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Determination of car on-road black carbon and particle number emission factors and comparison between mobile and stationary measurements

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Abstract

We have used two methods for measuring emission factors (EF) in real driving conditions on five cars in a controlled environment: the stationary method, where the investigated vehicle drives by the stationary measurement platform and the composition of the plume is measured; and the chasing method, where a mobile measurement platform drives behind the investigated vehicle. We measured EF of black carbon and particle number concentration. The stationary method was tested for repeatability at different speeds and on a slope. The chasing method was tested on a test track and compared to the portable emission measurement system. We further developed the data processing algorithm for both methods, trying to improve consistency, determine the plume duration, limit the background influence and facilitate automatic processing of measurements. The comparison of emission factors determined by the two methods showed good agreement. EFs of a single car measured with either method have a specific distribution with a characteristic value and a long tail of super emissions. Measuring EFs at different speeds or slopes did not significantly influence the EFs of different cars, hence we propose a new description of vehicle emissions that is not related to kinematic or engine parameters, rather we describe the vehicle EF with a characteristic value and a "super emission" tail.

1 Introduction

Regulation of the particulate air pollution addresses the health and climate effects of aerosolized matter (Ferrero et al., 2014; Ramanathan and Feng, 2009; Pope et al., 2009). One of the main sources of air pollution is traffic. To estimate the traffic contribution to air pollution different methods can be used (Perrone et al., 2014; Moroni et al., 2012; Wang et al., 2011; Kuhns et al., 2004). The bottom-up approach attempts to calculate the emissions from the vehicle fleet composition and vehicle emission factors (EF); however the difficulty of these methods lies in the details of different vehicle

types needed, like: vehicle types (cars, heavy and light duty); the maintenance of engines, their use in different environments (city, highway, regional roads) and weather conditions.

There have been many tests and statistical analyses performed on different vehicles and driving parameters to include these variables to the highest possible degree. Franco et al. (2013) summarized these in their review of EF development. They define EFs as different empirical functional relations of emitted pollutants to the activity that causes them. Most standardized and robust EFs were found to be produced in laboratories using dynamometer tests with prescribed driving cycles. These tests can produce aggregated or bag results with respect to mean speed or some other kinematic parameter (e.g. mean acceleration) of a driving cycle; or instantaneous emission data, where measured emissions values can be related to recorded instantaneous kinematic or engine covariates (Perrone et al., 2014).

The estimation of vehicle emissions increased in complexity as emission regulations came into effect; before regulation emissions were high and well correlated with vehicle average speed while, after the introduction of newer and more sophisticated engines and after-treatment systems, vehicle emission factors were no longer correlated to average driving speed (Ntziachristos and Samaras, 2000) and the instantaneous approach became more difficult to apply as well (de Haan and Keller, 2004). Ajtay et al. (2005) and Weilenmann et al. (2001) found that the measured engine parameters and emission measurements diverge as the exhaust gas is transported through the car, sampling lines and analyzers, resulting in up to 15 s delayed and broadened exhaust parameters. They found that further discrepancies arise from incompatible time resolutions of emission and engine related measurements: the time characteristic for emission related parameters depends on the operation of the engine and is of the order of magnitude of several tenths of a second, contrasting most emission related measurements which operate on one second time base.

In order to validate emission model predictions and to compare their performance to actual vehicle emissions, so-called "real world" EF measurement techniques have been

developed (Franco et al., 2013). These employ different techniques for measuring numerous in-use vehicles in actual traffic situations; e.g. measurements were performed on-road by employing remote sensing, chasing, or on-board detection, or in tunnels (some of the first such experiments were those of Hansen and Rosen, 1990; Bishop and Stedman, 1996; Weingartner et al., 1997).

The "real world" methods have been described as less precise than the dynamometer studies because the tests are not as repeatable as dynamometer ones due to absence of standard cycles and presence of additional source of variability, such as environmental or traffic conditions, driver behavior or highly transient operation (Franco et al., 2013). However, we believe these measured emission factors could also be a beneficial addition to models, if they would be properly incorporated with real world emission measurements of a large number of random in-use vehicles resulting in a proper representation of the whole vehicle fleet emissions.

Legislation limits the particle mass concentration (PM) both at the sources of emissions (e.g. vehicle emission standards; one of the latest is Euro 5 and Euro 6 directive for light passenger and commercial vehicles 715/2007/EC); and in ambient concentrations (2008/50/EC). However, it has been demonstrated that black carbon (BC) and particle number concentration (PN), both showed direct or indirect health and climate effects (e.g. Bond et al., 2013; Janssen et al., 2011; Vinzents et al., 2005, and references there-in).

Here a comparison of two methods for measuring the real-world BC and PN EFs of a single vehicle is presented. The first method is the stationary method where EFs are calculated from the instantaneous rise of pollutants as a vehicle passes the measuring station (Hansen and Rosen, 1990; Ban-Weiss et al., 2009; Dallmann et al., 2011); the second is the chasing method where vehicle emissions are measured by driving a mobile station behind the measured vehicle on the road (Wang et al., 2011, 2012). Both methods assume equal dilution of all emitted pollutants and use the carbon balance equation to calculate the EF as the pollutant mass per kg fuel consumed. Assuming complete combustion of fuel, where almost all carbon in fuel is oxidized to CO_2

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(Hansen and Rosen, 1990; Ban-Weiss et al., 2009; Dallmann et al., 2011), the fuel consumption can be estimated by measuring the CO_2 emissions. Wang et al. (2011) also measured CO, unburned hydrocarbons and soot to obtain a more accurate EF; however it has been demonstrated by Dallmann et al. (2011) that omitting these from calculation yields only a 5 % positive bias in calculated EF.

To improve the knowledge on "real world" EFs, we applied these methods, so far used on heavy duty vehicles and pre EURO1 passenger cars (91/441/EEC) (Hansen and Rosen, 1990; Ban-Weiss et al., 2009; Dallmann et al., 2011; Wang et al., 2011; 2012), to passenger cars in use in the European Union today. These have smaller fuel consumption and thus emit less CO_2. We compared the two methods in controlled conditions on a training track using different driving scenarios, to see how these real-world EFs are affected by vehicle speed, position of the measurement station and dynamics of the terrain. We further developed the two methods: the stationary method to improve detection of the plume duration; and the chasing method to obtain the time evolution of the EF.

While measuring emissions of a vehicle by chasing it on a road the dilution changes with vehicle shape, tailpipe position, the chasing distance, speed, and changing of the chased vehicle exhaust flow (Chang et al., 2009; Vogt et al., 2003). To investigate the effect of these parameters during the determination of the EF by chasing, and to further explain the results of the running integration calculation, we applied the chasing method to a vehicle equipped with portable emission measurement system (PEMS), which has shown to be well correlated with dynamometer measurements (Weiss et al., 2011; Johnson et al., 2009).

2 Experimental setup

Experiments were conducted on two test circuits: the training track of the Slovenian national automobile and motorcycle association and the Italian Monza racing circuit. On the Slovenian track both chasing and stationary measurements (Sect. 2.1) were

conducted on five in-use cars, while on the Italian circuit the chasing measurements were conducted on a vehicle that was equipped with PEMS (Sect. 2.2).

2.1 Chasing vs. stationary measurements

This part of the experiment was conducted on the training track of Slovenian national automobile and motorcycle association on 23 December 2011. The track is a 1.3 km long circuit with two sharp turns and a slope (3 % grade). Along the track two measuring sites for the stationary method have been set up: one on flat part of the road and one on the slope. In the stationary part of the experiment each car passed both stations in three different scenarios: with constant speed of 50 km h^{-1} and 90 km h^{-1}, and while accelerating from a stationary position about 10 m in front of the station. Each scenario was repeated five times. The second part of the experiment consisted of chasing a car with our mobile measuring station, making 5 laps on the training track circuit.

Both the chasing method (Wang et al., 2011) and the stationary method (Hansen and Rosen, 1990) were applied by measuring the exhaust of five different passenger cars. Three cars had a compression ignition engine (diesel) and were compliant with Euro 3 emission standard (98/69/EC) with production years 2000, 2002 and 2004; and two were compliant with Euro 5 standard (715/2007/EC), of which one had a compression (diesel) and the other spark ignition engine (gasoline). The vehicles are henceforth referred to as EURO3 (00), EURO3 (02), EURO3 (04), EURO5 (d) and EURO5 (g), respectively. The Euro 3 cars were each driven by their owners, the Euro 5 cars were driven by the same driver that drove the EURO3 (02), who was a professional driver.

This part of the study was designed: (a) to test the applicability and the reproducibility of the two measurement methods on personal vehicles (cars); (b) to see if EF's measured with the stationary technique would be higher or more repeatable on the slope where we would expect a more constant engine load; (c) to see the possible difference in EFs during stationary measurements having vehicles pass at two different speeds and while accelerating; and (d) to compare the results of the two methods.

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2.2 Chasing vs. tailpipe measurements

This part of the experiment was performed in Monza (Italy) on part of the Monza race track in 24 May 2012. The part of the track where we conducted measurements is flat and approximately 700 m long. In one chase the driver of the tested vehicle equipped with PEMS was instructed to steadily accelerate and shift up to the 4th gear, then re-duce speed, make a sharp U-turn and return to the starting position at the end of which the mobile measurement station lagged behind the chased vehicle. This was repeated three times. The chased vehicle in this experiment was a Euro 3 (98/69/CE) diesel van equipped with PEMS. This experiment was set up so we could better understand the results of the chasing approach.

2.3 Instrumentation

Instruments used for stationary measurements on the flat part of the training track were the Dekati Electrical Low Pressure Impactor (ELPI+) for measuring particle num-ber concentrations (PN) and number size distributions, an Aethalometer prototype AE33 (prototype ver. P10, made by Aerosol d.o.o.) for BC measurements, which was equipped with a cyclone inlet (aerodynamic diameter cut-off at 1.7 µm) to eliminate measuring re-suspended particles; and a Vaisala flow-through Carbocap GMP343 for measuring CO_2 concentrations. These instruments were operated with 10, 7 and 7 L min^{-1} flows respectively. Sampling inlets were positioned next to the road about 10 cm from the ground.

Stationary measurements on the sloped part of the track and chasing measure-ments were performed with the mobile measurement platform, which was a small van equipped with TSI Fast Mobility Particle Seizer (FMPS) for PN measurements and num-ber size distributions, a second Aethalometer AE33 and another Carbocap GMP343. These instruments also operated with 10, 7 and 7 L min^{-1} flows, respectively. The in-lets of the mobile station were attached under the backdoor window on the right-hand

side of the station. All equipment in the mobile station was powered by three 100 Ah batteries.

All instruments were operated with one second time resolution with the exception of Carbocap GMP343 which needs 2 s to perform each measurement.

All sampling lines were kept as short as possible (no longer than 2.5 m). Isokinetic sampling was not attempted as it had negligible effect on our measurements (Wang et al., 2011; Baron and Willeke, 2001).

As the PN was measured using two different measuring systems, the total PN measured by ELPI+ and the FMPS were compared before the field activity. The instruments were running overnight measuring ambient concentrations. These showed good correlation $R^2 = 0.91$, but because the slope was 8.15 and the intercept −6630.9 we adjusted the ELPI+ data. After the adjustment was made, the responses of instruments on the test night were the same and on the day of the training track measurements the two showed similar background concentrations.

The data from the two Aethalometers was compensated off-line for loading effects using the compensation algorithm implemented also on-line in the Aethalometer AE33 (Drinovec et al., 2014). After the compensation was applied the data from the two Aethalometers used in the training track experiment featured the regression slope 0.93 and intercept 0, with $R^2 = 0.99$. The two CO_2 probes also correlated well: $R^2 = 0.99$; slope 0.88 and intercept 0.

In the Monza test the chased vehicle was equipped with a Portable Emission Measurement System (PEMS) (Weiss et al., 2011); which consists of exhaust gas analyzers (CO_2, CO, THC, and NO_x), an Exhaust Flow meter (EFM), a Global Positioning System (GPS), and a weather station. The mobile platform had an Aethalometer AE33 (ver. P20) equipped with a cyclone inlet (aerodynamic diameter cut off at 1.7 µm) for the BC measurements, and flow through Carbocap GMP343 sensor for the CO_2 measurements; powered with one 100 Ah battery. The sampling line was again shorter than 2.5 m; the inlet was mounted on the right hand-side external mirror.

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3   Emission factor calculations and data processing

We calculated the EFs using the carbon balance method (Hansen and Rosen, 1990):

$$EF = \frac{\int_i^j (P_j - P_i)\mathrm{d}t}{\int_i^j (CO_{2j} - CO_{2i})\mathrm{d}t} \cdot w_c \qquad (1)$$

5 where w_c represents the carbon fraction of fuel and was set to 0.861 for diesel and 0.864 for gasoline (Huss et al., 2013); P in the equation represents the pollutant, either PN or BC; the subscripts i and j denote the time of the beginning and the end of the measured plume, respectively.

In stationary measurements rapid concentration increases following each pass of the
10 measured vehicle were measured. We included all peaks where the pollutant increase was higher than 3 standard deviations of the background concentrations. For each successful plume capture the background concentrations were calculated as an average of 20 s before the inflection point. This point marked the beginning of the plume. The end of the plume was set to the time when the pollutant concentration time derivative
15 reaches the minimum (Fig. 1). The data was smoothed with a 5 s running average and the time derivative calculated from 3 measurement points previous to the time of the calculated derivative. Our integrated peaks lasted around 10–20 s, similar to plume durations previously reported by Ban-Weiss et al. (2009) and Dallmann et al. (2011). We determined the beginning and the end of the plume from BC measurements rather than
20 from the CO_2 peak (Ban-Weiss et al., 2009). We found that performing the same integration analysis on all of the pollutants produced more consistent results. We noticed (similarly to Dallmann et al., 2011) that there was a difference in the response time of the instruments used.

During chasing measurements dynamic time resolved pollutant concentrations in the
25 plume of the investigated vehicle were measured: an example measured during one lap on the Slovenian track is reported in Fig. 2. The EFs were calculated and compared using two approaches. In the first, we subtracted the background concentrations and

then performed running integration with a 10 s time window to obtain a time resolved EF. The second approach was bulk plume integration performed according to Wang et al. (2011), who integrated each measured pollutant from the beginning to the end of the chase.

For the chasing method it was most important to find the correct background for CO_2, because the determination of the background influences the accuracy of the EF – as Wang et al. (2011) reported, changing the CO_2 background for 2 % changed the calculated EF for up to 40 % (only up to 5 % for changing BC or PN baseline). Thus, we analyzed the background and its influence on our calculations and, as reported in Sect. 4.2.2, we found a smaller variation of the EF due to the change of the CO_2 background using our EF determination. For each car chase we subtracted the background which we set as a linear trend between the 20 s average of data we measured before and after the chase period when no car emissions were measured. The background did not change much in this period (Fig. 2), but we expect that during real-life on-road measurements, where spatial concentrations may vary more than they did in the controlled conditions of the training track, the background might change – hence the need for the linear approximation of the background during the chasing period. To describe the background variation we calculated the standard deviation of the background from the measurements before the chase. Two standard deviations covered at least 90 % of the variation. We added and subtracted two standard deviations from the background to determine the influence of background variation on the calculated EFs; results are discussed in Sect. 4.2.2.

4 Results and discussion

Results from the experimental campaign are here reported and organized as follows: results from both the stationary and chasing methods are discussed in Sects. 4.1 and 4.2, respectively, while, a comparison between the two approaches is reported in Sect. 4.3.

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4.1 Stationary measurements

First interest of this research was to see if this method was applicable on today's in-use cars that have smaller fuel consumption and therefore emit less CO_2 than trucks, busses and pre-euro cars, on which this method was so far used (e.g. Ban-Weiss, 2009; Hansen and Rosen, 1990). To calculate an EF according to Eq. (1), we first need a valid measurement of CO_2. The capture of increased CO_2 concentrations in this experiment was high, out of 150 vehicle passes of the measurement stations we managed to measure 125 CO_2 peaks. This means we can detect "fuel consumption" of most vehicles passing the measuring site. We failed to detect the CO_2 transient increase 13 times during 90 km h^{-1} passes and 12 times during 50 km h^{-1} passes of different vehicles. Considering results of the chasing experiment performed in Monza (see Sect. 4.2.3) we could attribute this to a probable combination of low mass exhaust flow rate, high speed and consequently higher dilution rate.

To calculate an EF, an increase of pollutant is also required, in this study either BC or PN. Both Euro 5 vehicles had a small number of calculated EFs, due to the absence of BC and PN peaks. The few BC EFs of the Euro 5 vehicles that were measured were at least 10 times smaller than BC EFs of the Euro 3 vehicles. We could measure more PN EF of the two Euro 5 vehicles (15 in-total), of which EURO5 (d) had the highest PN EF of all vehicles measured with the stationary method, though its total number of PN EFs was the smallest (we could calculate only 3). The low values of the BC EFs for both EURO5 vehicles indicate improved technology in lowering soot emissions. The infrequent high PN emissions may be due to nucleation of the exhaust vapors after being emitted in the ambient environment, which would be suppressed in the presence of soot nucleation (Kittelson, 1998).

In total, 88 and 67 PN and BC EF respectively were calculated; during 9 passes the Aethalometer advanced the tape and no BC measurements were taken. Table 1 summarizes EFs for individual cars in terms of average, median, minimum, maximum and the count of the obtained EFs.

The average values and the span of the three EURO3 differed for both BC and PN EFs (Table 1). The highest BC emitter was EURO3 (04), whose average EF was twice as high as EURO3 (00), which had the lowest BC EF of the three EURO3 cars. EURO (04) had also the highest minimum and maximum BC EF of the three. The results are different for PN EF, where the highest emitter turned out to be EURO3 (00). We will further address the differences in vehicles and BC vs. PN EFs also in comparison to the chasing method results in Sect. 4.3.2.

Position of the measuring station on slope or flat road did not show significant changes in measured PN EFs, average on slope was 1.24 and $1.07 \times 10^9 \, \# \, kg^{-1}$ on flat road (Fig. 3), respectively. The median did not change for PN EF (0.88 and $0.87 \times 10^9 \, \# \, kg^{-1}$ on slope and flat road measurements, respectively). Average of BC EFs measured on the slope was $0.50 \, g \, kg^{-1}$ and on flat road $0.40 \, g \, kg^{-1}$; the median BC EF values were twice as high ($0.53 \, g \, kg^{-1}$) on slope as they were on the flat road ($0.27 \, g \, kg^{-1}$). Both BC and PN EFs feature tails toward the larger EF values. We also notice that we were more successful in measurements on the flat road where we measured 9 CO_2 peaks more than on the slope. This could be due to the position of the inlets – on the flat part of the track they were closer to the exhaust pipes of the passing cars, on the slope measurements, where inlets were mounted on the side of the mobile station, further away from the exhaust.

Vehicle passes of the measuring site with different speed regimes produced similar spans for all vehicles for both PN and BC EFs (Fig. 3c and d). Median PN EFs values were similar for 50 and 90 $km \, h^{-1}$ passes, 1.01 and $0.96 \times 10^9 \, \# \, kg^{-1}$ respectively and slightly lower for the acceleration regime where the median was $0.70 \times 10^9 \, \# \, kg^{-1}$. Median BC EFs were 0.37, 0.27 and $0.49 \, g \, kg^{-1}$ for driving regimes 50 $km \, h^{-1}$, 90 $km \, h^{-1}$ and accelerating respectively; the span of EFs was similar for all conditions.

These measurements show that a single stationary measurement isn't necessarily the representative EF of that particular vehicle, and that we can produce similar fleet PN and BC EFs distributions, regardless of the speed regime or slope on which the measurement station would be positioned. This suggests that a large number of

measurements should be taken to describe a representative fleet (like in Carslaw and Rhys-Tyler, 2013, for gaseous emissions), which should be possible using the stationary method.

In this respect, to assess the reliability and applicability of the stationary method, a comparison with the chasing method was performed and it is reported in Sect. 4.3. Moreover, the same chasing method was in turn validated through PEMS measurements (Sect. 4.2.3).

4.2 Chasing measurements

4.2.1 Chasing emission factors

Chasing results are summarized for each car in Table 2 where we report the average, median, minimum and maximum of the 10 s running integration technique, and the bulk EF calculated as an integral of the entire 5–6 min chasing run. In the same table we also report the influence of the determined background on the calculated EF, which we will elaborate on further in the next Sect. 4.2.2. We obtained almost the same values in bulk integration and median of the running integration for all vehicles (Table 2, EF columns for each car). This shows consistency between the two data processing methods, with the running integration providing the time evolution of the EF.

Because we omitted parts of data when values fell below the set background, the analysis turned out to be more difficult for the two EURO5 vehicles. In these two cases not only we had low signal to noise ratio for the CO_2 but also low signals of BC and PN. EURO5 (d) again had a single "super emission" peak of PN which lasted for almost entire first lap of the chase, thus the PN EF running integration maximum is high $27.1 \times 10^9 \# \, kg^{-1}$. EURO5 (g) emitted elevated particle concentrations throughout the chase but there was very little emitted BC. The calculated medians of both BC and PN EFs are low for the two technically advanced vehicles, about an order of magnitude smaller than median EFs of EURO3 vehicles.

4.2.2 Influence of background determination

To analyze the sensitivity of EFs to the determination of the background we calculated the EFs with three different background levels. First the background was set by calculating the linear trend between the 20 s average before and after the chase. Background variability was described with two standard deviations of the 20 s data before the chase, which covered at least 90 % of background variability. This value was then added to/subtracted from the background linear trend. In Table 2 the EFs calculated from background with added or subtracted two standard deviations are marked with EF+ and EF−, respectively. Median values of BC EF are again very consistent with the bulk integration values using the same background. The background changes affected both the median of the timer resolved measurements, and the bulk integration result, for about $-24/+26$ %. The changes had a bigger influence on the maximum EF values. PN EF did not match as well for the EF+ median but other than that the results were again similar for bulk and median of the running integration. Again we had tails of "super emission", here even more pronounced than for BC EF.

The advantage of the running integration method is that with it we can obtain an EF distribution. Both BC and PN EFs the distribution are best described with the median value, which has shown to be most characteristic for the vehicle; however the seldom high emission peaks should not be completely overlooked as their contribution to total emissions could be large. Our method may have difficulties in precisely determining the maximum value of the EF in the super emission tail, which can also depend on specific engine operation state that could or could not have been reproduced in the 5 lap chase.

4.2.3 Influence of dillution on chasing CO_2 measurements

The chasing method was further evaluated using tailpipe measurements of CO_2 by PEMS. In this test we wanted to see how mobile measurements match direct in-exhaust measurements of the chased vehicle emissions. From these measurements

we calculated the dilution rate (DR) as a ratio of the CO_2 measured by PEMS and by the chasing instrument (Chang et al., 2009) results are presented in Fig. 4. Our calculated DR values range from about 100, when we were in closer proximity of the chased vehicle and both vehicles speed was smaller; to the maximum value of about 72 000 when both emitted CO_2 and the exhaust mass flow rate drop. This occurred at the end of the track where we had to slow down to make a sharp U-turn. Excluding this period the maximum DR value was 8943 and the median 1077. This is similar to the measurements of Vogt et al. (2003), where they report dilution factors measured at approximately constant 14, 50 and 100 m distance from a diesel car going 50–100 km h^{-1} to be ranging from 926 to 9300.

The dilution does not affect the calculated EF BC, as we can see from Fig. 4. The BC EF is highest just before the highest cruising speed is reached; the dilution ratio is highest when the exhaust mass flow rate drops. This is consistent with finding of Chang et al. (2009) who report that the dilution ratio depends not only on speed but also on exhaust flow rate and other parameters, that are more important in near wake region. Omitted parts when CO_2 drops below the background overlap with parts when there is little to no CO_2 emitted from the exhaust pipe, thus the measured concentrations do not reach above the background level.

4.3 Comparing emission factors determined with different methods

4.3.1 Stationary and chasing emission factors

Another objective of this study was to compare the results of the chasing and the stationary methods. We plotted together the EFs of both methods for individual vehicles in Fig. 5, because the median values of the chasing method overlapped so well with the bulk integration, we omitted the later from the plot. On Fig. 5 are box plots for each car abbreviated as E3 (00), E3 (02), E3 (04), E5 (d) and E5 (g) for EURO3 (00), EURO3 (02), EURO3 (04), EURO5 (d) and EURO5 (g) respectively; letters C and S mark the chasing and stationary method respectively. Grey dots represent the actual data points:

there is a big difference in the measurement counts for both methods while chasing method consists of about 600 points, the stationary measurements feature only about 20 for EURO3 vehicles and a very scarce number of EFs for the two EURO5 vehicles.

We can see very good agreement between BC EF of EURO3 (00) and EURO3 (04), not only do the median values lie very close, but also the tails of both methods match (see also Tables 1 and 2). Here we can also see that the tails of the chasing method have only few points that are high enough to skew the distributions, this is even more pronounced with PN EFs. The EURO3 (02) stationary EFs are systematically lower than the chasing results. We believe this was because the EURO3 (02) driver was a professional driver who could produce the most repeatable stationary results, whereas for the chasing measurement we could capture the whole emission range of the vehicle. For "real world" measurements we wouldn't expect his driving to be representative, as most car drivers aren't professionals. Also a larger number of stationary measurements may show a distribution closer to the chasing one. PN EFs of the two methods match well only for the EURO3 (00), the other two vehicles PN EFs were much higher measured with the chasing method than those measured with the stationary method.

4.3.2 Particle number and black carbon emission factors

Comparing EF PN and EF BC that were calculated from the same vehicle drive-by (Fig. 6) we see that highest EF PN and highest EF BC do not coincide. This was also observed by Ban-Weiss et al. (2009) on their ∼ 300 vehicle fleet. In our study, EURO5 (d) produced highest stationary PN EF, EURO3 (04) highest BC EF, EURO3 (02) stationary EF's lie between these two, and EURO3 (00) features both high PN EF and BC EF, occurring at different vehicle passes. This is in-line with the theory that the soot particles inhibit formation of nucleation particles in the exhaust (Kittelson, 1998). The median of chasing EFs calculated with running integration method are also marked on Fig. 6 for comparison, they all match well with the stationary measurements.

To further explain the difference between PN and BC EF behavior, we segregated the PN EF to three size bins and calculated their individual EFs, using the running integration approach. Besides the total PN EF we now calculate EF PN for three aerodynamic particle diameter segments: less than 50 nm, 50 nm to 200 nm, and larger than 200 nm. We can see from Fig. 7 that PN EF from the 50–200 nm range coincides well with BC EF. PN EF from the 50–200 nm range is in this case very small compared to the EF of particles smaller than 50 nm. These dominated in parts of the EURO3 (02) plume e.g. one lap example of this vehicle in Fig. 7. We found similar super emissions of particles smaller than 50 nm in EURO5 (d) plume. We can see from Fig. 7 that from time 30 to 70 s total PN EF followed BC EF; these findings are consistent and complementary to our stationary observations on Fig. 6, where EF PN and EF BC did not overlap because the constituents of the exhaust changed with driving dynamics; similar results were reported in Sharma et al. (2005).

4.3.3 Black carbon emission factors measured in real driving conditions, emission standards and laboratory measurements

We wanted to know if our EURO3 vehicles are still compliant with their legislative standard and how our real world driving approximation matches the legislative one, therefore we recalculated the EURO3 emission standard from particulate mass (PM) in $(g\,km^{-1})$ to BC EF in $(g\,kg^{-1})$, assuming there is 47.6 % of elemental carbon (which we equate with BC in this particular campaign) in emitted diesel particulate mass (Landis et al., 2007, similar in Sharma et al., 2004); fuel density 0.832 $kg\,L^{-1}$ (mean of limit values of EN ISO 3675 and EN ISO 12185) and fuel consumption of 6 or 8 L/100 km. Car owners of the three EURO3 cars reported that their cars consume 7–9, 5–6 and around 8 L/100 km for EURO3 (00), EURO3 (02) and EURO3 (04) respectively. We did not use the lower limits as they applied for highway driving, which was not the case in our experiment. By the recalculation a EURO3 compliant car with 6 or 8 L/100 km fuel consumption could emit 0.48 or 0.36 g BC per kg fuel, we marked the limit lines on Fig. 5a. From these calculations the median EF BC determined by stationary measurements

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of the EURO3 (04) car is not expected to be compliant with legislation; neither is the median chasing BC EF. The median EURO3 (00) EF determined with the stationary and chasing technique is just below the limit value but has a long tail, where individual measurements lie well above the limit value. The median stationary EURO3 (02) BC EF is below the legislative limit, with only few measurements exceeding it, this however changes with the chasing technique where also the median BC EF of EURO3 (02) exceeds the legislative limit. The most probable cause of difference is the difference between the driving regime used here and the one in emission standard testing. In our case we had a track with slope and curves, while in the European emission standards the new European driving cycle is used where the changes in speed are gradual. Future emission standards should also address these "super emission" peaks, which may substantially contribute to total emissions.

BC EFs for a EURO5 gasoline and a EURO3 diesel car were recently measured in laboratory experiments by Platt et al. (2013) and Chirico et al. (2010). Platt et al. (2013) measured emissions of a single gasoline EURO 5 vehicle on a dynamometer using the new European driving cycle and report BC EF of $0.02\,\mathrm{g\,kg^{-1}}$. Chirico et al. (2010) measured BC EF of three in use cars with different after treatment devices. Most of their presented measurements are performed with the engine in idle mode, where BC EFs range from 0.466 to $0.763\,\mathrm{g\,kg^{-1}}$ fuel; one car was measured also at the speed of $60\,\mathrm{km\,h^{-1}}$ with reported BC EF of $0.512\,\mathrm{g\,kg^{-1}}$. These numbers are again consistent with the median EF values determined in our on-road measurements (see Tables 1 and 2).

5 Conclusions

Measuring vehicle EFs in real driving condition can bring further understanding of the actual fleet emissions. It can be a cost effective way to monitor fleet emissions by measuring a large number of vehicles and to check if measures taken to reduce emissions were effective. We further developed two such methods and tested them in a controlled

environment. We measured EF of two parameters: the particle number concentration (PN) and black carbon (BC), both being recognized as closer related to deleterious health effects of particulate air pollution than currently used legislative particle trait – their mass (PM) (eg. Janssen et al., 2011; Vinzents et al., 2005, and references there-in).

We have shown that the stationary and the chasing methods yield similar EFs and can thus be complementary. Both showed that a single stationary measurement of one vehicle, or the bulk EF of one chase, may not be representative of that vehicle EF span. Both methods showed how the emissions of a single vehicle changed with the driving conditions and further indicated that using a single number as a representation of that vehicle EF might not be appropriate, rather the distribution of that vehicle EF should be considered, where a single vehicle EF distribution has the most consistent number of measurements around the median value and is accompanied by a tail of "super emissions". These emission peaks may be similar to what Ajtay et al. (2005) found in their dynamometer study; and to NO_x emissions of different driving cycles reported in Franco et al. (2013) review of EF development. We found vehicle specific EF distribution for each of the investigated cars. In the stationary measurements EURO3 (00) and EURO3 (02) have similar average EF BC but their distributions are different – EURO3 (00) median is lower and tail longer than EURO (02). We found that measuring at different speeds or on different slope does not increase the EF (similarly to Ban-Weiss et al., 2009). Measuring during acceleration and in close proximity of the exhaust pipe improves the measurement collection efficiency. This knowledge on EF distributions could be beneficial, if included in models of traffic emissions.

With tailpipe measurements, we calculated the dilution ratio of CO_2 in the plume of the chased vehicle between the exhaust and the mobile measurement station. We found that the EF does not change with the dilution ratio, it is however sensitive to low exhaust CO_2 concentrations of the chased vehicle. If there is no CO_2 emitted from the chased vehicle, none can be measured and thus such parts should be omitted from the results.

The cost of both methods is low and each has its pros and cons – stationary method needs more measurements of a single vehicle to obtain the representative EF distribution or a measurement of a large number of vehicles to obtain a representative distribution for the fleet, but isn't affected by the background as much as the chasing method. With the later we can get the EF distribution in a single chase, but the data is more difficult to collect and analyze due to possible disturbances from other vehicles on the road, or sources close to the road, which may disturb the background. However, both are a valuable contribution to understanding fleet emissions as they can be used to measure the versatility of the in-use fleet. Using distributions of measured real world EFs may improve modeling of traffic emissions. This may be especially true for BC, since it is not as affected by environmental conditions as PN or particle mass. In disperse and variable air pollution source like traffic, it is advantageous to have different approaches and insights to the problem. By comparing results of different measurements like dynamometer and real world measurements, we can understand traffic emissions better. The methodology described here-in will be used to compile the EF inventory of the European in-use car fleet, measured in real driving conditions in winter on Slovenian roads.

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Table 1. Summarized emission factors (EF) of individual vehicles for **(a)** black carbon (BC) and **(b)** particle number (PN).

(a)	BC EF [g kg^{-1}]	EURO3 (00)	EURO3 (02)	EURO3 (04)	EURO5 (d)	EURO5 (g)
	AVERAGE	0.35	0.40	0.72	/	0.03
	MEDIAN	0.25	0.41	0.70	0.03	0.02
	MIN	0.06	0.16	0.22	/	0.01
	MAX	1.30	0.73	1.75	/	0.06
	COUNT	20	23	20	1	3
(b)	PN EF [10^9 # kg^{-1}]	EURO3 (00)	EURO3 (02)	EURO3 (04)	EURO5 (d)	EURO3 (g)
	AVERAGE	1.59	0.93	1.10	4.08	0.12
	MEDIAN	1.27	0.74	1.07	2.44	0.11
	MIN	0.32	0.34	0.49	2.22	0.04
	MAX	5.05	2.74	1.66	7.57	0.22
	COUNT	24	27	22	3	12

Table 2. Summarized results of the chasing measurements. BC **(a)** and PN EF **(b)** are reported as the average, median, minimum and maximum of the running integration method and the result of bulk integration. In column headers EF, EF −, and EF + denote the subtracted background used for the calculation of the EFs. For background influence analysis we first subtracted the linear fit of average background values measured before and after the chase (EF), then we tested it by subtracting (EF −) or adding (EF +) two standard deviations to the background value.

(a)	BC EF $[\mathrm{g\,kg^{-1}}]$	EURO3 (00)			EURO3 (02)			EURO3 (04)			EURO5 (d)			EURO5 (g)		
		EF −	EF	EF +	EF −	EF	EF +	EF −	EF	EF +	EF −	EF	EF +	EF −	EF	EF +
RUNNING I.	AVERAGE	0.26	0.36	0.67	0.47	0.72	1.11	0.51	0.69	1.08	0.06	0.07	0.10	0.06	0.10	0.08
	MEDIAN	0.24	0.30	0.40	0.42	0.60	0.82	0.51	0.67	0.87	0.05	0.05	0.04	0.04	0.04	0.03
	MIN	0.07	0.09	0.09	0.14	0.14	0.11	0.01	0.001	0.14	0.01	0.0007	0.00002	0.003	0.0005	0.0003
	MAX	0.65	3.07	15.11	2.50	4.18	17.97	1.36	1.89	10.66	0.42	0.51	5.36	0.34	1.84	1.24
BULK INT.		0.24	0.30	0.40	0.45	0.61	0.87	0.51	0.63	0.79	0.04	0.03	0.02	0.04	0.03	0.03
(b)	PN EF $[10^9\,\#\,\mathrm{kg^{-1}}]$	EURO3 (00)			EURO3 (02)			EURO3 (04)			EURO5 (d)			EURO5 (g)		
		EF −	EF	EF +	EF −	EF	EF +	EF −	EF	EF +	EF −	EF	EF +	EF −	EF	EF +
RUNNING I.	AVERAGE	0.99	1.44	2.71	1.87	3.26	3.3	1.41	2.00	3.13	0.80	1.02	1.68	0.15	0.26	0.36
	MEDIAN	0.86	1.05	2.71	1.17	1.52	3.3	1.27	1.64	3.13	0.10	0.12	1.68	0.12	0.15	0.36
	MIN	0.08	0.21	0.24	0.05	0.48	0.65	0.001	0.0001	0.25	0.004	0.002	0.003	0.01	0.02	0.02
	MAX	4.34	17.63	61.90	28.83	91.3	45.10	4.40	8.76	42.19	20.80	27.10	38.90	0.99	3.20	7.55
BULK INT.		0.85	1.10	1.49	1.29	1.78	2.61	1.27	1.58	2.03	0.68	0.82	1.03	0.12	0.15	0.19
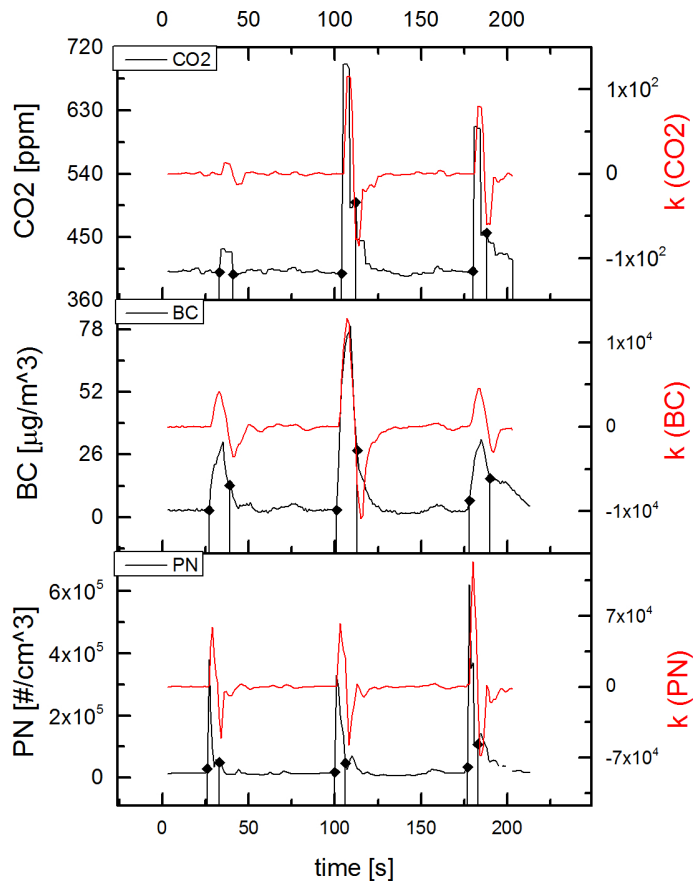
Figure 1. Examples of three consecutive stationary plume measurements. Marked with black dots and drop lines are the beginning and the end of each plume. The calculated slope (k) of each pollutant which was used to determine the end of the plume is plotted in red.
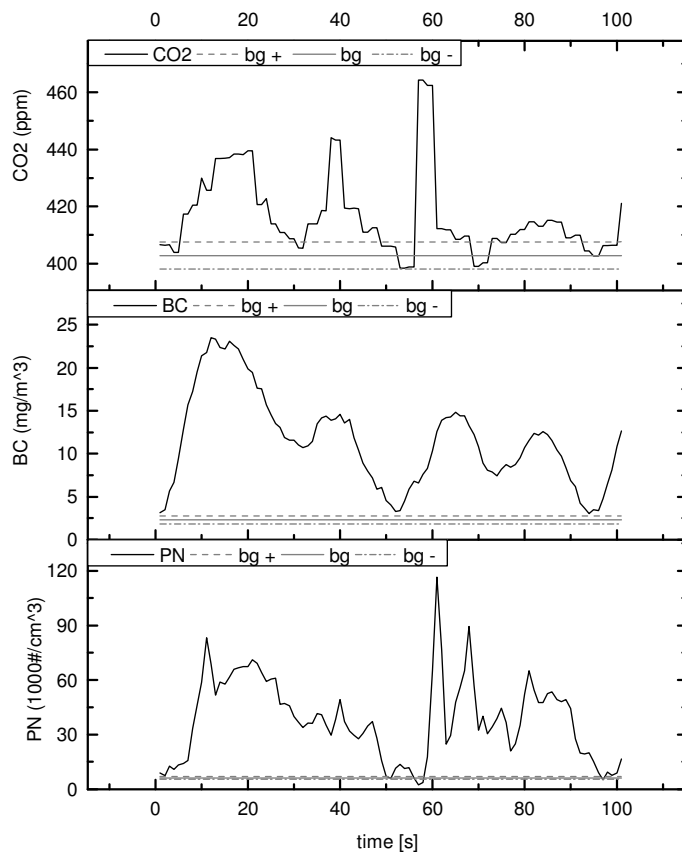
Figure 2. An example of chasing plume measurements (black solid line). Background (bg) and background + and − two standard deviations of background is shown in gray solid, dashed and dash-dotted line, respectively.
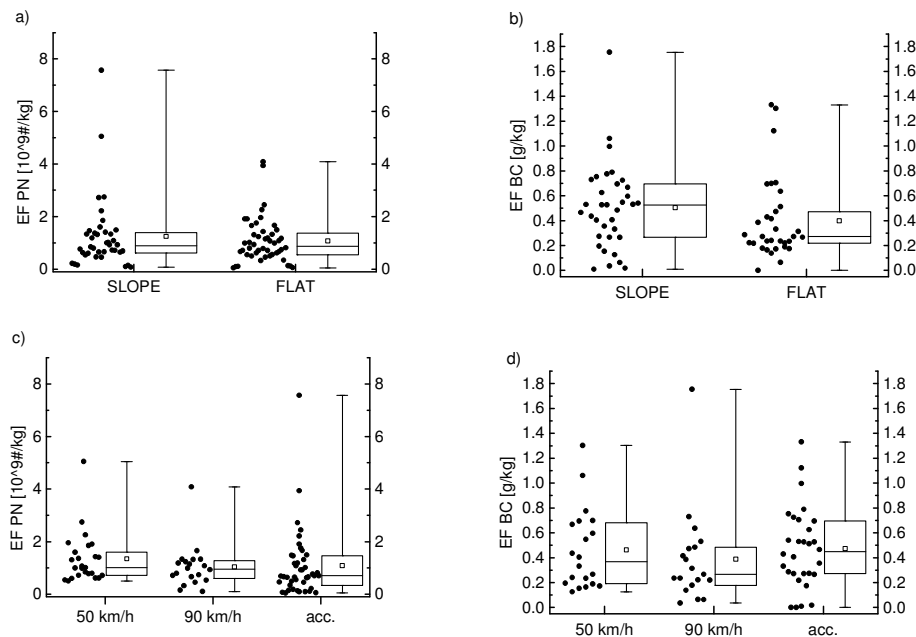
Figure 3. Stationary measurements of BC and PN emission factors for measurements performed on slope and flat road (**a** and **b**); and at different speeds of passing cars (**c** and **d**). Values marked are the minimum and maximum (–), 25th and 75th percentile (lower and upper box lines), median (middle box line), and mean (□). Black dots represent the actual data points.
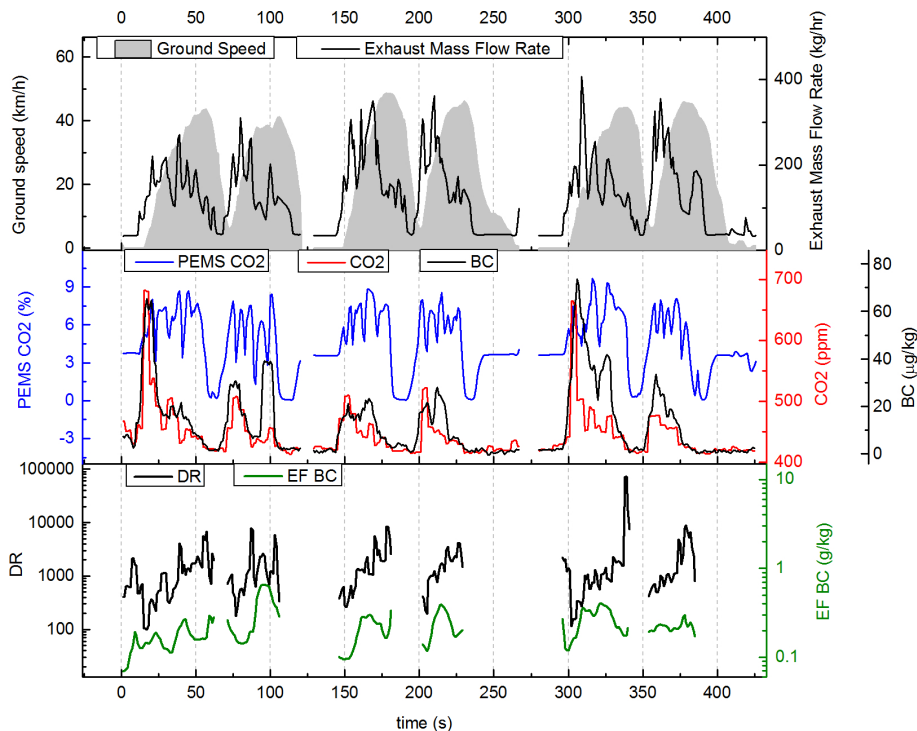
Figure 4. Tailpipe measurements performed with portable emission measurement system (PEMS) are ground speed (shaded grey) and exhaust mass flow rate (black) – top; and CO_2 (blue) – middle. Also in the middle plot are CO_2 and BC measured with mobile station in red and black, respectively. Calculated dilution ratio (DR) in black and BC EF in green – bottom. BC EF does not show any significant dependence on DR. Note the log scales for DR and EF.
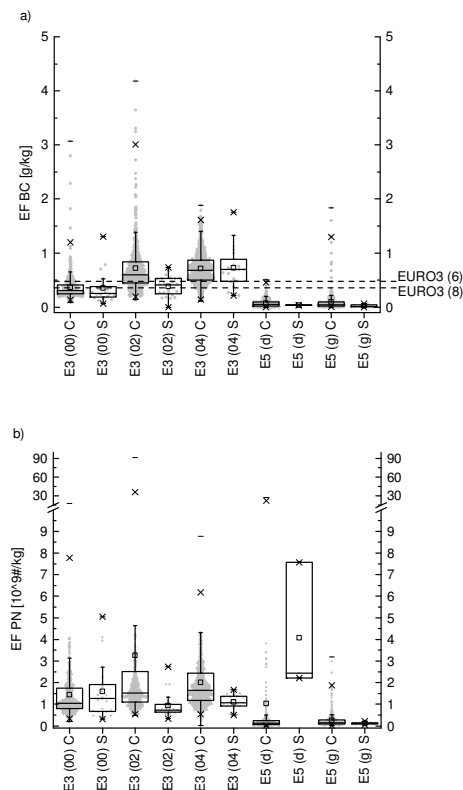
Figure 5. Black carbon **(a)** and particle number emission factor distributions **(b)** for each car measured with chasing (C) and stationary (S) method. Marked values represented are the minimum and maximum (–), 1st and 99th percentile (x), 5th and 95th percentile (whiskers), 25th and 75th percentile (bottom and upper box lines), median (middle box line), and mean (□). Grey dots represent the actual data points. The EURO 3 emission standards recalculated from particle mass to BC, assuming consumptions of 6 and 8 L/100 km is shown in **(a)**.
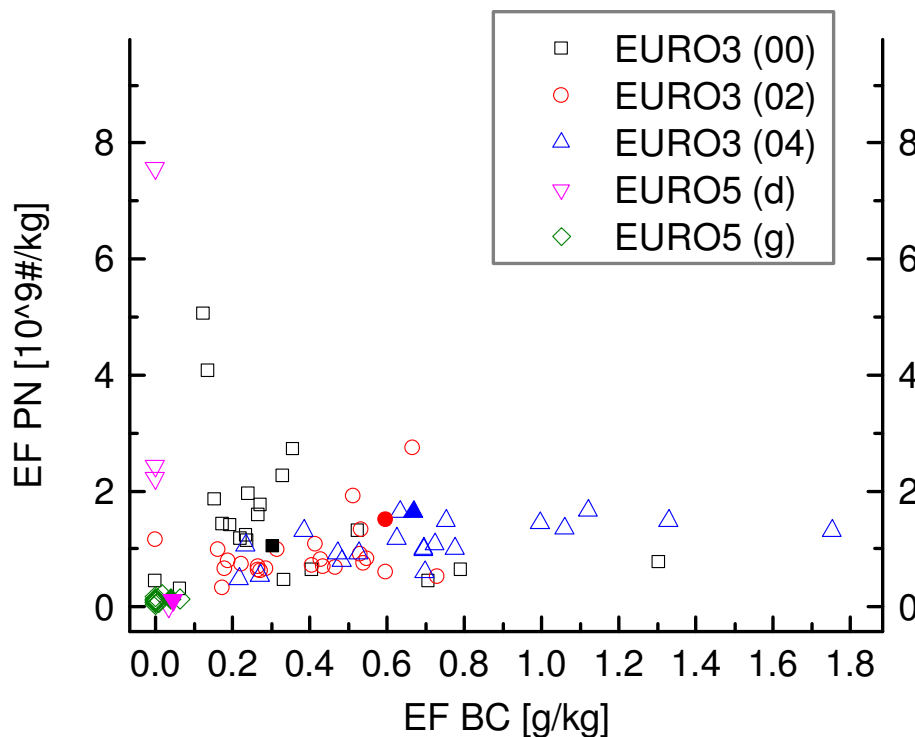
Figure 6. Particle number emission factors (EF PN) vs. black carbon emission factors (EF BC) measured at the same vehicle drive-by. Empty symbols represent measurements with the stationary method; full shapes denote the median of EFs measured with the chasing method. EURO5 (g) and EURO5 (d) overlap thus the latter is not visible.

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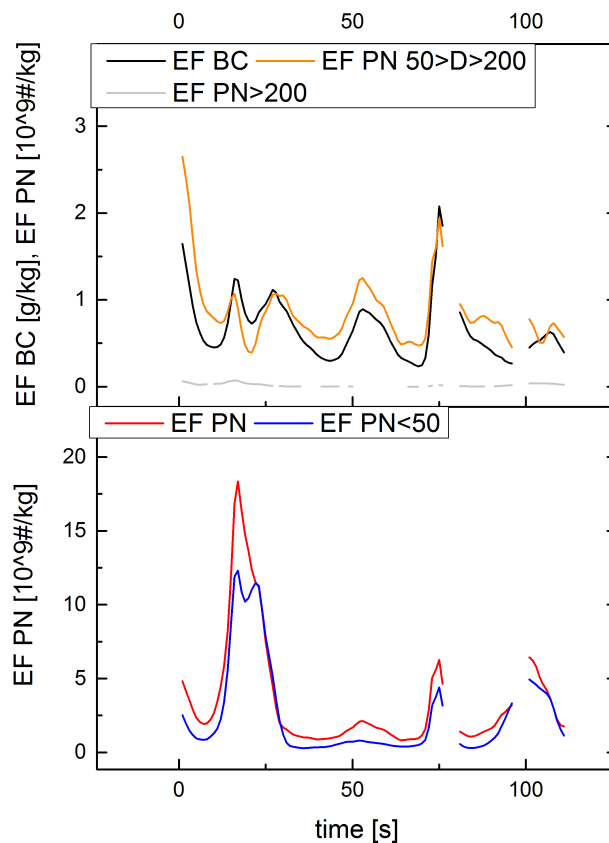Printer-friendly Version

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Figure 7. A sample of chasing emission factors for black carbon (BC) and particle number (PN) for different size fractions for EURO3 (02): BC (black), total PN (red), PN of particles smaller than 50 nm (blue), PN of particles from 50 nm to 200 nm (orange) and PN of particles larger than 200 nm (grey).