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Heavy Metal Estimation and Quality Assurance Parameters for Water Resources in the Northern Region of Pakistan

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Abstract: The current study investigates the water quality parameters of drinking water resources in District Neelam (DNLM), Azad Jammu & Kashmir (AJK), Northwestern Pakistan. The studied area has been recently reported with many waterborne diseases, which probed this analytical study. The samples were aseptically collected from springs, taps, and surface water bodies. The water quality parameters, such as physical, microbiological, anions, and heavy metals, were tested. Results showed that the electrical conductance (EC) and total dissolved solids (TDS), were 974.60 µS/cm and 912.10 mg/L, respectively, exacerbating the quality of drinking water in DNLM. For microbial water testing, we used 3M-Petrifilms as a detection source, which could separate coliform bacteria from *E. coli* by creating unique surface chromophores. Out of sixty collected samples, 76% had bacterial contamination. Nitrite, nitrate, and phosphate (9.8, 15.0, and 15.1 mg/L), were also surpassing the safe limits of the World Health Organization (WHO) standards for water quality measurement. The heavy metals, i.e., As, Cr, Cu, and Pb were also tested in current analysis. Pb and Cr (0.04 mg/L and 0.06 mg/L) exceeded from safe drinking water guidelines of the WHO and more than 50% of the collected samples had Pb as a major water pollutant in DNLM. Poor waste management, open sludge discharge, lack of municipality measures, and mineral leaching into the freshwaters of DNLM due to mining and metal extraction processes were the main sources of water pollution in the region. The inorganic pollutants were responsible for the sudden rise of different malignancies and other fatal diseases (vital organ failures and reproductive disorders) in the region, which has not been reported in the past. The current investigation yielded useful baseline data of the drinking water reserves of NW Pakistan that could help to develop techniques for the mitigation of water pollutants present in the region.

Keywords: waterborne diseases; mineral leaching; bacterial pollution; nitrites; nitrate; Pb

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1. Introduction

The rapid population growth and uncontrolled urbanization have led to serious environmental hazards, water contamination being one of them [1]. Climate changes such as, anthropogenic, geomorphological changes, erosion, and mining processes have significant effects on available water resources, i.e., springs, lakes and groundwater sources [2]. Excessive land usage and application of fertilizers have threatened species extinction and loss of biodiversity worldwide [3]. Therefore, water quality management has been regarded as a global issue and high concentrations of parameters such as physical, anionic, and heavy metals indicate the poor water quality conditions of the region. Heavy metals, i.e., Pb, Cr, Fe and Cd, are the major contributors to water pollution and chronic poisoning in aquatic organisms and humans [4]. Heavy metals, along with microbial agglomerations can further boost the impact of aqueous pollutants, affecting open-water systems [5]. Along with this the physical parameters also affect the health of living organisms using these waters [6]. Urbanization, inadequate sanitation, and unregulated waste disposals have affected the upper concentration values of water quality indicators which have degraded the quality of accessible water reserves [7]. Global warming is another potential issue that has widened the gap between the availability of clean drinking water and its demand [8]. Therefore, the current study has great importance and applicability, as there is no comprehensive information available on the water resources of District Neelam (DNLM), Azad Jammu and Kashmir (AJK), Pakistan.

Microbiological water testing is a global indicator of water quality determination [9]. Microbial species such as total coliforms and *E. coli* [10], *Enterococci* [11], and *C. perifirengens* are the reason for many waterborne diseases worldwide [12]. The existence of fecal coliforms in recreational waterways is another problem which is considered a major cause of health hazards in humans. Likewise, billions of people lack basic sanitation and clean water facilities around world, especially in poor and developing countries. As an example, more than three million infants die annually from diarrhea and other viral diseases in these countries caused by consuming polluted water [13]. Kapembo et al., reported the bacterial (fecal bacteria and *E. coli*) contamination in the Democratic Republic of the Congo (2016–2019), producing typhoid fever, amoebic dysentery, diarrhea, gastrointestinal problems, and cholera that affected more than 75% of the people [14]. According to the WHO (2018), waterborne illnesses kill 2 million people annually [15]. Hamad et al., showed the presence of Gram-negative bacteria (CFU/mL) as 20 CFU/mL followed by 485 and 283 CFU/mL (Alhilwa and Alwafi, Iraq), respectively [16]. Additionally, species such as *V. fluvialis, G. hollisae, V. hollisae,* and *V. mimicus* in water bodies extensively cause human gastrointestinal problems [17]. The presence of *Salmonella* a genus of Enterobacteriaceae in water causes diarrhea, nausea, and fever [18]. The drinking water also contains *Shigella*, another species of pathogenic bacteria, which causes fever, anorexia, tiredness, malaise, and stomach pains [19].

Higher anionic additions to the freshwater reservoirs degrade their quality. For example, the higher concentrations of anions (>MCL of anions) such as, F, Cl, Br, NO_2, NO_3, and SO_4 found in water samples from Kaohsiung, Taiwan, damaged the water quality of the region [20]. Djam et al., analyzed NO_3, F, Cl and SO_4 in bottled water and found the mean values as 0.13, 6.77, 25.1 and 20.9 ppb, respectively [21]. In Saudi Arabia water collected from bottles and local taps was tested for NO_3, NO_2 and SO_3 showed that in bottled water NO_3 was 5.42–12.14 mg/L, NO_2 was 1.08–4.37 mg/L and SO_3 was 6.84–32.45 mg/L, and tap water contained NO_3 at 1.27–16.11 mg/L, NO_2 at 0.43–10.77 mg/L and SO_3 at 7.14–36.10 mg/L [22]. Another study on the anions in the Dhaleshwari River (DR) India showed higher amounts of Cl, NO_3, SO_4, and PO_4 in the river water [23]. The high nitrate levels > 0.1 mg/L in the water were due to excessive use of nitrate fertilizer or other farming practices in Dhaleshwari India which produced many diseases, such as vascular disorders and ovarian malignancies [24], skin disorders [25], and other infectious diseases [26]. The US EPA/WHO (2009) has suggested that the MCL for nitrate in drinking water is 10 mg/L, and for nitrite, it is 0.1 mg/L. Anionic infiltrations degrade freshwater reservoirs.

Trace metals are the most severe cause of water contamination, especially in developing countries such as Pakistan [27]. Similarly, trace metals are posing significant challenges worldwide, e.g., in Fiji the high concentrations of Pb, Cd and Mn have been reported to be 0.03–0.53 g/L, 0.01–0.95 g/L and 0.01–3.66 g/L [28]. In a study by Abd Byty et al., they showed high concentrations of Zn, Cr, As, Pb, Ni, Co, Cd and Hg as; 2.31, 1.10, 5.78, 0.29, 3.35, 0.32, 0.07 and 5.62, respectively [29]. Gad et al. working on the water quality of the

Nile river, stated the presence of Al, Ba, Cr, Cu, Fe, Mn, Mo, Ni, and Zn at concentrations of 0.52, 0.045, 0.0046, 0.0046, 0.433, 0.047, 0.0092, 0.0183, and 0.0161 mg/L, respectively [30]. Another study on the Qaroun lake in Egypt also reflected the presence of Cd and Cu from industrial releases that contaminated the water quality of the lake [31]. Toxic trace metals, i.e., Zn, Cu, Fe, Mn, Cd, Ni, and Pb, can severely harm human health [32], producing dysfunctional vital body organs, i.e., kidneys and lungs [33].

This study aims to establish a connection between the current water quality conditions of the region from the perspective of regional pollutants (bacterial and mineral) and their impact on the health of resident communities. This investigation of the current water quality parameters in parochial water resources of AJK will help us to develop a relationship between the various fatal diseases that have recently occurred in AJK (such as malignancies, vascular disorders, reproductive anomalies, and gastrointestinal diseases, etc.) with water pollutants of the region. Further, from the analytical methods and results, we have discussed and uncovered important information about the untapped water resources in the AJK region, NW-Pakistan and related them to the recent upsurge in waterborne diseases.

2. Geology of the Studied Area

The district of Neelam (DNLM) is situated in the Himalayan gorge, also known as Neelam valley, which is located north to south of Muzaffarabad, the capital of Azad Jammu and Kashmir (AJK), Pakistan. Neelam valley expands on both sides of the Neelam River (NR) [34]. It is a long, arch-shaped valley home to several waterfalls, springs, gorgeous pine, cypress, and deodar trees (Figure 1). Rough mountains surround Neelam valley with an average height of 2554 m, and some peaks as high as 4570 m [35].

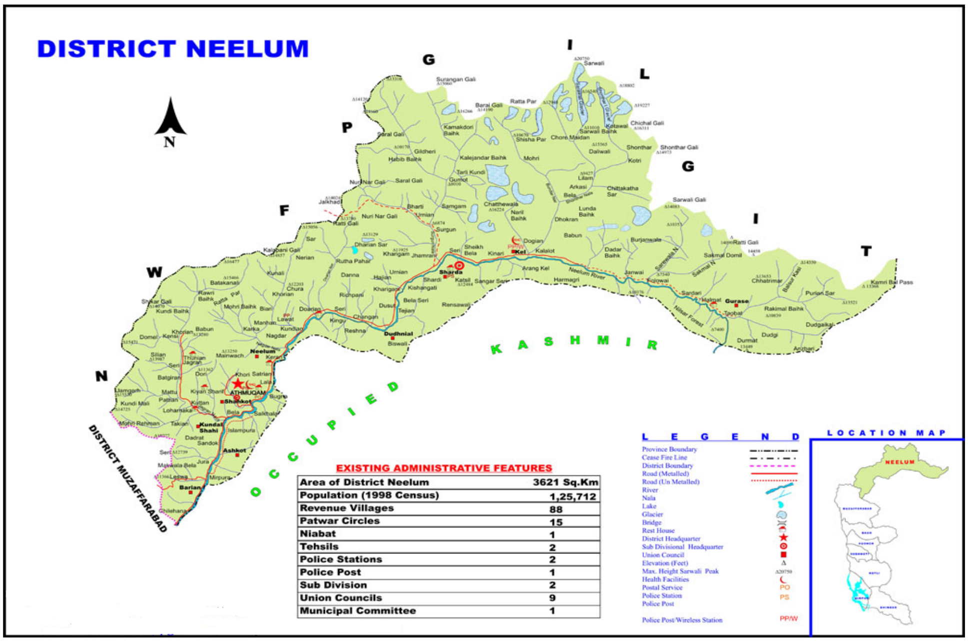

Figure 1. Geographical map showing the geolocation of the district of Neelam, AJK, Pakistan.

3. Materials and Methods

3.1. Sampling Area

The district of Neelam is a hilly region stretching 360 m to the south and 6325 m to the north [36]. It is very beautiful, but it is remote and a challenging area to reach. Most precipitation takes place in winter (average snowfall is 10–12 feet), and the human population relies heavily on natural resources for survival [37]. The current study area begins from Taubutt, Sharda (34°47′23.0″ N, 74°33′56.0″ W) and ends at Challeana, the boundary of DNLM with the district of Muzaffarabad, AJK [38]. The mountain tops are rich in different minerals, i.e., entonite, beryl, fluorite, garnet, hematite, limonite, margarite, mica, quartz, ruby, copper, lead and zinc [39]. Hydrology of the region is polymictic and has predominantly cold climate conditions. The exploration was carried out from September 2021 to November 2021. A total of 60 sampling sites were selected for quality assessment of useable water resources of DNLM.

3.2. Sampling and Analysis

Daylight timings were used for sampling and area analysis. The site locations were selected on the basis of their extensive use and long-term availability of water resources. Water was sampled in Teflon bottles (rinsed with HCl and HNO_3 and then washed with an excess of distilled water) by filling them slowly and replacing the caps to prevent the water from bouncing [40]. To avoid chemical changes during transportation, the samples were frozen in an ice slurry. The separation was made in collected water samples for heavy metal, inorganic, and microbiological specimens, which were placed in encoded bottles. Non-acidified water samples were used for microanalysis, while for heavy metal testing, the samples were acidified with 5% HNO_3 to inhibit bacterial growth. All detection methods used were approved by the WHO and APHA. Chloride was measured by volumetric titration (APHA 2320-B) [41,42], and a UV spectrophotometer XD-7500 (Lovibond, Germany) was used to measure nitrite, nitrate, sulphate and phosphate, showing good linearity with the standards [43] using specific Lovibond kits. The pH of the water samples was measured with a pH meter (Weilheium, Model WTW 82362, Germany), and an EUTECH-PCD650 (Thermo Fisher Scientific, Bremen, Germany) water checker was used to measure EC and turbidity in the collected water samples [44]. All instruments were calibrated against solutions of known concentrations and their accuracy (±0.001) was checked showing good linearity. The linearity was checked after every 5th measurement, hence validating the measurement processes. The ICP-OES Optima 7000-DV (PerkinElmer) was used to check the total concentrations of the heavy metals in the sampled water [45]. The Optima WinLab32™ software was used during the analysis and the response was initially measured by running standard analytes. The colony forming unit technique (CFU) was used to quantify the total colonies of both coliforms and *E. coli* strains using 3M-Petrifilms (3M-USA) and the colonies were distinguished by their different stain colors [46].

4. Results

4.1. Physical Parameters

The pH values, temperature, EC, TDS and turbidity were measured on-site (Figure S1). Table 1 presents the values for the physicochemical parameters obtained from district of Neelam, AJK.

Table 1. The mean concentrations of the selected physicochemical, anionic and heavy metal parameters in the drinking water samples collected from the district of Neelam, AJK.

Parameter	Number of Samples	Minimum	Maximum	Mean	Std. Deviation
EC (µS/cm)	60.0	6.60	974.6	200.1	79.10
Temp (°C)	60.0	9.00	27.00	17.79	3.400
pH	60.0	5.10	9.900	7.471	0.400
TDS (mg/L)	60.0	0.07	912.1	184.0	115.2
Turbidity (NTU)	60.0	0.00	8.300	0.970	0.801
Hardness (mg/L)	60.0	49.0	488.3	198.4	0.802
NO_2	60.0	0.00	9.800	1.320	108.7
NO_3	60.0	0.01	>15.0	3.060	6.930
F	60.0	0.01	4.500	0.410	0.531
Cl	60.0	0.27	30.00	8.720	9.373
SO_4	60.0	8.00	270.0	56.75	20.55
PO_4	60.0	0.50	15.10	4.340	20.55
As	60.0	0.00	0.009	0.0008	0.002
Cr	60.0	0.003	0.057	0.0007	0.003
Cu	60.0	0.002	0.048	0.0029	0.009
Pb	60.0	0.0003	0.037	0.0070	0.005

EC: electrical conductance, TDS: total dissolved solids.

4.2. Bacterial Analysis

The bacterial analysis of the drinking water samples of DNLM was performed with the help of 3M-Petrifilms with CFU expertise. Out of the tested water samples, 76% of were found to be heavily polluted with harmful bacteria. The water was hence rendered unsuitable for drinking or domestic consumption. Despite this, a group of twenty-three samples also contained fecal coliforms, making the regional water even worse for human use. Only fourteen samples were safe to drink, free from impurities. The accuracy and validity of the method was checked after every fifth measurements by running a bacteria-free sample (deionized water).

4.3. Anions

The main anions that were abundantly found in DNLM were nitrites, nitrates, and phosphates (Table 1). For nitrite (NO_2) the WHO recommended limit (2009) is 0.1 mg/L. Numerous samples (n = 22) in the DNLM exceeded the WHO's minimum allowable limit for NO_2. DNLMA-58 Taubutt main (9.80 mg/L), DNLMA-55 Gurase main (8.30 mg/L), DNLMA-47 Kail main (7.60 mg/L), and DNLMA-42 Kishanghati (7.51 mg/L) had the highest doses of nitrite in the water. These samples, including DNLMA-44 Sharda main (6.33 mg/L), DNLMA-53 Sardari (6.41 mg/L), DNLMA-43 Narhgali (5.47 mg/L), and numerous other water samples exceeded the allowable nitrite range. High nitrate (NO_3) concentrations were found at DNLMA-58 Taubutt main (15.0) and DNLMA-55 Gurase main (12.0 mg/L). While just two sites, DNLMA-33 Chaleyana village (270 mg/L) and DNLMA-14 at Bun Chatter (255 mg/L), had greater sulphate ion levels than the WHO recommended limit of 250 mg/L for drinking water.

Numerous samples gathered in DNLM revealed significantly high phosphate and nitrite concentrations. The USEPA established a permissible limit for phosphate ions in water in 1986, which ranges from 0.1 to 0.05 mg/L [47]. DNLMA-14 main Bazar Lawat had high levels phosphate ions (14.9 mg/L) and DNLMA-60 Dudgi Taubutt (12.3 mg/L). Other higher values were in DNLMA-38 Authai Narr (12.1 mg/L), DNLMA-23 Kasrian kerana (10.8 mg/L) and DNLMA-44 Sharda main (11.8 mg/L). More than 90% of the samples had higher concentrations of PO_4 than the WHO's maximum permissible limit.

4.4. Metal Cations

The ICP-OES was used to calculate the concentrations of heavy metals in water samples. Arsenic (As), chromium (Cr), copper (Cu) and lead (Pb) were tested using the ICP-OES technique. The highest concentration of As was 0.0091 mg/L in Mohalla Narr Athmuqam (DNLMC-11) and 0.0087 mg/L and 0.0083 mg/L at Barrian Challeana (DNLMC-1) and Shahkot (DNLMC-5), respectively. At some other sites, chromium exceeded the maximum allowable limits of the WHO (0.05 mg/L) for safe drinking water. Higher Cr uptake was found at 0.0566 mg/L in a distribution tank at Mohalla Authai Narr Athmuqam (DNLMC-38), followed by 0.5620 mg/L at main Bazar Lawat (DNLMC-25), 0.0547 mg/L Lala village (DNLMC-39) and 0.0514 mg/L in Mohalla Farashiyan Kundalshahi (DNLMC-32). Copper was only present in small amounts in the whole valley. The highest values for copper were 0.0487 mg/L at main Chashma upper Neelum (DNLMC-22), 0.0356 mg/L Mohalla Authai Narr Authmuqam (DNLMC-38) and 0.0344 mg/L main Bazar Lawat (DNLMC-25). In the current water analysis, high values of Pb ions (>0.1) were obtained at different locations of DNLM. The maximum values were 0.0371 mg/L in Mohalla Narr Authai THQ-Authmuqam (DNLMC-40), 0.023 mg/L Islampura Kundulshahi (DNLMC-3), and 16 more samples have surpassed the WHO's allowable range for Pb ion concentrations. The lowest Pb concentration was 0.0003 mg/L in Danjar upper Mohalla Authmuqam (DNLMC-18).

The approved WHO limits for water quality parameters are shown in Table 2.

Table 2. Drinking water quality guidelines given by the World Health Organization (2009).

Contaminant	WHO limits
P_H	6.5–8.5
EC	400 µS/cm
TDS	300–900 mg/L
Turbidity	1 NTU
Hardness	60–120 (mg/L)
Cl	250.0 (mg/L)
F	2.19 (mg/L)
NO_2	0.1 (mg/L)
NO_3	10.0 (mg/L)
SO_4	250.0 (mg/L)
PO_4	0.1 (mg/L)
As	0.01 (mg/L)
Cr	0.05(mg/L)
Cu	2.0 (mg/L)
Pb	0.01 (mg/L)
Bacteria	-

(-) indicates no information is available about this parameter.

5. Discussion

In the current study, more than 76% of the water samples were fouled with either total coliforms, *E. coli* or with both contaminants.

The microbial analysis is shown in Figure 2a–c. Aquatic pollution has been a major environmental problem that is threatening public interests on the global level [48]. Among the ecological point sources, bacterial pollution is considered a significant challenge to the modern world [49]. Bacteria, such as total coliforms and *E. coli*, are important contaminants to gauge aquatic pollution [50]. Statistics have explained that about 50,000 humans die across the globe a day due to bacterial diseases caused by polluted water [51]. A recent study that was carried out in Samoa (Upolu Island) showed the presence of many bacterial species, e.g., coliforms, etc. contaminating the water sources [52]. Another study on total coliforms and fecal coliforms was conducted in Mexico and showed the adverse effects on human communities [53]. A similar study was performed by Escamilla et al. in Mexico and showed that water bodies were heavily contaminated with Gram-negative bacteria [54]. These disease-causing microbes enter in fresh water through bacterial- and sewage-polluted estuary [55]. In 1991 a wave of hepatitis-E in Kanpur struck the city in the worst possible way due to the use of polluted drinking water. This disease wave led to nearly a hundred thousand infected people in India [51]. This is quite relevant to the present analysis in which heavy fluxes of biological pollutants were found in the water bodies of DNLM. These pollutants have made the indigenous water sources unfit for human usage. A related study was done by Karanis et al. in the USA on water contaminated with bacteria and showed that nearly half a million people were affected with waterborne illnesses due to polluted water [56]. The contaminated water had a phenomenal socio-economic consequence on the local communities in the USA [57]. Moreover, bacteria and microbes, such as *E. histolytica*, *G. intestinalis*, and *Cryptosporidium*, can cause many infections, i.e., skin diseases, and infections in eye membranes, ear, nose, and throat [58]. We used 3M-Petrifilms for bacterial detection and gauged the number of total coliforms and *E. coli* in the water based on the different colony colors of the CFUs. The microbial contamination in the region was linked to the poor infrastructure, untreated sludge and sewage disposals, openly fouling, animal remains and practically no sewerage system.

In DNLM, the values of anionic parameters were found to be substantially higher than other districts of AJK. The region's geography has a major role in the spread of mineral pollution in the region. High concentrations of anions, i.e., NO_2 in DNLM can be seen in Figure 3a. The places such as DNLMA-55 Gurase main (8.32 mg/L), DNLMA-47 Kail main (7.61 mg/L), DNLMA-42 Kishanghati (7.53 mg/L), DNLMA-44 Sharda main (6.31 mg/L), DNLMA-53 Sardari (6.42 mg/L) DNLMA-43 Narhgali (5.48 mg/L) had higher nitrite concentrations along with nitrate ions (NO_3). Similarly, nitrate increases were found in places DNLMA-58 Taubutt main (15.01 mg/L) and DNLMA-55 Gurase main (12.01 mg/L). The existence of nitrite and nitrate ions in DNLM has been a serious concern because it can affect the alimentary pathway and damage the esophagus and pharynx of the digestive system. Nitrate levels were found high in northern Italy, surpassing 50 mg/L on average with some nitrites [59]. Picetti et al. reported that in Europe, the nitrates and nitrites are involved in causing gastric cancer in humans [60]. Nitrites are carcinogenic, especially when they are combined with other organic species, i.e., proteins, and produce nitrosamines giving rise to malignancies (e.g., ovarian cancer in females and other malignancies) [61]. The studies have proven that continuous exposure to heavy doses of nitrogen compounds cause interthyroid reduction and produce goitrogen [62]. The high concentrations of nitrites and nitrates are related open littering and the use of nitrogen fertilizers in DNLM. These nitrogenous compounds can accumulate in green plants and produce indirect affects to the living ecosystem. The higher concentrations of PO_4 were also found in the water samples of DNLM (see Figure 3b). Phosphate can be dangerous to human health when in excess as it can produce kidney infractions and osteoporosis [63]. Rocks erosion and mineral leaching has been regarded as the main sources of phosphate spread in the region.

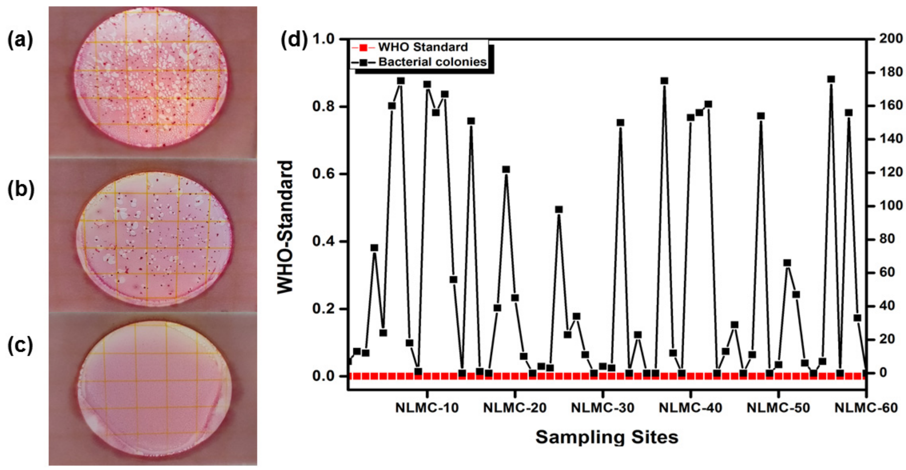

Figure 2. (**a**) shows the presence of total coliforms, (**b**) *E. coli* having different colony colors, (**c**) bacterial-free sample and (**d**) WHO standards for bacterial contamination and the number of colonies obtained at different sites of DNLM in the current experiments.

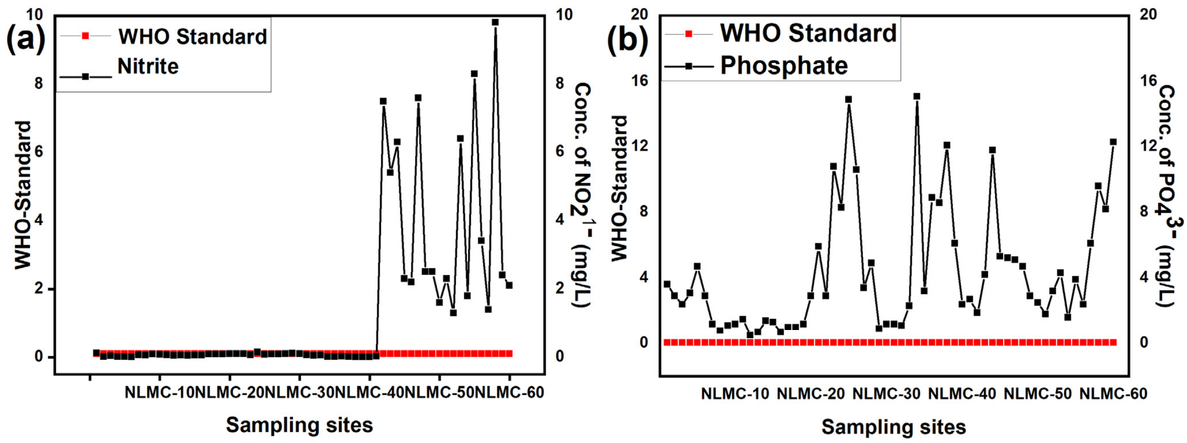

Figure 3. Shows the plot between the high concentrations of NO_2^{-1} vs. the standard values by the WHO at different sampling sites of the district of Neelam, AJK (**a**,**b**) the concentration of PO_4^{3-} against the WHO standard.

The quality and quantity of surface water sources mainly depend on the extent of heavy metal or trace metal content. The heavy metal content in the water samples of DNLM was determined by ICP-OES expertise.

The major heavy metals found in the District Neelam were As, Cr, Cu, and Pb (Figure 4). As and Cr were not found to be as high as the concentrations of Pb, but the presence of the former ions cannot be overlooked. Copper was found within the range of the WHO standards, and Cr showed a slight increase in two sites (DNLM-24 and 47). The highest concentration of arsenic was 0.009 mg/L present at Mohalla Narr Athmuqam (DNLMC-11), followed by 0.0087 mg/L and 0.0083 mg/L at Barrian Challeana (DNLMC-1) and Shahkot (DNLMC-5), respectively. Lead was the major heavy metal contaminant in the water sources of DNLM. In many places in the Neelam valley (i.e., >26% of the studied area), the prevalence of Pb was high. The maximum values were 0.0371 mg/L in Mohalla Narr Authai, THQ-Authmuqam (DNLMC-40), and 0.023 mg/L, Islampura Kundulshahi (DNLMC-3). There were 16 samples which surpassed the WHO allowable range for Pb ions in DNLM. The lowest concentration point was 0.0003 mg/L found in Danjar Upper Mohalla Authmuqam (DNLMC-18). A similar study on heavy metal testing was conducted by Nagajyoti et al., reporting that common heavy metals in drinking water that cause aqueous poisoning are Pb, Fe, Cd, Cu, As, Cr [64]. Another study by Dong et al., in New York (USA) measured high concentrations of Fe and Cd in the state water reservoirs [65]. In a Romanian river Calmuc et al. reported elevated amounts of Ni and Cd that were affecting water bodies in Romania [66]. Zohu et al. studied heavy metal pollution in global rivers. They found that mining, using fertilizers, pesticides and industrial effluents were the leading causes of heavy metal pollution in Asia, North and South America [67]. Nickson et al., reported As in the water samples of Muzaffargarh, a remote district of Pakistan, having As concentrations higher than 0.906 mg/L, especially in shallow sites, while in rural areas, the As concentration was 0.025 mg/L [68]. Another similar study was carried out by Zeng et al. on the Soan River in the district of Rawalpindi, Pakistan, and found that the Cd concentration was 0.1 mg/L, along with Zn at 0.09 mg/L, Co at 0.16 mg/L, Ni at 0.42 mg/L, and Cu at 0.24 mg/L [69]. Zeng et al. in 2020, also measured the concentrations of Cr as 0.11 mg/L, Fe as 0.4 mg/L, Pb as 1.10 mg/L and Mn as 0.16 mg/L in the surface waters of the district of Rawalpindi [70]. Alipour et al. reported Zn, Pb, and Ni as 7.20, 0.67, and 0.21 mg/L, respectively, in Pakistan [71]. An extensive heavy metal exploration study supporting the current data was conducted by Muhammad et al. in Allai Kohistan Pakistan, reported Co, Cu, and Cr as 0.04 mg/L, 0.44, and 1.15 mg/L, respectively [72]. The heavy metal data obtained in this study is comparable to the findings of Ali et al. in Muzaffarabad, AJK, showing that the concentrations of As was 0.097 mg/L, Cr was 0.566 mg/L, and a minute increase in Cu^{2+} to 0.487 mg/L [73]. In DNLM, higher amounts of Pb were found in the most populated areas increasing the risk factors in the area. Local inhabitants of DNLM suffer many diseases, such as kidney and nervous disorders, which can be linked to Pb and other mineral pollutants. Melnyk et al. reported the same toxic effects of Pb as a cytotoxic and neurotoxic element that is a possible human carcinogen of Group-B2. It affects mental development and growth in infants and young children [74]. The heavy metal sources in DNLM include leaded paints, Pb-containing minerals and other anthropogenic sources. In all the tested sites in DNLM (mountainous area), lead–sulfur minerals (PbS) were present that caused higher concentrations of Pb in DNLM. Moreover, muddled expansion and inadequate sewerage systems have also contributed to the inground leaching of harmful domestic wastes that have polluted underground water sources as well. Quick measures to prevent heavy-metal contamination (especially Pb) include reducing the use of plumbing materials, including copper or galvanized steel. These materials can be replaced with Teflon pipes or eco-friendly polymeric materials. The heavy-metal pollution in the area and other contributing pollutants have been the biggest challenge for the people living in the region.

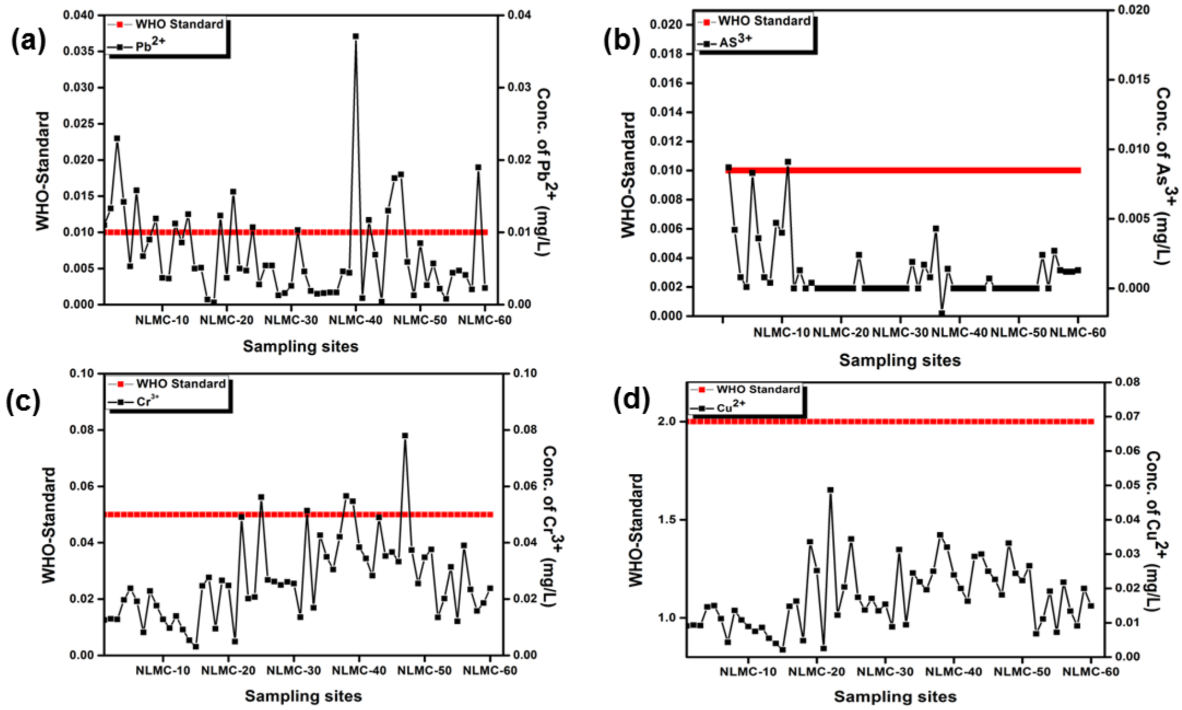

Figure 4. The plot between the concentration of heavy metals vs. the standard values by the WHO at different sampling sites of the district of Neelam, AJK. (**a**) Pb (**b**) As, (**c**) Cr^{3+}, and (**d**) Cu^{2+} against the WHO standards.

Many developing countries around the world are facing a significant challenge in providing clean water sources to its population [75]. Mostly, in these countries, aqueous pollutants are added to fresh water from industrial or pharmaceutical sources, which is quite understandable [76]. However, in DNLM, NW-Pakistan, there is no working industry; therefore, the chances of industrial or pharma-products seeping into the fresh drinking water resources are negligible. Hence, the natural causes of these inorganic mineral pollutants (anionic and toxic metals) in the region are metal extraction processes and poor-quality agriproducts that are ruining the useable water reservoirs. These potent contaminants have raised the risk of many fatal diseases (malignancies and other vital organ diseases) in the region, which were previously never reported. Therefore, there is an urgent need for efficient mitigation technology to neutralize the effects of these aquatic contaminants and ensure safe water supplies to the communities. Many techniques, such as filtration, precipitation, and adsorption, have been used to mitigate mineral and bacterial pollutants [77]. Many of these methods have been adopted by developing countries to decontaminate water pollutants. For example, in Indonesia and Bangladesh, MOFs based on CeO_2/Fe_3O_4 NPs were used to remove the heavy metals As, Cu, Cr, Cd, Pb and Hg from drinking water [78]. Prathna et al., used iron and silver nanoparticles and carbon-based materials to remove heavy metals, i.e., As, Cr and azo-dyes, from communal wastewater [79]. In China, Cu-based biosorbents have been used to treat contaminated water [80]. Recent developments in the field include biobased materials, such as nanocellulose, activated carbons and plant material impregnated with NPs, which have been used for water decontamination [81]. Nanoparticles of different materials, such as FeS, Ag, ZnS, TiO_2, etc., can be incorporated into the biomass, such as activated carbon which can be used later as a water purification material. In view of the literature, we also propose the use of carbon-based materials loaded with NPs (biomaterials) for the removal of heavy metals and bacteria from the parochial drinking water sources of DNLM. The carbon-based NPs have been used for water decontamination; Li et al. used CNTs with T_2O_3 NPs for multifunctional water purification [82]. Yang et al. also studied carbon-based membranes for water treatment [83].

The study was focused on the policy goal 'clean water for all' set by UN-SDG 6 [84], which was also adopted by Pakistan. Although this goal has not yet been achieved, the acquired results provide the blueprints of the active water pollutants in the region, which could be alleviated by applying a focused mitigation approach in the future. Furthermore, the policymakers must address this serious issue of water pollution in the region and minimize or modify the metal extraction procedures so that water pollution can be avoided. Effective and intelligent disposal of the extraction by-products is also needed to avoid contamination. Additionally, there should be a proper analysis of agriproducts used by the farmers in DNLM to find the exact concentrations of the contaminant ions contributed by the agriculture sector to the freshwater sources of the region. The study has provided baseline water quality data of the northern region of Pakistan, which was not reported before this study

6. Conclusions

The current study assessed the water quality indicators of the District Neelam, AJK, Pakistan. The available water resources were alkaline, with high EC, TDS, turbidity and hardness values. The mean concentrations of NO_2 and NO_3 ions surpassed the WHO thresholds but the real threat was the high-bacterial agglomeration (>76%) and heavy metals in most of the fresh-water sources of DNLM, posing more significant health risks, such as cancers and gastrointestinal and reproductive disorders to the regional population. The ICP-OES analysis of heavy metals showed an impactful concentration of lead metal ions. The high concentration of Pb in DNLM waters was due to the mineral diaspora of the region, metal extractions and anthropogenic sources. Additionally, the releases of drainage water, littering at tourist hotspots and poor sewage management downgraded the quality of the water sources in the district of Neelam, AJK, Northwest Pakistan. The study also elaborated on the cause(s) of the high prevalence of fatal diseases (e.g., malignancies) in recent years and indicated some mitigation technologies for future work. Hence, the current investigation has provided baseline data on the water resources of the region which will be helpful to developing efficient diagnostics for treatment solutions for the current health dilemma in DNLM. However, we recommend that more work is focused on the analysis and decontamination of the water resources of the northern regions of Pakistan.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/w15010077/s1, Figure S1: Plots show the concentrations of the physical parameters vs standard WHO values (a) pH, surpassing the standard values, (b) EC, with higher values than WHO-STD (c) TDS values with increments at some sites, (d) Turbidity increase vs WHO standard limit. Figure S2: Plots show the concentrations of the anions vs standard WHO values (a) $NO_3{}^{-1}$, surpassing the standard values at two sites, (b) $SO_4{}^{2-}$, with higher values than WHO-STD (c) F^{-1} values with increments at one site, (d) hardness increase vs WHO standard limit crossing almost at all sites. References [85–87] are listed in Supplementary Materials file.

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