Process Analysis of CMAQ v5.0 With and Without Bidirectional NH_3 Exchange

Gill-Ran Jeong[1], Jesse Bash[2],
Daven Henze[1], and Rob Pinder[2]

[1.] Department of Mechanical Engineering,
University of Colorado, Boulder
[2.] Atmospheric Modeling and Analysis Division,
NERL, US EPA

Motivation

NH_3 affects the earth environment :

- neutralizing acidic gases
- increasing atmospheric PM in fine mode
- degrading visibility → air pollution.

- inducing acidification of water body
- increasing algal growth
- degrading estuaries → water pollution.

playing an important role in
the nitrogen cycle of the earth system.

- NH_3 bidirectional mechanism arises from the thermodynamics and ubiquity of these substances in environment media due to wide scale fertilizer application.
- NH_3 bidirectional exchanges change deposition and ambient concentrations.

Physical Process Analysis :

- What are the governing processes that reflect NH_3 bidirectional exchange?
- How does the bidirectional mechanism affect the atmospheric lifecycle of NH_3?

Objectives and Goals

The objectives are

- to observe how the NH_3 bidirectional exchange changes ambient NH_3 concentrations and aerosol concentrations
- to investigate the physical processes both in base and bidi cases and how they changes due to the NH_3 bidirectional exchange
- to examine the changes in the range of influence of NH_3, horizontally and vertically

The goal of this study is

to characterize the physical processes related to NH_3 bidirectional exchange and to investigate how Process Analysis (PA) can inform the inverse modeling such as Air Mass Factor (AMF) method or Adjoint method.

Physical Process Analysis

$$\frac{\partial C_g}{\partial t} + \nabla\cdot\left(\vec{v}C_g\right) = (\nabla\cdot K_b\nabla)C_g + R_{emis} + R_{dep} + R_{washout(cloud)} + R_{het\_rxn(aerosol)}$$

advection diffusion emission dry deposition wet deposition (cloud) aerosol

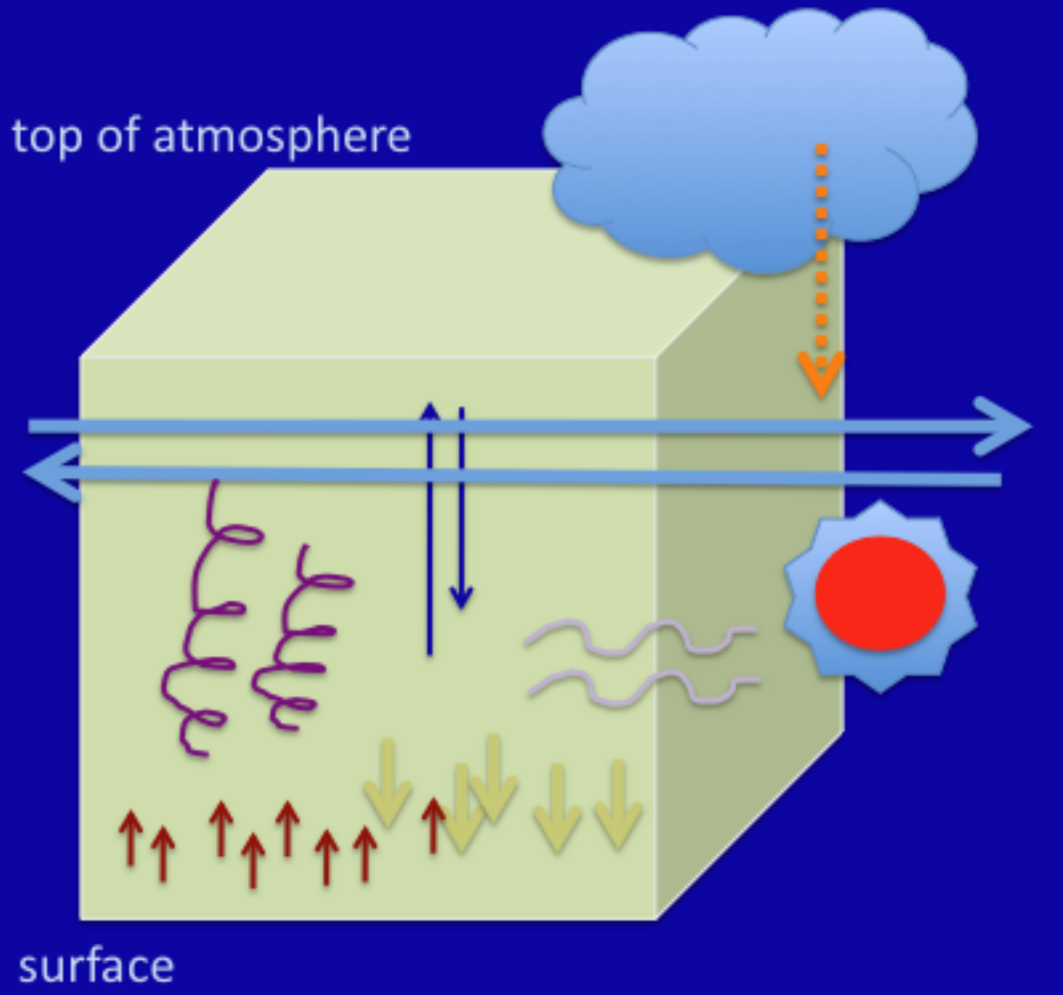
The changes in a species' concentration due to operator n.

$$(\Delta C)_n = \int_t^{t+\Delta t} L_n dt$$

L_n is the differential operator associated with a process.
Δt is the synchronized time step.

The concentration at the end of a time step

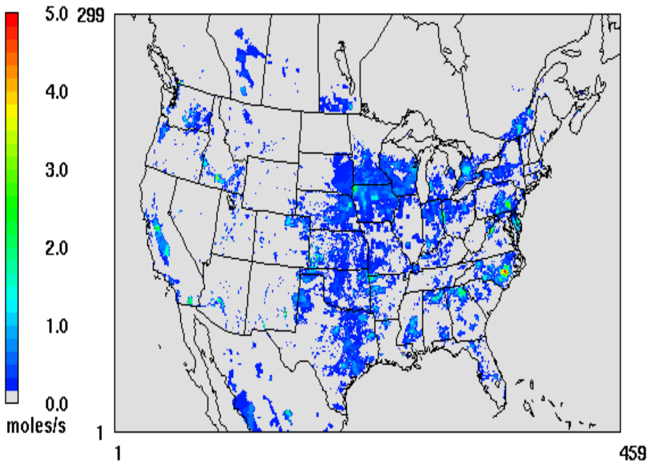
$$C(t+\Delta t) = C(t) + \sum_{n=1}^{N}(\Delta C)_n$$



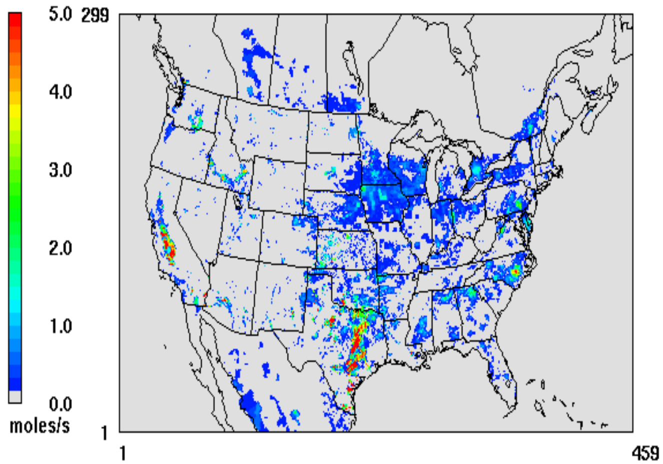
Emission Map

NH_3 emission in CONUS (August, 2009)
(NH_3 sources = agricultural fertilization + animal lots + industry)

Base

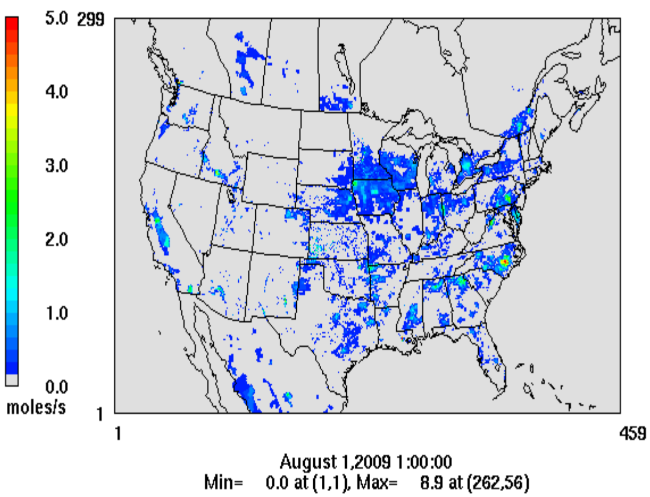


Bidi

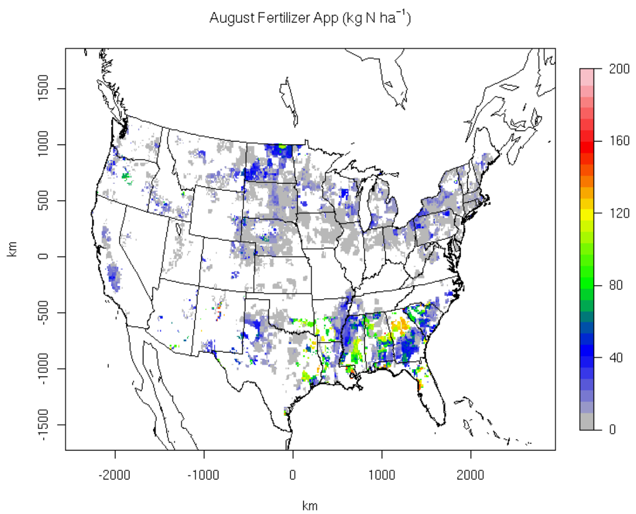


- NH_3 emissions are higher over the East US than the West US.
- Bidi emission map shows high NH_3 emission over Eastern Texas and San Joaquin valley, California.

no NH_3 Fertilization (moles/s)



NH_3 fertilization (kg N /ha)

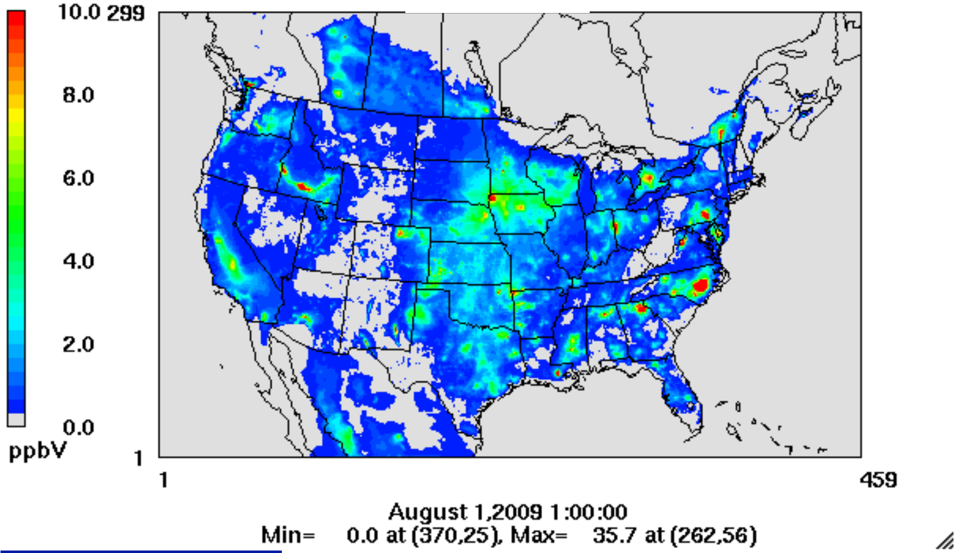


- Increased emissions correspond to fertilization application.
- In Midwest and around great lakes, animal or industry emissions are dominant.

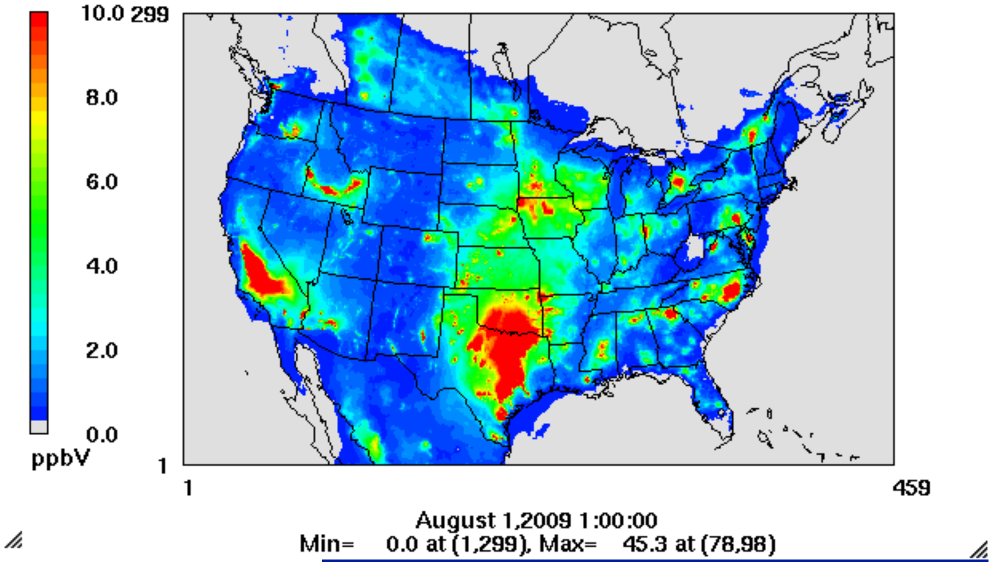
Changes in spatial distribution of NH_3 due to bidirectional mechanism

Monthly mean spatial distributions from NH_3 CMAQ output (August, 2009)

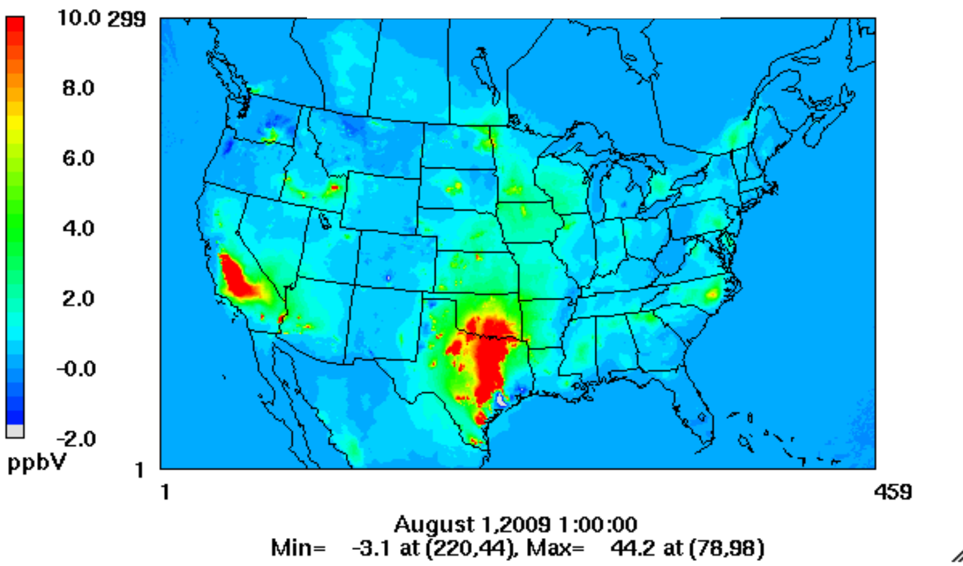
base



bidi



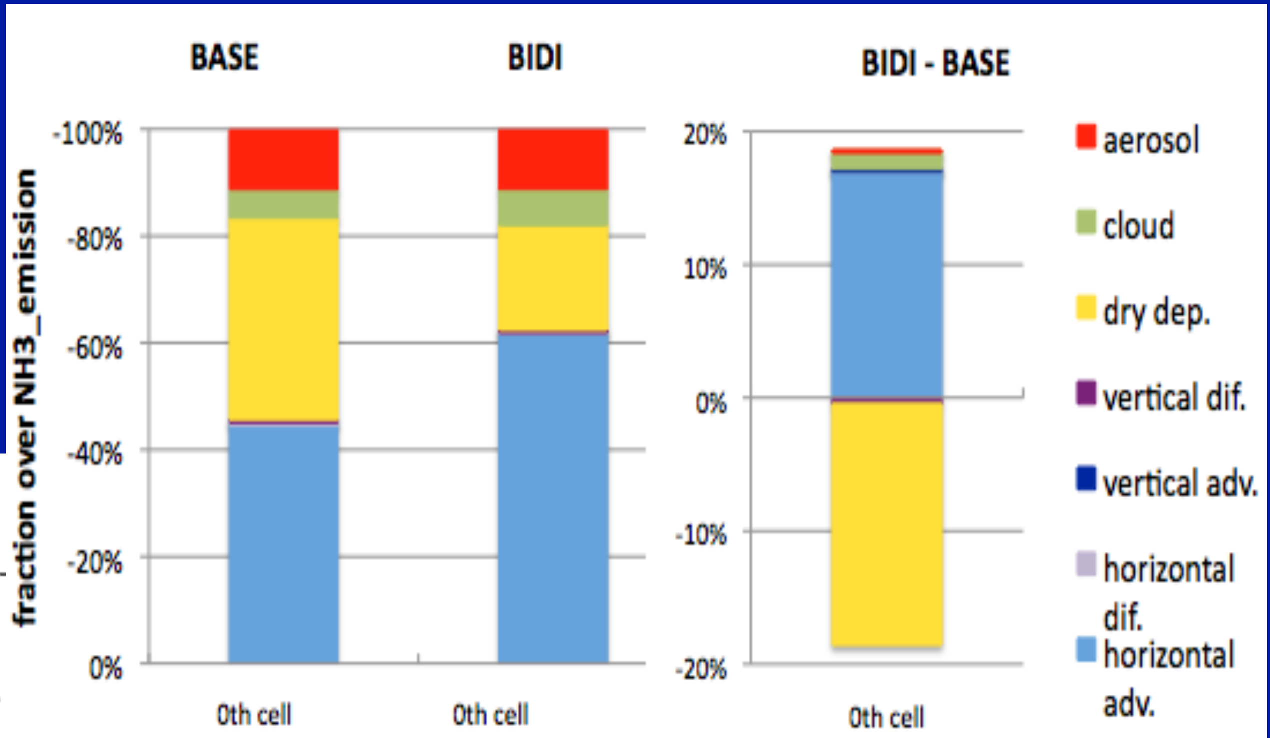
bidi - base

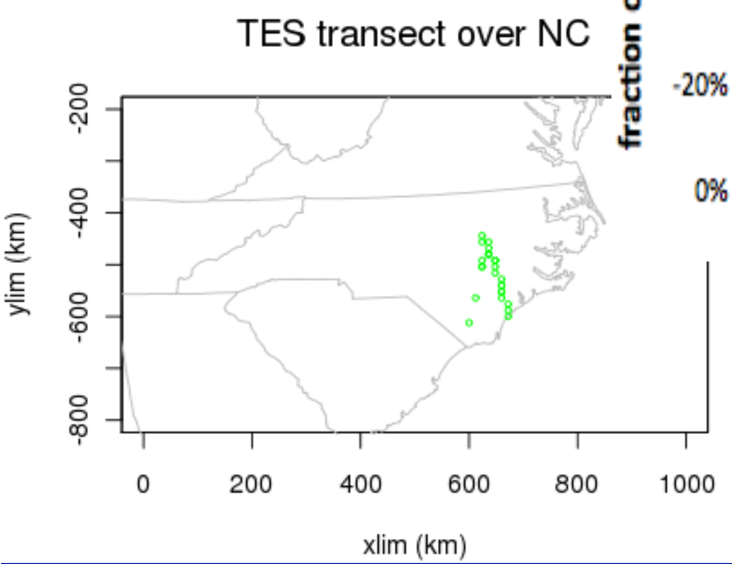


- Increases in NH_3 are present through the domain.
- The largest increases are over Texas and San Joaquin valley, California.
- In TES transect over NC shows moderate increase in NH_3.

NH_3 Physical Process Analysis in a cell

TES special observations over North Carolina August, 2009



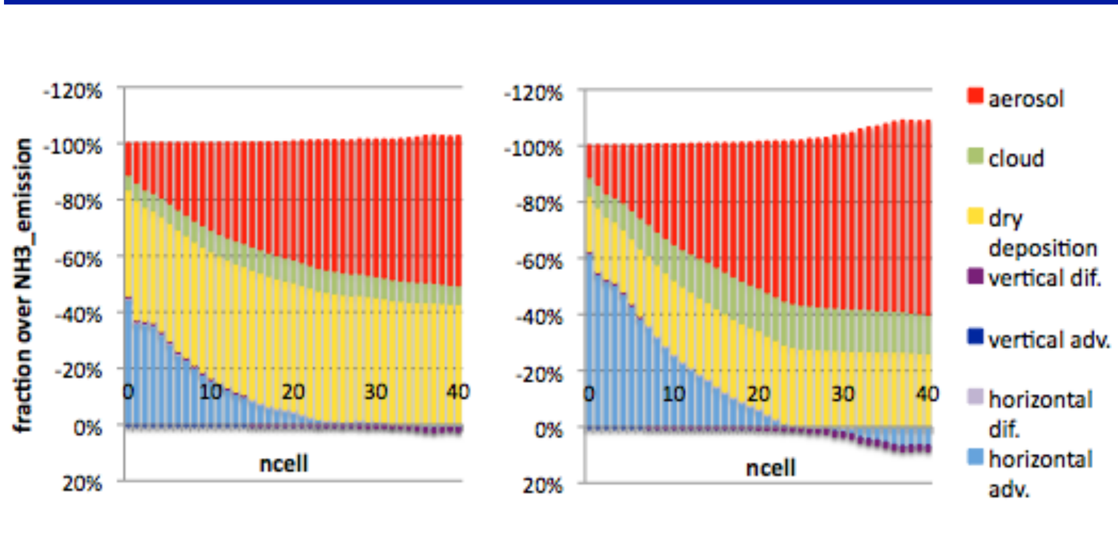


- Over a specific location, 0th cell,
- the fractions of physical processes with respect to NH_3 emission (a column averaged value)
- The loss of NH_3 emission are in order; horizontal advection > dry deposition > aerosol > cloud (wet deposition)

- When expanding the analysis from a single cell to ncell in N, S, E, and W direction ... (go next)

Fractions of Physical Processes with respect to NH_3_emission (Column Averaged Values)

BASE BIDI



NH_3

Loss process:

Dry deposition ↓

Aerosol ↑

Wet deposition ↑

Horizontal advection ↑

Fractions of Physical Processes with respect to NH_3_emission (Column Averaged Values)

BASE BIDI

NH_3

Loss process:
Dry deposition ↓
Aerosol ↑
Wet deposition ↑
Horizontal advection ↑

NHx

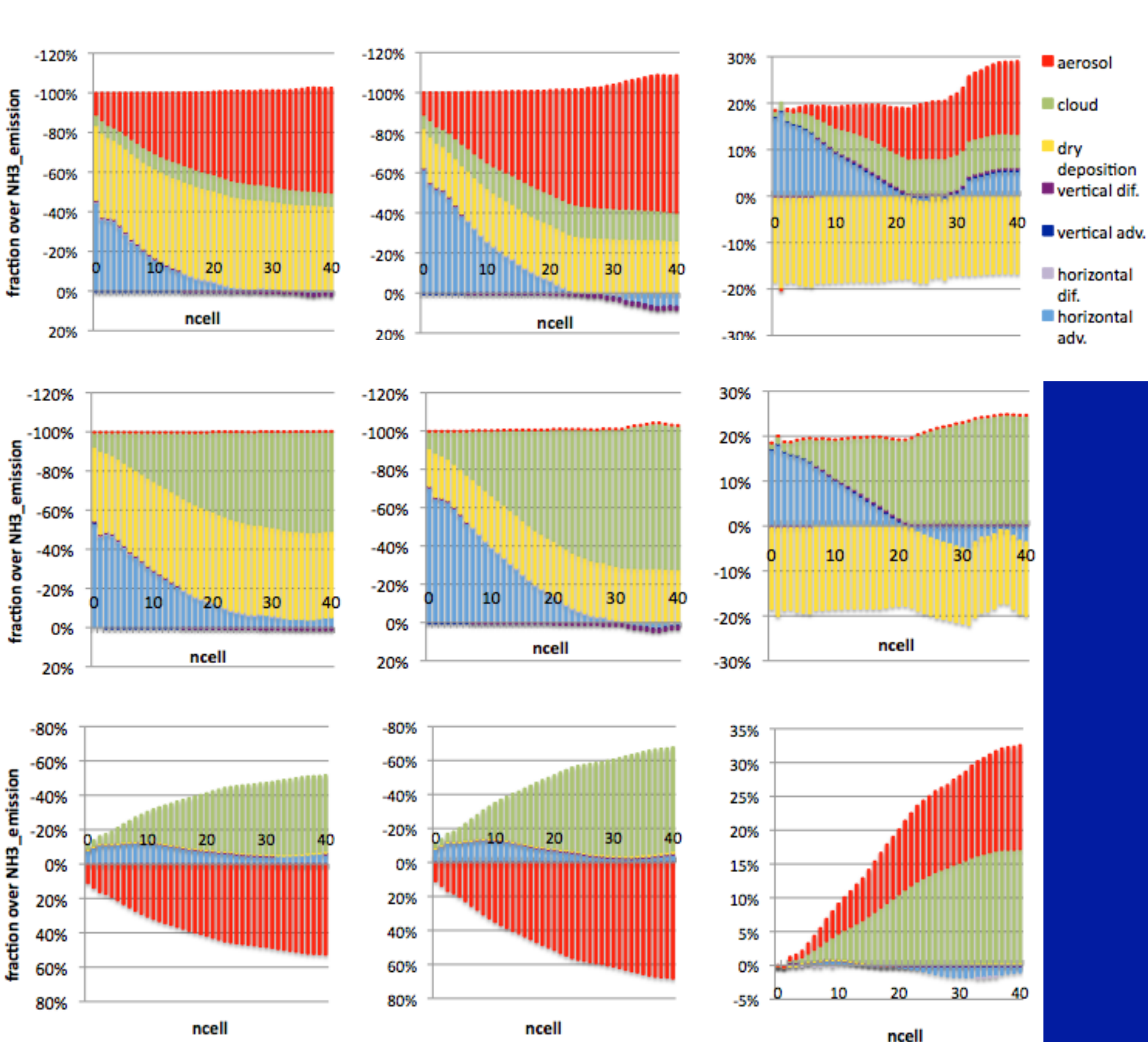
About 25th cell,
Horizontal advection
Becomes very small.

NH_4

gain process:
Aerosol ↑
Loss process:
Wet deposition ↑

Fractions of Physical Processes with respect to NH3_emission (Column Averaged Values)

BASE | BIDI | I BIDI I – I BASE I



NH_3

Loss process:
Dry deposition ↓
Aerosol ↑
Wet deposition ↑
Horizontal advection ↑

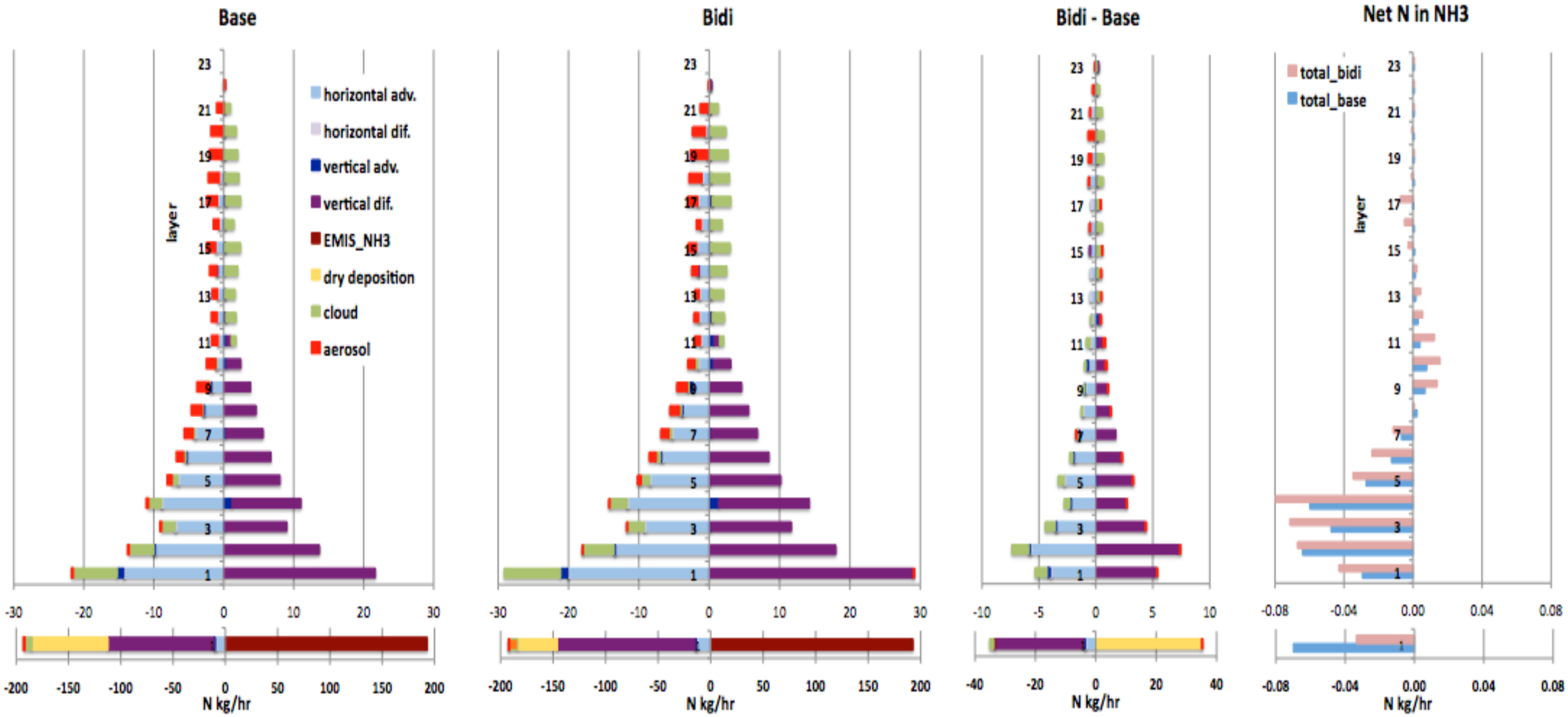
NHx

About 25th cell,
Horizontal advection
Is relatively not important.

NH_4

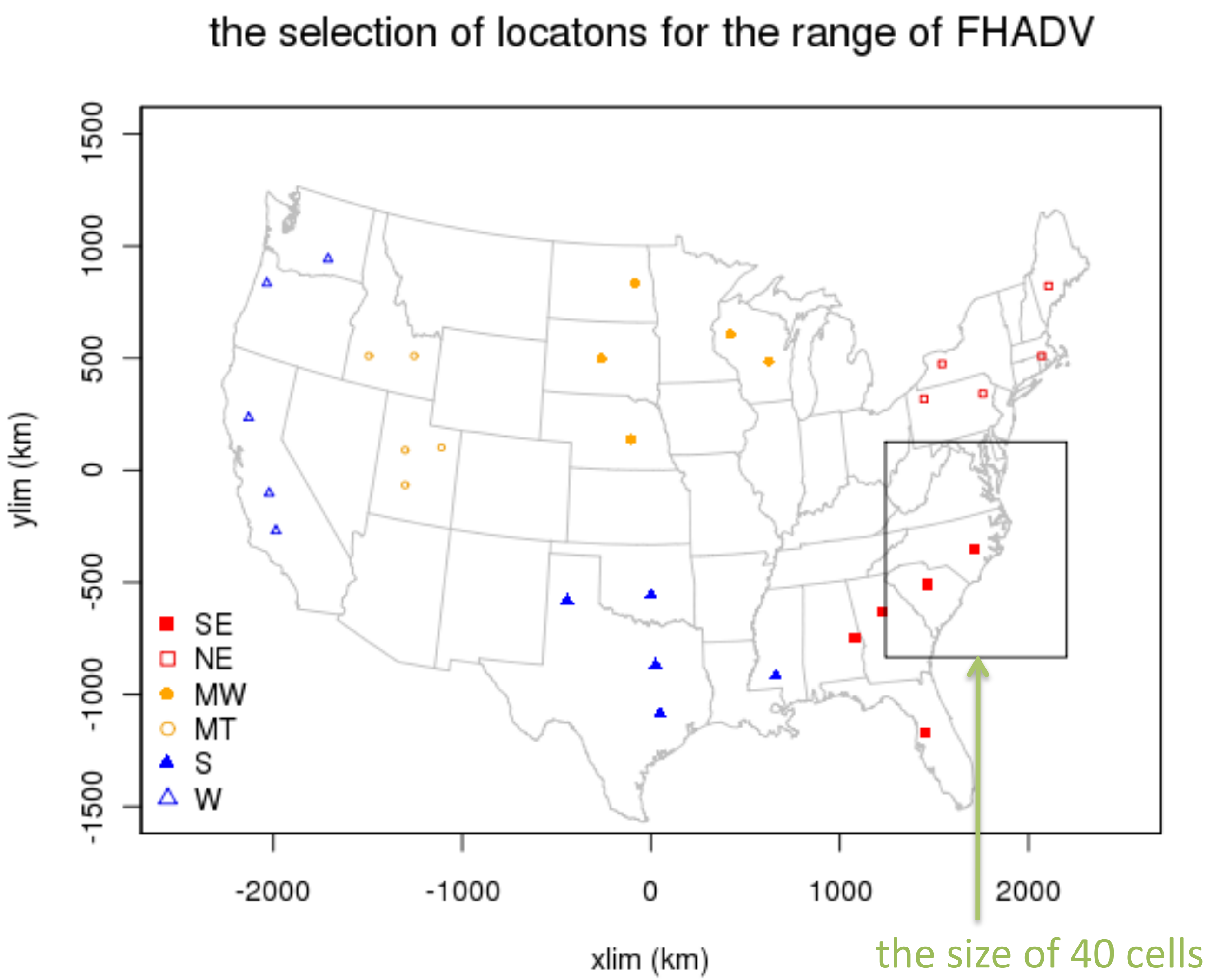
gain process:
Aerosol ↑
Loss process:
Wet deposition ↑

The vertical distribution of the fraction of individual processes over NC TES transect



- Surface layer : majority of NH_3 gain/loss occurs.
  In this local site, NH_3 loss in base case > in bidi case.
- Between 1st layer and above surface, maximum vertical gradient of N concentration.
- Above surface to 10th layer (about PBL), gain due to diffusion, loss due to horizontal advection
  ➔ NH_3 pumping (due to diffusion) and transported out NH_3 due to horizontal advection.
- Above PBL, aerosol gain/loss processes are dominant.
- Bidirectional exchange magnifies this phenomena.

How far is the range of horizontal advection processes due to bidirectional exchange?



FHADV :
the fractions of horizontal advection over NH_3 emission

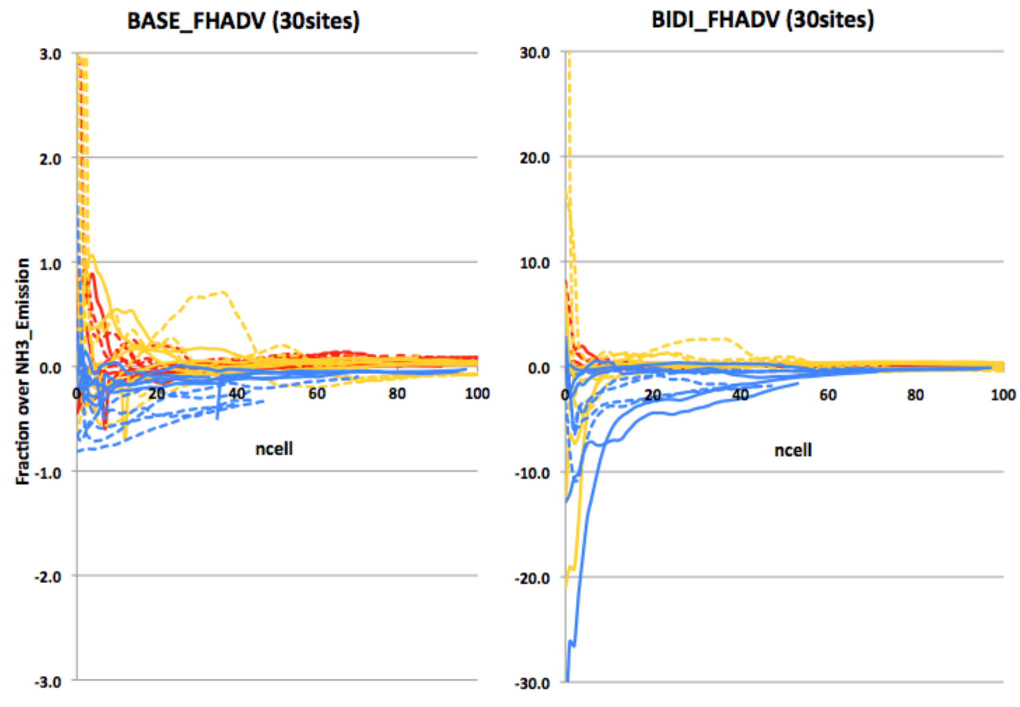
6 regions were selected based on fertilization area and geographical location

- SE: South East US
- NE: North East US
- MW : Midwest US
- MT: Mountain US
- S: South US
- W: West US

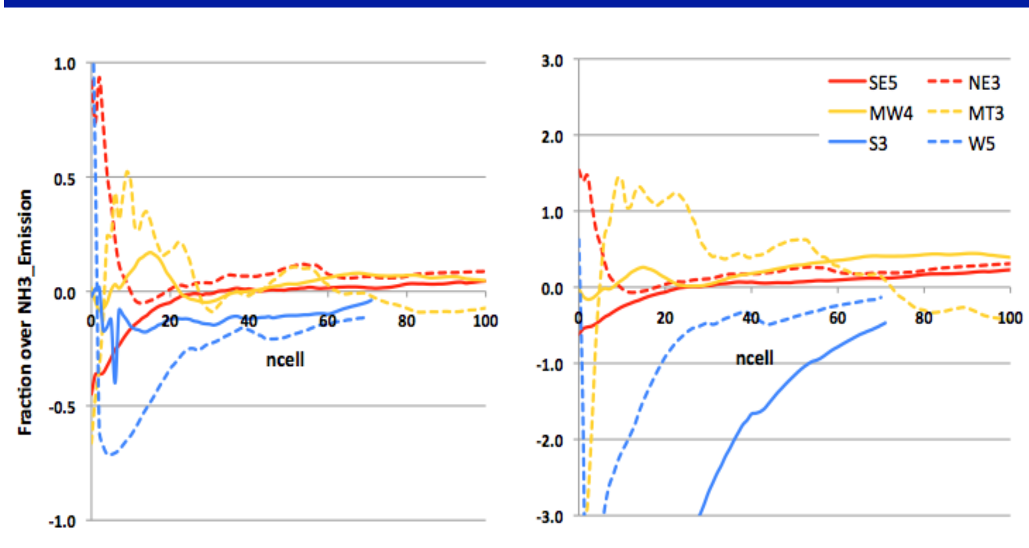
In the same region, the distance between source regions is more than 10 cells.

The area caused by ncells = [(ncells*(East+West) × 12km]×[(ncell*(North+South) × 12km]
where, East=West=North=South=1.

Ranges in the influence of horizontal advection



The sites selected the 6 regions



- Over United States, the ranges of NH_3 transport are up to 80 cells (~2000km).

- Due to NH_3 bidirectional exchange, the fractions of horizontal advection over NH_3 emission (FHADV) become large by a factor of 2 ~ 30 than base case.

- The ranges of influence horizontal advection depend on the land use types and the geographical distribution of NH_3 sources.

- NE, SE, MW : The variations of FHADV in bidi case are similar to base case but the magnitude of bidi case are twice larger than the base
- MT, S, W : The magnitude of FHADV of bidi are larger than base case by a factor of 20. NH_3 influxes from neighboring sources or large outflows affect over CONUS domain.

Conclusions and Implications

1. Domain wide

NH_3 bidirectional exchange:

$[NH_3]_{dry\_dep} \downarrow$ $[NH_3] \uparrow$ $[NH_4^+] \uparrow$ $[NO_3^-] \uparrow$ $[NH_4^+]_{wet\_dep} \uparrow$

2. From process analysis at a high CAFO emission site:

• Increase in NH_3 losses due to bidirectional exchange include aerosol production, wet deposition, and horizontal advection.

• Decrease in dry deposition losses

3. Changes in the range of advection influence depend on land use type and density of source region: The stronger NH_3 bidirectional exchange, the longer transport (e.g. Southern part of US and Western Part of US).

• NH_3 bidirectional mechanism is likely to increase lifetime of NH_3 through longer transport than the case of no NH_3 bidirectional exchange.

Implications for Inverse Modeling

• Simple inverse modeling such as AMF method is applicable in transformation dominant regime. If transport dominant regime needs to be included, the error estimation due to horizontal advection should be taken into account.

• Adjoint inversion: All the individual physical processes can be analyzed.
: The effects of NH_3 bidirectional exchanges can be proved in the characteristic processes of NH_3 bidirectional exchange, such as dry deposition and vertical diffusion, aerosol/cloud processes in transformation dominant regime, and advection process in transport dominant regime.

Bias and bidi effect in the EUS, August 2009

(base-obs)/obs (bidi-obs)/obs (bidi – base)/base

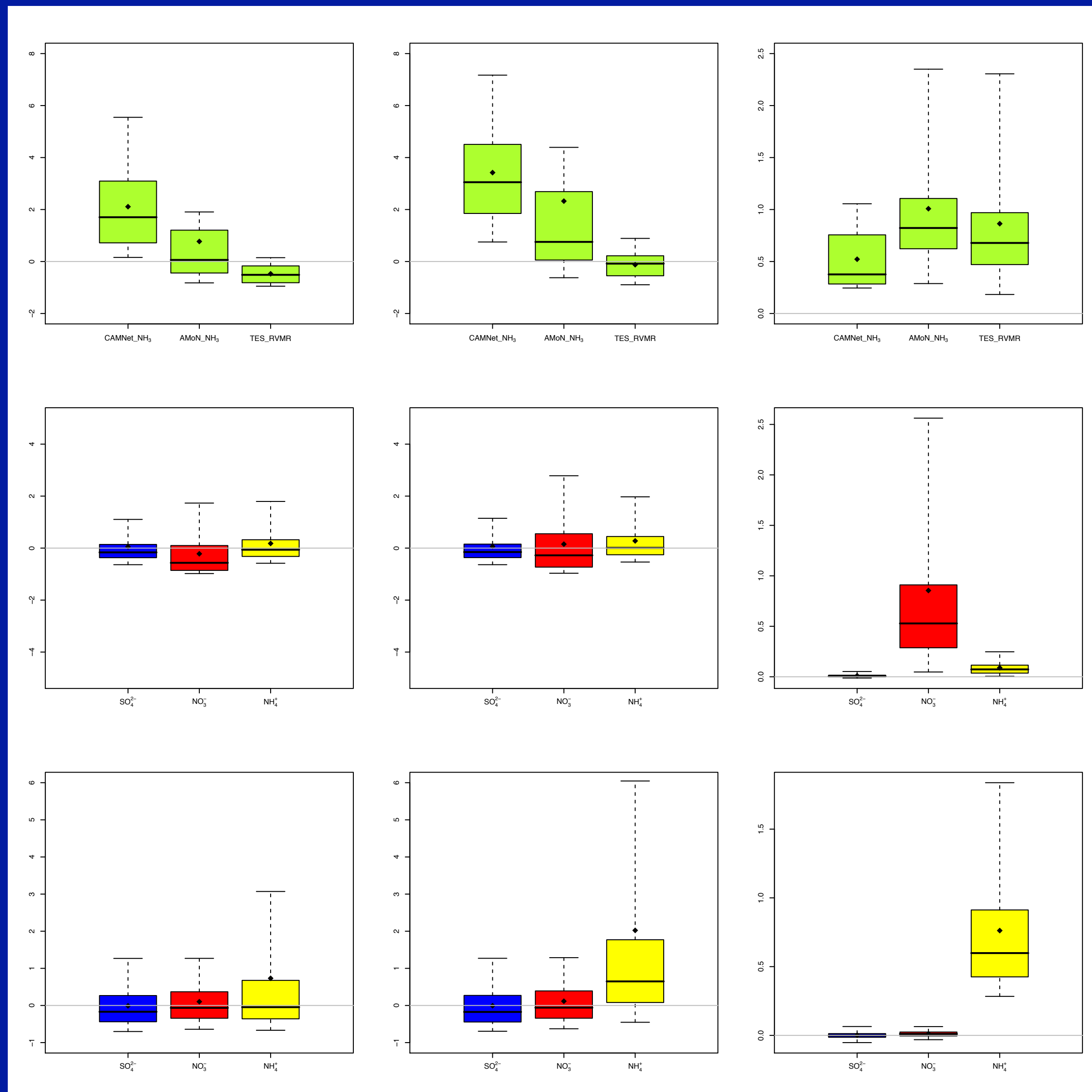
- NH_3 vs.
  1st CAMNet (NC)
  2nd AMoN (EUS)
  3rd TES RVMR (NC)

CMAQ base overestimates surface observation but underestimate TES RVMR

- SO_4^{-2}, NO_3^-, NH_4^+ vs. CSN (EUS)

- wet deposition
  SO_4^{-2}, NO_3^-, NH_4^+ vs. NADP (EUS)

CMAQ base aerosol concentrations are in good agreement with observations

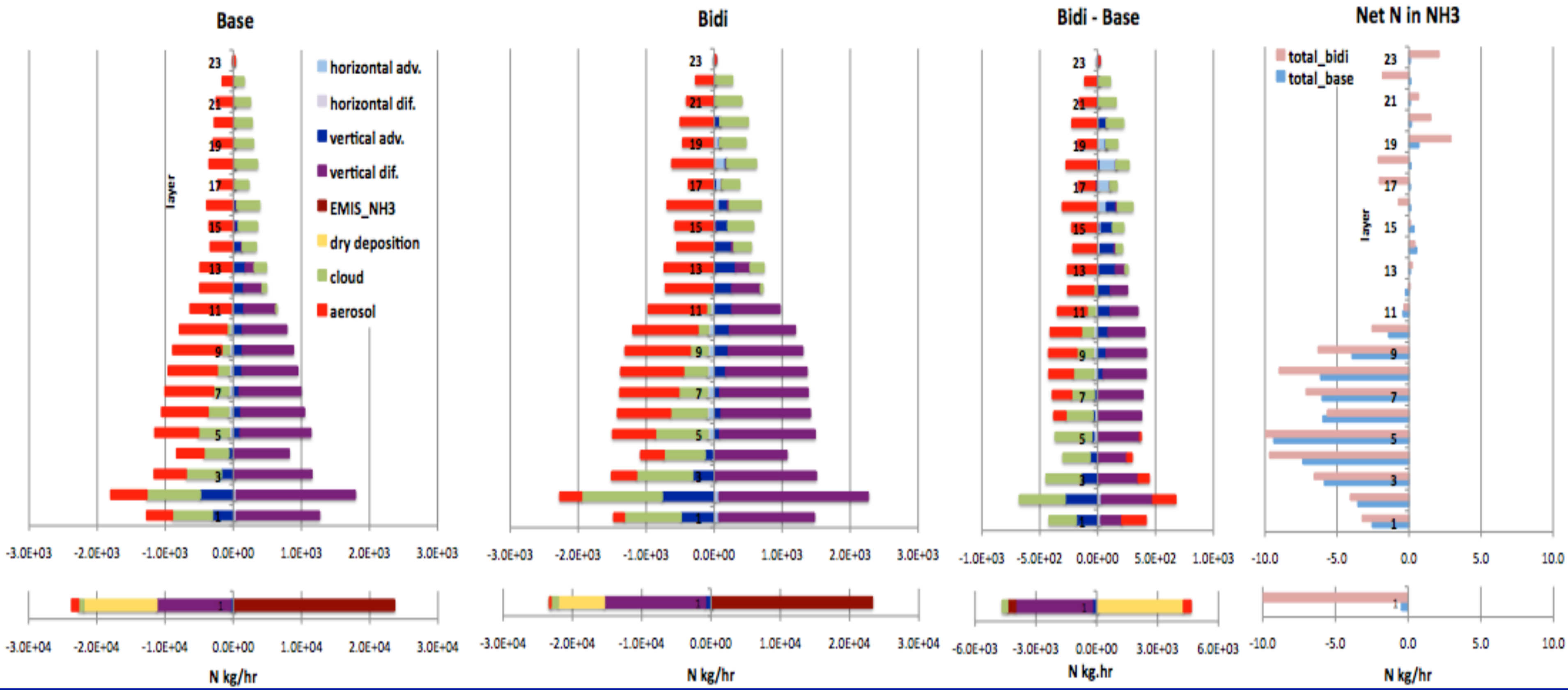


Bidi exchanges reduce the bias against TES

NO_3^- a lot increases by 30 ~ 90 %. NH_4^+ increases by 10~20%.

Wet deposition Of NH_4^+ increases By 50 ~ 90%

The vertical distribution of the fraction of individual processes apart from NC TES transect



•N loss due to removal processes: base case < bidi case by more than O(1)
•Above surface, gain due to diffusion, loss due to aerosol and cloud process.
➔Multi-phase process (transformation) is vigorous rather than transport in all the layers.
•The changes in N in NH_3 are net loss in lower atmosphere, net gain in upper atmosphere.
•The vertical changes in N mass due to physical process are less steep than those in NC transect.
•Cloud process causes a loss in the lower atmosphere but a gain in the upper atmosphere.
•The changes in the fraction of horizontal advection process by the extent of horizontal advection are important to determine NH_3 lifetime and aerosol growth.