Emissions of particle numbers in the Dutch Emission Inventory

TNO Intern ⟩ TNO 2024 R10541
26 April 2024

Energy & Materials Transition
www.tno.nl
+31 88 866 42 56
info@tno.nl

TNO 2024 R10541 – 26 April 2024

Emissions of particle numbers in the Dutch Emission Inventory

Author(s)	Sophie E.H. van Mil, Jeroen J.P. Kuenen, Antoon J.H. Visschedijk, Ruben Goudriaan, Emiel van Eijk, Jessica M. de Ruiter, Norbert E. Ligterink.
Classification report	TNO Intern
Title	TNO Intern
Report text	TNO Intern
Appendices	TNO Intern
Number of pages	30 (excl. front and back cover)
Number of appendices	1
Sponsor	RIVM
Project name	RIVM UFP inventory for ER
Project number	060.57656

Summary

Exposure to particulate matter has adverse impacts on human health. Recently attention for the health impacts of ultrafine particles (UFP) has been growing. To get a better insight into ultrafine particles and their possible impact on health, the Dutch Health Council ('Gezondheidsraad') advised to get a better understanding of UFP in a broad sense, including its sources. UFP are defined as particles with a diameter < 100 nm, or $PM_{0.1}$[1]. Due to their small size, the particles contribute little to total particulate mass. Therefore, the best way to estimate releases of these extremely small particles is by counting the number of particles emitted. However, particles above 100 nm, which are excluded from the UFP emission, also contribute to ambient particulate number and mass concentrations. They are therefore also relevant to take into account in the inventory, especially since size and composition of particles change upon release in the atmosphere.

This study provides first emission estimates of total particle numbers (TPN) in the Netherlands, as a part of the Dutch Emission Inventory. These total particle number (TPN) emissions include both solid and volatile particles. Furthermore, a distinction has been made between the total emitted number of particles TPN_{325} (number of particles up to 325 nm) and the part that is defined as ultrafine particles(TPN_{100}). An upper threshold of 325 nm is used, above which the total number of particles is negligible. The lower limit of 10 nm has been chosen because this will be the new lower limit for ambient air quality measurements, but also because it is increasingly difficult to measure emissions and concentrations of particles smaller than 10 nm. In addition their contribution to exposure is likely limited.

In former and ongoing European research projects, TNO has recently prepared first European emission inventories, which form a starting point for this study. Furthermore, emission for road transport, a key contributor to TPN emissions, are based on a methodology which takes into account emission measurements performed at TNO. Our results show that traffic and transport are the major sources for Dutch TPN emissions, with over 90% of the total emissions. Within the transport sector, sea shipping and aviation are the most significant contributors, followed by road transport and inland shipping. More than 90% of the TPN emitted is estimated to be smaller than 100 nm, hence within the definition of ultrafine particles.

The emission estimates in this report should be used with caution, as uncertainties for TPN are large. This study provides a first inventory, but future research is needed to improve emissions and decrease uncertainties. Next steps could include improving the understanding of the relation between solid and volatile particles, as well as improving insights of how these particles evolve and how to best quantify TPN emissions. Furthermore, the focus should be on improving emission estimates for major sources where emissions are obtained using generic methods, such as sea shipping and aviation.

[1] $PM_{0.1}$ refers to particle size expressed in µm (micrometer). Where 0.1 µm is equivalent 100 nm (nanometer)

Samenvatting

Blootstelling aan fijnstof heeft negatieve gevolgen voor de gezondheid. Recent is ook de aandacht voor gezondheidseffecten van ultrafijne deeltjes (UFP) toegenomen. Om beter inzicht te krijgen in deze ultrafijne deeltjes en hun mogelijke invloed op gezondheid, adviseerde de Gezondheidsraad om verschillende aspecten van UFP in kaart te brengen, waaronder de bronnen. UFP is gedefinieerd als deeltjes met een diameter <100 nm, oftewel $PM_{0.1}$[2]. Doordat ze zo klein zijn, dragen ze weinig bij aan de totale massa. De beste manier om het vrijkomen van deze extreem kleine deeltjes te schatten, is door het aantal geëmitteerde deeltjes te tellen. Deeltjes groter dan 100 nm, die dus niet onder UFP vallen, dragen echter ook bij deeltjes aantallen en massaconcentraties. Ze zijn daarom ook relevant om mee te nemen in de inventarisatie, zeker aangezien maat en compositie van deeltjes veranderen na het vrijkomen in de atmosfeer.

Deze studie geeft eerste emissieschattingen van het totaal aantal deeltjes (TPN, total particle number) in Nederland, als onderdeel van de Nederlandse Emissieregistratie. Deze deeltjesaantallen (TPN) emissies omvatten zowel vaste als vluchtige deeltjes. Daarnaast is ook onderscheid gemaakt tussen het totale aantal geëmitteerde deeltjes (TPN_{325}: aantal deeltjes beneden 325 nm) en het deel wat binnen de definitie van ultrafijne deeltjes valt (TPN_{100}). De bovengrens van 325 nm is gekozen omdat daarboven deeltjesaantallen te verwaarlozen zijn. Voor de ondergrens is 10 nm gekozen om praktische redenen (dit is de nieuwe standaard voor metingen in de buitenlucht, en daarbij is het heel lastig om voor de allerkleinste deeltjes emissies en concentraties te meten) maar ook omdat hun bijdrage aan blootstelling waarschijnlijk beperkt is.

In verschillende voltooide en lopende Europese onderzoekprojecten heeft TNO al eerste Europese emissie-inventarisaties gemaakt, die een startpunt vormen voor deze studie. Daarnaast zijn voor het bepalen van emissies van wegtransport, een belangrijke bron van TPN emissies, ook door TNO uitgevoerde emissiemetingen meegenomen. Onze resultaten laten zien dat verkeer en vervoer de grootste bron van Nederlandse TPN emissies is, met een bijdrage van meer dan 90% aan de totale emissies. Binnen deze sector zijn zeescheepvaart en luchtvaart verantwoordelijk voor de grootste bijdragen, gevolgd door wegtransport en binnenvaart. Meer dan 90% van de TPN emissieschattingen is kleiner dan 100 nm en valt dus onder UFP.

De emissieschattingen uit dit rapport moeten met behoedzaamheid worden gebruikt, aangezien onzekerheden van TPN emissieschattingen groot zijn. Deze studie geeft een eerste inventarisatie van emissies, maar toekomstig onderzoek is nodig om emissieschattingen te verbeteren en onzekerheden te verkleinen. Vervolgstappen omvatten onder andere het verbeteren van begrip van de relatie tussen vaste en vluchtige deeltjes, het beter begrijpen hoe deze deeltjes zich ontwikkelen, en hoe deze emissies het best te kwantificeren zijn. Daarnaast zou de focus voor het verbeteren van de emissieschattingen moeten liggen op de belangrijkste bronnen waarvoor nu nog generieke methodes gebruikt worden, zoals zeescheepvaart en luchtvaart.

[2] PM0.1 refereert aan deeltjes grootte uitgedrukt in µm (micrometer). 0.1 µm is gelijk aan 100 nm (nanometer).

Contents

Summary ... 3
Samenvatting ... 4
Contents ... 5
1 Introduction ... 6
2 Methodology ... 8
2.1 Transport ... 9
2.1.1 Road transport ... 9
2.1.2 Mobile machinery ... 11
2.1.3 Inland shipping ... 13
2.1.4 Railways ... 14
2.1.5 Aviation ... 14
2.1.6 Sea shipping ... 14
2.2 Other sources ... 15
3 Results ... 18
4 Discussion and next steps ... 21
5 Conclusions and recommendations ... 25
6 Bibliography ... 27
Signature ... 29

Appendix
Appendix A: Description of Electronic Appendices 30

1 Introduction

For many years it has been known that particulate matter in the atmosphere leads to adverse health impacts, and it is therefore important to mitigate its sources in order to reduce concentrations and therewith exposure to these small particles. In recent years however, it has become clear that a specific part of particulate matter, ultrafine particles or UFP, may be particularly relevant for negative impacts on human health. Ultrafine particles are defined as particles smaller than 0.1 µm. Since these particles are very small, their contribution to the particulate mass is typically negligible, but these particles may exist in large numbers. Therefore these are typically expressed in number of particles. This because the counting of numbers can be done much more accurate than weighing the mass, as is done for PM_{10} or $PM_{2.5}$. These particles are so small that they hardly have a mass.
To get a better insight into ultrafine particles and their possible impact on human health, the Dutch Health Council ('Gezondheidsraad') advised[3] to get a better understanding of UFP in a broad sense, which includes getting a better understanding of exposure to and health impacts of UFP, but also to reduce UFP emissions and where possible increase distance to sources. A key prerequisite for the latter is to get a good overview of the sources of UFP, for which the Health Council advised to include UFP in the Dutch Emission Inventory. This will facilitate a good understanding of the main sources of UFP, to quantify the sources with highest uncertainties and prioritize improvements, but also to monitor trends in UFP emissions over time.

In 2022 a preparatory study was performed by TNO[4], commissioned by RIVM, to propose an approach on how to incorporate UFP in the Dutch Emission Inventory. In the resulting note first rough estimates were given based on European research, and recommendations were given on how to best incorporate UFP in the Dutch Emission Inventory.

In multiple former and ongoing European research projects, TNO has prepared first European wide emission inventories for ultrafine particles, expressed as particle number, to support European scale modelling efforts for UFP. In the early 2010s the EUCAARI (Kulmala, 2011) and TRANSPHORM (Denier van der Gon, 2014) projects led to first particle number emission inventories (e.g. Paasonen, 2016), which are being updated in ongoing Horizon projects, such as RI-URBANS (Kuenen, 2022). These were for the European domain and covering the main anthropogenic emission sources for which also particulate matter emissions are estimated.

This study presents the first version of an emission inventory for ultrafine particles for the Netherlands, for the year 2022. It should be emphasized that with regard to emissions of ultrafine particles, there are major uncertainties, which are significantly higher than for typical air pollutants like NOx or PM2.5. This relates largely to the nature of ultrafine particles. These are small particles which may be emitted by sources, but immediately after emissions chemical reaction and physical transformation processes start which strongly affect concentrations of UFP in the short term.

[3] Risico's van Ultrafijnstof in de buitenlucht, Gezondheidsraad Advies Nr. 2021/38, https://www.gezondheidsraad.nl/documenten/adviezen/2021/09/15/risicos-van-ultrafijnstof-in-de-buitenlucht
[4] https://publications.tno.nl/publication/34640553/3jcCEh/kuenen-2023-ultrafijne.pdf

UFP is an overarching term for particles smaller than 100 µm. In practice, the emission of these particles includes solid particles, typically expressed as SPN (solid particle number), which includes the small particles already present in solid form in the exhaust gas at the time the hot flue gases exit the chimney or exhaust. At that time, however, the plume starts diluting and cools down, which typically triggers the generation of potentially many more particles, which are known as the volatile part. This part is especially difficult to quantify, since the actual processes creating volatile particles are in part dependent on the circumstances and hence measuring these emissions is more difficult to reproduce. The solid particle number on the other hand is a more stable parameter, which is therefore typically used in the type approval or testing procedures of new engines or vehicles. It is, however, important to stress that the population is exposed to the sum of both solid and volatile particles. While some of the volatile particles will have limited lifetime their contribution to total emissions is such that they cannot be neglected when in modelling concentrations of these particles, followed by exposure and health impact assessment.

The sum of the solid and volatile part is known as total particle number (TPN). We define TPN as the emission at the point of release into the atmosphere, but including the cooling and dilution until reaching ambient temperature conditions. Since TPN is the parameter that is in the end relevant for air quality and exposure, this is what we aim to estimate with this study, despite the uncertainties associated with this. This study is, therefore, clearly aimed to be a starting point, and TPN emission estimates provided in this study should be improved in the years to come. For clarity, in the remainder of this report we will therefore refer to TPN as the relevant substance instead of the more generic term of UFP. We further detail TPN in the following way:

- TPN_{100}: total number of particles in the size range 10-100 nm;
- TPN_{325}: total number of particles in the size range 10-325 nm.

While the total size range covered by this emission inventory is 10-325 nm, it should be noted that also particles smaller than 10 nm are emitted, in some cases in large quantities. These smaller particles are also recorded in UFP concentration measurements ambient air. These ranges are chosen as such mainly for practical reasons, since the smaller the particles become the more difficult, they are typically to measure. The 10 nm cut-off is in line with the latest ambient air quality measurement standards, and the latest motor vehicle standards; Euro-7, that are currently being agreed upon at European level, which also conforms to the 10 nm lower limit value. On the other side, the 325 nm upper limit is chosen because above this limit, the contribution of emitted particles to the total particle numbers is typically negligible (Harni S.D., 2023).Most of the mass of particles is typically above this size, but the number is not relevant anymore for these few large particles. TPN_{100} best matches with the definition of UFP, but for completeness as well as the interpretation of observations TPN_{325} is a valuable complementary source of information as it is considered to represent for TPN as a whole.

Based on the first version of the TPN emission inventory for the Netherlands presented in this report, future improvements can be prioritized based on the importance of the source and possible exposure (proximity of exposed people to sources).

In this report, the methodology of obtaining emission estimates are described in more detail in Chapter 2. This is split for different source groups, as methodologies vary. Results are discussed in Chapter 3. Chapter 4 includes the discussion and next steps. Chapter 5 concludes on the findings and provides recommendations for where the authors feel future improvements to TPN emissions should be targeted.

2 Methodology

The methodology for obtaining TPN emission estimates for the Netherlands is kept close to the approach common in the Dutch Emission Inventory. This way, easy uptake can be enabled and the format is kept similar. Furthermore, with high uncertainties of TPN emissions and ample improvements to be made, the focus of this study is to implement knowledge from previous studies. We aim to obtain best estimates for sources identified in earlier work as having high emissions, whereas more standard approaches are used for emission sources with lower TPN emissions.

In contrast to emissions of most other species in the Dutch national Pollutant Release and Transfer Register (PRTR) (hereafter referred to as the Dutch Emission Inventory), the TPN +emissions are not defined in mass, but as particle numbers. Both TPN_{100} and TPN_{325} are given in 10^{21} (Z, Zetta) particles.

Our emission estimate results are gathered and combined in a work field ("werkveld"), similar to all other Dutch Emission Inventory numbers. In this work field, which consists of an MS Access database, the table 'TPN_EMISSIES' contains all emission estimates. It also contains the compound definitions, dataset information, and fuel use per emission source and fuel type. Fuel use comes from the existing Emission Registration reporting and are in this study used as input for activity data when needed.

To align this inventory with the other Emission Registration work, an emission is obtained for each distinguished emission source ("emissie-oorzaak") and fuel type ("emissie verklarende variabele") for which there are relevant particle number emissions. In practice this means that emissions are obtained for each emission source ("emissie-oorzaak") for which particulate matter (PM) emissions are included in the Emission Registration.

Table EA.2 in the Electronic Appendix lists all emission sources considered in this study, together with the related sectors ("doelgroep"). These sectors are a way to group emission sources based on their origin. Each emission source is already assigned to a sector in the Emission Registration. As transport related sources are known for their key contributions to TPN emissions, we have split this sector into subgroups, e.g. for road transport, aviation, shipping. See Table EA.1 in the Electronic Appendix for the full split.

Methodologies for obtaining emission estimates may differ for various source groups. For sectors known to lead to high TPN emissions, more detailed methodologies were used to obtain emission estimates. For others, more general approaches using a set of emission factors and different types of activity data are used. In total, 8 different methodologies were used to obtain emission estimates. Table 2.1 shows which methodologies were used for which sector. Table EA.2 in the Electronic Appendix indicates the methodology used for each emission source and fuel type. For each sector, the related methodologies are explained in more detail below.

Table 2.1: Methodologies used to obtain emissions, for each sector group.

Sector group	Methodologies used
Transport	1 – Road transport exhaust 2 – Road transport brake wear 3 - Mobile machinery 4 – Inland shipping 5 – Railway 6 - Aviation 7 – Fuel based EF 8 - $PM_{0.95}$ based EF
Consumers	7 – Fuel based EF 8 – $PM_{0.95}$ based EF
Energy and industry	7 – Fuel based EF 8 – $PM_{0.95}$ based EF
Other	7 – Fuel based EF 8 – $PM_{0.95}$ based EF
Agriculture	Combustion emissions estimated using 7 – Fuel based EF Other emissions not estimated

For a number of emission sources, no TPN emission estimates were made. The nature of these emission sources is such that minimal to no TPN emissions are expected. This is the case for all sectors related to non-combustion sources in agriculture, as well as for example building fires, burning of candles, etc. Also, for some emission sources related to aviation, rail traffic, or road transport, no emissions were obtained. These are described in more details in the relevant sectors below.

2.1 Transport

2.1.1 Road transport

Emissions for road transport in the Netherlands are calculated bottom-up (Ligterink, Geilenkirchen, Eijk, & Ruiter, 2021). This means that the basis for the calculations is each individual vehicle that is registered. The emissions are calculated by multiplying the yearly activity of the vehicle (mileage or number of cold starts) with specific emission factors (emissions per km or per start). Activity data and Implied Emission Factors (IEF used for the calculation of wear emissions), aggregated per VERSIT+ vehicle category from the bottom-up calculation are used as input for the calculation of UFP emissions.

Particle emissions from road transport originate from four sources, i.e. tailpipe emissions, tire wear, brake wear and road wear. Tailpipe emissions are further subdivided in cold start emissions and emissions with a warm engine (Geilenkirchen, et al., 2023).

PN tailpipe emission factors for road transport are based on the current emission factors used for the calculation of SRM[5] emission factors (Eijk, Ligterink, Geilenkirchen, Ruiter, & Hoen, 2023). As exhaust emissions vary for different vehicles (ages, types, etc), the

[5] SRM: Standaard Reken Methode, official procedure to model air quality in the Netherlands, with definition for vehicle categories and driving behavior.

emissions are obtained on a more detailed level than per emission source and fuel type. These more detailed emissions are then later aggregated to obtain one TPN_{100} and one TPN_{325} emission for each emission source. The method used to obtain exhaust emissions, are referred to as *Method 1 – Road transport exhaust*. Cooling units of heavy duty vehicles are also considered, following this same method for road transport exhaust.

Emission factors used in prior analyses (Keuken, 2016), as well as new PN analysis (Ruiter J. d., 2024) and measurement results. UFP_{100} is approximated by

$$TPN_{100/325} = PN_{10}(\sim PN_{23}) + PN_{volatiles}$$
$$= (1 + f_{<23nm})\left(\frac{PM}{EC}\right)PN_{23} + \left(\frac{PN_{23}}{PM}\right)THC$$

where PM, EC, THC the relevant VERSIT+ road-type dependent emission factors in g/km, PN_{23} the particle number larger than 23 nm as currently measured for vehicle legislation, and $f_{<23nm}$ the estimated increase with respect to PN_{23}: 0.3 for diesel and 0.5 petrol (European Commission; Samaras & et al., 2020). With the pending transition from the lower limit for automotive measurements in Euro-7 from 23 nm to 10 nm, there has been a number of underlying comparative studies. Only for the highest particulate matter mass emissions, e.g., PM > 300 mg/kWh, there is a substantial part of the particles in accumulation mode, resulting in a difference between TPN_{100} and TPN_{325}.

Cold start emission factors are calculated by multiplying the cold start PM emission factor with the PN_{23}/PM fraction per VERSIT+ class.

For petrol vehicles the TPN_{325} emission factor is assumed to be equal to the TPN_{100} emission factor as nearly all particles are <100 nm. For diesel, over half of the emissions are found in the nucleation mode (~10nm) and a fraction of the accumulation mode (~70nm) above 100 nm (as can be seen in the size distribution in Figure 2.1). (Eastwood, 2008)
Therefore, only for diesel, TPN_{325} emissions are assumed to be 15% higher than TPN_{100} emissions.

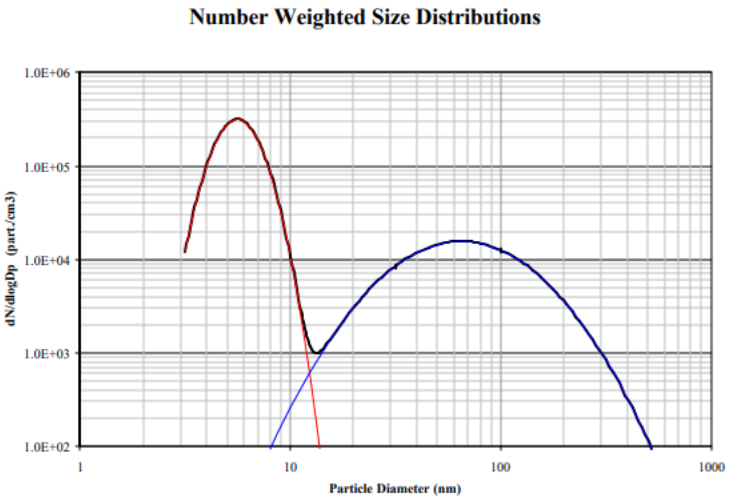

Figure 2.1: Size distribution diesel particles (Kittelson, Watts, Johnson, Baltensperger, & Burtscher, 2003)

Calculation of TPN based on HC (hydrocarbon) emissions in some cases leads to very high values, especially for older vehicles because of their high HC emissions. In the hot exhaust gas, the hydrocarbons are still gaseous, but they condensate directly after the exhaust line, when the exhaust gas cools in ambient conditions. These simple estimates, adding volatiles to non-volatiles, based on the mass ratio, are not realistic and emission factors are therefore truncated to maximum concentrations (see the explanation in 0). For high HC emission the process is expected to be condensation onto existing particles, thus not increasing particle numbers (Kittelson, Khalek, McDonald, Stevens, & Giannelli, 2022).
Table 2.2 shows the maximum values for different vehicle categories.

Table 2.2: Maximum TPN_{100} tailpipe emission factors

Vehicle category	Maximum emission factor [#/km]
Mopeds, motorcycles and other lcat vehicles	1 x10^{13} #/km
Passenger cars and vans	5 x 10^{13} #/km
Trucks and tractor-trailers	2 x 10^{14} #/km

Volatile particle emissions for vehicles on CNG, LNG and LPG are assumed to be zero as these fuels are gaseous in outside air and do not form particles, based on the vapour pressures. Such engines have particle emissions mainly associated with the burning of lubrication oil, and thus dependent on the type of oil, which follows the same trend as other engines, which all have the burning of lubrication oil (Samaras & et al., 2020).

Tire wear and road wear from road transport are not estimated in this study. Both mainly contribute to emissions of coarser particles and therefore TPN emissions of tires and road are expected to be of minor importance. TPN is in the extreme tail of their size distributions.

Brake wear emissions are a complex problem with far more and smaller particles, especially when the brakes warm up. At lower temperatures about 5x10^9 particles are measured per 1 mg of particulate matter (J. Wahlström, 2010). At higher temperatures this can increase to a factor 100 higher particle numbers per mg (we experience this in our own measurements as well). Assuming that about 2% of the mass is generated in higher temperatures, that leads to a conversion factor of 0.98x5x10^9 + 0.02 x 5 x 10^{11} = 1.5 x 10^{10} #/mg based on the PM2.5 wear emission factors for TPN_{325}. For TPN_{100} a factor of 1.2 x 10^{10} #/mg is assumed based on the particle size distribution. This method is referred to as *Method 2– Brake wear*.

2.1.2 Mobile machinery

TPN emissions for Non Road Mobile Machinery (NRMM) largely depend on the presence of a particulate filter. Especially smaller NRMM have no PN_{23} limit values which implies that from legislation perspective, there is no incentive to install a particulate filter.

For diesel engines without particulate filter we typically see maximal values of 10^7 #/cm3 or 10^{10} #/l (ambient conditions), regardless of the source (road or NRMM). This is caused by the fact that particles clump together based on the square of the given concentration (Smoluchowsk equation) (Eastwood, 2008). The residence time of gases in the exhaust line determines the concentration. (Hinds, 1999). For higher mass (PM2.5), the particle size, or the fraction in accumulation mode around 100 nm, increases. Basically, more particles are produced in the diesel engine itself, but they are coagulated, condensed and accumulated in larger particles.

Diesel engines typically have a range of a factor two in engine speed between the lowest and highest engine load, and thus in mass flow. Depending on turbo and EGR, that means that there is a factor 2-3 in volume flow between the lowest and highest engine power.

Lots of diesel engines without particulate filter (especially smaller NRMM) are oversized for the power they need to deliver (1.6-2 litre engines for 20kW are not uncommon). For larger engines the engine size increases slower with the engine power (typically 40kW per litre). Therefore, the engine size in litres is estimated by 1+0.025 x P_{rated}[kw].

Particle numbers for engines with high particulate matter emissions are mainly a function of exhaust volume flow, thus proportional engine size times engine speed. With higher particulate mass, the particles are larger, not so much higher in number. Hence, the determination below, is based on this principle.

Diesel engine speed is typically between RPM = 800 and 2400 [min^{-1}], and the filling degree, i.e., the amount of inlet air related to the engine volume, is 90%. Volume flow can thus be calculated as:

$$Q\left[\frac{liters}{hour}\right] = 60\ [min] * 0.9 * (1 + 0.025 * P_{rated}) * \frac{RPM}{2} = (27 + 0.067 * P_{rated}) * RPM$$

This lead to a TPN emission factor, depending on the requested power, for “land based” mobile sources without particulate filter of:

$$TPN_{100}\left[\frac{\#}{hour}\right] = 10^{10}\left[\frac{\#}{liter}\right] * Q\ \left[\frac{liters}{hour}\right] = 2.7 * 10^{11} * RPM + 6.75 *\ 10^{9} * P_{rated} * RPM$$

RPM does not directly correlate with requested power, for example during accelerations and hydraulics where engine torque can very strongly, but by approximation:

$$TPN_{100}\left[\frac{\#}{hour}\right] = 2 * 10^{14} + 4 * 10^{14} * \frac{P_{demand}[kW]}{P_{rated}[kw]} + 5 * 10^{12} * P_{rated}[kW] + 10^{13} * P_{demand}[kW]$$

Where 0 < P_{demand}[kW]/P_{rated}[kW] < 1 is the fraction requested engine load.

Many of the smaller diesel engines have a large engine volume compared to the power they supply, and, therefore, a relatively high exhaust gas rate. For larger engines, the engine volume is about 40 kW per litre engine. Therefore, the equation $Engine_{lires}$ = 1 + 0.025 * P_{rated}[kW] is used. Moreover, larger engines run slower, thus have a more limited increase in exhaust mass flow, compared to the rated power (P_{rated}) and power demand. See above.

For larger engines (>56kW) an approximation in the following form suffices:

$$TPN_{100}\ \left[\frac{\#}{hour}\right] = (5 * 10^{12} + 10^{13} * Load[\%]) * P_{rated}\ [kW]$$

The EMMA model uses engine maps to calculate emissions for different NRMM types (Dellaert, 2023). Based on the formulas given above, TPN_{100} emission factors have been estimated for the requested fields. They are summarized in table below (Table 2.3**).**

Table 2.3: The emission maps of a NRMM engine, with emission rates for different engine loads, proportional to the rated power.

Engine size	Load	0%	10%	20%	30%	40%	50%
>56 kW	#/s*P_{rated}	1.39E+09	1.67E+09	1.94E+09	2.22E+09	2.50E+09	2.78E+09
<56kW	#/s*P_{rated}	2.78E+09	3.33E+09	3.89E+09	4.44E+09	5.00E+09	5.56E+09
		60%	70%	80%	90%	100%	
>56 kW	#/s*P_{rated}	3.06E+09	3.33E+09	3.61E+09	3.89E+09	4.17E+09	
<56kW	#/s*P_{rated}	6.11E+09	6.67E+09	7.22E+09	7.78E+09	8.33E+09	

Diesel engines with particulate filter emit around 100.000 #/s*$P_{rated.}$ (Ruiter & Mensch, 2022). These are typical values from Dutch Periodic Technical Inspection (PTI or 'APK')) and PEMS-PN testing of diesel engines under load. (Kadijk, Elstgeest, Mark, & Ligterink, 2020) If the engine is idling, the values are lower, however, more regenerations with associated particle emissions may occur when the engines are operated much at low engine loads. However, given the insignificant values, these distinctions are irrelevant.

As for road traffic, 15% extra is added for TPN_{325}.

Since these diesel engines, with high air-fuel ratios emit mainly EC, and little OC or VOC, the formulas should be used without modifications for volatiles.

The method to obtain TPN emission estimates for mobile machinery as described here is referred to as *Method 3 – Mobile machinery.*

2.1.3 Inland shipping

For inland shipping and sea vessels, the formulae from Section 2.1.2 can be used with a factor 5 increase for high-speed engine and a factor 10 for low-speed (two-stroke) engines, due to a shorter residence time of the engine gas in the tailpipe itself. This only holds for engines with PM emission limits above 0.3 g/kWh and NO_X > 8 g/kWh, or no aftertreatment. With aftertreatment the formulae can be used directly.

With the lack of detailed information on engines and engine loads the current ratio of CO2 (424 kton) and PM2.5 (146 ton), and the typical CO2 concentration of 5% in exhaust gas, it translates to 0.34 g/kg CO2. In one litre exhaust gas for CCR0 and CRR2 , with little aftertreatment and residence time in the exhaust line, is 10^{11}. For newer CCR2 the value is 10^{10}. It is expected that CCR0 and CCR1 are dominant for UFP of inland shipping, and this yields with typical exhaust gas composition a ratio of **TPN/PM2.5 = 3 x 10^{15} [#/g]**. Particles are fine and therefore TPN_{100} = TPN_{325}.

The use of this PM2.5 based emission factor to obtain emissions is referred to as *Method 4 – Inland shipping*.

2.1.4 Railways

There are about 80 diesel-fuelled trains in use for passenger transport in the Netherlands and a similar amount of diesel locomotives. Passenger trains have some sort of aftertreatment and emit 10^{10} #/liter in exhaust gas, diesel locs do not and emit 10^{11} #/liter. Passenger trains have 2 x 15 = 30 liter engine and diesel locs 100 liter engines. They operate typically 10 hours per day and 250 days per year. This lead to:

- Passenger trains: 1.8 x 10^{21} #/year
- Diesel locomotives: 6 x 10^{22} #/year

These numbers represent total #/year, aligning with TPN_{325}. 62% of this is considered to be TPN_{100}.
This methodology is referred to as *Method 5 – Railway*.
Diesel locomotives are represented by emission source 0200100 'Exhaust gas, rail traffic – cargo'. Passenger trains are represented by emission source 0200300 'Exhaust gas, rail traffic – passengers'.
For all other rail related emission sources, such as wear and wheel dust, no PN emissions are estimated.

2.1.5 Aviation

In a turbine engine, e.g. for airplanes, the residence time is very short and most particles are still around 10 nm (limited accumulation). The concentrations are around 5 * 10^{11} #/liter. There is more air per kg fuel for jet engines, especially for low loads this leads to more particles per kg fuel. Therefore, airport remote sensing measurements (PTU, taxiing) can lead to overestimations of average UFP emissions per kg fuel in normal use. A typical value is 10^{16} #/kg fuel.
The short residence time and high concentration of small particles makes that the emissions from aircraft are difficult to quantify. Recent estimates show that the emissions of total particles (including the volatiles) may be as much as a factor 20 higher compared to only the solid particles from aircraft (Takegawa, 2021), however other estimates point to a lower share. Given the uncertainties around this, it was decided not to use such a factor to scale up the solid particles (which are already included in the CLEO emission model used in the Dutch Emission Inventory). Instead, it was chosen to apply an emission factor per LTO (Landing and Take-off cycle) which was previously used in European research studies, which is 1.2 x 10^{19} #/LTO which represents a more conservative approach with regard to the amount of volatile particles added (Denier van der Gon, 2014).
The methodology for this LTO based approach of obtaining TPN emissions for aviation, is referred to as *Method 6 – Aviation*.

Since no information was available for particle emissions from cruise flight, and also since these are not directly relevant for concentrations in the Netherlands, TPN emissions from cruise flight are not estimated. In addition, the emission factor applies to jet engines, which are believed to represent the vast majority of TPN emissions from air transport. Emissions related to piston engines are estimated using a $PM_{0.95}$ emission factor approach *(Method 8 – $PM_{0.95}$ based EF)*. However, these are expected to be minor with respect to jet engine emissions. Piston engines are used in small airplanes, which represent only a small fraction of Dutch air transport.

2.1.6 Sea shipping

Emissions from sea shipping are a major source of TPN emissions. A recent European research project (SCIPPER) provided an updated set of emission factors for main pollutants and also TPN for shipping in Europe based on an extensive literature review (Grigoriadis, 2021). An emission factor of 3.58 x 10^{15} #/kWh is provided as base emission factor for TPN at

European scale for liquid fuels (diesel-like). Assuming an efficiency of ~30% this would imply an emission factor of 3.0 x 10^{14} #/MJ. Similarly, for LNG an emission factor is provided at 2.3 x 10^{12} #/kWh, which would be equivalent to 1.9 x 10^{11} #/MJ when assuming the same 30% efficiency.

As means of a cross-check, these are compared to other estimates which were made in earlier EU research projects. Here, the emission factors were estimated dependent on the sulphur content of the fuel. This is important, since the sulphur content of the fuel is an important factor with a strong correlation to the amount of particles formed. The range provided here for shipping EFs (all liquid fuels) is between 16 and 50 x 10^{13} #/MJ (Visschedijk, 2022). Here, the lowest value is representative for fuels with a sulphur content < 0.1%, which would be the most representative for the North Sea which – together with the Baltic Sea – has the most strict limit values with regard to sulphur in Europe. This EF is around half the EF quoted from the SCIPPER study, which might be explained by the fact that this is from a European project which also included the Mediterranean region where a less strict sulphur regime applies. Hence, the lower emission factor is believed to be more representative for the Netherlands and North Sea at this point and this is the EF used for liquid fuels. For LNG, the SCIPPER emission factor quoted earlier in this section is used in the absence of an alternative emission factor.

For sea shipping, for all fuels, a direct emission factor based on fuel use is used to obtain emission estimates. This method is referred to as *Method 7 – Fuel based EF*. This methodology is described in more detail below, in 2.2 Other sources.

2.2 Other sources

For other sectors, relatively straight-forward methods were used to obtain emission estimates. As shown in Table 2.1, these consist of *Methodology 7 – Fuel based EF* and *Methodology 8 – $PM_{0.95}$ based EF*. For both cases, an emission factor approach is used, where an emission factor is multiplied with relevant activity data in order to get to a yearly total emission estimate. For these two methods, different types of activity data and corresponding emission factors are used.

The emission factors used in this study all originate from literature review. Earlier EU projects EUCAARI (Kulmala, 2011) and TRANSPHORM (Denier van der Gon, 2014) first European-wide emission inventories for TPN were made on this basis. This work originates from ~10 years ago and given technological developments these may therefore be on the high end of the range for a number of sources. Emission factors were collected for main sources, but it was quickly found that TPN emission factors are generally scarcely available. Therefore, an alternative method was developed by looking at $PM_{2.5}$ mass, estimating the smaller fraction therein (first $PM_{0.95}$ and then down $PM_{0.3}$, representing the PM mass below 0.95 µm and 0.3 µm, respectively) and then estimating the number of particles therein by means of a literature-based or assumed size distribution. The latter is mainly used for small sources, where a simple method suffices, but also for industrial sources where different processes causing both mass and number emissions are happening at the same time. By applying specific knowledge of the processes in each industry, a decision is made on which size distribution is the most suitable.

Table EA.2 in the Electronic Appendix gives an overview of the emission sources for which these methodologies are considered to obtain TPN emission estimates. For relevant emission sources where an emission factor approach is used (*Methodology 7 – Fuel based EF* or *Methodology 8 – $PM_{0.95}$ based EF*), the emission factors are also included here. Emission sources are often more detailed than TPN emission factors. These emission factors represent more general processes. Therefore, emission factors often applied to multiple emission sources. To link these two, assumptions have been made on the main processes prescribed in an emission source. Table EA.3 in the Electronic Appendix gives an overview of the various emission factors considered in this study. It includes a more general description of the process captured by this emission factor. This gives more insight into why emission factors are used for certain emission sources. Each emission factor is given a short label, which is also referred to in Table EA.2, allowing to cross-check which emission factor is used for which emission source, and to see which assumptions have been made when linking an emission factor to each emission source. Furthermore, Table EA.3 indicates in which project each emission factor was obtained; EUCAARI (Kulmala, 2011), TRANSPHORM (Denier van der Gon, 2014), RI-URBANS (Kuenen, 2022), or TNO study for UFP in the Rijnmond region (Visschedijk, 2022).

Methodology 7 – Fuel based EF

For some emission sources, a direct emission factor is considered, with the fuel use *('Emissie verklarende variabele hoeveelheid')* as activity data.

$$TPN_x\ emission = EF_{direct,x}\ \cdot\ fuel\ use$$

Where

- TPN_x is TPN_{100} or TPN_{325} in Z (10^{21}) particles.
- $EF_{direct,x}$ is a direct emission factor for TPN_{100} or TPN_{325}. Such an emission factor is considered for each fuel type and emission source combination. This emission factor is in 10^{16} particles/MJ fuel use.
- Fuel use is fuel use in MJ. This is also defined per emission source and fuel use in the Emission Registration. For this study, we have used the fuel use reported in the Dutch Emission Inventory for the year 2022 based on the final figures (Dataset "ER 1990-2022 Definitief").

Methodology 8 – $PM_{0.95}$ based EF

In other cases, the particle number emissions are deduced from the PM emissions. An indirect emission factor is then used, for which activity data is often $PM_{0.95}$ emissions (total mass of emitted particles smaller than 0.95 µm). Both the emission factor and $PM_{0.95}$ emission are defined for each combination of emission source and fuel type.
Therefore, for each emission source and fuel type, a specific TPN emission estimate is obtained as follows:

$$TPN_x\ emission = EF_{indirect,x}\ \cdot\ PM_{0.95}\ emission$$

Or

$$TPN_x\ emission = EF_{indirect,x}\ \cdot\ PM_{10}\ emission\ \cdot fraction\frac{PM_{0.95}}{PM_{10}}$$

Where

- TPN_x is TPN_{100} or TPN_{325} in Z (10^{21}) particles.
- $EF_{indirect,x}$ is an indirect emission factor for TPN_{100} or TPN_{325}. This is specific for a fuel type and emission source combination. This emission factor is in $10^{13}\ particles\ /\ kg\ PM_{0.95}$.
- $PM_{0.95}$ emission is the yearly total emission of $PM_{0.95}$. This can be obtained my multiplying the PM_{10} emission from the emission registration by a fraction for $PM_{0.95}/PM_{10}$. The PM_{10} emissions used are those reported in the Dutch Emission Inventory for the year 2022 based on the final figures (Dataset "ER 1990-2022 Definitief"). The fractions to obtain $PM_{0.95}$ can be found in Electronic Appendix Table EA.4. These are also based on existing fractions from the Dutch Emission Inventory.

$$PM_{0.95}\ emission =\ PM_{10}\ emission\ \cdot fraction\frac{PM_{0.95}}{PM_{10}}$$

Industry – 'NACE' ('ERI')
For industry, emission estimates in the Emission Registration are partly based on annual emission reports (AER) by individual facilities. This are included in the so-called 'NACE' ('ERI'). Emission factors related to this are also treated separately.

For the ERI emission sources, it was decided to obtain emissions on the level of the emission sources, and not obtain company specific emissions. Because of the high uncertainty of TPN emission estimates, it is more fitting to obtain estimates on an aggregated level rather than per individual company. However, this does imply that spatial distribution of the emissions is not possible for now.

Also, for industrial emissions, there is often not a clear split between process or combustion emissions. For these emissions reported by companies, the emissions are labelled as combustion if there is fuel used by the installation from which the emissions occur. However, the emissions reported may actually represent a sum of process and combustion emissions. For many of these emission sources related to industry, we use indirect emission factors and therefore reported PM emissions as activity data. However, if the reported PM emissions also include a significant contribution from process-based emissions, application of a fuel-based emission factor will lead to erroneous PN emission estimates. To overcome this issue, an implied emission factor was obtained and used to judge whether the PM emissions are mainly combustion- or process-based. These implied emission factors were obtained for natural gas and PM_{10} emissions. When this check indicated that PM emissions were too high based on the natural gas consumption, it was assumed process-based emissions play a key role in this process and a process-based EF was chosen for this emission source. This was then done for all fuel types linked this emission source, as it was assumed that then process-based emissions were dominant for all fuel types. When PM emissions were in the expected range for natural gas combustion, it was assumed that this emission source mainly contained combustion emissions, and therefore the relevant TPN emission factor for the combustion process was used.

As was explained before, already existing emission registration fractions of $PM_{0.95}/PM_{10}$ per fuel type and emission source were used to obtain $PM_{0.95}$ emissions (for these fractions, see EA.4). For emission sources for which emission estimates were obtained using *Methodology 8 – $PM_{0.95}$ based EF*, these $PM_{0.95}$ emissions are needed as activity data. However, when industries report both PM_{10} and PM2.5 emissions, both emissions are included in the ER. However, the $PM_{2.5}/PM_{10}$ fraction as follows from reported emissions may not align with the fractions in the ER. This as fractions in the ER are more general and may vary per industry. In some cases, this leads to complications, as now using PM10 emissions to obtain $PM_{0.95}$ lead to occurrences where $PM_{0.95} > PM_{2.5}$. For these cases, $PM_{0.95}/PM_{2.5}$ fractions were obtained and $PM_{2.5}$ emissions were used to obtain $PM_{0.95}$. These fractions are also included in Table EA.4.

3 Results

Table 3.1 shows TPN emissions for the different sectors (“Doelgroepen”). These sectors are a way to group emission sources based on their origin. Each emission source is already assigned to a sector in the Emission Registration. It becomes clear that the main source of TPN emissions in the Netherlands is traffic and transport, which is found to account for 91% of the TPN emissions when considering the size range up to 325 nm. When zooming in only on particles below 100 nm, the share of traffic and transport in total TPN emissions increases to 97%, since especially for combustion.

Table 3.1: TPN emissions for all source groups ('Doelgroepen')

Doelgroep	TPN_{100} (10^{21} #)	TPN_{325} (10^{21} #)
Agriculture	122	468
Chemical industry	2.7	17
Construction	4.5	17
Consumers	206	1188
Drinking water supply	0.1	0.2
Energy sector	9.8	18
Other industry	294	463
Refineries	131	144
Sewage systems and waste water treatment plants	1.7	1.8
Trade, Services, and Government	29	106
Traffic and transport	23504	23906
Waste disposal	5.5	14
Total	**24310**	**26343**

The TPN emissions are shown again in Figure 3.1 but now also including $PM_{2.5}$ emissions from the Dutch Emission Inventory in the same plot. In these figures the traffic and transport sector is split in the main subsectors, while some smaller source groups which are lumped together. It shows that the main contributors are sea shipping[6] and aviation, which account for 44% and 25% of TPN_{325} emissions, respectively. The other main contributors are road transport (12%) and inland shipping (10%).

Figure 3.1 also shows the corresponding $PM_{2.5}$ emissions as reported in the Dutch Emission Inventory for the year 2022 (based on Dataset “ER 1990-2022 Definitief”). Here it can be clearly observed that the main sources of particle number emissions are different from the main sources contributing to the PM mass. For PM mass, the main contribution is from consumers (incl. wood combustion) and energy and industry sectors which together represent more than half of the Dutch $PM_{2.5}$ emissions, however for TPN_{325} their combined

[6] Sea shipping includes all shipping activities happening on the Dutch Continental Shelf (or “Nederlands Continentaal Plat”), so also including ships that do not depart from or arrive in the Netherlands

contribution is only 7%. For mobile sources on the other hand, the contribution to total $PM_{2.5}$ emissions is 36% while for TPN_{325} the transport sectors contributes more than 90%.

The uncertainties of the estimated emissions have not been estimated at this point. However, as mentioned earlier in this report uncertainties for TPN are higher than for main air pollutants like PM. Since in many cases available measurements are scarce, there is little information to base uncertainty estimates on.

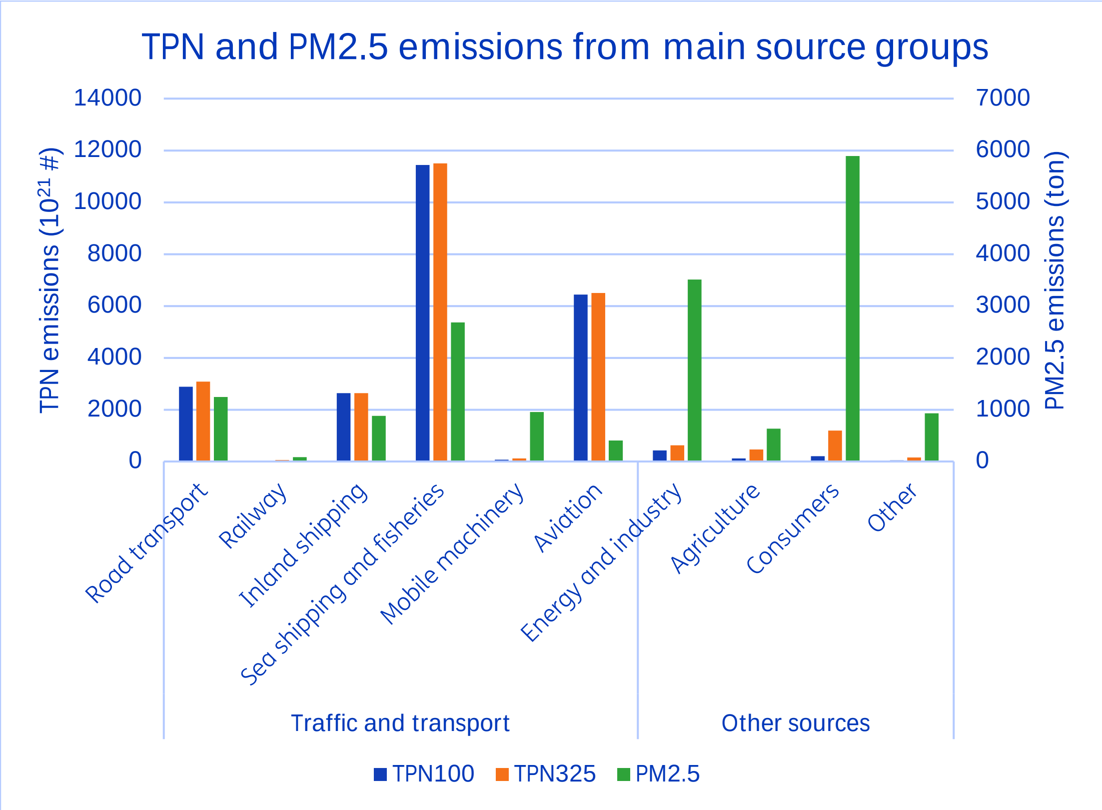

Figure 3.1: TPN emissions (below 100 and 325 nm, respectively) and PM2.5 emissions as reported in the Dutch Emission Inventory for the main contributing sectors to TPN emissions

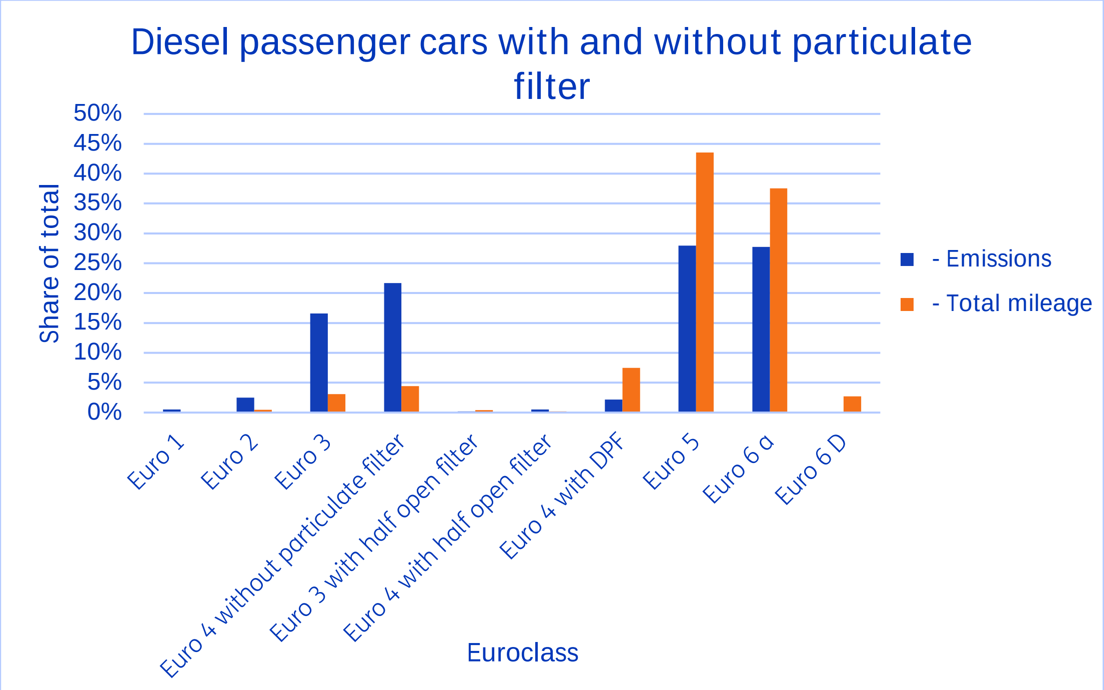

Figure 3.2: Share in mileage and TPN100 emissions of diesel passenger cars

Figure 3.2 partially shows the effect of particulate filters in TPN emissions of diesel passenger cars. The figure shows that diesel passenger cars without particulate filter (the 4 left most vehicle types) account for 8% of the total mileage of diesel passenger cars but 41% of the TPN emissions. On the other hand, vehicles with a particulate filter have a similar contribution to the total TPN emissions, but a significantly higher share in the vehicle kilometres. Note that the TPN emissions for vehicles with a particulate filter mainly consist almost exclusively of volatile particles, as emissions of solid particles are negligible.

4 Discussion and next steps

This study presents a first emission inventory for total particle numbers (TPN) for the Netherlands. It is also (one of) the first of its kind in Europe. TPN is further specified in TPN_{100} and TPN_{325}. TPN_{100} is the closest match with Ultrafine particles (UFP). The novelty of an emission inventory for TPN also implies that specific methodologies and emission factors are only scarcely available. Assumptions and expert judgement have been used to fill the gap for those sources for which no information was available. This allowed to arrive at a first complete emission inventory for TPN, for which the year 2022 has been selected.

Emissions were estimated for each individual emission source ("emissie-oorzaak") included in the Dutch Emission Inventory. Some sources were not estimated for now but in all these cases only negligible emissions of TPN are expected. This because no combustion or high temperatures is involved in the respective source. These include for instance agricultural sources (excl. fuel combustion).
The main goal of this first inventory is to be able to quantify what the main sources of TPN emissions in the Netherlands are, and to be able to prioritize, on that basis, where improvements to current emission estimates are most needed.
In addition, it is foreseen that TPN emissions will continue to be monitored in upcoming years. This allows monitoring of emissions and trends therein over time. It also provides input to modelling exercises which - combined with measurements of ultrafine particles – provides a framework to assess and improve the understanding of the exposure to ultrafine particles in the Netherlands.

A number of important considerations should be made with regard to monitoring of TPN emissions.

- For monitoring particle emissions from specific installations or engines in legislation, in most cases SPN (solid particle number) is used as the metric, since a SPN measurement is more easily quantified and reproducible. Hence, it would have been far more straightforward to derive a full emission inventory for SPN. However, one of the key goals of this emission inventory is to deliver particle emissions relevant for air quality and exposure as input to the monitoring and assessment of particle number concentrations near the sources and in the Netherlands as a whole. For this purpose, considering TPN – so including the volatiles – is a necessity.
- In relation to the above, small particles (especially those in the ultrafine range) are constantly evolving in the atmosphere. After the exhaust or stack exit, in addition to the solid particles already present in the plume, volatile particles are formed upon cooling and dilution of the exhaust gases, which are included in the definition of TPN. However, that is not the end of the story, as the particles travel further they agglomerate or otherwise evolve. These processes also contribute to the difficulty in quantifying TPN emissions. The number of particles is gradually reduced, dependent on circumstances but also the type and size of the particles concerned. Typically the smallest particles agglomerate together first, hence a high number emission of small particles may be reduced to a much smaller number of larger particles in a short time frame. These processes are not in the scope of this emission inventory, however they are of crucial importance to be considered when using the results from this emission inventory in modelling studies. To estimate exposure, the presented TPN emissions should be

processed with dispersion models which incorporate schemes to account for the evolution of TPN numbers after release in the atmosphere.

Results

The results show that the main sources of TPN emissions in the Netherlands in 2022 are in the transport sector, which constitutes more than 90% of total TPN_{325} emissions in the Netherlands in 2022. This is in contrast with particulate mass emissions, where the transport sectors accounts for 36% of $PM_{2.5}$ emissions, and consumers and industry are also important contributors.
The largest contributing sectors to TPN emissions typically generate a lot of very small particles (in the lower size range, typically 10-20nm). Given their small size these particles only contribute a small fraction to the particle mass. This is especially true for the two main contributors to TPN emissions in the Netherlands, aviation and shipping, but holds also for some other sectors. On the other hand, some sources like wood combustion contribute substantially to total particulate matter (PM10) and thus also emit particles, but these are typically larger in size. This implies that for reducing particle emissions, depending on whether the focus is on mass or number of particles or UFP the sectors to be targeted likely differ.

Methodological issues and uncertainties

As mentioned above, this TPN inventory is a novel development, which implies that applicable methodologies and emission factors to estimate emissions for each source do not exist in many cases. Different approaches have been used to quantify TPN emissions for all sources, to get a complete overview for the Netherlands.

- For most transport sources (excl. aviation and sea shipping) the amount of particles was estimated based on emission factors of solid particle number (SPN) as a basis, to which estimates of volatile particles and particles below 23 nm are added based on other emitted substances (mostly EC and organic substances). The advantage of this approach is that it constitutes a consistent method which builds on a large set of emission measurements. This approach to estimate TPN is based on a theoretical relation which is yet to be verified in real-world situations, especially for the unknown condensation mechanisms.
- For a number of sources, direct emission factors have been collected from literature. These typically include the more significant sources of particles, such as aviation, sea shipping, wood combustion and diesel combustion in various appliances. Typically only one emission factor is available which is assumed to represent the average technology level in the Netherlands.
- For most industrial sources and other sources, generally TPN emissions are relatively small, and in many cases no emission factors are available. Therefore, a specific methodology was developed to derive TPN emissions from PM mass emissions by first estimating the PM mass fraction below 300 nm, and then use a selected size distribution from literature combined with an assumed density to estimate the number of particles. This is generally a more uncertain method but need to be used in the absence of other options. For industrial emissions however this method is more suitable since combustion and process emissions are often mixed. By looking at the PM mass emissions combined with knowledge on the process causing the emissions, a first estimate can be made.

The following list provides a first suggestion for improvements

- Aviation and sea shipping are both key sources of TPN_{325}, together responsible for two thirds of the total estimated TPN emissions in the Netherlands in 2022. For both a generic emission factor is used, comparable to a simple Tier 1 approach, so the development of an improved methodology would be highly recommendable.

  - For aviation a large fraction of the particles is very small and difficult to quantify in emission measurements, especially the volatile part.
  - For shipping, a similar issue exists, but in addition also the dependency on the sulphur content of the fuel makes that emission measurements from outside the sulphur controlled areas in Northern Europe are difficult to use for the Netherlands.
- Road transport, rail and inland navigation: the current approach estimates TPN emissions based on available SPN emissions, to which the volatile particles are added based on an estimation including other emission factors (e.g. EC and organic substances). The advantage of this approach is that it builds on real-world measured emission factors, on the other hand the methodology assumes fixed ratios between EC, organics and the amount of volatile particles generated. In the international literature, also many measurement-based TPN emission factors are reported. Hence, an important next step would be to compare these different approaches in order to further refine and where necessary adjust the currently used TPN emission factors in the coming years. In addition the contribution of non-exhaust sources (in particular brake wear, and wear from the use of overhead wiring in rail transport) should be further investigated.
- Energy and industry: the current approach builds mainly on estimation of TPN by means of a particle size distribution for the smallest share of the particles ($PM_{0.3}$). For the sources that are not a large contribution to the overall TPN emissions, this approach could be justified. However, also in industry there are some relatively important sources that should be investigated in more detail. A key example is the iron and steel industry, which would need to get specific attention by distinguishing the different subprocesses therein and derive TPN emission factors for the most relevant of those.

Apart from the above sector specific improvements, also the analysis and interpretation of various ambient measurements available for UFP can provide a lot of additional insights from which our knowledge of the source of particles can be improved.

Implementation in the ER system

For the implementation of the TPN emissions in the Dutch Emission Inventory, it has been attempted to use current emission sources (“emissie-oorzaken”). However, the emission inventory also contains a number of specific sources which are based on individual facility reporting, especially in the industrial sector. These may be a mix of process and combustion activities, and since it is not always fully clear, currently the emission factor chosen is based on the assumption it is one or the other which introduces uncertainty. To improve this, a more detailed look should be taken at the specific industrial facilities to select the most appropriate emission factor, or split the individual facility emissions between combustion and processes, and then apply the proper emission factors for both. However, it should be recognized that this is difficult for many facilities, therefore priority should be given to those facilities which are the largest contributors to TPN emissions.

For many industrial emission sources (“emissie-oorzaken”), indirect emission factors based on $PM_{0.95}$ emissions are used as activity data (*Methodology 8 – $PM_{0.95}$ based EF)*. Following the fractions that are currently available in the Emission Registration, these are obtained from PM_{10} emissions. However, as explained before, this leads to some complications for ‘NACE’ (‘ERI’) emission sources. For now this was solved by using $PM_{2.5}$ emissions. However, for all other emission sources, it would also improve emission estimates if $PM_{2.5}$ emissions would be used to obtain $PM_{0.95}$. Next steps would be to obtaining these $PM_{0.95}$/ $PM_{2.5}$ for all emission sources, use these to obtain $PM_{0.95}$ emissions, and add these fractions to the Emission Registration framework.

Implementation in ultrafine particle modelling in the Netherlands

This emission inventory for TPN is developed to be used as input to the modelling of ultrafine particles. An estimate has been provided for each emission source so that emissions can be aggregated to different levels of sources, and hence be used in the different air quality models used for the Netherlands, such as OPS and LOTOS-EUROS. The models need a particle size distribution for this. An important step is to use these emissions in the modelling with some kind of pre-processor, which translates these emissions to the emissions at the spatial and temporal scale at which the model is operating (e.g. 1km spatial scale and hourly temporal scale, for instance). This is important especially for the sectors where typically emitted particles are in the smallest size ranges such as aviation.

5 Conclusions and recommendations

This study presents a first version of a TPN emission inventory for the Netherlands for the year 2022, considering all relevant sources, which is the first of its kind in Europe. TPN emissions are quantified for two size ranges: 10-325 nm and 10-100 nm, where the latter represents the ultrafine particle size range. The lower boundary of 10 nm was chosen to align with the ambient measurement standard for particle number and earlier work on this topic, but also since assessment of smaller particles becomes increasingly difficult while their contribution to exposure may be limited. Specific attention has been given to estimating TPN from the transport sector, which constitutes the main sources of TPN emissions. For road transport, mobile machinery and inland shipping, we build on results from the extensive measurement programs on the basis of which emission factors for many pollutants are determined in the Netherlands. These measurements do not include TPN but TPN could be estimated based on SPN plus EC and organic substances. Sea shipping and aviation were estimated using direct emission factors derived from literature sources. For industrial sources, the estimation of TPN is based on PM mass combined with density and size distribution information.

The results show that transport sources dominate TPN emissions, especially when considering the lower size range below 100 nm. Sea shipping (incl. fishing) accounts for 44% of TPN_{325} emissions and aviation for 25%. Road transport and inland shipping contribute 12% and 10%, respectively. The energy sector, industry and consumers combined account for just 7% of the emissions, whereas the other sectors account for the remaining 2%. These numbers are in strong contrast with the sector shares for PM2.5 (mass) emissions, where the importance of transport sources is significantly smaller and consumers and industrial sources play a more important role. Here the mass is mostly in larger particles, making the number relatively small, whereas for shipping and aviation a large number of very small particles is emitted which have a negligible contribution to the particle mass.
Given that this inventory of TPN is a novel development, uncertainties are relatively high as standardized methodologies for estimating TPN emissions do not (yet) exist. This means new methodologies were developed and assumptions and simplifications were needed in order to get to a complete emission inventory. However, despite all the uncertainties such a first inventory is useful to get a good understanding of the main sources and to prioritize sectors where improvements to the emission inventory are necessary.

Specifically, we recommend to:
- Compile the TPN emissions regularly in the years to come along with the regular Dutch Emission Inventory, to be able to monitor TPN emissions over time, and implement improvements to the methodologies in line with what is done for other substances.
- When possible, extend the scope of the inventory to also include particles smaller than 10 nm in the inventory, given their contribution to ambient air quality concentrations.
- Share the results of this first TPN emission inventory with the wider (European) inventory community and scientific community, in order to make it useful for others and also to collect feedback on how this inventory can be further improved.

- Review the available information from measurement campaigns specifically targeting TPN emissions from shipping and aviation. These two sources together represent around 70% of the TPN emissions in this inventory, but rely only on a single emission factor for each of them. Assessing the emissions in more detail for these sectors will allow for a better understanding and also the ability to propose specific emission reduction measures.
- Compare the methodologies, and the underlying circumstances and dependencies therein, used to estimate TPN emissions from road transport, inland shipping and mobile machinery to available TPN source measurements from literature where available, to identify possible differences and explore options to improve the current methodology.
- Investigate particulate matter emissions from specific industrial sources which may contribute significantly to TPN emissions, for instance the iron and steel industry, and transshipment of materials. They are all known to be substantial sources of particle emissions, but the share of TPN is uncertain.
- Acknowledging that the low hanging fruit from previous UFP and PN studies has been harvested and included, it should be realized that further improvements should include a stronger investment in research. For example, a better defining of the ratio between solid and volatile PN for specific sources is important and may need more observations or emission factor measurements.
- Another promising avenue is that recently a number of UFP measurement stations have been made operational in the Netherlands. Analysis of the trends and /or episodes with high UFP concentrations can be informative for inventory improvement. Ideally this is done in collaboration between measurement experts and inventory compilers, as both disciplines are needed for such an analysis.

6 Bibliography

Dellaert, S. L. (2023). *EMMA - MEPHISTO model Calculating emissions for Dutch NRMM fleet.* Utrecht: TNO.

Denier van der Gon, H. V. (2014). *European particle number emissions for 2005, 2020 and 2030 with special emphasis on the transport sector.* Garmisch-Partenkirchen, Germany: 9th International Conference on Air Quality – Science and Application.

Eastwood, P. (2008). *Particulate emissions from vehicles.* John Wiley & Sons.

Eijk, E. v., Ligterink, N. E., Geilenkirchen, G., Ruiter, J. d., & Hoen, M. '. (2023). *Emissiefactoren wegverkeer 2023.* Den Haag: TNO.

European Commission. (n.d.). *Technical studies for the development of Euro 7: Testing, Pollutants and Emission Limits, and its Annexes.* European Commission.

Geilenkirchen, G., Bolech, M., Hulskotte, J., Dellaert, S., Ligterink, N., Sijstermans, M., . . . Hoen, M. (2023). *Methods for calculating the emissions of transport in the Netherlands.* Den Haag: PBL.

Grigoriadis, A. M. (2021). *The SCIPPER Project. D4.1 - New set of emission factors and activity information.*

Harni S.D., S. S. (2023). *Effects of emission sources on the particle number size distribution of ambient air in the residential area.* Atmospheric Environment, Vol. 293, 119419.

Hinds, W. (1999). *Aerosol technology: properties, behavior, and measurement of airborne particles.* John Wiley & Sons.

J. Wahlström, L. O. (2010). Size, shape, and elemental composition of airborne wear particles from disc brake materials. *Tribology Letters*, 15-24.

Kadijk, G., Elstgeest, M., Mark, P. v., & Ligterink, N. (2020). *• Follow-up research into the PN limit value and the measurement method for checking particulate filters with a particle number counter.* TNO.

Keuken, M. P. (2016). Particle number concentration near road traffic in Amsterdam (the Netherlands): comparison of standard and real-world emission factors. *Atmospheric Environment, 132*, 345-355.

Kittelson, D., Khalek, I., McDonald, J., Stevens, J., & Giannelli, R. (2022). Particle emissions from mobile sources: Discussion of ultrafine particle emissions and definition. *Journal of aerosol sciencescience, 159, 105881*.

Kittelson, D., Watts, W., Johnson, J., Baltensperger, U., & Burtscher, H. (2003). *Gasoline vehicle exhaust particle sampling study (No. CONF-200308-126).* University of Minnesota (US); University of Wisconsin Paul Scherrer Institute (US); Institute and the University of Applied Sciences (CH).

Kuenen, J. V. (2022). *RI-URBANS. Milestone M13 (M3.2): Dataset on PM ultrafine and non-exhaust sectoral emission distribution over Europe and plot cities.* RI-URBANS, TNO.

Kulmala, M. A. (2011). General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales. *Atmospheric Chemistry and Physics, 11(24)*, 13061-13143.

Ligterink, N., Geilenkirchen, G., Eijk, E., & Ruiter, J. (2021). *Emissiefactoren wegverkeer: wijzigingen en uitbreidingen 2021.* Den Haag: TNO.

Paasonen, P. K. (2016). Continental anthropogenic primary particle number emissions. *Atmospheric Chemistry and Physics, 16(11)*, 6823-6840.

Ruiter, J. d. (2024). *Analysis of the Emission Performance of the Vehicles Tested within the Green NCAP programme.* TNO.

Ruiter, J. d., & Mensch, P. v. (2022). *Analysis of the emission performance of the vehicles tested for the Green Vehicle Index (GVI) project.* TNO.

Samaras, Z., & et al. (2020). Measuring automotive exhaust particles down to 10 nanometres. *International Journal of Advances and Current Practices in Mobility*, 539-550.

Takegawa, N. M. (2021). Characteristics of sub-10 nm particle emissions from in-use commercial aircraft observed at Narita International Airport. *Atmospheric Chemistry and Physics, 21(2)*, 1085-1104.

Visschedijk, A. D. (2022). *UFP emissie in de Rijnmond regio in 2019.* Utrecht: TNO.

Signature

TNO Energy & Materials Transition Utrecht, 26 April 2024

ValidSigned by Sam van Goethem on 2024-04-26 15:13:31

ValidSigned by Ruben Goudriaan on 2024-04-26 13:32:16

Sam van Goethem
Research Manager

Ruben Goudriaan
Project Manager

Appendix A
Description of Electronic Appendices

This report contains several Electronic Appendices (EA), all combined in one Excel file. This consists of 4 tables:

- EA.1 – Sector and Doelgroep (NL, Emission Registration)
  - This table indicates the sectors referred to in this study and how these relate to the “Doelgroep” as in the Dutch Emission Inventory. The Dutch Emission Inventory already links every emission source to a “Doelgroep”.
- EA.2 - All emission causes and fuel types considered in this study, together with its source group and methodology. For Methodology 7 - Fuel based EF and Methodology 8 - PM0.95 based EF, also emission factors are included.
  - This table provides information on the methodology for each emission cause and fuel type. For emission causes for which emissions are obtained using *Methodology 7 - Fuel based EF* and *Methodology 8 - PM0.95 based EF*, also the emission factors and a EF short code are included. The EF short code indicates which emission factor is used.
- EA.3 – All emission factors used in analyses, including a brief description and project reference.
  - This gives an overview of the various emission factors considered in this study. It includes a more general description of the process captured by this emission factor. For *Methodology 7 - Fuel based EF* and *Methodology 8 - PM0.95 based EF* , only a selection of emission is factor is used. Some emission factors are reused for multiple emission causes.
    In this table, each emission factor is assigned an EF short code, which is included in table EA.2 for referencing. Furthermore, this table also includes a brief description of the types of processes or sources that the emission factor represents. The emission factors used in this study originate from literature review in earlier projects. For each emission factor, the project reference is also included in this overview.
- EA.4 – Fractions used to obtain $PM_{0.95}$ estimates from PM_{10} or $PM_{2.5}$ reported emissions.
  - These fractions come from the Dutch Emission Inventory. However, this table is updated regularly by the Dutch Emission Inventory. Therefore, the fractions used for this study are also included here.

Energy & Materials Transition

Princetonlaan 6
3584 CB Utrecht
www.tno.nl