

1 **Appraisal of measurement methods, chemical composition and sources of**
2 **fine atmospheric particles over six different areas of Northern Belgium**

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13
14 **Abstract –**

15 Daily and seasonal variation in the total elemental, organic carbon (OC) and elemental carbon (EC)
16 content and mass of PM_{2.5} were studied at industrial, urban, suburban and agricultural/rural areas.
17 Continuous (optical Dustscan, standard tapered element oscillating microbalance (TEOM), TEOM
18 with filter dynamics measurement system), semi-continuous (Partisol filter-sampling) and non-
19 continuous (Dekati-impactor sampling and gravimetry) methods of PM_{2.5} mass monitoring were
20 critically evaluated. The average elemental fraction accounted for 2-6 % of the PM_{2.5} mass measured
21 by gravimetry. Metals, like K, Mn, Fe, Cu, Zn and Pb were strongly inter-correlated, also frequently
22 with non-metallic elements (P, S, Cl and/or Br) and EC/OC. A high OC/EC ratio (2-9) was generally
23 observed. The total carbon content of PM_{2.5} ranged between 3-77 % (averages: 12-32 %), peaking
24 near industrial/heavy trafficked sites. Principal component analysis identified heavy oil burning,
25 ferrous/non-ferrous industry and vehicular emissions as the main sources of metal pollution.

26
27 **Capsule:** This work compares PM_{2.5} monitoring methods to characterize PM_{2.5} over six locations of
28 different anthropogenic activities over Northern Belgium.

29
30 **Keywords:** respirable particles, PM₁₀, heavy metals, mineral content, EDXRF analysis, soot,
31 multivariate analysis, non-exhaust emission.

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33 1. Introduction

34 The fine fraction of suspended particulate matter (SPM) is generally referred as PM_{2.5} in the
35 literature, i.e., any particle with an aerodynamic diameter below 2.5 µm. Particulates in this size
36 range have been reported to be responsible for adverse health effects in humans (Pope and Dockery,
37 2006). PM_{2.5} aerosols also affect the local air quality of populated areas by impairing visibility and
38 contribution to acid precipitation (IPCC, 2007; Harrison et al., 2004). Fine particulates play a direct
39 role in global climate change by absorbing and scattering the solar radiation, thereby altering the total
40 radiation budget of the Earth-atmosphere system (IPCC, 2007), and an indirect role by changing the
41 depth and albedo of clouds (Twomey, 1974; IPCC, 2007). Due to the complexity in composition and
42 impacts of PM_{2.5} aerosols, in addition to the need for their physical and chemical characterisation
43 (e.g., metal content), it is also important to determine their sources, how they are formed, their
44 distribution, and local and global transport mechanisms to achieve a better understanding of these
45 aerosols, and to evaluate their risks to human health (Maenhaut, 2008).

46 Over most of the PM_{2.5} size range, carbonaceous particles in the atmosphere have been
47 shown to be composed of elemental carbon (EC) and organic carbon (OC) (Putaud et al., 2004). The
48 EC has a chemical structure close to that of impure graphite (Chen et al., 2003). EC originates from
49 direct emission of particles from various anthropogenic sources, mainly from combustion (e.g., diesel
50 engines) (Lim et al., 2002). OC originates from both direct emission of particles and atmospheric
51 transformation of organic (gaseous) compounds. Secondary OC has been reported to be generated by
52 condensation of low vapour pressure products during the photooxidation of hydrocarbons (Chow et
53 al., 1996).

54 In the present study, daily, seasonal and site-specific variations in the elemental, EC and OC
55 content and the mass of PM_{2.5} aerosols were evaluated to identify their chemical composition and
56 sources at six locations of diverse anthropogenic influence in Northern Belgium. Furthermore,
57 continuous, semi-continuous and non-continuous sampling and/or monitoring techniques and
58 analytical chemical methods were applied and critically assessed to aid the decision-making process
59 for PM_{2.5} monitoring and to reduce the bias in the environmental analyses performed.

60

61 2. Experimental

62 2.1. Sampling site characteristics

63 Six sampling sites, representative of different anthropogenic influences in Northern Belgium
64 (called as Flanders, Fig. 1), were selected: (1) Petroleumkaai (industrial site), the harbour of Antwerp
65 surrounded with several petroleum processing plants, oil refineries and other oil industrial plants; (2)
66 Borgerhout, one of the central districts of Antwerp with high density urban traffic (40-50 thousand
67 cars/day) and also influenced by a non-ferrous industrial site of Hoboken within 5 km distance – a
68 nearby district of Antwerp; (3) Zelzate (suburban site) – at the junction point of a busy main-road of
69 the town with three smaller roads (but lower traffic density than that at Borgerhout) and a petrol
70 station, close to a heavily travelled motorway (A11) and a highway (R4), and also nearby steel
71 industry; (4) Hasselt (suburban site, channel-side with modest traffic density, i.e., shipping at nearby
72 sluice gates and highway with medium/low traffic intensity); (5) Wingene (rural, agricultural area),
73 negligibly low traffic, but certain agricultural activities (animal farming and crops); and (6) Mechelen
74 (outskirts), relatively low traffic density, and some industrial impact, i.e., production of dyes and
75 washing powder. Each location was visited in two, non-concurrent campaigns in diverse seasons of
76 the year (Table 1). The most important meteorological parameters, such as wind-speed (WS), wind
77 direction (WD), relative humidity (RH), air temperature (T_a), air pressure (P_a) and the half hourly
78 amount of precipitation (PR) were also recorded during the sampling campaigns at weather stations
79 located near the sampling sites.

80

81 2.2. Sampling of particulate matter

82 2.2.1. Continuous monitoring

83 For the continuous monitoring of $PM_{2.5}$, Rupprecht & Patashnick (R&P, Thermo Fischer
84 Scientific, East Greenbush, NY, USA) Model 1400 tapered element oscillating microbalance
85 (TEOM) units were used, each fitted with a $PM_{2.5}$ Sharp-Cut Cyclone inlet. The inlet was preheated
86 to 40 °C or 50 °C, to eliminate the effect of condensation or evaporation of water particles. For a
87 short period in Borgerhout, a TEOM was equipped with a filter dynamics measurement system
88 (FDMS), which calculates PM-concentrations based on separate ambient air and reference (at 4 °C)

89 measurements. Using a splitter, the sampled air is alternately sent for 6 min to the measurement (at
90 the conditioning temperature of 30-50 °C), either directly, or after filtration at 4 °C. Thus it can be
91 used to detect the vaporization losses encountered with a standard TEOM. The concentration of
92 PM_{2.5} was also optically monitored (i.e., scattering measurements) with the application of an R&P
93 Dust Monitor (Dustscan).

94

95 2.2.2. Non-continuous sampling and gravimetry

96 Daily (24 h), size-fractionated aerosols at a height of ~1.8 m above ground level were
97 occasionally sampled using a four-stage Dekati-impactor with an airflow rate of 30 dm³ min⁻¹.
98 Nuclepore-filters with a diameter of 25 mm and a pore-size of 0.2 µm were fitted onto the three,
99 upper-stage impactor-plates, corresponding to >PM₁₀, PM_{10-2.5}, and PM_{2.5-1}, fractions, respectively. A
100 Whatman filter with a diameter of 47 mm was placed onto the bottom of the impactor to collect the
101 “back-up” (PM₁) fraction. The filters were weighed on a micro-balance (Sartorius model M5P-
102 000V001, Göttingen, Germany) before and after sampling according to the EN12341 protocol. The
103 TSP, PM₁₀, and PM_{2.5} data were calculated from the masses of diverse fractions deposited on the
104 different stages of the impactor.

105

106 2.2.3. Daily sampling of PM_{2.5} and PM₁₀ for EDXRF analysis

107 Automated R&P Model Partisol Plus samplers and an ESM Eberline Model FH95 SEQ
108 (Eberline Instruments GmbH, Germany) were used for the daily sampling of PM_{2.5} and PM₁₀,
109 respectively, with an airflow rate of 1 m³ h⁻¹. Various types of filters of 47 mm diameter and 0.8 µm
110 porosity were tested, e.g., Teflon, cellulose nitrate, and cellulose acetate. The latter filter material was
111 used for sampling the total elemental content, since it gave the lowest blanks for XRF-analysis. After
112 removing the samples from the sampling instruments, they were stored in a cooling box and
113 transported to the laboratory, where they were kept in a fridge (below 4°C) till sample processing.
114 The filters were subjected to gravimetric analysis (see Section 2.2.2.). However, the gravimetric
115 analysis of the cellulose acetate filters was influenced by the tendency to be electrostatically charged,

116 which resulted in a certain deviation in the measured filter masses. For this reason, only the
117 gravimetric results for Teflon-filters were taken into consideration.

118

119 2.3. Analytical methods and instrumentation

120 2.3.1. Energy dispersive X-ray fluorescence spectrometry (EDXRF)

121 The EDXRF analysis was performed on a Tracor Spectrace 5000 instrument, which uses a
122 low power Rh-anode X-ray tube (17.5 W). The characteristic X-ray radiation was detected by a
123 Si(Li) detector. For determining high-Z elements (starting from K), an accelerating voltage of 35 kV
124 and a current of 0.35 mA were used. The acquisition time was set at 10000 s. Low-Z elements (from
125 Al to Cl) were measured at 10 kV and 0.35 mA with an acquisition time of 4000 s. The measured
126 intensities were converted into elemental concentrations by the AXIL (analysis of X-ray spectra by
127 non-linear iterative least-squares) program (Vekemans et al., 1994). The limit of detection (LOD)
128 values for Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Se, Br, Rb, Sr and Pb ranged from 3.8 to 12.6
129 ng m⁻³ for air samples. The precision of the determinations expressed as the relative standard
130 deviation (RSD) was generally better than 1 % for the high-Z elements, but it was around 5 % for Al,
131 Si, P, S and Cl (Samek et al., 2002).

132

133 2.3.2. Monitoring of organic and elemental carbon

134 Particulate carbon was monitored with an Ambient Carbon Particulate Monitor (ACPM)
135 Model 5400 (R&P) fitted with a Stairmand PM_{2.5} cyclone head. Air was sampled at a flow-rate of
136 16.7 l min⁻¹ on the impaction plate (with a 50 % effective aerosol cut-off diameter of 0.14 µm) of the
137 ACPM preheated at 50 °C to avoid the adsorption of gaseous organic compounds. The sampling
138 interval was set at 2 h. As a first stage of the analysis, the samples were heated up to 340 °C to
139 decompose the organic species deposited. This step was followed by afterburning at 750 °C. After
140 each analysis step, performed in a closed loop under atmospheric air, the CO₂ concentration was
141 measured with a non-dispersive infrared detector, and the carbon content was calculated. The
142 concentrations obtained after the first and the second analysis stage correspond to the fractions of OC
143 and EC, respectively.

144 It is to be noted that the ACPM may suffer from positive sampling artifacts, which depend on
145 the working-temperatures of its impactor plate (Matsumoto et al., 2003). However, the influence of
146 these artifacts on the measurement of OC collected for longer sampling intervals (e.g., 2 h),
147 especially at its higher concentrations, and evolved at a higher temperature (250-340 °C) and EC in
148 aerosols could involve less uncertainty (Matsumoto et al., 2003). It should also be mentioned that EC
149 readings from the standard ACPM have been found to be lower than those from a modified-ACPM,
150 due to the fine aerosol cut-off, as above (ten Brink et al., 2005). Because of this negative bias, **the**
151 **standard ACPM technique is just reliable in a sense to make an estimation of the EC/OC values.**

152

153 2.4. Data evaluation and statistics

154 The analytical data were statistically treated using the SPSS software package (version 14.0).
155 Bivariate correlation analysis was performed by calculating the Pearson's correlation coefficient at
156 two-tailed significance levels (i.e., corresponding to correlations significant either at the $p=0.05$ or
157 $p=0.01$ levels, respectively). The daily metal levels were also processed using the principal
158 component analysis (PCA) with varimax rotation and Kaiser Normalization. Only principal
159 components (PCs) having an eigenvalue in the component data set higher than unity before varimax
160 rotation were retained.

161

162 3. Results and discussion

163 3.1. Assessment of PM_{2.5} dust monitoring methods

164 3.1.1. Bias experienced with optical methods

165 The temporal evolution of the PM_{2.5} mass curves from the optical Dustscan was very similar
166 to those of gravimetric methods, but its absolute values were strikingly higher (Fig. 2). Since this
167 mass deviation rather randomly changed, it was not possible to specify an exact factor/function for
168 this inaccuracy. Dustscan, like other optical methods, is usually calibrated against so-called "standard
169 aerosols". However, the morphology and composition of atmospheric aerosols can change rapidly
170 over time and location, which can cause a bias in the optical measurements.

171

172 3.1.2. TEOM versus gravimetric particulate measurements

173 The PM_{2.5} levels from gravimetry were significantly higher than those of the standard TEOM
174 (Fig. 3). This bias is due to the temperature conditioning of the air stream in the TEOM (cf. Section
175 2.2.1.), which treatment can cause mass loss through evaporation of volatile components in PM_{2.5},
176 e.g., ammonium nitrate (NH₄NO₃) (Charron et al., 2004), semi-volatile organic compounds (Vecchi
177 et al., 2009), and water attached to aerosol particles due to their hygroscopic nature, even at as low
178 temperatures as 25-30 °C. This finding was verified by simultaneous monitoring of PM_{2.5} in
179 Mechelen in the second campaign with two TEOM units, applying conditioning temperatures of 50
180 °C and 40 °C, respectively (Table 2). A Partisol sampler working at 20 °C was used as a reference.
181 No mathematical correction for NH₄NO₃ evaporation losses was applied in these experiments, as
182 usually recommended when applying standard TEOM methods (Charron et al., 2004). It is to be
183 noted that NH₄NO₃ has been found in PM_{2.5} in concurrent samples of the same sites (Bencs et al.,
184 2008). As expected, an increase in the conditioning/sampling temperature resulted in a decrease in
185 the daily PM_{2.5} mass (see Table 2 and Fig. 3).

186 Comparing the standard TEOM and TEOM-FDMS results, an obvious discrepancy can be
187 seen between the two PM_{2.5} data series (Fig. 3). On the other hand, the TEOM-FDMS values
188 approach very well those from Partisol sampling and gravimetry. Thus they exhibit a more accurate
189 monitoring of PM_{2.5} than the methods involving the use of mathematical correction factors.

190 Interestingly, the PM_{2.5} mass data in Mechelen showed a contradicting trend (Fig. 4), i.e., a
191 reverse pattern on most sampling days (i.e., Partisol < TEOM (40 °C) < TEOM (50 °C)). The same
192 trend could be observed for Petroleumkaai in the 1st campaign and for Wingene and Zelzate in the 2nd
193 campaign. Moreover, the PM₁₀ results of TEOM and Eberline appeared to behave the same way. The
194 change in the daily average temperature (T_{da}) may offer a possible explanation. On those days, when
195 the expected difference between PM_{2.5} data of TEOM and gravimetry did not occur, T_{da} was always
196 higher than 15 °C, which led to the following conclusions:

197 (1) When the ambient air temperature is high enough (e.g., T_{da} > 15 °C), a part of the volatile
198 compounds is already evaporated before/during sampling. Therefore, the temperature conditioning in
199 TEOM has a less pronounced, or even negligible effect on PM_{2.5} values, i.e., the data are in

200 agreement independently of the temperature of conditioning. Since the Partisol filters were measured
201 passing some days (sometimes two weeks) after the sampling, they were longer exposed to
202 fluctuating outdoor conditions and generally higher indoor (laboratory) temperatures than the outdoor
203 temperatures. This can cause additional evaporation of volatile sample components, which results in
204 $PM_{2.5}$ values lower than those obtained from TEOM.

205 (2) When T_{da} is lower than 15 °C, the conditioning effect in TEOM is larger and the Partisol
206 filters are preserved much better during their stay in the sampling units, due to the low evaporation
207 losses.

208 (3) The fact that PM_{10} concentrations are less dependent on T_{da} during summer campaigns,
209 could be explained by the fact that relatively coarser particles are present in the air and a considerable
210 part of NH_4NO_3 exists in air by evaporation, due to the drier and warmer weather. Certainly, the
211 coarse particles present in the $PM_{10-2.5}$ fraction are less prone to vaporization than most fine particles
212 in $PM_{2.5}$. [The fine particles can be more easily vaporized than coarse particulates, due to the changing
213 in their solid state characteristics with a decreasing aerodynamic diameter \(e.g., melting and boiling
214 point\).](#) That's why, their masses affect the PM_{10} measurements to a larger extent, and the influence of
215 T_{da} does not prevail so clearly. In cold periods with a high precipitation rate, such as in Mechelen in
216 the second campaign (Table 1), the less coarse particles remain in the air. Hence, PM_{10} could
217 evidently be more influenced by evaporation effects.

218

219 3.1.3. Site-specific variation in the levels of $PM_{2.5}$

220 The highest average daily concentrations of $PM_{2.5}$ were observed in Borgerhout, i.e., 29 ± 15
221 $\mu g m^{-3}$ and $45 \pm 22 \mu g m^{-3}$ for the first and second campaigns, respectively (Table 3). The highest daily
222 mean concentration of $PM_{2.5}$ was also experienced in Borgerhout ($99.7 \mu g m^{-3}$) in the second
223 campaign. A strong linear correlation was found between the $PM_{2.5}$ and PM_{10} concentrations. The
224 $PM_{2.5}$ contribution to the PM_{10} levels was the highest in Borgerhout (i.e., 71% on average; and 92%
225 maximum), which means that most of the particles were present in the fine to ultra-fine particle range
226 at this site.

227 In view of the new European Directive (ED), it might be interesting to evaluate the measured
228 concentrations and to check if they meet the proposed PM_{2.5} limit values of yearly average of 25 µg
229 m⁻³ (European Directive, 2008). Since only a maximum of three-month-long monitoring was carried
230 out at each location, it was only possible to make a prediction based on extrapolation with this
231 limited data set. These daily average PM_{2.5} values have already exceeded the ED-limit for three sites
232 in the first campaign and for Borgerhout in the second campaign (Table 3). However, if taking into
233 account the overall campaign averages, only the levels at Borgerhout (38 µg m⁻³) were found above
234 the ED-limit (Table 4).

235 The average yearly concentration of PM_{2.5} at each sampling location was calculated by an
236 extrapolation scheme using the average daily PM_{2.5} concentrations for the sampling campaigns at
237 each site and a correction factor based on the proportion of the average concentrations monitored
238 during the same period and over a whole year at other three reference locations. In this way, it was
239 possible to avoid over- or underestimations, due to unforeseen, general seasonal variations. The
240 yearly average PM_{2.5} concentration for the three reference locations (Mechelen-Zuid, Mechelen-
241 Nekkerspoel and the Brussels-Zaventem) in 2002 was 16 µg m⁻³; the average for the same period as
242 in Borgerhout was 21 µg m⁻³ (Table 4). These results are also in agreement with the annual mean
243 PM_{2.5} level of 17 µg m⁻³ reported for Menen, Belgium (Ravindra et al., 2008). Therefore, one could
244 already assume that the very high average value of 38 µg m⁻³ in Borgerhout was due to measurements
245 that were carried out in an episodic pollution period with very high PM_{2.5} levels. Thus the yearly
246 average for Borgerhout is expected to be lower than 38 µg m⁻³. According to the results of
247 extrapolation (Table 4), only the Borgerhout site would not meet the ED-limit.

248

249 3.1.4. Impact of meteorological conditions on the PM_{2.5} and PM₁₀ mass

250 As would be expected, an increase in wind speed, precipitation, and/or RH decreased the
251 PM_{2.5} and PM₁₀ levels (Supplementary Table 1). On the other hand, an increase in the air temperature
252 would be generally followed by an increase in the aerosol mass, i.e., particle accumulation.
253 However, this trend was experienced only for a couple of sites, like Borgerhout during the winter-
254 spring and Wingene in the summer campaign, while T_a was rather anti-correlated with both PM_{2.5}

255 and PM₁₀ aerosols at the other sites/campaigns. This was likely due to the fairly high percent of
256 volatile fraction of aerosols, e.g., NH₄NO₃ (Bencs et al., 2008), the increase in T_a allowed their
257 vaporization in both particulate fractions.

258

259 3.2. Elemental composition of PM_{2.5}

260 3.2.1. Ratio of elements in PM_{2.5}

261 The elemental content was found to be at a relatively low, but highly fluctuating percentage
262 in PM_{2.5}, ranging from 0.1 to 24 % with an average of 3.4 % (Table 5). As a general observation,
263 peak levels of elements in PM_{2.5} (i.e., values generally higher than 10 % of the total PM_{2.5} mass) were
264 found at sites with industrial impact (Zelzate, Mechelen and Petroleumkaai). In Hasselt, the
265 elemental fraction was higher only in the second campaign, which is likely due to some industrial
266 impact as well as the ship-traffic nearby this location, and the prevailing winds from the west. As
267 expected, at the rural site of Wingene a lower percentage of elements were found in PM_{2.5}, i.e., up to
268 6.3 %, when the maximum observations were considered. These findings show the importance of
269 industrial emission of metals and the lower impact of the traffic. The calculated total-element-to-
270 PM_{2.5} ratios are comparable with those observed for the city of Ghent in Belgium (3-4 %) (Viana et
271 al., 2007) and for Central European cities (2-5 %), but much lower than those for North and South
272 European cities with values of 15-40 % and 8-20 %, respectively (Querol et al., 2004).

273

274 3.2.2. Influence of anthropogenic activities on the elemental content of PM_{2.5}

275 The elemental content in PM_{2.5} showed high site-specific variations (Table 6). Non-metallic
276 elements, such as P, S, Cl, Se and Br, were also detected at significant air levels, ranging from 14-
277 302, 9.9-4600, 9-2400, 1.1-28 and 2-91 ng m⁻³, respectively. The highest total concentrations of
278 elements were found at sites under industrial influence (Zelzate, Petroleumkaai, and Borgerhout).
279 The cold seasons were characterized with increased metal content of PM_{2.5} (e.g., K, V, Fe, Ni, and
280 Zn). The total elemental content (200-1100 ng m⁻³), irrespective of the nature of the sampling site, is
281 commensurate with those data reported for Central European cities (Vallius et al., 2005) and for

282 Ghent (Viana et al., 2007), but generally lower than those from North and South European cities with
283 1000-6000 ng m⁻³, respectively (Querol et al., 2004).

284 Fairly high concentrations of Al, Si, K, Ca and Fe, i.e., up to a few hundred ng m⁻³, were
285 generally found at each site (Table 6). Most of these elements have a crustal origin (Maenhaut et al.,
286 2007), although some of them in PM_{2.5} may have an anthropogenic origin. For instance, Oravisjärvi
287 et al. (2003) have pointed out Li, F, Na, Mg, Al, K, Ca, Mn, Fe, Zn, Cd and Pb at enhanced
288 atmospheric levels in the vicinity of steel-works. Most of these elements were also found at increased
289 air levels at Zelzate (Table 6). At this area, F has also been observed at a bit enhanced level
290 compared to the other sites (Bencs et al., 2008). The highest K content was found at Borgerhout
291 during the cold season, which is likely due to the presence of biogenic materials (e.g., wood and coal
292 combustion). Similarly high K levels were observed in Zelzate during winter and also in Hasselt
293 during autumn.

294 Zn and Pb were present at lower levels than the former elements, but still at significant
295 concentrations (Table 6). The highest levels were at industrial/heavy trafficked sites (Zelzate and
296 Borgerhout). Pb is also associated with non-biomass burning sources, like fossil fuel combustion and
297 the non-ferrous industry (Nriagu and Pacyna, 1988). The nearby presence of a precious metal
298 refinery in Hoboken, i.e., within 5 km of the sampling site of Borgerhout, supports this possibility.
299 Also, coal-burning (e.g., in power stations) can be an emission source of Zn and Pb, as well as
300 increase the air concentrations of other heavy metals like Cr, Ni, Cu and Cd (Keegan et al., 2006). In
301 urban areas, the brake lining of vehicles is a major source of Cu, Zn, Cd, Sb and Pb (Hjortenkrans et
302 al., 2007).

303 Ti, V, Cr, Mn, Ni, Cu and Rb were present at levels lower than ~10 ng m⁻³ in PM_{2.5} (Table
304 6). Sr was measured in well detectable amounts only at Zelzate during the first campaign (average: 1
305 ng m⁻³), which is likely due to the nearby steel industry. Cu was found at elevated levels at the heavy
306 trafficked Borgerhout. V and Ti were detected at increased atmospheric concentrations at the
307 industrial/heavy trafficked sites. Interestingly, V was also observed at fairly enhanced levels at the
308 agricultural Wingene during both campaigns. This could be due to the heavy diesel emission from

309 vehicles used over the crops. The V-to-Ni ratio of ~2 supports this assumption (Maenhaut et al.,
310 2007).

311

312 3.3. Source identification of elements in PM_{2.5}

313 3.3.1. Correlation analysis

314 3.3.1.1. General trends over sites and campaigns

315 Some of the elements, mostly originating from anthropogenic emissions, like K, Mn, Fe, Cu,
316 Zn and Pb, further referred to as the “base-group”, were strongly correlated with each other
317 (Supplementary Table 1). Moreover, they were frequently correlated with non-metallic elements, like
318 P, S, Cl and/or Br, and also with EC and OC. The correlation of Fe, Zn, Cu, Ba and EC suggests
319 traffic/vehicular sources (Viana et al., 2008).

320 Generally, the anthropogenic elements were strongly correlated with elements mostly of
321 crustal origin, e.g., Al, Ca, and/or Si, Ti. Monitoring data from a background site indicate Si-Al-Fe-,
322 Si-, Ca-S-Si-, Ca-Si-, Fe-Si-, and Ti-Si-rich particles as typical soil elements (Hoornaert et al., 2004).
323 These elements in PM_{2.5} can also originate from anthropogenic sources. However, an enrichment
324 factor of higher than unity has been observed only for Ca and Ti over Uccle, an urban background
325 site in Belgium, i.e., 10 and 3, respectively (Maenhaut et al., 2007). Therefore, a rather crustal origin
326 has been presumed for the atmosphere over Flanders.

327 Cr was usually correlated with some elements of the base-group, which points to common
328 sources. Cr is mostly associated with traffic/vehicular emission (Viana et al., 2008; Hu et al., 2009)
329 and industrial combustion (e.g., coal-fuelled power plants (Keegan et al., 2006) and coke ovens
330 (Wang et al., 2003)). Ni was correlated with Cu over most of the sites, which can be interpreted as a
331 signature of oil combustion (Pio et al., 1989). Rb was generally well correlated with EC, OC and the
332 elements of the base-group at sites with industrial influence. Sr was usually correlated with Al and/or
333 Si, which shows its origin mostly of crustal sources. The most interesting site/campaign-specific
334 correlations are discussed as follows.

335

336 3.3.1.2. Site- and campaign-specific correlations

337 In Zelzate, besides the base-group, K was also correlated with Ni and Sr, in the winter
338 campaign, and with V and Rb in the summer-autumn campaign. These elements and Fe were closely
339 correlated to each other in both campaigns, thus they most probably originate from the neighbouring
340 steel industry. Interestingly, they were also correlated with Al and/or Si, which is usually of crustal
341 origin, but for Al studies also suggest combustion related sources in $PM_{2.5}$ (e.g., steel industry –
342 Oravisjärvi et al., 2003). In the winter campaign, Ca was correlated well with P, Ni and Cu, which
343 suggests that a part of the Ca likely comes from re-suspended road dust. In the winter campaign, V
344 was well correlated with Br and P, but in the summer-autumn campaign, it was strongly correlated
345 with Ni and Si. According to Maenhaut et al. (2007), V and Ni have a source of vehicular emission
346 and heavy oil burning. At Zelzate, V likely originates from the diesel traffic, but also, a part of it
347 might come from the re-suspended road dust, as observed by Oravisjärvi et al. (2003). Ti might
348 originate partly from the steel industry, since it was strongly correlated with Fe and the other steel-
349 industry related elements as above and EC in the summer-autumn campaign, but a part of it might
350 also come from crustal sources (Maenhaut et al., 2007) and/or re-suspended road dust (Oravisjärvi et
351 al., 2003).

352 The emission of a petrochemical plant has been characterized with increased S, V, Ni, Zn
353 and Se content of $PM_{2.5}$ (Bosco et al., 2005), which results in correlation of these elements. In
354 Petroleumkaai, the base-group, EC and OC were correlated with V and/or Ni, which points to a
355 source of the nearby heavy oil burning activities. In the autumn campaign, Ti was correlated with the
356 crustal Al, Si, but also with P and Se, while in the winter campaign, it was strongly correlated with V
357 and Si, thus it partly originates from the oil refinery emissions.

358 In the heavy trafficked Borgerhout, besides the base-group and crustal elements, Ca was also
359 correlated strongly with Ti, Cr, non-metallic elements and EC, thus they possibly come from road
360 dust re-suspension. Ti was correlated with V, Ni and P in the autumn-winter campaign, while it was
361 correlated with V, Cr, the base-group and crustal elements in the winter-spring campaign. Ni was
362 correlated well with EC and OC in the winter-spring campaign. Thus Ti and Ni were supposed to
363 originate from diesel emissions and heavy oil burning activities, e.g., the oil industry at
364 Petroleumkaai located northwest of Borgerhout.

365 The suburban/riverside site at Hasselt showed a pattern of strong correlations of the base-
366 group with crustal (Al, Si and Ca) and non-metallic (P, S and Br) elements in both campaigns.
367 Interestingly, Cl was strongly correlated with these elements too. Cl is of sea salt origin (Hoornaert et
368 al., 2003). However, at Hasselt, the origin of Cl might be found in the nearby paper industry, which is
369 a source of organic Cl. Interestingly, Ni was strongly correlated with most of the detected elements,
370 while Ti, V and Cr were somewhat poorly correlated with any of them. V has also been observed to
371 be poorly correlated in PM_{2.5} in Uccle, for which additional sources of Ni (e.g., non-ferrous industry)
372 were presumed to be responsible (Maenhaut et al., 2007). This is likely true for the Mechelen site.

373 The suburban Mechelen site showed a similar pattern of correlations of elements like that of
374 Hasselt, apart from the non-metallic elements, which were much less correlated with the others. For
375 example, Cl was correlated only with Ca and Ti, thus they likely originated from road dust re-
376 suspension. In the autumn-winter campaign, EC was correlated with Fe, Zn and Pb, .

377 In the rural/agricultural area of Wingene, the strong correlation of P, S, V, Ni and Br is likely
378 related to the use of diesel vehicles over nearby crops in the spring and summer seasons. In the spring
379 campaign, Ti was correlated with Ni and Br. Thus Ti likely also related to diesel fuel emissions. The
380 correlation of K, Cr, Mn, Fe, Zn, Pb and EC points to biomass burning (cf. Godoi et al., 2004).

381

382 3.3.2. Principal component analysis (PCA)

383 As the PCs were extracted successively, the first PC is more correlated with the variables
384 than the second (Supplementary Tables 2-7). Hence, only the major PCs from each site were assessed
385 for source characterization. The species having loadings above 0.7 were characterized "high" and
386 those below 0.4 "low". The species having PC loading of less than 0.4 were considered either not be
387 related to the other species or were explored in an additional PC (Costello and Osborne, 2005).

388 PC 1 for Petroleumkaai shows high loadings for P, S, K, Fe, Cu, Zn, and Pb. This PC
389 indicates the presence of petroleum refinery and other industrial combustion activities, which are
390 dominant in this area. Further, PC 2 has high loadings of Al and Si with moderate loadings of Ca and
391 Rb. This PC reflects the contribution of crustal elements. At Borgerhout, PC 1 is highlighted by Al,
392 Si, P, S and K with moderate loadings of Cl, Ca, V, Fe, Zn, Br and Pb. This PC shows a mixed

393 influence of vehicular emission and suspension of road dust. Zn has been identified in tire wear
394 emission and Fe in brake-drum abrasion (Manoli et al., 2002). PC 2 has high Fe and Cu loadings with
395 relatively lower loadings of Ca, Zn, Sr and Pb. This PC is indicative of non-exhaust emission sources
396 (e.g., wheel/brake abrasion of vehicles) with the influence of diesel exhaust, as well as the influence
397 of a nearby precious metal refinery. Although Pb has been phased out from petrol, generally, road
398 traffic emission remains one of the sources (Heal et al., 2005). Zelzate has high loadings of Al, Cl, K,
399 Zn, Br and Pb, Fe, with moderate loadings of Si, P, S, and Cu. This emission seems to be related to
400 the coke-oven industries in this area. PC 2 is represented by P, S and Cr suggestive of the combustion
401 activities in the vicinity.

402 PC 1 in Hasselt has high loadings of Si, S, Cl, K, Ca, Ni, Cu, Zn, Br and Pb, which shows the
403 mixed influence of natural and vehicular/ship emissions. PC 2 is represented by Cr and Fe. High
404 loadings of Fe and Cr are suggestive of road dust (Manoli et al., 2002), but considerable Cr comes
405 also from mobile sources (e.g., brake wear emissions) nearby heavily trafficked roads (Hu et al.,
406 2009). At Wingene, K, Fe, Zn and Pb show relatively higher PC loadings. In addition to the crustal
407 sources, K has been identified an important marker of biomass burning. Wingene is characterised as
408 a rural/agricultural area, thus biomass burning activities may be expected in this area. PC 2 has high
409 loadings of V, Mn and Ni. V and Ni are markers of fuel-oil (Heal et al., 2005), whereas the other
410 elements are likely due to local suspension of dust during agricultural practices. PC 1 at Mechelen is
411 highlighted by Al, Cl, K, Ca, Cu, Zn, Se and Br. Interestingly, high loadings of Br and Se were
412 noticed only at this location. Br is frequently used in dye industries; whereas Se is mainly used in
413 glass manufacturing chemicals and pigments. This PC shows the influence of local industrial
414 activities on PM_{2.5} composition. PC 2 has high loadings of Si, P, S, Fe, Rb and Pb. Some of these
415 elements can be related to the re-suspension of road dust (Heal et al., 2005), and hence, indicate their
416 relation with non-exhaust emissions. However, the presence of S and P also indicates industrial
417 combustion and biomass burning activities, respectively.

418

419 3.4. Elemental carbon and organic carbon content of PM_{2.5}

420 3.4.1. Site-specific variation of EC and OC

421 The daily concentrations of EC and OC determined non-concurrently with the ACPM over
422 various sites/campaigns are listed in Table 7. The minimum levels of EC ranged between 0.1-0.5 μg
423 m^{-3} , while the maximum EC values were found to be between 1.1-4.4 μg m^{-3} . Amongst the studied
424 sites, Borgerhout and Petroleumkaai reported the highest EC levels in the first campaign, with values
425 of 3.3 and 4.4 μg m^{-3} , respectively. The average level of EC was the highest also at the heavy
426 trafficked Borgerhout (1.3-1.5 μg m^{-3}), whereas sites with less traffic density showed much lower
427 concentrations (0.3-0.6 μg m^{-3}). The exception was Petroleumkaai, with an increased average value
428 of 1.1 μg m^{-3} in the first campaign, likely due to the nearby industrial emissions combined with
429 warmer weather conditions. The maximum daily OC values were found to be at Mechelen (10 μg m^{-3})
430 and Petroleumkaai (7.3-7.6 μg m^{-3}), and followed a similar pattern for the average levels of 4.1 and
431 4.2 μg m^{-3} , respectively.

432 In the literature, OC is often expressed as organic material (OM). To calculate OM,
433 multiplication factors of 1.6 and 2.1 for OC have been recommended for urban and non-urban
434 aerosols, respectively (Turpin and Lim, 2001). In this study, the factor of 1.6 was used for calculating
435 the OM values (Table 7). Fairly low average levels of EC and OC were observed for each
436 site/campaign, i.e., ranging between 0.4-1.5 and 2.3-4.2 μg m^{-3} , respectively (Table 7), which
437 correspond to an OM+EC range of 4.1-7.7 μg m^{-3} . This interval is similar to those OM+EC values,
438 reported for the total carbon content of $\text{PM}_{2.5}$ in Ghent, Belgium (3.9-9.7 μg m^{-3}) (Viana et al., 2007)
439 and to those OC+EC ranges for North European cities (1-6 μg m^{-3}) (Querol et al., 2004), but a bit
440 lower than those reported for Central European cities (3-16 μg m^{-3}) (Querol et al., 2004; Putaud et al.,
441 2004).

442 The two-hour peak values of EC and OC of 19 and 20 μg m^{-3} , respectively, were found at
443 Borgerhout (Table 8). A similar high OC value was found at Petroleumkaai in the first campaign;
444 whereas the EC value was lower (7.4 μg m^{-3}). The highest two-hourly minimum value of EC was
445 observed in Borgerhout (0.44 μg m^{-3}) in winter-spring, whereas the other sites yielded 1-2 orders of
446 magnitude lower values. Interestingly, the highest minimum OC values occurred in Mechelen (1.1 μg
447 m^{-3}) and Hasselt (0.86 μg m^{-3}), which are suburban areas (both influenced by industrial activities),

448 though the less intense, but continuous local emissions by car and ship traffic may contribute to these
449 increased background OC levels.

450

451 3.4.2. OC/EC ratios

452 The primary OC/EC ratio is defined as the atmospheric concentration of organic material
453 emitted directly into the air by anthropogenic sources and measured as primary OC divided by the
454 atmospheric level of EC. In early studies, a primary OC/EC value of 2.2 is assumed to indicate rising
455 secondary organic aerosol (SOA) levels in the atmosphere (Turpin and Huntzicker, 1991). An
456 OC/EC ratio of 2 has also been used to identify the formation of SOAs (Chow et al., 1996). A recent
457 study by Harrison and Yin (2008) suggests a minimum OC/EC ratio of 0.65.

458 For most of the sampling sites, a high OC/EC ratio (3.7-9) was generally observed (Table 7).
459 The exception was the heavy trafficked urban site of Borgerhout with a value of 1.9 in each
460 campaign. Especially high OC/EC ratios of 8.4 and 9 were observed at Mechelen and Hasselt,
461 respectively, the sites with medium-to-low traffic density and some industrial influence. The
462 experienced high OC/EC ratios indicate an intensive formation of SOAs or the contribution of local
463 sources to increase OC levels (e.g., wood burning).

464

465 3.4.3. Contribution of total carbon (EC+OC) to PM_{2.5} levels

466 The minimum levels of total carbon (TC) in PM_{2.5} ranged between 3 and 11 %, while its
467 maximum values were found to be between 22 and 77 % (Table 9). The average contribution of TC
468 was relatively high for Petroleumkaai (32 %), Hasselt (23%) and Mechelen (24 %), whereas the
469 remaining sites were usually below 14 %. Apart from the industrial site of Petroleumkaai, these
470 values are generally lower than those reported for the OM+EC content of PM_{2.5} for Ghent (32-39 %)
471 (Viana et al., 2007), as well as those reported for other European cities (20-45 %) (Viana et al., 2007;
472 Putaud et al., 2004; Querol et al., 2004).

473

474 **4. Conclusions**

475 The results obtained with the assistance of diverse physical and chemical characterisation
476 methods for PM_{2.5} generally showed very similar temporal and spatial changes in the pattern of the
477 studied aerosol components. The optical dust monitoring methods (e.g., Dustscan) produce higher
478 PM_{2.5} mass readings than the gravimetric methods, consequently, they can only be used for showing
479 concentration trends. The PM_{2.5} data from the standard TEOM monitoring are handicapped by
480 sampling artifacts (e.g., evaporation losses), whereas values from the TEOM-FDMS method agreed
481 well with the data from gravimetry. The monitoring over Flanders produced similar results for the
482 PM_{2.5} mass as for other European sites. However, a fairly large difference was found for the
483 elemental and total carbon (OC+EC) contributions in Flanders compared to higher percentages
484 observed in most of the Central European cities. This bias might be due to the use of a standard
485 ACPM in the present experiments, which is handicapped by lower EC/OC readings.

486 The metal content of PM_{2.5} indicated the high importance of industry-related emissions over
487 Flanders and a lower impact of the traffic emission. PCA and correlation analysis showed the
488 contribution of local traffic, heavy oil burning, combustion, and ferrous/non-ferrous industrial
489 emission, as the main sources to PM_{2.5} levels. It is to be noted that only one of the sampling sites
490 (Borgerhout with heavy traffic and some industrial influence) did not meet the EC's proposed annual
491 limit for PM_{2.5}. This is an indicative of "hot-spots" existing in the urban environment, which should
492 be locally monitored for PM_{2.5} levels. Moreover, local legislations on the air quality assurance of
493 these areas should be introduced for counteracting the pollution periods when the dust content is
494 expectable to overtake the daily allowable EC-limit, for example, by immediate actions of local
495 authorities (e.g., by reducing the density of the local traffic).

496

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651

Table 1. Time schedule of the sampling campaigns and average daily meteorological data with their variations (expressed as standard deviation – SD)

<i>Location – campaign</i>	<i>Sampling period (day/month/year)</i>	<i>Season</i>	<i>Average precipitation (mm day⁻¹)</i>	<i>Air temperature (°C)</i>	<i>Air pressure (hPa)</i>	<i>Relative humidity (%)</i>	<i>Wind speed (m s⁻¹)</i>	<i>Main wind direction*</i>	<i>n</i>
Petroleumkaai-1	18/09/2001-29/10/2001	A	3.0±6.0	14.9±1.8	1014±7	72±5	3.9±1.4	SW	36
Petroleumkaai-2	19/12/2002-23/02/2003	W	3.5±5.9	4.5±4.6	1013±13	73±7	4.4±1.4	S-SW/NE	52
Borgerhout-1	06/11/2001-10/12/2001	A-W	2.7±4.7	6.9±3.2	1024±11	75±5	3.6±1.6	SW	32
Borgerhout-2	10/02/2003-07/04/2003	W-Sp	0.9±1.8	7.1±3.9	1024±8	59±10	3.4±1.3	E/E-NE/SE	45
Zelzate-1	11/12/2001-30/01/2002	W	1.8±3.5	3.2±3.6	1022±13	81±25	5.5±2.6	SW/W-SW	51
Zelzate-2	13/08/2002-26/09/2002	Su-A	1.0±2.4	17±3.0	1018±4	83±7	3.3±1.2	N/N-NE	40
Hasselt-1	01/02/2002-26/03/2002	W-Sp	3.2±4.3	8.2±3.0	1010±9	67±7	5.5±2.1	SW/W-SW	46
Hasselt-2	27/09/2002-04/11/2002	A	2.1±3.0	11.5±2.1	1013±9	70±6	4.0±1.9	SW/W	38
Mechelen-1	16/05/2002-26/06/2002	Sp-Su	2.1±5.1	17.1±2.6	1014±7	60±6	3.7±0.9	SW	30
Mechelen-2	05/11/2002-03/01/2003	A-W	2.6±4.0	6.2±4.2	1012±11	74±7	3.8±1.4	S/SE/E	38
Wingene-1	27/03/2002-15/05/2002	Sp	1.3±2.3	10.2±2.5	1016±9	73±10	4.1±1.8	SW/N/N-NE	41
Wingene-2	27/06/2002-12/08/2002	Su	2.2±5.0	17.2±2.6	1014±5	79±6	3.6±1.1	SW/W/NW	45

Abbreviations: A – autumn, W – winter, Sp – spring, Su – summer,

n – the number of daily data used for calculating average concentrations

* – the main wind directions were extracted from the windrose of each site/campaign

Table 2. Daily average PM_{2.5}-data (µg m⁻³) from standard TEOM units

Location	Campaign 1			Campaign 2		
	<i>Min</i>	<i>Max</i>	<i>Mean ± SD</i>	<i>Min</i>	<i>Max</i>	<i>Mean ± SD</i>
Petroleumkaai	8.2	30.8	17 ± 6	8.3	31.9	16 ± 6
Borgerhout	6.2	34.9	20 ± 8	12.0	47.0	28 ± 11
Zelzate	6.2	53.1	21 ± 11	5.7	30.1	16 ± 7
Hasselt	6.8	34.4	13 ± 6	6.8	30.8	15 ± 6
Wingene	6.5	45.1	20 ± 10	6.5	27.2	13 ± 5
Mechelen	7.4	26.7	14 ± 5	9.9	44.5	21 ± 9
Mechelen	8.0	27.0	13 ± 4	8.0 ^a	51.0 ^a	22 ± 10 ^a

^a data from simultaneous sampling with a second TEOM unit operated at 40 °C

Table 3. Daily PM_{2.5}-data (µg m⁻³) from Partisol sampling and gravimetry (Teflon filters)

Location	Campaign 1			Campaign 2		
	<i>Min</i>	<i>Max</i>	<i>Mean ± SD</i>	<i>Min</i>	<i>Max</i>	<i>Mean ± SD</i>
Petroleumkaai	7.9	59.5	20 ± 11	6.3	61.6	21 ± 12
Borgerhout	0.2	62.7	29 ± 15	13.8	99.7	45 ± 22
Zelzate	1.6	67.7	26 ± 16	5.9	43.8	16 ± 8
Hasselt	5.5	59.0	19 ± 13	4.4	38.5	16 ± 8
Wingene	7.0	63.9	28 ± 16	5.2	23.2	11 ± 4
Mechelen	7.1	21.3	12 ± 7	9.2	60.1	24 ± 13

Table 4. Average PM_{2.5} concentrations for the monitoring period and extrapolated annual values

<i>Location</i>	<i>Number of days sampled</i>	<i>PM_{2.5} average (µg m⁻³)</i>	
		<i>Monitored with Partisol</i>	<i>Extrapolated yearly</i>
Petroleumkaai	89	21	23
Borgerhout	76	38	29
Zelzate	91	22	21
Hasselt	82	17	20
Wingene	94	20	18
Mechelen	71	19	19

Table 5. Percentage of PM_{2.5} due to elemental fraction (%)

Location	Campaign – 1			Campaign – 2		
	Min	Max	Mean ± SD	Min	Max	Mean ± SD
Petroleumkaai	2.4	10.9	6.3 ± 2.5	1.1	11.5	2.7 ± 2.0
Borgerhout	1.1	7.5	2.9 ± 1.2	0.9	4.0	2.2 ± 0.7
Zelzate	0.7	24.1	5.8 ± 5.5	1.0	12.9	4.0 ± 2.5
Hasselt	0.2	5.1	2.1 ± 1.1	1.8	14.6	4.3 ± 3.2
Wingene	0.4	6.3	2.1 ± 1.4	0.8	4.1	2.1 ± 0.8
Mechelen	0.1	15.1	2.8 ± 2.6	1.1	13.5	3.8 ± 2.8

SD – standard deviation

Table 6. Average elemental concentrations in PM_{2.5} over various sites and campaigns

<i>Location – campaign</i>	<i>Season</i>	<i>Average concentration (ng m⁻³)</i>														
		K	Ca	Ti	V	Cr	Mn	Fe	Ni	Cu	Zn	Rb	Pb	Al	Si	Total
Petroleumkaai-1	A	122	104	11	9	2	9	188	6	6	57	n.d.	13	122	366	1015
Petroleumkaai-2	W	139	3	5	16	1	4	66	7	4	31	1	26	83	58	444
Borgerhout-1	A-W	175	79	2	5	2	11	197	8	10	70	n.d.	28	55	186	828
Borgerhout-2	W-Sp	269	78	8	9	3	10	253	6	13	83	2	49	106	118	1007
Zelzate-1	W	247	111	6	6	1	13	279	9	7	84	1	24	60	302	1151
Zelzate-2	Su-A	104	63	6	8	1	3	95	4	5	20	1	15	51	181	557
Hasselt-1	W-Sp	93	32	2	n.d.	n.d.	4	56	3	3	33	n.d.	8	21	74	329
Hasselt-2	A	239	91	3	4	3	8	144	9	7	65	2	26	55	42	698
Mechelen-1	Sp-Su	69	34	1	1	2	5	60	4	5	31	1	8	29	74	324
Mechelen-2	A-W	183	91	3	3	3	9	127	8	9	66	2	30	46	35	615
Wingene-1	Sp	85	21	1	7	1	4	57	3	2	22	2	13	34	66	318
Wingene-2	Su	66	20	1	5	1	3	31	3	1	14	1	10	28	38	222
Overall average	-	149	61	4	7	2	7	129	6	6	48	1.4	21	58	128	626
SD	-	72	37	3	4	1	3	83	2	3	25	1	12	32	110	315

Abbreviations: A – autumn; W – winter; Sp – spring; Su – summer; n.d. – not detected

Table 7. Daily concentrations of EC and OC, and related OM and OC/EC ratios

Location – campaign	Season	EC ($\mu\text{g m}^{-3}$)			OC ($\mu\text{g m}^{-3}$)			OC/EC	OM*	OM+EC
		<i>Min</i>	<i>Max</i>	Mean \pm SD	<i>Min</i>	<i>Max</i>	Mean \pm SD	<i>Mean</i>	<i>Mean</i>	<i>Mean</i>
Petroleumkaai – 1	A	0.2	3.3	1.1 \pm 0.8	2.0	7.3	4.1 \pm 1.3	3.7	6.6	7.7
Petroleumkaai – 2	W	0.1	1.1	0.4 \pm 0.3	0.7	7.6	2.3 \pm 1.3	5.8	3.7	4.1
Borgerhout – 1	A-W	0.1	4.4	1.3 \pm 1.0	1.3	4.9	2.5 \pm 1.1	1.9	4.0	5.3
Borgerhout – 2	Wi-Sp	0.5	3.1	1.5 \pm 0.6	1.0	5.6	2.9 \pm 1.3	1.9	4.6	6.1
Zelzate – 1	W	0.0	1.9	0.6 \pm 0.5	1.1	5.9	2.6 \pm 1.5	6.5	4.2	4.8
Hasselt – 2	A	0.1	1.1	0.3 \pm 0.2	1.3	5.6	2.7 \pm 1.1	9.0	4.3	4.6
Mechelen – 2	A-W	0.1	1.5	0.5 \pm 0.3	2.0	10.2	4.2 \pm 1.6	8.4	6.7	7.2

Abbreviations: A – autumn; W – winter; Sp – spring; SD – standard deviation;

* OM was calculated with a multiplication factor of 1.6

Table 8. Minimum and maximum concentrations of EC and OC measured by the ACPM (2-hour data)

Location – campaign	EC ($\mu\text{g m}^{-3}$)		OC ($\mu\text{g m}^{-3}$)	
	<i>Min.</i>	<i>Max.</i>	<i>Min.</i>	<i>Max.</i>
Petroleumkaai – 1	0.014	7.42	0.014	18.5
Petroleumkaai – 2	0.025	1.72	0.391	8.75
Borgerhout – 1	0.001	19.2	0.001	19.9
Borgerhout – 2	0.444	5.05	0.465	10.3
Zelzate – 1	0.001	9.22	0.001	12.3
Hasselt – 2	0.006	3.09	0.858	8.06
Mechelen – 2	0.061	1.91	1.15	13.5

Table 9. Percentage of the PM_{2.5} concentration due to total carbon (EC+OC)

Location	Campaign – 1			Campaign – 2		
	<i>Min</i>	<i>Max</i>	Mean \pm SD	<i>Min</i>	<i>Max</i>	Mean \pm SD
Zelzate	3	77	14 \pm 17	-	-	-
Hasselt	-	-	-	11	49	23 \pm 8
Mechelen	-	-	-	7	71	24 \pm 13
Petroleumkaai	9	48	32 \pm 8	8	23	14 \pm 4
Borgerhout	8	48	14 \pm 8	4	22	12 \pm 4



Fig. 1. Map of Flanders showing the six sampling sites

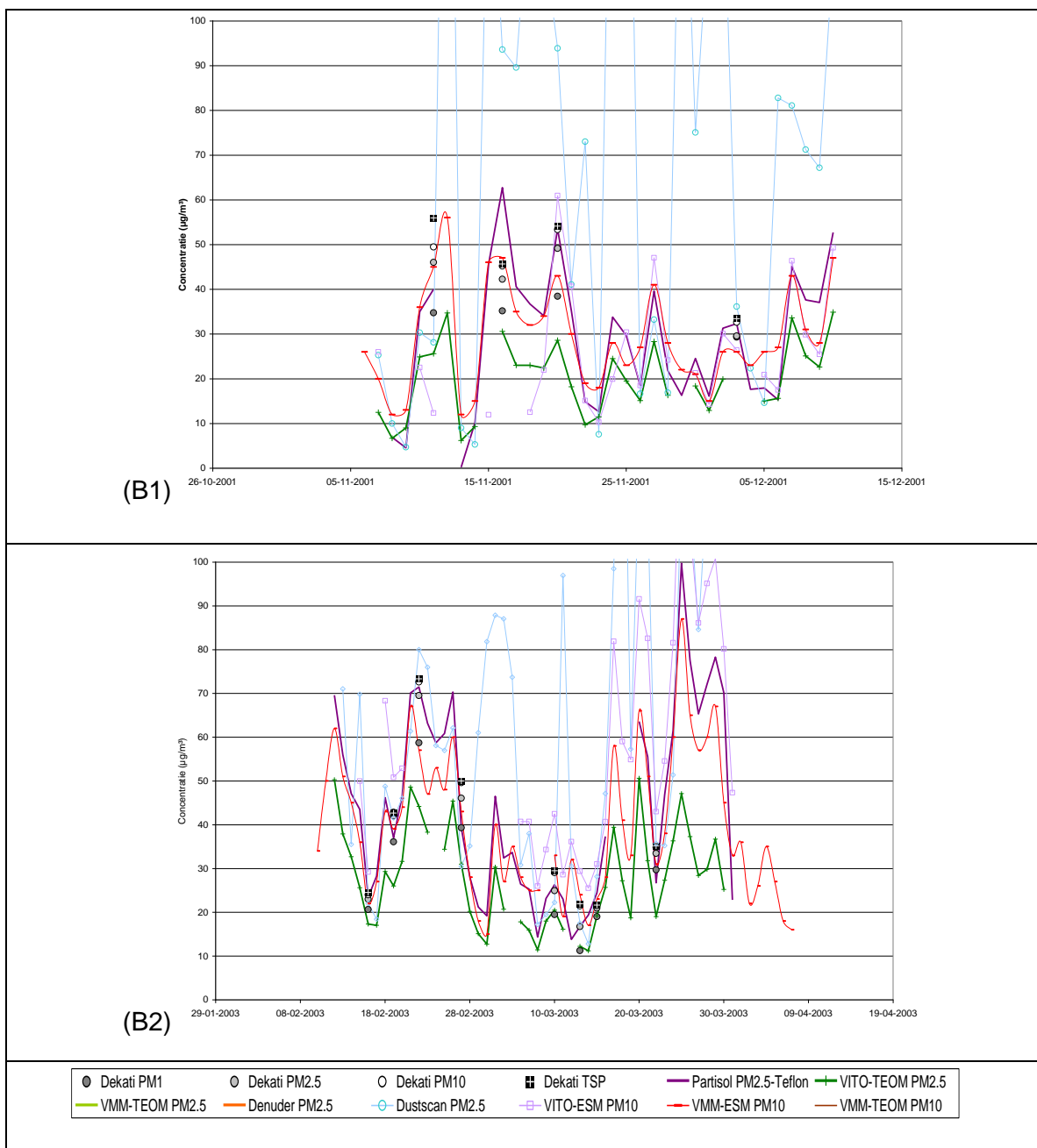


Fig. 2. Temporal variation of PM-concentrations in Borgerhout in campaign 1 (B1) and campaign 2 (B2) obtained with various methods

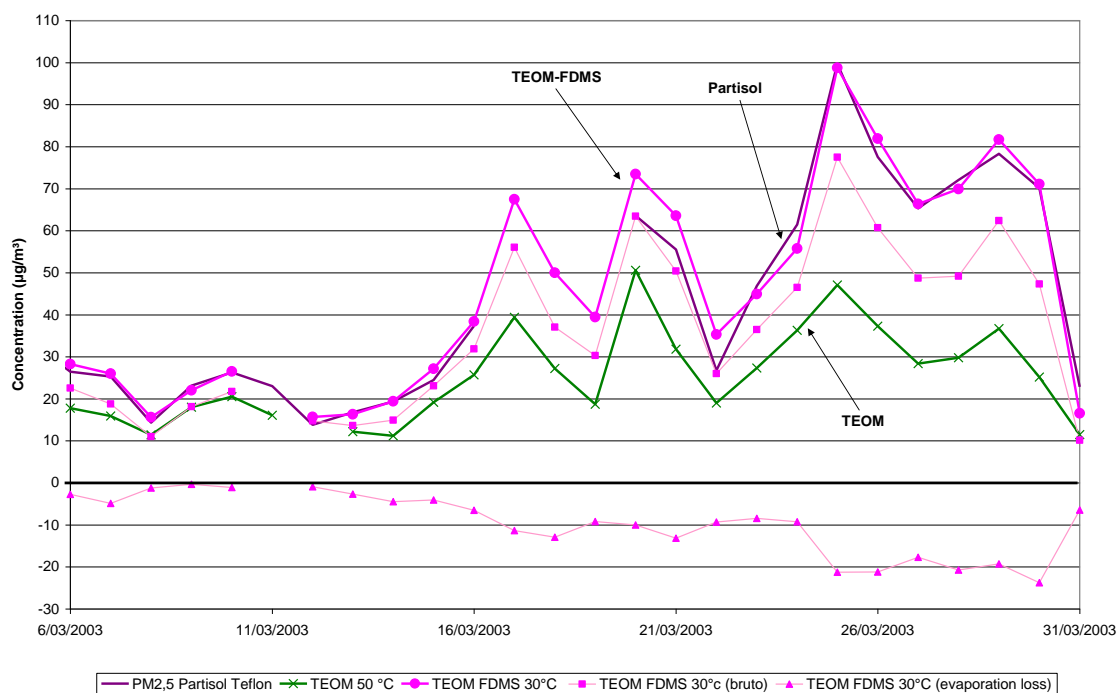


Fig. 3. Comparison of TEOM, TEOM-FDMS and Partisol sampling with gravimetry for the mass of $PM_{2.5}$ aerosols at Borgerhout in campaign 2 (bruto: the mass at 30 °C obtained after correction with the evaporation loss)

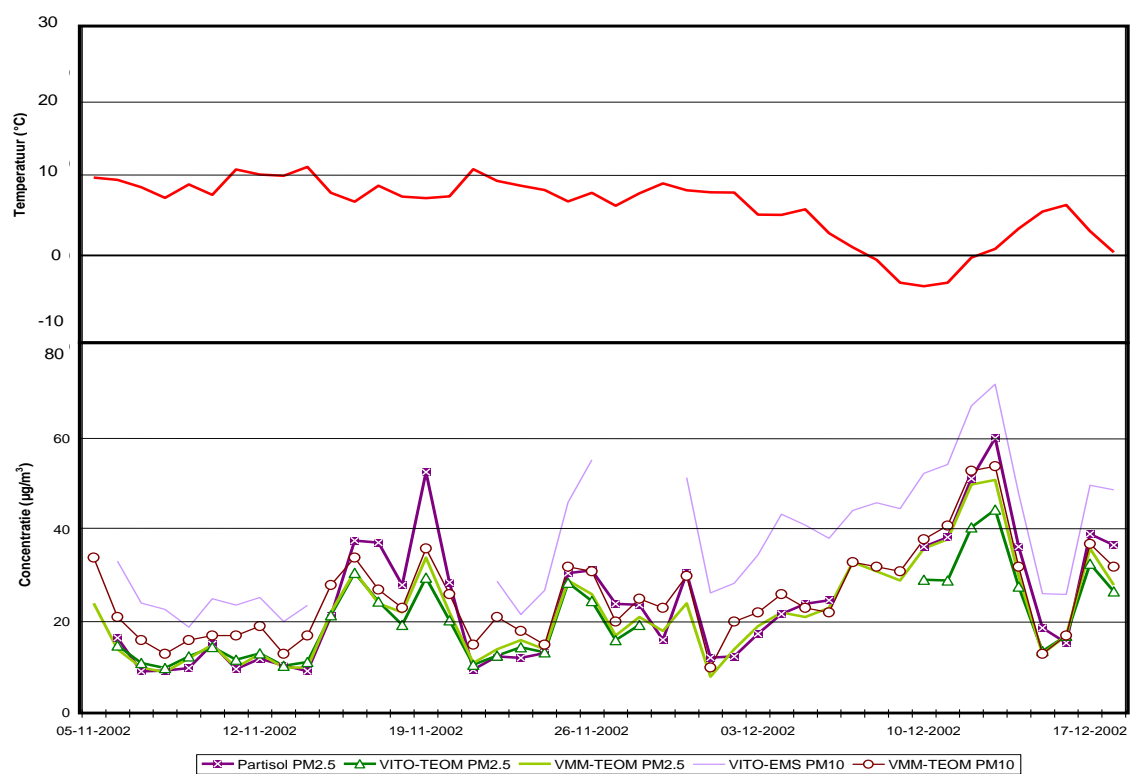


Fig. 4. Temporal fluctuation of the $PM_{2.5}$ and PM_{10} mass fractions and the average daily temperature in the second campaign in Mechelen

Supplementary Material

Supplementary Table 1

Correlation of PM elemental content, elemental and organic carbon, and meteorological parameters at various sampling sites and campaigns

	<i>Z1</i>	<i>Z2</i>	<i>B1</i>	<i>B2</i>	<i>P1</i>	<i>P2</i>	<i>H1</i>	<i>H2</i>	<i>M1</i>	<i>M2</i>	<i>W1</i>	<i>W2</i>
PM _{2.5}	EC, OC, TC, RH(a), PR(a), T(a), WS(a), P_a	T_a, EC	EC, PR(a), T(a), WS(a), P_a	EC, OC, TC, RH(a), PR(a), T_a, P_a(a), WS(a)	EC, OC, TC, RH, T_a(a), WS(a)	EC, OC, TC, RH(a), PR(a), T_a(a), P_a	EC, RH(a), PR(a), T_a(a), P_a, WS(a)	EC, OC, TC, RH, PR(a), T_a(a), P_a, WS(a)	PR, T_a(a), P_a(a)	EC, RH(a), PR(a), T_a(a), P_a, WS(a)	RH(a), P_a, WS(a)	EC, T_a, P_a
PM ₁₀	EC, OC, TC, RH(a), PR(a), T_a(a), WS(a), P_a	T, RH, EC	-	EC, OC, TC, RH(a), PR(a), T_a, WS(a)	EC, TC, RH, T_a(a), P_a, WS(a)	EC, OC, TC, RH(a), PR(a), T_a(a), P_a	EC, RH(a), PR(a), T_a(a), P_a, WS(a)	EC, OC, TC, T_a, P_a, WS(a)	PR(a)	EC, RH(a), PR(a), T_a(a), P_a, WS(a)	-	EC, RH(a), T_a
K	Al, Si, P, S, Cl, Ca, Mn, Ni, Cu, Zn, Br, Sr, Pb, EC, OC, TC	Al, Si, Ca, V, Mn, Fe, Cu, Zn, Rb, Pb, T_a	Al, Si, P, S, Cl, Ca, V, Mn, Fe, Cu, Zn, Br, Rb, Pb, EC, OC, TC, WS(a)	Al, Cr, Mn, Fe, Ni, Cu, Zn, Rb, Pb, EC, OC, TC, RH(a), PR(a), WS(a)	Al, Si, P, S, Ca, Mn, Fe, Ni, Cu, Zn, Rb, Pb, EC, OC, TC, WS(a)	Mn, Fe, Cu, Zn, Rb, Pb, EC, OC, TC, PR(a), T_a(a), P_a, WS(a)	Al, Si, P, S, Cl, Ca, Mn, Ni, Cu, Zn, Br, Pb, PM_{2.5}	Al, Ca, Mn, Ni, Cu, Rb, Sr, RH	Al, Si, Ca, Ti(a), Mn, Ni, Cu, Zn, Br	Fe, Cu	Si, Cr, Mn, Fe, Ni, Zn, Rb, Pb, T_a(a)	Al, Fe, Cu, Zn, Pb, EC
Ca	P, S, Cl, Ni, Cu, RH	Al, Si, Ti, Mn, Fe, Cu, Zn, Rb, Pb, EC	Al, Si, P, S, Cl, V, Mn, Fe, Ni, Cu, Zn, Br, EC	Al, Si, Ti, Cr, Mn, Fe, Ni, Cu, EC, T_a, WS(a)	Si, P, S, Mn, Fe, Ni, Cu, Zn, Se, Pb, EC, OC, TC, PM_{2.5}, WS(a)	-	Al, Si, P, S, Cl, Mn, Ni, Cu, Zn, Br, Pb	Al, Mn, Ni, Cu, Rb, Sr, RH, PR	Al, Si, Cl, Mn, Ni, Cu, Zn, Br	Mn, Ni, Cu, Sr, Pb(a), EC(a), PR, WS	Cr, RH	PR, T_a(a)
Ti	T_a	Al, Si, Cr, Mn, Fe, Zn, Pb, EC	P, V, Ni	Al, Si, V, Cr, Mn, Fe, Ni, Cu, EC, T_a, WS(a)	P, Al, Si, Se, PM_{2.5}	V, Si	PM_{2.5}, RH, P_a(a)	EC, PM₁₀, P_a	S(a), Cl, PM_{2.5}(a)	-	Cl(a), Ni, Br,	-

V	Al, P, S, Br	Al, Si, Ni, Rb, Sr, PM ₁₀	Al, Si, P, S, Cl, Mn, Fe, Ni, Br, EC	Al, Si, Fe, Ni, WS(a)	Ni, PM _{2.5} (a), WS(a), P _a	-	-	-	PM _{2.5}	-	P, S, Ni, Br, Pb,	Ni, PM _{2.5} , WS
Cr	Al, Si, Cl, Mn, Sr, RH(a), EC, TC	PM _{2.5} , WS(a)	Cl, Se	Al, Si, Mn, Fe, Cu, RH(a)	-	RH(a), WS(a)	-	Mn, T _a (a)	Mn, Ni, Cu, Zn, Br, PM _{2.5} (a)	Mn, Fe, Zn,	Si, Mn, Fe, Zn, Rb, Pb, PR	Zn, Pb, PM _{2.5}
Mn	Al, Si, Cl, Fe, Zn, Br, Rb, Pb, EC, TC	Al, Si, Fe, Cu, Zn, Rb, T _a	Al, Si, P, S, Cl, Fe, Zn, Br, Rb, Pb, EC, OC, TC	Al, Si, Fe, Ni, Cu, Zn, Rb, Pb, EC, OC, TC, RH(a), WS(a)	Si, P, S, Cl(a), Fe, Ni, Cu, Zn, Se, Pb, EC, OC, TC, PM _{2.5} , WS(a)	Fe, Cu, Zn, Rb, Pb, EC, OC, TC, PR(a), P _a , WS(a)	Al, Si, P, S, Cl, Ni, Cu, Zn, Br, Pb, PM _{2.5}	Al, Fe, Ni, Cu, Zn, Rb, Pb, PM _{2.5} , EC	Al, Si, Fe, Ni, Zn,	Fe, Ni, Cu, Zn, PR	Fe, Zn, Br, Rb, Pb, T _a (a)	Fe, Ni, Cu, Zn, Pb, RH(a)
Fe	Al, Cl, Zn, Rb, Pb, EC, OC, TC, WS, T _a , PR, P _a (a)	Al, Si, Zn, Rb, Pb, EC	Al, Si, P, S, Cl, Cu, Zn, Br, Pb, EC, OC, TC, T _a (a), P _a , PR(a), WS(a)	Al, Si, Ni, Cu, Zn, Rb, Pb, EC, OC, TC, RH(a), T _a , WS(a)	Al, Si, P, S, Ni, Cu, Zn, Br, Rb, Sr, Pb, EC, OC, TC, PM _{2.5} , WS(a)	Cu, Zn, Rb, Pb, EC, OC, TC, PR(a), P _a , WS(a)	S, Pb, EC, P _a , WS(a)	Zn, Pb, EC, OC, TC, WS(a), P _a	Al, Si, P, S, Sr, Pb, RH, T _a (a)	Si, Zn, Pb, EC	Al, Si, P, S, Cu, Zn, Br, Rb, Pb,	Al, Si, Cu, Zn, Pb, EC, RH(a), T _a , WS(a)
Ni	Si, P, Cl, Cu, Zn, Br, Sr, OC, T _a , RH, PR	Al, Si, PM ₁₀	Se, T _a	Al, Si, Cu, EC, OC, TC, T _a , WS(a)	P, Cu, EC, PM _{2.5} , WS(a)	-	Al, Si, P, S, Cl, Cu, Zn, Br, Sr, Pb,	Al, Cu, Rb, RH, PR	Al, Cu, Zn, Br, PR(a)	Cu, Pb(a), EC(a), RH, PR, WS	Al, P, S, Cu, Br, Pb, PM _{2.5}	PM _{2.5}
Cu	Si, P, Cl, Zn, Br, EC, OC, TC, T _a , RH, PR	Zn, Rb, Pb	Al, Si, P, S, Cl, Zn, Br, Pb, EC, OC, TC, PR(a), P _a , WS(a) WD(a)	Al, Si, Zn, Rb, Pb, EC, OC, TC, RH(a)	P, S, Zn, Br, Pb, EC, OC, TC, WS(a)	Zn, Rb, Pb, EC, OC, TC, PM _{2.5} , PR(a), WS(a)	Al, Si, P, S, Cl, Zn, Br, Pb, PM _{2.5}	Al, Rb, RH	Al, Zn, Br, Rb(a)	Sr, RH	Al, P	Al, Si, Zn, Pb, EC, T _a
Zn	Si, Cl, Br, Pb, EC, OC, TC, T _a , RH,	Al, Si, Rb, Pb, RH(a), T _a , EC	Al, Si, S, P, Cl, Br, Rb, Pb, EC, OC,	Al, Rb, Pb, EC, OC, TC, RH(a)	Si, P, S, Br, Pb, EC, OC, TC, PM _{2.5} ,	Rb, Pb, EC, OC, TC, PR(a), P _a ,	Al, Si, S, Cl, Br, Pb, P, PM _{2.5}	Al, Pb, EC	Al, Br	Si, EC	Si, Br, Rb, Pb, T _a (a)	Pb, EC, PM _{2.5}

	PR		TC, P_a, WS(a)		WS(a)	WS(a)						
Rb	Se, Pb, EC, OC, TC, PM₁₀	Si, Pb, PM_{2.5}	Al, Si, P, S, Cl, EC, OC, TC	Pb, EC, OC, TC, WS(a)	EC, OC, TC, WS(a)	Pb, EC, OC, TC, RH, PR(a), P_a	-	Al, Si(a)	Sr, PM_{2.5}, RH, PR	Al, EC(a)	S, Pb	-
Sr	Al, Si	-	-	-	Si	-	Al	Al	Al	-	-	P_a(a)
Pb	Al, Cl, EC, OC, TC	Al, Si, T_a, WS	Al, Si, P, S, Cl, PM_{2.5}, PM₁₀, EC, OC, TC, T_a(a), P_a, WS(a)	EC, OC, TC	Al, Si, P, S, EC, OC, TC, PM_{2.5}, WS(a)	Al, EC, OC, TC, PM_{2.5}, PR(a), WS(a)	Al, Si, P, S, Cl, PM_{2.5}	EC, OC, TC, WS(a), P_a	Al, Si, P, S, RH, T_a(a),	EC, RH, WS	Al, Si, P, S, T_a(a)	EC, PM_{2.5}, WS(a)
Al	Si, P, S, EC	Si, EC, T_a	Si, P, S, Cl, EC, OC, TC, PM_{2.5}, PM₁₀	Si, EC, OC, TC, RH(a), T_a, WS(a)	Si, P, S, OC, PM_{2.5}, T_a	Si	Si, P, S, Cl, PM_{2.5}	RH,	Si, PM₁₀, RH, T_a(a)	Si	P, S, PM_{2.5},	Si, EC, T_a, WS(a)
Si	P, S, Cl	-	P, S, Cl, EC, OC, TC	EC, T_a, WS(a)	P, S, Cl(a), EC, OC, TC, PM_{2.5}, T_a	-	P, S, Cl, PM_{2.5}	EC, PM₁₀, T_a, P_a	P, S, RH, T_a(a)	-	RH(a), T_a(a)	EC, RH(a), T_a
Br	Al, Si, P, S, Cl, Rb, Pb, EC, OC, TC	n.a.	Al, Si, P, S, Cl, Rb, Pb, EC, OC, TC, RH, WS(a)	n.a.	P, S, Pb, EC	n.a.	Al, Si, P, S, Cl, Pb, PM_{2.5}	n.a.	PM_{2.5}(a)	n.a.	P, S, Pb,	n.a.
S	P, Se, EC, P_a, T(a), WS(a)	n.a.	P, Cl, EC, OC, TC, WS(a)	n.a.	P, EC, OC, TC, RH, WS(a)	n.a.	P, Cl, PM_{2.5}	n.a.	P, RH, T_a(a), PR	n.a.	P, PM_{2.5}	n.a.
Cl	T_a, RH, PR, WS, P_a(a)	n.a.	P, EC, OC, TC, WS(a)	n.a.	OC(a), TC(a), T_a(a)	n.a.	P, PM_{2.5}	n.a.	T_a	n.a.	PM_{2.5}(a), RH	n.a.
P	P_a, WS(a), EC, OC	n.a.	EC, OC, TC, WS(a), RH	n.a.	EC, OC, TC, WS(a)	n.a.	PM_{2.5},	n.a.	RH, T_a(a)	n.a.	Se, PM_{2.5}	n.a.

Abbreviations: EC – elemental carbon, OC – organic carbon, TC – total carbon, RH – relative humidity, PR – precipitation, WS – wind-speed, T_a – air-temperature, P_a – atmospheric pressure, (a) – anti correlation, n.a. – not analyzed; bold letter: correlation is significant at the 0.01 level (strong correlation), normal letter: correlation is significant at the 0.05 level (some correlation)

Supplementary Table 2: PCA of elements at Petroleumkaai

Rotated Component Matrix [†]						
Species	Factor					
	1	2	3	4	5	6
Al	-0.01	0.94	-0.17	0.05	0.21	0.01
Si	0.09	0.91	-0.18	0.13	0.18	-0.13
P	0.73	0.58	-0.11	0.26	-0.07	-0.04
S	0.79	0.47	0.08	0.26	-0.08	-0.14
Cl	-0.27	-0.15	0.37	-0.22	-0.58	-0.11
K	0.92	0.11	0.26	-0.07	0.02	-0.02
Ca	0.55	-0.43	0.06	-0.38	-0.07	0.56
Ti	0.16	0.21	0.14	0.85	-0.15	0.17
V	-0.31	-0.04	-0.13	0.70	0.43	-0.10
Cr	-0.09	0.00	-0.57	0.04	-0.02	-0.76
Mn	0.00	-0.07	0.10	0.23	-0.27	0.80
Fe	0.90	-0.19	-0.17	-0.15	0.03	0.14
Ni	-0.07	-0.06	0.95	0.03	-0.02	0.15
Cu	0.87	-0.13	-0.12	-0.17	0.31	0.23
Zn	0.96	-0.12	-0.06	0.11	-0.02	0.07
Se	-0.26	-0.28	-0.30	0.11	-0.69	0.18
Br	0.48	0.20	0.04	0.51	0.48	0.17
Rb	0.01	-0.40	0.81	0.07	0.02	0.16
Sr	-0.28	0.07	-0.02	0.03	0.67	-0.30
Pb	0.88	0.38	-0.03	0.10	-0.01	-0.01
<i>Eigenvalue</i>	6.58	4.24	2.40	1.72	1.43	1.02
<i>% Variance Explained</i>	32.90	21.19	12.02	8.59	7.17	5.12
<i>Major Sources</i>	<i>Refinery</i>	<i>Crustal</i>	<i>Brake wear +</i>	<i>Heavy oil burning</i>		

*Extraction Method: Principal Component Analysis.
 Rotation Method: Varimax with Kaiser Normalization.
[†]Rotation converged in 14 iterations.*

Supplementary Table 3: PCA of elements at Borgerhout

Rotated Component Matrix [†]					
Species	Factor				
	1	2	3	4	5
Al	0.97	0.14	0.04	-0.09	0.13
Si	0.95	0.22	0.05	-0.01	0.15
P	0.96	0.20	-0.04	-0.08	-0.02
S	0.94	0.16	-0.09	-0.11	-0.06
Cl	0.70	0.47	0.10	-0.23	0.09
K	0.94	0.31	-0.01	0.02	0.00
Ca	0.66	0.60	0.05	0.16	0.26
Ti	-0.19	-0.33	0.14	0.88	-0.17
V	0.68	0.10	-0.11	0.54	-0.18
Cr	0.11	0.03	0.94	0.15	0.01
Mn	0.39	-0.29	-0.62	0.25	0.19
Fe	0.51	0.80	0.13	-0.07	-0.11
Ni	-0.46	0.26	-0.30	0.45	0.37
Cu	0.29	0.93	0.05	-0.16	0.03
Zn	0.74	0.58	-0.09	-0.07	-0.09
Se	0.10	-0.01	-0.12	-0.10	0.80
Br	0.86	0.20	0.07	-0.08	0.26
Sr	-0.13	0.63	-0.20	0.07	-0.65
Pb	0.49	0.71	0.29	-0.17	-0.13
<i>Eigenvalue</i>	9.96	2.57	1.59	1.52	1.12
<i>% Variance Explained</i>	52.40	13.52	8.39	8.01	5.9
<i>Major Sources</i>	<i> Vehicular </i>	<i> Non-exhaust </i>	<i> Non-ferrous industry </i>	<i> Heavy oil burning </i>	

[†] Rotation converged in 11 iterations.

Supplementary Table 4: PCA of elements at Zelzate

Rotated Component Matrix †					
Species	Component				
	1	2	3	4	5
Al	0.78	0.09	0.11	0.31	0.12
Si	0.66	-0.30	0.39	0.09	-0.08
P	0.70	0.64	-0.13	0.08	0.10
S	0.62	0.73	-0.03	0.01	0.12
Cl	0.88	0.02	0.16	-0.09	-0.34
K	0.89	0.00	0.23	-0.06	0.20
Ca	0.06	-0.06	-0.08	0.93	0.08
Ti	-0.22	-0.05	-0.03	-0.14	-0.77
V	0.07	0.34	0.79	-0.01	-0.16
Cr	-0.16	0.83	0.23	0.03	-0.15
Mn	0.38	-0.09	0.70	-0.18	0.34
Fe	0.77	0.21	0.20	0.47	0.12
Ni	0.45	0.39	0.43	0.26	0.10
Cu	0.67	0.14	-0.10	0.49	0.21
Zn	0.92	0.07	0.18	0.11	0.21
Br	0.79	0.42	0.30	0.08	0.08
Sr	-0.16	-0.37	-0.08	-0.50	0.49
Rb	0.22	0.49	0.36	0.02	0.40
Pb	0.88	0.20	0.02	0.04	0.24
<i>Eigenvalue</i>	8.87	2.11	1.79	1.30	1.13
<i>% Variance Explained</i>	46.70	11.12	9.40	6.82	5.96
<i>Major Sources</i>	Coke-Oven	Combustion	Traffic (diesel)	Soil/road dust	

† Rotation converged in 15 iterations.

Supplementary Table 5: PCA of elements at Hasselt

Rotated Component Matrix [†]					
Species	Component				
	1	2	3	4	5
Al	-0.07	-0.14	-0.11	0.15	-0.74
Si	0.99	-0.08	0.03	0.04	0.05
P	-0.09	-0.01	0.60	-0.23	-0.03
S	0.90	0.36	-0.09	0.04	0.03
Cl	1.00	-0.05	0.00	0.00	-0.01
K	0.99	0.02	0.01	0.07	-0.01
Ca	0.99	-0.09	0.04	0.01	-0.01
Ti	0.05	-0.53	-0.17	0.31	0.67
V	-0.10	-0.07	-0.06	-0.92	0.09
Cr	-0.07	0.80	-0.16	-0.06	0.37
Mn	-0.02	-0.45	-0.22	0.53	0.08
Fe	0.07	0.93	0.01	0.06	-0.16
Ni	0.99	-0.14	0.03	0.03	-0.01
Cu	0.99	-0.01	-0.01	0.04	-0.04
Zn	0.99	0.11	-0.03	0.06	-0.03
Br	1.00	-0.03	0.01	-0.01	0.03
Rb	0.38	0.03	0.78	0.25	-0.10
Sr	-0.24	-0.08	0.67	0.06	0.42
Pb	0.86	0.45	-0.09	0.08	0.06
<i>Eigenvalue</i>	<i>9.73</i>	<i>2.52</i>	<i>1.58</i>	<i>1.33</i>	<i>1.25</i>
<i>% Variance Explained</i>	<i>51.20</i>	<i>13.24</i>	<i>8.33</i>	<i>6.98</i>	<i>4.61</i>
<i>Major Sources</i>	<i>Vehicular</i>	<i>Road Dust</i>	<i>Biomass</i>	<i>Tyre wear / brake lining</i>	

[†] Rotation converged in 8 iterations.

Supplementary Table 6: PCA of elements at Wingene

Rotated Component Matrix †						
Species	Component					
	1	2	3	4	5	6
Al	0.39	-0.16	0.79	0.14	-0.06	-0.24
Si	0.47	-0.46	-0.11	-0.11	-0.06	-0.44
P	0.44	0.19	0.51	0.53	0.35	-0.02
S	0.47	0.33	0.25	0.56	0.47	-0.04
Cl	-0.22	-0.06	-0.03	0.89	0.00	-0.07
K	0.84	0.05	0.17	-0.20	-0.20	0.21
Ca	0.18	-0.15	0.19	0.03	0.20	0.81
Ti	0.17	0.04	-0.23	-0.13	-0.17	0.79
V	0.01	0.67	-0.10	0.56	-0.06	-0.19
Cr	-0.36	-0.22	0.08	0.08	-0.76	-0.01
Mn	0.05	-0.75	-0.07	0.07	0.01	-0.01
Fe	0.78	-0.22	0.38	0.05	0.11	-0.11
Ni	0.39	0.74	0.25	0.16	0.22	-0.02
Cu	0.06	0.13	0.78	-0.09	-0.06	0.26
Zn	0.84	-0.08	0.20	0.02	-0.03	0.16
Se	0.30	0.18	-0.02	0.54	-0.60	0.31
Br	0.19	0.43	0.67	-0.01	0.22	-0.13
Rb	0.67	0.35	-0.03	-0.13	0.12	0.16
Sr	-0.36	-0.05	0.11	0.25	0.65	0.15
Pb	0.88	0.22	0.15	0.23	0.06	0.09
<i>Eigenvalue</i>	5.98	2.99	2.05	1.79	1.52	1.39
<i>% Variance Explained</i>	29.88	14.97	10.25	8.95	7.61	6.94
<i>Major Sources</i>	Biomass	Fuel oil	Crustal	Sea salt		

† Rotation converged in 10 iterations.

Supplementary Table 7: PCA of elements at Mechelen

Rotated Component Matrix [†]					
Species	Component				
	1	2	3	4	5
Al	0.87	0.30	-0.09	-0.33	0.02
Si	0.50	0.84	-0.16	-0.09	0.07
P	-0.06	0.96	0.12	-0.14	-0.09
S	0.15	0.97	-0.12	0.09	0.09
Cl	0.69	0.19	0.22	-0.05	0.01
K	0.97	0.23	-0.03	0.02	-0.05
Ca	0.93	0.35	-0.02	0.02	-0.03
Ti	0.00	0.18	-0.25	-0.86	0.21
V	-0.14	0.24	0.11	-0.16	0.91
Cr	-0.23	-0.42	0.72	-0.13	0.49
Mn	-0.29	0.18	-0.59	0.63	-0.15
Fe	0.10	0.96	-0.18	-0.15	0.13
Ni	0.16	-0.13	0.91	0.18	-0.07
Cu	0.79	0.16	0.52	0.04	0.10
Zn	0.98	-0.09	-0.03	-0.01	-0.09
Se	0.83	-0.45	-0.21	0.13	0.00
Br	0.93	0.09	0.21	-0.01	-0.25
Rb	0.25	0.77	-0.13	0.44	0.25
Sr	-0.06	0.55	-0.13	0.62	0.49
Pb	0.15	0.97	-0.12	0.09	0.09
<i>Eigenvalue</i>	<i>8.09</i>	<i>5.35</i>	<i>2.35</i>	<i>1.88</i>	<i>1.11</i>
<i>% Variance Explained</i>	<i>40.44</i>	<i>26.75</i>	<i>11.77</i>	<i>9.39</i>	<i>5.57</i>
<i>Major Sources</i>	<i>Local Industry</i>	<i>Road Dust</i>	<i>Tyre wear / brake lining</i>		

[†] Rotation converged in 8 iterations.