Measurements and Emissions

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large variety of measurement techniques for atmospheric trace are available:

- **Gas chromatography (GC, universal technique, in-situ)**
- **Optical spectroscopy (universal technique, in-situ, remote sensing)**
- **Chemiluminescence (e.g. for the detection of NO or O3)**
- **Photoacoustic detection**
- **Electrochemical techniques**

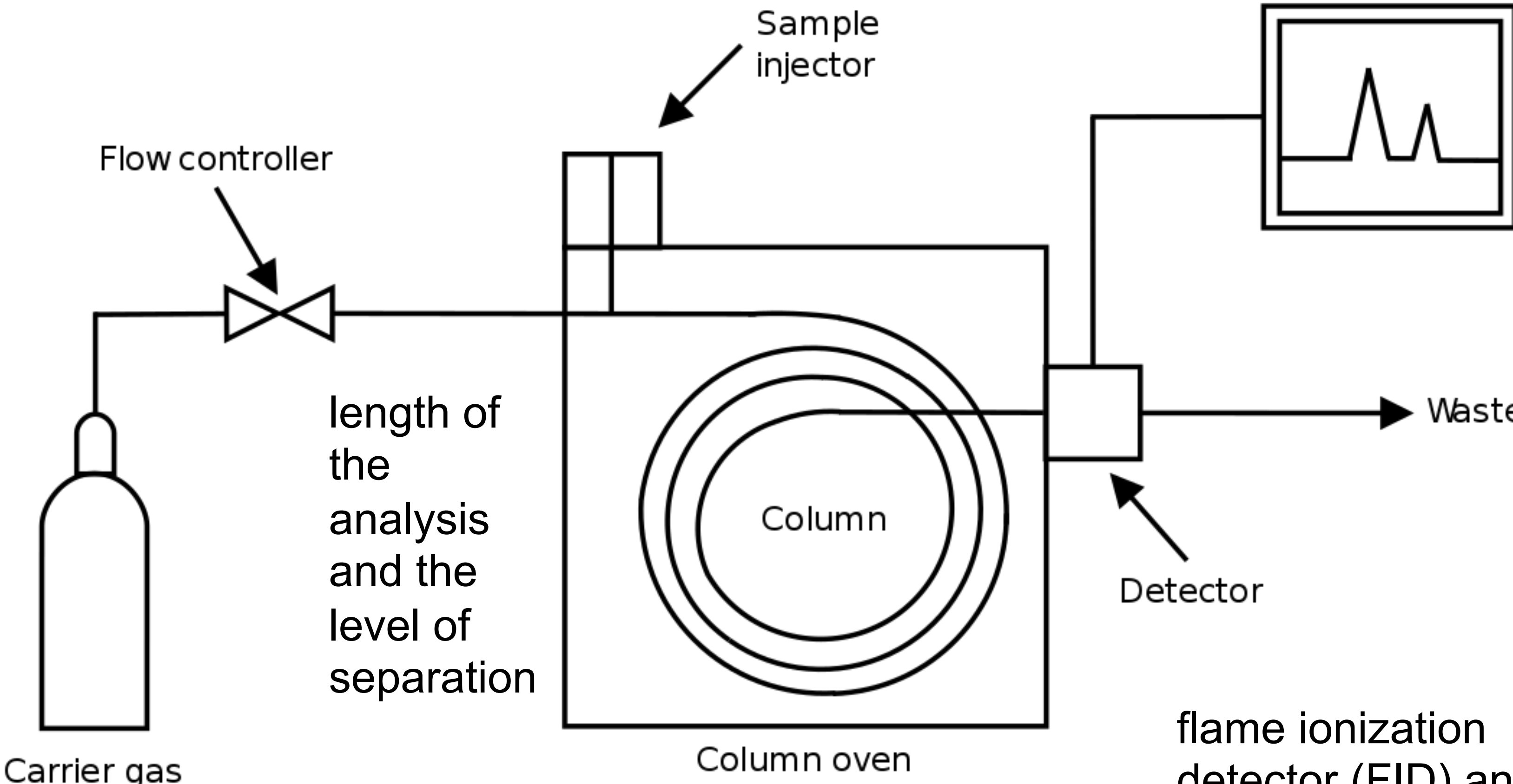
Spectroscopic techniques are a powerful variety.

These techniques are highly sensitive, very specific, universally usable, give absolute results, and have the potential for remote sensing.

Gas chromatography

Retention time

separating the different components of a mixture

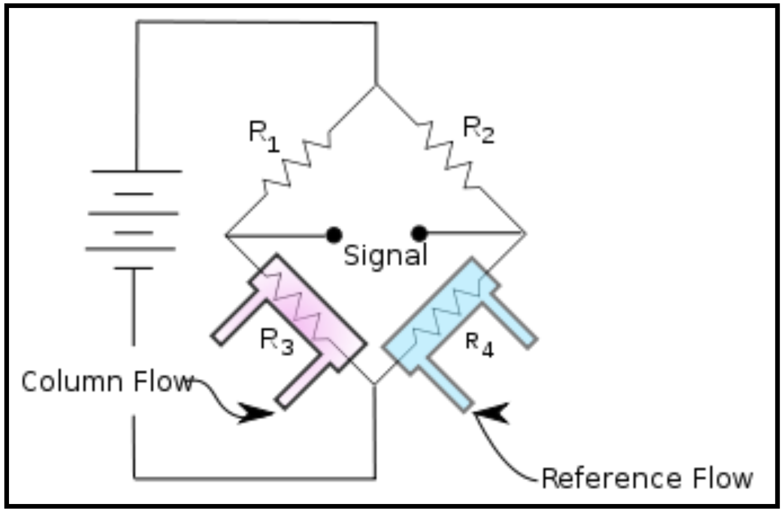


length of the analysis and the level of separation

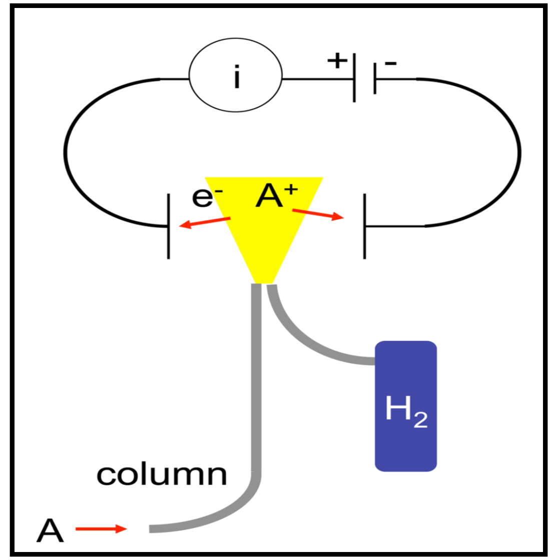
Mobile is a carrier gas,
inert gas such as helium
unreactive gas such as nitrogen

stationary phase
microscopic layer of polymer on an inert solid support, inside a piece of metal tubing called a column

flame ionization detector (FID) and the thermal conductivity detector (TCD) thermal conductivity detector (TCD)

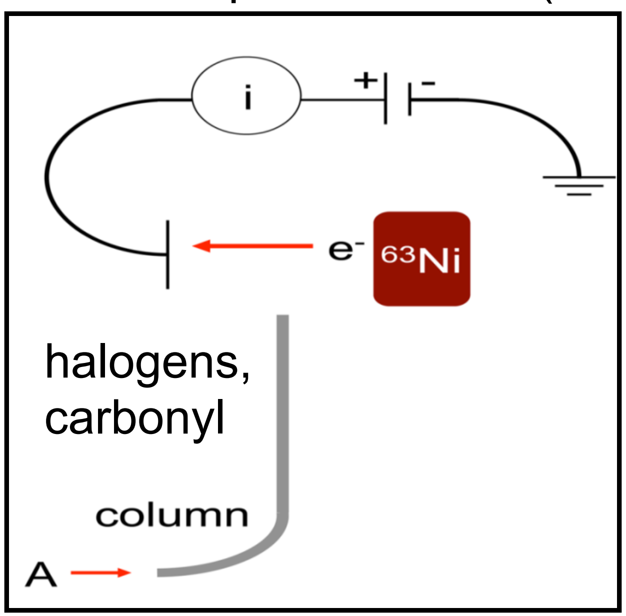


flame ionization detector (FID)



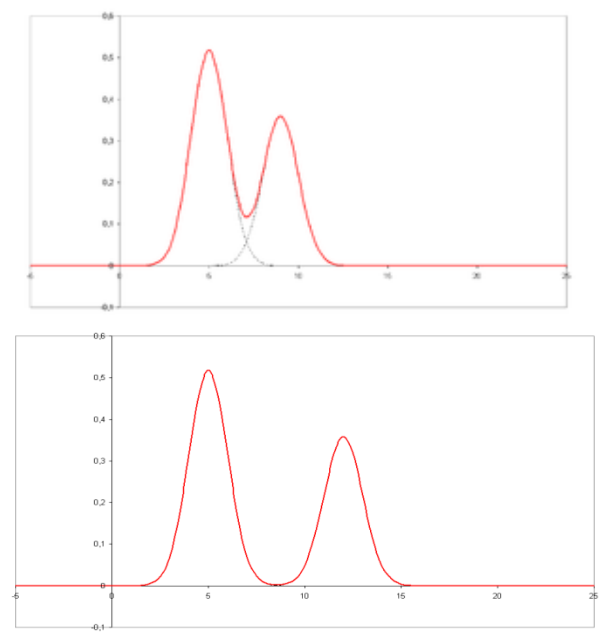
Mostly for Hydrocarbons

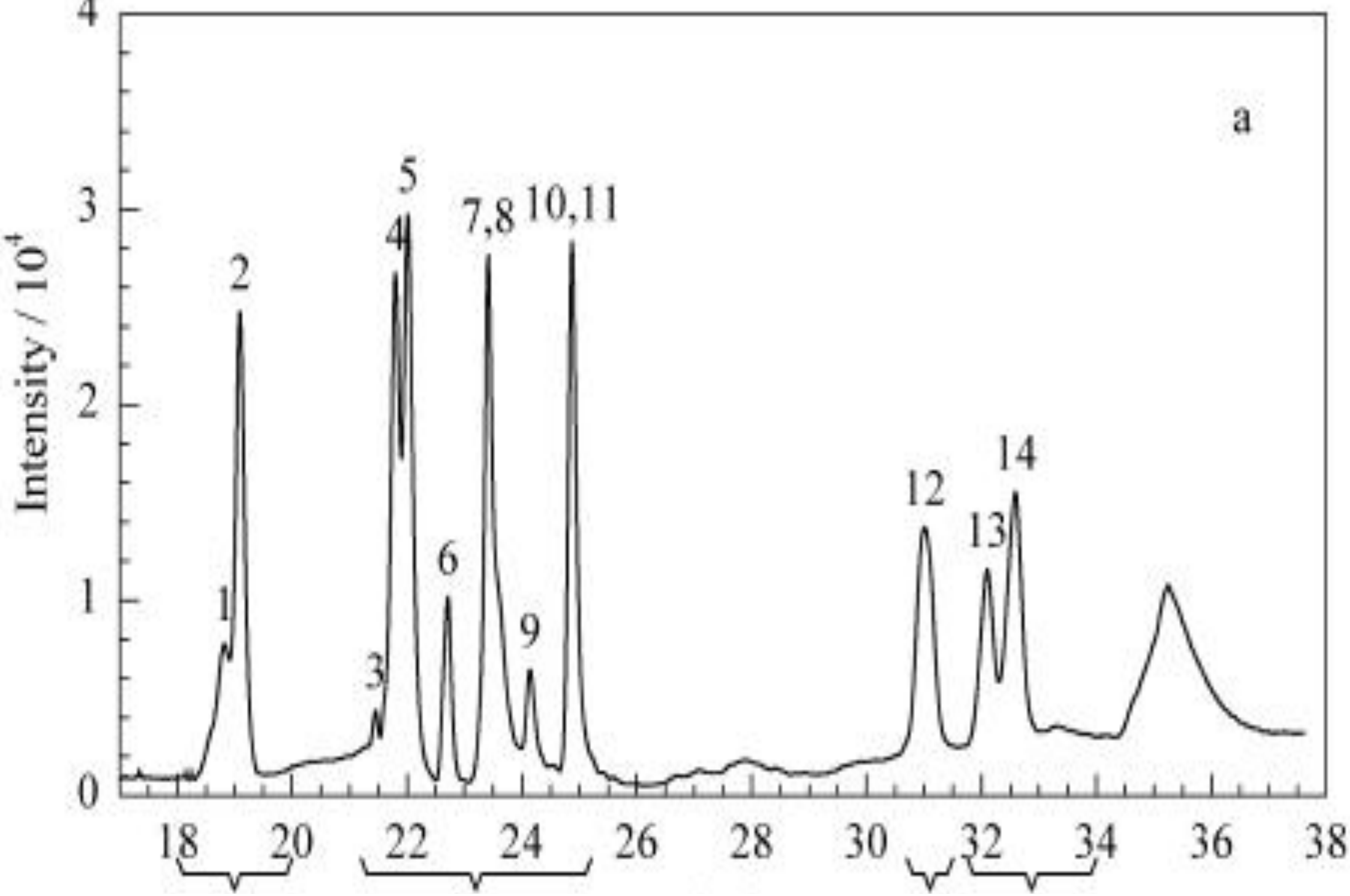
Electron capture detector (ECD)



Makeup gas N2 generates background current

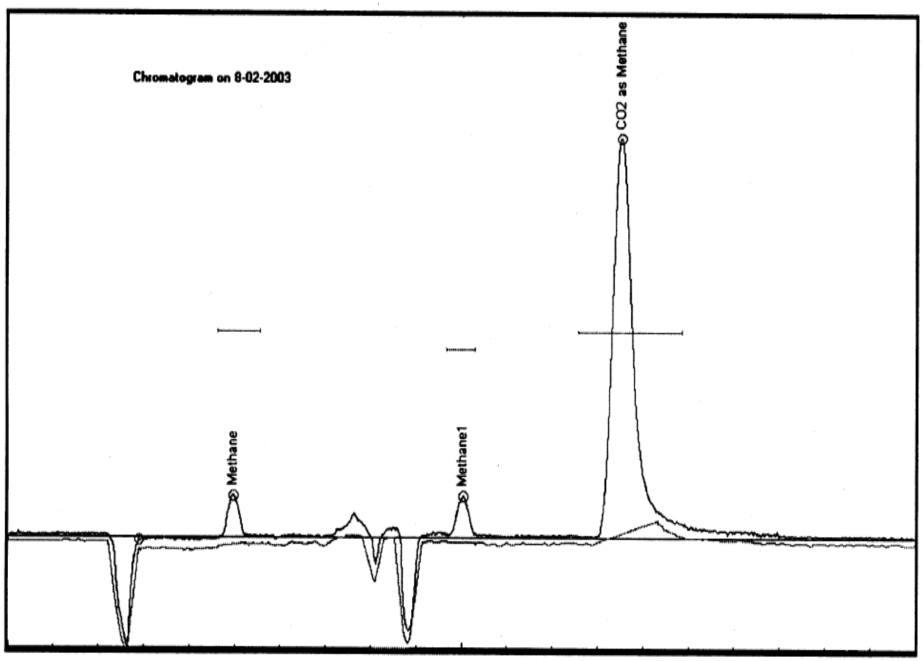
Chromatogram

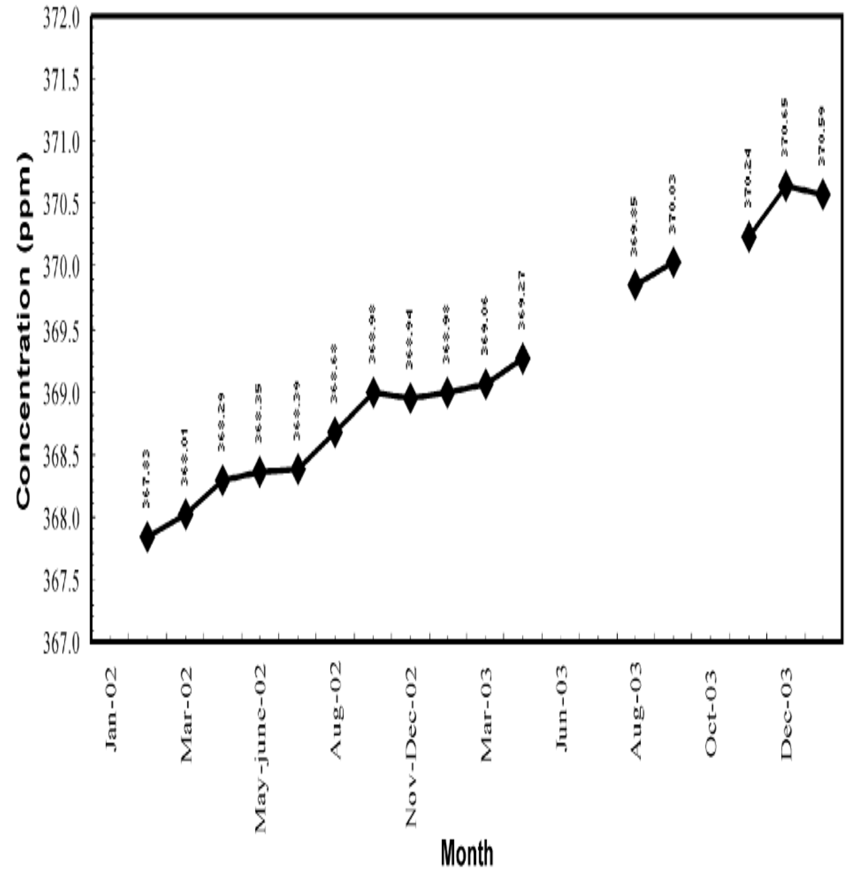




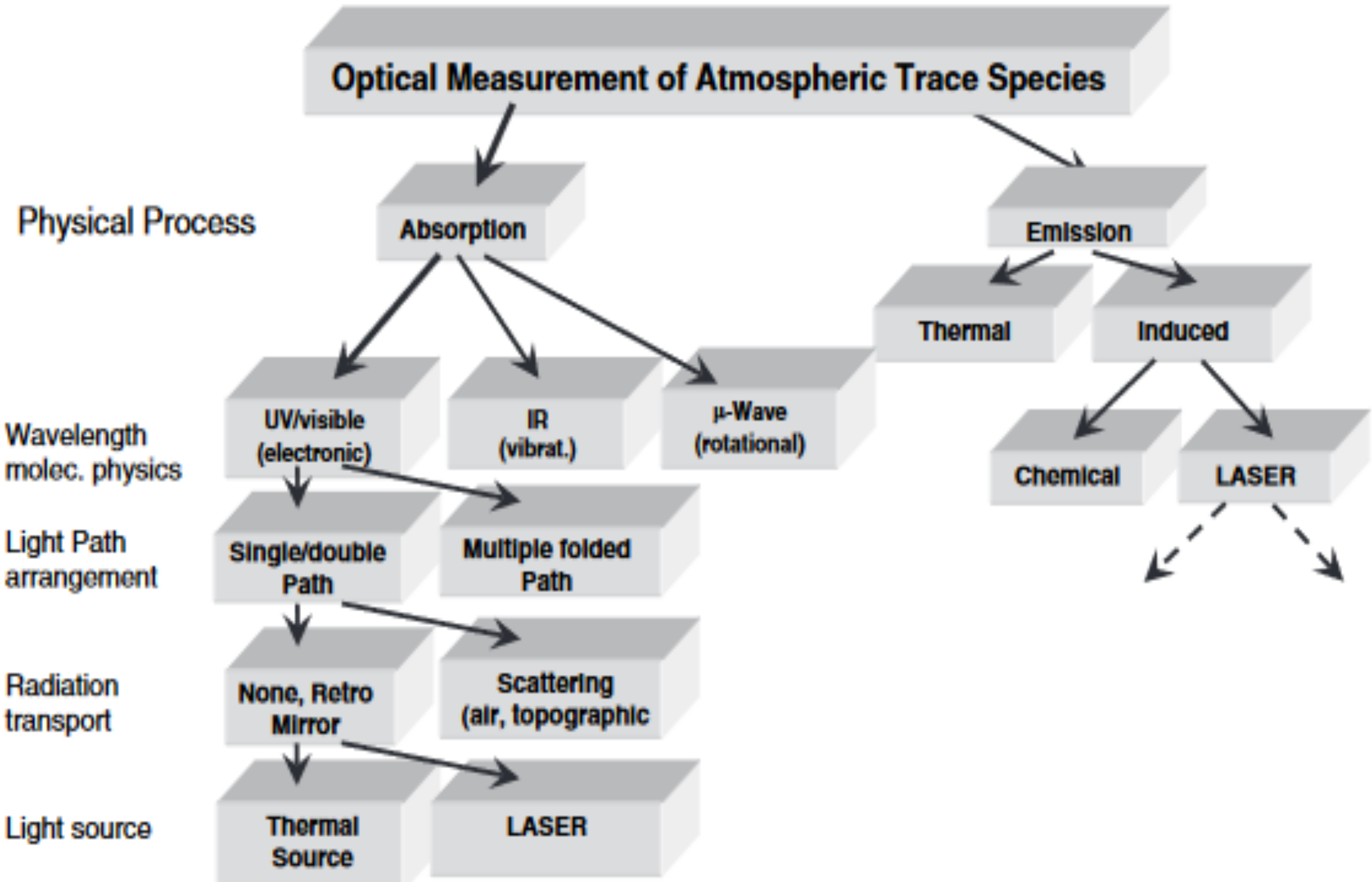
a
Intensity / 10^4
0
1
2
3
4
18
20
22
24
26
28
30
32
34
36
38
1
2
3
4
5
6
7,8
9
10,11
12
13
14

Typical Methane CO and CO_2 Chromatogram observed at Antarctica





Area of peak gives information about the concentration



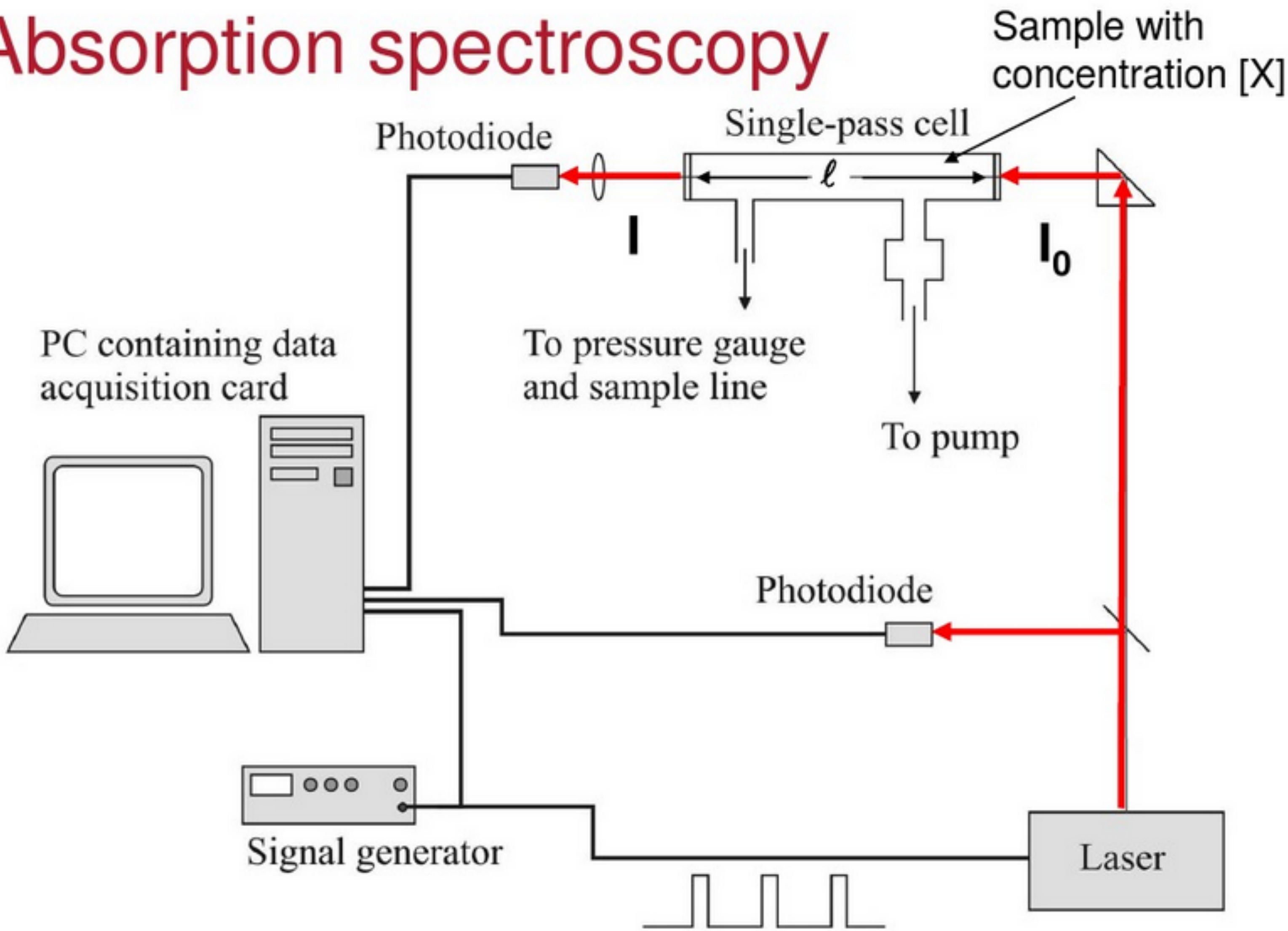
Optical Measurement of Atmospheric Trace Species
Physical Process
Absorption
Emission
Thermal
Induced
Wavelength molec. physics
UV/visible (electronic)
IR (vibrat.)
μ-Wave (rotational)
Chemical
LASER
Light Path arrangement
Single/double Path
Multiple folded Path
Radiation transport
None, Retro Mirror
Scattering (air, topographic
Light source
Thermal Source
LASER

The important technical criteria of spectroscopic instruments

(1) are the wave-length region used
(2) the physical principle (i.e. absorption or emission spectroscopy),
(3) the arrangement of the light path (path in the open atmosphere or enclosed – frequently folded – path)
(4) or the type of light source used.

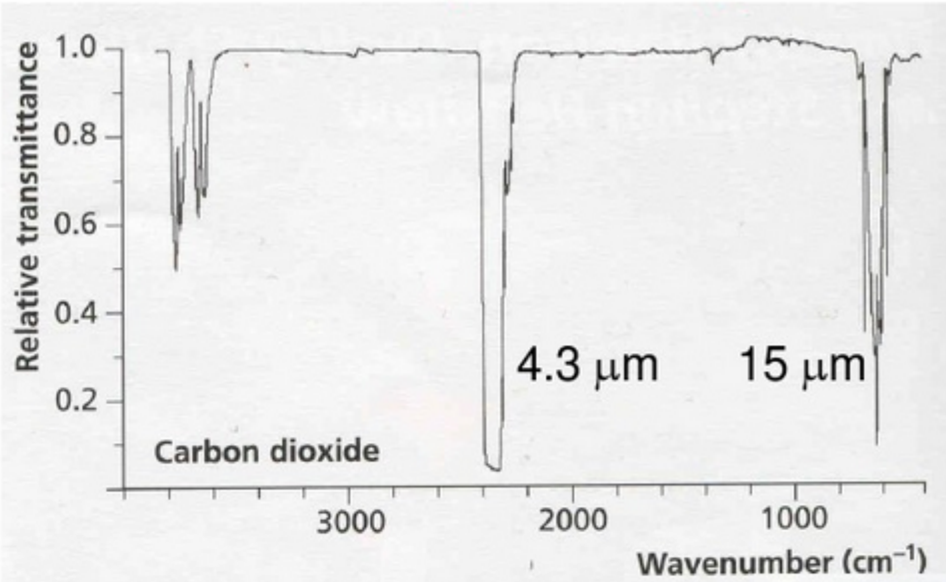
- Tunable Diode Laser Spectroscopy (TDLS)
- Cavity Enhanced Absorption Spectroscopy (CESA)
- Photo Acoustic Spectroscopy (PAS)
- Light Detection And Ranging (LIDAR)
- Differential Absorption LIDAR (DIAL)
- Laser-Induced Fluorescence (LIF)
- Cavity-Ringdown Spectroscopy (CRD)
- Cavity Attenuated Phase Shift (CAPS)
- Differential Optical Absorption Spectroscopy (DOAS)

Absorption spectroscopy



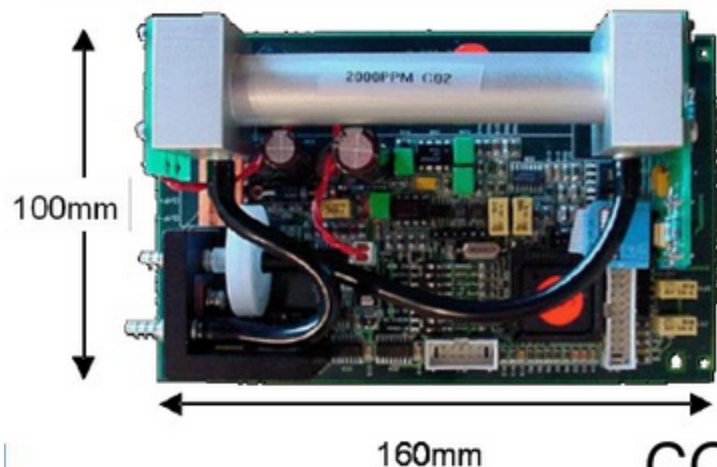
Beer-Lambert Law: $I = I_0 \exp\{-\sigma [X] \ell\}$

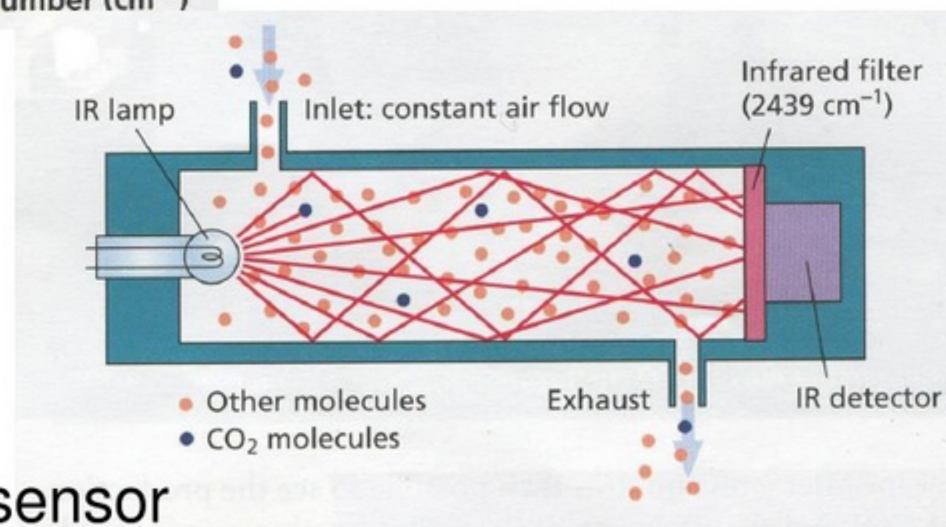
1. Absorption spectroscopy



Bending Vibration
$\lambda = 15\ \mu m$

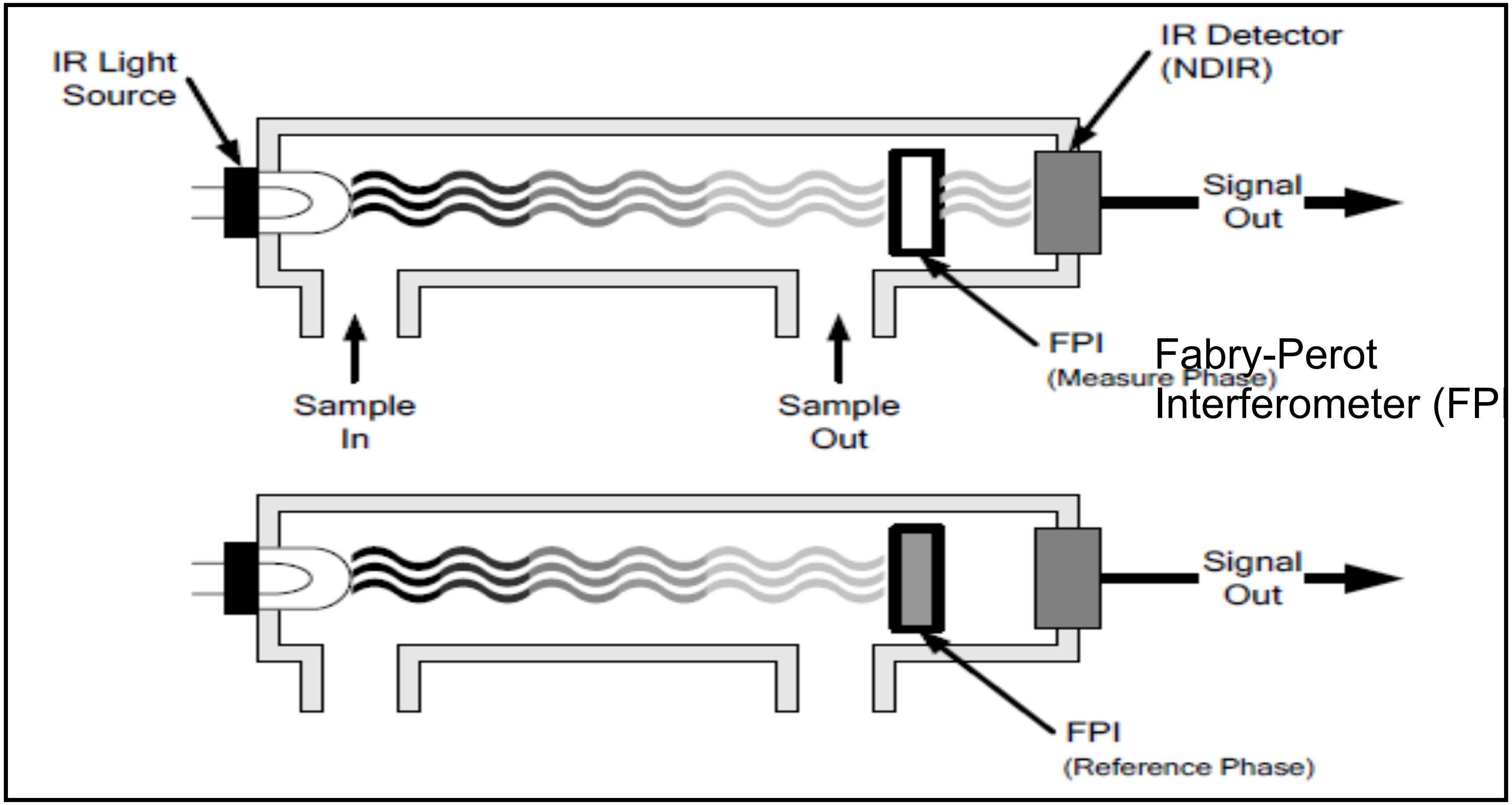
Asymmetric Stretch
$\lambda = 4.3\ \mu m$



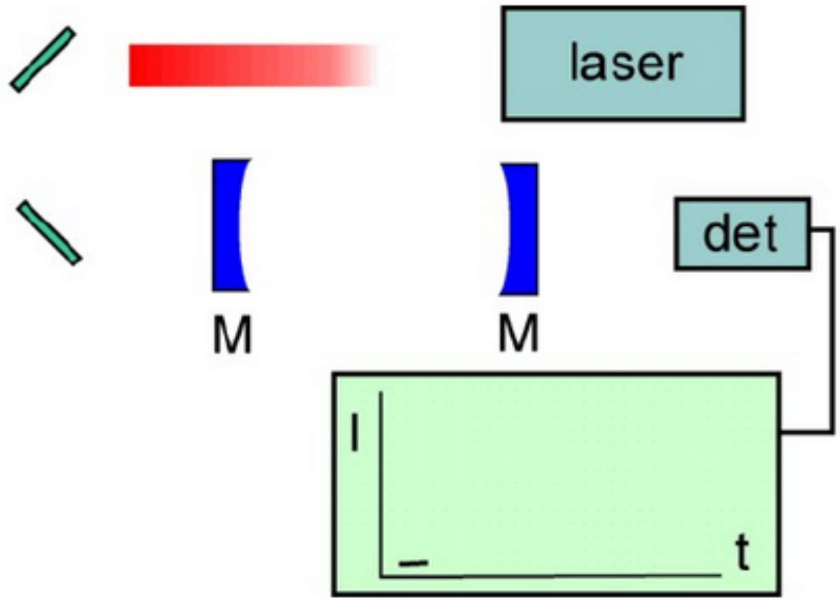


CO_2 sensor

Infrared CO_2 analyzer

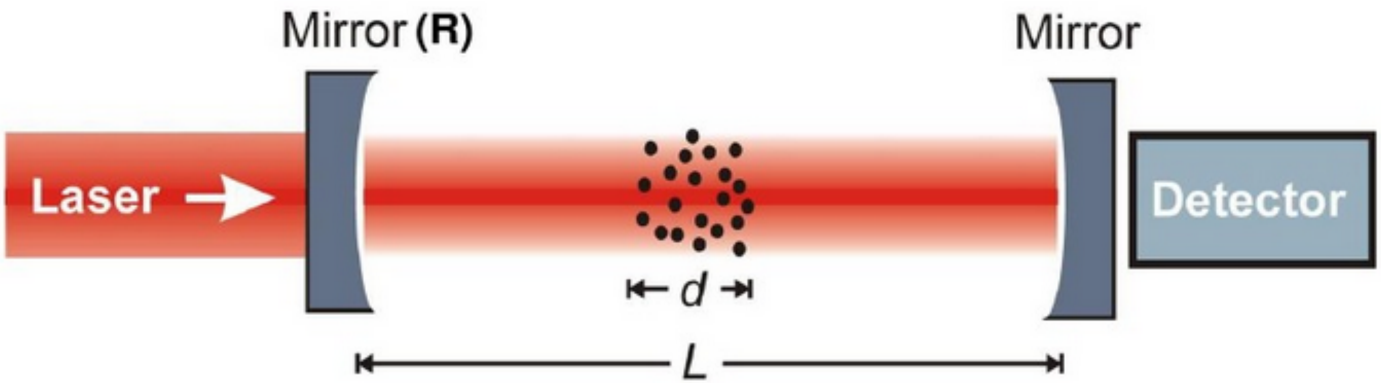


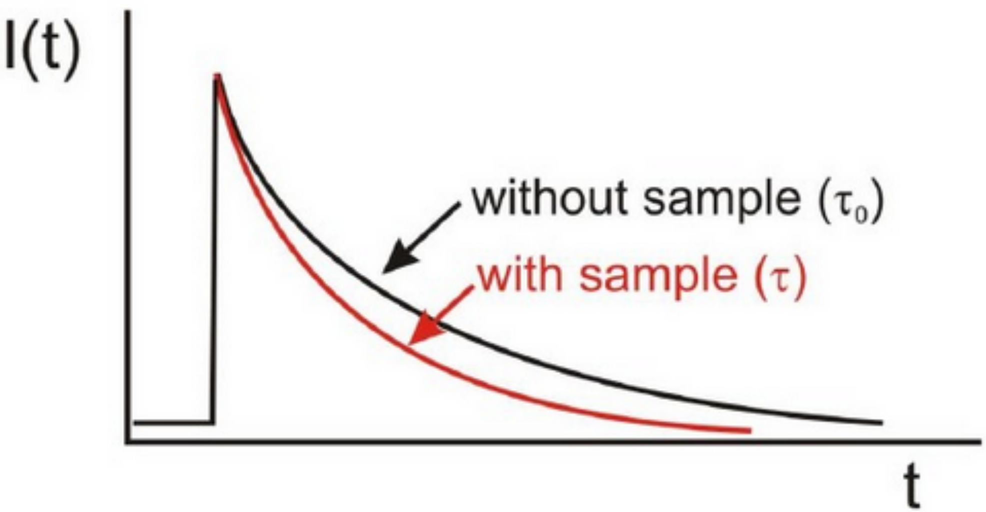
2. Cavity ring-down spectroscopy



- Trap a pulse of light between two high-reflectivity mirrors
- Place the gas sample between the mirrors (in the cavity)
- Measure the **lifetime** of the light pulse in the cavity – depends on concentration of sample

Cavity ring-down spectroscopy





For an empty cavity:

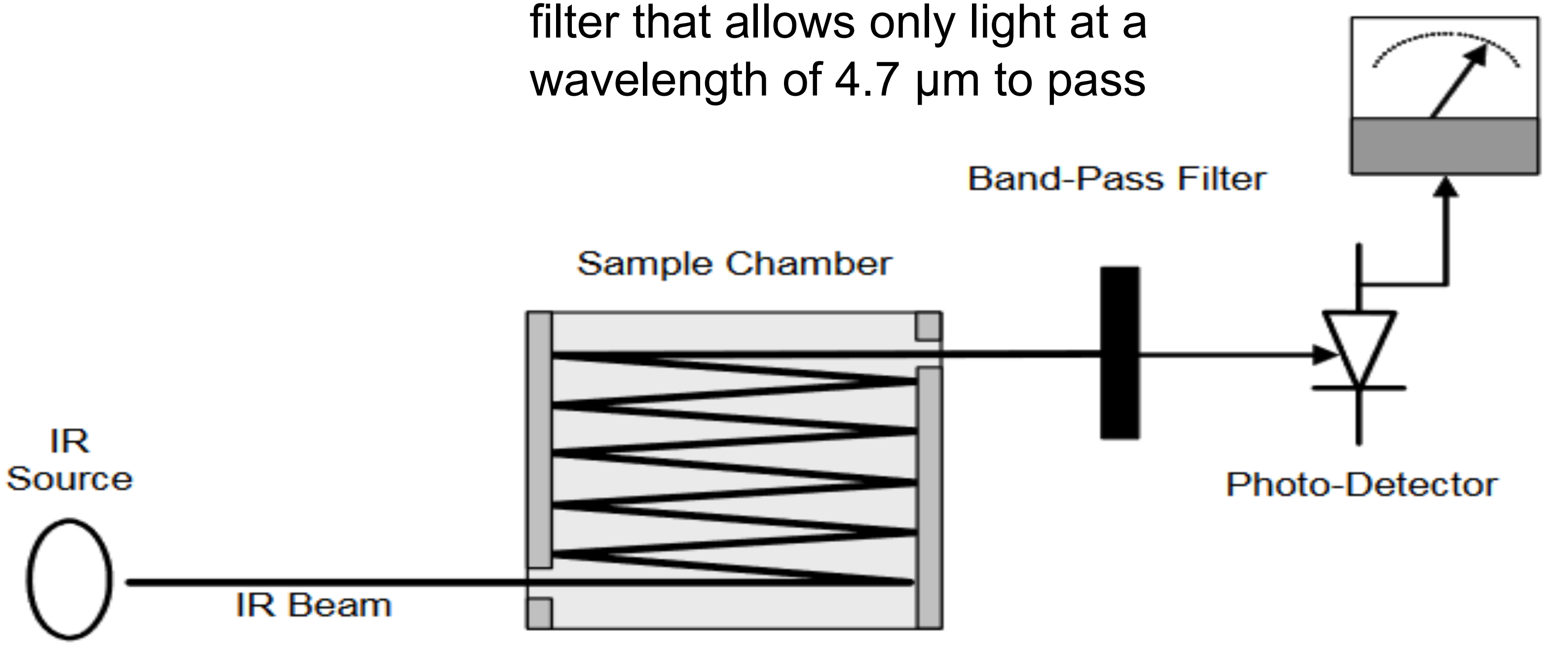
$$\tau_0 = \frac{L}{c(1-R)}$$

With an absorber:

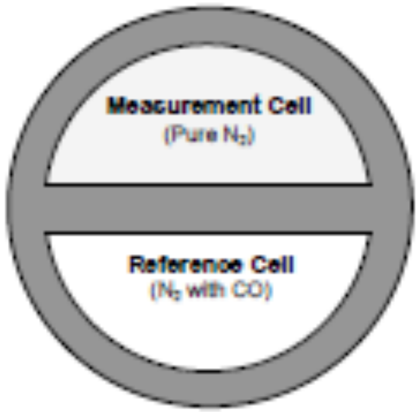
$$\frac{1}{\tau} - \frac{1}{\tau_0} = c[X]\sigma\frac{d}{L}$$

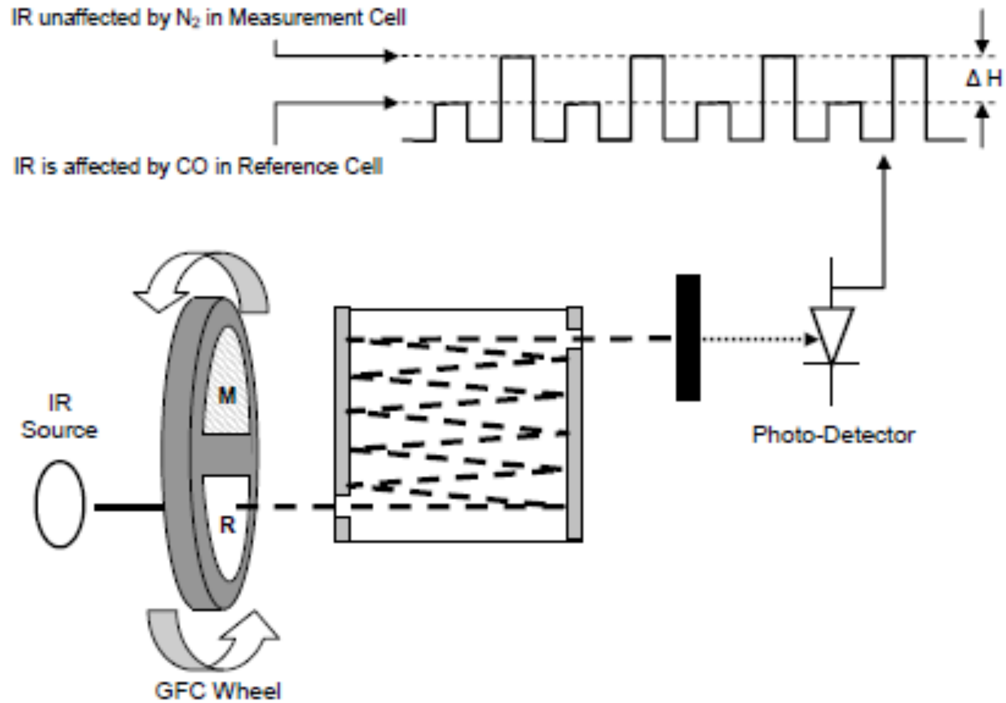
Measurement of Carbon Monoxide

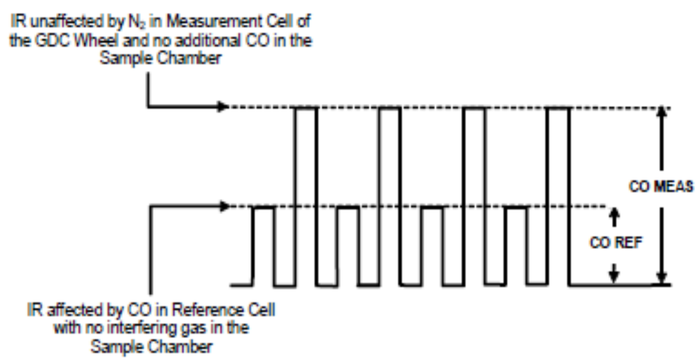
beam shines through a band-pass filter that allows only light at a wavelength of 4.7 µm to pass

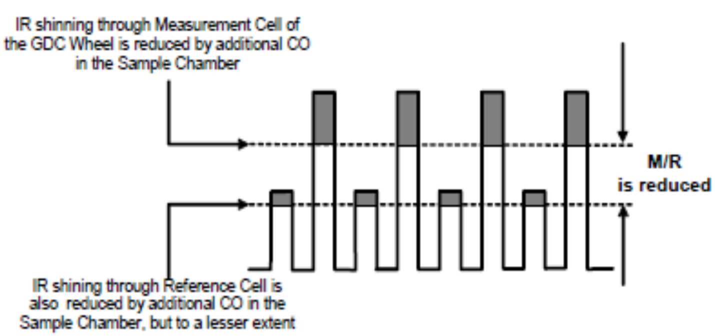


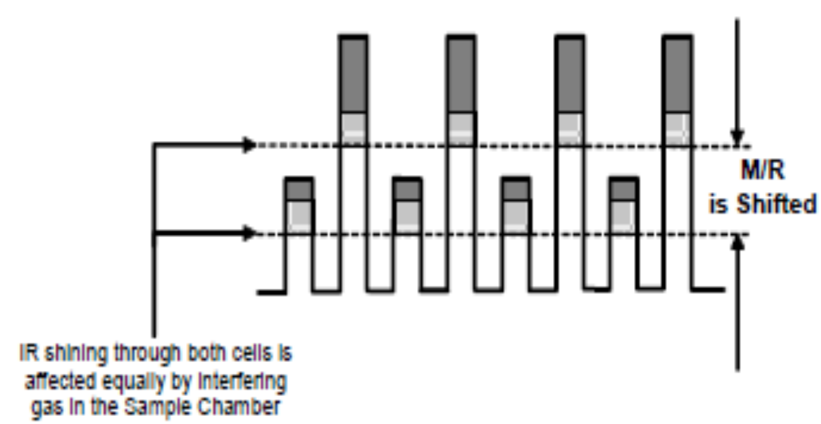
high-energy heated element to generate a beam of broad-band IR light


Measurement Cell
(Pure N_2)
Reference Cell
(N_2 with CO)

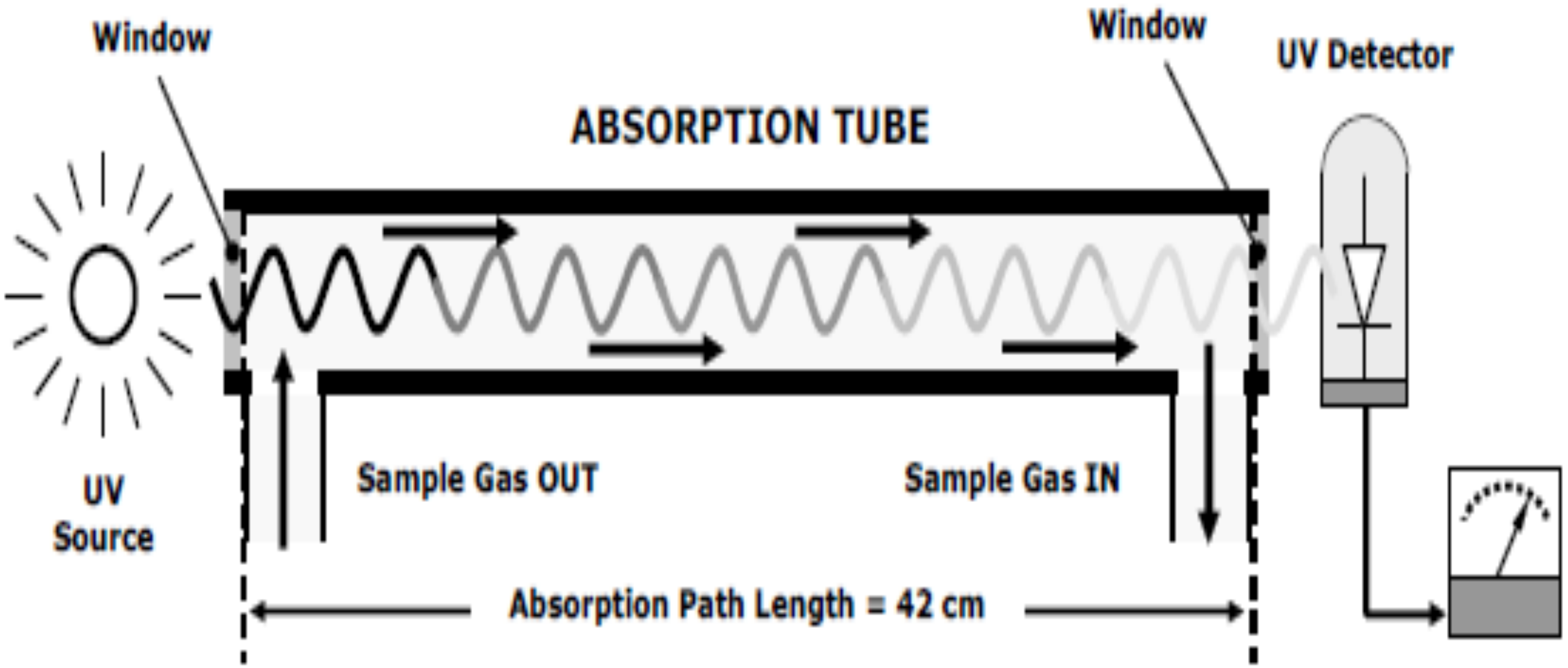

IR unaffected by N_2 in Measurement Cell
IR is affected by CO in Reference Cell
Δ H
IR Source
M
R
Photo-Detector
GFC Wheel


IR unaffected by N_2 in Measurement Cell of the GDC Wheel and no additional CO in the Sample Chamber
CO MEAS
CO REF
IR affected by CO in Reference Cell with no interfering gas in the Sample Chamber


IR shinning through Measurement Cell of the GDC Wheel is reduced by additional CO in the Sample Chamber
M/R
is reduced
IR shining through Reference Cell is also reduced by additional CO in the Sample Chamber, but to a lesser extent

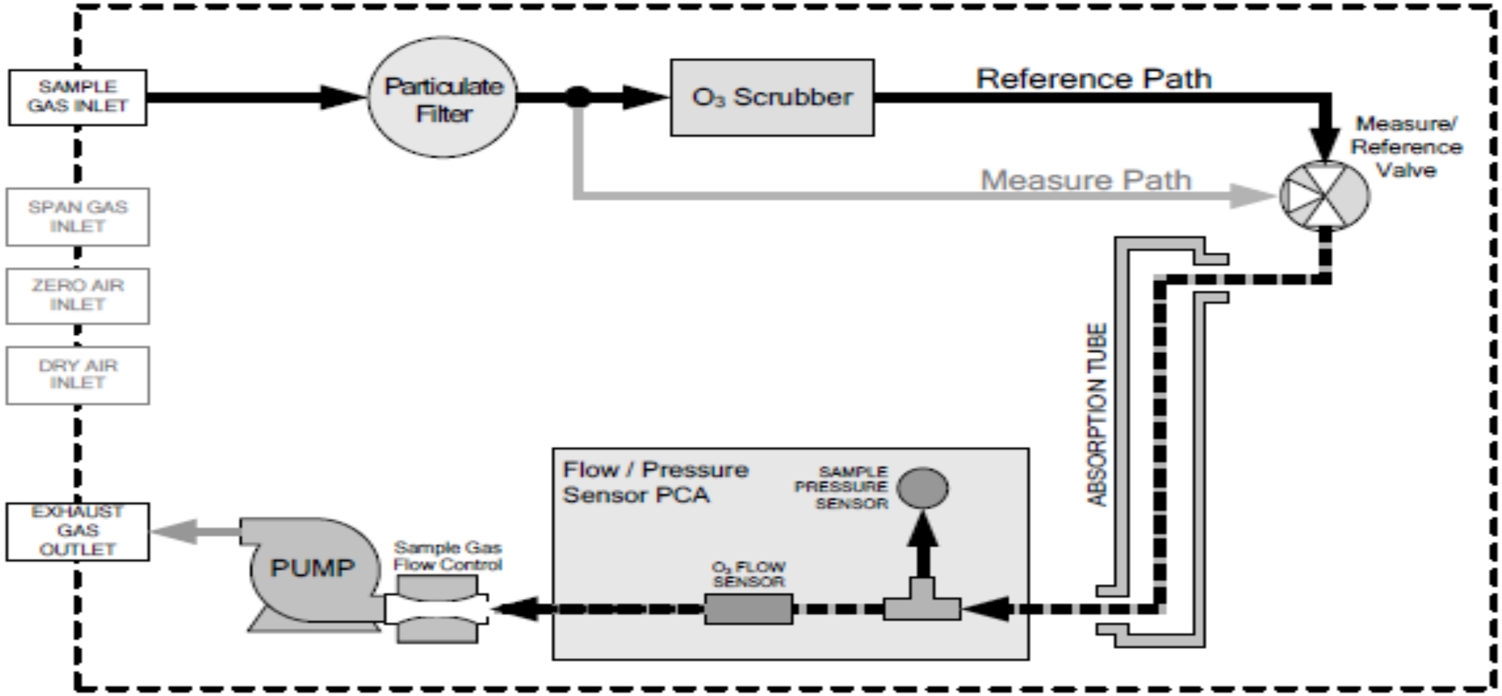

M/R
is Shifted
IR shining through both cells is affected equally by interfering gas in the Sample Chamber

Measurement of Ozone



uses a high energy, mercury vapor lamp to generate a beam of UV light 254nm

Measurement of Ozone



The Measurement / Reference Cycle consists of:

TIME INDEX	STATUS
0 seconds	Measure/Reference Valve Opens to the Measure Path.
0 – 2 seconds	Wait Period. Ensures that the Absorption tube has been adequately flushed of any previously present gasses.
2 – 3 seconds	Analyzer measures the average UV light intensity of O_3 bearing Sample Gas (I) during this period.
3 seconds	Measure/Reference Valve Opens to the Reference Path.
3 – 5 seconds	Wait Period. Ensures that the Absorption tube has been adequately flushed of O_3 bearing gas.
5 – 6 seconds	Analyzer measures the average UV light intensity of Non-O_3 bearing Sample Gas (I_0) during this period.
CYCLE REPEAT EVERY 6 SECONDS	

The Cavity Attenuated Phase Shift (CAPS) NO_2 monitor

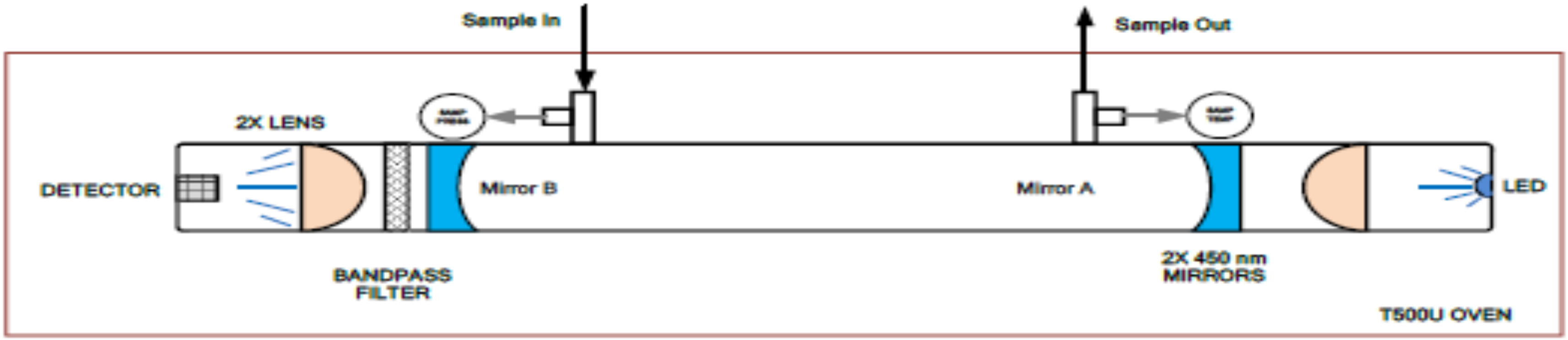
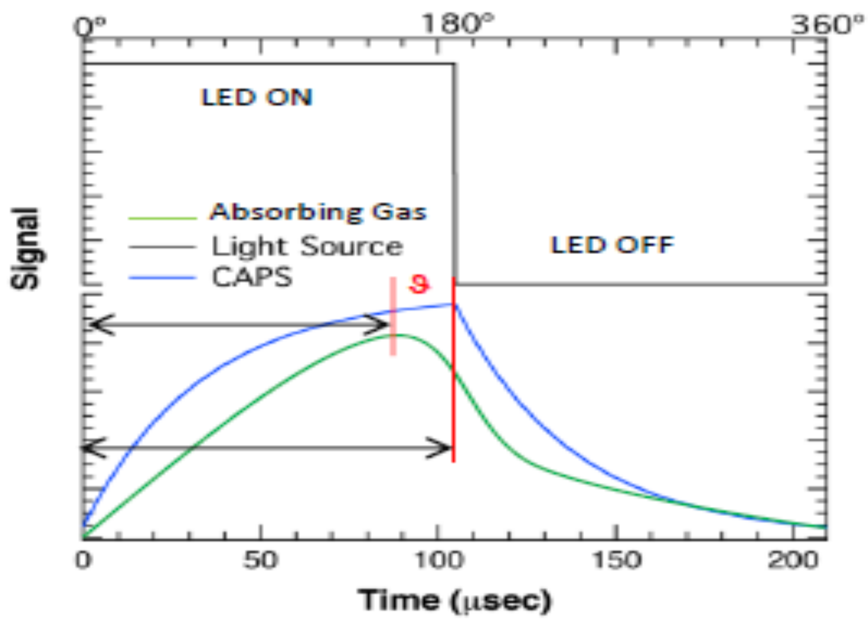
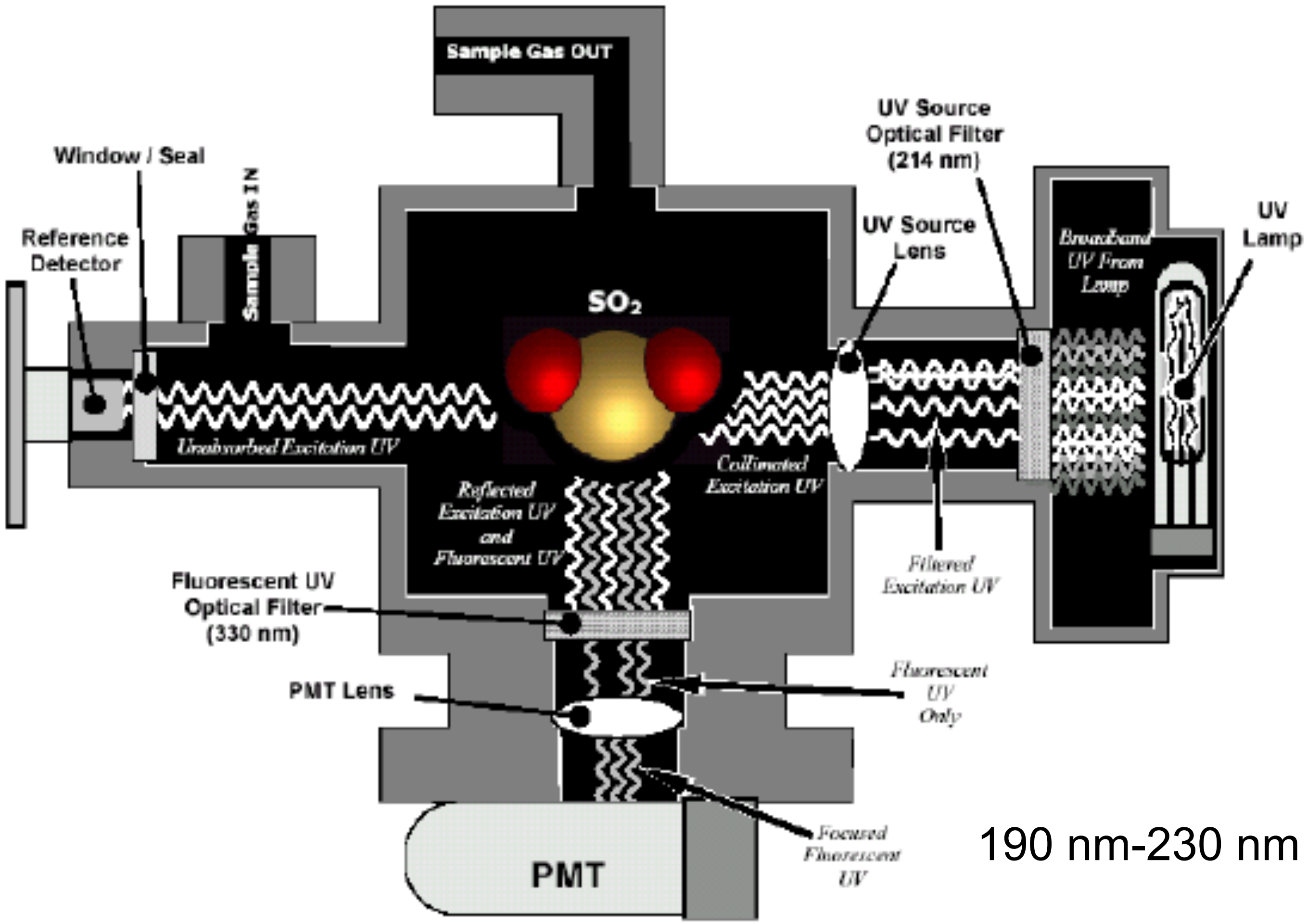


Figure 14-1: T500U Optical Absorption Cell

450 nm



UV Fluorescence SO_2 Analyzer



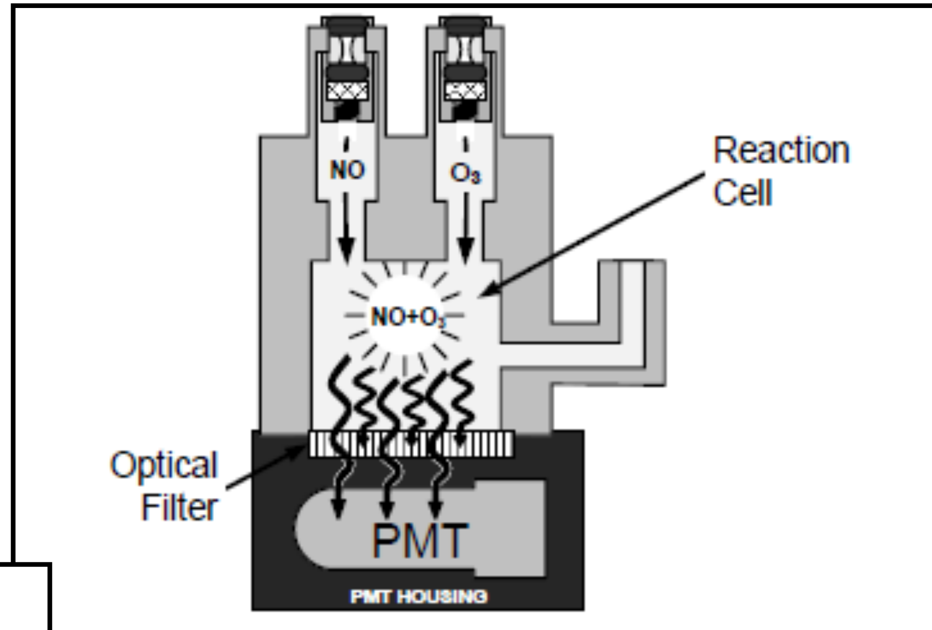
190 nm-230 nm

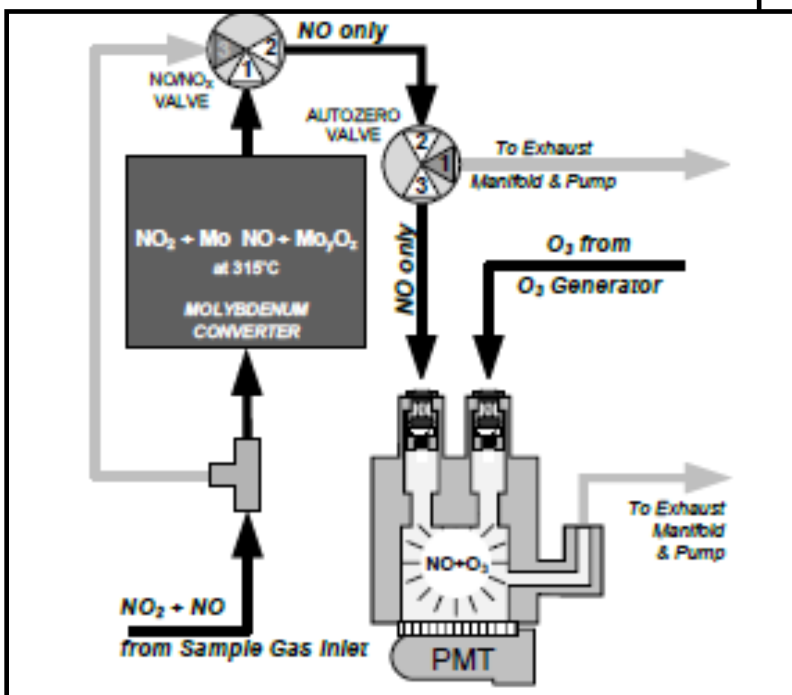
214 exication 330 emission

CHEMILUMINESCENCE NOx Analyzer

$$NO + O_3 \rightarrow NO_2^* + O_2$$

$$NO_2^* \rightarrow NO_2 + h\nu_{1200nm}$$

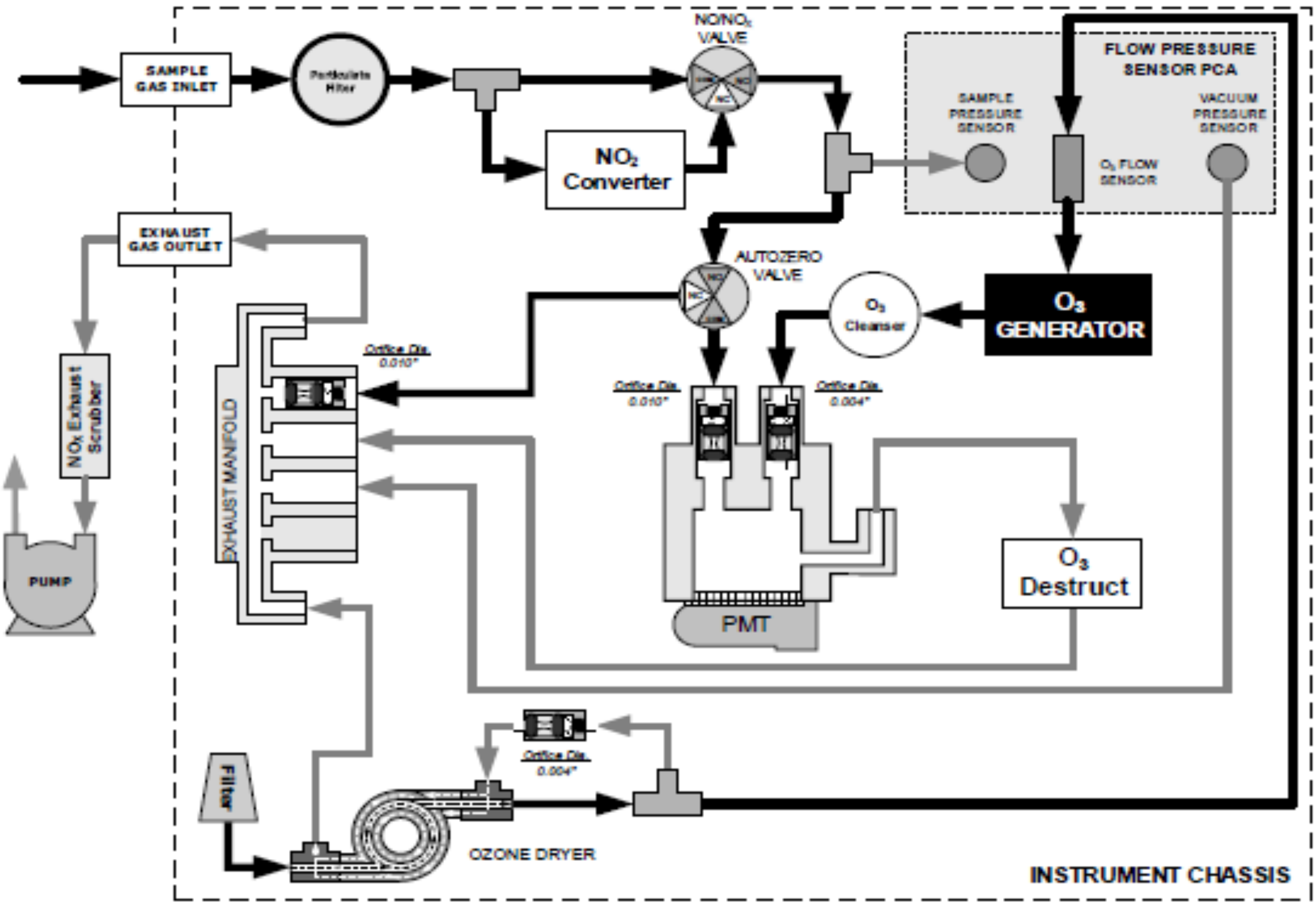




$$NO_2^* + M \rightarrow NO_2 + M$$

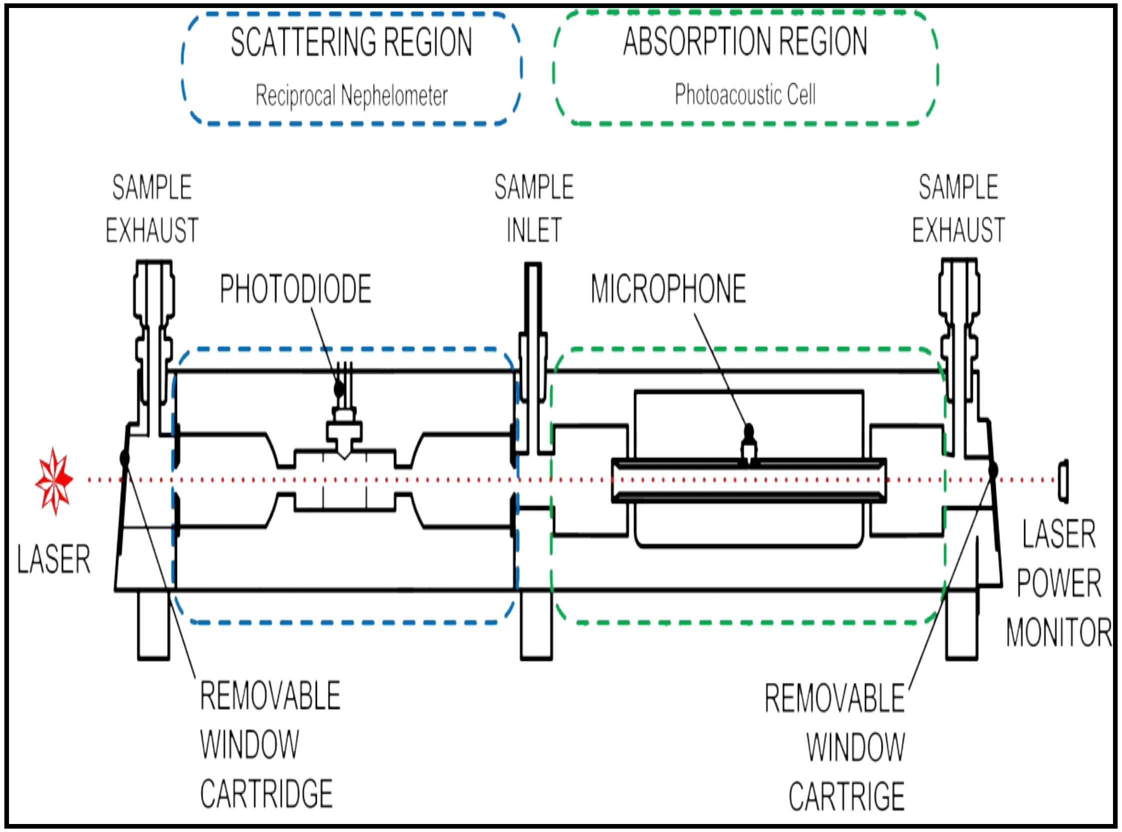
Less pressure (avoid collision)
increase ozone concentration

CHEMILUMINESCENCE NOx Analyzer



The Photoacoustic Extinctiometer (PAX)

simultaneously measure light scattering and absorption



Nephelometer to measure the light scattering.

The absorption through photoacoustic

Absorbing particles heat up and quickly transfer heat to the surrounding air. The periodic heating produces pressure waves that can be detected with a sensitive microphone.

Red (870 nm) – absorption is highly specific for black carbon (soot) particles; scattering best for large particles
Green (532 nm) – measures in the visual range, typically what the human eye observes
Blue (405 nm) – absorption correlates to the organic, or brown carbon content; efficient scattering for fine and ultrafine particles

Microwave Radiometer

Tropospheric Profiling
of temperature, humidity and liquid water

Water Vapour Monitoring
e.g. at astronomical sites

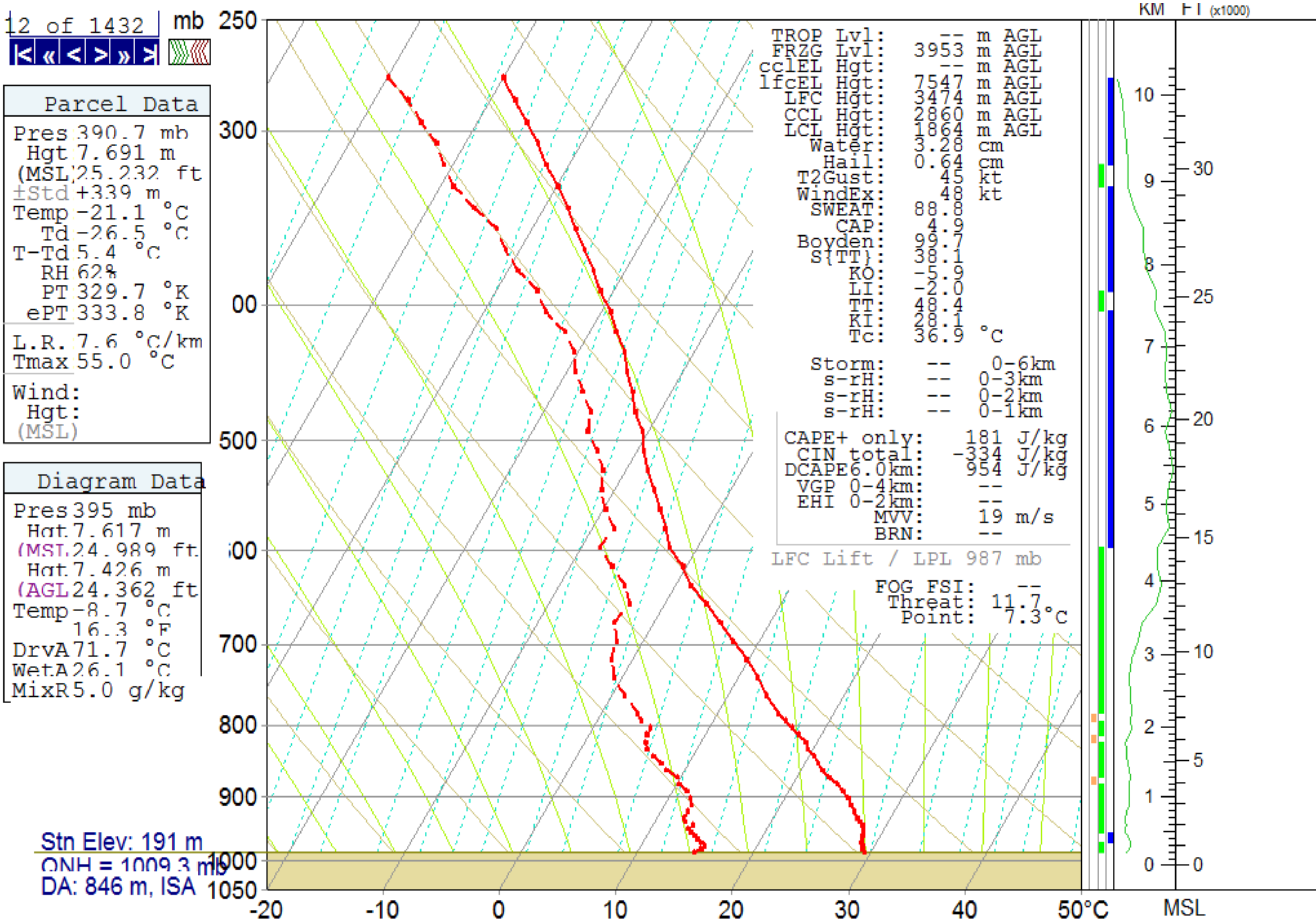
Now-Casting
- atmospheric stability
- severe weather

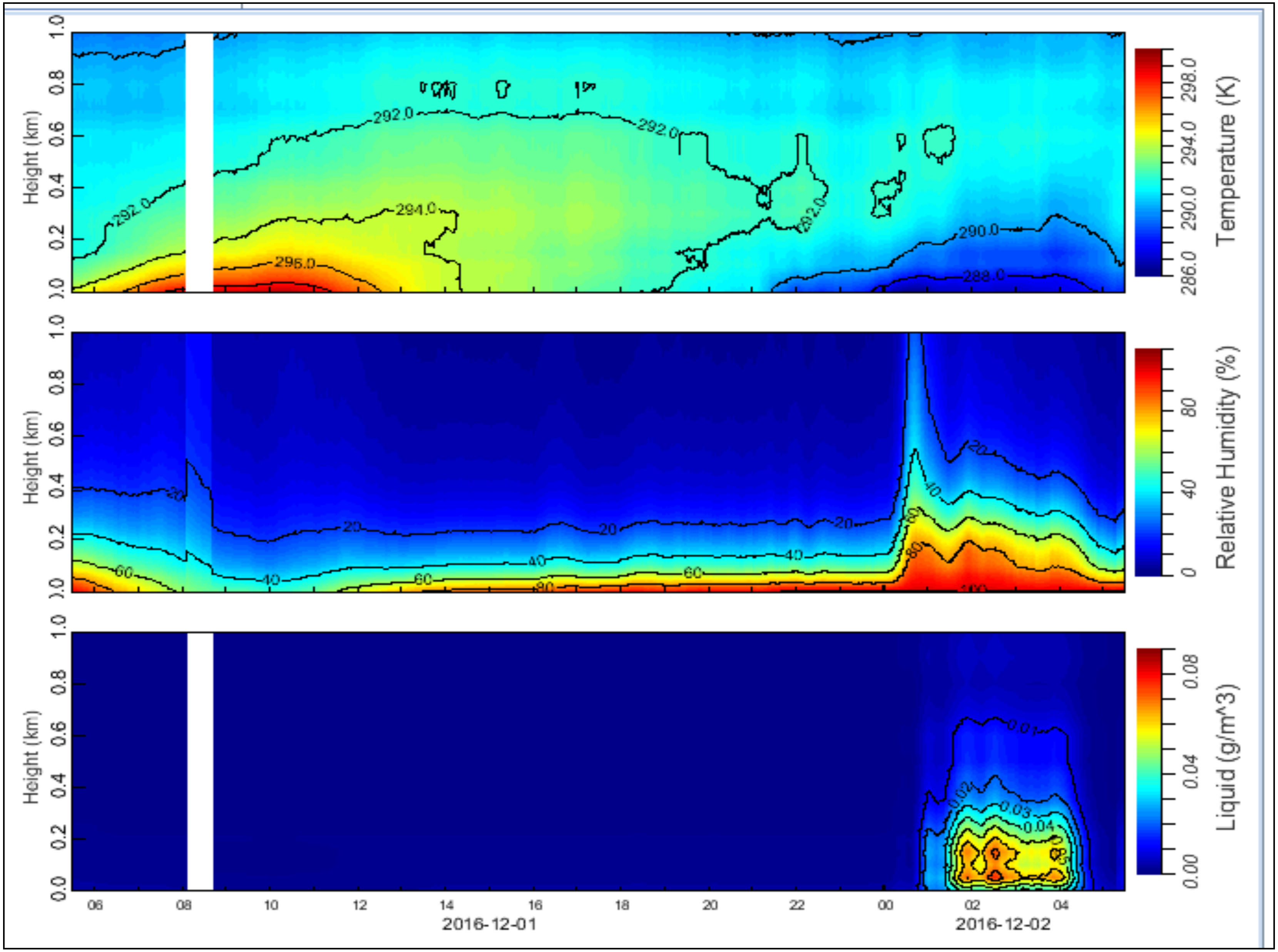
Data Assimilation
input for weather and climate models

Boundary Layer Profiling
- high-resolution temperature profiles (better than balloons)
- 24/7 monitoring of temperature inversions
- fog detection
- air pollution applications

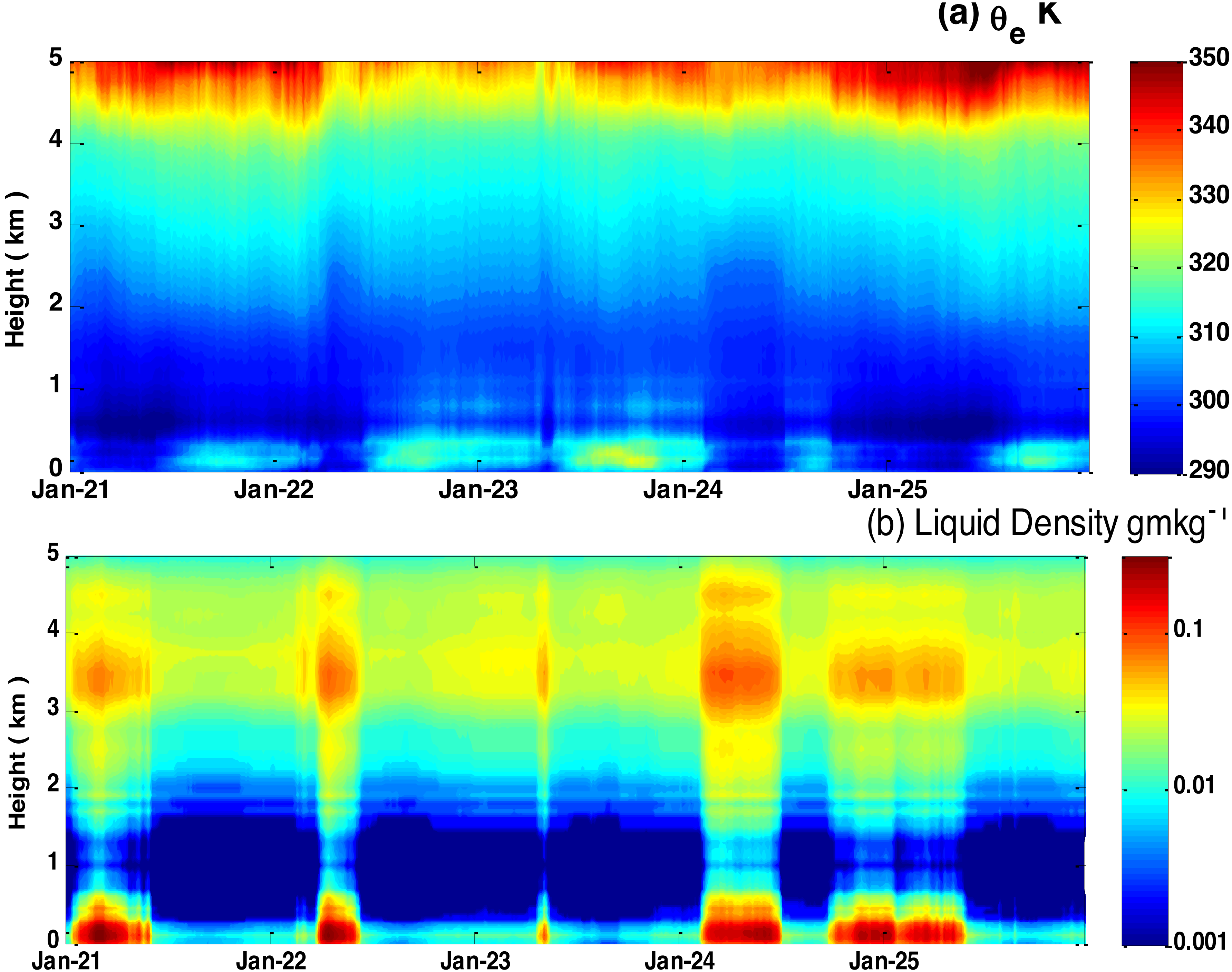
Temperature, Humidity and Liquid Profiler
21 K-band (22 to 30 GHz) channels
14 V-band (51 to 59 GHz) channels

that measures energy emitted at sub-millimetre-to-centimetre wavelengths


12 of 1432
mb
Parcel Data
Pres 390.7 mb
Hgt 7.691 m
(MSL) 25.232 ft
±Std +339 m
Temp -21.1 °C
Td -26.5 °C
T-Td 5.4 °C
RH 62%
PT 329.7 °K
ePT 333.8 °K
L.R. 7.6 °C/km
Tmax 55.0 °C
Wind:
Hgt:
(MSL)
Diagram Data
Pres 395 mb
Hgt 7.617 m
(MSL) 24.989 ft
Hgt 7.426 m
(AGL) 24.362 ft
Temp -8.7 °C
16.3 °F
DryA 71.7 °C
WetA 26.1 °C
MixR 5.0 g/kg
Stn Elev: 191 m
QNH = 1009.3 mb
DA: 846 m, ISA
250
300
400
500
600
700
800
900
1000
1050
-20
-10
0
10
20
30
40
50°C
TROP Lvl: -- m AGL
FRZG Lvl: 3953 m AGL
cclEL Hgt: -- m AGL
lfcEL Hgt: 7547 m AGL
LFC Hgt: 3474 m AGL
CCL Hgt: 2860 m AGL
LCL Hgt: 1864 m AGL
Water: 3.28 cm
Hail: 0.64 cm
T2Gust: 45 kt
WindEx: 48 kt
SWEAT: 88.8
CAP: 4.9
Boyden: 99.7
S{TT}: 38.1
KO: -5.9
LI: -2.0
TT: 48.4
KI: 28.1
Tc: 36.9 °C
Storm: -- 0-6km
s-rH: -- 0-3km
s-rH: -- 0-2km
s-rH: -- 0-1km
CAPE+ only: 181 J/kg
CIN total: -334 J/kg
DCAPE6.0km: 954 J/kg
VGP 0-4km: --
EHI 0-2km: --
MVV: 19 m/s
BRN: --
LFC Lift / LPL 987 mb
FOG FSI: --
Threat: 11.7
Point: 7.3°C
KM
FT (x1000)
10
9
8
7
6
5
4
3
2
1
0
30
25
20
15
10
5
0
MSL



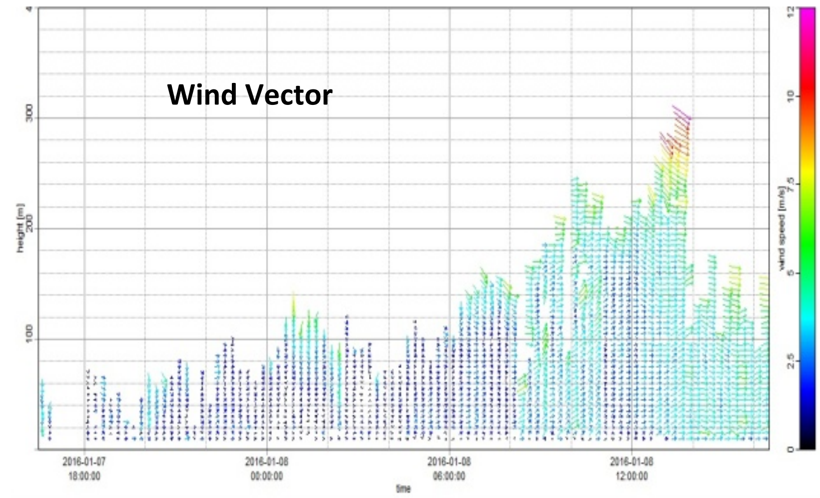
Height (km)
Temperature (K)
292.0
294.0
296.0
290.0
288.0
Relative Humidity (%)
20
40
60
80
100
Liquid (g/m^3)
0.01
0.02
0.03
0.04
06
08
10
12
14
16
18
20
22
00
02
04
2016-12-01
2016-12-02

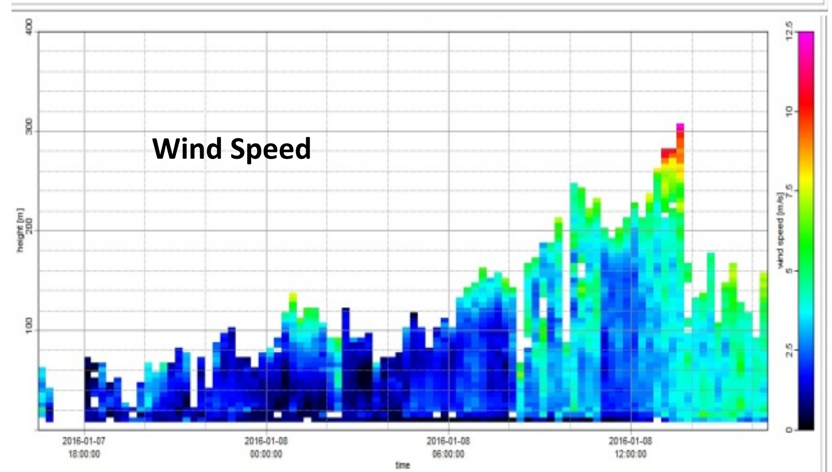


(a) θe K
Height (km)
Jan-21
Jan-22
Jan-23
Jan-24
Jan-25
350
340
330
320
310
300
290
(b) Liquid Density gmkg⁻¹
Height (km)
0.1
0.01
0.001

SODAR

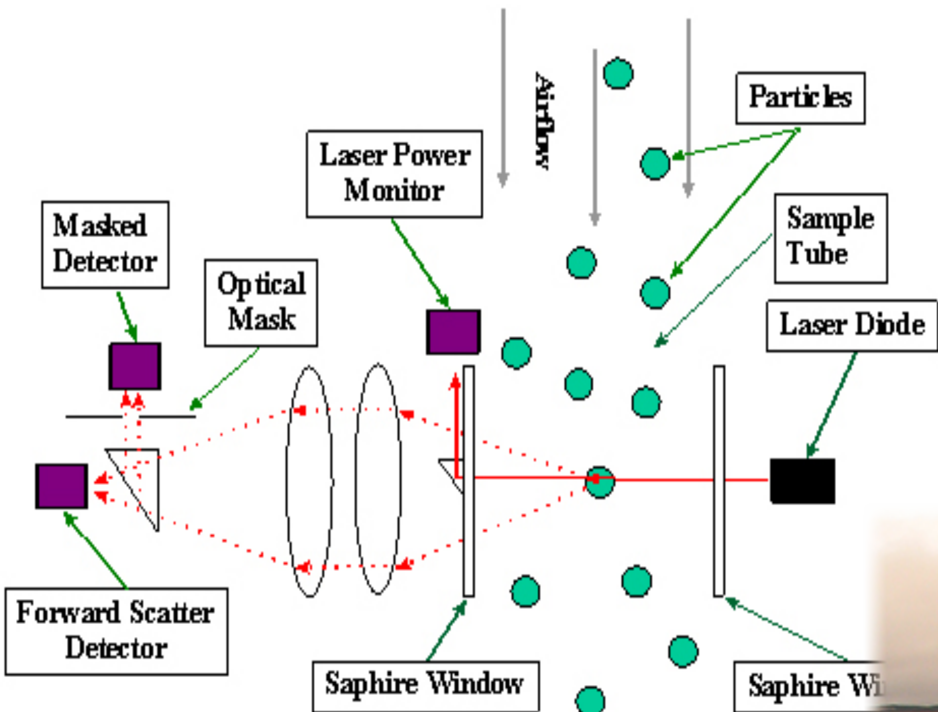
Used as a wind profiler to measure the scattering of sound waves by atmospheric turbulence





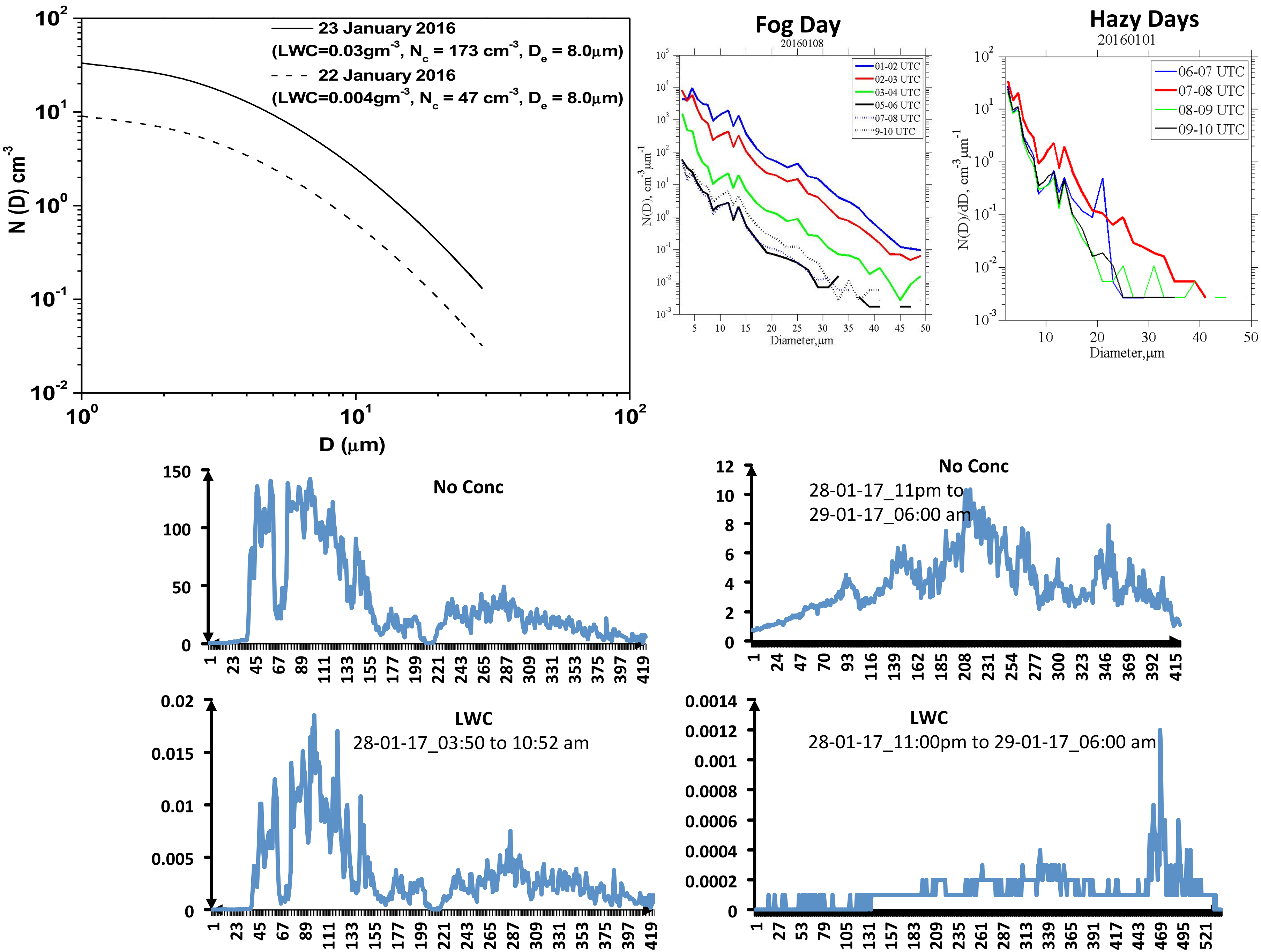
Fog Monitor (FM-120)

forward-scattering optical spectrometer



Droplets with 2-50 μm diameter

- Particle diameter
- Particle number concentration
- Liquid water content (LWC)
- Effective diameter (ED)
- Median volume diameter (MVD)



23 January 2016
(LWC=0.03gm-3, Nc = 173 cm-3, De = 8.0μm)
22 January 2016
(LWC=0.004gm-3, Nc = 47 cm-3, De = 8.0μm)
N (D) cm-3
D (μm)
Fog Day
20160108
01-02 UTC
02-03 UTC
03-04 UTC
05-06 UTC
07-08 UTC
9-10 UTC
N(D), cm-3 μm-1
Diameter,μm
Hazy Days
20160101
06-07 UTC
07-08 UTC
08-09 UTC
09-10 UTC
N(D)/dD, cm-3 μm-1
Diameter,μm
No Conc
No Conc
28-01-17_11pm to
29-01-17_06:00 am
LWC
28-01-17_03:50 to 10:52 am
LWC
28-01-17_11:00pm to 29-01-17_06:00 am

Emission

Outline

1. **What is an Emissions Inventory and why do we need one?**

    1. Building and bottom up emission inventory

2. **Satellite measurements and top down emission inventory**

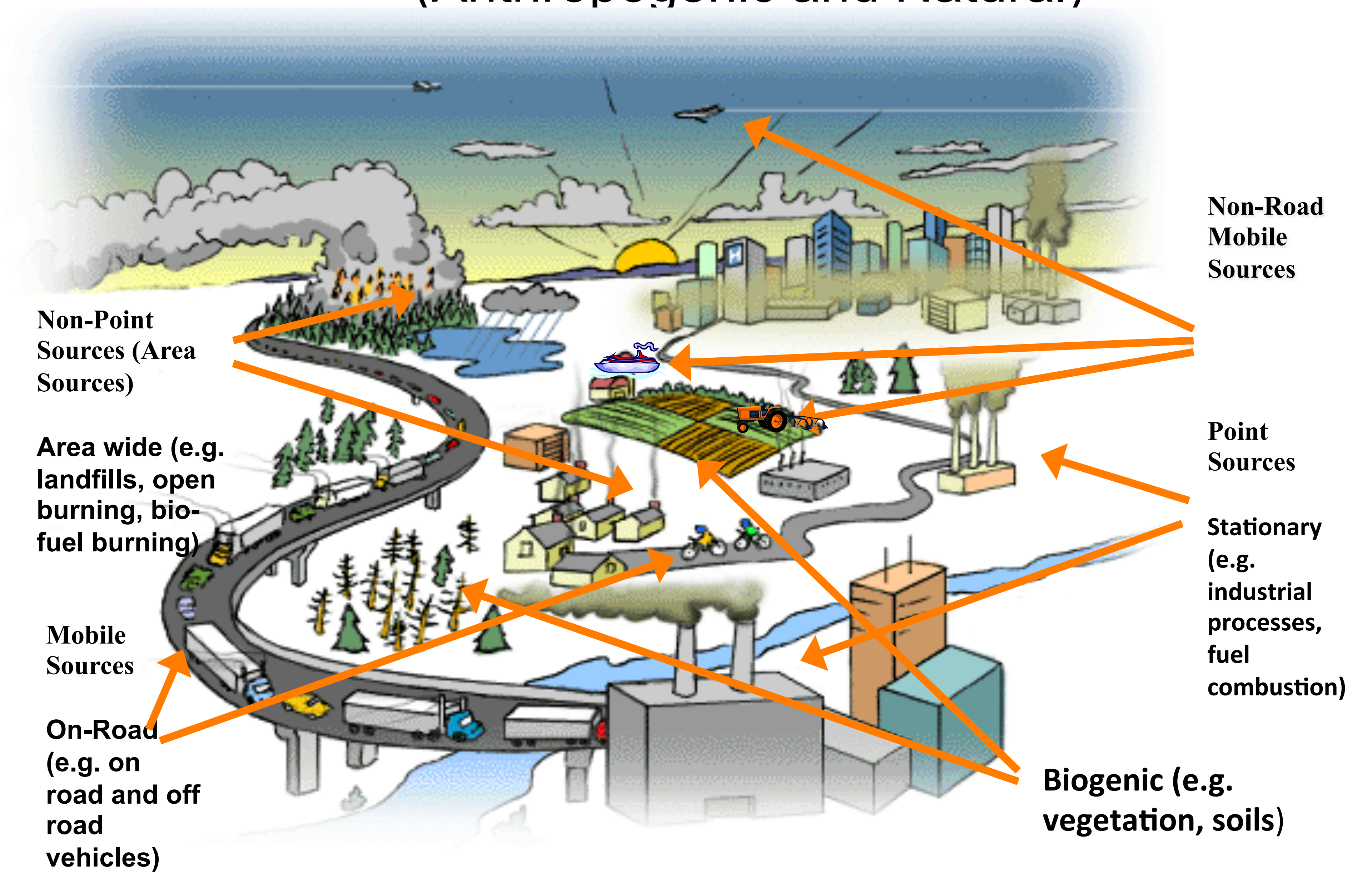
3. **Data base for Global and regional Emission Inventory data sources**

What is an Emission Inventory?

An emission inventory is an accounting of the amount of pollutants discharged into the atmosphere. An emission inventory usually contains the total emissions for one or more specific greenhouse gases or air pollutants, originating from all source categories in a

- Specific geographic area
- Certain period of time
- Organized by type of data (e.g., point, area, mobile, biogenic)

What are Emission Sources?
(Anthropogenic and Natural)



What information we get from EI?

- **Where air pollution is emitted**

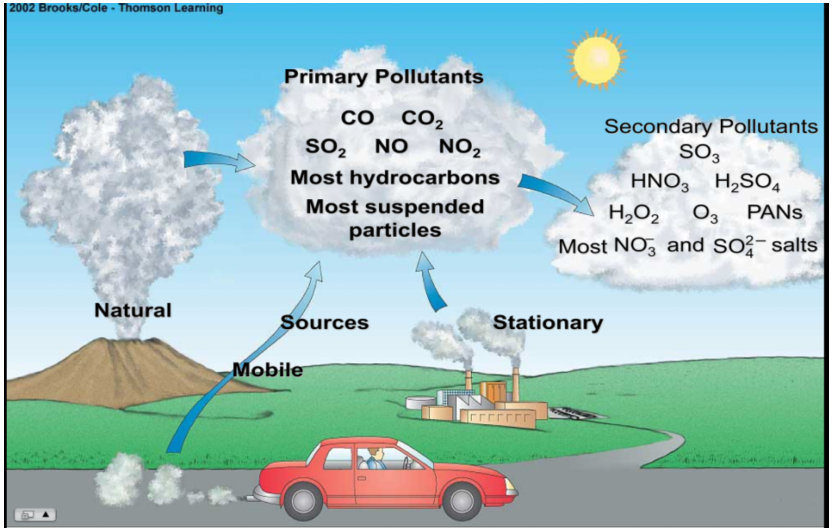
(**at surface:** e.g. from mobile and st. sources, ocean, biomass,
non- surface: e.g. Air craft, lightening, etc, Indoor activity or outdoor activity)

- **How much is emitted from each source**

- **What sources would be most effective to control**

Information we could not get from EI?

- The distance that air pollutant emissions are transported
- The amount of air pollution to which people are exposed
- The health risk from the air pollution

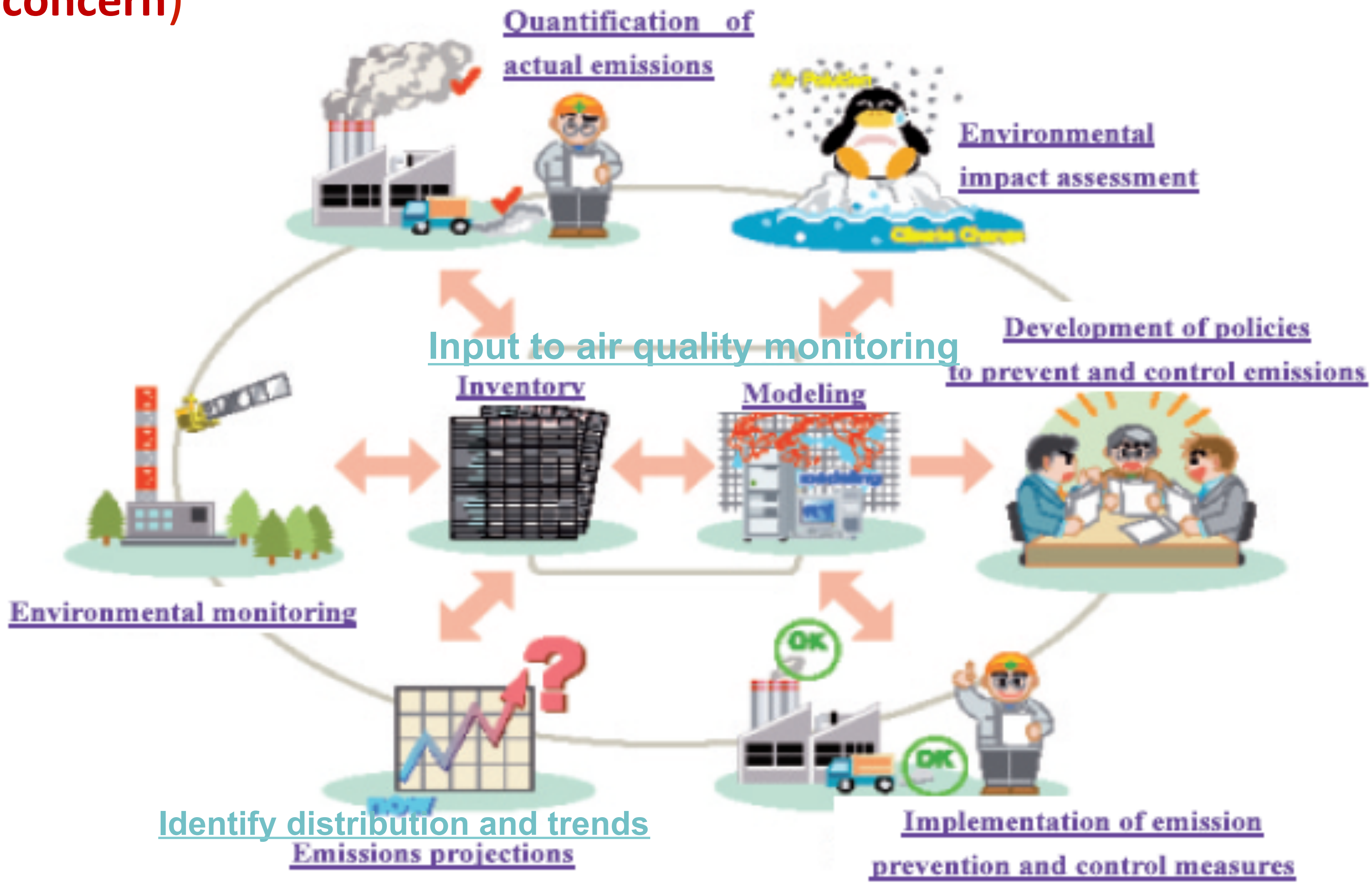


Formation of secondary pollutants (e.g. ozone, PAN, HNO3).

Windblown dust

Why is emission inventory needed?

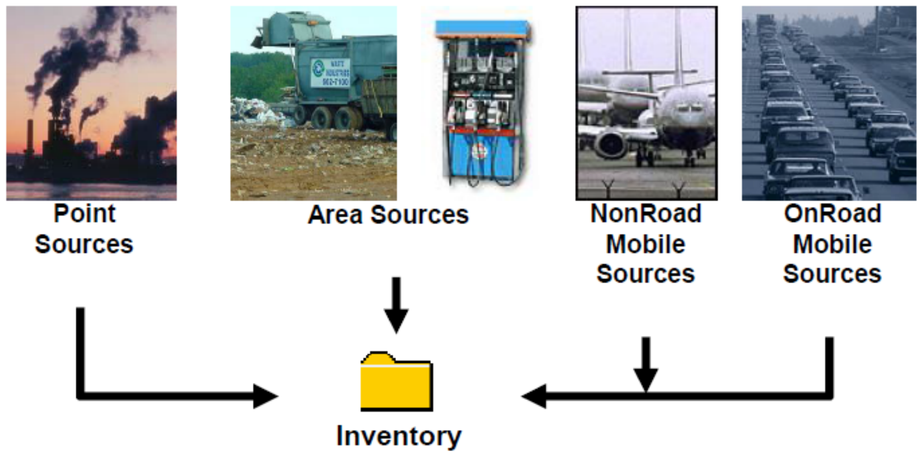
Air quality management (Identify sources of pollution and pollutants of concern)

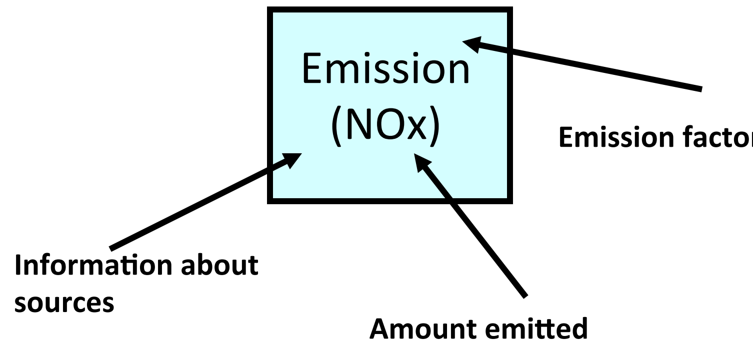


Types of Inventories

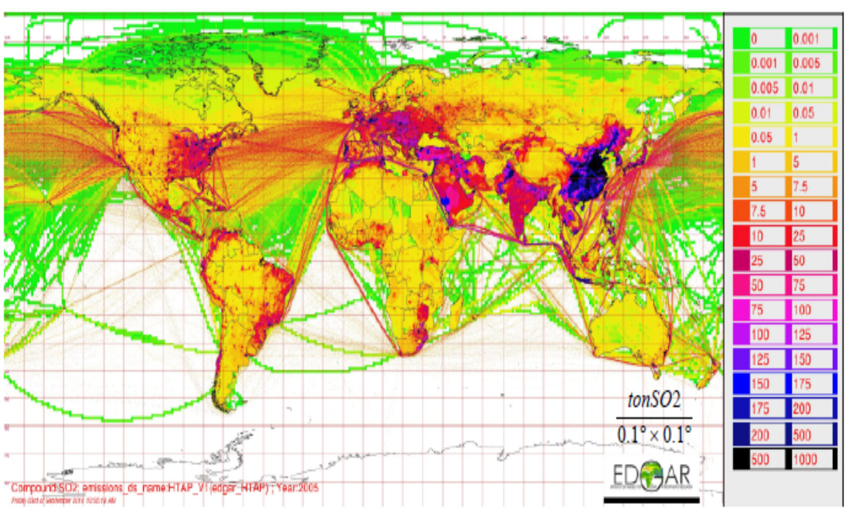
- **Bottom up EI** (emissions → concentrations)
- **Top Down** (concentrations → emissions)
    1. Direct estimate
    2. Combination of satellite and bottom up
- Annual average
- Seasonal inventories
- Forecasted - future estimates
- Gridded / Modeling

Building an bottom up Emission Inventory



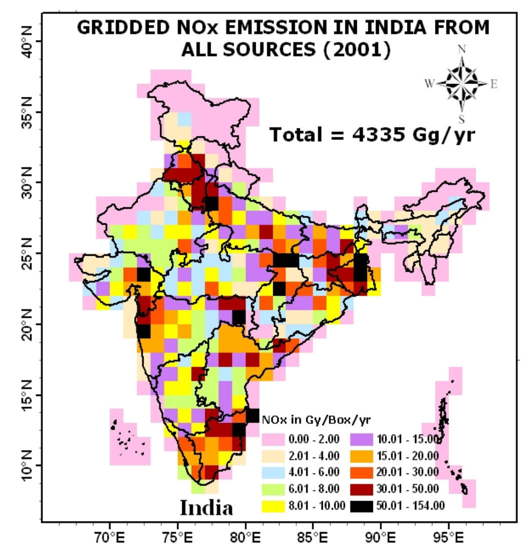


Edgar (global)



EDGAR-HTAPV1 SO_2 total emissions in ton per 0.1°x0.1°

India (regional)



Activity Data and Methodology

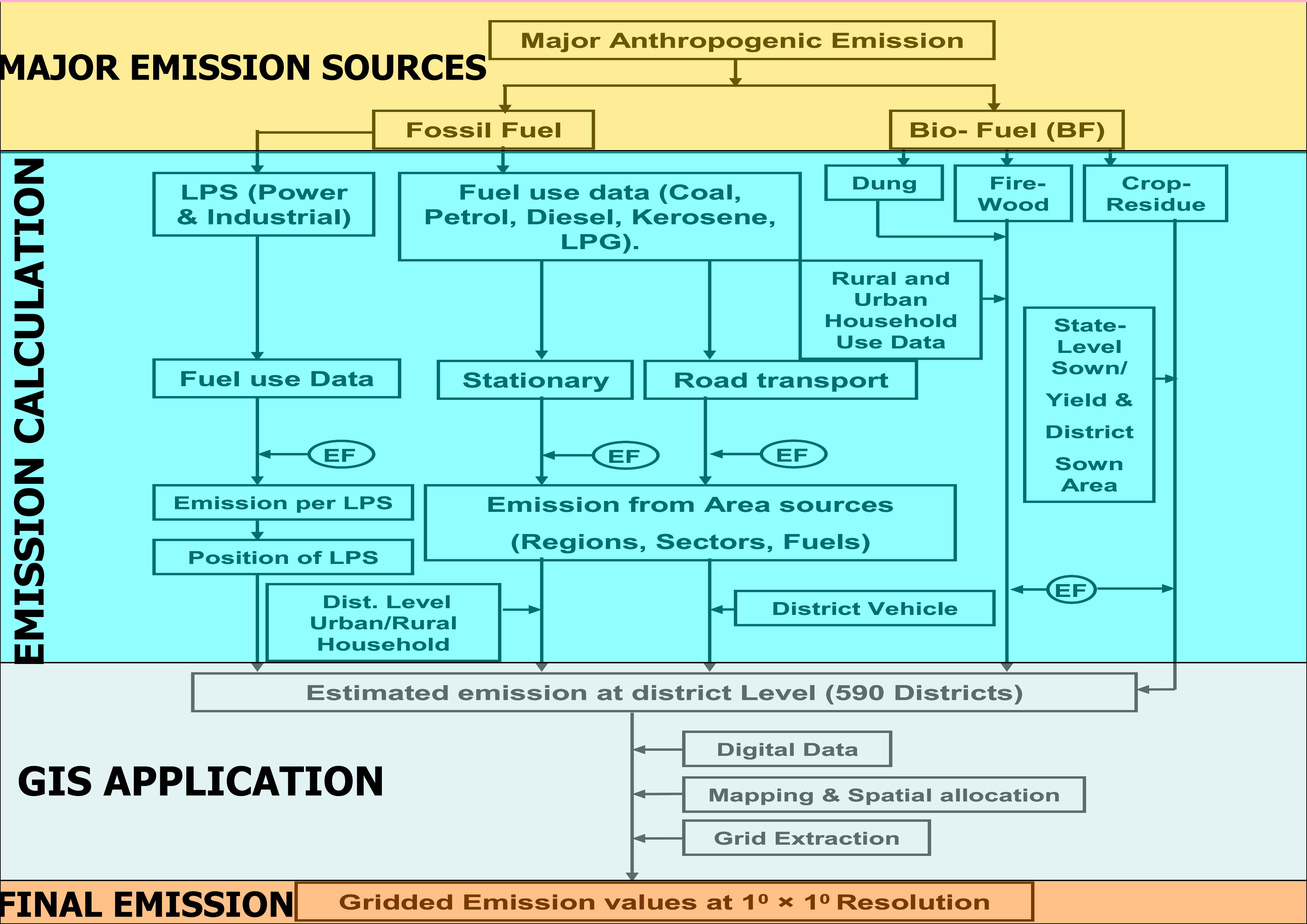
- **Methodology**

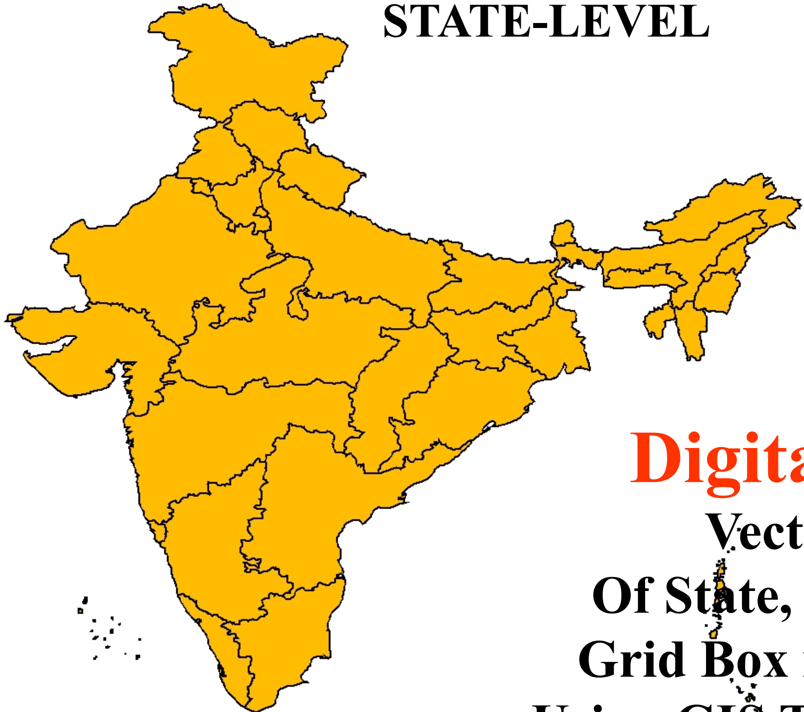
Emission Inventories are estimated as per the most widely used emission factor approach.

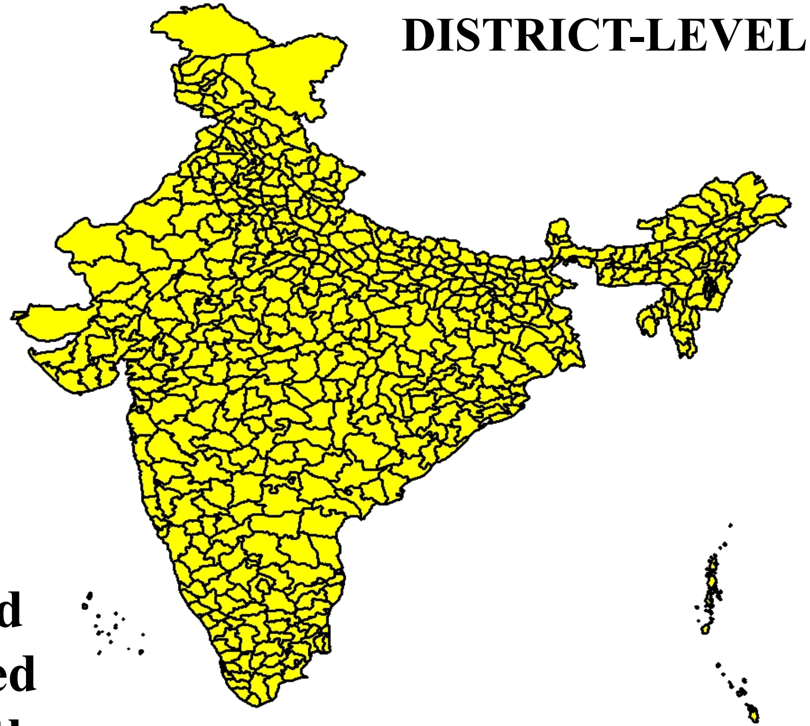
$$Em_{j,k} = \sum_{l}\sum_{m} FC_{k,l,m}\left[\sum_{n} EF_{j,k,m,n} X_{k,l,m,n}\right]$$

- *j,k,l,m,n* =Species, country, sector, fuel type, Technology
- Em = Total Emission
- FC = Fuel consumption , Kg/yr
- EF $\sum X$ = Emission factor specific to fuel/Technology
- X = Fraction of fuel for this sector consumed by a specific technology, where =1 for each fuel and sector

Schematic Methodology for the Development of Indian emission estimation

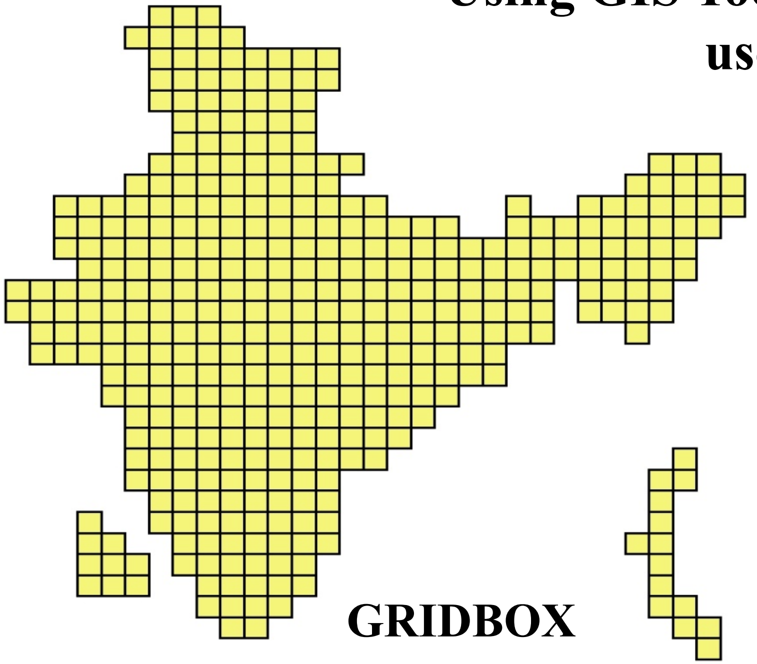


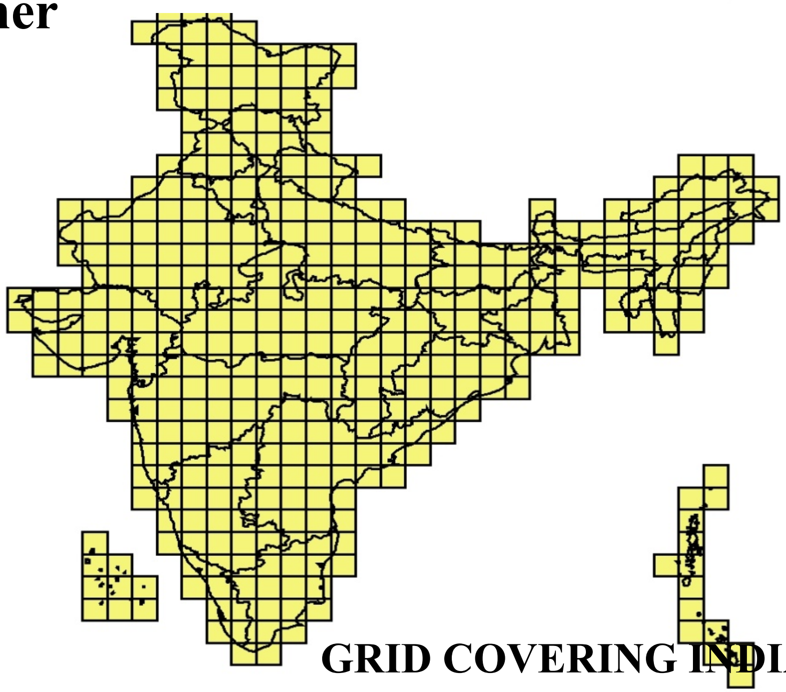




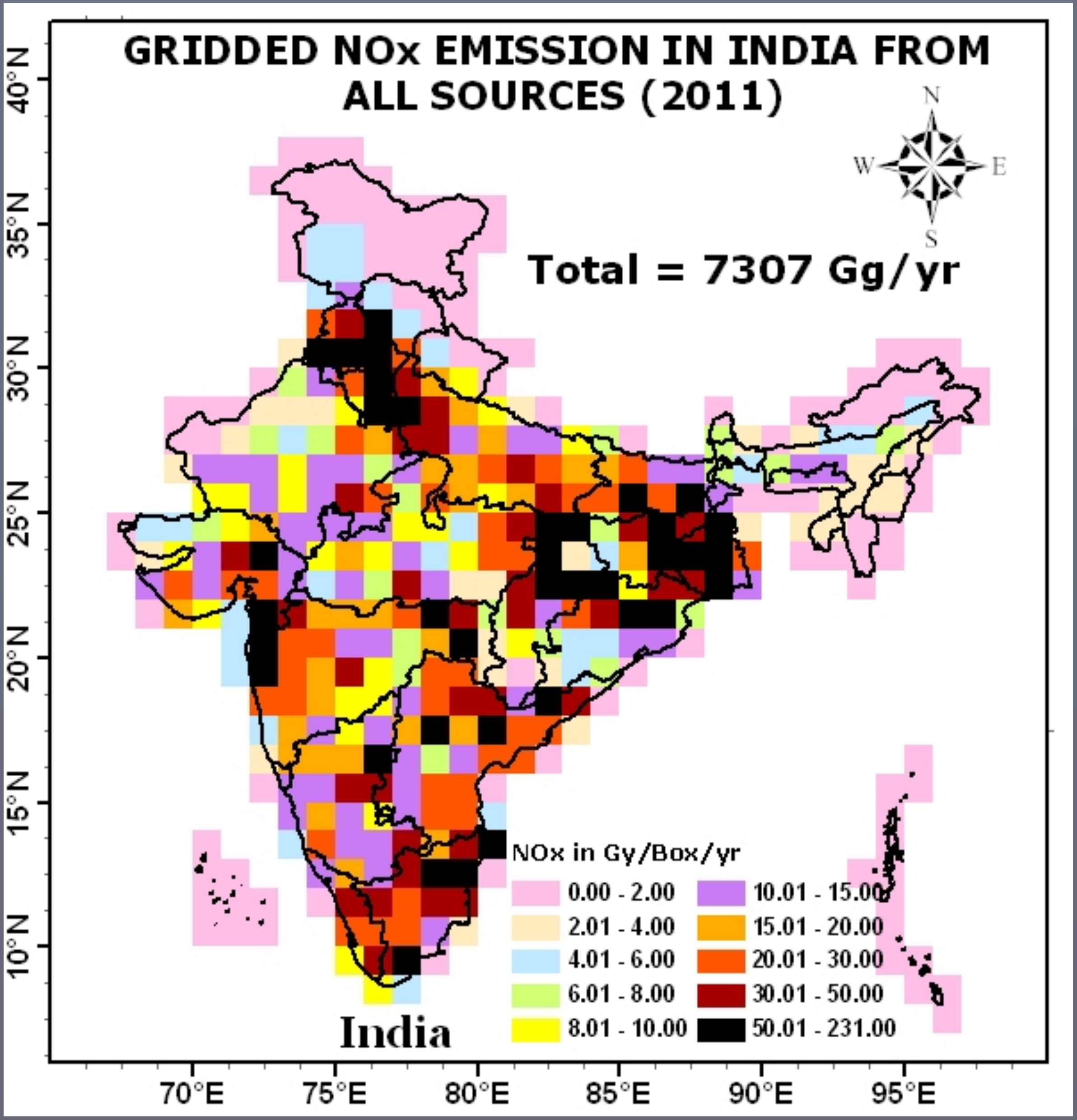
Digital Data:

Vector-Map Of State, District and Grid Box is Generated Using GIS Tool for further use.

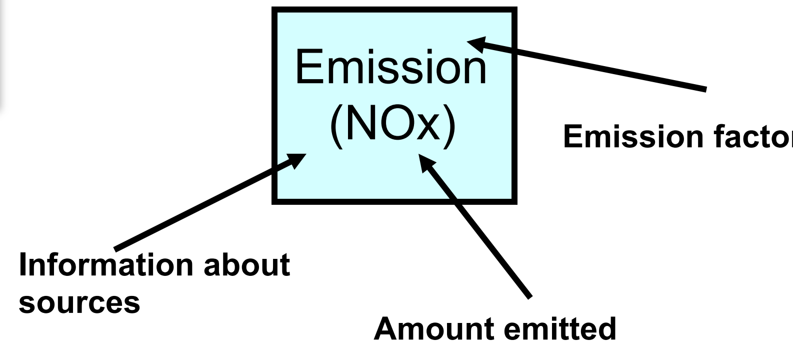




NO_x Emission in India (2011)



Drawbacks of bottom-up inventories



- Depend on the availability and reliability of the statistical information.
- Depend on historic information: easily out-dated.
- Uncertainties in spatial resolution if only area totals are available.

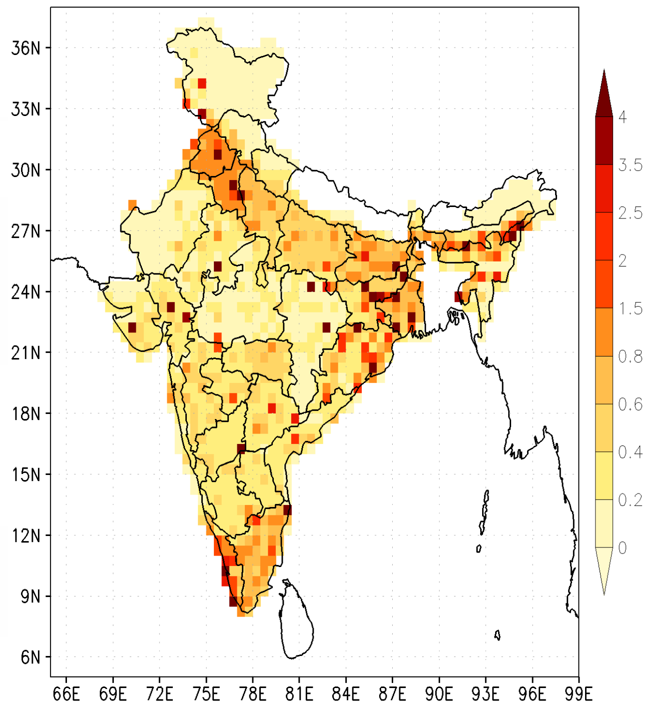
If you don’t like your result change your approach!

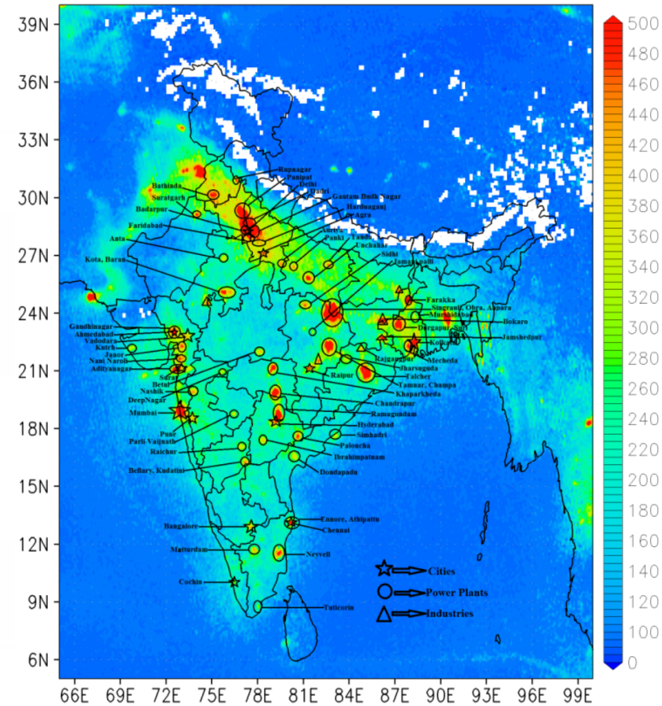
Constraining emissions with satellite observations

- Satellites have world-wide, homogeneous coverage.
- Correcting inventory for emission trends
- Detecting new (unknown) emission sources
- Emission trend analysis reveals effectiveness of air pollution policy
- Up-to-date emission inventories improve air quality forecasting

Top Down Approach

Emissions ← Concentrations





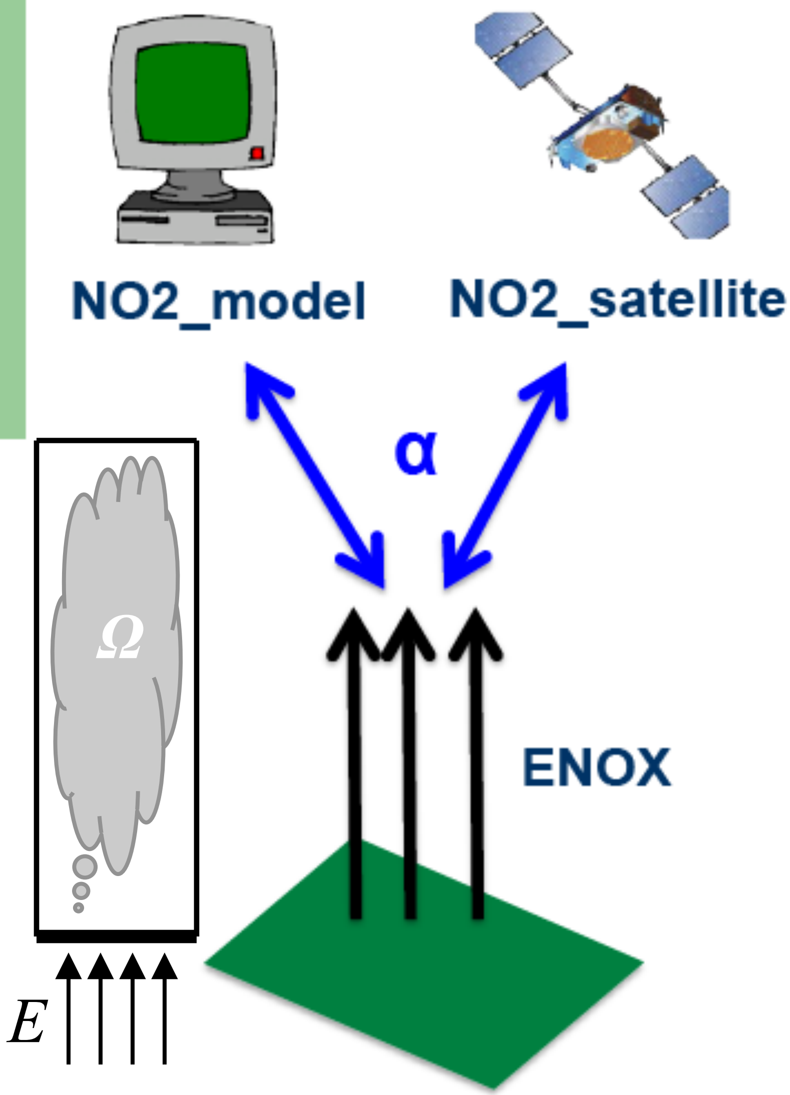
- **Top Down (concentrations → emissions)**

  1. **Direct estimate**

  2. **Combination of satellite and bottom up**

Top-down Approach

Martin et al. [2003, 2006]
Lamsal et al. [2011]



= Adjustment of the emissions with satellite observations to reduce the disagreement between model and observation.

Assuming that horizontal transport of NOx is negligible, a posteriori emissions can be derived as following:

α = **ENOX_apriori** / **NO2_model**

=> **ENOX_aposteriori** = α x **NO2_satellite**

ENOX = anthropogenic NOx emissions
NO2_model= Modeled NO2 Tropospheric Column
NO2_satellite= Satellite NO2 Tropospheric Column

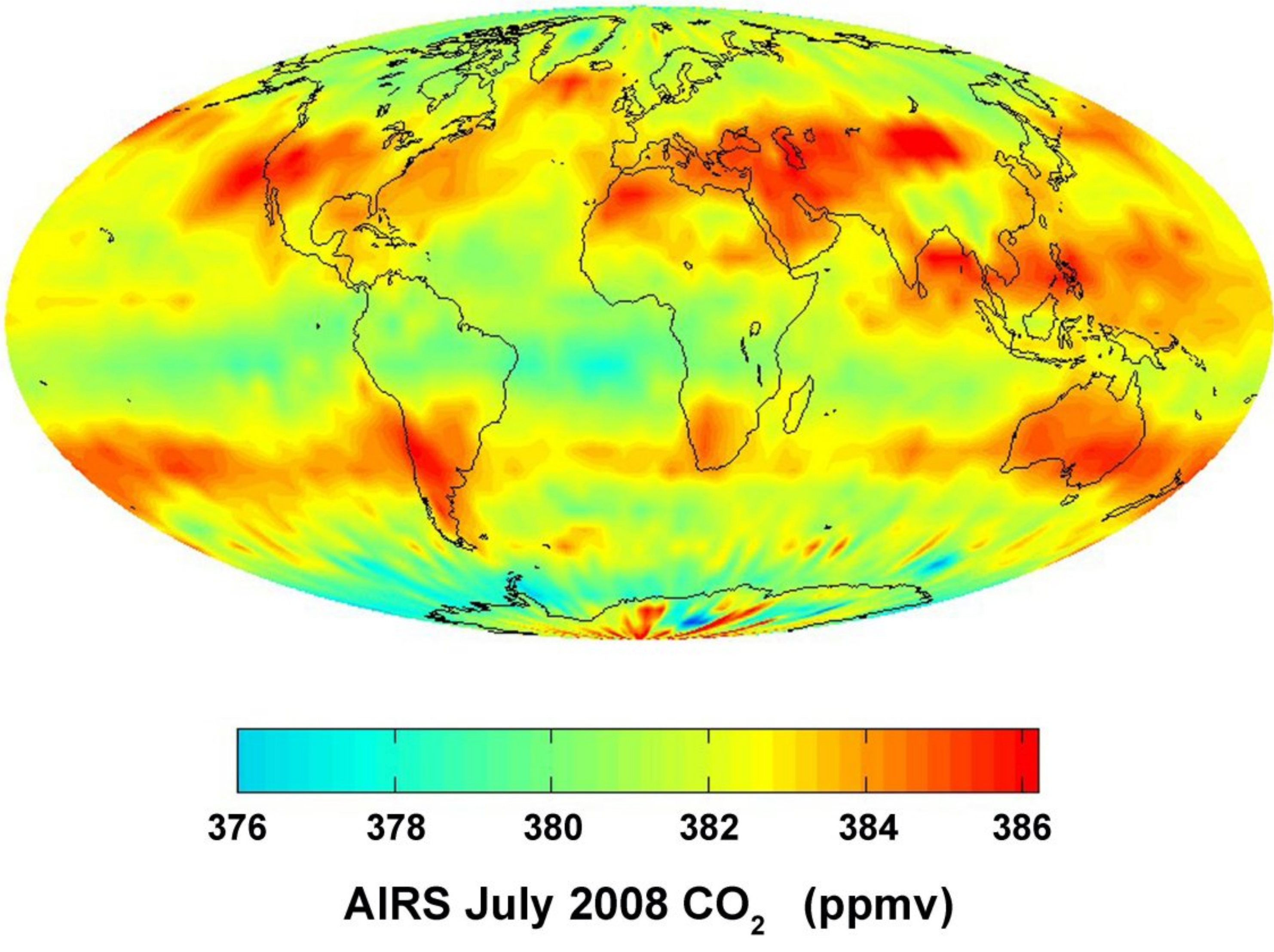
Satellite can only observe pollutant concentrations. These should be backtracked to their underlying emissions:

THIS IS AN INVERSE PROBLEM

Complicating factors:

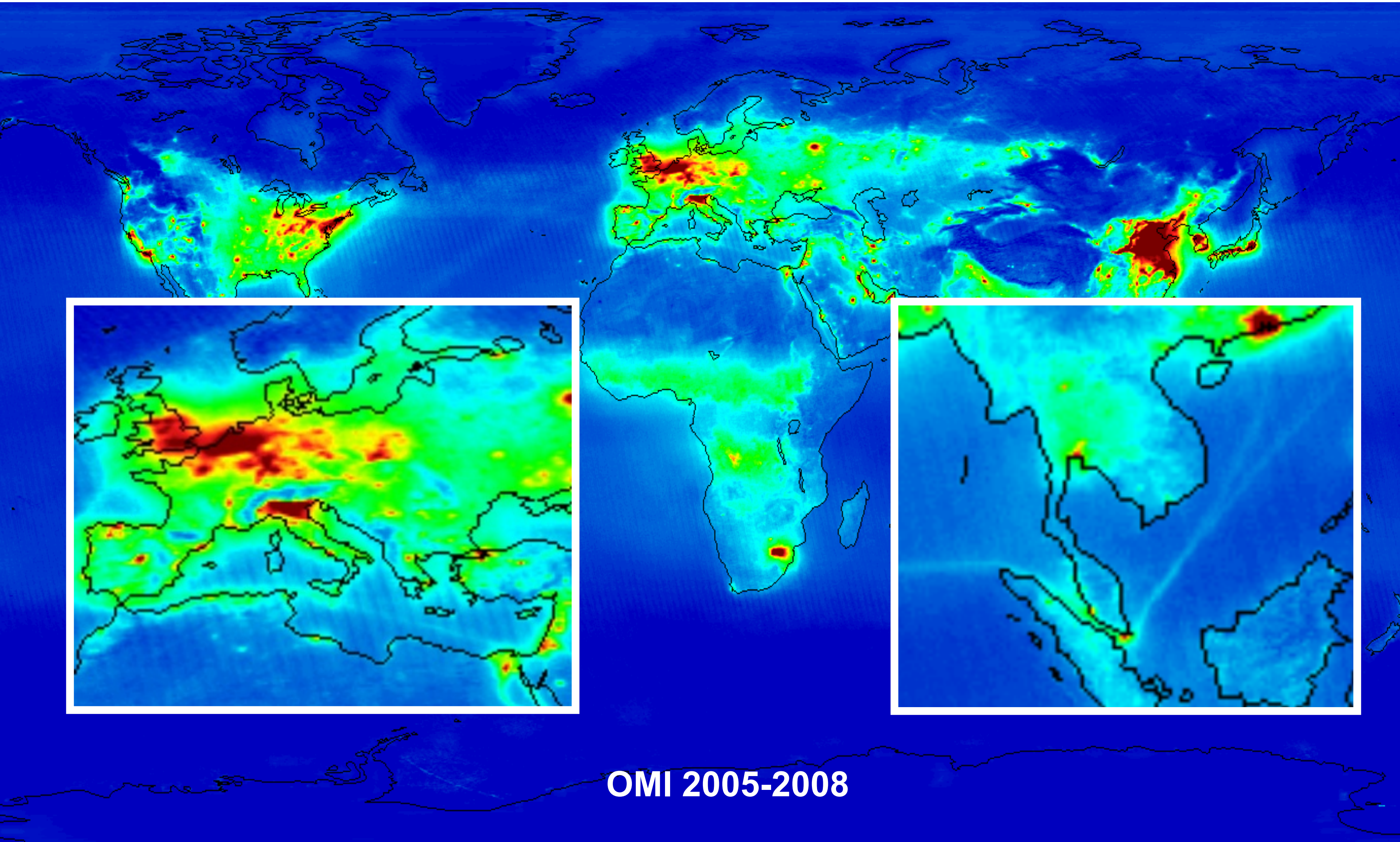
- Transport of pollutant away from the source
- Lifetime of pollutant
- Variability in lifetime (temperature, chemical composition…)

Lifetime example (1): CO_2



Lifetime example (2): NO_2

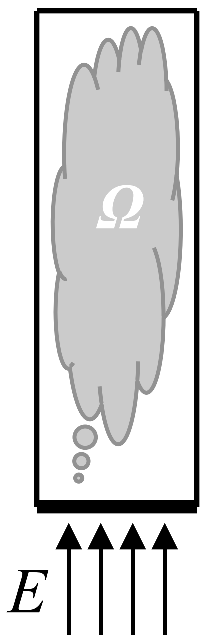
tropospheric NO_2 in summer: ~4h, in winter: ~10h



Local, linear relation concentration and emission

Martin et al. (2006) Space-based constraints on NOx emission, J. Geophys. Res.
Jaeglé et al. (2005) Global partitioning of NOx sources using (…), Faraday Discuss.

Assume linear relation between NO_x emission and NO_2 concentration:



Direct Approach

$$E_t = \alpha\Omega_{obs} \quad , \quad \alpha = (\Omega_{NO_X} / \Omega_{NO2}) / \tau_{NO_X}$$

Combine Approach

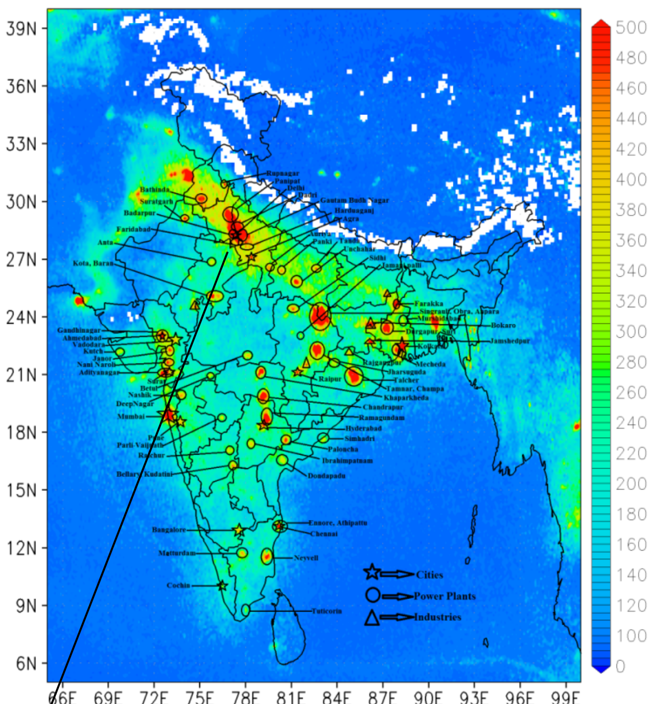
$$E = \alpha\Omega_{obs} \; , \quad \alpha = E_{a\,\mathrm{mod}} / \Omega_{No2m}$$

Local, linear relation concentration and emission

Direct Approach

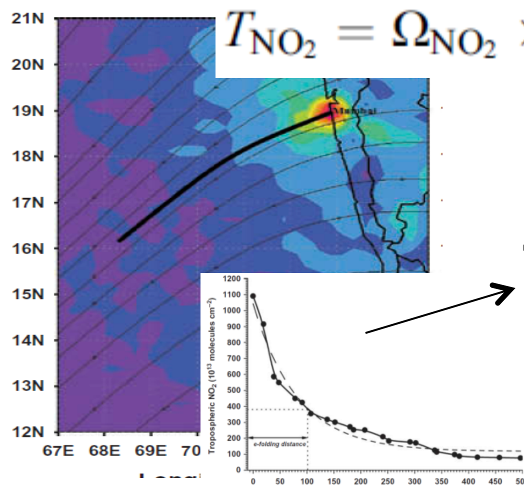
$$E_t = \alpha\Omega_{obs} \quad , \quad \alpha = (\Omega_{NO_X} / \Omega_{NO2}) / \tau_{NO_X}$$

From literature, measurements



1.7 x 10[15] mole/c m[2]
(25km x 25 km)

$$T_{NO_2} = \Omega_{NO_2} \times \exp(-t/\tau_{NO_x}),$$



11.2 hours = 11.2*3600 sec

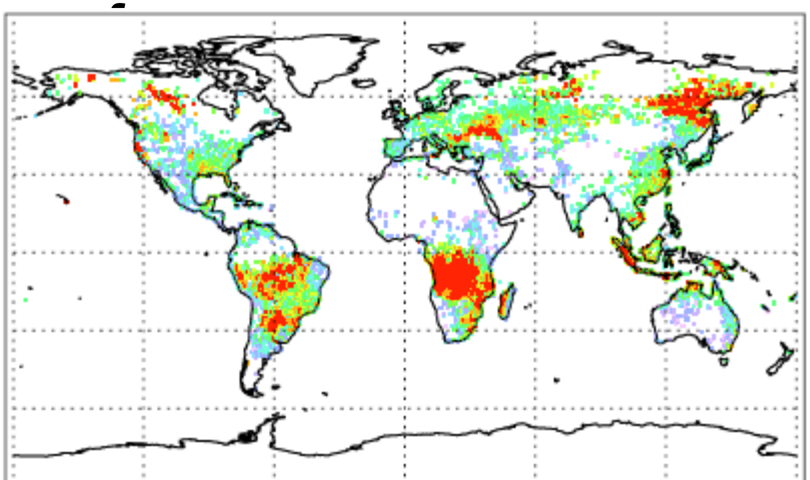
Et = (0.31/11.2)*1.7 x 10[15] mole/cm[2]

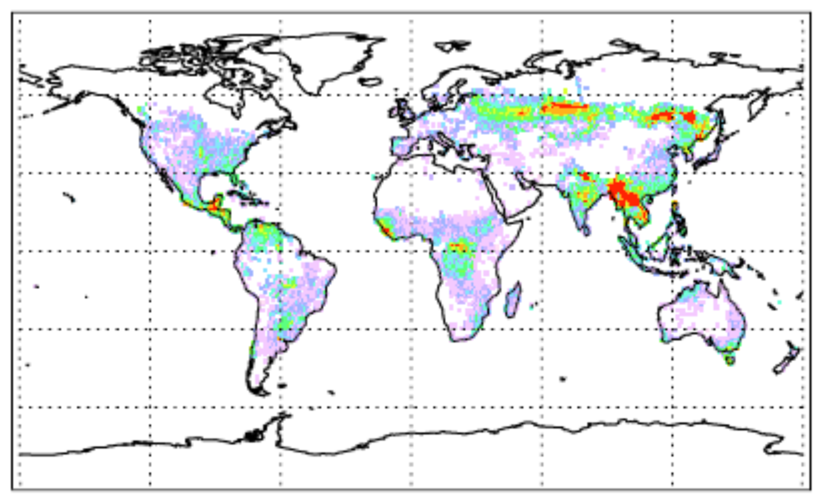
Combine Approach (Lab + Satellite)

Satellite based estimates of Fire emission

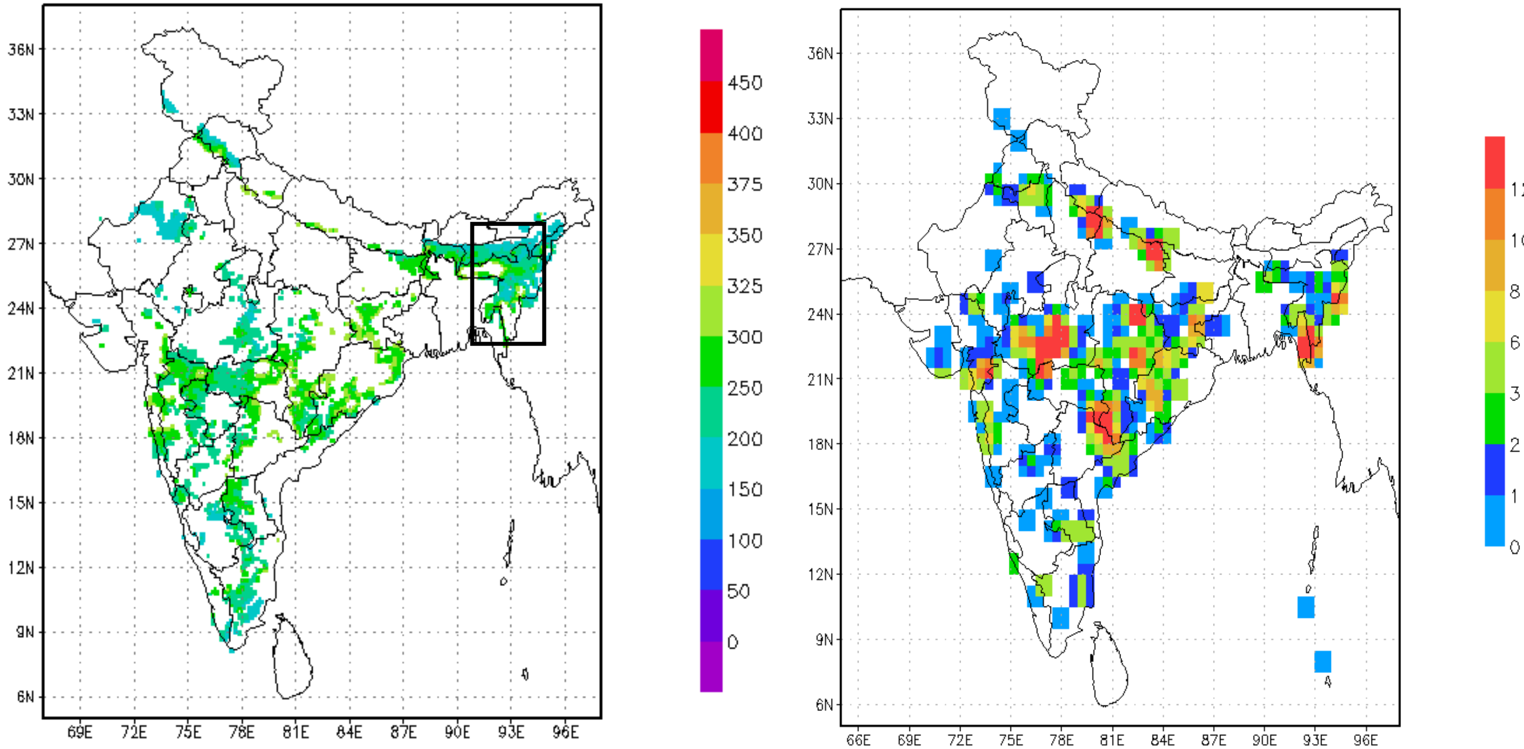
$$E_i = A(x,t) \times B(x) \times \mathrm{FB} \times \mathrm{ef}_i$$

Use of fire hot spots
Burnt area at location x and time t: A(x,t)
land cover maps (identify vegetation type)
Biomass loading : B(x)
consumption estimates at location x: FB
Emission factors (emission factors from filed lab experiment):





Complete Satellite based **Approach**



(a) Spatial distribution of OMI NO_2 (1 x 10^{13} molecules cm^{-2}) for 2005 for regions with a maximum in the seasonal cycle in tropospheric NO_2 during March-April. In these regions the dominant source type is estimated as biomass burning. **(b)** ATSR fire counts over the India region during March-April 2005 averaged over 0.5º ×0.5º.

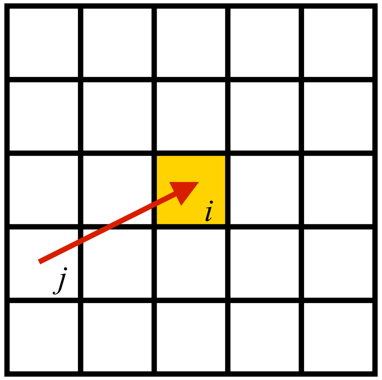
Local, linear relation concentration and emission

Advantages

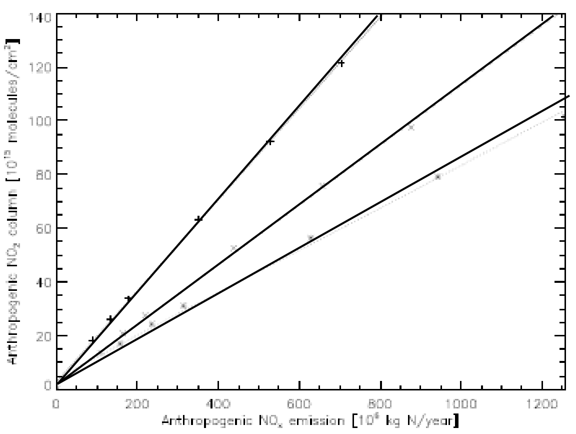
- Fast, no inverse modeling needed

Disadvantages

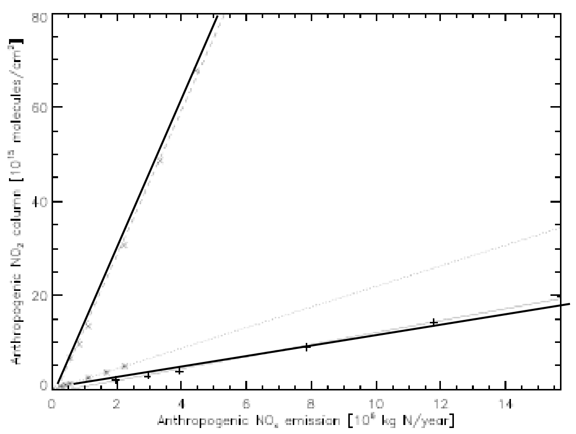
- Transport to neighbouring grid cells neglected
- Only one emission update possible
- No new sources detected if *a priori* emission is 0



Urban



Rural



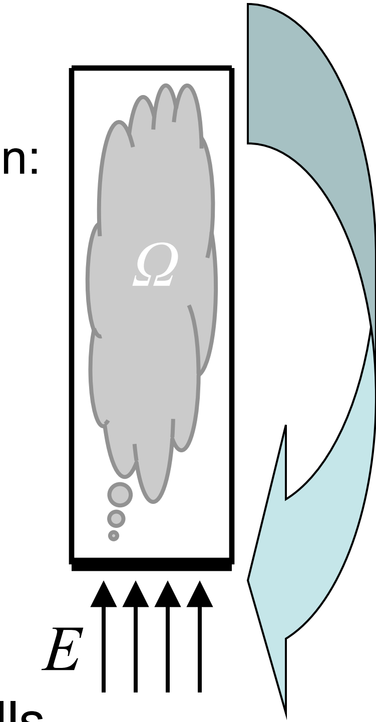
Local, linear relation applied iteratively

(Ghude et al., (2013), GRL

Assume linear relation between NOx emission and NO2 concentration:

$$E_{i+1} = \alpha \Omega_{obs,i} \quad , \quad \alpha = E_i / \Omega_i$$

Iterate until convergence criteria are met.



Advantages

- Iteration compensates for transport to neighbouring grid cells
- Accuracy of emissions improves

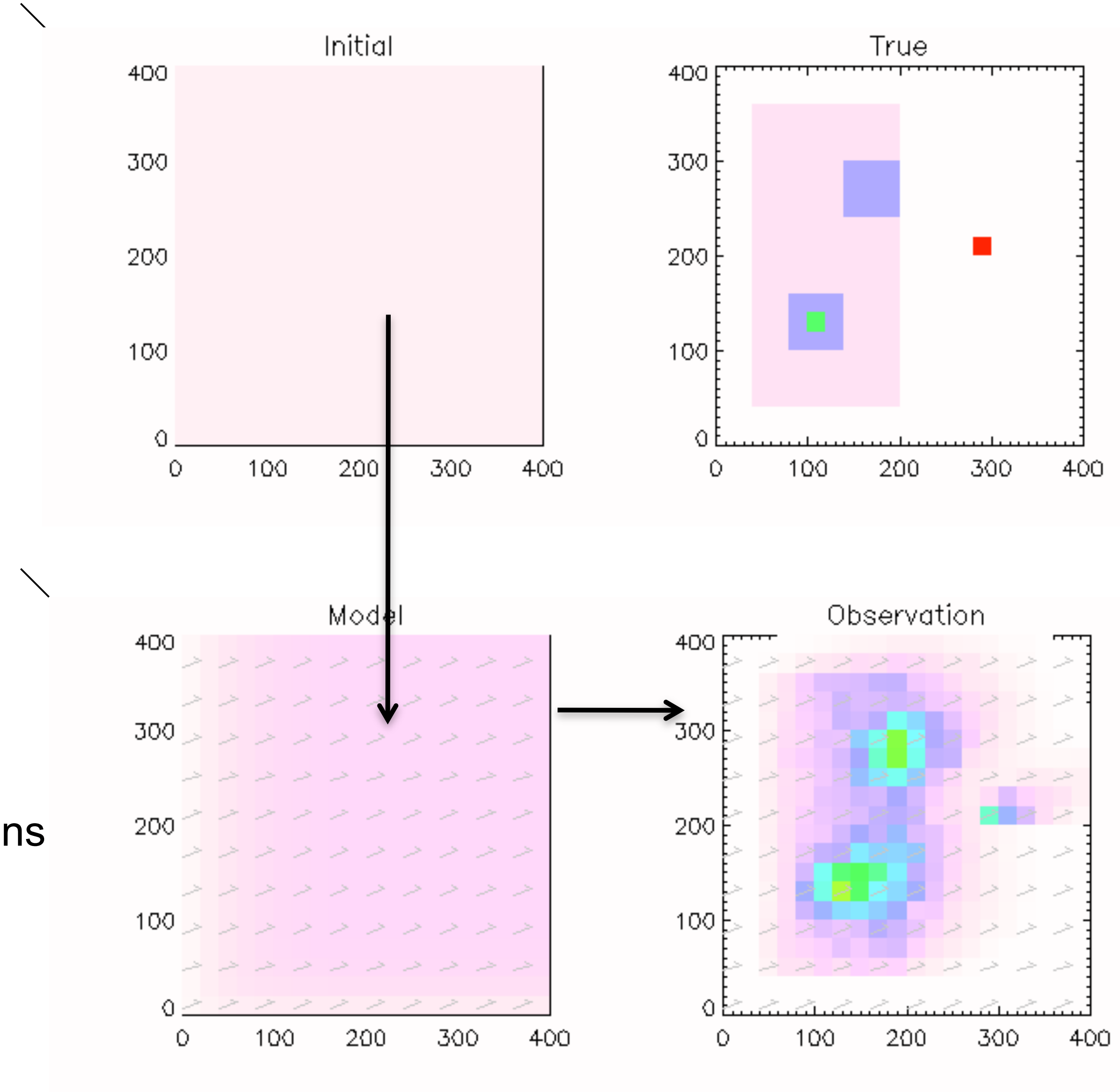
Disadvantages

- No new sources detected if *a priori* emission is 0

Flatland simulation

- “Toy” transport model in two dimension
- Simplified advection model allows analytic calculation of sensitivities

emissions

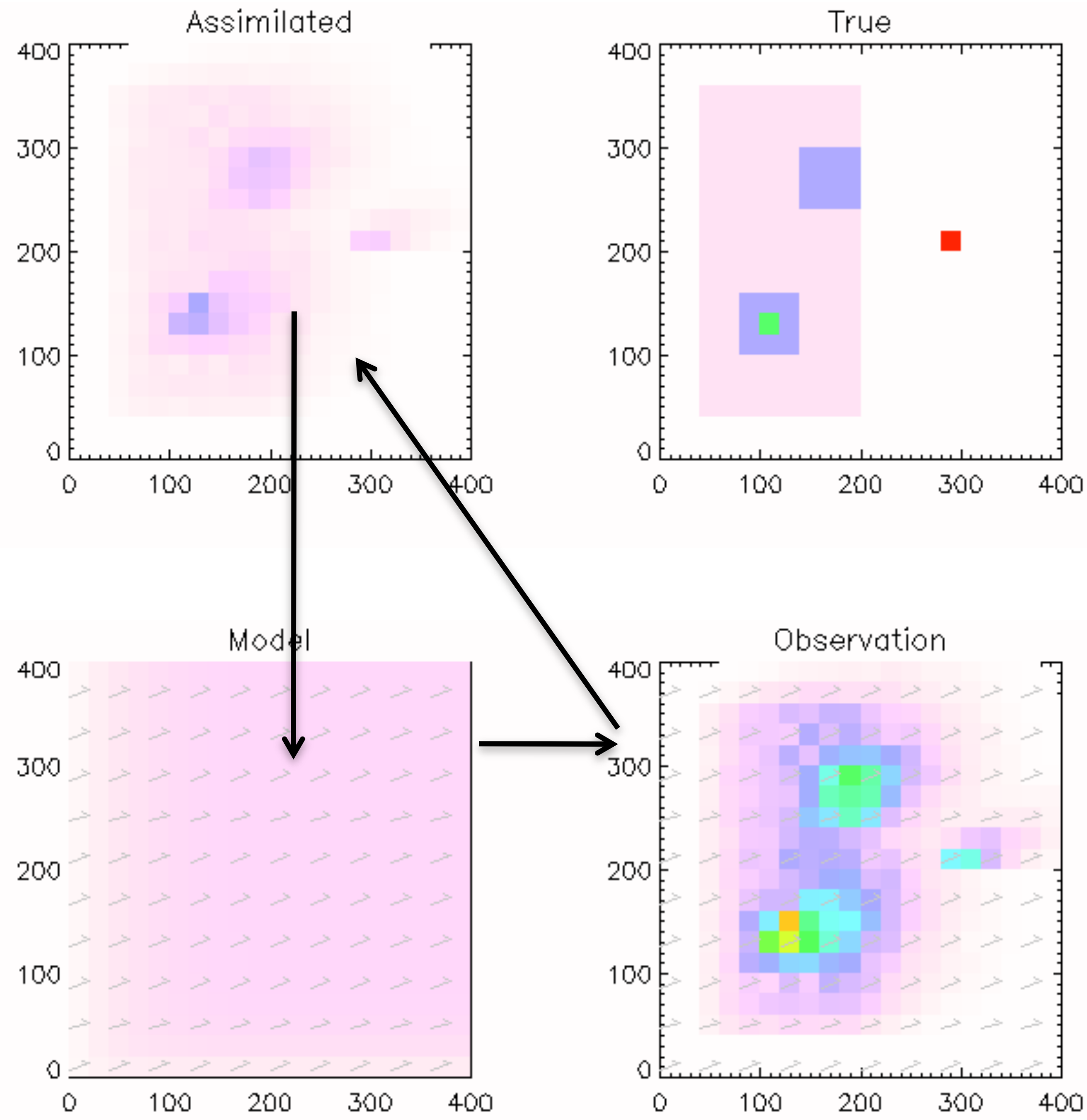


concentrations

Local, linear

emissions

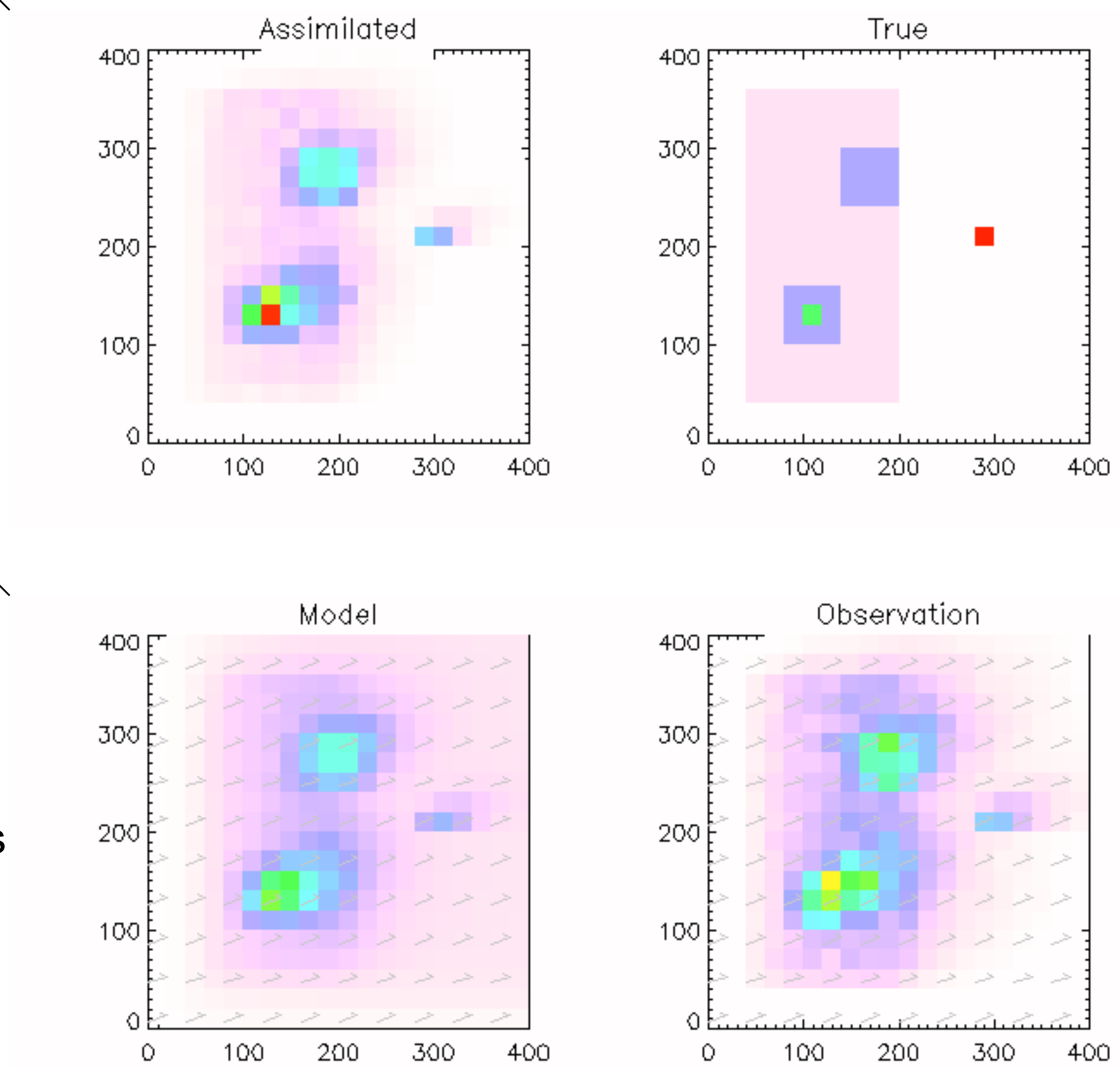
concentrations



Local, linear

emissions

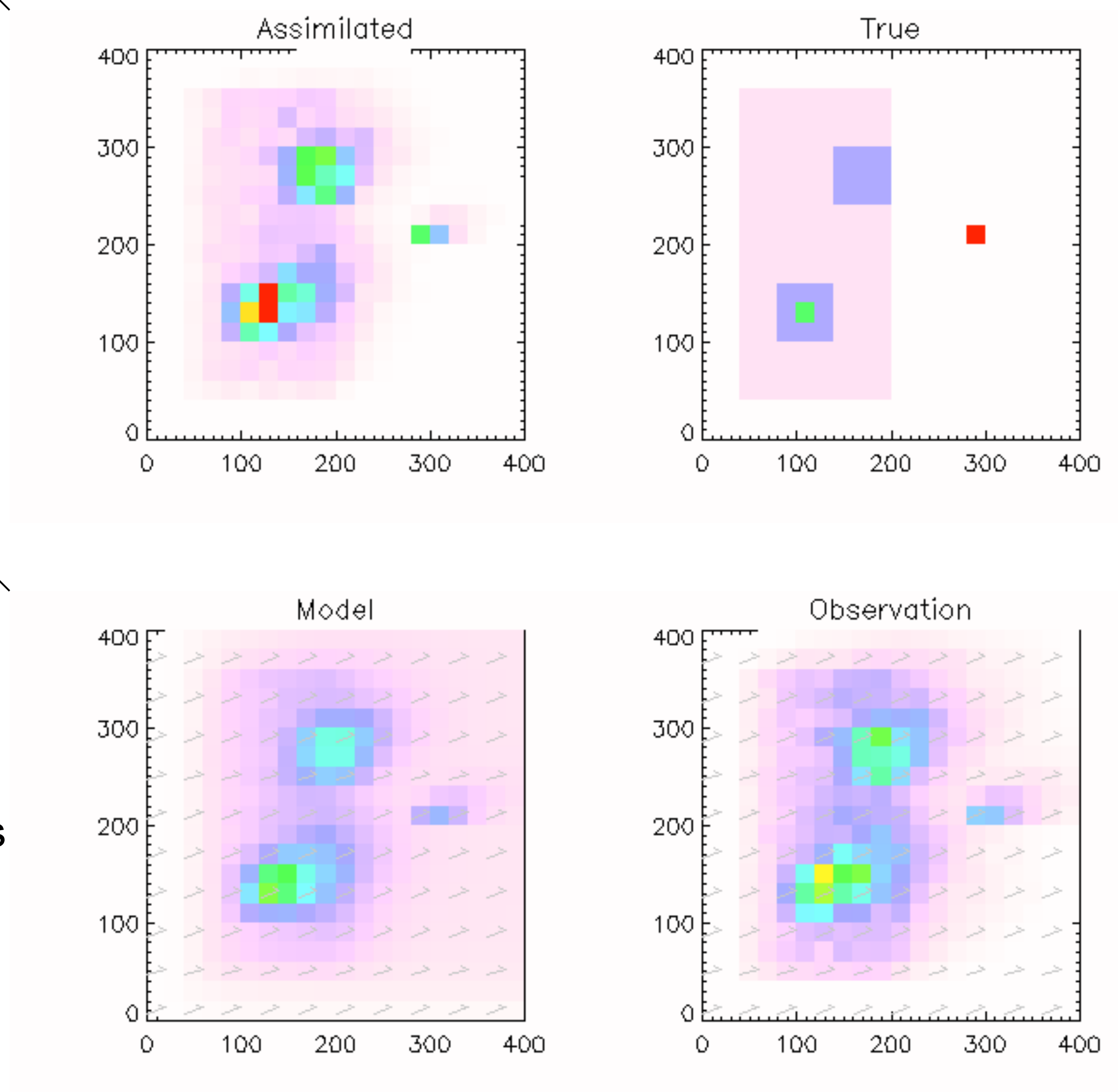
concentrations



Local, linear

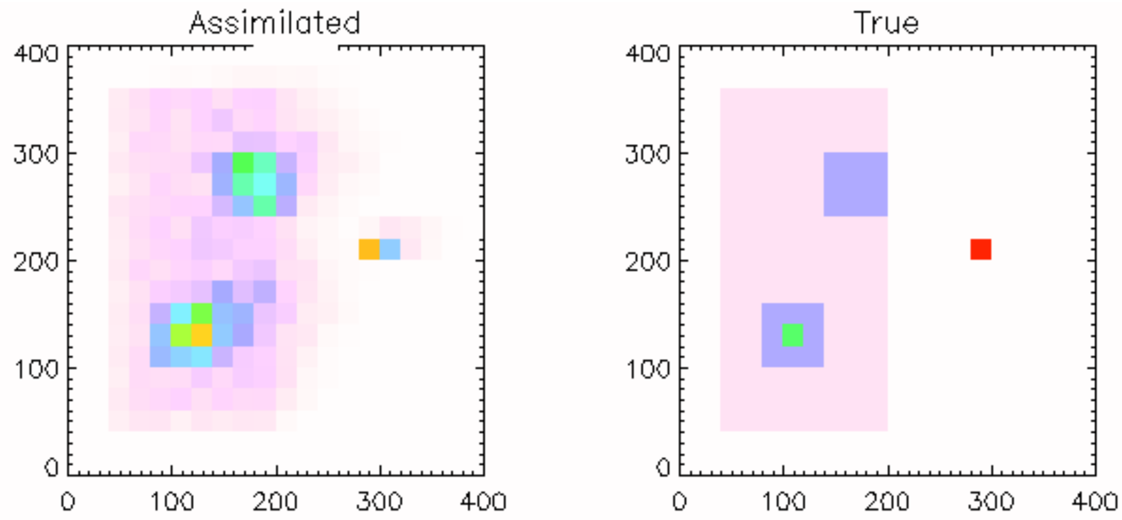
emissions

concentrations

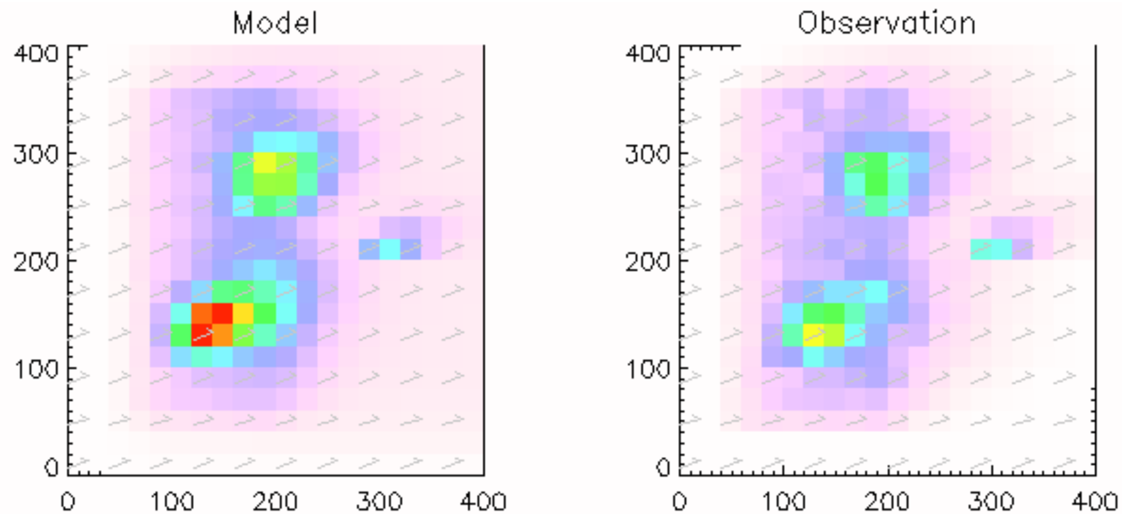


Local, linear

emissions

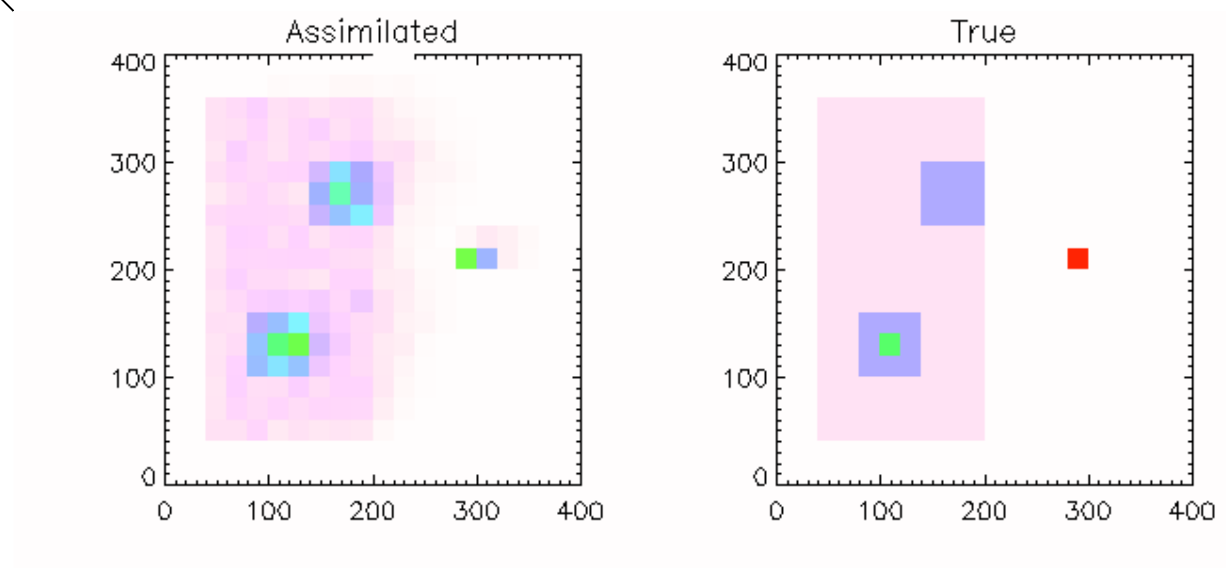


concentrations

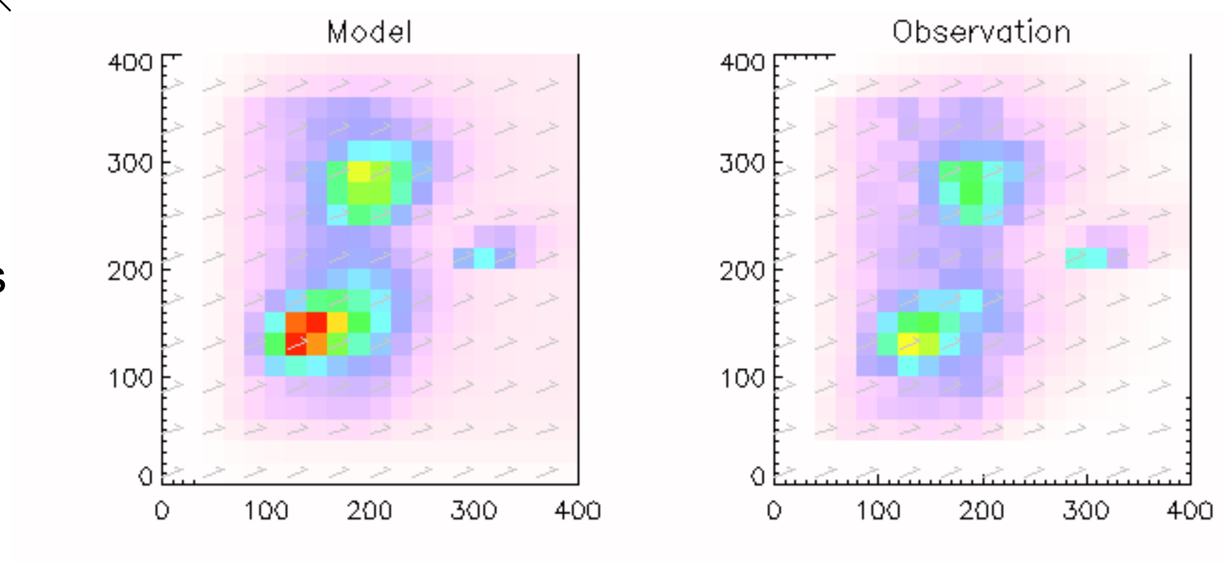


Local, linear

emissions



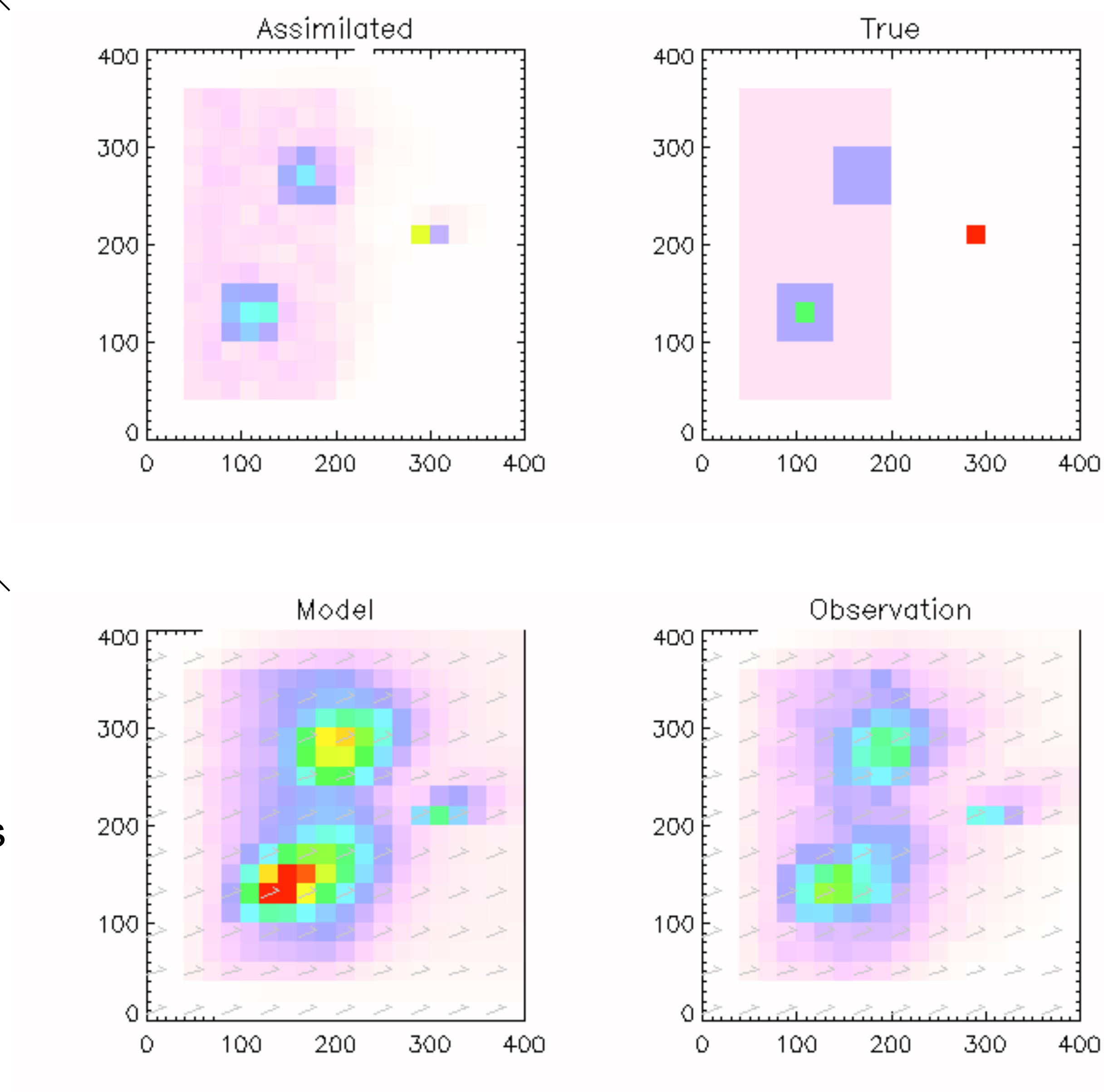
concentrations



Local, linear

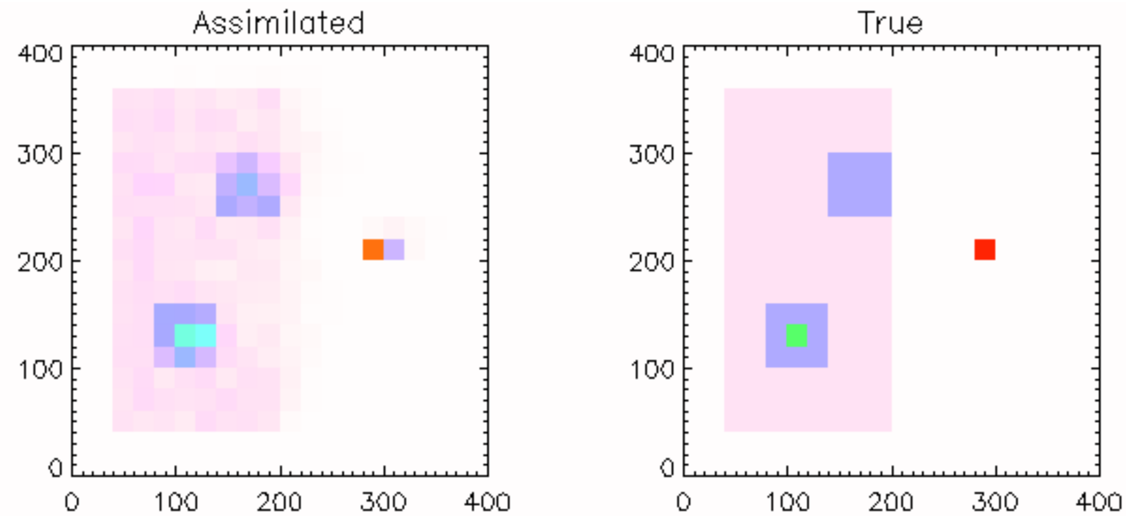
emissions

concentrations

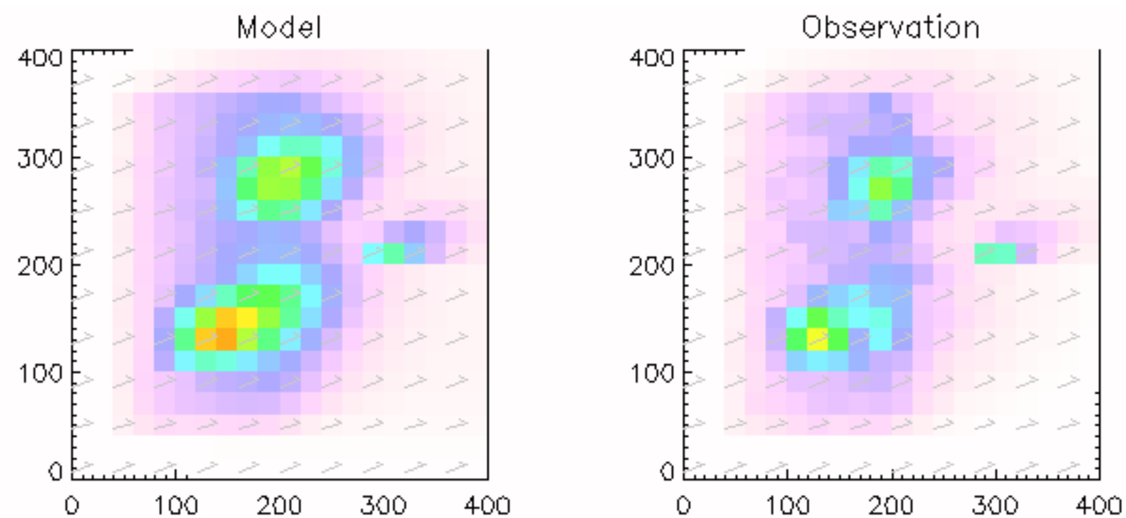


Local, linear

emissions

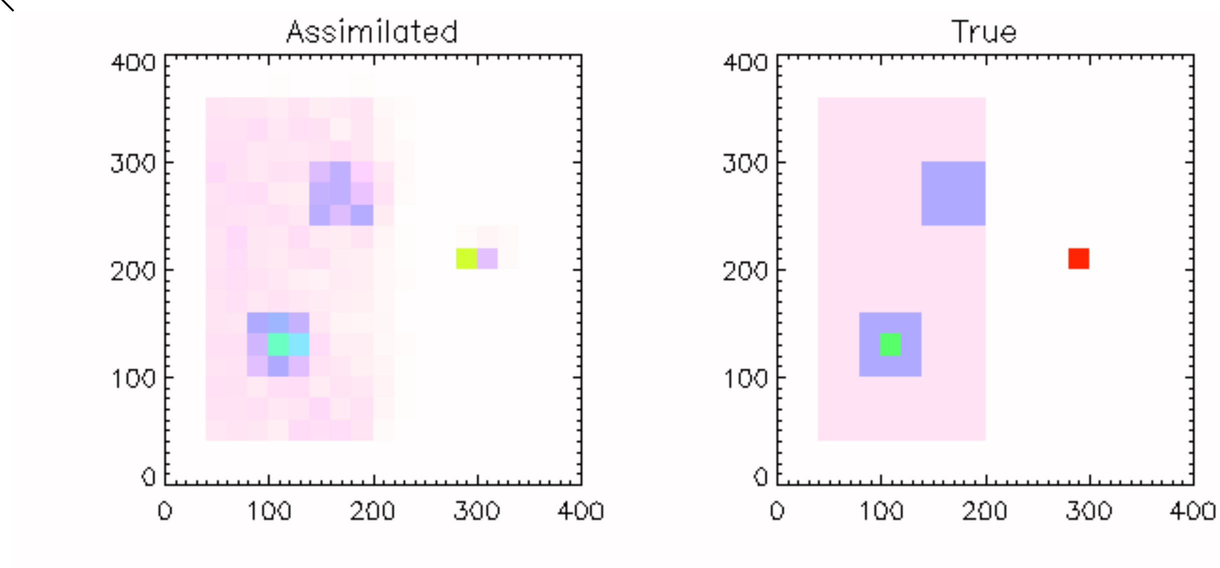


concentrations

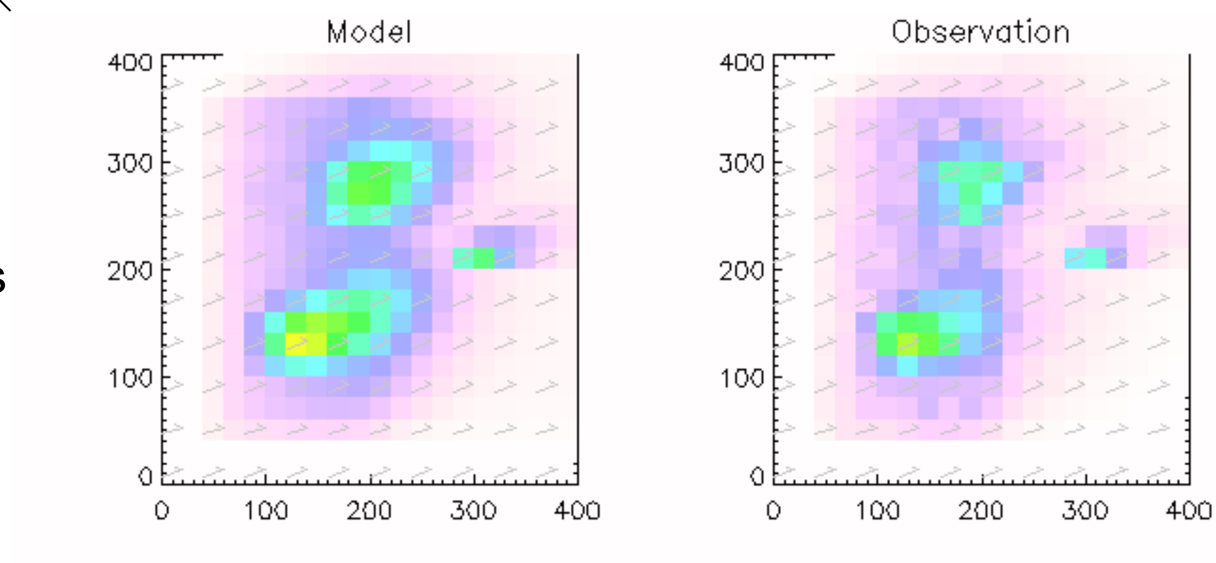


Local, linear

emissions

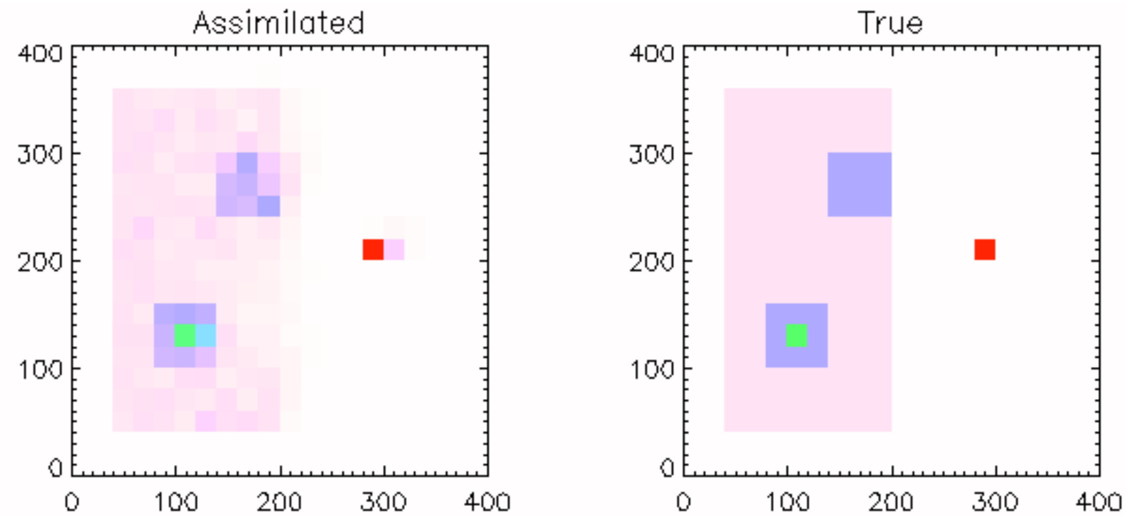


concentrations

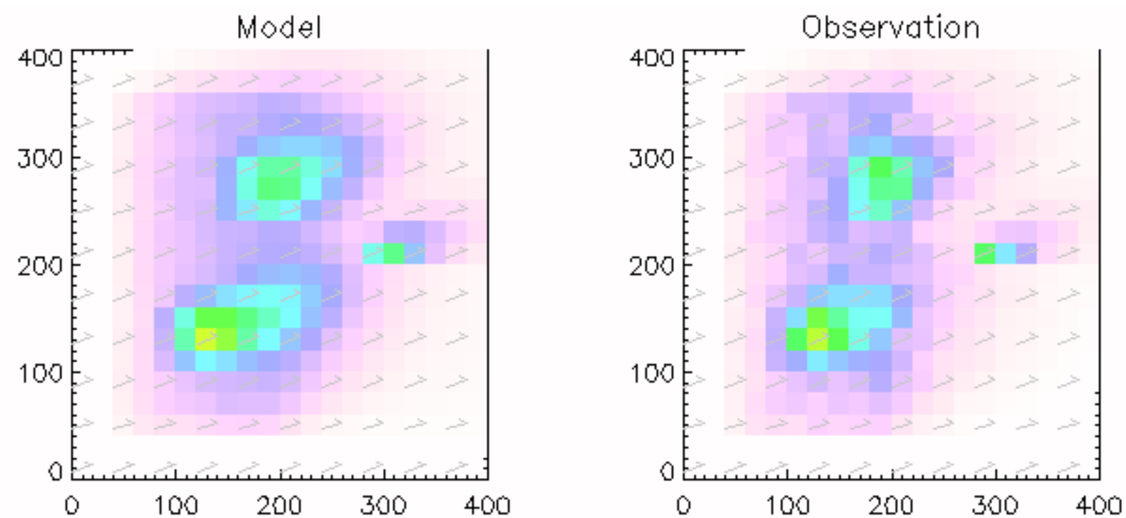


Local, linear

emissions

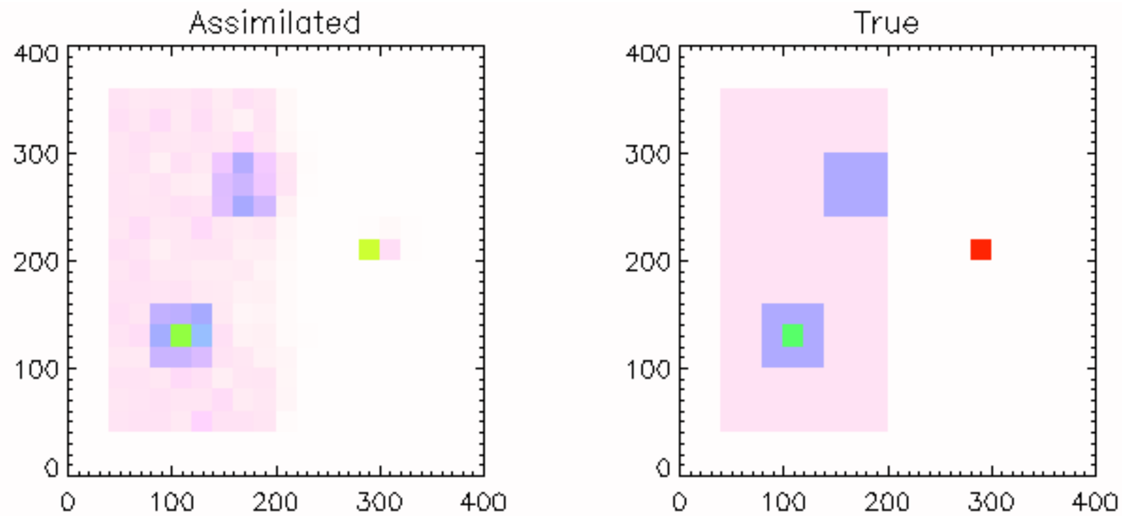


concentrations

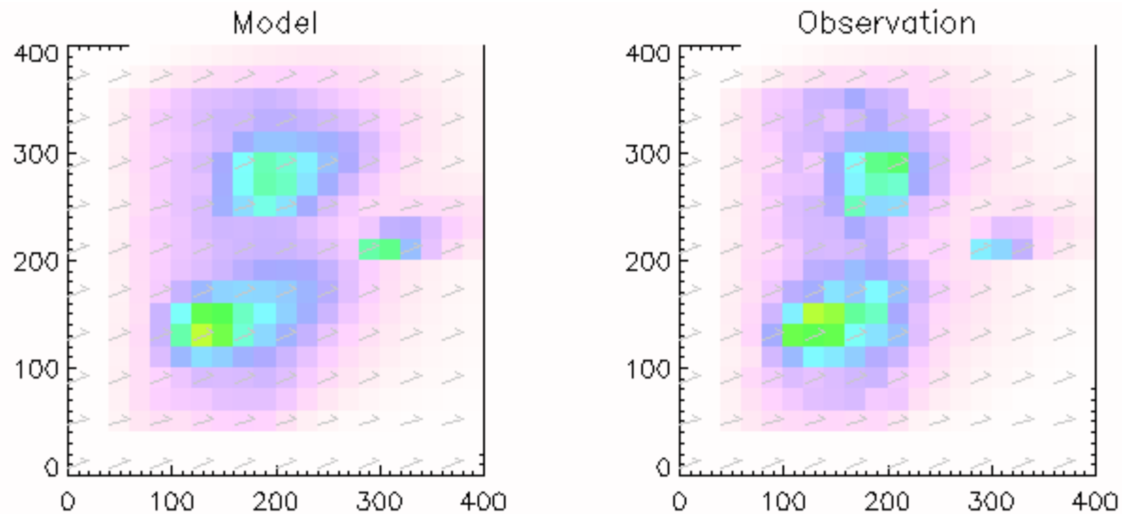


Local, linear

emissions

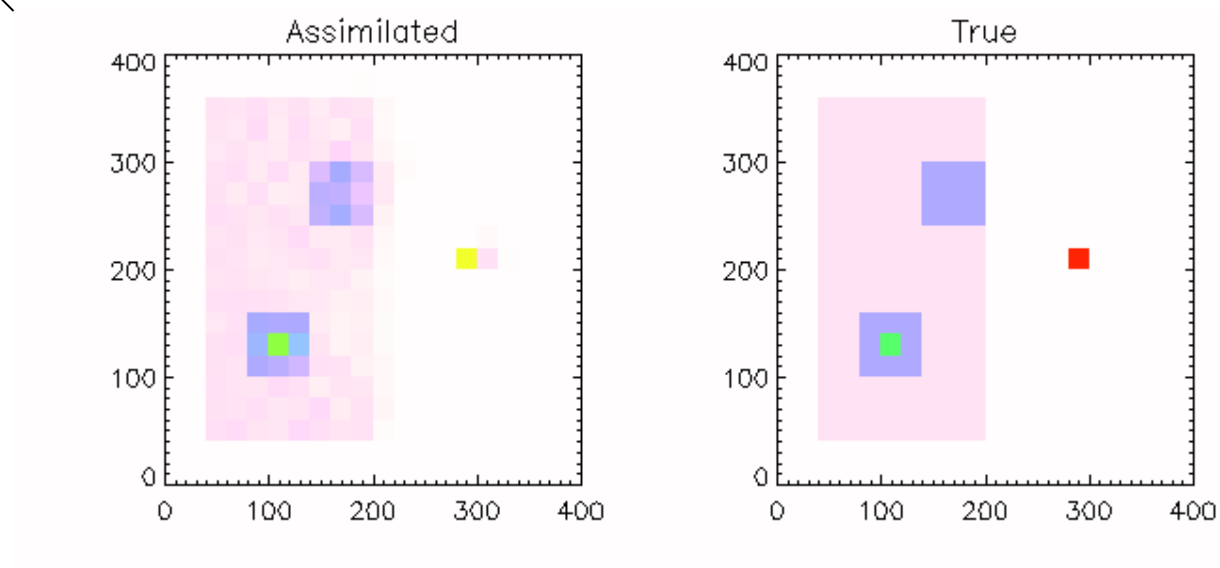


concentrations

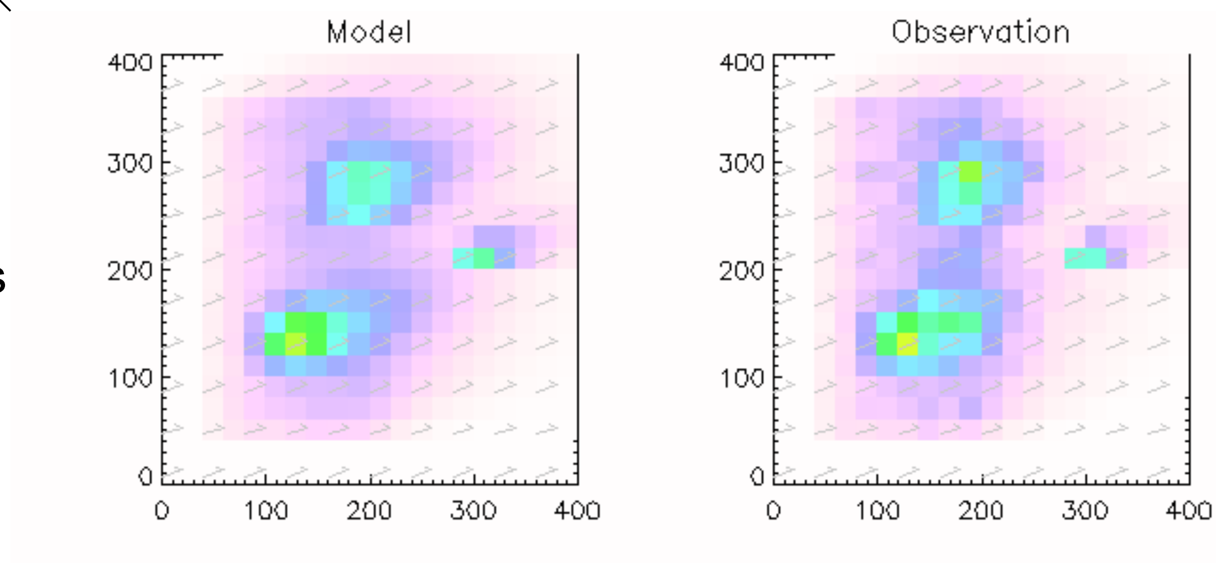


Local, linear

emissions

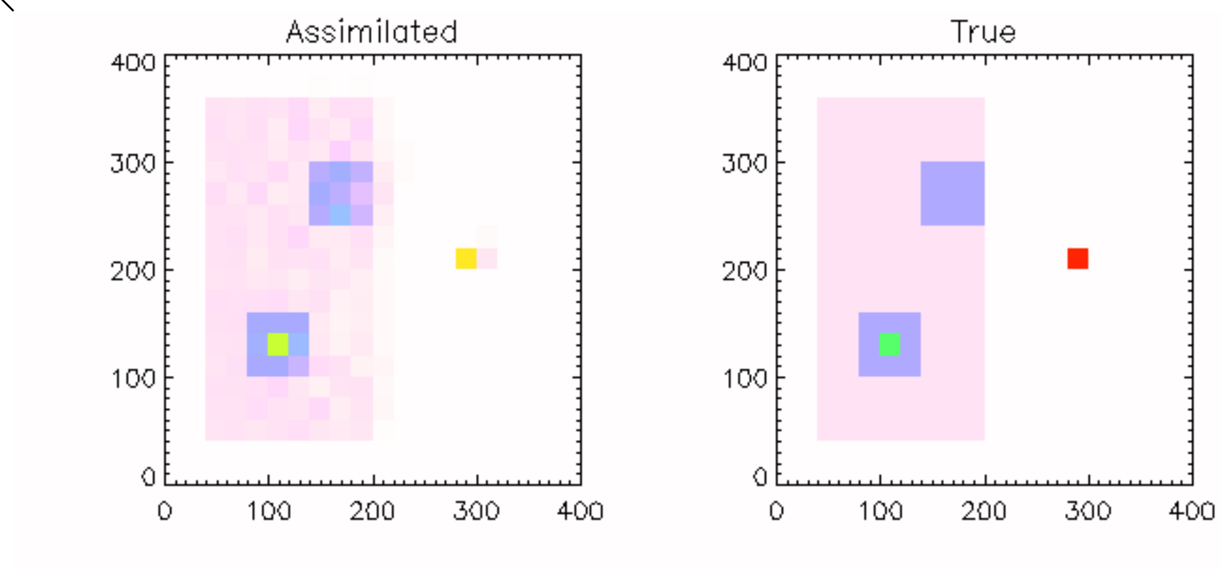


concentrations

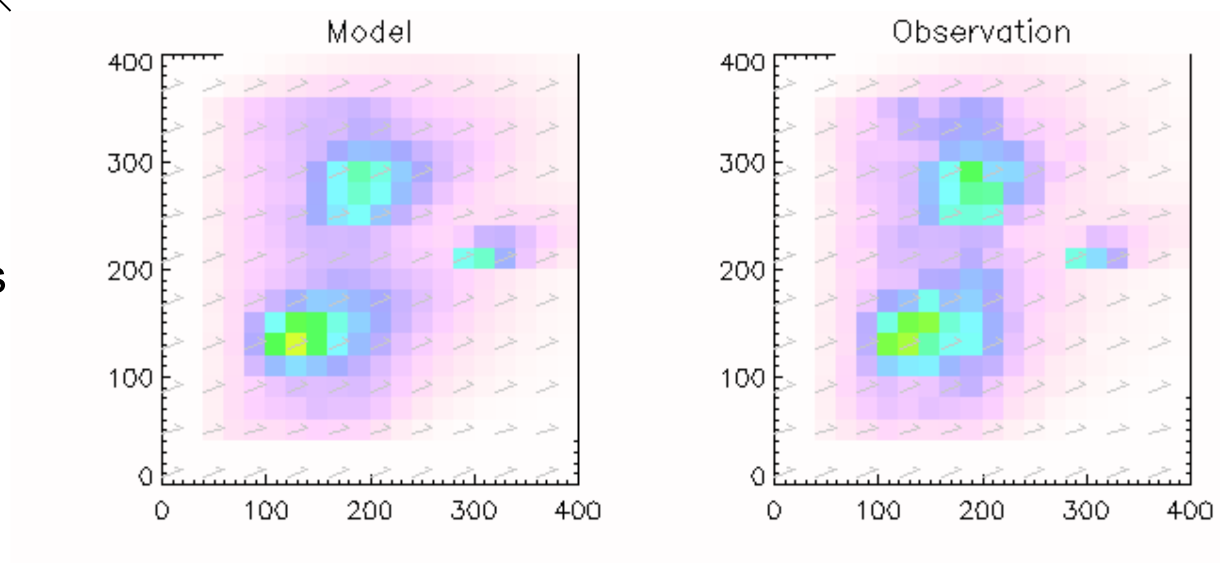


Local, linear

emissions



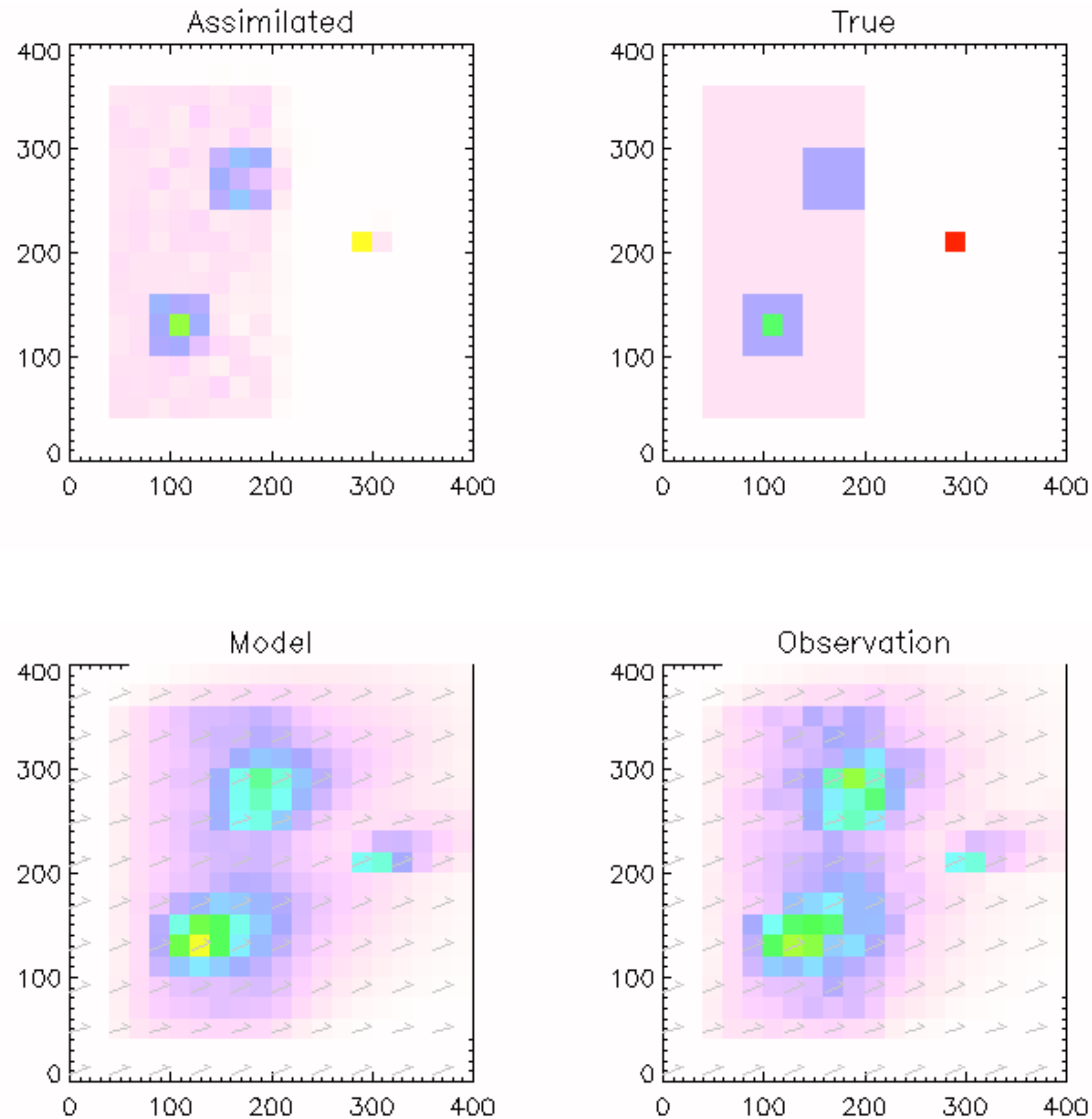
concentrations



Local, linear

emissions

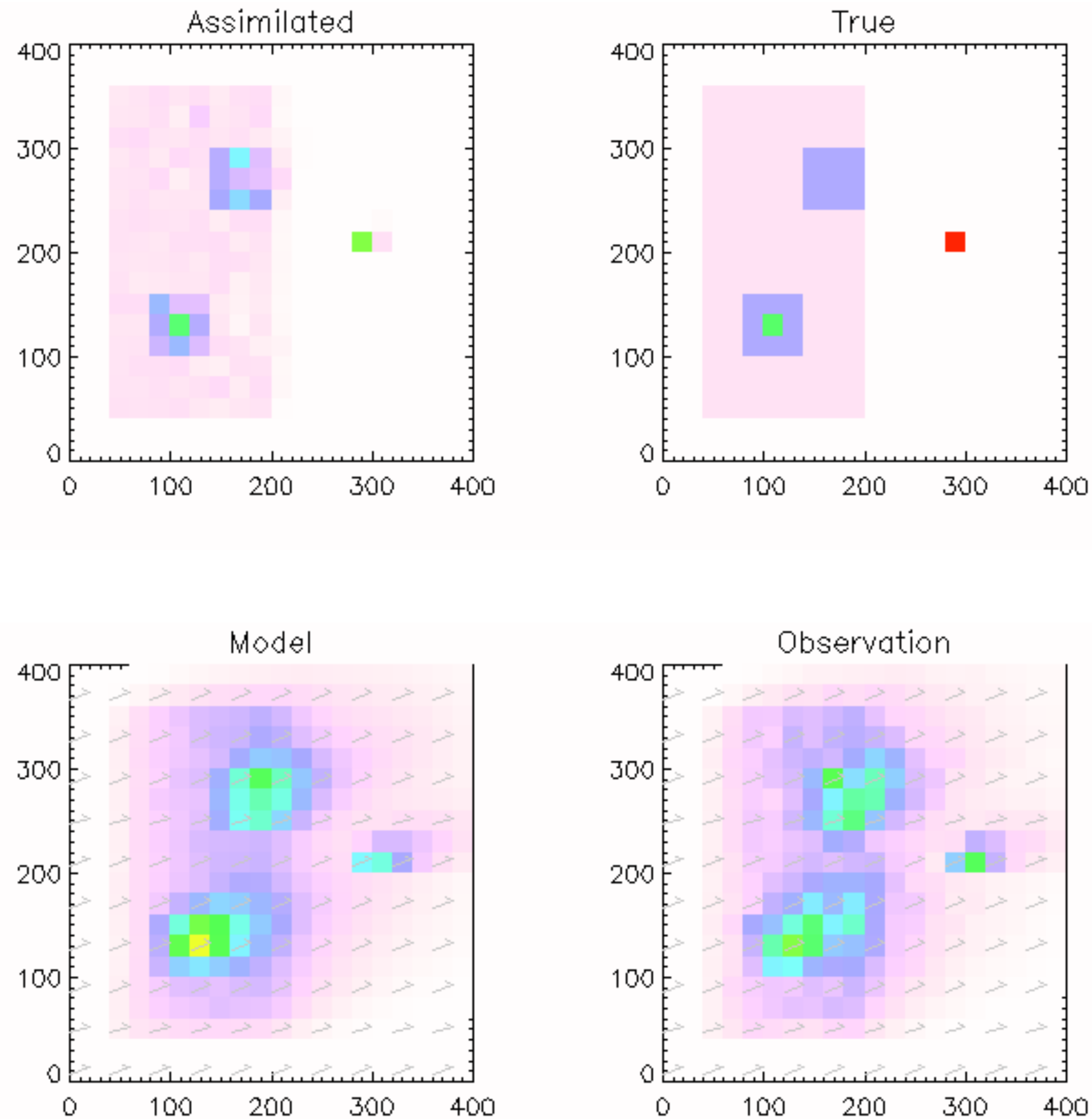
concentrations



Local, linear

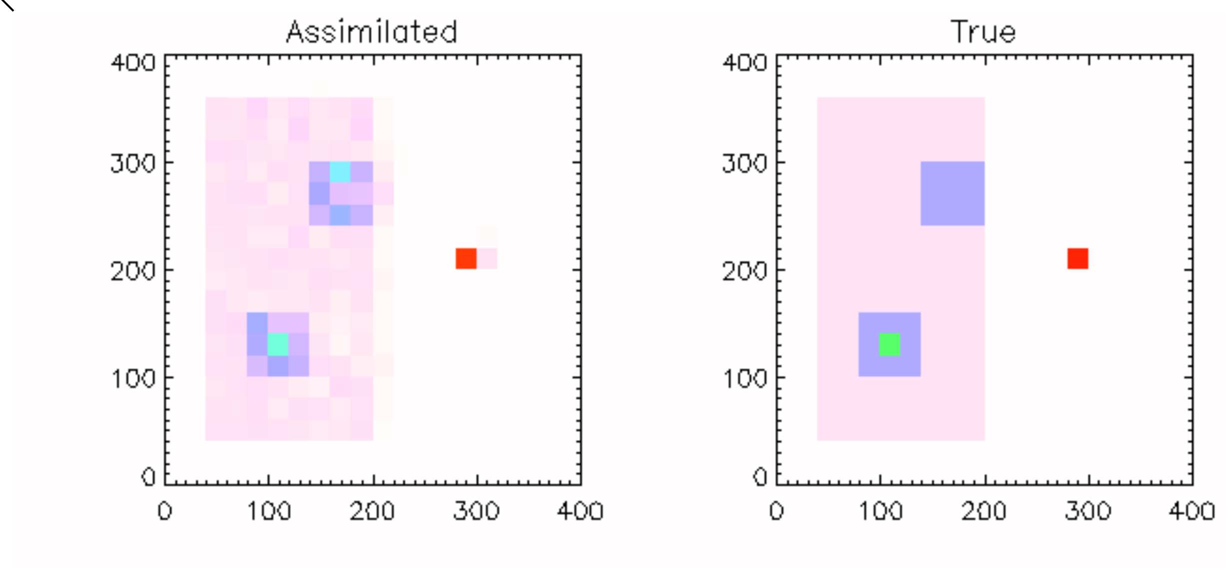
emissions

concentrations

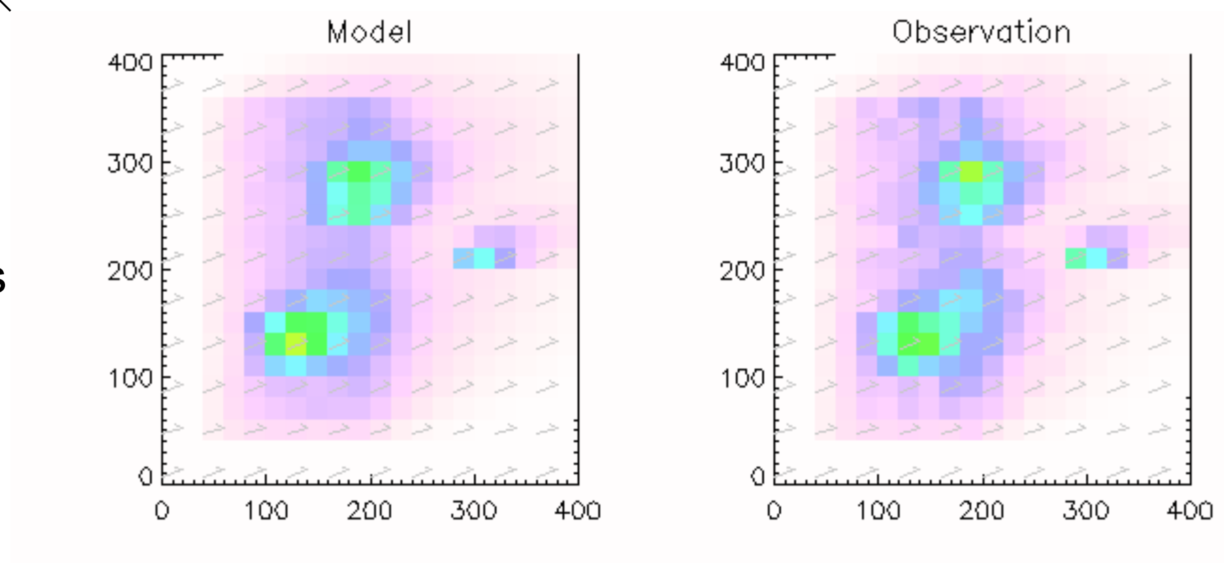


Local, linear

emissions

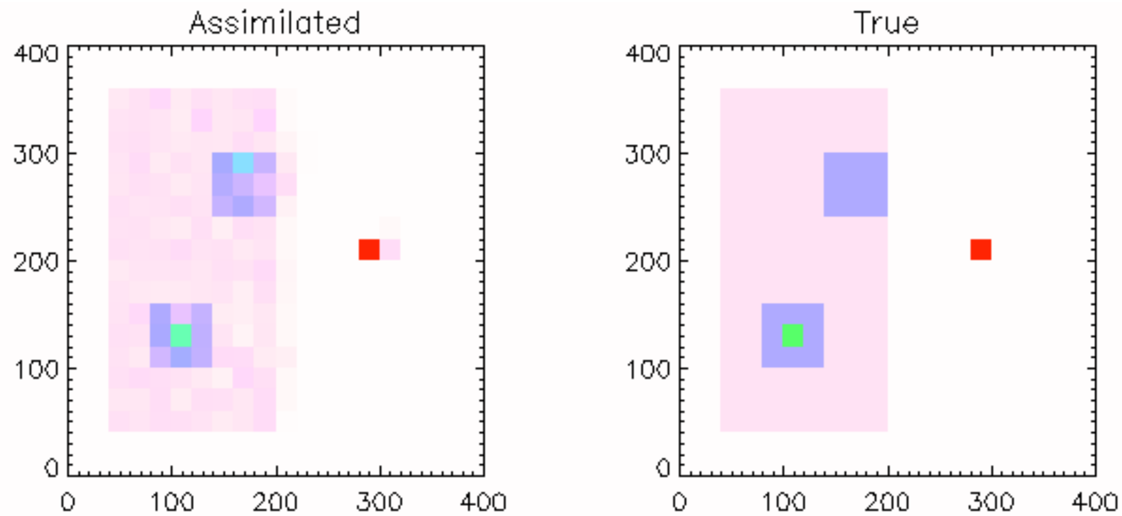


concentrations

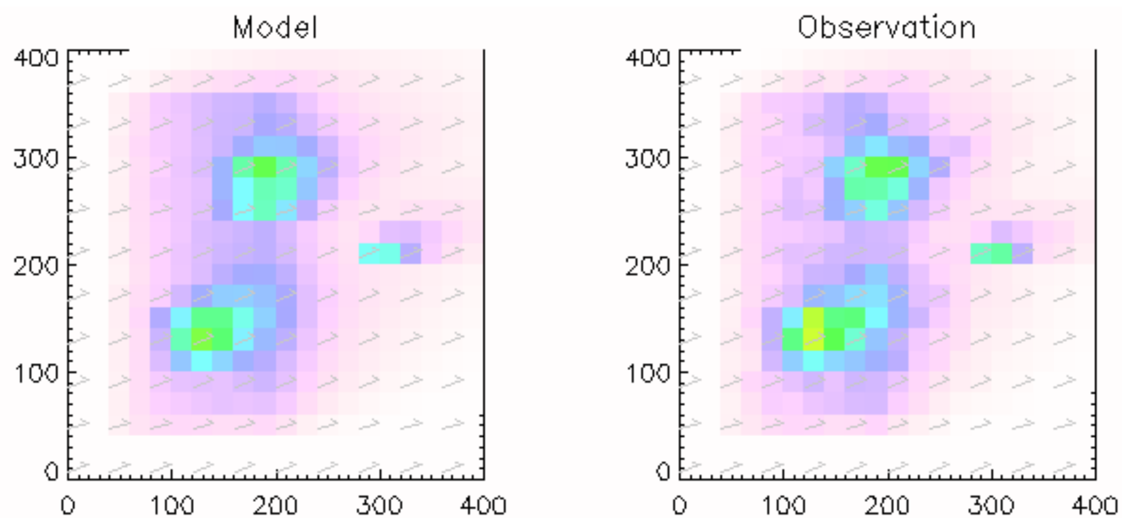


Local, linear

emissions

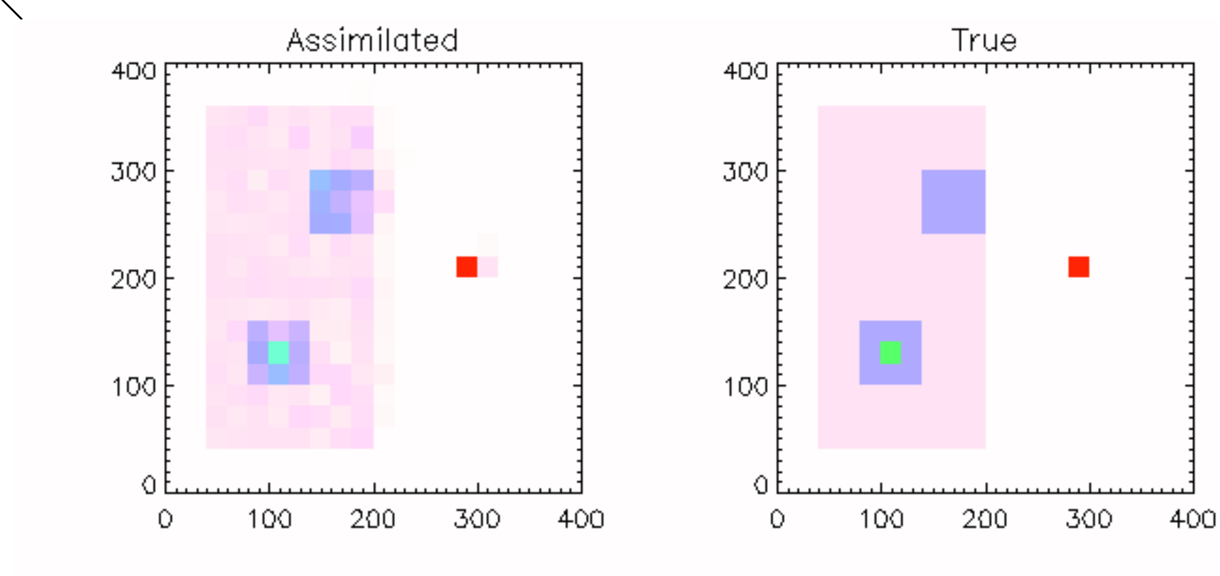


concentrations

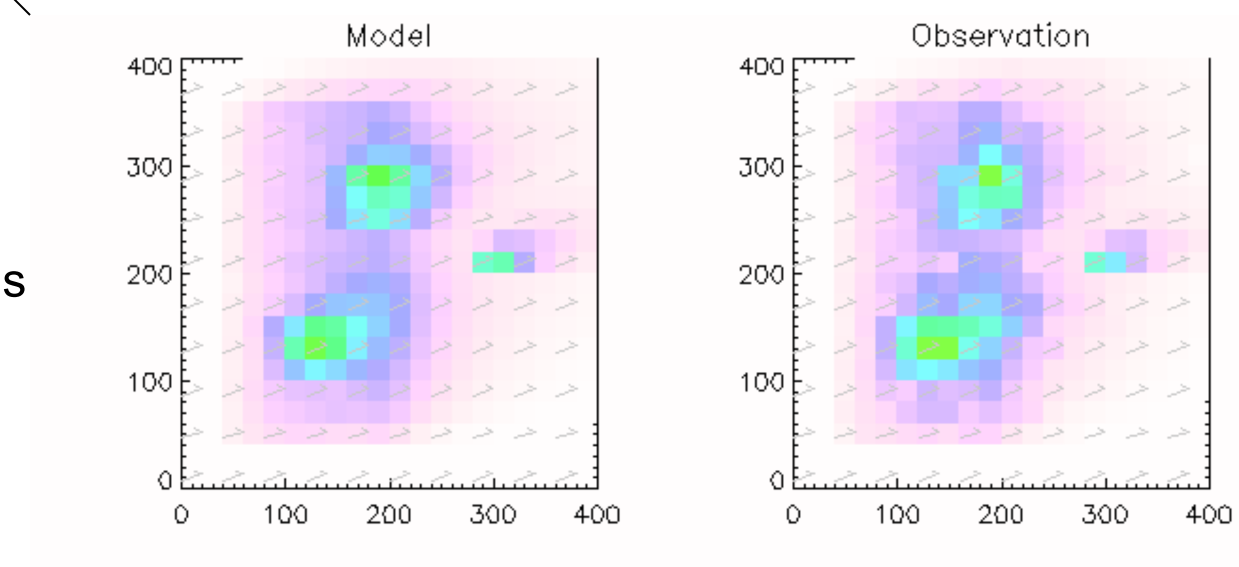


Local, linear

emissions

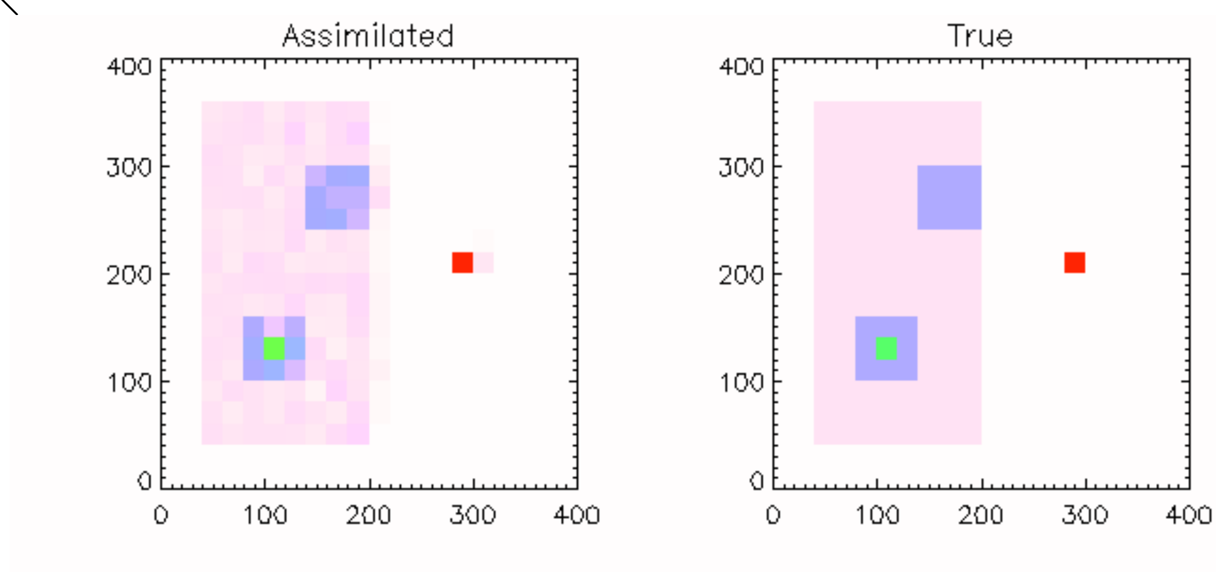


concentrations

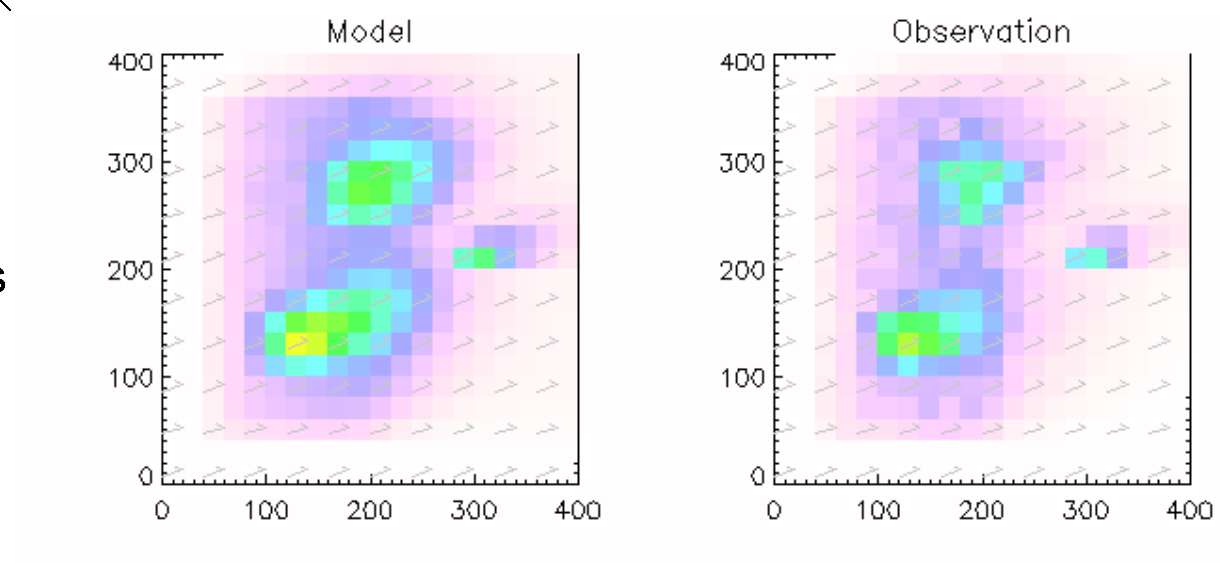


Local, linear

emissions

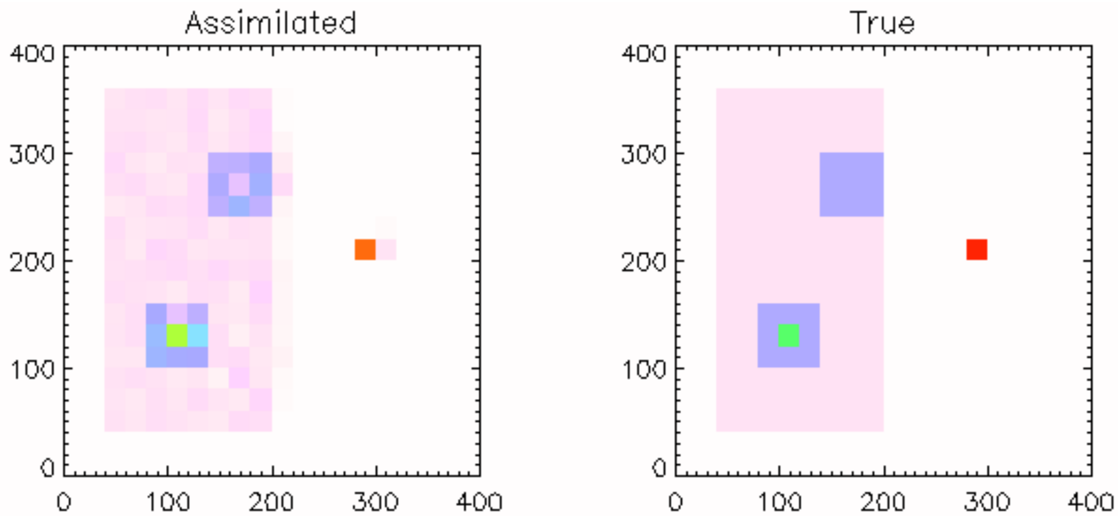


concentrations

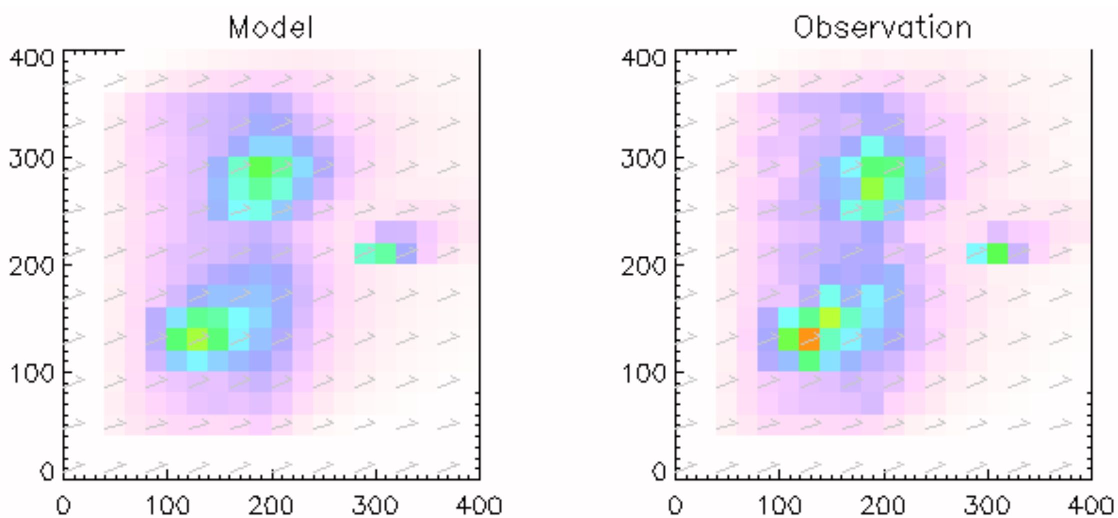


Local, linear

emissions



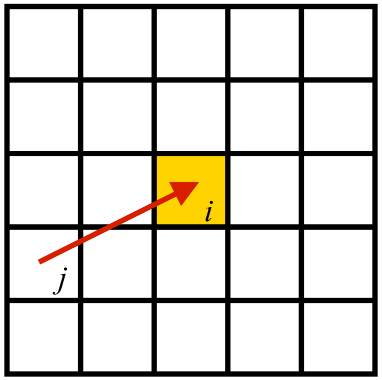
concentrations



Local, linear

More realistic inversion: Sensitivities

When transport is taken into account, emissions in all grid cells can contribute to the observed concentration:

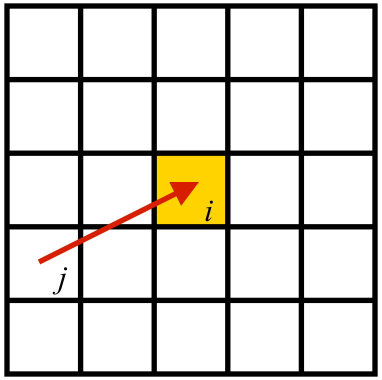


$$\Delta\Omega_i = \sum_j \alpha_{j\to i}\Delta E_j \quad , \quad \alpha_{j\to i} = \frac{\partial\Omega_i}{\partial E_j}$$

Monte Carlo method

Konovalov et al. (2006), Inverse modeling of NOx emission on a continental scale (2006), ACP

Perform model runs with random perturbations on the *a priori* emissions to get a set of linear equations from which the sensitivities can be solved:



$$\Delta\Omega_1^i = \sum_j \alpha_{j\to i}\Delta E_1^j$$

$$\Delta\Omega_2^i = \sum_j \alpha_{j\to i}\Delta E_2^j$$

$$\ldots$$

$$\Delta\Omega_N^i = \sum_j \alpha_{j\to i}\Delta E_N^j$$

The optimal number of random model runs depends on the desired accuracy.

For two next neighbours: N = 100

Monte Carlo method

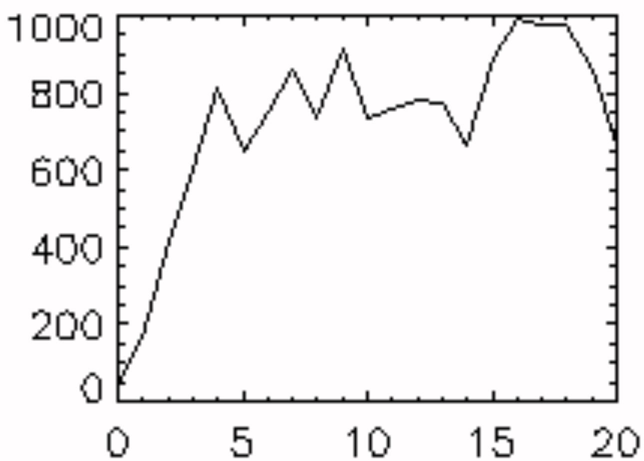
Advantages

- Takes transport of nearest neighbours into account

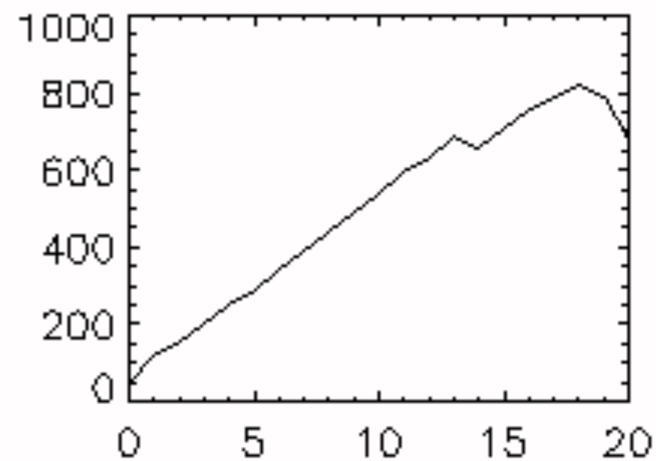
Disadvantages

- Time consuming calculations:
  ~100 model runs needed to solve transport from 2 nearest neighbours.

Convergence behaviour



Local, linear

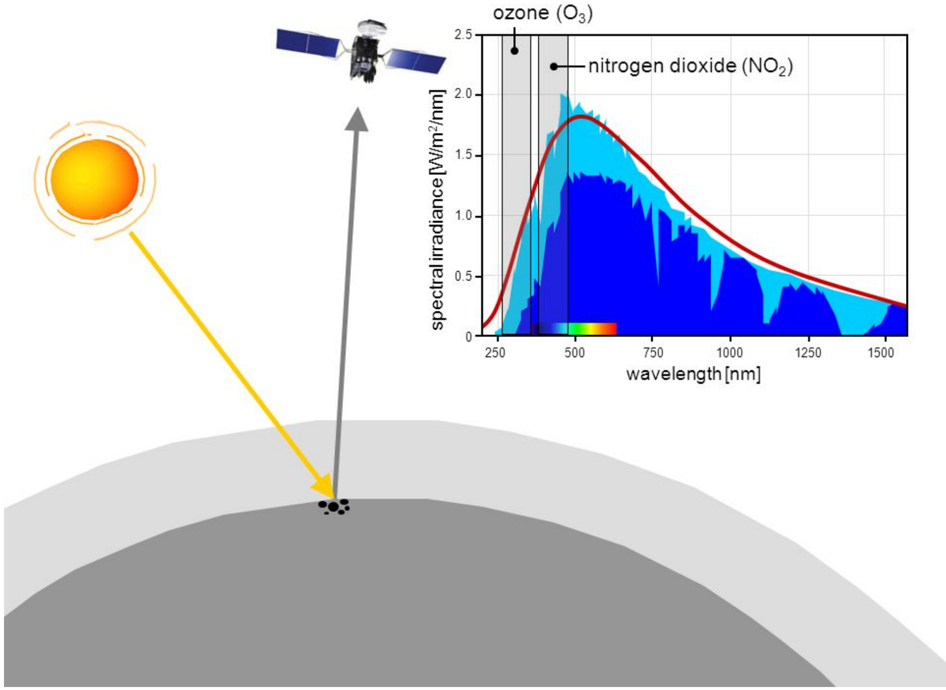


Kalman

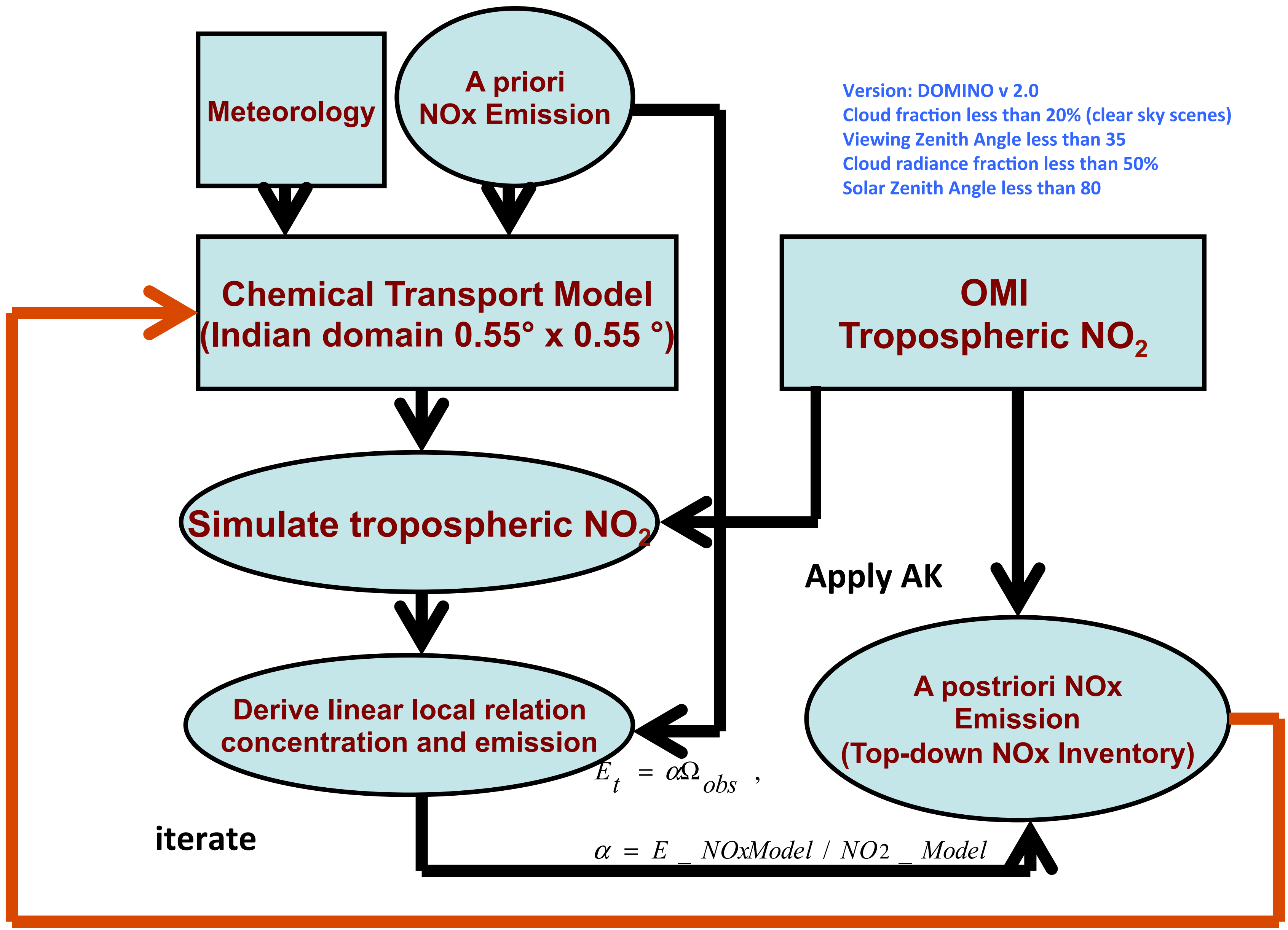
Top-Down Approach for NOx- emission over India

- **What we Need?**
- Satellite observations of NO_2
- Chemical Transport Model

Tropospheric NO_2 column is derived in three main steps involving the calculation of
(1) slant column (using Differential Optical Absorption Spectroscopy (DOAS) approach in the 405–465 nm spectral
window),
(2) tropospheric slant column (using modeling/ assimilation approach),
(3) tropospheric vertical column (using air mass factor—AMF).



Top-Down Approach for NOx- emission over India



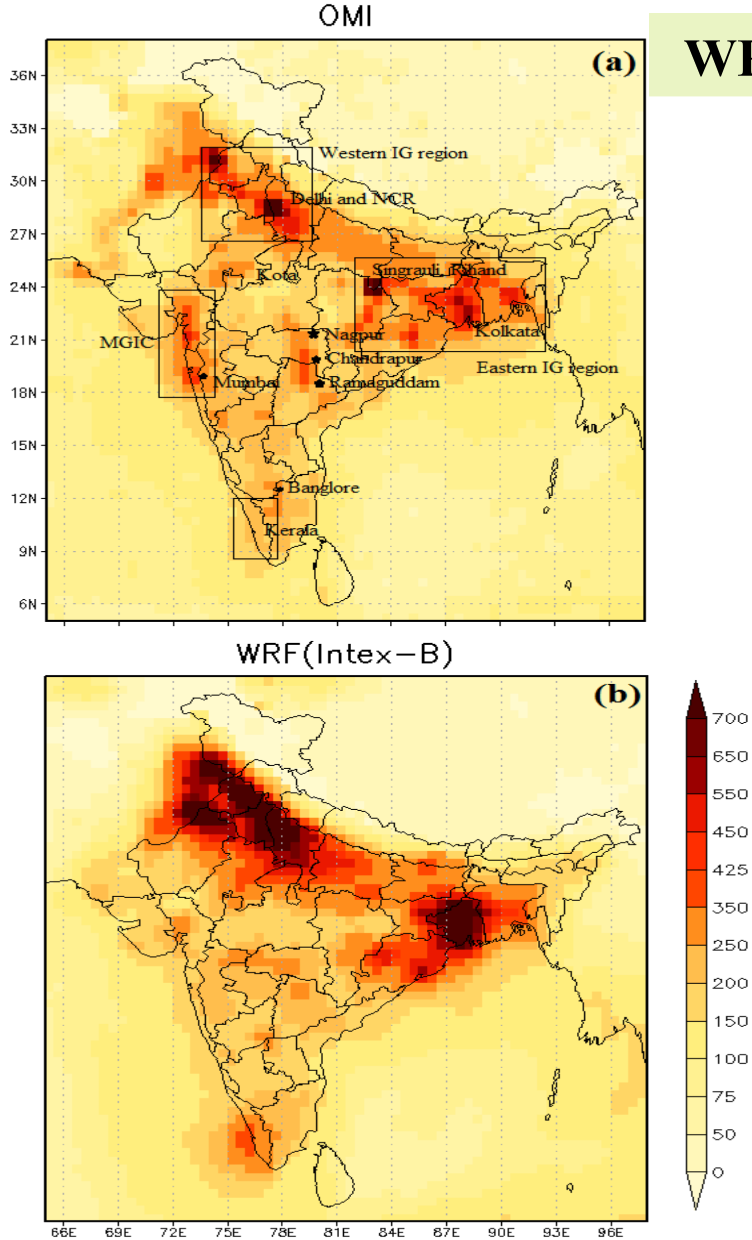
OMI Tropospheric NO_2 columns for the year 2005 Over India

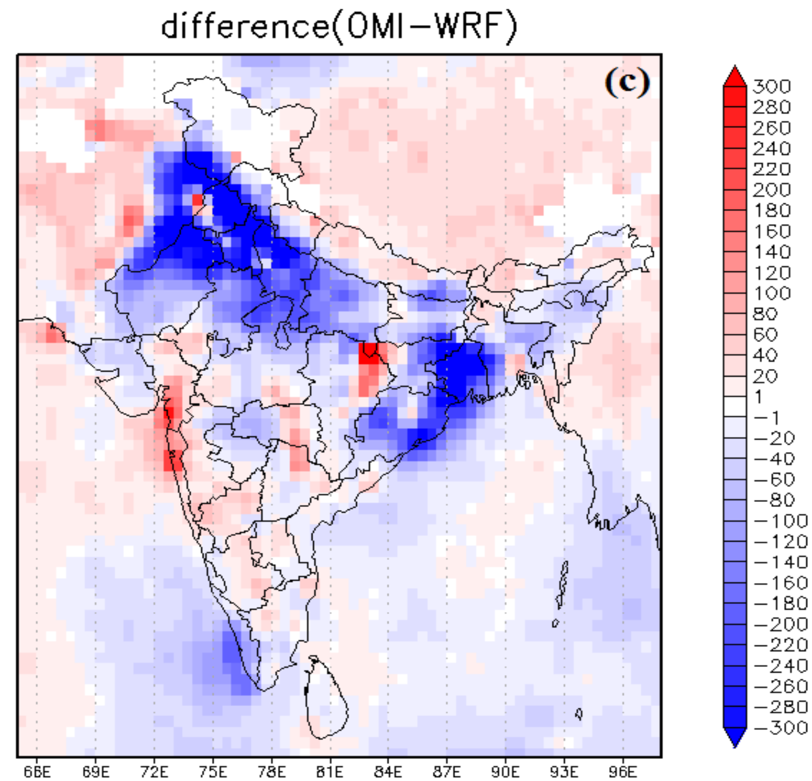
Version: DOMINO v 2.0 (Level 2 data set)
Cloud fraction less than 20% (clear sky scenes)
Viewing Zenith Angle less than 35 (pixel size smaller than $34x14km^2$)
Cloud radiance fraction less than 50%
Solar Zenith Angle less than 80

WRF-Chem Simulation for Jan 2005

Domain	: South Asia (0 - 45° N, 55 -110 ° E)
Period	: 2005
Resolution	: 55 km x 55 km
Emissions	: INTEX-B (A Priori)
Fire Emission	: NCAR Fire Inventory (FINN)
Biogenic	: MEGAN
Gas Ph. Chem	: MOZART
Aerosol Ph. Chem	: GOCART

WRF simulation with *a priori* emission (Intex-B)





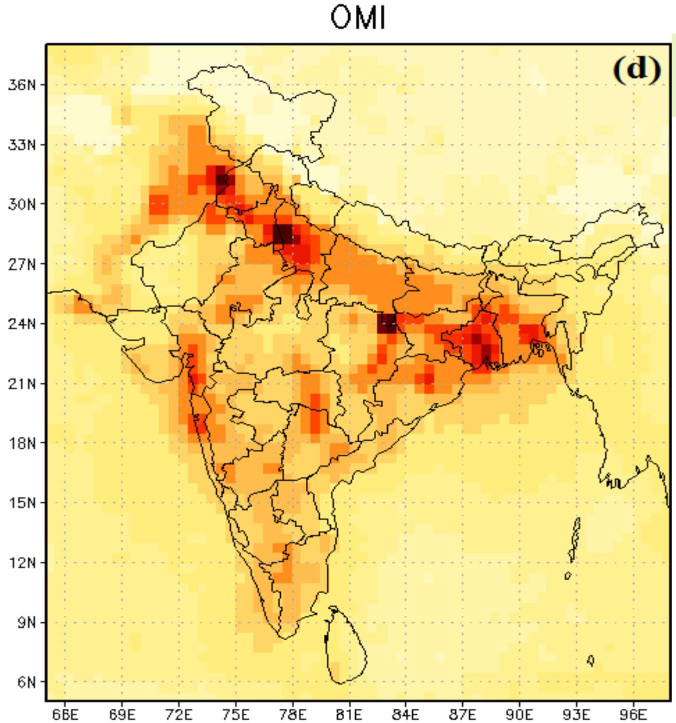
With OMI Averaging Kernel ←

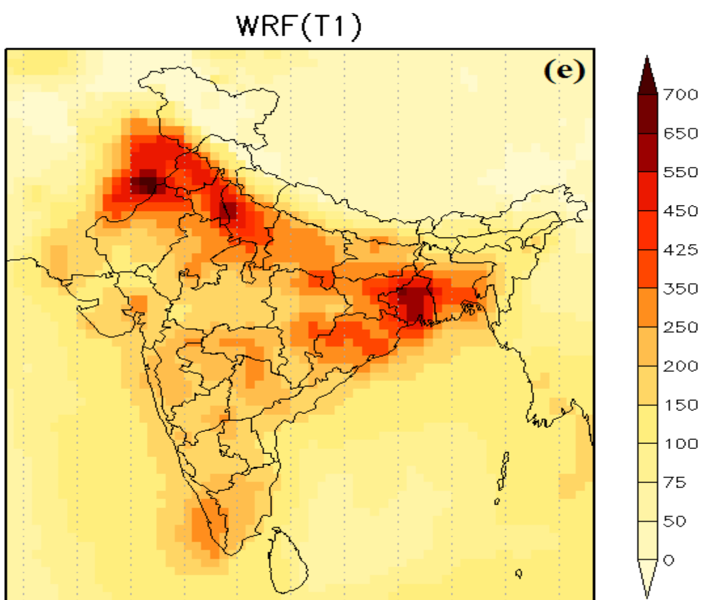
Correlation $R^2 = 0.68$

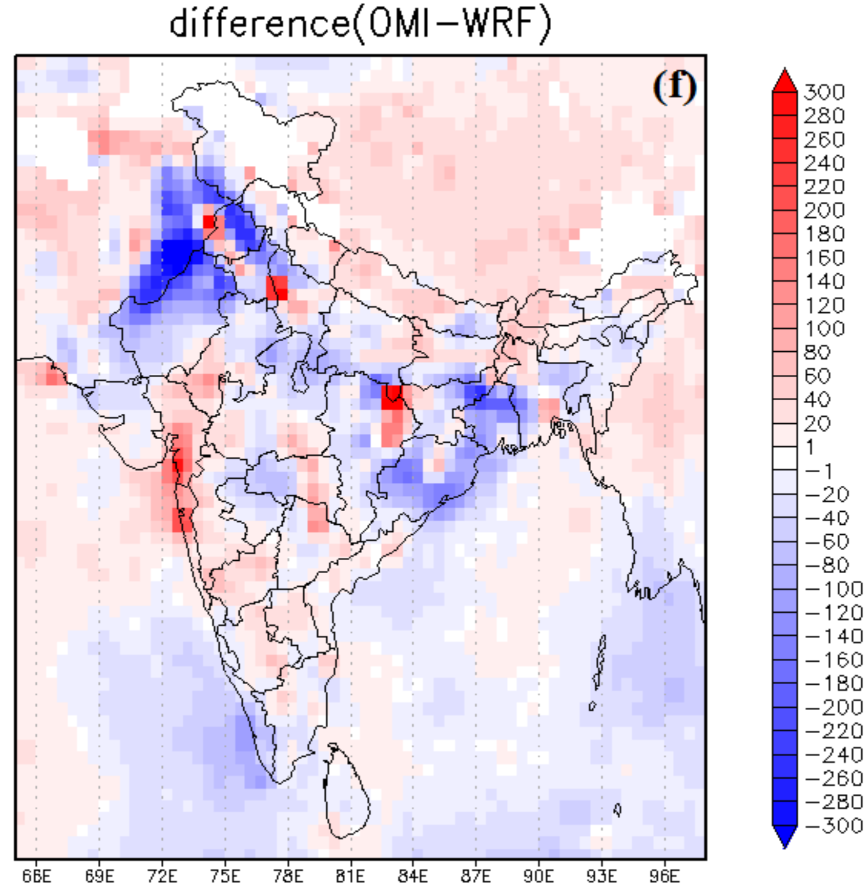
Spatial distribution of OMI and WRF-Chem tropospheric NO_2 column for 2005 and its difference. Tropospheric column NO_2 unit is 10^{13} molecules/cm^2/s.

Ghude S.Det al., GRL, 2013c

WRF simulation with *a postrioro* emission (T1)





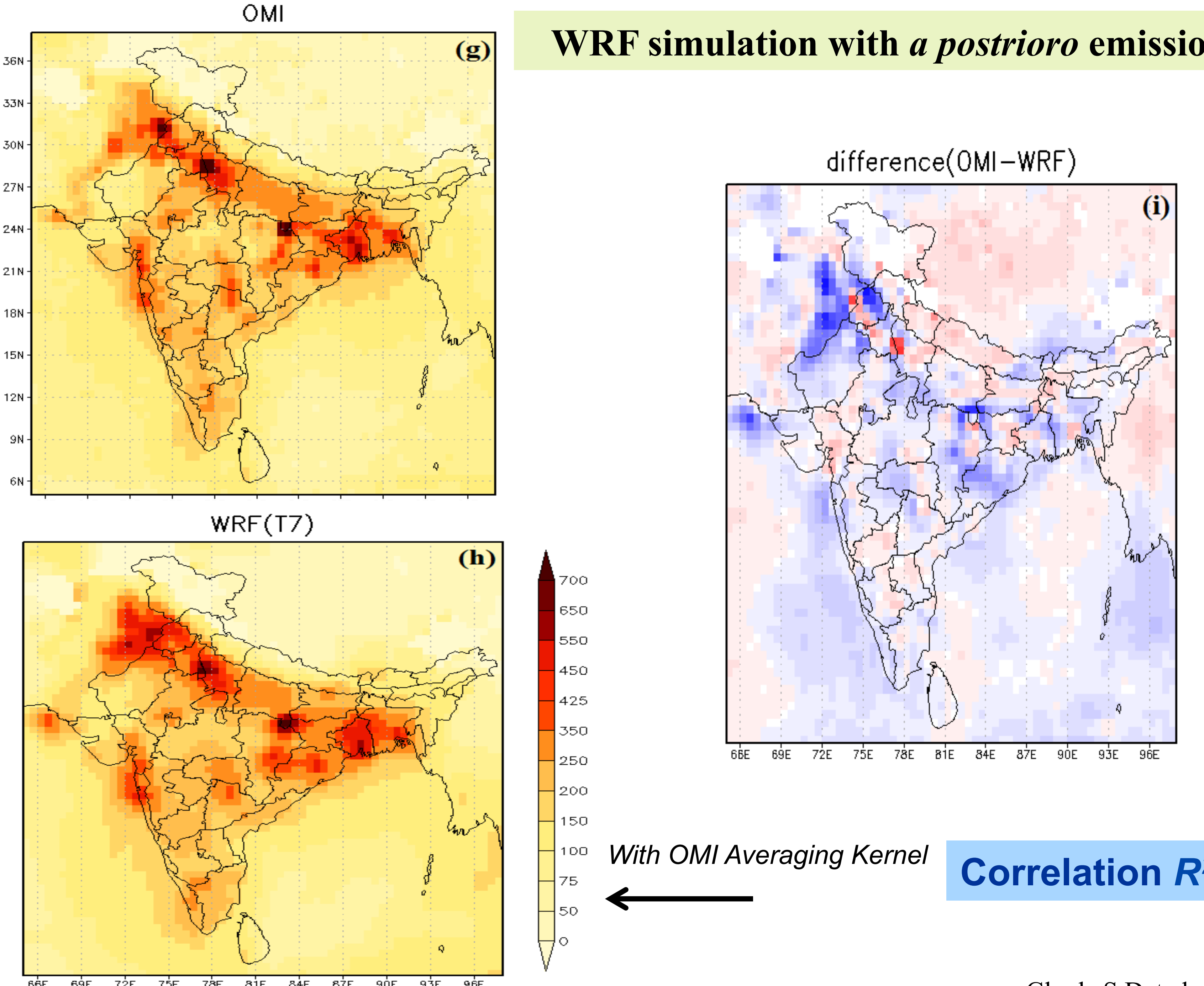


With OMI Averaging Kernel ←

Correlation R^2 = 0.76

Ghude S.Det al., GRL, 2013c

WRF simulation with *a postrioro* emission (T7)

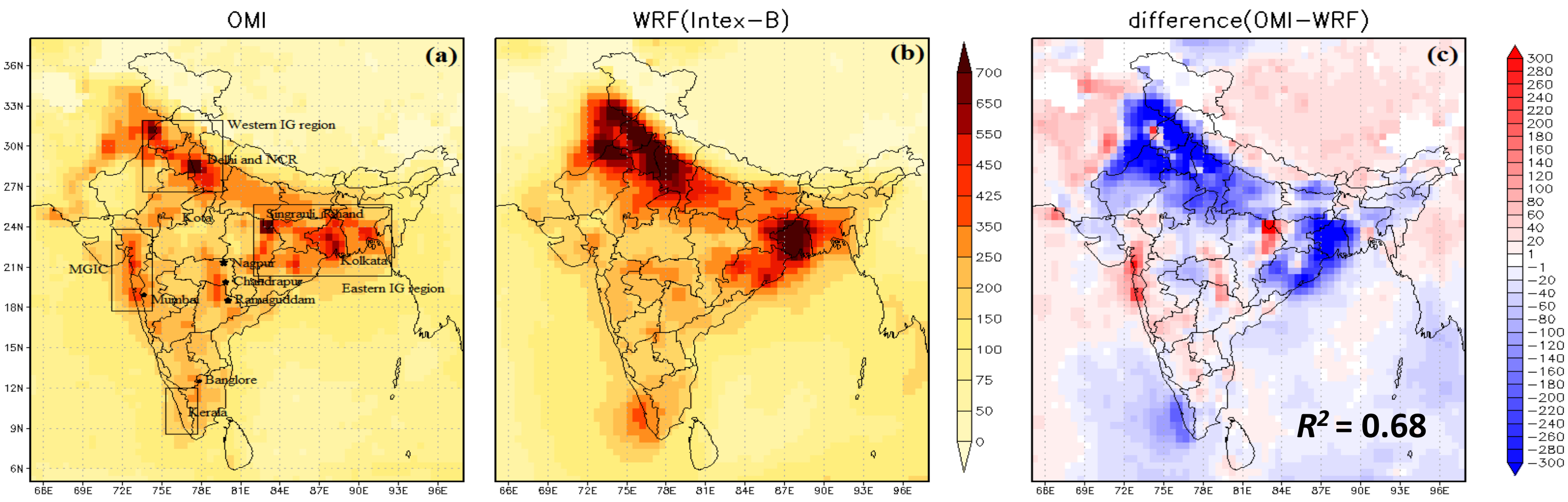


With OMI Averaging Kernel

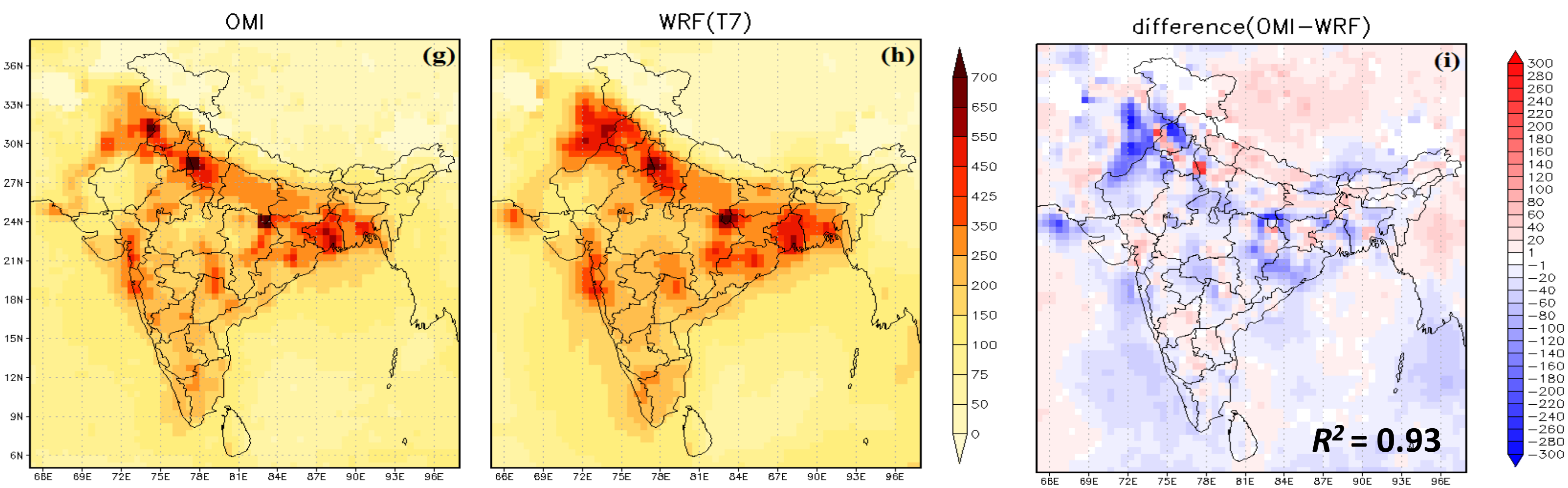
Correlation R^2 = 0.93

Ghude S.Det al., GRL, 2013

WRF simulation with *a priori* emission (Intex-B)

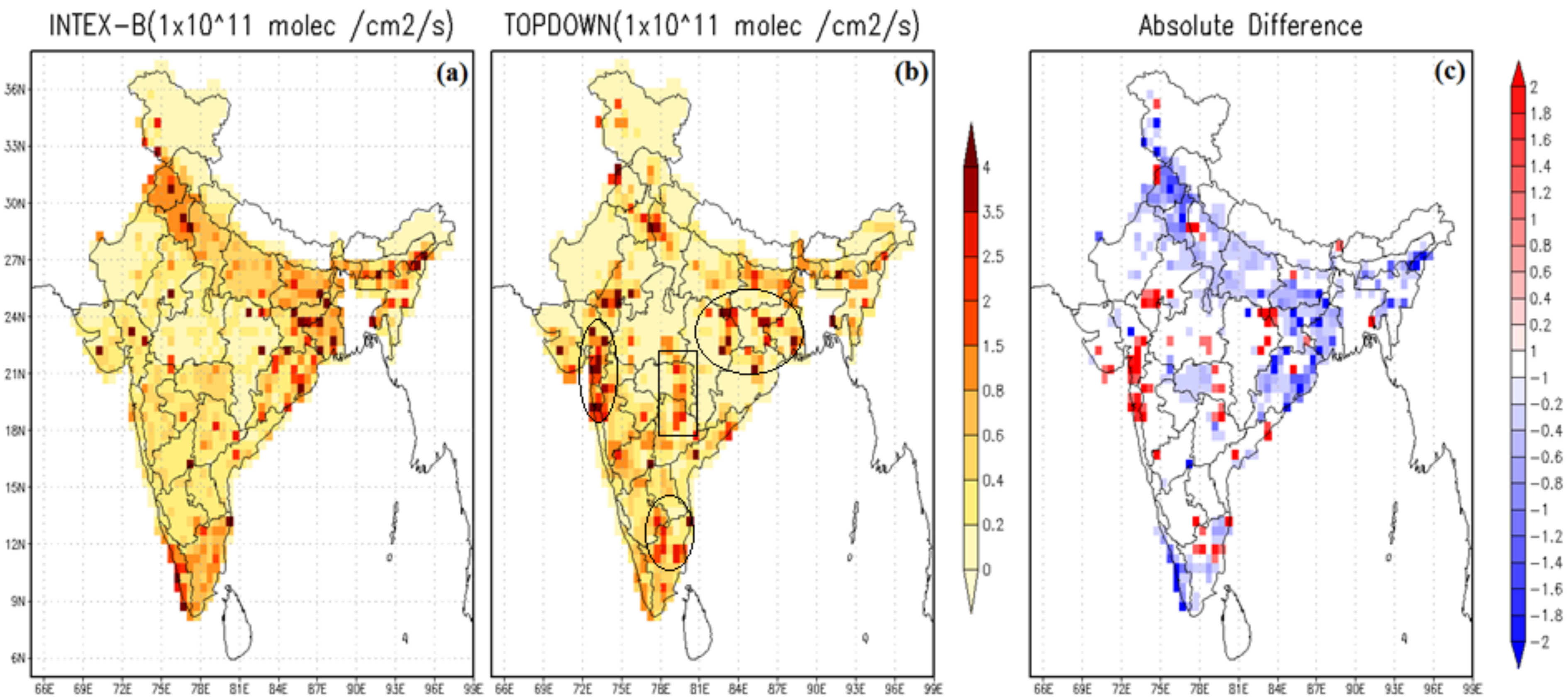


WRF simulation with *a postrioro* emission (T7)



Ghude S.Det al., GRL, 2013

A priori versus A posteriori Emissions

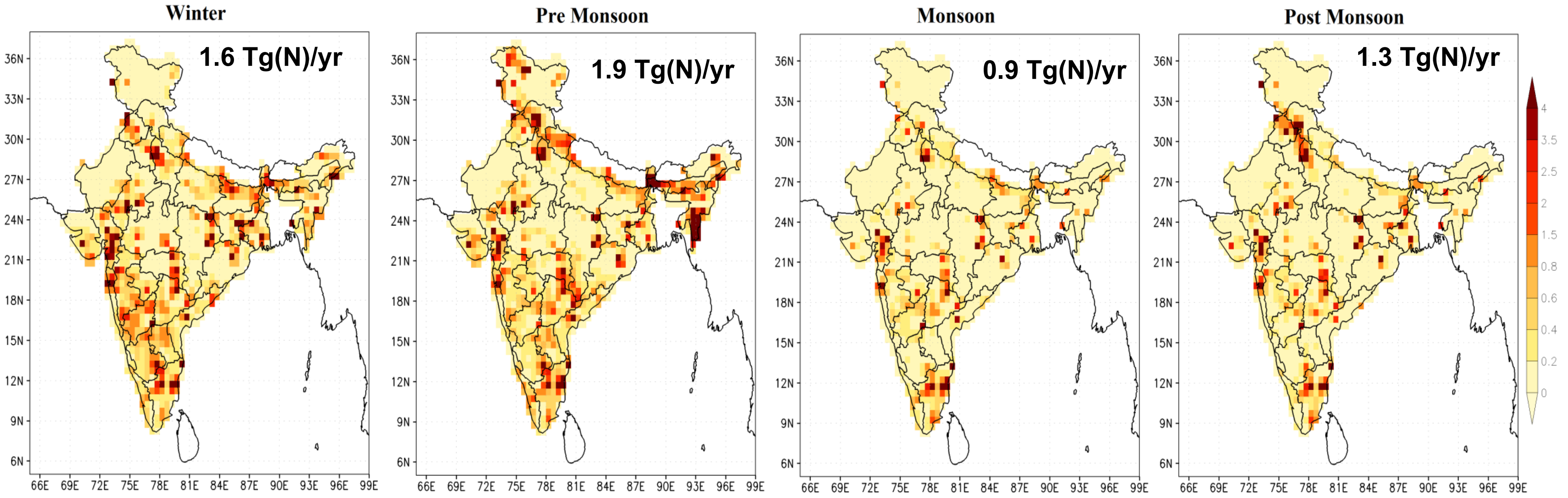


Spatial distribution of NO_X emission from (a) INTEX-B inventory, (b) optimized top-down Inventory and (c) their difference (emission unit is 10^{11} NO molecules/cm^2/s)

A priori emission	A posteriori emission
1.56Tg N /year	1.42Tg N /year

Ghude S.Det al., GRL, 2013c

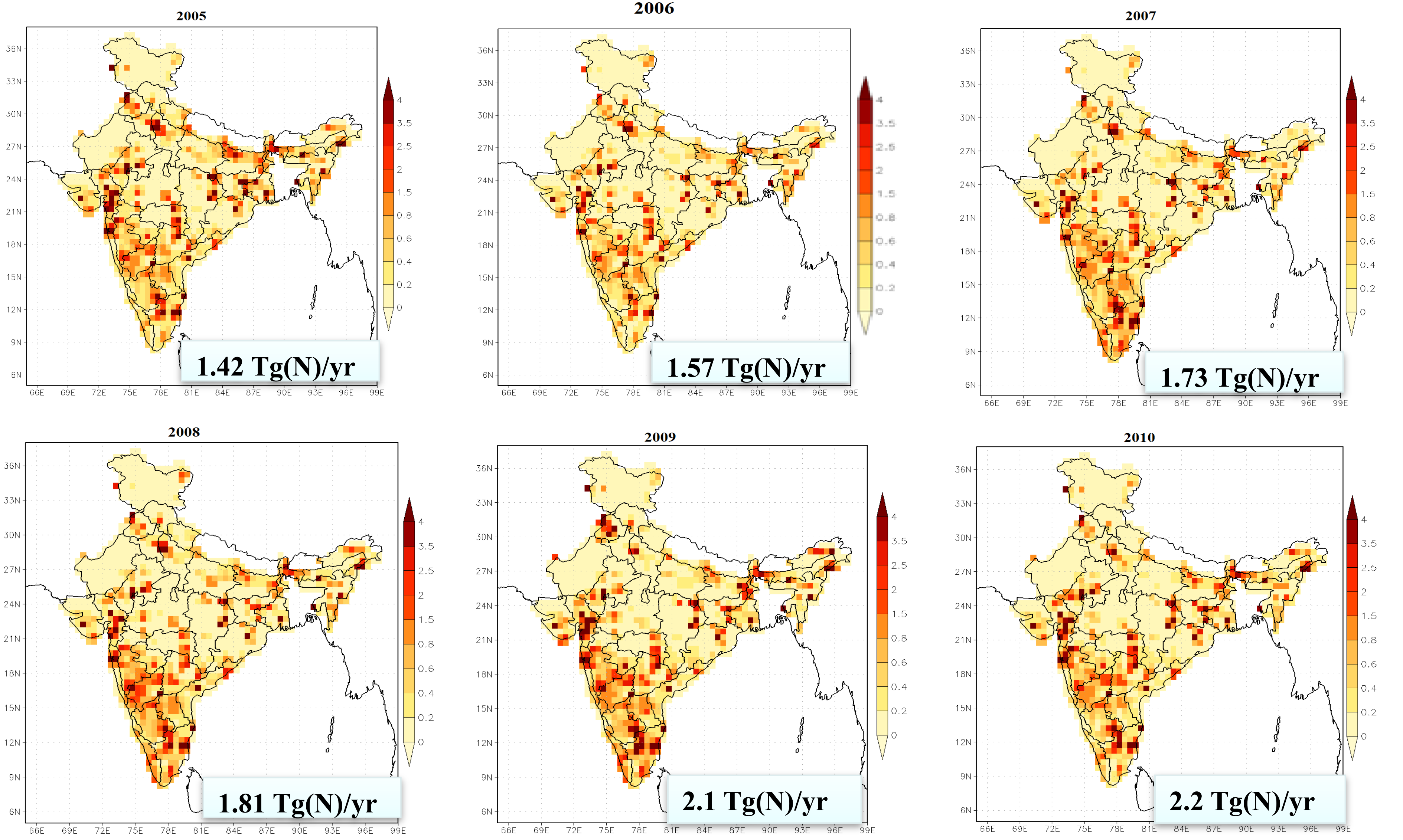
Seasonal Top-down Emission



Spatial distribution NO_X emissions from optimized Top-Down inventory (emission unit is 10^{11} NO molecules/cm^2/s) for winter, pre-monsoon, monsoon and post-monsoon seasons for 2005.

Jne et al., AE, 2014 (Under review)

Annual Top-down Emission



Spatial distribution of annual NO_X emissions from optimized Top-Down inventory (emission unit is 10^{11} NO molecules/cm^2/s) for 2005, 2006, 2007, 2008, 2009, 2010 years.

Sources of Uncertainties in Top-Down Inventory:

1. Uncertainty in satellite retrievals

OMI filtering error,
Air Mass Faction (clod fraction, cloud pressure, NO2 Profile shape, aerosols calculation, Stratospheric corrections, etc)

2. Model Uncertainties

- Error in model metrology
- Model profile shape
- Simulation of fate of Nitrogen emitted into the atmosphere
- (chemical mechanism and heterogeneous aerosol
- NOx reactions)
- dry and wet deposition

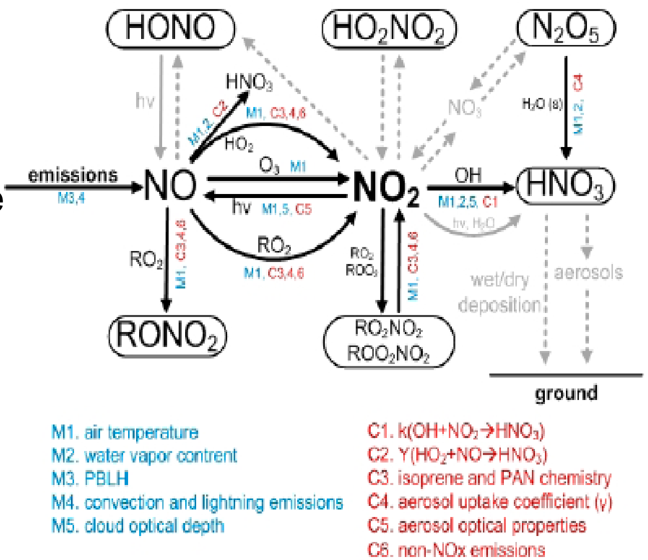


Fig. 1. Tropospheric chemistry involving NO_x and impacts of meteorological and chemical parameters evaluated in the present study. Processes shown in solid grey arrows are discussed without sensitivity simulations. Processes shown in dashed grey arrows are not discussed explicitly. Note that PBL mixing and convection affect vertical distributions of NO_x and related species. Heterogeneous uptake on aerosols depends on the amount of aerosol surfaces as well. Evaluation on the $RONO_2$ pathway is focused mainly on isoprene nitrates. Clouds and water vapor have indirect influences on radicals through effects on solar radiation.

Lin et al 2012

Conclusions

Every advantage has its disadvantage

Access to information on emissions:

The ECCAD Emissions Database

ECCAD = Emissions of Chemical Compounds and Compilation of Ancillary Data

Goal of ECCAD

Provide access to surface emissions and associated ancillary data to support many projects:

- International projects such as:
  - **GEIA** (Global Emission Inventories Activity)
  - **IGAC** (International Global Atmospheric Chemistry Project)
  - **CCMI, AEROCOM, , CMIP5/CMIP6, etc.**

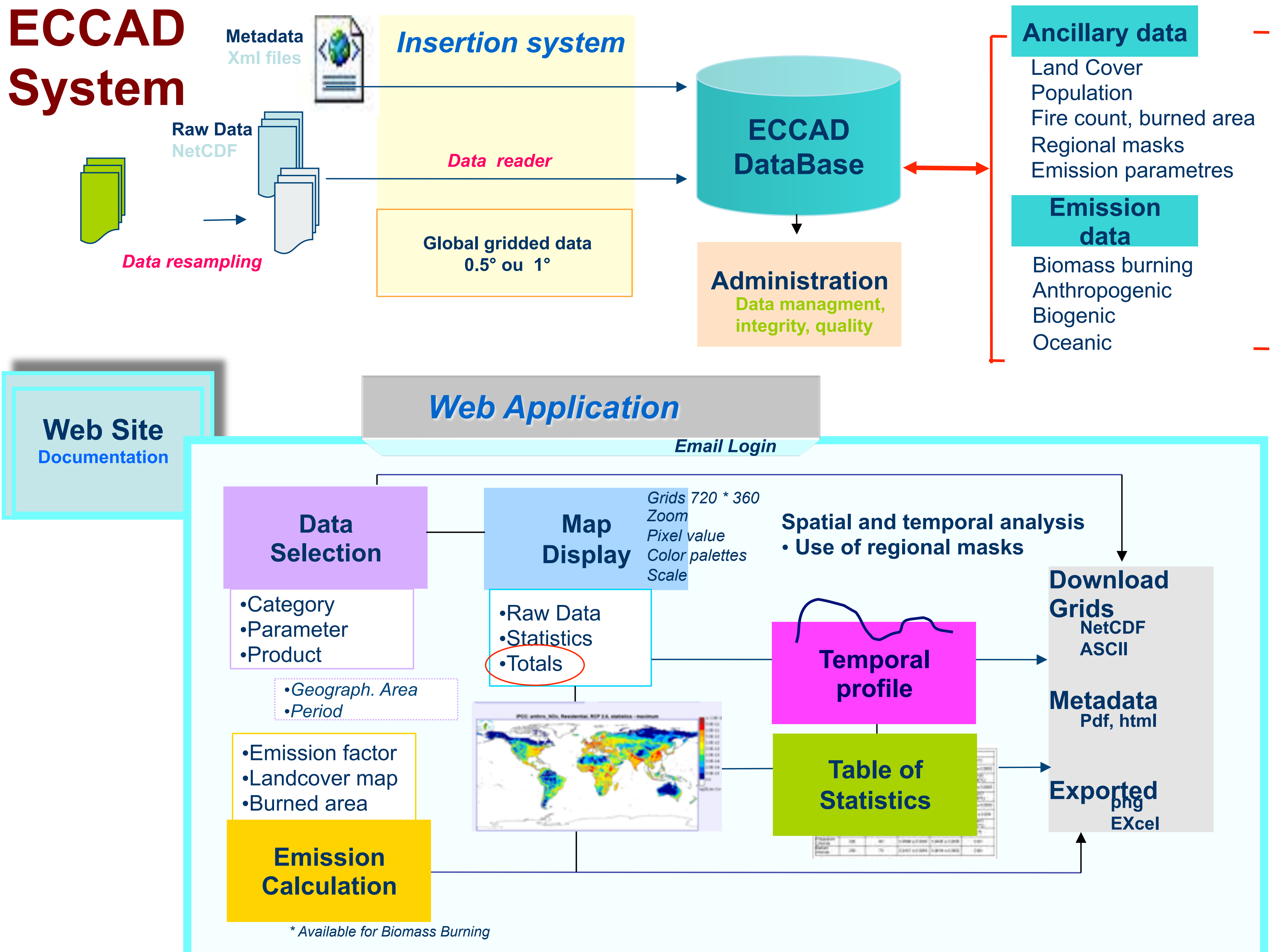
- European research projects such as:
  - **MACC-II** (Monitoring of the Atmospheric Composition and Climate)
  - **PANDA** (Forecasting Air Pollution in China)
  - **ChArMEx** (The Chemistry-Aerosol Mediterranean Experiment)
  - **ECLIPSE, PEGASOS, PANDA, ACCESS, etc.**

Website: pole-ether.fr/eccad

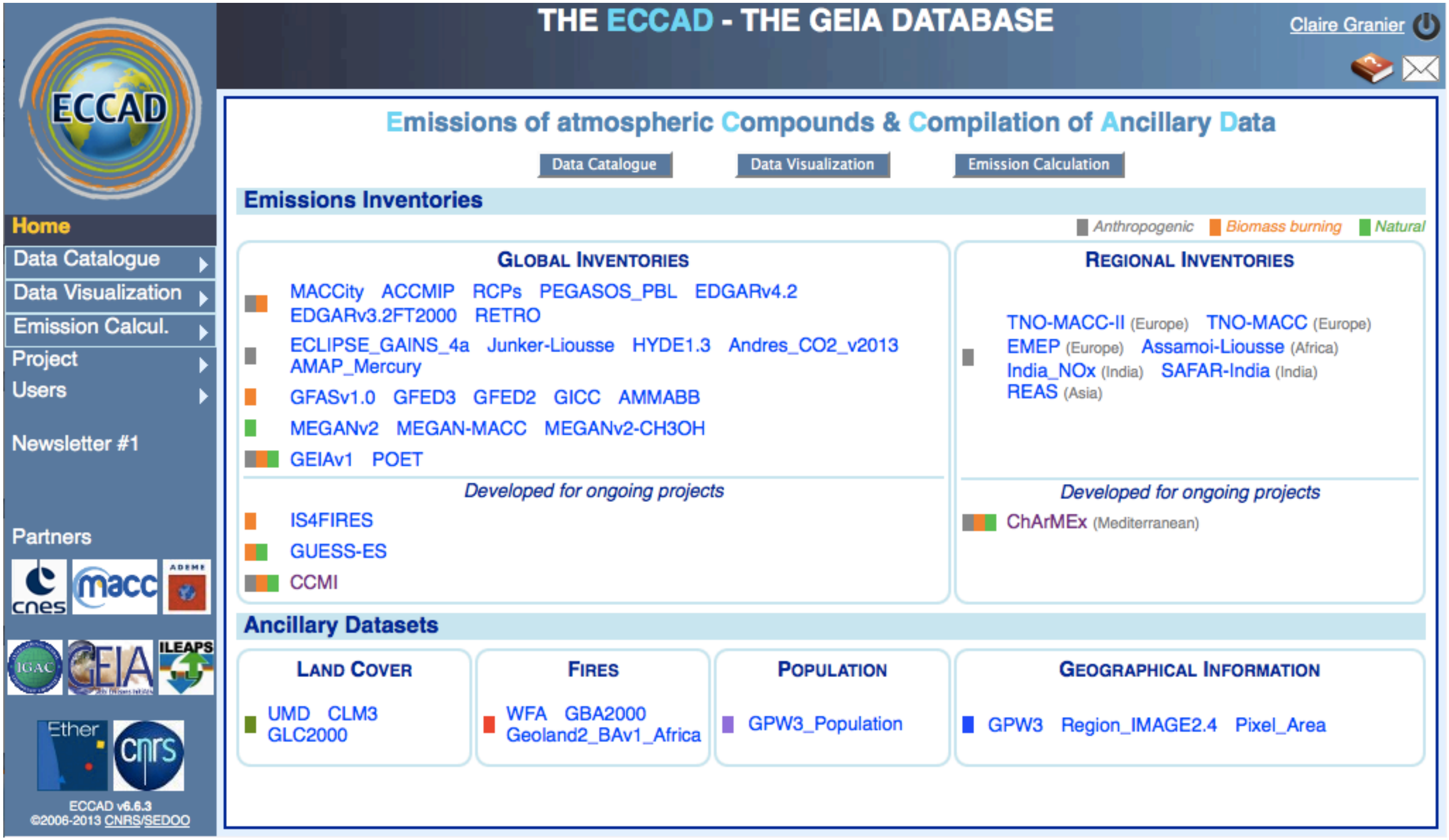
From the ECCAD website

The ECCAD database includes currently a large diversity of datasets, which provide global and regional surface emissions for a large set of chemical compounds. All the data are at a 0.5x0.5 or 1x1 degree resolution. ECCAD provides detailed metadata for each dataset, including information on complete references and methodology, and links to the original inventories.
Several tools are available for the visualization of the data, for computing global and regional totals and for an interactive spatial and temporal analysis. The data can be downloaded as NetCDF CF-compliant files.

NetCDF-CF = Climate and Forecast (CF) Metadata Convention
See http://cfconventions.org/


ECCAD System
Metadata
Xml files
Insertion system
Raw Data
NetCDF
Data reader
Data resampling
Global gridded data
0.5° ou 1°
ECCAD DataBase
Administration
Data managment, integrity, quality
Ancillary data
Land Cover
Population
Fire count, burned area
Regional masks
Emission parametres
Emission data
Biomass burning
Anthropogenic
Biogenic
Oceanic
Web Site
Documentation
Web Application
Email Login
Data Selection
•Category
•Parameter
•Product
•Geograph. Area
•Period
Map Display
Grids 720 * 360
Zoom
Pixel value
Color palettes
Scale
•Raw Data
•Statistics
•Totals
Spatial and temporal analysis
• Use of regional masks
Download Grids
NetCDF
ASCII
Metadata
Pdf, html
Exported
png
EXcel
Temporal profile
Table of Statistics
•Emission factor
•Landcover map
•Burned area
Emission Calculation
* Available for Biomass Burning

ECCAD = Emissions of Atmospheric Compounds and Compilation of Ancillary Data



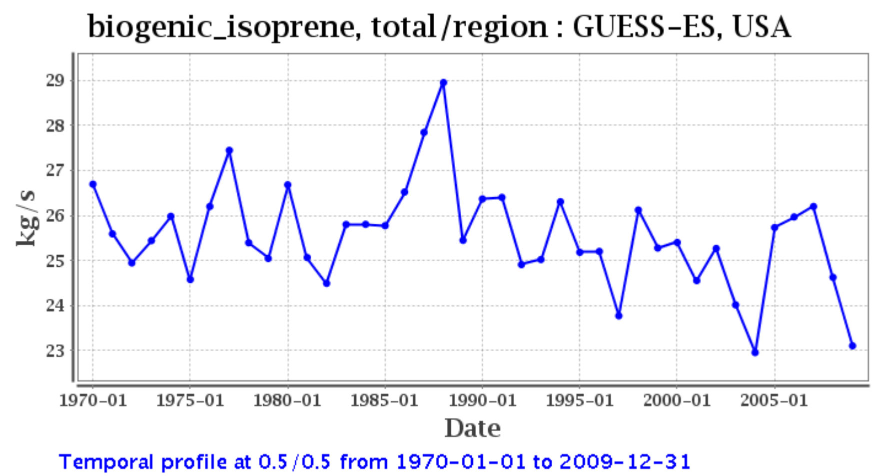
ECCAD is the GEIA emissions database

Regional Inventories (7)						
TNO-MACC-II **Europe** *2013*	2003 - 2009	yearly	Anthropogenic	0.5°	TNO innovation for life	PDF
TNO-MACC **Europe** *2009*	2003 - 2007	Yearly	Anthropogenic	0.5°	TNO innovation for life	PDF
EMEP **Europe** *2007*	1980 - 2020	Yearly	Anthropogenic	0.5°	Convention on Long-range Transboundary Air Pollution emep	PDF
Assamoi-Liousse **Africa** *2012*	2005 - 2030	Decadal	Total	0.5°	LA	PDF
India_NOx **India** *2012*	2005	Yearly	Anthropogenic	0.5°		PDF
SAFAR-India **India** *2012*	1991 - 2011	Decadal	Anthropogenic	1°		PDF
REAS **Asia** *2007*	1980 - 2020	Yearly	Anthropogenic	0.5°	REAS	PDF
Regional Inventories developed for ongoing projects (1)						
ChArMEx **Mediterranean** *2012*	2000	Varied	Anthropogenic Biomass burning Biogenic Oceanic Volcanic	0.25/0.5/1°	CHARMEX MISTRALS	PDF

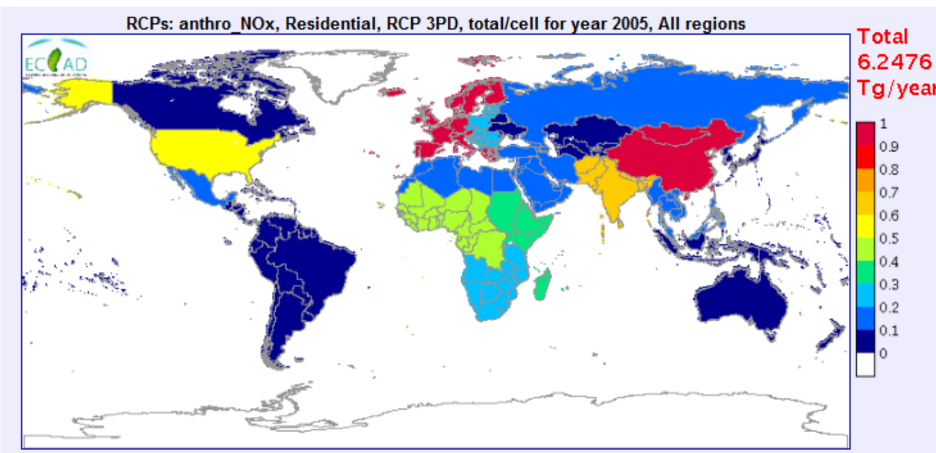
Many different inventories, including global and regional datasets
Only gridded inventories can be inserted currently in the database
New version under development (Thanks to EPA)
→ data at any resolution can be included

ECCAD tools

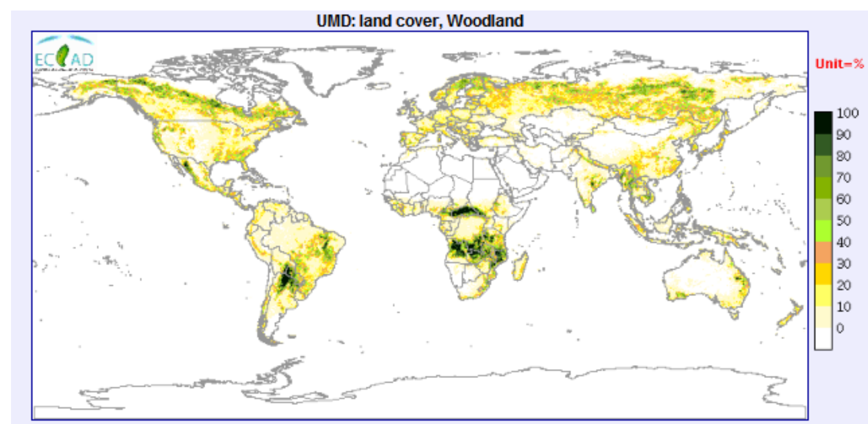
Temporal variation for a country over a period of time



Total emitted for different regions



Ancillary data



Currently under development: comparisons of maps with algrebraic calculations, scattered plots http://www.temis.nl/airpollution/no2.html

https://disc.gsfc.nasa.gov/Aura/data-holdings/OMI/omno2_v003.shtml

http://projects.knmi.nl/omi/research/product/product_generator.php?&info=intro&product=N

http://www.tropmet.res.in/emission/

http://eccad.sedoo.fr/eccad_extract_interface/JSF/page_map_instituts.jsf

Estimate Surface $PM_{2.5}$ From Satellite AOD

AOD is related to $PM_{2.5}$

...but how to separate surface aerosol from column concentrations?

$PM_{2.5}$ / AOD ratio is a function of:

- vertical structure
- aerosol type/hygroscopicity
- meteorology

Multiple approaches:

- surface monitors calibration
- empirical relations
- model output

Approach

satellite-based measurements of *aerosol optical depth* to *$PM_{2.5}$* using a global/regional chemical transport model

---

Following *Liu et al.*, 2004:

$$\text{Estimated } PM_{2.5} = \eta \cdot \tau$$

Combined MODIS/MISR Aerosol Optical Depth

Global/Regional CTM

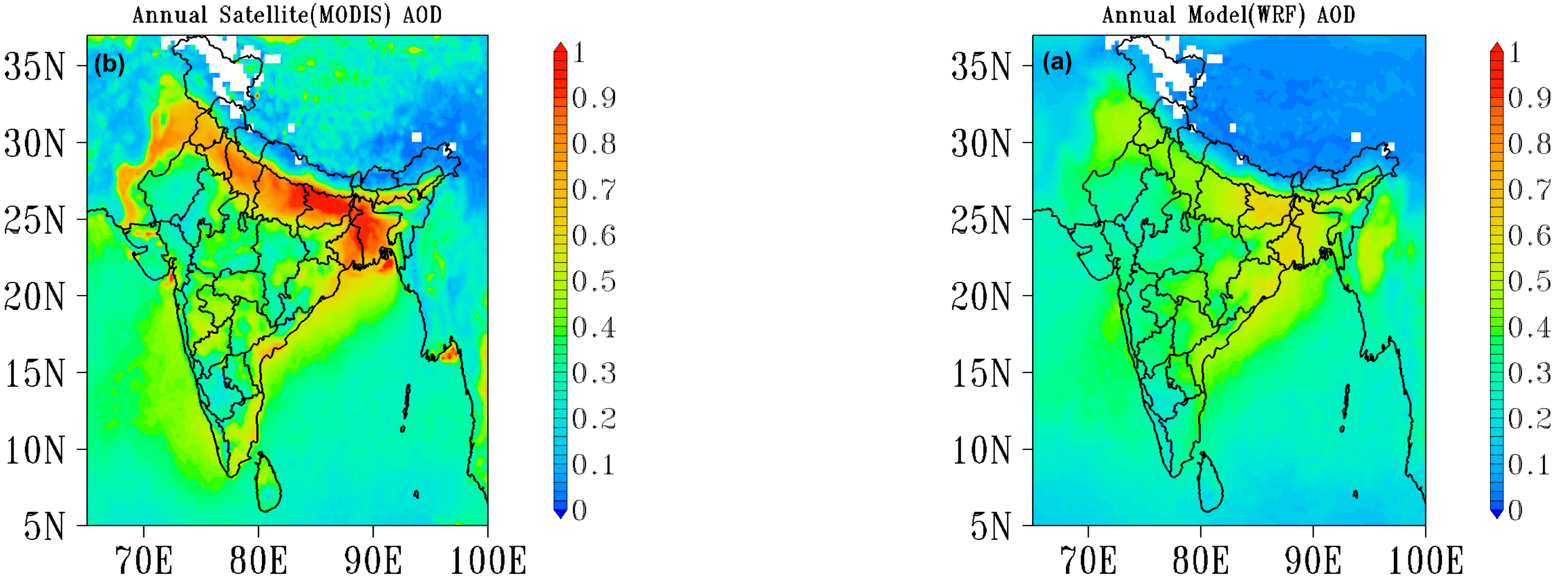
- AOD/$PM_{2.5}$ relationship (η)
- *A priori* values

Satellites provide:

- High resolution (0.1° × 0.1°)
- Observational constraint
- Column-integrated value

η

- vertical structure
- aerosol type
- meteorological effects
- meteorology
- diurnal effects



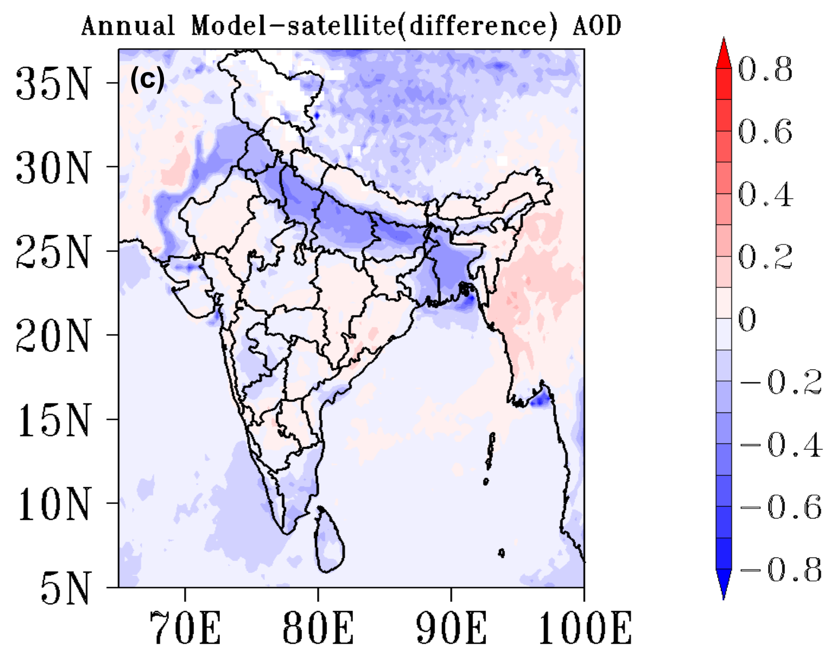


Fig. 2: Annual mean Aerosol Optical Depth (AOD) (a) MODIS Satellite, (b) WRF-chem Model, & (c) annual model-satellite (difference).

Estimated $PM_{2.5} = \eta \cdot \tau$

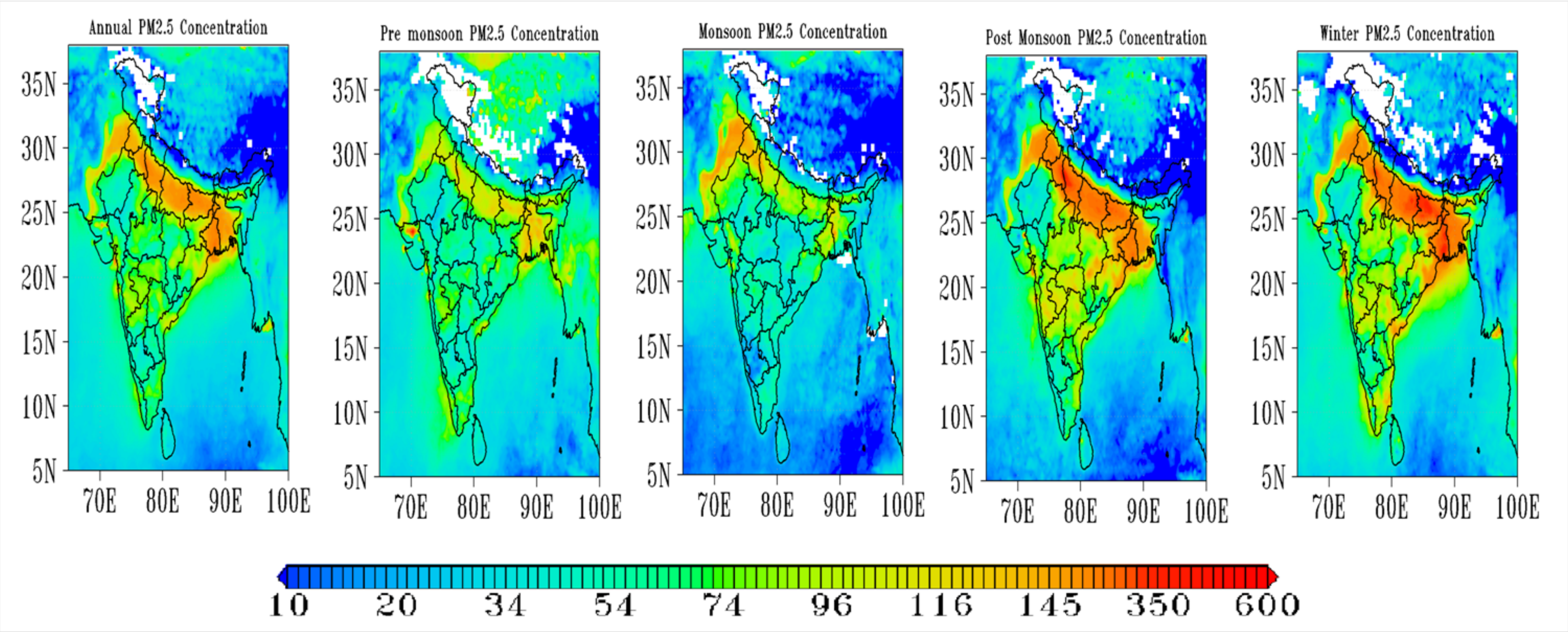


Fig. 7: Estimated $PM_{2.5}$ values for annual and seasonal during the study period.

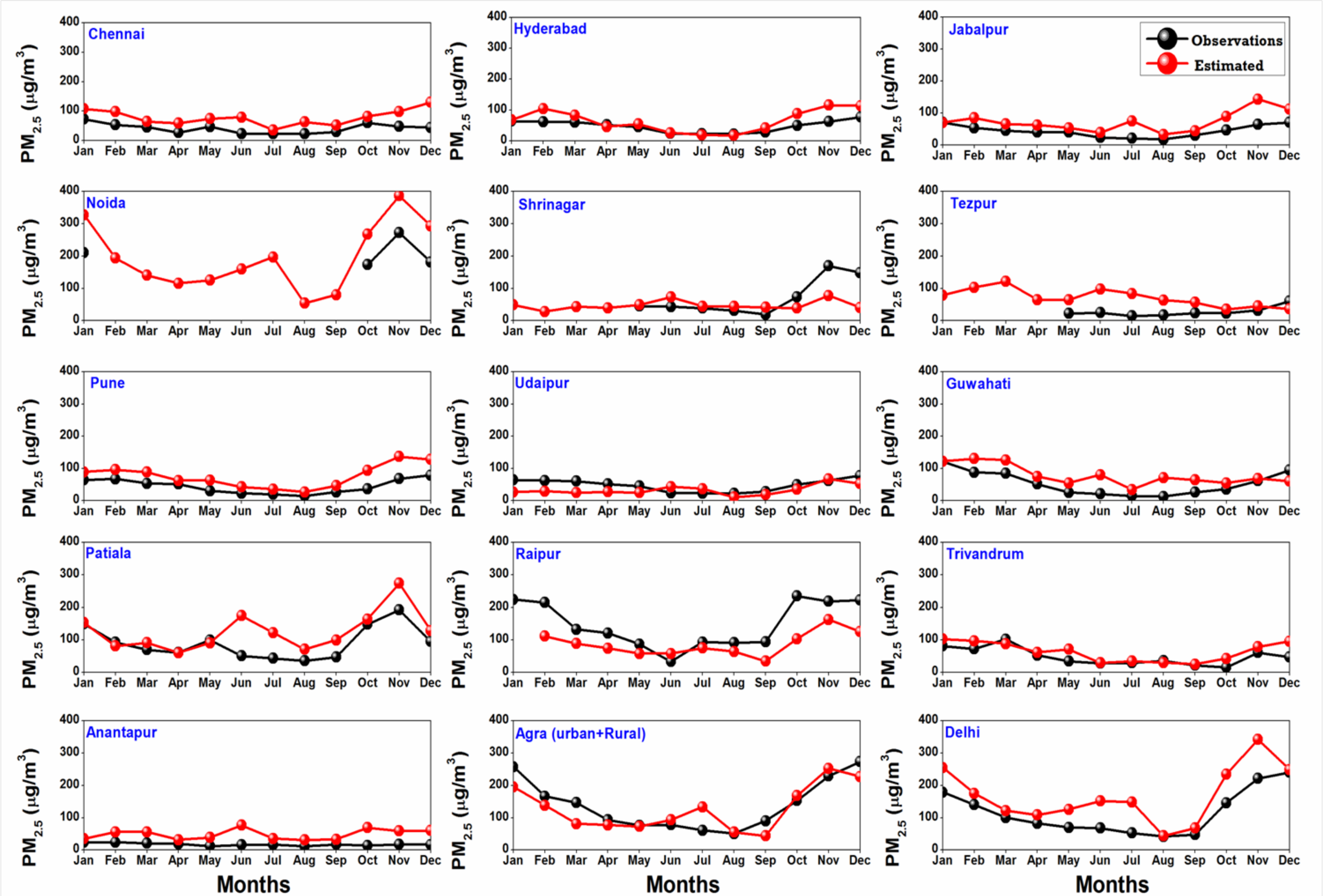


Fig. 3: Variability of monthly mean satellite derived (red), and observed (black) surface $PM_{2.5}$ (in μg/m^3) over 15 monitoring locations.

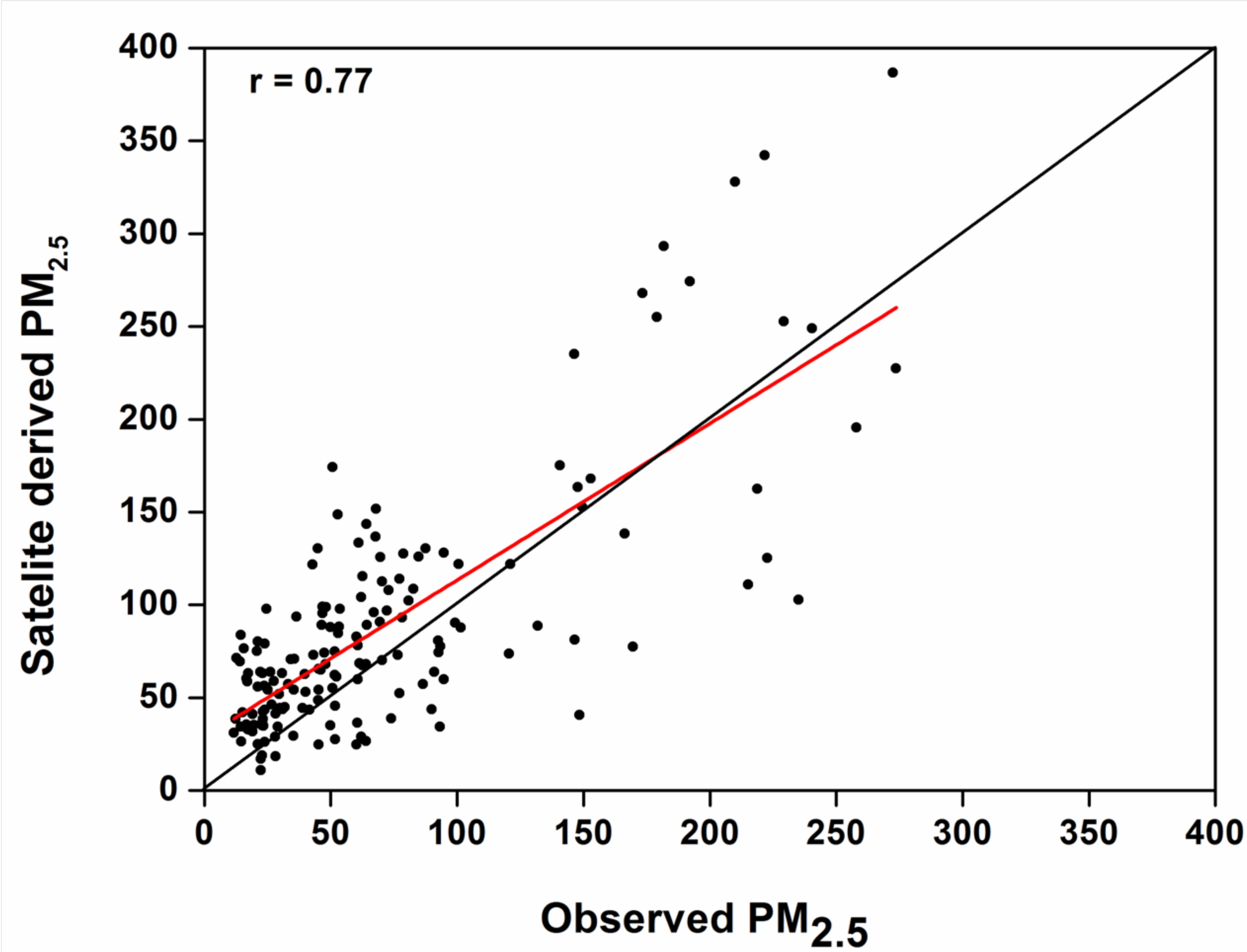


Fig. 4b: Correlation coefficients between observed (monthly mean of 15 stations) and estimated $PM_{2.5}$ concentrations

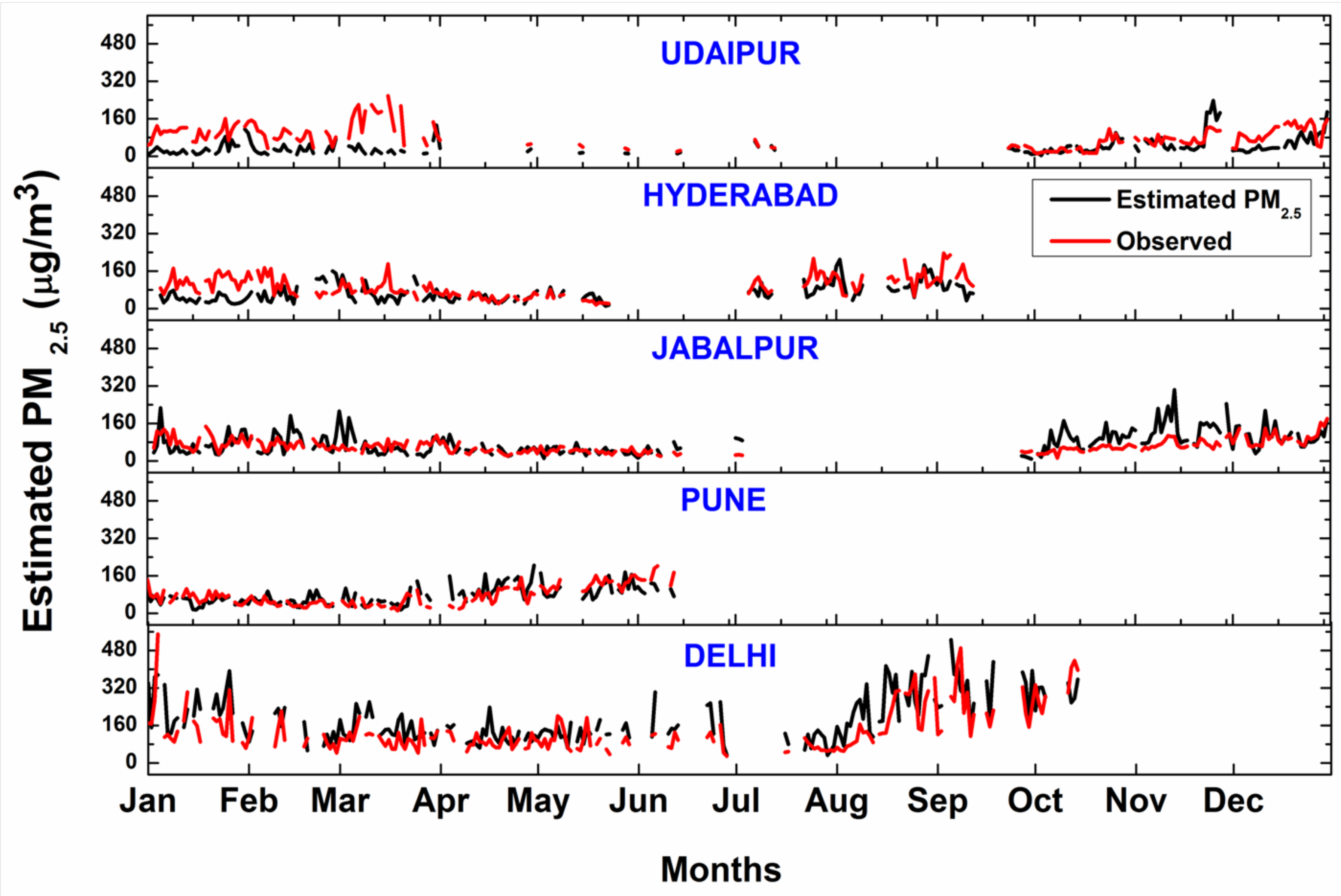


Fig. 5: Variability in observed and estimated $PM_{2.5}$ concentrations over Delhi, Pune, Jabalpur, Hyderabad and Udaipur

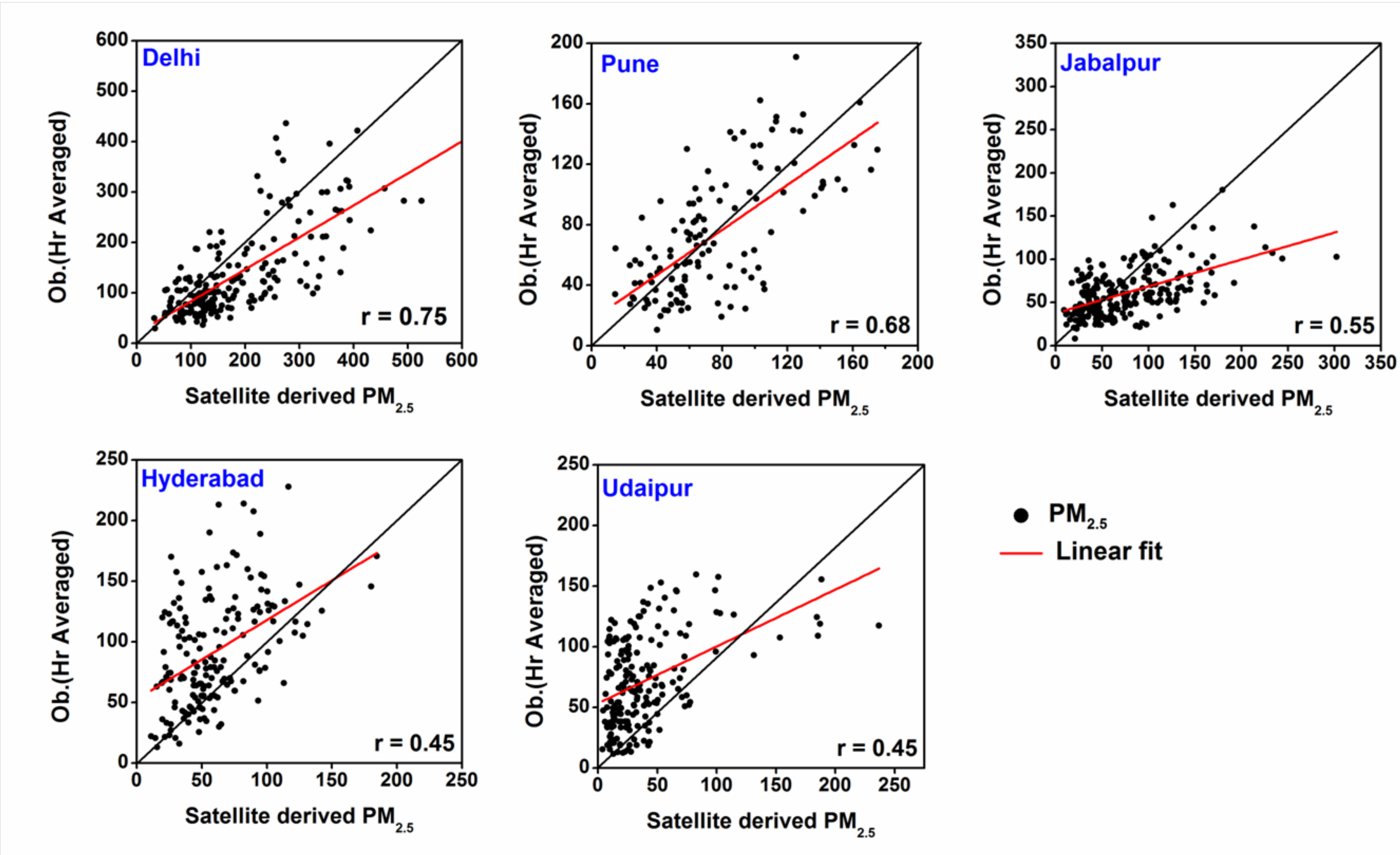


Fig. 6: Correlation coefficients values between observed and Satellite derived $PM_{2.5}$ concentrations over Delhi, Pune, Jabalpur, Hyderabad and Udaipur

Emission Exercise ---- Transportation Sector (Delhi)

Categories of vehicles

1. Two stroke two-wheeler (2W2S)
2. Four stroke two-wheeler (2W4S)
3. Two stroke three-wheeler (3W2S)
4. Four stroke three-wheeler (3W4S)
5. Four wheeler gasoline (4WG)
6. Four wheeler diesel (4WD)
7. Heavy Duty Diesel Low sulfur (HDDLS)
8. Heavy Duty Diesel High sulfur (HDDHS)

Pollutants for which Emission Factors have been determined

1. Carbon dioxide (CO_2)
2. Carbon monoxide (CO)
3. Oxides of Nitrogen (NO)

Computed Mass Emission Factors for Different Vehicles

Species		2W2S	2W4S	3W2S	3W4S	4WG	4WD	HDDLS	HDDHS
FC	g/km	11.0	9.7	22.1	25.9	84.3	92.7	195.2	195.2
	G/hr	254.9	225.2	511.2	599.8	1576.5	1733.2	3649.9	3649.9
CO_2	g/km	26.6	28.3	60.3	78.5	223.6	208.3	515.1	515.2
	G/hr	617.0	655.6	1397.2	1817.1	4181.3	3896.4	9633.3	9634.8
CO	g/km	2.0	1.4	5.25	2.0	24.8	2.0	4.7	4.7
	G/hr	46.4	33	121.6	46.9	462.9	36.8	88.4	87.4
NO	g/km	0.8	1.4	1.2	2.0	3.3	116.9	354.3	405.3
	G/hr	20.5	32.9	28.1	46.0	62.4	2185.5	6626.6	7579.2

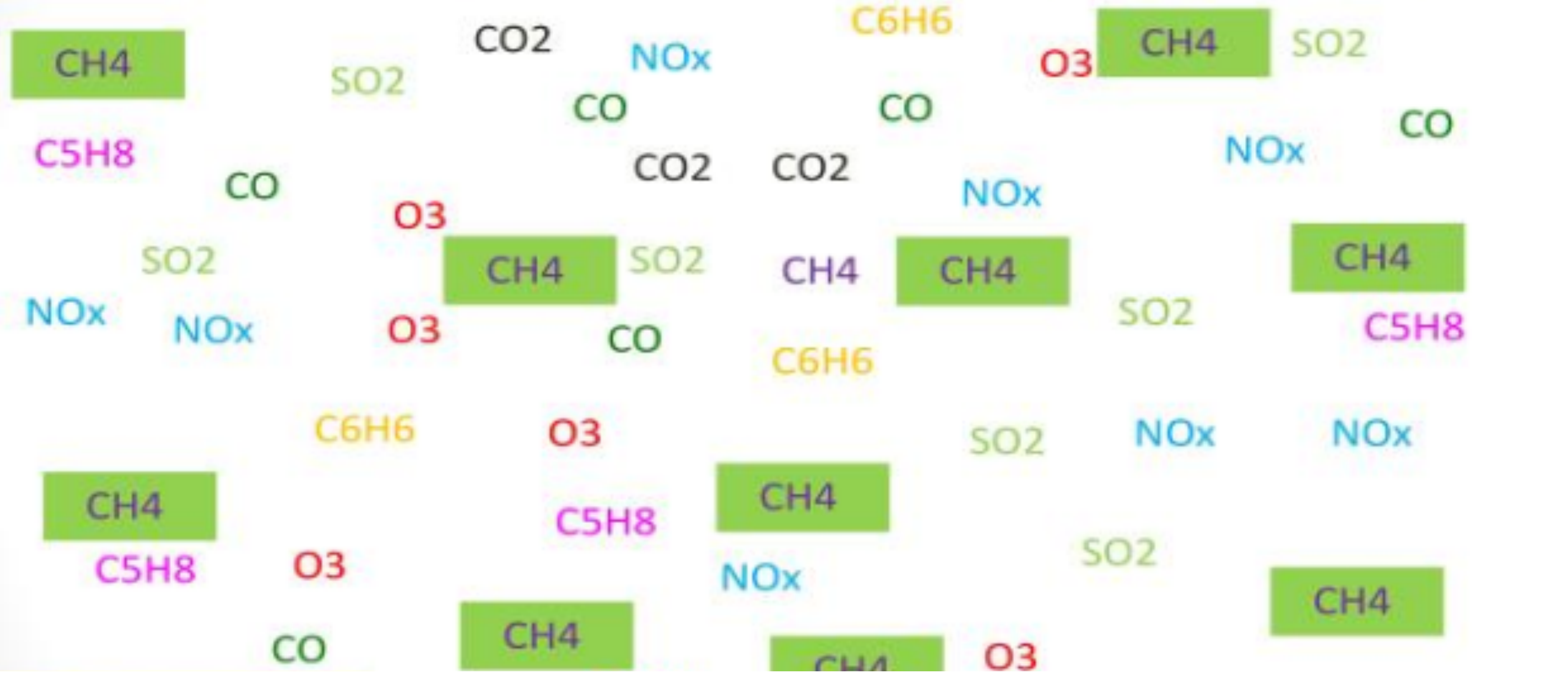
Area of Delhi: 36 km x 36 km

Number of total vehicles : 4.5 million

1.	Two stroke two-wheeler (2W2S): 10%	25km /day
2.	Four stroke two-wheeler (2W4S) : 10%	25km /day
3.	Two stroke three-wheeler (3W2S): 15%	50km /day
4.	Four stroke three-wheeler (3W4S):5%	50km /day
5.	Four wheeler gasoline (4WG): 30%	25km /day
6.	Four wheeler diesel (4WD): 10%	25km /day
7.	Heavy Duty Diesel Low sulfur (HDDLS): 10%	10km /day
8.	Heavy Duty Diesel High sulfur (HDDHS): 10%	10km /day

Calculate total annual emissions from the transportation secto

Challenge: How to measure one compound among many?



Category	Parameters	Instruments
Ground and surface properties	Temperature and water profile in the ground Ground heat flux	Soil moisture sensors
Surface layer meteorology	Temperature, humidity, pressure, wind speed, wind direction at1m, 2m, 5m, 10m, turbulence and precipitation	All in one whether station, 10m vertical mast, IMD-SAFAR AWS network, sonic anemometers
Radiation fluxes	Downwelling shortwave (SW) direct, diffuse, and global long wave (LW) irradiance	Sky Radiometer, Net radiometer, pyranometers
Atmospheric profiles	Temperature, humidity, pressure, wind speed, wind direction	Microwave radiometer, Tethersonde instruments, Radio-sonde (IMD)
Aerosols and fog optical properties	Visibility, fog and cloud base height, absorption coefficient, scattering coefficient, aerosol optical depth	RVR, fog detector, Photoacoustic Extinctiometer (PAX- aerosol absorption and scattering), Neflometer, sun-photometers, Aethalometer
Aerosols and fog microphysics	Aerosol particle size distribution, aerosol particle counter, Condensation Particle Counter/ Condensation droplet counter, CCN	SMPS, CPC/CDP, CCN counter, GRIMM
Aerosol/gas chemistry	Fog collector, Particle in Liquid Sampler (PILS), EC/OC analyser, VOCs, Aérosol filter collector, SO_2 gas analyzer , PM_1, $PM_{2.5}$, PM_{10}	Fog Collector, PILS, PTRMS EC/OC anayzer, SO_2 analyzer, PM analyzers

In brief, tropospheric NO2 column is derived in three main steps involving the calculation of
(1) slant column (using Differential Optical Absorption Spectroscopy (DOAS) approach in the 405–465 nm spectral window),
(2) tropospheric slant column (using modeling/ assimilation approach),
(3) tropospheric vertical column (using air mass factor—AMF).