Emissions: What, Why, Where, How?

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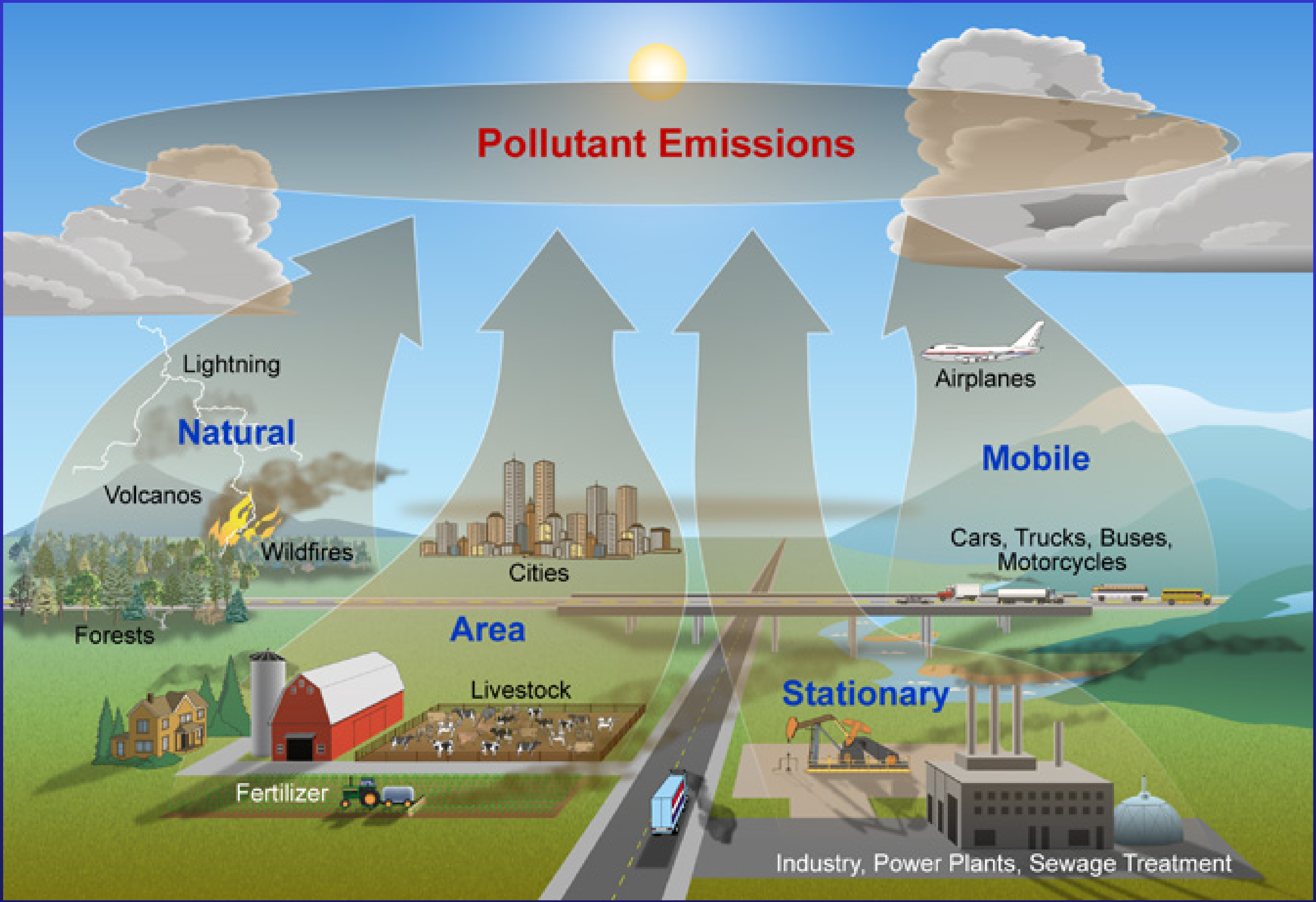
Topics

Part 1. Overview and Bottom-Up Inventories
Part 2. Inventory Evaluations and Top-Down Approaches
Part 3. Quantifying Specific Emissions Sources

NCAR ASP Colloquium, 03 August 2016

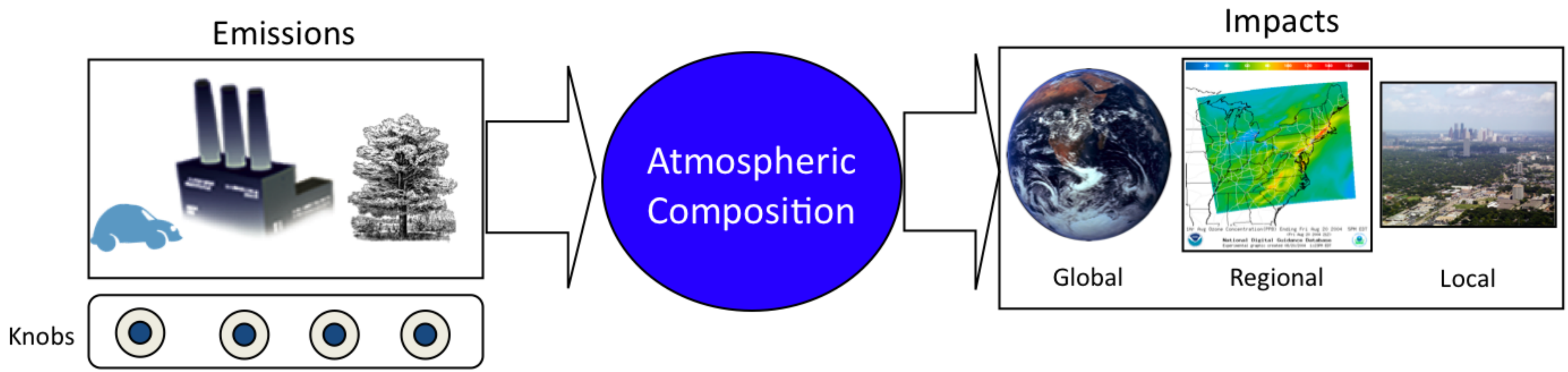
Part 1. Overview and Bottom-Up Inventories

- Sources of emissions to the atmosphere
- Emission Inventories: What are they and why are they important?
- How to build an inventory
- Available inventories
- Where to get inventory data


Pollutant Emissions
Lightning
Natural
Volcanos
Wildfires
Forests
Cities
Area
Livestock
Fertilizer
Airplanes
Mobile
Cars, Trucks, Buses, Motorcycles
Stationary
Industry, Power Plants, Sewage Treatment

Motivation for Understanding Emissions

Actions and decisions about the atmosphere focus on emissions

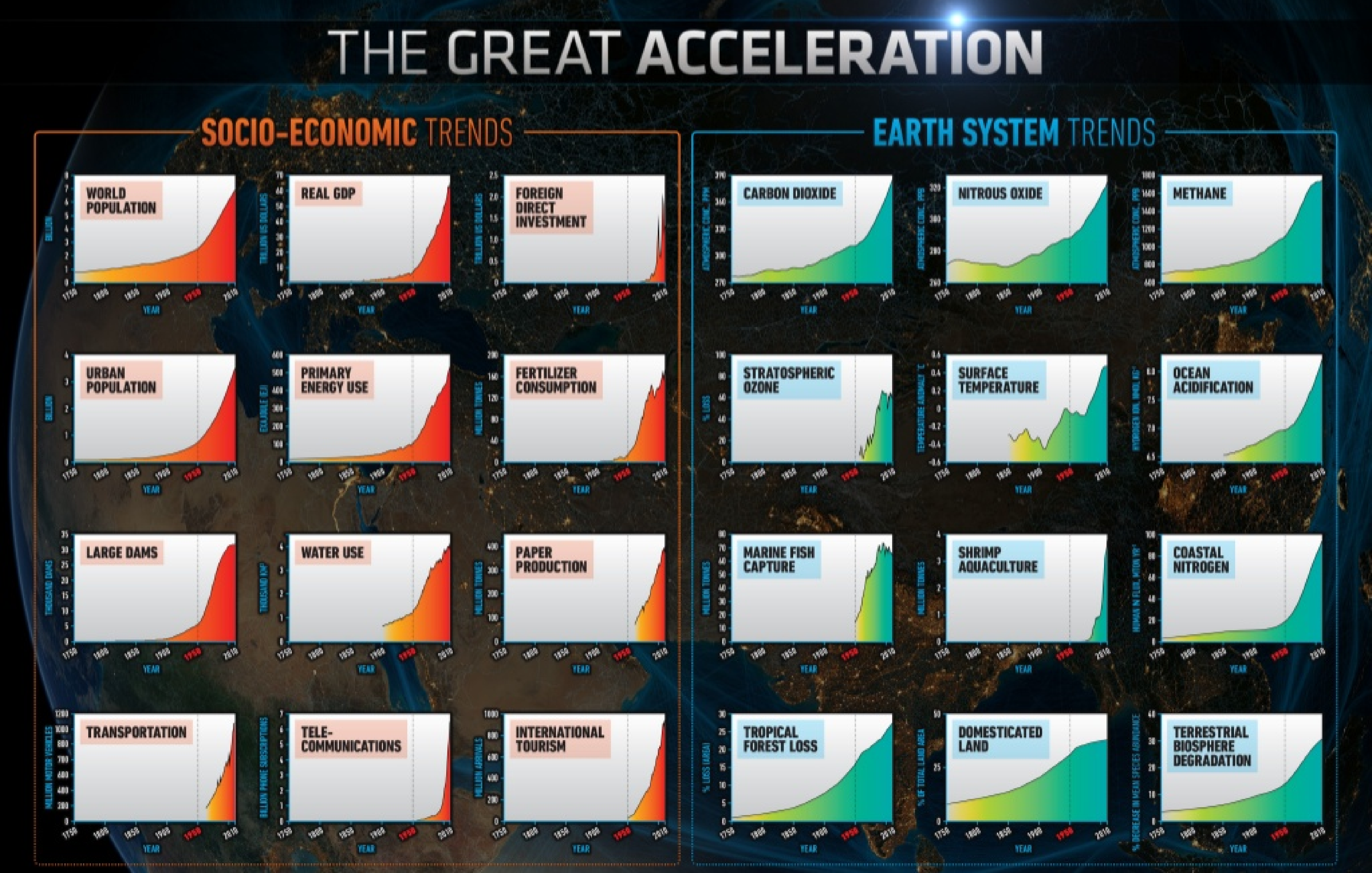


Accurate emission information is needed to:

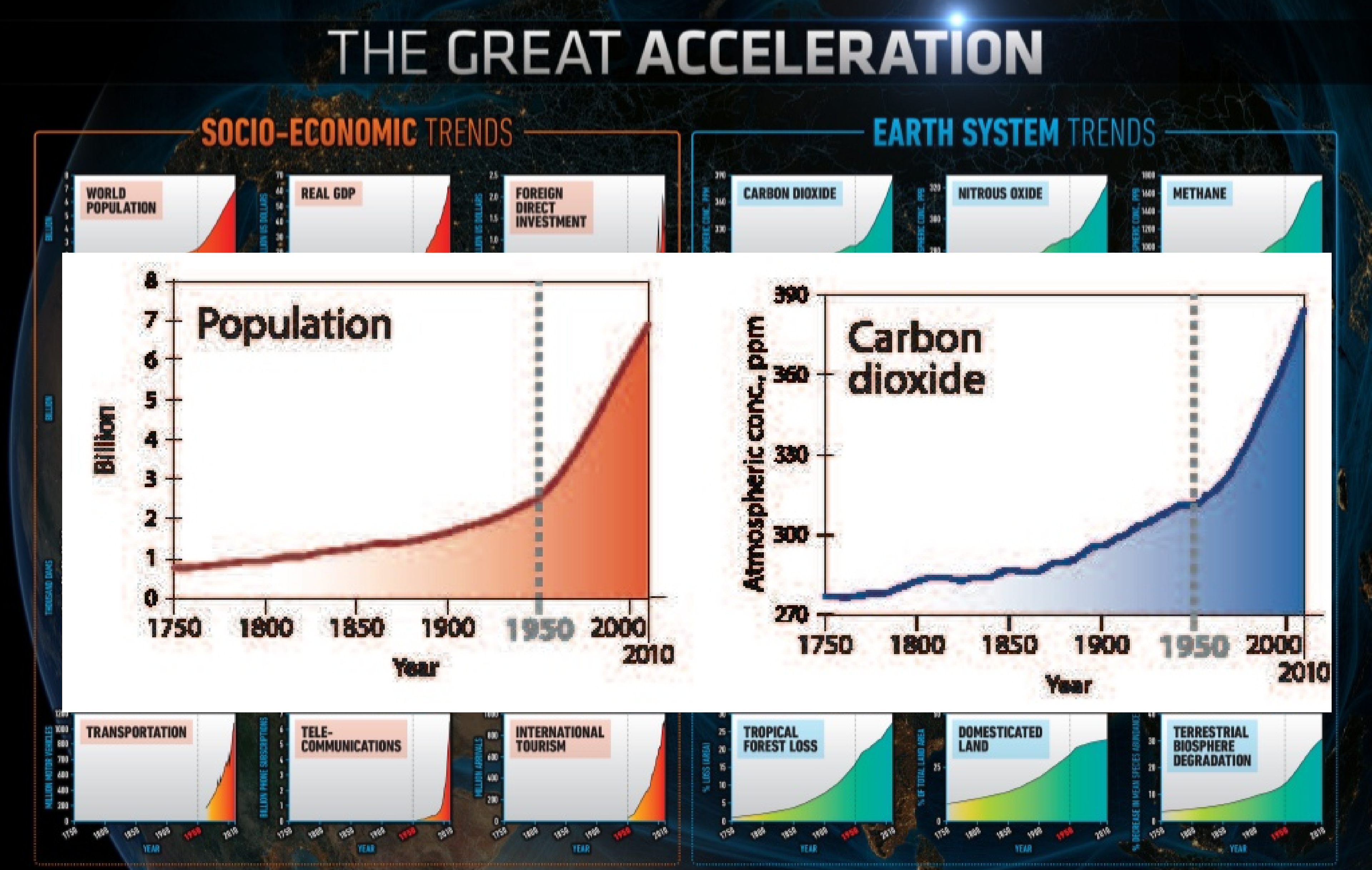
- Quantify and predict atmospheric composition
- Understand changes in air quality and climate
- Make choices about emission controls

We are in the "Century of Accountability"

Emissions are tools of regulation, economics, foreign policy, & international diplomacy http://www.futureearth.org/blog/2015-jan-16/great-acceleration



W. Steffen et al. (2015) The Anthropocene Review, doi: 10.1177/2053019614564785 http://www.futureearth.org/blog/2015-jan-16/great-acceleration



W. Steffen et al. (2015) The Anthropocene Review, doi: 10.1177/2053019614564785

Emissions & air pollution: is it a new problem ?

Not really = Smog episodes reported from the early 1900s

Los Angeles, **USA**, 1950s

Pittsburgh, **USA**, 1948

Meuse Valley, **Belgium**, 1930

Leeds, **UK**, 1896

London, **UK**, 1952

Major Smog Episodes Spurred Action to Clean Air in the US

Los Angeles, CA (1948)

Highest ozone recorded in 1955 (680 ppb)

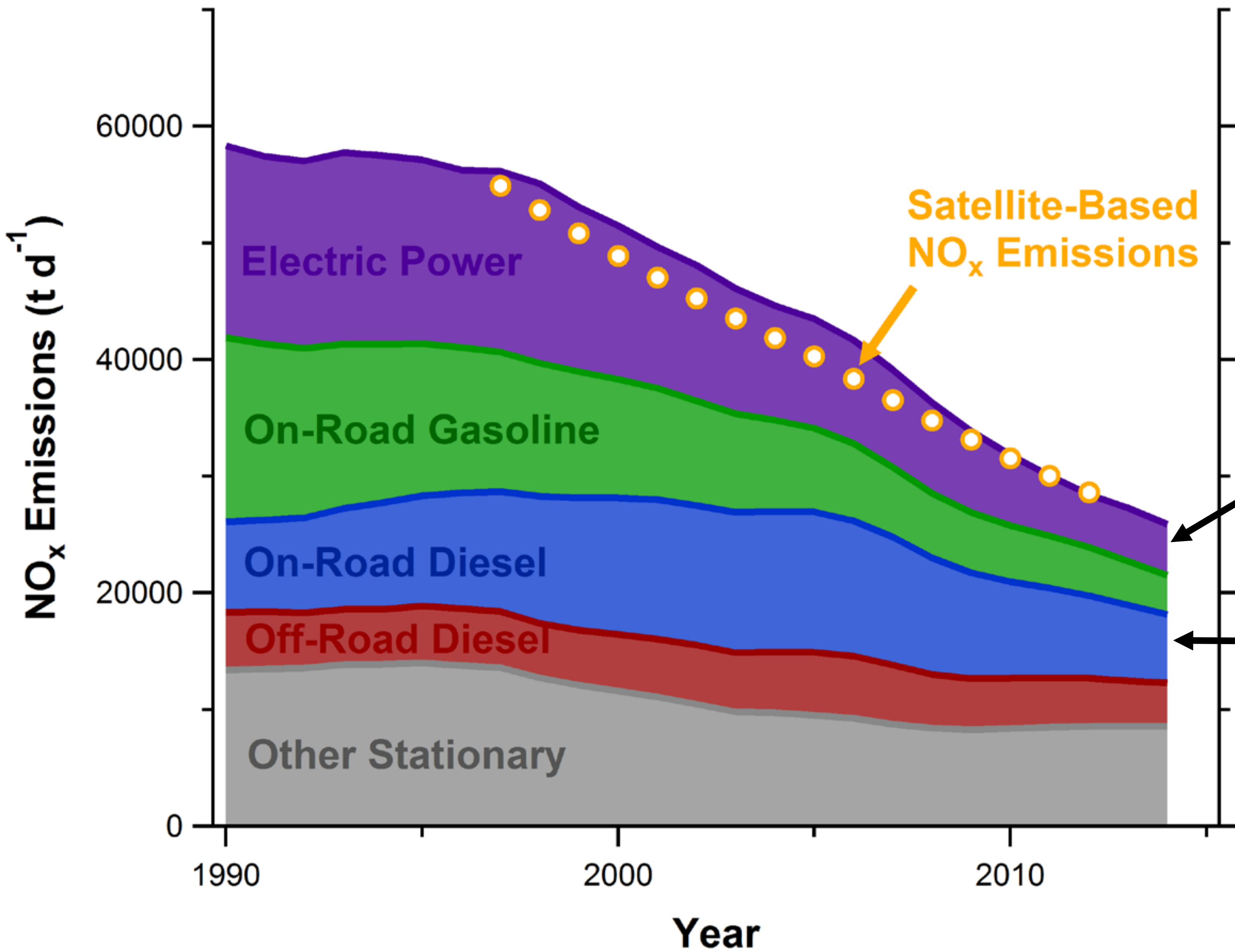
Automobiles

Donora, PA (1948)

20 people die from smog event

Stationary Sources

Trends in US Anthropogenic NO_x



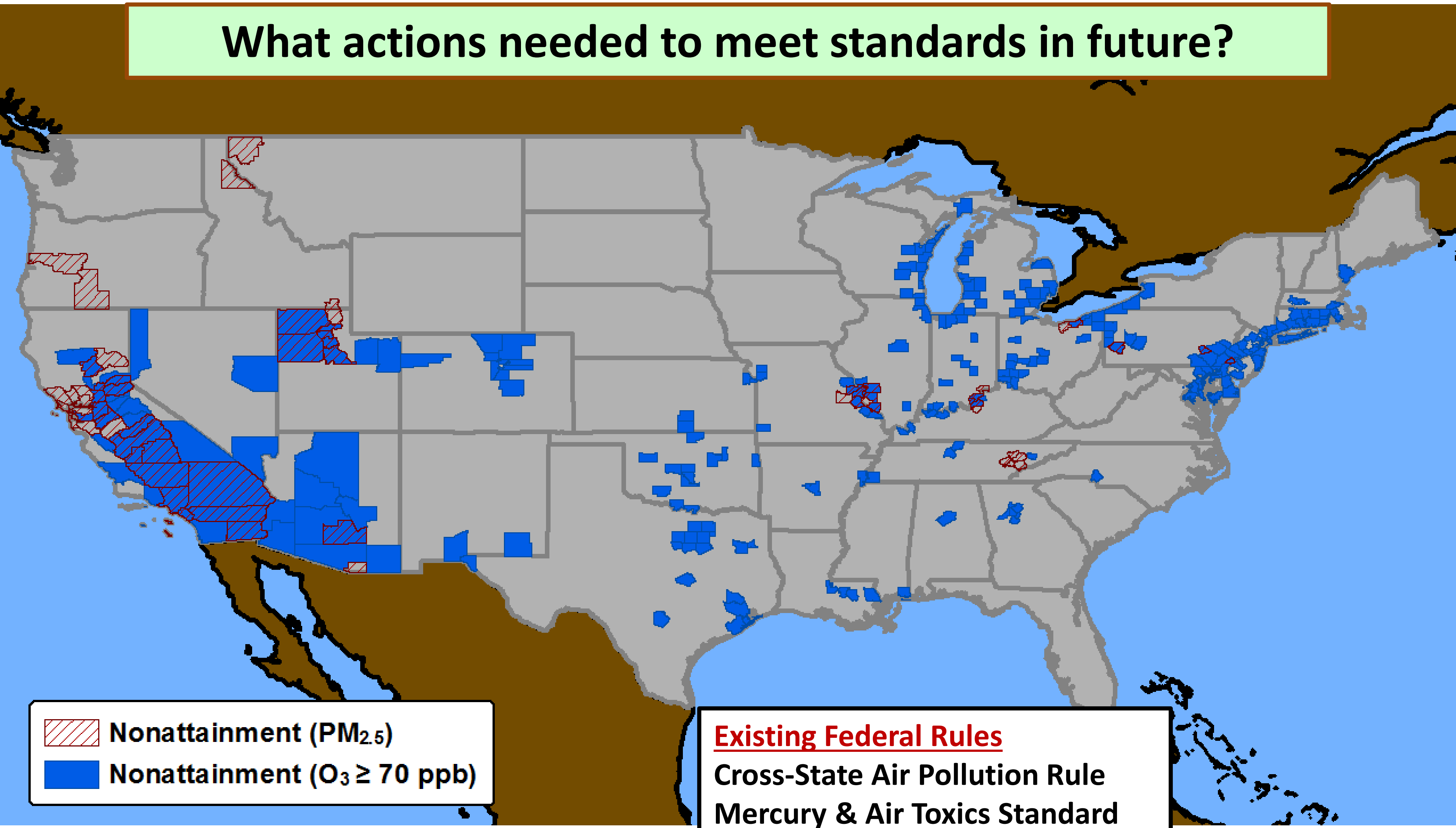
Largest reductions from cars and power plants

New controls implemented

Figure updated from McDonald et al. (2012) J. Geophys. Res.

Current US Non-Attainment Areas

What actions needed to meet standards in future?



Existing Federal Rules

Cross-State Air Pollution Rule
Mercury & Air Toxics Standard
Clean Power Plan
Vehicle & Fuel Standards

Recent observations/pictures of polluted regions

South America

from the
aqicn.org

Asia

Green: good **Yellow**: moderate
Red: unhealthy **Brown**: hazardous

Tiananmen Square, **Beijing**, 01/2013

What can be done ?
Identify and quantify the emissions of the compounds responsible for the pollution, understand their origin and reduce the corresponding emissions.

Emissions Needs in Atmospheric Research

- **Climate studies: impact of climate on emissions and of emissions on climate**
  - past/future realistic scenarios (**decades-century**)
  - **long-lived species**, aerosols and a few ozone precursors
  - emissions models or algorithms to take into account **land-use** and **human-related changes**

- **Distribution at global scale, impact long-range transport**
  - **limited number** of chemical species
  - moderate spatial (i.e. **1° x1°**) and temporal resolution
  - long-term variation (**a few decades**)
  - need some coupling emissions/meteorological conditions

- **Analysis and forecasting of atmospheric composition, observations from campaigns**
  - **wide range** of chemical species
  - high spatial (i.e. **0.1° x0.1° or few kms**) and temporal resolution

General Bottom-up Inventory Methodology

Inventories are an amalgam of calculations and measurements

Total mass emissions of compound X

$$E_X = \sum_S [EF_{X,S} \bullet A_S \bullet (1 - CE_{X,S})]$$

Sum up all sources S

Emissions factor = mass of compound X emitted by source S per unit activity; "representative" measurements or estimated

Activity of source S, e.g., amount of fuel burned, vehicle miles driven, etc.; measured or estimated

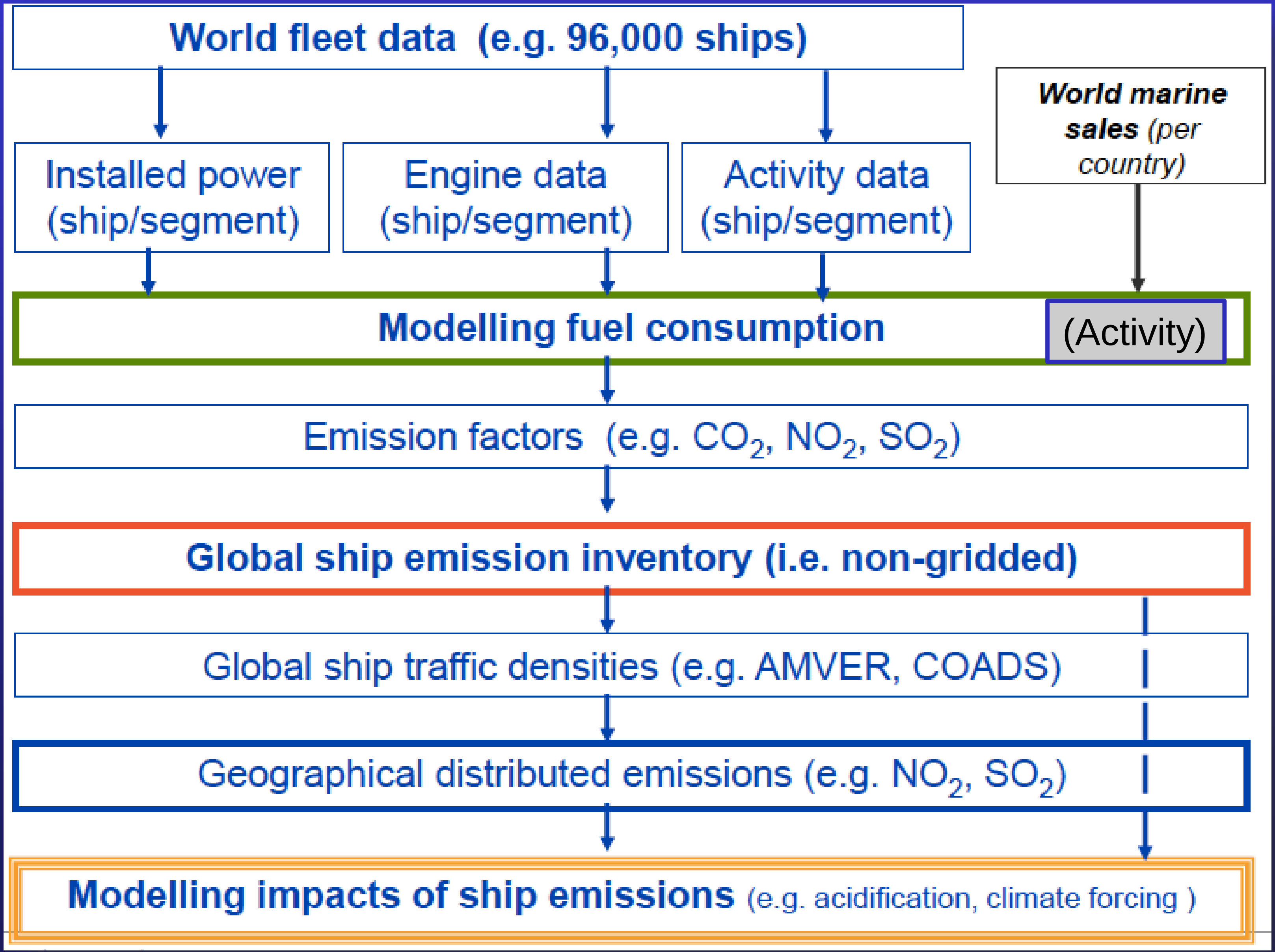
Effectiveness of control measures for cmpd X at source S; estimated or "representative" measurements

Above calculation carried out over a ***particular geographic region*** *(usually national scale, or state/country) for a* ***specific time period*** *(usually annual or monthly)*

Additional steps usually needed to use inventory in modeling or analysis

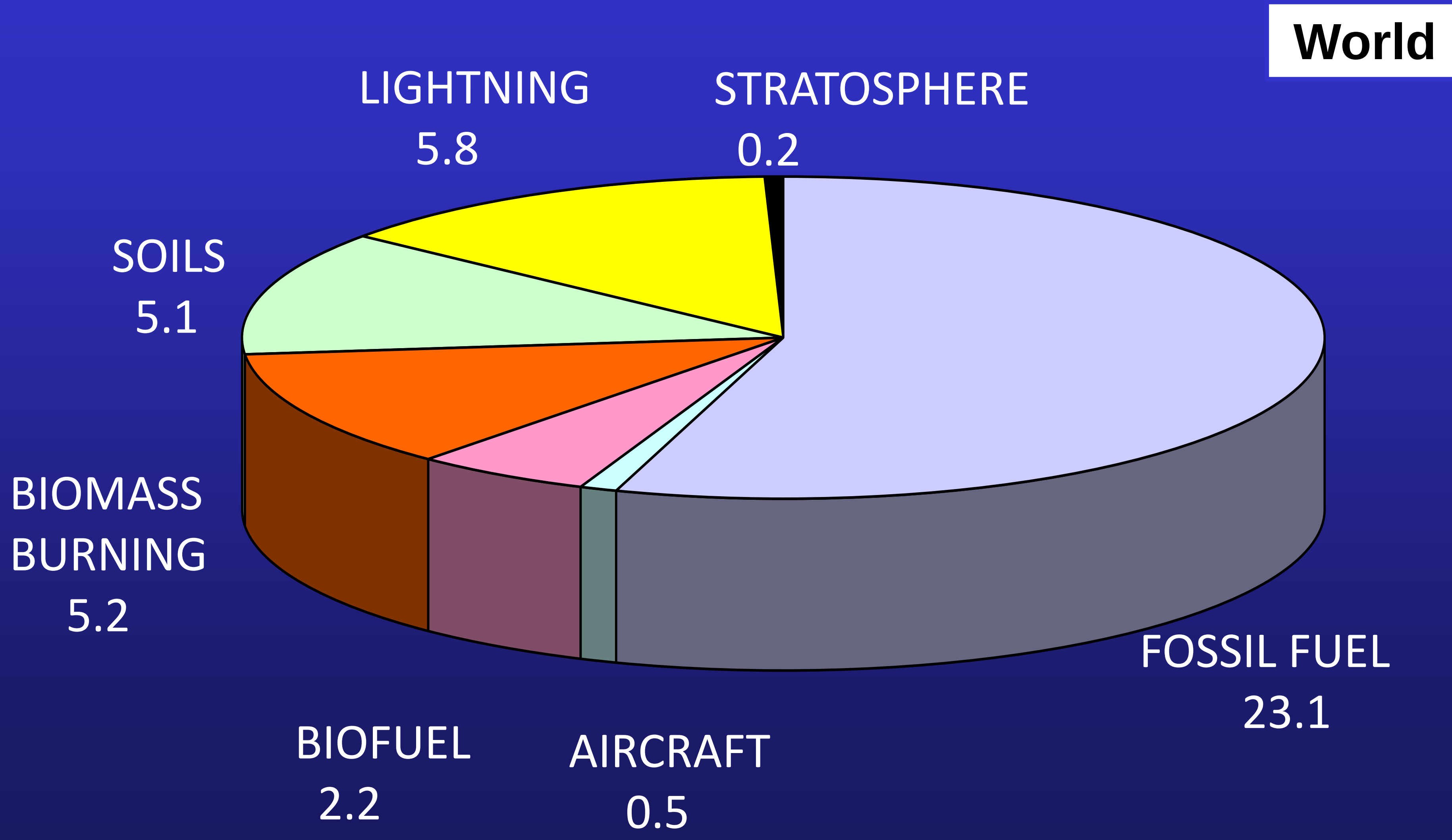
- ***Spatial Allocation***: gridding, spatial scaling
- ***Temporal Variability:*** between and subdivisions of inventory periods
- ***Temporal Extrapolation:*** projections, scenarios
- ***Speciation:*** VOCs, PM, …

Example: Emissions from ships



Emissions have to be quantified for many different sectors:

example => NO_x emissions (Tg N yr^{-1})



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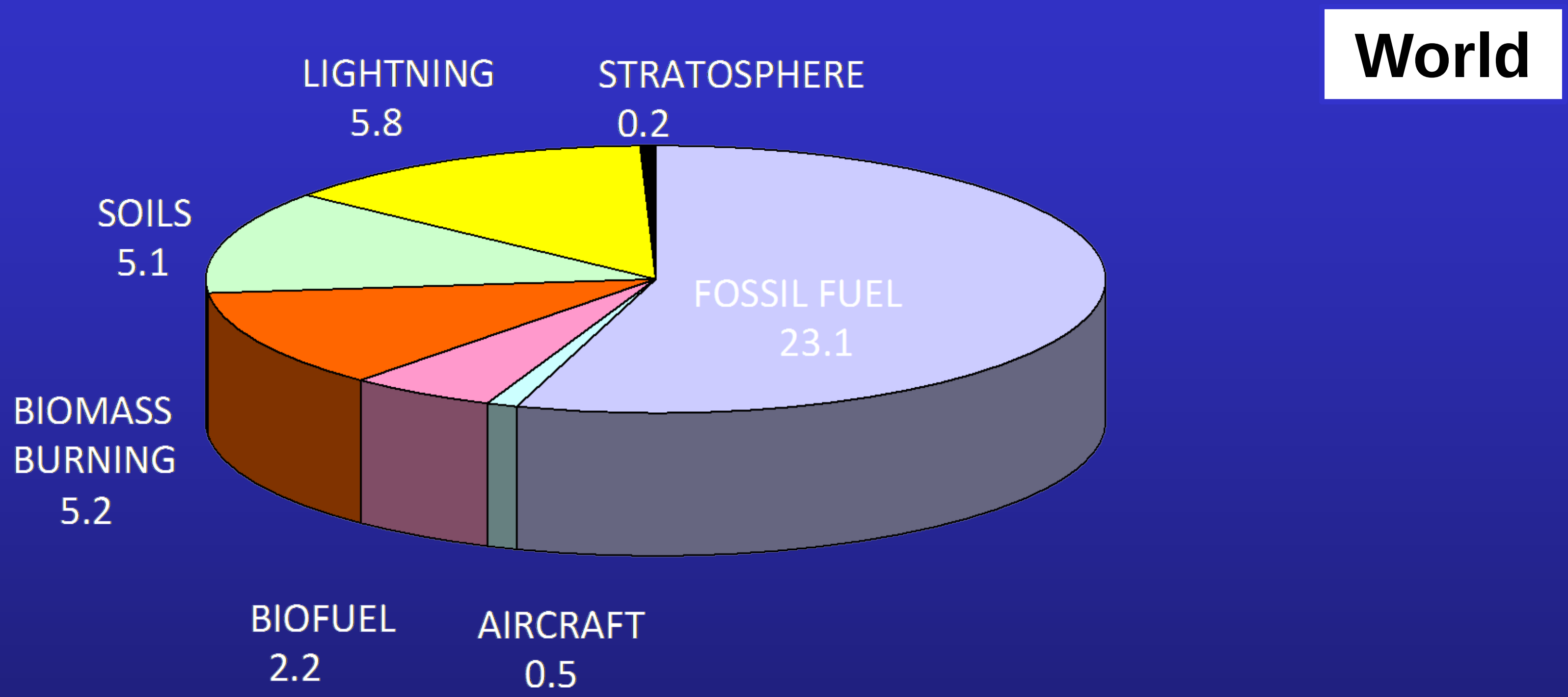
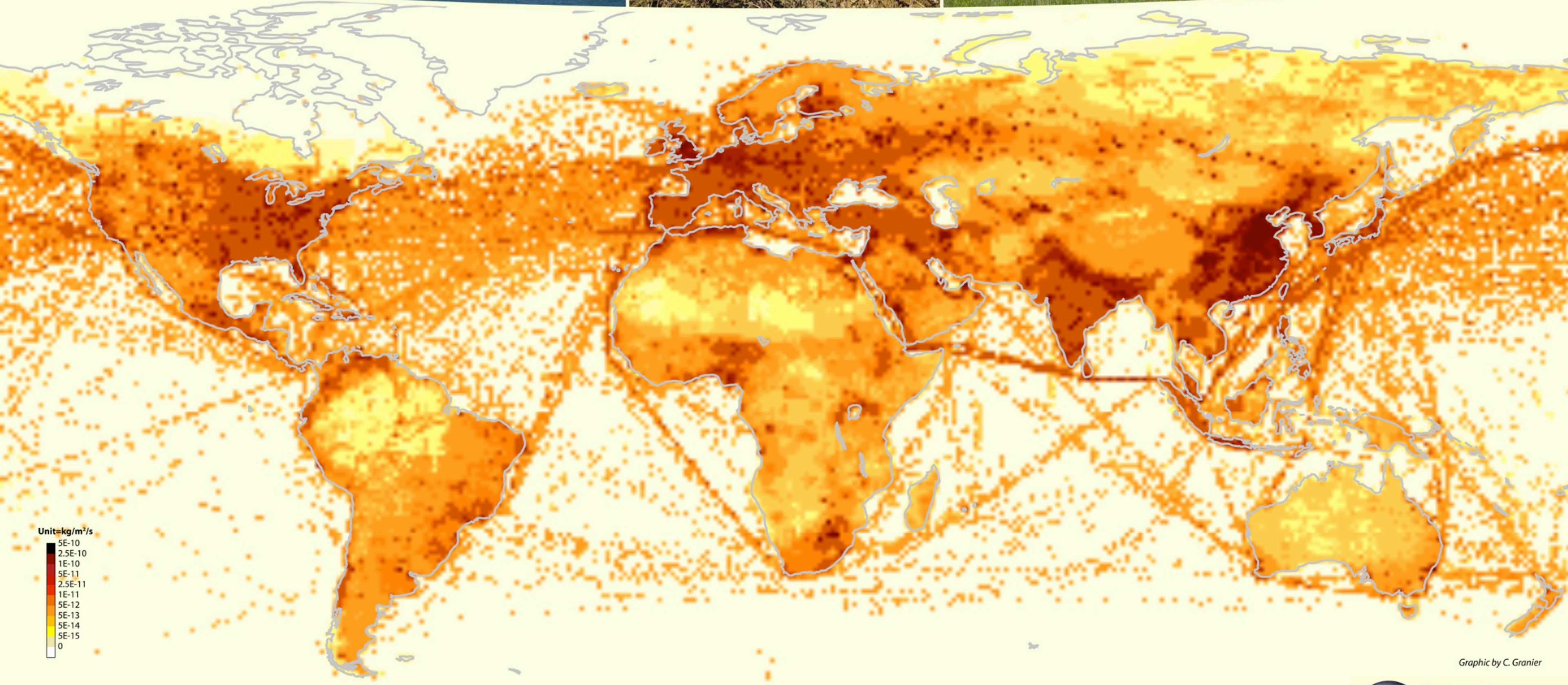


Be careful with units:

Fossil fuel emissions = 23.1 Tg N/yr
which means 49.5 Tg NO/yr or 75.9 Tg NO_2/yr

Very often, papers only say Tg NO_x/yr → ??



Graphic by C. Granier

Emissions of Nitrogen Oxides from all Anthropogenic Activities in 2010

Data provided by Claire Granier and Gregory Frost.

The MACCity inventory quantifies trace gases and aerosols emitted by a variety of human activities, including energy production, industry, transportation and agriculture.

GEIA
Global Emissions InitiAtive

A list of recent global inventories

Author	Acronym	Reference or Website	Years	Resolution
Lamarque et al.	ACCMIP	eccad.sedoo.fr	1900-2000	0.5x0.5
Riahi et al.	RCPs	eccad.sedoo.fr	2000-2100	0.5x0.5
Granier et al.	MACCity	eccad.sedoo.fr	2000-2015	0.5x0.5
Maenhout et al.	EDGAR4.2	edgar.jrc.ec.europa.eu	1970-2008	0.1x0.1
Crippa et al.	EDGAR4.3	edgar.jrc.ec.europa.eu	1970, 2010	0.1x0.1
Maenhout et al.	HTAPv2	edgar.jrc.ec.europa.eu	2008, 2010	0.1x0.1
Klimont et al.	ECLIPSE v4, v5	iiasa.ac.at	1990-2030	0.5x0.5
Schultz et al.	RETRO	juelich ftp	1960-2000	0.5x0.5
Bond et al.	Bond	Hiwater.org	1850-2000	country
Junker&Liousse	J&L	eccad.sedoo.fr	1860-2003	1x1
Huang Y. et al.	PKU	inventory.pku.edu.cn	1960-2009	0.1x0.1
Smith et al.	PNNL	sedac.ciesin.columbia.edu	1850-2005	1x1

Blue: inventories providing just a few species

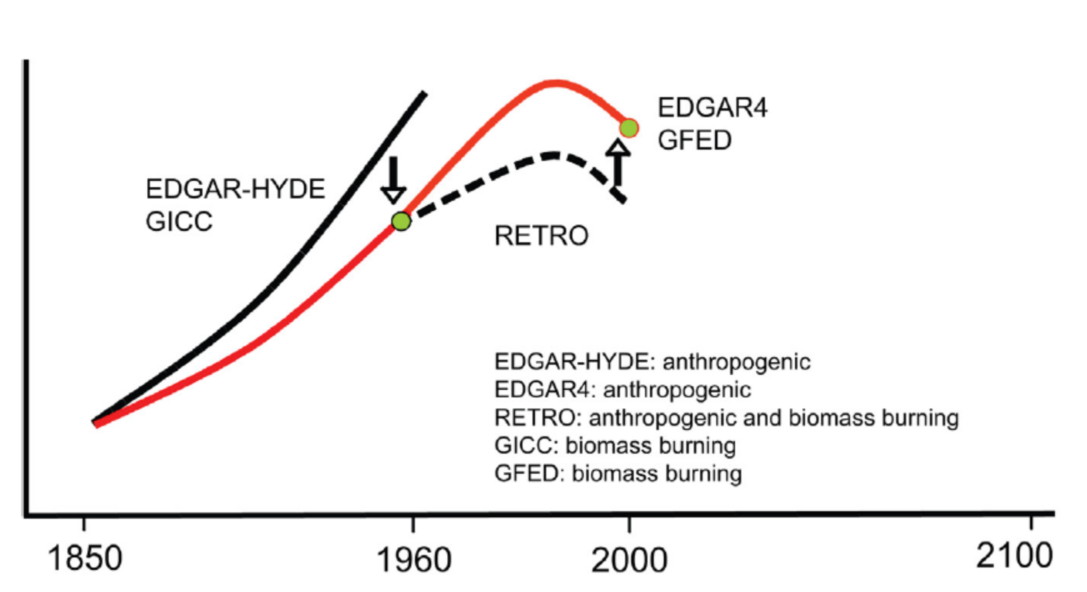
All the data are publicly available. Most available at: eccad.sedoo.fr, the database of the **Global Emissions InitiAtive (GEIA)**

ACCMIP = A long-term emissions dataset developed for IPCC AR5

(Details in Lamarque et al., ACP, 2010)

ACCMIP: Emissions for Atmospheric Chemistry and Climate Modeling Intercomparison Project

- Anthropogenic & biomass burning emissions
- Period: 1850-2000
- Available for each decade
- No seasonal variation
- 0.5° x0.5° resolution



IPCC: Intergovernmental Panel on Climate Change

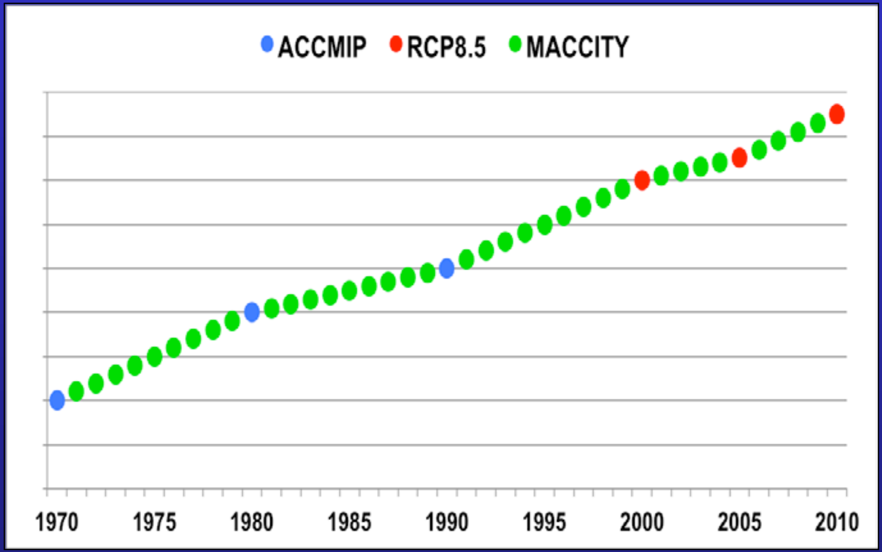
MACCity = anthropogenic and biomass burning emissions

(Details in Granier et al., Climatic Change, 2011)

Developed as part of MACC, MACCII and Cityzen

Uses RCPs IPCC scenario for 2005, 2010, 2015

- Period: 1960-2016; Monthly averages
- 0.5° x0.5° resolution
- Species: CH_4, CO, NO_x, SO_2, BC, OC, NH_3 & a large set of VOCs (new VOCs generated, based on users requests)
- different emissions sectors



Large community of users:

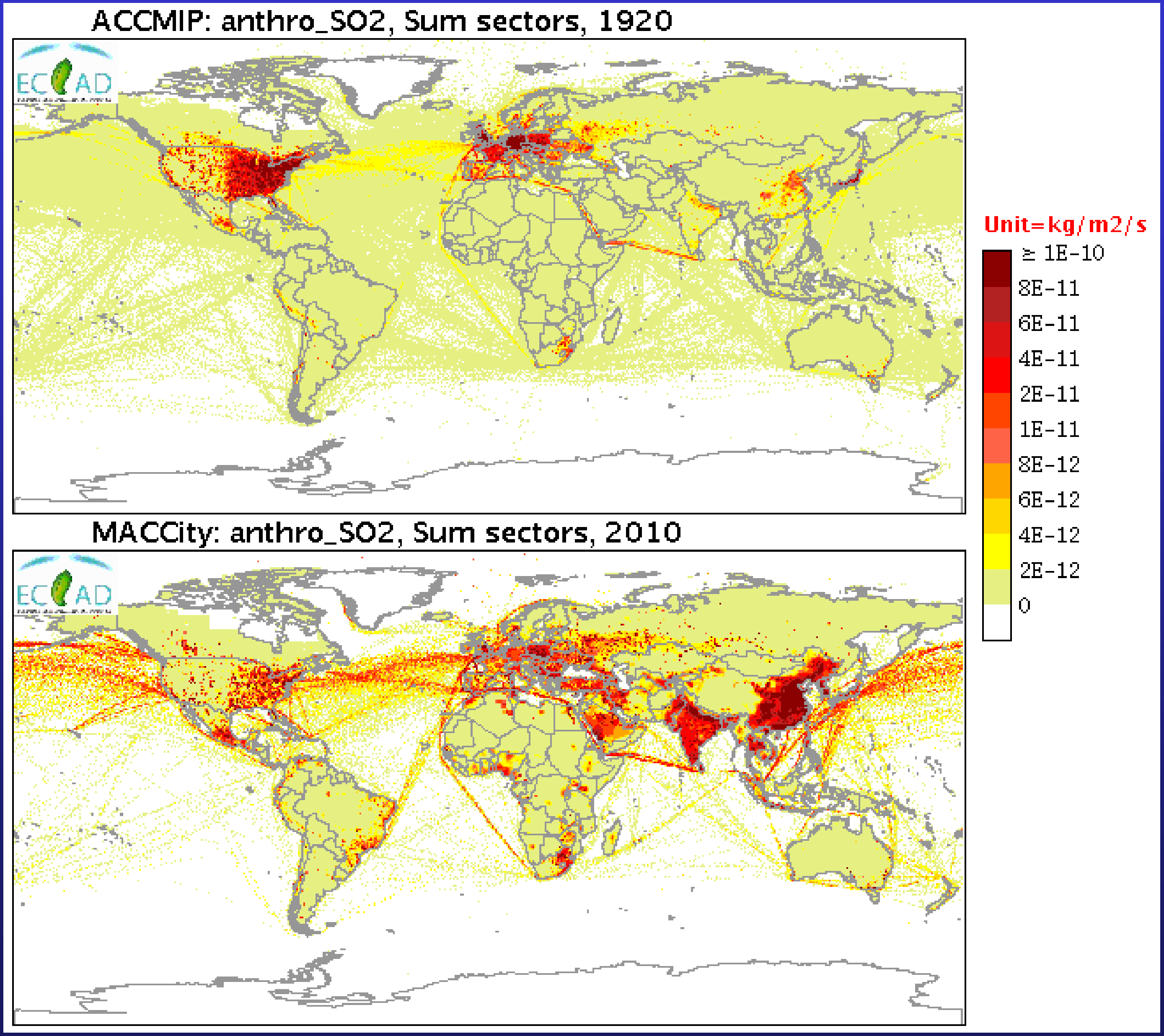
→ EU projects: PEGASOS, ACCESS, ACCENT-Plus, CAMS, etc.

→ International modelling projects: CCMI (chemistry-climate, hindcast), CMIP5 (IPCC), AEROCOM (Aerosols), etc.

→ International programs: IGAC, iLEAPS, GEIA, etc.

→ Individual laboratories in Europe and elsewhere

Example of the ACCMIP/MACCity emissions: change in SO_2 from 1920 to 2010



Developed at JRC, Ispra, Italy

EDGARv4.2 (1970-2008)

Direct greenhouse gases

CO_2, CH_4, N_2O,

HFCs, PFCs, SF_6, NF_3

Stratospheric ozone depletion substances

HCFCs

Ozone precursors

NMVOC, CO, NO_x, CH_4

Acidifying gases

NH_3, NO_x, SO_2

Primary Particulates

PM_{10}

Resolution: 0.1° x 0.1°

EDGARv4.2 FT2010 (2000-2010)

CO_2

CH_4

N_2O

F-gases:

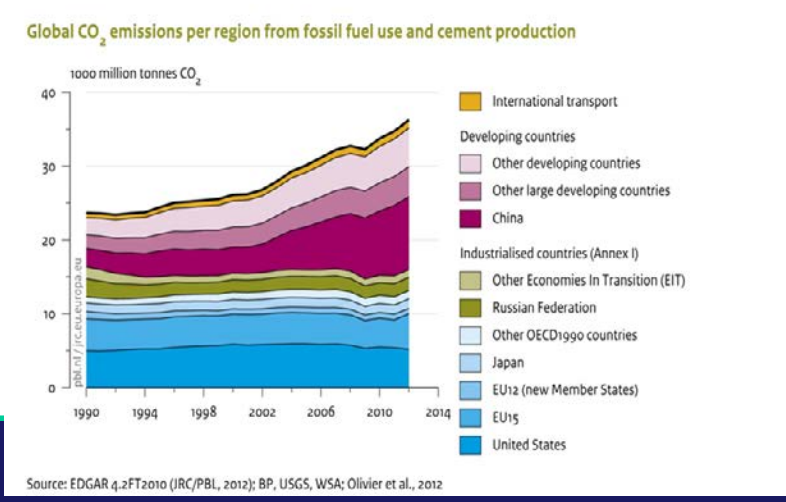
-HFC_s

-PFC_s

-SF_6

EDGAR CO_2 report

CO_2 trends until 2012

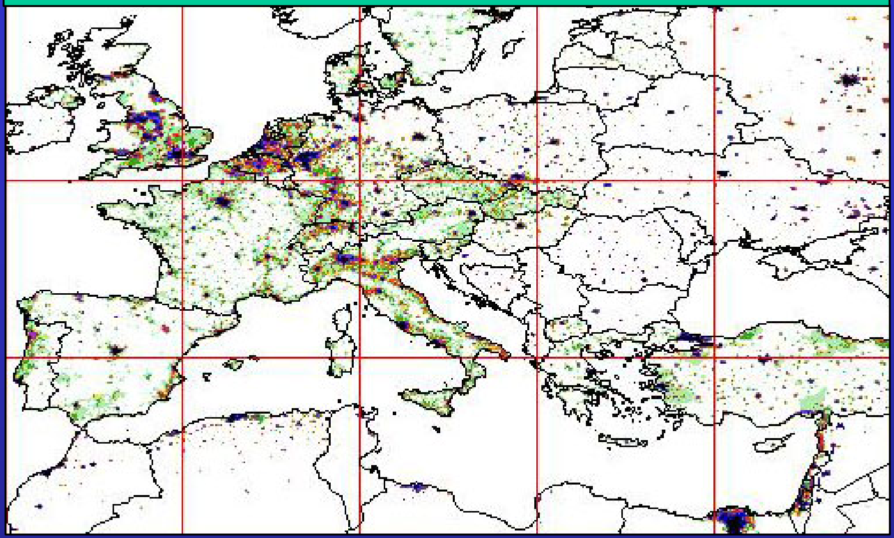


EDGAR

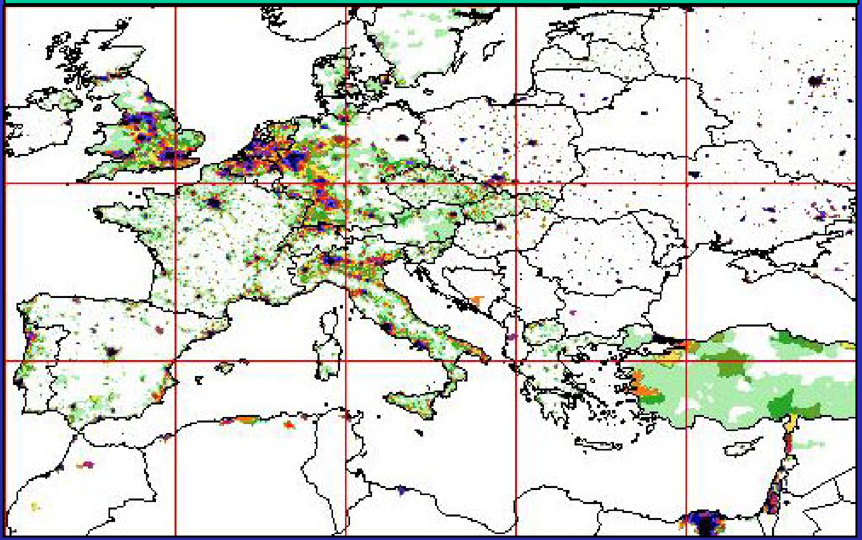
EMISSION DATABASE FOR GLOBAL ATMOSPHERIC RESEARCH

Downscaling→ 0.01°x0.01°

Urban population calculated using GRUMPv1

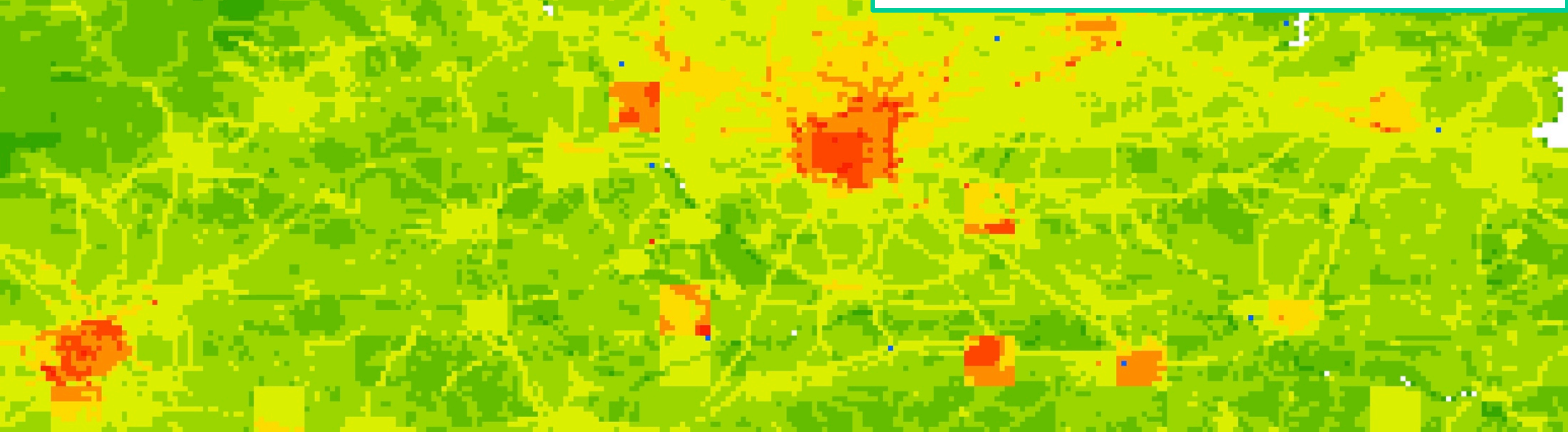


Urban population calculated using GPWv3



Turin-Milan CO_2 at 0.01°x0.01°

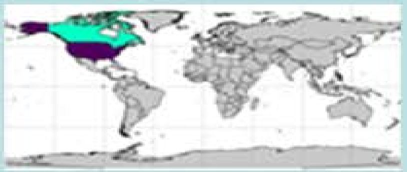
http://edgar.jrc.ec.europa.eu/



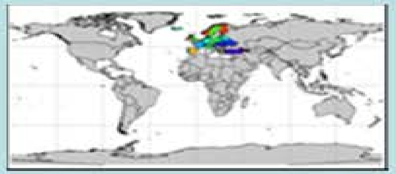
HTAP_v2

HTAPv2.2 = harmonised mosaic of official & semi-official 0.1°x0.1° monthly gridmaps, gapfilled for global coverage

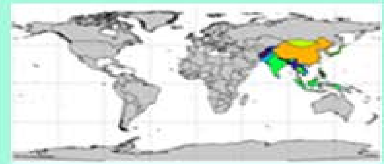
USEPA/EnvironCanada



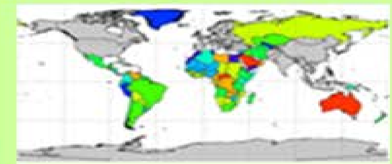
EMEP-TNO/MACC



MIX-Asia



EDGARv4.3



Online: HTAP_v2.2 of 18/12/13 on ftp://edgar@edgar.jrc.ec.europa.eu or ECCAD

	EMEP	TNO	USEPA	MIX	EDGAR
CO	grid /yr	grid /yr	grid /m /height	grid /m (no AWB)	grid /m
NMVOC	grid /yr	grid /yr	grid /m /height/species	grid /m	grid /m
NOx	grid /yr	grid /yr	grid /m /height	grid /m (no AWB)	grid /m
SO2	grid /yr	grid /yr	grid /m /height	grid /m (no AWB)	grid /m
NH3	grid /yr	grid /yr	grid /m /height	grid /m	grid /m
PM10	grid /yr	grid /yr	grid /m /height	grid /m (no AWB)	grid /m
PM2.5	grid /yr	grid /yr	grid /m /height	grid /m (no AWB)	grid /m
BC		grid /yr	grid /m /height	grid /m (no AWB)	grid /m
OC		grid /yr	grid /m /height	grid /m (no AWB)	grid /m

International Institute for Applied Systems Analysis
IIASA www.iiasa.ac.at

Austria

science for global insight

PEGASOS
Pan-European Gas-AeroSOls-climate interaction Study

ECLIPSE

Global air pollutant emission scenarios 1990-2050; ECLIPSE

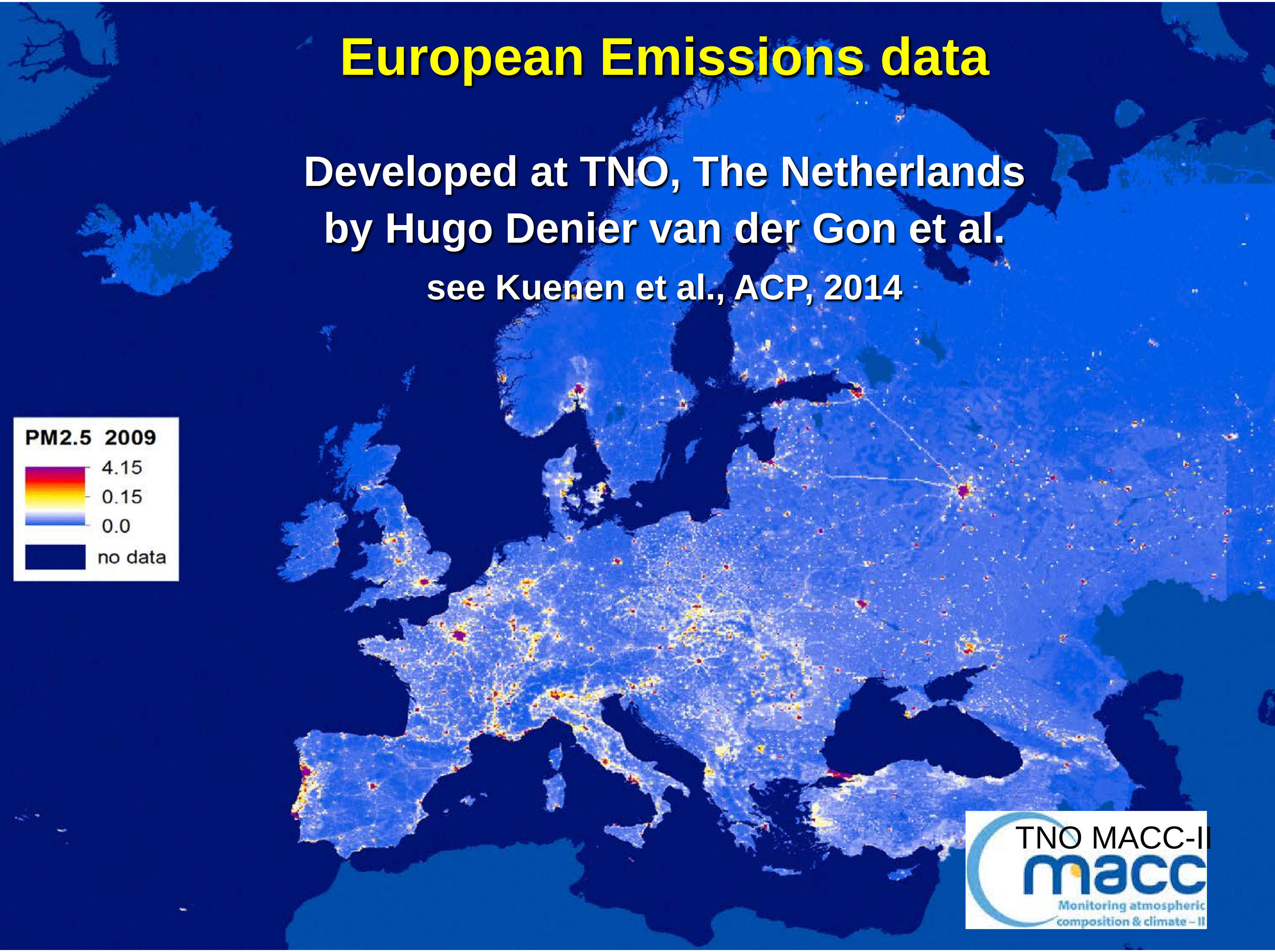
Z. Klimont, C. Heyes, L. Höglund-Isaksson, J. Cofala, P. Rafaj, W. Schöpp, P. Purohit, J. Borken, M. Amann, K. Kupiainen, W. Winiwarter, I. Bertok, R. Sander, B. Zhao, S. Wang

- Development driven by the need to improve aerosols emissions in long term IAM scenarios
- Multi-pollutant fine resolution inventory and projections including technology resolution (annual, monthly)
- Developed for a range of policies
- ‘New’ sources included, e.g., shale gas, gas flaring, wick lamps, diesel generators, super-emitters
- Public access to gridded data
- Platform for further set of scenarios including also estimate of mitigation costs

International Institute for Applied Systems Analysis
IIASA

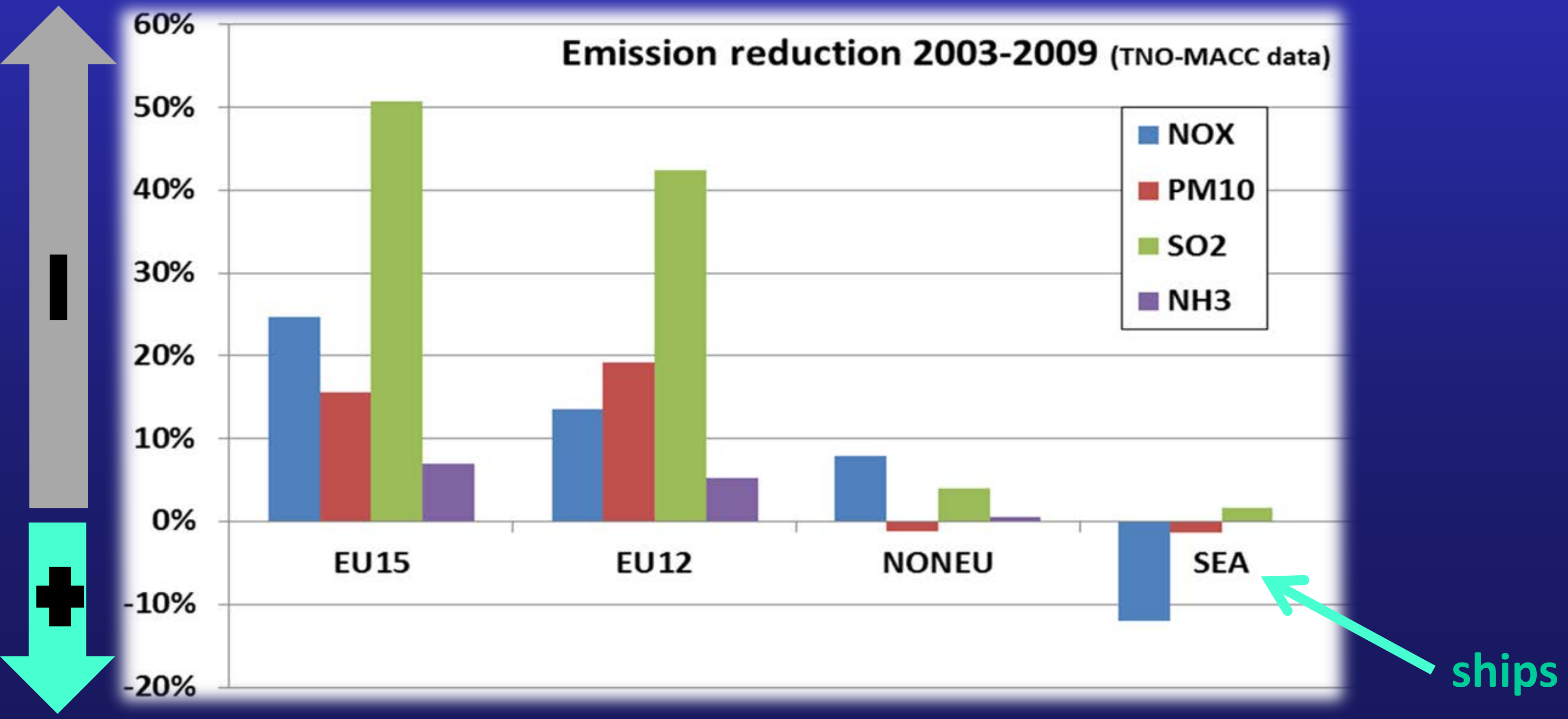
European Emissions data

Developed at TNO, The Netherlands
by Hugo Denier van der Gon et al.
see Kuenen et al., ACP, 2014



European anthropogenic emissions: TNO-MACC 2003-2009 emissions

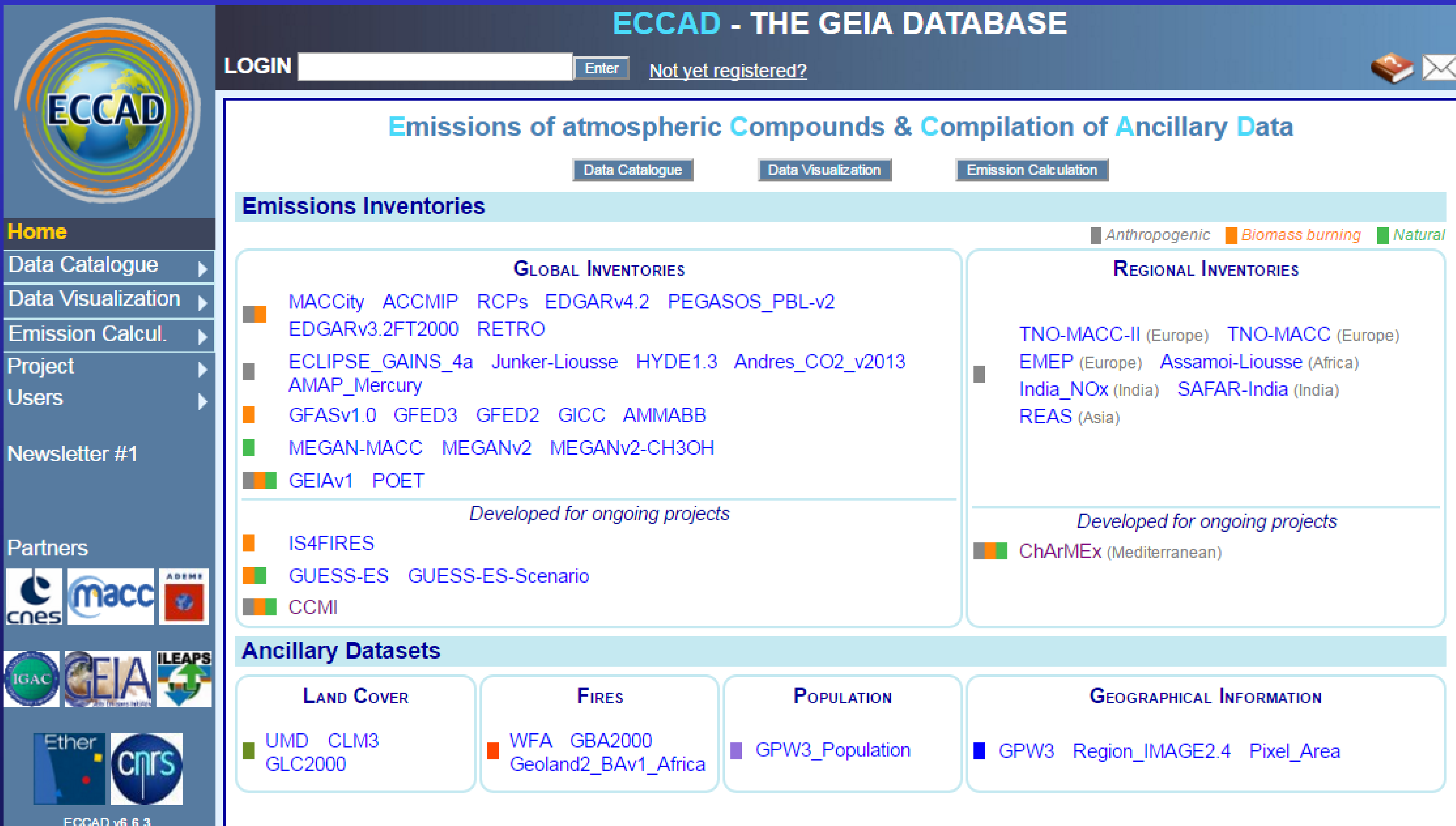
- "Processing" of country emission data to construct a consistent time series for **reanalysis & European forecasting**
- Resolution is 7x7 km and analysis of trends have been provided
- Inclusion of point source year-to-year changes



Where can you get most of these emissions ?

=> ECCAD : Atmospheric Compounds and Compilation of Ancillary Data

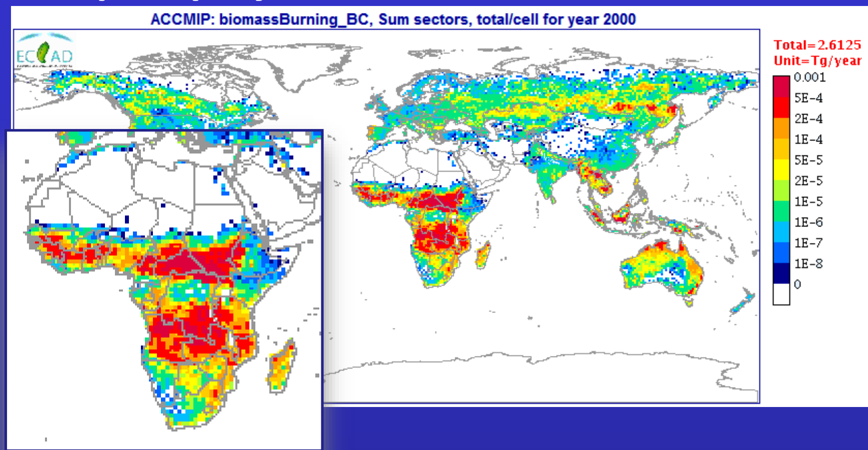
http://pole-ether.fr/eccad



ECCAD Visualisation and On-line Tools

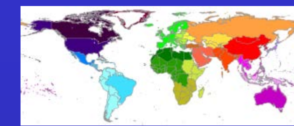
http://pole-ether.fr/eccad

Map display (zoom, scaling, color palette)

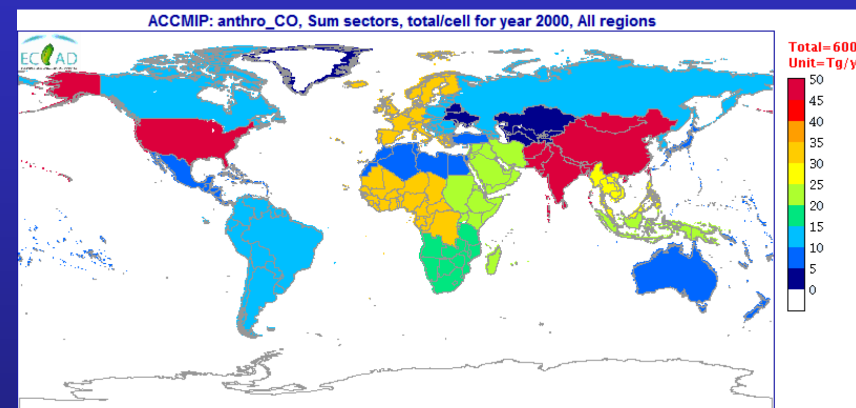


Regional Analysis

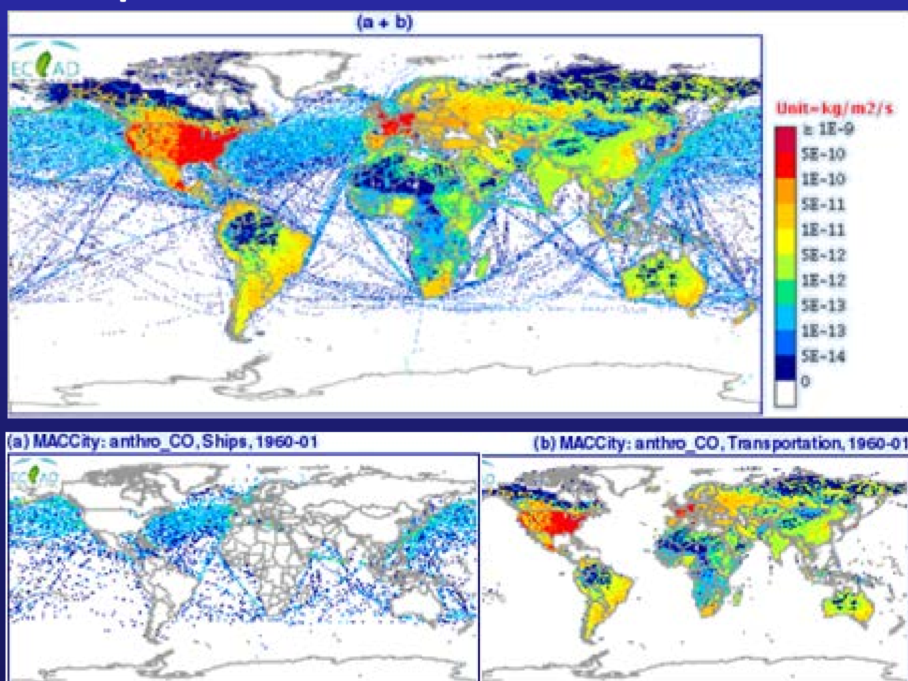
- Countries
- Continent
- User-defined regions



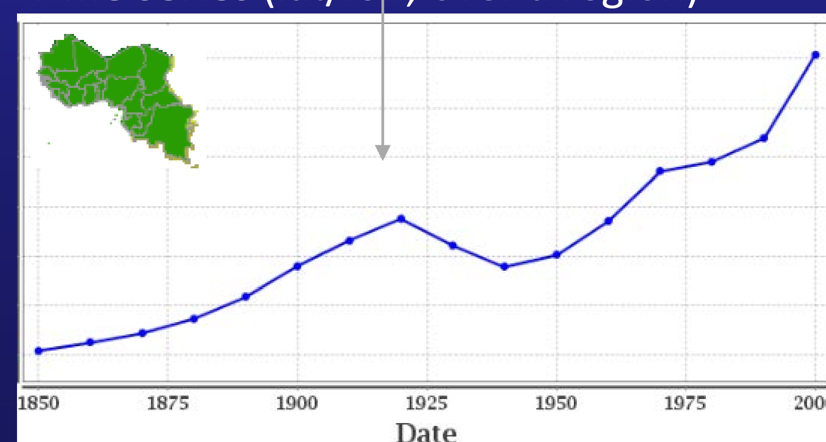
Total global/regional emissions



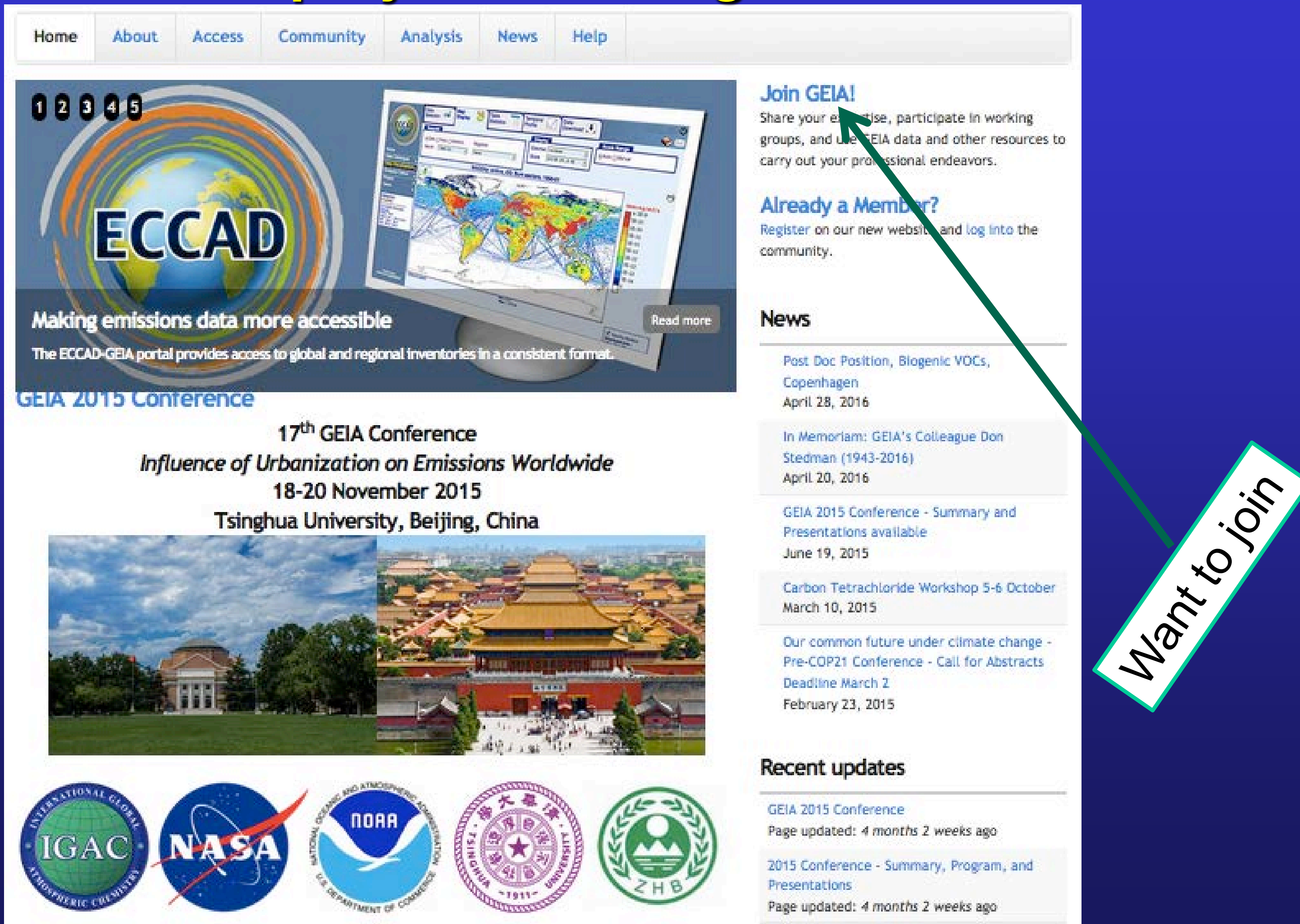
Comparisons of datasets



Time series (lat/lon, over a region)

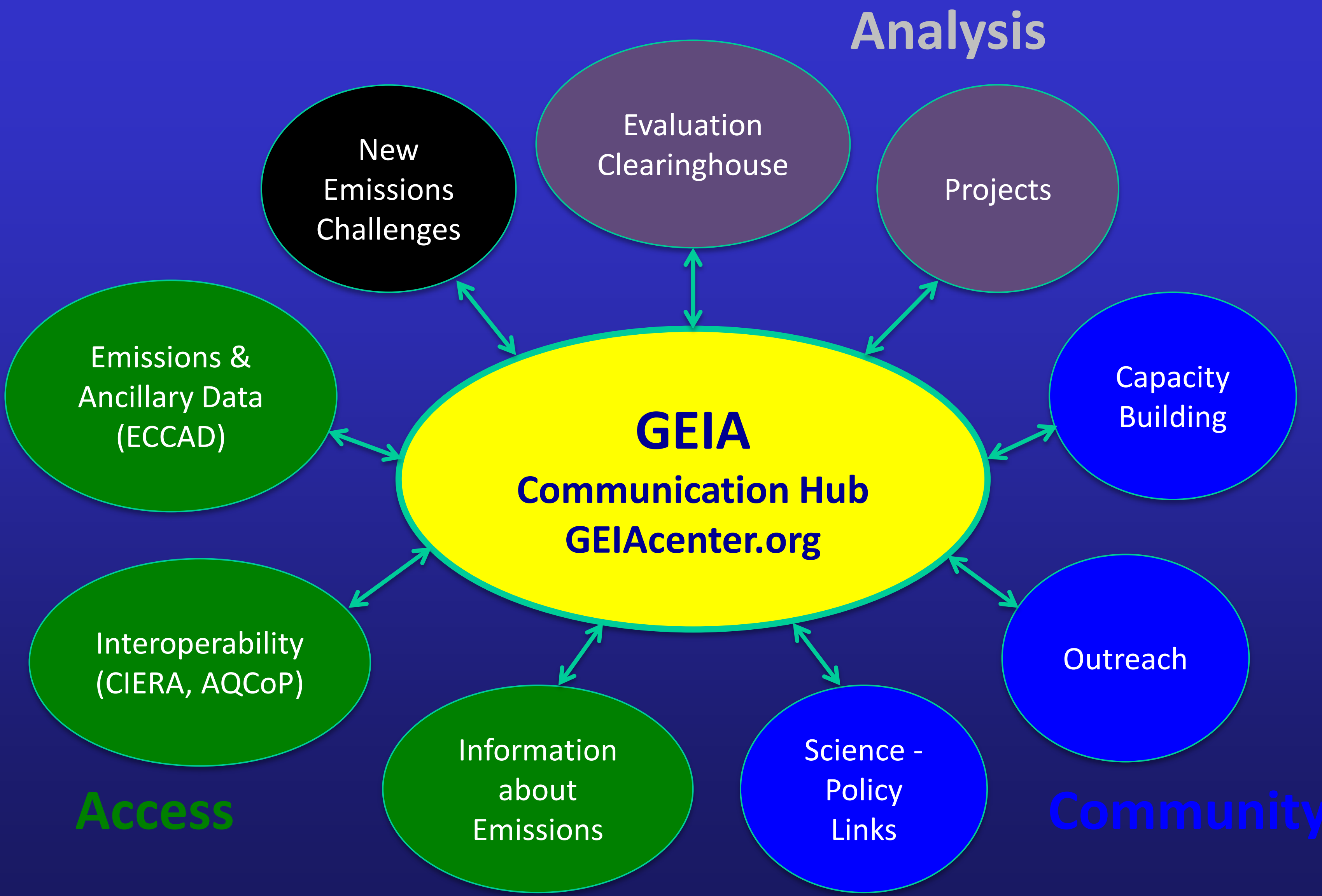


GEIA: The Global Emissions InitiAtive
International project focusing on emissions



GEIA Activities

http://www.geiacenter.org/



GEIA Working Groups

China Emissions WG

Contacts: Kebin He, Qiang Zhang, Yuxuan Wang

- Improving scientific basis for emissions from China
- Coordinating Chinese groups working on emissions
- Contributing to East Asian Emissions Assessment *(ACP)*

VOC Emissions WG

Contacts: Erika von Schneidemesser, Hugo Denier van der Gon

- Improve global understanding of VOC emissions speciation
- Leverage on-going VOC measurements, inventories, modelling
- Evaluate most important VOC source sectors

Latin America/Caribbean (LAC) Emissions WG

Contacts: Nicolas Huneeus, Laura Dawidowski, Néstor Rojas

- Develop and evaluate LAC-specific emissions information
- Create LAC regional emissions database and inventory
- Build LAC emissions expert community & link to global efforts

Urban Emissions WG

Contacts: Leonor Tarrasón

- Bring together techniques for urban emissions characterization
- Build capacity in megacities around the world

http://geiacenter.org

IGAC - Providing Global Leadership

IGAC's Activities

IBBI

GEIA
Global Emissions InitiAtive

ccmi
chemistry-climate model initiative

AICI
AIR-ICE CHEMICAL INTERACTIONS

PACES
air Pollution in the Arctic:
Climate Environment and Societies

DEBITS
Deposition of Biogeochemically Important Trace Species

FUNDAMENTALS
ATMOSPHERIC CHEMISTRY

TOAR
tropospheric ozone assessment report

OASiS

ACAM
ATMOSPHERIC COMPOSITION & THE ASIAN MONSOON

14th Biennial IGAC Science Conference
26-30 September 2016

www.igac2016.org

Part 2. Inventory Evaluations and Top-Down Approaches

- Inventory comparisons
- Challenges with bottom-up inventories
- Using top-down approaches to understand emissions
- Examples of atmospheric observations used in top-down analyses

1. How good are all these inventories?

2. Are any of these emissions accurate?

3. If I use one dataset vs. another in my model or analysis of observations, will I get better results?

→ A systematic evaluation of surface emissions has started within GEIA (Global Emissions InitiAtive): geiacenter.org

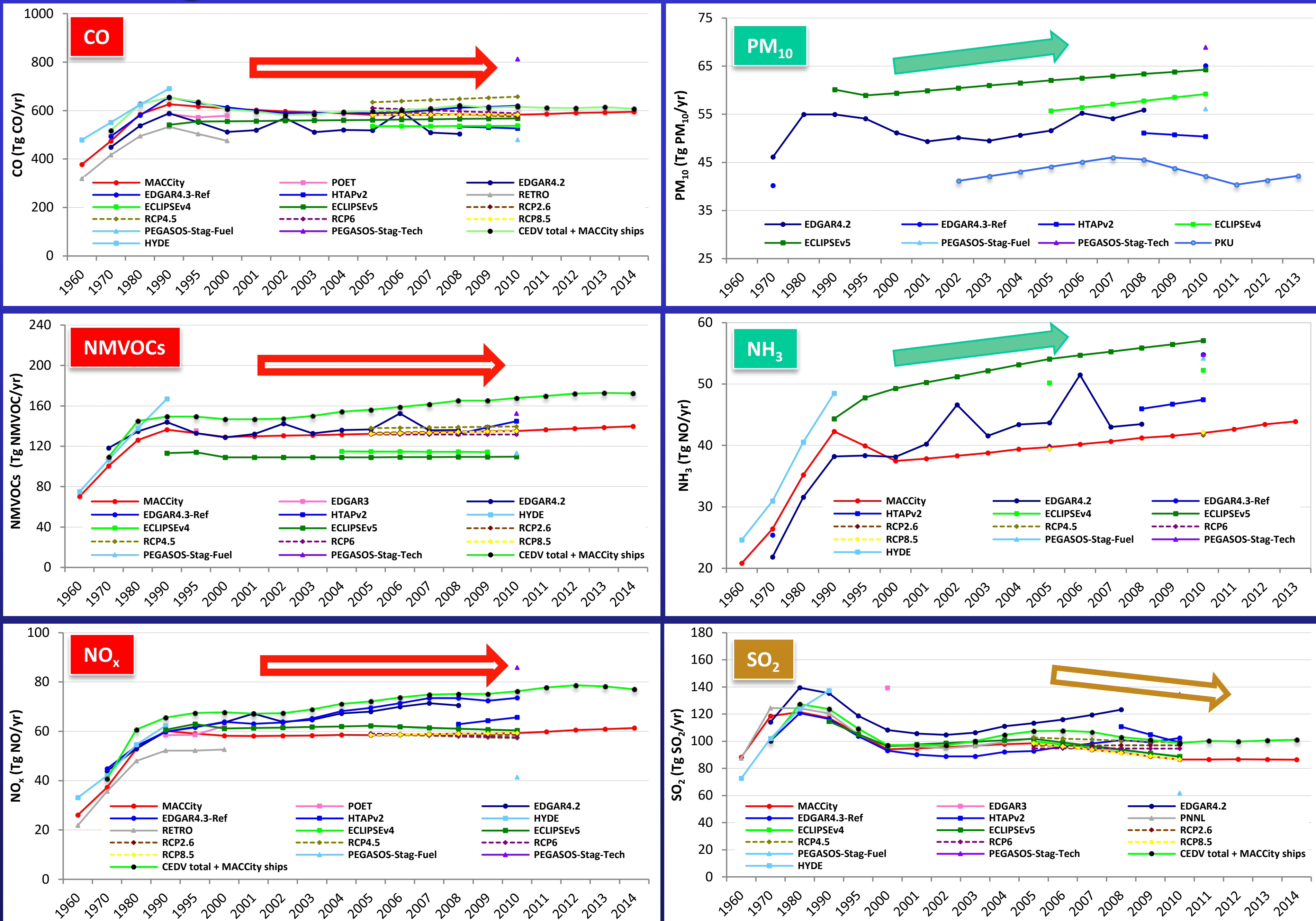
✓ Only publicly available gridded inventories considered (for China: global inventories + REAS v1, v2, MEIC/MIX)

✓ Focus on: CO, NO_x, SO_2, BC, total NMVOCs, NH_3 and PM_{10}

For more information, talk to Claire Granier (Claire.Granier@noaa.gov)

Total global emissions

Claire Granier et al.



Emissions for Asia considered for the evaluation

Author	Acronym	Reference or Website	Years	Resolution
Q. Zhang et al.	MEIC	Meicmodel.org	2008, 2010, 2012	0.25x0.25
Ohara et al.	REASv1	jamstec.go.jp	1980-2003	0.5x0.5
Kurokawa et al.	REASv2	nies.go.jp/REAS	2000-2008	0.25x0.25
Zhang et al.	TRACE-P INTEX-B	cgrer.uiowa.edu	2000, 2006	0.5x0.5
Li et al.	MIX	http://www.meicmodel.org/ dataset-mix	2008, 2010	0.25x0.25
B. Zhao et al.	ZhaoB	ACP, 13, 9869, 2013	1995-2010	country
Y. Zhao et al.	ZhaoY	ACP, 13, 4872013	2005, 2010	country
Cao	Cao	Atm. Env, 40, 6516, 2006	2000	0.2x0.2
X Huang et al.	HuangX	Glob. Bio. Cyc, 26, 2012	2006	1x1 km
Kang et al.	Kang	ACPD, 15, 26959, 2015	1980-2012	1x1 km
Bo et al.	Bo	ACP, 8, 7297, 2008	1980-2005	40x40 km
Lei et al.	Lei	ACP, 11, 931, 2011	1990-2005	30'x30'
Wei et al.	Wei	Atmos. Env., 2011	2005-2020	36x36 km
Su et al.	Su	Env. Sci. Tech.,	1990-2007	province
Lu et al.	Lu	ACP, 10, 6311, 2010	1996-2010	province
R. Wang et al.	WangR	Env. Sci. Tech., 46, 2012	1949-2007	0.1x0.1
S. Wang et al.	WangS	ACP, 14, 6571, 2014	2005-2030	country

+ PKU (Peking University) global inventory already listed before

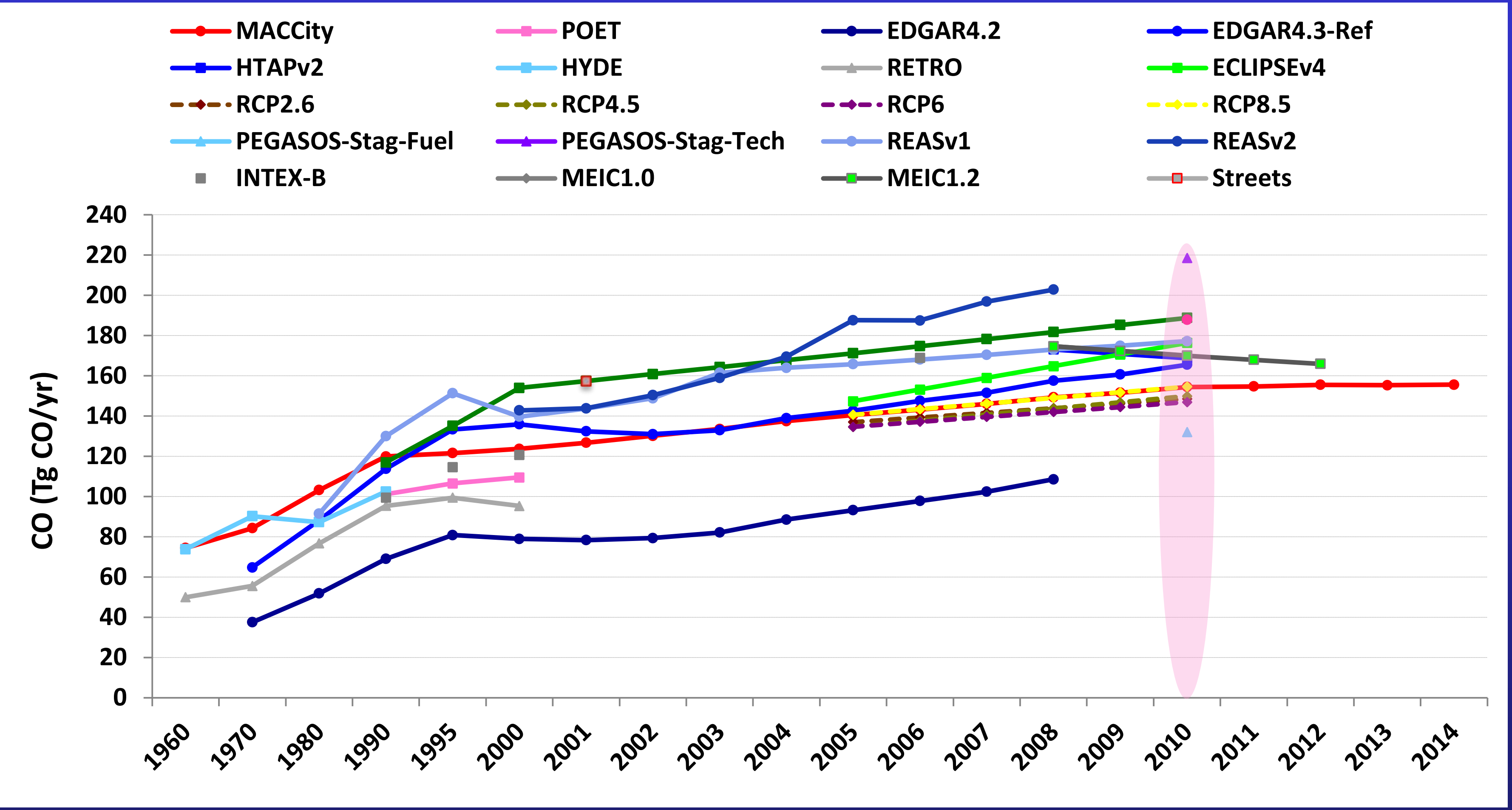
Inventories publicly available

Others: generally just one publication, no information if data are available somewhere

Claire Granier et al.

Trends in CO Anthropogenic Emissions

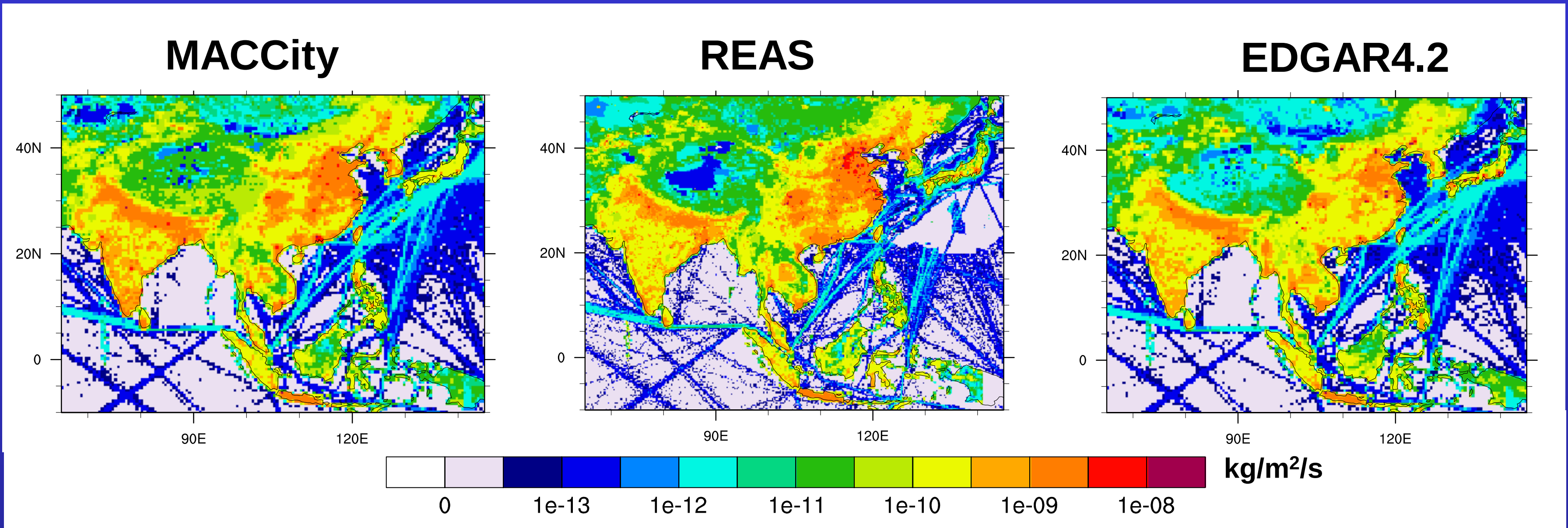
China



- ★ Large differences in emission levels as well as in trends
- ★ Some emissions agree very well with some IPCC scenarios or are very different for other scenarios

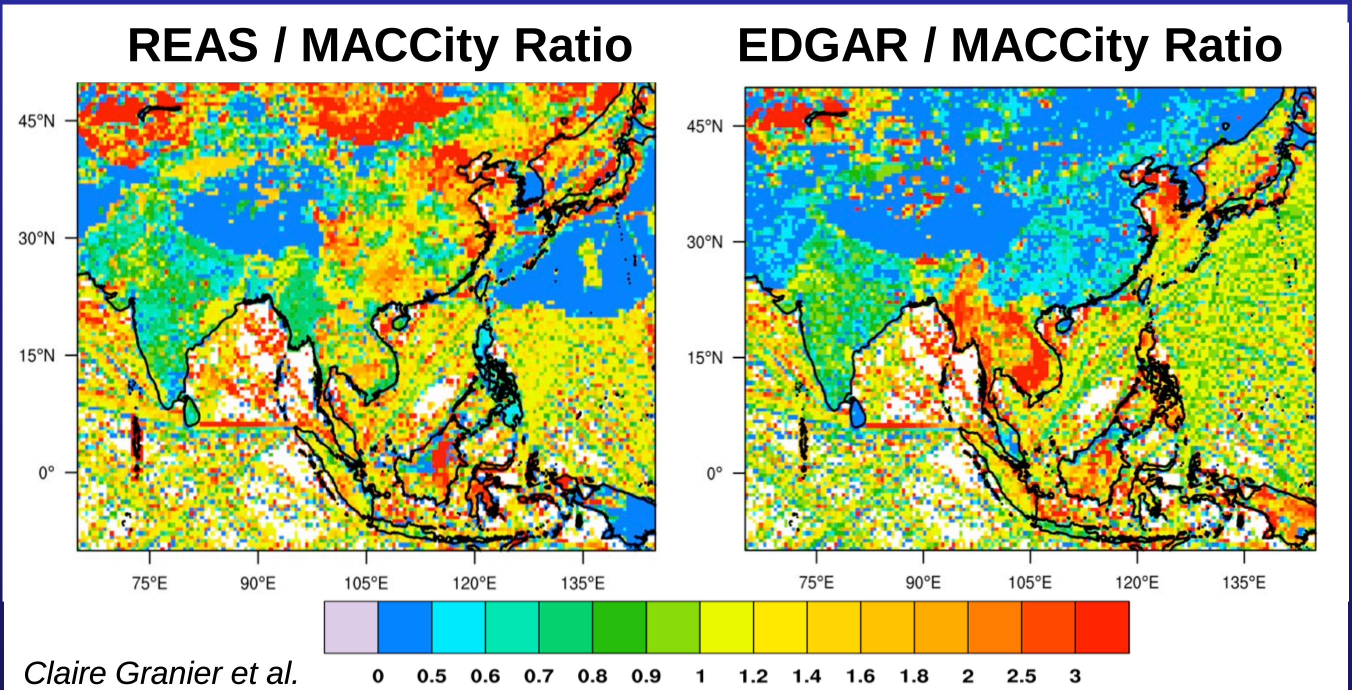
Claire Granier et al.

Spatial Difference in CO Anthropogenic Emissions - 2008



Very large differences in spatial distributions

→ More work needed to define the best spatial resolution, for example using global/regional models.



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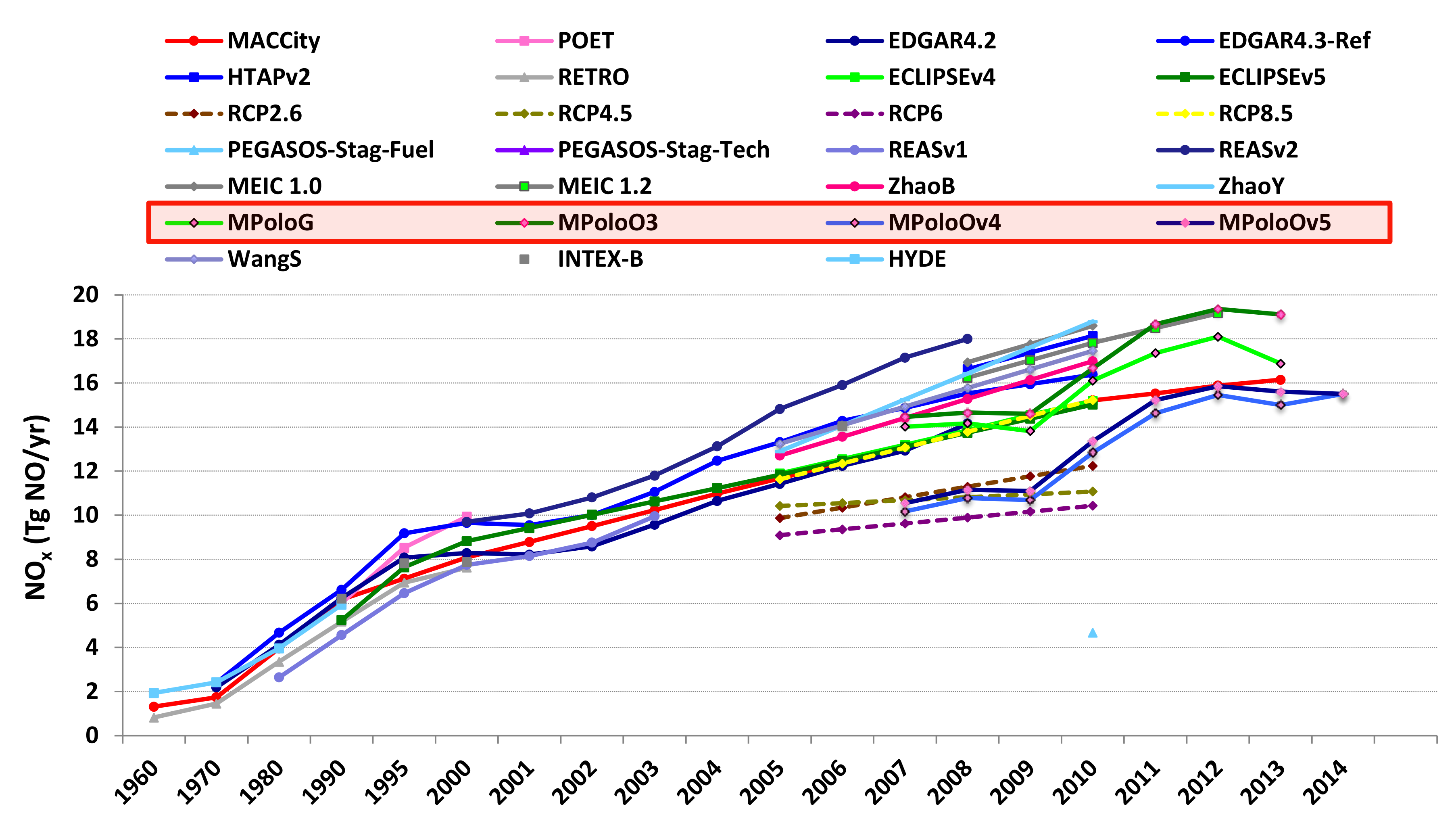
Trends in NO_x Anthropogenic Emissions

China



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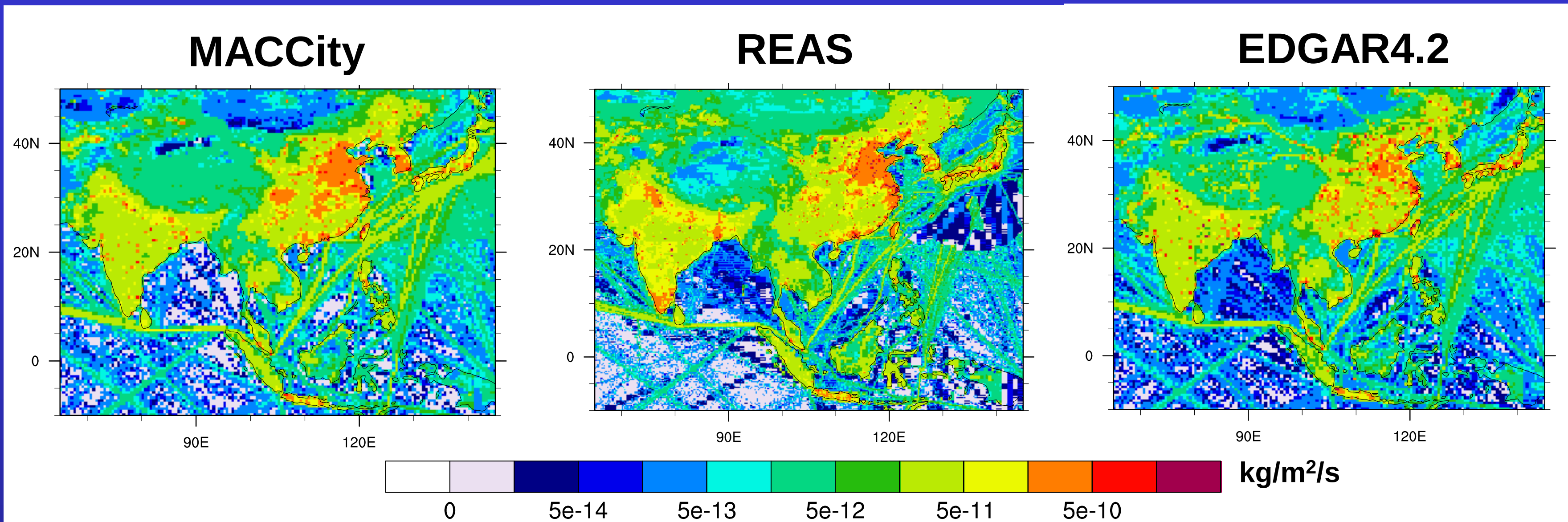
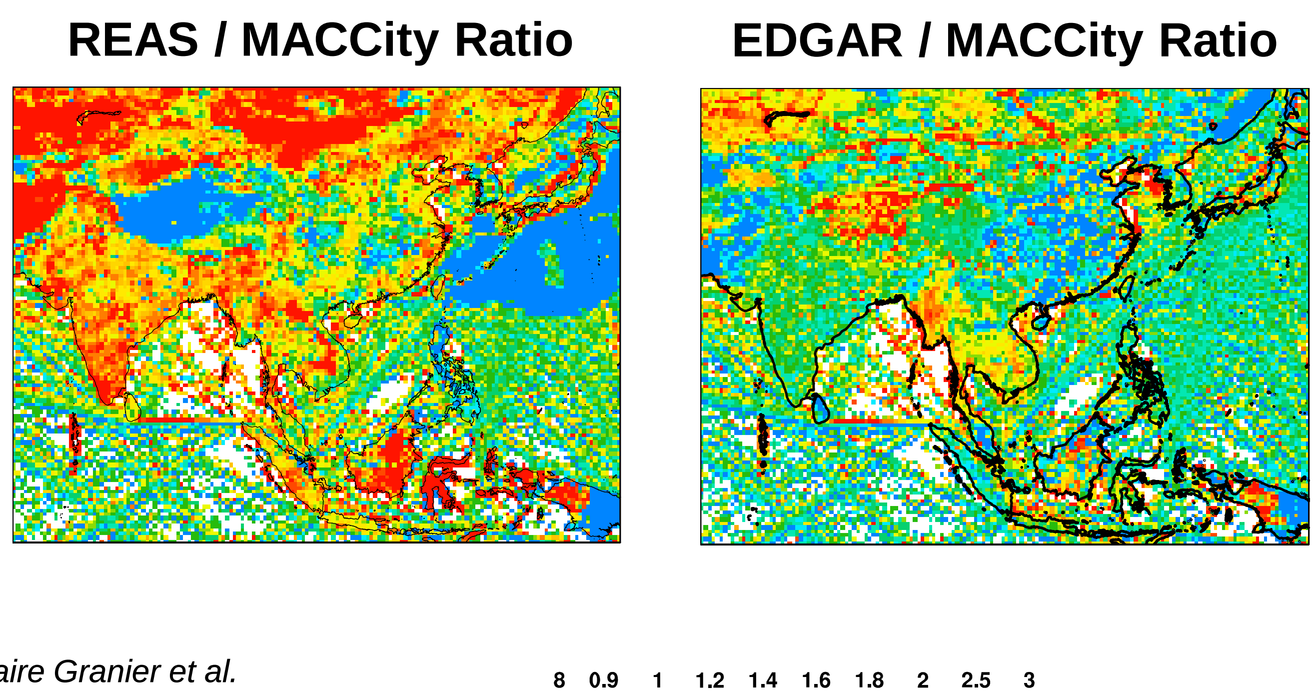
Spatial Difference in NO_x Anthropogenic Emissions - 2008



Significant differences in India and South East Asia

→ While MACCity is lower than REAS, the values are quite similar between EDGAR and MACCity



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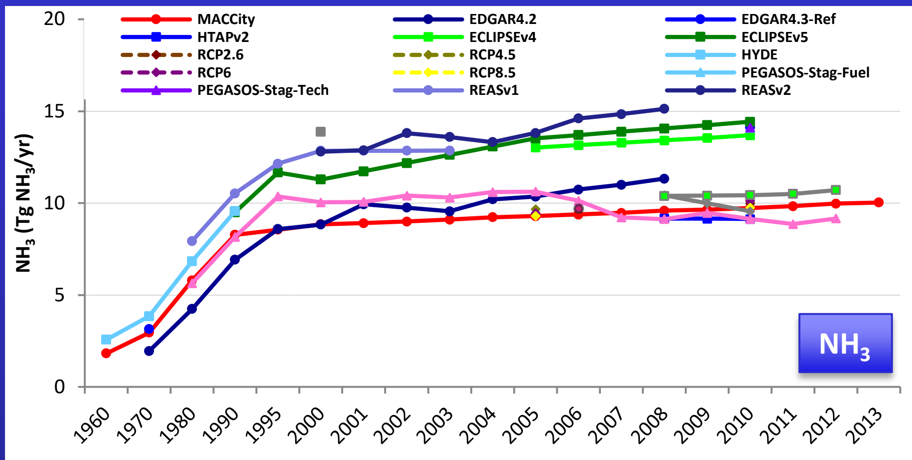
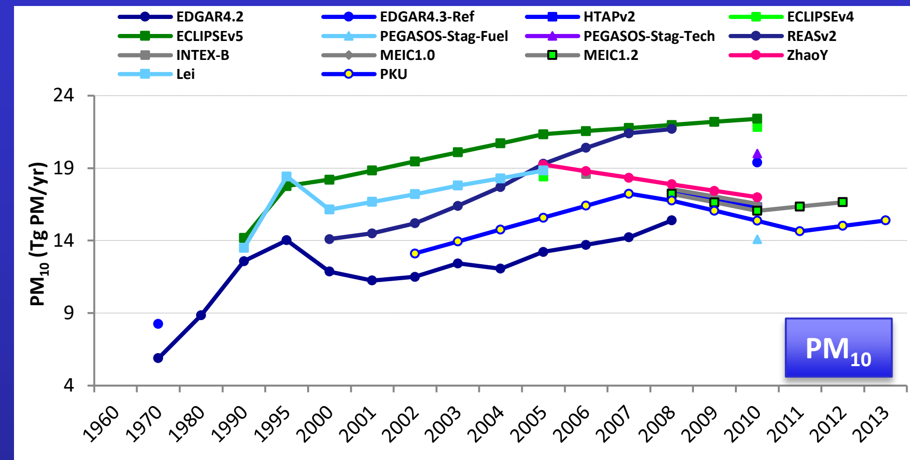
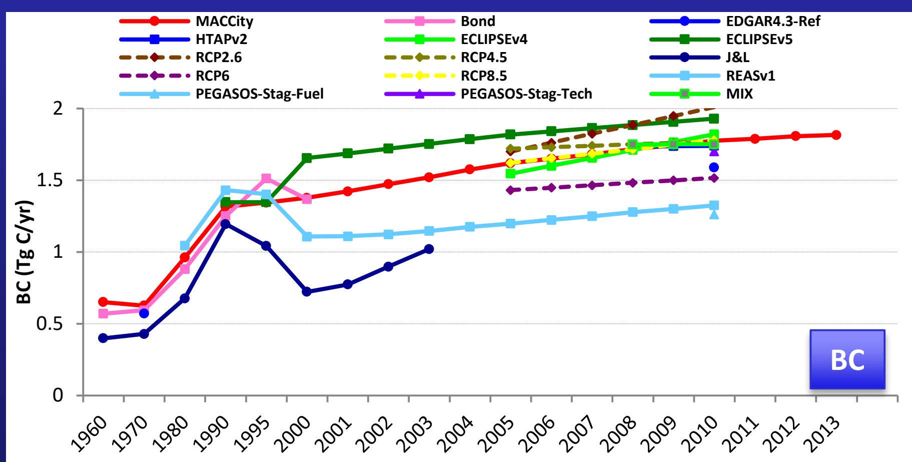
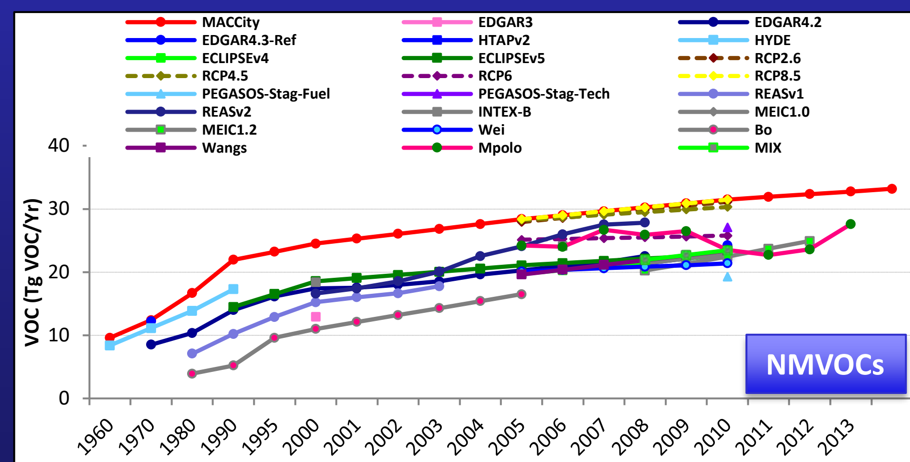
Emissions of Different species from Anthropogenic Emissions

China









- ★ Large differences between the inventories
- ★ No clear information on the trends

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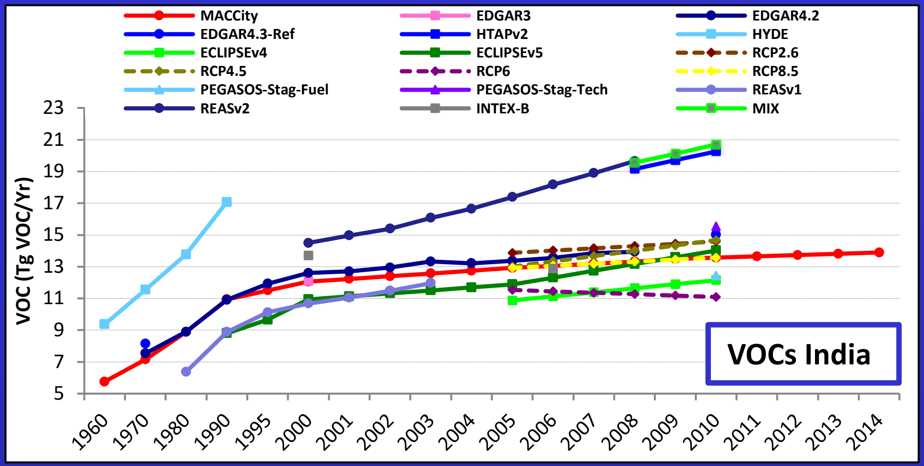
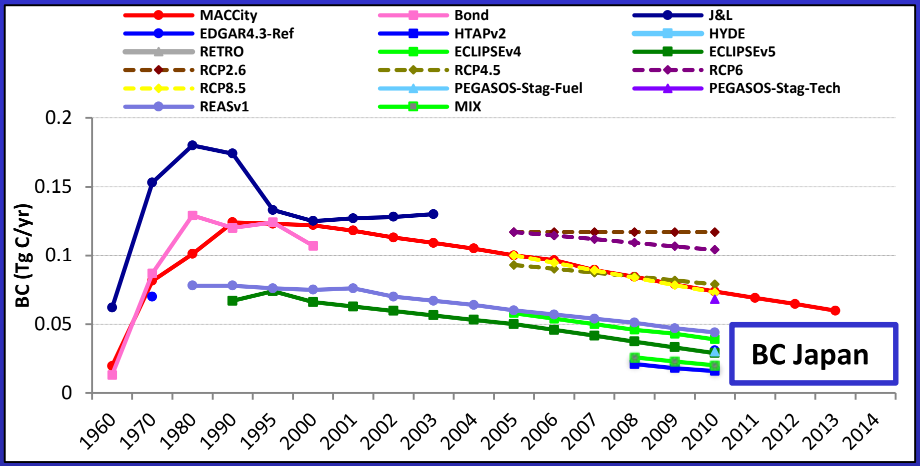
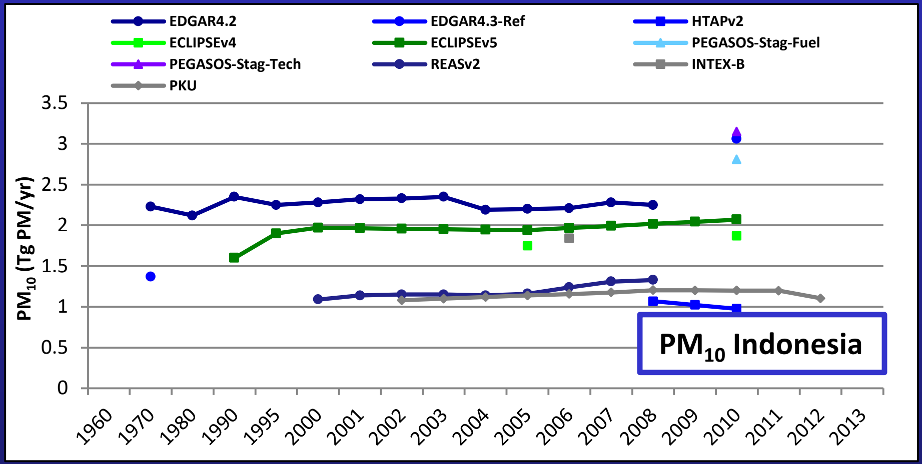
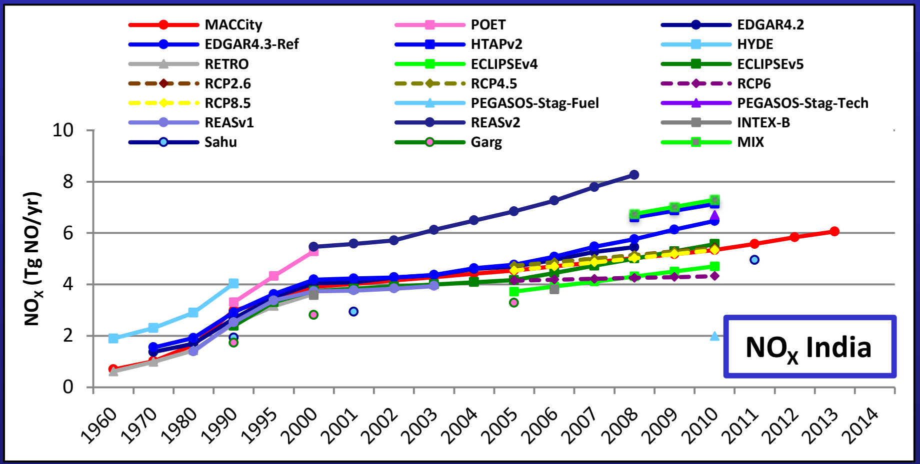
Trends in other Regions in Asia









- ★ For VOCs and NO_x in India, some inventories show large increases while other indicate almost constant emissions
- ★ For Japan, there is consistency in the decrease in BC emissions

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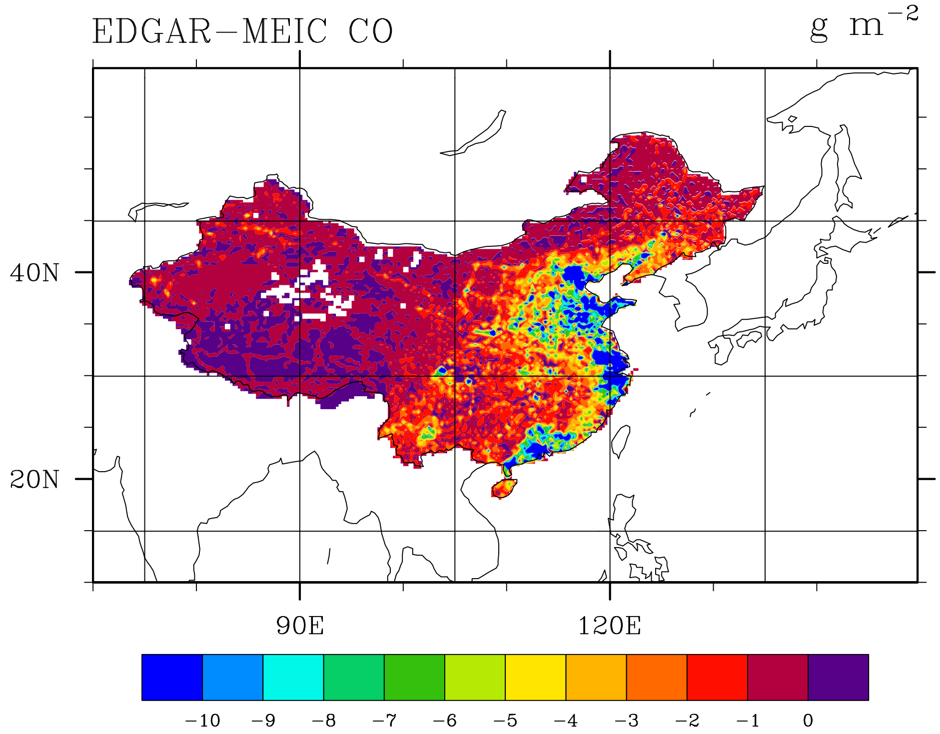
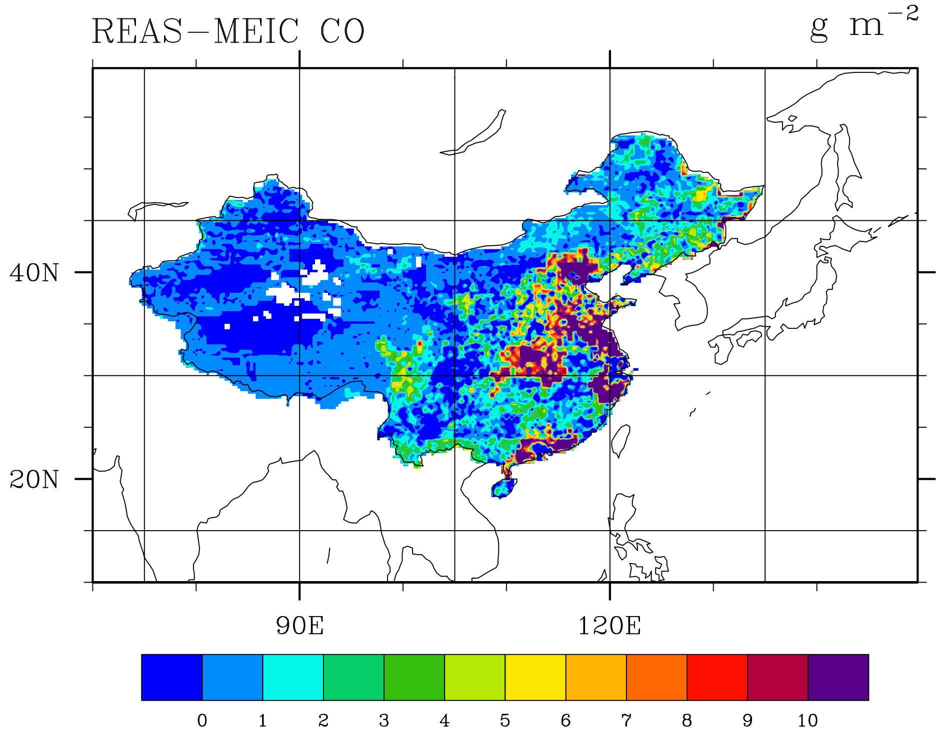
Transport sector CO emissions in 2008
(Is the definition of transport emissions the same ?)

From E. Saikawa, 2014 GEIA Conference

EDGAR - MEIC total: **-16.7 Tg**

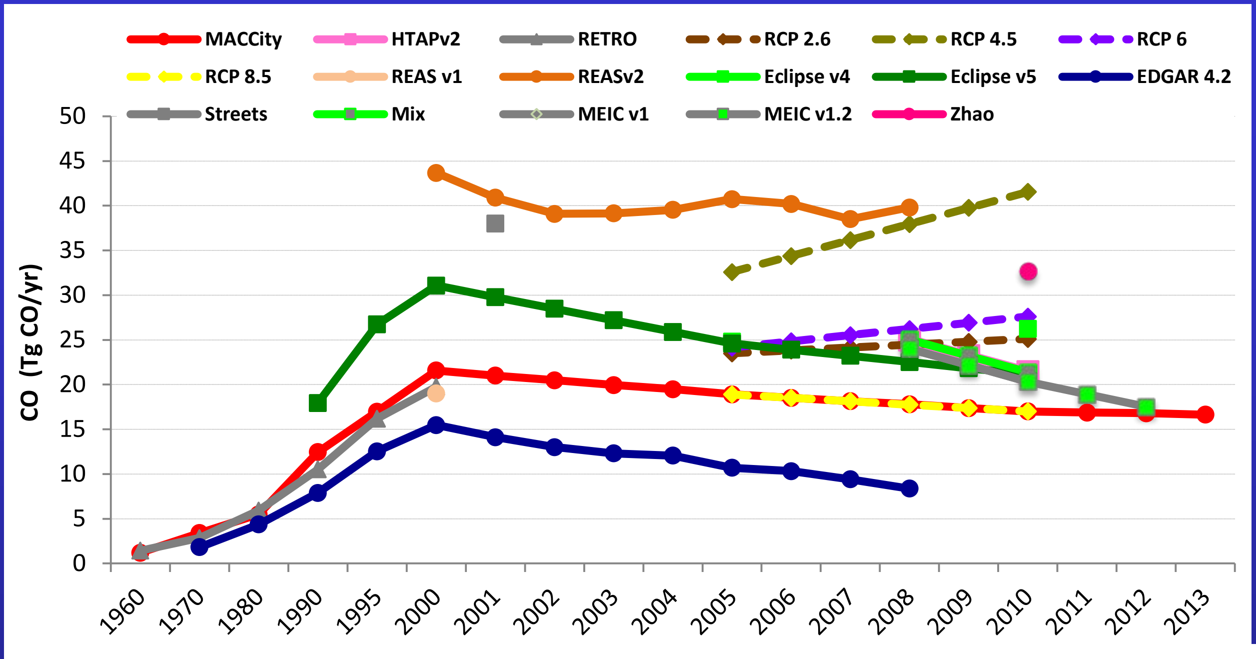


REAS - MEIC total: **20.3 Tg**



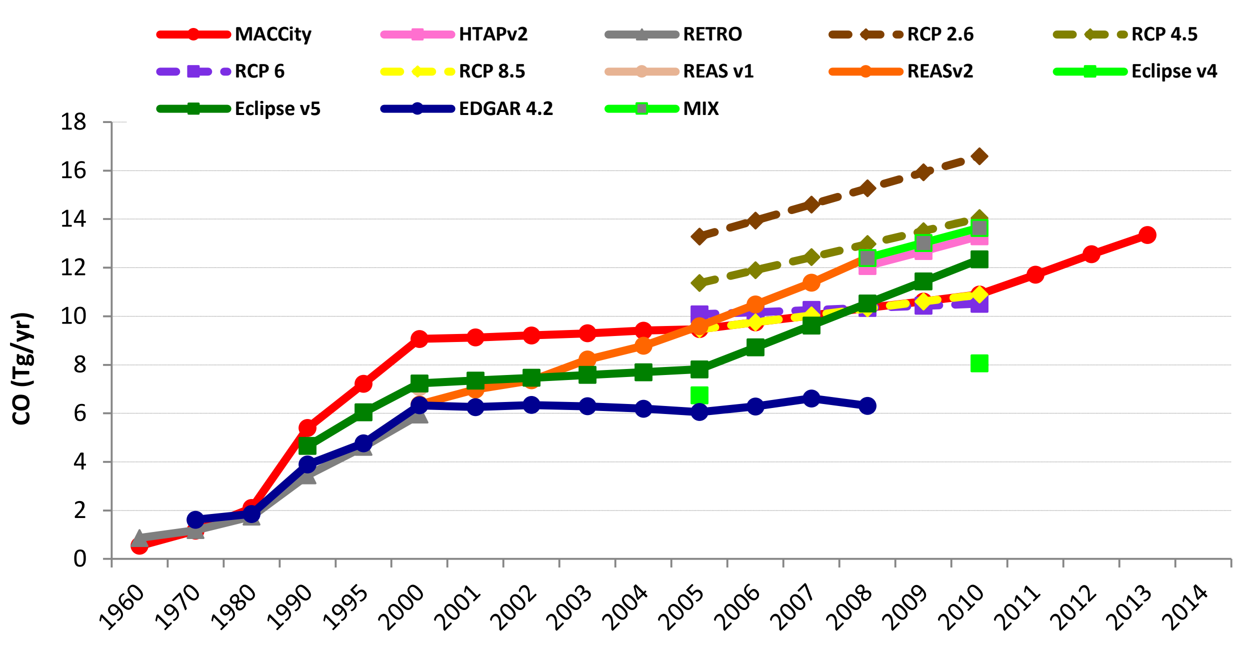
	EDGARv4.2	MEIC	REASv2
CO [Gg/year]	7,338	24,036	44,299

Trends in CO Emissions from Transport sector



China

=> Policies implemented



India

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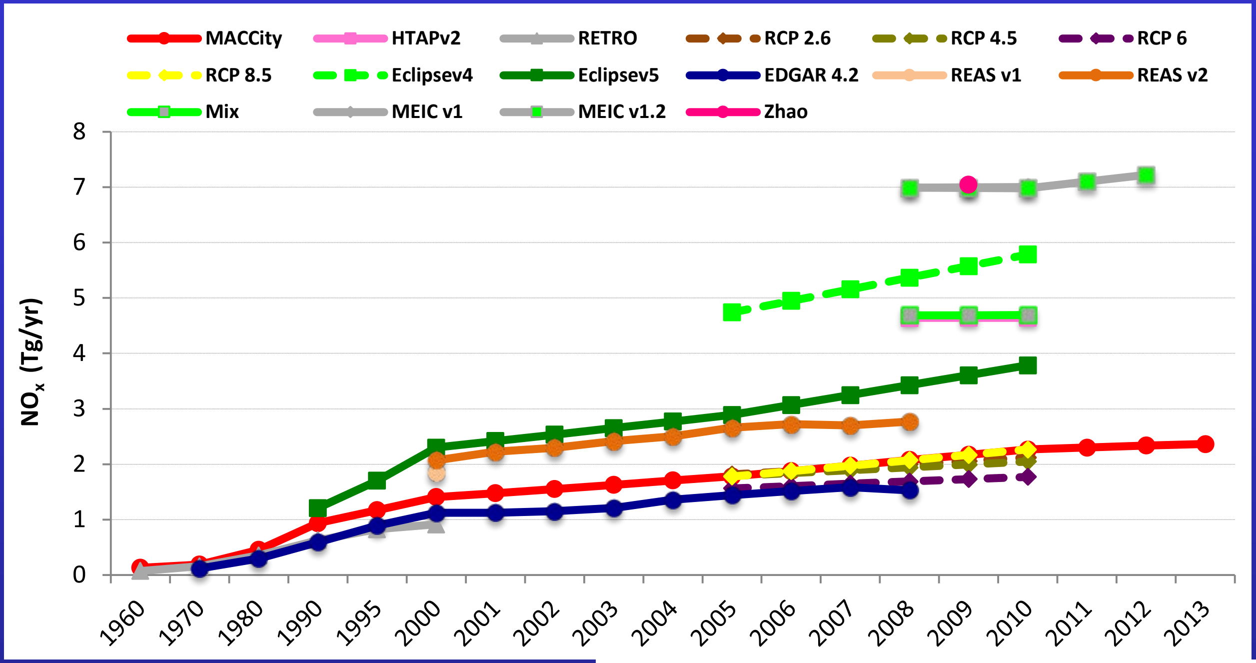
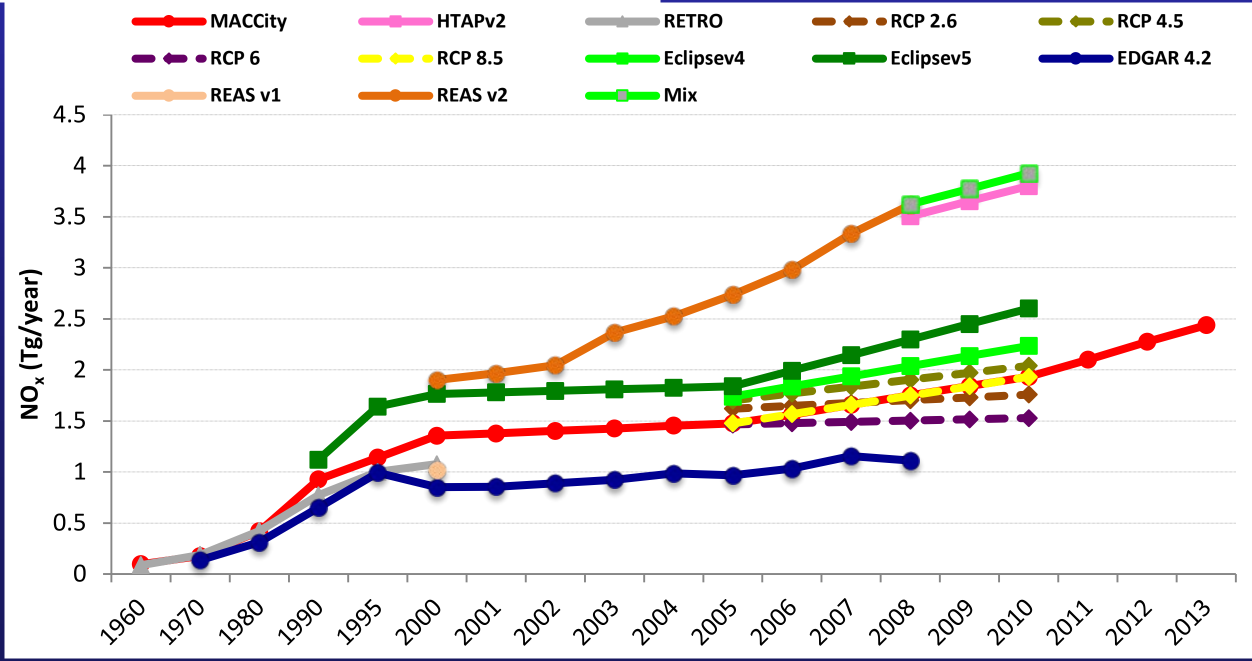
Trends in NO_x Emissions from Transport sector



China

India



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Emissions Information Challenges





Many emissions data requirements are common to air quality & climate research, regulation & policy:

- Transparency
- Uncertainty
- Timeliness
- Accuracy
- Consistency

At the same time, there are many issues and needs associated with emissions data:

Complexity

- Spatial/temporal scales
- Source types
- Interdisciplinary

Development

- Inconsistencies
- Timeliness
- Traceability

Analysis

- Evaluations
- Uncertainties
- Impacts

Communication

- Data access and sharing
- Literature access
- Producer – user feedbacks

Frost, G. J., et al. (2013) Atmos. Environ., doi: 10.1016/j.atmosenv.2013.08.063

A few of the many main issues in the determination of anthropogenic emissions

- Sector definitions
- Emission factors
- Short time scale variations
- Sporadic and rapidly evolving sources
- Gridding
- Speciation of VOCs emissions
- Speciation of PM emissions
- How to use inverse modeling

Comparisons for different sectors

→ Most global inventories use the IPCC sectors
→ Regional inventories developed by regulatory agencies use SNAP or other sectors

Sector number	Sector name
1	Energy production and distribution
2	Industry (combustion and non-combustion)
3	Land transport
4	Maritime transport
5	Aviation
6	Residential and commercial
7	Solvents
8	Agriculture
9	Agricultural waste burning on fields
10	Waste
11	Open vegetation fires in forests
12	Open vegetation fires in savanna and grasslands
13	Natural emissions

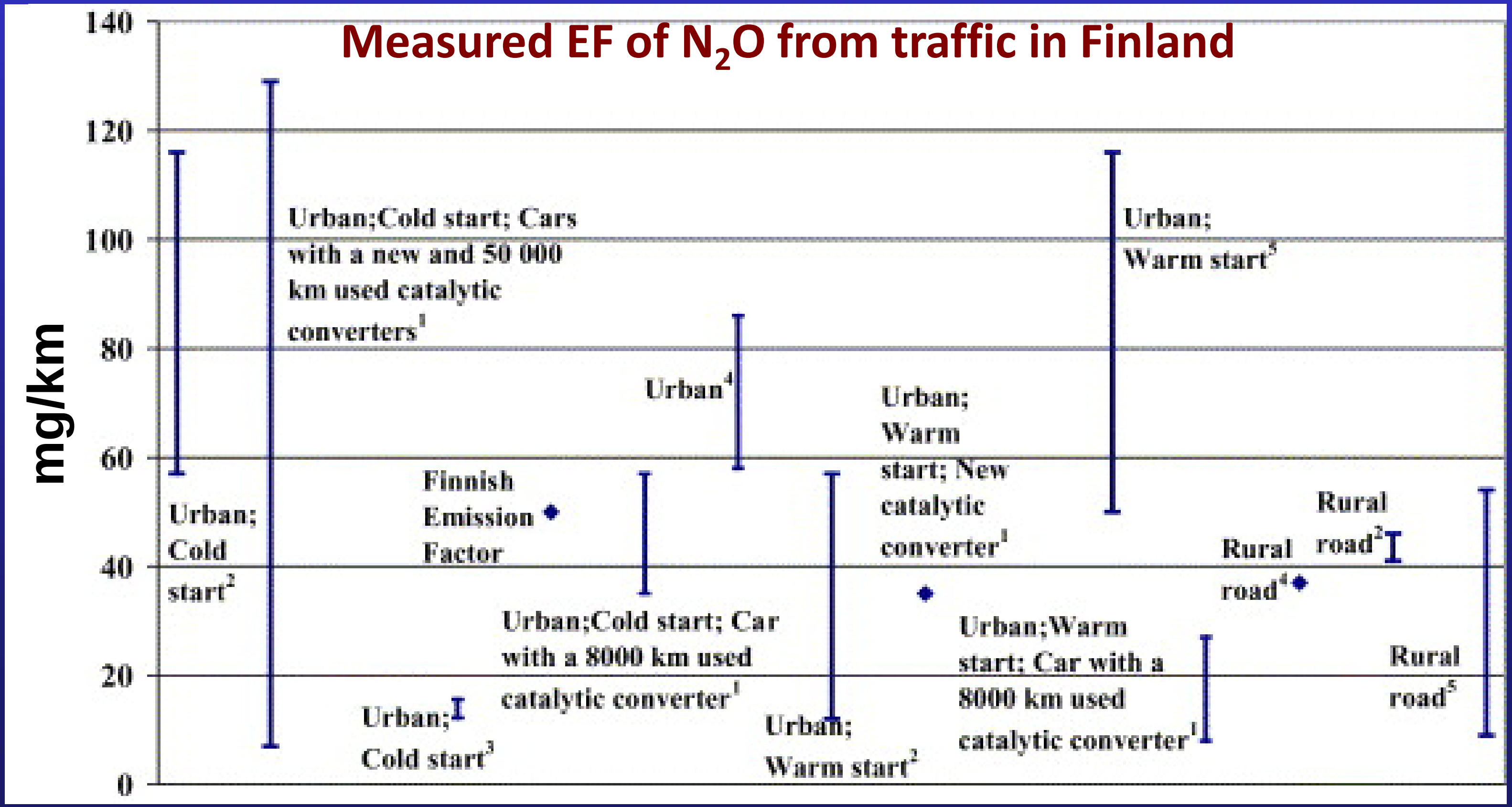
IPCC sectors used in some global inventories

SNAP sectors in the European inventories

SNAP	Description
1	Public electricity and other energy transformation
2	Small combustion plants
3	Industrial combustion and processes with contact
4	Industrial process emission
5	Fossil fuel production
6	Solvent and product use
7	Road Transport
8	Other (non-road) transport and mobile machinery
9	Waste disposal
10	Agriculture
11*	Nature

One of the main uncertainties: Emission Factors

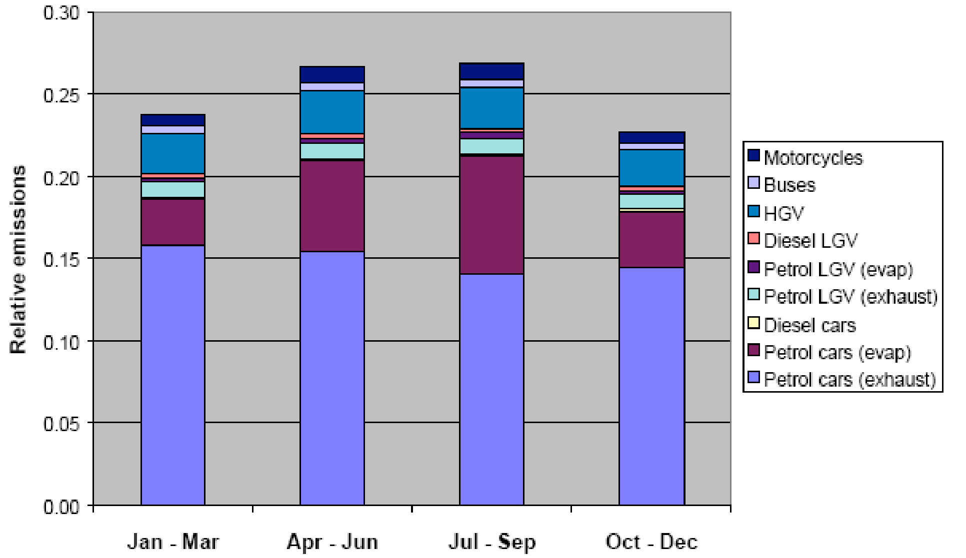
Main reasons:
- errors in definition / interpretation of definition
- errors in sampling / measurements
- errors in the scientific understanding of the processes



Monni, S. (2004) Uncertainties in the Finnish 2002 Greenhouse Gas Emission Inventory, VTT Working Papers 5

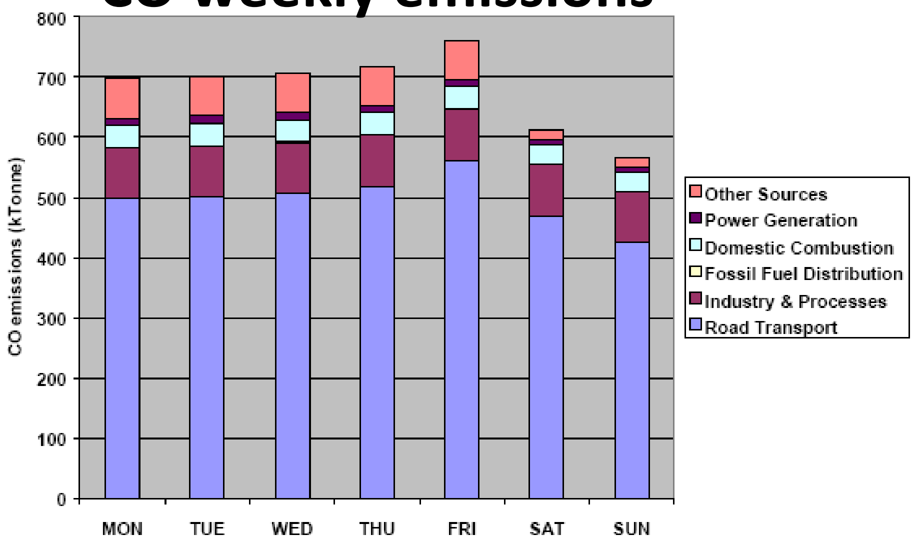
Temporal variation of fossil fuel

Seasonal VOCs

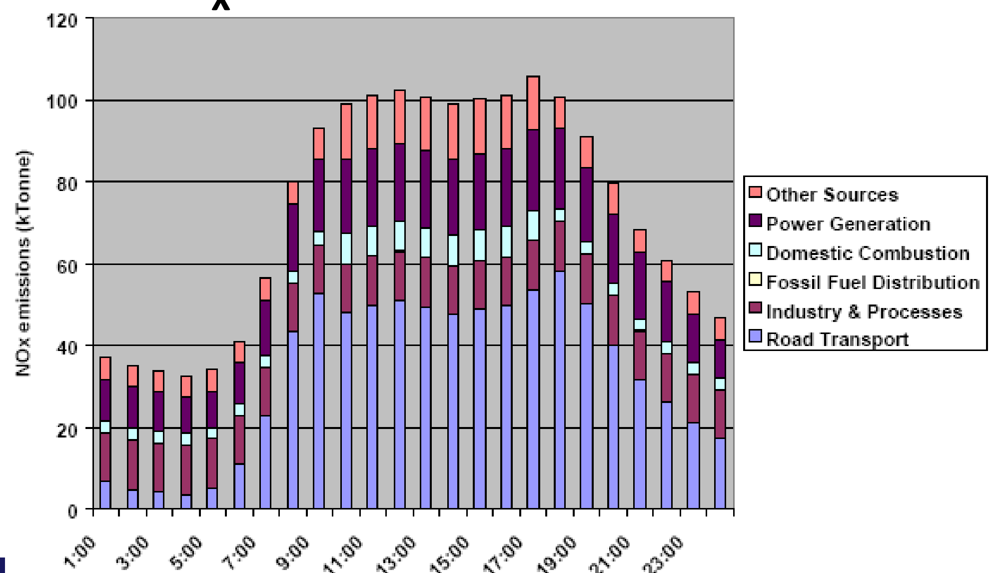


From TNO, The Netherlands:
not many details on differences between countries, different year, etc.

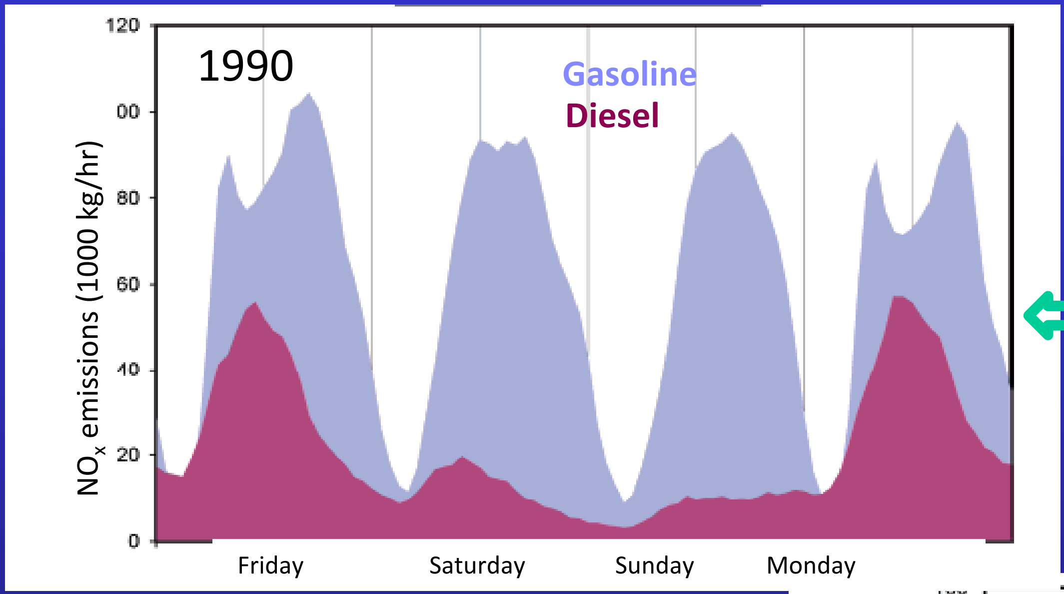
CO weekly emissions



NO_x diurnal emissions



Urban Day-of-Week Variations: Surface Observations, Fuel-Based Estimates

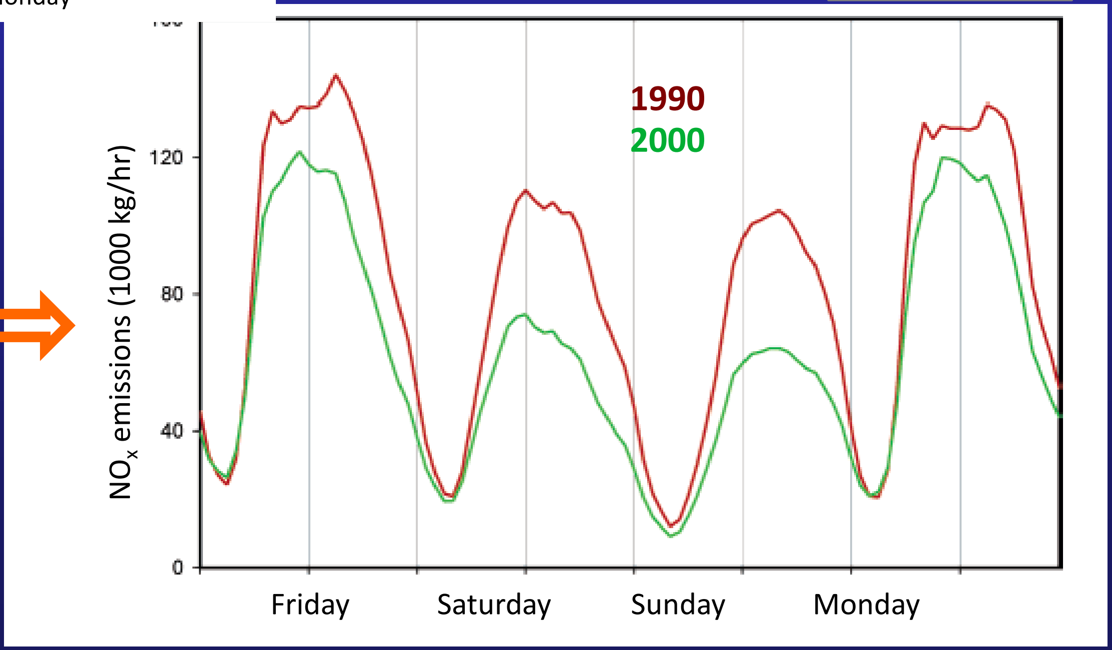


California statewide NO_x emissions from motor vehicles based on roadside monitoring, traffic counts, and fuel use

Weekend-weekday differences resulting from traffic patterns

All traffic

1990 - 2000 change in weekend-weekday difference:
cleaner gasoline vehicles



Harley, R., et al. (2005) Environ. Sci. Technol.

Top-Down vs. Bottom-Up Emissions Approaches

Top-Down (inverse modeling)
Atmospheric measurements and modeling

- Sector specific
- Time specific and speciated
- Results are model-dependent
- Generally one species
- Range of uncertainties
- Up-to-date

Relies on high quality atmospheric measurements of trace gas and measurements/modeling of the atmosphere mixing and wind characteristics

Bottom-Up (inventories)
Regulatory or research emissions inventories

- Sector specific/process specific
- Time specific and not always speciated (total VOCs)
- Regional or global scales
- Uncertainty estimates usually non-existent or low
- Rarely up-to-date

Relies on high quality activity data (routine and non routine), emissions factors, estimates of control effectiveness

Pétron, G., et al. (2012) *J. Geophys. Res.,* doi: 10.1029/2011JD016360

$E_X = \Sigma_S [EF_{X,S} \bullet A_S \bullet (1 - CE_{X,S})]$

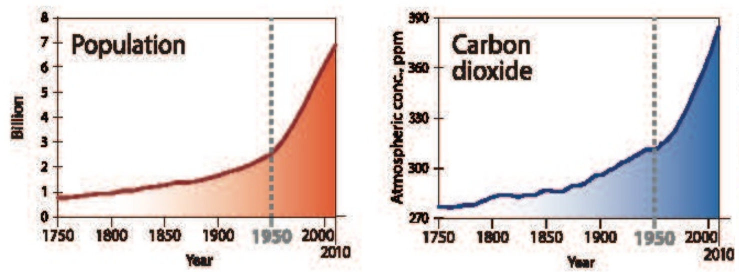
Need for Top-Down Methods for Quantifying Emissions

Actions about the atmosphere focus on emissions. Emission inventories are critical for understanding emissions in the past, present and future.

Inventories are inherently complex and therefore uncertain, and these uncertainties are difficult to quantify.

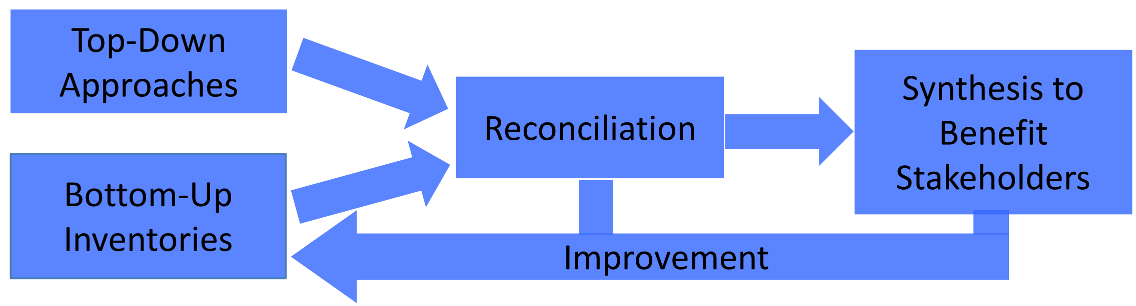
X±? ton/yr

Rapid societal development results in new challenges to emissions understanding. Similarly, efforts to circumvent controls may be difficult to detect.



W. Steffen et al., The Anthropocene Review, 2015

Top-down methods based on observations can provide objective tests of inventories and help improve the scientific basis of emissions.



We cannot manage what we can't measure – John Burrows

Why top-down emissions analyses are needed

How Engineers at West Virginia University Caught VW Cheating

By Philip E. Ross
Posted 22 Sep 2015 | 15:00 GMT

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VW diesel emissions investigation widened to include other brands

German regulators to run tests on possible 'unusual pollutants emissions' on other models including BMW, Mercedes and Ford

Twenty-three German and foreign car brands will be investigated on suspicion of further manipulation of nitrogen oxide emissions. Photograph: Robert Harding/Rex Shutterstock

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NATURE | CORRESPONDENCE

2015

Vehicle emissions: Diesel pollution long under-reported

Alastair C. Lewis, David C. Carslaw & Frank J. Kelly

Affiliations | Corresponding author

Nature **526**, 195 (08 October 2015) | doi:10.1038/526195c
Published online 07 October 2015
Correction (October, 2015)

Contents lists available at ScienceDirect

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Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

ATMOSPHERIC ENVIRONMENT

2013

New insights from comprehensive on-road measurements of NO_x, NO_2 and NH_3 from vehicle emission remote sensing in London, UK

CrossMark

David C. Carslaw [a,*], Glyn Rhys-Tyler [b,1]

[a] King's College London, Environmental Research Group, Franklin Wilkins Building, 150 Stamford Street, London SE1 9NH, UK
[b] Transport Operations Research Group, School of Civil Engineering and Geosciences, Newcastle University, Newcastle-upon-Tyne NE1 7RU, UK

Environmental Science & Technology

Article

pubs.acs.org/est

2015

Reactive Nitrogen Species Emission Trends in Three Light-/Medium-Duty United States Fleets

Gary A. Bishop* and Donald H. Stedman

Department of Chemistry and Biochemistry, University of Denver, Denver, Colorado 80208, United States

Supporting Information

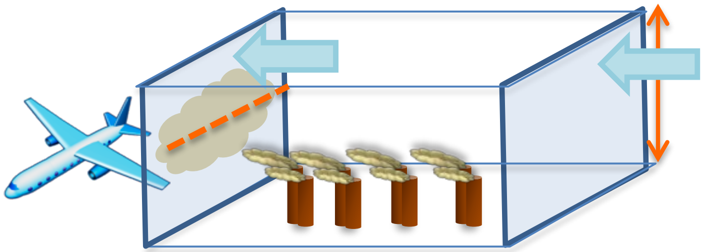
How top-down emissions analyses are carried out

Top-down emissions approaches

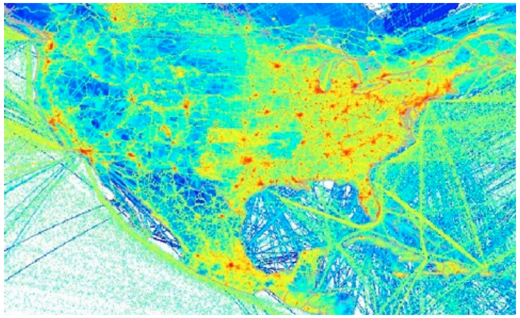
- Observations of atmospheric abundance
- Models: abundance ⇔ emissions
- Reconciliation with bottom-up inventories
- Quantifiable uncertainties
- Different methods → same answer
- Pollutants relevant to air quality & climate

What can be determined

Total emissions



Spatial mapping



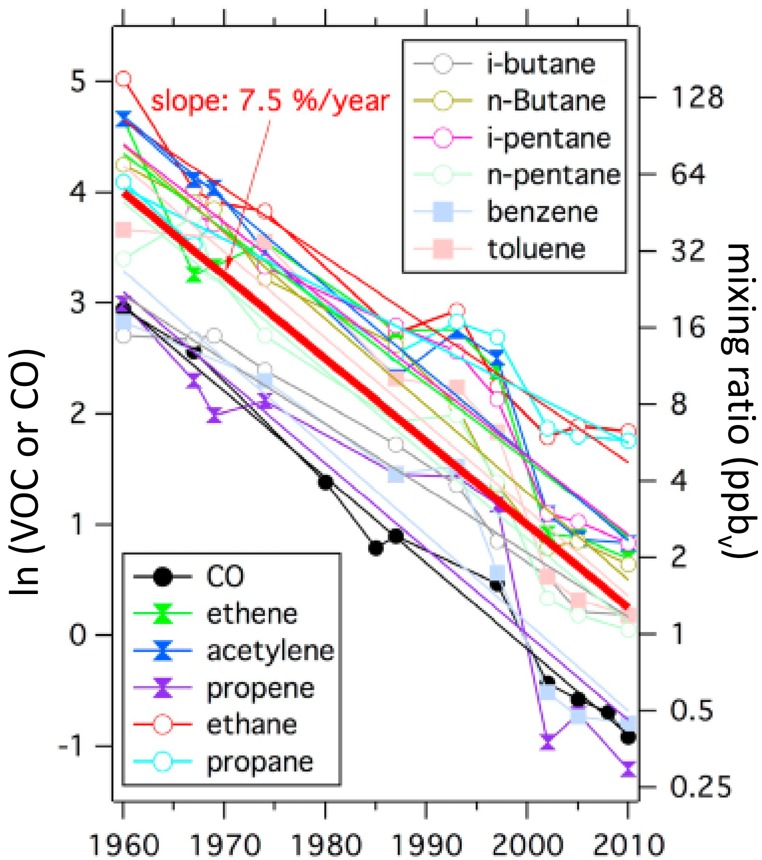
Temporal variation

Sector partitioning

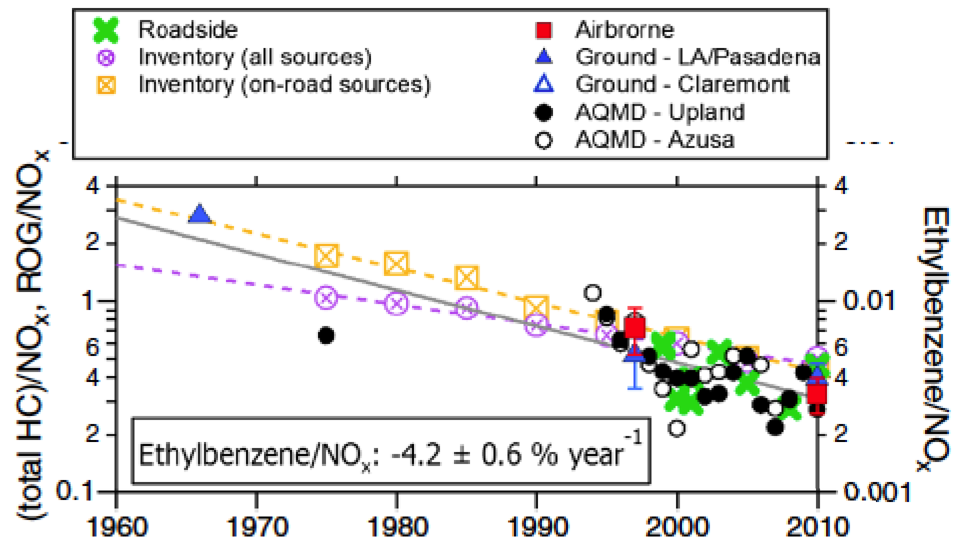
Atmospheric observations reflect emissions changes

Observations from Monitors & Field Campaigns

South Coast Air Basin (Los Angeles)



Warneke, C., et al. (2012) J. Geophys. Res., doi:10.1029/2012JD017899

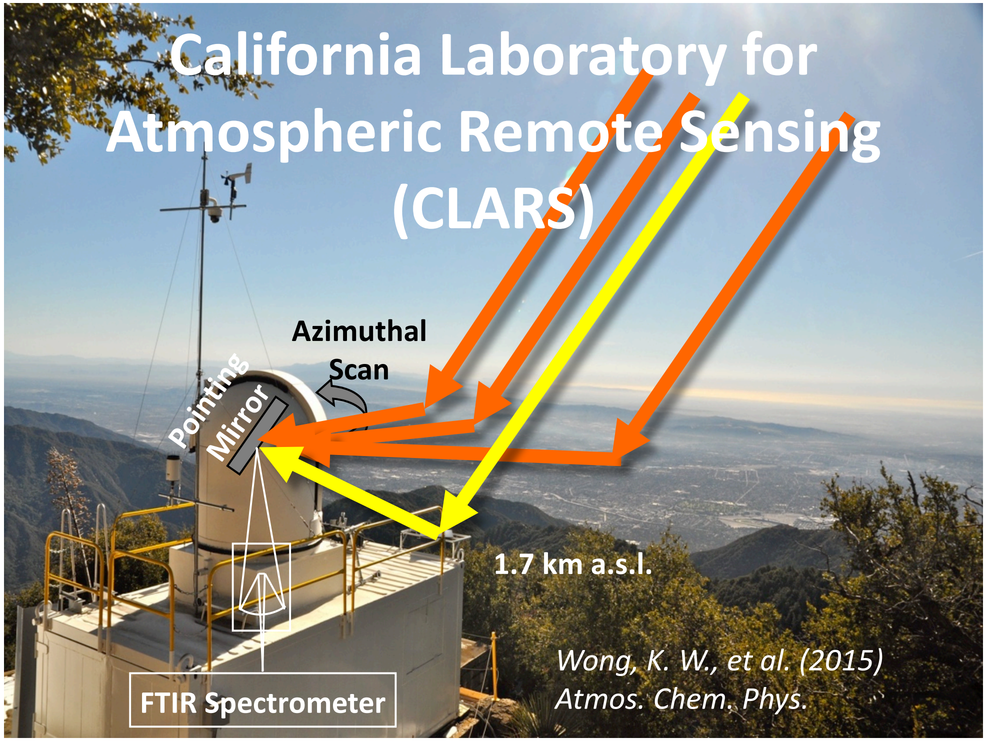


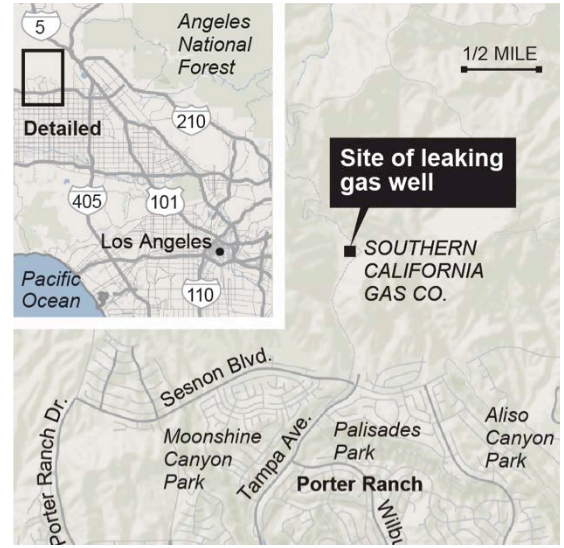
Pollack, I. B., et al. (2013) J. Geophys. Res., doi: 10.1002/jgrd.50472

Surface and airborne observations show consistency in interannual trends

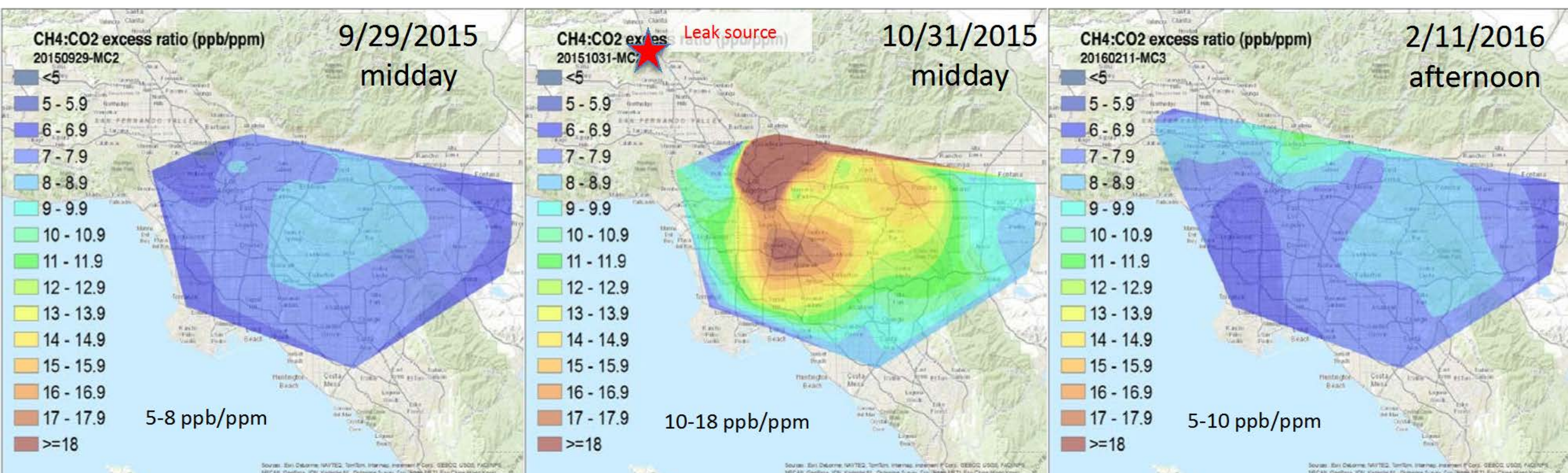
- ➢ Ambient NOx & VOCs decreasing for 50 yrs
- ➢ Declining motor vehicle emissions

Observing urban natural gas leaks

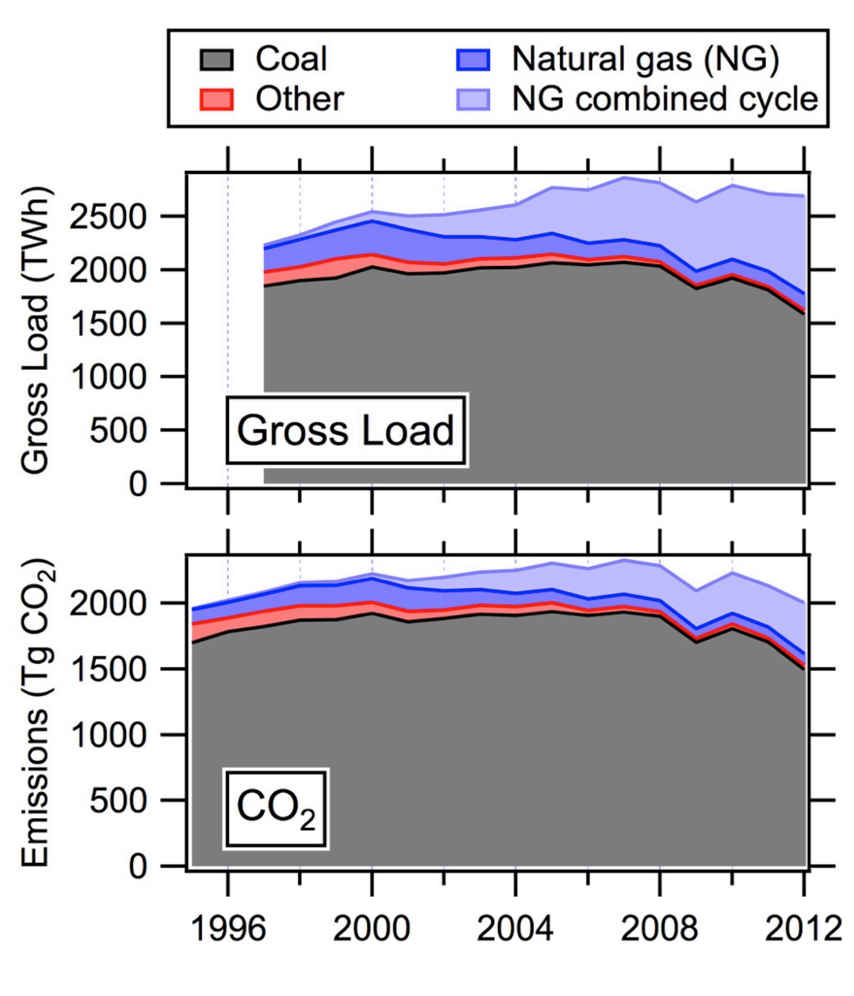




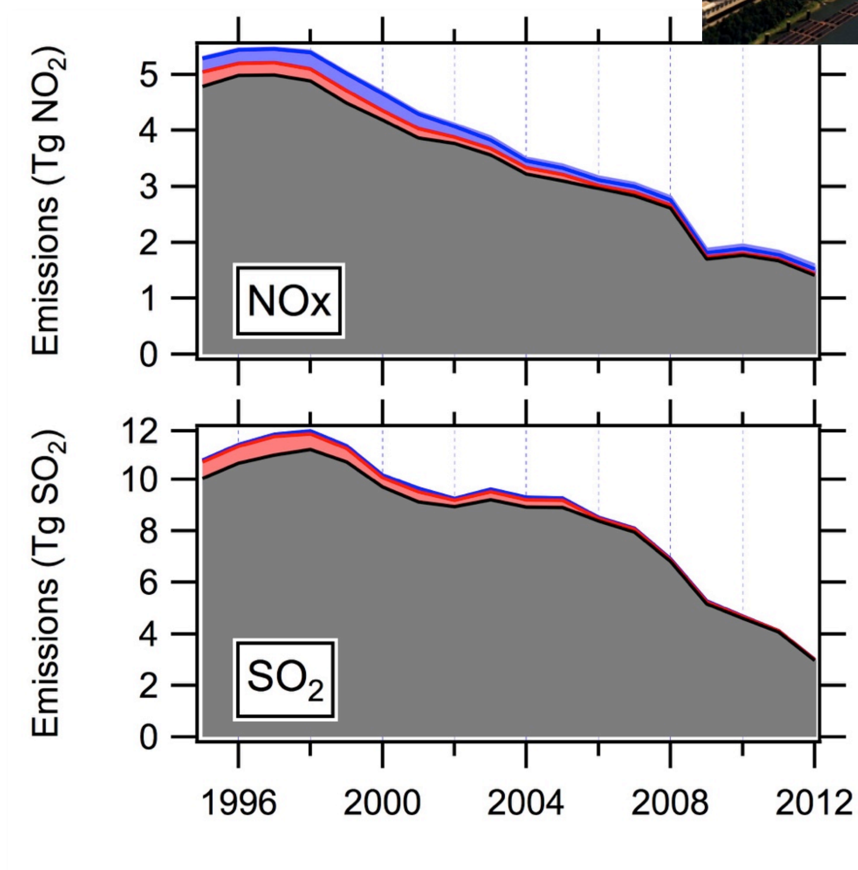
Prior to leak | Leak in progress | Leak terminated



US power plant emissions are monitored



http://ampd.epa.gov/ampd/

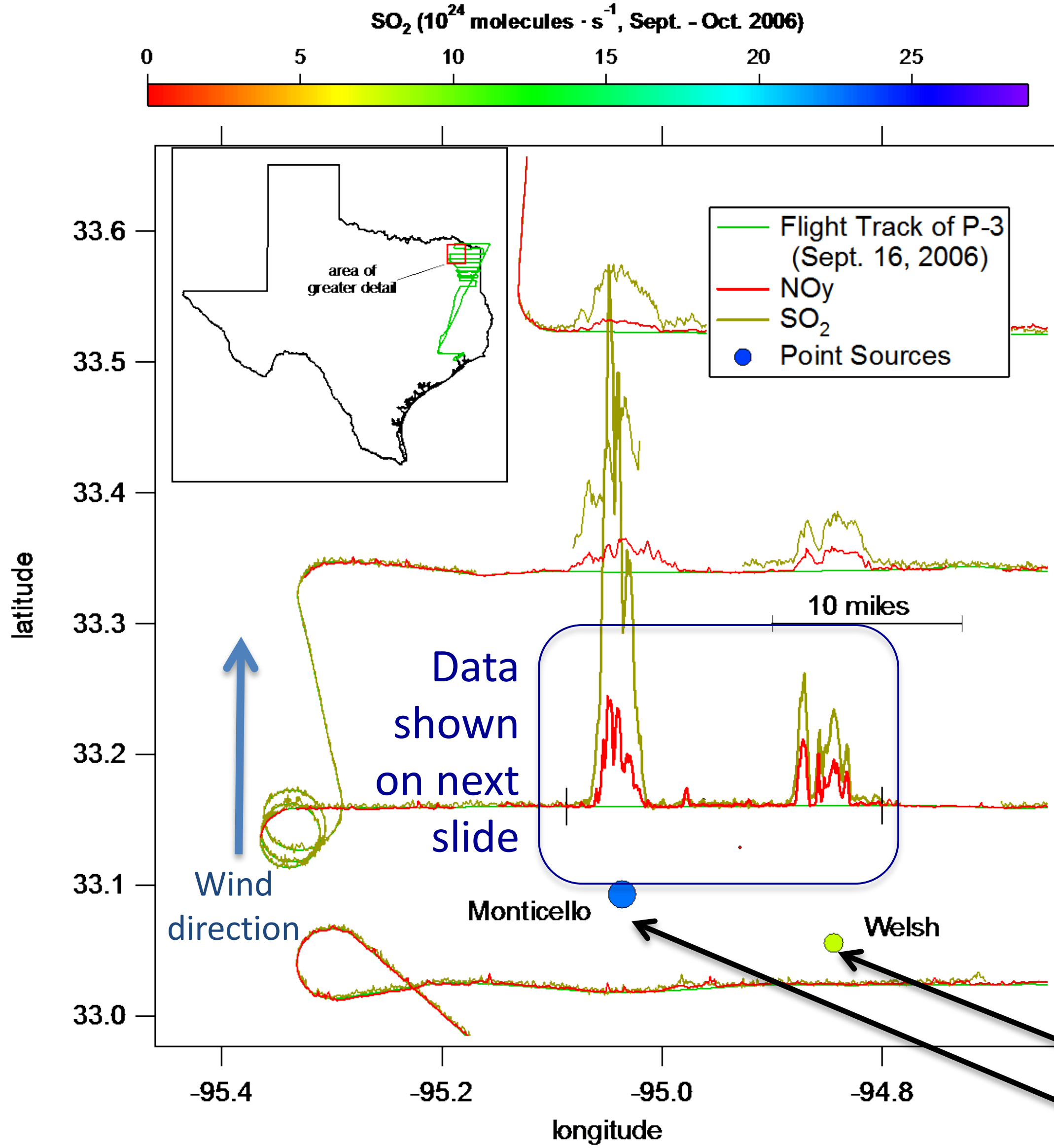


- Gross load had maximum in 2007; coal is being replaced by natural gas
- CO_2 decreased since 2007: increased use of natural gas
- NOx and SO_2 decreased: increased regulation and natural gas use

de Gouw, J., et al. (2014) Earth's Future, doi: 10.1002/2013EF000196

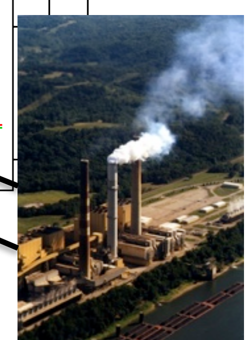
Power plant emissions from aircraft data

NOAA P-3 flight track & chemical data collected on 9/16/2006



Peischl, J., et al. (2010) J. Geophys. Res., doi: 10.1029/2009JD013527

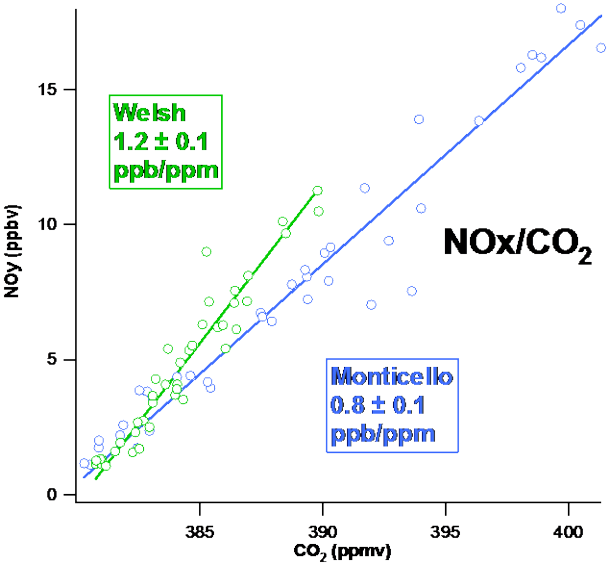
2006 Texas Air Quality Study

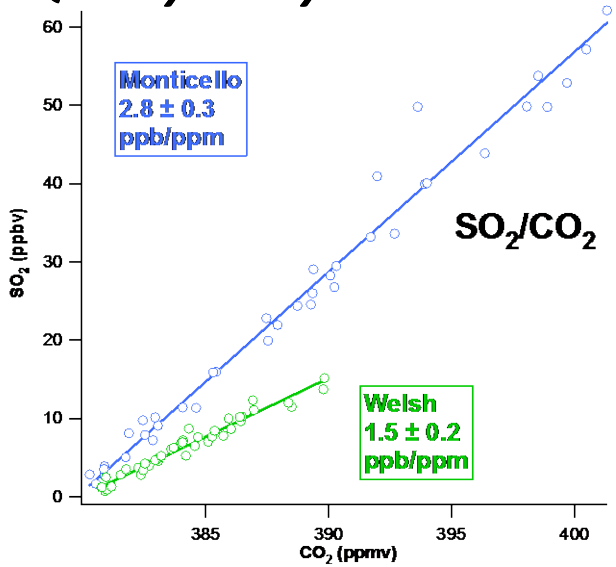


- Measured multiple chemical species on flights downwind of power plants
- Flights during daytime, well-mixed conditions

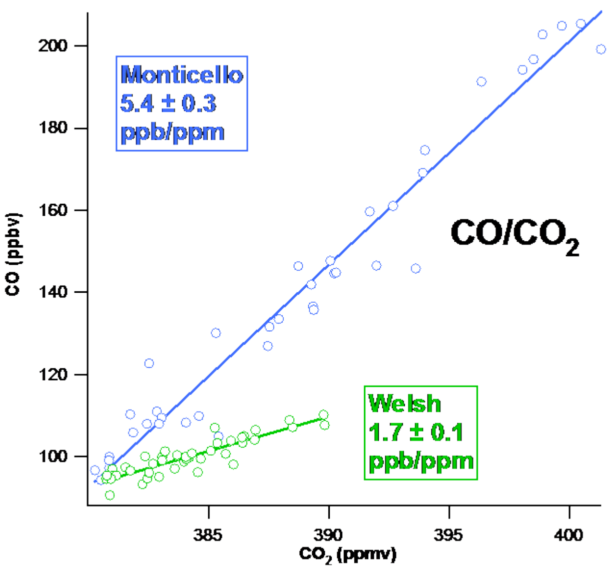
Power plant emissions from aircraft data

2006 Texas Air Quality Study





Slope of enhancement ratio correlations = Emissions ratio in molecules of species X per molecule CO_2



emission ratio		NOx/CO_2	SO_2/CO_2	CO/CO_2
Monticello	P-3	0.8 ± 0.1	2.8 ± 0.3	5.4 ± 0.3
	CEMS*	0.8	2.8	
Welsh	P-3	1.2 ± 0.1	1.5	1.7
	CEMS*	1.0	1.7	

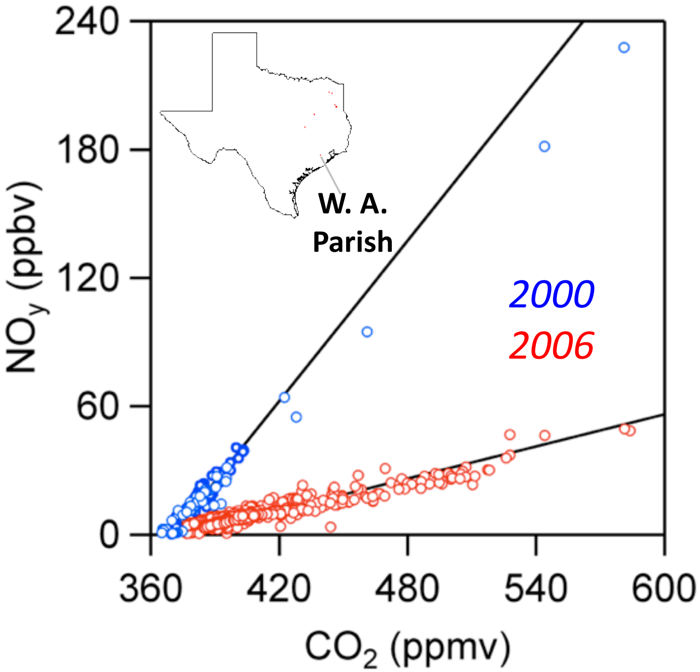
**CEMS = Continuous Emissions Monitoring Systems*

Peischl, J., et al. (2010) J. Geophys. Res., doi: 10.1029/2009JD013527

Power plant emissions from aircraft data

2000 and 2006 Texas Air Quality Studies

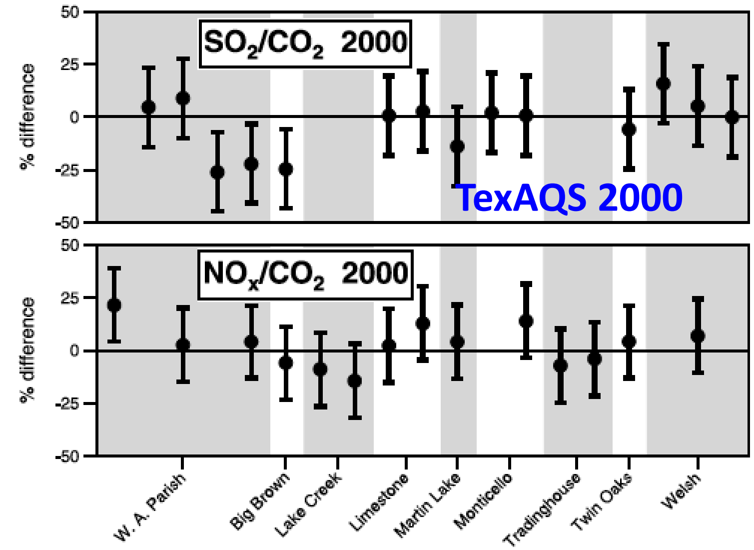
Comparing power plant emissions before & after NOx controls

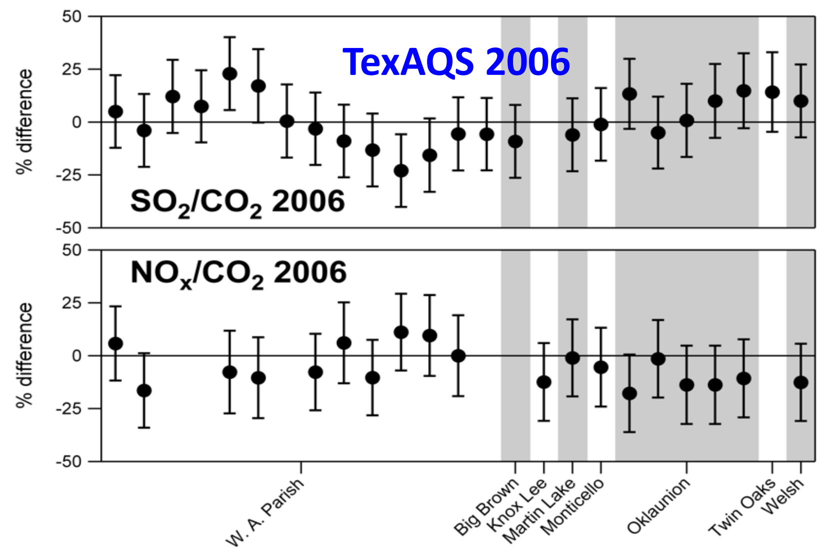


Measured reduction in NOx emissions resulting from controls

CEMS and aircraft-derived emissions agreed to within ±25%

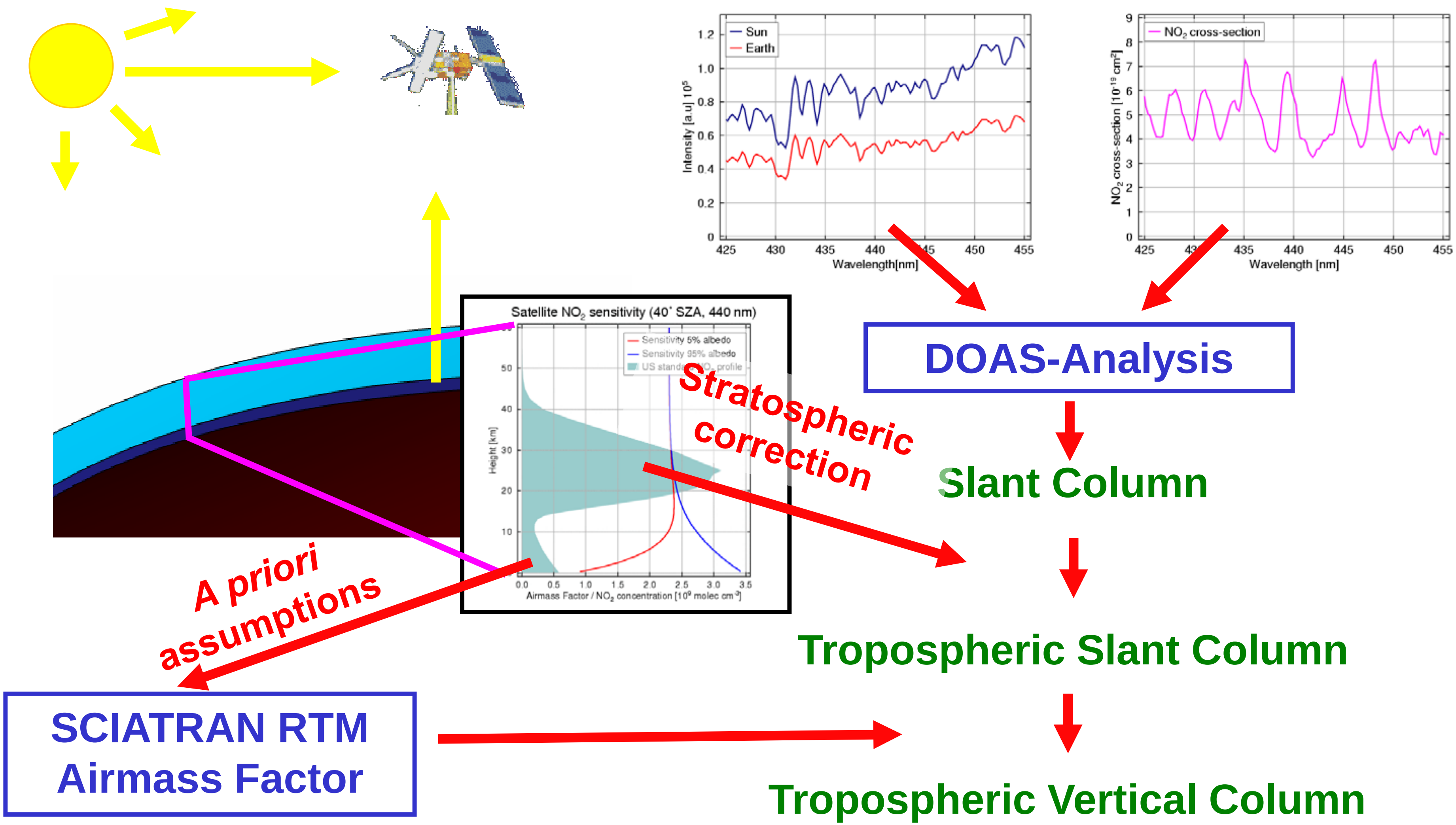
Power plant emissions differences = CEMS – aircraft



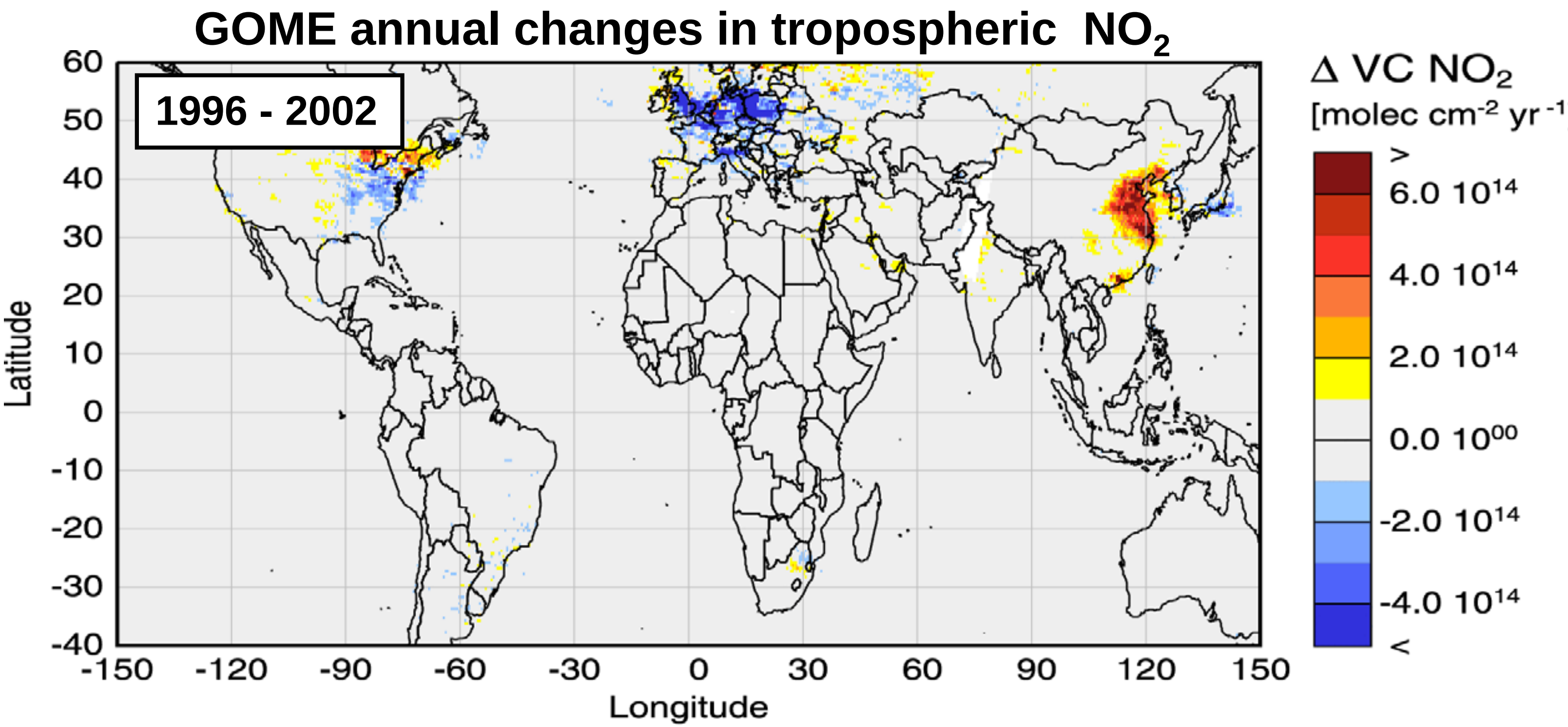


Peischl, J., et al. (2010) J. Geophys. Res., doi: 10.1029/2009JD013527

Satellite UV/VIS (NO_2) Measurements: Principle



Satellite NO_2 Trends: The Global Picture

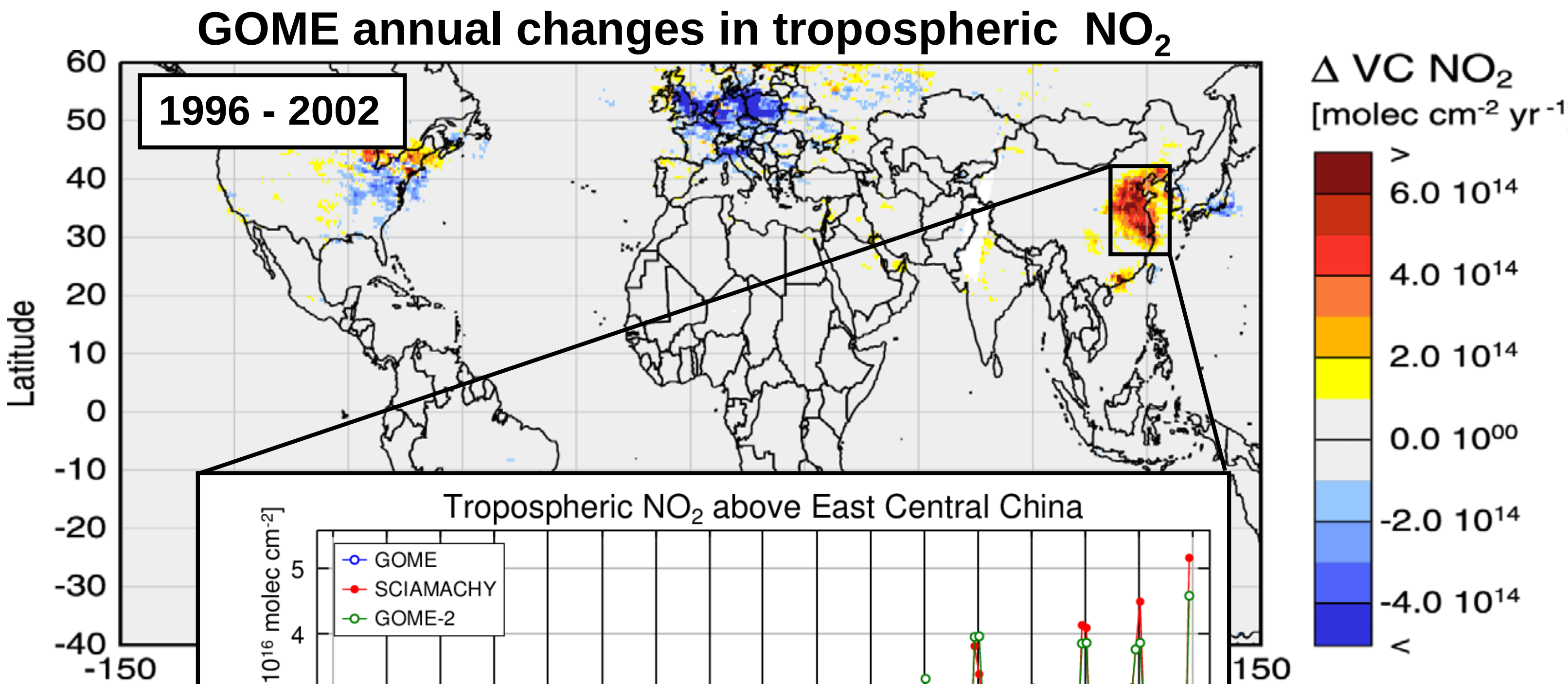


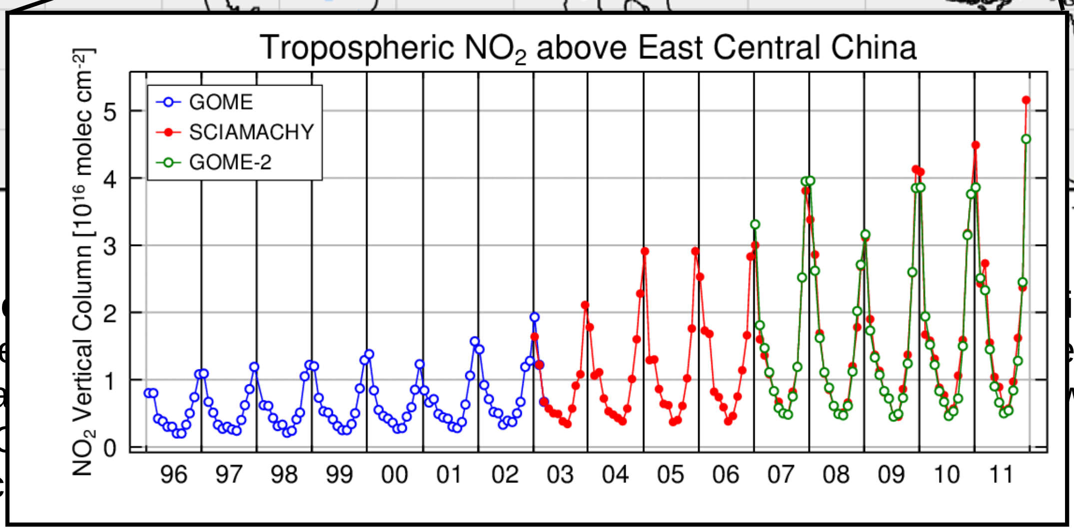
- 7 years of GOME satellite data
- DOAS retrieval + CTM-stratospheric correction
- seasonal and local AMF based on 1997 MOART-2 run
- cloud screening

- NO_2 reductions in Europe and parts of the US
- strong increase over China
- consistent with significant NO_x emission changes

A. Richter et al. (2005) Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437

Satellite NO_2 Trends: The Global Picture





- 7 years … ions in Europe and parts of the US
- DOAS r… ease over China
- seasona… with significant NO_x emission 1997 MC…
- cloud sc…

A. Richter et al. (2005) Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437

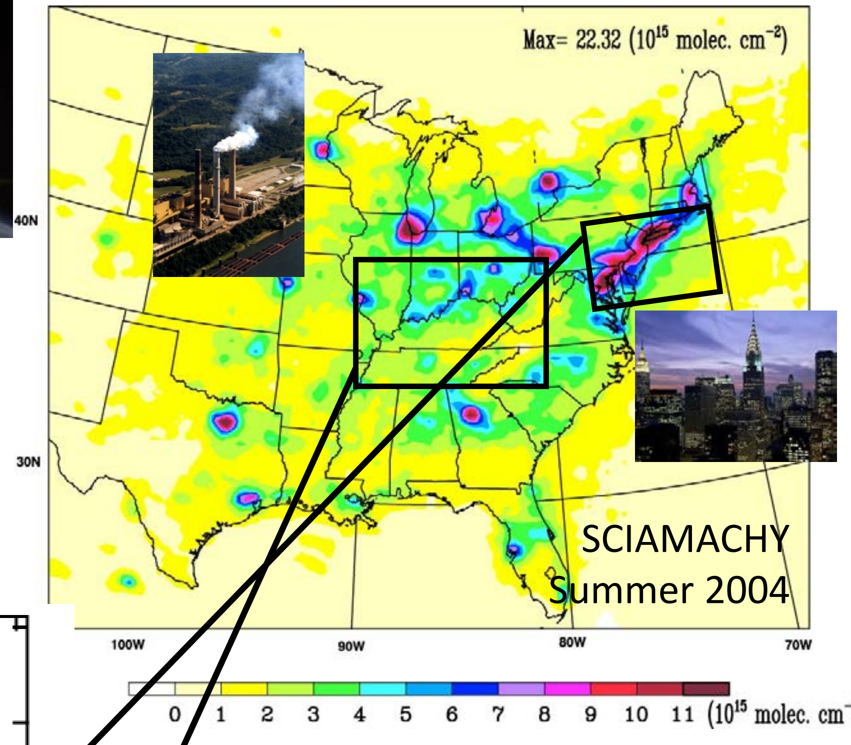
Universität Bremen
Andreas.Richter@iup.physik.uni-bremen.de

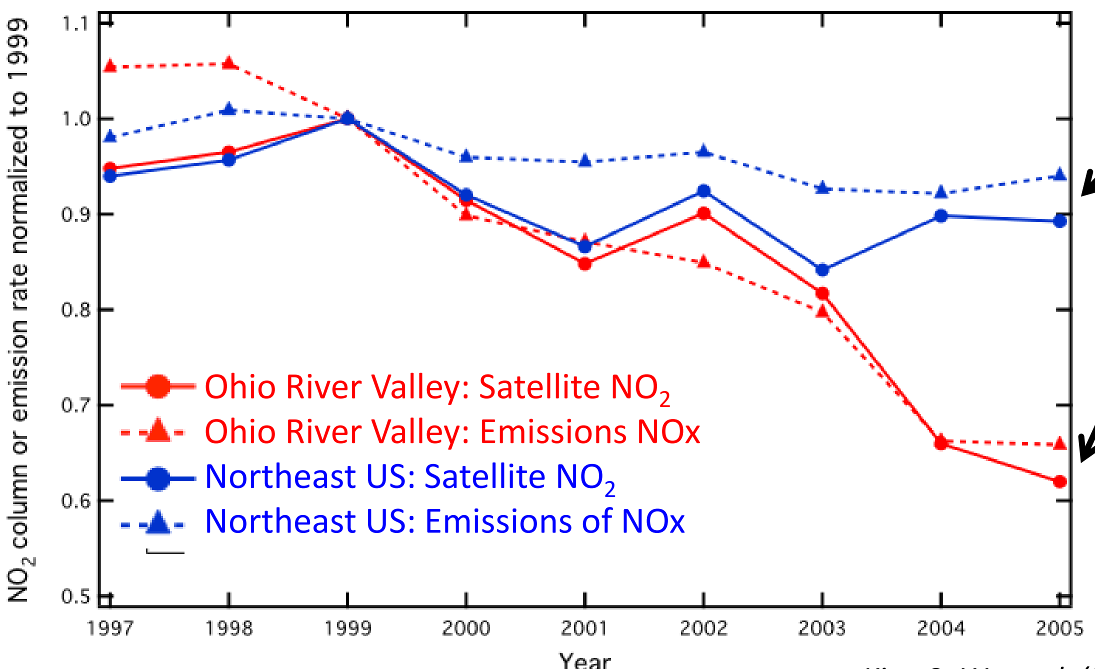
Using Satellites to Understand US Emissions Trends

Satellites measure solar radiance reflected from Earth

After accounting for atmospheric absorption and scattering, amount of NO_2 in troposphere can be derived

Satellite NO_2 signals are proportional to NOx emissions from surface sources



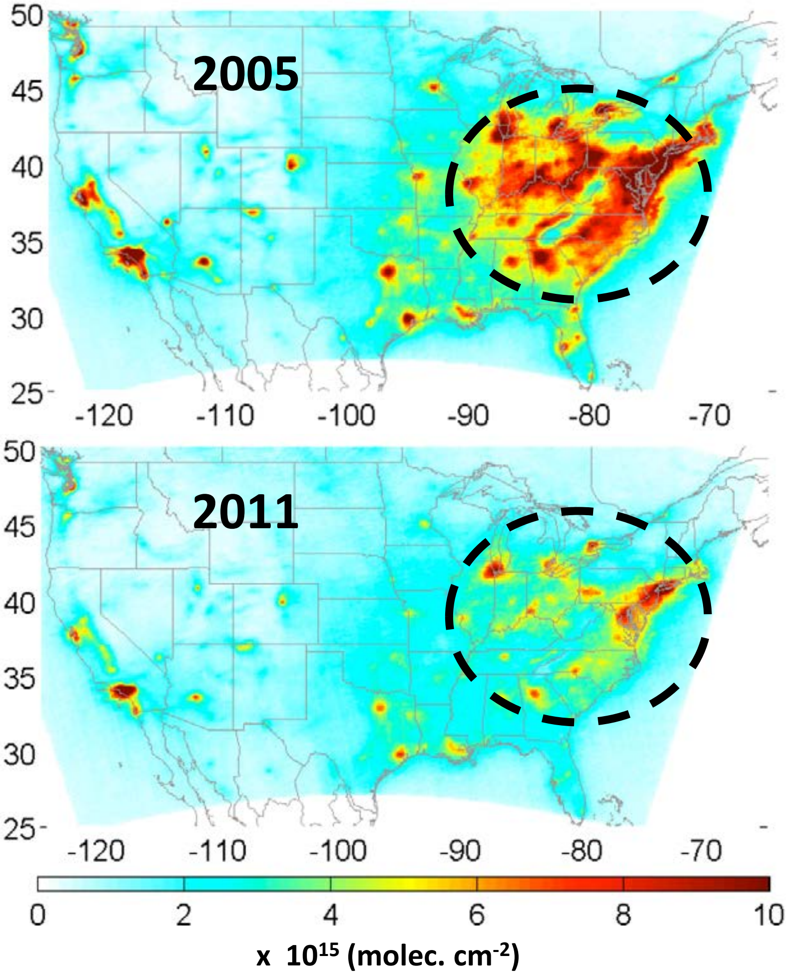


Trends in satellite NO_2 signals show impact of power plant NOx emission controls

Kim, S.-W., et al. (2006) Geophys. Res. Lett., doi: 10.1029/2006GL027749

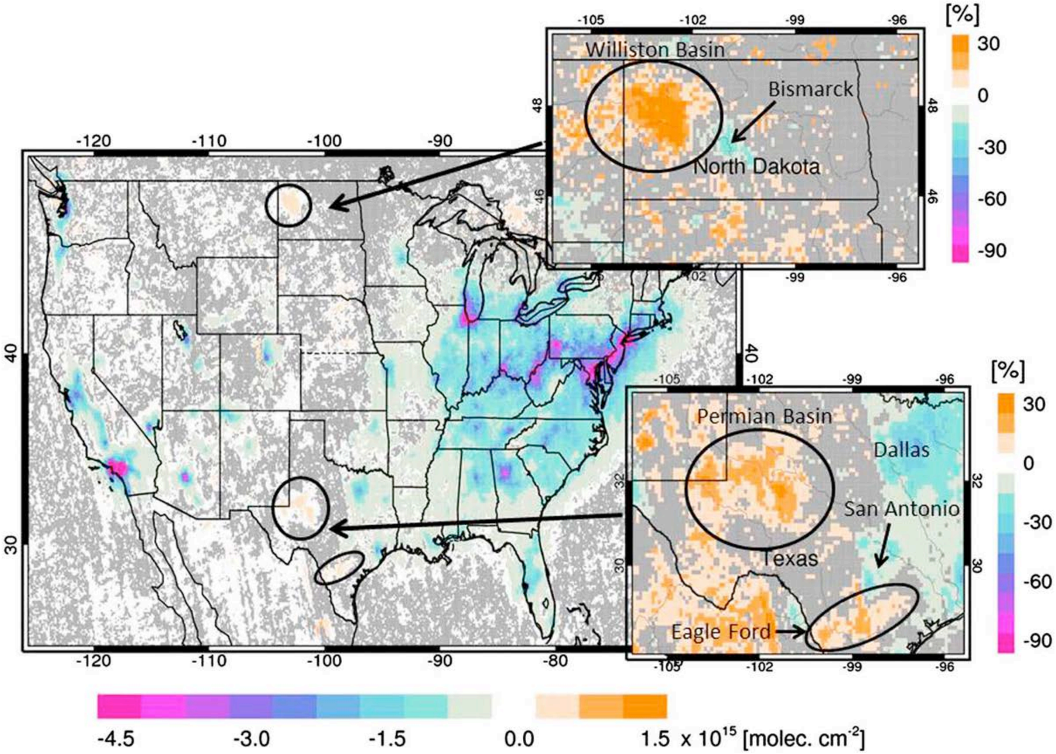
Using Satellites to Understand US Emissions Trends

Satellite (OMI) NO_2 Columns



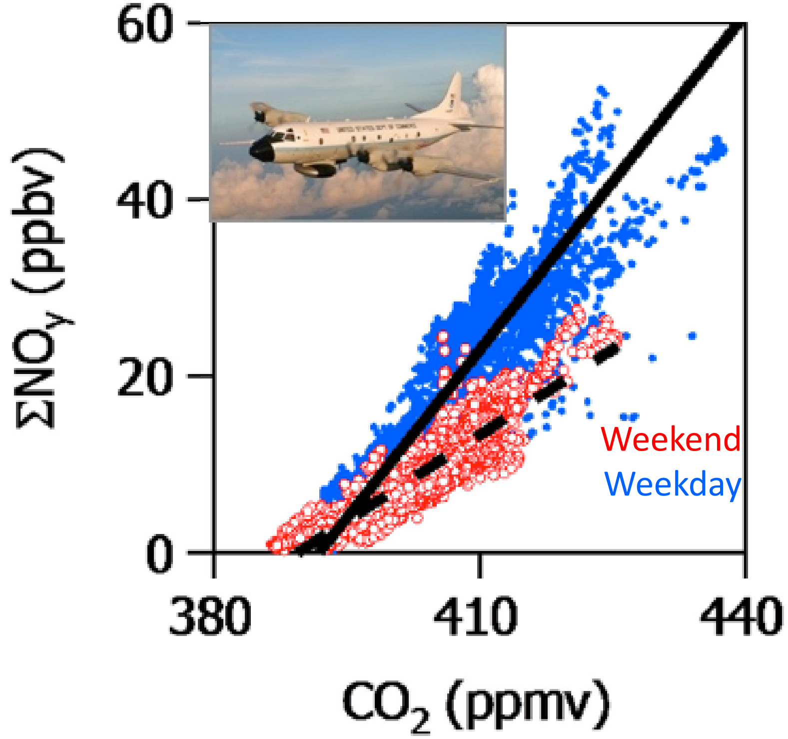
Russell, A. R., et al. (2012) Atmos. Chem. Phys.

Changes in OMI NO_2 columns, 2005-2014

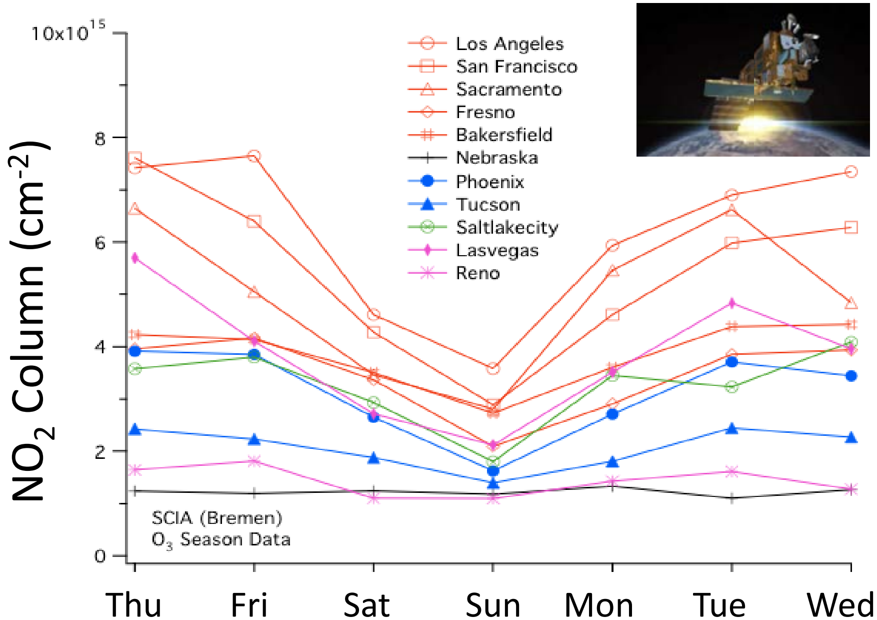


Duncan, B. N., et al. (2016) J. Geophys. Res.

Resolving day-of-week emissions variations with research aircraft and satellites

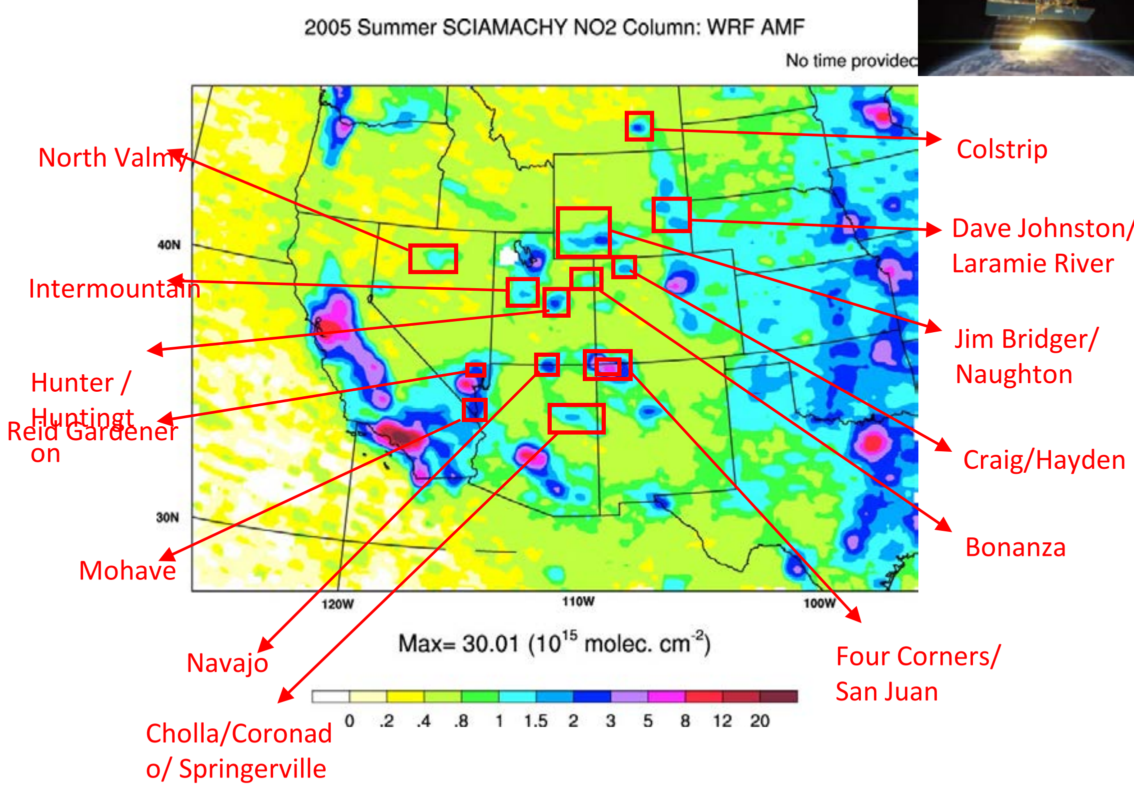


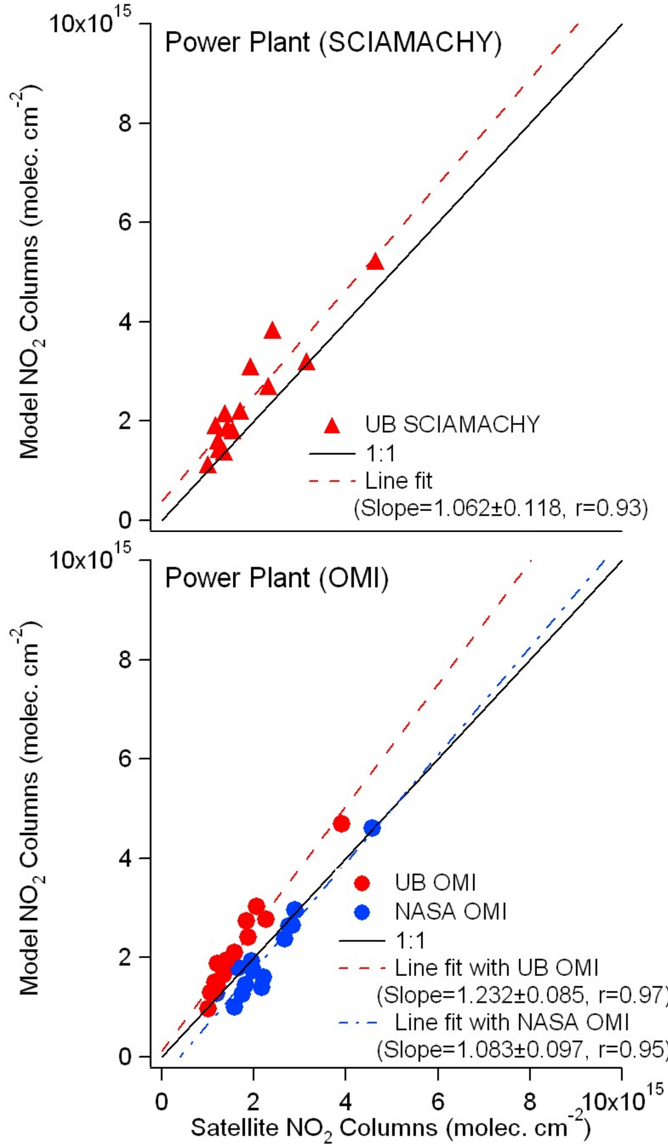
Pollack, I. B., et al. (2012) J. Geophys. Res., doi: 10.1029/2011JD016772



Kim, S.-W., et al. (2009) J. Geophys. Res., doi: 10.1029/2008JD011343

Satellite retrieval uncertainties impact emissions estimates

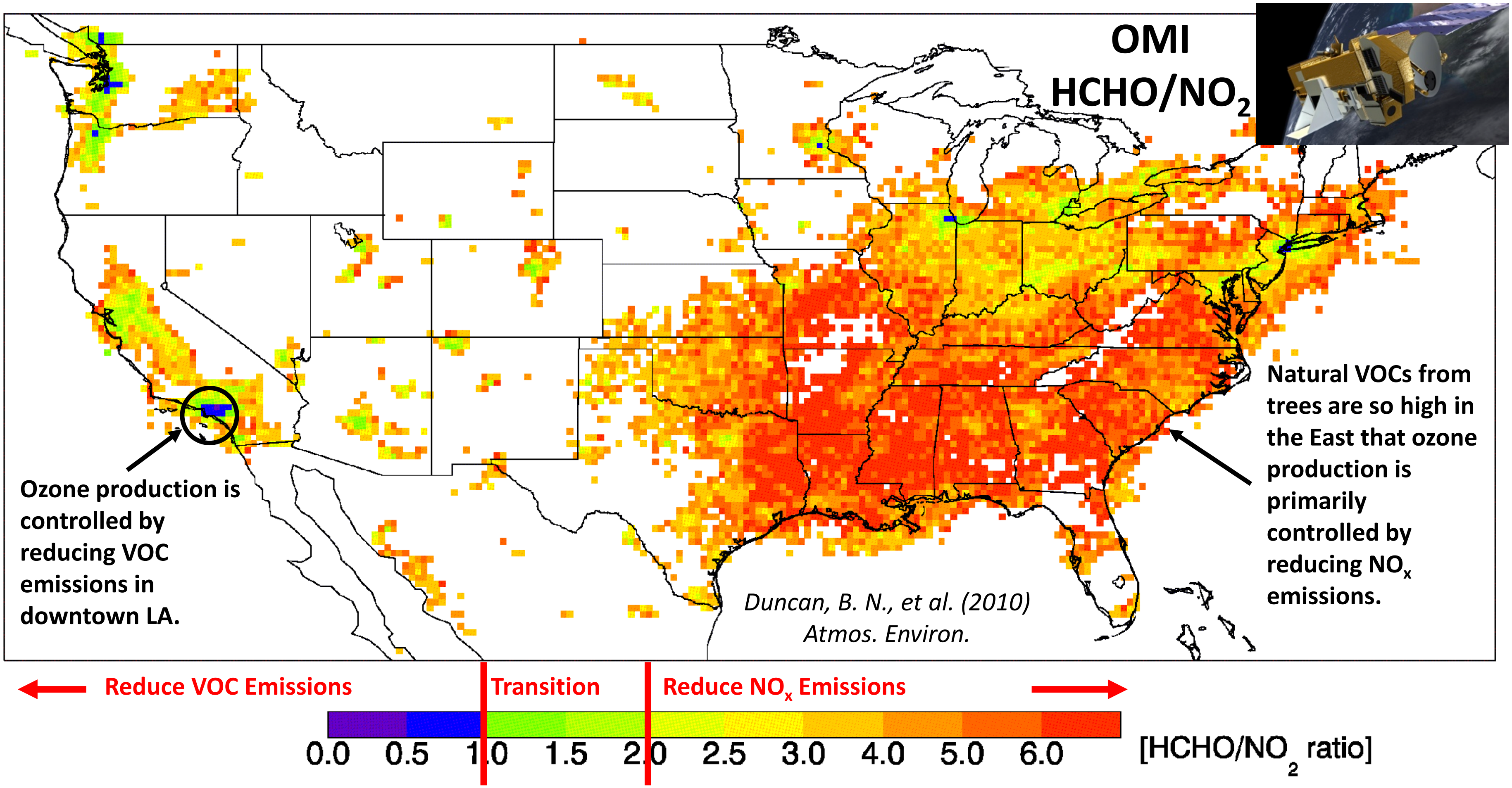




- US power plants directly monitor their emissions (CEMS)
- Leverage power plant satellite signals to understand retrieval uncertainties

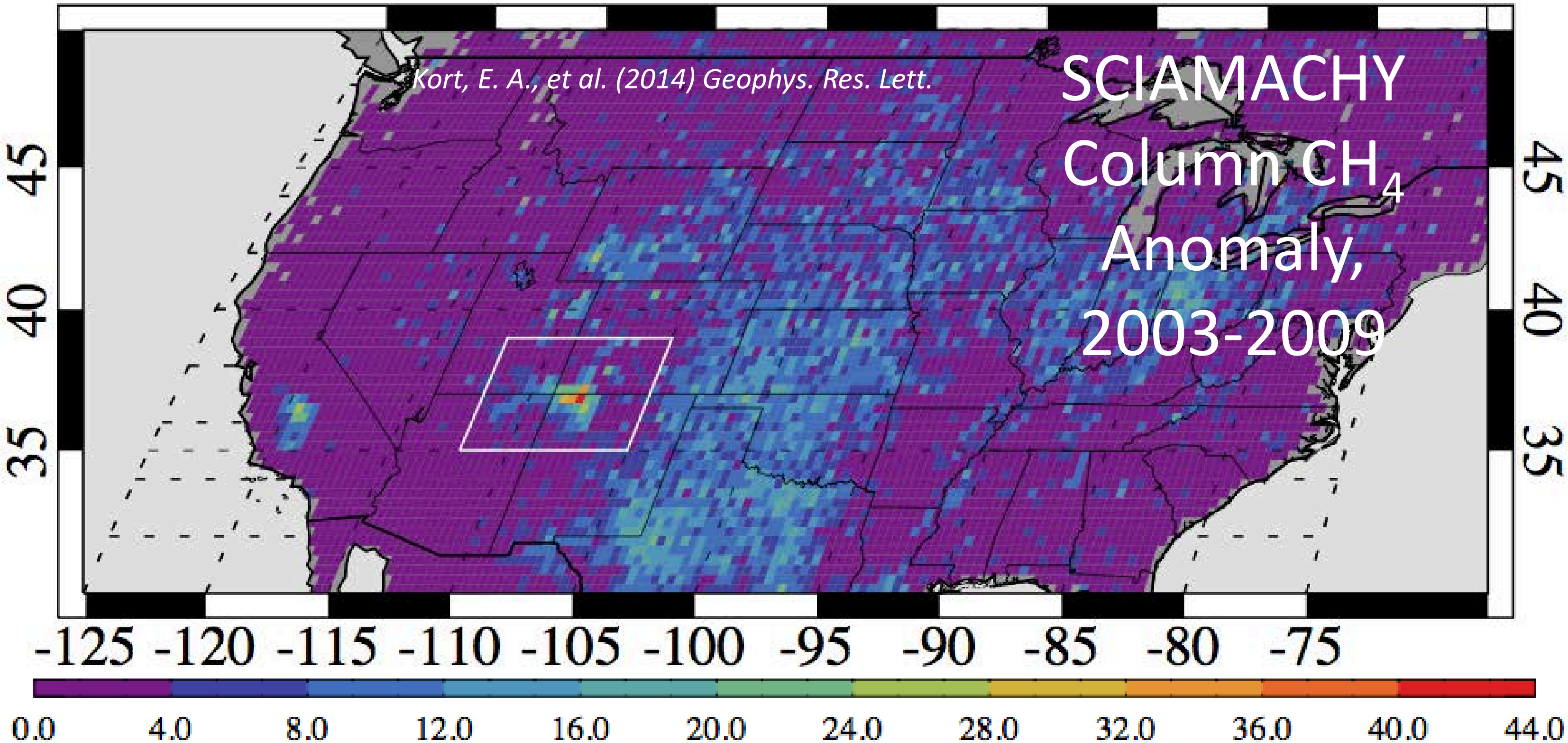
Kim, S.-W., et al. (2009) J. Geophys. Res., doi: 10.1029/2008JD011343

Using satellites to assess control strategies



- $HCHO/NO_2$ columns used to explore sensitivity of ozone formation to changes in VOC and NOx emissions
- Indicator ratios will vary with time of day, day of week, and temperature, reflecting emissions source mixtures and chemistry

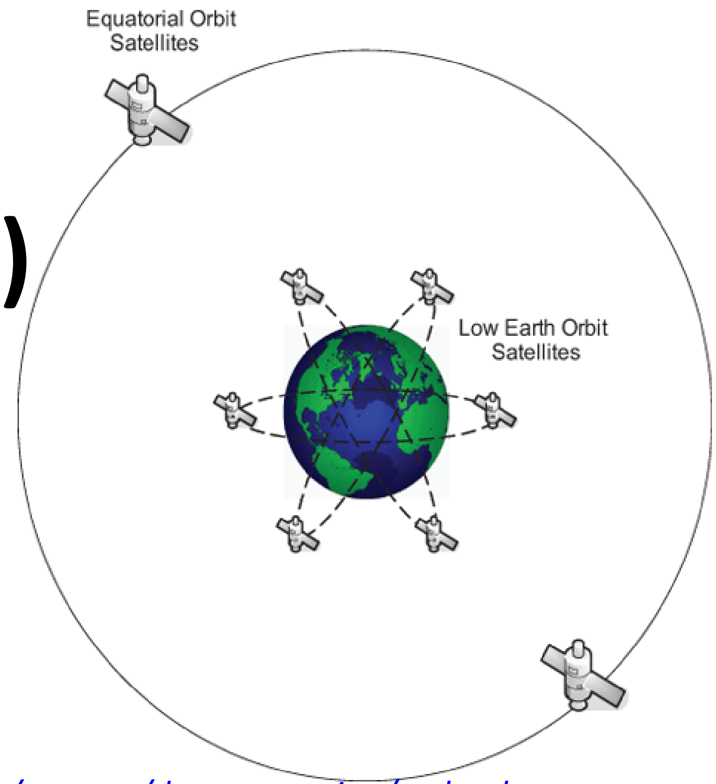
Satellite CH_4 over fossil fuel basins



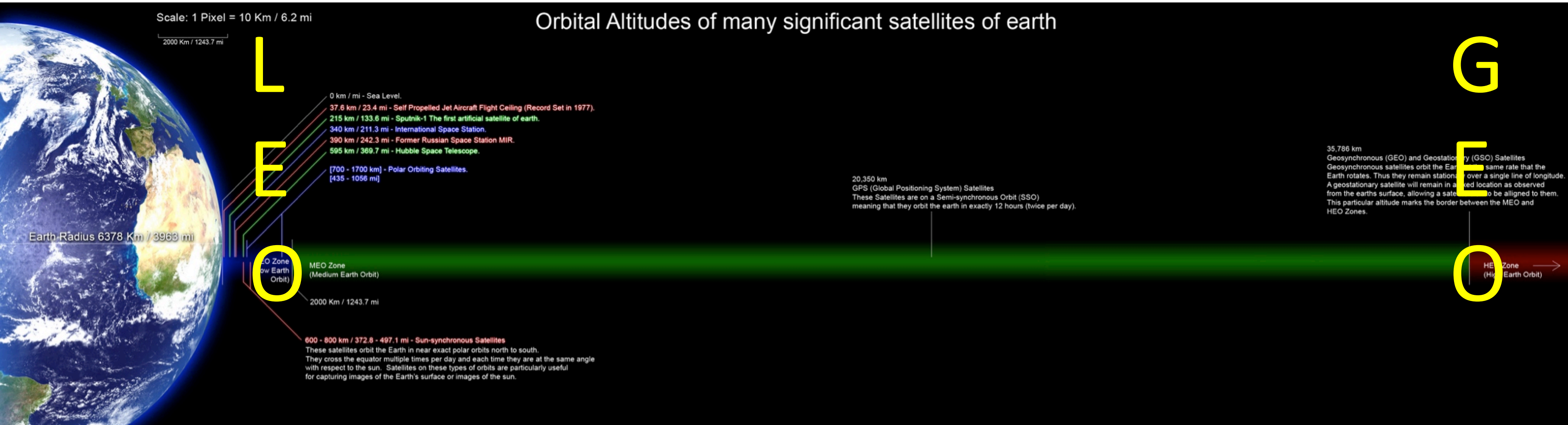
Column CH_4 anomaly (ppb)

- Largest methane anomaly in the US
- Quantified annual emissions
- Independent verification by aircraft
- What are the sources?

Low-Earth-Orbiting (LEO) vs Geostationary (GEO) Satellites

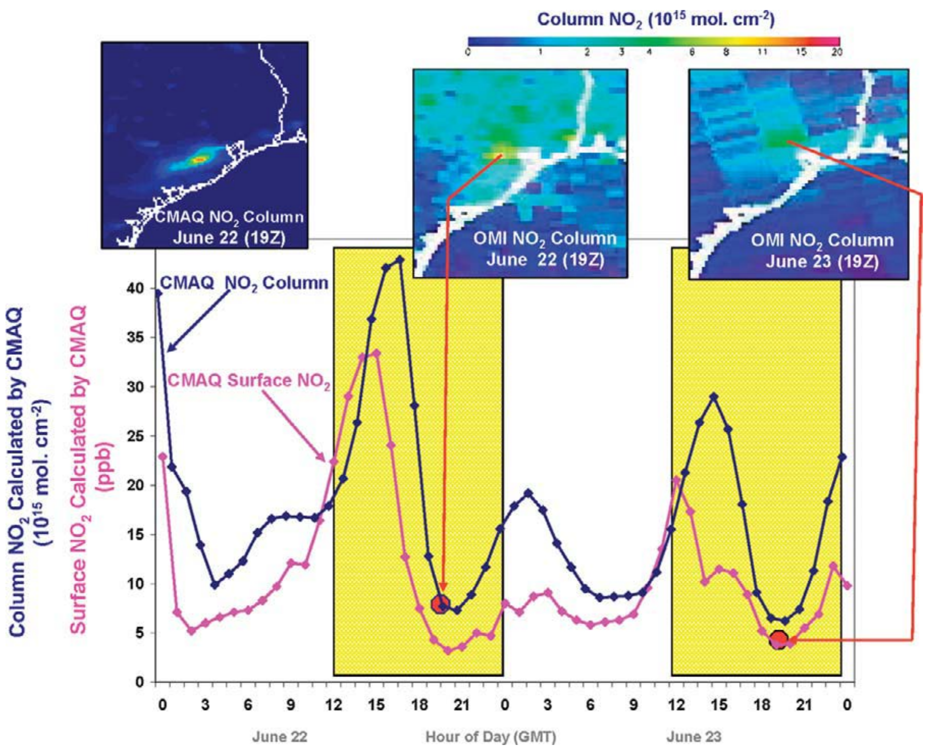


http://www.unidata.com.au/support/documentation/technology

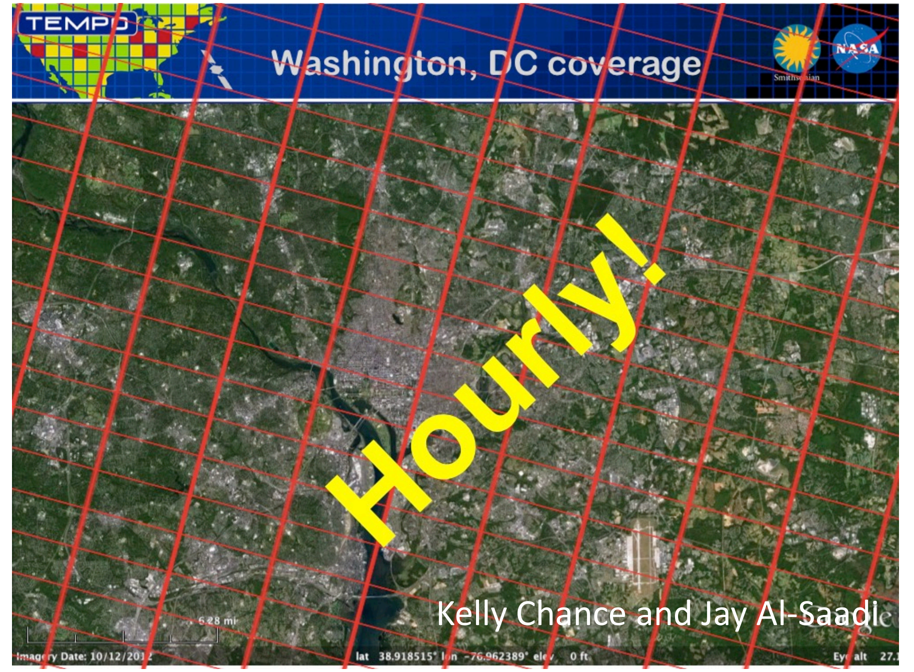


https://commons.wikimedia.org/wiki/File:Orbitalaltitudes.jpg#/media/File:Orbitalaltitudes.jpg

GEO should offer improvement over LEO for emissions constraints



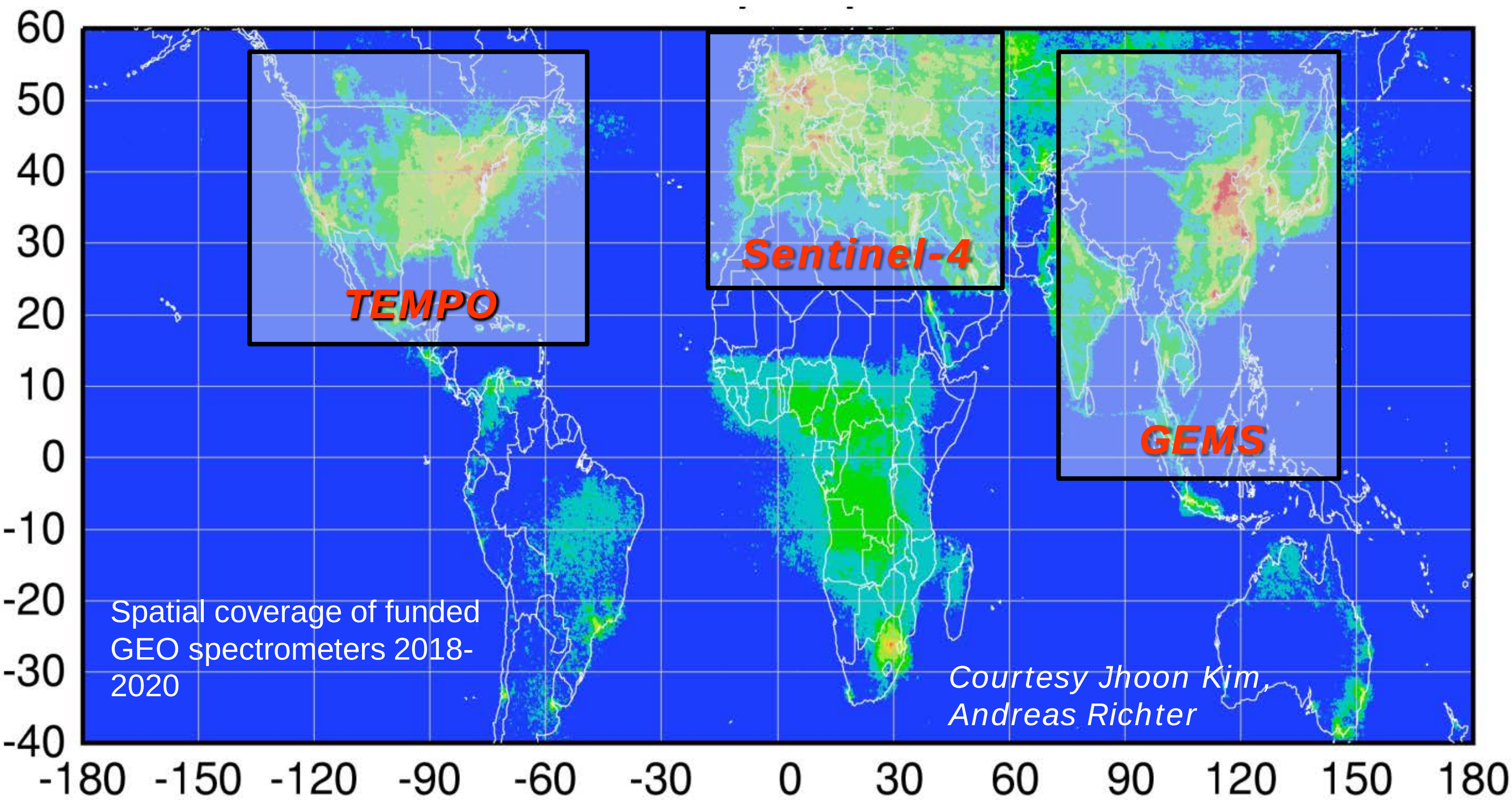
Fishman, J., et al. (2008), Bull. Amer. Meteorol. Soc.



TEMPO's hourly sampling and high spatial resolution will enable it to…

- Evaluate temporal variations, spatial mapping, and sector-specific emissions in unprecedented ways
- Resolve daytime cycles in emissions and chemistry
- Improve detection of lower frequency variations in emissions and processes

Global GEO monitoring constellation



Policy-relevant science and environmental services enabled by common observations

- Improved emissions at common confidence levels over Northern Hemisphere
- Improved air quality forecasts and assimilation systems
- Improved assessment of impacts

BREAK

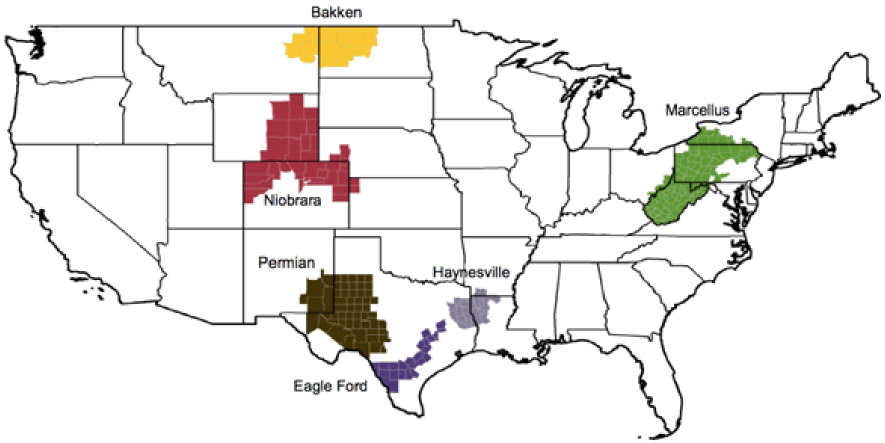
Part 3. Quantifying Specific Emissions Sources

- Oil and natural gas extraction
- Mobile sources
- Biogenic sources
- Fires and open burning

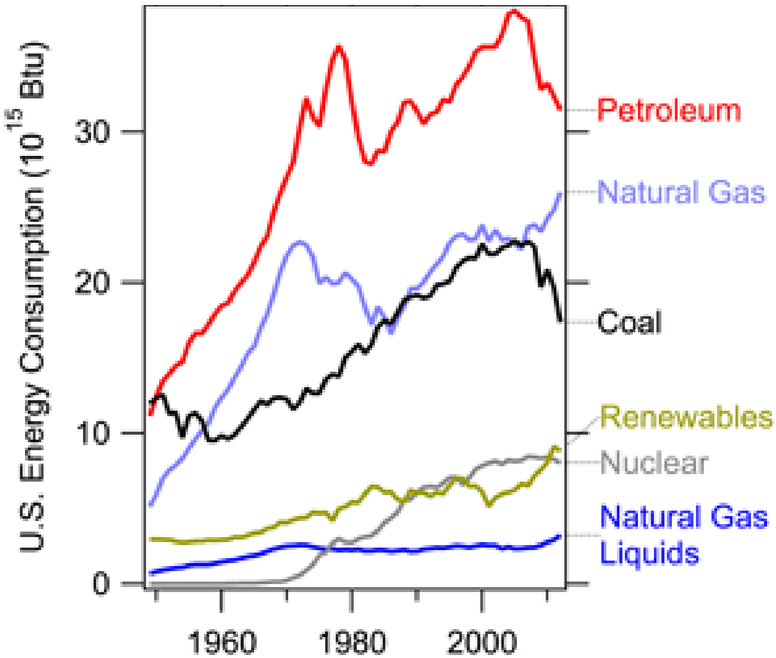
Energy in the US

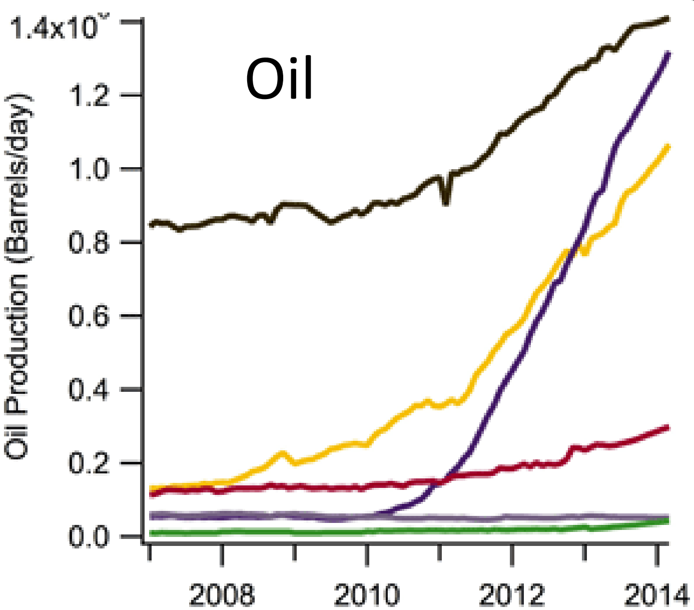
US Oil and Natural Gas Production

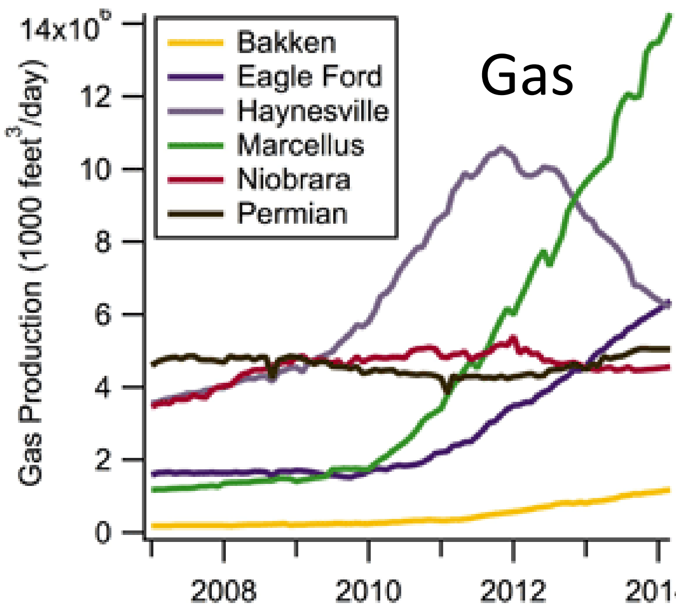
Some important tight sand/shale basins



US Energy Consumption

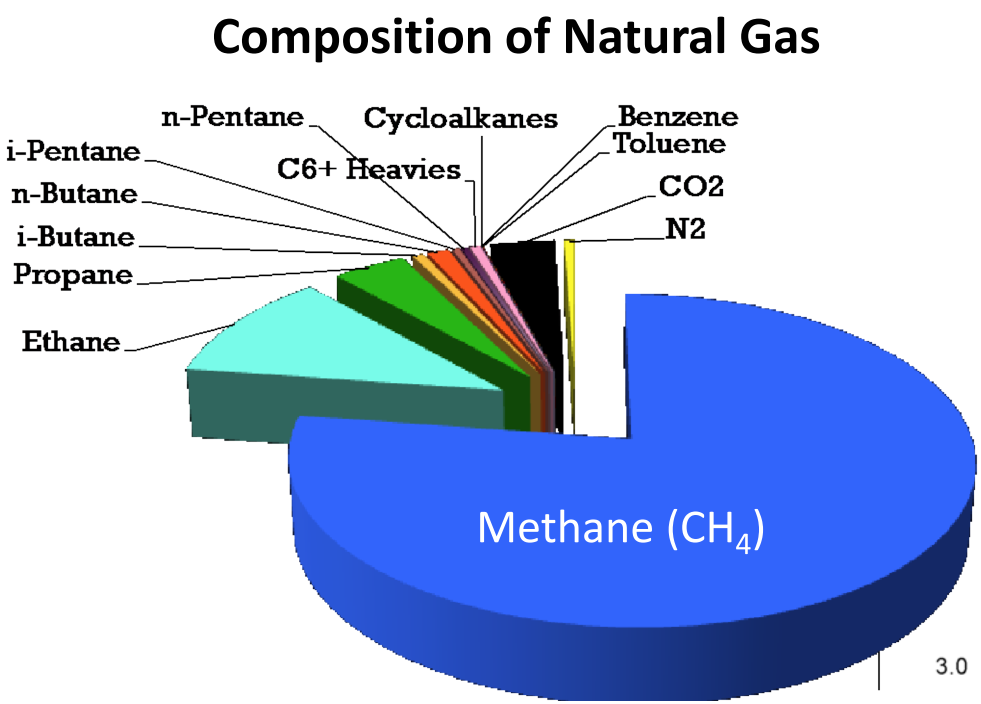






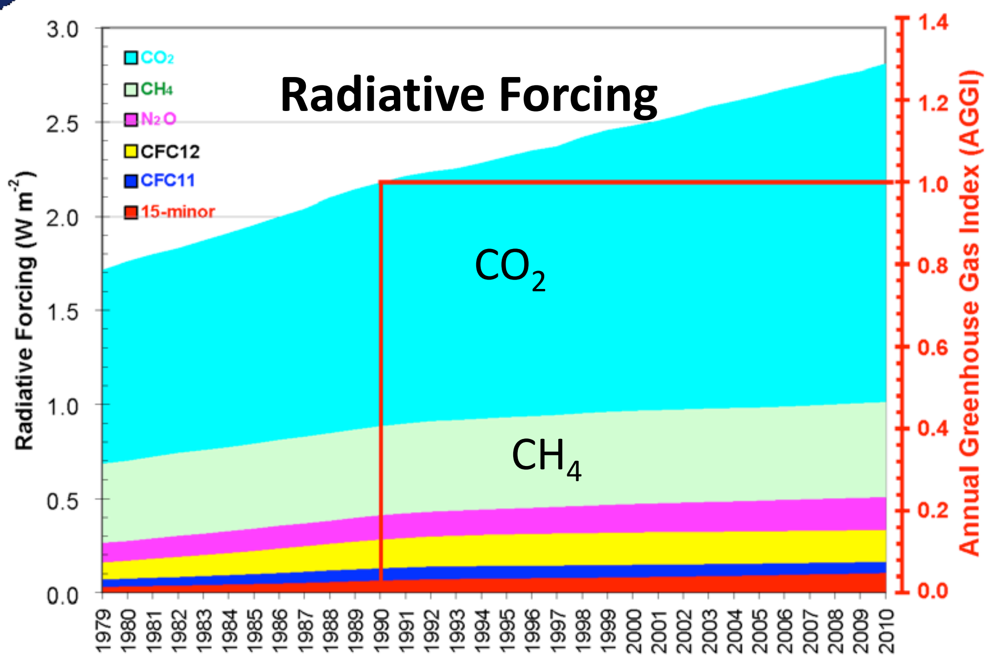
US Energy Information Administration

Potential Impacts from Oil & Gas Development

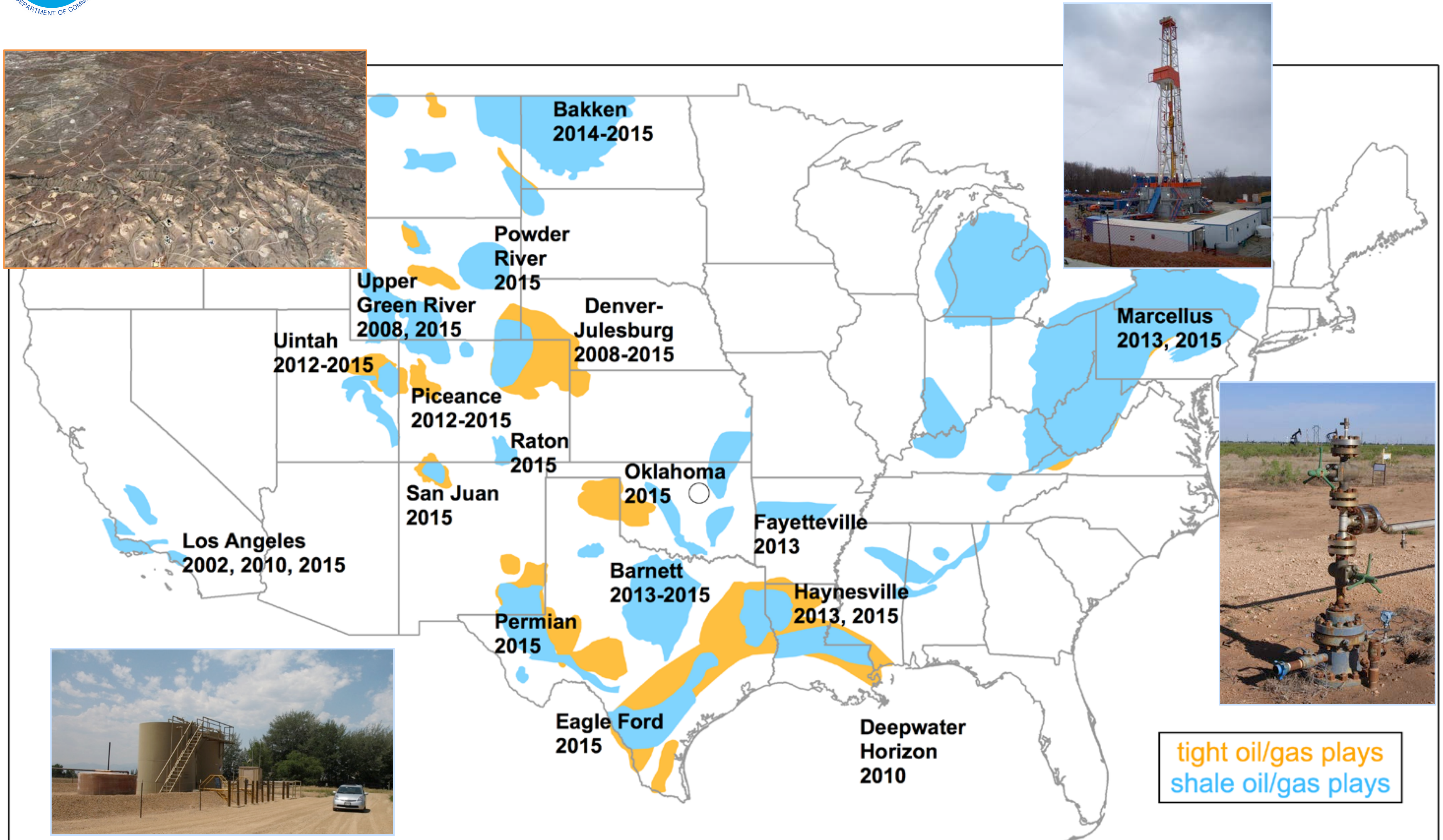


Air Quality
VOCs are precursors to ozone and particulate matter, the main constituents of smog. Many VOCs are also toxic to humans.

Climate
Methane is the 2nd most important greenhouse gas in terms of radiative forcing.



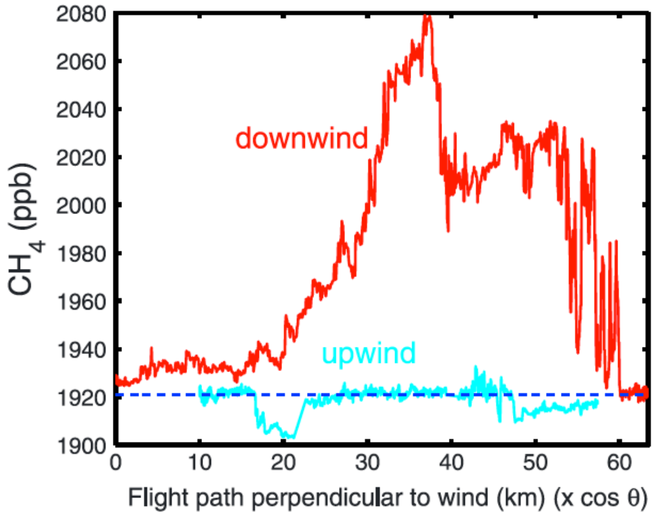
NOAA ESRL Oil/Gas Research Studies



US Energy Information Administration

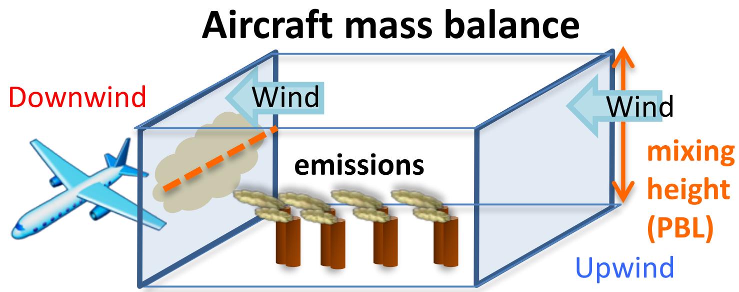
http://www.esrl.noaa.gov/csd/news/topics/oilandgas.html

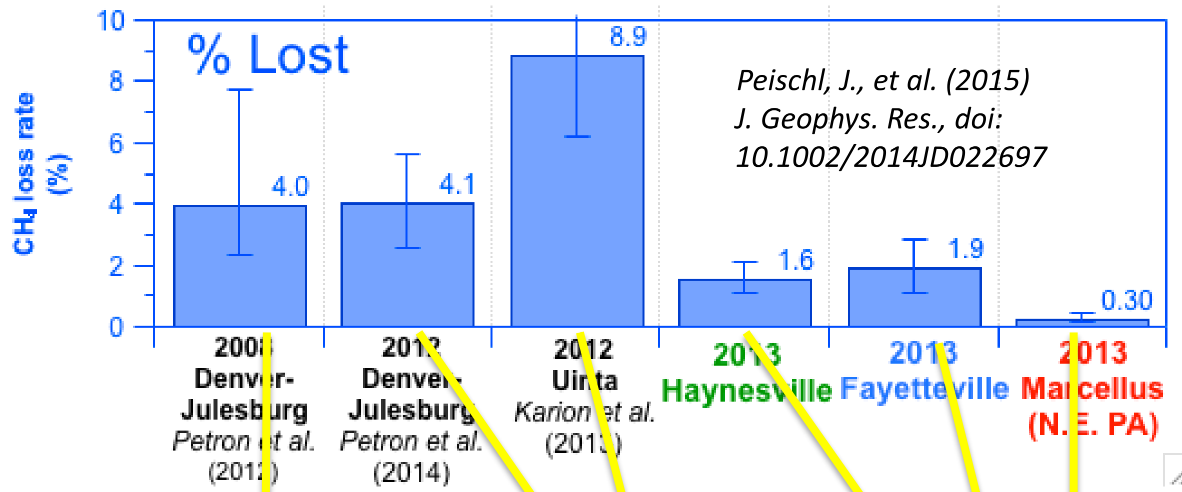
CH_4 Emissions in Oil/Gas Basins

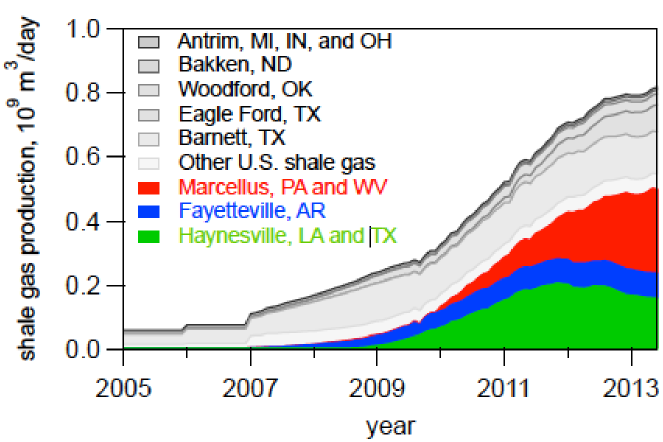


Karion, A., et al. (2013) Geophys. Res. Lett., doi: 10.1002/grl.50811

Hydrocarbon leak rates inferred from observations in oil & gas production basins



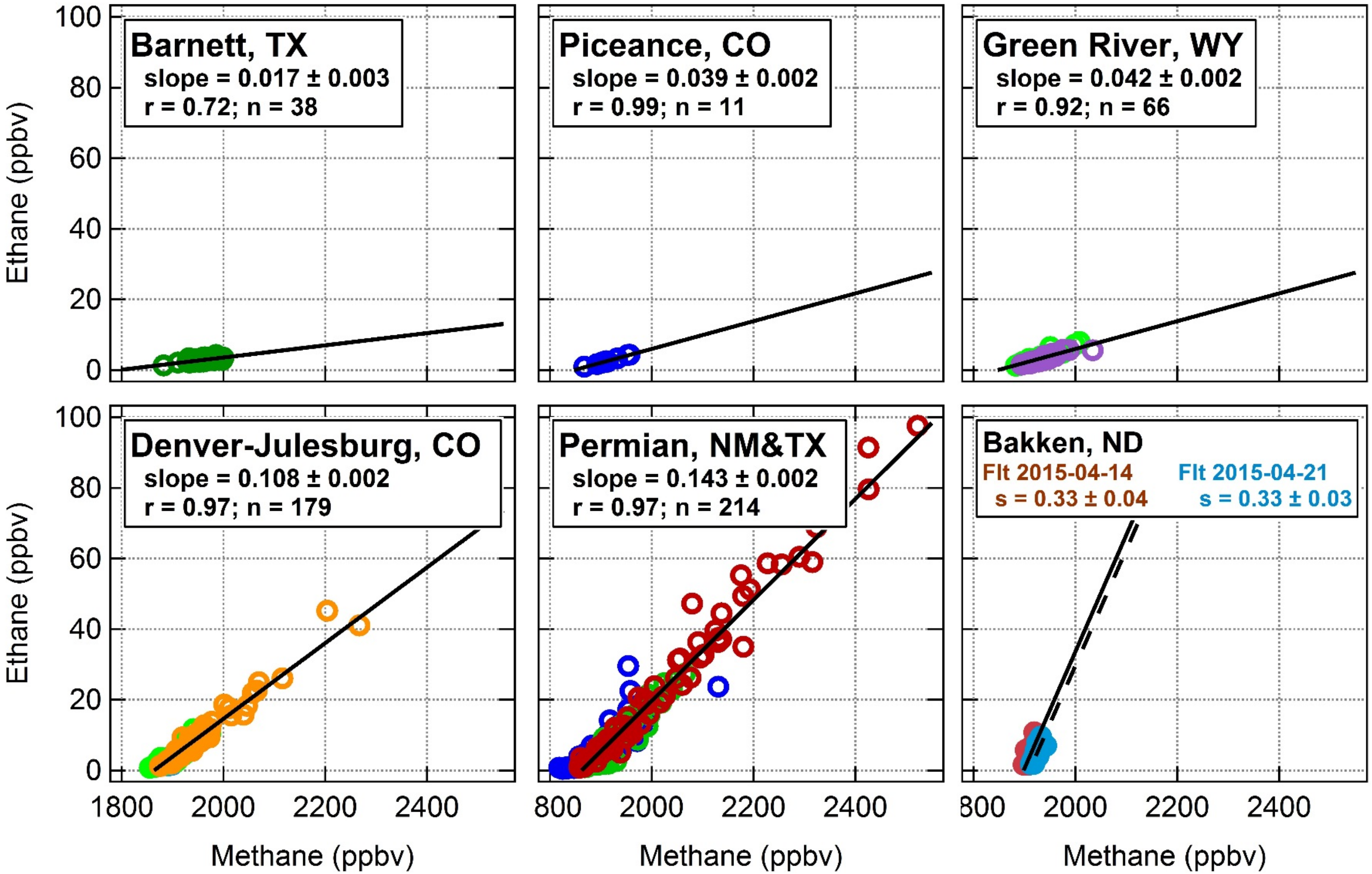




Emissions from different oil & gas basins can differ significantly

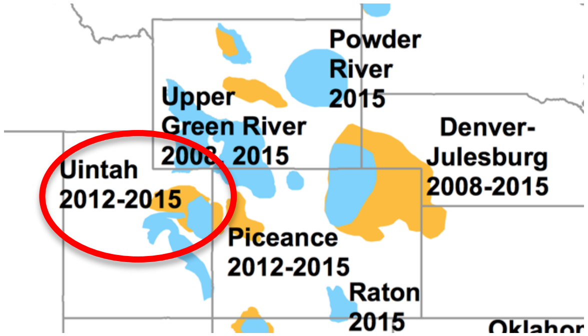
VOC/CH_4 Ratios: Chemical Fingerprint of Oil/Gas Activity

VOC Enhancement Ratios: minimize effects of air mass mixing and dilution



Jessica Gilman et al., in preparation

Atmospheric Measurements Improve Emissions in an Oil and Natural Gas Basin



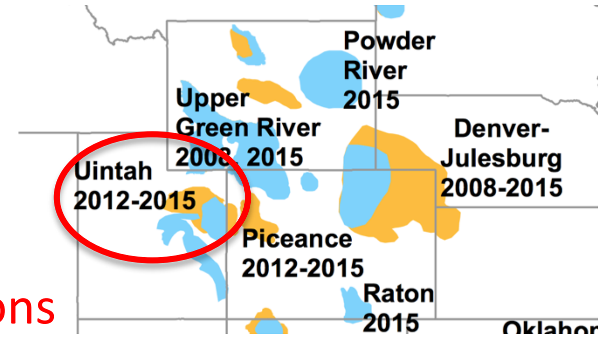
Emission datasets	Source	Methane (tons/year)	Non methane VOCs (tons/year)	NO_x (tons/year)
Bottom-up	EPA National Emission Inventory (NEI-2011)	100,279	101,184	16,448
Top-down	Based on the measurements	482,130	184,511	4,158

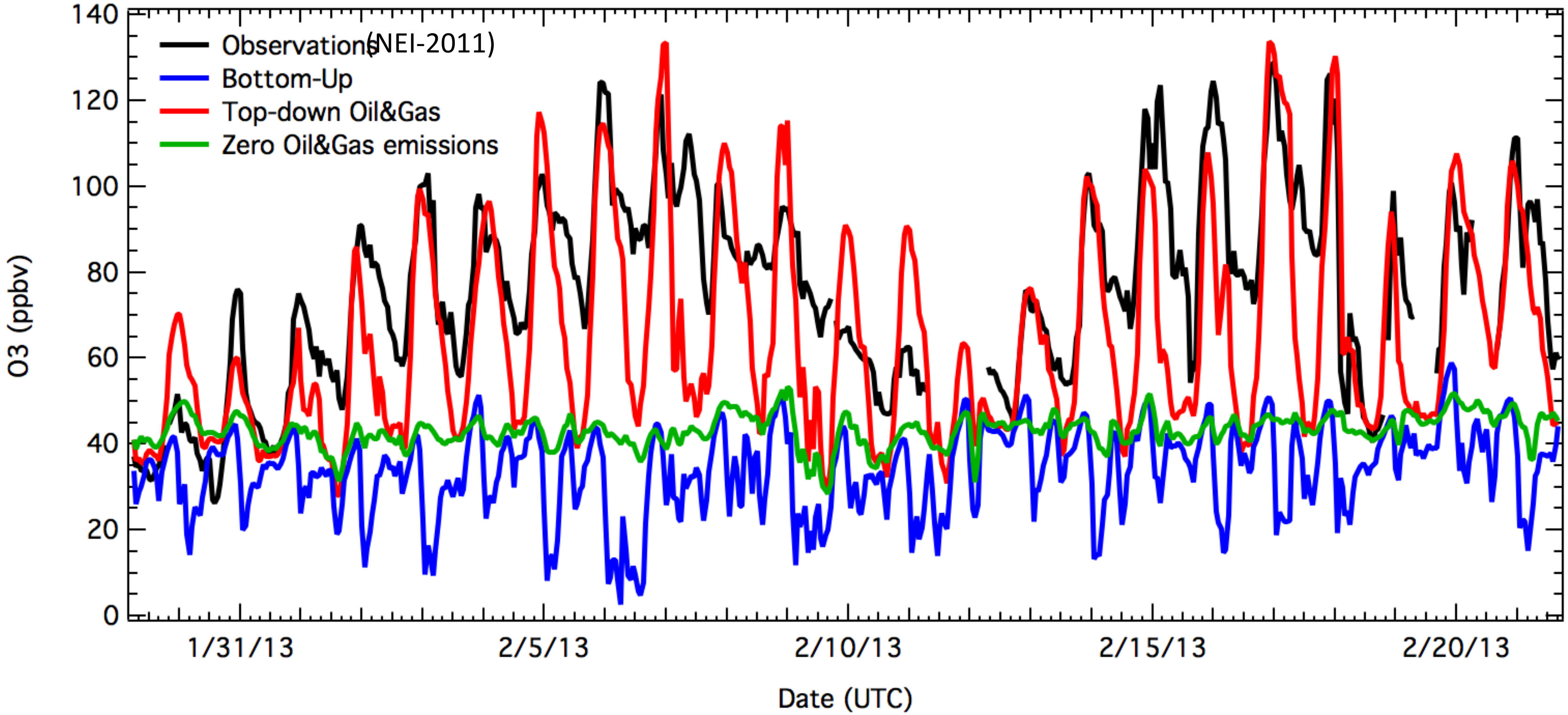
- ✓ Total top-down based methane flux estimate from *Karion et al., 2013*
- ✓ Total methane and other VOC emissions in NEI-2011 are lower by a factor of 4.8 and 1.8 than in the top-down estimates, respectively
- ✓ Conversely, NO_x emissions are 4 times higher in the NEI-2011 inventory
- ➢ Implications for air quality regulations and for climate and air quality impacts

Ahmadov, R., et al. (2015) Atmos. Chem. Phys., doi: 10.5194/acp-15-411-2015

Measurement-Based Emissions Improve O_3 Predictions in an Oil and Natural Gas Basin

- Multi-day buildup of surface O_3 during stagnation episodes
- Model using official bottom-up emissions inventory (NEI-2011) can't reproduce observed high O_3 levels
- Measured emissions case explains high O_3 levels
- High O_3 in this basin driven mostly by oil/gas emissions





Ahmadov, R., et al. (2015) Atmos. Chem. Phys., doi: 10.5194/acp-15-411-2015

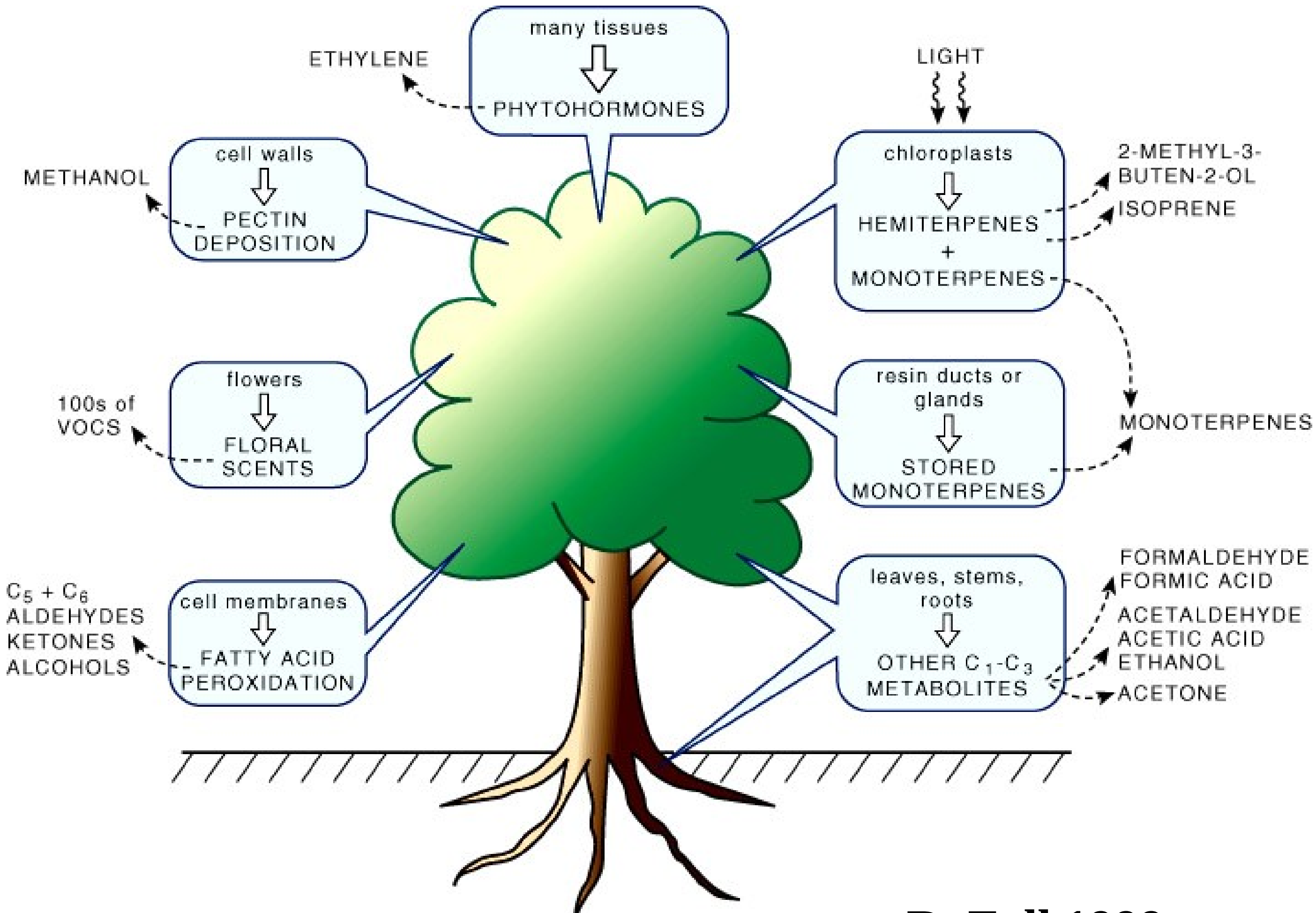
Mobile sources

Switch to Mobile Sources presentation file

Biogenic Emissions

Biogenic Emissions

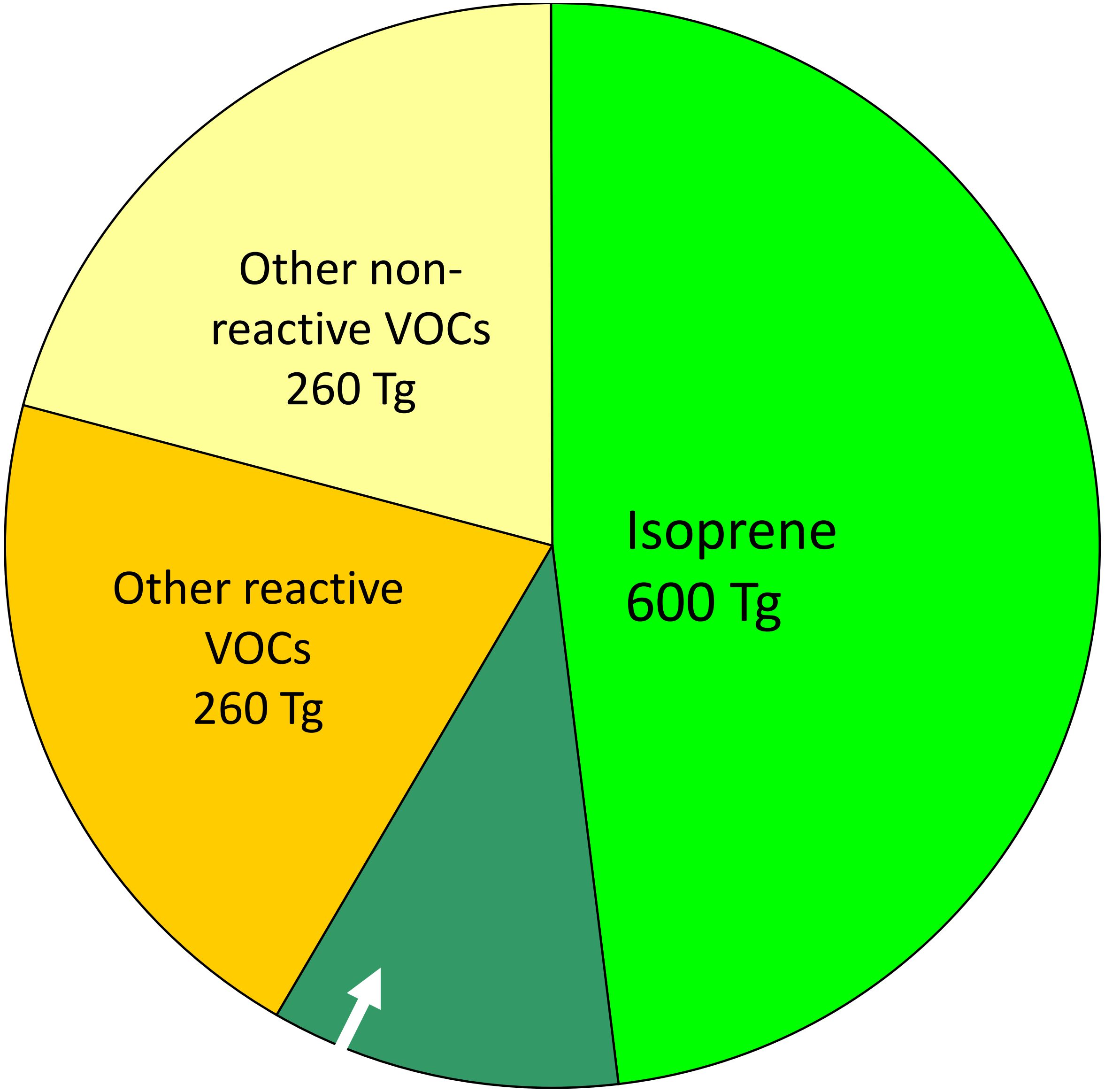
- What is emitted?



R. Fall 1999

Global Estimates of Non-Methane VOC Emissions

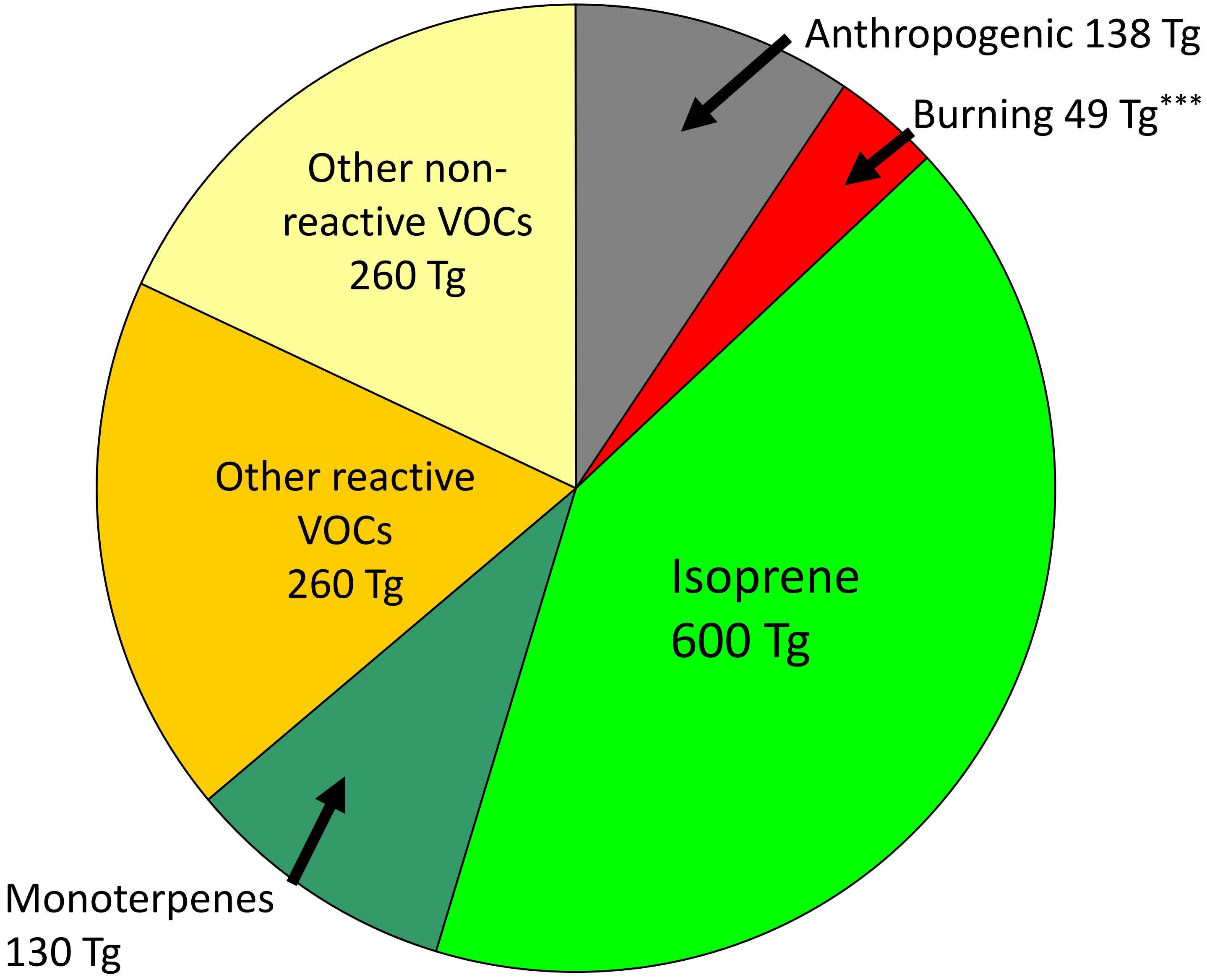
Total:
~1250 Tg yr^{-1}



Monoterpenes
130 Tg

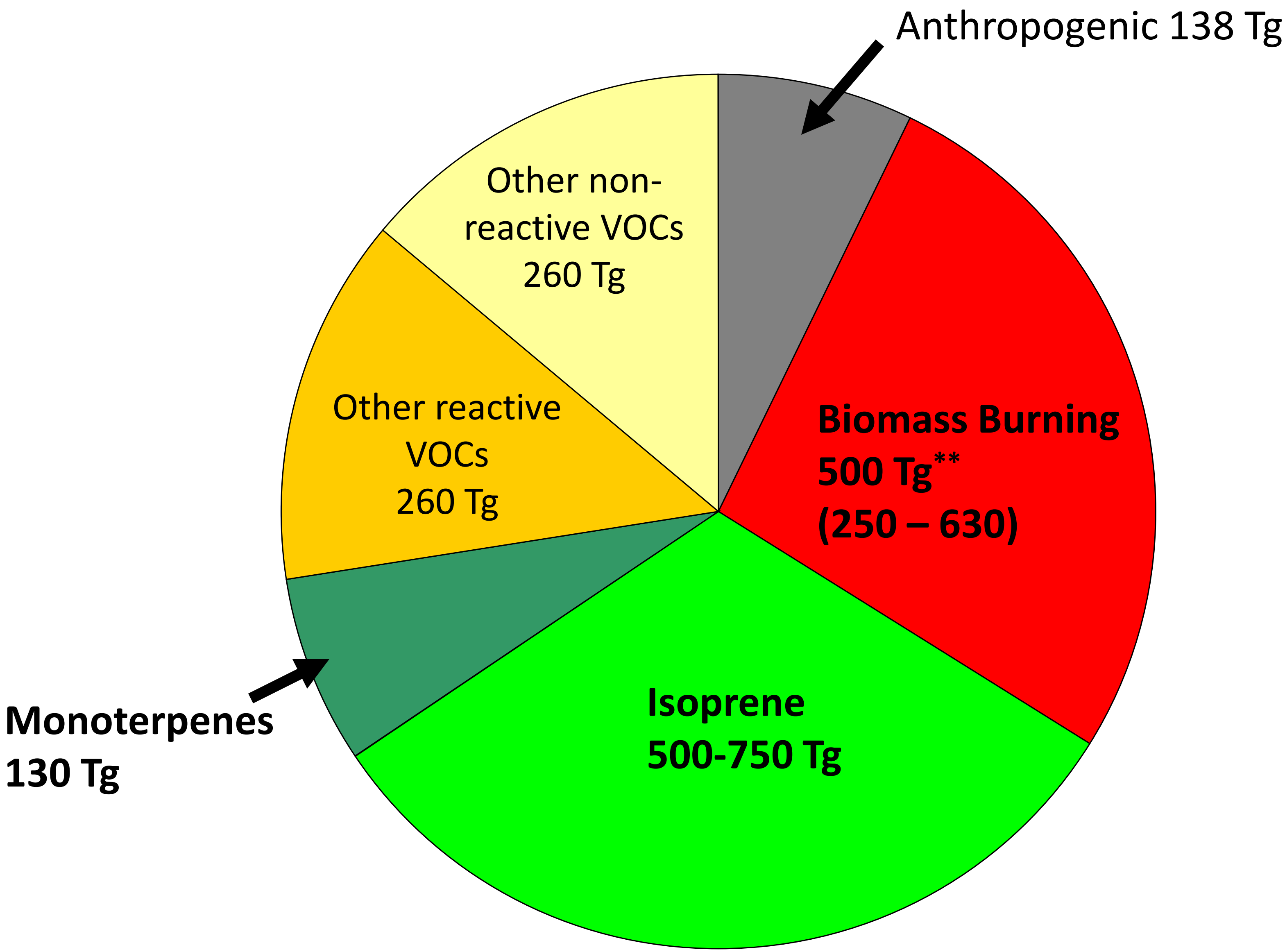
Guenther et al. 1995; Guenther et al. 2006

Global Estimates of Non-Methane VOC Emissions



EDGAR32FT2000; Guenther et al. 1995; Guenther et al. 2006

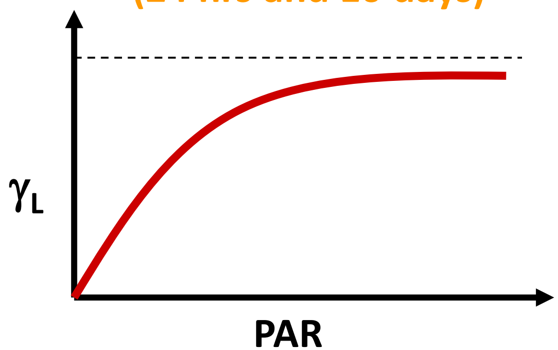
Global Estimates of Non-Methane VOC Emissions

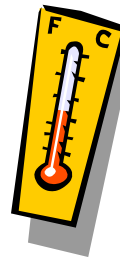


**Yokelson et al., *Atmos. Chem. & Physics, 2008*

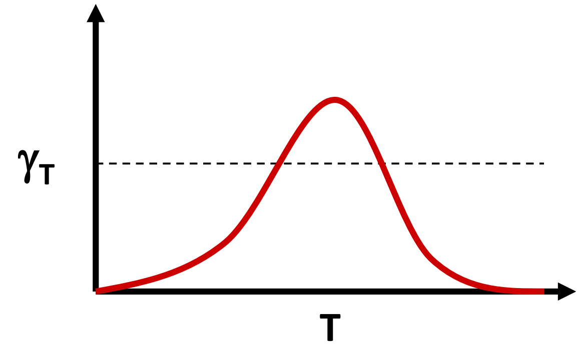
Meteorological and Phenological Variables Controlling Biogenic Isoprene Emissions

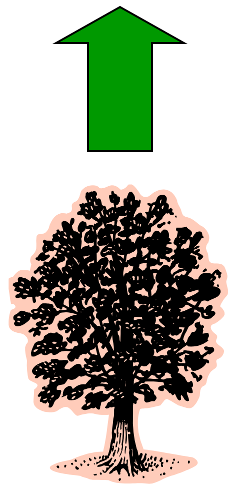
LIGHT
→Diffuse and direct radiation
→Instantaneous and accumulated
(24 hrs and 10 days)

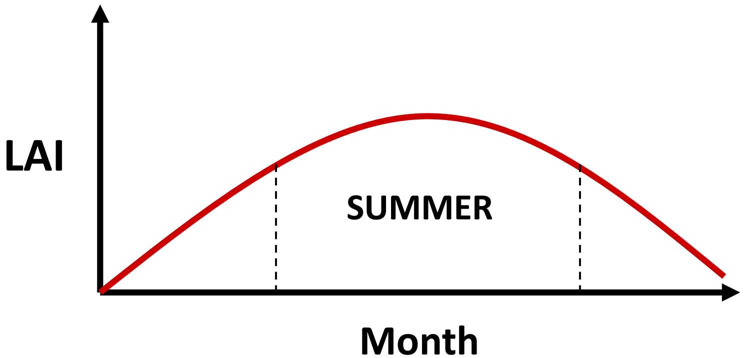




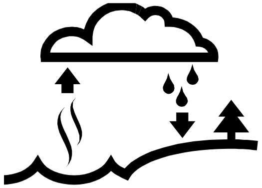
TEMPERATURE (Leaf-level)
→instantaneous and accumulated
(24 hrs, 10 days)







LEAF AGE
→Max emission = mature
→Zero emission = new



SOIL MOISTURE
→ suppressed under drought

Slide from C. Heald (CSU)

[Guenther et al., 2006]

Modeling Biogenic Emissions

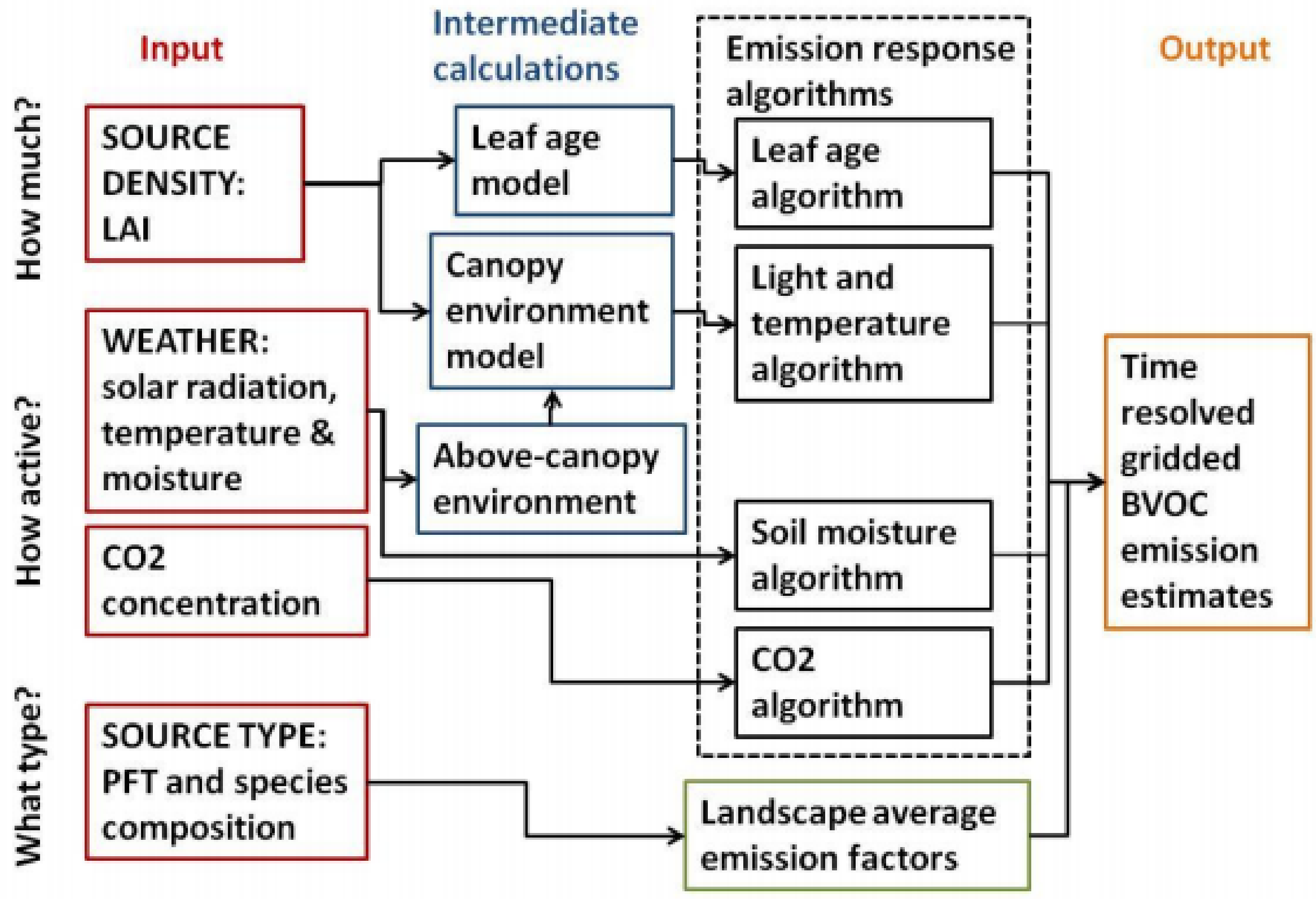


Fig. 1. Schematic of MEGAN2.1 model components and driving variables.

Guenther et al., 2012

$$EM = \varepsilon \bullet \gamma_{CE} \bullet \gamma_{age} \bullet \gamma_{SM} \bullet \rho$$

(Guenther et al., 2006)

EM: Emission ($\mu g\ m^{-2}\ hr^{-1}$)

ε: Emission Factor ($\mu g\ m^{-2}\ hr^{-1}$)

γ_{CE}: Canopy Factor

γ_{age}: Leaf Age Factor

γ_{SM} : Soil Moisture Factor

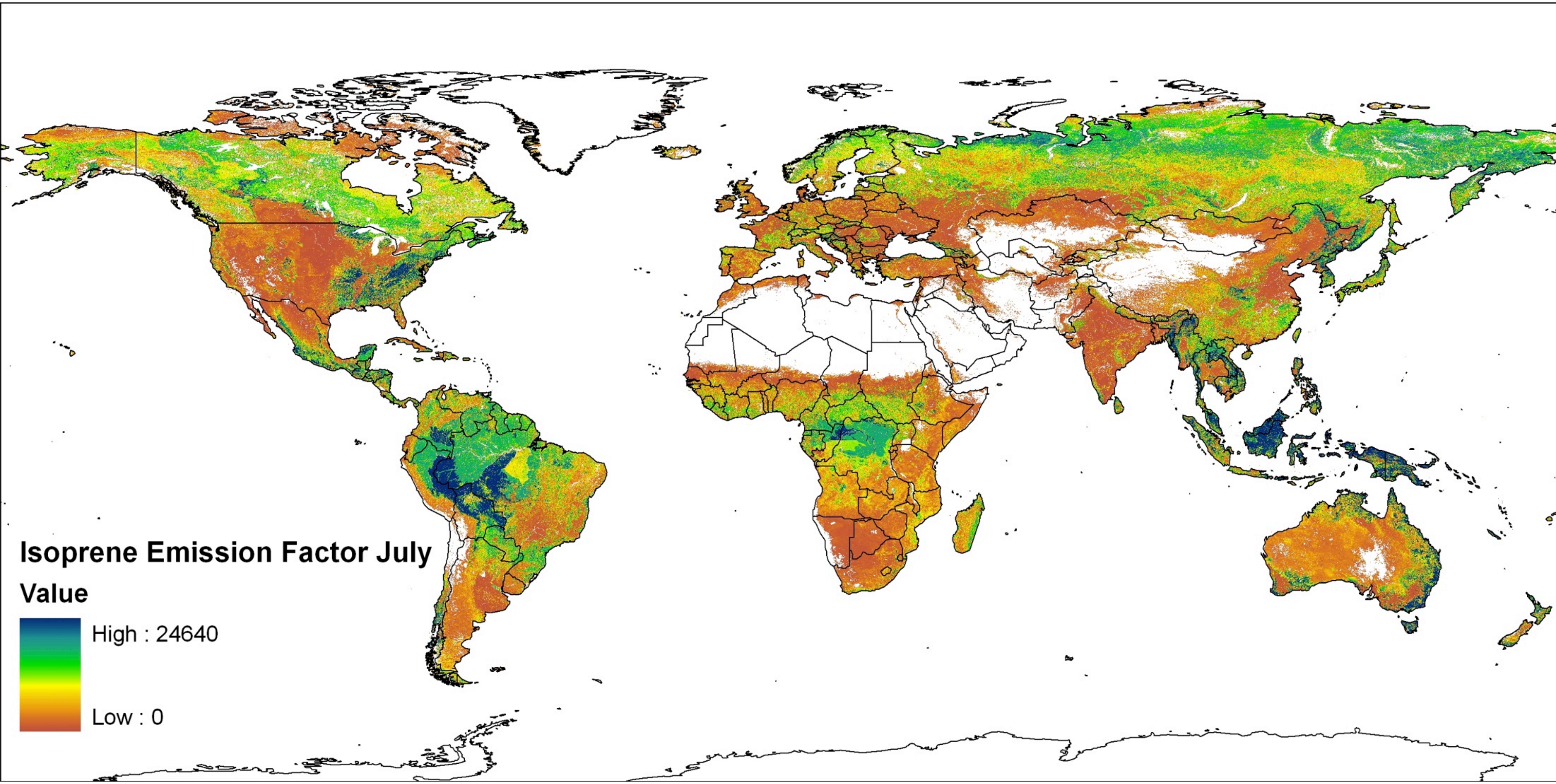
ρ: Loss and Production within plant canopy

Emission Factors assigned:

- By Plant Functional Type
- Area-Specific Map

Isoprene Emission Factor Map

@ Standard Conditions (July LAI)



$$EM = \varepsilon \bullet \gamma_{CE} \bullet \gamma_{age} \bullet \gamma_{SM} \bullet \rho$$

$$\gamma_{CE} = \gamma_{LAI} \bullet \gamma_{P} \bullet \gamma_{T}$$

(Guenther et al., 2006)

EM: Emission ($\mu g\ m^{-2}\ hr^{-1}$)

ε: Emission Factor ($\mu g\ m^{-2}\ hr^{-1}$) → Emission Factors assigned:
- By Plant Functional Type
- Area-Specific Map

γ_{CE}: Canopy Factor

γ_{age}: Leaf Age Factor

γ_{SM} : Soil Moisture Factor

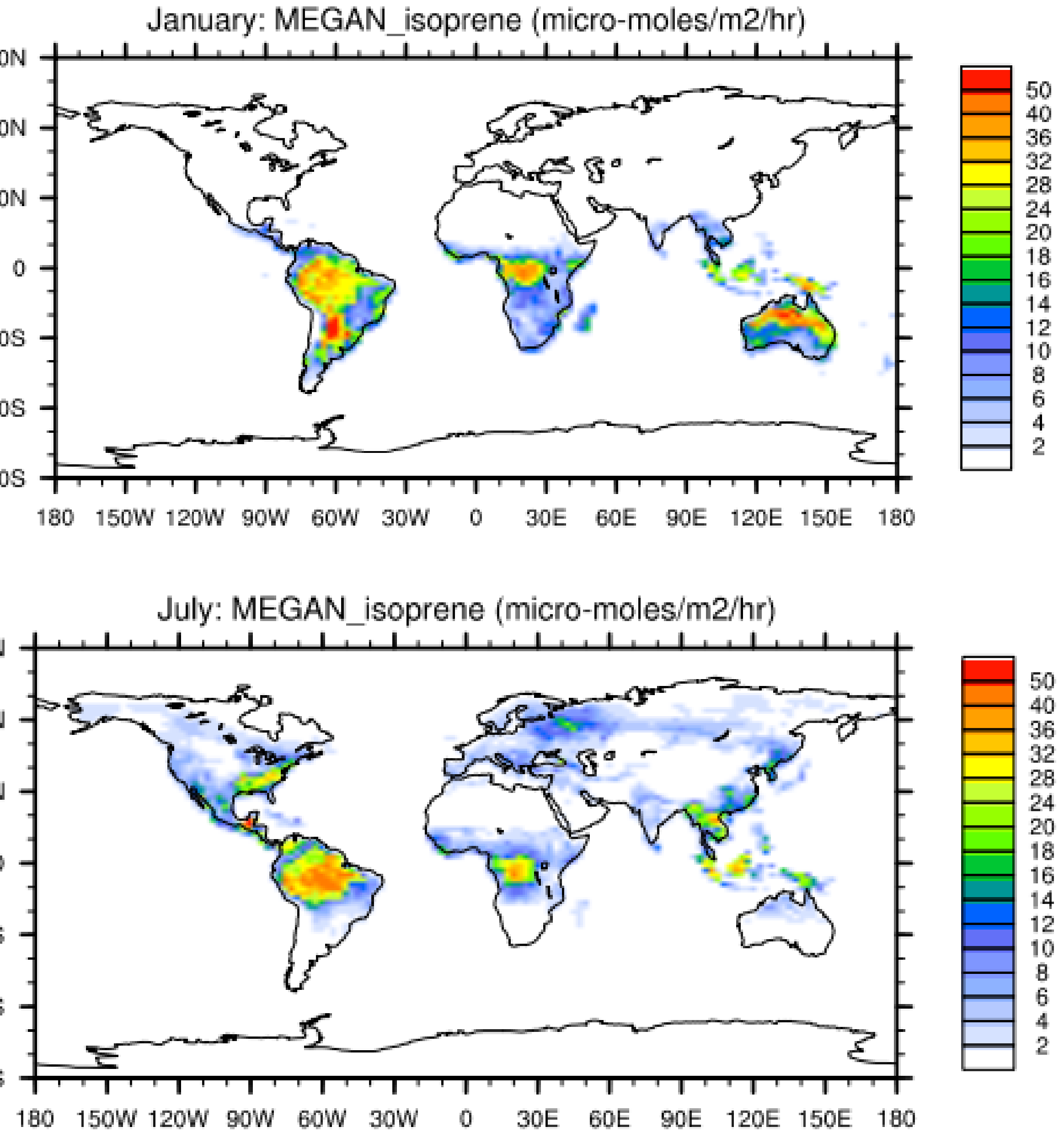
ρ: Loss and Production within plant canopy

γ_{LAI} : Leaf Area Index Factor

γ_{P} : PPFD Emission Activity Factor (light-dependence)

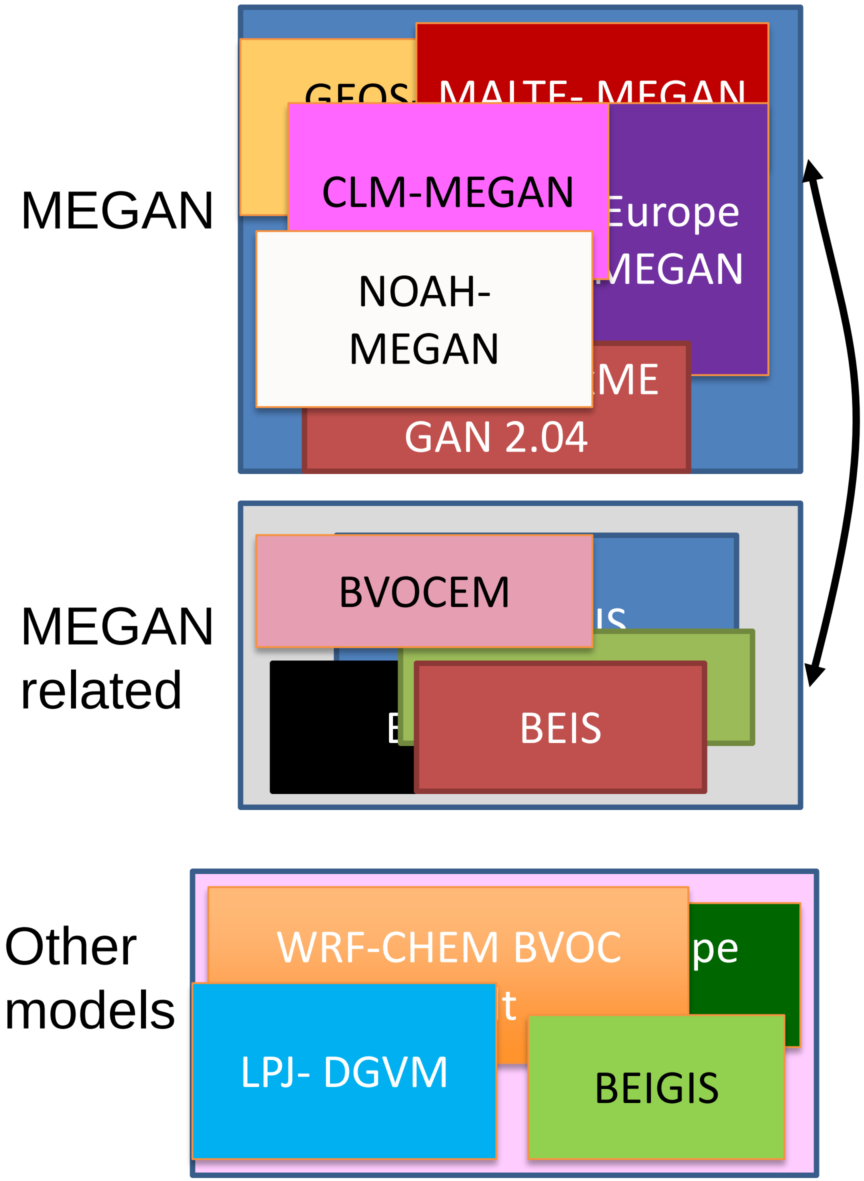
γ_{T}: Temperature Response Factor

Isoprene Emission Estimates (MEGAN v2.1)



Guenther et al. 2012

Biogenic VOC emission models



Although there are few comparisons……

Between-model emission differences are about the same as within-model differences (i.e. estimated with different versions of the same or similar model)

For example, Areneth et al. (2011) found that emission differences between MEGAN and the MEGAN-related BVOCEM model are greater than the differences between MEGAN and LPJ-DGVM

Different versions of MEGAN (or just using different inputs) result in even larger differences
(Guenther et al. 2006; Mueller et al. 2008; Barkley et al. 2008)

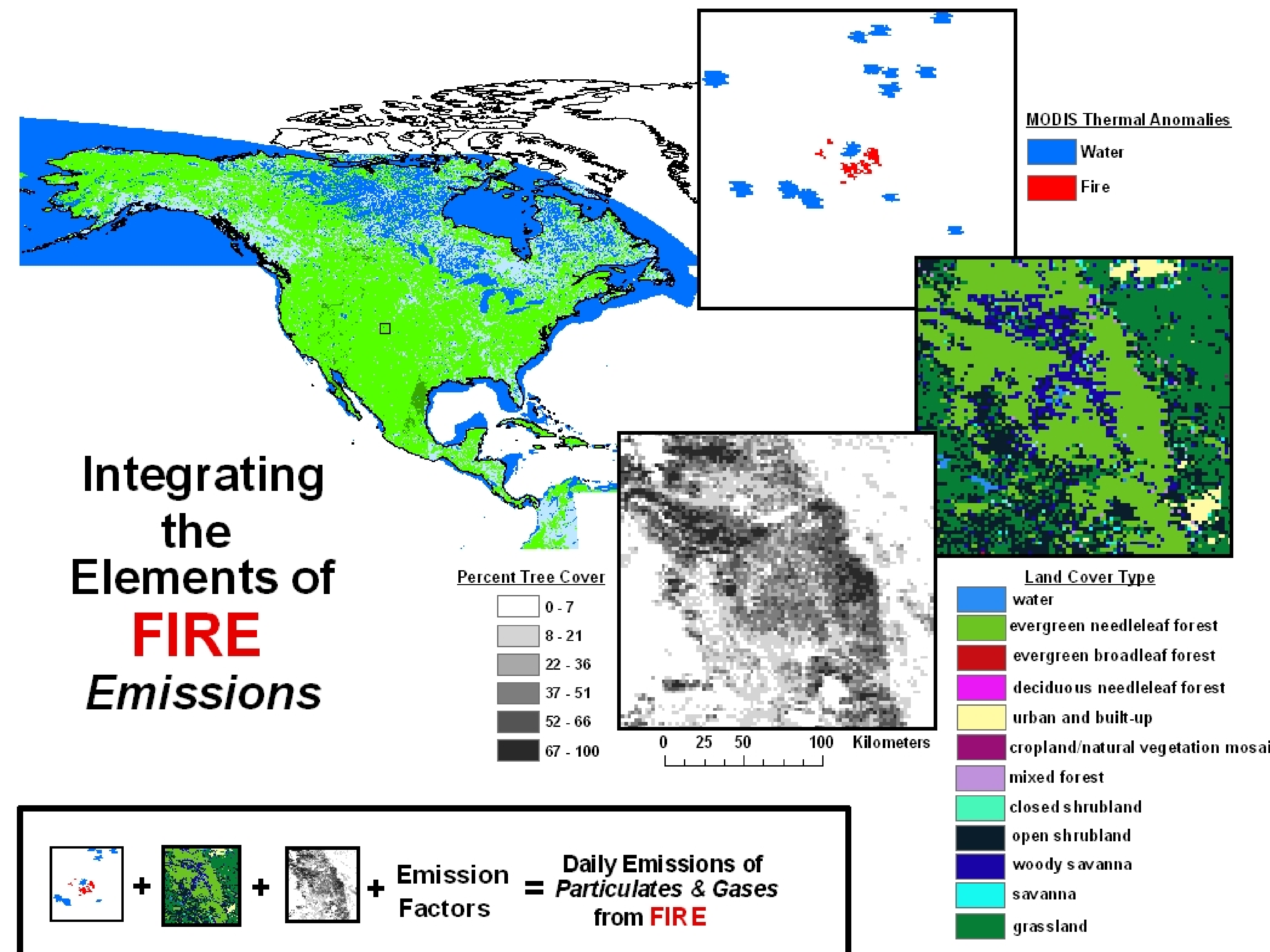

Fires and open burning

Modeling Fire Emissions requires multiple inputs:

- fire location/timing
- burned area
- fuel loading
- fuel burned
- emission factors

Many efforts to develop fire emissions inventories & models

- Global, regional, local
- Various resolutions
- Various species
- Driven by different data (remote sensing/reports/ both)



Integrating the Elements of FIRE Emissions
MODIS Thermal Anomalies
Water
Fire
Percent Tree Cover
0 - 7
8 - 21
22 - 36
37 - 51
52 - 66
67 - 100
0 25 50 100 Kilometers
Land Cover Type
water
evergreen needleleaf forest
evergreen broadleaf forest
deciduous needleleaf forest
urban and built-up
cropland/natural vegetation mosai
mixed forest
closed shrubland
open shrubland
woody savanna
savanna
grassland
+ + + Emission Factors = Daily Emissions of Particulates & Gases from FIRE

Modeling Fire Emissions

$$Emissions_i = f(A(x,t), B(x), EF_i)$$

A(x,t): Area burned

B(x): Biomass burned

- type of vegetation (ecology)
- fuel characteristics:
  - amounts of woody biomass, leaf biomass, litter
  - fuel condition
  - moisture content

EF_i: Emission factor (mass of emission$_i$ /biomass burned)

- fuel characteristics
- fuel condition

- Satellite Data
  - Location and timing of fires
  - Vegetation distribution
  - Density of vegetation
- Laboratory Measurements
- Field Observations

Estimating Emissions

Emission Factor
(g/kg)

- Based on laboratory and field measurements
- Dependent on measurement techniques
- Function of type of burning

Thomas Karl, TROFFEE Study, Brazil
Montana Fire Sciences Laboratory (B. Yokelson)

Emission factors for open and domestic biomass burning for use in atmospheric models

S. K. Akagi[1], R. J. Yokelson[1], C. Wiedinmyer[2], M. J. Alvarado[3], J. S. Reid[4], T. Karl[2], J. D. Crounse[5], and P. O. Wennberg[6]

Atmos. Chem. Phys., 11, 4039–4072, 2011
www.atmos-chem-phys.net/11/4039/2011/
doi:10.5194/acp-11-4039-2011

Published 2011
May 2013 Update at:
http://bai.acd.ucar.edu/Data/fire/

Fire Emissions

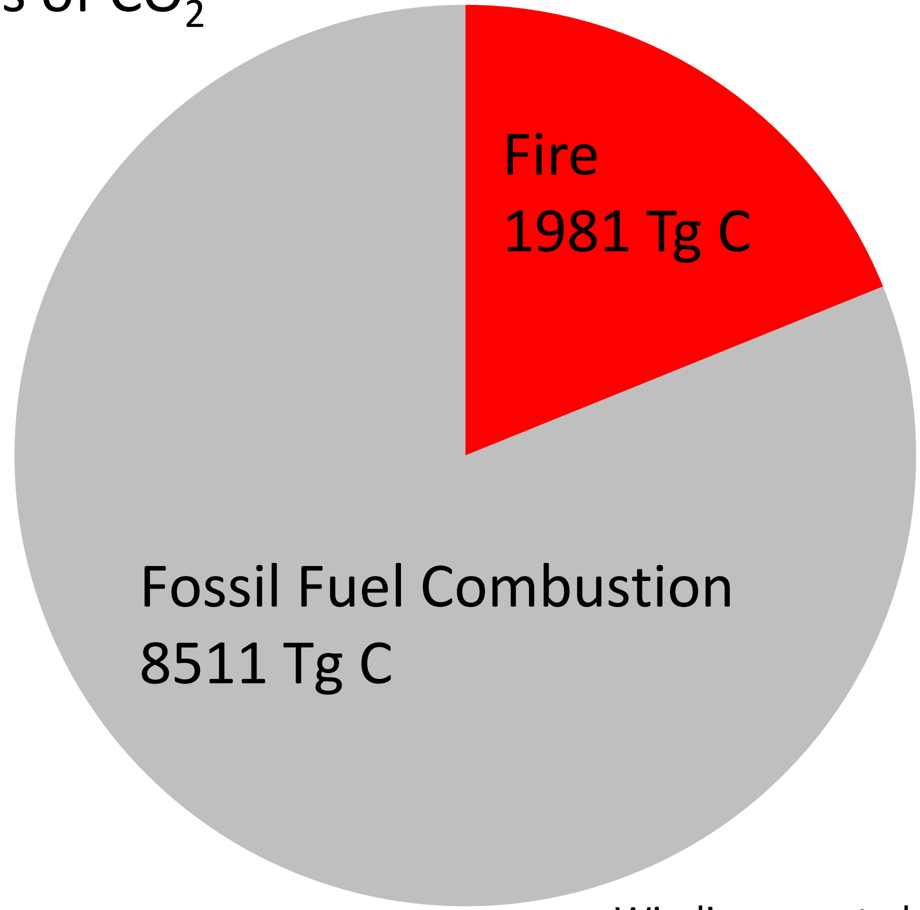
- Not just one thing emitted
- Many different compounds
- Some hazardous air pollutants
- Numerous impacts on air quality and climate

Carbon Dioxide (CO_2)	2-Methyl-1-Pentene (C_6H_{12})	Nitric Oxide (NO)
Methane (CH_4)	*n*-Hexane (C_6H_{14})	Nitrogen Dioxide (NO_2)
Hydrogen (H_2)	Heptane (C_7H_{16})	Nitrous Acid (HONO)
Carbon Monoxide (CO)	Benzene (C_6H_6)	Methyl Nitrate ($MeONO_2$)
Acetylene (C_2H_2)	Toluene ($C_6H_5CH_3$)	Ammonia (NH_3)
Ethylene (C_2H_4)	Xylenes	Hydrogen Cyanide (HCN)
Ethane (C_2H_6)	Ethylbenzene (C_8H_{10})	Acetonitrile (CH_3CN)
Propadiene (C_3H_4)	Methanol (CH_3OH)	Propenenitrile (C_3H_3N)
Propylene (C_3H_6)	Phenol (C_6H_5OH)	Propanenitrile (C_3H_5N)
Propane (C_3H_8)	Formaldehyde (HCHO)	Pyrrole (C_4H_5N)
1-Butene (C_4H_8)	Glycolaldehyde ($C_2H_4O_2$)	Carbonyl Sulfide (OCS)
1,3 Butadiene (C_4H_6)	Acetaldehyde (CH_3CHO)	Dimethyl Sulfide (C_2H_6S)
trans-2-Butene (C_4H_8)	Propanal (C_3H_6O)	Sulfur Dioxide (SO_2)
n-Butane (C_4H_{10})	Hexanal ($C_6H_{12}O$)	Methyl Bromide (CH_3Br)
i-Butane (C_4H_{10})	Acetone (C_3H_6O)	Methyl Iodide (CH_3I)
trans-2-Pentene (C_5H_{10})	Methacrolein (C_4H_6O)	Trichloromethane ($CHCl_3$)
cis-2-Pentene (C_5H_{10})	Crotonaldehyde (C_4H_6O)	OC
n-Pentane (C_5H_{12})	Methyl Vinyl Ketone (C_4H_6O)	BC
i-Pentane (C_5H_{12})	3-Pentanone ($C_5H_{10}O$)	Total PM
3-Methyl-1-Butene (C_5H_{10})	Furan (C_4H_4O)	Total Particulate Carbon
Isoprene (C_5H_8)	Formic Acid (HCOOH)	$PM_{2.5}$
Cyclopentane (C_5H_{10})	Acetic Acid (CH_3COOH)	PM_{10}

Akagi et al., *Atmos. Chem. & Phys.*, 2011

Fire Emissions

Annual (2005-2009)
Global Emissions of CO_2

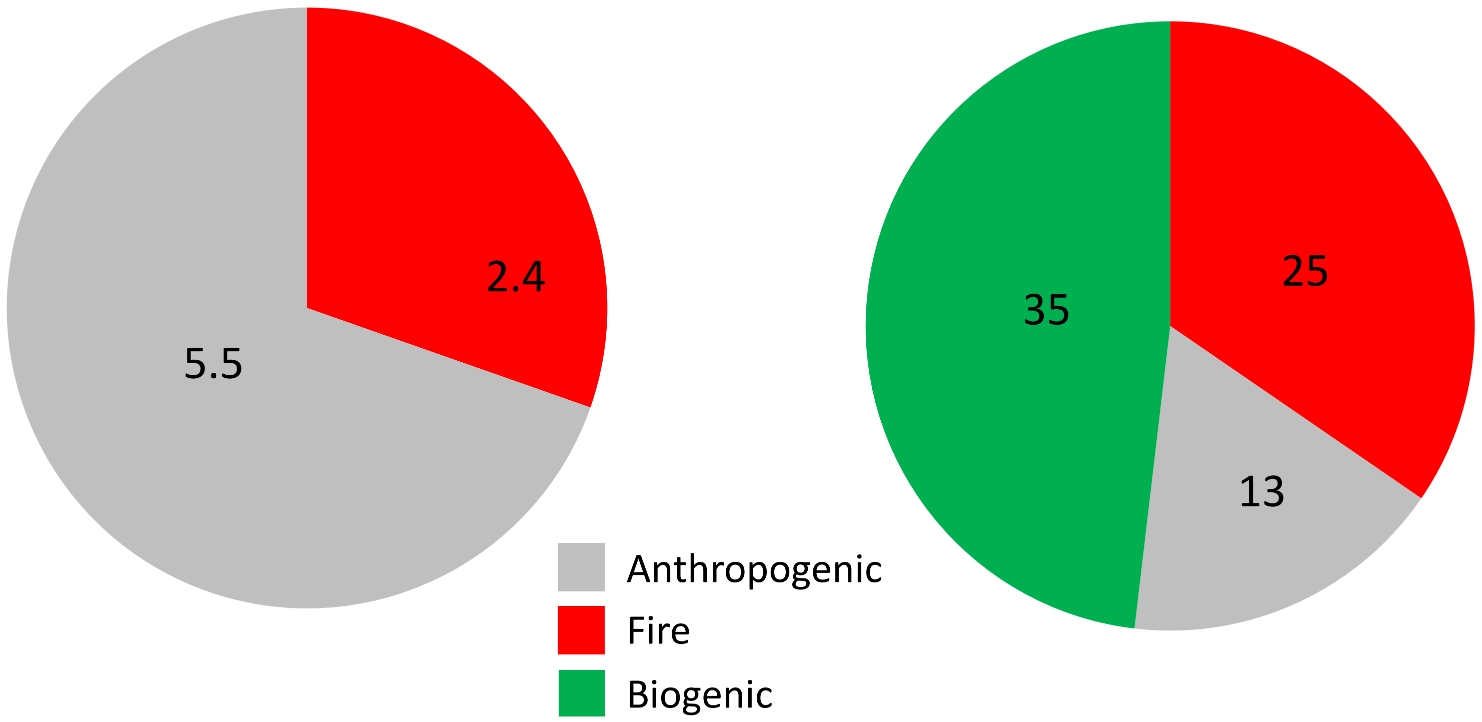


Wiedinmyer et al., *GMD*, 2011
http://cdiac.ornl.gov/trends/emis/glo.html

Fire Emissions

Particulate Black Carbon

Particulate Organic Carbon



$PM_{2.5}$ Emissions

45 Tg from Fire

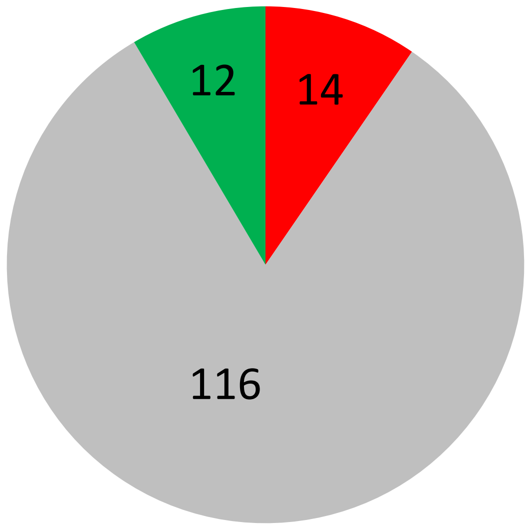
34 Tg from Anthropogenic

Wiedinmyer et al., *GMD,* 2011

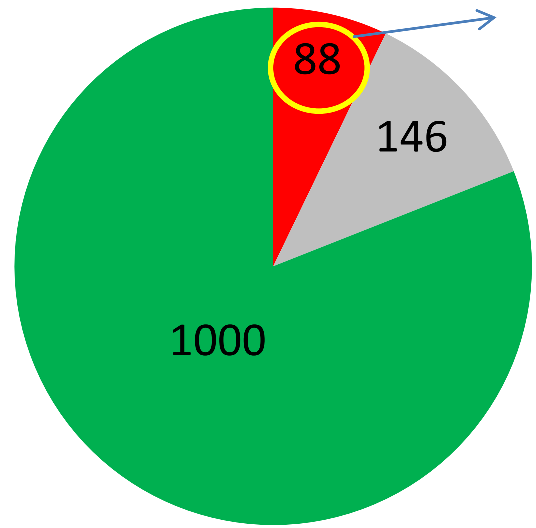
Andreae and Rosenfeld, *Earth Science Reviews*, 2008

Fire Emissions

Nitrogen Oxides

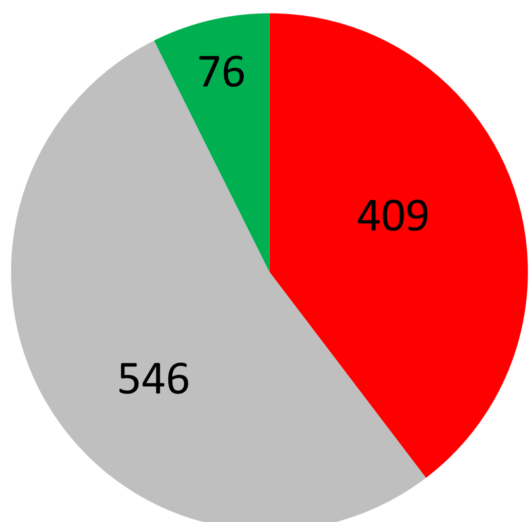


Non-Methane Organic Compounds



30% - 70% of NMOC were un-identified (Yokelson et al., 2013)

Anthropogenic
Fire
Biogenic



Carbon Monoxide

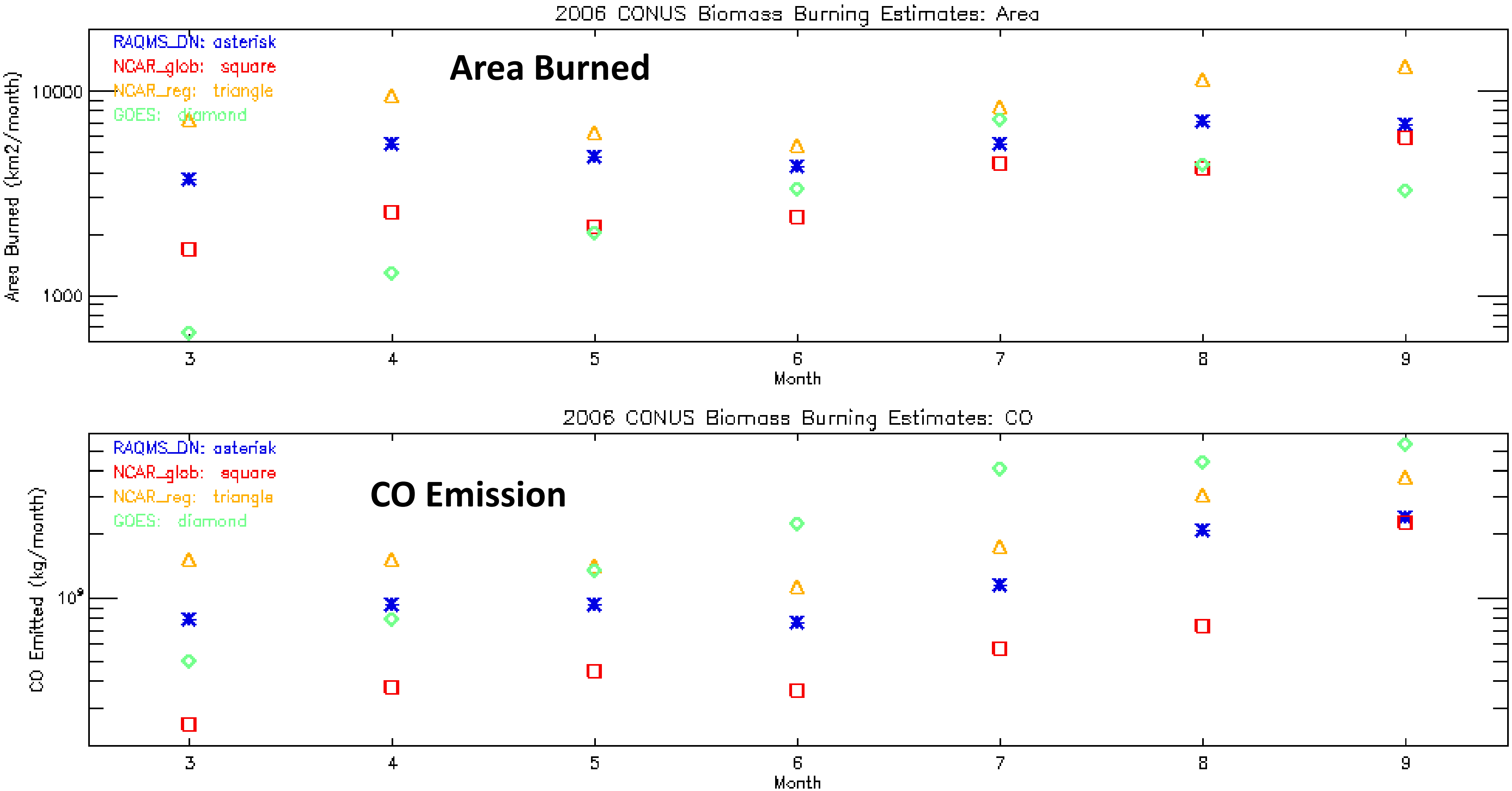
Wiedinmyer et al. *GMD* 2011
EDGARFT2000
Yan et al, GBC, 2005
Guenther et al., 1995; 2006; pers. comm.
Andreae and Merlet, GBC, 2001

Fire Emissions Inventories

- Many emission models exist
  - global/regional
  - Different drivers of activity

- Differences in emissions can be substantial

Uncertainties in Emission Estimates



Al-Saadi *Journal of Applied Remote Sensing* , 2008, DOI: 10.1117/1.2948785.

Take home messages….

- High quality emissions information is critical to understand the atmosphere and make good decisions about how to manage it
- Bottom-up inventories are integral to these efforts, but there are challenges associated with these complex datasets
- There are significant disagreements between different global and regional bottom-up inventories
- Top-down approaches based on atmospheric measurements and models provide objective information about emissions
- Combining top-down and bottom-up approaches improves process-based understanding of key emissions sources
- A broad community of scientists, regulators, and policymakers must collaborate to develop high quality emissions information

Thank you for your attention

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