

MEASUREMENT,
DETECTION AND
CONTROL OF
ENVIRONMENTAL
POLLUTANTS

PROCEEDINGS OF A SYMPOSIUM

15-19 MARCH 1976

PROCEEDINGS SERIES

**MEASUREMENT, DETECTION
AND CONTROL OF
ENVIRONMENTAL POLLUTANTS**

**PROCEEDINGS OF AN INTERNATIONAL SYMPOSIUM
ON THE DEVELOPMENT OF NUCLEAR-BASED TECHNIQUES
FOR THE MEASUREMENT, DETECTION AND CONTROL
OF ENVIRONMENTAL POLLUTANTS
HELD BY THE
INTERNATIONAL ATOMIC ENERGY AGENCY
IN VIENNA, 15-19 MARCH 1976**

**INTERNATIONAL ATOMIC ENERGY AGENCY
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MEASUREMENT, DETECTION AND CONTROL
OF ENVIRONMENTAL POLLUTANTS

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FOREWORD

Environmental pollution is the consequence of socio-economic activities and natural changes. Intensive industrialization, massive urbanization and accelerated consumption of fossil fuels have speeded up pollution to an alarming degree. There *must* be restrictions on the emission of air-borne particulate matter, release of noxious gases and dispersion of toxic substances in water systems and on land: it is necessary to take preventive measures to protect the meteorological and ecological systems against the threat of constant but unabated pollution. In this alarming situation the words of Ben Jonson seem particularly apt: "But if once we lose this light, 'tis with us perpetual night"

Following the Symposium on the Use of Nuclear Techniques in the Measurement and Control of Environmental Pollution in October 1970, the International Atomic Energy Agency convened the present Symposium to review the current status of the development of nuclear analytical methods and tracer techniques in their contribution to the monitoring of environmental pollutants. The participants heard and discussed 41 papers related to the activation analysis of air-borne particulate matter and water pollutants, X-ray fluorescence techniques and tracer technology in the investigations of air and water pollutants. In addition, two General Discussion sessions were arranged. In these, three topics were thrown open for an exchange of views: intercomparison studies in multielement analysis; analytical techniques based on electron excitation; and the role of tracer technology in pollution studies. It is hoped that the Proceedings will throw light on how more could be done to improve the quality of the environment.

EDITORIAL NOTE

The Editor of this journal has been advised by the International Atomic Energy Agency that the present Symposium is the first of a series of such symposia to be held in the field of environmental pollution. The Agency has expressed its interest in the development of the nuclear analytical methods and tracer techniques in their contribution to the monitoring of environmental pollutants. It is hoped that the present Symposium will be a landmark in the development of these techniques and that the results of the symposium will be widely disseminated through the present journal. The Editor of this journal has been advised by the International Atomic Energy Agency that the present Symposium is the first of a series of such symposia to be held in the field of environmental pollution. The Agency has expressed its interest in the development of the nuclear analytical methods and tracer techniques in their contribution to the monitoring of environmental pollutants. It is hoped that the present Symposium will be a landmark in the development of these techniques and that the results of the symposium will be widely disseminated through the present journal.

FOREWORD

The International Atomic Energy Agency (IAEA) was established in 1957 under the aegis of the United Nations. Its primary purpose is to promote the peaceful uses of atomic energy and to prevent the spread of nuclear weapons. The Agency's work is carried out through a number of technical cooperation projects, research and development activities, and the provision of technical assistance to member States. The Agency's activities are financed by contributions from its member States and by the sale of nuclear fuel and other products. The Agency's work is carried out through a number of technical cooperation projects, research and development activities, and the provision of technical assistance to member States. The Agency's activities are financed by contributions from its member States and by the sale of nuclear fuel and other products.

EDITORIAL NOTE

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Session I

**COMPREHENSIVE INVESTIGATIONS ON
THE COMPOSITION OF AIR POLLUTANTS**

Chairman

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ORIGIN AND MONITORING OF POLLUTANTS IN FOSSIL-FUEL FLAMES

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Abstract

ORIGIN AND MONITORING OF POLLUTANTS IN FOSSIL-FUEL FLAMES.

A review is given of the origin of pollutants in fossil-fuel flames. Burning of fossil fuels is the major cause of air pollution and significant reductions in levels of environmental pollution can be achieved by more effective control of combustion systems. The chemical kinetics of formation of unburned hydrocarbons, oxides of nitrogen, carbon monoxide and particulate matter are described, as well as the reactions which can lead to oxidation and destruction of these pollutants within the flame. The important influence of mixing and aerodynamics is discussed, together with methods of mathematical modelling and prediction methods. Practical problems arising in gas turbine engines, spark ignition engines and diesel engines are investigated in order to minimize the emission of pollutants while preserving fuel economy.

INTRODUCTION

The major cause of air pollution is the emission of pollutants from combustion systems burning fossil fuels. These arise in the combustion of oil, coal and gas in the furnaces of electricity generating power stations and industrial plant as well as in automobiles, trucks and aircraft. In large industrial complexes and urban areas, deterioration of the environment due to pollution has reached such high levels that the largest cities in the world have been threatened with suffocation. Damage to property and landscape has reached such high proportions that the public has demanded a reduction in emission of pollutants. Under pressure from environmentalist lobbies, legislation has been introduced in the United States and other industrialized countries restricting the quantity of pollutants emitted from engines and industrial plant. The restrictions have been imposed with increasing severity and huge financial resources are being expended on changing engineering design, as well as monitoring and controlling the emission of pollutants.

The environment in many major cities in England has been radically improved simply due to the control of domestic coal fire burning. Fog and smog levels have been reduced, sunshine has been increased, bird and plant life have reappeared and the quality of life has improved. In the largest Californian cities, where automobile exhaust fumes are the main cause of pollution, the rates of increase of levels have been reduced and can be expected to decrease in the near future. Some of the initial attempts at reducing pollutants from automobiles resulted in a decrease in combustion efficiency. The energy crisis of 1974 followed by the fivefold increase in the price of oil emphasized the need for fuel conservation so that solutions proposed for the reduction of emission of pollutants must also result in high combustion efficiency.

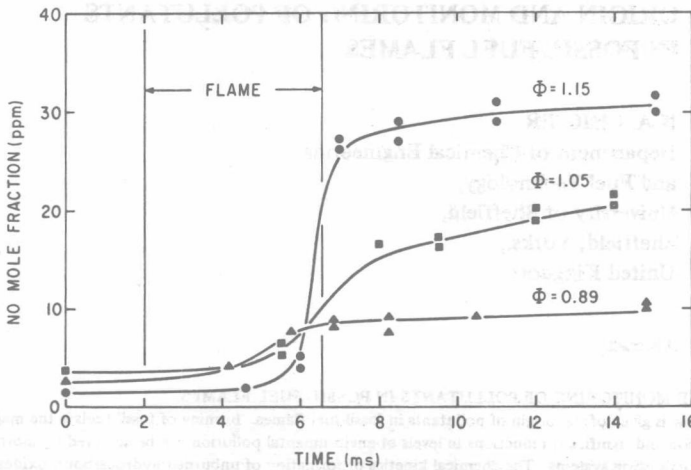


FIG.1. Measured nitric oxide profiles in methane-air flames demonstrating the importance of NO formation in the post-flame zone. NO formation increases as the fuel-air equivalence ratio, ϕ , increases (Bowman [3]).

The subject of pollution formation and destruction in flames has been examined in detail by a series of authors in Volume 1 of Progress in Energy and Combustion Science. It is demonstrated that very considerable reductions in emissions of pollutants can be achieved within the combustion chamber and that a large proportion of the pollution of the environment has been due to inefficient combustion of fossil fuels. The emphasis in this review is therefore on the reduction of emissions at source. More detail concerning the subject can be found in reference 1.

PRINCIPAL POLLUTANTS

Emissions from engine exhausts and chimney stacks are mainly in the gaseous phase with small quantities of solid particulate matter held in suspension. The gas constituents are made up of inert gases which have been passed through the combustion chamber unchanged, products of combustion and unburned fuel and oxidant. The solid particulate matter arises from the fuel and consists of metallic compounds and other materials which cannot burn and hydrocarbons which were not completely burned in the flame. Under very poor combustion conditions, liquid fuel may be emitted when full throttle is used during rapid accelerations. Since pollution is defined as the contamination of man's environment, all matter emitted from combustion chambers that alters or disturbs the natural equilibrium of the environment must be considered as a pollutant. The major concern is to prevent damage to human, animal and plant life and generally to create no artificial disturbance to the natural ecosystem.

Pollutants can arise both as products of combustion and also as impurities in the fuel which are not burnt during the process of combustion. Unburned hydrocarbons arise from the incomplete combustion of fuel due to the fuel rich operation or inadequate mixing of fuel and air, as does the formation of carbon during the combustion process. Oxides of nitrogen are formed during the combustion process, the source of nitrogen either being from nitrogen contained within the fuel or fixation of nitrogen from the air supply for the combustion process.

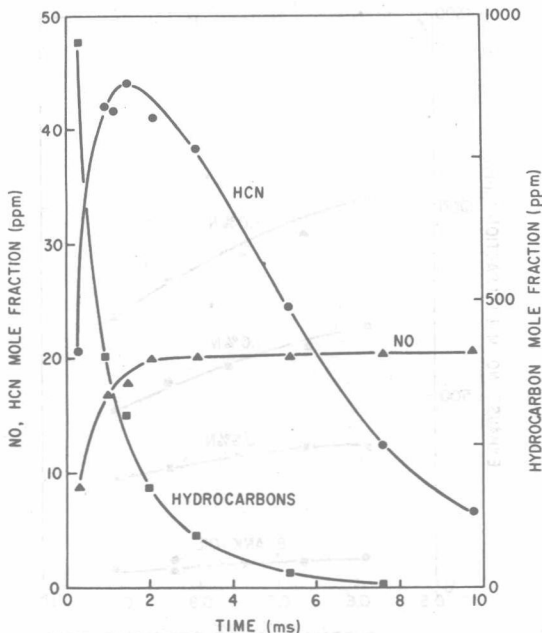


FIG.2. Large concentrations of HCN are measured near the flame reaction zone of fuel-rich flames. Rapid formation of NO corresponds with the appearance of HCN (Bowman [3]).

The four principal chemical classes of pollutant species emitted from combustion sources are nitrogen oxides, carbon monoxide, organic compounds (unburned and partially burned hydrocarbons), and sulphur oxides. As part of the combustion process, pollutant species are formed by chemical reaction and subsequent chemical reaction can also lead to their destruction or elimination. The formation of carbon monoxide, soot and other pollutant species, may be controlled to constitute intermediates within the combustion system, which can be destroyed by chemical reaction prior to emission from the system. At the temperature achieved in most flames, the presence of fuel and oxidant leads inevitably to the formation of some pollutants. These high concentration levels may be reduced by further chemical reaction as the temperature is reduced, but quenching leads to the freezing of certain species, resulting in high emission rates. Pollution formation, destruction and control is thus intimately connected with the combustion process. The general aims of maximizing combustion efficiency and minimizing pollutants are conflicting, since combustion efficiencies are maximized at, or close to, stoichiometric mixing conditions where the highest temperatures are achieved. These high temperatures lead to formation of high levels of pollutants. Optimization can only be achieved by very careful control of air-fuel ratio and temperature levels throughout the system.

Particulates

The particulate matter emitted from combustion chambers has three possible sources: (i) matter which was not combustible; (ii) matter which was capable of being burned but was not burned; and (iii) matter formed

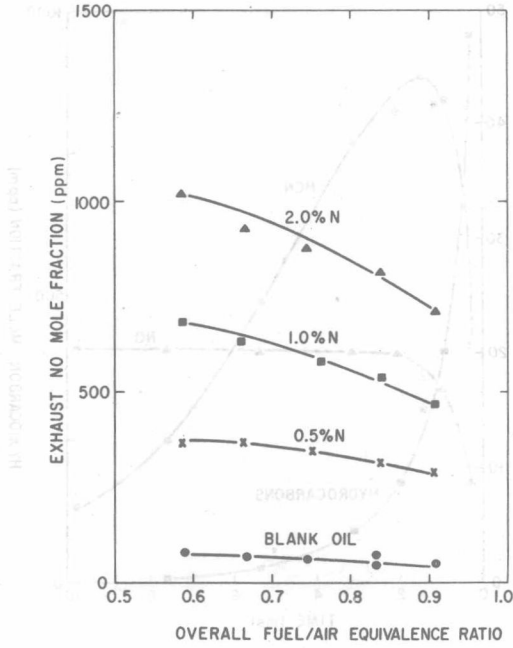


FIG. 3. NO concentrations in the flue gas of an oil-fired furnace show the effect of adding organic nitrogen compounds to the fuel (Bowman [3]).

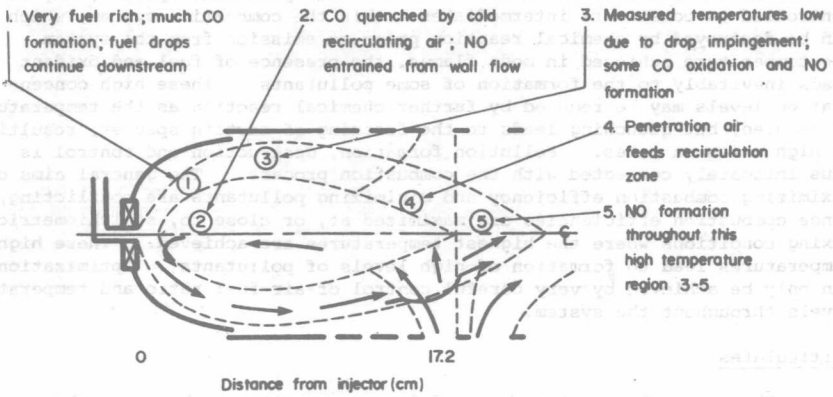


FIG. 4. Formation and oxidation of CO and NO in gas turbine combustion chambers (Mellor [6]).

during the process of combustion. Temperature conditions in most combustion chambers are sufficiently high for vaporization of liquids so that, except for conditions of very rich low temperature burning, all emitted particulate matter will be in the solid phase. Particulate matter can be deposited on surfaces within the combustion chamber or may be emitted with the exhaust gases. Most pollution problems arise from particulates which are sufficiently small that they are held in suspension as they are transported by the exhaust gases. The particulate matter may be clearly visible as smoke or water vapour, but small concentrations of particles can be significant sources of pollution without being clearly visible.

Almost all hydrocarbon fuels contain traces of metals and other solid matter, which normally do not burn and pass through the combustion chamber as an element or, more generally, as a metal oxide. Most of these metals are capable of burning and are even used as fuels in rockets and other high temperature devices. Carbon has a melting point of 3813K, yet solid carbon burns easily in air, due to its low thermal conductivity. Most of the particulate matter emitted from practical combustion devices is solid carbon. Solid carbon particles formed in flames are known as soot. Soot can be formed from purely gaseous fuels and is more commonly formed when liquid fuels are used. Carbon particles are the main cause of luminosity of flames and the emission from the yellow region of flames is due to carbon. For flames that rely on radiative heat transfer, attempts are thus made to promote the formation of soot particles. In automobile engines and other systems with water-cooled surfaces, coking, due to the deposition of hard carbon deposits, can readily arise when fuel is allowed to come into direct contact with the cooled surfaces. In gas turbine combustion chambers, formation of solid particles is generally considered to be undesirable because radiative heat transfer from the flame to the combustor-can walls would require additional cooling and also the presence of particles causes damage to gas turbine blades, either by direct impingement of the particles, causing pitting of the blades, or by deposition on the blades, leading to subsequent corrosion especially in marine applications.

In industrial furnaces, flames are separated into two main regions - a luminous region, where particles are present directly as fuel particles, or have been formed by the combustion process and, subsequently, a region where sufficient oxygen is supplied to the particles in order to complete their combustion before leaving the combustion chamber. In addition to the carbonaceous particles responsible for the main continuous radiation, some contribution from banded emission is made by large carbon and hydrocarbon molecules in regions close to the reaction zone. The mechanisms for the formation of soot involve the dehydrogenation of organic compounds and polymerization leading to formation of large carbonaceous particles. There are distinct differences in the formation mechanisms of diffusion flames and pre-mixed flames. For very small diffusion flames on circular burners, luminous regions appear as the mass flow is increased and further increase in mass flow leads to the formation of soot at the top. For paraffins, the tendency to smoke increases with molecular weight but the reverse is true for the olefine, di-olefine, benzene and naphthalene series. Primary alcohols form more soot as the molecular weight is increased and secondary alcohols produce more soot than primary alcohols. The C/H ratio is the most important parameter, but molecular structure also plays an important role. For the same C/H ratio, branched chain paraffins produce more smoke than the corresponding normal isomers. Thus iso-octane produces flames of higher luminosity than n-octane. Flame temperature affects soot formation in two ways; higher flame temperatures provide greater temperature gradients, which favour the formation of soot particles but also the higher flame temperatures lead to faster burning. The low luminosity found in benzene and toluene flames has been explained as being due to the reduction in flame temperature as a result of soot liberation,

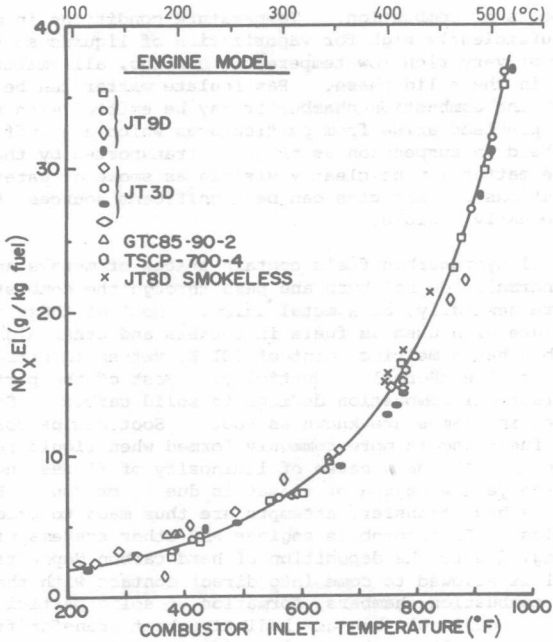


FIG.5. Increase in combustor inlet temperatures in gas turbine engines leads to substantial increase in NO_x emission index (Mellor [6]).

whereas in flames with oxygen, the fuel is decomposed in the preheating zone and there is no direct contact between undecomposed fuel and oxygen because they are separated by combustion products. Sufficient residence time is also required for carbon formation. For methane, carbon is formed in regions of relatively high temperature, where the particles are consumed rapidly without formation of soot. Acetylene decomposes at a lower temperature and carbon particles form in cooler regions and are more likely to form soot because of the lower concentration of oxygen-containing substances. Turbulence leads to an increase in the formation of carbon and also to a more rapid burning of the carbon particles. Control of the mixing distribution in a turbulent flame allows control of soot formation and soot burning.

In premixed flames where there is sufficient oxygen to react with carbon to form carbon monoxide, no carbon will be formed provided the temperature is sufficiently high and the residence time is sufficiently long. When there is insufficient oxygen to react with all the carbon to form CO, then carbon will be found at the end of the flame. The formation and burning of carbon is so dependent upon local temperature and mixture ratio conditions that, unless these are clearly known or specified, it is not possible to determine the local concentrations of carbon particles.

Carbon luminosity occurs at mixtures which are much less rich than those required to liberate free carbon under equilibrium conditions. On an air/fuel ratio basis, the tendency to form carbon increases in the order: aldehydes;