United States Environmental Protection Agency | Air and Radiation | EPA420-R-98-009
August 1998

Emissions of Nitrous Oxide from Highway Mobile Sources

Comments on the *Draft Inventory of U.S. Greenhouse Gas Emissions and Sinks, 1990-1996* (March 1998)

EPA420-R-98-009
August 1998

Emissions of Nitrous Oxide from Highway Mobile Sources

Comments on the *Draft Inventory of U.S. Greenhouse Gas Emissions and Sinks, 1990-1996* (March 1998)

Harvey Michaels
Assessment and Modeling Division
Office of Mobile Sources
U.S. Environmental Protection Agency

NOTICE

This technical report does not necessarily represent final EPA decisions or positions. It is intended to present technical analysis of issues using data which are currently available. The purpose in the release of such reports is to facilitate the exchange of technical information and to inform the public of technical developments which may form the basis for a final EPA decision, position, or regulatory action.

TABLE OF CONTENTS

EMISSIONS OF NITROUS OXIDE FROM MOBILE SOURCES:

COMMENTS ON THE *DRAFT INVENTORY OF U.S. GREENHOUSE GAS EMISSIONS AND SINKS , 1990-1996* (MARCH 1998)

August 13, 1998

1 INTRODUCTION

The estimate of the contribution of nitrous oxide from mobile sources to total U.S. emissions of greenhouse gases went from one-half percent in the last official inventory, published in 1997 (U.S. EPA) to three percent in the March 10, 1998, draft *Inventory of U.S. Greehouse Gas Emissions and Sinks 1990-1996* (U.S. EPA), which will be referred to in these comments as the *Draft Inventory*. The primary reason for this change is the use of much larger emission factors for gasoline highway vehicles, rather than increases in vehicle miles traveled. OMS believes that these emission factors are considerably larger than they should be. Therefore, these comments will focus primarily on the origin and validity of the emission factors used in the *Draft Inventory* and on the development of better ones.

The emission factors for passenger vehicles from the last official Inventory, from the 3/10/98 *Draft Inventory*, and from OMS's proposed revisions that are developed in this document, are listed in the following table.

Control Technology	N_2O Emission Factors for Passenger Vehicles		
	Last Official Inventory (g/km)	3/10/98 *Draft Inventory* (g/km)	OMS Revision (g/km)
LEV		0.040	0.018
Advanced 3 Way (Tier 1)	0.019	0.170	0.029
Early 3-way (Tier 0)	0.046	0.170	0.051
Oxidation Catalyst	0.027	0.075	0.032
Non-Catalyst	0.005	0.020	0.010
Uncontrolled	0.005	0.020	0.010

The *Draft Inventory* adopted the emission factors for U.S. vehicles from the *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 1997), which are referred to in these comments as the *IPCC Guidelines*. They list emission factors for European cars that are between four and thirty-four times lower than for similar U.S. vehicles:

Comparison of estimated emission factors in the IPCC *Guidelines* between U.S. and European passenger vehicles		
Control Technology	**Emission Factors (g/mi)**	
	U.S. Passenger Vehicles	**European Passenger Vehicles**
LEV	0.064	
Advanced 3 Way	0.274	0.08
Early 3-way	0.274	0.008
Oxidation Catalyst	0.121	0.008
Non-Catalyst	0.032	0.008
Uncontrolled	0.032	0.008

1.1 Control technology terminology

For U.S. vehicles, the following control technology designations are more appropriate than those used in the *Draft Inventory*:

- For "Early three-way catalyst," substitute "Tier 0."

- For "Three-way catalyst" or "Advanced 3 Way," substitute "Tier 1."

Tier 0, Tier 1 and LEV (Low Emission Vehicle) are not control technologies per se, but emissions regulations. They do, however, correspond to combinations of control technology and engine design. Tier 0 refers to standards earlier than Tier 1 that applied to vehicles equipped with three-way catalysts (TWCs). Tier 1s and LEVs both have TWCs, but the data show that their more stringent NO_X standards are associated with lower nitrous oxide emissions as well. The introduction dates for "early three-way catalysts" and "advanced three-way catalysts" in the *Draft Inventory* correspond approximately to the introduction of Tier 0 and Tier 1 emissions regulations (see Table C-7 in the *Draft Inventory* or Section 4.2 below). The assignments of control technologies to model years are revised in Section 4.2.

1.2 Purposes and overview

The purposes of these comments are 1) to review the data supporting the nitrous oxide emission factors used in the *Draft Inventory*, 2) to provide revised emission factors, 3) to recommend changes in other factors affecting nitrous oxide emissions, and 4) to recommend changes in future inventories.

Section 2 reviews the data sources and methods supporting the nitrous oxide emission factors used in the *Draft Inventory*. Section 3 presents the development of revised emission factors. This development is based on a review of the literature (Section 3.1) and on recent tests conducted at EPA's National Vehicle and Fuel Emissions Laboratory (NVFEL) (Section 3.2). Section 4 discusses other issues that affect the calculation of U.S. emissions of nitrous oxide from mobile sources. These include diesel emission factors (Section 4.1), assignment of control technology by model year (Section 4.2), distribution of model years in each calendar year (Section 4.3), and uncertainty (Section 4.4). Section 5 discusses further work to better evaluate the contribution of mobile sources to U.S. emissions of nitrous oxide. Section 6 is a consolidated list of the specific changes recommended for the *Draft Inventory*.

2 AN ANALYSIS OF THE DATA SOURCES AND METHODS USED TO OBTAIN THE EMISSION FACTORS FOR NITROUS OXIDE FROM GASOLINE HIGHWAY VEHICLES IN THE RECENT *DRAFT INVENTORY*

The trail of references from the *Draft Inventory* back to the original data sources is described briefly below. A more detailed analysis is provided in Appendix A.

- The grams/mile emission factors for U.S. mobile sources used in the *Draft Inventory* were taken from the *IPCC Guidelines*.

- The grams/mile emission factors for U.S. vehicles in the *IPCC Guidelines* come from a report prepared by Weaver and Chan (1996): "Mobile source emission factors for global warming gases."

- Weaver and Chan (1996) obtained their grams/mile emission factors from the last column of Table 7 in Ballantyne et al. (1994). The heading of this column is "Current Canadian Estimates: EPS Inventory."

- The reference for the last column of Table 7 in Ballantyne et al. is to Jaques (1992), *Canada's Greenhouse Gas Emissions: Estimates for 1990*, published by the Canadian Government. Ballantyne et al. obtained the grams/mile emission factors (for aged TWCs, new TWCs, and oxidation catalysts) in this column from the grams/kilogram emission factors presented in Jaques, by assuming fuel economies of 9.4, 11.9, and 6 km/L respectively and a standard value for the density of gasoline. The fuel economies are from Jaques's Table 16 and the gasoline density from Table 32. It is not clear where Ballantyne et al. obtained their grams/mile emission factor for non-catalyst vehicles, since it is roughly half the value that would be derived from Jaques by the method described.

- Jaques's emission factors for vehicles without catalysts, vehicles equipped with aged TWCs and vehicles equipped with new TWCs, are the averages of the first two lines of de Soete's (1989) Table XXIX. Jaques converts these averages, which are in units of g/km, to units of g/kg by assuming a uniform fuel economy of 8.5 km/L and a gasoline density of 0.75 kg/L. Jaques's emission factor for oxidation catalyst vehicles is the same as that for new TWC vehicles. Since none of his references support this emission factor for oxidation catalysts, it is possible that he simply adopted the emission factor for new TWCs. The average emission factor for oxidation catalysts from de Soete's Table XIV was 70% higher than the one Jaques uses.

- Lines 1 and 2 of De Soete's Table XXIX are averages of emission factors in his Table XIV. Line 1 is the average of Table XIV lines 2, 4-7, and 11-19, and represents the data from three studies which measured emission factors on a total of five cars tested without catalysts, with new TWCs, and with aged TWCs on various European dynamometer test cycles. Line 2 is the average of Table XIV lines 11-19, and represents the data from a single car tested without a catalyst, with eight new TWCs and with the same eight TWCs bench aged. Therefore, Jaques's average of lines 1 and 2 of de Soete's Table XXIX double weights lines 11-19 from Table XIV. Since the averages are of individual data points, and approximately 80% of the new and aged TWC data come from lines 11-19, Jaques's emission factors for TWCs are derived approximately 90% from a single study involving one car and eight non-production catalysts.

- De Soete's Table XIV lines 11-19 refer to one study, Prigent et al. (1991), in which one car was tested without a catalyst, with eight different non-production catalysts, and then with the same eight catalysts bench-aged. The catalysts were located 1.4 m from the engine.

- Table XIV line 2 refers to Lindskog (1988), in which one non-catalyst car and one car equipped with a TWC were tested on the Swedish driving cycle.

- Table XIV lines 4-7 refer to Prigent et al. (1989), in which two cars were each tested with and without new TWCs.

In summary:

- All the emission factors originate from testing done on five cars using European test cycles. Fuel sulfur content for these tests was unspecified.

- The new and aged TWC emission factors are based 90% 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% of the data for the TWC emission factors came from two studies and three more cars, all tested on European driving cycles only.

- The non-catalyst emission factors were derived from four cars.

- 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 TWCs.

3 IMPROVED ESTIMATES OF EMISSION FACTORS FOR GASOLINE HIGHWAY VEHICLES

Compared to regulated tailpipe emissions, there exist relatively few data that can be used to estimate nitrous oxide emission factors for gasoline highway vehicles. Nitrous oxide is not a criteria pollutant, and measurements of it in automobile exhaust are not routinely collected. Many of the recent measurements have been part of research efforts attempting to understand why and under what conditions TWCs produce nitrous oxide, rather than trying to characterize the U.S. fleet.

OMS determined emission factors for Tier 0 and earlier vehicles primarily from the published literature (Section 3.1). For Tier 1 vehicles and for LEVs, data was used from the recent testing program at NVFEL (Section 3.2). Section 3.3 discusses the limited data that we have for trucks. Section 3.4 summarizes our recommendations for emission factors by vehicle type and control technology.

3.1 Emission factors for Tier 0 and earlier passenger cars

In looking for a better estimate of emission factors, OMS has decided to review only published values for the composite of the standard FTP driving cycle, since it is the standard driving cycle for the U.S. To do otherwise would require reconciling alternative test cycles, tunnel studies, and remote sensing studies—an effort beyond the scope of this review.

To determine emission factors for Tier 0 and earlier vehicles, the following published studies were included in the analysis:

Prigent and de Soete (1989)
Dasch (1992)
Smith and Carey (1982)
Smith and Black (1980)
Urban and Garbe (1979)
Urban and Garbe (1980)
Ballantyne et al. (1994)
Barton and Simpson (1994)
Braddock (1981)

Also included were two measurements of one Tier 0 vehicle that the NVFEL included in its recent study of nitrous oxide emissions from Tier 1 vehicles and LEVs.

Light trucks are analyzed separately, since their emissions are significantly higher than passenger vehicles. The above studies that included light trucks also treated them separately from passenger vehicles. Emission factors for trucks are addressed in Section 3.3 below.

Some authors distinguish "dual bed" catalysts from TWCs, but the distinction is not clear, and we have followed most authors in considering dual-bed catalysts as a form of TWC.

There is evidence that aged TWCs emit more nitrous oxide than new ones. For this reason, we have separated the data into "new" and "aged" (or "old"). "New" means a vehicle that was supplied by a manufacturer for testing and has less than a few thousand miles on the odometer. Everything else is aged or old.

The results are summarized in the following table:

Catalyst age	Parameter	Catalyst Type			
		All	**None**	**Oxidation**	**3-Way**
All ages	mg/mi	56.	17.	51.7	60.5
	n	50	3.	11	36
	std. err. of mean	6.5	13.	19.1	6.8
New	mg/mi	42.7	17.	37.8	47.2
	n	29	3.	4	22
	std. err. of mean	4.8	13.	12.	5.4
Aged	mg/mi	74.2		59.7	81.5
	n	21		7	14
	std. err. of mean	13.2		29.7	13.8

The study by Ballantyne et al. has been excluded from our averaging, because the fuel they used contained 700 ppm sulfur, roughly double what might be expected in U.S. gasoline. The sulfur content of the fuel used in Braddock (1981) was 250 ppm. It was 290 ppm in Urban and Garbe (1979 and 1980), Smith and Carey (1982), and Smith and Black (1980). It was 500 ppm in Barton and Simpson. Sulfur in fuel has been shown to degrade catalyst performance with respect to conventional emissions (see, e.g., Lindhjem 1995 and Monroe et al. 1991). Newly acquired data at NVFEL, discussed below, indicates that emissions of nitrous oxide were significantly higher using Clean Air Act Baseline (CAAB) fuel, a fuel intended to represent a "normal" commercial fuel and which contained 285 ppm sulfur, than when using Indolene, a fuel used in vehicle certification and which contained 24 ppm sulfur. We believe that the higher nitrous oxide emissions were due to the higher sulfur content of CAAB fuel. The fuel analyses and our reasons for believing that the differences in nitrous oxide emissions were due to differences in sulfur content rather than to differences in other fuel parameters are detailed in Appendix B.

For comparison, the following table presents emission factors for new and aged TWCs for all data, for data excluding Ballantyne et al. (1994), and for Ballantyne et al. alone. Units are mg/mi, with the number of data points in parentheses.

	New TWC	Aged TWC
All	50.4 (25)	97.7 (22)
Without Ballantyne et al.	47.2 (22)	81.5 (14)
Ballantyne et al. only	74. (3)	126. (8)

Including Ballantyne would increase the aged TWC emission factor from 0.08 to 0.1 g/mi.

3.2 Emission factors for Tier 1 and LEV vehicles: recent measurements by the NVFEL

A measurement program was undertaken during June and July, 1998, to determine nitrous oxide emissions from aged Tier 1 and LEV vehicles using commercial fuels. 23 vehicles were tested: 18 Tier 1 vehicles, 4 LEVs, and one Tier 0 vehicle that was recruited in error. One of the Tier 1s was recruited specifically to verify the results for a single high-emitting pickup truck. Tier 1 odometers ranged from 16,000 to 75,000 miles. All four LEVs were obtained from their manufacturers. Three of the four were equipped with TWCs that had been bench-aged to 100,000 miles. Three of the odometers read about 5,000 miles; the fourth read about 169,000 miles. Vehicles were tested with air conditioning (A/C) off at 75°F and on at 95°F. All vehicles except one LEV and one Tier 1 were tested using CAAB fuel, a commercial fuel containing 285 ppm sulfur. All of the LEVs and three of the Tier 1 vehicles were tested with Indolene, a low-sulfur fuel used in vehicle certification. The testing schedule and fuel analyses are in Appendix B. The schedule included 23 vehicles and 50 samples.

In order to estimate the emission factors for Tier 1 vehicles, we averaged only tests run with CAAB fuel, and we omitted the second high-emitting pickup truck that was recruited specifically to verify the first one. The following table shows these results:

Tier 1 emission factors from NVFEL program

Vehicles included in average	**Emission factor (mg/mi)**	**Number of vehicles**	**Number of samples**	**Std. err. mean (mg/mi)**	**Range (mg/mi)**
All	63.6	17	29	7.1	24-167
Passenger vehicles	46.3	12	21	5.0	24-124
Light trucks and SUVs	108.9	5	8	11.8	80-167

The emission factor of 46 mg/mi for these Tier 1 passenger vehicles compares favorably with the emission factor of 82 mg/mi for Tier 0 vehicles equipped with TWCs.

The following summarizes the LEV emission factors under our test program. All the LEVs were obtained from their manufacturers. Three had catalysts bench-aged to 100,000 miles.

LEV emission factors from NVFEL program					
Fuel	**Emission factor (mg/mi)**	**Number of vehicles**	**Number of samples**	**Std. err. mean (mg/mi)**	**Range (mg/mi)**
CAAB Fuel	77.8	3	6	14.7	32.-116.
Indolene	28.3	4	8	2.5	14.-36.

LEVs are currently running only in California on low-sulfur fuel, so the emission factor using Indolene is the applicable one.

Emissions were always higher with CAAB Fuel than with Indolene.

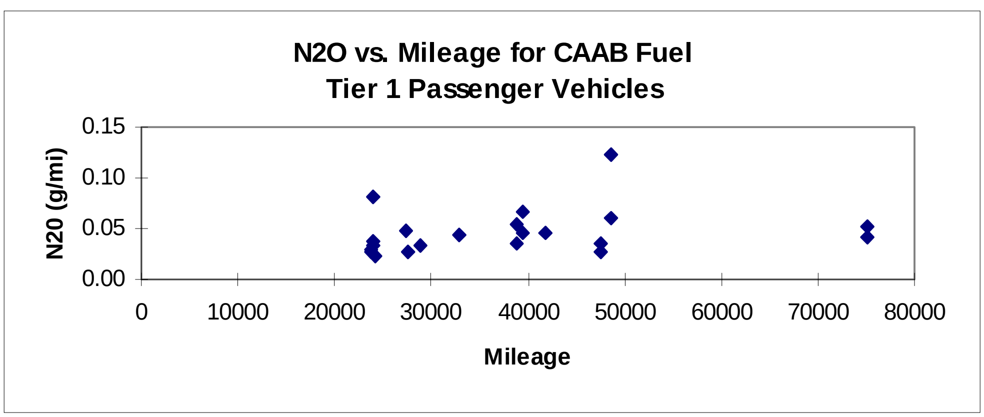
In 8 cases, tests were repeated with both fuels. Six of the tests were with LEVs, and two with Tier 1 vehicles. All showed higher emissions with CAAB than with Indolene. The ratio of nitrous oxide emissions using CAAB to those using indolene ranged from 1.2 to 4.4 and averaged 2.6. The mean of the ratio was significantly larger than 1 ($p<.01$). We believe that the basis for this difference is fuel sulfur content. The fuel analyses and some modeling results supporting this belief are in Appendix B.

Emissions were usually higher with A/C On at 95°F than with A/C off at 75°F

In 22 cases, tests were repeated under both A/C modes. In seventeen cases emissions were higher with A/C on, in five cases with A/C off. The ratio of nitrous oxide emissions with A/C on to those with A/C off ranged from 0.9 to 3.4 and averaged 1.5. The mean of the ratio was significantly larger than 1 ($p<.01$).

Nitrous Oxide was unrelated to the mileage of the vehicles.

A regression of nitrous oxide emission factors against mileage for Tier 1 passenger vehicles yielded a slight positive slope not significantly different from zero ($p<0.25$). R^2 was 0.06.



Barton and Simpson (1994) similarly did not find a significant relationship between nitrous oxide emissions and mileage. Their slope was negative.

Light-duty trucks had higher emissions than passenger vehicles. This result is in agreement with Ballantyne et al. (1994) and with Barton and Simpson (1994).

3.3 Emission factors for gasoline highway vehicles other than passenger cars

Only three of the reviewed studies include data on vehicles other than passenger vehicles. All the non-passenger vehicles were light duty trucks equipped with TWCs. The results are summarized in the following table:

Study	**Age**	**Emission factors (mg/mi) (number of vehicles)**		**Trucks/PVs (ratio)**
		Light-duty trucks	**Passenger vehicles**	
NVFEL	Old	109 (5)	46 (12)	2.4
Ballantyne et al. (1994)	All	188 (3)	111(11)	1.7
	Old	93 (1)	126 (8)	0.73
	New	236 (2)	74 (3)	3.2
Barton and Simpson (1994)	All	163 (3)	75 (11)	2.2
	Old	300 (1)	80 (11)	3.8
	New	95 (1)	55 (2)	1.7
Average				2.2

While the data are limited and not without exception, they are fairly convincing that light-duty trucks emit more nitrous oxide per mile than passenger vehicles.

In the absence of a better alternative, we recommend that emission factors for passenger vehicles be applied to other gasoline highway vehicles in proportion to their fuel economy, which is the same practice employed in the *Draft Inventory*. For this purpose, we have used the fuel economies specified by Weaver and Chan (1996) and incorporated into the *IPCC Guidelines*. They are listed in Appendix C. According to Chan (1998), they were obtained from MOBILE5 and then reduced by 15%. The use of fuel-consumption ratios to determine emission factors should be considered a temporary measure only, to be replaced as soon as real data are available.

3.4 Recommended emission factors for gasoline highway vehicles

Passenger vehicles

A list of the revised emission factors is presented in Section 6.1. Except for LEVs as specified, it is assumed that these vehicles are being operated on a standard commercial fuel containing about 300 ppm sulfur. Aged TWCs emit more than new TWCs, but we believe aging happens fairly early, so we assume most of the fleet is aged. There are no data to assign a mileage to this transition.

Control Technology	Emission Factor (mg/mi)	n	Std. Err. Mean (mg/mi)	Range (mg/mi)	Emission Factor (mg/km)*
Non-catalyst	16.6	3	13.0	2-42	10.3
Oxidation catalyst	51.7	11	19.1	8-233	32.2
Tier 0	81.5	12	13.8	6-190	50.7
Tier 1	46.3	21	5.0	24-124	28.8
LEVs on standard fuel	77.8	6	14.7	32-116	48.4
LEVs on low-S fuel	28.3	8	2.5	14-36	17.4
* Extra precision has been included so conversion between units does not introduce a significant difference.					

Summary of Sources:

Control Technology	Data Source
Non-catalyst	Prigent and de Soete (1989), Dasch (1992), and Urban and Garbe (1979)
Oxidation catalyst	Smith and Carey (1982), Urban and Garbe (1979)
Tier 0	Smith and Carey (1982), Barton and Simpson (1994), and NVFEL (1998) (one car). Only old cars were included. Ballantyne et al. (1994) was excluded because of high fuel sulfur content (700 ppm).
Tier 1	NVFEL (1998). CAAB fuel, both A/C modes.
LEVs on standard fuel	NVFEL (1998). CAAB fuel, both A/C modes.

LEVs on low-sulfur fuel	NVFEL (1998). Indolene fuel, both A/C modes.

<u>Gasoline highway vehicles other than passenger vehicles</u>

A list of the revised emission factors is presented in Section 6.1. We have used fuel-specific emission factors, as was done in the *Draft Inventory*. That is, we use the preceding emission factors for passenger vehicles, adjusted by the ratio of the fuel economies of passenger vehicles and the other vehicle type. The data that support this practice are that light trucks emit more nitrous oxide than passenger vehicles (see Section 3.3). The data are not good enough to say how much more, but fuel-specific emission factors seem an appropriate estimate at this time. The increasing proportion of light trucks in the U.S. fleet emphasizes the need to collect additional data.

We have used the fuel economies in the *IPCC Guidelines* for calculating fuel-specific emission factors. These fuel economies came from MOBILE5, reduced by 15% (Chan 1998). While it is likely that these estimates of fuel economy can be improved, it is only their ratios that are being used in this context. The use of fuel-consumption ratios to determine emission factors should be considered a temporary measure only, to be replaced as soon as real data are available.

Note that for Gasoline Heavy-Duty Vehicles the emission factors in Table C-8 of the *Draft Inventory* specified as Catalyst and Non-Catalyst Control were actually the fuel-specific values for Advanced 3-Way and Early 3-Way. This error is also present in the *IPCC Guidelines* and in Weaver and Chan (1996).

4 OTHER ISSUES

4.1 Diesel emission factors

Weaver and Chan (1996) cite Dietzmann et al. 1980 (SAE 801371) as the basis for nitrous oxide emission factors for heavy-duty diesel trucks, saying that they averaged the Dietzmann et al. values for heavy-duty trucks and estimated emission factors for lighter duty vehicles by assuming fuel-specific emission factors. Four engines were studied in Dietzman et al., one from 1977 and three from 1979. The 1979 engines were required to meet more stringent emissions standards. The average nitrous oxide emission factors for Dietzman et al.'s three 1979 engines were 31, 55, and 40 mg/mi. The 1977 engine emitted 76 mg/mi. The average of the four values is 50.5 mg/mi = 31.4 mg/km, which is the value Weaver and Chan use for uncontrolled HDDVs. Fuel-specific emission factors seem to have been applied inconsistently to other diesel classes. For example, 63 mg/km is assigned to light-duty diesels with moderate control. Application of fuel-consumption proportionality yields emission factors of about 10 mg/km for light-duty trucks and 8 mg/km for passenger vehicles. The *IPCC Guidelines* values for European diesels (Tables 1-37 to 1-39) are 30, 20, and 10 mg/km for heavy-duty, light-duty, and passenger vehicles respectively. The values in Dietzmann, Weaver and Chan, and the European tables in the *IPCC Guidelines* are all quite low and in the same range. Because of very limited data and greater European experience with diesel, OMS recommends taking the European values from the *IPCC Guidelines*: 30, 20, and 10 mg/km for heavy-duty, light-duty, and passenger vehicles respectively.

Vehicle type and control technology	***Draft Inventory* (g/km)**	**Fuel-specific based on Dietzmann et al. (1980) (g/km)**	**European (g/km)**
Diesel Passenger Cars			
Control Technology			
Advanced	0.0070	0.0068	0.0100
Moderate	0.0100	0.0071	0.0100
Uncontrolled	0.0140	0.0091	0.0100
Diesel Light Trucks			
Control Technology			
Advanced	0.0240	0.0094	0.0200
Moderate	0.0630	0.0095	0.0200
Uncontrolled	0.0310	0.0119	0.0200
Diesel Heavy-Duty Vehicles			
Control Technology			
Advanced	0.0250	0.0283	0.0300
Moderate	0.0250	0.0289	0.0300
Uncontrolled	0.0310	0.0314	0.0300

4.2 Control technologies and their assignment by model year.

A small section of Table C-7 of the *Draft Inventory* is shown below:

U.S. Greenhouse Gas Emissions and Sinks: 1990-1996 Page 152	
Table C-7: Control Technology Assignments for Highway Mobile Sources	
Vehicle Type/Technology	Model Years
Gasoline Passenger Cars and Light-Duty Trucks	
Uncontrolled	1966-1972
Non-catalyst controls	1973-1977
Oxidation catalyst	1978-1982
Early three-way catalyst	1983-1995
Three-way catalyst	1996
Low emission vehicle*	1996

The following control technology designations are more appropriate for U.S. vehicles:

- For "Early three-way catalyst," substitute "Tier 0."
- For "Three-way catalyst," which is referred to in Table C-8 (Emission Factors) as "Advanced 3 Way," substitute "Tier 1."

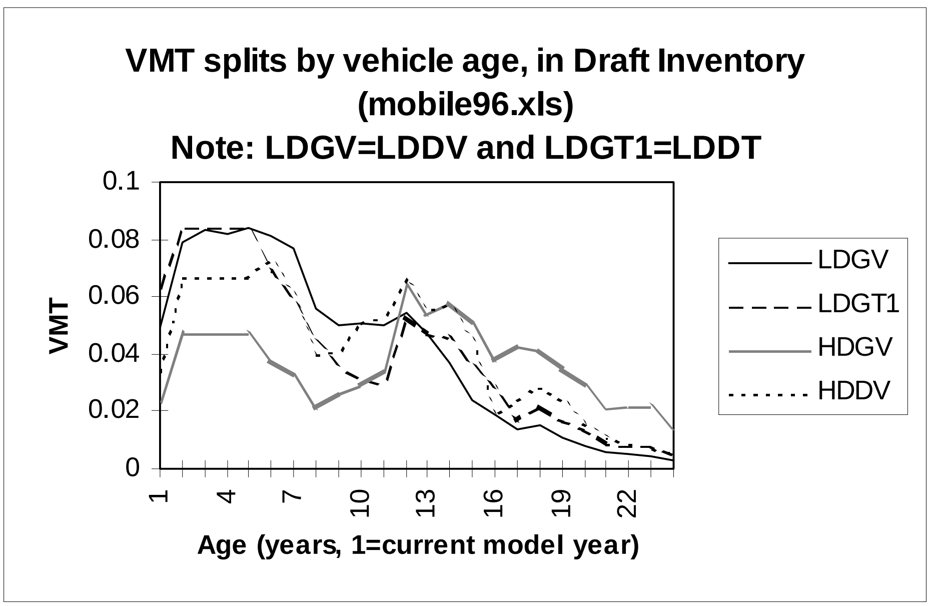
See Section 1.1 above for additional discussion of this issue.

Our revised assignment of technologies by model year are detailed in the tables in Section 6.2. Our principal source for this data is the "Compilation of Air Pollutant Emission Factors, Volume II: Mobile Sources" (U.S. EPA 1998), commonly referred to as AP42. Additional information concerning the phase-in of Tier 1 and LEV technologies and schedules for California have been provided by our MOBILE team.

A significant change from the way the *Draft Inventory* technology assignments were done is the splitting of a single model year between more than one technology. We felt it was especially important to do this for later model years, which make up a large proportion of the fleet. The effect of our revisions is to introduce technologies earlier than they were introduced in the *Draft Inventory*.

4.3 Distribution of VMT by vehicle age for each calendar year

The table of fraction of VMT by vehicle age that was used for all calendar years in the *Draft Inventory* is plotted in the figure below. Each vehicle type is plotted with a separate line.



The irregularity of the plot indicates that these values represents data for a particular year. However, the spreadsheet used in the *Draft Inventory* applies this table to all years from 1990 to 1996. The table has a large peak for vehicles that are eleven years old, reflecting large purchases of new vehicles in that model year. When this table is applied to other years than the one for which the data apply, this peak will be incorrectly associated with other model years. As a matter of documentation, the year from which the data for this table were taken and the source of the data should be specified.

4.4 Uncertainty estimates

Various places in the *Draft Inventory* contain discussions of uncertainty, but the Executive Summary and Annex C do not. The discussion of uncertainty on p. 27 should be repeated in both the Executive Summary and Annex C. The data in these locations otherwise give an impression of far greater precision than is warranted.

5 EFFORTS THAT WOULD IMPROVE FUTURE INVENTORIES

5.1 Measure the nitrous oxide emissions of in-use vehicles

There is a great need for additional data. Nitrous oxide emissions from in-use vehicles should be measured in as many testing programs as possible. In programs where an FTIR is being used, adding the analysis of nitrous oxide should be relatively simple.

Heavier gasoline vehicles should be tested to determine their emission factors. The light truck fleet is becoming a larger proportion of the U.S. total and therefore needs to be well characterized. The current stratagem of using fuel-specific emission factors is suitable only as a temporary measure.

The effect of sulfur on nitrous oxide emissions should be studied, on different vehicle types, with and without catalysts. It appears that sulfur has a strong effect on nitrous oxide emissions. Emission factors for vehicles with TWCs may prove to be a strong function of the sulfur content of the fuel used.

Diesel vehicles of all weight classes should be tested. Routine testing should include nitrous oxide. We need data on in-use vehicles, and, as new control technologies are developed, we will need data on how those technologies affect nitrous oxide emissions.

The large variability in nitrous oxide emissions should be understood. Such knowledge might lead to changes in catalyst design and configuration that would eliminate high emitters. Second-by-second studies of low and high emitters would probably yield good insight into the problem, and provide some productive hypotheses for further testing.

5.2 Refine estimates of fleet composition and activity

Separate tables of VMT fraction by vehicle age should be developed for each historical calendar year for which an inventory is prepared.

VMT estimates could benefit from close scrutiny and comparison between sources.

VMT and fuel-sales-based estimates should be reconciled.

5.3 Analyze additional sources in the literature

While only further testing will provide the real data we need, some additional value can be obtained by a more exhaustive review of the literature.

- Authors who tested vehicles using the FTP, but did not report the composite number we need for consistency, might be willing to supply that data if requested. For example:

  - Laurikko and Paivi (1995) tested five cars of different mileages at different temperatures on the FTP cycle, but only reported bags 1 and 3.

  - Joumard et al. (1996) tested 25 private cars, some with and some without catalysts, on a variety of driving cycles, including the FTP, but nitrous oxide was not reported for the FTP.

- Careful analysis of European and Japanese driving cycles could possibly yield data comparable to those from the FTP cycle.

5.4 Develop estimates of uncertainty

Estimates of uncertainty should be developed in future Inventories.

5.5 Include nitrous oxide as part of a future version of MOBILE

Incorporating nitrous oxide into MOBILE would assure that our knowledge of nitrous oxide emissions by mobile sources is represented in a way consistent with other mobile emissions. It would also simplify the generation of an annual inventory.

5.6 Integrate with the Trends process

The estimates of nitrous oxide emissions from mobile sources should be integrated with the process by which OAQPS produces the *National Air Pollutant Emission Trends Data Base*. This approach would avoid duplication of effort and improve consistency across EPA.

6 CONSOLIDATED LIST OF SPECIFIC CHANGES TO THE *DRAFT INVENTORY*

6.1 Revised nitrous oxide emission factors for highway mobile sources

The following table lists the revised emission factors. It corresponds to Table C-8 in Annex C of the *Draft Inventory*. The rationale for these emission factors is detailed in the body and appendices of these comments. We have included more significant figures than is warranted by their uncertainty to assure consistent calculations when using different units. Note that instead of

"Early" and "Advanced" TWCs, we use the terms "Tier 0" and "Tier 1".

Vehicle type and control technology	Nitrous Oxide Emission Factors	
	g/mi	g/km
Gasoline Passenger Cars		
Control Technology		
Low Emission Vehicles*	0.0283	0.0176
Tier 1	0.0463	0.0288
Tier 0	0.0815	0.0507
Oxidation Catalyst	0.0517	0.0322
Non-Catalyst	0.0166	0.0103
Uncontrolled	0.0166	0.0103
* Applicable to California VMT only		
Gasoline Light-Duty Trucks		
Control Technology		
Low Emission Vehicles*	0.0400	0.0249
Tier 1	0.0643	0.0400
Tier 0	0.1362	0.0846
Oxidation Catalyst	0.0673	0.0418
Non-Catalyst	0.0188	0.0117
Uncontrolled	0.0190	0.0118
* Applicable to California VMT only		
Gasoline Heavy-Duty Vehicles		
Control Technology		
Tier 0	0.2781	0.1729
Oxidation Catalyst	0.1400	0.0870
Non-catalyst	0.0412	0.0256
Uncontrolled	0.0432	0.0269
Diesel Passenger Cars		
Control Technology		
Advanced	0.0161	0.0100
Moderate	0.0161	0.0100
Uncontrolled	0.0161	0.0100
Diesel Light Trucks		
Control Technology		
Advanced	0.0322	0.0200
Moderate	0.0322	0.0200
Uncontrolled	0.0322	0.0200
Diesel Heavy-Duty Vehicles		
Control Technology		
Advanced	0.0483	0.0300
Moderate	0.0483	0.0300
Uncontrolled	0.0483	0.0300
Motorcycles		
Control Technology		
Non-Catalyst Control	0.0068	0.0042
Uncontrolled	0.0087	0.0054

6.2 Revised technology assignments by model year for gasoline highway vehicles except

motorcycles

For Gasoline Passenger Cars (light duty gas vehicles, LDGV), except California:

Model Year	Percentage of 49 States LDGV with each control technology				
	Uncontrolled	**Non-catalyst control**	**Oxidation**	**Tier 0**	**Tier 1**
≤1972	100				
1973-1974		100			
1975		20	80		
1976-1977		15	85		
1978-1979		10	90		
1980		5	88	7	
1981			15	85	
1982			14	86	
1983			12	88	
1984-1993				100	
1994				60	40
1995				20	80
1996					100

For Gasoline Light Duty Trucks (LDGT), except California:

Model Year	Percentage of 49 States LDGT with each control technology				
	Uncontrolled	Non-catalyst control	Oxidation	Tier 0	Tier 1
≤1972	100				
1973-1974		100			
1975		30	70		
1976		20	80		
1977-1978		25	75		
1979-1980		20	80		
1981			95	5	
1982			90	10	
1983			80	20	
1984			70	30	
1985			60	40	
1986			50	50	
1987-1993			5	95	
1994				60	40
1995				20	80
1996					100

For Gasoline Heavy-Duty Vehicles (heavy-duty gas vehicles, HDGV):

Model Year	Percentage of national HDGV with each control technology			
	Uncontrolled	Non-catalyst control	Oxidation	Tier 0
≤1981	100			
1982-1984	95		5	
1985-1986		95	5	
1987		70	15	15
1988-1989		60	25	15
1990-2003		45	30	25
2004				100

For California Gasoline Passenger Cars and Light-Duty Trucks (light duty gas vehicles and trucks, LDGV and LDGT):

Model Year	**Percentage of California LDGV and LDGT fleet with each control technology**					
	Uncontrolled	**Non-catalyst control**	**Oxidation**	**Tier 0**	**Tier 1**	**LEV**
≤1972	100					
1973-1974		100				
1975-1979			100			
1980-1981			15	85		
1982			14	86		
1983			12	88		
1984-1991				100		
1992				60	40	
1993				20	80	
1994					90	10
1995					85	15
1996					80	20

6.3 Document distribution of VMT by vehicle age for each calendar year

The existing table of VMT by vehicle age is for a particular but unspecified year. As a matter of documentation, the year from which the data for this table were taken and the source of the data should be specified.

6.4 Include a discussion of uncertainty in the Executive Summary and Annex C

Various places in the *Draft Inventory* contain discussions of uncertainty, but the Executive Summary and Annex C do not. The discussion of uncertainty on p. 27 should be repeated in both the Executive Summary and Annex C. The data in these locations otherwise give an impressions of far greater precision than is warranted.

7 REFERENCES

Ballantyne, Vera F., Peter Howes, and Leif Stephanson. 1994. "Nitrous Oxide Emissions from Light Duty Vehicles." SAE Paper 940304.

Barton, Peter and Jackie Simpson. 1994. "The effects of aged catalysts and cold ambient temperatures on nitrous oxide emissions." Mobile Sources Emissions Division (MSED), Environment Canada, MSED Report #94-21.

Braddock, James N. 1981. "Impact of low ambient temperature on 3-way catalyst car emissions." SAE paper 810280.

Chan, Lit-Mian. 1998. Phone call July 23.

Dasch, Jean Muhlbaler. "Nitrous Oxide Emissions from Vehicles." January, 1992. *Journal of the Air and Waste Management Association*, 42(1):63-67.

De Soete, Gerard G. 1993. "20. Nitrous oxide from combustion and industry: chemistry, emissions and control." In A. R. van Amstel (ed.) *Proceedings of an International IPCC Workshop on Methane and Nitrous Oxide : Methods in National Emissions Inventories and Options for Control*. RIVM Report No. 481507003, Bilthoven, The Netherlands.

De Soete, G. 1989. "Updated evaluation of nitrous oxide emissions from industrial fossil fuel combustion," draft final report prepared for the European Atomic Energy Community, Institut Francais du Petrole, Ref 37-559.

Dietzmann, Harry E., Mary Ann Parness, and Ronald L. Bradow. 1980. "Emissions from Trucks by Chassis Version of 1983 Transient Procedure." SAE Paper 801371.

IPCC/UNEP/OECD/IEA. 1997. *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories*. Paris: Intergovernmental Panel on Climate Change, United Nations Environment Programme, Organization for Economic Co-Operation and Development, International Energy Agency.

Jaques, A. P. 1992. *Canada's Greenhouse Gas Emissions: Estimates for 1990*. Environmental Protection Series, Report EPS 5/AP/4, December 1992. Environmental Protection, Conservation and Protection, Environment Canada.

Joumard, Robert, Robert Vidon, Laurent Paturel, and Gerard de Soete. 1996. "Changes in pollutant emissions from passenger cars under cold start conditions." SAE Paper 961133.

Laurikko, Juhani and Paivi Aakko. 1995. "The effect of ambient temperature on the emissions of some nitrogen compounds: a comparative study on low-, medium- and high-mileage three-way catalyst vehicles." SAE paper 950933.

Lindhjem, Christian E. 1995. "The effect of gasoline reformulation and sulfur reduction on exhaust emissions from post-1983 but pre-1990 vehicles." SAE paper 950778.

Lindskog, A. 1988. Data presented at the EPA/IFP European Workshop on the Emission of

Nitrous Oxide from Fossil Fuel Combustion, Figures 5-18 and 5-19. Rueil-Malmaison, France, June 1-2, 1988. EPA Report EPA/600/13.

Monroe, David R., Martin H. Krueger, Donald D. Beck and Michael J. D'Aniello, Jr. 1991. "The effect of sulfur on three-way catalysts." in A. Crucq (Editor), *Catalysis and Automotive Pollution Control II*, proceedings of the Second International Symposium (CAPoC 2), Brussels, Belgium, September 10-13, 1990. New York: Elsevier Science Publishers.

Neitzert, Frank. 1998. Personal communication.

Prigent, M., G. de Soete, and R. Doziere. 1991. "The effect of aging on nitrous oxide N_2O formation by automotive three-way catalysts," in A. Crucq (Editor), *Catalysis and Automotive Pollution Control II*, proceedings of the Second International Symposium (CAPoC 2), Brussels, Belgium, September 10-13, 1990. New York: Elsevier Science Publishers.

Prigent, Michel and Gerard De Soete. 1989. "Nitrous oxide N_2O in engines exhaust gases—a first appraisal of catalyst impact." SAE Paper 890492.

Prigent, M. and G. G. De Soete. 1992. "The increase of N_2O emissions caused by three-way automotive catalysts." NIRE/IFP/EPA/SCEJ 5th Interrnational Workshop on N_2O Emissions, 1-3 July 1992, Tsukuba, Japan, paper 10-3. We do not have this document. Data from it is referenced in de Soete (1993).

Smith, L. R. and F. M. Black. 1980. "Characterization of exhaust emissions from passenger cars equipped with three-way catalyst control systems." SAE paper 800822.

Smith, Lawrence R. and Penny M. Carey. 1982. "Characterization of exhaust emissions from high mileage catalyst-equipped automobiles." SAE paper 820783.

Urban, Charles M. and Robert J. Garbe. 1979. "Regulated and Unregulated Exhaust Emissions from Malfunctioning Automobiles." SAE Paper 79696.

Urban, Charles M. and Robert J. Garbe. 1980. "Exhaust Emissions from Malfunctioning Three-Way Catalyst-Equipped Automobiles." SAE Paper 800511.

U.S. EPA. 1997. *The 1997 U.S. Climate Action Report*, Submitted by the United States of America Under the United Nations Framework Convention on Climate Change.

U.S. EPA. 1998. *Inventory of U.S. Greehouse Gas Emissions and Sinks 1990-1996.* Draft, 3/10/98. U.S. EPA, Office of Policy, Planning, and Evaluation. EPA-230-R-97-002.

U.S. EPA. 1998. "Compilation of Air Pollutant Emission Factors, Volume II: Mobile Sources," commonly referred to as AP-42, pending 5th edition, last updated: 06 April 1998. Accessed from the web site: http://www.epa.gov/omswww/ap42.htm.

U.S. EPA. 1989. "EPA/IPF European Workshop on the emission of nitrous oxide from fossil fuel combustion (Rueil-Malmaison, France)," Prepared by Air and Energy Engineering Research, Research Triangle Park, Raleigh, North Carolina, Report No. EPA-600/9-89-089.

Weaver, Christopher S., Lit-Mian Chan. 1996. "Mobile source emission factors for global warming gases." Draft Final Report, June 24, 1996, submitted to ICF, Inc., 1850 K Street, NW, Suite 1000, Washington DC by Engine, Fuel, and Emissions Engineering, Inc., 9812 Old Winery Place, suite 22, Sacramento, CA 95827.

Weaver, Christopher S. 1998. Telephone conversation on July 10.

APPENDIX A

A DETAILED DISCUSSION OF THE ORIGIN OF THE EMISSION FACTORS FOR NITROUS OXIDE FOR GASOLINE HIGHWAY VEHICLES IN THE *DRAFT INVENTORY*

The following sections detail the number trail backward from the *Draft Inventory* to original sources, supporting the more limited description in the body of the comments.

A. The *Draft Inventory*, the *IPCC Guidelines*, Weaver and Chan (1996).

The *Draft Inventory* lists emission factors that are identical to those of the *IPCC Guidelines*, which in turn are identical to those of Weaver and Chan (1996), who are the source of these values (Weaver 1998). For light-duty passenger gasoline highway vehicles, the emissions factors are as follows:

	U.S. Draft Inventory		*IPCC Guidelines*				
	g/km	Calc[a] g/mi	g/kg	km/L	g/km	Calc[b] g/km	Calc[c] g/mi
Low Emission Vehicles	0.04	.064	0.453	8.5	0.040	0.04	0.064
Advanced 3 Way	0.17	.274	1.892	8.3	0.170	0.171	0.275
Early 3-way	0.17	.274	1.81	8	0.170	0.17	0.273
Oxidation Catalyst	0.075	.121	0.622	6.2	0.075	0.075	0.121
Non-Catalyst	0.02	.032	0.125	4.5	0.020	0.021	0.034
Uncontrolled	0.02	.032	0.13	4.7	0.020	0.021	0.033

[a] Calculated from g/km by conversion factor for km/mi
[b] Calculated from g/kg using km/L and 0.75kg/L
[c] Calculated from the calculated g/km by conversion factor for km/mi

The calculations in this table have been done to verify the internal consistency of the emission factors expressed in different units in the *IPCC Guidelines* and to examine how the precision shown affects the interconversion of units. The emission factors listed as g/km in the *Draft Inventory* are identical to the factors listed as g/km in the *IPCC Guidelines.* The tables of emission factors in the *IPCC Guidelines* are identical to those in Weaver and Chan (1996).

B. From Ballantyne et al. (1994) to Weaver and Chan (1996)

Weaver and Chan (1996) got their emission factors from the last column of Table 7 in Ballantyne et al. (1994), headed "Current Canadian Estimates: EPS Inventory:"

Comparison of control-technology terminology and emission factors between Ballantyne et al. (1994) and Weaver and Chan (1996)				
Ballantyne (1994) Table 7			**Weaver and Chan (1996)**	
Catalyst Type	**Current Canadian Estimates: EPS Inventory (mg/mi)**	**Units conversion to (g/km)**	**Control Technology**	**Emission Factor (g/km)**
New 3-way	60	.037	LEV	0.040
Aged 3-way	280	.174	Early 3-Way, Three-way	0.170
Oxidation	120	.075	Oxidation	0.075
None	32	.020	Uncontrolled, Non-Catalyst	0.020

Of the four sets of estimates in Ballantyne et al.'s Table 7, the reason for choosing this one, according to Weaver (1998), was the wide range of the estimates, the difficulty of reconciling them, and the fact that the Canadian emission factors appeared to be official government figures.

Weaver and Chan (1996) assumed both advanced and early TWCs to have the same nitrous oxide emissions properties. Weaver (1998) indicated that since catalyst aging occurs relatively quickly, all TWCs were assumed to be aged. He further indicated that he and Chan reasoned that LEVs would behave like new TWCs, because part of LEV technology was fine tuning catalyst placement and other factors to insure quick light-off, and that the aging effect was likely due to delayed light-off. Therefore LEVs would behave like new TWC vehicles.

C. From Jaques (1992) to Ballantyne et al. (1994)

The last column of Ballantyne et al.'s Table 7 originated, with some modification, from *Canada's Greenhouse Gas Emissions: Estimates for 1990* (Jaques 1992). Ballantyne et al. appear to have converted Jaques's units (g/kg) into g/mi by using the fuel economies in Jaques's Table 16 and assuming a fuel density of 0.75 kg/L (Jaques's Table 32), as we show in the following table:

Catalyst Type	Jaques (1992)			Ballantyne et al. (1994) Table 7 Current Canadian Estimates (mg/mi)
	Table 31 (g/kg)	Table 16 Fuel Economy (km/L)	Table 31/ Table 16 (mg/mi)	
New	0.6	11.9	61	60
Aged	2.2	9.4	282	280
Ox	0.6	6	121	120
None	0.31	6	62	32

A problem with this derivation is that it produces 62 mg/mi for non-catalyst vehicles, but Ballantyne et al. list 32 mg/mi. The table below shows the column of Ballantyne's Table 7 giving the range of de Soete's (1989) estimates. We have produced the last column below (Actual Ranges) directly from de Soete's (1989) Table XIV.

Catalyst Type	Ballantyne et al. (1994), Table 7		de Soete's (1989) Actual Ranges from Table XIV all points lines 2,4-7, 11-19 (mg/mi)
	Current Canadian Estimates (mg/mi)	de Soete, 1989 Range (mg/mi)	
New	60	60-170	54-141
Aged	280	260-355	50-1000
Ox	120	120	112-257
None	32	8-32	13-151

Ballantyne et al.'s listing of de Soete's (1989) ranges suggest that they took what they understood to be the high end of the non-catalyst range rather than the value provided by Jaques (1992), and mis-attributed it to Jaques. However, the last column of the above table shows de Soete's (1989) actual ranges as well as we have been able to determine them. One could suppose that Ballantyne et al. excluded outlier values, but the high end of Ballantyne et al.'s (1994) new TWC range and the low end of their non-catalyst range lie outside the actual ranges. Ballantyne, in an email through Stephanson (one of Ballantyne's co-authors), was unable to recall the origin of the non-catalyst emission factor.

Another problem in using the last column in Ballantyne et al.'s Table 7 to represent the Current Canadian Estimates is that Jaques derived his emission factors from g/km data in de Soete (1989) and converted them to g/kg by assuming a uniform fuel economy of 8.5km/L (Neitzert 1998). Ballantyne et al. then converted these numbers (except for the non-catalyst case) to units of g/mi by assuming a different set of fuel economies. The following table compares Ballantyne et al.'s "Current Canadian Estimates" to Jaques's, using the same fuel economy he assumed in deriving them.

Catalyst Type	Ballantyne et al. (1994) Current Canadian Estimates (mg/mi)	Jaques (1992) assuming 8.5km/L (mg/mi)
New	60	85
Aged	280	312
Ox	120	85
None	32	44

D. From de Soete (1989) to Jaques (1992)

The official Canadian estimates for 1990 (Jaques 1992) have been derived primarily from de Soete (1989). Neitzert (1998) said that the emission factors in Jaques (1992) for new TWC, aged TWC and non-catalyst vehicles were obtained by averaging lines one and two of Table XXIX (de Soete 1989) and assuming a fuel economy of 8.5 km/L and a fuel density of 0.75kg/L. The following table demonstrates this derivation and also shows emission factors in g/km and g/mi:

	Uncontrolled	New TWC	Aged TWC
line 1, Table XXIX (gN/km)	0.026	0.03	0.137
line 2, Table XXIX (gN/km)	0.008	0.037	0.11
line 1, Table XXIX (gN_2O/km)	0.041	0.048	0.216
line 2, Table XXIX (gN_2O/km)	0.013	0.059	0.173
line 1, Table XXIX (gN_2O/mi)	0.066	0.077	0.347
line 2, Table XXIX (gN_2O/mi)	0.021	0.095	0.278
avg of lines 1 and 2 (gN_2O/km)	0.027	0.053	0.194
avg of lines 1 and 2 (gN_2O/mi)	0.044	0.086	0.313
Assuming 8.5km/L,.75kg/L (gN_2O/kg)	0.307	0.604	2.201
Jaques (1992), Table 31 (gN_2O/kg)	0.31	0.6	2.2

Comparison of the last two lines shows that we have successfully reproduced Jaques's emission factors, except for the oxidation catalyst case.

Next, we must ask where Jaques's oxidation catalyst emission factor of 0.6 g/kg originated. De Soete's Table XXIX is a summary table for his Table XIV, which lists five emission factors for vehicles equipped with oxidation catalysts. The average of these is 0.092 g/km, which converts to 1.04 g/kg and does not match Jaques's emission factor. We believe the most plausible explanation of Jaques's oxidation catalyst emission factor is that, in the absence of data lower than the new TWC data, it was simply taken to be identical to the new TWC emission factor. Neitzert (1998) did not know how the oxidation catalyst emission factor was derived.

In the next section, we examine the original data sources.

E. De Soete's (1989) original sources

The averages in lines 1 and 2 of Table XXIX are referenced to lines 2, 4-7, and 11 of Table XIV. Each of the Table XIV lines referenced is actually a series of individual data points. Below are lines 1 and 2 of de Soete's Table XXIX:

Emission factor (g N_2O as N/km)	Uncontrolled (without catalyst)	With Three-Way Catalyst	
		New	Aged
Averaged over all cycles: ECE cold, ECE hot, EUDC and SDC (Table XIV, lines 2,4 to 7, 11 to 19)	0.0261	0.0304	0.1373
Averaged over all cycles: ECE cold, ECE hot and EUDC (Table XIV, lines 11 through 19)	0.0084	0.0374	0.1099

Note that the values in this table are averaged over European driving cycles only. There are a few FTP cycles included in Table XIV, but they are not included in the averages in Table XXIX.

Below, we have attempted to replicate Table XXIX by averaging the specified lines in Table XIV:

Lines of Table XIV used in average	Uncontrolled (without catalyst)	With Three-Way Catalyst	
		New	Aged
2,4 to 7, 11 to 19	0.0236	0.0355	0.1177
11 to 19	0.0085	0.0374	0.11
2,4 to 7	0.0284	0.0292	0.1486
Lines 2 & 3 of this table	0.0184	0.0333	0.1293

We have succeeded in replicating line 2 of Table XXIX, but not line 1.

This table shows the data sources contributing to Jaques (1992). Table XIV is in de Soete (1989).

Data Source	Data Points for[a]				Reference[b]
	no cat.	new TWC	aged TWC	ox cat.	
Table XIV, line 2	6 24% 1		6 20% 1		42. Lindskog, A., data presented at the EPA/IFP Workshop 1988, report on that meeting p. 103 and tables 5-18 and 5-19.
Table XIV, lines 4-7	13 52% 2	15 24%			43, see also 44 and 45. 43. Prigent, M., Doziere, R. and De Soete, G., unpublished document of the French Petroleum Institute, 1988. 44. Prigent, M., data presented at the EPA/IFP Workshop 1988, p. 103-109 and figs 5-7 and 5-8 45. Prigent, Michel and Gerard De Soete. 1989. "Nitrous oxide N_2O in engines exhaust gases—a first appraisal of catalyst impact." SAE Paper 890492.
Table XIV, lines 11-19	6 24% 1	48 76%	24 80%		47. Prigent, M.F., de Soete, G.G. and Doziere, R., "The effect of catalyst aging on nitrous oxide emissions from automobiles with a three-way catalyst," to be presented at the CAPoC Meeting, Brussels, 10-13 September 1990.
Total	25	63	30	0	

a. Top number is the number of data points. The next number is the percentage of total data points that reference comprises for the control category. The third number is the number of vehicles tested. If there is only one vehicle number in a row, the same vehicle or vehicles were used to test all control technologies.
b. The numbers are those used in Table XIV to indicate the references.

42. Lindskog 1988 reported on two cars on the SDC (Swedish driving cycle), a Volvo 240 without a catalyst, and a Volvo 260 with 10,000 km with a TWC.

45. Prigent and de Soete. 1989. "Nitrous oxide N_2O in engines exhaust gases—a first appraisal of catalyst impact." SAE Paper 890492 is apparently the same material presented at the EPA/IFP Workshop in 1988. Two cars were tested, a Citroen BX19GT and a Renault Fuego U.S. version,

with and without TWCs.

47. Prigent, M., G. de Soete, and R. Doziere. 1991. "The effect of aging on nitrous oxide N_2O formation by automotive three-way catalysts," in A. Crucq (Editor), *Catalysis and Automotive Pollution Control II*, proceedings of the Second International Symposium (CAPoC 2), Brussels, Belgium, September 10-13, 1990. New York: Elsevier Science Publishers. These tests were performed using 8 different catalysts and two similar 2.2L 4 cylinder engines. One was for aging the catalysts, the other, mounted in an unspecified chassis, was used for running the tests. The catalysts were mounted 1.4 m from the engine. They were not production catalysts but apparently were fabricated for these tests. This reference gives measurements in g/test graphically and ratios between catalyst and non-catalyst vehicles numerically. However, the individual test results in g/km are given in Table XIV of de Soete (1989). The test vehicle and engine are unidentified in the reference, but are identified in Table XIV as a "Fuego, 2.21 L engine equipped with electronic fuel injection + oxygen sensor, closed loop."

A subsequent publication by the same researchers apparently involving a different vehicle and the same or a similar set of new and aged catalysts, gives sharply lower nitrous oxide emission factors: the averages were 12 mg/mi for non-catalyst vehicles, 29 mg/mi for new TWCs, and 42 mg/mi for aged TWCs (Prigent and de Soete 1992). These are lower than their previously reported values by factors of approximately 4, 3, and 7, respectively.

F. Summary of the data sources

- All the emission factors originate from testing done on five cars using European test cycles. Fuel sulfur content for these tests was unspecified.

- The new and aged TWC emission factors are based 90% 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% of the data for the TWC emission factors came from two studies and three more cars, all tested on European driving cycles only.

- The non-catalyst emission factors were derived from four cars.

- 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 TWCs.

APPENDIX B

NVFEL TESTING PROGRAM

SUMMARY TABLE OF PRELIMINARY NVFEL TESTING RESULTS.

Nitrous oxide emissions are shown in grams per mile for the composite FTP cycle. CAAB is the Clean Air Act Baseline Fuel, which contained 285 ppm sulfur. Indolene contained 24 ppm sulfur.
PV = passenger vehicle
MV = mini-van
PU = pickup truck
SUV = sport utility vehicle

Vehicle Type	Odometer (miles)	Emissions Control	CAAB Fuel		Indolene Fuel	
			75° F A/C Off	95° F A/C On	75° F A/C Off	95° F A/C On
PV	75698	Tier 0	0.018	0.053		
MV	39539	Tier 1	0.046	0.067		
PV	48690	Tier 1	0.059	0.124		
MV	23914	Tier 1	0.038	0.033		
MV	27491	Tier 1	0.028	0.049		
PV	47461	Tier 1	0.027	0.035		
PV	38766	Tier 1	0.036	0.054		0.039
PV	75083	Tier 1	0.042	0.052		
PV	24086	Tier 1	0.024	0.081		
MV	23838	Tier 1	0.027	0.029		
PU	26262	Tier 1	0.227	0.203	0.115	
PU	41549	Tier 1	0.167	0.145		
PU	20585	Tier 1	0.082	0.082		
PU	16319	Tier 1	0.087	0.102		
PU	19251	Tier 1		0.080		
MV	32818	Tier 1		0.043		
PV	28935	Tier 1		0.033		
PV	41896	Tier 1		0.046		
SUV	20949	Tier 1		0.126	0.063	
PV	4959	LEV	**	**	**	**
MV	169311 *	LEV	**	**	**	**
PV	5038 *	LEV			**	**
MV	5038 *	LEV	**	**	**	**

* Catalyts bench-aged to 100,000 miles.
** Data not shown here, but included in averages.

ANALYSES OF THE FUELS USED IN NVFEL TESTING

Tested at NVFEL
CAAB Dispen. #2: tests conducted between 6/4 and 6/15/98
tank #21 cert (Indolene): tests conducted between 2/26 and 3/16/93

Test Code	Test method	CAAB	Indolene	UNITS
552	MTBE by OFID	0.099	0	Oxy Percent
562	ETBE by OFID	0	0	Oxy Percent
534	Ethanol by OFID	0	0	Oxy Percent
572	TAME by OFID	0	0	Oxy Percent
421	Sulfur in Gasoline by ASTM D 2622	285	24	Parts Per Million
62	Vapor Pressure by Appendix E Method 3	7.44	9.02	PSIA
65	Percent Evaporated at 200 Degrees F	39.5	36.4	Volume Percent
66	Percent Evaporated at 300 Degrees F	80.5	88.3	Volume Percent
48	Aromatics in Gasoline MSD D5769	38.261	49.437	Volume Percent
49	Olefins in by FIA D-1319-93	8.524	1.053	Volume Percent
64	Benzene in Gasoline by ASTM D 3606	1.22	0.2409	Volume Percent
46	Aromatics by FIA D-1319-93	30.6	30.1	Volume Percent
69	Specific Gravity @ 60 Degrees F	0.75352	0.74397	60/60F
692	Degrees API	56.28	58.69	Degrees API
691	Density @ 60 deg F	0.75278	0.74324	g/cm-03 @ 60 deg F
101	D 86 Initial Boiling Point	98.8	92.8	Degrees F
110	10 Percent	139.69	132.39	Degrees F
150	50 Percent	222.6	219.8	Degrees F
190	90 Percent	338.89	315.79	Degrees F
200	End Point	413.89	381.7	Degrees F
201	Residue	1.89	1	mL
202	Total Recovery	96.8	97.69	mL
203	Loss	1.29	1.29	mL
592	Volume Percent Oxygenates by MSD	0.55	0	Volume Percent
541	Methanol by MSD (Screen)	0	0	Volume Percent
591	Weight Percent Oxygen by MSD	0.1	0	Weight Percent
543	Methanol by OFID	0	0	Volume Percent
585	t-Butanol by OFID	0	0	Volume Percent
589	Isobutanol by OFID	0	0	Volume Percent
5802	n-Butanol by OFID	0	0	Volume Percent
593	Volume Percent Oxygenates by OFID	0.55	0	Volume Percent
59	Weight Percent Oxygen by OFID	0.09	0	Weight Percent
30	Lead in Gasoline by ASTM D 3237	0.001	na	Grams Pb per Gallon
32	Weight Fractioin Carbon ASTM D 3343-95	0.8673	0.8661	Weight Fraction
991	Phosphorus in Gasoline by ASTM D 3231	0	na	Grams per Gallon
73	Net Heat of Combustion ASTM D 3338-92	18428	na	BTU per Pound
221	Motor Octane	82.59	na	Motor Octane Number
220	Research Octane	92.09	na	Research Octane Number
218	Sensitivity	9.5	na	RON-MON

na = not analyzed

WHY WE BELIEVE THE NITROUS OXIDE DIFFERENCES BETWEEN THE TWO FUELS ARE DUE TO DIFFERENCES IN SULFUR CONTENT

In the NVFEL testing program, emissions of nitrous oxide average 2.5 times higher using CAAB fuel than using Indolene. There are many differences between these two fuels, as the previous table shows. In this section, we make our case—suggestive, rather than conclusive—that the difference in nitrous oxide emissions is primarily due to the difference in the sulfur content of the fuels.

The argument may be summarized that TWCs emit nitrous oxide when they are performing less efficiently than normal (e.g., at lower than normal operating temperatures or after aging), when they also emit more NO_X. Sulfur decreases the efficiency of TWCs and increases NO_X emissions. Therefore, sulfur is also likely to increase nitrous oxide emissions. Of the differences between CAAB fuel and Indolene, modeling shows that only sulfur accounts for the increased NO_X emissions of CAAB fuel. Therefore, that same difference probably also accounts for the increased nitrous oxide emissions.

We know of no published testing on the effects of sulfur content or other fuel parameters on nitrous oxide emissions. However, existing data suggest that for cars equipped with TWCs, conditions that enhance NO_X emissions (i.e., decrease the effectiveness of the catalyst) also enhance nitrous oxide emissions. For example:

1. Nitrous oxide emissions are maximal at around the light-off temperature of catalysts, when NO_X conversion is suboptimal (Prigent et al. 1991).

2. For the same cars equipped with TWCs, tuneups decrease both NO_X and nitrous oxide emissions (Smith and Carey 1982).

3. Running the FTP at a lower than normal temperature in two cases increased NO_X emissions and in two cases decreased them. In all cases the change in nitrous oxide emissions was in the same direction as the change in NO_X emissions (Braddock 1981).

Sulfur has been shown to decrease the effectiveness of NO_X conversion (Lindhjem 1995, Monroe et al. 1991).

Also, we have received an email from Matthias Tappe of the German Federal Environmental Agency - Environment and Traffic as follows: "Regarding the sulfur content your idea that the sulfur level in gasoline influences the N2O emissions has been confirmed by industry data available to us."

EPA's Complex Model shows that the sulfur difference between CAAB and Indolene is the only difference that strongly affects NO_X emissions

EPA's Complex Model was developed in conjunction with petroleum refiners and gasoline formulators to provide guidance as to how emissions would change when various gasoline components were altered. The model predicts that our CAAB fuel will emit 13% more NO_X than our Indolene fuel. The first column in the following table lists the components used by the Complex Model that are different between CAAB and Indolene. The second column shows the percentage change in NO_X emissions that results when the component in the first column is changed from its value in Indolene to its value in CAAB. The third column shows the percentage change in NO_X emissions when all components except the one in the first column are changed from their Indolene to their CAAB values.

Parameter	Percentage change in NO_X emissions from Indolene when:	
	Only this parameter is changed to its CAAB value	All parameters except this one are changed to their CAAB values
All parameters	13	0
MTBE	0	13
Sulfur	12	1
RVP	0	13
E200	0	13
E300	0	12
Aromatics	0	13
Olefins	1	12
Benzene	0	13

Sulfur is the parameter of greatest importance in diminishing the catalytic reduction of NO_X, and, therefore, we suspect, also the parameter of greatest importance in enhancing the production of nitrous oxide.

APPENDIX C

ASSUMED FUEL ECONOMIES WHOSE RATIOS WERE USED TO GENERATE EMISSION FACTORS FOR VEHICLES FOR WHICH THERE WERE NO DATA

The following of fuel economies and carbon dioxide emission factors are from Tables 1-27 through 1-33 of the *IPCC Guidelines*. The source of these tables is Weaver and Chan (1996). Chan (1998) said that their source for fuel economies was MOBILE5 reduced by 15%. The use of fuel-consumption ratios to determine emission factors should be considered a temporary measure only, to be replaced as soon as real data are available. In calculating emission factors, we used the ratios of carbon dioxide emission factors, rather than of fuel economies, because they were listed with more significant digits. The two are equivalent within rounding error, as is shown by the third column of the table below, which was obtained by multiplying the first two columns together.

Vehicle type and control technology	**Fuel Ec (km/L)**	**CO2 (g/km)**	**Test CO2*Fuel Ec (g/L)**
Gasoline Passenger Cars			
Control Technology			
Low Emission Vehicles*	8.5	280	2380
Tier 1	8.3	285	2366
Tier 0	8	298	2384
Oxidation Catalyst	6.2	383	2375
Non-Catalyst	4.5	531	2390
Uncontrolled	4.7	506	2378
* Applicable to California VMT only			
Gasoline Light-Duty Trucks			
Control Technology			
Low Emission Vehicles*	6	396	2376
Tier 1	6	396	2376
Tier 0	4.8	498	2390
Oxidation Catalyst	4.8	498	2390
Non-Catalyst	4	601	2404
Uncontrolled	4.1	579	2374
* Applicable to California VMT only			
Gasoline Heavy-Duty Vehicles			
Control Technology			
Tier 0	2.3	1017	2339
Oxidation Catalyst	2.3	1036	2383
Non-catalyst	1.8	1320	2376
Uncontrolled	1.8	1320	2376
Diesel Passenger Cars			
Control Technology			
Advanced	10	237	2370
Moderate	9.6	248	2381
Uncontrolled	7.5	319	2393
Diesel Light Trucks			
Control Technology			

Vehicle type and control technology	Fuel Ec (km/L)	CO2 (g/km)	Test CO2*Fuel Ec (g/L)
Advanced	7.2	330	2376
Moderate	7.2	331	2383
Uncontrolled	5.7	415	2366
Diesel Heavy-Duty Vehicles			
Control Technology			
Advanced	2.4	987	2369
Moderate	2.4	1011	2426
Uncontrolled	2.2	1097	2413
Motorcycles			
Control Technology			
Non-Catalyst Control	10.8	219	2365
Uncontrolled	8.9	266	2367