

## Modelling the Mixing of Size Resolved Traffic Induced and Background Ultrafine Particles from an Urban Street Canyon to Adjacent Backyards

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## ABSTRACT

By means of numerical simulations the aims of this study are as follows: (1) to investigate the dispersion and mixing of ultrafine particles (UFP) with pre-existing size resolved UFP in a street canyon and its vicinity with the ENVI-met 3D microscale model; (2) to show the effects of boundary conditions, like wind direction and traffic emissions, on the UFP concentration in the near vicinity; and (3) to evaluate the importance of deposition and coagulation at the street scale. The decrease in UFP concentration in nucleation mode particles (diameter < 30 nm) and Aitken mode particles (diameter between 30–100 nm) downwind of the street canyon is caused by the large differences in the size distributions of the emissions and the background. Based on the wind direction and traffic emissions, the UFP concentration over the rooftop increases by 23% for the South-West wind up to 165% for the Northern wind. The background distribution for the South-West wind direction remains mono-modal, with a peak in the Aitken mode as a result of the relatively low contribution of advected particles from the street canyon. For the Northern wind the background size distribution transforms to a bi-modal distribution, with peaks in the nucleation and the Aitken modes as a result of the high UFP concentration advected from the boulevard. The overall effect of deposition and coagulation on the UFP is negligible for the cases considered in this study. Overall, deposition is more efficient and faster than coagulation in removing particles, and especially those in the nucleation mode.

Keywords: Ultrafine particles; Size distribution; CFD; Street canyon; Aerosol dynamics.

## INTRODUCTION

From a population exposure point of view, air quality in street canyons is of a major importance, since the highest pollution levels are often concentrated there. Epidemiological and toxicological studies show the potential threat of ultrafine particles (UFP, diameter < 100 nm) on human health, associated with cardiovascular risk factors and changes in lung function (Oberdorster and Utell, 2002; Ibald-Mulli et al., 2002; Morawska et al., 2004; Donaldson et al., 2005; Crüts et al., 2008; Politis et al., 2008). These studies can benefit from the knowledge on the transport, mixing and aerosol dynamics phenomena at various scales that are essential for the dispersion and evolution of UFP. Regulatory measures for UFP at this point are missing despite the reported health implications, partly due to the insufficient knowledge related to the complex nature of these particles. Considerable efforts have been made in recent years to improve the scientific understanding of the dispersion and evolution of UFP. CFD (Computational Fluid Dynamics) models allow to simulate the flow characteristics and the

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dispersion of pollutants at different scales (e.g., vehicle's wake, street canyons, urban neighbourhood, city scale). CFD modelling may indeed be useful to investigate the details of flow characteristics at micro scale but it requires considerably large capacity of computer resources if adopted for large spatial scales (e.g., city). Modelling studies dealing with the air flow in areas with complex geometries are abundant (Di Sabatino *et al.*, 2007; Gromke *et al.*, 2008; Kumar *et al.*, 2008a; Narita *et al.*, 2008; Buccolieri *et al.*, 2009; Abdulsaheb and Kumar, 2010; Vos *et al.*, 2012b). However despite the large scientific interest and contribution to the topic of UFP for the last decade, the ultrafine particles still remain an area with open questions and even contradictory opinions.

A limited number of model studies, in particular CFD studies, accounts for the dispersion of ultrafine particles size distribution and total number of UFP. For example Gidhagen *et al.* (2004b) used a CFD code with implemented UFP dynamics in a street canyon study. They showed the importance of different transformation processes on the size distribution, the effect of traffic induced turbulence and emissions. Later, Kumar *et al.* (2009b) compared the measured vertical profiles of the total number of particles in a street canyon with CFD model results (without considering aerosol dynamics). They found a good agreement between modelled and measured total number of particles within a

factor 2–3. There is a general consensus that differences up to a factor 10 are not striking and unexpected for ultrafine particles (Beddows and Harrison, 2008; Morawska et al., 2008) due to the large uncertainties caused by the lack of standards for the physical modelling and field measurements of UFP. Nikolova et al. (2011b) showed how UFP disperse in a street canyon with the 3-D CFD model ENVI-met, but the focus was only on the total number of particles. Limited is also the number of UFP modelling studies with CFD considering the neighbourhood and city scales. Birmili et al. (2009) studied the UFP dispersion on a German highway with a micro scale model. They highlighted the increasing application of 3-D flow models in urban micro scale environments and their usefulness for determining trafficderived UFP concentrations. Gidhagen et al. (2004a) and Wang et al. (2008) demonstrated the fast decrease of total number of particles with distance from the source as a result of dilution.

The model studies of the UFP dispersion remain scarce as compared to other pollutant dispersion studies (for example particulate matters  $PM_{10}$  or  $NO_2$ ), especially when it comes to the dispersion of UFP in complex urban terrain. This is on the one hand caused by the limited number of dispersion models that can deal with ultrafine particles, closely related to the considerable efforts for model development. On the other hand the lack of network with continuous and standardized field measurements hampers the robust and thorough validation of the dispersion models. Various UFP measurement campaigns are reported in the literature, but the intercomparison of those campaigns is in most cases impeded due to for example different instrument limits for the particles diameter, different height of the instruments, distance from the source, etc.

In this paper we present an UFP module developed and implemented in the 3D CFD model ENVI-met that accounts not only for the total number of particles as presented in Nikolova *et al.* (2011b), but also for the UFP size distributions. Two scenarios are generated for different wind directions over the same domain aiming to evaluate the importance of traffic and boundary conditions on the downwind size resolved UFP concentration. The aim of this study is (1) to investigate, by means of numerical simulations, the size resolved dispersion and dilution of traffic emitted ultrafine particles with background UFP in a street canyon and the near surroundings, (2) to show the importance of the boundary conditions like wind direction and traffic emissions on the UFP concentration in the near surroundings and (3) to evaluate the importance of deposition and coagulation at street scale.

# METHODOLOGY AND THEORETHICAL FOUNDATIONS

## **CFD** Model

ENVI-met is a RANS (Reynolds Averaged Navier-Stokes) equations based, non-hydrostatic micro scale obstacleresolving model with advanced parameterizations for the simulation of surface-plant-air interactions (Bruse and Fleer, 1998). The model also has a module for the dispersion of polluting gases and particles, as well as their interaction with buildings and vegetation. For this study, ENVI-met is extended with an UFP module in order to account for the size resolved dispersion of ultrafine particles, including support for deposition and coagulation.

So far, all these features (CFD, vegetation module, micro climate, size resolved UFP, aerosol dynamics) are never combined into one single air quality and micro climate model. As such, this is the first model of this type to present the dispersion and mixing of size resolved UFP from street to the near surroundings.

#### UFP Dynamics in ENVI-Met

While the transport processes mainly affect the spatial distribution of UFP, processes like deposition and coagulation may have an effect on the UFP size distribution, i.e., the distribution of the UFP concentration in function of the particle diameter. In total 12 size bins have been implemented in ENVI-met, 10 between 1 and 100 nm, and 2 between 100 and 700 nm. Table 1 summarizes the main parameters for the size bin approach as well as the names of the size intervals used in the paper. Nucleation mode is the size interval for particles with diameters between 1 and 30 nm, Aitken mode is the size interval for particles diameter between 30 and 100 nm and accumulation mode is the size

Number of bins k	Bin boundaries *10 <sup>-9</sup> [m]	Mean particle diameter *10 <sup>-9</sup> [m]	Width of the size bin logDp	Name of modes
1	1-10	5	1	
2	10-20	15	0.301	Nucleation mode
3	20-30	25	0.176	
4	30–40	35	0.125	
5	40-50	45	0.097	
6	50-60	55	0.079	
7	60-70	65	0.067	Aitken mode
8	70-80	75	0.058	
9	80-90	85	0.051	
10	90-100	95	0.046	
11	100-200	150	0.301	A a annun lation mada
12	200-700	350	0.544	Accumulation mode

Table 1. Size bins parameters implemented in ENVI-met.

interval for all particles bigger than 100 nm in diameter. All size bin boundaries are fixed and all particles in a size bin share the same diameter. Additionally, all particles are assumed to have the same density (1 g/cm<sup>3</sup>). Kannosto et al. (2008, forest site) showed that the density of grown 33 nm particles was about 1.0 g/cm<sup>3</sup> with density variations between 0.9 and 1.3 g/cm<sup>3</sup> in the Aitken mode. The density of the nucleation mode particles at 17 nm was approximately 1.3 g/cm<sup>3</sup> with variations in the 15-30 nm size range between 0.5 and 1.5 g/cm3. The density of the ultrafine particles at roadsides should be expected to be higher due to the small amount of water in the composition of the particles (Chen et al., 2010). However, the above discussion is based on the assumption that all particles are spherical. In reality the majority of the atmospheric particles are nonspherical and multi-component in composition and the effective density is often used instead. Nonetheless, Park et al. (2003) found the effective density of diesel particles decreased with increasing particle sizes, which was around 1.0, 0.8, 0.6 and 0.5 g/cm<sup>3</sup> for particles of 50, 100, 150 and 300 nm in mobility diameter, respectively. In addition condensation/evaporation should also be considered if one wants to have a detailed dynamic representation of the density of ultrafine particles. Recent studies suggest that semi-volatile organic compounds (Robinson et al., 2007) play a role in the evolution of ultrafine particles via evaporative shrinkage of nanoparticles (Dal'Osto et al., 2010) that would have an effect on the particle density, too. However, in our simulations we do not account for the nonspherical, multi-component nature of the particles and the evolution of the size distribution via condensation/ evaporation.

#### Deposition

Deposition similarly to coagulation reduces the number of UFP. The scheme used here is the so called scheme of resistances described in details in Seinfeld and Pandis (1998). The scheme considers dry deposition only. The deposition velocity is a function of the aerodynamic resistance  $r_a$ , the quasi-laminar resistance  $r_b$  and the sedimentation speed  $v_s$  as follows:

$$v_{d} = \frac{1}{r_{a} + r_{b} + r_{a}r_{b}v_{s}} + v_{s}$$
(1)

Particles can deposit on different surfaces like ground, building roofs and vegetation. In contrast with the deposition of  $PM_{10}$  where the sedimentation speed plays a dominant role, the deposition for UFP is mainly affected by the aerodynamic and quasi-laminar resistances. Bruse (2007) details the technical implementation of deposition in ENVI-met whereas Wania *et al.* (2011) analysed the influence of different street vegetation on traffic induced particle dispersion using ENVI-met.

#### Coagulation

Coagulation is the process of collisions of particles due to their Brownian motion. When two particles collide it is assumed that they adhere. The collision rate CR  $[m^3/s]$ , referred from now on as coagulation rate, is calculated

following the mathematical description in Seinfeld and Pandis (1998):

$$CR_{ij} = \frac{2\pi (D_i + D_j) (D_{pi} + D_{pj})}{\frac{D_{pi} + D_{pj}}{D_{pi} + D_{pj} + 2\sqrt{g_i^2 + g_j^2}} + \frac{8(D_i + D_j)}{(D_{pi} + D_{pj})\sqrt{c_i^2 + c_j^2}}$$
(2)

where i and j refer to the corresponding size bins,  $Dp_{i,j}$  is the mean particles diameters per size bin [m],  $c_{i,j}$  is the thermal speed [m/s],  $D_{i,j}$  is the diffusion coefficient [m<sup>2</sup>/s] and  $g_{i,j}$  is a function of the diffusion coefficient and the mean free path. The net amount of coagulation is calculated according to:

Coagulation = 
$$\sum_{i=1}^{12} 0.5 \sum_{j=1}^{i-1} CR_{j,i-j} N_j N_{i-j} - N_i \sum_{j=1}^{12} CR_{i,j} N_j$$
 (3)

where  $N_{j,i}$  is the concentration of UFP per corresponding size bin. The first sum on the right is the production of particles and the second sum is the loss of particles due to collisions. The overall loss is bigger than the production leading to a reduction of the total number of particles.

#### Condensation and Nucleation

The nucleation and condensation of fresh particles in the car exhaust and in the vehicle wake are assumed to be included in the emission model. In addition, studying regional nucleation events and growth of particles with ENVI-met is out of the scope because of its spatial resolution. As a result, the processes of condensation and nucleation will not be considered further in this study.

#### Configuration of the CFD Model

The domain is based on a real world street canyon, described in details in Nikolova et al. (2011b). Briefly, the area is a typical busy urban commercial/residential area in the city of Antwerp, Belgium. It is located at 51°12'32.00"N and 4°25'56.00"E. The modelled street canyon is 12 m wide and approximately 120 m long and oriented northsouth. The average height of the buildings is 11m and the building-height-to-street-width ratio H/W is on average 0.92. In total 1.9 million cells covered the study area with a grid resolution of  $1 \times 1$  m in x and y directions. Vertically, z varied from 20 cm in the first 2 m to a few meters at the top of the model domain. The north exit of the street canyon connects to a busy boulevard. The emissions on the traffic lanes were estimated based on the parameterization of Nikolova et al. (2011a). The emission model UFPEM was evaluated against measurements of UFP in a tunnel in Nikolova et al. (2011a) and Vos et al. (2012a). Traffic count showed that in total 5690 and 277 passenger cars between 17 and 18 h (rush hour) were driving on the boulevard and in the street canyon on 28/07/2009, respectively. Respectively 80 and 10 Heavy Duty Vehicles were counted on the boulevard and the street canyon. This day was randomly selected from the 14 days measurement campaign in Nikolova et al. (2011b) who successfully validated the

ENVI-met model for total number of UFP by comparing measured and simulated results for four locations in the same street canyon. Background measurements were performed in a street, blocked for non-residential traffic, next to the canyon, such that the ultrafine particles level is not influenced significantly by any single source, but rather by the integrated contribution from all sources (e.g., by all traffic, combustion sources, etc.) upwind of the measurement point. During the measurement campaign, the prevailing winds were from south south-west, therefore the integrated contribution from the street canyon and the boulevard is minimized. Background total number of particles were measured with water based CPC (Condensational Particle Counter type 3786, TSI Inc., http://www.tsi.com/). The instrument measured the total number concentration from 3 to 1000 nm. However, the background size distribution was not measured. In order to overcome this drawback we combined a typical urban background size distribution found in the literature, with the total number concentration measured in the background location of the current domain. In this study we used the shape of the urban background size distribution presented in Ketzel et al. (2004), roof-top urban station HCOE. By fitting the log-normal roof-top size distribution we estimated the percentage of particles per size bin and distributed our UFP measured total numbers accordingly. The size distribution is mono-modal distribution with the majority of the ultrafine particles in the Aitken mode. The background size resolved UFP distribution and the emissions, estimated using UFPEM, on the boulevard and in the street canyon for 28/07/2009 at 17 h are given in Table 2.

The initial meteorological parameters for the two scenarios are presented in Table 3. Run #1 (SW wind) is representative of low to moderate UFP emission load in the street canyon. The meteorological conditions were the actual meteorological conditions at that day and hour, obtained from the local airport in Antwerp, Belgium. Run #2 is a case with a hypothetical Northern wind, having the same wind speed as Run #1. Wind speed was kept the same in both scenarios in order to evaluate the role of the aerosol transformation processes. This case represents a scenario with very high UFP emissions, mainly in the nucleation and Aitken modes, on the boulevard. With these two scenarios we evaluated the UFP concentration and size distributions in the proximity of the sources and in the empty space surrounded by the buildings, reffered hereafter as backyard. In street canyons with H/W ratio close or equal 1 and regional wind speed  $\geq 2$  m/s, the dynamics in the canyon is governed mainly by the wind direction and the creation of a vortex inside the canyon as discussed in Kumar et al. (2008a). There is a critical wind speed (below 1.5 m/s) under which the traffic induced turbulence plays a role, confirmed by several studies (Di Sabatino et al., 2003; Kastner-Klein et al., 2003; Mazzeo and Venegas, 2005; Solazzo et al., 2007). This can additionally reduce the total number UFP concentration in a street canyon (Gidhagen et al., 2004b; Ketzel et al., 2005). However traffic induced turbulence was not considered in this analysis due to the high regional wind speed.

## **RESULTS AND DISCUSSION**

## South West Wind (Run #1)

Fig. 1 presents a snapshot of the modelled UFP total number distribution for July, 28th 2009, 17 h. The red vector indicates the direction of the flow crossing the canyon. We investigate the total number UFP concentration as well as the evolution of the UFP size distribution along the vector. The distance considered up- and downwind of the canyon is about 20 m. The UFP total number concentrations upwind of the canyon is purely background with no signal from the traffic emitted UFP particles, assuming that the background concentration is homogeneously distributed in space and time. A clear increase in the UFP concentration is simulated in the canyon, related to the traffic emitted UFP. The peak concentration in the centre line from 1.5 m to 4.5 m decreases (not shown), caused by the mixing of particles with fresher air, but the concentration remains higher than in the background as a result of the reduced ventilation. Particles from the canyon are lifted due to the vortex inside the canyon and transported above rooftop. Above building's height, the concentration notably decreases caused by the

Table 2. Ba	ckground U	FP concentrati	ion per size b	in and siz	e resolve	d traffic	emissions	on the	boulevard	and in t	he street
canyon. Pass	senger cars/1	7 h: boulevard	15690, street	canyon 27	7; Heavy	Duty Vo	ehicles/17 h	n: boulev	vard 80, st	reet cany	on 10.

Sizo hin	Particles diameter	Background concentration	Emissions boulevard	Emissions canyon
Size olii	nm	#/cm <sup>3</sup>	#/(m.s)	#/(m.s)
1	5	0	8.22E+09	4.65E+08
2	15	1.16e+03	1.01E+11	5.88E+09
3	25	1.74E+03	3.64E+10	2.12E+09
4	35	1.48E+03	2.99E+10	1.73E+09
5	45	1.16E+03	2.41E+10	1.39E+09
6	55	8.82E+02	1.54E+10	8.72E+08
7	65	6.70E+02	8.78E+09	4.89E+08
8	75	5.11E+02	4.86E+09	2.63E+08
9	85	3.92E+02	2.69E+09	1.42E+08
10	95	3.04E+02	1.53E+09	7.76E+07
11	150	1.06E+03	2.38E+09	1.10E+08
12	350	2.14E+02	5.70E+07	2.20E+06
Total	concentration	9.57E+03	2.35E+11	1.35E+10

		P	
28/07/2009 17 h	Wind direction	Wind speed	Т
# Run	[°]	[m/s]	[K]
1	232	3	294
2	360		

 Table 3. Initial regional meteorological parameters to initialize ENVI-met for the selected time period.

dilution with fresher air, but remains higher as compared to the background level, an apparent contribution from the traffic. Background concentration level is reached at about 25m height above the ground. Downwind of the canyon (in the backyard) the UFP number concentration has increased with 23% in comparison with the background concentration due to the advection of particles from the canyon (from 9573 particles/cm<sup>3</sup> to 11760 particles/cm<sup>3</sup>).

The average size distributions in the background, within the street canyon and in the backyard are plotted in Fig. 2. The background size distribution is mono-modal with a peak in Aitken mode, based on the average urban background size distribution shown in Ketzel et al. (2004) and adapted to our study domain (Table 2). In the canyon the distribution is bi-modal with peaks in the nucleation and Aitken modes. The concentration of the simulated particles per size bin decreases with height but the two modal shape remains intact within the street canyon. This effect is expected due to the confined space in the canyon and the slow removal processes (deposition and coagulation) which we discuss in section 3.3. Above the canyon, particles in the nucleation mode almost disappear completely due to the dilution between the two mediums with large differences in concentrations. In the backyard, the distribution has the

mono modal peak in the Aitken mode similar to the background size distribution. It can be shown that the 23% increase in the background concentration is due to the advection of nucleation and Aitken mode particles from the canyon. The contribution of advected UFP per size bin from above rooftop is given in Fig. 4. In the first size bin (diameter 5 nm), all particles in the backyard are traffic related and advected from the canyon. Almost half of the particles in the second size bin (diameter 15 nm) are traffic related and transported into the backyard. For the rest the number of particles transported above rooftop decreases with the increase in the size bin diameter. Particles in the accumulation mode are mainly present in the atmosphere as aged particles and the UFP number concentration in the accumulation mode remains practically the same as compared to the background accumulation mode UFP concentration.

Due to the uncertainties connected with the missing measured background size distributions, the interpretation of the current results is based on the assumption that there are no background UFP in the first size bin. This representation might not be fully realistic, however, the cases considered in this study offer an opportunity to examine a scenario with sharp gradients in the concentrations that is often reported in literature. The main underlying driving force for the dilution and fast decrease in the UFP size bin concentration is the difference between the concentration per size bin in and above the canyon, also discussed in Kumar et al. (2008b). The UFP concentration decreases with height due to the removal of particles as a result of mass exchange between the street and the less polluted air from above. Kumar et al. (2009a) measured the vertical distribution of size resolved UFP in another street canyon. They showed that the shape of street level particle number distributions was similar for the



Fig. 1. Dispersion of UFP in the study domain with SW wind, snapshot at 17 h.



Fig. 2. Average size distribution in and downwind the canyon (backyard).



Fig. 3. Share of particles transported from the canyon that contribute to the background size distribution concentration.

selected heights (due to the slow transformation processes) with consistent decrease in the number concentration per size mode. Wang et al. (2008) and Zwack et al. (2011) modelled the dispersion of traffic emitted UFP (total number of particles) in a neighbourhood scale, showing that the concentration decays exponentially with distance. This is also confirmed by the measurements by Zhu et al. (2009), showing that the UFP concentration decayed exponentially with increasing distances with sharp concentration gradients observed within 100-150 m from the roadway. Within the measured size range, smaller particles (6-25 nm) decayed faster than larger ones (100-300 nm). He and Dhaniyala (2011) studied the vertical and horizontal distributions of ultrafine particles (total numbers) also near a highway and found that 50 m away from the highway and on a height above 8m the UFP concentration was uninfluenced by the traffic source from the highway. This could be explained by the fact that a wide open space (no obstacles like buildings, trees, etc.) enhances the dilution between the observation point and the traffic lane.

## North Wind (Run #2)

In Run #2 we investigate the dilution and transport phenomena from high UFP emission site on the boulevard to the background UFP concentration in the near surroundings, particularly in the backyard. Fig. 5 shows the spatial distribution of the total number UFP for northern wind. Similarly to the previous example, the UFP concentration and size distribution will be investigated along the red vector. The boulevard is 36 m wide street and the distance considered downwind of the boulevard (in the backyard) is about 15 m.

High number of simulated UFP are advected to the south, increasing the UFP number concentration in the surroundings. The simulated UFP number concentration on both lanes decreases rapidly but despite the mixing with fresh air and advection to the south, the concentration at 15 m above the ground (not shown) remains higher as compared to the background level. The background concentration level is reached at about 60m height above the ground. Downwind of the boulevard, in the backyard between the buildings, the UFP concentration drastically increases as compared to the urban background - from 9573 particles/cm<sup>3</sup> to 25400 particles/cm<sup>3</sup> or 165%. In comparison with run #1, here the traffic emissions have a strong and rather severe mark on the UFP background concentration. The evolution of the size distribution is shown in Fig. 5.

Despite the dilution, the UFP concentration in the nucleation and the Aitken modes remain high. The size distribution on the boulevard is dominated by nucleation mode particles while downwind of the boulevard (in the



Fig. 4. Dispersion of UFP in the study domain with northern wind, snapshot at 17 h.



Fig. 5. Average size distribution on the boulevard and downwind the boulevard (in the backyard).

backyard) nucleation and Aitken modes particles have almost equal share in the size distribution. The increase of 165% in the backyard concentration is due to the advection of particles mainly in nucleation and Aitken modes from the boulevard, shown in Fig. 6. The mono modal background UFP size distribution transforms to a two modal structure with well-defined nucleation and Aitken modes. From the first size bin (5 nm) to size bin with diameter 55 nm more than half of the particles are traffic induced and transported from the boulevard.

#### Importance of Deposition and Coagulation

The role of deposition and coagulation is evaluated with 4 model runs – one without deposition and coagulation,

one with deposition only, one with coagulation only and one with both deposition and coagulation. The UFP size distribution in the street canyon is analyzed at the location with the highest concentration (10 cm height).

Coagulation and deposition both reduce the total number of particles with 0.95% in the street canyon. Coagulation reduces only 0.3% of the total number of particles while deposition is slightly more efficient with 0.65% reduction. The smallest size bin losses about 4.3% of its particles mainly due to deposition that remains more effective in removing the nucleation mode particles than coagulation. Between the different modes, the coagulation is faster for the nucleation mode particles with minor effect on the concentration in Aitken and accumulation modes. This is



Fig. 6. Share of particles transported from the boulevard that contribute to the background size distribution concentration.

because coagulation is efficient between particles of different sizes with smaller particles having high mobility (due to their Brownian motion) and larger particles providing a large cross-section. We evaluated the role of coagulation in the case of increased number concentration per size bin on the boulevard and we found that the loss of particles due to coagulation increases but the overall net effect on the total number concentration is negligible (0.5%). The total loss of particles on the boulevard due to the combined effect of deposition and coagulation is 2.3%. This loss is dominated by deposition (1.8%) and the removal of particles in the 5 nm and 15 nm size bins reaches about 8% and 3%, respectively. For ultrafine particles (diameter < 100 nm) the settling velocity vs is rather small as compared to the settling velocities of fine (PM<sub>2.5</sub>) and coarse (PM<sub>10</sub>) particulate matters. An increase/decrease in particle's density (parameter that is used in the estimation of the settling velocity vs) would lead to a marginal increase/decrease of the settling velocity of ultrafine particles, however the deposition velocity of the ultrafine particles is mainly driven by the aerodynamic and quasi-laminar resistances. If we neglect for a moment the aerodynamic resistance (that depends on the state of the atmosphere), then the overall dry deposition velocity is vd = (1/rb) + vs (Seinfeld and Pandis, 1998). The value of 1/rb is large for very small particles (ultrafine size range) because of the efficient transport mechanism across the surface layer by Brownian diffusion. In other words the ultrafine particles experience faster Brownian motion and therefore less sublayer resistance in comparison with coarser particles. This also explains why particles in the nucleation mode deposit faster in our simulation in comparison with particles in the accumulation mode. For very large particles ( $PM_{2.5}$ ,  $PM_{10}$ ) impaction and interception are leading and effective mechanisms of removal in which density would have a more prominent effect on the deposition velocity.

In the literature there is a debate on which processes are important for the UFP size distribution and to what extent. The general consensus is that removal processes such as dilution and dry deposition should be considered at street scale (Gidhagen *et al.*, 2004a; Ketzel *et al.*, 2005). The relevance of these processes may vary depending on the concentration of the exhaust emissions, the meteorological conditions, complexity of the area, etc. (Wehner *et al.*, 2002;

Charron and Harrison, 2003; Kumar et al., 2008a). On the importance of the coagulation process, there exist rather contradictory opinions - some that favour its inclusion in dispersion models and others that do not. For example Gidhagen et al. (2004b) and Ketzel and Berkowicz (2004) show evidence in support of both but the underlining principles are dependent on a variety of factors already mentioned above. Moreover, the total net effect on the particle number concentration may not be affected notably (Kumar et al., 2008b, 2009a) due to the competition between the removal processes. Vignati et al. (1999) found that due to rapid dilution only very small particles (2 nm) have a coagulation time scale which is comparable with typical residence times of pollutants in a street. Zhu et al. (2002) reported that atmospheric dilution and coagulation near a highway played important roles in the rapid decrease of particles number concentrations and transformation of the size distribution. On the contrary, Zhang and Wexler (2002), Pohjola et al. (2003) and Gidhagen et al. (2005) showed that coagulation is too slow to alter the particles size distribution, also confirmed in the cases considered in this study. A method to determine the relative importance of various processes is the time scale analysis discussed in Ketzel and Berkowicz (2004). The dilution time at the street scale is a function of the canyon height H and the rooftop wind speed u<sub>roof</sub>, as follows:

$$\tau_{dilution} = \frac{H}{0.1 u_{roof}} \tag{4}$$

For H = 11 m and  $u_{roof} = 3$  m/s, the corresponding time scale is about 37 s. For low wind speed conditions ( $u_{roof} = 1$  m/s), the dilution time will be 110 s. The time scales for coagulation and deposition can be estimated as follows:

$$\tau_{coagulation} = \frac{N}{\partial N / \partial t} \approx 333 \text{s}$$
(5)

$$\tau_{deposition} = \frac{N}{\partial N / \partial t} \approx 154 \text{s}$$
(6)

where N is the total number concentration (#/cm<sup>3</sup>) and  $\partial N/\partial t$  is the overall amount of particles removed per second

 $(\#s^{-1}/cm^3)$  for coagulation and deposition, respectively. The time scale for coagulation in our study remains about two times lower than the time scale for deposition. Overall, the time scale analysis supports the general opinion in literature that dilution remains faster in comparison with deposition and coagulation, followed next by deposition.

## CONCLUSIONS

The aim of this study was to show, by means of numerical simulations, how traffic emitted size resolved UFP dilute and mix with the background UFP using the micro scale model ENVI-met and to evaluate the importance of deposition and coagulation on the UFP size distributions. The concentration of particles in the nucleation mode largely decreased than the concentration of particles in the Aitken mode due to the larger differences in concentrations between traffic emitted (high number concentration) and background nucleation mode particles (low number concentration). Depending on the traffic emissions and wind direction, background UFP concentration can increase from 23% (for SW wind) to 165% (for N wind) in the cases considered in this study. The background size distribution for SW wind remained mono-modal with a peak in Aitken mode while for N wind it was reshaped to a bi-modal with two peaks in nucleation and Aitken modes. This transformation was caused by the high number of particles in the nucleation mode advected from the boulevard with more than 50% contribution to the background nucleation mode. Particles in the accumulation mode did not show variance in space, because they are mainly present in the atmosphere as aged particles.

Once emitted in the atmosphere the ultrafine particles are subject to transformation processes like dilution, deposition and coagulation. In our simulations, dilution was found to be more important for the UFP concentration than removal from deposition and coagulation and the overall effect of deposition and coagulation is negligible. Deposition was found to be faster than coagulation in removing particles in the smallest size bin (5 nm, up to 8%). Coagulation has negligible impact on the UFP concentration (total number and size distribution) even for the very high number concentration simulated on the boulevard.

So far this is one of the few studies that can model UFP – total number of particles and size distributions altogether in real world environment. With the implementation and integration of knowledge in ENVI-met (UFP traffic emissions, aerosol dynamics, UFP size distribution) the UFP dispersion can be further investigated, e.g., for different emission scenarios, different meteorological conditions, different complexities of the built up area (buildings, vegetation, etc.). However, extensive measurement campaigns in street to neighbourhood scale devoted to both horizontal and vertical UFP size distributions are a necessary step for verification and model assessment.

## ACKNOWLEDGMENTS

This work and I. Nikolova are supported by VITO N.V.

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Received for review, June 27, 2013 Accepted, August 20, 2013