COMMISSION OF THE EUROPEAN COMMUNITIES

environment and quality of life

NITROGEN OXIDE EMISSIONS FROM ENERGY
GENERATION WITHIN THE EEC : 1970 – 1985

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1. SUMMARY

The most recent estimate of world-wide NO_x emissions from all sources is 540M tons p.a. of NO equivalent, of which 110M tons p.a. arises from pollution sources and the remainder from natural processes. NO is formed by the combination of atmospheric nitrogen and oxygen at the high temperature occurring in combustion flames, and this is further oxidised to NO_2 when the exhaust gases are diluted on release into the open air. At the point of emission, NO_x consists of a mixture of NO and NO_2, a typical ratio being 10 to 1 by volume. The conditions which favour formation of NO_x are high temperature and long residence time of the burning gases in the high temperature zone.

Emission factors (weight of NO_x formed per unit weight of fuel consumed) vary considerably, not only with type of fuel and type of combustion unit, but also between apparently identical units burning the same fuel. Quoted emission factors can therefore only be mean or typical values, and considerable variation is to be expected, depending on the mode of operation. This is particularly true of mobile sources, where engine load conditions are constantly changing.

However, using the emission factors given in Section 3 of the report, together with estimated fuel consumption in various sectors for the community member countries, it is concluded that the total NO_x emission in 1970 within the Community amounted to approximately 8M tons. On the basis of the predicted fuel growth pattern used in this report (Section 5) the total NO_x emissions are expected to rise to around 13M tons in 1985. Analysis by sector of fuel use shows that in 1970 general industry accounted for one third of the emissions, but by 1985 the largest emitting sector is predicted to be transport (again one third of the total). Emissions from transport sources would double between 1970 and 1985, assuming no controls. Analysis by type of fuel shows that in 1970 liquid fuels accounted for 60% of all emissions and this would grow to around 80% by 1985. These are mean values which are subject to considerable uncertainty, and of course vary if the emission factors and quantities of fuels consumed vary from the assumed figures. No account is taken

of the current oil crisis.

It must not be presumed that the importance of a particular source, so far as damaging effects are concerned, is related only to the quantity of NO_x emitted. It may·be that the manner of emission (e.g. whether at high or low level) is also an important factor, since it is this which often determines the resulting ground level concentration of NO_x. The dispersion and consequential affects of NO_x emissions are outside the scope of this report, but it should be borne in mind that any priorities for control policies ought to take into account the likely damage (if any) caused by NO_x levels from different sources.

The control technology for NO_x emissions is in its infancy, and much proving of techniques under real-life conditions remain to be done. For stationary sources, although it is doubtful if there are commercial plant operating with NO_x control anywhere at the present time, modification of combustion conditions or furnace design appear to hold most promise, and a reduction in NO_x emissions of 50% seems feasible with current technology, as applied to oil-or gas-fired plant. However coal-fired plant present problems where control of fuel/air ratios is called for. Greater reduction of NO_x is theoretically possible using catalysts, but a successful technique remains to be developed. For mobile sources, exhaust gas recirculation is already practised on automobiles in the USA. This reduces NO_x emissions by 50%. Again, further reduction could be achieved by catalytic means but a successful practical device has not yet been produced. However, indirect injection diesels, Wankels, and stratified charge engines all have lower NO_x emission levels. The ultimate answer to NO_x (and other) pollution from mobile sources may be to change the propulsion unit entirely - say to an electric drive, but with a penalty to overall fuel usage efficiency.

Reliable costs for the various methods of NO_x control are very difficult to obtain, and those quoted in Section 7 are based in the main on design studies and limited experimentation.

Table 1 summarizes the current state of knowledge existing on the topics covered later in the report.

Table 1

Current State of Knowledge on No$_x$ Topics

Topic	State of Knowledge
NO$_x$ Formation Processes	Details of reactions occurring in flames during formation of NO poorly understood. Theory underestimates actual formation rates
Emission Factors	Wide variability, even between apparently identical plant
NO$_x$ Emissions	Accuracy of forecasts depends on accuracy of: (1) Emission factors (see above) (2) Estimates of fuel consumption – current energy crisis introduces large element of uncertainty here
Control of NO$_x$ Emissions (1) Stationary Sources	(1) Theoretical methods understood. Practical application lacking
(2) Mobile Sources	(2) Exhaust gas recirculation practised in USA. Technology for high degree of control not yet established, but indirect injection diesels, Wankels, and stratified charge engines all give lower NO$_x$ emissions
Economics of No$_x$ control (1) Stationary Sources	(1) Idealised estimates exist (US origin) but require confirming under practical conditions
(2) Mobile Sources	(2) Some cost data exist for exhaust gas recirculation

2.   INTRODUCTION

The scope of this report is to consider a number of aspects of the emission of nitrogen oxides (NO_x) within the EEC from sources of energy generation. Thus the succeeding sections deal in turn with the following topics:

(a) The mechanism of formation of NO_x during the combustion of fuels;

(b) The emission factors for NO_x production from various fuels and types of combustion system;

(c) The estimated quantities of NO_x emitted from different classes of source, projected up to 1985 on the basis of forecast fuel consumption for the EEC;

(d) The methods for the control of NO_x emissions, both currently available and anticipated;

(e) The cost of the various methods of NO_x control.

Within recent years there has been a considerable increase in the interest shown towards NO_x emissions as a contribution to general air pollution. This upsurge of interest derives mainly from the part played by NO_x in the formation of photochemical smog, which has become especially notorious in the Los Angeles district of the United States. However it is not only in the formation of photochemical smog that NO_x poses a potential hazard. With the exception of the substantially inert nitrous oxide, N_2O, (which is not formed during the combustion process), all oxides of nitrogen are acidic and toxic at concentrations of several ppm. The principal oxide formed when fuels are burnt in air is nitric oxide, NO; this further reacts with oxygen to give nitrogen dioxide, NO_2. It is the sum of these two oxides which is conventionally represented by the term NO_x.

Because of the relatively recent attention which NO_x pollution has attracted, comprehensive data on air concentration levels are generally not available. In addition, the extent of NO_x formation depends critically on the precise conditions of temperature, fuel/air contact time, and fuel/air ratios which exist during combustion. All these can vary considerably, not only from one fuel to another, but also within a given fuel-burning appliance, and estimates of emission factors (weight of NO_x

emitted per unit weight of fuel burned) can show correspondingly wide variations.

The presence of these factors necessarily results in considerable uncertainty in any estimates which are made, either of overall NO_x emissions or typical ambient air levels in industrial environments. Nevertheless, although much investigational work remains to be done on all aspects of NO_x pollution, there is sufficient information available to enable a broad picture of the emission situation to be built up, both current and projected.

World-wide NO_x emissions have been estimated by Robinson and Robbins (1968), to be of the order of 270M tons p.a. (expressed as equivalent NO). Of this, only 35M tons p.a. arise from pollution sources, the remainder being generated as NO in the bacteriological decomposition of organic matter in soil. NO_x is also produced in nitrogen fixation by lightning, but only to a marginal extent (Junge, 1963).

Table 2 gives a breakdown of the world-wide NO_x emissions from pollution sources, as estimated by Robinson and Robbins, together with the emissions from within the EEC, as estimated in the present report.

Table 2

Comparison of Worldwide and EEC NO_x Emissions from man-made sources[a] (expressed as NO)

Fuel and Source		NO_x Emissions (M tons NO)	
		Worldwide[b] (1967)	EEC (1970)[e]
Coal:	Power Generation	8.0	1.4
	Industry	9.0	0.9
	Domestic/Commercial	0.7	0.2
Petroleum:	Refinery Production	0.5	- [c]
	Gasoline	4.9	2.2
	Kerosene	0.8	0.5
	Fuel Oil	2.3	1.4
	Residual Oil	6.0	0.6
Natural Gas:	Power Generation	0.4	0.1
	Industrial	0.7	0.2
	Domestic/Commercial	0.3	0.1
Others:	Incineration	0.3	- [d]
	Wood Fuel	0.2	- [d]
	Forest Fires	0.5	- [d]
Total		34.6	7.6

Notes (a) Virtually entirely from combustion processes. NO_x emissions from chemical plant can be ignored in this context.
(b) Source: Robinson and Robbins (1968).
(c) Included in figure for fuel oil.
(d) Not included.
(e) For the 9 EEC member countries, as calculated in section 5.

Although the emission estimates in Table 2 refer to slightly different years for the World and EEC, it is nevertheless interesting to note that on the basis of the above figures, Community countries account for around 20% of the world NO_x emissions. In both cases, around 5% of the total arises from natural gas combustion, while the EEC emits proportionally more NO_x from liquid fuels than the world as a whole. It would be unwise to place too much emphasis on the absolute values, especially since in a supplemental report, Robinson and Robbins (1971) revised their estimates upwards to 540M tons p.a. of equivalent NO total world emissions including 110M tons p.a. from pollution sources. They give no reason for this.

Finally, Table 3 compares the mean NO_x emissions from EEC sources in 1970 and in 1985, assuming no controls.

Table 3
Comparison of EEC[a] NO_x emissions in 1970 and 1985

Fuel	NO_x emissions (M tons NO)	
	1970	1985
Solid	2.5	1.7
Liquid	4.7	10.3
Gas	0.4	1.0
Total	7.6	13.0

Note (a) For the 9 EEC member countries, as calculated in section 5.

The total emissions are likely to have risen by 70%, although, reflecting decreasing usage, emissions from solid fuels could well fall by 30%. On the other hand emissions from both liquid and gaseous fuels will probable have more than doubled by 1985, the former by then accounting for three-quarters of all emissions. These conclusions are based on the fuel usage assumptions given in Section 5, and do not reflect the current oil crisis.

3.   FORMATION OF NO_x DURING COMBUSTION

At least six stable oxides of nitrogen are known, together
with one unstable compound. They are:

Nitrous Oxide, N_2O	Nitrogen Dioxide, NO_2
Nitric Oxide, NO	Nitrogen Tetroxide, N_2O_4
Nitrogen Sesquioxide, N_2O_3	Nitrogen Pentoxide, N_2O_5

and the unstable   Nitrogen Trioxide, NO_3.
The only oxides which are important as air pollutants are Nitric
Oxide and Nitrogen Dioxide, the basic mechanism of their formation
being the same for all forms of combustion, both for stationary
and mobile sources. At the high temperatures occurring during
the combustion of fuels in air, the nitrogen and oxygen present
react together to form NO almost exclusively. This is a colourless
odourless gas, slightly soluble in water, the physical properties
of which are not clearly understood. The formation of NO takes
place according to a number of different reactions between N_2 and
O_2, which may be simply represented as:

$$N_2 + O_2 \rightleftharpoons 2NO$$
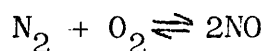
The final concentration of NO is dependent upon a number of factors:

(a) The concentrations of nitrogen and oxygen. The higher
the concentrations of O_2, (excess air conditions), the
more NO that is present at equilibrium;

(b) The temperature of combustion. Since the activation
energy of the reaction between N_2 and O_2 is high, the
higher the temperature, the higher the concentration of NO;

(c) The residence time of the gases in the various zones of
combustion. Longer residence times at high temperatures
tend to produce higher NO concentrations. Furthermore
when rapid cooling, say in an exhaust system, follows
combustion, the reverse reaction (dissociation of NO)
takes place only slowly and NO persists at higher concen-
trations than if slow cooling were to be permitted.
Rapid quenching often occurs after combustion in flames,
so that the level of NO in exhaust gases vented to the
atmosphere is more likely to be governed by conditions at
flame temperatures than by lower intermediate cooling or
ambient temperatures;

(d) The pressure of the reacting system. At higher pressures the maximum equilibrium concentration of NO is raised. Thus at 1 atm. the maximum equilibrium molar concentration of NO is 5.2% (achieved at $3000^{o}K$), while at 0.01 atm. the maximum concentration falls to 2.2% (Bagg 1971). (This result appears surprising from a thermodynamic point of view);

(e) The nature of the surfaces with which the reacting gases are in contact during combustion.

The full details of all the mechanisms involved in the formation of NO in flames are not completely known. In some cases much higher concentrations of NO are produced than would be expected from considerations of theoretical kinetics, the most likely reason being the formation of free radicals and atoms in greater than equilibrium concentrations.

It is generally assumed that NO_2 is produced by the further oxidation of NO with excess oxygen*, the simplest way of representing this being:

$$2NO + O_2 = 2NO_2$$
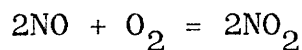
Although NO_2 exists in equilibrium with its dimeric form N_2O_4, the fraction of the latter existing at atmospheric concentrations of NO_2 can be neglected. NO_2 is a reddish-orange gas with a characteristic pungent odour; it is corrosive and highly oxidising. Physiologically it is both irritating and toxic at concentrations of a few ppm.

Figure 1 shows the equilibrium composition of air at various temperatures, from which it can be seen that NO_2 is present only as a very minor constituent at flame temperatures, $(2000^{o}-3500^{o}K)$ and immediately after combustion NO is by far the dominant oxide of nitrogen present in the gas steam. The pressure is 1 atm.

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*There is some recently reported evidence (Merryman E.L. and Levy A. "NO_x formation in combustion: Anomalous early NO_2 formation".^{x} Proc. 3rd Intl. Clean Air Congress, Düsseldörf, October 1973, C117) that NO_2 might be formed before the appearance of NO in some flame systems. However even the authors approach this observation with much scepticism.

Figure 1

Equilibrium composition of air at various temperatures

(Source: Bagg, 1971)
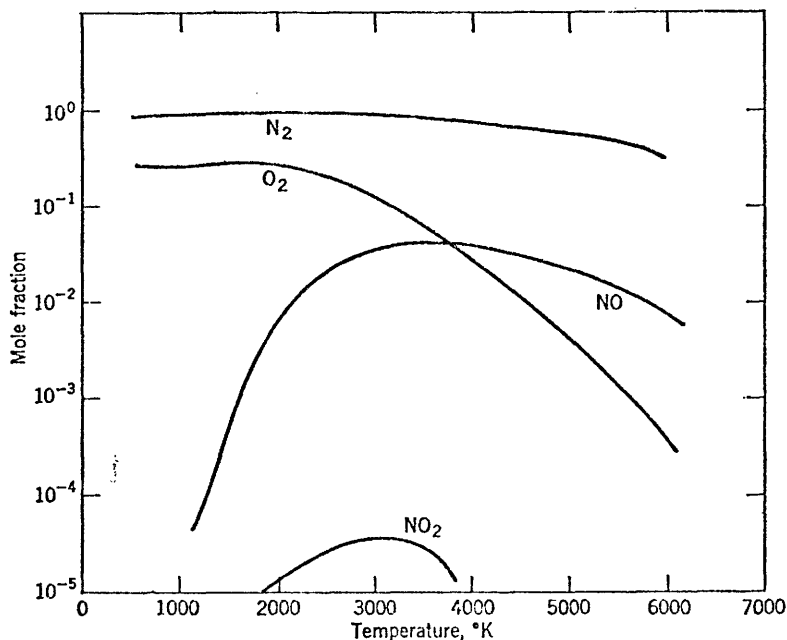
However, after the exhaust gases emerge into the atmosphere, oxidation of NO to NO_2 takes place in the presence of excess oxygen. The formation of NO_2 is determined by:

(a) The concentrations of the reactants. According to the equation:

$$2NO + O_2 = 2NO_2,$$
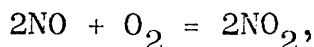
the rate of formation of NO_2 is proportional to the square of the NO concentration and directly to the O_2 concentration, i.e.

$$\frac{d}{dt}\left[NO_2\right] \propto \left[NO\right]^2 . \left[O_2\right]$$

(b) The rate constant of the fastest reaction process;

(c) The time during which the reactants are in contact.

The NO concentration in exhaust gases emerging into the atmosphere may range from 200 to 3000 ppm, but because of the low O_2 concentration, the NO_2 content is initially only of the order of one tenth this figure. In fact a ratio of $NO:NO_2$ of about 10:1 by volume is typically regarded as an appropriate overall figure for NO$_x$ emissions from combustion processes (Derwent and Stewart, 1973).

The initial rates of oxidation of NO to NO_2 in the open

atmosphere have been calculated on a theoretical basis and are given in Table 4.

Table 4

Rate of oxidation of NO to NO_2 in the atmosphere

(Source: Bagg, 1971)

Initial NO concentration (ppm)	Initial rate of oxidation to NO_2 (ppm/min.)
1	0.0003
10	0.03
100	3.0

Under certain conditions, for example those where photo-chemical smog formation is prevalent, such as in Los Angeles, much more rapid oxidation rates have been observed. Thus at an initial NO concentration of 3 ppm (which is the highest atmospheric concentration found in practice) theory would predict that only 6% would be oxidised within 1 hour. In Los Angeles however, oxidation is almost complete within 3 to 4 hours due to the enhancing effect of hydrocarbon pollutants. Nevertheless, the rate of dilution of exhaust gases after release into the atmosphere will have a strong determining effect on the oxidation rate. Slow dilution is conducive to rapid oxidation, whereas rapid dilution (as in exhaust gases from a moving car or from a high chimney in a wind) implies slow oxidation.

NO_2 is removed from the atmosphere principally by the scavenging action of water vapour which converts the NO_2 to a mixture of nitric acid and NO:

$$3NO_2 + H_2O \rightleftharpoons 2HNO_3 + NO$$

The HNO_3 vapour is rapidly removed by reaction with atmospheric ammonia and absorption on hygroscopic particles, thus all the HNO_3 eventually becomes nitrate salt aerosol. (Robinson and Robbins, 1968). The residence time for NO_x in the atmosphere is the order of one to two weeks.

4. EMISSION FACTORS

The previous section discussed the factors in the combustion process which determine the formation of NO_x, the principal ones being flame temperature, degree of excess air and gas residence time in the high temperature zone. Although under closely controlled conditions theoretical predictions of NO_x concentration in exhaust gases may agree with actual results, experience has shown that the NO_x emission levels from real-life combustion units cannot be predicted on a theoretical basis alone. Not only do emissions vary considerably between different units having similar operating conditions, but they can even differ widely in apparently identical units. It is not surprising therefore that different investigators can report widely differing emission factors (expressed, e.g. as weight of NO_x emitted per unit weight of fuel consumed). It is worthwhile quoting from the findings of Mills et al. (1961) in support. This worker made a study of the effect of operating variables on NO_x emissions from steam-raising power plants in Los Angeles County, USA, in which some 130 tests including 554 individual samples were considered. He said: "The rates of emission of NO_x from units as complex as these, with the possibility of a number of constantly fluctuating operating variables, may be assumed to be constantly fluctuating also. In these circumstances, the rate of emission of NO_x at any given instant in any plant may be different from the rate of emission at the next instant. Experience gained during the carrying out of the project has shown these assumptions to be true. A striking example of this variability is the fact that samples taken as nearly at the same time as possible from two probes in as close proximity to each other as possible show two different values for NO_x concentrations. It was found that actual rates of NO_x emissions from sister units may be different for operating conditions which are the same for each unit within the limits of ability to determine. This phenomenon has been verified repeatedly". For these reasons, and also because of the great variety of combustion units in use, there is no merit in an over-detailed analysis of the reported measurements of NO_x emissions from individual units. Also, it is only very recently that the more reliable chemi-luminescent method of measuring

NO and NO_2 concentrations has become available. Thus measurements of ten years ago may be subject to large errors.

For the purposes of this report, values for emission factors have been categorised according to fuel (solid, liquid, or gaseous) and fuel-consuming sector (electricity generation, domestic and commercial heating, general industry and transport). Table 5 gives these factors which have been derived from five representative, and as far as possible independent, sources. The variation in the estimates is clear; however since the figure given against each reference is in every case a mean or "typical" value, the real variation is much wider than implied by the quoted standard deviations. The derivation of the factors from the actual figures quoted by the reference sources is given in the Appendix.

Some general points relating to emission factors follow. Fuel category: For each of the three sectors of stationary combustion sources, the highest emission factors arise from solid fuel combustion, followed by liquid fuel, followed in turn by gas.

Stationary Sources: The study by Mills et al (1961) covered gas- and oil-fired equipment ranging from domestic heaters to large steam boilers, which enabled a relationship between gross heat input and NO_x emissions to be established. This is shown in Figure 2, which is to some extent idealised. The heat input includes heat in the fuel plus heat in the pre-heated air. The combustion pressure was around 1 atm. Again it must be stressed that these are average values, about which considerable variation is found.

The heat release rate in a given furnace volume markedly affects the flame temperature produced. If heat release rate increases but heat transfer does not keep pace, furnace temperature increases and so does NO_x formation. It is also true that NO_x emissions decrease with decrease in furnace loading, a direct result of heat release rate. Apart from this, it has been observed that in oil fired units, NO_x emissions vary with the firing technique used. Thus, for a given heat release rate, horizontal firing produces the most emissions followed by tangental firing (US: DHEW, 1970a). This is taken further in section 6 - see Figure 10.

Table 5

NOx Emission Factors (expressed as kg.NO(d) per tonne oil equivalent)

FUEL / SECTOR	Solid (a)							Liquid (b)							Gaseous (c)						
	Ref 1	Ref 2	Ref 3	Ref 4	Ref 5	Mean	Std Devn	Ref 1	Ref 2	Ref 3	Ref 4	Ref 5	Mean	Std Devn	Ref 1	Ref 2	Ref 3	Ref 4	Ref 5	Mean	Std Devn
Electricity Generation	9.7	11	17	14	11	12.5	3.0	7.6	11	13	13	13.5	11.5	2.4	–	6	7	7	6	6.5	0.6
Domestic Heating etc	2	6.8	4.3	6	2.9	4.4	2.0	4	2.8	1.8	5	8	4.5	2.4	1.7	3	1.3	2	1.8	2.0	0.6
General Industry	9	11	11	14	11	11	1.8	10	9	7.7	8.6	10	9.1	1.0	2.6	4	3.1	3.8	2.9	3.3	0.6
Transport								15				18	17	1.6							

References

1. Derwent and Stewart (1973)
2. CECD (1972)
3. US: EPA (1972a)
4. US: DHEW (1970a).
5. Bagg (1971)

Notes

(a) Assumes 1 ton coal or coke = 0.7 ton oil equivalent.

(b) Assumes 1 ton crude oil or oil products or natural gas liquids = 1 ton oil equivalent.

(c) Assumes $10^9 M^3$ natural gas = 0.9 tons oil equivalent.

(d) Unless otherwise stated it has been assumed that NO_x emissions consist of NO and NO_2 in the proportion of 10:1 by volume.

Figure 2

Relationship between heat input and NO_x emissions for gas-and oil-fired combustion equipment

(Source: Mills, et al, 1961)

AVERAGE RATE OF HEAT INPUT TO A
UNIT IN A GIVEN CLASS OF COMBUSTION
EQUIPMENT, Btu/hr
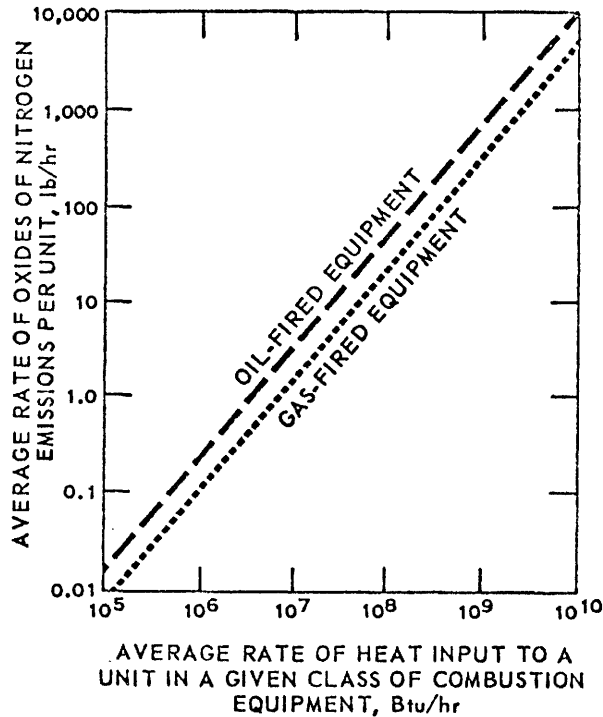
Mobile Sources: The most important source in this category is the internal combustion (i/c) engine. A considerable amount of investigatory work has been undertaken, which has established the way in which NO_x emissions vary with different operating conditions. In the case of petrol engines, Figure 3 illustrates the way in which NO_x, CO and HC (hydrocarbon) emissions in the exhaust gases, vary with air/fuel ratio. It shows clearly that maximum NO_x emissions occur when the engine is operating at the stoichimetric air/fuel ratio (i.e. at maximum flame temperature), but rapidly decreases both for richer and leaner mixtures. Different driving conditions require different air/fuel ratios for optimum engine performance. Thus when extra power is needed for acceleration, a richer mixture is produced, while under crusing conditions a leaner mixture can be used to give greater fuel economy. Laboratory studies, quoted by Bagg (1971), have also shown how NO_x emissions vary with ignition timing and manifold vacuum. Figure 4 shows the effect of these factors. Other factors which

Figure 3

Relationship between air-fuel ratio and pollutant emissions for petrol-fuelled i/c engines
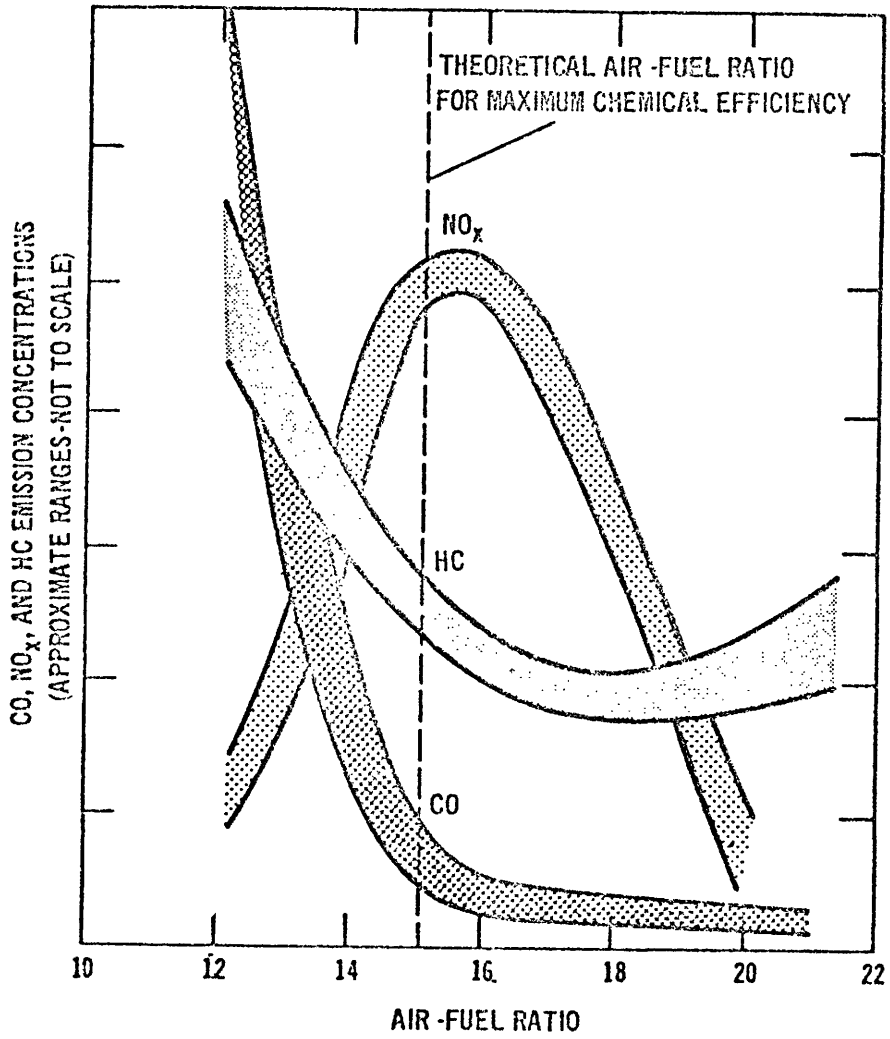
(Source: US: DHEW, 1970a)

THEORETICAL AIR-FUEL RATIO FOR MAXIMUM CHEMICAL EFFICIENCY

NO_x

HC

CO

CO, NO_x, AND HC EMISSION CONCENTRATIONS (APPROXIMATE RANGES-NOT TO SCALE)

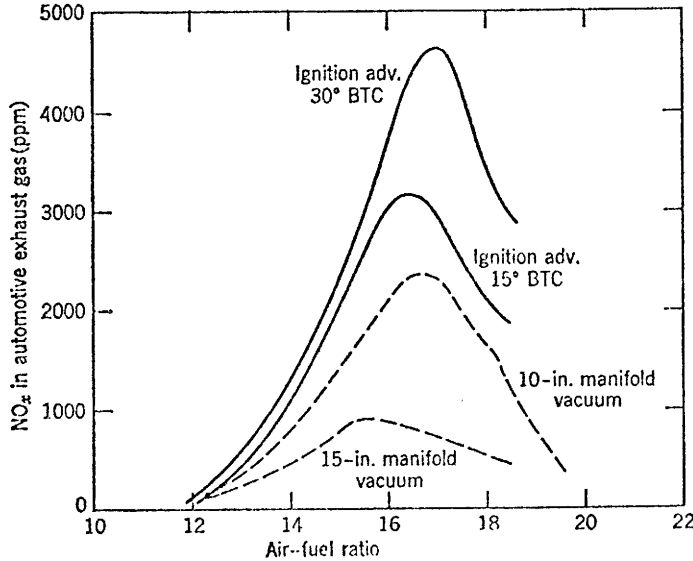10   12   14   16   18   20   22

AIR-FUEL RATIO

Figure 4

Effect of ignition timing and manifold vacuum on NO_x emissions
(petrol engines)

(Source: Bagg, 1971)

increase the emissions are increase in compression ratio, increase
in coolant temperature, and build-up of deposit in the combustion
chamber, all of which tend to produce higher combustion temperatures
or extend the time for which high temperatures persist. These
matters are discussed further, as they relate to emission control,
in section 6.

Table 6, quoted by Derwent and Stewart (1973), for a 1300cc
engine, shows the considerable range over which NO_x concentrations
in the exhaust gases can vary.

Table 6

NO_x emissions in exhaust from petrol engine under varying
driving conditions

(Source: Fussell, 1970)

Driving Operation	Average $NO^{(a)}$ in exhaust (ppm)
Idling	30
Acceleration	6000
Cruising	2000
Deceleration	60

Note (a): The NO_x consists almost entirely of NO upon emission
to the atmosphere.

Because of these wide variations and the different driving patterns applying in practice, it is very difficult to arrive at a weighted emission factor which can be applied to the total quantity of fuel consumed. Derwent and Stewart use a mean NO_x concentration of 1000 ppm, which leads to an emission factor of 16 Kg NO/tonne fuel consumed. Bagg (1971), reporting American tests on non-US compact cars quoted a mean figure of 1200 ppm NO_x ($\sigma = 580$ ppm). Considering the scope for variation, this is a not unsatisfactory agreement between the two independent sources of information.

In diesel engines, the fuel does not enter the cylinder mixed with air, but is injected into the cylinder under high pressure. Air-fuel ratios are higher than with petrol engines. Experiments have shown that direct-injection engines behave very similarly to petrol engines as far as NO_x emissions are concerned. Emissions from indirect-injection engines, on the other hand, are much lower and much less sensitive to air-fuel ratio. Figure 5 illustrates these findings.

Figure 5

Relationship between air usage and NO_x emissions in exhaust gases from diesel engines

(Source: Bagg, 1971)
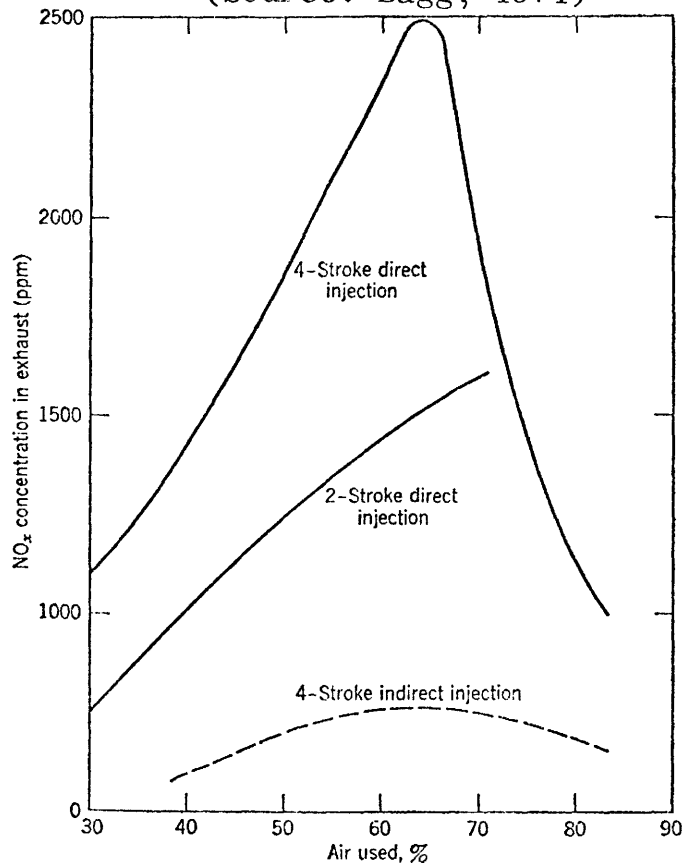
As with petrol engines, the NO_x emissions differ with the driving operation. Table 7 quotes average figures for a normally-aspirated diesel engine. Because of the difference in air-fuel ratio, Tables 6 and 7 cannot be compared directly.

Table 7

NO_x emissions in exhaust from normally-aspirated diesel engines under varying load conditions

(Source: Fussell, 1970)

| Load Condition	Average NO in exhaust (ppm)
Idling	120
No Load	110
Half Load	490
Full Load	920

However two points can be made. Firstly the NO_x concentrations in diesel exhaust gases are much lower, and secondly there is much less variation between the emissions in the various operating modes, than for petrol engines. Based on the above figures, Derwent and Stewart derived a mean NO_x concentration of 500 ppm, equivalent to 11kg NO/tonne fuel consumed, which is rather lower than petrol engine emissions. Bagg, on the other hand states that "under typical driving conditions the average weight emission from diesel-powered vehicles will be equal to or slightly greater than (US) gasoline powered vehicles" (Bagg, 1971). It seems fair to conclude therefore that, for the purposes of the present exercise, NO_x emission factors for the two sources can be taken as being of a similar order.

Gas turbines are the third main class of i/c engine used for propulsion - in this case, aircraft. Figure 6 shows how NO_x emission varies with air/fuel ratio for a Pratt and Whitney JT8 (rated at 14000 lbs thrust).

Figure 6

Relationship between air-fuel ratio and NO_x emissions for aircraft jet engine (Pratt and Whitney JT8)

(Source: Bagg, 1971)

Equivalence ratio ϕ

(ϕ = actual fuel-air ratio/stoichimetric ratio)
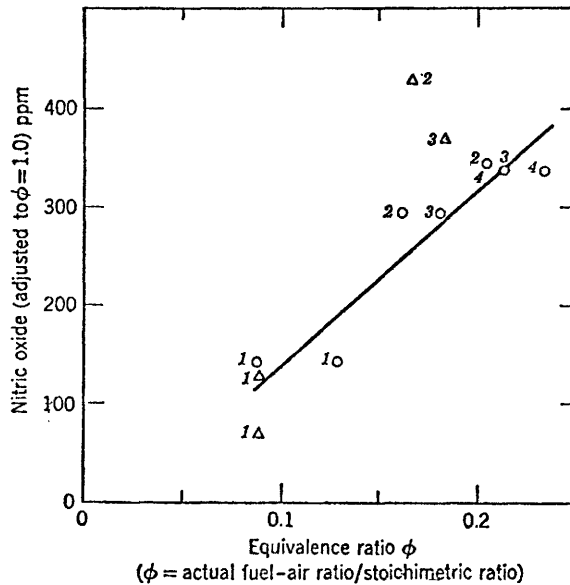
Key 1 = Idling	3 = Max. continuous power
2 = Cruising	4 = Max. short term power
References 0 = Smith et al (1968)	$\triangle$ = George & Burlin (1960)

The adjustment of emission concentrations to an air/fuel ratio of 1 is made to permit direct comparison with conventional road vehicle propulsion units, and it is immediately apparent that the emission factors for jet engines are much lower than for petrol engines. Typical NO_x emission factors for aircraft are summarized in Table 8, quoted by Derwent and Stewart.

Table 8

Typical NO_x emissions from aircraft (Kg NO/tonne fuel)

(Source: NREC, 1968)

Aircraft Type	Operating Mode	
	Taxi	Take-off, Landing, Climbing
Turboprop	3.7	3.1
Short-haul jets	2.0	4.3
Long-haul jets	2.0	4.3

5. NO$_x$ EMISSION ESTIMATES

Fuel consumption estimates broken down by sector of use up to 1985 are available from Commission documents for the Community of six, for oil (EEC, 1972a) for gas (EEC, 1972b), and for all three fuel types (EEC, 1972c). There are also some partly disaggregated data for coal consumption for the Community of 9 (Stief-Tauch, 1973). However the most complete set of statistics currently available for the Community of 9 are to be found from OECD sources (OECD, 1973), and the latter has been used as the main basis for calculating NO$_x$ emissions. Table 1 of OECD (1973) gives sectoral energy requirements for the years 1970, 1980 and 1985 in terms of million tons of oil equivalent (M t.o.e.) on the basis of 1M t.o.e. $\equiv$ 10^{13} K cal. It is worth stressing again, that the current oil supply crisis will certainly result in profound changes in these fuel estimates.

These data are presented in Table 9, broken down by fuel and by sector of use. The figures for 1975 have been interpolated. Using the fuel quantities in Table 9, multiplied by the emission factors in Table 5, NO$_x$ emissions can be calculated, and are shown in Table 10. Emission factors were assumed constant over the whole period, the mean values from Table 5 being used. No controls were assumed. Figures 7 and 8 show the same information graphically, upper and lower ($\pm 2 \sigma$) estimates as well as mean values being given separately.

A number of conclusions can be drawn from the data, the most important being:

(1) In 1970 the total NO$_x$ emissions amounted to around 7.6M tonnes, and this could rise to 13M tonnes by 1985, i.e. an increase of 70%;

(2) Analysis by sector of fuel consumption shows the following breakdowns for 1970 and 1985:

Table 9

EEC[a] SECTORAL PRIMARY ENERGY CONSUMPTION: 1970-1985

Million tons of oil equivalent (10^{13} K Cal.)

(Source: OECD, 1973)

YEAR / SECTOR \ FUEL	1970				1975[g]				1980				1985			
	Solid	Liquid	Gas	Total	Solid	Liquid	Gas	Total	Solid	Liquid	Gas	Total	Solid	Liquid	Gas	Total
Electricity Generation	113	51	19	183	102	95	30	227	95	136	39	270	90	182	47	319
Domestic Heating and Misc[a]	52	113	29	194	30	157	53	240	16	187	75	278	10	218	97	325
Industry[d]	85[b]	151[e]	67	303	68[b]	190[e]	97	355	63[b]	242[e]	125	430	58[b]	292[e]	153	503
Transport[c]	2	132	-	134	1	167	-	168	-	212	-	212	-	265	-	265
TOTAL[f]	252	447	115	811	201	609	180	990	174	777	239	1190	158	957	297	1412

Notes (a) Includes households, commercial, small industrial and agricultural undertakings, etc.

(b) Iron and Steel industry includes coke and other solid fuels.

(c) Including bunkers for ships.

(d) Including internal requirements of the energy-producing industries (refineries, coal mines, etc).

(e) Including refinery losses.

(f) Excludes primary fuels used in non-energy applications.

(g) Interpolated data using best fit curves on data for 1970, 1980 and 1985.

Table 10

EEC(9) NO$_x$ EMISSIONS: 1970 - 1985

(Million tonnes, mean values(b) assuming no controls)

SECTOR	1970				1975				1980				1985			
	Solid	Liquid	Gas	Total	Solid	Liquid	Gas	Total	Solid	Liquid	Gas	Total	Solid	Liquid	Gas	Total
Electricity Generation	1.4	0.6	0.1	2.1	1.3	1.1	0.2	2.6	1.2	1.6	0.3	3.1	1.1	2.1	0.3	3.5
Domestic Heating etc	0.2	0.5	0.1	0.8	0.1	0.7	0.1	0.9	0.1	0.8	0.2	1.1	-	1.0	0.2	1.2
Industry	0.9	1.4	0.2	2.5	0.7	1.7	0.3	2.7	0.7	2.2	0.4	3.3	0.6	2.7	0.5	3.8
Stationary Sources Total	2.5	2.5	0.4	5.4	2.1	3.5	0.6	6.2	2.0	4.6	0.9	7.5	1.7	5.8	1.0	8.5
Transport(a)	-	2.2	-	2.2	-	2.8	-	2.8	-	3.6	-	3.6	-	4.5	-	4.5
Total	2.5	4.7	0.4	7.6	2.1	6.3	0.6	9.0	2.0	8.2	0.9	11.1	1.7	10.3	1.0	13.0

Notes (a) Most of this arises from road transport. Derwent and Stewart (1973) report that NO$_x$ emissions from aircraft landing and taking off at Heathrow airport, London, amounted in 1970 to only about 40,000 tons.

(b) The variation (2σ) about the mean:-

   (i) arising from uncertainties in NO$_x$ emission factors, is 0.67 to 1.5 x the mean value for sectoral and fuel type totals, and 0.8 to 1.25x the mean values for total emissions.

   (ii) arising from uncertainties in fuel consumption, based on Stief-Tauch (1973), is 0.9 to 1.1 times the mean values.

Table 11

NO$_x$ Emissions: Sector Breakdown for 1970 and 1985 (Mean)

Year Sector	1970		1985	
	M Tons NO$_x$	%	M Tons NO$_x$	%
Electricity Generation	2.1	28	3.5	27
Domestic	0.8	10	1.2	9
Industry	2.5	33	3.8	29
Transport	2.2	29	4.5	35
Total	7.6	100	13.0	100

Thus, although in 1970 industry is the largest sectoral contributor, accounting for around one third the total, transport is the fastest growing sector, doubling from 2.2M tonnes in 1970 to 4.5M tonnes in 1985, by which time it could replace industry as the largest sector.

(3) Analysis by type of fuel consumed shows the following breakdowns:

Table 12

NO$_x$ Emissions: Fuel Type Breakdown for 1970 and 1985 (Mean)

Year Fuel	1970		1985	
	M Tons NO$_x$	%	M Tons NO$_x$	%
Solid	2.5	33	1.7	13
Liquid	4.7	62	10.3	79
Gas	0.4	5	1.0	8
Total	7.6	100	13.0	100

The most striking features are the growth of emissions from liquid fuels (almost half of which arises from mobile sources), and the decline of emissions from solid fuels. In absolute terms, the emissions from liquid fuels more than doubles, from 4.7M tonnes in 1970 to 10.3M tonnes in 1985; while solid fuel emissions fall by a third from 2.5M tonnes in 1970 to 1.7M tonnes in 1985. Emissions from gas-fired sources, whilst still a minor

contributor to the whole, nevertheless increase by 150%, from
0.4M tonnes in 1970, to 1M tonnes in 1985.

The figures from which the above conclusions are derived,
are subject to two sources of variation: variation in the actual
quantities of fuel consumed and variation in the values of the
NO_x emission factors themselves. The total variation is illustrated
in Figures 7B, 7C, 8B and 8C.

Taking the emission factors first, those used are mean values
and the possible variation about the means is indicated in Table 5
and in the Appendix. This can be quite large: a factor of two
from the lowest value to the highest value quoted in the five
references for a particular emission factor is not unusual.
Moreover, the value quoted by each reference is itself a mean or
"typical" value. Bearing in mind these large uncertainties and
the very small number of independent references quoted a detailed
statistical presentation would not be justified, but the true
values of the sectoral and fuel type emissions probably lie
between 0.7 and 1.5 times the indicated figures, while the true
total emissions probably lie between 0.8 and 1.2 times the
indicated figures (2 σ limits), given that the fuel consumption
figures are correct. The possible variations in fuel consumption
are less easy to define, for two reasons:

(a) The consumptions in each sector and for each fuel type
are not independent, but are related in a complex manner;

(b) The consumption of each type of fuel is dependent
ultimately on the price structure and availability, and
also on the rate of growth of the economy of the
Community. The current crisis affecting oil supplies
will of course have a profound effect on any future
estimates.

Stief-Tauch (1973) gives the most likely range of solid fuel
consumption in 1985 as 210-250M t.e.c., i.e. 147-175 M t.o.e.
This represents a variation of approximately $\pm$ 10% on the actual
figure of 158M t.o.e. quoted in Table 8, which is quite small
compared to the likely variation arising from uncertainties in
the NO_x emission factors. However, much larger variations in
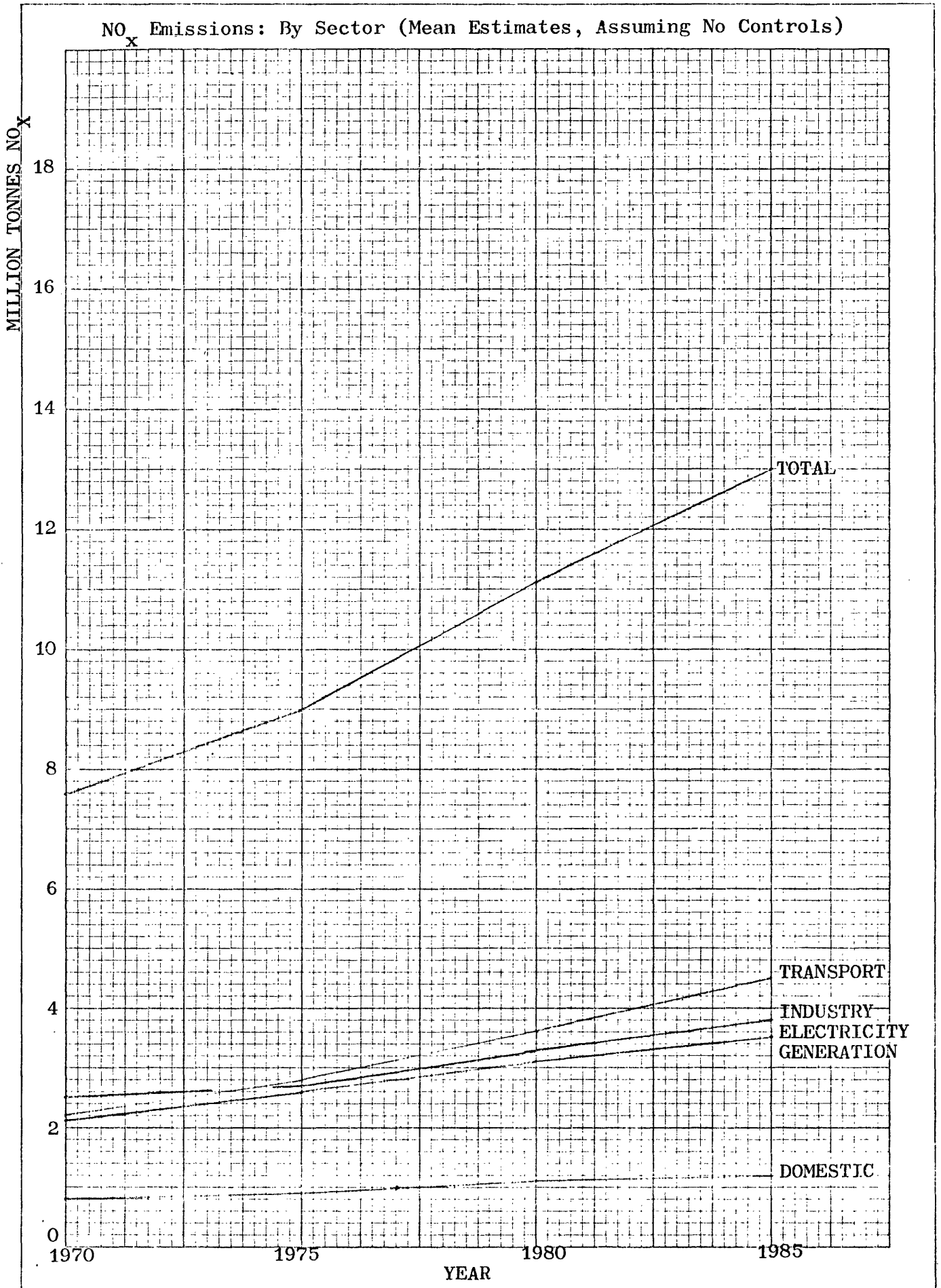fuel consumption might well be considered possible.

Figure 7A

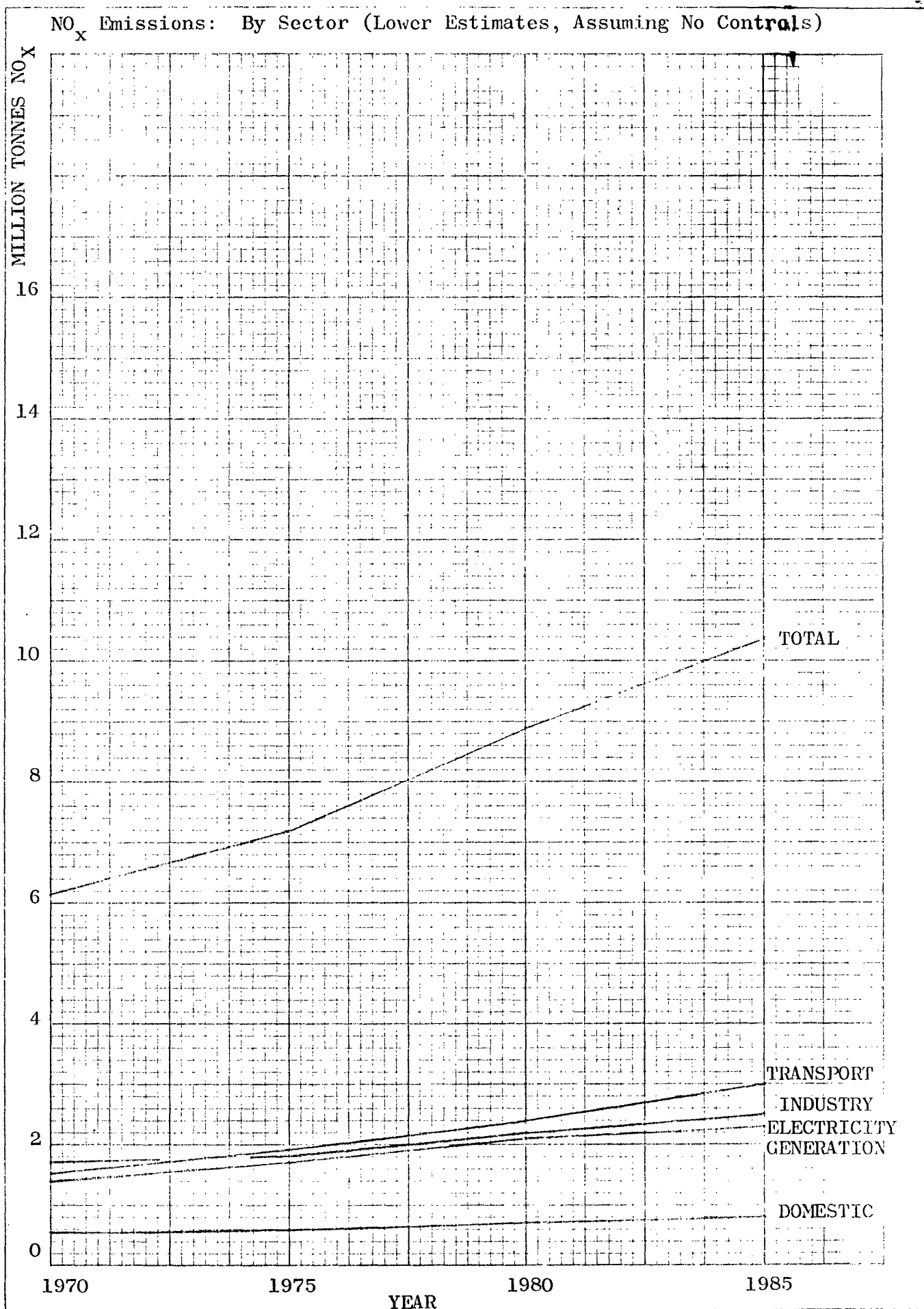NO$_x$ Emissions: By Sector (Mean Estimates, Assuming No Controls)

Figure 7B

NO$_x$ Emissions: By Sector (Lower Estimates, Assuming No Controls)

Figure 7C

NO$_X$ Emissions: By Sector (Upper Estimates, Assuming No Controls)
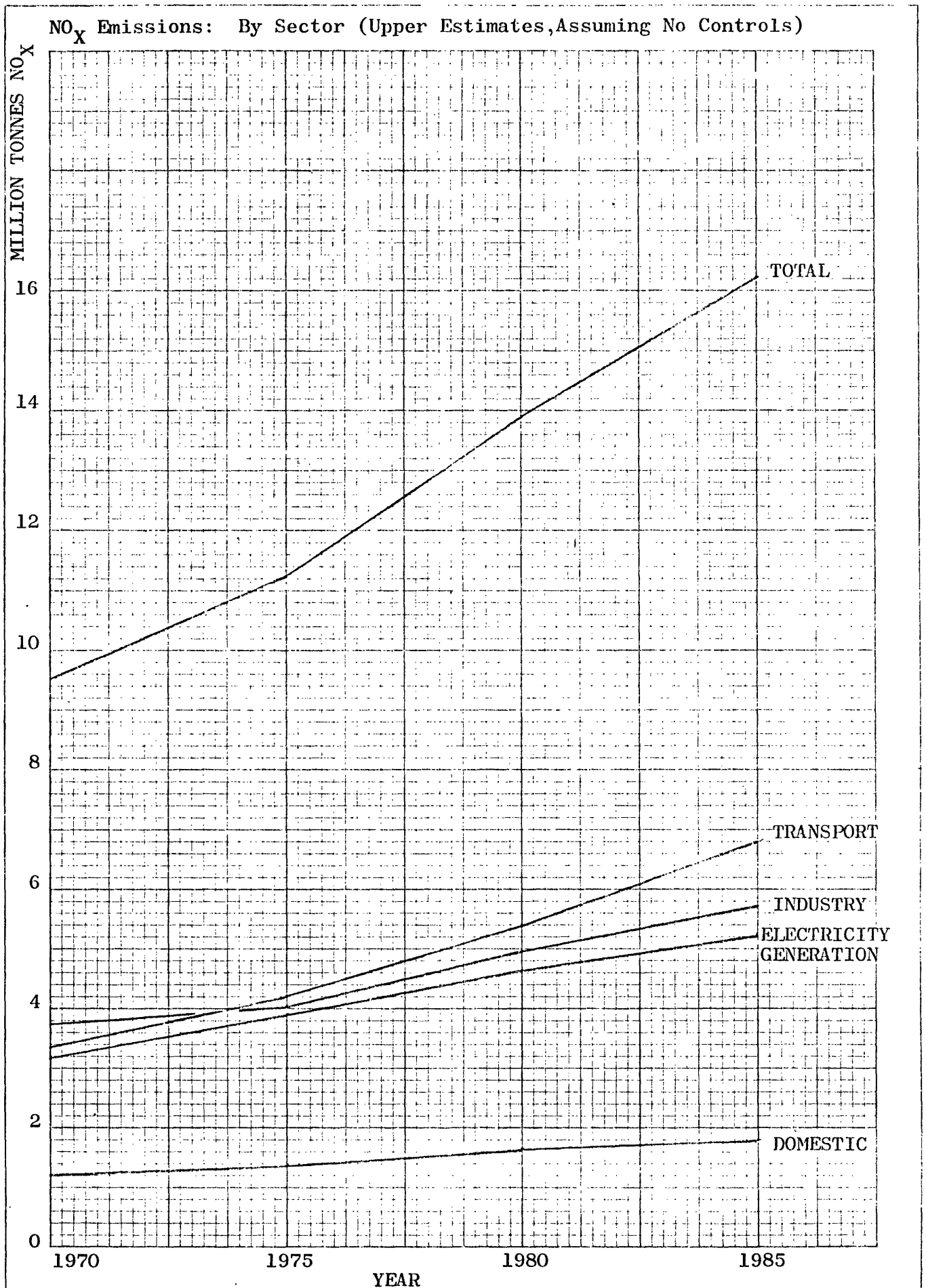
Figure 8A

NO$_x$ Emissions: By Fuel (Mean Estimates Assuming No Controls)
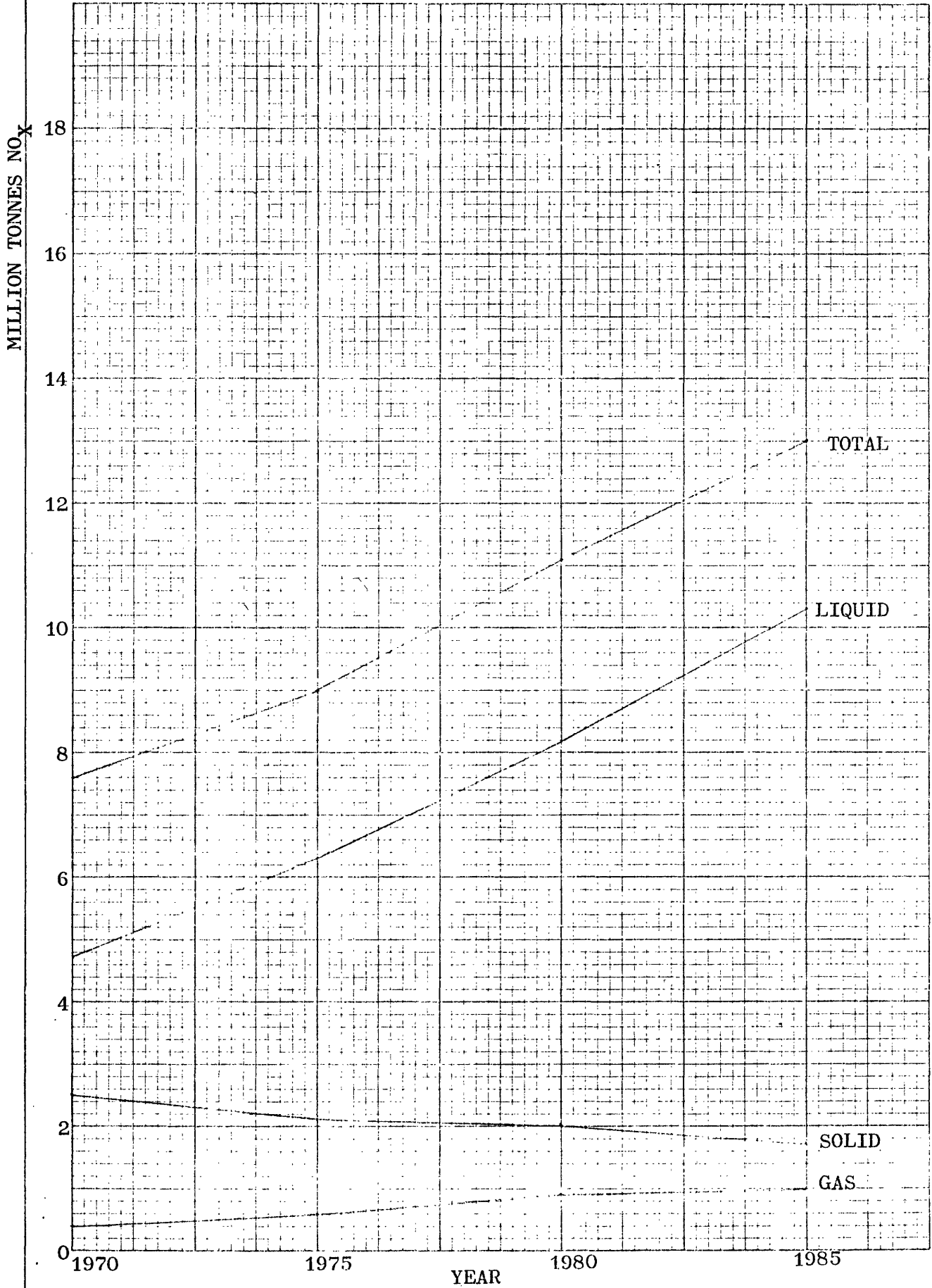
Figure 8B

NO$_X$ Emissions: By Fuel (Lower Estimates, Assuming No Controls)

Figure 8C

NO_X Emissions: By Fuel   (Upper Estimates, Assuming No Controls)
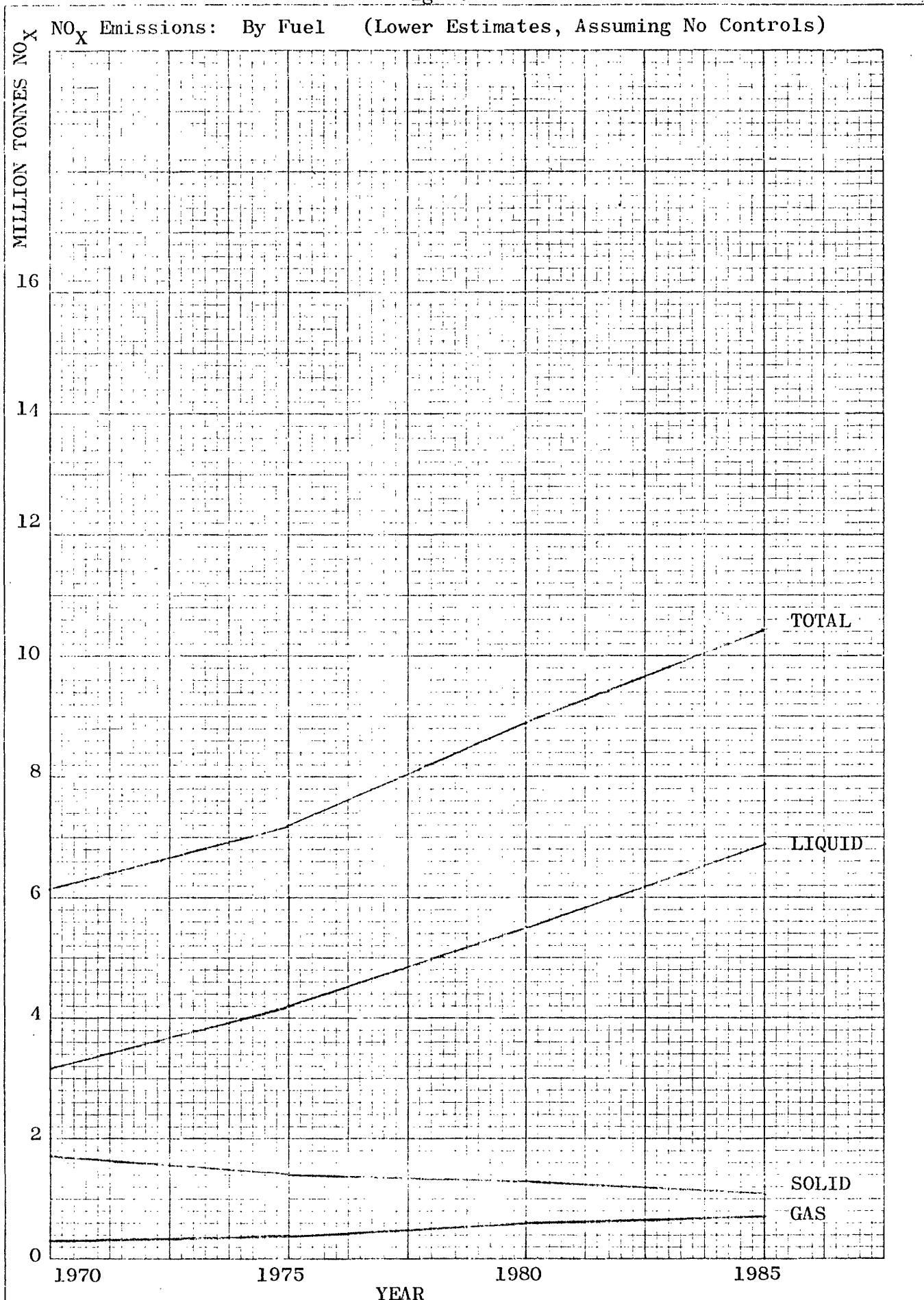
Figure 8C. NO_X Emissions: By Fuel (Upper Estimates, Assuming No Controls). Y-axis: MILLION TONNES NO_X (0 to 16+). X-axis: YEAR (1970 to 1985). Curves labeled TOTAL, LIQUID, SOLID, GAS.
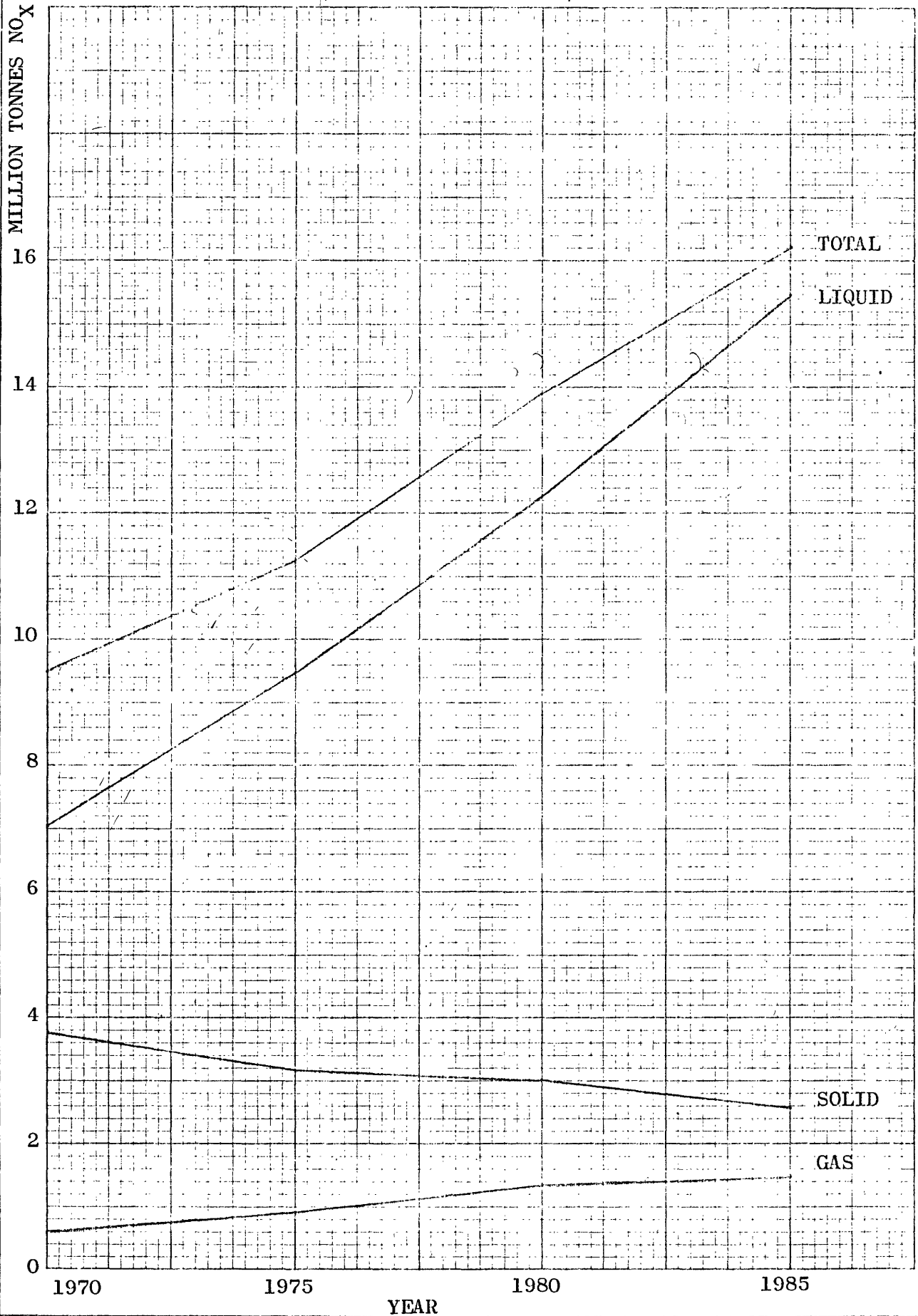
<u>Effect of Emission Controls</u>:   No statutory control of NO_x emissions are as yet planned either from stationary or mobile sources, within Community member countries.   However, as is pointed out in the succeeding section of the report, a reduction in the emission factors from stationary sources of around 50% is possible with current technology.   With "advanced" technology, a reduction of 75% in emission factors from stationary sources and 50% from mobile sources is feasible.   However it seems doubtful whether by 1985, the application of "advanced" control technology could have extended sufficiently to achieve much impact on total emissions.   On the other hand, if controls are applied it could well be that by 1980 some degree of reduction could be achieved through application of known methods.   This is discussed further in section 6 below.

Any logical control policy ought, of course, to take into account the damage, if any, arising from the emissions from various sources.   This is likely to be related to manner of emission (e.g. high or low level) as well as total quantity emitted, since the former will influence the resulting ground level concentrations.   However these matters go beyond the scope of this report.

6. <u>CONTROL OF NO$_x$ EMISSIONS</u>

This section is concerned with NO$_x$ control methods which have been developed or proposed. In order to place the topic in its proper perspective it is important to realise that, so far as is known, there are no stationary combustion sources operating at the present time with NO$_x$ emission control either in Western Europe or the USA. In the United Kingdom, however, the CEGB do practise low excess air combustion in certain fossil-fuelled electricity generating plant in order to reduce formation of sulphur trioxide (SO$_3$). This procedure also incidentally reduces NO$_x$ formation. The description of the control of NO$_x$ emissions from stationary sources which follows, refers wholly to experimental work which has been carried out in recent years. In the case of mobile sources however, exhaust gas recirculation is in operation on cars in the USA. Control of NO$_x$ emissions may be achieved by one or more of the following three techniques:

(a) Modification of fuel combustion conditions;

(b) Modification of combustion chamber design

or (c) Removal of NO$_x$ from the exhaust gases after formation. Although these alternatives apply equally to stationary and mobile sources, the actual techniques differ considerably between classes of source; the latter will therefore be considered separately.

<u>Stationary Sources</u>: (a) <u>The Combustion process may be modified</u> in the following ways:

(i) <u>Low excess air combustion</u>

This is one of the most promising and universally applicable methods of NO$_x$ reduction. It may be very effective where the fuel is gas or oil, but in coal-firing serious imbalances in fuel/air distribution could result and problems of unburned fuel or carbon monoxide emissions may limit its use in this type of plant. Figure 9 indicates the relationship between excess air and NO$_x$ emissions for oil-fired boilers which has been reported from US experiments.

Figure 9

Experimental Relationship between NO$_x$ emissions and excess air
for oil-fired boilers

(Source: US DHEW, 1970a)

OXYGEN IN FLUE GAS, percent
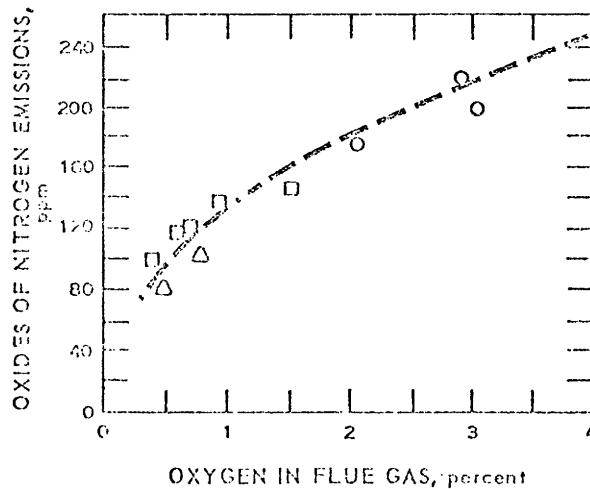
(ii) Two-stage combustion

This is a technique whereby the air supply to the primary
combustion zone is reduced to the theoretical (stoichimetric)
level, or just below, and a secondary air feed is made just beyond
the flame front where combustion is completed. It has been
reported that in experiments with horizontally-fired units, two-
stage combustion could reduce the NO$_x$ emissions by 50% compared
to a similar single-stage unit, and in tangentially-fired units
the corresponding reduction is 22% (Bagg, 1971). As with low
excess air combustion, coal-fired units would be difficult to
operate using this technique.

(iii) Flue gas recirculation

This is a technique whereby part of the flue gases
(10-20%) are diverted back to the furnace chamber. This has the
effect of decreasing the peak flame temperature by diluting the
primary flame zone with non-reactive gases. US data indicate
that NO$_x$ emissions could be reduced by at least 50% by applying
this technique to a tangentially-fired gas burning boiler
(Bagwell et al, 1970). However, the practicality of this approach
to the whole range of combustion units remain to be demonstrated.
Thus the recirculation of large quantities of gases in the
furnaces of electricity generating plant could pose handling
problems, in addition to increased capital and operating costs,
and efficiency losses.

(iv) <u>Water or steam injection</u>

This produces a similar condition to flue gas recirculation, i.e. the lowering of peak flame temperature. However although laboratory studies have reported some success, the practical drawbacks such as decreased efficiency and increased corrosion in the combustion chamber have not yet been fully assessed.

(b) <u>Turning to modification of combustion chamber design</u>, the principal aim here is to slow down the rate at which fuel and air mix. Thus more heat is removed from the gas before combustion is complete, resulting in a lower peak flame temperature and therefore less NO_x formation. There are essentially two different approaches which have been proposed:

(i) <u>Burner configuration</u>

In general it appears that highly turbulent conditions within the burner tend to favour high NO_x emissions while more streamline gas flows produce less NO_x. This explains why cyclone burners emit higher NO_x concentrations than tangential burners. Figure 10 illustrates the different emission levels produced by horizontal and tangential firing.

<u>Figure 10</u>

<u>Effect of firing made on NO_x emissions</u>

(Source: US DHEW 1970a)

HEAT RELEASE, 10^3 Btu/ft^3-hr
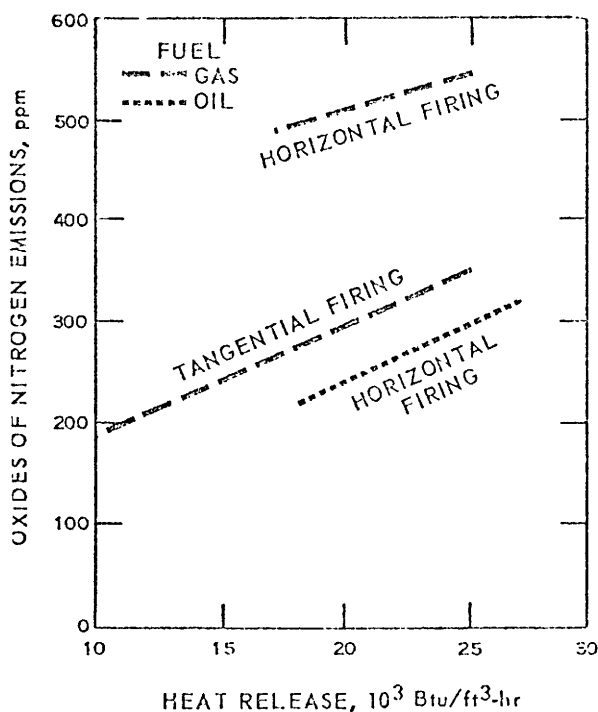
This shows that as much as 50-60% reduction in NO_x levels could be achieved by using tangential rather than horizontal firing.

(ii) <u>Fluidized bed combustion</u>

High heat transfer rates are possible in fluidized beds, which lead to lower peak temperatures and hence lower NO_x formations. The addition of limestone or similar material to the combustion zone has been suggested, since it has the potential of reducing SO_2 emissions as well, by chemical combination of the sulphur contained in the fuel. (Bartok et al 1971). However, practical equipment based on this principle is still in process of development.

(c) <u>Finally, the removal of NO_x from flue gases</u> provides, in theory, an alternative means of reducing NO_x released into the atmosphere. However, technology development is hampered by the very large volumes of gases to be treated and the low concentrations of NO_x present, compared to other gaseous pollutants. Four approaches have, to varying extents, been investigated.

(i) <u>Catalytic decomposition</u>

The principle here is that at the relatively low temperatures found in exhaust gases, the equilibrium condition of the reaction:

$$N_2 + O_2 \rightleftharpoons 2NO,$$
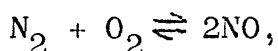
favours the decomposition of NO into nitrogen and oxygen. Unfortunately at these temperatures the decomposition rate is extremely slow, and no suitable catalyst has been discovered.

(ii) <u>Catalytic reduction</u>

This involves the use of a reducing agent which will convert the NO to nitrogen by chemically removing the combined oxygen. The technique has been successfully applied to the treatment of tail-gas from nitric acid plants, where addition of carbon monoxide or methane is made before passing over a platinum metal catalyst. Over 90% of NO_x can be decomposed by this method. Typical reactions are:

$$2NO + 2CO = N_2 + 2CO_2$$
$$4NO + CH_4 = 2N_2 + CO_2 + 2H_2O$$
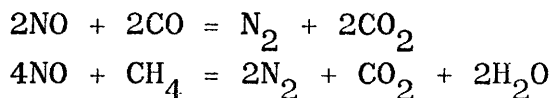
The reactions are exothermic, and the actual process conditions used depend upon the value attributed to recovered heat, the cost of heat exchangers etc, and the availability and cost of reducing gases.

However the applicability of this method to flue gases from combustion sources is very doubtful, due to the high catalyst costs, the emission of quantities of reductant gases (and even ammonia) into the atmosphere (which could themselves be regarded as pollutants), and the poisoning effect on the catalyst of sulphur-containing compounds present in the flue gas stream.

(iii) <u>Adsorption/Absorption</u>

NO is a relatively inactive gas chemically, compared to the more reactive dioxide, NO_2. If oxidation of NO to NO_2, which takes place slowly on release to the atmosphere, could be speeded up, the chemical removal of NO_2 might then be feasible. There has therefore been a search for suitable oxidising agents, and such adsorbents as silica gel, alumina, molecular sieves, char, and ion-exchange resins have all been shown to have the capacity of adsorbing NO, with associated oxidation to NO_2. Other solid sorbents include manganese and alkalised ferric oxides (Bartok et al, 1971). Technical problems include poisoning and mechanical attrition of the sorbent in the gas stream.

Rather less unpromising in theory is chemical adsorption using liquid scrubbing solutions or suspensions. Thus alkaline solutions such as calcium hydroxide or limestone suspensions in water are capable of removing not only NO_2 but other acidic gases such as SO_2 and SO_3 as well. Moreover, an equimolecular mixture of NO and NO_2 leads to the formation of the sesquioxide N_2O_3:

$$NO + NO_2 \rightleftharpoons N_2O_3$$
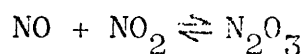
which is more reactive than either of the simpler oxides. Recycling of NO_2 into the flue gas can facilitate this reaction.

Concentrated sulphuric acid will also absorb NO and NO_2 mixtures, as well as SO_2.

Disadvantages of absorption by liquids include the low efficiency (less than 50%), the cost of reagents, and the problems of disposal of large volumes of liquid effluents, which could create pollution problems of different kind. The ideal solution would be to generate compounds which are commercially marketable, for example, as fertilisers. Another disadvantage is the cooling of the gas plume which leads to poor dispersion

and possible high local ground level concentrations of pollutant gases. There is also the possibility of mist formation.

(iv) Physical separation

A successful physical separation process would make use of one or more physical property of NO (or NO_2) which differs significantly from the corresponding property of other constituents in the flue gas. A number of possibilities have been investigated, including condensation temperature, magnetic susceptibility and molecular size, but without much success, (Bartok et al, 1971).

In summary, the most promising techniques for NO_x control in stationary combustion sources appear to consist in the modification of fuel combustion conditions or furnace design. A reduction in NO_x emissions of 50%, compared to unmodified plant is probably achieveable, and maybe more, but with loss of efficiency.

The necessary basic technology is available for oil and gas-fired plant, but coal-fired units could present grave problems where fine control of fuel/air ratios is required. Flue gas treatment is in principle applicable to all classes of fuel combustion, but the technology is much less well-developed. Nevertheless if legislation were to require the removal of NO_x to around 90%, it seems that catalytic decomposition or reduction methods must be developed. Perhaps the answer would lie in a combination of several different techniques. No stationary combustion sources are known to be operating in Western Europe or the USA at the present time which practise NO_x control.

Mobile Sources: The relationship between air/fuel ratio in i/c engines and NO_x emissions has already been discussed in Section 4 of the report, where it was seen that conditions which minimise emission of carbon monoxide (CO) and hydrocarbons (HC) produce high NO_x emissions, in an unmodified engine. The bulk of the development work on reduction of emissions from i/c engines has been done in the USA, which was the first country to impose gas emission standards from mobile sources. The current US limit set for NO_x emissions from automobiles is 3.1 gms/mile, equivalent to about 14 Kg/NO_x/t.o.e. for US-manufactured vehicles and 28 Kg/NO_x/t.o.e. for European cars. NO_x emission limits in future years were planned to become progressively more severe, but because of the difficulties foreseen in achieving the

originally proposed levels there has recently been a relaxation
of the intended limits. Thus the original intention was to
achieve a level of 0.4 gm/mile (= 1.8 Kg NO_x/t.o.e. for US cars
and 3.6 Kg NO_x/t.o.e. for European cars) by 1976, representing
a 90% reduction compared with 1971 level. At the time of
writing however, the 1976 models will be required to meet an
interim standard of 2.0 gm/mile. When (or even whether) the level
of 0.4 gms/mile will be reimposed remains to be seen.

No legislation limiting NO_x emissions from mobile sources
within Community countries is envisaged at the moment, but
reference to the Appendix shows that current uncontrolled emission
rates from West European cars average about 16 Kg NO_x/t.o.e. or
1.8 gm NO_x/mile, which meets the US interim standard for 1976.

In describing potential methods of NO_x emission control
in i/c engines, it is convenient to discuss the various types
of engine separately.

(a) Spark-ignition engine

The simplest way of reducing NO_x emissions is to reduce
the compression ratio, which has the secondary effect of reducing
the engine octane requirements, with some loss of power and
increase in fuel consumption.

Exhaust gas recirculation is currently practised on automobiles
in the USA. The fuel/air mixture is diluted with controlled
amounts of exhaust gas before entering the combustion chamber.
This reduces peak combustion temperatures and so NO_x formation
is lower. However considerable power loss is incurred, also
some induction system components can be corroded and plugged
by acid condensate and dirt in the recycled exhaust, but the
omission of lead from petrol would moderate these problems.

Exhaust gas recirculation is probably adequate to meet the
US interim 1976 standard of 2.0 gms/mile for US cars (a
reduction of 50% from the 1971 NO_x emission level) but the
original limit of 0.4 gms/mile would certainly require a
radically different approach. Probably this lower level can
only be achieved in a conventional engine (if at all) by the
use of some form of catalytic conversion of the NO_x, most likely
reduction to nitrogen. However, a satisfactory catalytic
converter has yet to be developed; the problems to be overcome
include damage by inadvertent overheating, mechanical erosion,
and poisoning from fuel additives such as lead.

It may be that a radical change in the design of spark-ignition engine is required, to achieve significant reductions in NO_x emission.

The Wankel engine, although having higher CO and HC than the conventional engine, has lower NO_x emissions in the region of 0.6 to 0.7 gms/mile, (US: OST, 1972), but at the expense of increased fuel consumption.

The Stratified charge engine utilises a rich area in the combustion chamber which is easily ignited and raises the temperature of the whole charge to a level where it burns more completely (thus reducing residual hydrocarbons). By this means, overall lean mixtures can be burned, giving low NO_x emissions. In addition, the fuel consumption is improved.

So far, consideration has been given to engine modifications or catalytic converters. A third approach remains, that of water injection into the intake manifold. This decreases the peak combustion temperature by virtue of the latent heat absorbed during the vaporisation of the water, but the disadvantages are the formation of sludge leading to a breakdown of lubrication, and increased engine wear, together with increased fuel consumption.

(b) Diesel engine

Broadly speaking, NO_x emissions (in terms of weight of NO_x emitted per unit weight of fuel) are of the same order for both petrol and diesel engines, as discussed in Section 4. However, indirect injection with the maximum possible injection time of the fuel will reduce NO_x emissions (see Figure 5) but with the penalty of some increase in fuel consumption. The discovery of an effective NO_x reduction catalyst seems unlikely at the present time, due to the higher oxygen content of the exhaust stream, compared to that from petrol engines.

(c) Gas turbine

Although this type of propulsion unit may find future application in railway engines and heavy goods road vehicles, the main use will probably continue to be in aircraft. Reduction of NO_x emissions can be achieved by reduction of maximum temperature in the primary zone of the combustor. Decreased air/fuel ratio may reduce combustor chamber temperature and

thereby reduce NO_x emissions. Apart from engine modifications, reduction of emissions of all kinds can be reduced by restriction of ground operation of the engines as much as possible. This could mean the use of auxiliary vehicles to tow aircraft to the take-off point, or alternatively the transport of passengers to the aircraft to reduce idling time.

(d) Unconventional systems

Some of the alternatives to the conventional i/c engines which could reduce NO_x emissions include the Rankine and Stirling heat engines. However the ultimate answer may lie in the electric car, provided a power system of greater overall energy usage efficiency can be developed.

In summary, although an enormous amount of effort in the last few years (mainly in the USA) has gone into the development of techniques for the reduction of NO_x emissions from conventional spark ignition engines, the only practically successful result so far has been achieved through exhaust gas recirculation. This has enabled US cars to reduce NO_x emissions by 50% from the 1971 levels (4 gms NO_x/mile). Reduction by 90% of the 1971 levels probably can only be achieved in conventional engines by the use of catalytic conversion, but no satisfactory solution is yet in sight. Further NO_x reduction can however be achieved by the use of the indirect injection diesel, the Wankel, or a stratified charge engine, but the final solution may lie in the development of a practical electric drive if overall fuel economy is sacrificed. (See US DHEW 1970b).

7.   **ECONOMICS OF NO_x CONTROL**

Since the application of control techniques to NO_x emissions in real-life situations is still extremely limited (except for exhaust gas recirculation in automobile engines), all published data on control costs are based on experimental work and design studies, and must be treated with caution. Furthermore, there are very few available data within Community member countries. The figures presented below are almost all from US sources, and should only be regarded as giving a broad indication of cost levels.

Stationary Sources: The most comprehensive presentation of the costs of NO_x emission control from stationary sources is made in Bartok et al (1971), from which the following analysis is taken.

Table 13 summarizes the degree of reduction in NO_x emissions theoretically achievable in large boilers of sizes varying from 120 to 1000MW, by the use of various combustion control techniques described in Section 6. In noting the figures in the table, which indicate that reductions in NO_x emissions of up to 90% are possible, emphasis must be placed on the word "theoretical", especially where coal-fired plants are concerned. As more development work is carried out, the figures given below can be expected to be drastically revised.

Table 13

Theoretical % NO$_x$ Reduction by Combustion Modification to Boilers of Various Sizes
(Source: Bartok et al, 1971)

Boiler size	LEA[1]	Two-stage	LEA+ two-stage	FGR[2]	LEA+ FGR	Water injection
1000 MW						
Gas	33	50	90	33	80	10
Oil	33	40	73	33	70	10
Coal	25	35	60	33	55	10
750 MW						
Gas	33	50	85	33	80	10
Oil	33	40	73	33	70	10
Coal	25	35	60	33	55	10
500 MW						
Gas	33	50	70	33	70	10
Oil	33	40	65	33	65	10
Coal	25	35	55	33	55	10
250 MW						
Gas	33	45	60	30	60	10
Oil	30	35	55	30	55	10
Coal	25	30	50	30	50	10
120 MW						
Gas	33	40	53	30	53	10
Oil	30	30	45	30	45	10
Coal	25	25	40	30	40	10

[1]LEA: Low excess air firing.

[2]FGR: Flue gas recirculation for NO$_x$ control.

Table 14 gives the estimated additional annual costs (capital amortised at 20% p.a., plus operating) of the modifications listed in Table 13. For 2-stage combustion the costs would range from zero for large gas and oil-fired units to the full cost of complete alteration or redesign for coal-fired plant. Flue gas recirculation costs assume a 15% maximum recirculation, and water injection is taken to incur a 1% loss in fuel efficiency. In those cases where installation of control equipment would result in a net saving, control costs are listed as negative.

Table 14

Estimated Extra Cost of Modified Boilers given in Table 13

(Source: Bartok et al 1971)

Control method	1000 MW plant	Annual control cost $1000	750 MW plant	Annual control cost $1000	500 MW plant	Annual control cost $1000	250 MW plant	Annual control cost $1000	120 MW plant	Annual control cost $1000
Low excess air	Gas	-95	Gas	-68	Gas	-35	Gas	-4	Gas	10
	Oil	-297	Oil	-226	Oil	-152	Oil	-44	Oil	-2
	Coal	-79	Coal	-59	Coal	-27	Coal	0	Coal	11
2-Stage combustion	Gas	0	Gas	24	Gas	44	Gas	52	Gas	54
	Oil	0	Oil	24	Oil	44	Oil	52	Oil	54
	Coal	299	Coal	240	Coal	176	Coal	103	Coal	60
Low excess air and 2-stage combustion	Gas	-95	Gas	-45	Gas	0	Gas	49	Gas	64
	Oil	-297	Oil	-203	Oil	-88	Oil	9	Oil	52
	Coal	220	Coal	181	Coal	148	Coal	104	Coal	72
Flue gas recirculation	Gas	202	Gas	160	Gas	109	Gas	58	Gas	24
	Oil	202	Oil	160	Oil	109	Oil	58	Oil	50
	Coal	202	Coal	160	Coal	109	Coal	58	Coal	30
Low excess air and flue gas recirculation	Gas	107	Gas	91	Gas	74	Gas	55	Gas	40
	Oil	-95	Oil	-67	Oil	-23	Oil	15	Oil	28
	Coal	123	Coal	101	Coal	82	Coal	59	Coal	41
Water injection	Gas	144	Gas	114	Gas	71	Gas	29	Gas	9
	Oil	179	Oil	141	Oil	87	Oil	56	Oil	11
	Coal	143	Coal	113	Coal	70	Coal	29	Coal	9

The apparent economic attraction of the low excess air method is clearly shown in the above figures.

Finally, Table 15 illustrates the derived control costs per ton of NO_x abated for the various control methods applied to a 1000 MW boiler operating at 70% load factor.

Table 15

Analysis of NO_x Control Costs for 1000MW Boiler given in Table 14

(Source: Bartok et al 1971)

Control method	% NO$_x$ Reduction	Fuel used	Cost per year $1,000	NO$_x$ Reduction 1000's of tons/yr	$/Ton NO$_x$	Mils/KWH
Uncontrolled (base case)	0	Gas	0	53.0	0	0
	0	Oil	0	30.0	0	0
	0	Coal	0	30.0	0	0
Low excess air	35	Gas	-95	17.5	-5	-0.02
	35	Oil	-297	9.9	-30	-0.05
	25	Coal	-79	7.5	-11	-0.01
Two-stage combustion	50	Gas	0	26.5	0	0
	40	Oil	0	12.0	0	0
	35	Coal	299	10.5	29	0.05
Low excess air plus 2-stage combustion	90	Gas	-95	47.7	-2	-0.02
	73	Oil	-297	21.9	-14	-0.05
	60	Coal	220	18.0	12	0.04
Flue gas recirculation	35	Gas	202	17.5	12	0.03
	35	Oil	202	9.9	20	0.03
	35	Coal	202	9.9	20	0.03
Low excess air plus flue gas recirculation	80	Gas	107	42.4	3	0.02
	70	Oil	-95	21.0	-5	-0.02
	55	Coal	123	16.5	8	0.02
Water injection	10	Gas	144	5.3	27	0.02
	10	Oil	179	3.0	60	0.03
	10	Coal	143	5.0	48	0.02

Control costs

For stationary gas engines, water injection into the intake manifold has promising potential for NO_x reduction. Table 16 shows the costs of such a system, based on water costs of 20c/1000 US galls., an injection rate of 1.25 lbs water/lb fuel, and 75% NO_x reduction.

Table 16
(Source: Bartok et al, 1971)

Theoretical NO_x Control Costs for Stationary Gas Engines

Engine hp	80	800	4200
Cost, $/year	60	800	930
Cost, $/ton NO_x eliminated	420	9.30	1.20

For small domestic combustion sources, a reduction of 60-70% in NO_x emissions is predicted by combining low excess air firing with flue gas recirculation. An anticipated conversion cost of $20 is quoted, though this seems a suspiciously low figure.

It must be stressed again that the above costs are based largely on design studies and limited experimentation. The technology remains to be proved on a large scale before the above costs can be considered firm. In particular the technical problems associated with modification of the combustion systems of coal fired plant may prove insuperable.

The alternative approach of flue gas treatment has also been costed. Table 17 illustrates the theoretical costs for a 1000MW coal-fired plant.

Table 17

Theoretical Costs of Flue gas treatment for 1000MW coal-fired plant

(Source: Bartok et al, 1971) (*)

Control process	Investment 10^6	Control costs		
		$/Ton coal	$/Ton NO_x	Mils/KWH
$Mg(OH)_2$ scrubbing	12.0	-0.06	-5	-0.02
H_2SO_4 scrubbing				
98% Scrubber efficiency	17.3	0.17	14	0.06
92% Scrubber efficiency	17.3	0.65	118	0.23
Limewater scrubbing[a]	8.5	0.50	45	0.18
NH_3 reduction and scrubbing[b]	15.0	0.92	66	0.33
H_2S reduction	11.7	0.98	77	0.34

[a] Single scrubber limewater control system with thermal regeneration of sorbent.

[b] Based on catalyst cost of $100/CF; space velocity 6000-7000 SCFH/CF.

Note (*) It is understood that a more recent paper by Bartok et al., presented at the International Congress on Chemical Engineering in the service of Man (Paris, September 1972) is much more pessimistic regarding the potential of flue gas treatment. Unfortunately the existence of this paper became known too late for its findings to be reviewed in the present report.

Mobile Sources: Information on the costs of NO_x emission control from mobile sources is very difficult to obtain, partially due no doubt to technical uncertainties but also to commercial sensitivity. The US Council on Environmental Quality has however published (CEQ 1971) estimates of the cost of meeting the original 1976 US standards (0.4 gms NO_x/mile); these are around $350 per vehicle in capital costs plus an

increase of $65 per year in operating and maintenance costs. In contrast to this, US automobile manufacturers have claimed (USAM, 1971) that the capital costs will be in the region of $500 per vehicle.

Somewhat lower costs are quoted in U.S. EPA (1972b), an extract from which is given in Table 18.

Table 18
Control Techniques and Estimated Investment Costs for Mobile
Source Emission Controls, 1967-1977
(Source: U.S. EPA 1972b)

Model Year	Autos and Light Duty Trucks. Typical Changes or Controls Added	Additional Cost per New Vehicle $	NO_x limit (g/mile)
1973-74	Exhaust gas recirculation (e.g.r.) for NO_x control, speed controlled spark timing.	48.00	3.1
1976-77	Low temperature NO_x decomposition catalyst unit (cost above controls supplanted, e.g. e.g.r.).	85.00	0.4
1976-77	e.g.r. increased to maximum for NO_x control. Modulation of recirculation. (Additional cost above previous simpler system).	14.00	0.4

The above costs are somewhat difficult to interpret and should be regarded with suspicion, especially since no successful NO_x catalyst has been developed.

In Western Europe, NO_x emission control costs have been estimated in France (Frybourg 1971) to be about 1% of the price of a car to achieve a 30% reduction in the presently uncontrolled levels of NO_x emission.

Increases in fuel consumption are estimated to have risen by 3 to 12% in the US to meet the current limit of 3.1 gms NO_x mile, over uncontrolled engines (U.S. NAS 1972) with

progressively higher fuel consumption penalties as the limits are tightened.

8. REFERENCES

BAGG J. (1971) "The Formation and Control of Oxides of Nitrogen in Air Pollution". Paper in: Air Pollution Control, Part 1 (Edited by W. Strauss). Wiley-Interscience, New York.

BAGWELL, F.A. ROSENTHAL, K.E., et al (1970). "Oxides of Nitrogen Emission Reduction Program for Oil and Gas Fired Utility Boilers". Paper presented at 32nd Annual Meeting of American Power Conf., Chicago, April 1970.

BARTOK W., CRAWFORD A.R., CUNNINGHAM A.R., HALL H.J., MANNY E.H., and SKOPP A., (1971). "Stationary Sources and control of Nitrogen Oxide Emissions". Paper presented at 2nd International Clean Air Congress, Washington D.C., U.S.A. Dec 6-11, 1970.

CEQ (1971) "Phase II of the Economic Impacts of Meeting Exhaust Emission Standards, 1971-1980". Report prepared for U.S. Council on Environmental Quality by Chase Econometric Associates, Inc., Dec 1, 1971.

DERWENT R.G. and STEWART H.N.M. (1973). Air Pollution from the Oxides of Nitrogen in the United Kingdom". Paper in: Atmospheric Environment Vol 7, No. 4, April 1973, Pergammon Press, Oxford, England.

EEC (1972a). The Community Oil Sector Medium-Term Forecast and Guidelines. Commission of the European Communities Report SEC(72)3173 final. Brussels, September 1972.

EEC (1972b). Medium Term Prospects and Guidelines in the Community Gas Sector. Commission of the European Communities Report SEC(72)3182 final. Brussels, September 1972.

EEC (1972c) Prospects of Primary Energy Demand in the Community (1975-1980-1985). Commission of the European Communities Report SEC(72)3283 final. Brussels, October 1972.

FRYBOURG M. (1971). "Automobiles et Nuisances: pour un programme d'action". Report to the Prime Minister of Ad Hoc Working Group, Paris, August 1971.

FUSSELL D.R. (1970). Atmospheric Pollution from Petrol and
Diesel engined Vehicles. Petrol Review, Vol 24,
pp192-202.

GEORGE R.E. and BURLIN R.M. (1960). Air Pollution from
Commercial Jet Aircraft in Los Angeles County.
Air Pollution Control District, Los Angeles County.

JUNGE C.E. (1963). Air Chemistry and Radioactivity, Academic
Press, New York, p123.

MILLS J.L., LEUDTKE K.D., WOOLRICH D.F., AND PERRY L.B. (1961).
Emissions of Oxides of Nitrogen from Stationary
Sources in Los Angeles. Report Nos 1-4, Los
Angeles County Air Pollution District.

NREC (1968)   Nature and Composition of Aircraft Engine Exhaust
Emissions. Report 1134/1, November 1968.

OECD (1972)   Report of Joint Ad Hoc Group on Air Pollution from
Fuel Combustion in Stationary Sources. Organisation
for Economic Cooperation and Development Report
PAC/70.7 (Final Revision), Paris, August 1972.

OECD (1973)   Energy Committee Forecasting Exercise: Up-Dating
of the 1980 Exercise and Sectoral Results for
1985. Organisation for Economic Cooperation and
Development Report DIE/E/EN/73.127 Scale B, Paris,
September 1973.

ROBINSON E., and ROBBINS R.C., (1968). Sources, Abundance and
Fate of Gaseous Atmospheric Pollutants. Stanford
Research Institute, USA, February 1968.

ROBINSON E., and ROBBINS R.C. (1971). Sources, Abundance, and
Fate of Gaseous Atmospheric Pollutants, Supplement.
Stanford Research Institute, USA, April 1971.

SMITH D.S., SAWYER R.F., and STARKMAN E.C. (1968). J. Air
Pollution Control Association, Vol 18, No. 30.

STIEF-TAUCH P., Letter to TAYLOR K. dated 5th October 1973,
enclosing "Perspectives de consommation de Houille
en 1985 (CEE à 9)".

U.S.A.M. (1971) U.S. Automobile Manufacturers' Communications to
Ad Hoc Committee on Cumulative Regulatory Effects
on the Cost of Automotive Transportation (RECAT).
Office of Science and Technology, February 1972.

U.S. DHEW (1970a). Control Techniques for Nitrogen Oxide Emissions from Stationary Sources. US Department of Health, Education and Welfare. National Air Pollution Control Administration Publication No. AP-67. Washington D.C., U.S.A., March 1970.

U.S. DHEW (1970b). Control Techniques for Carbon Monoxide, Nitrogen Oxide, and Hydrocarbon Emissions from Mobile Sources. US Department of Health, Education and Welfare. National Air Pollution Control Administration Publication No. AP-66. Washington D.C. U.S.A., March 1970.

U.S. EPA (1972a). Compilation of Air Pollutant Emission Factors (Review). US Environmental Protection Agency, February 1972.

U.S. EPA (1972b). "The Economics of Clean Air". Annual Report of the Administrator of the U.S. Environmental Protection Agency to Congress, March 1972.

U.S. OST (1972). Cumulative Regulatory Effects on the Costs of Automotive Transportation (RECAT). Final Report of the Ad Hoc Committee. Office of Science and Technology, February 1972.

U.S. NAS (1972). Semi-annual Report by the Committee on Motor Vehicle Emissions of the National Academy of Sciences, Washington, January 1972.

APPENDIX: NO$_x$ EMISSION FACTORS

This Appendix lists the basic data on NO$_x$ emission factors used to compile the figures in Table 4.

There are five reference sources:

1. Derwent and Stewart (1973).
2. OECD (1972).
3. US: EPA (1972a).
4. US: DHEW (1970a).
5. Bagg (1971).

Of these, reference 3: "Compilation of Air Pollutant Emission Factors (Revised)" by the US Environmental Protection Agency, 1972, probably covers the whole air pollution field more comprehensively than any other single document. However, the data refer to US conditions and must be used with care where the European situation is concerned. This applies particularly to automobile emissions.

The most up to date review of NO$_x$ emissions is to be found in reference 1: "Air Pollution from the Oxides of Nitrogen in the United Kingdom" by Derwent and Stewart (Atmospheric Environment, 7, 4, April 1973, pp 385-401).

Reference 3: "Report of Joint Ad Hoc Group on Air Pollution from Fuel Combustion in Stationary Sources" by OECD (PAC/70.7), Paris, 1972, is a valuable compendium of information on air pollution from OECD member countries.

Reference 4: "Control Techniques for Nitrogen Oxide Emissions from Stationary Sources" by the US Department of Health, Education and Welfare, 1970, is a well-known standard work on the subject.

Reference 5: "The Formation and Control of Oxides of Nitrogen in Air Pollution" by Bagg, which constitutes a paper in the book "Air Pollution Control", Part 1, Ed. Strauss (Wiley-Interscience 1971) gives much detailed discussion of NO$_x$ emission results not conveniently collected elsewhere. The basic data from these references is arranged below in the sectors of fuel use given in Table 4.

Electricity Generation

Ref 1 (Table 5, p.391)

Fuel \ NO$_x$	ppm in exhaust		g/Kg fuel	Kg/t.o.e.
	Range	Mean		
Oil	180–900	400	7.6	7.6
Coal	100–1460	400	6.8	9.7*

*Assumes 1 tonne coal $\equiv$ 0.7 tonne oil

Ref 2 (Table 7, p.12)

Fuel \ NO$_x$	% by weight o.e.		Kg/t.o.e.
	Range	Mean	
Solid	0.6 – 1.4	1.1	11
Liquid	0.7 – 1.5	1.1	11
Gas	0.3 – 1.4	0.6	6

Ref 3 (Tables 1-2, 1-5, 1-6)

Fuel \ NO$_x$	Kg/unit quantity of fuel		Kg/t.o.e.
	Range	Mean	
Solid	9–28/tonne	12	17
Liquid	12.6/M^3	12.6	13
Gas	6,250/10^{6}M^3	6,250	7

Ref 4 (Table 8-1)

Fuel \ NO$_x$	lbs/unit quantity of fuel	kg/t.o.e.
Solid	20/ton (US)	14
Liquid	104/1000 galls (US)	13
Gas	390/10^{6}ft^3	7

Ref 5

NOₓ / Fuel	lbs/unit quantity of fuel		kg/t.o.e.
	Range	Mean	
Solid	1.7- 26/1000 lbs	8	11
Liquid	13-14/1000 lbs	13.5	13.5
Gas	"half oil emissions"	6	6

Domestic and Commercial Heating

Ref 1 (Table 5)

NOₓ / Fuel	Kg/unit quantity of fuel	Kg/t.o.e.
Solid	1.4/tonne	2
Liquid	4/tonne	4
Gas	0.04g/MJ*	1.7

*42MJ per Kg oil

Ref 2 (Table 7, p.12)

NOₓ / Fuel	% by weight o.e. in exhaust		Kg/t.o.e.
	Range	Mean	
Solid	0.6 – 1.2	0.68	6.8
Liquid	0.06 – 6	0.28	2.8
Gas	0.09 – 1.43	0.30	3.0

Ref 3 (Tables 1-2, 1-5, 1-6)

NOₓ / Fuel	Kg/unit quantity of fuel		Kg/t.o.e.
	Range	Mean	
Solid	3/tonne	3	4.3
Liquid	$1.5/M^3$	1.5	1.8
Gas	$800-1600/10^6 M^3$	1200	1.3

Ref 4 (Table 8-1)

NO$_x$ / Fuel	lbs/unit quantity of fuel		Kg/t.o.e.
	Range	Mean	
Solid	8/ton (US)	8	5.7
Liquid	12-72/1000galls (US)	42	5 .
Gas	116/10^{6}ft^3	116	2

Ref 5

NO$_x$ / Fuel	lbs/unit quantity of fuel (mean)	Kg/t.o.e.
Solid	0.2/10^{6}BTU	2.9
Liquid	8/1000 lbs	8
Gas	0.1/10^{6}BTU	1.8

Industrial Heating

Ref 1 (Table 5)

NO$_x$ / Fuel	Kg/unit quantity of fuel	Kg/t.o.e.
Solid	6/tonne	8.6
Liquid	9.8/tonne	9.8
Gas	0.061g/MJ	2.6

Ref 2 (Table 7)

NO$_x$ / Fuel	% by weight o.e. in exhaust		Kg/t.o.e.
	Range	Mean	
Solid	0.6 - 1.4	1.1	11
Liquid	0.7 - 1.5	0.9	9
Gas	0.3 - 1.4	0.4	4

Ref 3 (Tables 1-2, 1-5, 1-6)

NOₓ / Fuel	Kg/unit quantity of fuel		Kg/t.o.e.
	Range	Mean	
Solid	7.5/tonne	7.5	11
Liquid	$4.8-9.6/M^3$	7.2	7.7
Gas	$1920-3700/10^6 M^3$	2810	3.1

Ref 4 (Table 8-1)

NOₓ / Fuel	lbs/unit quantity of fuel	Kg/t.o.e.
Solid	20/ton (US)	14
Liquid	72/1000 galls (US)	8.6
Gas	$214/10^6 ft^3$	3.8

Ref 5

NOₓ / Fuel	lbs/unit quantity of fuel (mean)	Kg/t.o.e.
Solid	$0.8/10^6$ BTU	11
Liquid	10/1000 lbs*	10
Gas	$0.16/10^6$ BTU	2.9

* Interpolated figure

Transport

US data are not relevant here, since in general automobile engines are much larger than for European cars. Also, NO_x emissions from vehicles are controlled whereas there is no comparable legislation in Western Europe. Thus, only references 1 and 5 apply.

Ref 1 (quoting Fussell, 1970) gives:-

Engine	ppm NO in exhaust		Kg/t.o.e.
	range	mean	
Petrol	30-6000	1000	16
Diesel	100-1000	500	11

Ref 5 (quoting results for non-US compact cars) gives:-

Engine	ppm NO in exhaust		Kg/t.o.e.
	Range	Mean	
Petrol	50-3400	1200	19
Diesel	30-850	450	11

In order to obtain an overall emission factor weighted relative to the amounts of petrol and diesel fuel used, the data in the Commission document SEC(72)3283 final: "Prospects of Primary Energy Demand in the Community, 1975, 1980, 1985" were used (Appendix 4, pp.39 and 42). From this it is seen that the weight ratio of petrol: diesel consumed rises from 2.0 in 1970 to 2.3 in 1985.