

Technical Report

Determination of a Range of Concern
for Mobile Source Emissions
of Formaldehyde
Based Only on its Toxicological Properties

by

Penny M. Carey

July 1983

NOTICE

Technical reports do not necessarily represent final EPA decisions or positions. They are 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.

U. S. Environmental Protection Agency
Office of Air, Noise and Radiation
Office of Mobile Sources
Emission Control Technology Division
Technical Support Staff
2565 Plymouth Road
Ann Arbor, Michigan 48105

Summary

This paper describes an effort by the Emission Control Technology Division of the EPA to suggest a range of concern for formaldehyde (HCHO) emissions from mobile sources. As defined in this report, the lower value of the range will be the lowest level at which there is some suggestion of adverse physiological effects. The upper level of the range of concern is that pollutant concentration above which the studies show that the pollutant causes so great a health hazard as to strongly suggest it be avoided. The region between these limits will be termed the "ambient air range of concern", indicating the range of adverse physiological effects caused by exposure to various concentrations of the pollutant. This range is also expressed in terms of a vehicle emission range of concern to show what levels of vehicle emissions would create ambient concentrations within the ambient air range of concern.

In light of the action called for in section 202(a)(4) of the Clean Air Act (CAA) (1)* and due to a concern within industry as to what emission levels will be used as the basis for the

*Numbers in parentheses denote references listed at end of report.

evaluation of current and future technologies, a methodology was developed prior to this paper for bracketing a range of concern for various unregulated pollutants (2). This paper coordinates the efforts from two EPA contracts in order to use that methodology specifically for an evaluation of formaldehyde.

Mathematical models were previously designed for various scenarios for which mobile sources are the overwhelming contributor to the exposure (such as garages, roadway tunnels, expressways, and street canyons). These models were used to calculate the ambient air concentrations resulting from various mobile source formaldehyde emission factors (mg/mile or mg/minute) (3). These models were also used to convert the ambient air range of concern to corresponding vehicle emission ranges of concern for the various exposure scenarios.

In conjunction with this, a formaldehyde health effects literature search was conducted by Midwest Research Institute under contract to EPA to aid in the determination of the suggested range of concern (4). The literature review focused on the toxicological (i.e., noncarcinogenic) properties of formaldehyde rather than on its carcinogenicity

to humans. The consideration of the potential carcinogenicity of formaldehyde is important but beyond the scope of this report. Some of the typical toxicological health effects noted were eye, nose, throat and respiratory tract irritation of various degrees, depending on exposure.

The results of the Midwest analysis suggest a range of concern for ambient formaldehyde concentrations of 0.03 mg/m to 1.0 mg/m³ (0.02 ppm to 0.8 ppm). Using the mathematical models developed for the roadway scenarios, this range of concern corresponds to motor vehicle emissions ranging from 10.5-350.1 mg/mile for a severe roadway tunnel situation to 714.3-23,809 mg/mile for a typical street canyon situation. For garage scenarios, the formaldehyde range of concern corresponds to motor vehicle emissions ranging from 0.4-15 mg/minute for a severe personal garage situation to 7.7-256 mg/minute for a typical parking garage situation. These ranges of vehicle emissions corresponding to the range of concern are then compared to fleet average emissions to determine if the fleet average emissions fall below, within or above the range of concern for the various scenarios. A summary of selected results can be found in Table S-I.

Table S-I

Summary of Selected Results*Severe Case

<u>Fleet Conditions</u>	<u>Ambient Air Scenario (Severe Case)</u>	<u>Fleet Average Emissions (mg/mile)</u>	<u>Emissions (mg/mile) Corresponding to Range of Concern</u>	<u>Relation of Fleet Average Emissions to Range of Concern</u>
CY 1978 Fleet (includes 25% malfunction)	Roadway Tunnel	25.41	10.5 - 350.1	Within
	Expressway	36.64	59.3 - 1976	Below
	Street Canyon	27.24	106.4 - 3546	Below
25% of CY 1978 Fleet Composed of Catalyst- Equipped Methanol-Fueled Vehicles (25% malfunction of both current fleet & methanol- fueled vehicles)	Roadway Tunnel	27.11	10.5 - 350.1	Within
	Expressway	36.44	59.3 - 1976	Below
	Street Canyon	31.55	106.4 - 3546	Below
100% of Fleet Methanol- Fueled, 25% Malfunction	Roadway Tunnel	32.21	10.5 - 350.1	Within
	Expressway	35.83	59.3 - 1976	Below
	Street Canyon	44.50	106.4 - 3546	Below

*Garage scenarios are not included in this table due to the preliminary nature of the test data. Refer to text of paper for discussion and results for the garage scenarios.

The term "CY 1978" as used in this report and given in Table S-I is defined as a calendar year (CY) 1978 fleet composed of gasoline- and diesel-fueled vehicles. Fleet average emissions for the CY 1978 fleet were calculated using vehicle miles traveled (VMT) fractions representative of a 1978 fleet together with available emission factor data for a variety of different model year gasoline- and diesel-fueled vehicles. Fleet average emissions for those conditions for which methanol-fueled vehicles are introduced into the fleet were also calculated using VMT fractions representative of a 1978 fleet and available emission factor data. Use of the 1978 VMT fractions results in the percentage of catalyst-equipped light-duty vehicles being that present in 1978 rather than a later year when more catalyst-equipped vehicles are on the road.

Referring to Table S-I, based on the available data, the estimated CY 1978 fleet emission factors are below the ranges of concern for the street canyon and expressway scenarios, and within the range of concern for the severe roadway tunnel scenarios. Similar results are obtained if 25 percent of the CY 1978 fleet is replaced with catalyst-equipped, methanol-fueled vehicles meeting current HC, CO and NOx emissions standards.

The "severe case" methanol fleet situation was determined to be that in which 100% of the fleet is methanol-fueled and catalyst-equipped (75% properly functioning, 25% malfunctioning). Due to the limited data, the malfunction chosen for the light-duty methanol-fueled vehicles was removal of the catalyst. Since data were available for a heavy-duty methanol-fueled engine operated with the partial failure of a catalyst, this malfunction was chosen for the heavy-duty engines. For the "severe case" methanol fleet situation, the fleet emission factors are within the range of concern for the roadway tunnel, but fall below the ranges of concern for the street canyon and expressway scenarios.

Based on tests conducted with light-duty gasoline- and diesel-fueled vehicles, parking and personal garage exposures, under severe conditions, would fall within, but not above the range of concern. With catalyst-equipped, methanol-fueled vehicles, parking and personal garage exposures would fall below the range of concern, based on the limited number of tests that have been run. The fact that idle formaldehyde emissions from the single methanol-fueled vehicle tested were lower than those from the gasoline-fueled vehicles tested suggests the small sample size may be producing misleading results.

It must be stressed that the range of concern for formaldehyde suggested in this report is based on its toxicological properties and not its potential carcinogenicity. In addition, this report does not consider the photochemical reactivity of formaldehyde; it is known that formaldehyde has relatively high photochemical reactivity. Consideration of the carcinogenicity and atmospheric photochemical reactions of formaldehyde and its end products is important but beyond the scope of this report.

I. Introduction

Aldehydes are a class of partially oxidized hydrocarbons emitted from many sources, including mobile sources. Formaldehyde, the most prevalent aldehyde in vehicle exhaust, is currently unregulated from mobile sources. Formaldehyde in vehicle exhaust is formed by the incomplete combustion (partial oxidation) of the fuel.

Due to its toxic properties, characteristically pungent odor and photochemical reactivity, tests have been conducted to characterize formaldehyde emissions as a function of driving cycle, fuel and emission control system. The results of these emissions tests along with health effects data, as summarized later in this report, are used to suggest the conditions under which formaldehyde emissions could be of concern with respect to public health and welfare.

In addition to examining formaldehyde emissions from diesel- and gasoline-fueled vehicles, this report examines formaldehyde emissions from methanol-fueled vehicles. This was done because of the potential for increased use of methanol as an automotive fuel. The Clean Air Act requires EPA to evaluate the health risks of new vehicle technologies.

In the interest of establishing a range of concern for levels of formaldehyde in motor vehicle exhaust, Midwest Research Institute (MRI) compiled information on the noncarcinogenic health effects of formaldehyde at different concentrations (4). The results of that work form the basis for the range of concern suggested later in this report.

This report does not consider the photochemical reactivity of formaldehyde; it is known that formaldehyde has relatively high photochemical reactivity. Consideration of the atmospheric photochemical reactions of formaldehyde and its end products is beyond the scope of this particular report. The methodology presented in this paper was developed for analysis of an unregulated pollutant regarding only its toxicological properties. It has not been applied to the evaluation of any carcinogenic properties a pollutant might possess. Therefore, this report also does not consider the potential carcinogenicity of formaldehyde. Some animal tests have indicated that formaldehyde may cause an increased incidence of squamous cell (nasal) cancer in rats. The federal government is currently developing a general policy for use by federal agencies in regulating carcinogens. It is expected that any needed regulations for mobile source emissions of formaldehyde due to its potential carcinogenicity would be handled under the general policy being developed.

II. General Information on Formaldehyde

Formaldehyde (HCHO) is a colorless gas with a characteristically pungent odor. It is highly irritating to the exposed membranes of the eyes, nose and upper respiratory tract. Formaldehyde is the most common and important aldehyde emitted into the air.

Several billion pounds of formaldehyde are produced commercially each year in the United States (7). Partially because of formaldehyde's antiseptic properties, it is used in the medical, brewing and agricultural industries. About half the formaldehyde produced is used in the preparation of urea-formaldehyde and phenol-formaldehyde resins. These resins, in turn, are used in the production of plywood, particleboard, foam insulation, and a wide variety of molded or extruded plastic items.

Under certain conditions, formaldehyde can be released into the environment over a prolonged period from resinous products. These products include urea-formaldehyde foam insulation, particle board and some plywoods. Additional sources of formaldehyde include automotive exhaust, cigarette smoke, incinerators and photochemical generation in the ambient air.

Formaldehyde is known to be a component of photochemical smog formation. Photochemical smog is a form of air pollution which arises from the reactions of oxides of nitrogen and hydrocarbon compounds in the presence of sunlight. Formaldehyde can be photooxidized with a nitrogen oxide mixture in air to yield ozone, which is also toxic. Smog often results in eye and throat irritation, odor, plant damage and decreased visibility. Formaldehyde may account for a large fraction of the eye irritation associated with photochemical air pollution. As mentioned previously, the formation or destruction of formaldehyde by photochemical reactions is an important consideration but beyond the scope of this report. This report will consider only that formaldehyde directly emitted from vehicles.

In an automotive system, formaldehyde is formed by the incomplete combustion (partial oxidation) of the fuel. Formaldehyde emissions, in general, have been shown to decrease when a catalyst is used for emission control. Control of HC and CO emissions brings about a corresponding reduction in formaldehyde emissions for most emission control systems.

III. Legislative Background

When the Clean Air Act was amended in August 1977, the additions included sections 202(a)(4) and 206(a)(3) which deal with mobile source emissions of hazardous pollutants from vehicles manufactured after 1978. These sections are stated below:

202 (a)

"(4) (A) Effective with respect to vehicles and engines manufactured after model year 1978, no emission control device, system or element of design shall be used in a new motor vehicle or new motor vehicle engine for purposes of complying with standards prescribed under this subsection if such device, system, or element of design will cause or contribute to an unreasonable risk to public health, welfare, or safety in its operation or function.

(B) In determining whether an unreasonable risk exists under subparagraph (A), the Administrator shall consider, among other factors, (i) whether and to what extent the use of any device, system, or element of design causes, increases, reduces, or eliminates emissions of any

unregulated pollutants; (ii) available methods for reducing or eliminating any risk to public health, welfare, or safety which may be associated with the use of such devices, systems, or elements of design which may be used to conform to standards prescribed under this subsection without causing or contributing to such unreasonable risk. The Administrator shall include in the consideration required by this paragraph all relevant information developed pursuant to section 214."

206 (a)

"(3) (A) A certificate of conformity may be issued under this section only if the Administrator determines that the manufacturer (or in the case of a vehicle or engine for import, any person) has established to the satisfaction of the Administrator that any emission control device, system, or element of design installed on, or incorporated in, such vehicle or engine conforms to applicable requirements of section 202(a)(4).

(B) The Administrator may conduct such tests and may require the manufacturer (or any such person) to conduct such tests and provide such information as is necessary to carry out subparagraph (A) of this paragraph. Such

requirements shall include a requirement for prompt reporting of the emission of any unregulated pollutant from a system device or element of design if such pollutant was not emitted, or was emitted in significantly lesser amounts, from the vehicle or engine without the use of the system, device, or element of design."

Prior to these amendments, EPA's guidance to the manufacturers regarding hazardous unregulated pollutants were contained in the Code of Federal Regulations, Title 40, section 86.078-5b. This subsection is stated as follows:

"Any system installed on or incorporated in a new motor vehicle (or new motor vehicle engine) to enable such vehicle (or engine) to conform to standards imposed by this subpart:

(i) Shall not in its operation or function cause the emissions into the ambient air of any noxious or toxic substance that would not be emitted in the operation of such vehicle (or engine) without such system, except as specifically permitted by regulation; and

(ii) Shall not in its operation, function, or malfunction result in any unsafe condition endangering the motor vehicle, its occupants, or persons, or property in close proximity to the vehicle.

(2) Every manufacturer of new motor vehicles (or new motor vehicle engines) subject to any of the standards imposed by this subpart shall, prior to taking any of the action specified in section 203 (a)(1) of the Act, test or cause to be tested motor vehicles (or motor vehicle engines) in accordance with good engineering practice to ascertain that such test vehicles (or test engines) will meet the requirements of this section for the useful life of the vehicle (or engine)."

Before certification can be granted for new motor vehicles, manufacturers are required to submit a statement, as well as data (if requested by the Administrator), which will show that the technology for which certification is requested complies with the standards set forth in section 86.078-5(b). This statement is made in section 86.078-23(d).

The EPA issued an Advisory Circular (AC) (5) in June 1978, to aid the manufacturers in complying with section 202 (a)(4). Manufacturers were asked to continue providing statements showing that their technologies did comply with the vehicle emission standards and also will not

contribute to an unreasonable risk to public health. Another Advisory Circular (6) was issued in November of that year continuing these procedures for 1980 and later model years. At that time, EPA began work to develop and implement a methodology which would provide a preliminary assessment of potential mobile source unregulated pollutant hazards in order to assist the manufacturers in deciding which, if any, unregulated pollutants are of particular concern.

Up to this time, several preliminary assessments have been made covering sulfuric acid, hydrogen cyanide and ammonia. In each of these cases, the preliminary assessment found no reason for suspecting a public health problem from the current fleet emissions of these pollutants, and recommended that further monitoring may be appropriate to be sure that new vehicle/emission control system configurations did not result in greatly increased emissions.

IV. Methodology Overview

Along with the previously mentioned activities, EPA, with the input from several interested parties, has developed a methodology which is one possible approach to implementing section 202 (a) (4) of the CAA. This approach is explained in detail in EPA report number EPA/AA/CTAB/PA/81-2, "An Approach for Determining Levels of Concern for Unregulated Toxic Compounds from Mobile Sources" (2). Only a brief summary of this method will be presented in this report.

Under contract to EPA, Southwest Research Institute (SwRI) and Midwest Research Institute (MRI) have provided valuable information for this effort. SwRI developed or modified mathematical models for predicting ambient air concentrations of mobile source pollutants for a variety of exposure scenarios including enclosed spaces, street canyons, and expressways. Once vehicle emission factors for various vehicle categories have been determined for a particular pollutant, these models can then be used to calculate corresponding ambient air values for both severe and typical exposure situations for each scenario.

Health effects literature searches have been conducted by MRI in an attempt to aid EPA in suggesting a range of concern for

various selected pollutants. With adequate information, the limits for this range can be chosen. The upper level of the range will be that pollutant concentration above which the studies show that the pollutant causes so great a health risk as to strongly suggest it be avoided. The lower value of the range will be the lowest level at which there is evidence of adverse physiological effects. The region between these limits will be termed the "ambient air range of concern", indicating scattered data points providing evidence of adverse or physiological effects caused by exposure to various concentrations of formaldehyde. Using the ambient air vs. emission factor model developed earlier, the ambient air range of concern can be expressed in terms of a vehicle emission range of concern for each scenario. Any technology emitting a pollutant falling within the range of concern should be subject to closer scrutiny. Technologies with emission levels which fall above the highest value of the range should be considered "high risk" with respect to human health.

For the purpose of this report, this particular methodology has been used to develop a range of concern specifically for motor vehicle emissions of formaldehyde.

V. Vehicle Emissions of Formaldehyde

Formaldehyde exhaust emissions have been measured for a variety of vehicle types. The EPA recommended procedure for this measurement is described in an EPA report entitled, "Analytical Procedures for Characterizing Unregulated Pollutant Emissions from Motor Vehicles" (8). The recommended procedure, commonly referred to as the 2,4 dinitrophenylhydrazine (DNPH) procedure, includes use of a gas chromatograph (GC) and flame ionization detector (FID) for analysis of formaldehyde and other individual aldehydes. The DNPH procedure was used to obtain all the formaldehyde data in this report. In some cases a high pressure liquid chromatograph (HPLC) was used rather than a GC-FID for analysis of formaldehyde. The use of two different analytical techniques should not significantly affect the results.

Formaldehyde emission factors for various vehicle types were collected from several available sources and are listed in Table I. Emission factor data were obtained for a variety of different model year gasoline- and diesel-fueled vehicles.

Table I

Gasoline- and Diesel-Fueled Vehicles-No MalfunctionFormaldehyde Emission Factors^a

<u>Vehicle Category</u>	<u>Formaldehyde (mg/mile)</u>		
	<u>FTP</u>	<u>Hot FTP</u>	<u>HFET</u>
Light-Duty Diesel Vehicles	21.21	15.24	12.44
Light-Duty Diesel Trucks ^b	21.21	15.24	12.44
Light-Duty Gasoline Vehicles			
Non-Catalyst; no air pump	48.79	47.83	34.97
Non-Catalyst; air pump	15.62	11.39	17.47
Oxidation Catalyst;no air pump	2.28	1.51	1.36
Oxidation Catalyst;air pump	4.65	4.25	1.37
3-way Catalyst; no air pump	0.35	0.11	0.11
3-way Plus Ox. Cat.; air pump	2.57	2.91	3.04
Light-Duty Gasoline Trucks ^c			
Non-Catalyst; air pump	15.62	11.39	17.47
Catalyst; no air pump	2.28	1.51	1.36
	<u>Transient FTP</u>	<u>Hot Trans. FTP</u>	<u>7 Mode Steady State</u>
Heavy-Duty Diesel Trucks ^d	36.75	35.31	123.85
Heavy-Duty Gasoline Trucks ^d	62.48 ^e	45.56 ^e	174.38

a References 9, 10, 11, 12, 13, 14, 15, 16.

b Due to a lack of sufficient data, these values are assumed to be the same as those given for light-duty Diesel vehicles.

c These values are assumed to be the same as those given for light-duty gasoline vehicles.

d Heavy-duty engine data expressed as g/kW-hr converted to mg/mile using road fuel consumption test data from other heavy-duty engines.

e Due to a lack of sufficient data, these values are assumed to be the same as those given for non catalyst, light-duty gasoline vehicles, with an air pump, adjusted by a factor of 4 for approximate differences in fuel consumption.

Emission factors for low mileage light-duty vehicles were compiled for the Federal Test Procedure (FTP), hot start FTP, and Highway Fuel Economy Test (HFET) driving schedules. Emission factors for the heavy-duty vehicles were compiled for the transient FTP, hot FTP and 7 mode steady state driving schedules.

The available data for the light-duty gasoline-fueled vehicles list emission levels from both unmodified (i. e., properly tuned) and malfunctioning vehicles. The malfunction modes evaluated for the non-catalyst and catalyst-equipped vehicles were 12% misfire and disconnected EGR and/or O₂ sensor, respectively. These malfunction modes resulted in the greatest increase in formaldehyde emissions. Average malfunction emissions for each light-duty gasoline vehicle category are given in Table II. These malfunction emissions will be used when calculating fleet average emission factors as discussed later in this report.

The emissions found for the malfunction modes are especially important to this effort due to the fact that formaldehyde emissions tend to increase under malfunction conditions. Maximum emission rates have been listed below for three vehicle categories.

Table II

Average Malfunction Emissions
Light-Duty Gasoline Vehicles*

<u>Vehicle Category</u>	<u>Formaldehyde (mg/mile)</u>		
	<u>FTP</u>	Hot Start <u>FTP</u>	<u>HFET</u>
Non-cat.; no air pump	208.80	237.49	199.34
Non-cat.; air pump	121.06	101.95	242.22
Ox. cat.; no air pump	7.03	4.04	4.38
Ox. cat.; air pump	8.08	7.77	9.01
3-way cat.; no air pump	1.58	1.30	0.56
3-way plus ox. cat.; air pump	1.42	1.43	0.48

*References 12, 13, 14, 15.

Maximum Reported Formaldehyde Emission Rates a,b
(Highest Values Reported From Any Source On Any Single Test)

<u>Light-Duty Gasoline Vehicle Category</u>	<u>FTP</u>	<u>Mg/Mile Hot FTP</u>	<u>HFET</u>
Non-catalyst	340.38	369.30	338.29
Oxidation Catalyst	27.37	23.99	54.40
3-Way Catalyst	39.80	19.50	6.87

a References 12, 18, 19.

b Formaldehyde emissions from in-use vehicles and/or vehicles operating under malfunction modes.

Data from in-use vehicles operating with or without malfunctions were also examined and, where appropriate, included in the above table. The maximum reported emissions for the non-catalyst-equipped vehicles are higher than those of the other two categories, and they are also much higher than any of the vehicle categories listed in Table I.

Table III lists the formaldehyde emissions found for light- and heavy-duty methanol-fueled vehicles. The heavy-duty numbers are based on tests of only one engine (M.A.N. 100% methanol/spark ignition engine). Formaldehyde emissions from this heavy-duty engine were lower than those from the light-duty vehicles when the transient FTP engine dynamometer cycle was used. This appears to be an anomaly; the heavy-duty emission factors given in Table III should be updated as additional tests are run. The

Table III

Methanol-Fueled Vehicles
Formaldehyde Emission Factors ^a

<u>Vehicle Category</u>	<u>Formaldehyde (mg/mile)</u>		
	<u>FTP</u>	<u>Hot FTP</u>	<u>HFET</u>
Light-Duty Methanol Vehicles			
Unmodified (no malfunction) Oxidation or 3-Way Catalysts	21.67	6.79	2.20
Malfunction Non-catalyst (catalyst- equipped vehicles tested without catalysts)	123.55	116.98	83.25
	<u>Transient FTP</u>	<u>Hot Trans. FTP</u>	<u>7 Mode Steady State</u>
Heavy-Duty Methanol Engines ^b			
Unmodified (no malfunction) Oxidation Catalyst	3.09	0.00	158.78
Malfunction Partial Failure of Oxidation Catalyst	9.27 ^c	0.00	476.27

^aReferences 20,21,22,23.

^bHeavy-duty engine data expressed as mg/kW-hr converted to mg/mile using road fuel consumption test data from other heavy-duty engines.

^cSince data on the transient cycle are not available for this malfunction, this emission factor was obtained by applying the three fold increase found for the 7 mode data to the data on the transient cycle obtained with the oxidation catalyst (3.09 x 3 = 9.27 mg/mile).

formaldehyde emission factors given in Table III come from vehicles or engines using 100% methanol fuel; formaldehyde emissions from vehicles or engines using a gasoline-methanol or diesel fuel-methanol mixture are not considered in this paper. It should be noted that the formaldehyde emission factors for the light-duty methanol-fueled vehicles are based on tests of only a few vehicles. Formaldehyde emissions from these vehicles appear to be well controlled. Additional light-duty methanol-fueled vehicles should be tested to confirm these findings.

It is assumed that current HC, CO, and NO_x emission standards will apply to future light-duty methanol-fueled vehicles, and that these vehicles will require catalysts to meet these emission standards. The hydrocarbon (HC) standard (e.g., 0.41 g/mile for light-duty vehicles on the FTP) would presumably apply only to the HC portion of any unburned alcohol in the exhaust or evaporative emissions. Since HC comprises only 50% of the mass of methanol, the standard to be met for actual methanol emissions would in effect be double that for gasoline (e.g., 0.82 g/mile for light-duty vehicles on the FTP). Only light-duty methanol-fueled vehicles which were equipped with catalysts and which met the existing federal emission standards were used to generate the

data in Table III. Conclusions in this report are therefore not applicable to methanol-fueled vehicles which do not have catalysts or methanol-fueled vehicles whose emissions without malfunction would not meet current standards.

Data for the heavy-duty engine were given in terms of mg/kW-hr. To put these data in terms of mg/mile, they were first converted to g/kg fuel by dividing by the fuel consumption (kg fuel/kW-hr). Then, using fuel economy data from other heavy-duty diesel engines averaging roughly 56 liters/100 km (24), and adjusting for the different energy content of methanol vs. diesel fuel, the corresponding methanol emission factors were calculated. Like the gasoline- and diesel-fueled vehicles, formaldehyde emissions from methanol-fueled vehicles are shown to decrease substantially when a catalyst is used for emission control.

A certain percentage of in-use vehicles typically operate in a less-than-optimum condition, referred to in this report as a malfunction condition. As discussed previously, malfunction data are available for light-duty gasoline-fueled vehicles. For methanol-fueled vehicles, however, data are very limited. The malfunction data used for the light-duty methanol-fueled vehicles were those data obtained when the catalyst-equipped vehicles were tested without catalysts.

Table III shows the dramatic increase in formaldehyde emissions that results with this severe malfunction. The "malfunctioning" (no catalyst) vehicle formaldehyde emissions are roughly an order of magnitude greater than emissions from non-malfunctioning catalyst-equipped vehicles. Limited data are available for a methanol-fueled catalyst-equipped 1981 Ford Escort tested with the air injection and/or EGR disconnected (23). Formaldehyde emissions with both the air injection and EGR disconnected are comparable to the formaldehyde emissions obtained when the vehicle was tested without a catalyst.

Seven mode steady-state data exist for a heavy-duty methanol-fueled engine operated with partial failure of the oxidation catalyst. The data in Table III show formaldehyde emissions to increase roughly three times with the partial failure of the catalyst as compared to the functioning catalyst. Since data on the transient cycle are not available for this malfunction, the three fold increase found for the 7 mode data was applied to the data on the transient cycle obtained with the functioning catalyst. Unfortunately, no data are available for the engine operated without the oxidation catalyst.

The potential effect of these malfunctions on ambient air concentrations of formaldehyde is shown in columns 4 and 5 of Table VII (in Section VI) with 25% of the vehicles assumed to be malfunctioning.

The driving cycles considered in this report and given in Tables I, II, and III were chosen to represent various exposure scenarios. These scenarios and the driving cycles chosen for each scenario will be discussed in the following section (section VI). In addition to the driving cycles given in Tables I, II, and III, available formaldehyde idle emissions data (mg/minute) were used to estimate formaldehyde exposures in garage scenarios. This will also be discussed in section VI.

Fleet Average Emissions

Using the formaldehyde emission factor data presented in Tables I, II, and II, it is possible to calculate fleet average emission factors. The additional information used to make these calculations is listed in Table IV. A fraction of the vehicle miles traveled (VMT) is listed for each vehicle class. These data were derived from information presented in the Pedco Report of 1978 (25), and the EPA report, "Mobile Source Emission Factors: For Low Altitude Areas Only" (26).

Table IV gives the VMT mix for the 1978 fleet. This mix tends to change from year to year with the introduction of new engines and emission control systems, so the fleet average emissions can be updated by modifying the fleet VMT mix data used in the calculations.

An example of this would be the quantity of non-catalyst gasoline VMT relative to the VMT of catalyst-equipped vehicles. Based on Table IV which reflects the makeup of a 1978 fleet, 57% of the total VMT (for light-duty and heavy-duty vehicles) would be from catalyst-equipped vehicles (with or without an air pump) and 24.5% would be from non-catalyst-equipped light-duty vehicles. In later years, the non-catalyst fraction of the total VMT is expected to decrease. As a result, the formaldehyde fleet average emission factors for later years (if based on a total VMT composed of diesel and gasoline-fueled vehicles) are also expected to decrease.

Each vehicle class VMT fraction is multiplied by the corresponding emission factor (EF) for that class, giving a fraction quantity of pollutant emitted from a particular vehicle category in comparison to other vehicle categories in the fleet. The $EF \times VMT$ fractions for each vehicle class are calculated and then summed to obtain a total fleet average.

Table IV

Fraction VMT For 1978 Fleet*

<u>Vehicle Category</u>	<u>Fraction VMT</u>
Light-Duty Diesel Vehicles	0.015
Light-Duty Diesel Trucks	0.002
Light-Duty Gasoline Vehicles	
Non-Catalyst; no air pump	0.147
Non-Catalyst; air pump	0.098
Ox Cat.; no air pump	0.289
Ox Cat.; air pump	0.261
3-Way Cat.; no air pump	0.012
3-Way plus Ox. Cat.; air pump	0.008
Light-Duty Gasoline Trucks	
Non-Catalyst	0.096
Catalyst	0.010
Heavy-Duty Diesel Trucks	0.027
Heavy-Duty Gasoline Trucks	0.035

*References 25 and 26.

The total fleet average then is based on VMT fractions for the 1978 fleet together with emission factor data for a number of different model year vehicles. For formaldehyde emissions from the calendar year (CY) 1978 fleet (composed of gasoline- and diesel-fueled vehicles), the fleet average ranges from 13.63 mg/mile to 18.98 mg/mile, depending on the driving cycle chosen. This takes into account only those vehicle classes listed in Table IV. Of course, as mentioned previously, should any of these categories change, so would the total fleet average.

It is difficult to predict exactly what percentage of vehicle categories will make up the entire fleet at any one time. In order to account for differing proportions of malfunctions and technologies, Table V was devised. Table V presents fleet averages for the CY 1978 fleet and the CY 1978 fleet with 25% of the light-duty vehicles malfunctioning. The latter fleet average is based on the assumption that 25% of the vehicle fleet operates in some malfunction mode (i.e., misfire, disconnected O₂ sensor, etc.) at any given time (17)*. Further work may identify a more accurate percentage.

*Previous reports (38,39) on specific compounds evaluated for Section 202(a)(4) of the Clean Air Act also used 25% malfunction. This percentage is confirmed as a realistic upper bound based on the 1982 EPA Office of Mobile Sources Field Operations Support Division tampering survey results. These results indicate that, for vehicles at the 50,000 mile point in non-I/M areas, the tampering rate is approximately 26% when catalyst removal, disconnected air pump, and habitual misfueling are considered.

Table V

Total Fleet Averages

	<u>Formaldehyde (mg/mile)</u>		
	<u>FTP</u> ^a	<u>Hot FTP</u> ^b	<u>HFET</u> ^c
CY 1978 Fleet (no malfunction)	15.66	13.63	18.98
CY 1978 Fleet (25% malfunction)	27.24	25.41	36.64
25% of CY 1978 Fleet ^d Composed of Catalyst- Equipped Methanol- Fueled Vehicles (no malfunction)	16.87	11.82	17.21
25% of CY 1978 Fleet ^d Composed of Methanol-Fueled Vehicles (both fleet and methanol-fueled vehicles contain 25% malfunction)	31.55	27.11	36.44
100% of Fleet ^d Methanol-Fueled Vehicles (25% malfunction)	44.50	32.21	35.83

^aIncludes LD FTP and HD Transient FTP emission factors.

^bIncludes LD Hot FTP and HD Hot Transient FTP emission factors.

^cIncludes LD HFET and HD 7 Mode Steady State emission factors.

^dBased on a VMT mix of 93.8% LD/6.2% HD.

Table V also presents fleet averages for hypothetical situations in which 25% and 100% of the CY 1978 vehicle fleet is replaced with light- and heavy-duty methanol-fueled vehicles. For the two situations in which 25% of the CY 1978 fleet is replaced with methanol-fueled vehicles, 25% of the CY 1978 light-duty fleet was replaced with light-duty methanol-fueled vehicles and 25% of the CY 1978 heavy-duty fleet was replaced with heavy-duty methanol-fueled vehicles. The introduction of light-duty methanol-fueled vehicles into the CY 1978 fleet is expected to have more effect on formaldehyde emissions than the corresponding introduction of heavy-duty methanol-fueled vehicles because light-duty vehicles are estimated to comprise 93.8% of the fleet.

From examining Table V, it can be seen that substituting 25% of the CY 1978 fleet with light and heavy-duty catalyst-equipped, methanol-fueled vehicles has little impact on resulting fleet average formaldehyde emissions. This occurs in spite of the fact that light-duty catalyst-equipped, methanol-fueled vehicles emit greater quantities of formaldehyde than their gasoline-fueled counterparts. The reason is, by substituting 25% of the CY 1978 fleet with catalyst-equipped, methanol-fueled vehicles, a portion of the non-catalyst-equipped, gasoline-fueled vehicles are in the end result displaced. Non-catalyst-equipped, gasoline-fueled vehicles emit greater quantities of

formaldehyde than catalyst-equipped, methanol-fueled vehicles. In reality, however, methanol-fueled vehicles would be replacing primarily catalyst-equipped vehicles rather than non-catalyst-equipped vehicles. In addition, the number of non-catalyst-equipped vehicles will continue to decrease in the future. These are weaknesses of the simplistic partial replacement of the fleet with methanol-fueled vehicles. Because of these weaknesses, this replacement scenario does not provide an adequate one-to-one comparison with continued current fleet sales.

The compiled fleet averages given in Table V will be used in comparing vehicle emissions to the suggested range(s) of concern. In subsequent steps, these fleet averages will be used to calculate ambient concentrations of formaldehyde for each situation.

VI. Formaldehyde Ambient Air Concentrations

The formaldehyde emission factor information provided in Tables I through V can be used in conjunction with the modeling techniques developed by Southwest Research Institute (SwRI) (3), in order to calculate the ambient air concentrations produced by varying levels of formaldehyde vehicle emissions for different microscale exposure situations. Future work may identify other scenarios which would also be appropriate for the assessment of human exposure to exhaust pollutants, but, for this task, only five exposure scenarios were investigated: personal garages, parking garages, roadway tunnels, street canyons, and urban expressways. Actual locations and receptors representing typical and severe exposure levels were chosen for each of these scenarios. The mathematical models for each different situation were chosen from the literature. No attempt was made to develop new models, although existing models sometimes required modification or use in a new manner to most accurately define the ambient air concentrations. For localized area sources, the literature search for models produced several models that predicted concentrations downwind of area sources, but none that predicted concentrations within the area source itself; therefore, this exposure situation, while possibly important, will not be

considered. Each situation has been considered separately, and, therefore, no cumulative effects have been determined at this point. Reference (3) discusses in detail the reasoning behind using these specified scenarios as well as the information used in the determination of the modeling techniques. It should be noted that another approach would be to use an air quality model for a region as a whole; however, EPA has not used this approach for unregulated emissions, preferring localized situations since they are of greatest concern.

Fleet averages for CY 1978 fleet and various methanol situations, listed in Table VI, were used to estimate the corresponding formaldehyde ambient air concentrations given in Table VII. Table VII presents ambient air concentrations of formaldehyde, as a function of vehicle emissions, for seven ambient situations.

Garage scenarios are not included in the table, but are described in the text because idle emissions are expressed in terms of mg/minute rather than mg/mile, and are available for only a limited number of vehicles.

Table VI

Total Fleet Averages for Various Exposure Situations^a

	<u>Formaldehyde (mg/mile)</u>			
	<u>Roadway tunnel^b</u>		<u>Street Canyon^c</u>	<u>Expressway^d</u>
	<u>Typical</u>	<u>Severe</u>		
CY 1978 Fleet (no malfunction)	18.98	13.63	15.66	18.98
CY 1978 Fleet (25% malfunction)	36.64	25.41	27.24	36.64
25% of CY 1978 Fleet Composed of Catalyst-Equipped Methanol-Fueled Vehicles (no malfunction)	17.21	11.82	16.87	17.21
25% of CY 1978 Fleet Composed of Methanol-Fueled Vehicles (both fleet and methanol- fueled vehicles contain 25% malfunction)	36.44	27.11	31.55	36.44
100% of Fleet Methanol-Fueled Vehicles (25% malfunction)	35.83	32.21	44.50	35.83

^aTotal fleet averages taken from Table V.

^bTotal fleet average for the HFET cycle was chosen to represent the typical case tunnel situation. Total fleet average for the hot start portion of the FTP was chosen to represent the severe case tunnel situation.

^cTotal fleet average for the FTP cycle was chosen to represent both the typical and severe case street canyon situations.

^dTotal fleet average for the HFET cycle was chosen to represent the typical, severe and close proximity expressway situations.

Table VII

Ambient Air Scenarios *
Formaldehyde Concentrations (mg/m³)

	<u>CY 1978 Fleet (no malfunction)</u>	<u>CY 1978 Fleet (25% malfunction)</u>	<u>25% of CY 1978 Fleet Composed of Catalyst- Equipped Methanol-Fueled Vehicles (no malfunction)</u>	<u>25% of CY 1978 Fleet Composed of Methanol- Fueled Vehicles (25% malfunction)</u>	<u>100% of Fleet Methanol- Fueled (25% malfunction)</u>
Roadway Tunnel					
Typical	0.021	0.041	0.019	0.041	0.040
Severe	0.039	0.073	0.034	0.077	0.092
Expressway					
Typical	0.002	0.005	0.002	0.005	0.004
Severe	0.010	0.019	0.009	0.018	0.018
Close Proximity	0.002	0.004	0.002	0.004	0.004
Street Canyon					
Typical	0.001	0.001	0.001	0.001	0.002
Severe	0.004	0.008	0.005	0.009	0.013

*Garage scenarios are not included in this table due to the preliminary nature of the test data. Refer to text of paper for discussion and results for the garage scenarios.

Each scenario is intended to represent a specific type of situation. The typical personal garage situation represents a 30 second vehicle warm-up time and the severe situation simulates a five minute vehicle warm-up time. Both of these cases, of course, take place within a residential garage with the door open, and are intended to correspond to summer and winter conditions, respectively.

The typical parking garage case simulates an above the ground, naturally ventilated garage in which it is assumed that a vehicle spends an equal amount of time on both the parking level and ramp level. The severe case represents an underground garage wherein the exposed population is assumed to be at parking level five (lowest level). It is also assumed that this exposure occurs 20 minutes after a major event in which the parking structure is emptying from an essentially full condition. The initial concentration of formaldehyde is assumed to be low (0.001 mg/m^3).

In order to more closely assess public exposure to formaldehyde in garage situations, idle and very low speed emissions data were collected from six production vehicles (27). The vehicles included a 1970 non-catalyst-equipped vehicle, 1978 and 1980 oxidation catalyst-equipped vehicles, 1981 and 1982 three-way, catalyst-equipped vehicles, and a

1981 diesel vehicle. Idle data were collected to simulate the personal garage situations. A modified version of the New York City Cycle (NYCC) was developed to simulate low speed operation that may be encountered in a typical parking garage. The modified NYCC is 12 minutes in duration, has a maximum speed of 21 miles per hour, an average speed of 2.5 miles per hour, and contains 68 percent idle operation. With the exception of the diesel vehicle, the vehicles were tested unmodified and under malfunction operation. Formaldehyde emissions at idle ranged from 0.00 to 3.43 mg/minute for the unmodified vehicles, and from 0.00 to 3.97 mg/minute for the malfunctioning vehicles. Formaldehyde emissions at low speed ranged from 0.00 to 2.86 mg/minute for the unmodified vehicles and from 0.12 to 1.97 mg/minute for the malfunctioning vehicles. Assuming worst case conditions (idle: 0.00-3.97 mg/minute, low speed: 0.12-2.86 mg/minute), formaldehyde ambient air concentrations for each of the garage situations would be as listed below:

Diesel and Gasoline-Fueled Vehicles
Formaldehyde Ambient Air Concentrations (mg/m³)

<u>Personal Garage</u>		<u>Parking Garage</u>	
<u>Typical</u>	<u>Severe</u>	<u>Typical</u>	<u>Severe</u>
0.000-0.031	0.000-0.266	0.000-0.011	0.007-0.159

Currently, idle emission data for methanol-fueled vehicles are available from one vehicle, a VW Rabbit with a 3-way catalyst (22). Average formaldehyde emissions at idle are 0.26 mg/minute with the catalyst and 17.49 mg/minute without the catalyst. Resulting formaldehyde ambient air concentrations for the methanol-fueled vehicle for each of the garage situations would be as listed below:

Methanol-Fueled VW Rabbit
Formaldehyde Ambient Air Concentrations (mg/m³)

	<u>Personal Garage</u>		<u>Parking Garage</u>	
	<u>Typical</u>	<u>Severe</u>	<u>Typical</u>	<u>Severe</u>
With Catalyst	0.002	0.017	0.001	0.014
Without Catalyst	0.138	1.172	0.068	0.974

Two specific tunnel designs were chosen to estimate the two roadway tunnel cases. A newly designed, two lane roadway tunnel, with moderate traffic flow, is used for the typical condition, while an old design, heavily traveled roadway tunnel is used for the severe condition. The HFET driving cycle, with an average speed of 48.2 mph, was chosen to represent the typical case tunnel scenario. For the severe case tunnel scenario the average speed is 25 mph, so of the data available, the hot start portion of the FTP was chosen as representative.

The street canyon situations are simulated by examining the parameters of two street canyons. The most sensitive parameter in this model appears to be the number of traffic lanes within the canyon. The typical condition is calculated for a two-lane street canyon with a traffic load of 800 vehicles per hour and a sidewalk location of the exposed population. The severe condition is based on a six-lane street canyon with a 2400 vehicles per hour traffic load, and the exposed population is located inside the vehicles. The FTP was chosen to represent the typical and severe street canyon. The FTP, with an average speed of 19.6 mph, simulates urban driving conditions including cold and hot starts and stop and go driving.

Three different cases were considered in order to cover the possible range of exposures in an expressway situation. The typical, on road exposure is based on a four-lane expressway with a traffic load of 1400 vehicles per hour and a westerly wind (perpendicular to roadway) of 1.0 meter per second (representing the most severe wind condition). In this situation, the exposed population is located inside the vehicle. The severe case represents a heavily traveled (3600 vehicles/hour), ten-lane freeway with a 1.0 meter/second westerly wind (perpendicular to roadway), and an in-vehicle

location of the exposed population. The third case is the off-road case which estimates an exposure involving a close proximity to the highway (i.e., living or working close to a heavily traveled freeway). This case is calculated on a short term basis (rush hour) for a distance of 100 meters downwind of the roadway. The HFET was chosen to represent the expressway scenarios.

From examining Table VII, resulting ambient air concentrations of formaldehyde for the roadway scenarios range from 0.001 to 0.092 mg/m³ depending on the scenario and fleet situation chosen. Of the scenarios examined, the severe roadway tunnel results in the highest formaldehyde concentrations. Similarly, of the fleet situations examined, the 100% methanol fleet situation (with 25% malfunction) results in the highest formaldehyde concentrations.

VII. Formaldehyde Health Effects

A literature review concerning the health effects of formaldehyde was performed as an input to the determination of a suggested range of concern for mobile source emissions of this compound (4). The literature review and this report focus on the noncarcinogenic effects of formaldehyde rather than on its carcinogenicity to humans. The latter is an unresolved question of much importance that will be discussed briefly but is beyond the scope of this report.

Interpretation of the health effects of formaldehyde must consider not only the concentration, but also the duration of exposure. The literature review examined both acute and chronic exposure studies of animals and humans. Results of selected acute and chronic exposure studies will be briefly discussed.

Numerous studies have shown that formaldehyde is irritating to the eyes and upper respiratory tract of laboratory animals. The minimal adverse effects seem to be local irritation and subsequent tissue reactions, especially in the pulmonary system. Such adverse effects generally appear at levels at or above 1 mg/m^3 (0.8 ppm), whether the animals

were acutely or chronically exposed. In chronic studies, biochemical and inflammatory changes have been reported in rats exposed for only 8-12 weeks to formaldehyde levels as low as 0.012 mg/m^3 (0.01 ppm) (4).

Formaldehyde is strongly irritating to the human eye, nose, and throat and capable of causing allergic sensitization. Acute human experimental exposure to formaldehyde concentrations of $1.25\text{-}17.3 \text{ mg/m}^3$ (1.00 - 13.8 ppm) results in moderate to severe irritation of the eye, nose, and throat (4). Exposure times ranged from 1.5 minutes (with multiple exposures) to 5 hours. Clear irritation occurs among subjects exposed to formaldehyde concentrations at or above 1.0 mg/m^3 (0.8 ppm). At exposures of approximately 1.0 mg/m^3 for 10 minutes or 5 hours, eye and respiratory tract irritation is slight, odor is perceived, and other effects such as changes in breathing rhythm and alpha-rhythms occur (29,30,31). Slight eye, nose, and throat discomfort occurs at a formaldehyde concentration of 0.3 mg/m^3 (0.24 ppm) when exposed 5 hours (29). The threshold for eye irritation is $0.2\text{-}0.25 \text{ mg/m}^3$ (0.16 - 0.20 ppm) based on a single exposure of 300 seconds (32). The reported odor thresholds range from 0.4 mg/m^3 (0.32 ppm) to roughly 0.05 mg/m^3 (0.04 ppm) for sensitive subjects (33). Mood changes have been reported for subjects exposed to formaldehyde levels as low as 0.0024 mg/m^3 (0.0019 ppm) (34).

Repeated exposure to formaldehyde can cause sensitization in certain individuals (such as people with allergies, asthmatics and others with hyper-reactive airways). Sensitization is an allergic process caused by repeated exposure to certain substances. When exposed to formaldehyde, these sensitized persons may exhibit allergic dermatitis or mild to severe asthmatic reactions. There are indications that some of the sensitized individuals may develop increasingly severe reactions from subsequent exposure to formaldehyde. It is estimated that fewer than 20% but perhaps more than 10% of the general population may be susceptible to formaldehyde and may respond to extremely low levels of formaldehyde (7).

In occupational and residential studies, formaldehyde levels of $0.036 - 4.98 \text{ mg/m}^3$ ($0.029 - 3.98 \text{ ppm}$) have been associated with health effects such as eye, nose and throat irritation, nausea, vomiting, diarrhea, headaches, irritability and skin rashes (35). Case studies in mobile homes which used particle board in the construction predict that 20 percent of the adult population would experience eye irritation at a formaldehyde level of 0.25 mg/m^3 (0.2 ppm) (36). In one group of mobile homes where consumers had health complaints, 90 percent of the formaldehyde concentrations measured were below 0.12 mg/m^3 (0.10 ppm)

(35). Using data available to the Consumer Product Safety Commission as of April 1981, the average level of formaldehyde measured in homes with urea formaldehyde foam insulation was 0.14 mg/m^3 (0.12 ppm) and in homes without urea formaldehyde foam insulation the level was 0.036 mg/m^3 (0.03 ppm) (35). Many (31.6 percent) of the complaint residences with urea formaldehyde foam insulation in which formaldehyde measurements were made had levels of formaldehyde at or below 0.13 mg/m^3 (35).

Nonsmoking and smoking humans have been found to contain formaldehyde in the breath at levels as high as 0.1 mg/m^3 (0.08 ppm), formaldehyde being a normal metabolite and a metabolite of exogenous substances (37). The American Industrial Hygiene Association (AIHA) recommends an outdoor ambient air formaldehyde standard of 0.12 mg/m^3 (0.1 ppm).

Preliminary results of a 24-month chronic-inhalation study sponsored by the Chemical Industry Institute of Toxicology (CIIT) have shown that formaldehyde is a carcinogen in rats. Groups of 120 male and 120 female rats were exposed by inhalation to 0, 2, 6, or 15 ppm formaldehyde vapor 6 hr/day, 5 days/week for 24 months. After 18 months, 36 of 240 rats exposed to a formaldehyde level of 15 ppm were found to have squamous cell carcinomas in the nasal cavities. Similar

tumors were not detected in rats exposed for 18 months to 2 or 6 ppm or in mice exposed to 2, 6, or 15 ppm formaldehyde. The frequency of nasal cancers through the 18-month sacrifice was reported by Swenberg et al.(28). Later, the CIIT reported at the Formaldehyde Symposium on November 20-21, 1980, in Raleigh, N. C., that nasal cancer had been observed in two rats exposed at 6 ppm for 24 months and in two mice exposed at 15 ppm for 24 months. By the end of 24 months, 95 rats exposed to 15 ppm had developed nasal cancers. Although there is no direct evidence of the carcinogenicity of formaldehyde in humans, these results provide evidence that formaldehyde might represent a carcinogenic risk to humans.

VIII. Determination of the Range of Concern and Conclusions

The definition of "range of concern" is that range of exposure concentrations suspected (but not confirmed) to be detrimental to human health. The lower value of this range would be the lowest concentration at which there is some suggestion of adverse physiological effects. The upper value of this range would be that level above which the studies show that the pollutant causes so great a health risk as to strongly suggest it be avoided. Although it would be more appropriate to indicate the exposure time relative to its corresponding concentration which tends to cause adverse health effects, exposure times vary considerably among the available studies. The determination of the range of concern was based primarily on acute human experimental studies since these were thought to most closely simulate the exposure situations examined in this report.

The range of concern for formaldehyde is based on an examination of relevant studies pertaining to noncarcinogenic health effects, primarily acute human experimental studies. Because formaldehyde is a strong irritant of the eyes, nose, and throat and is also capable of causing an allergic sensitization among the exposed population, special emphasis will be given to the levels of formaldehyde found to cause discomfort, where ordinarily this type of effect may not be considered an "adverse" health effect.

The range of concern is suggested to be 0.03 mg/m^3 - 1.0 mg/m^3 (0.02 - 0.8 ppm). The suggested upper level is 1.0 mg/m^3 because of the wide evidence of clear irritant effects among individuals acutely exposed at this level. The suggested lower level is 0.03 because the numerous animal data, if directly extrapolatable to humans, dictates a level of 0.01 - 0.04 mg/m^3 . In addition, a human odor threshold as low as 0.05 mg/m^3 has been reported for sensitive populations. The capability of formaldehyde to affect allergic sensitization cannot be overemphasized as an additional rationale for caution. In relation to the Threshold Limit Value* (TLV) of 3 mg/m^3 for formaldehyde, the lower level is 1/100th the TLV. This lower limit is somewhat conservative considering that formaldehyde levels in homes without urea formaldehyde foam insulation average 0.036 mg/m^3 and that formaldehyde in human breath is as high as 0.1 mg/m^3 .

Between the chosen limits of the range (0.03 - 1.0 mg/m^3), there are scattered data points providing evidence of adverse physiological effects caused by exposure to various concentrations of formaldehyde.

*The Threshold Limit Value, set by the American Council of Governmental Industrial Hygienists, is the recommended maximum time weighted average concentration to which workers can be exposed for an 8-hour work day or 40-hour work week.

The next step in making use of this range of concern is to translate it into terms of automotive emission factors for each public exposure scenario. Table VIII lists the fleet average emission factors which correspond to the upper (1.0 mg/m^3) and the lower (0.03 mg/m^3) limits of the suggested ambient air range of concern. Inspection of this table shows that the scenarios result in a wide range of emission factors corresponding to the health effects range of concern of 0.03 mg/m^3 to 1.0 mg/m^3 . From this table the severe cases, especially for the tunnel scenario, are the ones which require further investigation. Using the fleet average emission factors from Table VI the emission factors for each scenario can be compared to the corresponding range of concern. This comparison is given in Table IX. The fleet average emission factors for the two "worst case" fleet situations were selected for comparison. (To compare the other fleet situations their fleet average emission factors in Table VI can be compared to column A of Table IX). Garage scenarios will be considered separately.

As shown in Table IX, even if it is assumed that the CY 1978 fleet is operating with 25% malfunction, the fleet average emission factor could be within, but not above the range of concern for the severe roadway tunnel situation. Formaldehyde emissions for the street canyon and expressway scenarios appear to be below the range of concern for the current fleet situations explored in this report.

Table VIII

Emission Factors Corresponding to the
Lower and Upper Limits of the Formaldehyde Range of Concern

Ambient Air Scenario	Emission Factor (mg/mile) corresponding to a 0.03 mg/m ³ exposure	Emission Factor (mg/mile) corresponding to a 1.0 mg/m ³ exposure
Roadway Tunnel		
Typical	26.7	890.5
Severe	10.5	350.1
Street Canyon		
Typical	714.3	23,809
Severe	106.4	3546
Expressway		
Typical	241.9	8065
Severe	59.3	1976
Off Road	285.7	9524
Parking Garage*		
Typical	7.7	256
Severe	0.5	18
Personal Garage*		
Typical	3.8	127
Severe	0.4	15

*Emission factors are given in mg/minute for garage exposures.

Table IX

Range of Concern Compared to Potential Emissions

	A	B	C
	Range of Concern (Severe Case) i.e., Fleet Average Emissions (mg/mile) Needed To Be of Concern ^a	Fleet Average Emissions(mg/mile) Assuming CY 1978 Fleet with 25% Malfunction ^b	Fleet Average Emissions(mg/mile) Assuming 100% Methanol Fueled Catalyst-Equipped Vehicles with 25% Malfunction ^b
Roadway Tunnel	10.5-350.1	25.41	32.21
Street Canyon	106.4-3546	27.24	44.50
Expressway	59.3-1976	36.64	35.83

^aFrom Table VIII.^bFrom Table VI.

The highest emission rate reported for formaldehyde is 369.30 mg/mile from a non-catalyst-equipped vehicle. This is above the range of concern for the severe roadway tunnel, but within the range of concern for the severe street canyon and expressway scenarios. Highest formaldehyde emission rates for oxidation catalyst-equipped vehicles (54.40 mg/mile) and 3-way catalyst-equipped vehicles (39.80 mg/mile) fall within the range of concern for the severe roadway tunnel scenario but fall below the range of concern for the severe street canyon and expressway scenarios. These emission rates are for unique vehicles, and it is extremely unlikely that the average emission rate of vehicles in a tunnel would ever be so high.

Referring again to Table IX, for the "worst case" methanol fleet situation given, the fleet average emission factors are within, but not above the range of concern for the severe roadway tunnel situation. As with the CY 1978 fleet situation, the street canyon and expressway scenarios do not appear to present any possible problem regarding formaldehyde exposure for the methanol situations explored in this report.

Garage scenarios were discussed in Section VI. Based on low speed and idle tests conducted with light-duty gasoline- and diesel-fueled vehicles, parking and personal garage

exposures, under severe conditions would fall within, but not above the range of concern. Parking and personal garage exposures with methanol-fueled, catalyst-equipped vehicles would on average fall below the range of concern, based on the limited number of tests that have been run. Emissions from a malfunctioning, methanol-fueled vehicle (i.e., one in which the catalyst was removed) could fall above the range of concern for the severe personal garage situation; however, it is extremely unlikely that emissions from a methanol-fueled vehicle in a garage would ever be so high.

References

- 1) "Clean Air Act as Amended August 1977," Public Law 88-206, 89-272, 89-675, 90-148, 91-604, 92-157, 93-319, 95-95, 95-190.
- 2) "An Approach for Determining Levels of Concern for Unregulated Toxic Compounds from Mobile Sources," R. Garbe, EPA Technical Report No. EPA/AA/CTAB/PA/81-2, July 1981.
- 3) "Ambient Pollutant Concentrations from Mobile Sources in Microscale Situations," M. Ingalls, R. Garbe, SAE Paper 820787, June 1982.
- 4) "Formaldehyde Health Effects," EPA Report No. EPA-460/3-81-033 by Midwest Research Institute under EPA Contract No. 68-03-2928, EPA Project Officer Robert Garbe.
- 5) U.S. EPA Advisory Circular 76, June 1978.
- 6) U.S. EPA Advisory Circular 76-1, November 1978.
- 7) "Formaldehyde and Other Aldehydes," National Research Council report based on EPA Contract No. 68-01-4655, Project Officer Dr. Alan P. Carlin, 1981.
- 8) "Analytical Procedures for Characterizing Unregulated Pollutant Emissions from Motor Vehicles," EPA Report No. EPA-600/2-79-017, February 1979.
- 9) "Investigation of Diesel-Powered Vehicle Emissions VII," EPA Report No. EPA-460/3-76-034, K. Springer, Southwest Research Institute, February 1977.
- 10) "Characterization of Gaseous and Particulate Emissions from Light-Duty Diesels Operated on Various Fuels," EPA Report No. EPA-460/3-79-008, C. Hare, Southwest Research Institute, July 1979.
- 11) "Characterization of Sulfates, Odor, Smoke, POM and Particulates from Light and Heavy Duty Engines - Part IX," EPA Report No. EPA-460/3-79-007, K. Springer, Southwest Research Institute, June 1979.
- 12) "Unregulated Exhaust Emissions from Non-Catalyst Baseline Cars Under Malfunction Conditions," EPA Report No. EPA-460/3-81-020, C. Urban, Southwest Research Institute, May 1981.

- 13) "Regulated and Unregulated Exhaust Emissions from Malfunctioning Non-Catalyst and Oxidation Catalyst Gasoline Automobiles," EPA Report No. EPA-460/3-80-003, C. Urban, Southwest Research Institute, January 1980.
- 14) "Regulated and Unregulated Exhaust Emissions from Malfunctioning Three-Way Catalyst Gasoline Automobiles," EPA Report No. EPA-460/3-80-004, C. Urban, Southwest Research Institute, January 1980.
- 15) "Regulated and Unregulated Exhaust Emissions from a Malfunctioning Three-Way Catalyst Gasoline Automobile," EPA Report No. EPA-460/3-80-005, C. Urban, Southwest Research Institute, January 1980.
- 16) "Emission Characterization of an Alcohol/Diesel Pilot Fueled Compression Ignition Engine and It's Heavy Duty Diesel Counterpart," EPA Report No. EPA-460/3-81-023, T. Ullman and C. Hare, Southwest Research Institute, August 1981.
- 17) "Inspection and Maintenance for 1981 and Later Model Year Passenger Cars," D. Hughes, SAE Paper 810281, February 1981.
- 18) "Characterization of Exhaust Emissions from High Mileage Catalyst-Equipped Automobiles," EPA Report No. EPA-460/3-81-024, L. Smith, Southwest Research Institute, September 1981.
- 19) "Hydrocarbon and Aldehyde Exhaust Emission Species from Three-Way Catalyst Vehicles with Feedback System Disablements," P. Wuebben, J. Wood, and M. Porter, South Coast Air Quality Management District Draft Report, EPA Grant #A00904813, August 26, 1982.
- 20) "Characterization of Exhaust Emissions from Methanol-and Gasoline-Fueled Automobiles," EPA Report No. EPA-460/3-82-004, L. Smith, C. Urban, Southwest Research Institute, August 1982.
- 21) "Emission Characterization of a Spark-Ignited Heavy-Duty Direct-Injected Methanol Engine," Draft Final Report No. EPA-460/3-82-003, T. Ullman, C. Hare, Southwest Research Institute, April 1982.
- 22) Memo: Methanol VW Catalyst Weekly Update Report #8, T. Penninga to C. Gray, U.S. EPA/OMS/ECTD, October 19, 1982.

- 23) State of California Air Resources Board, Alcohol Fueled Fleet Test Program, Project 3T8001, Fleets No. 2 and No. 3, Fourth Interim Report, MS-82-11, August 1982.
- 24) "Heavy Duty Diesel Particulate Emission Factors," T. Baines, J. Somers, C. Harvey, Journal of the Air Pollution Control Association, 29:6, June 1979.
- 25) "Air Quality Assessment of Particulate Emissions from Diesel-Powered Vehicles," PEDCO Environmental, Inc., EPA Contract No. 68-02-2515, Project Officer, J. Manning, March 1978.
- 26) "Mobile Source Emission Factors: For Low Altitude Areas Only," EPA Report No. EPA-400/9-78-006, March 1978.
- 27) "Unregulated Emissions for Vehicles Operated under Low Speed Conditions," L. Smith, Draft Report for EPA Contract No. 68-03-3073, October 1982.
- 28) "Induction of Squamous Cell Carcinomas of the Rat Nasal Cavity by Inhalation Exposure to Formaldehyde Vapor," J. A. Swenberg et al., Cancer Research 40, 3398-3402, 1980.
- 29) "Formaldehyde in the Indoor Environment-Health Implications and the Setting of Standards", I. Anderson, In: Indoor Climate, Effects on Human Comfort, Performance, and Health in Residential, Commercial, and Light-Industry Buildings, Proc. of the First Int. Indoor Climate Symp., Copenhagen, Denmark. August 30-September 1, 1978, P. O. Fanger and O. Valbojorn, Eds. Danish Building Research Institute, Copenhagen, Denmark, pp. 65-87.
- 30) "Hygienic Evaluation of Formaldehyde as an Atmospheric Air Pollutant", V.P. Melekhina, In: USSR Literature on Air Pollution and Related Occupational Diseases-A Survey, NTIS TT64-11574, National Technical Information Service, U. S. Department of Commerce, Springfield, VA., 1964.
- 31) "Influence of Small Concentrations of Formaldehyde Vapors on the Human Organism", A.K. Sgibnev, Gig. Tr. Prof. Zabol. 12(7):20-25, 1968.
- 32) "Effects of Photochemical Air Pollution on the Human Eye-Concerning Eye Irritation, Tear Lysozyme and Tear pH", N. Okawada et al., Nagoya J. Med. Sci. 41 (1-4):9-20, 1979.

- 33) "Combined Action of Six Air Pollutants on the Human Body", M.T. Takhirov, Gig. Sanit. No. 5:100-102 (Russ); Chem. Abstr. 81:110854, 1974.
- 34) "Materials (Information) for Revision of the Maximal Permissible Concentrations of Formaldehyde in the Interior Atmosphere of Industrial Premises", G. N. Zaeva, et al., Gig. Tr. Prof. Zabol. 12(7):16-20 (Russ) 1968; English translation available from John Crerar Library, Chicago, Illinois. Order No. 74-13625-06J.
- 35) "Ban of Urea-Formaldehyde Foam Insulation", Consumer Product Safety Commission, 16 CFR Part 1306, Vol. 47, No. 64, 14366-14421, Friday, April 2, 1982.
- 36) "A Random Sample Survey of Wisconsin Mobile Homes: Formaldehyde Concentrations and Health Effects", L.P. Hanrahan, et al., Wisconsin Division of Health, Department of Health and Social Services, Madison, Wisconsin, 1980.
- 37) "Contaminants in the Air Exhaled by Man", Yu. G. Nefedov, et al., Kosm. Biol. Med. 3(5): 71-77 (Russ), 1969.
- 38) "Determination of a Range of Concern for Mobile Source Emissions of Ammonia", R. Garbe, EPA Technical Report No. EPA/AA/CTAB/PA/81-20, August 1981.
- 39) "The Determination of a Range of Concern for Mobile Source Emissions of Methanol", C. Harvey, EPA Technical Report No. EPA/AA/CTAB/PA/82-10, September 1982.