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ORIGINAL PAPER

NO_2-immission assessment for an urban hot-spot by modelling the emission–immission interaction

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Abstract
Urban air quality and climate protection are two major challenges for future mobility systems. Despite the steady reduction of pollutant emissions from vehicles over past decades, local immission load within cities partially still reaches heights, which are considered potentially hazardous to human health. Although traffic-related emissions account for a major part of the overall urban pollution, modelling the exact interaction remains challenging. At the same time, even lower vehicle emissions can be achieved by using synthetic fuels and the latest exhaust gas cleaning technologies. In the paper at hand, a neural network modelling approach for traffic-induced immission load is presented. On this basis, a categorization of vehicle concepts regarding their immission contribution within an impact scale is proposed. Furthermore, changes in the immission load as a result of different fleet compositions and emission factors are analysed within different scenarios. A final comparison is made as to which modification measures in the vehicle fleet offer the greatest potential for overall cleaner air.

Keywords Air quality · Zero impact · SubZero · Emission · Immission · Emission–immission-interaction · Synthetic fuel · OME

Abbreviations

CI	Compression ignition
CO_2	Carbon dioxide
DOC	Diesel oxidation catalyst
EFA	Emission factor
EHC	Electrically heated catalyst
FEP	Future emission potential
HBEFA	Handbook emission factors
HDV	Heavy-duty vehicles
HLNUG	Hessian agency for nature conservation, environment and geology
LDV	Light-duty vehicles
NO_2	Nitrogen dioxide
NO_x	Nitrogen oxides
OME	Polyoxymethylene dimethyl ether
PCV	Passenger car vehicles
RDE	Real driving emissions
SCR	Selective catalytic reduction
sDPF	SCR substrate on diesel particulate filter

List of Symbols

e_w	Emission factor, weighted average
e_i	Emission factor of vehicle segment
x_i	Market share of vehicle segment

1 Introduction

Global efforts seek to minimize urban air pollution. According to research in the field of general health, air pollution is considered to be a major cause of many severe diseases that potentially cause premature deaths [1–4]. Unlike other reasons for adverse health effects, in the case of air pollution, individuals have in most cases no direct choice to avoid exposure, given its ubiquity. In addition to the influences on human healthiness, further negative aspects, such as the consequences for the environment, have led to the introduction of measures for air quality control in the past. As a result, significant improvements in urban air quality were realized up to today [5–8]. However, under special conditions exceedances of the legal limits of NO_2 or particulates remain locally and besides other influencing factors and emitters, vehicle fleet emissions continue to be a major influencing factor [5, 9, 10].

Despite the fact of the high importance of the vehicle emission influence, there is still a lack of knowledge about

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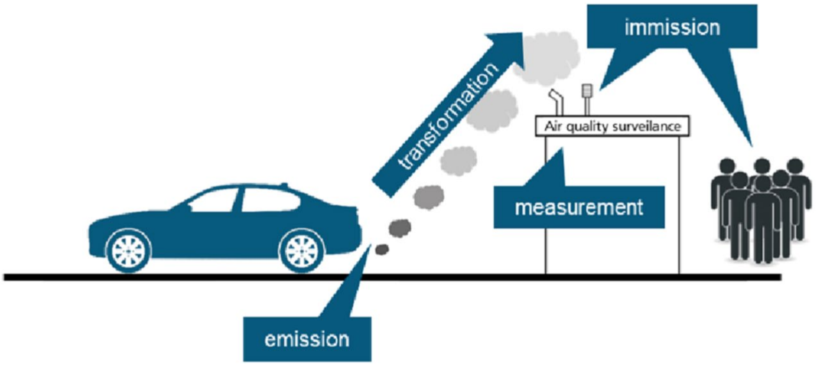

Fig. 1 Emission-Immission interaction for a measurement site—emissions leave vehicles, are being transformed within the ambient air and then exposed to e.g. pedestrians or air quality surveillance stations as immission

the exact interaction between the overserved immission and how it corresponds to the vehicle fleet emission leading to it. Based on an empirical model, this paper addresses the question raised and gives an estimation of the immission impact due to emissions caused by certain vehicle fleets for NO_2. The model is derived on the basis of recorded data for a hot spot measurement station within the city of Darmstadt, Germany, which surpassed the legal limit in the past repeatedly. With the derived interaction, immission potentials for certain technologies can be quantified on the basis of the model, which is done for current type approval technologies. Further, an emission level scale is introduced that rates the influence of vehicle categories based on the attributed immission.

1.1 Emission–immission interaction

Pollutants emitted by combustion engine-powered vehicles leave the tailpipe in a certain concentration referred to as emission. Over time, the pollutants mix with ambient air, get diluted due to spatial expansion, react with other substances and reach the respiratory passages of city inhabitants in a different concentration, referred to as immission (see Fig. 1). The effects of emission and transformation on the immission depend on numerous factors and boundary conditions [11–15]. To control the health-relevant immission level in cities with high traffic volume, the European Parliament and council defined limit values for certain air pollutants within Directive 2008/50/EC [16]. Air quality is hence monitored close to traffic in hot-spot areas with high immission concentration as well as in urban and rural background areas.

For the purpose of a detailed analysis of the prevailing immission load, it is indispensable to precisely describe the interaction between the place of origin and the place of observation, considering the influencing parameters. The pollutants emitted by vehicles first enter a primary area, which is subsequently referred to as a driving tube. It is defined here as the volume immediately behind a vehicle that results from spanning its frontal area along the longitudinal direction of the road. Subsequent vehicles draw their inlet air from this tube and transformative effects of the emission components already occur here. These continue in the further course of the secondary area between the driving tube and the measuring point.

1.2 Investigated measurement site

The measurement scenario investigated in this paper is the local, near-traffic situation at the immission hot-spot Hügelstraße in the city of Darmstadt, Germany. For this purpose, data of three air quality measurement stations for near-traffic (1), urban background (2) and rural background (3) is considered [18–19]. The urban background station is equipped with additional sensors for environmental conditions and located within a small park [18]. The rural background station is located approximately 30 km outside the city on a field [19]. Additionally, the number of vehicles passing the near-traffic measurement station is measured by an inductive loop integrated into the pavement which also allows for vehicle classification. The considered parameters for the investigation of the emission-immission interaction, which are available as measurement data, are listed in Table 1.

Table 1 Regarded parameters and used data sources based on HLNUG (public and non-public data) [20]

Value	Measurement station
Traffic volume	Near-traffic (1)
Temperature	Urban background (2)
Air pressure	Urban background (2)
Wind direction	Urban background (2)
Wind velocity	Urban background (2)
Humidity	Urban background (2)
Solar radiation	Rural background (3)
O_3 immission	Urban background (2)
NO_2 rural background	Rural background (3)
NO_2 urban background	Urban background (2)
NO_2 near-traffic	Near-traffic (1)

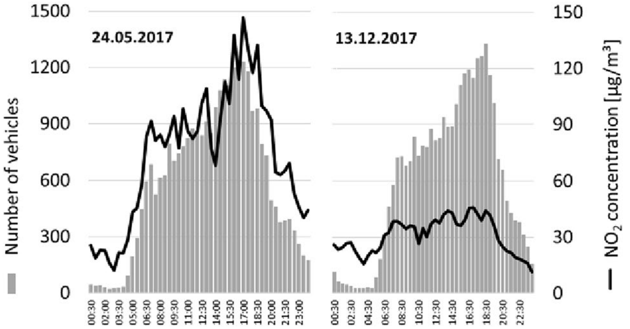

Fig. 2 Exemplary days with strong (summer) and weak (winter) Traffic-NO_2-immission correlation at the near-traffic air quality measurement station Darmstadt Hügelstraße (1). Measured values based on 30-min averages

All listed parameters are provided with a time resolution of 30 min as half-hour average values, except for the number of passing vehicles which is tracked as absolute count [18–19]. Unless explicitly stated otherwise, all subsequent analyses are based on the 30-min resolution of the raw data. First raw data analysis allows to determine possible influencing factors and the magnitude of the impact. These parameters are essential as they have to be integrated into the traffic emission-immission model to be built and derived in the following sections of this paper.

Being the main emitter of nitrogen oxides (NO_x) within cities, vehicles show a strong correlation by their number in terms of the prevailing NO_2-immission load. Increasing NO_2 concentrations as a result of high traffic are described throughout various publications [5, 22–27] and this relation is adopted. However, cases exist where the measured NO_2-immission seems to be decoupled from the number of vehicles passing the measurement station.

Figure 2 shows two exemplary days with similar traffic intensity. While the NO_2 concentration on the 24.05.2017 (avg. 16 °C) corresponds strongly to the number of vehicles, the monitored immission on the 13.12.2017 (avg. 3 °C) seems to be almost independent of it. Since both days are of the same year, where vehicle emissions are expected to be comparable, it becomes obvious that additional factors influence the prevailing immission load. In this particular case, there was prolonged rainfall in the second half of the day on December 13th [28], which can lead to the leaching of ambient NO_2. However, this effect cannot be explicitly read off in the measured NO_2.

Further analysis shows increasing NO_2 concentration with higher ambient temperatures and lower humidity, illustrated in Fig. 3. These environmental influences are attributed to fundamentally effective interactions and chemical reactions within the air. To account for further transformative effects, the solar radiation intensity and ozone (O_3) concentration as a reaction product are considered as model input parameters.

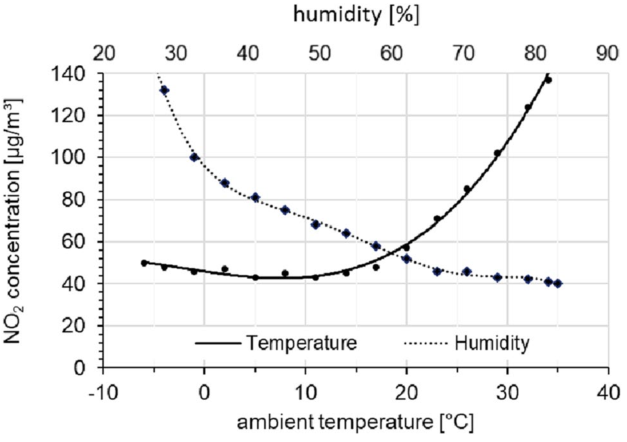

Fig. 3 Average NO_2 concentration over temperature and humidity at the near-traffic air quality measurement station Darmstadt Hügelstraße (1), (2017–2018)

Besides, the dataset used is also subject to specific local influences. A highly relevant one is a tunnel exit, located approximately 100 m westwards, in the single direction of travel (one-way street) just before the measurement station. Due to unfavourable spatial dilution and the single direction of travel towards the station, it is assumed that the tunnel acts as a chimney. Moreover, the street is ascending significantly prior to the measurement station. The stated situation is likely to be one of the main causes why this particular station is among Germanys hot spot stations with the highest limit violations [29]. The described chimney effect is also subject to weather influences. Analysis of the prevailing wind directions proof high immission concentrations when the wind is coming from north-westerly directions, as shown in Fig. 4, which coincides with the orientation of the tunnel exit and the measurement station. Vice versa, the immission load remains significantly lower for wind coming from southern-easterly directions, whereby pollutants are transported in a direction away from the station.

To sum up, according to expectations, the measured immission value is strongly influenced by the prevailing ambient conditions. Due to superposition effects, a direct attribution of single influencing factors to the measured immission value is not possible. Intersection analyses can provide valuable information about direct dependencies, but at the same time considers other influencing factors as well. Likewise, a direct causal attribution based on the isolated consideration of influencing factors is generally inadmissible, since correlations might falsely be assumed as possible causalities. A good example is the plot of ambient NO_2 concentration versus ambient temperature in Fig. 3: physically, no direct explanation exists why the NO_2 concentration should decrease at lower temperatures with comparable traffic intensity. On the contrary, it is to be expected that

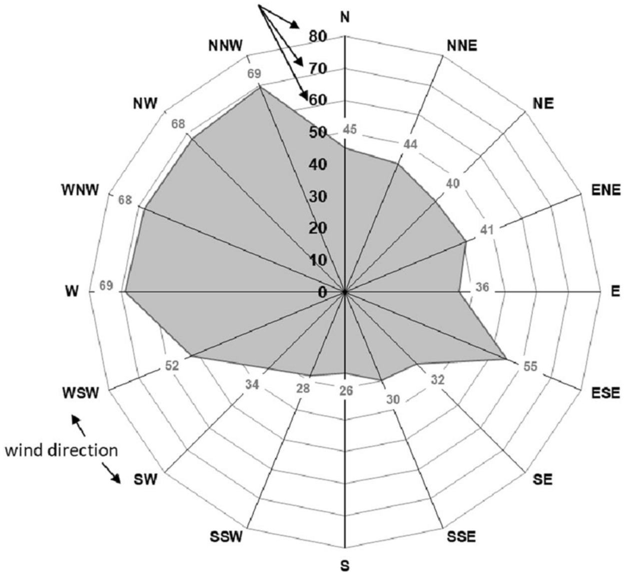

Fig. 4 Average NO_2 concentration in relation to the wind direction at the near-traffic air quality measurement station Darmstadt Hügelstraße (1), (2017–2018)

adapted combustion engine operation (lower exhaust gas recirculation rates) in combination with residential heating will lead to an increase in the prevailing NO_2 load during winter time. The measurement data analysis shows that significantly higher wind speeds are recorded in the winter half-year, but at the same time, for example, the intensity of the global radiation is significantly weakened. In total, the result is a measured reduction, which, however, cannot be described on the basis of a single influencing factor. It can therefore only be depicted correctly if the interaction of all influencing factors is considered accordingly. To achieve this goal, the approach of a model for the description of the emission-immission behaviour under consideration of the prevailing ambient conditions is pursued.

2 Methodology

When it comes to gaseous pollutants, the healthiness of humans and nature is affected by the exposure to a certain concentration over a certain time, known as immission. To a great extent, high immission levels are observed in areas with a high level of anthropogenic activity [30, 31]. Mobility enables and drives human activities in terms of economy but for private purposes as well. Today's mobility systems rely to great extent on the usage of fossil, carbon-based fuels whose combustion is related to the release of pollutants. Hence, it can be concluded and has been shown in the past, that inner-city immission levels are impacted by the local traffic volume. However, it remains challenging to quantify the interaction between vehicle emissions and the exact amount of resulting immission load due to the complex mechanisms that effect the dilution and further reaction of pollutants leaving a vehicle [5, 22–27, 30]. For a given vehicle fleet with corresponding emissions, the observed immission concentration depends on numerous environmental conditions like the ambient temperature, humidity, wind direction, wind velocity, air pressure, solar radiation and the distance between emission source and immission measurement point. However, current legal limits for vehicles are not derived on the basis on their associated effects in terms of immission but on their direct emission instead [32]. To ensure fast immission reduction, also taking additional influencing factors such as technology availability, market penetration or cost considerations into account, a valid quantification basis for the emission-immission interaction can help to identify key paths towards cleaner air. The approach presented in this paper is based on the following three steps:

1. *Immission modelling* In order to calculate the effects of traffic emission on inner-city immission, an empirical model is derived on the basis of measurement data for one representative, near-traffic hot spot measurement station. Besides measured immission concentrations, data of relevant influencing factors (see Table 1) is included, to account for dilutive and especially conversion effects. The traffic-related emission is considered on the basis of measurement data for the number of vehicles passing the measurement point in combination with specific vehicle emission factors taken from HBEFA.
2. *Estimation of future emission values* For the purpose of projecting future emission values, both, public sources and own investigations with alternative, potentially CO_2 neutral fuel are evaluated. Part of this analysis is the transfer of concentration-based measurements to legally compliant, distance specific emission values.
3. *Scenario analysis* Finally, the results from steps 1 and 2 are combined in this final step, where different scenarios for fleet development and replacement are being investigated. This includes the upgrade to today's newest technology, successor technology, and also considering electric vehicles. On the basis of these finding, the corresponding emission classes are categorized, based on their impact for urban air quality.

3 Immission modelling

With the goal to estimate the impact of different traffic scenarios on the measured NO_2 concentration for the local near traffic station *Darmstadt Hügelstraße*, an immission simulation model is proposed in this paper. Based on the data available, an empirical model is chosen over a physical dispersion model. Whereas physical models deliver comparably robust results as well and are used commonly for air pollution control plans, the effort for modelling is comparably high and deviations remain [13]. Moreover, emission quantification, as taken from HBEFA, is itself subject to inaccuracies if influencing factors and mechanisms are not correctly represented in there. By choosing an empirical model, it is possible that such error influences are compensated for by implicit model relations derived during the model training. Hence, such error compensations become therefore part of the model through the calibration process during model training.

Kohoutek [33] gives an overview of different immission modelling techniques and the achievable accuracies. He concludes, that degrees of certainty up to 80 are feasible with neural networks. Neuronal network models also showed high degrees of determination for immission modelling in [34], where the influence of traffic is modelled implicitly. Compared to physical models, the advantage is that indirect and unknown effects, as well as possible data errors, can, under certain circumstances, be considered within the implicit model generation, as long as they are reflected in the input data. Thus, the procedure is partly more robust against errors. For the given purpose, a 4th order robust neuronal network model (RNN) is selected. To determine the impact of synthetic or future traffic scenarios, the emission value in the proposed model has to be considered explicitly.

The primary task of the model is to reproduce the transformation process within the ambient air from the emitters to the immission measurement point. This process is subject to a variety of influences, some of which also interact with each other. The mechanisms that are effective here can be divided into conversion, dispersion and deposition, which are in turn influenced by local conditions such as the arrangement of the building, but above all by meteorological influences. The prevailing, ambient NO_2 concentration is subject to two fundamental reactions, which are responsible for its formation and degradation [35]. NO_2 is formed as a reaction product of NO on the following three paths:

$$2NO + O_2 \rightarrow 2NO_2,$$
$$NO + O_3 \rightarrow NO_2 + O_2,$$
$$NO + \text{peroxide radicals} \rightarrow NO_2 + \text{radicals}.$$

Likewise, atmospheric NO_2 can react with atmospheric oxygen to form ozone under the presence of solar radiation

$$NO_2 + O_2 + \text{solar radiation} \rightarrow NO + O_3.$$

For this reason, NO_2 is also considered a precursor substance for O_3. To sum up, at a traffic hotspot, the NO_2 concentration is critically dependent on the primary emitted nitrogen monoxide NO_xide (NO), the photochemically produced ozone (O_3) and the intensity of global radiation [33]. For this reason, the discussed parameters are being used as model inputs besides general weather influences (wind, temperature, humidity, pressure). All used parameters are listed in Table 1. The interactions and reactions are not described explicitly but expected to be represented within the model. In comparison to other modelling approaches, the tailpipe NO-NO_2 ratio is not used as explicit model input. It is assumed that the model also represents the underlying reactions from emitted NO_x to ambient NO_2 at the measurement station. Nevertheless, the tested integration of the NO–NO_2 ratio as an additional model input parameter is carried out, with the result that, according to the stated expectations, no significant change in the modelling quality was observed.

Determination of the total vehicle fleet emission, relevant for the resulting immission value, is calculated from each vehicle's emission and the number of vehicles over time

$$\mathrm{em}_{\mathrm{total}} = \sum_i \mathrm{EFA}_i \cdot n_i,$$

where i represents each vehicle category, EFA_i the category's subsegment emission factor and n_i the vehicle number of each vehicle category. This assumption means that a low number of vehicles with a high emission factor has the same impact on the immission as a high number of vehicles with a respective low emission factor. The EFA is considered as specific NO_x emission in g/km. It is taken from HBEFA, as is the fleet composition (vehicle categories) and their specific emission standards. All stated inputs from HBEFA are considered as constant for each individual year.

In order to obtain traffic immission values, the model is generated within a training process, exported and afterwards used for scenario calculation, as illustrated in Fig. 5. For the training phase, the available measurement data from the near traffic station *Darmstadt Hügelstraße* [20] is used and combined with HBEFA 3.3 fleet emission factors. The used HBEFA version was the newest version available by the time of model training. The model is trained with data from six consecutive years (2012–2018) for the near-traffic measurement station, *Darmstadt Hügelstraße* (1). The time resolution of the input variables and the model used is 30 min, both in the training and in its later use. An analysis showed that delay times of predominant effects are within the measurement resolution, as they correspond directly to the measured NO_2 concentration. Hence, auto-correlation effects are assumed to be non-significant and no further regarded. Compared against the measured concentrations during the training period, the overall coefficient of determination is 85%, with a root mean square error (RMSE) of 15.9 $\mu g/m^3$ which equals normalized (NRMSE) 4.7%, displayed in Fig. 6. Validation

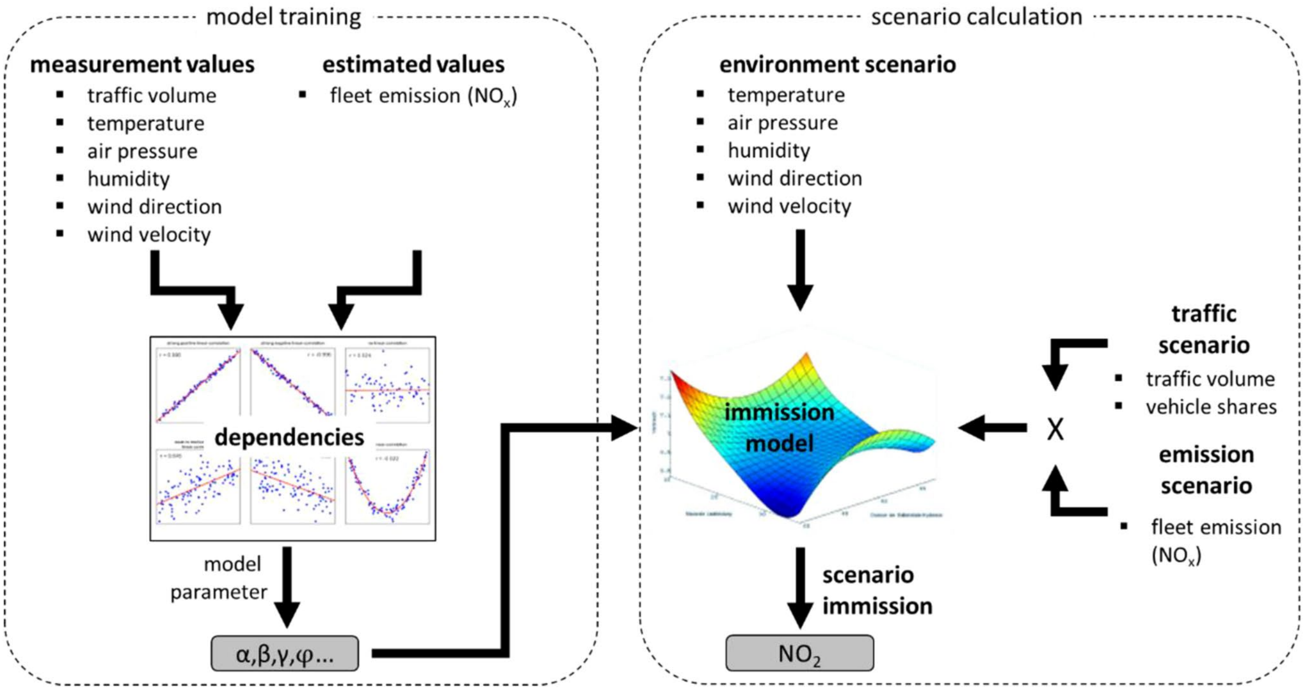

Fig. 5 Schematic description of traffic-induced immission modelling approach for both phases—model training (left) and scenario calculation (right)

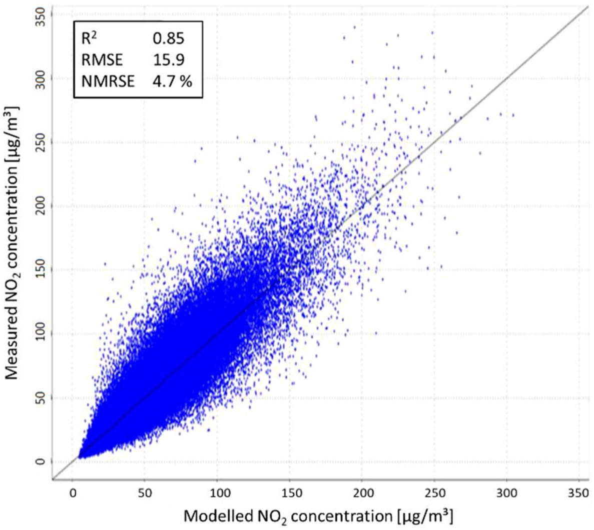

Fig. 6 Correlation of RNN immission model and measured values (30 min resolution) for *Darmstadt Hügelstraße*

of the model is based on the year 2011, which has not been used within the training, by comparing the whole year series of measurement to simulation data. With regard to the regulated, annual mean value, the simulated concentration falls short 1.2 µg/m^3 of the measured value of 64.0 µg/m^3 (1.9%).

4 Emission factors

As described before, the developed immission model utilizes emission values from the Handbook of Emission Factors, HBEFA, 3.3 [36]. These factors rely on different measurement sources which serve as input for an underlying model, that outputs emissions for specific vehicle categories in specific driving situations under consideration of environmental influencing factors [37]. With advances of new emission technology corresponding to imposed emission standards, emission factors of the vehicle fleet are subject to constant change. The procedure implies various uncertainties with regard to the derived emission result. Especially for new technologies with few or no measurement data serving as input for model training, HBEFA values are subject to higher derivation [38]. For future technologies that are in the stage of research and development, the used HBEFA 3.3 offers no outlook data. As a result, the authors present a state-of-the-art screening on the basis of publicly available RDE measurement data to reduce uncertainty regarding the EFA for the current technology level. To consider emission factors reachable in the future, public sources and own results from tested measurements serve as an input basis.

Table 2 Average, weighted urban NO_x emission for passenger cars with emission standard EU 6d, based on RDE measurements listed in ACEA database [40, 44], vehicle shares based on [43]

Segment	B	C	$D+E$	M	J	Avg	σ
Diesel (mg/km)	61.3	76.2	47.3	76.6	76.6	59.5	4.5
Share (%)	3	18	29	30	20		
Gasoline (mg/km)	24.2	33.9	20.8	24.3	35.9	27.5	2.1
Share (%)	23	28	10	31	8		

4.1 State of the art screening

In the immission scenario under consideration, the main share of the traffic-related emissions is attributable to passenger cars (Fig. 7). The EFA of this vehicle category assigned in HBEFA 3.3 should be validated in accordance with the latest Euro 6d-TEMP emissions standard, as there remains uncertainty [38, 38]. For this purpose, RDE data from vehicle manufacturers is used [40],[1] who are obliged to publish the test result data during type approval and in-service operation [42]. With the statistical composition of the passenger car fleet according to total vehicle registrations [43], the weighted average emission value e_w can be calculated as

$$e_w = \sum_{i=1}^{n} e_i x_i,$$

where i is the index variable for different segments, e_i and x_i the respective EFA and market share for the reference year 2018. The sample consists of a total of 827 data sets, 406 for diesel PC and 421 for gasoline PC. Table 2 lists the resulting averages of considered segments, based on their market share and the resulting weighted average for the urban part of the RDE measurements.

For both combustion principles, the legal emission limit [44] is met on average without the consideration of conformity factors (CF) [32]. If the total test distance is considered, specific emissions are even lower. For the following analysis, all EFAs are taken from HBEFA 3.3, except for EU 6d passenger car vehicles (PCV), where the weighted average values of Table 2 serve as input.

4.2 Mid-term technology potential

If combustion engine technology is to be ready for use within the next few years, proof of functionality at the system level must be provided today. That means, fully working prototypes are available and serve as a basis for specific emission value estimates. Among promising technologies for further emission reduction in these concepts for diesel PCV are heated catalysts, pre-turbo-catalyst-systems, higher fuel injection pressure, double-urea-dosing alongside with the overall trend to electrification [46–49, 50]. A comparative study ranges the resulting tailpipe NO_x emissions in urban areas from 10 to 50 mg/km [51].

4.3 Long-term technology potential

To estimate long-term emission levels, own investigations on component level readiness are conducted. Cost aspects and production capacities are not regarded. Considering the challenges of climate change and urban pollution, future engine concepts should ideally be CO_2 free and emit a negligible amount of pollutants. Consequently, within the scope of this paper an engine concept with the potentially CO_2-neutral, synthetic fuel of the class of polyoxymethylene dimethyl ethers (OME) is addressed, which emits a minimum of pollutants due to the integration of modern exhaust after-treatment systems. The potentials for this concept in hybrid powertrains applications for the high market share volumes C and J have been demonstrated in [52]. Based on testbed measured, steady operating points, extrapolated in a full vehicle simulation for a hybrid power-split topology, the weighted average, minimum potential for NO_x emissions was determined at 2.3 mg/km for the urban share of RDE-valid trips. This value is assumed to mark the lowest achievable emission potential for the subsequent scenario analysis and is referred to as future emission potential (FEP).

5 Immission assessment

On the basis of the developed empirical immission model (neural networks) in combination with the attributed emission factors based on HBEFA, it is now possible to investigate the immission impact for different scenarios in emitter changes. All subsequent considerations are based on the reference year 2018, as it is the last year the model is explicitly trained for. In order to quantify just the traffic-related immission load, the background immission level is kept constant at the measured yearly average for the year 2018. In this way,

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[1] All data retrieved from [40] except from Volkswagen AG, who publish their data individually via [42.].

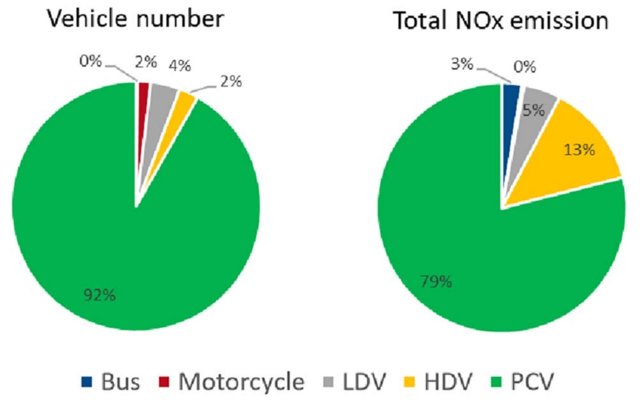

Fig. 7 Allocation of total vehicle number and total fleet emission to vehicle categories in the respective baseline scenario for the year 2018

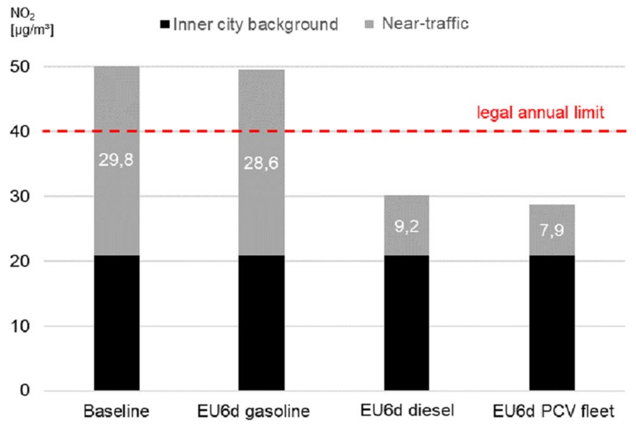

Fig. 8 Immission reduction potential for EU6d passenger car vehicles

the upper boundary of the total immission load is determined, as potential background reductions by diminished traffic emissions are neglected. The inner-city background immission, compromising of the rural background and the additional city background, adds up to 20.9 µg/m^3 ppb NO_2. In combination with traffic emissions, total immission in close street proximity at Darmstadt Hügelstraße station averages to 50.7 µg/m^3 NO_2 in 2018.

The selected, near-traffic, inner-city scenario is characterized by a high proportion of passenger cars vehicles (PCV) in the entire vehicle fleet. As depicted in the left diagram of Fig. 7, 92% of the vehicles are PCV, of which 49% are operated with gasoline and 51% with diesel. The remaining shares, sorted by size, are divided into light-duty vehicles (LDV), heavy-duty vehicles (HDV), buses and motorcycles. Based on the total vehicle number, the greatest reduction seems to be attributable to the PCV segment. In total, more than 7.6 million vehicles pass the measurement station during the year.

However, the comparison of total emission shares (right diagram of Fig. 7) already hints to the fact, that the interaction of both, emission factor and number of emitters has to be regarded simultaneously, in order to account for their total impact. For instance, with a 2% share of all vehicles, the HDV cause 13% of the total NO_x emission. The total emission presented here is based on an annual average where the vehicle fleet emits 199,000 mg/km NO_x on average for every half our sample (added up) which is equivalent to 420 mg/km per vehicle.

The following scenario figures all refer to the reference year 2018. Derived from it is the associated inner-city NO_2 background, which is considered constant in all scenarios at 20.9 µg/m^3 or 26 ppb respectively (black bars). The additional pollution caused by traffic is always indicated in grey. With their high proportion in traffic composition and proven high NO_x emissions in real driving operation, diesel passenger cars are regarded as the main source of inner-city NO_x emissions [37]. In order to classify the potential of the latest technology available on the market, the first step is to model how the additional pollution will develop if only cars with the EU6d emission standard are in use.

As it could be expected, there is a significant reduction potential for diesel passenger cars due to the high NO_x emissions to date, while there is hardly any improvement for gasoline passenger cars, see bar chart Fig. 8. In total, a reduction of 73% in additional pollution (41% total pollution) is achieved solely through improvements in the PCV sector. Prospectively, this improvement will be achieved through natural fleet replacement and without any further measures. For further immission reduction, it may seem consequent to follow the chosen path and aim for even lower emissions in PCV segment. Keeping in mind the previously mentioned interaction of vehicle number and the EFA, there obviously exists a tipping point where improvements in one vehicle category's EFA have only limited impact on air quality when even a small number of vehicles can account for large contributions in total emission due to their high EFA.

This phenomenon can be observed in Fig. 9, where the immission from PCV for EU6d and long-term technology (FEP) is compared. The resulting reduction of 38% (83% to baseline) falls short to the PCV EFA improvement (−95%) due to a masking effect by other vehicle categories, mainly HDV. With significantly lower emissions for PCV, other emitters become dominant and supersede the specific improvement. With just 2% of vehicle share, HDVs would then contribute almost 50% to the total emission. Hence, it can be concluded that focusing on a single emitter category of the vehicle fleet is only feasible within certain limits. For this reason, EFAs according to the latest European emission limits for all vehicle categories have furthermore been considered under EU6d vehicle fleet in the comparison shown in Fig. 10. This adds up to a reduction of 94% against the

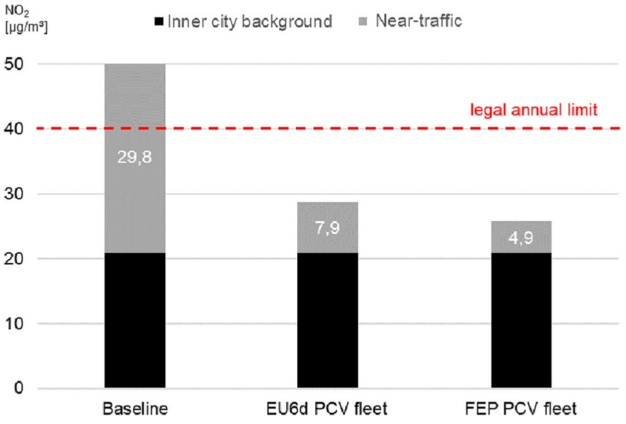

Fig. 9 Immission reduction potential with a focus on the passenger car vehicle fleet for present (EU6d) and future emission potential technology (FEP)

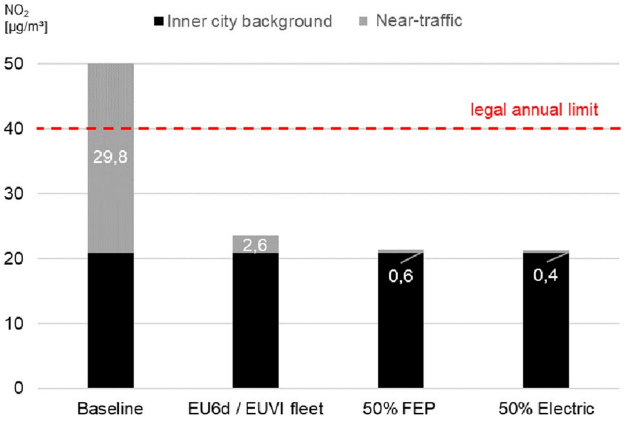

Fig. 10 Immission reduction potential with a focus on the entire vehicle fleet for present (EU6d/EUVI) and future emission potential technology (FEP) compared to electrification

baseline in contrast to the 73% reduction achieved through EU6d PCVs only.

Besides these boundary considerations for the passenger car fleet, regarding the overall fleet development, it is expected that battery electric vehicles and future powertrain concepts with e.g. electrified combustion engines operated with (partly synthetic) fuels, such as the presented long-term technology level, will gain significant market shares to also reach the climate goals [38]. Again, as a boundary estimation, the market share for both technologies is assumed to be 50%. As Fig. 10 shows, this would result in a further reduction of 79% (Long-term technology/FEP) respectively 86% (Electric) compared to the EU6d fleet immission impact. Within case EU6d, the most recent emission standard according to HBEFA 3.3 is applied to all vehicle categories, e.g. EU VI for heavy-duty vehicles. The resulting order of magnitude lies within the measurement uncertainty of the model and marks the point, where additional immission load as a consequence of urban traffic becomes neglectable.

On the basis of these results, there is no significant advantage in terms of urban air quality from the use of electric vehicles over FEP combustion engine technology. With the previously dominant effect of negative air quality impacts for CI-engines weakening, other properties of this drive system such as its high efficiency and robustness are coming to the fore. In combination with synthetic, potentially CO_2 free fuel a sustainable propulsion can be realized. Once again it should be noted that the discussed effect of a lowering of the urban background immission level by very low emissions from vehicles was not addressed in the stated scenarios but kept constant (black bars). If considered, the total immission load should therefore fall even shorter. This aspect is part of ongoing activities for the extension and update of the proposed model.

5.1 Estimation of intermediate transformation

Since the previously introduced empirical model approach does not allow for conclusions about the concentrations more closely to the emitters, the concentration in the driving tube is derived through further analysis.

In a series of 20 individual measurements, the concentration in the driving tube was measured by means of a remote sensing vehicle, using the same measurement device as the immission measurement station *Darmstadt Hügelstraße* [53]. The probe is located within the front bumper, close to the fresh air inlet of the engine room and used for the determination of the driving tube concentration. In addition to drive-through measurements, five continuous measurements were carried out with a stationary vehicle in the vicinity of the measuring station. The mean values of these mobile measurements are put in relation to the immission recorded by the measurement station at the same time. This results in an average factor of 17 between the measurement station and the driving tube. Respectively, the further transformation ratio from the driving tube to the average fleet tailpipe emission (based on HBEFA) is calculated at 565.

Since the classification of emitters, based on their immission impact, demands for a conversion of distance-specific emissions into concentrations directly at tailpipe, an estimate based on previous work and real driving emission (RDE) measurements under urban conditions is derived. According to [54, 55] a ratio of 35 $\frac{\text{ppm}}{\text{g/km}}$ NO_x is obtained for vehicles operated stationary and in load changes. However, transients in full real driving operation lead to significantly higher ratios. Within 379 RDE measurements with 10 different vehicles (EU6) conducted by TU Darmstadt, this ratio ranges from 163 to 434 $\frac{\text{ppm}}{\text{g/km}}$ for the urban share of all trips.

Table 3 Immission impact vehicle categories

Zero Impact	Emission of the vehicle fleet has no influence on the immission measurement result in specific measurement scenario
Levelling	Emission of the vehicle fleet reduces the immission result in specific measurement scenario in and near the driving tube
SubZero	Emission of the vehicle fleet reduces the immission result in specific measurement scenarios and also reduces the background level

According to the corresponding mean value of 309 $\frac{\text{ppm}}{\text{g/km}}$, the ratio in urban real driving operation is from here on considered at 300 $\frac{\text{ppm}}{\text{g/km}}$.

5.2 Derivation of impact categories

Based on the described challenges regarding urban air quality, the aim of future measures must be to keep the emission-related exposure on the population as low as possible. In combination with the described methodology of emission-immission interaction, it becomes possible to categorize emitting vehicles, based on their potential immission impact contribution. The derived definition of vehicle categories is shown in Table 3. The impact is assessed in the driving tube, which means in a distance much closer to the vehicle tailpipe than the probe position of air quality surveillance stations and closer than average pedestrians come to the vehicles. Hence, the transformation from tailpipe to the driving tube has to be considered. The guiding idea behind the definition is best explained within an exemplary, fictious vehicle fleet for each kind:

Zero impact vehicles have no influence on the driving tube concentration in the scenario, e.g. if all vehicles consist of this type, no significant immission is attributable to traffic. Supposedly, concentrations are somewhere in the range of the natural fluctuation of the rural immission background. Beyond this point, vehicles do reduce the prevailing immission load as their emission in combination with dilutive effects and chemical reactions result in lower concentrations than the prevailing conditions. In the first stage, this applies to levelling vehicles, which smoothen the immission load of high emitters but cannot account for a significant, overall reduction. In contrast, SubZero vehicles would reach such low emission values, that they are able to even reduce the prevailing background immission in the specific scenario.

Since the specific boundary conditions of a measurement scenario do influence the interaction, this aspect is part of the definition as well. Further, with the ambient concentration being another variable part of the definition, the classification is not fixed. That means, a vehicle defined as SubZero in the scenario *Darmstadt Hügelstraße* could be a polluter in the countryside. However, the classification within Hot-Spot-scenarios, which are responsible for the majority of immission limit violations, seems a reasonable approach to assess emission technology potentials. The range of corresponding, distance-specific emission values to the definition given in Table 3 under consideration of the previously described intermediate transformation from tailpipe to driving tube, is illustrated in the scale of Fig. 11. For comparison reasons included are the present European emission standards limit values for EU6d. Since the quantitative determination of the stated emission values is based on the immission measurement data, which is associated with relatively wide dispersion and furthermore dependent on the scattering, approximated intermediate transformation factor, the categories are interpreted as ranges.

According to the scale of Fig. 11, an average emission of 21 mg/km NO_x would lead to a driving tube concentration in the range of the inner-city background level at 21 µg/m^3 (Zero impact, equiv. to approximately 11 ppb), which results in a measured value at the station of approximately 1 µg/µ^3 (≈0.5 ppb). A driving tube concentration of half the prevailing background immission load (levelling) is reached at an emission level of 10 mg/km. Below the levelling range then starts the SubZero range at 5 mg/km NO_x emission which leads to an average driving tube concentration of approximately 3 µg/m^3 (≈ 1.6 ppb). Further reaction and dilution to the measurement station result in values significantly smaller than 1 µg/m^3. The derived values correspond to other

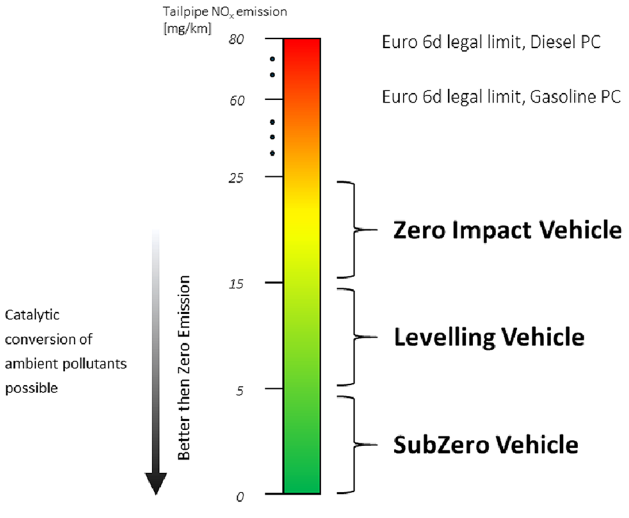

Fig. 11 Proposed classification of low emission vehicles and corresponding specific tailpipe emission ranges based upon their immission impact, valid for the Darmstadt Hügelstraße scenario

investigations of NO_2 immission load as a result of vehicle emissions such as [58–58].

Based on this scale it becomes possible to evaluate vehicles not only according to their tailpipe emission, but rather their immission contribution in terms of type approval. If a pollutant concentration balance within the driving tube of the given scenario is conducted, zero-emission vehicles have no influence whereas ultra-low emission vehicles have the potential to reduce the prevailing immission load in hot spot scenarios. Thus, among other measures, vehicles of these type bear the potential for contributing to higher urban air quality in the situation with high NO_2 immission loads.

5.3 Classification of obtained results

On the basis of the derived transformation mechanisms and emission factors, the obtained scenario results can be classified in a final step. With the scenario being part of the categorization, the following classification is only valid for the investigated scenario of *Darmstadt Hügelstraße*.

A EU6d-Temp passenger car fleet emits 44 mg/km NO_x in average per vehicle. This distance-specific emission converts to approximately 13.200 ppb at Tailpipe. Based on the earlier introduced intermediate transformation, the corresponding driving tube concentration ranges around 23 ppb. Further transformation to the measurement station results in an immission load from this vehicle class of 1.4 ppb NO_2, with the rural background in a fluctuation range of 3…5 ppb. The results show that EU6d-Temp passenger cars are able to partly but not entirely reach the range of Zero impact immission defined here. This corresponds roughly to a similar categorization approach of Eichlseder, who defines a contribution of passenger cars to the immission load at highly frequented roads in the order of clean rural background value (here 3.5 $\mu g/m^3 \approx 1.7$ ppb NO_2) as Rural Background Emission Level (= RuBEL) [59]. According to the immission impact scale of Fig. 11, mid-term technologies reach the range of Zero impact and Levelling vehicles, with an associated driving tube immission of 5.1–25.2 ppb (station: 0.3–1.6 ppb). The specific emission of the long-term technologies results in driving tube concentrations of down to 1 ppb (station: 0.1 ppb), whereby even the inner-city background is undercut strongly and the SubZero level is reached.

Within the proposed classification scale in Fig. 11, the contribution to air pollution control at highly polluted locations can also be illustrated. Zero-emission vehicles, e.g. battery electric vehicles, which have no influence on the prevailing pollutant concentrations in terms of local NO_2, are also unable to change them. On the other hand, internal combustion engines with state-of-the-art exhaust gas after-treatment systems are able to reduce the prevailing concentrations through catalytic conversion. However, this is only the case in locations with significant pollution. Conversely, health impairments as a result of high pollution levels are relevant precisely in those situations.

6 Summary

The paper at hand examines the influence of vehicle emissions on the prevailing nitrogen dioxide immission load in near-traffic areas using a local hot-spot station as an example.

For this purpose, an empirical NO_2-emission-immission model is developed, which allows for the determination of the additional pollution load close to traffic on the basis of passing vehicles under consideration of the prevailing environmental conditions with a time resolution of 30 min. The achieved accuracy ($R^2 = 87\%$) by training with measurement data from six consecutive years can be classified as high compared to known empirical modelling approaches and those of physical modelling. Nevertheless, uncertainty remains with regard to special effects such as atmospheric inversion conditions or heavy rainfall. All in all, a high-quality representation of the immission load at the place of observation is achieved.

Scenario analysis on the basis of the introduced model shows significant reductions in NO_2 immission loads for vehicles with the EU6d emission standard, especially as a result of the significant improvements for CI-engines. Moreover, medium- and long-term technologies offer further potential for reducing additional traffic-related pollution to a level comparable to that in the case of widespread fleet electrification. In all the scenarios considered, the currently valid annual mean limit value of 40 $\mu g/m^3$ will be undercut strongly without considering expected improvements in the prevailing background immission. The findings are finally transferred to a proposed classification scale of vehicle immission impact categories.

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Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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