# Evaluation of CMAQ simulated $NH_3$ and $PM_{2.5}$ concentration in Taiwan with dynamical $NH_3$ emission parameterization

Chia-Hua Hsu, Fang-Yi Cheng

*Department of Atmospheric Science, National Central University*



# Outline

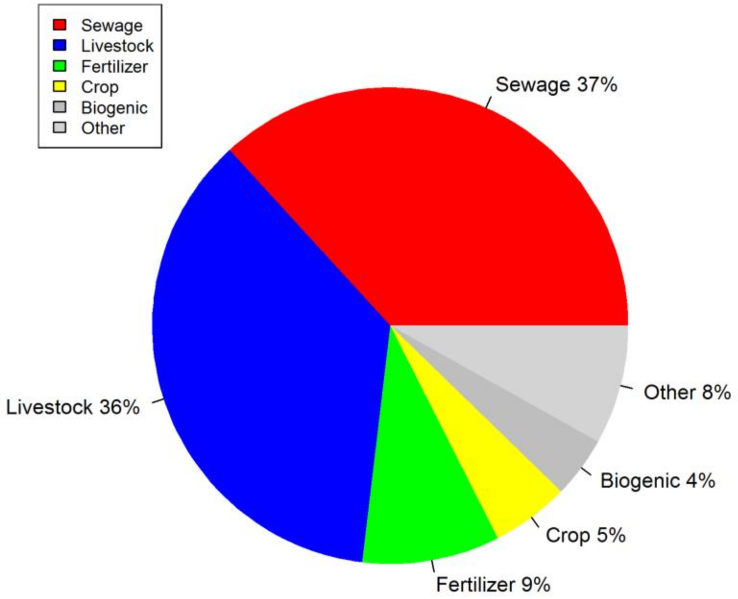
- Introduction
- Observation and Method
- Model Configuration & Experiment design
- Model and observation data comparison
- Summary and Conclusion

# Introduction

- Ammonia ($NH_3$) is an important precursor of $PM_{2.5}$ and studies have shown that $NH_3$ have prominent diurnal and seasonal variation. (Gilliland et al,. 2006; Paulot et al., 2014, Pinder et al., 2006, )

- Apply diurnal or seasonal variation to $NH_3$ emission could reduce the $NH_3$ and $PM_{2.5}$ simulated bias. (Pinder et al., 2006; Zhu et al., 2015)

- Based on 2010 Taiwan emission inventory (TEDS8.1), ammonia is majorly emitted by urban sewage (37%), livestock operation (36%) and agriculture activities (14%).



# Introduction

- With the constant $NH_3$ emission rate, model create large bias in the simulated $NH_3$ and $PM_{2.5}$, especially induce high nitrate concentration.

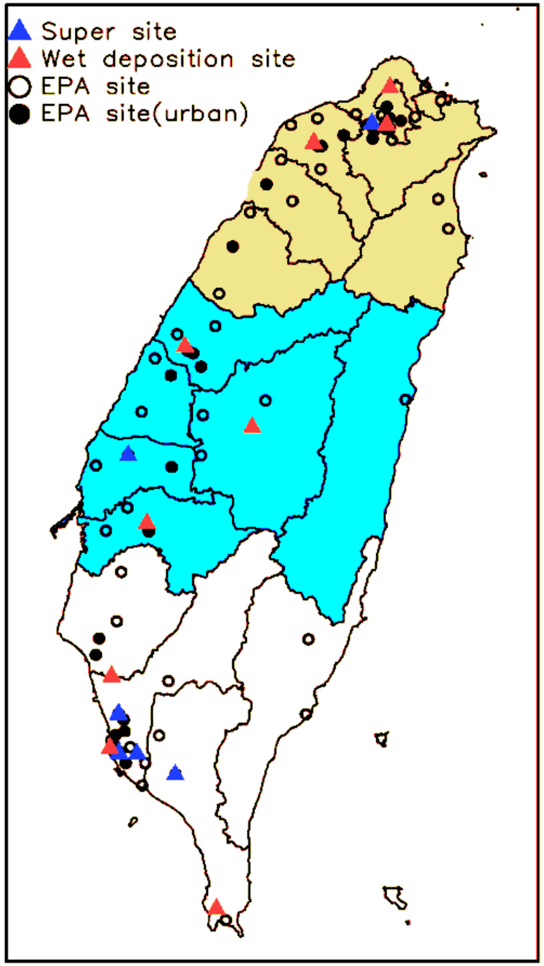
- Urban sewage accounts for an extremely large portion of $NH_3$ emission (37%) in Taiwan compared to other emission inventory. (Huang et al., 2012, Bouwman et al., 1997, Kang et al., 2016, Street et al., 2004).

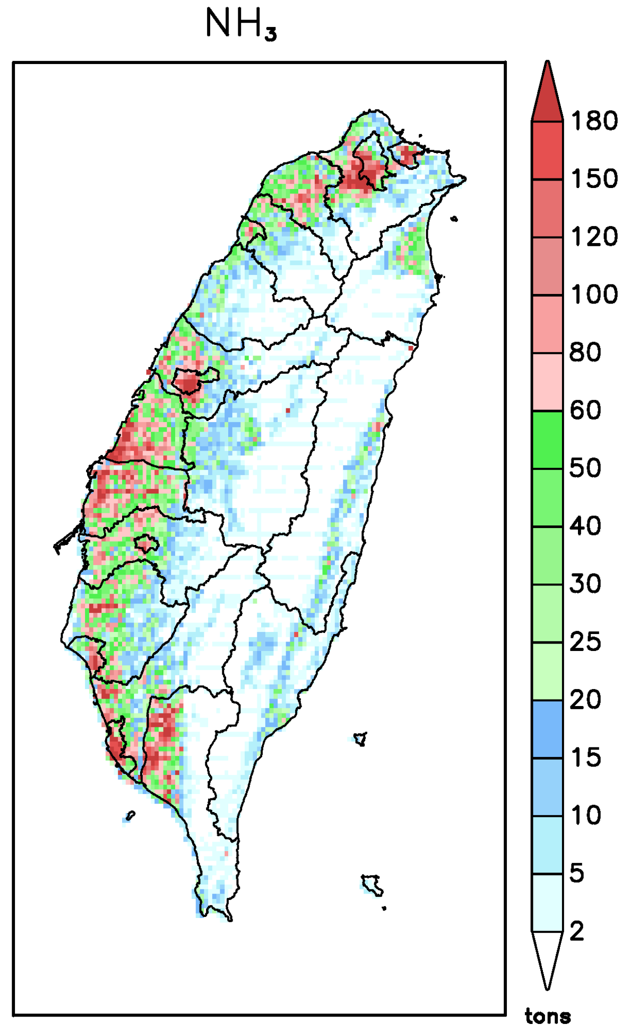
- The object of this study is :

1. Adopted a dynamical $NH_3$ emission parameterization to improve the diurnal and seasonal variation of $NH_3$ and nitrate.

2. Conduct an emission reduction experiment to clarify the problem of urban sewage emission.

# Observation and Emission Data

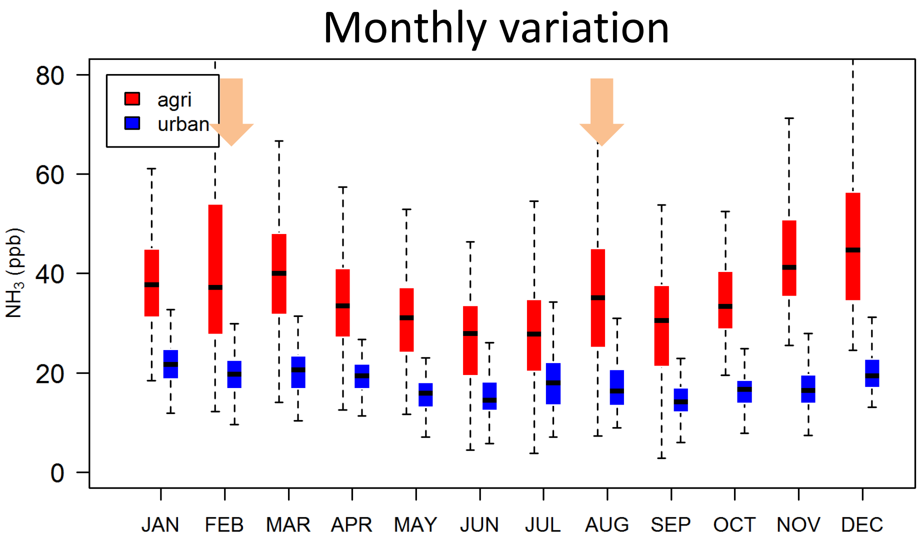


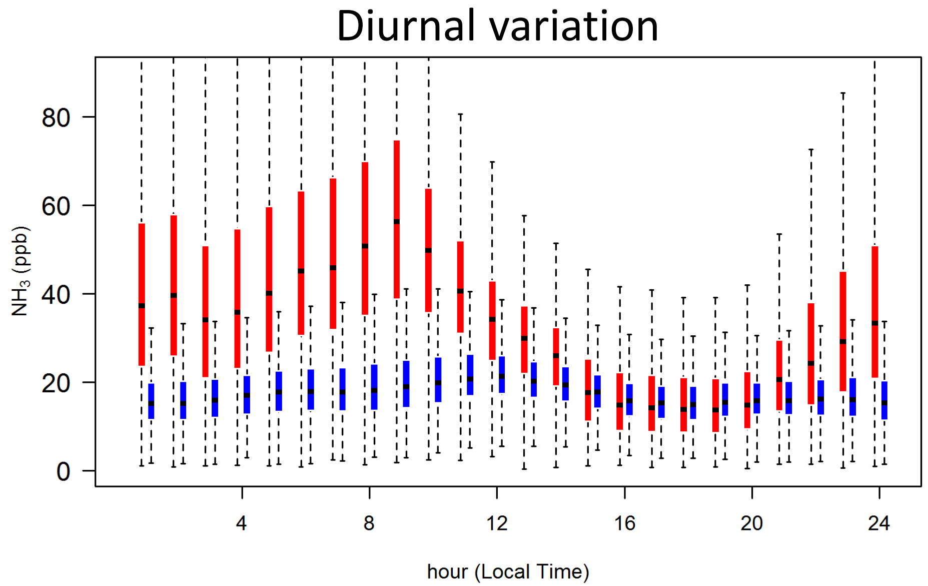




| Observation Network | | |
|---|---|---|
| **Type** | **number** | **Data frequency** |
| **EPA stations** | **66** | **hourly** |
| **Super Sites Network** | **6** | **hourly** |
| **Acid Rain Network** | **9** | **daily** |

# Diurnal and seasonal variation of observed $NH_3$

- Based on 2009-2010 observation, $NH_3$ have two peak in FEB-MAR and AUG in the agricultural region which reflect the major growing season, while urban region $NH_3$ remain at similar level.
- $NH_3$ has significant diurnal variation with higher(lower) $NH_3$ concentration during nighttime (daytime) in Taiwan .

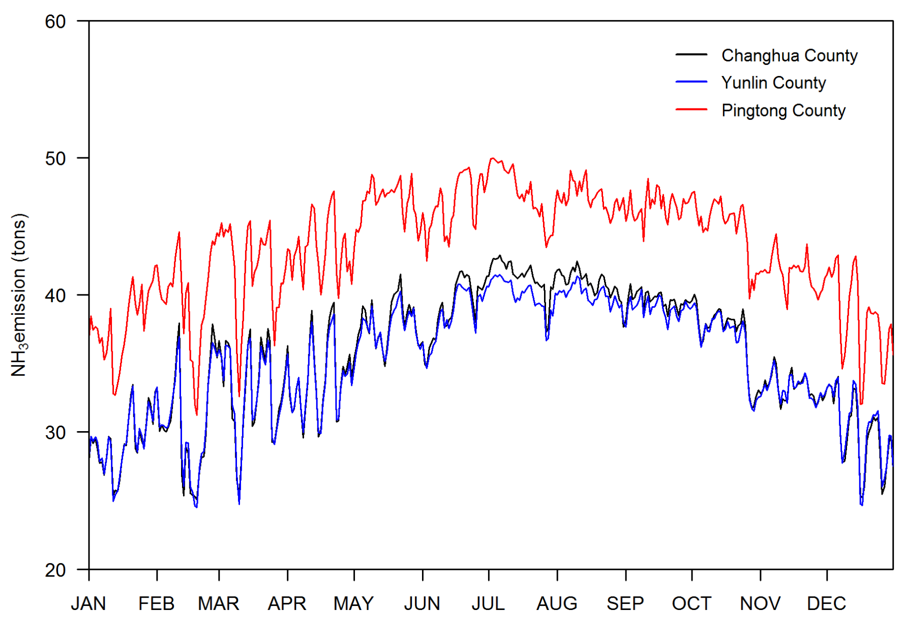


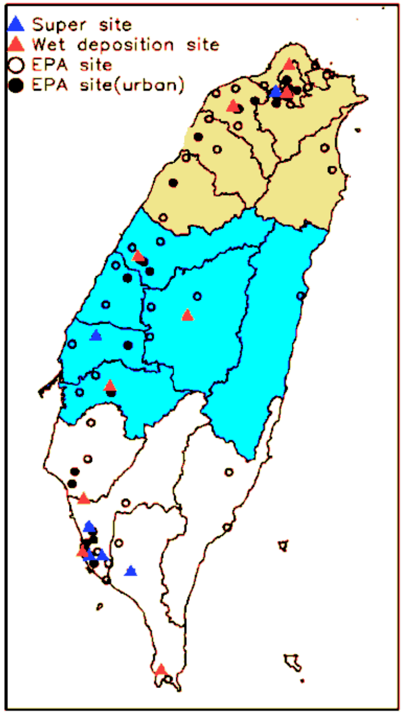



# Method – daily $NH_3$ emission allocation

- We calculate daily $NH_3$ emission following Gyldenkærne et al.(2005).
- Daily livestock emission of each county are calculated based on observed daily mean temperature (*T*) and wind speed (*V*), where *E* denoted the annual total emission and $E_{d,s}$ is daily emission, a(b) is 0.89(0.26):

$$E_{d,s} = E \frac{T_s{}^a V_s{}^b}{\sum_{s=1}^{365} T_s{}^a V_s{}^b}$$



# Method – daily $NH_3$ emission allocation

- Daily rice fertilizer ($E_f$) and rice crop emission ($E_c$) are calculated based on Gaussian function and growing degree day (GDD)
- Daily other crop fertilizer emission are allocated following the monthly variation in MASAGE $NH_3$ inventory (Paulot et al., 2014)

$$E_f = E * exp^{0.0223T} exp^{0.0419W} \frac{1}{\sigma\sqrt{2\pi}} \times e^{(\frac{(t-\mu)^2}{-2\sigma^2})}$$

$$E_c = E * \frac{1}{\sigma\sqrt{2\pi}} \times e^{(\frac{(t-\mu)^2}{-2\sigma^2})}$$

$$GDD = \sum_a^b \frac{(T_{max} + T_{min})}{2} - 10$$

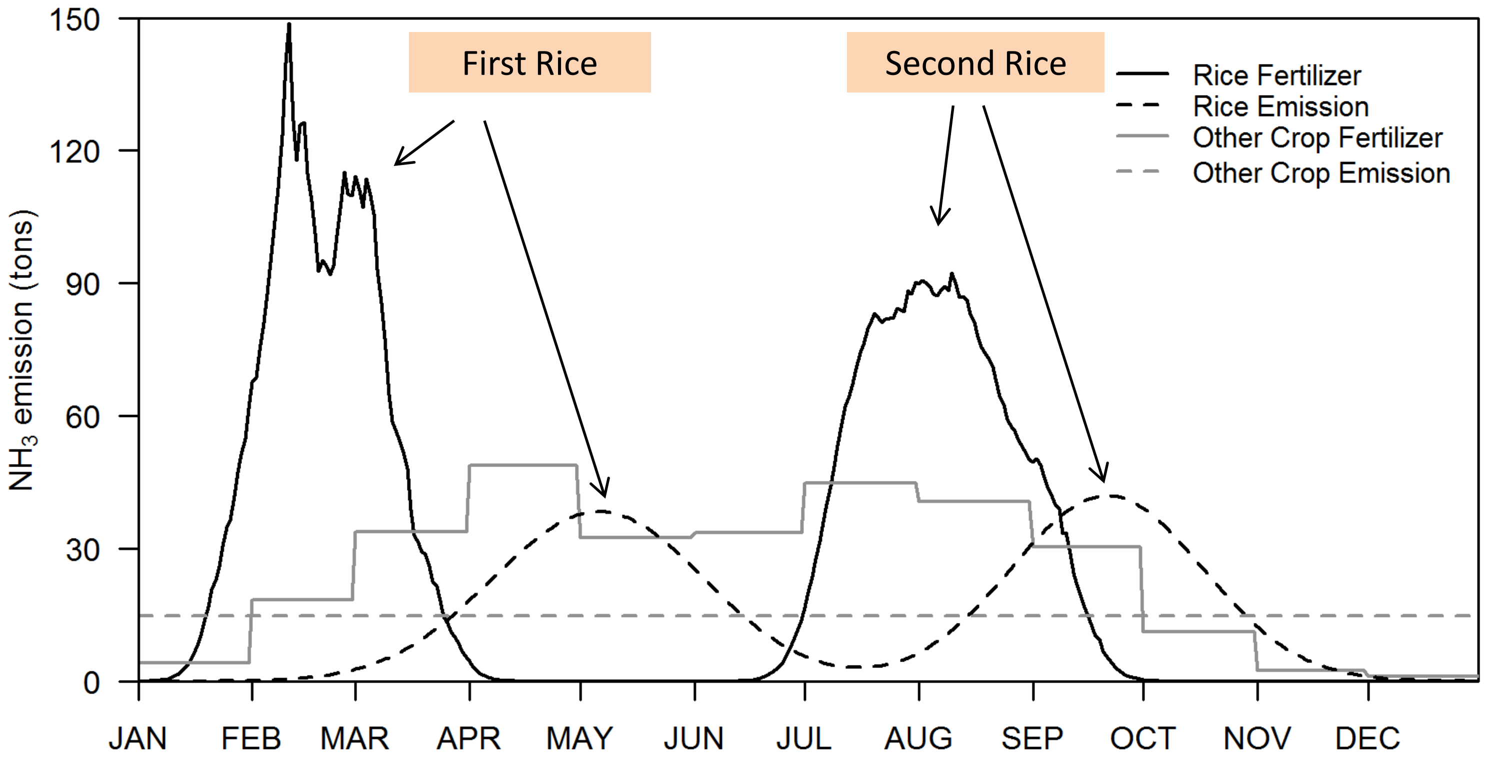
| Region | First Rice | Second Rice |
|---|---|---|
| Northern Taiwan | 60 | 211 |
| Central Taiwan | 46 | 201 |
| Southern Taiwan | 32 | 191 |

Rice Transplanting Day (Day of year)

| | Fraction (%) | μ (mean of Gaussian function) | σ |
|---|---|---|---|
| 1st | 25 | Transplanting Day - 3 | 7 |
| 2ed | 25 | 176.3 (205.6) | 7 |
| 3rd | 30 | 337.5 (515.5) | 7 |
| 4th | 20 | 698.7 (890) | 7 |
| Rice | - | 968.25 (1228.25) | 25 |

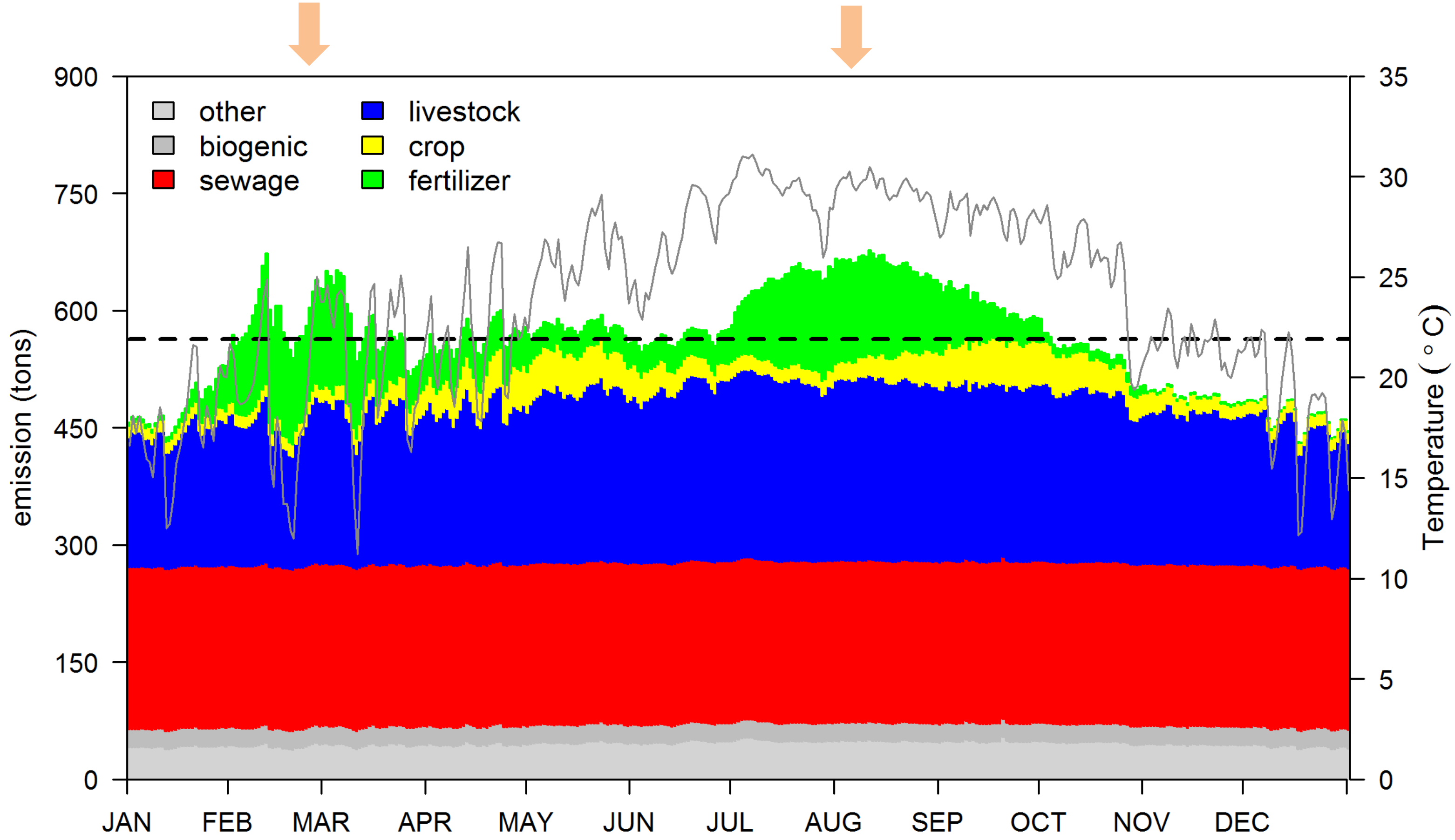
# Method – daily $NH_3$ emission allocation

- Daily rice fertilizer ($E_f$) and rice crop emission ($E_c$) are calculated based on Gaussian function and growing degree day (GDD)
- Daily other crop fertilizer emission are allocated following the monthly variation in MASAGE $NH_3$ inventory (Paulot et al., 2014)



# Method – daily $NH_3$ emission allocation

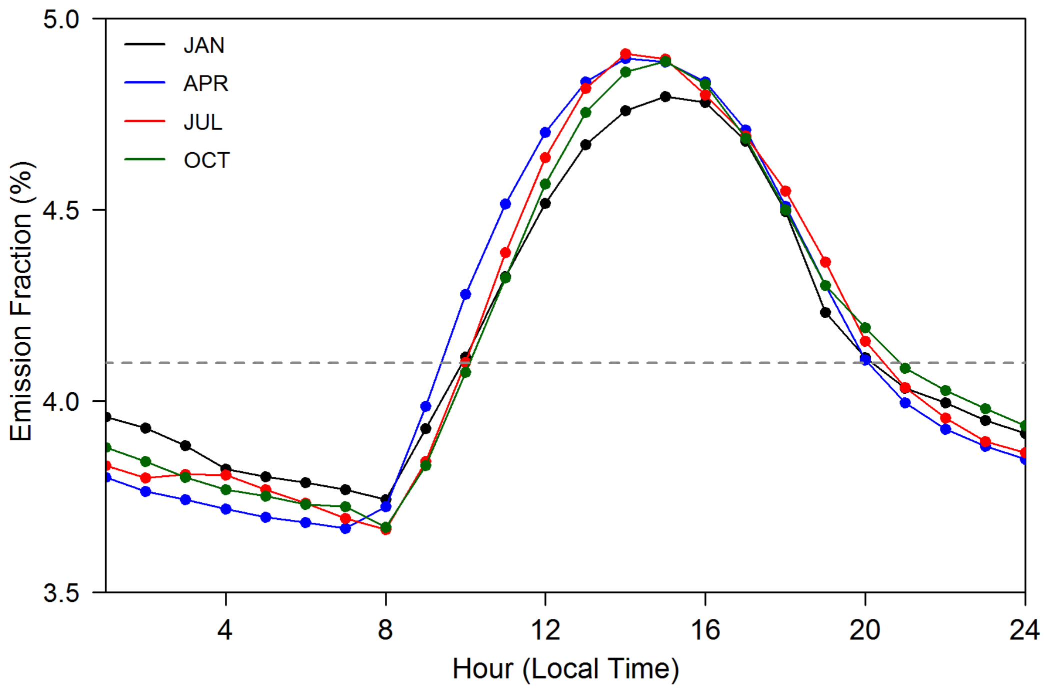
- Other $NH_3$ sources (mobile, industry, biogenic, sewage) are equally distributed to each month.
- $NH_3$ emission have higher value in summer months and growing seasons (Feb - Mar, Jul - Aug) .



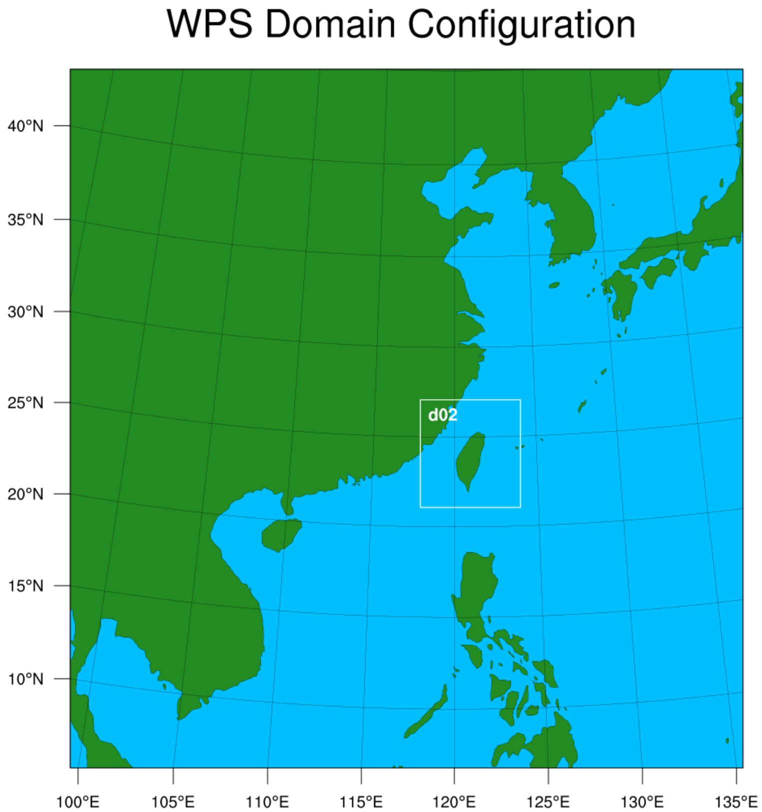
# Method – diurnal $NH_3$ emission allocation

- The diurnal variation of $NH_3$ follows the same method as daily $NH_3$ emission treatment, but *T* and *V* now are provided by WRF, $E_h$ ($E_d$) is hourly (daily) emission of each grid.
- Emission from mobile is applied by a double peak temporal profile.
- Emission from industry assume fixed emission rate throughout the day.

$$E_h = E_d \frac{T^a V^b}{\sum_{s=1}^{24} T^a V^b}$$



# Model Configuration & Experiment design



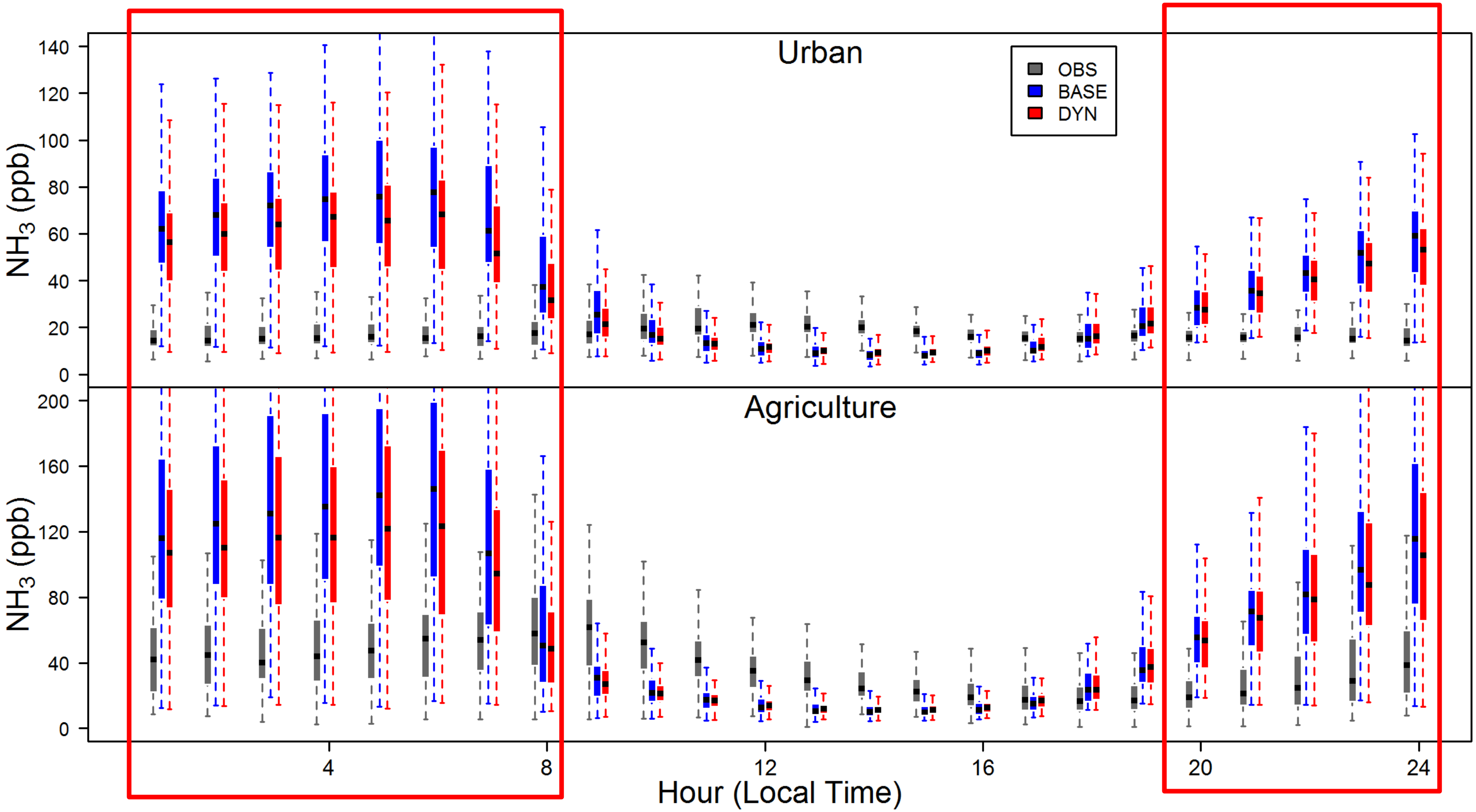

- In this study, we propose three experiments (**BASE**, **DYN**,**DYN1**) to investigate the result of $NH_3$ treatment. DNY1 reduce 80% of urban sewage emission (29.4 % of total $NH_3$).
- Based on the weather classification (Hsu and Cheng, 2016), we choose studying cases that include weak synoptic forcing days → have higher pollutants level and less LRT pollutants.

| WRF version | WRF3.7.1 | |
|---|---|---|
| WRF setting | D01 | D02 |
| resolution | 15 km | 3km |
| CMAQ version | CMAQv5.2 | |
| Anthropogenic EMIS | TEDS8.1 | |
| ASIA EMIS | MICSASIA 2010 | |
| Chemical mechanism | CB05e51-AE6 | |
| Biogenic EMIS | MEGAN2.04 | |

| Experiment | diurnal variation | seasonal variation | $NH_3$ reduction |
|---|---|---|---|
| **Static(BASE)** | - | - | - |
| **Dynamic (DYN)** | v | v | - |
| **Dynamic1 (DYN1)** | v | v | v |

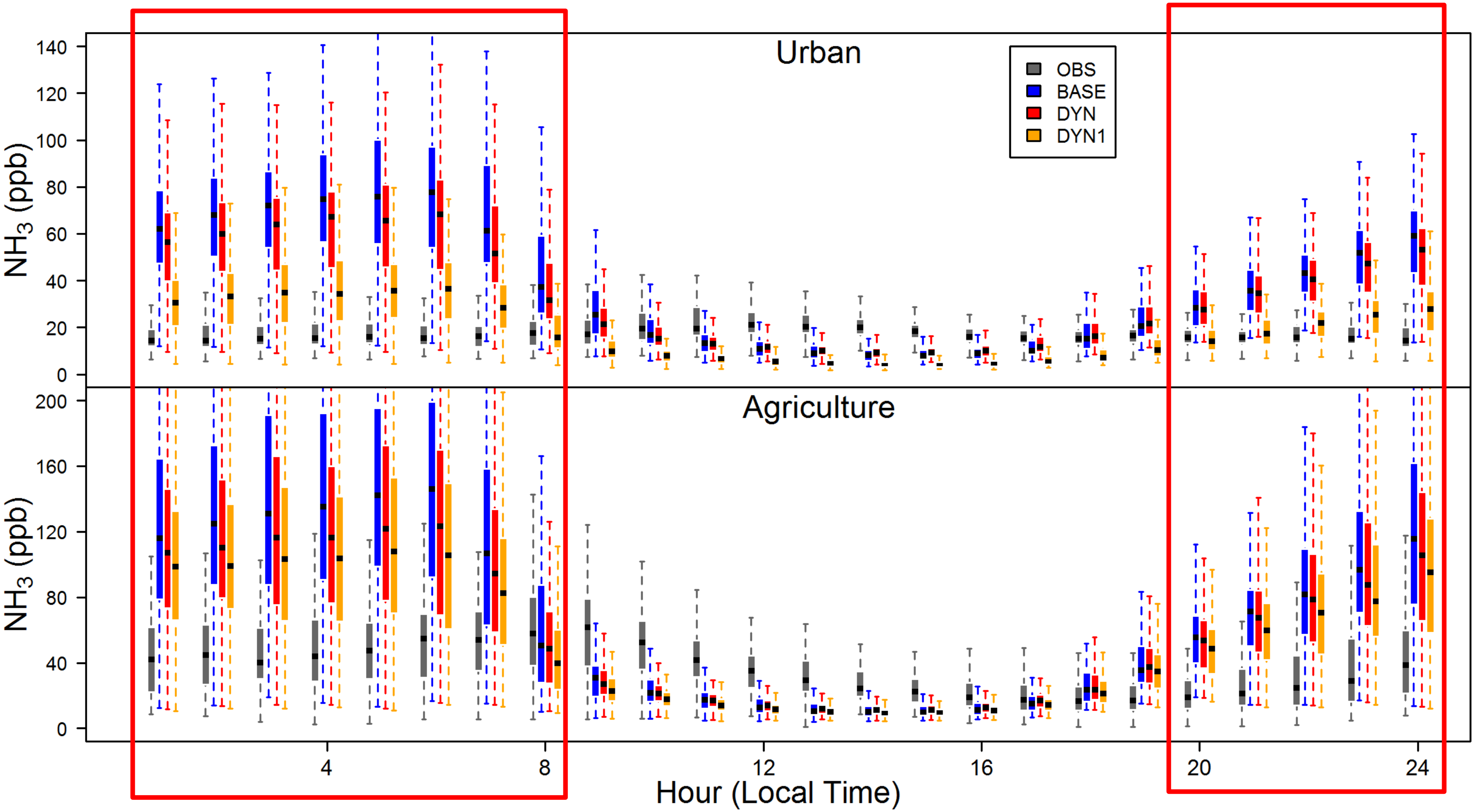
| Simulation Time Period | |
|---|---|
| Time Duration | Weak Synoptic Forcing Days |
| 2010/01/02 - 2010/01/08 | 2 |
| 2010/02/03 - 2010/02/10 | 6 |
| 2010/02/19 - 2010/02/28 | 6 |
| 2010/03/09 - 2010/03/19 | 6 |
| 2010/05/26 - 2010/05/31 | 2 |
| 2010/06/22 - 2010/06/28 | 0 |
| 2010/07/15 - 2010/07/25 | 8 |
| 2010/08/22 - 2010/08/28 | 5 |
| 2010/10/05 - 2010/10/14 | 3 |
| 2010/11/10 - 2010/11/14 | 1 |
| 2010/11/25 - 2010/11/30 | 3 |
| 2010/12/08 - 2010/12/13 | 6 |
| 2010/12/16 - 2010/12/21 | 5 |

# Result - Diurnal variation of $NH_3$




- BASE case overestimate $NH_3$ significantly during nighttime.
- DYN case improve the simulation result but still overestimate $NH_3$ by a factor of 3 in the urban region. → Too much emission in the urban region

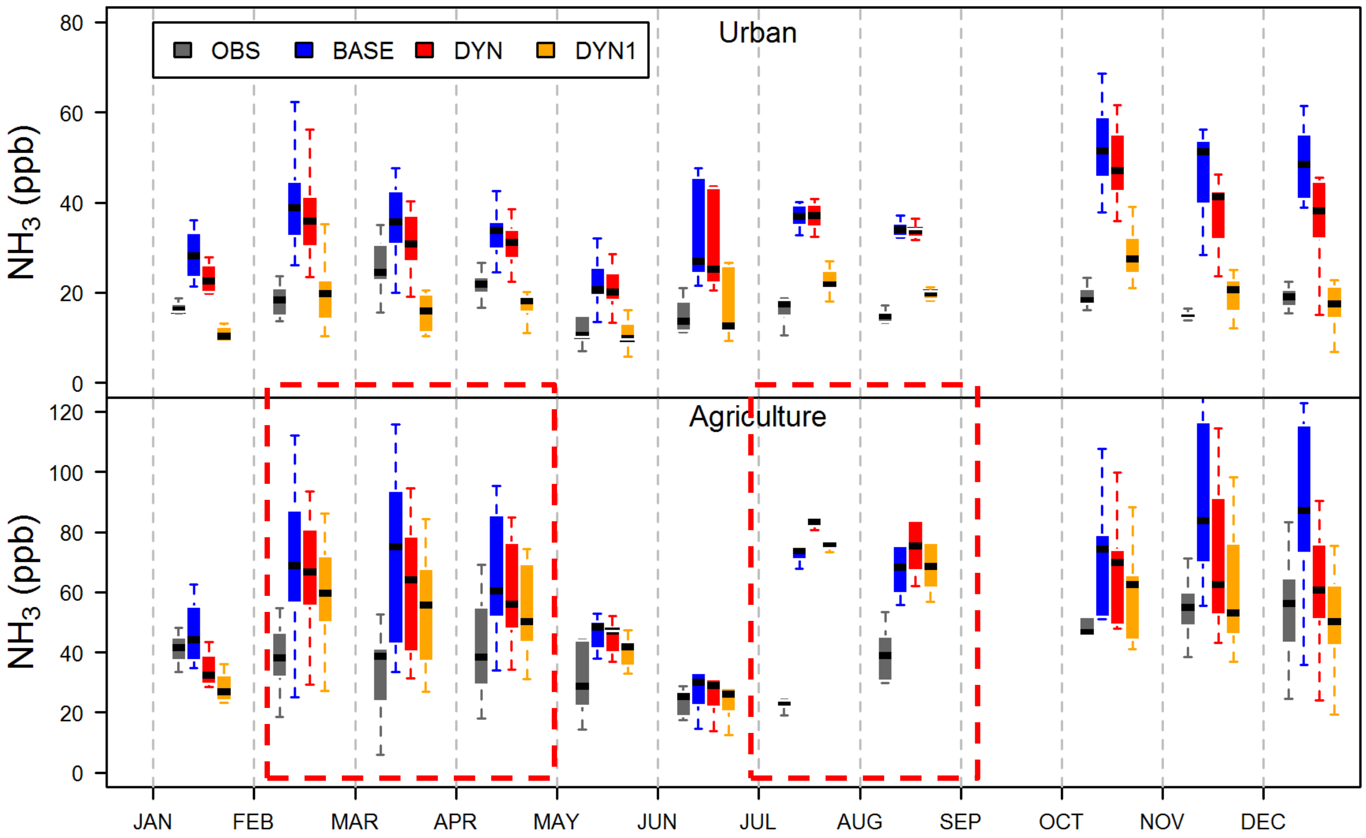
# Result - Diurnal variation of $NH_3$




- BASE case overestimate $NH_3$ significantly during nighttime.
- DYN case improve the simulation result but still overestimate $NH_3$.
- In urban region, DYN1 is much closer to the observation, but the overestimation still exist in agriculture region.

# Result - Monthly variation of $NH_3$

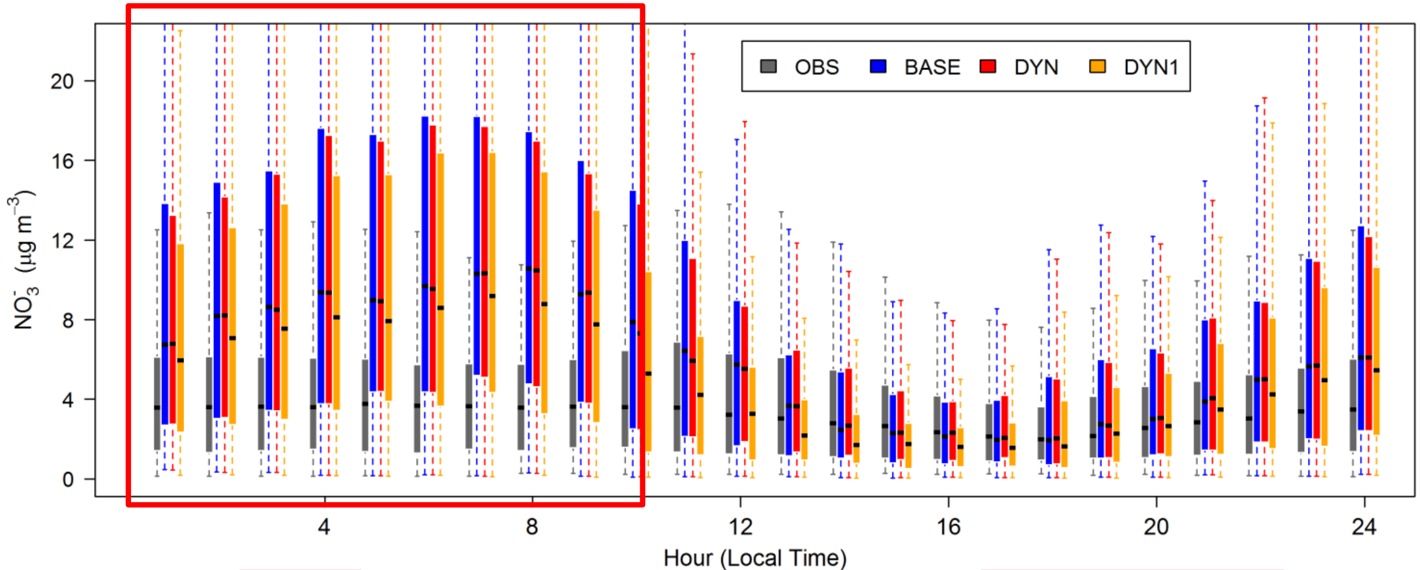
- Dynamical $NH_3$ treatment improve the seasonal variation of simulated $NH_3$.
- In urban region, DYN1 shows good agreement with observation.
- In agricultural region, DYN and DYN1 over-predict $NH_3$ in growing season.

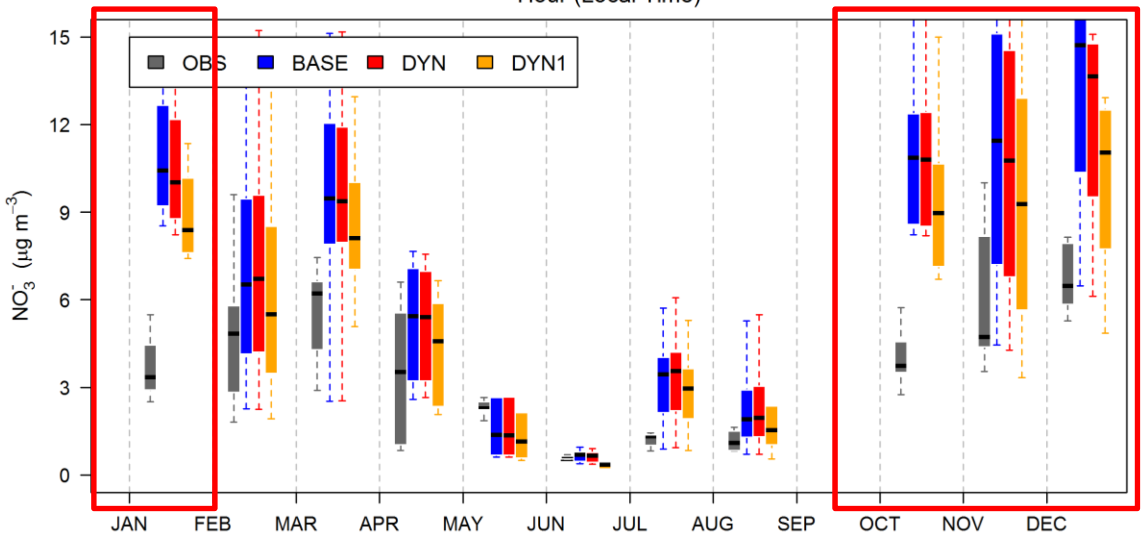



| | Average | MB |
|---|---|---|
| Total Mean | | |
| OBS | 26.07 | - |
| STD | 49.48 | 24.06 |
| DYN | 44.58 | 18.83 |
| DYN1 | 31.42 | 5.72 |
| Urban Region | | |
| OBS | 18.35 | - |
| STD | 39.27 | 19.64 |
| DYN | 35.14 | 15.43 |
| DYN1 | 19.08 | -0.30 |
| Agri Region | | |
| OBS | 41.50 | - |
| STD | 69.91 | 32.88 |
| DYN | 63.48 | 25.65 |
| DYN1 | 56.12 | 17.75 |

# Result – Monthly and diurnal variation of $NO_3^-$

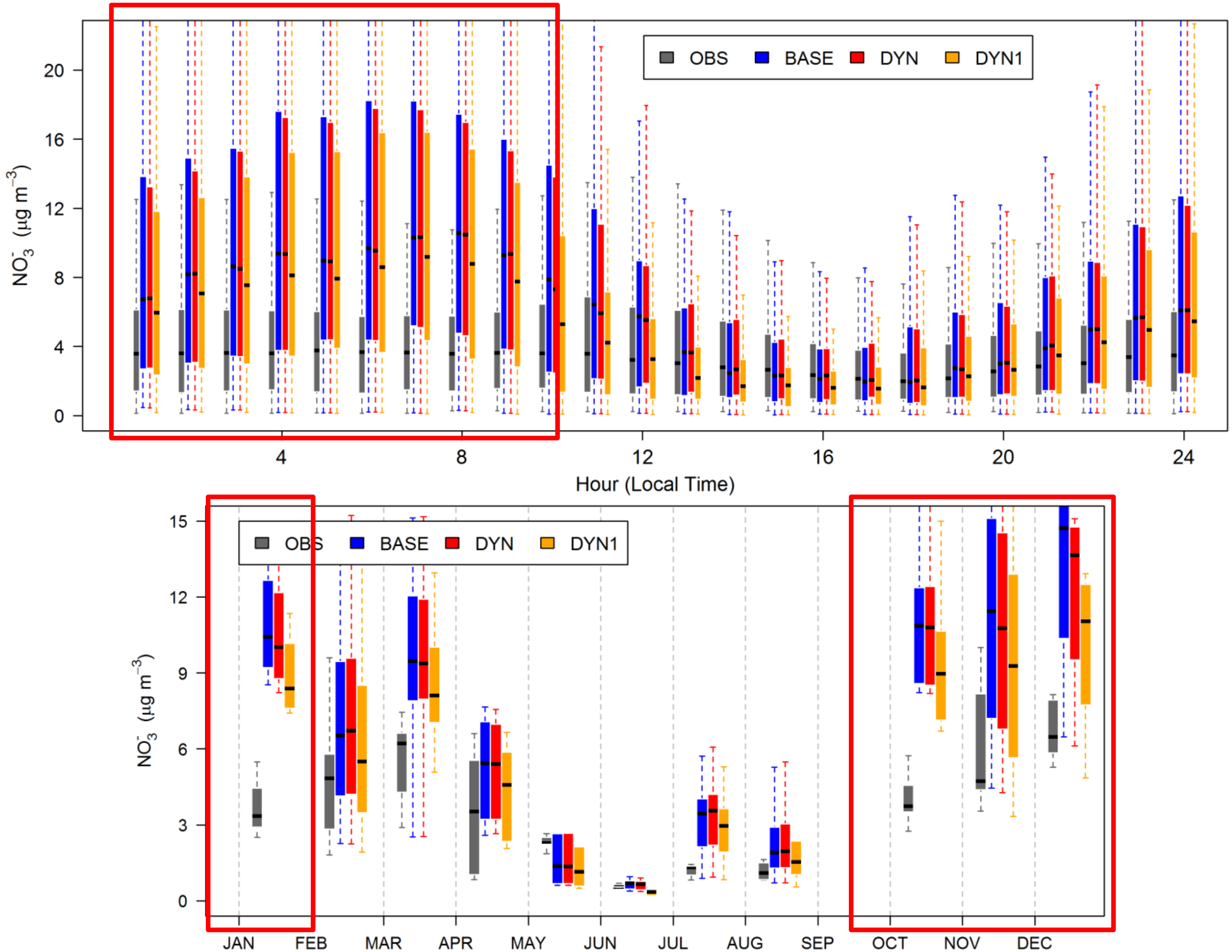
- BASE overestimate $NO_3^-$ during nighttime due to high $NH_3$.
- DYN1 still over-predict $NO_3^-$ → overestimate of NOx and $NH_3$



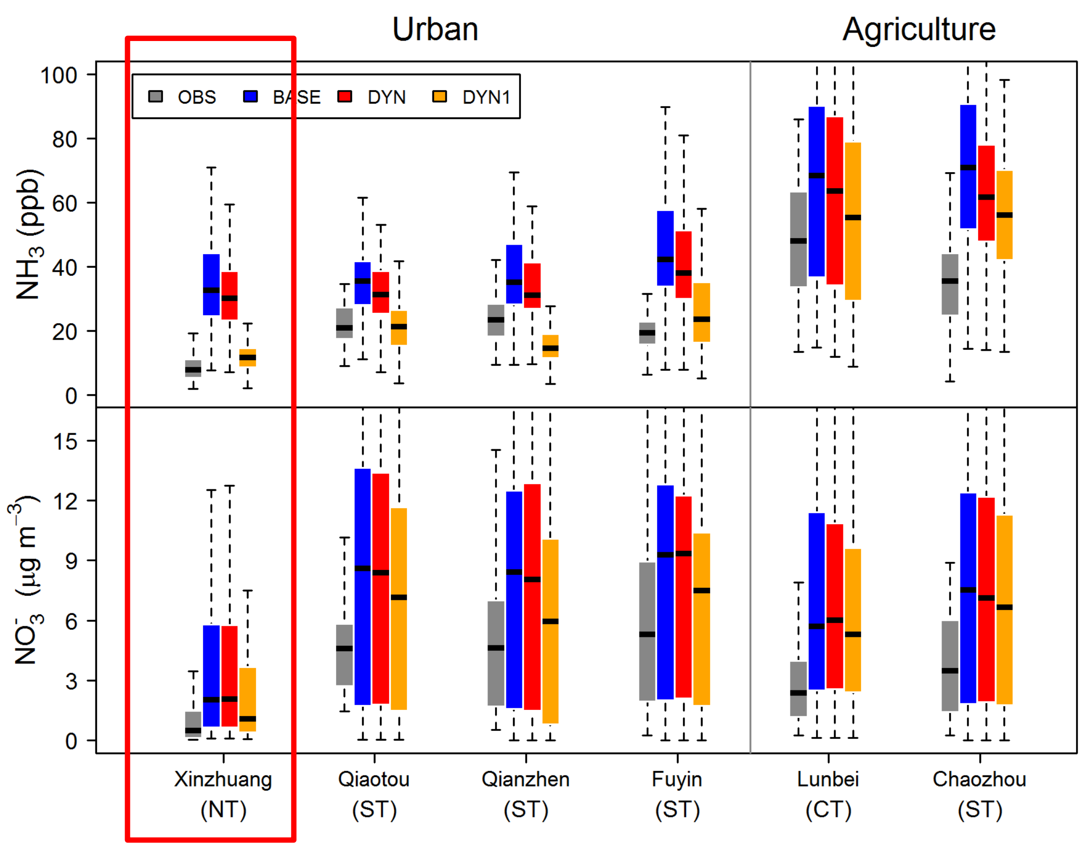
# Result – Monthly and diurnal variation of $NO_3^-$

- BASE run overestimate $NO_3^-$ during nighttime due to high $NH_3$.
- Large bias occurs from October to January



# Overall simulation result of $NO_3^-$ and $NH_3$

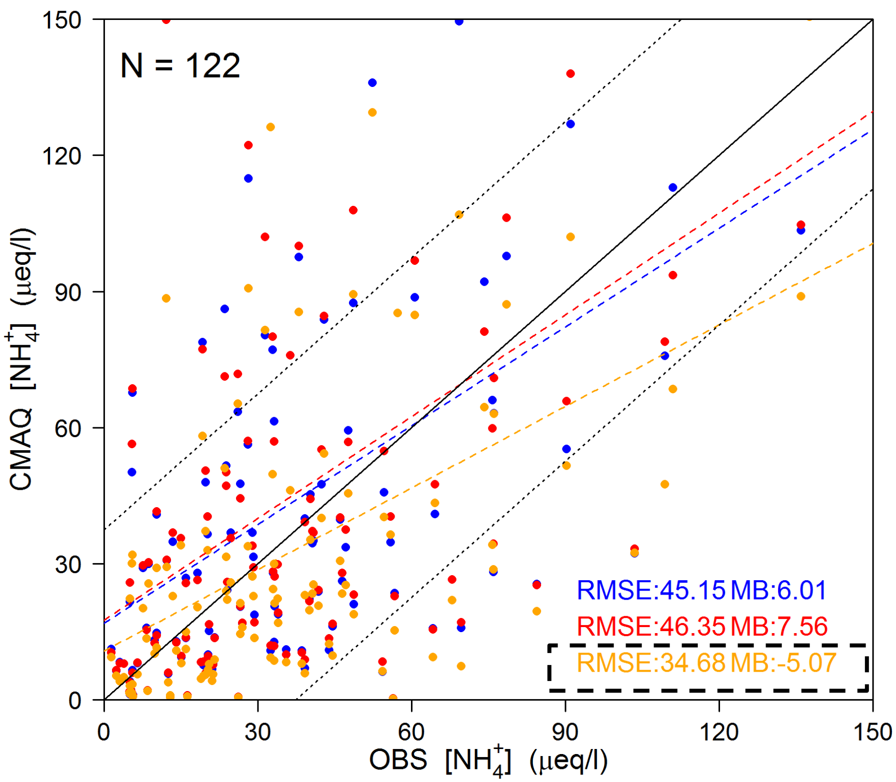
- $NO_3^-$ shows large difference in urban, particular in northern Taiwan.
- In agricultural region, $NO_3^-$ did not show large difference.
- The dynamical $NH_3$ treatment has little impact on $SO_4^{2-}$ concentration.



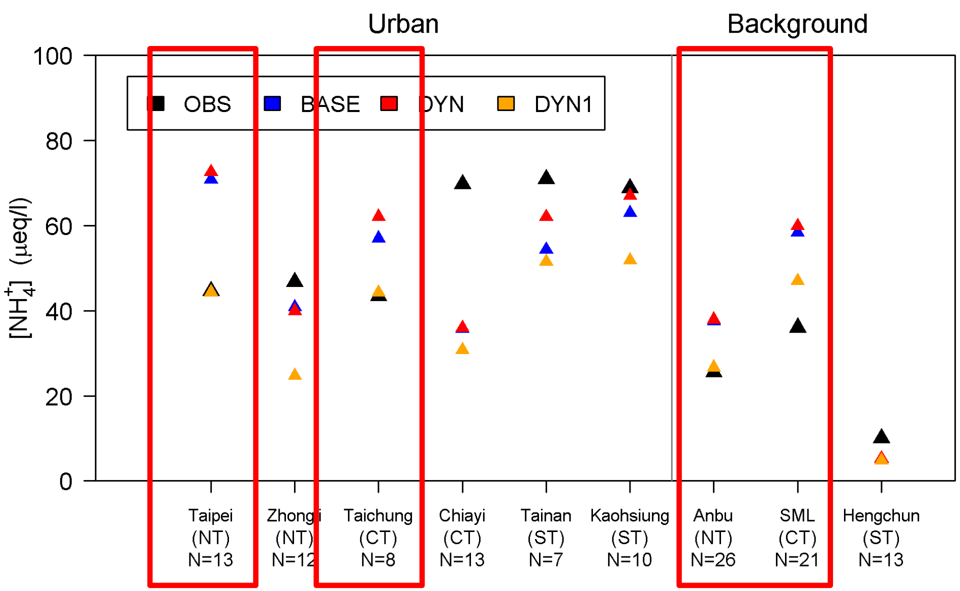

| | Average | RMSE |
|---|---|---|
| $NO_3^-$ ($\mu g/m^3$) | | |
| OBS | 4.01 | - |
| BASE | 7.84 | 10.56 |
| DYN | 7.71 | 10.35 |
| DYN1 | 6.50 | 8.97 |
| $SO_4^{2-}$ ($\mu g/m^3$) | | |
| OBS | 7.05 | - |
| BASE | 3.89 | 7.23 |
| DYN | 3.86 | 7.13 |
| DYN1 | 3.64 | 6.83 |

# Model and observed $NH_4$ wet deposition comparison

- Only the data with $0.25 \le \frac{P_{mod}}{P_{obs}} \le 4$ are used in comparison.
- BASE and DYN overestimate $NH_4$ wet deposition, DYN1 reduce the bais.
- Large improvement occurs in northern Taiwan (Taipei, Anbu) and SML sites.
- In the central to southern Taiwan, model tend to underestimate $NH_4$ wet deposition.




| | RMSE | MB |
|---|---|---|
| BASE | 45.15 | 6.01 |
| DYN | 46.35 | 7.56 |
| DYN1 | 34.68 | -5.07 |



# Summary

- Implementing dynamical $NH_3$ emission parameterization :

→ Improve diurnal and seasonal variation of $NH_3$ and nitrate

→ Reduced positive bias of nitrate

- In the DYN1, $NH_3$ show good agreement in the urban region and simulated $NH_4$ wet deposition is improved → too much urban sewage emission

- DYN and DYN1 still overestimate $NH_3$ in agricultural region

→ Overestimated of livestock emission ?

→ Other crop fertilizer and crop emission treatment

→ Apply $NH_3$ bi-directional model to better simulate $NH_3$ land-atmosphere interaction in the agriculture region

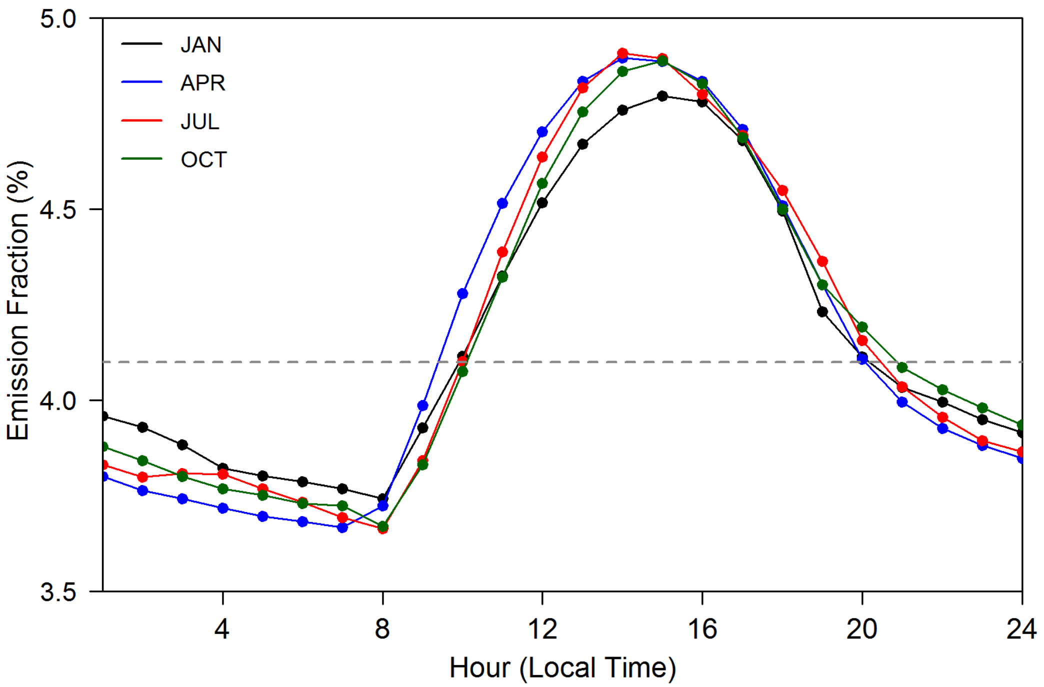
- Need process-base model or inverse modeling to provide a better $NH_3$ emission inventory in Taiwan.

The End

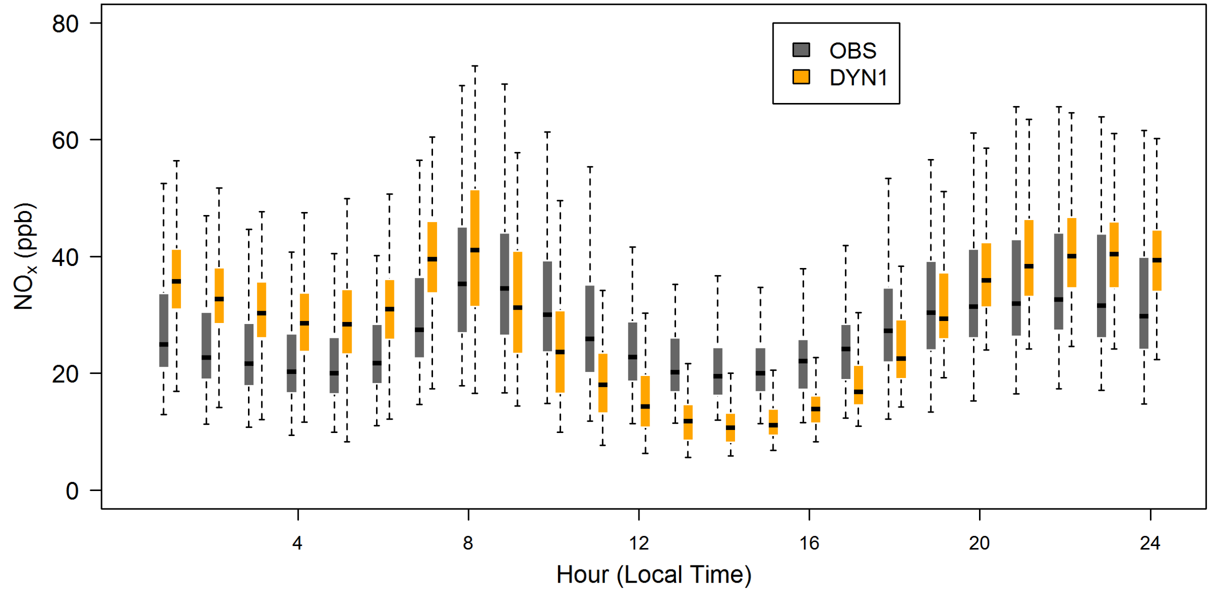
Q & A

- Why urban (household) sewage emission is overestimated ?
  → super high emission factor (50.4 kg/$10^6$ L) ~ 50.4 g/$m^3$

- Why DYN1 still over-predict $NH_3$ during night time?
  → diurnal profile
  → low PBLH during night time ( < 30 meter )



| | | Average | R | MB | NMB(%) | NME (%) | RMSE |
|---|---|---|---|---|---|---|---|
| **T2M ( °C)** | | | | | | | |
| | OBS | 23.14 | - | - | - | - | - |
| | WRF | 22.80 | 0.97 | -0.34 | -1.13 | 5.39 | 1.51 |
| **WS10 (m/s)** | | | | | | | |
| | OBS | 1.53 | - | - | - | - | - |
| | WRF | 2.38 | 0.54 | 0.85 | 139.14 | 153.22 | 1.45 |
| **$PM_{2.5}$ (μg/m$^3$)** | | | | | | | |
| | OBS | 35.68 | - | - | - | - | - |
| | STD | 26.79 | 0.54 | -8.89 | -15.59 | 57.13 | 24.78 |
| | DYN | 26.65 | 0.54 | -9.03 | -15.83 | 57.07 | 24.71 |
| | DYN1 | 24.31 | 0.54 | -11.37 | -22.82 | 55.94 | 24.26 |
| **$SO_2$ (ppb)** | | | | | | | |
| | OBS | 4.73 | - | - | - | - | - |
| | STD | 3.80 | 0.29 | -0.93 | -3.15 | 77.66 | 4.66 |
| | DYN | 3.80 | 0.29 | -0.92 | -2.94 | 77.70 | 4.66 |
| | DYN1 | 3.90 | 0.29 | -0.83 | 0.19 | 77.52 | 4.64 |
| **NOx (ppb)** | | | | | | | |
| | OBS | 27.70 | - | - | - | - | - |
| | STD | 29.64 | 0.49 | 1.94 | 23.46 | 71.88 | 24.73 |



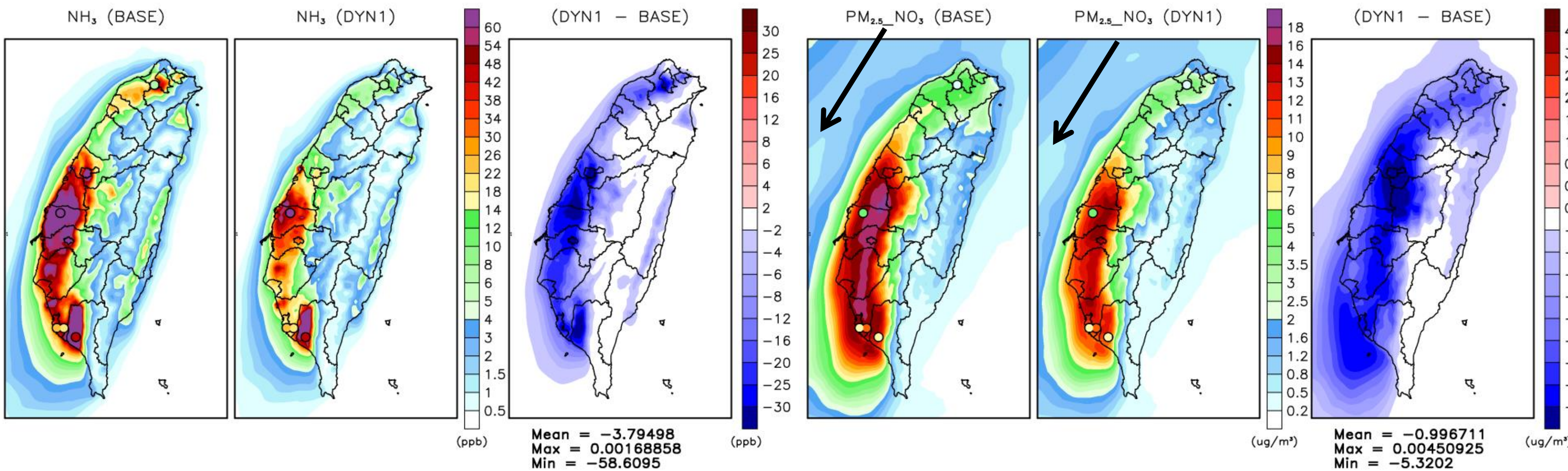
Why DYN1 still over-predict nitrate during winter ?

→ Excessive precursor ( $NH_3$, NOx)

→ Meteorological condition ( low PBLH, stagnant wind field)

**Oct - Dec**

**$NH_3$** **Nitrate**



# Model and observed $PM_{2.5}$ speciation data comparison

- Observed $NH_4^+ = \frac{(SO_4^{2-} \times 2)}{96} + \frac{NO_3^-}{62}$
- BASE run overestimate $NO_3^-$ ($NH_4^+$) during nighttime due to high $NH_3$ value.