BIOLOGICAL TREATMENT OF COKE PLANT WASTE UTILIZING AN INTEGRAL CLARIFICATION CONCEPT

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INTRODUCTION

Armco's Hamilton plant is located in New Miami, Ohio, on the Great Miami River. The plant produces molten pig iron, metallurgical coke, coke gas, and coking by-products.

The Hamilton Coke Plant consists of four Koppers-Becker underjet design batteries with a total of 110 ovens. The oldest battery was constructed in 1928 and the newest started up in 1947. During this period the batteries have been rebuilt several times.

In 1976, Armco initiated an extensive modification and rehabilitation program for all four coke batteries. A major part of this program was the installation of state-of-the-art air and water pollution control facilities. The water pollution control program included the collection and treatment of sanitary sewage, ammonia still waste, benzol plant waste, quench tower waste, and high temperature noncontact cooling water. This paper specifically deals with the treatment of the sanitary sewage, ammonia still waste, and the benzol plant waste.

BACKGROUND

The wastewaters generated at Armco's Hamilton Coke Plant are primarily from the flushing liquor system and the benzol plant. The flushing liquor is hot water which is sprayed directly into the collecting mains to quench coke oven gas as it leaves the ovens. A circulating liquor system is used to cool the gas in direct spray primary coolers. Water evaporated from the coal is condensed in the main and primary coolers creating excess flushing liquor. This highly contaminated excess liquor from the two systems is collected in storage tanks prior to treatment.

Several sources of wastewater from the benzol plant are collected in a common oil separation sump. The largest source is condensate from wash oil/crude light oil distillation operations. The wash oil is purified by steam stripping to remove crude light oil that was absorbed in the light oil gas scrubbers. The steam condensate is discharged to the benzol sump as a contaminated waste stream. See Table I for the design volumes and chemical composition of the raw excess ammonia liquor and benzol plant wastewater.

At the outset of the program, a study was conducted to determine the best approach for treating coke plant wastewater. Alternatives studied and rejected included physical-chemical treatment with activated carbon and joint treatment in a publicly owned treatment works (POTW). The physical-chemical scheme offered a lower capital investment but a much higher operating cost and was thus rejected. The joint treatment scheme was rejected because of the remote location of the coke plant and the unique configuration of the POTW. Thus, a combination of physical-chemical treatment followed by biological treatment was chosen. The major treatment objective was compliance with NPDES permit requirements. In order to meet this objective, each major waste stream had to be pretreated by physical-chemical methods to remove incompatible pollutants prior to biological treatment.

Table I. Raw Waste Water - Design Composition

	Excess Ammonia Liquor			Benzol Plant Waste		
	(mg/1)	(kg/day)	(#/day)	(mg/1)	(kg/day)	(#/day)
Average Flow GPD		58,300			43,100	
Ammonia-N	4625	1020	(2247)	13	2.3	(5)
Cyanide	25	5.5	(12)	19	3.2	(7)
Thiocyanate	1400	310	(680)	18	2.7	(6)
Oil & Grease	10	1.8	(4)	45	7.3	(16)
Phenol	1140	250	(550)	114	19	(41)
Sulfide	23	5	(11)	11	1.8	(4)
Suspended Solids	60	14	(30)	40	6.4	(14)
COD	8180	1820	(4000)	550	90	(200)
TOC	2350	520	(1140)	540	88	(195)
pH		9.0			7.2	

PHYSICAL-CHEMICAL PRETREATMENT

The benzol plant waste contains large quantities of oil. During the original survey the majority of this oil was free or floatable oil with less than 30 mg/1 of emulsified oil. However, the light oil recovery operation has since been modified, and the waste stream now contains 800 mg/1 of emulsified oil. The free oil is partly removed in the existing oil separation sump. However, this sump is not capable of handling large oil spills. Because of the potential detrimental effect of large quantities of free oil on the biological treatment plant, additional oil removal equipment was installed to help contain spills. This equipment consists of a prepackaged gravity oil/water separator. The separator is installed in series with the existing sump and designed to remove free oil that passes through the primary separation tank. After treatment in this separator, the water is pumped to the biological treatment plant. To date, the emulsified oil has caused no apparent problems at the bioplant.

Excess ammonia liquor contains large quantities of ammonia, sulfide, cyanide, and other compounds which can inhibit biological oxidation [1,2]. The ammonia is present in two forms, commonly referred to as "free" and "fixed" ammonia [3]. Free ammonia, including ammonium hydroxide, ammonium carbonate, ammonium sulfide, ammonium cyanide, etc., is easily dissociated and removed by steam stripping. Fixed ammonia salts, including ammonium chloride, ammonium thiocyanate, ammonium sulfate, etc. [3] are dissociated and removed by raising the pH with an alkaline material and steam stripping. To enable the final effluent to meet the NPDES permit requirements a steam distillation system was installed to remove the free and fixed ammonia, cyanide, and sulfide. This system was chosen over other concepts being used in the industry because of economics and site specific factors.

The alkaline material used to dissociate the fixed ammonia at Hamilton is caustic soda. A solution of 50% sodium hydroxide is injected directly into the still without additional dilution. Caustic soda was chosen over the more traditional material, milk of lime, because of simplified operation and fewer maintenance problems. The addition of caustic requires only the installation of a storage tank and a metering pump, rather than the complex feeding system required for lime, with a resultant lower capital cost. The pH at the top of the fixed still can be controlled, thus eliminating the swings in pH at the bottom of the still due to the long lag time in the still, and the problem of fouling the still with lime has been eliminated. Another advantage, as yet not fully evaluated, is that caustic eliminates suspected problems caused by high concentrations of calcium in the bioplant feed associated with lime stills. The major disadvantage of caustic is that it costs several times as much as an equivalent amount of lime.

An ammonia still originally installed in 1954 as part of the Middletown Coke Plant and retired in 1976 was relocated to the Hamilton Coke Plant. The still contains 5 free ammonia (free leg) trays and 10 fixed ammonia (fixed leg) trays. The trays are all standard cast iron single bubble cap plate sections. The system is equipped to use 50% sodium hydroxide as the alkaline material necessary to dissociate fixed ammonia. A second "standby" ammonia still was installed for use during maintenance and cleaning of the primary still. The second still is a standard pressure vessel column with float valve type, tower filler trays. The ammonia still system includes an automatic pH monitoring and control system which measures and records the pH of the still discharge as well as controlling the amount of caustic fed to the fixed leg.

Excess ammonia liquor is injected into the free leg of the ammonia still near the top of the still column. After passing through the free leg, the liquor is removed and caustic added and mixed with a motionless mixer. The high pH liquor is then injected into the fixed leg. Low pressure exhaust steam is injected at the base of the still, which is bubbled through the descending flow of liquor to strip the ammonia. The ammonia vapors and other acid gases are collected at the top of the still, cooled to condense excess water, and discharged into the coke gas downstream of the primary cooler. The ammonia is later recovered from the gas as ammonium sulfate, a by-product. The still wastewater with little remaining ammonia is pumped to the biological treatment plant.

DESIGN CONSIDERATIONS

The biological system was designed from actual wastewater flows and analysis (Table I), data reported in the literature [1,4-9], and information gathered while inspecting most of the operating coke plant biological treatment systems in North America. The most significant findings of this predesign investigation were: (a) the need for extended equalization, primarily to equalize the wide fluctuation in wastewater chemistry; (b) a minimum aeration detention time of 24 to 48 hours; (c) the need for completely mixed aeration to minimize the concentration of toxic parameters; (d) the possibility of achieving both carbon oxidation and nitrification in a common aeration tank; (e) the need to add phosphorus to support bio growth; (f) the universal problem with aerator foaming; and (g) the need to control pH and temperature. In addition, the investigation uncovered an innovative clarification system that might be used to great advantage in coke plant biotreatment. The system incorporates an integral clarifier with scraper mechanism to direct the settled sludge back into the aeration basin. In late 1976, there were no similar operating systems in the United States, although several were operating in Canada. In December 1976, a trip was taken to observe operating systems in Quebec [10] and Ontario, Canada [11]. Following this trip, it was concluded that the concept offered several advantages, including potential capital cost savings, elimination of a separate sludge return system, potential for improved treatment, and substantial land savings. Therefore, in February 1977, the engineering firm of Burns & McDonnell was instructed to proceed with the design of the biological treatment system utilizing this integral clarifier concept with provisions to achieve nitrification and with the capability to add a second stage nitrification reactor should it be required.

In November 1977, construction commenced with the clearing of the site. The work proceeded very slowly through the winter months, and in March 1978, the first major pour of concrete was made. The work progressed through the summer of 1978 and because of many delays caused by a wet spring and summer, construction carried over into the winter of 1979. Fortunately, the winter of 1979 was relatively mild and the plant was ready to be started in March of 1979.

DESCRIPTION OF FACILITIES

The treatment plant process flow schematic is shown in Figure 1. The incoming waste is received at the plant in one of two surge tanks. The process surge tank, which is 40 feet in diameter by 40 feet tall with a working capacity of 300,000 gallons, receives waste from the ammonia still system and the benzol plant. The tank provides flow and chemical equalization by operating at 50% full and using the side entering mixer shown in Figure 2. Process waste is normally stored in the surge tank for approximately one day before it is pumped to the aeration basin. The sanitary surge tank shown on Figure 3 receives the sanitary sewage generated in the coke plant area, and gas seal water from the flare stack. This tank is agitated with air to insure a non-septic waste. After equalization, the sanitary waste is combined with the process waste and discharged into the aeration basin.

The ammonia liquor from the still is approximately 105 C and must be cooled to approximately 25 C [4] for optimum ammonia removal in the bioplant. This cooling is achieved in part by flash cooling at the still and natural heat loss in the surge tank, with the final cooling achieved in two parallel spiral flow heat exchangers. To compensate for the hot blower air used in the aeration process, the liquor is cooled to around 20 C during the summer months.

In order to maintain the nutritional balance [1] of the aerobic system, phosphoric acid is added to the liquor just after the heat exchangers. At the same point, sulfuric acid is added for pH control should the basin pH exceed the desired 7.8 operating point. Following chemical addition, the total flow is discharged into two parallel aeration/clarification basins.

The treatment systems utilizes a completely mixed, activated sludge extended aeration concept with an integral clarifier. The aeration portion of the plant contains two cubical concrete basins each with its own clarifier section. Each basin has one submerged turbine aerator to achieve complete mixing and oxygen transfer. Three rotary lobe positive displacement air blowers shown on Figure 4 are used to generate the supply of air to the submerged turbine aerator. The aeration-clarification configuration shown on Figure 5 allows the aerator to develop a horizontal velocity along the surface of the aeration basin. This, in turn, causes a downward flow of approximately 10 times the once-through flow in the "chimney" or the space provided between the aeration section and the clarification section. The majority of this flow passes back into the aeration basin, carrying with it the solids settled in the clarifier section. This action provides a theoretical recycle rate of 200%. The flow into the clarifier is equal to the incoming waste. This flow enters at the bottom along the length of the clarifier, passes vertically up through the basin, and is finally discharged from the system. A scraper mechanism is used to move the settled sludge down the sloped clarifier bottom to the chimney area where it is resuspended and carried back to the aeration basin.

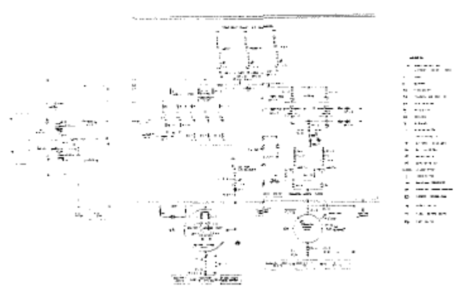

Figure 1. Schematic diagram of treatment plant.

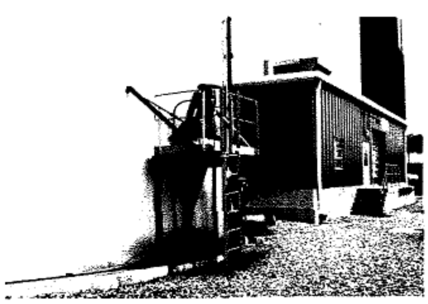

Figure 2. Process surge tank with side entering mixer.

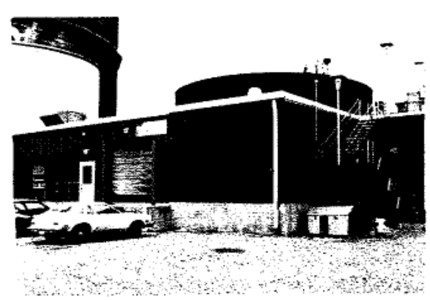

Figure 3. Control building and surge tanks.

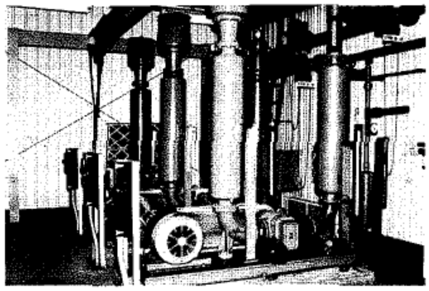

Figure 4. Positive displacement air blowers.

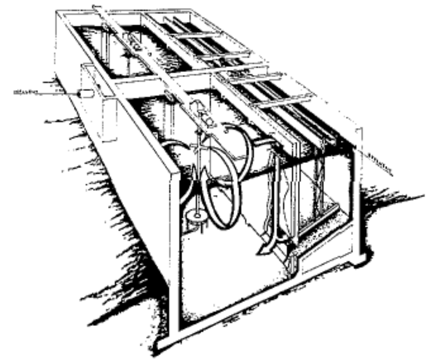

Figure 5. Schematic of aeration/clarification system.

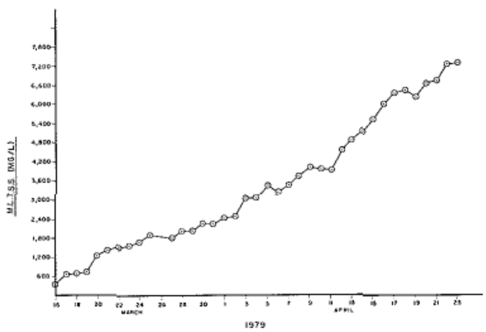

Figure 6. Mixed liquor total suspended solids during start-up.

The treatment plant graphic control panel enables the operator to monitor the physical operations of the plant including flows, temperatures, tank levels, etc., and to make minor corrections as required. The system continuously monitors the aeration dissolved oxygen, temperature and pH and automatically controls the latter two. Based on these controls and chemical analysis made in the treatment plant laboratory, adjustments are made to the system.

SHAKEDOWN AND START-UP

The checking and testing of the system conducted during February and early March of 1979 went smoothly. The only major problems encountered were a bad vibration in the south aerator mixer and an excessive pressure drop in the air supply system. The pressure drop problem was easily corrected by a modification to the sparge ring, but the vibration problem was not fully corrected until late August. Additional construction delays were encountered in the ammonia still area and the benzol yard area so that those areas were not fully operational until July and September, respectively.

On March 14, 1979, the north basins and process surge tank were filled with clean water. Waste ammonia liquor was then discharged to the surge tank which resulted in an extremely dilute solution of feed stock. Approximately 20 gpm of this dilute feed was pumped into the aeration basin which had been seeded with approximately 3000 gallons of activated sludge from Middletown, Ohio's, POTW. During the following days, additional truckloads of sludge were pumped into the system until the mixed liquor was at 1200 mg/1 MLTSS. Because the Middletown POTW treats wastes from Armco's Middletown Coke Plant, the microorganisms were acclimated to coke plant waste. As shown on Figure 6, the total solids in the mixed liquor started to climb on day one and has continued to increase. At the end of thirty days, the mixed liquor had increased to 6000 mg/1 and as shown on Figures 7 and 8, the total solids inventory has continued to climb while the

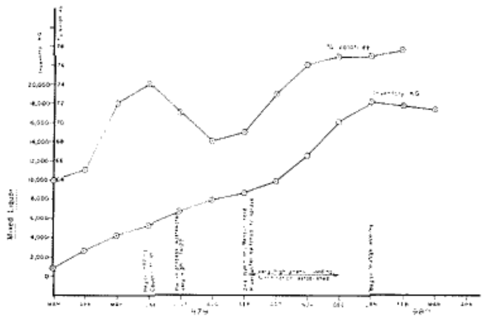

Figure 7. Mixed liquor/total biomass inventory and percent volatiles.

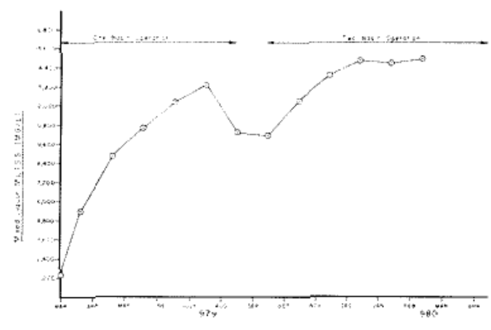

Figure 8. Mixed liquor daily average concentration MLTSS.

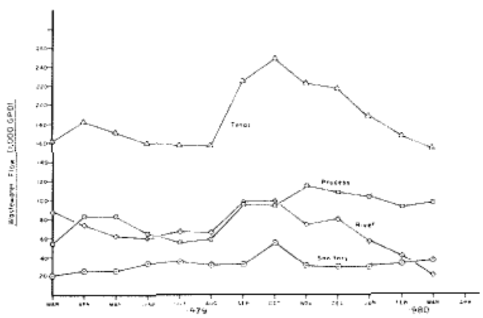

Figure 9. Average monthly wastewater flows.

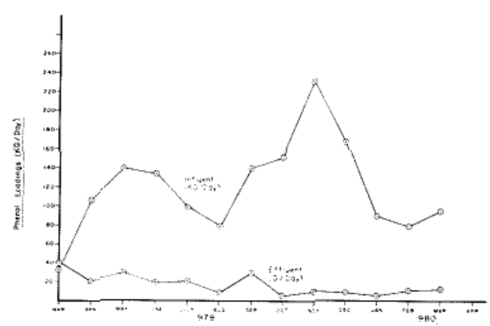

Figure 10. Phenols: influent and effluent average daily loadings.

volatiles have generally been above 70%. The dip in MLTSS concentration shown on Figure 8 is caused by the start-up of the second aeration basin after correction of the vibration problems and installation of a foam spray system. While operating with only one aeration basin, the system consistently removed phenol and thiocyanate with little or no removal of ammonia.

DISCUSSION OF OPERATING DATA

Figure 9 shows the wastewater flows that have been treated in the first year of operation. The sharp increase in September is caused by the introduction of the benzol yard waste for the first time and the use of river water in a foam spray system. Because the system experienced severe foaming while operating on one basin, a river water spray header was installed around the basins to help knock down the foam. After the second aeration basin was started, and as the MLTSS continued to climb, the foam subsided allowing decreased use of river water during early 1980.

The system has functioned well in treating phenol even during upset or shock loading conditions. Figure 10 shows the phenol loadings that have been treated. The influent phenol monthly average has varied from a low of 80 kilograms per day (180#/day) to a high of 230 kg/day (500#/day) with no effect on the effluent quality. The highest monthly average discharge to date has been 30 grams per day (1 ounce per day).

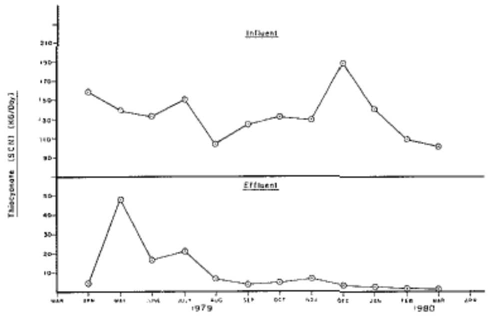

Figure 11. Thiocyanate (SCN): influent and effluent average daily loadings.

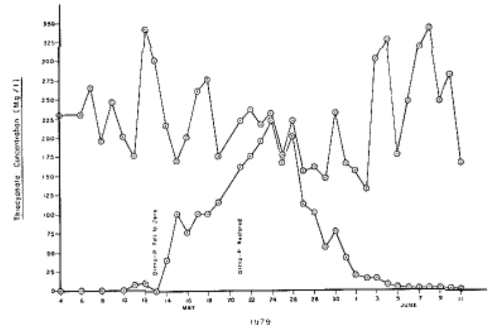

Figure 12. Upset conditions due to lack of phosphorus nutrient as indicated by thiocyanate.

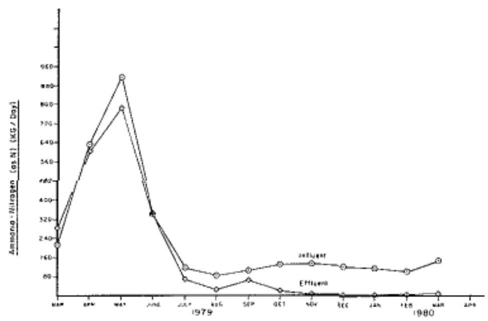

Figure 13. Ammonia nitrogen: influent and effluent total daily loadings.

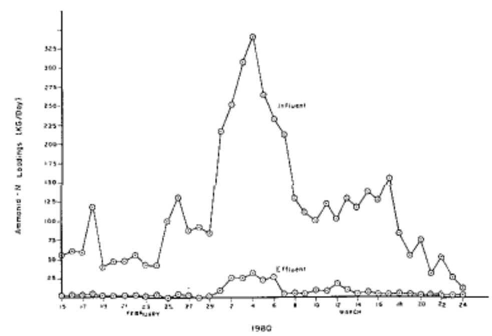

Figure 14. Ammonia upset: influent and effluent total daily loadings.

Thiocyanate removal has not been as spectacular as is shown on Figure 11. Thiocyanate has proven to be the hardest parameter to remove and the most sensitive to varying operating conditions. For this reason, and the fact that the wet chemical test for thiocyanate is easy, it has been used to determine the relative health of the system. Figure 12 shows the daily influent and effluent concentrations during "upset" conditions. In May, phosphate concentrations in the system were inadvertently depleted. The phosphate levels were undectectable for more than a week before corrective measures were taken. Figure 14 shows the removal of thiocyanate has stabilized, possibly due to the start-up of the second basin in September, with the resultant increased biomass inventories, and no upsets have occurred since.

During the first seven months of operation, ammonia removal, as shown on Figure 13, was poor due to the lack of caustic in the ammonia still and operation of only one aeration basin. The ammonia still system was started before the caustic feed system and did not achieve fixed ammonia removal until late May. The bioplant operated on one aeration basin until September 15, when the second basin was put into service. During that four-month period, June-September, ammonia removal was erratic and the system difficult to operate. The plant would achieve nitrification, a drop in the pH would occur, and the next day there would be no evidence of nitrification. On September 15, 1979, the second aeration basin was put into service which stabilized the system. Soon nitrification began to occur consistently and the system has achieved excellent ammonia removal to date. Since mid-October the system has had an average influent loading of 85 kg/day (190#/day) of ammonia nitrogen and 130 kg/day (285#/day) of thiocyanate and has discharged an average for the six months of only 6 kg/day (13#/day) of ammonia-nitrogen (Tables II and III). An indicator of the stability of the system is shown on Figure 14, which shows a shock loading

Table II. Biological Treatment Plant Influent

	Design Composition			Actual Composition[a]		
	(mg/1)	(kg/day)	(lb/day)	(mg/1)	(kg/day)	(lb/day)
Flow:						
Ammonia Still Waste		64,600 GPD			-	
Benzol Yard Sump		43,100 GPD			-	
Misc. Waste		10,900 GPD			-	
Total Process		118,600 GPD			102,500 GPD	
Sanitary		21,800 GPD			35,100 GPD	
Total Contaminated Waste Water		140,400 GPD			137,600 GPD	
Dilution Water		73,800 GPD			61,400 GPD	
Total Plant Feed		214,200 GPD			199,000 GPD	
Ammonia-N	80	70	(150)	115	85	(190)
Cyanide	5	3.6	(8)	3	2.3	(5)
Thiocyanate	400	320	(700)	175	130	(285)
Oil & Grease	12	10	(22)	45	35	(75)
Phenol	250	200	(440)	180	135	(300)
Suspended Solids	40	32	(70)	70	55	(120)
COD	1800	1500	(3300)			
TOC	600	500	(1100)			
pH		7.0-10.0				
Detention time[b] - total feed		- 30 hours			32 hours	
Detention time[b] - process & sanitary		- 45 hours			46 hours	
Clarifier Overflow Rate-Total Feed				300 GPD/SF	275 GPD/SF	
Clarifier Overflow Rate-Process & Sanitary				200 GPD/SF	190 GPD/SF	
Clarifier Weir Loading-Total Feed				4,100 GPD/LF	3,800 GPD/LF	
Clarifier Weir Loading-Process & Sanitary				2,700 GPD/LF	2,650 GPD/LF	

[a]October 1979-March 1980: 6-month average of 30-day averages.
[b]Based on aeration tank volume (including chimney) of 265,000 gallons.

Table III. Biological Treatment Plant Effluent

	Design Composition			Actual Composition[a]		
	(mg/1)	(kg/day)	(lb/day)	(mg/1)	(kg/day)	(lb/day)
Flow:		214,200 GPD			199,000 GPD	
Ammonia-N	180	145	(316)	7	6	(13)
Cyanide	4.5	3.6	(8)	1.5	1	(2.5)
Thiocyanate	-	-	-	4	3	(7)
Oil & Grease	12	10	(22)	5	4	(8)
Phenol	3.0	2.3	(5)	0.001	0.007	(0.015)
Suspended Solids	70	60	130	32	25	(55)
pH		6.0-9.0				

[a]October 1979 - March 1980: 6-month average of 30-day averages

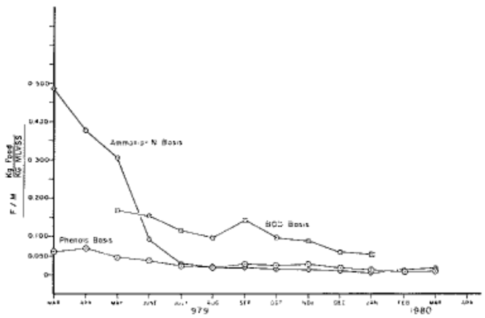

Figure 15. Food/mass ratios.

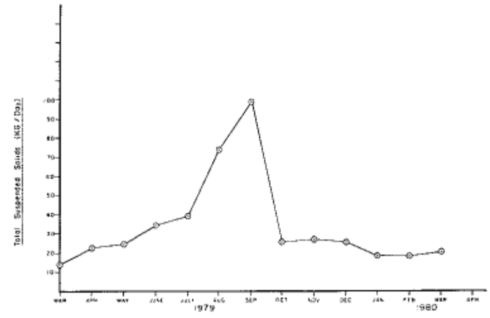

Figure 16. Effluent total suspended solids average daily discharge.

in early March, 1980. Around February 29, the last free tray of the ammonia still became plugged with tar and pitch. After minor modifications and tuning, the standby still was put on stream March 5 and operated until March 18. Although the standby still did not exhibit the removal efficiency of the primary still, it operated well enough to bring ammonia loadings at the bioplant back into range, eliminating the need for backup storage lagoons or other treatment. During the first week of March, the treatment plant ammonia feed was more than tripled to 340 kg/day (750#/day), yet the biosystem achieved over 90% removal of ammonia. The highest quantity discharged during this time was 30 kg/day (66#/day).

Figure 15 shows the food-to-mass ratio expressed in kilograms of BOD_5, phenol, and ammonia fed to the plant over kilograms of mixed liquor volatile suspended solids. As is shown, the F:M ratios are currently very low, with BOD_5 in the range of 0.06 kg/kg MLVSS, phenol at 0.01 kg/kg MLVSS and ammonia in the range of 0.015 kg/kg MLVSS. These low ratios are primarily due to the high inventory of mixed liquor solids.

The suspended solids in the effluent as shown on Figure 16 have generally been quite low. With the exception of one peak period, solids in the effluent have been less than 40 kg/day (90#/day), and have averaged 25 kg/day (55#/day) for the last six months. In August and September, solids were very high in the one operating aeration basin when flows through the basin were high. Subsequently, the sludge blanket in the clarifier section was nearly at the water surface and a carry-over of solids resulted. In January, a sludge wasting program was initiated and approximately 35 kg/day (77#/day) of solids are currently wasted. Mass balance calculations indicate that biological growth has been equivalent to approximately 0.25 kg/kg of phenol removal. The excellent performance in the clarifier is in part credited to the design of the integral clarification concept. No polymers are added and the flow receives no mechanical flocculation in the clarifier. The absence of sludge recirculation pumps has prevented breakup of the floc as it is returned to the aeration basin. The low effluent suspended solids may be attributable in part to the use of sodium hydroxide in the ammonia still rather than milk of lime. This contributes a much lower inorganic solids loading to the system, thus allowing the microorganisms to form a better floc with less "fine" solids carry-through to the effluent.

OPERATING TRAINING AND RESPONSIBILITIES

The successful operation of this plant must be credited to outstanding performance by the operators. The plant is under the direct control of one day foreman with one operator present around the clock and an extra operator on day turns. All of these men underwent an extensive three-week classroom training program, with additional on-hand training before and during the start-up stages. In addition, the operators are obtaining the required experience and reviewing additional training material in preparation for obtaining State certification as licensed operators. The operators are conducting the majority of the chemical analyses with only cyanide and oil & grease analyses contracted to outside labs. By manning the plant around the clock, the various operations can be closely monitored

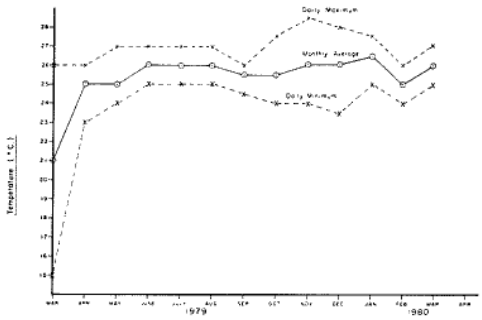

Figure 17. Aeration basin temperature.

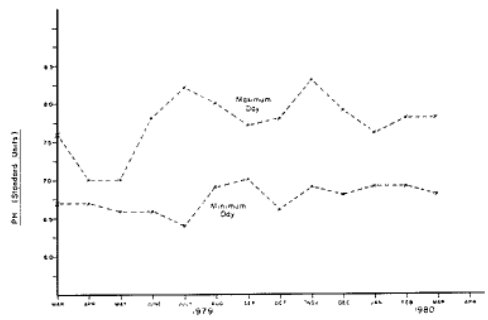

Figure 18. Aeration basin maximum and minimum pH data.

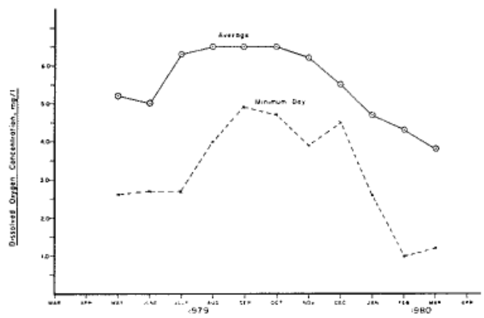

Figure 19. Aeration basin dissolved oxygen data.

and controlled. Figures 17, 18, and 19 show the close control that has been achieved on temperature, pH, and dissolved oxygen. The average temperature has been controlled at 25 C plus or minus 1 C throughout the last year. By close observation, the operators can make corrections in the cooling water system before troubles develop. Even though the pH on Figure 18 varies from 6.6 to 8.3, the operators were able to correct this by adding acid or alkaline materials to bring the system back into specification. The dissolved oxygen as shown on Figure 19 is not usually a controlled parameter, but is used as a monitoring tool. The operator can detect changes in the D.O. which may signal a pending upset and take corrective action.

CAPITAL AND OPERATING COST

The biological treatment system as originally constructed cost $2.15 million, with an additional $1.35 million for collection and ammonia still systems. An additional $1.5 million was spent for miscellaneous sumps, cooling towers, and related projects, bringing the total project cost to $5.0 million. Included in this figure is an estimated $800,000 for modifications and relocations to retrofit the existing coke plant to accomodate the new treatment plant. Based on the total contaminated wastewater design flow of 140,000 GPD, the cost of the biological treatment plant was $15/gallon. The ammonia stills and collection system add $10/gallon, for a total capital cost for the bioplant and associated pretreatment systems of $25/gallon. Direct operating cost for the treatment plant and the ammonia stills for the period July-December 1979 was about $18/1000 gallons of process liquor treated, or approximately $1.30/ton of coke produced. The added cost of capital recovery makes the treatment cost $30/1000 gallons of process liquor or $2.20/ton of coke produced.

CONCLUSIONS

The treatment of coke plant waste liquors to achieve phenol and ammonia removal in a single stage reactor has proven to be a viable treatment method, although expensive when used in series with a caustic soda ammonia still. Control of pH has been the most difficult factor because of the formation of acid in the treatment process and the destruction of the available alkalinity.

Negative effects on nitrification or phenol removal by the introduction of emulsified oil has not been a problem. Emulsified oil in the effluent is averaging less than 5 mg/1 with an average inlet loading rate of over 40 mg/1.

Operating the system with the extremely high mixed liquor and long sludge ages in the aeration basin has not been a problem. During periods of high flow, some carry-over of solids is evident but there is no indication of a problem during normal operations.

Although the operations of this plant have been extremely smooth, a degree of caution must be exercised if this data is to be considered for other treatment plants. At this writing, the plant has operated thirteen months with only six months of satisfactory nitrification. Nitrification has only occurred during the winter months. It is essential for complete demonstration of the plant to obtain a full year of operating data.

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