



› **EMISSION INVENTORIES FOR AIR QUALITY  
AND CLIMATE MODELLING IN EUROPE**  
HIGHLIGHTING SOME RECENT ISSUES, IMPROVEMENTS AND TRENDS

Hugo Denier van der Gon

**TNO** innovation

**with a lot of help from my colleagues**

*Prettige Kerstdagen*



*Gelukkig Nieuwjaar*

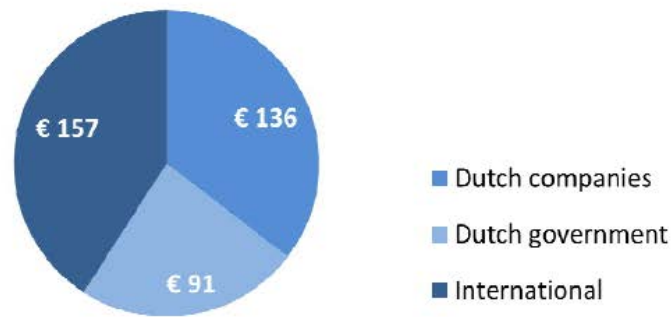
# WELCOME TO TNO

- › Number of employees (effective average) 2.926
- › Consolidated turnover € 518 million

**Consolidated turnover (million)**



**Consolidated market turnover (million)**



Trends Transitions

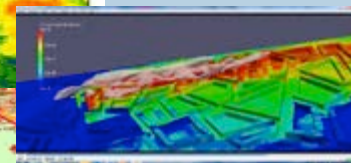
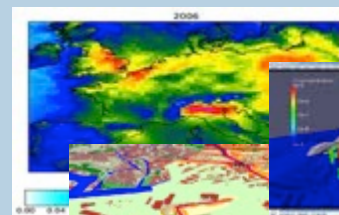
# Environment & Sustainability



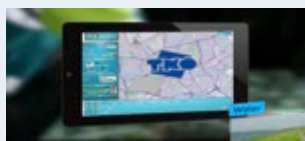
Lab Facilities

## Sensors

- ▣ Air
- ▣ Water
- ▣ Noise
- ▣ Nano
- ▣ Heat



Emissions, Models & Simulation



ICT, (big) data



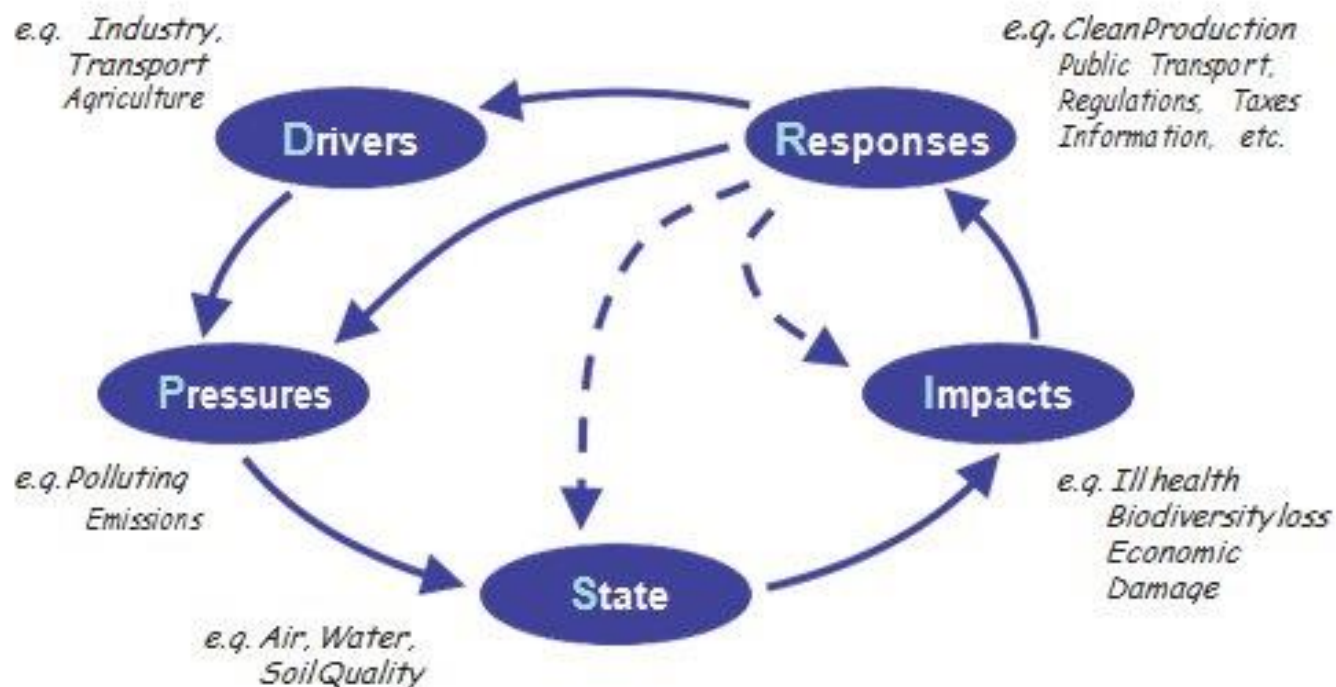
Field experiments

# OUTLINE

- › Introduction
- › Why & How make an inventory ?
- › A selection of topics...
  - › Greenhouse gases
  - › Non-exhaust road transport emissions
  - › Woodburning PM
  - › Emission timing
  - › Validate and Verify
- › Questions & Discussion

# DPSIR FRAMEWORK....

## › Emissions are key!



# USE OF EMISSIONS DATA: INPUT FOR AIR QUALITY FORECASTS

[HTTP://ATMOSPHERE.COPERNICUS.EU/ABOUT-CAMS](http://atmosphere.copernicus.eu/about-cams)

ABOUT CAMS

NEWS & MEDIA

EVENTS

CATALOGUE

RESOURCES

TENDERS

USER SUPPORT

## HOURLY ENSEMBLE MAPS

Home > Services > RegionalAirQuality > Ensemble Analysis and Forecast > Hourly Ensemble Maps

Data access

Ensemble Analysis  
and Forecast

Individual Analyses

Individual Forecasts

Verification of  
Analyses & Forecasts

Ensemble reanalysis

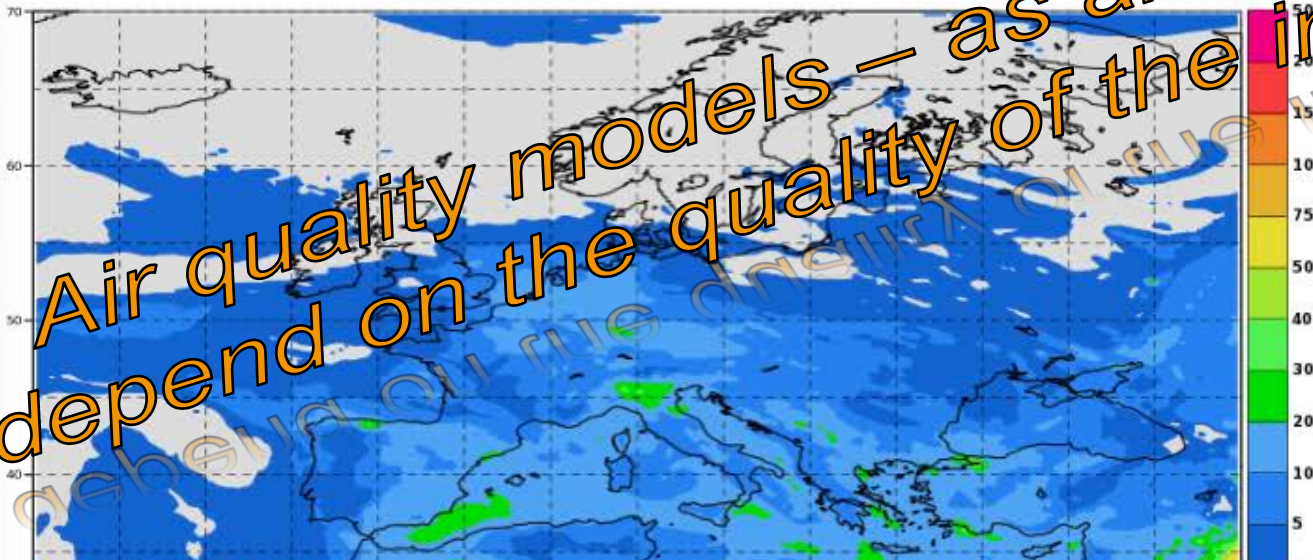
Documentation

Daily mean and maximum

Hourly Ensemble Maps

EPSgrams

Wednesday 12 April 2017 00UTC CAMS Forecast t+063 VT: Friday 14 April 2017 15UTC  
Model: ENSEMBLE Height level: Surface Parameter: PM2.5 Aerosol [  $\mu\text{g}/\text{m}^3$  ]



MACC-III is a Coordination & Support Action (2014-2015) funded by the European Union under the Horizon 2020 Programme.

It is coordinated by the European Centre for Medium-Range Weather Forecasts and operated by a 36-member consortium.

<http://macc-raq-op.meteo.fr/?category=ensemble>

# EMISSION INVENTORIES....

**EMISSION = ACTIVITY X EMISSION FACTOR**

› **Uhhhh OK, but is it.....?**



**or**



# BOTTOM-UP EMISSION INVENTORIES AND HYBRID, COMPILED INVENTORIES

## › Classic Emission inventory

$$Emission_{pollutant} = \sum_{activities} Activity\ rate_{activity} \times Emission\ factor_{activity, pollutant}$$

## › EMEP/EEA air pollutant emission inventory guidebook: Technical guidance to prepare national emission inventories

<http://www.eea.europa.eu/publications/emep-eea-guidebook-2016>

› Examples: Dutch Emission Registration; The Emissions Database for Global Atmospheric Research (EDGAR) <http://edgar.jrc.ec.europa.eu/>

## › Hybrid, compiled Emission inventory (collecting various different bottom-up estimates, possibly including expert judgements)

› Example TNO-MACC (Kuenen et al., ACP, 2014)

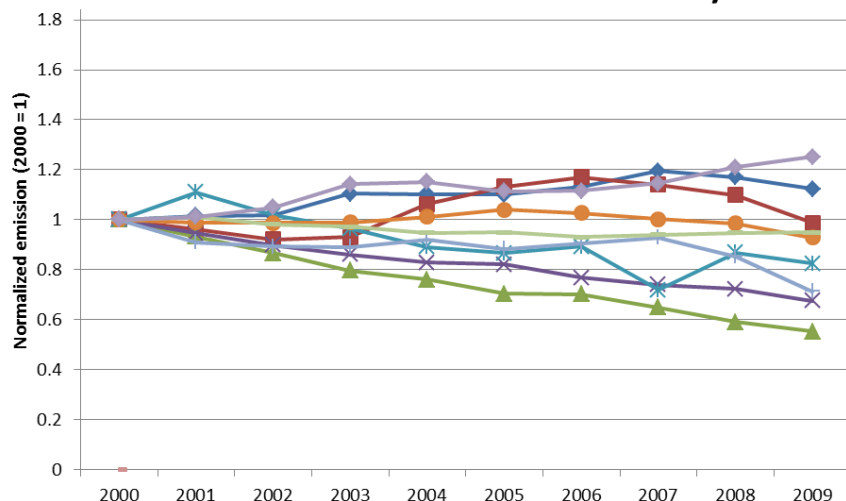
## WHY IS “EXTRA” WORK NEEDED?

- › In Europe Countries shall make their own inventories and shall provide these to EMEP (The European Monitoring and Evaluation Programme EMEP) (& also in gridded form for modelling).
- › Can be accessed at <http://www.ceip.at/>
- › But.....
- › Some countries do not comply (or only partly)!
- › Time series may be incomplete
- › The spatial distribution is not consistent between countries
- › ....

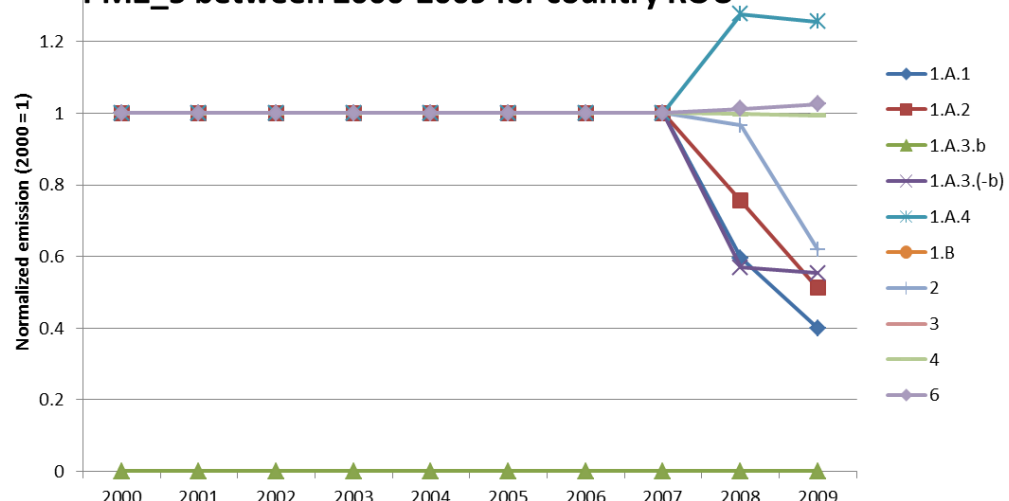
Note: Slide is no longer up to date – example of past analysis

# IS THERE A PROBLEM ?

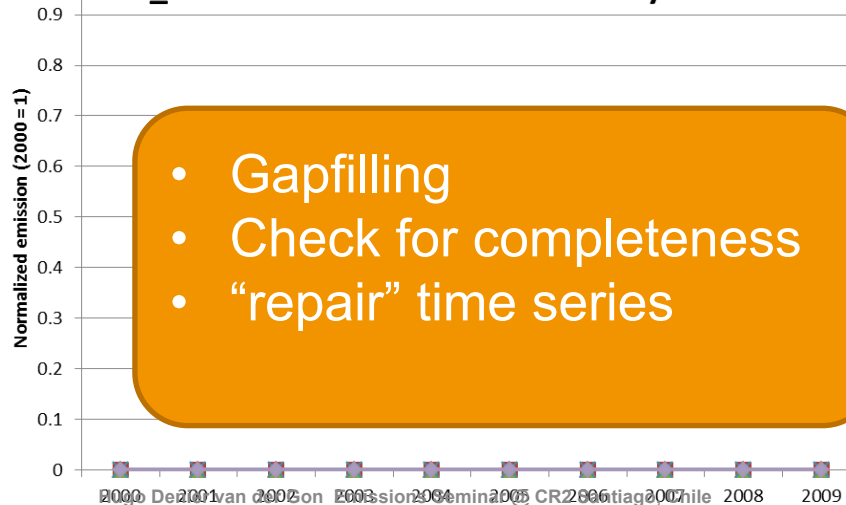
Emissions of NOX between 2000-2009 for country DEU



PM2\_5 between 2000-2009 for country ROU

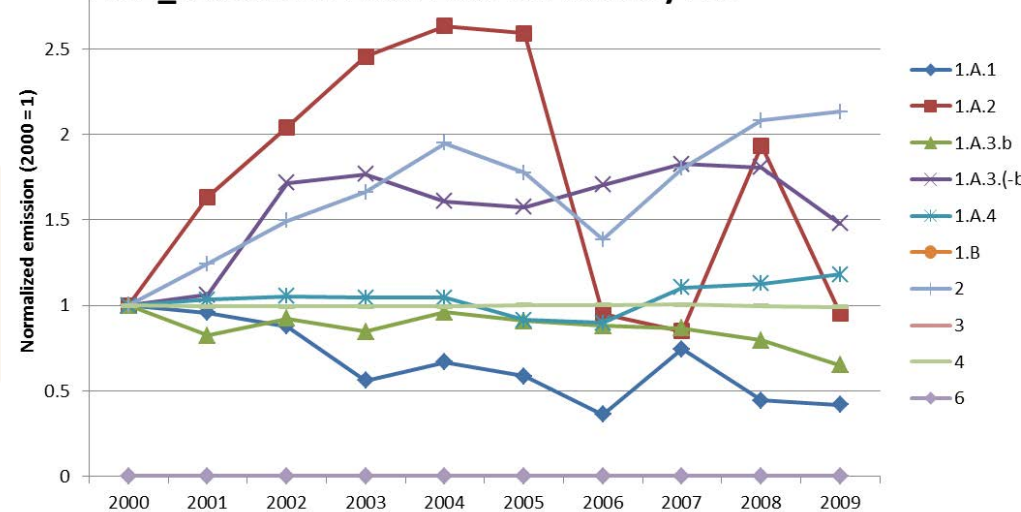


PM2\_5 between 2000-2009 for country GRC



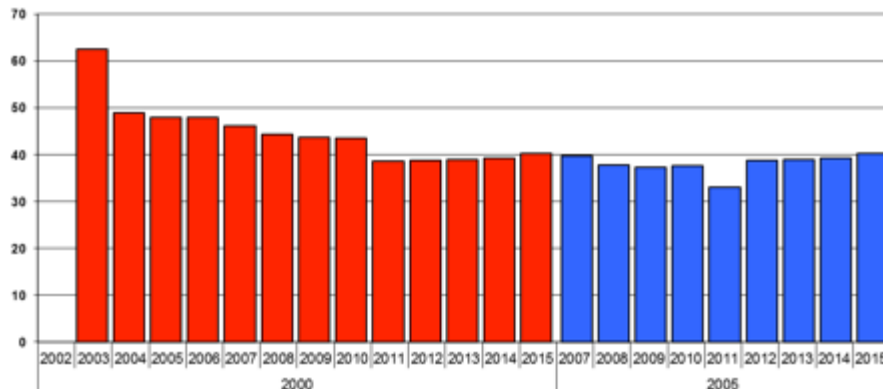
- Gapfilling
- Check for completeness
- “repair” time series

PM2\_5 between 2000-2009 for country EST

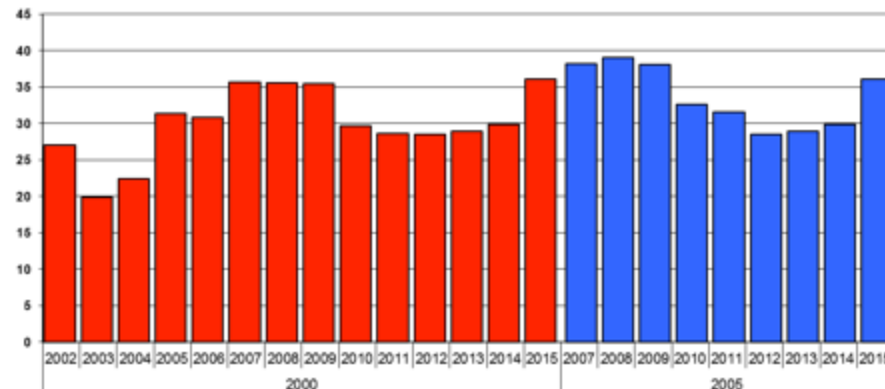


## OFFICIALLY REPORTED EMISSIONS OF PM10 (KT)

Officially reported emissions of PM10 (Gg) in Netherlands



Officially reported emissions of PM10 (Gg) in Denmark



### Important consequences:

- Need to “freeze” the data set at a chosen moment because the emissions for a given year are not static.
- If the time series is extended, all years need to be updated
- Needs good communication and explanation!

*Thanks to EMEP- CEIP for keeping the data and making it available!*

# TNO-MACC-III EMISSION INVENTORY

**We make liberal use of country reported emissions** (EMEP-CEIP) and IIASA GAINS but cater for the AQ models and user specific needs:

- › Pollutants covered:
  - › NO<sub>x</sub>, SO<sub>2</sub>, NMVOC, NH<sub>3</sub>, CH<sub>4</sub>, CO, PM<sub>10</sub>, PM<sub>2.5</sub> (and components)
  - › Add or “do” components that are not or incompletely reported like EC
  - › High resolution (~ 7x 7 km), consistent distribution patterns for road transport, agriculture, power plants, industry, etc.
  - › UNECE-EUROPE... not limited to EU27
  - › Collaborate in interpretation and provide description of input.

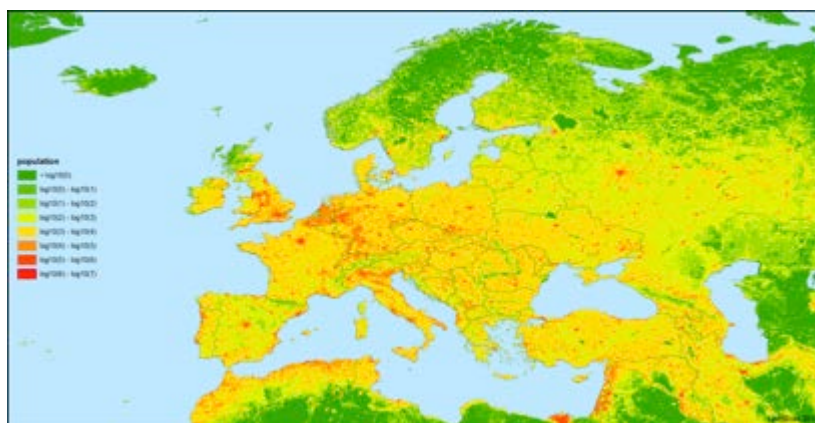
See *J. Kuenen et al., ACP, 2014*

<http://www.atmos-chem-phys.net/14/10963/2014/acp-14-10963-2014.pdf>

# SPATIAL DISTRIBUTION

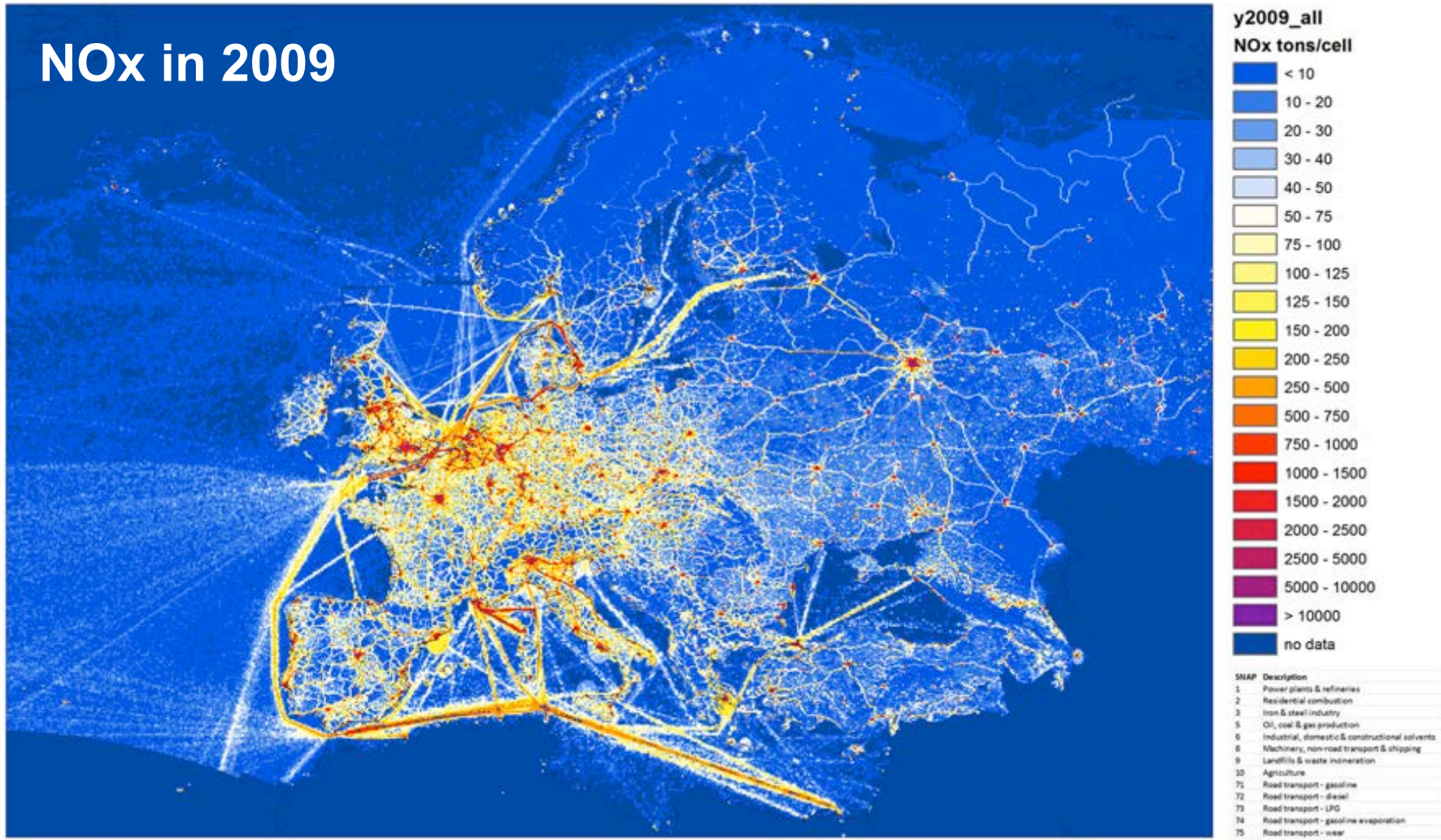
- › Use generic spatial distribution system throughout Europe
- › All emission sources distributed using proxy parameters, e.g.

*Population density (total, rural, urban)*



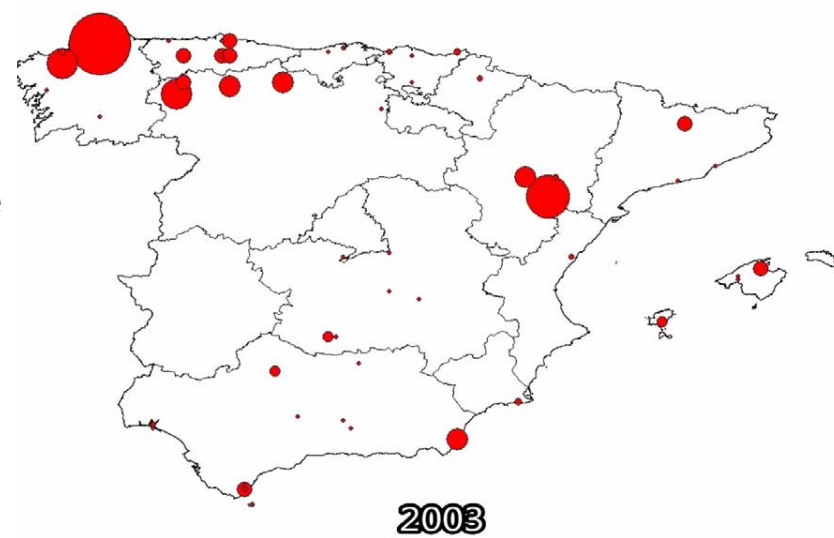
# RESULTING EMISSION MAPS

**NO<sub>x</sub> in 2009**



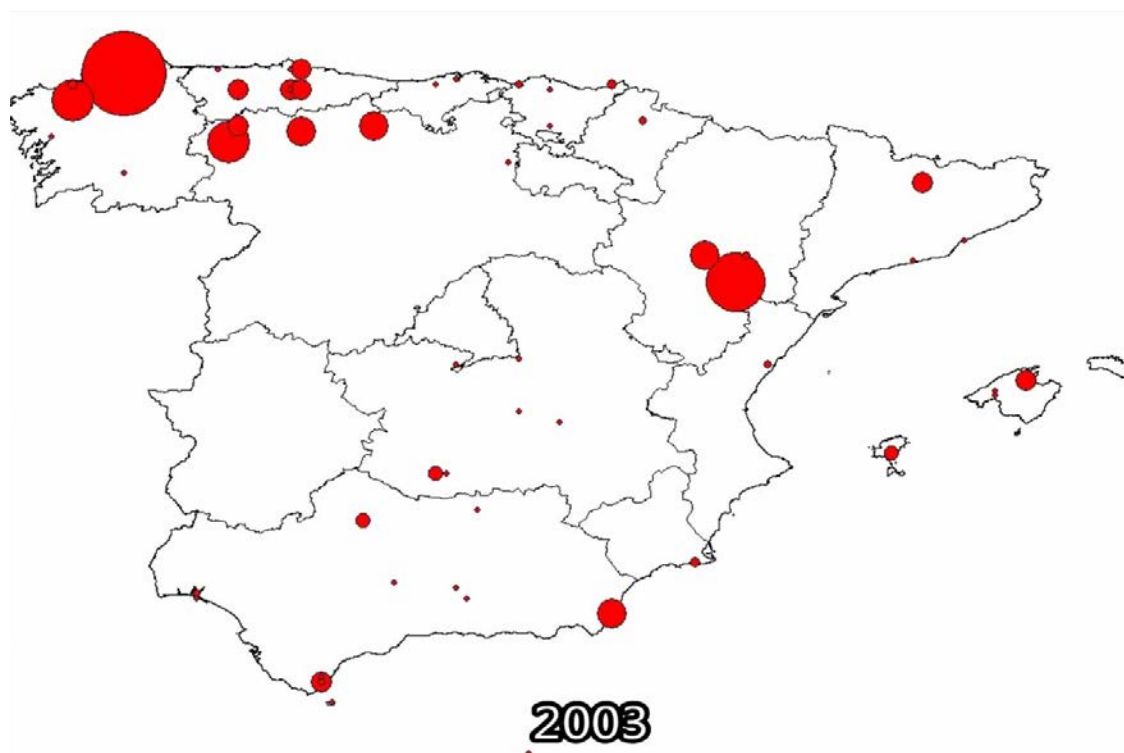
# POINT SOURCES

- › Use of European Pollutant Release and Transfer Register (and predecessor EPER): <http://prtr.ec.europa.eu>
  - › EPER: 2001 and 2004
  - › E-PRTR: annual data from 2007 onwards
- › Used in TNO-MACC-II as relative proxy for distributing emissions for specific sector/fuel combinations (2004 proxy for years 2003-2005, 2007 also for 2006)
- › Where E-PRTR data not available or not suitable, TNO PS info from TNO\_MACC-I has been used as proxy for distributing emissions



# POINT SOURCES

- › Use of European Pollutant Release and Transfer Register (and predecessor EPER): <http://prtr.ec.europa.eu>
- › EPER: 2001 and 2004; E-PRTR: annual data from 2007 onwards

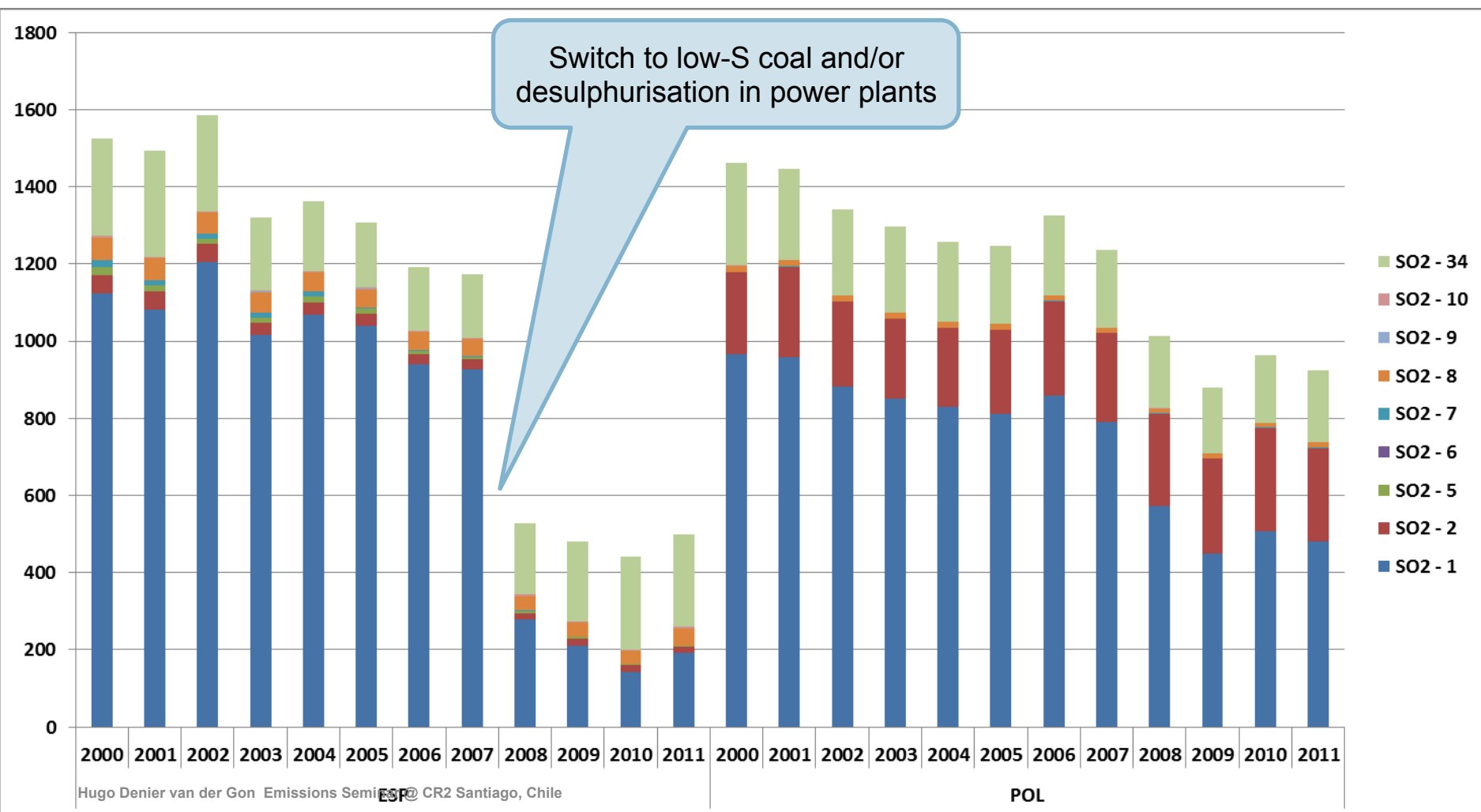


# TNO-MACC\_III PM10 TRENDS 2000-2011

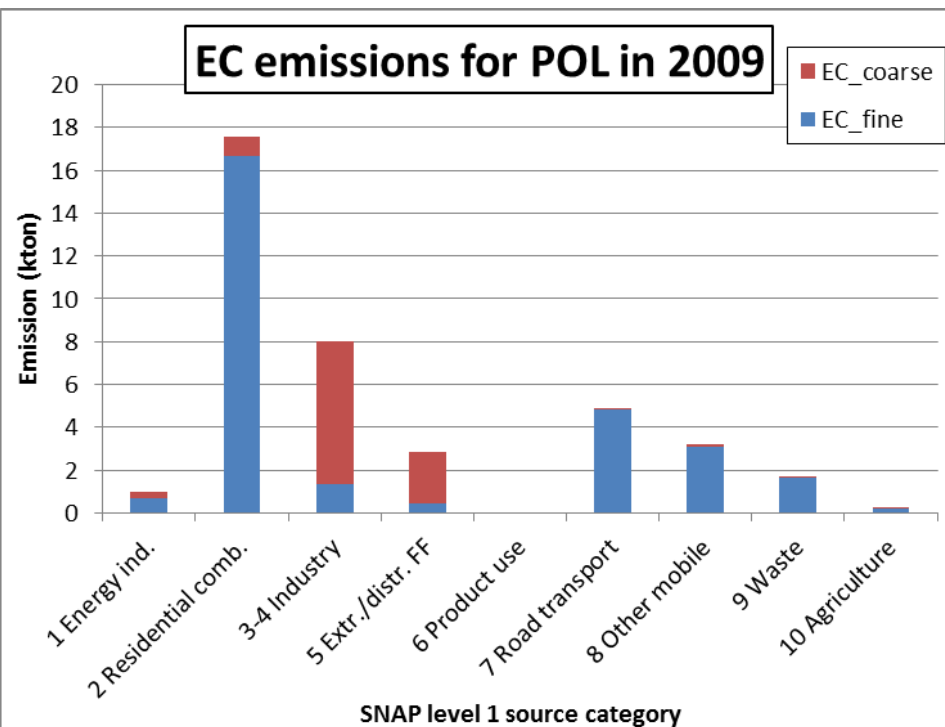
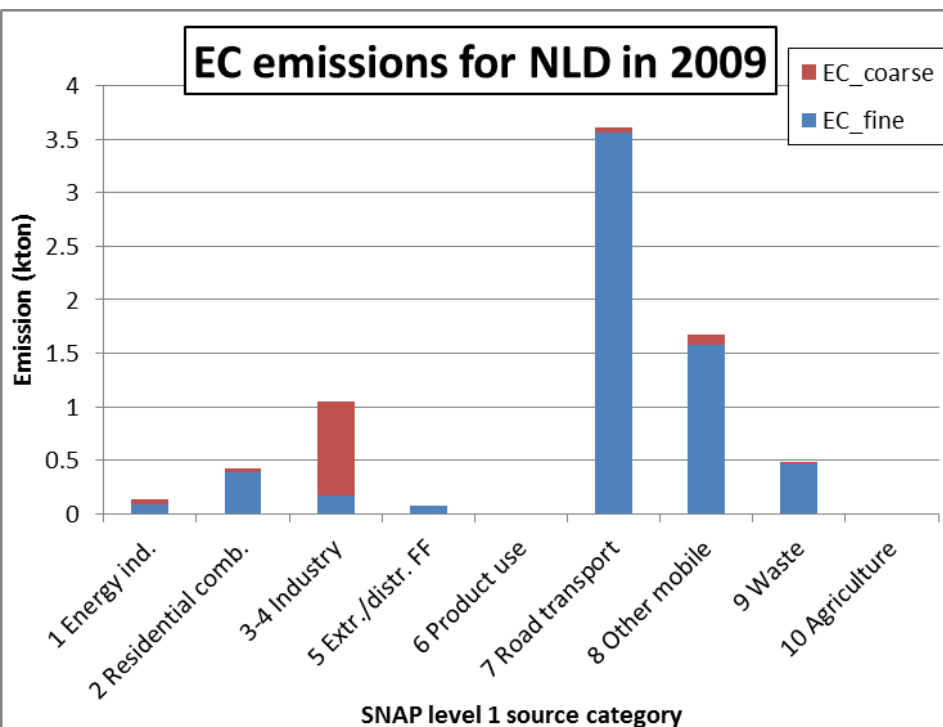
Sum of SumOfEmission



## CONSISTENT EMISSIONS DATA BROKEN DOWN BY SOURCE SECTOR BY COUNTRY. EXAMPLE SO<sub>2</sub> FOR SPAIN AND POLAND.

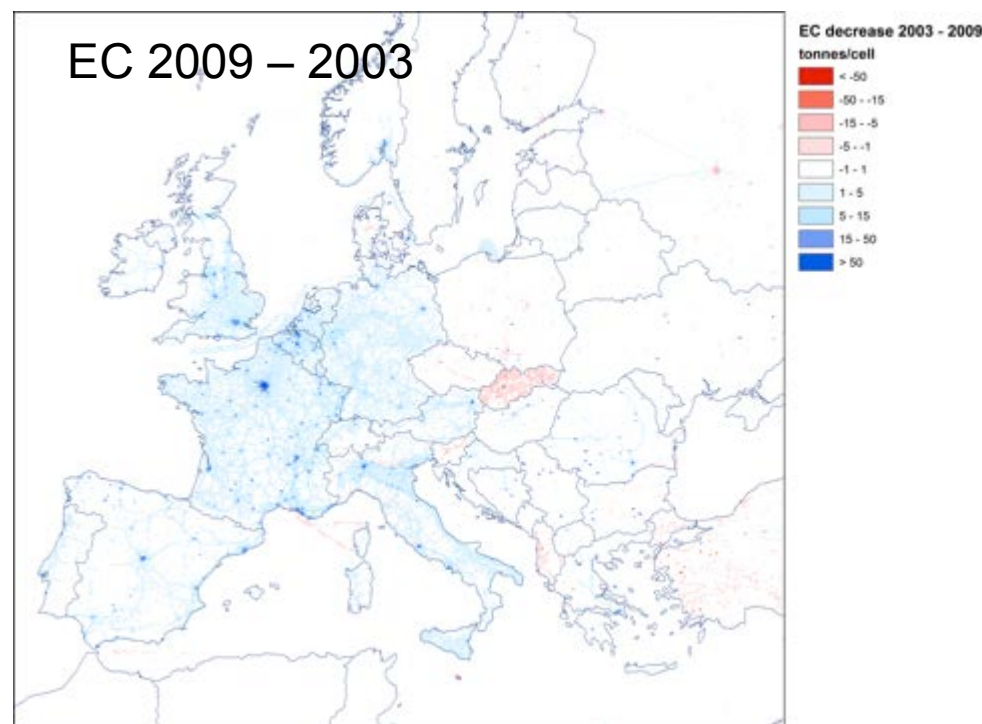
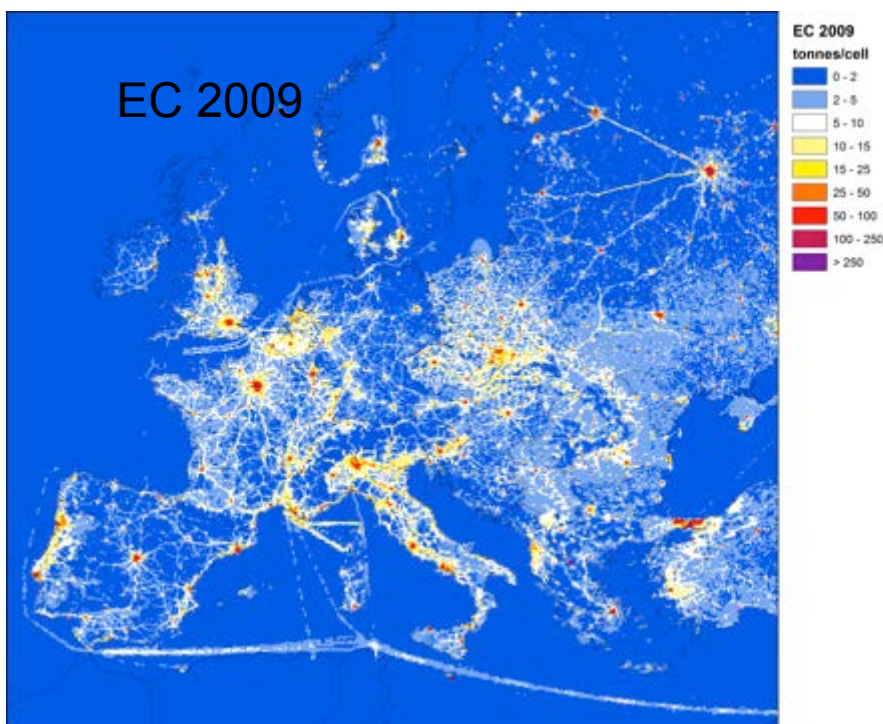


## Example of derived Elemental carbon emissions by country by sector



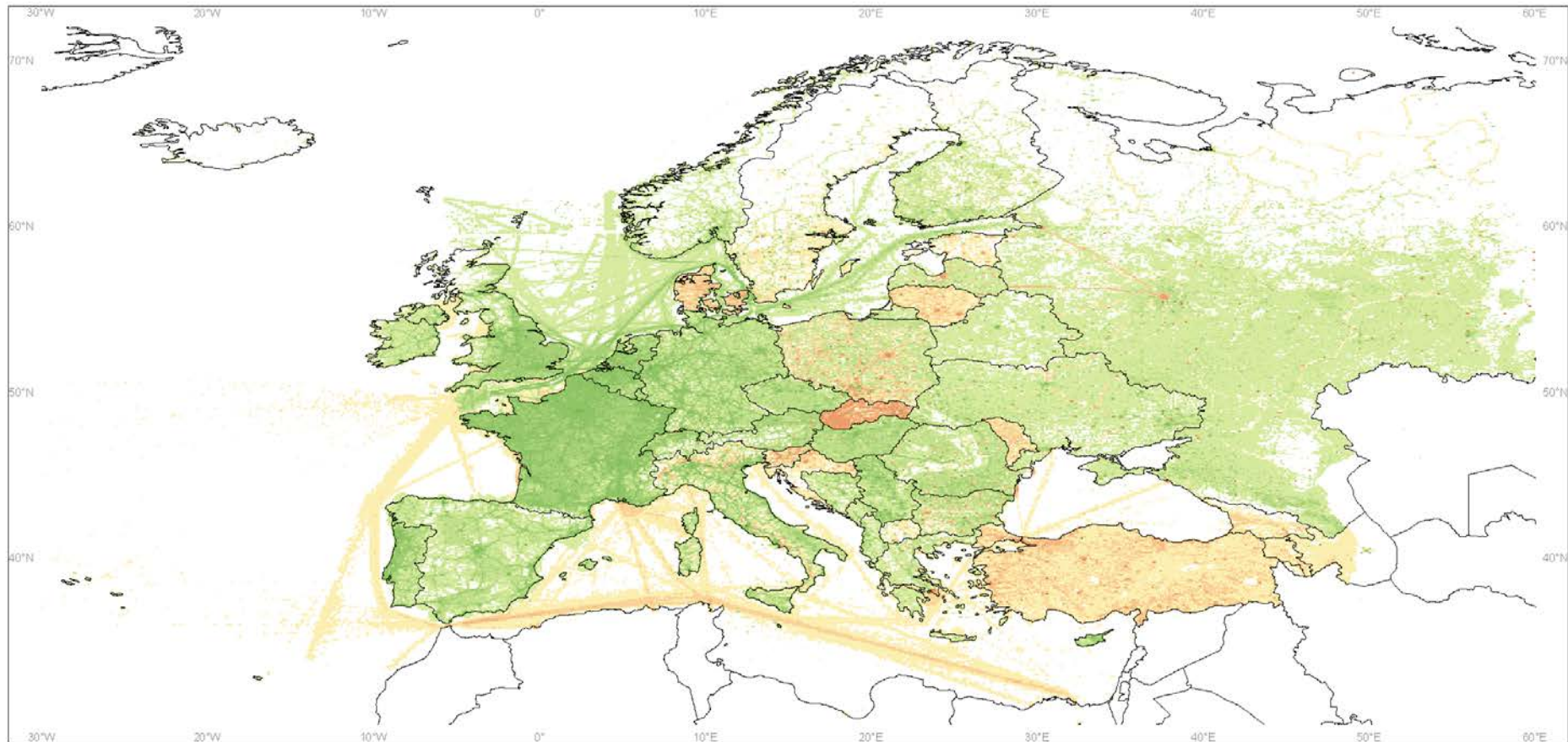
Dominant sources vary by country!  
Effective policy in Netherlands addresses road transport,  
in Poland focus should be on residential combustion

# SPATIAL DISTRIBUTION AND CHANGE OVER TIME (TNO-MACC EMISSION INVENTORY)

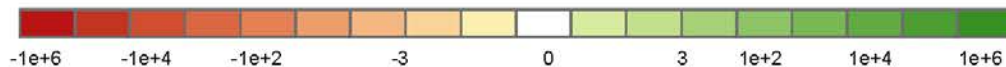


**Source:** Kuenen, J., A. Visschedijk, M. Jozwicka, and H. Denier van der Gon, TNO-MACC\_II emission inventory; a multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling ACP, 14, 10963–10976, 2014

# CHANGE IN PM2.5 EMISSION BETWEEN 2000 AND 2011 (GREEN = REDUCTION; RED = INCREASE)

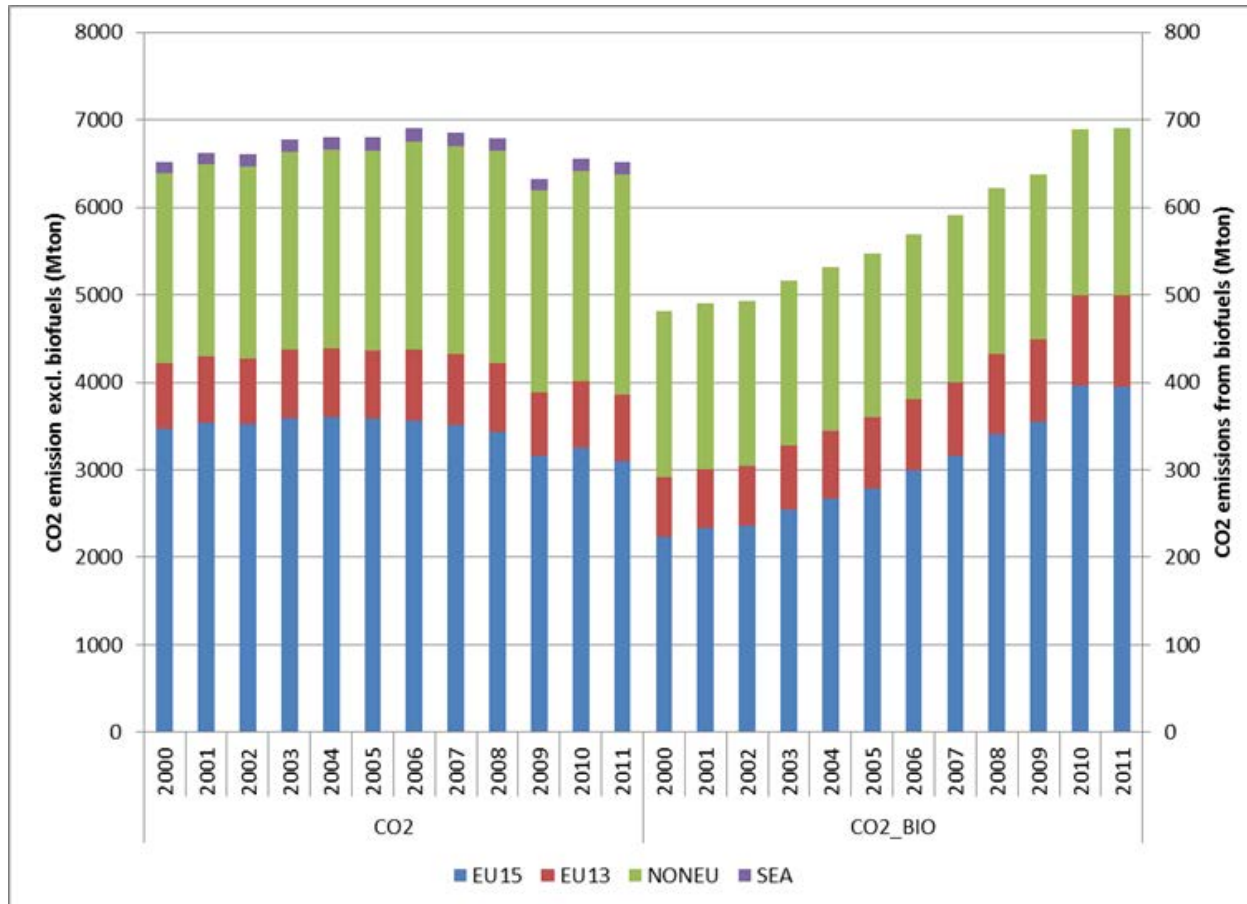


PM2.5 2000-2011 All Sectors 01 MACC-III



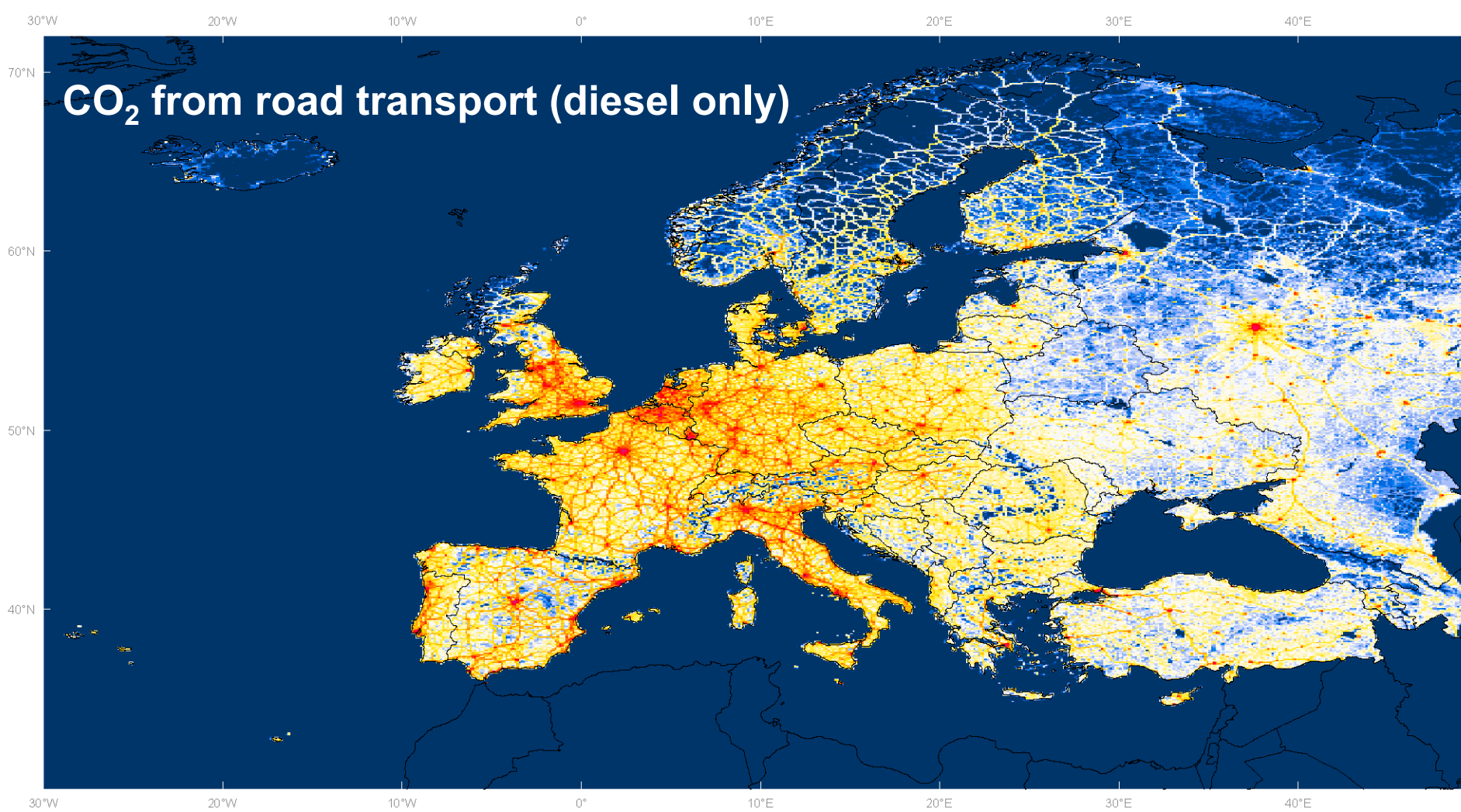
tonnes/cell

# CO<sub>2</sub> EMISSIONS EXCLUDING BIOFUEL COMBUSTION & CO<sub>2</sub> FROM BIOFUEL COMBUSTION (ON RIGHT AXIS)

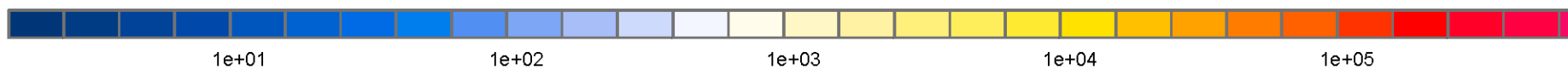


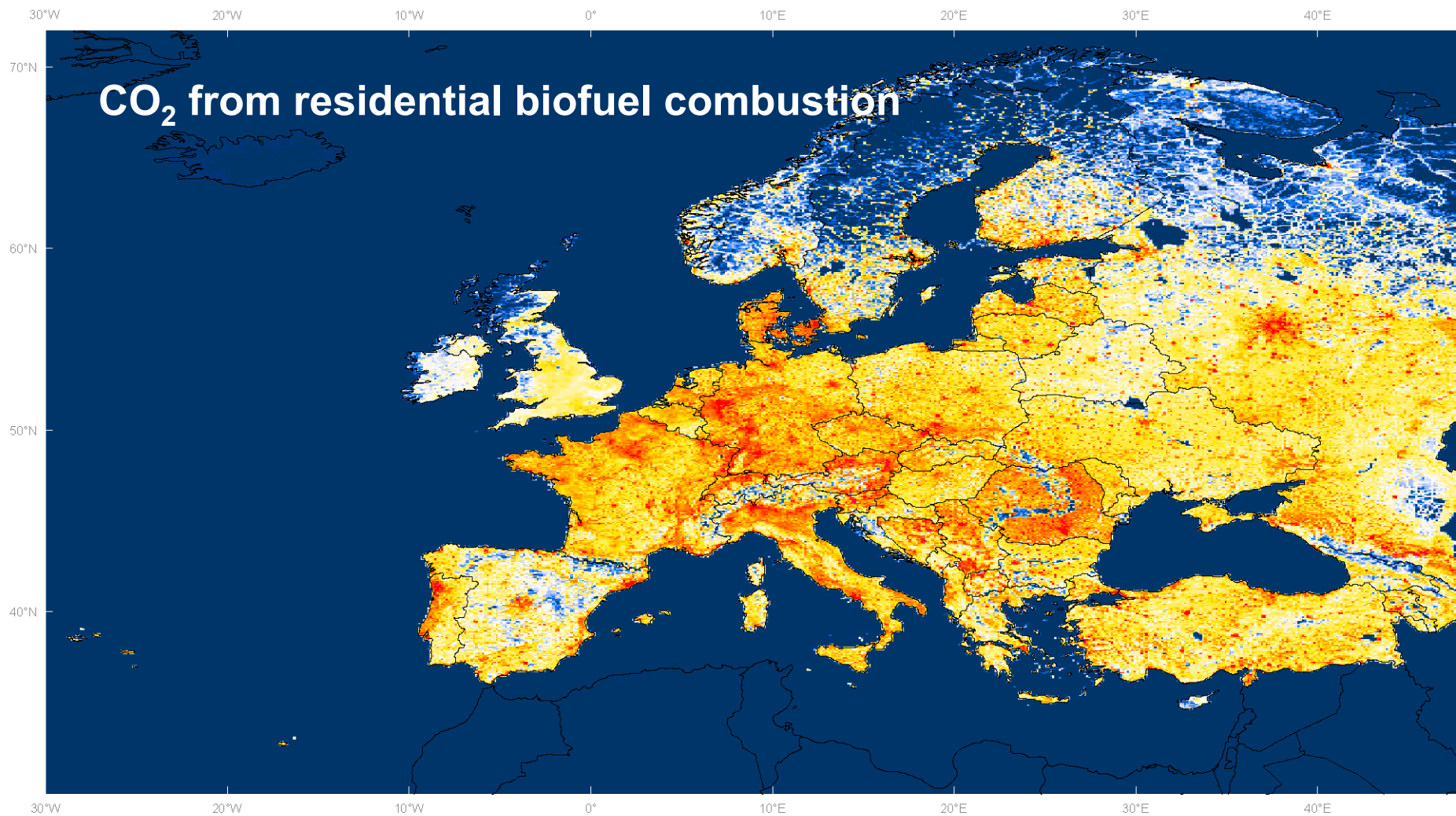
per year and per country group  
(EU15 = EU Member States as of 1-1-1995 plus Norway and Switzerland, EU13 = New EU Member States joined after 1-1-1995, NONEU = all other countries, SEA = sea regions).

**Rather stable Fossil CO<sub>2</sub> (note the crisis in 2009);  
increasing biofuels but still factor 10 difference**



2009 Sector 72 CO<sub>2</sub>





2009 Sector 02 CO2 BIO





# THE (RE)NEW(ED) OPPORTUNITY...



# HISTORIC PARIS AGREEMENT ON CLIMATE CHANGE

Paris, 12 December 2015 –

An historic agreement to combat climate change and unleash actions and investment towards a low carbon, resilient and sustainable future was agreed by 195 nations in Paris today.



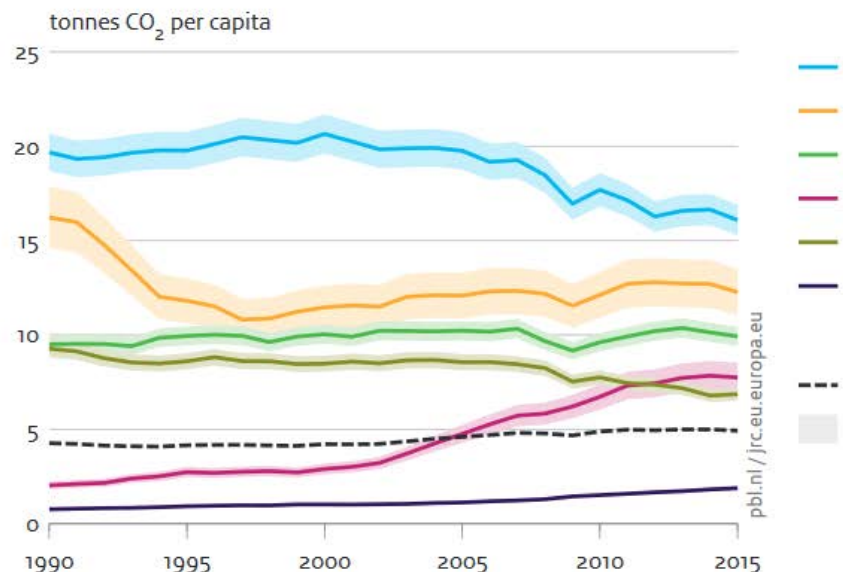
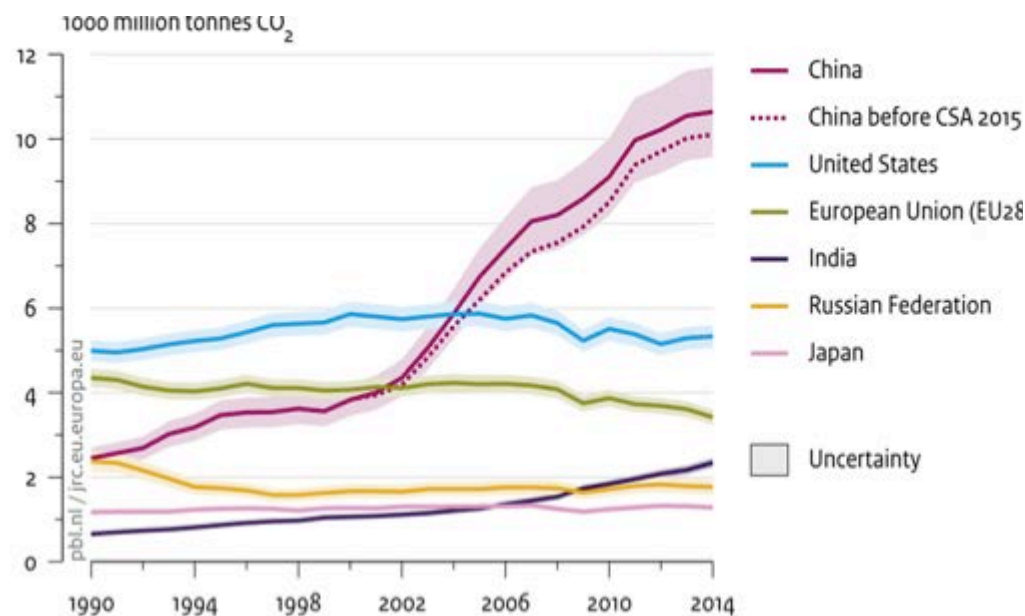
# GREENHOUSE GASES

- › CO<sub>2</sub> and biofuel CO<sub>2</sub> in the European TNO inventories
- › **Towards independent verification**
- › Measuring concentrations around Rotterdam (CO<sub>2</sub>, CO, CH<sub>4</sub>)
- › Developing quick scans for CH<sub>4</sub> emissions from landfills
- › Campaign for Oil and Gas CH<sub>4</sub> leakage in Groningen gas field
- › Cooperation with satellite experts (SRON) to verify point sources
- › Identifying tracers and markers e.g. ethane for fossil NG leakage
- › Carbon footprint of different fuels

# Complexity? –

## zooming out makes the answers simpler....

### CO<sub>2</sub> EMISSIONS FROM FOSSIL-FUEL USE AND CEMENT PRODUCTION IN THE TOP 5 EMITTING COUNTRIES AND THE EUROPEAN UNION



Source: EDGAR v4.3.2 FT2015 (JRC/PBL 2016; IEA 2014 (suppl. with IEA 2016 for China, BP 2016, NBS 2016); UNPD 2015 (WPP, Rev. 2015)

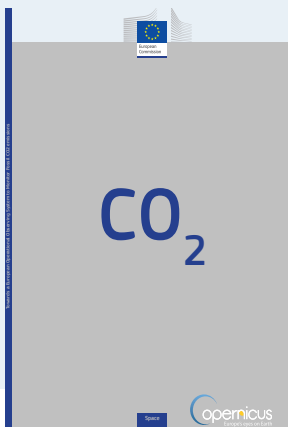
Source: EDGAR v4.3.2 FT2015 (JRC/PBL 2016; IEA 2014 (suppl. with IEA 2016 for China, BP 2016, NBS 2016); UNPD 2015 (WPP, Rev. 2015)



**Climate Change... the bigger challenge!**

# CO<sub>2</sub>

1. What are the **critical uncertainties** and limitations of **current inventories** of fossil CO<sub>2</sub> emissions based on fuel use statistics?
2. How could inventories be improved using independent **space-borne measurements of atmospheric CO<sub>2</sub>**?



## “CO<sub>2</sub> report” : Towards a European Operational Observing System to Monitor Fossil CO<sub>2</sub> Emissions

<http://www.copernicus.eu/main/towards-european-operational-observing-system-monitor-fossil-co2-emissions>

# The top-down atmospheric approach

# Improving emissions inventories

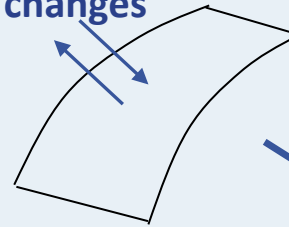
Top-down

Measurement systems for atmospheric CO<sub>2</sub>



Inverse modeling

Sources and sinks for CO<sub>2</sub> on a gridded basis with temporal changes



Improved Inventories

UNFCCC

Regional authorities

Emission Trading markets

Effort Sharing Decision sectors

Public

Bottom-up

Emission inventories from statistical data

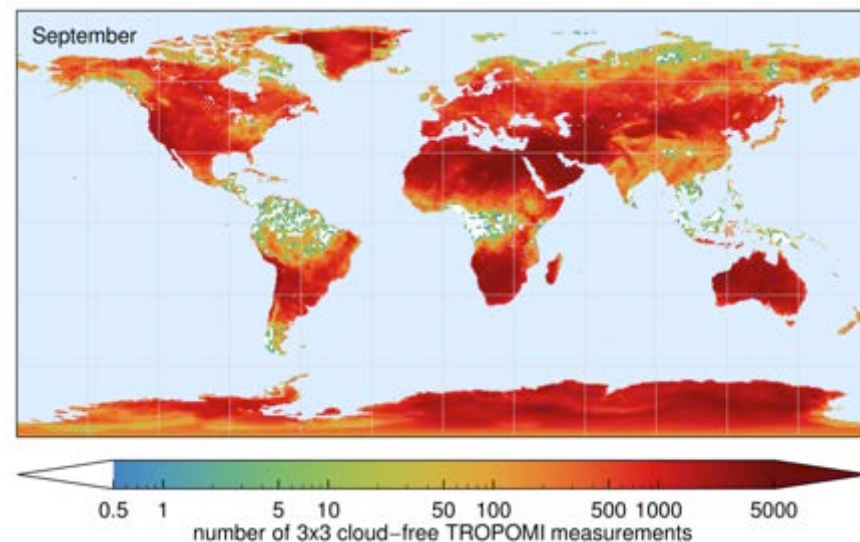
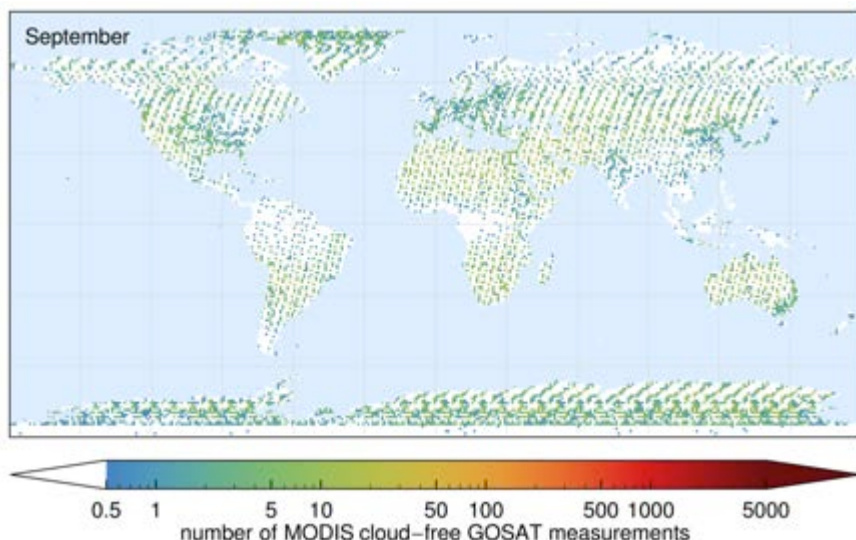


Maps of emissions per sector of human activity

- CO<sub>2</sub> emissions of subnational governments / regions need also to be mapped within a regionally complete picture (covering all human activities).

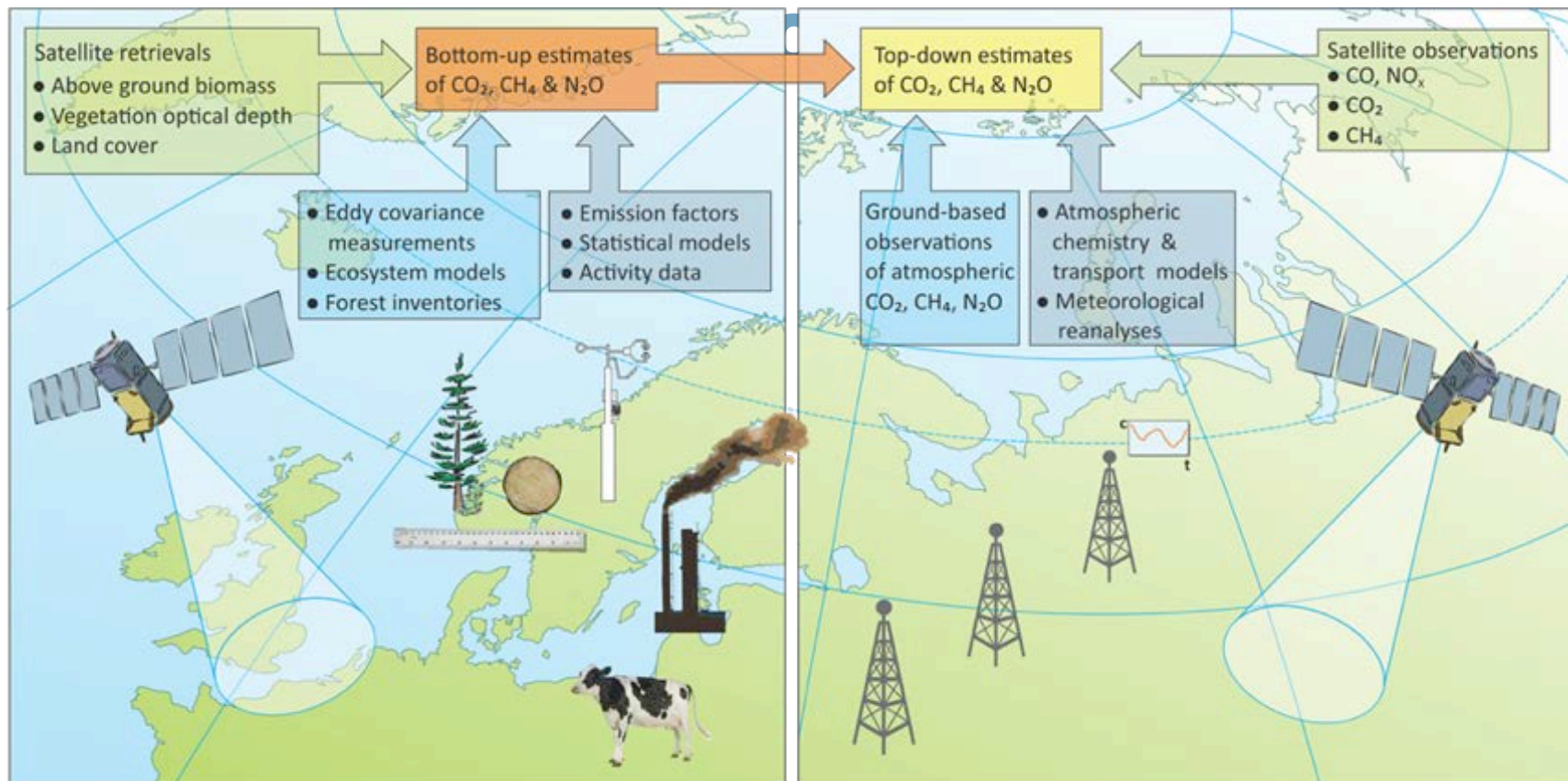
	active period	Type observation	Footprint size (km x km)	# meas. per time unit	Sensitivity per meas. (XCH <sub>4</sub> %)	Coverage
SCIAMACHY	2002-2012	Scanning (960 km)	~60x30 ( <b>1800 km<sup>2</sup></b> )	1.35 per sec ( <b>6</b> )	low	Global in 6 days
GOSAT	2009-...	pointing	Diameter ~10 km ( <b>80 km<sup>2</sup></b> )	1 per 4.5 sec ( <b>1</b> )	moderate	Global but with holes
TROPOMI	2017-...	Pushbroom (2600 km)	~7x7 ( <b>50 km<sup>2</sup></b> )	216 per sec ( <b>980</b> )	moderate	Global in 1 day

Important : **coverage** (incl. absolute number of useful meas.), footprint size, sensitivity  
(Only few % cloud free observations)



**TROPOMI should be much better suited to detect point sources CH<sub>4</sub>**

# TOWARDS VERIFICATION OF REPORTED



# **EARTH OBSERVATION DATA...**

## **NEW OPPORTUNITIES FOR EMISSION INVENTORIES**



# AIR POLLUTION WHAT IS IT MADE OF?

NO<sub>x</sub> emissions = NO + NO<sub>2</sub>;  
*It is what it is....*

**PM = ....?**



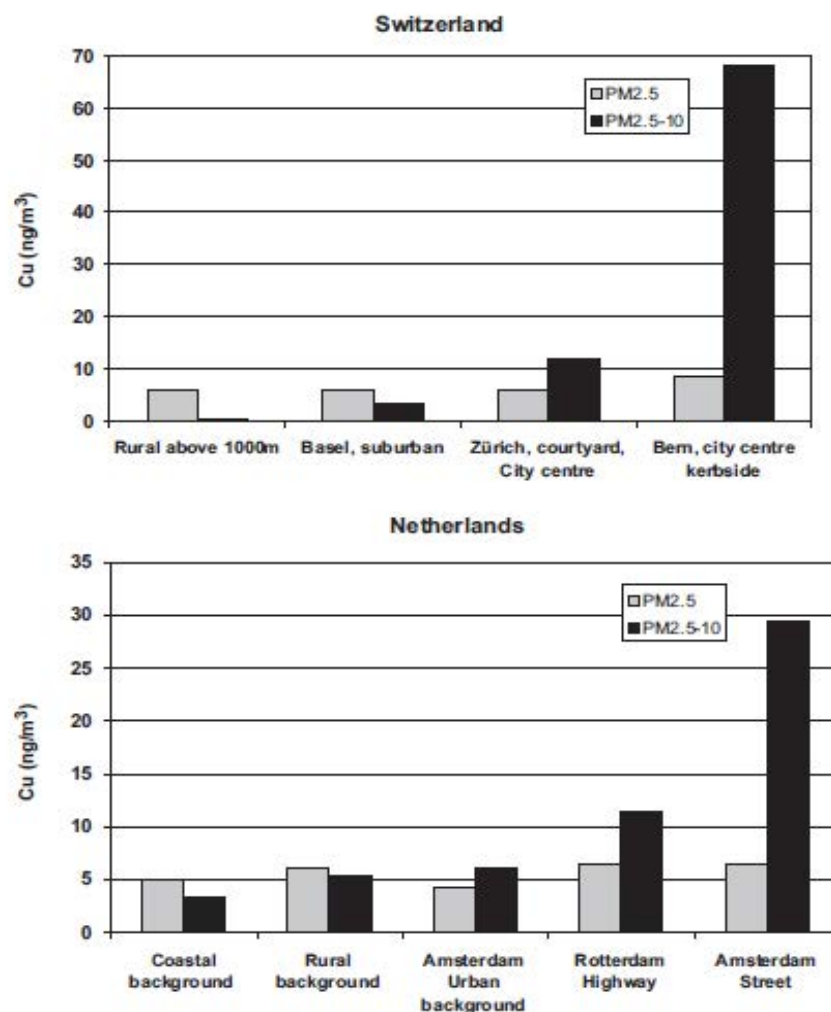


Fig. 4. Concentration of Copper in ambient PM samples at various locations in the coarse fraction (PM2.5–10) and the fine fraction (PM2.5) for Switzerland (top) and the Netherlands (bottom). (Recalculated from data by Hueglin et al., 2005 and Visser et al., 2001, respectively.)

**WHEN BRAKE  
WEAR TURNED  
OUT TO BE THE  
MISSING  
COPPER  
SOURCE....**

*Denier van der Gon et al.,  
Atmospheric Environment 41 (2007)  
8697–8710*



## ***Study about traffic particles and cardiovascular health***

- NC highway patrol troopers
- Troopers work up to 9 hrs inside their cars
- Health effects associated with metals in PM<sub>2.5</sub> (brake wear)

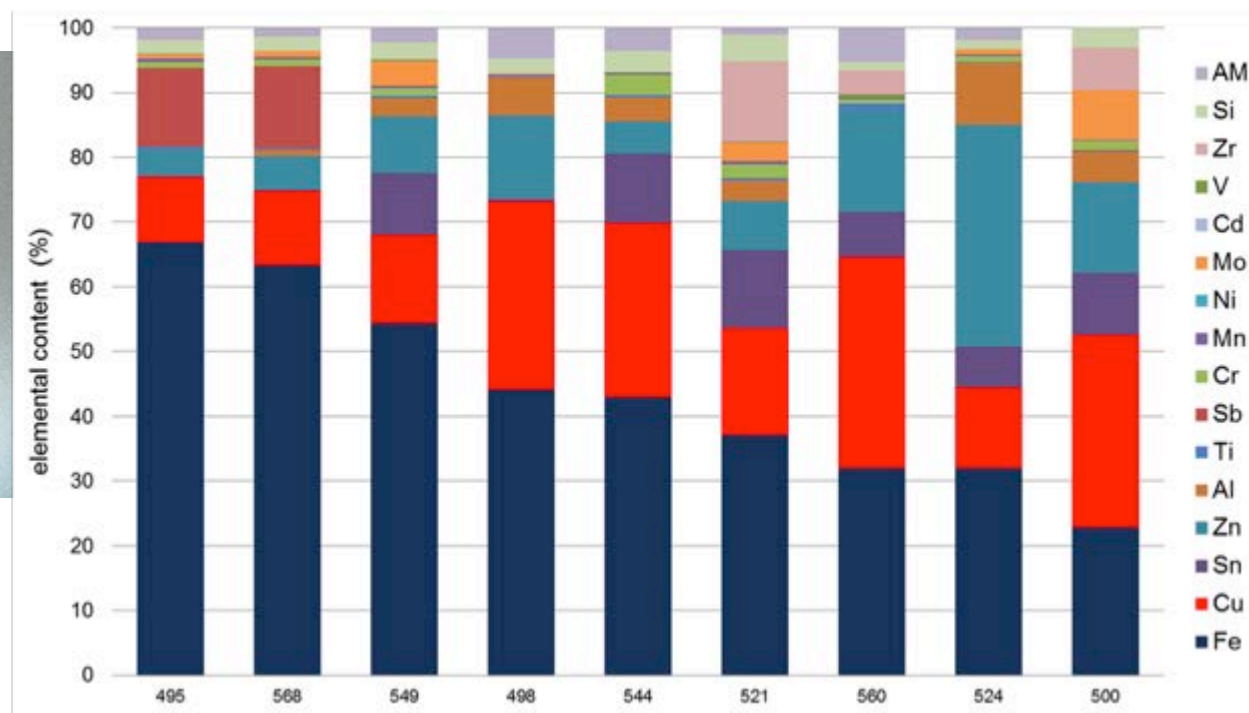


*Riediker, et al. 2003. Environ Sci Technol. 37:2084-2093.*

*Riediker, et al. 2004. Am J Respir Crit Care Med. 169:934-940.*

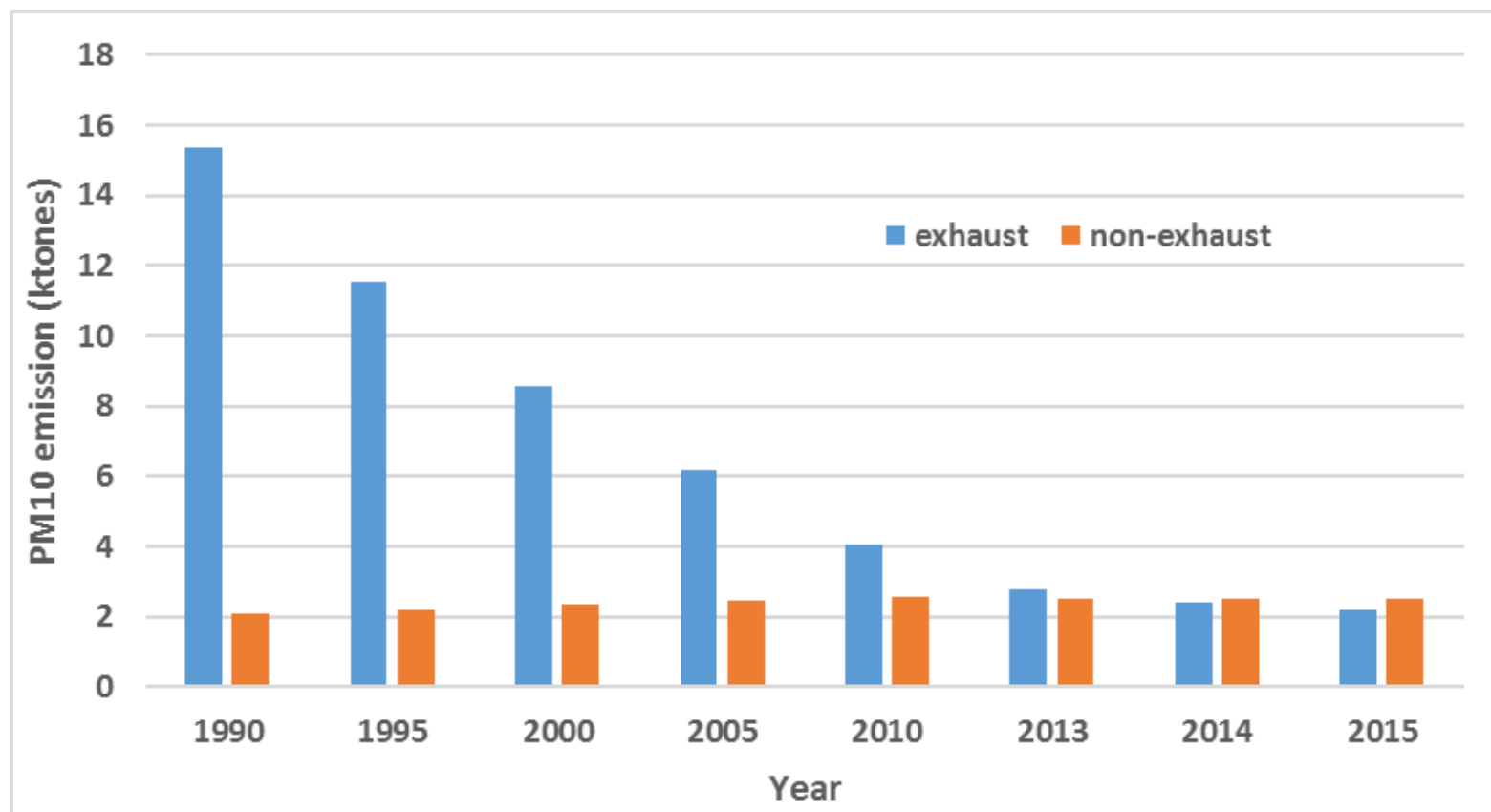
*Riediker et al. 2004. Part Fib Toxicol. 1:2.*

# METAL CONTENT (% M/M) OF SELECTED BRAKE PAD SAMPLES BY XRF ANALYSIS



Hulskotte, J.H.J., G.D. Roskam, H.A.C. Denier van der Gon, Elemental composition of current automotive braking materials and derived air emission factors, Atmospheric Environment, 99, 436–445, 2014

# TREND IN ROAD TRANSPORT PM EMISSIONS IN THE NETHERLANDS 1990-2015



A major change in composition – from combustion to wear dominated.....

## MORE ON NON-EXHAUST EMISSIONS....



### REVIEW PAPER

The Policy Relevance of Wear Emissions from Road Transport, Now and in the Future—An International Workshop Report and Consensus Statement

Hugo A.C. Denier van der Gon,<sup>1,\*</sup> Miriam E. Gerlofs-Nijland,<sup>2</sup> Robert Gehrig,<sup>3</sup> Mats Gustafsson,<sup>4</sup> Nicole Janssen,<sup>2</sup> Roy M. Harrison,<sup>5,6</sup> Jan Hulskotte,<sup>1</sup> Christer Johansson,<sup>7,8</sup> Magdalena Jozwicka,<sup>1</sup> Menno Keuken,<sup>9</sup> Klaas Krijgsheld,<sup>10</sup> Leonidas Ntziachristos,<sup>11</sup> Michael Riediker,<sup>12</sup> and Flemming R. Cassee<sup>2,13</sup>

***Journal of the Air & Waste Management Association, 63(2):136–149, 2013.***



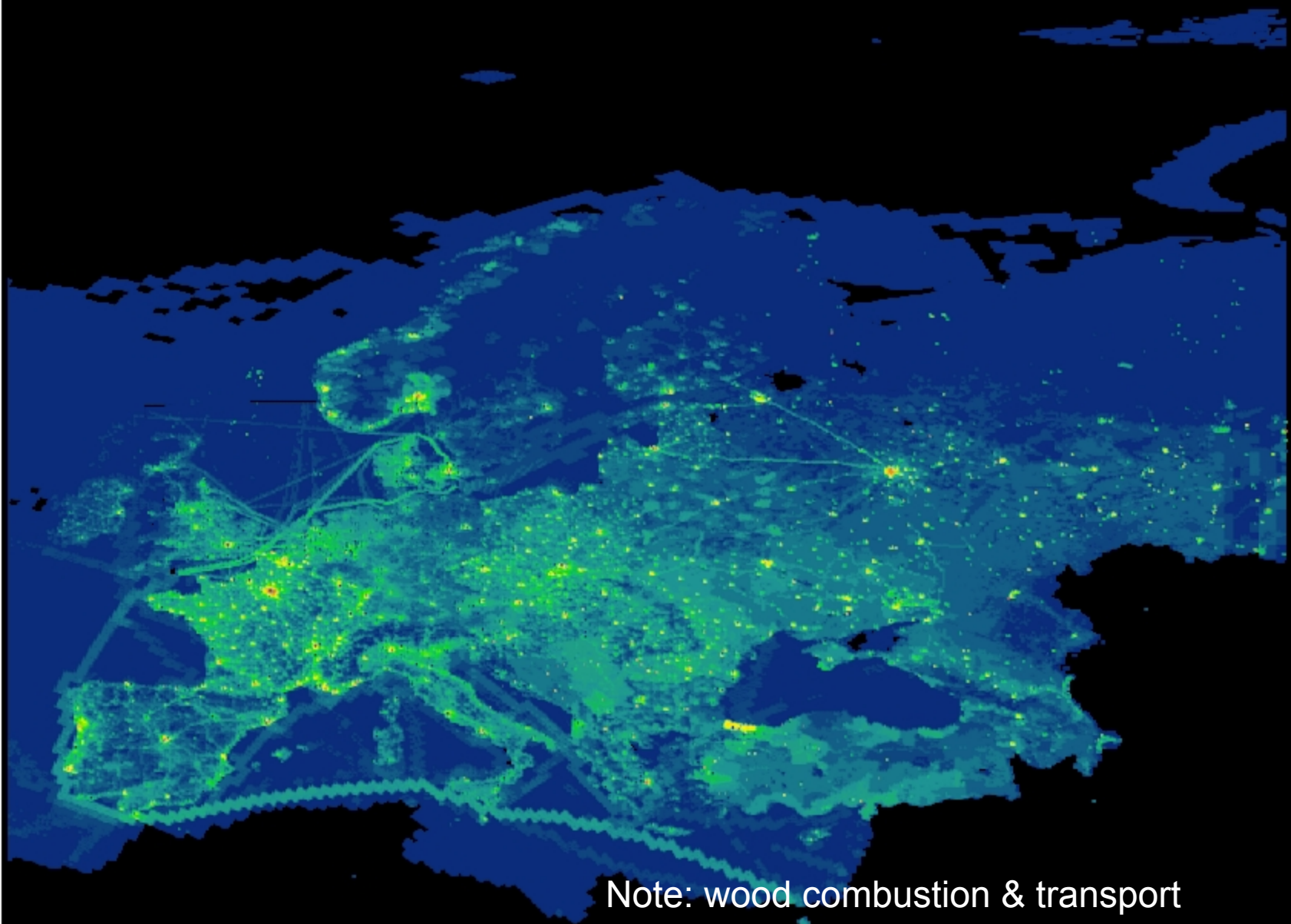
### Review

Urban air quality: The challenge of traffic non-exhaust emissions

Fulvio Amato<sup>a,\*</sup>, Flemming R. Cassee<sup>b,1</sup>, Hugo A.C. Denier van der Gon<sup>c</sup>, Robert Gehrig<sup>d</sup>, Mats Gustafsson<sup>e</sup>, Wolfgang Hafner<sup>f</sup>, Roy M. Harrison<sup>g,k</sup>, Magdalena Jozwicka<sup>c</sup>, Frank J. Kelly<sup>h</sup>, Teresa Moreno<sup>a</sup>, Andre S.H. Prevot<sup>i</sup>, Martijn Schaap<sup>c</sup>, Jordi Sunyer<sup>j</sup>, Xavier Querol<sup>a</sup>

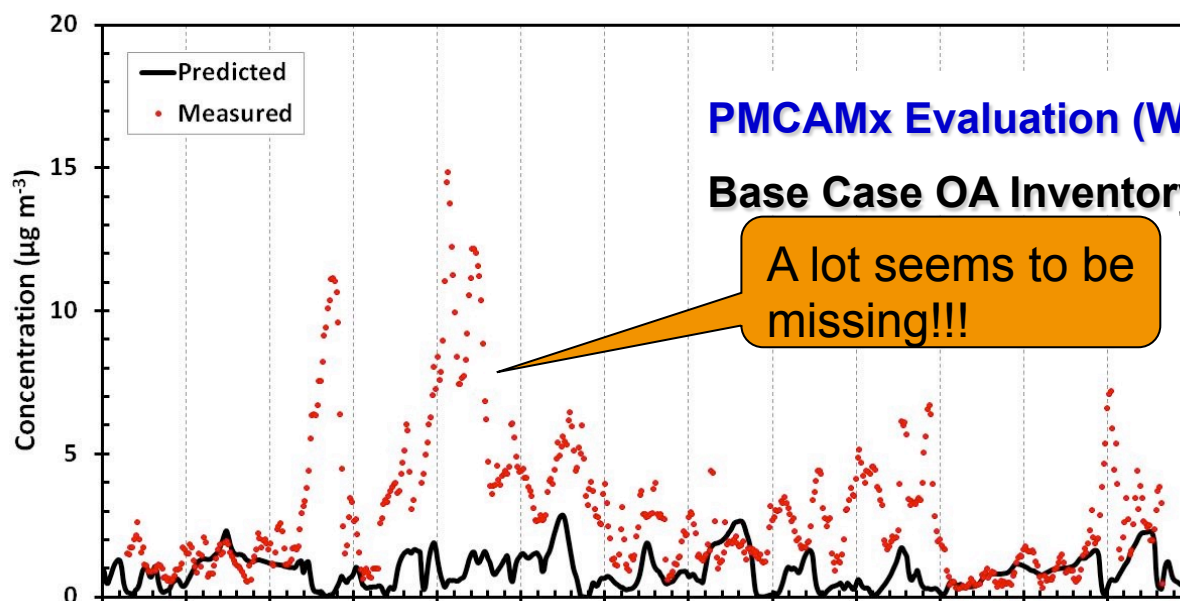
***Journal of Hazardous Materials 275 (2014) 31–36***

# PM\_OC\_2.5



# WHY RETHINK PM FROM WOOD COMBUSTION?

- › In 2010 good results EUCAARI EC / OC inventory but....major model vs observed discrepancies during episodes influenced by residential wood combustion (RWC).



**PMCAMx Evaluation (Winter 2009)**

**Base Case OA Inventory; site Vavihill (Sweden)**

A lot seems to be missing!!!

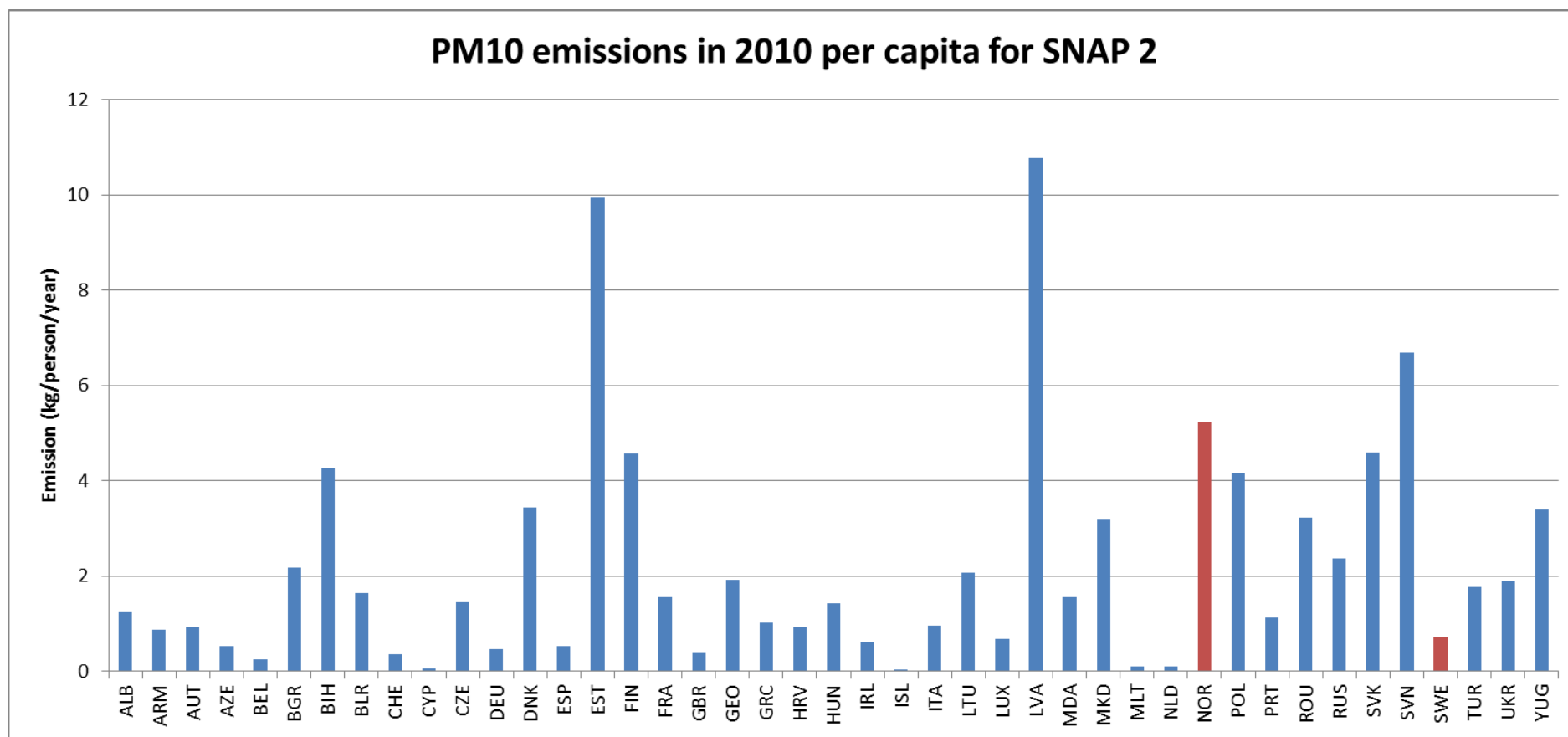
Similar performance in  
e.g. Payerne, Melpitz

Regional CTMs have a problem with mass closure for PM – mostly an “OC problem”. At the time we thought the VBS approach would solve it, now we see it helps but there is still a substantial gap, especially close to the sources.....

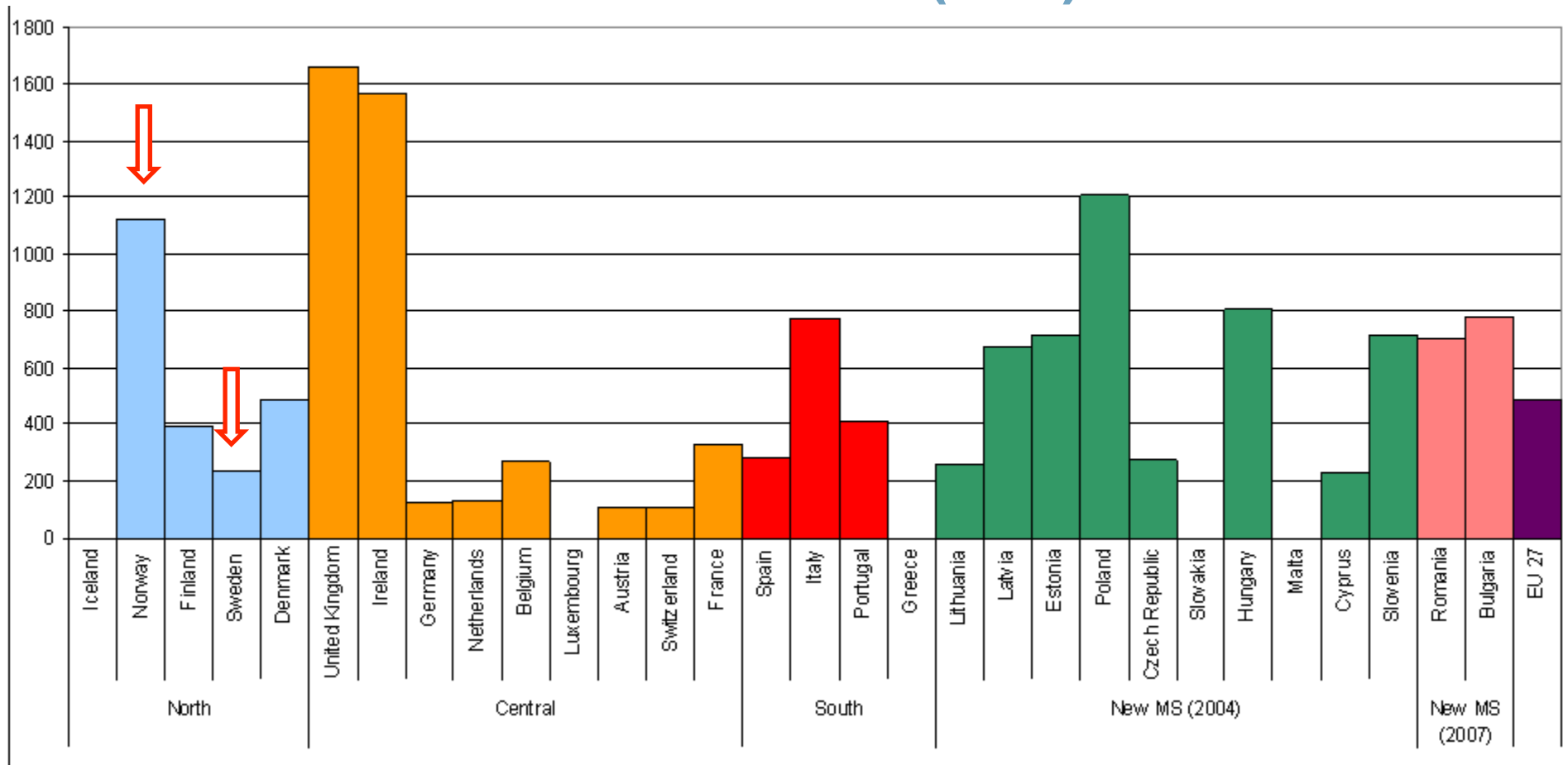
# RETHINK THE RWC INVENTORY

- › A first comparison/ analysis showed incompatibility of different country approaches (not “right or wrong” )
- › Start partly from scratch
- › Don’t use national reported data
- › Do not define EC and OC as a fraction of PM10 or PM2.5 that should “fit” in the official reported data
- › Make a selection of emission factors and apply in a consistent way for all countries

# PM10 EMISSION PER CAPITA FOR RESIDENTIAL COMBUSTION IN 2010

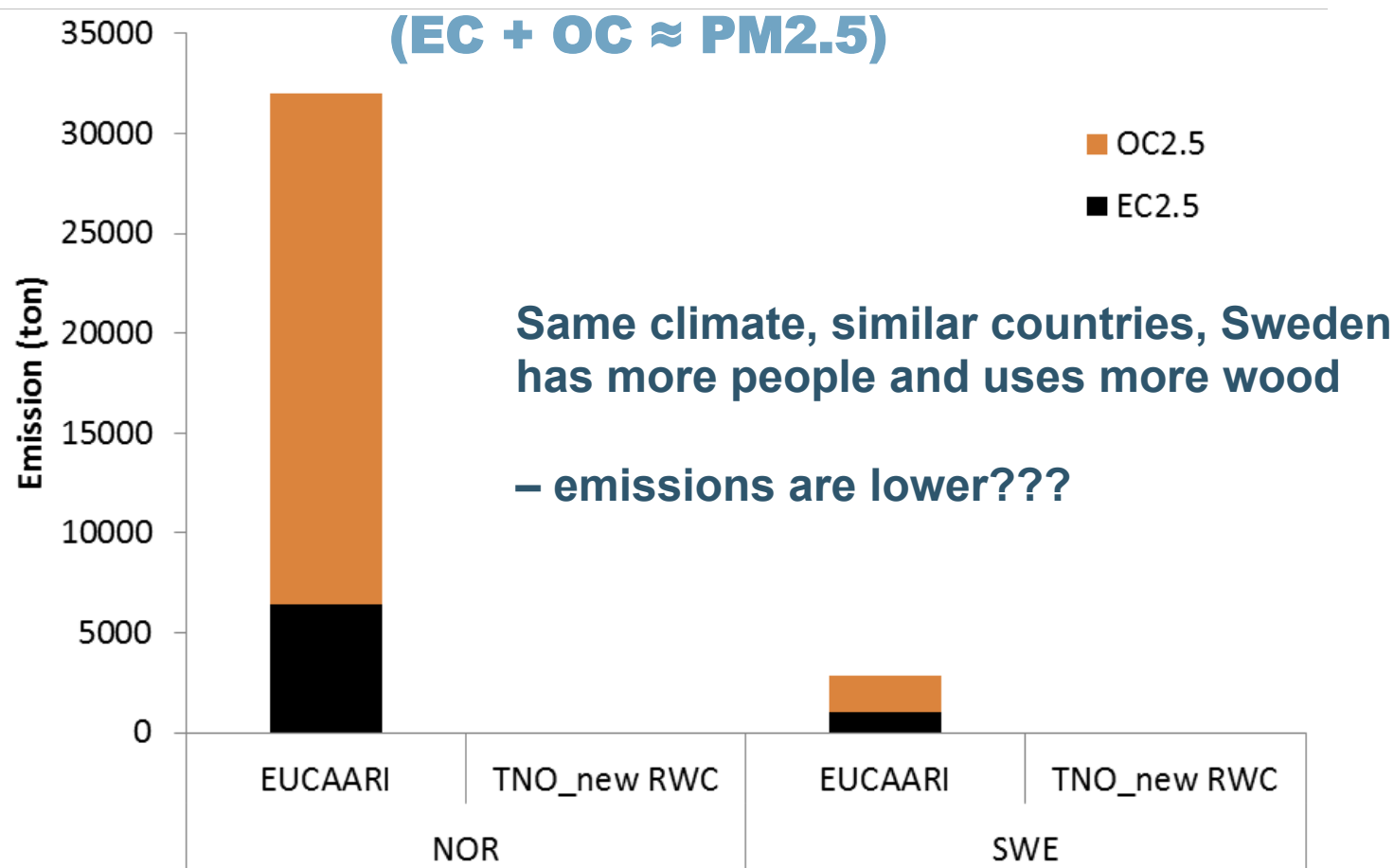


# EMISSIONS OF PM10 PER UNIT OF WOOD COMBUSTED IN THE RESIDENTIAL SECTOR IN 2009 (G/GJ)



› Large differences in “implied” emission factors

# NORWAY & SWEDEN: TOTAL RESIDENTIAL WOOD COMBUSTION EMISSIONS IN 2005



**EUCAARI inventory approach:**

- 1) Estimate PM2.5
- 2) use fractions to estimate EC and OC

# FILTERABLE PM AND CONDENSABLE PM

- › the US EPA defines particulate matter (PM) as consisting of a filterable fraction (FPM) and a condensable fraction (CPM).



Filterable PM is directly emitted:

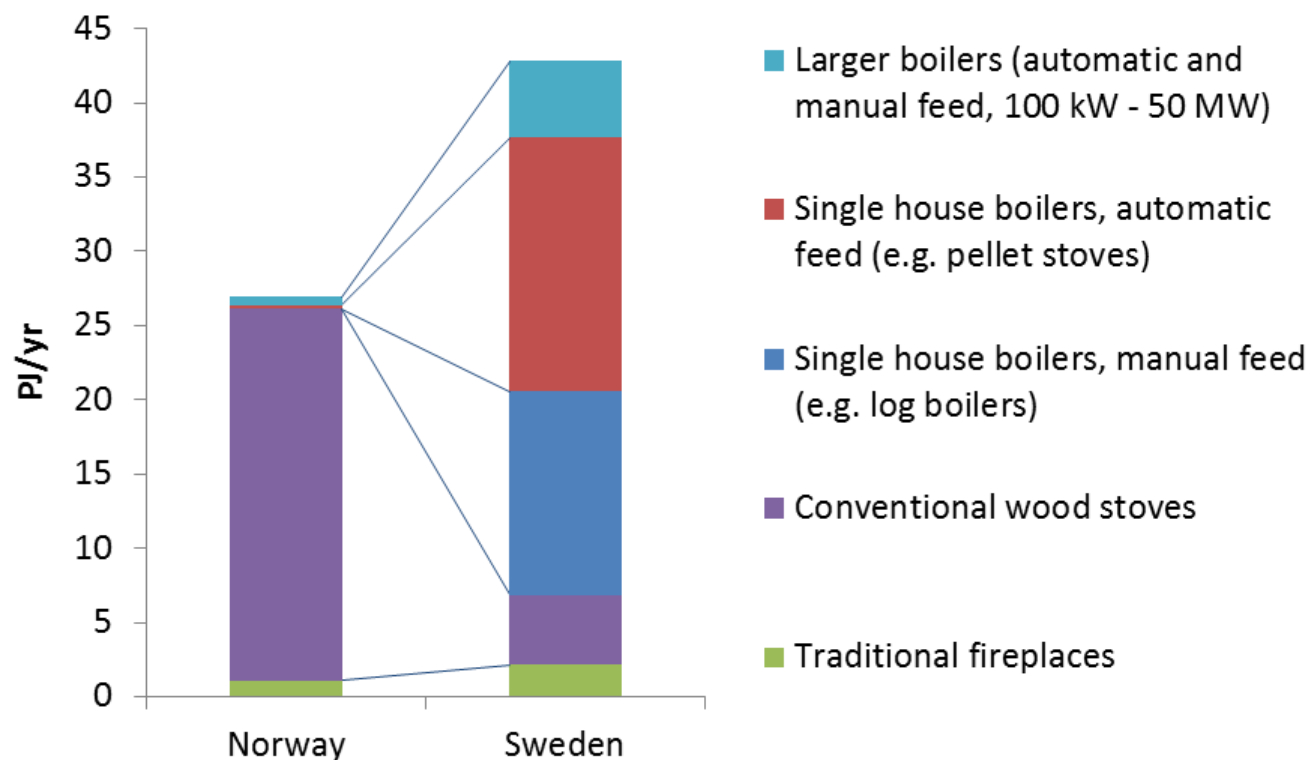
- Solid or liquid
- Captured on filter
- PM<sub>10</sub> or PM<sub>2.5</sub>

Condensable PM is in vapor:

- Reacts upon cooling and dilution
- Forms solid or liquid particle
- Always PM<sub>2.5</sub> or less

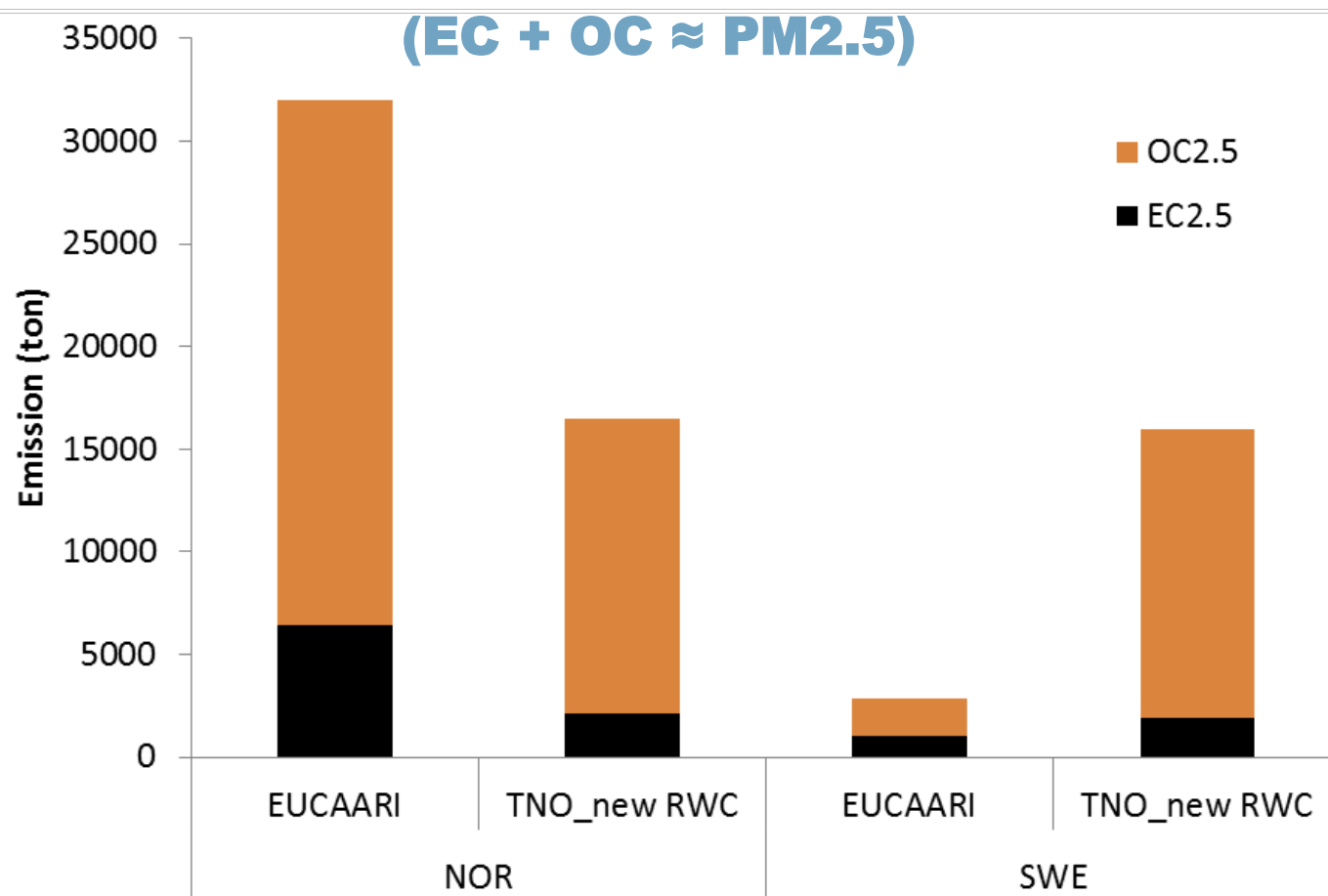
*where should the PM mass be that forms almost instantaneously?*

# WOOD USE IN NORWAY & SWEDEN

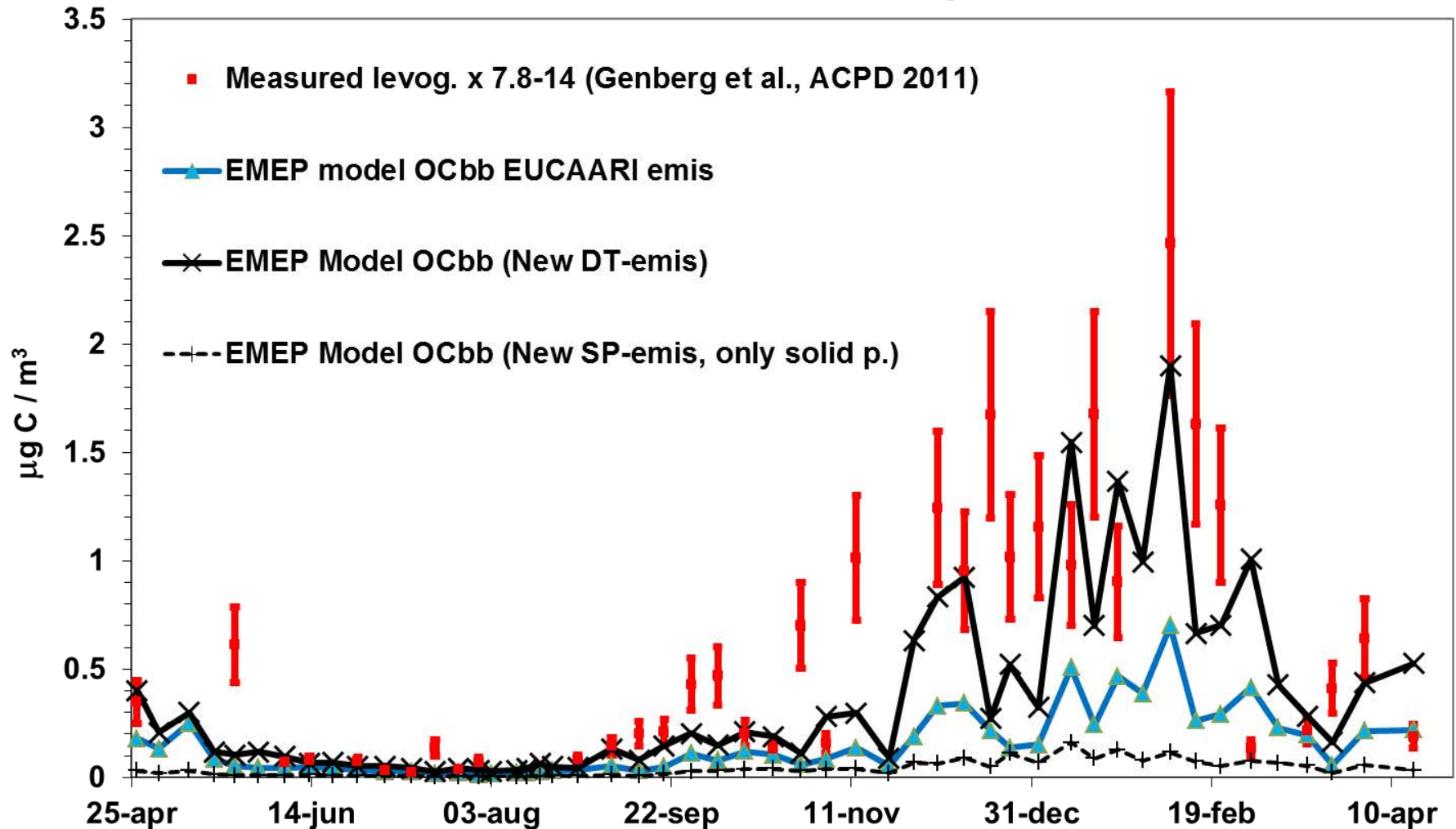


- › Wood use in Sweden larger but different appliance
- › Countries use their own methods to calculate PM emissions

# NORWAY & SWEDEN: TOTAL RESIDENTIAL WOOD COMBUSTION EMISSIONS IN 2005



# Vavihill OCbb and levoglucosan 2008-2009

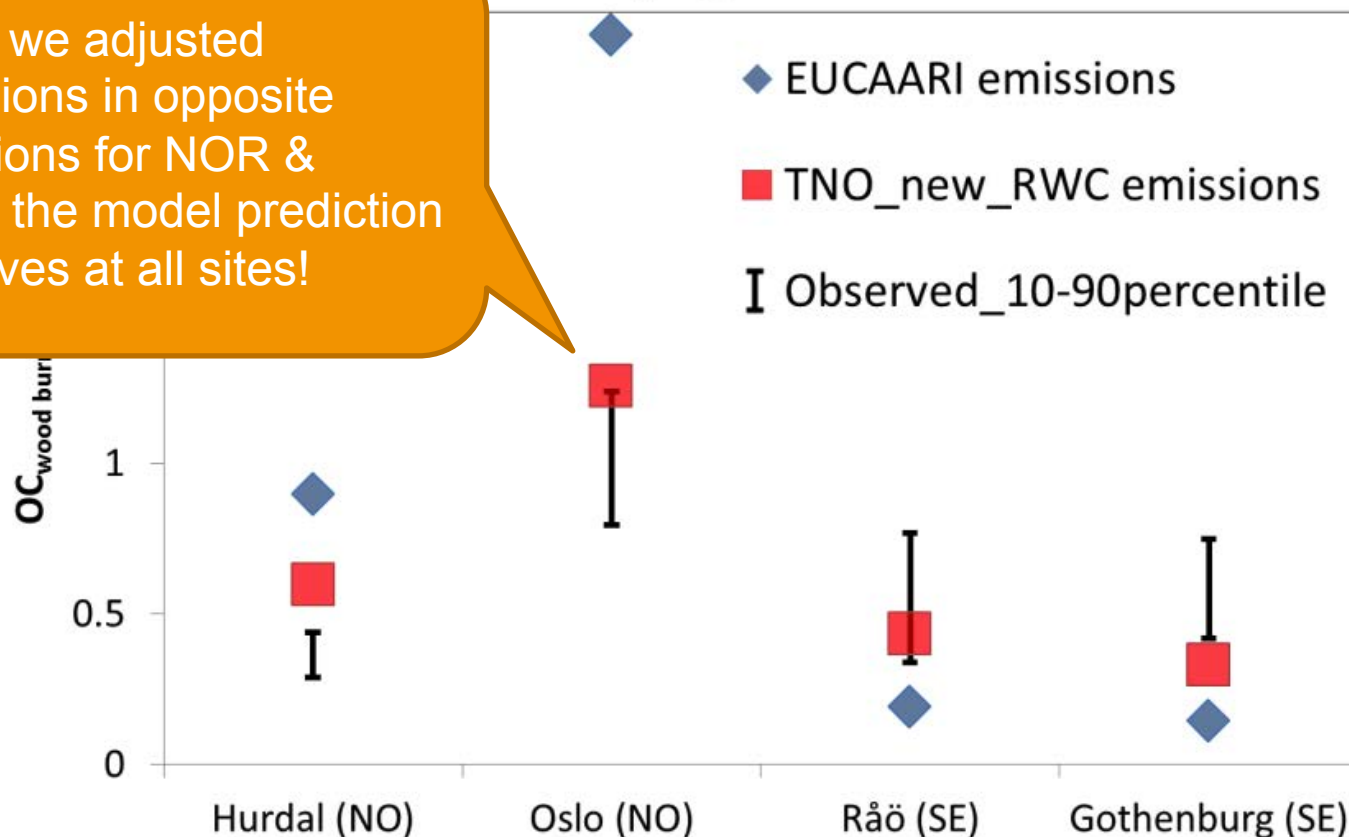


*Denier van der Gon et al., 2015*

## Wood Burning OC - source-apportionment studies

Winter campaigns in Scandinavia

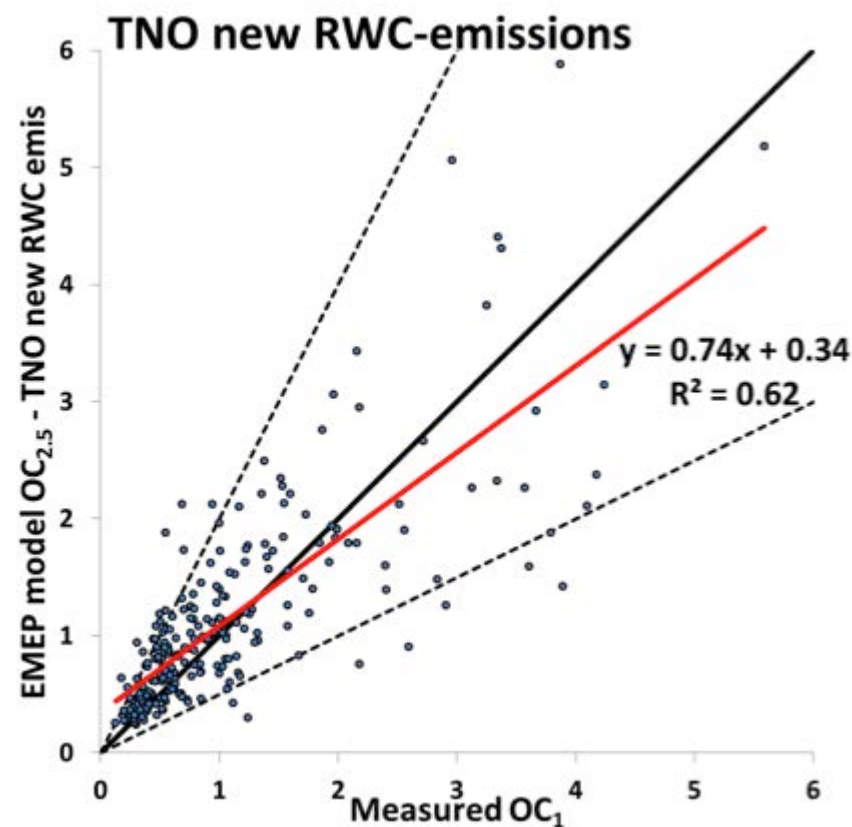
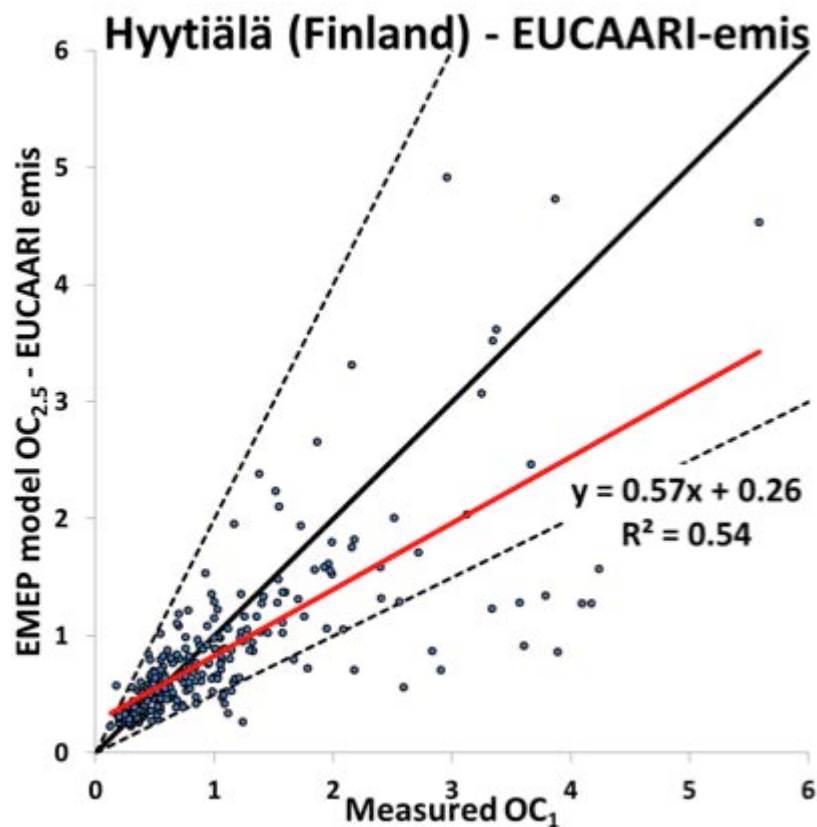
While we adjusted emissions in opposite directions for NOR & SWE, the model prediction improves at all sites!



*Comparison of model calculated OC from wood burning to source-apportionment data from measurement campaigns during winter in Norway and Sweden*

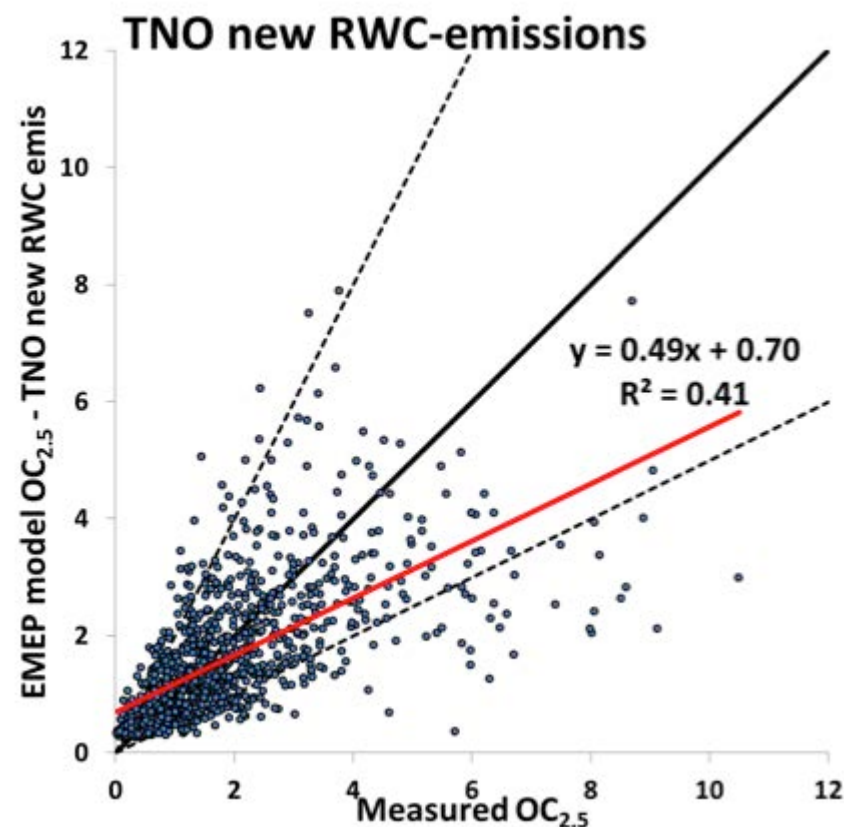
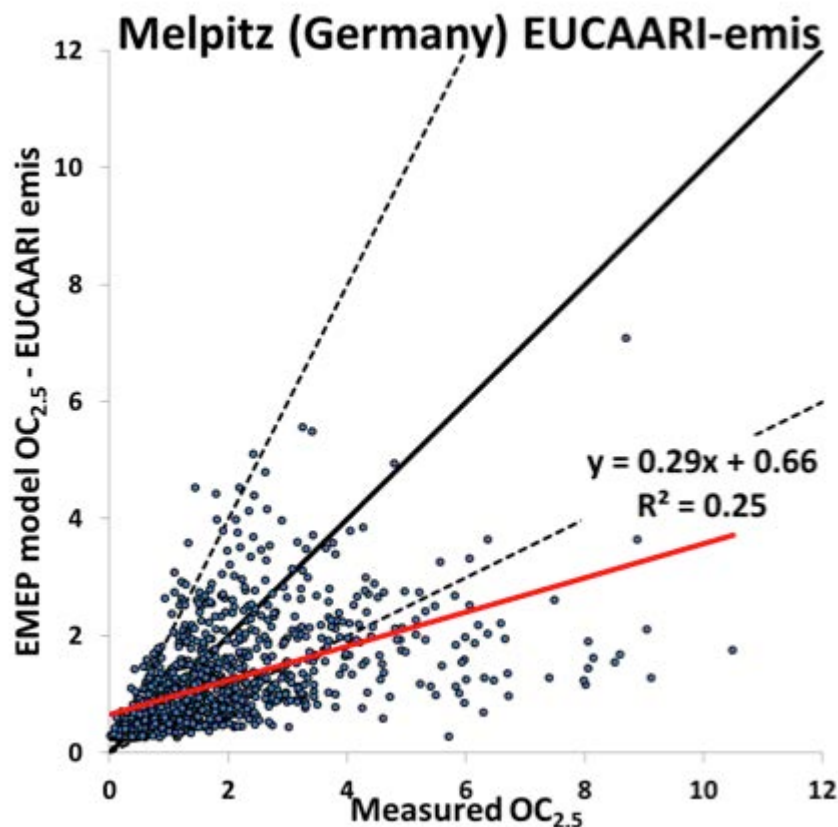
(SORGA, 1 - 8 March 2007, Yttri et al., 2011) (GÖTE, 11 Feb - 4 Mar 2005, Szidat et al., 2009). Unit:  $\mu\text{g}(\text{C}) \text{ m}^{-3}$

# MEASURED AND MODELLED PM<sub>OC</sub> CONCENTRATIONS WITH THE EMEP MSC-W MODEL



*Denier van der Gon et al., ACP, 2015*

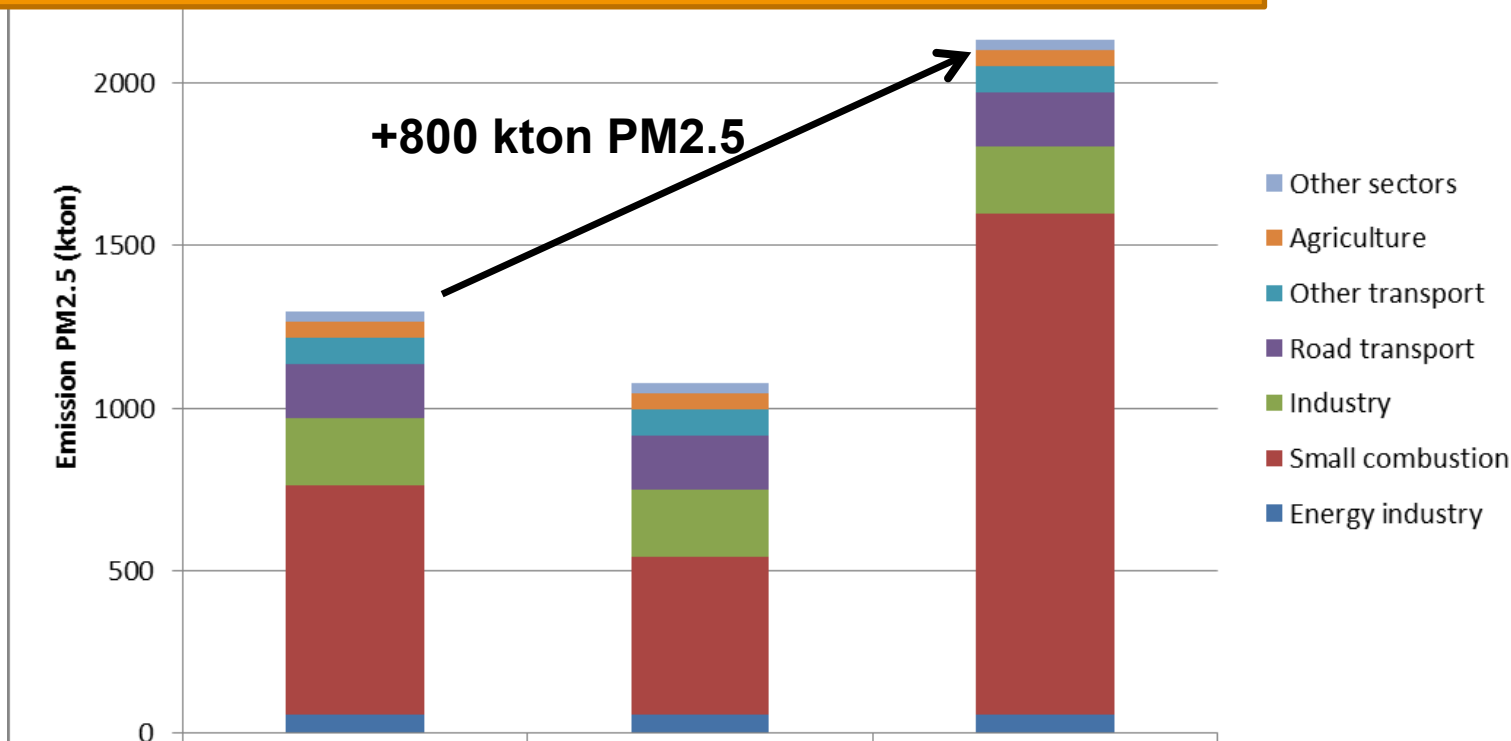
# MEASURED AND MODELLED PM<sub>OC</sub> CONCENTRATIONS WITH THE EMEP MSC-W MODEL



*Denier van der Gon et al., ACP, 2015*

# IMPLICATIONS FOR EU-28 (2013)

**+ 60% more PM<sub>2.5</sub> after adjusting one source**



**This is a major impact and needs further investigation and verification!**

Research is needed both at

- **the emissions side** (PM condensable by source type, better activity and appliance types information and emission factors)
- **the model side** (PM, NMVOC, SVOC, IVOC and volatility base set approach)

# EMISSION TIMING!

- › Models need spatial and temporal explicit emissions.
- › Impact of temporal variation is underestimated and undervalued.
- › Fixed (no year to year variation) default temporal profiles are much better than no temporal profile but.....
- › But, especially for episodes the timing of emission can be crucial.....
- › Often debated – is it “emission” work or “model” work; - Answer: Both!

On going work, difficult to get funded and very suitable for collaboration and/or PhD projects because you can learn much about the system...

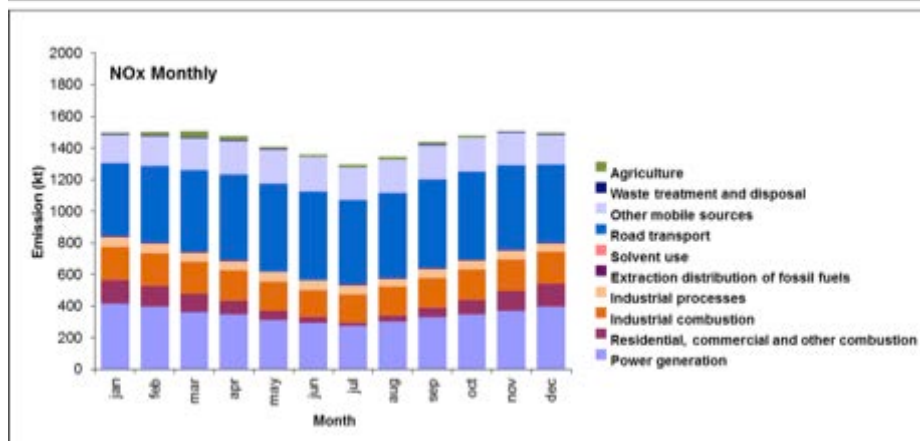
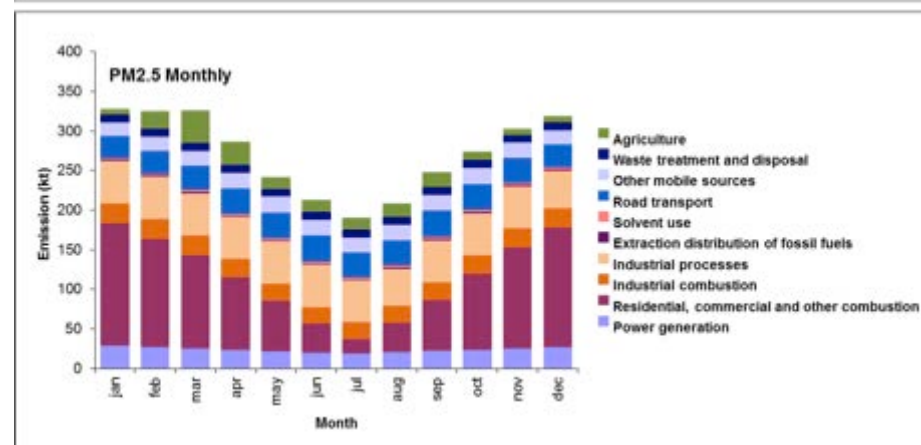
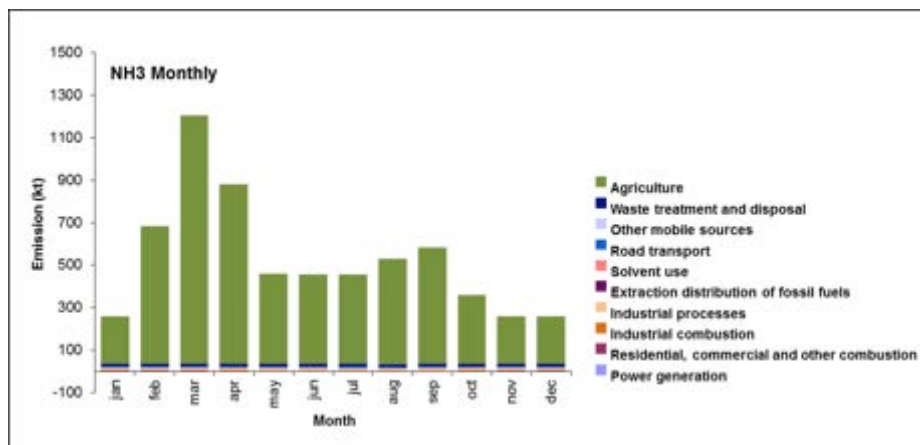
Most prominent examples

- › NH<sub>3</sub> volatilization after manure / fertilizer spreading)
- › Timing of domestic heating
- › Dependencies of traffic on weather conditions

# EMISSION TIMING!



# DEFAULT TEMPORAL PROFILES



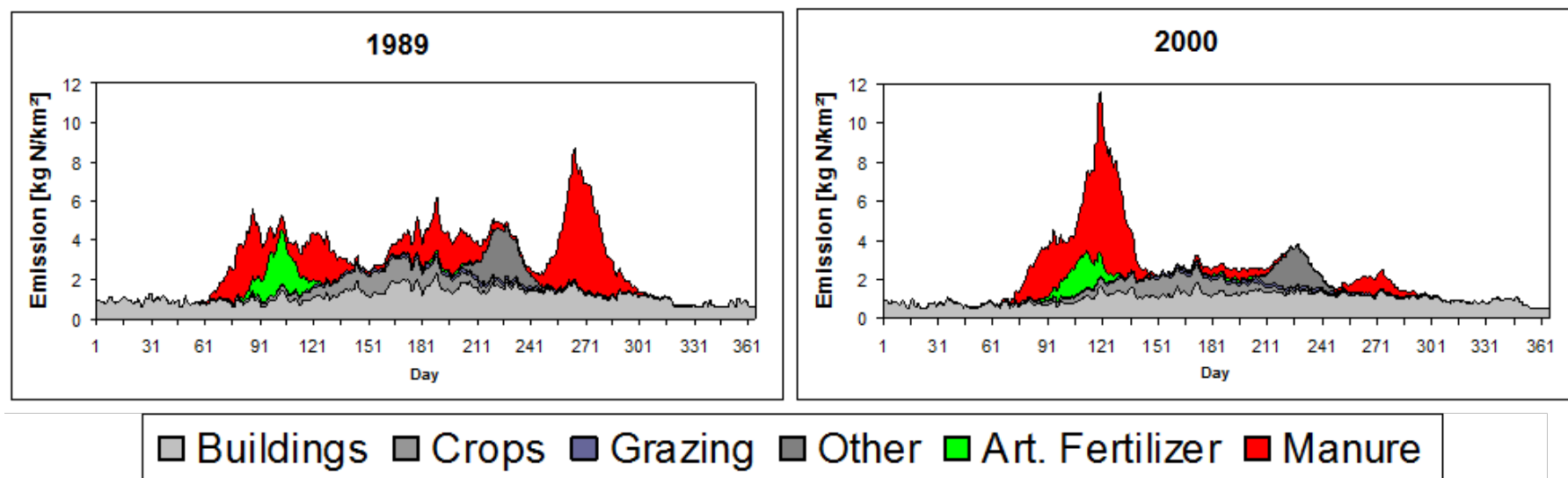
Much better than nothing but.....

- Same for all countries
- Same for all years

Known to be unrealistic

- Impact of improvements on annual average modelled concentrations in general limited

# HOURLY AMMONIA EMISSIONS AS A FUNCTION OF TIME FOR 1989 AND 2000 IN DENMARK (SKJØTH ET AL., 2008).

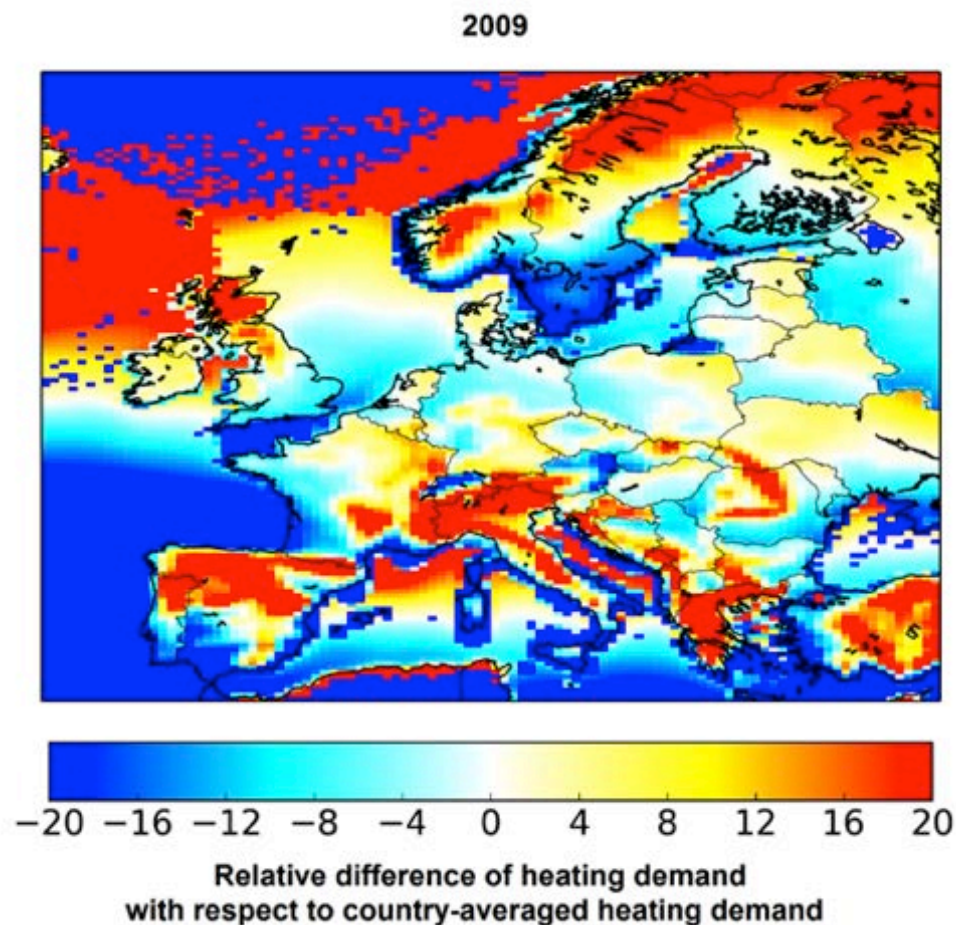


A large shift in emission timing during the year is observed due to a change in the regulation in Denmark and the subsequent change in the agricultural practice

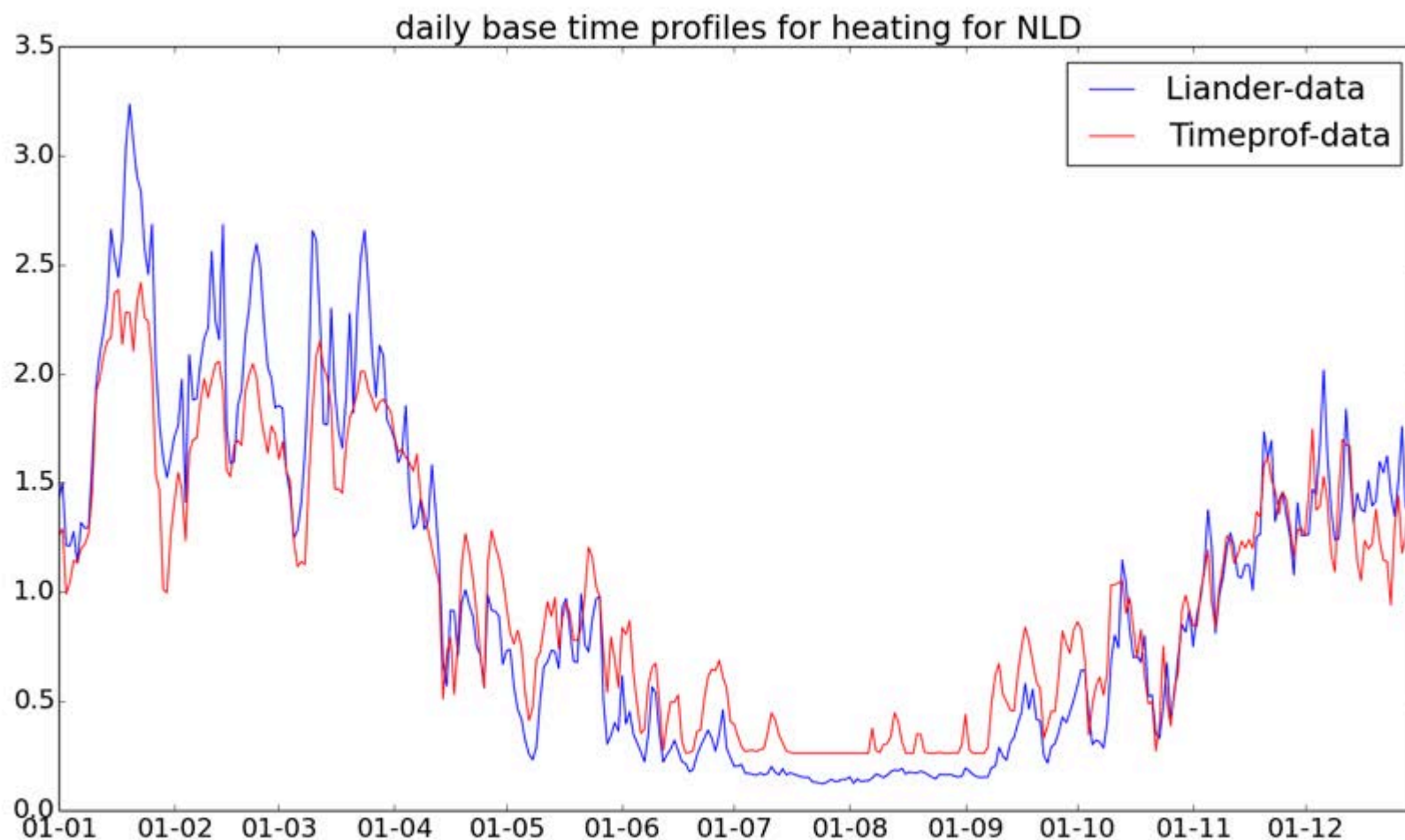
- › We know how to implement but several “complications”
- › In the model? In the emission database?

# RESIDENTIAL COMBUSTION EMISISON TIMING

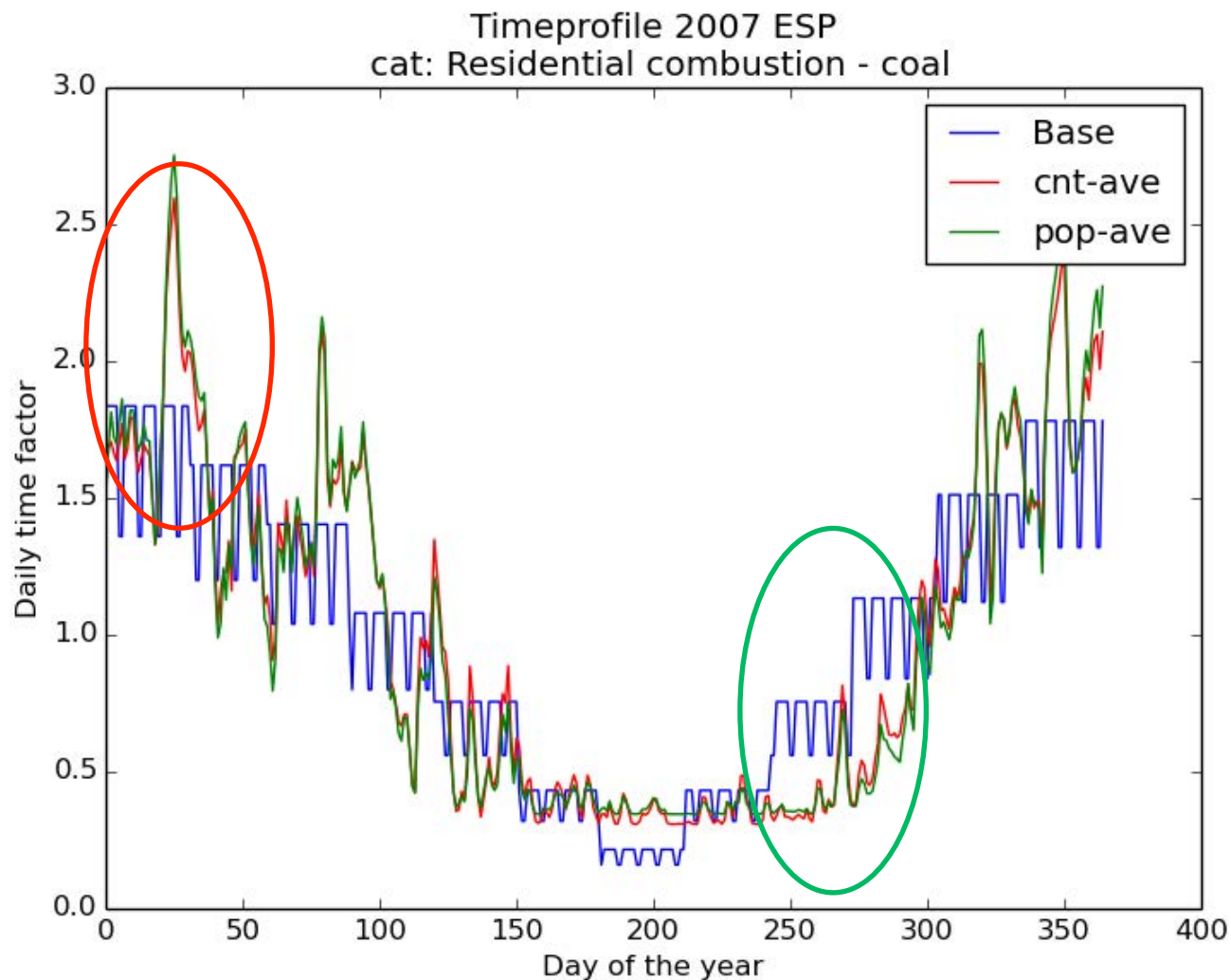
- › Heating is related to outside temperature (for space heating purposes)
- › The default profile takes seasonal climate into account
- › Changes between years can be large (e.g. March, April, October, November)
- › An alternative approach is to make the heating demand depend on actual temperature using a “degree day” approach
- › A 2<sup>nd</sup> refinement is to do this on a grid basis, taking into account gradients within the country (e.g. North vs. South)



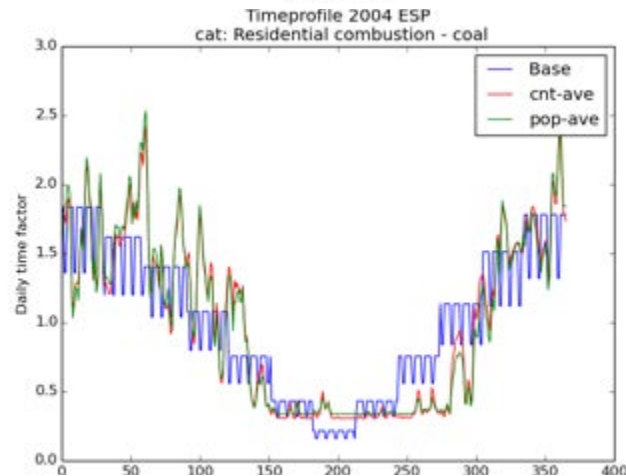
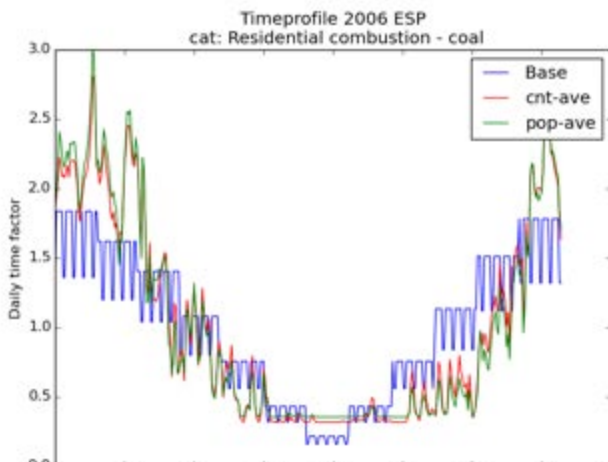
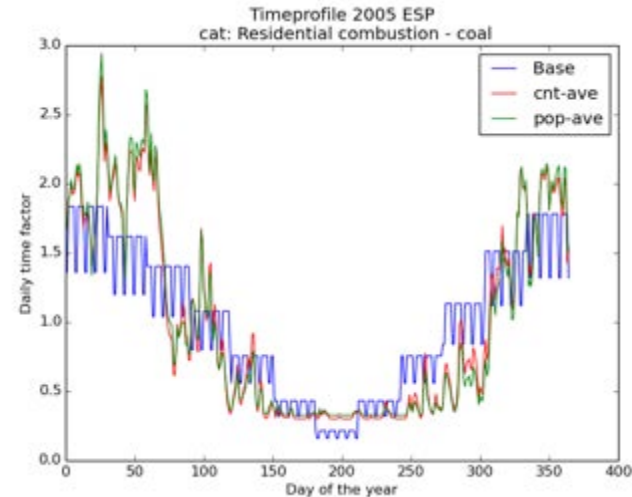
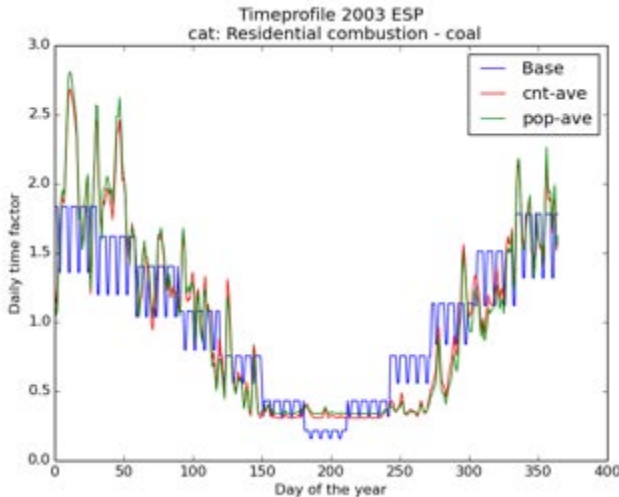
# TESTING THE APPROACH USING REAL DEMAND DATA



# APPLICATION EXAMPLE – SPAIN.



# APPLICATION EXAMPLE - SPAIN

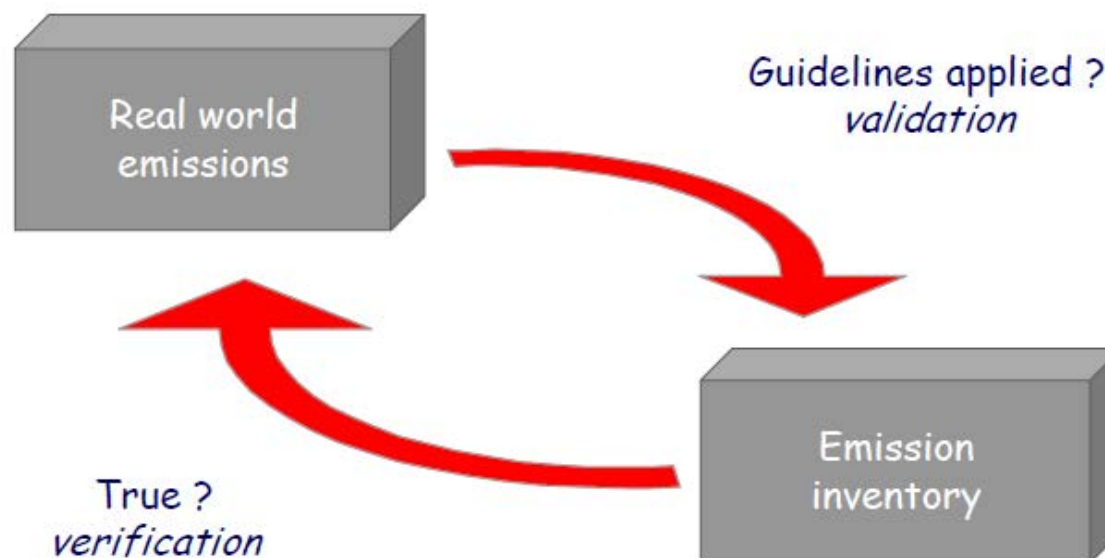


- › Degree day approach supports general pattern of default profile
- › Episodes can be dramatically different

## TO SUMMARIZE....

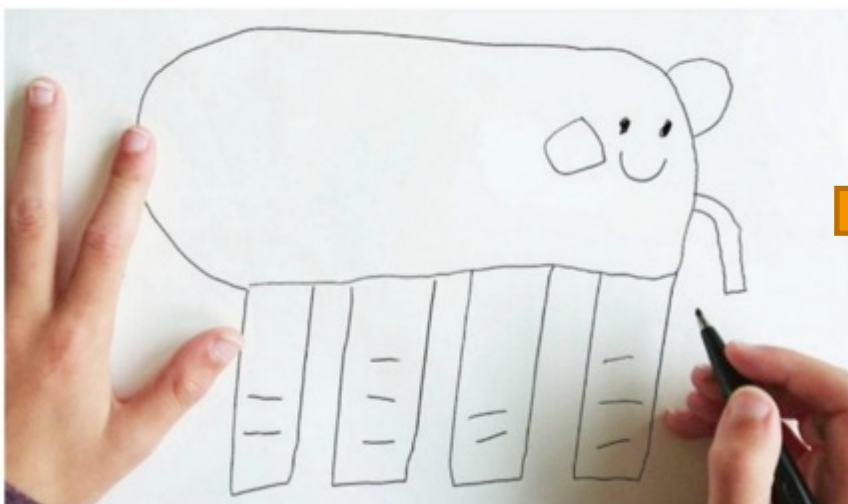
- **Good emission data are crucial**
- Significant room for improvement for every substance but not the same!
- Cooperation model– emission experts can highly beneficial
- Latest year 2011 but updates expected in 2017 (depending on acceptance of CAMS proposal)
- The TNO-MACC data have a country identifier and a SNAPcode in the grid; easy to make adjustments
- Easy to implement your own country data and use TNO-MACC to represent all other countries
- **If you do.... we are interested to learn about your national (gridded) data and you findings**

# VALIDATION AND VERIFICATION



- › **Validation** checks whether or not the guidelines have been applied, whereas **Verification** checks whether the data are true.

# A MODEL IS A MODEL....



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A close-up photograph of a llama's head and neck, looking upwards towards a thought bubble. The background is a bright blue sky with wispy white clouds. The llama has light brown and white fur.

Questions?

**To Summarize:**

**Yes!**

**Emissions can be fun science!**

**Thank you for your attention**

Contact:

[hdeniervandergon@gmail.com](mailto:hdeniervandergon@gmail.com)

[Hugo.deniervandergon@tno.nl](mailto:Hugo.deniervandergon@tno.nl)