Keynote address

Ref 205

Progress in pollution abateme ... n cokemaking industry

R. Fisher

This paper reviews some recent research and development work that has been carried out on the control of air and water pollution within the European cokemaking industry. In the carbonisation process itself, diffuse emissions from battery doors, lids, and ascension pipe seals and from battery operations, such as oven charging and pushing and coke quenching, are significant sources of pollution. Collaborative investigations undertaken by various research groups are reported which demonstrate the effectiveness of modern battery design backed up by good operating practices. In the purification of carbonisation effluents, considerable advances have been made in the control of treatment processes. This is highlighted by developments in the control of ammonia stripping operations and by the enhancement of biological treatment facilities to include nitrification and denitrification of the waste water. However, as environmental constraints become tougher, there will be an increasing need to consider new concepts in cokemaking technology and waste water treatment. IS/1026

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INTRODUCTION

Over the past 20 years or so, a large amount of research and development work has been carried out by the carbonisation industry aimed specifically at improving pollution control. In Europe, the European Coal and Steel Community (ECSC) has provided generous financial support for developments in pollution control carried out by the European cokemaking industry. Much of the research work carried out under the auspices of the ECSC has been on a multinational basis, with complementary studies being undertaken by two or more research groups.

The European Coking Industry Environmental Working Group has prompted a number of these collaborative research studies under ECSC sponsorship. Other members of the Group have also carried out individual ECSC research projects. The Industry Working Group was set up in 1978 as a subcommittee of the European Cokemaking Committee, which had been established four years earlier as a platform for sharing knowledge and experience to overcome technical problems in cokemaking. During its early meetings, the European Cokemaking Committee realised rapidly that there was a need for a specialist group to deal specifically with the increasing environmental pressures facing the industry; hence, the Environmental Working Group came into being. The objectives of this paper are twofold. First, to review some of the important environmental studies that have been carried out during the last decade and, second, to demonstrate the value of ECSC research funding in investigating and developing practical solutions to environmental problems.

AIR POLLUTION CONTROL

Sources of air pollution on coking plants may be defined broadly as either continuous or fugitive. The main sources of continuous emission are those that originate from combustion processes such as battery underfiring, steam raising, and ammonia incineration. Fugitive emissions may be subdivided into three categories. First, the discontinuous emissions that are linked to specific operational processes such as the charging and pushing of ovens and the wet quenching of hot coke. Second, the diffuse emissions that arise from leakages at the charging holes, ascension pipes, and oven doors, or from vents on storage tanks in the byproducts plant. Finally, the fugitive emissions that occur from open sources, such as stockpiles, under windy conditions. In most countries, it is the fugitive emissions which generally give rise to complaints from the general public and which, because of the way in which they arise, are usually the most difficult to control.

Continuous emissions

The main pollutants present in the underfiring waste gases are oxides of sulphur (principally sulphur dioxide), oxides of nitrogen (NO_x, principally in the form of nitric oxide), and particulates. Sulphur dioxide emissions are best controlled by desulphurisation of the coke oven gas when it is used for underfiring. The processes for achieving this are well known and do not require further description here. The most widely applied methods in Europe are probably ammonia based removal processes[1,2] and the Stretford process.[3] A major disadvantage of the latter is disposal of the blowdown, owing to its volume and high concentration of ammonium or sodium thiocyanate. The liquor is biodegradable at high dilutions, but sufficiently high dilution is usually only possible in large scale biological treatment plants, such as municipal sewage treatment facilities. An alternative approach, adopted at the Serémange Coke Works of Lorfonte in France, is to dispose of the waste liquor by adding it to the coal blend.[4] Operating in this way has not caused any problems so far with oven refractories or coke quality.

The control of NO_x emissions cannot be achieved so easily, since the NO_x is formed principally by oxidation of nitrogen in the combustion air under the high temperatures prevailing in the heating flues (thermal NO_x), rather than the oxidation of chemically combined nitrogen in the fuel (fuel NO_x). Low NO_x burners have been applied successfully at British Steel General Steels' Scunthorpe Works and at other European coking plants. The new Kaiserstuhl III coking plant, Dortmund, Germany, which is currently under construction, includes a new type of oven design[5] that incorporates well established NO_x control techniques, such as staged combustion and partial waste gas recirculation.

It also includes a new type of silica refractory brick, having a higher thermal conductivity that may lead to temperature reductions of 50–80°C in the heating flues while maintaining the same rate of advance of the coking process.

Particulate matter in stack emissions is mainly soot arising from incomplete combustion. Sometimes, this may be the result of poor setting or maintenance of the burners. However, the main cause is gas leakage from the ovens to flues or in regenerators and is usually found only in older batteries. The application of silica welding[6] can help to overcome these problems and extend battery life.

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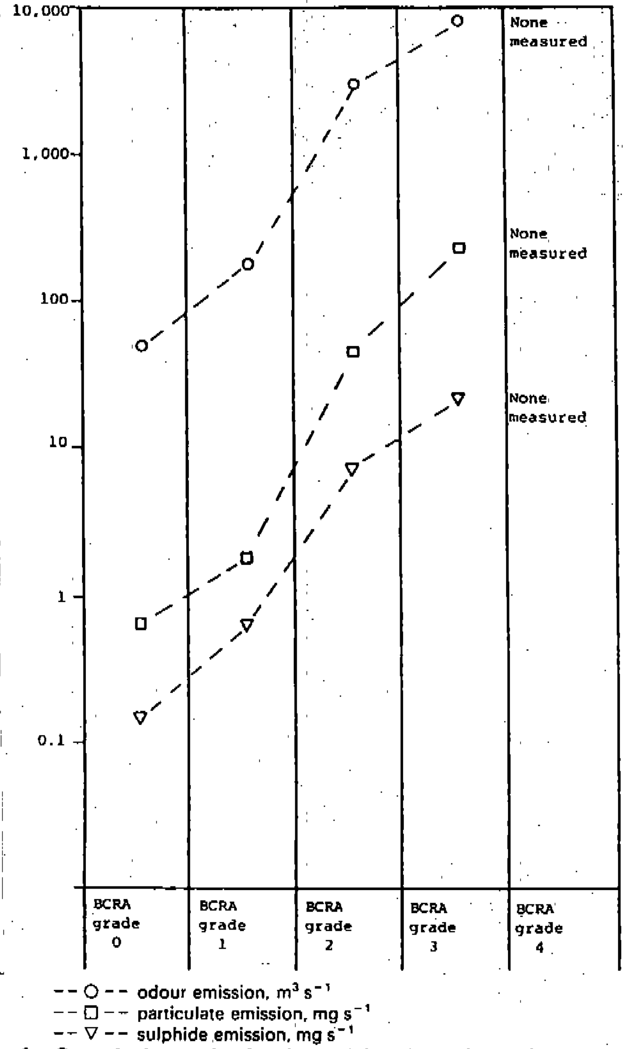
Belgium in the work on heavy metals, and by the Spanish organisation Empresa Nacional Siderúrgica SA (ENSIDESA) for part of the work on PAHs. The odour impact studies were carried out jointly by LECES, BCRA Scientific and Technical Services Ltd from the UK, and Société Belge de Filtration (SBF) from Belgium.

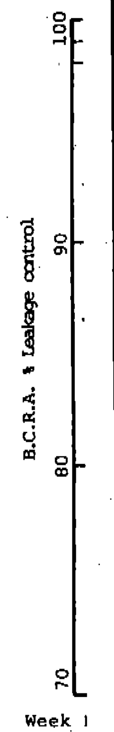
The fate of heavy metals in the coking process was investigated, because the German TA Luft regulations specify strict limits on various heavy metals in three classes. During the investigation, it was established that beryllium, arsenic, cobalt, nickel, antimony, chromium, copper, vanadium, and manganese were largely retained in the coke, whereas cadmium, mercury, thallium, lead, and zinc were more concentrated in the tar. Heavy metal concentrations in the waste water were negligible. It was demonstrated that the emissions of heavy metals from wet quenching towers also met the TA Luft limits for all classes of heavy metals. Heavy metal exposures in the workplace were several orders of magnitude less than the specified occupational exposure limits.

The main source of PAHs was the battery itself; the contribution of the byproducts plant was small compared with the overall level of PAH emissions. On old 4·5 m coke ovens, the total emission of PAHs amounted to 7·8 g of PAH per tonne of coke produced, of which approximately 48% was attributed to door leakage and 42% to leaking charge hole lids. Similarly, with BTX and odour emissions, leakages from oven doors and lids made significant contributions to the overall emission level, although emissions from storage tank vents in the byproducts area were also important with regard to these compounds. In the work carried out by BCRA on odoriferous emissions, door leakages were sampled, using a specially designed hood to capture the emissions, and very useful correlations were established between the emissions of odour, particulates, and hydrogen sulphide and the visible appearance of the smoke leakage as assessed by the BCRA method[11] (*see* Fig. 1). It is fairly certain that similar correlations would exist between PAHs, BTX, and visible smoke leakage assessment.



1 Correlations obtained on coke oven doors between visible grades of smoke leakage (BCRA 0–4) and measured mean rates of odour, particulate, and sulphide emission

In all of these studies, it was demonstrated that the emissions of BTX, PAHs, and odoriferous compounds were reduced considerably on modern coking plants having selfsealing doors.[12,13] With regard to PAHs, for example, emissions from tall ovens with modern selfsealing door designs were typically only 1–2% of those from ovens having older doors. Battery emissions of BTX on modern plants were also reduced to a level of less than 6% of those from older installations. However, good door design in itself is not sufficient to ensure good leakage control performance. Of equal importance is good plant management and a strict routine for cleaning all sealable ports. At British Steel General Steels' Redcar Works, oven door and oven top leakage control have averaged greater than 99% since start-up of the rebuilt ovens in 1984. The installation of water sealed ascension pipe caps has also produced a 100% record on ascension pipe cap leakage control. This performance has generally been attributed to the use of high pressure water jet oven door cleaning,[14] together with the strict discipline of cleaning every time a door is removed. The importance of the door cleaning schedule was demonstrated vividly during work carried



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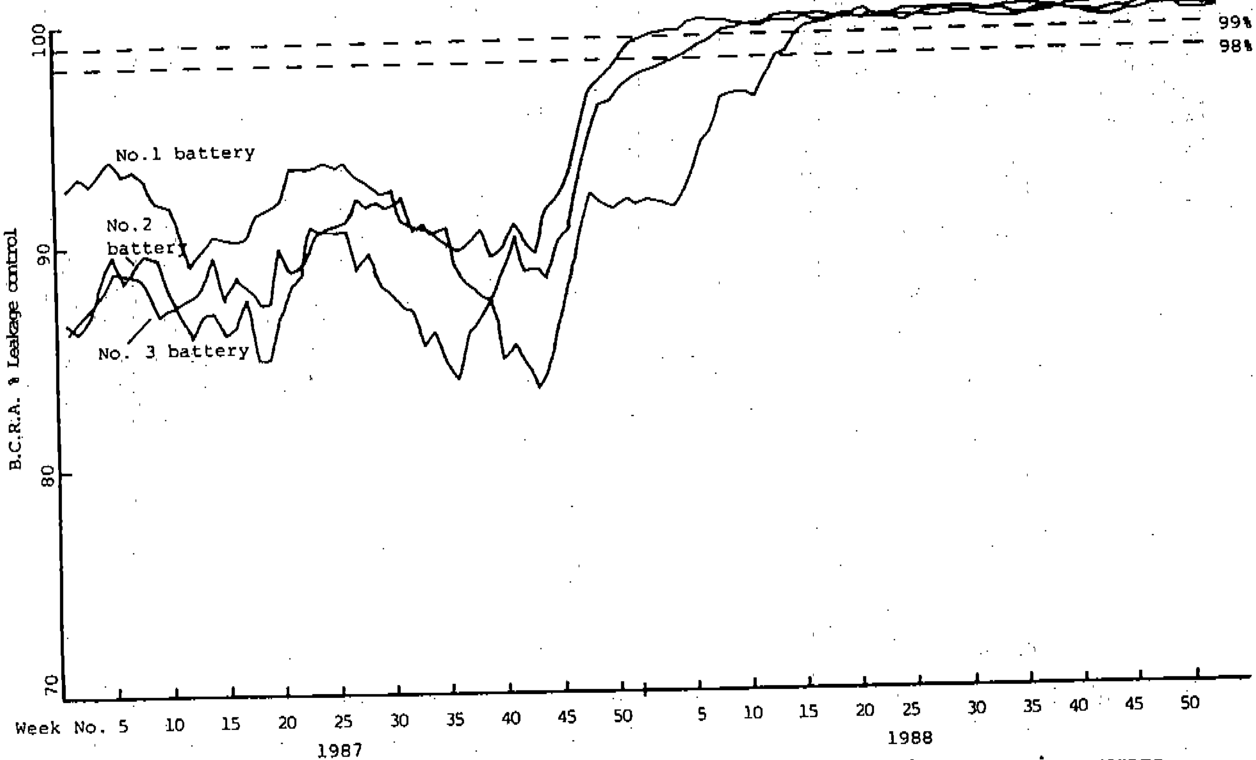


2 Coke oven monitoring at British Steel Scunthorpe Works, Appleby: coke oven doors – running average

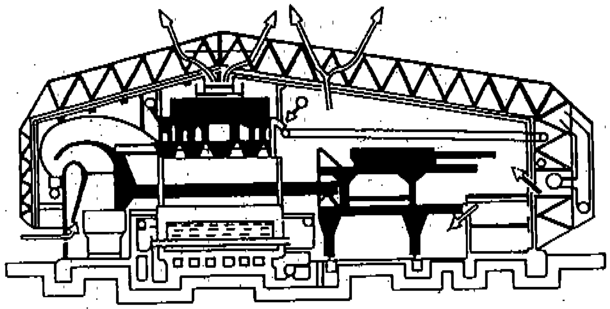
out by BCRA at British Steel's Appleby Frodingham Coke Works, Scunthorpe in connection with their odour emissions study. To improve door leakage control, all aspects of door handling, door cleaning, and door maintenance were examined and an entirely new discipline was implemented. The existing manual method of door cleaning was stopped and all doors were water jetted. In addition, a more rigid programme of door maintenance in which every door was removed for full inspection and overhaul at approximately eight week intervals was initiated. The changes that occurred following the review can be seen in Fig. 2, which shows the results of routine visual door leakage surveys during the 1987–88 period. Before Autumn 1987, all three batteries had door leakage control valves in the range 85–93%, but, when the changes in door maintenance and cleaning started to take effect, the performances were routinely greater than 99%. A remarkable feature of this performance was that two of the batteries concerned had old fashioned Wolff type rigid knock-up seals.

It should be pointed out that measurements made downwind show that the influence of a coking plant on background levels of PAHs and BTX had a range of 1–2 km. When the coking plant is situated in the middle of an integrated iron- and steelworks, this distance is likely to be within the Works' boundary. Dispersion calculations on odoriferous emissions suggest that there is a similar range over which they are likely to be detectable by the human nose.

The problem of diffuse emissions from oven openings (lids and doors) is related directly to the length of the sealing surfaces and the number of openings per day. The philosophy being applied in the latest German coke oven development, Kaiserstuhl III, is to minimise both of these factors by enlarging the chamber volume. In Kaiserstuhl III, the chamber volume will be 80 m^3, which will lead to the production of 48 t of coke per charge. The plant has 120 ovens in 2 batteries and will replace the existing Hansa and Kaiserstuhl plants, which have 469 ovens in 9 batteries. For a coke production of 2 Mt/year, the number of pushing cycles is reduced from 652 to 115 per day and, as a result, the number of openings of lids and doors is reduced from 5868 to 1035 per day. At the same time, the length of sealing surfaces that requires cleaning decreases from 14 to 5·3 km/day. In addition, the ovens will be equipped with leakage extraction systems situated above the coke side and ram side doors. More detailed accounts of the developments in pollution control built into the Kaiserstuhl III coking plant are available in Refs. 15 and 16.

Open source emissions

In an integrated iron- and steelworks, large areas of land are devoted to the stocking and blending of raw materials such as coal, iron ores, and fluxes. Because of their openness, the stockpiles are vulnerable to material losses under strong wind conditions. Losses of this type are a particular problem at the Sollac coking plant at Fos-sur-Mer, owing to the mistral, which blows through that region. Similar problems have been encountered at the Taranto Works of ILVA in Italy and at some of British Steel's Works. Work carried out by Constant *et al.*[17] at Fos-sur-Mer showed that the losses could vary from 170 kg h^{-1} under a wind speed of 30 km h^{-1} to 2800 kg h^{-1} for a mistral blowing at 50 km h^{-1}. Given the average strength and frequency of the mistral, stockpile losses were estimated to amount to 6000 t/year. One approach to controlling stockpile losses is the use of water sprays, as installed at Hoogovens' steelworks in The Netherlands. However, the use of water sprays demands very careful management and their effectiveness is limited when strong winds may persist for several days. A technique that has been applied successfully at Fos-sur-Mer, Taranto, and within British Steel is to stabilise the stockpiles by spraying them with a polymer filming agent, which effectively seals the surface of the stockpile and prevents material lift-off. It is not possible to maintain permanently sealed surfaces on stockpiles because of the need to remove material from them, but, by methodical working of the stockpiles, the



3 Combisystem (cross-section)

area of broken surface can be minimised. The application of filming agents reduces stockpile losses by a factor of between 3 and 8.

FUTURE DEVELOPMENTS IN AIR POLLUTION CONTROL

The techniques currently used for air pollution control in coking plants have, in most instances, been available for several years. Any further developments must depend to a large extent on new concepts in cokemaking technology, as discussed by Nashan.[18] To meet this challenge, the European Development Centre for Cokemaking Technology has been established, with Ruhrkohle AG, Essen as the major shareholder. The basis of the development will be the construction of a new jumbo coking reactor as a modular unit with both bottom arranged and side arranged regenerators, incorporating a cassette type coke pushing and quenching system.

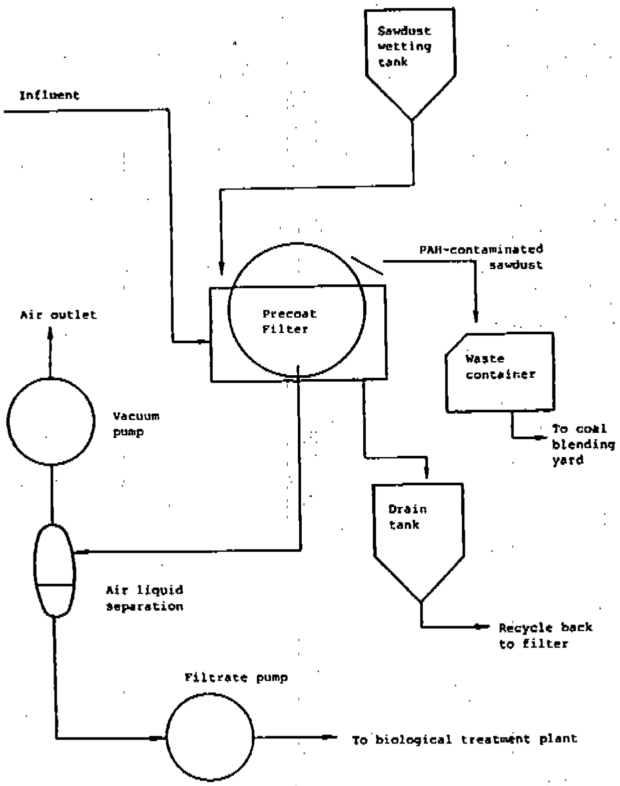
An alternative that may be examined in the future is the possibility of totally enclosing coke oven batteries in a roofed building, as shown in Fig. 3. This so-called 'combisystem' incorporates sectionwise ventilation of the various compartments, namely, coke side, battery top, and ram side. Clearly, such a development would enable external emissions to be controlled more effectively, but great attention would be required to ensure that adequate workplace conditions were maintained.

WATER POLLUTION CONTROL

Traditionally the treatment of coke oven waste water has been a three stage process, involving pretreatment for the separation of oils and tars, steam stripping for the removal of ammonia, and, finally, biological oxidation for the destruction of phenols, thiocyanates, cyanides, thiosulphates, and other compounds that contribute to the chemical oxygen demand (COD) of the effluent. However, as legislation has become tougher in the member states of the EC, the treatment of carbonisation effluents has increased in complexity and may now incorporate additional stages either before or after the biological oxidation process. Moreover, the biological treatment process has been extended, in some instances, to meet stricter discharge limits for nitrogen compounds. An example of the direction in cokeplant waste water standards is given in Table 1, which outlines the limits currently under discussion in Germany.

Pretreatment of cokeplant effluents

The main objective of the pretreatment of cokeplant waste water is to render it amenable to the subsequent treatment stages. In most instances, this consists of the gravity separation of oils and tar by sedimentation and decantation in holding tanks. Two developments in pretreatment of waste water have taken place in Europe. At Hoogovens' steelworks in The Netherlands, a vacuum filtration system



4 Hoogovens' PAH filtration system

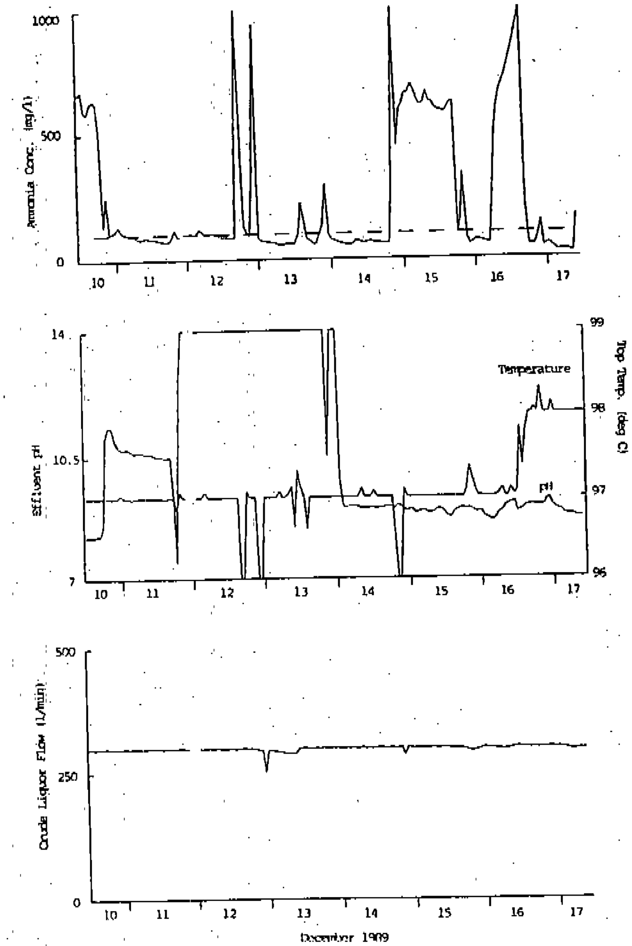
has been developed[19] for the removal of PAHs from waste waters. The system (*see* Fig. 4) is based on precoat vacuum filtration with the use of wood flour (particle dia. ≈ 250 μm) as a filter aid. The wood flour is prewetted and applied to the surface of the filter drum as a filter cake. The drum, which rotates at 1 rev min^{-1}, is immersed in liquor such that 60% of the filter area is submerged and available for filtration. As the precoat material (initially 55–60 mm thick) becomes contaminated, the outer layer is removed gradually at a rate of 1 mm h^{-1} until a residual cake of 15 mm thickness remains. The contaminated sawdust is recycled back to the coal blend for disposal. The system is capable of removing 97–99% of PAHs and typical residual concentrations of PAHs in the filtrate are 20 to 90 ppb.

At the Carling Coke Works in France, a dissolved air flotation process was employed[20] to remove residual tar particles before steam stripping. The system was installed essentially to eliminate fouling of the packed columns of

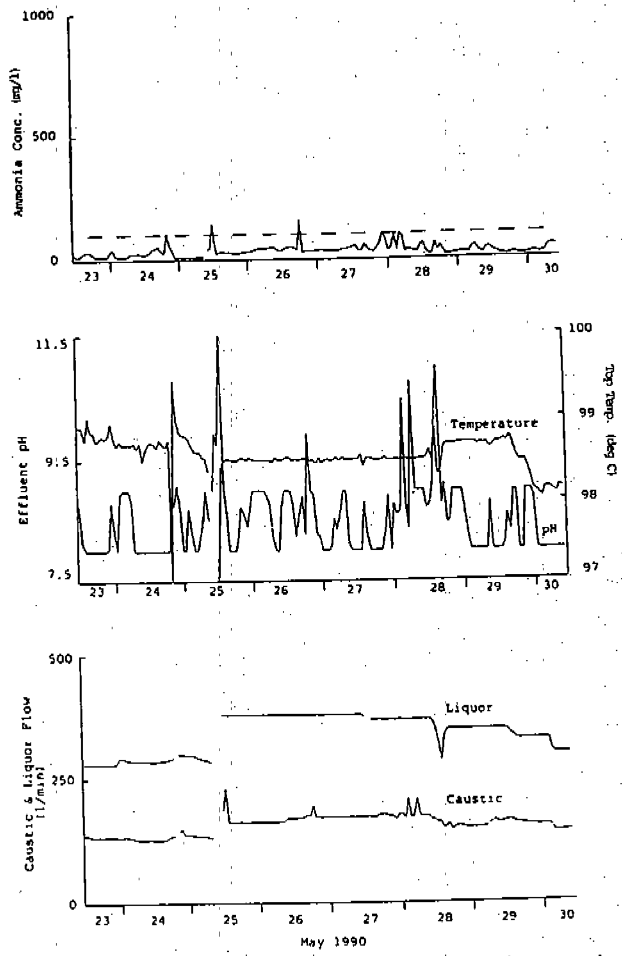
Table 1 Coking plant waste water standards at present under discussion in Germany

	Concentration	
Item	mg l^{-1}	g/t coke
Suspended solids	50	15
COD	200	60
BOD	30	9
Fish virulence	4	
Nitrogen (inorganic):		
NH_4^+, NO_2^-, NO_3^-	80	24
Nitrogen (total)	100	30
Phenol	0·5	0·15
Phosphorus	3	0·9
CN	0·1	0·03
SCN	20	6
Adsorbable halogenated hydrocarbons	1	0·3
BTX	0·1	0·03
BaP	0·1	0·03
Sulphide	0·1	0·03

COD chemical oxygen demand; BOD biological oxygen demand.



5 Typical one week period of operation under manual ammonia still control



6 Typical one week period of operation under automatic ammonia still control

the ammonia still, but brought with it the additional benefit of PAH removal. For naphthalene and acenaphthene, the extraction rates were about 35–37%, but, for higher PAHs, extraction efficiencies were greater than 81·5%. The removal efficiency increased with increasing molecular weight and, for PAHs with more than three aromatic rings, removal rates of 97–99% were achieved.

Ammonia stripping

Throughout the world, steam stripping remains the primary technique for removal of the bulk of ammonia present in cokeplant effluents. However, the process is costly and may account for more than half of the overall treatment costs. Therefore, good process control is an essential prerequisite for the cost effective performance of ammonia stills.

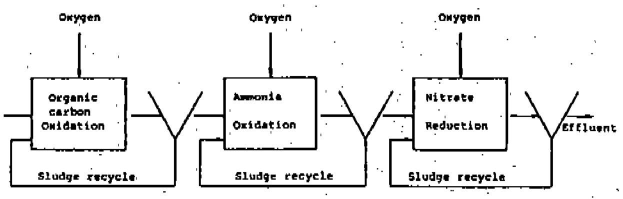
During the 1980s, the ECSC provided financial support to two research projects, carried out by British Steel's Research Laboratories at Teesside and Rotherham, that were concerned with providing more accurate control of the stripping process. In the first project,[21] which was carried out by Teesside Laboratories, instrumentation was developed for continuous monitoring of the residual ammonia content and pH of the stripped liquor. Following its development, the system was applied successfully throughout British Steel for the manual control of its ammonia stills. More recently, the ammonia monitor has been used as the basis of an automatic control scheme[22] (also developed with the aid of ECSC funding).

The control strategy developed in the project acts as an expert system to determine when changes of steam or alkali reagent addition (in this case caustic soda) are required and the magnitude of such changes. Software was developed to enable the plant based process management computer to examine the process data (namely, effluent pH and ammonia content, vapour temperature, and liquor and caustic soda flowrates), to run through the control logic, and to implement any required changes via independent PID (proportional, integral, and derivative action) controllers that were linked to a control valve in the steam supply line and to a variable stroke caustic additions pump.

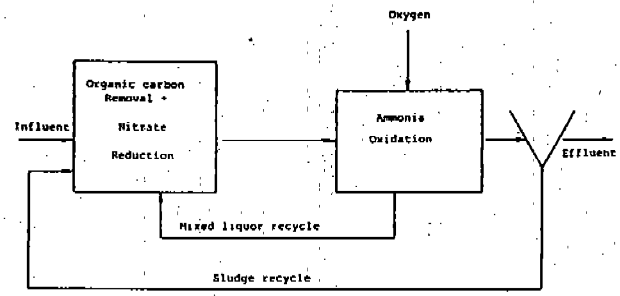
Following the introduction of automatic control, it proved possible to treat the crude liquor to lower and more consistent levels of ammonia and pH than previously. Typical performances under manual and automatic control are shown in Figs. 5 and 6, respectively. Under automatic control, satisfactory treatment was achieved with the use of less caustic soda, but slightly more steam, than previously. The quantity of caustic soda required to treat a given volume of crude liquor was reduced by 15–16%, which is equivalent to annual savings of £25 000 in reagent costs.

Biological treatment

Biological treatment processes have been accepted for a number of years as being the most effective means for the final treatment of cokeplant effluents. Generally, the main purpose of the treatment has been to reduce the effluent COD to acceptable levels by the destruction of phenols, thiocyanates, cyanides, and thiosulphates. However, in many countries in the EC, there is a general trend towards stricter discharge limits for ammonia. The bulk of the ammonia is removed by steam stripping and, although low levels of ammonia ($<20\ \mathrm{mg\,l^{-1}}$) are achievable by this



7 Three sludge process for biological control of nitrogen



8 Single sludge preDN–N process for biological control of nitrogen

means, operating costs increase sharply as the target ammonia level is reduced. One point that is often overlooked is that, in conventional biological effluent treatment plants, nitrogen in the form of thiocyanate, cyanide, and organonitrogen compounds is converted to ammonia in the treatment process, so that the ammonia content of the treated effluent is usually higher than that of the feed liquor. Therefore, very low effluent ammonia levels can be achieved only by extending the biological treatment to include the destruction of ammonia.

The complete biological destruction of ammonia is a two stage process, involving nitrification to yield oxidised forms of nitrogen (predominantly nitrate), followed by denitrification in which the oxidised forms of nitrogen are reduced to gaseous nitrogen. Very different operating conditions are required in these processes: nitrification requires aerobic conditions, whereas denitrification takes place most readily under an anoxic regime. Biological nitrogen control has been the subject of a number of ECSC funded projects. One approach, studied at laboratory scale at the Centro Sviluppo Materiali (CSM), Rome, Italy, involved a three stage treatment with removal of organics, nitrification, and denitrification taking place in separate reactors, each having its own biomass separation and recycling system, as shown in Fig. 7. However, this approach, which requires three separate biomasses and an external source of organic carbon for denitrification, is expensive in terms of both capital and operating costs. Then, in 1980, the results of laboratory studies carried out at the Wastewater Technology Centre, Burlington, ON were presented[23] on the so-called predenitrification–nitrification (preDN–N) process, the flowsheet of which is shown in Fig. 8. This process, which had been applied to certain chemical industry wastes, had not hitherto been attempted on coke oven wastes.

The preDN–N process utilises a single sludge for the destruction of both COD and nitrogen compounds. Influent is fed continuously into the first stage reactor, which is maintained under anoxic conditions, from where mixed liquor is pumped continuously to the second stage aerobic reactor. A portion of the mixed liquor in the aerobic reactor is recycled back to the anoxic reactor, while the overflow from the aerobic basin is passed to the clarifier. Ammonia is nitrified in the aerobic reactor and the resulting oxidised nitrogen in the recycled liquor is reduced to gaseous nitrogen in the anoxic reactor, with the biomass using nitrate as an oxygen source and organic carbon in the feed as an energy source. Surplus organic carbon in the denitrified liquor leaving the anoxic cell is oxidised in the usual way in the aerobic reactor. Operation in this manner has the dual advantages of eliminating the need for an external carbon source and reducing the net oxygen requirement. Moreover, savings in reagents required for pH control are possible, owing to the alkaline balance of the system.

The process was investigated in a joint ECSC project, involving British Steel, Hoogovens, and Dutch State Mines (DSM), which included a full scale demonstration[24] of the technology at British Steel's Scunthorpe Works. The first permanent full scale implementation of the process took place at British Steel's Orgreave Coke Works[25] in 1988 and this was operated successfully until the closure of the Works in August 1990. The long term consistency achievable can be seen in Fig. 9 which shows the outlet ammonia concentration during the first year of operation. The preDN–N system has subsequently been applied successfully at the coke works of NV Sidmar in Belgium and is being considered at other coking plants in Europe. An alternative nitrification–denitrification (N–DN) process has been developed by British Steel in which both processes are carried out in a single reactor by alternating between aerobic and anoxic conditions. This process was developed at British Steel's Orgreave Works and operated in parallel with the preDN–N process configuration. Although generally less stable than the preDN–N process, the single cell N–DN process is capable of producing acceptable ammonia discharge levels, as shown in Fig. 9*b*.

Alternative biological treatment processes

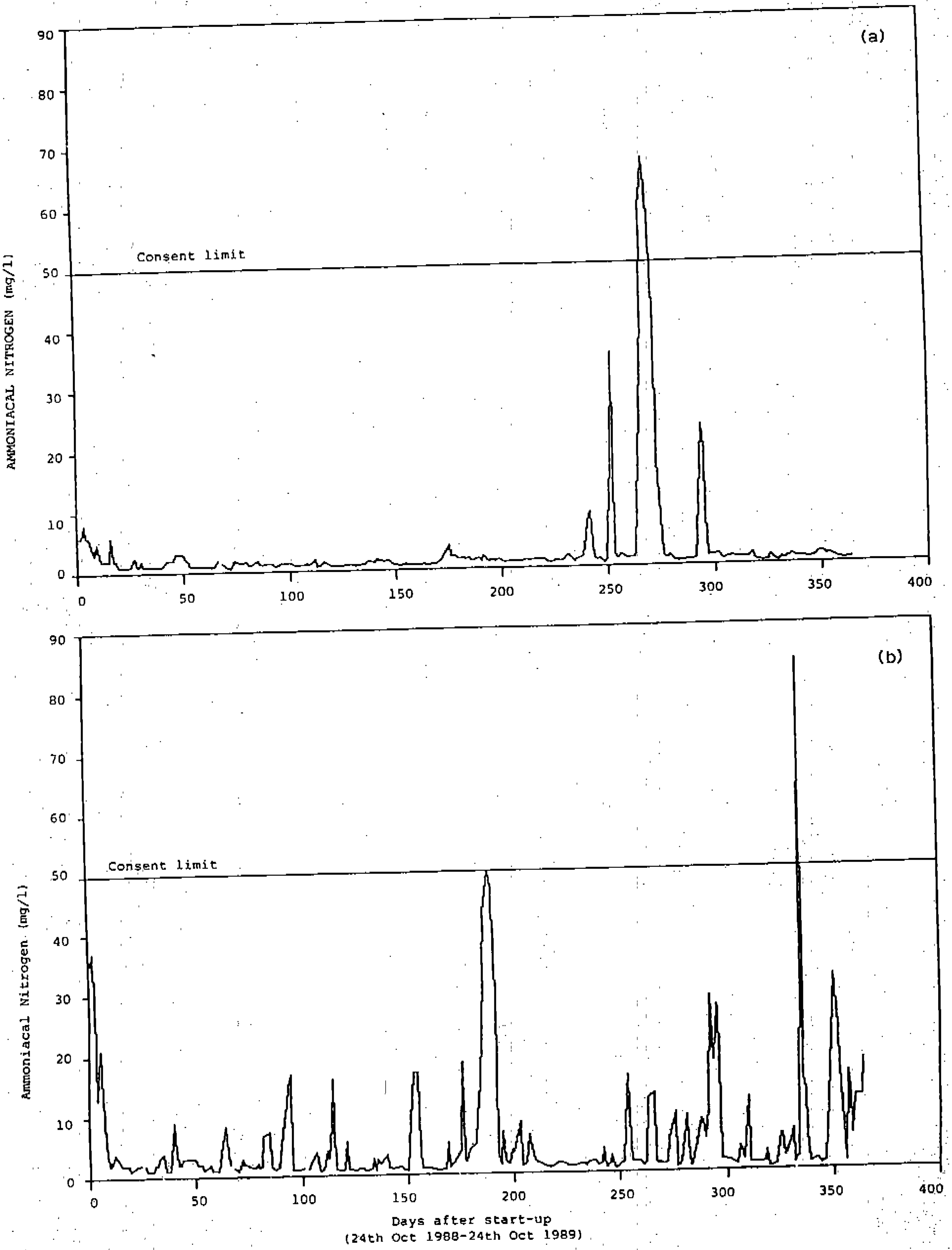
Two developments of interest in biological treatment have taken place during the last five years. The first is the installation of a fluidised bed[26] treatment system at Hoogovens. This type of reactor has the potential for process intensification and is particularly attractive where space is limited.

The second development is the application of reed beds to coke oven effluent treatment. This system, developed in Germany, exploits the natural ability of the *Phragmites* reed to transfer oxygen from the atmosphere to its root zone, where bacteria in the soil effect biological removal of pollutants present in the groundwater. A system of this type is being developed at British Steel's Llanwern Works. In complete contrast to a fluidised bed treatment plant, reed beds require large areas, e.g. the system at Llanwern occupies about 18 hectares, but they have considerable potential for energy savings and are ecologically attractive.

As is often the case with pioneering technological developments, there have been some initial problems with both of these processes. However, these are by no means insurmountable and it is expected that both schemes will achieve their full operating potential in the near future.

Tertiary treatments

Tertiary treatment processes are designed to provide final polishing of effluents before discharge. Perhaps the most common cause of non-compliance with discharge limits is high effluent suspended solids concentrations. It is often the case that high suspended solids concentrations may also produce concomitant effects on the effluent COD or PAH levels. Equally, the consent limits set for the COD or PAHs may actually require the effluent suspended solids concentration to be controlled to a greater extent than is required by the effluent discharge limits. At British Steel General Steels' Scunthorpe Works, a dissolved air flotation unit has been installed after the clarifier of the biological



a preDN–N process; *b* single cell N–DN system

9 Nitrification performance during first year of operation at British Steel's Orgreave Works

effluent treatment plant to control the effluent suspended solids concentrations. The unit is only brought into operation when problems occur and a continuous suspended solids monitoring system has been installed to give early warning of elevated suspended solids concentrations, so that the effluent flow can be diverted to the dissolved air flotation unit. Considerable pilot scale work has been carried out at NV Sidmar with the aim of developing a system for improving effluent suspended solids concentration.

FUTURE DEVELOPMENTS IN WATER POLLUTION CONTROL

Thus far, it has proved possible to meet the ever increasing demands of new legislation for water pollution control by

incorporating additional treatment stages or by modifying existing treatment processes. As treatment schemes grow in complexity, they become more costly and require more attention from operators to ensure that they are working at optimum efficiency. Clearly, the ideal situation would be to avoid completely the need to discharge waste water from the plant. This possibility has been investigated by DMT in an ECSC sponsored pilot scale study[27] by application of reverse osmosis. To optimise the technique, new methods for gas cooling and surplus waste recovery have been developed. The system is still being developed at the Kaiserstuhl Coking Plant, Dortmund in conjunction with Still Otto GmbH. It is confidently expected that the new system for recovery of process water from carbonisation surplus water will be fully developed and operating within the next two years.

CONCLUSIONS

Considerable progress has been made in the carbonisation industry in both air and water pollution control during the past 10–20 years. As further environmental improvements are required by the authorities, it will be necessary to consider alternative production and pollution control technologies.

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