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Aerosol particle number emissions and size distributions: Implementation in the GAINS model and initial results

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Abstract

Particulate matter affects our health and climate. In addition to well based knowledge on the adverse health effects related to particle mass concentrations, there is increasing evidence showing that the number concentrations of ultra-fine aerosol particles, with diameters below 0.1 µm, have negative health impacts, which are significantly different from those caused by larger particles with sizes over 1 µm. Particles with diameters between 0.1 and 1µm can also be activated as cloud droplets; thereby, higher number concentrations can increase the cloud albedo and thus the proportion of solar radiation reflected back to space, causing a cooling aerosol climate effect. In addition to this indirect effect, aerosol particles affect Earth radiation budget directly by either scattering solar radiation (e.g. sulphate aerosol, cooling effect) or absorbing it (black carbon aerosol, warming effect).

Currently, European air quality legislation on particulate matter is mainly focussing on particle mass, although emission standards for particle numbers have been introduced for mobile sources. Mass concentration is dominated by particles larger than 0.1 µm, and it is not well associated with number concentration, due to the often different formation mechanisms of ultra-fine and larger particles. For combustion sources, some emission control technologies affect mainly large particle emissions, and may even increase emissions of ultra-fine particles. Hence, in order to comprehensively estimate health and climate effects of anthropogenic aerosol particles, it is necessary to quantify their emissions with both mass and number based metrics, including information on their size distribution.

This report describes the implementation of size segregated particle number emission calculations in the GAINS model. Results show that in 2010 in Europe more than 60 % of particle number emissions emerge from road transport, even though their share in total PM1 mass emissions (i.e., the mass of emitted particles with diameters below 1 µm) is only 12 %. Particle number emissions from road transport are expected to decrease rapidly in the future due to further tightening of exhaust emission legislation (EURO-standards). Due to the envisaged more pronounced particle number emission reduction in the road transport sector compared to the currently second and third largest source sectors, shipping and combustion of fuel wood and coal in the residential sector, emissions from the latter two sectors are anticipated to exceed road transport emissions by 2025. Estimated shares in total European emissions in 2025 range, depending on the applied future scenario, from 35 to 41 % for shipping, from 26 to 29 % for residential combustion and from 17 to 21 % for road transport. The presented initial results are, however, subject to significant uncertainties, primarily due to limited measurement data for several emission sources.

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Aerosol particle number emissions and size distributions: Implementation in the GAINS model and initial results

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1 Introduction

Current air quality legislation in the European Union concerning aerosol particles is focussing on the total mass of particles with diameters (d_p) below 2.5 µm (PM2.5) or below 10 µm (PM10), with the exception of source-specific emission limit values for solid particle numbers from new vehicles. However, there is increasing evidence that adverse health effects of aerosols are also partly associated with the number concentration of ultrafine particles (UFP) with d_p < 0.1 µm (WHO, 2013, more details e.g. in Samet et al., 2009 and references 13-26 therein). These particles can enter deep into human lungs and further to the cardiovascular system, and affect the central nervous system. Thereby, health effects of UFP differ from those of particles in micrometre size scales. On the other hand, climate effects of aerosol particles are strongly dependent on the number concentrations of particles with d_p > 0.1 µm. All aerosol particles have direct climate effects: they either scatter solar radiation thus cooling the climate (e.g. sulphur aerosol) or absorb solar radiation (e.g. black carbon aerosol) which warms the atmosphere. However, particles with approximately d_p > 0.1 µm also form cloud droplets and thus cool the climate (indirect aerosol climate effects) by increasing the share of solar radiation reflected back to space (Forster et al., 2007). Whereas the anthropogenic direct climate effects of aerosols are estimated from slightly negative to slightly positive, it is agreed that the indirect cloud forming effects turn the total aerosol forcing (at least most likely) to negative, i.e. cooling (Forster et al., 2007, Murphy et al., 2009, Bond et al., 2013).

The particle number concentration is not directly related to PM2.5 or PM10, because mass concentrations are dominated by particles with d_p > 0.1 µm and number concentrations by particles with d_p < 0.1 µm. Sources of particles in different size ranges are often different: particles larger than roughly 0.3 µm are directly emitted to the atmosphere, whereas a significant share of the smaller particles is formed from vapour molecules during the nucleation process, e.g., in exhaust plumes or in the atmosphere (secondary particles). Also number emissions of particles > 0.1 µm differ significantly from particle mass emissions, because particles with diameters close to or over 1 µm have a strong effect on mass emissions, but in numbers their emissions are far less significant.

Number emissions and concentrations of aerosol particles are dominated by UFP. Particle number emissions differ from mass emissions not only because of different origins of particles, but also because the concentrations of UFP are partly controlled by larger particles. Basically in all combustion processes of organic matter that dominate anthropogenic emissions, potential nucleating vapours are formed. When the trace gas exits, e.g., the tailpipe or chimney, the decline in trace gas temperature decreases the volatility of these vapours, allowing for their condensation and nucleation. At this stage the aerosol population entering the atmosphere is very concentrated, and the larger particles form an efficient condensation/coagulation sink for nucleating vapours as well as for the freshly nucleated particles (Kulmala et al., 2001). Thus, the decrease in the emissions of larger particles *i*) leads to enhanced formation of UFP, since more vapours are available for nucleation, and *ii*) extends the lifetime of UFP by decreasing the rate of their scavenging to the larger particles.

In addition to the total number of particles, mainly determined by UFP, also the size of particles has prominent impact on the effects of particles. Particles with diameters of some tens of nanometres enter efficiently deep into lungs and are transported from there to the cardiovascular system, whereas most of the particles with diameters of some hundreds of nanometres are exhaled (Politis et al., 2008). Particles in the micrometre size range are mostly deposited to the nasal and larynx areas. On the other hand, particles with diameters close to or over 0.1 µm (100 nm) have more impact on climate than the smaller ones due to their cloud forming potential. In addition to the health and climate effects, particle size is important for the evolution of the aerosol particle population due to the size dependence of their coagulation scavenging, and thus of their lifetime. Hence, the knowledge of the number size distribution of aerosol particles is an essential input parameter for climate and air quality models: in absence of data on size distributions, several models assume one for distributing the particle number to different sizes.

Within the last year, we have introduced the calculation of particle number emissions into the GAINS model. Our analysis is based on the results of the EU FP6 EUCAARI project (Kulmala et al., 2011), which provided a first European size-resolved particle number emission inventory (H. Denier van der Gon et al., 2009, H. Denier van der Gon et al., 2010). Despite the limited information on the number and size distributions of anthropogenic emissions, we demonstrate that current knowledge allows a reasonable analysis of particle number emissions and their anticipated changes due to emission control strategies that focus on PM-mass.

2 Methods

2.1 GAINS structure and scenarios

The GAINS (Greenhouse gas – Air pollutant Interactions and Synergies) model (Markus Amann et al., 2011) is an integrated assessment model that brings together information on the sources and impacts of air pollutant and greenhouse gas emissions

and their interactions. GAINS combines data on economic development, the structure, control potential and costs of emission sources, the formation and dispersion of pollutants in the atmosphere and an assessment of environmental impacts of pollution.

GAINS addresses air pollution impacts on human health from fine particulate matter and ground-level ozone, vegetation damage caused by ground-level ozone, the acidification of terrestrial and aquatic ecosystems and excess nitrogen deposition of soils, in addition to the mitigation of greenhouse gas emissions.

GAINS describes the inter-relations between these multiple effects and the range of pollutants (SO_2, NO_x, PM, NMVOC, NH_3, CO_2, CH_4, N_2O, F-gases) that contribute to these effects at the European scale. GAINS assesses, for each of the 43 countries in Europe, more than 1000 measures to control the emissions to the atmosphere. It computes the atmospheric dispersion of pollutants and analyses costs and environmental impacts of pollution control strategies. In its optimization mode, GAINS identifies the least-cost balance of emission control measures across pollutants, economic sectors and countries that meet user-specified air quality and climate targets.

In GAINS, emissions from different sources are calculated with three basic input parameters (Z. Klimont et al., 2002):

- volumes of yearly activities (*A*) in a given sector, corresponding to certain fuels (e.g., fuel wood used (burned) per year in domestic single house boilers),
- the shares (*X*) of abatement technologies applied to fuel consumption of the activity (e.g., improved boilers with accumulation tank, pellet boilers, boilers with electrostatic precipitator, etc.) such that ∑X=1,
- the emission factors (EF) for each sector-fuel-technology –combination (emissions per activity unit).

The yearly emissions *E* in region *i* are calculated as

$$E_i = \sum_{ijkm} E_{ijkm} = \sum_{ijkm} A_{ijkm} X_{ijkm} \mathrm{EF}_{ijkm} \,, \qquad (1)$$

where the indices *j* refer to source sector, *k* to fuel and *m* to abatement technology.

Within GAINS, future emissions are estimated for different scenarios of anthropogenic activities (e.g., energy use), for which shares *X* of different technology levels for all emission sources are assumed. This report employs estimates of historic and future emissions that have been produced for the analyses supporting the revision of the Thematic Strategy on Air Pollution of the European Commission. In particular, particle number emissions are calculated for scenarios that are presented in TSAP Report #10 (Markus Amann et al., 2013).

2.2 Emission factors for particle numbers

The determination of emission factors (EF_N) for particle number (PN) emissions and particle size distributions (PSD) is based on the European particle number emission

inventory developed by TNO (H. Denier van der Gon et al., 2009, H. Denier van der Gon et al., 2010) during the EUCAARI project (Kulmala et al., 2011). In that work, as well as here, the PSDs present the size segregation of the number emissions into size classes, i.e., the proportions P_i of the total number of emitted particles in each size sector *i*. Thus, the emission factor for a single size class *i* is written as

$$\mathrm{EF}_{\mathrm{N},i} = P_i \mathrm{EF}_{\mathrm{N}}, \quad (2)$$

and the sum over all P_i:s equals one. Values for the proportions P_i are calculated from modal presentations of PSDs, consisting of one to three lognormal modes.

In the TNO study, $\mathrm{EF_N}$:s were determined through two alternative ways. For some source sectors, especially for traffic and domestic combustion, both $\mathrm{EF_N}$:s and PSDs were determined from the literature directly. For other source sectors, $\mathrm{EF_N}$:s were based on PM1 mass emission factors ($\mathrm{EF_{PM1}}$), which were originally adapted from an earlier version of the GAINS model (M. Kulmala et al., 2011, H. Denier van der Gon et al., 2010). The latter group of $\mathrm{EF_N}$:s was formed by first determining particle mass and number size distributions and estimating the particle densities from the literature, then converting $\mathrm{EF_{PM1}}$ to (mass) emission factors of particles smaller than 300 nm ($\mathrm{EF_{PM0.3}}$) based on the mass size distributions, and finally resolving the $\mathrm{EF_N}$:s that yield the $\mathrm{EF_{PM0.3}}$:s by applying the particle number size distributions.

In our first analysis, we employ for many source sectors the emission factors and size distributions provided in the TNO study. However, for sources that are most important for particle numbers, such as road transport and wood combustion in the domestic sector, we developed improved emission factors and size distributions. The modifications to the TNO study are described below.

Firstly, we extend the PSDs in GAINS to cover sizes from electrical mobility diameter (d_M) of 3 nm up to aerodynamic diameter (d_A) of 1 µm, whereas the particle size range in the TNO study was from d_M =10 nm to d_A =300 nm. The size range was extended to larger sizes in order to allow for comparison between the emission factors for particle number and PM1 mass, the latter being determined as the total mass of particles with $d_A \leq 1$ µm. Additionally, even though the share of particles larger than 300 nm in all emitted particles is negligible, large particles are important in some source sectors. The extension towards smaller diameters was made to provide the whole particle size range for climate model calculations. These extensions of the particle size ranges required recalculation of the $\mathrm{EF_N}$:s for source sectors that were originally based on PM0.3 emission factors, with the formula

$$\mathrm{EF_N} = \frac{1}{\rho \sum_i P_i \frac{\pi}{6} d_i^3} \mathrm{EF_{PM1}}, \quad (3)$$

where ρ is the estimated density of the emitted particles, P_i is the proportion of particles in size class *i* out of the total number of emitted particles and d_i is the geometric mean diameter of the particles in size class *i*. The values for ρ and PSDs were taken from the TNO analysis, with the exception of the PSDs mentioned below.

Secondly, new PSDs were introduced for road transport sources with the highest activities (diesel heavy duty trucks and busses, both diesel and gasoline light duty

trucks and passenger cars), based on the EU FP7 project TRANSPHORM database (Vouitsis et al., 2013 and 2014). Also EF_N:s and PSDs for domestic wood combustion (including pellet burning and medium size district heating boilers) and for shipping emissions were updated (domestic sector: Gaegauf et al., 2001, Emma Hedberg et al., 2002, L. S. Johansson et al., 2004, C. Johansson et al., 2008, Kinsey et al., 2009, Lamberg et al., 2011, Bäfver et al., 2011, C. Boman et al., 2011**,** Pettersson et al., 2011**,** Chandrasekaran et al., 2011; shipping: Hobbs et al., 2000, Sinha et al., 2003, Petzold et al., 2008, Murphy et al., 2009, Moldanova et al., 2011, Diesch et al., 2013), as well as for two stroke vehicles in road transport (Ntziachristos et al., 2005, Etissa et al., 2008). New PSDs were introduced also for flaring in gas and oil industry (Canteenwalla et al., 2006) and for coke production (Weitkamp et al., 2005). The EF_N for tire wear, previously based on $EF_{PM0.3}$, was replaced with a direct PN emission factor (Dahl et al., 2006).

2.3 Particle number size distributions

Emission factors include the total number emission factor (EF_N) and the particle number size distribution (PSD) in 15 size classes (see Table 1).

Table 1. Particle diameter ranges of the size classes in nanometres (0.001 µm). Diameters are electrical mobility diameters, except for * which is aerodynamic diameter (see text below).

Size class number	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Min d_p	3	6	10	14	19	26	36	50	70	100	142	200	285	405	577
Max d_p	6	10	14	19	26	36	50	70	100	142	200	285	405	577	1000 *

PSDs are initially determined with a size distribution function f_N, which is set by one to three lognormal modes. The modal size distributions are converted to particle concentrations N_i in 15 size bins i, with $\sum N_i$=1. The class of the smallest particles has a minimum diameter of 3 nm and a maximum of 6 nm (electric mobility diameters, d_M), whereas the largest size class extends from d_M = 577 nm to 1 µm in aerodynamic diameter (d_A). The electrical mobility diameters are applied for all size class boundaries except 1 µm, because it is the diameter relevant to the PSD measurements typically made with Scanning/Differential Mobility Particle Sizers (SMPS/DMPS). Aerodynamic diameter is chosen for the upper diameter limit of the largest size class (1 µm) to allow comparison between the number emissions and PM_1 mass emissions, determined as the total mass of particles with $d_A \leq 1$ µm. For calculating the concentration of the largest size class, the upper diameter limit in d_M is calculated with

$$d_M = \frac{d_A}{\sqrt{\rho/1000}}, \quad (4)$$

where ρ is the estimated density of particles in units [kg m^{-3}]. Because the upper size limit of the largest size class is in d_A, concentrations N_{15} are not directly comparable (e.g., for particles with the highest estimated density, ρ = 3000 kg m^{-3}, the upper size limit equals the lower size limit d_M=577, resulting in N_{15}=0). For the total particle number this causes, however, only negligible inaccuracy, since particle number emissions are much higher in smaller size bins.

It is planned to implement EF_N for the total number emissions and $EF_{N>0.1}$ for the emissions of particles with d_p > 0.1 µm for routine calculations in the GAINS model. The latter emission factor is relevant for, e.g., climate model calculations addressing particles active in cloud formation processes (Dusek et al., 2006). Additionally, emissions in all 15 size classes are available upon request.

2.4 Examples of particle emission factors and size distributions

Emission factors applied for different technologies of heavy duty diesel trucks (MD HDT) and domestic wood (FWD) and hard coal (HC1) fuelled stoves are presented as examples in Table 2. The related particle size distributions multiplied with the emission factor EF_N are presented in Figure 1. For hard coal combustion the emission factors are PM1 mass based with identical PSDs, whereas for the other emissions presented in Figure 1 direct EF_N:s and individual PSDs for technologies are applied.

Table 2. Examples of particle number emission factors: heavy duty diesel truck emissions (MD HDT) for uncontrolled (NOC) and EURO I-VI vehicles, and domestic stove emissions for wood (FWD) and hard coal (HC1) fuelled old (NOC), new, improved (IMP), electrostatic precipitator equipped (ESP) and pellet burning (PELL) stoves.

Technology	**EF_N (10^{22} #/PJ)**
MD HDT NOC	6.85
MD HDT EURO I	10.52
MD HDT EURO II	12.88
MD HDT EUROIII	8.21
MD HDT EURO IV	2.02
MD HDT EURO V	0.92
MD HDT EURO VI	0.01
FWD STOVE NOC	3.20
FWD STOVE NEW/IMP/ESP	4.80
FWD STOVE PELL	5.64
FWD STOVE PELL+ESP	0.05
HC1 STOVE NOC	20.33
HC1 STOVE IMP	16.26
HC1 STOVE NEW	11.62

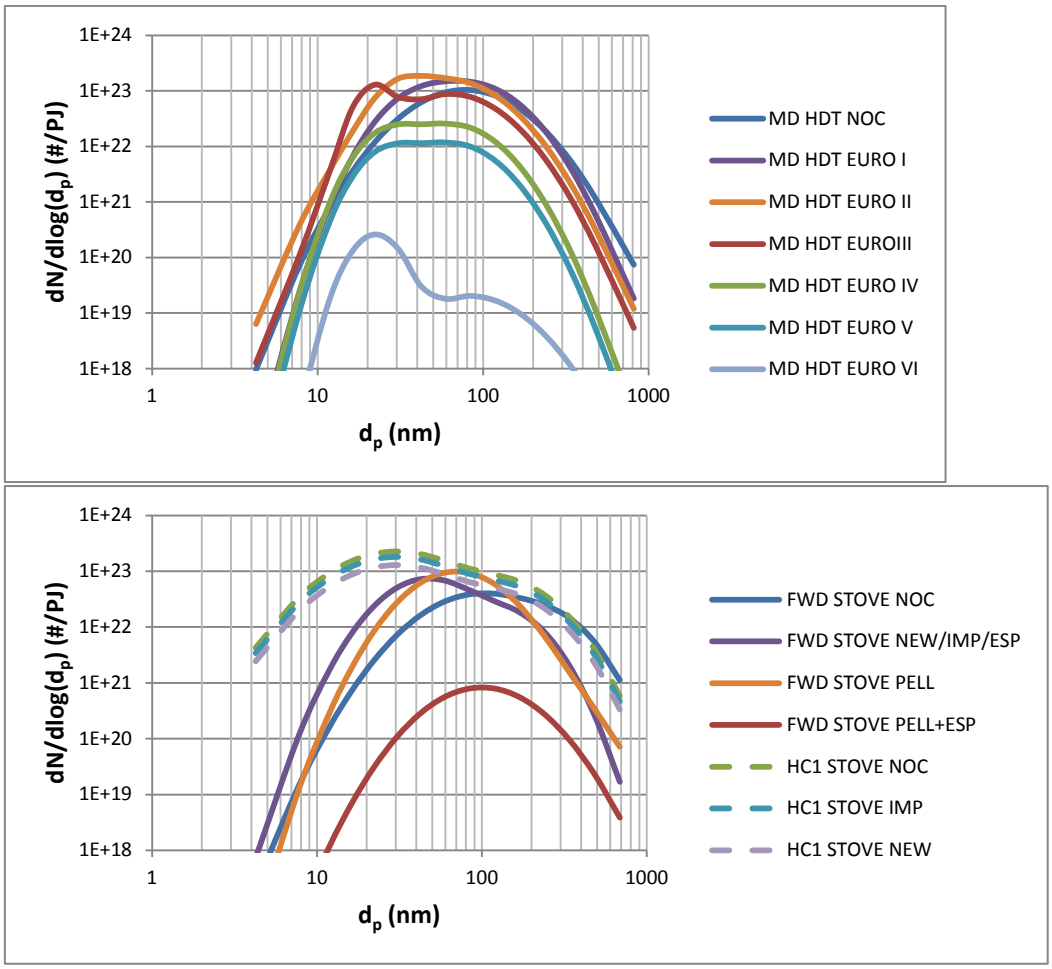

Figure 1. Examples of emission factor size distributions. Upper panel: heavy duty diesel truck emissions for uncontrolled (NOC) and EURO I-VI vehicles. Lower panel: domestic stove emissions for wood (FWD) and hard coal (HC1) fuelled old (NOC), new, improved (IMP), electrostatic precipitator equipped (ESP) and pellet burning (PELL) stoves.

3 Initial results

In this section, we present first results of the particle number emission calculations with the GAINS model. It is important to note that these are first estimates and are subject to significant uncertainties. The reasons behind the uncertainties are discussed in the Discussion section.

We compare particle number emissions with mass emissions of particles with diameters below 1 µm, because these emissions correspond to different metrics of the same particles. PM2.5 mass emissions are less correlated with the number emissions than PM1 emissions.

3.1 Comparison of number and mass emission factors

The inaccuracy of EF_{PM1} for estimating particle number emissions is illustrated in Figs. 2a-b. The maximum variability of EF_N from different sources with similar EF_{PM1} is roughly four orders of magnitude (depicted with an arrow in Fig. 2b). This means that from two different sources emitting the same mass of particles per activity unit, the other may have larger number emissions by a factor of 10,000. The difference arises, naturally, from the difference in sizes of the emitted particles. The relation between emission factors of particles larger than 0.1 µm ($EF_{N>0.1}$) and total number emission factor (EF_N) is depicted in Fig. 2c, showing that also these factors are not coupled.

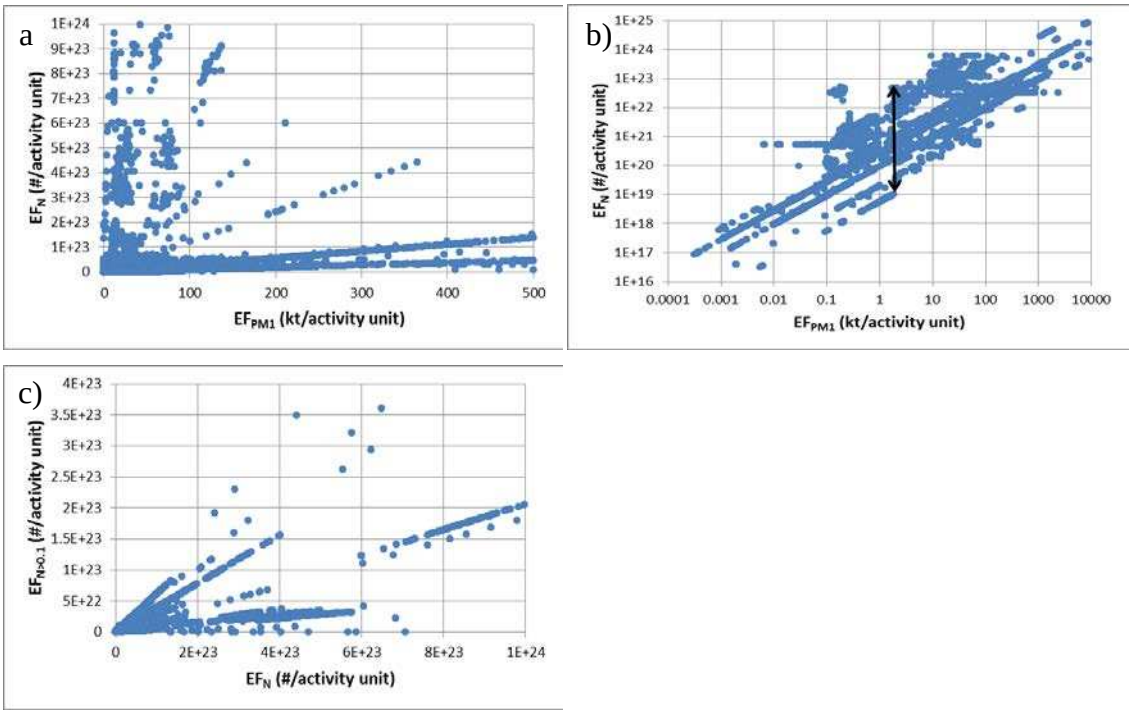

Figure 2. Particle number emission factors EF_N vs. particle mass emission factors EF_{PM1} in linear (a) and logarithmic (b) scale. The arrow in (b) shows the maximum variability of EF_N:s corresponding to similar EF_{PM1}:s. Panel c) illustrates the lack of a direct coupling between number emissions of particles with diameters above 0.1 µm ($EF_{N>0.1}$) and total particle number emissions. Panels (a) and (c) do not present the whole ranges of the emission factors.

3.2 Current and future particle number emissions in the EU28

Estimates of total particle number emissions (N_{tot}) and emissions of particles with d_p > 0.1 µm ($N_{>0.1}$) are presented in Table 3 for the 28 EU countries (EU28). The table includes estimates for the years 2010, 2020 and 2025, based on the assumption that all measures included in current legislation will be effectively implemented according to current schedule (the current legislation, CLE, scenario). For 2025, estimates are also provided for the central policy scenario A5 analysed in TSAP Report #10 and for the maximum feasible reductions (MTFR) scenario (Amann et al., 2013). Changes in

emissions relative to 2010 are presented as well, including the corresponding changes in PM1 mass emissions for comparison. Results for European non-EU countries are presented in Appendix A.

Particle number emissions are clearly dominated by the ultrafine particles (UFP, d_p<0.1 µm) in all the source sectors excluding waste treatment and agriculture, which can be seen from the average factor of five difference between N_{tot} and $N_{>0.1}$.

Table 3. Total particle number emissions (N_{tot}) and emissions of particles with d_p>0.1 µm ($N_{>0.1}$) for EU28 countries, by source sector. Emissions are presented for years 2010, for the current legislation scenario (CLE) in 2020 and 2025, and for 2025 for the central ambition level (A5) and the maximum technically feasible reductions (MTFR) scenarios. Percentage changes from year 2010 refer also to PM1 mass emissions for comparison.

	Emissions (x10^{23} particles)										**Change from 2010** (%)											
	2010		CLE 2020		CLE 2025		A5 2025		MTFR 2025		CLE 2020			CLE 2025			A5 2025			MTFR 2025		
	N_{tot}	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	N_{tot}	$N_{>0.1}$	PM_1	N_{tot}	$N_{>0.1}$	PM_1	N_{tot}	$N_{>0.1}$	PM_1	N_{tot}	$N_{>0.1}$	PM_1
Power generation	**176**	42	**139**	34	**139**	34	**78**	11	**68**	3	-21	-20	-37	-21	-19	-38	-56	-75	-79	-62	-93	-93
Domestic sector	**1639**	347	**1494**	315	**1357**	286	**1307**	243	**1114**	209	-9	-9	-13	-17	-18	-23	-20	-30	-46	-32	-40	-66
Industrial comb.	**109**	24	**118**	29	**104**	25	**66**	10	**49**	5	9	22	10	-5	6	-4	-40	-59	-54	-55	-78	-71
Industrial proc.	**218**	16	**199**	16	**197**	17	**58**	10	**56**	9	-9	4	3	-9	6	4	-73	-36	-32	-74	-43	-38
Fuel extraction	**0.14**	0.14	**0.13**	0.13	**0.12**	0.12	**0.12**	0.12	**0.12**	0.12	-4	-4	-4	-14	-14	-14	-14	-14	-14	-14	-14	-14
Road transport (RT)	**7741**	1243	**1889**	286	**894**	132	**894**	132	**894**	132	-76	-77	-64	-88	-89	-74	-88	-89	-74	-88	-89	-74
Shipping	**1824**	12	**1776**	10	**1788**	10	**1788**	10	**1788**	10	-3	-19	-8	-2	-19	-8	-2	-19	-8	-2	-19	-8
Other non-road trans.	**597**	157	**393**	84	**338**	63	**338**	63	**338**	63	-34	-46	-57	-43	-60	-73	-43	-60	-73	-43	-60	-73
Waste treatment	**38**	28	**39**	29	**39**	29	**30**	22	**30**	22	2	2	2	3	3	3	-22	-23	-21	-22	-23	-21
Agriculture	**262**	162	**303**	187	**303**	187	**0**	0	**0**	0	15	15	15	15	15	15	-100	-100	-99	-100	-100	-99
Total excl. RT & ships	**3039**	776	**2685**	694	**2478**	641	**1877**	359	**1656**	311	-12	-11	-13	-18	-17	-20	-38	-54	-52	-46	-60	-66
Total	**12603**	2031	**6350**	990	**5159**	783	**4559**	501	**4337**	453	-50	-51	-19	-59	-61	-27	-64	-75	-54	-66	-78	-66

Table 4. Same as Table 3, but segregating the emissions between EU28 countries.

	Emissions (x10^{23} particles)									
	2010		CLE 2020		CLE 2025		A5 2025		MTFR 2025	
	N_{tot}	$N_{>0.1}$	N_{tot}	$N_{>0.1}$	N_{tot}	$N_{>0.1}$	N_{tot}	$N_{>0.1}$	N_{tot}	$N_{>0.1}$
Austria	**265**	49	**87**	18	**65**	14	**54**	10	**51**	10
Belgium	**269**	47	**100**	14	**73**	10	**65**	7	**65**	7
Bulgaria	**110**	29	**74**	20	**59**	17	**40**	6	**37**	6
Cyprus	**25**	4	**11**	1	**6**	1	**6**	0.7	**5**	0.7
Czech Rep.	**254**	51	**159**	31	**128**	26	**109**	18	**99**	17
Denmark	**193**	28	**99**	11	**86**	8	**85**	7	**83**	7
Estonia	**33**	10	**20**	6	**16**	5	**15**	3	**12**	2
Finland	**188**	33	**108**	17	**93**	15	**93**	13	**81**	10
France	**2190**	297	**1014**	105	**849**	74	**790**	63	**749**	55
Germany	**1313**	215	**340**	60	**256**	47	**218**	40	**214**	38
Greece	**377**	45	**317**	29	**288**	25	**265**	13	**263**	12
Hungary	**146**	28	**69**	14	**44**	10	**39**	7	**36**	6
Ireland	**165**	25	**107**	21	**75**	13	**67**	11	**62**	11
Italy	**1753**	227	**991**	95	**839**	68	**795**	51	**788**	49
Latvia	**50**	13	**38**	9	**30**	8	**26**	5	**22**	4
Lituhania	**69**	19	**56**	15	**44**	12	**37**	7	**33**	6
Luxembourg	**59**	10	**12**	2	**5**	1	**5**	1.0	**5**	0.9
Malta	**5**	1	**1**	0.3	**1**	0.2	**1**	0.1	**1**	0.1
Netherlands	**302**	42	**84**	11	**67**	8	**64**	7	**63**	7
Poland	**1306**	230	**906**	162	**717**	132	**646**	105	**593**	96
Portugal	**299**	53	**100**	21	**63**	16	**48**	6	**45**	6
Romania	**247**	78	**197**	64	**178**	59	**111**	20	**94**	17
Slovakia	**100**	21	**50**	10	**40**	8	**35**	5	**31**	5
Slovenia	**51**	11	**18**	5	**14**	4	**12**	2	**12**	2
Spain	**1270**	254	**753**	168	**557**	137	**401**	49	**391**	47
Sweden	**218**	38	**109**	18	**98**	17	**93**	16	**71**	8
UK	**1290**	159	**504**	50	**448**	39	**425**	23	**420**	22
EU27	**12550**	2018	**6324**	981	**5139**	775	**4546**	498	**4327**	450
Croatia	**53**	14	**26**	9	**21**	8	**13**	3	**11**	3
EU28	**12603**	2031	**6351**	990	**5160**	783	**4559**	501	**4337**	453

	Change from 2010 (%)											
	CLE 2020			CLE 2025			A5 2025			MTFR 2025		
	N_{tot}	$N_{>0.1}$	PM_1	N_{tot}	$N_{>0.1}$	PM_1	N_{tot}	$N_{>0.1}$	PM_1	N_{tot}	$N_{>0.1}$	PM_1
Austria	-58	-63	-22	-69	-71	-29	-74	-79	-63	-75	-80	-71
Belgium	-63	-70	-30	-73	-79	-34	-76	-85	-53	-76	-86	-61
Bulgaria	-33	-31	-18	-46	-40	-26	-64	-78	-67	-67	-80	-79
Cyprus	-58	-62	-48	-77	-79	-62	-78	-81	-71	-78	-82	-74
Czech Rep.	-38	-39	-17	-49	-48	-16	-57	-64	-47	-61	-67	-63
Denmark	-49	-60	-48	-56	-71	-56	-56	-75	-71	-57	-76	-79
Estonia	-39	-38	-27	-52	-48	-31	-54	-70	-63	-64	-76	-81
Finland	-43	-48	-29	-51	-55	-37	-50	-59	-52	-57	-70	-73
France	-54	-65	-28	-61	-75	-39	-64	-79	-48	-66	-81	-65
Germany	-74	-72	-29	-80	-78	-33	-83	-82	-49	-84	-82	-55
Greece	-16	-36	-28	-24	-44	-28	-30	-72	-69	-30	-73	-76
Hungary	-53	-50	-25	-70	-65	-34	-73	-76	-65	-75	-78	-77
Ireland	-35	-16	-6	-55	-48	-14	-60	-55	-35	-62	-57	-44
Italy	-43	-58	-25	-52	-70	-35	-55	-78	-58	-55	-78	-64
Latvia	-24	-27	-13	-41	-41	-24	-48	-63	-52	-55	-69	-76
Lituhania	-19	-23	-10	-36	-36	-25	-47	-65	-60	-52	-68	-76
Luxembourg	-80	-80	-46	-91	-90	-50	-91	-90	-58	-91	-91	-66
Malta	-75	-72	-59	-86	-83	-65	-88	-89	-74	-89	-90	-77
Netherlands	-72	-73	-29	-78	-80	-32	-79	-82	-41	-79	-83	-45
Poland	-31	-30	-6	-45	-43	-18	-51	-55	-42	-55	-58	-55
Portugal	-66	-59	-25	-79	-69	-29	-84	-88	-65	-85	-90	-73
Romania	-20	-18	-15	-28	-24	-23	-55	-74	-65	-62	-78	-81
Slovakia	-50	-52	-18	-60	-60	-21	-65	-74	-57	-69	-78	-78
Slovenia	-63	-51	-18	-72	-59	-19	-76	-79	-73	-77	-78	-84
Spain	-41	-34	-9	-56	-46	-12	-68	-81	-67	-69	-82	-75
Sweden	-50	-52	-17	-55	-57	-18	-57	-57	-31	-67	-79	-69
UK	-61	-69	-1	-65	-75	-11	-67	-85	-51	-67	-86	-57
EU27	-50	-51	-19	-59	-62	-27	-64	-75	-54	-66	-78	-66
Croatia	-50	-34	-11	-61	-43	-19	-75	-77	-59	-80	-81	-73
EU28	-50	-51	-19	-59	-61	-27	-64	-75	-54	-66	-78	-66

The distribution of number emissions N_{tot} and $N_{>0.1}$ and PM1 mass emissions in different source sectors for 2010 is depicted in Figure 3. Over 60 % of total particle number emissions originate from road transport, 14 % from shipping and 13 % from the domestic sector. This is in strong contrast to PM1 mass emissions, for which the domestic sector is dominant with a share of 56 %, and road transport and shipping contribute only 12 % and 1 %, respectively. Non-road transport (excluding shipping) contributes 5 % to N_{tot} emissions, whereas the shares of each of the remaining sectors are 2 % or smaller.

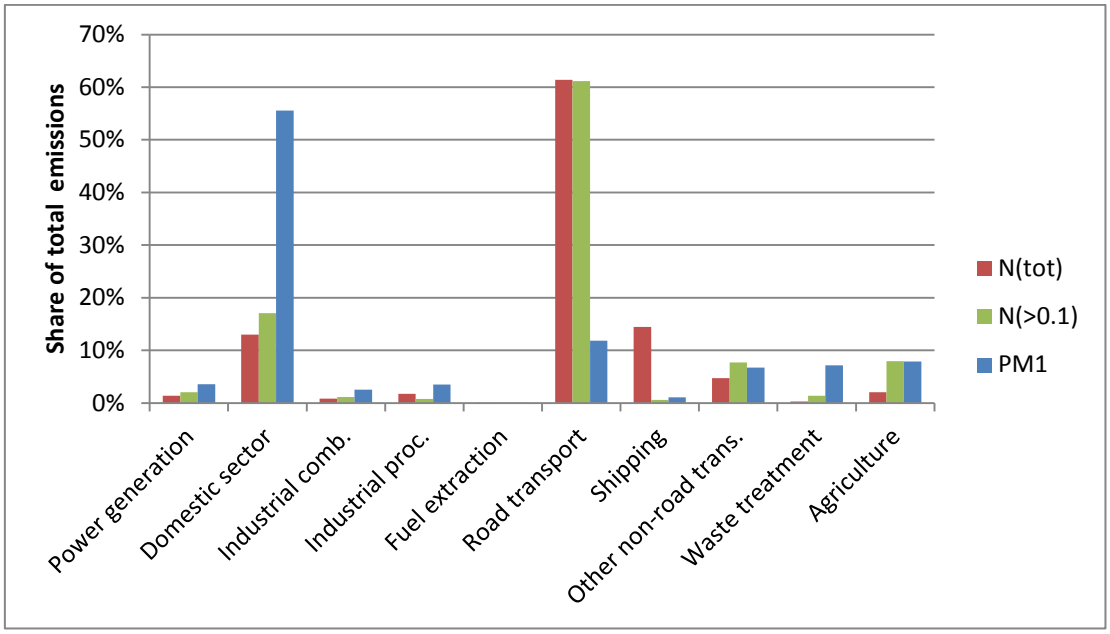

Figure 3. The distribution of number emissions N_{tot} and $N_{>0.1}$ and mass emissions PM1 in different source sectors in EU28 for year 2010.

The on-going and planned developments in road transport technologies are expected to cut PN emissions drastically. The estimated evolution of total particle number emissions is depicted in Figure 4 (note that road transport emissions for 2010, 7.7×10^{26} #/year, are far above the emission scale of the figure). Road transport PN emissions are estimated to decrease by 76 % from 2010 until 2020 and by 88 % until 2025 (scenarios for 2025 are similar to each other in terms of road and non-road transport emissions). Number emissions from the domestic sector are anticipated to decrease until 2025 far less than those from road transport, from 17 to 32 % depending on the scenario. PN emissions from shipping remain in these estimations practically constant. Emissions per ship decrease due to the decreasing share of ships using heavy fuel oil, which contains more sulphur and thus causes higher PN emissions than diesel oil (e.g. Diesch et al., 2013), but the increase in total activity evens out this decrease. However, due to lack of appropriate measurements we have not considered here the effect of limiting the sulphur content of ship fuel to 0.1 %, which will most probably decrease the emissions from national shipping in future. According to this analysis, particle number emissions from both domestic combustion and shipping will be higher than road transport emissions by 2025 (Table 2). The shares of shipping, domestic combustion and road transport in 2025 will be 35 %, 26 % and 17 %, respectively, in the CLE scenario, and 39 %, 29 % and 20% in the A5 scenario, and 41 %, 26 % and 21 % in the MTFR scenario.

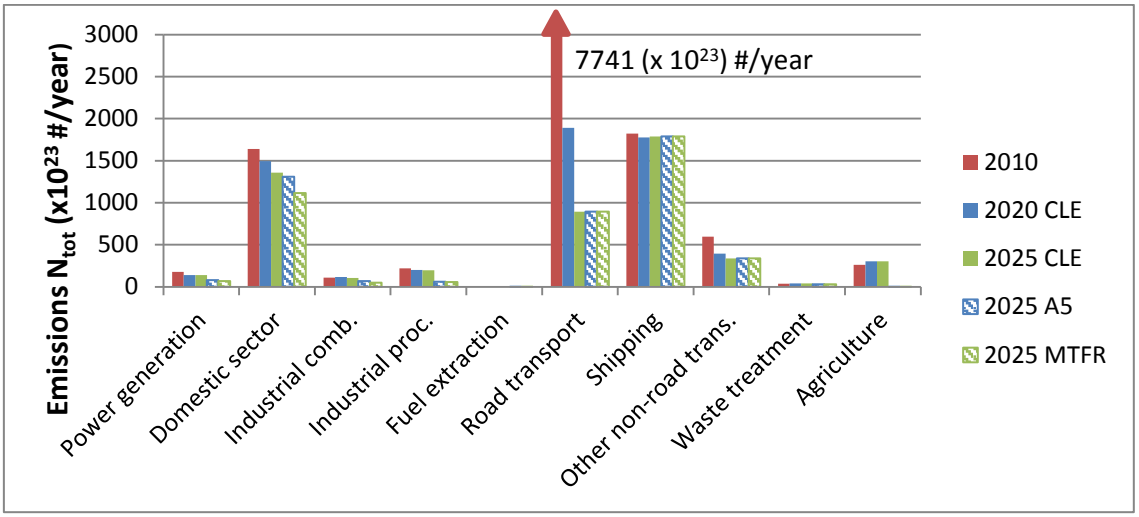

Figure 4. The yearly total particle number emission under the investigated scenarios.

Road transport is the major factor of future change in particle number emissions, and reductions in number emissions N_{tot} and $N_{>0.1}$ and mass emissions PM1 from road traffic are quite similar (see right hand side of Table 3). Also in the source sectors other than road transport and shipping the decreases in these different emission metrics are quite similar under the CLE scenario, as can be seen in the changes from year 2010 in Table 3, row 'Total excluding road transport and ships'. However, in total emissions the decrease of PM1 mass emissions is much smaller than of number emissions, because PM1 emissions are dominated by the domestic sector (see Figure 3). Under the more ambitious scenarios the reductions of emissions from other sources than road transport and shipping are far more effective for PM1 and $N_{>0.1}$ than for N_{tot}, and also the total reductions in $N_{>0.1}$ are over 10 percentage points higher than in N_{tot}. Higher reductions of emissions of larger particles in the A5 and MTFR scenarios arise mainly from the banning of open burning of agricultural waste, which emits mostly particles with diameters over 0.1 µm.

3.2.1 Detailed size segregation of estimated particle number emissions

More detailed size segregation of the reductions in particle number emissions under the different scenarios is depicted in Figure 5, in which emissions from road transport, shipping, domestic sector and the other source sectors are presented together with total emissions. In Figure 5a, the dominant role of road transport emissions in 2010, as well as their significant decrease, is clearly visible. In road transport emissions the highest emissions are of particles with diameter around 60 nm, which emerge from both diesel and gasoline vehicles. The nucleation mode particles, which form the 'elbow'-shape between 20 and 40 nm, originate mainly from diesel fuelled vehicles. As can be seen from Figure 1 the nucleation mode is dominant for EURO-III and VI heavy duty diesel vehicles. For older vehicles, from uncontrolled to EURO-II, the high emissions of larger particles seem to have scavenged the nucleating vapours and nucleation mode particles from the exhaust plumes. Improved technology in EURO-IV and newer heavy duty vehicles seems to decrease efficiently both the larger particle and nucleation mode particle emissions.

For the domestic sector (Figure 5c), only one mode with mean diameter between 40 and 50 nm is visible. This is the size range in which residential combustion of both wood and hard coal has high emissions, even though the peak diameter of hard coal emissions is in smaller sizes and that of conventional stoves in larger sizes (see Figure 2). In Figure 5c, for other source sectors than domestic and road transport, the decrease in emission in the CLE scenario emerges from non-road transport sources. Banning open burning of agricultural residuals causes a drastic difference between the CLE and the more ambitious A5 and MTFR scenarios.

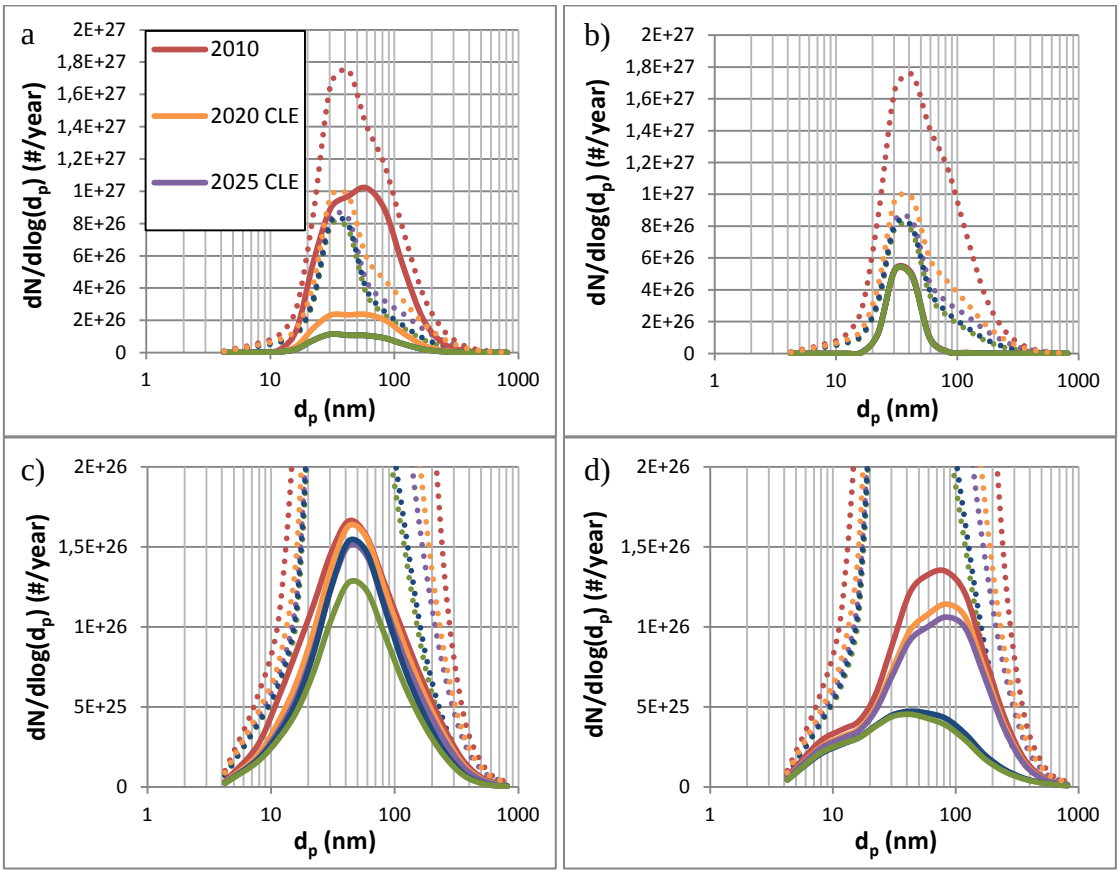

Figure 5. Size resolved PN emissions in EU28 under the studied scenarios, which are indicated in panel (a). Total emissions are shown with dotted lines in all panels, whereas the solid lines indicate PN emissions from (a) road transport, (b) shipping, (c) domestic combustion and (d) other source sectors. Note the order of magnitude difference in scales between upper and lower panels.

4 Discussion

4.1 Uncertainties in the estimates

The results presented above are the first estimates of particle number emissions with the GAINS model. While compiled to the best of our knowledge given time and budget constraints, they are subject to significant uncertainties, and there are numerous

possibilities for further improvements. However, we are confident that the applied emission factors and size distributions give reasonable estimates of *i*) the overall magnitudes of the particle number emissions, *ii*) the significance of different source sectors and *iii*) of the size ranges of emitted particles from these source sectors.

A comparison between the emission estimates for 2005 developed within the EU FP6 EUCAARI project by TNO and the GAINS results show reasonable agreement in all the above points *i-iii*), especially when corrected for the adjusted new PN emission factors.

In general, uncertainties in PN emission estimates originate mainly from the following reasons:

- Particle number emissions are dominated by nucleation mode particles in the ultra-fine particle size range. Most of these particles have formed from vapours through nucleation processes and are not in the solid phase. The level of nucleation mode emissions depends *i)* on air temperature, as at higher temperatures the nucleation of vapours is less effective, and *ii)* on the rate at which the aerosol is mixed with surrounding air, as the scavenging of vapours and nucleated particles to larger particles is most efficient before and immediately after the highly concentrated aerosol is introduced to the surrounding air. Thus, emission factors and size distributions are dependent on the measurement conditions in terms of temperature and dilution rate, but also the actual atmospheric emissions vary under different atmospheric conditions, mostly in terms of air temperature.
- Another main uncertainty relates to the scarcity of particle number emission data for several important source sectors. The majority of emission factors apart from road transport and domestic wood combustion are derived from PM1 mass emissions by distributing the mass according to the applied particle size distribution (PSD). Because a uniform PSD is applied for different technologies in these sources, the decrease in mass emissions results in a similar relative decrease in number emissions. This is, however, not correct in many cases: when emissions of large particles (which determine the total mass emissions) decrease, the sink for nucleating vapours and nucleation mode particles is reduced, leading to a smaller decrease or even an increase in total number emissions. This is visible in the data for domestic wood combustion, for which total number emissions increase and diameters of emitted particles decrease with advancing technologies.

Up to some point, these uncertainties can be reduced by a more detailed review of the most recent literature, but it is obvious that in order to derive quantitative estimates for particle number emissions more measurements are needed.

4.2 Future strategies for emission reductions

Even with a prime focus of emission control strategies on particle mass, it is important to monitor the concurrent evolution of particle number emissions, as their development can follow different trends. As mentioned earlier, the decrease in emissions of larger particles can in some cases lead to increasing number emissions of nucleation mode particles. It is possible that the increase in number emissions from newer/improved stoves compared to old/uncontrolled ones arises partly from this

phenomenon. It has been proposed that similar effect would take place in diesel vehicle emissions when the particle mass emissions decrease drastically while DPF systems are introduced (e.g. Arnold et al., 2012), but in the TRANSPHORM database this effect is not visible. This is most probably due to the very low sulphur content in diesel fuel in Europe, and the situation may be very different in countries in which diesel fuel contains more sulphur.

4.3 Climate-relevant particle number emissions

Particles with the highest climatic impact, with respect to their size and typical levels of concentration, fall in the size classes roughly between 0.1 and 1 µm, which are least efficient in depositing into the human respiratory system (Politis, M. et al., 2008). Roughly half of the number concentrations of >0.1µm particles in the European continental boundary layer originate directly from anthropogenic emissions, whereas the other half has grown to this size from smaller particles through condensation of biogenic vapours on smaller bio- and anthropogenic particles (Paasonen et al., 2013). Climate effects of aerosol particles are various. In addition to the capability of >0.1 µm particles to form cloud droplets thus cooling the climate (indirect effects), particles have direct climate effects: they either scatter solar radiation (also a cooling effect) or exert a warming impact by absorbing solar radiation (e.g., black carbon particles). Since also the share of black carbon emissions on total anthropogenic particle emissions can be quantified with GAINS (Kupiainen and Klimont, 2004 and 2007, Bond et al., 2013), estimation of the overall climate effects of reducing particle emissions will be possible in future studies.

Our initial analysis on particle number emissions suggests that in the current legislation scenario the decrease of emissions of particles in the size range > 0.1 µm is very similar to the decrease in total number emissions. However, in the more ambitious A5 and MTFR scenarios, emissions of particles > 0.1 µm are reduced by over 10 percentage points more than the corresponding total number emissions. This size resolved information is important when estimating the climatic impacts of emission reductions.

4.4 Next steps for calculating particle number emissions in GAINS

While a first step has been made to estimate current and future particle number emissions in Europe, improvements in the calculation methodology for several source sectors could significantly reduce uncertainties. This requires more detailed and/or more representative direct size resolved particle number emission measurements. Especially, more detailed descriptions of emissions from shipping, domestic coal combustion, non-road transport and industrial processes may alter the results significantly. The currently PM1 based emission factors in these source sectors should be replaced with direct emission factors corresponding to the different technologies considered in the GAINS model. Additionally, emissions from vehicles using alternative fuels, e.g., natural gas, biodiesel, etc., should be addressed in more detail. Finally, more detailed technology segregation for wood combustion emissions in the household sector may lead to slightly different conclusions on future emission trends.

It is also important to examine the representativeness of the current emission factors and size distributions, especially at the global scale. For instance, current emission factors for domestic combustion are derived from experiments for typical European and North-American burners and stoves. For global road transport, some of the vehicles outside the EURO standards are not well described, and the effect of fuel sulphur content on number emissions should be addressed. Additional efforts to improve the knowledge base will be warranted. However, even without such improvements, preliminary calculations at the global scale would be valuable as especially the climate relevance of particles active in cloud formation is as much a global as a regional issue.

5 Summary and conclusions

Particle number emission factors and the particle number size distributions related to these emissions have been introduced into the GAINS model. Addition of particle number emissions to the online GAINS model is envisaged during 2013.

Results show that road transport is currently the dominant source of particle number emissions in Europe, contributing more than 60 % to total emissions. Roughly 85 % of the particles emitted from road transport are ultra-fine particles (UFP), i.e., have diameters <0.1 µm (100 nm). This is in agreement with earlier particle number emissions estimates (H. Denier van der Gon et al., 2010a). However, the road transport share in particle numbers is rather different from the shares in PM2.5 or PM1 mass emissions, which are dominated by the domestic combustion sector. Also particles emitted from shipping, which is the second largest particle number source, are in the UFP size range and contribute minimally to mass based metrics.

The current legislation (CLE) scenario would reduce particle number emissions until 2025 by close to 60 %, mainly as a consequence of the 88 % reduction in road transport emissions. Shipping emissions are estimated to remain quite constant and domestic sector emissions to decrease by 17 %. Consequently, in 2025 road transport emissions would account for only 17 % of total emissions, whereas shipping emissions would contribute 35 % and the domestic sector 26 %.

Under the CLE scenario, the overall decrease in particle number emissions is estimated to be much stronger than the decrease in particulate mass emissions, due to the major impact of road transport emission mitigation measures especially on particle number emissions. However, for the more ambitious A5- and maximum technologically feasible reductions scenarios, particle number emissions from other sectors are estimated to decrease clearly less (from 14 to 20 percentage points) than the particle mass emissions. The different response of particle number and particle mass to changes in technologies can be expected, and in some cases a reduction in mass can even increase the number emissions.

These new estimates provide input for the calculation of indirect climate impacts of aerosols, which are heavily affected by number concentration of particles > 0.1 µm.

The current estimates of particle number emissions are subject to significant uncertainties, arising partly from the difficulties in estimating emissions of non-solid ultrafine particles, and partly from the limited number of available data for several source sectors. In order to reduce these uncertainties, new size resolved measurement data on particle number emissions from several important sources and (new) technologies are required.

It should be realized that our knowledge on the role and impact of UFP and particles > 0.1 µm in ambient air for both human health and climate controls through cloud formation are still not perfect. However, for making progress in these research fields, it is important to improve emission estimates. With increasing knowledge on health and climate effects of particle number concentrations, it is expected that in the future the number and size of emitted particles will play an important role in addition to the mass based metrics of PM10 and PM2.5.

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7 Appendix A: Emissions in European non-EU countries

Total particle number emissions and the emissions of particles > 0.1 µm for non-EU countries are presented in Tables A1 and A2. Under the applied scenarios for year 2025 there was no difference in activities or technologies for non-EU countries.

Table A1. The estimated total particle number emissions (N_{tot}) and the emissions of particles with d_p>0.1 µm ($N_{>0.1}$) for EU28 countries, divided to source sectors. Estimated emissions are presented for years 2010, 2020 and 2025 under the current legislation scenario (CLE). In percentage changes from year 2010, on right, corresponding changes in PM1 mass emissions are shown for comparison.

	Emissions (x10^{23} particles)						**Change from 2010 (%)**					
	2010		2020		2025		2020			2025		
	N_{tot}	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	N_{tot}	$N_{>0.1}$	PM1	N_{tot}	$N_{>0.1}$	PM1
Power generation	**111**	48	**102**	44	**106**	46	-8	-8	-11	-4	-3	-10
Domestic sector	**578**	136	**548**	126	**541**	124	-5	-7	-7	-6	-9	-9
Industrial comb.	**174**	53	**218**	70	**235**	76	25	32	24	35	44	37
Industrial proc.	**1509**	125	**1527**	135	**1530**	139	1	8	8	1	11	11
Fuel extraction	**0.12**	0.12	**0.12**	0.12	**0.12**	0.12	0	0	0	6	6	6
Road transport (RT)	**1784**	321	**1057**	163	**645**	98	-41	-49	-48	-64	-69	-61
Shipping	**280**	1	**290**	1	**299**	1	4	-1	2	7	6	7
Other non-road trans.	**1223**	114	**1221**	113	**1178**	113	0	-1	-1	-4	-1	-2
Waste treatment	**37**	30	**38**	31	**38**	31	2	2	2	3	3	2
Agriculture	**660**	408	**651**	402	**643**	397	-1	-1	-1	-3	-3	-3
Total excl. RT & ships	**4292**	913	**4305**	922	**4271**	927	0	1	1	0	1	2
Total	**6357**	1236	**5652**	1085	**5215**	1026	-11	-12	-1	-18	-17	0

Table A2. Same as Table A1, but segregating the emissions between non-EU countries.

	Emissions (x10^{23} particles)						Change from 2010 (%)					
	2010		2020		2025		2020			2025		
	N_{tot}	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	**N_{tot}**	$N_{>0.1}$	N_{tot}	$N_{>0.1}$	PM1	N_{tot}	$N_{>0.1}$	PM1
Albania	**32**	9	**23**	7	**19**	6	-27	-23	-4	-41	-33	-8
Belarus	**129**	43	**144**	43	**156**	44	12	1	5	21	4	6
Bosnia Herzegovina	**30**	10	**19**	6	**15**	5	-35	-37	-32	-48	-45	-35
Iceland	**24**	1	**27**	1	**28**	0	10	-40	13	16	-51	12
Macedonia	**21**	8	**14**	5	**11**	4	-34	-38	-48	-46	-47	-57
Moldova	**25**	9	**25**	9	**22**	8	-1	-5	0	-15	-11	0
Norway	**317**	23	**196**	10	**174**	8	-38	-54	-6	-45	-66	-2
Russia (Eur)	**3213**	597	**2946**	536	**2588**	491	-8	-10	2	-19	-18	2
Serbia Montenegro	**156**	49	**123**	37	**108**	34	-21	-24	-26	-31	-30	-30
Switzerland	**115**	20	**47**	8	**30**	5	-59	-58	-37	-74	-73	-47
Turkey	**1110**	277	**833**	238	**798**	230	-25	-14	4	-28	-17	4
Ukraine	**1208**	193	**1282**	185	**1294**	190	6	-4	-3	7	-1	1
Total non-EU	**6357**	1236	**5652**	1085	**5215**	1026	-11	-12	-1	-18	-17	0