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Item 3 (d) of the provisional agenda
Methodological issues
Issues relating to greenhouse gas inventories

Estimation of emissions from road transport

Note by the secretariat*

Summary

Road transport is an important source of greenhouse gas emissions (primarily carbon dioxide and nitrous oxide) for a number of Parties included in Annex I to the Convention. The *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories* (1996 IPCC Guidelines), as elaborated by the *IPCC Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories* (IPCC good practice guidance), contain methodologies that are being used by many Parties to estimate emissions from road transport. These methodologies range from tier 1 methods using default emission factors to complex models using country-specific emission factors. Although the 1996 IPCC Guidelines and the IPCC good practice guidance have proved to be useful tools for the estimation of emissions from road transport, some areas for further improvement of the IPCC methodologies have been identified. This document contains suggestions that could be considered by the IPCC in its work on the development of the *2006 IPCC Guidelines for National Greenhouse Gas Inventories and by Parties in the preparation of national GHG inventorie*s and by Parties in the preparation of national greenhouse gas inventories.

* This note was prepared by the secretariat on the basis of input provided by Mr. Wiley Barbour and Mr. Michael Gillenwater.

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I. Introduction

A. Mandate

1. The Subsidiary Body for Scientific and Technological Advice (SBSTA), at its seventeenth session, invited the Intergovernmental Panel on Climate Change (IPCC) to revise the *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories* (1996 IPCC Guidelines) taking into consideration the relevant work under the Convention and the Kyoto Protocol, and to aim to complete the work by early 2006.[1] In response, the IPCC initiated this work in 2003 and agreed on the terms of reference, table of contents and work programme for the development of the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (2006 IPCC Guidelines).

2. The SBSTA, at its nineteenth session, considered the initial information on methodological issues relating to the preparation of national greenhouse gas (GHG) inventories by Parties, contained in document FCCC/SBSTA/2003/INF.10, and decided to forward it to the IPCC for consideration. It also requested the secretariat to continue to cooperate with the IPCC and provide more detailed information based on the latest available GHG inventory submissions by Parties and the results of the technical review of GHG inventories. Such information could serve as input to the planned IPCC meetings that will take place during the development of the 2006 IPCC Guidelines.[2]

B. Scope of the note

3. This note addresses methodological issues relating to the estimation of GHG emissions from road transport, focusing in particular on nitrous oxide (N_2O) emissions.[3] It provides an overview of the mechanics of N_2O formation, brief descriptions of models that are being used by Parties to estimate emissions from road transport, and information on the results of research on the development of emission factors for N_2O emissions from transport.

C. Possible action by the Subsidiary Body for Scientific and Technological Advice

4. The SBSTA is invited to consider the information in this note and forward it to the IPCC for its consideration. Parties may wish to consider the information in this note when preparing their national GHG inventories.

II. Background

5. The direct GHGs emitted by road transport activities are carbon dioxide (CO_2), methane (CH_4), N_2O, and the hydrofluorocarbon HFC-134a. Essentially all Parties to the Convention estimate emissions of CO_2, CH_4, and N_2O in a similar fashion, which involves the multiplication of some form of activity data (e.g., quantity of fuel consumed or vehicle kilometres travelled) by emission factors per unit of activity.[4]

6. Emission factors for estimating CO_2 emissions from fossil fuel combustion for road transport are based on a combination of each fuel's carbon contents and an adjustment factor for the fraction of that carbon that remains unoxidized (i.e., post-combustion residual soot or ash). For all typical fuel

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[1] FCCC/SBSTA/2002/13, paragraph 14 (f).

[2] FCCC/SBSTA/2003/15, paragraphs 17 (a) and (c).

[3] Methodological information on other sectors is provided in documents FCCC/SBSTA/2004/INF.2, which deals with fugitive emissions from fuels and FCCC/SBSTA/2004/INF.4, which deals with emissions from agriculture.

[4] Although not the focus of this paper, the quality of the activity data collected by a Party is, in most cases, the most important variable affecting the quality of its final emission estimates for road transport, given that default emission factor data are available and country-specific values are not likely to diverge greatly from defaults.

combustion technologies, when averaged nationally, the fraction oxidized is unlikely to vary outside the range of 97–100 per cent. Carbon content of specific fuel types also does not vary greatly, as fuels tend to be fairly uniform in their chemical compositions and the amount of carbon can be determined by basic chemical analysis or stoichiometry. When normalized by the fuel's energy content, the variation in carbon content for a given fuel type is reduced even further. In general, the overall uncertainty in the factors used to estimate CO_2 emissions from road transport is unlikely to be very large and will be primarily associated with the quality of the activity data used.

7. Unlike CO_2 emissions from fuel combustion, CH_4 and N_2O emissions from road transport are less easily estimated. In addition to activity data, CH_4 emissions are a function of a number of factors that can vary considerably, such as combustion conditions, post-combustion emission controls, fuel composition, and driving practices. Similarly, N_2O emission rates can vary dramatically and have been found to be primarily functions of the type and operating temperature of catalytic emission control equipment, which can be affected by several variables.

8. Although there are many complex factors that affect the emission rates for both CH_4 and N_2O from road transport vehicles, CH_4 emission factors have been more thoroughly studied because of the concern over hydrocarbon emissions and their role in the formation of ground-level (tropospheric) ozone. Emissions of non-methane hydrocarbons (NMHCs) or non-methane volatile organic compounds (NMVOCs) have been the focus of most research. Methane, unlike most other hydrocarbons, does not contribute very much to ozone formation. In the process of developing emission factors and speciation profiles for hydrocarbon emissions, much insight has been gained into the mechanics of CH_4 emissions from road transport and appropriate emission factors for estimating CH_4emissions.

9. In contrast to the number of measurements that have been performed of pollutants regulated in many countries (e.g., carbon monoxide (CO), nitrous oxides (NO_X) and hydrocarbons), fewer measurements have been made of N_2O emissions from road vehicles. Emissions of N_2O are not regulated in most countries, and no Party is known to have mandatory regulations currently in place limiting emissions of N_2O from road transport.[5] The N_2O emission factors for road transport that are available are generally considered highly uncertain – for application to national inventories – and have not been developed at a high level of detail. The small number of measurements that have been performed on emissions of N_2O from road vehicles have shown that emissions are dominated by the formation of N_2O in catalytic converter systems. It has been found that little or no N_2O is typically produced in automotive internal combustion engines. Most of the N_2O emitted is formed within the catalyst bed (Riemersma et al., (2003b)). Catalyst systems control tailpipe emissions of NO_X (i.e., nitric oxide (NO) and NO_2) by catalytically reducing NO_X to N_2. Sub-optimal catalyst performance, primarily at lower temperatures, results in incomplete reduction and the conversion of some NO to N_2O rather than to N_2. The design of three-way catalysts, which are used on most modern light-duty vehicles with post-combustion emission controls in Parties included in Annex I to the Convention, is determined mainly by the need to reduce NO_X emissions.

10. Because of the relatively greater uncertainty associated with N_2O emission factors, they are the focus of this note. Emissions of hydrofluorocabons (HFCs) and chlorofluorocarbons (CFCs) from mobile refrigeration and air conditioning systems are not addressed here.

[5] California Assembly Bill 1493 (Pavley) was passed in 2002 and requires the California Air Resources Board (CARB) to develop greenhouse gas emission standards for vehicles, applicable beginning with model year 2009. The bill requires CARB to develop and adopt regulations by 2005. http://www.arb.ca.gov/cc/ab1493.pdf

III. Mechanics of nitrous oxide formation

11. Nitrous oxide has not been found to be an important component of exhaust gases from most internal combustion engines used in road transport vehicles (Feijen-Jeurissen et al., (2001); Gense and Vermeulen, (2002)). The temperatures and pressures within internal combustion engines have not been found to be favourable to producing N_2O. Instead N_2O is primarily formed through reactions of the exhaust gas constituents with post-combustion catalysts found in emission control equipment.

12. Three-way catalysts are designed to reduce NO to N_2 through reactions with CO, hydrocarbons, and a metal catalyst. This conversion is a temperature-dependent process, generally starting at around 200ºC, peaking at around 90 per cent at about 350ºC and holding quite steady at higher temperatures.

13. The generation of N_2O inside the three-way catalyst occurs within a particular temperature range when NO molecules associate with individual nitrogen atoms on the surface of the metal catalyst instead of two nitrogen atoms associating with each other and forming N_2. Nitric oxide molecules can only adsorb onto the catalyst surface at lower temperatures. At higher temperatures NO dissociates into separate N and oxygen atoms. Therefore, higher catalyst temperatures favour the formation of molecular nitrogen over N_2O both through limits on the formation of N_2O in the first place and through the decomposition of the N_2O further along the catalyst bed.

14. The temperature window within the three-way catalyst bed that has been found to be optimal for N_2O formation is typically 250–400ºC. Operating conditions that tend to maintain the catalyst within this temperature window will thus tend to produce greater N_2O emissions (Odaka et al., (2002)). Catalysts typically pass through this temperature window after an engine start-up. Other factors, such as cold starts or low ambient air temperatures (<20˚C), may also cause the catalyst to operate in this temperature window for a longer period of time.

15. Odaka et al. (2002) found that N_2O generation begins when the three-way catalyst bed temperature reaches 150°C and thereafter increases approximately linearly as temperature rises, peaking at around 320°C. As the temperature rises further, the theoretical reaction conditions are less favourable to the formation of N_2O and some of the N_2O that does form may decompose, thereby leading to minimal N_2O emissions at temperatures beyond 500°C (see figure 1). Basically, the more time that a catalyst bed operates in the 200–500°C temperature range, especially between 280 to 350°C, the greater the N_2O emissions from the vehicle. However, under more realistic operating conditions, when oxidizing conditions exist within the catalyst, N_2O may continue to be emitted even at higher catalyst bed temperatures (Gense and Vermeulen, (2002)).

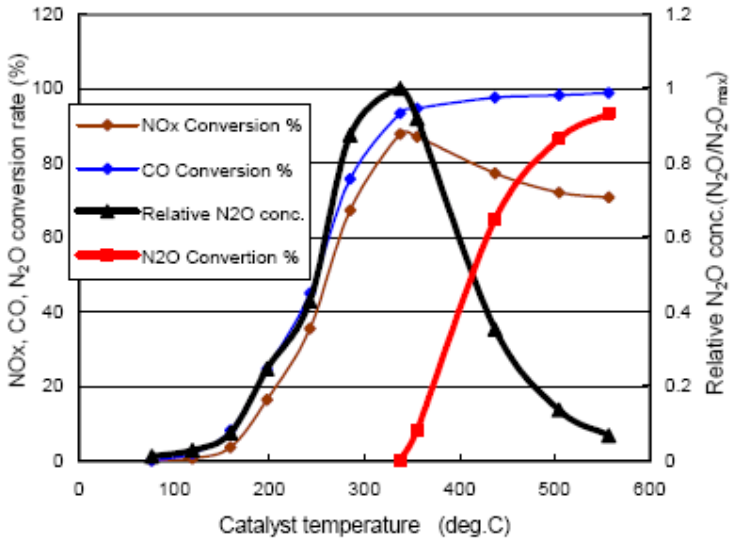
16. Oxidation catalysts – used in heavy-duty vehicle engines and older vehicles – and platinum (Pt) catalysed particulate filters (e.g., Continuously Regenerating Technology (CRT) filter and catalytic soot filter (CSF)) are also expected to produce N_2O (Riemersma et al., 2003b). SCR-deNO_x catalysts that use urea or ammonia (NH_3) with an active and selective catalyst (e.g., titania-vanadia) are not expected to produce N_2O at temperatures below 450º C; however, above that temperature NH_3 could be oxidized into N_2O (Riemersma et al., (2003b)).

17. For a more thorough discussion of the mechanics of N_2O formation and conversion in catalysts see Koike and Odaka (1996); Koike et al. (1999); Lipman and Delucchi (2002); Meffert et al. (2000); Odaka et al. (1998); Odaka et al. (2002) and Riemersma et al. (2003b).

18. A brief discussion on some of the factors that potentially affect N_2O emissions from road vehicles is contained in annex I to this note. The annex focuses on recent research (since 1998). For a detailed discussion of the literature on N_2O emissions from road transport vehicles see Feijen-Jeurissen

et al. (2001). Riemersma et al. (2003a) provide a detailed review of emissions from heavy-duty diesel vehicles. Lipman and Delucchi (2002) present a summary of research findings from testing up to 1998.

Figure 1: Relationship between catalyst bed temperature and N_2O formation



Source: Odaka et al. (2002).

IV. Mobile source emission models

19. In this chapter, mobile source emission models for the transport sector, such as the MOBILE and MOVES models of the United States Environmental Protection Agency (EPA) and the COPERT model of the European Environment Agency (EEA) are discussed. These models have historically focused on estimating emissions of regulated air quality pollutants and not direct GHGs. The purpose of this chapter is to discuss how these models currently address GHG emission factors and what improvements are planned for the immediate future.

A. MOBILE

20. The MOBILE model is a software program that provides estimates of current and future emissions from highway motor vehicles. MOBILE6 is the latest in a series of MOBILE models that date back to 1978. The most recent version, MOBILE6.1/6.2, calculates average in-use fleet emission factors for hydrocarbons, CO, NO_X, exhaust particulate matter (which consists of several components), tyre wear particulate matter, brake wear particulate matter, sulphur dioxide (SO_2), (NH_3,) six hazardous air pollutants (HAP), and CO_2 for gasoline-fuelled and diesel highway motor vehicles and for certain specialized vehicles such as natural-gas-fuelled or electric vehicles that may replace them. It bases these emission factors on vehicles from the 25 most recent model years, and is capable of developing factors for calendar years between 1952 and 2050.

21. MOBILE6.1/6.2, released in 2003, was the EPA's first highway mobile source emissions model to include estimates of CO_2 emissions. However, the method and data for estimating CO_2 emission factors are "very simplistic" according to the EPA. MOBILE6.1/6.2 does not include the capability to estimate N_2O or directly estimate CH_4 emission factors, but it is capable of estimating CH_4 emission factors in some cases. The EPA plans to include non-CO_2 GHGs (CH_4 and N_2O) and improved factors for CO_2 in MOBILE6.3.

22. Emission factors for CO_2 in MOBILE6.1/6.2 are based on fuel economy performance estimates built into the model or supplied by the user. These fuel economy estimates assume that all of the carbon in the fuel is oxidized to CO_2, including carbon initially emitted as hydrocarbons or CO, but it does not account for the fraction remaining unoxidized as particulate matter, soot, or ash. The IPCC default value for the fraction of petroleum fuels left unoxidized during combustion is 1.5 ± 1 per cent by mass.

23. The factors for hydrocarbons, CO, NO_X, particulate matter (PM) and air toxics in MOBILE6.1/6.2 are "final" according to the EPA; however, the fuel economy and CO_2 estimates produced by the model are still considered draft. EPA (2002) discusses the current fuel economy estimates used in the model.

24. Although fuel economy is known to be affected by a variety of factors, the estimates of fuel economy in MOBILE6.1/6.2 and MOBILE6.3 depend only on vehicle type and model year. Therefore, unlike most other MOBILE6 emission factor estimates, the CO_2 emission factor estimates are not adjusted for speed, temperature, fuel content, or the effects of vehicle inspection maintenance programmes. This means that MOBILE6.1/6.2 cannot be used to model the effects of these parameters on CO_2 emissions. Instead, the EPA intends for MOBILE6.3 to be used to model large-scale (e.g., national annual) emissions for which CO_2 variations due to variations in factors such as vehicle speed and ambient temperatures can reasonably be expected to "average out".

25. Although MOBILE6.1/6.2 does not directly estimate CH_4 emission factors, it can be used to do so for some categories where total hydrocarbons and NMHC estimates are available. Methane estimates can be developed by subtracting total hydrocarbons and NMHC values. However, care should be taken in doing so because these factors were not developed for the purpose of estimating CH_4 emissions. The EPA also warns that total hydrocarbons and NMHC outputs from the model for natural gas vehicles should not be subtracted from each other to estimate CH_4 emissions. This limitation should be solved with the release of MOBILE6.3, which will explicitly include both CH_4 and N_2O emission factor estimates.

26. For more information on the calculation procedures in the MOBILE6 model see EPA (2003) and http://www.epa.gov/otaq/models.htm.

B. MOVES

27. The United States EPA is also developing, in parallel to MOBILE6, a new generation mobile source emission model called the Motor Vehicle Emission Simulator (MOVES). This new platform will model both on-road and non-road emission sources and will address all air quality pollutants addressed in the MOBILE model. When complete, MOVES will also support the development of emission inventories and projections at the country-level for CO_2, CH_4, N_2O, and air-conditioner refrigerants (e.g., HFC-134a).

28. MOVES will calculate emission inventories – a step that is currently performed outside the MOBILE model. The MOBILE model produces only emission factor estimates. It does not handle activity data and, therefore, cannot produce actual inventories. The MOVES model will take into account factors such as vehicle speed, effects of inspection maintenance programmes, extent of air conditioning usage, ambient temperature, roadway types, roadway grades, and vehicle weight (including load) to the extent that those factors are found to be relevant.

29. The schedule for the development of the MOVES model is summarized below:

(a) MOVES2004 (planned completion mid-2004)

(i) Draft vehicle fleet and activity data components

(ii) Draft on-road fuel consumption and CO_2, CH_4, and N_2O emissions

(iii) United States county level inventories for 1999 forward

(b) MOVES2005 (planned completion early 2005)

(i) Draft emission estimates for aircraft, commercial marine transport, and locomotives

(c) MOVES2006 (planned completion early 2006)

(i) Final emission estimates for aircraft, commercial marine transport, and locomotives

(ii) Draft on-road emission estimates for criteria pollutants

(d) MOVES2007 (planned completion mid 2007)

(i) Final on-road model

(ii) Draft non-road model.

30. More information on the status of the MOVES model can be found at http://www.epa.gov/otaq/ngm.htm.

C. COPERT

31. The COPERT model was financed by the EEA, under the framework of the activities of the European Topic Centre on Air and Climate Change. COPERT was primarily developed to estimate emissions from road transport for annual national inventories. The current version of the model, COPERT III, has been available since 2000.

32. COPERT estimates emissions of all regulated air pollutants (e.g., CO, NO_X, volatile organic compounds (VOC), and particulate matter) produced by different vehicle categories (passenger cars, light-duty vehicles, heavy-duty vehicles, mopeds, and motorcycles) as well as CO_2 emissions on the basis of fuel consumption. Emissions are also calculated for other non-regulated pollutants, including CH_4, N_2O, NH_3, SO_2, heavy metals, polycyclic aromatic hydrocarbons (PAHs) and persistent organic pollutants (POPs). Additionally, the model provides NMVOC emissions allocated to several individual species.

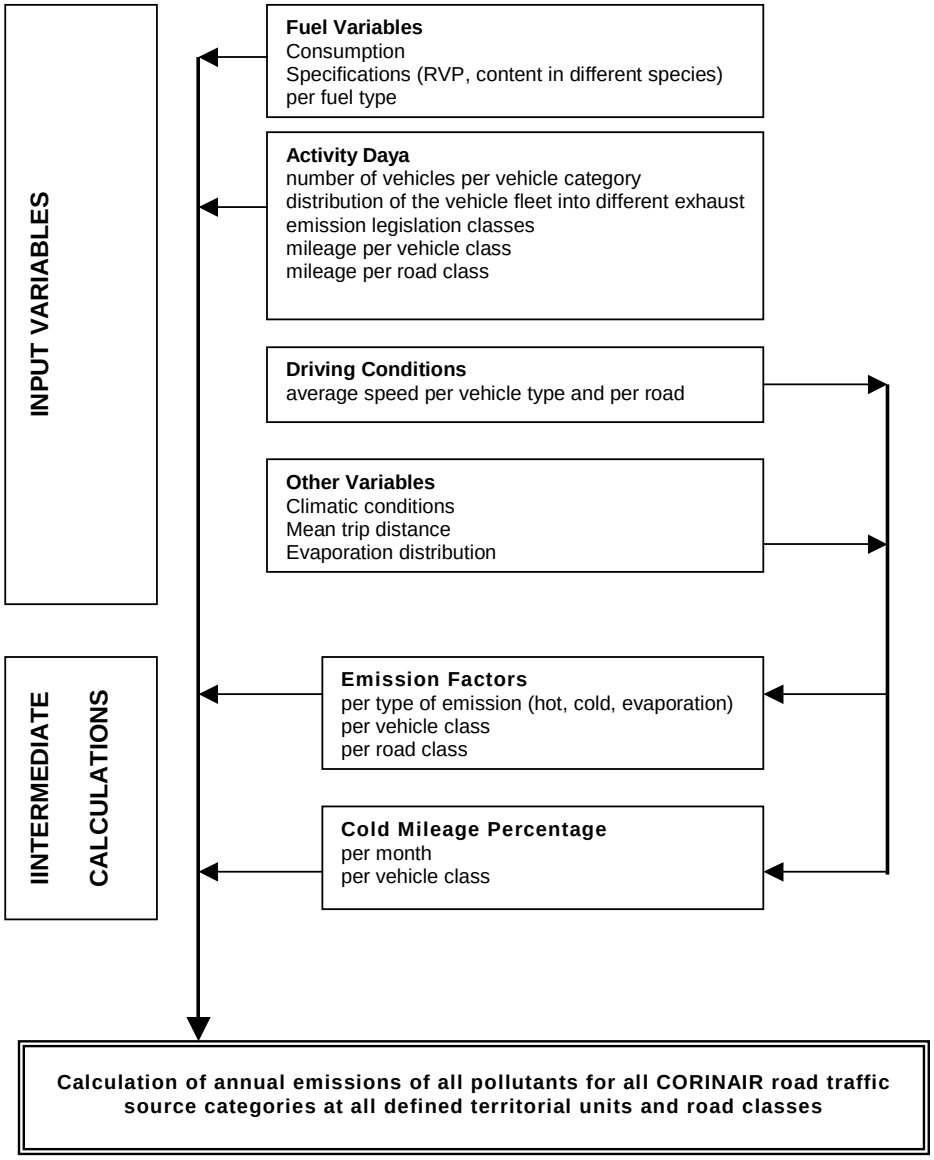
33. Emissions in the model are estimated from three general processes: emissions produced during thermally stabilized engine operation (hot emissions); emissions occurring during engine start from ambient temperature (cold-start and warming-up effects); and NMVOC emissions due to fuel evaporation. Total emissions are calculated as a product of activity data provided by the user and speed-dependent emission factors calculated by the model.

34. The model also distinguishes between urban, rural and highway driving to account for variations in driving performance. Different activity data and emission factors are attributed to each driving situation. Cold-start emissions are attributed to urban driving because the assumption is made that most vehicles start any trip in an urban area. An overview of the factors captured in the COPERT model is provided in figure 2.

35. Like the MOBILE model, COPERT bases CO_2 emissions on fuel consumption and assumes that all the carbon in the fuel is fully oxidized into CO_2 (i.e., it does not address the fraction left unoxidized).

However, unlike MOBILE, COPERT's fuel consumption factors, and therefore CO_2 emission estimates, are speed-dependent.

Figure 2: Overview of the COPERT model



Source: EEA (2000).

36. The CH_4 emission factors for light-duty vehicles in COPERT depend on driving mode (urban, rural, and highway) and speed. The CH_4 factors for heavy-duty vehicles, however, are not speed-dependent.

37. Emission factors for N_2O in COPERT are not speed-dependant and vary by driving mode only for light-duty gasoline vehicles. Cold-start emissions are not estimated separately, but are assumed to be incorporated in the bulk emission factors.

38. More information on COPERT can be found at http://vergina.eng.auth.gr/mech/lat/copert/copert.htm.

D. Comparing models

39. MOBILE is designed to estimate only emission factor values. It does not include methods for handing vehicle activity data, and therefore does not actually produce emission estimates. Emission estimates are calculated outside the MOBILE model. COPERT is capable of handling both activity data and emission factors. When complete, MOVES will include activity data, and therefore be more similar to the COPERT model.

40. Overall, however, both MOBILE/MOVES and COPERT use essentially the same methodology to estimate historical and projected GHG emissions. Currently MOBILE does not explicitly estimate CH_4 and does not estimate N_2O emissions. Both models estimate CO_2, based on fuel consumption factors, but MOBILE's data are preliminary and it does not account for vehicle driving practices (speed). Neither model addresses the fraction of carbon left unoxidized when estimating CO_2 emissions.

V. Comparison of nitrous oxide emission factors

41. A summary of selected N_2O emission factor values used by Parties to the UNFCCC or available in recent research publications is presented in table 1. An attempt was made in this table to arrange factors such that they can be readily compared with similar values from other studies and Parties. There is much variation in the N_2O emission factors available.

42. The following sections briefly discuss each set of N_2O emission factor values presented in table 1. The most recent results presented in this table are from the testing done by TNO Automotive (Riemersma et al., (2003b)). Aggregate results from the recent testing by Meffert et al (2000) and Odaka et al. (2002) on the effect of colder ambient temperatures, and by Huai et al. (2003) on emissions from alternatively fuelled vehicles, were not available in the form necessary for the table and are therefore not presented. Disaggregated emission factors by model year or control technology type were also not available from Behrentz et al. (2004).

A. COPERT

43. The N_2O emission factor values in COPERT III were estimated on the basis of literature review for all vehicle categories (for example, de Reydellet, (1990); OECD, (1991); Perby, (1990); Potter, (1990); Prigent and De Soete, (1989); Zajontz et al., (1991)). EEA (2000) states that the N_2O data in the model "are still quite unreliable and need further confirmation by measurements." The studies used to develop N_2O emission factors are relatively old (nothing later than (1991)). However, it is anticipated that a new version of COPERT with revised N_2O emission factors will be available by the end of 2004.

B. United States inventory and National Vehicle and Fuel Emissions Laboratory testing

44. The EPA, at its National Vehicle and Fuel Emissions Laboratory (NVFEL), measured N_2O emission rates from tier 1 and low emission vehicle (LEV) gasoline-fuelled passenger cars and light-duty trucks equipped with three-way catalysts (EPA, (1998)).

45. According to EPA's regulatory classification for light-duty vehicles, tier 0 is equivalent to "early three-way catalysts," and tier 1 is equivalent to "three-way catalysts" and "advanced three-way catalysts." LEV is representative of tighter restrictions beyond tier 1. Each of these categories actually represents emission regulations (same as Euro 1, etc.) that generally correspond to combinations of emissions control technology design, including post-combustion equipment.

Table 1: N_2O emission factor summary (mg/km)

COPERT III Bulk (hot+cold)				*U.S. Inventory**			*Lipman & Delucchi, 2002*		*1996 IPCC Guidelines European Vehicles*	
Passenger Cars	Urban	Rural	Hwy	**Passenger Cars**	2003	2004	**Passenger Cars**		**Passenger Cars**	
Conventional	5	5	5	Uncontrolled & NonCatalyst	10.3		Uncontrolled	0-12	Uncontrolled & NonCatalyst	5
				Oxidation	32.2				Oxidation	5
Euro I onwards	53	16	35	Tier 0	50.7		Tier 0	12-62		
				Tier 1	28.8		Tier 1	31	TWC	5
				LEVs	17.6					
Diesel	27	27	27	Diesel	10.0		Diesel	6-37	Diesel	10
LPG	15	15	15	LPG	9.4	0.5	LPG	Same as gasoline		
				Methanol	3.9		Methanol (Advanced)	Same as gasoline		
				Ethanol	4.7		Ethanol	Same as gasoline		
				CNG	7.0		CNG (Advanced)	75% of gasoline		
2 - stroke	5	5	5						2 - stroke	5
Light Duty Vehicles				**Light Duty Trucks**			**Light Duty Trucks**		**Light Duty Vehicles**	
Conventional	6	6	6	Uncontrolled	12.9		Uncontrolled	0-12	Uncontrolled	6
				NonCatalyst	12.9					
				Oxidation	40.3					
Euro I onwards	53	16	35	Tier 0	63.5		Tier 0		Moderate Control	6
				Tier 1	36.1		Tier 1	68		
Diesel	17	17	17	Diesel	20.0		Diesel	6-37	Diesel	20
				LEVs	22.0					
Heavy Duty Vehicles				**Heavy Duty Vehicles**			**Heavy Duty Vehicles**		**Heavy Duty Vehicles**	
Gasoline > 3.5 t	6	6	6	Uncontrolled	22.0	29.8	Uncontrolled	31-62	Uncontrolled	6
				NonCatalyst	22.0					
				Oxidation	68.9	93.2				
				Tier 0	109	147				
				Tier 1	86.6					
Diesel	30	30	30	Diesel	30.0		Diesel	12-56	Diesel	30
Buses & Coaches (Diesel)	30	30	30							
							Methanol (Advanced)	Same as gasoline		
							CNG (Advanced)	75% of gasoline		
				LEVs	70.4					
Motorcycles				**Motorcycles**					**Motorcycles**	
Uncontrolled >50 cm³ 2 strk	2	2	2	Uncontrolled & NonCatalyst	4.4	4.5			Uncontrolled < 50 cm³	1
Uncontrolled >50 cm³ 4 strk	2	2	2						Uncontrolled >50 cm³ 2 strk	2
Uncontrolled < 50 cm³	1	1	1						Uncontrolled >50 cm³ 4 strk	2

Note: Gasoline fuelled unless indicated otherwise.

* 2004 emission factor values for the 2004 United States inventory were the same as the 2003 inventory unless indicated.

Table 1: N_2O emission factor summary (mg/km)

1996 IPCC Guidelines U.S. Vehicles		*Koike & Odaka, 1996*		*TNO Automotive*					*Dutch Inventory, 2003 (based on older TNO Automitive results)*			
Passenger Cars		**Passenger Cars**		**Passenger Cars**	Urban hot	Urban cold	Rural	Motorway	**Passenger Cars**	Urban	Rural	Motorway
Uncontrolled & NonCatalyst	20	Uncontrolled	8	NonCatalyst (ECE 15-04)	0	0	0	0	Uncontrolled & NonCatalyst	5	5	5
Oxidation	75	Oxidation (New-Aged)	27-58									
Early TWC (Tier 0)	170			Euro I	21	38	13	8	Euro I	40	20	20
TWC (Tier 1)	170	TWC (Aged / Average)	144 / 48	Euro II	13	24	4	2	Euro II	25	5	5
LEVs	40			Euro III	5	9	2	1	Euro III	10	5	5
Diesel (Uncontrolled)	7			Diesel (Unctrl ECE 15-04)	0	0	0	0	Diesel (Conventional)	5	5	5
Diesel (Moderate)	10			Diesel (Euro I)	2	0	4	4	Diesel (Euro I)	5	5	5
Diesel (Advanced)	14			Diesel (Euro II)	4	3	6	6	Diesel (Euro II)	10	10	10
				Diesel (Euro III)	9	15	4	4	Diesel (Euro III)	10	10	10
				LPG	--- Same as gasoline ---				LPG	--- Same as gasoline ---		
		CNG (with TWC)	16									
Light Duty Trucks									**Delivery Vans**			
Uncontrolled	24								Conventional	Same as pasgr cars		
NonCatalyst	23									Same as pasgr cars		
Oxidation	97											
Early TWC (Tier 0)	227								Euro I onwards	Same as pasgr cars		
TWC (Tier 1)	236											
Diesel	24-63								Diesel	20	20	20
									LPG	Same as pasgr cars		
LEVs	58											
Heavy Duty Vehicles		**Heavy Duty Vehicles**		**Heavy Duty Vehicles**					**Lorries**			
Uncontrolled	54											
NonCatalyst	591											
Oxidation												
TWC	606											
Diesel (Unctrl & Moderate)	25	Diesel	11	Diesel	--- See detailed table in paper ---				Diesel	30	30	30
Diesel (Advanced)	31											
									Buses & Tractors (Diesel)	30	30	30
Motorcycles									**Motorcycles**			
Uncontrolled & NonCatalyst	2								Uncontrolled	2	2	2
									Mopeds	1	1	1

Note: Gasoline fuelled unless indicated otherwise

46. Based on the tests at NVFEL, the EPA concluded that for vehicles with three-way catalysts:

(a) N_2O emissions are likely to increase with higher-sulphur-content gasoline fuels

(b) Emissions are greater with air-conditioning operating than without it operating

(c) Light-duty trucks exhibited greater emission factors than did passenger cars

(d) N_2O emissions were unrelated to vehicle mileage (i.e., catalyst age)

(e) Vehicles with catalyst and engine designs meeting the more recent tier 1 and LEV standards exhibited reduced emission rates of both NO_X and N_2O compared with tier 0 vehicles.

47. The tests at NVFEL and a review of the literature were used to develop the N_2O emission factors for the United States GHG road transport emission inventory. The following references were used by EPA for gasoline-fuelled highway passenger cars:

(a) LEVs. Tests performed at NVFEL (EPA (1998))[6]

(b) EPA tier 1. Tests performed at NVFEL (EPA (1998))

(c) EPA tier 0. Barton and Simpson (1994), Smith and Carey (1982), and one car tested at NVFEL (EPA (1998))

(d) Oxidation catalyst. Smith and Carey (1982), Urban and Garbe (1980)

(e) Non-catalyst. Dasch (1992), Prigent and De Soete (1989), and Urban and Garbe (1979).

48. Nitrous oxide emission factors for other types of gasoline-fuelled vehicles (light-duty trucks, heavy-duty vehicles and motorcycles) were estimated by adjusting the factors for gasoline passenger cars by their relative fuel economies. EPA (2004) reports that data from the literature and tests performed at NVFEL support the conclusion that light-duty trucks and other vehicles have higher emission rates than passenger cars, and acknowledges that the use of fuel-consumption ratios to determine N_2O emission factors is "considered an estimate, with a moderate level of uncertainty."

49. Nitrous oxide emission factors for tier 1 and LEV heavy-duty gasoline vehicles were estimated from the ratio of NO_X emissions to N_2O emissions for EPA tier 0 heavy-duty gasoline trucks. An NO_X to N_2O ratio of 60 was applied to the NO_X emissions from EPA tier 1 and LEV heavy-duty gasoline vehicles.[7] The N_2O emission factors employed by EPA for gasoline road vehicles are less than the United States default values in the 1996 IPCC Guidelines, but are greater than the European default values.

50. Nitrous oxide emission factors for all classes of diesel highway vehicles were taken from the European default values found in the 1996 IPCC Guidelines.

51. EPA's N_2O emission factors for light-duty alternative fuel vehicle are based on data in Argonne National Laboratory's GREET 1.5 Transportation Fuel Cycle Model (Wang, (1999)) and the conclusions reported in Lipman and Delucchi (2002) for some light and heavy-duty alternative fuel vehicles. EPA

[6] LEVs were assumed to be operated using low-sulphur fuel (i.e., Indolene at 24 ppm sulphur). All other NVFEL tests were performed using a standard commercial fuel (CAAB at 285 ppm sulphur). Emission tests by NVFEL consistently exhibited higher N_2O emission rates from higher sulphur fuels on EPA tier 1 and LEV vehicles.

[7] From the United States national inventory report, it is not clear whether this ratio was applied to exhaust gas concentrations or mass emissions.

assumes most alternative fuel vehicles use catalysts similar to EPA tier 1 gasoline cars and applies an NO_X to N_2O ratio of 5.75 to tier 1 gasoline car emission factor values. Lipman and Delucchi (2002) found NO_X to N_2O ratios for light-duty alternative fuel vehicles with three-way catalyst systems to vary from 3 to 5.5 for older technology.

52. Overall, Lipman and Delucchi (2002) analysed existing N_2O studies with testing done prior to 1999. They found that N_2O emission factors for light-duty vehicles equipped with three-way catalysts meeting United States tier 1 standard were, in general, lower than default values in the 1996 IPCC Guidelines, but perhaps somewhat higher than those in EPA (1998) and used in the United States GHG inventory.

53. Meffert et al. (2000) also reported, based on testing results in EPA (1998) and Becker (1999) and for the same fuel type, that there is no support for EPA's finding that LEVs emit less N_2O than tier 1 vehicles or that tier 1 vehicles emit less than tier 0 vehicles.

C. Intergovernmental Panel on Climate Change Guidelines for United States and European vehicles

54. The N_2O emission factors for road vehicles in the 1996 IPCC Guidelines are based on a small number of tests from the late 1980s and early 1990s (Riemersma et al., (2003); EPA, (1998)). After tracking the origins of the N_2O factors for United States light-duty gasoline vehicles in the 1996 IPCC Guidelines, EPA (1998) found that:

(a) All the emission factors originate from testing done on five cars using European test cycles. Fuel sulphur content for these tests was unspecified;

(b) The new and aged three-way catalysts emission factors were based 90 per cent on a single study using a single car with eight non-production catalysts, new and bench-aged, with the catalysts located 1.4 m from the engine. The other 10 per cent of the data for the three-way catalyst emission factors came from two studies and three more cars, all tested on European driving cycles;

(c) The non-catalyst emission factors were derived from four cars;

(d) The emission factor for oxidation catalyst vehicles does not appear to be based on testing, but is instead the same emission factor used for new three-way catalysts;

(e) The references used by the IPCC were Ballantyne et al. (1994), De Soete (1989), Jacques (1992), Lindskog (1988), and Prigent and De Soete. (1989).

55. The IPCC good practice guidance (IPCC, 2000) did not provide a comprehensive update of N_2O emission factors, although data were available (e.g., the EPA (1998) study summarized data for United States vehicles). The emission factors in the IPCC good practice guidance are not given in terms of travel activity (e.g., g N_2O/km), which is commonly used for the actual compilation of emissions, but they are converted and presented in terms of energy units (kg N_2O/TJ). This conversion was done using an average fuel economy factor, thereby implying that they could be applied directly to fuel consumption data. Both the 1996 IPCC Guidelines and the IPCC good practice guidance do not include any emission factors for alternative fuel vehicles.

D. Koike and Odaka

56. Summary N_2O emission factor values were presented in Koike and Odaka (1996) for a small number of vehicle categories. However, it is assumed that the authors would be able to provide revised summary factors based on their subsequent work.

E. TNO Automotive

57. TNO Automotive has undertaken an extensive research and testing programme on N_2O emissions from road transport. Feijen-Jeurissen et al. (2001) includes a detailed review of the literature on the process of N_2O formation in three-way catalysts and a variety of other types of catalysts. Based on their analysis, they expected that European N_2O emission factors for vehicles with three-way catalysts should decrease from 1990 to 2000 due to improved control over air-fuel ratios, lower sulphur content of fuel, and improved activity and stability of catalysts. They also concluded that N_2O is not formed during combustion in diesel-fuelled vehicles.

58. Riemersma et al. (2003b) summarizes this work as including 45 passenger cars using gasoline, diesel, and liquefied petroleum gas (LPG) fuels and a European driving cycle. TNO also tested one hybrid vehicle and 33 heavy-duty diesel trucks and buses. Another 10 gasoline passenger cars were tested using both the European driving cycle and the Common Artemis Driving Cycle; the latter is meant to simulate more realistic driving practices. Measurements were made using hot starts and cold starts at 20 and 9°C. New catalysts were tested and then retested after ageing 3,000 km. High and low sulphur fuels were also tested.

59. TNO Automotive's results for diesel heavy-duty trucks is presented in table 2; however, they indicate that due to considerable uncertainty in their methods these values were not to be taken as definitive estimates.[8] They concluded that N_2O emission factors for Euro 1 to 3 heavy-duty diesel trucks were much less than the IPCC default factor (30 mg/km) (Riemersma et al., (2003a)).

60. TNO Automotive concluded that future regulatory restrictions and emission control technology improvements focused on NO_X emissions are likely to also reduce N_2O emission rates for light-duty vehicles. However, as an increasing number of diesel vehicles include catalytic emission control equipment, emissions of N_2O from diesel vehicles will greatly increase.

61. Detailed discussions on the work at TNO Automotive can be found in Feijen-Jeurissen et al. (2001), Gense and Vermeulen (2002), and Riemersma et al. (2003a).

F. Dutch inventory

62. N_2O emission factors used to estimate road vehicle emissions in the Netherlands are based on older measurements by TNO Automotive (Feijen-Jeurissen et al., (2001)). These emission factors do not incorporate the most recent data discussed in Riemersma et al. (2003b). The Dutch inventory reports that testing at TNO indicates that heavy-duty diesel engines emit very little N_2O, and considerably less than the IPCC default values for European vehicles, which are also used by the United States. The values presented in table 1 were taken from a methodological companion document to the Dutch inventory (Spakman et al., (2003)). TNO has indicated that future inventory submissions by the Netherlands will probably incorporate the more recent emission factor results in Riemersma et al. (2003b).

G. University of California Los Angeles/California Air Resources Board testing

63. Behrentz et al. (2004) used dynamometer testing in conjunction with high resolution Fourier Transform Infrared spectroscopy to measure N_2O emissions from 37 light-duty vehicles at the California Air Resources Board. The vehicles studied, including passenger cars, sport utility vehicles, and light-duty trucks, were a subsample of a fleet that represented California's in-use vehicle fleet. Two driving cycles were used: Standard United States Federal Test Procedure (FTP-75) urban dynamometer driving schedule (UDDS) driving cycle; and Unified Cycle (UC).

[8] TNO reported an asymmetric uncertainty range of about plus 25 per cent and minus 50 per cent (Riemersma et al., (2003a)).

Table 2: TNO Automotive test results for "real-life" N_2O emission factors for heavy-duty diesel vehicles

Euro 1			**N_2O emission factors (mg/km)**		
Vehicle type	**GVW (tonne)**	**Load condition**	**Urban**	**Rural**	**Highway**
Light truck	6.8	fully loaded	<6	<5	<3
Medium truck	13.6	fully loaded	<11	<9	<7
Heavy truck	37.1	half loaded	<17	<14	<10
Heavy truck	37.1	fully loaded	<19	<16	<11
Truck with trailer/semitrailer	40.0	half loaded	<18	<15	<11
Truck with trailer/semitrailer	40.0	fully loaded	<20	<17	<11
Urban bus	15.2	fully loaded	<12	<10	<7

Euro 2			**N_2O emission factors (mg/km)**		
Vehicle type	**GVW (tonne)**	**Load condition**	**Urban**	**Rural**	**Highway**
Light truck	6.8	fully loaded	<5	<5	<3
Medium truck	13.6	fully loaded	<11	<9	<6
Heavy truck	37.1	half loaded	<17	<14	<10
Heavy truck	37.1	fully loaded	<18	<16	<10
Truck with trailer/semitrailer	40.0	half loaded	<18	<15	<10
Truck with trailer/semitrailer	40.0	fully loaded	<20	<17	<11
Urban bus	15.2	fully loaded	<12	<10	<7

Euro 3			**N_2O emission factors (mg/km)**		
Vehicle type	**GVW (tonne)**	**Load condition**	**Urban**	**Rural**	**Highway**
Light truck	6.8	fully loaded	<3	<3	<2
Medium truck	13.6	fully loaded	<5	<5	<4
Heavy truck	37.1	half loaded	<8	<8	<6
Heavy truck	37.1	fully loaded	<10	<9	<7
Truck with trailer/semitrailer	40.0	half loaded	<9	<9	<7
Truck with trailer/semitrailer	40.0	fully loaded	<11	<10	<7
Urban bus	15.2	fully loaded	<6	<6	<5

SCRdeNO$_X$			**N_2O emission factors (mg/km)**		
Vehicle type	**GVW (tonne)**	**Load condition**	**Urban**	**Rural**	**Highway**
Light truck	6.8	fully loaded	18	24	21
Medium truck	13.6	fully loaded	36	49	41
Heavy truck	37.1	half loaded	55	75	63
Heavy truck	37.1	fully loaded	89	93	69
Truck with trailer/semitrailer	40.0	half loaded	59	81	68
Truck with trailer/semitrailer	40.0	fully loaded	96	100	74
Urban bus	15.2	fully loaded	40	55	46

CRT			**N_2O emission factors (mg/km)**		
Vehicle type	**GVW (tonne)**	**Load condition**	**Urban**	**Rural**	**Highway**
Light truck	6.8	fully loaded	19	14	9
Medium truck	13.6	fully loaded	38	29	17
Heavy truck	37.1	half loaded	58	44	26
Heavy truck	37.1	fully loaded	59	41	24
Truck with trailer/semitrailer	40.0	half loaded	63	47	28
Truck with trailer/semitrailer	40.0	fully loaded	64	44	25
Urban bus	15.2	fully loaded	43	32	19

Source: Riemersma et al. (2003a).

64. Behrentz et al. (2004) found that catalyst type, driving cycle, and vehicle type were the most important factors determining the N_2O emission rates from gasoline powered light-duty vehicles. However, their results indicated that the presence of a pre-catalyst, the type of transmission (automatic or manual), and the engine configuration (L or V) did not greatly affect N_2O emissions. The biggest differences in N_2O emissions were observed between the different phases (i.e., bags) within the driving cycles, indicating that operating conditions such as driving patterns and catalyst temperature play a major role in the formation of N_2O. They also suggested that although overall N_2O/NO_X emission ratios could be used to estimate N_2O emission factors, more sophisticated analyses should be applied to control for confounding variables.

65. Among the vehicles tested, the average N_2O emission factor from pilot study was 20 ±4 mg/km, although factors ranged from 2 mg/km for a 2001 model (LEV) passenger car to 100 mg/km for a 1991 model (tier 0) light-duty truck. Average N_2O emission factors from their study are presented in table 3. They also reported that they expected light-duty vehicles to exhibit decreasing N_2O emissions with increasingly stringent NO_X control technologies.

Table 3: Average N_2O emission factors

		Average N_2O (mg/km)			
Selection criteria	Number of vehicles	Phase 1	Phase 2	Phase 3	Entire cycle [a]
UDDS/ethanol/LDT/TWC/double bed/no pre-catalyst	1	6	4	7	5
UDDS/summer/PC/TWC/single bed/no pre-catalyst	16	32	14	35	23
UDDS/winter/PC/TWC/single bed/no pre-catalyst	6	27	7	27	16
UC/summer/PC/TWC/single bed/no pre-catalyst	6	57	10	63	16
UC/winter/PC/TWC/single bed/no pre-catalyst	6	56	12	51	17
UC/summer/PC/oxidizing	2	25	20	60	23
UC/winter/PC/TWC/single bed/pre-catalyst	2	53	13	64	19
UDDS/winter/LDT/TWC/single bed/no pre-catalyst	4	54	22	61	39

Source: Behrentz et al. (2004).
[a] Weighted average computed according to 40 CFR §83.144.90.

66. The results presented in Behrentz et al. (2004) are from a pilot study that is part of a more extensive study of vehicle N_2O emissions. The investigators have recently finished the full testing programme (350 dynamometer tests, 140 vehicles, and 5 driving cycles) and are currently analysing data.

VI. Conclusions

67. There remains a need for additional research and testing on N_2O emissions from road vehicles equipped with post-combustion catalyst emission control equipment. The existing versions of both MOBILE and COPERT require updating of CO_2, CH_4 and N_2O emission factors. The detailed activity data addressed in these emission models should facilitate the application of better N_2O emission factors. Further, these models should integrate default unoxidized fraction assumptions for estimating CO_2 emissions, which should be consistent with IPCC guidance. These default unoxidized fraction values should be based on actual measurement data, where available.

68. The results of the most recent testing (e.g., Riemersma et al. (2003); Odaka et al. (2002)) and the expected results from ongoing testing (e.g., UCLA/CARB) could be evaluated in developing new default N_2O emission factors for the 2006 IPCC Guidelines. In addition, the IPCC could consider the following issues:

(a) Improvement of the methodologies for estimating N_2O emissions from road transport vehicles. In particular:

(i) Use activity data and emission factors based on vehicle kilometres travelled (VKT) and taking into account the vehicle types and emission control technology type/standard.[9] Testing by TNO Automotive indicates that for gasoline light-duty vehicles with three-way catalysts, improvements in NO_X emission controls over time (Euro 1, 2, 3, etc.) has and will continue to lead to lower average N_2O emission rates from vehicles;

(ii) Provide advice to Parties that may need to use aggregate fuel data instead of VKT data to estimate N_2O and CH_4 emissions on how to transparently document their assumptions for average fleet fuel economy and convert their fuel consumption data into vehicle travel activity data (VKT) in order to apply emission factors;

(iii) Focus on factors that affect the temperature of the catalyst bed (e.g., ambient temperatures, frequency of cold starts, catalyst placement);

(iv) Provide advice to Parties on how to incorporate a factor (or factors) to estimate the penetration of various types of catalytic control technology equipment in the country's vehicle fleet.

(b) Development of N_2O emission factors for all vehicle classes and environments. In particular:

(i) At a minimum, separate emission factors for the portion of the vehicle fleet that has catalytic emission control equipment and the portion that does not could be included in the 2006 IPCC Guidelines;

(ii) Focus on the time that the vehicle's catalyst remains in the relevant temperature window that has been found to generate large N_2O emissions (250 to 500ºC, with a peak around 280 to 350ºC);

(iii) Take into account the operation of vehicles in cold climates or cold seasons, for which higher emission factors are expected;

(iv) Take into account the frequency in the use of air conditioning. N_2O emission factors should be higher for regions where air conditioning is frequently used, or a correction factor should be included in the IPCC methodology. EPA testing found emissions in gasoline passenger cars to be on average 50 per cent higher with air conditioning operating;

(v) Take into account information on catalyst ageing. Catalyst ageing does have an effect on N_2O emissions, but ageing appears to happen relatively early to new catalysts. Whether it leads to increased emission rates depends upon the typical driving cycle and catalyst type. Activity data should not be separated into aged and unaged categories. Emission factors should be based on aged catalysts and there does not appear to be a sound basis using existing data to adjust N_2O emission factors for catalyst age;

(vi) Develop N_2O emission factors for light-duty and heavy-duty trucks instead of simply basing them on their relative fuel-economy to passenger cars or NO_X ratios;

[9] Vehicle model year may be used as a proxy for control technology type/standard in cases where specific regulatory deadlines have been implemented.

(vii) Given the post-combustion control technology used, engine size of the vehicle, climate, and driving cycle, it appears that N_2O emission factors for Europe are not much different from those in the United States.

69. The IPCC may further consider the following issues and decide whether additional guidance should be incorporated in the 2006 IPCC Guidelines:

(a) National emission factors may need to account for large differences in the sulphur content of fuels used between countries in vehicles equipped with three-way catalysts, as well as for any regulatory changes that may affect the sulphur content of fuels. Parties with high-sulphur fuel may exhibit much higher N_2O emissions;

(b) Diesel vehicles do not appear to produce much N_2O emissions, in the absence of catalytic emission control technology. As an increasing number of diesel vehicles install catalytic emission control equipment, the importance of N_2O emission factors for diesel engines will increase considerably. IPCC default N_2O emission factors may need to be lowered for light-duty and heavy-duty diesel vehicles without catalysts but should anticipate the penetration of new diesel vehicles with catalysts and higher emissions in the future;

(c) Minimal data are available on N_2O emissions from alternatively fuelled vehicles (e.g., ethanol, methanol, LPG, compressed natural gas (CNG)). The evidence available appears to indicate that emissions from ethanol, methanol and LPG vehicles with and without three-way catalysts are similar to those fuelled with gasoline. There is some conflicting information on the similarity between CNG and gasoline-fuelled vehicles, with CNG vehicles appearing to have lower emissions in some studies;

(d) Use of travel activity-based models (e.g., MOBILE, MOVES, and COPERT) to estimate fuel consumption, and therefore CO_2 emissions for national GHG inventories in order to provide higher resolution data for reporting. However, these CO_2 estimates should be calibrated with CO_2 emission estimates from more aggregate fuel consumption data (e.g. sales data) and carbon content values. As a rule, the total calculated fuel consumption based on travel activity should also be equal to the consumption statistics found in the Parties' energy balance.

70. Finally, Parties may wish to consider the following regarding the estimation of emissions from road transport:

(a) Using the same data and modelling approach for estimating CH_4 and N_2O emissions as they use to estimate air quality pollutant (e.g., hydrocarbons, CO, and NO_X) emissions to ensure consistency in inventory estimates from road transport;

(b) Reporting vehicle travel activity data (e.g., vehicle kilometres travelled) in the CRF submissions, instead of fuel consumption data, for better comparison with actual emission factors.

Annex I

Factors that potentially affect nitrous oxide emissions from road vehicles

A. Ambient temperature

1. The operating temperature of catalytic converters is affected significantly by its location in the vehicle. The placement and insulation of three-way catalysts has generally been optimized to minimize the time required for the catalyst bed to reach operating temperature. However, the design and placement of the catalyst must also prevent the catalyst from deteriorating as a result of exposure to excessively high temperatures (Feijen-Jeurissen et al., (2001)).

2. Lipman and Delucchi (2002), based on an examination of existing studies, did not find an effect of colder ambient temperatures on the catalyst bed temperature, although they did state that ambient temperature could affect engine load and combustion conditions, which might affect N_2O emissions.

3. Odaka et al. (2002) tested vehicles with three-way catalysts at ambient temperatures of –4 and –5°C and continuously measured N_2O emissions and temperatures at various points along the exhaust system and catalyst using a 2001 model Toyota Corolla bench test configuration. The catalyst in each test was placed immediately adjacent to the exhaust manifold. They found that colder ambient temperatures lead to an increase in the amount of time it took the catalyst to fully heat up from a cold start. They also found that ambient temperature affected the time it took the catalyst to warm up more than did the driving pattern (i.e., type of driving cycle).

4. The ultimate conclusion in Odaka et al. (2002) was that "N_2O emissions may drastically increase in colder cities and winter city traffic conditions." Specifically, they found that N_2O emissions at a cold ambient temperature (–4 to –5°C) were "at least three times" the amount emitted at warmer ambient temperatures (25°C).

5. In vehicles where the catalyst is installed farther from the engine exhaust manifold and more of the exhaust system leading to the catalyst is exposed to convective and radiative heat transfer, the effect of lower ambient temperatures may be greater. If colder ambient temperatures prevent the catalyst bed from heating up fully, and the catalyst is then regularly operated below 500°C, much greater amounts of N_2O could be generated.

6. TNO Automotive also found that N_2O emission rates were greater after a cold start than after hot start, and "colder" cold starts (9ºC) produced larger emission factors than room temperature starts (20ºC). Specifically, they found that the average emission factor for 10 gasoline passenger cars with three-way catalysts after hot, 20°C, and 9°C starts were 20, 35, and 41 mg N_2O/km, respectively (Gense and Vermeulen, (2002)).[1] They concluded that the lower ambient temperatures caused the catalyst to operate for a longer time within the temperature window favourable to N_2O formation (Riemersma et al., (2003b); Gense and Vermeulen, (2002)). Similar effects were found by Laurikko et al. (1994).

7. Behrentz et al. (2004) reported that the importance of cold starts differed when vehicles were separated into high, intermediate, and low emitters. They found that for high emitting vehicles, the emission rate was highest when the catalyst was at the fully warmed-up operating temperature. They concluded that the catalysts on these high emitting vehicles were not functioning optimally. Emissions

[1] The results in Gense and Vermeulen (2002) were based on tests for the following vehicles: Honda Accord, Opel Astra, Opel Astra Station, Opel Corsa, Peugeot 306, Renault Clio, Saab 9-3, Toyota Corolla Wagon, Toyota Corolla, and VW Lupo.

after a hot start actually increased the amount of time that the vehicle operated under temperatures that resulted in high emission rates.

B. Catalyst ageing

8. EPA (1998) found from its testing campaign that N_2O emissions from vehicles with three-way catalysts were unrelated to vehicle mileage. This finding can be interpreted as implying that there is no effect on N_2O emissions from catalyst age. However, the data set examined by EPA did not include any new (i.e., unaged) catalysts. The lowest mileage vehicle tested still had an odometer reading of more than 16,000 miles and mileages on most vehicles were more than 20,000 miles.

9. Behrentz et al. (2004) also did not find significant correlations between vehicle mileage and N_2O emissions. However, they noted that their results could be affected by the relatively small number of vehicles used to analyse the effect of vehicle mileage.

10. Although EPA (1998) and Behrentz et al. (2004) did not show a clear catalyst ageing effect (i.e., mileage effect), other studies have (e.g., Koike et al., (1999); Meffert et al., (2000); Odaka et al., (1998); Riemersma et al., (2003b)). Specifically, Odaka et al. (1998) and Koike et al. (1999) found that ageing generally increases N_2O emissions, but that ageing can decrease emissions depending upon the driving cycle assumed, the type of catalyst and the related shifts in the temperatures favourable to N_2O formation as the catalyst ages. The shift in this temperature window varies with the types and quantities of metals used in the catalyst (e.g., Pt and Rhodium), but some of the differences between metals seem to disappear as the catalyst deteriorates through ageing (Odaka et al., (1998)). TNO Automotive found that passenger cars equipped with aged three-way catalysts emit relatively more than vehicles with fresh catalysts due to deactivation over time; however, they were not able to quantify the effect (Gense and Vermeulen, (2002)).

11. Meffert et al. (2000) also found that ageing (i.e., 10,000 versus 60,000 miles) caused changes in N_2O emissions at various stages of the United States FTP driving cycle. They reported that these changes appeared to be caused by a shifting of the temperature distribution favourable to N_2O formation in the catalyst.[2]

12. It appears that most of the effect of ageing on a catalyst's tendency to produce N_2O occurs somewhat early in its lifetime. Therefore, most of a vehicle's operating life will be spent with a catalyst in a relatively aged condition with respect to N_2O emissions. Although ageing appears to increase N_2O emission rates more often than decrease them, the effect of ageing depends strongly on the relationship between shifts in the temperature window and the vehicle's operating temperature profile.

C. Fuel formulation

13. The reaction mechanisms in three-way catalysts are sensitive to the concentrations of sulphur, oxygen, water vapor, CO, and NO. Nitrous oxide emission rates in vehicles with three-way catalysts have been shown to increase with the sulphur content in fuels. Based on their own testing, EPA (1998) concluded that higher-sulphur fuels were likely to increase N_2O emissions.

14. TNO Automotive, based on their measurement data, were unable to confirm that the use of higher-sulphur gasoline in vehicles with three-way catalysts leads to higher N_2O emissions; however, they stated that such an effect is still anticipated (Riemersma et al., (2003b)).

15. Meffert et al. (2000) reported that "factors that lower NO_X conversion efficiency tend to increase N_2O emissions". Specifically, they found that the use of the fuel additive methyl cyclopentadienyl

[2] Meffert et al. (2000) tested 1997 Crown Victorias equipped with tier 1 compliant three-way catalysts. They found that N_2O emissions averaged 78 mg/km at 60,000 miles.

manganese tricarbonyl (MMT) could reduce vehicle N_2O emissions over time by reducing the deposition of phosphorus on Pt catalysts, and thereby minimizing the effect of ageing.

D. Air/fuel ratio

16. In order for three-way catalysts to efficiently convert CO, hydrocarbons, and NO, proper stoichiometric ratios of these pollutants and oxygen in the exhaust gas must be maintained. It is the function of the lambda (λ) control sensor, which is located just before the catalyst, to maintain the catalyst at optimal stoichiometric conditions. The result is that the catalyst oscillates between slightly oxidizing and reducing reaction conditions. TNO Automotive has reported that N_2O emission rates increase when an engine operates at a lean air/fuel ratio, thereby producing more oxidizing reaction conditions for the exhaust gas ($\lambda<1$). This effect is due to the removal of CO, which is used as a reduction species for NO (Feijen-Jeurissen et al., (2001)). As emission control regulations on NO_X have tightened for newer vehicles, λ-control has also improved. In addition to helping to reduce regulated pollutants, Feijen-Jeurissen et al. (2001) found that these improvements also led to lower N_2O emission rates.

17. Meffert et al. (2000) also reported that under real driving conditions with three-way catalysts "[t]he presence of some N_2O in exhaust, either as a result of innate catalyst behaviour or oscillating air-fuel ratio…should be expected even with a warmed-up low mileage catalyst."

18. Vehicles with three-way catalysts have been regularly observed to emit N_2O even at fully heated operating temperature (~500°C), although at rates less than at lower temperatures. Emission rates at these higher operating temperatures have been found to vary greatly across vehicles (Gense and Vermeulen, (2002)). One of the reasons for the continued emission of N_2O at these higher temperatures may be that catalysts in some vehicles operate at more oxidizing reaction conditions.

E. NO_X and N_2O correlation

19. Because N_2O is formed from NO, it is expected that N_2O emissions should be somewhat correlated with NO_X emissions and NO throughput from the engine. Meffert et al. (2000), though, found that N_2O/NO_X emission ratios were highly variable in the vehicles they tested. TNO Automotive found that tailpipe N_2O emissions did not straightforwardly correlate with engine-out NO_X emissions rates in gasoline and diesel light-duty and heavy-duty vehicles with various catalyst configurations and should not be based on NO_X emissions (Riemersma et al., (2003b)). However, they did observe a relationship between NO_X conversion efficiency of three-way catalysts and tailpipe N_2O emissions (Gense and Vermeulen, (2002)).

20. After dynamometer testing 37 in-use vehicles, Behrentz el al. (2004) found an overall N_2O/NO_X emissions ratio of 0.095 ± 0.035. The vehicles tested exhibited a relatively small variability in NO_X emission factors (300 ± 70 mg/km). However, they report that whereas N_2O formation is highly dependent on catalyst temperature, NO_X production is highly dependent on engine temperature, and these two conditions are not necessarily correlated throughout the entire course of a testing cycle.

21. Differences in the temperature dependent mechanics of NO_X conversion and N_2O generation and conversion in three-way catalysts suggest that N_2O emissions and tailpipe NO_X emissions are not easily correlated and depend on a number of other key variables. Their use for determining N_2O emission factors is uncertain and the subject of ongoing scientific debate.

F. Driving cycle

22. Assumptions regarding the typical driving cycle can dramatically affect the emissions rate for N_2O because over a complete cycle – assuming that the catalyst warms up beyond 500°C – the catalyst

tends to operate within the N_2O temperature window for only a small fraction of the entire cycle time. Changes in the assumed driving cycle that extend or reduce the time that the vehicle catalyst is operated at these lower temperatures (e.g., frequency of cold starts) will have a roughly proportional effect on the overall N_2O emissions rate. The sensitivity of the catalyst bed's temperature to driving cycle – and possibly the placement of the catalyst along the exhaust system – may explain some of the variation found in N_2O emission rates across vehicles and tests.

23. TNO Automotive found that for gasoline cars, N_2O emission rates using a more "realistic" driving cycle were lower than those using the standardized European Driving Cycle. The more "realistic" driving cycle produced higher engine loads (e.g., faster accelerations), and therefore led to a more rapid heating up of the catalyst and a reduced amount of time that the catalyst is operated or falls into the temperature window favourable for N_2O formation (Riemersma et al., (2003b)).

24. Behrentz et al. (2004) determined that the driving cycle selected has a major effect on N_2O emission rates. A more aggressive cycle (i.e., the unified cycle) yielded the highest emissions compared to the United States standard driving cycle. EPA (1998) also found that N_2O emission rates were greater when air-conditioning was in operation than without it operating. It is assumed this effect is due to increased engine load and greater exhaust output.

G. Vehicle and fuel type

25. Light-duty trucks have, on average, been found to have higher emission rates than passenger cars (EPA, (1998)). It is assumed that the reason for this is primarily because of the increased throughput of exhaust gas in most light-duty trucks.

26. Behrentz et al. (2004) reported that vehicle type played a major role in determining N_2O emission rates. Specifically, light-duty trucks exhibited much higher N_2O emission rates compared to passenger vehicles. They also found that light-duty trucks and passenger cars produced similar amounts of N_2O per unit of NO_X, and concluded that the absolute differences in N_2O emissions between vehicle types were likely to be caused by less stringent NO_X emissions standards for light-duty trucks in the United States.

27. TNO Automotive concluded that N_2O is not formed during combustion in diesel-fuelled vehicles (Feijen-Jeurissen et al., (2001)). Specifically, diesel fuelled light-duty vehicles without catalysts emitted almost no N_2O after both cold and hot starts (Gense and Vermeulen, (2002)). However, they concluded that oxidation catalysts and catalysed soot filters on diesel engines do produce N_2O emissions, although at lower rates than gasoline-fuelled vehicles with three-way catalysts.

28. Both diesel and gasoline fuelled heavy-duty trucks, in most countries, do not operate with three-way catalysts, and therefore tend to have lower N_2O emissions. TNO Automotive reported that diesel heavy-duty vehicles emission rates were much less than vehicles equipped with catalysts and were much less than IPCC default values, which were based on emission factors for light-duty vehicles and an assumed correlation with NO_X emissions (Riemersma et al., (2003b)). They also found that N_2O emission rates after cold starts were not different than after a hot start (Riemersma et al., (2003a)). However, they did find that the use of after treatment systems such as SCR-deNOx and EGR with CRT filter (e.g., Euro IV and V vehicles) for heavy-duty vehicles is likely to increase N_2O emission rates (Riemersma et al., (2003b))

29. Few data are available on N_2O emissions from alternatively fuelled vehicles (e.g., ethanol, methanol, LPG, and CNG). Most of the evidence that is available appears to indicate that emissions from ethanol, methanol and LPG vehicles with and without three-way catalysts are similar to those for vehicles fuelled with gasoline. Based on their analysis of existing studies, Lipman and Delucchi (2002) concluded that N_2O emission factors being developed for CNG-fuelled vehicles with catalysts were, on

average, 75 per cent less than those for similar gasoline fuelled vehicles. Koike and Odaka (1996) also showed lower emission rates for CNG vehicles.

30. However, more recent testing by Huai et al. (2003) did not appear to show lower emission rates from CNG vehicles than gasoline fuelled vehicles (see table below).

Table: N_2O emission factors for alternatively fuelled vehicles

Model year	Make	Model	Fuel	N_2O (g/mi)
1999	Honda	Civic GX	CNG	<MDL
1995	GMC	Sonoma PU	CNG	0.022
1994	Dodge	Caravan Minivan	CNG	0.008
1994	Dodge	Ram 350 Van	CNG	0.077
1994	Dodge	Ram 350 Van 2	CNG	0.016
2000	Ford	F-150 XL	LPG gasoline	0.017
1999	Ford	F250 XLT	LPG gasoline	0.012
1992	Chevrolet	S10 PU	LPG	0.006
1994	Ford	Taurus FFV	M85	0.059
1992	Dodge	Spirit FFV	M85	0.004

Source: Taken from Huai et al. (2003), who also reported an average of 35.5 mg N_2O/km across all alternatively fuelled vehicles tested.

31. TNO Automotive, based on testing of one low mileage vehicle installed with a three-way catalyst and a NO_X storage catalyst, found that a hybrid car emitted low levels of N_2O during the entire test cycle (Feijen-Jeurissen et al., (2001)).

Annex II

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