Evaluation of bi-directional ammonia exchange in GEOS-Chem using in-situ observations

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Impact of NH_3

- Human health impact
- $PM_{2.5}$ causes **bronchitis**, **asthma**, **premature mortality**…
- Environmental impact

- **Haze**, decreases visibility**.**
- **Eutrophication** of surface waters:
  - Alga blooms;
  - Hypoxia;
  - Cloudy, colored water.
- **Soil acidification:**
  - Nitrification of NH_4^+ into NO_2^-, releasing H^+.

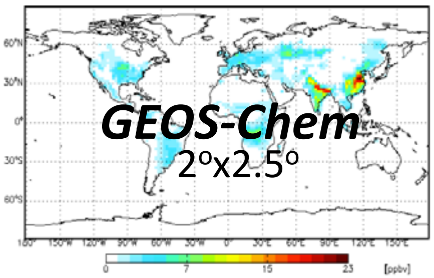
Smog

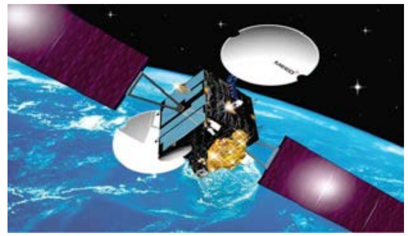
Eutrophication

Soil acidification

Large uncertainties in NH_3 inventories.

Assimilating remote sensing NH_3 from TES





Modeled NH_x emissions

Modeled NH_x con., NO_3^- con.

VS

Measured NH_x con., NO_3^- con.

- 4D-Var inversion (GEOS-Chem adjoint) to adjust NH_3 emissions.
- Prior NH_3 profiles lower than TES retrieval.
- Optimized NH_3 profiles increased towards TES retrieval.
- Optimized model still underestimates TES retrievals.

Zhu et al., 2013

Evaluation: model NH_3 vs AMoN obs

al

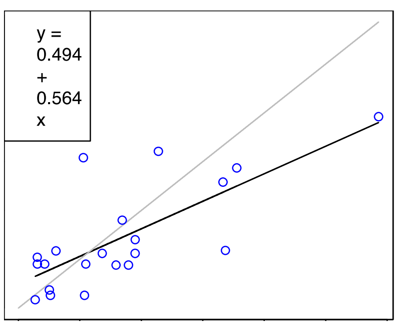
April

July

GC (ppb)

GC (ppb)

October



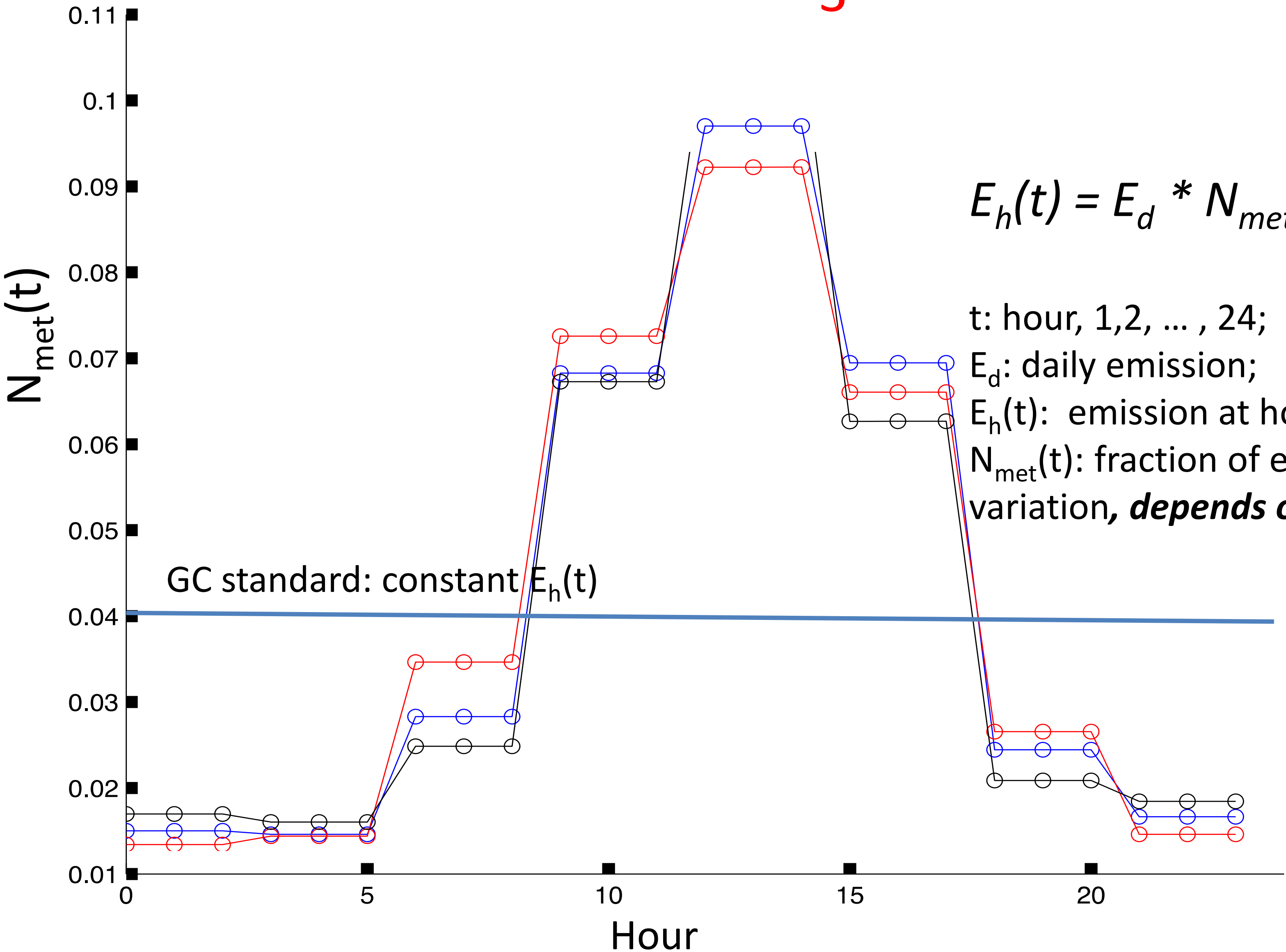
AMoN (ppb)

AMoN: 21 sites with 2-week long observation, Middle & Eastern US

- Slopes are all close to unity.
- R^2 all increased.
- In July, the model estimates are biased high.
- Possible reasons for July bias:
  - TES overpass time (1:30 at day and night) lead interest to diurnal variability of model NH_3 (Similar diurnal variability issue in CMAQ [Jeong submitted].)
  - Bi-directional exchange.

Zhu et al., 2013

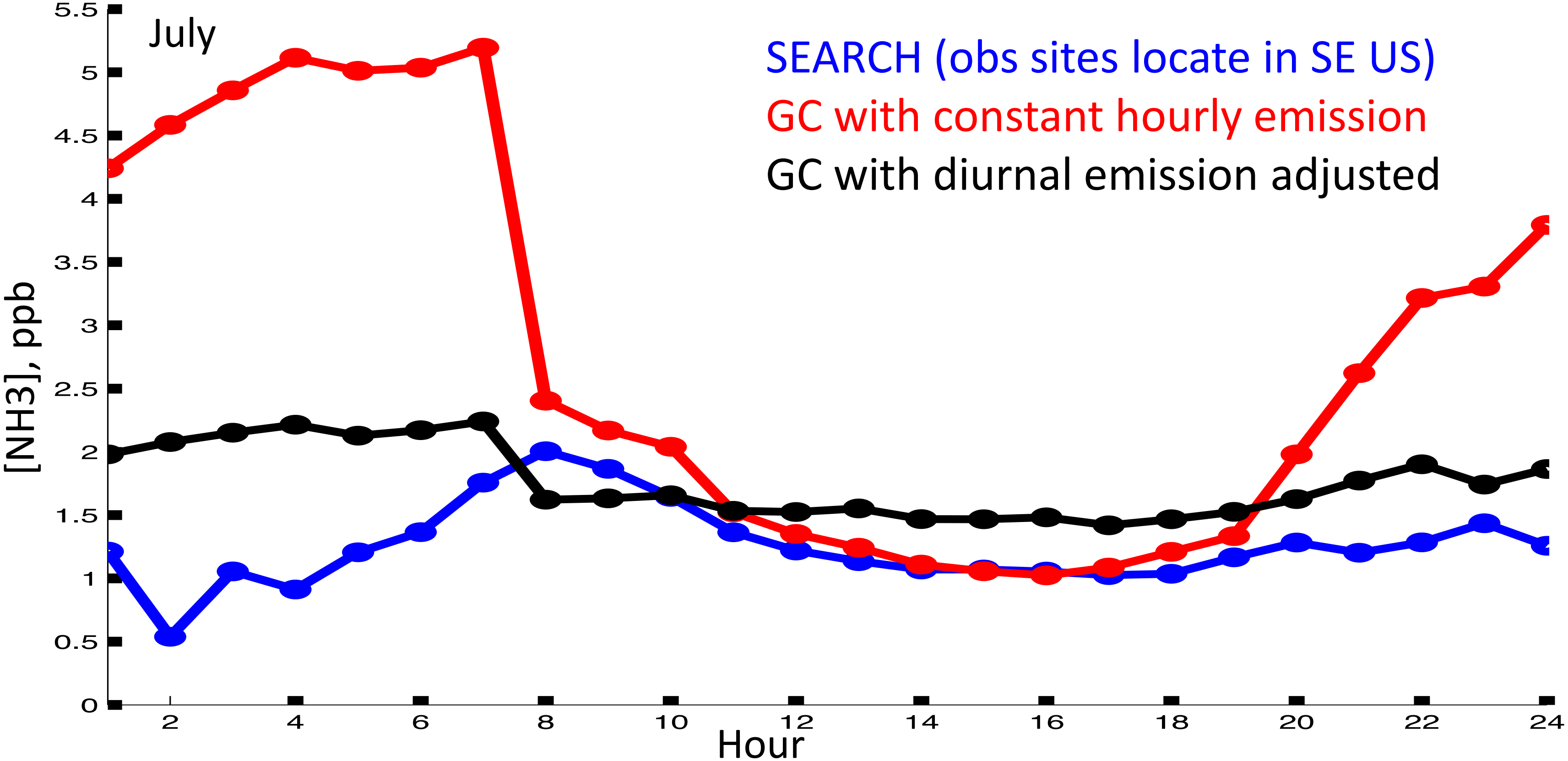
Implementing diurnal variability for livestock NH_3 emissions



$$E_h(t) = E_d * N_{met}(t)$$

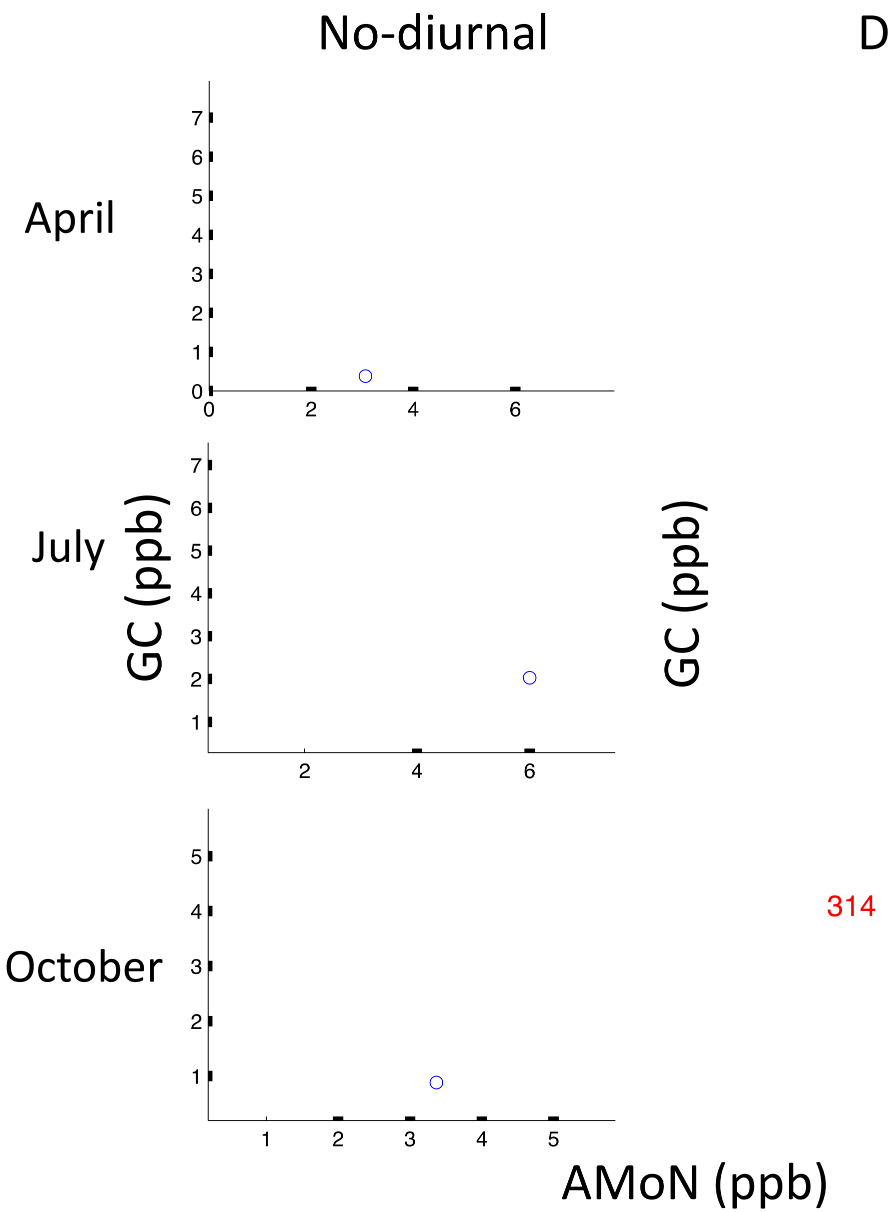
t: hour, 1,2, … , 24;
E_d: daily emission;
$E_h(t)$: emission at hour t;
$N_{met}(t)$: fraction of emission diurnal variation***, depends on T, $R_{aerodynamic}$***

Surface NH_3 diurnal variability



- NH_3 decreased at night by several ppb; increased in day up to 1 ppb.
- Monthly average surface NH_3 (and NO_3^-!) decreased.
- NH_3 concentration (at TES overpass time 13:30) can be impacted without changing total emissions.
- Improves TES assimilation results compared to Zhu et al. 2013.

Diurnal variation impact: model NH_3 vs AMoN obs



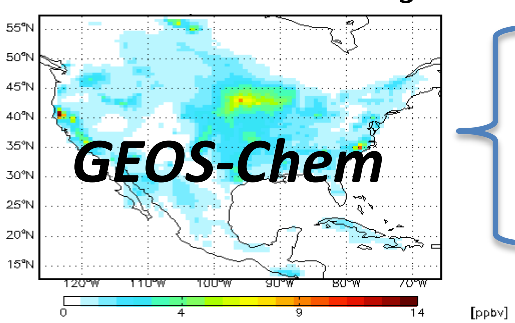
AMoN: 21 sites with 2-week long observation, Middle & Eastern US

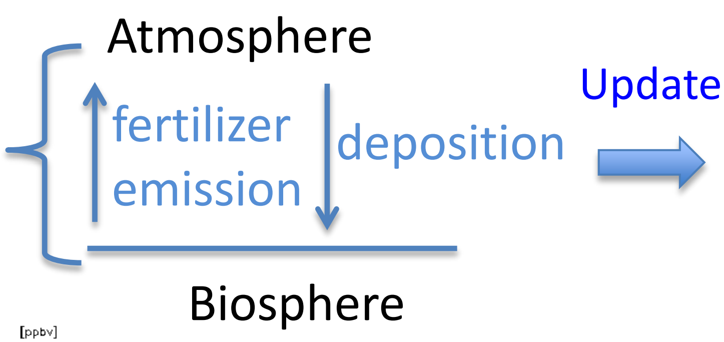
- R^2 increased in July.
- Slope decreased due to NH_3 monthly average decreased in July.

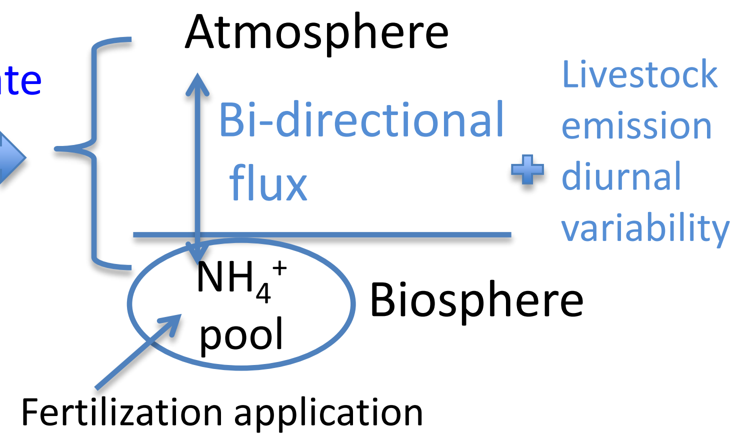
Process based treatment of NH_3 sources

- Bidirectional surface exchange between atmosphere & biosphere is key part of NH_3 cycle
- Most current air quality models (e.g., GEOS-Chem) do not account for:
  - bi-directional surface fluxes of NH_3
  - NH_4^+ pool in soil
  - diurnal variability of livestock emissions

Surface level NH_3



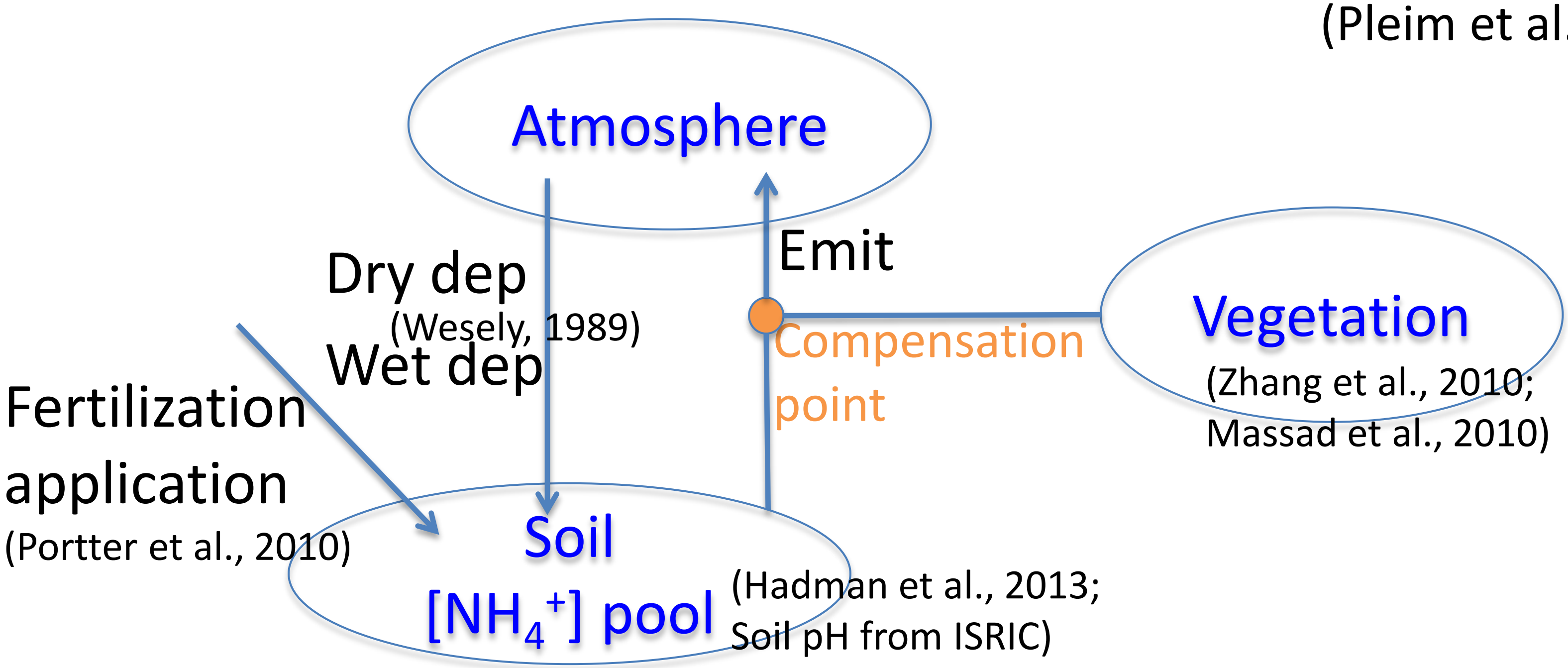




- Update GC to include these 3 processes
- Applied to:
  - 0.5° x 0.667°, GEOS-5: April, July, October of 2008
  - Focus on US domain

Bi-directional exchange schematic

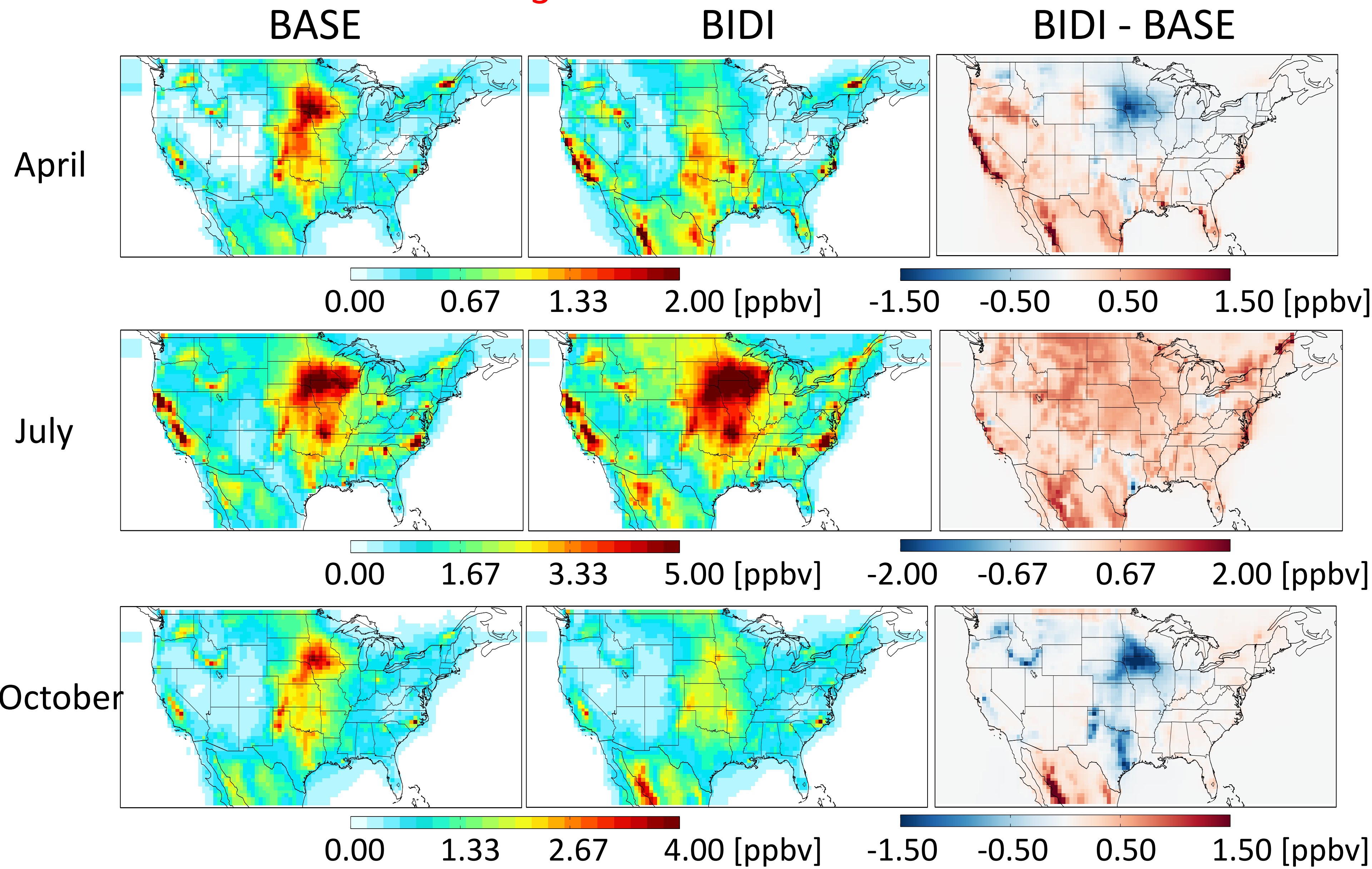
(Pleim et al., 2013)



- $[NH_4^+]_{soil}$ reflects instant changes due to fertilization application and bi-directional exchange.
- $[NH_3]_{compensation\ point}$ reflects instant changes of $[NH_3]$ in atmosphere, soil & vegetation.
- Overall impact: NH_3 lifetime will be effectively extended.

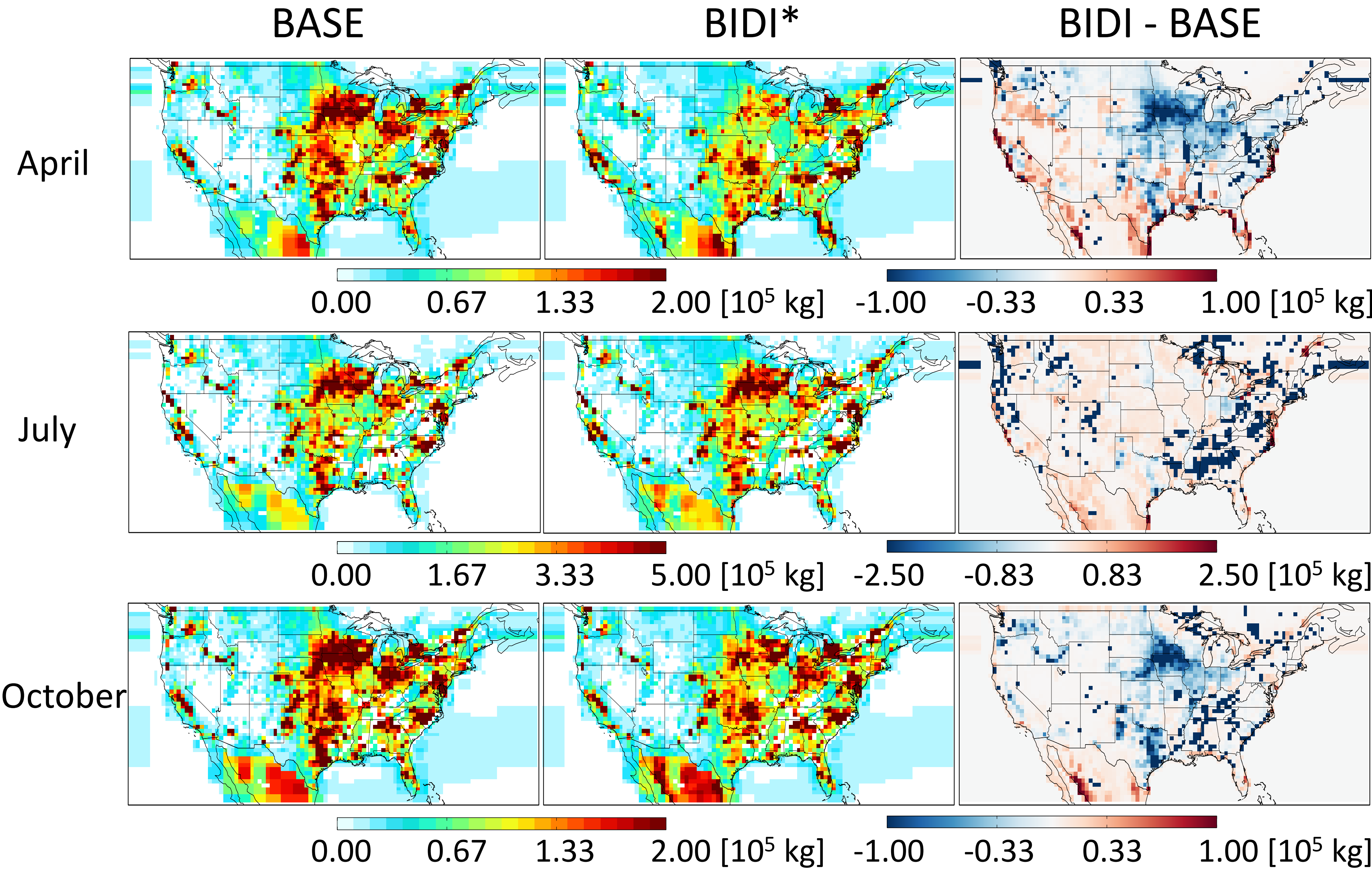
References: Cooter et al., 2010; Zhang et al., 2010; Massad et al., 2010; Pleim et al., 2013; Bash et al., 2013; Hadman et al., 2013; Portter et al., 2010.

NH_3 surface concentration



Changes similar as CMAQ (Jeong et. al, 2013, submitted)

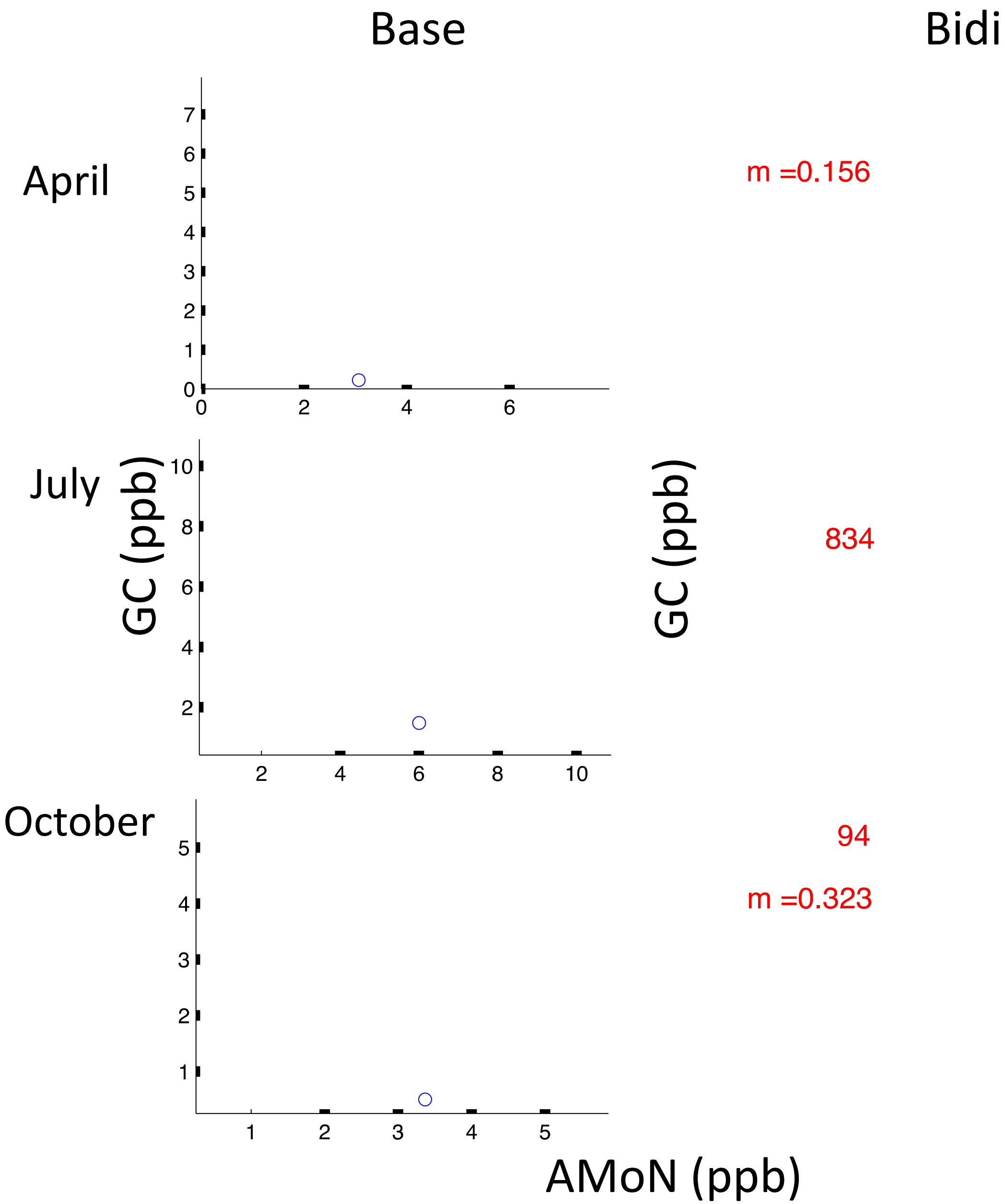
NH_3 emission



BIDI*: similar as CMAQ (Jeong et al., 2013, submitted)

Changes in emission = upward flux from soil and vegetation

BIDI exchange impact: model NH_3 vs AMoN obs



AMoN: 21 sites with 2-week long observation, Middle & Eastern US

- BIDI NH_3 increased slightly in July, improve the R^2 and the slop.
- BIDI NH_3 decreased in some sites in April & October.
- BIDI improvement depend on locations.

BIDI exchange impact: model NH_3 vs AMoN obs

Optimized (Zhu et al 2013)

Bidi with optimized emissions

8

m =0.163

April

July

GC (ppb)

GC (ppb)

October

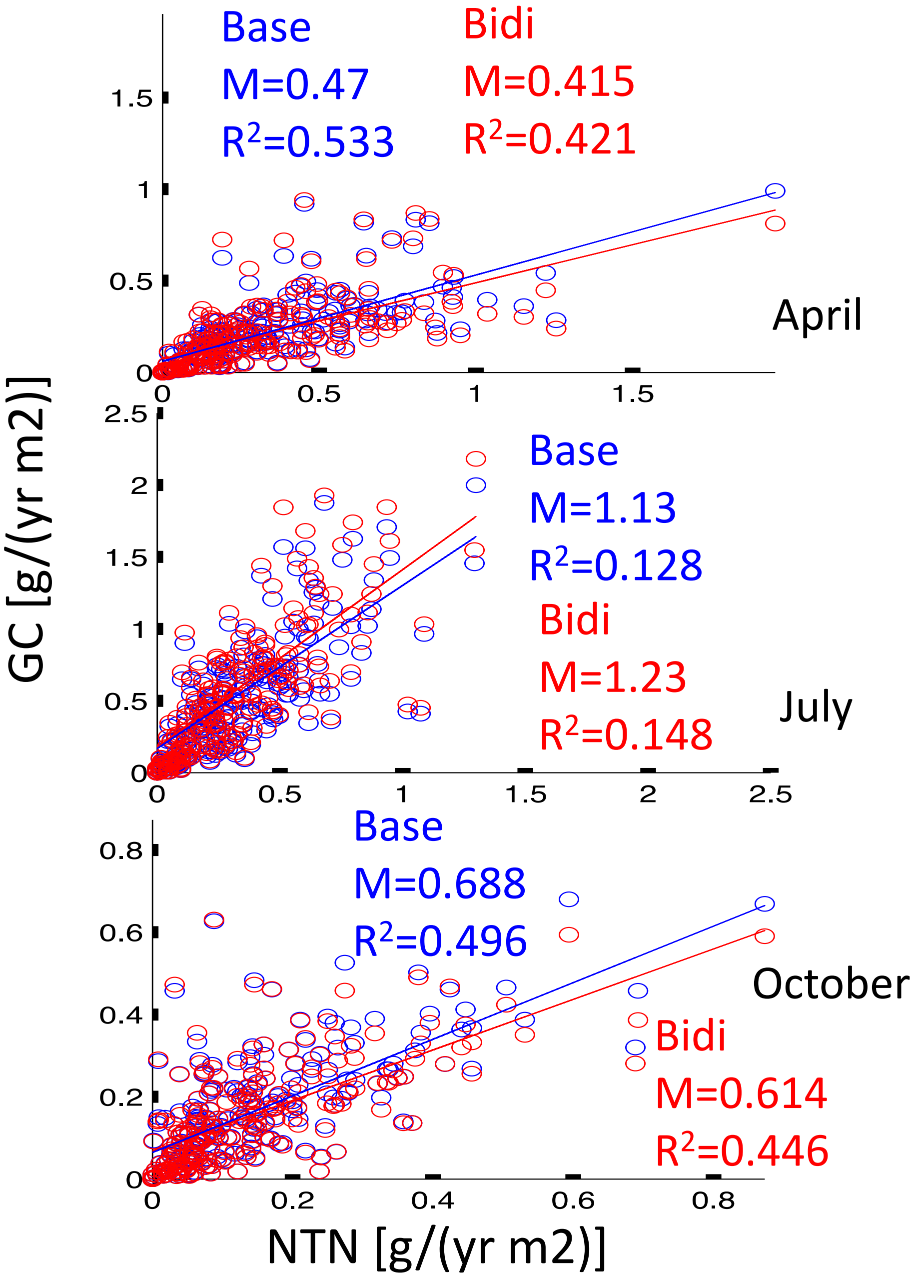
604

m =0.404

AMoN (ppb)

- Replacing the emission inventory in BIDI with optimized emissions from Zhu et al 2013.
- NH_3 decreased in all three months.
- BIDI decreased the NH_3 overestimate in July.
- TES assimilation with BIDI is in progress.

BIDI exchange impact: model NH_x vs NTN obs



NTN: > 200 sites in US, week long observation

- Bidi NH_3 not improved the slope.
- Bidi NH_3 still overestimate in July.

Conclusions

- Bi-directional exchange increased the NH_3 monthly mean concentration in July through out U.S., decreased in North and Middle U.S. in April & October.
- Spatial distribution differences between the BASE and BIDI are similar as CMAQ.
- Bi-directional exchange's impact mainly depending on the locations, will try case studies in fertilizer emission dominated region (e.g., Central Great Plain, South Texas, California).
- Data sources of important parameters (e.g., soil pH, fertilizer application rate) in bi-directional exchange need to be further investigated.
- Changes to NH_3 from bi-directional exchange need to be further resolved with results from top-down constraints (Zhu et al., 2013).

Thanks !

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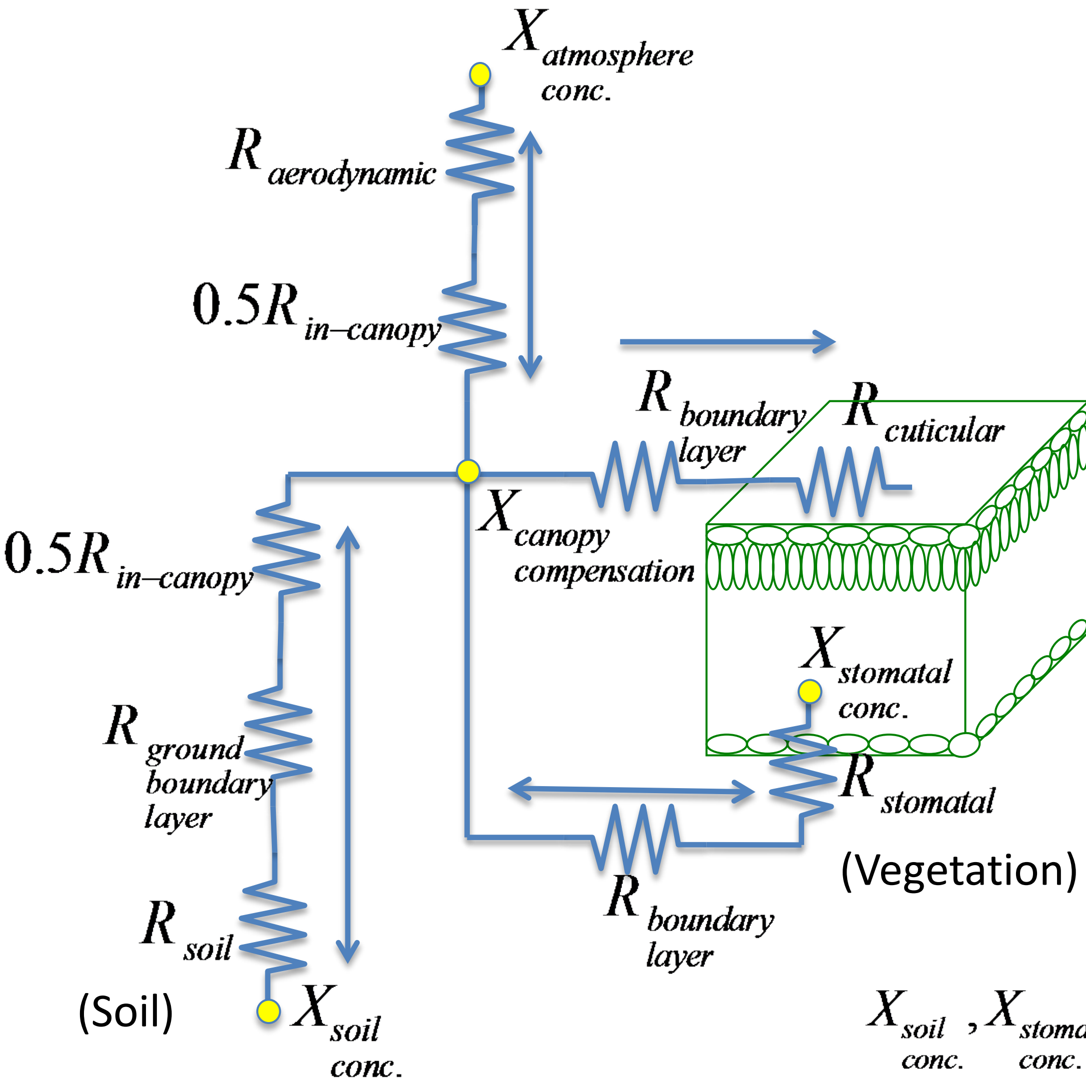
Bi-directional exchange schematic



(Pleim et al., 2013)

$$F_{total} = \frac{X_{\substack{canopy\\compensation}} - X_{\substack{atmosphere\\conc.}}}{R_{aerodynamic} + 0.5R_{in-canopy}}$$

- Direction of flux:

$$X_{\substack{canopy\\compensation}} \leq X_{\substack{atmosphere\\conc.}} : \text{deposition}$$

$$X_{\substack{canopy\\compensation}} > X_{\substack{atmosphere\\conc.}} : \text{emission}$$

Emission potential: $\Gamma = \dfrac{[NH_4^+]}{H^+}$

$$X_{\substack{soil\\conc.}}, X_{\substack{stomatal\\conc.}} = \frac{A}{T_{soil,stomatal}} \exp\left(-\frac{B}{T_{soil,stomatal}}\right) \Gamma_{soil,stomatal}$$

$$A = 2.7457 \times 10^{15}, B = 10378$$