Modeling Ozonation and Chlorination in potable Water Treatment

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Abstract

This article presents SimOx, the new simulator being currently developed by Anjou Recherche / ENSCR for oxidation steps in potable water treatment works.
Whereas designing this new prediction engine was originally motivated by difficulties encountered when simulating a plant under on-site conditions, an increasingly stringent legal context and the emergence of micropolluants of concern clearly confirmed the need for a performing and innovative tool. The experience gained with the development of previous simulators is of advantage and opens interesting possibilities, in particular calculation capabilities upon which SimOx partly bases. Nevertheless, given the very characteristics of the new simulator SimOx (capacity to deal with sparse on-site data under changing water matrixes and hydrodynamic conditions), special modelling and calculation procedures are being implemented.

Keywords

Modelling, Oxidation, potable Water Treatment, Boundary Value Problem, Disinfection By-Products, Micropollutants Removal.

Introduction

In regards of increasingly stringent rules on toxicity (e.g. regulations 1DBPR for the USA and 98/83/EC for the EU), simulation softwares for potable water treatment have become a necessity. This particularly applies to oxidation steps, where disinfection must be guaranteed without surpassing the legal thresholds on potentially harmful by-products, and has therefore lead to the building up of a new simulator for both chlorination and ozonation, SimOx.
Management of oxidation steps involved in potable water treatment works is presently based on few measurements, partially collected at the outlet of the processes, as residual ozone. Now, the vast majority of oxidation simulators that have been developed perform their calculation downstream assuming the whole initial state, i.e. the inlet, to be known. Considering this contradiction, we therefore propose to build up an innovative simulator adapted to on-site conditions: easy-to-use and effective, provided only with few measurements (from the system

boundaries: inlet and outlet), its indications should insure a good level of disinfection combined with an acceptable by-products formation rate. It has also to be able to predict micropollutants fate. Previous research has lead to the development of simulation devices that predict concentration profiles, basing on the knowledge of the inlet composition though [Savary, 2002]. Our aim is hence to ameliorate and develop them further, adapting their modelling and calculation procedures to on-site conditions (see figure **1**).

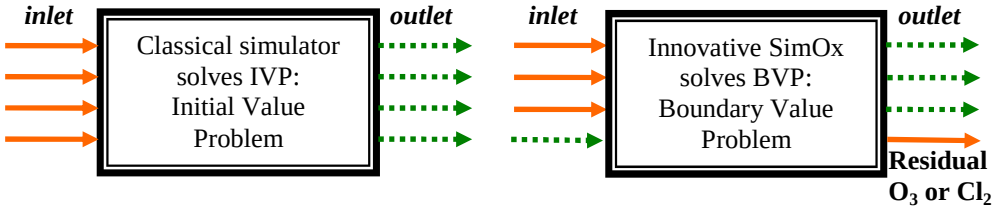

Figure 1 Comparison between a typical available simulator, working under normal conditions (left) and SimOx working under conditions encountered on site (right). The solid arrows represent the known or observed data (simulation inputs) whereas the dotted arrows represent the unknowns (calculated simulation outputs).

Such project implies a multi-disciplinary approach: besides chemical kinetics, hydraulics and simulation issues, other constraints (legal context, user-friendliness, data availability) have to be taken into account as well. We analyse them in the first section and demonstrate the need for an innovative tool as SimOx giving its simplified scope statement. We then present simulators already developed by Anjou Recherche / ENSCR and highlight the present prediction capabilities for oxidation in potable water treatment works. Lastly, we give more insight into SimOx, shortly describing its operating principle.

I A context requiring new tools

Setting the rapidly changing context of potable water oxidation clearly stresses the challenges that are faced when developing a simulator. We review in this part the main constraints on oxidation steps in water treatment works and confront them to the objectives of a simulator.

Rigorous legal frame

Although widely used, chlorine (or its related compounds) and ozone as powerful oxidising agents have shown application limits. Actually, brought into natural water, these species, reacting with naturally occurring organic substances, are prone to form so-called DBPs (Disinfection By-Products), potentially harmful.
The USEPA regulatory instance has therefore set levels of authorized contamination for various suspected species. Target compounds with their limits were listed in Stage 1 Disinfectants and Disinfection By-Products Rule (Stage 1 DBPR), which was issued in 1998. In 2006, Stage 2 was built upon Stage 1 DBPR to address higher risk public water systems for protection measures beyond those required for existing regulations. The MCLs (Maximum Contaminant Levels) defined by Stage 1 DBPR and the MCL recommendations (MCL Goals) of Stage 2 DBPR are given in table **1**.

Table 1 Level of authorized contamination – Stage 1DBPR and 2DBPR (source: USEPA)

Contaminant	**MCL ($mg.L^{-1}$)** ***fixed by 1DBPR***	**MCLGoals ($mg.L^{-1}$)** ***targeted by 2DBPR***
Total Trihalomethanes (TTHM)	0.080	*not affected*
Five Haloacetic Acids (HAA5)	0.060	*not affected*
Bromate	0.010	0
Chlorite	1.0	0.8

With a slight delay, the European Instances followed the American position. So, the European Council emitted on November 3, 1998 the 98/83/EC directive on the quality of water intended for human consumption, which regulates water quality at the tap [Roccaro et al., 2005], [Duguet et al., 2006]. The European MCLs are gathered in table **2**.

Table 2 Level of authorized contamination – 98/83/EC (source: European Portal, http://europa.eu)

Contaminant	**MCL ($mg.L^{-1}$)**	
	December 25, 2003	December 25, 2008
Total Trihalomethanes (TTHM)	0.150	0.100
Bromate	0.025	0.010
Chlorite	0.200	

Emerging micropollutants

Since the 1990's, pharmaceutical substances have been increasingly found in aquatic environment and thus recognised as an important class of organic pollutants. These emerging micropollutants are repeatedly detected in surface and ground waters at concentration ranging from $ng.L^{-1}$ to $\mu g.L^{-1}$ (see e.g. [Serensen et al., 1998]), depending on the drugs ease of being degraded. It is worth mentioning that the pharmaceutical residues belong to various classes of drugs: antibiotics, anti-epileptics, analgesics, antineoplastics, pharmaceuticals acting as endocrine disruptors, contraceptives… [von Gunten et al., 2005].

Participating in the European TECHNEAU project, within a work area devoted to cost effective technologies and system schemes development, SimOx benefits from common research efforts on emerging issues in drinking water. In this frame, partners have agreed upon an experimental comparison of micropollutants removal effectiveness for different processes (including oxidation processes and AOPs). This global approach gives opportunity to share knowledge on target molecules commonly studied: besides sulfamethoxazole (antibiotic agent) and carbamazepine (anticonvulsant), synthetic organic contaminants. Pesticides and herbicides such as alachlor, atrazine and its degradation by-products DEA and DIA are being investigated as reference compounds.

Already existing simulators

All the existing water treatment plant simulators propose, more or less, same functionalities: design, process optimisation, operator training, educational purposes, automation. Some of them also include cost savings investigation

modes. Furthermore, the simulators are very similar in their use. The interface allows building up one's own model and then to run the simulation, having specified certain characteristics (regarding the water, the processes etc…). Table **3** gives an overview of common simulators currently available.

Table 3 Comparison of potable water treatment works simulators

Name	Developed by	Highlights/ Strenghts	Drawbacks/ Weaknesses	Chemical models
OTTER	WRc	Readily extensible by users familiar with FORTRAN/C/C++	Large data needs	Semi-empirical relations
Stimela	TU Delft	Online access	Simple oxidation models	Semi-empirical relations
Metrex	University of Duisburg	Particle removal	Not tested on site	Mechanistic + correlations
Watpro	Hydromantis	Disinfection-DBPs	Long calibration time: 1 year of data	USEPA correlations
WTPmodel	USEPA	Removal of NOM-DBPs	Limited validity domain	Empirical relations

Contrary to SimOx, all the simulators presented in table **3** are not oxidation-specific. They all aim at simulating a whole water treatment works. This is why, even though focalising on disinfection problematics (DBPs), their use has given evidence of the lack of precision in their predictions for single processes [Dudley and Dillon, 2005]. As it appears in table **3**, the main drawback of common simulators lies moreover in their poor adaptability to specific on-site conditions. When adaptable, the simulators require a very long calibration period. This is mainly imputable to the choice of basing the models on correlations or empirical relations, of which role is not to be physically valid but to fit simulation results to experimental data. Obviously, one cannot simply eliminate such correlations (especially dealing with NOM (Natural Organic Matter)), but our efforts are directed towards reducing their number. Moreover, the hydraulic representations offered by the various existing prediction engines remain simple, modeling all flow conditions uniquely with CSTRs (Completely Stirred Tank Reactors).
Finally, one should keep in mind on-site specificities: (i) only few measurements, (ii) available at various locations of the process (inlet, outlet). This aspect, presently not considered by the over-listed simulators, is a key feature of SimOx.

Scope statement

Having set the global context of oxidation for potable water treatment and its modelling, we now exhibit the challenges that have to be faced, considering the characteristics of SimOx.
Besides providing reliable tendencies even when extrapolating outside the calibration domain - which is difficult with a correlation-based simulator - the main functionalities of the simulator can be seen as answers to the challenges previously expressed. This is schematically summarised in figure **2**.
The main challenges are:

- The various conditions the simulator is dealing with (water quality, hydrodynamical conditions)

- The incomplete knowledge of the process parameters (when measured, some concentrations are observed at the outlet, some other at the inlet)
- This tool is designed to be used on-site

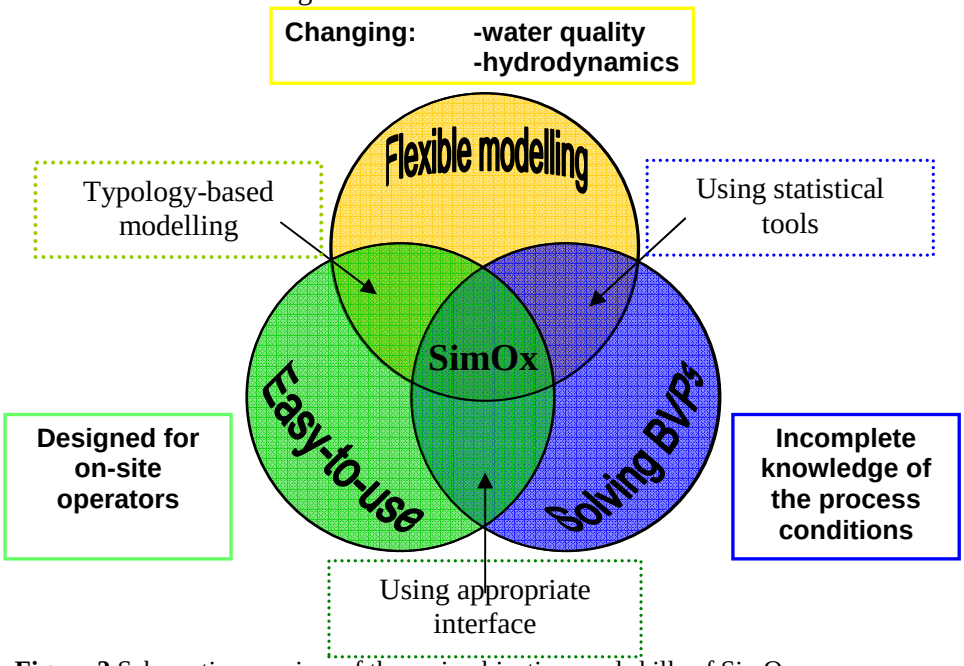

Figure 2 Schematic overview of the main objectives and skills of SimOx.

To those issues, SimOx' conception gives following answers:

- A flexible model, with a limited set of adjustable parameters adapting to specific conditions. In order to systematise the modelling, a typology-based approach was chosen.
- Special solving capabilities: the simulator is able to handle incompletely defined initial states. Statistical tools are also implemented to get a more effective resolution and to permanently upgrade the adjustable modelling part.
- A user-friendly interface allowing one to choose between different configurations (chemically or hydrodynamically).

II Tools already available : $SimO_3$ and $SimuCl_2$

History

During the nineties, with rising concern about micropollutants and DBPs, be it for ozonation or chlorination, it became clear that a reactor modelling based only on the Ct concept was no longer sufficient. This approach had been chosen, for safety reasons, to underestimate the inactivation efficiency of a given reactor, using the outlet oxidant concentration as average concentration and the t_{10} as residence time. Later, some improvements were included in order to give inactivation credits for each contact chamber. Even though, a plug flow pseudo first order degradation model can only be of limited help in predicting the DBPs formation resulting from successive oxidations like THMs or bromates : it works, not because of the validity of underlying concepts, but because the parameter fitting led it to.

Based on it, but with a large scope, practical global correlations were developed basing on lab experiments and on-site measurements (e.g. [Song et al, 1996], [Ozekin and Amy, 1997]) with a limitation: unexpected or uncertain results might come out when using correlations outside the range of conditions for which the parameters were identified (e.g. hydraulic behaviour of the reactor, extreme temperatures, nature of the organic matter…). For a group like Veolia Water, with plants all over the world, a more generic approach was sought.

At about the same period, on one hand, a major development entered the water treatment world: the detailed hydrodynamic modelling of the reactors using computational fluid dynamics, CFD, ([Doquang 1993], [Doquang et al., 1996], [Murrer et al., 1995], [Dumeau de Traversay, 2000]). On the other hand, the growing complexity of the true chemical reaction mechanisms and their kinetics for hydroxyl radical and bromate formation were described in controlled conditions in the labs ([von Gunten and Hoigné, 1994], [Westerhoff et al., 1998]).

The solution of an oxidation reactor model combining hydraulics and chemical kinetics is extremely difficult to find given the very discrepancies in time scales. Even if softwares like FLUENT CFX, etc… are theoretically able to solve such problems, it would be overly resource consuming, - computational time and memory -, for a frequent use. The need for a tool able to solve an approximate but realistic model emerged, from both chemical and inactivation points of view.

Principle

Considering that a local, microscopic, knowledge of velocity and concentration fields of all chemical entities might not be relevant, it was decided that the simplification would essentially concern hydraulic models. It is a current practice in chemical engineering, to describe complex reactors by their RTD (Residence Time Distribution), and to use, as model, a systemic representation exhibiting a similar RTD. A systemic representation is a network of ideal reactors, CSTRs or PFRs (Plug Flow Reactors). Such a network may include by-passes, recycling flows, etc… It should be regarded as an integrated form of the flow field. Hence the RTD is not the only criteria, localised turbulent areas should be modelled as such and placed accordingly in the network, as it will be shown later. In figure **3** we present an example of the hydraulic study of a 381 m^3 ozonation reactor with 4 chambers. The 3D computational fluid dynamic simulation was performed using FLUENT and compared to on-site tracer test. These results, local flowrates and turbulent areas, were then used to build the 2D systemic network. Characteristic times and residence time distributions are given as well.

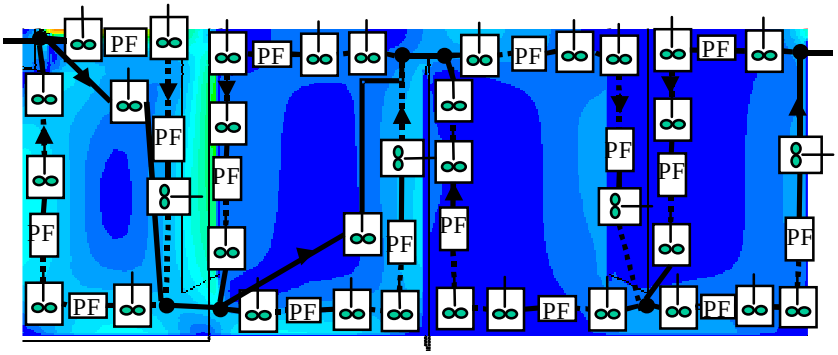

Figure 3.a CFD flow pattern and systemic network

	Tracer test	Fluent	Systemic
t_a (min)	3.2	2.4	2.4
t_{10} (min)	4.9	3.8	3.8
$\bar{t}$ (min)	12.3	11.8	11.8
τ (min)	13.4	12.1	12.1
t_{90} (min)	21.7	21.8	21.8

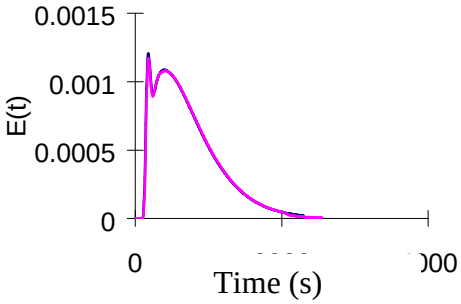

Figure 3b and 3c Example of an hydraulic study of a 381 m^3, 4 chambers, industrial tank at 2200 $m^3.h^{-1}$

The chemical transformations, represented by several sets of chemical equations associated with mixed order kinetic rates models and Arrhenius-type temperature dependence, may be simplified as well. But this should be done with caution. Some pathways, inactive under moderate conditions, could namely become important by a simple but unusual change in temperature, pH or concentration of some compounds. Each set corresponds to a consistent group of reaction forming a part of the chemical description: for example one set for the O_3/°OH mechanism, one for the reactions implying carbone-containing mineral entities (ions, radicals …), one for the reactions implying bromine-containing entities, one per identified micro-pollutant, etc… One special set handles the inactivation of bacteria or oocysts with reaction-like equations.
In figure **4**, we present an example of consistent chemical description with 3 identified groups: the core reactions, the interactions with the NOM ("instantaneous" ozone demand, ozone consumption, promotion and scavenging of radicals), the targets (i.e. bromate, micropollutant, inactivation).

Figure 4 Example: a reaction sets collection for a consistent chemical description

First developed for single-phase reactors, the software $SimuCl_2$, has been designed as a generic reactor simulator (i.e. it could be used as well for contact chambers in ozonation reactors) but specifically applied and validated for post-chlorination reactors ([Mahé et al., 2000]). A second version, $SimO_3$, includes two phases ideal reactors with mass transfer ([Savary, 2002]). Gas and water have separate flow patterns interconnected only through reactive contact reactors. Global mass transfer coefficients K_La can be given locally or deduced from various empirical correlations. Gas-liquid equilibria (O_3, but also CO_2 and other compounds if required) obey Henry's Law with a Van't Hoff temperature dependence.
Figure **5** summarises the information flow for the definition and the solution of a problem. Numerous information can be gathered from the literature, especially concerning the chemistry and mass transfer data; the remainder can be obtained through experiments, calculations (CFD), and exploitation of existing on-site measurement history.

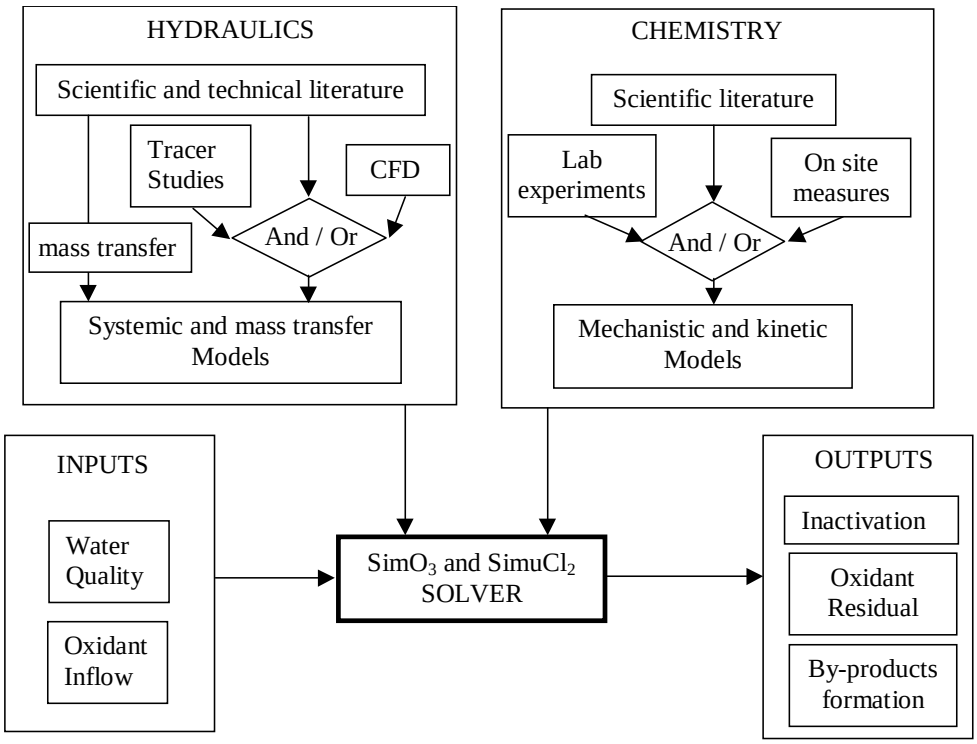

Figure 5 Solvers environment.

Results and needs for improvements

Finally, figure **6** presents the comparison of a series of ozonation simulations performed using the previous approach and compared to experimental data obtained, over a period of 5 months in 1999, for a reactor with a 15 to 33 minutes hydraulic residence time, ozone doses from 0.9 to 3.2 $mg.L^{-1}$. The reactor is fed by sand-filtered water of the Neuilly-sur-Marne plant and ozonated air.
The reaction set was similar to the one presented in figure **4**. The hydraulic network was not determined on site but based on similarities with an other reactor. Small atrazine spikings were performed to have a feedback for the °OH radical profile. The only fitting that was performed once for the whole data set, concerned

the °OH promoting effect of the NOM, which explains the good correlation of the atrazine results.

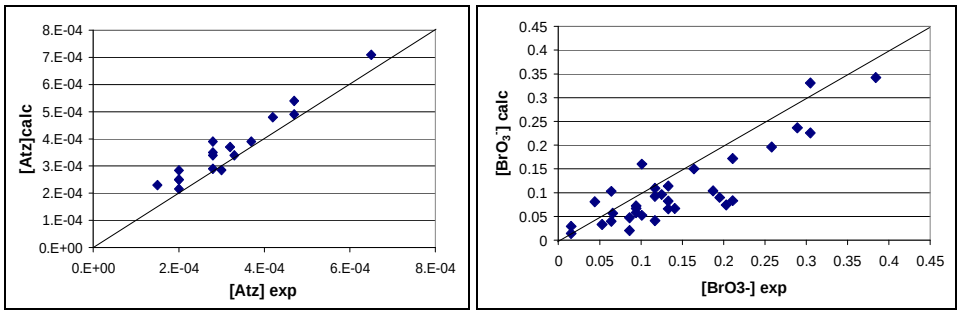

Figure 6 : Comparison of calculated versus experimental residual molar atrazine levels and bromate formation for a real water (Ozone dose : 0.9 to 3.2 mg.L^{-1}, $[O_3]_G$ = 4.8 to 26 mg.NL^{-1}, L/G ratio = 5 to 11, TOC : 1.3 to 1.7 mg.L^{-1}, pH : 6.5 to 7.4, τ = 15 to 33 min.)

The atrazine and bromate results were contrasted but encouraging. The main tendencies and effects were clearly reproduced (not detailed here). However, the lack of precision, especially for low concentrations, will have to be addressed. This illustrates the importance of local adaptation of the model: among the possible local improvements in this case: "instantaneous" demand and O_3 demand and their impact of the °OH formation, better knowledge of the hydraulics.
Moreover, like most simulators, both versions $SimuCl_2$ and $SimO_3$ require full knowledge, be it exact or hypothesised, of the inputs: the concentration of all species, pH, temperature, flowrates… and produce a completely determined global output.

III A new type of modelling

In this section, we review the key features of SimOx explaining how they fulfil the objectives set in the scope statement and overcome the limitations of the simulators previously presented in section II. The simulator still being under development, there will not be any consideration on its interface. Hence, this section is essentially focused on the simulator's operating principle.

A three-stage resolution

Confronted to a situation where only sparse information on the process conditions is available, SimOx operates in three steps, using different physico-chemical models:

-Firstly, the simulator tries to get quick knowledge on the missing inlet data **X** basing on the known data. This is done by means of statistical tools such as Artificial Neural Networks using a robust simplified physico-chemical model. The purpose of this step is to recreate an initial state likely to have led to the observed outlet oxidant concentration.

-Secondly, by iteratively solving the problem of determining the best value for **X** to fit to the outlet oxidant concentration. This is achieved using solving procedures (relaxation, optimal control) on a model including all the reactions significantly altering the oxidant profile.

-Lastlty, SimOx handles a classical problem with all initial conditions known. A complete model including all possible reactions (including micropollutants and DBPs) is implemented.
This is schematically illustrated by figures **7**, **8** and **9** using the same representation conventions as figure **1**.

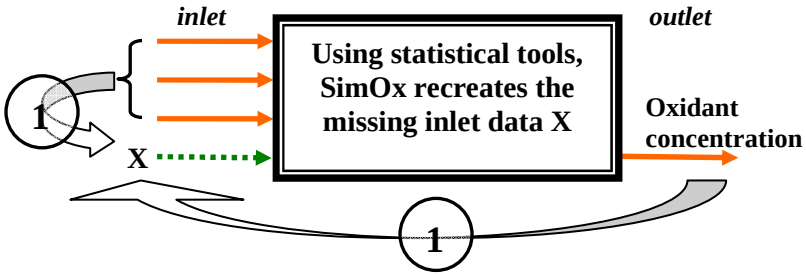

Figure 7 Schematic representation of the first step in SimOx' solving procedure. The simulator initialises the solving procedure illustrated in the next figure with a simplified model (1st model).

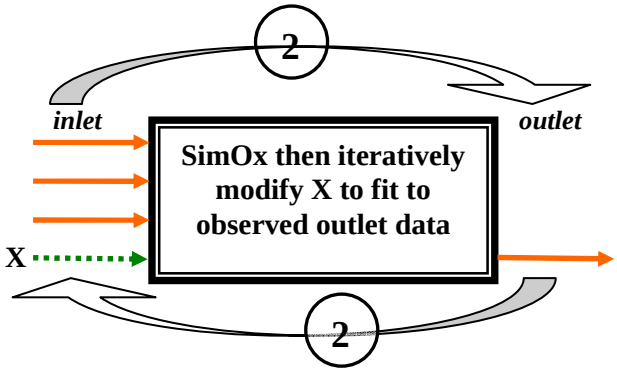

Figure 8 Schematic representation of the second step in SimOx' solving procedure: finding the value for **X** with a model including only reactions relevant to the oxidant profile (2nd model).

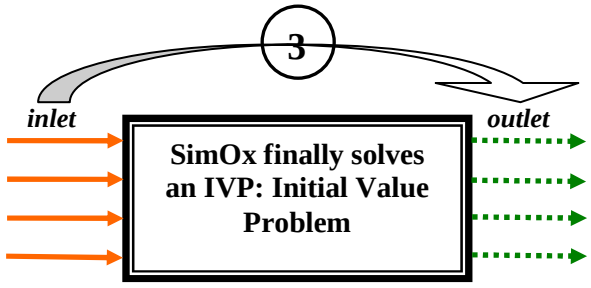

Figure 9 Schematic representation of the third step in SimOx' solving procedure. A complete model is being implemented.

Simplified calibration procedures

Considering the very specific nature of water constituents, there presently cannot be any modelling for oxidation steps in potable water treatment without calibration phase. Calibrating SimOx through an adaptation of its adjustable model's parameters is therefore a necessary step to insure performing predictions. The experience of the previous simulators (see section II) has shown that an overly complex calibration is not suited for on-site application. An important part of the

development efforts has thus been devoted towards a simplification of the calibration procedure. The idea is to:

-Determine the relevant parameters uneasily predictable, of which knowledge provides good prediction for the oxidant - and radicals, in the case of ozonation - profile

-Find the easiest testing procedures to assess their values. Typically, these are bench-scale measurements of chemico-physical water properties (batch testings or Ozotests [Roche et al., 1994]).

In SimOx, specific water conditions uniquely affect the oxidant profile. Hence, calibration only concerns the two first models, in a different fashion though: whereas the simplified model is directly calibrated from on-site measurements, the second model is mostly affected by bench-scale testing results. The calibration of the second model may also require additional information from on-site measurements. This is summarised in figure **10**.

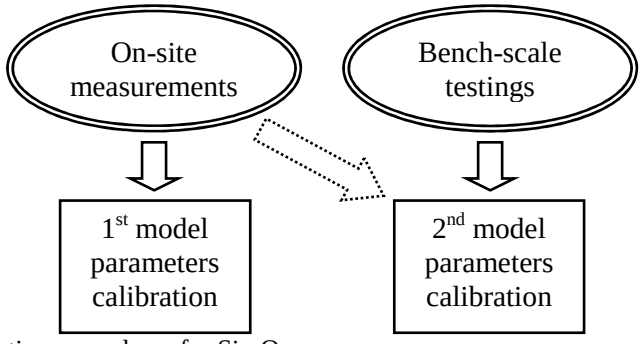

Figure 10 Calibration procedures for SimOx.

Typology-based approach

In introduction to this paper, we specified the very general requirements of SimOx. Amidst them, adaptability plays a key role. Adaptability to on-site conditions means both (i) adaptability to chemical characteristics (i.e. water quality) and (ii) to hydrodynamic specificities (e.g. reactor geometry, equipment…). This has much to do with calibrating and has led to simplified calibration procedures, but also to typology-based modelling.

Thereby SimOx gives the user the possibility to choose between different configurations (chemical, hydraulic). In doing so, the operator disposes of two types of lists: a list of reaction pathways coming into play according to water quality; and a list of typical reactors representing all the hydrodynamic conditions that can be encountered on site. Following the prescribing of calibration procedures (precedent paragraph), the simulator development was focused on establishing such classifications. Based on various (e.g. technical, physical, geometrical…) considerations, this typology has to be drawn up carefully. Again, the simplicity of use and editing has to be balanced with the accuracy of the simulator's prediction; the length of the list has to be balanced with its ability to describe satisfactorily all possible configurations.

Finally, it has to be noted that SimOx is a readily extensible tool, offering the user the possibility to create its own chemical or hydraulic sets if specific configuration are needed.

Conclusion

Facing typical issues encountered under on-site conditions, SimOx represents a promising and innovative simulator for the optimisation of plant performance and thus improvement of drinking water quality.
After the laboratory testing phase that already begun and gave preliminary results, SimOx shall be tested on-site within a year. In the same time, the graphical user interface will be developed at Anjou Recherche. The complete simulator shall be operational within a year and a half on the first potable water treatment works.

Acknowledgements

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- European Portal, http://europa.eu
- USEPA website, http://www.epa.gov/