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US experience of hydrocarbon and NO_x emissions control

Prepared by the CONCAWE Secretariat based on the paper presented by Dr. L.J. McCabe at the CONCAWE seminar on atmospheric emissions.

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1. FOREWORD

This paper summarises the presentation given by Dr. L.J. McCabe in October 1985 at the CONCAWE Seminar on "Atmospheric emissions and their effects on the environment in Europe with particular reference to the role of hydrocarbons".

It covers the US experience on control of hydrocarbon and NO_x emissions as main precursors to ozone formation, the relevant control technologies, and focuses on the effectiveness of various control strategies and measures.

The original motives for ozone level control in USA and Europe are substantially different (urban ambient concentration of oxidants in California and concern about health effects, while in Europe the concerns mainly come from forest damage) such that the enforcement of USA regulations in Europe may not be appropriate. CONCAWE believes that this paper provides a valid contribution to the debate on the best way to approach similar regulatory measures currently under consideration by some European countries and the EEC.

The paper recalls basic rational principles that are generally agreed and reviews field experience which will help to identify the appropriate path to follow for an effective and well balanced approach to the problem of atmospheric emissions control.

The main issues and associated considerations are:

- Any control policy should be based on sound scientific information. Since rural ozone levels in the US have not been subject to control - as opposed to urban ozone concentrations - much uncertainty surrounds efforts to predict effects of proposed strategies on future rural ozone levels. Appropriate models are in the early development stage both in the USA and Europe and great efforts still seem required before they can help formulating any sensible control policy.

- An effective control strategy should begin with the largest sources. In the USA (as in Europe) the main sources of hydrocarbon emissions are motor vehicles and solvents evaporation (about 80% of total man-made volatile organic compound emissions also in Western Europe). It was for these sources that the USA first enforced control measures. Only after these measures were found not sufficient were other controls considered, such as for refuelling emission control (only 2% of total man-made emissions in USA as well as in Europe).

- The adoption of specific control measures should be preceded by an adequate evaluation of the various technologies available to allow the identification of the most cost-effective option.

To cope with car refuelling emissions, the installation of vapour recovery at service stations (Stage 2) and the alternative of onboard carbon canisters are both under consideration by the EPA. Both systems have pro's and con's, but according to the Author, in the USA carbon canisters appear by far more cost-effective.

It is necessary that European legislators give serious consideration to these alternatives at an early stage. CONCAWE believes that this paper can constructively contribute to the debate.

2. ABSTRACT

Ozone formation in the atmosphere is attributed to complex chemical pathways involving reaction of hydrocarbons and nitrogen oxides in the presence of sunlight. Thus, control strategies to reduce ozone levels involve control of hydrocarbons and NO_x.

This review traces the development of hydrocarbon and nitrogen oxide (NO_x) controls as they relate to the control of high ambient concentrations of oxidants once observed in most US urban areas. First in California, then throughout the US, urban ambient concentrations of oxidants were observed which are associated with alleged health effects. This prompted legislation to control ozone and other ambient air pollutants. In Europe, the motivation for controlling ozone appears to have come from the association of ozone with forest damage at relatively lower rural ozone concentrations. Urban ozone concentrations are similar in Europe and the US; therefore, similar precursor controls should be effective. Since rural ozone levels in the US have not been subject to control, much uncertainty surrounds efforts to predict effects of proposed strategies on future rural ozone levels. Models in both the US and Europe that would handle transport of mostly urban precursor hydrocarbon and NO_x emissions and mixing with rural natural hydrocarbons to predict rural ozone levels are still in the early development stage.

Effective control strategy should begin with the largest sources. In the US motor vehicle emission controls of hydrocarbons and NO_x have been the backbone of ozone control strategies, beginning first in California in the 1960s, then becoming increasingly more stringent as technology was developed. Vehicle hydrocarbon emissions have been second most important for hydrocarbon reduction; controls have involved reduction in use, reformulation, and recovery.

Petroleum industry sources represented only about 10% of uncontrolled hydrocarbons and even less of NO_x. Recently, hydrocarbon emissions have been controlled selectively in high ozone areas. Controls in refineries have included better tank seals and increased monitoring and maintenance of valves and fittings. In gasoline marketing, vapour recovery has been required for transfer operations from bulk tanks to tank trucks and to retail outlets. Vapour recovery in vehicle refuelling is in place in California and is under consideration as a nationwide requirement. Limits on gasoline volatility exist in California and are under consideration in other states and at the national level. Control of stationary source NO_x emissions has been limited to the few areas in violation of health-based standards for NO_x. Limits on refinery boiler and process heater NO_x emissions have been part of the control strategy.

Review of in-place petroleum industry hydrocarbon control techniques indicates that vehicle emission controls have had a larger impact on the petroleum industry through requirements to market unleaded gasoline than direct industry emission controls. Availability of unleaded gasoline was mandated when exhaust emission control catalysts were introduced. The unleaded gasoline mandate also carried restrictions on sulphur and phosphorus content and on usage of oxygenates and fuel additives. The more recent introduction of oxidation/reduction (three-way) catalysts that combine catalytic control of NO_x emissions with control of hydrocarbon and carbon monoxide is bringing pressure for low-phosphorus engine oils.

Concern about the performance of exhaust catalysts in service may bring about a complete ban on leaded gasoline to prevent catalyst poisoning through misuse of leaded gasoline in catalyst-equipped vehicles. Concerns about the effectiveness of evaporative emission controls may lead to gasoline volatility restrictions.

In the United States, regulations to reduce hydrocarbon and NO_x emissions to control ambient ozone have had a substantial impact on the petroleum industry. If further ozone control is necessary worldwide, regulations should take account of cost-effectiveness of alternative control strategies. Any policy must be based on sound scientific information and the application of value judgements that consider key social and economic factors.

3. "US EXPERIENCE OF HYDROCARBON AND NO_x EMISSIONS CONTROL"

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I appreciate the opportunity to be with you today to participate in this very important CONCAWE seminar on atmospheric emissions and acid rain as related to the alleged causes and effects of forest damage. This seminar is very timely, since various legislative control options are being considered in Europe to address the forest damage problem.

As we proceed towards the completion of this seminar, it is important to emphasise that any regulations should be based on sound scientific evidence and should be on a cost-effectiveness basis. Any policy should take into account value judgements that consider key social and economic factors. Research results, such as those presented by Professor Zöttl on the probable causes of forest damage and possible revitalisation of damaged trees, should receive wide distribution, especially to those responsible for developing regulations to control forest damage.

In my presentation, I will discuss the U.S. experience of hydrocarbons and NO_x control, covering the reasons for ozone controls in the U.S., control technology, and the effectiveness of control measures.

In the early 1950's, Professor Arie Haagen-Smit at California Institute of Technology conducted a series of chamber experiments in which he showed that the hydrocarbons and nitrogen oxides in auto exhaust could react in the presence of sunlight to form oxidant or ozone similar to Los Angeles smog. His experiments showed these substances cause eye irritation, crop damage, and rubber cracking, indicative of a highly oxidising medium.

Haagen-Smit recognised that Los Angeles' ozone levels were much higher than could be explained by the photolysis of the available NO_x. He speculated that a free radical mechanism involving the formation of organic peroxides must be involved. We now feel confident that the hydroxyl radical is basic to the understanding of the amplifying effect that hydrocarbons have on ozone formation from NO_x (Fig. 1). These radicals, present in the early morning atmosphere, can be formed from the photoreaction of residual ozone with water vapour, or from the photolysis of nitrous acid formed at night by as yet poorly understood NO_x reactions. The simplified steady-state reactions illustrate ozone formation by NO_2 photolysis, alkyl peroxyl radical formation by hydroxyl radical attack, and radical oxidation of NO to replenish the NO_2. This scheme ignores partial loss of NO_x through nitric acid, peroxyacetylnitrate (PAN) and organic nitrate formation, but illustrates the possibilities fot high levels of ozone formation with little consumption of NO_x.

The pseudo-equilibrium indicated in the photolysis reaction partially explains the apparent depression of ozone formation with increasing NO_x levels at constant HC levels.

As illustrated by Dr. Cox, organics can be ranked as to their ozone-forming potential roughly in proportion to the rate of reaction with hydroxyl radicals. Such a ranking has formed the basis for some strategies for reformulation of solvents to provide less reactive emissions.

Throughout the 1960's, first in California and then through Congressional action, controls were imposed on automotive emissions and, to a much lesser degree, on some petroleum sources of hydrocarbon vapours. Enforcement of other than new car controls was lax, and time tables for achieving emissions reductions were essentially non-existent. In 1970 and 1977, riding a wave of public sentiment for environmental legislation, Congress passed amendments to existing environmental laws mandating EPA to set national health-based air quality standards. EPA was given the authority to withhold federal funding for state projects and prohibit new construction, if the States failed to meet the standards. In the 1970 Amendments to the Clean Air Act, Congress mandated reduction of automotive emissions by 90% over the 1970 levels.

EPA set the ozone standard at 0.08 ppm (1-hr average) in 1971 based on hastily assembled support data and relaxed the standard by 50% to 0.12 ppm in 1979 on the basis of a more thorough examination of health effects observed at low ozone concentrations. This action was precipitated by an API lawsuit. To achieve these levels, EPA advised that peak morning hydrocarbon reduction was the most effective control strategy. This, coupled with NO_x reductions through automotive emissions control, would achieve the desired ozone levels.

As shown in Fig. 2, early 1970's ozone levels in the U.S. were not too different from those currently observed in European urban areas. Since transport from urban areas is currently felt to be the largest source of rural ozone, although sporadic stratospheric ozone may contribute to peak levels, presumably control strategies which work in the U.S. will work in Europe.

In 1979, the U.S. EPA recognised that both hydrocarbon and NO_x concentrations influence ozone formation and that the HC/NO_x ratio is very important. Fig. 3 illustrates how ozone levels are dependent on both hydrocarbon and NO_x concentrations. A slight reduction in NO_x may require a substantial reduction in hydrocarbons in order to reduce ozone from 0.2 ppm to 0.12 ppm; thus the hydrocarbon-NO_x levels must be balanced.

An effective ozone control strategy should begin with control of the largest source of precursor emissions. Fig. 4 illustrates the contribution of major U.S. hydrocarbon sources as a percent of total 1970 hydrocarbon emissions. In 1970, about 40% of the total hydrocarbons were from motor vehicles (highway transportation). By

1982, motor vehicle hydrocarbon sources had been reduced to about 22% of the total hydrocarbons. The second major source was solvent evaporation, which was reduced from 23% of total hydrocarbons in 1970 to 15% in 1982 by reduction in the use of solvents, reformulation, and solvent recovery. Petroleum sources were only 10% of the hydrocarbon burden in 1970 and thus there is little nationwide control of petroleum sources. However, in high ozone areas, petroleum source hydrocarbon control is extensive.

About 50% of the NO_x emissions in the U.S. have been due to power generation (Fig. 5). In 1970, about 40% of the NO_x emissions resulted from motor vehicles and the contribution increased by 1982 due to the increase in vehicle miles travelled. Petroleum sources are a minor contribution to NO_x emissions. Overall, NO_x emissions have not been controlled as effectively as hydrocarbon emissions because of the unavailability of cost-effective NO_x control technology.

As noted above, motor vehicles are major sources of hydrocarbon and NO_x emissions. Major milestones in U.S. motor vehicle emissions controls are shown in Fig. 6. Crankcase hydrocarbons and carbon monoxide were controlled by use of positive crankcase ventilation (PCV) valves, and in 1968 hydrocarbons were reduced further by engine detuning and use of air pumps to promote oxidation. In 1970, controls were implemented to control evaporative hydrocarbon emissions resulting from diurnal heating of the fuel in the vehicle tank and evaporation losses from the carburetor after the engine is heated up and allowed to stand. Evaporative emissions have been controlled by installing a carbon canister onboard the vehicle to collect the vapours and then purging the vapours to the combustion process. In 1972, exhaust gas recirculation was introduced to reduce vehicle NO_x emissions by reducing peak combustion temperatures.

A major step in control exhaust hydrocarbons and carbon monoxide was the introduction of noble metal oxidation catalysts on vehicles in 1975, requiring the use of unleaded gasoline. Three-way catalysts were introduced nationwide in 1981 to effect better control of NO_x emissions. Meeting the evolutionary stringent vehicle emissions standards required a substantial effort by both the automotive and petroleum industries. There was give and take on both sides, resulting in a substantial impact on both industries. The most significant impact on the petroleum industry was the requirement of unleaded gasoline on catalyst-equipped cars.

Motor vehicle emissions controls have been effective. Trends in U.S. mobile source emissions and vehicle miles travelled (VMT) are shown in Fig. 7. These are plotted as percent of 1970 emissions for the period 1970-1982. From 1970 to 1982, vehicle miles travelled increased by about 35%. For the same period, hydrocarbon emissions decreased substantially. Nitrogen oxide emissions increased during this period reflecting the increase in vehicle miles travelled. But we are now seeing a downward trend as a result of the implementation of NO_x controls.

The next largest source of hydrocarbon emissions is solvents. Solvent sources and controls are illustrated in Fig. 8. Controls include reduced solvent content, reformulation, and solvent recovery.

Although petroleum sources are a minor contributor to the hydrocarbon burden, there has been selective control in areas of ozone standard non-attainment. Production, storage, and refinery process sources and controls are shown in Fig. 9. These controls are very costly. Control of fugitive emissions in the refinery is especially burdensome and not very cost-effective because of the manpower requirements for daily monitoring and extensive reporting requirements.

There are also substantial and costly controls for hydrocarbons in the marketing of gasoline as shown in Fig. 10. At gasoline terminals, vapours are controlled mainly by condensation and incineration. At terminals, gasoline tank truck vapours are controlled by vapour recycling to the terminal and by bottom loading. In bulk delivery at the service station, the vapours displaced from the service station tank are recycled to the tank truck and subsequently disposed of at the bulk terminal. This is referred to as Stage I service station control.

One source of hydrocarbon emissions which has not been widely controlled up to now is vehicle refuelling emissions at service stations. The U.S. EPA is weighing the merits of controlling refuelling emissions nationwide. Two alternative control technologies are being considered: vapour recovery at the service station pump (Stage II vapour recovery) and vapour collection on the vehicle, designated onboard refuelling emission control. The State of California and Washington, D.C., have already adopted Stage II refuelling emissions controls, and other states are considering doing so as a step toward meeting ozone standard. Hydrocarbon emissions from refuelling gasoline vehicles account for only about 2% of total hydrocarbon emissions in the U.S. For European vehicles, with less severe exhaust emission regulations and presently no evaporative emission control requirements, refuelling emissions represent a smaller fraction of vehicle hydrocarbon emissions.

Without controls, when gasoline is transferred into a vehicle tank, vapours are displaced and vented into the atmosphere. Service station vapour recovery systems (Stage II) are designed so that the vapour displaced during refuelling flows back into the station storage tank being emptied. Liquid and vapour volumes remain closely in balance so that little vapour is ever vented from the system. A Stage II system is illustrated in Fig. 11. Experience with Stage II controls in the U.S. shows that customers have difficulty using the equipment, that vapour-capture components of the nozzles have limited durability, and that regular maintenance is needed to ensure that the equipment operates with the expected efficiency.

The feasibility of Onboard control of refuelling emissions has been demonstrated by Mobil and Exxon in work sponsored by API. The essential components of the system are a seal in the fill-pipe to prevent escape of vapours, a canister containing activated carbon to absorb the fuel vapours, a means to regenerate the canister by desorbing or purging vapours during driving, and a suitable value in the vapour line between the fuel tank and canister to prevent transfer of liquid fuel to the canister or leakage in event of vehicle roll-over. A schematic of the Mobil Onboard system is shown in Fig. 12. The fill-pipe seal shown is a liquid seal similar to the vapour trap in a common sink drain and that has proven to be very effective. The carbon canister for the Onboard system was about three times the volume of that used on the production vehicle to control evaporative emissions. The Onboard system serves the dual purpose of controlling evaporative emissions. Canister purging was controlled by modifying the production vehicle's electronic control systems. A solenoid valve is opened at specified engine operating conditions. For the Mobil Onboard car, the roll-over valve (Fig. 12) was designed to be closed, except during vehicle refuelling. A vapour/liquid separator was also added to prevent carry-over of liquid fuel droplets with vapour to the canister during refuelling.

The results of the studies by Mobil and Exxon have shown that a vehicle Onboard system can control refuelling emissions with greater than 98% efficiency; control of evaporative emissions was improved to the extent that the present standard of 2.0 g/test prescribed for a 9 psi RVP certification fuel was met with a fuel of 11.5 psi RVP, typical of commercial gasoline; and the Onboard system had no adverse effect on exhaust emissions. The Onboard system is compatible with existing gasoline dispensing nozzles. It is simple in design and construction and should be readily adaptable to common automotive production methods and materials. Analysis of long-term cost and effectiveness show that the Onboard technology is more cost-effective than service station Stage II control and Onboard systems can control substantially more hydrocarbons than Stage II controls, since Onboard systems will also control evaporative emissions.

Vehicle emissions controls have had substantial effects on the petroleum industry (Fig. 13). The introduction of noble metal exhaust catalysts mandated the marketing of unleaded gasoline. To meet octane requirements, gasoline pool octane must be increased. The capital costs for this are not recoverable due to the current oversupply of gasoline. Oxygenate extenders are of limited utility due to limitations imposed by emission control systems.

The use of catalysts has also resulted in restricted fuel sulphur and phosphorus levels and in additive usage. In addition, autobuilders are pressing for low phosphorus engine oils. Stringent evaporative emissions standards have resulted in proposals for more controls on gasoline volatility.

The effectiveness of hydrocarbon and NO_x control strategies on U.S. urban ozone trends is illustrated in Fig. 14 for the period 1979-1984 for some major U.S. cities. The second daily high ozone level (ppm) is shown for each year. While there are excursions in ozone due to meteorological factors, in general there is a downward trend in ozone levels. The progress to date has been moderated by lack of reliable models to predict precursor effects on ozone formation. We would also point out that an ozone standard based on a daily peak level is impractical and the standard should be based on averaging techniques.

Acknowledgement

The contributions of Dr. W.J. Koehl and Dr. S.S. Wise in preparing this paper are gratefully acknowledged.

4. SUMMARY

- U.S. began to control ozone because of health concerns at urban ambient concentrations.
- Europe is proposing controls because of forest damage at relatively lower rural concentrations.
- Urban ozone concentrations are similar in Europe and U.S.; similar controls should be effective.
- Effective control strategy should begin with the largest sources.
- In U.S., motor vehicle emission controls have been the backbone of ozone control strategies.
- Solvent evaporation control has been second most important for HC reduction.
- Petroleum industry sources represented only about 10% of uncontrolled HC and even less of NO_x. They have been controlled selectively in high ozone areas.
- Vehicle emission controls have had a larger impact on the petroleum industry through unleaded gasoline than direct industry emission controls.
- Regulations to reduce hydrocarbon and NO_x emissions have had a substantial impact on industry.
- Future regulations should be based on sound-scientific evidence and take account of cost-effectiveness of alternative control strategies.

FIGURE 1

HYDROCARBON ENHANCEMENT IN OZONE FORMATION

Simplified Initiation:

$$O_3 + \text{Light} \xrightarrow{[H_2O]} 2OH^{\cdot} + O_2$$

Simplified Steady-State:

$$2NO_2 + \text{Light} \overset{[O_2]}{\rightleftharpoons} 2NO + 2O_3$$

$$RH + OH^{\cdot} \xrightarrow{[O_2]} RO_2^{\cdot} + H_2O$$

$$RO_2^{\cdot} + NO \xrightarrow{[O_2]} R'CHO + HO_2^{\cdot} + NO_2$$

$$HO_2^{\cdot} + NO \Longrightarrow OH^{\cdot} + NO_2$$

- -

Simplified Stoichiometry:

$$RH \xrightarrow{[O_2]} R'CHO + H_2O + 2O_3$$

FIGURE 2
EUROPEAN VS U.S. OZONE LEVELS

	Range			
	50%	98%	99%	Max
Europe (22 Cities) (1979-1981)	4-45 ppb	14-145 ppb		42-489 ppb
U.S. (33 Cities) (Non-California 1971)	0-35 ppb		10-120 ppb	60-360 ppb
U.S. (California 1971)	10-40 ppb			50-530 ppb

FIGURE 3
OZONE CAN BE CONTROLLED THROUGH BOTH HYDROCARBONS AND NO x

STANDARD EKMA OZONE DIAGRAM

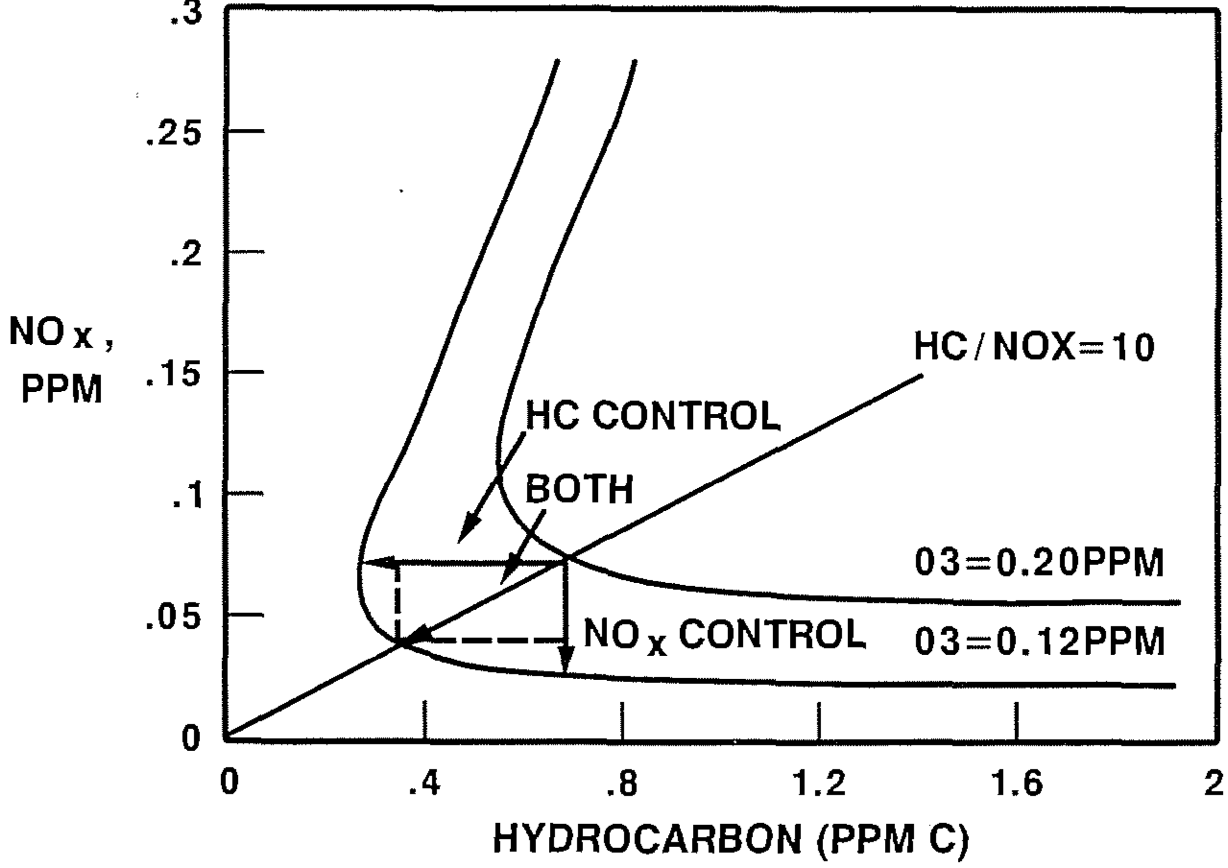

FIGURE 4

1970 / 1982 U.S. HC EMISSIONS

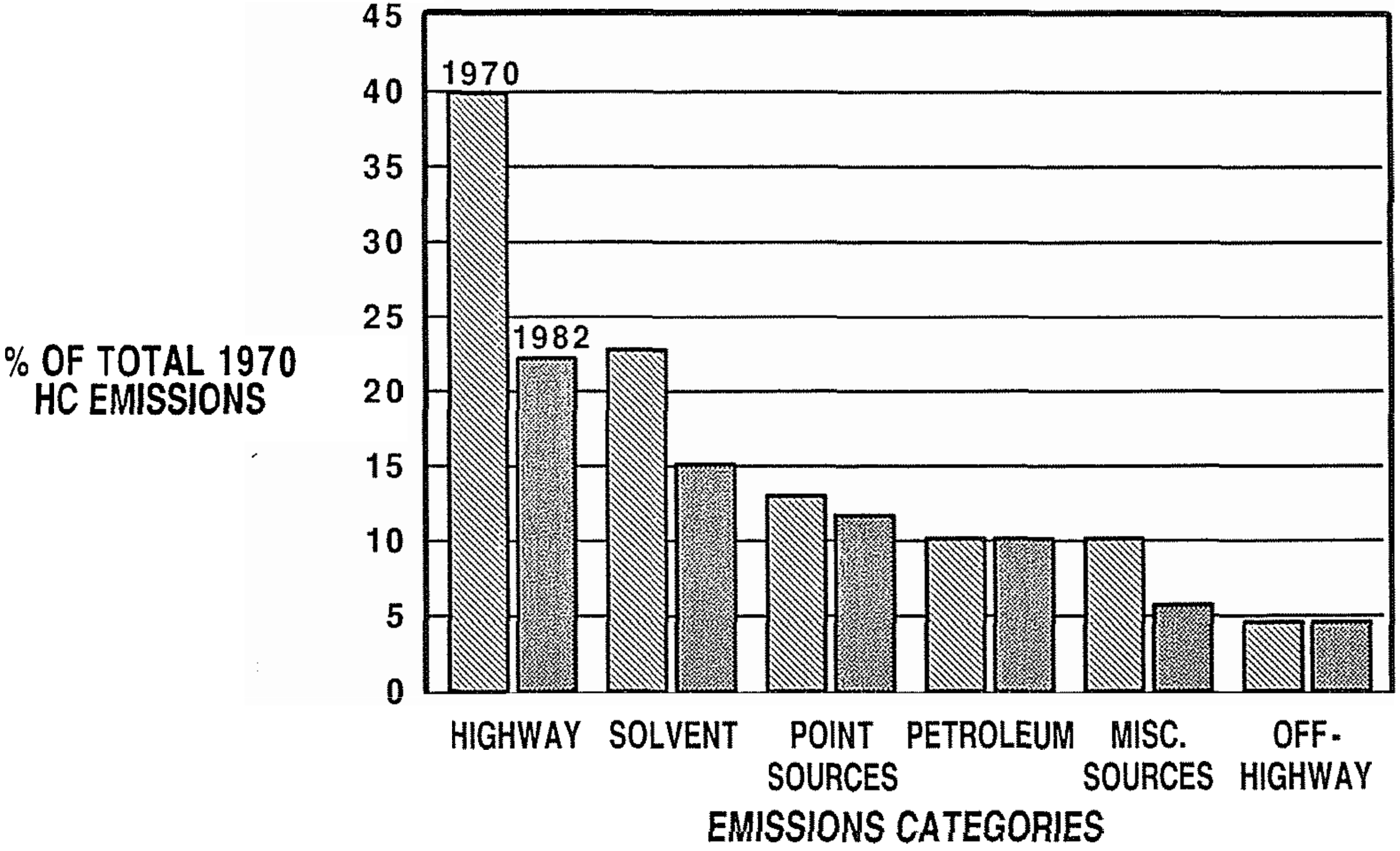

FIGURE 5
1970 / 1982 U.S. NOX EMISSIONS

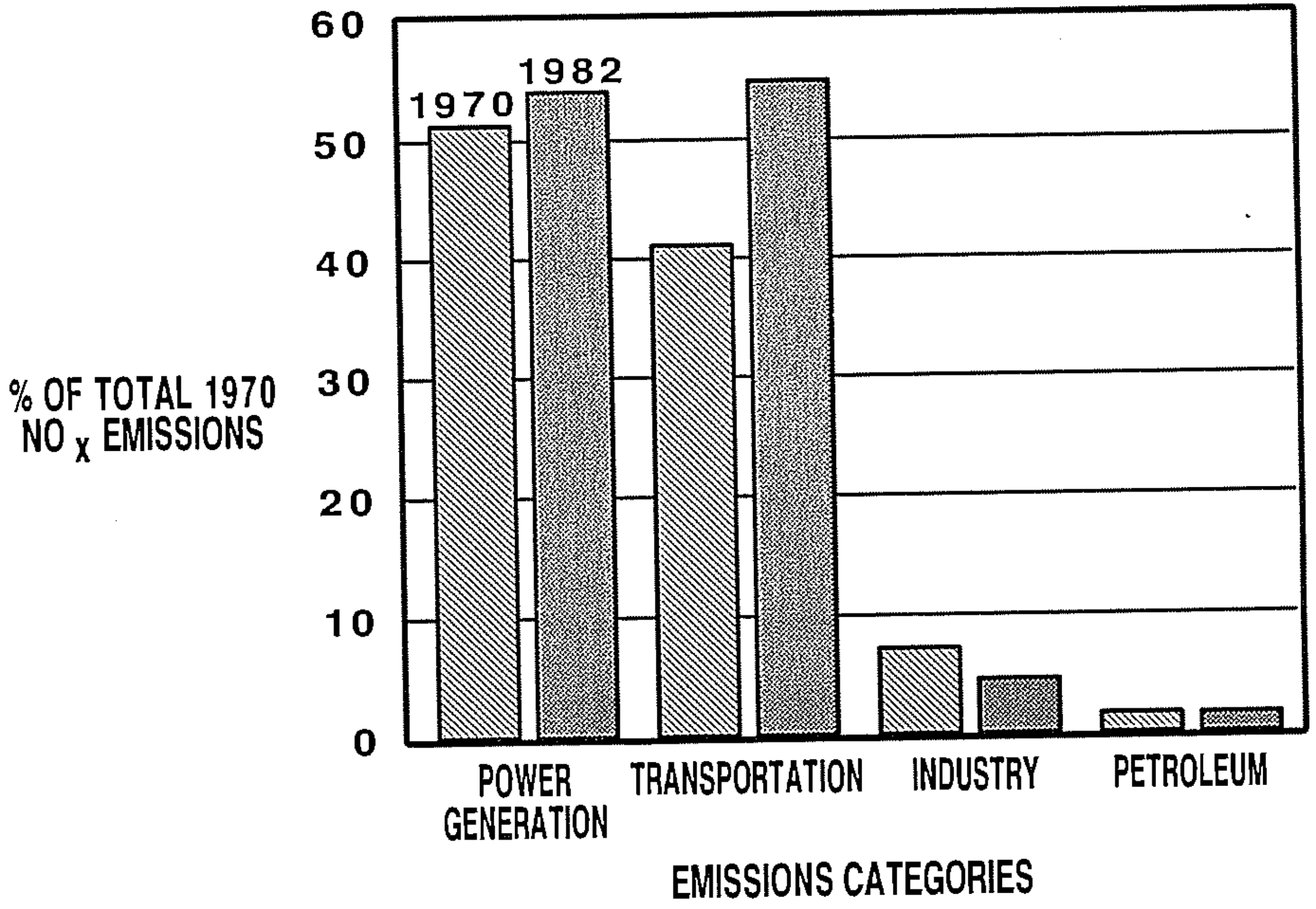

FIGURE 6

U.S. MOTOR VEHICLE EMISSIONS CONTROLS MAJOR MILESTONES

Source	Controls
1963 - Crankcase HC & CO	PCV Valve Blowby to Intake System
1968 - Exhaust HC	Engine Detuning & Air Pumps
1970 - Evaporative HC	Charcoal Canisters
1972 - Exhaust NO_x	EGR
1975 - Exhaust HC & CO	Oxidation Catalysts & Unleaded Gasoline
1981 - Exhaust NO_x	Oxidation - Reduction (3-Way) Catalysts

FIGURE 7
TRENDS IN U.S. MOBILE SOURCE EMISSIONS AND VMT

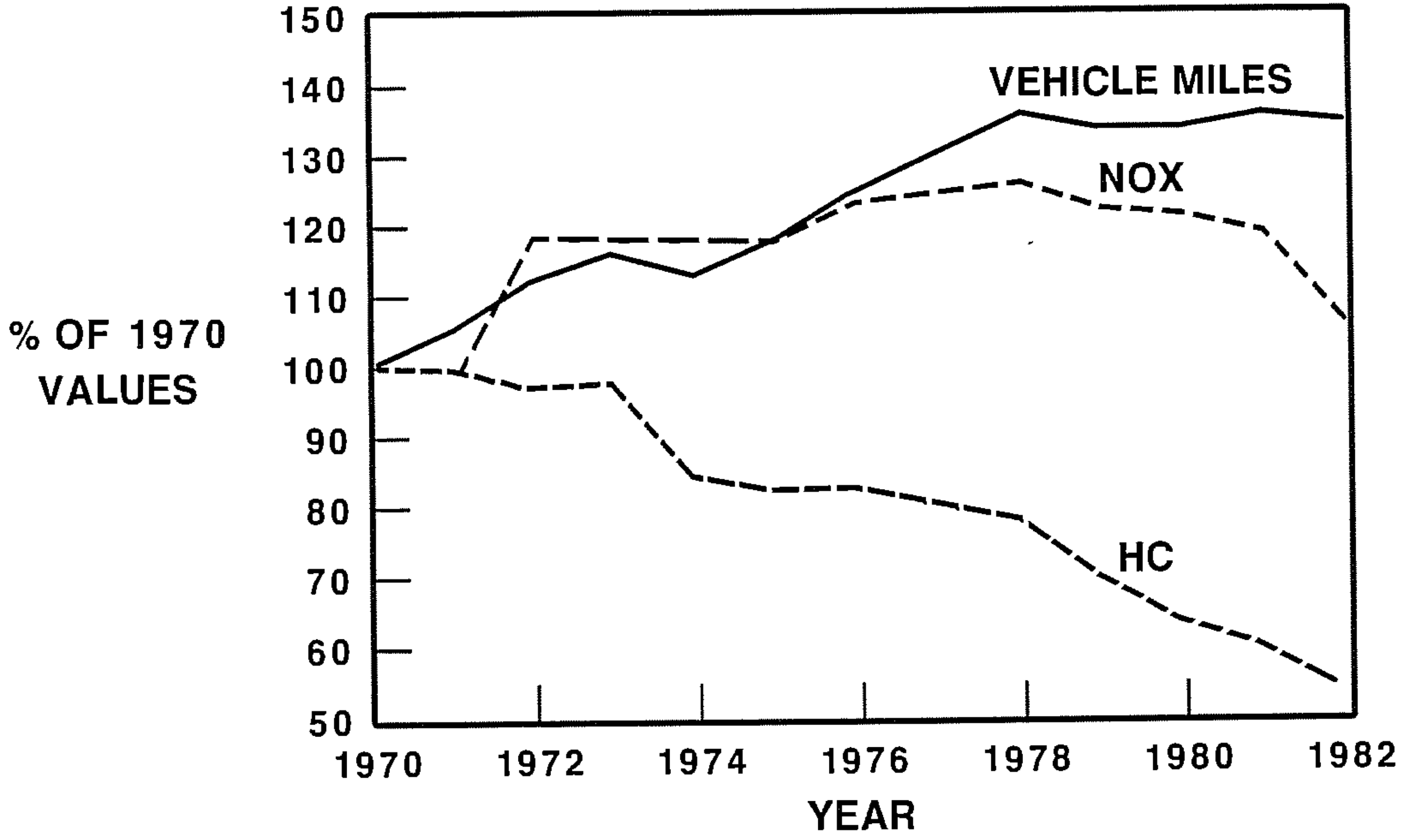

FIGURE 8

CONTROLS ON SOLVENT USAGE

Source	Controls
Asphalt Emulsions	Reduce Solvent Content
Printing Inks	Reduce Solvent Content, Solvent Recovery
Automotive Paints	Water Emulsions, Reduced Solvent Content, Solvent Recovery
Furniture Paints	Reformulation
Exterior Paints	Water Emulsions, Reduced Solvent Content
Dry Cleaning Solvents	Solvent Recovery
De-greasing Solvents	Solvent Recovery, Low RVP Solvents

FIGURE 9
PETROLEUM INDUSTRY HYDROCARBON SOURCES SUBJECT TO CONTROL

Source	Controls *
Production & Storage	
Fixed Roof Tanks	Vapor Recovery & Internal Floater
Floating Roof Tanks	Secondary Seals
Fugitive Emissions	Periodic Monitoring & Housekeeping
Refinery Processes	
Vacuum Producing Systems	Hot Well Covers & Vapor Incineration
Wastewater Separaters	Cover
Process Unit Turn-Arounds	Vapor Holding & Recovery
Fugitive Emissions	Periodic Monitoring & Housekeeping

* Required as of 1982 in all U.S. ozone nonattaiment areas, but applied earlier in some states.

FIGURE 10
MARKETING HYDROCARBON CONTROLS

Source	Controls
Bulk Gasoline Terminals *	Condensation, Incineration
Gasoline Tank Trucks *	Vapor Recycling to Terminal & Bottom Loading
Service Station	
● Bulk Delivery *	Vapor Recycling to Tank Truck (Stage I)
● Vehicle Refueling **	Vapor Capture at Station or on Vehicle

* Controls required as of 1982 in all U.S. ozone nonattainment areas, but applied earlier in some states.

** Pending in U.S. EPA, but service station controls used in California since 1974.

FIGURE 11

STAGE II

BALANCED DISPLACEMENT SYSTEM

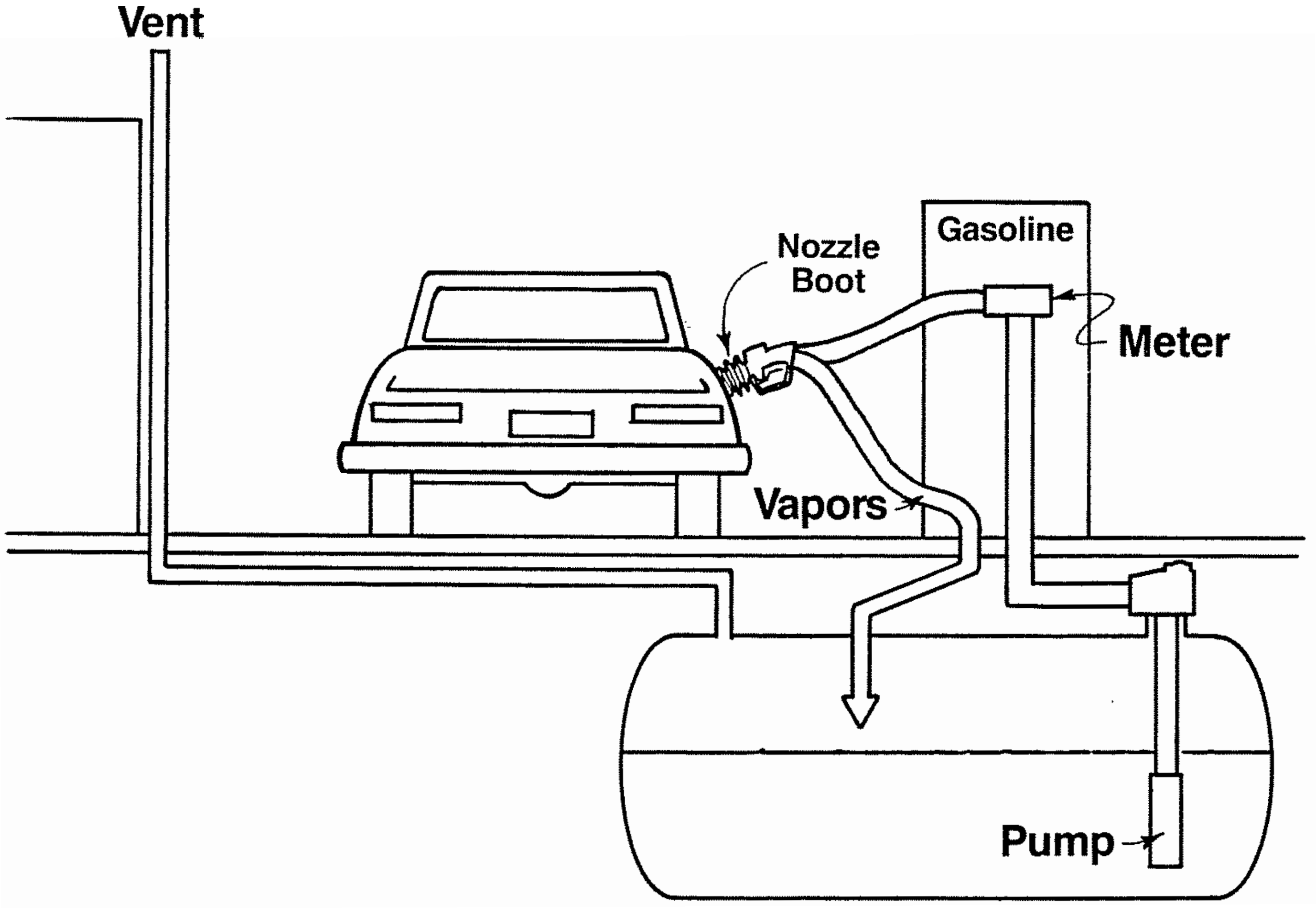

FIGURE 12

ONBOARD REFUELING EMISSION CONTROL SYSTEM
1985 BUICK CENTURY

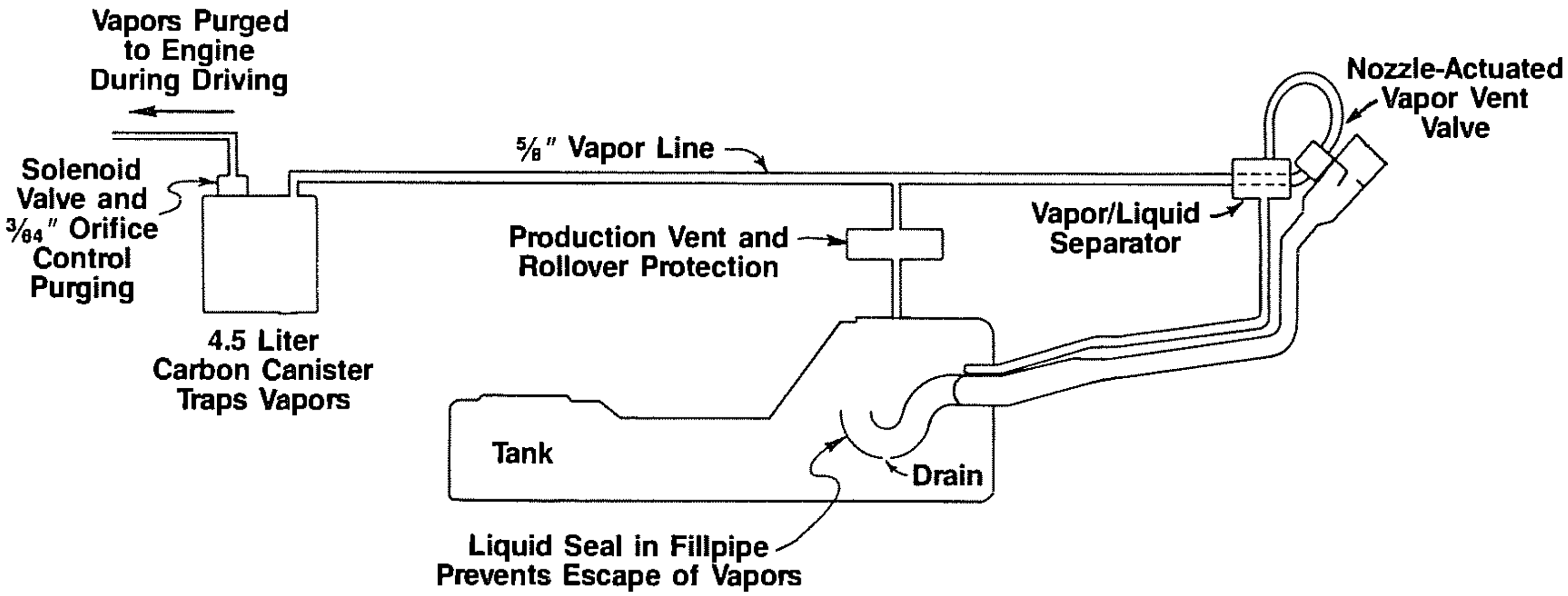

FIGURE 13

VEHICLE EMISSIONS CONTROLS AFFECT INDUSTRY

- Oxidation Catalysts
  - Unleaded Gasoline Mandated
  - Fuel Sulfur Level Restricted
  - Fuel Phosphorus Restricted
  - Additive and Oxygenate Usage Restricted
- Oxidation / Reduction Catalysts
  - Auto Builders Pressing for Low Phosphorus Engine Oils
- Evaporative Controls
  - Gasoline Volatility Limits Proposed

FIGURE 14
1979-1984 U.S. URBAN OZONE TRENDS