



RIGA TECHNICAL  
UNIVERSITY

**Sandis Dejus**

## **ONLINE DRINKING WATER QUALITY MONITORING**

Summary of the Doctoral Thesis



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Riga 2018

**RIGA TECHNICAL UNIVERSITY**

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Institute of Heat, Gas and Water Technology

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MONITORING**

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# **DOCTORAL THESIS PROPOSED TO RIGA TECHNICAL UNIVERSITY FOR THE PROMOTION TO THE SCIENTIFIC DEGREE OF DOCTOR OF ENGINEERING SCIENCES**

To be granted the scientific degree of Doctor of Engineering Sciences, the present Doctoral Thesis has been submitted for the defence at the open meeting of RTU Promotion Council on 13 December 2018, 12:00 at the Conference Hall of Riga Technical University, 6 Azenes street, floor 11.

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## **DECLARATION OF ACADEMIC INTEGRITY**

I hereby declare that the Doctoral Thesis submitted for the review to Riga Technical University for the promotion to the scientific degree of Doctor of Engineering Sciences is my own. I confirm that this Doctoral Thesis had not been submitted to any other university for the promotion to a scientific degree.

Name Surname ..... (signature)

Date: .....

The Doctoral Thesis has been written in Latvian. It consists of Introduction; 4 chapters; Conclusion; Recommendations; 18 figures; 11 tables; 9 appendices; the total number of pages is 120. The Bibliography contains 186 titles.

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# 1. GENERAL DESCRIPTION

## 1.1. Introduction. Topicality

Currently, about half of the human population lives in cities. However, forecasts show that in 30 years two-thirds of the world's population will be living in cities and agglomerations. Population growth in cities leads to increase of risks associated with public drinking water supply systems and water regarding safety and amount. Although during the last century there have been ambitious improvements regarding drinking water supply systems and technologies, there are still cases of public drinking water supply system contamination events that from time to time lead to illnesses or deaths of consumers. Scientific studies have shown that about 30 % of all gastrointestinal illnesses recorded in urban areas are related to consumption of inadequate quality drinking water from centralized systems. Application of drinking water quality evaluation methods that are regulated by legislation have not been able to detect those contamination events. Therefore, online drinking water quality monitoring systems with early warning function have been developed. In the case of a detected contamination event, the systems automatically trigger an alarm. Such systems are commercially available, but experimental studies show that the average contamination event detection accuracy is 82 %. It means that consumers are still exposed to potential threats. Moreover, evaluation of microbiological drinking water quality is not integrated into such a system, because the use of existing microbiological methods requires 18–24 hours. Consequently, it is necessary to improve early warning systems by implementing new and more accurate contamination detection algorithms and investigations of most appropriate drinking water quality parameters that would be required for online drinking water quality monitoring.

## 1.2. The objective and main tasks

The objective of this Thesis is to develop a solution for drinking water quality monitoring system including the measurement of physicochemical and microbiological parameters and to evaluate its precision in the detection of various contamination events.

The main tasks are as follows:

- identification of physicochemical and microbiological drinking water quality parameters that theoretically should be included in online drinking water quality monitoring systems;
- development, adaption, and evaluation of an automatic drinking water contamination event detection algorithm;
- identification of combination of drinking water quality parameters that provides the most accurate detection of various contamination events by pilot scale experimental studies.

### **1.3. Scientific novelty and application**

Within the framework of this Thesis, a new solution for online drinking water quality monitoring has been developed. It contains monitoring of physicochemical and microbiological drinking water quality parameters and an automatic contamination detection algorithm. Since the solution introduces the most promising microbiological drinking water quality investigation methods such as flow cytometry and adenosine triphosphate, it is innovative and unique. The results of this study spotlight the necessity of further experimental and theoretical studies on online measurements of these microbiological parameters and methods. The study also demonstrated that the introduction of these microbiological parameters in early warning systems has a positive impact on the accuracy of the contamination detection. Moreover, by implementing the developed solution, it is possible to reduce the time required for determination of the microbiological conformity of drinking water for safety requirements from 18 to 24 hours to 5 minutes. Prior to application of the developed solution in real scale systems it is necessary to examine it by implementing online measurement systems and automated microbiological measurement methods, however experimentally obtained results indicate the essential benefits of the use of this solution in real water systems.

## **2. LITERATURE REVIEW**

In order to ensure the safety of drinking water, comprehensive drinking water monitoring must be performed at each stage of the drinking water supply system. In developed countries, including Latvia, drinking water quality after drinking water treatment facilities meets all requirements, but during storage and distribution, its quality may deteriorate [1]. Statistical data shows that 30–60 % of all complaints and contamination events in drinking water supply systems are related to the distribution network [2]. The reasons for drinking water quality deterioration can also be unfortunate accidents as well as intentional actions. Insufficient drinking water quality can cause problems related to health, social, psychological, economic and reliability issues [3], [4]. Unfortunately, there is no perfect water supply system that does not create any potential risks for its users in the world. Therefore, the objective of water utilities is to minimize the potential risks of contaminated or insufficient quality water and protection of consumers by implementing drinking water quality monitoring [5].

Despite the risks, in most of the world, the drinking water quality monitoring is carried out by the implementation of standard methods for grab sampling and laboratory investigations. Therefore, the actual results of drinking water quality are established with a relatively long time shift [6], [7] that might lead to additional threats to consumers. For example, the determination of presence and concentration of pathogenic microorganisms in drinking water samples by the classical cultivation method requires several days. Consequently, the risk of supply of contaminated drinking water may be increased [8]–[10]. Use of existing standard methods does not provide any information regarding the dynamics and compliance to the regulation of drinking water quality within the scope of hours, days, or weeks. Moreover,

previous research shows that through current monitoring programmes [6], the presence of microbiological contamination can be found with a probability of less than 5 % [11]. In previous studies it was concluded that a human resource of 0.37 to 3.58 full-time equivalents is needed to detect and investigate one microbiological contamination event [12].

For more accurate and comprehensive drinking water quality monitoring, online drinking water quality monitoring systems with early warning function are applied more often. Such a system, in the case of contamination event, triggers an automatic alarm and warns the drinking water supply system operator that actions should be taken to minimize the potential risks [10]. The costs of installation of such a system are relatively high [2]. To reduce the installation costs, cheap and robust sensors for monitoring of physicochemical parameters are used. The measurement results are processed with mathematical algorithms that can determine whether the water complies with drinking water safety requirements or not. In case of non-compliance, alerts to the operator of the water supply system are made in order to ensure that appropriate operational activities are undertaken in a timely manner [10]. Implementation of such a system can reduce the time needed for contamination event detection to the minute [10], [13].

Although during the last decade significant improvements of early warning systems have been made, the contamination detection and classification accuracy are highly dependent on the type of contaminant and detection algorithm integrated into it. It is necessary to perform experimental studies with simulations of contamination events to improve these algorithms, determine their stability and choose the parameters to be monitored [13]–[15]. The experimental tests of commercially available systems that include monitoring of physicochemical parameters or combinations of them show the contamination event detection in 82 % of cases. This indicates the need for improvement of these systems [16]. There are algorithms reported in scientific papers that have a capability to detect the contamination with 100 % accuracy. However, in most cases, they are evaluated with artificial data sets or experimental data from chemical contamination events [13], [17], [18]. Moreover, there have not been studies with the implementation of microbiological parameters or methods for development of early warning systems. Consequently, in most cases, the quality of microbiological drinking water remains unknown, but microbiological quality is the most common cause of different diseases [7], [19], [20]. The reason for the lack of studies might be that the currently applied methods of detection of microbiological parameters are time-consuming (24 h to 48 h) and labour intensive, but new and relatively faster methods are being appropated relatively slowly [7], [19]–[23]. Introduction of new and faster microbiological methods in early warning systems could improve the accuracy of the detection of contamination incidents, particularly in the case of microbiological hazards [7], [24], [25].

Extensive research on the improvement and new implementation of existing microbiological examination methods for drinking water is being carried out worldwide. There have been more than 10 innovative methods proposed by scientists [7]. In terms of this Thesis, because of the relatively short time consumption and high precision, adenosine triphosphate and flow cytometry measurements are performed.



### 3. MATERIALS AND METHODS

#### 3.1. The evaluation method of contamination detection algorithms

The accuracy of contamination detection algorithms indicates its ability to identify pollution incidents or automatically classify “clean” and “contaminated” water. In order to assess the accuracy of operation of the pollution detection algorithms, it is necessary to evaluate the conformity of their classification results with water quality in the water supply system (Fig. 3.1) [18], [27], [28].

		Quality of Water	
		Clear	Contaminated
Decision made by algorithm	Clear	True negative (TN)	False negative (FN)
	Contaminated	False positive (FP)	True positive (TP)

Fig. 3.1. Possible contamination detection algorithm results in relation to water quality.

The evaluation of results made by the contamination detection algorithm is used to estimate the sensitivity of detection ( $PD$ ), false alarm rate ( $FAR$ ), and classification accuracy ( $P$ ) values for the algorithm. The sensitivity of the detection algorithm describes its ability to detect a contamination event. The false alarm rate indicates the probability of false classification results. Classification accuracy describes the overall accuracy of the algorithm to correctly classify the measurements in the presence of “clean” or “contaminated” water in the system. All resulting algorithm performance assessments range from 0 to 1. In the case of  $PD$  and  $P$ , the value 1 corresponds to 100 % accuracy. For  $FAR$  value 0 indicates correct classification results.

#### 3.2. Experimental study of drinking water contamination events in a pilot scale water supply system

Within the framework of this Thesis the experimental evaluation of drinking water quality parameter variations by simulating various contamination event scenarios was performed. Also, the assessment of the contamination detection algorithm’s ability to detect a contamination event was accomplished. In terms of experimental studies, a pilot scale drinking water supply system (Fig. 3.2) was constructed at the premises of Riga Technical University. In the system, the contamination events were simulated. Systems of such type and scale are used to prevent threats to consumer health. Experiments can be carried out within controlled hydraulic conditions but in the context of a real centralized water supply system.

### The pilot scale water supply system

The pilot scale water supply system (Fig. 3.2) consists of a 200 metre long polyvinyl chloride (PVC) pipeline with an internal diameter of 25 mm. The pipe material fulfills the requirements for food production, and it is assumed that it minimizes the possible bacterial growth impact that might be found in results. The total volume of the system is 98.2 liters.

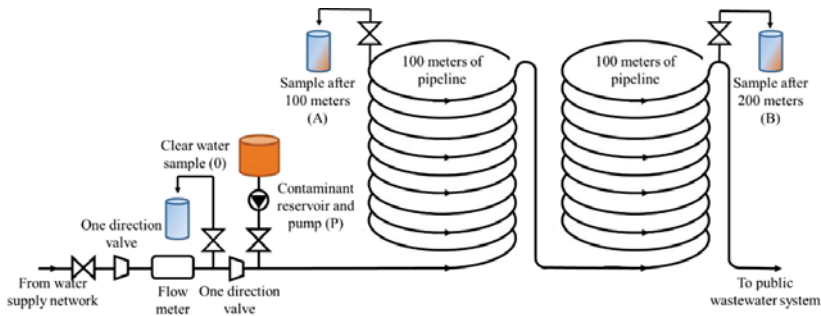


Fig. 3.2. Schematic representation of the pilot scale water supply system.

In total there are three sampling sites installed in the system, accordingly before the contamination dosage connection (0), 100 meters after (A) the contamination and 200 meters after (B) the contamination dosage connection. The clear water sampling site is placed before the contaminant dosing point and is intended for monitoring water quality changes in the water supply system of the city of Riga [29]. At the potentially contaminated water sampling sites, A and B, drinking water quality changes during various contamination scenarios are observed. To maintain a steady flow and hydraulic conditions, an ultrasonic flowmeter and valve are installed at the inflow of the system. To simulate the contamination events, a contaminant dosing system with contaminant reservoirs and dosing pump (P) are used.

### Parameters of drinking water quality monitoring

To evaluate how physicochemical drinking water quality parameters change, cheap and robust analysis methods and sensors that are widely used in early warning systems are used: temperature ( $T$ ), electrical conductivity (EC), oxidation-reduction potential (ORP), pH, turbidity (NTU), and total organic carbon (TOC). Measurement results collected from such sensors can relatively accurately indicate non-typical conditions in a drinking water supply system. For microbiological drinking water surveys promising alternative microbiological test methods, flow cytometry (FCM) and adenosine triphosphate (ATP) measurements are used.

The method of grab sampling and laboratory analysis has been used during the experiments. To exclude potential problems related to the operation of physicochemical online sensors and data transmission, such as signal disturbances and “noises”, online measurements are not used. Grab sampling is also required for the use of selected microbiological methods.

### Drinking water contamination event scenarios

During the experimental investigation, 4 contamination scenarios were simulated: surface water, groundwater, wastewater, and pathogenic bacteria model contamination.

In the surface water scenario, a situation is simulated where there is an operational disturbance in the drinking water treatment plant and raw surface water enters into the water supply system. In the groundwater and wastewater scenarios, a damaged water supply pipeline installed 1 meter below the natural groundwater level, with an orifice diameter of 1.5 mm, is simulated. Regarding these scenarios the development of vacuum and subsequent intrusion of groundwater and groundwater with wastewater, leakage has been assumed. In the contamination scenario of the pathogenic bacterial model, a deliberate case of drinking water contamination by injection of *Escherichia coli* microorganisms with the broth into the system is simulated.

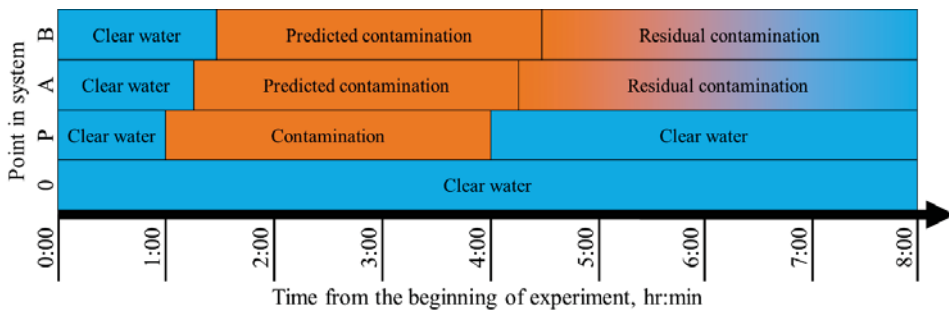


Fig. 3.3. Schedule of contamination scenario simulation.

The scenarios were selected on the basis of operation of the centralized water supply system and the most common problems and issues described in the literature. During all scenarios, a plausible microbiological threat to consumer health and the possible presence of pathogenic microorganisms are simulated.

The contamination flow (5 mL/s) during all the experiments is sustained as 10 % of total water flow in a pipe (50 mL/s). Sustained flow velocity in the pipeline is 0.1 m/s. The simulated contamination flow is a corresponding 1.5 mm pipeline rupture and negative pressure in the water supply system. In scenarios not related to intrusions, an identical flow is maintained to ensure the comparability of results. The total duration of the experiment is 8 hours. Each contamination event lasted for 3 hours (P) (Fig. 3.3). During each contamination scenario, 50 liters of contamination was dosed into the system.

Before dosage of contaminant (Fig. 3.3, time period “clean water”), at all sampling points (0 – sample of clean water, A — sample after 100 meters of pipeline, B — sample after 200 meters of pipeline, P — contamination dosing site) and during the whole experiment at sampling site 0 samples were taken at a 15 minute interval. The predicted contamination period was calculated on the basis of steady flow rate and assuming that there is no diffusion (16 minutes and 40 seconds at sampling site A, and 33 minutes and 20 seconds at sampling site B). For each contamination scenario, three repetitions have been done.

### 3.3. Contamination detection algorithm

The obtained results of the quality parameters' changes during simulated contamination were processed with the Mahalanobis distance detection algorithm [28]. The algorithm was selected on the basis of previously performed literature analysis. In previous studies this algorithm has shown the highest accuracy of the detection and classification of contamination. The Mahalanobis distance algorithm is based on a cluster analysis. It means that the data sets are grouped into classes, depending on their similarity. The Mahalanobis distance is a non-dimensional measure that describes the distance from a certain object to some selected point in multidimensional space. Water quality measurements performed at each time step are combined in one characteristic parameter. These are defined as an object; several similar objects can be defined as a class.

The principle of experimentally acquired data processing is shown in Fig. 3.4. The data processing can be divided into two stages.

In the first stage, the measurements done in the first repetition of the relevant contamination event scenario experiments are processed. Based on the theoretical calculation of water retention time at each sampling site, the measurements are divided into two groups. The group "clear water" consists of all readings ( $n = 56$ ) corresponding with the theoretical presence of clear water at each sampling site. The group "contaminated water" accordingly consists of all readings ( $n = 37$ ) corresponding with the theoretical presence of contaminated water at sampling sites A and B. To establish the value of the certain parameter in day-to-day water supply conditions, an average value from readings within the class "clear water" for each parameter is calculated. To remove the possible scale and amplitude impact of each parameter on the results, the ratio ( $RB$ ) of each reading and previously calculated average value of each parameter value is calculated. For each time step, the calculated parameter ratios  $RB$  (8 parameters) is combined within one representative vector characterized by 8 independent figures. The vectors, according to the previously performed grouping of "clean water" and "contaminated water", are arranged into two classes that are used in the next stage for the detection of drinking water contamination events.

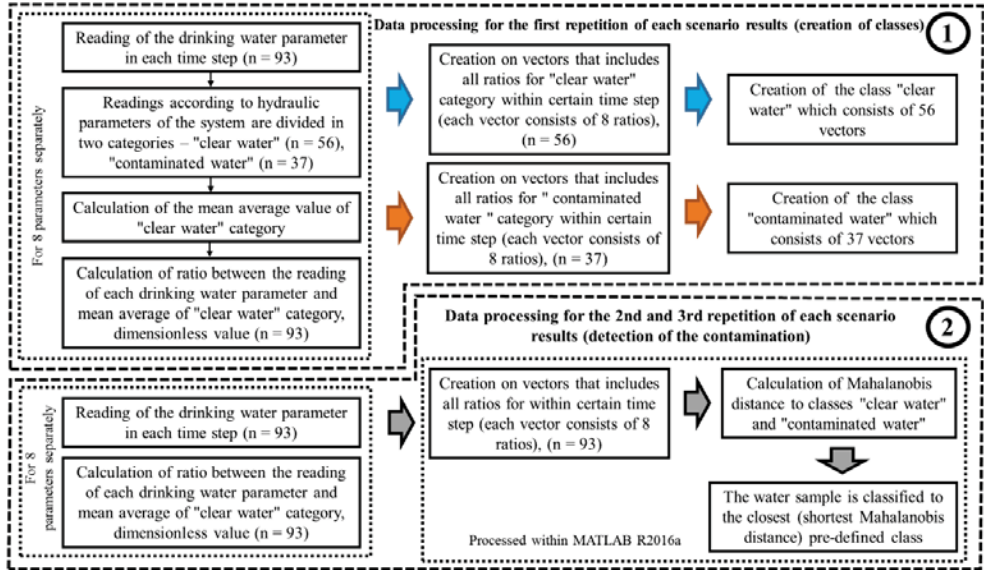


Fig. 3.4. The principle of drinking water quality measurement data processing.

In the second stage, similar to the first stage, relative ratios of  $RB$  are found for measurement results in each time step. The ratios acquired in each time step are combined into the representative vector. To determine the similarity of each vector to previously defined classes the Mahalanobis distance (3.1) is calculated. The measurements performed in the particular time step are classified to the class that they have the smallest Mahalanobis distance to. The data processing process is partly automated and developed using mathematical calculation software *MATLAB R2016a* tools.

$$D_M(p, c) = \sqrt{(p - \mu_c)^T \times S^{-1} \times (p - \mu_c)}, \quad (3.1)$$

where

- $D_M(p, c)$  – Mahalanobis distance between the measurement vector and defined class;
- $p$  – vector of measurements in  $n$ -dimensional space;
- $\mu_c$  – vector average parameter value within the particular class in  $n$ -dimensional space;
- $S$  – covariation matrix for vector  $y$ ;
- $T$  – transpose function.

## 4. RESULTS AND DISCUSSION

### 4.1. Comparison of contamination detection algorithms

In the scope of this Thesis during literature studies a comparison of 11 various drinking water contamination event detection algorithms has been done. To compare these algorithms, an evaluation of the sensitivity of detection ( $PD$ ) and false alarm rate ( $FAR$ ) for each algorithm is accomplished. The parameters describing the algorithms are summarized in

Table 4.1. There are *PD* and *FAR* values, the source of data for evaluation of algorithm in corresponding research, type of contamination event, and drinking water quality parameters used for detection contamination event.

Table 4.1

Comparison of contamination detection algorithms

Algorithm	<i>PD</i>	<i>FAR</i>	Data source	Type of contamination	Parameters
<b>VEA</b>	0.52	0.22	E	Cadmium nitrate	<i>T</i> , pH, NTU, EC, ORP, UV-254, nitrates, phosphates
	0.52	0.88	R	Phenol	pH, DO, COD, nitrates, phosphates, TOC, EC, NTU, <i>T</i> , fluorides
	0.84	0.61	M	Undefined	
	0.89	0.41			
<b>LPF</b>	0.38	0.94	E	Cadmium nitrate	<i>T</i> , pH, NTU, EC, ORP, UV-254, nitrates, phosphates
	0.68	0.82	R	Phenol	pH, DO, COD, nitrates, phosphates, TOC, EC, NTU, <i>T</i> , fluorides
	0.92	0.24	M	Undefined	
	0.90	0.25			
<b>PE</b>	0.97	0.025	E	Cadmium nitrate	<i>T</i> , pH, NTU, EC, ORP, UV-254, nitrates, phosphates
	0.83	0.33	R	Phenol	pH, DO, COD, nitrates, phosphates, TOC, EC, NTU, <i>T</i> , fluorides
	0.74	0.78	M		
	0.69	0.87			
	0.10	0.80	E	Glyphosate	<i>T</i> , pH, NTU, EC, ORP, UV-254, nitrates, phosphates
	1.00	0.00		Cadmium	
	1.00	0.00		Atrazine	
	0.76	0.00		Nickel	
	0.79	0.00		Chromium	
<b>KKA</b>	0.97	0.00	E	Acrylamide	
<b>MEK</b>	0.44–1.00	–			
<b>MNTVM</b>	0.08–0.59	0.001–0.09	M		<i>T</i> , pH, NTU, EC, TOC, total chlorine
	0.38–0.99	0.04–0.15			
<b>MNTDR</b>	0.30–0.99	–			
	0.58–0.98	–			
<b>AVM</b>	0.82	0.14			
<b>CANARY</b>	0.89	–			
	0.68–0.96	0.01–0.50			
<b>MA</b>	0.33–1.00*	–	E	Glyphosate	<i>T</i> , pH, NTU, EVS, ORP, UV-254, nitrates, phosphates
	0.20–0.95*	–		Sodium fluoride	
	0.94–0.95*	–		Cadmium nitrate	
<b>DSM</b>	0.21–1.00	0.032–0.39	M	Ferric ammonium sulphate	pH, EVS, ammonia
	0.52–1.0	0.006–0.39		Potassium ferricyanide	

Legend: M – artificial data set, R – contamination results from real-scale water supply system, E – contamination results from contamination experiments pilot-scale water supply system, *PD* – probability of detection, *FAR* – false alarm rate, *T* – temperature, NTU – turbidity, EC – electrical conductivity, ORP – oxidation-reduction potential, UV-245 – ultraviolet light sensor, TOC – total organic carbon, COD – chemical oxygen demand.

\*Classification result, detection accuracy *PD* for all contamination events are 1.00.

Several of the evaluated contamination event detection algorithms (MEK, MNTVM, MNTDR, AVM, CANARY, DSM) have not been tested with experimental measurement data

that describes the drinking water quality alterations during contamination events. In those assessments, the concentrations of contaminants and alterations of parameters are artificially created data sets. Consequently, there is no link to the possible measurements of water quality parameters and contamination events with concentrations that can be representative of real scale systems. Therefore, these assessments of the capabilities of contamination detection cannot be considered as comprehensive. In most of these data sets the contamination event is simulated as a rapid alteration of parameter values with mutual correlations during the contamination event. Moreover, the duration of the artificial contamination events is long enough to be detected by algorithms. The use of these kind of data sets and the established accuracy of the contamination detection may be misleading, as well as significantly lower in real data processing. In order for the comprehensive assessment of the algorithm to respond to a real contamination event, pilot-scale experiments should be carried out by monitoring water quality parameter alterations that are not necessarily correlated with each other.

A very significant component of the contamination detection algorithms is threshold values that must be set for most of the algorithms by the user. Usually, those values are based on previous experience and knowledge regarding parameter alterations within the drinking water supply system. Consequently, these values are subjective and may vary considerably. The appropriate setting of these threshold values has a crucial effect on the accuracy and sensitivity of contamination detection. This kind of approach relies on the knowledge of water utility staff.

PE and MA algorithms have shown the best contamination detection sensitivity results by processing the results from experimental studies. In certain cases the *PD* values reach 1.00, meaning that all of the contamination events have been detected and *FAR* values are 0.00, meaning that no false alarms have been triggered. In those cases, it can be concluded that the accuracy of those algorithms is reaching 100 %.

According to trends in the improvement of early warning systems within water supply systems, the only algorithm that is considered capable of not only detection with high sensitivity (*PD* = 1.00), but also classification between different types of contaminants, is the Mahalanobis distance (MA) algorithm. This algorithm does not include any threshold values. The disadvantage of the algorithm is the need for class definition that is related to long-term and repetitive contamination experiments in pilot-scale systems.

During the literature studies, none of the contamination detection algorithms has been considered as ready to be included in real scale and commercial early warning systems. In all of the studies, the need for additional research and evaluation of detection precision with experimental simulations of contamination events is reported.

In terms of all analyzed studies where the contamination detection algorithms have been tested in pilot scale water supply systems, they are tested with injections of specific chemical contamination solutions. No scientific studies have been reported where an experimental examination of microbiologic contamination event has been done. Also, microbiological parameters and methods have not been included in the evaluation of algorithms.

## **4.2. The alterations of drinking water quality parameter measurements during contamination events**

The average values of drinking water quality parameter measurements of clear and contaminated water samples allow evaluating their conformity to safe drinking water quality requirements. The results of drinking water contamination events show that using monitoring of physicochemical parameters and assessing their compliance with the requirements does not lead to the detection of the contamination event in most of the cases. The only scenario where any of the physicochemical parameter values exceeded the regulated values in Latvia and the EU is the wastewater contamination scenario. In this scenario, the average turbidity value of 5.26 NTU exceeded the regulated 3 NTU value. In addition, according to recommendations in Canada, where the permissible turbidity is 1 NTU, insufficient values would also be detected in the simulations of surface and groundwater scenarios. Only in those cases any deviations from the regulated values theoretically could be estimated by implementing routine drinking water quality monitoring. During the duration of all other simulations, the values of parameters that are regulated by the legislation did not exceed any limits.

The alterations of microbiological parameters are relatively large and are well represented during the contamination events. Nevertheless, the adenosine triphosphate and flow cytometry measurement methods and allowed concentrations are not regulated by law. It means that these parameters would not be monitored during routine monitoring programmes. Consequently, the contamination event would not be detected. The exceeded parameter values of cell count and indicator organisms most likely would be detected by classical cultivation methods.

The observed alterations of drinking water quality parameters indicate the potential of EC, turbidity, TOC, ATP and FCM implementation in contamination event detection, as well as relatively low alterations of ORP, *T* and pH parameters, except for the wastewater scenario where the noticeable decrease in ORP values was observed.

## **4.3. Definition of contamination types**

To implement the automatic drinking water contamination event detection Mahalanobis distance algorithm, according to methodology (Fig. 3.4, stage 1), it is necessary to define the classes for clear and contaminated water. The average *RB* (contaminated and clear water ratio of the relevant parameter) values of each contamination class are calculated using the methodology described above. If those values are arranged in a “spider” type chart, they form “fingerprints” of the relevant type of contamination (Fig. 4.1).

The parameters that were most specific for various contaminants are: the combination of NTU, FCM and ATP for the surface water contamination event, the combination of EC, ATP and NTU for the groundwater contamination event, ORP (towards the center), ATP and FCM combination for the wastewater contamination event, and the combination of ATP and FCM for the pathogenic bacterial model contamination event.



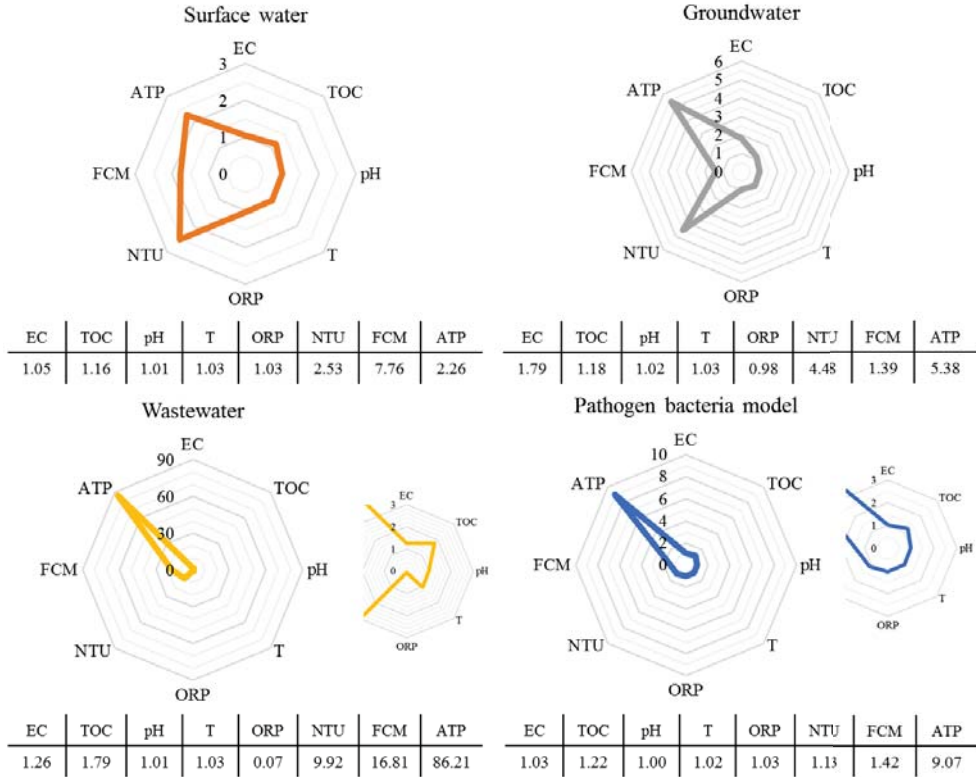


Fig. 4.1. *RB* values for classification of various types of contaminants.

Acquired *RB* values are combined in vectors consisting of the number of variables corresponding to the number of parameters in the number set. If all eight drinking water quality parameters are included in the algorithm, then the vector consists of a set of eight figures. For example, in the case of a surface water scenario, the vector classifying the contaminant is combined as  $p(V) = (1.05; 1.16; 1.01; 1.03; 1.03; 2.53; 7.76; 2.26)$ .

#### 4.4. The accuracy of contamination detection by single parameters

In order to identify parameters that show the highest contamination detection accuracy, the evaluation of the sensitivity of detection (*PD*), false alarm rate (*FAR*) and classification accuracy (*P*) was accomplished for measurements of each separate parameter. The comparison of classification accuracy is summarized in Fig. 4.2. The highest possible classification accuracy is 1.00, which refers to 100 % precise classification of clear and contaminated water measurements. The average classification accuracy acquired by measurements of separate parameters was 0.79 (79 % correct classification of clear and contaminated water). This result is close to the previously reported experimental assessments of early warning systems where 82 % accuracy was described.

The best contamination event detection and classification results were acquired by the implementation of EC and TOC parameters, which, in the case of all scenarios, have been identified with a higher classification accuracy than the average classification accuracy of each contamination scenario. The NTU, FCM and ATP parameters have shown such accuracy in three of four scenarios. During the contamination event with pathogenic bacteria model, the ATP, unlike all other parameters, demonstrated relatively high classification accuracy and therefore justifies its application in cases of specific contamination events. The acquired results of the sensitivity of contamination detection ( $PD$ ) by the implementation of pH (0.29),  $T$  (0.49) and ORP (0.25) parameters showed lower results of any sensitivity of contamination detection than previously described in the literature.

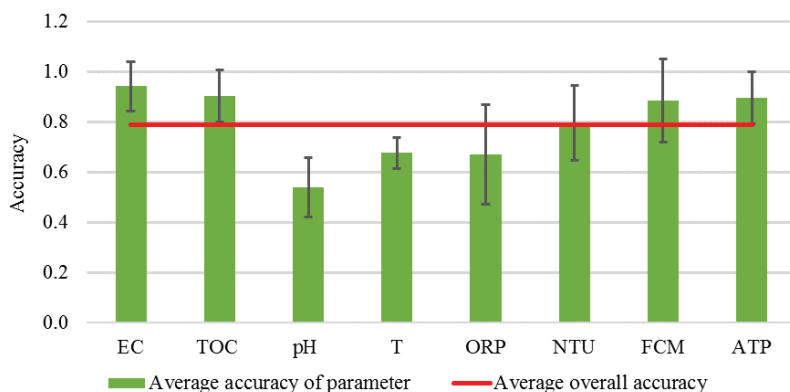


Fig. 4.2. Classification accuracy by the implementation of separate parameter measurements.

Moreover, in the previous studies, the authors acknowledge that the alteration of  $T$ , pH and ORP parameters during the normal conditions in real scale water supply systems are relatively high. Therefore, it is relatively complicated to distinguish between the normal alterations and contamination event created alterations. For that reason, it was decided to exclude  $T$ , ORP and pH parameters from the following contamination detection and data processing studies in this Thesis. The following data processing and contamination detection studies include EC, TOC, NTU, FCM and ATP parameters.

#### 4.5. The accuracy of contamination detection by various combinations of parameters

The measurements of the previously selected 5 parameters are combined in all 25 possible combinations. The purpose of setting up the combinations is to improve the contamination detection and classification accuracy. Also, the impact of microbiological parameter introduction in early warning systems is investigated. The obtained classification accuracy ( $P$ ) results are summarized in Fig. 4.3. In order to interpret the possible improvements more precisely, instead of standard deviations, there are a minimal and maximal classification accuracies acquired within the various contamination scenarios displayed (Figs. 4.3 and 4.4).

Therefore, it is possible to analyze not only the improvement of the average contamination classification accuracy but also its improvement in cases of different critical or less critical scenarios and situations.

The average drinking water contamination event classification accuracy ( $P$ ) obtained by implementation of various parameter combinations is 0.91. It is higher compared to the average classification accuracy of 0.79 acquired by measurements of separate parameters. It leads to the conclusion that the implementation of various drinking water quality parameter monitoring increases the contamination detection and classification accuracy compared to separate parameter monitoring.

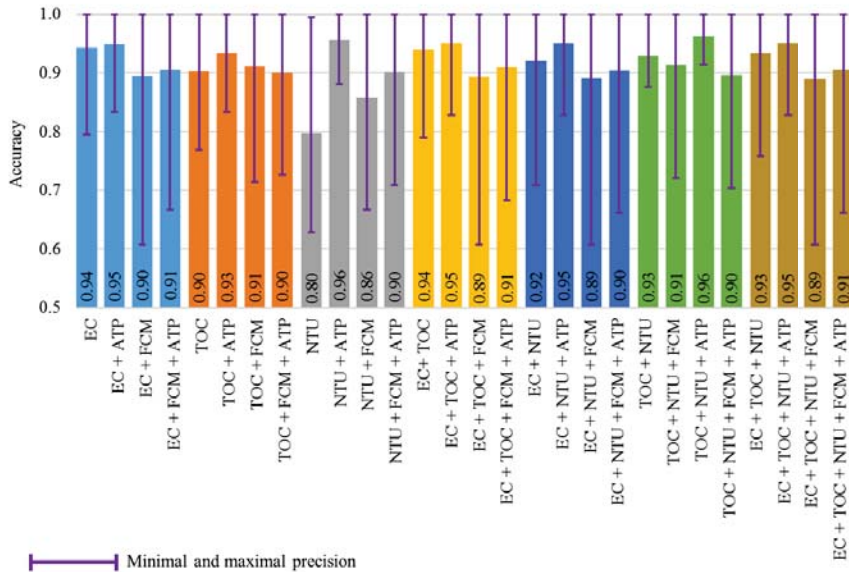


Fig. 4.3. Classification accuracy by the implementation of various parameter combinations.

The highest classification accuracy was acquired by NTU and ATP and NTU, ATP and TOC parameter measurement combinations. Accordingly, the measurement of those parameter combinations and the processing of the measurement results by detection and classification algorithm allows classifying the contamination events with an average accuracy of  $0.96 \pm 0.04$ . The combination of NTU and ATP measurements has classified the contamination event with 1.00 accuracy within the wastewater scenario and 0.99 within the groundwater scenario. In the case of pathogenic bacteria model scenario the accuracy was 0.95, and in the surface water scenario it was 0.88. Whereas the combination of NTU, ATP and TOC parameters showed an accuracy of 1.00 in the wastewater scenario, in the groundwater and pathogenic bacteria model scenarios it was 0.97, but in the surface water scenario 0.91. Although the results of the combination of both parameters are very similar, the combination of NTU, ATP and TOC parameters is chosen as the best combination because its minimum classification accuracy is higher. The minimum accuracy is critical because it

represents the possibility of detecting the contamination event within the worst case and preventing a supply of potentially insufficient quality water.

The influence of microbiological parameters to the contamination detection and classification accuracy is assessed by comparing the accuracy results obtained by combinations of physicochemical parameters with results obtained by these combinations joined with one or both microbiological parameters. The average drinking water contamination event classification accuracy results and improvements gained by the addition of microbiological parameters are shown in Fig. 4.4.

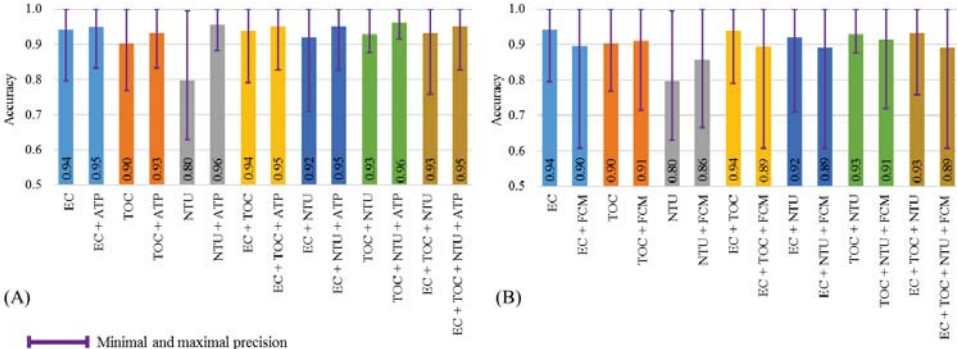


Fig. 4.4. The adjustments of classification accuracy by addition of ATP (A) and FCM (B) parameters to physicochemical parameter combinations.

The inclusion of microbiological parameters in a drinking water pollution classification algorithm does not provide unambiguous results. By the addition of ATP measurements to any combination of physicochemical parameters, the average accuracy of the contamination event classification is improved or maintained at an existing level. Moreover, in all cases of the addition of the ATP, the accuracy of the minimal contamination classification increased compared to the results obtained by relevant parameters. The addition of ATP to physicochemical parameter measurements improves the contamination classification accuracy so that it can be useful in drinking water quality online monitoring and early warning systems.

By the addition of the FCM parameter to physicochemical parameter measurements the accuracy of the contamination event classification can improve the accuracy results, but also in specific cases there has been the reduction of accuracy obtained. The addition of FCM to any combination of physicochemical parameters, in cases of surface water and groundwater scenarios, improved the accuracy of the contamination event classification on average by 0.05, in the case of the wastewater scenario, it was maintained at an existing 1.00 level, but in the case of pathogenic bacteria model contamination, it decreased by an average value of 0.16. Besides, the addition of FCM to measurements of physicochemical parameters may reduce the minimal accuracy of the contamination event classification that is obtained by the relevant parameter combination, consequently aggravating the total detection results of a contamination event.

In cases where both the ATP and FCM measurements are added to the measurements of physicochemical parameters, the average pollution detection accuracy remains at the current level. In surface water and groundwater scenarios, accuracy was improved by 0.02 and 0.08, respectively. In the wastewater scenario, it remained 1.00. However, in the case of the pathogenic bacterial model contamination scenario, it decreased by 0.12. Addition of both ATP and FCM measurements to physicochemical parameter measurements can improve the contamination event classification accuracy. However, the experimentally obtained results show unstable improvements in accuracy and even a reduction. The simultaneous inclusion of FCM and ATF parameters in the online monitoring and early warning systems of drinking water may not be useful.

#### **4.6. Contamination detection accuracy and operation of early warning systems**

Although in pilot scale experiments it is not possible to fully simulate potential contamination events in public water supply systems, this work has experimentally demonstrated the effectiveness and potential of the application of various drinking water quality monitoring parameters in the detection of diverse drinking water contamination events. For the correct interpretation of obtained contamination detection and classification results, it must be taken into account that it was acquired during limited and specific experimental setups and the contaminant flow was 10 % of the total flow in the system. In public water supply systems, contamination events may be more heterogeneous regarding flows and contamination periods. Experimental studies were carried out in a straight pipeline, without connections and branches that models a relatively simple part of the drinking water distribution system. In the real scale water supply systems, the contaminant concentrations, mixing with clear water and flows, are much more challenging to be predicted and detected. Besides, during experimental studies grab sampling and laboratory investigations were done instead of online measurements.

However, the experimental results and the contamination event classification accuracy of  $0.96 \pm 0.04$  by implementation measurements of ATP, turbidity and TOC parameters are relatively high if compared to the accuracy of the previously experimentally tested commercial early warning systems of 0.82. Those systems, by processing artificial datasets, can reach an accuracy of 0.98. Whereas the algorithms proposed by scientists (DSM, PE, MEK, MA algorithms), which are not yet included in commercial early warning systems, have shown up to 1.00 high contamination detection sensitivity (*PD*). These results are achieved by processing artificial measurement data sets or simulating chemical contamination events. In terms of this Thesis similar – 1.00 contamination detection sensitivity (*PD*) with simulations of real contamination events and subsequent measurement data processing (ATP, turbidity and TOC parameters) was achieved in wastewater and groundwater scenarios. Moreover, the simulated contamination event scenarios in this work are close to real conditions, thus showing detection of contamination even in relatively complex circumstances. Still, the cost of such online drinking water quality monitoring station sensors

and equipment could be relatively high. The online turbidity sensor costs approximately EUR 3000–6000. The price of the TOC online sensor depends on the method of analysis that is integrated into it, but it ranges between EUR 8000 and EUR 60 000. The ATP's online measurement equipment is not commercially available, but their approximate construction costs could be estimated at EUR 10 000–20 000. However, with improvements in the technological design, sensors might become more accessible, and equipment prices could be reduced that would lead to the increased application of them.

The results obtained demonstrate the possible use of the Mahalanobis distance algorithm in the detection of various contamination events within water supply systems. Still, before the application of the proposed solution in real scale water supply systems, the results of the contamination detection and classification must be approved in different circumstances by changing hydraulic and pollution parameters. As well as in future experiments, it is necessary to include online measurements that would bring their results closer to the application of selected drinking water quality parameters in early warning systems. The algorithm that was developed and adapted in terms of this Thesis can be easily automated and included in early warning systems. The disadvantage of the proposed algorithm is a large amount of data required to define pollution classes.

In most cases, the impact on consumer health after consumption of drinking water is related to insufficient microbiological quality on drinking water. Therefore, the research that is linked to the application of microbiological parameters in early warning systems is getting more topical. Unfortunately, there are relatively large alterations of drinking water quality parameters in water supply systems that may affect the detection of contamination events. Experimentally obtained alterations of ATP during normal operating conditions of the system were  $\pm 39.6\%$ . These alterations were relatively higher than they were for all analyzed parameters in this study. The addition of the ATP measurement to the physicochemical parameter measurements improves the contamination event detection and classification accuracy. In terms of experimental studies, it was proved that the potential solution of automated ATP monitoring equipment can improve the contamination detection and classification accuracy by online early warning systems. The automation of ATP measurements would lead to a relatively simple method for the continuous monitoring of microbiological quality of drinking water. This would allow to timely detect the contamination event and significantly reduce the risk to consumer health. However, the cost of the current method, compared to classical methods, is relatively high. To reduce the operational and maintenance costs of the ATP measurement application in real scale systems, the frequency of analysis might be reduced. Reducing the operational and servicing costs of the equipment, the regularity of the analysis of the ATP parameter may be reduced. Additionally, during the implementation of ATP measurements, there are liquid wastes produced that are required to be discharged into the sewer, therefore in real scale systems, it is necessary to provide a more sophisticated equipment installation design.

In previous studies, cases are described where with application of flow cytometry (FCM) and statistical analysis of measurement results it is possible to detect contamination events within experimental simulations of a number of microorganism increase by 4%. During

experimental studies within this Thesis  $\pm 23.1\%$  ( $6.59 \times 10^5 \pm 1.52 \times 10^5$  cells/mL) alterations of the number of cells in the normal operating conditions of the water supply system are similar to alterations of  $1.62 \times 10^5$  cells/mL to  $1.07 \times 10^6$  cells/mL reported in previous studies done in the Riga water supply system. The amplitude of such alterations in normal operating conditions prevented the detection of low-pollution events and reduced the accuracy of contamination event classification by the implementation of FCM measurements. Therefore, the effectiveness of the addition of FCM parameter to the physicochemical parameters for the detection of contamination incidents depends on the alterations of microorganism concentration in the water supply system during normal operating conditions. Before application of FCM measurements in public water supply systems, there must be comprehensive studies regarding the accomplished concentration of microorganisms in drinking water. FCM's relatively low results of contamination detection may also be associated with the presence of suspended particles or increased turbidity in water. During drinking water analyses, the suspended particles in FCM measurements may be treated as microorganisms, thus lowering the accuracy of the actual measurements.

Accurate detection of water supply system contamination events is not sufficient to motivate water utilities to apply online drinking water quality monitoring and early warning systems. The use of these systems currently is not economically feasible because of the relatively high installation and maintenance costs. However, a study carried out in the USA has shown that more than 30 % of all recorded gastrointestinal diseases may be associated with drinking water supply systems. Every year more than \$10 billion is spent for the treatment of these diseases. Therefore, investments in early warning systems cannot be cost-effective within the framework of water utilities, but they can be highly effective in terms of general public health and economic benefits.

The studies that have been carried out so far and further studies in the field of drinking water quality monitoring lead to more intensive applications of online and early warning solutions. Their possible implementation not only for the detection of contamination incidents but also for improvement of drinking water treatment and distribution would increase the number of applications all over the world.

## 5. CONCLUSIONS

1. The drinking water contamination event with an average classification accuracy ( $P$ ) of  $0.96 \pm 0.04$  can be detected by measuring an adenosine triphosphate, turbidity and total organic carbon in drinking water and processing the acquired data with the Mahalanobis distance contamination detection and classification algorithm. This implies that the presence of clean and polluted water in the system can be classified with the average accuracy of 96 %. In case of a wastewater intrusion, the contamination event can be classified with the accuracy of 100 %.
2. Temperature, oxidation-reduction potential and pH parameters tests demonstrate the lowest accuracy of contamination event classification —  $0.68 \pm 0.06$ ,  $0.67 \pm 0.20$  and  $0.54 \pm 0.12$ , respectively. These accuracies are lower than the average  $0.79 \pm 0.14$



obtained from readings of separate parameters. Therefore, the tests of these parameters are not suitable for a contamination event classification. Still, monitoring of these parameters can be used for the drinking water requirements evaluation.

3. By adding the data of adenosine triphosphate (ATP) readings to any combination of the physicochemical parameters examined in the study, the average classification accuracy (*P*) increases by 0.06 or 6 %. In case of the pathogen bacteria model contamination event, this can lead to the improvement of up to 0.32 or 32 %.
4. Addition of the flow cytometer (FCM) readings to any combination of the physicochemical parameters examined in the study do not influence the average accuracy of all kinds of contamination event scenarios. Though, in the case of the wastewater and surface water contamination events, the accuracy improves by 0.19 and 0.21 or 19 % and 21 % accordingly. The accuracy of the contamination event classification using the flow cytometry data is influenced by relatively significant fluctuation of the initial cell count  $\pm 23.1$  % within the day-to-day public water supply system operation.
5. Addition of the microbiological parameters and methods to the early warning system can enhance the accuracy of the contamination event detection. However, long-term readings of these parameters must be acquired to determine their natural oscillation.
6. Implementation of automated microbiological methods in the early warning systems would decrease the time necessary for microbiological contamination event detection from more than 18 hours, accordingly to current legislation, down to about 1–5 minutes. This could decrease the time the consumers could be supplied with the drinking water not complying with the drinking water requirements.

## **6. RECOMMENDATIONS AND FUTURE STUDIES**

Application of the early warning systems in the water-supply systems is inevitable as the urban population grows that poses potential threats to the water requirements. Consequently, scientific studies for the improvement of these systems are crucial.

This study develops and adopts detection and classification algorithm of the drinking water contamination events. However, only particular contamination scenarios have been tested. Furthermore, the algorithm should be improved with a contamination type classification step that would likely enable detection of the contamination source and type. To test and upgrade the algorithm with other types of contamination (like pesticides or toxic chemical compounds), repeated experiments with various pollution types, flows (flow rate of this study was 0.1 m/s), and concentrations shall be performed, as well as a contamination database should be developed. Moreover, the number of online drinking water quality monitoring stations and the place of installation in real scale water supply system have a critical impact on the contamination detection accuracy. Therefore, place and number of the online drinking water monitoring stations installation shall be well considered at the planning stage as faulty allocation could lead to system uselessness.

The best combination of the drinking water quality parameters has been identified in this study that comprises readings of the adenosine triphosphate, turbidity and total organic



carbon, which shall be suitable for the contamination event detection and classification. Unfortunately, in the experimental study, the grab sampling and analysis method were used, it means that the acquired results are not declaring the accuracy of contamination event detection with online sensors and real scale early warning system. Pilot scale experiments with the introduction of online sensor readings must be performed to make these statements affirmative. The major challenge of these experiments would be readings of the adenosine triphosphate due to limited availability of such online sensors in the market. However, scientific publications provide information regarding the list of prototype systems that could be applied. Automation of the adenosine triphosphate readings and making the equipment available for online applications would cause the extensive use of this method for the drinking water microbiological surveys. Moreover, increased supplies would decrease the price of these devices, thus making them more attractive for the water utilities.

In comparison to the classical cultivation methods, alternative and relatively quick methods of the microbial drinking water tests (like flow cytometry and adenosine triphosphate) significantly increase the quantity of collected information on the microbiological drinking water quality and its dynamics. Consequently, a list of pilot and real scale experiments could be executed that would allow exploring the probable correlation between dynamics of microbial and physicochemical parameters at day-to-day operation and contamination events. These methods shall be included in legislation to motivate their more extensive use. Likewise, currently flow cytometry is included in the legal list of the drinking water microbiological test methods of Switzerland.

Introduction of relatively expensive early warning systems depends on the motivation of water utilities to invest in new technologies and equipment. To raise the awareness of each water utility on potential threats, it is crucial to implement the risk assessment and evaluation approach. This kind of approach would allow the water utilities to recognize existing and potential risks and should motivate them to implement early warning systems. The risk assessment is a part of the Water Safety Plan strategy implementation regulated by the European Union. Though, implementation of these plans is voluntary under the law in force.

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