The traffic emission-dispersion model for a Central-European city agrees with measured black carbon apportioned to traffic

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\textbf{A B S T R A C T}

The bottom-up traffic emission model EMISENS is used to calculate hourly black carbon (BC) and nitrogen oxides (NO\textsubscript{x}) emission rates on an average workday in Maribor (Slovenia), using emission factors that were previously determined with the on-road chasing measurements in Slovenia. From modeled NO\textsubscript{x} emission rates and in-situ NO\textsubscript{x} measurements we empirically determined the hourly dispersion of traffic emissions and applied it to model BC concentrations using BC emission rates. We compared the modeled BC concentrations with in-situ BC concentration measurements over three periods in winters 2011–2012 and 2012–2013, a total of 67 workdays. Measured BC concentrations were first apportioned to traffic using the bottom-up Aethalometer model. We found that the bottom-up modeled BC concentrations overestimated the top-down apportioned values by only 19%, 32% and 6% in each of the three investigated time periods, respectively. We did not find any influence of meteorology on the performance of the model. This is the first time that BC source apportionment results were used to evaluate traffic emissions calculated using the bottom-up modeling approach. We demonstrate that the two independent approaches yield similar results. We use thus validated emission inventory for evaluating different emission reduction scenarios. We show that excluding 10% of vehicles that are highest BC or NO\textsubscript{x} emitters would reduce the total BC or NO\textsubscript{x} emissions from traffic in Maribor by 39% and 33% respectively.

1. Introduction

Traffic emission models have been used to backup discussions related to improvements in air quality and to define mitigation policies in cities. They are especially useful when designing air quality plans and quantifying the effects of hypothetical interventions on the fleet collective emission rates. Numerous models were developed. An overall review of the methodologies that are available and used in Europe to compile local and regional air quality plans is proposed by Thunis et al. (2016). Choosing the right model depends on the required spatial and temporal detail of the investigated subject, and on the available resources in terms of time, finance and available data. While some models may oversimplify the traffic situation, more complex models require more detailed input data, which may be more susceptible to errors in estimations, measurement or to assumptions. As Grote et al., 2016, remind, the quality of the model’s results depends on the quality of the input data.

Two kinds of approaches to model traffic emission rates can be distinguished: the bottom-up approach relies mostly on local activity estimates collected over the area of interest (e.g. traffic counts for road segments in a city); the top-down approach distributes emission totals (country or region, e.g. derived from total fuel sales) spatially according to gridded proxies (e.g. population, land use ...). Thunis et al., 2016, mention the importance of favoring bottom-up approaches detailed at the spatial and temporal resolutions adequate for the modeling purposes.

The EMISENS traffic model (Ho et al., 2014) was developed to insure the coherence between bottom-up and top-down approaches. It is an inexpensive alternative to road traffic emission models as it computes traffic emission inventory with less input information by introducing vehicle and street categories; and it calculates the uncertainties in the input data to help facilitate their improvement. The model formulation is based on COPERT IV methodology (Ntziachristos et al., 2007), and originally uses COPERT IV emission factors (EF).

Nevertheless the model could also benefit of real world EF measurements. Those data can be issued from the large measured sample of vehicles and the variability introduced with the use of the real world EF, including the influence of driver behavior or highly transient...
operations. One can note that this might be at the expense of precision since various real-world EF measurements methods were characterized as being less precise than the dynamometer studies (Franco et al., 2013). Nonetheless, the use of the most recent and relevant EF reported for the investigated area allows to better model hourly traffic emission rates.

To validate traffic emission models’ results different techniques may be used. Several approaches in linking emission inventories and ambient measurements have been described and evaluated by (Monks et al., 2015). In fact measurements of real-world EF were initially developed to evaluate and identify the potency improvement of the road traffic emission models (Franco et al., 2013). Other approaches to evaluate traffic emissions are widely based on direct comparison of the measured ambient pollutant concentrations to the results of traffic emission-dispersion modelling (Smit et al., 2010, and references therein). The advantage of using ambient measurements is that they capture a range of driving conditions and a large vehicle sample. The disadvantage is the combined use of emission and dispersion models that increases uncertainties, and the influence of non-traffic sources. Uncertainties are even greater when the environmental simplifications such as perfect mixing or steady-state wind conditions are made (Smit et al., 2010).

Traffic emission models vary in complexity. Smit et al. (2008) proposed the following categories: Average-speed models (e.g. COPERT, MOBILE, EMFAC), Traffic-situation models (e.g. HBEFA, ARTEMIS), Traffic-variable models (e.g. TEE, Matzoros model), Cycle-variable models (e.g. MEASURE, VERSIT +), and Modal models (e.g. PHEM, CMEM). The proposed categories are increasing in complexity as the amount of input variables and their complexity is increasing, i.e. in the average speed models like COPERT and EMISENS variables like average vehicle speed, EF and number of vehicles are simple and easier to obtain than variables for i.e. stop and go traffic needed for traffic situation models, or queue length and signal settings needed for traffic variable models, etc.

A more complex model doesn’t necessarily mean that it would perform better than a less complex model. Smit et al., 2010, found that less complex models had lower prediction errors for particulate matter (PM). Model prediction errors for PM along with carbon monoxide varied within a factor of 3 (also up to 5), whereas their prediction error for CO₂ emissions was 1.3, and around 2 for hydrocarbons (HC) and nitrogen oxides (NOₓ). They attributed the large prediction error for PM to the discrepancies between different definitions of PM between models and observations (e.g. size, measurement principle, exhaust/non-exhaust contribution).

Although particulate mass concentration (especially PM10) is a regulated aerosol parameter (European parliament, 2008), it has been shown that BC is more relevant metric of traffic pollution than PM10 when demonstrating the effectiveness of air quality mitigation measures (Invernizzi et al., 2011; Titos et al., 2015). Because BC absorbs visible light, is resistant to chemical transformation (Goldberg, 1985; Ogren and Charlson, 1983), and has been shown to have a higher relation to deleterious health effects (Janssen et al., 2012), it became one of the key targets for current research on the aerosol impact on climate and health and the associated mitigation strategies (Petzold et al., 2013, and references there in).

Unlike PM, which is a mix of primary and secondary aerosols, BC is primary only and can be directly linked to its sources. Its two main sources in many European urban areas are traffic and domestic heating using biomass burning in small inefficient stoves. Their contributions to ambient air pollution can be quantitatively discriminated based on aerosol light absorption properties with the so called “Aethalometer model” (Sandradewi et al., 2008a). This source apportionment model uses the notion that the light absorption coefficient of aerosols is inversely proportional with the wavelength at which it is measured and that this dependence can be described with the Angstrom exponent, which is source specific. It has been demonstrated that, for aerosol particles produced in fossil fuel combustion, the Angstrom exponent is 1.0 ± 0.1 (Bond and Bergstrom, 2006); and for wood burning fraction the Angstrom exponent is 2.0–0.5/ +1.0 (Day et al., 2006; Kirchstetter, 2004; and references therein).

In this research we use the EMISENS traffic inventory model with BC and NOₓ EF collected by Ježek et al. (2015a), in real driving conditions to simulate BC and NOₓ traffic emission rate. NOₓ emission rates modeled by EMISENS coupled with NOₓ in-situ measurements are used to evaluate the dispersion of the traffic pollutants in the atmosphere. Then using the dispersion factor the concentrations of BC are estimated using the modeled BC emissions. Finally we evaluate the resulting modeled BC concentrations with other traffic BC evaluations based on in-situ BC measurements. Indeed in-situ BC measurements were appropriated to traffic using the Aethalometer model. The objective of the paper is to use this first comparison to proceed to a cross validation of the apportionment methodology, the EF measurement data, and the traffic emission model. Since there are many uncertainties on BC emissions around the world, we suggest applications of combined methodologies for planning the abatement strategies related to traffic emissions in cities.

The paper is structured as follows: In Section 2 we describe the city of Maribor and the positions of traffic count measurements sites, the positions of the ambient air pollution monitoring stations and the Aethalometer model. In Section 3 we in detail describe the input data used in the EMISENS traffic model and propose four emission reduction scenarios. In Section 4 we describe how we determine the dispersion of BC emissions to calculate BC concentrations in the city center. In Section 5 we present the results of this paper, which are spatial and temporal traffic emission rates on an average workday in the city of Maribor (Section 5.1), validation of the modeled BC concentrations with the ambient measurements of BC apportioned to traffic with the Aethalometer model (Section 5.2), the results of the emission reduction scenarios in the city center (Section 5.3), followed by a discussion on the results (Section 5.4) and conclusions of the paper (Section 6).

2. Study area and in-situ measurements

The city of Maribor is the second largest settlement in Slovenia. It is a small city positioned in a basin that is opened to South-East, transacted by the Drava River (Fig. 1). The city center lies to the north of the Drava River; to the south of the river we find the industrialized areas, a shopping center and residential districts. The Maribor municipality had approximately 112000 inhabitants and was covering an area of 147.5 km² in years 2008–2015. In the same time period the Maribor settlement had approximately 95000 inhabitants (year by year statistic from Republic of Slovenia Statistical office web site can be found in Supplement Table 1) covering an area of 41 km².

Our modeling domain was a 5 × 5 km grid over Maribor, with a spatial resolution of 0.5 × 0.5 km² (Fig. 2). The size of our domain is small because Maribor is a small city, moreover we wanted to model with high spatial resolution because BC concentrations are highly spatially heterogeneous (Enroth et al., 2016; Invernizzi et al., 2011; Ogrin et al., 2016; Pirjola et al., 2012; Titos et al., 2015) and we wanted to avoid averaging traffic emissions over a too large area. The 0.25 km² grid box was in our opinion good area to model traffic emissions in Maribor because it included the entire street canyon in which the city center air pollution measurement station was positioned and the few closest streets (Fig. 2), which may influence the pollutant concentration.
levels there. Ogrin et al., 2016, found that ~200 m away from a highway, BC and NOx concentrations drop to urban background levels; similar observations were made in Finland by Enroth et al. (2016). The resulted methodology could be applicable to other cities with similar size. This means about half of all European cities, according to a study made in 2011 by European commissions’ Directorate-General for Regional and Urban Policy and Organization for Economic Co-operation and Development (OECD), which found about half of all cities in Europe are relatively small with a center between 50000 and 100000 inhabitants (Dijkstra and Poelman, 2012). We found similar in Eurostat’s data. We used the last reported population for a city from years 2008–2015; where there was data for the city and greater city area, we used the latter; and we excluded all cities with population less than 50000. With these constraints we obtained the data for 907 European cities. 68% of European cities had fewer than 200000 inhabitants. These cities contain 30% of the European population living in cities.

2.1. Traffic count sites and traffic situation

Six traffic count sites were set up in and around the Maribor city center as part of Slovenian Infrastructure Agency national monitoring. The data for each traffic count site (TCS) included vehicle counts with 15 min time resolutions for eight vehicle categories: cars, light trucks (LT), medium trucks, heavy trucks, and heavy trucks with trailers, semi-trailer trucks, busses and motorcycles. In our study our final two categories were light vehicles (LV) and heavy vehicles (HV). The LV category included cars and LT; these are all vehicles lighter than 3.5 t. The HV included medium trucks, heavy trucks, and heavy trucks with trailers, semi-trailer trucks and busses, which were all vehicles heavier than 3.5 t.

Fig. 1. The top left image shows a relief map of the city area; the top right is the exact same area in its orthophoto version. The small white rectangle in the bottom right corner of both top maps denotes a distance of 1.5 km. The red rectangles mark the area shown enlarged in the bottom map, where the city center above the Drava river is shown and the positions of the urban background and the urban air pollution measurement stations are marked with an orange “x” and a blue “x”, respectively. The surroundings of the urban background ambient air monitoring station (orange frame) looking southwest, towards the city; the closest road would be to 135 m to the left of the observer; and the urban ambient air monitoring station (blue frame), facing south along the Titova cesta, which was at the time unusually empty. The maps are used with the permission of Geopedia.si. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)
The positions of the six TCSs are marked in Fig. 2. The actual location of site 802 is positioned on the A1 highway (European route E57), before the highway splits into three branches (observing the route from south to north). One of the branches is the H2 expressway, which passes through the city via TCS 15 and 16; the second is the continuation of the A1 highway, which bypasses the city via TCS 889; and the third is the A4 highway, which leads south toward the border with Croatia (not depicted in Fig. 2). The expressway H2 and highway A1 branches of the motorway merge further north (off the map in Fig. 2) and lead to Austria and Hungary.

Maribor is connected to Ljubljana (capital of Slovenia) and Graz (second largest city in Austria) via the A1 highway. The part of this highway that is bypassing Maribor was built in 2009 (the route in Fig. 2 with TCS 889), channeling most of the transient traffic away from the expressway (the route in Fig. 2 with TCSs 15 and 16). The change of traffic is evident in Supplement Figure 1, where we show how the annual average daily traffic (AADT) for LV and HV changed in the period 2006 to 2012 (Statistical Office Republic Slovenia). The LV traffic on the TCS 15 and 16 was reduced by approximately 10000 LV and 5000 HV during 2009 whereas the new highway segment started off with approximately 20000 LV and 5500 HV.

The new traffic situation affected mostly the north-south transient traffic and did not affect the traffic in the city center. In a cordon - license plate tracking study from 2005, the results showed that the transient traffic in the direction east-west was weak (only 500 per day), with most of the transit traffic through Maribor in the direction north-south (Lep and Mesarec, 2013). In a later 2010 cordon study performed in the city center, they found that for 83% travels in the city center either the destination or the origin of the travels was there-in; 17% of traffic was transient, most of which was on Titova cesta (Lep and Mesarec, 2013), the road next to which the city center air pollution monitoring station was positioned (Fig. 2 and 1). The fact that the traffic situation in the city center was not greatly affected is evident also in Supplement Figure 1, TCS 18, where both LV and HV AADT is stagnant through the years 2006–2012.

Another important finding in Lep and Mesarec (2013) relevant to
this study, was that the traveling time in the city during rush hours does not increase significantly, meaning that congestion during rush-hours is not severe and would thus not impact traffic emissions significantly in those time periods (Grote et al., 2016).

Traffic count data was used first to investigate fleet composition (section 3.1.1); second, to verify the AADT on the road map (section 3.1.2.1 and Supplement material 2); third, to calculate the diurnal traffic flux profiles for LV and HV (section 3.1.3), and finally to calculate the weights for LT and cars in LV category (section 3.1.4). The data was provided by the Slovenian Road Agency at the Ministry of Infrastructure and Spatial Planning, for three winter periods 2011, 2012 and 2013. We used only data for the measurement time periods, listed in Table 1.

Table 2

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The BC and NOx ambient measurements were performed at two air pollution measurement stations in and around Maribor. The urban measurement site lies in the city center (15°39′5″ 46°34′33″) next to a busy road (Titova cesta). The urban background site is positioned just outside the city (Vrbanski plateau, 15°37′38″ 46°34′7″). The stations with their surroundings are shown in Fig. 1; their positions are marked in Figure 1 and Figure 2. The closest road to the urban background station is approximately 135 m away and it is not very busy with traffic (~3000 vehicles per day, Drev et al., 2009). The distance between the two stations is approximately 2.2 km as the crow flies (Fig. 1). The urban station is operated by the Slovenian Environment Agency and the urban background station by the National Laboratory of Health, Environment and Food. The two organizations provided quality controlled NOx measurement data for their respective stations.

The source apportionment was made using the Aethalometer model (Sandradewi et al., 2008a) with most frequently used Angstrom exponent pair of 1.0 for traffic and 2.0 for wood burning. The source apportionment is possible because the Aethalometer, model AE31, measures light absorption at different wavelengths. These measurements enable us to discriminate between sources’ contribution to BC using the source specific Angstrom exponent, which describes how the aerosol particle absorption coefficient varies with the wavelength (Sandradewi et al., 2008b). Completely black particles which may be found in fresh diesel exhaust feature an Angstrom exponent close to 1. Smoke produced from burning wood contains aerosolized substances that absorb strongly in the blue and ultraviolet part of the light spectrum, so we expect an Angstrom exponent around 2 (Sandradewi et al., 2008b) or even more if the combustion is very inefficient. For a more detailed description of the Aethalometer operational principle see (Drinovec et al., 2014; Hansen et al., 1984), and for a more detailed description on the Aethalometer source apportionment see (Favez et al., 2010; J. Sandradewi et al., 2008a; Sandradewi et al., 2008b).

We used the data from three winter periods, when we obtained valid NOx and BC measurements available from both the urban and the background stations: (i) 2nd – 16th November 2011 (11 workdays); (ii) 1st February– 14th March 2012 (27 workdays); and (iii) 26th November 2012–31st January 2013 (29 workdays). Summary of the investigated time periods is in Table 1.

3. Modelling the traffic emission rates

We used the EMISENS traffic model (Ho et al., 2014) to calculate hourly BC and NOx emissions produced by traffic on an average workday in Maribor. In the following subsections we will examine input data for the EMISENS model and the analysis of traffic in Maribor to check the influence of the input data on the model results. A more detailed description of the model may be found in Ho et al. (2014).

3.1. Input data for the EMISENS model

The input data for the EMISENS model were:

a) Traffic count data on 6 TCSs in Maribor in winters 2011, 2012 and 2013, described in section 2.1
b) A road map taken from the noise study made for Maribor municipality in 2009 (Drev et al., 2009), which provides information on the street lengths, speed limits and AADT for LV and HV

c) On-road EF in terms of g of pollutant per km driven, were calculated from EF in Ježek et al. (2015a), which were reported in terms of g of pollutants per kg fuel consumed, and the fuel consumption data from EMEP/EEA guide book (Ntziachristos et al., 2014).
In the following subsections we describe the use of these datasets in the model.

3.1. The fleet composition

Table 2 shows the average number of traffic per workday at each location and the percentage of vehicles in LV and HV for the three investigated time periods, a more detailed table with the percentage of vehicles in each vehicle category separately is in Supplement Table 3. Cars represent the majority of the traffic at all sites. The portion of cars is similarly high (86–93%) on all non-highway counter sites (TCS 15, 16, 18, and 61, hence forth referred to as city traffic count sites or CTCS). On the two highway counters 802 and 889, it is about 20–34% lower (70% and 56%, respectively). The traffic on highways differs from other locations, with a 3–4 fold higher share of all goods vehicles and in terms of the proportion of semi-trailer trucks, which is ten times higher than on other counter locations. This reflects the transient interstate traffic of goods on the A1 highway. The CTCS should be more representative for the city fleet composition as they are positioned closer to the city center, and they differ in terms of percentage of respective categories by a maximum of 7% (cars) and a minimum of 0.8% (medium sized trucks) from each other. Owing to the higher share of goods vehicles on the highway counters, we can see that the fleet composition on the highway differs from the fleet composition in the city. Our main goal was to see how our model would perform in the city center, which is where the ambient station is positioned. Because the A1 highway is not in our modeling domain and much too distant to influence the concentrations in the city (Ogrin et al., 2016), we use only the traffic count data from the CTCS to set the weights for EF of cars and LT the final LV category.

From Table 2 we can see that out of all goods vehicles, the most frequent were light trucks with a 4–7% share, with the others represented with 0.1–2%. The number of motorcycles on all traffic count sites was very small, with up to 0.1% all sites and time periods. We excluded them from further analysis because we did not have EF measurements for motorcycles and excluding them would not yield a systematic bias due to their very small number and low EF (Zardini et al., 2014), but we treat all streets as one category. We do not expect that this would influence the results around the urban ambient air pollution monitoring station, nor 87% of domain where the expressway is not present.

3.1.2. The road map

The road map from the noise study (Drev et al., 2009) provided the information on the street lengths, speed limits and AADT for LV and HV. This data was provided by the Department of Geographic Information System and Data Processing at the Office of Utility, Transport and Spatial Planning of the Municipality of Maribor.

3.1.2.1. Modeling domain. We used the road map of the city from the 2009 noise study (Drev et al., 2009) which included information on street lengths, AADT (Supplement Figure 2) for LV and HV on each street segment, and speed limits (Supplement Figure 3). All roads with traffic flow higher than 1000 vehicles per day and 184 road segments with traffic flow lower than that were included in the noise study road map. The difference between the road map from the 2009 noise study (Drev et al., 2009) and all roads in the area is evident in Fig. 2, where we added an underlying road map from the year 2012 provided by Slovenian ministry of environment and spatial planning. The roads missing in the city center are local city roads that were either closed for traffic or were used for access to closed spatial units (such as residential neighborhoods, individual constructions, industrial estates, shopping and recreation centers, etc.). Also missing in the 2009 map was the segment of A1 highway that was opened for traffic in August 2009 (road with traffic counter 889 in Fig. 2), that overtook most of the transient traffic from the city center. We discuss the effects the new highway section had on the city traffic situation in more detail in Supplement material 2.

3.1.2.2. Speed limit. The speed limit (for LV) on the highway, where the TCSs 802 and 889 are positioned is 130 km/h; on the expressway, where TCSs 15 and 16 are positioned, the speed limit was 100 km/h; on TCSs 18 and 61, which are positioned on main city roads, the speed limit varies from 70 to 90 km/h; on Titova cesta, next to the air pollution monitoring station, the speed limit is 50 km/h. The speed limit over the domain is quite homogeneous, on ~70% of street segments it is 50 km/h, ~12% of roads have speed limit 70 km/h and ~8% more than that (summary in Supplement Table 2). Supplement Figure 3 indicates where those roads are. We can observe that in the grid cell with the city center ambient air measurement station and the grid cells around it, all streets have speed limit of 50 km/h or less. Because the speed limit over the entire domain and especially in the grid cell with the ambient monitoring station is homogeneous, and because Ježek et al., 2015b showed that the median EF value was the most representative for individual vehicle emissions, regardless of the speed regime, we did not classify streets according to the speed limit, as was done previously using the EMISEN model (Ho et al., 2014; Rahal et al., 2014), but we treat all streets as one category.

3.1.3. The diurnal traffic flux profile and uncertainties on street mileage

We calculated the average workday hourly traffic profile for both LV and HV vehicle categories on CTCSs. An example from the (ii) investigated time period is in Fig. 3. These demonstrated that there is different traffic burden of LV and HV on the four sites, but a similar diurnal variation of the number of vehicles. We described this daily variation on each site relative to the respective site’s average daily traffic (the average number of vehicles per day traveling on the street segment, calculated for the investigated time period). Averaging thus obtained profiles gives a common diurnal traffic flux curve for LV and HV, represented in Fig. 3, graphs c and d. The traffic flux and the standard deviations were calculated from the combined data of all three investigated time periods. If we calculated the traffic flux for individual time period, we got the same results as for all three time periods together.

The standard deviation of LV and HV within an hour changed during the day and was smaller for LV than for HV. For LV it was highest in the hour just before the morning peak, for HV it was high during the day. This indicates that there are small differences between the street segments’ LV flux and slightly larger differences for HV traffic. However, since LV are dominant and Maribor is a small city, we think it is safe to assume the traffic flux would be similar on all street segments.

The uncertainty of the LV and HV street mileage factor was estimated by a study of the total daily traffic data distribution around a mean profile. We excluded all weekends and holidays, and two outliers from HV data. The distributions are presented in Fig. 4, where we can see that the distribution total daily traffic for LV was slightly more symmetrical than that for HV, and both were skewed slightly to the left. The uncertainty on the LV and HV street mileage factor is evaluated using the standard deviations, that are 0.08 and 0.14 for LV and HV respectively.
3.1.4. Weighted average emission factor for vehicle categories

For both LV and HV categories used in the emissions model, the EF were calculated with a weighted average EF of their subcategories of vehicles. The EF of all the vehicles in their respective subcategories were taken from the on-road chasing study of Ježek et al. (2015a). The individual vehicle’s EF from Ježek et al. (2015a), were converted from g/kg units to g/km units, using EMEP/EEA fuel consumption values (Ntziachristos et al., 2014), which were 70 g/km, 60 g/km, 80 g/km and 240 g/km for gasoline cars, diesel cars, light trucks, and HV respectively (Table 3). Because we wanted the EF categories to represent all the vehicles in the category as well as possible with a single number, we integrated the EF distribution of individual subcategories to obtain a single number. We split the vehicles in each subcategory in bins in way that each bin included as close as 10% of vehicles according to their EF.

Fig. 3. An example of average workday diurnal traffic counts for light vehicles- LV, (a) and heavy vehicles – HV, (b), on four city traffic count sites (CTCS) from the (ii) time period. Hourly diurnal traffic flux calculated relatively to the average daily traffic on traffic count sites for LV (c) and HV (d) in three different time periods (i, ii, iii); the error bars are standard deviations calculated for each hour for all workdays from all CTCS.

Fig. 4. Distributions of relative workday traffic on all city traffic count sites for light (LV) and heavy (HV) vehicles.
EMEP/EEA. In the study of Pranjc (2013), they calculated different fractions of gasoline and diesel cars when dividing Maribor in different sized areas. In the center they found that the proportion of gasoline and diesel cars was 71 and 29%. Increasing the area from the city center to the more industrialized area under the Drava River would respectively change the percentage to 61 and 39%. In the broad Maribor municipality the percentage of gasoline and diesel cars would respectively change to 68 and 32%. We kept the national percentage of gasoline and diesel cars in the fleet because they were somewhere in between the narrow and broad area of Maribor, and because there were recorded about 12.5% of vehicles from the cities in near proximity of Maribor and 17.4% from other cities travelling to the Maribor city center (Lep and Mesarec, 2013).

In the next step we weighed the cars and LT, with 0.94 and 0.06, respectively. Here the weights were taken from the traffic count data average of the four CTCS (Table 2). The HV category already included different sized trucks so it remained as one category. The resultant subcategory’s EF for BC and NOₓ and their respective weight factors are listed in Table 3.

The uncertainties of all EF were set to 25%. Although Ježek et al., 2015b, determined only the uncertainty for BC EF, we expect it would be the same for NOₓ, since the uncertainty depends mostly on the CO₂ measurements.

### 3.2. Emission reduction scenarios

With the EMISENS model we tested few emission reduction scenarios in the city center where the air pollution monitoring station was positioned. We set up 4 scenarios:

a) excluding high BC emitters;

b) excluding high NOₓ emitters;

c) excluding vehicles older than 15 years;

d) replacing HV with LT.

We know that high BC emitters are not also high NOₓ emitters (Ban-Weiss et al., 2009; Irena Ježek et al., 2015a; Wang et al., 2012), therefore we tested the co-benefits of excluding either high BC (scenario a) or high NOₓ (scenario b) emitters from the fleet. We did this by calculating the EF distributions in the same manner as described in subsection 3.1.4, only we first excluded 10% of highest BC (or NOₓ) emitting cars (regardless of the fuel) and then integrated each distribution. As there are differences in the purpose and the necessity of heavy, light trucks and cars in the city, we eliminated top 10 percent in each category. We did not however exclude 10% of highest emitting gasoline and 10% highest emitting diesel cars respectively, because their purpose of use is similar. In the interest of lowering BC emissions in the city excluding high emitting cars results in eliminating high emitting diesel cars. A similar argument holds for NOₓ, only here some of the high emitting gasoline cars do emit as much as the high emitting diesel cars (Fig. 5).

In scenario -c- we excluded vehicles older than 15 years (their age in year 2011 when the EF measurements were conducted). For cars this measure would exclude those that were compliant with the Euro 1 (or less) vehicle emission standard. A similar, measure was already taken in

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<th>Subcat. II</th>
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<th>EF NOₓ</th>
<th>Weight factor α</th>
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<td>0.560</td>
<td>0.63</td>
<td>0.94</td>
</tr>
<tr>
<td></td>
<td>Diesel cars</td>
<td>60</td>
<td>0.072</td>
<td>1.036</td>
<td>0.36</td>
<td></td>
</tr>
<tr>
<td>LT</td>
<td></td>
<td>80</td>
<td>0.066</td>
<td>2.222</td>
<td>/</td>
<td>0.06</td>
</tr>
<tr>
<td>HV (&gt; 3.5t)</td>
<td></td>
<td>240</td>
<td>0.237</td>
<td>7.917</td>
<td>/</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 5. Showing the emission factor distribution on a logarithmic scale for four vehicle categories, using consecutive 10 percentile EF values as bin delimiters.
Table 4

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Vehicle category</th>
<th>BC EF (g/km)</th>
<th>NOx EF (g/km)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial situation in Maribor</td>
<td>LV</td>
<td>0.078</td>
<td>2.029</td>
</tr>
<tr>
<td></td>
<td>HV</td>
<td>0.237</td>
<td>7.917</td>
</tr>
<tr>
<td>a) excl. high BC emitters</td>
<td>LV</td>
<td>0.062</td>
<td>1.637</td>
</tr>
<tr>
<td></td>
<td>HV</td>
<td>0.241</td>
<td>8.002</td>
</tr>
<tr>
<td>b) excl. high NOx emitters</td>
<td>LV</td>
<td>0.05</td>
<td>1.637</td>
</tr>
<tr>
<td></td>
<td>HV</td>
<td>0.087</td>
<td>8.002</td>
</tr>
<tr>
<td>c) excl. vehicles older than 15 years</td>
<td>LV</td>
<td>0.062</td>
<td>1.317</td>
</tr>
<tr>
<td></td>
<td>HV</td>
<td>0.243</td>
<td>5.954</td>
</tr>
<tr>
<td>d) replacing HV with LT</td>
<td>LV</td>
<td>0.078</td>
<td>2.029</td>
</tr>
<tr>
<td></td>
<td>HV</td>
<td>0.066</td>
<td>2.222</td>
</tr>
</tbody>
</table>

Maribor in 2013. It excluded vehicles compliant with standards Euro 1 or less in the narrow city center. The results did not show any improvement in air quality (Jezek et al., 2013), probably because the measures were only vaguely declared and the execution was not strictly enforced.

With scenario -d- we wanted to test by how much the emissions would be reduced, if there were no heavy trucks in the city center. We wanted to see how much influence the heavy trucks have on the air pollution, since there are not many in the city to start with (73% of street segments have 5% of HV or less). We replaced heavy trucks with light trucks because we expect that goods would still need to be delivered in the city center. This is of course a very simplified reflection of the taken measure. It would probably not be possible to exclude all trucks – i.e. trucks that are used for waste removal. Also removing heavy trucks would probably result in higher number of light trucks, which would be needed to deliver the same amount of goods in the city. The purpose of this scenario is to investigate the influence of HV on traffic contribution to air pollution, because we found that the HV AADT on the road map was over estimated. EF used in the emission reduction scenarios are summarised in Table 4.

4. Dispersion of pollutants

The ambient concentrations of pollutants are dependent on the intensity of their sources and the meteorological conditions, which ultimately determine the time, location and magnitude of the exceedances (Seinfeld and Pandis, 2006). The dispersion of pollutants in the atmosphere depends on the action of turbulent eddy motion and advection of wind, heat and humidity.

Local concentration $C$ of pollutant changes with time due to the advection of pollutant with wind; turbulent dispersion in horizontal and vertical directions; pollutant sources $Q$ and sinks (Rakovec and Vrhavec, 2007). The dispersion calculation may be based on the Gaussian dispersion model, where the ambient pollutant concentrations are calculated from the source intensity and atmospheric advection and dispersion, which includes different meteorological parameters. Since BC is an inert pollutant and is removed from the atmosphere by deposition only, and there is not a lot of sunlight, the temperature is low and there is little ozone ($O_3$) in the winter when, NOx can also be assumed as a non-reactant tracer in 1 h time intervals (Rakovec and Vrhavec, 2007), we can simplify the dispersion equation for both pollutants into:

$$C(x, y, z) = \frac{Q}{D} \quad (1)$$

where all meteorological parameters are combined in dispersion $D$. Even though this is a very simplified case, we used the Gaussian dispersion model because it is simple, it gives results that agree well with experimental data, and has thus frequently been used in air pollution modelling and environmental studies (Hanna et al., 1982; Turner, 1971).

We empirically determined $D$ from the intensity of the NOx traffic emissions $Q_{NOx}$ and the traffic related ambient NOx concentrations $C[NOx traffic]$.

$$D = \frac{Q_{NOx}}{C[NOx traffic]} \quad (2)$$

For the source intensity of NOx, we used the hourly NOx emissions modeled with the EMISENS model for the grid cell where the urban ambient air monitoring station was located. We calculated the traffic related ambient NOx concentrations $C[NOx traffic]$ from NOx concentrations measured at the urban and background ambient air monitoring station (described in Section 2) by subtracting the NOx concentrations measured at the background station from those measured in the city, thereby excluding from our dispersion calculation the influence of NOx coming from other sources and traffic plume history.

$$C[NOx traffic] = C[NO, urban] - C[NO, background] \quad (3)$$

For the purpose of converting NOx from ppb concentration to its air ratio ($\mu g/m^3$), we used standard conditions (100 kPa and 0 °C), the molar mass for NOx was set with 20% NO2 and 80% NO (Monks et al., 2015). NOx and BC emitted by traffic have the same sources. We therefore assume that, in 1 h observation intervals, meteorology affected the emissions of both pollutants the same way, and apply the same dispersion to the modeled traffic BC emissions ($Q_{BC}$).

$$C[BC model] = \frac{Q_{BC}}{D} = \frac{C[NOx traffic]}{Q_{NOx}} \cdot Q_{BC} \quad (4)$$

The comparison of the thus modeled traffic BC concentrations $C[BC model]$ was made to the BC ambient concentrations measured at the urban ambient air monitoring station. These were source apportioned to traffic using the Aethalometer model ($BC_{at}$); we also accounted for the traffic plume history by subtracting the $BC_{at}$ measured at the background site from the urban $BC_{at}$ measurements:

$$C[BCff traffic] = C[BCff urban] - C[BCff background] \quad (5)$$

A similar method was previously used in reverse for calculating emission factors from ambient pollutant concentrations and traffic counts (Imhof et al., 2005).

We calculated the modeled BC concentrations for three winter periods for which we had valid NOx and BC measurements available at the city center and urban background sites. We limited our research to workdays because there is more traffic in workdays and the sources are better defined during these days. The ambient measurements of NOx and the source apportioned BC for the investigated time periods are shown in Supplement Figure 6-8. We can see that on urban station, the measured NOx and $BC_{at}$ concentrations have similar dynamics, with the exception of some midday NOx peaks, while at the background station their dynamics are not as closely related and the BC peaks are more pronounced.

5. Results

5.1. Spatial and temporal traffic emission rates

The results of the traffic emission model are hourly emission rates on an average workday, spatially distributed over the domain. The diurnal emission variations of BC and NOx for the grid cell with the urban measurement stations are shown in Fig. 6. Because cars are the dominant vehicle category in the city, the temporal variability of emission rates is very similar to the LV traffic flux (Fig. 3 c). The first
emission peak occurs at 7:00 CET and the second at 15:00 CET, which reflects people travelling to and from work.

An example of spatially distributed BC emissions is plotted in Fig. 7, where we show the map of emissions at 15:00 CET, the highest daily emission peak. We can see that the grid cell with the urban measurement station is not the most laden with traffic emissions; instead we can see that most traffic emissions are produced in the red, orange, yellow and green colored areas, which cover the expressway, the highway and other main city aortas, reflecting the AADT in Supplement Figure 2.

EMISENS model computes also the uncertainties on the modeled emissions if uncertainties on mileage and EF can be estimated. Taking into account a sensitivity module based on a Monte Carlo simulation, it is also possible to compare the influence of several parameters. Therefore, the input information that contributed most to the model uncertainty on average were the uncertainty in LV EF (21% and 20%, for BC and NOx emission rates respectively), followed by LV street mileage (7% and 6% for BC and NOx emission rates respectively), HV EF (5% and 6%, for BC and NOx emission rates respectively), and lastly the HV street mileage (3% for both BC and NOx emission rates). The uncertainty of the calculated emissions in the grid box with the ambient air pollution measurement station is on average during the day 21% and 24% for BC and NOx emission rates respectively.

5.2. Comparison of ambient measurements and modeled results

We present the linear regression analysis between the ambient BC measurements (which were apportioned to fossil fuel burning with the Aethalometer model, and from which the traffic plume history was subtracted) and BC concentrations that were the results of emission/dispersion model for each of the three investigated time periods in Fig. 8. The intercept was forced through zero. In all three time periods we get an excellent agreement between the two, Pearson’s $r$ is 0.95 or
and by 10% would change the traffic emission-dispersion model’s BC results (BCmodel) and BC apportioned to traffic with the Aethalometer model (BCif traffic) in the three investigated time periods: (i) 2nd – 16th November 2011; (ii) 1st February – 14th March 2012; and (iii) 26th November – 31st January 2013.

### Table 5

| Total workday traffic black carbon (BC) and nitrogen oxides (NOx) emissions in the grid cell with the urban air pollution monitoring station and the effectiveness of four emission reduction scenarios where we exclude vehicles that are high emitters (HE) of BC or NOx; vehicles that were older than 15 years, or replace heavy vehicles (HV) with light trucks (LT). |
|---|---|---|---|---|
| BC | 3907.9 | 39% | 17% | 18% | 9% |
| NOx | 105451.8 | 16% | 33% | 16% | 12% |

0.96, and the slope is 1.19, 1.32, and 1.06 for the three investigated time series, which indicated that our models’ results are good. There is some bias for the three time periods: 1.05, 1.36 and 0.52° respectively, which contribute to the error reflected in root mean squares for the three periods: 1.75, 2.31 and 2.01.

Aethalometer measurements assume MAC values to convert the measured optical absorption to BC mass. The exact MAC depends on aerosol mixing state and the reference method used to determine it (Zanatta et al., 2016). In the second time period we tested the influence of changing MAC by ±10%. Increasing MAC by 10% would decrease BC concentrations and BCif 10%, while decreasing MAC would increase it by 10%. The resulted slope between emission/dispersion model and the Aethalometer model would change by 3%. Changing MAC wouldn’t influence the source apportionment because MAC is the same for fossil fuel and wood burning sources (Zotter et al., 2017), but changing the 2 Å exponents used in the Aethalometer model would.

The time series of the BC concentration measurements (BCif traffic), which were apportioned to fossil fuel burning with the Aethalometer model and the NOx measurements from both urban and background air pollution monitoring stations, together with the modeled BC concentrations (BCmodel) for the three investigated time periods are presented in Supplement Figures 6–8. We can see that there are time periods where the model performs better or worse.

We investigated the influence of meteorology on our models results by comparing several weather parameters measured by Slovenian Environmental Agency at Maribor airport (the data was accessed on their web site http://meteo.arso.gov.si/met/) to the residual relative to in situ measurements ((BCif traffic)−[BC model])/BCif traffic). We did not find any relation to the measured temperature, pressure, wind direction nor precipitation, and a weak relation to wind speed (and maximum wind speed), relative humidity and global (and diffused) radiation. We present the latter in Supplement Figure 9 (maximum wind speed and diffused radiation are very similar to wind speed and global radiation so we omitted those) together with relation of in-situ measurements to the relative residual. We get the highest negative relative residuals when the traffic related BC concentrations are low during night time when there is high relative humidity and low wind speed. These high negative values represent only small portion of data.

5.3. Results of different emission reduction scenarios

We tested four emission reduction scenarios to see how different emission reduction measures would affect emission rates in the city center where the city center air pollution measurement station is positioned. The results of different measures are depicted in Fig. 5 next to EMISENS model results for an average workday and summarized in Table 5. In the first scenario (a) where we excluded 10% of highest BC emitting vehicles in categories cars, LT and HV, daily BC emissions were reduced by 39% and NOx emissions by 16%. In the second scenario (b) we excluded 10% of highest NOx emitting vehicles. This reduced NOx emissions by 33% and BC emissions by 17%. In scenario (c) we excluded vehicles older than 15 years. With this measure we got 18% and 16% reduction for BC and NOx emissions respectively. Excluding either high BC or NOx emitters yielded better results for the targeted pollutant than targeting vehicle age, while producing similar results for the not-targeted pollutant.

In scenario (d) we replaced all HV with LT. Doing this produced 9% reduction in daily BC emission rates and 12% reduction in daily NOx rates. Even though HV EF are 4 and 3.5 times as high as light trucks BC and NOx EF respectively, they represent a small fraction of vehicles in the city center (5% of vehicles on 73% of road segments). Because trucks have high EF in terms of pollutant per km driven, it would be somewhat efficient to use trucks that pollute less even if there aren’t...
many of such vehicles in the fleet to start with. Replacing HV with LV is not exactly a realistic scenario. We should have accounted for the increase in number of LV because a single HV can transport more than a single LV. We did not explore this further because the main question we wanted to answer with this scenario was to see how grave misrepresenting the amount of trucks in the fleet is. We found that it can create a small bias.

These strategies would have to be tested again for a different city, where the fleet composition and traffic situation differs from that in Maribor. However for Maribor it indicates how effective each measure would be on an average workday in the city center, if fully implemented.

5.4. Discussion

In this study we have used the on-road EF measurements in an emission/dispersion model and validated the model results with measured BC concentrations apportioned to traffic with the Aethalometer model. Using a primary pollutant (BC) we aimed to reduce the uncertainty due to discrepancy between different definitions of PM between emission models and observations (Smit et al., 2010). Using background measurements and the Aethalometer model to apportion BC to a single source enabled us to reduce the influence of non-traffic sources on the emission/dispersion model results.

The spatial and temporal results of EMISENS model reflect the AADT and traffic flux. When testing the input data we found that the LV category was dominant in the city (according to the CTCS and the road map data) and that the workday diurnal traffic flux had small variation (standard deviation of the relative LV daily traffic was 0.08). HV were not as abundant as LV in the city, 73% of street segments had less than 5% of HV in the fleet and the rest had less than 20% (Supplement Figure 5). Because there were fewer HV in the city fleet, the variation would be higher (standard deviation of the relative HV daily traffic was 0.14), however their influence on the traffic emission models uncertainty would be low (around 5% regarding the EF and 3% regarding the calculated street mileage).

We evaluated our traffic emission models’ results with in situ BC measurements that were apportioned to traffic using the Aethalometer model and by empirically determining the dispersion of pollutants with NOx in situ measurements and modeled NOx emission rates. We found excellent agreement between the results of traffic emission/dispersion model and the in situ measurements. For the three time periods we found that our model overestimated the in situ measurements by 19%, 32% and 6% (Fig. 8).

Although there has been research performed where they could explain why during increased wind episodes the measurements at the background station would underestimate the dispersion and thus overestimate the modeled BC concentrations (Price et al. (2014) found that there was replenishment of fine particles from the traffic when the wind was parallel to the canyon. And Jones et al. (2010) found an inverse behavior of NOx and elemental carbon when the wind speed exceeded 2.5 m/s, which they attributed to the effects of complex flow pattern within the street canyon and to the buoyancy of the exhaust emissions; and to the fact that the wind speed in the street canyon may not be well represented with wind speed measurements elsewhere (the airport with wind speed measurements was in our case within 25 km of the measurement site). We did not find any weather parameter that would systematically cause our model to perform better or worse. The premise that the weather influences both BC and NOx in winter in 1 h time periods the same way turned out to be valid.

The results of the emission reduction scenarios show how the emissions would be impacted, if we excluded specific vehicle groups. Targeting high BC or high NOx emitters would respectively bring highest BC (39%) or NOx (33%) emission reductions in the city. Excluding the similar percentage of vehicles in 2011, while excluding vehicles based on their age, would reduce BC and NOx traffic emissions by 18% and 16% respectively. These results show that not all old vehicles are super emitters and not all high BC emitters are also high NOx emitters. Targeting high emitters instead of the old vehicles would be in the interest of air quality a more efficient and just measure.

When we were eliminating high emitters we excluded 10% of them in car, LT and HV categories. Excluding vehicles older than 15 years would in the year 2011 exclude 12% of cars, 10% of busses and 11% of different sized trucks (according to Republic of Slovenia Statistical office). In the following years, this percentage would decrease and with new Euro emission standards in place, these reductions would no longer hold. Newer vehicles have to comply with more stringent emission standards, thus they are equipped with better emission regulation mechanism. These can deteriorate with time or in individual new vehicles may not be working properly, in such instance identifying and excluding high emitters could turn out to be even more beneficial in reducing traffic emission rates.

6. Conclusions

We calculated the traffic emission rates of BC and NOx for an average workday in a small Central European city (Maribor, Slovenia) using the EMISENS model. The empirical dilution factor of traffic emissions was calculated from the ambient measurements of NOx and modelled NOx emission rates. This factor was used to calculate BC concentrations from traffic emission rates. The modeled BC concentrations were then compared to BC from the in-situ measurements apportioned to traffic. A good agreement between the two approaches was found, with the modelled BC concentrations overestimating the measured BC by 19%, 32% and 6% in three different winter time periods. This was the first time the source apportionment of BC using Aethalometer model had been used to evaluate the results of emission inventory model results.

We did not find any influence of meteorology on the performance of the model. However, since we made numerous estimations, several factors could have caused the relatively small discrepancies between the modeled concentrations and ambient measurements. Our background measurements could have not been a representative background for the urban measurement site. The investigated pollutants BC and NOx could have mixed differently, or there could have been loses to either of the two pollutants, by which we would underestimate our background levels.

The modeling domain we used was small, the area was in a more or less flat valley, and the street speed limits were homogenous, which simplified the input information for the model. To confirm that the BC concentration predictions are accurate over the entire modeling domain, additional ambient measurements would be required especially in cells that include highway and expressway traffic emissions.

With the EMISENS model we also tested 4 emission reduction scenarios, where we first excluded 10% of highest BC emitters, second we excluded 10% of the highest NOx emitters, third we excluded vehicles older than 15 years and lastly we replaced HV with LT. We found that even though trucks are not numerous on the streets of Maribor, replacing HV with LV would reduce the emission rates and hence local BC and NOx concentrations by 9% and 12% respectively. This indicates that there is a small bias when misestimating the number of HV in the city center. If the measure to exclude vehicles older than 15 years would be enforced in the year 2011, it would deny about 10% of all vehicles entrance to the city center. This would reduce BC and NOx emission rates by 19% and 17% respectively. If on the other hand 10% of highest BC (or NOx) polluters would be excluded, the emission rates of the pollutant would be reduced by 39% (or 33%). The co-benefits on NOx traffic emission rates when excluding high BC emitters, and vice versa on BC emission rates when excluding high NOx emitters, would be similar to excluding vehicles that were older than 15 years in 2011.

This work presents a detailed methodology to determine traffic emissions in a city and a simple method to verify the emission models
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results. Such verified model can then be used to simulate the most efficient traffic emission reduction measure for the investigated city.

Conflicts of interest

At the time this research was conducted I. Ježek and G. Močnik were employed in Aerosol d.o.o. where Aethalometers are developed and manufactured.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.

References


