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A statistical approach for identification of potential pollution incidents due to lignite mining activity in a surface water stream

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ABSTRACT

The present study aims to the development of a methodology for the identification of potential pollution incidents in surface water bodies, by the utilization of statistical techniques, using monitoring data collected from a multi-parametric sensors system. Demonstration of the proposed methodology carried out in a surface water stream located within a lignite mine area in Northern Greece; an on-line water quality monitoring system using multi-parametric sensors was installed in three representative sites of the study area, and the corresponding measured parameters included pH, temperature, conductivity, nitrate ions (NO_3^-), sulfate ions (SO_4^{2-}), and chloride ions (Cl^-) for a period of about 10 months. This system comprised of a probe containing the measuring elements (electrodes and sensors) and additional hardware/software for data collection, processing and transfer through the internet (wireless telemetry). The data collected from the monitoring station were treated using a statistical approach, aiming to reveal potential correlations between the physicochemical parameters and the volumetric flows of water drained from the lignite mines into the surface stream, in order to identify and justify pollution incidents from lignite mining activity. The results indicated that 14 potential pollution incidents were recorded during the entire monitoring period, between May 2010 and October 2010. Most active months within the study period, for the appearance of potential pollution incidents were May, June, and July for pH, Cl^-, and SO_4^{2-}; however conductivity was not considered as an index of potential pollution incident, since it was not correlated to significant changes in water quality. The detailed analysis of the periods where potential pollution incidents were identified revealed that similar trends in the variation of the physicochemical parameters were observed in 12 out of 14 cases, justifying the proposed statistical approach as a method for the identification of potential pollution incidents.

Keywords: Identification of pollution incidents; Lignite mining activity; Statistical technique; Surface water stream quality; Multi-parametric sensors system; Ptolemaida—Greece

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1. Introduction

Coal mining activities may have significant environmental impacts, affecting the nearby aquatic ecosystems. Drainage from areas with active or abandoned coal mines or from hazardous waste landfills where mining and byproducts from thermoelectric power stations (i.e. ash) are disposed result in wastewater runoffs with high pollutant content, contributing to surface and underground water aquifers contamination. Especially, the combustion of coal in thermal power stations for electricity generation results in the production of large amounts of combustion residues and particularly fly ash; these byproducts are accumulated in on-site piles and ash ponds within the mines, leading to serious environmental problems, and particularly in the contamination of ground and surface waters due to leaching of trace elements [1,2].

The effective monitoring of the water properties of surface and groundwater aquifers has become a requirement in these areas, aiming to the assessment of their quality according to the Water Frame Directive (2000/60/EC) [3]. The utilization of an on-line water quality monitoring system provides a database, containing information of water characteristics that can be further handled and published in an open access website. However, a large number of raw data are deduced by on-line monitoring sensors in these cases, making their interpretation a difficult task. Nevertheless, the experimental data can be subjected to further statistical analysis aiming to evaluate variations by time, to determine potential trends and more significantly to identify the occurrence of potential pollution incidents, if a significant correlation with other relevant parameters (i.e. drainage flow rate) can be established.

On-line monitoring sensors have been employed in several cases, for the characterization of water quality in environmentally sensitive areas; various statistical analysis methods have been utilized for data handling: data collected from various surface water bodies in the basin of the Nestos river in Greece were treated for the determination of potential trends using the non-parametric Spearman's criterion [4]. The effect of lignite mining activities on water quality was studied by other researchers, in the area of Ptolemais basin in Northern Greece; these workers used least squares linear models for the simulation of the experimental data by the time [1].

In another study, monitoring data from the Zarka river (Jordan) during the period 1988–2000 were used to forecast the water quality and quantity of King Talal Reservoir [5]. The exponential growth model yielded the least percentage of mean error for the prediction of TSS and T-P concentration, while the ARIMA model yielded the least percentage of mean error for BOD_5 forecasting. The COD content was best described by the quadratic method, the T-N was best predicted by the linear method and the auto regression method was used for the prediction of the volumetric flowrate [5].

However, limited research efforts have been paid towards the identification of potential pollution incidents using appropriate statistical techniques, considering a large number of on-line measurement data of surface water characteristics; the complexity and difficulty of this problem is straightforward.

The objective of this study was the on-line monitoring of the water quality of the aquatic resources in an environmentally "sensitive" lignite mine area of Ptolemaida basin in Western Macedonia Region—Northern Greece, through the installation and operation of an on-line water quality monitoring stations network, the examination of the water properties variation by time and the investigation of potential pollution incidents due to water drainage from mines and ash disposal sites.

The study area was the lignite mines area of Ptolemaida, located in Western Macedonia Region in the northern part of Greece. In this area, there are significant underground and surface water reservoirs (i.e. lakes Zazari, Cheimaditida, Petron, Vegoritida, Polyfitou etc.). The main activity that is directly or indirectly deemed to affect the quality characteristics of the aquifers is the operation of Public Power Corporation's (PPC) lignite mines and the corresponding thermal power stations. Other activities (agricultural, livestock, municipal etc.) might affect the water quality of the region to a rather limited extent, in comparison to the strong environmental impact due to PPC activity.

2. Materials and methods

2.1. *On-line water quality monitoring network based on multi-parameter sensors*

The continuous measurement of the water quality in the target area was carried out by the utilization of a multi-parametric sensor unit (Watertool II - Terramentor E.E.I.G.). This system comprises a probe containing the measuring elements (electrodes and sensors) protected by a robust cylindrical metal casing (Fig. 1). The system includes additional components for data collection and transfer, including the digitizing and control/storage unit and a data transfer unit. This apparatus can be used for the simultaneous measurement of up to 15 physicochemical parameters; however, in this study, the following six parameters were monitored: temperature,

Fig. 1. The multi-parameter sensors system (Watertool II).

Fig. 2. Special metal boxes constructed for the protection of the measuring equipment.

conductivity, pH, nitrate ions (NO_3^-), chloride ions (Cl^-), and sulfate ions (SO_4^{2-}). These particular physicochemical parameters were considered as representative of the water quality characteristics, as they were directly related to mine drainage pollution (particularly pH and SO_4^{2-}). The values of these parameters were recorded every 10 min. The concentration of the corresponding parameters was measured by ion-selective electrodes; while conventional open cell-type sensors were used for the measurement of conductivity and temperature [6,7].

The unit was equipped with appropriate hardware (wireless telemetry 3G ADSM) for data transfer through the internet. Furthermore, the unit was supported by software for automatic measurement of certain parameters, electrodes calibration, performance of diagnostic tests and verification, processing and visualization of the data. This software allows for early warning signals, when pollutant concentrations exceed certain predefined thresholds. Each multi-parametric unit was placed into specific metal frame insulated air-conditioned box (dimensions 1 m × 1 m × 1 m), in order to ensure their efficient operation and protection (Fig. 2). Calibration and operational control of the electrodes/sensors was performed for each electrode/sensor separately monthly, using standard solutions, in order to ensure the appropriate operation of the probes and the measurements reliability. However, sulfate ions electrodes were periodically calibrated by the supplier, depending on their performance efficiency [6–9].

The network of the monitoring stations consisted of three units. After a thorough study of the lignite mines area, three representative locations were selected for the installation of each unit, as shown in Fig. 3 [2,6]:

(1) The first monitoring station was installed in "Soulou" water stream near a bridge within the lignite mines area.
(2) The second monitoring station was installed in a water drilling within the lignite mine area (near the first station).
(3) The third monitoring station was installed in the effluent stream of Ag. Dimitrios PPC Thermal Station.

The main criteria taken under consideration for the selection of the most appropriate area for the installation of the monitoring stations were the distance from the lignite mines, the proximity to surface and ground water aquifers, previous measurement records, their accessibility, safety etc. All three monitoring stations were installed to the corresponding areas within March 2010.

Monitoring of the water quality of "Soulou" stream was considered as the most important process, due to its environmental significance; this stream received the discharges from lignite mines. Drainage from lignite mines due to rainfalls or groundwater inflows is collected in open ponds at the lower area of the mine. Water from each pond is then discharged to "Soulou" water stream by certain water pumps [10,11]. During summer period, the water from "Soulou" stream is used for irrigation of agricultural areas. "Soulou" water stream is discharged in Lake Vegoritis; "Soulou" stream and Vegoritis Lake are considered as "sensitive" water bodies, according to current national legislation [12].

In the current study, lignite mines drainage volumetric flow data into "Soulou" stream were utilized

Fig. 3. Location of the water quality monitoring stations in lignite mines area of Ptolemaida basin, Northern Greece (Source: Adapted by the authors from Map Background Image © 2015 CNES / Astrium © 2015 Google).

for the year 2010 from two drainage points: "Notio Pedio" lignite mine drainage point (onp) and "Kardia" lignite mine drainage point (kar), as presented in Fig. 3. However, water quality data measurements were not available for these points.

2.2. Statistical analysis

Raw data measurements were processed for their quality and consistency using non parametric descriptive statistics (box-plot analysis); therefore, a preliminary study took place aiming to exclude extreme values (i.e. due to electrodes operational problems, malfunctions etc.).

On-line measurement data recorded from "Soulou bridge" monitoring station were related to lignite mines drainage volumetric flow into "Soulou" stream for the two drainage points (kar, onp). Due to the long distance between the drainage points and the monitoring station, an appropriate time lag was calculated and considered in the analysis.

After preliminary data handling for the exclusion of extreme data, the monitoring period was divided into shorter intermittent time intervals, consisting of groups with continuous sets of records. As a result, a spreadsheet was prepared, containing data corresponding to all physicochemical parameters values and water volumetric flows for the two drainage points (onp, kar).

The values of the water volumetric flows in the two drainage points (onp, kar) were divided in two groups. The first group included all values higher than the overall mean volumetric flow rate per drainage point and per period examined, and was coded as 2, indicating a potential pollution incident. The second group included values lower than the overall mean and was coded as 1 representing a normal period with no incident occurring. This technique complies with the assumption that an increase in the corresponding volumetric flow regime of the drainage might change the concentration of the measured physicochemical parameters in the water stream.

Mean daily values of each physicochemical parameter were estimated per drainage point, allocated to incident groups 1 and 2, and were finally plotted as a function of time per each monitoring period. This procedure allows for the utilization of a two-factor analysis of variance (days and type of incident) and statistically significant effects were checked at 0.05 probability level [13]. When a significant effect was detected ($p \leq 0.05$), the mean values at each interaction level (time × incident) were plotted against time and group level, including the corresponding 95% confidence intervals calculated from the error mean square derived from the analysis of variance. A significant statistical difference between mean values was considered, when the confidence intervals did not overlap, in pair-wise comparisons [13]. The statistical procedure was performed via the MINITAB statistical software version 17.0.

It should be noted that the key point when applying the proposed methodology is to determine potential simultaneous significant changes in concentration and volumetric flow values for each examined time period, rather than to identify periods of high concentrations; such a correlation would indicate towards the occurrence of a pollution incident. The drainage volumetric flow classification in groups (1, 2) should be related to statistically different parameters concentration mean values (group 2 value > group 1 value) in the drainage points (kar, onp) in order to have an indication of potential pollution incident. All the available periods containing a complete set of physicochemical data were statistically analyzed for potential pollution effects.

Specific days within periods in which statistically significant effects were drawn, were further analyzed on a 24-h time frame for the detection of particular trends, related to parameters and drainage volumetric flow changes. Polynomial equations were utilized to describe potential trends by time, with parameters and volumetric flows being the dependent variables (Y) and the time variation (in minutes) the independent one (X). The coefficient of multiple determination of fit line was chosen as the main criterion of equation reliability; therefore, higher values of the coefficient (from 0 towards 100%) indicated improved model reliability. It was assumed that similar changes of patterns between volumetric flows and physicochemical variables with time indicate uniform conditions of effects and therefore approximately same relationships.

3. Results and discussion

The target area represents the primary area of coal exploitation in Greece for power generation. Residues from coal mining, as well as ash byproducts from thermal power stations are disposed in open areas resulting in significant environmental burden of the area. "Soulou" stream is a small river where drainage from the surface mines and the disposal sites is discharged; therefore, particular efforts were paid in this study, to monitor the water quality of "Soulou" stream, aiming to identify potential pollution incidents.

The approach used towards the determination of pollution incidents included the preliminary arrangement of raw data, according to their quality values and consistency, in order to identify common time periods where all available physicochemical parameters were measured. Unfortunately, due to operational problems, NO_3^- on-line measurement data were few and thus they were not considered in the following analysis. In the corresponding time periods, statistical analysis was carried out for the determination of potential correlations between the on-line physicochemical parameters data and the corresponding volumetric flow of lignite mines water drainage into "Soulou" stream.

It should be mentioned that "Soulou" stream is an open system, receiving influents from various external pollution sources, in addition to those associated with lignite mining activities, such as agricultural and livestock runoff, municipal wastewater and wastewater from other industrial activities; therefore interferences with mining activities cannot be excluded [6]. However, the development of the proposed methodology took place based on the assumption that pollution incidents were mainly attributed to lignite mining activity, rather than to other potential pollution sources.

The daily mean changes of water volumetric flow with time in drainage points kar and onp for the whole monitoring period, are presented in Fig. 4.

In the drainage point kar, the volumetric flow is rather constant up to October, following an increasing trend by the end of 2010 exhibiting a variation between 230 and 270 m^3/h. Four low and one high peak values were observed at certain time periods for drainage point onp, while a nearly constant pattern at about 160 m^3/h was observed for the remaining periods. The variation in the drainage water volumetric flow is attributed to the time-dependent operation of the water pumps in the lignite mines ponds, according to the requirement for discharging certain water volumes.

The corresponding water quality results are shown in Fig. 5. Each physicochemical parameter presents a unique profile by the time during the monitoring period. An increase in conductivity values in the period March–June is evolved, followed by an abrupt decline during August and a period of almost constant conductivity values. The pH values showed a similar

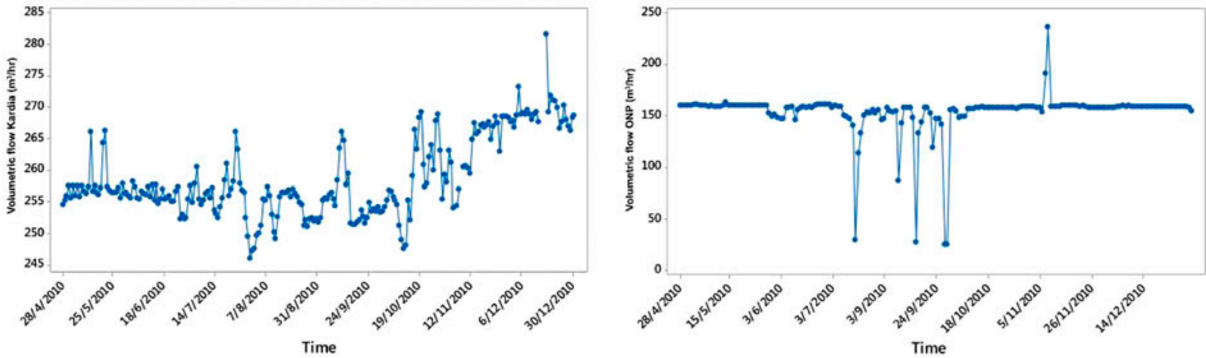

Fig. 4. Change of water volumetric flow (mean daily values) with time in drainage points kar and onp for the whole monitoring period.

pattern consisting of high values that were reduced to about 8; this pattern was repeated three times in the entire monitoring period. Chloride ions presented strong variations from August till October 2010. Sulfates, on the other hand, did not show any trend in their time pattern.

From the entire monitoring period, the preliminary analysis revealed certain time intervals where all parameter values were available; from these periods, 12 intermittent intervals were recorded for pH concentrations, 16 for chloride ions, 4 for sulfate ions and 10 for conductivity. Nevertheless, statistically significant indications of potential pollution incidents were justified in 13 time intervals. The variation of the daily mean values of the monitored parameters along with the corresponding days, categorized into two

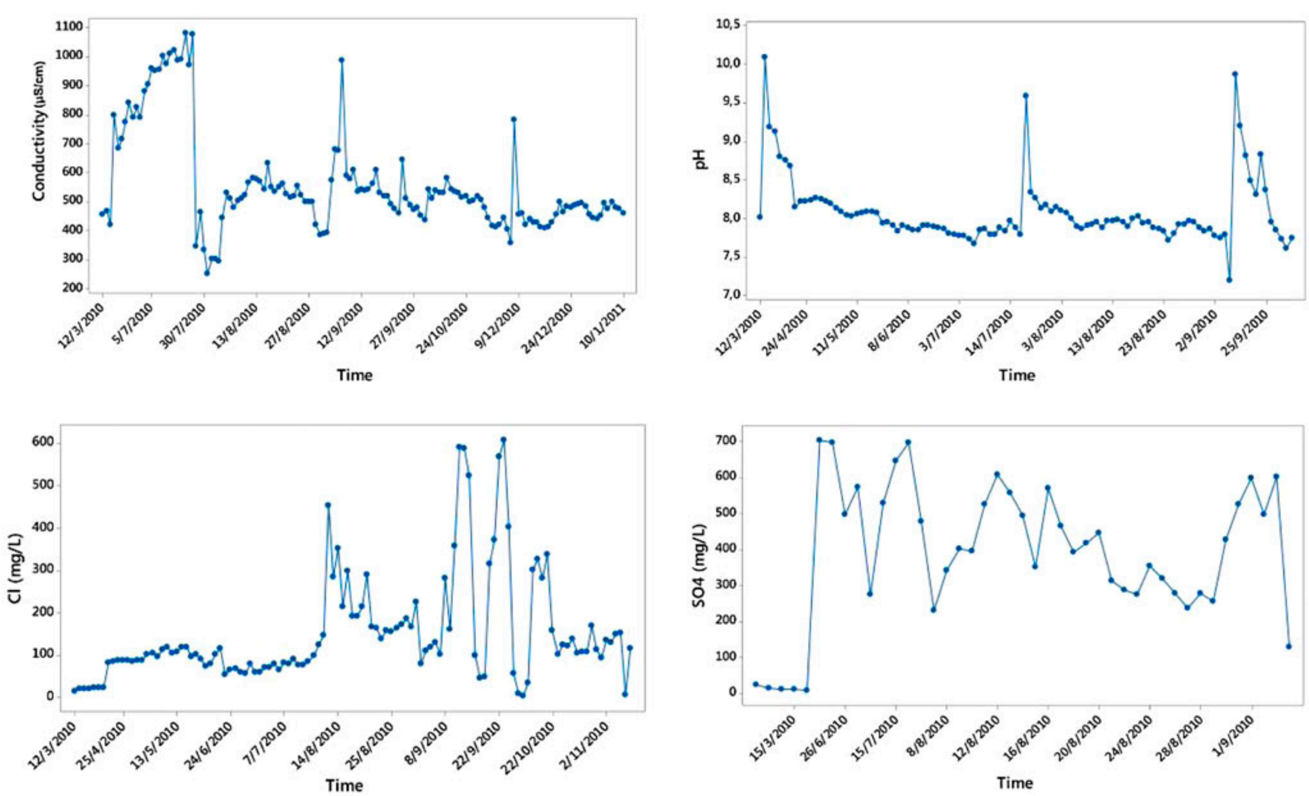

Fig. 5. Change of conductivity, pH, Cl^-, and SO_4^{2-} concentration (mean daily values) with time in “Soulou bridge” monitoring station for the whole monitoring period.

volumetric flow groups of incidents (1, 2) in drainage points kar and onp, are presented for each significant monitoring period in Figs. 6–9 for pH, chloride ions, and sulfates, respectively. The 95% confidence intervals of means, calculated from the error mean square of a two-factor analysis of variance, are presented in these graphs. The horizontal dashed red line represents the overall mean parameter value for the examined period. Parameters mean values corresponding to volumetric flow group 1 are graphically presented with continuous blue line, while parameters mean values corresponding to volumetric flow group 2 are graphically presented with dashed red line. The particular days where potential pollution incidents have been identified, are shown in a yellow circle in these graphs; these days were identified according to the proposed methodology, where a significant statistical difference was considered between mean values, when their intervals did not overlap, in pair-wise comparisons.

Specifically, four days (9 May 2010, 3 June 2010, 11 Juy 2010, and 28 August 2010) were detected with potential pollution incidents due to pH values variation, as presented in Fig. 6. Eight potential pollution incidents (7 May 2010, 12 May 2010, 5 June 2010, 2 July 2010, 3 July 2010, 16 July 2010, 25 September 2010, and 16 October 2010) were detected for chloride ions concentrations as presented in Figs. 7 and 8. The highest significant chloride ions concentrations, higher than 500 mg/L, were observed during autumn (end of September and middle October), as presented in Fig. 8. The lowest values, well below 100 mg/L, were observed early in June and July, as shown in Fig. 7.

Sulfate ions concentration overloadings were observed exclusively in the "Kardia" lignite mine drainage point (kar) during July (Fig. 9). Two statistically significant potential pollution incidents were identified on 5 and 16 July 2010, with the former presenting sulfate ion concentration three times lower (228 vs. 629 mg/L) than the latter.

The analysis of the conductivity values did not reveal any significant variation that could be attributed to potential water surcharge in the monitoring periods; thus, conductivity was not considered as an index of potential pollution incident. The mean values between the codes 1 (no incident) and 2 (incident) did not differ significantly at any day of the monitoring periods.

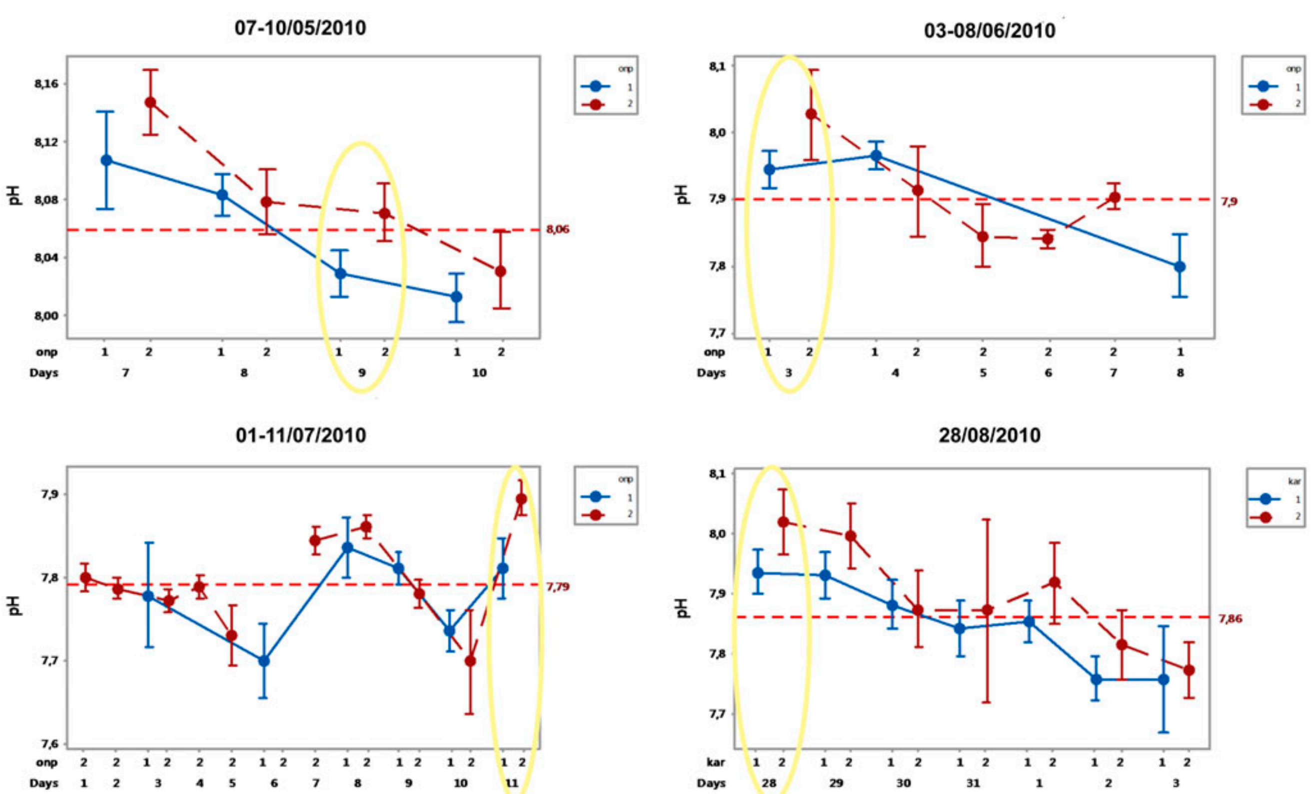

Fig. 6. Mean changes of pH along with the days of particular monitoring periods categorized into two volumetric flow groups (1, 2) in drainage points kar and onp.

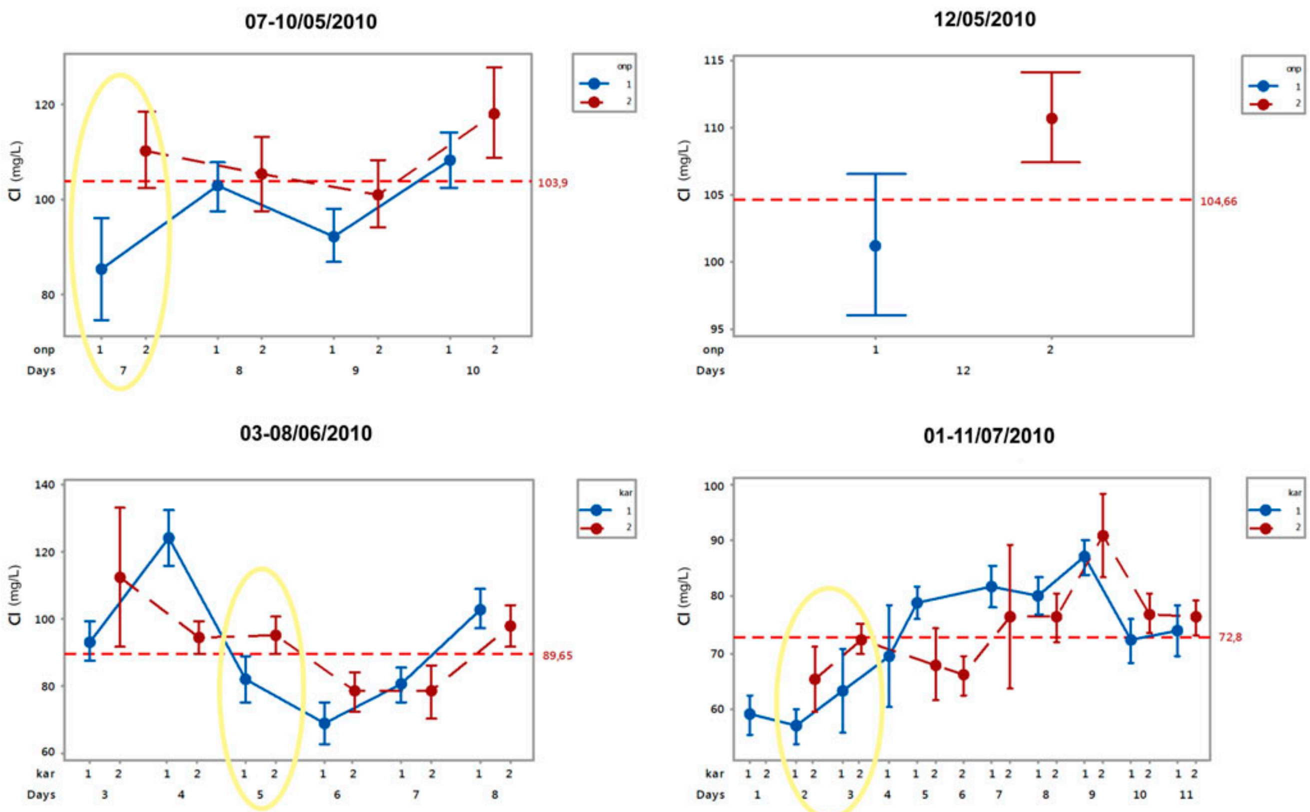

Fig. 7. Mean changes of chloride ions concentration along with the days of particular monitoring periods categorized into two volumetric flow groups (1, 2) in drainage points kar and onp.

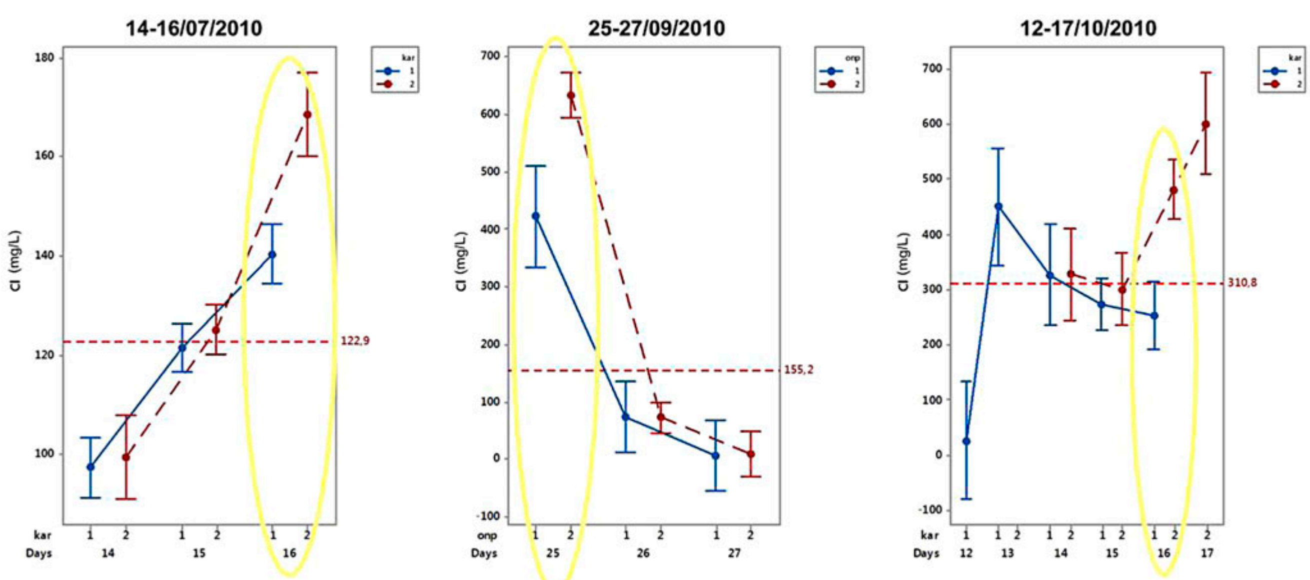

Fig. 8. Mean changes of chloride ions concentration along with the days of particular monitoring periods categorized into two volumetric flow groups (1, 2) in drainage points kar and onp.

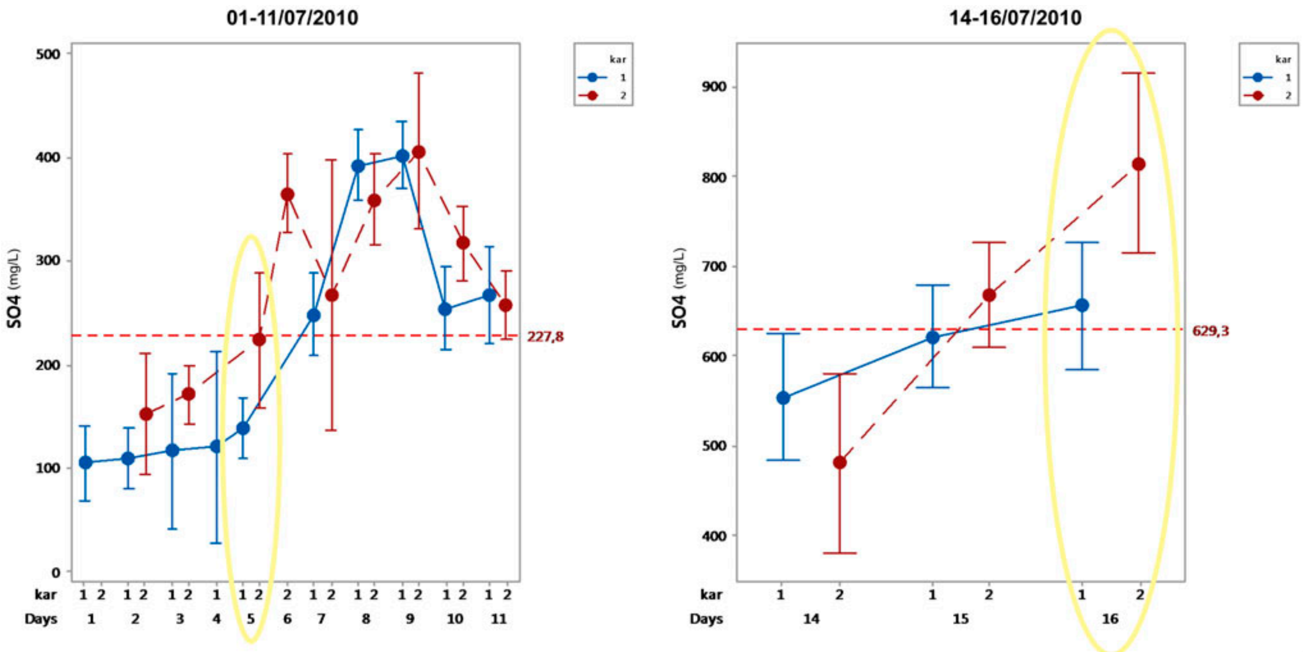

Fig. 9. Mean changes of sulfate ions concentration along with the days of particular monitoring periods categorized into two volumetric flow groups (1, 2) in drainage points kar and onp.

Fourteen potential pollution incidents observed during the whole monitoring period, between May 2010 and October 2010, are given in Table 1. Six of those were observed in "Notio Pedio" lignite mine drainage point (onp), and eight in "Kardia" lignite mine drainage point (kar). Potential pollution incidents were mainly observed during the following months: May, June, and July. Especially, pH and chloride pollution incidents were observed during May (7–10 May 2010) and June (3–8 June 2010), chloride and sulfate ions incidents during July (14–16 July 2010), while pollution incidents attributed to all three parameters were observed during July.

Data measured during time periods where evidence for pollution incidents was identified, as given in Table 1, were further analyzed aiming to the justification of pollution incidents. This approach involved the determination of potential relationships between physicochemical and drainage volumetric flow variations on a daily basis. It was assumed, that similar trends in daily variations would validate the occurrence of potential pollution evidence; the variation of a physicochemical parameter and drainage volumetric flow under a similar time pattern, would allow the establishment of a positive or negative correlation.

Efforts were given towards the development of polynomial equations in order to determine the best fit of the above variables by the time; second ($\hat{Y} = b_0 + b_1X + b_2X^2$) and third-degree ($\hat{Y} = b_0 + b_1X + b_2X^2 + b_3X^3$) equations were employed in this approach. These functions are clearly depicted in Figs. 10–12, corresponding to one or two curves, respectively. The reliability of each polynomial equation was checked against the coefficient of determination R^2 of fit line, and the corresponding results are given in Table 2. Values of R^2 approaching 100%, corresponded to points in each graph close to the polynomial fit line, resulting therefore, to equation functions with low variability [13].

As shown in Table 2, best fit polynomial equations were achieved in 12 out of 14 d, indicating that 12 pollution incidents were justified from the entire period of monitoring.

The pH relation to pollution incidents was completely validated in all 4 d as shown in Fig. 10, as deduced by three second-order and one third-order polynomial equations. The corresponding reliability varied between 30 and 90%, mostly due to the few pH values on a certain day, 3 June 2010 (R^2 = 29.6%).

Chloride ions concentrations showed similar positive correlation to water volumetric flow on a 24 h period as shown in Fig. 11. Two out of eight days were not validated as periods of potential pollution incidents, since no clear trends were detected by time, mainly due to the presence of few and sparsely distributed chloride ions records. Three second and three

Table 1
Summary of identified potential pollution incidents

A/A	Examined period	Date with identified potential pollution incident	Parameter	Drainage point
1	07–10 May 2010	09 May 2010	pH	onp
2	03–08 June2010	03 June 2010	pH	onp
3	01–11 July 2010	11 July 2010	pH	onp
4	28 August–03 September 2010	28 August 2010	pH	kar
5	07–10 May 2010	07 May 2010	Cl^-	onp
6	12 May 2010	12 May 2010	Cl^-	onp
7	03–08 June 2010	05 June 2010	Cl^-	kar
8a	01–11 July 2010	02 July 2010	Cl^-	kar
8b	01–11 July 2010	03 July 2010	Cl^-	kar
9	14–16 July 2010	16 July 2010	Cl^-	kar
10	25–27 September 2010	25 September 2010	Cl^-	onp
11	12–17 October 2010	16 October 2010	Cl^-	kar
12	01–11 July 2010	05 July 2010	SO_4^{2-}	kar
13	14–16 July 2010	16 July 2010	SO_4^{2-}	kar

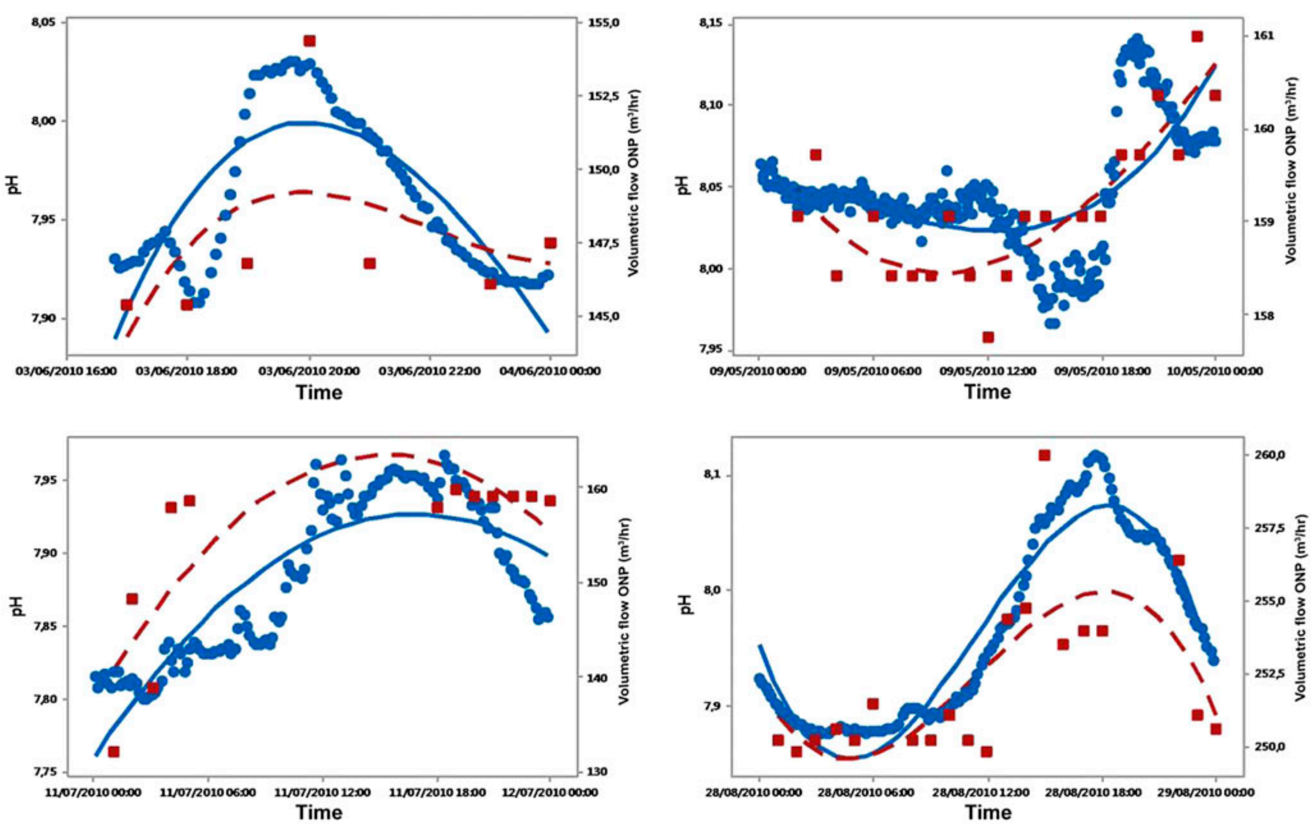

Fig. 10. Change of pH concentration (red line) and drainage volumetric flow (blue line) in days of potential pollution incidents on a 24-h basis.

third-order polynomial equations were established, showing a degree of reliability higher than 80% for the physicochemical parameters; however, the degree of reliability on 3 July 2010 dropped down to 41.7%.

A strong positive correlation was observed between the changes in sulfate ions concentration and drainage volumetric flow by time, as shown in Fig. 12. Two second-order polynomial equations were

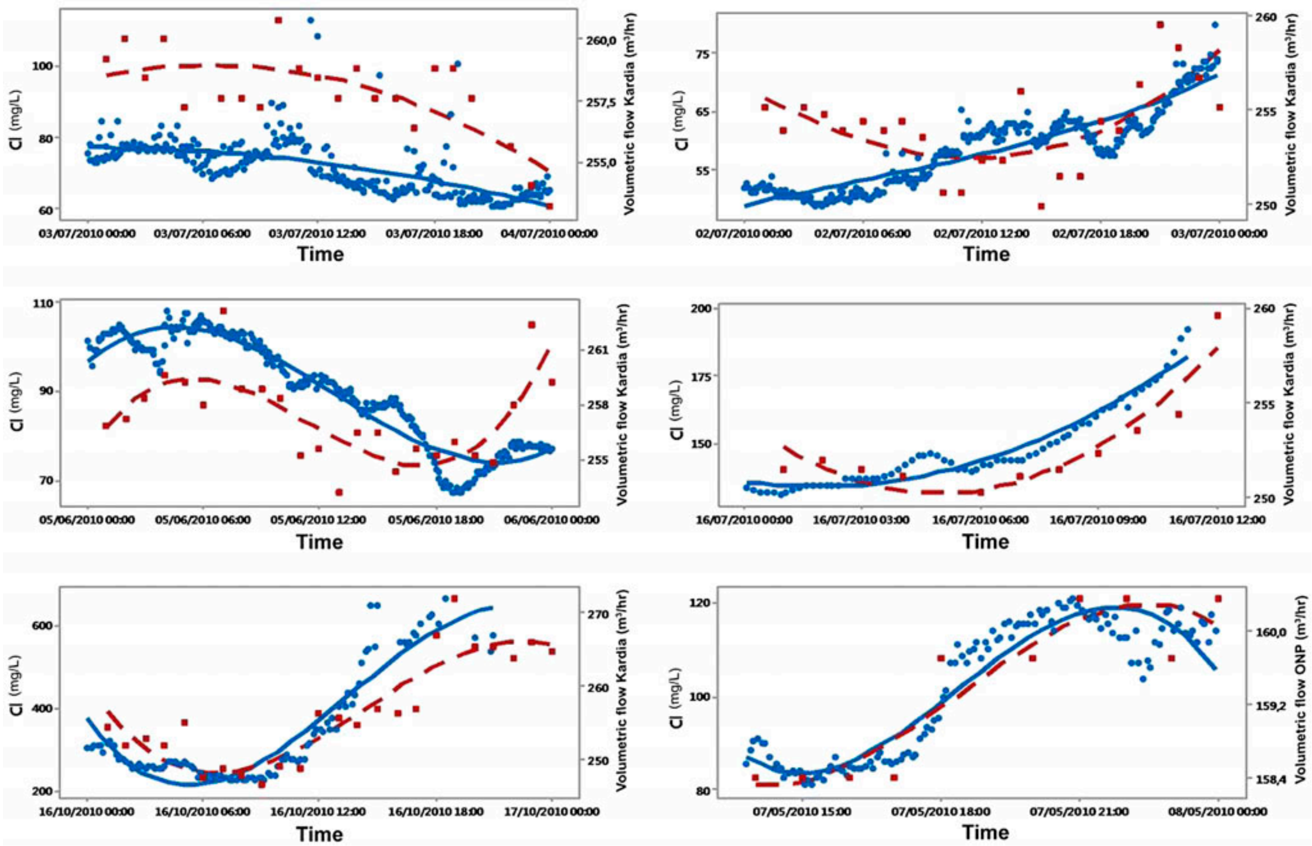

Fig. 11. Change of chloride ions concentration (red line) and drainage volumetric flow (blue line) in days of potential pollution incidents on a 24-h basis.

Table 2
Confirmation of identified potential pollution incidents

A/A	Date with identified potential pollution incident	Parameter	Confirmation of potential pollution incident	R^2 (%) Parameter	R^2 (%) Vol. flow
1	09 May 2010	pH	Yes	39.5	76.5
2	03 June 2010	pH	Yes	67.5	29.6
3	11 July 2010	pH	Yes	74.5	65.8
4	28 August 2010	pH	Yes	93.0	52.8
5	07 May 2010	Cl^-	Yes	89.1	86.0
6	12 May 2010	Cl^-	No	–	–
7	5 June 2010	Cl^-	Yes	92.2	59.9
8	2 July 2010	Cl^-	Yes	83.0	45.2
9	3 July 2010	Cl^-	Yes	41.7	55.0
10	16 July 2010	Cl^-	Yes	94.3	87.8
11	25 September 2010	Cl^-	No	–	–
12	16 October 2010	Cl^-	Yes	88.8	82.9
13	05 July 2010	SO_4^{2-}	Yes	84.5	41.1
14	16 July 2010	SO_4^{2-}	Yes	98.1	87.8

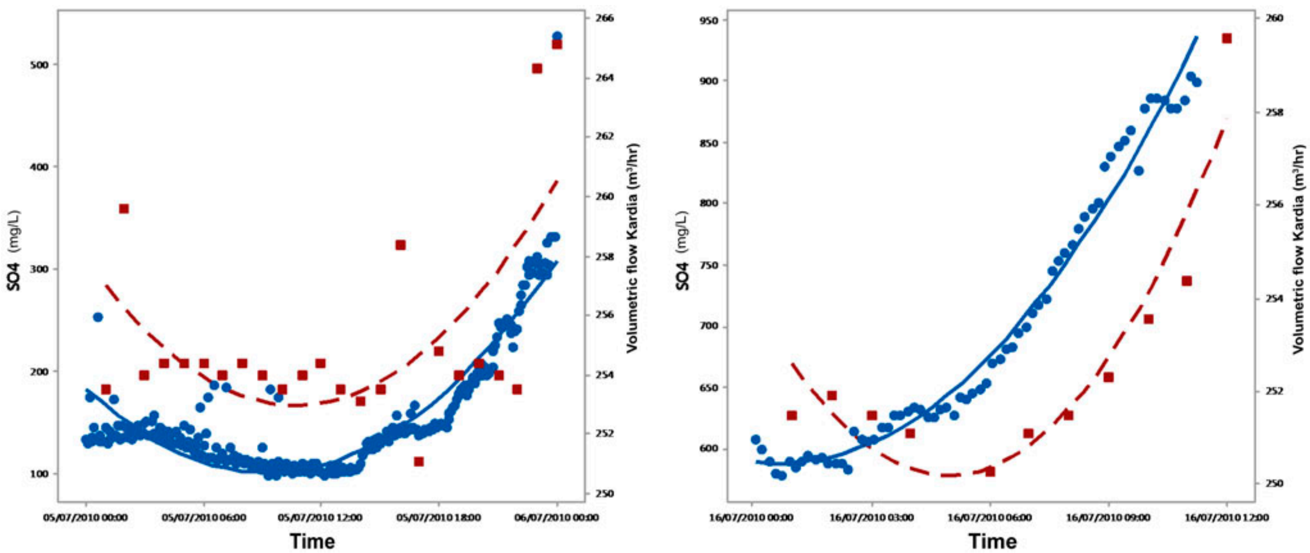

Fig. 12. Change of sulfate ions concentration (red line) and drainage volumetric flow (blue line) in days of potential pollution incidents on a 24-h basis.

produced with fit lines of high reliability (>80%) for the physicochemical parameters and high degree of proportional change by the time.

It can be concluded from the above analysis that the application of the proposed statistical approach into a large database of water quality values (containing about 80.000 data in the current study) can contribute to the identification of potential pollution incidents; nevertheless, such a huge amount of raw data is very hard to be handled with other conventional methods.

4. Conclusions

During the current study, a network of three multi-parameter sensor systems was installed in the Ptolemaida lignite mines region, in order to monitor the water quality of the aquatic resources of the area, and to investigate potential impacts due to pollution incidents. The data of physicochemical parameters concentration were collected for a period of about 10 months and were combined with available data of lignite mines drainage volumetric flow into a surface water stream, the "Soulou" stream, for the same period; certain statistical techniques were applied in order to develop a methodology for the identification of potential pollution incidents.

From the analysis of a large number of raw data, the following conclusions can be obtained:

(1) Potential pollution incidents were identified in monitoring intermittent periods, where the 95% confidence intervals of the physicochemical parameter mean values corresponding to the two drainage volumetric flow groups did not overlap in pair-wise comparisons (incident/not incident).
(2) Fourteen potential pollution incidents were revealed in the whole monitoring period, between May 2010 and October 2010, by applying the proposed statistical approach. Six of the potential pollution incidents were attributed to "Notio Pedio" lignite mine drainage point, and eight to "Kardia" lignite mine drainage point.
(3) Most active months for the occurrence of pollution effect were May, June, and July. pH and chloride ions pollution incidents were observed in May and June, chloride and sulfate ions in July, and all three parameters between first and eleventh of July.
(4) Conductivity did not reveal any significant trends during the monitoring period, thus it was not considered as an index of potential pollution incident.
(5) The detailed analysis of the days identified as periods of potential pollution incident revealed similar trends in drainage volumetric flows and physicochemical parameters variations, justifying therefore the occurrence of a pollution incident. The polynomial equations used in order to find the best fit of the corresponding variables, showed strong positive correlation between the change of pH, Cl^-, and SO_4^{2-} concentration and drainage volumetric flows; according to this approach, 12 pollution incidents were verified out of 14

potential ones. Thus, the proposed statistical approach could be used as a tool in order to identify potential pollution incidents.

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