TECHNISCHE UNIVERSITÄT WIEN
Vienna | Austria

Diplomarbeit

Nitrogen oxides and ammonia in a traffic tunnel – determination of emission factors

Ausgeführt am Institut für
Chemische Technologien und Analytik
der Technischen Universität Wien

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Wien, 02.05.2018

"Remember to look up at the stars and not down at your feet. Try to make sense of what you see and wonder about what makes the universe exist. Be curious. And however difficult life may seem, there is always something you can do and succeed at.

It matters that you don't just give up."

– Stephen Hawking

ACKNOWLEDGMENTS

The present work was carried out within a project conducted by Laboratorium für Umweltanalytik GmbH in cooperation with the Vienna University of Technology, Institute of Chemical Technologies and Analytics. The project was funded by ASFINAG Bau Management GmbH.

At this point, I would like to express my gratitude to the whole team from Laboratorium für Umweltanalytik GmbH, especially to Reinhard Ellinger and Christian Hübner for giving me the chance to prove my skills and to gain experience in working life. Thank you for all the things I learned and for the great time I spent with you.

Moreover, I thank ASFINAG Bau Management GmbH for the financial support of this work.

I would also like to thank sincerely Anne Kasper-Giebl for giving me the opportunity to work in her research group and for always supporting me with her knowledge. Thank you for your interesting inputs and discussions and for answering all my questions with so much patience.

A big thank you to all members of our research group for making my time here so joyful, for always helping me and listening to my problems. Thank you for all the great and funny moments, which enriched my life. Special thanks to Dette Kirchsteiger for pushing me through the last stressful weeks.

My sincerest gratitude goes to my family and my friends. I thank my parents for making my education possible and, together with my sister, my partner and all my friends, for always supporting me. Thank you for your patience and understanding, especially when life was hard and stressful.

ABSTRACT

Vehicular emissions for nitrogen oxides (NO_x) have gained considerable attention because of revelations concerning failures of operation in latest diesel exhaust after-treatment systems in real-word driving conditions. The aim of the presented study is to evaluate real-world emission factors for NO_x for the current Austrian vehicular fleet and to compare them with emission factors provided by the "Handbook Emission Factors for Road Transport" (HBEFA). Emission factor models, i.e. HBEFA, are widely used for a variety of applications, for example emission inventories and environmental impact assessments. The given values for nitrogen oxides are questioned especially for diesel-powered passenger cars with Euro 4, 5 and 6 standards since the diesel scandal became popular. Moreover, the emission factor for ammonia (NH_3) is determined as well because it is used as reducing agent for nitrogen oxides in the selective catalytic reduction system for diesel vehicles. The functionality of this exhaust after-treatment system was revealed to be restricted in real-world operations. Ammonia is yet still a non-regulated compound, however it has influence on ecosystems due to its nitrogen input, leading to acidification and eutrophication. In addition, ammonia plays an important role in the formation of secondary particulate matter, which is associated with adverse health effects.

A measurement campaign from October to December 2017 in tunnel Kaisermühlen was conducted in order to calculate emission factors for nitrogen oxides (NO_x), nitrogen dioxide (NO_2) and ammonia (NH_3). The tunnel is part of an inner-city highway (A22) in Vienna, Austria. The campaign included concentration measurements of nitrogen oxides and ammonia inside as well as outside (background levels) the tunnel. Furthermore, the longitudinal air velocity inside the tunnel was measured and traffic data was provided by ASFINAG.

Daily traffic volume in tunnel Kaisermühlen is 52.000 vehicles in one direction with an average share of heavy-duty vehicles of 5,3%. During workdays passenger cars show a pronounced diurnal profile with peaks during rush hours. The longitudinal air velocity correlates well with traffic density.
Concentrations inside the tunnel for nitrogen oxides (NO_x) and nitrogen dioxide (NO_2) and ammonia (NH_3) are 1170 µg NO_2/m^3 and 226 µg/m³ and 13,5 µg/m³, respectively. Background levels are 36 µg NO_2/m^3, 26 µg/m³ and 1,7 µg/m³ for NO_x, NO_2 and NH_3, respectively.

The average fleet emissions factors are 0,45 g NO_2/vkm for NO_x, 78 mg/vkm for NO_2 and 4,7 mg/vkm for NH_3. The data set for nitrogen oxides and nitrogen dioxide can be divided in two groups according to different traffic conditions occurring in the tunnel Kaisermühlen.

Furthermore, the NO_x and NO_2 emission factors depend strongly on the share of heavy-duty vehicles in the vehicular fleet. Therefore, linear regression analysis delivered separate emission factors for passenger cars and heavy-duty vehicles. The emission factors for ammonia do not correlate with the proportion of heavy-duty vehicles.

The calculated emission factors for nitrogen oxides and nitrogen dioxide agree well with values given in HBEFA for fleet emission factors as well as for emission factors for passenger cars and heavy-duty vehicles separately. Hence, the emission factors provided by HBEFA represents real-world emissions adequately. Regarding emission factors for ammonia, the given value in HBEFA exceeds the calculated factor three times. Since ammonia is non-regulated, the data set for modelling is very meagre.

A comparison with other tunnel studies shows that emission factors for nitrogen oxides are at the same magnitude as in recent investigations found in literature. Former tunnel studies calculated higher emission factors for nitrogen oxides, which is most probably due to another composition of the vehicular fleet and other exhaust after-treatment technologies. Emission factors for ammonia are compared with literature as well. The presented study delivers the lowest emission factor. It has to be noted that the vehicular fleet in other tunnel studies is mainly powered with gasoline, whereas in Austria diesel is the dominant fuel used. Gasoline vehicles emit more ammonia, which is formed over the three-way catalyst, than diesel-powered passenger cars equipped with lean NO_x trap or selective catalytic reduction system with aqueous urea as reducing agent.

Following points sum up and highlight the findings of the presented work:

- A measurement campaign was conducted in the tunnel Kaisermühlen, Vienna, Austria in order to determine real-world emission factors
- Emission factors for nitrogen oxides, nitrogen dioxide and ammonia were determined successfully
- The emission factors for nitrogen oxides and nitrogen dioxide agree well with emission factors provided by the Handbook Emission Factors for Road Transport
- HBEFA represents real-word emissions for nitrogen oxides and nitrogen dioxide
- The emission factor for ammonia is lower than it is given in HBEFA
- Comparison with literature shows comparable results for NO_x emission factors
- Lower emission factors for ammonia are determined compared to other tunnel studies; however, differences of the composition of fuels used has to be kept in mind

KURZFASSUNG

Seit der Aufdeckung des Diesel-Skandals im Jahr 2015 haben die verkehrsbezogenen Emissionen von Stickstoffoxiden (NO_x) enorme Aufmerksamkeit erhalten. Der Skandal betrifft die bewusste Manipulation der Abgasnachbehandlungssysteme von Diesel-Fahrzeugen, die im realen Fahrbetrieb nur eingeschränkt funktionieren. Das Ziel der vorliegenden Arbeit liegt in der Bestimmung realer Stickstoffoxidemissionen der aktuellen österreichischen Verkehrsflotte und dem Vergleich der gefundenen Werte mit jenen aus dem „Handbuch für Emissionsfaktoren des Straßenverkehrs" (HBEFA). Modelle für Emissionsfaktoren, wie eben das HBEFA, finden in einer Reihe von Projekten Anwendung, wie zum Beispiel bei Emissionsinventuren oder im Rahmen von Umweltverträglichkeitsprüfungen. Als Folge des Diesel-Skandals werden speziell die NO_x-Emissionsfaktoren von dieselbetriebenen PKW mit den Emissionsklassen Euro 4, 5 und 6 aus dem HBEFA in Frage gestellt. Zusätzlich werden die Emissionsfaktoren von Ammoniak (NH_3) bestimmt, da Ammoniak als Reduktionsmittel in der selektiven katalytischen Reduktion zur Stickstoffoxidminderung eingesetzt wird. Die Funktionalität eben dieses Abgasnachbehandlungssystems wurde im realen Fahrbetrieb eingeschränkt. Verkehrsbezogene Ammoniakemissionen sind bislang nicht limitiert, dennoch trägt die Deposition von Ammoniak zur Versauerung und Eutrophierung bei und hat somit Einfluss auf Ökosysteme. Weiters spielt Ammoniak als wichtigste basische Komponente in der Atmosphäre eine wichtige Rolle bei der Bildung von sekundären Aerosolpartikeln, welche mit nachteiligen Gesundheitsaspekten assoziiert sind.

Zur Ermittlung der Emissionsfaktoren für Stickstoffoxide (NO_x), Stickstoffdioxid (NO_2) und Ammoniak (NH_3) wurde eine Messkampagne im Zeitraum vom Oktober bis Dezember 2017 im Tunnel Kaisermühlen durchgeführt. Der Tunnel ist Teil der innerörtlichen Autobahn A22 in Wien, Österreich. Im Rahmen der Messkampagne wurden Konzentrationen der Stickstoffoxide und Ammoniak sowohl im Tunnel als auch außerhalb (Hintergrundbelastung) bestimmt. Zusätzlich wurde die Längsgeschwindigkeit im Tunnel gemessen, Daten zum Verkehrsaufkommen wurden von der ASFINAG zur Verfügung gestellt.

Das tägliche Verkehrsaufkommen im Tunnel Kaisermühlen in eine Fahrtrichtung liegt bei 52.000 Fahrzeugen mit einem durchschnittlichen LKW-Anteil von 5,3%. An Werktagen zeigen PKWs Spitzen zu den durch den Berufsverkehr bedingten Stoßzeiten. Die Längsgeschwindigkeit korreliert sehr gut mit der Verkehrsdichte.
Konzentrationswerte im Tunnel liegen bei 1170 µg NO_2/m^3 für NO_x, 226 µg/m^3 für NO_2

und13,5 µg/m³ für NH_3. Die Hintergrundbelastung beträgt 36 µg NO_2/m³ für NO_x, 26 µg/m³ für NO_2 und 1,7 µg/m³ für NH_3.

Der durchschnittliche Flotten-Emissionsfaktor pro Fahrzeug ist 0,45 g NO_2/km für NO_x, 78 mg/km für NO_2 und 4,7 mg/km für NH_3. Für Stickstoffoxide und Stickstoffdioxid kann der Datensatz in zwei unterschiedliche Verkehrszustände eingeteilt werden. Weiters zeigen die Emissionsfaktoren von NO_x und NO_2 eine starke Abhängigkeit vom vorliegenden LKW-Anteil in der Verkehrsflotte. Durch lineare Regressionsanalyse können daher separate Emissionsfaktoren für PKW und LKW berechnet werden. NH_3-Emissionsfaktoren zeigen keine Korrelation mit dem LKW-Anteil.

Die berechneten NO_x- und NO_2-Emissionsfaktoren stimmen sehr gut mit den Werten aus dem HBEFA überein, sowohl für die Flotte als auch für PKW und LKW getrennt. Daraus kann geschlossen werden, dass das HBEFA die realen Emissionen gut abbildet. Der angegebene Emissionsfaktor für Ammoniak ist dreimal so hoch wie der experimentell gefundene. Man muss aber beachten, dass die Datengrundlage für die nicht-limitierte Komponente Ammoniak sehr gering ist.

Ein Vergleich mit anderen Literaturdaten zeigt für Emissionsfaktoren von NO_x und NO_2 gute Übereinstimmungen mit neueren Studien. Ältere Studien hingegen weisen höhere Emissionsfaktoren auf, was auf eine andere Verkehrsflottenzusammensetzung und andere Abgasnachbehandlungssystemen zurückzuführen ist. Ammoniak weist im Vergleich zu anderen Tunnelstudien die geringsten Emissionsfaktoren auf. Allerdings sind die untersuchten Verkehrsflotten nicht vergleichbar, da in dieser Studie vor allem Diesel als Treibstoff eingesetzt wird und in allen anderen der Anteil an Benzin dominiert. Benzinbetriebene PKW mit 3-Wege-Katalysatoren emittieren mehr Ammoniak als dieselbetriebene Fahrzeuge, die mit NO_x-Speicherkatalysatoren und selektiver katalytischer Reduktionssystemen zur NO_x-Emissionsminderung ausgestattet sind.

Folgende Punkten fassen die vorliegende Arbeit zusammen:

- Eine Messkampagne zur Ermittlung realer Emissionsfaktoren wurde im Tunnel Kaisermühlen, Wien, Österreich durchgeführt
- Emissionsfaktoren von Stickstoffoxiden, Stickstoffdioxid und Ammoniak wurden erfolgreich bestimmt
- NO_x- und NO_2-Emissionsfaktoren stimmen gut mit den Werten aus dem Handbuch für Emissionsfaktoren aus dem Straßenverkehr überein
- HBEFA bildet die realen Emissionen für NO_x und NO_2 gut ab
- Der Emissionsfaktor für Ammoniak ist geringer als der angegebene Wert im HBEFA
- NO_x-Emissionsfaktoren sind vergleichbar mit Werten aus der Literatur
- Geringere NH_3-Emissionsfaktoren wurden verglichen mit anderen Tunnelstudien bestimmt, allerdings müssen Unterschiede in der Treibstoffzusammensetzung der Flotte berücksichtigt werden

LIST OF ABBREVIATIONS

EF	Emission factor
ERMES	European Research on Mobile Emission Sources
HBEFA	Handbook Emission Factors for Road Transport
HDV	Heavy-duty vehicle
LNT	Lean NO_x trap
RDE	Real driving emission
PEMS	Portable emissions measurement system
PM	Particulate matter
ppm	Parts per million
SCR	Selective catalytic reduction
SIA	Secondary inorganic aerosol
SOA	Secondary organic aerosol
WLTP	Worldwide Harmonized Light Duty Test Procedure
vkm	Vehicle kilometre

TABLE OF CONTENTS

1 INTRODUCTION

Vehicular transportation of individual persons has become an implicitness in industrialised states all over the world and therefore also in Austria. The number of motorised vehicles, including mopeds, motorcycles, passenger cars, heavy-duty vehicles (HDVs) and buses, has increased strongly during the last decades. [1] Despite the benefits of vehicle motorisation, there is one main disadvantage one has to consider: the usage of fossil fuel based vehicular transportation is an emission source for anthropogenic air pollutants. Today, the reduction of exhaust gas emissions is one major objective the automotive industry has to face, also due to strict legislation.

Regarding the emissions of nitrogen oxides of diesel vehicles, a new reduction system was introduced. Detailed explanation will be presented in another section. Shortly, this exhaust after-treatment is based on the selective catalytic reduction (SCR) of nitrogen oxides with ammonia, as it is well established in power plants. The reducing agent is an aqueous solution of urea – better known under the brand name "AdBlue" –, which is injected into the hot exhaust gas stream, where thermal decomposition of urea takes places yielding the needed ammonia.

In 2013 an investigation conducted in the United States revealed a discrepancy concerning the emission of nitrogen oxides between tests on a chassis dynamometer and measurements under real driving conditions. After some more research was made, manufacturer "Volkswagen AG" was convicted of manipulate the above-mentioned SCR system using a defeat device. This defeat device is able to recognize whether the vehicle is on a chassis dynamometer or not. During the approval procedure on the chassis dynamometer, the urea reduction system is normally operated. So, the tested vehicle or motor type complies with the legal thresholds for nitrogen oxides. In real-world operation the urea-SCR runs with limited functionality, resulting in much higher nitrogen oxides emissions. [2]

As a consequence of this so-called "diesel scandal", the emission factors for nitrogen oxides provided by the "Handbook Emission Factors for Road Transport" (HBEFA), which are used for example in environmental impact assessments, are questioned. It is alleged that the listed values in HBEFA for nitrogen oxides are too low. Consequently, this would lead to an underestimation of the additional pollution within an impact analysis, for example, a road construction project.

The aim of the present master thesis was to evaluate the emission factors for nitrogen oxides for the current Austrian vehicular fleet by conducting a measurement campaign. Afterwards, the results were compared with the values provided by the "Handbook Emissions Factors for Road Transport" and other literature.
In addition, the emission factors for ammonia were determined. Ammonia is used as reducing agent for nitrogen oxides in the urea-SCR technology and is therefore a possible secondary air pollutant. Although ambient air concentrations of ammonia are usually well below any values indicating a harm for human health (e.g. the MAK value is 20 ppm NH_3 on daily average [3]), ammonia plays an important role in the formation of secondary particulate matter and has influence on ecosystems due to its nitrogen input. Presently no ambient oar limit values for ammonia exist.

This work covered practical tasks, data analysis and literature research. Planning and conducting the measurement campaign as well as sample analysis in the laboratory were the main parts of this master thesis. Data analysis and comparison of the emission factors found experimentally with values given in HBEFA and other literature made up another important task of the presented work.

2 BACKGROUND INFORMATION

2.1 The Austrian vehicular fleet

The Austrian vehicular fleet has increased massively during the last decades, also the mode of driving for passenger cars has changed as can be seen in Figure 01(a) and Figure 01(b), respectively.

Development of the Austrian vehicular fleet since 1960

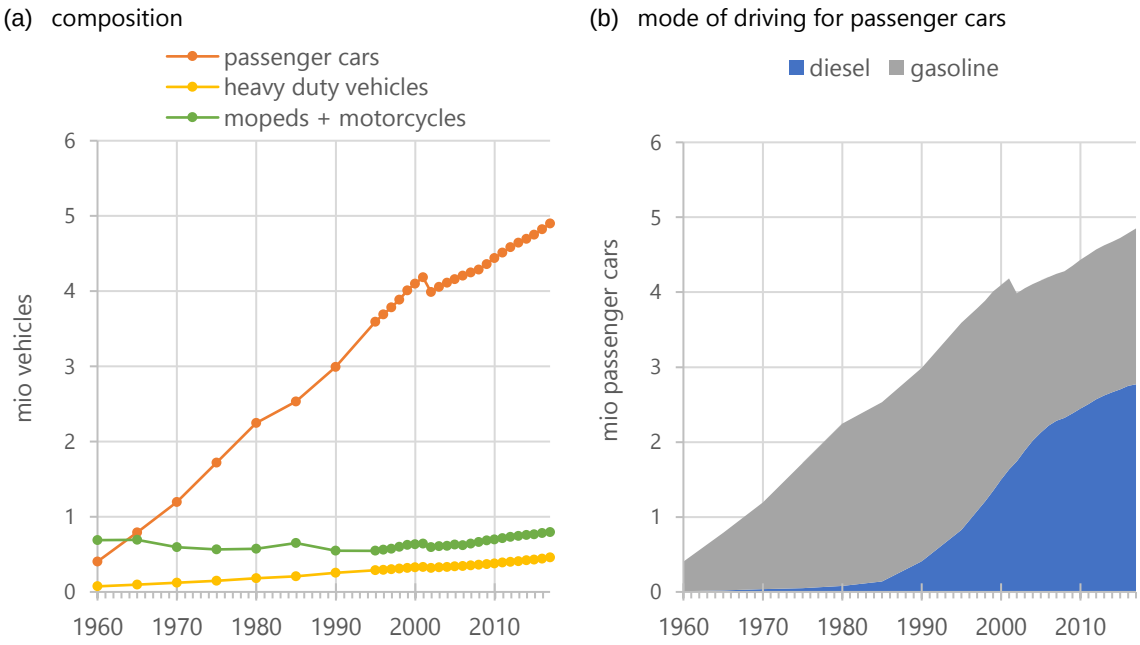


Figure 01: Development of the Austrian vehicular fleet, divided into (a) class of vehicles and (b) mode of driving for passenger cars [1]

The number of passenger cars in Austria has increased more than ten times since 1960. At first, they were driven with gasoline, since 1985 the proportion of diesel-powered vehicles raised rapidly and from 2005 on, diesel constitutes more than 50% of all driving modes (Figure 01(b)).

The composition of the Austrian vehicular fleet in 2017 is shown in Figure 02(a). Passenger cars constitute 72% of all vehicle classes, motorcycles and HDVs come second and third with 8% and 7%, respectively. Passenger cars are mainly powered with diesel (57%) and gasoline (42%). Other modes of driving, like electric power, liquified petroleum gas (LPG), neutral gas, hydrogen or hybrid technologies, comprise only 1% (Figure 02(b)).

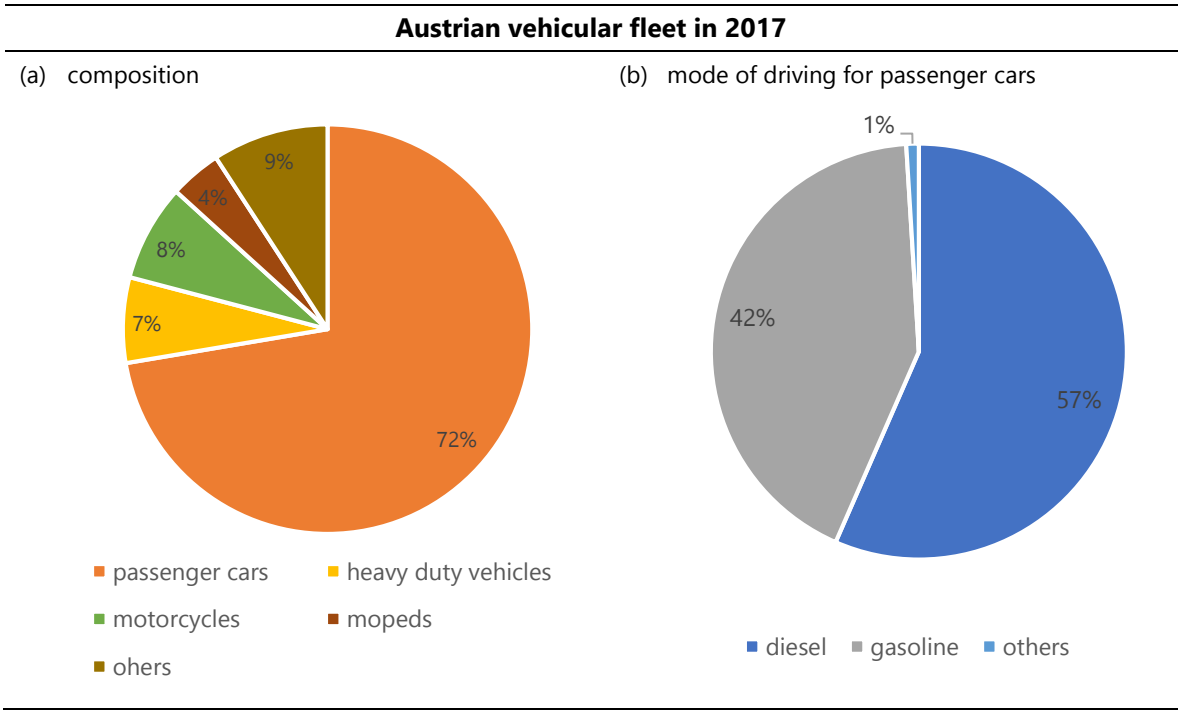


Figure 02: Austrian vehicular fleet in 2017, divided into (a) class of vehicles and (b) mode of driving for passenger cars [1]

2.2 Vehicular exhaust gas emissions

With the motorisation of vehicles, a new anthropogenic emission source for air pollutants has emerged. During the combustion of fossil fuels many air pollutants are generated, which can be divided into two groups, namely primary and secondary pollutants. Primary pollutants are emitted directly out of the exhaust pipe, such as gaseous compounds nitrogen oxides (NO_x), carbon monoxide (CO) and carbon dioxide (CO_2), as well as particulate matter (PM). Secondary pollutants are formed from the primary pollutants through chemical reactions in the atmosphere, for example ozone (O_3) and secondary particles. Other exhaust gas pollutants are hydrocarbons (HC) and volatile organic compounds (VOCs). [4] [5] This VOCs are precursors for secondary organic aerosols (SOA). [6] Secondary inorganic aerosols (SIA), i.e. ammonium sulphate ($(NH_4)_2SO_4$) and ammonium

nitrate (NH_4NO_3) are formed via neutralization reactions with atmospheric ammonia (NH_3), the most abundant basic compound in the atmosphere. [7]

Many studies are related to exhaust gas emissions of fossil fuel-driven vehicles, i.e. vehicles powered with gasoline and diesel, and their impact on human health and environment. Both short-term and long-term exposure lead to adverse health effects. Immediate symptoms are, for example, cough, headache, nausea and irritation of eyes. Diseases concerning the respiratory system, including lung cancer and asthma, are a consequence of long-term exposure to air pollutants. [5]

Regarding exhaust emissions from diesel-driven vehicles, nitrogen oxides are of great concern. NO_x emissions result from the fixation of nitrogen in air during combustion at high temperatures. Many studies have revealed a relation between increased NO_x concentrations and adverse health effects, such as increased asthma incidence [8], asthma exacerbations [9] and deficits in lung function growth [10], especially when it comes to children.

The NO_x emissions in Austria are declining since 1990. The main reason for this are reduced emissions from the transportation sector due to progress in automotive technologies, for example exhaust gas after-treatment systems. In 2015, the transportation sector was responsible for 51% of all nitrogen oxides emissions, from which 52% are contributed to diesel-powered HDVs. [11]
The NO_x concentrations also show a declining trend all over Austria. Concentrations observed near traffic related sites are 9% lower in 2015 compared to 1998. However, the reduction of NO_x emissions is more pronounced (- 26% from 1998 to 2015). [12]

2.3 Reduction technologies for vehicular NO_x

The reduction of the air pollutant NO_x is today of great importance due to its adverse health effects. The legislation in Austria and in the European Union in general has become more and more strict. Therefore, various exhaust gas after-treatment systems for vehicles running on fossil fuels have been invented. These technologies are based on catalytic converters and are presented in the following sections.

2.3.1 Reduction technology for gasoline-powered vehicles

The first catalytic converters for gasoline-driven vehicles were introduced in 1975. They were so-called two-way catalysts, because of their ability of oxidizing CO and unburned hydrocarbons (HC) into the non-harmful compounds CO_2 and water vapour. Five years later, in 1980, these converters were enhanced with the possibility to reduce NO_x into non-toxic gaseous nitrogen (N_2). This three-way catalyst (TWC) is composed of a ceramic block

covered with a thin film of platinum group metals, all embedded in a stainless steel container. The TWC works properly at exhaust gas temperatures of about 350°C. Beneath this temperature neither the oxidation nor the reduction occurs in a sufficient way. In addition, the air-fuel ratio λ is another parameter influencing the conversion efficiency of the TWC. The highest conversion efficiencies for all three compounds are achieved at the stoichiometric point λ = 1. [13]
Studying the selectivity of the reduction of nitrogen oxides revealed the TWC as source for secondary ammonia (NH_3) emissions. The emissions of NO and NH_3 were found to be anticorrelated. The highest concentrations of ammonia were observed during fuel-rich combustion conditions (λ < 1). [14]

2.3.2 Reduction technologies for diesel-powered vehicles

Unfortunately, the well-established TWC is not suitable with diesel-driven vehicles due to the lean combustion conditions (λ > 1) where excess oxygen is contained in the exhaust gas. Therefore, other reduction mechanisms have to be applied.

Lean NO_x trap (LNT)
The LNT has two operating modes. In the first step, NO_x is oxidised on precious metals and bonded to storage compounds with basic properties (mainly Ba) in form of nitrates. The NO_x storage only occurs under oxidising conditions. In the second step, the stored NO_x is reduced. At first, the formed nitrates are decomposed into NO_x under stoichiometric and reducing conditions. Afterwards, the emitted NO_x is reduced into N_2 on precious metals. HC, CO and H_2, which are formed under fatty conditions (λ < 1), act as reducing agents. After this regeneration step, the LNT is fit for the next cycle of storing and reducing NO_x. [15]
Studying the selectivity of NO_x reduction showed an increased selectivity to ammonia with increasing H_2-NO_x ratio as well as increasing regeneration time. [16]

Selective Catalytic Reduction with urea (urea-SCR)
A common process in reducing NO_x emissions in power plants is the selective catalytic reduction (SCR) with ammonia, but for small installations, i.e. motor vehicles, there are problems with storage and safety of this toxic compound. As an alternative reducing agent urea ($CO(NH_2)_2$) is supposed because of its non-toxicity and its easy handling. [17]

The commercial available aqueous urea solution for SCR systems has a concentration of 32,5 wt.% and is better known under the brand name “AdBlue”. “AdBlue” is sprayed into the hot exhaust gas stream in front of the catalytic unit. The mixture is thermally decomposed yielding ammonia and isocyanic acid, the latter being hydrolysed on the catalysts' surface, forming additional ammonia. Finally, ammonia serves as the actual NO_x reducing agent. The chemical reactions of ammonia formation and NO_x reduction are given in equations (1) to (6): [18]

Formation of ammonia		
1. Evaporation of water:	$CO(NH_2)_{2\,(aq)} \rightarrow CO(NH_2)_{2\,(s)}$	(1)
2. Thermolysis of urea:	$CO(NH_2)_{2\,(s)} \rightarrow NH_{3\,(g)} + HNCO_{(g)}$	(2)
3. Hydrolysis of isocyanic acid:	$HNCO_{(g)} + H_2O_{(g)} \rightarrow NH_{3\,(g)} + CO_{2\,(g)}$	(3)
Selective catalytic reduction of NO_x (gaseous phase):		
	$4\,NH_3 + 4\,NO + O_2 \rightarrow 4\,N_2 + 6\,H_2O$	(4)
	$NO + NO_2 + 2\,NH_3 \rightarrow 2\,N_2 + 3\,H_2O$	(5)
	$4\,NH_3 + 2\,NO_2 + O_2 \rightarrow 3\,N_2 + 6\,H_2O$	(6)

The efficiency of urea pyrolysis (equation (2)) is dependent on the exhaust gas temperature. The hotter the exhaust gas stream, the higher the thermal decomposition of urea leading to a maximum of ammonia formation. The high reactivity of isocyanic acid may leads to undesired products: the addition of isocyanic acid to urea yields biuret and the trimerization of isocyanic acid leads to cyanuric acid. However, the emission of these high molecular substances is negligible when the temperature of the exhaust gas is high enough. Ammonia slip is observed when the reducing agent is in excess compared to NO_x. [18]

2.3.3 Secondary ammonia emissions caused by NO_x reduction

The above-mentioned catalytic converters are well-established in fossil fuel powered vehicles. Despite their ability of reducing nitrogen oxides in a sufficient way, they all serve as a source for secondary ammonia emissions.
Especially the TWC used in gasoline light duty vehicles is mentioned as the main source for vehicle-related ammonia emissions. Ammonia is formed via reaction of nitrogen monoxide (NO) with hydrogen (H_2), which is formed according to the water-gas shift reaction. The emission amount was found to be higher with lower ambient temperatures. Ammonia formation is further enhanced at fatty combustion conditions ($\lambda < 1$) because of the higher concentrations of CO and H_2. These conditions occur mainly during acceleration actions. [19] Regarding the urea-SCR technology used in diesel-driven passenger cars, excess of urea, low temperatures and catalyst degradation are the main reasons for secondary ammonia emissions. The ammonia emission from vehicles equipped with urea-SCR are lower compared to gasoline-powered vehicles using TWC for NO_x reduction. [20]

Ammonia plays an important role in particle formation and growth in both urban and rural areas. The presence of NH_3 in vehicular exhaust gas enhances particle formation. Moreover, it is observed, when urban plumes reach rural areas, NH_3 from rural sources, i.e. livestock waste and fertilizer application, promote PM formation. [21]
Ammonia also has a negative effect on ecosystems due to its nitrogen input. Elevated

deposition of ammonia can lead to damages and reduction of some plant species. Both wet and dry deposition affect ecosystems. [22]

Nevertheless, ammonia from vehicular exhaust emissions is not yet regulated. In 2015, the transportation sector was responsible for only 2% of all NH_3 emissions in Austria. [11]

2.4 Diesel scandal and its consequences

In 2013 a measurement campaign was conducted by the Center for Alternative Fuels, Engines & Emissions (CAFEE) from West Virginia University. Three diesel-powered vehicles were tested on their real-world emissions over defined routes with different driving conditions using a portable emissions measurement system (PEMS). All three vehicles were equipped with NO_x after-treatment technologies, including one LNT and two urea-SCR systems.
Real-world emissions of nitrogen oxides were observed to exceed the legal threshold for the vehicle equipped with LNT by a factor of 15 – 35 and for one vehicle equipped with urea-SCR by a factor of 5 – 20. For the second vehicle with urea-SCR system, the emissions were at or below the standard. Interestingly, the two high-emitting vehicles complied with the certification cycle on the chassis dynamometer. [2]

Differences in the NO_x emission behaviour between test cycle on a chassis dynamometer and real-world driving conditions are justified to a certain extent because in real-world driving may occur high-performance operations, for example strong acceleration and steep incline, that do not appear in the test cycle. However, it is strongly forbidden to use a so-called defeat device, which is able to recognize whether the vehicle is on a chassis dynamometer undergoing a certification test or not. An intervention into the exhaust gas after-treatment system to comply with the emission limits on the test cycle, whereas malfunction of the exhaust gas after-treatment during real-world driving operations is considered as wilful manipulation. [23]

In 2015 the Volkswagen AG was convicted of using such a defeat device. Thus, explaining the observed discrepancy for NO_x emissions between test cycle and real-world driving conditions by Thompson G. J. et al. (2014) [2], since the two high-emitting vehicles were manufactured by Volkswagen AG. As a consequence of this diesel scandal, which had and still has an immense media presence, the European Union passed a new approval procedure for passenger cars. The new test cycle – called "Worldwide Harmonized Light Duty Test Procedure" (WLTP) – includes real driving emission (RDE) measurements beside the classic dynamometer test. [24] The maximum emission limit for NO_x emitted from passenger cars in the European Union is 80 mg/km. Real driving emissions of nitrogen oxides are permitted to exceed the dynamometer emissions by a factor 2,1 till January 2020, afterwards the allowed factor for exceedance figures 1,5. [23] [25]

2.5 Emission factors

Tailpipe emissions of road traffic can be described by emission factors (EFs), i.e. emitted mass per travelled distance and vehicle. The determination of emission factors can be achieved via chassis dynamometer tests of single vehicles or motor types. The main disadvantage of this simple method is the requirement of many measurements under different conditions for obtaining a certain representativeness.
Another possibility for the determination of emission factors representing an ensemble of vehicles is to conduct tunnel measurements. Such measurements are said to describe the "real-world emissions", though the results of tunnel studies cannot be generalized to other vehicle ensembles and other driving conditions. [26]

In the present master thesis, the emission factors for nitrogen oxides and ammonia for the Austrian vehicular fleet were determined by carrying out a measurement campaign in the tunnel Kaisermühlen. The calculation of emission factors is based on equation (7) used in Staehelin J. et al. (1997). [26]

$$EF_x(t) = \frac{\Delta C_x(t) * u(t) * q * t}{N(t) * d} \quad (7)$$

EF_x	...	Emission factor of compound x [g/vkm]
ΔC_x	...	Difference in concentration of compound x (tunnel – background) [g/m^3]
u	...	Air velocity [m/h]
q	...	Tunnel cross-section [m^2]
t	...	Duration of time interval [h]
N	...	Number of vehicles [-]
d	...	Distance between the measurement sites

For the calculation of emission factors, a few measurements have to be carried out. First of all, the concentration of the investigated compound is needed. In addition, the background levels have to be considered. Furthermore, the longitudinal air velocity through the tunnel has to be determined. Taking the tunnel cross-section into account, the emission mass flux for a certain time interval is obtained.
In a next step, this emission mass flux is divided through the number of vehicles and the travelled distance from tunnel entrance to the measurement point, thus yielding the emission factor in emitted mass per vehicle kilometre [g/vkm].

2.5.1 Handbook Emission Factors for Road Transport (HBEFA)

The "Handbook Emission Factors for Road Transport" (HBEFA) was launched by six Environmental Agencies of Austria, France, Germany, Norway, Sweden and Switzerland. The first version was published in 1995 by ERMES (European Research on Mobile Emission Sources) group. The HBEFA provides emission factors for all vehicle classes on a variety of

traffic situations, including all regulated and some non-limited air pollutants as well as fuel consumption and CO_2.

The diesel affair raised concern on the given emission factors of diesel-powered light-duty vehicles. Thus, leading to an adaption of emission factors for nitrogen oxides for diesel-driven passenger cars with emission standards EURO 4, 5 and 6 in the latest version 3.3, which was published in April 2017. The results are based on recent real-world measurements conducted in different countries. [27]

The values listed in the HBEFA are used for a wide field of applications, including the determination of vehicular emissions within an environmental impact assessment and preparation of the annual Austrian Air Pollution Inventory. [28]

3 EXPERIMENTAL SECTION

3.1 Measurement Sites

The measurement campaign was carried out in the tunnel Kaisermühlen in 1220 Vienna, Austria. The 2.150 m long tunnel is part of the highway A22 and consists of two separate bores with three lanes in each direction. Speed limits are 80 km/h and 60 km/h for passenger cars and HDVs, respectively. There is a special traffic situation due to the "Section Control", which leads to a constant travel with few acceleration and braking actions. Furthermore, the tunnel jet and the ride in other vehicles' slipstream may reduce the driving resistance, thus leading to lower emissions. This is not in accordance with the usual driving pattern on open highways, so the results cannot be generalized. However, tunnel measurements are a well-established method for determining emission factors because they provide emission factors for an entire fleet under real-world driving conditions.

Measurements for the determination of emission factors were conducted inside as well as outside the tunnel (background concentrations).

3.1.1 Tunnel Kaisermühlen

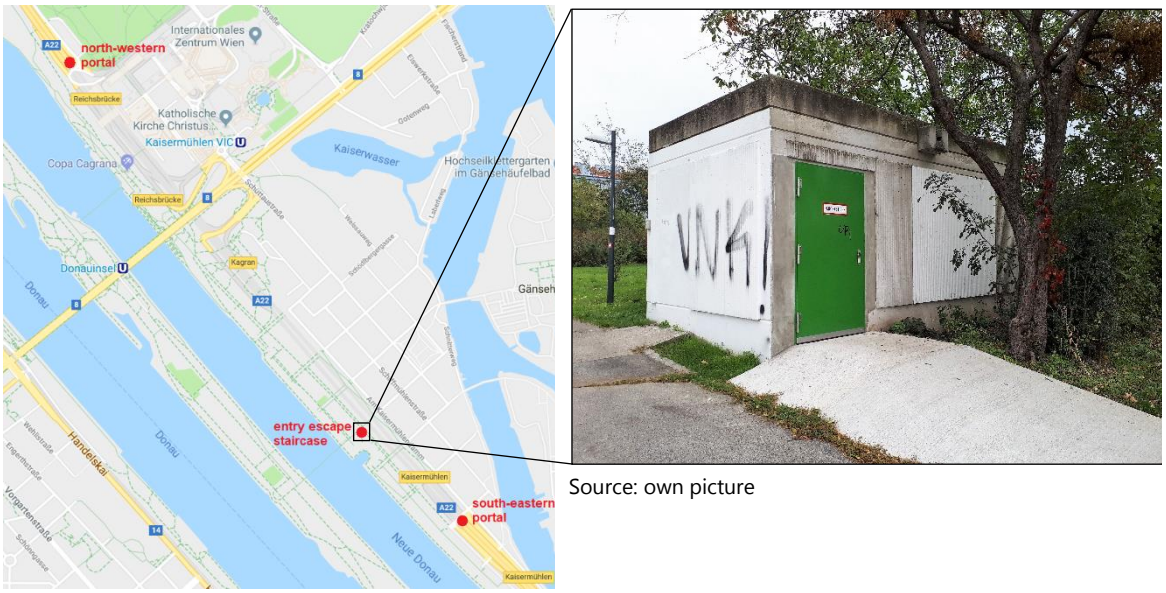
The measurements were conducted in the south-eastern bonded bore of the tunnel Kaisermühlen with the measurement point 1.644 m after the northern tunnel entrance. The measurement instruments were situated in a separate room right behind the tunnel wall. This room was reachable over the escape staircase. Hence, the measurement campaign was feasible without disturbing the tunnel operation and without having any security limitations.

Figure 03 shows the geographical location of tunnel Kaisermühlen with the tunnel portals as well as the entry to the escape staircase marked.

Continuous measurements were performed for nitrogen monoxide (NO) and nitrogen dioxide (NO_2). The concentrations were obtained on half-hour average. The longitudinal air velocity was measured with an impeller anemometer, which was installed inside the tunnel near the south-eastern portal. Furthermore, traffic data, including number and velocity of vehicles, divided into passenger cars and HDVs, were provided by ASFINAG, the Austrian autobahn and highway financing stock corporation.

Discontinuous measurements included the sampling of particulate matter (PM) for the determination of particulate ammonium nitrate (NH_4NO_3) on a daily basis. In addition, gaseous ammonia (NH_3) was sampled on impregnated filters for two or three hours, depending on the time of the day. The analysis of the discontinuously collected samples was performed with suppressed isocratic ion chromatography and took place in the laboratory of TU Wien (Vienna University of Technology, Institute of Chemical Technologies and Analytics).

Geographical location of the tunnel Kaisermühlen with entrance to the escape staircase



Source: Google Maps

Source: own picture

Figure 03: Map of Kaisermühlen in 1220 Vienna, Austria, indicating the tunnel portals and the entrance to the escape staircase

3.1.2 Measurement site for background levels – "Am Kaisermühlendamm"

The measurements of background concentration levels were carried out in "Am Kaisermühlendamm" in 1220 Vienna, Austria. The instruments used for sampling or online measurements were partly also situated in the separate room or in the adjacent staircase. In both cases with the inlet lines leading into the open air.

Analogous to the tunnel measurements, concentrations of nitrogen oxides (NO and NO_2) were recorded as half-hour averages. Filter samples for the determination of particulate ammonium nitrate as well as gaseous ammonia were sampled on a daily basis.

3.2 Measurement methods

The experimental setup is shown in Figure 04. The individual elements are described in detail in the following sections.

Experimental setup

- Nitrogen oxides
  Tunnel
  Background
- Ammonia
  Tunnel
  Background (not shown in this figure)
- Particulate matter
  Tunnel
  Background (not shown in this figure)
- Longitudinal air velocity
  Inside the tunnel (not shown in this figure)

Source: own picture

Figure 04: Experimental setup for the measurement campaign

3.2.1 Continuously measured parameters

3.2.1.1 Nitrogen oxides

An overview of the applied method for the continuous determination of nitrogen oxides (NO and NO_2) is presented in Table 01.

Table 01: Overview of the measurement of nitrogen oxides

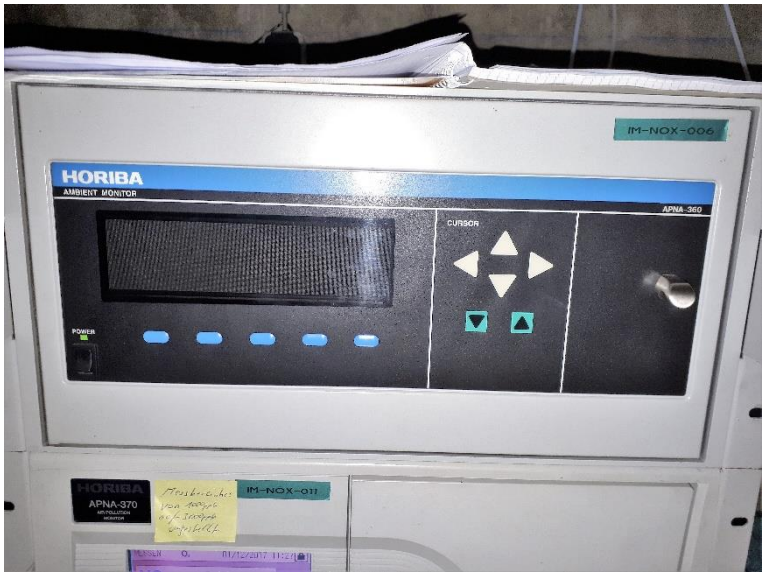
Instrument	Principle	Range	Site
Horiba APNA-360 Ambient Monitor	Chemiluminescence	0 – 1000 ppb	Background
Horiba APNA-370 Air Pollution Monitor	Chemiluminescence	0 – 3000 ppb	Tunnel

For the continuous measurement of nitrogen oxides (NO and NO_2) in the tunnel air as well as in ambient air, two instruments by Horiba with different measurement ranges were used. Both worked on the same principle of chemiluminescence.

The quantification is based on the measurement of the specific light emission of excited nitrogen oxide, which is formed during the reaction between nitrogen monoxide and ozone. The analyser comprises two channels and an ozone generator. In the first channel NO is oxidised with ozone (O_3). In the second channel, NO_2 in the air is reduced over a molybdenum converter previous the reaction chamber, therefore NO_x (sum of NO and NO_2) is measured. The difference of NO_x and NO concentration gives the concentration of NO_2.

Figure 05 shows one of the used analyser for nitrogen oxides.

Horiba APNA-360 Ambient Monitor



Source: own picture

Figure 05: Used instrument APNA-360 Ambient Monitor by Horiba for the measurement of nitrogen oxides

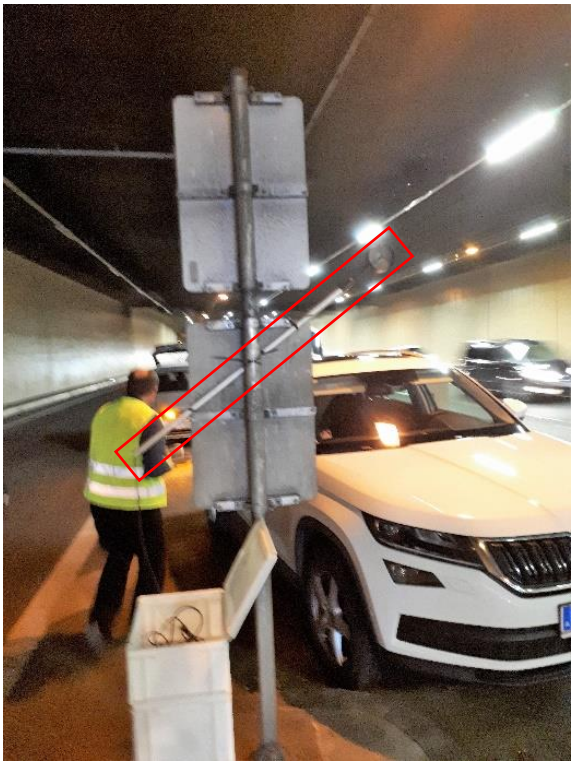
3.2.1.2 Longitudinal air velocity

An impeller anemometer was used for the determination of the longitudinal air velocity inside the tunnel. The measurement instrument was installed in the south-eastern bonded tunnel bore near the exit portal and can be seen in Figure 06.

Within a former investigation, also carried out in the tunnel Kaisermühlen, the longitudinal air velocity inside the tunnel was measured with two methods. On the one hand with a propeller anemometer and via tracer gas measurement on the other hand. For the latter, nitrous oxide (N_2O) was used as tracer compound. The longitudinal air velocity was

calculated from the running time of the tracer and compared with the data of the anemometer. As a result, the longitudinal air velocity obtained by the anemometer was validated through the tracer gas experiment. [29]

Impeller anemometer



(a) [30]

(b) [31]

Source: own picture

Figure 06: Impeller anemometer for determining the longitudinal air velocity inside the tunnel Kaisermühlen

3.2.2 Discontinuously measured parameters

In addition to the continuous measurements, a daily sample of particulate matter (PM) for the determination of particulate ammonium nitrate (NH_4NO_3) was collected onto a Teflon filter from 11 am till 10 am on the following day. Furthermore, gaseous ammonia (NH_3) inside the tunnel was collected with impregnated filters for two or three hours, depending on the time of day. Background levels of ammonia were sampled from 11 am till 10 am on the following day. Cellulose filters impregnated with oxalic acid were used for the collection of ammonia.

Table 02 sums up the sampling and analysis methods for the discontinuously measured parameters.

Table 02: Overview of the sampling and analysis methods for discontinuously measured parameters

Parameter	Sampling method	Analysis method	Site
Particulate ammonium nitrate in PM	Teflon filter Zeflour™ Supported PTFE 1 µm pore size, Ø 47 mm PALL Corporation Life Sciences	Suppressed isocratic ion chromatography	Tunnel and background
Gaseous ammonia	Filter pack with Teflon filter and two cellulose filters coated with oxalic acid, Ø 47 mm Whatman™, GE Healthcare Life Sciences	Suppressed isocratic ion chromatography	Tunnel and background

3.2.2.1 Particulate ammonium nitrate in PM

A filter sample of particulate matter was collected from 11 am to 10 am on the following day for the determination of particulate ammonium nitrate (NH_4NO_3) in tunnel air as well as in ambient air. The purpose of this investigation was to see whether the gaseous vehicular emissions of nitrogen oxides and ammonia lead to enhanced particle formation inside the tunnel.

PM was collected on Teflon filters "Zeflour™ Supported PTFE" produced by PALL Corporation Life Sciences with a flow rate of approximately 8 l/min and 4 l/min for tunnel air and ambient air, respectively.

The analysis of the samples took place in the laboratory with suppressed isocratic ion chromatography and will be discussed in a latter section.

3.2.2.2 Gaseous ammonia

The sampling of gaseous ammonia (NH_3) is based on a chemical reaction with oxalic acid forming ammonium oxalate.

Tunnel air as well as ambient air was sucked over cellulose filters, which are coated with oxalic acid. The preparation of these filters took place in the laboratory. The cellulose filters (produced by Whatman™, GE Healthcare Life Sciences) were impregnated with an aqueous solution containing 3% w/w oxalic acid and 5% w/w glycerol. But beforehand, the filters had to be cleaned. The filters were put into a narrow beaker glass, where they were covered with washing solution and were put in the ultrasonic bath for at least 10 minutes. The procedure included three washing steps with distilled water and one step with diluted coating solution (1:5). The impregnation step itself took also place in the beaker glass with the coating solution covering the cellulose filters and sonification for 10 minutes. The prepared filters were dried in a vacuum desiccator and were stored airtight until usage. [32]

The collection of gaseous ammonia was performed by using a filter pack system. Two impregnated cellulose filters were placed in series. A Teflon filter was put ahead to remove particulate matter avoiding interferences with ammonium salts, i.e. ammonium nitrate. Sampling flow rate was approximately 13 l/min and 4 l/min for tunnel air and ambient air, respectively. Analysis of the formed ammonium oxalate took place in the laboratory with suppressed isocratic ion chromatography and will be discussed in the next section.

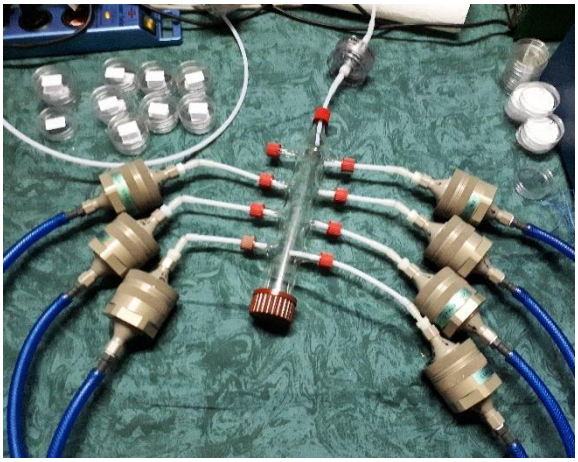
Gaseous ammonia in tunnel air

The collection of gaseous ammonia inside the tunnel took place with a time resolution of two or three hours, depending on the time of the day. Unlike the continuous measurement of nitrogen oxides (half-hour averages), ammonia had to be sampled for a longer period in order to ensure a proper analysis with ion chromatography. Blank values were in the range of 0,05 – 0,74 µg NH_3 per filter. Preliminary investigations indicated a time resolution of two hours during rush hours in the morning and evening and three hours over midday (lower traffic volume) as a good compromise between high time resolution and reliable analysis. Gaseous ammonia was not sampled during night time (22 pm to 6 am) due to the very low traffic volume (see section 5.1).

A special sampling system was used for the collection of gaseous ammonia in tunnel air, which is depicted in Figure 07. Figure 07(a) shows the distribution unit with seven filter packs, each containing two impregnated cellulose filters. The removal of particulate matter occurred in front of the distribution unit with a Teflon filter. The instrument showed in Figure 07(b) regulated the sampling duration of each filter pack with an included time switch. With this sampling system the impregnated cellulose filters had to be changed just once a day. The timetable for one measuring day is given in Table 03.

Sampling system for gaseous ammonia in tunnel air

(a)



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(b)

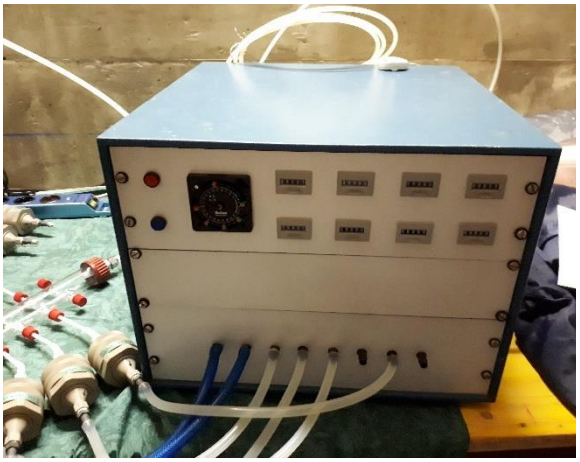

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Figure 07: Sampling system for gaseous ammonia in tunnel air with (a) the distribution unit with filter packs containing the impregnated cellulose filters and (b) the instrument for sequential sampling of the filter packs

Table 03: Timetable of discontinuous sampling of gaseous ammonia for one measurement day

Valve	Filter pack	Sampling time	Comments
1	1	11 am – 2 pm	Measurement start
2	2	2 pm – 4 pm	
3	3	4 pm – 6 pm	
4	4	6 pm – 8 pm	
5	5	8 pm – 10 pm	
6	-	10 pm – 6 am	No sampling during night time
7	6	6 am – 8 am	
8	7	8 am – 10 am	
-	-	10 am – 11 am	Changing cellulose filters

Gaseous ammonia in ambient air

The collection of gaseous ammonia in ambient air was carried out with the same sampling method as it was used for gaseous ammonia in tunnel air. As a difference, only one filter pack was used for sampling from 11 am to 10 pm on the following day, because of the lower concentrations.

3.2.2.3 Suppressed isocratic ion chromatography

For the quantification of sampled ammonium nitrate in particulate matter and gaseous ammonia in form of ammonium oxalate, suppressed isocratic ion chromatography was used. The filters were transported into the laboratory and put into 4 ml polyethylene vials.

After the extraction process – which will be discussed for Teflon filters and impregnated cellulose filters separately – the extracts were transferred into 1,5 ml Eppendorf tubes and analysed with ion chromatography.

Analysis of Teflon filters

Particulate ammonium nitrate contained in PM on the Teflon filters was extracted with 0,2 ml acetone and 3,4 ml deionized water. Acetone was necessary in order to make the hydrophobic Teflon filters wettable. The soaked filters were put in the ultrasonic bath for 30 minutes, afterwards the extracts were ready for analysis. Each filter was analysed for the cation ammonium (NH_4^+) and the anion nitrate (NO_3^-).

The limits of detection for the investigated compounds are listed in Table 04.

Table 04: Limits of detection for ammonium and nitrate in particulate matter

Site	**Ammonium (NH_4^+)**	**Nitrate (NO_3^-)**
Background	0,05 µg/m³	0,14 µg/m³
Tunnel	0,02 µg/m³	0,06 µg/m³

Analysis of impregnated cellulose filters

Gaseous ammonia reacted with the oxalic acid on the cellulose filter to ammonium oxalate. The formed ammonium oxalate on the filters was extracted with 3,6 ml deionized water and put into ultrasonic bath for 30 minutes. Each filter was analysed for the cation ammonium (NH_4^+).

The limit of detection for ammonium on the impregnated cellulose filters within the analysis with suppressed isocratic ion chromatography is 0,50 µg/ml NH_4^+. This means minima concentrations of atmospheric ammonia of 0,14 µg/m³ and 0,35 µg/m³ for tunnel air and ambient air, respectively.

The collection efficiency of the cellulose filters impregnated with oxalic acid for discontinuous ammonia sampling was investigated putting two filters in series. For some tunnel samples (including the measurement days 26.11.2017 – 29.11.2017) both filters were analysed separately. The concentration of formed ammonium oxalate on the second impregnated cellulose filter was found to be below the limit of detection, thus indicating a sufficient collection efficiency of the first filter. Therefore, all the other tunnel samples as well as the samples taken at the background site were analysed with both filters together.

External standards were used for quantification of the investigated compounds. Table 05 lists the characteristics of the suppressed isocratic ion chromatography systems used.

Table 05: Characteristics of used suppressed isocratic ion chromatography systems

Suppressed isocratic ion chromatography		
	Cations	**Anions**
Instrument	Dionex ICS-3000	Dionex ICS 1100
Analytical column	Dionex Ion Pak CS16A	Dionex Ion Pac AS22A
Precolumn	Dinoex Ion Pac CG16A	Dionex Ion Pac AG22A
Eluent	38 mM methane sulfonic acid	4,5 mM sodium carbonate / 1,4 mM sodium hydrogen carbonate
Flow	1 ml/min	1 ml/min
Suppressor	Dionex CRS 500 – 4 mm	Dionex ARS 300 – 4 mm
Regeneration agent	Eluent (circulated)	Eluent (circulated)
Sample loop	150 µl	100 µl
Detection	Conductivity detector	Conductivity detector
Evaluation software	Chromeleon 6.80/7.2.6	Chromeleon 6.80/7.2.6

3.3 Timetable for measurement campaign

Since the measurements were not all carried out at the exact same time period, the following graphic (Figure 08) should give an overview of the timetable for the conducted measurements.

Concentration measurements of nitrogen oxides inside the tunnel were carried out from 23.10.2017 till 11.12.2017, whereas background levels are obtained from 16.11.2017 till 04.12.2017. Regarding the measurement of particle matter for the determination of particulate ammonium nitrate as well as the collection of gaseous ammonia, the concentrations were measured from 24.11.2017 till 04.12.2017, except with the 25.11.2017 where a failure occurred.

The longitudinal air velocity inside the tunnel was measured from 16.11.2017 till 09.12.2017. Traffic data was obtained from 15.11.2017 till 12.12.2017.

Timetable for the measurement campaign

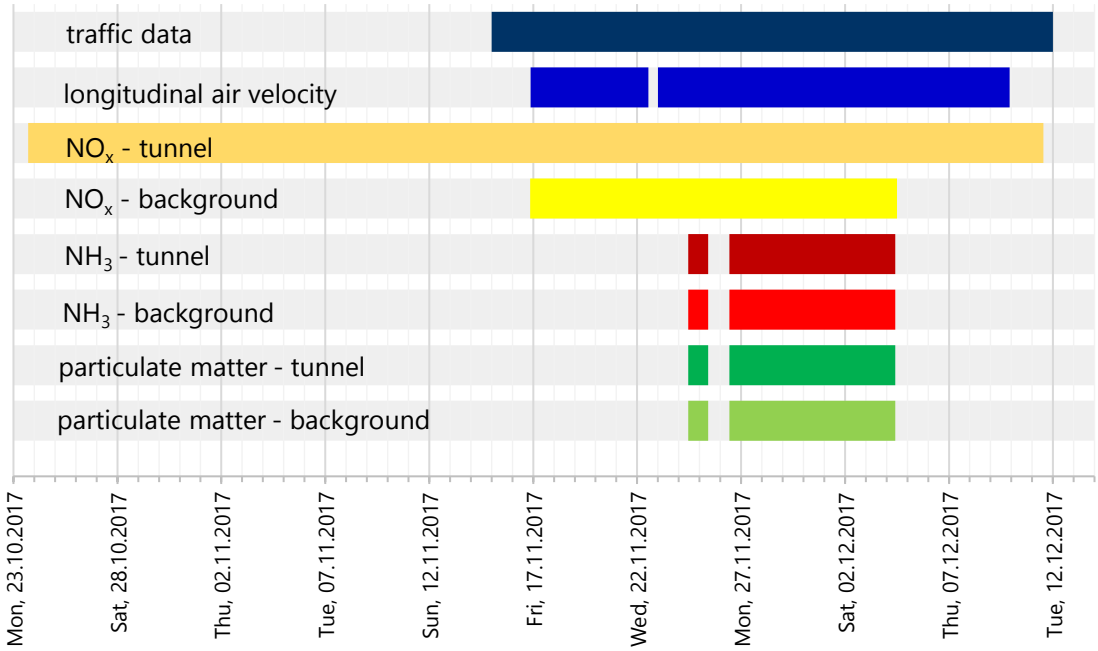


Figure 08: Timetable for the measurement campaign

4 RESULTS

4.1 Traffic volume and fleet composition

Traffic data, including number and speed of vehicles, which could be further divided into passenger cars and heavy-duty vehicles (HDVs), inside the tunnel Kaisermühlen is recorded by an automatic counting station and was provided by ASFINAG (the Austrian autobahn and highway financing stock corporation). Traffic counting is based on measurements with loop detectors, which are embedded in the road surface. It has to be noted, that with this method the differentiation between passenger car and HDV happens according to the length of the vehicle.

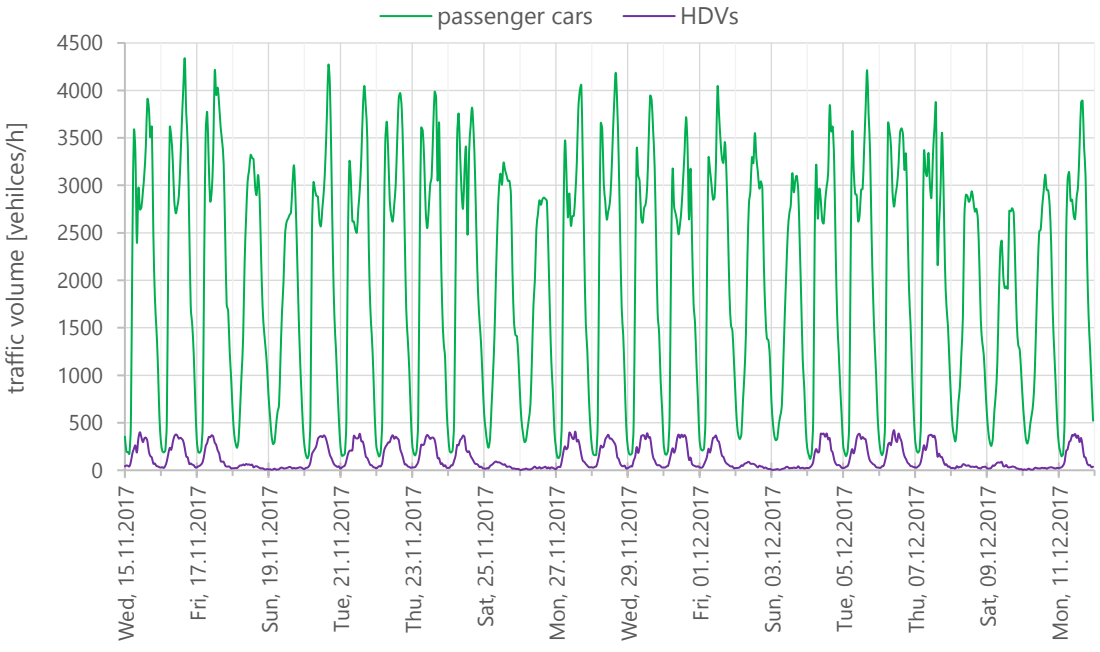


Figure 09: Diurnal trend of traffic volume, differentiated into passenger cars and HDVs, for the south-eastern bonded bore of tunnel Kaisermühlen from 15.11.2017 – 11.12.2017

Figure 09 shows the diurnal trend of the traffic volume observed in the tunnel Kaisermühlen in the period from 15.11.2017 – 12.12.2017. Regarding passenger cars, typical diurnal trends

are observed. During workdays two peaks indicate the rush-hour traffic, whereas on the weekend and on the public holiday (08.12.2017) the total number of passenger cars is lower. For HDVs, the driving ban on weekends [33] (on Saturdays from 3 pm till midnight and on Sundays and public holidays from midnight till 10 pm) as well as the driving ban during night time [34] (every night from 10 pm till 5 am on the next day) is clearly visible.

Average daily traffic volume in south-eastly direction is 52.000 vehicles with an average share of HDVs in the vehicular fleet of 5,3% during the investigated period. Considering workdays only, the HDV portion is 7,0%, whereas the percentage on Saturdays and Sundays – including the public holiday – figures 2,3% and 1,4%, respectively. Between 6 am and 10 pm 90% of the total daily traffic volume occurs. The vehicle number exceeds 1.000 vehicles/h at 5 am, 6 am and 9 pm on workdays, Saturdays and Sundays, respectively.

Table 06 figures the daily vehicle number during the investigated period in the south-eastern bonded bore of tunnel Kaisermühlen.

Table 06: Daily traffic volume in the south-eastern bonded bore of tunnel Kaisermühlen

Date		**Total vehicles**	**Passenger cars**	**HDVs**
Wednesday	15.11.2017	57765	53464	4225
Thursday	16.11.2017	59886	55488	4347
Friday	17.11.2017	61627	57809	3801
Saturday	18.11.2017	46449	45565	884
Sunday	19.11.2017	38090	37623	467
Monday	20.11.2017	55853	51615	4238
Tuesday	21.11.2017	55643	51440	4162
Wednesday	22.11.2017	58164	53827	4302
Thursday	23.11.2017	58269	53904	4365
Friday	24.11.2017	58269	54560	3575
Saturday	25.11.2017	46052	44981	1071
Sunday	26.11.2017	38052	37565	487
Monday	27.11.2017	55911	51497	4370
Tuesday	28.11.2017	57820	53614	4206
Wednesday	29.11.2017	56087	51822	4265
Thursday	30.11.2017	53962	49674	4254
Friday	01.12.2017	58582	54960	3622
Saturday	02.12.2017	47839	46762	1077
Sunday	03.12.2017	39130	38661	469
Monday	04.12.2017	54877	50548	4329
Tuesday	05.12.2017	57256	53019	4194
Wednesday	06.12.2017	58573	54117	4427
Thursday	07.12.2017	58398	54145	4165
Friday	08.12.2017*	42919	42058	861
Saturday	09.12.2017	37983	37007	976
Sunday	10.12.2017	38445	38002	443
Monday	11.12.2017	55634	51144	4490

* Public holiday

4.2 Longitudinal air velocity

The vehicles passing through the tunnel Kaisermühlen generate a tunnel jet, which is normally sufficient for tunnel ventilation. Forced ventilation with axial fans is only performed when high CO values are detected, for example during congestions.

The trend of the longitudinal air velocity inside the south-eastern bonded tunnel bore obtained from an impeller anemometer is shown in Figure 10. In addition, the diurnal trend of traffic volume is depicted.

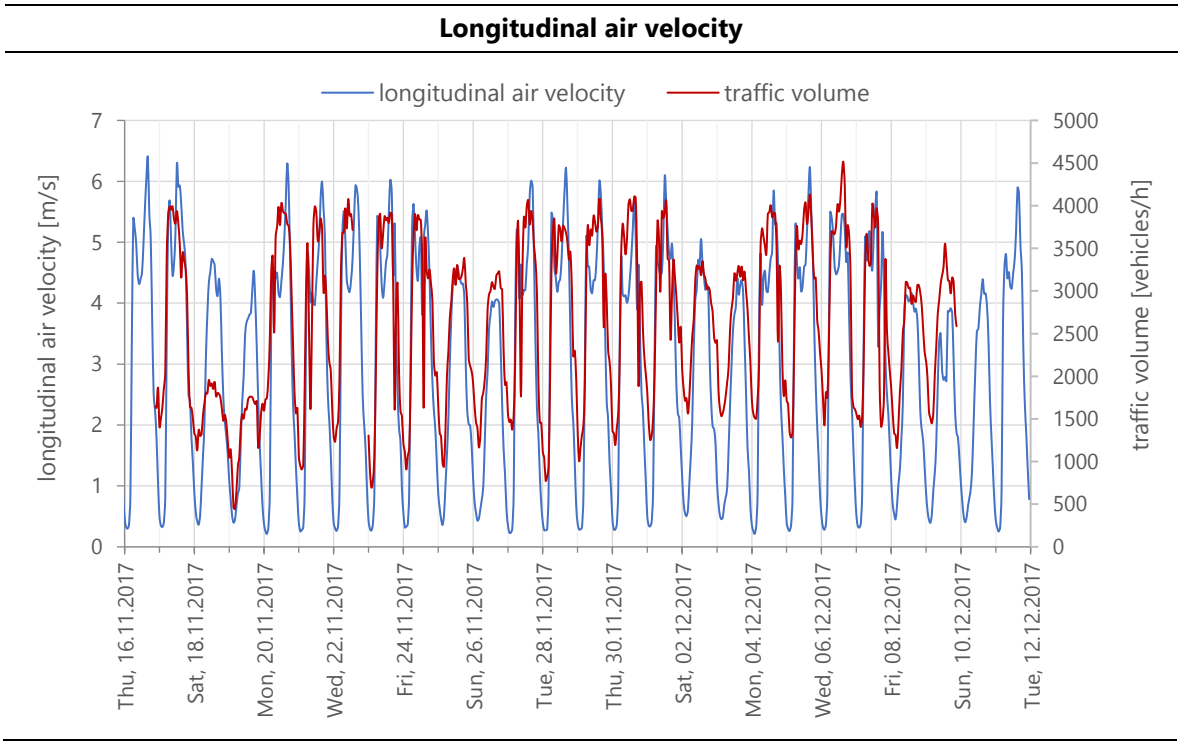


Figure 10: Diurnal trend of longitudinal air velocity for the south-eastern bonded bore of tunnel Kaisermühlen from 16.11.2017 – 09.12.2017 with corresponding traffic volume

The measured longitudinal air velocity shows a good correlation with the corresponding traffic volume. Maximum longitudinal air velocities of approximately 5,5 m/s are reached at traffic peaks according to rush hours during workdays. As with traffic numbers, the air velocity inside the tunnel is lower on the weekend. During night time the longitudinal air velocity reaches a minimum, however, the tunnel jet hardly falls below 1 m/s.

The volume flow inside the tunnel Kaisermühlen is obtained by multiplying the longitudinal air velocity by the tunnel cross-section, which is 117 m². Therefore, the maximum and minimum volume flows are about 2.300.000 m³/h for high traffic numbers and 420.000 m³/h during night time. For the calculation of emission factors, only values where the traffic

density reaches more than 1000 vehicles/h are taken into account (see section 5.1). The volume flow corresponding to 1000 vehicles/h is approximately 670.000 m³/h.

4.3 Concentrations of air pollutants

An overview of the average concentrations inside the tunnel as well as background levels for the investigated pollutants, i.e. nitrogen oxides, ammonia and particulate ammonium nitrate, is given in Table 07. Sulphate concentrations in particulate matter are also given. Data during night time (10 pm – 6 am) are not taken into account due to the very low traffic volume and therefore low longitudinal air velocities (see section 5.1).

Table 07: Average concentrations of measured air pollutants inside the south-bonded bore of tunnel Kaisermühlen and at the background site "Am Kaisermühlendamm" (excluding data during night time 10 pm till 6 am)

Pollutant	Site	Concentration	Period
NO_x	Tunnel	1170 µg NO_2/m³	23.10.2017 – 11.12.2017
	Background	36 µg NO_2/m³	16.11.2017 – 04.12.2017
NO_2	Tunnel	226 µg/m³	23.10.2017 – 11.12.2017
	Background	26 µg/m³	16.11.2017 – 04.12.2017
NH_3	Tunnel	13,5 µg/m³	24.11.2027 – 04.12.2017
	Background	1,7 µg/m³	
Ammonium (NH_4^+) in particulate matter	Tunnel	2,2 µg/m³	24.11.2027 – 04.12.2017
	Background	1,9 µg/m³	
Nitrate (NO_3^-) in particulate matter	Tunnel	5,3 µg/m³	24.11.2027 – 04.12.2017
	Background	4,9 µg/m³	
Sulphate (SO_4^{2-}) in particulate matter	Tunnel	1,8 µg/m³	24.11.2017 – 04.12.2017
	Background	1,5 µg/m³	

4.3.1 Nitrogen oxides

4.3.1.1 Nitrogen oxides inside the tunnel Kaisermühlen

The concentrations of nitrogen oxides (NO, NO_2 and NO_x) inside the tunnel Kaisermühlen for the investigated period from 23.10.2017 – 11.12.2017 are shown in Figure 11.

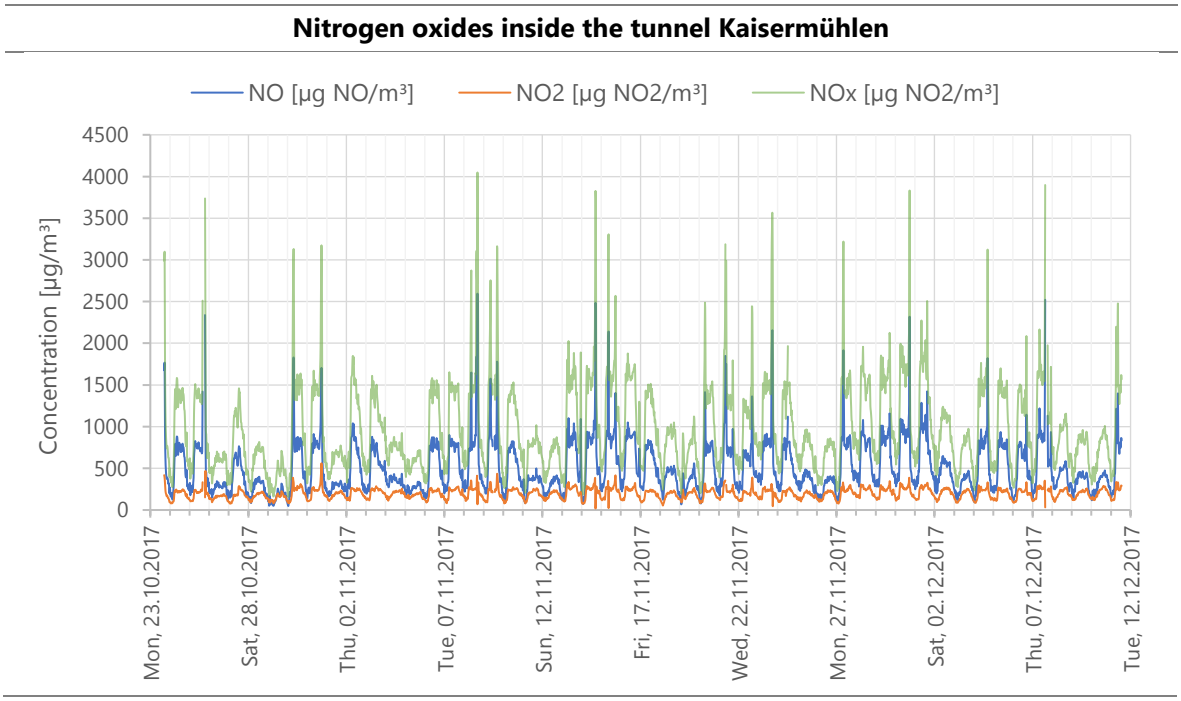


Figure 11: Diurnal trend of concentrations of nitrogen oxides in the south-eastern bonded bore in tunnel Kaisermühlen from 23.10.2017 – 11.12.2017

The concentrations of nitrogen oxides underlie a pronounced diurnal trend and show a good correlation with traffic volume. The average concentrations over the investigated period (23.10.2017 – 11.12.2017, 6 am – 10 pm) are 1170 µg NO_2/m³ and 226 µg/m³ for NO_x and NO_2, respectively. Regarding NO_x, the average concentration during workdays is 1400 µg NO_2/m³. Concentration peaks of 3000 µg NO_2/m³ and even more result due to rush-hour traffic, where it comes to conjunctions and, consequently, lower tunnel jets. NO_x concentrations are clearly lower on the weekend, namely 740 µg NO_2/m³ on average. This means a decrease in concentration. of about 47%. The average proportion of NO_2 to NO_x concentration is 22%, whereby on workdays a higher share in concentration (28%) and on weekend a lower one (18%) is observed.

4.3.1.1 Nitrogen oxides at the background site "Am Kaisermühlendamm"

The concentrations of nitrogen oxides (NO, NO_2 and NO_x) measured at the background site for the investigated period from 16.11.2017 – 04.12.2017 are shown in Figure 12.

The average concentrations during the investigated period (16.11.2017 – 04.12.2017, 6 am – 10 pm) are 36 µg NO_2/m³ and 26 µg/m³ for NO_x and NO_2, respectively. Therefore, the concentrations inside the tunnel exceed background levels 30 times for NO_x and 9 times for NO_2. The average portion of NO_2 to NO_x concentration is generally lager, namely 84%.

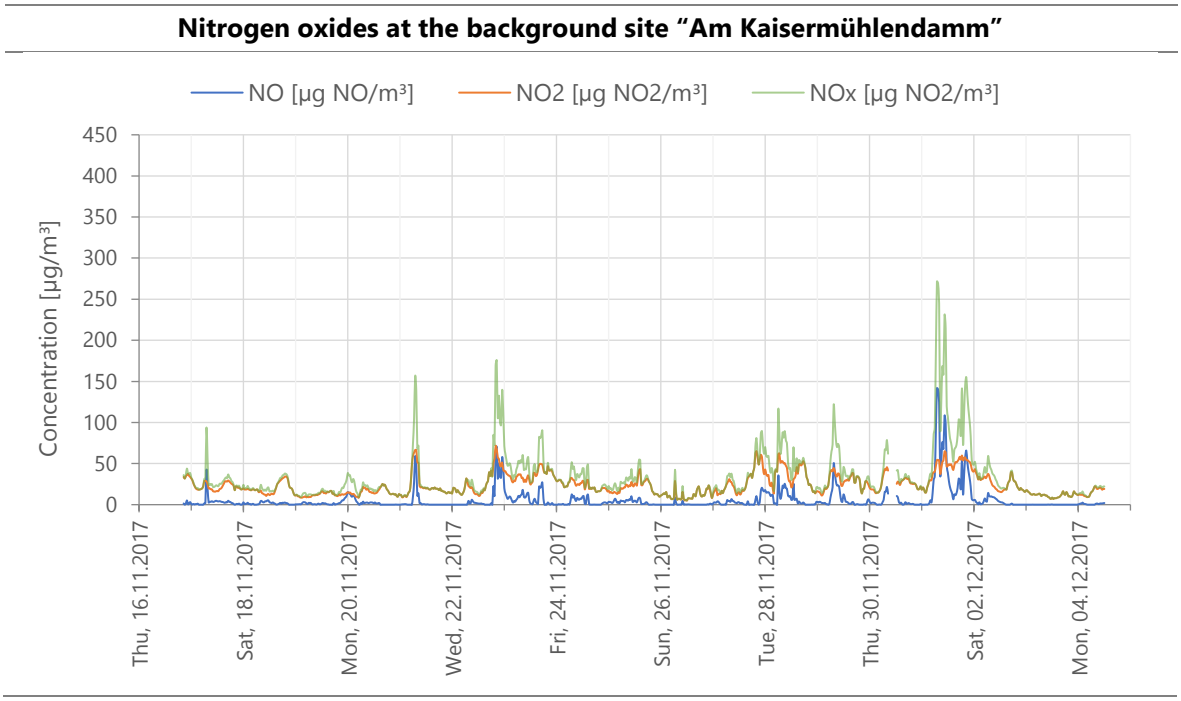


Figure 12: Diurnal trend of concentrations of nitrogen oxides at the background site "Am Kaisermühlendamm" from 16.11.2017 – 04.12.2017

4.3.2 Ammonia

4.3.2.1 Ammonia inside the tunnel Kaisermühlen

The concentration of gaseous ammonia (NH_3) inside the tunnel Kaisermühlen for the investigated period from 24.11.2017 – 04.12.2017 is shown in Figure 14 (page 29). Discontinuous measurements of ammonia were carried out with a time resolution of two and three hours, dependent on the time of day. The average concentration of gaseous ammonia inside the tunnel Kaisermühlen is 13,5 µg/m³. Compared to nitrogen oxides, the difference in concentration between workdays (14,3 µg/m³) and weekend (11,7 µg/m³) is less pronounced for ammonia. A diurnal trend according to traffic volume is not recognisable. However, maximum NH_3 concentrations were observed between 6 pm – 8 pm on some workdays.

4.3.2.2 Ammonia at the background site "Am Kaisermühlendamm"

The concentration of gaseous ammonia (NH_3) measured at the background site for the investigated period from 24.11.217 – 04.12.2017 is shown in Figure 13.

Ammonia in ambient air was collected as daily samples from 11 am till 10 am on the following day. The average concentration is 1,7 µg/m³. Therefore, the concentration inside the tunnel Kaisermühlen exceeds background levels approximately 8 times.

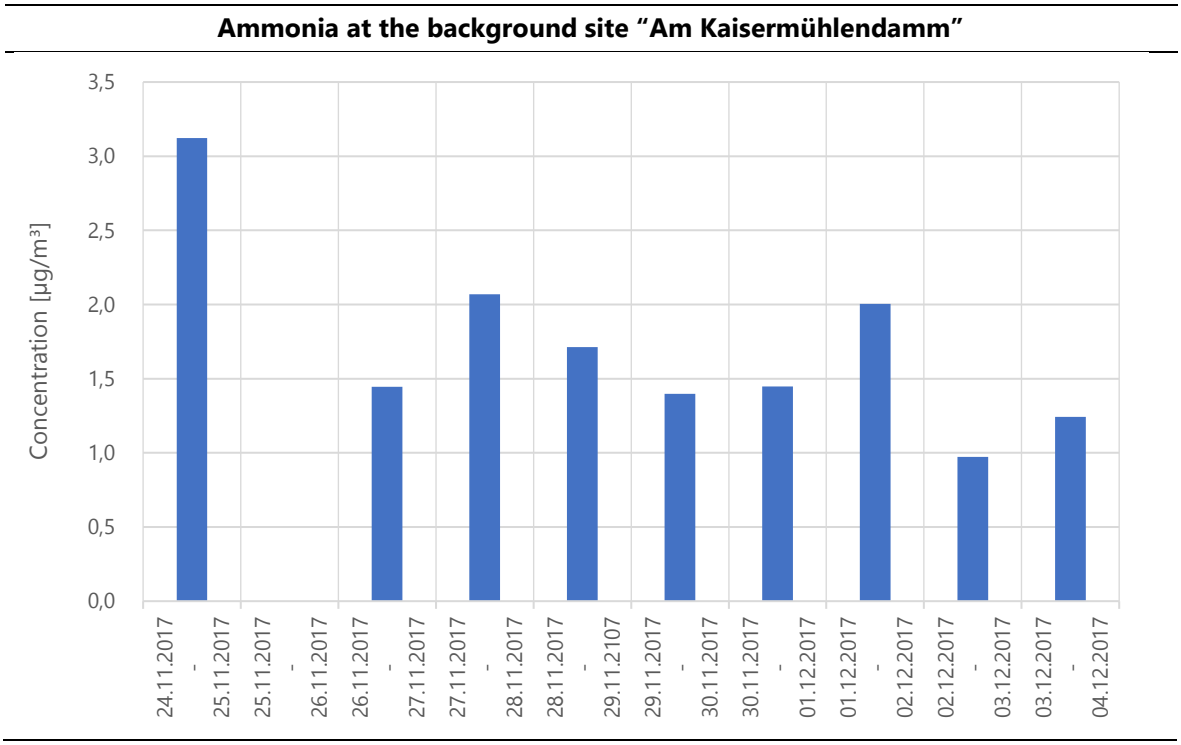


Figure 13: Concentration of ammonia at the background site "Am Kaisermühlendamm from 24.11.2017 – 04.12.2017

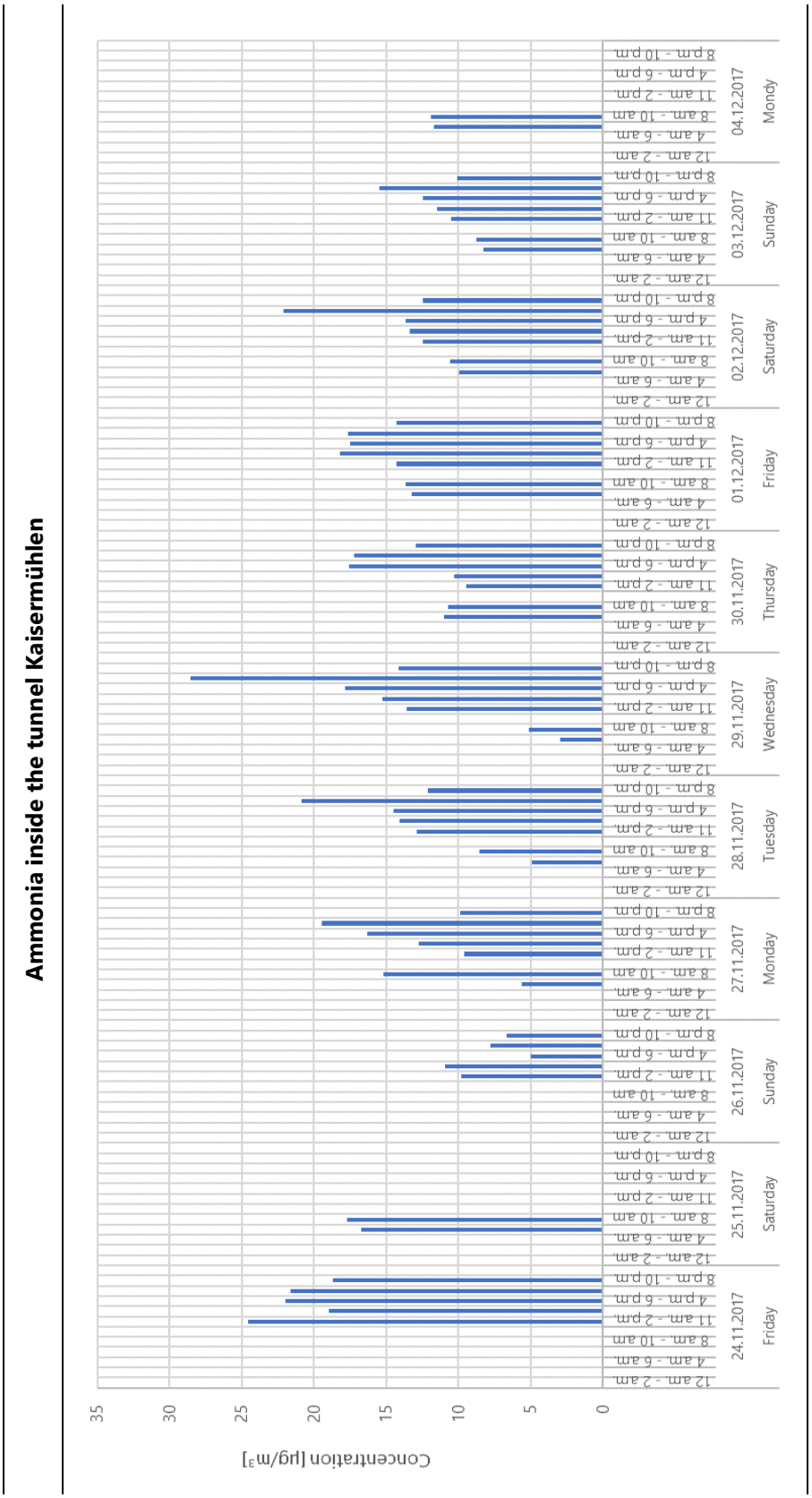


Figure 14: Concentration of ammonia in the south-eastern bonded bore of tunnel Kaisermühlen from 24.11.2017 – 04.12.2017

4.3.3 Ammonium nitrate in particulate matter

Particulate matter for the determination of particulate ammonium nitrate (NH_4NO_3) was collected as daily samples from 11 am till 10 am on the following day over the investigated period from 24.11.217 – 04.12.2017 at both measurement sites.

The obtained concentrations of ammonium (NH_4^+) and nitrate (NO_3^-) inside the tunnel Kaisermühlen are compared to background levels. The results are presented in Figure 15. The values from 25.11.2017 till 26.11.2017 are missing due to a failure in operation. Missing values elsewhere indicate concentrations below limits of detection (Table 04, page 19).

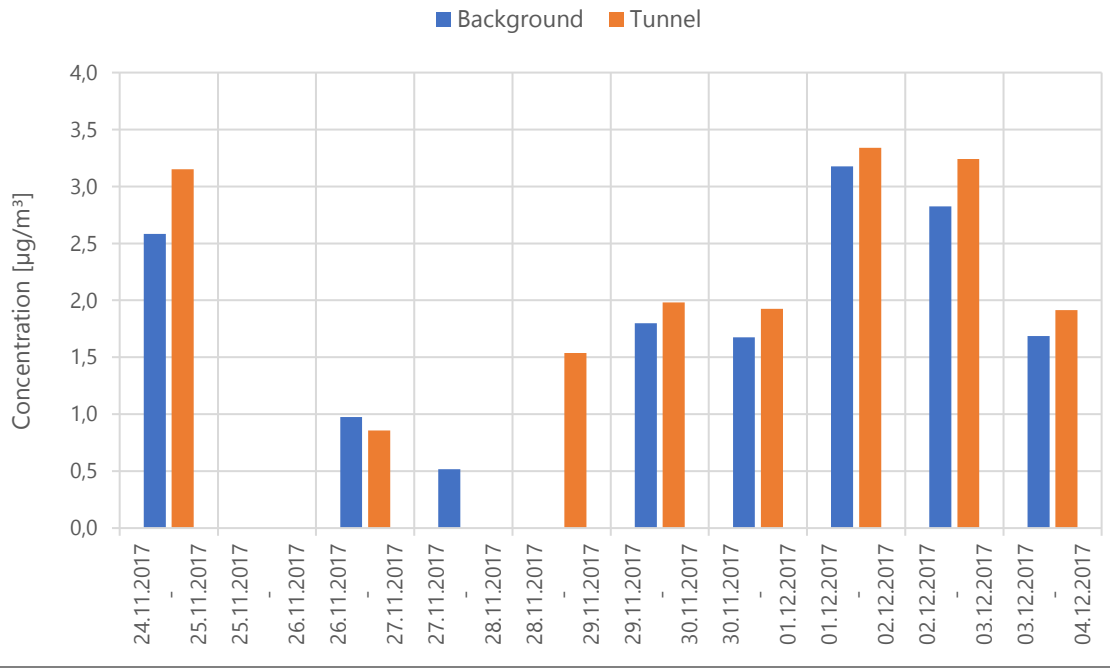
Regarding ammonium, the average concentrations are 2,2 µg/m³ and 1,9 µg/m³ inside the tunnel and at the background site Kaisermühlen, respectively. For nitrate, the average concentration inside the tunnel is 5,3 µg/m³, whereas background levels are 4,9 µg/m³ on average. Taking concentrations of sulphate in particulate matter (Figure 16) into account, the molar ratio of ammonium and the sum of nitrate and sulphate is 1. Sulphate only comprises approximately 17% at both measurement sites.

The increase in concentration for both ammonia and nitrate between ambient air and tunnel air is approximately 10%. Therefore, a reaction between the emitted compounds nitrogen oxides (NO_x) and ammonia (NH_3) forming particulate ammonium nitrate happens to a lesser extent inside the tunnel Kaisermühlen. The proportion of particulate bonded ammonia to gaseous ammonia is inside the tunnel is 16%. The sum of ammonia and ammonium at the background site is 3,6 µg/m³, which is only a fifth part of the concentration observed inside the tunnel Kaisermühlen (15,7 µg/m³).

Emission factors of particulate ammonium nitrate were not calculated due to the small data set.

Ammonium and nitrate in particulate matter

Ammonium



Nitrate

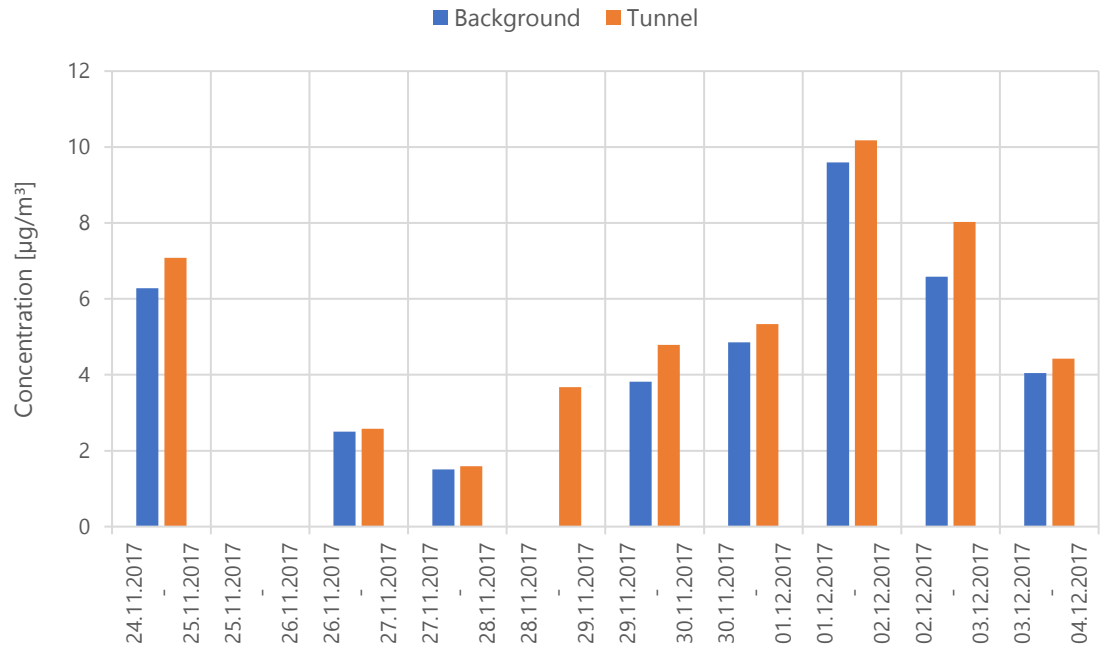


Figure 15: Concentrations of ammonium and nitrate in particulate matter in the south-eastern bonded bore of tunnel Kaisermühlen and at the background site "Am Kaisermühlendamm" from 24.11.217 – 04.12.2017

Sulphate in particulate matter

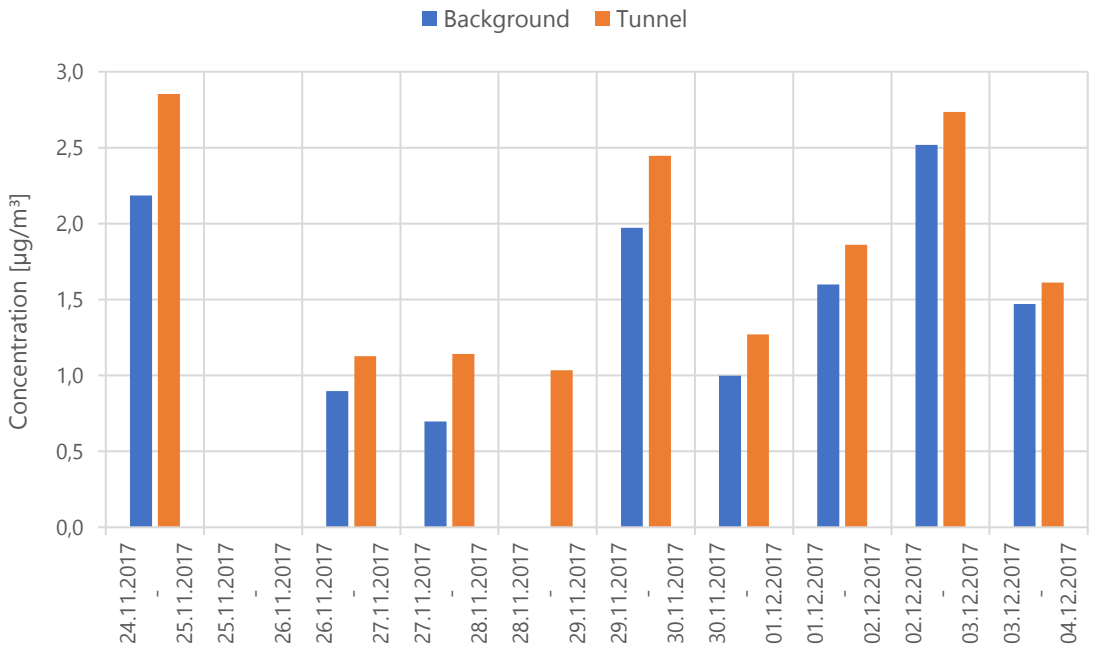


Figure 16: Concentrations of sulphate in particulate matter in the south-eastern boned bore of tunnel Kaisermühlen and at the background site "Am Kaisermühlendamm" from 24.11.2017 – 04.12.2017

5 EMISSION FACTORS

5.1 General aspects

As mentioned in section 2.5, the equation of Staehelin et al. (1997) [26] was used for calculating of emission factors:

$$\mathbf{EF_x(t)} = \frac{\mathbf{\Delta C_x(t) * u(t) * q * t}}{\mathbf{N(t) * d}} \quad (7)$$

EF_x	...	Emission factor of compound x [g/vkm]
ΔC_x	...	Difference in concentration of compound x (tunnel – background) [g/m^3]
u	...	Air velocity [m/h]
q	...	Tunnel cross-section [m^2]
t	...	Duration of time interval [h]
N	...	Number of vehicles [-]
d	...	Distance between the measurement sites

At first, the measured background levels were subtracted from the concentrations inside the tunnel Kaisermühlen in order to obtain the emissions from vehicular traffic only. Together with the volume flow – the product of measured longitudinal air velocity and tunnel cross-section (117 m²) – the emission mass flux was calculated. This emission mass flux was divided by the number of total vehicles driving through the tunnel and the distance from tunnel entrance to measurement point (1.644 m).

For the calculation of emission factors, only measurement data was taken into account, where the according traffic volume exceeds 1000 vehicles/h. Less traffic numbers lead to a low tunnel jet, which makes the emission mass flow difficult to quantify due to the greater uncertainty.

5.2 Nitrogen oxides (NO_x)

5.2.1 Emission factors for nitrogen oxides (NO_x)

The calculated emission factors for nitrogen oxides (NO_x) in dependence on the traffic volume are shown in Figure 17. In addition, the emission factors were differentiated

according to fleet velocity. Furthermore, the emission factors were differentiated according to the proportion of heavy duty vehicles (Figure 18).

NO_x emission factors differentiated according to fleet velocity

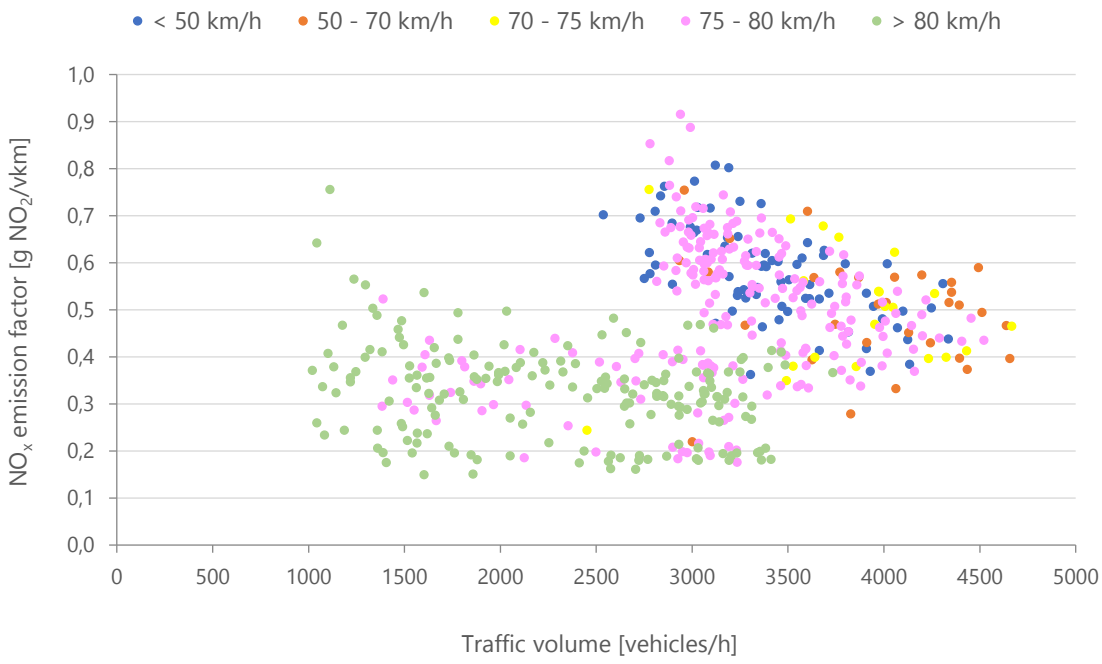


Figure 17: Emission factors for NO_x dependent on traffic volume with differentiation into fleet velocity

NO_x emission factors differentiated according to HDV proportion

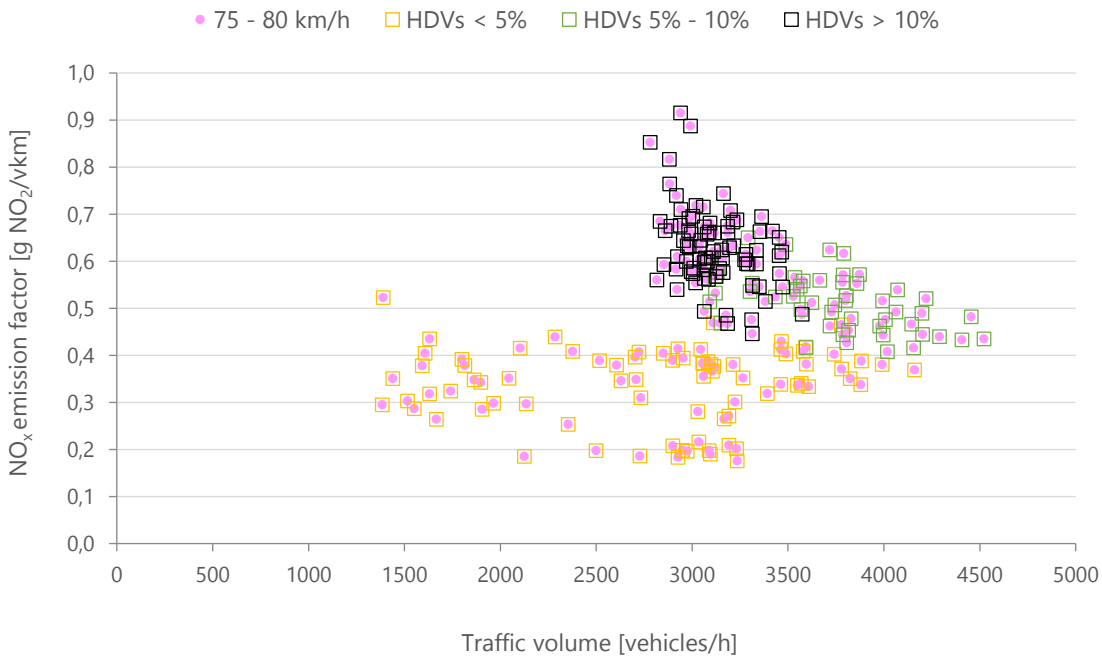


Figure 18: Emission factors for NO_x dependent on traffic volume with differentiation into HDV proportion for an average fleet velocity of 75 – 80 km/h

The average emission factor for NO_x is 0,45 g NO_2/vkm for the actual Austrian vehicular fleet passing through the south-eastern bonded bore of tunnel Kaisermühlen with an average HDV portion of 5,2% during the investigated period from 16.11.2017 – 04.12.2017 (6 am – 10 pm).

High traffic volume lead to lower fleet velocities and vice versa. The calculated emission factors at low traffic volumes and higher fleet velocities are generally lower than the emission factors measured at high traffic numbers. Therefore, the scatter plot in Figure 17 can be divided into two traffic conditions, further explanation will follow shortly.

Another point worth mentioning is, that the proportion of HDVs in the vehicular fleet has a large influence on the emission factor. For the sake of clarity, only data points at an average fleet velocity between 75 - 80 km/h were further differentiated into HDV proportion and presented in Figure 18. A positive correlation can be observed: the higher the proportion of HDVs in the vehicular fleet, the higher the emission factor of NO_x. Consequently, the emission factors measured on weekends are lower than those on workdays.

As a consequence of the relations discussed for Figure 17 and Figure 18, the following conclusions can be drawn:

- The proportion of HDVs in the vehicular fleet has an immense influence on the emission factor for NO_x. The smaller the HDV portion the lower the emission factor. Hence, separate emission factors for passenger cars and HDVs can be calculated by linear regression analysis (see section 5.2.2).
- Two different traffic conditions occur in the tunnel Kaisermühlen due to different fleet velocities. "Fluid traffic" is observed with higher fleet velocities, whereas at high traffic volumes – and therefore lower fleet velocities – the traffic condition can be described as "saturated traffic". This differentiation into two traffic conditions will be used for further data analysis.
- HDV proportion determines also the fleet velocity. The smaller the HDV portion the higher the average fleet velocity. The reason for this is the difference in speed limits: passenger cars are allowed to go 80 km/h, whereas the speed limit for heavy-duty vehicles is 60 km/h. In addition, the HDV portion influences the average velocity of passenger cars. Passenger cars on the right lane have to adapt the driving speed of HDVs, whereas on the left lane the passenger cars may go hardly unaffected. Consequently, the traffic condition can also be characterised by the proportion of HDV in the vehicular fleet. "Fluid traffic" is observed with lower HDV portion, which occurs in the evening of working days and on weekends particularly. Fleets with a higher share of HDVs are considered as "saturated traffic", which appears mainly on workdays from 6 am till 6 pm.

5.2.2 Linear regression analysis of emission factors for NO_x

As mentioned above, separate emission factors for passenger cars and heavy-duty vehicles can be calculated by linear regression analysis when a dependency of the emission factor on the HDV proportion in the vehicular fleet is given. The regression model is based on following equation (8): [35]

$$\mathbf{EF_x(t) = \alpha + \beta * pHDV(t) + \varepsilon(t)} \quad (8)$$

EF_x	...	Emission factor of compound x
α, β	...	Parameters of linear regression
pHDV	...	Proportion of HDVs
ε	...	Random error

In Figure 19 the calculated emission factors for NO_x were put in dependence on the corresponding HDV proportion. The regression results for the differentiated traffic conditions "fluid traffic" and "saturated traffic" are depicted in Figure 20. Extrapolation of the regression line to pHDV = 0 and pHDV = 1 yields the separate emission factors for passenger cars and HDVs, respectively.

NO_x emission factors dependent on HDV proportion

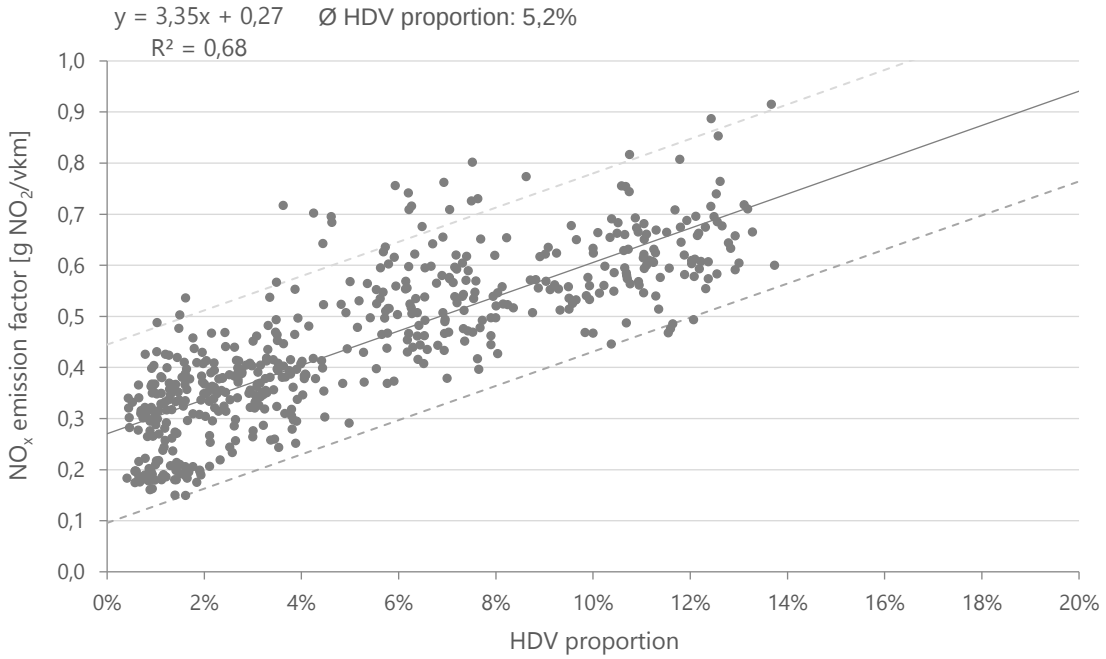
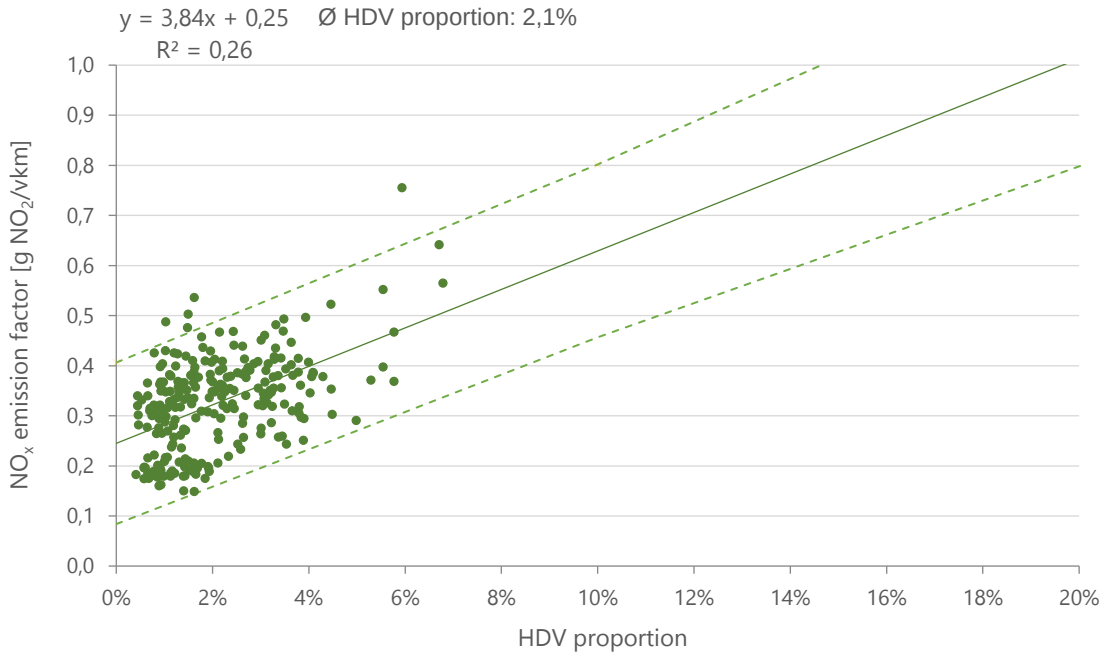


Figure 19: Emission factors for NO_x dependent on HDV proportion with linear regression analysis; dashed lines mark the 95% confidence interval

NO_x emission factor dependent on HDV proportion according to different traffic conditions

Fluid traffic



Saturated traffic

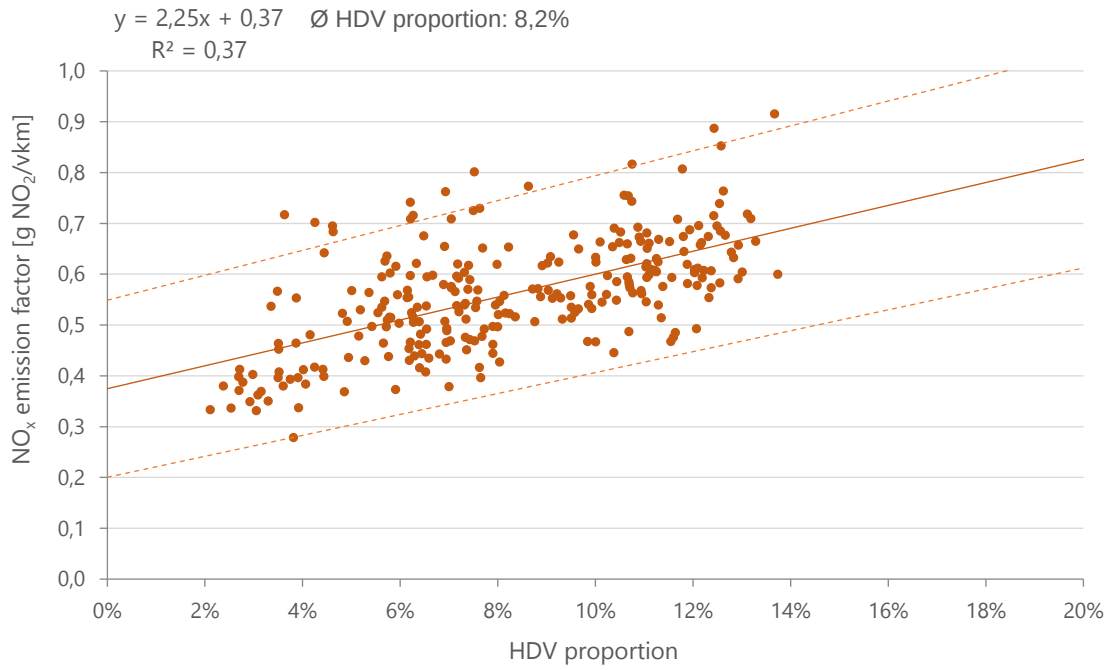


Figure 20: Emission factors for NO_x dependent on HDV proportion differentiated into different traffic conditions with linear regression analysis; dashed lines mark the 95% confidence interval

The average emission factor for NO_x is 0,45 g NO_2/vkm. The extrapolated emission factors for passenger cars and HDVs are 0,27 g NO_2/vkm and 3,62 g NO_2/vkm, respectively.

For the traffic condition described as "fluid traffic", the average emission factor for NO_x is 0,33 g NO_2/vkm. The extrapolated emission factor for passenger cars is 0,25 g NO_2/vkm. For the vehicle class HDV no emission factor was calculated, because the accuracy of the extrapolation is too low with low HDV proportion (2,1%).

For the traffic condition described as "saturated traffic", the average emission factor for NO_x is 0,56 g NO_2/vkm. The extrapolated emission factors for passenger cars and HDVs are 0,37 g NO_2/vkm and 2,62 g NO_2/vkm, respectively.

5.3 Nitrogen dioxide (NO_2)

5.3.1 Emission factors for nitrogen dioxide (NO_2)

The calculated emission factors for nitrogen dioxide (NO_2) in dependence on the traffic volume are shown in Figure 21. In addition, the emission factors were differentiated according to fleet velocity. Furthermore, the emission factors were differentiated according to the proportion of heavy duty vehicles (Figure 22).

The average emission factor for NO_2 is 78 mg NO_2/vkm for the actual Austrian vehicular fleet passing through the south-eastern bonded bore of tunnel Kaisermühlen with a HDV proportion of 5,2% during the investigated period from 16.11.2017 – 04.12.2017.

The discussed relations between emission factors, share of HDVs in the vehicular fleet and fleet velocity for NO_x were also observed for NO_2. However, the relations found for NO_2 are less pronounced compared to NO_x.

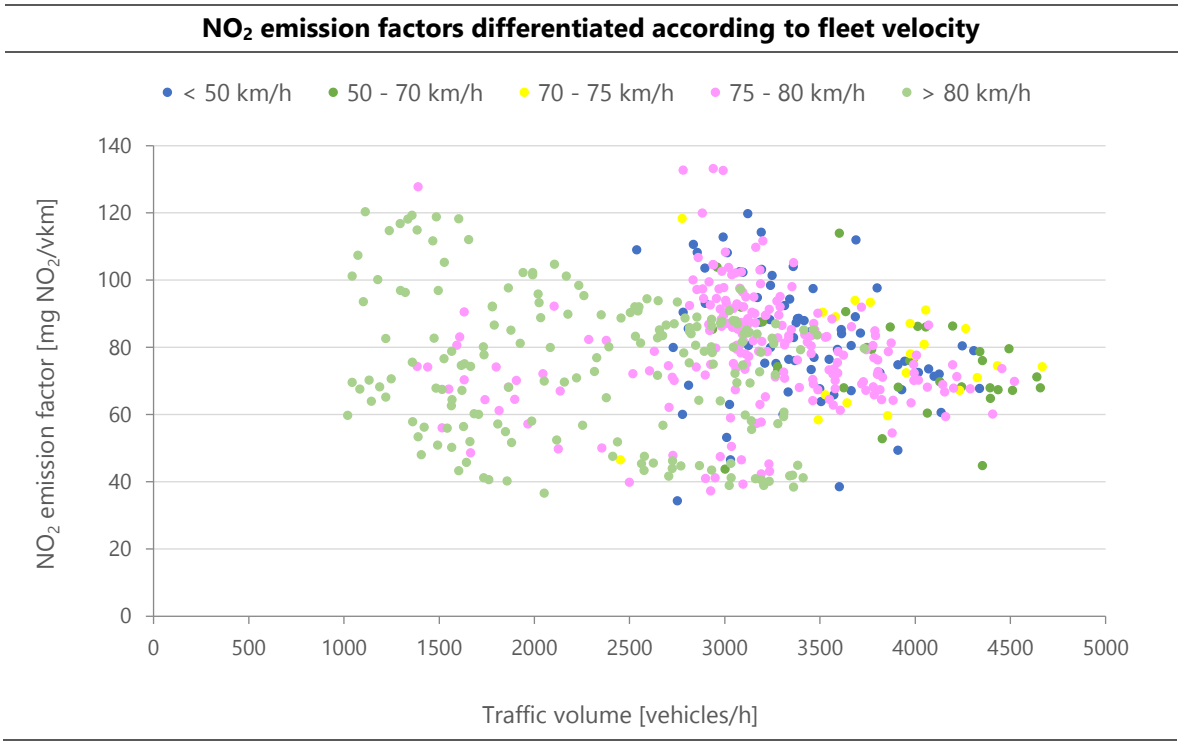


Figure 21: Emission factors for NO_2 dependent on traffic volume with differentiation into fleet velocity

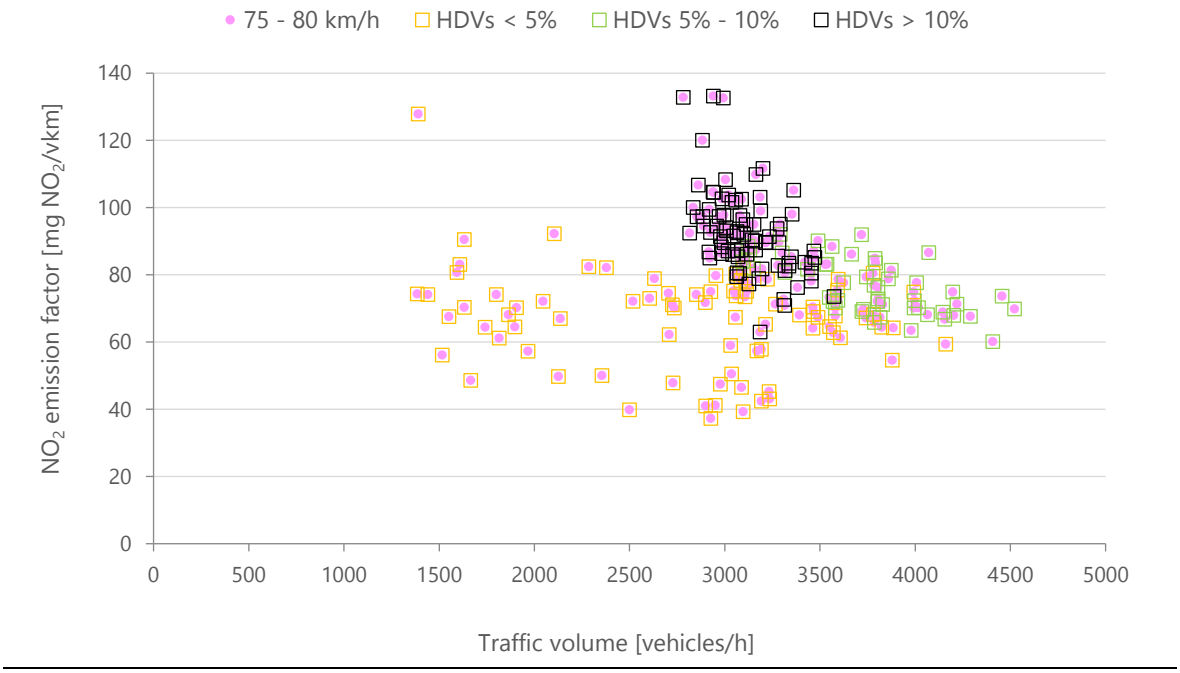


Figure 22: Emission factors for NO_2 dependent on traffic volume with differentiation into HDV proportion for an average fleet velocity of 75 – 80 km/h

5.3.2 Linear regression analysis of emission factors for NO_2

Analogous to the emission factors for NO_x, the calculated NO_2 emission factors were put into dependence on the HDV proportion (Figure 23). The regression results for the differentiated traffic conditions "fluid traffic" and "saturated traffic" are shown in Figure 24.

NO_2 emission factors dependent on HDV proportion

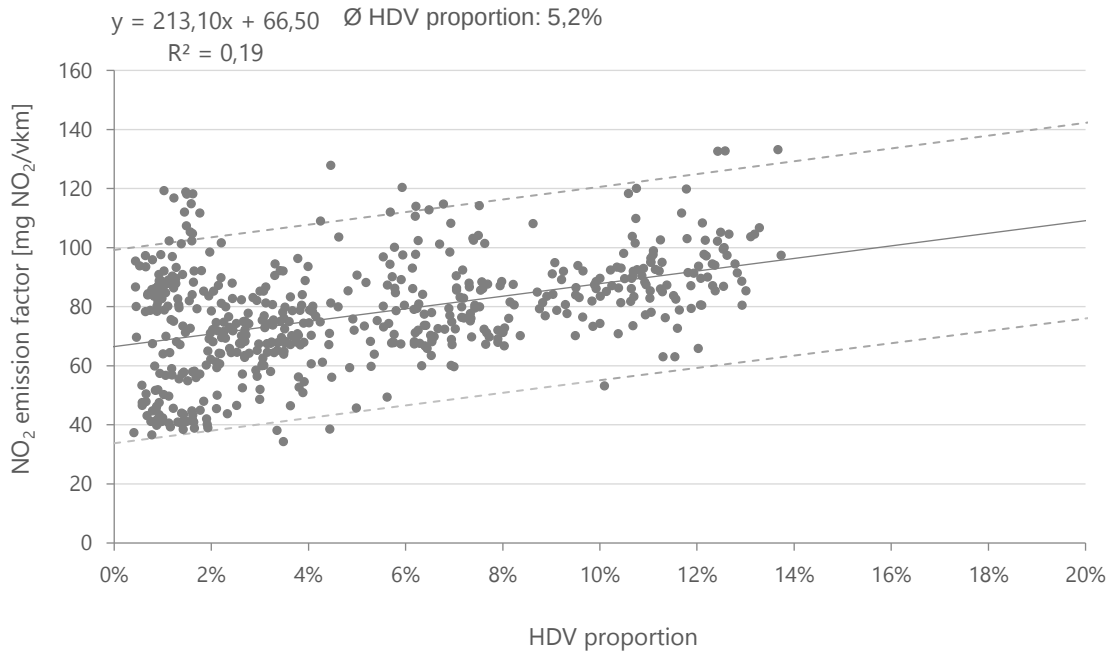


Figure 23: Emission factors for NO_2 dependent on HDV proportion with linear regression analysis; dashed lines mark the 95% confidence interval

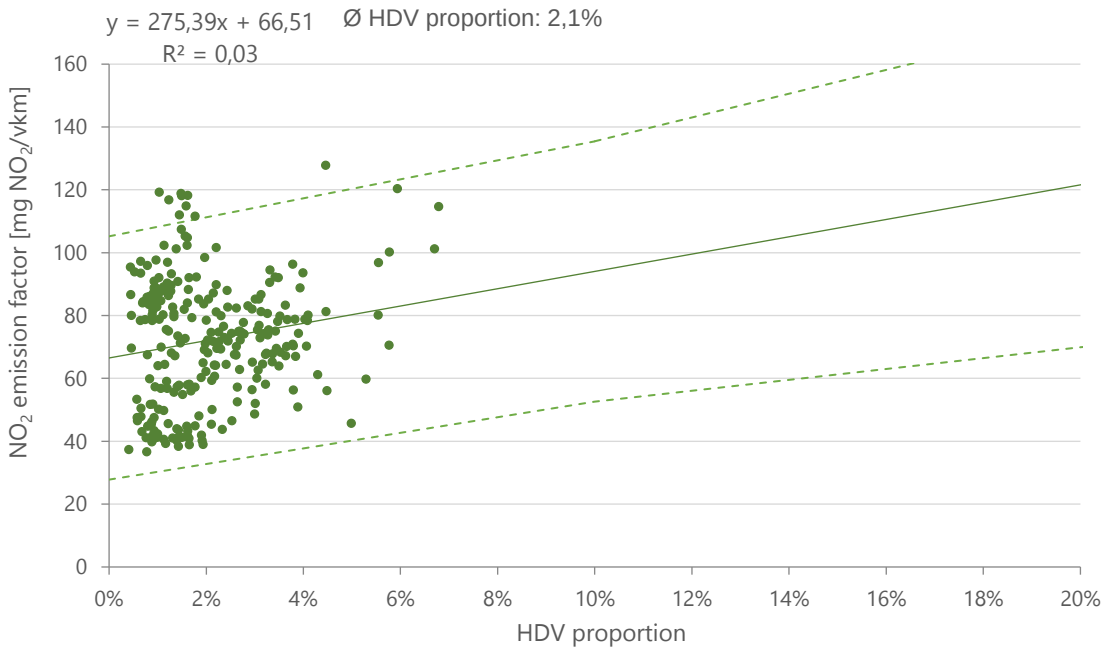
The average emission factor for NO_2 is 78 mg NO_2/vkm. The extrapolated emission factors for passenger cars and HDVs are 67 mg NO_2/vkm and 280 mg NO_2/vkm, respectively.

For the traffic condition described as "fluid traffic", the average emission factor for NO_2 is 72 mg NO_2/vkm. The extrapolated emission factor for passenger cars is 67 mg NO_2/vkm. For the vehicle class HDV no emission factor was calculated, because the accuracy of the extrapolation is low with low HDV proportion.

For the traffic condition described as "saturated traffic", the average emission factor for NO_2 is 83 mg NO_2/vkm. The extrapolated emission factors for passenger cars and HDVs are 59 mg NO_2/vkm and 350 mg NO_2/vkm, respectively.

NO_2 emission factor dependent on HDV proportion according to different traffic conditions

Fluid traffic



Saturated traffic

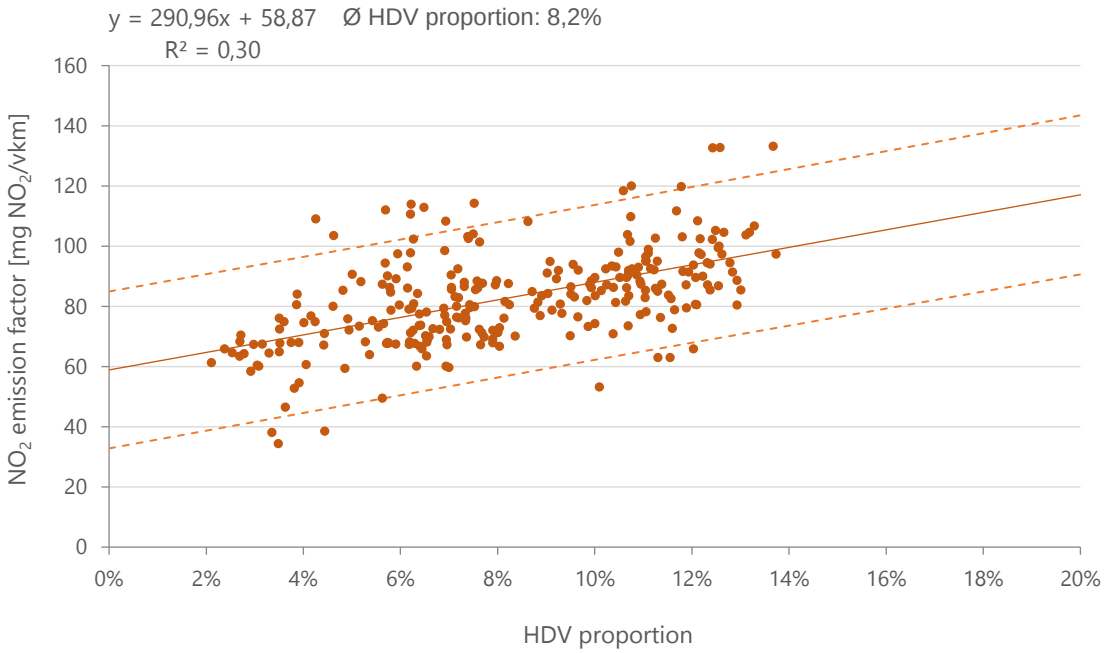


Figure 24: Emission factors for NO_2 dependent on HDV proportion differentiated into different traffic conditions with linear regression analysis; dashed lines mark the 95% confidence interval

5.4 Ammonia

5.4.1 Emission factors for ammonia (NH_3)

The calculated emission factors for ammonia (NH_3) in dependence on traffic volume are shown in Figure 25. In addition, the emission factors were differentiated according to fleet velocity. Furthermore, the emission factors were differentiated according to the proportion of heavy duty vehicles (Figure 26).

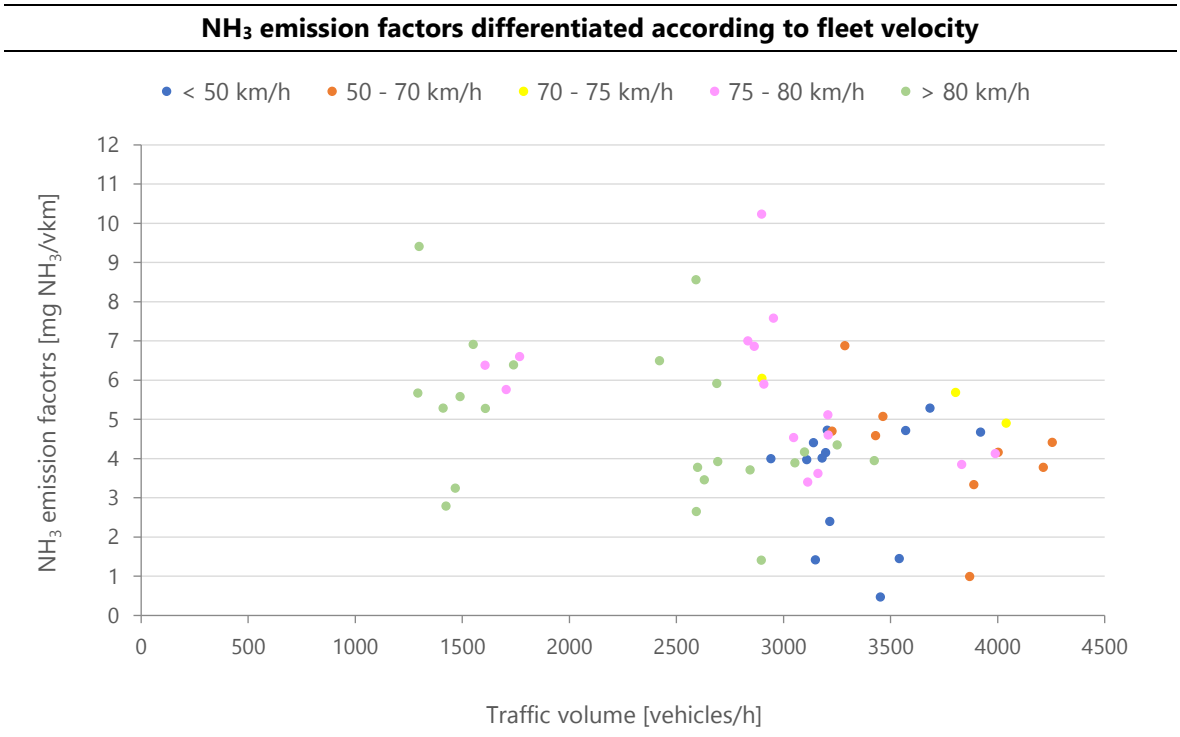
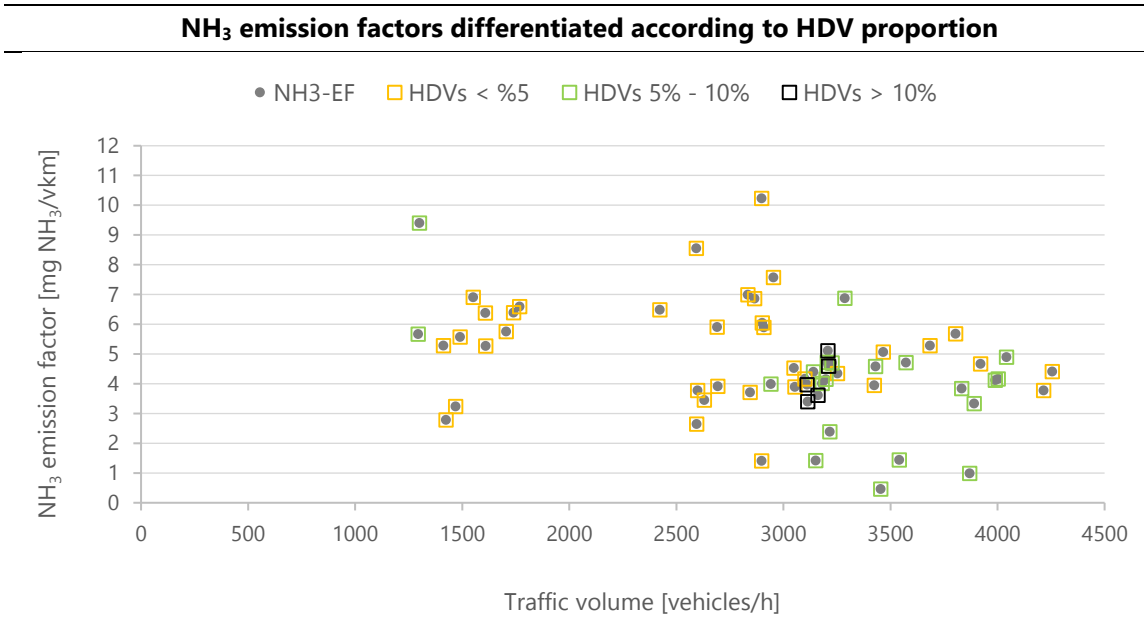


Figure 25: Emission factors for NH_3 dependent on traffic volume with differentiation into fleet velocity

The average emission factor for NH_3 is 4,7 mg NH_3/vkm for the actual Austrian vehicular fleet passing through the south-eastern bonded bore of tunnel Kaisermühlen with a HDV portion of 4,8% during the investigated period from 24.11.2017 – 04.12.2017.

On contrast to the nitrogen oxides NO_x and NO_2, the data set for ammonia could not be divided into different traffic conditions. Figure 26 indicates, that the share of HDVs in the vehicular fleet does not influence the emission factor for NH_3, as it was observed for NOx and NO_2 in Figure 18 and Figure 22,respectively.

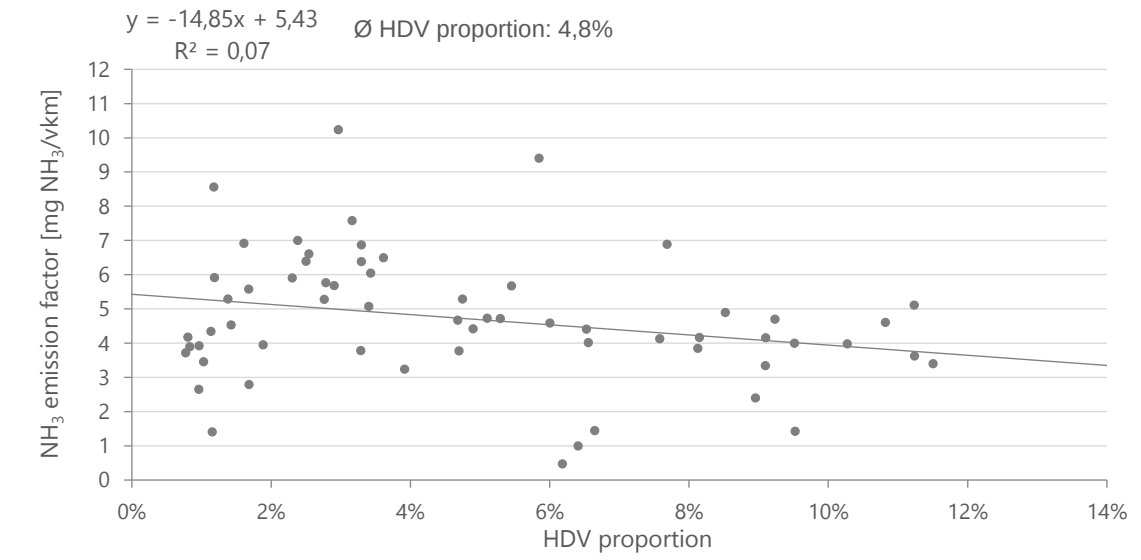


NH_3 emission factors differentiated according to HDV proportion

Figure 26: Emission factors for NH_3 dependent on traffic volume with differentiation into HDV proportion

5.4.2 Linear regression analysis of emission factors for NH_3

Analogous to the nitrogen oxides NO_x and NO_3, the calculated emission factors for NH_3 were put into dependence on the HDV proportion (Figure 27). A linear dependence of the emission factor for NH_3 on the share of HDVs in the vehicular fleet is not given according to Figure 27. Therefore, the calculation of separate emission factors for passenger cars and HDVs is not possible.



NH_3 emission factors dependent on HDV proportion

Figure 27: Emission factors for NH_3 dependent on HDV proportion with linear regression analysis

5.5 Summary of calculated emission factors

Table 08 gives an overview of the calculated emission factors for NO_x, NO_2 and NH_3 for the actual Austrian vehicular fleet based on measurements in the south-eastern bonded bore of tunnel Kaisermühlen.

Table 08: Summary of the calculated emission factors for NO_x, NO_2 and NH_3 for the actual Austrian vehicular fleet

	Nitrogen oxides NO_x	**Nitrogen dioxide NO_2**	**Ammonia NH_3**
Investigated period	16.11.2017 – 04.12.2017	16.11.2017 – 04.12.2017	24.11.2017 – 04.12.2017
Average HDV proportion	5,2%	5,2%	4,8%
Average emission factor for the entire fleet	0,45 g NO_2/vkm	78 mg NO_2/vkm	4,7 mg NH_3/vkm
• Fluid traffic	0,33 g NO_2/vkm	72 mg NO_2/vkm	-
• Saturated traffic	0,56 g NO_2/vkm	83 mg NO_2/vkm	-
Average emission factor for passenger cars	0,27 g NO_2/vkm	67 mg NO_2/vkm	-
• Fluid traffic	0,25 g NO_2/vkm	67 mg NO_2/vkm	-
• Saturated traffic	0,37 g NO_2/vkm	59 mg NO_2/vkm	-
Average emission factor for HDVs	3,62 g NO_2/vkm	280 mg NO_2/vkm	-
• Fluid traffic	-	-	-
• Saturated traffic	2,62 g NO_2/vkm	350 mg NO_2/vkm	-

6 COMPARISON WITH EMISSION FACTORS FROM HBEFA 3.3

6.1 General aspects

The calculated emission factors for NO_x, NO_2 and NH_3 for the actual Austrian vehicular fleet obtained from a measurement campaign conducted in the south-eastern bonded bore of tunnel Kaisermühlen are compared with emission factors from the "Handbook Emission Factors of Road Transport" (HBEFA) in the latest version 3.3 published in April 2017 by ERMES group.

The HBEFA is an electronic database which provides emission factors for all vehicle classes on a variety of traffic situations, including all regulated and some non-limited air pollutants, such as NH_3. Traffic situations are characterised by four parameters: area (for example rural or urban), road type (for example highway), speed limit and traffic condition (for example fluid traffic or saturated traffic). However, no traffic situation for an inner-city tunnel with "Section Control" is available.

The traffic situations listed in Table 09 best reflect the situations inside the tunnel Kaisermühlen and are used for comparison. The choice is based on the different speed limits for passenger cars and HDVs as well as on the observed fleet velocities and high traffic volumes.

Table 09: Overview of traffic situation used in HBEFA 3.3 for comparison

Vehicle class	Area	Road type	Speed limit	Traffic condition	Abbreviation
Passenger car	Urban	Inner-city highway	80	Fluid traffic	PC/80/fluid
			80	Saturated traffic	PC/80/saturated
			60	Saturated traffic	PC/60/saturated
HDV	Urban	Inner-city highway	60	Fluid traffic	HDV/60/fluid
				Saturated traffic	HDV/60/saturated

For all the traffic situations in Table 09 the corresponding emission factors for the investigated compounds NO_x, NO_2 and NH_3 are selected and compared with the calculated emission factors. The emission factors for the entire vehicular fleet are calculated weighting

the separate emission factors for passenger cars and HDVs from the HBEFA 3.3 according to the average proportion of HDVs observed in the investigated period.

It has to be noticed that the definition of HDVs used at the automatic counting station differs from the one used in HBEFA 3.3. HDVs by means of the HBEFA 3.3 are vehicles with a permissible maximum weight of more than 3.500 kg, whereas HDVs at the automatic counting station are defined by their length. Therefore, other vehicle classes, such as light duty vehicles or passenger cars with trailers, may be classified as HDVs as well. The portion of these vehicles classes with longer wheelbase, however, is relatively low, so they have no significant impact and are negligible.

The emission factors of the investigated compounds according to HBEFA 3.3 are given in Table 10 for each of the above-mentioned traffic situations.

Table 10: Emission factors for NO_x, NO_2 and NH_3 according to HBEFA 3.3 (2017)

Vehicle class	Traffic situation	NO_x [g NO_2/vkm]	NO_2 [mg/vkm]	NH_3 [mg/vkm]
Passenger car	PC/80/fluid	0,29	89	15
	PC/80/saturated	0,35	110	
	PC/60/saturated	0,35	108	
HDV	HDV/60/fluid	2,22	213	3
	HDV/60/saturated	2,70	259	

6.2 Nitrogen oxides (NO_x)

The comparison of the calculated fleet emission factor for NO_x as well as the separate emission factors for passenger cars and HDVs – with differentiation in two traffic conditions – is presented in the following subsections.

Fleet emission factors for NO_x

Figure 28 shows the comparison of the calculated fleet emission factors for NO_x with the ones obtained from the HBEFA 3.3.

The average calculated emission factor for NO_x for the entire fleet with an HDV proportion of 5,2% is 0,45 g NO_2/vkm. For fluid traffic the calculated emission factor is 0,33 g NO_2/vkm, which equals the emission factor from HBEFA 3.3. For saturated traffic the calculated emission factor is 0,56 g NO_2/vkm. In HBEFA 3.3 the corresponding emission factor is 0,54 g NO_2/vkm for both considered speed limits. This means a slightly higher emission factor as it is given in HBEFA 3.3 (+ 4%). All in all, the fleet emission factors for NO_x obtained experimentally agree well with the values according to HBEFA 3.3.

Comparison of fleet emission factors for NO_x with differentiation into traffic situations

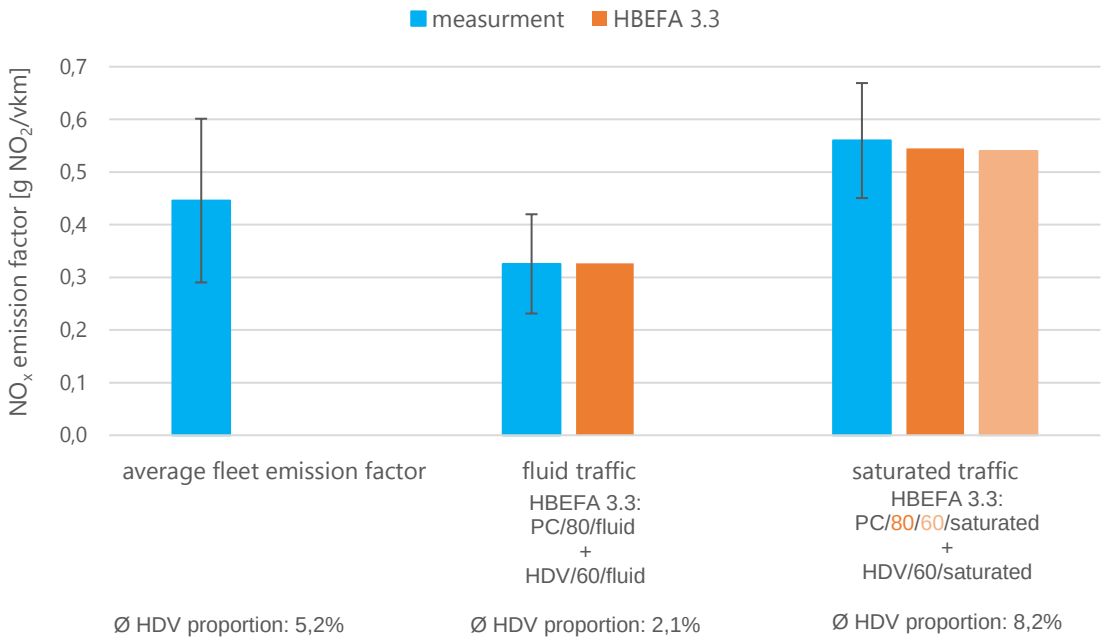


Figure 28:: Comparison of fleet emission factors for NO_x obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

Passenger car emission factors for NO_x

Figure 29 shows the comparison of the calculated passenger car emission factors for NO_x with the ones obtained from the HBEFA 3.3.

The average calculated emission factor for NO_x for passenger cars is 0,27 g NO_2/vkm. For fluid traffic the calculated passenger car emission factor is 0,25 g NO_2/vkm. In HBEFA 3.3 the corresponding emission factor is 0,29 g NO_2/vkm. Therefore, the emission factor found experimentally is 14% lower than the given value in HBEFA 3.3. For saturated traffic the calculated passenger car emission factor is 0,37 g NO_2/vkm, whereas the emission factors according to HBEFA 3.3 is 0,35 g NO_2/vkm for both considered speed limits. The calculated emission factor is 6% higher compared to the emission factors from HBEFA 3.3.

In general, the calculated emission factors for NO_x for passenger cars are comparable to the values given in HBEFA 3.3.

Comparison of passenger car emission factors for NO_x with differentiation into traffic situations

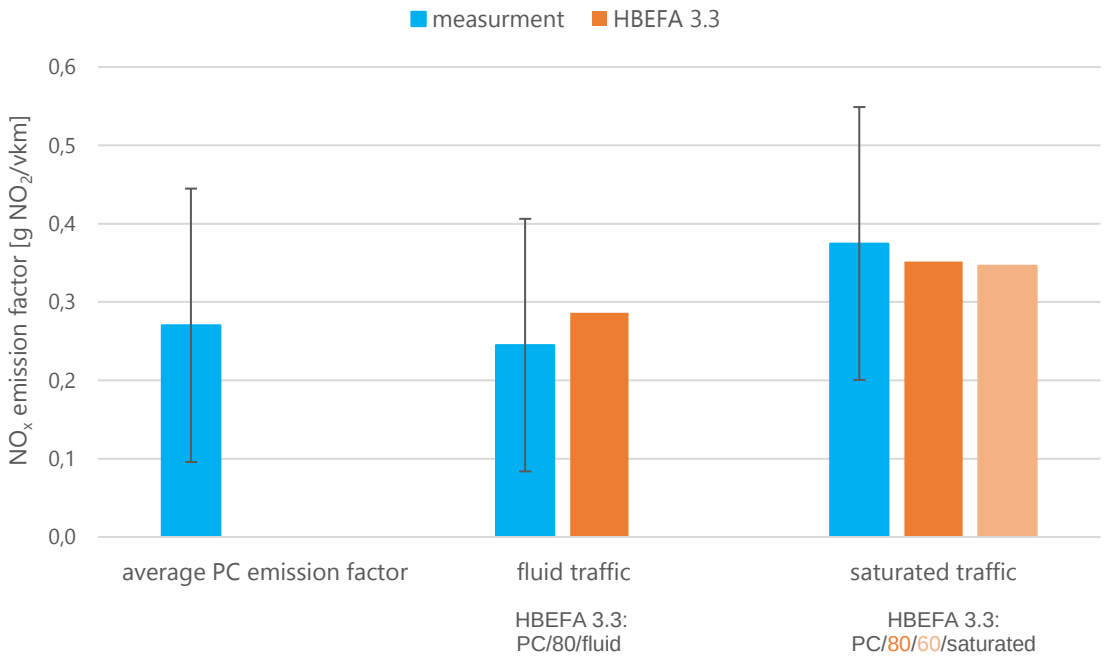


Figure 29: Comparison of passenger car emission factors for NO_x obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

HDV emission factors for NO_x

Figure 30 shows the comparison of the calculated passenger car emission factors for NO_x with the ones obtained from the HBEFA 3.3.

The average calculated emission factor for NO_x for HDVs is 3,62 g NO_2/vkm. For saturated traffic the calculated HDV emission factor is 2,63 g NO_2/vkm, which is slightly lower than the emission factor provided by HBEFA 3.3 with 2,70 g NO_2/vkm (- 3%). Regarding the traffic situation considered as fluid traffic the emission factor for NO_x for HDVs is not calculated due to the very low HDV proportion of 2,1%. Therefore, extrapolating to 100% HDV proportion is afflicted with great uncertainty.

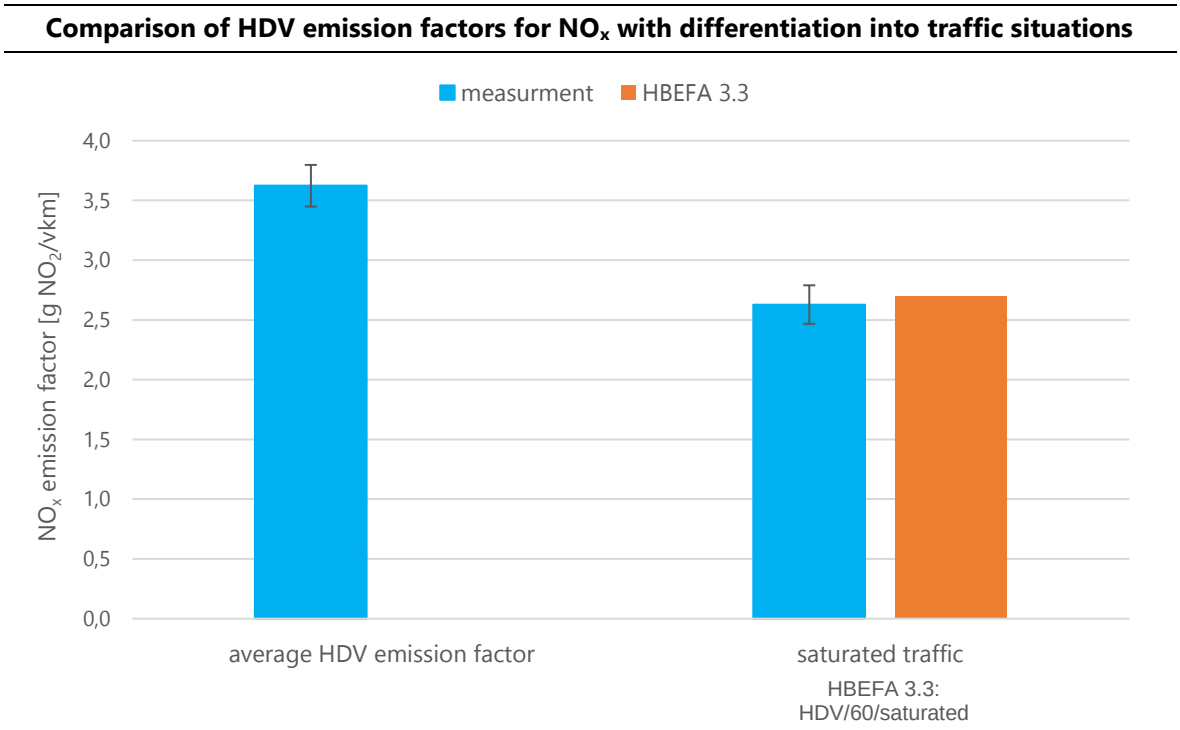


Figure 30: Comparison of HDV emission factors for NO_x obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

6.3 Nitrogen dioxide (NO_2)

The comparison of the calculated emission factors for NO_2 as well as the separate emission factors for passenger cars and HDVs – with differentiation in two traffic conditions – is presented in the following subsections.

Fleet emission factors for NO_2

Figure 31 shows the comparison of the calculated fleet emission factors for NO_2 with the ones obtained from the HBEFA 3.3.

Comparison of fleet emission factors for NO_2 with differentiation into traffic situations

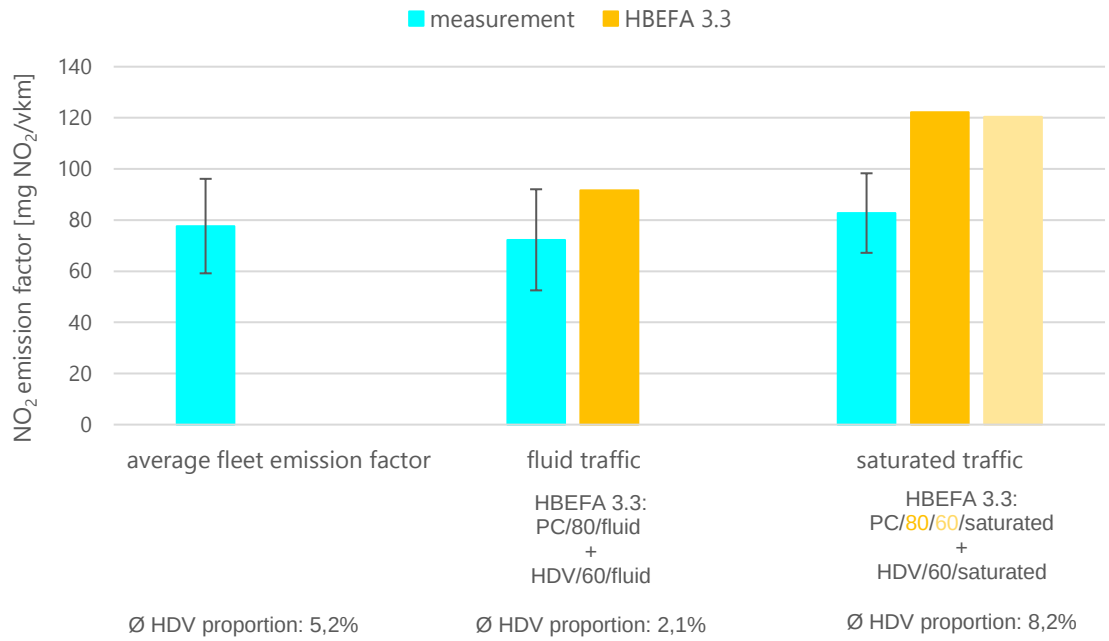


Figure 31: Comparison of fleet emission factors for NO_2 obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

The average calculated emission factor for NO_2 for the entire fleet with an HDV proportion of 5,2% is 78 mg NO_2/vkm. For fluid traffic the calculated emission factor is 72 mg NO_2/vkm. In HBEFA 3.3 the corresponding emission factor is 92 mg NO_2/vkm. Therefore, the emission factor obtained experimentally is 22% lower. For saturated traffic the calculated emission factor is 83 mg NO_2/vkm, which is significantly lower than the emission factors given in HBEFA 3.3. The given values are 122 mg NO_2/vkm and 120 mg NO_2/vkm with a speed limit for passenger cars of 80 km/h and 60 km/h. Therefore, the deviations are - 32% and - 31%, respectively.

The fleet emission factors for NO_2 obtained from the measurement campaign are generally lower than it is given in HBEFA 3.3.

Passenger car emission factors for NO_2

Figure 32 shows the comparison of the calculated passenger car emission factors for NO_x with the ones obtained from the HBEFA 3.3.

Comparison of passenger car emission factors for NO_x with differentiation into traffic situations

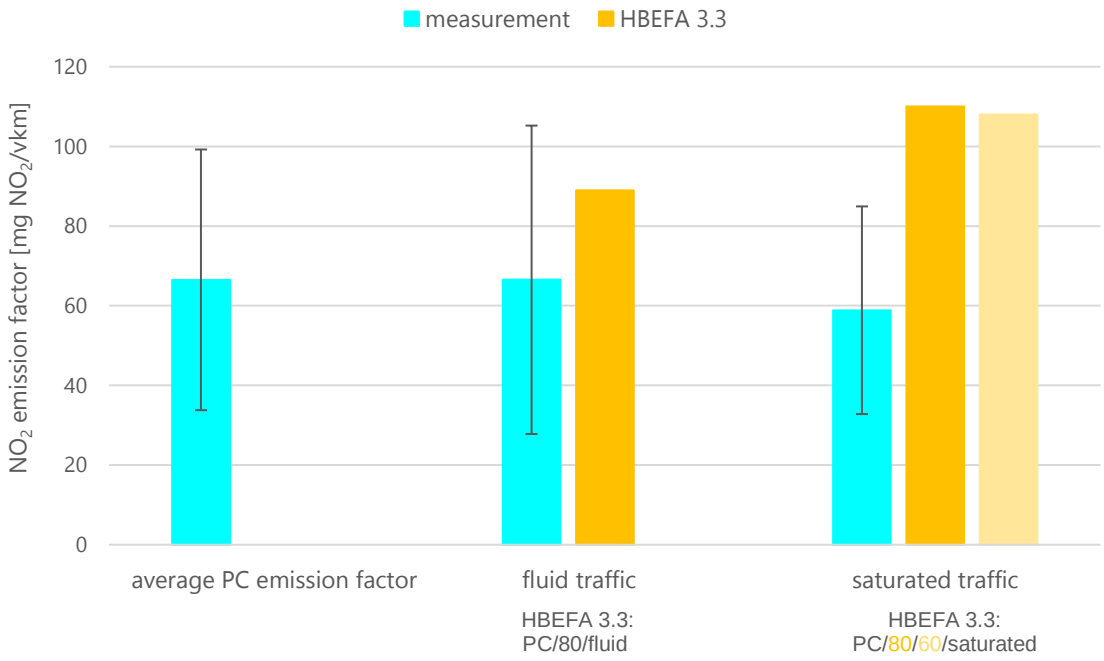


Figure 32: Comparison of passenger car emission factors for NO_2 obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

The average calculated emission factor for NO_2 for passenger cars is 67 mg NO_2/vkm. For fluid traffic the calculated passenger car emission factor is also 67 mg NO_2/vkm. In HBEFA 3.3 the corresponding emission factor is 89 mg NO_2/vkm. Therefore, the emission factor found experimentally is 25% lower than the given value in HBEFA 3.3. For saturated traffic the calculated passenger car emission factor is 60 mg NO_2/vkm, which is significantly lower than the emission factors given in HBEFA 3.3. The given values are 110 mg NO_2/vkm and 108 mg NO_2/vkm with a speed limit for passenger cars of 80 km/h and 60 km/h. Therefore, the deviations are - 46% and - 44%, respectively.

The passenger car emission factors for NO_2 obtained from the measurement campaign are generally lower than it is given in HBEFA 3.3.

HDV emission factor for NO_2

Figure 33 shows the comparison of the calculated HDV emission factors for NO_x with the ones obtained from the HBEFA 3.3.

Comparison of HDV emission factors for NO_2 with differentiation into traffic situations

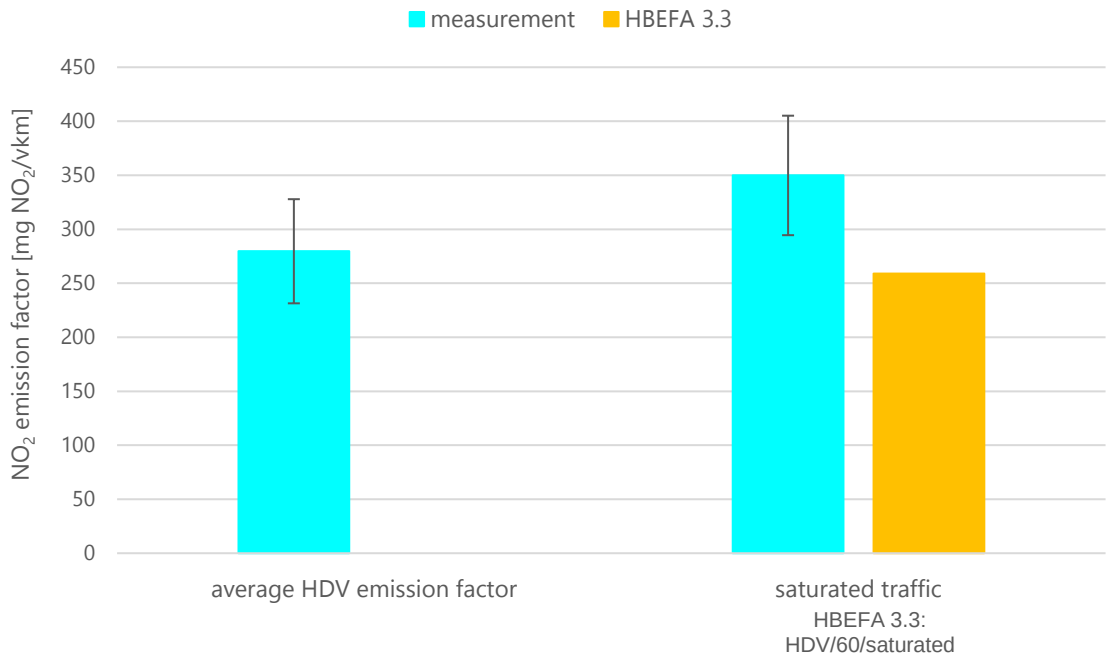


Figure 33: Comparison of HDV emission factors for NO_2 obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

The average emission factor for NO_2 for HDVs is 279 mg NO_2/vkm. For saturated traffic the calculated HDV emission factor is 350 mg NO_2/vkm. In HBEFA 3.3 the corresponding emission factor for HDVs is 259 mg NO_2/vkm, which means a deviation of + 35% for the calculated HDV emission factor. Regarding the traffic situation considered as fluid traffic the emission factor for NO_2 for HDVs is not calculated due to the very low HDV proportion of 2,1%. Therefore, extrapolating to 100% HDV proportion is afflicted with great uncertainty.

In general, the emission factors for NO_2 for HDVs obtained from the measurement campaign are higher compared to the values provided by the HBEFA 3.3.

6.4 Ammonia (NH_3)

Since for ammonia only an average fleet emission factor could be determined, the comparison has to be done in another way. Figure 34 shows the calculated fleet emission factor together with the emission factors provided by HBEFA 3.3 as well as the given emission factors for passenger cars and HDVs, separately.

Comparison of fleet emission factors for NH_3 with differentiation into traffic situations

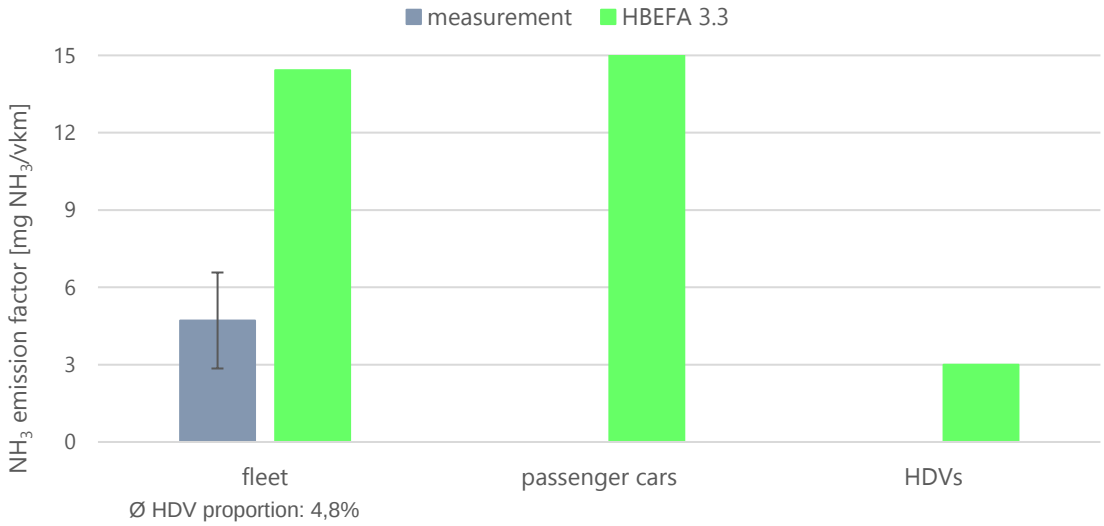


Figure 34: Comparison of fleet emission factor of NH_3 obtained from a measurement campaign in the south-eastern bonded bore of tunnel Kaisermühlen and values from HBEFA 3.3 (2017)

The average calculated fleet emission factor for NH_3 is 4,7 mg NH_3/vkm. According to HBEFA 3.3 the corresponding fleet emission factor is 14,4 mg NH_3/vkm, which is three times higher than the measured emission factor.

Regarding the emission factors for NH_3 provided in HBEFA 3.3 (Table 10), the same value is attributed to each of the considered traffic situations for passenger cars (15 mg NH_3/vkm) as well as for HDVs (3 mg NH_3/vkm). The reason for this is, that ammonia is yet still a non-regulated parameter, therefore, the data set, which the calculations are based on, is very meagre – compared to nitrogen oxides and other regulated pollutants, which are well investigated. Having a closer look on the emission factor for NH_3 for passenger cars in HBEFA 3.3, it can be seen, that this value of 15 mg NH_3/vkm is obtained by linear combination of the separate emission factors for diesel- and gasoline-driven passenger cars. The given emission factors for passenger cars powered by diesel and gasoline are 1 mg NH_3/vkm and 36 mg NH_3/vkm, respectively. The distribution of mode of driving for passenger cars in 2017 according to HBEFA 3.3 was 60% diesel and 40% gasoline, thus yielding the weighted emission factor for NH_3 for passenger cars of 15 mg NH_3/vkm.

7 COMPARISON WITH LITERATURE

7.1 Nitrogen oxides

7.1.1 Background levels of NO_2

The concentration measurement network in Austria is very dense, monitoring of air pollutants is conducted by the nine states and the Austrian Federal Environmental Agency. The observed nitrogen dioxide concentrations at the background site "Am Kaisermühlendamm" are compared to measured concentrations at a nearby monitoring site for urban background concentrations. The concentration trend at "Stadlau" is depicted in Figure 35. The NO_2 concentrations at both background sites are well comparable. Concentrations between 10 and 60 µg/m³ are observed at "Am Kaisermühlendamm" and at "Stadlau". [36]

Urban background concentrations for nitrogen dioxide measured at "Stadlau"

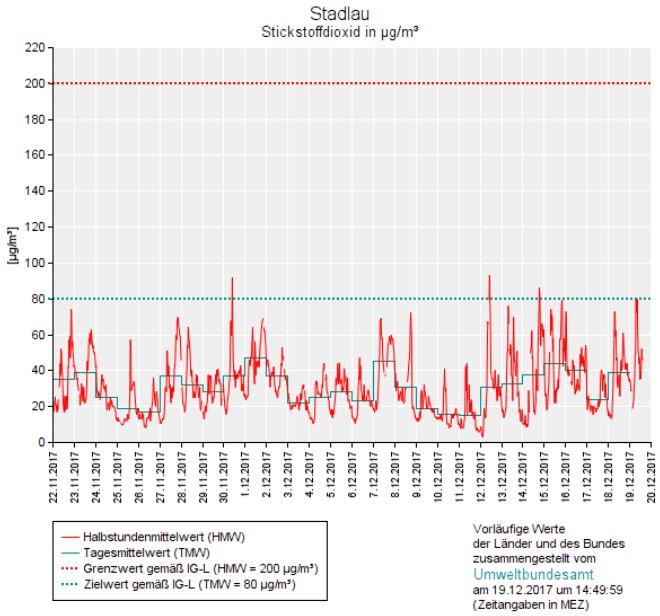


Figure 35: Urban background concentrations for nitrogen dioxide measured at "Stadlau" from 22.11.2107 till 20.12.2017 [36]

7.1.2 Concentrations and emission factors for NO_x inside tunnels

The concentrations of NO_2 and NO_x as well as the calculated emission factors are compared to values from other tunnel studies found in literature. Table 11 lists the literature used for comparison.

Table 11: Overview of concentrations and fleet emission factors for NO_x and NO_2 found in literature

	Urban H. (2006) [37]	**Staehelin J. et al. (1995) [38]**	**Ait-Helal W. et al. (2015) [39]**	**Present study**
Year	2005	1993	2011-2013	2017
State	Austria	Switzerland	Belgium	Austria
Tunnel	Tunnel Kaisermühlen	Gubrist tunnel	Leopold II tunnel	Tunnel Kaisermühlen
Traffic volume [vehicles/day]	41.000		24.000	52.000
Share of HDVs	10%	< 1% (Sunday) – 24% (workday)	HDV ban	5%
Concentration [µg NO_2/m^3]				
NO_x	1180			1170
NO_2	210			226
Emission factor [g NO_2/vkm]				
NO_x	0,67	2,25	0,54 ± 0,20	0,45
NO_2	0,07			0,08

Urban H. (2006) [37] conducted a similar measurement campaign in tunnel Kaisermühlen. The used method for determining concentrations of nitrogen oxides was the same as in the present study. Observed concentrations inside the tunnel were quite the same as found in this investigation. However, the traffic volume with a daily average of 41.000 vehicles and 10% HDVs was lower than it was during this campaign (52.000 vehicles per day and 5% HDVs). The concentration of NO_2 is found to be slightly higher than it was measured by Urban H., indicating a higher proportion of primary emitted nitrogen dioxide.
Regarding the calculated emission factors for NO_2 the values have not changed (0,07 g NO_2/vkm in 2005 and 0,08 g NO_2/vkm in 2017). Emission factors for NO_x, however, were found to be 33% lower.

Measurements conducted in the Gubrist Tunnel, Switzerland (Staehelin J. et al. (1995) [38]) in 1993 delivered an emission factor for NO_x of 2,25 g NO_2/vkm. The passenger cars in the vehicular fleet under investigation were mainly powered by gasoline, the portion of diesel-driven passenger cars was only 2,1%. Approximately 70% of gasoline vehicles were equipped with an TWC. A more recent investigation conducted by Ait-Helal W. et al. (2015) [39] in a traffic tunnel only restricted to light-duty vehicles calculated an average emission factor for nitrogen oxides of 0,54 g NO_2/vkm. Diesel (64%) and gasoline (36%) were the fuels

used for LDVs, which is well comparable to the Austrian composition of passenger cars in this study. Therefore, similar emission factors were found. It can be seen, that NO_x emission factors are decreasing with time, which is due to changes in the vehicular fleet, i.e. a higher proportion of diesel-driven passenger cars, and improved exhaust after-treatment systems.

Ntziachristos L. et al. (2016) [40] aimed to validate emission factors from models, i.e. the Handbook Emission Factors for Road Transport and to identify whether they adequately reflect real-word emissions in reaction to the revelations on NO_x emission control failures for diesel-driven passenger cars. Unlike the presented investigation, where emission factors from an entire fleet via tunnel study were calculated, a collective of 22 Euro 5 passenger cars and 17 Euro 6 passenger cars were tested in the laboratory as well as on-road using PEMS. Laboratory tests were divided into on-road cycles and regulated approval cycles. The results showed that for Euro 5 passenger cars the emission factors provided by COPERT, which is similar to the HBEFA, agree well with the experimentally found ones. This is because the modelling of emission factors is based on real-world driving cycles and on-road measurements with PEMS and not on type-approval test cycles. Regarding Euro 6 passenger cars, the measured emissions exceeded emission factors. However, it is predicted that on-road emissions will decrease, since manufacturers have to meet RDE standards. [24]

The comparison with literature shows comparable emission factors for NO_x with recent tunnel studies conducted in Europe. Former investigations, i.e. Staehelin J. et al. (2005) [38] show higher emission factors. However, the vehicular fleet and exhaust after-treatment systems have changed over time, which has to be considered for comparison. Ntziachristos L. et al. (2016) [40] also showed, that emission factors from models like HBEFA reflect real-world emissions very well, although passenger cars with Euro 6 standard emit higher amounts of NO_x in on-road measurements. It has to be noted, that modelling emission factors for Euro 6 passenger cars is based on a small data set which will be improved in near future.

7.2 Ammonia

The comparison of the calculated fleet emission factor for NH_3 with the values provided by HBEFA 3.3 does not show a good concordance (see Figure 34). Therefore, the calculated fleet emission factor for NH_3 is further compared with other emission factors obtained from tunnel studies found in literature.

So far, only few tunnel studies for determining fleet emission factors for NH_3 were conducted. Table 12 gives an overview of the investigations found in literature.

Table 12: Overview of fleet emission factors for NH_3 found in literature

	Moekli M. A. et al. (1996) [41]	**Vieira-Filho M. S. et al. (2016) [42]**	**Liu T. et al. (2014) [43]**	**Chang Y. et al. (2016) [44]**	**Present study**
Year	1995	2011	2013	2014	2017
State	Switzerland	Brazil	China	China	Austria
Tunnel	Gubrist tunnel	JQ tunnel	Zhujiang Tunnel	Handan tunnel	Tunnel Kaisermühlen
Traffic volume	38.000 vehicles/day	2.247 vehicles/hour	1.782 vehicles/hour	120.000 vehicles/day	52.000 vehicles/day
Share of HDVs	4%	10%	11,8%		5%
Measurement method	Photoacoustic Detection System	Impinger – 1mM H_2SO_4	Chemiluminescence	Impinger – 5 mM H_2SO_4	Filter pack - impregnated filters
Time resolution	10 min	1 hour	1 min	2 hours	2 – 3 hours
NH_3 concentration					
Background	~ 10 ppb	15,2 ± 11,3 µg/m³	53 ppb	5,6 ± 2,5 µg/m³	1,7 µg/m³
Tunnel	~ 390 ppb	46,5 ± 17,5 µg/m³	729 ppb	64,9 ± 11,5 µg/m³	13,5 µg/m³
EF [mg/vkm]	15 ± 4	44 ± 22	230 ± 14	28 ± 5	4,7 ± 1,9

Moekli M. A. et al. (1996) [41] conducted a measurement campaign for the determination of fleet emission factors for ammonia and ethene in 1995. The tunnel of interest was situated in the north of Zurich, Switzerland.
The Gubrist Tunnel is a 3,2 km long tunnel consisting of two bores with two lanes each. A daily traffic volume of 38.000 vehicles in each direction was observed during the measurement period. The speed limit inside the tunnel was 100 km/h. Proportion of HDVs never exceeded 15% with an average of 4,4%. Unfortunately, no information about the fuel types of the vehicular fleet was given.

The measurement of gaseous ammonia was achieved with a mobile photoacoustic trace gas detection system.
The calculated emission factor for ammonia inside Gubrist Tunnel was 15 ± 4 mg NH_3/vkm.

In May 2011 a measurement campaign for determining gaseous ammonia among other pollutants was carried out in a vehicular tunnel in the metropolitan area of São Paulo, Brazil by Vieira-Filho M. S. et al. (2016). [42]
The speed limit inside the 1,9 km long tunnel is 70 km/h. The vehicular fleet consisted mainly of light duty vehicles (LDVs) and motorcycles (approximately 90%), which were powered by gasohol, a mixture of gasoline and anhydrous ethanol. HDVs, by contrast, were driven with diesel. Traffic volume for LDVs was on average 2247 vehicles/h, the proportion of HDVs was negligible.
Ammonia sampling was performed by impinger systems with 1 mM H_2SO_4 as absorbent solution at a flow rate of 1 l/min.
Ammonia concentration inside the tunnel exceeded the measured background levels of a factor 3, figuring 46,5 µg/m³ and 15,2 µg/m³ for tunnel concentrations and background levels, respectively. The calculated fleet emission factors ranged from 2,6 to 104 mg NH_3/vkm with an average of 44 ± 22 mg NH_3/vkm.

Determination of a fleet emission factor for NH_3 was the aim of a tunnel study conducted in August 2013 by Liu T. et al. (2014). [43]
The measurement campaign was carried out in the 1,2 km long Zhujiang Tunnel in Guangzhou, China. The tunnel had two bores with each two lanes. Traffic volume was 1782 vehicles/h during the investigated period and speed limits were 15 – 50 km/h inside the tunnel. On average, 88,2% of the vehicles were powered by gasoline, HDVs were diesel-driven and were banned during rush hours and night time.
Ammonia was continuously measured by a chemiluminescence analyser.
Concentrations of gaseous ammonia found at the exit of the tunnel were 13,8 times that at the inlet. In addition, NH_3 concentrations showed a good correlation with total traffic density, whereas no correlation between ammonia concentration and diesel trucks was observed. The calculated emission factor for NH_3 was 230 ± 14 mg NH_3/vkm.

Chang Y. et al. (2016) [44] conducted a measurement campaign for the determination of emission factors for NH_3 in 2014.
The tunnel under investigation was Handan tunnel in Shanghai, China, a 720 m long tunnel with two bores and four lanes each.120.000 vehicles passed the tunnel daily with a speed limit of 80 km/h. Light-duty vehicles comprised 85% of the vehicular fleet. Unfortunately, no information about fuel types was given.
Sampling of ammonia was performed by an impinger system with 5 mM H_2SO_4 as absorbing agent and a flow rate of 1 l/min.
Concentrations were found to increase with distance from the tunnel entrance with values

at the deepest measurement point inside the tunnel exceeding the ammonia concentrations at the tunnel entrance by a factor 5. The calculated emission factor for NH_3 was 28 ± 5 mg NH_3/vkm.

The fleet emission factor for ammonia calculated within this study is 4,7 ± 1,9 mg NH_3/vkm and is the lowest NH_3 emission factor compared to the conducted tunnel investigations presented above.

It is noteworthy, that a variety of measurement principles for determining atmospheric ammonia concentrations as well as different methods for calculating emission factors were used. Unlike nitrogen oxides, where the measurement of chemiluminescence has established as the measurement principle of choice, determination of ammonia concentrations can be achieved with a variety of measurement methods. As can be seen in Table 12, five investigations, where four different methods were used, were compared. In addition, the time resolution strongly varies. The presented study shows the lowest time resolution (2 – 3 hours), whereas the highest figures 1 minute.
Furthermore, there are major differences in the vehicular fleet composition among the studies investigated and the fuel types differ to a large extent. Regarding this study, the Austrian fleet consists of approximately 75% passenger cars, of which 60% are diesel-powered, whereas in the above-mentioned investigations the vehicles are mainly driven with gasoline. According to HBEFA 3.3 passenger cars powered with gasoline emit more ammonia than diesel cars, namely 36 mg NH_3/vkm compared to 1 mg NH_3/vkm. Other literature [19] [20] also indicated TWC-equipped gasoline cars as bigger contributors to vehicular ammonia emissions than diesel-powered passenger cars. Thus, explaining the higher ammonia concentrations inside tunnels as well as higher fleet emission factors for NH_3 found in tunnel studies, where gasoline is the main fuel type used.

8 DISCUSSION

In this section the results found and presented previously, are examined critically and discussed.

Starting with nitrogen oxides, the calculated emission factors agree well with the ones provided by HBEFA 3.3 as well as with recent European tunnel studies found in literature. Also, for nitrogen dioxide, the emissions factors calculated and given in HBEFA 3.3 are comparable, though the experimentally found values for the entire vehicular fleet and passenger cars are generally lower. The calculation of emission factors for NO_x and NO_2 for heavy-duty vehicles is afflicted with a greater uncertainty due to the low proportion of HDVs in the vehicular fleet. The share of HDVs never exceeded 15% during the measurement campaign, therefore extrapolating to 100% HDVs is very uncertain. For the same reason, it was decided to forego the calculation of the HDV emission factor for a traffic situation describe as fluid traffic (only 2,1% HDV).

The calculated fleet emission factor for NH_3 is much lower than it is given in HBEFA 3.3. On the one hand, measurements on which the modelling of the emission factor in HBEFA 3.3 is based on are few because ammonia is yet still a non-limited compound. On the other hand, the data set obtained from the conducted measurement campaign is also very meagre.
Separate emission factors for ammonia for passenger cars and HDVs could not be determined by linear regression analysis, due to the poor correlation (see Figure 27). However, a poor correlation between emission factor for ammonia and diesel trucks was also found by Liu T. et al. (2014) [43]. Thus, giving evidence that the share of heavy-duty vehicles in the vehicular fleet does not have a significant influence on the emission factor for ammonia, like it was observed for nitrogen oxides.
The wide range of emission factors for ammonia found experimentally may be explained by the variability of the passenger cars' composition during the measurement campaign regarding the fuel burned. Gasoline-powered vehicles emit more ammonia than diesel cars, as it was observed in other tunnel studies mentioned in section 7.2 as well as it is given in HBEFA 3.3. The ratio of passenger cars driven with diesel to ones driven with gasoline surely varied during the investigation period. Unfortunately, the provided traffic data only distinguishes between passenger cars and HDVs and gives no further information about the composition of the fleets' fuel types. It is assumed that the emission factor for ammonia is higher with higher share of gasoline-powered passenger cars in the vehicular fleet. However, no further data analysis for proving this assumption was possible.

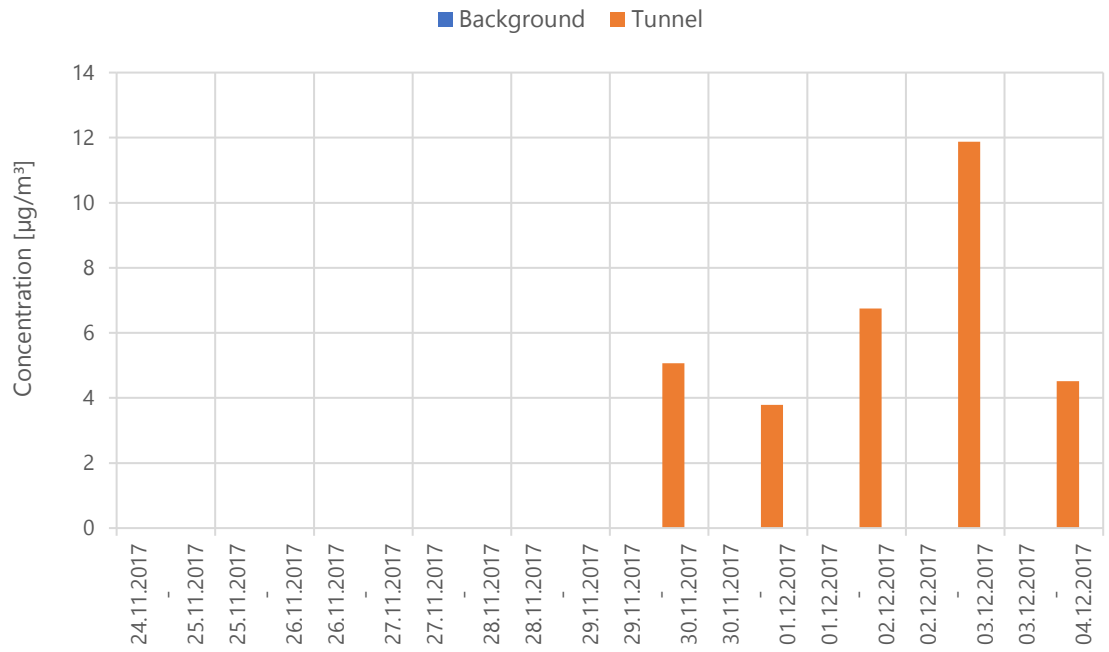
It has to be mentioned that the measurement site for determining background levels is not optimal chosen. According to Staehelin J. et al. (1997) [26] the sampling point should be right after the tunnel entrance. Thus, ensuring only emissions from vehicles going through the tunnel are considered for calculating emission factors. Ambient circumstances which may influence the emission factors are hence excluded.
For example, wind speed and its direction may affect the calculated emission factors. North-westerly wind is in line with the vehicles driving towards the tunnel. Exhaust gas emitted in front of the tunnel entrance may be drown into the tunnel, thus leading to a higher background level. In addition, stronger winds from north or north-east may blow the exhaust plume leaving the north-western bonded tunnel bore in southerly direction, where it could be sucked into the measuring bore due to the piston effect. Both effects lead to an overestimation of the emission factors, since these additional emissions are not excluded through the actual background measurement site "Am Kaisermühlendamm". In this case, the calculated emission factors for NO_x would be lower compared to the values given in HBEFA 3.3. However, it was not possible to investigate the influence of ambient wind, because appropriate wind data was neither measured nor available.

Within the analysis of particulate matter for the determination of particulate ammonium nitrate by suppressed isocratic ion chromatography, other water-soluble components are identified and quantified as well.
Concentrations of water-soluble sodium and chloride in particulate matter are quantified for both measurements sites. In Figure 36 it can be seen, that background levels of sodium are below the limit of detection (0,4 µg/m³) and chloride concentrations are below 0,4 µg/m³. Regarding the first measurement days inside the tunnel Kaisermühlen, sodium concentrations are also below the limit of detection (0,2 µg/m³) and chloride concentrations are relatively low. In the second half of the investigated period, concentrations of both analytes increase immense, due to application of deicing salt. However, this increase is not observed for background levels at the measurement site "Am Kaisermühlendamm".

This finding indicates, that the sampling point for background levels is not optimal for determining vehicular emission factors. Background concentrations right after the tunnel entrance may lead to lower emission factors. However, conducting measurements right after the entrance of tunnel Kaisermühlen was not possible.

Sodium and chloride in particulate matter

Sodium



Chloride

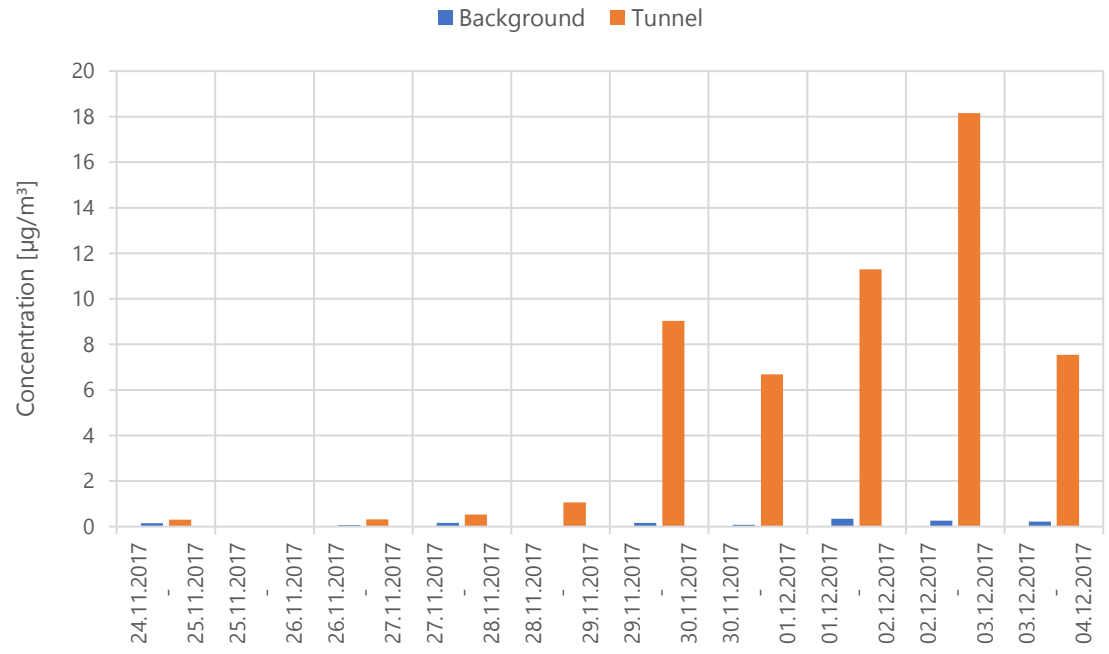


Figure 36: Concentrations of sodium and chloride in particulate matter in the south-eastern bonded bore of tunnel Kaisermühlen and at the background site "Am Kaisermühlendamm" from 24.11.2017 – 04.12.2017

9 SUMMARY AND OUTLOOK

The determination of vehicular emission factors for nitrogen oxides, nitrogen dioxide and ammonia through a measurement campaign conducted in the tunnel Kaisermühlen was performed successfully.
The emissions of those air pollutants are today of great concern, particularly since the manipulation of an exhaust after-treatment system, the selective catalytic reduction of nitrogen oxides via urea or rather ammonia, became popular. The principle of the urea-SCR system is presented in detail in section 2.3. Briefly, the aqueous urea solution is injected into the hot exhaust gas stream, where thermal decomposition takes place. In the next step, the formed ammonia acts as reducing agent for nitrogen oxides, which are generated during high-temperature combustion of fossil fuels, i.e. diesel fuel. The reduction reaction takes place on the surface of a catalytic converter.

A variety of parameters is required for calculating emission factors for an entire vehicular fleet. Therefore, measurements within a tunnel study were conducted in the period from October to December 2017. The tunnel of interest, tunnel Kaisermühlen, is 2.150 m long and consists of two separate bores, carrying the traffic in opposite directions on three lanes each.

The longitudinal air velocity inside the south-eastern bonded bore of tunnel Kaisermühlen was measured with an impeller anemometer, which was installed near the exit portal. Traffic data was provided by ASFINAG and included number of vehicles and average velocity for the entire fleet, as well as for passenger cars and HDVs separately.
Measurements of air pollutants were also carried out in the south-eastern bonded bore with the sampling point 1.644 m after the entrance. Nitrogen oxides were measured by continuous chemiluminescence analyser and discontinuous filter samples were collected for determining concentrations of gaseous ammonia (cellulose filters impregnated with oxalic acid) and ammonium nitrate in particulate matter (Teflon filters). The analysis of the extracted filters with suppressed isocratic ion chromatography took place in a laboratory of TU Wien.
Background levels of the investigated compounds were measured at "Am Kaisermühlendamm". The sampling methods were the same as for the measurements inside the tunnel.

Measured concentrations of nitrogen oxides and the longitudinal air velocity inside the tunnel Kaisermühlen showed pronounced diurnal trends and correlated very well with traffic density. Maximum values occurred with rush hours, whereas the lowest values were observed during night time. Average concentrations of nitrogen oxides and nitrogen

dioxide in tunnel air exceeded background levels 30 times and 9 times, respectively. Regarding the concentrations of gaseous ammonia inside the tunnel, a poor correlation with traffic number was found. Background levels were approximately 8 times lower. Concentrations of ammonium nitrate in particulate matter inside the tunnel increased about 10% compared to background levels. However, the concentrations were relatively low and were not further considered.

The calculated fleet emissions factors are 0,45 g NO_2/vkm and 78 mg NO_2/vkm for NO_x and NO_2, respectively. Two traffic conditions occurred inside the tunnel Kaisermühlen: "fluid traffic" and "saturated traffic". The main parameter influencing the traffic condition is the share of heavy-duty vehicles in the vehicular fleet. Consequently, separate emission factors for the two traffic conditions could be calculated. In addition, the emission factors showed a good correlation with the proportion of HDVs. Linear regression analysis provided separate emission factors for passenger cars and HDVs.
The calculated emission factors for NO_x and NO_2 were compared with corresponding values provided by the "Handbook Emission Factors for Road Transport" (HBEFA 3.3, 2017). Nitrogen oxides showed a good agreement, whereas for nitrogen dioxide lower emission factors were determined, except for HDVs, which were higher. These findings confirmed the emission factors given in HBEFA 3.3. Comparison with other tunnel studies found in literature showed good agreement as well. In Europe, a decrease of the emission factor for NO_x was observed which is most probably due to implementation of improved exhaust gas-after treatment systems for diesel-powered vehicles.

For ammonia, the calculated fleet emission factor is 4,7 mg NH_3/vkm. Whether a distinction into different traffic conditions nor a linear regression analysis for emission factor depending on the share of HDVs in the vehicular fleet was possible.
The fleet emission factor for ammonia provided by HBEFA 3.3 exceeded the calculated one more than three times. Other emission factors determined within tunnel studies found in literature were taken for comparison. It was found, that the present investigation conducted in tunnel Kaisermühlen gives the lowest fleet emission factor for ammonia. That is because the Austrian vehicular fleet comprises 60% diesel-driven vehicles, whereas the investigated fleets in other tunnel studies mainly consist of vehicles powered by gasoline. Gasoline cars emit much more ammonia than vehicles burning diesel fuel.

Although the emission factors for NO_x and NO_2 found experimentally agree well with them provided by HBEFA 3.3, it has to be mentioned that the measurement site for background levels was not positioned right after the tunnel entrance, as it should have been according to Staehelin J. et al. (1997) [26].
Ambient wind speed and its direction may have an influence on the concentration entering the tunnel Kaisermühlen. Winds coming from the north-west, north or north-east increase

the background levels, which are not considered at the actual measurement site "Am Kaisermühlendamm". Hence, the calculated emissions factors could be overestimated.

For further investigations, a measuring point right after the tunnel entrance should be installed. This would ensure, that only vehicular emissions emitted inside the tunnel are considered for calculating emission factors.

Regarding the determination of emission factors for ammonia, a longer sampling period would provide a bigger data set. Since no linear correlation between emission factor and the share of HDVs in the vehicular fleet was observable, the traffic data should contain more detailed information concerning on the fleets' fuel type. It is assumed that the emission factor correlates directly with the proportion of gasoline-powered passenger cars. However, for proving this assumption the appropriate information is needed.

10 LIST OF FIGURES

11 LIST OF TABLES

12 REFERENCES

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