

Detection of drinking water contamination event with Mahalanobis distance method, using on-line monitoring sensors and manual measurement data

S. Dejus, A. Nescerecka, G. Kurcalts and T. Juhna

ABSTRACT

Concerns about drinking water (DW) quality contamination during water distribution raise a need for real-time monitoring and rapid contamination detection. Early warning systems (EWS) are a potential solution. The EWS consist of multiple conventional sensors that provide the real-time measurements and algorithms that allow the recognizing of contamination events from normal operating conditions. In most cases, these algorithms have been established with artificial data, while data from real and biological contamination events are limited. The goal of the study was the event detection performance of the Mahalanobis distance method in combination with on-line DW quality monitoring sensors and manual measurements of grab samples for potential DW biological contamination scenarios. In this study three contamination scenarios were simulated in a pilot-scale DW distribution system: untreated river water, groundwater and wastewater intrusion, which represent realistic contamination scenarios and imply biological contamination. Temperature, electrical conductivity (EC), total organic carbon (TOC), chlorine ion (Cl⁻), oxidation–reduction potential (ORP), pH sensors and turbidity measurements were used as on-line sensors and for manual measurements. Novel adenosine-triphosphate and flow cytometric measurements were used for biological water quality evaluation. The results showed contamination detection probability from 56% to 89%, where the best performance was obtained with manual measurements. The probability of false alarm was 5–6% both for on-line and manual measurements. The Mahalanobis distance method with DW quality sensors has a good potential to be applied in EWS. However, the sustainability of the on-line measurement system and/or the detection algorithm should be improved.

Key words | contamination detection, drinking water quality, early warning systems, on-line monitoring

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INTRODUCTION

Drinking water (DW) supply systems are vulnerable to deliberate and accidental water contamination events, and DW contamination is still an issue all over the world. For example in China, on average 1,906 accidents per year were related to DW contamination, according to data obtained over several years (Liu *et al.* 2015b). Contamination events with high impact on society were detected in the USA, e.g., the spill containing crude 4-methylcyclohexanemethanol in 2014

that contaminated the Elk River and influenced the water supply to 300,000 consumers (Liu *et al.* 2016). Moreover, at least 400,000 people were affected by the distribution of pathogens (Corso *et al.* 2003) during the outbreak reported in Milwaukee (USA) in 1993, which led to hundreds of hospitalizations and even deaths. Serious chemical contamination caused by ammonia spillage was reported in Tel Aviv (Israel) in 2001 (Winston *et al.* 2003) and contamination with

aluminum sulfate occurred in Camelford (UK) in 1988 (Altmann *et al.* 1999). In turn, 2,300 people became seriously ill, and seven died as a result of DW microbial contamination in Walkerton (Canada) in 2000 (Hrudey *et al.* 2003). These accidents resulted from a combination of different direct and indirect factors, including the human factor, and complete prevention of system failures is hardly possible. However, in most cases, the number of affected people could be smaller if the response to the contamination event was faster. Hence, it highlights a need for fast and reliable contamination detection systems.

On-line DW quality monitoring and early warning systems (EWS) were designed to provide remote and continuous DW monitoring and to notify the water utilities and consumers about contamination events in the DW supply system, and therefore to prevent possible contaminant exposure (Storey *et al.* 2011). An EWS consists of a DW quality sensors set, data collection and analysis system and alarm triggering algorithm (Liu *et al.* 2014, 2015a, 2016). Measurement tools usually consist of non-specific compound sensors, such as temperature, oxidation-reduction potential (ORP), pH, conductivity, etc. (Yang *et al.* 2009; Liu *et al.* 2015a). Advantages of non-compound specific sensors are their accuracy, relative simplicity, and low installation and operational costs. Moreover, it is not possible to predict the type of contamination which could occur in the system, and to install corresponding compound-specific sensors. Thus the precision and fast response of the non-compound specific sensors have higher contamination detection potential.

A critical factor for adequately working EWS is the detection algorithm (Liu *et al.* 2014). Mathematical algorithms have been developed to recognize contamination events between normal periodic fluctuations of DW quality. There are various emerging algorithms such as canonical correlation analysis (Li *et al.* 2016), minimum ellipsoid classification (Oliker & Ostfeld 2014a, 2014b), extended Dempster-Shafer method (Hou *et al.* 2013) and others, which differ in precision, reliability, and requirement of computing resources. The main principle is common to most methods. Event detection methods are evaluated by the trade-offs between false positive (FP) and false negative (FN) decisions as a function of the detection method, or in other words, by its ability to place the current measurement

of water quality parameters into one of two classes: background – clear and safe water, event – contaminated water (Liu *et al.* 2014, 2015a, 2016). The Mahalanobis distance method is an event detection and object classification algorithm, which is based on the assumption that similar objects have close values for a set of dimensions. If the distance from an object is shorter than the distances to other classes, then the object is deemed to belong to that class. The method is promising for DW EWS, since it has low false-alarm probability and high contamination-event detection precision, and allows classification of contamination events (Liu *et al.* 2015a). Moreover, this method has been already used for detection and classification of the most common water chemical pollutants reported in China (Liu *et al.* 2015a); however, the Mahalanobis method has not been tested for biological contamination detection. In general, there is a lack of on-line monitoring and EWS data from real-scale contamination events and particularly biological contamination detection (Perelman *et al.* 2012; Hou *et al.* 2013; Oliker & Ostfeld 2014a, 2014b; Liu *et al.* 2015a, 2015b). The latter is especially important, because biological contamination agents could cause illness of consumers in very short time, and therefore should be detected as soon as possible. Additionally, despite the generally high precision of non-compound-specific sensors, continuous measurements of flowing water might affect the accuracy of the measurements. Therefore, an experimental study of different biological contamination scenarios, close to real contamination conditions, was needed to evaluate the performance of previously proposed mathematical algorithms and systems.

The aim of this study was to evaluate the event detection performance of the Mahalanobis distance method in combination with on-line DW quality monitoring sensors and manual measurements of grab samples for potential DW biological contamination scenarios.

MATERIALS AND METHODS

Pilot-scale drinking water network

The pilot-scale DW distribution network (PSDN) was constructed (Figure 1). The PSDN is located in Riga Technical

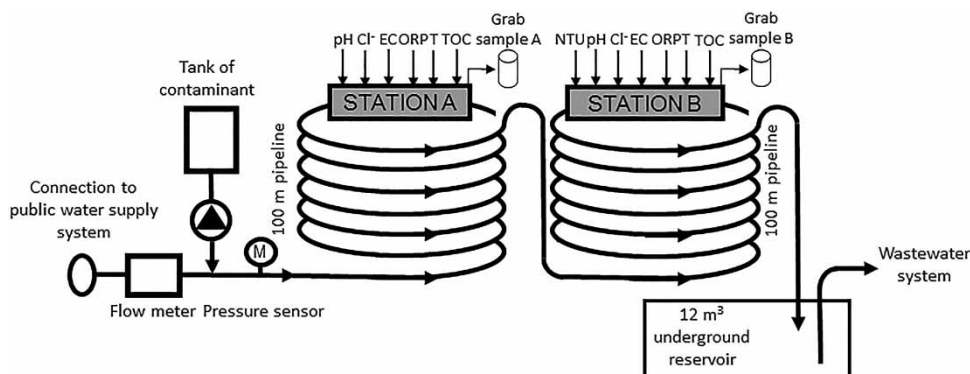


Figure 1 | Experimental setup of a pilot-scale DW distribution network.

University (RTU) Water Research Laboratory at RTU campus: water source – public DW supply system (approximately 12 km from DW treatment plant Daugava with a retention time of 15 hours), water outlet – water from the network is collected in a 12 m³ water reservoir below it and afterwards discharged into a public wastewater collection system. The PSDN consists of a 200 m pipeline: material – PVC, inner diameter – 25 mm, total volume – 98.2 l. Hydraulic conditions: flow – 0.20 m³/h, velocity – 0.1 m/s, pressure – 1 bar (limited by quality sensor durability). The PSDN has two on-line DW quality monitoring points; one is installed after 100 m from the connection to the public DW supply system, the other one after 200 m (100 m after point 1). Each of the monitoring points is equipped with temperature (T), electrical conductivity (EC), total organic carbon (TOC), chlorine ion (Cl-), ORP and pH sensors. Selection of the monitored parameters was described in the Dejus et al. (2015), where the main criteria were to keep low capital investments, low maintenance requirements and a good response to most possible contamination types, e.g. *Escherichia coli*, pesticides and arsenic contamination (Che & Liu 2014; Hou et al. 2014). Also, each of the monitoring points is equipped with a tap for grab sampling. Additionally, the turbidity is monitored at station B.

A contamination port in the PSDN is installed right after the water inlet and flow meter (Figure 1). It is equipped with a peristaltic pump LongerPump BT-600-2 J to maintain constant contaminant flow during the simulations of contamination events; the flow is set as 0.33 l/min.

Contamination scenarios

Three contamination scenarios were simulated. River water was added to the PSDN to imitate failure in the DW treatment plant. The addition of groundwater represented leakage and negative pressure in the supply pipeline, while wastewater was added to show DW supply and sewer system cross-connection (wastewater). All these scenarios represent the risk of biological pollution of the DW supply system. Contamination agents were added three times during each scenario with 2 h intervals between two events. For that 5 l of contaminants were added to the PSDN during each event within 15 min, which resulted in 10% (v/v) dilution in DW. On-line parameters were collected continuously, and grab samples for physical, chemical and biological measurements were taken at fixed time points (Table 1).

Table 1 | Grab sampling schedule: A – grab sample at monitoring station A, B – grab sample at monitoring station B (Figure 1)

	Time from addition of contaminant (h:min)													
Sampling station	0:00	0:10	0:15	0:20	0:25	0:30	0:35	0:40	0:45	0:50	1:00	1:30	2:00	
	A, B	A, B	A	A, B	A	A, B	B	A, B	B	A, B	A, B	A, B	A, B	

On-line and grab sample measurements were performed with different frequencies and various parameters were analyzed as shown in Table 2. On-line measurements were analyzed at 1 min intervals, and grab samples were taken with 5–10 min time steps, according to the sampling schedule, demonstrated in Table 1.

On-line measurements

On-line measurements were performed using EC, temperature (T), TOC, chloride (Cl^-), pH and turbidity sensors. EC, T and TOC sensors were developed by Adrona Ltd (Latvia); pH was measured with combination pH electrode HI1230B (Hanna Instruments Ltd), ORP – with ORP electrode HI3230B (Hanna Instruments Ltd), Cl^- electrodes – Ion Selective Electrode (ISE) K-27502-13 (Cole-Parmer Ltd). Relative instrumentation errors for sensors: EC: $\pm 2\%$, T: $\pm 3\%$, TOC: $\pm 2\%$, pH: $\pm 3\%$, ORP: $\pm 2.5\%$, Cl^- : $\pm 2\%$. Pressure sensor – MBS 3000 (Danfoss), flow meter FLOW 38 (COMAC CAL s.r.o.). For turbidity measurements, a TubiGuard photometer with additional logging machine Sicon (Sigris Photometer AG) was used. Calibration of sensors was done before the experiments. The data from the on-line monitoring system was read with a time step of 1 min, thus representing the instantaneous value measured for each parameter. Data from the sensors were read, processed and stored directly on a personal computer (PC) as .csv (Microsoft Excel Comma Separated Values File) files allowing integration of the data into various modeling, processing or warning tools.

Intensive grab sampling and manual measurements

The grab samples were taken in parallel with on-line measurements with time steps of 5 or 10 min based on the

phase of the experiment (Table 1). More frequent sampling (5 min intervals) was performed within a period between 5 min before and 15 min after the predicted time of the contamination event at each point depending on the water residence time at each of the stations and the duration of the contamination event. The total volume of the sample was 100 ml, which was divided into smaller volumes to measure different parameters. The schedule of grab sampling at each point is shown in Table 1. The grab samples were analysed for TOC (Formacs^{HT} TOC Analyser, Skalar Analytical B.V.), EC, temperature, ORP, pH (Multimeter Multi 9420 with Tetracon[®]925, SenTix ORP 900 and SenTix[®] pH 940 electrodes, WTW Xylem Analytics Germany Sales GmbH & Co.), turbidity (2100P ISO Turbidimeter, Hach Company Ltd) – identical to the on-line monitoring system. Also the adenosine triphosphate (ATP) and flow cytometric (FCM) total and intact cell counts were measured to monitor biological parameters.

Fluorescent staining and FCM of water samples

FCM analysis was based on methods described previously (Berney *et al.* 2008; Hammes *et al.* 2008; Prest *et al.* 2013). For total cell count (TCC) staining, a working solution of 100x diluted SYBR[®] Green I (SG) was used; 1 mL of the sample was stained with SG working solution at $10 \mu\text{L mL}^{-1}$ and incubated for 10 minutes at 35°C before analysis. For intact cell count (ICC) staining, propidium iodide (PI; 30 mM) was mixed with the SYBR[®] Green I working solution (SGPI) to a final PI concentration of 0.6 mM; 1 mL of the sample was stained with SGPI at $10 \mu\text{L mL}^{-1}$ and incubated for 15 minutes at 35°C before analysis. Before FCM analysis, the water samples were diluted (10% v/v) with $0.22 \mu\text{m}$ filtered commercially available bottled water (Evian, France). FCM measurements were performed on the CyFlow[®] SL, equipped with a blue 25 mW solid-state laser emitting light at a fixed wavelength of 488 nm. Green fluorescence was collected at $520 \pm 10 \text{ nm}$, red fluorescence above 630 nm, and high-angle sideward scatters (SSC) at 488 nm. The trigger/threshold was set in the green fluorescence channel and data were acquired on two-parameter density plots, while no compensation was used. All data were processed with the FCM propriety software, and

Table 2 | Analyzed parameters with on-line sensors (on-line) and manual measurements (grab sample), where X – parameter was analyzed, O – parameter was not analyzed

Parameter	EC	TOC	pH	ORP	T	NTU	FCM	ATP	Cl^-
On-line	X	X	X	X	X	X	O	O	X
Grab sample	X	X	X	X	X	X	X	X	O

electronic gating was used to separate positive signals from instrument and water sample background (Prest et al. 2013).

Adenosine triphosphate (ATP) analysis

Total ATP was determined using the BacTiter-Glo™ reagent (Promega Corporation), and a luminometer (Hygiene™) with minor adaptations from the method described elsewhere (Hammes et al. 2010; Nescerecka et al. 2016). In short: a water sample (750 µL) and the ATP reagent (50 µL) mixed with 8 µL of 1 M MgCl₂ prior to analyses were warmed to 38 °C simultaneously in separate sterile Eppendorf tubes. The sample and the reagent were then combined and then the luminescence was measured after 20 s reaction time at 38 °C. The data were collected as relative light units (RLU) and converted to ATP (nM) by means of a calibration curve made with a known ATP standard (rATP, 10 mM, Promega Corporation). For extracellular ATP analysis, each sample was filtered through a 0.1 µm sterile syringe filter (Millex®-GP, Millipore), followed by analysis as described above. The intracellular ATP was calculated by subtracting the extracellular ATP from the total ATP for each sample.

Mahalanobis distances for event detection method and performance evaluation

The Mahalanobis distances contamination detection method is based on cluster analysis. This is a process of grouping objects into previously defined classes with similar parameters. In this study, two classes were defined: clean water, parameters of which were typical for water without contamination (DW), and contaminated water. A set of the parameters, measured for the sample, is defined as one object in a multidimensional space, where the number of dimensions correspond to the quantity of parameters and the scale is normalized relative to all parameters. The distance from the current object to the previously set class (point in multidimensional space) is calculated. If the distance from an object is shorter than the distances to the other classes, then the object is deemed to belong to that class (Liu et al. 2015a). Mahalanobis distance can be used to identify and gauge the similarity of an unknown sample set (object) to a known one (class).

If object $p = (p_1, p_2, \dots, p_n)$ and class $c = (\mu_{c1}, \mu_{c2}, \dots, \mu_{cn})$, where μ_c is the mean of all instances in class c , are points in n -space, then the Mahalanobis distance from p to c or from c to p is given by

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

where D_M is the Mahalanobis distance, S is the covariance matrix of q , T is the transposer. The distance to the previously assumed and set amount of classes is calculated. It is deemed that the object p belongs to the class with the shortest distance D_M to it.

For the convenience of data comparison, the absolute values of the parameters were expressed as relative changes of each parameter:

$$RB_x = \frac{R_x}{B_x} \quad (2)$$

where R_x is the raw reading or measurement of parameter x , B_x is the average value of the baseline for parameter x , and RB_x represents the relative change of the sensor against its baseline (Liu et al. 2015a). A mathematical algorithm addressing the estimation of D_M for each measurement time step within MatLab R2016 software was developed specifically for this study.

To evaluate the ability of contamination detection by on-line and grab sample monitoring, a performance test was done. It consisted of the calculation of true positive rate (TPR) and false positive rate (FPR) values by

$$TPR = \frac{TP}{TP + FN}, \quad (3)$$

$$FPR = \frac{FP}{FP + TN} \quad (4)$$

where TP (true positive) is the detection of an actual event, FP (false positive) is the setting of the alarm on if there is no contaminant in the system, FN (false negative) is the situation when an actual event is not detected, and TN (true negative) is the keeping of the alarm off while there is only clear water in the system (Perelman et al. 2012; Arad et al. 2013; Liu et al. 2014).

RESULTS AND DISCUSSION

Response of different parameters to contamination

During the study 195 samples were analyzed manually, and 722 were measured on-line during simulation experiments with three contamination scenarios. Most of the monitored parameters showed changes during contamination events, and the extent of changes depended on the type of contaminant. As expected, the most dramatic changes were observed during the wastewater scenario experiment. A representative example of a response to contamination is demonstrated in Figure 2. EC, TOC, ORP, turbidity, FCM-TCC and ATP measurements of grab samples showed clear peaks during water contamination and indicated biological contamination. Strong correlations were obtained between these parameters (not shown). However, temperature and pH did not show any specific fluctuations in the presence of a contaminant. Interestingly, ORP changes were obvious only at station A (15 min water retention

time), and no changes were observed at station B. Further study is needed to explain the relation between changes of ORP and water retention time, and whether this parameter is critical for DW monitoring.

For quantitative comparison of different parameters with different contamination scenarios, RB_x values were calculated for each measurement. At each on-line reading time step and for each grab sample, the RB_x values have been computed and merged to create a multidimensional vector for analysis of Mahalanobis distance. Average raw reading values and RB_x values for each type of contaminant are demonstrated in Table 3. TOC, turbidity, EC, ATP and FCM-TCC showed the best response to all types of contamination. The deviations regarding clear water parameters ($RB_x = 1$) for TOC were in the range 0.17–0.67, turbidity: 0.82–5.33, EC: 0.01–0.11, ATP: 0.08–52.57 and FCM-TCC: 0.06–12.36. Similar to the graphical representation in Figure 2, RB_x values for the wastewater scenario were the most different from the baseline with an average RB_x value of 9.84, in comparison with 1.18 and 1.33 for river

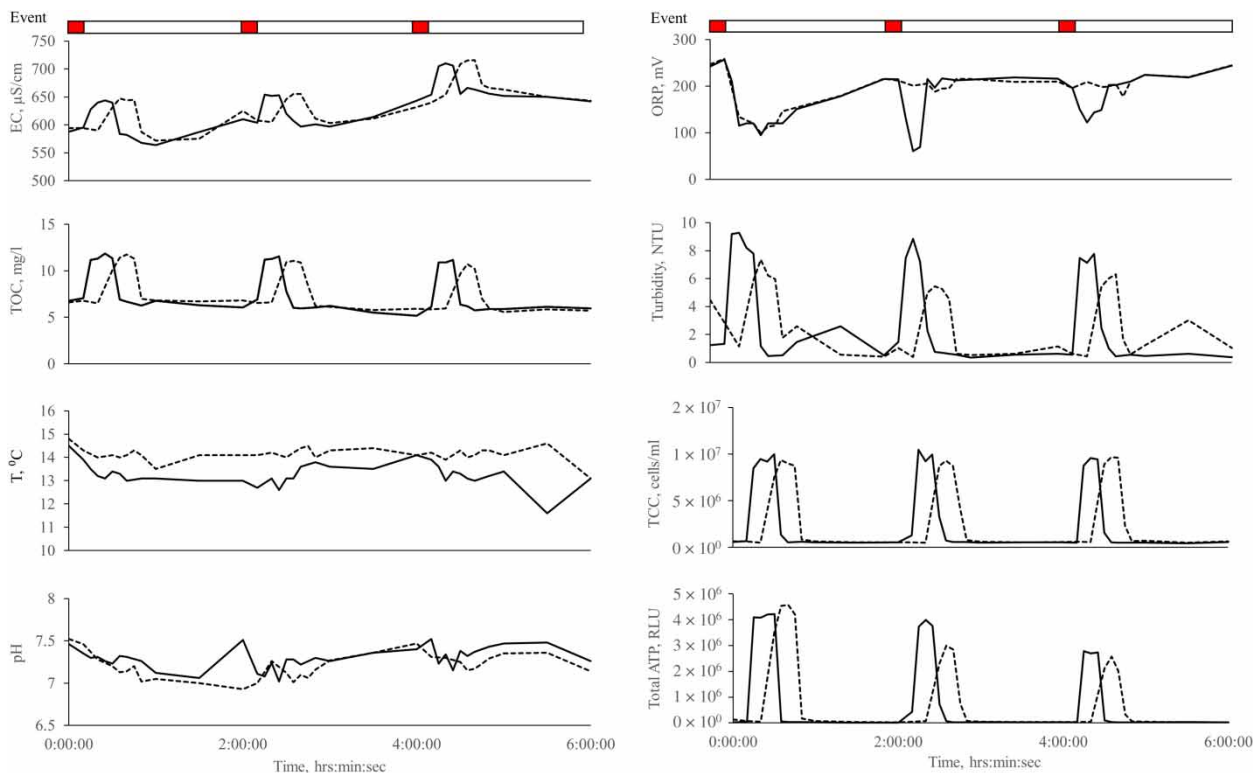


Figure 2 | Raw data from wastewater scenario with grab sample monitoring: solid line – station A (retention time 15 min), dashed line – station B (30 min), red bars – contamination event at contamination point. Please refer to the online version of this paper to see this figure in color: <http://dx.doi.org/10.2166/ws.2018.039>.

Table 3 | Relative values of grab sampling results for different types of contaminant expressed as RB_x values. Clean water measurements are shown as raw values ($RB_x = 1$). FCM-TCC – total cell count, ATP – total ATP

Contaminant	EC μS/cm	TOC mg/l	pH –	T °C	ORP mV	Turbidity NTU	FCM-TCC cells/ml	ATP RLU
Clean water	627	7.57	7.29	14.0	221.1	1.27	460,668	31,127
Relative values of the parameters against the baseline (RB_x) – dimensionless								
River water	0.99	1.17	1.00	0.98	1.02	1.82	1.19	1.29
Groundwater	1.11	1.21	0.99	0.98	0.99	3.37	1.06	0.92
Wastewater	1.07	1.67	0.99	1.00	0.70	6.33	13.36	53.57

water and groundwater events, respectively. Results show that the biological measurements tend to have higher deviations from baseline, which can enhance the detection of a contamination event. As far as biological methods were used only for grab samples, this means that the possibility of river water intrusion detection with on-line sensors is rather low. Nevertheless, TPR was 0.88 and FPR = 0.05 for grab sampling during the river water scenario, which confirms the ability of the proposed algorithm to detect even low-concentration contamination events. TPR and FPR values for the groundwater scenario with grab sampling were 0.91 and 0.19, respectively. Wastewater events showed the best results with TRP of 1.00 and FPR of 0.05.

Evaluation of detection system performance

The event detection algorithm with the Mahalanobis distance method was evaluated by determination of TPR and FPR values. TPR and FPR values were determined for overall system performance, considering all measured parameters (Table 4). The contamination detection was more reliable with grab sampling, resulting in TPR 0.89 (meaning that 89% of events should be detected), than the on-line monitoring, which detected only 56% of contamination cases. The false-alarm rate for both methods was similar: 5% and 6% for grab sampling and on-line monitoring, respectively. Detection performance by grab

sampling was rather similar to the TPR results of 0.62–0.78 for glyphosate, sodium fluoride and 0.52–0.94 for cadmium nitrate contamination reported in recent works (Liu et al. 2014, 2015a).

The on-line system showed rather modest results (TPR = 0.56) that could be explained by relatively high fluctuations of the values during normal system operation, i.e., without contamination. The fluctuations could be explained by measurement errors, related to instrumental errors, possible deviations from calibration points in long-term measurements, possible distortion of measurements by sensors installed nearby and signal processing from the sensor to system phase during the measurement. The relative instrumental errors for sensors used in the on-line system are higher than for sensors used in grab sampling, e.g., the instrumental errors for the on-line and manual measurement sensors were as follows: ORP: $\pm 2.5\%$ and $\pm 0.1\%$, EC: $\pm 2\%$ and $\pm 0.5\%$, pH: $\pm 3\%$ and $\pm 0.05\%$, respectively. These errors could influence the process of clear and contaminated class definition, for instance, as low as 1% deviation could indicate bacterial contamination, according to the data from Table 2. Possible distortion of measurements most likely could be related to the EC electrode, which releases electrical charge into the water, which can affect the measurements of nearby sensors. Also, the signal processing quality within the on-line monitoring system can be doubted regarding signal transposing from mV gathered from electrodes to units of certain measurement. The achieved results underline the importance of careful definition and description of the baseline class that could improve the accuracy of calculations and possibility of contamination detection. Thus, while different parameters showed a good response to contamination events, the on-line measurement system still has to be improved by implementation of more precise sensors and electrodes, evaluation of resilience of calibration in long-term experiments and

Table 4 | Results of evaluation of contamination detection performance

Type of monitoring	Number of measurements	Results					
		TP	FP	TN	FN	FPR	TPR
Grab sampling	195	64	6	117	8	0.05	0.89
On-line monitoring	722	54	35	591	42	0.06	0.56

measurements. Besides, despite the regular maintenance and cleaning of sensors, the possible impact of biofilm growth on electrodes should not be neglected.

CONCLUSIONS

- The Mahalanobis algorithm is suitable for EWS. However, it should be improved, considering normal baseline fluctuations to reach higher levels of confidence about contamination detection.
- The proposed method resulted in 89% positive detection alarms and 5% false positive alarms for biological contamination events, when measured manually, which indicates a good potential for the applied measurement tools and measured parameters to be implemented in EWS.
- The Mahalanobis distance method in combination with on-line sensors showed only 56% positive detection alarms, and 6% false positive alarms for biological contamination events. Although more than a half of contamination events were detected, the sustainability of the measurement system and/or the detection algorithm should be improved.
- Manual measurements showed better results than the on-line sensors. This highlights the importance of using high-precision sensors for continuous measurements and thorough maintenance and frequent calibration.
- TOC, turbidity, EC, ATP and FCM-TCC showed the best response to all types of contamination. The deviations regarding clear water parameters ($RB_x = 1$) for TOC were in the range 0.17–0.67, turbidity: 0.82–5.33, EC: 0.01–0.11, ATP: 0.08–52.57 and FCM-TCC: 0.06–12.36.
- Future studies and experiments with different pollutants, contaminant concentrations, flows and other physical and chemical properties should be done to define different contamination types, classes and determine the limits of the detection method.

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