

Article

Multisource Groundwater Contamination under Data Scarcity: The Case Study of Six Municipalities in the Proximity of the Naameh Landfill, Lebanon

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Abstract: Lebanon is affected by a protracted environmental and solid waste crisis that is threatening the water resources and the public health of its communities. This study is part of a public participatory research project that aims to evaluate the impacts of solid waste disposal practices on water, air, and health in six villages of Lebanon, stigmatized by the presence of a regional landfill. Community mapping enabled the selection and testing of seven springs and three wells in the upstream basin and 11 wells in the lower basin, covering a broad list of chemical, physical, and bacteriological parameters. Two water quality indices (WQ-1 and WQ-2) were used to assess water quality in the study area. The results for the upstream wells and springs showed a significant bacteriological contamination, while the results in the lower wells showed high levels of conductivity, chlorides, and zinc along with the occurrence of organic micropollutants in trace concentrations. The comparison between the experimental data, with the natural background value established in the same area, did not show major differences, except for zinc and bacteriological indicators. The bacteriological contamination is most likely related to sewage infiltration into groundwater at the time of the assessment. Zinc may result from landfill leachate infiltration but also well corrosion. Saltwater intrusion affecting the coastal basin is masking the results for conductivity, chlorides, and sulfates, whereas the presence of small traces of organic micropollutants in the coastal aquifer may be related to leachate infiltration. WQI-1 results, which included bacteriological indicators, showed highly degraded water quality in the C1-C3 inner basin. In contrast, WQI-2, which includes physio-chemical indicators only, showed good water quality, slightly deteriorating in the coastal area, downstream of the Naameh landfill.

Keywords: water quality; *E. coli*; community mapping; landfill; multisource contamination

1. Introduction

Nations and communities approaching the second decade of the new millennium are facing a multi-tiered challenge to improve their waste management practices and control the health impacts from the release of environmental pollutants [1]. Everyday news brings to light a new tragedy of the commons, exposing the public health consequences of unsustainable environmental practices, especially in the developing world. While the access to safe water as an inalienable human right has been recognized by the United Nations General Assembly Resolution 64/292 [2] and is at the center of the Sustainable Development Goals, there is still a long way to reach the targets of Goal 6: Clean Water and Sanitation for all by 2030 [3]. Lebanon is currently struggling with several socioeconomic and environmental challenges. These are linked, among other factors, to (a) a chronic environmental management deficit at the national level due to outdated regulations [4], (b) poorly implemented environmental policies [5], (c) the burden of a consumerist culture [6], and (d) the rapid influx and the permanence of 1.5 million Syrian refugees after the 2011 Syrian conflict [7]. As a result, several hazardous solid waste disposal and wastewater discharge practices are exacerbating the stressed Lebanese environmental resources and threatening the hosting and displaced communities' health [8,9]. Concerning solid waste management, most of the municipalities in rural areas are currently open dumping or open burning a significant amount of waste [10–12]. In addition, landfilling has been practiced as a solution for waste disposal for the Beirut broader metro area, home to over 2.2 million residents (half of Lebanon's population). Several international and some Lebanese studies have clearly demonstrated the negative effects of open burning, open dumping, and landfilling for the environment and public health of the nearby communities [9,13–15]. Another threat to the environment and public health is represented by poor sanitation in most of Lebanon, considering that 92% of the country's wastewater is discharged untreated in surface water bodies or septic tanks or cesspits [16]. Untreated wastewater discharge in the environment is particularly hazardous in mountain areas, often marked by a well-developed karst and by a rapid connection between surface water and subsurface flow over large distances, e.g., [17].

Understanding the effects of waste disposal across the Lebanese water systems is therefore crucial considering the abovementioned implications for public health. Geochemistry has been found as an important tool to differentiate anthropic and natural geochemical phenomena occurring in aquifers [18–21]. At the same time, there is a growing consensus on the need to integrate scientific investigation with community knowledge and concerns [22,23].

This study is part of a public participatory multidisciplinary research described in [24]. The work was initiated by six municipalities in Aley District in the aftermath of the closure of the regional landfill of Naameh in 2015, which led to a national solid waste crisis that spanned till 2016 (Waste Crisis 2015–2016 [10]). The municipalities reached out to the American University of Beirut (AUB) for support in assessing the environmental (air, water, and health) damages from waste disposal and improve their waste management practices [24]. In particular, their interest stemmed from a public health scare that emerged from the long-awaited closure of the landfill after years of operation beyond its design capacity. The present study endeavors to answer two questions raised by the six communities:

- In view of the solid waste crisis of 2015–2016, are there any residual damages on the quality of surface and groundwater resources (springs and wells)?
- What are the other threats affecting the water quality in the area?

To answer these research questions, a water quality testing campaign was performed during high flow period in the hydrological year (November and April). A large spectrum of physical, chemical, and microbiological parameters were tested for 18 water sources. While national published studies focused on anthropic impacts on water resources and related public health implications [14,25–27], no research attempted to establish the effects on the groundwater resources from landfilling, open dumping, or wastewater disposal. This study is therefore the first of its kind in Lebanon.

Moreover, our data and approach have some broader implication: data scarcity undermines the capacity of local authorities and communities in the developing world to make decisions based on sound evidence [28]. Hence, there is a particular need for site-specific water quality assessments in a data-scarce context like Lebanon for well-informed water management strategies.

2. Materials and Methods

2.1. Study Area

The study area is located 15–20 km south of Beirut, Lebanon. It covers the cadastral area of six municipalities in Aley District, Mount Lebanon (Figure 1). The climate is Mediterranean, characterized by hot, dry summers, and short winters [29]. The annual rainfall in the area is between 700 and 900 mm [30]. The major surface water body in the area is the Damour River on the southern border of the area, while the central and the northern parts are characterized by the presence of seasonal surface water bodies (gullies and streams).

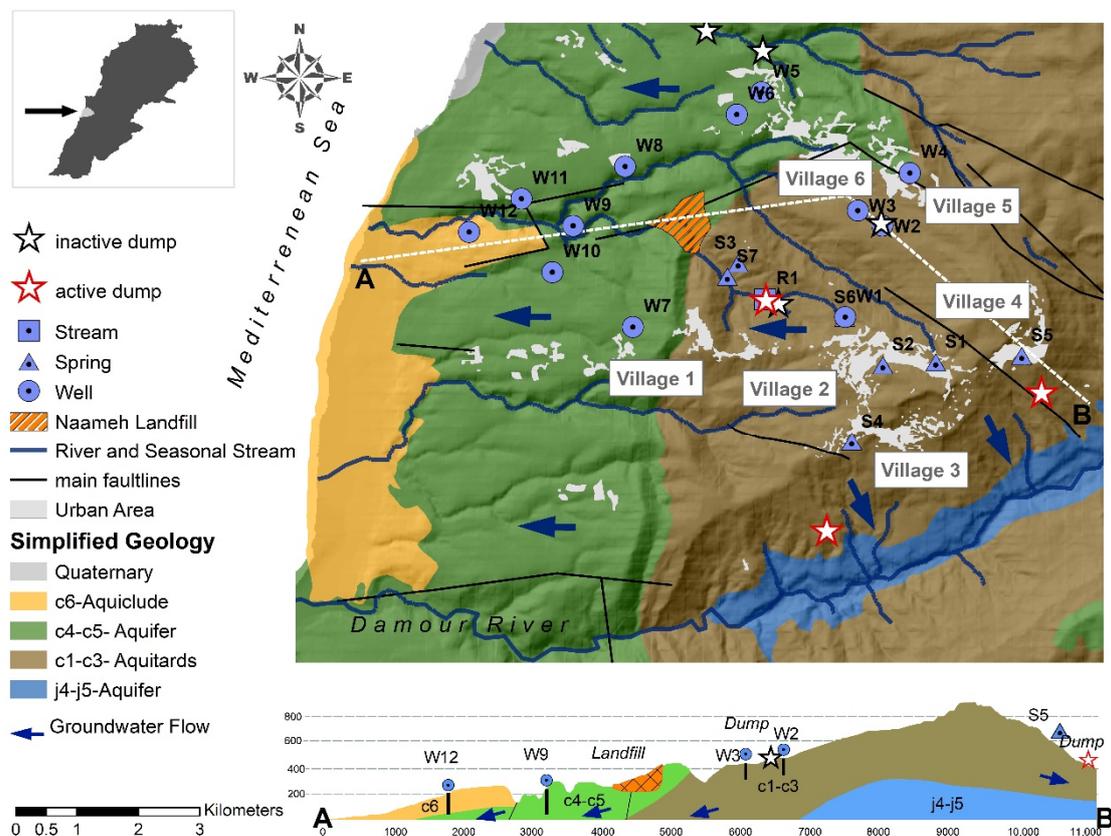


Figure 1. Geological map and profile sketch of the study area, location of water sources, and solid waste hazards. Geology data from [31] Centre National de la Recherche Scientifique (CNRS) and hydrogeology from [32] United Nations Development Program – Ministry of Energy and Water (UNDP-MOEW, 2014) and [33] Khadra 2014.

As shown in Figure 1, the area is geologically located in the flank of a regional anticline, giving a general slope of the layers EW dipping [31–35]. The coastal area is defined by the C4-C5 aquifer, part of the 19b Sarafand-Khaldi Cretaceous basin [32]. This aquifer is a strategic resource for the local communities and the Beirut metropolitan area (supplying around 40% of its water demand) [32]. The C4-C5 coastal basin has been facing saltwater intrusion since the 1960s [36,37]. The inland is a mountainous area, defined by alternating lithologies between sandstone (C1 formation), marls and

locally limestone (C2a, C3 formation), and limestone (C2b formation). The general groundwater direction is east to west, with quality deterioration westward due to saltwater intrusion [36].

The boundary between the two basins is defined by the Hammana formation (C3), dominated by marl layers forming an impermeable base of the C4-C5 basin. While the C1-C3 basin is hydrogeologically defined by [32] UNDP-MOEW (2014) as an unproductive basin for groundwater (Basin 31-Aptian Albian), many seasonal and some perennial springs are present in the area. The local communities use these springs for irrigation, water supply in the dwelling and, in some cases, drinking. The Naameh landfill is located at the western edge of the C4 basin, at a lower hydrographical and hydrogeological position than the upper municipalities (Figure 1). Therefore, effects on the surface and groundwater from the landfill are not expected to occur in the C1-C3 Aptian Albian Basin. However, several open dumps are present in the C1-C3 used by the local municipalities in emergency cases. In fact, the pop up of municipal dumps in Mount Lebanon after 2015 is one of the consequences of the closure of the Naameh landfill in 2015 [10]. It is worth mentioning that while the Naameh landfill is an engineered landfill provided with a bottom liner and leachate draining and methane collection systems, local dumps do not have any environmental control in place.

The area has been studied by Khadra and Stuyfzand (2014) [33]. Their study defined the main hydrosomes and the hydro-geochemical facies, based on an extensive sampling for major cations and anions, isotopes, and heavy metals, and followed Stuyfzand's, 1993 [38] methodologies. In this work, the authors identified one area, north-west of the landfill, to be likely affected by the landfill leachate percolation in the aquifer [33] (Khadra and Stuyfzand, 2014). The presence and extent of the contamination indicated by these authors was based on the results from one well, due to its elevated concentrations of chlorides, boron, lead, and zinc. Moreover, the sample was plotted below the boron/chloride (B/Cl) reference line constructed for the salinized samples, which reflected higher chloride content compared to the corresponding boron.

2.2. Methods

Guided by the local municipalities, mapping of the main water sources and local solid waste dumping sites in the area was conducted between October and November 2017. A total of 18 springs, 28 public and private wells, and seven municipality dumpsites (four inactive and three active) were identified. The selection of the sampling points from all mapped wells was based on the following two considerations: (a) prioritize water sources of public concern (municipal wells and private wells used for water trucking and municipal springs used for drinking) and (b) prioritize wells located in the proximity of the Naameh landfill (for the sources located downstream). Accordingly, 12 wells and seven springs were selected for sampling. Samples were also collected from a seasonal stream in the proximity of a local dumpster to estimate the impacts of open dumping on surface water. A summary table of the locations discussed with the community can be found in supplementary information Table S2.

Two wells (W4 and W5) were sampled twice during the campaign, due to their importance for the local community. In fact, W4 is used by many residents of Village 5 as a source of drinking water and W5 is the main water source for the largest municipalities in the area, serving approximately 15,000 citizens (information from the municipality).

A detailed list of the analytical methods deployed for the water quality measurements is presented in the supplementary information (Table S1). Analyses were performed at the Environmental Core Lab at AUB and at Eurofin Analytico, Netherlands. Samples were collected at the source outlet using sterile vials and bottles and strictly followed the laboratory standard procedures. Shipped samples were kept refrigerated and delivered within two days to the Netherlands. All bacteriological analyses were conducted at AUB laboratories.

The sample from the local river (R1) in the proximity of an active municipal dumpsite was tested for physical parameters (pH, conductivity, and temperature), general chemical parameters (chlorides, nitrate, phosphates, and sulfates), oil and grease, metals, microbiology (total and fecal coliforms

and *Escherichia coli* (*E. coli*) and volatile organic compounds (VOCs). Springs in the C1-C3 basin were tested for physical parameters, general chemical parameters, heavy metals, microbiology, total petroleum hydrocarbons (TPH), and adsorbable organic halides (AOX). AOX is a sum parameter used as a proxy for persistent organic pollutants in water. Possible sources of AOX include landfilling with high organic waste fraction [39], open dumping and burning practices [40], or other sources including sludge and sewage infiltration [41,42]. It is worth mentioning that the use of AOX as an indicator of pollution is disputed by a portion of the scientific community [43]. The same tests performed for the C1-C3 springs were also conducted for the three wells located in the C1-C3 basin. The wells located in the C4-C5 basin—the unit that hosts the Naameh landfill—were tested for a more extensive range of organic compounds. These include mono aromatic hydrocarbons, benzene, toluene, ethylbenzene and xylene (BTEX), phenols, poly aromatic hydrocarbons (PAH), halogenated hydrocarbons (volatile halogenated hydrocarbons, chlorinated benzenes, chlorinated phenols, polychlorinated bisphenols (PCBs)), chloronitrobenzenes, pesticides (chlorine, nitrogen and phosphorus based), and total petroleum hydrocarbon (TPH).

Each result was compared to the relevant national and international guidelines for drinking water Ministry of Health (MOH), 1999 [44] and World Health Organization (WHO), 2008 [45]. Sampling was conducted in November 2017, February and April 2018, corresponding generally to high flow period in the study area.

To evaluate the anthropogenic impacts on water quality, a comparison was made with the natural background concentration established in the same area by Khadra and Stuyfzand (2014) [33]. For each hydrogeological area and type of water source (springs or wells), a 95% confidence level interval was calculated from the results. For bacteriological parameters that were not included in the study of [33] Khadra and Stuyfzand (2014), a natural background value was assigned equivalent to the limits of detection (1 colony-forming unit (CFU) for total and fecal coliforms).

To establish the water quality across the area of study, two water quality indices (WQI) referred to as WQI-1 and WQI-2 respectively, adapted from Muzenda et al. (2019) [46] and Soltan et al. (2013) [47], were determined from the experimental water quality data (Appendix A, Equations (1)–(6)). Both indices compare the water quality results to a reference water quality standard. The Lebanese Ministry of Health (MOH) Standard 1999 [44] was adopted in this study. The parameters included in WQI-1 are conductivity, pH, chlorides, nitrate, phosphorous, barium, chromium, selenium, zinc, and fecal and total coliforms. The parameters for WQI-2 were almost identical to WQI-1 but excluded the total and fecal coliforms results. WQI-1 has a relatively high weight for the bacteriological parameters (fecal and total coliforms) and is therefore more appropriate as an indicator for possible sewage and wastewater effect on water quality. A value for WQI-1 that exceeds 100 indicates that at least one of the parameters is above the referenced standard. On the other hand, WQI-2 averages the ratio between the results and the reference standard adopted. Accordingly, a value of 100 for WQ-2 indicates that several parameters exceeded the WQ referenced standard. WQI-2 is used therefore to indicate differences in physicochemical water quality across the area (the two methods are described in detail in Appendix A).

The results for water quality were interpolated using the Kriging operator of the Spatial Analyst toolset of ArcGIS Desktop 10, ESRI [48] (ordinary Kriging method, spherical variogram with variable searching radius, output cell size 100 m). All statistical analysis was performed using R software. Plots and graphs were created using ggplot open source packages for R [49].

3. Results

The results of the water quality parameters are presented in Tables 1–3 and will be elaborated below according to the different sampling areas and sources.

3.1. Stream

The sample R1 was collected in the immediate proximity of a local municipality dumpster. R1 showed six flagged values against Ministry of Health (MOH) [44] maximal concentration limits

(MCL) for wastewater disposal in surface water; nominally, conductivity (1576 uS/cm), nitrate, sulfides, manganese, oil and grease in water (Table 1). The microbiology analysis showed concentrations above the limits, with fecal coliforms and *E. coli* above 10,000 CFU.

Table 1. Results from water quality testing of a stream (R1) located in the proximity of a dumpster.

Analysis Group	Analysis	Unit	Sample R1	MOH-MCL-Surface Water
Field data	Conductivity	uS/cm	1576	-
	pH		7.4	6–9
	Temperature	°C	20	
General chemical	Ammonia	mg/L	5.8	10
	Fluoride	mg/L	0.45	25
	Nitrate (NO ₃)	mg/L	117.3	90
	Orthophosphates	mg/L	1.51	5
	Phosphorous	mg/L	0.08	16
	Sulfides	mg/L	0.185	<0.005
	Sulfates	mg/L	48.5	1000
Other chemical	Total organic carbon (TOC)	mg/L	100	75
	Total suspended solids	mg/L	67.5	200
Metals	Aluminum	ug/L	3	10,000
	Barium	ug/L	8	2000
	Cobalt	ug/L	30	500
Microbiology	Manganese	ug/L	1680	1000
	<i>E. coli</i>		>10,000	2000
	Fecal coliforms	CFU/250 mL	>10,000	2000
	Other bacteria	P/A	Present	-
	Salmonellae	P/A	Present	Absent

Cyanides, As, Cd, Cr, Cr⁶⁺, Cu, Fe, Hg, Pb, Sb, Sn, Zn, and VOCs are below detection limit.

3.2. C1-C3 Groundwater Basin

The results for the C1-C3 groundwater basin (Table 2) revealed values of conductivity and pH within the standards for both springs and wells. Nitrate and phosphates spiked in three samples (two springs and one well). Barium is present at low concentration (5.4–18 ug/L) in all the samples, selenium and chromium are present in one well at trace concentration, and zinc is above 100 ug/L in two wells. Low concentrations of TPH were detected in one spring and low levels of oil and grease were reported for one of the wells. AOX was not detected in any of the samples. All the samples contained total and fecal coliforms and six of the 10 samples were positive for *E. coli*. Full results are presented as supplementary information (Table S3: results from water quality testing of springs in C1-C3 basin and Table S4: results from water quality testing of wells in C1-C3 basin).

Table 2. Results from water quality testing of wells and springs in C1-C3 basin. Natural background from [33] Khadra and Stuyfzand (2014), values for the mountain hydrosome ¹.

Analysis Group	Analysis	Unit	N of Tests—Wells	Average Wells +/- 95% CI	N of Tests—Springs	Average Springs +/- 95% CI	Natural Background	EPA/WHO MCL	MOPH MCL
Field data	Conductivity	uS/cm	3	604.7 +/- 122.7	7	619.3 +/- 50.5	519	-	1500.00
	pH	-	3	7.4 +/- 0.3	7	7.6 +/- 0.2	7.2	6–8	6–8
	Temperature	°C	2	17.4 +/- 3.8	6	16.7 +/- 1.9	-	-	-
General chemical	Chlorides	mg/L	3	17.4 +/- 10.6	7	30.6 +/- 7.4	24.1	250	200
	Nitrate (NO3)	mg/L	3	1.6 +/- 3	7	16.9 +/- 12.9	1.2	45	45
	Phosphorous	mg/L	3	0 +/- 0	7	0 +/- 0	0.1	-	-
	Orthophosphates	mg/L	1	NA	6	0 +/- 0.1	-	0.03	1
	Diphosphorus Pentoxide	mg/L	2	0 +/- 0	6	0 +/- 0.1	-	-	-
	Sulfates	mg/L	1	0	-	-	12	250	250
	Barium	ug/L	3	13.3 +/- 8.2	7	6.4 +/- 2.4	8.4	2000	2000
Metals	Chromium	ug/L	3	0.2 +/- 0.3	7	0 +/- 0.1	0.6	100	-
	Selenium	ug/L	3	0.2 +/- 0.3	7	0 +/- 0.1	<0.05	50	10
	Zinc	ug/L	3	103.3 +/- 113.3	7	0 +/- 0.1	4.1	5000	5000
Petroleum hydrocarbons	TPH (Total)	mg/L	1	NA	6	0.7 +/- 1.4	-	-	-
	<i>E. coli</i>	P/A	3	Present (1/3)	7	Present (5/7)	-	A	A
Microbiology	Fecal coliforms	CFU/250 mL	3	5.7 +/- 11.1	7	27.7 +/- 25.5	-	<1 cfu	<1 cfu
	Total coliforms	CFU/100 mL	3	50.7 +/- 48.6	7	67 +/- 31	-	<1 CFU	<1 CFU
	Other	P/A	3	Present (3/3)	7	Present (7/7)	-	-	-

¹ Ammonia, fluoride, nitrate equivalent, nitrite (NO₂), nitrite as No₂-N, P₂₀₅, PO₄, cyanides, DOC, TOC, total suspended solids, all other metals (Al, As, Be, Cd, Co, Cr₆₊, Cu, Fe, Hg, Mo, Ni, Pb, Sb, Sn, V), AOX, biphenyl, phenols, total PCBs, and VOCs are below detection limits.

3.3. C4-C5 Groundwater Basin

Three wells in C4-C5 basin exhibited high values of conductivity, chlorides, and sulfates (Table 3). One well showed moderate nitrite levels (0.095 mg/L). For the metals, trace concentration levels of arsenic, chromium, copper, mercury, and molybdenum were detected in different sources in the aquifer. As for the C1-C3 aquifer, barium was present as background in all samples, while zinc spiked in three wells (W4, W8, and W9). For the organic compounds, low levels of TPHs were detected in four wells, in association with phenol and trichloromethane (chloroform). Biphenyl was detected at very low concentration in two wells. Phenols were also detected at higher concentration (400 ug/L) in one sample (well W5A) but were not detected in the same well when sampled again (W5B). Two of the wells showed high levels of fecal and total coliforms and positive *E. coli*, while low levels of bacteria were detected in two wells (Fc 1–2 CFU/250 mL; TC 3–4 CFU/100 mL). Full results are presented as supplementary information (Table S5: results from water quality testing of wells in C4-C5 basin).

Table 3. Results from water quality testing of wells in C4-C5 basin. Natural background from [33] Khadra and Stuyfzand (2014), values for the Coastal Hydrosome ¹.

Subgroup	Analysis	Unit	N of Tests—Wells	Average Wells +/- 95% CI	Natural Background	EPA/WHO MCL	MOPH MCL
Field data	Conductivity	uS/cm	11	1189 +/- 616	757	-	1500.00
	pH		11	7.4 +/- 0.1	7.1	6-8	6-8
	Temperature	°C	9	20.3 +/- 2.4	-	-	-
General chemical	Bromide	mg/L	6	0.4 +/- 0.7	0.3	-	-
	Chlorides	mg/L	11	115.5 +/- 127.9	82.1	250	200
	Nitrate (NO3)	mg/L	11	8.3 +/- 3.6	5.5	45	45
	Nitrite (NO2)	mg/L	4	0 +/- 0	-	1	0.05
	Sulfates	mg/L	8	51.4 +/- 34	71.9	250	250
	Arsenic	ug/L	11	0.1 +/- 0.2	0	50	50
	Boron	ug/L	6	16.7 +/- 32.7	42	-	-
	Barium	ug/L	11	24.2 +/- 14.4	51.1	200	200
	Chromium	ug/L	11	0.4 +/- 0.5	0.6	100	100
	Copper	ug/L	10	0.8 +/- 1.6	0.9	1300	1000
Metals	Mercury	ug/L	11	0 +/- 0	0	2	5
	Molybdenum	ug/L	9	0.4 +/- 0.8	0.2	-	-
	Nickel	ug/L	11	0.5 +/- 1.1	0.2	20	10
	Zinc	ug/L	11	228.7 +/- 214.9	3.3	5000	5000
	Miscellaneous organic compounds	Biphenyl	ug/L	7	0 +/- 0	-	-
Petroleum hydrocarbons	TPH (total)	ug/L	7	16.3 +/- 12.6	-	-	-
	Phenols	Phenol	ug/L	7	64.3 +/- 116.2	-	-
Volatile halogenated hydrocarbons	Trichloromethane	ug/L	7	0.2 +/- 0.2	-	100	100
	<i>E. coli</i>	P/A	5	Present (2/5)	-	A	A
Microbiology	Fecal coliforms	CFU/250 mL	5	7.2 +/- 9.5	-	<1 CFU	<1 CFU
	Total coliforms	CFU/100 mL	5	41.4 +/- 46.94	-	<1 CFU	<1 CFU
	Other (present or absent)	P/A	5	Present (5/5)	-	-	-

¹ Ammonia, ortho-phosphates, P, P205, PO4, cyanides, total suspended solids, all other metals (Al, Be, Cd, Co, Cr6+, Mn, Pb, Sb, Se, Sn, V), cresols, chlorophenols, PAH, VOCs, chlorinated VOCs, and pesticides are below detection limit.

3.4. WQI

Water quality indices WQI-1 and WQI-2 are presented in Table 4. The springs of the C1-C3 basin have very poor water quality as WQI-1 is higher than 100 for all locations and has an average of 1121. Similarly, the wells in the C1-C3 and C4-C5 basin have poor water quality, with an average WQI-1 of 444 and 377, respectively. More generally, WQI-1 is significantly lower for wells than for springs. On the other hand, values of WQI-2, (which do not include total and fecal coliform values),

were below 100 for all locations. Full results for the water quality indices computation are presented as supplementary information (Table S6: W_i and K value for WQI-1 and Table S7: Result from WQI-1 Analysis; Table S8: Result from WQI-2 Analysis).

Table 4. Summary of the water quality index (WQI) analysis adopted from [46] Muzenda et al. (2019) (WQI-1) and [47] Soltan et al. (1999) (WQI2).

Hydro-Geological Setting	Water Source	WQI-1 ¹	WQI-2 ²	
Springs C1-C3	S1	177	16	
	S2	1598	23	
	S3	1455	18	
	S4	617	30	
	S5	212	24	
	S6	1382	20	
	S7	2404	18	
	Average Springs C1-C3	1121	21	
Wells C1-C3	W1	1066	19	
	W2	129	16	
	W3	136	15	
	Average Wells C1-C3	444	17	
Wells C4-C5	W4	874	24	
	W5	102	17	
	W6	114	20	
	W7	-	17	
	W8	-	21	
	W9	1145	81	
	W10	-	22	
	W11	1	36	
	W12	28	44	
		Average Wells C4-C5	377	31

¹ WQI-1 is based on pH, chlorides, nitrate, phosphorous, barium, chromium, selenium, zinc, and total and fecal coliforms. ² WQI-2 is based on pH, chlorides, nitrate, phosphorous, barium, chromium, selenium, and zinc.

4. Discussion

4.1. Comparison with Natural Background

In order to evaluate the extent of the anthropogenic effects in the tested wells and springs, the results recorded in this work were compared with the natural background level (NBL) established by Khadra and Stuyfzand 2014 [33]. Figure 2 presents the water parameters results normalized by the natural background levels for mountain and coastal hydrosomes. The results for springs and wells in the C1-C3 basin were normalized by the natural background for the mountain hydrosome while results for C4-C5 wells were normalized by the coastal hydrosome background levels. Total and fecal coliforms were not included in the study of Khadra and Stuyfzand 2014 [33]; an arbitrary background value of 1 CFU (the limit of quantification) was assigned to those two parameters.

As shown in Figure 2, most results fall close to the background values (corresponding to 1 in the graph) except for zinc (in wells C1-C3 and wells C4-C5), fecal coliforms (in springs C1-C3), and total coliforms in all sampling areas. High values of nitrate in the C1-C3 springs were also observed. Nevertheless, the data does not show large deviations from the natural background values.

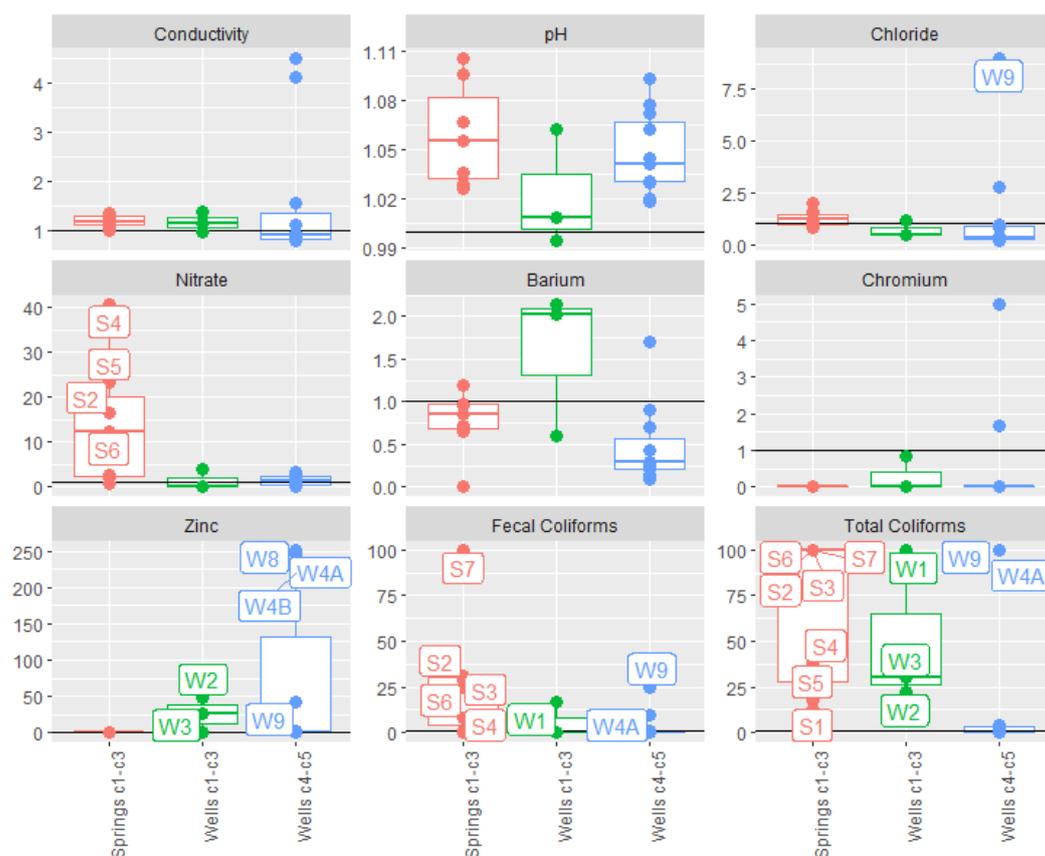


Figure 2. Results for the different hydrogeologic settings normalized by the natural background level (NBL) established by Khadra and Stuyfzand (2014) [33]. The y-axis therefore represents the number of folds that the results are higher or lower than the NBL.

4.2. Possible Sources Of Contamination

Different studies in Lebanon suggest metals (Zn, Cd, Cr, and Pb) as possible marker indicators for solid waste contaminants in surface water bodies [27,50,51]. Additionally, elevated values of conductivity, chlorides, and sulfates are commonly used as indicators for leachate contamination in groundwater [52,53], while AOX are used by some authors as a proxy to evaluate the presence of persistent organic compounds. The results in the upper area (C1-C3 basin) did not flag any of these markers except the stream sample R1, sampled in the proximity of a local dumpsite, which showed high levels of manganese, oil and grease, and nitrate. At the same time, the presence of very high values of *E. coli* in the same sample (>10,000 CFU/250 mL) suggests a strong contribution of sewage. This interpretation was also confirmed by the local municipalities in the area at the time of the assessment. C1-C3 springs and wells results are also consistent with a hypothesis of prevailing sewage contamination rather than solid waste, as indicated by the presence of nitrate, phosphorous, and bacteriological indicators (e.g., [54]). It can be argued that fecal coliforms in water may not necessarily indicate the presence of feces and hence sewage infiltration [55]. Nevertheless, the fact that 7/8 samples that were positive for fecal coliforms were also positive for *E. coli* is an indication that sewage infiltration from septic tank or sewage lines is likely to be the major source of bacteriological contamination in the area. Another possible contribution to the contamination of the springs could be agricultural runoff (e.g., [56]), even though most of the springs sampled were located in the proximity of urban/peri urban areas (Figure 1). Agricultural runoff can be a second contribution of bacteriological contamination particularly in the sources located at lower hydrographic level from agricultural areas (W11, S7). For the coastal area (C4-C5 basin), relatively high values of conductivity, sulfates, and chlorides were recorded along with trace concentrations of organic compounds (phenols, TPHs, and trichloromethane). Zinc,

at values higher than 100 ug/L, was measured in three wells (W4, W8, and W9). The high values of chlorides and sulfates may either be the result of saltwater intrusion, as indicated by different studies in the area [36,37], or leachate percolation in the groundwater [52,53].

The result for the water sources in close proximity of the landfill were plotted in a ternary diagram for sulfates, nitrate, and chlorides (Figure 3a). Together with the samples collected in this research, the values of some relevant hydrogeochemical facies identified by Khadra and Stuyfzand [33] are displayed in this same diagram. The facies are (a) the natural background for mountain and coastal hydrosomes, (b) the salinized facies of the aquifer, (c) the sample suspected to be affected by landfill leachate. Nitrate levels range from 0% to 10% for all samples except W5B (23%). This suggests that agricultural runoff was probably not affecting the aquifer at the time of the assessment. A group of sources clusters around the natural background values. These sources are in the upper region of the aquifer with an exception of W12 which is located in the coastal plain. Four wells (W7, W9, W10, and W11) have a higher chloride sulfates ratio than the rest of the wells, ranging from 60% to 90% of chlorides. Furthermore, the well W9 falls exactly in the region of the hydrosomes affected by seawater intrusion [33]. Noteworthy, none of the samples is close to the landfill leachate region, characterized by a higher fraction of sulfates. However, it is necessary to have a larger sample of data and conduct further testing (analysis including the major cations and anions that were not part of this assessment) to come to a conclusive answer.

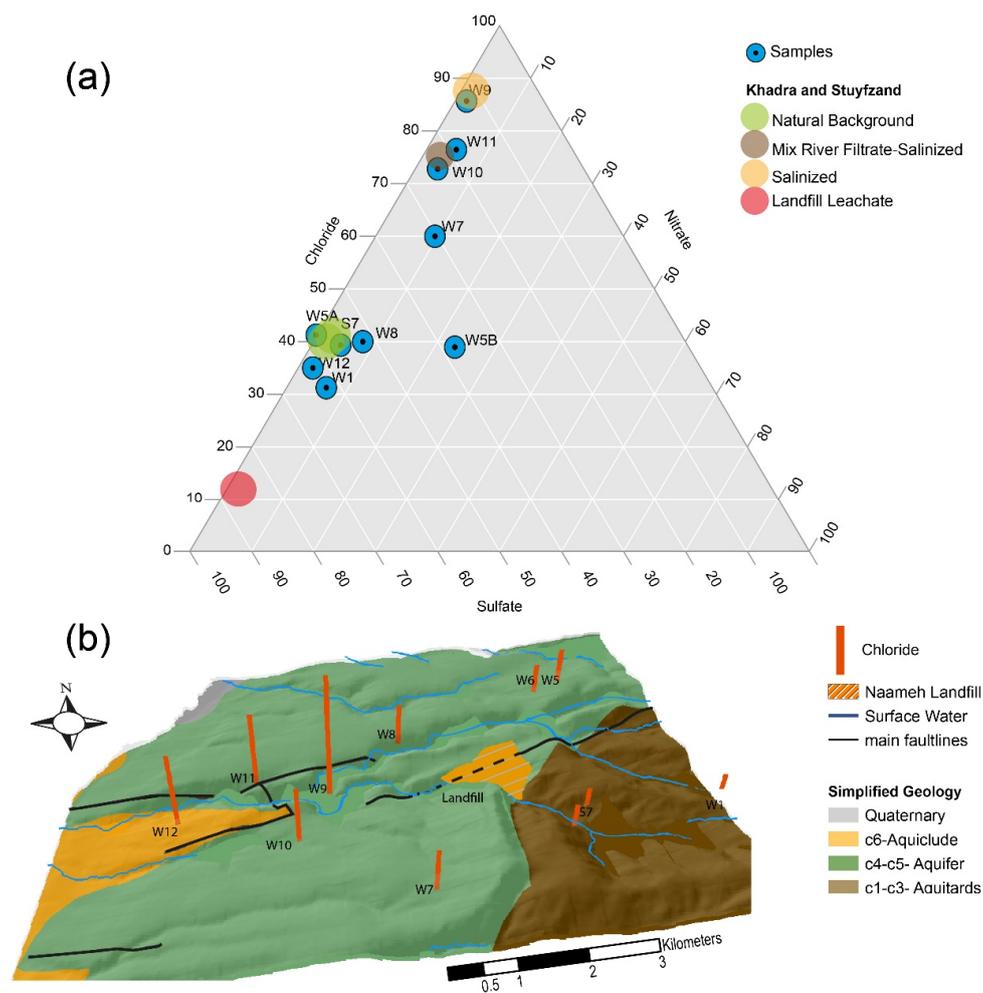


Figure 3. (a) Ternary plot for chlorides, sulfates, and nitrate for the sources surrounding the Naameh landfill, and comparison with some of the hydrogeochemical facies suggested by Khadra and Stuyfzand [33]. (b) Geology and sampling location.

To further evaluate the possibility of a non-marine anomaly responsible for the chloride concentration found in the wells, a Cl/Br ratio was used (as illustrated by [57] Kelly et al. (2010)) for W9 and W11. Cl/Br ratio was employed in different studies to characterize the hydrogeochemical facies of groundwater bodies [57–59]. The ratios for the chloride concentration versus chloride boron ratio were plotted for W9 and W11 in Figure 4.

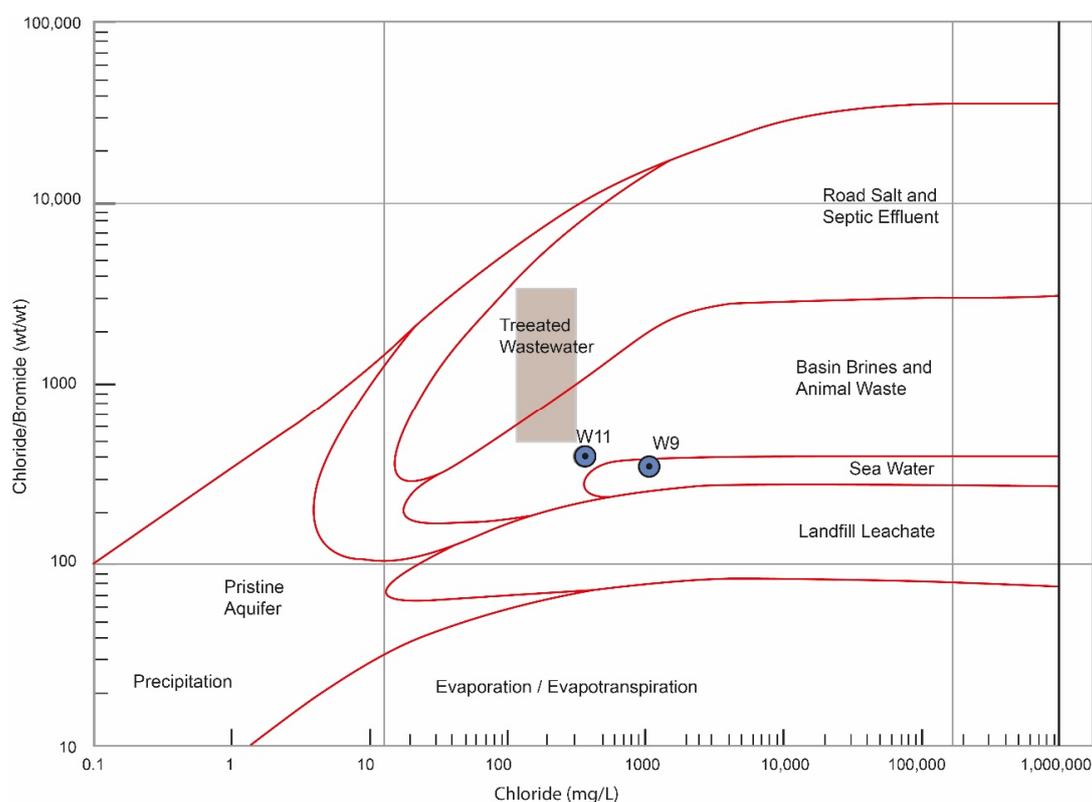


Figure 4. Diagram adopted from Kelly et al. (2010) [57], identifying sources for chloride/bromide ratio fields vs. chloride concentration. Samples W11 and W9 fall between seawater and the basin brines.

As can be seen in Figure 4, the W9 sample falls in the seawater classification, while the W11 sample falls in the basin brines and animal waste classification but is still very close to the seawater classification.

Figure 5a presents the results for the trace elements concentration (metals and organic compounds) in the C4-C5 aquifer. Zinc spikes in three wells downstream of the landfill. However, high values of zinc were also recorded in other wells upstream (W4A and W3). The zinc anomaly could be the result of corrosion of the casing of the sampled wells, some of which were older than 15 years (information from the Municipalities). Barium occurs at background levels. As for molybdenum, it is present in only one of the wells. Wells W8, W9, W10, and W11, all located downstream of the landfill, present trace concentrations of TPHs, phenols, and trichloromethane. Furthermore, TPH was found in association with phenol, suggesting a possible common origin. Studies suggest that trace concentrations of phenol can either be of natural origin or result from leachate infiltration in groundwater [52,60], as well as sewage infiltration and agricultural and industrial runoff [61,62]. Phenol was also detected in high concentrations in one sample upstream of the site (W4A 400 ug/L in November 2017), although the results were not confirmed in the second sample in the same well (W4B). Trichloromethane (chloroform) was detected in W9 and W10 at trace concentrations (0.39 and 0.66 ug/L respectively). Trichloromethane as well could be the result from the infiltration of landfill leachate as well as other sources, including industrial and agriculture runoff and sewage infiltration. In order to better understand possible interrelations between the detected trace elements and sources of contaminants in Figure 5b, the sources in the lower C4-C5 basin along with some of the land use-land cover for the area from CNRS, 2011 [63]

are presented. The first observation is that W7, located in a prevalently woodland area, presents only barium. This observation rules out the natural sources for the origin of the phenol. On the other hand, the wells presenting phenol and TPH are in proximity with roads and seasonal streams. The combined presence of these trace elements might result from the mix between the seasonal stream, contaminated with sewage, and the local aquifer. This is in line with Figure 3, that shows that wells W10 and W11 have levels for chloride/sulfate ratios between the natural background and the sea water intrusion region of the plot.

Overall, considering the very low values detected and the limited size of the samples in space and time, the results do not clearly confirm the hypothesis of a contact between the landfill leachate of the Naameh landfill and the C4-C5 aquifer. At the same time, indication of wastewater infiltration in the lower area is suggested by the presence of *E. coli* in three out of five samples tested for bacteriology (Table S4: Results from water quality testing of wells in C4-C5 basin). It is highly likely that the contamination occurred through the subsurface infiltration from the seasonal streams, though contamination from other sources (leaks from the sewage lines along the road network and septic tanks) is not to be excluded.

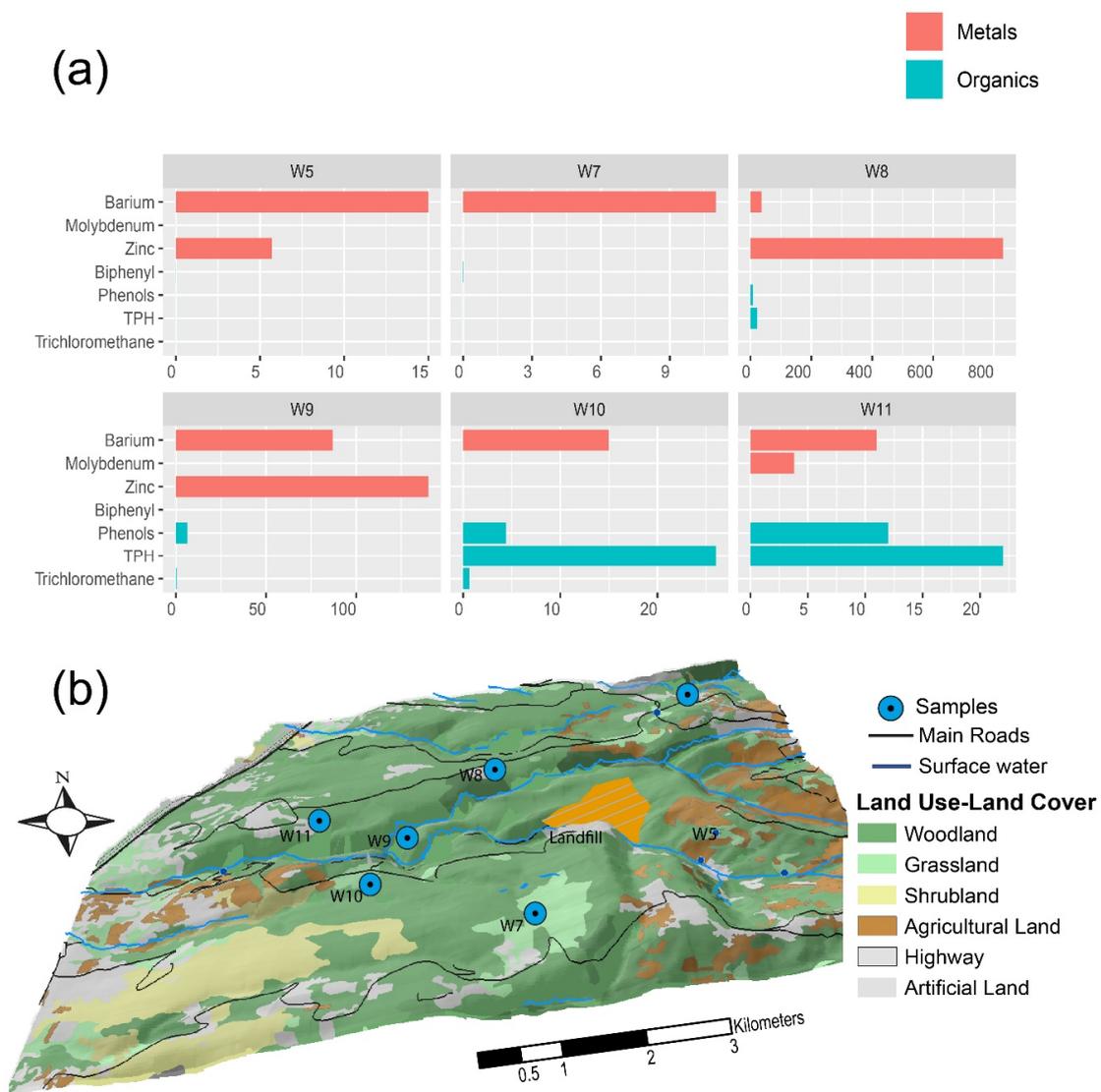


Figure 5. (a) Trace elements concentration in the wells of the C4-C5 aquifer around the Naameh landfill. Units are in ug/L. (b) Land use-land cover map of the area downstream of the Naameh landfill and sampling locations [63].

4.3. Water Quality Index and Community Public Health

Concerning the water quality index analysis, WQI-1 reflects what was previously discussed in Sections 4.1 and 4.2 as it indicates an extensive bacteriological contamination in the area, particularly in the upper C1-C3 basin, upstream of the Naameh landfill (Figure 6a). Nevertheless, WQI-2 (Figure 6b), which excludes the contribution of the bacteriological results, shows an overall good water quality across the region ($WQI-2 < 100$), with higher values in the coastal area (average 31, max 81). The higher WQI-2 values in the coastal region are the result of high values of chlorides, conductivity, and zinc observed in the C4-C5 wells. It can therefore be concluded that the physicochemical water quality in both the mountain range and the coastal area were, at the time of the observations, of an average good quality.

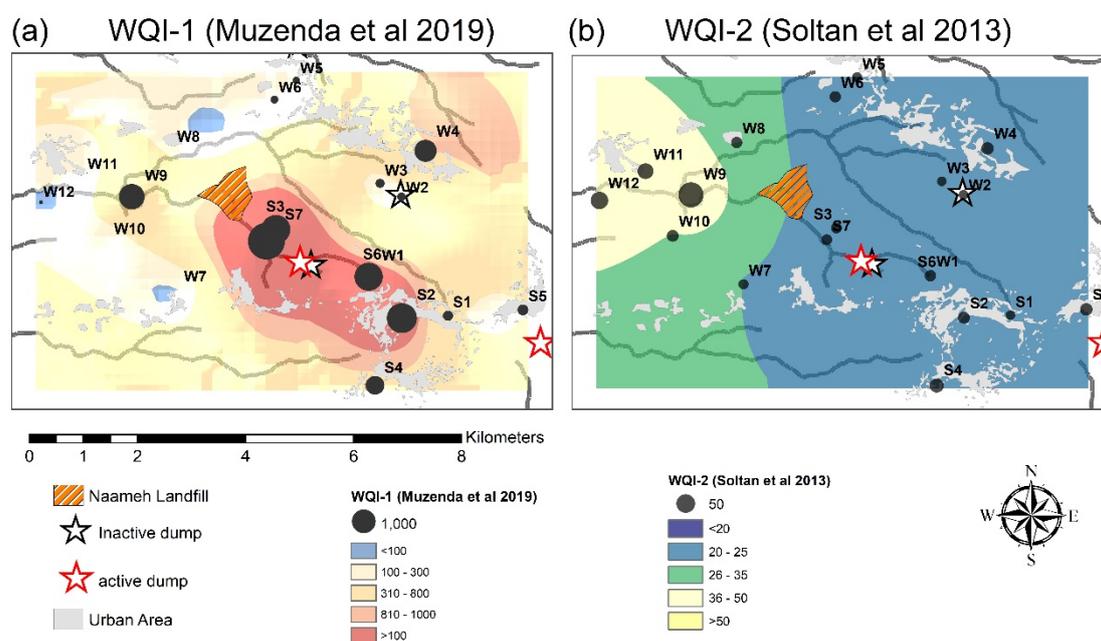


Figure 6. Water quality indices (WQI-1 and WQI-2) across the area of study. The predicted water quality was determined using Kriging on the WQI respective values.

In parallel to this work, an observational epidemiological study was carried out in the area. Surveys were collected from 2720 citizens from the same six municipalities involved between November 2017 and January 2018. The participants were asked to self-report the frequency of occurrence of symptoms and diseases associated with solid waste malpractices (Table S8: Health Assessment Summary). The methodology to collect the health data was based on an air pollution model, described in [24]. The model simulated the exposure to NO_x concentrations (used as representative of landfill gas emission) and the results were presented as percentages from the maximum modeled concentration at the Naameh landfill. In the exploratory analysis, a significant correlation between some of the symptoms and the landfill TAPM exposures were found (see supplementary information Table S8: Health assessment summary). The results reported in this work could provide a second possible explanation for part of the health ailments as some of the reported symptoms and diseases may have been caused by contact with water contaminated with wastewater (particularly skin rashes). As shown in Figure 7, Villages 1 and 2, served by different water sources with recorded bacteriological contamination (S1, S2, S6, W1), are also the ones with the highest exposure from the air pollution model. In other words, air pollutants from the landfill and water pollutants from wastewater may have contributed to the increase of skin rashes in the areas of Villages 1 and 2.

While this result is important, it should be stressed that tap water from the households was not tested and, therefore, the link with the health symptoms can only be inferred and should be taken as

preliminary. Further research to understand and better define the correlation between waste disposal, water, and health shall be conducted to better unravel the causalities.

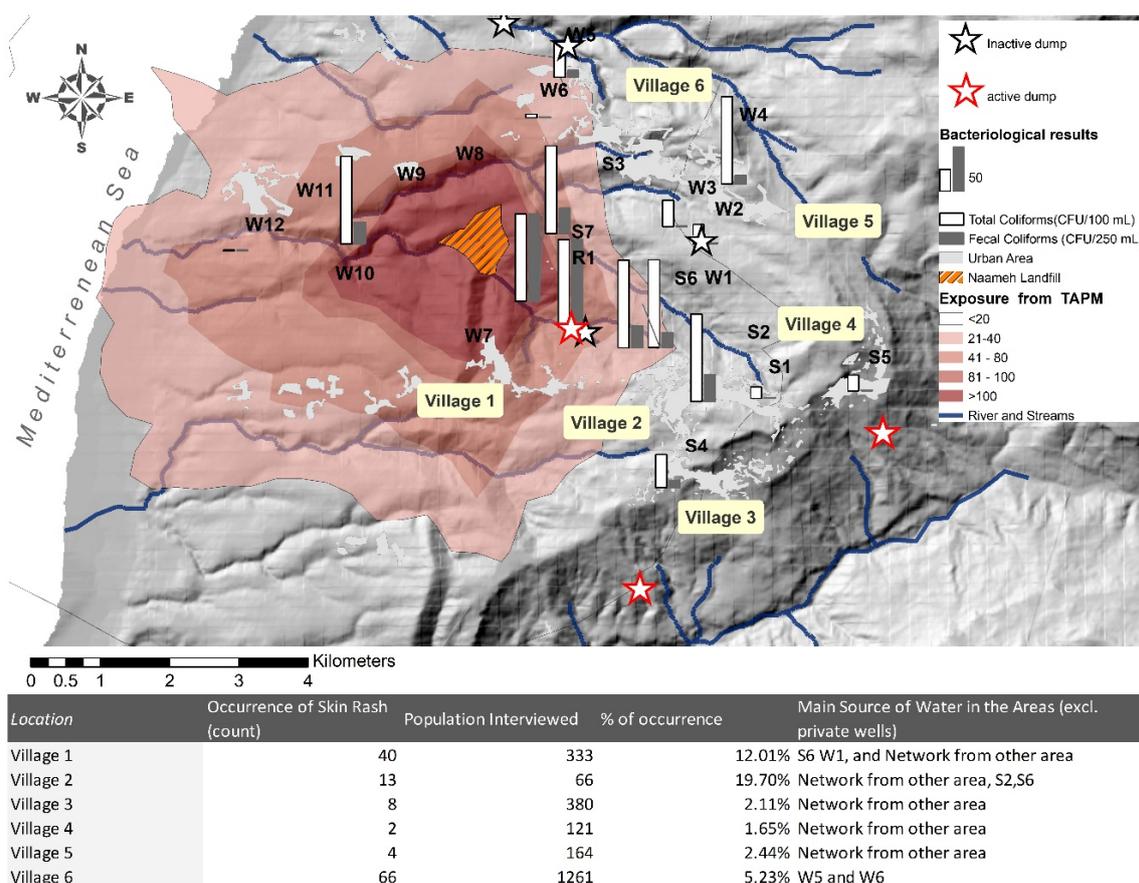


Figure 7. Bacteriological results, air exposure from the air pollution model (TAPM) [64], showing high bacteriological contamination in the proximity of Village 1 and Village 2. S1, W12, W2, W3, W5, fecal coliforms < 1 CFU, W12 total coliforms < 1 CFU. TAPM exposures are presented as the percentages of modeled air pollutants concentration from the maximum modeled concentration of at the landfill source.

The findings on water quality from the C1-C3 and C4 basins provide an answer to the research questions raised by the local communities. While there is no major evidence of solid waste related contamination in the area, the presence of bacteriological contamination (total and fecal coliform) associated with *E. coli* indicates an intensive wastewater contamination.

Several objections may be raised on the process that led to the results reported in this work, amongst which are the following: (a) the sampling did not consider seasonal variation of water quality; degradation in the water quality is expected during the low flow season and (b) source appropriation for landfill leachate was not possible. While this study does not claim to carry out a complete model on contamination and transport in the groundwater resources in the study area, it provides an important snapshot of data for the status of water quality in one of the most important and controversial areas for water supply and solid waste disposal in Lebanon.

5. Conclusions

This study answers two questions raised by the six communities of the study area: (1) do the groundwater resources present evidence of solid waste contamination related to the Naameh landfill and to the local dumpsite in the area? (2) What are the other sources of contamination affecting the groundwater resources? From a broader perspective, the study assessed water quality degradation

across different aquifers and sources, from a relatively small sample (18 sources, one sampling campaign). The analysis conducted included various physical, chemical, and microbiological parameters and sources of different levels of importance for the community (drinking, public water supply, and water trucking). Seven springs and three wells in the C1-C3 basin and 11 wells in the C4-C5 basin were tested for general chemical physical parameters (conductivity, chlorides, sulfates, nitrate, etc.), microbiology (total and fecal coliforms), metals (nickel, lead, arsenic, zinc, copper, etc.). The wells located in the aquifer underlying the landfill were tested for a large spectrum of organic compounds. For the C1-C3 basin, the result indicated a significant bacteriological contamination, along with relatively high values of nitrate and phosphates in local springs. The results for the C4-C5 aquifer showed higher levels of conductivity, chlorides, and zinc along with the occurrence of organic micropollutants in trace concentrations. The water quality index analysis revealed that, considering the bacteriological indicators, both aquifers have extremely deteriorated water quality ($WQI > 100$). When looking at the different sources, the springs recorded the poorest water quality ($WQI > 1000$ for several springs). On the other hand, WQI-2 that do not consider microbiology showed a generally good water quality, slightly deteriorating in the coastal wells downstream of the Naameh landfill.

In conclusion, we can state that at the time of the analysis, the hypothesis of a solid waste diffuse contamination to the groundwater sources in the area is not fully supported by the data. However, the presence of organic compounds at low concentration and the presence of zinc in some of the wells may be an indicator of contamination by leachates. On the other hand, a major bacteriological contamination, resulting from release of untreated wastewater in septic tank and surface water bodies, is a major source of pollution of the upper C1-C3 basin, where most of the municipalities are located. While the limited number of observations both in space distribution and time (limited to a high flow period between November and March) represent a limit to this research, the data presented in this work has provided an improved insight of the current status of the water quality in the study area. The findings can be used to formulate two recommendations. The first one consists of improvements in the wastewater management in the upper municipality, essential to decrease the release of untreated wastewater (a recommendation which has been partly taken into consideration from the local authorities, as the municipality in the area will be connected to the main sewer network linked to Al Ghadir treatment plant south of Beirut). The second one includes the continuous monitoring of the Naameh landfill and, specifically, the establishment of a monitoring network of selected wells to be regularly tested. Finally, the following set of recommendations could be provided for research groups conducting similar assessments. First, it is extremely important to engage the communities involved at all the stages of the project [65]. Second, results from the water quality indices depend on the parameters chosen. In this case, a major role has been played by bacteriological indicators; hence the importance of selecting the most appropriate water quality index according to the target contaminants in order not to disseminate confusing results. Third, new studies could build on the results provided in this manuscript and its supplementary material to enhance the resolution (spatial or temporal) of the observations.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2073-4441/12/5/1358/s1>, Table S1: List of standards and methods for water testing, Table S2: Results from water quality testing of springs in C1-C3 basin, Table S3: Results from water quality testing of wells in C1-C3 basin, Table S4: Results from water quality testing of wells in C4-C5 basin, Table S5: Wi and K values for WQI-1, Table S6: Result from WQI-1 Analysis, Table S7: Result from WQI-2 Analysis, Table S8: Health assessment summary.

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Appendix A

WQI-1

Following [46] Muzenda et al. (2019), the WQI-1 is defined by the following equation:

$$WQI = \text{antilog}_{10} \left[\sum_i^n w_i \log_{10} q_i \right] \quad (A1)$$

where w_i is the weight of the i th parameter, q_i is the quality ranking of the i th parameter. Equations (2) and (3) define w_i , while Equation (A4) defines q_i :

$$w_i = \frac{k}{v_i} \quad (A2)$$

where v_i is the maximum allowed value for the i th parameter, in our case according to [44] Lebanese Ministry of Health standard for drinking water, while k is a constant defined as follows:

$$k = \frac{1}{\sum_i^n \frac{1}{v_i}} \quad (A3)$$

Finally, q_i is defined as per Equation (A4)

$$q_i = 100 \times \frac{va - vs}{vi - vs} \quad (A4)$$

where va is the water quality result, vs is the ideal water quality result (7 for pH and 0 for all other parameters)

WQI-2

Following [37] Soltan et al. (2013), the average water quality index i.e., WQI-2 is calculated by the following Equation (A5):

$$WQI - 2 = \frac{\sum_{i=1}^n q_i}{n} \quad (A5)$$

where q_i is the quality rating for the i th parameter, defined as below (Equation (A6)):

$$q_i = 100 \times \frac{V_i}{S_i} \quad (A6)$$

where v_i is the water quality result of the i th parameter and s_i is the water quality standard from Ministry of Health (1999) [44].

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