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Project Summary

The Fate of Organic Pollutants in a Wastewater Land Treatment System Using Lagoon Impoundment and Spray Irrigation

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Michigan's Muskegon County Wastewater Management System (MCWMS) is one of the largest facilities of its kind, treating on the average of 125 thousand cubic meters of wastewater by extended aeration, lagoon impoundment, and spray irrigation. Over 70% of the influent originates from industrial sources including several organic chemical manufacturers. This study was undertaken to determine the fate of the organic compounds within the treatment system. The influent, which is comprised of about 150 organic chemicals at low μg/l to low mg/l concentrations, enters the system and is treated, initially, in the biological cells. Over 90% of the total organic compounds are removed from the water at this stage by volatilization, sedimentation into sludge and biological breakdown. Compounds preferentially settling in the sludge are 3,3'-dichlorobenzidine, 2-chloroaniline, alkyl benzenes and 2,4'-diamino-3,3'-dichlorobiphenyl at average concentrations ranging from 26 to 44 mg/kg. These compounds are resistant to biodegradation. A few biodegradable compounds like phenanthrene and naphthalene are also found in the sludge at less than 1 mg/kg, due possibly to their affinity to the oils and greases commonly found in the sludge. The water treated in this manner enters two storage lagoons (344 hectare each). Impoundment for approximately five months helps to further remove organics by further volatilization, sedimentation, biological breakdown and photodecomposition. The organic compounds surviving after this treatment are in most cases less than 1% of their influent concentrations. The compounds 2,2'-dichloroazobenzene and diethoxychlorobenzene, however, do not readily volatilize, are not biodegradable, and do not accumulate in the sludge, thus appearing to elude treatment during the initial two steps of aeration and lagoon impoundment. Spray irrigation of the lagoon impounded water, however, removes virtually all remaining organic matter. The draintiles which collect the soil percolated water show only sporadic low concentrations (1 μg/l) of chloroform, 1,2-dichloroethane and phthalate esters. In addition, after eight years of irrigation, the soils are almost free of organic compounds except for 2,2'-dichloroazobenzene and phthalates detected in some fields. The analysis of corn samples did not detect any uptake of man-made organics.

The discharges, except for occasional (μg/l) levels of some organics, have been clean. Isolated incidences are apparently due to lagoon seepage, which also contributes to the discharge. Minimzation at the industrial source has kept the levels of contamination in the lagoon seepage under control.

This Project Summary was developed by EPA's Robert S. Kerr Environmental Research Laboratory, Ada, OK, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

When Muskegon County, Michigan, initially proposed land irrigation treatment of its wastewater, the idea was relatively new and untested. There was skepticism as to the effectiveness of the treatment, not to mention the fear of unknown hazards. Now, after many years of operation, it is clear that the system is quite effective. The impact on the environment, to say the least, has been very positive. The lakes and the rivers are many orders of magnitude cleaner than in the past. All wastewater used to be discharged directly or indirectly to river and lake systems in the county with little or no treatment. Now, after collection, the wastewater is treated by aeration in the biological cells and impoundment before being used as water for agricultural irrigation. The soil acts as an excellent adsorbing medium, while the nutrient value is reclaimed by the crops in one of the largest farming operations in the country. In addition to high quality wastewater treatment, the crops provide a substantial source of revenue, thereby reducing the ultimate cost to the users. (The user fee at the time of the report was 25.4 cents per 1000 gallons).

The United States Environmental Protection Agency (EPA), the Michigan Department of Natural Resources, the County of Muskegon and academic institutions (among them, University of Michigan and Michigan State University) have examined the fate of the conventional parameters such as nutrients and trace metals in this land treatment and without exception found it very effective. The socioeconomic impact was also found to be very favorable. Within the last decade, considerable attention has been drawn to the toxic organic waste and its effect on the environment. This has created renewed interest in the Muskegon system. The Robert S. Kerr Environmental Research Laboratory, in 1977, conducted a preliminary survey of toxic pollutants in the system and identified 56 chemicals, including priority pollutants in the influent to the treatment works. This was not surprising, since over 70% of the influent is comprised of wastewater from pulp and paper, chemical, and general manufacturing operations in the county. This information prompted the present investigation of the fate of these and other organic chemicals in the Muskegon system. The principal objectives of this study are:

1) The qualitative and quantitative identification of the organic chemicals in the influent to the system.
2) Elucidation of the treatment performance at all the intermediate treatment points in the system, including the seasonal variations.
3) Determine the ultimate fate of the organic chemicals in the system and define any potential environmental impact.
4) Suggest improvements in the design and operation of the land treatment and identify the need to minimize certain organic compounds by pretreatment at the source.

This study, as expected, has been instrumental in the local industrial regulatory programs and in the management of the treatment system. This report undoubtedly could be an excellent guide to other land treatment facilities.

Facility and Operations

The Muskegon County Wastewater Management System employs extended aeration, lagoon impoundment and spray irrigation to treat an average of 125 thousand cubic meters (TCM) of municipal and industrial wastewater. The system operates as a regional treatment facility, serving 13 municipalities and over 23 major industries including nine chemical-related industries. Over 70% of the influent can be attributed to industrial discharge. The system is located on 11,000 acres (4,455 hectares) of sandy, unproductive land in the northeast corner of Muskegon County. The wastewater is collected by six pumping stations and delivered to a central pumping station with a maximum capacity of 212 cubic meters per minute. The combined wastewater is pumped through 17.2 kilometers of reinforced concrete pipe to the wastewater system.

The design features of the system are illustrated in Figure 1. Initial treatment of the wastewater is provided by two of the three extended aeration biological cells. Each cell has a surface area of eight acres (3.24 hectares) and a holding capacity of 159 TCM. The cells contain 12 floating aerators and 6 stationary platform mixers. The holding time in the two cells is roughly 2.5 days. After aeration, the treated wastewater passes down a concrete spillway to the two storage lagoons, 344 hectares each, with a combined storage capacity of 19.3 million cubic meters. The lagoons are encircled by dikes 4.6 meters high and 61 meters wide at the base. A border strip 122 meters wide from the top of the dike is lined with 20 cm of compacted clay, with remaining bottom area left as native soil. The treated water from the aeration cell is stored in these lagoons for approximately 5 months. The water seeping through lagoon walls is intercepted by a ditch system surrounding the lagoons. This water, if necessary, could be pumped back into the storage lagoons or, if the National Pollutants Discharge Elimination System (NPDES) water quality criteria are met, it may be discharged to the receiving streams. During irrigation months, the water from the lagoon with the highest water quality is discharged into an outlet lagoon with about 6 hectares of surface area and 4 meters in depth. Approximately 2,300 hectare of land is irrigated using this water by 54 center-pivot irrigation rigs at an application rate of 6-10 cm per week. The rigs move in a circular path making one revolution in one to seven days. Most of the land is Rubicon, Roscommon, and AuGres sand and the remaining is Granby, a loamy sand, and Nester-Tonkey clays. The irrigation fields are drained by an elaborate network of perforated nylon-socked tiles and a series of collection ditches. The drainage network consists of 114 kilometers of draintile, 31 kilometers of drain pipe and 16 kilometers of ditches. This system acts to protect the groundwater table, direct water flow and to recollect the renovated water, thus preventing water logging of the land. Over 300 monitoring wells have been installed to ensure that the groundwater quality is being maintained. Corn grown on the irrigation fields serves as a source of revenue and also removes the nutrients. In spite of the unproductive nature of the soil, with the help of irrigation over 43 hectoliters per hectare corn is produced annually.

The collected water from the draintiles is discharged to the north via an outfall (SW05) to Mosquito Creek. Water from the fields on the southern part of the project is discharged to Black Creek (SW34). The project has operated since May 1973.

Experimental Features and Results

The study period lasted from December 1979 through November 1981. The first year of the study was designed to provide intensive data concerning treatment performance to identify any potential problem areas and to verify the quality of the data through a rigorous QC/QA program. During the second year along with treatment performance, attention was focused on soil accumulation and lagoon seepage. Water samples were collected from various treatment points in the system monthly during December 1979 through April 1980, then biweekly

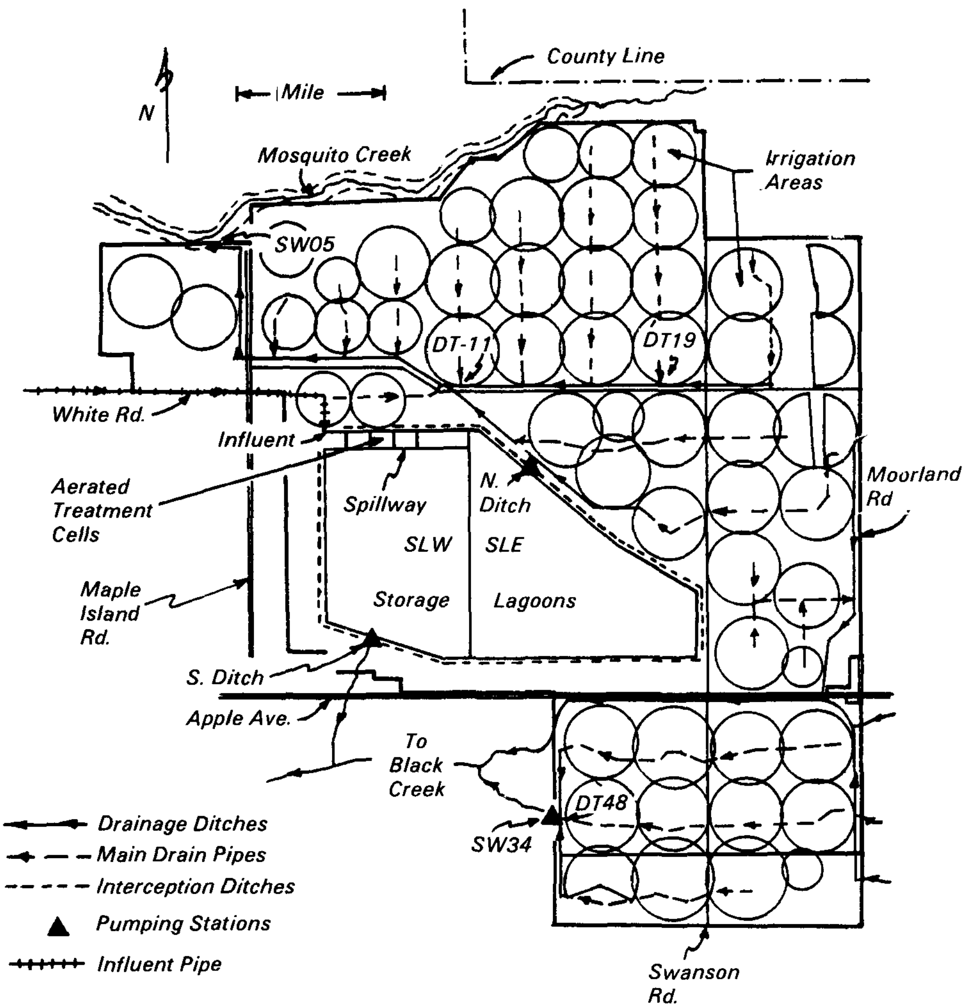

Figure 1. *Sampling locations.*

during May 1980 through November 1980 and then monthly the rest of the time. The water samples were either 2 week composites or 4 point composites. From the aeration cell and lagoon sludge, grab samples were acquired quarterly. The soil and corn samples were composites collected annually. All of the sample collection methods were standard methods designed to protect the integrity of the samples. The analytical procedures applied were standard EPA methods backed by extensive daily quality control. Most analyses were performed by gas chromatographic/mass spectrometric methods. Analysis of 1,2-dichloroethane for the lagoon seepage study was performed using gas chromatography with Hall detector. Analysis of 1-chloroaniline, 3,3'-dichlorobenzidine and 2,4'-diamino-3,3'-dichlorobiphenyl in the lagoon seepage wells was done by HPLC with an electrical conductivity detector. Complete details of the experimental conditions can be found in the full report.

The results of the treatment performance are summarized in Table 1. Out of 150 or so organic compounds detected in the influent, 28 priority pollutants and 33 additional organic compounds were over 1 $\mu g/l$ on the average. Benzene, chloroform, 1,2-dichloroethane, tetrachloroethylene, 2-chloroaniline, 2,4'-diamino-3,3'-dichlorobiphenyl, dimethyldisulfide, 2-propanol and thiobismethane were present in the influent in the range of 100 $\mu g/l$ to 1000 $\mu g/l$. Only toluene and acetone exceeded this range. Most of these pollutants are traced back to various industrial sources. Analysis of the influent serves as a check along with industrial monitoring which is performed independent of this study. Many volatile priority pollutants like benzene, chloroform, 1,2-dichloroethane, tetrachloroethylene, toluene, and 1,1,1-trichloroethane originate from chemical manufacturing, paint manufacturing or from general manufacturing. The sulfides are predominantly from the pulp and paper industry. Other compounds can be traced to chemical industries. The influent composition may vary at any given time depending upon factors such as batch dumping, sanitary uses, etc.

The spillway concentrations listed in Table 1 reflect the extent of treatment in the biological cells over the period of about 2.5 days. The areation in these cells is effective in volatilizing many organics. Other processes operative at this stage of the treatment are breakdown by bacterial metabolism and sedimentation into the sludge. As can be seen in Table 1, many compounds are removed from the water to nondetectable or near nondetectable levels. Priority pollutants surviving over 10 $\mu g/l$ average levels are benzene (11 $\mu g/l$), chloroform (86 $\mu g/l$), 1,2-dichloroethane (164 $\mu g/l$), methylene chloride (31 $\mu g/l$), tetrachloroethylene (31 $\mu g/l$) and toluene (34 $\mu g/l$). Most of these are removed by over 90%. Methylene chloride and 1,2-dichloroethane, however, are removed by 28% and 78% respectively. Among the additional organic compounds 1,258 $\mu g/l$ of acetone, 92 $\mu g/l$ of 2-chloroaniline, 52 $\mu g/l$ of 2,4'-diamino-3,3'-dichlorobiphenyl, 19 $\mu g/l$ of alkyl substituted benzenes, 18 $\mu g/l$ of 2,2'-dichloroazobenzene, 50 $\mu g/l$ of dimethyl disulfide, 589 $\mu g/l$ of 2-propanol and 30 $\mu g/l$ of thiobismethane survive this phase of treatment on the average of over 10 $\mu g/l$. Acetone, 2,4'-diamino-3,3'-dichlorobiphenyl, 2,2-dichloroazobenzene and 2-propanol are among the compounds with average removal of less than 60% of their influent concentrations. The compounds that elude the aeration cell treatment are either water soluble, relatively nonvolatile or noncompatible to bacterial consumption. Table 2 provides a list of compounds found in the aeration cell II sludge. Appearance of compounds such as 2-chloroaniline, 3,3'-dichlorobenzidine and 2,4'-diamino-3,3'-dichlorobiphenyl is probably due to their resistance to biodegradation. Alkyl substituted benzenes probably originate from linear alkyl benzene sulfonate detergents. These compounds, in addition to their low biodegradability, have lower water solubility. Naphthalene, phenanthrene and related compounds are biodegradable. Presence of these compounds in the sludge may be attributed to their affinity to oils and greases common to the sludge.

Table 1. *Treatment Performance During 1980-81*

Additional Organic Compounds	Average Influent Concentration (µg/l)	Average Spillway Concentration (µg/l)	Average Outlet Concentration (µg/l)	Average Outfall SW05 Concentration (µg/l)	Average Outfall SW34 Concentration (µg/l)
Acetanilide	5	< 1			
Acetovanillone	16	1			
Acetone	2,664	1,258			
Alkyl substituted benzenes	53	19	1.0	0.1*	<0.1*
Aniline	24	1			
Atrazine**				0 2	0.3
2-Chloroaniline	567	92	1.7	1.2	<0.1*
Cresol	10	1			
2,4'-Diamino-3,3'-dichlorobiphenyl	118	52			
2,2'-Dichloroazobenzene	38	18	4.7		
Diethoxychlorobenzene	21	10	0.3		
Diisopropoxychlorobenzene	4	1	0.1		
Dimethoxybenzene	1	< 1			
3,4-Dimethoxyphenol	6	2			
Dimethylbenzaldehyde	2	< 1			
Dimethyldisulfide	180	50			
Dimethylnaphthalenes	14	4			
2-Ethoxypropane	3				
Ethylaniline	2	1			
Isopropylidene dioxyphenol	8	4	1.0		
2-Methoxyphenol	40	3			
Methyl aniline	37	13			
Methyl naphthalenes	8	2			
Naphthol	1				
N,N-Dimethylaniline	5	1	<0.1*		
2-Propanol	842	589			
Simazine**				0.2	0.2
Benzene	153	11			
Bis(2-ethylhexyl)phthalate	23	9	2.6	2 0	2.0
Bromodichloromethane	2				
Butyl benzyl phthalate	1				<0 1*
Chlorobenzene	23	< 1			
2-Chlorophenol	2	< 1			
Chloroform	747	86	2.6	0.4	
4-Chloro-3-methylphenol	3				
1,2-Dichlorobenzene	4	< 1			
1,4-Dichlorobenzene	1				
3,3'-Dichlorobenzidine	23	6			
1,1-Dichloroethane	3	< 1			
1,2-Dichloroethane	730	164	1.8	0 6	
2,4-Dichlorophenol	2	< 1			
2,4-Dimethylphenol	1				
Dimethyl phthalate	4		<0.1*		
Ethyl benzene	14	< 1			
Fluoranthene	18	< 1			
Methylene chloride	43	31		0.6*	
Naphthalene	4	< 1			
Phenol	5		0.1*		
Tetrachloroethylene	361	31	0.8	0.4	
Toluene	1,964	34	0.1		
Trans-1,2-dichloroethylene	6	< 1			
1,1,1-Trichloroethane	93	9	0.5		
Trichloroethylene	36	2	0.1*		
2,4,6-Trichlorophenol	2	1	<0.1		
Vinyl chloride	1				
Substituted benzoic acids	1				
Substituted benzaldehyde	2	1			
Thiobismethane	171	30	0.1		
Tributyl phosphate	30	12	0.8	<0.1*	
Trimethylnaphthalenes	11	5			
Vanillin	6	< 1	<0.1*		

* Single occurrence.
**Only those compounds above 1 µg/l in the influent are listed, except for Atrazine and Simazine.

Table 2. Average Concentration of Organic Chemicals in Cell II Sludge (µg/Kg) for 1980 and 1981

Priority Pollutants	*Average*
Butyl benzyl phthalate	*280*
Benzo(a)anthracene	*60*
Bis(ethylhexyl)phthalate	*4,200*
1,4-Dichlorobenzene	*200*
1,2-Dichlorobenzene	*320*
Di-n-butyl phthalate	*340*
3,3'-Dichlorobenzidine	*44,000*
Fluoranthene	*230*
Fluorene	*140*
Naphthalene	*580*
Phenanthrene	*810*
Phenol	*240*
Pyrene	*200*
Toluene	*730*
Additional Organic Compounds	
2-Chloroaniline	*26,600*
Cresol	*1,100*
2,2'-Dichloroazobenzene	*1,400*
Diethoxychlorobenzene	*4,000*
Dimethyl naphthalenes	*9,400*
2,4'-Diamino-3,3'-dichlorobiphenyl	*35,000*
Methyl naphthalenes	*2,600*
Substituted alkyl benzenes	*40,000*
Trimethyl naphthalenes	*7,300*
Tributyl phosphate	*800*

The wastewater treated in the aeration cell enters the storage lagoons via the spillway. Long-term impoundment in the lagoons is very effective in further treating the organic compounds through waste stabilization action. Processes similar to the aeration cell, i.e. volatilization, sedimentation and biodegradation are also active here. In addition, due to the large surface area photodecomposition is greatly facilitated. The tremendous volume of lagoon system also provides a large buffer capacity to adsorb any short-term shock load that may pass through the extended aeration treatment. About 20 organic compounds are detected in the storage lagoon Their concentrations are dependent upon two main factors, which are the loadings and the season During winter months due to slower rate evaporation due to colder weather and ice cover, the volatiles tend to remain at higher concentrations. However, only chloroform and 1,2-dichloroethane were near 50 µg/l in 1980 and acetone and 2-chloroaniline at that level in 1981. Other compounds are near their detection limit.

The system is designed to prevent lagoon seepage from migrating off the project by two interception ditches surrounding the north, east, and south boundaries and purge wells along the west boundary. Monitoring of the lagoon seepage wells on the western side has discovered 5 µg/l of 1,2-dichloroethane and migration is suspected. Continued operation of the purge wells should restrict the migration; however, future plans include the construction of an interception ditch in this area. Analysis of the wells situated between the two storage lagoons indicates that vertical contamination does not proceed to the 68 feet level. The north interception ditch is mixed with groundwater and drain tile effluent to form the north discharge (SW05) to Mosquito Creek South interception ditch (or discharge to HH&G Drain) represents a direct discharge of groundwater and lagoon seepage to Black Creek. A consistent 4 to 6 µg/l of 1,2-dichloroethane was found in the south ditch along with concentrations of 2-chloroaniline that varied from 86 µg/l to 1 µg/l in 1980. In the north ditch during 1981, the levels of 2-chloroaniline dropped from 28 µg/l to <1 µg/l by May. Monitoring of groundwater wells beyond the interception ditch indicates that the ditch is acting as an effective barrier since no contamination has been detected.

Water is drawn from the storage lagoons into the outlet lagoon before irrigation. Table 1 shows the average concentrations of organic compounds in the outlet lagoon. Compounds appearing above 1 µg/l are bis(2-ethylhexyl)phthalate 2.6 µg/l, chloroform 2.6 µg/l, 1,2-dichloroethane 1.8 µg/l, alkyl substituted benzenes 1.0 µg/l, 2-chloroaniline 1.7 µg/l, 2,2'-dichloroazobenzene 4.7 µg/l and isopropylidinedioxyphenol 1.0 µg/l. Most compounds are treated better than 99%. Of the priority pollutants, chloroform and 1,2-dichloroethane are the only two compounds of concern since phthalates with their widespread use are analytically questionable. The county has placed limits on the discharge of chloroform and 1,2-dichloroethane among other compounds. As mentioned earlier, the irrigation water drains through the soil and is then collected by drain tiles. Many organic compounds are adsorbed by the soil where they are subject to bacterial action or evaporation Very few compounds actually remain in soil. Soil analyses at various points in the system indicated that besides questionable phthalates, 2,2'-dichloroazobenzene, xylene, benzyl alcohol, phenyl ethanone, N-phenylaniline, ethenyl benzene, arochlor 1016, phenol, benzaldehyde, trichloroethylene, toluene, 1,1,1-trichloroethane, phenylacetic acid, simazine, atrazine, vanillin, and in an isolated case 1,2-dichloroethane were detected at levels 1 µg/kg-600 µg/kg, not more than six of these being present in a single sample. Atrazine and simazine are introduced in the soil by means of the herbicides used in the farming and are detected in the soil in the growing season. The drain tile water analyses showed only sporadic presence of low 1-4 µg/l levels of chloroform, 1,2-dichloroethaneazobenzene, atrazine, simazine, etc., none of which were persistent.

The outfall discharge data is also listed in Table 1. In the north outfall (SW05) only compounds above 1 µg/l average concentration are bis(2-ethylhexyl) phthalate 2 µg/l and 2-chloroaniline 1.0 µg/l. In the south outfall (SW34) only bis(2-ethylhexyl)phthalate is above 1 µg/l level (2.0 µg/l). The presence of phthalate, as discussed earlier, may be the plastic lining in the ditches and drain tiles.

The results of the overall study show that Muskegon County Wastewater Management System is very effective in treating a variety of chemicals and industrial wastes by utilizing extended aeration, lagoon impoundment, and spray irrigation. From almost 150 organic compounds identified in the µg/l to mg/l range, over 90% are removed in the first stage of the treatment, the extended aeration in the biological cells. Lagoon impoundment further removes the organic compounds and prior to irrigation, over 99% of the organic concentration is removed by either sedimentation, volatilization, biodegradation, or photodecomposition. The water percolated through the soil profile and collected as drain tile effluent is virtually free of organic compounds. The outfalls for the most part do not contain organic compound. Except for small quantities of certain organic compounds previously mentioned, the soil has not accumulated organic compounds. Two chemicals, 2-chloroaniline and 1,2-dichloroethane have been detected in some lagoon seepage wells. The levels have significantly declined during the study period and in recent samples average 2 $\mu g/l$. None of the priority pollutants were detected in the corn sample.

The feasibility of sludge application is also investigated as part of this study. The results of this study are being reported separately.

Recommendations

The County has identified two areas from the study that require management changes to maintain the integrity of the treatment works and decrease the possibility of environmental discharge of chemicals. These areas are lagoon seepage and sludge concentration. The lagoon seepage question is two-fold; first, as it relates to the interception ditches, and second, the northwest corner of the storage lagoons.

In February 1981, the County issued management guidelines, limiting the discharge of organic chemicals to the system by industrial users. Utilizing the extensive data base of industrial loadings and treatment performance data collected by the County laboratory during the study period, limits were established for chemicals that persist by concentrating in the sludge or that survive treatment and thus have a potential for discharge to the environment. The limits became effective January 31, 1982 and should insure future operation without lagoon seepage or sludge disposal questions being raised. In addition, if monitoring indicates any significant trends which suggest that present limits need to be revised or new compounds need to be controlled, the necessary action will be taken by the County through the powers outlined in the Amendments to Exhibit D (County Wastewater Ordinance).

Lagoon seepage in the northwest corner of the storage lagoon system will be examined in the 201 Facilities Expansion Plan hydrogeological study. When the groundwater dynamics of this area are defined, management strategies will be developed to control or contain any migration from the storage lagoons. In examination of the sludge accumulation question, results of lysimeter sludge application studies indicate that the organic chemicals in the sludge will not leach through the soil column and that they will undergo some biodegradation. A 25-acre field pilot study is being proposed for spring of 1982. If results are favorable, the sludge may be land applied on a larger scale. In order to prevent any future questions from arising concerning sludge management, chemicals accumulating in the sludge will be limited as part of the industrial effluent guidelines. These limits are again subject to the results of future monitoring programs by the County. The studies that were undertaken at Muskegon will help the understanding of the behavior of many of the man-made organics at treatment facilities employing extended aeration, lagoon impoundment and spray irrigation. Other than volatilization, the two major removal mechanisms include detoxification by bacteria and photo-oxidation by solar radiation. The mechanism of bacterial action should be investigated further with a goal of isolating the working bacteria.

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The complete report, entitled "The Fate of Organic Pollutants in a Wastewater Land Treatment System Using Lagoon Impoundment and Spray Irrigation," (Order No. PB 83-259 853; Cost: $25.00, subject to change) will be available only from:

National Technical Information Service
5285 Port Royal Road
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