

TNO report

TNO 2014 R11513 CO₂ emission from urea consumption in SCR after-treatment systems in heavy-duty vehicles

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Summary

Since about 2005 SCR (Selective Catalytic Reduction) after-treatment systems for the reduction of NO_x emissions are commonly used in diesel-fuelled heavy-duty vehicles such as Euro V and VI trucks and busses. In an SCR system a urea-based additive solution, commercially available as AdBlue, is used for the catalytic reduction of NO_x. The reduction of NO in the SCR process is performed with ammonia as reducing agent. The ammonia is delivered by the hydrolysis of one molecule of ureum delivering two molecules of ammonia and one molecule of carbondioxide.

Within the framework of the Dutch emission inventory program the need to quantify this new source of CO_2 emission from traffic was felt. Hence, a concise study was performed by TNO to estimate road type specific CO_2 emission factors from the use of urea-additives.

The 2006 IPCC Guidelines for National Greenhouse Gas Inventories include CO_2 emissions from the use of urea-based additives in catalytic convertors as a new source of greenhouse gas emissions (see Ref. [1]). The method described in the 2006 IPCC Guidelines for calculation of CO_2 emissions due to the use of urea-additive based selective catalytic reduction (SCR) of NO_x emissions, is translated into a practical calculation method relating the urea-additive CO_2 emissions to the consumption of urea-additive and diesel fuel.

The trip and driving behavior dependencies of urea-additive consumption are investigated by examining PEMS-based model data for the NO_x/CO_2 ratio of trucks. Some minor dependencies are found, justifying the approach to neglect these for first order estimates of road specific urea-additive CO_2 emission factors.

Combining the developed practical calculation method with the Dutch SRM¹ diesel fuel CO₂ emission factors for Euro-V and Euro-VI trucks, estimated road specific urea-additive CO₂ emission factors are derived (see Table 1). Based on a urea-additive (AdBlue) consumption of 6 vol. % (Euro V) or 3 vol. % (Euro VI) of the diesel fuel consumption, the urea-additive CO₂ emissions are calculated to be 0.6 % or less (Euro V) or 0.3 % or less (Euro VI) of the diesel fuel CO₂ emissions.

It is estimated that the uncertainty margins in urea consumption, and hence in the associated urea-additive CO_2 emission, are 25% on the lower side and 10 to 25% on the upper side.

Arguments are provided that different CO2 emission practical calculation rules have to be developed for light duty diesel vehicles are to be equipped with ammonia based SCR NO_x emission reduction systems.

¹ SRM stands for 'Standaard Reken Methode' and refers to the Dutch standard calculation methodology (SRM 1 and 2) for road type specific traffic emission factors on urban, rural and motorway type roads.

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1 Introduction

The SCR (Selective Catalytic Reduction) after-treatment system for the reduction of NO_x emissions, which is commonly used in diesel fuelled heavy-duty vehicles since 2005, consumes a urea-solution, commercially available as AdBlue. The reduction of NO to N₂ in the SCR process is performed by means of ammonia (NH₃) as reducing agent. The ammonia (NH₃) is formed by the hydrolysis of one molecule of urea: $(NH_2)_2$ CO, delivering two molecules of ammonia (NH₃) and one molecule of carbon dioxide (CO₂).

As the consumption of urea solution by trucks with an SCR system is only a fraction of the consumption of diesel fuel, the CO_2 emission from urea solution is not expected to be a major source of CO_2 by traffic. Nevertheless, within the framework of the Dutch emission inventory program the need to quantify this new source of CO_2 emission from traffic was felt for some time already. Therefore a concise study on this subject was performed by TNO.

This study consists of three parts:

- 1. Firstly (Chapter 2), the IPCC guideline on the calculation of CO₂ from ureaadditive is translated into a practical calculation method by combining it with available data on urea solution consumption for Euro V and VI trucks.
- Secondly (Chapter 3), the trip and driving behavior dependency for CO₂ from urea-additive is investigated using PEMS model data on the NO_x/CO₂ ratio for trucks.
- 3. Finally (Chapter 4), road type specific *CO*₂-*from-urea-additive-emission-factors* are derived by combining the developed practical calculation method for CO₂ from urea-additive with road type specific *CO*₂-*from-diesel-fuel-emission-factors*.

An IPCC guideline in reference [1] prescribes how the CO_2 emission from ureaadditive should be accounted for, separate from the CO_2 emission produced by the diesel fuel combustion. The guideline gives a calculation rule for the CO_2 emission in terms of the amount of urea-additive used. By combining this calculation rule with the available trip averaged data for urea solution consumption in terms of diesel fuel consumption for Euro V and Euro VI trucks, a practical calculation method is developed relating the urea-additive CO_2 emissions to the consumption of ureaadditive and diesel fuel.

2 Calculation methodology for CO₂ from urea

According to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories the equation to calculate CO_2 emissions from urea-based catalytic converters reads (see eq. 3.2.2. on page 3.12 of Ref. [1]):

Emission = Activity
$$\bullet$$
 (12/60) \bullet Purity \bullet (44/12) (1)

Here 'Emission' is the emitted CO_2 mass due to the use of urea for selective catalytic reduction (SCR) of NO_x emissions in combustion engine exhaust gas. I.e. the CO_2 directly coming from the carbon in urea, i.e. the C in $CO(NH_2)_2$. The 'Activity' designates the used mass of urea-based additive and the 'Purity' the mass fraction of urea in the urea-additive. The default purity of urea additives is usually taken as 0.325 according the guideline. This value also holds for a commonly used urea-based additive like AdBlue.

For the calculations to come, equation 1 is rewritten in a more compact form as:

$$\mathsf{E}_{\mathsf{UA}} = \mathsf{M}_{\mathsf{UA}} \bullet \mathsf{MF}_{\mathsf{U}} \bullet (44/60) \tag{2}$$

Where E_{UA} is the emitted CO_2 mass due to the use for urea-additive (UA), M_{UA} the mass of used urea-additive and MF_U the mass fraction of urea therein (i.e. $MF_U = M_U/M_{UA}$).

For vehicles, e.g. trucks and buses, the consumption of urea-additive, as specified or measured, is usually given as a drive averaged volume fraction (VF) with respect to the diesel fuel consumption:

$$VF_{UA} = V_{UA}/V_D \tag{3}$$

Expressed as a volume percentage VF_{UA} ranges from about 2 to 6 % (according to various internet sources, e.g. see <u>http://en.wikipedia.org/wiki/Adblue</u>) for a properly functioning SCR and depends on vehicle type and mass and driving behavior. A non-properly functioning SCR usually uses less urea-additive.

Similarly the consumption of urea-additive can also be defined as a mass fraction (MF):

$$MF_{UA} = M_{UA}/M_D \tag{4}$$

Using the mass densities of urea-additive and diesel fuel and equations 3 and 4, the mass consumption of urea-additive in terms of the mass consumption of fuel follows as:

$$M_{UA} = (\rho_{UA} / \rho_D) \bullet VF_{UA} \bullet M_D$$
(5)

Substituting this last expression for M_{UA} into equation (2) then yields:

$$E_{UA} = (\rho_{UA} / \rho_D) \bullet VF_{UA} \bullet M_D \bullet MF_U \bullet (44/60)$$
(6)

This expression relates the CO₂ emission due to the use of urea-based additive directly to the diesel fuel consumption, two known densities and fractions VF_{UA} and MF_U which are known (MF_U = 0.325) or approximately known (VF_{UA} \approx 0.02 to 0.06).

As an example, the ratio E_{UA}/M_D is calculated from equation (6) while using the following density² and fraction values:

$$\label{eq:rho_UA} \begin{split} \rho_{UA} &= 1090 \ \ kg/m^3 \qquad \rho_D = 832 \ \ kg/m^3 \\ \\ VF_{UA} &= 0.06 \ \ m^3/m^3 \qquad MF_U = 0.325 \ \ kg/kg \end{split}$$

Additionally assuming that $VF_{UA} = 0.06$ and by application of formula 6 the ratio E_{UA}/M_D is calculated to be about 0.0187 kg (or 18.7 gram) CO_2 due to urea-based additive per kg diesel fuel.

Now compare the above example value of the CO_2 emission due to urea-based additive to the CO_2 emission from the combustion of diesel fuel itself. This has an E_D/M_D ratio which is usually (see Ref. [2]) taken as 3.16 kg CO_2 per kg of combusted diesel fuel, i.e.:

$$E_{\rm D} = 3.16^* M_{\rm D}$$
 (7)

Hence, the drive averaged urea-based CO_2 emission is only about 0.6 % (=18.7/3160) or less of the CO_2 emission due to the associated diesel fuel combustion.

² For the density of diesel fuel, see for example Ref. [2]. For the density of AdBlue urea-additive, see for example the AdBlue specification data at <u>http://www.iso22241.org/</u>.

3 Trip and driving behavior dependencies of urea consumption

From PEMS real world driving measurements of CO_2 and NO_x emissions of heavy duty (HD) vehicles like trucks and buses it is well known that these may vary widely depending on vehicle mass and payload, speed and associated road, traffic and driver induced vehicle dynamics (accelerations and decelerations). Though the individual emissions may vary widely, emission ratios usually vary much less. Emission ratios relative to CO_2 have the advantage of being directly related to fuel consumption. Hence, the NO_x/CO_2 emission ratio will be considered here.

Using an emission model derived from real world driving PEMS data sets (see Ligterink et al. in Ref. [3]), the NO_x/CO_2 emission ratio for a truck with a kerb weight of 15 ton, a gross vehicle weight of 30 ton and a vehicle rated power of 281 kW, depending on average vehicle speed (i.e. including dynamics typical for that average speed) and varying payload can be calculated as depicted in Figure 1 (see page 13).

Figure 1 shows that the NO_x/CO_2 ratio is highest at very low average speeds, then more or less constant from 10 to 70 km/h and decreasing beyond that. The NO_x/CO_2 ratio also decreases with increasing payload. The increase at low speeds is limited, i.e. in the order of 20 %, with respect to the more or less constant value between 10 to 70 km/h.

As a plausibility check of these results, it is interesting to see how a certain value of the NO_x/CO_2 emission ratio³, e.g. of say 8 g NO_x per kg CO_2 , relates to the urea-additive which would be needed to fully convert it with SCR. Then assuming for now that all NO_x is converted, the urea-additive consumption relative to the fuel consumption as a volume fraction is readily found as:

$$VF_{UA} = (\rho_D / \rho_{UA}) \bullet (M_{NOx} / E_D) \bullet (M_{UA} / M_{NOx}) \bullet (E_D / M_D)$$
(8)

The stoichiometric mass ratio M_{UA}/M_{NOx} for conversion of NO_x with urea into CO_2 , H_2O and N_2 is known to be about 2 when $MF_U = 0.325$ (see Ref. [4]). This means that for each converted gram of NO_x about two grams of urea-additive (provided $MF_U = 0.325$!) are needed.

Now substituting the following values:

$\rho_D = 832 \text{ kg/m}^3$	$\rho_{UA} = 1090 \text{ kg/m}^3$	
$M_{NOx}/E_D = 0.008 \text{ kg/kg}$	$M_{UA}/M_{NOx} = 2.06$ kg/kg	$E_D/M_D = 3.16 \text{ kg/kg}$

into equation (7) yields $VF_{UA} \approx 0.04$ or 4 %. This is in good agreement with the empirically known consumption range of urea solution specified as 2 to 6 vol. % of the fuel consumption.

 $^{^3}$ Note that in the notation previously introduced, E_D refers to the CO_2 emission due to diesel fuel consumption of diesel fuel mass M_D . Hence, the NO_x/CO_2 mass ratio, or M_{NOx}/M_{CO2} , is in this notation written as M_{NOx}/E_D .

Finally, note that the stoichiometric mass ratio M_{UA}/M_{NOx} assumes 100 % NO_x conversion efficiency. Hence, it gives a theoretical lower limit for the urea-additive usage necessary to convert a certain amount of NO_x and the actual usage will be somewhat higher. Using an M_{UA}/M_{NOx} ratio of 2 (i.e. $MF_U = 0.325$!) together with equation (1), this lower limit and the associated CO₂ emission can be elegantly expressed as follows:

Where the NO_x and CO_2 of course refer to the NO_x converted and the CO_2 caused by this conversion with urea solution.

Based on the previously shown moderate dependency of the NO_x/CO₂ emission ratio and the plausibility check, it seems justified to use constant, i.e. independent of road type, relative⁴ urea-additive consumption values as first order estimates of the SRM⁵ road type specific urea-additive CO₂ emission factors.

In a previous communication (see Ref. [5]), the SRM⁵ emission factors for CO_2 and NO_x for sets of heavy duty Euro V (with SCR) and VI trucks (with SCR and EGR) have been reported (based on measurements in 2012 and 2013). In Table 1 (see page 14) the CO_2 emission factors per SRM road type are given for the same vehicle types but now using the recently published 2014 SRM values (see Ref. [6] or Ref. [7]).

Using equation (7) the fuel consumption mass M_D , per km in this case, is readily calculated from the CO_2 from diesel fuel emission factors given in Table 1. Next, using equation (6) the CO_2 emission factors from urea-additive then easily follow as given in the table for road type WT3 (motorway). Please note the used parameter values given at the bottom of the table. Also note that the urea-additive consumption, expressed as volume fraction MF_{UA} was set to 0.06 for Euro V trucks and to 0.03 for Euro VI trucks⁶. I.e. assuming optimal SCR and maximum urea-additive consumption. For Euro VI trucks this was done for all road types but for Euro V trucks only for WT3. For Euro V and WT1 and WT2 corrections were made as explained in the following.

It seems likely that especially for road types WT1 (urban) and WT2 (rural) and Euro V trucks, an assumed urea-additive consumption of 0.06 (i.e. $VF_{UA} = V_{UA}/V_D = 0.06$) is too high. During WT1 type trips, and a to a lesser extent during WT2 trips, the engine of Euro V trucks (without EGR) will get less warm as a result of the lower engine loads and hence the SCR will more often be shut down by the temperature sensor. This results in lower urea-additive usage and a (relatively) higher NO_x emission. The significantly higher NO_x emission factors of the Euro V trucks for especially road type WT1, see Table 1, seem to confirm this.

It is therefore assumed that the fixed urea-additive consumption of 0.06 for Euro V trucks is a fair estimate for road type WT3 (motorway) and that the CO_2 and NO_x emission factor values per road type with equation (9) may be used to calculate corrected urea-additive CO_2 emission factors in the following way:

$$E_{UA_corr} = (\rho_{UA} / \rho_D) \bullet VF_{UA} \bullet M_D \bullet MF_U \bullet (44/60) - E_corr$$
(10a)

⁴ Relative to the diesel fuel consumption.

⁵ SRM stands for 'Standaard Reken Methode' and refers to the Dutch standard calculation methodology (SRM 1 and 2) for road type specific traffic emission factors on urban, rural and motorway type roads.

⁶ These representative values of AdBlue consumption for Euro V and Euro VI were taken from real world AdBlue consumption data from unpublished TNO research.

Note that the correction value E_corr is zero for WT3 and hence equation (10a) is then identical to equation (6). From E_{UA_corr} , a corrected urea-additive volume consumption VF_{UA_corr} can be readily calculated as:

$$VF_{UA \text{ corr}} = (\rho_D / \rho_{UA}) \bullet (60/44) \bullet (1/MF_U) \bullet (E_{UA \text{ corr}} / M_D)$$
(11)

Urea-additive consumption uncertainties

Of course the SCR urea-additive consumption, i.e. the used volume fractions of 0.06 and 0.03 for Euro V and Euro VI trucks, and hence the associated CO_2 from urea-additive emission have uncertainty margins. The currently available data on urea solution consumption of SCR equipped HD vehicles is not sufficient for a reliable quantitative estimation of these uncertainty margins. A qualitative approach to estimate these margins is as follows.

The lower limit for urea solution consumption is zero when the SCR is either defective or out of urea solution, in which case the urea associated CO_2 emission is zero as well. As this is not likely to occur permanently, it is estimated that the lower uncertainty limit is 25 % at maximum.

On the upper side, overdosing of urea solution is not very likely to occur for two reasons. First of all overdosing costs money as more urea solution is used than necessary. Secondly, overdosing will likely to be detected soon as it leads to NH_3 emission, for which strict legislative emission limits hold and which is easily smelled or measured when setting or testing an SCR equipped HD engine. Hence, it is estimated that the upper uncertainty limit is 10 to 25 % at maximum.

Summarizing, it is estimated that the uncertainty margins in urea solution consumption, and hence in the associated CO_2 -from-urea solution emission, are 25 % on the lower side and 10 to 25 % on the upper side.

SCR developments for LD

Looking at the technology developments for diesel fuelled light duty (LD) vehicles, there seem to be two trends in NO_x reduction.

The first trend is that LD diesel vehicles are equipped with an SCR NO_x emission reduction system and urea solution in a canister, of say 20 liters volume, which should hold enough urea solution to last the period between services at say 20,000 km intervals. Thus the associated urea solution consumption in terms of the Urea solution/diesel volume fraction would be in the order of 0.02, i.e. somewhat less than for HD vehicles. Hence, for SCR equipped diesel vehicles the CO_2 -from-urea solution emission relative to the CO_2 -from-diesel emission is expected to be about 0.2 %.

The second trend is that LD diesel vehicles are equipped with NH_3 storage (safely stored in some solid state crystal structure) for NO_x emission reduction by direct NH_3 injection. In this case the associated CO_2 emissions due to NO_x reduction would of course be zero.

5 Conclusions

From the presented material the following conclusions can be drawn.

- The IPCC guideline (in Ref. [1]) for the calculation of the CO₂ from the use of urea-additive for selective catalytic reduction (SCR) of NO_x emissions from diesel combustion engines was readily translated into a practical calculation method relating the urea-additive CO₂ emissions to the ureaadditive and diesel fuel consumptions (see equation (6)).
- 2. Using the practical calculation method and two fixed urea-additive consumptions, i.e. 6 vol.% (w.r.t. fuel) for Euro V and 3 vol.% for Euro VI, the urea-additive CO₂ emission factors of Euro V and Euro VI trucks were estimated by the diesel fuel CO₂ emission factors for WT3 and from these for WT1 and WT2 using their diesel fuel CO₂ and NO_x emission factors for a correction of the urea-additive CO₂ emission factor and urea- additive consumption (see Table 1).
- Relative to the associated diesel fuel CO₂ emission the urea-additive CO₂ emission is estimated to be 0.6 % or less for Euro V trucks and 0.3 % or less for Euro VI trucks.
- 4. It is estimated that the uncertainty margins in urea solution (commercially available under the name "AdBlue") consumption, and hence in the associated urea-additive CO_2 emission, are 25 % on the lower side and 10 to 25 % on the upper side.
- 5. When light duty diesel vehicles are to be equipped with urea-additive based SCR NO_x emission reduction systems the urea-additive CO₂ emission relative to the diesel fuel CO₂ emission is expected to be about 0.2 % or less. If instead NO_x emission reduction by direct NH₃ injection from stored NH₃ is to be used, this will not generate any extra CO₂ emissions. Amminex from Danmark, for example, uses such an NH₃ storage technology.

6 References

[1] 2006 IPCC Guidelines for National Greenhouse Gas Inventories, Volume 2 Energy.

[2] JRC, 2007, WELL-TO-WHEELS ANALYSIS OF FUTURE AUTOMOTIVE FUELS AND POWERTRAINS IN THE EUROPEAN CONTEXT (TTW Report 010307).

[3] Ligterink, 2012, A velocity and payload dependent emission model for heavyduty road freight transportation, Transportation Research Part D 17 (2012), pp. 487-491.

[4] Relatie_Adblue-FC-NOx_RVB.xls. TNO internal document.

[5] Kuiper et. al., 2013, Verwachtingen voor heavy duty emissiefactoren Euro-V/VI 2013, PowerPoint presentatie "Heavy duty emissiefactoren 2013 v2.pptx".

[6] Detailemissiefactoren_SRM_20140212_all.xlsx. TNO internal document.

[7] Klein et al., 2014, Methods for calculating the emissions of transport in the Netherlands, May 2014, Report and Excel table with data. See http://www.cbs.nl/en-GB/menu/themas/natuur-milieu/methoden/dataverzameling/overige-dataverzameling/2014-methods-for-calculating-the-emissions-of-transport-in-the-netherlands-pub.htm.

7 Figure and Table



Figure 1 NO_x/CO₂ ratio for a truck of 15 ton with maximum 15 ton payload and a vehicle rated power of 281.25 kW.

VERSIT+ Vehicle Class	CO2	CO2 from fuel in g/km		CO2 from AdBlue in g/km			NOx in g/km			AdBlue Consumption in vol. %		
	WT1	WT2	WT3	WT1	WT2	WT3	WT1	WT2	WT3	WT1	WT2	WT3
MVADEDESSCRICH	510	339	287	2.2	16	17	4.65	2.76	1.68	4.4	4.8	6.0
MVADEDE5SCR7WA	1023	680	568	4.4	33	3.4	8.98	5.24	3 16	4,4	4,0	6.0
MVADEUG5SCRICH	447	294	254	19	1.4	15	4 66	2 67	1 77	43	4,5	6.0
MVADEUG5SCRZWA	922	601	502	4,1	3,1	3,0	8,57	4,85	3,22	4,5	5,2	6,0
ZVADEDE5SCR	1426	947	770	6,2	4,7	4,6	11,73	6,61	3,88	4,4	5,0	6,0
ZVADEDE5ANHSCRLCH	1260	811	719	5,6	4,4	4,3	9,08	4,20	3,07	4,5	5,5	6,0
ZVADEDE5ANHSCRZWA	1754	1126	989	7,7	6,1	5,9	11,88	5,29	3,64	4,4	5,5	6,0
ZVADEUG5SCR	1344	860	682	6,5	4,6	4,0	10,20	5,64	3,72	4,9	5,4	6,0
ZVADEUG5ANHSCRLCH	1272	763	632	6,8	4,4	3,7	6,32	3,16	2,43	5,4	5,9	6,0
ZVADEUG5ANHSCRZWA	1843	1077	870	10,1	6,3	5,2	7,09	3,40	2,57	5,5	5,9	6,0
ZTRDEDE5SCRLCH	1406	933	760	6.1	4.6	4.5	11.62	6.57	3.86	4.4	5.0	6.0
ZTRDEDE5SCRZWA	2096	1388	1039	9.0	7.2	6.2	13.66	6.61	3.34	4.3	5.2	6.0
ZTRDEUG5SCRLCH	1320	846	673	6.4	4.5	4.0	10.18	5.64	3.72	4.9	5.4	6.0
ZTRDEUG5SCRZWA	2245	1368	924	12,2	8,0	5,5	7,03	3,10	2,00	5,5	6,0	6,0
MVADEURGICH	393	273	241	12	0.8	0.7	0.28	0.19	0 17	3.0	3.0	3.0
MVADEUR6ZWA	818	557	479	2,4	1,6	1,4	0,58	0,39	0,34	3,0	3,0	3,0
ZVADEUR6	1208	798	656	3.6	2.4	1.9	0.86	0.56	0.46	3.0	3.0	3.0
ZVADEUR6ANHI CH	1170	721	617	3.5	21	1.8	0.83	0.51	0.44	3.0	3.0	3.0
ZVADEUR6ANHZWA	1708	1018	858	5,5	3,0	2,5	1,21	0,72	0,61	3,0	3,0	3,0
	1186	785	647	35	23	10	0.84	0.56	0.46	3.0	3.0	3.0
ZTRDEUR6ZWA	2089	1275	911	6,2	3,8	2,7	1,48	0,90	0,40	3,0	3,0	3,0
	rho UA	rho D	VF UA		MFU			M D/F D				
	kg/m^3	kg/m^3	m^3/m^3		kg/kg			,				
	1090	832	0,06	Euro V	0,325	0,018734	Euro V	0,316456				
			0,03	Euro VI		0,009367	EuroVI					
	For Furo V trucks, the CO2 from AdBlue for WT3 was calculated with equation (6) and for WT1 and WT2 with equation (10)											
	For Euro V	/I trucks. th	e CO2 from	AdBlue w	as calcula	ted with ea	uation (6)	for all road	types.			
	The AdBlue consumption given in the last three columns was calculated with equation (11).											
			-					•				

Table 1 Road type specific CO2 emission factors caused by urea consumption

Note that the urea CO_2 emission factors were calculated from the diesel fuel CO_2 emission factors using equation (6) for Euro VI trucks for all road types, and for Euro V trucks with equation (6) for road type WT3 with equation (10) for road types WT1 and WT2.

Road types WT1, WT2 and WT3 refer to urban, rural and motorway roads as used in the Dutch standard calculation methodology SRM 1 and 2 for road type specific emission factors.

8 Signature

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