Disproportionate Water Quality Impacts from the Century-Old Nautanen Copper Mines, Northern Sweden

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Received: 23 January 2020; Accepted: 11 February 2020; Published: 13 February 2020

Abstract: Pollution from small historical mining sites is usually overlooked, in contrast to larger ones. Especially in the Arctic, knowledge gaps remain regarding the long-term mine waste impacts, such as metal leakage, on water quality. We study the small copper (Cu) mines of Nautanen, northern Sweden, which had been in operation for only six years when abandoned approximately 110 years ago in 1908. Measurements from field campaigns in 2017 are compared to synthesized historical measurement data from 1993 to 2014, and our results show that concentrations of Cu, Zn, and Cd on-site as well as downstream from the mining site are order(s) of magnitude higher than the local background values. This is despite the small scale of the Nautanen mining site, the short duration of operation, and the long time since closure. Considering the small amount of waste produced at Nautanen, the metal loads from Nautanen are still surprisingly high compared to the metal loads from larger mines. We argue that disproportionately large amounts of metals may be added to surface water systems from the numerous small abandoned mining sites. Such pollution loads need to be accounted for in sustainable assessments of total pollutant pressures in the relatively vulnerable Arctic environment.

Keywords: abandoned mines; mine waste; metal mass flows; Arctic

1. Introduction

Historical mining operations commonly deposited waste rocks, tailings, and metallurgical slags on pristine ground or in nearby lakes or streams [1]. As a consequence, historical mine waste has direct contact with terrestrial and aquatic environments and ecosystems, which implies a high risk for the development of acid mine drainage if the waste contains sulfidic material. Compared to present mining operations in most countries, where mine effluents are becoming increasingly controlled, abandoned historical mine waste represents an uncontrolled source of metal pollution at the landscape level [2], which hampers long-term sustainable resource management.

Long-term contamination from historical mining is usually reconstructed from bed sediments sampled from downstream surface water systems. For instance, sediments from the Big River, central USA, have elevated metal concentrations along a 170-km stretch as a result of 120 years of mining [3]. Mass flows of metals through surface water, on the other hand, directly reflect continued loads of mobile metal phases into freshwater ecosystems. Considering riverine mass flows of copper (Cu), the historical mining districts in the Odiel drainage basin in Spain have, for example, been estimated to contribute as much as 1300 tons of Cu per year to the surface water system [4]. Within the drainage basin, which covers 2300 km$^2$, mining operations began in prehistoric times [5], which could explain...
the magnitude of the Cu mass flows. A release of up to 160 tons of Cu per year was estimated for the Britannia mine, in south-western Canada, to coastal areas along an 80 km underground tunnel system [6]. The more than 1000-year-old Falun mine, in central Sweden, released (pre-remediation) about 30 tons Cu per year to downstream surface waters [7]. Analogously, much of the existing literature on historical mining regards large scale and relatively well-defined “worst-case” pollution sites where a massive metal pressure on aquatic and terrestrial ecosystems is obvious.

Research on long-term (> 100 years) mine waste degradation and spreading processes are essentially lacking from the Arctic, in contrast to the relatively well documented impacts of historic mines, e.g., in the Mediterranean or from the temperate climate zones of Europe or North America. Follow-up long-term monitoring and groundwater data are most often not available from northern latitudes [8]. This knowledge gap should be of increasing concern, as the Arctic is likely to face high pressures from mineral extraction in the near future [9]. In addition, the Arctic is already facing multiple pressures on the biotic and abiotic environment due to climate change and other societal activities [10].

Current knowledge of historical mine waste impacts in the Arctic has mostly been gained from sites that were abandoned during the last few decades. For example, the Black Angel mine in western Greenland closed in 1990 and its subsea tailing deposit was found to contribute to metal accumulation in local fish tissue [11]. Studies on older mine waste are rare, which hampers the understanding of long-term impacts of Arctic mining operations. Existing (70 years old) examples include the Sherritt-Gordon mine in central Canada and the Laver mine in northern Sweden, which, currently, continue to release metals to their respective local environment [12,13]. Furthermore, the impact of the numerous, small mining sites (e.g., < 1 million tons of produced tailings) are poorly investigated, which inhibits, e.g., the upscaling of total historical loads to regional scales.

The Nautanen Cu mining site in northern Sweden is a clear example of a small historical site comprising mine waste from only six years of active production more than 100 years ago (1902–1908). Existing monitoring data from the Nautanen site provide us with a rare opportunity to study the long-term environmental impacts from a well-defined abandoned Arctic mining site. This may increase the understanding of historical mining impacts in other regions of the Arctic, including Russia [14,15] and Finland [16]. For example, soil Cu contamination has previously been found impact biota in the vicinity of the mine tailings, e.g., lowering the cold tolerance of the oligochaete worm [17]. The water-borne spreading of Cu and its potential consequences for ecosystem functions needs to be further studied, especially in colder climates. The hereto unexploited Cu ore at Nautanen is currently under consideration for extraction, which highlights the need for a thorough characterization of the site’s current metal contamination. The main objectives of this study were therefore to examine (i) the present situation and potential temporal trends in how the Nautanen mines impact surface water metal concentrations (relative to regional backgrounds), (ii) present and cumulative metal riverine mass flows since the mine closed, and (iii) the downstream mass flows in relation to the amount of deposited mining waste within the catchment, facilitating a comparison of impacts with other Arctic mines (of various sizes).

2. Materials and Methods

2.1. Site Description

2.1.1. Historical Background

The Nautanen copper mines were owned by the company Nautanen Kopparfält AB. Mining production began in 1902 and was in operation only for six years before shutting down in 1908 due to unexpectedly limited ore reserves [18]. About 72,000 tons of unprocessed ore were mined during this short period, which resulted in 5700 tons of Cu concentrate and 4600 tons of iron (Fe) concentrate [19]. Assuming that all the mined Cu concentrate was chalcopyrite (CuFeS₂ with 35% Cu [20]), which is the most common Cu mineral at Nautanen, the total Cu production was approximately 2000 tons. The
mining site included five larger mines named: Dagny, Hoppet, Maria, Max, and Fredrik (Figure 1), a small residential area and a cable railway system. The crusher, smelter, and power station were located in the central parts of the mining site, hereafter referred to as the industrial area (Figure 1) [21]. As of 2017, there was about 10,000 tons (5500 m$^3$) of unconfined tailings and up to 10,000 tons (5000 m$^3$) of metallurgical slag products in the industrial area [22]. After the mining site closed, the buildings and infrastructure components were removed and Nautanen was abandoned a few years later. While in operation, the mines at Nautanen were estimated to have produced a total of 68,000 tons of waste rock, which mostly were dumped outside the Max and Maria mines [18]. As the waste rock was found to contain a relatively high Cu grade, most of the dry-lying heaps were removed in 2005–2008 for re-processing at the nearby, currently active Cu mine, Aitik, which is owned by Boliden AB [23]. Waste rocks were still present in 2017 in the Maria Lake (Figure 1) and in smaller heaps spread across the mining site. So far, no thorough remediation has been implemented at Nautanen. The area is primarily used for recreational purposes and winter-spring reindeer herding by the Baste ˇ cearru Sami community [24]. An exploration permit has allowed Boliden AB to investigate the Nautanen area for possible new mining since 2009 [25]. Some relatively recent publicly available documents therefore describe the ore deposits and potential mining operations [26].

2.1.2. Climate and Environmental Setting

The annual average temperature is $-1.6^\circ C$ with summer (June–August) and winter (December–February) temperatures averaging around 11 °C and $-13^\circ C$ respectively (based on data of the 25-year period 1993–2017 from the Climatic Research Unit (CRU) at the University of East Anglia described by Harris et al. [27] (see Supplementary Materials S1 for details). The same dataset showed that annual average temperatures have increased by 1.0 °C during the last 110 years (1908–2017). The annual average precipitation is 560 mm/yr (for 1993–2017) and compared to the first quarter of the century (1901–1925), total annual precipitation has increased by 23% (based on CRU data). Nautanen is located within a larger active mining district where the Malmberget mine (Fe) and Aitik mine (Cu) are the two largest mines in operation within a 10–15 km distance (Figure 1). Nautanen and Aitik are connected through the Nautanen Deformation Zone (north-north-west direction), which consists of metamorphosed volcanoclastic sedimentary rocks with Cu-Au ± Fe mineralization [28]. The formation hosts mainly sulfide deposits of chalcopyrite (CuFeS$_2$), pyrite (FeS$_2$), and magnetite (Fe$_3$O$_4$) [20]. The landscape is characterized by Veiki moraine (ice walled lake plains) where the geomorphology of the Nautanen area is characterized by exposed bedrock or till [29]. The headwaters of the Imetjoki stream system originate from two smaller lakes situated about 1.5 km north of the Nautanen mining site (Figure 1). The Imetjoki stream flows along the northern side of the mining site partly passing through wetlands. The stream drains the Northern Lake and Imetjärvi, the latter being the outlet for the Maria Lake. In the industrial area, Imetjoki flows across the tailings where the groundwater level has been measured to be located at only 0.5–1.5 m depth [18]. Downstream of the Max mine the groundwater level was estimated to be located below 3 m depth [22]. Imetjoki joins the Nietsajoki stream at 4 km downstream of the mining site, which gives the Imetjoki a drainage area of 6.6 km$^2$. Nietsajoki joins the Lina River another 8 km downstream and then eventually drains into the Kalix River and the Baltic Sea.
Figure 1. Map of the Nautanen mining site in the Imetjoki drainage basin, where sampling zones for the measurement campaigns in May and August 2017 are marked. Local background samples were taken within the green rectangles (in the upstream part of the Imetjoki drainage basin and in the reference rivers Nietsajoki and Lina River). Samples of the main mining site were taken within the red rectangles, and downstream samples were taken within the blue rectangles.
2.2. Methods

2.2.1. Field Measurements and Data Synthesis

Two field campaigns were conducted at Nautanen in 2017; one during May (snowmelt conditions) and one in August (after ground thawing; Figure 1). Water quality sampling and discharge measurements were performed simultaneously at nine sampling points, complemented with seven additional sampling points for water quality only. The sampling points were chosen to represent four zones: (i) local background conditions at locations judged to be unaffected by the past mining activities (i.e., in the Imetjoki stream, upstream of the main mining zone, and in two reference rivers in the close vicinity; Figure 2a), (ii) at the main mining zone (Figure 2b), (iii) directly downstream of the main mining zone (Figure 2c; D1 in Figure 1), and (iv) 4 km downstream of the main mining zone (Figure 2d; D2 in Figure 1).

Figure 2. Photographs showing conditions at the sampling zones: (a) local background (here specifically the upstream lake of the Imetjoki stream); (b) main mining; (c) directly downstream of the main mining zone, D1; and (d) 4 km downstream of Nautanen mining site, D2.
The two reference rivers were the Nietsajoki stream and the Lina River (Figure 1). Water samples for analyses of metal concentrations and base chemistry were taken in the streams in triplicates for total concentration and dissolved concentration (having passed a 0.22 µm Filtropur filter), and acidified according to the sampling protocol in the Supplementary Materials S2. pH, electric conductivity, alkalinity, water temperature, and turbidity were also measured in situ (see Section S2 in the Supplementary Materials for instruments and sampling details). The stream discharge at each point was estimated from measuring water velocity with an A. Ott Kempten propeller current meter (at 60% of the total depth from water surface at each width increment) by wading along a cross section of the stream.

In addition to the conducted local Nautanen measurements, we considered regional background concentrations reported by Herbert et al. [30], reflecting element concentration ranges observed in northern Sweden between 1996 and 2007 in streams judged to be unaffected by point pollutant sources. Existing historical field measurement data on water quality in the Nautanen region were also synthesized from publicly available documents both online and in state archives, mostly in the form of technical consultancy reports ordered by governmental agencies. More specifically, water quality measurements have been conducted at Nautanen on several occasions and open data were available from 1993 to 1994 [19,31], 2001 [18], 2005 to 2009, and 2014 [23]. Laboratory analyses and details of all measured and historical samples are described in Section S2 in the Supplementary Materials. Specifically, the elements copper (Cu), zinc (Zn), cobalt (Co), cadmium (Cd), nickel (Ni), molybdenum (Mo), barium (Ba), manganese (Mn), aluminum (Al), and iron (Fe), together with dissolved organic carbon (DOC) and sulfate (SO₄), were analyzed in detail due to their common association with mining activities and related geochemical processes [32–34].

2.2.2. Mass Flow Analysis

Mass flows of elements at Nautanen were calculated in two ways considering the Imetjoki stream and its tributaries; as snapshot mass flows, and as long-term average mass flows. In addition, the cumulative element mass carried by Imetjoki was estimated over the approximately 110 years that has passed since the mine closed in 1908. The quantifications considered conditions in four zones (or locations): (i) upstream of the main mining zone, (ii) at the main mining zone, and downstream of the main mining zone at locations (iii) D1 and (iv) D2 (Figures 1 and 2).

The snapshot element concentration within each sampling zone was calculated by averaging the inflowing tributary water with the main Imetjoki stream water. Specifically, for sampling points within a considered zone $j$, with measured discharge $q_1$ and element concentration $c_1$, where Imetjoki was joined by a tributary with measured discharge $q_2$ and measured total element concentration $c_2$, the snapshot element concentration in Imetjoki immediately after the confluence point was calculated as a flow weighted average concentration, $\bar{c}_j$. This was done for each of the four sampling zones ($j$; upstream, main mining zone, D1 and D2) along the main Imetjoki stream according to:

$$\bar{c}_j = \frac{c_1 q_1 + c_2 q_2}{q_1 + q_2}. \quad (1)$$

The corresponding snapshot element mass flow for sampling zone $j$, $mf_j$, was calculated as:

$$mf_j = \bar{c}_j (q_1 + q_2). \quad (2)$$

Concentration enrichment factors and relative mass flow increases were calculated by normalizing the respective zonal concentration and mass flow values by the corresponding values for the upstream zone. Since upstream sampling could not be performed in the May field campaign (due to snow conditions), we only used results from the August field campaign (where sampling in all zones were performed) in the mass flow analysis.
The long-term average mass flows ($\overline{MF}_{L,j}$) for a 25-year period (1993–2017) in each of the sampling zones $j$ were calculated by:

$$\overline{MF}_{L,j} = \overline{C}_{L,j} \overline{Q}_{L,j},$$

(3)

where $\overline{C}_{L,j}$ is the long-term average concentration for each of the sampling zones, and $\overline{Q}_{L,j}$ is a corresponding long-term average discharge. Regarding $\overline{C}_{L,j}$, we used the assumption of (approximate) geochemical stationarity, i.e., that the inter-annual metal concentration variation is typically low compared to the much higher variability in discharge [35]. This allowed us to estimate $\overline{C}_{L,j}$ as approximately equal to $\overline{C}_j$, i.e., that long-term average concentration would be equivalent to our average zonal snapshot concentration ($\overline{C}_j$). Concerning the long-term average discharge, $\overline{Q}_{L,j}$, we used a water balance approach based on monthly precipitation and temperature measurements provided by the Climatic Research Unit (CRU) at the University of East Anglia [27] to estimate the average discharge at the downstream location D2 ($\overline{Q}_{L,D2}$; see Section S3 in the Supplementary Materials for water balance calculations), for the period 1993–2017. Our resulting evapotranspiration (estimated from equations from Turc [36] and Langbein [37]) was furthermore compared to independent evapotranspiration quantifications based on available, region-specific precipitation and stream flow data from the Swedish Meteorological and Hydrological Institute (SMHI). $\overline{Q}_{L,D2}$ was further scaled to quantify the discharge in the upstream sampling zones, based on the relative catchment area of each sampling zone ($A_j$), yielding the following equation for calculating long-term average discharge of sampling zone $j$:

$$\overline{Q}_{L,j} = \overline{Q}_{L,D2} \frac{A_j}{A_{D2}}.$$  

(4)

The average mass flows at the downstream location D2 ($\overline{MF}_{L,D2}$) were furthermore compared to the total mass of deposited tailings and slag products at the industrial area to yield a so-called “stream load-to-tailings” ratio. The industrial area was considered as the main pollutant source within the catchment [22] and the ratio was, therefore, calculated by dividing the amount (mass) of tailings and slag products with $\overline{MF}_{L,D2}$. This stream load-to-tailings ratio can be seen as a first-order indicator, facilitating order-of-magnitude comparisons of environmental impacts between mining sites of different sizes. The ratio is mainly relevant when particulate flows are negligible (as in Nautanen), in which case, the geochemical impacts from land disturbance can be approximated by the mass of deposited wastes. For comparison, under conditions where particulate flows are dominant, one can alternatively consider surface erosion rates per unit of disturbed lands (e.g., Walling and Fang [38]).

Last, the 110-year (1908–2017) cumulative mass for each sampling zone ($M_{110,j}$) was estimated by:

$$M_{110,j} = \overline{C}_{100,j} \overline{Q}_{110,j} \cdot 110.$$  

(5)

Similar to Equation (3), $\overline{C}_{100,j}$ is here used as equivalent to the snapshot $\overline{C}_j$. $\overline{Q}_{110,j}$ is the average discharge for the 110-year period (1908–2017; see Section S3 in the Supplementary Materials).

3. Results

3.1. Water Quality

The stream discharge measured at Nautanen in May 2017 was relatively low for the season. Much of the winter snow still remained on the ground, without contributing to melt water-related flow peaks that frequently occur in May. The August 2017 discharge levels were consistent with previously recorded measurements (August–September in 2001–2002 [18]). Both in May and August, most elements existed predominantly in the dissolved phase (a $t$-test showed no significant difference at $p = 0.05$ between dissolved and total concentrations). The pH ranged between 3.1 and 6.4 at the industrial area (i.e., where the tailings are located within the main mining zone). The same pH-range (3.8–6.3) was measured at the lake outlets in the upstream zone. Other sampling points within the
main mining zone had a pH around 6.1, while the downstream locations D1 and D2, as well as the reference Nietsajoki and Lina rivers had pH closer to 6.8 (see Tables S2 and S3 in the Supplementary Materials for a complete list of all water quality parameters and discharge).

Both of the field campaigns conducted in 2017 (May and August) are represented in Figure 3, where the total (unfiltered) concentrations for selected elements are grouped by sampling zone. Each element is shown together with its corresponding regional background concentration range (shown in grey; no regional values were available for Mo, Ba, and SO₄). Our results show that, except for Fe, all metal concentrations in the water samples collected from the main mining zone (red boxes; Figure 3) were significantly higher compared to local background concentrations (green boxes) and concentrations at the downstream locations D1 and D2 (blue boxes). For instance, the median Cu, Zn, Co, and Ni concentrations at the main mining zone exceeded median local background concentrations by as much as one to three orders of magnitude. Many metals (Cu, Zn, Co, Cd, and Mn) additionally showed considerably higher mining zone concentrations than the regional background concentrations. Even at the downstream locations D1 and D2, the metal concentrations were in most cases elevated compared to background values. Notably, the trend of Fe differed from other metals, following instead the same trend as DOC in that the downstream concentrations in general exceed those of the mining zone.

![Figure 3](image)

**Figure 3.** Boxplots of total log concentrations (µg/L) of selected parameters from Nautanen in May and August 2017 per sampling zone: local background (incl. upstream and reference rivers), mining, and downstream. Whiskers represent 1.5 times the inter-quartile length and black points show values outside that range. Grey areas indicate regional background ranges for northern Sweden (except for Mo, Ba, and SO₄ where no regional background values were found). All concentrations are given in Table S3 in the Supplementary Materials.

### 3.2. Concentration Enrichment and Element Mass Flow Increases

Concentration enrichment factors and snapshot mass flow increases are presented in Figure 4 for the selected elements considering the main mining zone (red bars) and downstream locations (blue bars), relative to upstream conditions (green area), where the enrichment factors and relative increases of mass flows per definition are of equal unity. The highest concentration enrichment factor of all investigated elements was found for Cu at the main mining zone, with a value of about 140 (Figure 4a). The mass flow of Cu increased even more than that, by 290 times over the main mining zone (Figure 4b), which is due to a higher stream water discharge at the main mining zone than upstream of it. Many elements (Cu, Zn, Co, Cd, Ni, Mn, and SO₄) showed considerable concentration enrichment (factors
was calculated to 120 kg from upstream of the main mining zone, 43 tons for the main mining zone.

At the downstream location D1, these elements displayed distinctly lower relative mass flow increases, although the increases were still greater than or equal to 8 (average 26). The downstream location D2 displayed similar mass flow increased relative to the upstream conditions at the D1 location. In contrast, due to dilution effects, the concentration enrichments were much less pronounced at the D2 location than at D1. Other elements, like Fe and DOC displayed no or low concentration enrichments (factors near unity) over the mining site, whereas mass flows still increased all the way along the main stretch of the stream, due to the element supply with inflowing water from tributary streams.

Figure 4. Relative change (in log scale) from upstream to the main mining zone (red color) and between the upstream and downstream sampling zones (blue colors) shown as (a) concentration enrichment factors and (b) as relative mass flow increases at Nautanen in August 2017. Values >1 represent an increase, <1 represents a decrease, and values equal to 1 indicates no change.

Considering the surveyed 25-year period (1993–2017), the average mass flow of Cu leaving the upstream zone was estimated at 1.2 kg/yr, whereas the mass flow of Cu leaving the main mining zone was estimated at 450 kg/yr. At both the downstream locations D1 and D2, the mass flow of Cu was lower, although still considerable at about 70 kg/yr. The identical average mass flow values of Cu at these two locations (D1 and D2) implies that no major net addition or net removal of Cu occur along the 4 km pathway that separates the points. These mass flows were based on long-term average discharges of 450,000 m³/yr at the D1 location and of 2,000,000 m³/yr at the D2 location for the period 1993–2017 (i.e., Q_{L1D1} and Q_{L2D2}, respectively). During this 25-year period, the annual discharges showed a coefficient of variation of 0.23. Q_{L2D2} was in turn derived from a calculated actual evapotranspiration (ET\textsubscript{a}) of 260 mm/yr, which is within the reported range of 260–310 mm/yr for 1981–2010 for watersheds close to Gällivare City, as calculated by the Swedish Meteorological and Hydrological Institute [39]. Previous estimates of annual mass flows of Cu ranged between 78 and 240 kg/yr for the main mining zone. The total Cu mass that has been transported through each sampling zone during the approximately 110 years that has passed since the mine closed (1908–2017) was calculated to 120 kg from upstream of the main mining zone, 43 tons for the main mining zone,
and 6.7 tons for the downstream locations D1 and D2. During these 110 years the discharge was estimated to increase with approximately 35% from the first quarter of the century (1901–1925) to the last (1993–2017), in response to changing hydroclimatic conditions. Finally, the stream load-to-tailings ratio for Nautanen was estimated at $10^5$ per year based on the average Cu load in surface waters 4 km downstream of the main mining zone (70 kg/yr) and the amount of deposited tailings and slag products in the industrial area (20,000 tons).

3.3. Historical Cu Measurements

The historical measurements of total Cu concentrations in Nautanen are synthesized in Figure 5, which shows the results from 1993 to 2014, together with the present field campaign results from 2017. The scattering in local background levels reflects different sampling locations between the various historical measurement campaigns. This is also the case for the wide spread Cu concentrations at the mining zone. The highest on-site Cu value was measured in 2017, pertaining to a tributary stream flowing directly over the tailings that had not been monitored before. A general spread in concentrations across the main mining zone has been seen throughout the different measurement campaigns conducted between 1993 and 2017. However, the Cu concentration values at the downstream locations D1 and D2 has been similar over this time period. The lowest D1 concentration in Figure 5 (at 20 µg/L) pertains to a small stream downstream of the Fredrik mine (Figure 1), and unlike the other D1 measurement locations, is therefore not representative of the main Imetjoki channel. The Cu concentrations after Imetjoki and Nietsajoki joins (smaller dark blue points in Figure 5) dropped by one order of magnitude, due to dilution from the much higher discharge of Nietsajoki. These concentrations (3.9–7.9 µg/L) are still higher than most of the local background values (see Table S4 in the Supplementary Materials for a full list of synthesized historical data). For other elements than Cu the historically available records show little information. Zn and Cd were measured at a few locations in 1993–1994, 2001, and 2014, and the results show similar concentration ranges in their corresponding sampling zone as were found in 2017 (Figure 3).

![Figure 5. Total Cu concentrations (in log scale µg/L) from water samples from Nautanen taken in 1993–1994, 2001, 2005–2009, 2014, and 2017, grouped by sampling location (local background, mining zone, and downstream locations). All concentrations and measurement locations are outlined in Tables S3 and S4 in the Supplementary Materials.](image-url)
4. Discussion

The historical activities at the Nautanen mining site cover a short time period (1902–1908) and are well documented. From the results of this study, we inferred that Cu concentrations were relatively constant during the surveyed 25-year period (1993–2017). For example, average Cu concentrations just downstream of the Nautanen mining site, and 4 km downstream of it, were 140 and 43 µg/L, respectively, according to measurements during the period of 1993–2014. These concentrations were similar to the average values of the conducted 2017-campaigns (130 and 38 µg/L respectively). Furthermore, our results showed that the average Cu loads in surface waters 4 km downstream of the main mining zone (70 kg/yr during 1993–2017) relative to the amount of tailings and slag products produced (20,000 tons), i.e., the stream load-to-tailings ratio, was to $1 \times 10^5$ per year.

The much larger abandoned Laver Cu mine in Sweden (1.2 million tons tailings) and the Sherritt-Gordon Cu mine in Canada (7.4 million tons tailings) both have stream load-to-tailings ratios of $1 \times 10^7$ per year, based on the stream mass flows of Cu at similar downstream distances as for Nautanen [12,13]. This means that these mining sites, compared to Nautanen, release significantly less Cu to surface waters relative to their total waste production. The tailings of the Sherritt-Gordon mine are, for instance, roughly 370 times larger than at Nautanen, whereas the Cu release rate is only about six times larger. Both the Laver and Sherritt-Gordon sites share similar characteristics as Nautanen; e.g., climatic conditions, a long time (70 years) since abandonment, and (mostly) un-remediated waste.

Our results therefore demonstrate that the combined impact of the remaining factors is large, including, e.g., impacts of varying abundance and availability of Cu in the host rock, weathering potential of the waste, and hydrological flow paths, to mention a few [32]. Notably, the existence of systematic differences in stream load-to-tailings ratios between small and large mining sites cannot be excluded. Some governing factors may, for example, be scale dependent, such as the surface area to volume ratio of the waste heaps, which, in turn, can impact weathering rates. The fact that the small Nautanen mining site had disproportionately high environmental impact, compared to the few existing quantifications of larger Arctic mining sites, illustrates a need to increase the general understanding of complex hydrogeochemical processes across different Arctic mining sites.

In Sweden, closed and abandoned sulfide mines have been estimated to load surface waters with a total of about 12 tons/yr of Cu [40]. This estimate is, however, highly uncertain and is based on a survey where the relevant County Administrative Boards provided data for monitored sulfide mines, which only represent a small fraction of the roughly 1000 known abandoned sulfide mines in Sweden [41]. Most of the smaller mining sites are unmonitored and could, therefore, not be included in those data sets. A similar regional-scale synthesis of historical mining pollution in England and Wales estimated the Cu loading of surface waters to be 19 tons/yr [2]. Thus, pollution from abandoned mines is associated with a substantial metal mass flux at the landscape-level and unaccounted-for smaller mines (similar to Nautanen) may increase this estimate even more.

Whereas the above-discussed metal concentrations and loads regard conditions in the surface water systems downstream of the main mining zone of Nautanen, our results additionally showed that the on-site stream water, at the middle of the mining site, has much higher aqueous concentrations and hence carries considerably higher metal loads (with, for example, a flow weighted average Cu concentration of 990 µg/L and a load of 450 kg/yr). Considering the observed absence of temporal trends during the last quarter of a century, together with the assumption that aqueous concentrations and metal loads should not have been lower than that in the beginning of the 20th century, when the mine was new, we estimate the total, on-site Cu loading of the stream corresponds to at least 43 tons since the closure of the mine (1908–2017).

For comparison, the total amount of produced Cu until 1908 was 2000 tons. Thus, for every ton of produced Cu, about 20 kg has been dissolved into the on-site stream water. Taken together, apart from observed spreading of metals to downstream waters, the above results reflect an on-going and considerable spatial re-distribution of metals within the local mining site. Specifically, (I) a net dissolution of metals in the upper parts of the main mining zone, leading to the observed
order-of-magnitude in-stream mass flow increases (the difference between the green and red bars in Figure 4b), and (II) a net retention of metals in the lower parts leading to order-of-magnitude in-stream mass flow decreases (the difference between the red and blue bars in Figure 4b). In particular, there was a small stream that carried very high mass flows of Cu, Zn, Co, Cd, and Ni in the middle of the Nautanen mining site, just upstream of the junction with the main stream Imetjoki.

The observed decrease in Cu mass flows of about 380 kg/yr (450 – 70 = 380) as the small stream joins Imetjoki could potentially be explained by precipitation from the aqueous phase, e.g., as a result of diffuse inflows of groundwater with a different chemical composition (as previously seen in, e.g., Kimball et al. [42] and Palumbo-Roe and Dearden [43]). Even if the precipitated metals (Cu) would have accumulated primarily in the stream sediments, they are at risk of re-mobilizing in the future due to changing biogeochemical conditions or due to extreme weather events. In any case, the vegetation at and near the main mining zone of Nautanen has already been shown to be negatively impacted by the mining activities [23].

Projected future warming (+4 and +5.6 °C for RCP 4.5 and RCP 8.5, respectively) by the year 2100 for Norrbotten County is expected to shorten the period during which streams and lakes are frozen [44]. Specifically, winter (December-February) temperatures are predicted to increase the most, which will increase the likelihood of precipitation occurring as rain instead of snow, thus enabling an increased movement of water and metals through the water system over the year. This also implies that the water equivalent of the accumulated winter snow pack may decrease, which, in turn, would reduce the amplitude of the spring snowmelt runoff [10]. Even if high meltwater flushes were becoming less prevalent, peak precipitation events have the potential to increase erosion and the transport of material from the freely exposed tailings at Nautanen. Furthermore, a potential effect from a projected increased and more variable precipitation (by +20% and +30% for RCP 4.5 and RCP 8.5, respectively) is a higher mean groundwater table and larger level fluctuations around the mean value. In such conditions, the washing-out of metals from tailings to the mobile aqueous phase at Nautanen would increase [12].

5. Conclusions

From the results of the field measurements at the Nautanen mining site (1993–2017), we conclude that:

- Despite the small spatial scale of the Nautanen mining site, the short duration of operation, and the long time (approximately 110 years) since closure, the average concentrations of Cu, Zn, and Cd on-site (990, 280, and 1.0 µg/L, respectively) and downstream of the mining site (150, 50, and 0.2 µg/L, respectively) were generally considerably above local (7.2, 3.0, and 0.010 µg/L, respectively) and regional (1.6, 11, and 0.10 µg/L, respectively) background values. In particular, downstream Cu concentrations were consistently high throughout the surveyed 25-year period (1993–2017).
- The on-site mass flows of Cu, Zn, Co, and Cd were estimated to be between 100 and 300 times higher than upstream of the mining site. For Cu, the average mass flows were 1.2 kg/yr upstream of the main mining zone, 450 kg/yr on the main mining zone, and about 70 kg/yr 100 m to 4 km downstream of the site. Many metals exhibited a similar spatial pattern, indicating an on-going considerable retention of metals at or near the main mining zone, corresponding to 380 kg/yr for Cu.
- Compared to other major abandoned mines in the Arctic, the metal loads from the Nautanen mining site were found to be unexpectedly high relative to the (small) amount of tailings and slag products produced. More generally, our approach based on stream load-to-tailing ratios show that small abandoned mining sites, which are numerous, could add disproportionately large amounts of metals to the surface water systems. Such effects need to be accounted for in assessments of total pollutant pressures in the relatively sensitive Arctic environment.
Supplementary Materials: The following are available online at http://www.mdpi.com/2071-1050/12/4/1394/s1. This supplementary material provides a description of the field measurement sampling protocols and water balance calculations. It also tabulates water quality parameters from the field measurements in 2017, and synthesizes historical data (period: 1993–2014). S1: Temperature and precipitation data, S2: Water sampling procedure and laboratory analyses, Table S1: Water sampling protocol for the Nautanen field campaign 2017, S3: Water balance calculations, Table S2: Results in base chemistry of surface water samples from the 2017 measurement campaign at Nautanen, Table S3: Results of total element concentrations in surface water samples from the 2017 measurement campaign at Nautanen, Table S4: Synthesis of historical field measurement campaigns at Nautanen.

Author Contributions: All authors participated in the field work; formal analysis: S.F.; data curation: S.F.; Supervision: J.J. and G.R.; writing-original draft: S.F.; writing-review and editing: J.J., G.R. and S.R.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Nordforsk Centre of Excellence for Resource Extraction and Sustainable Arctic Communities (REXSAC, project no. 76938) with additional funding from Göran Gustafsson Foundation for field work. S.C. was supported by Russian Science Foundation project no. 18-17-00086.

Acknowledgments: We acknowledge Carl-Magnus Mörth (Dept. of Geological Sciences), Marcus Sundbom, and Pär Hjelmquist (Dept. of Environmental Science) at the laboratories at Stockholm University for analyzing water quality data. We also thank Camilla Winqvist for providing historical archive data.

Conflicts of Interest: The authors declare no conflict of interest.

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