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1 A tunnel study to validate motor vehicle emission prediction software in Australia

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7 Abstract

- 8 A tunnel emissions study was conducted to (partially) validate the Australian vehicle emissions software
- 9 COPERT Australia and PIARC emission factors. The in-tunnel fleet mix differs substantially from the average
- on-road fleet, leading to lower emissions by factor of about 2. Simulation with the $P\Delta P$ software found that in-
- tunnel air-flow roughly compensates for road gradient impacts on NO_x emissions. PIARC emission factors are
- 12 conservative and exhibit the largest prediction errors, except for one very good agreement for LDV NO_x.
- 13 COPERT Australia is generally accurate at fleet level for CO, NO_x, PM_{2.5} and PM₁₀, when compared with other
- international studies, and consistently underestimates emissions by 7% to 37%, depending on the pollutant.
- Possible contributing factors are under-representation of high/excessive emitting vehicles, inaccurate mileage
- 16 correction factors, and lack of empirical emissions data for Australian diesel cars. The study results demonstrate
- 17 a large uncertainty in speciated VOC and PAH emission factors.

18 Graphical abstract



Highlights

22	Tunnel studies are useful to partially validate vehicle emissions software
23	Air flow in tunnels can compensate the impacts of road gradients on vehicle emissions
24	Local fleet mix is an essential factor in validation studies
25	Keywords
26	Motor vehicle; emissions; tunnel; validation; road traffic
27	1. Introduction
28	Motor vehicles are a major source of air pollution and greenhouse gas (GHG) emissions in urban areas around
29	the world. The close proximity of motor vehicles to the general population makes this a particularly relevant
30	source from an exposure and health perspective. This is illustrated by Caiazzo et al. (2013) who estimated that
31	total combustion emissions (particulates, ozone) in the U.S. account for about 210,000 premature deaths per
32	year, with motor vehicles being the largest contributor, contributing to around 58,000 premature deaths per year,
33	despite the fact that road transport only contributes about 7% to total $PM_{2.5}$ emissions.
34	Comprehensive measurement of vehicle emissions in urban networks is cost prohibitive due to the large number
35	of vehicles that operate on roads with different emission profiles, large spatial and temporal variability in vehicle
36	activity and many real-world factors that influence emission levels (Smit et al., 2008). The environmental
37	impacts of road traffic are therefore commonly evaluated at different scales using transport and emission models
38	and, in the case of air pollution, dispersion and exposure models. Models are also required to make projections
39	into the future.
40	Vehicle-emission prediction software is well-established in Europe and the US. However, these models have
41	been found to not adequately represent Australian conditions in terms of fleet mix, vehicle technology, fuel
42	quality and climate. Large errors of up to a factor of 20 have been reported when overseas models were directly
43	applied to Australian conditions without calibration (Smit and McBroom, 2009). Therefore, two software
44	packages have been developed specifically for Australian conditions, using comprehensive empirical data from
45	major Australian laboratory emission testing programs. COPERT Australia has been developed to estimate

46	motor vehicle emissions at a regional and national level, while a power-based model (P Δ P) was developed for
47	local assessments, as will be discussed in section 2.2.
48	As models are simplifications of reality, their limitations and accuracy should be clearly established. This paper
49	presents results of a tunnel emissions study that was conducted in Brisbane, Australia.
50	2. Method
51	2.1 Tunnel studies
52	There are several methods used to (partially) validate vehicle emission models, such as on-board emission
53	measurements (PEMS), remote sensing, near-road air quality measurements and tunnel studies (Smit et al.,
54	2010). Like all validation methods, tunnel studies have specific strengths and weaknesses. A strength is that
55	emissions are derived from a large sample of the on-road fleet under relatively controlled conditions, thereby
56	adequately capturing inter-vehicle variability in emissions. The spatial resolution aligns better with distance-
57	based emission factors (g/km) commonly used in vehicle emission models, as compared with localised
58	validation methods such as remote sensing and near-road air quality measurements.
59	However, there are also some challenges with tunnel studies. They represent only a limited range of operating
60	conditions (typically 'smooth', uncongested, high-speed driving). As a consequence, validation results cannot be
61	directly translated, for example, to commonly occurring urban driving conditions at lower speeds. Tunnels may
62	also have significant uphill and downhill gradients, and in-tunnel air-flows affecting emissions. Furthermore,
63	assumptions relating to the unknown proportion of vehicles in cold-start mode and actual vehicle loads are
64	required to make a comparison with model predictions. Nevertheless, tunnel studies provide a useful approach to
65	(partially) validate vehicle emission models for specific traffic situations.
66	Tunnel studies have been extensively used around the world to compare model predictions with observed values
67	(e.g. De Fré et al., 1994; Hausberger et al., 2003; Geller et al., 2005). In these studies, emission factors,
68	expressed as grams of pollutant per vehicle kilometre (g/veh.km, subsequently denoted as g/km), are determined
69	using the differences between the concentration levels at the tunnel entrance and exit, combined with tunnel
70	features (e.g. road length), traffic flow and traffic conditions, as well as either measured tunnel air-flow or a
71	dilution factor based on a tracer gas (e.g. SF ₆). Regression analysis is often used to develop mean emission
72	factors (g/km) by time of day for basic vehicle classes (e.g. light-duty vehicle, LDV and heavy-duty vehicle,
73	HDV). License plate information is typically recorded to obtain a detailed breakdown of the on-road fleet. In
74	tunnels with distinct traffic flow patterns (e.g. separate bores for trucks), separate emission factors can be

75	produced. Tunnel lengths vary from a few hundred metres to 10 km. Several studies are done in tunnels with		
76	significant road gradients up to 4.2%. The averaging time of measurement is typically one hour and total		
77	sampling times vary from 10 hours to a month (Smit et al., 2010).		
78	2.2 Australian vehicle emissions software		
79	COPERT (COmputer Program to calculate Emissions from Road Transport) is a globally used software tool		
80	used to calculate air pollutant and GHG emissions produced by road transport, and its scientific development is		
81	managed by the European Commission. A dedicated Australian version of COPERT was developed to reflect		
82	local fleet composition and driving characteristics and provide vehicle emission estimates for the Australian		
83	situation (Mellios et al., 2013; Smit and Ntziachristos, 2013a). The software has been adopted by the National		
84	Pollutant Inventory as the recommended model for motor vehicle emission inventories and has been used to		
85	estimate motor vehicle emissions for all states and territories in Australia (UQ, 2014).		
86	COPERT Australia estimates emissions for 122 air pollutants and greenhouse gases. The software estimates		
87	emissions of both cold-start and hot-running exhaust and non-exhaust pollutants. COPERT Australia predicts		
88	emissions for 226 individual vehicle classes, which are defined in terms of vehicle type (e.g. small passenger car		
89	large SUV, heavy bus, rigid truck, articulated truck), fuel type (petrol, E10, diesel, LPG) and 'emission control		
90	technology level' or ADRs (Australian Design Rules), which are the vehicle emission standards adopted in		
91	Australia (equivalent to Euro standards since 2003). The software accounts for various other factors such as		
92	driving conditions (average speed), fuel quality, impacts of ageing on emissions and meteorology (ambient		
93	temperature and humidity).		
94	The P Δ P software uses engine power (P, kW) and the change in engine power (Δ P, kW) to simulate fuel		
95	consumption and CO ₂ and NO _x (hot-running) emissions for 73 Australian vehicle classes for each second of		
96	driving (Smit, 2013). P Δ P has adopted the vehicle classification used in COPERT Australia, but with a focus on		
97	the most important vehicle classes. Similar to COPERT Australia, the software was developed using empirical		
98	data from a verified Australian emissions database with about 2,500 second-by-second emission tests (1 Hz) and		
99	about 12,500 individual aggregated 'bag' measurements using real-world Australian drive cycles. Multivariate		
100	time-series regression models have been fitted to these data using P and ΔP as predictor variables. The input to		
101	the model is speed-time data (1 Hz) and information on road gradient, wind speed, vehicle loading and use of air		
102	conditioning (on/off). This information is used to compute the required (change in) engine power for each		
103	second of driving, and subsequently predict second-by-second fuel consumption and emissions. The software		
104	has been used to estimate vehicle emissions in small urban networks using output from a microscopic transport		

model. The purpose was to estimate the impacts of a safety intervention programs on vehicle emissions using on-road GPS measurements and to assess the impacts of dynamic speed limits on emissions (Smit, 2014). The software is ideally suited to examine the combined impacts of vehicle speed, road gradient and piston air-flow in tunnels on emissions for all major on-road vehicle types (cars, SUVs, LCVs, rigid trucks, buses, articulated trucks).

PIARC (Permanent International Association of Road Congresses) publishes country-specific emission factor tables that are widely used around the world to estimate emission levels generated in tunnels, and assess ventilation requirements to maintain acceptable in-tunnel air quality and visibility (PIARC, 2012) .PIARC provides CO and NO_x emission factors (g/h), and opacity factors (m³/h, proxy for particulate matter) specifically for the Australian on-road fleet. Emission rates are provided for a range of speeds and road gradients for four vehicle classes, i.e. petrol and diesel passenger cars, light-duty vehicles (petrol/diesel mix) and diesel heavygoods vehicles.

2.3 Measurements in the Brisbane CLEM7 tunnel

Brisbane's Clem Jones Tunnel (CLEM7) has 4.8 km of twin one-directional 2-lane tunnels, with a cross-sectional area of about 60 m², linking major Brisbane roads. To control for portal emissions, the tunnel is subjected to forced ventilation through a combination a of jet fans inside the tunnel and exhaust fans located near the portals. Air monitoring equipment was installed in the north tunnel ventilation vent on 25 August 2014, as is shown in Figure 1.



Figure 1 – Installing measurement equipment in the CLEM7 Northbound tunnel vent.

125	Air-monitoring data (five-minute average) was collected in the vent for 9 days for CO, NO, NO ₂ , NO _x , PM _{2.5} ,
126	PM ₁₀ , speciated volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs), as well as
127	variables quantifying conditions in the tunnel vent (temperature, relative and absolute humidity, atmospheric
128	pressure).
129	Nitrogen oxides (NO, NO2, NOx) were measured using a light emission (chemiluminescent) analyser (Teledyne
130	API200). Carbon monoxide (CO) was measured with an infrared absorption instrument utilising the gas filter
131	correlation technique (Teledyne API300). Particle concentrations were measured with a Thermo Scientific 1405-
132	DF TEOM Continuous Dichotomous Ambient Air Monitor to simultaneously measure $PM_{2.5}$ and PM_{10} .
133	In contrast to high-resolution (5-minute) measurements of CO, NO _x and PM, sampling periods for VOCs and
134	PAHs are 24 hours or longer and they were not conducted for the full measurement period. Evacuated canisters
135	fitted with timers and critical orifices were used to take VOC samples over a 24-hour period on sequential days
136	in the tunnel vent. Some canisters experienced problems with the timer and did not provide a sample. Samples
137	were successfully collected for a total of four days. The canisters were then sent for laboratory analysis using gas
138	chromatography and mass spectrometry (GC/MS) in accordance with the US EPA Compendium TO-15 analysis
139	method. PAHs have been collected using a low-volume air sampler in combination with a frit and a sorbent
140	cartridge (XAD-2 resin) over an approximately nine-day sampling period. After sampling, they were extracted
141	together to obtain the gas-phase and particle-associated PAH concentrations. Gas Chromatography - High-
142	Resolution Mass Spectrometry was used for the PAHs analysis.
143	The pollutant monitoring data were checked by pre- and post-test calibration, as daily calibration for zero and
144	span values could not be carried out during the test period. Particulate matter monitoring data collected with the
145	TEOM instrument were verified according to Australian Standard AS/NZS 3580.9.13:2013.
146	Tolling statistics are continuously collected at the exit of the northbound tunnel using camera-imaging
147	technology. License plate numbers (LPNs) are collected and date-time stamped for each vehicle that passes the
148	cameras. Each vehicle is then classified as a motorcycle, car, light commercial vehicle (LCV) or heavy
149	commercial vehicle (HCV) using height, length and width of each vehicle, which are determined when the
150	vehicles travel through a specific zone on the road. Figure 2 shows the variation of traffic volumes going through
151	the tunnel by hour of day and day of the week.

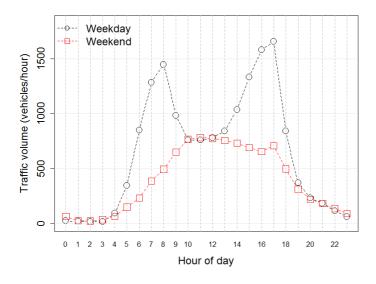


Figure 2 – Average total traffic count by hour of day and by day of the week.

Figure 3 shows a time-series plot of measured NO_x concentration levels ($\mu g/m^3$) in the north ventilation stack, including the urban background concentration levels measured at South Brisbane station. The daily variation in traffic flows is clearly visible in the concentration data, as is the difference between weekdays and weekend (30 and 31 August).

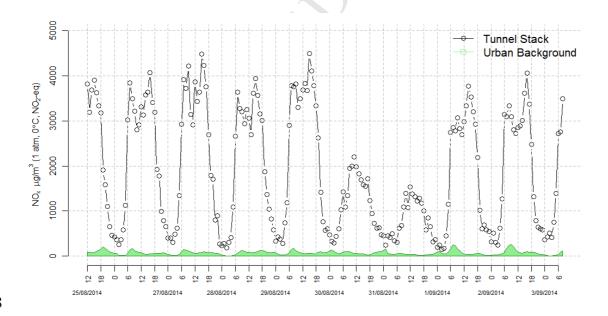


Figure 3 – Hourly averaged tunnel vent NO_x concentrations (NO_2 -equivalents) and urban background concentrations.

161	2.4 Emission computation
162	Examination of five-minute data was performed to check the quality and validity of the raw concentration
163	measurements, before hourly averaged values were computed. Tunnel emissions were computed by multiplying
164	hourly-averaged measurements of time-aligned and background-corrected concentrations by tunnel air-flow data
165	(m³/h). Ambient concentration data from nearby monitoring stations were used to estimate concentrations at the
166	tunnel entrance point. Hourly vehicle travel in the tunnel is quantified with a variable called 'vehicle kilometres
167	travelled' (veh.km/h). Hourly VKT were computed by multiplying total traffic volume (veh/h) derived from
168	tolling statistics with total distance (km). NO _x emissions were corrected for humidity.
169	2.5 In-tunnel fleet mix
170	The LPN data were cross-referenced with vehicle registration information from the Queensland Department of
171	Transport and Main Roads, and individual vehicles were allocated to one of the 226 vehicle classes used in
172	COPERT Australia. About 13% of LPN could not be matched with Queensland vehicle registration data,
172	reflecting unidentified license plates and the portion of inter-state and unregistered vehicles. A comparison
174	between the average Queensland fleet (UQ, 2014) and the in-tunnel fleet based on analysis of license plate
175	numbers revealed that there are significant differences.
175	numbers revealed that there are significant differences.
176	Whereas the VKT weighted proportion of diesel and petrol/E10 vehicles is similar (~ 29% and ~70%,
177	respectively), the tunnel has higher proportion of medium passenger cars and SUVs, as compared with the
178	Queensland average fleet. Importantly, the vehicle fleet in the tunnel is substantially younger with better engine
179	and emission-control technology, as compared with the average 2010 Queensland fleet. This is partly explained
180	with the difference in base year, but also expected to reflect a tendency for newer vehicles to use tolled tunnels.
181	The impact of fleet mix on emissions is further discussed in section 3.3.
182	2.5 Road gradient and air-flow
183	In-tunnel air speeds are measured continuously and vary from 7 to 18 km/h, depending on the time of day. The
184	road gradient profile of the tunnel was determined from tunnel design maps. In-tunnel driving behaviour was
185	recorded and analysed in a brief measurement campaign of traffic conditions by driving a car in and around the
186	CLEM7 tunnel on 27 August 2014 in the morning peak hour (8:30am – 10:00am) using the ATLAS II ¹ smart
187	app (Safi et al., 2015). Driving behaviour in the tunnel can be characterised as 'free-flow freeway conditions'
188	with an 80 km/h speed limit. Using this information, the $P\Delta P$ software was run to quantify the combined impact

 $^{^{\}it I}$ Advanced Travel Logging Application for Smartphones II.

of road gradient, (piston) air-flow and tunnel driving conditions on vehicle emissions. Two input files were created: 1) a second-by-second input file for in-tunnel vehicle speed, (variable) air speeds and road gradient (Figure 4), and 2) a second-by-second input file with the same vehicle speed profile but with zero air speed and zero road gradient ('base case').

Total emissions (grams of NO_x) were then calculated for each tunnel journey for a range of in-tunnel wind speeds, and divided by total tunnel distance to compute average emission factors (g/km) for LDVs and HDVs. By dividing these composite emission factors with the 'base case' values, correction factors were computed as a function of in-tunnel air speed. The results are shown in Figure 5. COPERT Australia emission factors for NO_x are corrected with these values.

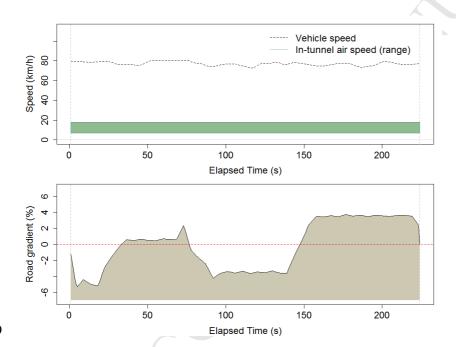


Figure 4 – Visualised second-by-second input file for the $P\Delta P$ software, including in-tunnel air speed (range), road gradient profile and vehicle speed for tunnel.

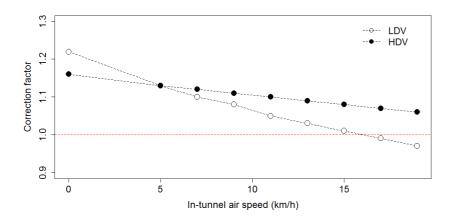


Figure 5 – Correction factors for combined road grade and in-tunnel air-flow impacts on vehicle emissions in the CLEM7 tunnel for LDVs and HDVs as computed with the $P\Delta P$ software.

The road gradient effect on in-tunnel emissions is substantial with an approximately 20% net increase in NO_x emissions (air speed is zero km/h). However, in-tunnel air speed is predicted to have a significant impact on emissions: it roughly compensates for the impacts of road gradient at higher air speeds due to reduced aerodynamic drag. The average correction factor for the full measurement period, accounting for variable intunnel air-flows, is therefore small: 1.01 and 1.08 for LDVs and HDVs, respectively.

2.6 Start emissions

in-tunnel vehicles are in cold-start mode is difficult to determine, and would require a detailed analysis of start location and distance driven to the tunnel entry. This information is not readily available. However, given that the bulk of cold-start emissions are typically emitted in the first minute of driving (Smit and Ntziachristos, 2013b) and the long length of the tunnel, it is expected that most vehicles will be driving in hot-running

Cold starts contribute significantly to total vehicle emission loads, on average, 42%, 31%, 7% and 5% to total

emissions of CO, VOCs, NO_x and PM_{2.5}, respectively, for the Queensland fleet (UQ, 2014). The extent to which

conditions. As a result, the unknown impact of cold-start conditions is expected to be insignificant.

219	2.7 High-emitters
220	Vehicle ageing has a significant and unavoidable effect (increase) on vehicle emissions, and this is aggravated
221	by poor maintenance and tampering. Vehicle fleet emissions are dominated by a small percentage of 'high-
222	emitters' with excessive emission levels, which has been confirmed by different types of emission studies
223	including laboratory test programs (e.g. Sjödin and Lenner, 1995; Pierson et al. 1999, Choo et al., 2007; RTA,
224	2009) and remote sensing studies (e.g. Zhang et al., 1995, NIWA, 2008; 2015). Studies have shown that
225	emissions from 'high-emitting' vehicles can be at least 50 times higher than a properly functioning catalyst car
226	(e.g. Sjödin et al., 1997), and improper maintenance (and tampering) has been indicated as the principal reason
227	for the skewness of vehicle emission distributions. The latter will remain unchecked and unverified in the
228	absence of inspection and repair programs, as is the case in Australia.
229	Recent remote sensing studies (Park et al., 2012) suggest that the skewness of (~1 Hz) emission distributions for
230	CO, hydrocarbons (HC) and NO _x has increased over the last decade due to high-emitting vehicles, whereas fleet-
231	averaged emissions have decreased considerably. Bishop et al. (2012) reported that 1% of on-road vehicles in the
232	USA contributed about 10% to total vehicle emissions in the late 1980s, and that this contribution of 1% of on-
233	road vehicles now has increased to about 30%. This is, to some extent, also caused by the the irregular emissions
234	behavior of modern cars, which is increasingly characterised with low emission levels and brief and large
235	emission peaks (e.g. De Haan and Keller, 2000; Smit, 2013).
236	This change in on-road emission profiles reflects two main trends 1) the penetration of cleaner vehicles into the
237	fleet over time due to increasingly strict emission standards and improved control technologies with irregular
238	emissions behaviour, and 2) the presence of vehicles that are badly tuned or have been tampered with, have
239	engine issues and/or have malfunctioning or partly functioning emission control systems (catalysts, lambda
240	sensors, faulty fuel caps, fuel injector malfunction, worn turbochargers, clogged air filters etc.). It is noted that
241	there could be other reasons for the occurrence of vehicles with excessive emission levels than tampering, engine
242	(tuning) issues and malfunctioning emission control systems, such as poorly retrofitted fuel systems and to some
243	extent even heavy loads.

3. Results and discussion

 $3.1 \, NO_2 \, to \, NO_x \, ratios$

 NO_2 to NO_x ratios are of interest as they quantify the proportion of NO_x that is directly emitted as NO_2 . The box-and-whisker plot in Figure 6 shows that NO_2 to NO_x ratios are typically 0.15 during times of day with significant traffic volumes (6 AM – 8 PM). This is in line with expected mean primary NO_2 emissions at fleet level (e.g. Soltic and Weilenmann, 2003; Carslaw and Beevers, 2005). COPERT Australia predicts an average ratio of 0.13.

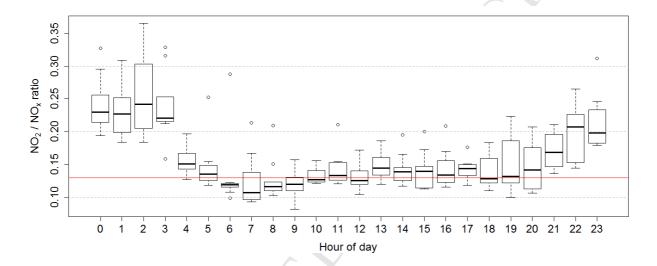


Figure 6 – Box-and-whisker plot of measured NO_2 to NO_x ratios in the tunnel by hour of day. Red line shows the predicted ratio with COPERT Australia.

Atmospheric photo-oxidation produces ozone in the urban atmosphere, which reacts with NO in e.g. vehicle exhaust producing NO_2 . This reaction occurs relatively fast in heavy trafficked areas, including tunnels, resulting in reduced ozone concentrations and elevated NO_2 concentrations (e.g. McConnell et al., 2006). At night, NO_x concentrations are substantially reduced (Figure 3) and ozone formation ceases. On balance, NO_2 to NO_x ratios can be high at low ambient concentration levels (typically 0.75 to 0.90). This effect is visible in Figure 6, where NO_2 to NO_x ratios in the tunnel are higher at night (around 0.25). At night, traffic volumes in the tunnel are small (Figure 2) and ratios are more affected by ambient ratios. This suggests that for model validation, hours with small traffic volumes should not be used as these measurements can be significantly impacted by potential errors in estimated background concentration levels.

264 3.2 Model prediction errors

Figure 7 shows hourly emission predictions and observations in goodness-of-fit plots for each pollutant. A dot point represents one hourly value. The grey dashed 45° lines indicate a perfect fit without bias. Any dot points on this line show model predictions that are equivalent to observations. If a point lies below the 45° line, the model under-predicts, and if is lies above the 45° line, the model over-predicts.

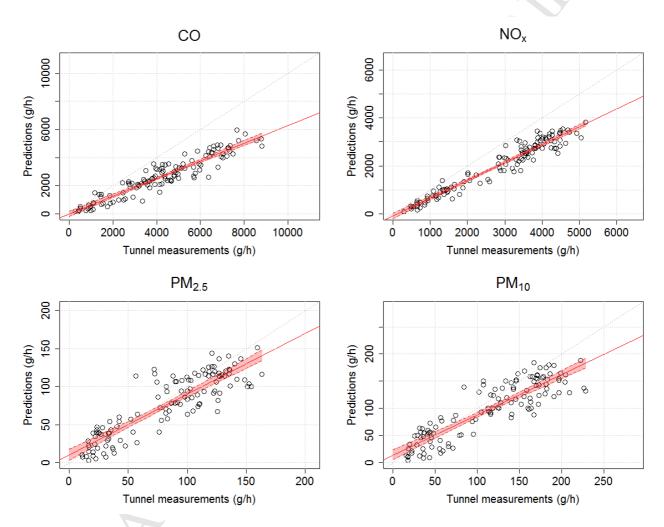


Figure 7 – Hourly COPERT Australia predictions versus measured tunnel emissions by pollutant (red line = linear regression line, red shading = 95% confidence intervals).

A linear ordinary least-squares (OLS) regression model was fitted to these data:

273
$$P = \beta O + \varepsilon$$
 Equation 1

In this model **P** represents a vector of hourly predictions, **O** the vector of hourly observations, $\boldsymbol{\beta}$ is a vector of regression coefficients (β_0 , β_1) and $\boldsymbol{\varepsilon}$ is the vector of error terms.

This model is useful as the slope (β_I) can be used to estimate the systematic error or bias in COPERT predictions in relation to the measured tunnel emissions. The coefficient of determination (R^2), estimated intercept (b_0) and slope (b_I) and bias are shown in Table 1.

Table 1 – Model performance statistics showing fitted regression coefficients (\pm standard error, p-value), coefficient of determination and bias.

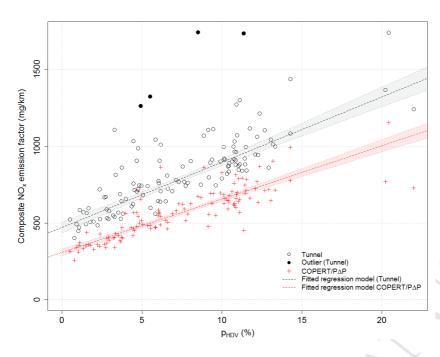
Pollutant	b_0	b_1	R^2	Bias
CO	$+5.66 \pm 99.61 \ (p = 0.955)$	$0.63 \pm 0.02 \ (p < 0.001)$	0.88	-37%
NO _x	$-89.40 \pm 56.13 \ (p = 0.114)$	$0.74 \pm 0.02 \ (p < 0.001)$	0.93	-26%
PM _{2.5}	$+10.10 \pm 3.61 \ (p = 0.006)$	$0.80 \pm 0.04 \ (p < 0.001)$	0.78	-7% ^{a)}
PM_{10}	$+13.96 \pm 4.63 \ (p = 0.003)$	$0.74 \pm 0.04 \ (p < 0.001)$	0.78	-14% ^{b)}

^a bias for an average concentration value of 77 μ g/m³, bias is a function of observed concentration and ranges from +52% at the lowest measured concentration to -13% at the highest measured concentration, ^b bias for an average concentration value of 114 μ g/m³, bias is a function of observed concentration and ranges from +32% at the lowest measured concentration to -19% at the highest measured concentration.

Fitted intercepts are expected to be zero as zero emission predictions (no vehicles in the tunnel) should correspond to zero emission measurements. The intercepts are not significantly different from zero for CO and NO_x , but are significantly different for PM. One contributing factor is that background concentration levels are relatively high for PM (on average 6 μ g/m³ for PM_{2.5} and 14 μ g/m³ for PM₁₀). As a consequence, errors in background concentration data can significantly impact on the results. There are also significant differences between the empirical base for the COPERT software and the tunnel results that may significantly affect measured PM mass concentrations, and can distort expected relationships with regard to traffic volume, driving conditions and fleet mix. Whereas laboratory emission measurements are conducted under strictly defined and controlled conditions, the tunnel PM samples measure particles that have aged (typically 8 minutes after emission from exhaust pipe) and have undergone several processes such as nucleation, coalescence and condensation, as well as absorption to and re-entrainment from tunnel walls. Tunnels are also uncontrolled in relation to non-exhaust PM emissions, and could be significantly influenced by e.g. trucks carrying dusty loads.

The regression model suggests that the prediction software under-estimates emissions by 7 to 37%, depending on the pollutant. These validation results appear to be relatively good. For instance, a review of 50 international vehicle emission model validation studies showed that reported model prediction errors are generally within a

299	factor of 2 for NO _x and within a factor of 3 for CO and PM, although differences as high as a factor of 5 have			
300	been reported (Smit et al. 2010).			
301	A plausible factor for this consistent underestimation may be (in part) an incomplete representation in the			
302	COPERT Australia emission factors of vehicles with high or even excessive emissions ('high-emitters'), as was			
303	discussed in section 2.7. This issue has been reported in other studies. For instance, NRC (2000) considered that			
304	under-representation of emissions from high-emitters in the US MOBILE model was one of the chief reasons for			
305	MOBILE under-predicting real-world fleet emissions. A related factor could be inaccurate computation of			
306	ageing effects of Australian vehicles. Although COPERT Australia simulates the effects of ageing with generic			
307	mileage-correction algorithms, they are based on limited non-Australian data. In fact, recent research suggests			
308	that these correction algorithms underestimate ageing effects on emissions substantially and thus require further			
309	calibration (Borken-Kleefeld and Chen, 2015).			
310	3.3 Model prediction errors by vehicle class			
311	Consideration of vehicle class specific prediction errors facilitates cost-effective and focused vehicle emission			
312	measurement programs that target specific vehicle classes, which show substantial discrepancies between			
313	observed and predicted emission factors. Composite emission factors (g/km) were computed by dividing hourly			
314	tunnel emissions (g/h) by total hourly travel (veh.km/h). Hours with reduced average speeds less than 75 km/h			
315	(e.g. due to tunnel maintenance) were removed to ensure homogeneous and comparable traffic conditions.			
316	Hourly data with less than one vehicle going through the tunnel per minute were also removed. This is important			
317	because hourly data with a small number of vehicles can be significantly influenced by errors in urban			
318	background concentrations, in particular for pollutants with relatively high background levels such as particles			
319	(PM), as was discussed previously.			
320	The hourly composite emission factors (e) are plotted against the percentage of heavy-duty vehicles (p _{HDV}). An			
321	example for NO_x is shown in Figure 8. Figure 8 shows the hourly tunnel data, as well as the hourly predictions			
322	with the COPERT Australia and P Δ P model. The significant variation in the COPERT Australia and P Δ P model			
323	predictions reflects the impact of the changing fleet mix for each hour of the sampling period inside the tunnel.			



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Figure 8 - Measured and predicted NO_x composite emission factors for each hour, fitted regression models with 95% prediction intervals and outliers.

A two-step approach was employed in the regression analysis for the tunnel data. The occurrence of excessive-

emitters in a particular hour is expected to substantially increase the composite emission factor (g/km) and will

show up as outliers in the computed emission factors. It is important to include these valid outliers in the

determination of composite emission factors from the in-tunnel measurements. However, this poses specific

Therefore a robust weighted linear modelling (RWLM) approach was first used to identify these outliers. This

regression is weighted with the total VKT for each hour to account for the higher accuracy of data points with

more vehicles. Any hourly emission value that exceeds the median value plus three times the (robust) standard

As the second step, a (VKT-)weighted ordinary least squares (OLS) linear regression was performed on the data

Here e_h is the mean of the hourly emission values that were tagged as outliers, and p_h is the proportion of outliers

in the data. It is thus assumed that high-emitters 1) form a small portion of the fleet and occur randomly in time,

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Equation 2

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 $e = e_h p_h + \beta p_{HDV} + \varepsilon$

without outliers. The regression model is defined as:

issues in the model-fitting process that need to be addressed.

deviation is tagged as an outlier (shown as black solid dots in Figure 8).

and 2) are not significantly affected by the proportion of HDVs. For the CLEM7 data the number of hours with outliers (significant 'high-emitter impacts') was 2% for PM, 3% for NO_x and 4% for CO. This percentage is in line with overseas reports. For instance, Choo et al. (2007) analysed 837,829 Inspection and Maintenance (I/M) test results and found that approximately 4.6% of all vehicles are labelled as 'gross polluters'. The high-emitter offset ($e_h \times p_h$) in equation 2 typically adds an offset value of 10-15% to the mean emission factor ($p_{HDV} = 5\%$). A similar weighted ordinary least squares (OLS) linear regression model was fitted to the COPERT

Australia/PΔP model predictions (Equation 2, but without the high-emitter offset term).

After model fitting, light-duty and heavy-duty emission factors were computed by using p_{HDV} values of 0% and 100%, respectively, in the linear regression models for each pollutant. Table 2 shows predicted and observed emission factors for LDVs and HDVs, including 95% confidence limits.

Table 2 – Composite emission factors (mg/km) in hot-running conditions for LDVs and HDVs, including 95% confidence limits and comparison with COPERT Australia/PΔP and PIARC.

Pollutant COPERT Australia/PΔP		PIARC	CLEM7 Tunnel		
Light-duty vehicles ($p_{HDV} = 0\%$)					
CO	718 ±29 (1,662) ^{a,c} *	2,486 *	1,370 ±79		
NO_x	307 ±23 (681) ^{a,b} *	504	519 ±36		
PM _{2.5}	13 ±1 (26) ^a *	· -	15 ±2		
PM_{10}	18 ±1 (32) ^a	_	21 ±3		
Heavy-duty vehicles ($p_{HDV} = 100\%$)					
CO	941 ±340 (1,055) ^a	1,308 *	-90 ±939		
NO _x	3,780 ±273 (6,634) ^{a,b} *	7,538 *	4,771 ±435		
PM _{2.5}	124 ±12 (134) ^a	_	137 ±26		
PM_{10}	142 ±12 (149) ^a *	_	210 ±36		

^a prediction for Queensland average fleet within brackets, ^b COPERT prediction includes PΔP correction for tunnel road gradient and air-flow,

Table 2 shows that the fleet mix in the tunnel has a large impact on predicted emission factors. This was already visible in Figure 8, which shows the variation in predictions solely due to variation in the in-tunnel fleet mix. In addition, COPERT Australia predictions for the *average* Queensland fleet produce LDV and HDV emission factors that are a factor of 1.7-2.3 and 1.1-1.7 higher, respectively, as compared with the in-tunnel fleet. These results shows the sensitivity of model predictions to the local fleet mix, and indicates that detailed local fleet mix information should be explicitly considered in validation studies.

 $^{^{\}circ}$ 962 mg/km if corrected for road gradient impacts, * statistically significant difference with observations (p < 0.05)

361	The PIARC emission factors for CO and NO _x reflect the in-tunnel fleet mix and are substantially higher than
362	COPERT in all cases, varying from a factor of 1.6 to 3.5, depending on the pollutant and vehicle class. These
363	results indicate that PIARC emission factors are generally conservative.
364	In terms of prediction errors, comparison of the measured and predicted emission factors show that in several
365	cases the differences between observations and predictions are not significantly different from zero ($p < 0.05$),
366	i.e. PIARC: LDV NO _x , COPERT Australia: HDV PM _{2.5} , LDV PM ₁₀ , HDV CO, as is shown in Table 2.
367	The PIARC and COPERT Australia CO emission factors for LDVs are 81% higher and 48% lower, respectively
368	than the value measured in the tunnel, and these differences are statistically significant (p < 0.05). A possible
369	reason for the underestimation of CO emissions in COPERT could be additional emissions due to cold starts and
370	road gradient. The unknown impact of cold-start conditions is expected to be insignificant, as discussed in
371	section 2.6. The PIARC method suggests an increase in the CO LDV emission factor of 34% due to road
372	gradient effects in the tunnel. Correcting COPERT Australia predictions with this correction factor reduces the
373	prediction error for COPERT Australia from -48% to -30%. It is suggested that high-emitting vehicles in the on-
374	road fleet and possibly inaccurate mileage correction factors play a significant role in the underestimation, as
375	will be discussed later.
376	A negative HDV emission factor is estimated for CO with the tunnel model, with a 95% confidence interval of -
377	1.0 to +0.8 g/km. This large uncertainty is the result of substantial variability in observed CO emissions and
378	significant extrapolation ($p_{HDV} > 0.22$). The COPERT Australia CO emission factor for HDVs is 0.9 ± 0.3 g/km
379	and is not statistically significant (p < 0.05). Cold-start effects on the CO HDV emissions are expected to be
380	insignificant. The PIARC method applied to the CLEM7 tunnel suggests an increase in the CO HDV emission
381	factor of 6% due to road gradient effects. The computed PIARC CO emission factor for HDVs is about 40%
382	higher than the COPERT Australia value.
383	COPERT Australia predicts an average LDV NOx emission factor of 0.7 g/km for the Queensland fleet, but a
384	substantially lower value of 0.3 g/km for the actual fleet mix in the tunnel. These values have been corrected for
385	the impacts of road gradient and piston air-flow in the tunnel, using the $P\Delta P$ software (Section 2.5). The
386	corrected LDV COPERT Australia NO _x emission factor is 40% lower than the (humidity-corrected) value
387	measured in the tunnel and this difference is statistically significant (p < 0.05). This may reflect a higher-than-
388	expected proportion of (diesel) vehicles with maintenance issues. The result is of interest as there is a lack of
389	empirical vehicle emissions test data for Australian light-duty diesel vehicles in particular. This is in contrast to
390	light-duty petrol vehicles for which extensive emission test programs have been carried out in Australia. As a

391	consequence, European emission algorithms for diesel cars were directly used in COPERT Australia, and it is
392	the only vehicle type for which Australian vehicle emission measurements have not been available.
393	The PIARC NO _x emission factor for LDVs is almost equivalent to the observed value. However, PIARC
394	overestimates the NO_x emission factor for HDVs with 60% and this difference is statistically significant (p <
395	0.05). The tunnel measurements produce a composite HDV NO _x emission factor of 4.7 g/km. COPERT
396	Australia predicts an average HDV NO _x emission factor of 6.6 g/km for the Queensland fleet, but a substantially
397	lower value of 3.8 g/km for the actual fleet mix in the CLEM7 tunnel. These values have been corrected for the
398	impacts of road gradient and piston air-flow in the tunnel using the $P\Delta P$ software. The corrected HDV COPERT
399	Australia NO_x emission factor is 19% lower than the value measured in the tunnel, and the difference is
400	statistically significant (p < 0.05). This may reflect heavy-duty diesel vehicles with e.g. maintenance issues and
401	elevated NO _x emissions that are not yet fully reflected in the software.
402	COPERT Australia predicts an average LDV PM _{2.5} and PM ₁₀ emission factor of 26 and 32 mg/km for the
403	Queensland fleet, and a substantially lower value of 13 and 18 mg/km for the actual fleet mix in the tunnel,
404	respectively. This value is about 10-15% lower than the value observed in the tunnel, but this difference is only
405	statistically significant for $PM_{2.5}$ (p < 0.05). COPERT Australia predicts an average HDV $PM_{2.5}$ and PM_{10}
406	emission factor of 134 and 149 mg/km for the Queensland fleet, and a lower value of 124 and 142 mg/km for the
407	actual fleet mixes in the tunnel, respectively, which is about 10% and 30% lower than the observed values. The
408	difference is statistically significant (p < 0.05) for PM $_{10}$ only. These results indicate that overall prediction errors
409	(under-estimation) for PM are small, but more significant for HDVs. Given the range of factors that complicate
410	validation for PM that were discussed before, these results show a remarkably good performance of COPERT
411	Australia.
412	The analysis of vehicle-class specific prediction errors has shown that largest prediction (%) errors for COPERT
413	Australia are observed for LDVs for the majority of pollutants (CO, NO _x , PM _{2.5}), except for PM ₁₀ where HDVs
414	have the highest (relative) error. PIARC emission factors generally show the largest prediction errors, except for
415	one very good agreement for LDV NO _x . Composite emission factors in COPERT Australia are not significantly
416	different (p < 0.05) from those observed in the tunnel in 25% of the cases. COPERT Australia emission factors
417	for LDVs and HDVs have prediction errors ranging from about 10-40%. It is suggested that high-emitting
418	vehicles in the on-road fleet play a significant role in the underestimation.
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420	3.4 VOC emission factors

Individual hydrocarbons include gas-phase VOCs, gas-phase semi-volatile hydrocarbons (also commonly called SVOCs) and particulate-phase hydrocarbons, where condensation of semi-volatile HCs on aerosols occurs. The exact definition of the hydrocarbons varies in literature and depends on the measurement equipment used. VOCs are roughly defined as being C_1 - C_{12} hydrocarbons, SVOCs as C_{10} - C_{26} (mainly alkanes and aromatics) and particulate phase hydrocarbons as C_{14+} . Analysis of the VOC canisters identified 28 individual VOCs above the limit of detection, which are mainly alkanes, alcohols and aromatics. Table 3 shows the results.

Table 3 – Measured and predicted emission factors (mg/km) for speciated VOCs (± standard error).

Speciated VOC	Tunnel	COPERT Australia	Error
Ethanol	27.0 (±5.6)		_
Acetone	15.9 (±0.9)	0.2	-99%
Toluene	14.0 (±1.0)	4.7	-66%
Xylene (m-& p-)	11.5 (±3.1)	3.9	-66%
2-Methylbutane	7.6 (±0.8)		_
Pentane	6.1 (±0.8)	3.4	-44%
Benzene	5.6 (±0.8)	2.1	-62%
Methylene-chloride	4.7 (±3.6)	_	_
1,2,4-Trimethylbenzene	3.9 (±1.5)	1.1	-71%
Hexane	3.0 (±0.6)	1.1	-62%
Xylene (o-)	3.0 (±1.0)	1.9	-36%
Undecane	2.6 (±1.4)	0.3 ^a	-88%
4-Ethyltoluene	2.5 (±1.5)	_	_
Cyclohexane	2.4 (±0.8)	0.3 ^b	-86%
p-Diethylbenzene	2.4 (±1.4)	-	_
Decane	2.4 (±1.2)	0.2	-93%
Octane	2.2 (±1.3)	0.2	-93%
1,3,5-Trimethylbenzene	2.2 (±1.2)	0.4	-83%
2,2,4-Trimethylpentane	2.2 (±1.2)	_	_
Nonane	2.1 (±1.1)	0.1	-97%
Styrene	2.0 (±1.1)	0.2	-87%
Heptane	1.9 (±1.1)	0.8	-59%
Ethylbenzene	1.8 (±1.0)	2.3	+24%
Acrolein	1.8 (±0.1)	0.3	-85%
1,3-Butadiene	1.5 (±0.1)	0.5	-67%
Isopropylalcohol	1.2 (±0.3)	_	_
Methylethylketone (MEK)	1.0 (±0.2)	0.1	-93%
Methyltert-butylether (MTBE)	$0.7 (\pm 0.0)$	_	_
Σ	135	24	

⁴²⁸ a COPERT Australia category "alkanes C_{10} - C_{12} ", b COPERT Australia category "cycloalkanes"

The difference between COPERT Australia and the tunnel measurements is large. COPERT substantially underestimates emission factors for individual VOCs, typically with a factor of 5, but in some cases an order of magnitude lower.

In addition, 43 VOCs for which COPERT Australia provides emission factors, were either not included (aldehydes; 3.3 mg/km as predicted with COPERT) or were not measured above the limit of detection in the tunnel. As a consequence, only 33% of the sum of speciated VOCs predicted with COPERT Australia is reported in Table 3 (24 mg/km). The sum of speciated VOCs has an observed value of 135 mg/km in the tunnel, which is 46% lower than the *total* value predicted with COPERT Australia (72 mg/km).

The VOC profiles are quite different as shown in Figure 9. The proportion of alcohols in the observations is substantial (21%) and absent in the COPERT predictions. The high observed values are related to the use of E10 in Queensland. The COPERT Australia VOC profile is dominated by alkanes and alkenes (47% and 17%), whereas the tunnel observations have proportions of 22% and 1%, respectively. The aromatics content is more similar with 36% (observed) and 29% (predicted). The observed proportion of ketones (12%) is however substantially higher than the predicted value of 0.4%.

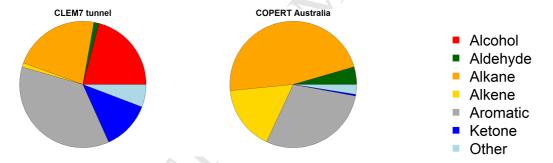


Figure 9 – Proportion of VOCs in fleet emission factors by VOC class as observed in the tunnel ("CLEM7 tunnel") and the complete VOC profile as predicted with COPERT Australia ("COPERT Australia").

 These results demonstrate the large uncertainty in speciated VOC emission factors, and this suggests that further studies to improve VOC profiles and associated emission factors are warranted.

451 3.4 PAHs emission factors

Observed and predicted PAH emission factors are presented in Table 4.

Table 4 – Measured and predicted emission factors (ng/km) for speciated PAHs.

Speciated PAH	Tunnel	COPERT Australia	Error
Napthalene	4,793,031	700,352	-85%
Phenanthrene	5,264	20,841	+296%
Pyrene	2,510	9,405	+275%
Fluoranthene	1,377	10,625	+672%
Anthracene	648	2,094	+223%
Benzo(a)anthrancene	194	1,159	+497%
Chrysene	194	2,599	+1,237%
Benzo(e)pyrene	194	1,814	+833%
Benzo(b)fluoranthene	154	1,094	+611%
Benzo(g,h,i)perylene	134	1,354	+913%
Benzo(a)pyrene	121	682	+462%
Benzo(k)fluoranthene	81	948	+1,071%
Indeno(1,2,3-cd)pyrene	77	767	+897%
Dibenzo(a,h)anthracene	49	132	+173%
Σ	4,804,028	753,869	

COPERT substantially overestimates emission factors for almost all PAHs, typically with a factor of 3-13, except for napthalene, which is underestimated with a factor of 7. In addition, 12 PAHs for which COPERT Australia provides emission factors, were not measured above the limit of detection in the tunnel. As a consequence, only 97% of the sum of PAHs predicted with COPERT Australia is reported in Table 3 (753,896 ng/km). The sum of PAHs has an observed value of 4.80 mg/km in the tunnel, which is 516% higher than the value predicted with COPERT Australia (0.78 mg/km), and due to the discrepancy for naphtalene which makes up the bulk of total PAHs. These results demonstrate the large uncertainty associated with PAH emission factors, and suggests that further studies to improve PAH profiles and associated emission factors are needed.

464	5. Conclusions
465	This paper presents results of a tunnel emissions study that was conducted in Brisbane, Australia, to (partially)
466	validate the Australian vehicle emissions software COPERT Australia and PIARC emission factors. Emissions
467	of NO _x , NO ₂ , PM _{2.5} , PM ₁₀ , CO, VOCs and PAHs generated in the 4.8 km-long tunnel were monitored for 9 days
468	in the north ventilation vent. Other data were collected including traffic counts, license plates, in-tunnel air-flow,
469	speed-time profiles using a smart app, tunnel design maps and background concentrations.
470	Analysis found that the the in-tunnel fleet mix differs substantially from the average on-road fleet, with a larger
471	proportion SUVs and younger vehicles, leading to lower emissions by factor of about 2.
472	The $P\Delta P$ software was run to examine and quantify the combined impact of road gradient, (piston) air-flow and
473	tunnel driving conditions on NO _x emissions. The road gradient effect on in-tunnel emissions is substantial with
474	an approximately 20% increase in NO _x emissions. However, in-tunnel air-flow roughly compensates for road
475	gradient impacts at higher air speeds due to reduced aerodynamic drag. On average, NOx emissions are increased
476	by 1-8%.
477	Typical measured in-tunnel NO2 to NOx ratios were 0.15, which is close to 0.13 predicted with COPERT
478	Australia. The results suggest that the COPERT Australia is generally accurate at fleet level for CO, NO_x , $PM_{2.5}$
479	and PM_{10} , when compared with similar international studies. COPERT underestimates emissions by 7% to 37%,
480	depending on the pollutant. These findings apply only to the specific measurement conditions in the tunnel, i.e. a
481	free-flow speed of about 80 km/h, the particular road gradient profile and ventilation conditions (piston effect)
482	and the specific young fleet mix. As a consequence, these results cannot be used to make generic statements
483	about accuracy of the software. Instead, other studies are required to quantify prediction accuracy in other urban
484	conditions, using for instance remote sensing or near-road air-quality measurements.
485	COPERT Australia composite LDV/HDV hot-running emission factors for CO, NO _x , PM _{2.5} and PM ₁₀ are not
486	significantly different (p $<$ 0.05) from those observed in the tunnel in 25% of the cases. For the other cases,
487	emissions are consistently underestimated by ~ 10-40%, depending on the pollutant and vehicle class. The
488	largest prediction errors are observed for LDVs for the majority of pollutants (CO, NO_x , $PM_{2.5}$), except for PM_{10}
489	where HDVs have the highest (relative) error. It seems plausible that three factors play a role in the
490	underestimation: 1) under-representation of high/excessive-emitting vehicles in the model due to the absence of
491	Inspection and Maintenance (I/M) programs in Australia, 2) lenient vehicle ageing (mileage) correction factors
492	in the COPERT software, and 3) lack of empirical emissions data for Australian diesel LDVs.

Nevertheless, these validation results appear to be relatively good in comparison with other international

494	validation studies. This is particularly the case for PM, which tends to have the lowest prediction errors, despite
495	the range of factors that complicate validation for PM. In comparison with COPERT Australia, PIARC emission
496	factors are conservative and exhibit the largest prediction errors, except for one very good agreement for LDV
497	NO_x .
498	In regard to speciated VOCs, the difference between COPERT Australia and the tunnel measurements is large,
499	with substantially different VOC profiles. COPERT substantially underestimates emission factors for individual
500	VOCs, typically with a factor of 5, but in some cases an order of magnitude lower. COPERT substantially
501	overestimates emission factors for almost all PAHs, typically by a factor of 3-13, except for napthalene, which is
502	underestimated with a factor of 7. These results demonstrate the large uncertainty in speciated VOC and PAH
503	emission factors, which suggests that further studies to improve local VOC and PAH profiles and associated
504	emission factors are required.
505	The results indicate that further targeted emissions testing for diesel vehicles using e.g. PEMS would benefit
506	vehicle emission modelling and air-quality assessments in Australia. Other tunnel datasets in other cities,
507	preferably of longer duration than a week, could be analysed in a similar fashion to see if these results are
508	confirmed.
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