TRANSPHORM
transport pollution health

SEVENTH FRAMEWORK PROGRAMME

TRANSPHORM submitted deliverable

Applying the size-segregated PM SIZEx model: ESCAPE cohort site particle uptakes in the lung

D2.5.4

Partners: THL, UH, NILU, FMI, AUTH, JRC, HMGU, DMI

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Applying the size-segregated PM SIZEx model: ESCAPE cohort site particle uptakes in the lung

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Executive summary

This TRANSPHORM report D2.5.4 presents annual lung deposition (uptake) estimates for FINRISK cohort members in Helsinki and Vantaa, Finland. FINRISK one of the European cohorts used in the epidemiological ESCAPE study on long-term effects of particulate matter exposures on human health. Due to the in-house collaboration in THL on the cohort (Chronic Disease Epidemiology and Prevention Unit, Unit of Environmental Epidemiology) and exposure model development (current work; Unit of Assessment and Modelling) and availability of geographical data on the survey participants, the FINRISK was selected as the target cohort for demonstrating the WP2.5 exposure and dose model applicability in an epidemiological setting. Detailed additional work on short term exposures and doses on two other cohorts, the ULTRA2 cohort in Helsinki, Finland, and KORA cohort in Augsburg, Germany, was presented in the previous reports (D2.5.1 and D2.5.2).

The current work integrates these previous TRANSPHORM WP2.5 steps in developing particle size dependent approach for estimating the impact of infiltration and respiratory tract deposition on the contribution of particles of different sizes to the uptake of an independent cohort.

The model is capable of estimating particle uptakes in five different regions of the human respiratory tract (including extra thoracic, bronchiolar, alveolar), defined by the ICRP (1994) lung deposition model, and in five alternative metrics including besides mass, also particle number and particle surface area that have been suggested as relevant for the health risks.

This report gives an overview of the methodology and a summary of the estimated FINRISK cohort member doses for year 2010. The actual database of size disaggregated annual doses for the 5700 cohort members is provided as an electronic appendix.

Deliverable D2.5.4

Work package summary and the task completed in the current Deliverable

Work package number	2.5	**Start date or starting event:**			12	
Work package title	Improved modelling of exposure to transport related pollution for combined air quality and exposure modelling					
Activity Type	RTD					
Participant number	1	4	5	8	9	**12**	14
Participant short name	UH	NILU	FMI	AUTH	JRC	**THL**	HMGU
Person-months per participant:	12	1	5	3	5	**16**	2
Participant number	16						
Participant short name	DMI						
Person-months per participant:	1						

	Year 1				Year 2				Year 3				Year 4			
	I	II	III	IV	I	II	III	IV	I	II	III	IV	I	II	III	IV
WP2.5 Exposure modelling																

Del. no.[1]	**Deliverable name**	**WP no.**	**Lead beneficiary**	**Estimated indicative person-months**	**Nature**[2]	**Dissemination level**[3]	**Delivery date**[4]
D2.5.1	Size segregated particle infiltration: a modelling framework for micro-environment seasonal ventilation and infiltration patterns	WP2.5	THL	12	R	PU	18
D2.5.2	Development and validation of the size-segregated micro-environmental model	WP2.5	THL	18	R	PU	24
D2.5.3	Evaluation of combined dispersion and exposure modelling systems	WP2.5	FMI	18	R	PU	30
D2.5.4	Exposure calculations for ESCAPE cohorts and target cities	WP2.5	THL	12	R	PU	30

Task 2.5.4: Prediction of numerical results, especially for the ESCAPE cohort sites (**THL**, FMI, HMGU, AUTH, JRC)
We will predict the pollutant concentrations for ESCAPE cohort members, and compute other predicted results for targeted cities. Exposure estimates will include integrated exposure to size distributed mass concentrations, particle number concentrations and source specific concentrations (in particular for transport sources). The results from this task will provide data directly for SP3 cohort studies.

Nomenclature and abbreviations

The following nomenclature and abbreviations have been used in the D2.5.4.

Particle properties

D_p particle diameter (μm), indicator of particle size; commonly either aerodynamic diameter (equivalent diameter of density 1 g cm^{-3} particle) or mobility diameter

Particle fraction definitions used here

ultrafine particles (Aitken + nucleation mode):	particles with diameter in between 0.001 - 0.1 μm
accumulation mode particles:	particles with diameter in between 0.1 - 1 μm
supermicron mode particles:	particles with diameter between 1-2.5 μm

Particle size distribution parameters

GMD geometric mean diameter (μm)

GSD geometric standard deviation

Aerosol concentration (xC), exposure (xE) and uptake (xU) metrics

$mC(D_p)$ particle size dependent particle mass concentration

$nC(D_p)$ particle size dependent particle number concentration

$lC(D_p)$ particle size dependent particle length concentration

$SC(D_p)$ particle size dependent particle surface area concentration

$VC(D_p)$ particle size dependent particle volume concentration

Traditional particle concentration metrics

$PM_{2.5}$ particle mass concentration for particles smaller than 2.5 μm in aerodynamic diameter

Infiltration model variables

F_{inf} infiltration factor (dimensionless)

$F_{inf}(D_p)$ particle size dependent infiltration factor (dimensionless)

Lung deposition parameters

DE(Dp) deposition efficiency (fraction) of particles of given size at defined regions of the respiratory tract:

Al alveolar region including respiratory bronchioles

ET1 anterior nasal region

ET2 extrathoracic region

BB bronchial region

bb bronchiolar region

1 Introduction

Particulate matter, especially $PM_{2.5}$, has been estimated to be responsible for a substantial burden of disease and loss of life years in various risk assessments based on epidemiological concentration response relationships. Similar concentration-response functions have been presented for coarse particles ($PM_{2.5-10}$), black carbon (BC) and particle number concentrations representing various fractions of inhalable particle size distributions. However, not all particles that are inhaled remain in the respiratory tract, substantial but not constant part of them is immediately exhaled. The probability of a particle being captured in the respiratory system is strongly dependent on the particle size.

It is reasonable to assume that only particles that are deposited in the respiratory tract actually affect a person's health. The chain of events from emission sources trough environmental and concentrations and exposure leading to doses is depicted in Figure 1, highlight also the concentration-response relationship observed in ambient epidemiology.

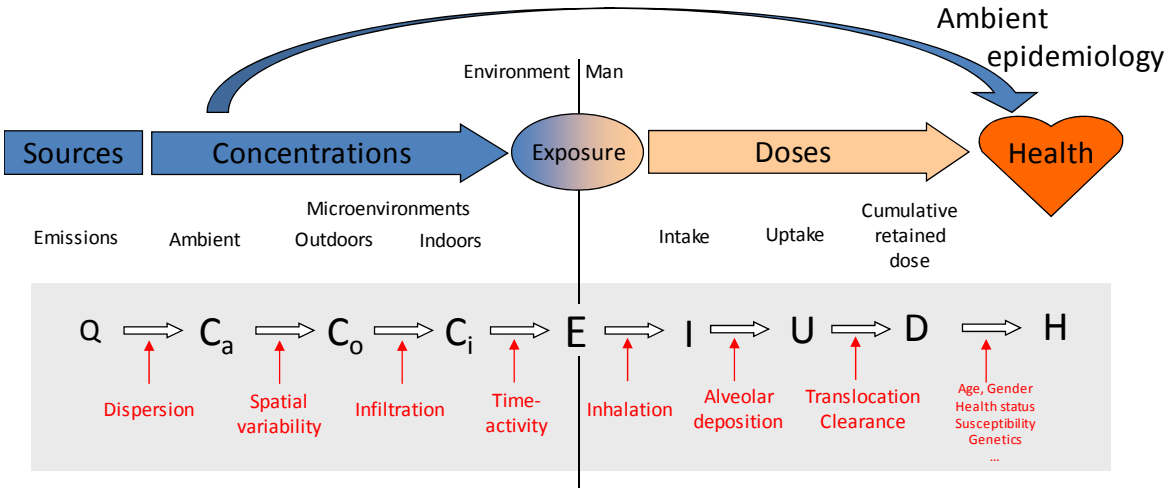


Figure 1. Ambient epidemiology links concentrations (*Ca*) with health, spanning over several nonlinear particle size dependent processes.

The first phase along the exposure chain in Figure 1, where the airborne particle size distribution is modified from the ambient one is infiltration of the aerosol indoors. As indoor environments represent a majority of the time-activity of urban populations, it becomes necessary to understand in more detail how the infiltration modifies exposures (Figure 2).

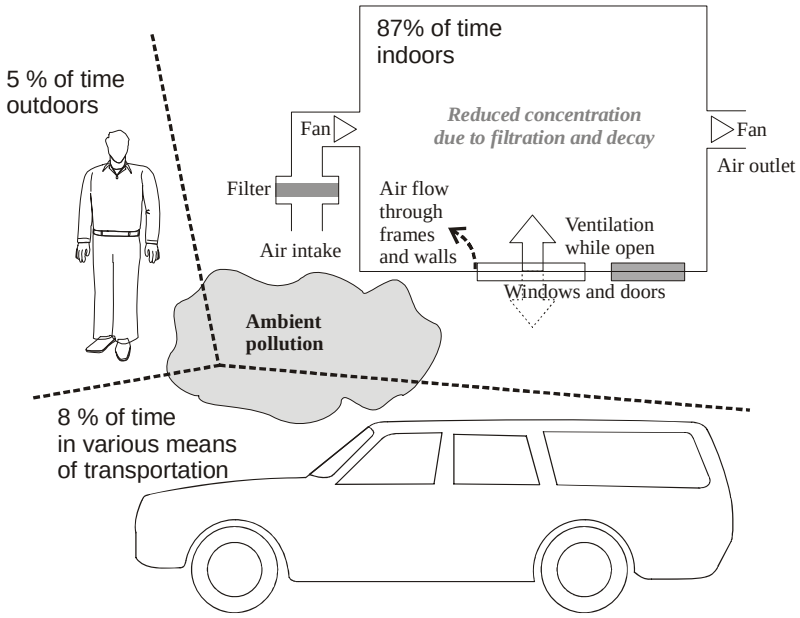


Figure 2. Schematic diagram of main microenvironments and their role for exposure to traffic generated particles for European adults (Hänninen et al., 2005).

Infiltration-caused particle size distribution changes affect especially ultrafine and coarse particles due to physical phenomena, such as diffusion and gravitational settling, that cause them to deposit on surfaces when moving through the building envelope and indoors. Infiltration efficiency has been found to be highest for the accumulation mode particles ($0.1 < D_p < 1$ μm) and to diminish efficiently for ultrafine ($D_p < 1$ μm) and supermicron particles ($D_p > 1$ μm) (Hänninen & Sorjamaa, 2011).

A particle size specific infiltration modelling approach was developed in TRANSPHORM WP2.5 and reported in Deliverable D2.5.1 (Hänninen & Sorjamaa, 2011). The infiltration component was then supplemented by an established human respiratory tract model (ICRP, 1994) in the next phase, reported in D2.5.2 (Sorjamaa & Hänninen, 2012), including also a time-activity approach for covering a number of microenvironments.

The aim of the current work is now to apply the previously developed model components (Figure 3) for the estimation of personal particle uptakes of epidemiological cohort members based on their residential addresses. The results will account for particle size distribution modification by infiltration and lung deposition and the model characterizes the doses in alternative aerosol metrics including number, length, surface area, volume and mass uptakes in different regions of the respiratory tract (extrathoragic, bronciolar and alveolar, with total of five subregions)

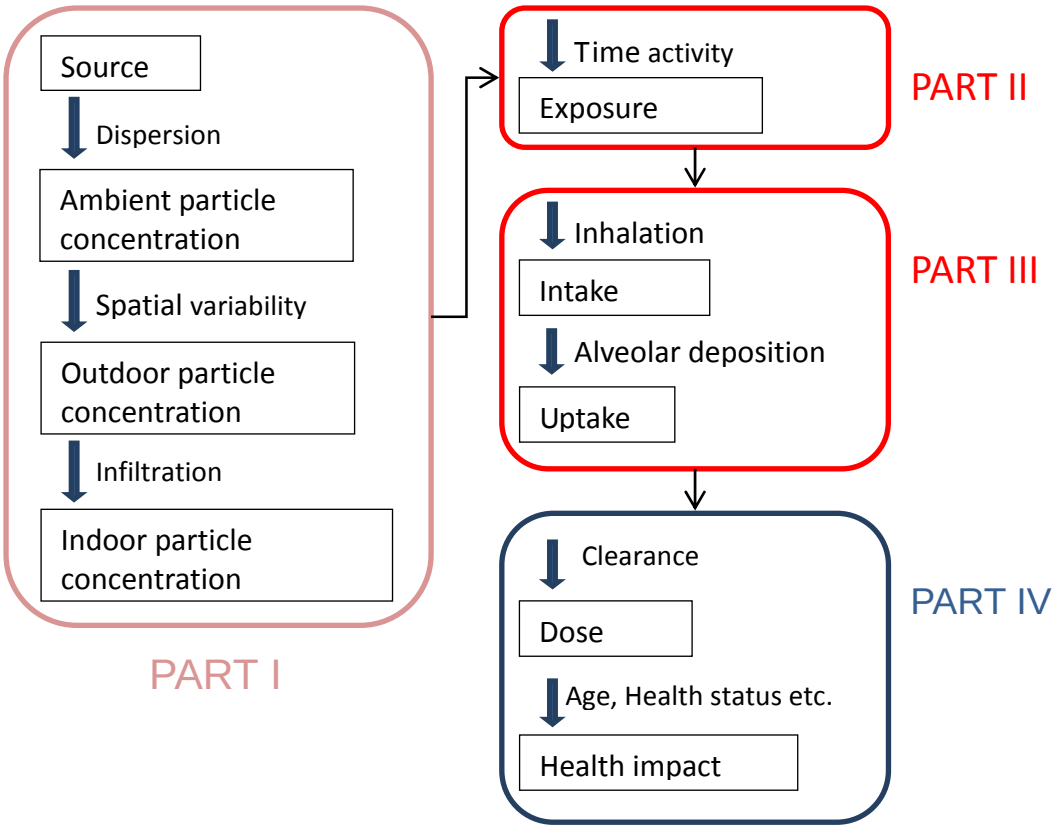


Figure 3. Diagram of the particle size specific exposure and dose model components. Parts I – III were implemented within the TRANSPORM project setting particle uptake as the current output variable.

2 ESCAPE Cohort studies for air pollution epidemiology

One of the main objectives of TRANSPHORM project is to provide additional exposure data for the ESCAPE cohorts. ESCAPE is relying on land use regression techniques in the native exposure estimation using residential locations of the cohort members. The main interest from the WP2.5 point of view is in particle size-specific non-linear exposure transformation processes that cannot be fully characterized by adjustment of regression coefficients.

ESCAPE incorporates a large number of European cohorts (Table 1) and the amount of data available on the occupational status, commuting habits etc. of the cohort members is limited and variable. The challenge in combining the microenvironmental approach with cohort studies is that the cohort studies have been designed for other purposes than air pollution epidemiology. Therefore there is very limited or no data on the daily time-activity, type of professional activities, location of the workplace, commuting habits, building types, and ventilation patterns. In such cases the missing parameters have to be replaced by population mean estimates, reducing the inter-individual contrasts in exposures that are needed for improving the epidemiological analysis. The only factor on which the improvement is directly possible is accounting for the particle size distribution differences at the residential locations by using aerosol properties from the dispersion model and then estimating the best estimate of long-term infiltration factor.

The FINRISK cohort from Finland was selected for the test case to demonstrate the use of the particle size dependent exposure and dose model in an epidemiological cohort setup.

Table 1. List of European cohorts participating the ESCAPE study. The TRANSPHORM target cities are highlighted in yellow. Additionally the two data sets used in the TRANSPHORM WP2.5 model development and evaluation are highlighted in green (ULTRA not being involved in ESCAPE and TRANSPHORM city London missing in ESCAPE).

	Epidemiologic Study Acronym (see WP descriptions for more details)	Study Area	Measurements	Contacts: Epidemiology	AQ data & modelling in TRANSPHORM
1	APREG	Györ HU	PM+NOx		
2	EPIC	Florence I	NOx		
3	EPIC, SIDRIA, ECRHS	Turin I	PM+NOx		
4	EPIC	Varese I	NOx		
5	ECRHS	Verona I	NOx		
6	ECRHS	Pavia I	NOx		
7	GASPII, SIDRIA	Rome I	PM+NOx		
8	ECRHS, INMA	Barcelona ES	PM+NOx		
9	EPIC, INMA, ECRHS	San Sebastian, Galdakao ES	NOx		
10	ECRHS	Huelva ES	NOx		
11	ECRHS, INMA	Oviedo ES	NOx		
12	REGICOR	Girona ES	PM+NOx		
13	EPIC	Athens GR	PM+NOx		
14	RHEA	Heraklion GR	PM+NOx		
15	EPIC, ECRHS, UK 1946 cohort	Oxford, Norfolk, Norwich, Ipswich UK	PM+NOx		
16	BIB	Bradford UK	NOx		
17	MAAS, UK 1946 cohort	Manchester UK	PM+NOx		
18	EPIC	Utrecht NL	PM+NOx		
19	EPIC, ABCD	Amsterdam NL			
20	EPIC	Doetinchem NL			
21	EPIC	Maastricht NL	PM+NOx		
22	PIAMA	Rotterdam NL			RIVM, UU
23	ECRHS	Antwerp BE			
24	EPIC	Heidelberg GE	NOx		
25	ECRHS	Erfurt GE	NOx		
26	SALIA, RECALL	Ruhr Area GE	PM+NOx		
27	LISA + GINI	Munich GE	PM+NOx		
28	KORA	Augsburg GE	<Pers.NC, ambient PM, gases>	Peters/HMGU	Cyrys, Pitz/HMGU
29	SAPALDIA	Lugano SU	PM+NOx		
30	SAPALDIA, ECRHS	Basel SU	NOx		
31	SAPALDIA	Geneva SU	NOx		
32	VHM&PP	Vorarlberg AU	NOx		
33	DCH, National Birth Cohort	Copenhagen DK	PM+NOx		
34	HUBRO, MOBA	Oslo NO	PM+NOx	NPHI	NILU
35	BAMSE, TWINGENE, 60 year olds	Stockholm SE	PM+NOx		
36	ECRHS, EPIC	Umea SE	NOx		
37	ECRHS, EPIC, GAZEL, EGEA	Paris FR	PM+NOx		
38	ECRHS, EGEA, GAZEL	Grenoble FR	NOx		
39	EPIC, EGEA, GAZEL	Marseille FR	NOx		
40	EPIC, EGEA, GAZEL	Lyon FR	NOx		
41	EDEN	Nancy, Poitiers FR	NOx *		
42	FINRISK	Helsinki, Turku FI	PM+NOx	Lanki, Yli-Tuomi/THL	Karppinen, Kukkonen/FMI
43	HAPIEE	Cracow PL	PM+NOx		
44	KANC	Kaunas LI	PM+NOx		
	ULTRA-2	Helsinki FI	PM2.5, UF	Lanki/THL	Lanki, Yli-Tuomi/THL
		London missing!			

2.1 FINRISK cohort study

One of the cohorts involved in the ESCAPE study is the FINRISK study in Finland, which was selected for detailed exposure and uptake modelling in TRANSPHORM. FINRISK is a national population survey on risk factors of chronic diseases, especially cardiovascular diseases, cancer, asthma, allergy and diabetes. Environmental factors are also considered, even though the study was not specifically designed to investigate air pollution effects. The study is coordinated by the Chronic Disease Epidemiology and Prevention Unit of National Institute for Health and Welfare (THL).

FINRISK is based on a stratified random sample drawn from the general population aged from 25 to 64 years from the population register. The samples were stratified according to the WHO MONICA protocol so that at least 250 subjects of each sex and 10-year age group were chosen in each area (4 age groups; 2 genders=8 groups x 250=2000 subjects per each of six geographical areas)

In ESCAPE, outdoor concentrations of air pollutants are modelled to home addresses of FINRISK cohort members in order to evaluate the health effects of long-term exposure to air pollutants. In ESCAPE, four cross-sectional population surveys (1992, 1997, 2002, 2007) and two study areas were used yielding to approximately 11200 subjects from the Helsinki metropolitan area and Turku city with its larger surroundings.

The Greater Helsinki Area consists of cities Helsinki (592,000 inhabitants) and Vantaa (202,000), while the Turku-Loimaa area includes the city of Turku (177,000), the town of Loimaa (17,000) and the surrounding municipalities. Both areas are characterized by minor altitude differences.

Population density varies from 20 inhabitants/km^2 in Loimaa to 30,000 inhabitants/km^2 in the most dense apartment blocks in Helsinki. Major sea ports are present in Helsinki and Turku. Two regional background monitoring sites were selected in countryside settings to capture regional differences resulting from long-range transport. Within ESCAPE, eighteen urban background and twenty street sites were sampled mainly in the residential areas and centres of these cities.

The FINRISK baseline study consisted of a clinical visit and a set of questionnaires. The clinical visit contained a physical examination with blood pressure and anthropometric measurements and a venous blood specimen was taken to determine e.g. serum total cholesterol, high density lipoprotein (HDL) cholesterol, triglyserides, gammaglutamyltransferase (GGT) and C-reactive protein (CRP).

The applied questionnaires covered socio-economic status, health status and weight, physical activity, smoking and consumption of alcohol, and nutrition.

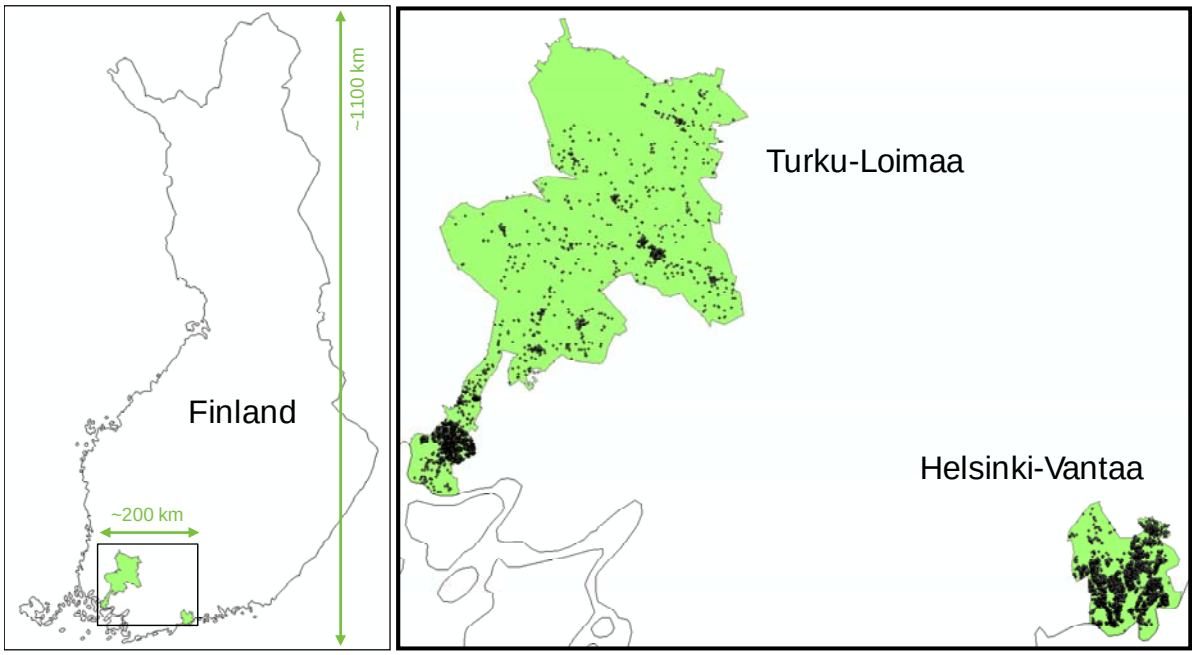


Figure 4. The Helsinki-Vantaa and Turku-Loimaa regions were included in the ESCAPE air pollution study. The detailed map on the right shows the locations of the residences of the cohort members at the time of the clinical visit.

The cohorts were followed up at 2009 (up to 16 years) for fatal and nonfatal coronary and stroke events and total mortality. Data was collected from Specific myocardial infarction and stroke registers, The National Hospital Discharge Register, and National Causes of Death -registry. The National Social Security Institute's drug reimbursement records to identify subjects who have developed diabetes or hypertension during the follow-up period were also included.

Health endpoints covered included inflammation, hypertension and blood pressure, incidence of coronary and cerebrovascular disease, gene-environment interactions, biomarkers, gene-environment interactions and other environmental exposures, and total mortality.

2.2 Land-use regression based ESCAPE exposure assessment

In the ESCAPE study the assessment of the long-term exposures is based on land use regression (LUR) modelling. An overview of the approach is given here as a background for understanding the differences with the current particle size dependent modelling approach.

The land use data from Helsinki area includes digital road data, local DigiRoad –road network with linked traffic intensities, population and household density, land use categories and heating system

of the buildings. Proximity to roads, traffic intensity and traffic load and road length in multiple buffers around the residential locations of the cohort members were used as predictor variables together with population density, ports and urban green areas and forests.

Targeted measurements were conducted for development and calibration of the land use regression models including NO_2, NO_x, $PM_{2.5}$, PM_{10}, $PM_{2.5abs}$ at 20 sites (12 in Helsinki-Vantaa and 8 in Turku-Loimaa) and additionally NO_x only was measured at 20 more sites (Figure 5). Additionally, one reference site was used.

At each site, measurements were conducted for three periods of two weeks in the cold season, the warm season and an intermediate temperature season. At the reference site, PM and NOx were measured using the same instruments continuously in two-week periods during a complete year. The data from the reference site was used to adjust the discontinuous site measurements to the true long-term average for the observation period (Jan 2010-Jan 2011 in Finland). At each time five sites + reference site were monitored simultaneously. At each site the sampling pump was on for 15 min during each two hours resulting to 42-hour sample in two weeks.

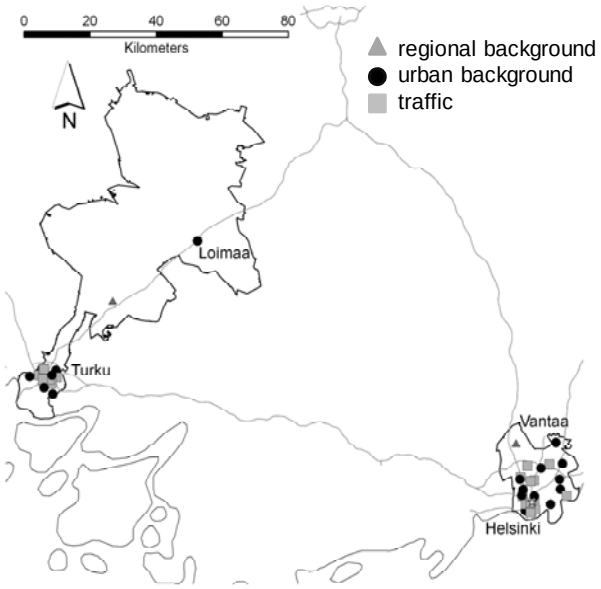


Figure 5. Locations of the ESCAPE sampling for land use regression modelling.

3 Methods for modelling particle size-specific lung deposition

The current work estimates the annual particle size specific particle uptakes for year 2010 of the FINRISK cohort members living in the Helsinki-Vantaa area. The overall modelling approach is depicted in Figure 6. There were 5700 cohort members with necessary data available.

Ambient $PM_{2.5}$ concentrations were estimated for the residential addresses by combining air quality model results calculated by Finnish Meteorological Institute (FMI) for exhaust and suspension particles and regional background measurements from Luukki monitoring station, a rural background station in Espoo, managed by the Helsinki Region Environmental Services Authority (HSY). These outdoor $PM_{2.5}$ concentrations at the residential locations were then transformed into semi-continuous particle size distributions for infiltration and lung deposition calculations by fitting to observed data using 68 bins. The previously developed infiltration model (Hänninen & Sorjamaa, 2011) was then applied to estimate the corresponding indoor concentration size distributions. Finally, a size dependent human respiratory tract model (ICRP, 1994) was then used to estimate the lung deposition in five regions.

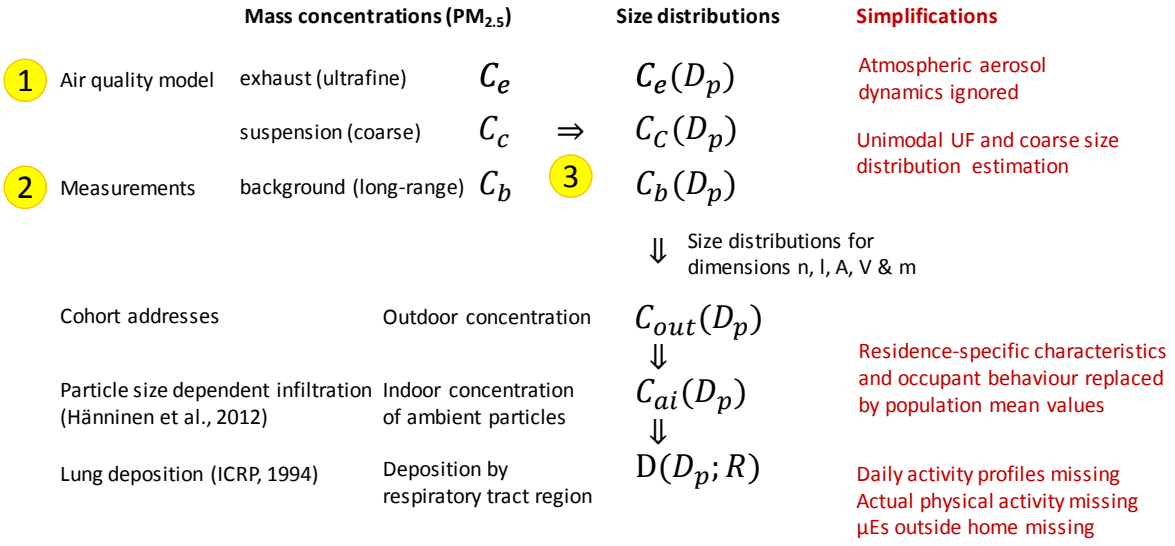


Figure 6. Overview diagram of the dose modelling.

In the current work the uptakes were estimated assuming staying indoors at home for the whole year due to lacking time-activity data. Actually 3658 of the included subjects had reported being employed, but no additional data on the locations of their employment was available. The remaining 2048 subjects were retired, homemakers/housewifes or unemployed and for them it can be assumed that the uptakes are slightly more representative. The microenvironmental methods based on time-activity data, presented in TRANSPHORM report D2.5.2 (Sorjamaa & Hänninen, 2012), were not applied for this cohort.

For the purpose of characterizing the model the variables and attributes presented in Table 2 were developed. Variables describe particular inputs used to calculate the estimates for each cohort member while the attributes are characteristics of each exposure and dose result; i.e. each result can be characterized by using all the alternative attribute dimensions.

Table 2. Variables and attributes used in the model. Variables represent inputs to the model. Attributes are dimensions into which each intermediate (concentration, exposure) and final result (human respiratory tract deposition) can be disaggregated. Internally the particle size distribution is presented by 68 bins.

Variables		**Attributes**	
Sources	Exhaust	Particle size fraction	Ultrafine (UF, <0.1µm)
	Suspension		Accumulation mode (0.1-1µm)
	Regional background		Coarse 1-2.5 µm
			Total (≤2.5 µm)
Buildings	Location (-> outdoor pollution)	Source fraction	Exhaust
	Particle size specific infiltration		Suspension
			Regional background
			Total
Person	Age, gender	Aerosol metric	Number (n)
	Time-activity (µEs)		Length (l)
	Physical activity		Surface area (S)
			Volume (V)
			Mass (m)

3.1 Air quality modelling for $PM_{2.5}$

Air quality modelling for year 2010 meteorology and emissions was conducted by FMI for the residential address locations of the cohort members. The model is described in more detail in the cited papers with a short summary of the approach given below.

The traffic flows where modelled for the street network of the Helsinki Metropolitan Area using the EMME/2 interactive transportation planning package. The model generates traffic demand on the basis of given scenarios, and allocates the activity over the links (i.e. segments of road or street) of this network, according to predetermined set of rules and individual link characteristics (Laurikko et al., 2003). The exhaust emissions are computed for each link using average speed-dependent functions, determined separately for each vehicle category. Vehicle categories for 14 different vehicle types are passenger cars, divided to petrol cars with or without a catalytic converter, and diesel-fuelled vehicles, as well as busses and heavy duty vehicles. The division of the vehicles within the passenger car category is based on the registration statistics. Suspension emissions were estimated using the methods presented by Kauhaniemi et al. (2011) using actual meteorology for 2010.

The dispersion modelling is based on the combined application of the Urban Dispersion Modelling system (UDM-FMI) and the road network dispersion model (CAR-FMI), developed at the Finnish Meteorological Institute (FMI) (Karppinen et al., 2000; Kousa et al., 2001; Härkönen, 2002). A main limitation of such so-called second generation Gaussian dispersion models is that they do not allow for the detailed structure of buildings and obstacles. Approximately 5000 road and street links were included in the computations. The model uses meteorological parameters from the FMI database and computes ambient air pollution concentrations for each hour over each year.

The air quality results were calculated as monthly and annual averages. Monthly averages would be useful in assessing the seasonal trends and changes in the impacts of ventilation behaviour and building heating. However, in the current work only the annual averages are reported.

3.2 Forming particle size distributions

The outdoor particle concentrations were estimated as (i) exhaust, (ii) suspension and (iii) regional background components. Based on the general finding that exhaust and suspension particle size distributions tend to follow lognormal shapes (e.g. Kleeman *et al.*, 2000, Hussein *et al.*, 2005, 2008), the estimated mass concentrations were transformed for the purpose of the modelling particle size specific infiltration and lung deposition into parametric size distributions. According to Kleeman et al., (2000) Hussein et al. (2005, 2008) the geometric mean particle diameter and corresponding geometric standard deviation were set to 0.2 μm and 1.8 for the exhaust particle mode, and to 4 μm and 2, respectively, for the suspension particle mode. The total particle mass up to aerodynamic particle diameter of 2.5 μm was set to match the $PM_{2.5}$ mass concentrations obtained from the CAR-FMI model and the density of all the particles was assumed to be 1.5 g cm^{-3} (Hänninen et al., 2011ab, Sorjamaa et al., 2011).

Aerosol processes such as coagulation and condensation cause the particle diameter of the aerosol to shift towards larger sizes. It was assumed that the shifts in the size along the dispersion are limited and that the applied distribution shapes are valid for rough estimation of the order of magnitude of the particle size specific processes.

More detailed data on the shape of the background particle size distribution was available from the SMEAR II rural background station located in Hyytiälä, Finland. The monthly particle size distributions measured in Hyytiälä with DMPS and APS are shown in Figure 7 for particle number and in Figure 8 for mass concentrations. The mass distributions are clearly bimodal with one peak in the accumulation mode regime and another in the coarse fraction.

No extensive analysis on the relation of exhaust, suspension and background particle $PM_{2.5}$ mass concentrations or mass distributions is done for this Deliverable, but the monthly average $PM_{2.5}$ mass concentrations and examples of different mass distributions are shown and briefly described below.

Monthly $PM_{2.5}$ mass concentrations for exhaust, suspension and background (measured at Luukki station) particles are presents in Table 3. The values for exhaust and suspension particle concentrations are averages of concentrations at cohort locations. The traffic emitted mass concentrations at cohort locations are clearly smaller than the background concentrations. Thus also the background size distributions dominate over the exhaust and suspension particle mass

distributions. However, there are some exceptions as can be seen in Figure 9-Figure 12 for the ESCAPE monitoring locations.

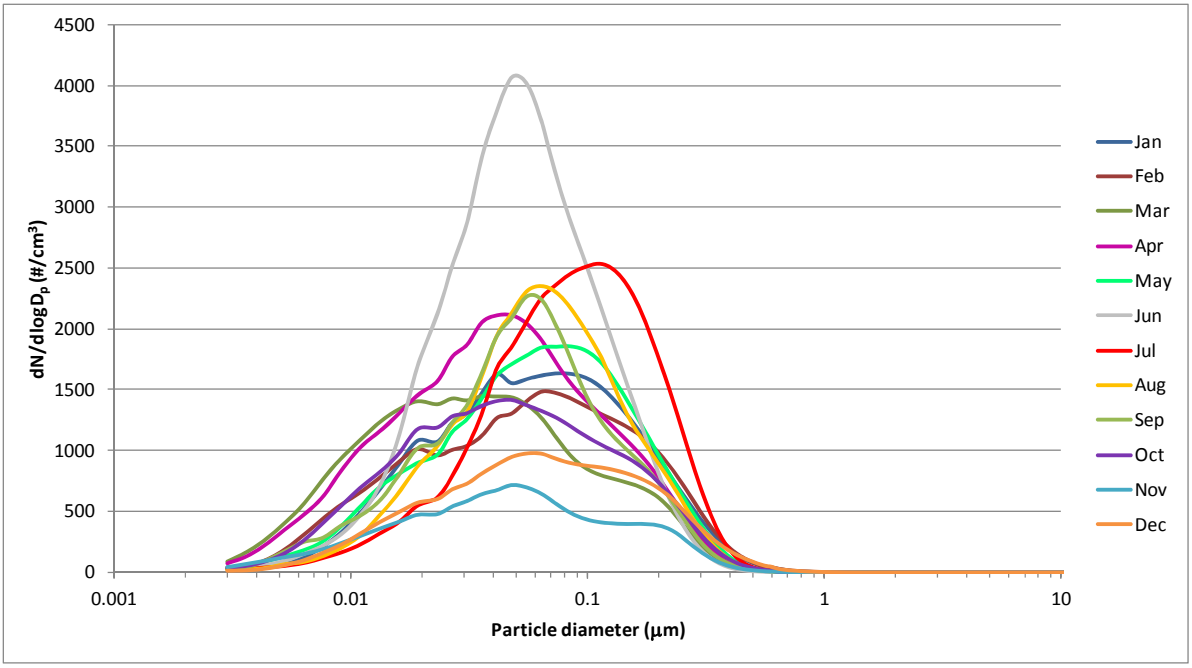


Figure 7. Monthly particle number size distributions from SMEAR II station located in Hyytiälä, Finland, for year 2010.

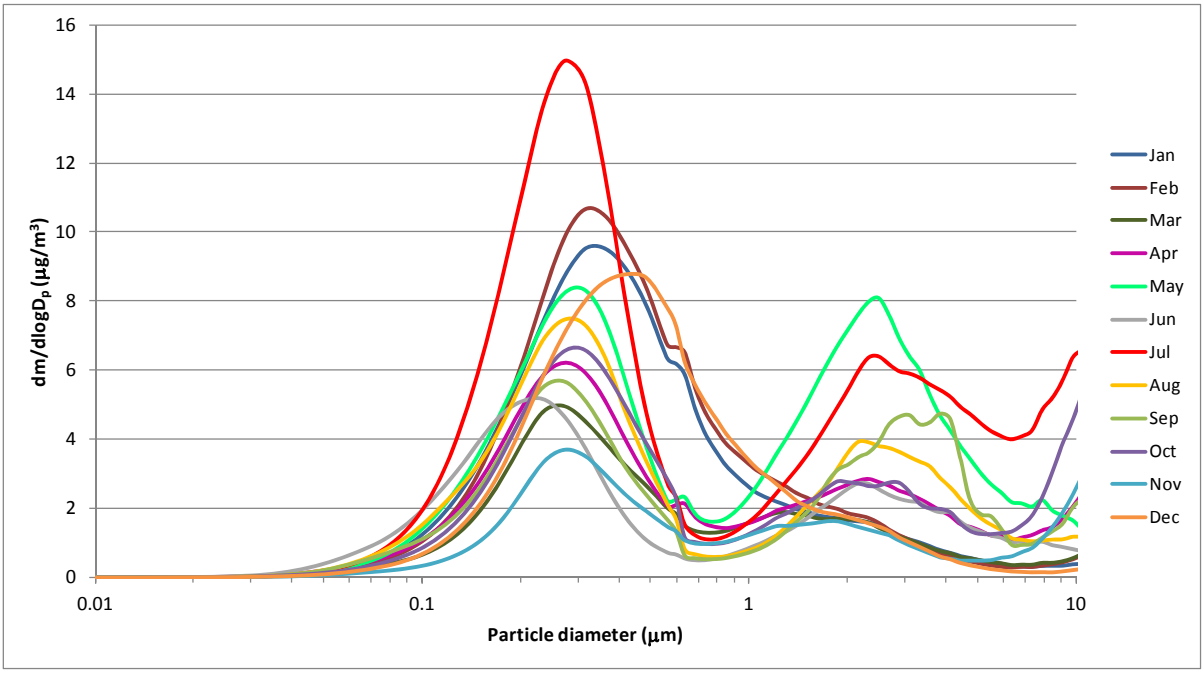


Figure 8. Monthly particle mass distributions from SMEAR II station located in Hyytiälä, Finland, for year 2010.

Table 3. Monthly and yearly averages for exhaust and suspension $PM_{2.5}$ mass concentrations as an average of all the cohort locations and for background station Luukki.

	Exhaust $PM_{2.5}$ ($\mu g/m^3$)	Suspension $PM_{2.5}$ ($\mu g/m^3$)	Background $PM_{2.5}$ ($\mu g/m^3$)
January	0.44	0.14	11.50
February	0.37	0.12	12.10
March	0.28	0.17	5.60
April	0.25	0.20	7.40
May	0.19	0.12	9.20
June	0.18	0.09	5.30
July	0.13	0.07	12.50
August	0.21	0.08	8.80
September	0.26	0.10	5.90
October	0.24	0.10	6.50
November	0.29	0.07	3.50
December	0.43	0.13	9.60
Year	0.27	0.12	8.20

Figure 9 shows average particle mass distributions in July 2010 for one of the FINRISK cohort sites. In most of the cases the situation is similar to this example: the background particles dominate the particle concentrations for all particle sizes. Even though the accumulation mode peak in Hyytiälä data may be enhanced by the particulate emissions from the surrounding forests, the difference to exhaust emissions is still huge. Yet there are cases where the exhaust and suspension particle concentrations exceed the background particle concentration in some of the particle size classes. Figure 10 shows an example of a situation where the mass concentration of the ultrafine particles caused by the tailpipe emissions exceed the corresponding background particle concentrations, while Figure 11 depicts a similar situation for suspension particles. High suspension particle concentration explained by the time of the year as March is usually the month when the snow cover melts in Finland and thus causes the road dust emissions to rise. There are also cohort sites with both exhaust (ultrafine fraction) and suspension particle concentrations exceeding the background concentrations as can be seen in Figure 12. The sum of exhaust, background and suspension particle mass distributions returns the total mass distribution. From now on the presented and discussed distributions are total mass distributions if not otherwise stated.

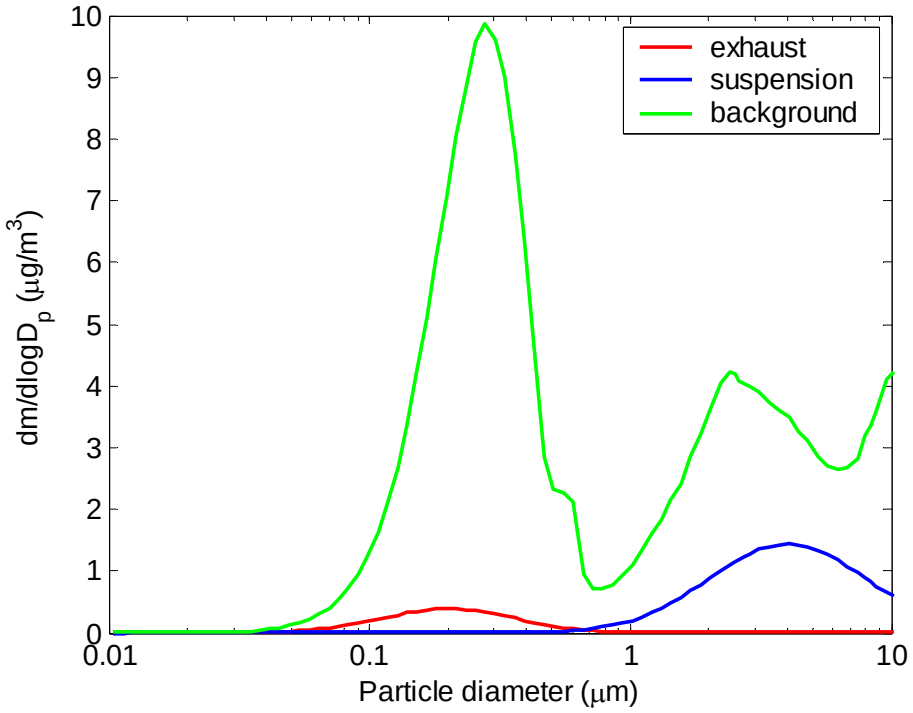


Figure 9. Monthly mass distributions for a FINRISK cohort site 7 in July 2010.

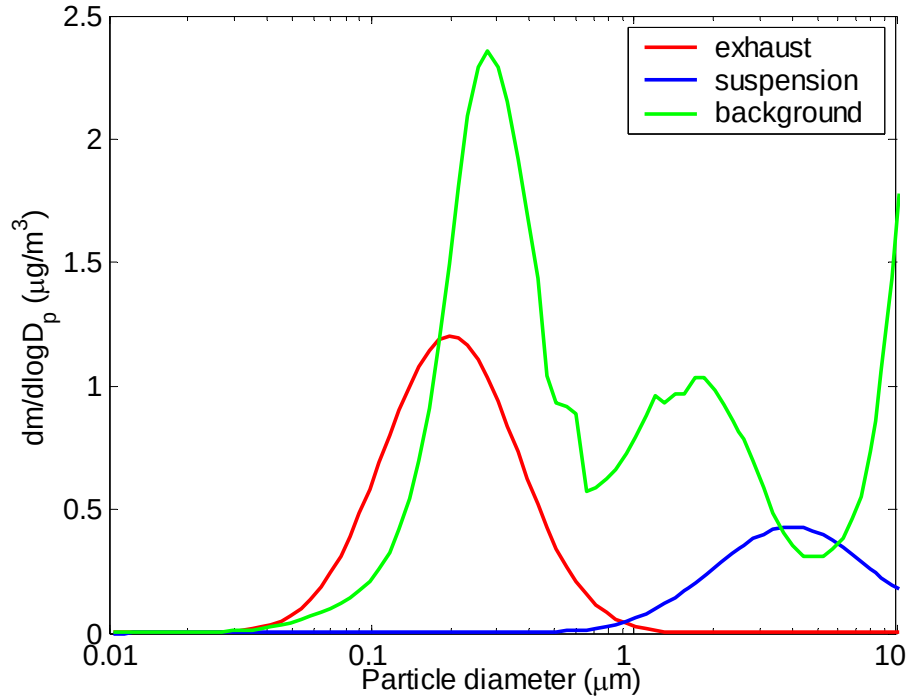


Figure 10. Estimated November mass distributions for a FINRISK cohort site 2.

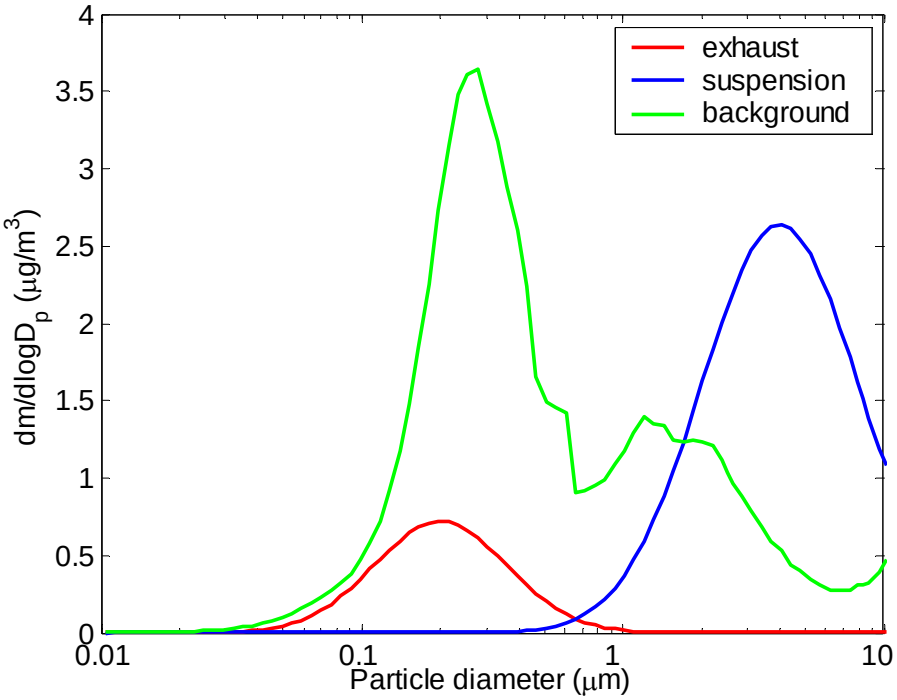


Figure 11. Estimated March mass distributions for a FINRISK cohort site 6.

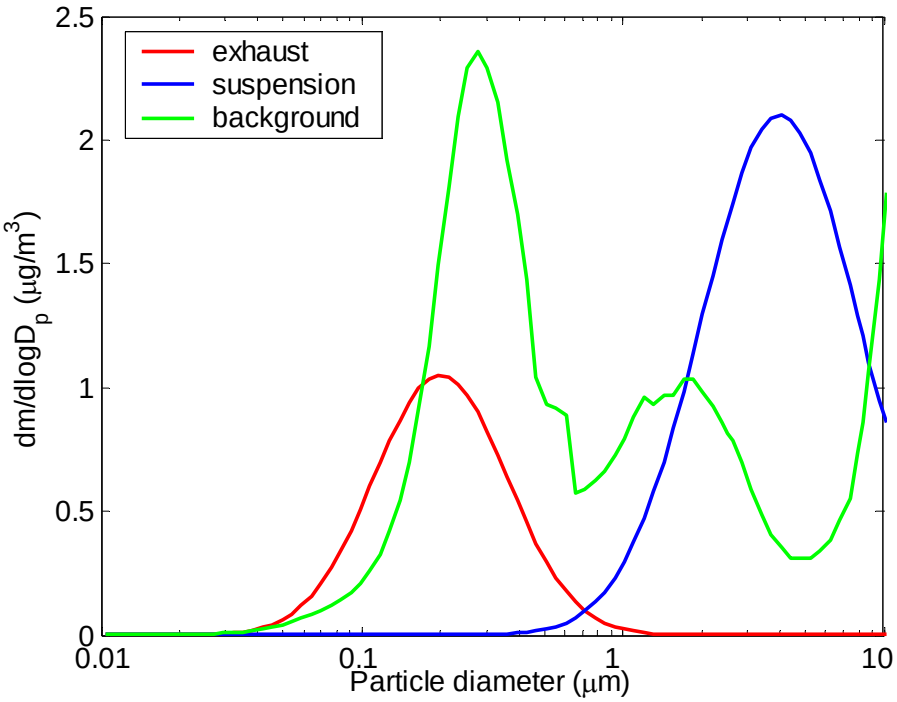


Figure 12. Estimated November mass distributions for a FINRISK cohort site 10.

3.2.1 Size-segregated infiltration factor $F_{inf}(D_p)$ and indoor concentrations

Infiltrated outdoor particle size distributions in cohort locations were calculated using the average of the size-segregated infiltration factors ($F_{inf}(D_p)$) estimated in detailed re-analysis of the Ultra data presented in TRANSPHROM Deliverable 2.5.1 (Hänninen & Sorjamaa, 2011). The Ultra mean infiltration factor as a function of particle diameter is shown in Figure 13.

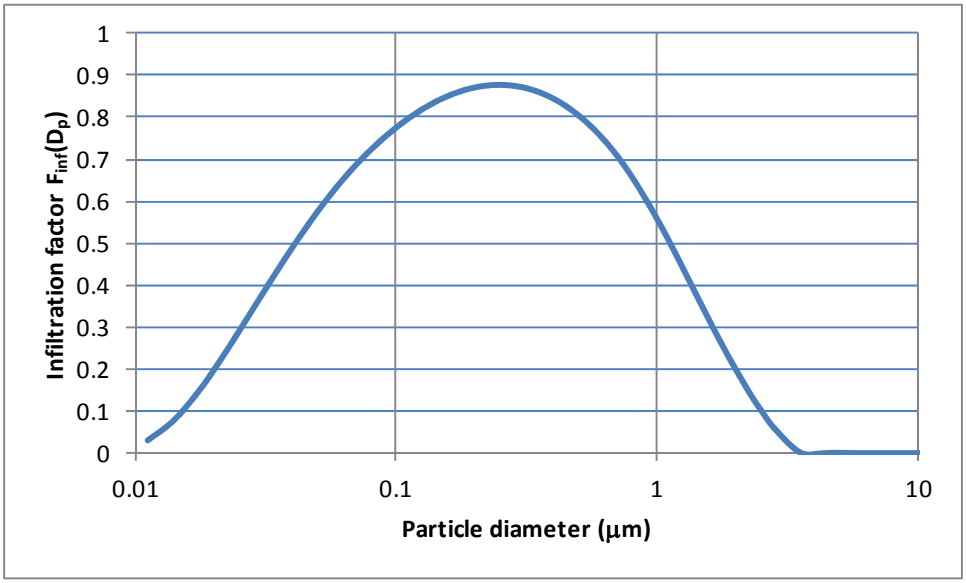


Figure 13. Average size-segregated ULTRA infiltration factor applied in the current study (Hänninen & Sorjamaa, 2011).

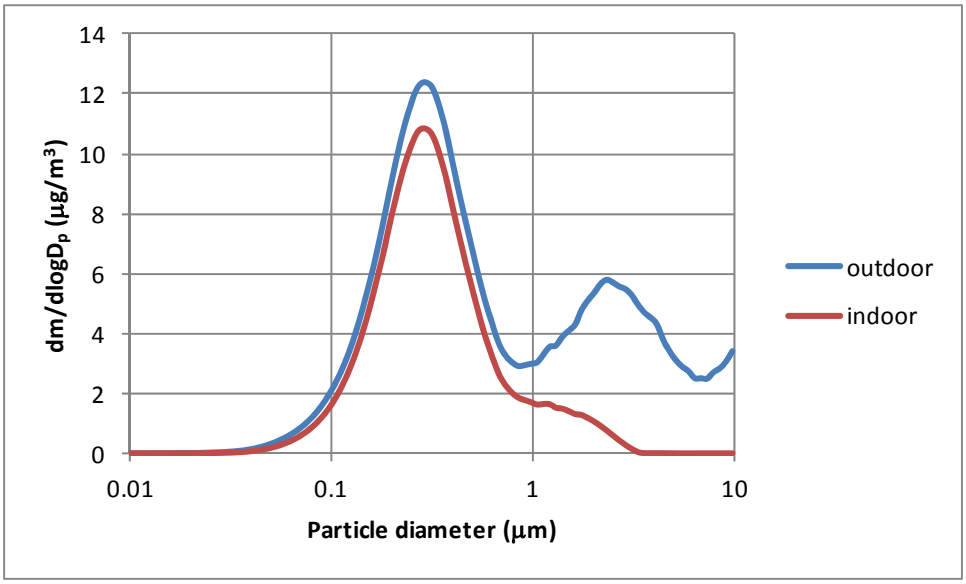


Figure 14. Estimated average outdoor and indoor particle mass distributions for the FINRISK cohort locations.

Figure 14 shows the estimated average particle mass distributions outside and inside the FINRISK cohort buildings. The decrease in the particle concentration due to infiltration in the fine particle fraction is not as enhanced as in the coarse fraction, but still clearly seen. As can be seen e.g. from Figure 9-Figure 12, most of the mass of the suspension particles is in the coarse particle fraction, which is efficiently removed in the infiltration process. Also most of the small exhaust particles are deposited, but the largest of the exhaust particles infiltrate efficiently.

3.2.2 Uptake in the human respiratory tract

Respiratory tract depositions were estimated using the International Commission for Radiological Protection model (ICRP, 1994). Overview of the probability of a particle to be deposited in different regions of the human respiratory tract is shown in Figure 15 as function of the particle diameter. In the alveolar region particles with diameter of few tens of nanometers have the highest deposition probability. Supermicron particles have a comparable high deposition probability in the extra-thoracic region.

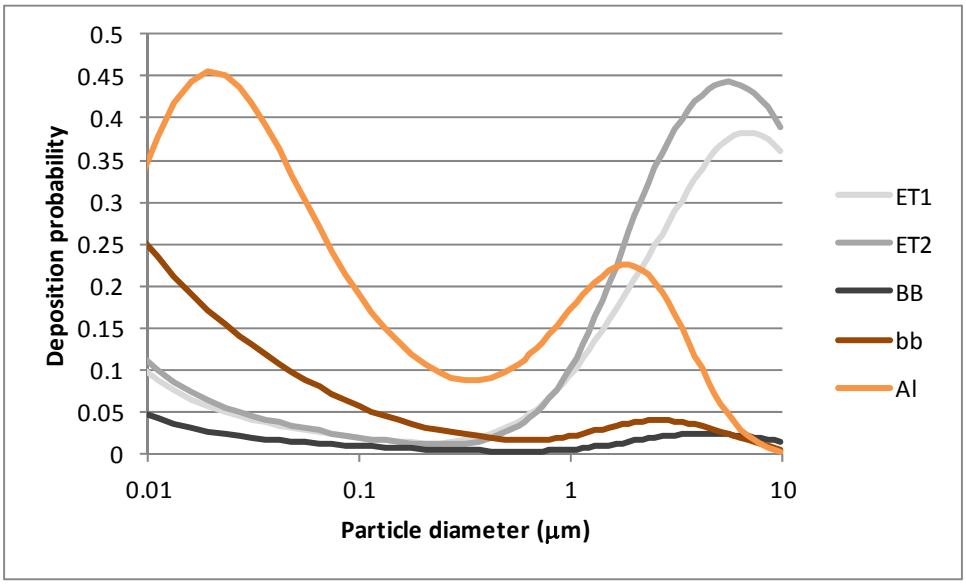


Figure 15. Particle deposition probability as a function of particle size in different lung regions for an adult male sitting and breathing through his nose. ET1 =anterior region ET2 = exthra thoracic region, BB = bronchial region, bb = bronchiolar region, and Al = alveolar region.

The ICRP (1994) model allows for adjusting the breathing rates according to various levels of physical activity ranging from sleep to heavy (Figure 16). In the current case no activity diaries were available for the FINRISK cohort members and therefore the physical activity was set to a constant value corresponding to sitting corresponding to breathing rate 0.39 and 0.54 m^3 h^{-1} for female and male subjects, respectively. A more detailed physical activity model could be easily developed for cases when time-activity data would be available.

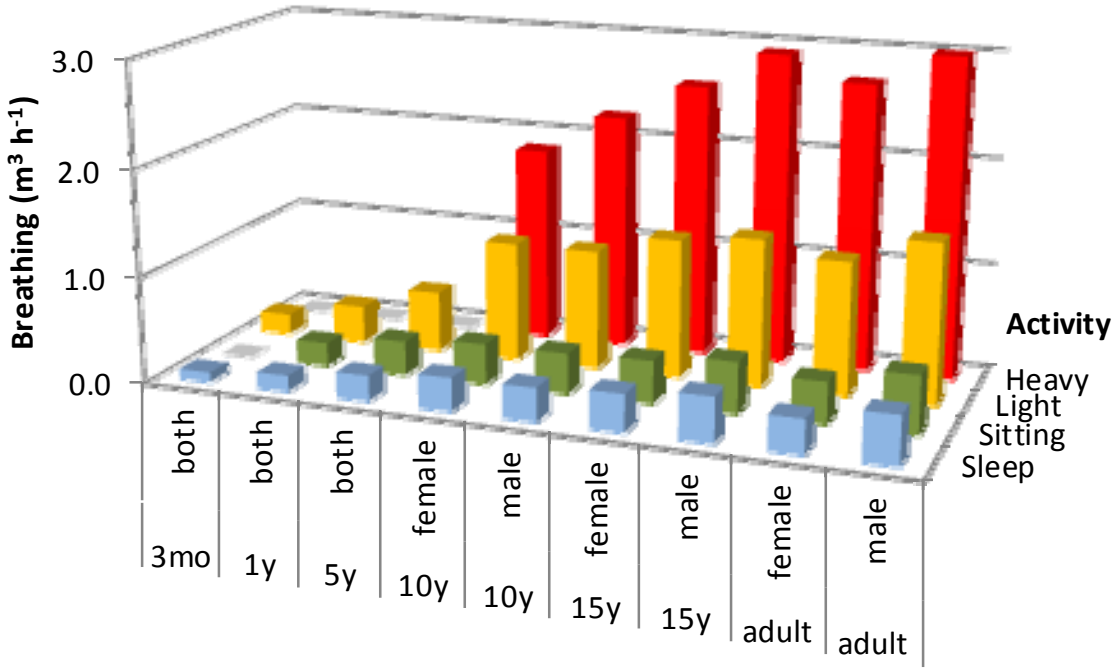


Figure 16. Relationships of age, gender and physical activity level with the breathing volume (ICRP, 1994).

4 Results on particle deposition in the human lungs

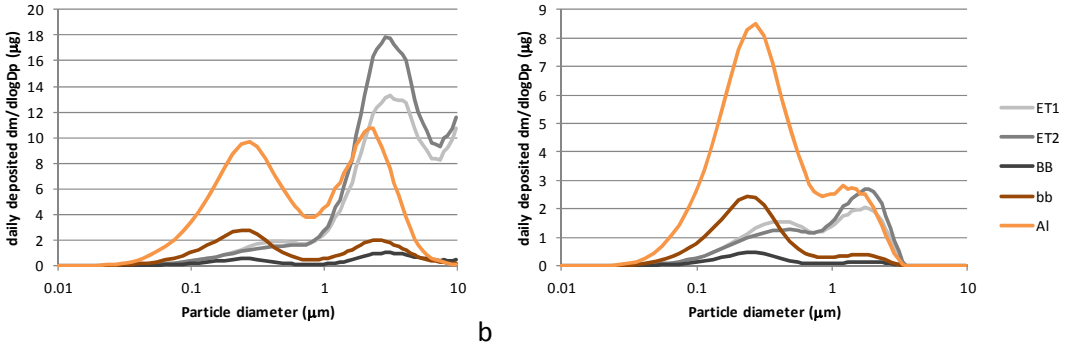
Annual respiratory tract uptake of particles were calculated for 2920 female and 2780 male subjects (total n=5700) FINRISK cohort members residing in Helsinki-Vantaa area in Finland. Even though 3658 subjects reported being employed, due to the lack of data on occupational and other activities outside the homes, only annual uptakes accounting for the time spent home indoors were estimated. Air quality model run by the Finnish Meteorological Institute was used to estimate the exhaust and suspension particle mass concentrations ($PM_{2.5}$) at the residential locations of the cohort members. Regional background concentration was accounted for by using $PM_{2.5}$ monitoring data from Luukki station. These three $PM_{2.5}$ particle fractions were transformed into estimated full size distribution and input into the infiltration and dose model. The full results of the model runs consists of annual exposures and uptakes of the cohort members divided into 68 particle size bins, expressed in mass, volume, particle surface area, particle length and particle number concentrations for each of the three particle fractions and the total PM2.5 concentration. Thus the raw database contains 3 x 68 x 5 = 1020 exposure values and corresponding uptakes for each five respiratory tract regions, totalling 5100 dose variables for each cohort member (Table 4). For presentation and analysis purposes the size distributions are re-aggregated into ultrafine (10-100 nm), accumulation mode (100 nm – 1 µm) and supermicron (1 – 2.5 µm) fractions.

Table 4. Dimensions of the exposure and uptake estimates.

	Raw categories	Regrouping
Sources	Exhaust, suspension, regional background	Total
Size categories	68 bins	UF, accumulation, coarse
Respiratory tract regions	ET1, ET2, BB, bb, AI	ET, BB, Abb
Metric	Number, length, surface, volume, mass	n/a

Particle size distribution is modified significantly in two phases of the exposure process: (i) when infiltrating into a confined space and (ii) when deposited onto the surfaces in the human respiratory tract. The effects of the former are highlighted in the comparison of a daily particle mass uptake estimated assuming (a) spending all time outdoors or (b) indoors (Figure 17). The

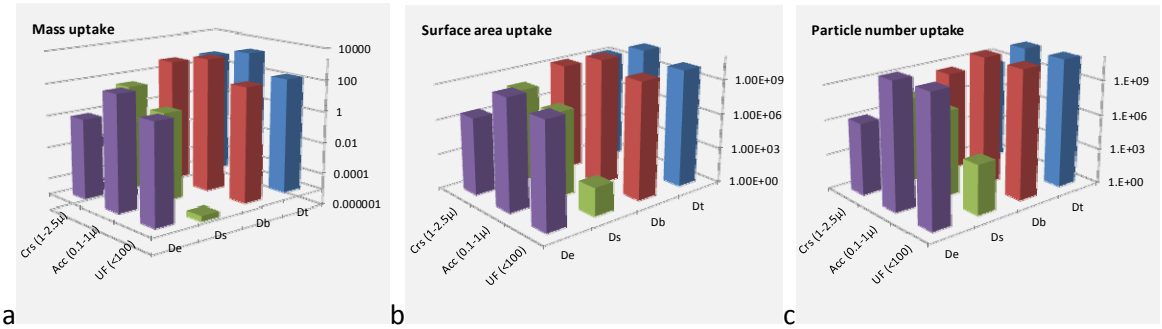
graphs highlight very well the impact of a building removing especially the coarse particles from the air, therefore reducing the corresponding uptakes while indoors.



a b

Figure 17. Continuous size distributions of daily particle mass uptake for (a)outdoor and (b) indoor aerosol (sitting adult male, nose breathing).

The average uptake estimates are characterized for alveolar uptake in Figure 18 in mass, surface area and particle number metrics. It can be seen that all total doses (blue columns) are dominated by regional background particles (red columns). In mass (graph a) the role of the exhaust (De) is slightly larger than suspension (Ds) and has significantly different size distribution. Surface area and particle number uptakes have significant contributions from the exhaust component, even though the regional background still remains higher.



a b c

Figure 18. Contribution of different sources (exhaust (De), suspension (Ds), background (Db), total (Dt)) and ultrafine, accumulation mode and coarse particles to the annual average uptake (dose) using (a) mass (µg), (b) particle surface area (μm^2), and (c) particle number as measure.

Annual uptake of particle mass (µg) in different regions of the respiratory tract (Figure 19) peaks for accumulation mode particles in the alveolar region, which is roughly five times larger than any

other deposition fraction. Accumulation mode has also most significant contribution to the bronchiole (bb) uptake, commonly counted in the total alveolar component. It is notable that in contrast to often stated hypothesis, the supermicron fraction (1-2.5 µm) contributes more to the alveolar uptake mass than the ultrafines (<100 nm) in the current indoor-based settings.

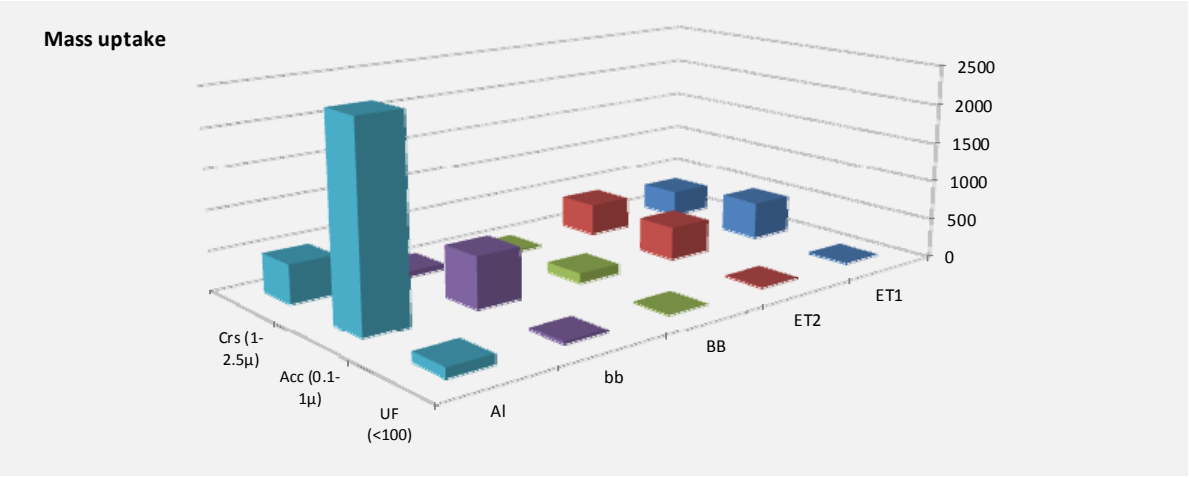


Figure 19. Annual uptake of particle mass (µg) in different regions of the respiratory tract.

5 Discussion and uncertainties

The model developed in PM SIZEx and TRANSPHORM WP2.5 is capable of handling full characterization of particle size distributions, visits to numerous microenvironments, variability in physical activity, different building characteristics and variation in ventilation rates and behaviour of the occupants modifying the infiltration of outdoor particles indoors. However, in the current application of the model for estimation of the particle uptakes of the 5700 members of the FINRISK cohort, lacking data on almost all these activities did not allow for utilizing the model to the full potential. Therefore the uptake estimates presented contain several simplifications that could be overcome in future studies by refining the data collection procedures. An overview of the simplifications was given in Figure 6. Some of them are discussed in more detail below.

Time-activity and spatial mobility. Personal time-activity in traffic and other microenvironments takes place at variable indoor and outdoor locations. Due to the lack of time-activity data on the cohort members the respiratory tract uptakes were estimated assuming that they spend the whole year in their homes. However, more than half of the cohort members reported to be employed,

being likely to spend at least part of their time at work. Moreover, even retired, housemaker and unemployed subjects regularly spend time in traffic and other places, which was not included in the model due to the lack of data. As the concentrations in traffic and at central locations where many of the visits outside from home are likely to head are expected to be higher, it seems probable that the uptakes calculated here are underestimated and moreover, in case of working subjects, they may be underestimated more than for the subjects spending more time at home.

Underestimation of variance. Due to the lack of individual data, mean estimates had to be used for the generation of the size distributions and infiltration factors that actually vary by building type and occupant ventilation behaviour. This is likely to lead to underestimation of variability between the subjects.

6 Conclusions

The current results demonstrated the usability of a semi-continous particle size distribution based model for exposure and dose estimation. The uptake (dose) characterization using alternative metrics for the particles in a size segregated manner, including mass, number, and particle surface area uptakes, produces a rich set of uptake characterization variables, analysis of which can give new insights to the composition of the human particle uptakes even accounting for the uncertainties inherent to the results concerning individual cohort members.

Most importantly, as highlighted in Figure 19, it can was seen that the total particle uptake is dominated by accumulation mode particles in the alveolar region. This is particularly interesting because the accumulation mode particles have lowest deposition efficiencies in the alveolar region (Figure 15). However, the result becomes understandable when looking at the size dependence of the particle infiltration from outdoor air indoors (Figure 13), where the accumulation mode size range represents the highest values. Overall, in the current model where the subjects were assumed to stay indoors all the time, the result is contrastingly high deposition of accumulation mode in the alveolar region. It will be very interesting to study the impacts of a more complex time activity patterns on the regional deposition profiles.

Overall, the work demonstrated that the model is ready for more detailed applications, but that in the case of most of the currently available cohort studies the amount of data on time activity and other behavioural factors is too limited for efficient utilization of the model capabilities.

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