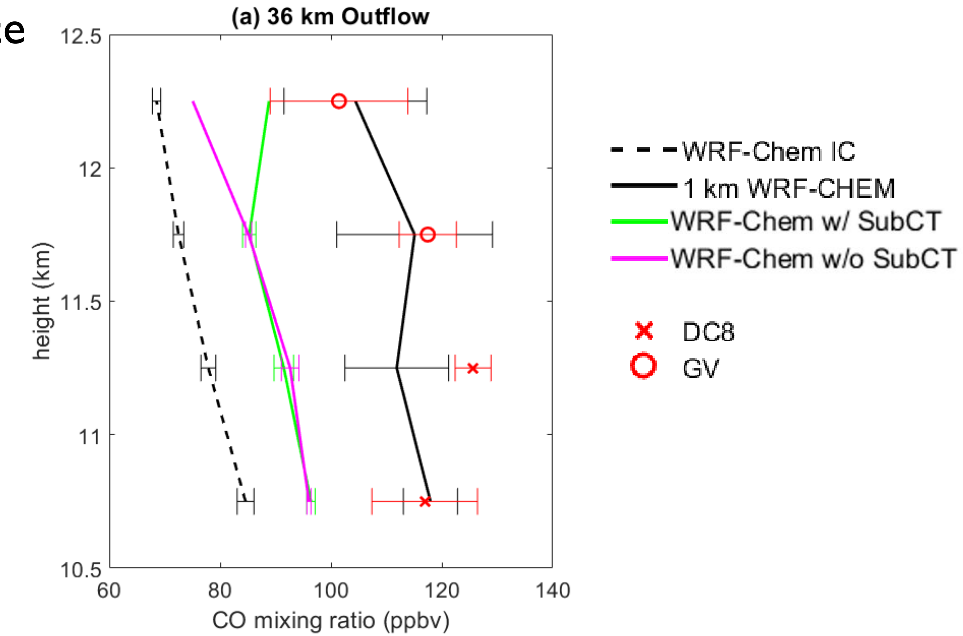
I. DEEP CONVECTIVE TRANSPORT OF TRACE GASES

- Conducted WRF-Chem V3.9 simulations at various spatial resolutions

	Cloud Resolved Run	Cloud Parameterized Run
Meteorology Initial/Boundary Conditions	NAM 18 UTC	
Chemistry Initial/Boundary Conditions	DC-8 measurement to generate I.C. & B.C.	MOZART scaled
Horizontal/ Vertical/ Time Resolution	1 km/ 89 levels/ 3s	36, 12 km/ 90 levels/ 120s, 60s
Cumulus Parameterization	-	GF with KF closure
Microphysics	Morrison	
PBL	YSU	MYJ
Short/longwave radiation	RRTMG	
Lightning Schemes	Price and Rind (1992) lightning flash rate scheme based on maximum vertical velocity	Price and Rind (1992) lightning flash rate scheme based on level of neutral buoyancy (Wong et al., 2013)
Chemistry option	MOZCART	
Fire/ Anthropogenic/ Biogenic Emissions	FINN/ NEI/ MEGAN	

I. DEEP CONVECTIVE TRANSPORT OF TRACE GASES

- Conducted WRF-Chem simulations at various spatial resolutions (36, 12, and 1 km), using CO as an example tracer.
- The cloud resolved simulations (1 km) of deep convective transport of CO compare well with aircraft observations.
- The cloud parameterized simulations (36, 12 km) underestimate the transport of CO
  - **Problem**: the cumulus parameterized convective transport of trace gases in the **chemistry part of WRF-Chem** **is not consistent** with the cumulus parameterization in the **meteorological part of WRF-Chem** (some regions have convective clouds but no convective transport of trace gases).
    - In the chemistry part of the model the subgrid convective transport is based on the GD scheme, while in the meteorological part other convective schemes may be selected (e.g. GF scheme).

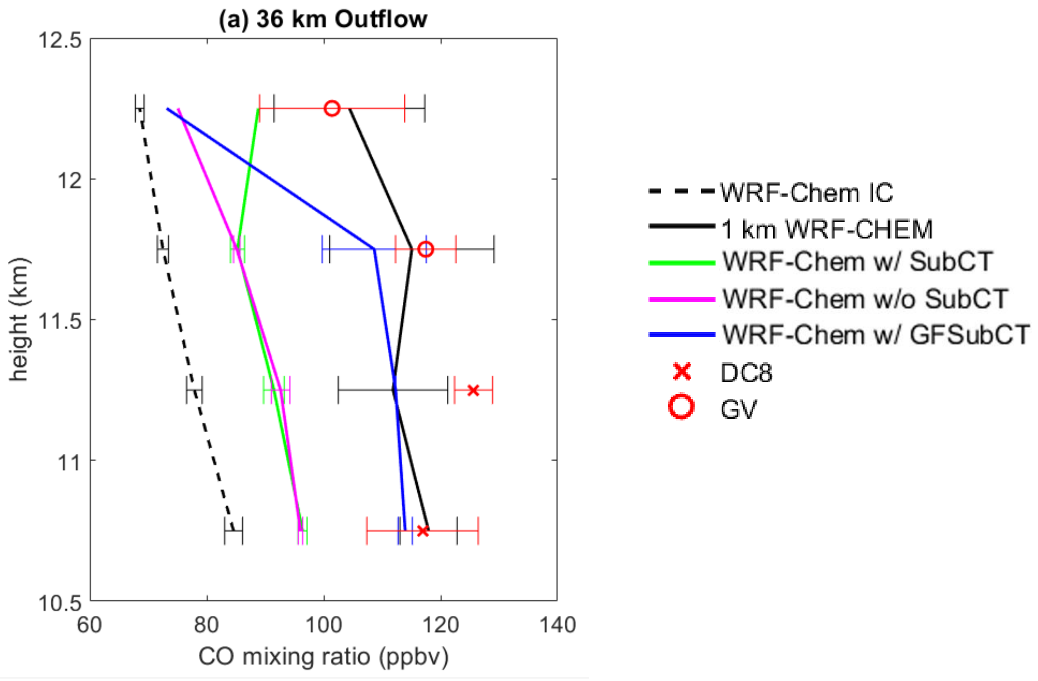


I. DEEP CONVECTIVE TRANSPORT OF TRACE GASES

- The cloud parameterized simulations (36, 12 km) underestimate the convective transport of CO
  - **Solution:** Modify the subgrid convective transport code to be compatible with the Grell-Freitas scheme:
    - using mass flux related variables from GF cumulus parameterization to calculate the subgrid convective transport of trace gases in a new routine within the meteorological part of the model using the following equation (Grell & Freitas, 2014):

$$\left(\frac{\partial C}{\partial t}\right)_{\text{subgrid}} = -\frac{1}{\rho}\frac{\partial}{\partial z}\left[m_u(C_u - C_e) - m_d(C_d - C_e)\right]$$

  - Reduces the RMSE by 39%. Improvement would have been even better if cloud top height had not been underestimated



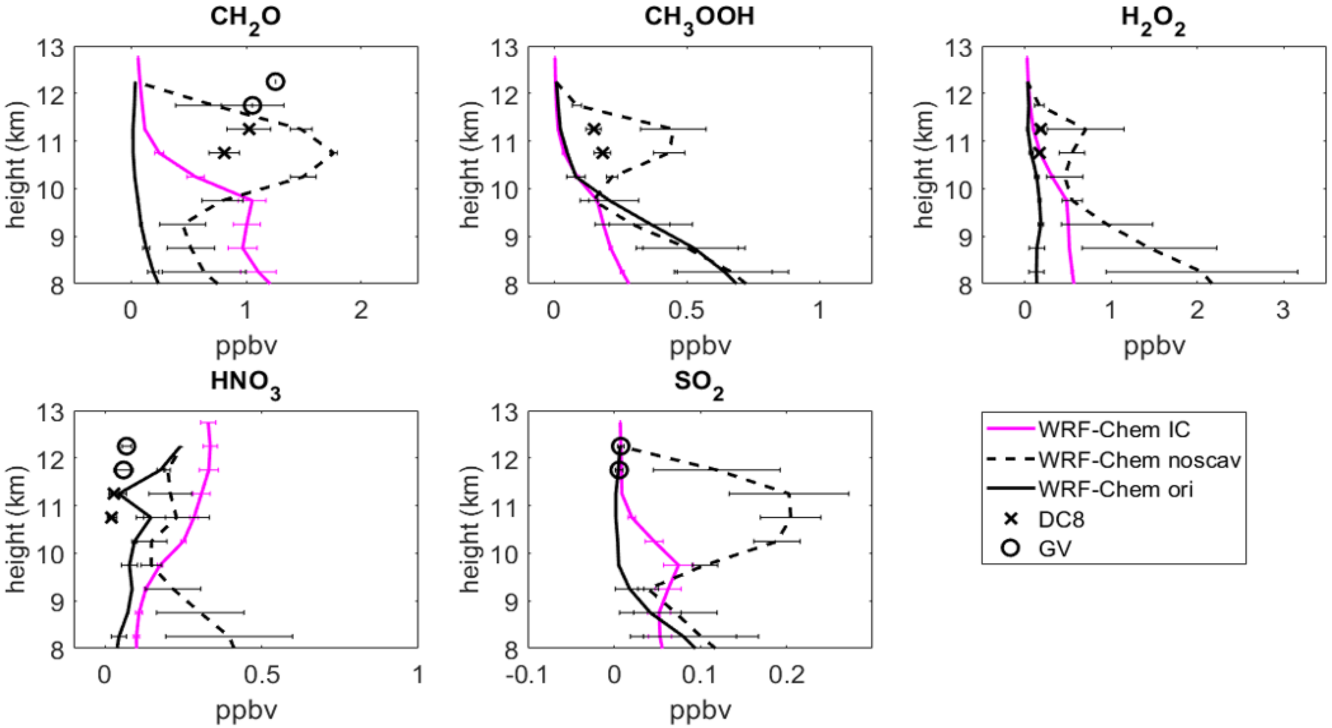
Li et al., 2017, 2018, JGR

II. DEEP CONVECTIVE TRANSPORT OF SOLUBLE SPECIES (WET SCAVENGING)

- Deep convective transport of **soluble species** (wet scavenging)
  - The amount of O_3 and aerosol formed in the UT depends on the net convective transport of gases that are **soluble** and reactive in the aqueous phase.
  - A number of physical processes within the convective core and anvil affect the net convective transport of soluble species, including:
    - General vertical transport (just like insoluble species)
    - Wet scavenging sink:
      - Dissolution in cloud water
      - Removal by precipitation
      - Evaporation and release of dissolved gases
      - Freezing of droplets - part of the dissolved gases may be released and part retained in ice
  - Previous studies found that a primary source of wet scavenging simulation uncertainty is the **fraction of gases that are released from ice during hydrometeor freezing**.

II. DEEP CONVECTIVE TRANSPORT OF SOLUBLE SPECIES

- WRF-Chem is employed at **cloud parameterized resolution** (36 km) with chemistry and emissions.
- We focus on wet scavenging processes of **five soluble species (CH_2O, CH_3OOH, H_2O_2, HNO_3, and SO_2)**
- **Problem:** Default WRF-Chem underestimate the mixing ratio of soluble trace gases in the upper troposphere.
- The subgrid scale wet scavenging depends on the **solubility of the tracer** and on the **conversion rate of cloud water to rain water**.



Li et al., 2019, JGR

II. DEEP CONVECTIVE TRANSPORT OF SOLUBLE SPECIES

- **1st modification**: Introduce **ice retention factor** to **change the solubility of the tracer** when temperature is below the freezing point
  - When droplets freeze, part of the dissolved gases may be released and part retained in ice.
- In the original equation for the subgrid scale wet scavenging sink, C_{si}, all soluble gases retained in ice.
- New equation for the subgrid scale wet scavenging sink (with ice retention factor r, *Bela et al., JGR,2017*):

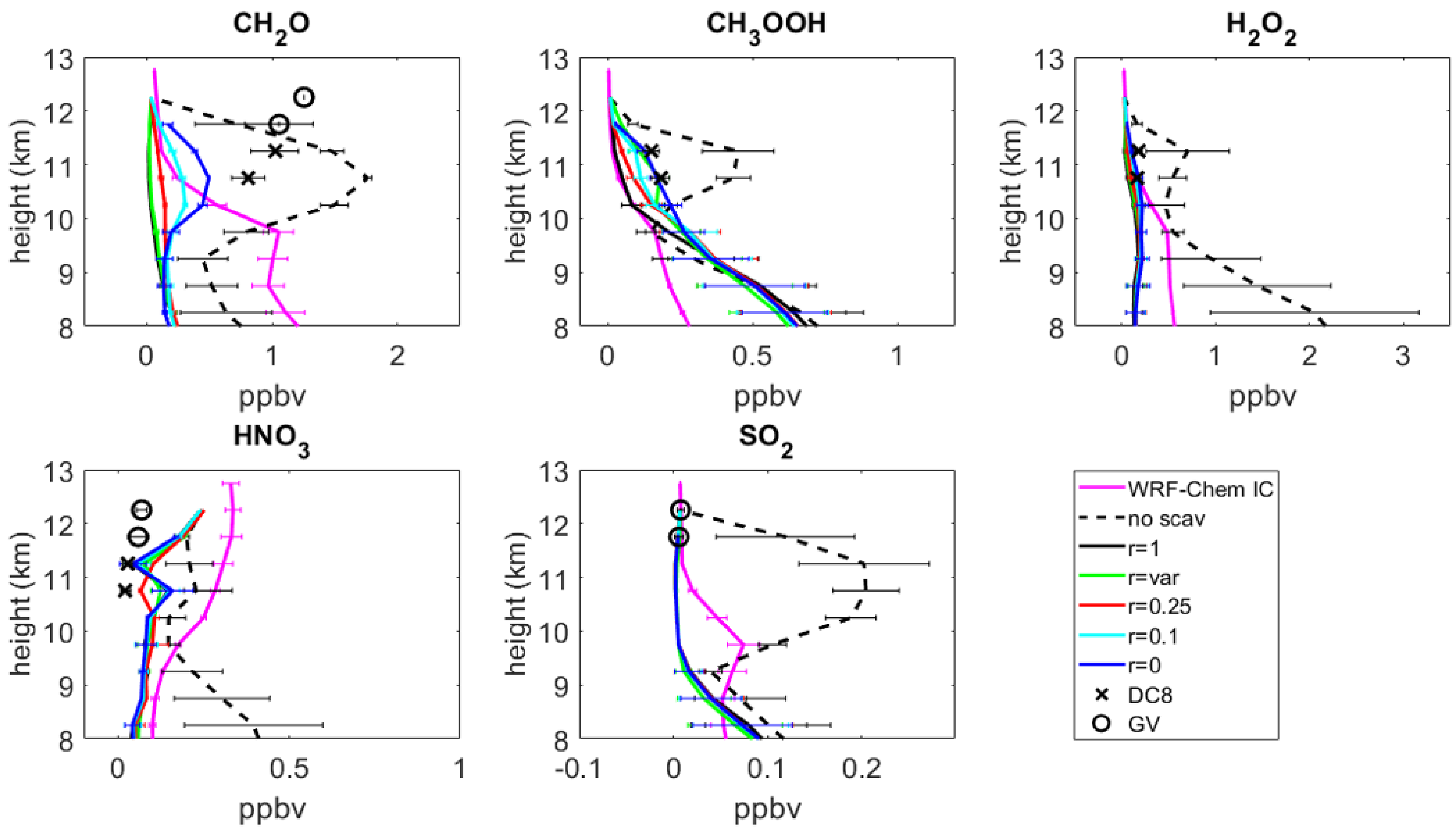
$$C_{si} = \begin{cases} C_{si} & T \geq 273.15\ K \\ rC_{si} & T < 273.15K \end{cases}$$

If r=0: all goes out; If r=1: all stays in ice

- Based on the new equation, we conducted 5 sensitivity tests using different ice retention factors (r):

	CH_2O	CH_3OOH	H_2O_2	HNO_3	SO_2
scav. r=0	**0**	**0**	**0**	**0**	**0**
scav. r=0.1	**0.1**	**0.1**	**0.1**	**0.1**	**0.1**
scav. r=0.25	**0.25**	**0.25**	**0.25**	**0.25**	**0.25**
scav. r=1	**1**	**1**	**1**	**1**	**1**
scav. r=var (Leriche et al., 2013)	**0.64**	**0.02**	**0.64**	**1**	**0.02**

II. DEEP CONVECTIVE TRANSPORT OF SOLUBLE SPECIES



- Compared to the default WRF-Chem results (r = 1.0), when using r=0, the differences between observation and simulation were reduced by 24%, 87%, and 77% for CH_2O, CH_3OOH and H_2O_2

II. DEEP CONVECTIVE TRANSPORT OF SOLUBLE SPECIES

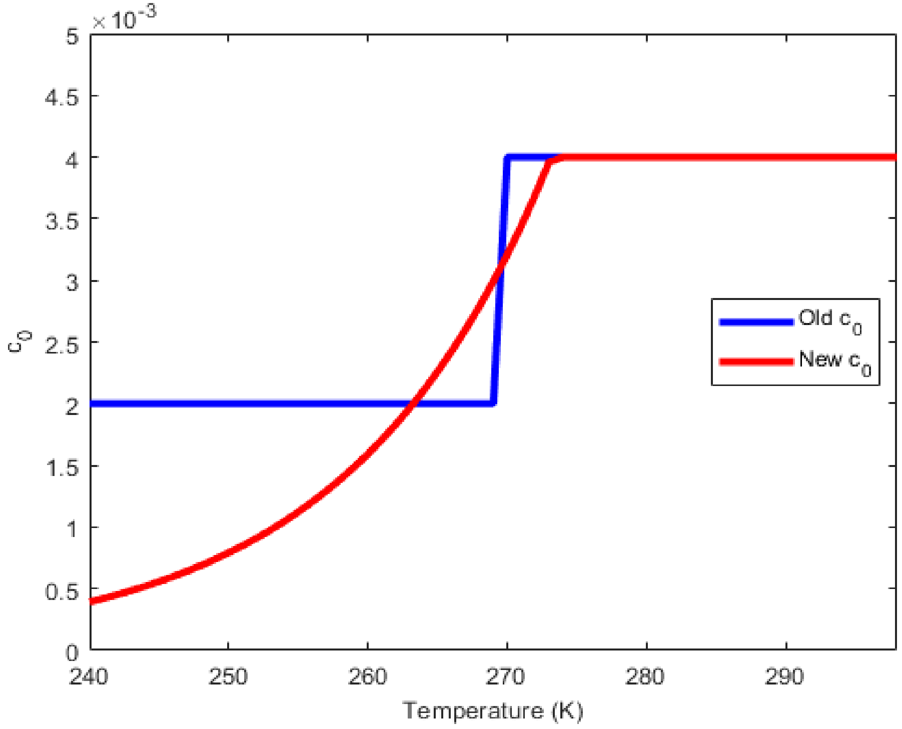
- **2nd modification**: conversion rate of cloud water to rain water.
- Original conversion rate of cloud water to rain water (c_0):

$$c_0 = \begin{cases} 0.004 & T \geq 270\ K \\ 0.002 & T < 270K \end{cases}$$

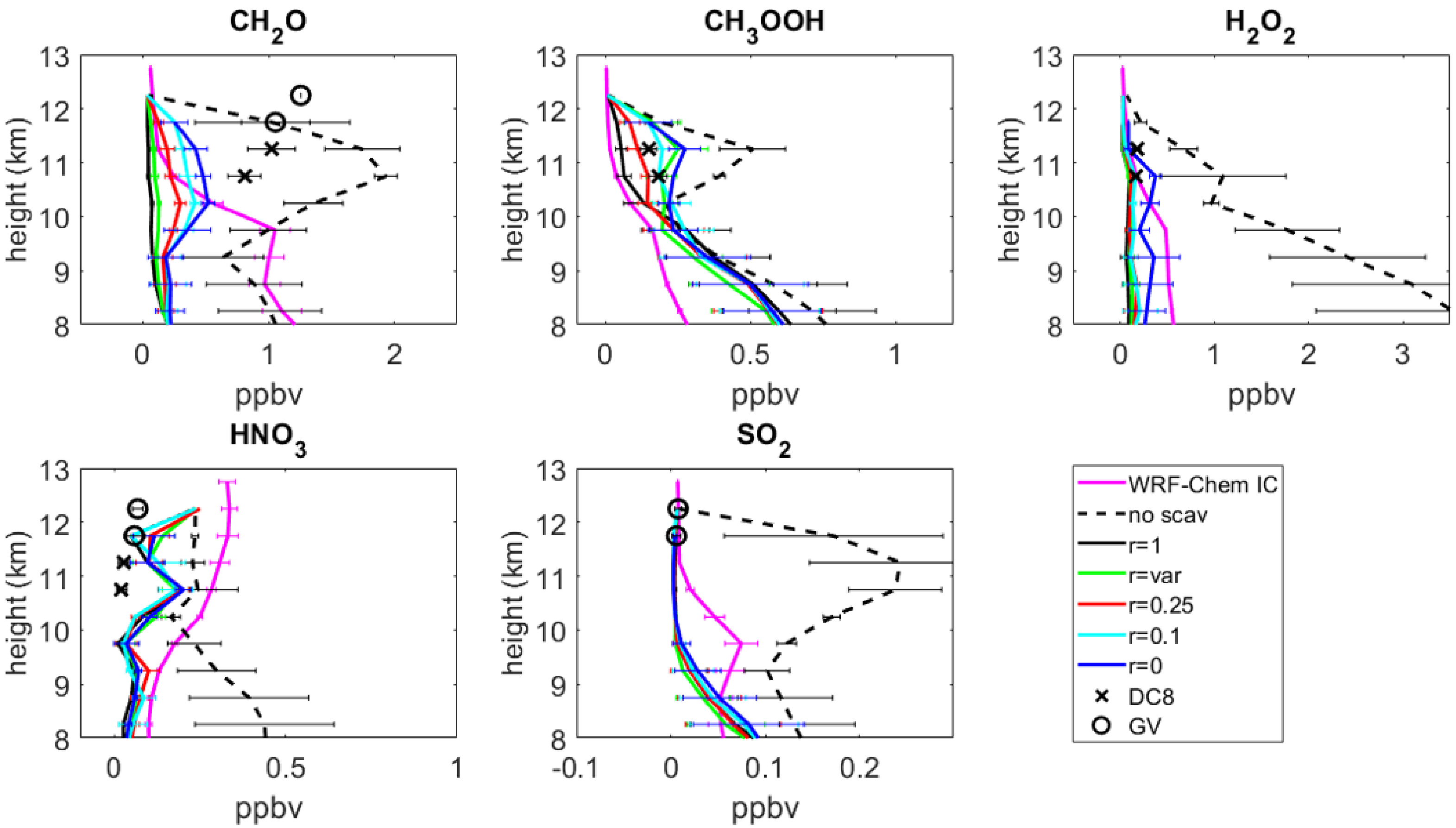
- New c_0 (Han et al., 2016):

$$c_0 = \begin{cases} 0.004 & T \geq 273.15\ K \\ 0.004e^{[a(T-273.15)]} & T < 273.15K \end{cases}$$

  - a=0.07
  - Reduces the conversion rate of cloud water to rain water below 260K



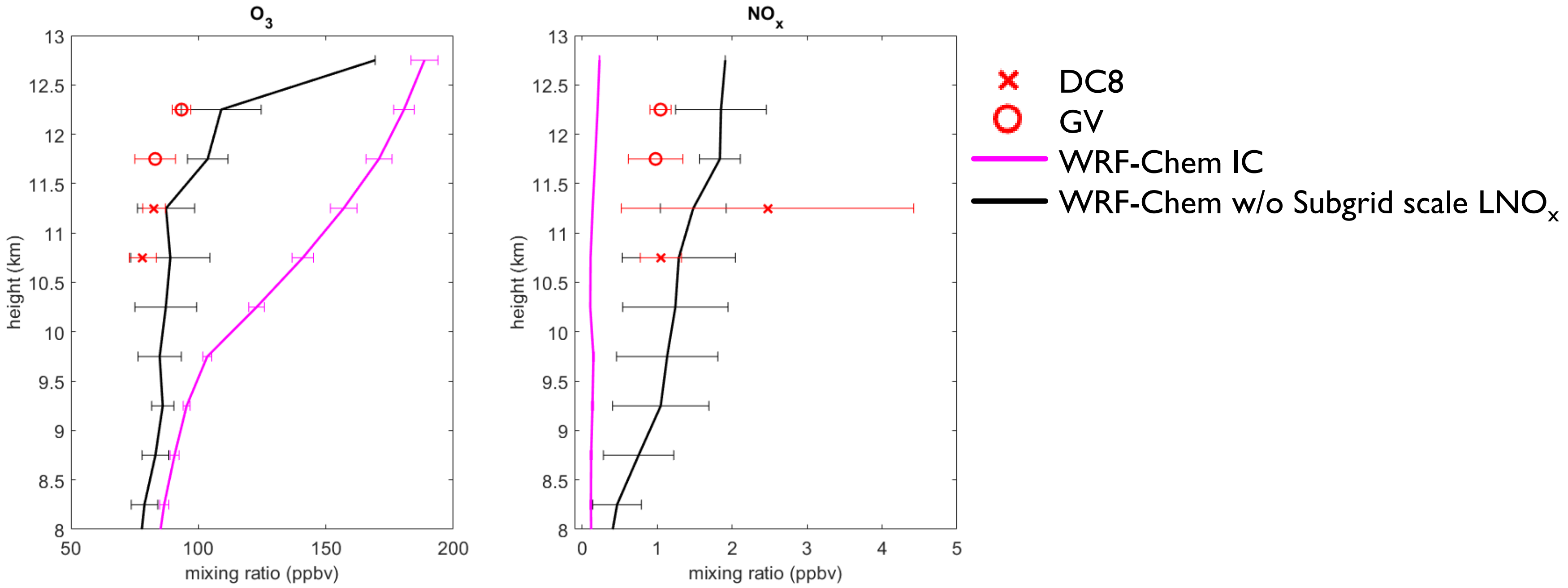
II. DEEP CONVECTIVE TRANSPORT OF SOLUBLE SPECIES



- The use of the new conversion rate increases the UT CH_2O, CH_3OOH and H_2O_2 mixing ratios when using the same ice retention factors.

III. SUBGRID SCALE LNO_x SCHEME

- Lightning is the dominant source of the NO_x in the UT.
- Model fails to simulate the peak in UT NO_x due to underestimate of the convective transport of the LNO_x, which is caused by lack of proper consideration of LNO_x in the subgrid scale convective transport module.



III. SUBGRID SCALE LNO_x SCHEME

- **Problem:**
  - In the subgrid scale convective transport module, the trace gas mixing ratio in the updraft/downdraft depends on the trace gas mixing ratio at the updraft/downdraft initiation level and the subgrid scale entrainment/detrainment rate at each level above/below.
  - In the UT, the subgrid scale entrainment rate is very small. Therefore, the LNO_x produced in the grid-scale only has a small impact on the subgrid-scale NO_x mixing ratio in the parameterized updraft.
- **Solution:** Added LNO_x transport in the GF updrafts and downdrafts.

$$NO_{x_{u,k}} = \frac{((NO_{x_{u,k-1}} + \boldsymbol{LNO_{x_{u,k-1}}})(1-0.5\mathrm{det}_u) + ent_u NO_{x_{e,k-1}})}{1-0.5\mathrm{det}_u + ent_u}$$

$$NO_{x_{d,k}} = \frac{((NO_{x_{d,k+1}} + \boldsymbol{LNO_{x_{d,k+1}}})(1-0.5det_d) + ent_d NO_{x_{e,k-1}})}{1-0.5det_d + ent_d}$$

$$\left(\frac{\partial C}{\partial t}\right)_{\mathrm{subgrid}} = -\frac{1}{\rho}\frac{\partial}{\partial z}[m_u(C_u - C_e) - m_d(C_d - C_e)]$$

- LNOx in the updraft and downdraft are calculated using Ott et al. (2010) scheme with LMA measured combined IC+CG vertical distributions.

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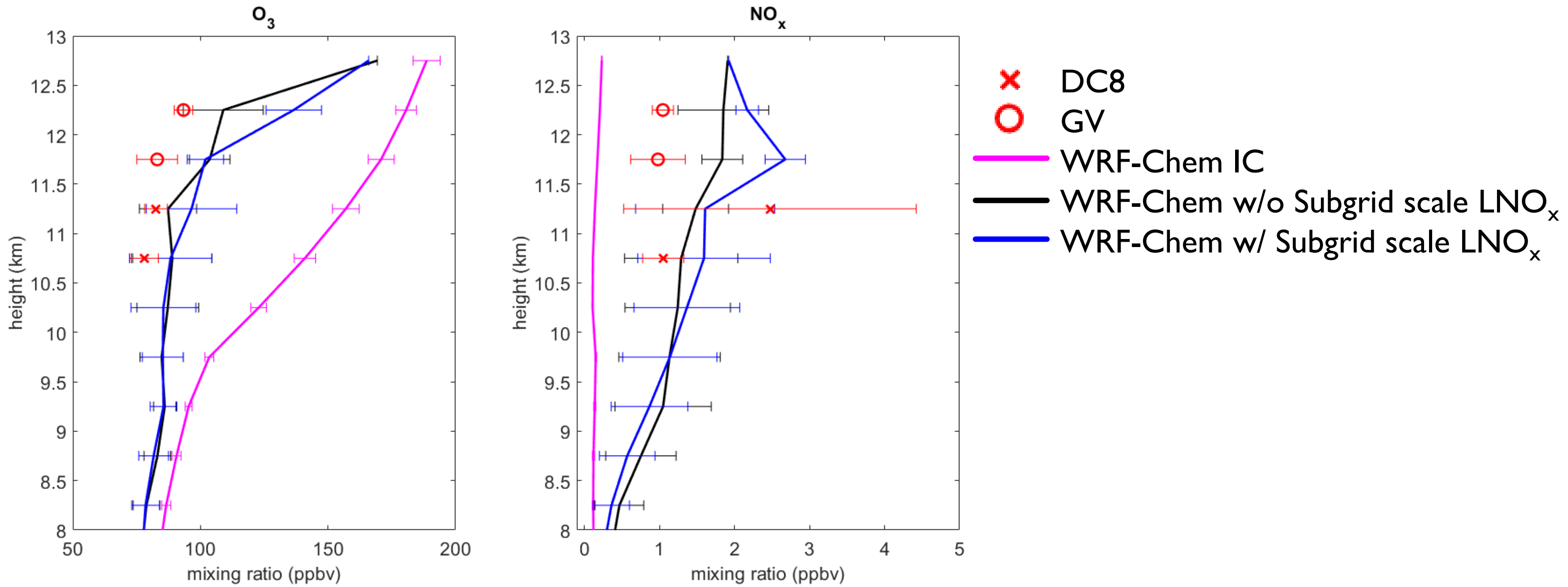

CONCLUSION

- To better reproduce the transport of trace gases, subgrid convective transport needs to be consistent with the convective cloud parameterization in the meteorological model that drives the chemical transport.

- The default WRF-Chem removed too much CH_2O, CH_3OOH, and H_2O_2 in the upper troposphere.
- The introduction of ice retention factor and the usage of new conversion rate of cloud water to rain water improved the model simulation of soluble trace gases.

- The default WRF-Chem failed to simulate the UT NO_x peak due to the underestimate of the subgrid scale LNO_x convective transport.
- Adding subgrid scale LNO_x in the parameterized updraft and downdraft improves the simulation of UT NO_x.


Thank you!