

Article

Mapping the Complex Journey of Swimming Pool Contaminants: A Multi-Method Systems Approach

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Abstract: Swimming pool owners worldwide face the challenging task of keeping their pool water balanced and free from contaminants. However, swimming pool water (SPW) quality management is complex with the countless processes and interactions of interlinked system variables. For example, contamination with sunscreen residues is inevitable as users apply sunscreen to protect their skin from damaging ultraviolet (UV) radiation. Nanoparticulate titanium dioxide (nano-TiO₂) is one such residues that have received criticism due to potential human health and environmental risks. Despite ongoing research studies, management strategies of nano-TiO₂ in swimming pools are still limited. Therefore, this paper focuses on developing a multi-method approach for identifying and understanding interdependencies between TiO₂ particles and an aquatic environment such as a swimming pool. Given the complexity of the system to be assessed, the authors utilise a systems approach by integrating cross-matrix multiplication (MICMAC) and Systems Thinking techniques. The developed conceptual model visually depicts the complex system, which provides users with a basic understanding of swimming pool chemistry, displaying the numerous cause-and-effect relationships and enabling users to identify leverage points that can effectively change the dynamics of the system. Such systems-level understanding, and actions will help to manage nano-TiO₂ levels in an efficient manner. The novelty of this paper is the proposed methodology, which uses a systems approach to conceptualise the complex interactions of contaminants in swimming pools and important pathways to elevated contaminant levels.

Keywords: titanium dioxide; engineered nanoparticles; inorganic UV filter; sunscreen; swimming pool water chemistry; aquatic pollution; fate and behaviour modelling; systems thinking; causal loop diagram; structural analysis-MICMAC



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1. Introduction

Public and private swimming pools are widely used across the world as swimming is a popular activity due to its health benefits; pools are also essential for practicing other water sports like surfing, snorkelling, or canoeing [1–3]. For years, the number of swimming pools installed globally has been on the rise [4], especially since the beginning of the coronavirus pandemic in 2019 [5]. This upward trend has particularly been observed in the private residential swimming pool market in countries such as Australia [6], the USA [5,7], and in the European Union [5,8].

Swimming pool water (SPW) management is complex and requires trained personnel to ensure their compliance with health and safety standards [9,10]. While public swimming pools are well regulated and professionally monitored, this is not the case for private residential swimming pools, which are often poorly maintained due to maintenance costs,

a lack of understanding and knowledge, and the owners' laxity. As a result, the risk of SPW contamination is increased and so is the health risk to pool users [11]. Contaminants can be bather-derived (body fluids, hair, skin, personal care products), maintenance-derived (disinfectants, disinfection by-products, algicides, chemicals to adjust the SPW chemistry) or originate from environmental sources (bird or other animal excretions, dust, soil, leaves, grass) [12,13].

Potential health risks arising from SPW contamination have widely been investigated in the literature [14,15]. However, most studies have focussed on monitoring the levels of common contaminants such as body fluids [16,17], disinfectants and disinfection by-products [18–22], whereas information regarding the SPW contamination by personal care products is rather scarce. Only a few researchers have examined the occurrence and fate of selected personal care products including preservatives [20,23], insect repellents [24], as well as organic [20,25] and inorganic UV filters [26–28]. Yet even these studies focus on explicit fragments of the whole picture, for example, by measuring the total concentration of the contaminant, and fail to identify or account for the complex interplay of factors that may influence the contaminant's fate.

Nanoparticulate titanium dioxide (nano-TiO₂) is commonly used as an inorganic UV filter in sunscreens [29,30], but can also be found in a range of diverse, new and established applications such as photovoltaics, batteries, phytoremediation and photocatalysts, drug delivery systems, self-cleaning and self-sterilising coatings, or as colorant in paint and plastics [31,32]. Recently, debate about the safe usage of nano-TiO₂ has resurfaced with the European Food Safety Authority (EFSA) re-classifying TiO₂ as no longer safe as a food additive (E171) [33]. Nonetheless, nano-TiO₂ is still allowed up to a maximum concentration of 25% in sunscreens [34], even though the risks to humans and the environment following nano-TiO₂ exposure have not yet been conclusively assessed [35,36].

Previous research has shown that nano-TiO₂ can be found in both natural (e.g., estuarial waterways, freshwater bodies) [37–40] and man-made (e.g., swimming pools) [27,28] aquatic environments. However, the challenge of differentiating engineered nano-TiO₂ from naturally occurring particles in nature remains [41]. One way to approach this limitation is by investigating the fate and behaviour of nano-TiO₂ in a relatively small and enclosed environment such as a swimming pool. In particular, private residential swimming pools provide a suitable setting for investigating the variety of interactions occurring between the pool users' behaviour (pool and sunscreen usage), nano-TiO₂ and the chosen ecological system.

Similar to natural systems, this socio-ecological system also comprises many interacting and interdependent factors that change over time. Accordingly, in order to capture these interactions and conceptualise such systems meaningfully, we need to employ tools and techniques that enable closed, interdependent parametrisation of specific environments that are also adaptable and expandable. In this context, a systems approach offers a suitable platform to combine multiple tools and techniques for capturing this complexity [42,43]. Compared to other modelling techniques, a systems approach has clear advantages for providing a holistic understanding of the nano-TiO₂ fate in the environment.

In light of the above discussion, the overarching goal of this research is to develop an integrated conceptual framework to capture the complex dynamics of the interactions between sunscreen derived nano-TiO₂ and SPW using multi-method systems approaches, namely Systems Thinking (ST) [44,45] and Matrice d'Impacts Croisés Multiplication Appliquée à un Classement (MICMAC; Cross-Impact Matrix Multiplication Applied to Classification) [46,47].

2. Method

As discussed above, socio-ecological systems are complex systems characterised by dynamic and nonlinear interdependencies between their social and ecological components. That is, a change in any system component triggers a chain of effects between related variables. To capture the complexity of such systems, in this research, a multi-methods

systems approach was selected to tackle the problem in hand. Figure 1 shows the framework development steps using two systems approach techniques, ST and MICMAC. These steps are discussed in the following sections in greater detail.

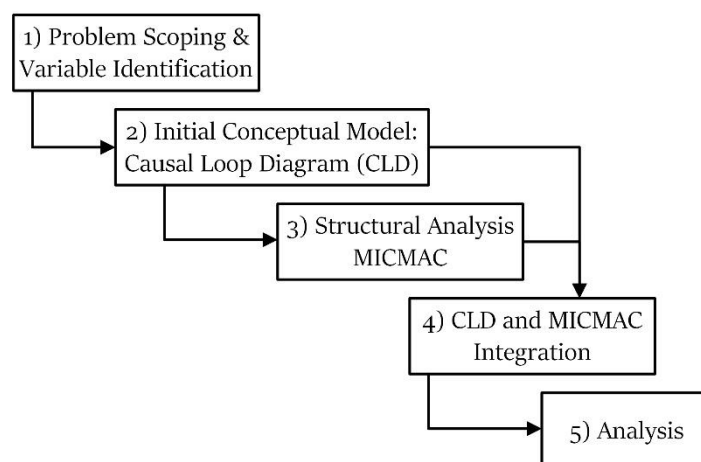


Figure 1. A multi-method systems approach development framework.

While there is no prescribed recipe for successful modelling, this research paper adopts an approach based on some commonly used modelling steps described by Sterman [48] and Voinov [49]. As the statistician George Box famously noted, the important question is not whether models are right or wrong but whether they are illuminating and useful. Therefore, it should be expected that this conceptual framework and any model subsequently built based on this framework can be modified over time as knowledge of the system improves.

2.1. Problem Scoping and Variable Identification

The first step in developing a conceptual framework is to define the overall research objective, thus, characterising the purpose of the model. The second step involves identifying important variables through an extensive literature review and expert workshops. Primary variables are key factors that describe the problem and are critical to telling the story. Further, it is necessary to identify the causal chains of secondary, interacting variables for each primary influencing variable. In this study, the primary variables were identified by individual or limited interdependencies reported in the literature.

2.2. Initial Conceptual Model: Causal Loop Diagram (CLD)

CLDs, or feedback loop diagrams, are a part of the ST toolbox and help to visualise the complex structure of a system. Mapping the formerly identified system variables and their causal links helps to recognise the underlying structure of the system and identify leverage points that enable efficient and effective interventions [50]. The aim when creating a CLD is not necessarily to include all relevant variables that tell the story but rather to reconsider including variables that may not change or change very slowly relative to the defined time horizon of the viewed system, or those that are irrelevant to the overall objective of the system [51], thereby simplifying the causal relationships to key variables." Conversely, excessive omission of variables may result in the misrepresentation of relationships and lack of clarity to the observer [48]. Therefore, when constructing a CLD, the level of detail, physical and temporal offsets or boundaries have to be carefully considered and selectively defined [52].

As shown in Figure 2, causal relations are illustrated in a CLD as arrows between variables, which can either be positive (+) or negative (−). A positive link between cause-and-effect stands for a change in the same direction, which means, if the cause increases (or decreases), the effect increases (or decreases) further to what it would have been [48]. For example, as illustrated in Figure 2, "the higher (or lower) the number of contaminants, the higher (or lower) the concentration of total dissolved solids in the SPW" or "the

higher (or lower) the concentration of total dissolved solids in the SPW, the more (or less) the necessity to drain and replace the SPW". In contrast, a negative link represents a change in the opposite direction. Referring to Figure 2, "the more (or less) the pool owner drains and replaces the SPW, the lower (or higher) the number of contaminants", or "the higher (or lower) the capacity to disinfect or sanitise the SPW, the lower (or higher) the number of contaminants". In some cases, there might be a time delay between cause-and-effect, graphically shown as "//". For example, some contaminants like by-products from intentionally added pool chemicals, bather-introduced substances, or debris, degrade slowly; thus, they gradually add to the level of total dissolved solids. Delays are important to consider as they create a complex dynamic behaviour that is often counter-intuitive and difficult to solve [44,53]. For example, adjusting the chemicals in a pool is challenging as some substances take time to dissolve or react with other constituents, or simply require time to be evenly distributed in the pool.

If a causal relation retrospectively affects the variable of origin, it reflects a closed sequence of cause-and-effect, called a feedback loop [54]. Feedback loops can either be positive (reinforcing, R) or negative (balancing, B) where a positive feedback amplifies the direction of change and a negative feedback constrains or self-corrects change in the system [55]. Each feedback loop influences the overall behaviour of the system, which underlines the importance of identifying feedback to understand the system as a whole [56].

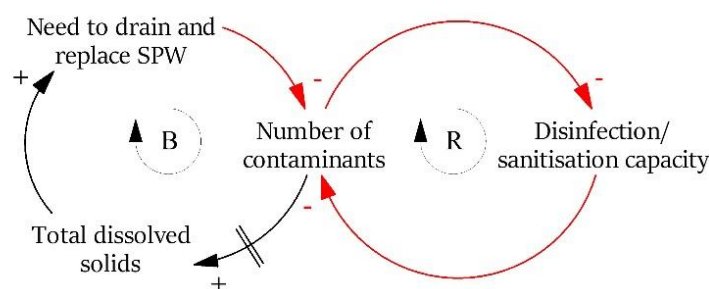


Figure 2. Example CLD with a negative (balancing, B) and a positive (reinforcing, R) feedback loop, and a delay.

2.3. Structural Analysis (MICMAC)

Often, it is convenient to adopt a somewhat informal and haphazard approach to formulating a conceptual framework of systems. The problem with this approach is that it tends to focus on variables and connections with which we are already most familiar. To take a more rigorous approach to the model conceptualisation process, tools like MICMAC are used allowing a more systematic formulation and analysis of a system. Such structural analysis ranks the identified variables or variable groups based on their overall importance (direct and indirect influence and dependence) in the system and determines the strength of their interdependencies [57].

The MICMAC analysis includes the three steps: (1) listing identified primary and secondary variables; (2) describing the relationships between variables by populating a dual entry structural analysis matrix; and (3) identifying key variables that are essential to formulating a conceptual system's framework [58,59].

The aim of the first step is to convert the identified variables from the initial conceptual model (CLD) into a matrix with horizontal and vertical data entries, as exemplarily shown in Figure 3a. In the second step, expert consultations are conducted to quantify the direct relationships between variables by populating a matrix of direct influence (MDI). To assess the direct relationship between a pair of variables, the question to be asked is whether, for example, variable 2 (column) is directly influenced by variable 1 (row). The relationship assessment includes four intensity levels: no influence (0); weak influence (1); moderate influence (2); and strong influence (3), see Figure 3a. The purpose of this process is to rank each variable or variable group based on their influences and dependencies, which aids in identifying the most important variables of the system.

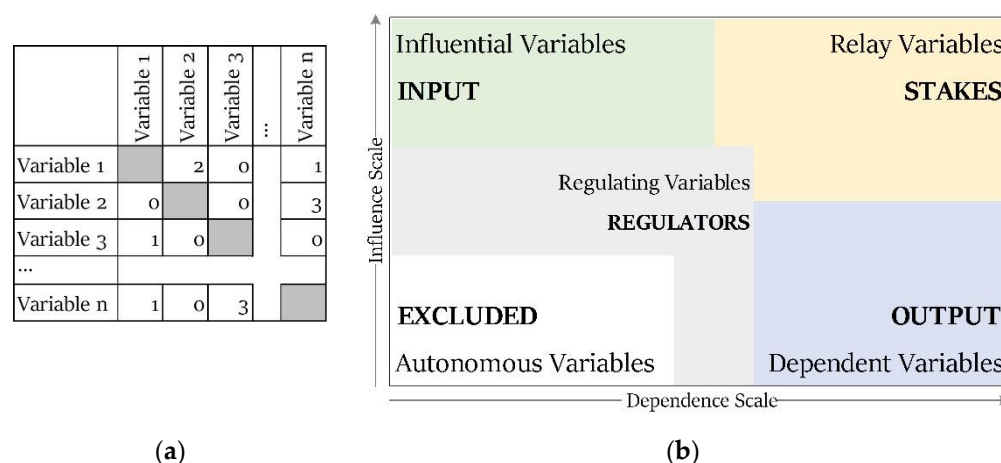


Figure 3. (a) Schematic illustration of a matrix of direct influence; (b) MICMAC influence-dependence map.

In the third and final step, the MICMAC analysis not only identifies the key variables, but also creates (direct and indirect) influence-dependence maps [60] that place the variables into a quadrant depending on their overall influence-dependence rating. The variables are sorted into four categories, as shown in Figure 3b: (1) *Influential (input) variables* are variables that show a minor dependence but a strong influence on other variables and the system as a whole when changing. Therefore, variables in this group are important to consider when discussing systems' leverage points and should be included when developing the system's framework. (2) *Relay variables or stakes* are variables that are both highly influential on the system and also highly dependent on other variables. These variables are considered unstable or dynamic variables as any change may cause feedback through other variables [57]. Hence, they are also referred to as linkage variables. (3) *Dependent variables* represent the system's output, which means they are variables that are highly influenced by other variables and the system. (4) *Autonomous variables* are neither influential nor dependent and are not controlled by the dynamics of the system. These variables are usually excluded from further analysis as they have comparatively weak and limited systems links. Variables that are sitting at the cross-sections of the four quadrants are regarded as *regulating variables or regulators*. These variables cover the averagely influential and/or dependent variables and can control the system to a certain degree but have an overall lower priority in the system [58].

If unsure whether to include or exclude a variable, comparing the variables' placement in the direct and indirect influence-dependence maps may provide a remedy. Overall, a MICMAC analysis helps to investigate the relationships between the variables within a system in a structured manner, and additionally, reduces inherent biases that can occur when reflecting on the system as a whole [59].

2.4. CLD and MICMAC Integration

Both techniques, CLD and MICMAC, have their weaknesses and strengths. While a CLD is a useful tool to tell the system's story and sufficiently displays the causal relationships and interdependencies between system variables, CLDs fail to clearly represent the strength between causal links and do not show the extent to which a variable is important in the system. On the other hand, MICMAC has strong capabilities to determine key variables and quantify the strength of their interactions. However, a significant drawback of this technique is the inability to explicitly show causal links and polarity. Therefore, combining these two techniques provides a more powerful method to draw a clearer picture of the system. The benefit of this integration is that the 'final CLD' includes the strength and polarity between links and reveals the categorisation of the variables based on the influence-dependence map.

2.5. Analysis

The final step of this multi-method systems approach is to identify the leverage points and relevant feedback loops of the system. As D. Meadows stated, ‘*Before you disturb the system in any way, watch how it behaves.*’ ([44], p. 170). Accordingly, leverage points that influence the system effectively can only be found when the system’s behaviour and its underlying structure are understood. As a result, strategies to implement the interventions can be discussed and recommendations made.

3. Results

3.1. Problem Scoping and Variable Identification

The overarching goal of this research was to map the feedback structure of a system that illustrates the causal relations of released nano-TiO₂ from sunscreens in residential private outdoor swimming pools. Primary and secondary variables were identified through a comprehensive literature review [61] and later verified in a workshop with experts. The selected variables cover all aspects that are essential for telling the story of how nano-TiO₂ enters, behaves, and potentially leaves private residential outdoor swimming pools. This includes factors such as nano-TiO₂ and sunscreen properties, human behaviour, pool operating and monitoring parameters, external environmental factors, or the swimming pool chemistry. A detailed variable description can be found in Appendix A, Table A1.

3.2. Initial Conceptual Model: Causal Loop Diagram (CLD)

The initial conceptual model consists of three interlinked CLDs, namely (1) essential swimming pool chemistry, (2) supplementary swimming pool chemistry, and (3) TiO₂ concentration in SPW. The purpose of developing three CLDs, rather than one, combined CLD, was to simplify the modelling process without losing the required level of detail necessary to understand the logic of the causal relations.

The first CLD depicts the components that typically make up private residential outdoor swimming pools (Figure 4), as the behaviour and fate of nano-TiO₂ are significantly governed by the properties of the aqueous environment to which they are exposed. The commonly used Langelier Saturation Index (LSI) that assesses the water balance in swimming pools served as a core component of the CLD. The LSI comprises of five factors (see grey boxes in Figure 4): (1) pH value; (2) water temperature (T); (3) calcium hardness (CaH); (4) total alkalinity (TA); (5) total dissolved solids (TDS); and a correction factor that accounts for the presence of cyanuric acid (CYA) in outdoor pools [62]. The LSI is also expressed as a formula that links the five factors:

$$\text{LSI} = \text{pH} + \text{T} + \text{CaH} + [\text{TA} - (\text{CYA correction factor @ pH})] - \text{TDS} \quad (1)$$

Even though there are numerical values assigned to the factors, the LSI formula was sufficient to provide the polarity of the causal links between the factors and the LSI. The pool water is considered chemically balanced if the LSI is (close to) zero [9]. As shown in Figure 4, below or above the target LSI, the disequilibrium can result in calcite crystal formation, plaster etching, surface fading or calcium carbonate (CaCO₃) scale formation [62,63]. Therefore, the primary goal for pool operators is to keep the LSI close to its target of zero and thus prevent irreparable pool damage or significant (time and money) costs in pool maintenance. Additional chemical equations are included in Appendix A, Table A1.

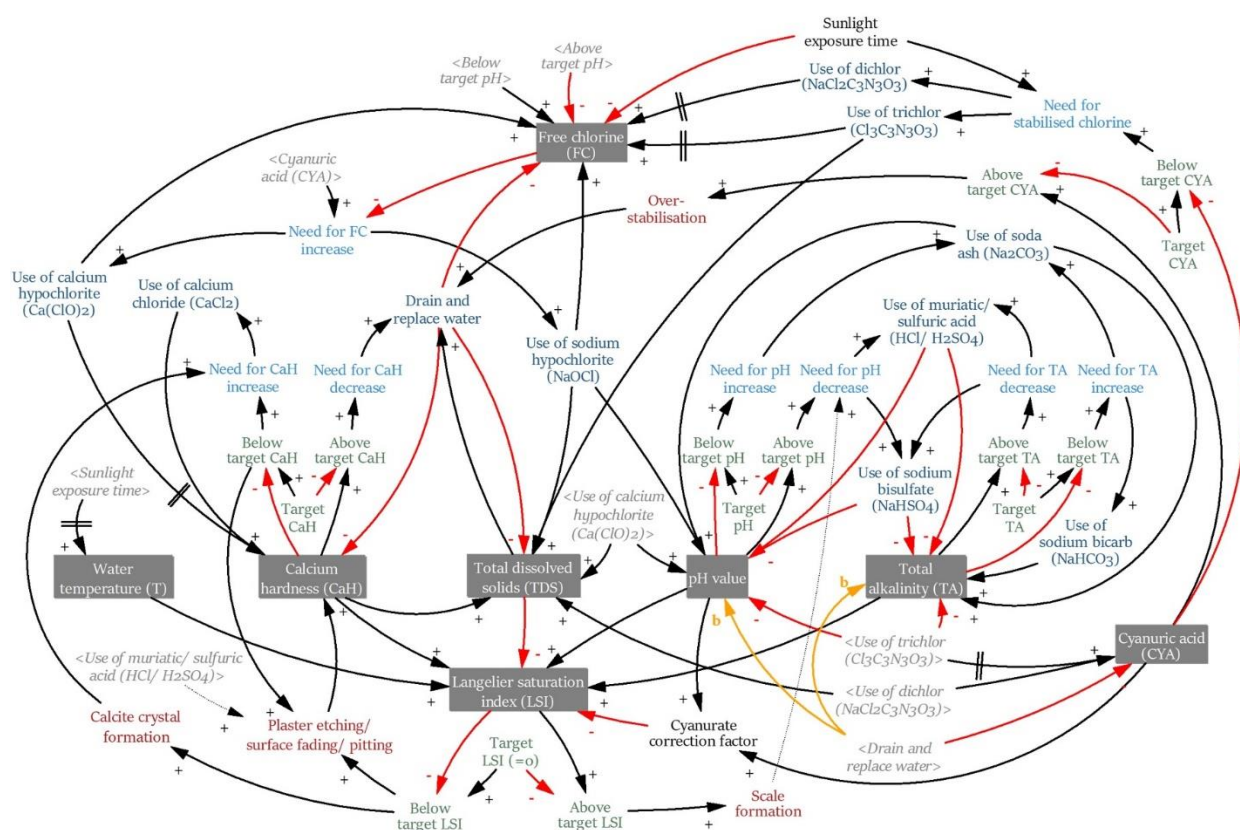


Figure 4. CLD 1: Essential swimming pool chemistry. Colour code (variables): red—SP(W) issues, green—desired state of variable, blue—SP(W) solutions, action to achieve a desired state, grey/italic—‘shadow variable’, i.e., a variable that is described elsewhere in the CLD; and (arrows): yellow (b)—both directional (+ or −). Dotted—link may accidentally be established, but occurrence is unlikely.

The approach to maintaining a balanced LSI may differ, but a good starting point is to keep the controllable factors such as CaH, pH value, or TA, close to their desired ranges (see Figure 4, targets, below and above targets). The following example illustrates their importance. Outdoor swimming pools require CYA (cyanuric acid (CYA)) as a chlorine stabiliser to prevent the photolysis of hypochlorite (OCl^-) and hypochlorous acid (HOCl) [64]. This is achieved by the formation of chlorinated isocyanurates, which are impervious to photolysis [63]. With sunlight (sunlight exposure time) breaking down the free chlorine, the chlorinated isocyanurates (use of trichlor, dichlor) release chlorine over time to compensate for the loss of free chlorine [65]. A CYA concentration range of 15–50 ppm (target CYA) is considered ideal for stabilising free chlorine [66,67]. However, at the same time, using CYA as a chlorine stabiliser substantially reduces the oxidising and sanitising capacity of chlorine [68]. The free chlorine concentration has to be increased to counterbalance the low effectiveness of chlorine when used in conjunction with CYA [69]. Even so, once the CYA level has reached 70 ppm (above target CYA), adding more free chlorine has a negligible effect on the disinfection and sanitisation capability [70]. In other words, once the drawbacks of the reduced disinfection capability of chlorine outweigh the benefits of stabilising chlorine, the swimming pool is considered over-stabilised (over-stabilisation) [71]. The most economical way to fix over-stabilisation is by draining and replacing the SPW (drain and replace water), hence lowering the high CYA level [72].

Mapping the entire “web” of effects resulting from variable adjustments in a CLD assists all levels of users (from researchers to pool service technicians) to understand the complexity with ease compared to traditional explanations based on chemical equations. In this regard, using a CLD does not replace the traditional approach; on the contrary, it

plays a complementary and supporting role when tackling the challenge of regulating the complex pool chemistry.

The second CLD, shown in Figure 5, extends the first CLD by including variables in pool operation (e.g., turnover rate, filtration efficacy) and other chemical additives beyond LSI (e.g., clarifiers, sequestering agents) that are used to solve specific undesirable swimming pool or SPW conditions (e.g., scale formation, contamination). These new variables were displayed in bold to be easily distinguished from variables of the first CLD. Furthermore, a few of the variables used in the first CLD were omitted in the second CLD or included as shadow variables shown in grey and italics to obtain a better overview.

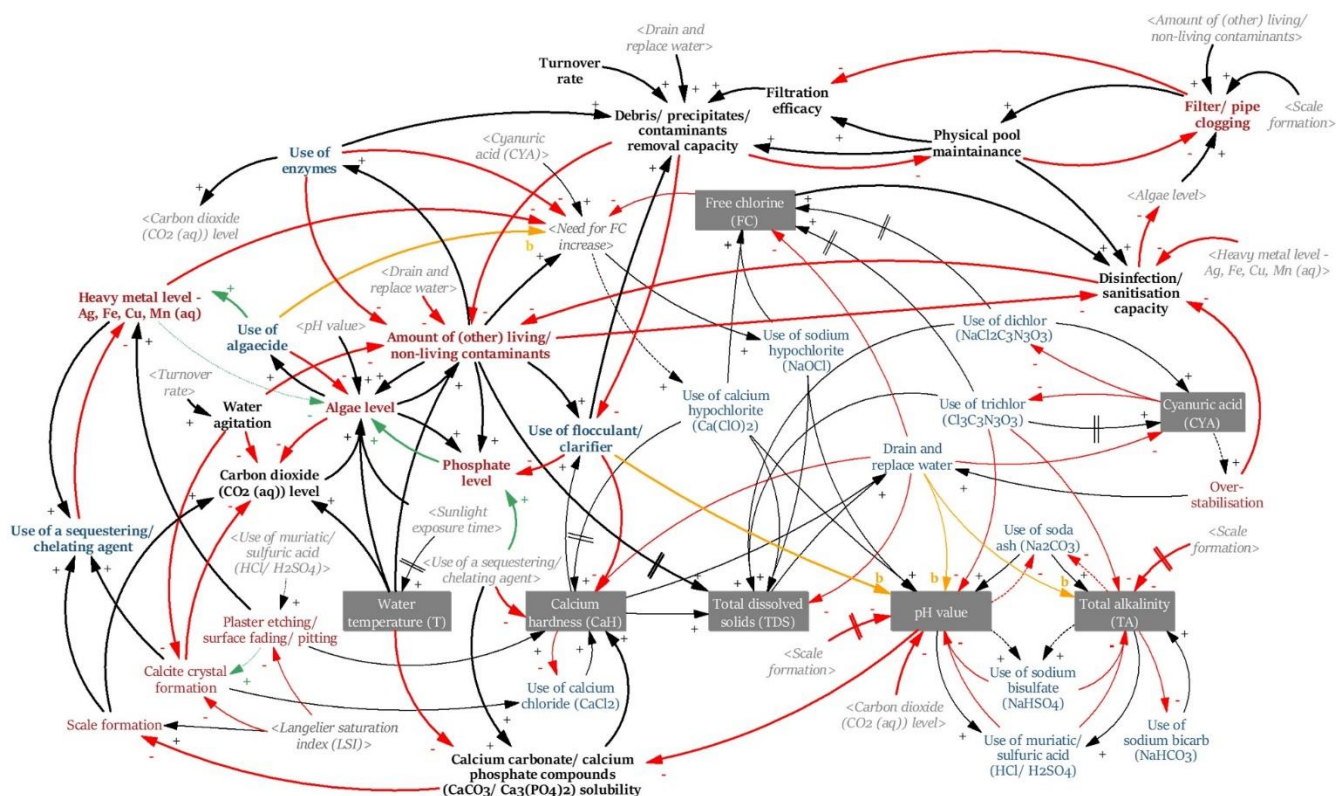


Figure 5. CLD 2: Supplementary swimming pool chemistry. Colour code is the same as in Figure 4 caption, additionally: green arrows represent a situation where either there is a relationship (+ or −) between two variables or no relationship at all, depending on the chemical composition of the affecting variable.

The main foci of the second CLD are (1) to comprehend the occurrence and build-up of various SPW contaminants/pool conditions, (Figure 5, variables displayed in red font), and (2) to explore removal options (displayed in blue). It helps to identify the cause of the problem and explicit intervention points together with their potential benefits and (unintentional) side effects. For instance, algae growth (*algae level*) is a function of *water temperature (T)*, *sunlight exposure time*, *pH value*, *carbon dioxide (CO_{2(aq)}) level*, *phosphate level*, *amount of other living/non-living contaminants*, *disinfection/sanitisation capacity*, and *use of algaecide* [73,74]. Of these, the two obvious options to remove algae and inhibit regrowth are using algaecides (*use of algaecide*) and striving for higher disinfectant concentration levels (*disinfection and sanitisation capacity*). However, algae may still regrow rapidly as these two interventions may not necessarily address the source of the problem and may be outweighed by other positive variables for algal growth. Thus, additional measures to reduce algae growth are required. For example, even though the primary purpose of chlorine (*free chlorine (FC)*) as a disinfectant is to kill pathogens or their hosts (such as algae), 90% of chlorine is used to oxidise organic contaminants (*amount of other living/non-living contaminants*) such as body fluids or personal care products [75]. This directly reduces the

capacity to control algal growth. If the killing rate of the disinfectant becomes too limited, the growth rate of algae can predominate. Thus, instead of increasing the disinfectant concentration to excessive levels, hence unnecessarily escalating costs, removing other organic contaminants through, for example, the *use of enzymes* helps the disinfectant to eliminate algae. Furthermore, after effectively removing algae, depriving the SPW of nutrients such as dissolved carbon dioxide (CO_2 ; *carbon dioxide* ($\text{CO}_2(\text{aq})$) level), nitrates (NO_3^-), or orthophosphates (PO_4^{3-} ; *phosphate level*) is essential to prevent an almost immediate new algae outbreak [76]. This can be achieved by (1) sufficiently agitating the SPW (*water agitation*), which lowers the *carbon dioxide* ($\text{CO}_2(\text{aq})$) level, consequently preventing a rise of the *pH value*; (2) keeping the *pH value* in the recommended range of 7.2–7.8 [11]; and (3) using a phosphate remover (*use of flocculant/clarifier*) [77]. Even though these measures do not help with an existing algae outbreak, they do address the root problem and are, therefore, great for prevention. Using algacides other than *free chlorine* (FC) to kill algae in the pool should be the last resort as they can cause significant health and environmental impacts [12]. In addition, copper-based algacides can cause metal stains or turn the SPW into a clear green solution when copper is oxidised by chlorine [10]. The *use of algacides* also impacts the *need for FC increase*, as, on the one hand, the oxidation of algacide by-products requires more free chlorine, while, on the other hand, algacides reduce the ‘workload’ of free chlorine, hence increasing its effectiveness to kill algae [9].

This example shows how easy it is to mismanage just one particular problem in a swimming pool, as the first course of action is usually to treat the obvious problem regardless of potential side effects, rather than understanding and solving the problem’s origin. Therefore, when tackling a problem, the supplementary pool chemistry CLD (see Figure 5) should also be considered to identify its root cause and to assess the potential risks and unforeseen consequences resulting from an intervention.

Finally, the third CLD, shown in Figure 6, integrates variables related to the sunscreen formulation and TiO_2 particle properties as well as the bathers’ behaviour. Although the main purpose of this CLD is to determine the *total TiO_2 concentration in the SPW*, the CLD also reveals which variables are critical for understanding the input, behaviour, fate, and removal of TiO_2 .

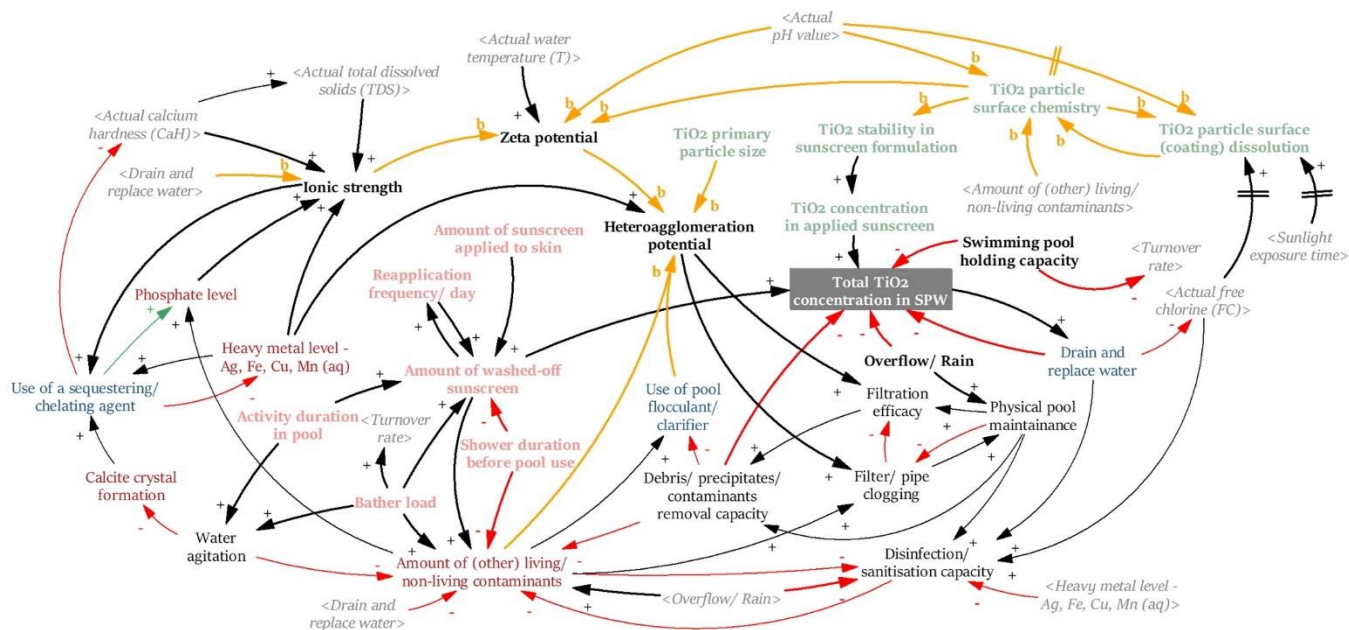


Figure 6. CLD 3: TiO_2 concentration in SPW. Colour code is the same as in the previous two figures. Additionally: pink variables represent the bathers’ behaviour, whereas green variables refer to sunscreen formulation and TiO_2 particle properties.

When developing the CLD, sunscreen usage was assumed to be the only source of TiO_2 found in the SPW. Therefore, the TiO_2 input can be traced back to the variables *TiO_2 concentration in the applied sunscreen* and the *amount of washed-off sunscreen* (see Figure 6). Both variables can be manipulated to a certain degree by either the sunscreen manufacturer (e.g., by adequately stabilising the TiO_2 particles in the formulation (*TiO_2 stability in sunscreen formulation*)) or the bather (e.g., by applying more or less sunscreen to the skin (*amount of sunscreen applied to skin*)). Therefore, the impact of these variables on the *total TiO_2 concentration in the SPW* can be defined and predicted well. However, the situation changes considerably once the TiO_2 particles enter the SPW environment as their behaviour and fate greatly depend on the individual SPW chemistry (e.g., *ionic strength*) [78–80] and TiO_2 particle properties (e.g., *TiO_2 particle surface chemistry*) [81,82] that can additionally change over time (*TiO_2 particle surface (coating) dissolution*) [83,84]. As the behaviour and fate depend on multiple factors, a high number of both directional arrows (+ or –; see Figure 6, yellow arrows (b)) were displayed in the third CLD. Both of these directional arrows indicate that a causal relationship can be established, but the direction of their relationship has to be assessed on a case-by-case basis. Even so, the third CLD reveals which variables impact the *total TiO_2 concentration in the SPW*. The visual presentation of these relationships will be helpful for researchers to design further experimental or modelling investigations.

Furthermore, the third CLD clearly shows that some TiO_2 removal strategies (e.g., *physical pool maintenance*) can reduce the *total TiO_2 concentration in the SPW* irrespective of how strongly variables related to the SPW chemistry and TiO_2 particle properties influence the TiO_2 particle fate and behaviour.

3.3. Structural Analysis (MICMAC)

In the third step of the framework, the MICMAC analysis, the relative importance of identified variables in the system and the strength of their relationships were quantified.

Firstly, the variables to be used in the MICMAC analysis (Table 1) were determined in the initial step of building the conceptual model. To manage the large and extensive set of variables, they were also grouped into eight categories: (1) essential SPW chemistry (monitoring parameters); (2) supplemental SPW chemistry (chemicals); (3) SP/SPW conditions; (4) essential SPW properties; (5) operating parameters; (6) external monitoring parameters; (7) variables related to the bathers' behaviour; and (8) sunscreen formulation/ TiO_2 particle property.

Table 1. List of variables used in the MICMAC analysis.

Category	Variables (Long Label)	Variables (Short Label)
Essential SPW chemistry (monitoring parameters)	Langelier saturation index	LSI
	Water temperature	T
	Calcium hardness	CaH
	Total dissolved solids	TDS
	Total alkalinity	TA
	pH value	pH
	Free (active) chlorine	FC
	Cyanuric acid	CYA
	Carbon dioxide (CO_2) level in water	CO_2 (aq)
Supplementary SPW chemistry (chemicals)	Use of sequestering/chelating agent	SEQ Agent
	Use of flocculant/clarifier	FLOC/CLAR
	Use of algaecide	Algaecide
	Use of enzymes	Enzymes
	Drain and replace water	Replace H_2O

Table 1. Cont.

Category	Variables (Long Label)	Variables (Short Label)
SP/SPW conditions	Over-stabilisation	Over-stab
	Scale formation	Scale
	Calcite crystal formation	Calcite
	Plaster etching/surface fading/pitting	Etching
	Filter/pipe clogging	Clogging
	Heavy metal level	HMetal Lev
	Algae level	Algae
	Phosphate level	Phosphate
	Amount of washed-off sunscreen	WashedOffS
Essential SPW property	Total TiO ₂ concentration in SPW	TiO ₂ concSP
	Amount of (other) non-living/living contaminants	CONTAM
	Calcium carbonate/calcium phosphate compounds solubility	Solubility
	Heteroagglomeration potential	AggloPot
Operating parameter	Ionic strength	IS
	Zeta potential	ZetaPot
	Physical pool maintenance	Maintain
	Turnover rate	Turnover
	Water agitation	Agitation
	Filter efficacy	FilterEffi
Monitoring parameter (external)	Swimming pool holding capacity	SP HCap
	Debris/precipitates/contaminants removal capacity	RemovalCap
	Disinfection/sanitisation capacity	DisinfeCap
Bathers' behaviour	Sunlight exposure time	Sunlight
	Amount of rain/overflow	RainOverfl
	Bather load	Batherload
	Shower duration before pool use	ShowerDur
	Activity duration in swimming pool	ActivDur
Sunscreen formulation/TiO ₂ particle property	Reapplication frequency/day	ReappliSS
	Amount of sunscreen applied to skin	AppliedSS
	TiO ₂ stability (chemical and physical) in sunscreen formulation	TiO ₂ stabSS
	TiO ₂ concentration in applied sunscreen	TiO ₂ concSS
	TiO ₂ primary particle size	TiO ₂ PPS
	TiO ₂ particle surface chemistry	TiO ₂ SChem
	TiO ₂ particle surface (coating) dissolution	TiO ₂ SDis

Secondly, a dual entry structural analysis matrix was used (Appendix B, Table A2) to assess the direct relationship between a pair of variables. Based on this entry, the MICMAC software calculates a value for each variable indicating its strength of influence (or dependence) and then uses the values to rank each variable. The outcome of the variable ranking is shown in Appendix B, Table A3. The variables were grouped into eight categories and sorted based on their global ranking (number in brackets). To rank the groups, the values of all group members were summed up and divided by the number of variables within a group to calculate the strength of each group's influence (or dependence). The resulting ranking is shown in Table 2.

Table 2. Direct and indirect influence and dependency rating of variable groups according to the MICMAC method.

Influence Rank				Dependence Rank			
Direct		Indirect		Direct		Indirect	
1.	Supplementary SPW chemistry (chemicals)	1.	Supplementary SPW chemistry (chemicals)	1.	SP/SPW conditions	1.	Operating parameter
2.	Essential SPW chemistry (monitoring parameters)	2.	Essential SPW chemistry (monitoring parameters)	2.	Essential SPW property	2.	SP/SPW conditions
3.	Monitoring parameter (external)	3.	SP/SPW conditions	3.	Essential SPW chemistry (monitoring parameters)	3.	Supplementary SPW chemistry (chemicals)
4.	SP/SPW conditions	4.	Monitoring parameter (external)	4.	Operating parameter	4.	Essential SPW property
5.	Operating parameter	5.	Operating parameter	5.	Supplementary SPW chemistry (chemicals)	5.	Essential SPW chemistry (monitoring parameters)
6.	Essential SPW property	6.	Essential SPW property	6.	Sunscreen formulation/TiO ₂ particle property	6.	Sunscreen formulation/TiO ₂ particle property
7.	Bathers' behaviour AND Sunscreen formulation/TiO ₂ particle property	7.	Bathers' behaviour	7.	Bathers' behaviour	7.	Bathers' behaviour
		8.	Sunscreen formulation/TiO ₂ particle property	8.	Monitoring parameter (external)	8.	Monitoring parameter (external)

As shown in Table 2, the two most direct and indirect influential categories are the *supplementary* and *essential SPW chemistry*. Variables belonging to either of these categories are important to consider as both categories are very influential and moderately dependent. Hence, all variables of both groups were incorporated in the final CLD. *SP/SPW conditions* is another critical category as variables in this category are quite influential but, on top of that, also highly dependent. Variables in this group were marked as likely to be included in the final CLD. The variables *sunlight exposure time* and *amount of rain/overflow* make up the group *monitoring parameter (external)*, which was ranked in the intermediate range in terms of influence. However, since these parameters are extrinsic in source, the direct and indirect dependence rank of this category was the lowest. The categories *bather's behaviour* as well as *sunscreen formulation/TiO₂ particle property* were placed at the bottom of the list with a very low influence and dependency score. Variables that are part of either group were marked as likely to be excluded in the final CLD.

Finally, the variables were placed in direct (Figure 7) and indirect (Appendix B, Figure A1) influence-dependence maps to visualise their overall influence-dependence rating. Variables that were positioned in any of the three quadrants (1) *influential (input) variables*, (2) *relay variables* or *stakes*, or (3) *dependent (output) variables* were included in the final CLD.

Most variables, however, were positioned in the quadrant *autonomous variables*, which means that they have a limited overall influence-dependence rating. In particular, the variables close to the graphs' origin (lower left corner) are stand-alone variables with weak and a limited number of links to other system variables. Therefore, variables placed in this quadrant were for the most part excluded in the next step of the systems approach development framework.

Using both direct and indirect impact maps, as well as individual variable and variable group scorings, the following variables were excluded from the final CLD: (1) *plaster etching/surface fading/pitting*, (2) *heavy metal level*, (3) *zeta potential*, (4) *turnover rate*, (5) *water agitation*, (6) *swimming pool holding capacity*, (7) *shower duration before pool use*, (8) *activity duration in swimming pool*, (9) *reapplication frequency/day*, (10) *amount of sunscreen applied to skin*, (11) *TiO₂ stability (chemical and physical) in sunscreen formulation*, (12) *TiO₂ concentration in applied sunscreen*, and (13) *TiO₂ primary particle size*. The remaining variables were used to develop the final CLD, as detailed in the following section.

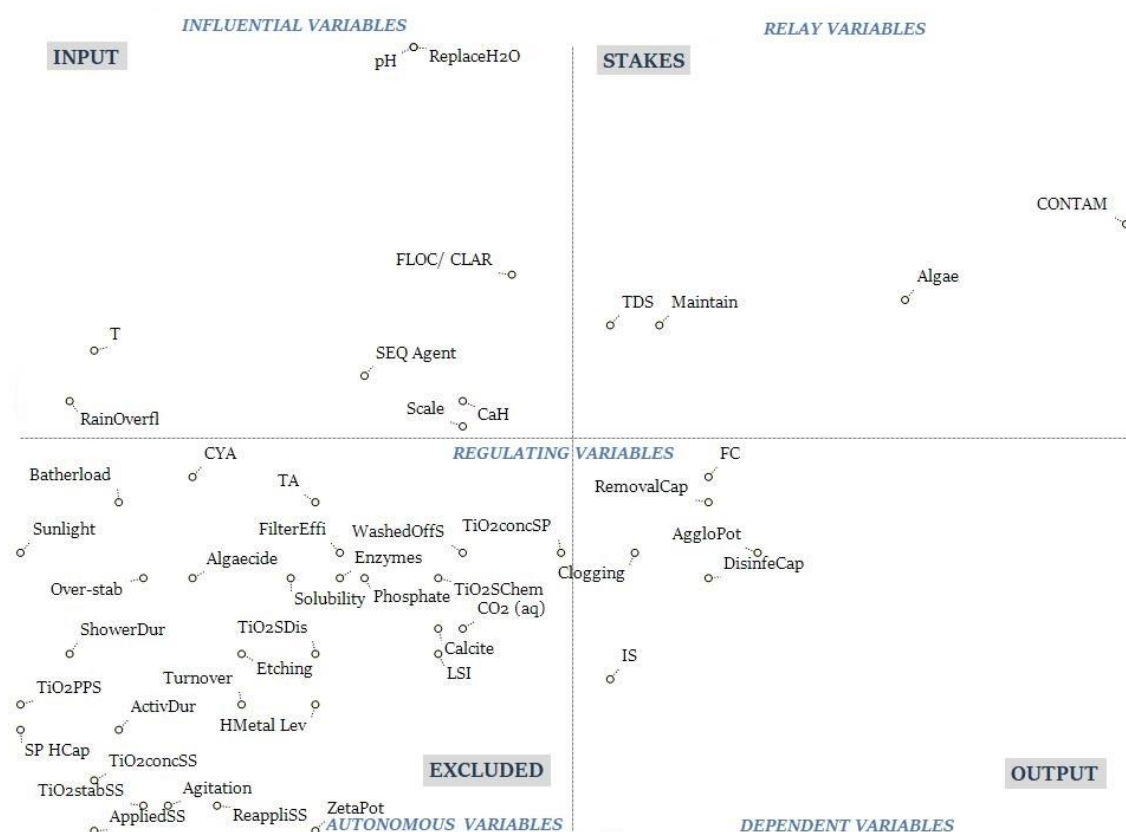


Figure 7. Direct impact map of the model variables, their quadrant allocation and categorisation.

3.4. CLD and MICMAC Integration

In the fourth step of the multi-method systems approach, a final CLD (Figure 8) was developed by integrating the findings from the MICMAC analysis into the initial CLDs. Specifically, reducing the number of variables through a MICMAC analysis allowed the integration of the three initial CLDs into a more focused, final CLD. Furthermore, the variables were regrouped into four categories based on their position in the quadrants of the direct impact map (Figure 7) instead of clustering the variables into eight contextual categories. By doing so, the degree of influence (or dependence) of the variables became obvious at first glance. The overall influence-dependence ranking score was also emphasized by using different font sizes. For instance, the bigger the font size of a variable, the stronger its effect. Similarly, using the structural analysis matrix entry (Appendix B, Table A2) and the overall influence-dependence rating, the weight (i.e., thickness) of the arrows between variables were adjusted to indicate the strength of their relationship. For example, the thicker the arrow, the stronger the cause-effect relationship. These modifications in the final CLD simplified the identification of intervention points to change the dynamics of the system.

For example, as shown in Figure 8, compared to previous CLDs, it is much easier to spot which variables are most effective in removing non-living/living contaminants (CONTAM) from the SPW merely by identifying connecting input variables (green) with bigger font sizes. In this case, the two most viable options are to either drain and replace the SPW (*ReplaceH₂O*) or to use a flocculant/clarifier (*FLOC/CLAR*).

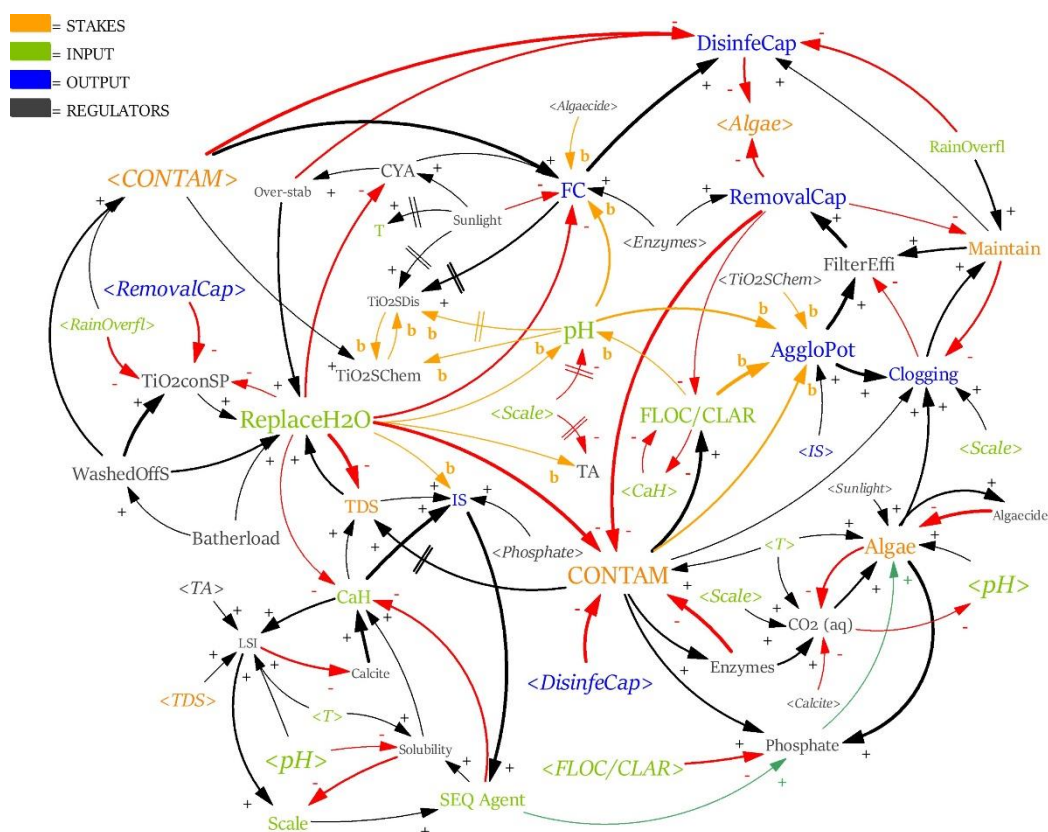


Figure 8. Final CLD: TiO_2 concentration in SPW. Variable colour code is based on the variable placement in the direct impact map (see Figure 7); arrow colour code is the same as in the previous CLDs.

3.5. Analysis

In the final step of the framework, key feedback structures and strategies containing high leverage points were identified and discussed. Figure 9 shows five feedback loops that were extracted from the final CLD (see Figure 8) to explicitly demonstrate the two strategies to manage non-living/living contaminants (CONTAM) in the SPW.

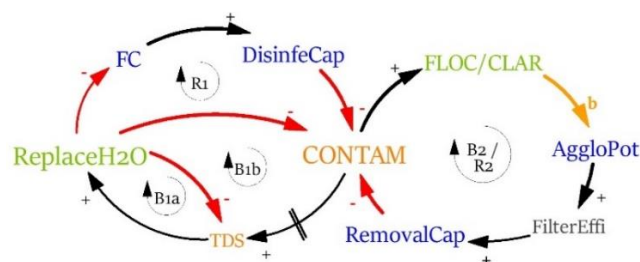


Figure 9. Five feedback loops extracted from Figure 8 illustrating two strategies to remove contaminants in SPW.

The first strategy to remove both living and non-living contaminants is by draining and replacing SPW (*ReplaceH₂O*), which is represented in the two balancing loops (*B1a*, *B1b*) and one reinforcing loop (*R1*). The balancing loop *B1b* consists of three variables (*CONTAM*, *TDS*, *ReplaceH₂O*) and regulates, as intended, the number of contaminants. With contaminants degrading over time, the *TDS* level in SPW rises as well (shown by the delay sign on the arrow between *CONTAM* and *TDS*). Since the only way to effectively remove *TDS* from SPW is by draining and replacing the SPW (*ReplaceH₂O*; see *B1a*), it is advisable to keep contamination levels low in the first place.

Furthermore, as illustrated in the reinforcing loop *R1* (*CONTAM*, *TDS*, *ReplaceH₂O*, *FC*, *DisinfeCap*), this strategy does not necessarily lower the number of contaminants. Without adjusting the SPW chemistry, draining and replacing the SPW (*ReplaceH₂O*) also decreases the free chlorine (*FC*) level, which again reduces the system's disinfection capacity (*DisinfeCap*). Even though the number of contaminants will still be affected, the extent to which the contamination level stagnates, increases or decreases can only be subsequently analysed in a quantitative model. Due to high replacement costs (vast water volume, additional chemicals) and wasting water, the first strategy may not be economically and environmentally the most viable option.

The second strategy to manage living/non-living contaminants (*CONTAM*) is to use a flocculant or clarifier (*FLOC/CLAR*). Depending on the amount and type of flocculant or clarifier used, this is represented by either a balancing (*B2*) or reinforcing (*R2*) loop. For example, negatively charged (anionic) contaminants are attracted by positively charged (cationic) clarifiers (e.g., quaternary ammonium chlorides (*QACs*)), which, if dosed correctly, neutralise the surface charge and thereby coagulate the contaminant (*AggloPot*) [85]. Consequently, the filter efficacy (*FilterEffi*) increases, and so does the debris/precipitates/contaminants removal capacity (*RemovalCap*). Therefore, if used precisely, using a flocculant or clarifier reduces the number of contaminants (*CONTAM*), which is represented in the balancing feedback loop *B2*.

However, if overdosed, the surface charge of the contaminants can reverse, which results in the contaminants repelling each other. Consequently, due to the lower heteroagglomeration potential (*AggloPot*), it is more difficult to filter or vacuum contaminants out of the SPW (*FilterEffi*, *RemovalCap*). Therefore, using more flocculant or clarifier than necessary can even lead to an increase in contamination (*R2*).

4. Conclusions

The overarching goal of this research was to develop an integrated conceptual framework to capture the complex dynamics of the interactions between sunscreen derived nano-TiO₂ and SPW. This was achieved by employing a multi-method systems approach, which combined ST and MICMAC techniques. The framework assembled three CLDs and the MICMAC structural analysis tool to build a final CLD, which was then applied to (1) identify the most influential (or dependent) system variables, (2) visually depict the interactions between identified variables and feedback structures for a broad audience, and (3) quantify the relevance of their relationships. Although it is impossible to predict, let alone control, the TiO₂ fate and behaviour in this complex system, the developed framework will help identify variables that can be used to develop intervention strategies to achieve contaminant mitigation with minimal effort (leverage points).

The proposed framework has implications for both researchers and pool service technicians. For researchers, this novel multi-method approach has not yet been used in this field and provides a novel perspective on nano-TiO₂ fate that can also be adapted for various other water contaminant management issues. Because the research crosses traditional disciplinary lines, it sets a precedent for other domains in chemistry to map complex relationships. This systems approach is viewed as complementary to traditional methods rather than replacing them. Another important characteristic of this framework is that it can address other swimming pool water quality issues, not just pertaining to contaminants. Furthermore, the holistic visualisation of the complex relationships in the form of a CLD could help to identify the focal points for laboratory work and consequently improve the effectiveness of subsequent experiments. For pool service technicians, this developed framework can be used to design more focussed workflow charts and visual maintenance plans that could enhance maintenance strategies, design more efficient filtration systems, and assist with water quality improvement recommendations and related chemical adjustments.

Although the integration of ST and MICMAC is relatively new, it has been used by researchers in various domains as an alternative to the qualitative approach to identi-

ifying “beyond the system” and “within the system” variables and as a way of presenting the interaction between the variables to explain dynamics of a system. For example, Dhirasasna et al. [86] examined the mental wellbeing of PhD students and Omran et al. [87] applied the method in food security and water security domains. The framework is a powerful analytical tool that can be easily used in any other system. However, clearly, like many other methods, the proposed method has also its limitations as this is not a plug-and-play framework. The main limitation of the proposed method is that it does not create predictive patterns as dynamic simulation approaches. However, it generates a great deal of information about the leverage points of the systems.

There are some limitations in this study that could be addressed in future research. First, the study only focused on sunscreens as the source of TiO_2 . Future work should also investigate other potential sources to provide a full picture. Additionally, future studies could shift the focus to the fate and behaviour of other contaminants such as by-products of disinfection processes instead of TiO_2 . Second, this framework only reveals the system as a static picture but does not capture the temporal dynamics of the system. However, by using this causal framework as a base, dynamic or probabilistic simulation models such as System Dynamics can be developed and implemented for scenario analysis and decision-making.

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Appendix A. Variable Descriptions

Table A1. Variable descriptions.

Variable (CLD)	Variable (MICMAC)	Variable Description	Reference
Activity duration in swimming pool	ActivDur	The time swimming pool user (after applying sunscreen) spends in the pool for leisure activities.	
Algae level	Algae	The algal density present in the SPW. Algae are microscopic plant-like organisms that contain chlorophyll and utilise photosynthesis to grow. Rain and wind introduce algae spores into the pool. Algae are typically not pathogenic but can provide an ideal substrate for bacteria. Algae control differs depending on the type of algae present in the SPW.	[9]
Amount of (other) non-living/living contaminants	CONTAM	Includes environmental/human, inorganic/organic contaminants like body oils, sweat, urine, personal care products/cosmetics, dust, dirt, bacteria, and disinfection by-products. Algae and TiO ₂ are also SPW contaminants. However, their interactions are separately described in more detail.	[12]
Amount of rain/overflow	RainOverfl	The amount of rain/excess water that overflows the sides of the pool. Assumption: rain/excess water does not contain TiO ₂ .	
Amount of sunscreen applied to skin	AppliedSS	The amount of sunscreen applied to the skin per person. Assumption: sunscreen contains TiO ₂ .	
Amount of washed-off sunscreen	WashedOffS	The amount of sunscreen residues (including other UV filters and sunscreen constituents like antioxidants, emollients) that are released into the SPW.	
Bather load	Batherload	The maximum number of swimming pool users in the pool during a given amount of time.	
Calcium hardness	CaH	The concentration of dissolved calcium (Ca ²⁺) in the SPW, expressed in ppm.	
Calcium carbonate/calcium phosphate compounds solubility	Solubility	The maximum amount of calcium carbonate (CaCO ₃)/calcium phosphate (Ca ₃ (PO ₄) ₂) that will dissolve in a given amount of SPW at a specific temperature. The exact relationships are more complex as, for example, the carbonate/bicarbonate equilibrium (pH dependent) or interactions with other ions (e.g., Mg ²⁺) in the pool have to be considered.	[63,88]
Calcite crystal formation	Calcite	Calcite crystals (CaCO ₃) form when the SPW is unbalanced (LSI < 0.3) mostly due to a lack of dissolved calcium (Ca ²⁺). Therefore, to increase the concentration of dissolved calcium (Ca ²⁺), the SPW extracts calcium from the pool plaster.	[89]
Carbon dioxide (CO _{2(aq)}) level	CO _{2(aq)}	Dissolved carbon dioxide (CO _{2(aq)}) is in equilibrium with carbonic acid (H ₂ CO ₃). As H ₂ CO ₃ is slightly acidic, the more CO _{2(aq)} , the lower the pH. H ₂ CO ₃ rapidly dissociates to hydrogen (H ⁺), bicarbonate (HCO ₃ [−]), and carbonate (CO ₃ ^{2−}) ions. This combination of alkaline (HCO ₃ [−] , CO ₃ ^{2−}) and acidic (H ⁺) substances helps to control the pH in the SPW (see TA). In addition, due to the equilibrium of dissolved CO _{2(aq)} and atmospheric CO _{2(g)} , CO ₂ will naturally outgas, and hence, cause the pH to rise to approximately 8.2. CO ₂ will leave the SPW until the equilibrium of dissolved CO _{2(aq)} and atmospheric CO _{2(g)} is reached. This causes the pH to rise naturally to approximately 8.2. Similarly, the pH will not rise above 8.2 as the atmospheric pressure will force CO ₂ back into solution (Henry's Law).	[90]

Table A1. Cont.

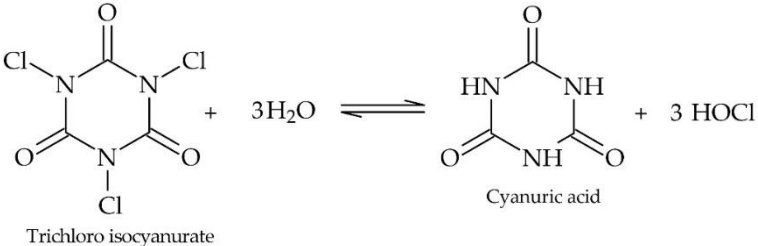
Variable (CLD)	Variable (MICMAC)	Variable Description		Reference
Cyanuric acid (Cyanurate correction factor)	CYA	 <p>Trichloro isocyanurate</p> <p>Cyanuric acid</p>	<p>The cyanuric acid (CYA) concentration in the SPW, expressed in ppm. CYA reduces the photochemical degradation of free chlorine by UV rays. However, using CYA impairs the oxidation capacity of free chlorine by reducing the oxidation-reduction potential (ORP). Also, as CYA forms alkaline ionisation products, the cyanurate alkalinity contributes to the total alkalinity, thus increasing the buffering capacity of SPW.</p>	[63,70, 91,92]
Debris/precipitates/contaminants removal capacity	RemovalCap	The ability to remove debris/precipitates/contaminants from the SPW. Their nature can be of organic or inorganic particulate matter.		
Disinfection/sanitisation capacity	DisinfeCap	The ability of a disinfectant to disinfect/sanitise the SPW. Primarily, disinfectants reduce the number of harmful microorganisms, but they can also oxidise, for example, bath waste.		
Drain and replace water	ReplaceH ₂ O	Drain and replace a specified volume of SPW with fresh tap/well/rainwater.		
Filter efficacy	FilterEffi	The ability of the chosen filtration system (cartridge/sand filters) to remove suspended particulate matter from the SPW. The filter medium, the water flow rate per unit area, regular filter cleaning (i.e., hosing down, soaking in a clean fluid for cartridge filters/backwashing for sand filters), the pore size (cartridge filter)/grade of filter sand size, single or multi-grade beds (sand filter) are key determinants.		[10,12, 13]
Filter/pipe clogging	Clogging	The degree to which the filter media/pipe is clogged through excessive use of flocculants/coagulants, scale/debris build-up. This impairs the water flow rate and filter efficacy.		
Free (active) chlorine	FC	<p>(1a) $\text{NaOCl} + \text{H}_2\text{O} \rightleftharpoons \text{HOCl} + \text{Na}^+ + \text{OH}^-$ Sodium hypochlorite Hypochlorous acid</p> <p>(1b) $\text{Ca}(\text{ClO})_2 + 2\text{H}_2\text{O} \rightleftharpoons 2\text{HOCl} + \text{Ca}^{2+} + 2\text{OH}^-$ Calcium hypochlorite</p> <p>(2) $\text{HOCl} \rightleftharpoons \text{O-Cl}^- + \text{H}^+$</p>	<p>Free (active) chlorine (FC) is a form of chlorine that can act as a sanitiser (and oxidiser) and can be added in various forms to the SPW, for example, as sodium or calcium hypochlorite—see equation (1a) and (1b), respectively. All forms produce HOCl (hypochlorous acid) in water, which is the most desirable, active form of FC. The pH predominantly determines the degree of dissociation of HOCl to H⁺ (hydrogen ion) and OCl[−] (hypochlorite ion, a less effective form of FC) in the SPW, see equation (2).</p>	[10,73]
Heavy metal level	HMetal Lev	The heavy metal level corresponds to the concentration of dissolved heavy metals such as silver (Ag ⁺), iron (Fe ²⁺), copper (Cu ²⁺), and manganese (Mn ²⁺) in the SPW. In high, non-chelated concentrations, these metals can be easily oxidised, for example, by disinfectant products, which can lead to metal stains or turn the colour of the water.		[10]
Heteroagglomeration potential	AggloPot	The heteroagglomeration potential is the likelihood of individual, separated suspended particulate matter to form assemblages. This mechanism can often be induced by adding salts or other chemicals such as coagulants/flocculants.		
Ionic strength	IS	The ionic strength represents the concentration of all ions present in the SPW and is equal to half of the sum of each ion's molar concentration multiplied by their valence squared.		

Table A1. Cont.

Variable (CLD)	Variable (MICMAC)	Variable Description	Reference
Langelier saturation index	LSI	The Langelier Saturation Index (LSI) is a measure to indicate the SPW balance. If perfectly balanced, the LSI is zero. Undersaturated SPW (low LSI) will seek to dissolve calcium from, for example, the pool plaster surface, whereas oversaturated SPW (high LSI) will deposit any form of calcium carbonate (CaCO_3) into the SPW to reduce the amount of dissolved calcium (Ca^{2+}). The LSI can be calculated as $\text{LSI} = \text{pH} + \text{T} + \text{CaH} + [\text{TA} - (\text{CYA correction factor @ pH})] - \text{TDS}$.	[62]
pH value	pH	Abbreviation for “potential or power of hydrogen”. The pH is defined as the negative decadic logarithm of the hydrogen ion activity. The pH value indicates the basicity or acidity of water on a scale from zero (the most acidic) to 14 (the most basic).	[93,94]
Over-stabilisation	Over-stab	Over-stabilisation is the build-up of cyanuric acid (CYA) in SPW that results from the use of chlorinated isocyanurates. As a result, the effectiveness of chlorine in killing pathogens is significantly impaired.	[95]
Phosphate level	Phosphate	The phosphate level comprises various types of organic and inorganic phosphorus compounds. However, most phosphorous compounds will eventually break down and convert to orthophosphates (PO_4^{3-}), which serve as nutrient for algae. Test kits usually only test for PO_4^{3-} as most abundant phosphate type in the SPW.	[96–98]
Physical pool maintenance	Maintain	The physical pool maintenance includes activities such as cleaning (skimming, scrubbing, vacuuming) pool surfaces/filter/pipes as per the manufacturer’s guidelines.	
Plaster etching/surface fading/pitting	Etching	Plaster etching/surface fading/pitting is generally caused by unbalanced SPW or wrong usage of acid products, which results in irreversible damage to the pool.	
Reapplication frequency/day	ReappliSS	The number of sunscreen reapplications per day per person.	
Scale formation	Scale	Scale formation refers to three types of calcium scale deposition resulting from chemically unbalanced SPW, namely calcium carbonate (CaCO_3)—and sometimes calcium phosphate ($\text{Ca}_3(\text{PO}_4)_2$) or calcium sulfate (CaSO_4).	
Shower duration before pool use	ShowerDur	The duration of the shower taken by the pool user before using the pool. Assumption: sunscreen applied by the pool users is partially washed off during the shower.	
Sunlight exposure time	Sunlight	This variable describes how long the SPW is exposed to full sunlight.	
Swimming pool holding capacity	SP HCap	The maximum water holding capacity in a SP.	
TiO ₂ concentration in applied sunscreen	TiO ₂ concSS	The TiO ₂ concentration in the sunscreen that is applied to the user’s skin.	
TiO ₂ particle surface chemistry	TiO ₂ SChem	The TiO ₂ particle surface can either be hydrophilic or hydrophobic depending on the sunscreen type (oil-in-water or water-in-oil).	
TiO ₂ particle surface (coating) dissolution	TiO ₂ SDis	The percentage of the depletion/dissolution of the TiO ₂ particle surface (coating); e.g., aluminium oxide (hydroxide) $\text{Al}_2\text{O}_3/\text{Al}_2(\text{OH})_3$ dissolution caused by chlorine.	[83,84]

Table A1. Cont.

Variable (CLD)	Variable (MICMAC)	Variable Description	Reference
TiO ₂ primary particle size	TiO ₂ PPS	The primary particle size of TiO ₂ particles used in the sunscreen.	
TiO ₂ stability (chemical and physical) in sunscreen formulation	TiO ₂ stabSS	The TiO ₂ particle stability to maintain dispersion in the sunscreen formulation.	
Total alkalinity	TA	The total alkalinity (aka buffering capacity) is the concentration of dissolved alkaline substances, e.g., hydroxide (OH [−]), carbonate (CO ₃ ^{2−}), bicarbonate (HCO ₃ [−]) ions in the SPW.	
Total dissolved solids	TDS	Total dissolved solids are the total amount of dissolved matter, including salts, minerals, metals, and contaminants in the SPW.	[99]
Total TiO ₂ concentration in SPW	TiO ₂ concSP	The total particulate TiO ₂ concentration in SPW.	
Turnover rate	Turnover	The turnover rate refers to the time during which a net volume of SPW passes through the filtration system.	[10]
Use of algaecide	Algaecide	The use of an algaecide refers to the use of natural/synthetic substances used for killing and controlling algae.	
Use of enzymes	Enzymes	The use of enzymes refers to proteins that accelerate the chemical reactions of other substances (e.g., the breakdown of oil or other non-living contaminants) without being used up or altered.	
Use of flocculant/clarifier	FLOC/CLAR	The use of a flocculant or clarifier enhances the agglomeration of suspended particulate matter in the SPW.	
Use of sequestering/chelating agent	SEQ Agent	The use of a sequestering or chelating agent refers to the use of chemicals that control the formation of scale or stains by preventing the precipitations of metal ions.	
Water agitation	Agitation	Water agitation refers to promoting gas exchange, which increases the rate at which oxygen (O ₂) dissolves in but also carbon dioxide (CO ₂) releases from SPW. This can be done purposefully through fountains and springs etc. or caused by the movement of pool users.	
Water temperature	T	The temperature of the SPW.	
Zeta potential	ZetaPot	The zeta potential refers to the electrical potential at the slipping plane, which separates the mobile fluid from the fluid attached to the particle surface. This is an important factor to indicate the stability of a colloidal dispersion.	[100]

Appendix B. MICMAC

Table A2. Variable descriptions.

<div>0 = No influence 1 = Weak influence 2 = Moderate influence 3 = Strong influence</div>																																																				
	1: ActivDur	2: AppliedSS	3: Batherload	4: ReapplSS	5: ShowerDur	6: CaH	7: CO2 (aq)	8: CYA	9: FC	10: LSI	11: pH	12: TA	13: TDS	14: T	15: Solubility	16: AggloPot	17: IS	18: ZaaPot	19: RainOverfl	20: Sunlight	21: RemovalCap	22: DisinfCap	23: FilterEffi	24: Mainain	25: SP HCap	26: Turnover	27: Agitation	28: TiO2concSS	29: TiO2SDis	30: TiO2SChem	31: TiO2PPS	32: TiO2stabsS	33: ReplacedH2O	34: SEQ Agent	35: Algaecide	36: Enzymes	37: FLOC/CLAR	38: Algae	39: CONTAM	40: WashedOHS	41: Calcite	42: Clogging	43: HMetal Lev	44: Over-stab	45: Phosphate	46: Etching	47: Scale	48: TiO2concSP				
1: Activity duration in swimming pool		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	0	0	0	0	1	3	0	0	0	0	0	0	0	0	0			
2: Amount of sunscreen applied to skin	0		0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0	0	0	0	0	0			
3: Bather load	0	0		0	0	0	0	0	1	0	0	0	1	0	0	0	0	0	1	0	0	1	0	1	0	0	2	1	0	0	0	0	0	0	0	0	0	1	1	0	3	3	0	0	0	0	0	0	0	0		
4: Reapplication frequency /day	0	0	0			0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	3	0	0	0	0	0	0	0	0	0			
5: Shower duration before pool use	0	1	0	1		0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	3	0	0	0		0	0	0	0	0			
6: Calcium hardness	0	0	0	0	0		0	0	0	3	0	0	2	0	0	2	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	1	0	0	2	0	0	2	0	0	0	0	1	2	0	0			
7: Carbon dioxide (CO2) level in water	0	0	0	0	0	1		0	0	0	3	2	0	0	1	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	1	0	0	0	0	0	0	0	0	0			
8: Cyanuric acid	0	0	0	0	0	0	0		2	2	1	1	1	0	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	1	0	0	0	1	0	0	0	2	0	0	0	0	0	0	3	0	0	1	0	0		
9: Free (active) chlorine	0	0	0	0	0	0	0	0	0	0	1	0	1	0	0	0	1	1	0	0	0	3	0	1	0	0	0	0	3	0	0	0	0	0	0	1	1	0	2	2	0	0	0	0	0	0	0	0	0	0		
10: Langelier saturation index	0	0	0	0	0	0	0	0			0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0		3	3	0		
11: pH value	0	0	0	0	0	1	2	0	2	3	2	0	0	3	1	1	3	0	0	0	1	0	1	0	0	0	0	0	1	2	0	0	0	0	0	0	1	0	2	0	0	2	0	0	0	1	3	2	0	0		
12: Total alkalinity	0	0	0	0	0	0	1	0	1	3	2	1	0	1	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	1	0	0	1	1	0	0	0	1	0	1	0	0		
13: Total dissolved solids	0	0	0	0	0	0	0	0	0	3	1	2	0	0	0	2	3	0	0	0	1	0	1	0	0	0	0	0	0	0	0	0	0	3	0	0	0	1	0	1	0	2	0	0	0	1	2	0	0			
14: Water temperature	1	0	0	0	0	0	2	0	1	3	1	0	1		2	0	0	1	0	0	1	0	0	0	0	0	0	0	0	0	3	0	0	0	0	0	1	0	3	0	0	1	0	0	0	0	0	1	0			
15: Calcium carbonate/calcium phosphate compounds solubility	0	0	0	0	0	3	1	0	0	0	0	1	1	0		1	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0		1	0	0	0	1	0	3	0
16: Heteroagglomeration potential	0	0	0	3	0	0	0	0	0	0	0	0	0	0		0	0	0	0	0	3	0	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	1	0	0	3	0	0	0	0	0	0			
17: Ionic strength	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3		3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0			
18: Zeta Potential	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0			
19: Amount of rain/overflow	1	1	1	1	0	1	0	1	1	0	1	1	1	0	0	0	1	0		0	1	1	1	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	1	0	1	0	0	1		
20: Sunlight exposure time	1	1	1	1	0	0	0	0	3	0	0	0	0	2	0	0	0	0	0		1	0	0	0	0	0	0	0	2	1	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0			
21: Debris/precipitates/contaminants removal capacity	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0			2	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	3	3	3	0	0	1	0	0	0	0	0	3			
22: Disinfection/sanitisation capacity	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0			0	1	0	1	0	0	0	0	0	0	0	0	1	1	1	1	3	3	0	0	0	0	0	0	0	0	0			
23: Filtration efficacy	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0		1	0	0	0	0	0	0	0	0	1	0	0	0	2	2	2	0	0	1	0	0	0	0	0	2				
24: Physical pool maintenance	0	0	0	0	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	2	3		0	0	1	0	0	0	0	0	0	0	0	0	0	0	2	3	0	1	3	0	0	0	0	1	2			
25: Swimming pool holding capacity	0	0	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0		3	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2			
26: Turnover rate	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0		2	0	0	0	0	0	0	0	0	0	0	0	1	1	0	0	0	0	0	0	0	0	1			
27: Water agitation	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0			0	0	0	0	0	0	0	0	0	0	0	1	1	0	1	0	0	0	0	0	0	0			
28: TiO2 concentration in applied sunscreen	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0		0	0	0	0	1	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	3			
29: TiO2 particle surface (coating) dissolution	0	0	0	0	0	0	0	0	0	0	0	1	0	0	2	1	1	0	0	1	0	0	0	0	0	0	0	0		0	0	3	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0			
30: TiO2 particle surface chemistry	0	0	0	0	0	0	0	0	0	0	0	0	0	0	2	0	3	0	0	1	0	0	0	0	0	0	0	0	3		0	0	3	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0			
31: TiO2 primary particle size	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0	1	0		1	0	0	0	0	0		0	0	1	0	0	0	0	1	0	0	0	0	1	0	0	0	0	0	0				
32: TiO2 stability (chemical and physical) in sunscreen formulation	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0			0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0				

Table A2. Cont.

	1: ActivDur	2: AppliedSS	3: Batherload	4: ReapplSS	5: ShowerDur	6: CaH	7: CO2 (aq)	8: CYA	9: FC	10: LSI	11: pH	12: TA	13: TDS	14: T	15: Solubility	16: AggloPot	17: IS	18: ZeaPot	19: RainOverfl	20: Sunlight	21: RemovalCap	22: DisinfCap	23: FilterEffi	24: Maintain	25: SP HCap	26: Turnover	27: Agitation	28: TiO2concSS	29: TiO2SDis	30: TiO2SChem	31: TiO2PPS	32: TiO2stabSS	33: ReplacedH2O	34: SEQ Agent	35: Algaecide	36: Enzymes	37: FLOC / CLAR	38: Algae	39: CONTAM	40: WashedOffS	41: Calcite	42: Clogging	43: HMetal Lev	44: Overstab	45: Phosphate	46: Etching	47: Scale	48: TiO2concSP	
0 = No influence 1 = Weak influence 2 = Moderate influence 3 = Strong influence																																																	
33: Drain and replace water	0	0	0	0	0	2	0	3	3	0	1	1	3	1	1	0	1	0	0	0	2	1	0	0	0	0	0	0	0	0	0	0	0	1	1	1	1	1	3	0	0	0	1	2	1	0	0	3	
34: Use of a sequestering/chelating agent	0	0	0	0	0	3	0	0	0	0	1	0	1	0	2	0	3	0	0	0	1	0	0	0	0	0	0	0	0	1	1	0	0	0	0	0	0	0	0	0	2	0	3	0	1	0	2	0	
35: Use of algaecide	0	0	0	0	0	0	0	0	2	0	1	0	1	0	0	0	1	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	3	1	0	0	0	2	0	0	0	0	
36: Use of enzymes	0	0	0	0	0	0	2	0	2	0	0	0	0	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	0	1	1	0	0	0	0	0	0	0	3	1	0	0	0	0	0	0	1	0	
37: Use of flocculant/clarifier	0	0	0	0	0	3	0	0	0	0	1	0	1	0	1	3	1	0	0	0	3	0	0	2	0	0	0	0	0	0	1	0	0	1	0	0	0	0	1	2	0	0	2	1	0	1	0	0	1
38: Algae level	1	0	0	0	0	0	3	0	1	0	0	0	2	0	0	1	0	0	0	0	0	2	0	2	0	0	0	0	0	1	0	0	0	0	3	0	0	0	3	0	0	2	0	0	3	0	0	0	
39: Amount of (other) living/non-living contaminants	0	0	0	0	0	0	0	0	2	0	0	0	2	0	0	1	0	0	0	0	0	3	0	3	0	2	0	0	0	2	0	0	0	0	0	3	3	1	0	0	3	0	0	2	0	0	0	0	
40: Amount of washed-off sunscreen	0	0	0	2	0	0	0	0	1	0	0	0	1	0	0	1	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	3	0	1	0	0	0	0	0	3		
41: Calcite crystal formation	0	0	0	0	0	1	2	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	3	0	0	0	0	0	0	0	0	0	3	0	0	0	0	0	0	1	0	0	0	0	0	0		
42: Filter/pipe clogging	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	3	3	0	0	0	0	0	0	0	0	0	1	0	1	1	1	0	0	0	0	0	0	0	0	1		
43: Heavy metal level	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	1	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	0	0	1	0	0	0	0	0	0	0	0	0		
44: Over-stabilisation	0	0	0	0	0	0	0	3	3	0	0	0	0	0	0	0	0	0	0	0	0	2	0	0	0	0	0	0	0	0	0	0	3	0	0	0	0	1	1	0	0	0	0	0	0	0	0		
45: Phosphate level	0	0	0	0	0	0	0	0	1	0	0	0	2	0	0	1	2	0	0	0	1	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	2	3	0	0	0	0	0	0	0	0	0		
46: Plaster etching/surface fading/pitting	0	0	0	0	0	2	2	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	3	0	2	0	0	0	0		
47: Scale formation	0	0	0	0	0	1	2	0	0	0	1	2	0	0	0	0	1	0	0	0	0	0	1	2	0	0	0	0	0	0	0	0	0	3	0	2	0	0	0	0	3	0	0	1	0	0	0		
48: Total TiO2 concentration in SPW	0	0	0	0	0	0	0	0	0	0	1	0	0	0	2	0	0	0	0	0	1	1	1	0	1	0	0	0	0	0	0	0	2	0	0	0	1	0	2	0	0	1	1	0	0	0	0		

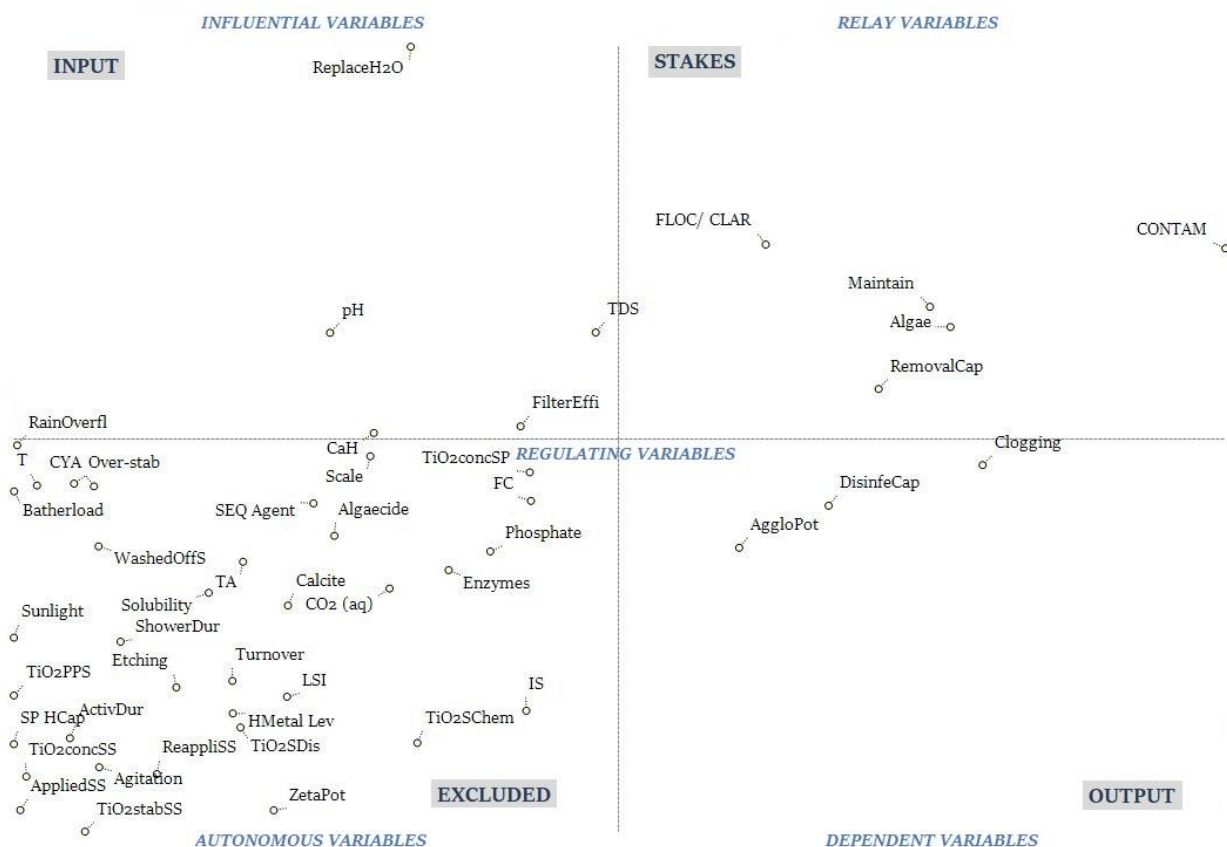


Figure A1. Indirect influence matrix of the model variables, their quadrant allocation and categorisation.

Table A3. Direct and indirect influence and dependency rating of all variables according to the MICMAC method—variables are ranked within each category as well as overall, that is compared to all 48 variables (shown in parentheses).

Category	Influence Rank		Dependence Rank	
	Direct	Indirect	Direct	Indirect
Essential SPW chemistry (monitoring parameters)	1. pH (1) 2. TDS (6) 3. T (8) 4. CaH (10) 5. CYA (13) 6. FC (14) 7. TA (16) 8. CO ₂ (aq) (31) 9. LSI (34)	1. TDS (6) 2. pH (7) 3. CaH (10) 4. T (16) 5. CYA (17) 6. FC (19) 7. TA (26) 8. CO ₂ (aq) (28) 9. LSI (36)	1. FC (4) 2. TDS (9) 3. CaH (13) 4. CO ₂ (aq) (14) 5. LSI (17) 6. pH (20) 7. TA (26) 8. CYA (34) 9. T (42)	1. TDS (9) 2. FC (10) 3. CO ₂ (aq) (18) 4. CaH (19) 5. pH (22) 6. LSI (25) 7. TA (27) 8. CYA (37) 9. T (41)
Supplementary SPW chemistry (chemicals)	1. ReplaceH ₂ O (2) 2. FLOC/CLAR (4) 3. SEQ Agent (9) 4. Algaecide (27) 5. Enzymes (28)	1. Replace H ₂ O (1) 2. FLOC/CLAR (2) 3. SEQ Agent (20) 4. Algaecide (22) 5. Enzymes (27)	1. FLOC/CLAR (12) 2. Replace H ₂ O (21) 3. SEQ Agent (22) 4. Enzymes (25) 5. Algaecide (35)	1. FLOC/CLAR (7) 2. Enzymes (15) 3. Replace H ₂ O (17) 4. Algaecide (21) 5. SEQ Agent (23)

Table A3. Cont.

Category	Influence Rank		Dependence Rank	
	Direct	Indirect	Direct	Indirect
SP(W) conditions	1. CONTAM (3) 2. Algae (5) 3. Scale (12) 4. WashedOffS (21) 5. Clogging (22) 6. TiO ₂ concSP (23) 7. Over-stab (29) 8. Phosphate (30) 9. Calcite (32) 10. Etching (36) 11. HMetal Lev (40)	1. CONTAM (3) 2. Algae (5) 3. Scale (12) 4. Clogging (13) 5. TiO ₂ concSP (14) 6. Over-stab (15) 7. WashedOffS (23) 8. Phosphate (25) 9. Calcite (30) 10. Etching (34) 11. HMetal Lev (38)	1. CONTAM (1) 2. Algae (2) 3. Clogging (8) 4. TiO ₂ concSP (11) 5. WashedOffS (15) 6. Scale (16) 7. Calcite (19) 8. Phosphate (23) 9. HMetal Lev (29) 10. Etching (32) 11. Over-stab (38)	1. CONTAM (1) 2. Clogging (2) 3. Algae (3) 4. TiO ₂ concSP (11) 5. Phosphate (14) 6. Scale (20) 7. Calcite (24) 8. HMetal Lev (29) 9. Etching (32) 10. WashedOffS (36) 11. Over-stab (39)
Essential SPW property	1. AggloPot (18) 2. Solubility (24) 3. IS (37) 4. ZetaPot (48)	1. AggloPot (24) 2. Solubility (29) 3. IS (37) 4. ZetaPot (47)	1. AggloPot (3) 2. IS (10) 3. ZetaPot (27) 4. Solubility (30)	1. AggloPot (8) 2. IS (12) 3. ZetaPot (26) 4. Solubility (31)
Operating parameter	1. Maintain (7) 2. RemovalCap (17) 3. FilterEffi (20) 4. DisinfeCap (25) 5. Turnover (38) 6. SP HCap (42) 7. Agitation (45)	1. Maintain (4) 2. RemovalCap (8) 3. FilterEffi (9) 4. DisinfeCap (21) 5. Turnover (33) 6. SP HCap (42) 7. Agitation (43)	1. RemovalCap (5) 2. DisinfeCap (6) 3. Maintain (7) 4. FilterEffi (24) 5. Turnover (31) 6. Agitation (36) 7. SP HCap (47)	1. Maintain (4) 2. RemovalCap (5) 3. DisinfeCap (6) 4. FilterEffi (13) 5. Turnover (30) 6. Agitation (35) 7. SP HCap (47)
Monitoring parameter (external)	1. RainOverfl (11) 2. Sunlight (19)	1. RainOverfl (11) 2. Sunlight (31)	1. RainOverfl (45) 2. Sunlight (46)	1. RainOverfl (44) 2. Sunlight (46)
Bathers' behaviour	1. Batherload (15) 2. ShowerDur (33) 3. ActivDur (41) 4. ReappliSS (44) 5. AppliedSS (47)	1. Batherload (18) 2. ShowerDur (32) 3. ActivDur (40) 4. ReappliSS (44) 5. AppliedSS (46)	1. ReappliSS (33) 2. ActivDur (39) 3. Batherload (40) 4. AppliedSS (41) 5. ShowerDur (44)	1. ReappliSS (33) 2. ShowerDur (34) 3. ActivDur (40) 4. AppliedSS (43) 5. Batherload (45)
Sunscreen formulation/TiO ₂ particle property	1. TiO ₂ SCchem (26) 2. TiO ₂ SDis (35) 3. TiO ₂ PPS (39) 4. TiO ₂ concSS (43) 5. TiO ₂ stabSS (46)	1. TiO ₂ PPS (35) 2. TiO ₂ SDis (39) 3. TiO ₂ SCchem (41) 4. TiO ₂ concSS (45) 5. TiO ₂ stabSS (48)	1. TiO ₂ SCchem (18) 2. TiO ₂ SDis (28) 3. TiO ₂ stabSS (37) 4. TiO ₂ concSS (43) 5. TiO ₂ PPS (48)	1. TiO ₂ SCchem (16) 2. TiO ₂ SDis (28) 3. TiO ₂ stabSS (38) 4. TiO ₂ concSS (42) 5. TiO ₂ PPS (48)

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