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# Non-exhaust particle emissions under various driving conditions: Implications for sustainable mobility

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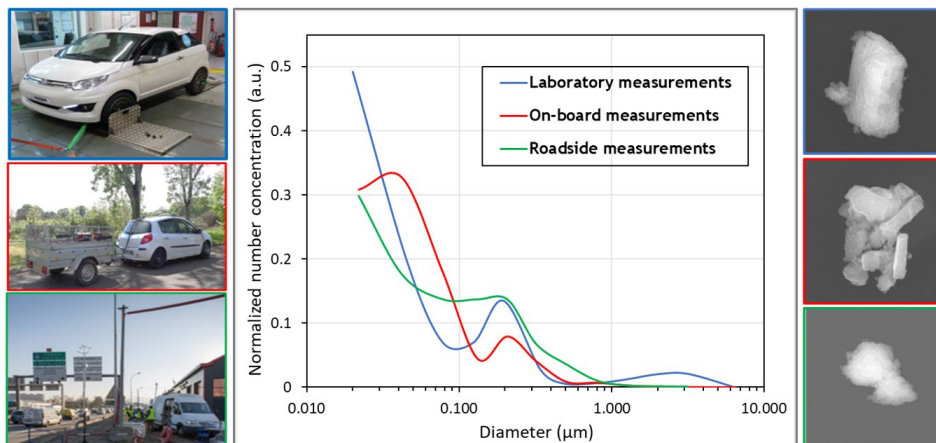
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**Keywords:** Non-exhaust emissions; brake wear particles; tire-road contact particles; size distribution; chemical composition; air quality

## Highlights

- The mass of non-exhaust particles (NEP) is distributed between the accumulation and coarse modes.
- The number concentration of NEP is shared between the nucleation and accumulation modes.
- Brake wear particle emissions are highly dependent on brake pad temperature.
- Tire-road contact emissions are determined by vehicle speed, acceleration and deceleration.
- The chemical composition and shapes of non-exhaust particles differ according to the size modes.

## Graphical abstract



26 **Abstract**

27 Non-exhaust particle (NEP) emissions from road traffic contribute significantly to Particulate  
28 Matter (PM) pollution in urban areas. The primary objective herein is to develop the  
29 knowledge required to move toward more sustainable mobility. NEP emissions are studied by  
30 means of complementary experiments on chassis dynamometers, on test tracks and at the  
31 roadside. Laboratory tests demonstrate that brake wear particles (BWP) emissions can change  
32 with braking force and frequency. A brake pad temperature threshold exists, above which the  
33 rate of ultrafine particle emissions is quite high. Below this threshold, the BWP emissions are  
34 dominant in the accumulation and coarse modes. Test track measurements have  
35 demonstrated that tire-road contact particle (TRCP) emissions considerably modify the  
36 atmospheric PM background especially for the supermicron fraction. Their number size  
37 distribution highlighted an ultrafine and accumulation modes centered at about 40 nm and  
38 200 nm, respectively. The TRCP level increases with vehicle speed and during the acceleration  
39 and deceleration phases. Roadside measurements in the urban environment confirm the  
40 presence of NEP in significant proportions, in both the accumulation and coarse modes. The  
41 chemical composition of NEP differs depending on the size mode: BWP mainly stem from the  
42 degradation of brake pad lining materials, while TRCP are a mixture of tire tread wear and re-  
43 suspended dust. The presence of Fe-rich particles nevertheless serves as a good indicator of  
44 their contribution to PM at the roadside. Lastly, in considering the parameters influencing NEP  
45 emissions, a series of recommendations are offered in order to achieve a more sustainable  
46 mobility.

47

48 **1. Introduction**

49 Traffic emissions have been responsible for air quality degradation for many years.  
50 Atmospheric Particulate Matter (PM) has been cited as causing some 430,000 premature  
51 deaths per year in Europe (European Environment Agency, 2015), and road traffic contributes  
52 significantly to their emissions, particularly in urban areas (Padoan and Amato, 2018).  
53 Ultrafine particles (in the nanometer size range) have particularly harmful effects on human  
54 health, by virtue of penetrating into the deep lung (Stone et al., 2017) and blood vascular  
55 system (Geiser and Kreyling, 2010), thereby increasing respiratory and cardiovascular  
56 morbidity, as well as lung cancer mortality through the mechanisms of oxidative stress and  
57 inflammation (Pope et al., 2002; Sun et al., 2010; Valavanidis et al., 2008).

58 Traffic-related particle emissions can be separated into exhaust emissions, resulting from  
59 incomplete fuel combustion and lubricant volatilization during the combustion procedure  
60 (Vouitsis et al., 2009), and non-exhaust emissions produced by brakes, tire and road surface  
61 wear, or by road dust resuspension (Thorpe and Harrison, 2008). Depending on the  
62 environment under consideration, exhaust and non-exhaust sources can contribute almost  
63 equally to total particle emissions from traffic (Harrison et al., 2001; Querol et al., 2004;  
64 Bukowiecki et al., 2010). However, as exhaust emissions controls have become more  
65 stringent, the relative contribution of non-exhaust emissions has increased commensurately  
66 (Kousoulidou et al., 2008; Van der Gon et al., 2013; Amato et al., 2014) and may now dominate  
67 in PM<sub>2.5</sub> (i.e. particles with an equivalent aerodynamic diameter < 2.5 μm) (Lawrence et al.,  
68 2016). These non-exhaust particles (NEP) have various sizes, shapes and chemical  
69 compositions (Grigoratos and Martini, 2014), depending on their origin (brake wear, tire wear,  
70 road surface wear, resuspension) or source-intrinsic parameters (e.g. brake and tire type,  
71 pavement texture).

72 The most abundant non-exhaust source is often assumed to be brake wear (Kwak *et al.*, 2013;  
73 Grigoratos and Martini, 2015), with a contribution of up to 55% by wt. of the non-exhaust  
74 PM<sub>10</sub> emissions near a major road in London (Harrison *et al.*, 2012). Brake wear particle (BWP)  
75 emissions depend on many factors, including: a) the type of brake pad (low metallic, semi-  
76 metallic, non-asbestos organic pads), more specifically the lining material; b) the type of brake  
77 assembly (drum brakes, disc brakes); and c) the vehicle operating conditions, e.g. initial speed,  
78 deceleration and braking force (Garg *et al.*, 2000; Hagino *et al.*, 2016, 2015; Iijima *et al.*, 2008,  
79 2007; Sanders *et al.*, 2003).

80 The emissions of tire-road contact particles (TRCP) constitutes another major NEP source  
81 (Kwak *et al.*, 2013; Grigoratos and Martini, 2015), including the resuspension of road dust as  
82 well as the wear of both tires and road surfacing materials. These emissions are likely to  
83 change as a result of many factors, such as meteorological and traffic conditions, ambient  
84 particulate pollution, driving style, variations in tire composition and texture, and road surface  
85 characteristics (Lowne, 1970; Etyemezian *et al.*, 2003; Dahl *et al.*, 2006; Gustafsson *et al.*,  
86 2008; Hussein *et al.*, 2008; Pirjola *et al.*, 2010; Sjödin *et al.*, 2010; Grigoratos *et al.*, 2018; Park  
87 *et al.*, 2018).

88 All these NEP are found not only in the coarse and fine fractions, but also in the ultrafine size  
89 range (< 100 nm) (Dahl *et al.*, 2006; Mathissen *et al.*, 2011; Sjödin *et al.*, 2010), particularly at  
90 high braking forces (Garg *et al.*, 2000; Mathissen *et al.*, 2011; Wahlström *et al.*, 2010b). Several  
91 difficulties arise however when studying NEP due to a lack of standardized sampling  
92 procedures and measurement techniques, which leads to non-comparable results (Grigoratos  
93 and Martini, 2015). Moreover, many studies have focused on BWP emissions, whereas those  
94 targeting TRCP emissions are relatively scarce due to the high contribution of resuspended

95 road dust, hence complicating application of the measurement and sampling methodologies  
96 (Grigoratos and Martini, 2014; Foitzik et al., 2018).

97 In this context, our initial goal is to determine the physical and chemical nature of BWP and  
98 TRCP, by means of implementing similar sampling and measurement methodologies that yield  
99 comparable results on the coarse ( $1\ \mu\text{m} < d < 10\ \mu\text{m}$ ), fine ( $100\ \text{nm} < d < 1\ \mu\text{m}$ ) and ultrafine  
100 ( $d < 100\ \text{nm}$ ) fractions. NEP emissions can be measured in different ways, namely: in the  
101 laboratory under controlled testing conditions (Pelkmans and Debal, 2006; Hagino *et al.*, 2016;  
102 Park *et al.*, 2018), on-board under semi-controlled testing conditions, and along the roadside  
103 by integrating the PM emissions from a wide array of vehicles and under different traffic  
104 conditions (Kwak *et al.*, 2014; Q. Zhang *et al.*, 2017). An original aspect of this work is studying  
105 NEP emissions under these three test conditions so as to draw conclusions on particle  
106 identification in various environments. BWP emissions have been studied by laboratory tests  
107 conducted on a chassis dynamometer using a common car of the French fleet, which entailed  
108 extracting new braking profiles from the ARTEMIS European driving cycles (André, 2004) and  
109 considering different parameters like vehicle speed and brake pad temperature. On-board  
110 measurements have been carried out on a test track to investigate TRCP emissions collected  
111 at the rear of a wheel, in considering a range of driving conditions and distances from the  
112 emission area. Lastly, roadside measurements, along a major French highway, allowed  
113 characterizing NEP in an actual urban environment and comparing them with previous  
114 findings under controlled and semi-controlled test conditions. In conclusion, through similar  
115 sampling / measurement methods and complementary test conditions, the factors influencing  
116 NEP characteristics are discussed and recommendations proposed in order to improve road  
117 mobility within the framework of sustainable development.

118

## 119 **2. Materials and methods**

### 120 2.1 Instrumentation

121 Particle size distributions were acquired by means of real-time measurements using three  
122 different instruments: an Electrical Low Pressure Impactor (ELPI™), a Fast Mobility Particulate  
123 Sizer (FMPS™), and an Optical Particle Counter (OPC Grimm™). The data are presented in this  
124 study as concentrations normalized by the integral of the particle counts. The ELPI™ (Dekati™,  
125 ref. Classic ELPI™) measures the number size distribution for particles with an aerodynamic  
126 diameter between 7 nm and 10 µm, at a time step of 1 s. This device operates at a flow rate  
127 of 10 L.min<sup>-1</sup>, with 12 size channels. For additional measurements of the ultrafine fraction (<  
128 100 nm), the FMPS™ (TSI™, ref. 3091) was used to measure electrical mobility diameters  
129 between 5.6 nm and 560 nm, at a time resolution of 1 s and a 32-channel size resolution; this  
130 instrument is equipped with a cyclone head (50% cut-off aerodynamic diameter = 1 µm) and  
131 operates at a sampled air flow rate of 10 L.min<sup>-1</sup> and a sheath air flow rate of 40 L.min<sup>-1</sup>. It  
132 relies on an electrical mobility measurement technique associated with a low-noise  
133 electrometer for particle detection. For these two particle sizers, mass size distributions are  
134 calculated from the number size distribution, in considering the particles to be spheres with a  
135 density of 1 g.cm<sup>-3</sup>, not so far from the effective density of brake wear particles measured by  
136 Nosko and Olofsson (2017). Both devices had previously been calibrated by their  
137 manufacturers, and the similarity between normalized size distributions measured in parallel  
138 has been verified (Figure S1). The main modes are well coinciding for the two measurements  
139 although there is an apparent overestimation of particle counts in the nano-size range of ELPI,  
140 and therefore an underestimation of larger ones, which has already been reported and  
141 attributed to particle bounce (Virtanen et al., 2010; Leskinen et al., 2012). An Optical Particle  
142 Counter (OPC Grimm™, ref. EDM 1.108) is introduced to probe, on a complementary basis,

143 the accumulation and coarse mode particles. This device outputs real-time measurements of  
144 particles with an optical diameter lying between 0.35 and 22.5  $\mu\text{m}$ , 15 size channels, a flow  
145 rate of 1.2  $\text{L}\cdot\text{min}^{-1}$  and a step time of 6 s. The analyzer had been calibrated by its manufacturer  
146 using dolomite dust. Only number concentrations data from this instrumentation were used  
147 in this study.

148 For the chemical characterization step, atmospheric particles were collected on nickel  
149 Transmission Electron Microscopy (TEM) grids using a 3-stage cascade impactor (air flow: 28  
150  $\text{L}\cdot\text{min}^{-1}$ ) with nominal cut-off sizes (i.e. aerodynamic diameters at 50% of collection efficiency)  
151 of 10, 1.0 and 0.1  $\mu\text{m}$ . Individual particle analyses were performed by Scanning Electron  
152 Microscopy with Energy-Dispersive X-ray spectrometry acquisition (SEM-EDX). Several  
153 hundred particles per impaction stage were detected and analyzed using a JEOL 7100F FEG-  
154 SEM equipped with a transmitted electron detector and three ultrathin-window (30  $\text{mm}^2$ )  
155 energy-dispersive X-ray detectors (Bruker XFlash 6/30), enabling the analysis of elements with  
156 an atomic number higher than boron ( $Z \geq 5$ ). Volatile and semi-volatile particles are likely to  
157 vaporize in the high vacuum environment of the SEM and are not observed. Therefore, the  
158 data from the SEM-EDX are only for non-volatile metallic constituents. Spectral acquisitions  
159 were performed at 15 kV, 300 pA for 20 s. The resulting large databases (particle size,  
160 morphological parameters *i.e.* particle projected area and Feret diameters, and elemental  
161 composition) were then statistically analyzed by means of Hierarchical Ascending  
162 Classification (HCA), using the FactoMineR software, clustering particles with similar  
163 characteristics, *i.e.* of comparable origin.

## 164 2.2 Sampling and measurement strategy



165 BWP emissions were studied in the laboratory on a chassis dynamometer (presented in  
166 Supplementary Information - Figure S2). Measurements and sampling were conducted on the  
167 side of the vehicle, about 8 cm from the wheel, using a conical sampling head (46 cm in  
168 diameter) centered on the wheel rim. To obtain a cooling pattern similar to real-world  
169 conditions, the engine compartment was cooled by a blower discharging a large-diameter  
170 stream of air toward the vehicle, with a flow rate proportional to the speed of the roller.  
171 Considering this sampling design, collection aspiration efficiencies ( $\eta$ ) were calculated using  
172 an empirical approach developed by (Baron and Willeke, 2001), in a turbulent regime  
173 ( $Re > 4 \times 10^3$ ) and where the sampling inlet is oriented at an angle  $\theta$  with respect to the air  
174 streamlines, using Equation 1:

$$175 \quad \eta = 1 + \left[ \left( \frac{U_0}{U} \right) \cos\theta - 1 \right] \left[ 3Stk \sqrt{\frac{U}{U_0}} \right] \quad (Equation1)$$

176 Where  $U$  = inlet velocity,  $U_0$  = flow velocity in the free-stream,  $Stk$  = Stokes number (calculated  
177 for a particle density of  $1 \text{g.cm}^{-3}$ ) and  $\theta$  = angle between the sampling inlet and the air  
178 streamlines ( $\theta = 53^\circ$  in our case). The calculated aspiration efficiencies (Table S 1) are higher  
179 than 90% whatever the particle size and the simulated vehicle speed. The ELPI is directly  
180 connected in the continuity of the sampling head outlet by an 80 cm antistatic tube, without  
181 bend, so the loss of particles during this path between the sampling head and the ELPI is  
182 negligible. In parallel, the FMPS is connected to the side of the sampling head, using two  $90^\circ$   
183 bends. Nevertheless, the fraction penetration in the bent section calculated according to  
184 Baron and Willeke (2001) is higher than 99% for particle diameters smaller than 300 nm (high  
185 detection limit of FMPS). In conclusion, the collection efficiency of BWP is higher than 90%  
186 whatever the particle size ( $< 10 \mu\text{m}$ ) and the particle concentrations were then not corrected  
187 for this loss.

188 To avoid contamination from exhaust emissions, the emissions were extracted at the tailpipe  
189 and evacuated to the outside after filtration. Three braking profiles, derived from the ARTEMIS  
190 (Assessment and Reliability of Transport Emission Models and Inventory Systems) driving  
191 cycles (André, 2004), were created to facilitate the qualitative characterization of brake wear  
192 particle emissions: urban (maximum speed = 50 km.h<sup>-1</sup>), suburban (maximum speed = 70 km.h<sup>-1</sup>),  
193 and highway (maximum speed = 120 km.h<sup>-1</sup>). These profiles were designed to be  
194 representative of actual driving conditions in Europe (André, 2004), yet with the high braking  
195 frequencies necessary to generate a sufficient amount of braking particles for sampling and  
196 analysis (Khardi and Nuel, 2017). They were performed by a same person. For these  
197 experiments, we used a light-duty vehicle (Renault Clio III, 1.6 i 16V, 111 hp, one of the most  
198 widely sold models in France) which was equipped with semi-metallic brake pads (TRW 8200  
199 432 336/ 32 336188 PES) and cast iron discs. Brake discs are traditionally made of grey cast  
200 iron with a pearlitic matrix (Cueva et al., 2003). Semi-metallic brake pads were purchased on  
201 the mass market and correspond to an intermediate price range. They are available in most  
202 EU countries (MSR, 2019). The brake temperature was monitored using an infrared  
203 temperature sensor (Texense, Ref: IRN2-V800-5-V2) positioned directly in front of the brake  
204 pads, 3 cm from the pad/disc contact surface.

205 TRCP emissions (including the re-suspension of road dust and wear of both tires and the road  
206 surface) were studied under semi-controlled conditions on a test track (2.3 km long; max.  
207 speed: 140 km.h<sup>-1</sup>). The road surface of the test track is consisting of semi-coarse (0/10)  
208 asphalt concrete with medium macrotexture (0.82-1.25 mm texture depth) and microtexture  
209 (skid resistance of 49.1-56.1) levels. The constituent rocks of the aggregates used in the  
210 asphalt concrete include both igneous rocks of felsic composition (rhyolite) and  
211 metamorphosed igneous rocks (gneiss). The chemical composition of these rocks is typically

212 70-77% silica ( $\text{SiO}_2$ ), 11-13% alumina ( $\text{Al}_2\text{O}_3$ ) and K, Na, Ca Fe and Mg as minor elements. The  
213 tests were conducted under dry weather conditions ( $T = 20\text{-}25^\circ\text{C}$  with 50%-80% humidity) and  
214 without significant wind ( $< 20 \text{ km}\cdot\text{h}^{-1}$ ). To preserve the same driving behavior throughout the  
215 testing campaign, the same pilot / co-pilot team was used. Vehicle data (speed, speed  
216 variations, engine parameters, etc.) were recorded at a frequency of 10 Hz regardless of  
217 driving conditions. A trailer containing the scientific instrumentation, including two power  
218 generators, was hitched. The inlet of the TRCP sampling probe was placed in the axis of the  
219 wheel 15 cm behind the tire and 7 cm from the ground (Figure S2).

220 As for the laboratory experiments on a chassis dynamometer, the collection aspiration  
221 efficiencies ( $\eta$ ) were calculated using an empirical approach developed by (Baron and Willeke,  
222 2001) using Equation 1 ( $\theta = 75^\circ$  in this case). The calculated aspiration efficiencies (Table S 2)  
223 are close to 100% for submicron particles (i.e. for the large majority of particles by number)  
224 whatever the speed of the vehicle (including when the vehicle is still for background  
225 measurement). For coarser particles (5-10 $\mu\text{m}$ ), these efficiencies are slightly less good,  
226 especially for 10 $\mu\text{m}$  particles (65.2% in average). However, the relative evolution of this  
227 efficiency for a particle size as a function of vehicle speed is  $\approx 10\%$ , which is relatively low, that  
228 is why experimental particle concentrations have not been corrected for the aspiration  
229 efficiency. The influence of tubing on measured number concentrations was investigated prior  
230 to the sampling campaigns. The tubing used to convoy particles is a 5m long antistatic hose  
231 with an inner diameter of 8 mm. The data showed that the particle loss amounts to 1.5%/m  
232 and that it is homogeneously distributed between fine and coarse particles (between <1% and  
233 3% variation in the ratio between fine and coarse concentrations). Therefore, the particle  
234 concentrations were not corrected for this loss.

235 Background concentrations were obtained when the vehicle was idle, facing the wind. To  
236 assess NEP dispersion, measurements were also performed at the edge of the test track using  
237 the same instrumentation. The sampling points were located at 0.45 and 3.0 m from the  
238 driving lane and 0.45 m from the ground. The vehicle was fitted with summer tires (Michelin™  
239 Energy Saver, 185/60 R 15 88H) purchased in the mass market. They have asymmetrical  
240 structures with three straight grooves that run around the center of the silica-based rubber  
241 tread. The elemental composition of the tire surface essentially consisted of C and O. Si, S, Zn,  
242 Fe and Ca were present as other major elements, i.e. ca. (in wt.%): 14%, 5%, 4%, 2% and 2%  
243 of the remaining, respectively. Al, Na, Cl, K and Mg were present as trace elements (i.e. less  
244 than 2wt.% of the total, once C and O have been discarded) or concentrated in localized areas.  
245 The tire surface was mostly amorphous with Si or Fe-enriched spots or inclusions. The shapes  
246 of inclusions were varied, ranging from spherical to cylindrical or highly amorphous.

247 Additional measurements and sampling were carried out at the roadside, along the A43  
248 motorway near Lyon, France. To provide a representative braking situation, the measurement  
249 site was located at the "Saint Quentin Fallavier" toll plaza, offering a traffic-influenced site  
250 (approx. 81,000 vehicles/day) exempt from industrial emissions with an urban atmospheric  
251 background. The instrumentation was set up in March 2017, downwind a few meters from the  
252 road, under the direct influence of road traffic emissions. Sampling and measurement heads  
253 were installed at the top of a 2-m high mast, so as to represent the air inhaled by humans.  
254 During this measurement campaign, the same instruments and sampling techniques used  
255 during laboratory and test track experiments were implemented, thus yielding comparable  
256 results.

257

### 258 **3. Results and discussion**

## 259 3.1 Brake wear particle (BWP) emissions

### 260 3.1.1 Particle size distribution, shape and chemical composition

261 The BWP size distribution is variable and may depend on many factors. The effect of repetitive  
262 braking with the same force during urban braking profiles was investigated first. Figure 1  
263 shows the evolution of this number size distribution during two successive urban profiles  
264 (about 900 seconds of continuous braking events). During the first profile (Urban 1 section), a  
265 bimodal distribution is observed with an accumulation peak centered at approx. 200 nm and  
266 a coarse peak at 2  $\mu\text{m}$ . The accumulation mode is most certainly generated by mechanical  
267 friction, while coagulation processes result in the coarse mode formation. Consequently, this  
268 coarse mode is in all likelihood essentially formed by agglomerated wear particles. As braking  
269 events accumulate, the particle size distribution evolves into an additional emission of  
270 ultrafine particles (Figure 1, Urban 2.A section) and ultimately results at the end of the second  
271 profile (Urban 2.B section) in a unimodal distribution with a peak at a diameter less than 30  
272 nm. Nanoparticle emissions were already observed when increasing the braking force at high  
273 vehicle speeds (Namgung *et al.*, 2016, 2017), but not with a constant braking force.  
274 Continuous braking events produce an increase in the friction surface temperature, as  
275 indicated by the increase in brake pad temperature (Figure 1). At a threshold temperature  
276 (about 250°C), the dual processes of brake pad thermal degradation and volatilization occur,  
277 and it can reasonably be assumed that this sequence leads to the abundant formation of  
278 nanoparticles by nucleation and condensation processes. This threshold temperature,  
279 associated with a significant change in BWP size distribution, has been confirmed for the three  
280 braking profiles studied in the next section and most likely depends on both the brake pad  
281 characteristics and temperature measurement techniques (Nosko and Olofsson, 2017).

282 The chemical composition of braking systems provides a good indication of the origin and  
283 formation processes of all these brake wear particles. Brake pads have a complex chemical  
284 composition that drastically varies depending on market price and manufacturer (Von Uexküll  
285 et al., 2005). The great diversity of pad compositions has already been highlighted in the  
286 literature, although certain elements considered as tracers for brake wear such as iron, copper  
287 or barium are more often mentioned (Garg et al., 2000; Sanders et al., 2003; Birmili et al.,  
288 2006; Thorpe and Harrison, 2008; Wahlström et al., 2010a; Pant and Harrison, 2013;  
289 Grigoratos and Martini, 2014; Hagino et al., 2016). Elemental maps of the brake pads used in  
290 our experiments have underscored the heterogeneous elemental distribution at the  
291 micrometric scale (Figure S3): some areas feature iron-rich components, while others are  
292 mainly composed of carbonaceous materials or mixed carbonaceous/metal agglomerates (C,  
293 O, Al, Si, S, Fe, Cu, etc.). This elemental distribution heterogeneity within the pad will probably  
294 generate a wide diversity of BWP, depending on the formation mechanism and pad area  
295 involved.

296 The cluster analyses from SEM-EDX data result in 7 clusters for BWP, whose names start by  
297 BW (Table 1). As expected, the individual BWP analysis shows a great diversity in chemical  
298 composition depending on the size modes considered. For the coarse mode, most BWP (BW-  
299 C3 type, with a relative abundance of P = 76%) form a complex mixture of carbon, oxygen and  
300 iron, with copper also being contained to a lesser extent and aluminum and silicon as minor  
301 elements. Generated by friction mechanisms and aggregation processes, these particles  
302 display Cu contents comparable to brake pads (i.e. 6.4% vs. 5.3%, respectively) as well as an  
303 agglomerated shape (see Figure 2). 17% of coarse particles (BW-C4 type) are iron-rich but do  
304 not contain copper. The absence of iron concomitants coupled with a carbon deficit would  
305 suggest disc wear as their likely source. Lastly, a small proportion of particles from this coarse

306 mode is carbonaceous particles also containing both iron and copper (BW-C1, P = 5%), or  
307 particles containing in Al (BW-C2, P = 2%). This would reflect either an origin to the carbon-  
308 based materials of the brake pads (including graphite, rubber and organic fibers) or, more  
309 rarely, vermiculite and other inclusions enriched with aluminum and iron. For the  
310 accumulation mode, most BWP (BW-A2 and BW-A3, P = 71%) are carbonaceous particles also  
311 containing iron to a varying extent (10.3 to 23.4 wt. %) and copper in a less extent. They also  
312 contain aluminum and silicon as minor elements. Their chemical compositions, which are  
313 quite similar to those of the brake pads introduced, exclude a brake disc origin and are nearly  
314 exclusively composed of iron. In addition, 29% of BWP from this accumulation mode (BW-A1,  
315 Table 1) are carbonaceous, while containing nitrogen and sulfur as minor elements. Mainly  
316 associated with a spherical shape (Figure 2), they probably stem from the degradation of the  
317 carbonaceous areas of brake pads. BWP of the nucleation mode (< 100 nm), formed from  
318 gaseous precursors, are too small to emit a signal detectable by SEM-EDX; however, when  
319 considering the chemical composition of brake pads and discs, it can reasonably be assumed  
320 that the majority of these BWP are carbonaceous, with carbon being among the less refractory  
321 elements in braking systems. A significant fraction of these nanometric particles might also be  
322 metal-rich (Liati *et al.*, 2019).

323 All these data from a vehicle equipped with semi-metallic brake pads clearly show that BWP  
324 are primarily the result of brake pad degradation (*i.e.* chemical and mechanical breakdowns)  
325 and not due to disc wear, except for a small fraction (17%) of coarse particles. Most BWP  
326 contain Fe (Table 1), hence iron, as a major element at the particle scale, can be considered  
327 as a tracer for BWP in the absence of other anthropogenic sources for this element. In the  
328 case of semi-metallic and non-asbestos organic brake linings, iron powder and steel wool are  
329 the major sources of iron (Thorpe and Harrison, 2008). For vehicles equipped with low-

330 metallic brake pads, the brake disc or drum may be the main contributors of Fe-containing  
331 particle emissions (Sanders *et al.*, 2003). In this latter case, Hulskotte *et al.* (2014) found that  
332 70% of brake wear originates from the discs and only 30% from brake pads. Several other  
333 elements are proposed in the literature as tracers for BWP, e.g. antimony, barium, lead or zinc  
334 (Iijima *et al.*, 2007; Charron *et al.*, 2019). Their identification requires more sensitive yet global  
335 analytical techniques, which prove to be ineffective for individual particle analysis.

### 336 337 3.1.2 Influence of braking force and brake pad temperature on BWP emissions

338 The effect of braking intensity on BWP emissions has been studied using three driving profiles  
339 that differ according to the braking forces applied by the driver (urban, suburban and highway,  
340 see Figure S4), associated with maximum speeds of 50, 90 and 120 km.h<sup>-1</sup>, respectively.  
341 Figures 3a, 3b and 3c show the temporal evolution of the size distribution of BWP emissions  
342 across various time intervals after the starting point of each test profile. These real-time  
343 measurements of number concentrations indicate that BWP emissions are not systematic for  
344 each brake use, and peak emissions are intermittently observed. PM resulting from brake  
345 wear likely occurs mainly as condensed nuclei and growing particles that partially stick to the  
346 available surfaces, including the disc (Augsburg *et al.*, 2011). This freshly-generated fraction  
347 could either be discharged by the air stream used for cooling the braking system or trapped  
348 for an indefinite period of time, thus explaining sporadic separation from the braking system  
349 surface. Regardless of braking profile, the BWP size distribution is similar with a highly  
350 dominant mode centered at 200 nm and a second highly tenuous mode centered at 10 nm.  
351 Nevertheless, the BWP concentration clearly depends on the braking profile being applied. On  
352 average, between 200 and 300 s after the braking profile starting point, number  
353 concentrations of  $2.7 \cdot 10^3$ ,  $2.0 \cdot 10^4$ , and  $1.0 \cdot 10^5$  cm<sup>-3</sup> were observed for the urban, suburban



354 and highway profiles, respectively. In sum, stronger braking forces lead to the generation of  
355 BWP with the same mean size (near 200 nm), yet in higher quantities.

356 Brake pad temperatures corresponding to the size distributions presented in Figures 3a, 3b  
357 and 3c are on average 120°, 170° and 220°C, respectively. These temperatures, typically  
358 encountered during normal braking under actual driving conditions (Garg *et al.*, 2000), lie  
359 below the point where nanometric particle generation is observed. However, if the number  
360 of times applying the brakes were to be drastically increased, like during the highway type  
361 experiment, then the temperature would increase to 400°C after 500-600 s and the BWP size  
362 distribution would change significantly (Figure 3d), with a strong emission of nanoparticles  
363 centered around 10 nm. Previous studies also reported that the size distribution when braking  
364 occurs at a vehicle speed of around 120 km.h<sup>-1</sup> displays a maximum at approx. 11 nm  
365 (Mathissen *et al.*, 2011; Kwak *et al.*, 2014). Although such temperatures are not commonly  
366 observed during actual driving, these results do confirm our previous assumption of a brake  
367 pad temperature threshold above which ultrafine BWP (core mode centered at about 10 nm)  
368 are generated in great numbers regardless of the braking force. This threshold differs  
369 according to the braking profile under consideration and, therefore, according to the braking  
370 forces applied: it equals approx. 250°, 300° and 350°C for urban, suburban and motorway  
371 profiles, respectively (Figure S5). Of course, these thresholds probably depend on the type and  
372 composition of the brake pads used. These values are therefore specific to a semi-metallic  
373 brake pad and with a temperature measurement methodology close to the one used here.

## 374 3.2 Tire-road contact particles (TRCP)

### 375 3.2.1 Particle size distribution, shape and chemical composition

376 The formation of ultrafine TRCP has already been reported in the literature, with diameters of  
377 30-60 nm and less than 30 nm for studded and non-studded tires, respectively (Dahl *et al.*,  
378 2006; Å. Sjödin *et al.*, 2010; Foitzik *et al.*, 2018), along with the emission of a coarse fraction  
379 (Kwak *et al.*, 2013; Å. Sjödin *et al.*, 2010; Hussein *et al.*, 2008). In this study, the vehicle used  
380 was fitted with summer tires. At a constant vehicle speed anywhere in the range from 50 to  
381 90 km.h<sup>-1</sup>, the TRCP number size distribution after background removal is bimodal (Fig. 4a),  
382 with a distinct ultrafine mode centered around 30-60 nm and a more minor accumulation  
383 mode centered around 200 nm. The mass size distribution is also bimodal, with an  
384 accumulation mode centered at 300 nm and a major coarse mode centered at 2-4 µm (Fig.  
385 4a). The particle diameters at which these emission modes are centered do not depend on  
386 vehicle speed, but their relative proportional values vary with it (Fig. 4b). The TRCP-to-  
387 background number concentration ratio is close to one for submicron particles (on average,  
388 1.44 and 1.65 for the 10 - 100 nm and 100 nm - 1 µm size ranges, respectively), and it only  
389 increases slightly with vehicle speed. These TRCP emissions consequently give rise to just a  
390 limited change in the submicron fraction of the atmospheric aerosol at the rear of the wheel  
391 for ambient concentrations commonly found in European urban environments (7,100  
392 particles.cm<sup>-3</sup> including a very large fraction of ultrafine particles and 25-30 µg.cm<sup>-3</sup> dominated  
393 by particles from the accumulation and coarse modes). In contrast, the TRCP-to-background  
394 number concentration ratio is higher for the 1-10 µm size range (Fig. 4b) and increases  
395 substantially with vehicle speed, up to approx. 5.7 at 90 km.h<sup>-1</sup>. This finding clearly highlights  
396 an increase in the proportion of coarse TRCP at high speed, i.e. with more turbulent air  
397 streams and shear stresses developing at the tire-road pavement interface. A significant  
398 change thus occurs in the supermicron fraction of the wheel rear atmospheric aerosol due to  
399 TRCP emissions.

400 The evolution of the TRCP-to-background number concentration ratios vs. vehicle speed ( $\alpha$ ,  
401 in km.h<sup>-1</sup>) can be fit in the 0 - 90 km.h<sup>-1</sup> range by use of an exponential law, as follows:

$$402 \quad \frac{\text{TRCP}}{\text{Bkgd}} = 1 + \exp (A\alpha) \quad (\text{Equation2})$$

403 Where Bkgd is the background number concentration for the day of measurements and “A”  
404 an empirical proportionality factor. The following values with the associated coefficient of  
405 determination ( $r^2$ ) were obtained for the different size ranges:  $(A, r^2)_{10-100\text{nm}} = (0.0050, 0.92)$ ,  
406  $(A, r^2)_{100\text{nm}-1\mu\text{m}} = (0.0070, 0.93)$ ,  $(A, r^2)_{1-10\mu\text{m}} = (0.0204, 0.99)$ , the latter being semi-quantitative  
407 considering the lower collection efficiency of particles above 5 $\mu\text{m}$  (Table S 1). This result and  
408 previous research (Etyemezian et al., 2003; Dahl et al., 2006; Hussein et al., 2008; Gustafsson  
409 et al., 2008; Foitzik et al., 2018) serve to confirm that driving conditions, including speed,  
410 should not be ignored in the strategies implemented to reduce NEP emissions from road  
411 traffic.

412 The cluster analyses of data from SEM-EDX result in 8 clusters for TRCP, whose names start by  
413 TRC (Table 1). As observed for BWP, the TRCP chemical composition differs by size mode.  
414 Accumulation mode particles were mostly carbonaceous, containing silicon as a minor  
415 element (64% / TRC-A2). 18% of them (TRC-A1) were mixed carbonaceous-aluminosilicate  
416 particles and 19% (TRC-A3) were mixed carbonaceous/Fe-rich particles, which is consistent  
417 with a mixture of road surface dust and tire components. Carbon is the main component of  
418 asphalt binders but also of tire treads. Silicon, found in high quantities in some tire  
419 formulations, is used in tire reinforcing filler materials (up to 66% in superficial micrometric  
420 fibers). These materials also include iron and zinc as minor components (Adachi and Tainosho,  
421 2004; Camatini *et al.*, 2001; Gustafsson *et al.*, 2008). The relative stability of zinc content in

422 tires makes this trace element a common tracer of tire wear emissions (Councell *et al.*, 2004;  
423 Davis *et al.*, 2001), but it is not sufficiently present in the TRCP to be detectable at the particle  
424 scale by SEM-EDX. For the coarse mode, 40% of TRCP were mixed carbonaceous/salt  
425 (containing NaCl) particles, most likely re-suspended sea-salt particles (Table 1 / TRC-C5)  
426 which are expected due to the presence of the Atlantic Ocean coast 20 km southwest of the  
427 test track and predominant southwest winds during our experiments. Approximately 20% of  
428 TRCP were aluminosilicates containing K and Na as minor elements (TRC-C1), which are  
429 terrigenous particles stemming from dust resuspension. 14% of coarse TRCP (TRC-C4) consist  
430 of aluminosilicate particles that also contain Na, Mg, Ca and Fe. The iron content in 19% of  
431 TRCP (TRC-C3) (35.5 wt. %) is too high to correspond to mineral dust, hence these particles  
432 are probably iron-rich from brake or tire wear potentially mixed with terrigenous dust on the  
433 road surface (the iron content of the metal inclusions apparent on the surface of the tire and  
434 brake pads exceed 65% and 90% by mass, respectively). Pavements normally have a rocky  
435 ballast, which result in wear particles composed of minerals dominated by elements such as  
436 Si, Ca, K, Al and Fe (Lindgren, 1996) as well as other minor components like titanium (from <  
437 0.1% to 33%). All these observations suggest that a high proportion of coarse TRCP are re-  
438 suspended particles.

439

### 440 3.2.2 Influence of acceleration and deceleration on TRCP emissions

441 As traffic intensifies, both accelerations and decelerations can exert a significant impact on  
442 TRCP emissions. As such, strong TRCP concentrations were recorded for both the  
443 accumulation and coarse modes during acceleration/deceleration cycles (Figure 5). Although  
444 the increase in TRCP-to-background number concentration ratios was close to that during the  
445 mildest accelerations, these ratio values rapidly increased above 2.45 m.s<sup>-2</sup>: TRCP

446 concentrations measured at  $3.10 \text{ m.s}^{-2}$  exceeded by a factor of close to 12 those measured at  
447  $2.30 \text{ m.s}^{-2}$  (Figure 5). TRCP emissions were not constant during the accelerations: the strongest  
448 accelerations (i.e.  $> 2.50 \text{ m.s}^{-2}$ ) were actually characterized by an initial maximum in number  
449 concentrations containing all the previously identified modes (i.e. ultrafine, 55-60%;  
450 accumulation, 3-10%; and coarse, 10-15%), followed by a sudden and sharp increase in  
451 particles from the ultrafine mode (85-95% of total measured particles). This sequence resulted  
452 in significantly smaller mean diameters than those obtained for constant-speed driving or  
453 during small accelerations, i.e.  $< 100 \text{ nm}$  vs.  $150\text{-}340 \text{ nm}$ , respectively. The pronounced  
454 increase in ultrafine mode level probably accounted for differing generation mechanisms, e.g.  
455 tire wear at higher temperature or associated with a more intense road dust fragmentation.

456 For decelerations, the TRCP concentration increased with both braking intensity (Kim and Lee,  
457 2018; Lee *et al.*, 2013) and speed at the start of braking (i.e. with braking power) (Figure 5).  
458 For instance, at similar deceleration rates ( $6.9\text{-}7.0 \text{ m s}^{-2}$ ) experienced at initial speeds of  $50$   
459  $\text{km.h}^{-1}$  and  $110 \text{ km.h}^{-1}$ , the TRCP-to-background number concentration ratios calculated over  
460 the  $0.35\text{-}10 \mu\text{m}$  range rose by a factor of about 3, thus corresponding to a threefold increase  
461 in TRCP emissions since the background remains relatively constant during the tests. Unlike  
462 accelerations and despite the predominance of submicron particles, the proportion of coarse  
463 particles climbed significantly, from 16% to 33%.

464 Let's finally note that BWP emissions are most likely to account for a small proportion of TRCP  
465 concentrations: this is estimated from changes in the concentration ratio  $<1 \mu\text{m}$  to  $>1 \mu\text{m}$  to  
466 be around 10% between measurements at rear of the tire and a few centimeters downstream  
467 of the brake system. After emission, BWP are rapidly incorporated into the cloud of  
468 resuspended particles and then removed by the airflow downstream of the vehicle. However,

469 the calculated percentage is only a lower limit. It is, indeed, not currently possible to make a  
470 strict distinction between direct emissions of BWP and those resulting from resuspension.

471 In sum, compared to constant-speed traffic, accelerations and decelerations significantly raise  
472 TRCP and BWP emissions. Traffic fluidization should therefore be adopted as an imperative  
473 objective during measurement campaigns aimed at developing sustainable mobility.

474

### 475 3.2.3 Dispersion of TRCP at various distances from the driving lane

476 To assess the dispersion of BWP and TRCP, additional measurements were recorded at the  
477 edge of the test track, 0.45 and 3.0 m from the driving lane, thus from the TRCP measurement  
478 point when the vehicle passes in front of the roadside instrument. Measurements  
479 corresponding to the vehicle passing in front of the instrument (called "sample") were  
480 extracted and compared to the atmospheric background (i.e. measured near the test track  
481 when the vehicle is idle with its engine turned off). These "samples" incorporate the  
482 background, NEP (BWP+TRCP) and exhaust particle emissions. Interestingly, for ambient  
483 concentrations commonly found in European urban environments (i.e.  $7,100 \pm 800$  particles  
484  $\text{cm}^{-3}$ , or  $25\text{-}30 \mu\text{g m}^{-3}$ ), the sample-to-background concentration ratios recorded at 0.45 and  
485 3.0 m from the edge of the test track (Figure 6) were close to one another and did not vary  
486 significantly with vehicle speed. For the coarse fraction (1-10  $\mu\text{m}$ ), whose contribution of fresh  
487 exhaust emissions can be considered negligible in light of their submicron size (Li *et al.*, 2018;  
488 Lu *et al.*, 2012), NEP emissions accounted for an average increase of 55% and 15% in the  
489 number concentration of background particles at 0.45 m and 3.0 m, respectively. In  
490 considering that there is no BWP emission at constant speed (i.e. without brake application),  
491 the decrease in the NEP so in the TRCP concentrations observed between the rear of the tire  
492 (Figure 4) and 0.45 m from the driving lane (Figure 6) demonstrates here that the TRCP were

493 largely removed from the immediate vicinity of the driving lane (i.e. 0.45m), most likely  
494 through deposition of the coarser particles and rapid dilution in incoming air during turbulent  
495 dispersion. The decrease in TRCP number concentration with increasing distance from the  
496 road is more pronounced at higher speeds: for the 1-10  $\mu\text{m}$  fraction, the TRCP-to-background  
497 ratios were strongly correlated with vehicle speed (Figure 4), while the sample-to-background  
498 ratios at 0.45 m and 3.0 m from the driving lane remained for the most part unchanged (Figure  
499 6). This finding underscores the increased turbulence in the mixing of air masses around the  
500 vehicle, thus resulting in higher dilution and dispersion rates, which ultimately counteract the  
501 TRCP concentration step-up in back of the wheel with vehicle speed. TRCP emissions from a  
502 single vehicle do not therefore contribute to a greater increase in PM level within the roadside  
503 atmosphere. However, a constant traffic flow would promote chronic impregnation of the  
504 roadside atmosphere with freshly emitted TRCP and/or more evolved particles (i.e. mixtures  
505 of exhaust and non-exhaust particles that have undergone one or more deposition, alteration  
506 and re-suspension cycles).

507

### 508 3.3. Implications for air quality in urban environments

509 To the best of our knowledge, no studies have sought to compare the micro-characteristics of  
510 particles collected both in an urban environment and during laboratory experiments  
511 conducted on brake wear and tire-road contact emissions. Many studies however have  
512 focused on ambient PM measurements in the urban environment, close to roadways, in  
513 identifying potentially high particulate levels from road traffic. The contribution of non-  
514 exhaust emissions to roadside PM concentrations, as generally evaluated from trace metals,  
515 is highly variable according to the studies, from a few to several tens of percent (Cahill et al.,  
516 2016; Charron et al., 2019; Jeong et al., 2019; Lawrence et al., 2016). This variability can be

517 explained by the distance from the roadside, with a reduction of up to about 40% of the  
518 particle number concentration at a distance of 10 m, but more likely by the driving mode, local  
519 topography and weather conditions (Fujitani *et al.*, 2012; Kumar *et al.*, 2014; Oakes *et al.*,  
520 2016; X. Zhang *et al.*, 2017).

521 PM mass concentrations measured during our experiments at a toll plaza on a heavily-  
522 trafficked motorway, with stop-and-go traffic conditions, exhibit a large submicron particle  
523 fraction (PM<sub>1</sub> counts for 75% of PM<sub>10</sub>). The mean PM<sub>1</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> concentrations were  
524 of approx. 22.5  $\mu\text{g}\cdot\text{m}^{-3}$ , 26.7  $\mu\text{g}\cdot\text{m}^{-3}$  and 30  $\mu\text{g}\cdot\text{m}^{-3}$ , respectively. These values are close to the  
525 local urban background measured by the air quality monitoring network over the same period  
526 at about 20 km away (daily mean PM<sub>2.5</sub> and PM<sub>10</sub> values of 27  $\mu\text{g}\cdot\text{m}^{-3}$  and 33  $\mu\text{g}\cdot\text{m}^{-3}$ ,  
527 respectively). This finding demonstrates the relatively low impact of direct traffic emissions  
528 from the road adjacent to the measurement site on PM mass concentrations, as expected by  
529 our previous experiments on test tracks at variable distances from the driving lane (Section  
530 3.2.3.). Another explanation lies in both the width of the toll plaza (some 120 m for 32 gates)  
531 so the far distance between the emission and measurement points and the smooth  
532 topography with no buildings in the near field, which favors higher dispersion of local  
533 emissions. Roadside studies systematically showed a rapid decrease in pollutant levels with  
534 increasing distance from the roadside (Cahill *et al.*, 2016; Enroth *et al.*, 2016; Oakes *et al.*,  
535 2016). The concentration gradient is variable and depends strongly from the air mass  
536 dynamics. For example, Enroth *et al.* (2016) observed half-decreases in PM<sub>1</sub> concentration  
537 ranging from 3 m to 75 m from the highway depending on the environment considered, which  
538 can lead to roadside PM concentrations relatively close to those of the urban background.  
539 Thus, the PM collected here is considered to be representative of urban particulate matter,



540 whose overall emissions from the entire dense urban road network certainly make a  
541 significant contribution.

542 The number size distribution is bimodal (Figure S6) with one mode at a diameter less than 30  
543 nm (nucleation mode) and another at 100-200 nm (accumulation mode). The finer mode  
544 corresponds to nanoparticles freshly formed from gaseous precursors, generally mainly  
545 consisting of volatile organic, nitrogen and sulfur compounds; it may stem from regional  
546 particle nucleation but also from traffic exhaust emissions or else include NEP (Agudelo-  
547 Castañeda et al., 2018; Enroth et al., 2016; Jeong et al., 2015). The mass size distribution is  
548 also bimodal (Figure S6) with maximum concentration around 0.4-0.6  $\mu\text{m}$  and above 3  $\mu\text{m}$ .  
549 The coarse mode, which is more abundant than the finer one, may include a significant  
550 proportion of NEP, which notably would fit with the non-exhaust coarse particles observed  
551 during our laboratory and test track experiments (Figure 1 and Figure 4) and has been reported  
552 in the literature (Harrison *et al.*, 2012).

553 In recognizing the absence of industrial iron sources at the regional scale, the presence of Fe-  
554 rich particles is a good indicator of the contribution of non-exhaust emissions to ambient PM  
555 levels in the urban environment, as already observed (Achad et al., 2013; Bucko et al., 2011;  
556 Charron et al., 2019; Sanderson et al., 2016; Weinbruch et al., 2014). The proportion of Fe-  
557 containing particles at the studied site is relatively high, i.e. around 60% of analyzed particles  
558 for the coarse fraction (aerodynamic diameter > 1  $\mu\text{m}$ ). This proportion is lower (by about 10%  
559 among the analyzed particles) for the submicron fraction, which is essentially composed of  
560 carbonaceous particles. These Fe-containing particles are basically iron oxide particles  
561 enriched to a various degree in carbon (Table S2); most of them also contain silicon in low  
562 amounts, owing to the NEP collected from brake wear and tire-road contact during our

563 experiments (Table 1). About 58% of the coarse Fe-containing particles (FeR-C1) consist  
564 primarily of iron oxides, and only 23% of the coarse Fe-containing particles (FeR-C2) are  
565 aluminosilicates, enriched in Ca and Fe (Table S2). All these coarse Fe-containing particles are  
566 mostly angular and typically consist of aggregates, which suggests the presence of a friction  
567 production mechanism. In addition, when normalized to C content, the chemical composition  
568 of FeR-C1 and FeR-C2 particles approximates that of commercial brake pads with high (> 5%)  
569 and low (< 5%) Fe content, respectively. Ba and/or Sb, also present in brake emissions  
570 (Sanderson *et al.*, 2016; Gietl *et al.*, 2010; see Figure 7), are sometimes detected as minor  
571 elements. In sum, Fe is a ubiquitous element in urban areas and, besides local sources (traffic,  
572 home heating, waste incineration, etc.), the long-range transport of both mineral dust and  
573 anthropogenic particles may contribute to Fe concentrations at the urban scale, especially for  
574 fine and ultrafine particles. However, in considering Fe-rich particle abundance as an indicator  
575 of the presence of NEP in the coarse PM fraction, our results suggest that for these  
576 supermicron particles, non-exhaust emissions do make a significant contribution to PM levels  
577 in urban environments.

578

#### 579 **4. Conclusion and recommendations**

580 In conclusion, the testing of three distinct environments (controlled in the laboratory, semi-  
581 controlled with a test track and in an actual urban environment) using the same  
582 instrumentation, sampling protocol and data processing methodology has made it possible to  
583 obtain complementary results on NEP. To summarize, for urban and suburban driving  
584 conditions, the emission of ultrafine BWP (< 100 nm) is low in comparison with particles  
585 emitted in the 100-200 nm range (accumulation mode) and 2.5-4  $\mu\text{m}$  range (coarse mode) due  
586 to the limited increase in brake pad temperature. The elemental composition of these BWP

587 clearly indicates that over 70% of them are due to brake pad wear. Although tire and  
588 pavement wear generates ultrafine particles, it is mainly the supermicron fraction of TRCP  
589 emissions that alters the composition of the atmospheric background noise. Most TRCP  
590 associated with the accumulation mode stem from tire wear, whereas a large proportion of  
591 the coarse TRCP consist of re-suspended road dust, as well as more evolved particles  
592 originating from the tire, brake and road surface wear. BWP and TRCP generated under  
593 controlled and semi-controlled conditions have thus been extensively characterized;  
594 moreover, particles with similar characteristics (size, morphology and chemical composition)  
595 have been detected along the roadside, thereby highlighting the real impact of NEP on urban  
596 air quality.

597 Information gleaned on the key parameters controlling NEP emissions can be used to issue  
598 recommendations intended to reduce non-exhaust emissions and improve sustainable  
599 mobility. Among the parameters tested, BWP emissions and TRCP levels markedly increase  
600 with both speed and speed variations. Accordingly, limiting driving speeds to 70 km h<sup>-1</sup> and/or  
601 smoothing traffic flow would help reduce NEP emissions. This goal can be achieved without  
602 any excessive investment, simply by means of various strategies typically implemented to  
603 improve driving safety or reduce exhaust emissions (through speed limit monitoring, road  
604 space rationing, congestion pricing, etc.). Eco-driving practices, combined with a dedicated  
605 infrastructure that reasonably limits speed and both the intensity and frequency of speed  
606 variations, are particularly able to reduce NEP levels in the near-road field.

607 TRCP emissions are conditioned locally by the degree of contamination and chemical  
608 composition of the dust deposited on the pavement (including the recycling of exhaust and  
609 non-exhaust emissions). This approach necessitates on-board and roadside devices capable of

610 detecting situations prone to non-exhaust emissions and warning drivers to adapt their speed.  
611 Moreover, given that the chemical composition of NEP is correlated with the composition of  
612 both brake pads and tires, making progress in the composition of these pads and in tire tread  
613 toward less toxic and more environmentally-friendly materials would be a positive step.

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904

905 **List of Tables**

906

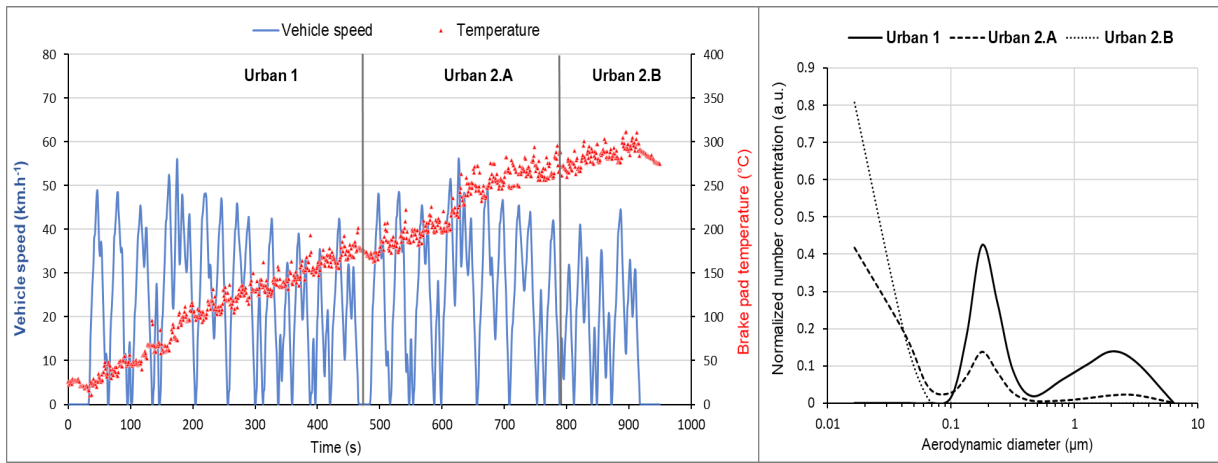
	Brake Wear (BW) particles							Tire-Road Contact (TRC) particles							
	Accumulation mode (A) (n=612)			Coarse mode (C) (n=195)				Accumulation mode (A) (n=274)			Coarse mode (C) (n=360)				
	BW-A1	BW-A2	BW-A3	BW-C1	BW-C2	BW-C3	BW-C4	TRC-A1	TRC-A2	TRC-A3	TRC-C1	TRC-C2	TRC-C3	TRC-C4	TRC-C5
	P=29%	P=25%	P=47%	P=5%	P=2%	P=76%	P=17%	P=18%	P=64%	P=19%	P=21%	P=6%	P=19%	P=14%	P=40%
<b>Dm</b>	0.16	0.21	0.52	2.16	2.62	2.61	2.58	0.64	0.37	0.87	1.95	1.42	1.76	1.78	1.48
<b>C</b>	76.8	64.1	41.5	54.7	18.1	21.1	17.1	39.9	69.6	37.2	33.9	47.7	29.6	40.1	45.4
<b>N</b>	2.3											15.2			2.9
<b>O</b>	18.1	22.4	28.5	32.6	35.7	29.7	26.6	43.9	27.2	29.1	45.0	30.8	29.8	39.7	23.1
<b>Na</b>											1.4		1.5	1.3	14.1
<b>Mg</b>														1.4	
<b>Al</b>		0.8	1.0		17.1	1.8		2.7			4.2			1.9	
<b>Si</b>		1.1	1.3	1.2		1.9	1.5	9.5	1.4	1.3	11.8	1.3	1.1	4.8	
<b>S</b>	1.8			1.8											
<b>Cl</b>															12.0
<b>K</b>											1.7				
<b>Ca</b>														4.6	
<b>Fe</b>		10.3	23.4	8.7	25.2	38.3	53.8	1.3		31.1		2.5	35.5	3.9	
<b>Cu</b>		1.2	3.6	0.8	3.2	6.4									

907

908 *Table 1: Mean diameter (Dm, in μm) and mean elemental composition (wt. %) for particles emitted from brake wear and*  
 909 *tire-road contact. The number of particles analyzed (n) by SEM-EDX and the relative proportion (P) of the various particle*  
 910 *types are indicated for each size mode. Only those elements with a mean concentration of at least 0.8% are reported.*

911 **List of figures**

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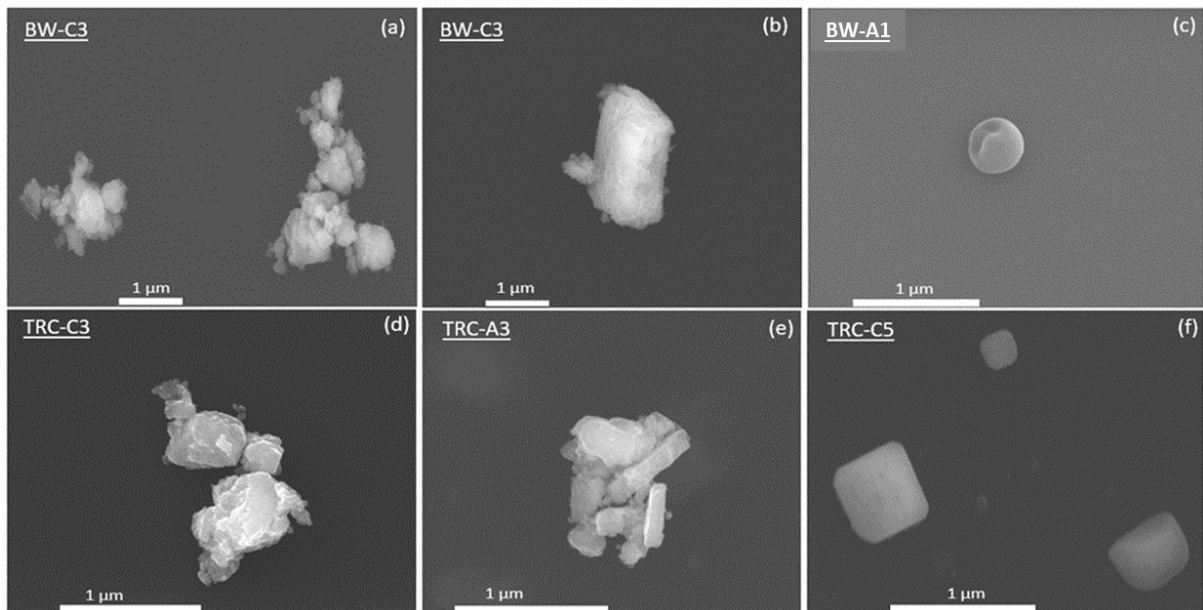


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914 *Figure 1: Evolution of brake pad temperature during two successive urban profiles (left) and mean number size distributions*  
 915 *of BWP after the removal of atmospheric background for the corresponding profile periods (right)*

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919 *Figure 2: Typical morphologies of brake wear (BW) and tire-road contact (TRC) particles: agglomerated (a), flaky (b),*  
 920 *spherical (c), agglomerated (d), agglomerated (e), angular (f). The identification of these particles (top left of each SEM-EDX*  
 921 *image) corresponds to the particle types in Table 1.*

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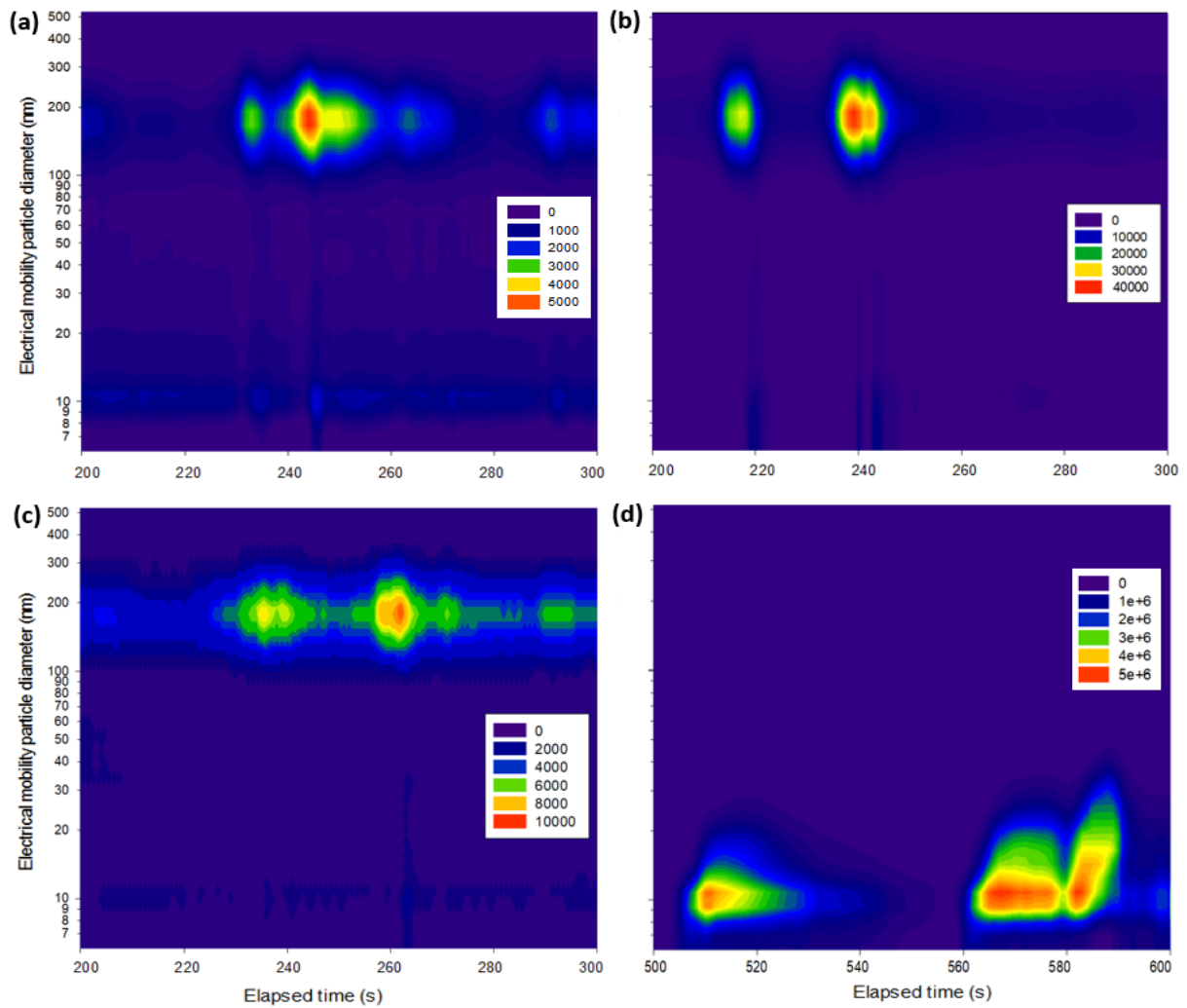
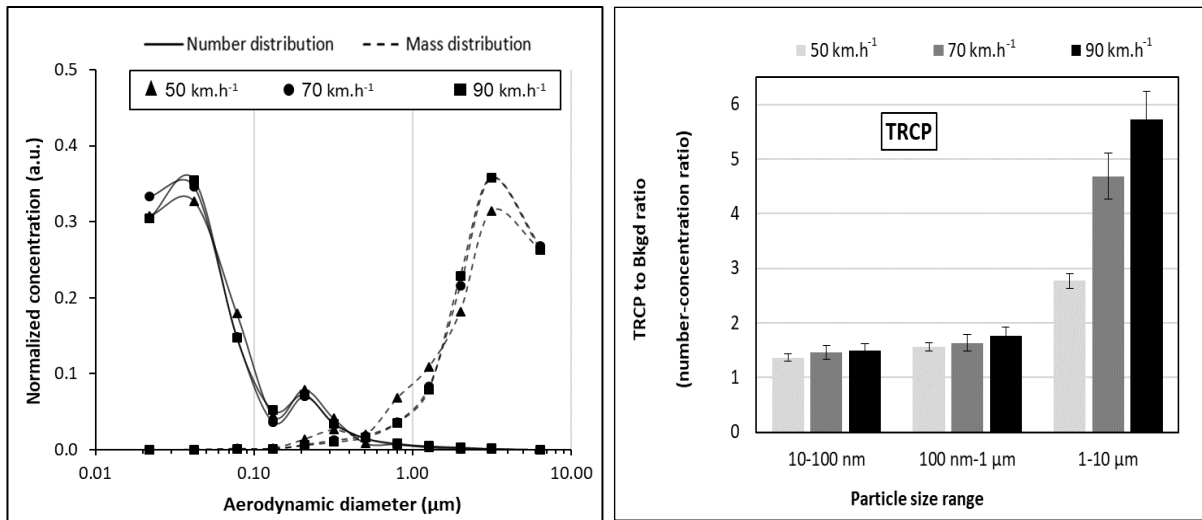


Figure 3: Temporal evolution of the size distribution of BWP emissions for the (a) urban, (b) suburban, (c) highway braking profiles 200 s after the profile starting point, and (d) for the highway braking profile 500 s after the starting point.

All concentrations are given in  $\#.\text{cm}^{-3}$ .

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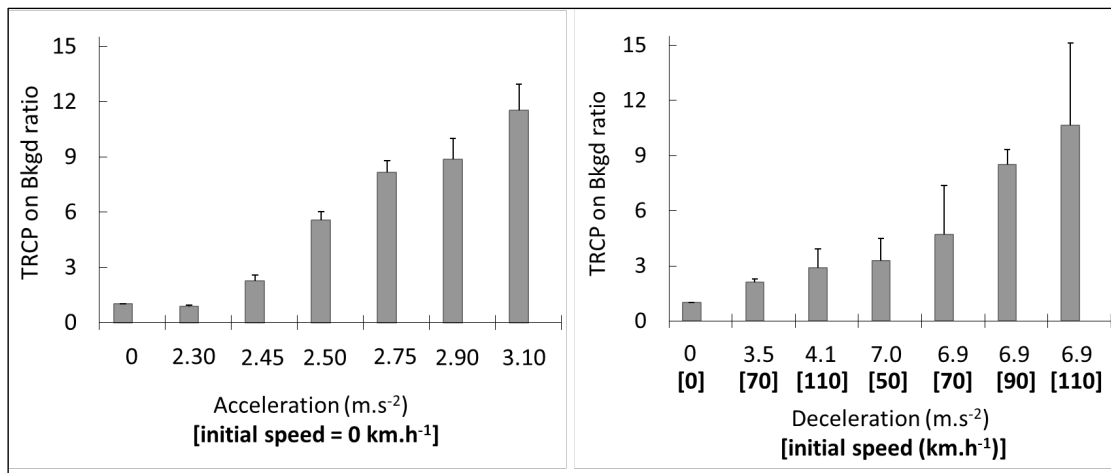


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936 *Figure 4: Normalized number and mass size distributions of TRCP after background removal (left) and TRCP-to-background*  
 937 *(Bkgd) concentration ratios (right) in considering the 0.01-10 μm particle size range (aerodynamic diameters). The error bars*  
 938 *account for standard errors calculated over the dataset from five successive tests.*

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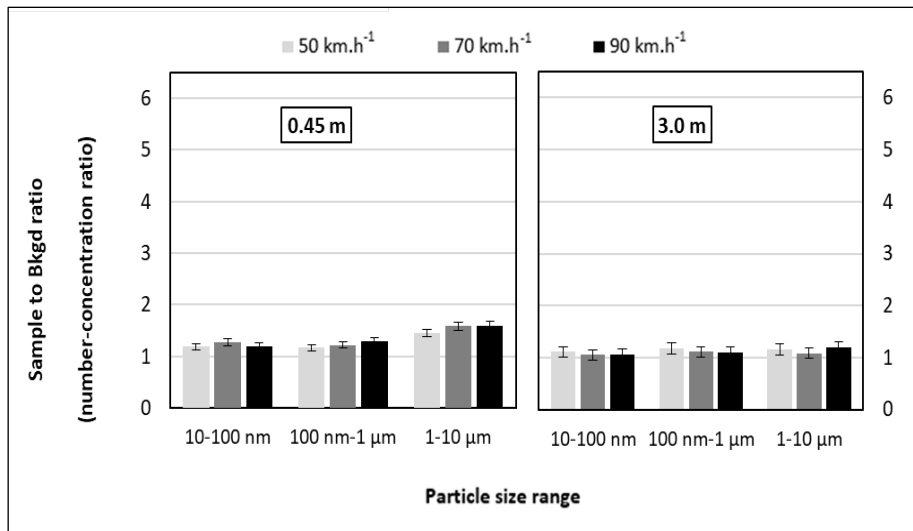


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942 *Figure 5: Evolution of TRCP-to-background (Bkgd) concentration ratios in considering the 0.35-10 μm size range of optical*  
 943 *diameter for increased acceleration (a) or deceleration (b) powers. The error bars account for standard errors calculated*  
 944 *over the dataset from five successive tests.*

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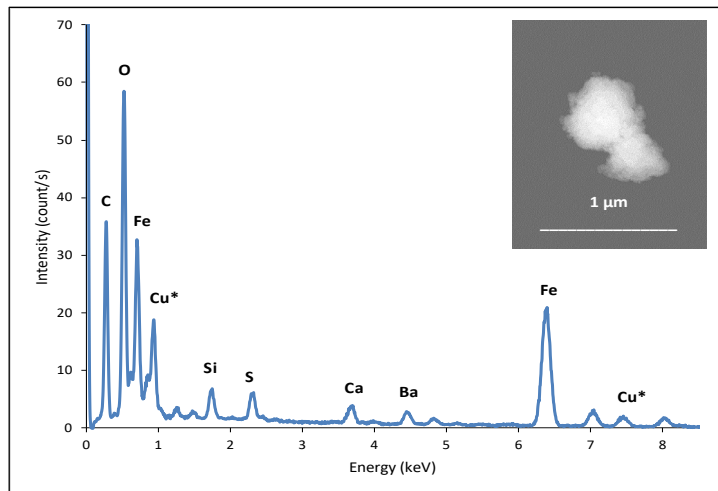


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948 *Figure 6: Sample-to-background (Bkgd) concentration ratios measured at the edge of the test track at an increasing distance*  
 949 *from the driving lane (0.45 and 3.0 m) in considering the 0.01-10 µm particle size range (aerodynamic diameters). The error*  
 950 *bars account for standard errors calculated over the dataset from five successive tests.*

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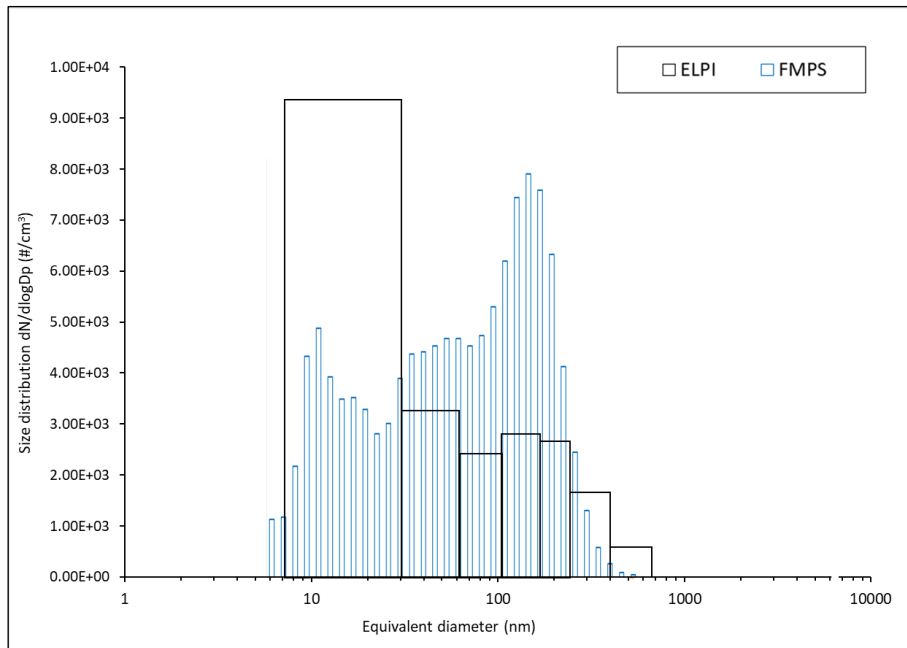
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954 *Figure 7: SEM image and EDX spectrum for a typical Fe-containing particle collected along the roadside and classified in the*  
 955 *FeR-A1 cluster (\*: Cu is from the grid holder)*

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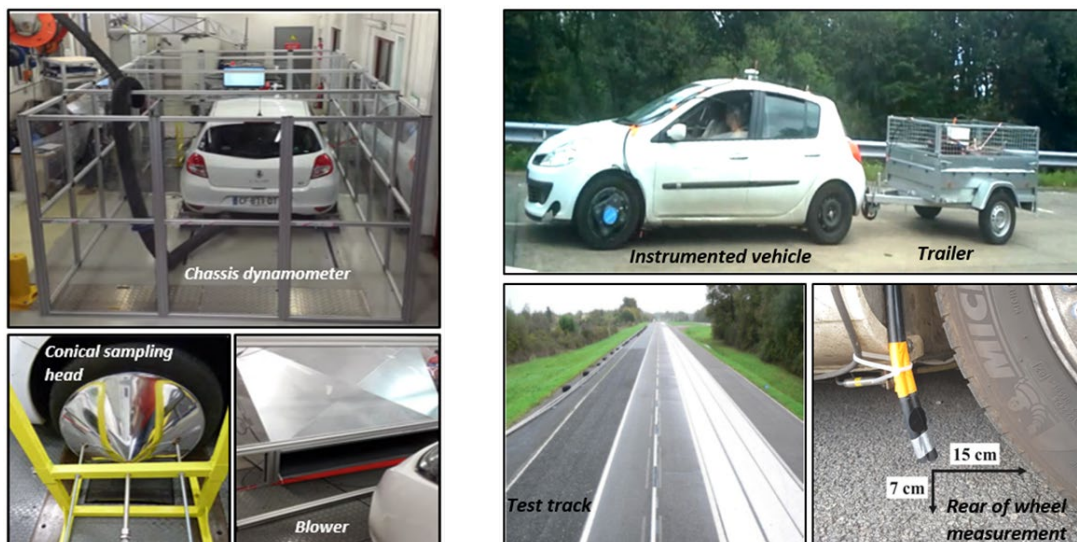
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### Supplementary information



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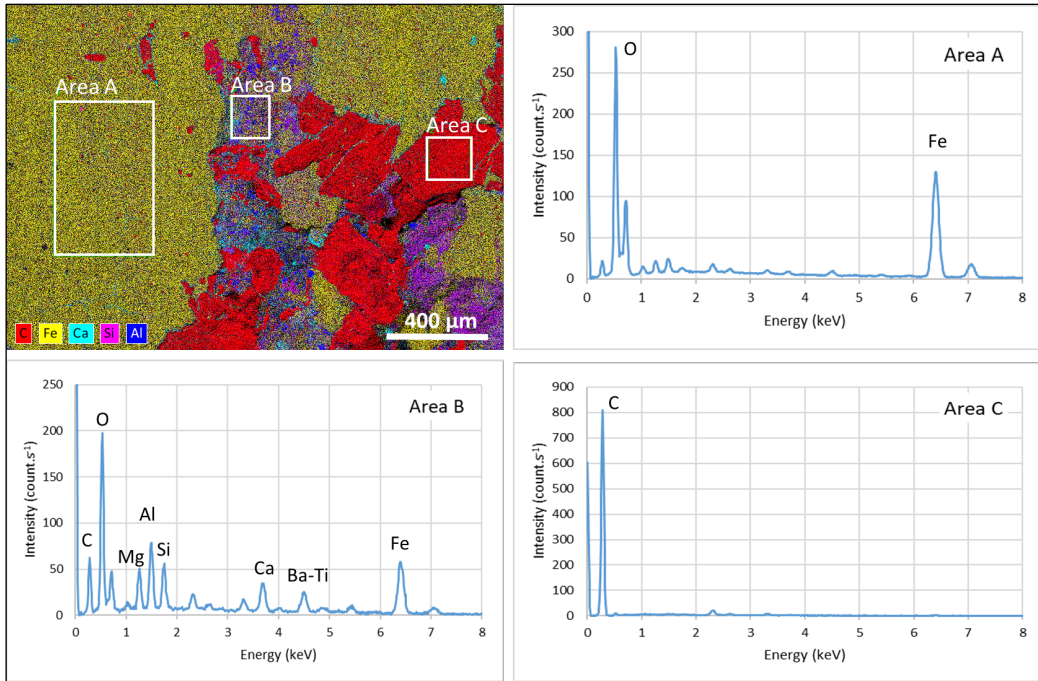
Figure S1: Example of comparison of aerodynamic diameters measured by ELPI (in black) with mobility diameters measured by FMPS (in blue). The two main modes are well coinciding for the two measurements although there is an apparent overestimation of particle counts in the nano-size range of ELPI, and therefore an underestimation of larger ones, which was already been reported and attributed to particle bounce (Leskinen et al., 2012; Virtanen et al., 2010).



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Figure S2: Photograph of the laboratory test on the chassis dynamometer (left) and measurements conducted on the test track using a trailer loaded with instruments (right)





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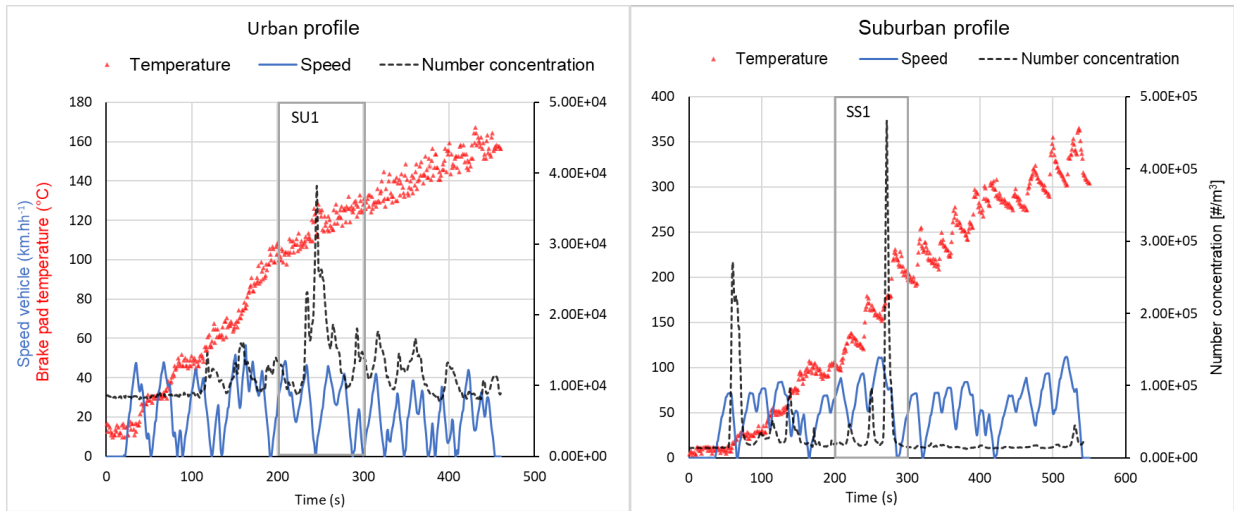
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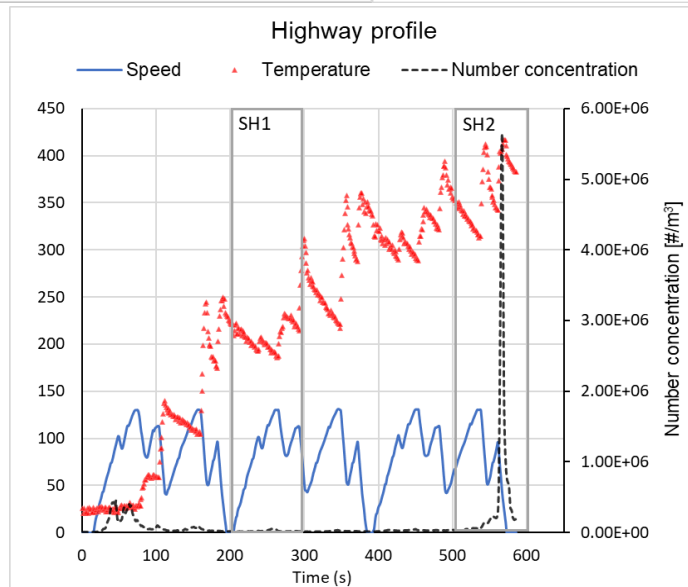
Figure S3: Elemental map of a brake pad surface for a semi-metallic pad and EDX spectra for the corresponding areas (A, B and C)

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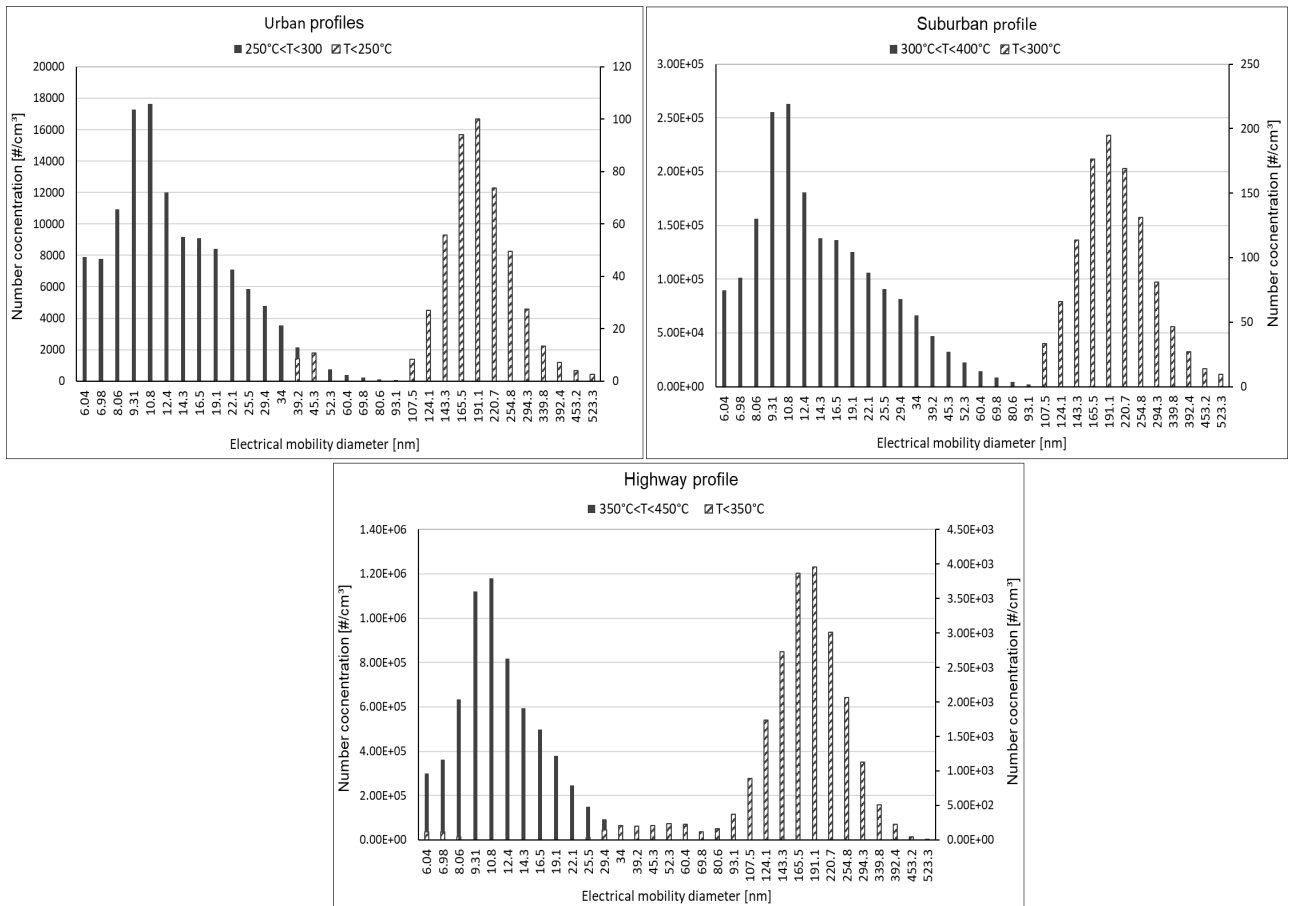
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Figure S4: Brake pad temperature evolution (in red) and PM concentration (black) vs. vehicle speed (blue) for three braking profiles: urban, suburban, and highway. The gray areas correspond to the temporal evolutions of the BWP size distributions presented in Figure 3.

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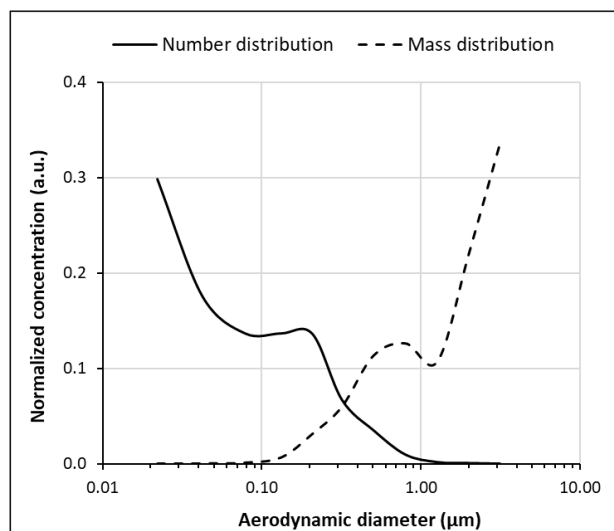
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Figure S5: Average number size distribution (after removal of the atmospheric background) of BWP emitted during urban, suburban and highway profiles vs. brake pad temperature

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Figure S6: Normalized number and mass size distributions of ambient particles along the roadside

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Vehicle speed	50 km/h	90 km/h	110 km/h
$U_0$ (m/s)	1.39	2.5	3.06
$U$ (m/s)	6.632	6.632	6.632
$\eta$ for particle diameter of 1 $\mu\text{m}$	1.000	1.000	1.000
$\eta$ for particle diameter of 5 $\mu\text{m}$	1.000	0.995	0.988
$\eta$ for particle diameter of 10 $\mu\text{m}$	0.996	0.949	0.908

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*Table S 1 : Aspiration efficiency for particles in the range 1-10 $\mu\text{m}$  for different simulated vehicle speeds. Considering the experimental configuration, the air flow velocity in the free-stream ( $U_0$ ) at the sampling head is estimated to be 10 times lower than the simulated vehicle speed due to air dilution.*

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Vehicle speed	50 km/h	70 km/h	90 km/h
$U_0$ (m/s)	3.345	4.344	5.345
$U$ (m/s)	3.316	3.316	3.316
$\eta$ for particle diameter of 1 $\mu\text{m}$	0.997	0.994	0.989
$\eta$ for particle diameter of 5 $\mu\text{m}$	0.928	0.892	0.867
$\eta$ for particle diameter of 10 $\mu\text{m}$	0.715	0.639	0.603

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*Table S 2: Aspiration efficiency for particles in the range 1-10 $\mu\text{m}$  for different vehicle speeds. The flow velocity in the free-stream ( $U_0$ ) was evaluated considering the relation between the vehicle speed and the flow velocity behind the front tire established by (Kwak et al., 2013).*

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Fe-containing particles along the Roadside (FeR)					
	Accumulation mode (A) (n=51)		Coarse mode (C) (n=442)		
	FeR-A1	FeR-A2	FeR-C1	FeR-C2	FeR-C3
	P=56%	P=44%	P=58%	P=23%	P=19%
<b>Dm</b>	0.87	0.65	3.50	2.40	2.15
<b>C</b>	23.1	37.1	33.8	22.1	17.8
<b>N</b>					7.6
<b>O</b>	29.5	37.8	30.8	45.6	46.0
<b>Na</b>				1.3	6.5
<b>Mg</b>				0.9	1.1
<b>Al</b>				2.5	
<b>Si</b>	1.1	4.13	1.0	6.4	1.4
<b>S</b>			1.0	1.6	1.4
<b>Ca</b>		2.5		3.4	4.3
<b>Fe</b>	45.6	16.1	31.0	15.3	13.1

1006 *Table S3: Mean diameter (Dm, in  $\mu\text{m}$ ) and mean elemental composition (wt. %) of Fe-containing particles collected along the*  
 1007 *roadside for the PM1-10 and PM0, 1-1  $\mu\text{m}$  stages (classification by means of clustering analysis from 51 coarse and 442 fine*  
 1008 *particles analyzed with SEM-EDX). The number of particles (n) analyzed by SEM-EDX and the relative proportion (P) of the*  
 1009 *various particle types are indicated for each size mode. Only those elements with a mean concentration of at least 0.8% are*  
 1010 *reported.*