CHEMICAL ENGINEERING TRANSACTIONS
VOL. 81, 2020
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it
Guest Editors: Petar S. Varbanov, Qiuwang Wang, Min Zeng, Panos Seferlis, Ting Ma, Jiří J. Klemeš

ISBN 978-88-95608-79-2; **ISSN** 2283-9216
DOI: 10.3303/CET2081025

Pinch Approach for Targeting in Multi-Contaminant Material Recycle/Reuse Network

Hon Huin Chin[a,*], Petar Sabev Varbanov[a], Peng Yen Liew[b], Jiří Jaromír Klemeš[a]

[a]Sustainable Process Integration Laboratory – SPIL, NETME Centre, Faculty of Mechanical Engineering, Brno University of Technology – VUT Brno, Technická 2896/2, 616 69 Brno, Czech Republic
[b]Department of Environmental Engineering and Green Technology, Malaysia-Japan International Institute of Technology (MJIIT), Universiti Teknologi Malaysia (UTM), Jalan Sultan Yahya Petra, 53100, Kuala Lumpur, Malaysia
chin@fme.vutbr.cz

Sophisticated approaches have been proposed to reduce material utilities such as water and hydrogen by designing resource allocation networks. For resource targeting, graphical methods have been developed for single quality material recycle/reuse. Up to now, systematic approaches to targeting fresh resources in a multi-contaminant material recycling network ahead of the detailed design are not readily available. This work presents a rigorous resource targeting procedure in the domain of multiple constraints (e.g. multi-contaminants). The problem is formulated as a resource-allocation superstructure model and modified to determine the model characteristics and obtain the optimal solution. These conditions are then transformed into a table and graphical forms to identify the minimum resource target. By inferring from the mathematical formulations, multiple cascades with individual contaminant/quality have to be analysed sequentially. The main issues are the identification of the proper assignment of material sinks or sources as well as their ranking into each cascade. An example is solved to illustrate the accuracy and applicability of the method. The method also provides insights into the problem as it could identify the limiting constraints for individual sinks.

1. Introduction

The increasing depletion of natural resources poses threats to processing facilities and strongly affects the socio-economic development. Process industries have pursued material conservation as the main strategy, advancing toward Circular Economy (Fan et al., 2019). Extending Heat Integration pioneered by Linnhoff et al. (1982) and his UMIST team, including the Total Site Heat Integration (Klemeš et al., 1997). El-Halwagi and Manousiouthakis (1989) introduced the problem of mass exchange networks (MENs) synthesis. El-Halwagi et al. (2003) later provided a single-stage targeting method to identify minimum resources for a single contaminant water network, with the solution strategy identified through rigorous analysis. An important variation of MENs was introduced in 1994 by Wang and Smith (1994). This methodology results in better constructions to recycle/reuse and regeneration. Both methods rely on the basic principle of concentration driving force. A review of the historical development of Pinch Analysis (PA) in water networks has been published by Foo (2009). Klemeš et al. (2018) have conducted a comprehensive overview of various extensions of PA in Mass Integration, including water and hydrogen integration. PA is well-established for a single contaminant water network, but the previous work on targeting procedure for multiple contaminant problems only locates the approximate freshwater flowrates, rather than the precise minimum.
Various attempts have been used to solve the recycle/reuse problems for multicomponent systems. Alva-Argáez et al. (1999) developed mixed-integer linear programming (MILP) based multi-contaminant transhipment model used for targeting. Their model could tackle the general problem of mass exchange networks, but it could not evaluate the mixing effect of multi-contaminant problems. To address this problem, Liu et al. (2009) proposed the Concentration Potential concept. The concept is based on the overall allocating possibility of source streams to demand streams. Li et al. (2017) provide a review of this approach with their extension and applicability. Fan et al. (2019) proposed an iterative approach to design the water network, including regeneration, recycle/reuse and wastewater treatment. Zhao et al. (2019) utilised the concept of designing heat-integrated water networks.

Paper Received: 02/04/2020; Revised: 16/05/2020; Accepted: 22/05/2020
Please cite this article as: Chin H.H., Varbanov P.S., Liew P.Y., Klemeš J.J., 2020, Pinch Approach for Targeting in Multi-Contaminant Material Recycle/Reuse Network, Chemical Engineering Transactions, 81, 145-150 DOI:10.3303/CET2081025

Other attempts include the work of Castaño and Higuita (2016), who used the property of turbidity (which sums the number of contaminants) in the design of water networks. Turbidity is regarded as the key measured parameter that correlates with the concentration of the suspended solids. Mabitla and Majozi (2019) presented a hybrid of graphical and mathematical approaches in solving multi-contaminant water and regeneration networks. The graphical approach involves the pre-processing steps to identify minimum water target and regenerator removal ratios. The multi-contaminant problem is solved using the reference contaminants approach from Calixto et al. (2015). However, their approach could only account at most two contaminants.
The previous works mainly applied mathematical design procedures for the multicontaminant recycle/reuse problem. None has presented a single-stage and systematic graphical targeting procedure. The calculation for designing multiple contaminant water networks can be fast with Mathematical Programming. However, it is often difficult to understand how the optimal solutions are obtained and determine the limiting bottlenecks from these models. In many cases, it is important to identify the target for minimum fresh resources usages ahead of the network configurations, providing a benchmark for detailed design. The capability of PA to target the resources and identify bottlenecks through visualisation provides added merits for its applicability in this type of problem. Realising the research gap, the main novelties in this work include:

(i) Developing a graphical targeting procedure for multi-contaminant water network through rigorous analysis.
(ii) Identifying the limiting contaminants based solely on the concentrations of available sinks and sources
(iii) Extending the application of the PA targeting framework from single to multiple quality indicators.

2. Methodology

The methodology consists of two sections. The mathematical analysis of the multi-contaminant water network problem is explained in section 2.1. This section shows the brief derivation of the equations used to identify, e.g. (a) the limiting contaminants, (b) the rankings of sources/sinks and (c) allocation of sinks to contaminant cascade. The hierarchical framework of the graphical targeting procedure is explained in section 3.

2.1 Mathematical analysis of the multi-contaminant water network

A typical source-sink allocation model of the water network is presented in Figure 1, with four main governing equations Eqs(1-4):

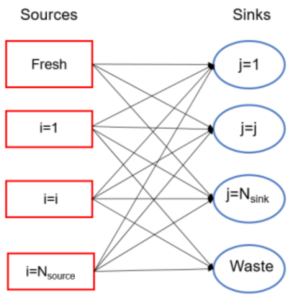

Figure 1: A superstructure material source-sink allocation model representation (El-Halwagi et al., 2003)

$$Min\ F_{FWT} = \sum_j F_{Fwj} \quad (1)$$

$$F_{SRi} = \sum_j F_{SRi,SKj} + F_{SRi,Waste} \quad \forall\ i \quad (2)$$

$$F_{SKj} = \sum_i F_{SRi,SKj} + F_{Fwj} \quad \forall\ j \quad (3)$$

$$\sum_i F_{SRi,SKj} C_{k,SRi} + F_{Fwj} C_{k,Fw} \leq F_{SKj} Z_{k,SKj} \quad \forall\ j\ \forall\ k \quad (4)$$

Where F_{FWT} represents the total freshwater flowrate, F_{Fwj} is freshwater to sink 'j' flowrate, F_{SRi} is source 'i' flowrate, $F_{SRi,SKj}$ is source 'i' to sink 'j' flowrate, $F_{SRi,\ Waste}$ is source 'i' to waste flowrate, F_{SKj} is the sink 'j' flowrate, $C_{k,\ SRi}$ is the concentration of contaminant 'k' in source 'i', C_{Fw} is the concentration of contaminant 'k' in freshwater, and Z_{SKj} is the concentration of contaminant 'k' in sink 'j'.
Eq(1) is the objective function to be minimised, which is the freshwater target, Eq(2) and Eq(3) are the mass balances for sources and sinks, while Eq(4) represents the contaminant constraints for individual sinks. These

equations are crucial to understanding the model characteristics to derive the equations for obtaining the optimal freshwater target. The main characteristics are explained in sections 2.1.1 to 2.1.2.

2.1.1 Determine the limiting contaminant for a source-sink pair

For a problem with 1 source and 1 sink. Using Eq(3) and Eq(4), the mass sink balance and its contaminant constraint are:

$$F_{SK1} = F_{SR1,SK1} + F_{FW1} \quad (5)$$

$$F_{Fw1}C_{k,Fw} + F_{SR1,SK1}C_{k,SR1} \leq F_{SK1}Z_{k,SK1} \quad \forall \text{k} \quad (6)$$

Eqs(5) and (6) can be rearranged to obtain the freshwater target. First, express $F_{SR1,SK1}$ in terms of F_{SK1} and F_{FW1} in Eq(5), and substitute into Eq(6). Assuming pure freshwater ($C_{k, FW} = 0$) and move F_{FW1} to the left-hand side, the minimum freshwater requirement is formulated- see Eq(7). The water requirement is equal to the maximum value among the requirements across all of the contaminants. The maximum value is used as one contaminant may require more dilution than another and becomes limiting.

$$F_{Fw1} \geq \max_k \left(F_{SK1} \left[1 - \left(\frac{Z_{k,SK1}}{C_{k,SR1}} \right) \right] \right) \;\; => \;\; F_{Fw1} \geq F_{SK1} \left[1 - \min_k \left(\frac{Z_{k,SK1}}{C_{k,SR1}} \right) \right] \quad (7)$$

If $Z_{k,SK1}$ is less than $C_{k,SR1}$, that means the freshwater is needed, and the Source can be used in the sinks, but not all sources due to contaminant constraint. Now if $Z_{k,SK1}$ is larger or equal to $C_{k,SR1}$, freshwater is not needed. The Source can be fully reused contaminant is not the constraint. To identify the limiting contaminant for the source-sink pair, one just needs to check the ratio of sink concentration to the source concentration for each contaminant. The minimum value across all contaminants 'k' occurs for the limiting contaminant. The method agrees but differs slightly with the concentration potential concept proposed by Liu et al. (2009).

2.1.2 Ranking of sources and sinks

Assume a case with 2 contaminants (k1 and k2), 2 sources (SR1 and SR2) are used on 2 sinks (SK1 and SK2) and $C_{k1,SR1} < C_{k1,SR2}$ (SR1 is cleaner than SR2 on contaminant 1) and $C_{k2,SR1} > C_{k2,SR2}$. (SR2 is cleaner than SR1 on contaminant 2). Using Eq(3) and Eq(4), for SK1, express $F_{SR2,SK1}$ in terms of $F_{SR1,SK1}$ and F_{FW1}, the freshwater target for SK1 is shown in Eq(8). Similarly for SK2 but express $F_{SR1,SK2}$ in terms of $F_{SR2,SK2}$ and F_{FW2}, with its freshwater target shown in Eq(9).

$$F_{Fw1} \geq \max_{k \in k1,k2} \left(F_{SR1,SK1} \left[\frac{C_{k,SR1}}{C_{k,SR2}} - 1 \right] + F_{SK1} \left[1 - \left(\frac{Z_{k,SK1}}{C_{k,SR2}} \right) \right] \right) \text{, the freshwater target for SK 1} \quad (8)$$

$$F_{Fw2} \geq \max_{k \in k1,k2} \left(F_{SR2,SK2} \left[\frac{C_{k,SR2}}{C_{k,SR1}} - 1 \right] + F_{SK1} \left[1 - \left(\frac{Z_{k,SK2}}{C_{k,SR1}} \right) \right] \right) \text{, the freshwater target for SK 2} \quad (9)$$

Notice for SK1- Eq(8), for contaminant 'k1', since $C_{k1,SR1} < C_{k1,SR2}$, the $F_{SR1,SK1}$ is preferred to be maximised to minimise the freshwater target. If this is a single contaminant problem, it proves the sources with lower concentration should be prioritised first. While for contaminant 'k2', $F_{SR1,SK1}$ is preferred to be minimised as it would contribute to more freshwater requirement. This creates a conflicting issue: whether to maximise or minimise $F_{SR1,SK1}$. However, this decision strongly depends on the right-hand side term as it is a constant term. If ($Z_{k1,SK1}$/ $C_{k1,SR2}$) < ($Z_{k2,SK1}$/ $C_{k2,SR2}$), the constant term for contaminant 'k1' is larger than 'k2', and $F_{SR1,SK1}$ is preferred to be maximised, This suggests that SK1 is likely to be limited by contaminant 'k1'. In this case, SK1 would follow the order with contaminant 'k1', and typical targeting approach for contaminant 'k1' can be performed. Similar inference can be obtained for SK2 by observing Eq(9). Assuming SK1 is limited by contaminant 'k1' and SK2 is limited by contaminant 'k2'. The total freshwater target becomes:

$$\begin{aligned} \textstyle\sum_j F_{Fwj} = F_{Fw1} + F_{FW2} \geq \; & \left(F_{SR1,SK1} \left[\frac{C_{k1,SR1}}{C_{k1,SR2}} - 1 \right] + F_{SK1} \left[1 - \left(\frac{Z_{k1,SK1}}{C_{k1,SR2}} \right) \right] \right)_{contaminant\ k1} + \\ & \left(F_{SR2,SK2} \left[\frac{C_{k2,SR2}}{C_{k2,SR1}} - 1 \right] + F_{SK1} \left[1 - \left(\frac{Z_{k2,SK2}}{C_{k2,SR1}} \right) \right] \right)_{contaminant\ k2} \end{aligned} \quad (10)$$

By deducing from Eq(10), multiple cascades are required to identify the freshwater target. Using the example above, SK1 should follow the cascade for contaminant 'k1' and SK2 follows the cascade for contaminant 'k2'. The sources should be arranged with ascending order of the concentration for each contaminant cascades. For contaminant 'k1', the sources should be arranged with the order of concentration 'k1' (SR1- > SR2); while for contaminant 'k2', the sources should be arranged in the order of (SR2- > SR1).

In a number of cases are sinks are limited by more than one contaminant. Let us say SK1 is limited by contaminant 'k1', and it was established that $F_{SR1,SK1}$ should be maximised- Eq(8). However, assume that the

SR1-SK1 pair is limited by contaminant 'k2' (can be determined by Eq(7)), there is a maximum amount of $F_{SR1,SK1}$ and $F_{SR2,SK1}$ that can be allocated to SK1, and this creates a scenario that SK1 is both limited by contaminants 'k1' and 'k2'. The maximum amount of $F_{SR1,SK1}$ can be determined by equating the freshwater target for both contaminants, as presented below:

$$F_{SR1,SK1}^{\max}\left[\frac{C_{k1,SR1}}{C_{k1,SR2}}-1\right]+F_{SK1}\left[1-\left(\frac{Z_{k1,SK1}}{C_{k1,SR2}}\right)\right]=F_{SR1,SK1}^{\max}\left[\frac{C_{k2,SR1}}{C_{k2,SR2}}-1\right]+F_{SK1}\left[1-\left(\frac{Z_{k2,SK1}}{C_{k2,SR2}}\right)\right] \quad (11)$$

3. Multi-contaminant Pinch Analysis

Based on the mathematical analysis in Section 2, the overall steps for freshwater targeting using PA is derived and presented as follow:

(i) For each sink 'j', (a) Identify which contaminant is limiting for the sink. This is done by evaluating the concentration ratio for each source 'i' for each contaminant 'k', i.e. $\min_k(\frac{Z_{1,SKj}}{C_{1,SRi}},\ldots,\frac{Z_{k,SKj}}{C_{k,SRi}})$. If contaminant 'k' corresponds to the minimum value of the ratio, then the sink is limited by contaminant 'k' for the specific source 'i' - see Eq(7). This ratio shows the recycling potential of the sources to the specific sink. (b) Allocate the current sink j to the cascade of its limiting contaminant. If it has other limiting contaminant, identify the maximum source flowrate that can be allocated to the contaminant cascade using Eq(11).

(ii) For each contaminant cascade, arrange the allocated sinks and all sources in ascending order of their concentrations. The order for the sources may differ among the contaminant cascades.

(iii) For each contaminant, construct the Source and the Sink Composite Curves (CCs). Perform Pinch Analysis/Cascade Table Analysis based on the assigned 'Sinks' with the remaining 'Sources' sequentially.

4. Case study

A pulp and paper mill case study from Lovelady et al. (2007) is used to demonstrate the proposed method. Tables 1 and 2 shows the water sources and sinks data for the case study.

Table 1: Available water sources sand sinks

Sources (SRi)	F_{SRi} (t/h)	C_{SRi} (ppm)			Sinks (SKj)	F_{skj} (t/h)	C_{skj} (ppm)		
		Cl^-	K^+	Na^+			Cl^-	K^+	Na^+
SR1	8,901	0	0	0	SK1	13,995	34.4	12.7	89.4
SR2	1,450	308.6	115.5	840.3	SK2	1,450	241	2.4	241
SR3	1,024	0	0	0	SK3	5,762	0	0	0
SR4	4,990	500	5	500	SK4	30,990	3.7	1.1	3.6

For step (i), of the algorithm in section 3, the limiting contaminant is first identified for each sink by calculating the individual sink-to-source concentration ratio. The freshwater is assumed to be free from contaminants (i.e. $C_{k,FW}$ = 0 for k = Cl, K, Na). The concentration ratio is used as the identifier for the limiting contaminants. The contaminant that corresponds to the minimum ratio among impurities is the limiting contaminant for a particular stream. Table 3 shows the results of the identification and the allocation of sinks. SR1, SR3 and SK3 are neglected since they are technically pure water streams. For SK1, as Na^+ correspond to the lowest ratio for SR2 and Cl^- correspond to the lowest ratio with SR4 among the contaminants group, which shows the recycling potential for the sources to SK1 is limited by Cl^- and Na^+. SK2 is limited by K^+, and SK4 is limited by Na^+.

Table 3: Identification of limiting contaminants for sinks (highlighted in bold represent the minimum values)

Sources (SRi)	SK1 ($Z_{k,SK1}/C_{k,SRi}$)			SK2 ($Z_{k,SK2}/C_{k,SRi}$)			SK4 ($Z_{k,SK3}/C_{k,SRi}$)		
	Cl^-	K^+	Na^+	Cl^-	K^+	Na^+	Cl^-	K^+	Na^+
SR2	0.11	0.109	**0.106**	0.78	**0.021**	0.287	0.012	0.01	**0.004**
SR4	**0.07**	2.54	0.179	0.482	**0.48**	0.482	0.0074	0.22	**0.0072**

The step (ii) involves the ranking of the sinks and sources for each contaminant. The ranking of the streams is to plot the Source and Sink CCs for each contaminant. As shown in Table 3, only a single sink stream is assigned to each contaminant cascade, while SK3 can be assigned to any of the group. The ranking of the sources for each contaminant is: Cl^- = {SR1- > SR3- > SR2- > SR4}, K^+ = {SR1- > SR3- > SR4- > SR2} and Na^+ = {SR1- > SR3- > SR4- > SR2}.

To assign the sinks to the specific contaminant CCs, the identification of limiting contaminant(s) play a key role. SK3 can be assigned to any of the contaminant cascades since it technically requires pure water stream. For SK1, it can be assigned to the contaminant ‘Cl^-’ CC or ‘Na^+’ CC, since SK1 is likely to be limited by both contaminants, as shown in Table 3. In this study, SK1 is assigned to the ‘Cl^-’ CC. As the flowrates of SR1 and SR3 are insufficient to fully cover SK1, SR2 has to be used for SK1. This indicates that SR2 cannot be assigned only to the ‘Cl^-’ CC. It is preferable to use SR2 and SR4 so that both contaminant loads would reach the maximum sink impurity limits to maximise the source reuse. Using Eq(11), the maximum amount of SR2 that can be allocated to SK1 in Cl^- cascade is calculated as 1,447.67 t/h (comparison between SR2 and SR4, with contaminant ‘Cl^-’ and ‘Na^+’), slightly less than the flowrate of SR2: 1,450 t/h.
PA is performed as the last step for individual contaminant sequentially. Figure 2 illustrates the Load vs Flowrate Composite Curves (CC) for each contaminant based on the sequence ‘Cl^-’ to ‘K^+’ and ‘Na^+’. The sequence for the contaminant is not important since it would yield a similar water target.

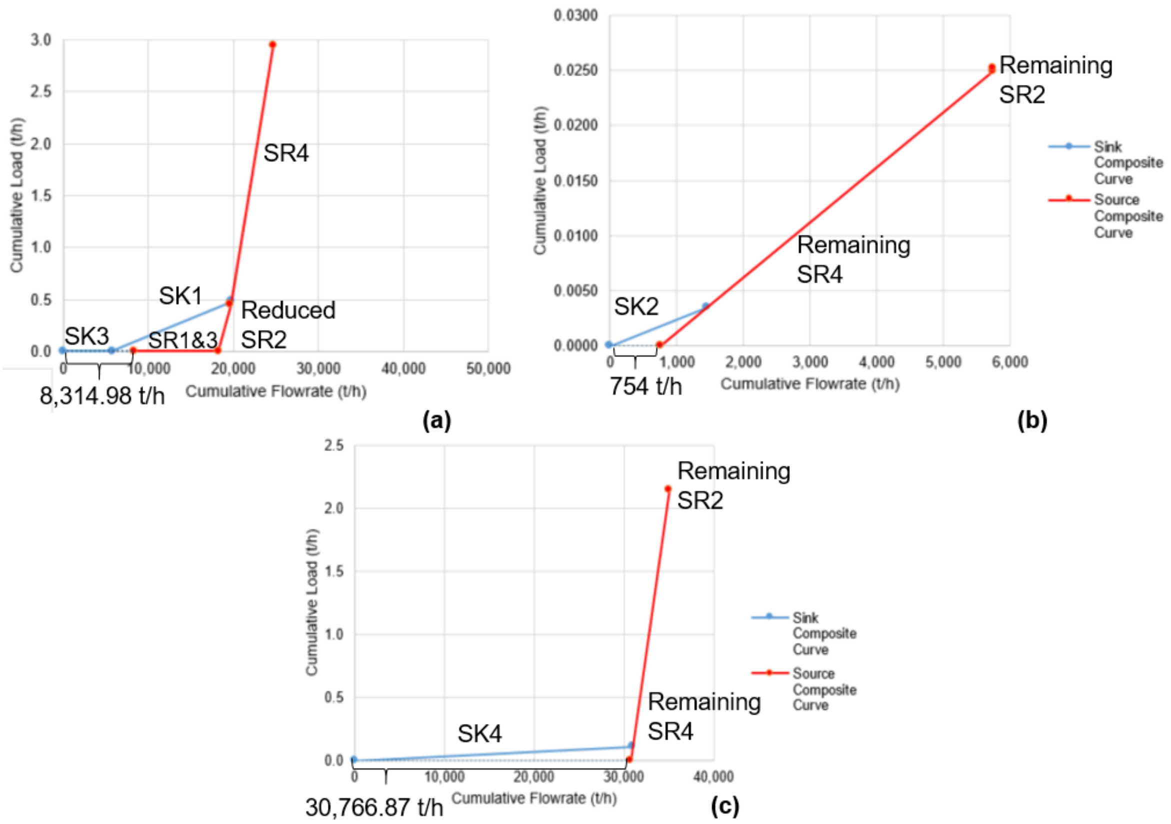

Figure 2: Source and Sink Composite Curves for contaminants (a) Cl^- (b) K^+ (c) Na^+

For the ‘Cl^-’ CC, the Source CC is plotted according to the ranking: SR1->SR3->SR2->SR4 (based on ranking developed in step ii). Only 1,447.67 t/h of SR2 can be included in this Source CC, as the pair of SR2-SK1 is also limited by Na^+. The ‘Cl^-’ Sink CC is plotted for SK3->SK1. SK3 requires a pure water stream so it can be assigned to any of the contaminant groups. If there is more than one sink limited by the same contaminant, arrange them with ascending order of the limiting contaminant’s concentration and plot the Sink CC. The Source CC is then shifted until it is on the right side of the Sink CC, as shown in Figure 2a. Subsequently, for ‘K^+’ CC, the Source CC should be plotted with the order of SR1- > SR3- > SR4- > SR2. Since SR1 and SR3, as well as part of SR2, are used up in ‘Cl-‘ CC, they are not included in ‘K^+’ CC. The remaining SR2 and SR4 are used to construct the Source CC. The small remaining amount of SR2 is stacked after SR4 (Figure 2b) The Source CC is also shifted until it is on the right side of the Sink CC. A similar procedure is then performed for contaminant Na^+ (Figure 2c). Since the only SR4 is used for ‘K^+’ CC, the remaining SR2 is still carried forward to ‘Na^+’ CC. The minimum freshwater target is identified as the sum of all freshwater targets for each contaminant CCs. The total freshwater target is 39,835.85 t/h, which is consistent with the result obtained by solving the superstructure model using GAMS. The solutions from GAMS also show that SK2 is limited by K^+ and SK4 is limited by Na^+, while SK1 is limited by both Cl^- and Na^+. This proves the accuracy and applicability of the proposed method.

5. Conclusion

This work identified a graphical targeting procedure for multi-contaminant material recycle-reuse networks, mainly for water networks. By analysing the rigorous source-sink allocation model, sequential PA or cascade analysis for each contaminant is required to identify the true minimum freshwater target. The main trick is to identify the allocation of sinks to the proper cascade by determining its limiting contaminants. The ratio between the sink concentration to maximum source concentration of the contaminant serves as the identifier for limiting contaminant. Another important part is to determine the other limiting contaminant for the sink constrained by the sources, as this limit the number of sources that can be used for each contaminant cascade. The allocated sinks and sources are then arranged with ascending order of their concentrations. After the assignment of sinks and sources to each contaminant cascade, the typical PA can then be performed for each cascade sequentially. The proposed method is validated with a study from a pulp and paper mill, which is a three contaminants water recycle/reuse problem. The freshwater target obtained is 39,835.85 t/h, which agrees well with the solutions obtained from solving the superstructure model. The limiting contaminants for each sink are also consistent with those solved by mathematical optimisation method. This proves the accuracy and legitimacy of the method. The limitation of this study is that manual curve shifting to identify the target can be tedious. Future research should focus on a Multi-Contaminant Cascade Table Analysis. It can be developed for automated resource targeting. Water regeneration potential and impure fresh resource should be incorporated into the analysis as well.

Acknowledgements

The EU supported project Sustainable Process Integration Laboratory – SPIL funded as project No. CZ.02.1.01/0.0/0.0/15_003/0000456, by Czech Republic Operational Programme Research and Development, Education, Priority 1: Strengthening capacity for quality research in collaboration with Universiti Teknologi Malaysia (UTM) is gratefully acknowledged.

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