Norwegian Waters:
Results from a Long-term Study of Metal Concentrations at a Test Center

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ABSTRACT

Regular and consecutive annular monitoring of metal concentrations in streams and standing waters at the Nammo Raufoss Test Center (NRTC) are summarized for a period close to 30 years. The sampling was done at a century-old shooting range which today has evolved into a complete test center. This substantial dataset provides understanding of the metal concentrations in standing waters, groundwater seepage and streams in relation to testing of ammunition and rocket motors. Results conclude that the mean annual metal concentrations in streams, with a few exceptions, are within the environmental quality standards, and with a trend towards overall lower concentrations in recent years. As part of a national monitoring program on metal-runoff from military firing ranges and demolition sites, annual mean concentrations of copper and lead at this test center were in the lower end among the investigated sites. The zinc concentration, in particular has been reduced after local countermeasures were taken. Including annular investigations of stream biota (macroinvertebrates) into the future monitoring scope may further strengthen the conclusion, that NRTC does not cause measurable negative effects on the downstream macroinvertebrate fauna.

1.0 INTRODUCTION

1.1 Nammo Raufoss Test Center in Context

Nammo is a producer of rocket motors and ammunition with several production facilities in Europe and in the US. The corporate office is located in Raufoss, Norway currently the largest site within the company. The corporate sites span a wide range of production facilities for energetic material, metal processing, assembly as well as R&D and additional functions. The Raufoss-site was established in 1896 and has since then experienced a continuous line of activities with technical advances and an expanding product portfolio. The company has a proximite test center, and the monitoring of metal concentrations in streams and waters at this this particular site represents the subject of this study. Nammo Raufoss Test Center (NRTC) currently covers an area of 4.000 da (4 km^2). This company-owned piece of land (Bradalsmyra) contains more than 20 shooting ranges and test facilities. The test facility was established in 1918 and has for more than a century expanded both in area, product range (Nammo Handbook, 2020), local infrastructure and measurement technology.

1.2 Novel Activities at Nammo Raufoss - Including Testing at NRTC in Recent Years

Nammo has atop of is publicly known product range lately broadened its set of activities and product portfolio considerably. The product range of solid rocket motors and ammunitions has added successful development of propulsion systems based on Hydrogen Peroxide (H_2O_2). This “green propellant” decomposes in an exothermal reaction resulting in hot gasses and water vapor. In Nammo-developed monopropellant, and hybrid technology systems the H_2O_2-based propulsion has shown its insensitivity to pyroshocks at NRTC (Krogstie, 2015) and passed technology maturation testing at NRTC. Initial tests at NRTC lay ground for successfully launching the sounding rocket “Nucleus”; reaching space (Faenza, 2019). The awareness on environmental impacts has increased in recent decades. Currently this test center is an established part of the ISO14001 certified local entity Nammo Raufoss AS.

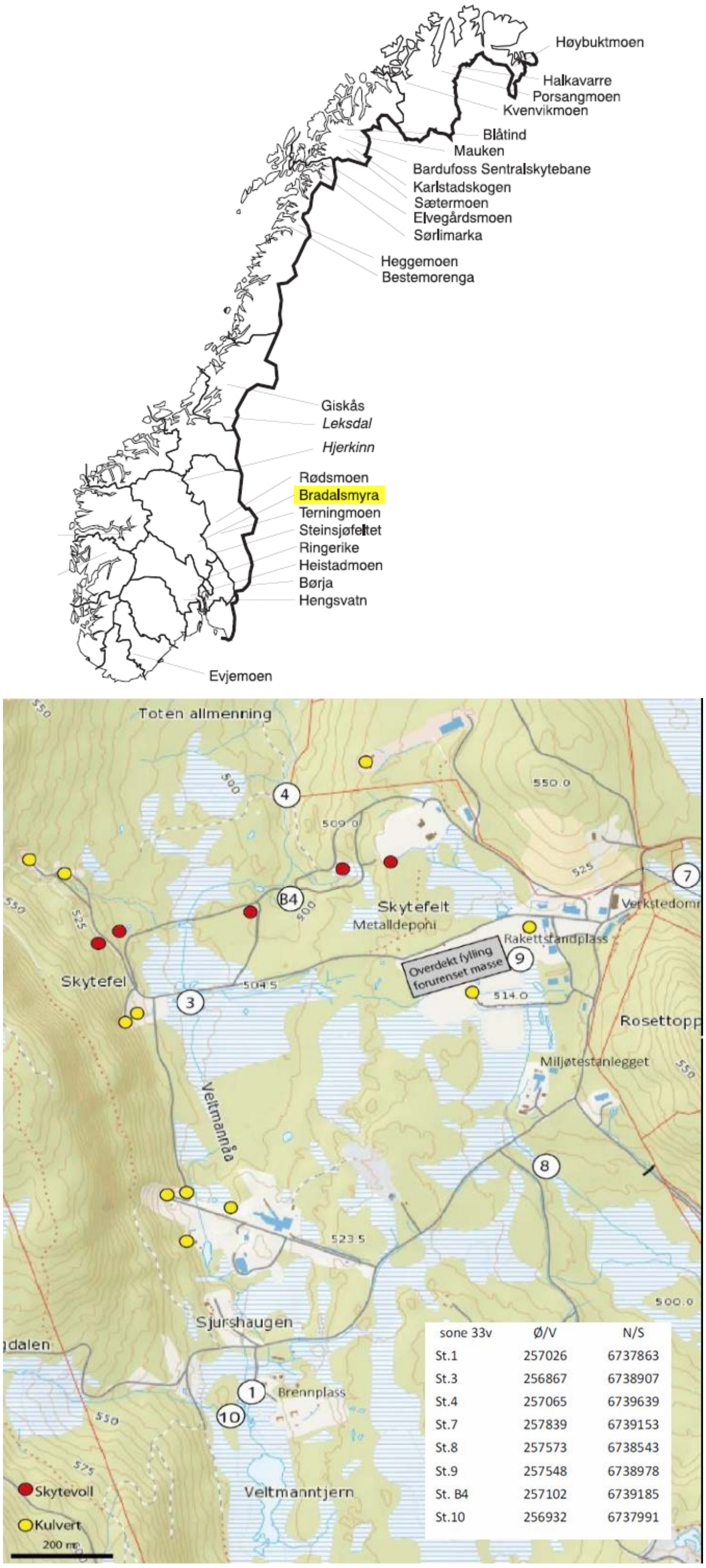
2.0 METHODOLOGY

2.1 Objective & Geography

The purpose of this evaluation of metal concentrations in streams, groundwater and standing waters is to assess whether the Test Center adds a significant environmental impact on streams draining the Test Center. Initially, monitoring at NRTC (aka. Bradalsmyra) was part of a larger national monitoring program on metal run-off from military training fields across Norway (Rognerud 2003) as shown in figure 1a. This national program was later divided into individual local monitoring programs.

NRTC (Bradalsmyra) is located from 500 to 700 m above sea level in a valley with steep hillsides towards the west, and lower hillsides with less elevation towards north, east as well as southwest (Figure 1b). The monitoring of metal concentrations in streams and waters in this area has been continuous since 1991. The sampling locations (stations – here marked “St.”) have been slightly adjusted throughout the years based on results from the monitoring program, as well as detection of historical metal deposits. Some of the adaptions are listed below:

- In 2004 5 sample stations were established east of B4 to monitor a recently identified metal deposit.
- These 5 stations were later replaced by station B4 after a modification of the shooting range.
- In 2006 sampling station, St.3, was established to monitor the run-off from a metal deposit.
- In 2011 St.9, was established to monitor metal-concentrations close to the metal deposit (above).
- The comparison of St.9 (local) vs. St.8 (run-off towards south) adds understanding on downstream dilution.
- In 2016 sampling station 10 (St.10) was established in order to investigate the geological contribution to background-levels.



sone 33v	Ø/V	N/S
St.1	257026	6737863
St.3	256867	6738907
St.4	257065	6739639
St.7	257839	6739153
St.8	257573	6738543
St.9	257548	6738978
St. B4	257102	6739185
St.10	256932	6737991

Figure 1 (a,b): Overview of the full scope of the study outlining the Test Center of Bradalsmyra / NRTC from Rognerud, 2003 (Fig.1a, left) Overview of the TestCenter and Water sampling locations (Økelsrud, 2019) (Fig. 1b, right).

2.2 Sample Stations & Methods

The area for the Test Center is mainly drained by the stream “Veltmannåa” to the north (St.4). In addition, the area is drained by two small streams, one to the east (St.7) and one to the south (St.8). Currently there are eight active monitoring stations where samples are taken monthly in the accessible time of year (typically April – November due to frozen conditions during winter). These stations are:

- St.1: Upstream in the main stream Veltmannåa starting from the lake “Veltmanntjern”.
- St.3: Veltmannåa in the center of the Test Center flowing towards north.
- St.4: Northern station of Veltmannåa as it flows out of the property.
- St.7: Eastern station where a minor stream flow out of the property.
- St.8: Southern point where a stream flow out of the property towards south.
- St.9: Small wetland/marsh area with periodically stagnant waters adjacent to a historical deposit.
- St.10: The stream representing the main influx to Veltmanntjern (the pond).

Water samples for determination of metal concentrations were collected in acid-washed plastic bottles, while the samples for pH and total organic carbon (TOC) / dissolved organic carbon (DOC) were collected in separate plastic bottles. The sampling, analysis of water chemical results and reporting were carried out by an independent approved institute for water research (Norwegian institute for water research, NIVA) and the water chemical results for each station are publicly available on the internet (Vannmiljø, 2020).

2.3 Variations in Monitoring Methods from Earlier Years to Present

During the first years of the monitoring program (1991-2005) detection limits of the analytical methods used (Atomic Absorption Spectroscopy) was close to water quality threshold values, which introduced some uncertainty in the assessment of environmental risk. Hence the preferred method at that time was the use of moss bioindicators such as *Fontinalis antipyretica* which has the ability to bioaccumulate metal concentrations with bioconcentration factors in the order log4-log4.3 (Rognerud, 2006). In the first years of the monitoring program of environmental metal concentrations at NRTC, effort was made to detect correlations between metal concentrations in moss growing on stones in streams and standing water with the water in which the moss grew (Figure 2). However, as mosses were never implemented in the national water quality guidelines with specific threshold values, the concentrations in water and moss was used in the early years to determine water quality. Later on only water sampling was used to classify the chemical and ecological status both from the early years and up to present day. As from 2006 the measurements in moss were left out completely from the monitoring program. An explanation for this is to be found in a variety of factors, some scientific (increased capacity of accumulating metals in moss at lower TOC), which also has led to the practice that water mosses never were implemented as a matrix for monitoring metals in the national water quality guidelines, which in later years (since 2008) follow the EU water frame directive (established in 2000). In this sense one can perceive the monitoring at NRTC as developing from a more “testing of different methods approach”, with some anchoring in guidelines and set threshold values, to a monitoring program strictly following the national water quality guidelines (see also Ch. 2.4 below).

Figure 2: Moss bioindicator *Fontinalis* antipyretica (left), physical sampling (middle) and reported correlations (right) between concentrations of Cu and Pb in moss and water in the early years of sampling program (Rognerud, 1994).

2.4 Adaption of Threshold Levels over the Years

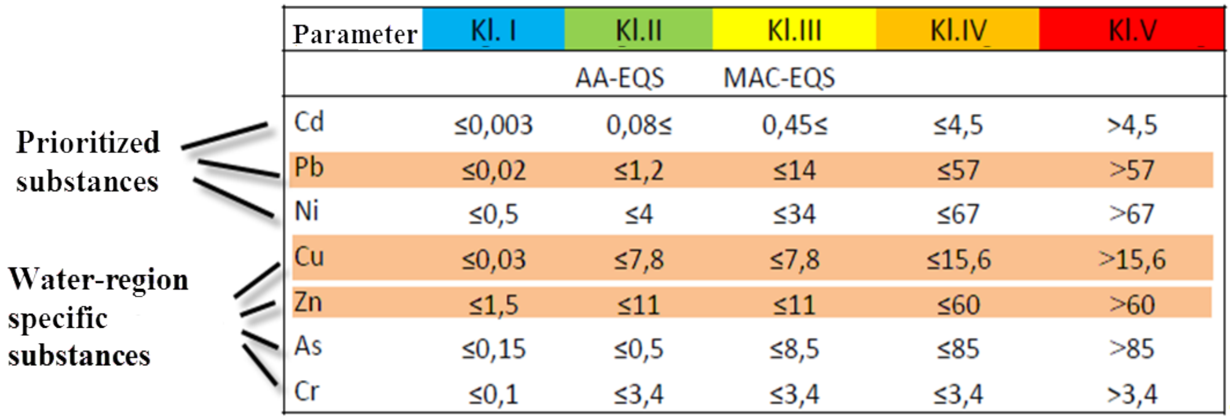
Starting from 1992 the Norwegian pollution agency developed a system dividing the water quality into different classes with related colour codes (Holtan & Rosland 1992). The quality classes (Table 1 – from Norwegian) included levels and colour codes; blue (good), green (less good), yellow (rather bad), red (bad), violet (very bad).

Table 1: Water quality classes according to (Holtan & Rosland 1992)

	Tilstandsklasse/ fargekode	God (I) blå	Mindre god (II) grønn	Nokså dårlig (III) gul	Dårlig (IV) rød	Meget dårlig (V) fiolett
Cu	Kobber (µg/l)	<2	2-5	5-15	15-50	>50
Pb	Bly (µg/l)	<1	1-3	3-5	5-10	>10
Zn	Sink (µg/l)	<10	10-30	30-60	60-110	>110

After a few years with various national water quality guidelines, internationally standardized methods and threshold levels were introduced in the (Directive 2013/39 EU) by using the threshold values for annual average concentrations (AA-EQS) and maximum annual concentrations (MAC-EQS). AA-EQS therefore represents the threshold value for the annual average of all samples from a given location (i.e. sampling station) to reach either good chemical (priority substances) or good ecological status (water-region specific substances). MAC-EQS is the threshold value for the maximum annular concentration, which then represents the threshold value for individual samples at the given location. Table 2 below is taken from Rognerud (2017). The references for classifying the results are divided into five classes (I-V) from EQS (Environmental Quality Standards) with background in AA-EQS and MAC-EQS. The classification of Cadmium is dependent on the concentration of lime (calcium) in the rivers, and this was accounted for in accordance to respective threshold limits (I: background, II: good, III: modest, IV: bad, V: very bad).

Table 2: Water quality classes (Vanndirektivet, 2013 / 2018)



Prioritized substances

Water-region specific substances

Parameter	Kl. I	Kl.II	Kl.III	Kl.IV	Kl.V
		AA-EQS	MAC-EQS		
Cd	≤0,003	0,08≤	0,45≤	≤4,5	>4,5
Pb	≤0,02	≤1,2	≤14	≤57	>57
Ni	≤0,5	≤4	≤34	≤67	>67
Cu	≤0,03	≤7,8	≤7,8	≤15,6	>15,6
Zn	≤1,5	≤11	≤11	≤60	>60
As	≤0,15	≤0,5	≤8,5	≤85	>85
Cr	≤0,1	≤3,4	≤3,4	≤3,4	>3,4

2.5 Attention to Additional Variables

Throughout the survey attention to additional details arose. Examples are listed below:

- Investigations on the influence of natural sources of metals (St.10) from 2016 (Rognerud, 2017).
- Measurements of TOC and pH to add understanding of local effects (Rognerud, 2017), (Rognerud, 2005b).
- Correction factors due to methodology shift away from moss sampling (Rognerud, 2007).
- Metal concentration levels relativized to local geology effects (Cambr.-Silurian minerals) (Økelsrud, 2019).
- Additional attention to benthic macroinvertebrates (Økelsrud, 2019).

Generally, the monitoring program has added novel aspects throughout the duration of the monitoring period in order to improve the understanding, and to deepen explanations up to local geology and in-stream water chemistry.

3.0 RESULTS AND DISCUSSION

An extensive series of reports has been published during the years of the monitoring program: (Kjellberg, 1992); (Rognerud, 1993) surveys NRTC only. Following publications (Rognerud, 1994), (Rognerud, 1995), (Rognerud, 1996), (Rognerud, 1998), (Rognerud, 2000), adds further military ranges of interest. The comprehensive 10-year summary (Rognerud, 2001) holds a rich lierature reference and several visual time-series. The further reports such as (Rognerud, 2002), (Rognerud, 2003), (Rognerud, 2004), (Rognerud, 2005b) hold several correlation-plots across the parameters in the data-sets. As from the first report focusing on NRTC only (Rognerud, 2005a), both a shift in methodology and closer focus on local effects were made. The 15-year national report (Rognerud, 2006) closes the national program. The most recent reports (Rognerud, 2007), (Rognerud, 2009), (Rognerud, 2011), (Rognerud, 2012) then solely focuses at NRTC, though with different timeframes such as the years 1991-2014 in (Rognerud, 2014), the years 1991-2017 in (Rognerud, 2017) , the timeframe 2004-2018 in (Økelsrud, 2019). The program continues in 2020.

3.1 Copper and Lead

Copper (Cu) and lead (Pb) was a focus on in the results early on in the monitoring program, as these metals typically occur in traditional ammunition (Figure 3). NRTC followed the national sampling program up until

Rognerud (2005b) and was from there on left out of the national program (Rognerud, 2006). Specific measurements continued at NRTC (Økelsrud, 2019). The singe-event high exposure of Pb at NRTC in 1997 was noticeable but challenging to trace back to the specific source.

As a comparison, typical values of other test ranges are listed in (Figure 3) below. These areas called "S" and "M" represents the ranges "Steinsjøfeltet" and "Mauken" respectivelty. For this comparison however the NRTC is clearly at the lower range. Concentrations for Pb were within what equals background levels at the NTRC for most years was, while Cu was within, what was then termed the threshold value for "less good". This threshold has since been changed and for Cu this equals "good ecological status today. The other ranges have more frequent measured concentrations, for some stations, well above the earlier threshold value for "less good" and the present EQS for both water-region specific substances (ecological status) and priority substances (chemical status).

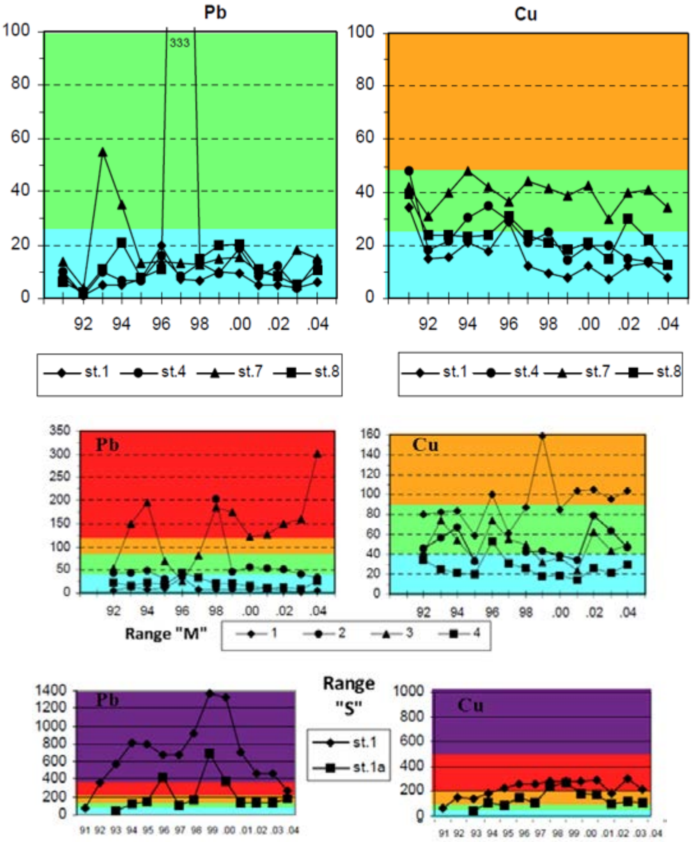
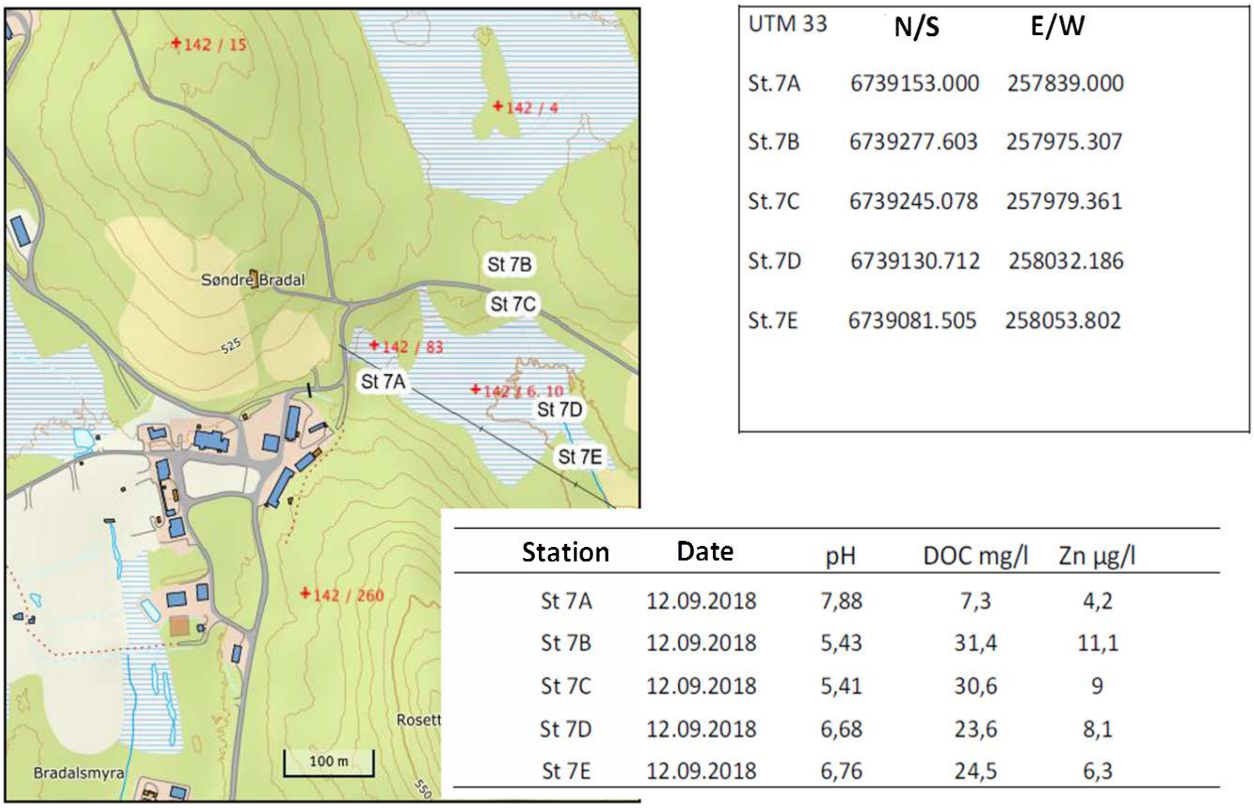


Figure 3: Concentrations (µg / g dry weight) of Cu and Pb in moss given as annual arithmetic avearge for the monitoring period, NRTC (left), Mauken (right top) and Steinsjøfeltet (right low). Colour codes relates to water quality classes in water and the thresholds are estimated from the relationship in moss and water concentrations.

3.2 Source Tracing of Zinc

Based on the reports from previous years the Norwegian Environment Agency (Miljødirektoratet) called for a source tracing of zinc (Zn) in the area nearby sampling station. St.7 (Rognerud, 2017). This investigation (Fig. 4) revealed that sampling station 7B had the highest measured concentration of Zn as well as the lowest pH-level. The results from the stations downstream from 7B, the stations 7C, 7D, and 7E are worth extra attention. They clearly illustrate how the levels of Zn decreases as well as the increasing pH. The results from these downstream stations indicate influence from both the upstream stations 7B and 7A. Station 7A, which is situated within NRTC had the lowest measured concentration in the source tracing study. It was concluded (by measuring pH, Zn, and DOC) that the contribution from the Test Center draining into the station 7A likely was below the natural contribution from outside the NRTC. This underlines that measured concentrations above the threshold to achieve "good ecological status", in particular for Zn, inside the Test Center may be explained by natural contribution by geological sources.



UTM 33	N/S	E/W
St.7A	6739153.000	257839.000
St.7B	6739277.603	257975.307
St.7C	6739245.078	257979.361
St.7D	6739130.712	258032.186
St.7E	6739081.505	258053.802

Station	Date	pH	DOC mg/l	Zn µg/l
St 7A	12.09.2018	7,88	7,3	4,2
St 7B	12.09.2018	5,43	31,4	11,1
St 7C	12.09.2018	5,41	30,6	9
St 7D	12.09.2018	6,68	23,6	8,1
St 7E	12.09.2018	6,76	24,5	6,3

Figure 4: Additional measurement points for origin tracing of zinc (Økelsrud et. al. 2019)

3.3 Other Influencing Effects

The results after 14 years of investigations adds a deeper understanding of the sources and reasons for variations in measured metal concentrations (Rognerud, 2005b, page 42). Typically, the main reasons are:

- Variations in local geology
- Variation in activity level from year to year
- Variation in activity level throughout the year
- Overall duration (years of active use) of the test range
- Different degree of mechanical interruptions / manipulation in the ground.

In general, the corrosion accelerates with lower pH. The pH-level at NRTC appears to be more close to pH 7 than many of the other test ranges in the study. This is beneficial for the metal concentration level at NRTC.

The reports from 2009 and 2010 add more details on the level of TOC (Total Organic Carbon) (Rognerud, 2009 & 2011). Metals are, in general, positively correlated with TOC, in particular antimony (Sb), mercury (Hg), bismuth (Bi), arsenic (As) and Pb form strong associations with organic matter (Rognerud and Fjeld, 2001). In other words, run off containing high concentrations of TOC will increase the influx of metals to streams as well as instream transport, as organic particles acts as vectors for metal ions. Metals forming complexes with organic matter becomes less bioavailable, and therefore less toxic to aquatic biota. This relationship however is to a large degree governed by pH, where pH when lowered will increase the ion exchange, meaning that the metal-ions dissociates from organic matter in change of hydrogen ions (Figure 5). Gundersen and Steinnes (2003) studying the influence of pH and TOC concentration on Cu, Zn, Cd, and Al speciation in mining polluted rivers, reported that less than 10% of Cu, Zn, and Cd were sorbed on particles or colloids in two rivers with average pH at 3.1 and 5.1, whereas 46%, 21%, and 21% of Cu, Zn, and Cd, respectively, occurred in sorbed form in six pH neutral rivers. The measured pH from sampling stations at NRTC ranges from around to 6 to around 8. This means that a considerable fraction of the measured Cu, Zn and Cd are complexed to organic matter. In general, pH, TOC and Ca influences on both mobility (transport) and bioavailability (hence toxicity) of metals in a complex manner. Analyses of results from Bradalsmyra in 2018 have shown that the correlation between DOC and TOC is close to 1, therefore from 2019 only DOC was reported.

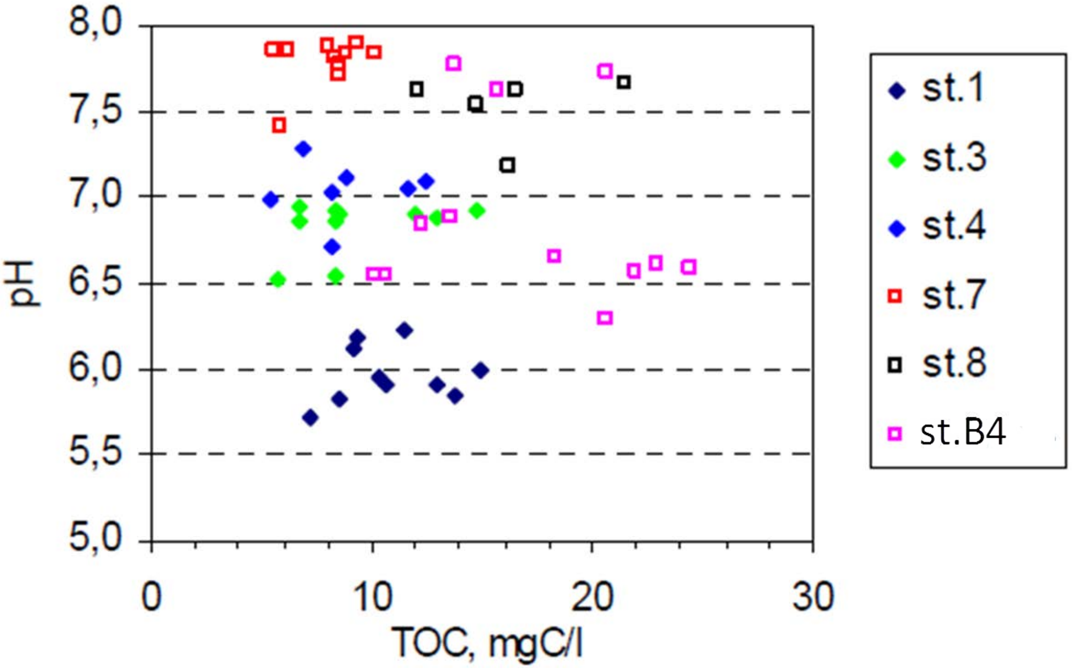


Figure 5: TOC vs. pH for NRTC in the years 2007-2010 (Rognerud, 2011)

3.4 Background Levels

Apparently, both the local geology and long-range atmospheric transport can add to the metal concentrations in soils and waters. This is also summarized after 11 years of monitoring by Rognerud (2002). A substantial fraction of the bedrock underlying the Test Center at Bradalsmyra consists of alum shales (Lutro and Nordgulen, 2004), that naturally contain increased levels of metals (Jeng and Bergseth 1992). Typically, soil and water adjacent to alum shales contain increased concentrations of Cu, Zn, Ni and Cd, as well as Pb, when compared to non-alum shale soils (Jeng, 1992). Erstad (2017) reported that Zn, Ni, Cu and Cd show a high mobility and aqueous concentrations when assessing the long- and short-term leaching potential of alum shale through an outdoor bulk- and a laboratory column experiment. Concentrations that far exceeded the Norwegian standards for drinking water and environmental quality were reported in that study. Pb on the hand had low mobility under similar pH regime. This may explain why Pb appears to be less prone to exceed the EQS than for example Zn in main streams draining the NRTC. The potential geological contribution has also been assessed when interpreting the measured metal concentrations at NRTC. However, the water quality guidelines do not differentiate between sources, and environmental status is strictly based on measured concentrations (according to guidelines). As discussed in Chapter 3.3, natural contribution may occasionally be higher than expected point sources. However, these background concentrations do not appear to be at the same level as the higher concentrations found adjacent to known point sources, such as at station 9 (St.9).

The other potential contributor to “background levels” of metals in soils and water may be long-range atmospheric transport. As metals are volatilised at high temperatures in processes such as fossil fuel combustion, for example Pb and Zn, are either redistributed onto smaller particles or emitted directly in the gas phase. These polluted aerosols may be transported over large distances in the atmosphere and deposited via precipitation far from emission sources (Steinnes, 2009). Surface sediment in lakes in the region, which in Bradalsmyra is situated, show increases of metal concentrations, particular for Pb and Cd, when compared to reference sediments (30-50 cm depth) from the preindustrial period (Rognerud and Fjeld, 2001). This signifies some contribution from long-range atmospheric deposited metals to the area.

3.5 Effects on benthic macroinvertebrates in streams

In 2018, for the first time in the monitoring programme at Bradalsmyra, an investigation on the benthic macroinvertebrate fauna (benthos) in the streams draining Bradalsmyra was conducted (Økelsrud and Rognerud, 2019). This was initiated in order to assess the overall diversity, and presence or absence of invertebrate groups sensitive to metal-pollution. Three of the stations in the regular water sample monitoring programme were sampled for benthos in late April. These three stations are identical with St. 1 and 4 in the Veltmannåa stream, and St. 8, downstream of St.9, adjacent the rocket firing-stand (Figure 2). In addition, one station was sampled further downstream of St.4 (~ 2,5 km downstream) and served as background station (due to dilution and sedimentation/absorption of metals). The results differed between stations, with sensitive Ephemeroptera’s (mayflies) only present at St.4 and at the downstream background station. It was concluded that St.8 did not have the appropriate morphology, size or substrate for a diverse benthic fauna. It was suggested that the lack of mayflies at St.1 could potentially be a result of run-off of heavy metals, or as a result of periods of drying out of the streams, and/or episodes of acidification. However, while mayflies were not present at St.1, but at St.4 (*Baetidae* spp.), the results indicated that the activities at the NRTC (including potential leaching from deposits) had not caused measurable negative effects on the macroinvertebrate fauna in streams draining Bradalsmyra during the autumn/winter 2018.

4.0 SUMMARIZING DISCUSSION & PROPOSED FURTHER WORK

The extensive dataset from the many years of monitoring at Bradalsmyra fully documents the historical development and the relative level of metal concentrations at NRTC compared to other military test ranges. The low levels of metal concentrations at NRTC can be explained by a series of factors:

1. Type of activity.
2. Type of ammunition and products tested.
3. The use of the surrounding land area close to the measurement points.
4. Geology and other local factors.

On an overall level we can state that these are typical pros for the NRTC in explaining lower levels of exposure compared to several other test ranges. Firstly, the activity is, despite of its diversity, rather well defined. Testing occurs at up to 20 distinct ranges each shooting or operating towards well defined caverns. The exposure is hence very local and the exposure area can to a large extent be fully identified. Secondly the company product contains a very little amount of Pb. The typical products are designed for harder targets hence holding more hardened steel, and less the traditional lead-type ammunition. Thirdly the use of the land area is rather well defined. Transport is conducted on well-defined roads and passages and overland transport of heavy machines hardly occur. Fourthly the the geology appears to contribute to somewhat increased background concentrations for some of the metals, especially for Zn. Mainly stemming from the Cambrian-Silurian period. Fifthly the company has taken efforts to replace cavern masses including metal residuals.

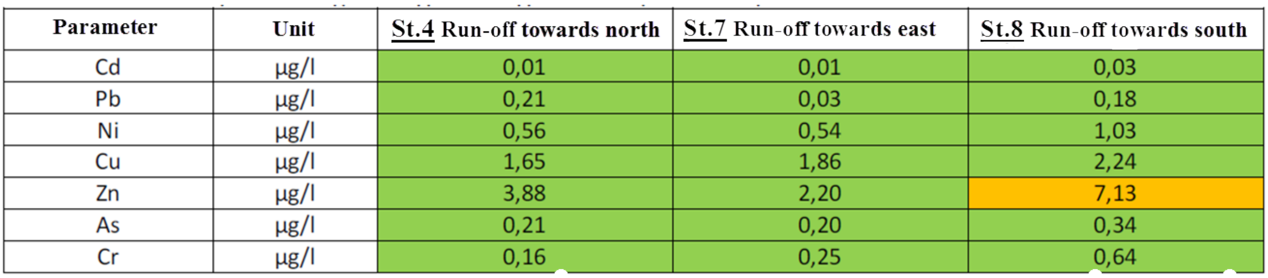
Finally the development focus for the later years adds increased emphasis on “green propulsion” typically using hydrogen peroxide as propellant.

Further work may include more in-depth studies of historical trends and further mapping of how local sources of metal content adds up with the level of background contribution from soil, rocks and other sources in the surrounding area. Future monitoring may also include studies of stream biota in out-flux areas or around other specific areas of interest. As the long-term monitoring up to date has provided a sustainable data-set it is of interest to continue the monitoring already in place in order to continue these data-series. It is also of pertinent interest to continue ensuring non-harmful metal concentrations in streams draining the test centre.

5.0 CONCLUSION

Regular monitoring of metal concentrations have been done for close to 30 years at Nammo Raufoss Test Center has provided a rich dataset for understanding the levels of metal concentrations in standing waters, groundwater seepage and streams. Results show that the average annual values for the metal concentrations are within the EQS with a trend towards reduced levels (Table 3).

Table 3: MAC-EQS Status at NRTC in 2018 after close to 30 years of measurements (Økelsrud, 2019)



Parameter	Unit	St.4 Run-off towards north	St.7 Run-off towards east	St.8 Run-off towards south
Cd	µg/l	0,01	0,01	0,03
Pb	µg/l	0,21	0,03	0,18
Ni	µg/l	0,56	0,54	1,03
Cu	µg/l	1,65	1,86	2,24
Zn	µg/l	3,88	2,20	7,13
As	µg/l	0,21	0,20	0,34
Cr	µg/l	0,16	0,25	0,64

Only one single sample resulted in a concentration above the EQS for good ecological status in streams draining the Test Center in 2018. When performing a source tracing study of the Zn-level in the area close to station 7 (inside the NRTC), the measured concentration inside NRTC were lower than two stations located in a stream flowing from a hillside and into the NRTC. The results indicate that the natural background concentrations can be elevated as a result of input from gelogical sources in the area.

The overall conclusion is hence that the Test Center has comparable, or lower contribution of metal in run-off to surrounding waters compared to other military test ranges in Norway. The Nammo Raufoss test activities, even after more than 100 years of concecutive test-activity, does not add an unacceptable contribution to measured metal concentrations in the downstream areas, nor harmfull concentrations of metals to biota downstream.

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