Global anthropogenic aerosol particle number emissions

Pauli Paasonen[1,2,3], Kaarle Kupiainen[2,3], Zbigniew Klimont[2], Antoon Visschedijk[4], Hugo Denier van der Gon[4] and Markus Amann[2]

[1]University of Helsinki, Finland [2]International Institute for Applied Systems Analysis (IIASA), Austria
[3]Finnish Environment Institute (SYKE), Finland [4]TNO, the Netherlands

4th International Workshop on Uncertainties in Atmospheric Emissions, Krakow

Short description of the work

- Implementation of emission factors (EF) for particle number (PN) emissions in the GAINS (Greenhouse gas – Air pollution Interactions and Synergies) emission scenario model
  - Global emissions from continental anthropogenic sources
  - PN size distributions for particles with diameters from 0.003 to 1 µm
- First results on emissions and effect on simulated atmospheric PN concentrations

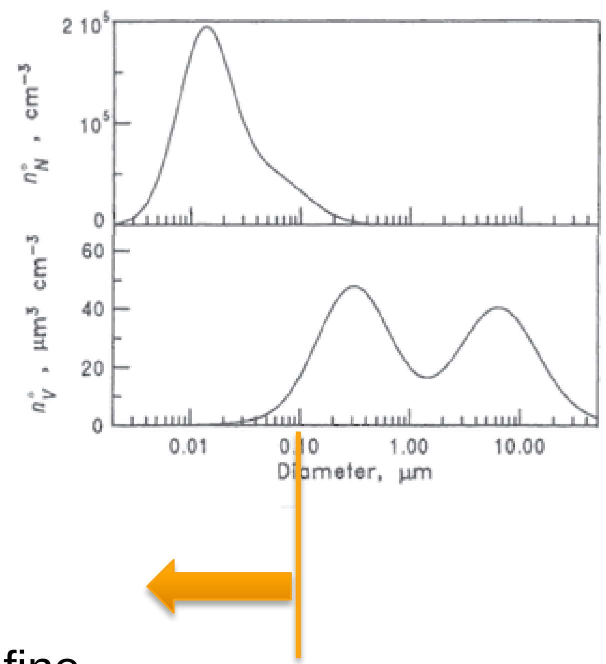
Outlook

- Why number emissions matter?
- Why new method is better than old?
- First PN results from GAINS model
- Preliminary comparison to AeroCom emissions
- Uncertainties

Why particle number emissions matter?

Number vs. mass concentration

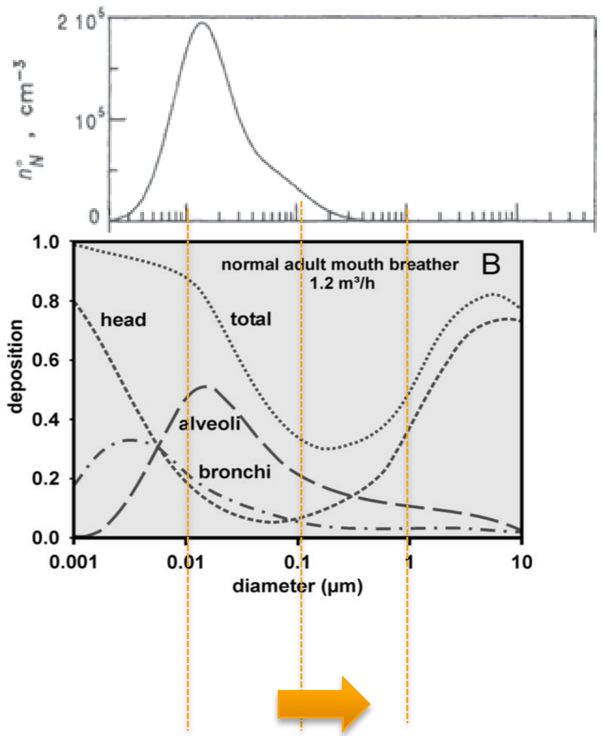
- Same particle population, different measures:
  - Number distribution
  - Volume (mass) distribution



Ultrafine particles (UFP)

Effects of particle number concentration

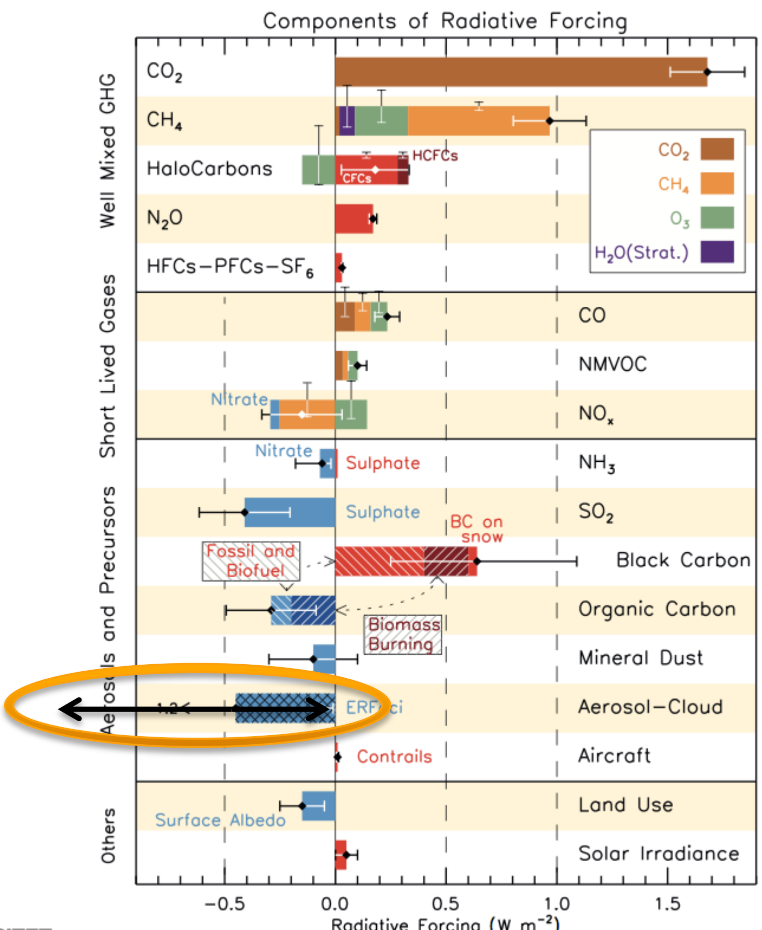
- Number distribution
- Deposition in lungs
- Cloud droplet formation



Aerosol – cloud interactions

- Each cloud droplet is formed on a aerosol particle with large enough diameter, roughly > 0.07-0.1 µm
  - Cloud Condensation Nuclei (CCN)
- Clouds reflect solar radiation back to space
- More CCN
  - -> smaller cloud droplets
  - -> higher reflectivity and longer droplet lifetime
  - -> negative radiative forcing
- Arneth et al. (2005): “Clean the air, heat the planet?”

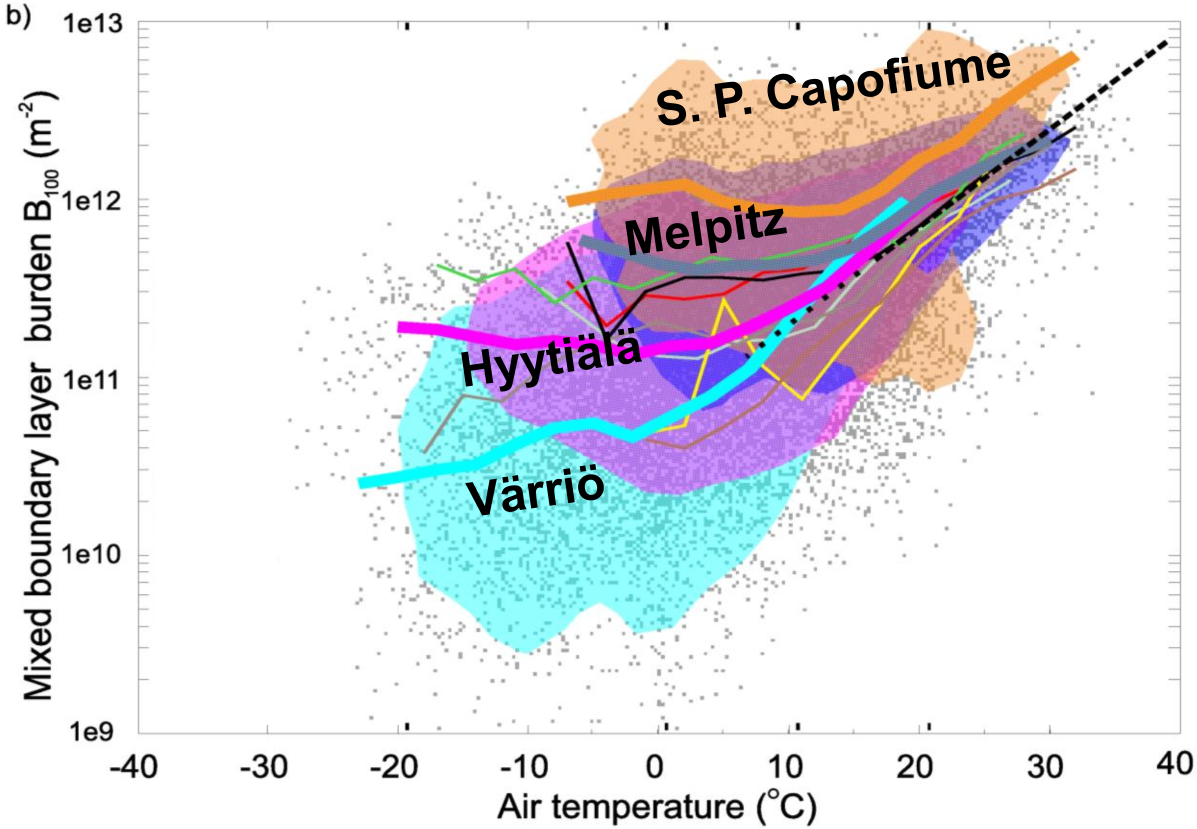
Aerosol – cloud interactions



- IPCC AR5: aerosol-cloud interactions are
  - Major source of negative anthropogenic radiative forcing
  - Major source of uncertainties in radiative forcing

Anthropogenic and biogenic CCN formation

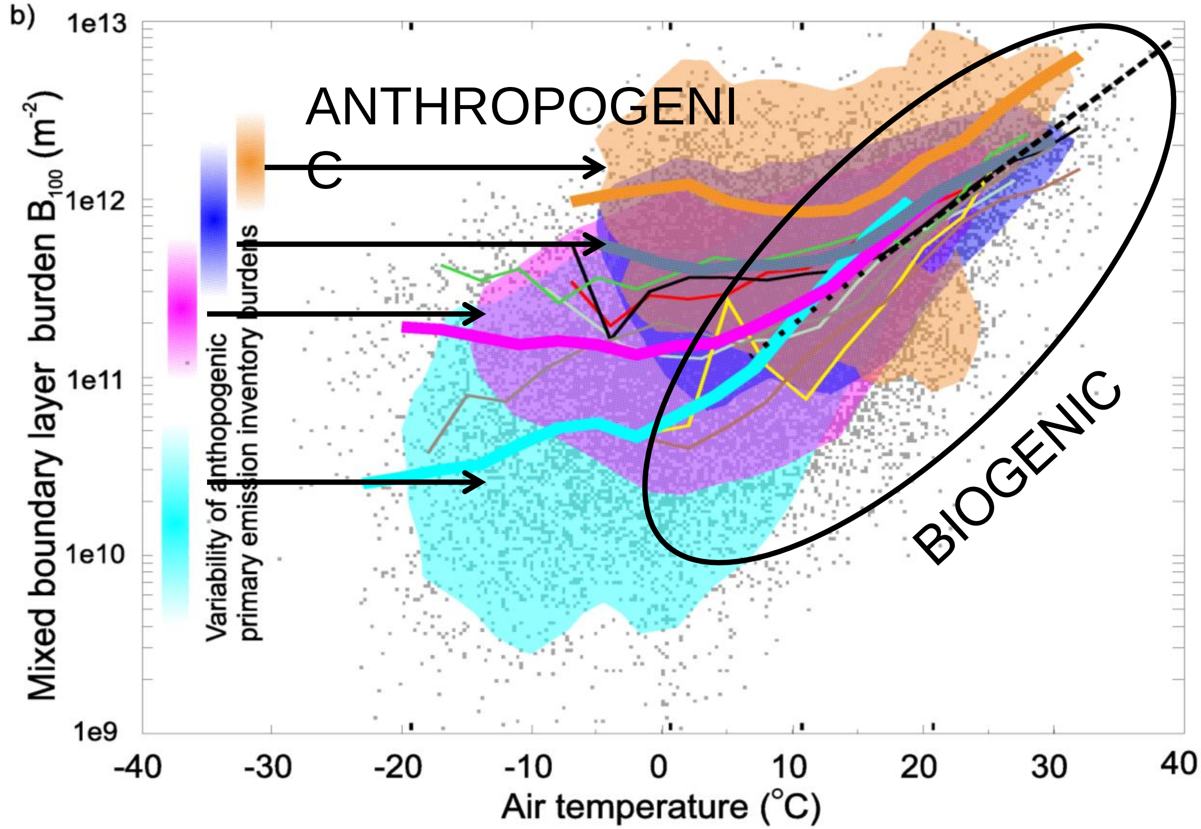
Boundary layer burden of CCN (i.e. d_P>0.1µm)



Paasonen et al., Nat. Geosci. 2013

Anthropogenic and biogenic CCN formation

Boundary layer burden of CCN (i.e. d_P>0.1μm)



Paasonen et al., Nat. Geosci. 2013

Why direct particle number emission factors and size distributions?

In climate modeling, mass emissions converted to numbers

Table 2. AeroCom anthropogenically modified (full molecular mass) emissions for the year 2000.

type	data source	time re-solution	aero type	injection altitude	$r_m[\mu m]$	σ	$r_f[\mu m]$	flux[Tg/yr] AeroCom	flux [Tg/yr] IPCC-TAR
wild-fire	GFED	monthly	BC	6 layers [H]	0.040	1.8	)95	3.1	5.7 [5–9]
	GFED	monthly	POM	6 layers [H]	0.040	1.8	)95	34.7	54 [45–80]
	GFED	monthly	S +	6 layers [H]	0.040	1.8	)95	4.1	2.2 [1–6]
biofuel	SPEW	yearly	BC	surface	0.040	1.8	)95	1.6	in wild fire
	SPEW	yearly	POM	surface	0.040	1.8	)95	9.1	
domestic	IIASA	yearly	S +	surface	0.015	1.8	)36	9.6	11.4[B]
fossilfuel	SPEW	yearly	BC	surface	0.015	1.8	)36	3.0	6.6 [6–8]
	SPEW	yearly	POM	surface	0.015	1.8	)36	3.2 (+19.1)	28 [10–30][A]
roads	IIASA	yearly	S +	surface	0.015	1.8	)36	1.9	3.6 [B]
shipping	EDGAR	yearly	S +	surface	0.500	2.0	56	7.8	7.3[B]
off-road	IIASA	yearly	S +	surface	0.015	1.8	)36	1.6	1.9[B]
industry	IIASA	yearly	S +	100–300	0.500	2.0	56	39.2 [I]	67.5[B]
power-pl.	IIASA	yearly	S +	100–300	0.500	2.0	56	48.4	53.6[B]

* S [illegible] *m(SO_2,) or m(S)=0.33*m(SO_4)], POM-particu[illegible])*orga[illegible]lack carbon

+ 2.5% of sulfur should be emitted as particulate SO_4, most sulfur (S) is emitted as gaseous SO_2

[H] 0–100 m, 100–500 m, 500–1000 m, 1–2 km, 2–3 km, 3–6 km, assignment according to Table 4

[A] Cooke et al. (Cooke et al., 1999) report a more moderate amount of 10.1 Tg OC-C/yr.

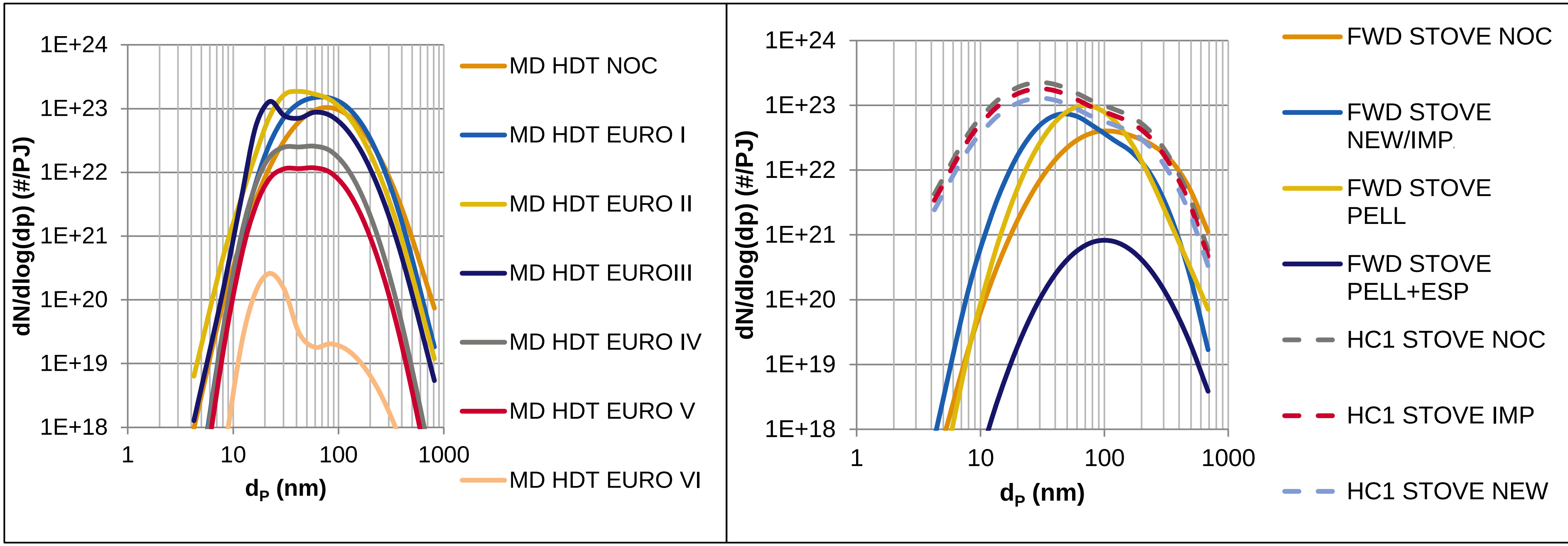
[B] based on EDGAR3.2 FT2000 (http://www.rivm.nl/edgar) and Olivier et al. (2005).

[I] probably not all industrial sources are included in the IIASA inventory.

From Dentener et al. (2006)

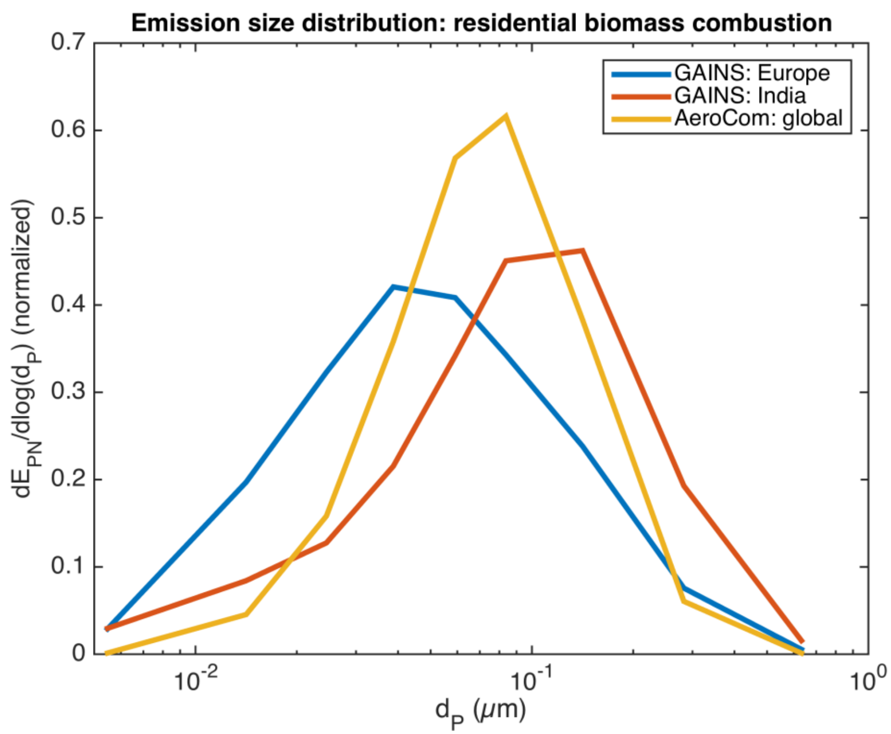
Technology impacts EF:s and size distributions

- Examples of EF_{PN}s and size distributions for low fuel sulphur content heavy duty diesel trucks (left) and residential firewood and hard coal stoves (right)



Fuel type (and quality) impacts EF:s and size distributions

- E.g. residential biofuel combustion: in India, lot's of agricultural residues and dung, in Europe firewood



- 1 kg aerosol mass distributed to particle sizes <0.4 μm, number emissions:
  - AeroCom: 1.0×10^{18}
  - GAINS Europe: 1.1×10^{18}
  - GAINS India: 0.46×10^{18}

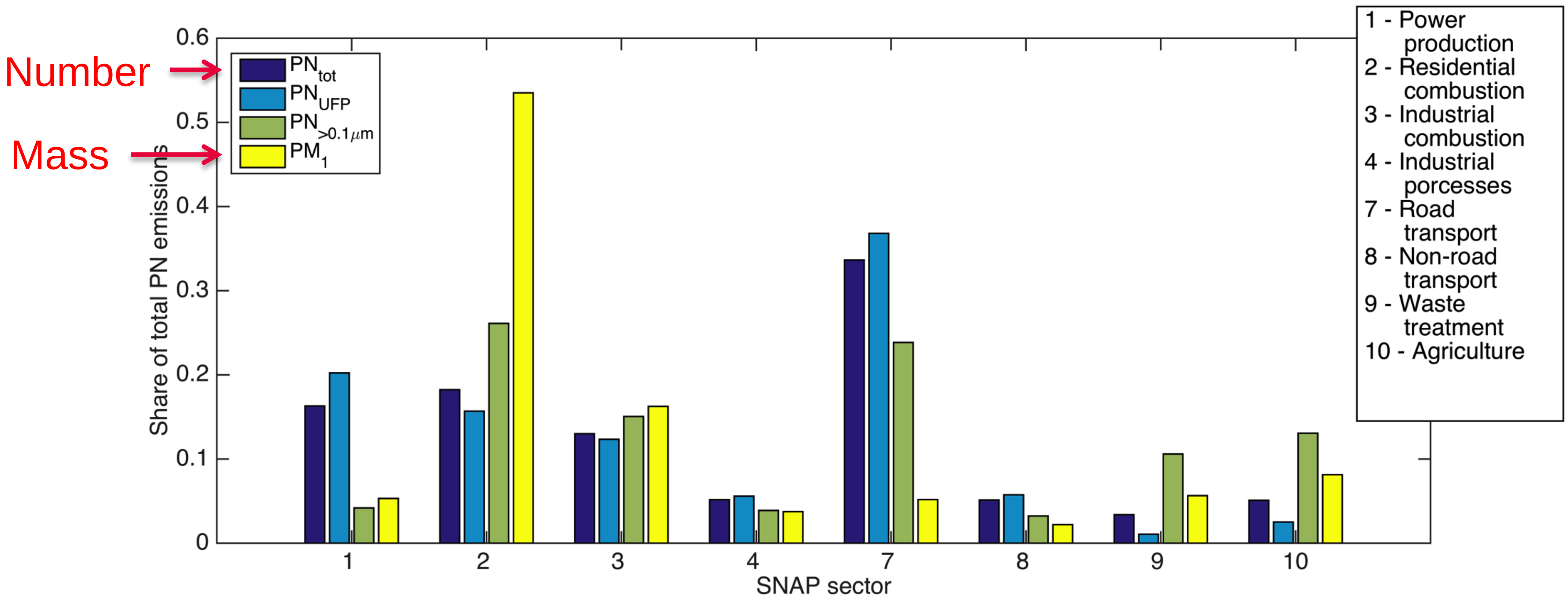
First global PN emission results from GAINS

GAINS model

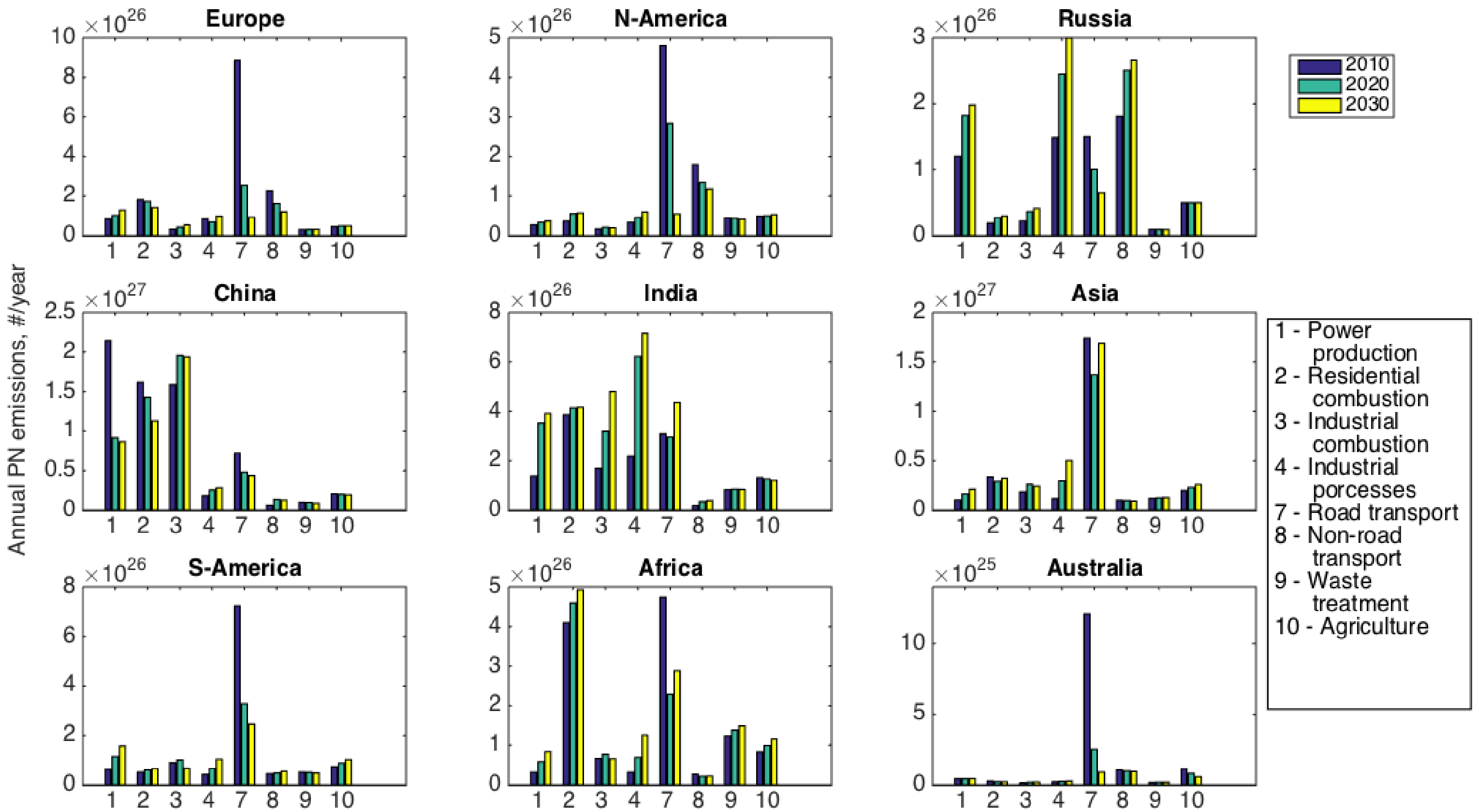
- GAINS (Greenhouse gas - Air pollution INteractions and Synergies) emission scenario model
  - Operated by MAG –program (Mitigation for Air pollution and Greenhouse gases) at IIASA (International Institute for Applied Systems Analysis, Austria)
  - Emissions: CO_2, CH_4, N_2O, NO_X, aerosols (mass: PM_{10}, $PM_{2.5}$, PM_1, BC, OC), SO_2, VOC, NH_3, CO, F-gases
  - Two space resolution levels: countries/regions (162) and gridding to 0.5°x0.5° longitude-latitude cells
  - Emissions for 1990-2050 (or -2030, depending on the scenario) in 5 years intervals

Emissions: particle mass and particle number

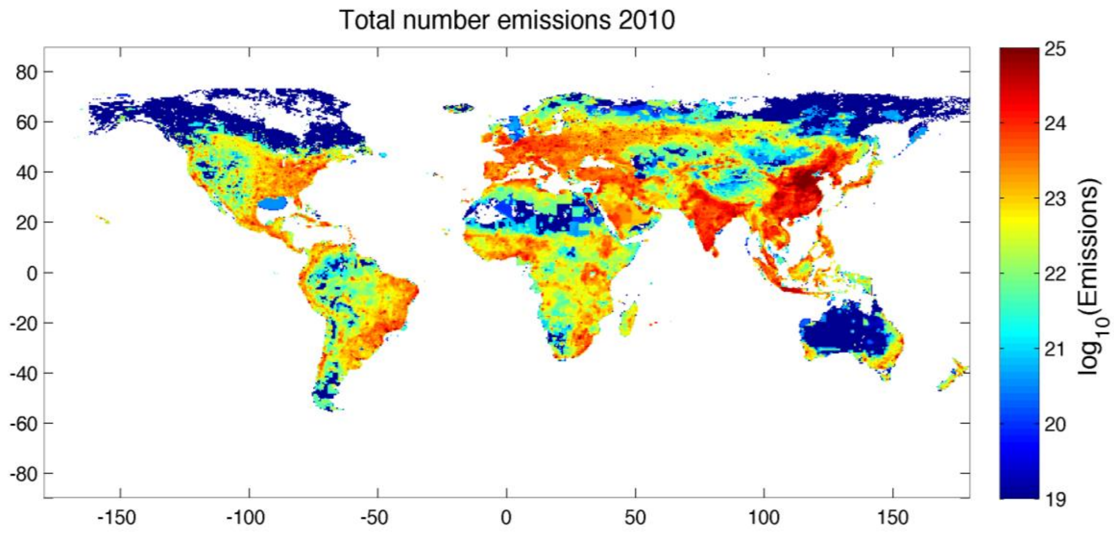
- Global mass emissions dominated by residential combustion – number emissions by road traffic

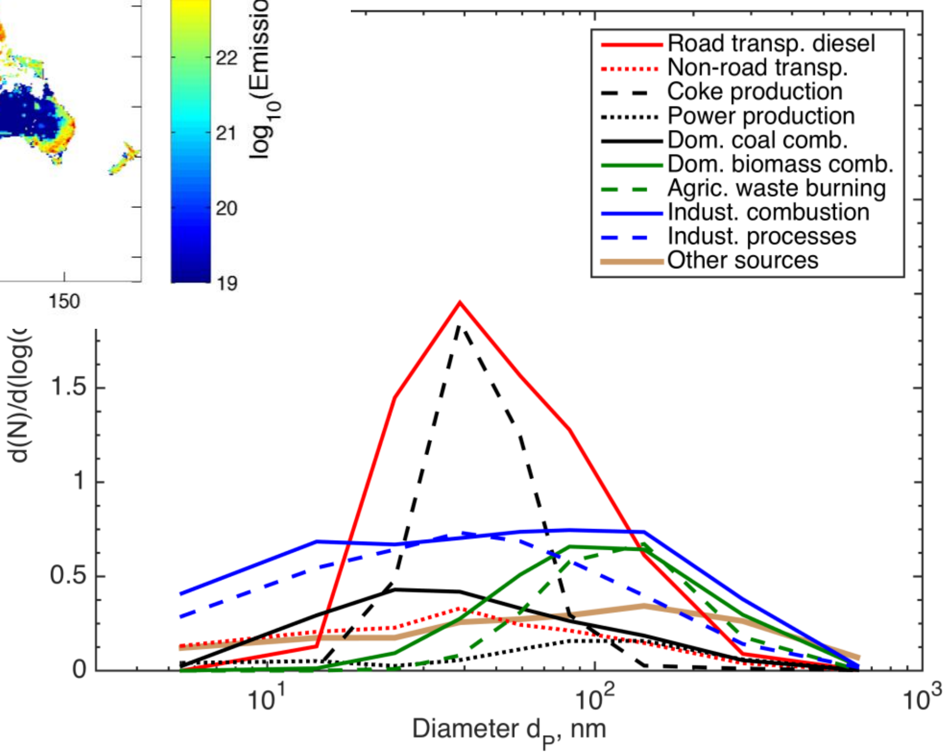


PN emissions – regional trends 2010-2030



PN emission spatial and size distribution

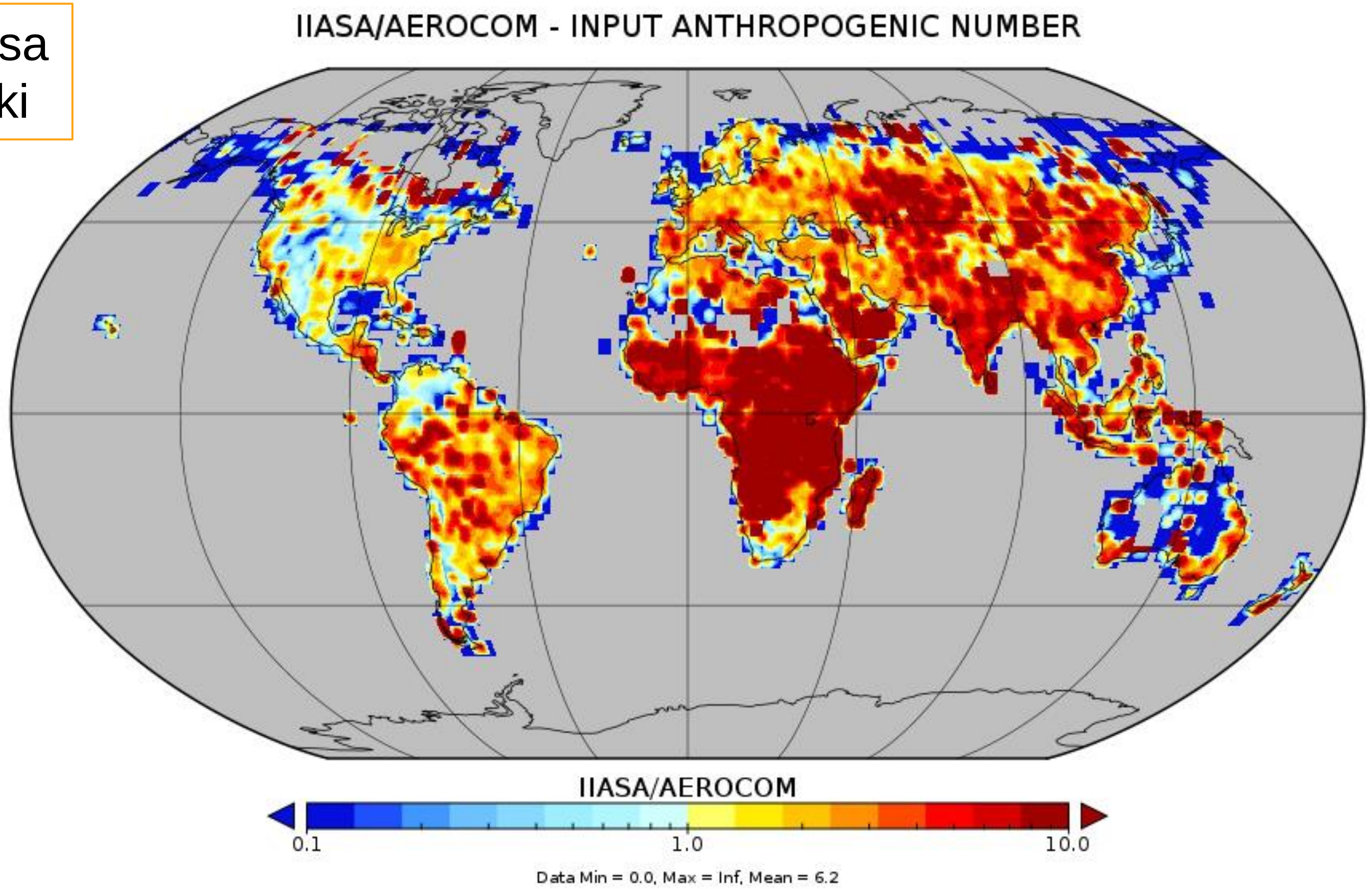




Preliminary comparison of AeroCom and GAINS emissions in ECHAM5.5

PN emissions GAINS / AeroCom

By Filippo Xausa
Univ. of Helsinki



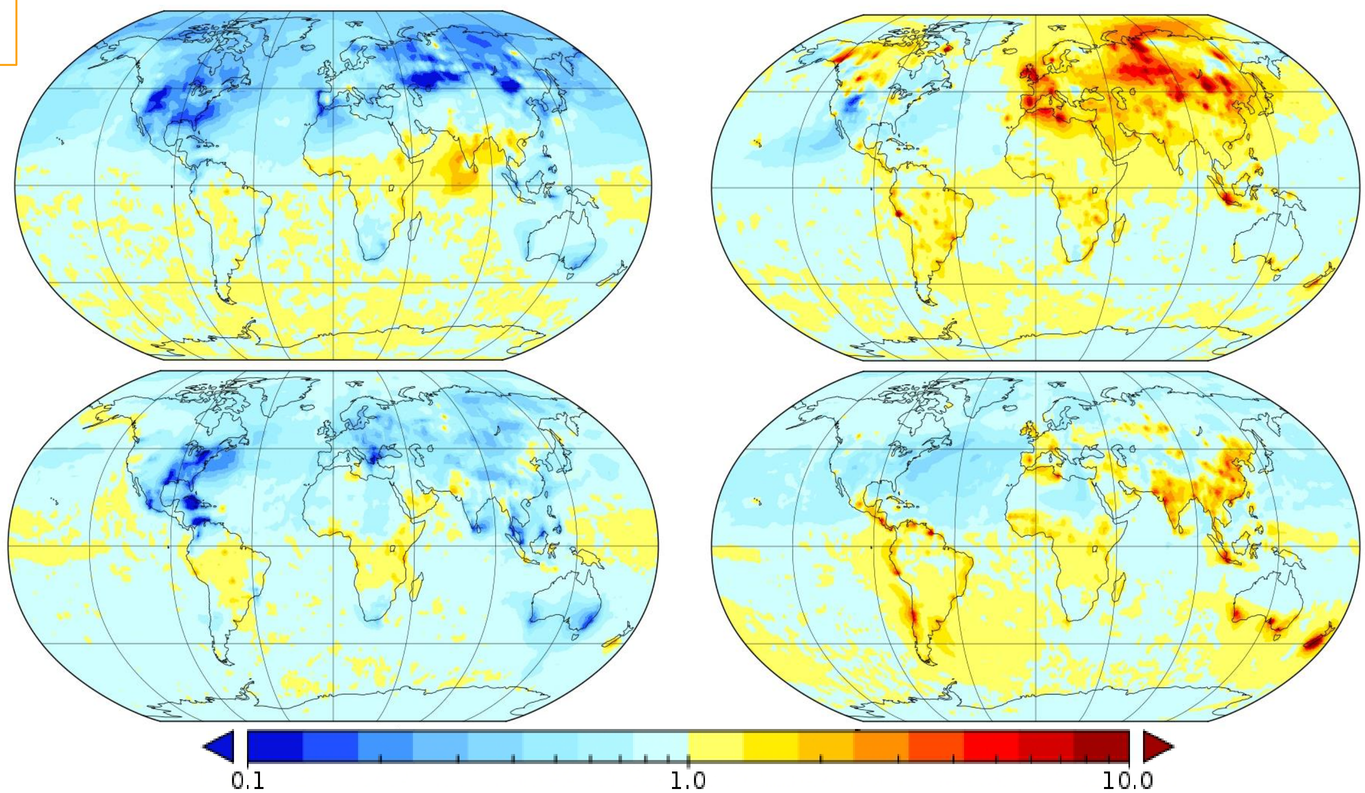
Concentration from ECHAM 5.5 Ratios: IIASA / AeroCom emissions

By Filippo Xausa
Univ. of Helsinki

d_P = 0.01-0.1 μm | d_P > 0.1 μm

January

July



Uncertainties in GAINS PN-emissions

- Main reason is uncertainties in EF:s, which are due to lack of references
  - EF:s for some major sources (e.g. coke production and resid. coal combustion) especially in China based on only one publication
  - Technology-specific EF:s for many sources (not dominant in Europe) are based on mass emission factors
  - Emissions of < 0.01 μm particles often not well described
  - Effects of varying fuel sulphur concentrations only in road transport EF:s
  - Distance from and time after emission crucial
  - PN behaves logarithmically – emission levels depend on combustion and ambient temperatures, user practices etc.
- Regional uncertainty levels vary, worst case in Asia and Africa

Additional uncertainties: adaptation of PN emissions

- Particle number size distribution evolves rapidly near the source, e.g. in street canyon, local and regional scale – sub-grid processes in AQ or Earth System Models
- Introducing the number emission to AQ and ES Models (in consistence with mass and composition) requires some work
  - Black carbon and organic carbon (aerosol mass) emissions also in GAINS, allowing for estimating black carbon particle size distribution and organic carbon / other PM shares in different particle sizes

Summary

- Particle number EF:s and size distributions implemented to GAINS
  - Improves (most probably) estimates of anthropogenic influence on cloud formation and human exposure to ultrafine particles
- Uncertainties in EF:s in certain sources are significant – geographic variation of uncertainties large
  - New measurements of PN EF:s needed
- Comparison to ambient concentration measurements to be done…

Thank you!

- Contact:
  - pauli.paasonen@helsinki.fi
- Funding:
  - Academy of Finland Centre of Excellence
  - EC 7th Framework projects ECLIPSE, PEGASOS, TRANSPHORM and ‘Assessment of hemispheric air pollution on EU air policy’
- Thanks:
  - Leonidas Ntziachristos and Ilias Vouitsis, Aristotle University of Thessaloniki (Greece)
  - Professor Qiang Zhang, Tsinghua University (Beijing, China)