



Project Summary

Chlorine Dioxide for Wastewater Disinfection: A Feasibility Evaluation

Paul V. Roberts, E. Marco Aleta, James D. Berg, and Bruce M. Chow

Chlorine dioxide was compared with chlorine for disinfecting wastewater in laboratory experiments. Chlorine dioxide disinfection was also demonstrated at a full-scale wastewater treatment plant. The criteria compared included coliform kill, inactivation of poliovirus and other indicators, and formation of halogenated organic byproducts.

Laboratory experiments were conducted using mass doses of disinfectant and contact time as independent variables. The fractional survival of coliform bacteria was correlated with the project of disinfectant residual \times contact time.

In general, chlorine dioxide accomplished a given fractional kill of total coliforms with a smaller product (residual \times time) than did chlorine. For a given contact time, the residual required to achieve a given fractional kill of coliforms was 2 to 70 times smaller for chlorine dioxide than for chlorine. Considering both required residual and disinfectant demand, the required doses of the disinfectants were estimated to satisfy three assumed coliform disinfection levels with two types of effluents: conventional activated sludge and filtered, nitrified activated sludge. The required mass doses of the disinfectants were approximately equal for treating conventional activated sludge effluent. The required dose of chlorine was approximately 2 to 10 times greater

than that of chlorine dioxide for treating filtered, nitrified effluent, depending on the coliform standard. The results of studies conducted at a full-scale plant generally agreed within a factor of two with the predictions from laboratory studies, when compared on the basis of the product (residual \times time) required to accomplish a given fractional kill.

For the cases likely to be most typical in practice, chlorine dioxide is approximately two to five times as expensive as chlorine for disinfection. On the other hand, chlorine dioxide forms much lower quantities of halogenated byproducts and is more effective in inactivating viruses than is chlorine.

This Project Summary was developed by EPA's Municipal Environmental Research Laboratory, Cincinnati, OH, 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 the back).

Introduction

The purpose of this report was to evaluate ClO_2 as an alternative to conventional chlorination for the disinfection of wastewater.

The specific objectives were:

1. To assemble and evaluate the available information concerning the chemistry of ClO_2 generation and its behavior in aqueous solu-

tion, the technology and costs of manufacture, its effectiveness as a disinfectant, and the possible side effects of its use.

2. To establish the dose-effectiveness relationship for ClO_2 as a disinfectant of wastewater after secondary treatment and after various stages of advanced treatment, using the survival of coliform bacteria as a criterion.
3. To compare the effectiveness of ClO_2 with that of Cl_2 , using a variety of indicators.
4. To demonstrate continuous generation of and disinfection with ClO_2 to fulfill coliform requirements under conditions representative of wastewater treatment.
5. To prepare a preliminary design for treatment plants of 0.04, 0.22, 0.44, 2.2, and 4.4 m^3/s capacity and to estimate the costs of construction and operation.
6. To obtain preliminary evidence as to whether the formation of chlorinated organic byproducts during wastewater disinfection conforms to the results from studies of water disinfection.

Procedures and Results

In the laboratory, ClO_2 was generated by reacting NaClO_2 solution with H_2SO_4 to produce gaseous ClO_2 , which was purified by passing through a NaClO_2 tower before being absorbed into water. The concentration of ClO_2 in the prepared solution was approximately 2 g/L. In field experiments, ClO_2 was generated by continuously mixing NaClO_2 solution with either H_2SO_4 , HCl , or Cl_2 gas in a commercially available reactor system. Chlorine species in the reaction product were determined by a series of methods that entailed measurement of ultraviolet absorbance at 360 nm to determine ClO_2 ; amperometric titration at pH 7 to determine ClO_2 and Cl_2 ; iodometric titration at pH 2 to determine ClO_2 , Cl_2 , and ClO_2^- ; iodometric titration in concentrated acid to determine ClO_2 , Cl_2 , ClO_2^- , and ClO_3^- ; and chloride measurement by use of the Mercuric Nitrate Method. The sensitivity and precision of the determination are summarized in Table 1.

The yields of ClO_2 using several generation schemes are summarized in Table 2. The yield of ClO_2 from a continuous generator at full-scale (2 kg ClO_2/hour) was higher than the yield observed in a batch process in the

Table 1. Sensitivity and Precision of Analyses for the Reaction Yield Study

Species	Detection Limit, Mol/L	Precision	
		Coefficient of Variation, %*	At Mean Concentration, Mol/L
Chlorine Dioxide	3×10^{-5}	0.1	0.02
Chlorine	5.6×10^{-5}	0.6	0.015
Chlorite	3×10^{-4}	1.7	0.013
Clorate	1.8×10^{-4}	0.4	0.01

* $(\text{Standard deviation divided by mean}) \times 100$ for 5 replicate measurements in distilled water.

Table 2. Yield of Chloride Dioxide

Type of Generation	Ratio of Reactants	Initial Chlorite Conc., M	Final pH	Yield, %*	n**
$\text{H}_2\text{SO}_4 + \text{NaClO}_2$, batch (laboratory)	$\frac{1.7 \text{ Mol H}_2\text{SO}_4}{\text{Mol ClO}_2^-}$	0.083	1.8	48	1
$\text{H}_2\text{SO}_4 + \text{NaClO}_2$, continuous (field)	$\frac{2 \text{ Mol H}_2\text{SO}_4}{\text{Mol ClO}_2^-}$	1.56	2.1	51	1
$\text{HCl} + \text{NaClO}_2$, continuous (field)	$\frac{1.4 \text{ Mol HCl}}{\text{Mol ClO}_2^-}$	1.35	2.2	78.4 ± 4.4	4
$\text{Cl}_2 + \text{NaClO}_2$, continuous (field)	$\frac{0.52 \text{ Mol Cl}_2}{\text{Mol ClO}_2^-}$	1.92	5.3	95 ± 3.5	2

*Yield as $(\text{Mol ClO}_2 \text{ produced} / \text{Mol ClO}_2^- \text{ feed}) \times 100$; mean \pm standard deviation.

**n = Number of trials.

laboratory; this is attributed to the higher concentration of NaClO_2 used in the full-scale, continuous system. The yield from continuous generation was increased when HCl was substituted for H_2SO_4 in the acid-chlorite process. The yield from the chlorine-chlorite process in the continuous generator was 95%, even though a small excess of Cl_2 (4% above the stoichiometric requirement) was used.

Disinfection experiments were carried out using secondary wastewater samples from three plants (Figure 1). The levels of treatment were: conventional activated sludge; nitrified activated sludge; and filtered, nitrified activated sludge.

Laboratory disinfection experiments were conducted in a 4-L batch reactor with disinfectant dose levels of 2, 5, and 10 mg/L and contact time levels of 5, 15, and 30 min as the independent variables in a full factorial design. The density of total coliforms was determined by the membrane filter method; disinfectant residual concentrations were determined by amperometric titration. The results were correlated as survival ratio, $N(t)/N(0)$, versus the product of residual \times contact time ($R \times t$). Typical results from a set of experiments using

ClO_2 to disinfect conventional activated sludge effluent are shown in Figure 2.

Full-scale disinfection experiments were carried out at a treatment plant to confirm the reliability of the laboratory data to predict plant performance. Laboratory and field data agreed well for both ClO_2 and Cl_2 .

ClO_2 was found to be a more effective disinfectant than Cl_2 when treating conventional (nonnitrified) activated sludge effluent (Figure 3). This difference was shown to be statistically significant by analysis of variance. When comparing ClO_2 to Cl_2 at a given survival ratio, a lesser value of $R \times t$ product suffices to achieve a given degree of coliform inactivation when ClO_2 is used (Figure 3). A similar comparison made for nitrified effluent indicated no appreciable difference between the two disinfectants to inactivate coliforms. When these comparisons were made for nitrified filtered effluents, however, ClO_2 was more effective than Cl_2 based on the same criteria.

The costs of disinfection with ClO_2 are compared with those of Cl_2 for six cases corresponding to two levels of pretreatment and three total coliform standards

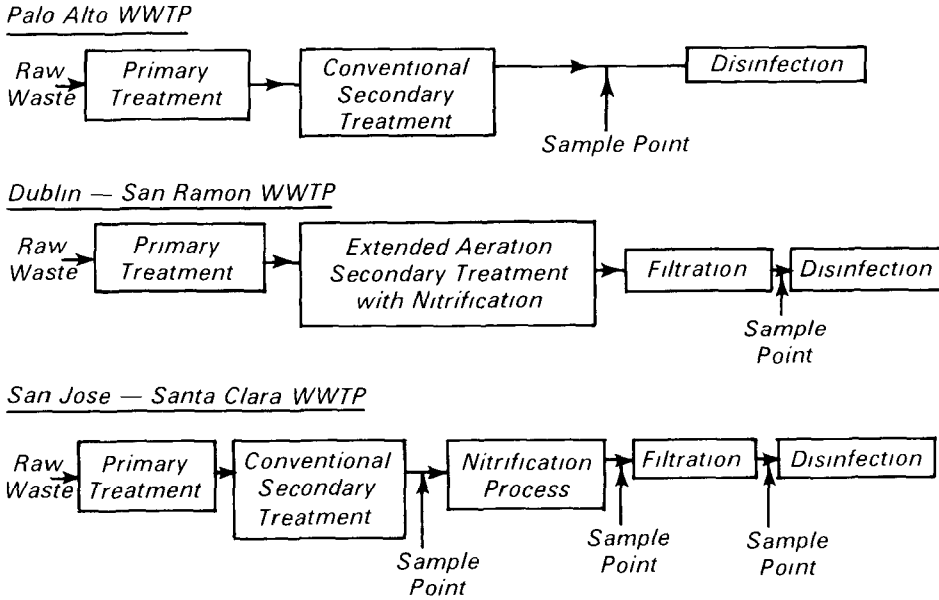


Figure 1. Wastewater treatment plant flow schemes and sampling points for disinfection experiments.

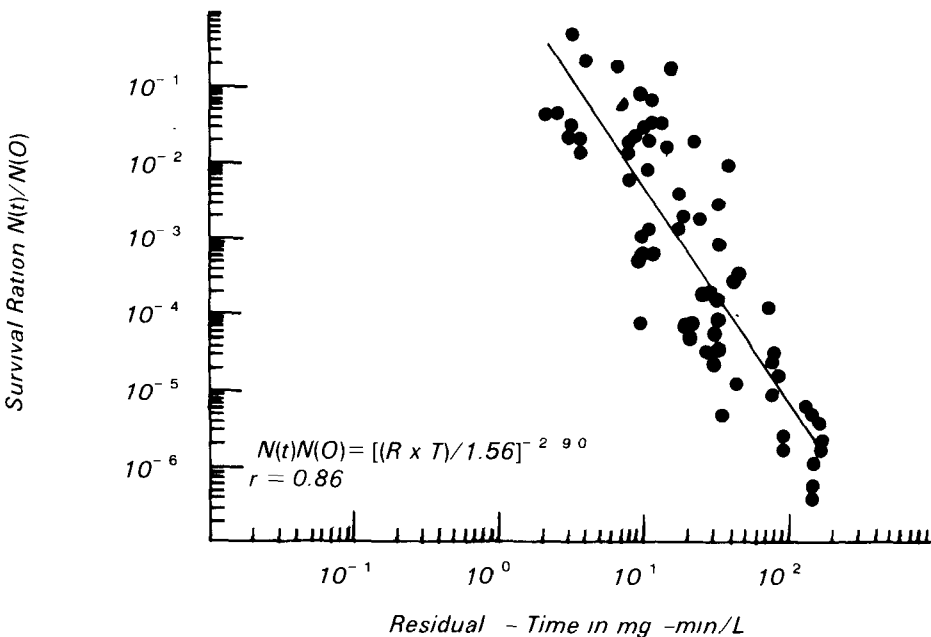


Figure 2. Data correlation for disinfection of conventional activated sludge effluent with chlorine dioxide (laboratory experiments).

(Table 3). Estimates for a 0.44 m³/s plant are shown in Table 3; the report presents cost estimates for a range of capacities from 0.04 to 4.4 m³/s. The high cost of NaClO₂ is responsible for the generally high cost of disinfection with ClO₂.

ClO₂ offered advantages that compensate for its cost being higher than that of chlorine. ClO₂ was found to be more effective for inactivating inoculated Poliovirus 1 and natural populations of coliphage than was Cl₂ in both non-nitrified and nitrified filtered wastewater effluents. ClO₂ treatment formed no measurable amounts of trihalomethane byproducts, whereas Cl₂ treatment formed 0.5 to 5 μMol/L of trihalomethanes, chiefly chloroform, in experiments using wastewater effluents. Moreover, ClO₂ formed negligible amounts of the broader class of halogenated organics measured collectively as total organic halogen (TOX)—less than 10% as much TOX as did chlorine. These advantages of ClO₂ should be considered, along with the cost-effectiveness comparison based on coliform kill, to reach decisions on using ClO₂ as a disinfectant in wastewater treatment.

The full report was submitted in fulfillment of Grant No. R-805426 by Stanford University under the sponsorship of the U.S. Environmental Protection Agency.

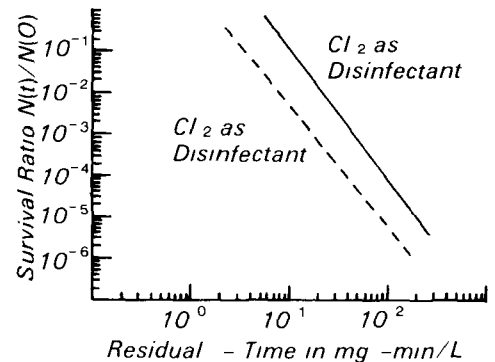


Figure 3. Comparison of coliform inactivation by chlorine dioxide and chlorine in conventional (nonnitrified) activated sludge effluent.

Table 3. Summary of Dose Requirements and Costs

Type of Effluent	Case	Total Coliform Standard, N/100 ml	Required Disinfectant Dose, mg/L		Total Costs,* ¢/m ³	
			Cl ₂	ClO ₂	Cl ₂	ClO ₂
Activated Sludge	A	2.2	7.89	7.92	0.82	8.72
	B	200.	2.45	2.90	0.58	3.43
	C	1000.	1.70	2.17	0.55	2.24
	D	2.2	11.15	5.52	0.95	6.21
Filtered, Nitrified Activated Sludge	E	200.	2.61	0.60	0.63	1.11
	F	1000.	2.60	0.14	0.58	0.61

*January 1980 costs, 0.44 m³/s plant.

Paul V. Roberts, E. Marco Aieta, James D. Berg, and Bruce M. Chow are with the Civil Engineering Department, Stanford University, Stanford, CA 94305. **Mark C. Meckes** is the EPA Project Officer (see below).

The complete report, entitled "Chlorine Dioxide for Wastewater Disinfection: A Feasibility Evaluation," (Order No. PB 81-213 357; Cost. \$14.00, subject to change) will be available only from:

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Project Summary

Wastewater Treatment by Rooted Aquatic Plants in Sand and Gravel Trenches

Pamela R. Pope

A patented* process developed by the Max Planck Institute (MPI) of West Germany to treat industrial wastes was evaluated as an energy-efficient method to treat municipal wastewater. The major goal was to achieve effluents meeting the U.S. Federal Effluent Standards using this novel biological treatment process that requires a minimal amount of mechanical equipment and manpower for normal operation.

The Moulton Niguel Water District (MNWD) of Laguna, California, constructed and operated an earthen trench system using rooted aquatic plants for the treatment of wastewater. Two trenches in series were planted with the reed *Phragmites* and the bulrush *Scirpus*, respectively.

A 2-month study using conventional secondary effluent as the trench influent showed the system was not effective for removing nitrogen and phosphorus components.

An 11-month study demonstrated that raw screened wastewater applied to the trench system at a rate not exceeding 95 m³/d (25,000 gpd) could be treated to secondary effluent quality. Spatial requirements were about the same as for a septic tank system.

This Project Summary was developed by EPA's Municipal Environmental Research Laboratory, Cincinnati, OH, to announce key findings of the research project which is fully docu-

mented in a separate report of the same title (see Project Report ordering information at back).

Introduction

The MPI process utilizes higher aquatic plants, such as reeds and bulrushes, for the treatment of wastewater. The system consists of two earthen trenches, lined with impervious membranes, operated in series. The first, designated as the filter trench, removes coarse suspended solids from the wastewater. The second, designated as the elimination trench, removes dissolved materials from the effluent of the first trench.

The MNWD services a residential area, and the wastewater is domestic in nature. The MPI system, as it was installed at the MNWD 3A facility, consists of two filter trenches and two elimination trenches. Two species of plants were used—a reed *Phragmites communis* in the filter trench and a bulrush *Scirpus lacustris* in the elimination trench. A view of the elimination trench system during construction is shown in Figure 1.

Filter Trenches

The two filter trenches are each 25 m (75 ft) long, 4 m (12 ft) wide, and 1.3 m (4 ft) deep. They are filled with three layers of gravel—150 mm (6 in.) of 50-mm (2-in.) gravel on the bottom; 225 mm (9 in.) of 19-mm (¾-in.) gravel in the middle,

*U.S. Patent 3,770,623, November 6, 1973

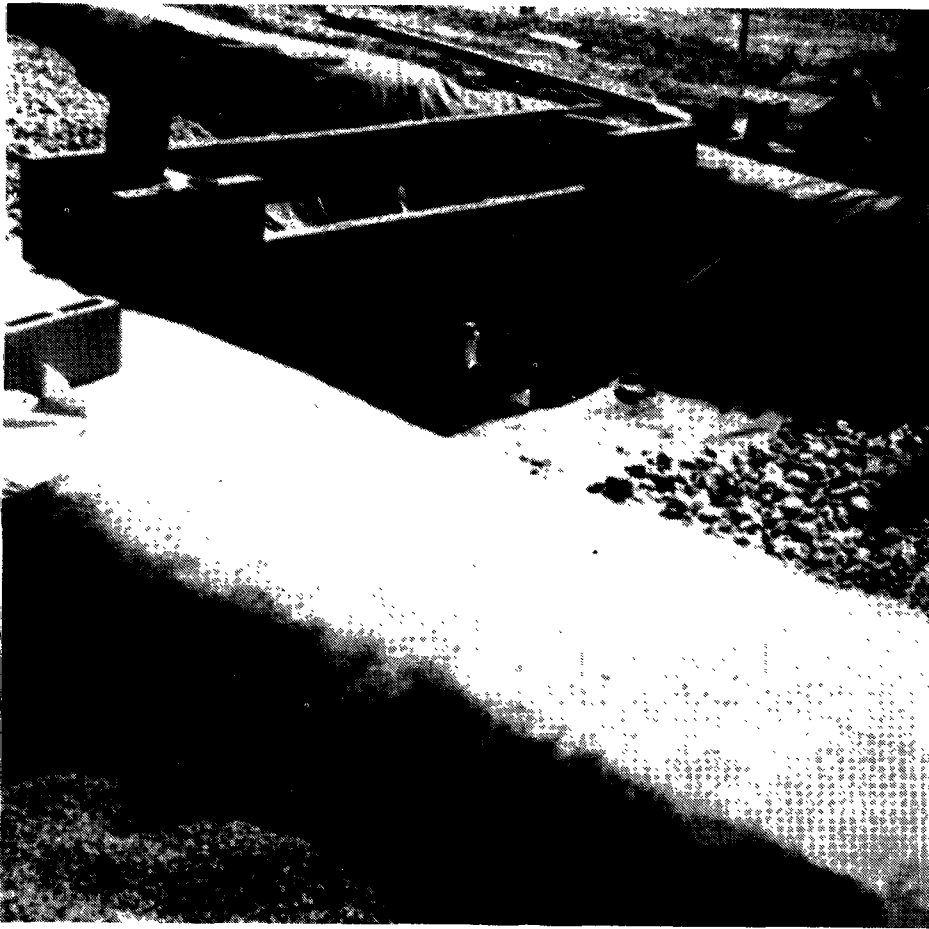


Figure 1. Construction of elimination trench.

and 75 mm (3 in.) of pea gravel on top. A 75-mm (3-in.) layer of silica sand covers the top. The raw wastewater enters the MNWD 3A facility at the north side of the plant and is passed through a rotor-strainer to remove large particles.

The screened influent flows down an influent channel and is pumped to the MPI system at a rate of approximately 8.8 to $9.5 \times 10^{-4} \text{ m}^3/\text{s}$ (14-15 gpm). The influent then flows south through a control valve into the east end of the filter trenches. Every 24 hr the flow is alternated between the two trenches. Each filter trench contains a central 150-mm (6-in.) plastic pipe with an open slit running its entire length; alternating long and short 100-mm (4-in.) plastic pipes extend from it for even dispersion of the influent. The wastewater flows down the central pipe through the extending pipes and onto cement splash pads located directly below. The wastewater percolates

through the trench in a vertical filtering action leaving a sludge layer on top. The sand filters out the suspended solids while the plant system draws moisture and nutrients. Slow drying of the deposited solids occurs, and extensive growth of the plant rootlets and runners aid in degrading the sludge layer on top of the sand. Each trench has a perforated 100-mm (4-in.) plastic pipe extending the entire length of the trench from the surface to the bottom in a "U"-shaped configuration. Flow from the underdrain goes into this pipe and through pipe extensions and a butterfly valve into a sump. This pipe not only transports the flow but allows aeration to the bottom since it has openings to the surface. The sump is a 1.3-m (4-ft) concrete pipe 3.3 m (10 ft) in height and is buried 2.6 m (8 ft). A small 0.25-kW (0.33-hp) pump, activated by a float, periodically pumps the flow to the elimination trenches.

Elimination Trenches

Normally, this process would be a total gravity flow system, with the elimination trenches so placed as to facilitate this, but because of site conditions at this location, the elimination trenches had to be constructed 90 m (100 yd) north of the filter trenches. The two elimination trenches are 50 m (150 ft) long, 4 m (12 ft) wide, and 0.75 m (2.5 ft) deep. They are divided in the center by a weir designed to allow composite sampling in this area and to aid in aeration. The filter trench effluent enters two 150-mm (6-in.) plastic pipes 4 m (12 ft) long set perpendicular to the trenches at the south side. The side facing the trenches is open, and the liquid is allowed to flow out into an area 1.3 m (4 ft) by 4.0 m (12 ft) in a waterfall-like action. This area is filled with 15 mm ($\frac{5}{8}$ in.) gravel held in place by 50- X 100-mm (2- X 4-in.) wood baffles. A later observation (by BWP of New York Inc.) showed that eliminating the baffles reduced the operational problems. The liquid percolates down in a horizontal manner at a level approximately up to 50 mm (2 in.) below the surface of the trenches. The trenches are filled with 15-mm ($\frac{5}{8}$ -in.) gravel with 75 mm (3 in.) of pea gravel on top. The flow passes through the weir and runs into two standpipes that lead into a sump. The level of the liquid flow is governed by raising or lowering the standpipes. A valve in the bottom of the weir allows periodic draining of the liquid in the lower portions of the trenches. The total retention times for the entire system are estimated to be 6 hr and 8.5 hr at flows of $133 \text{ m}^3/\text{d}$ and $95 \text{ m}^3/\text{d}$, respectively.

Test of MPI System as a Tertiary Treatment Process

Secondary effluent from the MNWD extended aeration plant was introduced to the MPI system on July 1, 1978, at a loading rate of $56 \text{ m}^3/\text{d}$ (15,000 gpd); the flow was increased to $95 \text{ m}^3/\text{d}$ (25,000 gpd) in August.

The analytical results for samples obtained during the time extended aeration effluent was applied to the system are shown in Table 1.

Overall removal of BOD_5 , VSS, and TSS was about 50 percent each month. COD reduction was about 40 percent. Ammonium nitrogen removal of 67 percent during August was superior to the 40 percent removal in July. Consideration of the $\text{NH}_4\text{-N}$, $\text{NO}_3\text{-N}$, and $\text{NO}_2\text{-N}$ values between the 2 months indicates

Table 1. Tertiary Treatment Results for MPI System, Average Values

Item, mg/L	MNWD Secondary Effluent	Filter Trench Effluent	Elimination Trench Effluent	Overall Removal, %
Flow of 25,000 gpd (95 m ³ /d)				
BOD ₅	15	9	7	53
TSS	15	8	7	53
VSS	10	6	4	60
COD	58	50	35	39
NH ₄ -N	12	8	6	50
NO ₂ -N	0.8	2	1.1	--
NO ₃ -N	2.1	5.5	7.2	--
TP	11.5	10	10	13

nitrification and subsequent denitrification were more active in the system during the warmer month of August. TKN samples were not collected during this period. The MPI system operated as a tertiary process did not efficiently remove TP. The extended aeration effluent applied to the system was of good quality. The filter trench achieved the greater part of the overall pollutant removals; the elimination trench showed only marginal incremental removal.

Test of MPI System as a Secondary Treatment Process

Screened raw wastewater was used as the influent to the MPI system in mid-September at a rate of 56 m³/d (15,000 gpd). The flow was increased to 95 m³/d (25,000 gpd) in October and 133 m³/d (35,000 gpd) in January, remaining at this rate through July. The growth of the bulrush *Scirpus* in the elimination trench is shown in Figure 2.

A thin sludge layer built up and completely covered the filter trenches by the end of October. At this time, the *Phragmites* had not spread throughout the trenches. Algae also started to grow on the sludge layer and may have contributed to later clogging problems. By mid-December, algae covered a large portion of the sludge layer, which was not drying or breaking up as the flow was directed into the alternate trench. The problem occurred equally in both filter trenches. Operation was suspended at this time to consider this

problem and to harvest the *Phragmites* from the filter trenches. The *Phragmites* had turned brown because of unusually cold weather and had begun to lay over because of their mature height and weight. When the sludge layer was skimmed off a little at a time, about 25 mm (1 in.) of sludge was found deposited on the filter media; this sludge layer was wet and becoming septic. Below this was a layer of compacted organic matter and fibers that were black in appearance and felt greasy; this intermingled with the sand, formed an almost impermeable layer. If a hole was poked through this layer, the liquid held in the sludge layer immediately drained through the remaining sand. The only solution at this time was to allow the filter trenches to dry after the harvesting and then to rake out the semidry top sludge layer carefully. Figure 3 shows the plants after harvesting.

Subsequent tests conducted in Long Island, New York, by BWP of New York, Inc., showed that using four parallel filter trenches to allow increased drying time and that draining the filter trenches three times a week minimized this sludge problem.

The major objective of this project was to evaluate the MPI system as a low-cost wastewater treatment alternative that would satisfy federal discharge requirements. These requirements are attained if final effluent BOD₅ and SS concentrations do not exceed 30 mg/L for 30 days average values, or 85 percent overall removal, whichever is more stringent. The fate of nitrogen and

phosphorus was also monitored. Table 2 summarizes all the data collected.

The system was evaluated for secondary treatment effectiveness for 11 months. For 5 of the months, the flow through the system was 95 m³/d (25,000 gpd) or less; for 6 months, 133 m³/d (35,000 gpd). Secondary treatment requirements for BOD₅ and SS were achieved all 5 months at the lower flow SS residuals and percent removals met secondary requirements all 6 months at the higher flow rate; however, the BOD₅ requirement was not achieved for 5 of the 6 months. The effluent violated both the concentration and percent removal requirements three times (January, April, and July); the percent removal requirement only was violated twice (May and June).

There was little difference in overall COD removal for the two application rates.

The NH₄-N and Org-N concentration values in the effluent during the periods of 95 m³/d application were representative of conventional secondary treatment residuals. Variations in percent removals were because of fluctuations in influent concentrations. The overall removal of total nitrogen varied from 61 percent in September to 32 percent in March.

During application of 133 m³/d, the Org-N residuals were about twice the values of the lower flow rate results. During February, a negative removal of Org-N was noted. The overall removal of total nitrogen was much lower than during the 95 m³/d application, 18 percent in January to 36 percent in June.

Nitrite and nitrate nitrogen concentrations for all the sample periods show that nitrification did not occur to any significant extent at either of the two flow rates.

The MPI system during both the 95 m³/d and the 133 m³/d application rates was not effective for total phosphorus removal. During the higher application rate, 2 months (January and June) showed negative removals.

The major increment of BOD₅, SS, VSS, and COD removal occurs at the filter trench, and the elimination trench serves as a polishing process (Table 2). Both trenches in series are necessary for satisfactory treatment.

The MPI system operated with raw screened wastewater at an application rate of 95 m³/d did achieve secondary effluent quality. Using the trench measurements, the spatial requirements of



Figure 2. Aquatic plant growth in elimination trench.

the MPI system equate to $0.02 \text{ m}^3/\text{m}\cdot\text{d}$ ($0.5 \text{ gpd}/\text{ft}^2$). Assuming a per capita wastewater discharge of 378 L (100 gal), the area required is $2 \text{ m}^2/\text{capita}$ ($21 \text{ ft}^2/\text{capita}$). These two values are very similar to spatial requirements of a septic tank system located in a satisfactory percolating soil.

Several operating problems, expected with new technology development, were experienced during this demonstration study. Many of the same operational problems were encountered at the Long Island, New York.

Several operating problems, expected with new technology development, were experienced during this demonstration study. Many of the same operational problems were encountered at the Long Island, New York, installation. Remedial measures applied at Long Island included:

1. Provision for increased area for the filter trenches, thereby allowing longer idle times for drying.

2. Recommend harvesting plants not more than once a year. Frequent harvesting of the plants used in the system promotes extra growth of the root systems and this contributes to clogging.
3. If plant growth becomes excessive during the year, individual plants are culled by pulling to thin the growth.

This initial assessment of the efficiency and spatial considerations for the MPI system for secondary treatment indicates it is worthy of further development.

The full report was submitted in fulfillment of Grant No. R-805279 by the Moulton Niguel Water District under the sponsorship of the U.S. Environmental Protection Agency.



Figure 3. Filter trench after harvest of plants.

Table 2. Secondary Treatment Results for MPI System, Average Values in mg/L

Sample Location	BOD ₅	TSS	COD	NH ₄ -N	Org-N	NO ₂ -N	NO ₃ -N	TP
Flow of 25,000 gpd (95 m ³ /d)								
Influent Wastewater	210	225	405	24	13	0.0	0.1	13
Filter Trench Effluent	77	41	179	19	9	0.4	0.9	12
Elimination Trench Effluent	26	20	86	16	5	0.1	0.4	12
Overall Removal, Percent	88	91	79	33	31	--	--	8
Flow of 35,000 gpd (133 m ³ /d)								
Influent Wastewater	171	181	405	25	17	0.0	0.3	13
Filter Trench Effluent	68	48	157	21	13	0.6	1.2	13
Elimination Trench Effluent	35	19	93	19	11	0.3	0.6	13
Overall Removal, Percent	80	89	77	24	35	--	--	0

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Ronald F. Lewis is the EPA Project Officer (see below).

The complete report, entitled "Wastewater Treatment by Rooted Aquatic Plants in Sand and Gravel Trenches," (Order No. PB 81-213 241; Cost: \$6.50, subject to change) will be available only from:

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