Exhaust Emissions from Light- and Heavy-duty Vehicles: Chemical Composition, Impact of Exhaust after Treatment, and Fuel Parameters

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This paper presents results from the characterization of vehicle exhaust that were obtained primarily within the Swedish Urban Air Project, "Tätortsprojektet." Exhaust emissions from both gasoline- and diesel-fueled vehicles have been investigated with respect to regulated pollutants (carbon monoxide [CO], hydrocarbon [HC], nitrogen oxides [NO_x], and particulate), unregulated pollutants, and in bioassay tests (Ames test, TCDD receptor affinity tests). Unregulated pollutants present in both the particle- and the semi-volatile phases were characterized. Special interest was focused on the impact of fuel composition on heavy-duty diesel vehicle emissions. It was confirmed that there exists a quantifiable relationship between diesel-fuel variables of the fuel blends, the chemical composition of the emissions, and their biological effects. According to the results from the multivariate analysis, the most important fuel parameters are: polycyclic aromatic hydrocarbons (PAH) content, 90% distillation point, final boiling point, specific heat, aromatic content, density, and sulfur content. — Environ Health Perspect 102(Suppl 4):13–23 (1994).

Key words: chemical characterization, chemometrics, exhaust emissions, fuel impact, multivaviate data analysis, PAH, regulated pollutants, unregulated pollutants

Introduction

The impact of internal combustion engines, Otto, and diesel engines on the environment and our lifestyles has been considerable. In Europe, research and development work on engines within the last two to three decades has been strictly focused on engine performance in terms of power output, fuel economy, reliability, etc., but not on engine emissions. During recent decades, pronounced interest has focused on exhaust emissions and their impact on health and the environment (*1–4*). Due to the rapid increase in the number of vehicles in use, especially in urban areas, engine emissions have become suspected culprits for some of the health effects observed in urban populations (*5*).

Most of the interest in emissions has been focused on passenger cars and other light-duty vehicles, because these categories of vehicles exist in much greater numbers than the heavy-duty vehicles. Vehicle emissions are usually divided into categories of regulated and unregulated pollutants. Regulated pollutants consist of carbon monoxide (CO), nitrogen oxides (NO_x, mainly nitrogen monoxide and nitrogen dioxide), unburned fuel, or partly oxidized hydrocarbons (HC), and particulates. These pollutants are specified by law in most of the industrially advanced countries. Unregulated pollutants are defined as compounds that are not specified by law. However, these unregulated pollutants may well belong to the group of unburned hydrocarbons, but not as individual compounds.

Several of the compounds present in diesel and gasoline engine exhausts are known to be carcinogenic and/or mutagenic (*5*). A group of compounds most often associated with this carcinogenic/mutagenic property are the polycyclic aromatic compounds (PAC) (*6*). This is of interest because unregulated pollutants are generally measured under the same driving conditions as those developed for regulated pollutant evaluations. Consequently, the unregulated pollutant measurements are made under the same engine operation conditions as for regulated pollutants, although the exhaust emission measurements obtained have no legal bearing.

This publication focuses primarily on vehicular pollutants that have been investigated and characterized within the Swedish Urban Air Project (SUAP), Tätortsprojektet (1979 to 1991), founded by the Swedish Environmental Protection Agency (SwEPA); these data were obtained largely from the time period 1985 to 1991. The effects of pollutants emitted from mobile sources are dependent on several factors that will be discussed herein.

Results and Discussion

Gasoline-fueled Vehicles

Organic Halides. Particle-associated organic halides have been identified in exhausts from gasoline and diesel-fueled vehicles (*7*). The most abundant bromofluorenone isomer determined as present in gasoline exhaust corresponds to an emission of approximately 2 µg/km. Haglund et al. (*8*) have investigated the presence of halogenated polycyclic aromatic hydrocarbons (PAH) in urban air, snow, and automobile exhaust, determined qualitatively in all three samples. A possible source of bromine and chlorine in leaded gasoline is the addition of scavengers, such as ethylene dichloride (EDC) and ethylene dibromide (EDB) (*7,8*). However, the presence of bromine in diesel fuel has also been determined (*7*), indicating bromine to be a natural constituent in crude oil. Another explanation for the presence of bromine in diesel fuel may be the storage conditions of the crude oil. Since storage is often in underground cavities on a bed of sea water, bromine present in the sea water may leach into the oil store. Further, polyhalogenated dioxins and furans have been detected in the exhaust emissions of leaded gasoline-fueled

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Table 1. Mean emission factors ($N = 2$ to 4) from constant cruising speeds, 70 and 90 km/hr. Vehicles investigated: carburetor without exhaust after treatment (V1); three-way catalyst system, λ-sond, turbo, and inter-cooler (V2); three-way catalyst system and λ-sond (V3) (14).

	Vehicle type					
	V1		V2		V3	
	70[a]	90[a]	70[a]	90[a]	70[a]	90[a]
CO, g/km	6	6	0.6	1.1	0.22	0.16
HC, g/km	0.50	0.42	0.07	0.1	0.05	0.05
NO$_x$, g/km	1.2	1.2	0.11	0.42	0.04	0.02
Particles, mg/km	2.8	30	0.7	4.2	0.6	5.3
Methanol, mg/km	3.1	7.8	0.07	0.1	0.05	0.1
Ethanol, mg/km	ND	ND	0.05	0.03	0.03	0.03
Benzene, mg/km	42	37	5	7.7	3.9	2.9
Toluene, mg/km	66	39	2.9	12	4.1	2.7
Particulate phase						
PAH, µg/km[b]	1.8	7.6	0.4	2.8	0.2	2.7
Benzo[*a*]pyrene, µg/km	0.05	0.2	0.01	0.15	0.02	0.12
1-Nitropyrene, µg/km	0.08	0.8	0.03	0.16	0.01	0.5
Strain, revertants/m						
Particulate phase						
TA98–S9	8.2	93	1.8	23	0.08	38
TA98+S9	2.9	73	0.9	15	0.3	39
TA100–S9	6.2	66	1.7	13	Toxic	24
TA100+S9	3.3	75	4.5	16	Toxic	27
TA100NR–S9	Toxic	15	1.9	14	Toxic	7.9

ND, not determined. [a] Cruising speed, km/hr. [b] Sum of 29 PAH, three-ringed to six-ringed PAH.

cars (9,10). Haglund and co-workers (10) determined bromfurans to be more prominent than bromdioxins in leaded gasoline exhausts (10). Emissions of polychlorinated dibenzodioxins (PCDD) and polychlorinated dibenzofurans (PCDF) from gasoline-fueled vehicles are not considered to be a major environmental pollutant, as group profiles differ significantly from those obtained in ambient air samples and in the biota (11). This indicates that when emitted from mobile sources, organic bromine derivatives might have a greater impact on the environment than organic chlorine derivatives.

Engine Operating Conditions. The amount of exhaust emitted from light-duty vehicles depends on driving conditions such as load and speed. This correspondence has been investigated with respect to regulated exhaust emissions (12). It was demonstrated that a correlation between increased speed of the vehicles and CO and NO$_x$ emissions was evident for speeds over 50 km/hr for vehicles without a catalyst. However, studies of exhaust emissions from gasoline-fueled passenger vehicles, with and without a three-way catalyst, have been carried out in combination with bioassay tests to obtain emission factors for both regulated and unregulated pollutants (13,14). Both constant-cruising-speeds (70 and 90 km/hr) and transient driving conditions were investigated. Mean value emission factors and biological activity data for this investigation are given in Table 1. As expected, emissions from catalyst-equipped vehicles showed a dramatic decrease compared with those from the vehicle without a catalyst. Although the emission of CO, HC, and NO$_x$ did not change significantly when results from the 70 km/hr and 90 km/hr tests were compared, there was a significant change in the particle phase emissions. However, it must be pointed out that, despite precautions taken to reduce sample-to-sample variation, relatively high standard deviations (in some cases up to 100%) were observed for the emission factors of unregulated pollutants and for detected mutagenic effects. The variability is discussed in more detail by Westerholm et al. in another paper (14).

Fuel–PAH-related Emissions. The effect of fuel-PAH content in gasoline was investigated with respect to fuel-related (unburned) PAH and to PAH formed during the combustion process (15). The vehicle used was equipped with fuel injection and oxygen sensor (λ-sond), but the catalyst section was removed. Four fuels with PAH content ranging from less than 0.1 mg/L to 54 mg/L were investigated in transient driving conditions (FTP-73). Both particle- and semi-volatile phase-associated PAH were sampled and quantified. A decrease in fuel-PAH content resulted in a decrease of emitted PAH.

Using a simple mathematical method to distinguish PAH present in the fuel and PAH formed in the exhaust emission, the intercept value can be interpreted as PAH formed in the combustion process (15,17). Using this method, we found that a major portion (>50%) of emitted PAH is formed

Table 2. Emission of PAH (µg/km), particulate phase associated, $N = 4$, mean value +/– standard deviation (SD) (%), semi-volatile associated, $N = 2$, mean value +/– SD (%). Lead-free gasoline, US FTP-73 test, λ-sond without catalyst (15).

	Particulate-associated		Semi-volatile-associated	
	Mean value	SD	Mean value	SD
Naphthalene	ND	–	23	18
Biphenylene	ND	–	8.3	20
Acenaphthylene	ND	–	28	35
Fluorene	ND	–	42	35
Phenanthrene	ND	–	91	45
Anthracene	ND	–	28	36
1-Methylphenanthrene	ND	–	16	26
Fluoranthene	2.5	6	18	31
Pyrene	4.6	9	17	34
Methylpyrene	ND	–	0.32	39
Benzo[*g,h,i*]fluoranthene	2.8	24	0.69	57
Cyclopenta[*c,d*]pyrene	3.3	14	ND	–
Benzo[*a*]anthracene	3.2	14	1.1	20
Chrysene/triphenylene	3.5	18	0.51	13
Benzo[*b,k*]fluoranthene	4.8	28	ND	–
Benzo[*e*]pyrene	2.7	17	ND	–
Benzo[*a*]pyrene/benzo[*c,d*]pyrenone	6.6	15	ND	–
Indeno[*1,2,3-c,d*]pyrene	1.6	15	ND	–
Benzo[*g,h,i*]perylene	5.5	10	ND	–
Coronene	2.4	22	ND	–
PAH[a]	<44.0		<274.00	

ND, not detected. [a] Sum of 22 PAH, three-ringed to six-ringed PAH.

Table 3. Emission of PAC (µg/km), particulate phase-associated, *N* = 6, mean value +/– standard deviation (SD) (%), semi-volatile-associated, *N* = 2, mean value +/– SD (%) (*27*).

	Particulate-associated		Semi-volatile-associated	
	Mean value	SD	Mean value	SD
Phenanthrene	29	48	266	4
Anthracene	3.3	13	10	7
3-Methylphenanthrene	30	27	95	8
2-Methylanthracene	40	24	103	9
4- and 9-Methylphenanthrene	48	21	106	7
1-Methylphenanthrene	41	20	87	2
Fluoranthene	17	11	5.6	6
Pyrene	11	14	2.1	23
Benzo[*a*]fluorene	2.4	32	0.18	20
2-Methylpyrene	2	19	0.13	12
1-Methylpyrene	0.75	35	0.10	20
Benzo[*g, h, i*]fluoranthene	1.5	19	<0.01	–
Cyclopenta[*c, d*]pyrene	0.18	30	<0.01	–
Benzo[*a*]anthracene	0.47	54	<0.01	–
Chrysene/triphenylene	2.8	18	<0.01	–
Benzo[*b, k*]fluoranthene	0.29	31	<0.01	–
Benzo[*e*]pyrene	0.15	28	<0.01	–
Benzo[*a*]pyrene	<0.06	–	<0.01	–
Perylene	<0.01	–	<0.01	–
Indeno[*1, 2, 3-c, d*]fluoranthene	<0.09	–	<0.01	–
Indeno[*1, 2, 3-c, d*]pyrene	<0.04	–	<0.01	–
Picene	<0.01	–	<0.01	–
Benzo[*g, h, i*]perylene	<0.13	–	<0.01	–
Coronene	<0.01	–	<0.01	–
PAH[a]	<23	22	<678	6
1-Nitropyrene	1.6	60	<0.02	
Dibenzothiophene	<0.02		0.15	27
4-Methyldibenzothiophene	0.06	21	0.22	18
3-Methyldibenzothiophene	0.10	35	0.34	24

[a] Sum of 24 PAH, three-ringed to six-ringed PAH.

in the combustion process. It was found that when a full range of aromatics (mono, di, and larger aromatics) was present in the fuel, more mutagens were produced in the combustion process. Comparison of PAH distribution between the particle- and the semi-volatile phases, where samples were collected using the dilution tunnel technique or from ambient air revealed that PAH profiles were similar in both of the collection methods: sampling of vehicle gasoline exhaust using the dilution technique generated particle- and semi-volatile phase-associated PAH profiles similar to those measured in urban environments (*18*).

The evaluation of biological activity of the semi-volatile phase leads initially to controversial results. Stump et al. showed that an adsorbent trap enriched very low amounts of mutagenic material compared to the level in the filter extracts (*19*). Schuetzle has also reported similar results (*20*). However, using the cryogenic condenser technique (*21*) the mutagenic activity of the semi-volatile phase was determined to be approximately 30 to 50% of the total mutagenicity (*15,22,23*). These results demonstrate the need to investigate both particle- and semi-volatile phases when characterizing exhaust emissions from gasoline-fueled vehicles. This statement is also valid for characterization of diesel exhaust, further discussed in the section "Diesel-fueled Vehicles." In Table 2, particulate-associated and semi-volatile-phase-associated PAC originating from the dilution tunnel technique are presented (*16*).

Diesel-fueled Vehicles

Characterization. A literature survey of identified and tentatively identified constituents in diluted diesel exhaust emissions was performed within the SUAP (*16*), which indicated that approximately 450 individual compounds have been detected in diesel exhaust. Quite a few new compounds have been identified or tentatively identified in diesel exhaust samples since 1987 (*24*). However, several compounds present in diesel exhaust have not been identified and investigated with respect to possible environmental and health effects.

Our evaluation of diesel exhaust emissions under the SUAP has employed a methodology designed for heavy-duty diesel exhaust emission characterizations, which was developed in close cooperation with the former SwEPA Emission Research Section in Studsvik (*25*). As for gasoline-fueled vehicles earlier presented, the evaluation, in conjunction with bioassay tests, characterizes both regulated and unregulated pollutants. Samples obtained consist of particulate material, semi-volatile material, and gaseous components. Vehicles are operated during transient driving conditions on a chassis dynamometer, and prior to sampling, the exhaust is diluted in accordance with the *Federal Register* (*26*). The transient driving schedule used is the "bus cycle," which simulates public transportation conditions within a city. The bus cycle was originally developed at the Technical University of Braunschweig (Germany). The development of sampling methodology, chemical analysis of the particulate and semi-volatile associated unregulated pollutants and bioassay tests are described in more detail elsewhere (*25,27*). A particulate filter was used downstream from the diluted diesel exhaust for collection of the particulate phase, and polyurethane foam plugs were used for sampling of the semi-volatile phase.

Sampling results indicated that the semi-volatile phase PAH content was approximately 280% higher than the particle-phase PAH. Most of the gas phase PAH mass consisted of three-ring compounds (*27*). The contribution of semi-volatile-phase-associated mutagenicity to the total mutagenicity was approximately 20% in strain TA100 +/–S9; in strain TA98–S9, the contribution was 10%; and in strain TA98+S9, 37%. Both particle- and semi-volatile phase crude extracts were fractioned into five fractions by a method originally developed for crude gasoline extracts (*28,29*). The greatest mutagenic activity was found in both phases in the fractions containing mono- and dinitro-PAH (*27*). Chemical analyses of PAC measured in particulate and semi-volatile phases as determined from diluted diesel exhaust that was sampled from the bus cycle are displayed in Table 3.

Fuel Effects. Eight diesel fuels and two heavy-duty vehicles (bus denoted vehicle 1, truck denoted vehicle 2, respectively) were selected for investigation of fuel composition for regulated pollutants, unregulated pollutants, and biological effects (via mutagenicity and TCDD-receptor affinity tests) in the exhaust emissions. Exhaust emission tests were repeated in triplicate for each fuel blend and vehicle. The investigation also focused on specification of fuel parameters in order to obtain less polluting diesel fuels for vehicles operating in cities. One common, commercial, standard diesel fuel was used as a reference for commercial

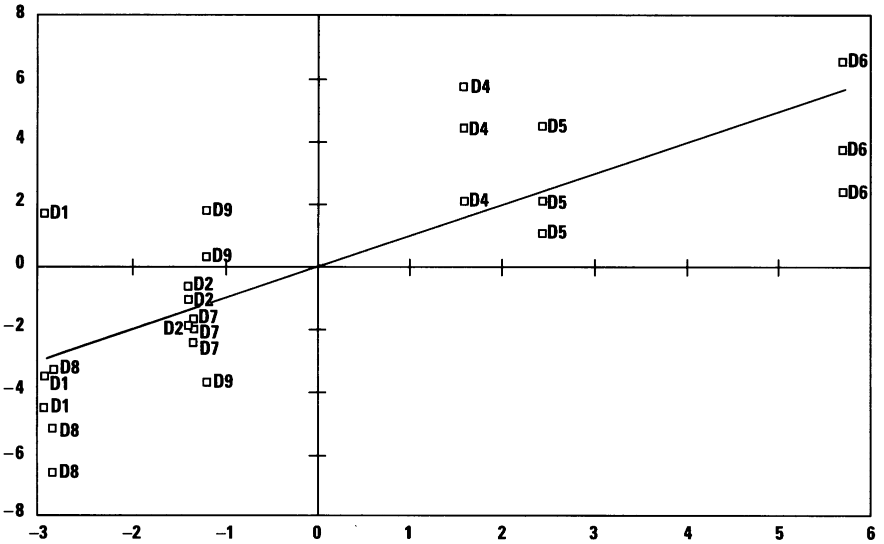

Figure 1. A partial least squares (PLS) regression model of different fuel blends (D1–D9) for vehicle 2, where the x-axis is the PLS-component $X1$ (representing the fuel variables), and the y-axis is the PSL-component $Y1$ (representing the regulated pollutants). The percentage within the brackets indicates how much of the variance is explained by that PSL-component (i.e., $X1$ [35.9] and $Y1$ [23.1]). PSL regression coefficient 0.33, correlation coefficient $R = 0.79$.

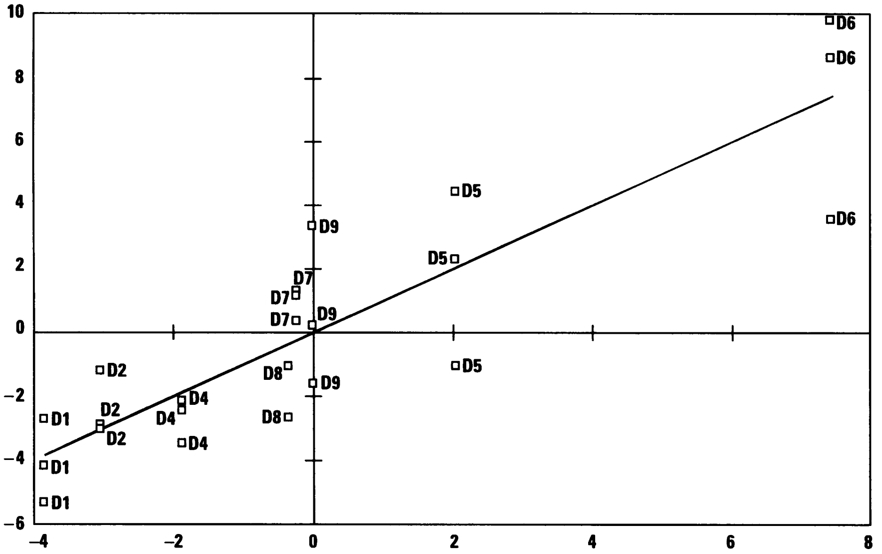

Figure 2. A partial least squares (PLS) regression model of different fuel blends (D1–D9) for vehicle 2, where the x-axis is the PLS component $X1$ (representing the fuel variables), and the y-axis is the PLS-component $Y1$ (representing the unregulated compounds). The percentage within the brackets indicates how much of the variance is explained by that PLS component (i.e., $X1$ [45.1] and $Y1$ [20.1]). PLS regression coefficient 0.64, correlation coefficient 0.88.

fuels available on the Swedish market; three are commercially available and commonly used for city buses, and four are test fuels. The entire investigation is described in detail in an earlier publication (*25*).

This presentation is a very condensed version of the results and findings from our investigation of the bus cycle, using a chassis dynamometer and the dilution tunnel technique. Chemometrics or multivariate analysis methodologies were used to help analyze the large quantities of data produced by this study (*25*). In order to build quantitative relations between different blocks of data, such as fuel parameters, regulated pollutants, unregulated pollutants, and bioassay emission data, partial least squares regression (PLS) was used (*30*). The relationships demonstrated were relatively vehicle-independent.

Figure 1 shows the PLS regression of the fuel blends (denoted by Daud numbers 1, 2, 4, 5, 6, 7, 8, and 9, respectively); the x-axis is the PLS component $X1$ representing fuel parameters, and the y-axis is the PLS component representing regulated emissions (HC, CO, NO_x, and particulate). The PLS regression coefficient $K = 0.33$ (correlation coefficient $R = 0.79$) was obtained. The PLS regression is displayed in Figure 2, in which the x-axis is the PLS component $X1$ representing fuel parameters, and the y-axis is the PLS component $Y1$ representing unregulated pollutants emitted (aldehydes, olefins, light aromatics, PAH, 1-nitropyrene). The PLS regression coefficient $K = 0.64$ (correlation coefficient $R = 0.88$) was obtained. In a similar manner, the PLS regression model for biological effects is displayed in Figure 3, where the y-axis represents the variables of biological effects measured in the emission (via Ames test and TCDD receptor affinity test, in both the particulate and semi-volatile phases). The PLS regression coefficient $K = 0.58$ (correlation-coefficient $R = 0.89$) was obtained. According to the results from the PLS analysis, the most important fuel parameters are: PAC content, 90% distillation point, final boiling point, specific heat, aromatic content, density, and sulfur content. All these fuel parameters should be decreased, except the specific heat, which should be increased to reduce emissions.

We concluded from the study that there exists a quantifiable relationship between the variables of the diesel-fuel blends and the variables of the chemical emission and their biological effects. Figure 4 illustrates the z-axis, the PLS-component $X1$ that represents the emission of both regulated and unregulated exhaust components, and the y-axis, the PLS-component $Y1$ that represents measured biological effects in the exhaust emissions. From the figure, we observe how the PLS regression coefficient $K = 0.62$ (correlation coefficient $R = 0.85$) is obtained. An important emission parameter with respect to biological activity is the sum of PAH in both the particle and semi-volatile phases. Diesel fuel-related emission factors emanating from a standard diesel fuel (D6, Swedish summer diesel-fuel quality), a diesel fuel for city buses (D8), and a test fuel (D1) are presented in Table 4. The PLS regression coefficients obtained in Figures 3, 4, and 5 indicate that there is a greater impact from unregulated pollutants and biological effects in the exhaust emissions, compared to regulated pollutants by selection of fuel.

Diesel Exhaust after Treatment. NO_x and particles are two diesel emission components that have an adverse effect on urban air quality. A possible technique to reduce particulate emissions is the use of an exhaust after treatment system, such as a particulate trap, catalyst, or a combination of both. In Table 5, emission results from two different particle traps are presented (*31*). Table 6 shows the emission results of exhaust that is after treated with a catalyst in combination with a particulate trap (*32*). From these results it can be concluded that diesel exhaust after treatment reduces emissions. For the wire mesh particulate trap (*31*), although general NO_x emissions were unaffected, nitrogen dioxide appeared at increased levels in certain modes with increased load and speed. This was investigated in a 13-mode diesel engine driving cycle, which does not include transient driving conditions. A general issue regarding particulate traps is the need to investigate in more detail the regeneration process with respect to chemical composition and biological effects of the exhaust gases.

Alternative Fuel Vehicles. Besides gasoline and diesel, other fuels can be used as the energy source in automotive combustion engines. Egebäck and Westerholm made a literature survey of Swedish research programs to evaluate possibilities and present aspects of using alcohols as automotive fuels (*33*). Research programs for alternative fuels depend strongly on political considerations, as well as the price and access to crude oil. The use of alcohol fuels depends not only upon possible replacement of some share of imported motor fuels and crude oils with domestic fuel constituents, but also on possible environmental improvements, such as reduction of exhaust emissions.

Table 7 lists emissions data from light-duty vehicles, originating from the U.S. FTP-72 driving cycle (*34*) fueled with 15 and 95% methanol in gasoline. Despite the measures taken for light-duty vehicles, there is still a need to reduce exhaust emissions from heavy-duty vehicles, especially vehicles intended for operation in urban areas. The technologies used in Sweden for compression ignition engines are a dual fuel system (*35,36*), and engines run on alcohol with an ignition improver (*36,37*). Of these two engine concepts, the latter seems to have the greater potential to reduce emissions when used in vehicles operated in cities. Presented in Table 8 are emission factor data from heavy-duty diesel vehicles fueled with alcohols.

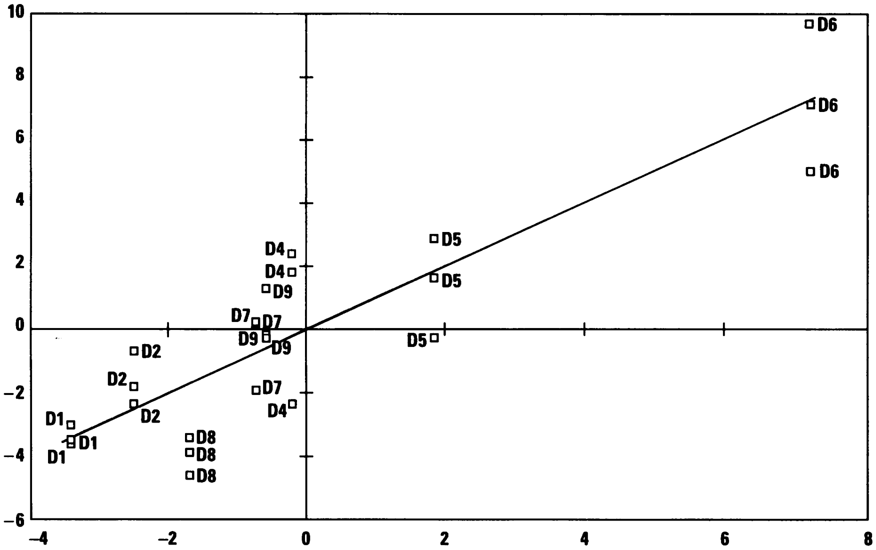

Figure 3. A partial least squares (PLS) regression model of different fuel blends (D1–D9) for vehicle 1, where the x-axis is the PLS component *X*1 (representing the fuel variables), and the y-axis is the PLS-component *Y*1 (representing the variables of biological effect). The percentage within the brackets indicates how much of the variance is explained by that PLS component (i.e., *X*1 [44.5] and *Y*1 [43.6]). PLS regression coefficient 0.58, correlation coefficient 0.89.

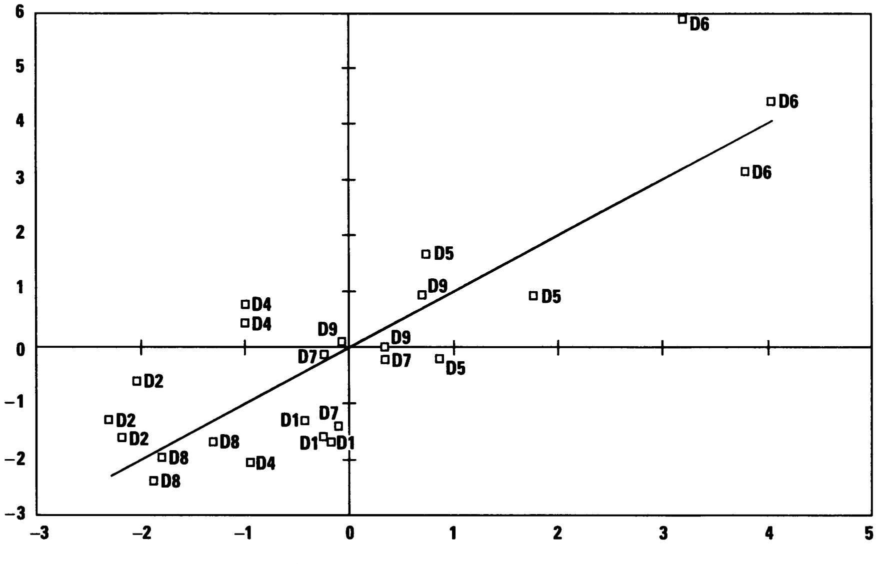

Figure 4. A partial least squares (PLS) regression model of different fuel blends (D1–D9) for vehicle 1, where the x-axis is the PLS component *X*1 (representing the chemical emission variables), and the y-axis is the PLS-component *Y*1 (representing the variables of biological effect). The percentage within the brackets indicates how much of the variance is explained by that PLS component (i.e., *X*1 [23.3] and *Y*1 [33.5]). PLS regression coefficient 0.62, correlation coefficient 0.85.

Impact of Ambient Air Temperature on Exhaust Emission. Egebäck and co-workers (*47*) investigated the impact of ambient temperatures as low as −10°C on the exhaust emissions from cars fueled with either gasoline or a 15% methanol-gasoline blend. As can be seen in Table 9 there is an increase of emissions at lower ambient temperatures. This shows that published vehicular emission factors that are normally obtained at approximately 20°C are underestimated for those areas with mean ambient temperatures below 20°C. This fact must be considered when performing risk assessment of air pollutants.

Technical Development

Engines and Vehicles

Engine and vehicle development are discussed in this section, and the benefit of using improved and alternative fuels is considered. Lead in fuel has long been regarded as an

Table 4. Diesel fuel-related exhaust emissions obtained in the bus cycle driving conditions ($N = 3$). Two vehicles investigated: a heavy-duty diesel passenger bus (V1) and a heavy-duty diesel truck (V2). Only three of eight fuels are presented here (i.e., commercial, standard diesel fuel) (D6), commercial diesel fuel for city buses (D8), and one of the test fuels (D1) (*25*).

	Fuel type					
	D6		D8		D1	
	V1 [a]	V2 [a]	V1 [a]	V2 [a]	V1 [a]	V2 [a]
CO, g/km	3.1	8.6	2.8	6	2.4	7.2
HC, g/km	1.3	1.2	1.6	1.3	1.2	1.4
NO_x, g/km	13.2	16.4	11.9	15.4	11.8	15.4
Particles, g/km	0.51	0.58	0.47	0.47	0.40	0.50
Methanol, mg/km	ND	ND	3	59	3.8	ND
Ethanol, mg/km	ND	ND	63	42	25	63
Ethylene, mg/km	33	135	41	64	ND	92
Propylene, mg/km	7.9	6.4	7.4	11	ND	9.6
Benzene, mg/km	7.2	13	4.3	8.7	8.4	8.3
Toluene, mg/km	4.9	14	1.9	4	9.2	4.9
Formaldehyde, mg/km	0.07	0.12	0.13	0.07	0.12	0.06
Acetaldehyde, mg/km	0.02	0.10	0.08	0.11	0.05	0.04
Particulate phase						
PAH, μg/km [b]	220	160	73	89	44	49
Benzo[*a*]pyrene, μg/km	0.19	0.47	0.17	0.57	0.07	0.22
1-Nitropyrene, μg/km	1.1	1.8	0.2	0.4	0.6	0.3
Semi-volatile phase						
PAH, μg/km [b]	230	200	67	35	110	26
Mutagenicity, revertants/m						
Particulate phase						
Strain						
TA98–S9	48	197	6.6	52	7.3	25
TA 98+S9	59	73	6.5	19	8.3	12
TA 100–S9	176	596	13	263	26	203
TA 100+S9	156	311	32	115	27	82
Semi-volatile phase						
Strain						
TA 98–S9	1.6	2.2	3.7	8.8	Toxic	5.6
TA 98+S9	10	543	83	20	Toxic	13
TA 100–S9	17	40	3.8	105	Toxic	63
TA 100+S9	23	192	33	58	7.3	42
TCDD receptor affinity test, IC_{50} [c], m/ml						
Particulate phase	0.03	0.03	0.11	0.08	0.24	0.09
Semi-volatile phase	0.02	ND	0.15	>0.5	0.09	>0.5

ND, not determined. [a] Vehicle type. [b] Sum of 27 PAH, three-ringed to six-ringed PAH. [c] IC_{50} relative binding affinities are expressed as the amount of sample required to compete for 50% of the [^{3}H]TCDD-binding sites.

air toxin; it is also harmful to the catalysts used to clean the exhaust emitted from vehicles. Also, there are components in diesel fuel that should be removed or reduced. The requirement to reduce automotive emissions has led to a new strategy for the development of internal combustion engines and vehicles. For a long time, the main parameters for improvement of internal combustion engines were fuel consumption and driveability, and these parameters are still of high priority for the engine developer. The challenge is now to integrate the emission requirement into engine design, and to minimize the possible conflict between good emission performance and high fuel efficiency. When discussing emission technology, the vehicle itself also must be integrated into the discussion. The design of the vehicle body, rolling friction coefficient, and the weight and load are greatly important since they impact emissions, as well as fuel consumption.

Gasoline-fueled Vehicles. In Europe, air pollution from motor vehicles had become a problem by the early 1960s. The number of vehicles, especially passenger cars, increased rapidly, and the emissions levels of carbon monoxide, hydrocarbons, and lead from the individual vehicles became high. Common understanding among the European authorities about the health and environmental effects of automotive pollution was lacking, and therefore there was no fast process for developing standards and regulations in order to reduce the tailpipe emissions. Sweden enacted the first set of standards and regulations for light-duty vehicles of 1971 and later models. These standards were active until 1976, when the U.S. standards for vehicles of 1973 and later models were introduced. These standards were in force from 1976 to 1988 models of passenger cars; the 1989 U.S. standards since have been in force on a mandatory basis. Table 10 presents estimated emission data for gasoline-fueled vehicles, ranging from pre-1971 to 1989 models.

Since the 1992 model, there also have been standards for light-duty trucks and buses. In Sweden, the main technology used until the 1988 model of light-duty vehicles has been engine modifications. In 1987, vehicles using three-way technology were introduced for voluntary use on the Swedish market. In order to speed up the introduction of the low-emission vehicles, the Swedish government introduced economic incentives giving the buyers of these vehicles a credit of approximately $1000.

Diesel-fueled Heavy-duty Vehicles and Engines. In many countries where actions have been taken to reduce emissions from light-duty vehicles, there is now a growing concern about the emissions from heavy-duty vehicles. Since almost all heavy-duty vehicles in Europe, including Swedish vehicles, are equipped with diesel-driven engines, much of the interest is focused on this engine type. Although different types of engines exist in Europe (the indirect injection and the direct injection engines), the focus here is on the direct injection engine, since the heavier heavy-duty vehicles are commonly equipped with this engine type. Swedish manufacturers have successfully developed fuel-efficient, turbo-charged engines with intercooler. Today the great majority of new engines for heavy duty vehicles on the Swedish market are turbo-charged. Since the early 1980s, repeated investigations of emissions from heavy-duty diesel-fueled vehicles in Sweden have yielded some important results, which have been presented here. Since Sweden has not yet introduced mandatory emission standards for heavy-duty vehicles (except for smoke), there is great variation in emission behavior from one type of vehicle to another. The manufacturer of the vehicle has not had any other obligation for technical development than to meet the customer's demand (i.e., to develop a fuel-efficient and durable vehicle). For public city buses, a new voluntary requirement has been agreed upon; the customers, which are primarily community-owned bus companies, require that the new buses meet certain emission standards. In the 1993 model, Sweden introduced mandatory emission requirements for all heavy-duty vehicles meaning that they will be required to meet the following standards: 1.2 g/kWh, 4.9 g/kWh, 9.0 g/kWh, and 0.4 g/kWh for HC, CO, NO_x, and particulates, respectively, when measured according to the 13-mode test method defined by the Stockholm group.

The technologies that are being developed to reduce emissions from heavy-duty diesel-fueled vehicles include engine modification, improved diesel-fuel, and exhaust-gas after treatment (*38*). The question of which technology should be used also depends on the air quality objectives. If the objective is to reduce those substances in the exhaust suspected of increasing cancer incidence, improvement of fuel quality, in combination with a catalyst or a particulate filter most probably will have to be used. On the other hand, to meet the 1993 Swedish emission standards, the only measure the vehicle manufacturer will have to take is to modify the engine, a modification that is not clearly defined. There is a full range of technical improvements available, such as turbo-charging, intercooling, in-cylinder air motion or turbulence (swirl or no-swirl) matched with the type of fuel injection system, and combustion chamber configuration (*48*). No- or low-swirl motions require high injection pressures and multi-hole injectors in order to atomize the fuel, and to achieve a complete air-fuel mixture.

Other engine modifications include injection duration and time, as well as combustion duration. An advanced technology is the use of electronically governed fuel injection of the engine. All measures taken to reduce the peak temperature in the combustion chamber during the combustion mode will decrease NO_x emission. Several possibilities for reducing the peak temperature include turbo-charging in combination with intercooling (if not used to increase the power output of the engine), retarded ignition timing, and exhaust gas recirculation (EGR). On the other hand, there is the risk that retarded ignition timing and EGR will increase particle emission. The increase of the smoke level because of retarded ignition timing can be overcome by an increase of the injection rate and in-cylinder air motion. The example shows that those techniques which do not increase other emission components must be used. It also shows that optimization of engine and combustion parameters is possible. Hydrocarbon emis-

Table 5. Mean exhaust emission factors from a heavy-duty diesel truck with and without two particulate traps (A and B) originating from measurements in the bus cycle (*31*).

Compound	Without trap	Trap A	Trap B
CO, g/km	5.5	4.9	1.6
HC, g/km	3.7	1.9	2.2
NO_x, g/km	16	13.6	15.1
Particles, g/km	0.7	0.1	0.2
Formaldehyde, mg/km	140	170	110
Acetaldehyde, mg/km	48	60	30
Particulate phase			
PAH, µg/km [a]	73	20	21
Benzo[*a*]pyrene, µg/km	1.6	0.2	0.2
1-Nitropyrene, µg/km	0.9	0.4	0.3
Mutagenicity, revertants/m			
Particulate phase			
Strain			
TA98–S9	200	100	70
TA98+S9	230	105	60
TA100–S9	450	130	220
TA100+S9	460	200	200
TA100NR–S9	220	110	130

[a] Sum of 14 PAH, three-ringed to six-ringed PAH.

Table 6. Mean exhaust emission factors from a heavy-duty diesel truck equipped with a catalyst and a particulate trap, originating from measurements in the New York City cycle (*32*).

Compound	Without trap or catalyst	With trap and catalyst
CO, g/km	10.5	5.3
HC, g/km	2.1	0.55
NO_x, g/km	34.8	34.4
Particles, g/km	0.73	0.26
PAH, µg/km [a]	85.00	4.5
Benzo[*a*]pyrene, µg/km	0.57	0.05
1-Nitropyrene, µg/km	2.4	00.75

[a] Sum of 14 PAH, three-ringed to six-ringed PAH.

Table 7. Mean emission factors levels obtained in the U.S. FTP-72 driving cycle using different light-duty passenger vehicle fuel/engine concepts: lead free, 15% methanol (V1); lead free, 15% and three-way catalyst λ-sond (V2); and 95% methanol and catalyst without feedback control (V3) (*34*).

	Vehicle type		
	V1	V2	V3
CO, g/km	16	3	5.6
HC, g/km	1.6	0.36	0.66
NO_x, g/km	1.8	0.20	0.19
Methanol, mg/km	125	9	1320
Ethanol, mg/km	4.3	6.4	ND
Ethylene, mg/km	80	20	1.8
Propylene, mg/km	28	6.7	ND
Benzene, mg/km	110	35	ND
Toluene, mg/km	230	41	ND
Formaldehyde, mg/km	35	1.3	19
Methylnitrite, µg/km	104	3 5	630
PAH, µg/km [a]	150	34	3.4
Benzo[*a*]pyrene, µg/km	4.1	1.5	<0.1
Mutagenicity, revertants/m			
Particulate phase			
Strain			
TA98–S9	29	3.4	0.5
TA98+S9	15	1	0.7
TA100–S9	25	2.1	<0.1
TA100+S9	40	5.3	1.3

ND, not determined. [a] Sum of 14 PAH, three-ringed to six-ringed PAH.

Table 8. Mean emission factor levels obtained in the bus cycle using different heavy-duty vehicle fuel/engine concepts: ethanol diesel dual fuel system (V1) (*36*); ethanol/ethyl hexyl nitrate and an oxidizing catalyst (V2) (*37*); and ethanol/polyethylene glycol and an oxidizing catalyst (V3) (*37*).

	Vehicle type		
	V1	V2	V3
CO, g/km	14	2.2	3.9
HC, g/km	1.8	1.8	1.4
NO_x, g/km	16	17	17
Ethanol, mg/km	ND	240	360
Formaldehyde, mg/km	160	79	89
Acetaldehyde, mg/km	59	450	340
PAH, µg/km	170	3	5
Benzo[*a*]pyrene, µg/km	1.6	<0.07	<0.03
1-Nitropyrene, µg/km	ND	0.45	0.39
Mutagenicity, revertants/m Particulate phase Strain			
TA98–S9	ND	87	176
TA98+S9	ND	36	49
TA98NR	ND	35	77
TA100–S9	830	100	120
TA100+S9	ND	94	106

ND, not determined. [a] Sum of 14 PAH, three-ringed to six-ringed PAH.

sion levels from a well functioning diesel engine are usually low if compared to the standards. Specific components in the exhaust, such as PAC, light aromatics, aldehydes, etc., can be efficiently reduced with a catalytic converter (*39*). Catalysts coated with precious metals (platinum, palladium, rhodium, and silver) are effective in oxidizing CO and gaseous HC, particulate soluble organic fraction (SOF), and other components, such as aldehydes, ketones, etc., but are not efficient in oxidizing the soot emissions (*40*). There are differing opinions among the heavy-duty vehicle manufacturers about particulate emissions. Some manufacturers are convinced that the U.S. 0.1 g/Bhph (gram/brake horse power hour) particulate emission standard can be met by an efficient engine modification in combination with an oxidation catalyst. Other manufacturers believe that a particle filter may solve the problem by trapping the particles with a specially designed filter element. This filter poses the difficulty of getting rid of the trapped particles to regenerate the filter. Different ideas for filter regeneration have been tested or are under investigation (*41*). Many solutions are under consideration, such as heating the filter with a burner or electricity, or by using a special additive. One solution may be to coat the filter element with a precious metal.

Fuels

In this section we discuss gasoline, diesel fuel, methanol, ethanol, and natural gas as automotive fuels. There are alternative fuels to methanol, ethanol, and natural gas, such as rape oil and hydrogen, but there is very little tested experience with these fuels. Important gasoline fuel parameters are octane numbers (RON and MON), density, lead and benzene content, volatility, vapor pressure and content of aromatics, olefins, and paraffins (*42*). To meet car manufacturers' requirements for a high-octane fuel, lead was added to the fuel. There has also been a strong demand for a highly volatile fuel, especially during winter.

It became evident 20 to 30 years ago that measures had to been taken to improve fuel quality for environmental reasons. Lead in fuel has been proven to cause brain damage in children living in areas with high levels of pollution from gasoline-fueled vehicles. Another reason for keeping lead out of gasoline is that lead acts as a poison to the catalyst. In the section of this article titled Organic Halides, we showed that the scavengers used in connection with lead contribute to the emission of dioxins as well. It is well known that benzene is a harmful substance to man, and that a high level of aromatics in the fuel increases the exhaust emission of benzene and PAH. It has also been shown that high fuel volatility increases the evaporative emission, and that a certain level of olefin may cause a buildup of deposits in the engine's intake system (carburetor and intake valves).

To improve the quality of gasoline, different measures have already been taken and more will be taken in the future. The most progressive step taken has been an agreement between the U.S. Environmental Protection Agency and the oil industry to establish rules and implementation guidelines for introduction of reformulated gasoline (*43*). The proposal states:

> Reformulated gasoline produced before March 1, 1997, will be certified by EPA if it results in no increase in oxides of nitrogen (NO_x); contains no more than 1.0 volume percent of benzene; contains at least 2.0 weight percent of oxygen; contains no heavy metals unless valved; and meets or is below the following Reid Vapor Pressure (RVP) specification during the ozone season: In Class B areas, 7.2 psi RVP, and in Class C areas, 8.1 psi RVP.

Similarly, in Sweden, a new standard that specifies both summer and winter gasolines has been approved (*44*). The summer-grade fuel will have an RVP of 45 to 75 kPa (6.53 to 10.88 psi) and the winter grade fuel 65 to 95 kPa (9.43 to 13.78 psi). The specification for benzene remains at 5.0% by volume.

Although diesel fuels can be of different blends, there is a well-pronounced demand from engine and vehicle manufacturers that the diesel fuel fulfill a certain standard. Important parameters from the fuel and engine manufacturers' points of view are cetane number/cetane index, density, distillation Coald Filtering Pluging Point (CFPP), viscosity, specific energy, and sulfur content. From the environmental point of view, there are certain fuel parameters of special importance: sulfur content, aromatic content (including the PAH content), initial boiling point (IBP), 95% distillation point, density, and cetane number.

In the discussion between environmental authorities and engine manufacturers, much emphasis has focused on sulfur and aromatic content in fuels. The impact of sulfur and aromatics in the fuel on the exhaust emissions has been the subject of investigations (*45,46*). The impact of sulfur has been stressed particularly, since sulfur has been shown to contribute to the particulate emission because of sulfate formation in the exhaust. A secondary sulfur reaction contributes to the deterioration of catalysts and particle filters. Reactions between sulfur and components such as phosphorus and calcium in the oil or fuel may also clog the exhaust treatment systems. In the earlier section, Fuel Effects, we discussed the impact of aromatics and PAH in diesel fuel. Results from an extensive investigation of eight fuels used in two different Swedish manufactured vehicles have clearly shown that aromatics and PAH have a measurable and certain impact on the emissions (*25*). Since the vehicles in the cited

Table 9. Ambient temperature-dependent exhaust emission levels, U.S. FTP-72 driving cycle, gasoline and 15% methanol-gasoline blend. Five different vehicles fitted with carburetor (*47*), mean values.

	Gasoline[a]			15% methanol[b]		
Temperature°C	22	5	−10	22	5	−10
CO, g/km	16	16	25	15	17	24
HC, g/km	1.6	1.7	2.5	1.6	2.0	2.8
NO_x, g/km	17	17	35	20	18	45
Particulate, mg/km	17	17	35	20	18	45
Formaldehyde, mg/km	4.4	4.2	6.3	3.6	4.8	8.7
Acrolein, mg/km	4.4	4.2	6.3	3.6	4.8	8.7
PAH sum of 14, µg/km[c]	110	140	610	210	400	930
Benzo[*a*]pyrene, µg/km	4.4	6.7	23	11	19	38

[a] Commercially available gasoline with 40 ppm; sum of 14 PAH. [b] 15% methanol-gasoline blend with 100 ppm; sum of 14 PAH. [c] Sum of 14 PAH, three-ringed to six-ringed PAH.

Table 10. Estimated emission data for gasoline-fueled passenger vehicles. The emission data are estimated to be mean values for vehicles in service. Evaluations and calculations were made by Karl-Erik Egebäck.

Model year	CO, g/km	HC, g/km	NO_x, g/km
Pre-1971 model[a]	39	3.9	2.0
1971–1975[b]	29	2.5	2.3
1976–1988[c]	19.0	2.0	1.7
1989–[d]	4.7	0.7	0.35

[a]Data prepared for the Special Guidance Group (*49*). [b]Egebäck K-E (*50*). [c]Egebäck K-E and Tejle G (*51*). [d]Egebäck K-E and Hedborn A (*52,53*).

investigation were not equipped with exhaust after treatment devices, the question of whether the impact of aromatics will diminish with use of such exhaust after treatment devices remains to be answered.

Methanol and ethanol are two alternative fuels which have been tested in both spark ignition and compression ignition engines. A widely accepted idea is that the use of alcohols as automotive fuels will decrease exhaust emissions. Many investigations have supported this idea, especially those concerning the emission of PAH and NO_x from alcohol-fueled vehicles. The investigations have also shown that emission components such as aldehydes, alcohols, and alkylnitrites will increase to a certain extent. Therefore, all vehicles fueled with methanol or ethanol should also be equipped with an efficient catalyst in order to perform as clean vehicles. Physically, methanol and ethanol are more suitable fuels for ignition combustion engines than for compression ignition engines, since both methanol and ethanol have a high octane number and a low cetane number. To be used in a compression engine, either some type of ignition improver must be used, or the engine must be equipped with a spark plug or glow plug, or be modified in some other way. In addition, it is common that the compression ratio must be increased to take advantage of alcohols as automotive fuels.

Natural gas is used both as a raw material in the methanol production process and as an automotive fuel. Attempts to use natural gas as a fuel for compression ignition engines have not been successful, especially when the fuel has been used as a means to reduce emissions. Natural gas has a low cetane number, so ignition must be supported in some way. In practice, this can be done by using a dual fuel system, in which diesel fuel can support the ignition of the gas. Since natural gas is a clean fuel, there is a great potential in using it in ignition compression engines to reduce harmful emissions. The problem with natural gas is that more emission of nitrogen oxides may occur than when using gasoline. The vehicle should therefore be equipped with a closed-loop fuel system in combination with a three-way catalyst.

Liquefied petroleum gas (LPG) is also used as an automotive fuel. Most of what has been said about natural gas can be applied to LPG. Tests have confirmed that the level of the NO_x emission can be three times higher when using LPG than when using diesel fuel in a heavy-duty engine. On the other hand, the level of the emission of particles, PAH, and some other components was very low, as was the mutagenicity (*37*).

Ignition Compression Engine Development

The technical development of the ignition compression engine and catalyst systems has been very successful. The emission of all except a few components, such as nitrous oxide and hydrogen sulfide, has been reduced to very low levels, as has the associated mutagenicity. The need still remains to take advantage of the potential of the three-way catalyst system; the emission during startup and warmup of the vehicle must be improved, especially the cold-start behavior. The emission standards applied in Sweden, as well as in other European countries and the United States, should be strengthened in order to force car manufacturers to take advantage of the three-way catalyst technology.

Today, the compression ignition engine is the most energy-efficient engine. The problem remains that the emission potential is not as good as for the ignition compression engine. Despite the fact that some important improvements have been reached as a result of engine modifications which so far have been made, much more can be done. The possibilities for environmentally improving the engine are found in further engine modifications, exhaust after treatment devices, and improved diesel fuels or alternative automotive fuels. There is still considerable potential to reduce the emission from heavy-duty vehicles. Methanol, ethanol, natural gas, or LPG are alternative automotive fuels with a certain cleaner emission potential. Ethanol is meant to be a typical biofuel (i.e., it is not manufactured from petroleum products). To take advantage of the emission potential of the alternative fuels, there is a need for extensive development and adaptation of the ignition compression engine and of the compression ignition engine. There is also a need for extensive development of new catalytic converter systems. When developing the engines and exhaust gas after treatment devices, the difference between alcohols and gaseous fuels must be considered.

Conclusions

In this project, the importance of measuring the contribution of semi-volatile associated compounds in automotive exhaust has been confirmed for both gasoline- and diesel-fueled engines. Furthermore, exhaust emissions must be thoroughly characterized, including both regulated and unregulated exhaust emission constituents and bioassay tests. Gasoline-fueled vehicles were investigated at different cruising speeds, and an increased emission of PAH, 1-nitropyrene, particulates, and mutagenic activity was determined at higher cruising speeds. Catalyst-equipped vehicles showed a dramatic decrease of these emission factors when compared with those from the vehicles without catalyst.

Fuel-dependent gasoline exhaust emissions exhibited an increase in PAH (particulate- and semi-volatile) emission in relation to increased fuel PAH content. The increase was linearly dependent, giving a term that can interpreted as the PAH formed in the combustion process. A large proportion of fuel PAH (>95%) is decomposed in the combustion process. Lowering the ambient temperatures will result in an increase of exhaust emissions (both regulated and unregulated pollutants) from engines (cold-start). It is important to consider this fact when performing risk assessment of air pollutants specifically for countries with mean ambient temperatures lower than 20°C. It has been confirmed from a diesel-fuel investigation that there exists a quantifiable relationship between fuel variables of the diesel-fuel blends, and the variables of the chemical emissions and their biological effects. Important diesel-fuel parameters are: density, 90% distillation point, final boiling point, specific energy, total aromatics, di-aromatics, tri-aromatics, and PAH contents.

From multivariate analysis performed, the PLS regression coefficient for regulated pollutants was found to be lower than that for unregulated pollutants; biological activity indicates that improving the quality of the fuel can have a greater impact on the unregulated pollutants in the exhaust emission and the biological effects of constituents in the exhaust. The PLS regression coefficients obtained are most likely different if the vehicles are equipped with exhaust after treatment. A general issue with the use of particulate traps is the need to investigate in more detail the regeneration process with respect to chemical composition and biological effects of the exhaust gases. It has been shown for a particulate trap that an increased nitrogen dioxide emission was measured in modes with increased load and speed compared to no exhaust after treatment.

Our study indicates that there is a need to update emission factors from research and today's engines/vehicles that are alternatively fueled, with respect to both regulated and unregulated exhaust emissions, to be performed in conjunction with bioassay tests. The future engine for heavy-duty vehicles will probably be an Otto-cycle–equipped-engine with a three-way catalyst, or some other type of engine, such as a lean burn, stratified charge or Sterling engine in a hybrid system, especially for city buses. Because of extended research and increased understanding of the health impact of exhaust emission constituents, and the increasing variety of fuels that may be used, the list of regulated pollutants is expected to be extended in the future. Further studies are needed on formed secondary pollutants that originate from mobile sources to estimate future potential environmental and health impacts.

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