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ON THE USE OF LINEAR MODELS FOR THE DESIGN OF WATER UTILIZATION SYSTEMS IN PROCESS PLANTS WITH A SINGLE CONTAMINANT

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This paper addresses the optimum design of water utilization systems when a single contaminant is present. The application of the necessary conditions of optimality allows an LP or MILP formulation depending on the objective function of choice. Several examples are presented to illustrate the proposed methodology and to point out that several alternative solutions are available.

Keywords: water reuse; waste water minimization.

INTRODUCTION

Wastewater streams in refineries and chemical plants (paper mills, semiconductor washing, etc) contain several contaminants (suspended solids, phenols, sulfides, ammonia, benzene, oil, etc.) that can create an environmental pollution problem.

Belhateche[1] offers a complete discussion of end-of-pipe wastewater clean-up solutions. However, these do not reduce water, they just clean it more efficiently. Attack the problem at its roots, i.e. generation of pollutants, process simulation has been proposed (Sowa[2], Hilaly and Sikdar[3]). However, for many processes the reduction of the generation of many pollutants is not possible. The concept of reusing water has received the name of Water/Wastewater Allocation Planning (WAP) problem. The search for optimal wastewater reuse solutions was addressed by industry itself more than twenty years ago (Carnes *et al.*[4], Skylov and Stenzel[5], Hospondarec and Thompson[6], Mishra *et al.*[7], Sane and Atkins[8]). Takama *et al.*[9] used mathematical programming to solve a refinery example. A superstructure of all water using operations and cleanup processes was set up and an optimization was then carried out to reduce the system structure by removing irrelevant and uneconomical connections. The model is transformed into a series of problems without inequality constraints by using a penalty function and finally solving it using the Complex method. Wang and Smith[10] presented a method based on targeting. The basic concept underlying the methodology is mass exchanger network (MEN) technology, which was in turn first proposed by El-Halwagi and Manousiouthakis[11] and was applied to the removal of phenol from refinery wastewater (El-Halwagi and Manousiouthakis[12]). Wang and Smith[10] proposed a methodology that can effectively pick optimal reuse solutions. They also explored options of regenerating wastewater even when the pollutant level has not reach end-of-pipe conditions, or has not be reused throughout the entire process. The authors approach the WAP using targeting graphical representations and techniques on superstructures of alternative designs. The targeting graphical method exploits the idea of plotting the cumulative exchanged mass vs. composition for a set of rich and lean streams, a concept first presented by El-Halwagi and Manousiouthakis[11] for synthesizing Mass Exchanger Networks. Once the target flowrate is obtained, the authors proposed a matching method similar to the procedure followed when constructing Heat Exchanger Networks (Linnhoff and Hindmarsh[13]). Finally, the loops created by the matching procedure are removed to render a realistic network of the water-using operations.

Additional work has been done to solve the problem when multiple contaminants are present. Doyle and Smith[14] presented a superstructure optimization, which is solved by a special iterative procedure. Kuo and Smith[15] approach the WAP problem in combination with the Wastewater Clean-up Allocation Planning (WCAP) problem and use graphical representations and techniques on superstructures of alternative designs. Alva-Argaez *et al.*[16] presented a solution approach for multicomponent systems based on mathematical programming. Quite recently, Huang *et al.*[17] presented an alternative NLP formulation. Finally, Alva-Argaez *et al.*[18] showed that using certain simplifying assumption, a transshipment model can be formulated.

The above work constitutes important attempts to solve the single and multiple contaminant WAP problem. However, each of these methods has either numerical or some other problems that prevent them from being robust. In this work, we propose an LP formulation for the optimal solution of the single contaminant WAP problem and a series of MILP problems to design different network alternatives. The new approach is based on previously developed (Savelski and Bagajewicz[19,20]) necessary conditions of optimality. The paper is restricted to problems with single contaminants. Despite this limitation, there are industrial cases where the method is applicable. The pulp and paper industry uses massive amounts of water and there the concern is on suspended solids only. The painting industry as well as the steel and semiconductor industries also care about total levels of a single contaminant.

This paper also shows explicitly that several alternative solutions of the problem exist, a point that has been made in the past, and a procedure is offered to pick the most convenient one, which previous techniques, notwithstanding their intrinsic value, have not been able to offer. Finally, a very important feature, which is the partial regeneration of wastewater to further reduce the freshwater use, is considered.

PROBLEM STATEMENT

Given a set of water-using/water-disposing processes, it is desired to determine a network of interconnections of water streams among the processes so that the overall fresh water consumption is minimized while the processes receive water of adequate quality. This is what is referred to as the Water/Wastewater Allocation Planning (WAP) problem. A more stringent-version of this problem was presented by Takama *et al.*[9] and later used by Wang and Smith[10]. In this version, limits on inlet and an outlet concentration of pollutant are imposed a-priori on each process and a fixed load of contaminants is used. These inlet and outlet concentration limits account for corrosion, fouling, maximum solubility, etc. Necessary conditions of optimality for this problem were first presented were presented by Savelski and Bagajewicz[19] and later proved rigorously by Savelski and Bagajewicz[20].

Targeting

The Water Allocation Planning problem can be modelled as follows:

$$
\begin{aligned}
& Min \sum_j F_j^w \\
& \text{s.t.} \\
& F_j^w + \sum_{i \in P_j} F_{i,j} - \sum_{k \in R_j} F_{j,k} - F_{j,out} = 0 && \forall j \in \boldsymbol{N} \\
& F_h^w - \frac{L_h}{C_{h,out}^{\max}} = 0 && \forall h \in \boldsymbol{H} \\
& \sum_{i \in P_j} F_{i,j}(C_{i,out} - C_{j,in}) - F_j^w C_{j,in} = 0 && \forall j \in \overline{\boldsymbol{H}} \\
& \sum_{i \in P_j} F_{i,j}(C_{i,out} - C_{j,out}) - F_j^w C_{j,out} + L_j = 0 && \forall j \in \overline{\boldsymbol{H}} \\
& C_{j,in} \le C_{j,in}^{\max} && \forall j \in \overline{\boldsymbol{H}} \\
& C_{j,out} \le C_{j,out}^{\max} && \forall i \in \boldsymbol{N} \\
& F_j^w, F_{i,j}, F_{j,out} \ge 0 && \forall i, \forall j \in \boldsymbol{N}
\end{aligned}
\tag{1}
$$

The problem has bilinear terms in those equality constraints where flowrate and concentration are simultaneously present. However, these bilinearities can be eliminated using the necessary condition of maximum outlet concentrations, that is, setting outlet concentrations to their maximum values.

The constraints can now be combined as follows:

$$
\left.
\begin{aligned}
\sum_{i \in P_j} F_{i,j}(C_{i,out}^{\max} - C_{j,in}) - F_j^w C_{j,in} = 0 \\
C_{j,in} \le C_{j,in}^{\max}
\end{aligned}
\right\} \Longleftrightarrow
\left\{ \sum_{i \in P_j} F_{i,j}(C_{i,out}^{\max} - C_{j,in}^{\max}) - F_j^w C_{j,in}^{\max} \le 0 \quad \forall j \in \overline{\boldsymbol{H}} \right.
\tag{2}
$$

The resulting problem is:

$$
\begin{aligned}
& Min \sum_j F_j^w \\
& \text{s.t.} \\
& F_j^w + \sum_{i \in P_j} F_{i,j} - \sum_{k \in R_j} F_{j,k} - F_{j,out} = 0 && \forall j \in \boldsymbol{N} \\
& F_h^w - \frac{L_h}{C_{h,out}^{\max}} = 0 && \forall h \in \boldsymbol{H} \\
& \sum_{i \in P_j} F_{i,j}(C_{i,out}^{\max} - C_{j,in}^{\max}) - F_j^w C_{j,in}^{\max} \le 0 && \forall j \in \overline{\boldsymbol{H}} \\
& \sum_{i \in P_j} F_{i,j}(C_{i,out}^{\max} - C_{j,out}^{\max}) - F_j^w C_{j,out}^{\max} + L_j = 0 && \forall j \in \overline{\boldsymbol{H}} \\
& F_j^w, F_{i,j}, F_{j,out} \ge 0 && \forall j \in \boldsymbol{N}
\end{aligned}
\tag{3}
$$

This problem is linear. Consequently, the optimal water flowrate and a feasible realizing network are both simultaneously obtained. Furthermore, when setting up the problem, the number of variables can be reduced by not including non-monotone connections as suggested by the monotonicity necessary condition.

Example 1: Targeting the Fresh Water Usage

Consider the following problem, which involves ten water-using processes. Table 1 shows the limiting data.

In this problem, after monotonicity is applied, the number of possible interconnections reduces from 72 to 40. Table 2 shows the results obtained using LINDO and Figure 1 shows the resulting realizing network. The fresh water savings are over 34%. It is worth noting that the optimization may render a network that, although feasible, could require too many interconnections. Moreover, some of them may be even impractical. For example, the flowrate from process 9 to 7 is only $1.2952\,\text{ton}\,\text{h}^{-1}$, which requires a 15-mm ID pipe, if an economical velocity of $2.0\,\text{m}\,\text{s}^{-1}$ is assumed. In this example, there are a total

Table 1. Limiting data for Example 1.

Process number	Mass load of contaminant (kg/h)	$C_{in}^{\max}$ (ppm)	$C_{out}^{\max}$ (ppm)	Minimum fresh water flowrate without reuse (ton/h)
1	2.0	25	80	25.0
2	2.88	25	90	32.0
3	4.0	25	200	20.0
4	3.0	50	100	30.0
5	30.0	50	800	37.5
6	5.0	400	800	6.25
7	2.0	400	600	3.3333
8	1.0	0	100	10.0
9	20.0	50	300	66.6667
10	6.5	150	300	21.6667
Total minimum flowrate (ton/h)				252.4167

Table 2. Solution of Example 1.

Process number	$F_{i,j}$ (ton/h)	Minimum fresh water flowrate with reuse (ton/h)	Wastewater flowrate (ton/h)
1	0.0	25.0	0.0
2	0.0	32.0	0.0
3	$F_{1,3}$ = 7.14286	15.7143	0.0
4	$F_{1,4}$ = 17.8571	26.4286	0.0
5	$F_{4,5}$ = 20.0	20.0	40.0
6	$F_{7,6}$ = 4.16667 $F_{10,6}$ = 8.33333	0.0	12.5
7	$F_{3,7}$ = 4.02857 $F_{9,7}$ = 1.29524	0.0	1.15714
8	0.0	10.0	0.0
9	$F_{2,9}$ = 32.0 $F_{4,9}$ = 11.20	36.80	78.7048
10	$F_{3,10}$ = 18.8286 $F_{4,10}$ = 13.0857 $F_{8,10}$ = 10.0	0.0	33.5710
Total minimum freshwater usage (ton/h)		165.9424	

of 12 interconnections among processes and 24 when counting connections from the fresh water source to the processes and connections from the units to the wastewater treatment plant.

ALTERNATIVE SOLUTIONS

Once the target is obtained, different network alternatives can be sought. To do that, different objective functions are proposed and the minimum fresh water usage is added as a constraint. These different cases are considered below.

CASE 1: MINIMUM NUMBER OF INTERCONNECTIONS

The flowsheet obtained in Example 1 has 12 interconnections ($F_{i,j}$), 7 connections from the fresh water source to the FWU (F_j^w) and 5 connections between terminal processes and the wastewater treatment plant ($F_{j,out}$).

In a grassroots design, it would be desirable to minimize the number of interconnections as a means of reducing piping costs as well as instrumentation. Therefore, a new model can be proposed where the objective function is no longer the fresh water consumption but the summation of the number of possible connections among processes. The definitions of head process, monotonicity and maximum outlet concentrations are also applied to reduce, the number of variables. The formulation of the problem follows:

$$
\begin{aligned}
& Min\left(\sum_{i,j} Y_{i,j} + \sum_{w,j} Y_{w,j} + \sum_{j,o} Y_{j,o}\right) \\
& \text{s.t.} \\
& F_j^w + \sum_{i \in P_j} F_{i,j} + \sum_{k \in R_j} F_{j,k} - F_{j,out} = 0 \qquad \forall j \in \boldsymbol{N} \\
& \sum_j F_j^w = \alpha \\
& F_h^w - \frac{L_h}{C_{h,out}^{\max}} = 0 \\
& \sum_{i \in P_j} \boldsymbol{F}_{i,j}(C_{i,out}^{\max} - C_{j,in}^{\max}) - F_j^w C_{j,in}^{\max} \le 0 \qquad \forall j \in \overline{\boldsymbol{H}} \\
& \sum_{i \in P_j} \boldsymbol{F}_{i,j}(C_{i,out}^{\max} - C_{j,out}^{\max}) - F_j^w C_{j,out}^{\max} + L_j = 0 \qquad \forall j \in \overline{\boldsymbol{H}} \\
& F_{i,j} - UY_{i,j} \le 0 \qquad \forall j \in \overline{\boldsymbol{H}} \\
& F_{\mathrm{j}}^{\mathrm{w}} - UY_{w,j} \le 0 \qquad \forall j \in \overline{\boldsymbol{H}} \\
& F_{j,out} - UY_{j,o} \le 0 \qquad \forall j \in \boldsymbol{N} \\
& Y_{i,j};\ Y_{w,j};\ Y_{j,o} \in \{0, 1\} \\
& F_j^w, F_{i,j}, F_{j,out} \ge 0 \qquad \forall j \in \boldsymbol{N}
\end{aligned}
\tag{4}
$$

Two new constraints have been added to the problem:

$$\sum_j F_j^w = \alpha \tag{5}$$

where α is the optimal fresh water flowrate obtained solving the targeting problem, and

$$F_{i,j} - UY_{i,j} \le 0 \quad \forall j \in \overline{\boldsymbol{H}},\ \ i \in \boldsymbol{P}_j \tag{6}$$

which relate the inter-processes flowrates with the integer variables. In these constraints, number larger than any feasible value of $F_{i,j}(\forall i, j)$. For this problem, the value of U was chosen to be larger than the targeted fresh water flowrate, α. The above sequence of models follows in spirit the same strategy as the transshipment model introduced by Papoulias and Grossman[21]. After a targeting phase where the total water consumption is obtained, a minimization of the number of connections is proposed.

Using this model, three different examples were run. The first one minimizes the number interconnections among processes, $F_{i,j}$, the second minimizes the connections between the fresh water source and the processes. Finally, all type of connections including those between the processes and the wastewater treatment plant is minimized.

Example 2: Minimization of the Number of Interconnection Among Processes

Consider the same limiting data as in Example 1 and its corresponding target value $\alpha = 165.9424\ \text{ton h}^{-1}$. The problem formulation contains only the integer variables $Y_{i,j}$. The solution yields 9 interconnections instead of the 12 previously obtained for Example 1. Since the minimum fresh water usage is the same, this solution network constitutes a better alternative than the one of Example 1. Figure 2 and Table 3 show the new design network and the results, respectively. The solution was obtained using LINDO/CIPLEX.

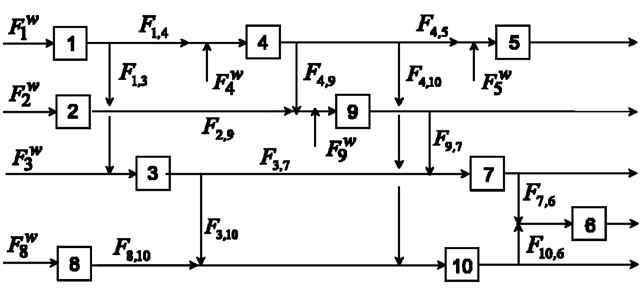

Figure 1. Solution network for LP formulation of Example 1.

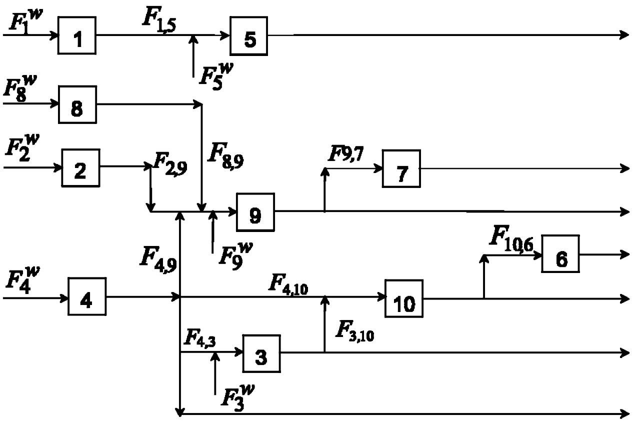

Figure 2. Minimum number of interconnections.

Example 3: Minimization of the Total Number of Connection

In this case, all three types of connections are considered. The minimum number of connections, for the given minimum fresh water target, is 22. Table 4 shows that, upon optimization, the number of total interconnections was reduced by one. Figure 3 shows the final network for this case. The purpose of solving the same example using different objective functions is to show how many different alternative flowsheets can be obtained for the same optimal condition of minimum fresh water usage. When designing, the awareness of these alternatives is highly advantageous. Another important feature to observe from these solutions is that, without imposing flowrate constraints the last optimization removed all negligible streams from the flowsheet. The solution networks for Examples 2 and 3 have flowrates lower than 1.5 tons h^{-1}, which can be considered uneconomical. None of them appears in the last solution network. In all the following examples, GAMS has been used as the equation modelling system and GAMS/CIPLEX as the MILP solver.

CASE 2: MINIMUM FIXED COSTS

Reducing the number of interconnections is relevant for a cost-effective design. However, the networks obtained in previous examples do not guarantee minimum cost but only minimum number of connections. Since not all interconnections will require the same fixed capital investment, it seems most appropriate to minimize the fixed annualized cost. For this example, without loss of generality, a series of single cost values representing each interconnection is given as shown in Table 5. These values can be understood as the corresponding calculations of the annualized installed costs of piping, valves and pumps. The new objective function can be written as:

$$\sum_{i,j} c_{i,j} Y_{i,j} \tag{7}$$

The rest of model (3) remains the same. The integer variables chosen are the interconnections among processes and those between processes and the wastewater treatment plant. The connections from the fresh water source cannot be different from those previously obtained hence; they have been excluded from the optimization. The optimal cost is \$53.159; the resulting network is shown in Figure 4 and Table 6.

Table 3. Solution of Example 2.

Process number	$F_{i,j}$ (ton/h)	Minimum fresh water flowrate with reuse (ton/h)	Wastewater flowrate (ton/h)
1	0.0	25.0	0.0
2	0.0	32.0	0.0
3	$F_{4,3}$ = 5.7143	17.1429	1.1905
4	0.0	30.0	1.4190
5	$F_{1,5}$ = 25.0	15.0	40.0
6	$F_{10,6}$ = 10.0	0.0	10.0
7	$F_{9,7}$ = 6.6667	0.0	6.6667
8	0.0	10.0	0.0
9	$F_{2,9}$ = 32.0	36.80	73.3333
	$F_{4,9}$ = 1.20		
	$F_{8,9}$ = 10.0		
10	$F_{3,10}$ = 21.6667	0.0	33.3333
	$F_{4,10}$ = 21.6667		
Connections	9	7	7

Table 4. Solution of Example 3.

Process number	$F_{i,j}$ (ton/h)	Minimum fresh water flowrate with reuse (ton/h)	Wastewater flowrate (ton/h)
1	0.0	25.0	0.0
2	0.0	32.0	0.0
3	$F_{2,3}$ = 6.349	16.508	0.0
4	0.0	30.0	0.0
5	$F_{1,5}$ = 25.0	15.0	40.0
6	$F_{3,6}$ = 4.029	0.0	9.194
	$F_{9,6}$ = 5.166		
7	$F_{10,7}$ = 6.667	0.0	6.667
8	0.0	10.0	0.0
9	$F_{2,9}$ = 25.651	37.435	74.834
	$F_{4,9}$ = 6.914		
	$F_{8,9}$ = 10.0		
10	$F_{3,10}$ = 18.829	0.0	35.248
	$F_{4,10}$ = 23.086		
Connections	10	7	5

CASE 3: FORBIDDEN/COMPULSORY MATCHES

It is quite often found that connections between certain processes are not allowed or must be imposed due to design or retrofit strategies. For example: heating and/ or cooling limitations may render certain connections beneficial or undesirable; low flowrate interconnections, typically below 2.0 $m^3 h^{-1}$, may be discarded due to economical or controllability reasons; finally, distance

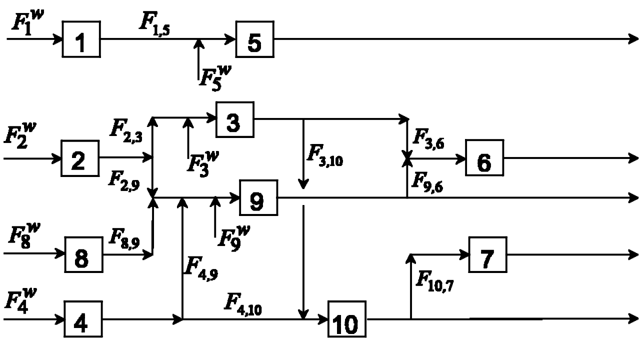

Figure 3. Network solution to Example 3.

Table 5. Cost data in $ per year.

Process	1	2	3	4	5	6	7	8	9	10	WWT
1	–	2.419	2.981	3.169	3.544	3.544	3.544	–	2.981	2.794	5.419
2	–	–	2.794	2.981	3.544	3.544	3.544	–	3.169	2.981	5.419
3	–	–	–	–	2.981	3.169	3.544	–	3.544	3.544	4.669
4	–	–	2.419	–	2.794	2.981	3.544	–	3.544	3.544	4.669
5	–	–	–	–	–	–	–	–	–	–	3.919
6	–	–	–	–	–	–	–	–	–	–	3.919
7	–	–	–	–	2.981	2.794	–	–	–	–	3.919
8	–	–	3.544	–	3.169	2.981	2.419	–	2.794	2.981	3.919
9	–	–	–	–	3.544	3.544	2.981	–	–	–	4.669
10	–	–	–	–	3.544	3.544	3.169	–	–	–	4.669

Table 6. Solution of fixed cost problem.

Process number	$F_{i,j}$ (ton/h)	Minimum fresh water flowrate with reuse (ton/h)	Wastewater flowrate (ton/h)
1	0.0	25.0	0.0
2	0.0	32.0	0.0
3	$F_{4,3}$ = 5.714	17.143	0.0
4	$F_{2,4}$ = 32.0	26.80	0.0
5	$F_{1,5}$ = 25.0	15.0	40.0
6	$F_{3,6}$ = 4.028	0.0	9.194
	$F_{10,6}$ = 5.166		
7	$F_{9,7}$ = 6.667	0.0	6.667
8	0.0	10.0	0.0
9	$F_{4,9}$ = 30.0	40.0	73.333
	$F_{8,9}$ = 10.0		
10	$F_{3,10}$ = 18.829	0.0	36.749
	$F_{4,10}$ = 23.086		
Connections	10	7	5

and space limitations may become decision variables as well. When imposing such restrictions to any of the previous cases we can expect either feasible or infeasible solutions. The infeasibility may arise because no network can be found for the fresh water flowrate fixed at its minimum target.

Two example problems are presented below. Example 5 is a variance of Example 4 where a forbidden match between processes 2 and 3 ($Y_{2,3} = 0$), and a compulsory match between processes 4 and 3 ($Y_{4,3} = 1$) are imposed. Figure 5 and Table 7 show the new realizing network and the obtained flowrates, respectively. Example 6 illustrates the increase in fixed costs when no process is allowed to send wastewater to more than two processes (including wastewater treatment units). Reducing the number of stream splitting makes the system easier to control. This is accomplished by adding the constraint $\sum_j Y_{i,j} \le 2 \forall i \in N$. The new fixed cost is $53.534 that is, only 0.7% higher than the optimal. Figure 6 and Table 8 depict the corresponding network and flowrates, respectively.

USE OF DEGENERACY

Although the previous examples provide different alternatives, they are all built based on the necessary conditions of maximum outlet concentrations. There are solutions to the problem that satisfy the target minimum fresh water usage, but have outlet concentrations lower than their maximum. These solutions feature the same total fresh-water intake. We will use the existence of these degenerate solutions to reduce the number of connections even further.

Consider the example problem proposed by Olesen and Polley[22]. The limiting data is shown in Table 9. The authors solved this problem by inspection and proposed the network solution illustrated in Figure 7. The reported fresh water target is 157.14 ton h^{-1}.

From Figure 7 it can be observed that process 1 is consuming 15.0 ton h^{-1} more fresh water than the minimum required to pickup its entire load. Consequently, its outlet concentration is 50 ppm instead of its possible maximum of 80 ppm. Therefore, Olesen and Polley[22] solution can be understood as a degenerate solution. Its equivalent is the same network where process 1 reaches its maximum outlet concentration and $F_1^w = 25.0$, $F_5^w = 15.0$, with the rest of the values being the same. The wastewater produced by process 1 would be at 80 ppm.

Figure 8 shows a design network obtained when applying the LP targeting model. One can explore degenerate solutions where the fresh water intake of either process 3 or 4 is eliminated. Consider we want to eliminate the fresh water connection to process 3. In

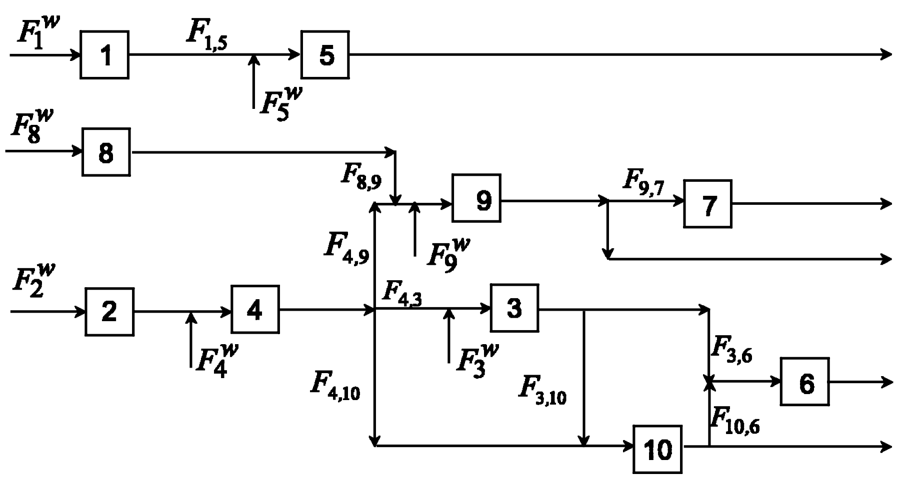

Figure 4. Solution network for minimum fixed cost.

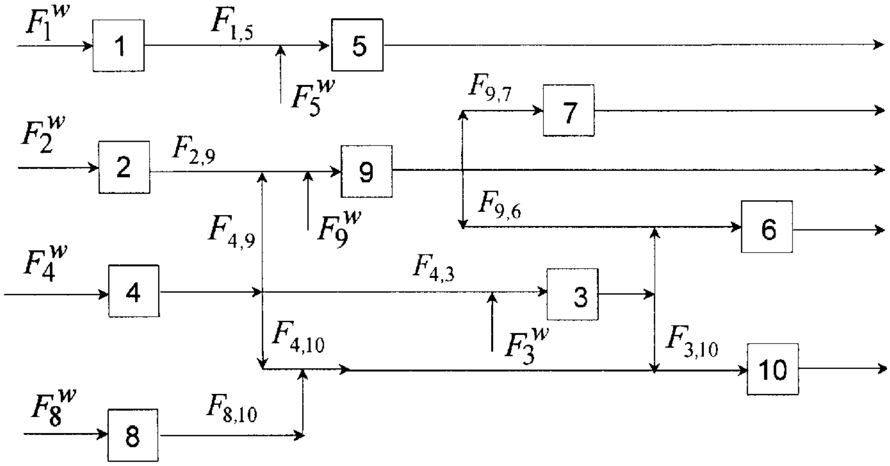

Figure 5. Solution network for Example 5.

this case, wastewater available at 25 ppm, which is the maximum inlet concentration allowed for process 3, is needed. To do that, the fresh water intake to process 1 should be increased to 80.0 ton h $^{-1}$. The minimum necessary reuse between units 1 and 3 can be calculated to be 22.8571 ton h $^{-1}$. The remaining wastewater at process 1, 57.1429 ton h $^{-1}$ can then be sent to process 4 reducing its fresh water needs to 7.1428 ton h $^{-1}$. Thus, a new equivalent and feasible network can be obtained by increasing F_1^w, reducing F_4^w and F_3^w until the last one is reduced to zero.

One can always explore degenerate alternatives once the LP model network has been obtained. The procedure involves increasing the freshwater of a process and reducing the freshwater of the receiver. As stated by the necessary conditions, these changes will not alter the overall freshwater consumption of the network, so they present no incentive in this regard. In addition, there is a disadvantage: the amount of water circulating through the processes for which the freshwater is increased also increases. This leads sometimes to a higher volume for this operation and consequently a higher cost. Since equipment cost is not considered in this paper, this point is left to be discussed in future work. Finally, they are infinite in number so enumeration is futile. One is therefore left with only one reason to use such degenerate solutions: they can be used to eliminate freshwater piping. As it was pointed out above in the example, the procedure to perform such trade-off studies is simple: Pick a process and a set of receivers. Increase the freshwater in the process and determine the corresponding reduction in each of the receivers. No mathematical programming model can make this simple procedure more efficient.

In making the above remarks, one should remember that they are only true if the assumptions of the problem hold. In particular, the assumption that the processes behave the same (same load of pollutant being picked up) for different water flowrates should be taken into account. This has been pointed out as a limitation of all this line of work by Bagajewicz[23], who suggested that the relationship between the outlet concentration and the load should be considered in more detailed modelling of this problem. Such modelling is non-linear and the aforementioned necessary conditions of optimality no longer hold. Thus, a trade-off exists between the ability of easily generating alternative solutions offered by the models in this paper and the incremental model accuracy that more elaborate models can offer at the expense of the known numerical difficulties this non-convex problems exhibit. Thus, the solutions presented in this paper may serve as a good starting point to solve the latter models.

REGENERATION

We now consider the issue of regeneration of wastewater so that it could be reused in processes. If one considers such treatment, the revised problem can be formulated as follows.

$$
\begin{aligned}
& Min \sum_j F_j^w \\
& \text{s.t.} \\
& F_j^w + \sum_{i \in P_j} F_{i,j} + \sum_{t \in T} F_{j,t} - \sum_{k \in R_j} F_{j,k} - \sum_{t \in T} F_{t,j} \\
& \qquad - F_{j,out} = 0 && \forall j \in \boldsymbol{N} \\
& F_h^w - \frac{L_h}{C_{h,out}^{\max}} = 0 && \forall h \in \boldsymbol{H} \\
& \sum_{i \in P_j} F_{i,j}(C_{i,out}^{\max} - C_{j,in}^{\max}) + \sum_{t \in T} F_{t,j}(C_{t,out}^{T} - C_{j,in}^{\max}) \\
& \qquad - F_j^w C_{j,in}^{\max} \le 0 && \forall j \in \overline{\boldsymbol{H}} \\
& \sum_{i \in P_j} F_{i,j}(C_{i,out}^{\max} - C_{j,out}^{\max}) + \sum_{t \in T} F_{t,j}(C_{t,out}^{T} - C_{j,out}^{\max}) \\
& \qquad - F_j^w C_{j,out}^{\max} + L_j = 0 && \forall j \in \overline{\boldsymbol{H}} \\
& \sum_{i \in N} F_{i,t} - \sum_{i \in N} F_{t,i} - F_{t,out} = 0 && \forall t \in \boldsymbol{T} \\
& F_j^w, F_{i,j}, F_{i,t}, F_{t,j} F_{j,out}, F_{t,out} \ge 0 && \forall i\ \forall j \in \boldsymbol{N}, \forall t \in T
\end{aligned} \tag{9}
$$

This problem is not linear, as the outlet concentrations of the treatment units are not known. There are, however, lower bounds of this concentration $C_{t,out}^{T^*}$ given by the treatment technology limitations. We now prove that this is actually the value that should be used in order to minimize the freshwater used.

Theorem: *If a solution to the WAP is optimal and a process exists that receives treated water as well as freshwater, then*

Table 7. Solution of compulsory matches, Example 5.

Process number	$F_{i,j}$ (ton/h)	Minimum fresh water flowrate with reuse (ton/h)	Wastewater flowrate (ton/h)
1	0.0	25.0	0.0
2	0.0	32.0	0.0
3	$F_{4,3}$ = 5.714	17.143	0.0
4	0.0	30.0	0.0
5	$F_{1,5}$ = 25.0	15.0	40.0
6	$F_{3,6}$ = 4.028	0.0	9.194
	$F_{9,6}$ = 5.166		
7	$F_{9,7}$ = 6.667	0.0	6.667
8	0.0	10.0	0.0
9	$F_{2,9}$ = 32.0	36.80	68.168
	$F_{4,9}$ = 11.20		
10	$F_{3,10}$ = 18.829	0.0	41.914
	$F_{4,10}$ = 23.086		
	$F_{8,10}$ = 10.0		
Connections	10	7	5

the outlet composition of the treatment should be the minimum possible, or this last condition is degenerate.

In other words, given a process j that receives treated water, that is $F_{t,j} > 0$, then $C^{T}_{t,out} = C^{T^*}_{t,out}$.

Proof: The proof is by contradiction:
Suppose that $C^{T}_{t,out} > C^{T^*}_{t,out}$. From the component mass balance over a process j that receives treated water, an expression of the freshwater water flow F^{w}_{j} follows:

$$F^{w}_{j} = \frac{\sum_i F_{i,j} C^{\max}_{i,out} + F_{t,j} C^{T}_{t,out} + L_j}{C^{\max}_{j,out}} - \left(\sum_i F_{i,j} + F_{k,j} \right) \tag{10}$$

For the case of minimum outlet concentration in the regeneration unit, we obtain:

$$F^{w^*}_{j} = \frac{\sum_i F_{i,j} C^{\max}_{i,out} + F_{t,j} C^{T^*}_{t,out} + L_j}{C^{\max}_{j,out}} - \left(\sum_i F_{i,j} + F_{k,j} \right) \tag{11}$$

Subtracting Equation (7) from (8) and considering that we maintain all the rest of the flows and concentrations unaltered, then:

$$F^{w^*}_{j} - F^{w}_{j} = \frac{F_{k,j}(C^{T^*}_{t,out} - C^{T}_{t,out})}{C^{\max}_{j,out}} \tag{12}$$

which is negative. If process $F_{j,out} \geq F^{w}_{j} - F^{w^*}_{j}$, then the proof is complete. However, if $F_{j,out} < F^{w}_{j} - F^{w^*}_{j}$, then there is a smaller amount of water available to be sent to the set of receivers downstream. In such case, the new value of the flowrate sent to final treatment is: $F^{*}_{j,out} = 0$, and the new flowrate from process j to the receivers is $F^{*}_{j,R_j} = F_{j,R_j} + (F^{w^*}_{j} - F^{w}_{j}) + F_{j,out}$, which is smaller than F_{j,R_j}. We now consider the adjustment of the freshwater on the receivers so that the total flowrate to the receivers is the same. Thus:

$$F_{j,R_j} + F^{w}_{R_j} + F_{P_j,R_j} = F^{*}_{j,R_j} + F^{w^*}_{R_j} + F_{P_j,R_j} \tag{13}$$

From these two equalities we obtain:

$$F^{w^*}_{R_j} - F^{w}_{R_j} = F_{j,R_j} - F^{*}_{j,R_j} = (F^{w}_{j} - F^{w^*}_{j}) - F_{j,out} \tag{14}$$

Thus the total change in freshwater intake is:

$$\Delta F^{w} = (F^{w^*}_{j} - F^{w}_{j}) + (F^{w^*}_{R_j} - F^{w}_{R_j}) = -F_{j,out} \tag{15}$$

which is negative. Finally, one needs to verify that the inlet concentration of the receivers has not increased. Thus,

$$C^{*}_{R_j,in} = \frac{F^{*}_{j,R_j} C_{j,out} + F_{P_j,R_j} C_{P_j,out}}{F^{*}_{j,R_j} + F^{w^*}_{R_j} + F_{P_j,R_j}} \tag{16}$$

But $F^{*}_{j,R_j} + F^{w^*}_{R_j} + F_{P_j,R_j} = F_{j,R_j} + F^{w}_{R_j} + F_{P_jR_j}$. Therefore, since $F^{*}_{j,R_j} < F_{j,R_j}$, we have:

$$C^{*}_{R_j,in} = \frac{F^{*}_{j,R_j} C_{j,out} + F_{P_j,R_j} C_{P_j,out}}{F_{j,R_j} + F^{w}_{R_j} + F_{P_j,R_j}} < \frac{F_{j,R_j} C_{j,out} + F_{P_j,R_j} C_{P_j,out}}{F_{j,R_j} + F^{w}_{R_j} + F_{P_j,R_j}} < C_{R_j,in} \tag{17}$$

Q.E.D.

When the regenerated water is sent to processes for which fresh water is not needed, then this has no influence in the objective function and the choice of outlet concentration is irrelevant. As a consequence of the above theorem and the previous comment, one is justified in using $C^{T^*}_{t,out}$ instead of $C^{T}_{t,out}$, substitution that makes the problem linear. However, if one wants to also reduce the amount of water sent through the treatment process, it is trivial to see that the amount of treated water reduces as the outlet concentration of the treatment process is reduced. Another case that needs to be addressed is when no freshwater is used in a process but water from the treatment unit is used. In this case, one should realize that:

- The maximum outlet concentration necessary condition holds when the freshwater used has some contaminant in it. This can be easily checked by revisiting the proofs presented by Savelski and Bagajewicz[20]. This makes the constraints in Equation (9) valid, even for the cases when freshwater is not used in a process.
- It is advantageous to continue using the minimum concentration possible at the outlet of the treatment. This is also straightforwardly proved if one considers the water from the treatment unit as another source of 'freshwater', even though it has some contaminant and its availability is constrained by how much water can be sent to this unit. Thus, for every unit that consumes treated water, but not freshwater, one can write:

$$F_{i,j} = \frac{\sum_i F_{i,j} C^{\max}_{i,out} + L_j - \left(\sum_i F_{i,j}\right) C^{\max}_{j,out}}{(C^{\max}_{j,out} - C^{T}_{t,out})} \tag{18}$$

which is minimum when $C^{T}_{t,out} = C^{T^*}_{t,out}$.

Finally, Equation (9) has no control over the amount of water sent to the treatment unit. Thus, to keep the amount of water going to treatment to a minimum, we proposed to solve (9) to obtain the target freshwater consumption (α). Afterwards, the solution to the problem with minimum

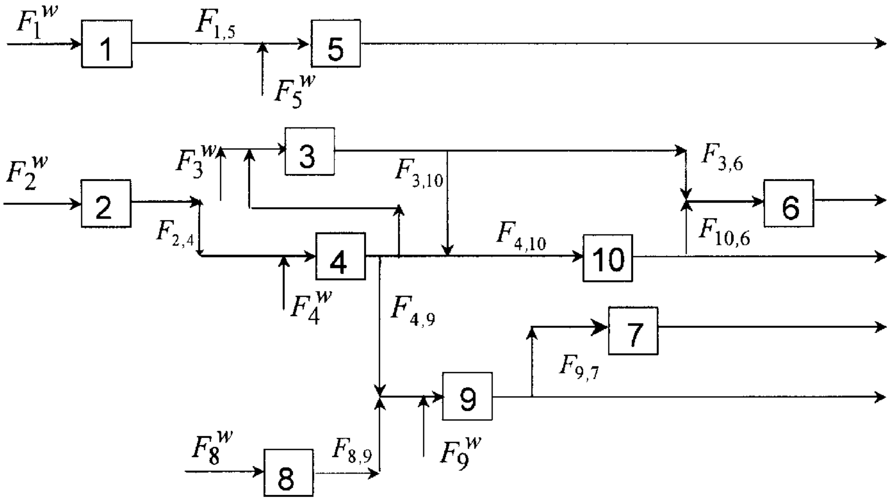

Figure 6. Solution network for Example 6.

Table 8. Solution of fixed cost with limited stream splitting.

Process number	$F_{i,j}$ (ton/h)	Minimum fresh water flowrate with reuse (ton/h)	Wastewater flowrate (ton/h)
1	0.0	25.0	0.0
2	0.0	32.0	0.0
3	$F_{4,3}$ = 5.714	17.143	0.0
4	$F_{2,4}$ = 32.0	26.80	0.0
5	$F_{1,5}$ = 25.0	15.0	40.0
6	$F_{3,6}$ = 4.028	0.0	9.194
	$F_{10,6}$ = 5.166		
7	$F_{9,7}$ = 6.667	0.0	6.667
8	0.0	10.0	0.0
9	$F_{4,9}$ = 30.0	40.0	73.333
	$F_{8,9}$ = 10.0		
10	$F_{3,10}$ = 18.829	0.0	36.749
	$F_{4,10}$ = 23.086		
Connections	10	7	5

Table 9. Limiting data from Olesen and Polley[22].

Process number	Mass load of contaminant (kg/h)	C_{in}^{max} (ppm)	C_{out}^{max} (ppm)
1	2.0	25	80
2	5.0	25	100
3	4.0	25	200
4	5.0	50	100
5	30.0	50	800
6	4.0	400	800

water running through the treatment unit is obtained by solving

$$\begin{aligned} &Min\left\{\sum_j F_{j,T}\right\} \\ &\text{s.t.} \\ &\sum_j F_j^w = \alpha \\ &\textit{All}\ \text{constraints of Equation (9)} \end{aligned} \tag{19}$$

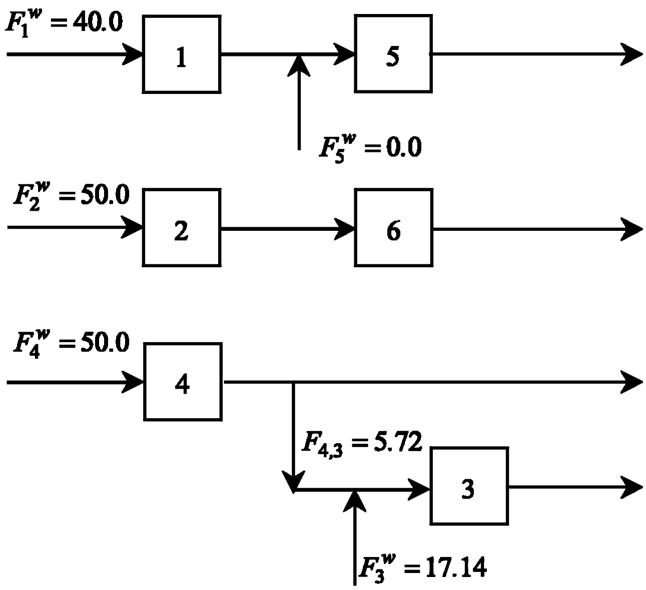

Figure 7. Design network proposed by Olesen and Polley[22].

Example 7: Regeneration

We now consider the use of one and two regeneration units in the case of Example 1, using a treatment outlet concentration of 5 ppm. When the above problem was solved the target obtained was 10 ton h $^{-1}$. The solution obtained using model (19) is shown in Figure 9 and Table 10.

The above solution contains recycles, that is, several processes are involved in a recycle containing the treatment unit. While all the flows can be, in principle, predicted by the graphical procedure developed by Wang and Smith[24], also well illustrated by Mann and Liu[25], this paper provides a straightforward and easy to implement procedure to obtain both the target flowrates and the corresponding network. Moreover, the present LP procedure can be extended to an MILP formulation to obtain minimum cost alternative solutions.

Interestingly, the above solution also suggests that given the appropriate availability of treatment units, the

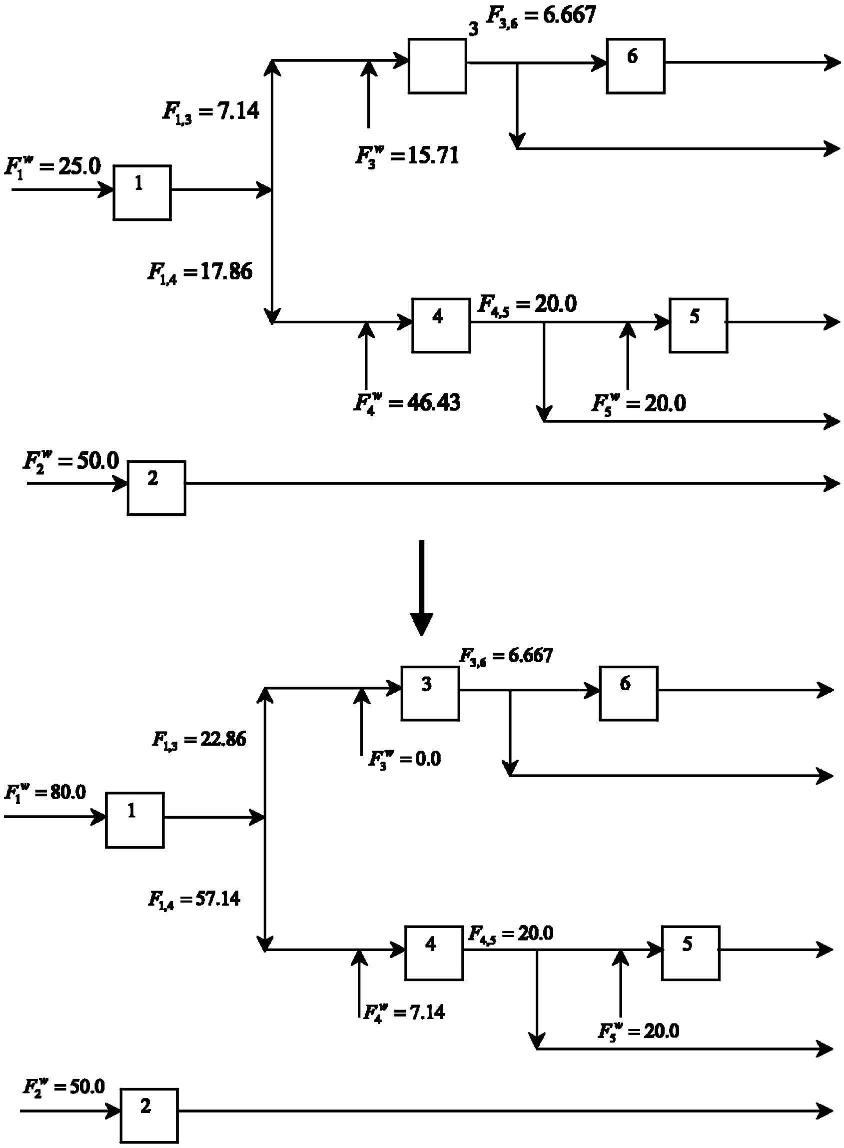

Figure 8. LP design network for the Olesen and Polley[22] problem with a degenerate alternative.

above procedure can be used to obtain zero-liquid discharge solutions. In fact, if process 8 has an inlet maximum concentration of 5 ppm or more, then the procedure renders zero freshwater consumption.

REGENERATION WITHOUT RECYCLES

To obtain solutions without recycles some binary variables and constraints need to be added. We introduce the binary variable Z_j to indicate whether water feeding unit j comes from the treatment unit or comes from units that are fed by the treatment unit. Therefore, to identify such situations one writes:

$$F_{t,j} - UZ_j \le 0 \quad \forall j \in N, \quad t \in T \tag{20}$$

$$Z_j \ge Y_{i,j} + Z_i - 1 \quad \forall i, \quad j \in N \tag{21}$$

Constraint (20) forces Z_j to be one when unit j is fed by treated water, while constraint (21) forces Z_j to be one when unit i is fed by treated water and unit i sends water to unit j, while is free to take any value otherwise. In addition, this does not prevent $F_{t,j}$ from being zero in all such cases. Finally, to eliminate the recycles, one must write:

$$Z_j + Y_{j,t} \le 1 \quad \forall j \in \boldsymbol{N}, \quad \forall t \in \boldsymbol{T} \tag{22}$$

which will prevent wastewater from being sent to the treatment if its precursors had been fed by treated water.

Example 8: Regeneration without Recycles

We revisit Example 1 using a treatment outlet concentration of 5 ppm and incorporating constraints (20) through (22). The target obtained was 91.123 ton h^{-1}, which compares very well with the solution obtained without regeneration. The solution obtained is shown in Figure 10 and Table 11.

The non-monotone connection between processes six and ten was set by the model in response to the maximum outlet concentration constraint of process ten. Its presence is actually irrelevant and has no effect in the removal of the

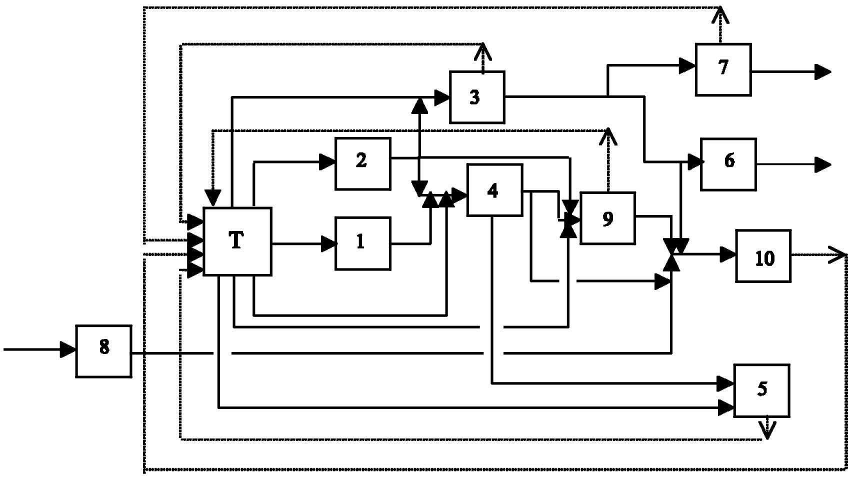

Figure 9. Solution of Example 1(with regeneration).

Table 10. Solution of Example 1.

Process number	$F_{i,j}$ (ton/h)	F_j^w (ton/h)	$F_{j,out}$ (ton/h)
1	$F_{1,4}$ = 26.667	0.0	0.0
2	$F_{2,3}$ = 5.378, $F_{2,4}$ = 8.235, $F_{2,9}$ = 20.269	0.0	0.0
3	$F_{3,6}$ = 8.333, $F_{3,7}$ = 5.000, $F_{3,10}$ = 2.414, $F_{3,T}$ = 7.110	0.0	0.0
4	$F_{4,5}$ = 18.947, $F_{4,9}$ = 19.759, $F_{4,10}$ = 21.293	0.0	0.0
5	$F_{5,T}$ = 40.00	0.0	0.0
6	0.0	0.0	8.333
7	$F_{7,T}$ = 3.333	0.0	1.667
8	$F_{8,10}$ = 10.00	10.00	0.0
9	$F_{9,10}$ = 9.627, $F_{9,T}$ = 70.373	0.0	0.0
10	$F_{10,T}$ = 43.333	0.0	0.0
T	$F_{T,1}$ = 26.667, $F_{T,2}$ = 33.882, $F_{T,3}$ = 17.479, $F_{T,4}$ = 25.096, $F_{T,5}$ = 21.053, $F_{T,9}$ = 39.972	0.0	0.0

Table 11. Solution of Example 1 (with regeneration and no recycles).

Process number	$F_{i,j}$ (ton/h)	F_j^w (ton/h)	$F_{j,out}$ (ton/h)
1	$F_{1,3}$ = 2.667, $F_{1,5}$ = 24.000	0.0	0.0
2	$F_{2,9}$ = 43.111	33.111	0.0
3	$F_{3,6}$ = 6.667, $F_{3,7}$ = 5.000, $F_{3,10}$ = 11.190	11.523	0.0
4	$F_{4,3}$ = 3.313, $F_{4,10}$ = 28.266	0.0	0.0
5	0.0	0.0	40.000
6	$F_{6,10}$ = 0.545	0.0	11.122
7	$F_{7,6}$ = 5.000	0.0	0.0
8	$F_{8,2}$ = 10.000	10.000	0.0
9	$F_{9,T}$ = 79.600	36.489	0.0
10	0.0	0.0	40.001
T	$F_{T,1}$ = 26.667, $F_{T,3}$ = 5.354, $F_{T,4}$ = 31.579, $F_{T,5}$ = 16.000	0.0	0.0

load in process ten. This process is a terminal process that utilizes neither fresh water nor treated wastewater.

One important thing that can be pointed out about the solution obtained is that the graphical methods rarely consider the use of freshwater in conjunction with regenerated water, which is what this last example portraits.

CONCLUSIONS

A method to solve the water allocation problem in process plants using linear programming has been presented. Examples have shown that the problem has several alternative optimal solutions, including degenerate cases where flows of water through the processes are larger than the minimum needed. Forbidden and compulsory water transfers from one process to another have also been solved. The findings of this paper have important consequences for the design and

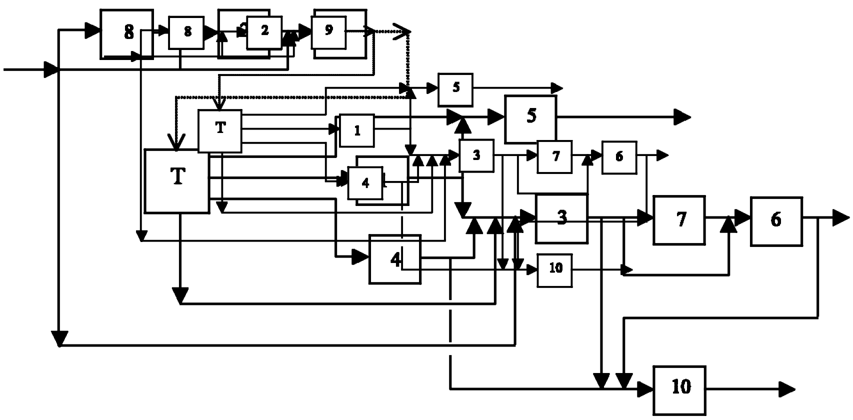

Figure 10. Solution of Example 1 (with regeneration and no recycles).

retrofit of water utilization systems, as it provides a tool to obtain several alternative solutions. Finally, the important problem of distributed treatment, which is known to reduce water usage even further, is considered. A new necessary condition of optimality is introduced and new LP/MILP formulations are introduced.

NOMENCLATURE

Sets

$\underline{\boldsymbol{N}}$	Set of all processes
$\overline{\boldsymbol{H}}$	Head processes
P_j	Precursors of process *j*
R_j	Receivers of process *j*
$\boldsymbol{T}$	Treatment processes

Variables

c	fixed cost of interconnection, $ per year
C	concentration of contaminant, ppm
F	water flowrate, ton h^{-1}
L	contaminant load, g h^{-1}

Subscripts

in:	at inlet
out:	at outlet or to final treatment
j:	process *j*

Superscripts

min:	minimum
max:	maximum
*:	additional sources
w:	fresh water
T:	treatment

REFERENCES

1. Belhateche, D. H., 1995, Choose appropriate wastewater treatment technologies, *Chem Eng Prog*, August, 32.
2. Sowa, C. J., 1994, Explore waste minimization via process simulation, *Chem Eng Prog*, November, 40.
3. Hilaly, A. K. and Sikdar, S. K. 1996, Process Simulation Tools for Pollution Prevention, *Chem Eng*, January, 98.
4. Carnes, B. A., Ford, D. L. and Brady, S. O., 1973, Treatment of Refinery Wastewaters for Reuse, *Nat Conf Complete Water Reuse*, Washington D.C.
5. Skylov, V. and Stenzel, R. A., 1974, Reuse of Wastewaters—Possibilities and Problems, *Proceeding of the Workshop (AIChE)*, 7.
6. Hospondarec, R. W. and Thompson, S. J., 1974, Oil-steam System for water Reuse, *Proceeding of the Workshop (AIChE)*, 7.
7. Mishra, P. N., Fan, L. T. and Erickson, L. E., 1975, Application of Mathematical Optimization Techniques in Computer Aided Design of Wastewater Treatment Systems, *Water-1974 (II), AIChE Symp. Series*, 71: 145.
8. Sane, M., Atkins, U. S. and partners, 1977, Industrial Water Management, *Int Chem Eng Symp Series*, 52.
9. Takama, N., Kuriyama, T., Shiroko, K. and Umeda, T., 1977, Optimal Water Allocation in a Petroleum Refinery, *PACHEC*, Denver.
10. Wang, Y. P. and Smith, R., 1994, Wastewater Minimization, *CES*, 49(7): 981.
11. El-Halwagi, M. M. and Manousiouthakis, V., 1989, Mass Exchanger Networks, *AIChE Journal*, 35(8): 1233.
12. El-Halwagi, M. M. and Manousiouthakis, V., 1990, Automatic Synthesis of Mass Exchanger Networks with Single Component Targets, *CES*, 45(9): 2813.
13. Linhoff, B. and Hindmarsh, E., 1983, The Pinch Design Method for Heat Exchanger Networks, *Chem Eng Sci*, 38(5): 745.
14. Doyle, S. J. and Smith R., 1997, Targeting water reuse with multiple contaminants, *Proc Safe Env Prot*, 75(3): 181–189.
15. Kuo, W.-C. J. and Smith, R., 1998, Designing for the Interactions between Water-Use and Effluent Treatment, *Trans IChemE Part A*, *Chem Eng Res Des*, 76: 287–301.
16. Alva-Argaez, A., Kokossis, A. C. and Smith, R., 1998, Automated Design of Industrial Water Networks, *AIChE Annual Meeting, paper*, 13f, Miami.
17. Huang, C.-H., Chang, C.-T., Ling, H.-C. and Chang, C.-C., 1999, A Mathematical Programming Model for Water Usage and Treatment Network Design, *Ind Eng Chem Res*, 38: 2666.
18. Alva-Argaez, A., Vallianoatos, A. and Kokossis, A., 1999, A Multi-Contaminant Transshipment Model for Mass Exchange Networks and Wastewater Minimization Problems, *Comput Chem Eng*, 23: 1439–1453.
19. Savelski, M. J. and Bagajewicz, M. J., 1999, WATERSAVE. A New Approach to The Design of Water Utilization Systems in Refineries and Process Plants, *AIChE Spring Meeting*, paper 43 b, Houston.
20. Savelski, M. J. and Bagajewicz, M. J., 2000, On the optimality conditions of water utilization systems in process plants with single contaminants, *CES*, 55(21): 5035–5049.
21. Papoulias, S. A. and Grossman, I. E., 1983, A Structural Optimization approach in Process-synthesis-II: Heat recovery networks, *Comput Chem Eng*, 7: 707.
22. Olesen, S. G. and Polley, S. G., 1997, A Simple Methodology for the Design of Water Networks Handling Single Contaminants, *Trans IChemE Part A*, *Chem Eng Res Des*, 75.
23. Bagajewicz, M., 2000, A Review of Recent Design Procedures for Water Networks in Refineries and Process Plants, *Comput Chem Eng*, 24(9): 2093–2115.
24. Wang, Y. P. and Smith, R., 1994, Design of Distributed Effluent Treatment Systems, *Chem Eng Sci*, 49(18): 3127.
25. Mann, J. G. and Liu Y. A., 1999, Industrial Water Reuse and Eastewater Minimization, (McGraw-Hill, New York)

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