European Emission Inventory of four indicator PAHs for 2005

- Benzo(a)pyrene,
- Benzo(b)fluoranthene,
- Benzo(k)fluoranthene
- Indeno(1,2,3-cd)pyrene

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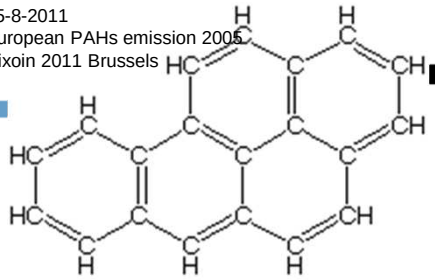
A new bottom-up inventory - Contents

- Introduction: Indicator PAHs of the POP protocol
- Anthropogenic sources – methodology & result
- Spatial Distribution
- Trends
- Natural sources
- Conclusions



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Introduction

- Polycyclic aromatic hydrocarbons (PAHs): a group of compounds composed of two or more fused aromatic rings.
- PAHs are recognized as being directly toxic to biota and possess toxic/carcinogenic properties.

- The Policy response: the 1998 UNECE POP Protocol (ECE/EB.AIR/60). Ultimate objective: to eliminate any discharges, emissions and losses of POPs (including PAHs).

- POP protocol (Annex III) specified 4 indicator PAHs for inventories:

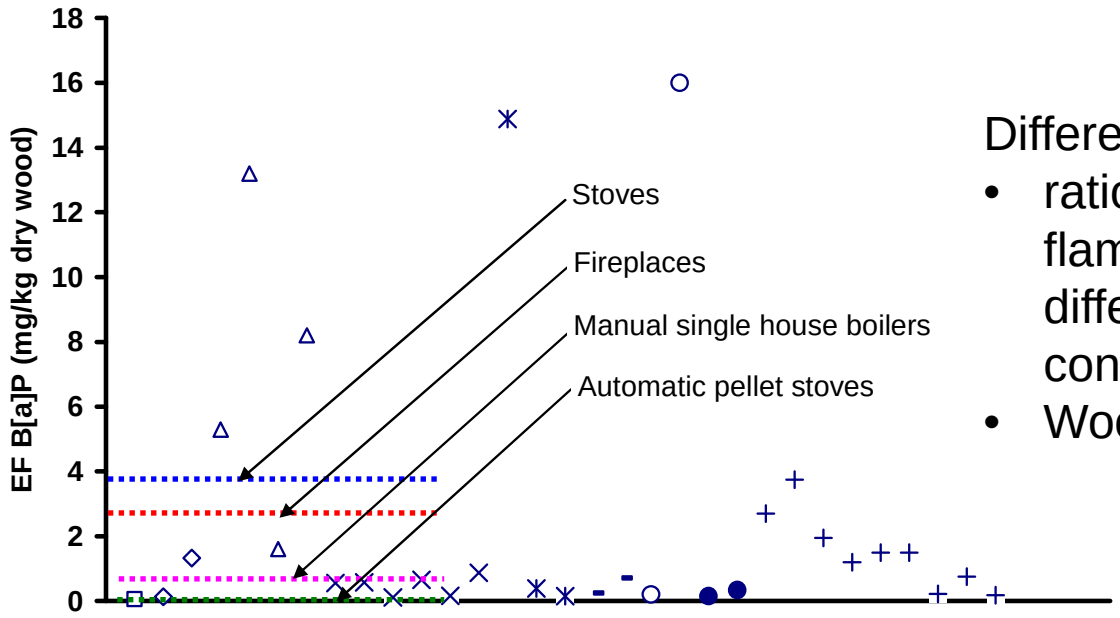
  Benzo(a)pyrene (BaP) B**enzo(b)fluoranthene** (BbF)

  Benzo(k)fluoranthene (BkF) **Indeno(1,2,3-cd)pyrene** (indeno)

Methodology = detailed review of key sources (activity x EF):

Residential combustion of wood

Recent years a lot of new studies were published (biofuels)
wide range in emission factors: For instance B[a]P in small units (< 50 kW):



Differences between studies:

- ratio between smoldering vs. flaming combustion (factor ~5 difference) - Smoldering conditions are often overlooked.
- Wood moisture contents

Activity data: Wood is non-commercial fuel; data uncertain - reassessed
-> a 20% higher fuel wood use in Europe was estimated (mostly in Eastern European countries). This causes proportional increase of PAHs emissions.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Methodology Residential combustion of Coal

PAH emission due to evaporation and incomplete combustion of volatile components in coal
Many new studies were published (China, Poland) : EFs vary more than 5 orders of magnitude (e.g. **B[a]P: 0.0002 – 90 mg/kg coal** for small units (<50 kW))

PAH emission depends on

Coal type: in order of increasing emission factors:

1. Anthracite
2. Smokeless fuels/coke,
3. Sub-bituminous coal;
4. Bituminous coal (most used in Europe)

Example conventional stoves
B[a]P EFs for anthracite are ~ 1.5% of EFs for bituminous and sub-bituminous coal

Appliance type / combustion conditions: EFS decrease with thermal capacities
e.g. B[a]P emission factors for medium boilers (5 to 50 MW) are only 2.5% of that of stoves and single house boilers (< 50 MW)

Conclusion: Correct classification of coal type and appliance type is crucial


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Coke ovens - I

- EF Coke ovens: 20 – 5000 mg B[a]P/tonne produced – split in
- 4 emission factor classes based on observed PAH emissions during:
  - Class 1 5000 the 1970s/1980s
  - Class 2 750 1980s/1990
  - Class 3 100 1990s/2000s
  - Class 4 20 2000s/Best available technology
-
- Appropriate emission factor class for each point source is based on
  - Stack measurement data
  - Real world information on technological standards
  - Socio-economic parameters
- Pictures of each plant on the Internet provide supportive information for high / low emitter, on a plant by plant basis.

Coke ovens II - Crucial to classify each facility –

visual judgement provides important supportive information

Modern plant in Taranto, Italy.
The location of the coke ovens is indicated by two red circles (No visible smoke is seen).

Class 4

Magnitogorsk iron and steel complex, Russia
Coke ovens (green arrow) & sinter factory on the far right. -> Identified as high emitter

Class 1

Söderberg aluminium Smelters

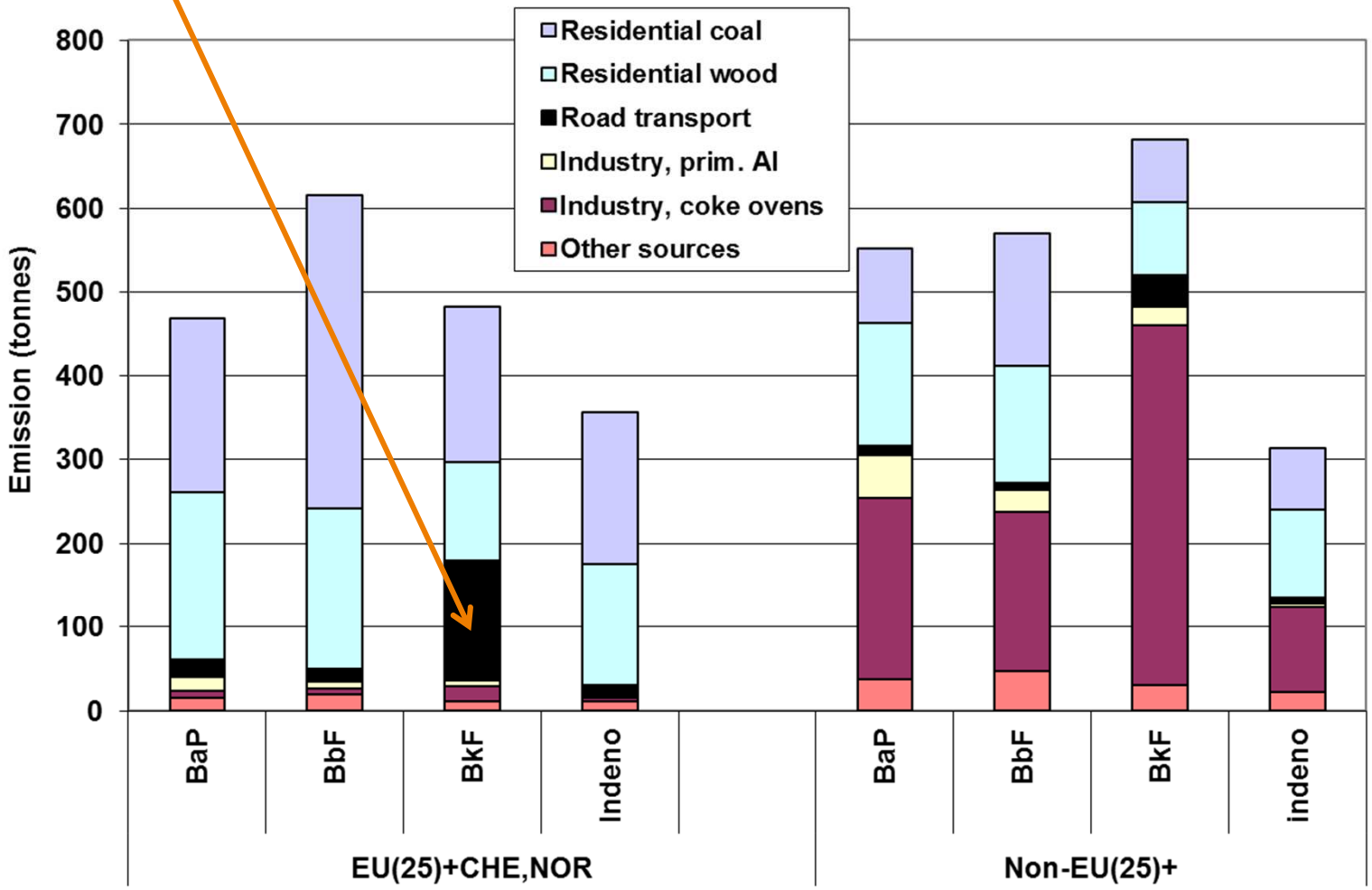
Review: Range in EFs =
10 – 120 mg B[a]P/tonne produced

The Sunndalsøra plant, Norway
modern (partly) Söderberg-based aluminium smelter: Class 4 low emitter

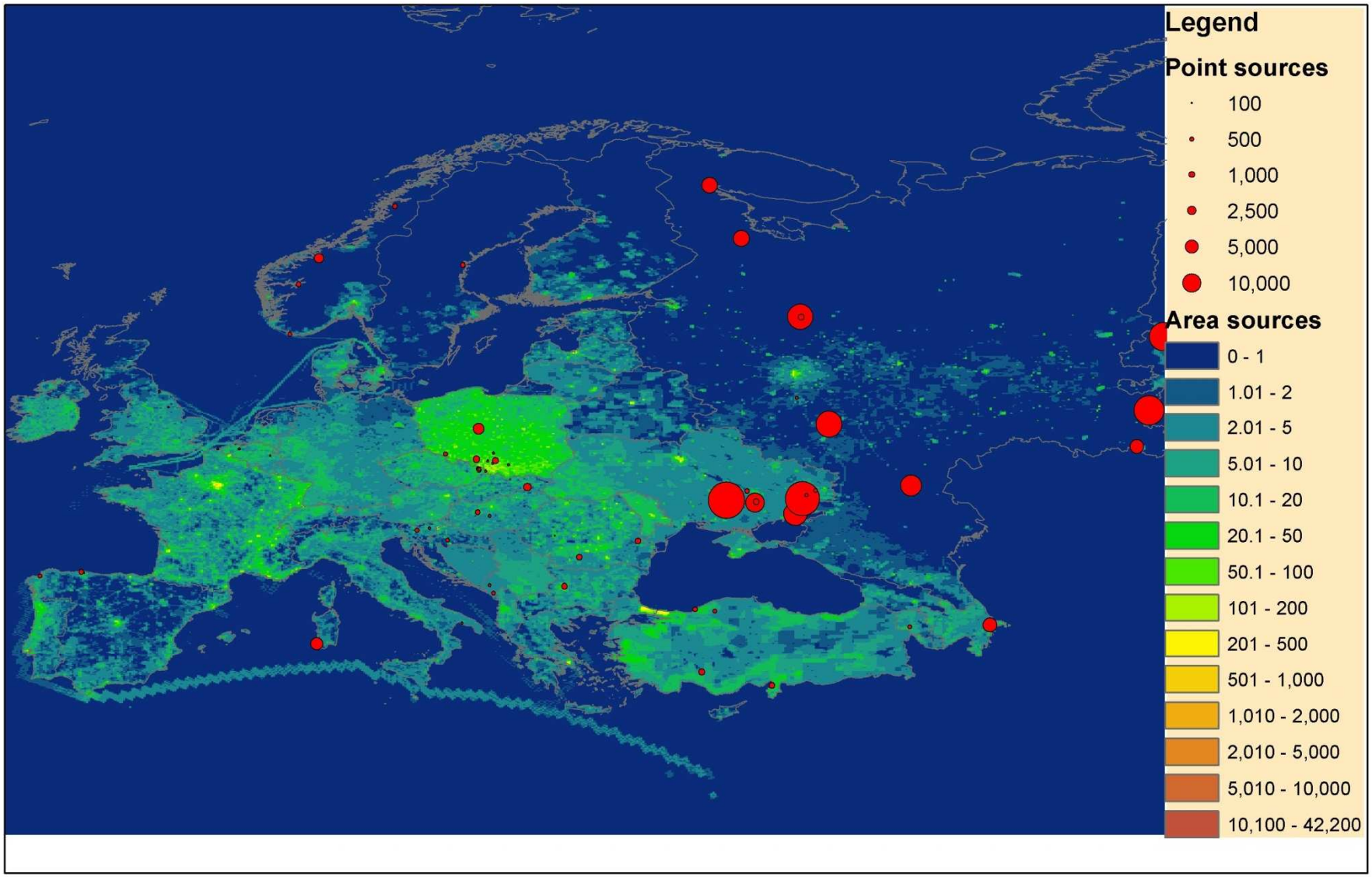
Krasnoturinsk, Russia
Söderberg-based plant, high emitter Class 1

PAHs emissions in 2005 excl. international shipping

Remarkable but based on very limited data “uncertain” ***or contributions***

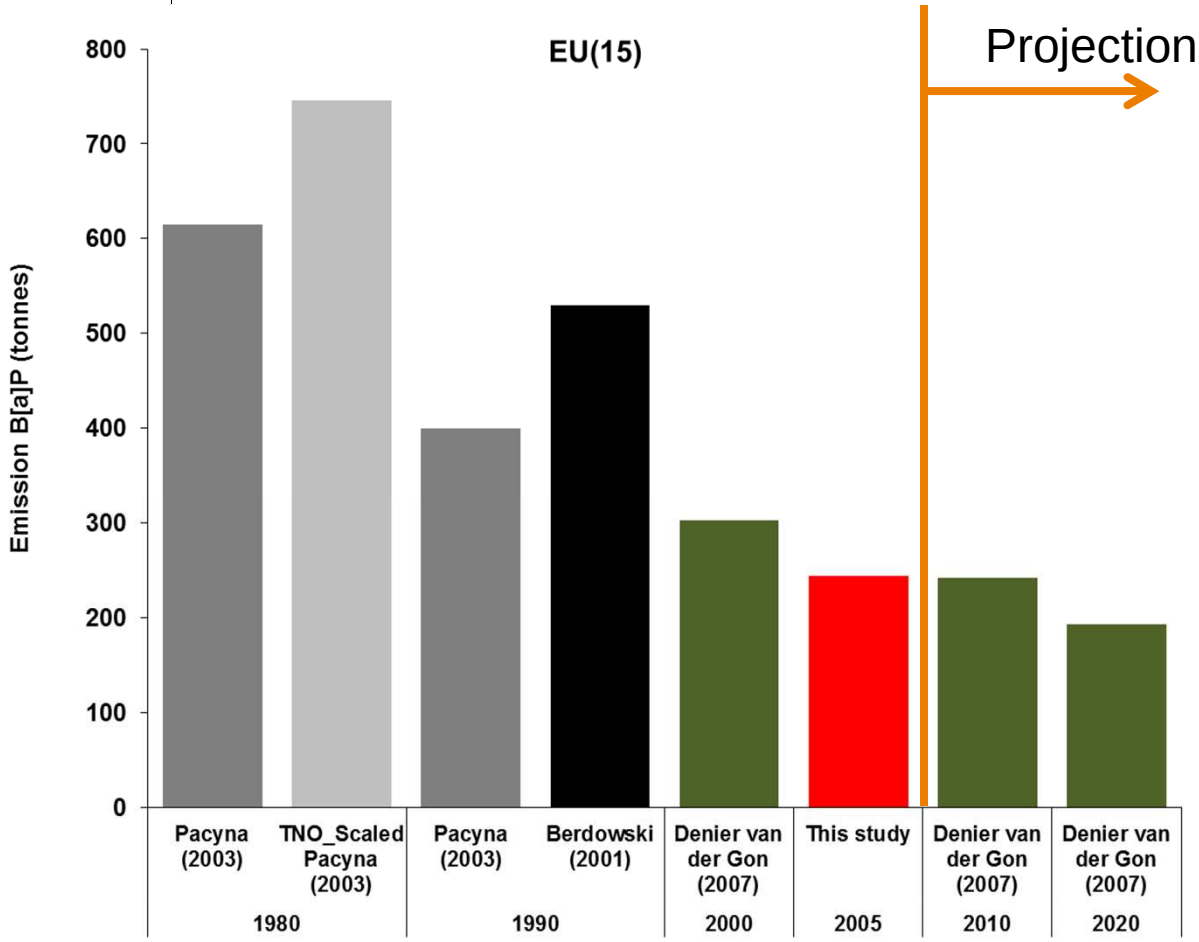


Spatial distribution of BaP emission



B[a]P emission trend in the EU 15 for 1980-2020

(excluding natural emissions)



Clear decline over time, mostly due to reduction of industrial emissions

Discrepancy 1980 and 1990 estimates due to emission factors for wood combustion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How important are natural (fire) emissions? *Methodology:*

- Available satellite observations are merged in a Global Fire Assimilation System (GFAS) to obtain fire emission estimates with global coverage at high temporal resolution (Kaiser et al., 2009; MACC project).
- From GFAS various biomass burning (BB) properties can be derived including an estimate of kg dry matter being burned.
- Review and collect PAHs emission factors / kg dry matter burned and calculate (daily) emission of PAHs
- Use a mask to derive the emissions per country and for the same domain as the anthropogenic emissions

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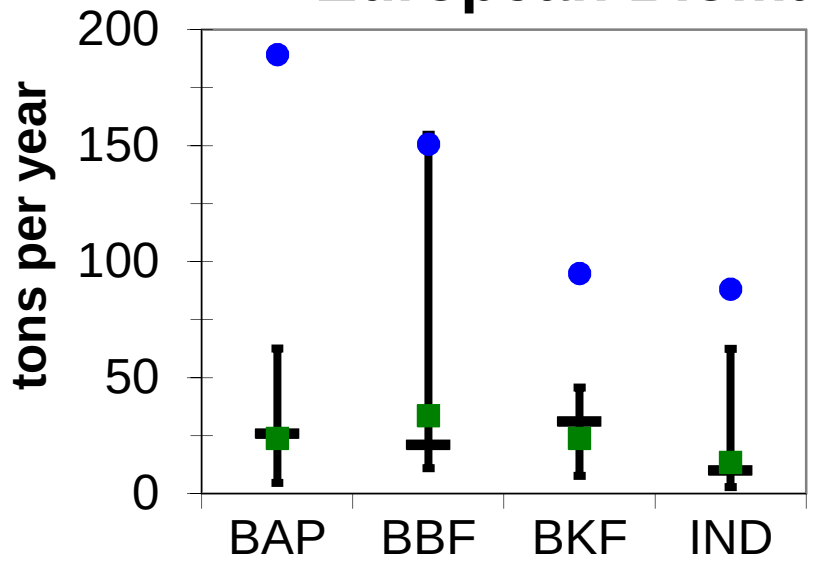
PAH emission in 2005 due to European Biomass burning (forest fires)

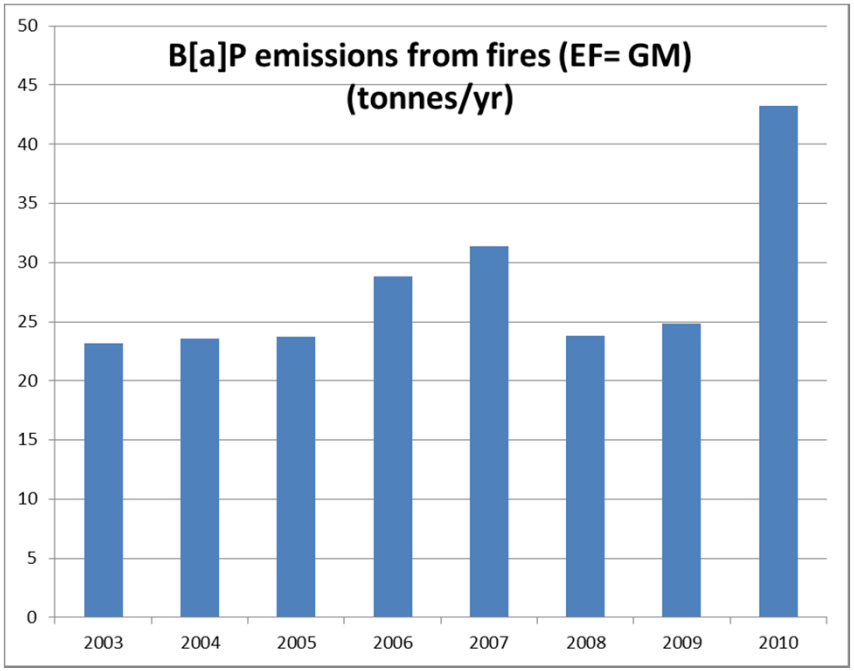


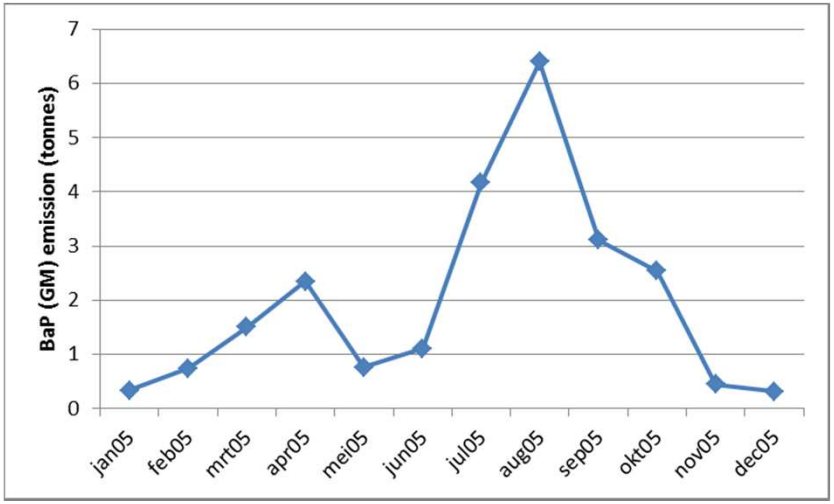
EF data contain important outliers – Result is very different means: GM or median is most robust

Estimate based on Geometric mean
BaP ~ 24 t/yr;
BBF ~ 33 t/yr ;
BKF ~ 24 t/yr
IND ~ 14 t/yr

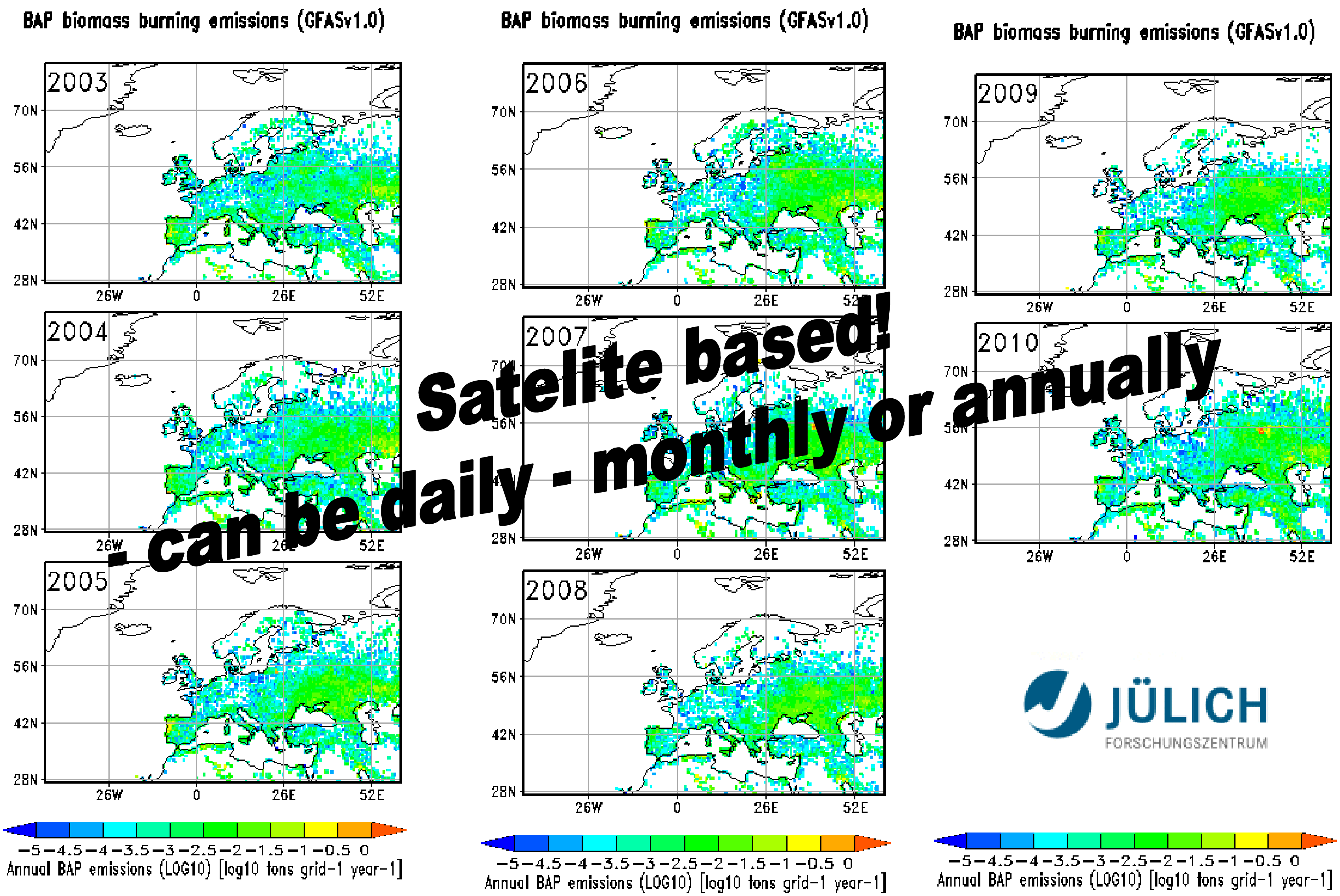
Small contribution compared to anthropogenic sources:
PAHs mainly formed between 700 - 900ºC, Biomass fuels burn often below 700℃ (savannah fires) or above 900°C (for extratropical forest fires), consequently not in the maximum of PAH production – explains limited importance

European PAH emission estimates from fires by year and monthly variation





Annual variation is limited but extremes occur (2010)
Emission pattern within the year is very important



Satelite based!

- can be daily - monthly or annually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Strengths of this new inventory for high molecular PAH

- Recent base year (2005)
- Thorough review of emission factors for most important sources :
  - *Residential wood combustion (including new activity data)*
  - *Residential coal combustion*
  - *Industry: Primary iron and steel (incl coke ovens) & Primary aluminium smelters (incl. method to identify high emitters)*
- Transport (over time from major to minor source)
- Compatible emissions from forest fires now available
- State-of-the-art spatial distribution including exact location of point sources and update point source list: many coke ovens and aluminium smelters have recently been closed in the EU


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Conclusions

- Total European emissions of BaP, BbF, BkF and Indeno in 2005 were estimated as 1020, 1186, 1164 and 669 tonnes/yr, respectively.
- The emissions are strongly dominated by residential combustion (EU) and industrial emissions (Non-EU countries)
- Forest fires / BB is a modest source of PAHs if (~2-4% of anthropogenic emissions) but sensitive to assumptions: Arithmetic mean (15-20% additional emissions).
- Transport is a minor source; although for Int. Shipping a factor of 100 between EFs – new TRANSPHORM data will be useful
- Importance of residential combustion is not expected to change
- Potential for reduction from industrial facilities in non-EU countries
- These 4 indicator PAHs are “heavy” molecules making fugitive emissions a minor source – this does not apply to all PAHs!!!


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THANK YOU FOR YOUR ATTENTION

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TRANSPHORM transport pollution health

macc Monitoring atmospheric composition & climate

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