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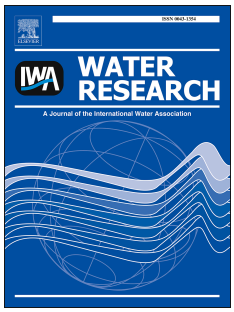

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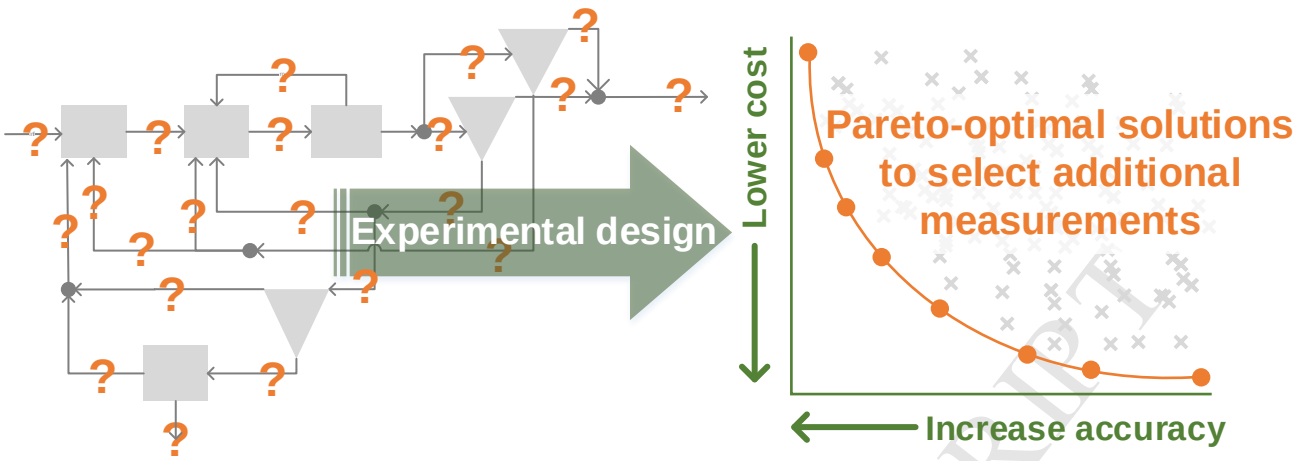

Experimental design for evaluating WWTP data by linear mass balances

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Abstract

A stepwise experimental design procedure to obtain reliable data from wastewater treatment plants (WWTPs) was developed. The proposed procedure aims at determining sets of additional measurements (besides available ones) that guarantee the identifiability of key process variables, which means that their value can be calculated from other, measured variables, based on available constraints in the form of linear mass balances. Among all solutions, i.e. all possible sets of additional measurements allowing the identifiability of all key process variables, the optimal solutions were found taking into account two objectives, namely the accuracy of the identified key variables and the cost of additional measurements. The results of this multi-objective optimization problem were represented in a Pareto-optimal front.

The presented procedure was applied to a full-scale WWTP. Detailed analysis of the relation between measurements allowed the determination of groups of overlapping mass balances. Adding measured variables could only serve in identifying key variables that appear in the same group of mass balances. Besides, the application of the experimental design procedure to these individual groups significantly reduced the computational effort in evaluating available measurements and planning additional monitoring campaigns. The proposed procedure is straightforward and can be applied to other WWTPs with or without prior data collection.

Keywords: experimental design; data validation; mass balances; data reconciliation; wastewater treatment plant;

1 Introduction

The importance of reliable data for wastewater treatment plant (WWTP) design, process optimization, operator training, developing control strategies, benchmarking and simulation is commonly advocated (Meijer et al., 2015, 2002; Puig et al., 2008; Rieger et al., 2010; Spindler, 2014; Villez et al., 2013a). Typical data in this respect concern flows and concentrations of components. Depending on the objectives, available historical data are complemented with additional data obtained through one or more intensive monitoring campaigns using classic sampling followed by laboratory analyses and/or online sensors.

Data reconciliation is a proven technique to evaluate the consistency of collected data (Crowe, 1996; Ozyurt and Pike, 2004). It involves a procedure of optimally adjusting estimates for variables such that these estimates satisfy the conservation laws and other constraints (Crowe, 1996) and are therefore more accurate than the original values. Data reconciliation is often accompanied by statistical tests for gross error detection (measurement validation), which verify whether the deviation between each estimate and its measurement is acceptable compared to the measurement error.

Even though data reconciliation has been widely applied in (bio)chemical engineering for decades (Madron et al., 1977; Madron and Veverka, 1992; van der Heijden et al., 1994b), this concept so far has received relatively little attention in wastewater treatment process engineering. Some studies applied the concept of redundancy analysis and variable classification, which are closely related to the principles and objectives of data reconciliation, for sensor fault detection (Villez et al., 2016, 2015, 2013b) or for describing redundancy in the data set (Spindler, 2014). In other studies, data reconciliation was directly applied for the validation of a WWTP process data set for modelling, process optimization or plant performance evaluation (Behnami et al., 2016; Meijer et al., 2015, 2002; Puig et al., 2008). The effects of erroneous data on modelling errors was investigated by Lee et al. (2015), applying gross error detection. Rieger et al. (2010) put the concept of data validation by mass balancing in a general data collection framework, stressing the importance of measurement planning to guarantee a successful subsequent data validation for WWTP. Besides full-scale processes, data reconciliation was also applied to long-term data of a lab scale wastewater treatment reactor to identify different anabolic reactions pathways (Lotti et al., 2014).

The abovementioned studies explicitly or implicitly pointed out that it is vital for a measurement plan to satisfy the redundancy and steady-state conditions, as important prerequisites for successful data reconciliation. While obtaining data fulfilling the steady-state condition was discussed in detail by Meijer et al. (2015), this work focuses on the redundancy requirement.

Redundancy of variables means that their measured values can also be calculated from other (measured) variables. However, for many WWTPs, there are often not sufficient initially measured data available and additional measurements typically need to be carried out (in a monitoring campaign) to ensure the required degree of redundancy for data reconciliation. In this respect, “overlapping mass balances” and “closed mass balances” are typically aimed at. The term “overlapping mass balances” refers to mass balances over single or combined subsystems that share one or more mass flows or have at least one variable in common. The term “closed mass balances” refers to mass balances in which all variables are measured and which can typically be set up for conserved quantities such as total mass flows or total phosphorus mass. This practice of adding overlapping and closed mass balances increases the overall system redundancy and has therefore been commonly applied for data reconciliation in wastewater process engineering (Lee et al., 2015; Meijer et al., 2015, 2002; Puig et al., 2008).

However, increasing the *overall* system redundancy does not guarantee the possible identification of specified key variables (van der Heijden et al., 1994a). The approach of Meijer et al. (2015, 2002) and Puig et al. (2008), aiming at increasing redundancy by adding measurements to set up overlapping and closed mass balances, therefore, involved the risk of adding trivial mass balances and associated unnecessary additional measurements. For WWTP data reconciliation, the question remains in what manner and to which extent additional measurements, entailing additional overlapping and closed mass balances, effectively lead to reliable and improved estimates of the key variables under concern.

This work provides a practical stepwise procedure to determine sets of additional measurements that guarantee the possible identification of key process variables, which means that their value can be calculated from other, measured variables. More specifically, these sets of additional measurements satisfy the required degree of redundancy for data reconciliation considering constraints in the form of linear mass balances. The focus of this work is on the experimental design, i.e. the determination of additional measurements allowing the identification of key variables. The actual application of data reconciliation to obtain reliable and improved estimates for key variables is the topic of a follow-up paper. The redundancy of measurements was analysed to gain insight in the way measured variables are related through linear mass balances. Particular attention was paid to the contribution of additional overlapping and closed mass balances. Through this comprehensive redundancy analysis, shortcomings of previous studies in selecting meaningful additional measurements were overcome. Moreover, the accuracy of the reconciled results and the cost of additional measurements were considered in finding optimal sets of additional measurements. The procedure was demonstrated for a full-scale WWTP.

2 Experimental design procedure

An experimental design procedure for practical application to wastewater treatment processes was derived (Figure 1).

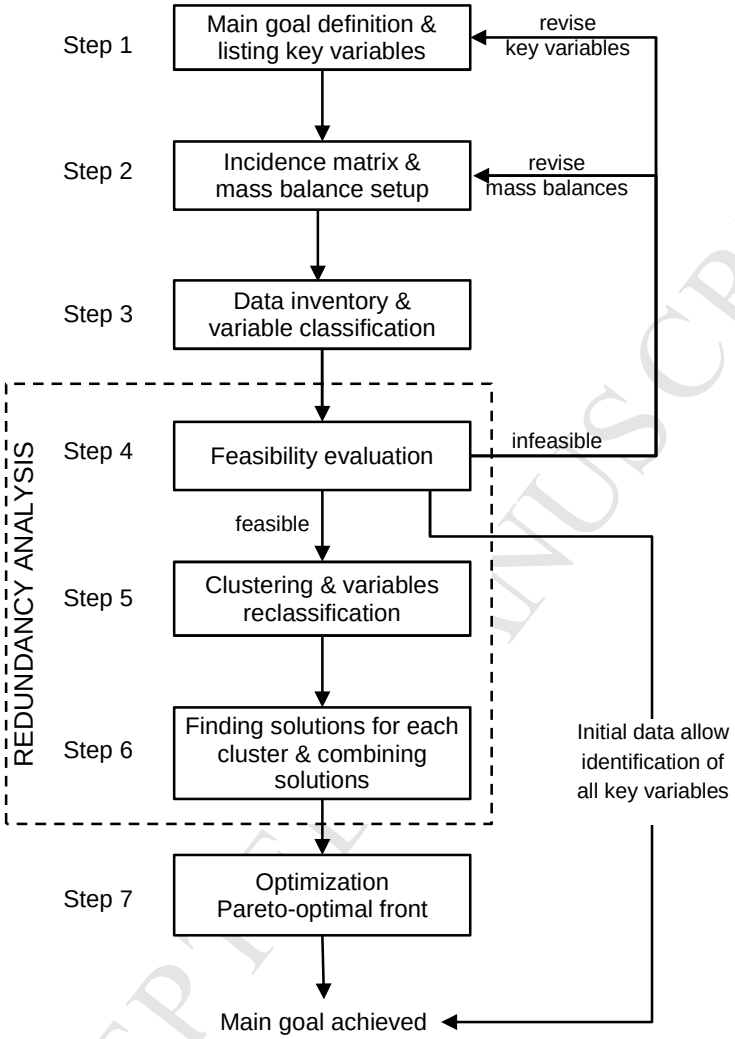


Figure 1. Experimental design procedure for the selection of sets of additionally measured variables that allow the identification of key variables.

The key variables are defined first (Step 1), followed by the set-up of an incidence matrix and mass balances based on the process flow diagram (Step 2) and the inventory of available data (Step 3). Even though these 3 steps have been addressed previously in an intuitive approach for data collection (Meijer et al., 2015), they were now included in a more formal experimental design procedure, focusing on key variables, simplifying the mass balance set-up and reducing associated efforts. Moreover, a comprehensive redundancy analysis has been added in this study (Steps 4-6), to overcome the shortcoming of previous studies. It is now

checked up-front that the list of key variables and/or the set of set up mass balances are relevant in the sense that key variables are identifiable (Step 4). Mass balances and their corresponding variables are clustered (Step 5), which greatly improves the efficiency of finding all solutions, i.e. sets of additionally measured variables that satisfy the defined main goal (Step 6). Finally, a procedure to select the optimal solution in terms of additional measurement costs and accuracy of identified key variables has now been provided as well (Step 7). Step 4 to Step 7 were implemented in MATLAB 2014b (MathWorks®). More details on the individual steps are given below. Details on the applied procedures and on the theoretical background are provided in Supplementary Material A and B, respectively.

Step 1. Main goal definition - listing key variables

Data reconciliation can be applied to identify key process variables and at the same time detect possible gross errors. Key variables may be measured or not; their identification means that improved estimates of their values are obtained. These new estimates meet all the constraints (i.e., fit all mass balances) and are therefore considered more reliable and accurate (have a smaller standard deviation or error) than the original values. In case a key variable is measured, the new estimate is considered improved compared to the original measurements. In case the key variable is not measured, the new estimate is considered improved compared to the value directly calculated from original measurements (using the available set of mass balances).

In this step, all key process variables are listed. Typical examples of key process variables in a WWTP that need to be known with high accuracy concern influent and effluent mass flow rates of the activated sludge process (biological reactor) as well as the waste activated sludge mass flow rate. The oxygen requirements for chemical oxygen demand (COD) and nitrogen removal are also important process variables and therefore typically need to be calculated – they are typical unmeasured key variables.

The constraints which the new estimates of key variables need to meet, are in the form of linear mass balances, consisting of mass flow terms. For this reason, key variables, denoted as K^* are expressed in terms of total mass flows and mass flows of individual components (as indicated by the superscript *). It is important to note that the mass flow of a certain component at a certain place is only considered measured if both the corresponding flow rate and component concentration are measured.

The experimental design procedure aims at determining one or more sets of additional measurements that guarantee the identification of all key variables, while minimizing the cost of additional measurements and maximizing the accuracy of the identified key variables.

Step 2. Incidence matrix and mass balance setup

The process flow diagram of WWTP is translated into a so-called incidence matrix, which is a mathematical description of the flow network. The columns of the incidence matrix represent process streams and the rows represent individual or combined unit processes. The elements of this matrix are:

- 1, if stream enters a unit process,
- -1, if stream leaves a unit process,
- 0, if stream is not incident with a unit process.

To visualize the spatial distribution of the interrelated subsystems, it is advised to number and arrange the flows and unit processes in the matrix following the water line, starting from the influent and primary tanks and ending towards the dewatered sludge. In this way, the matrix diagonal represents the water flow through the WWTP (Meijer et al., 2015).

Following the setup of the incidence matrix, linear mass balances of total mass flows ($\rho \times Q$, or Q when assuming the same density ρ for all streams in that mass balance) and individual mass flows, e.g. total phosphorus (*mTP)*, COD (*mCOD*) and total nitrogen (*mTN*), are set up for all subsystems considered. These subsystems could either be individual or combined unit processes. The resulting mass balances need to contain all key variables listed in Step 1. More detailed practical guidance on the selection of conservative quantities is provided by Meijer et al. (2015).

Step 3. Data inventory and variable classification

Once the mass balances are set up, an inventory is made of initially measured and initially unmeasured process variables that appear in the mass balances. The values of measured variables are obtained from routine lab analyses or through online monitoring. These are typically flows (Q) and concentrations of individual components such as COD, total nitrogen (TN) and total phosphorus (TP).

For optimization purposes, the expected measurement costs of all unmeasured variables (in the form of flow and concentration) and the measurement errors (standard deviation of the mean of the measurements in the form of mass flow) of all variables are also inventoried. In case the measurement error of a variable is not known or cannot be realistically estimated from expert knowledge, one could use a small error compared to those of other variables, essentially assuming a relatively good measurement, which still allows to track the

error propagation to the identifiable variables. Note that the relative magnitude of the measurement errors is of importance, rather than their exact values.

Let M be the set of initially measured variables and U be the set of initially unmeasured variables resulting from the data inventory. Part of the initially unmeasured variables are unmeasurable; they constitute the subset X of U. The remaining initially unmeasured variables could potentially be measured and constitute a complementary subset P_a of U ($P_a = U - X$). The aforementioned variables are typically expressed in terms of (volumetric) flows and concentrations.

Step 4. Feasibility evaluation

The feasibility of satisfying the main goal, i.e. of identifying all listed key variables, is evaluated for two extreme cases of measurement availability:

(**i**) All potential additionally measured variables P_a are measured additionally. It is thus checked whether all key variables are identifiable for the largest set of potential (additional) measurements and for the given set of mass balances. In case one or more key variables are not identifiable, it is recommended to first review the set of mass balances. The mass balances need to contain all key variables. Besides, non-identifiability could also result from mistakenly neglected flows or because of an oversimplified plant layout. If revising mass balances does not result in the identifiability of all key variables, there is insufficient redundancy in the system and it is advised to remove unidentifiable key variables, i.e. to return to Step 1. Once all key variables are identifiable considering the largest possible set of additional measurements, possibly after revising mass balances and/or key variables, the second extreme case of measurement availability is evaluated.

(ii) Only initially measured data are available. If all key variables are identifiable from the set of initial measurements, the main goal is fulfilled *a priori* and there is no need for additional measurements. If this is not the case, the procedure proceeds to Step 5 and Step 6 to determine sets of additional measurements resulting in the identifiability of all key variables. The existence of such sets of additional measurements is guaranteed by (i), which ensures the best possible definition of mass balances and removed key variables that are not identifiable *a priori*.

The identifiability of key variables is checked through redundancy analysis, based on the procedures of van der Heijden et al. (1994) and Klamt et al. (2002), as detailed in the Supplementary Material (section B1 for the theoretical background and section A1 for the practical implementation).

Step 5. Clustering and variables reclassification

Once the identification of key variables has been evaluated feasible, it will be investigated which set(s) of additional measurements are required to this end. This procedure is simplified by clustering the mass balances in groups of overlapping mass balances, i.e. mass balances that have at least one variable (total or individual mass flow rate) in common.

Clustering is based on redundancy analysis, involving the set-up of redundancy equations (see Supplementary Material B1). The redundancy equations are obtained from the original set of mass balances by eliminating all unmeasured variables, such that only measured variables remain. Variables that appear in a single redundancy equation will be used in data reconciliation to identify each other. When redundancy equations are interrelated by one or more variables, they will also be used to identify the variables in the related equations. The identifiability of variables in a group of interdependent redundancy equations is independent from the identifiability and measurement availability of variables in the other groups.

In order to cluster the mass balances in groups of overlapping mass balances, the redundancy equations are derived assuming all variables are measured. In this way, the maximum number of relations between (measured) variables can be identified, allowing subsequent variable reclassification clustering in groups of interdependent variables. First, the redundancy equations are clustered in groups of redundancy equations that are related by one or several variables. Second, groups of variables that belong to the corresponding groups of redundancy equations are formed (variable reclassification). Finally, based on groups of variables, the mass balances are clustered in group of overlapping mass balances that only contain variables of the corresponding groups.

After clustering the mass balances in groups of overlapping mass balances, variable classification was retaken for each group. Each group has its own measured variables (M), unmeasured variables (U), potential additionally measured variables ($P_a = U - X$), unmeasurable key variables (X), and key variables (K^*) that contribute to mass flow terms in the overlapping mass balances of that group. It is important to realize that flow variables (Q) are implicitly taken up in the individual mass flows (*mTP*, *mCOD*, *mTN*). For this reason, concentration variables (*TP*, *COD* or *TN*) always appear together with the flow rate (Q) of the corresponding stream while clustering. It is thus possible that a single (flow) variable appears in multiple groups.

The routine of clustering and variable reclassification is provided in Supplementary Material A2.

Step 6. Finding solutions

Clustering mass balances into groups of overlapping mass balances significantly simplifies the procedure of finding solutions. Indeed, the identifiability of key variables in one group of overlapping mass balances is independent from the measurement availability of variables in other groups; the measurement availability of a variable in a group of overlapping mass balances only helps identifying other variables in that group. The solutions for each group of overlapping mass balances can thus be derived separately and then combined.

Solutions are found by checking for all potential sets of additional measurements (per group) whether they guarantee the key variables (of that group) to be identifiable. The identifiability of key variables is checked through redundancy analysis, based on the procedures of van der Heijden et al. (1994) and Klamt et al. (2002), analogously as in Step 4 (see Supplementary Material B1). Since the key variables of all groups need to be identified simultaneously, the overall solutions are derived by combining the solutions for the individual groups of overlapping mass balances, while discarding duplicates. Step 6 is detailed in Supplementary Material A3.

Step 7. Optimization

In Step 7, the costs and accuracy are calculated for all solutions. Each set of additional measurements that guarantees all key variables K^* to be identifiable, is referred to as a solution and is characterized by a $1 \times n_P$ row vector $A = (a_1 \dots a_{np})$ consisting of binary decision variables a_j that indicate whether the corresponding potential additionally measured variables in P_a were selected to measure additionally ($a_j = 1$) or not ($a_j = 0$).

For every solution, the corresponding cost of additional measurements is calculated as the sum of the individual costs w_{aj} of additional measurements a_j, similar to the approach of Villez et al. (2016):

$$f_C(A) = \sum_{j=1}^{n_P} w_{aj}.a_j = W_a.A' \quad \text{(Eq.1)}$$

$W_a = (w_1 \dots w_{np})$ is a $1 \times n_P$ weighing vector, in which each element is w_{aj}.

The average variance of new estimates of key process variables (inversely related to accuracy), is calculated relative to the variance for the so-called reference solution, according to Eq. 2, and is termed $f_V(A)$. The reference solution, expressed as a $1 \times n_P$ vector $A_r = (1)$, is the solution obtained when all possible additional measurements P_a are measured additionally.

242 $$f_V(A) = \frac{1}{n_k}\sum_{i=1}^{n_k} v_i/v_i^r = \frac{1}{n_k}\sum_{i=1}^{n_k} w_{vi}.v_i = \frac{1}{n_k} W_v.V' \quad \text{(Eq.2)}$$

243 $V = (v_1 \dots v_{nk})$ denotes a $1 \times n_k$ vector of variances of new estimates ($v_i \geq 0$) of the key variables (hereafter
244 referred to as variance of key variables) when the solution A is implemented. The calculation of V is detailed
245 in the Supplementary Material (B2 for theoretical background and A4 for practical implementation). $W_v =$
246 $(w_{v1} \dots w_{nk})$ is a $1 \times n_k$ vector of non-negative weights, $w_{vi} = 1/v_i^r$, in which v_i^r represents the variance of the
247 key variables i when the reference solution is implemented.

248 Adding measurements to an existing set of measurements results in a smaller variance of new estimates
249 obtained through data reconciliation (van der Heijden et al., 1994). Therefore, the reference solution A_r is a
250 best known solution, which results in the smallest variance v_i^r (highest accuracy) of new estimates of the
251 key variables ($\forall i \in \{1,2,\dots,n_K\}: v_i^r \leq v_i$). The objective function $f_V(A)$ is a variation on the V-optimality choice
252 in the experimental design theory (Pukelsheim, 2006). Essentially, the use of relative variances of a solution
253 to a best known solution is a relevant choice to circumvent the problems due to the different units in which
254 different key variables are expressed. The division by number of key variables (n_K) makes this objective
255 such that in the best case the objective function $f_V(A)$ equals unity.

256 Finding an optimal solution is a multi-objective optimization problem consisting of finding the solution that
257 minimizes both $f_C(A)$ and $f_V(A)$. In this study, the Pareto-optimal solutions were determined, for which a
258 lower cost can only be obtained at the expense of a lower accuracy and vice versa. The implementation of
259 this step is detailed in Supplementary Material A4.

260

3 Application to a full-scale WWTP

WWTP under study

The proposed experimental design procedure was applied to WWTP Houtrust, The Hague, The Netherlands. Figure 2 displays a simplified configuration of this plant including all important streams; comprising a “three stage Phoredox process” or A2/O design. The full configuration of this plant is given in Supplementary Material C1; a more extensive plant description can be found in Meijer et al. (2015).

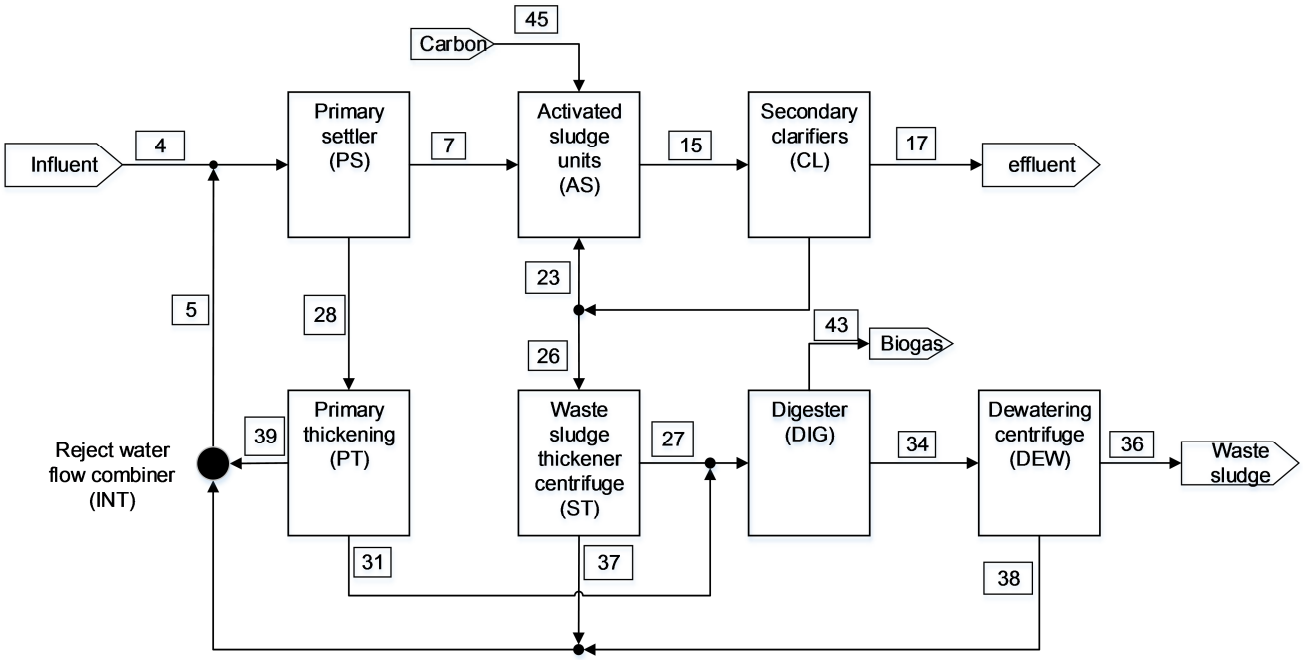


Figure 2. Simplified process flow diagram of WWTP Houtrust.

Step 1. Main goal definition - listing key variables

The experimental design procedure aims at determining one or more sets of additional measurements that guarantee the identifiability of all key variables, while minimizing the cost of additional measurements and maximizing the accuracy of the identified key variables. More specifically, key variables in the form of total mass flow and individual mass flows (COD, total nitrogen and total phosphorus) of the following streams had to be identifiable:

- Settled influent, i.e. influent of the activated sludge process (stream 7),
- WWTP influent (stream 4) and effluent (stream 17),
- Waste activated sludge (stream 26),

- WWTP waste sludge (stream 36),
- Reject water (stream 5),
- Biogas (stream 43, in this case only the mass flow of COD had to be identified).

Besides, the following unmeasurable key variables related to process performance had to be identifiable:

- Required oxygen for the oxidation of COD (OCcod, $kg.day^{-1}$),
- Amount of denitrified nitrogen (DENI, $kg.day^{-1}$),
- Primary sludge flow and associated mass flows of COD, total nitrogen and total phosphorus (stream 28).

Step 2. Incidence matrix and mass balance setup

The incidence matrix of WWTP Houtrust was set up based on the simplified process flow diagram (Figure 2), representing the WWTP layout by the minimum numbers of subsystems and streams but still contained all the variables of interest. The resulting matrix (Table 1) contained 8 rows (or subsystems) and 17 columns (or streams).

Table 1. Incidence matrix of the WWTP Houtrust. Ingoing and outgoing streams are denoted by '1' and '-1', respectively.

Subsystems ↓	Stream number in process flow diagram																
	4	5	7	15	17	23	26	27	28	31	34	36	37	38	39	43	45
	Total Influent	Reject water	Settled influent	Inflow secondary clarifiers (CL)	WWTP effluent	Return activated sludge	Waste activated sludge (WAS)	Thickened WAS to digester	Primary sludge	Thickened primary sludge	Digested sludge	Evacuated sludge	Centrate WAS thickening	Centrate dewatering	Overflow primary thickener	Biogas	External carbon source
Primary settler (PS)	1	1	-1						-1								
Activated sludge units (AS)			1	-1		1											1
Secondary clarifiers (CL)				1	-1	-1	-1										
Waste sludge thickener (ST)							1	-1					-1				
Primary sludge thickener (PT)									1	-1					-1		
Digester (DIG)								1		1	-1					-1	
Dewatering centrifuge (DEW)											1	-1		-1			
Reject water flow combiner (INT)		-1											1	1	1		

In simplifying the full process flow diagram (Supplementary Material C1), the activated sludge unit processes (selector, predenitrification, anaerobic, anoxic, aeration and de-aeration tanks) were grouped into a combined unit (AS, Figure 2), since they involve the unmeasured loss and supply of components through the gas phase (N_2, CO_2 and O_2), which do not need to (and cannot) be distinguished among them. Buffer units were not explicitly considered, reasonably neglecting accumulation, separation and/or conversion of components in these units. The small streams, such as clean water stream (stream 40), ferric chloride sulfate ($FeClSO_4$) added for phosphorus removal (stream 44 to selector and 46 to digester) and grit removed from the primary sludge (stream 41), were neglected. Bypass streams not used during normal operation (Q18, Q19 and Q20) were not considered either.

Based on the incidence matrix, 32 linear mass balances were set up (Supplementary Material C2). Four main types of mass balances were accounted for, describing the conservation of total flow (Q) and individual mass flows of total phosphorus (*mTP*), chemical oxygen demand (*mCOD*) and total nitrogen (*mTN*) around individual subsystems. The external carbon source (stream 45) and the biogas (stream 43) were reasonably assumed to represent only COD; their total mass flow rates were neglected (in mass balances #2 and #6, respectively). The oxygen required for COD removal (OCcod) and the amount of denitrified nitrogen (DENI) were taken into account in the COD balance of the activated sludge unit (mass balance #18). Note that, the resulting set of mass balances contains all key variables, as required.

The question may arise whether adding mass balances containing off-gas measurements would lead to additional solutions. This will be the case when the added mass balances contain key variables or stay in the same group with other mass balances that contain the key variables. Sampling in the gas phase, however, is typically difficult and associated with a large uncertainty (all the reactors are open and off-gas is dispersed over a large surface area) and significant costs. For these reasons and to limit the complexity of the given example, it was therefore decided not to consider mass balances containing off-gas measurements for demonstrating the experimental design procedure in this study.

Step 3. Data inventory and variable classification

An overview of the initially measured and initially unmeasured data of WWTP Houtrust in terms of flows and concentrations is given in Table 2.

Table 2. Data inventory in terms of flows (*Q*) and concentrations of total phosphorus (*TP*), chemical oxygen demand (*COD*) and total nitrogen (*TN*) for WWTP Houtrust. *mTP*, *mCOD* and *mTN* present mass flow terms.

PFD(*)	Short Name	*Q*			*TP*		*mTP*	*COD*		*mCOD*	*TN*		*mTN*
		m	*c*	*σ*	*m*	*c*	*σ*	*m*	*p*	*σ*	*m*	*p*	*σ*
4	WWTP influent	1	11	2,000	1	75	20	1	35	1500	1	75	40
5	Reject water	1	11	100	1	75	35	1	35	900	1	75	60
7	Settled influent		11	2,000		75	35		35	100		75	70
15	Inflow secondary clarifiers	1	11	3,000		75	15	1	35	2000	1	75	35
17	WWTP effluent		11	2,000	1	75	10	1	35	150	1	75	50
23	Return activated sludge	1	11	2,000		75	15	1	35	1500		75	50
26	Waste activated sludge	1	11	15		75	15	1	35	550		75	30
27	Thickened WAS		11	50		75	65		35	1500		75	100
28	Primary sludge		11	100		75	40		35	1500		75	45
31	Thickened primary sludge		11	15		75	20	1	35	600		75	50
34	Digested sludge		11	50		75	80	1	35	1200		75	35
36	WWTP waste sludge	1	11	15	1	75	80	1	35	2000	1	75	40
37	Centrate WAS thickening		11	50		75	35		35	600		75	25
38	Centrate dewatering		11	50		75	55	1	35	400		75	30
39	Overflow primary thickener	1	11	100		75	90		35	300		75	25
43	Biogas	1	11	100	1	75	2	1	35	200	1	75	2
45	External carbon source	1	11	2	1	75	2	1	35	150	1	75	2

(*) Stream number in process flow diagram (Figure 2).
m = indicating whether this flow/concentration variable is initially measured (1) or not (empty);
σ = estimated error of the corresponding mass flow of the measurements (standard deviation of the mean, used in Step 7);
c = weighing factor represents the cost of a single measurement.
Unit: flow and concentration = $m^3.day^{-1}$ and $g.m^{-3}$; mass flow: kg. day^{-1}

Errors of the measurement or the standard deviations of the mean measurements of all variables (in terms of total and individual mass flow) were estimated based on previous monitoring campaign (Meijer et al., 2015). From initial data, variables were classified into 4 groups: initial measured variables (M), initial unmeasured variables (U), unmeasurable variables (X) and potential additionally measured variables (P_a) (Table 3).

Table 3. Variable classification

	Description	**Corresponding variables**
M	Initially measured variables ($n_m = 34$)	*Q4, Q5, Q15, Q23, Q26, Q36, Q39, Q43, Q45,* *TP4, TP5, TP17, TP36, TP43, TP45,* *COD4, COD5, COD15, COD17, COD23, COD26, COD31, COD34, COD36, COD38, COD43, COD45,* *TN4, TN5, TN15, TN17, TN36, TN43, TN45*
U	Initially unmeasured variables ($n_u = 34$)	*Q7, Q17, Q27, Q28, Q31, Q34, Q37, Q38,* *TP7, TP15, TP23, TP26, TP27, TP28, TP31, TP34, TP37, TP38, TP39,* *COD7, COD27, COD28, COD37, COD39,* *TN7, TN23, TN26, TN27, TN28, TN31, TN34, TN37, TN38, TN39*
X	Unmeasurable variables ($n_X = 4$)	*Q28* *TP28* *COD28* *TN28*
P_a	Potential additionally measured variables ($n_p = 30$)	*Q7, Q17, Q27, Q31, Q34, Q37, Q38,* *TP7, TP15, TP23, TP26, TP27, TP31, TP34, TP37, TP38, TP39,* *COD7, COD27, COD37, COD39,* *TN7, TN23, TN26, TN27, TN31, TN34, TN37, TN38, TN39*
K^*	Key variables ($n_K = 31$)	*Q4, Q5, Q7, Q17, Q26, Q28, Q36* *mTP4, mTP5, mTP7, mTP17, mTP26, mTP28, mTP36*

		mCOD4, mCOD5, mCOD7, mCOD17, mCOD26, mCOD28, mCOD36, mCOD43, mTN4, mTN5, mTN7, mTN17, mTN26, mTN28, mTN36, DENI, OCcod

337 *OCcod = required oxygen for COD removal; DENI = denitrified nitrogen*

338 While the classification of variables and the measurement cost quantification are rather straightforward, the
339 estimation of the measurement accuracy may be more difficult. Any expert knowledge and/or information
340 from previous monitoring campaigns is most useful in this respect. Keeping in mind that the relative
341 magnitude of the error terms is more important than their absolute values, it is interesting to note that, e.g.,
342 the error term on the volumetric mass flow of the influent (Q4) is of the same magnitude as the error term on
343 its COD mass flow (mCOD4), on its turn being one magnitudes higher then COD mass flow in the effluent
344 (mCOD17).

345 **Step 4. Feasibility evaluation**

346 The feasibility evaluation for the WWTP Houtrust confirmed that the identification of key variables is feasible,
347 at least in the case that all potential additionally measured variables (all variables in P_a) are measured
348 additionally. However, the initial data were not sufficient to identify all key variables. Therefore, the procedure
349 is continued to find all sets of additional measurements that allow the identification of key variables and
350 select the optimal solutions in terms of cost and accuracy.

351 **Step 5. Clustering and variable reclassification**

352 The redundancy equations were set up and analysed in view of clustering (Supplementary Material C3). A
353 first group of redundancy equations contains only variables in terms of flows (equations #1-8 in), a second
354 group express the relations between total phosphorus loads (equations #9-16). A third group of redundancy
355 equations (equation #17-32) contains variables from both the *COD* and nitrogen balances; they can be used
356 to identify both *mCOD* and *mTN* variables. The *COD* and total nitrogen balances need to be considered
357 together because they are related through the amount of denitrified nitrogen, *DENI*.. Consequently, the mass
358 balances were also clustered into three corresponding groups.

359 Variable classification was retaken for each group (Table 4). Each group has its own measured variables (M),
360 unmeasured variables (U), potential additionally measured variables (P_a), unmeasurable key variables (X),
361 and key variables (K^*) that appear in the set of (overlapping) mass balances of that group. Consider, for
362 example, the group of overlapping mass balances of flow *Q* (Supplementary Material C3, mass balances #1-
363 8). In this group, seven key variables K^* need to be identifiable are flow measurements: WWTP influent (Q4),

reject water (*Q5*), settled influent (*Q7*), WWTP effluent (*Q17*), waste activated sludge (*Q26*), primary sludge (*Q28*) and waste sludge (*Q36*). Their identifiability needs to be checked for all subsets of potential additionally measured variables P_a = (*Q7, Q17, Q27, Q31, Q34, Q37, Q38*) in this case being 2^7 = 128 (with 7 the number of elements in P_a of this group) .

For the group of total phosphorus mass balances (Supplementary Material C3, mass balances #9-16), there are seven key variables K^*, namely, the total phosphorus mass flow in the influent (*mTP4*), reject water (*mTP5*), settled influent (*mTP7*), WWTP effluent (*mTP17*), waste activated sludge (*mTP26*), primary sludge (*mTP28*) and waste sludge (*mTP36*). Their identifiability needs to be checked for all subsets of potential additionally measured variables P_a = (*Q7, Q17, Q27, Q31, Q34, Q37, Q38, TP7, TP15, TP23, TP26, TP27, TP31, TP34, TP37, TP38, TP39)* in this case being 2^{17} = 131,072 (with 17 the number of elements in P_a).

Analogously, variable classification was applied to the group of chemical oxygen demand and total nitrogen balances. Note that, as the volumetric flows Q contribute to all individual mass flow terms, they are part of potential additionally measured variables of each group (Table 4).

Table 4. Variable classification for each group of overlapping mass balances. n_m, n_u, n_X, $n_p$and n_K represent the number of measured variables M, unmeasured variables U, unmeasurable key variables, potential additionally measured variables P_a, key variables K^*

	Description	**Group of overlapping mass balances**		
		Flow *(Q)*	Total phosphorus (*TP*)	Chemical oxygen demand and total nitrogen (*COD & TN*)
M	Set of measured variables	*Q4, Q5, Q15, Q23, Q26, Q36, Q39, Q43, Q45*	*Q4, Q5, Q36, Q43, Q45 TP4, TP5, TP17, TP36, TP43, TP45*	*Q4, Q5, Q15, Q23, Q26, Q36, Q39, Q43, Q45 COD4, COD5, COD15, COD17, COD23, COD26, COD31, COD34, COD36, COD38, COD43, COD45, TN4, TN5, TN15, TN17, TN36, TN43, TN45*
	$(n_m = 34)$	$(n_m = 9)$	$(n_m = 11)$	$(n_m = 28)$
U	Set of unmeasured variables	*Q7, Q17, Q27, Q28, Q31, Q34, Q37, Q38*	*Q7, Q17, Q27, Q28, Q31, Q34, Q37, Q38 TP7, TP15, TP23, TP26, TP27, TP28, TP31, TP34, TP37, TP38, TP39,*	*Q7, Q17, Q27, Q28, Q31, Q34, Q37, Q38 COD7, COD27, COD28, COD37, COD39, TN7, TN23, TN26, TN27, TN28, TN31, TN34, TN37, TN38, TN39*
	$(n_u = 34)$	$(n_u = 8)$	$(n_u = 19)$	$(n_u = 23)$
X	Set of unmeasurable key variables	*Q28*	*Q28 TP28*	*Q28, COD28, TN28*
	$(n_X = 4)$	$(n_X = 1)$	$(n_X = 2)$	$(n_X = 3)$
P_a	Set of potential additionally measured variables	*Q7, Q17, Q27, Q31, Q34, Q37, Q38*	*Q7, Q17, Q27, Q31, Q34, Q37, Q38, TP7, TP15, TP23, TP26, TP27, TP31, TP34, TP37, TP38, TP39,*	*Q7, Q17, Q27, Q31, Q34, Q37, Q38 COD7, COD27, COD37, COD39, TN7, TN23, TN26, TN27, TN31, TN34, TN37, TN38, TN39*
	$(n_p = 30)$	$(n_p = 7)$	$(n_p = 17)$	$(n_p = 20)$

Description	Group of overlapping mass balances		
	Flow *(Q)*	Total phosphorus (*TP*)	Chemical oxygen demand and total nitrogen (*COD & TN*)
Set of key variables K^* $(n_K = 31)$	*Q4, Q5, Q7, Q17, Q28, Q26, Q36* (*) $(n_K = 7)$	*mTP4, mTP5, mTP7, mTP17, mTP26, mTP28, mTP36* $(n_K = 7)$	*mCOD4, mCOD5, mCOD7, mCOD17, mCOD26, mCOD28, mCOD36, mCOD43, OCcod, mTN4, mTN5, mTN7, mTN17, mTN26, mTN28, mTN36, DENI* $(n_K = 17)$

(*) Key variables expressed in volumetric flows are directly equivalent to key variables in total mass flows as the same density is assumed for all streams.

Overall, three distinct groups of overlapping mass balances and associated groups of variables were determined: the flow (*Q*), the mass of total phosphorus (*mTP*) and the combined group of mass of chemical oxygen demand (*mCOD*) and mass of total nitrogen (*mTN*). Each group of mass balances can be effectively used to identify variables that appear in that group – only those and no other ones.

Step 6. Finding solutions

The determination of sets of additional measurements that guarantee the identification of key variables was performed separately for each group of overlapping mass balances and the obtained results were merged subsequently.

For instance, the set of overlapping mass balances for total phosphorus contains seventeen potential additionally measured variables ($n_p = 17$, Table 4), corresponding to 2^{17} = 131,072 subsets (combinations of variables) of P_a to be analysed. By applying the algorithm (Supplement Material A3), 337 out of 131,072 subsets of P_a were found as the solutions A allowing the identification of key variables K^* (Table 4) of this group. Similar interpretation can be done for other groups.

Since the key variables of all groups need to be identifiable simultaneously, 80,004 overall solutions A were derived by combining the solution vectors of one group to the ones of others, considering all possible combinations.

A non-clustering approach, analysing all possible combinations of initially unmeasured variables and the complete set of mass balances, without distinguishing between groups – essentially skipping Step 5 - was also performed for comparison. The results are summarized in Table 5.

Table 5. Summary of solution of clustering and non-clustering approach.

Group	n_p [(1)]	possible subsets	Number of solution[(2)]	Execution

			of P_a (2^{n_p})	A_z	A	time[3]
Clustering	*Q*	7	128	100		
	TP	17	131,072	337	80,004	47 s
	COD & TN	20	1,048,576	200		
Non-clustering		30	1,073,741,824		80,004	7486 s

(1) n_p is number of potential additionally variables in terms of flow and concentration
(2) A_z is solutions for each group of overlapping mass balances
A is final solution after combining solutions of individual groups (duplicates were removed)
(3) Procedures were implemented by using Matlab 2014a on desktop CPU i7-4770, RAM 8GB.

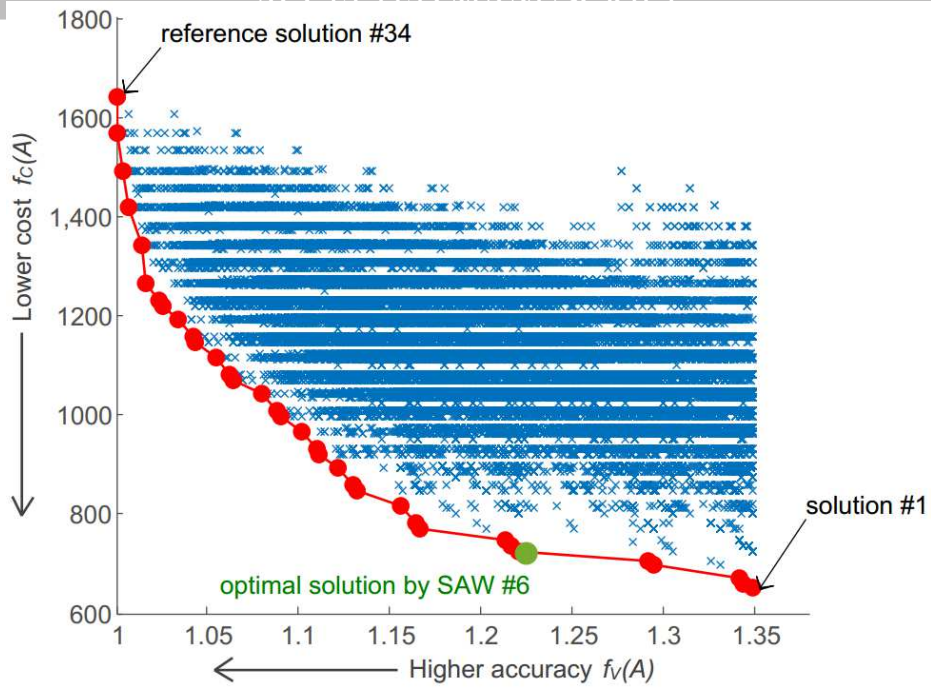
The total number of subsets to be analysed (total number of P_a of each group) in the clustering approach amounted to *1,179,776 (= 128 + 131,072 + 1,048,576*), compared to all 2^{30} = 1,073,741,824 subset of P_a in non-clustering approach (Table 5). It is clear that clustering significantly reduced computational effort, which enables the finding solutions to perform much faster, in this case by a factor of about 150 (47s versus 7486s).

The more potential additionally measured variables the system has, the greater advantage of clustering will be. For example, in case of 20 initially measured variables and 40 potential additionally measured variables (compared to 30 initially measured variables and 30 potential additionally measured variables in the presented case study), the number subsets of P_a to be checked in the non-clustering approach would be 2^{40} (about 1 x 10^{12}). With an average speed of analysing of 150,000 subsets/s with available computational resources, it would take about 80 days for non-clustering approach to solve the problem, while the clustering approach took about 2 hours to complete. The execution time for finding solution greatly depends on the number of initial measurements and the number of key variables.

From the 2^{30} = 1,073,741,824 combinations (subsets) of additional measurement analysed, 80,004 of them, i.e. a fraction of less than 10^{-4}, were found to be solutions that will allow key variables to be identified.

Step 7. Optimization

The cost and accuracy objective functions were calculated for all 80,004 solutions and are displayed in Figure 3. The Pareto-optimal front is also visualized, containing thirty-four (34) optimal solutions. For these Pareto-optimal solutions, a lower cost can only be obtained at the expense of a lower accuracy and vice versa, a higher accuracy can only be obtained at the expense of a higher cost. The specifications of the Pareto-optimal solutions are listed in Supplementary Material C4.



427

428 **Figure 3.** Solutions A are expressed in terms of cost $f_C(A)$ (the lower, the better) and accuracy $f_V(A)$ (the
429 lower the value, the higher accuracy of the solution or smaller variance of new estimate of key variables).
430 Each x represents a solution; the line with filled circles (red) represents the Pareto-optimal front, containing
431 all optimal solutions. The green filled circle denotes the optimal solution #6 selected by the simple additive
432 weighting method (SAW), see Supplementary Material C4 (for interpretation of the references to colour in
433 this figure, the reader is referred to the web version of this article).

434 The most accurate (but also most expensive) Pareto-optimal solution is the reference solution #34, for which
435 all 30 potential additionally measured variables P_a are measured additionally. The reference solution is
436 characterized by an accuracy $f_V(A)$ = 1.00 and cost $f_C(A)$ = 1642. The cheapest and least accurate Pareto-
437 optimal solution is solution #1 with *a* = 14, $f_V(A)$ = 1.35 and $f_C(A)$ = 650. An accuracy of 1.35 means that the
438 average variance ($f_V(A)$, see Eq. 2) identified through this solution is 35% higher than the lowest possible
439 variance, i.e. that of the reference solution and the cost of 650 is the total cost of 14 additional
440 measurements. An analogous interpretation holds for the other solutions.

441 From the 34 Pareto-optimal solutions, the user can select a favourite one. For instance, applying the additive
442 weighting method (SAW) results in optimal solution #6 (green-filled circle, Figure 3), requiring *a* = 15
443 additional measurements and characterized by an accuracy $f_V(A)$ = 1.22 and cost $f_C(A)$ = 725. While a
444 minimum number of 14 additionally measured variables is required to have enough redundancy to identify all

445 key process variables, the SAW optimal solution only requires one more additional measurement to offer a

446 better accuracy.

4 Discussion

Experimental design procedure in view of data reconciliation for wastewater treatment plants.

This contribution presents an experimental design procedure to determine set(s) of additional measurements, which should be carried out to guarantee the identifiability of key variables, meaning that their value can be calculated from other variables based on available constraints – in this case linear mass balances. The identifiability of key variables is a prerequisite for subsequent data reconciliation, through which the reliable and improved estimates for key variables are obtained. The focus on a predefined (limited) number of key variables is very relevant for monitoring campaigns at WWTPs since typically only a few volumetric flow rates and/or components mass flows should be estimated with high accuracy and high reliability while others are not of interest.

Experimental design for WWTP data collection has been addressed previously, e.g. by Meijer et al. (2015), Puig et al. (2008) and Rieger et al. (2010). In these studies, measurements and/or mass balances were added such that the number of constraints (independent mass balances) was higher than the number of unknown variables, i.e. aiming at an overdetermined system. In this way, redundancy was considered as a "global property" of the system. This approach, however, does not guarantee the identifiability of all specified key process variables, which is required for the subsequent improvement of their estimates through data reconciliation. It also involves the risk of adding measurements without added value in planned monitoring campaigns. Redundancy is indeed not a "global property" but rather is a property of individual variables (van der Heijden et al., 1994a).

In this study, the shortcomings of previous studies (Meijer et al., 2015, 2002; Puig et al., 2008; Rieger et al., 2010) are overcome by unambiguously checking the identifiability of all key variables through the application of redundancy analysis. The feasibility of identifying key variables for the given set of mass balances is checked upfront; mass balances and/or key variables are redefined if needed. The proposed procedure also simplified the set-up of mass balances. In previous studies, it was not always clear to which extent additional mass balances actually provided additional information, i.e. whether they were linearly independent from the previous ones. By applying a feasibility evaluation through redundancy analysis as proposed in this study, one can be confident that the key variables are identifiable for the given set of mass balances.

In this work, redundancy analysis was performed following the method of van der Heijden et al. (1994a) and Klamt et al. (2002). This analysis comprises the set-up of redundancy equations, which are derived by

eliminating unmeasured variables and linear dependencies from the set of mass balances. Graph-based methods (Kretsovalis and Mah, 1988)**,** as applied by Villez et al. (2016) to determine the optimal layout of flow sensors, constitute an alternative way to analyse redundancy. Graph-based method is intuitive (directly related to topology) and may avoid numerical problems in matrix inversion (particularly when dealing with larger and sparse matrices). Nevertheless, the set-up of redundancy equations and mass balances will still be required as they make up a fundamental part of the data reconciliation procedure. In addition, setting up redundancy equations (redundancy matrix R) allows the identification of groups of overlapping mass balances (clustering) and allows quantifying the accuracy by which key variables can be identified (variance matrix V). For all of these reasons, equation-based redundancy analysis is preferred in this work.

Clustering mass balances in groups of overlapping mass balances

In this work, clustering mass balances in groups of overlapping mass balances was proposed for the first time as an essential part of the experimental design procedure. Clustering significantly reduces the computational effort in finding sets of additional measurements that allow the identification of key variable. Solutions are determined independently for each group and the results for individual groups are subsequently combined. This decomposition makes that a much smaller number of sets of potential additionally measured variables need to be analysed. The advantages of clustering are more pronounced as the number of potential additionally measured variables increases. The number of additional measurement layouts to be analysed exponentially increases (2^n) with the increasing number of potential additionally measured variables (n).

In addition, clustering reveals dependencies between variables. The identifiability of variables in one group of overlapping mass balances is independent from the measurement availability of variables in other groups. Therefore, increasing the number of measured variables in one group only helps identifying other variables in the same group. There was not always full awareness of this in previous studies. Moreover, additional measurements of conservative quantities are not always as useful as they were thought to be. For instance, mass flow measurements of total phosphorus, combining measured flow and concentration, are often added to increase system redundancy (Meijer et al., 2015, 2002; Puig et al., 2008). While those measurements increased the number of total phosphorus mass flow variables that could be identified, however, they do not have a direct influence on the identifiability of COD and total nitrogen mass flow variables. An additional measurement of total phosphorus mass flow (flow rate and concentration) could, however, help identifying the key variables in other groups in the coincidental case that the (volumetric) flow rate of the corresponding

stream was not initially measured and corresponds to key variables in other groups (mass flows of COD and total nitrogen) of which the concentrations were already measured. Flow measurements contribute more to the identifiability of key variables than concentration measurements in the sense that they contribute to all mass flows of individual components and thus appear in more groups of overlapping mass balances.

Selecting the optimal solutions among alternatives

Among all solutions, the optimal solutions were found considering two objectives, namely the costs of additional measurements and the accuracy of identified key variables. The results of this multi-objective optimization problem were represented in a Pareto front. It is interesting to note that number of Pareto-optimal solutions is very small compared to total number of solutions (fraction of less than 10^{-3}) and represents an even smaller fraction of the total number of possible combinations of additional measurements (less than 10^{-7}). The Pareto-front is a valuable decision tool from which the user can simply select the preferred optimal solution based on expected accuracy and/or monitoring campaign budget. Alternatively, the trade-off between cost and accuracy could be made based on mathematical methods such as simple additive weighting (SAW), multiplicative exponent weighing (MEW), grey relational analysis (GRA), technique for order of preference by similarity to ideal solution (TOPSIS), etc. (Wang and Rangaiah, 2016).

The Pareto-optimal solutions are guaranteed to be globally optimal because an exhaustive search was applied: (1) all possible combinations (2^{30} in total) of additional measurements were analysed (through redundancy analysis) to find the solutions for the given set of mass balances and given data inventory, and (2) an accuracy $f_V(A)$ and a cost $f_C(A)$ were calculated for every possible solutions (80,004) found under (1) to find the Pareto-front (i.e., a discrete optimization problem).

To maximize the accuracy, this work aims to minimize the average variance of key process variables relative to those of the reference solution (i.e., the solution for which all possible additional measurements are measured additionally, leading to the smallest variance). Other options to maximize accuracy could be to maximize the determinant of the covariance matrix of key variables (D-optimality) or to maximize its minimum eigenvalue (E-optimality). This objective function then needs to be reformulated accordingly.

Application to other WWTPs

The proposed experimental design procedure is simple to apply to other similar WWTPs since it consists of a fixed sequence of steps, all of which are fully explained and documented. Step 1 to step 3 require inputs from the user (for listing key variables, setting up mass balances and inventorying data) following the

guidelines. Step 4 to step 7 are fully automated for any problem that can be formulated in the first 3 steps; these steps do not require user intervention except in case there is one or more key variables that cannot be identified for the given set of mass balances and key variables following the indication of Step 4.

The procedure was described as a retrofitting problem, in which initial measurements are already available and standard error of variables could be estimated/collected easily. The proposed experimental design procedure remains applicable in case no initial measurements are available, e.g. in case of a WWTP in the design phase. In this case, the standard error of the variables need to be estimated relying on expert knowledge, keeping in mind that their relative values are more important than the absolute values.

5 Conclusions

- An experimental design procedure for WWTP is proposed to determine sets of additional measurements, which guarantee that key variables can be identified in the sense that they can be calculated from other measurements and therefore, more reliable and improved estimates of these variables can be found through reconciliation.
- The comprehensive redundancy analysis takes advantage of independent groups of overlapping mass balances to decompose a large system to smaller independent sub-systems, which then significantly reduces computational effort for finding sets of additional measurements that allow the identification of key variables.
- The search for optimal sets of additional measurements is solved as a multi-objective optimization problem involving cost of additional measurements and accuracy of the improved estimates of key variables. The final result is the enumerated Pareto-optimal front of additional measurements, which is valuable for monitoring planning.
- The proposed procedure is straightforward and demonstrated for a case study and can easily be applied to other WWTPs, even if no initial measured data are available.

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616

Highlight

- Step-wise measurement planning procedure to obtain reliable data from wastewater treatment plants (WWTPs)
- Right combinations of measurements guarantee improvement of key variables
- Clustering in groups of overlapping mass balances speeds up calculation
- Optimal solutions are trade-off between measurement cost and variable accuracy
- Procedure is demonstrated for case study and straightforward to apply to other WWTPs