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Synergy between Emission Verification for Climate and Air Quality:

CMAQ Modeling Analysis over the Contiguous US domain

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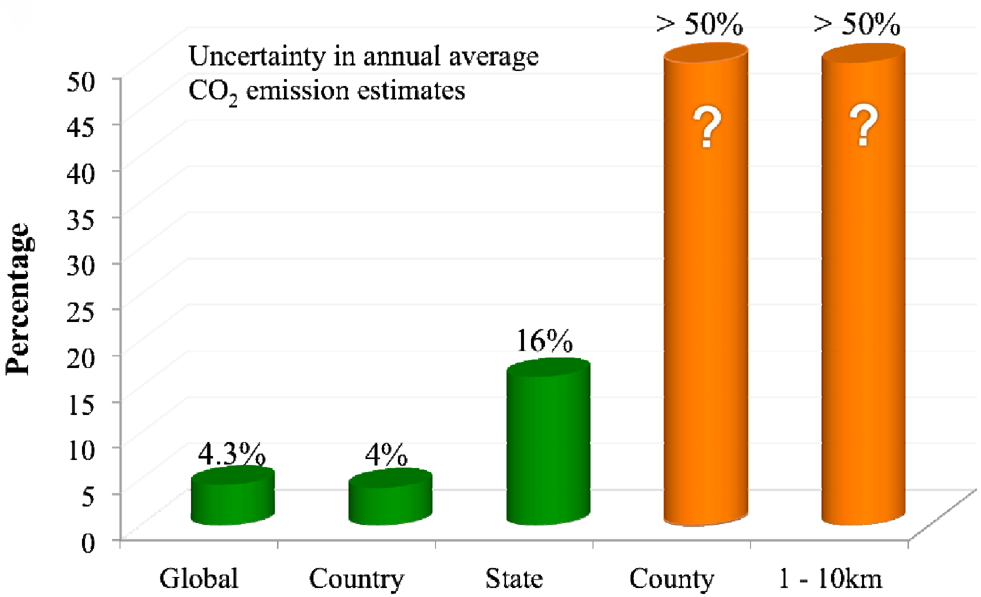
Fossil-fuel CO_2 emission verification:

uncertainties in fossil-fuel CO_2 emissions

spatiotemporal distributions

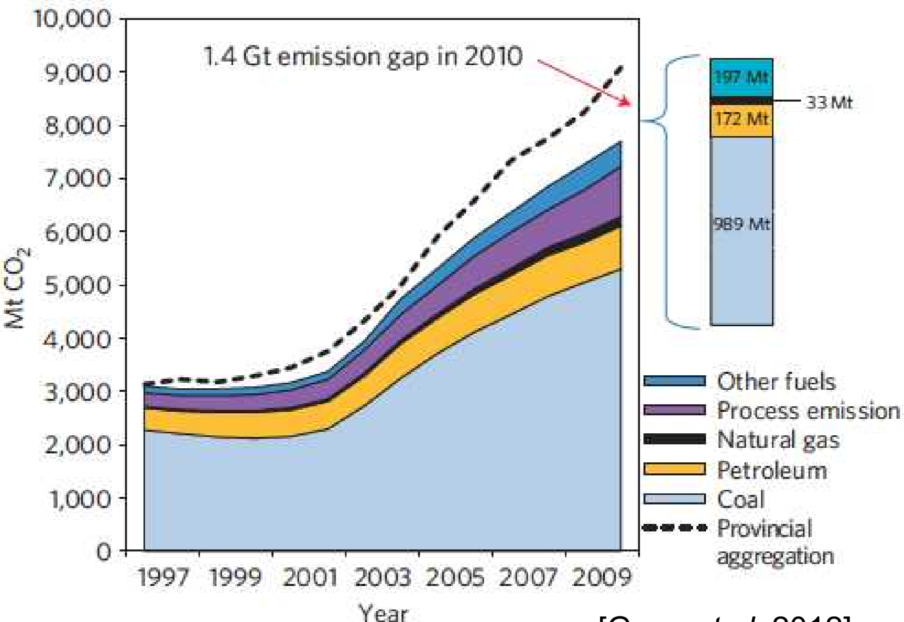


higher uncertainty after temporal allocation!

Sources: **Global:** NRC [2010]; **Country**: EPA [2012]; **State, county and 1-10km**: Gurney et al. [2009]

national emission trends

The gigatonne gap in China's CO_2 inventories



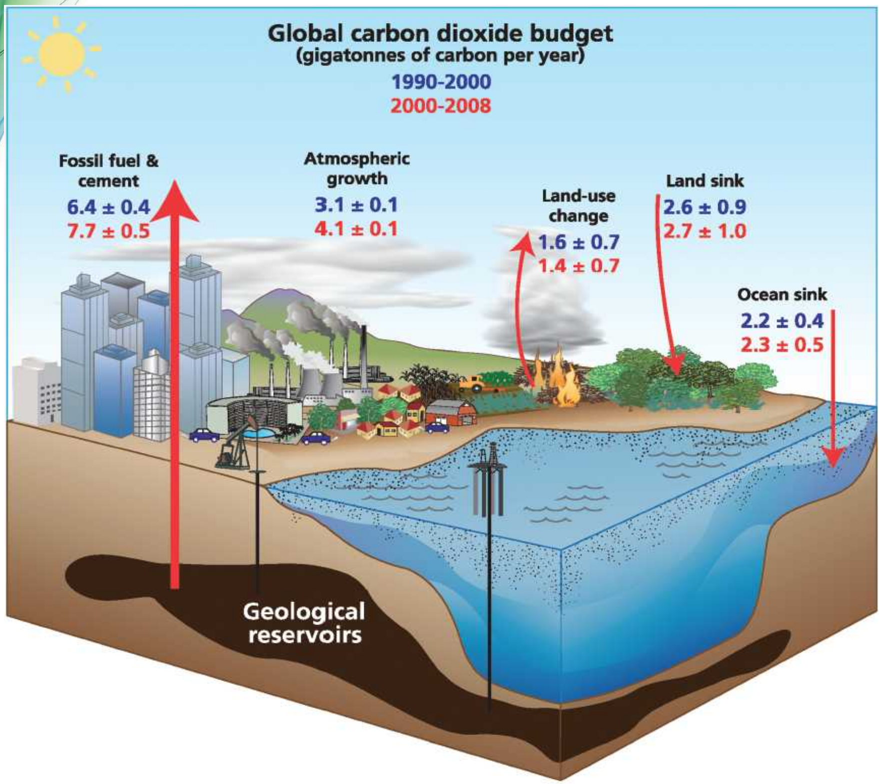
[Guan *et al*. 2012]

Verifying fossil CO_2 emissions is

"firmly on the agenda of science, politics, and business".

[Marland, 2008]

Challenges in atmospheric CO_2 source attribution



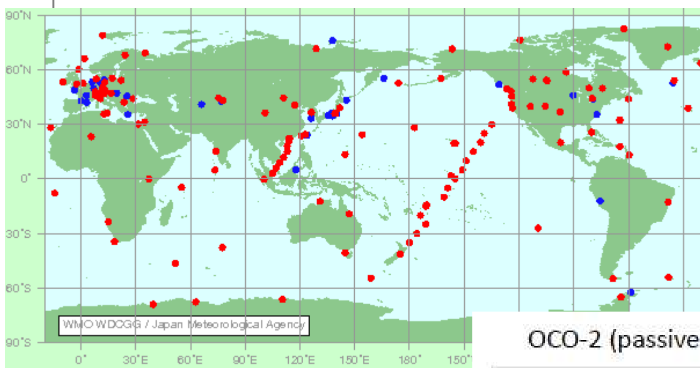
NRC, 2010: *Verifying Greenhouse Gas Emissions: Methods to Support International Climate Agreements, pp16, Fig. 1.3*

$$[CO_2]_{OBS} = [CO_2]_{natural} + [CO_2]_{fossil} + [CO_2]_{background}$$

Fossil-fuel emission

Terrestrial carbon cycle (highly uncertain)

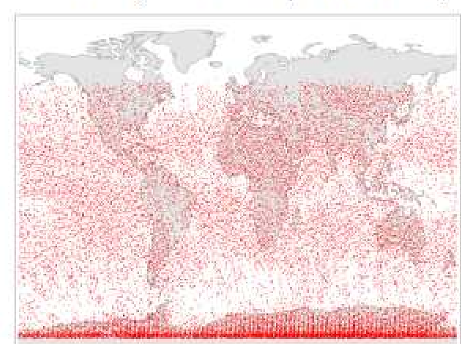
In situ network



http://ds.data.jma.go.jp/gmd/wdcgg/cgi-bin/wdcgg/map_search.cgi

satellites

OCO-2 (passive SWIR, launch 2014)



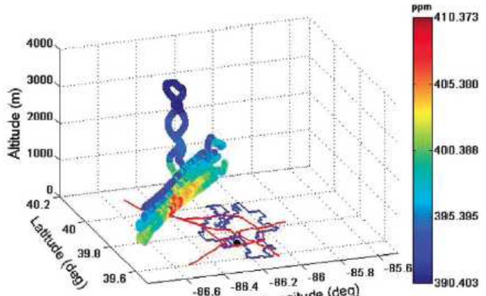
Caias et al., 2013

Quantifying fossil-fuel emissions is difficult due to:

(1) Large and uncertain natural carbon fluxes
(2) Sparse/inadequate observational data
(3) Transport model biases

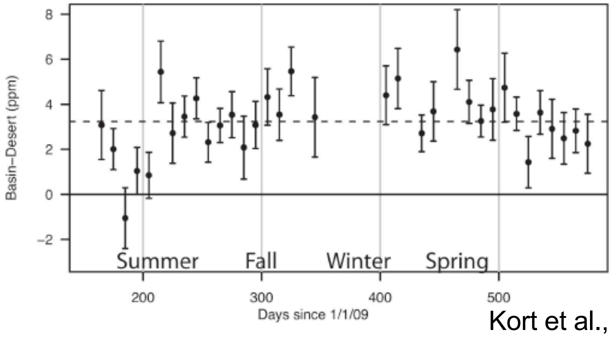
Current CO_2 emission verification activities and motivations for regional CO_2 modeling

Approach I: in situ observations



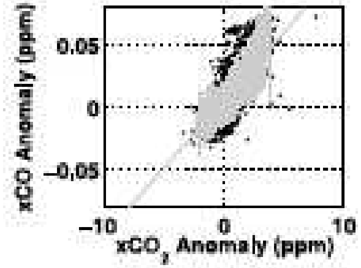
Mays et al., 2009

Approach II: satellite observations



Kort et al., 2011

Approach III: tracer correlation/ratio



Wunch et al., 2009

Research needs

Understanding the fine-scale spatial distribution of CO_2 and influence of the biosphere

Characterizing the observational system

Understanding the CO_2:tracer relationships

CMAQ-CO_2 Model configuration

CMAQ model setup

✓ **Version**	:	CMAQ v5.0
✓ **Domain**	:	CONUS/36km/22L
✓ **Meteorology**	:	WRF v3.1.1
✓ **Simulation year**	:	2007

CO_2 simulation

✓ **BC and IC**	:	CT-2011 (1°× 1°; 3-hourly)
✓ **Biosphere fluxes**	:	(1) CT-2011 (1°× 1°; 3-hourly) (2) CASA (1°× 1°; 3-hourly) (3) CLM4MIP (0.5°× 0.5°; 3-hourly)
✓ **Fossil fuel emissions**	:	(1) Vulcan (2002; 10km; hourly) in the US (2) CDIAC (2007; 1°× 1°; monthly) outside
✓ **Fire emissions**	:	GFEDv3.1 (0.5°× 0.5°; 3-hourly)
✓ **Ocean fluxes**	:	CT-2011 (1°× 1°; 3-hourly)

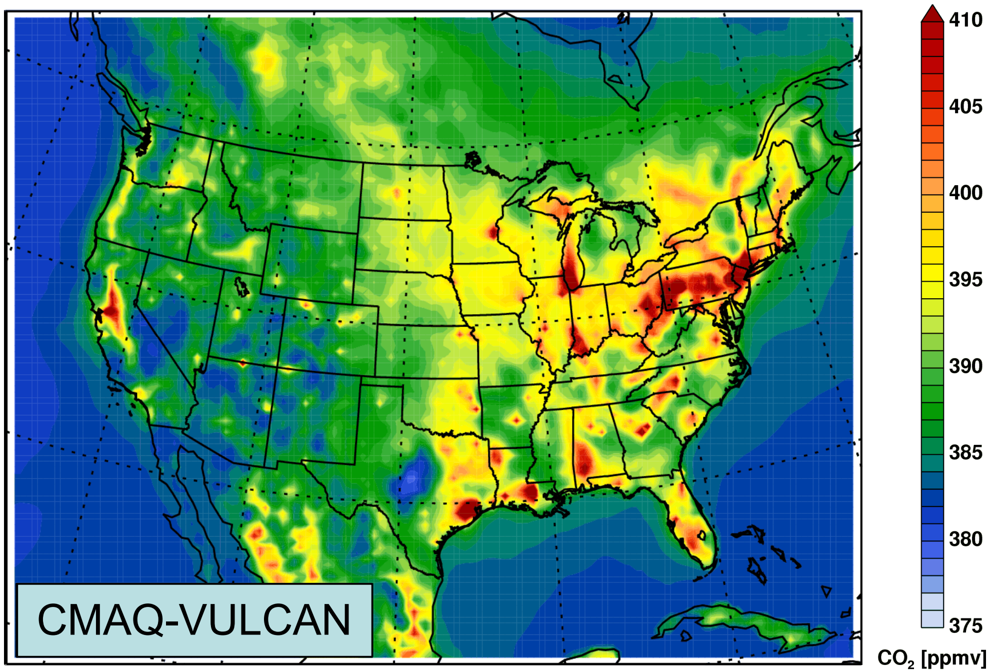
Chemistry simulation

✓ **BC and IC**	:	GEOS-Chem v8.2.1
✓ **Biogenic emissions**	:	MEGAN v2.1
✓ **Anthropogenic emissions**	:	NEI-2005
✓ **Fire emissions**	:	Blue Sky + FEPS
✓ **Chemistry**	:	CB-05

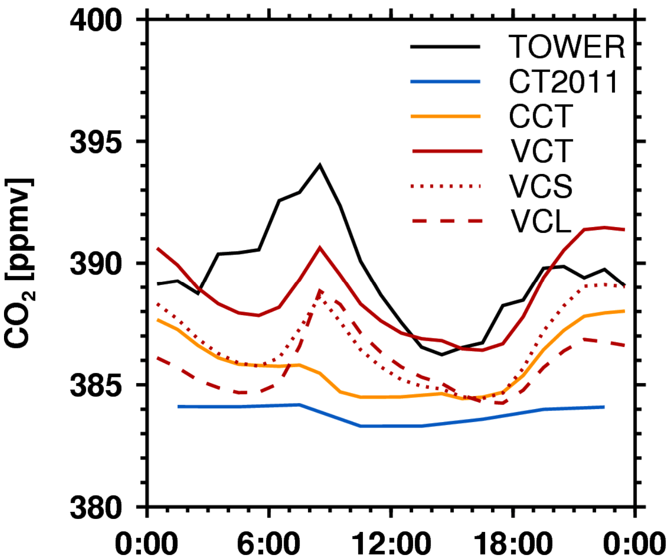
(For details refer to Liu *et al.*, 2013, JAWMA CMAS-2013 special issue)

CMAQ-CO_2 model evaluation

From CarbonTracker-2011 to CMAQ-CO_2
(Surface CO_2 in October 2007)



Model evaluation using tall tower data



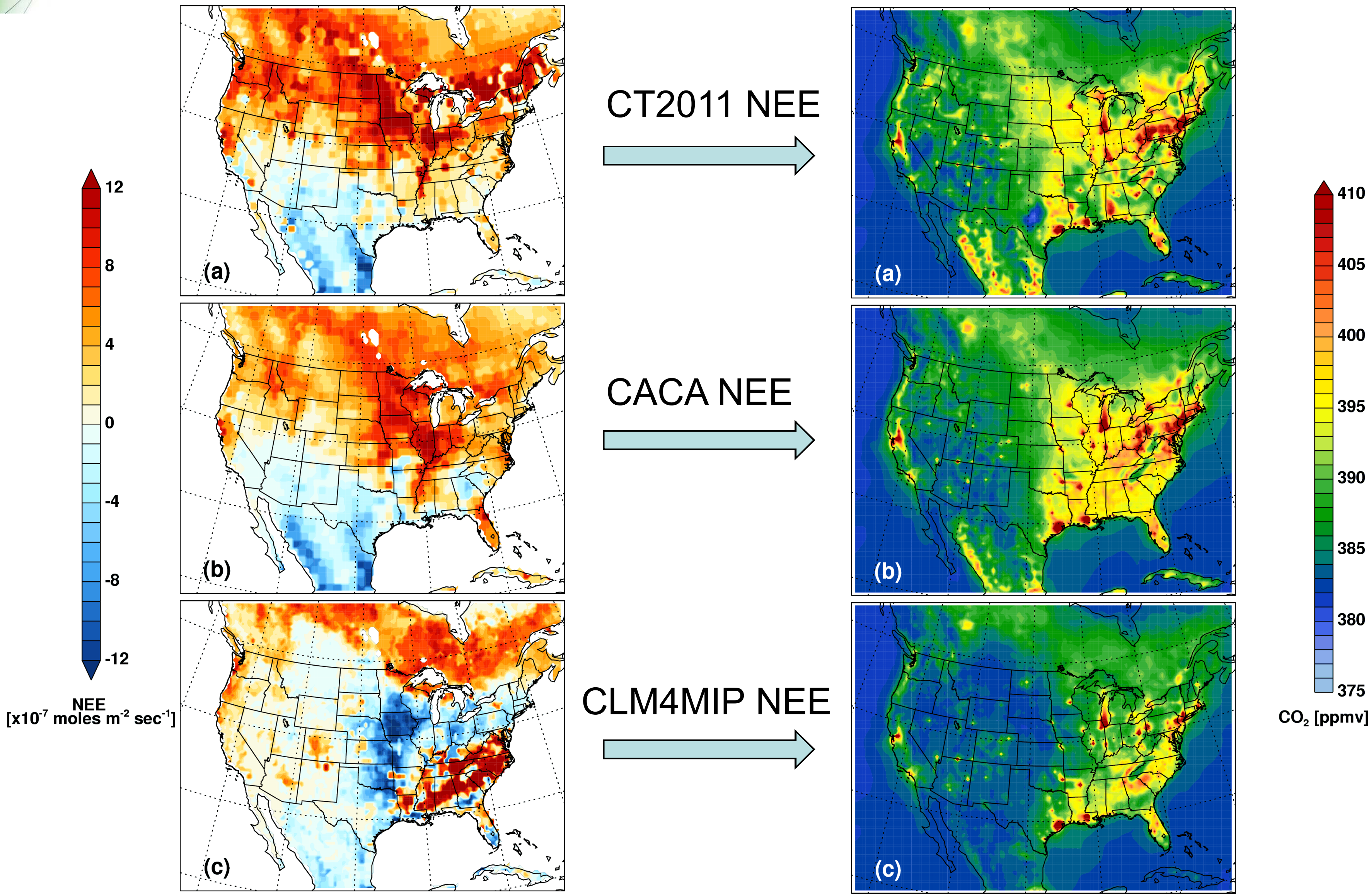
Monthly mean diurnal profiles of CO_2 in October 2007 observed at Boulder Atmospheric Observatory (BAO) (TOWER) and simulated by CT2011 and CMAQ with different configurations:
CCT: CDIAC FF emissions and CT2011 NEE;
VCT: Vulcan FF emissions and CT2011 NEE;
VCS: Vulcan FF emissions and CASA NEE;
VLM: Vulcan FF emissions and CLM4VIC NEE.

Examples of 'First-order' scientific questions to address with CMAQ:

✓What are the impacts of biosphere (and its uncertainties) on regional/urban CO_2 distribution?
✓How well/difficult can we see fossil-fuel CO_2 ($FFCO_2$) from space?
✓How similar/different are the distributions of $FFCO_2$ and combustion byproducts?

Uncertain biosphere-atmosphere exchange

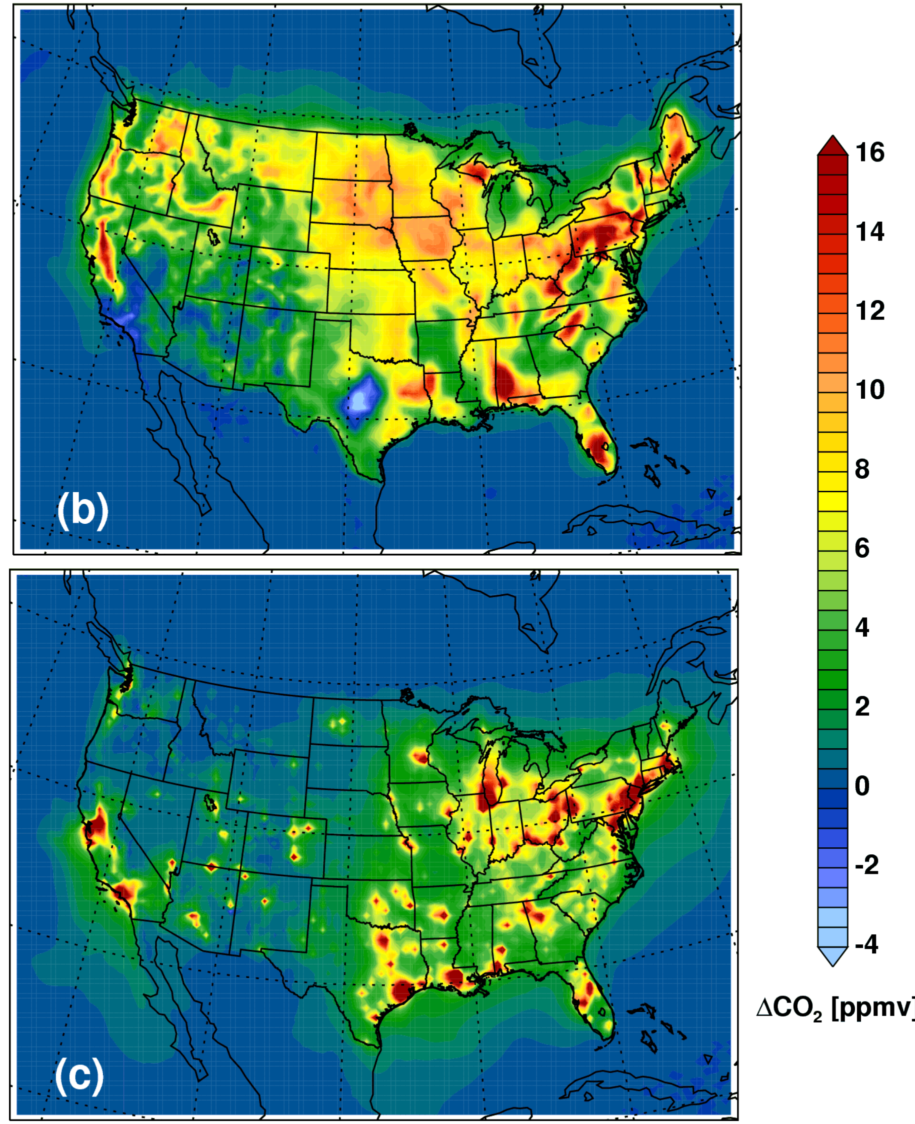
Sensitivity to different NEE inputs



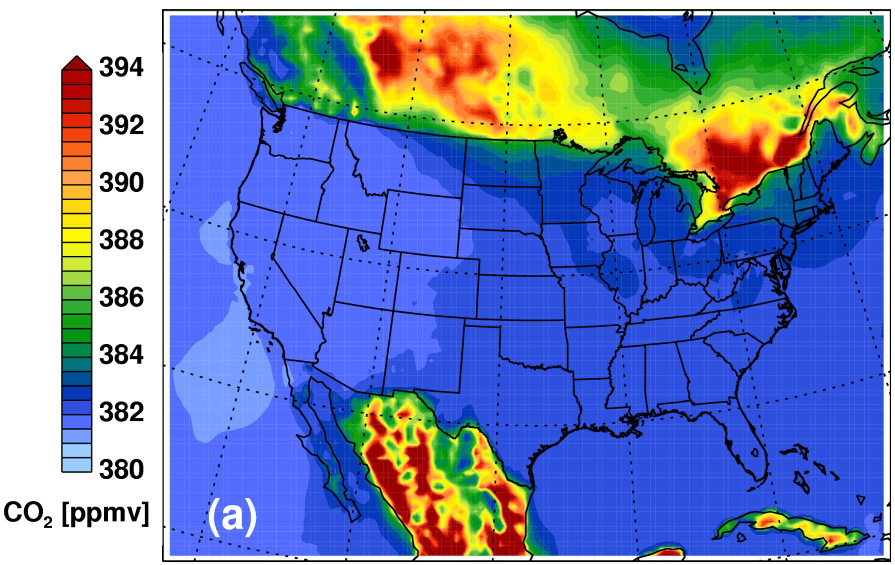
Decomposition of CO_2 components

Background, biosphere and fossil-fuel CO_2 components (October 2007)

Biospheric CO_2

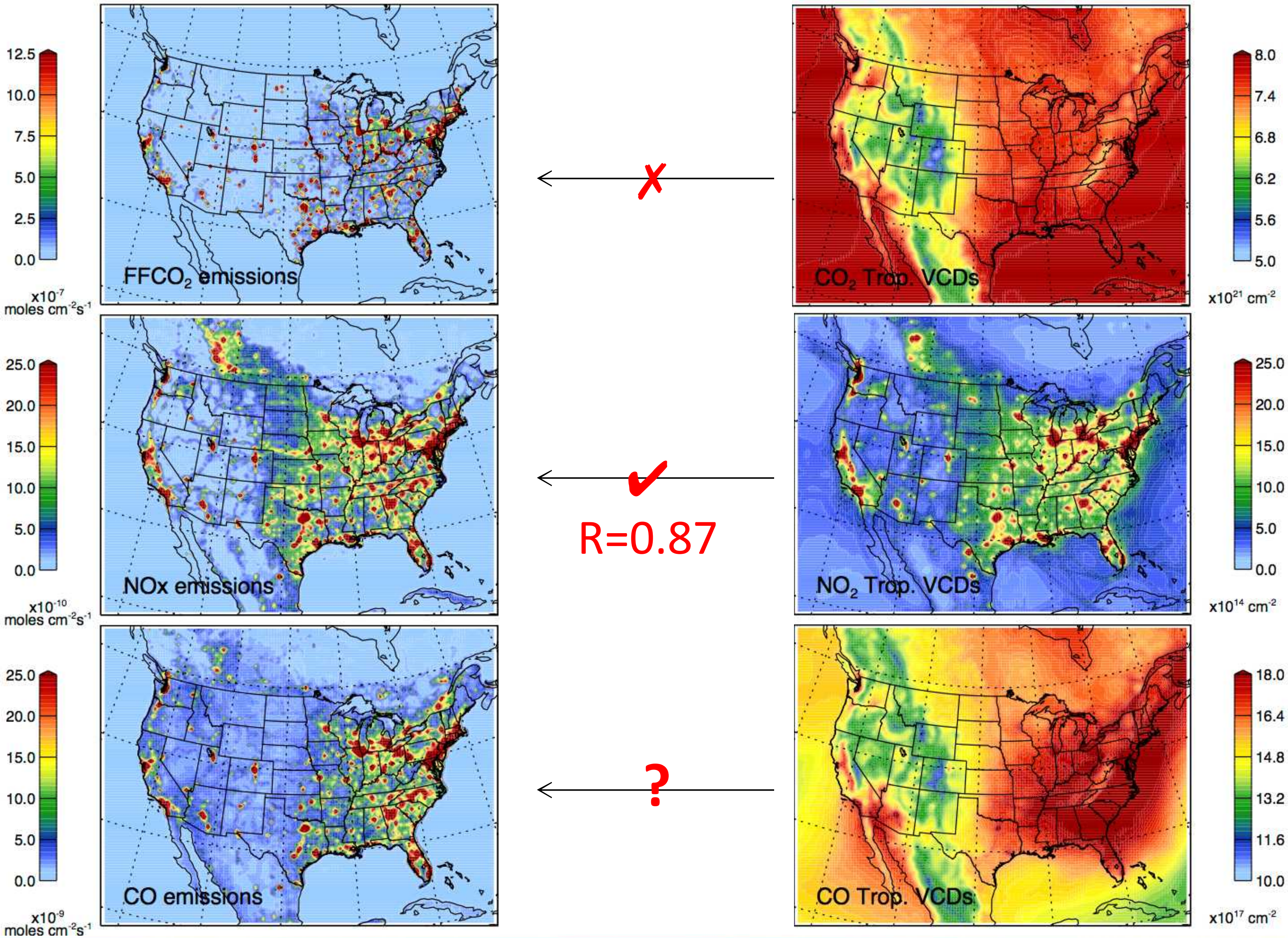


Background (w/o US fluxes)



Fossil-fuel CO_2 →

Using CMAQ to simulate space-observed CO_2 and combustion tracers (July)



Using tracers to constrain fossil-fuel CO_2 emissions: theoretical considerations

(1) The emission ratio approach $E_{CO_2} = \beta E_{tracer}$:

[e.g., Berezin *et al*. 2013]

Requirements/circumstances:

a. abundant and reliable observational data for the tracer (not necessary for CO_2)

b. good knowledge of the emission ratio (β)

Research needs:

a. quantify emission ratios for different sources

b. method uncertainty quantification

(2) The concentration slope approach $C_{CO_2} = \alpha C_{tracer}$:

[e.g., Prather et al., 1985; Brioude et al., 2012; Silva et al., 2013]

Requirements/circumstances:

a. observation data for both CO_2 and tracer

b. CO_2 and tracer has almost identical sources

Research needs:

a. understand factors that affect the tracer slope and correlation.

b. method uncertainty quantification

(3) The formal joint inversion approach:

[e.g., Wang *et al*. 2009]

Requirements/circumstances:

a. observation data for both CO_2 and tracer

b. knowledge of the transport and prior error correlations between CO_2 and tracer

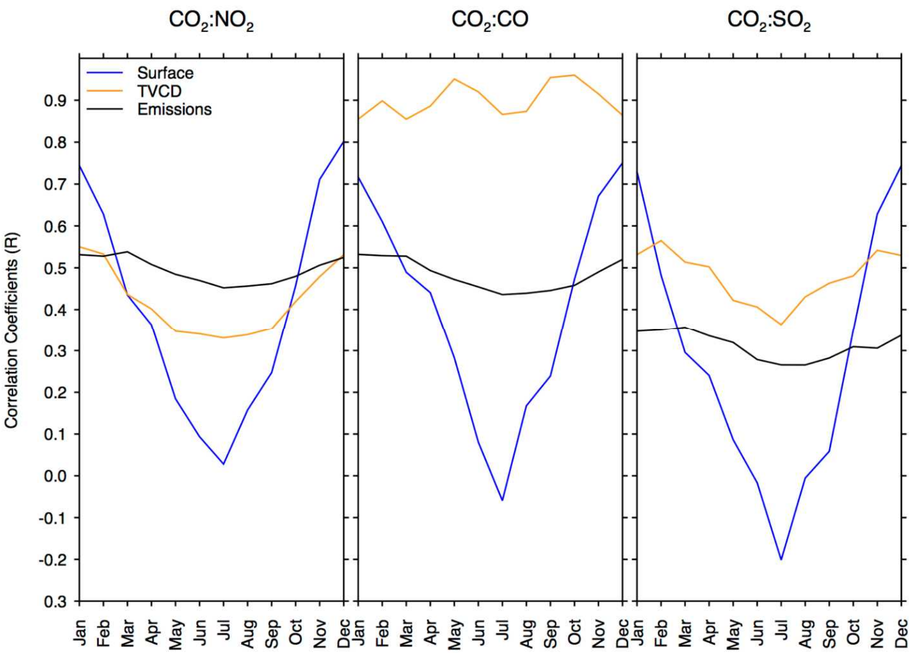
Research needs:

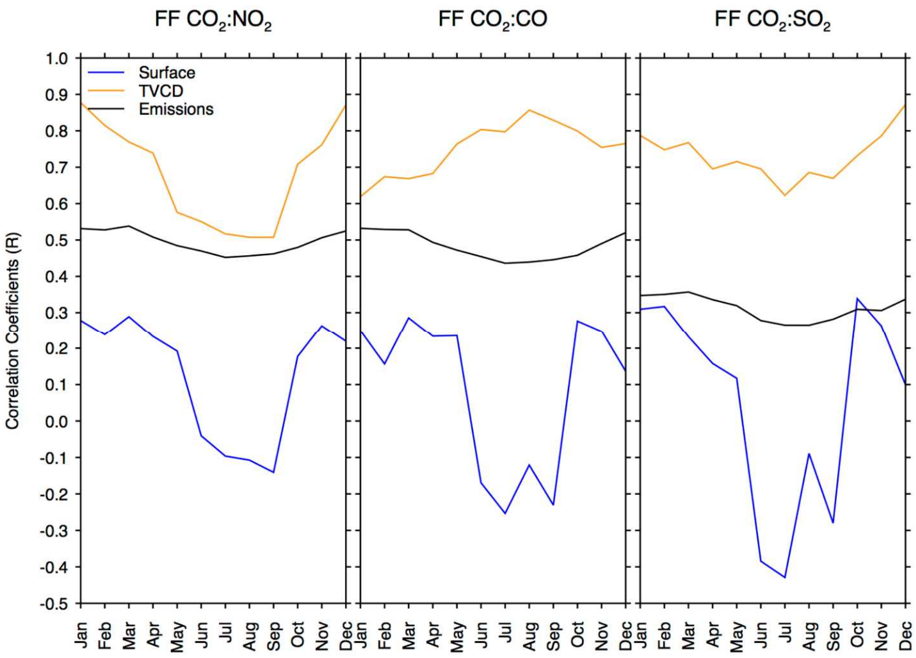
a. quantify transport and and prior error correlations

CMAQ-simulated spatial correlations of CO_2 and combustion tracers

We examined spatial correlations in the US domain for

(1) emissions (monthly mean)
(2) surface concentrations (local time 14:00)
(3) tropospheric vertical column densities (TVCDs, local time 14:00)
(4) both CO_2 and its fossil-fuel CO_2 component (FF CO_2)



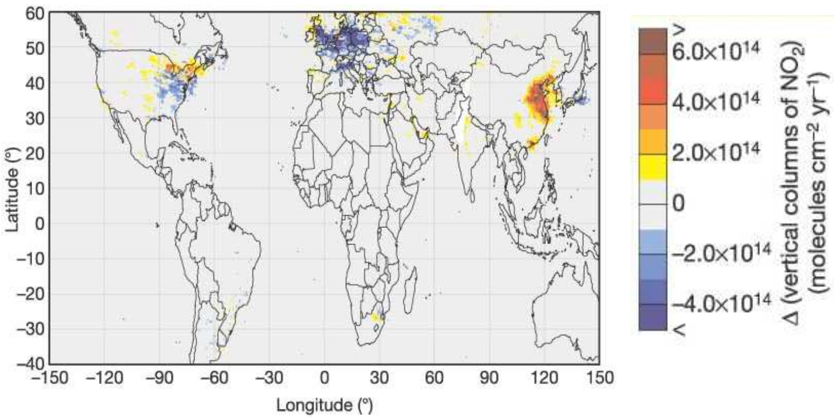


- ✓ Moderate to low emission correlations due to source differences
- ✓ FF CO_2 and combustion tracers are more closely related when observed from space
- ✓ Transport is a major factor shapping the atmospheric concentration patterns and correlations

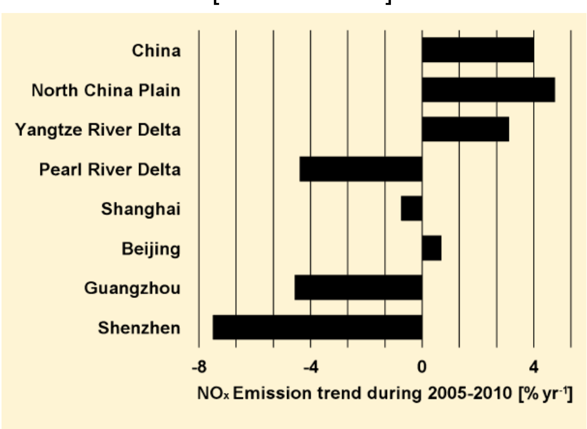
Tropospheric NO_2 VCDs: a good fossil-fuel CO_2 tracer?

Satellite NO_2 as an indicator of fossil-fuel emissions

1996-2002 GOME NO_2 trends [Richter *et al.* 2005]

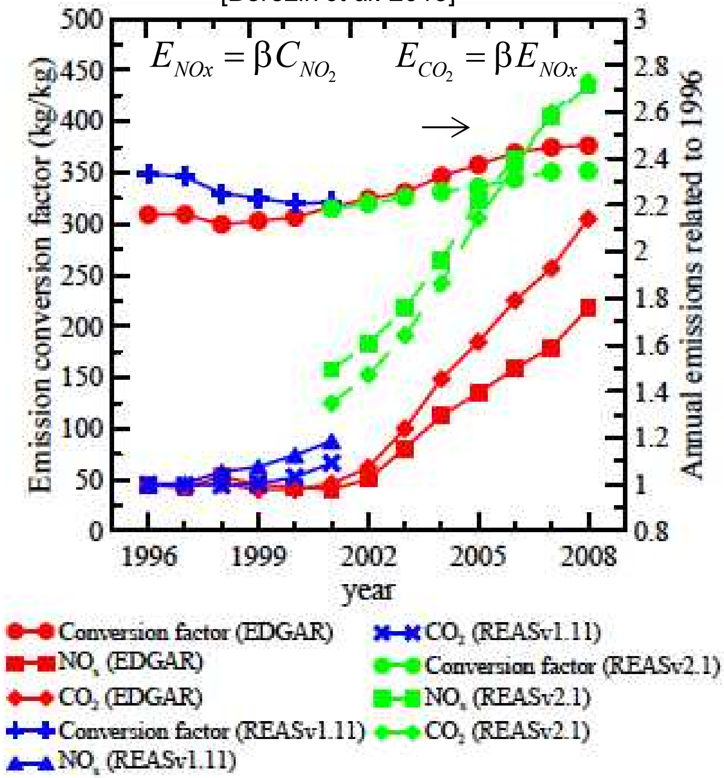


2005-2010 emission trends using OMI NO_2
[Gu *et al.* 2013]



Estimate CO_2 emissions trends using satellite NO_2

1996-2008 CO_2 emission trends from GOME and SCIAMACHY NO_2
[Berezin *et al.* 2013]

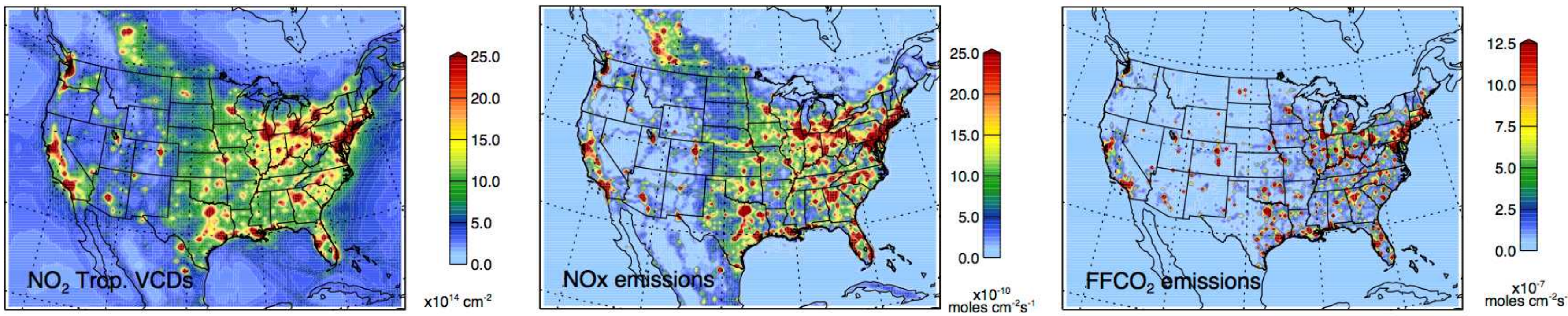


Using tropospheric NO_2 VCDs to constrain fossil-fuel CO_2 emissions

Theoretical and practical considerations:

1) Satellite-observed tropospheric NO2 VCDs closely relate to NOx emissions;
2) abundant data and experience are available from NOx emission verification by the air quality research community.

Steps:



Step 1: → NOx emission inversion

Step 2: → NOx-to-CO_2 emission conversion

Key technical development (*on going*):

✓ Carefully compiled CO_2:NOx emission ratio data;

✓ An inversion scheme properly taking into account the non-linear dynamics involving NOx emissions and chemistry, NO_2 vertical profiles, and tropospheric NO_2 vertical column densities (VCDs);

✓ Uncertainty quantification and comparison with the prior emission uncertainties

Summary

- We demonstrated how a regional chemical transport model like CMAQ can help understand regional CO_2 variability and facilitate emission verifications.
- We showed the similarities between fossil-fuel CO_2 and those combustion byproducts, which have long been studied by the air-quality community.
- Using a combustion byproduct as a tracer could help mitigate the challenges in atmospheric CO_2 source attribution.
- We outlined an approach to using satellite NO_2 observations to constrain fossil-fuel CO_2.

Acknowledgements

- CarbonTracker-2011 results are provided by NOAA ESRL (http://carbontracker.noaa.gov).
- Tower CO_2 data are provided by NOAA GMD.
- The CLM4VIC output is a product of the MsTMIP project
- WRF output and non-CO_2 emission data are shared by the SESARM project (http://www.metro4-sesarm.org).
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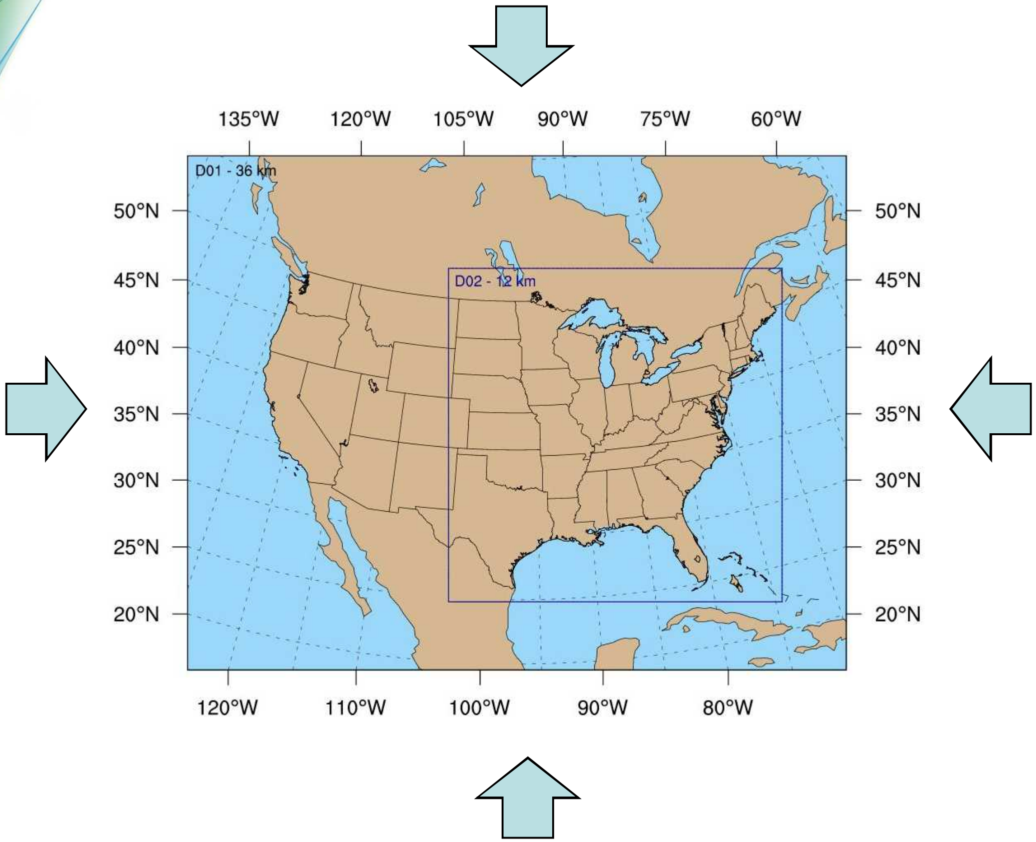
Thank you for your attention!!

Backup slides

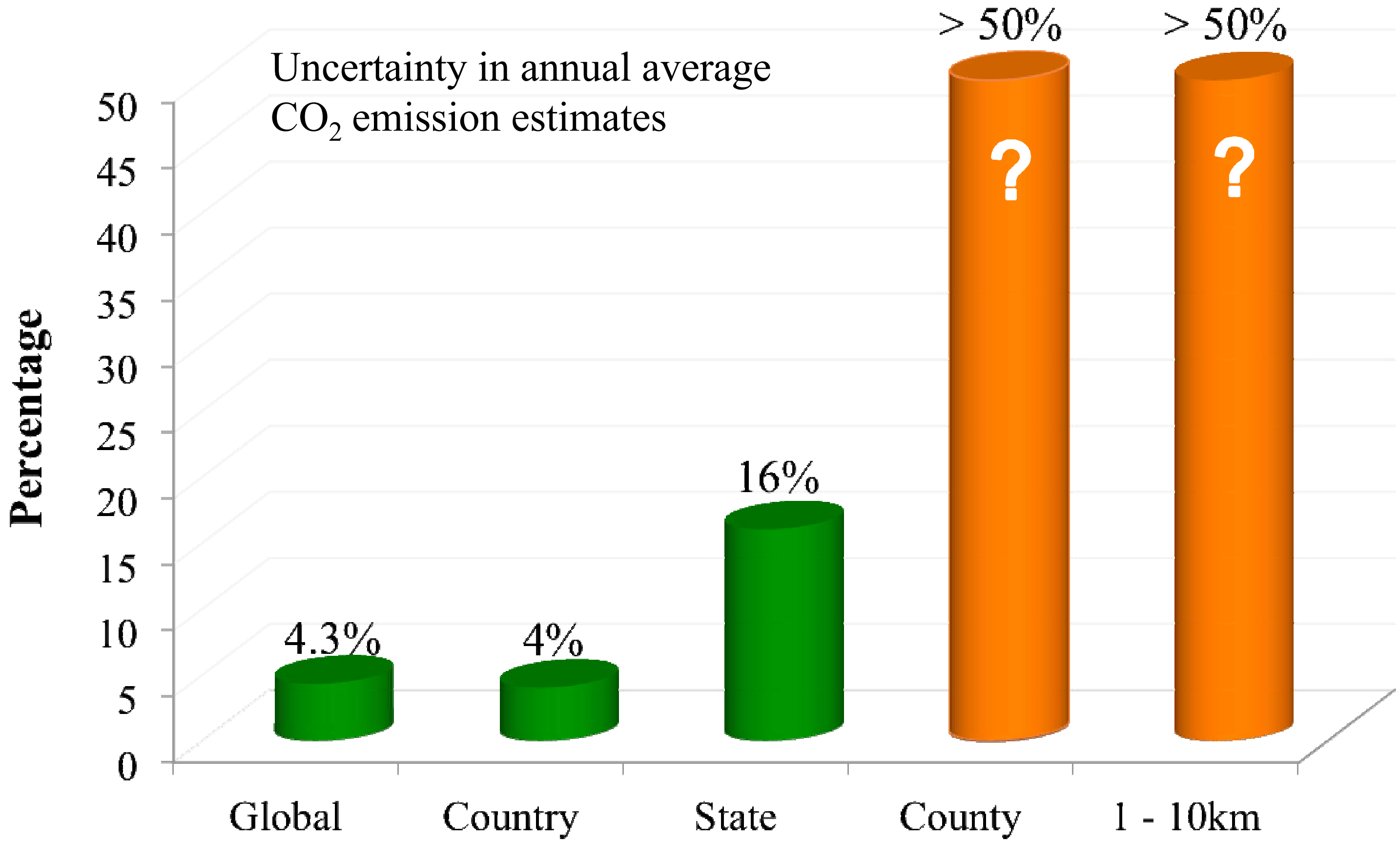
CMAQ-CO_2 model configuration



CO_2 Initial/boundary conditions: CT-2011 (1°× 1°; 3-hourly)

- **Meteorology and resolution**
  WRFv3.1.1 assimilated meteorology for 2007
  36km resolution;
- **CO_2 simulation**
  - ✓ **BC and IC**:
    CT-2011 (1°× 1°; 3-hourly)
  - ✓ **Biosphere fluxes** :
    (1: STD) CT-2011 (1°× 1°; 3-hourly)
    (2) CASA (1°× 1°; 3-hourly)
    (3) CLM4MIP (0.5°× 0.5°; 3-hourly)
  - ✓ **Fossil fuel emissions**:
    (1: STD) Vulcan (2002; 10km; hourly) in the US
    (2) CDIAC (2007; 1°× 1°; monthly) outside the US
  - ✓ **Fire emissions**:
    GFEDv3.1 (0.5°× 0.5°; 3-hourly)
  - ✓ **Ocean fluxes:**
    CT-2011 (1°× 1°; 3-hourly)
- **Chemistry simulation**
  - ✓ **BC and IC:**
    GEOS-Chem v8.2.1
  - ✓ **Biogenic emissions:**
    MEGAN v2.1
  - ✓ **Anthropogenic emissions:**
    NEI-2005
  - ✓ **Fire emissions:**
    Blue Sky + FEPS
  - ✓ **Chemistry:**CB-05

Uncertainty of fossil-fuel CO_2 emissions



higher uncertainty after temporal allocation!

Sources: **Global:** NRC [2010]; **Country**: EPA [2012]; **State, county and 1-10km**: Gurney et al. [2009]