Aerosol Processes in PAH Infiltration and Population Exposure in Rome

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Outline

- Scope: PAH exposures and risks in Rome, Italy, Children and elderly
- Aim: Advanced exposure modelling
  - outdoor and indoor sources of exposures
  - behavioral exposure determinants
- Implementation:
  - Analysis of exposure determinants
    - emission inventory, population time-activity measurement
    - exposure and microenvironment measurements
  - Analysis of infiltration and indoor sources

EXPAH Project

http://www.ispesl.it/expah/index.asp

- EU Life+, 2010-2013

- Focus: PAH exposures in Rome, Italy
  - risk assessment, modelling
  - chronic and short-term epidemiology

- Seven partners:

Department of Epidemiology Lazio Regional Health Service

INAIL

CNR-IIA

ARPALAZIO

ISAC

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ARIANET

Kick-off meeting in Rome, 2010

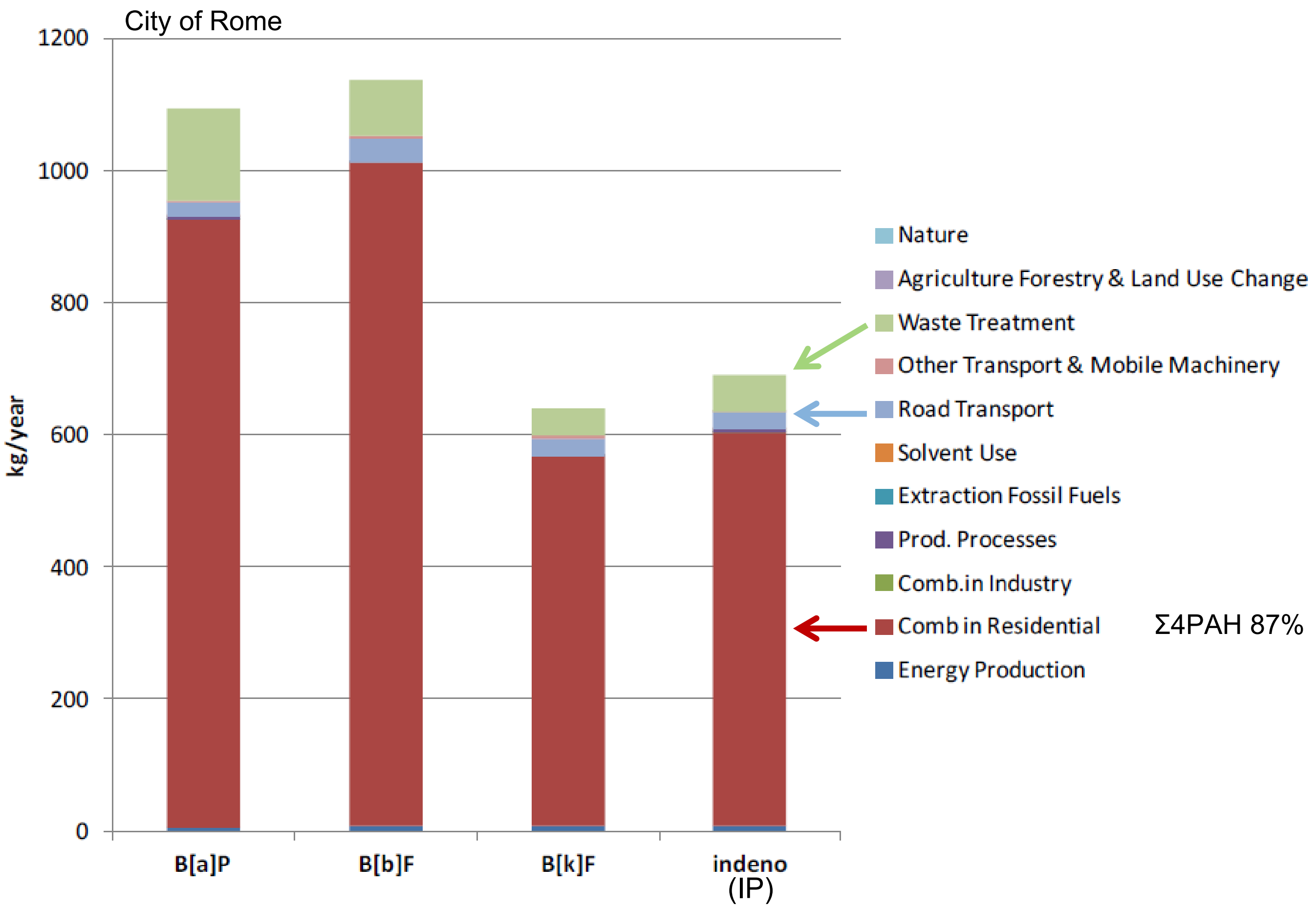
Human PAH exposure and dose

- PAHs are toxic compounds classified by the IARC as probable and possible human carcinogens.
- Health effects depend on
  - Duration of exposure (time-activity)
  - Magnitude of exposure (concentration)
  - Uptake of PAH particles in the lung
  - Toxicity of the PAH compounds
  - Health status, gender, genetics, age, …
- The aim of this work is to quantify the main processes that modify exposures, especially infiltration
  - statistical and aerosol-based modelling of infiltration
  - respiratory tract deposition estimation

EXPOSURE ANALYSIS

1

Emission inventory – 3 main sources of PAH



Source: EXPAH emission inventory (Radice et al., 2012)

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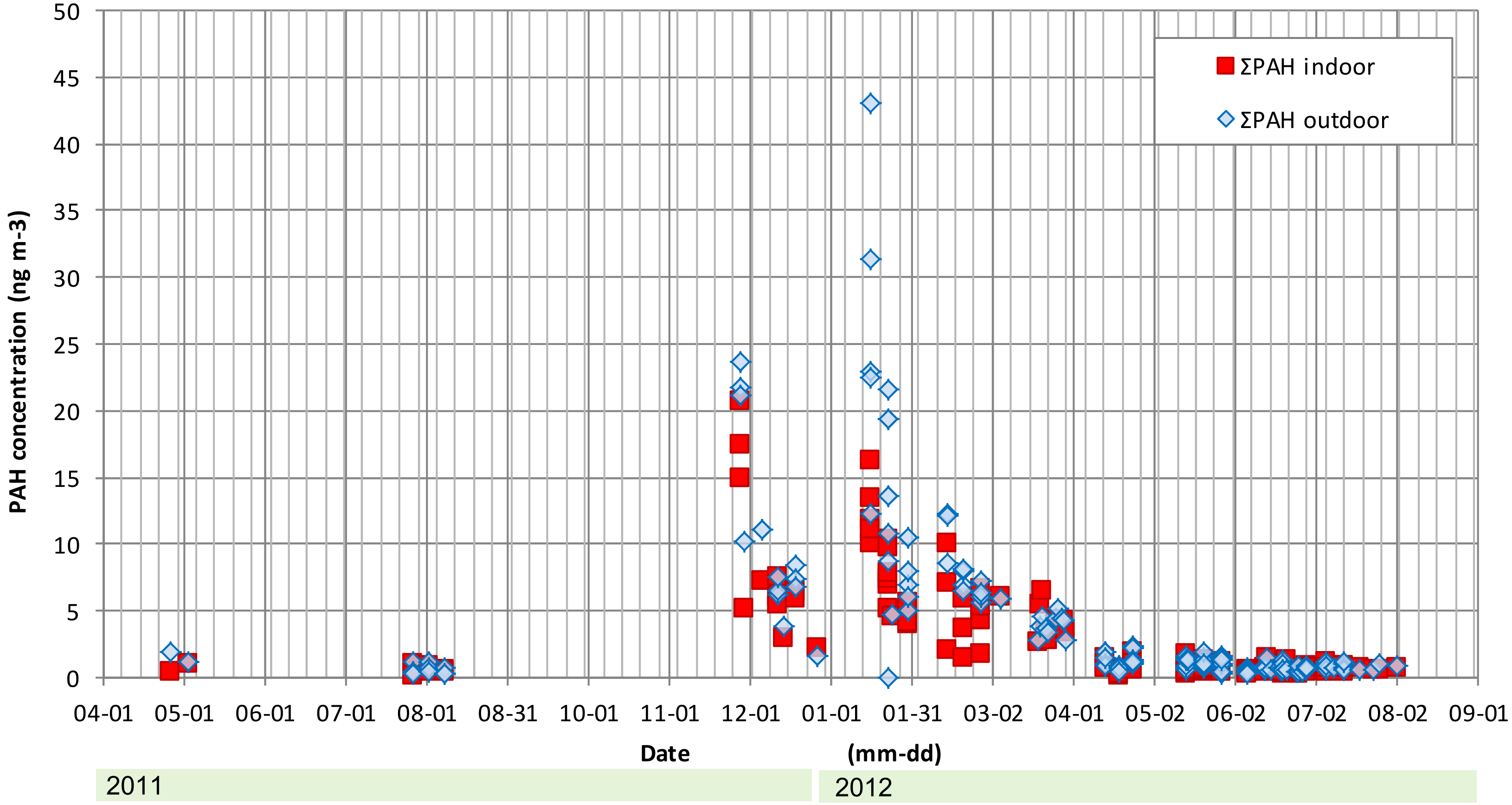
Weekly PAH concentrations by season



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Time-activity

- Questionnaire covered
  - last Wednesday and Sunday
  - two campaigns (in spring/summer and autumn/winter)
- Children (7-8 yr)
  - Mothers interviewed in face-to-face asking them to refer to the last Wednesday and the last Sunday
- Elderly (65-85 yr)
  - Letter sent to the selected people to fill in the diary of the time-activity relating to the past Wednesday and Sunday
  - After one week from the dispatch of the letter an operator called by phone the participants to record the responses

Codice: ______________

Data: Mercoledì ____________________

	TIPO DI ATTIVITA'												
	IN MOVIMENTO					NON IN MOVIMENTO							
	piedi o	motorino	macchina	autobus	metro o	Casa		Scuola		Attività sportive		Altro	
Ora	bicicletta		o taxi	o tram	treno	chiuso	aperto	chiuso	aperto	chiuso	aperto	chiuso	aperto
8.00-8.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
8.30-9.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
9.00-9.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
9.30-10.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
10.00-10.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
10.30-11.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
11.00-11.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
11.30-12.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
12.00-12.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
12.30-13.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
13.00-13.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
13.30-14.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
14.00-14.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
14.30-15.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
15.00-15.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
15.30-16.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
16.00-16.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
16.30-17.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
17.00-17.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
17.30-18.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
18.00-18.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
18.30-19.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
19.00-19.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
19.30-20.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
20.00-20.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
20.30-21.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
21.00-21.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
21.30-22.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
22.00-22.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
22.30-23.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
23.00-23.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
23.30-24.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
24.00-6.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
6.00-6.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
6.30-7.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
7.00-7.30	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐
7.30-8.00	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐	☐

Diary translated adopted from EXPOLIS study

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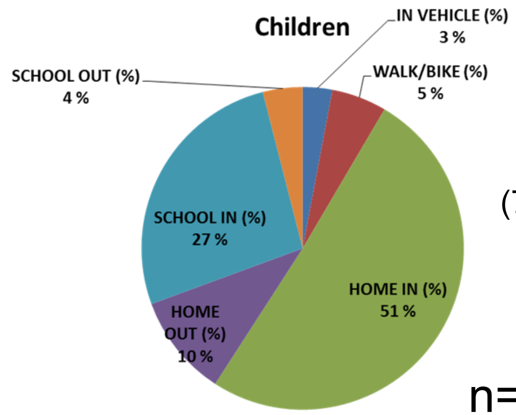
Time-activity

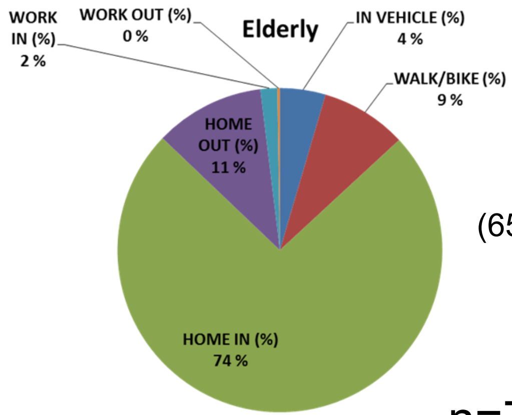
- Large differences in time-activity between age groups:
  - Elderly spend more time at home
  - and children at school (obviously)

  - children spend slightly more time outdoors but also slightly less walking and biking



(7-8 yr)

n=483



(65-85 yr)

n=707

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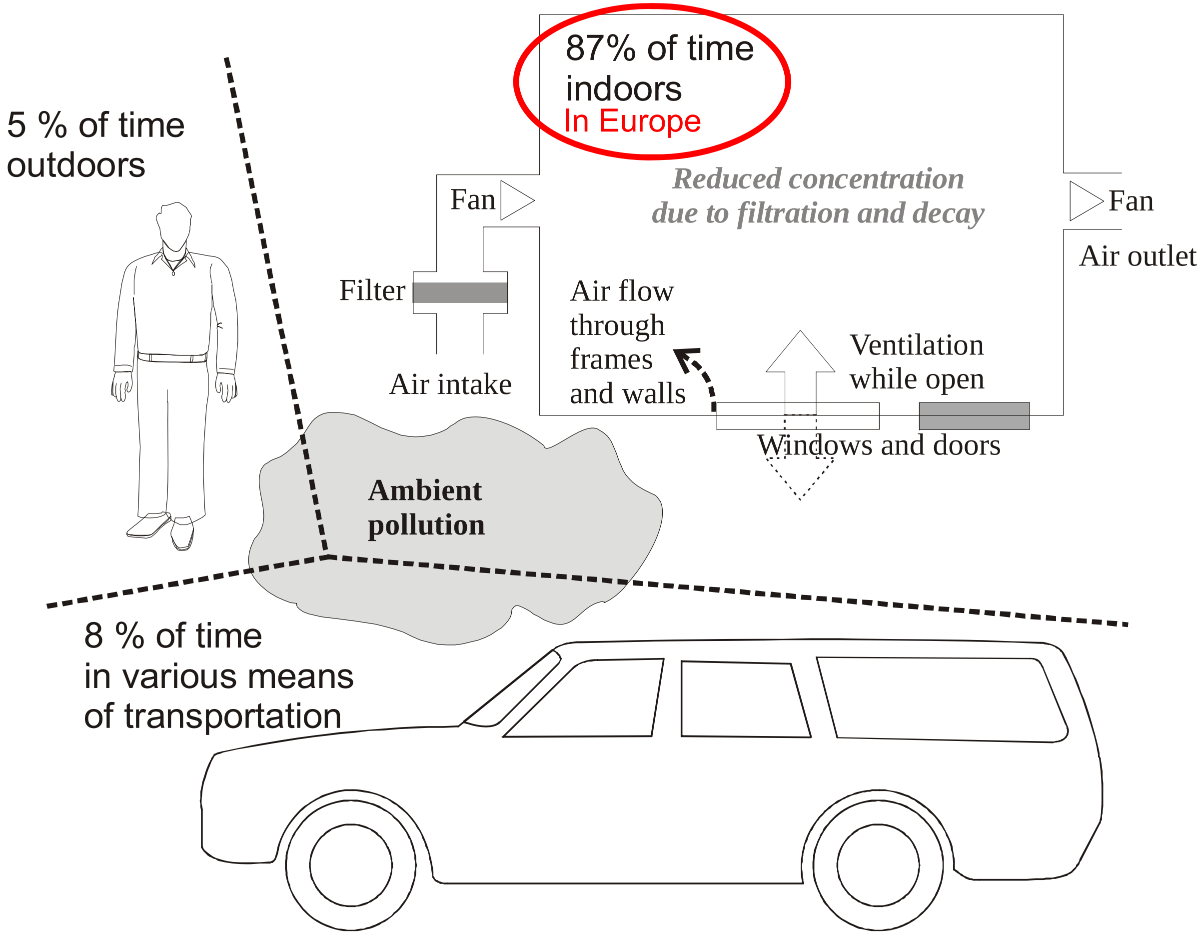
Infiltration



Source: Hänninen et al., 2005, Indoor Air

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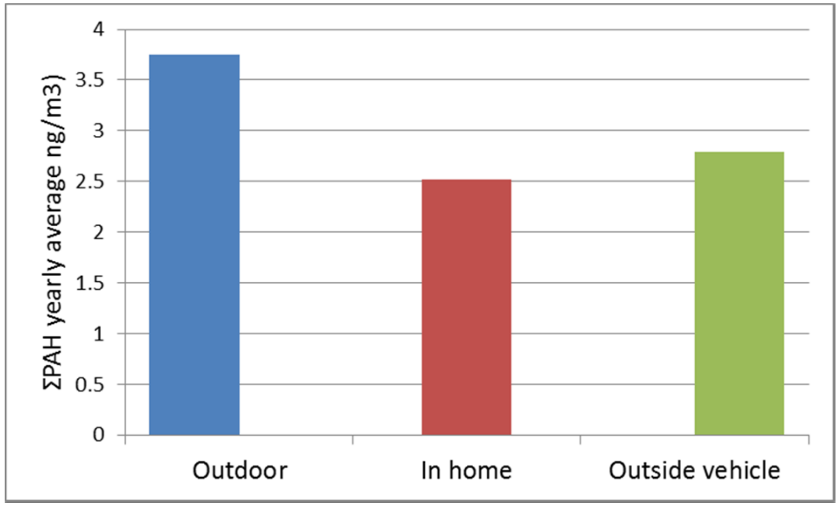
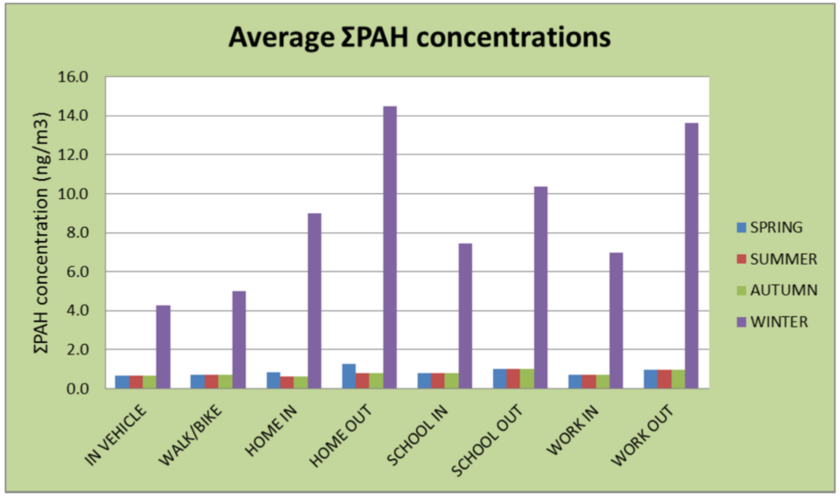
Average concentrations in microenvironments

- Overall observed average concentrations in
  - Home indoors
  - Outdoors (home)
  - Traffic

- Seasonal differences dominating:
  - Winter levels up to an order of magnitude higher





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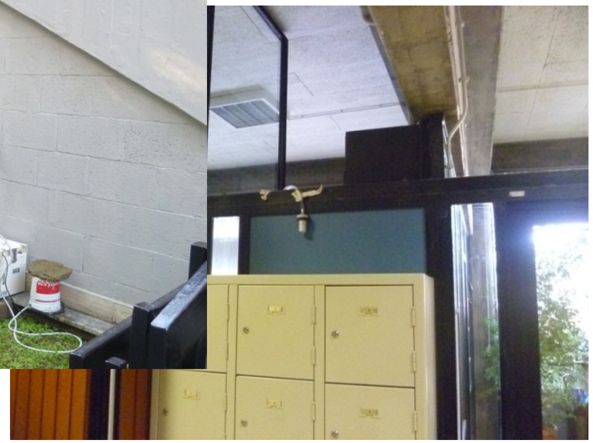
Microenvironments	Sites
Home	10
Office	4
School	6
Vehicles	3

Microenvironments measured

- Four microenvironment types:
  - homes (10)
  - schools (6)
  - offices (4)
  - vehicles (3)



- indoors and outdoors (146 successful measurement pairs)

- 1-7 day sampling with $PM_{2.5}$ inlets

Pollutants measured

- 11 individual PAH components
- 3 integrated groups (optional)

- 5 summed PAH groups
- 3 calculated ratios
- 3 other PM-bound pollutants

Groups		Compounds
PAH compounds	1	BaA
	2	BaP
	3	CH
	4	BPE
	5	DBA
	6	IP
	7	PE
	8	BeP
	9	BbF
	10	BjF
	11	BkF
Integrated peaks		BbjF
		BjkF
		BbjkF
Summed groups		ΣBbjF
		ΣBbjkF
		ΣBjkF
		PAHs (4 compounds)
		ΣPAH (all compounds)
Ratios		BaP/ΣPAH
		BaP/PAHs
		BaP/BeP
Other pollutants		PM2.5
		EC
		OC

Measurement data

	Compound		Outdoor measurements			Indoor measurements		
			mean	sd	n	mean	sd	n
Carcinogenic								
1	BaP	ng m^{-3}	0.56	0.80	145	0.41	0.55	146
2	BaA	ng m^{-3}	0.31	0.40	145	0.17	0.22	146
3	BbF	ng m^{-3}	0.90	1.24	48	0.51	0.70	54
4	BjF	ng m^{-3}	0.09	0.02	24	0.08	0.03	24
5	BkF	ng m^{-3}	0.06	0.02	27	0.06	0.02	24
6	CH	ng m^{-3}	0.71	1.04	55	0.33	0.35	56
7	DBA	ng m^{-3}	0.10	0.14	130	0.07	0.08	132
8	IP	ng m^{-3}	0.62	0.84	145	0.46	0.57	146
Non-carcinogenic								
9	BPE	ng m^{-3}	0.69	0.96	145	0.53	0.67	146
10	PE	ng m^{-3}	0.35	0.55	50	0.16	0.19	48
11	BeP	ng m^{-3}	0.68	1.00	55	0.55	0.64	56
Combined peaks								
12	BbjF	ng m^{-3}	0.16	0.10	3			
13	BjkF	ng m^{-3}	2.12	2.25	24	1.18	0.93	30
14	BbjkF	ng m^{-3}	0.91	1.29	94	0.59	0.65	92
Summed								
	cPAHs[a]	ng m^{-3}	3.35			2.08		
	ΣC×TEF[b]	ng m^{-3}	0.76			0.54		
	sPAH[c]	ng m^{-3}	7.55	13.46	11	2.34	3.59	12
	ΣPAH[d]	ng m^{-3}	4.15	6.50	146	2.85	3.77	146
Ratios								
	BaP/ΣPAH	%	13 %	5 %	145	14 %	5 %	146
	BaP/PAHs	%	5 %	1 %	2	9 %	2 %	5
	BaP/BeP	%	13 %	30 %	49	15 %	31 %	50
Particulate matter air pollution								
	PM25	µg m-3	22.6	13.1	143	22.5	15.6	140
	EC	µg m-3	2.2	1.3	28	1.8	1.5	28
	OC	µg m-3	6.7	6.0	28	6.6	3.0	28
Calculated sums[e]								
	ΣBbjF	ng m-3	0.31	0.82	146	0.20	0.49	146
	ΣBbjkF	ng m-3	0.32	0.82	146	0.21	0.49	146
	ΣBjkF	ng m-3	0.03	0.06	146	0.02	0.05	146

[a] sum of carcinogenic PAHs (1-8)
[b] sum of Concentration × TEF for carcinogenic PAHs (using Cal-EPA TEF factors)
[c] sum of BaP, IP, B(b,j,k)F
[d] sum of compounds (1-11) (including combined peaks when individual peaks were not observed)
[e] calculated for samples for which individual peaks were available for comparison with the combined peaks data

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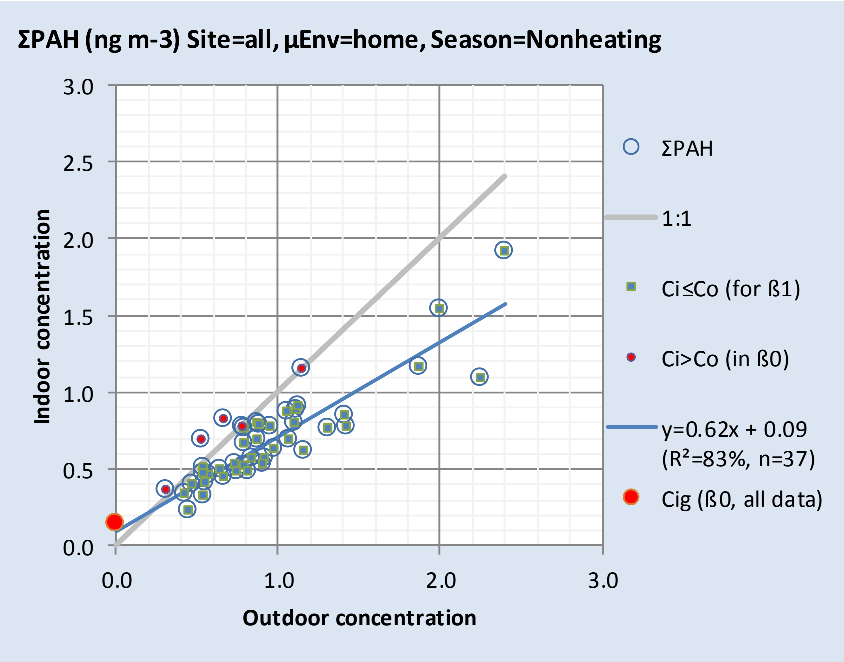
Identification of determinants of infiltration



for each

- PAH compound and group
- microenvironment type
- season
  - winter, spring, summer, autumn
  - heating, non-heating

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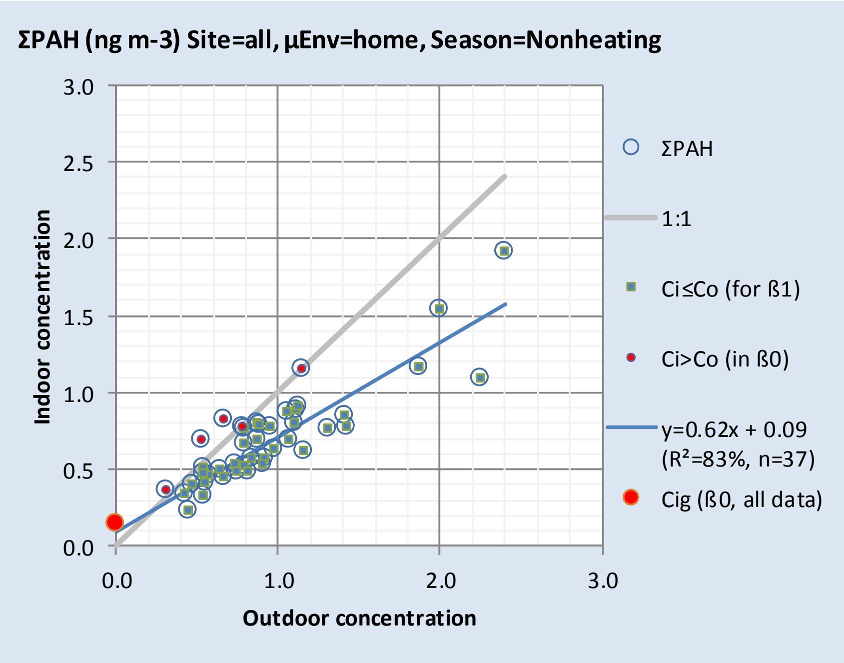
Statistical estimation of infiltration



Graphical tool for the analysis:

- selection µE, compound & season
- slope ($ß_1$) estimates Finf
- constant ($ß_0$) estimates concentration from indoor sources (Cig)
- calculation of standard errors for the estimates

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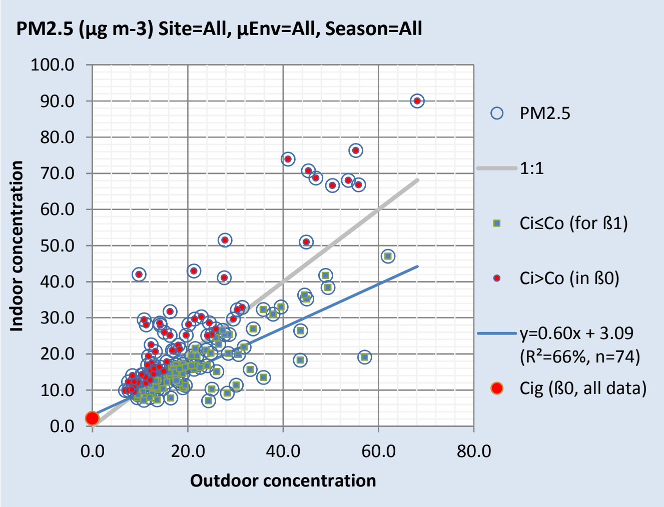
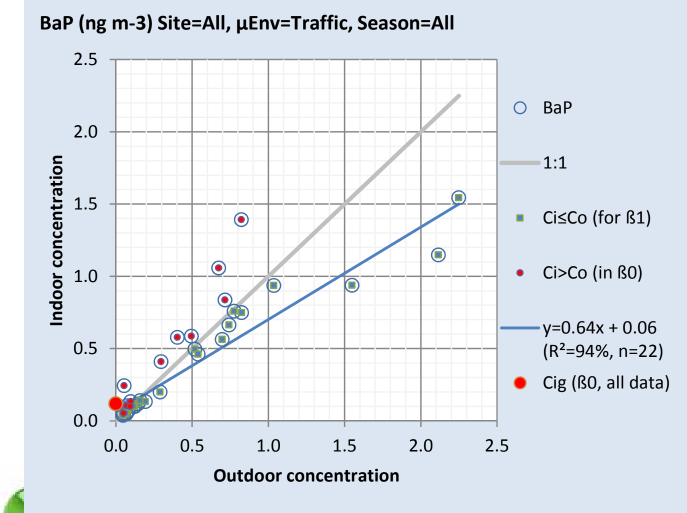
Statistical estimation of infiltration





- indoor source contribution (Cig = $ß_0$) is estimated from the data set containing all data points
- In the estimation of Finf ($ß_1$) points representing clear indoor source contribution (i.e. Cin>Cout; points shown in red) have been excluded

I/O regression analysis

- Tool for infiltration factor regression analysis
  - β1=Finf estimate (SE1=standard error for β1), β0=Cig estimate (SE0 = standard error for β0)
  - Analysis by compounds and compound groups
  - Unfiltered dataset = all data included, Ci<C0 = indoor sources removed from data

Run set 2: home by seasons					Regression results for Ci<Co dataset								Unfiltered dataset		Ci<Co -dataset	
					Finf(Ci<Co dataset)				Cig (Full dataset)				p(ß0=0)		p(Finf(µe)=Finf(All))	
	Compoun	Site	µEnv	Season	ß1	SE1	R2	n	ß0	SE0	R2	n				
31	PM2.5	all	home	all	0.647	0.066	78.3 %	29	4.382	2.260	53.1 %	53	0.026	**	0.500	.
32	PM2.5	all	home	heating	0.582	0.096	94.9 %	4	14.501	10.575	33.3 %	11	0.085	*	0.341	.
33	PM2.5	all	home	nonheating	0.324	0.068	49.8 %	25	12.272	2.138	5.8 %	42	0.000	***	0.008	***
33	PM2.5	all	home	winter	0.582	0.096	94.9 %	4	14.501	10.575	33.3 %	11	0.085	*	0.341	.
33	PM2.5	all	home	spring	0.617	0.097	67.9 %	21	8.899	2.325	16.9 %	34	0.000	***	0.427	.
33	PM2.5	all	home	summer	0.216	0.113	64.8 %	4	18.227	5.707	0.2 %	8	0.001	***	0.008	***
33	PM2.5	all	home	fall												
33	BaP	all	home	all	0.550	0.036	85.3 %	43	0.056	0.032	79.3 %	53	0.042	**	0.500	.
33	BaP	all	home	heating	0.433	0.155	56.5 %	8	0.457	0.217	50.5 %	11	0.017	**	0.269	.
33	BaP	all	home	nonheating	0.638	0.050	83.3 %	35	0.016	0.007	79.0 %	42	0.011	**	0.154	.
33	BaP	all	home	winter	0.433	0.155	56.5 %	8	0.457	0.217	50.5 %	11	0.017	**	0.269	.
33	BaP	all	home	spring	0.640	0.057	81.7 %	30	0.015	0.009	78.2 %	34	0.049	**	0.168	.
33	BaP	all	home	summer	0.868	0.136	93.2 %	5	-0.005	0.018	67.9 %	8	0.389	.	0.032	**
33	BaP	all	home	fall												
33	ΣPAH	all	home	all	0.529	0.031	86.6 %	48	0.608	0.262	81.1 %	54	0.010	**	0.500	.
33	ΣPAH	all	home	heating	0.348	0.099	58.1 %	11	4.899	1.410	52.9 %	12	0.000	***	0.081	*
33	ΣPAH	all	home	nonheating	0.618	0.048	82.8 %	37	0.148	0.055	76.5 %	42	0.003	***	0.128	.
33	ΣPAH	all	home	winter	0.348	0.099	58.1 %	11	4.899	1.410	52.9 %	12	0.000	***	0.081	*
33	ΣPAH	all	home	spring	0.618	0.049	84.2 %	32	0.108	0.055	81.5 %	34	0.024	**	0.131	.
33	ΣPAH	all	home	summer	0.727	0.068	97.5 %	5	0.242	0.148	73.4 %	8	0.051	*	0.022	**
33	ΣPAH	all	home	fall												

Infiltration estimation

- For ΣPAH estimates
  - Summer infiltration is higher than all season combined
  - Weak evidence for difference for heating season and winter (lower infiltration as could be expected)
- Limited evidence in the data on statistically significant seasonal differences in infiltration
- For ΣPAH the infiltration seems to be slightly higher during the non-heating season

	Season	
Microenvironment	Non-heating	Heating
Homes	0.62	0.62
Schools	0.82	0.68
Offices	0.51	0.37
Car and bus	0.83	0.75
Walk and bike	1.00	1.00

Aerosol modelling of infiltration

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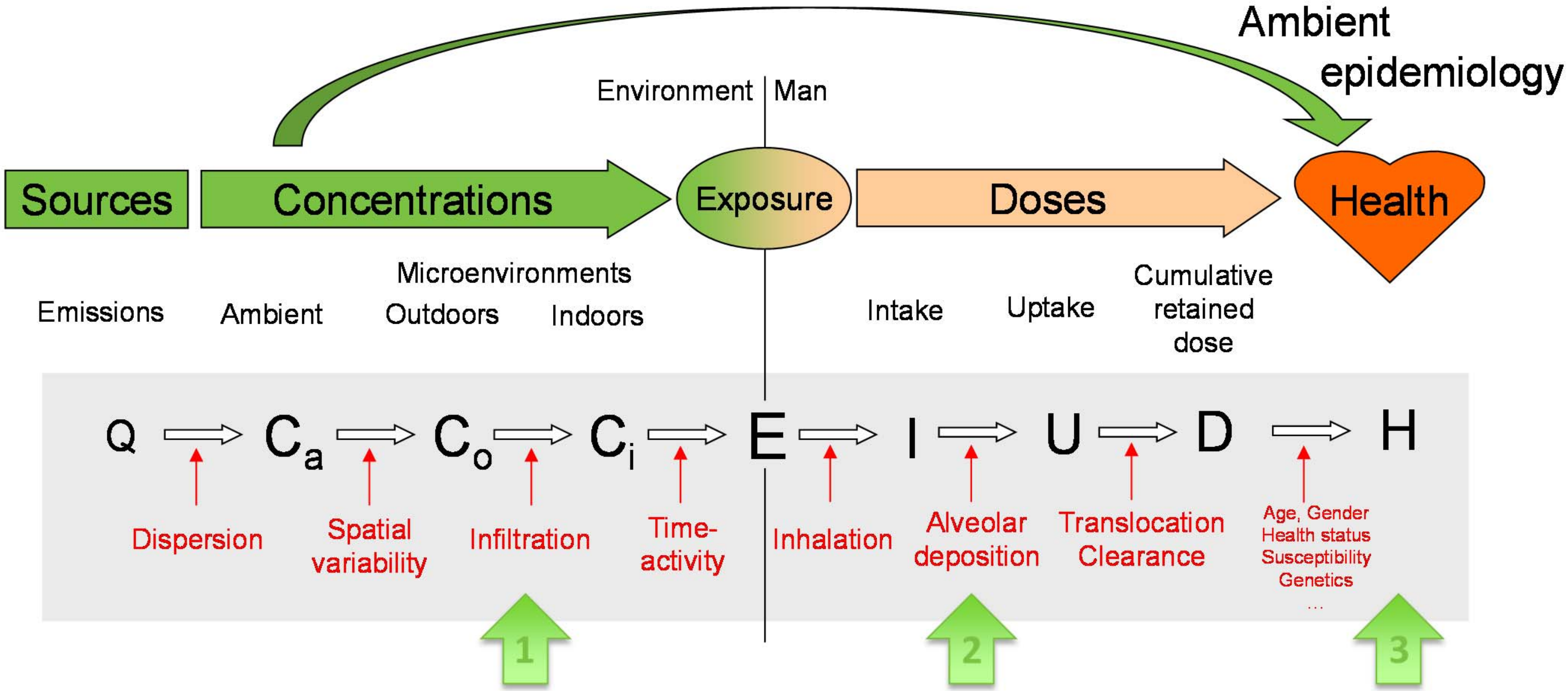
- Particle size dependence of infiltration
- Particle size dependence of respiratory tract uptake

Exposure and effect chain



Paper on evaluation of aerosol-based infiltration model

Aerosol-based modelling of infiltration of ambient $PM_{2.5}$ and evaluation against population-based measurements in homes in Helsinki, Finland

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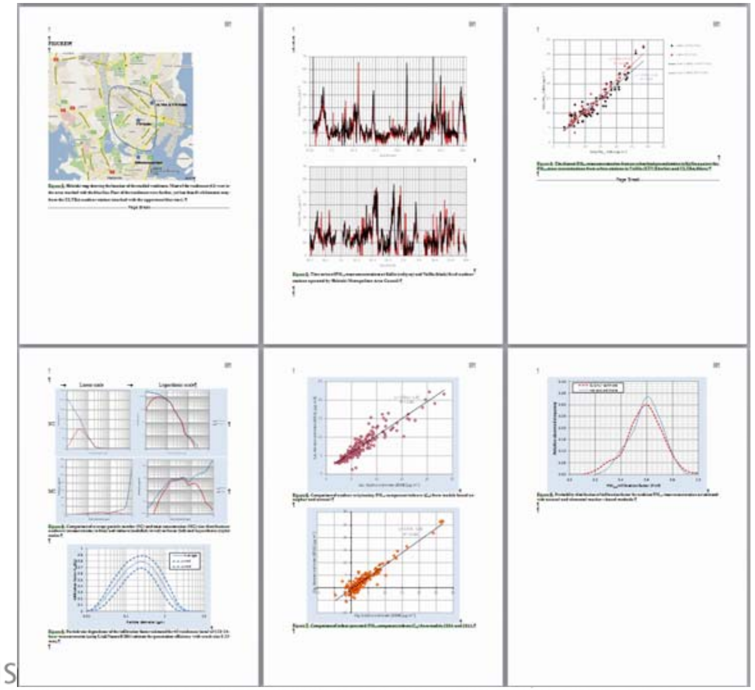
Hänninen et al., 2013.
J Aerosol Science
In print.



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Probability distribution of $PM_{2.5}$ infiltration

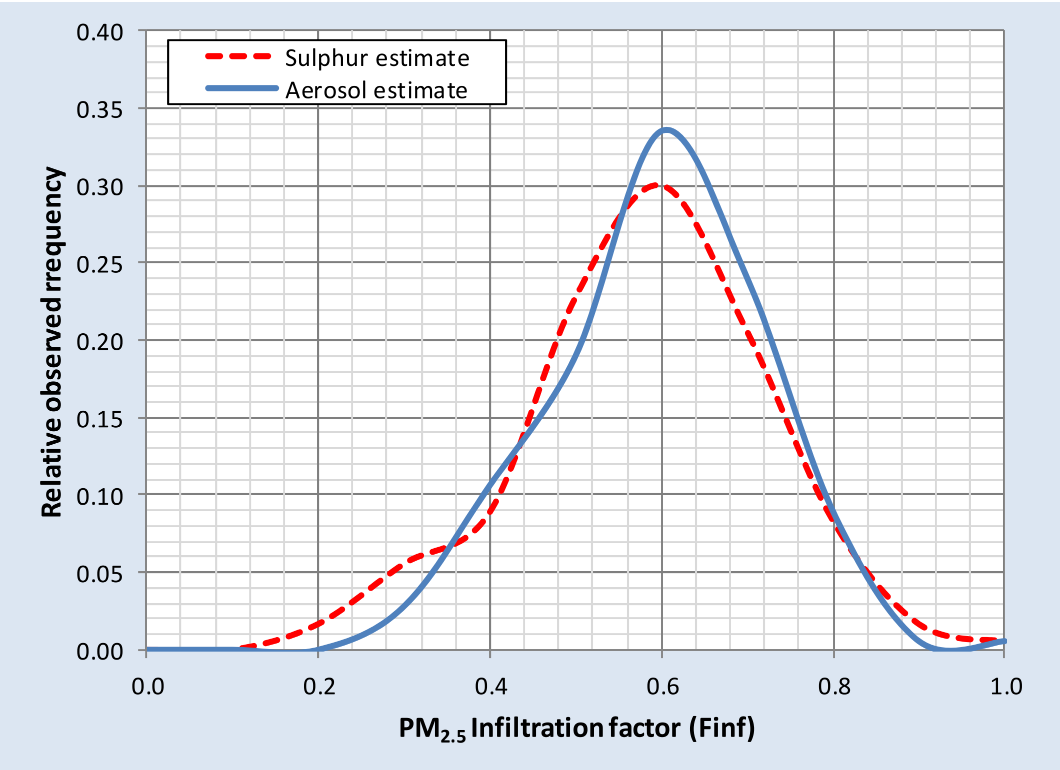


Figure 1. Probability distribution of infiltration factor for ambient $PM_{2.5}$ mass concentration as estimated with aerosol and elemental marker –based methods.

Ultra-Helsinki data

- 47 homes, ~3 measurements per home

Two methods compared:

- Elemental marker (S)

- Aerosol model (EAS, 12 channels)

Sources:
TRANSPHORM Deliverable D2.5.2
Hänninen et al., 2013

Wilson et al., 2000
Wilson & Brauer, 2006

Infiltration: mass-balance

$$C_i = F_{\inf} C_a + C_{ig}$$

(Eq. 1) $$\overline{C_i} = \frac{Pa}{a+k}\overline{C_a} + \frac{\overline{Q}}{V(a+k)} - \frac{\Delta C_i}{\Delta t(a+k)}$$

where
C_i = indoor concentration (µgm^{-3})
C_a = ambient (outdoor) concentration (µgm^{-3})
P = penetration efficiency (dimensionless)
a = air exchange rate (h^{-1})
k = decay rate indoors (h^{-1})
Q = source strength (µg h^{-1}) (symbol used by Dockery and Spengler was S)
V = interior volume of the building (m^3)

(Eq. 2) $$\overline{C_{ai}} = \frac{Pa}{a+k}\overline{C_a} = F_{\inf} C_a$$

(Eq. 3) $$\overline{C_{ig}} = \frac{\overline{Q}}{V(a+k)}$$

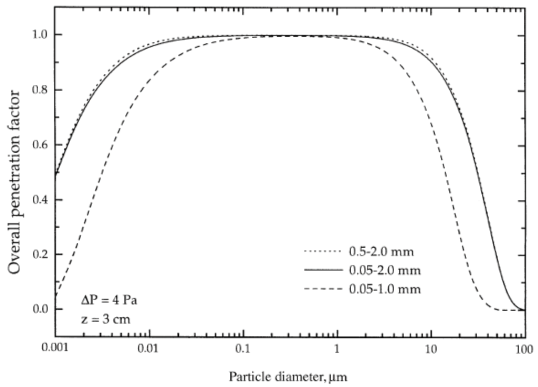
Infiltration

$$F_{\text{inf}} = \frac{Pa}{a+k}$$

P = penetration efficiency [0..1]
a = air exchange rate [h^{-1}]
k = deposition rate [h^{-1}]

Penetration and deposition modelling

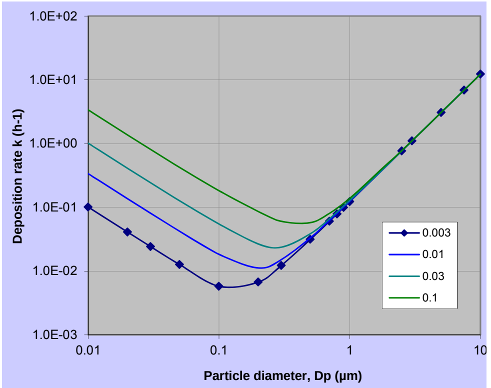
- The Liu & Nazaroff model for penetration was used to estimate the effect of building envelope on the particle size distribution
- The Lai & Nazaroff analytical model was used to estimate the particle size dependent deposition indoors



Penetration efficiency
(Liu & Nazaroff, 2001)

Parameter	Parameter value in model
Particle density (g/cm3)	1.5
Indoor temperature (°C)	20
Crack dimensions d, w, z (m)	0.003, 1, 0.03
Pressure difference (Pa)	4
Deposition surfaces AvV, AuV, AdV (m2 / m3)	2.9, 0.75, 0.75
Friction velocity (m/s)	0.03

Air exchange rate of 0.5 was used



Deposition rate
(Lai & Nazaroff, 2000)

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Particle size dependence of infiltration factor (Finf)

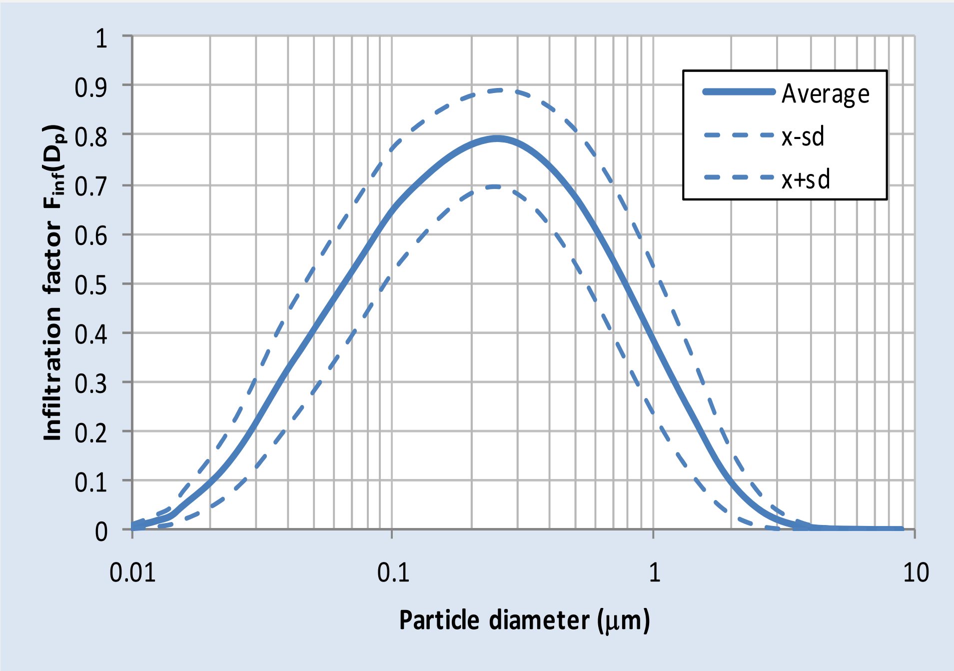


Figure 1. Particle size dependence of the infiltration factor estimated for 45 residences (total of 120 24-hour measurements) (using Liu&Nazaroff 2001 estimate for penetration efficiency with crack size 0.25 mm).

SD shown representing variation due to the variable particle size distribution and air exchange rate (not z, u, rho)

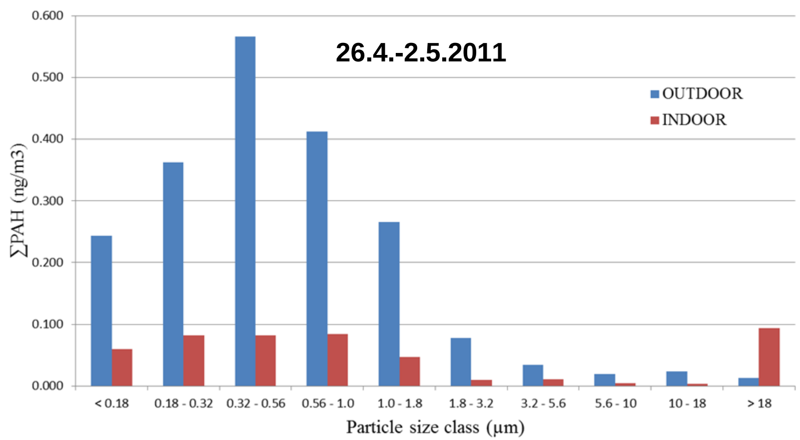
Strong particle size dependence

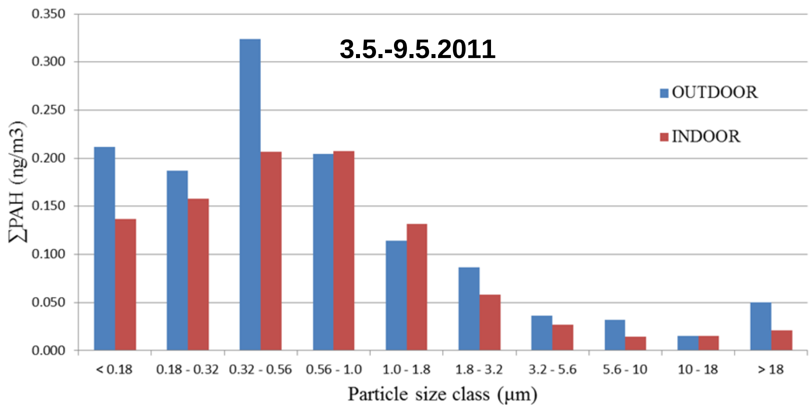
UF and coarse infiltrate poorly!
=> outdoor exposures likely to dominate!

Acc.mode ($PM_{2.5}$) infiltrates well
=> indoor exposures dominate

Sources:
TRANSPHORM Deliverable D2.5.2
Hänninen et al., 2013

Observed PAH size distributions





- Two 1-week indoor and outdoor size distribution measurements were conducted at institutional Montelibretti measurement site using a Berner ten-stage cascade impactor collecting particles from <180 nm up to >18 µm

	26.4.-2.5.2011		3.5.-9.5.2011	
	OUT	IN	OUT	IN
Ultrafines < 180nm	12 %	13 %	17 %	14 %
Accumulation 180nm – 1µm	66 %	52 %	57 %	59 %
Coarse > 1µm	21 %	35 %	26 %	27 %

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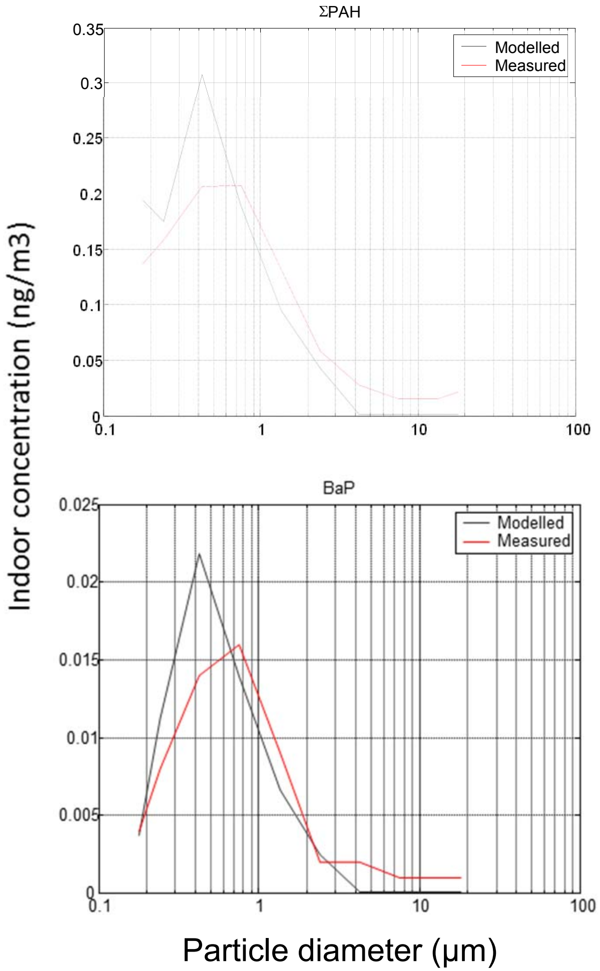
Aerosol infiltration model testing

- PAH size distribution data was available from one measurement site (Montelibretti) for two weekly averages in spring (26.4.-2.5.2011 and 3.5.-9.5.2011)
- Observed size distributions were used to test the aerosol model parameters for estimating infiltration
- Results showed relatively good match with observed indoor concentrations



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Aerosol infiltration modelling

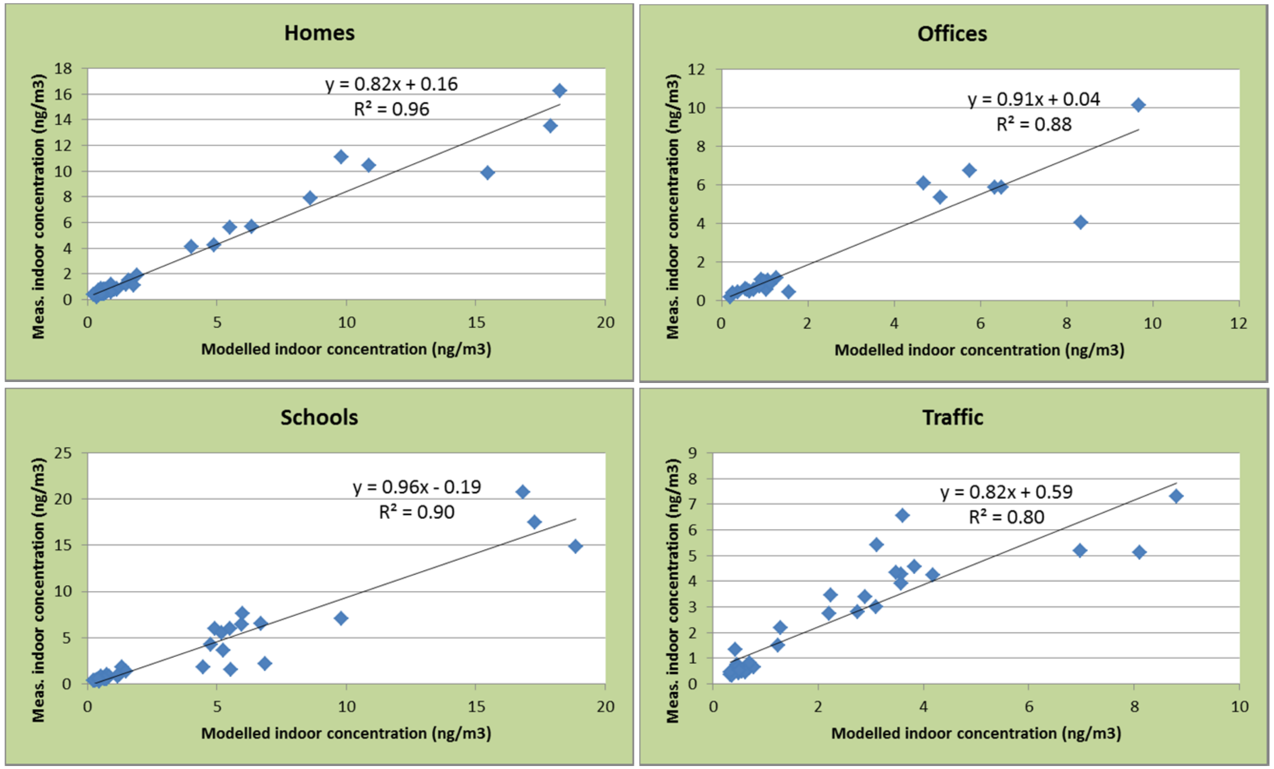
- After testing the infiltration model parameters, model was used to all measured sites (homes, schools, offices and traffic)

- Each monitored microenvironment, site and season was assumed to reflect similar particle size distribution as observed at Montelibretti site

- Measured outdoor PAH concentrations were divided into size classes, and aerosol-based infiltration model was used to predict the indoor concentrations in homes, offices, schools and traffic

Aerosol infiltration modelling

- The model captures the indoor concentration in all microenvironments quite well (R^2 values are between 0.80 and 0.96 in all cases).
- However, the model slightly over predicts the observed indoor concentrations in homes
  – Smaller air exchange rate in homes?



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Microenvironment model for personal exposure

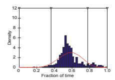
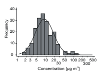
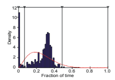
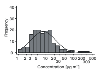
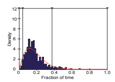
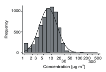
3

- Microenvironment model:

$$E = \sum f_i \times C_i$$

Selection of relevant microenvironments

Corresponding time-activity data

Microenvironment	Fraction of time distribution		Concentration distribution	Partial Exposure
1 Home indoors		x		$E_1=f_1 \times C_1$
2 Workplace/school		x		$E_2=f_2 \times C_2$
3 Traffic		x		$E_3=f_3 \times C_3$
Average personal exposure:				$Etot=E_1+E_2+E_3$

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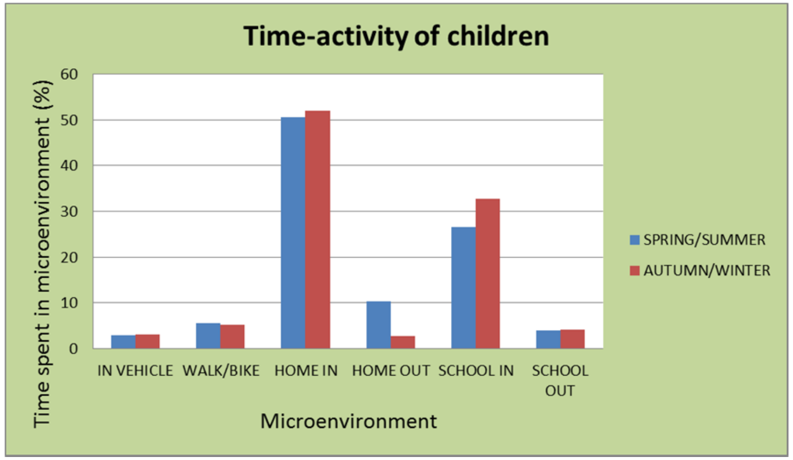
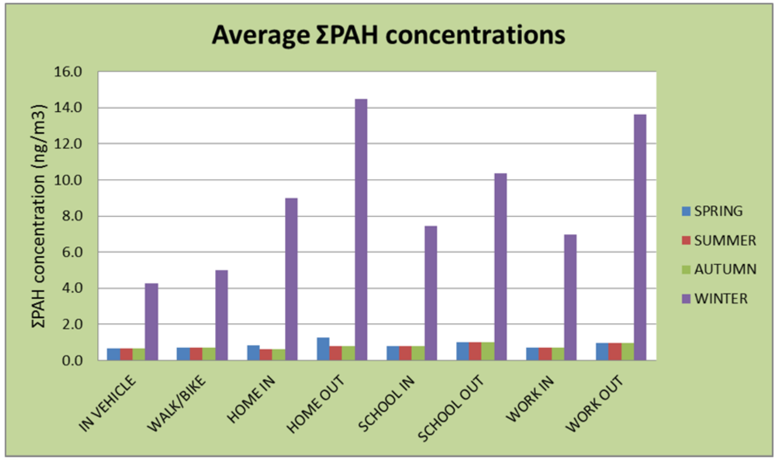
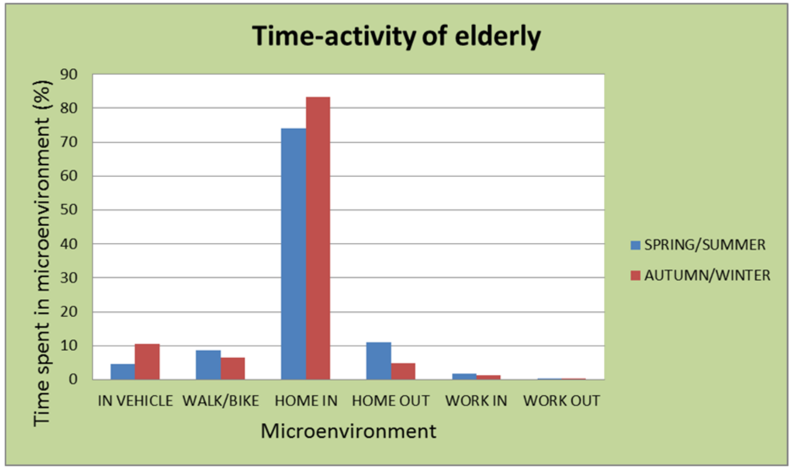
Exposure to PAH

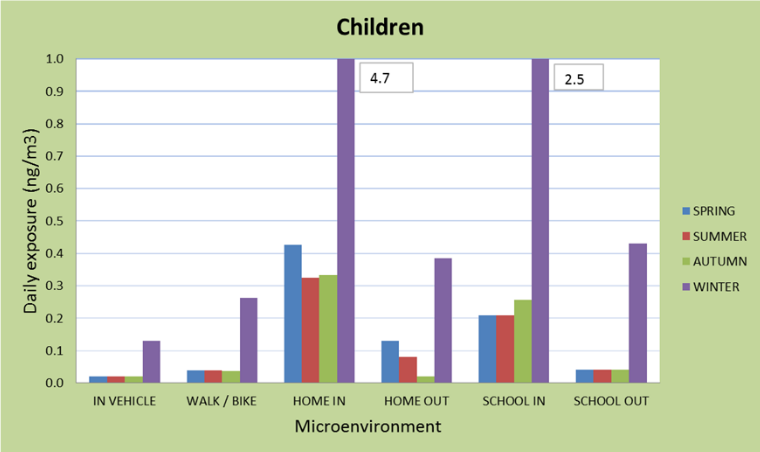
- Children and elderly PAH exposure was calculated by combining
  - Time-activity data from questionnaires (483 children and 707 elderly)
  - Average concentration levels in different microenvironments, indoors and outdoors







Exposure to PAH



- Children and elderly PAH exposure was calculated by combining the time-activity data from questionnaires (483 children and 707 elderly), with average concentration levels in different microenvironments, indoors and outdoors
- In both cases, winter season strongly dominates the exposure. In summer season people spend more time outdoors than in winter time, but PAH concentrations are much lower in summertime.

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Exposure to PAH

- Children's PAH exposure
  - Contribution of winter is 79% of the total annual exposure
  - Home indoor environment contributes 54% of the exposure

- In case of elderly PAH exposure
  - Contribution of winter is 80% of the total annual exposure
  - Home indoor environment contributes 80% of the exposure

Summary 1

- Outdoor levels are dominated by regional combustion sources
  - Winter levels an order of magnitude higher and represented 80% of annual exposures
  - Impact of traffic was close to negligible
- Indoor concentrations and exposures were observed to be strongly linked with outdoor concentrations
  - Indoor sources may occur, but played a minor role
- Seasonal differences are small in the infiltration
  - In winter time windows are kept close, but temperature difference increases natural ventilation via cracks

Summary 2

- Aerosol model estimated indoor concentrations surprisingly well, accounting for roughly estimated parameters and that…
- Particle size distribution data was limited
  - Ultrafine and accumulation modes contain majority (70-80%) of PAH mass
- Lung deposition model was demonstrated with the limited data
  - Daily deposited mass in the elderly lungs is almost 20% higher than in children
- Further work is needed
  - Description of particle size distributions related to particle infiltration and lung deposition modeling
  - Especially, seasonal variation of PAH compounds particle size distribution is worth of studying
  - Also, air exchange rates and deposition surfaces needs further study