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EMISSION, FATE AND RESPIRATION EXPOSURE RISK OF POLYCYCLIC AROMATIC HYDROCARBONS IN CHINA

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1. INTRODUCTION
2. EMISSION INVENTORY
3. OUTFLOW FROM CHINA
4. INHALATION EXPOSURE RISK

CONTAMINATION OF PAHs IN CHINA

- **16 parent PAHs on US EPA priority pollutant list**
  Nap, Acy, Ace, Flo, Phe, Ant, Fla, Pyr, BaA, Chr, BbF, BkF, BaP, IcdP, DahA, and BghiP
- **Incomplete combustion**
  industry, motor vehicles, residential solid fuel, wildfire etc.
- **important in China and other developing countries**
  Regionally based assessment of PTSs – UNEP Chemicals, 2003

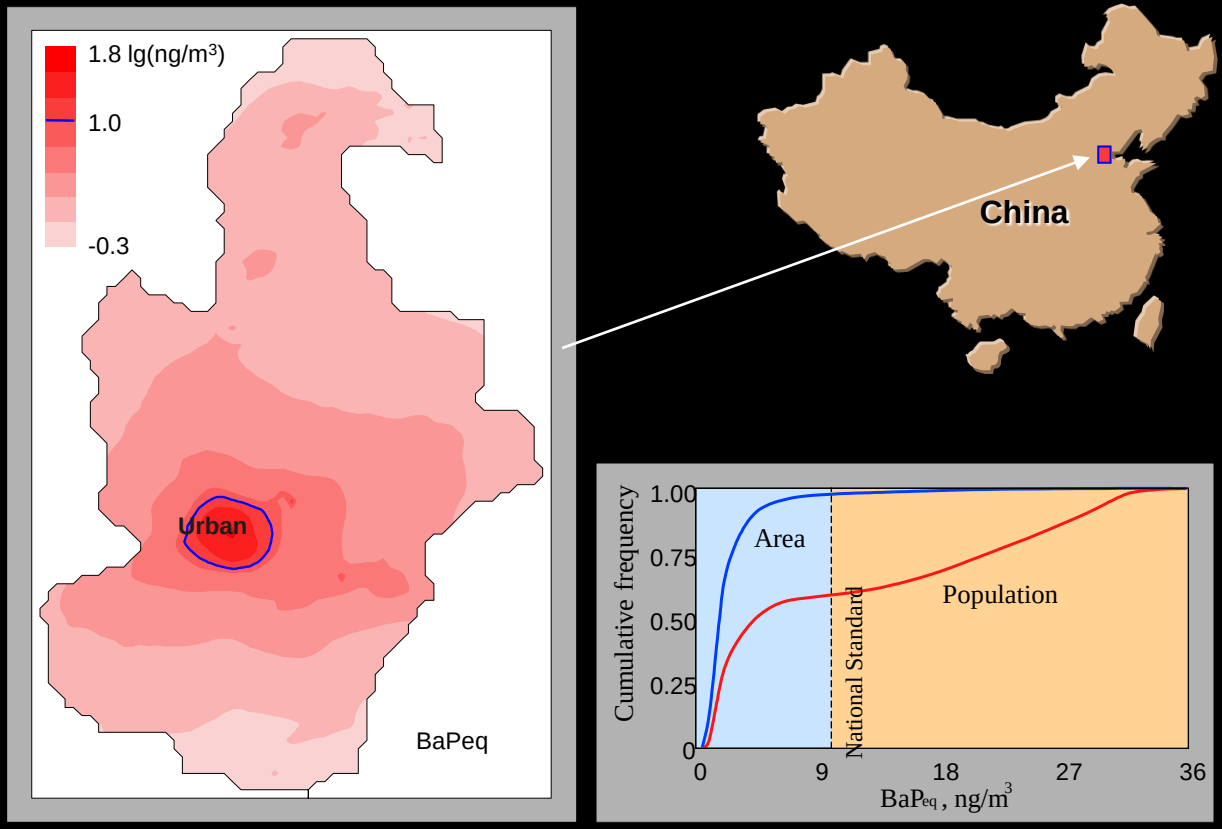



LOCAL CONTAMINATION

- **severe contamination in Tianjin**

  based on a short-term air transport modeling

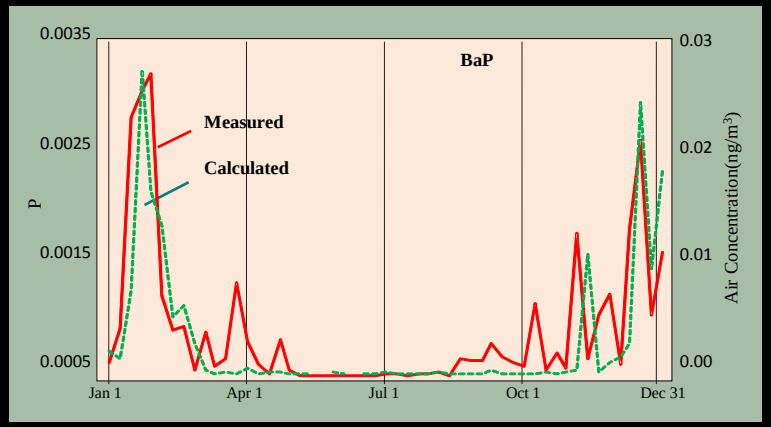
  over 40% population live in urban area with annual mean BaP_{eq} above 10 ng/m^3

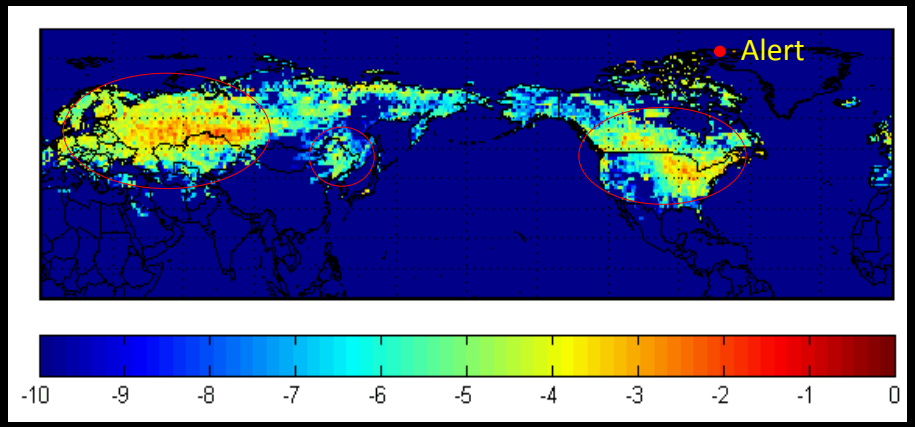


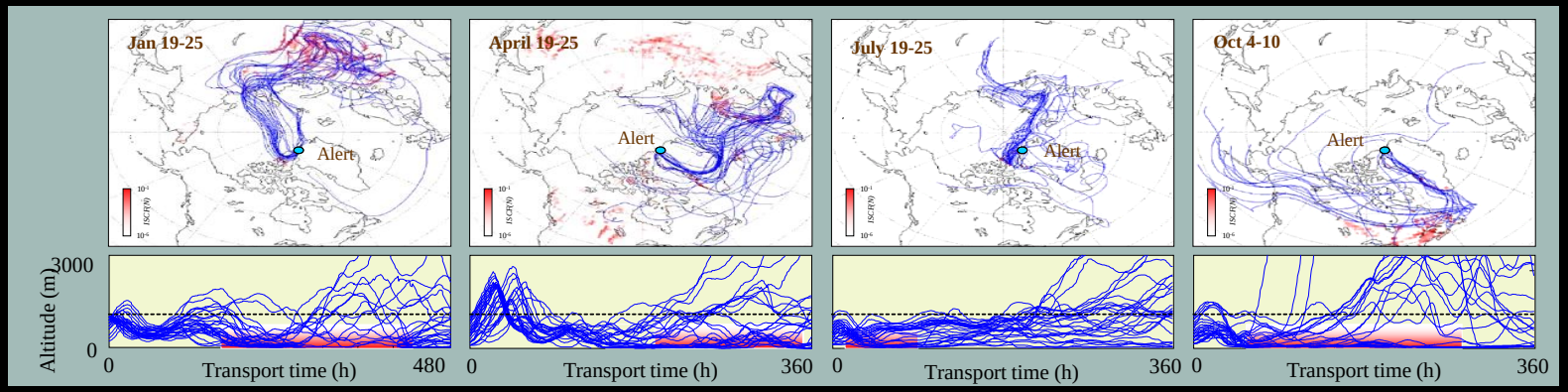
Tao et al., ES&T, 2006

LONG-RANGE TRASNPORT POTENTIAL

- **sources and pathways of PAHs observed at Alert, the Arctic**
  based on emission inventory, trajectory calculation, and a probabilistic model (ISCF) from almost everywhere in North Hemisphere







Wang et al., ES&T, 2010

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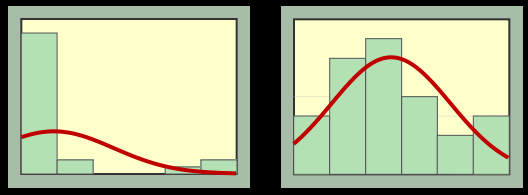
DEVELOPMENT OF PAH EMISSION INVENTORY

- **16 parent PAHs on USEPA priority pollutant list**
  from naphthalene to benzo(g,h,i)perylene

- **major emission activities**
  firewood, straw, domestic coal, industrial coal, coke ovens, motor vehicles, natural gas, consumer product usage, aluminum production, iron and steel production, petroleum refinery, gasoline distribution etc.

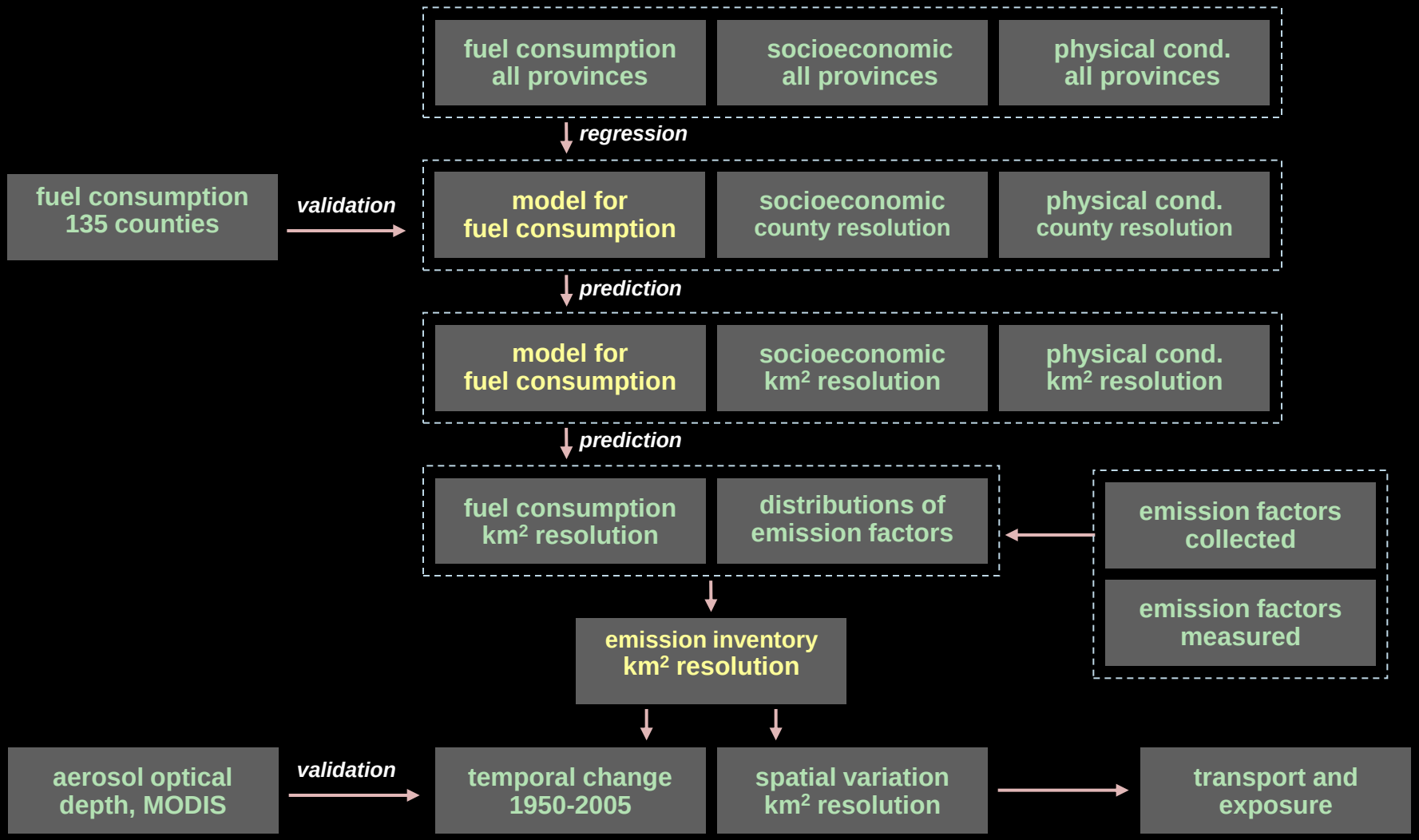
- **emission factors**
  a thorough literature review, medians and distributions used



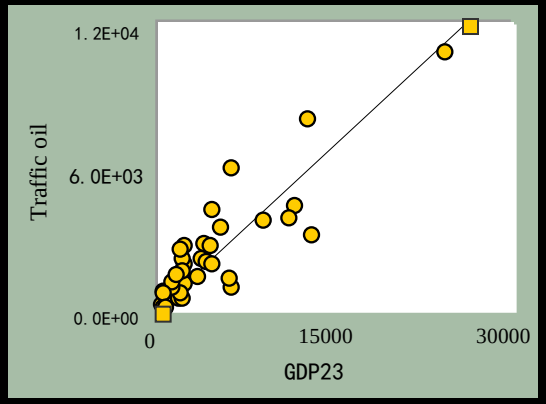
banzo[a]pyrene, traffic oil, n=33

- **emission activities**
  available data were those for all provinces and only 135 counties in China

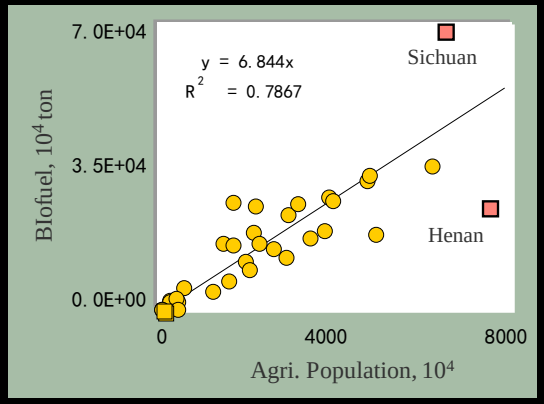
ENERGY CONSUMPTION PREDICTION



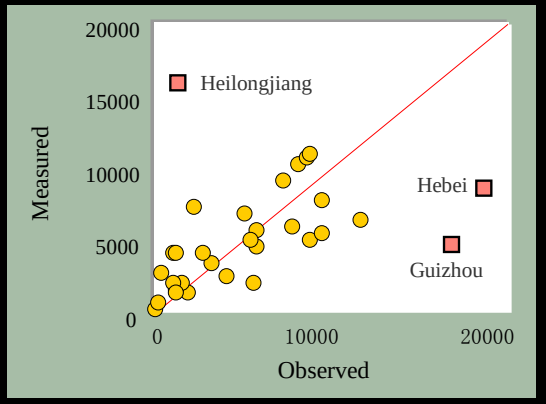
REGRESSION MODELS



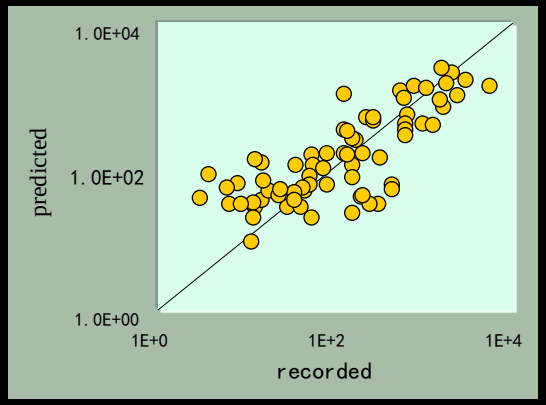
traffic oil



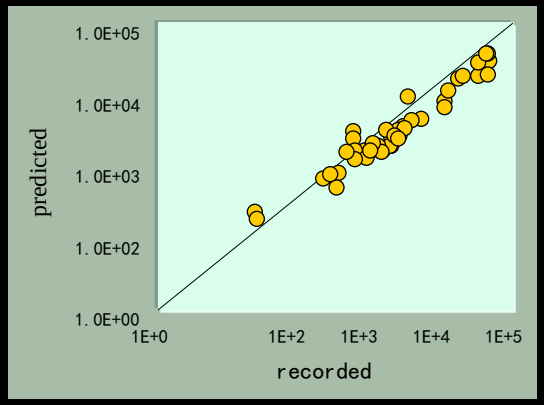
biofule



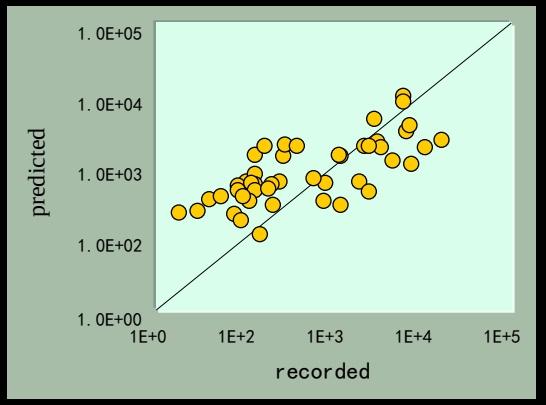
Domestic coal vs. population/T



industrial coal

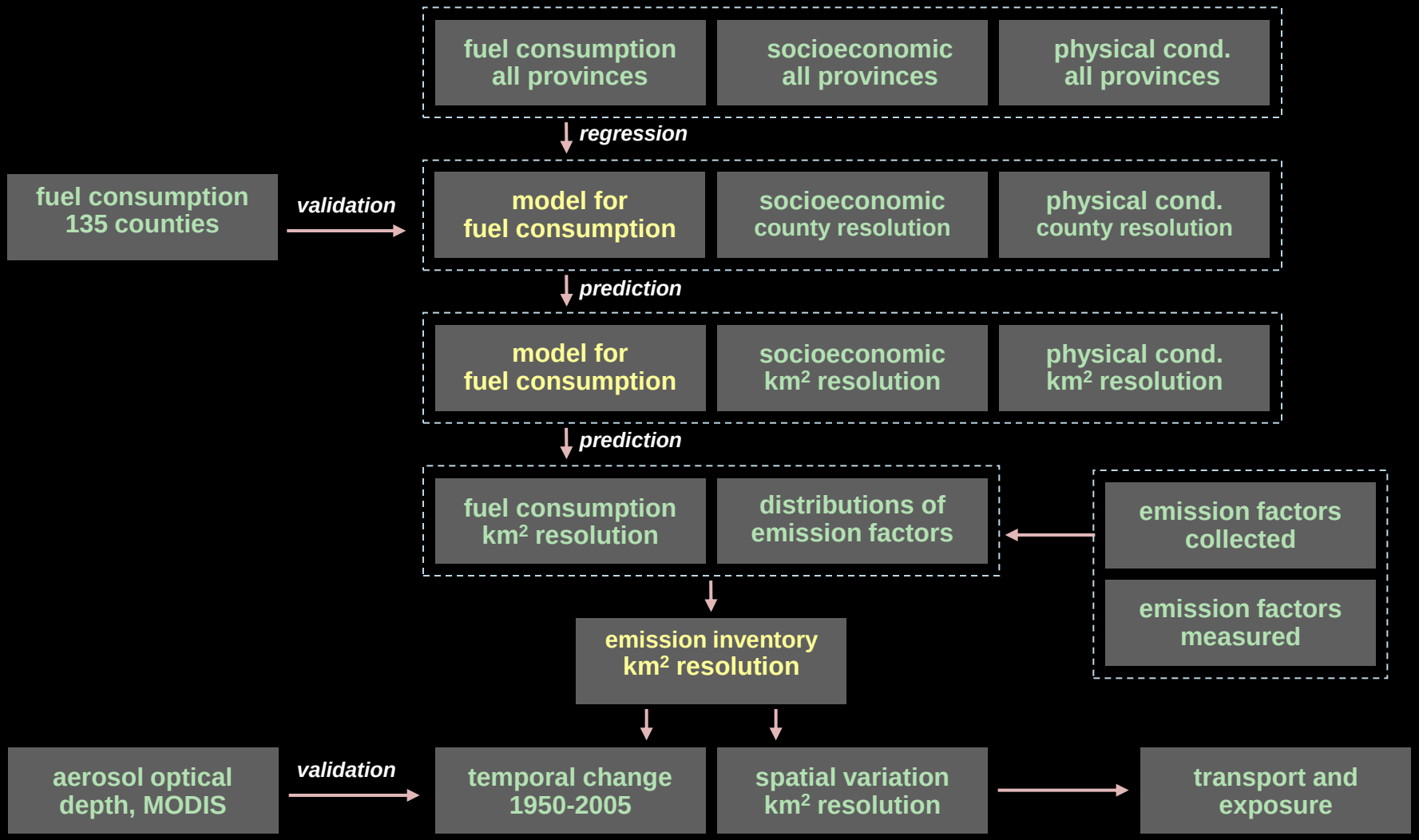


industrial petroleum



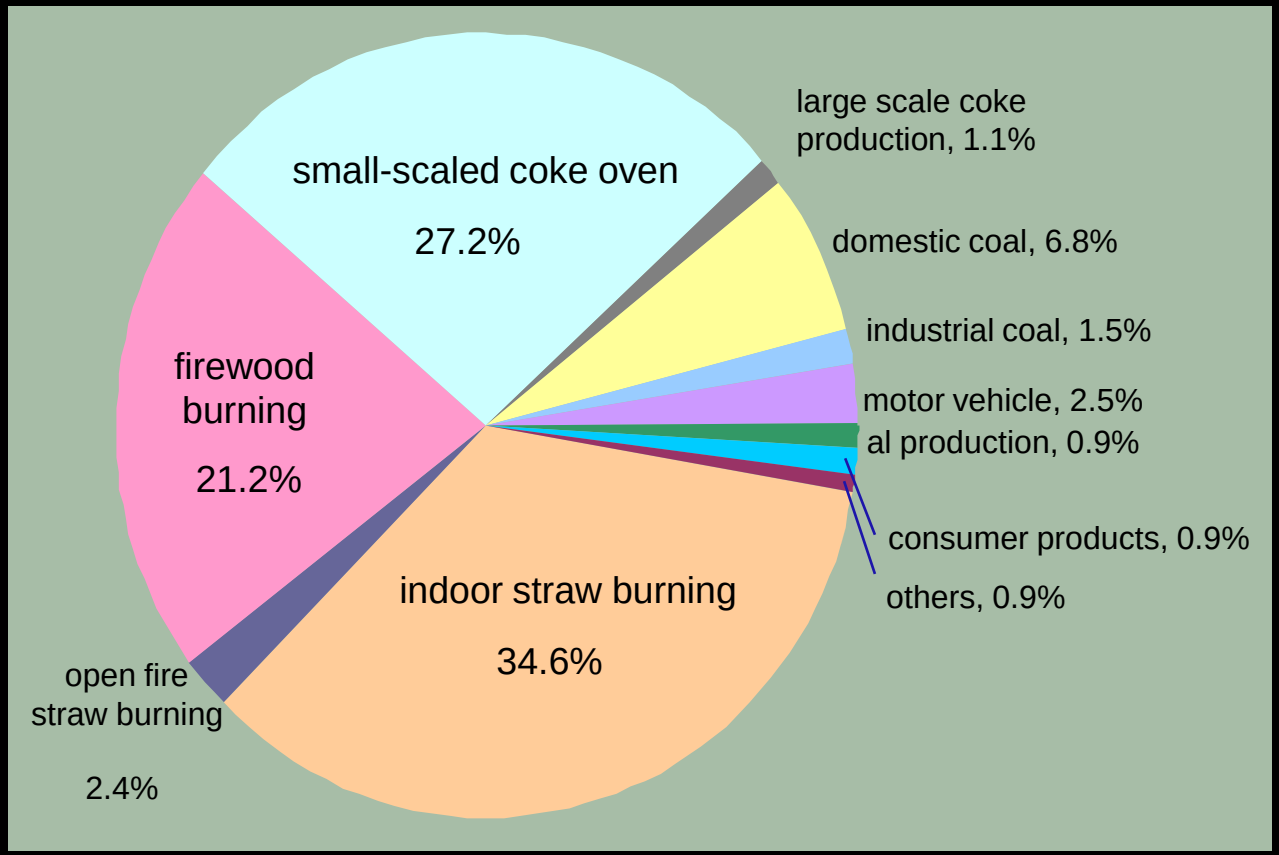
biomass burning

ENERGY CONSUMPTION PREDICTION



TOTAL EMISSION AND SOURCE PROFILE

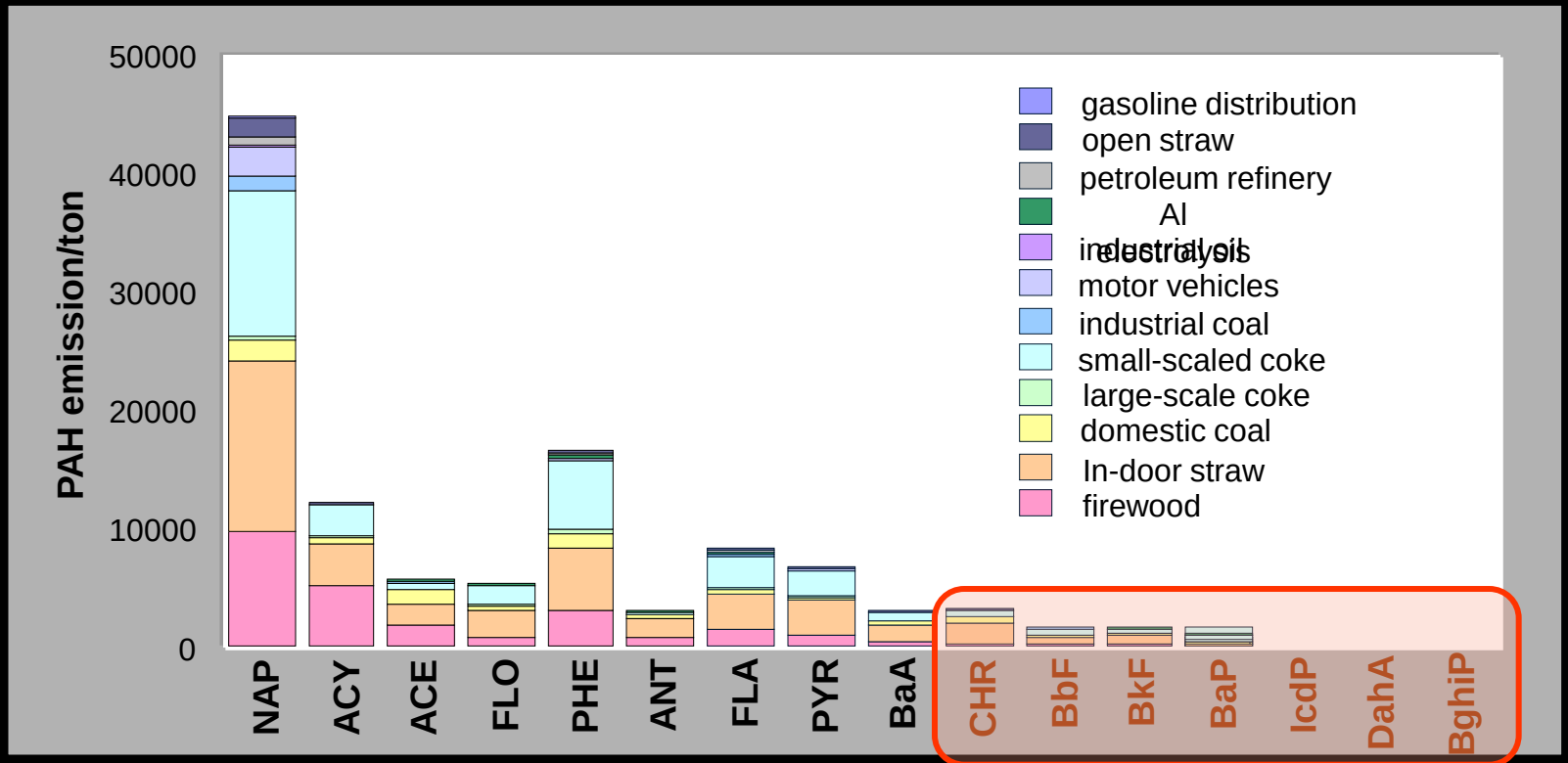
- annual emission was 116,000 ton in 2003
- primarily from small-scaled coke ovens and indoor biomass



COMPOSITION PROFILE OF THE EMISSION

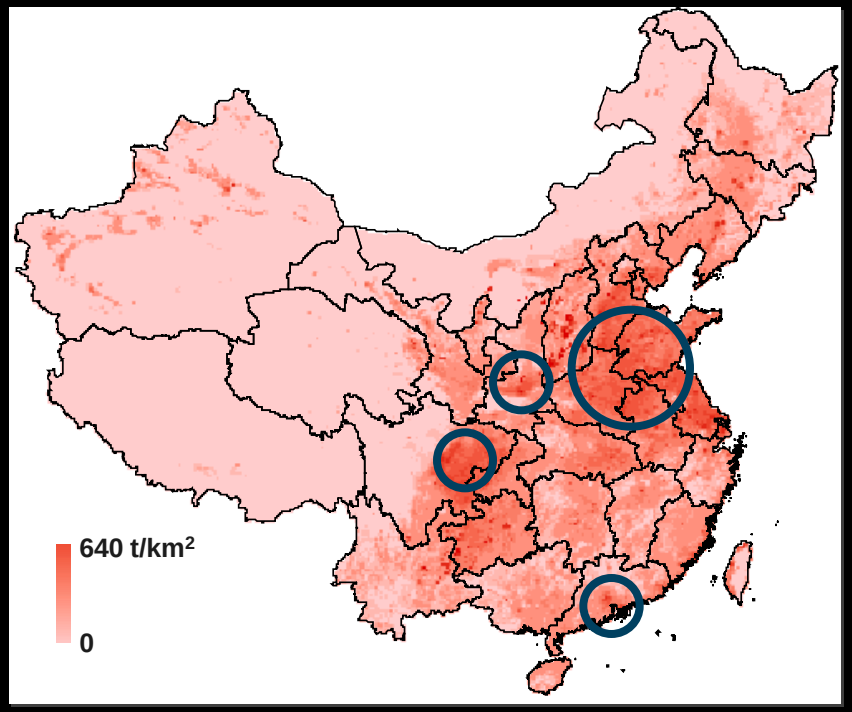
- **10% was carcinogenic compounds**

  compared to 2.7% in the Great Lake area

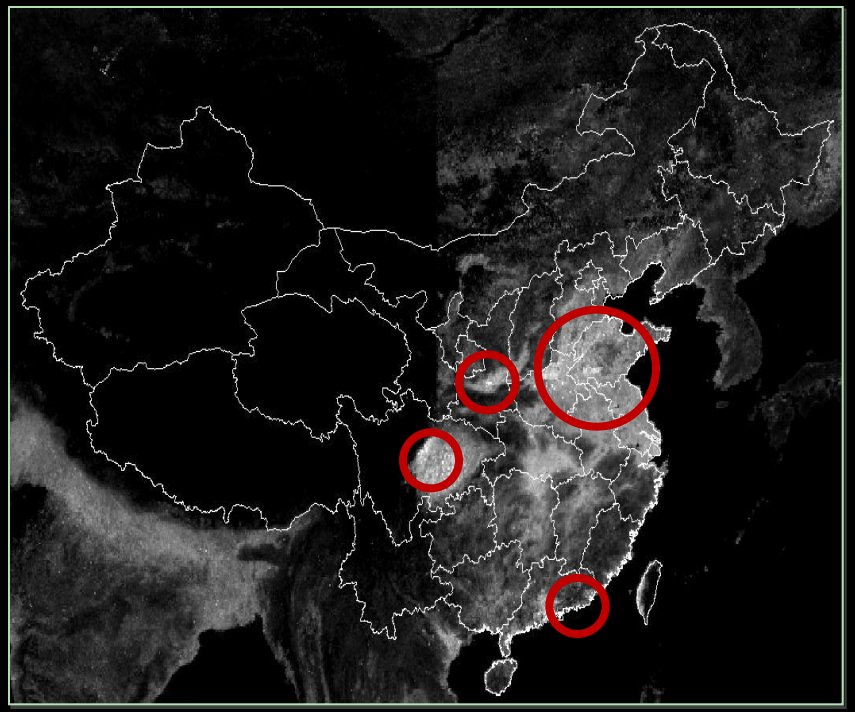


GEOGRAPHICAL DISTRIBUTION

- similar to aerosol optical depth in spatial pattern



PAH emission density with 1x1 km² resolution, 2003



aerosol optical depth (MODIS), 2003
Li et al., J Int. Remote Sensing, 2010

SUMMARY

- **total PAH emission in China: 116,000 ton in 2003**
- **major sources: indoor biomass burning, small-scaled coke ovens**
- **10% of total emission in China were carcinogenic**

Environmental Science & Technology 2006, 40, 702-708
Environmental Science & Technology 2007, 41, 683-687
Atmospheric Environment 2008, 42, 6828-6835
Science of Total Environment 2008, 1140, 1-21
Environmental Pollution 2008, 156, 657-663
Anna New York Acad Sci, 2008, 1140, 218-227
Atmospheric Environment 2009, 43, 812-819

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POTENTIAL RECEPTOR INFLUENCE FUNCTION

- **forward air mass trajectory calculation based**

  HYSPLIT

- **Potential Receptor Influence Function – PRIF**

  a probability density function

  idea borrowed from Potential Source Contribution Function (PSCF) ***Hafner et al., 2003***

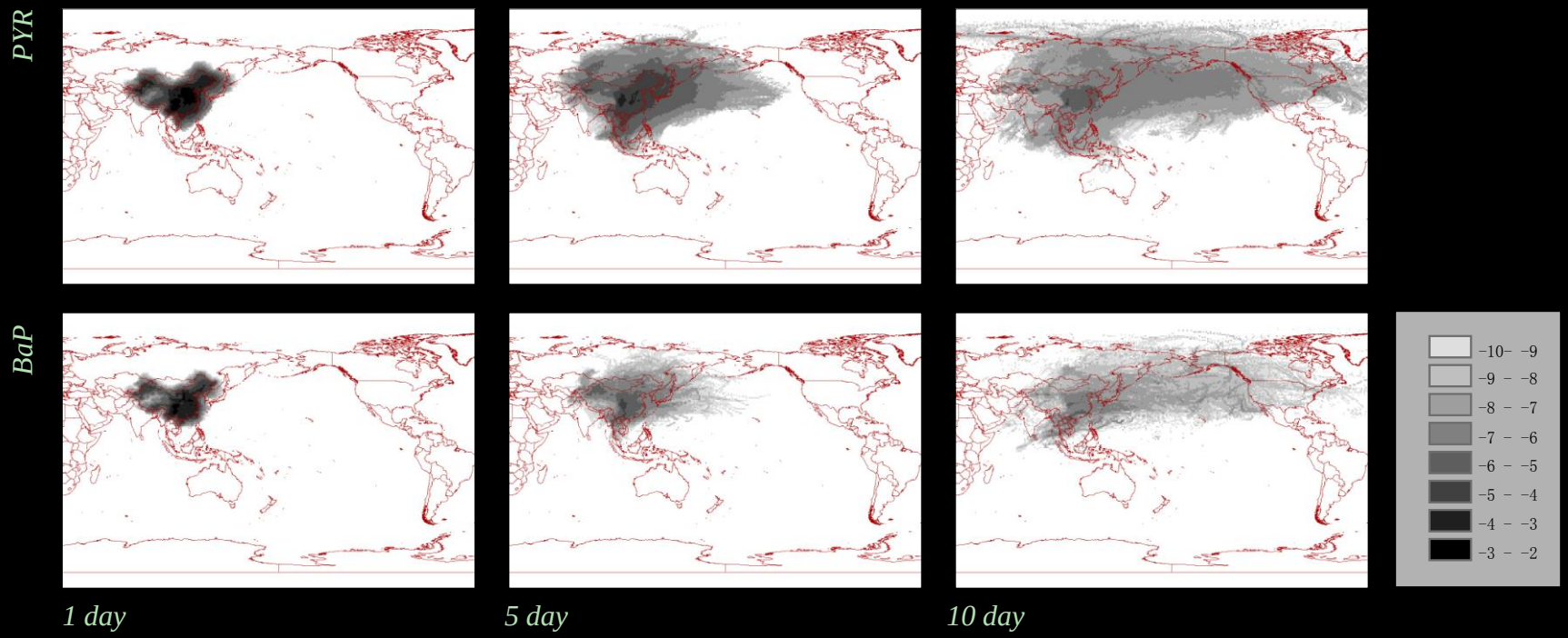
  the probability of PAHs arriving at a receptor site during a given emission duration and a known period of transport time

  processes (emission, partition, degradation, deposition) included

- **outflow quantification**

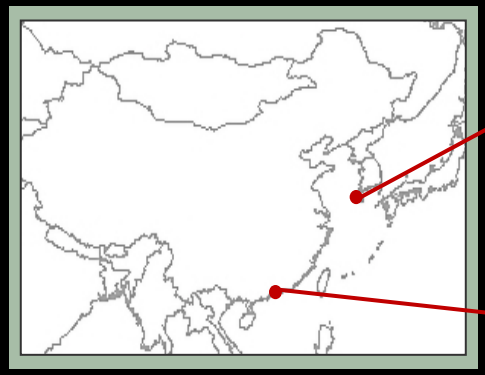
ANNUAL PAH OUTFLOW FROM CHINA

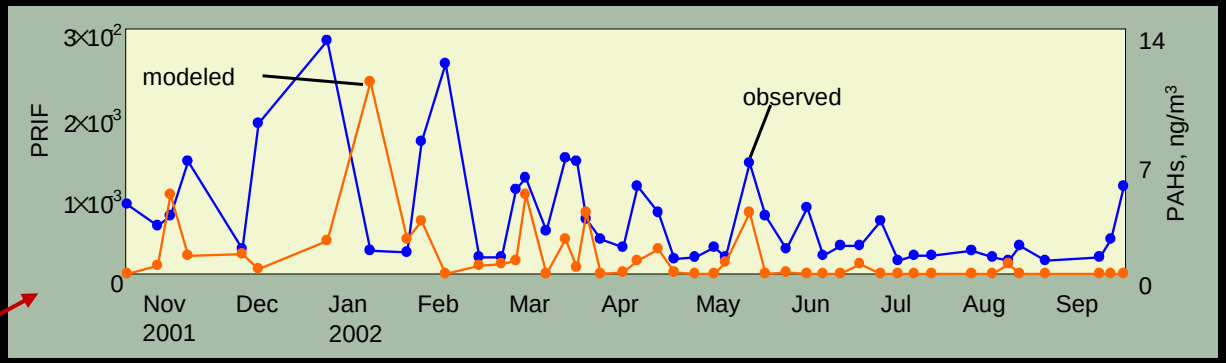
- **naphthalene, phenanthrene, pyrene, benzo(a)pyrene**
  3511 ton (8.0%), 1119 ton (7.2%), 663 ton (10.5%), 47.8 ton (3.3%)



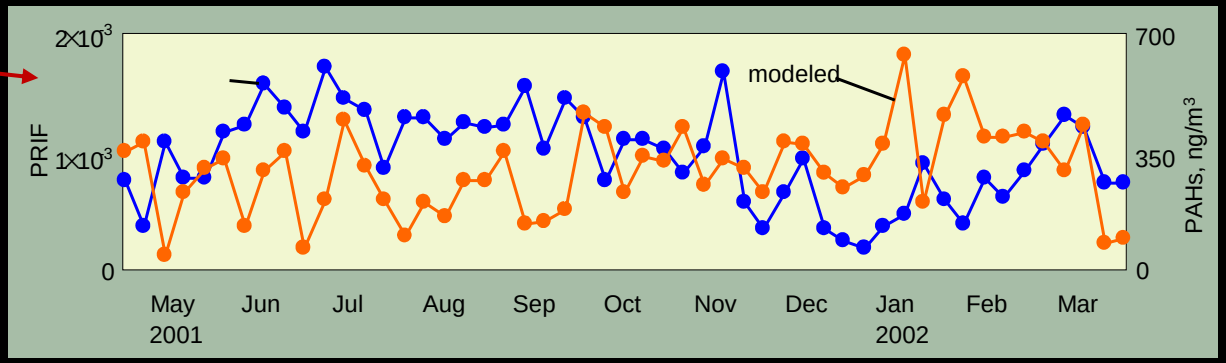
MODEL VALIDATION

- time series concentration data from two sites





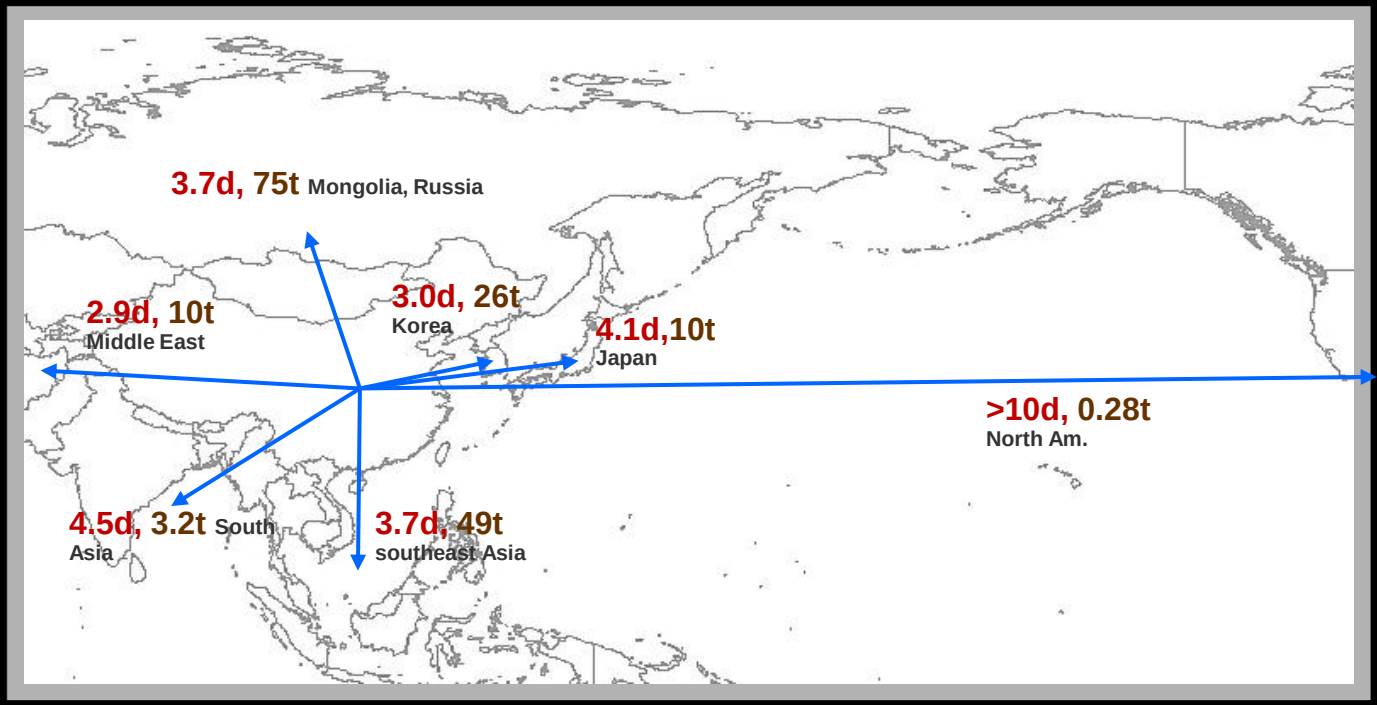
Gosan (33 °N, 126 °E), Lee et al., J. Geophys. Res. Atmos. 2006



Guangzhou (23 °N, 113 °E), Le et al., Sci Total Environ. 2006

OUTFLOW QUANTIFICATION

- total emission and PRIF calculation based
- net input of pyrene to various countries
- mean travel time and annual input



INTERANNUAL FLUCTUATION

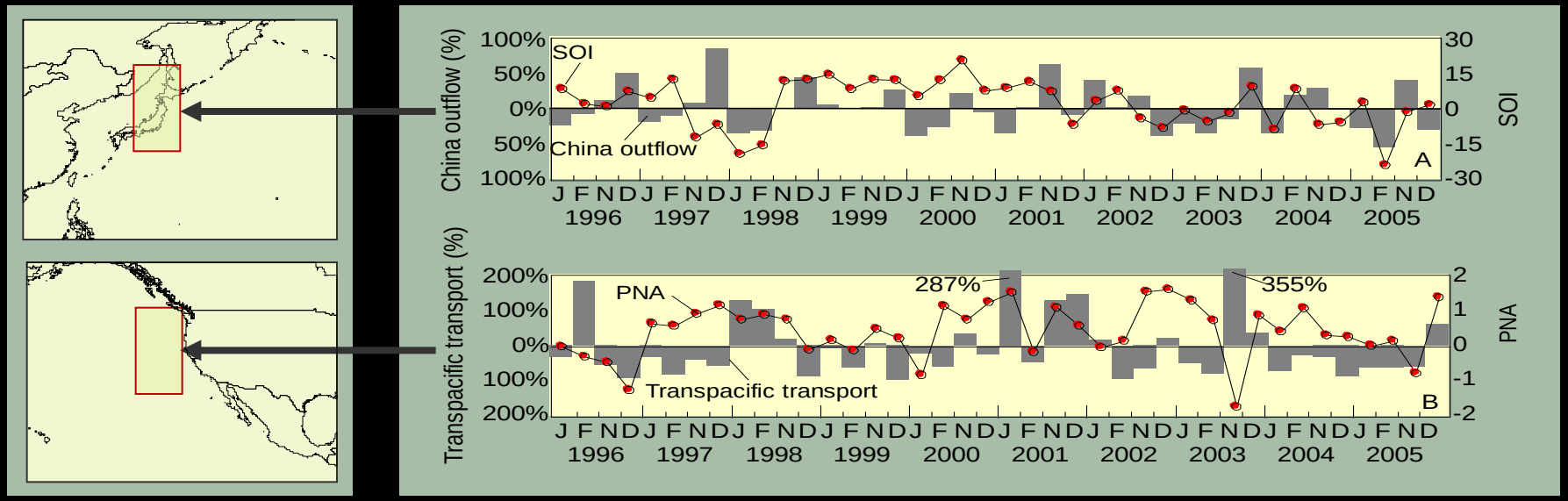
- **standardized monthly anomalies at two receptor areas**

  35°N-45°N – 135°E-145°E (outflow); 135°W-145°W (LRT), PRIF of pyrene

- **correlations**

  outflow – Southern Oscillation Index

  LRT – Pacific North America Index



SUMMARY

- **PRIF can be used to characterize PAH outflow**
- **outflow accounted for 3~11% of the total emission**

Environmental Science & Technology 2007, 41, 8370-8375

Atmospheric Environment. 2007, 41, 8370-8379

Environmental Science & Technology 2008, 41, 5196-5201

Environmental Science & Technology 2010, 44, 1017-1022

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ATMOSPHERIC TRANSPORT MODELING

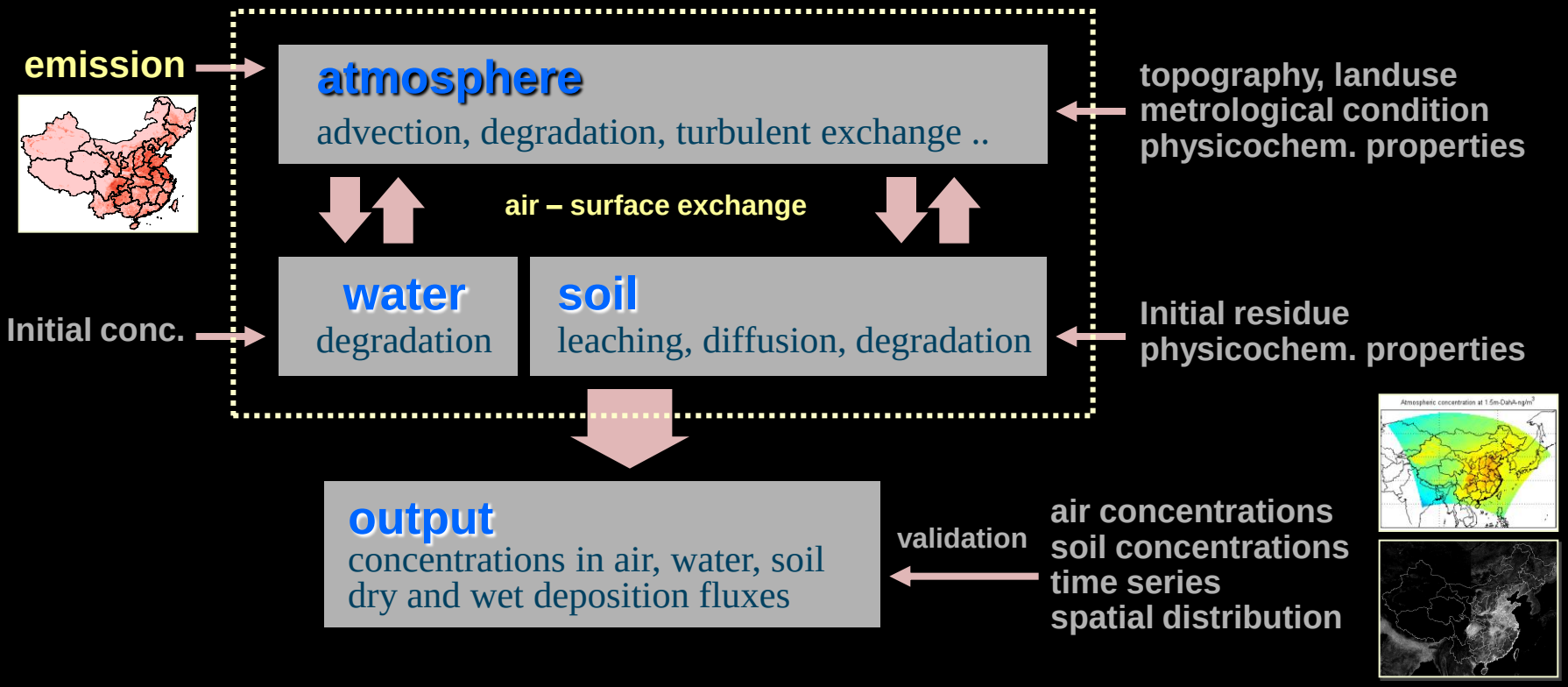
- **atmospheric transport model** *Ma, 2003*

  modified by including processes of air/surface exchange, partition, and degradation

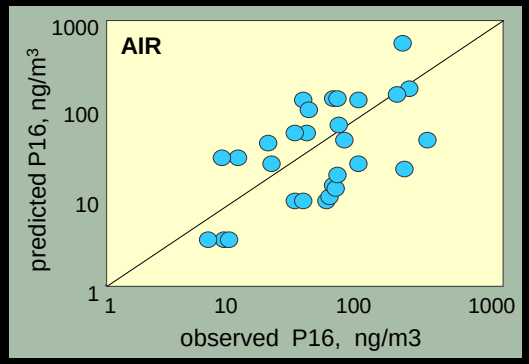
  atmospheric transport module (24km x 24km x 12 layer)

  soil-air exchange and soil module (fugacity based)

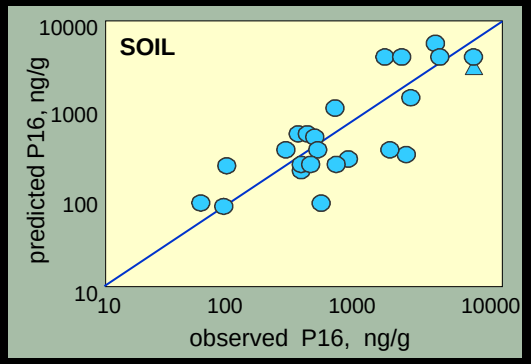
  water-air exchange and water module (stagnant film model)



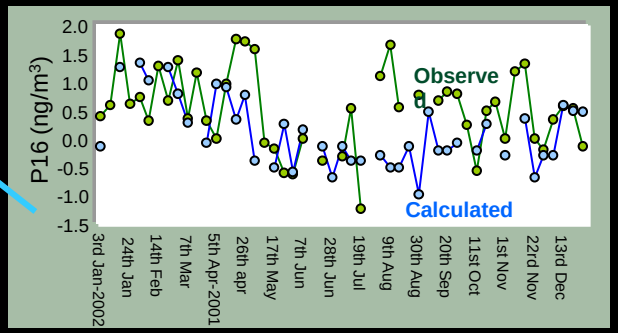
VALIDATION



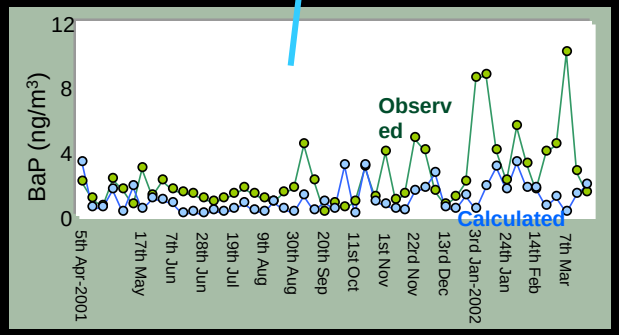
Ambient air at 35 sites, 1.5m



Surface soil at 24 sites



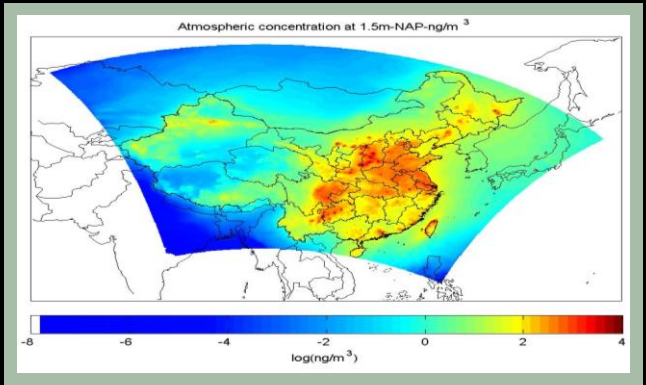
Gosan, Korea, Lee et al., JGR, 2006

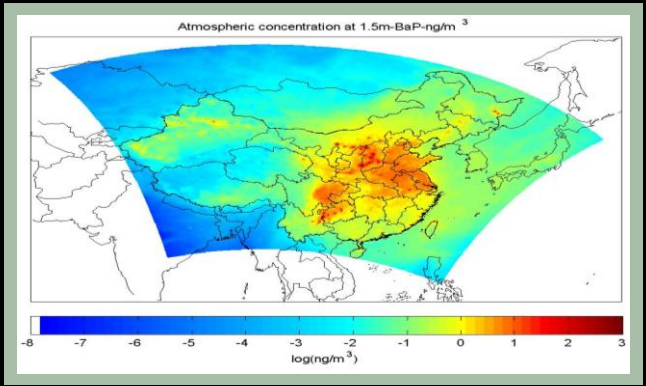


Guangzhou, China,, Li et al., 2003

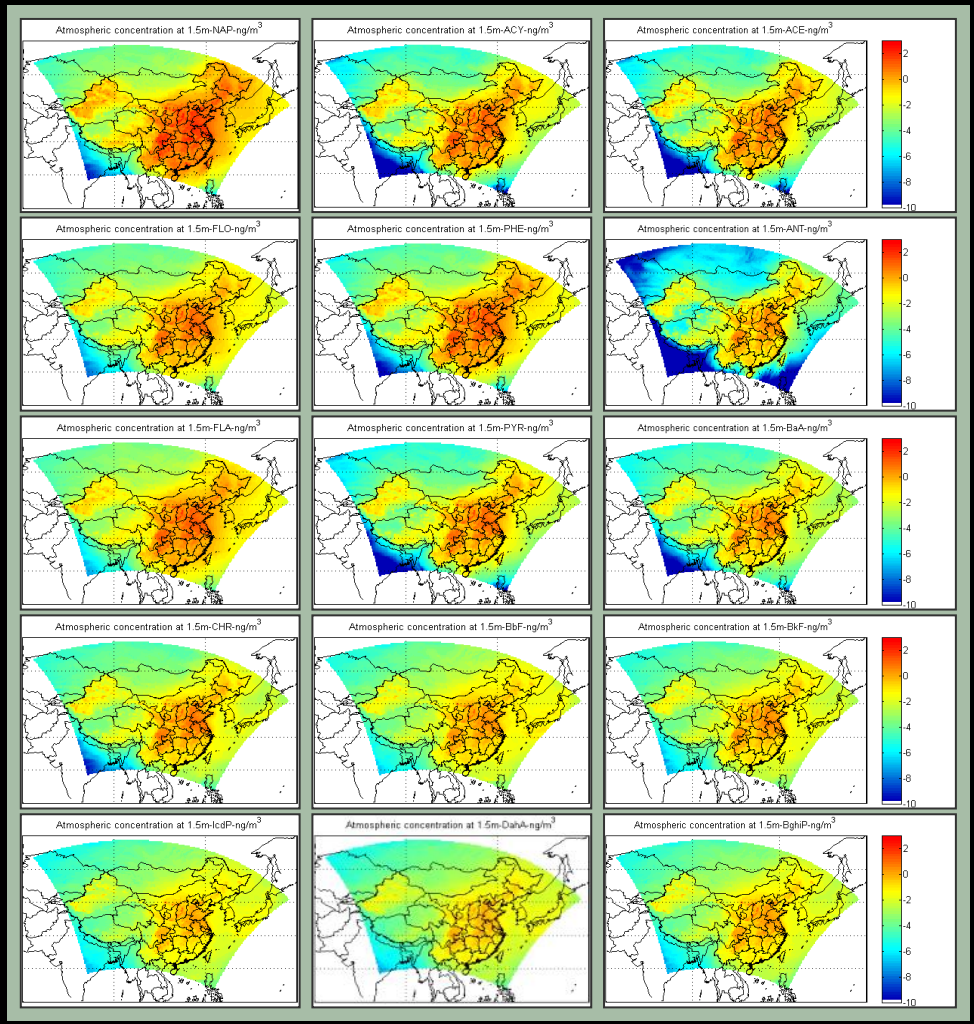
GEOGRAPHICAL DISTRIBUTION - AIR

- China, 2003, 15 PAHs





surface soil, naphthalene and benzo(a)pyrene



Ambient air , annual means, 1.5 m above the ground

AMBIENT AIR BaPeq

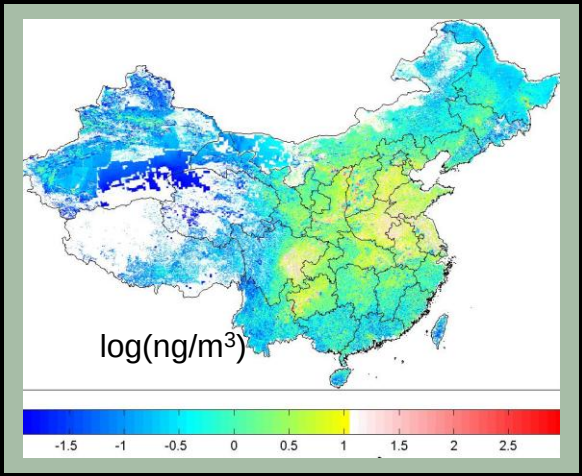
- **ambient air concentration of BaPeq** *1.5 m above the ground*

  area based mean: **2.4 ng/m^3**

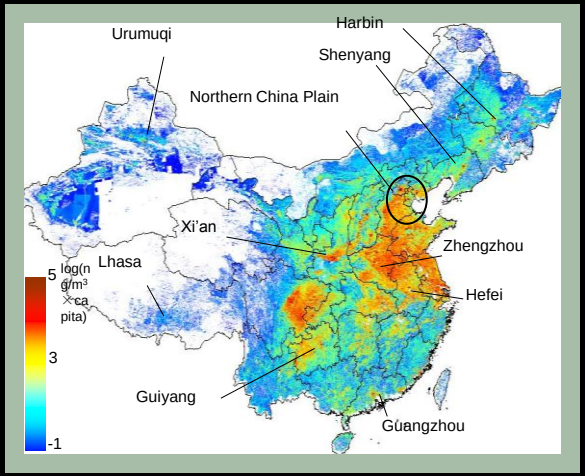
  population weighted mean: **7.6 ng/m^3**

  over 10 ng/m^3 China standard: **30% of population**

  over 1 ng/m^3 WHO guideline: **77% of population**



BaPeq, area based



BaPeq, population weighted

RE-CALCULATED RISK

- **lung-cancer risk due to inhalation exposure to PAHs**

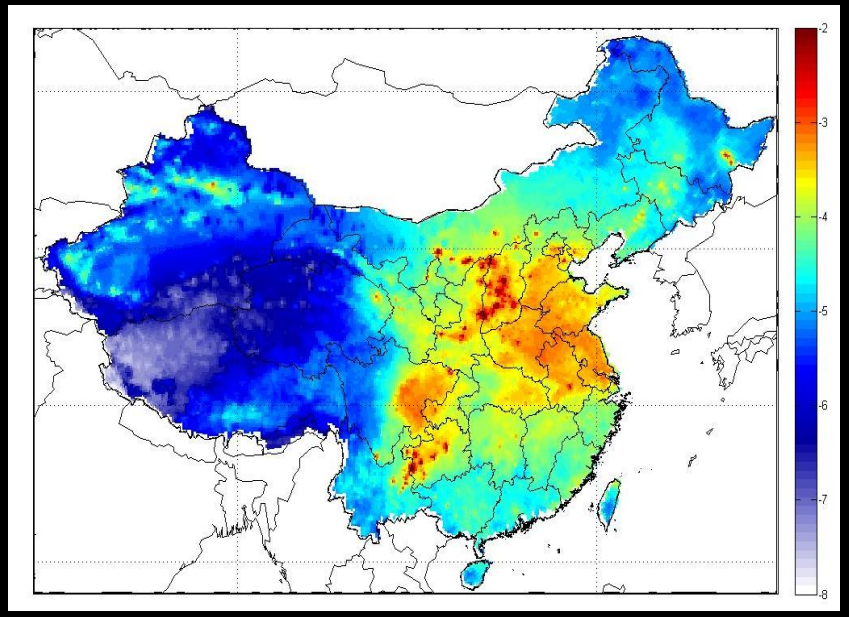
  population attributable fraction (PAF) **1.6 (0.91~2.6)%**

  population weighted extra lifelong lung cancer morbidity **51.5 (27.3~95.3)/100 000**

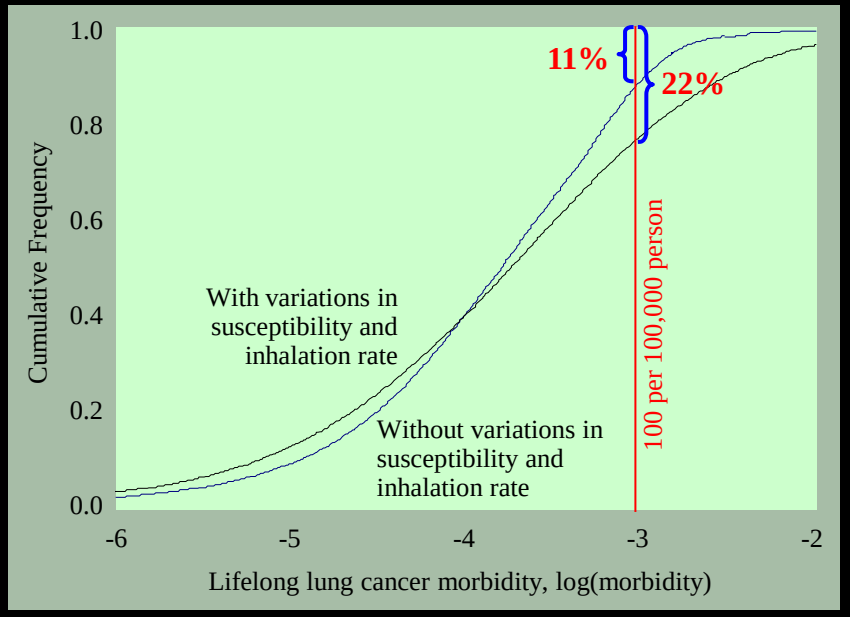
- **variation among individuals**

  22% of population have the risk greater than **100/100,000**

  underestimation for high-risk population without the uncertainty analysis



Spatial distribution of lifelong risk



Accumulative distribution of lifelong risk

SUMMARY

- population weighted annual mean BaPeq: 7.6 ng/m^3
- exposure of 30% population exceeded national standard
- population attributable fraction: 1.6% = 45.5/100,000
- high risk of vulnerable population

Environmental Science & Technology 40, 2006, 4586-4591

Chemosphere 55, 2007, 1922-1928

Science of Total Environment 383, 2007, 98-105

Proceedings of the National Academy of Sciences of the USA 106, 21063-21067

FUTURE STUDIES

- uncertainty
- indoor air
- other PAHs
- ingestion
- global

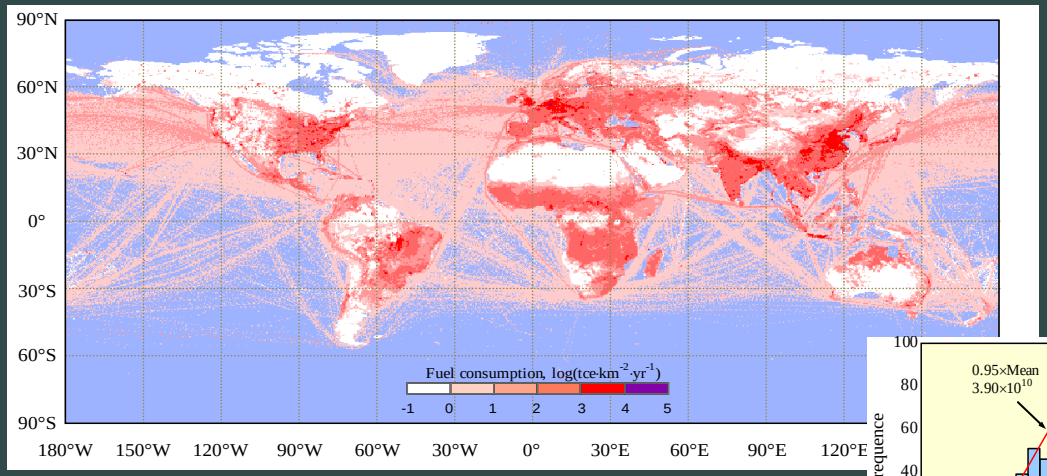
Emission factor database

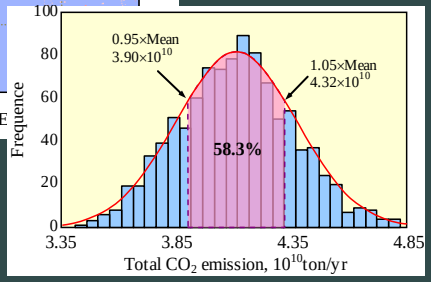
Combustion database

0.1°×0.1° combustion fuel inventory

64 fuel types

a by product of CO_2 emission inventory





ACKNOWLEDGEMENT

FINANCIAL SUPPORT

NATIONAL SCIENTIFIC FOUNDATION OF CHINA