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RANKING OF COAGULANTS FOR WASTEWATER TREATMENT USING PARTIAL ORDER THEORY

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SUMMARY

Jar-test is a useful tool for chemicals selection for physical–chemical wastewater treatment. The results show the treatment efficiency in terms of suspended matter and organic matter removal. However, in spite of having all these results, coagulant selection is not an easy task because one coagulant can remove efficiently the suspended solids but at the same time increase the conductivity. In this paper, the use of Partial Order Scaling Analysis (POSA) is proposed to help on the selection of the coagulant and its concentration in a sequencing batch reactor (SBR). An evaluation of two commonly used coagulation-flocculation aids was conducted and based on jar tests and POSA model, Ferric Chloride (100 ppm) was the best choice.

Keywords: *Coagulation, Jar Test, Partial Order Scaling Analysis, Treatment Selection.*

1. INTRODUCTION

One of the most commonly used methods for the removal of suspended solids in wastewater is the addition of coagulant and flocculation aids, such as Alum, Ferric Chloride, and long chain polymers (Ebeling, Sibrell, Ogden and Summerfelt, 2003). Coagulation, flocculation and clarification, followed by filtration, are the key steps in conventional wastewater treatment systems. This is a well-proven technology for the significant removal of color and particulate matter including protozoa (e.g. Cryptosporidium oocysts and Giardia cysts), viruses, bacteria, and other micro-organisms. Iron, manganese, tastes and odors may also be removed from the water by these processes (Aguilar, Saez, Llorens, Soler and Ortuno, 2002; Aguilar, Saez, Llo-

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rens, Soler and Ortuno, 2003). The reuse of treated wastewater in irrigation provides a cost effective and environmentally friendly alternative to wastewater disposal. When wastewater is adequately treated it can be safely applied to crops, and poses no greater risk to the consumer than conventional sources (Oron, Gillerman, Lal, Manor, Braude and Bick, 2010).

The treatment has several distinct stages: a coagulant is rapidly and uniformly dispersed through the mass of water. The processed water will then enter a flocculation chamber and a gentle mixing during this stage allows particles to agglomerate and form settleable flocs. Clarification usually follows the flocculation process and involves sedimentation or settling, which allows the formed flocs to be separated for subsequent removal as sludge. Clarification is then followed by filtration which provides a second, polishing step for particulates that were not removed during the clarification step (Friedler, Katz and Dosoretz, 2008).

Nearly every water treatment plant uses Aluminum-based coagulants [e.g. (Alum) or Poly-Aluminum Chloride (PACl) or iron-based coagulants (Ferric Chloride or Ferric Sulphate)] (Duan and Gregory, 2003; Al-Mutairi, Hamoda and Al-Ghusain, 2004; Selçuk, Kaptan and Meriç, 2004; Dominguez, de Heredia, Gonzalez and Sanchez-Lavado, 2005; Jung, Chanudet, Ghanbaja, Lartiges and Bersillon, 2005; Matilainen, Lindqvist and Tuhkanen, 2005; Rodríguez, Kennedy, Diepeveen, Prummel and Schippers, 2008; Yan, Wang, Qu, Ni and Chow, 2008; Silva, Hornes, Mitterer and Queiroz, 2009). Alum has been used for several centuries in water treatment and is probably the most well known and commonly used coagulant. The chemical is prepared by the reaction of certain clays with H_2SO_4 acid and delivered in granular or powder form (Selçuk *et al.*, 2004).

Ferric Chloride is highly acidic and the solution contains free hydrochloric acid. The solution is highly corrosive to nearly all normally used metals including all grades of stainless steel and needs to be stored, pumped and conveyed in synthetic corrosion-resistant materials. The chemical is normally supplied as a solution of about 40% strength as $FeCl_3$ with specific gravity of about 1.4 and a pH of less than 1.0 (Dominguez *et al.*, 2005).

The best approach for determining the treatability of a water source and determining the optimum parameters (most effective coagulant, required dose rates, pH, and flocculation times) is by using a jar-tester (Tatsi, Zouboulis, Matis and Samaras, 2003).

It is always preferable to carry out tests on a number of samples and if possible, under different conditions to establish the most reliable product. Having selected a suitable product, the use of routine jar tests remains necessary for a number of reasons: (i) the nature and quality of the raw water may change, which may affect the coagulant dose, (ii) it is necessary to check that the plant dosage matches the demand established in the laboratory, and (iii) different batches of coagulant may vary and the use of comparative jar tests using some of the original product sample is a useful quality check. The normal procedure when conducting a jar-test is**:** (i) initially to find the best performing coagulant and dose rate, and (ii) to determine the optimum pH for the chosen coagulant and dose rate. Coagulation-flocculation performance is usually judged on the basis of turbidity, color and organic constituent removal (Kim, Moon and Lee, 2001; Franceschia, Girou, Carro-Diaz and Maurette, 2002; Guida, Mattei, Rocca, Melluso and Meriç, 2007; Xiao, Ma, Yi and Huang, 2008).

Conventional partial order ranking may appear insufficient to solve problems involving a high number of parameters as the number of parameters may well appear to be prohibitive for developing a robust model (Carlsen, 2008). A possible improvement is to apply weights within a step-by step procedure or to use fuzzy partial order concepts (Brüggemann, Voigt, Restrepo and Simon, 2008a).

The objective of this study was to define a simple procedure useful in selecting the best coagulant and the relative dose for the treatment of wastewater. The specific aim was to design and optimize coagulation-flocculation process for the treatment of Municipal Solid Waste (MSW) in Hiria (Israel): (i) to investigate a first set of jar tests using different concentrations of Alum and Ferric Chloride for wastewater treatment, (ii) to add economical parameters in order to weight these parameters, according to the judgments of wastewater experts, (iii) to maximize the removal of organic constituents, minimize coagulant dose and cost and optimize the performance of a wastewater system which was equipped with a coagulation-flocculation process, and (iv) to estimate the application of the Partial Order Scaling Analysis for decision making in water and wastewater technology.

2. MATERIALS AND METHODS

2.1 *Analytical methods*

Samples were collected from the drainage of a sequencing batch reactor (SBR) plant of the ‘‘Arrow Ecology’’ in Hiria (Israel) and the tests were carried out on the same day. Coagulation and flocculation studies were performed in a standard jar test apparatus (Velp Scientifica JLT6) comprised of six paddle rotors and equipped with 6 beakers of 1 L volume. Samples were diluted 1:50 with potable water and thoroughly agitated (100 rpm) for re-suspension of settled solids before any tests were conducted. Chemical reagents used as coagulants are commercially available from ‘‘Arrow Ecology’’ company and include Alum [$Al_2(SO_4)_3.18H_2O$] and Ferric Chloride ($FeCl_3$).

The initial rapid mixing for all experiments was taken as 5 minutes (100 rpm) and for slow mixing at 25 minutes (25 rpm). After settling (duration 30 minutes), about 50 mL of supernatant was withdrawn using a plastic syringe from the point located about 4 cm below liquid level for the determination of COD, TSS, TFS, and TVS (Analyses were made in triplicates).

Pure Alum is white and gives a water-white solution. However the presence of iron as an impurity is common which gives the chemical or the solution a yellow or even an orange color. The wastewater plant uses cheap Alum that was prepared from low-grade clays and from waste acid. This leads to the presence of undesirable concentrations of heavy metals in the solution. Metals are not a problem as they are coagulants in their own right and tend to assist the coagulation process. Ferric Chloride is originally a waste product from spent pickling solution and the wastewater plant uses a cheap product that was prepared from reacting scrap iron with hydrochloric acid and supplied as a solution of about 40% strength as $FeCl_3$.

Coagulant quality control was performed by the measurement of the specific gravity which is both rapid and simple using a hydrometer. If this parameter differs by more than (5%) other parameters such as the pH and the viscosity were checked.

Standards of good laboratory practice such as the maintenance and periodic assessment of equipment, instrumentation, consumable supplies were applied. Precautions were taken to ensure that (i) the workers were familiar with the dangers and treatment associated with these coagulants, and (ii) there were minimum interferences caused by changes in raw water conditions.

TABLE 1. - *Indicators and cost data*

Indicator	Alum	Ferric Chloride	Remarks
Coagulant concentration			Based on jar test
Cost **(*)** $ per ton (dry)	253	472	Based on ‘‘Arrow Ecology’’ data and chemical market price in 2009
COD	-	-	Based on jar test
TFS	-	-	Based on jar test
TVS	-	-	Based on jar test
TSS	-	-	Based on jar test

* Technical grade, based on database of purchase prices in 2009 delivered by ‘‘Arrow Ecology’’.

2.2 *Management modeling and Partial Order Scaling Analysis (POSA)*

2.2.1 Coagulant selection: models

There is a tremendous need for research and models in the field of wastewater treatment and a great deal of research is required to accurately define the magnitude of adverse effects of the waste generated from various plants that use coagulation technology (especially clinical laboratories and multi-specialty hospitals) (Gautam, Kumar and Sabu, 2007). The limitations of using jar tests for determining optimum coagulant doses can be overcome by using mathematical models. Concerning the academic literature, there are few economic, statistical and multivariate models for optimal coagulant selection (Silva *et al.*, 2009). The technical and the economic studies are based on: (i) the identification of the coagulants for various design and operating parameters, and (ii) computation of the total annual cost (for selected coagulant, capacity and waste characteristics) for diverse pH at different coagulant doses (Ahmed, 2007).

Concerning statistical models, there are two types for coagulant selection: (i) simulation (linear and multifactor nonlinear), and (ii) process models. Contrary to the multiple regression models, the general linear model can analyze simultaneously more than one dependent indicator. The selection of the optimum coagulation conditions is carried out by post hoc analysis using Duncan test. Post hoc analysis determines if a certain difference between removal efficiencies is actually significant or not (Bhatia, Othman and Ahmad, 2007).

In conventional multifactor experiments, optimization is usually carried out by varying a single factor while keeping all other factors fixed at a specific set of conditions. It is not only time-consuming, but also usually incapable of reaching the true optimum due to ignoring the interactions among indicators. On the other hand, the Response Surface Methodology (RSM) has been proposed to determine the influences of individual factors and their interactive influences. The RSM is a statistical technique for designing experiments, building models, evaluating the effects of several factors, and searching optimum conditions for desirable responses. With RSM, the interactions of possible influencing parameters on treatment efficiency can be evaluated with a limited number of planned experiments (Park, Seo, Whang and Lee, 2000; Ahmad, Ismail and Bhatia, 2005; Ahmad, Wong, Teng and Zuhairi, 2007; Wang, Chen, Ge and Yu, 2007; Moghaddam, Moghaddam and Rami, 2010).

When process models are used, the data include process inputs (e.g. raw water quality parameters) and process control parameters (e.g. coagulant dose, pH) and the outputs of the process that is being modeled (e.g. treated water quality parameters) (Maier, Morgan and Chow, 2004; Bae, Kim and Kim, 2006; Chen and Hou, 2006; Al-Abri and Hilal, 2008). Although the utilization of process models overcomes the limitations of using jar tests for determining the optimal Alum or Ferric Chloride dose, the development of such models is not a trivial task. This is because water treatment processes are governed by complicated, nonlinear relationships (Al-Abri and Hilal, 2008).

2.2.2 Partial Order Scaling Analysis (POSA)

Scaling is based on a quasi-order that is a binary relation with the following characters: it is reflexive $(a \leq a \,\forall\, a)$, and transitive $(a \leq b)$ and $(b \leq c) \Rightarrow (a \leq c)$. If it is also anti-symmetric $(a \leq b)$ and $(b \leq a) \Rightarrow (a = b)$ then the quasi-order is called partial order. A partially ordered set (poset) is an ordered pair $(P, \leq)$, where P is a set and $\leq$ is a partial order relation on P (Annoni, Brüggemann and Saltelli, 2011).

Partial order as a discipline of discrete mathematics appears to be a promising tool for decision-making particularly in environmental issues (Voigt, Brüggemann, Scherb, Shen and Schramm, 2010). It has been argued that partial order theory may be the most objective way to rank a set of elements (Carlsen, 2004; Brüggemann and Voigt, 2011). The objectivity lies in the fact that in contrast to other multi-criteria methodologies, there is no need to unify the descriptors using weighting coefficients in any kind of functional relationships.

Data analysis is based on the the definitions of criteria and indicators: (i) criteria describe a numerical realization by a set of indicators, and (ii) indicator values characterize any coagulant by a vector, with components 1,.., m. A data matrix results with the rows characterizing the coagulants and the columns realizations of the indicators. A transformation is performed giving the entries of the data matrix discrete values. The transformed data matrix is the starting point for POSA. POSA determines the placement of coagulants along two perpedicular axes, the two - dimensional representation is called mapping diagram and is used for the selecton of the optimal coagulant.

Several POSA models are in use for academic research (POSAC, HUDAP, and CoPlot). The weak monotonicity coefficient, denoted by μ is of special use by POSA software (Shye, 1995). It varies between –1.00 and +1.00. $\mu = +1.00$ implies a perfect monotone trend in a positive direction; $\mu = -1.00$ implies a perfect monotone trend in a negative or descending direction. Given N pairs of data $\{(p_i, q_i);\ i = 1, 2, ..., N\}$ on two numerical indicators p and q, the weak coefficient of monotonicity μ between p and q is defined as follows:

$$\mu_{pq} = \frac{\sum_{i=1}^{N}(p_i - p_j)(q_i - q_j)}{\sum_{i=1}^{N}\sum_{j=1}^{N}|p_i - p_j||q_i - q_j|} \tag{1}$$

The aim of POSA is to present the data in two dimensions, preserving the partial order as well as possible in a mapping diagram in order to classify individuals. It is clear that for, empirical data, this procedure introduces some error in the mapping diagram. So, a coefficient which measures the goodness of fit must be defined. POSA gives such coefficient (CORREP, proportion of coagulat-pairs CORrectly REPresented):

$$\text{CORREP} = \frac{m_C + m_I}{M_C + M_I} \tag{2}$$

where for input coagulants: M_C is the number of comparable coagulant pairs and M_I the number of incomparable coagulant pairs referring to the original indicators. A similar notation holds for output (m_C and m_I). Two additional coefficients which measure the goodness of fit are defined: (i) CORREP1, proportion of comparable pairs that are CORrectly REPresented, and (ii) CORREP2, proportion of incomparable pairs that are CORrectly REPresented.

$$\text{CORREP1} = \frac{m_C}{M_C} \tag{3}$$

$$\text{CORREPE2} = \frac{m_I}{M_I} \tag{4}$$

In general a CORREP coefficient above 0.80 is regarded as indicating of a meaningful mapping diagram (Shye, 1995). The interpretation of POSA output is further illustrated in the literature (Brüggemann, Voigt and Pudenz, 2008b; Porter and Alison, 2001).

There has been a limited attempt in the literature to use POSA software for engineering design. In this paper, the proposed methodology adopted POSA software HUDAP to solve a decision design problem. The objective is to find the best coagulation performance frontiers from a set of jar test alternatives considering both quantitative and qualitative data.

2.3 *Coagulation-flocculation process: database*

Environmental engineering is concerned with waste in a wide range of natural waters and wastewaters and the source of pollution: domestic and industrial wastewater. All organic compounds, except few, can be oxidized by the action of strong oxidizing agents under acid conditions, regardless of the biological assimilability of the substances. This test Chemical Oxygen Demand (COD) is a rapid and precise method: oxidation by potassium dichromate in acid solution and the excess dichromate is titrated with standard ferrous ammonium sulphate using ferroin as an indicator (Singh, Malik, Mohan, Sinha and Singh, 2005). COD is used for determination of aggregate organic matter and measures organic concentration of both domestic and industrial wastewater.

The comparative performance of coagulation-flocculation process, carried out by the various alternative methods is very complex and is very complex and is highly dependable on various site-specific operational and economic parameters. In order to solve this problem, a pilot plant can be added in line to realize the origins of the effluent.

The process is subject to a series of specific site constraints and indicators (Table 1): (i) insertion of chemicals (coagulant concentration), (ii) coagulant cost, (iii) return for improved Chemical Oxygen Demand (COD), (iv) return for improved Total Fixed Solids [TFS-residue that remains after sample has been evaporated and dried at 103-105^{o}C and later ignited at 50050^{o}C], (v) return for improved Total Volatile Solids [TVS-Solids that volatized and burned off after sample has been evaporated and dried at 103 to 105^{o}C and later ignited at 500 50^{o}C], and (vi) return for improved Total Suspended Solids [TSS-portion of solids that retained on filter with specified pore size (1.58 μm), measured after sample has been evaporated and dried at 103-105^{o}C].

Chemical prices can be found at the internet (e.g. ICIS Chemical Business). The posted prices do not necessarily represent level at which transactions may have actually occurred, nor do they represent bid or asked price. The prices are intended as a guide and not to be used as a basis for negotiations between producers and customers. According to ICIS Chemical Business the coagulant prices are: Alum 331-425$/ton (100lb. bags, technical grade, 17% Al_2O_3) and Ferric Chloride 300-351$/

ton (Tanks, technical grade, 100% basis). In this research analysis the coagulants prices were based on database of purchase price in 2009 delivered by ‘‘Arrow Ecology’’: Alum, $253 per ton (dry) and Ferric Chloride $472 per ton (dry).

Wastewater treated by coagulation-flocculation process is defined by the following chemical characteristics based on Israel National Carrier feed: electrical conductivity 1.0dS/m, Na^{+1} 60-100mg/l, Ca^{+2} 45-50mg/l, Mg^{+2} 20-25mg/l, Cl^{-1} 200-220mg/l, SO_4^{-2} 17-20mg/l, HCO_3^{-1} 250-300mg/l (Yermiyahu, Tal, Ben-Gal, Bar-Tal, Tarchitzky and Lahav, 2007).

3. RESULTS AND DISCUSSION

3.1 *Jar test results*

The characteristics of raw drainage and Alum coagulation-flocculation are presented in Tables 2 and 3. In terms of COD and TSS, there is a high concentration of organic matter. Equal volumes (1,000 ml) of measured sample were delivered into each of the jars. Orion Model 420A was used for pH measurements and reagent grade chemical solutions (hydrochloric acid and Sodium hydroxide) were used for controlling the pH of samples.

As reported COD, TSS, TFS and TVS concentrations were specific to Alum test (Table 2) and Ferric Chloride test (Table 3), e.g. the mean values of COD in the effluent varied from 55 mg/l (Ferric Chloride treatment) to 84 mg/l (Alum treatment). COD removal efficiency in both treatments was very high (Ferric Chloride, 99.7% and Alum 99.6%). According to the test results the removal efficiency was almost stable (more than 95%) regarding the organic indicators COD, TSS, TFS and TVS. Test result pointed out that coagulation-flocculation process can guarantee high rejection of organic constituents for wastewater treatment plants.

TABLE 2. - *Samples treated with Alum*

Sample	Code	Alum (ppm)	Cost (Cent/m^3-feed*)	COD (ppm)	TSS (ppm)	TFS (ppm)	TVS (ppm)
1 Blank(**)	AL1	0	0	0	0	0	0
2 (***)	AL100	100	2.5	116	820	412	408
3 (***)	AL200	200	5.1	84	692	392	300
4 (***)	AL350	350	8.9	136	880	160	720
5 (***)	AL450	450	11.4	48	872	424	448
6 (***)	AL550	550	13.9	36	848	412	436

* 253$/ton (dry) technical grade, based on database of purchase prices in 2009 delivered by ‘‘Arrow Ecology’’. The blank sample in is referred to potable water (supplied by the Israel National Water Carrier) that contains no organic constituents.

** pH blank = 7.00.1.

*** pH samples = 7.80.1.

TABLE 3. - *Samples treated with Ferric Chloride*

Sample	Code	Ferric (ppm)	Cost (Cent/m^3-feed*)	COD (ppm)	TSS (ppm)	TFS (ppm)	TVS (ppm)
1 Blank(**)	FC1	0	0	0	0	0	0
2 (***)	FC100	100	100	20	564	116	448
3 (***)	FC200	200	200	40	960	388	572
4 (***)	FC350	350	350	120	944	268	676
5 (***)	FC450	450	450	44	1,204	536	668
6 (***)	FC550	550	550	52	1,096	268	828

* 472$/ton (dry) technical grade, based on database of purchase prices in 2009 delivered by ''Arrow Ecology''. The blank sample referred to potable water (supplied by the Israel National Water Carrier) that contains no organic constituents.
** pH blank = 7.00.1.
*** pH samples = 6.50.1.

3.2 *Statistical analysis*

A very important point in decision theory is that the values of the indicators are uncertain because of measurement errors. The only important element of the measure is the order they create among the elements of the domain. Indeed the orders are invariant under any strictly monotonous transformation. Therefore, a change of scale will not affect the conclusions. In order to examine the performance across all coagulants of interaction, the given set of measurements (Tables 2 and 3) were transformed using monotonous transformation (Zhu and Ding, 2007) (with the support of Dr. Amar from the Hebrew University of Jerusalem) to a new set of features (Table 4). In other words, what is important is the rank and not the quantity of distance between the various steps. If the transform is suitably chosen, transform domain features can exhibit high information packing properties compared with the original input samples. The basic reasoning behind transform-based features is that an appropriately chosen transform can exploit and remove information redundancies, which usually exist in the set of samples obtained by the measuring devices.

The transformed data (Table 4) was analyzed by partial order analysis technique {HUDAP software (MPOSAC)} (Amar, 2001; 2005). The MPOSAC algorithm provides a coefficient for the goodness of fit for the representation of the partial order: (i) CORREP coefficient [that displays the proportion of comparable pairs that are correctly represented, (Equation 2)] equals 0.9697, (ii) CORREP1 coefficient [that displays the proportion of profilies-pairs that are correctly represented (Equation 3)] equals 1.0000, and (iii) CORREP2 coefficient [that displays the proportion of incomparable pairs that are correctly represented (Equation 4)] equals 0.9474.

The coefficient of weak monotonicity (Equation 1) between each coagulant and the partitions along the X and the Y axis is presented in Table 5: a high correlation indicates a common direction. According to the results, indicator TFS goes with Y,

TABLE 4. - *Data analyzed*

Code	Coagulant concentration	Cost	COD	TSS	TFS	TVS
AL100	1	1	8	3	7	2
AL200	2	3	7	2	6	1
AL350	3	4	10	6	2	9
AL450	4	6	5	5	9	4
AL550	5	7	2	4	7	3
FC100	1	2	1	1	1	4
FC200	2	5	3	8	5	6
FC350	3	8	9	7	3	8
FC450	4	9	4	10	10	8
FC550	5	10	6	9	3	10

while COD goes with X. This means that the mapping diagram is essentially spanned by these two indicators.

The statistical analysis shows the power of POSA to organize coagulants simultaneously according to level of the sorted criteria and as functions of the content meaning of the base coordinates X and Y (Figure 1).

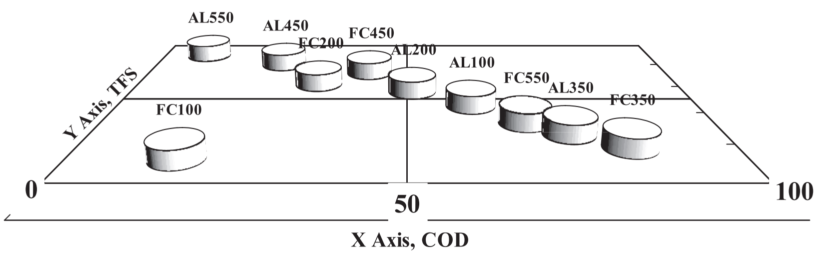

FIGURE 1. - *HUDAP software: output mapping*

TABLE 5. - *HUDAP software output: coefficient of weak monotonicity between each coagulants and the partitions along the X and the Y axis (*)*

Indicator	X	Y
Coagulant concentration	-0.07	0.65
Cost	0.22	0.37
COD	**0.97**	-0.44
TSS	0.51	0.25
TFS	-0.47	**0.96**
TVS	0.70	-0.51

* High correlation (bolt numbers) indicates a common direction

Overall, based on jar tests, and HUDAP software, 100 ppm Ferric Chloride (Code FC100) is the best system among all systems studied which shows the highest efficiency in terms of economic aspects and reduction of COD, TSS and TFS.

4. CONCLUSIONS

The design of a cost effective wastewater treatment process to achieve a desired good quality for irrigation can be very difficult, as a large number of treatment options are available. This process is further compounded by the many criteria that are needed to be considered in the selection course of action. A jar-test was used in order to test the performance of two commonly employed coagulation-flocculation aids (Alum and Ferric Chloride) for the treatment of the drainage of a sequencing batch reactor (SBR) plant of the ''Arrow Ecology'' in Hiria (Israel).

In order to get an optimal solution, an expert systems software based on partial order methodology were used. This study demonstrates the possibilities and appropriateness of using POSA for selection of the optimal coagulant and provides a systematical decision making framework with several characteristics: (i) the importance of different performances of treatment systems can be evaluated using multiple criteria – both quantitative and qualitative – rather than profitability alone, (ii) the use of ratings makes it possible to evaluate the applicability of different alternatives for the end user, (iii) the use of POSA method provides an effective way of documenting the management process and points out the importance of two indicators (TFS and COD), and (iv) the proposed approach forms the basis for a continuous process of planning and managing technology selection, so that the priorities of the treatment processes can easily be modified and updated.

Overall, based on jar tests and POSA models, 100 ppm Ferric Chloride is the best system among all systems studied which shows the highest efficiency in terms of reduction of COD, TSS, TVS and TFS. This study evidenced once again that coagulation process can assure the limits of organics for municipal wastewater treatment plants providing high removal efficiency using relatively low level of Alum or Ferric Chloride if the process is well optimized and operated.

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