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Cover photo: The aluminium raft equipped with special magnetic device which was used for identification and exact positioning of dumped ammunition in lakes (refer page 5 in present report) .

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Environmental Risk Assessment of Dumped Ammunition in Natural Waters in Sweden - a Summary

Abstract (not more than 200 words)

After World War II left ammunition was dumped, and in Sweden it was mainly discarded in lakes as well as at sea. By order of the Swedish Armed Forces, the Swedish Defence Research Agency (FOI) has conducted an environmental risk assessment regarding ammunition dumped in lakes and at sea.

In order to enable such an assessment, the following steps were involved; literature studies, field work, chemical analyses and laboratory tests (simulation of TNT release), with respect to retention of the ammunition to lake and sea sediment, respectively. Tests have also been carried out in lakes and at sea where the sensitivity to sediment and water polluted with ammunition in organisms living on the seafloor was analyzed. From an environmental point of view, the risk of leakage of TNT (trinitrotoluene) and its degradation products as well as heavy metals from the dumped ammunition is of paramount importance.

The present report summarizes and discusses the most important findings from investigations by FOI on the impact of dumped ammunition in natural waters.

Keywords

TNT (trotyl), sediment, retention, toxicity tests, seafloor fauna, risk assessment

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Rapportens titel (i översättning)

Miljöriskbedömning av dumpad ammunition i sjö och hav - en sammanställning

Sammanfattning (högst 200 ord)

Efter andra världskriget dumpades ammunition som inte längre behövdes och i Sverige skedde dumpningen främst i insjöar men också i havsområden. På uppdrag av Försvarsmakten har FOI genomfört en miljöriskbedömning av dumpad ammunition i insjöar och hav.

För att kunna göra denna bedömning har litteraturstudier, fältarbeten, kemiska analyser och laboratorieförsök (simulering av TNT-utsläpp) med avseende på sprängämnens fastläggning till sjö- respektive havssediment genomförts. Även försök för att utreda bottenlevande djurs känslighet för sprängämnesförorenat sediment och vatten, gällande både sjö och havsmiljö, har utförts. Ur miljösynpunkt är det främst risken för läckage av TNT (trinitrotoluen) och dess nedbrytningsprodukter samt tungmetaller från den dumpade ammunitionen som är av betydelse.

Föreliggande rapport sammanfattar och diskuterar de mest framträdande resultaten från FOIs undersökningar om miljöeffekter av dumpad ammunition i sjö och hav.

Nyckelord

TNT (trotyl), sediment, inbindning, giftighetstester, bottenfauna, riskbedömning

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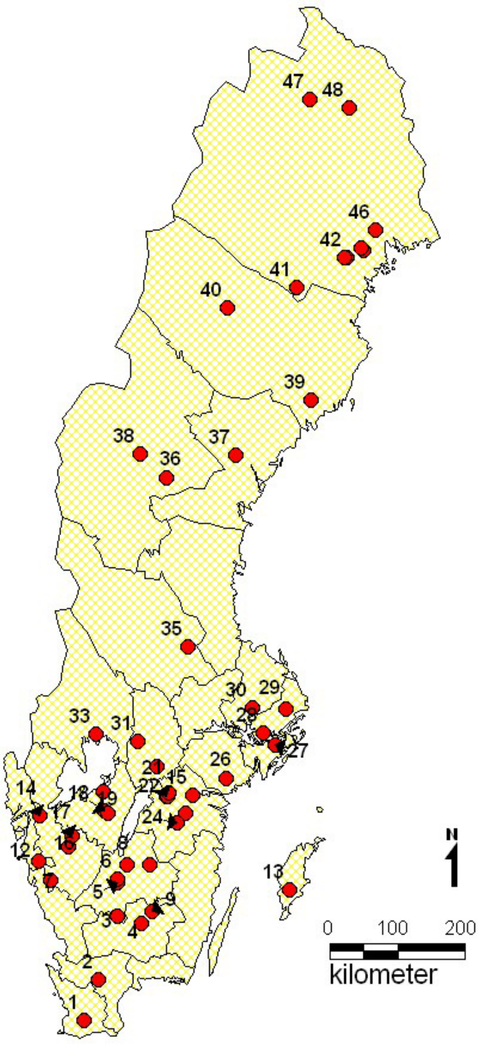
Background

There are 106 mapped grounds in Sweden where ammunition was discarded in the 1940s' to the 1960s'. The method of discarding substantial amounts of ammunition, unreliable and out-of-date, was rapid and cheap and along with some mines and locations on land the sea was the main dumping ground.

In the context of dumped ammunition the environment is supposedly affected by wrappings as well as by ammunition casings and explosives (Dave, 1997).

The amount of ecologically harmful substances in wrappings and casings is considered to be negligible (Sjöström et al., 1999). In order for the explosives to leak to the environment the casings must have rusted. Factors generally expediting the corrosion attacks on metals in the water environment are; higher temperature, salt content, oxygen content and/or lower pH value. The constitution of the ammunition and the amount dumped is defined by the number of hazardous substances (e.g. heavy metals) brought along by the dumping. Some of the organic substances and metals present in ammunition can in free form cause chronic and acute damage to nature (Berglind & Liljedahl, 1998). Explosives are generally grouped into initiators (for example lead azide), propellants (gun powder), explosives (e.g. TNT) and pyrotechnical sets (e.g. smoke sets).



A map of Sweden showing lakes containing dumped ammunition. The numbers indicate lakes selected by the Swedish Armed Forces (from a total of 65 lakes) for more detailed investigations. Refer Table 1 for further information and basic data. Note that ammunition also has been dumped in a handful abandoned mineshafts as well as at sea along the eastern and western coastline (not presented here).

Table 1. General information on the lakes with dumped ammunition presented in Figure 1. Data from Larsson (1999) for lake numbers 1-41, and from Sjöström et al (1999) for numbers 42-48.

Number	Lake	Coord. Y	Coord. X	Lake area (km^2)	Catchment area (km^2)	Average depth (m)	Turn over time (month)	pH	Cond (s)	Total alkalinity (ekv/l)	Temp (C^0)	Amm kg dump	Pb+Hg kg dump	Cu kg dump
1	Bökesjön	616320	135060	0,0	0,4	2,2	8	6,7	64,9	80	10,5	200	0,5	3000
2	Finjasjön	622380	137085	10,6	243,3	4,5	8	7,2	195,0	952	9,0	200	0,5	2000
3	Flåren	631800	139950	35,5	997,9	5,7	8	6,9	95,3	264	11,8	5	0,01	200
4	Helgasjön	630764	143570	49,5	626,1	5,1	16	6,7	92,7	30	12,7	55	0,07	1510
5	Östersjön	636790	139890	0,0	17,7	1,1	<1	5,3	43,3	40	3,2	10	0,01	1
6	Bohultasjön	637280	139970	0,1	1,1	2,8	18	5,5	55,0	136	6,3	5	0,01	10
7	Boglösasjön	637220	130020	0,1	1,0	2,7	40	5,7	50,7	180	6,8	5	0,02	10
8	Tenhultasjön	639500	141350	3,0	32,8	5,0	27	6,6	112,6	472	7,3	105	0,12	1000
9	Hunsnäsen	632500	145120	0,8	79,8	2,4	1	6,7	147,0	728	8,7	1	0,01	1
10	Soåsasjön	639540	144750	0,3	1,1	3,8	32	6,6	124,1	592	9,7	5	0,02	10
11	Kokhusgölen	639450	144800	0,0	0,4	2,7	7	6,3	68,3	384	9,3	5	0,02	5000
12	Landvettersjön	640100	128300	2,3	184,6	3,9	2	6,3	117,3	200	6,2	50	0,01	1000
13	Rammaträsk	646740	128440	10,1	78,9	5,3	26	6,4	86,0	104	4,6	1	0,01	200
14	Öresjön	642200	132600	12,0	179,5	4,5	12	6,0	101,1	212	6,9	20	0,02	1000
15	Glan	643780	133180	0,0	0,2	1,1	1	5,2	43,0	32	9,0	1	0,01	1
16	Sjön Säven	647570	137970	0,0	0,5	1,7	1	6,3	360,0	400	7,8	5	0,02	10
17	Trollsjön	647080	138580	0,0	0,3	2,2	11	5,1	17,9	16	10,9	5	0,02	10
18	Skarsjön	647130	138550	0,0	0,1	1,2	1	6,0	59,2	248	9,7	5	0,03	300
19	Brattesjön	649720	147290	0,0	0,4	1,8	2	4,6	35,2	0	10,2	5	0,01	1
20	Lillsjön	649620	147420	0,6	5,1	3,3	14	5,9	55,7	152	12,4	1	0,01	1
21	Bogölen	647213	150153	2,0	47,8	3,0	5	7,6	167,0	1096	12,0	1	0,01	1
22	Hjässasjön	645680	148830	0,0	2,0	1,9	1	6,4	94,0	352	14,9	500	1	4000
23	Svinstadsjön	650200	147650	0,1	2,2	1,9	4	4,8	38,1	0	12,4	1	0,01	10
24	Lumpgölen	652230	156180	0,5	3,1	3,5	18	7,0	188,7	1320	11,8	5	0,01	300
25	Opplången	657350	163480	0,8	6,0	3,4	17	7,9	483,0	1896	10,7	5	0,03	2000
26	Eglasjön	659160	161720	0,7	15,3	2,4	4	7,3	371,0	2616	11,9	2	0,02	1500
27	Ältasjön	657750	143000	1,6	59,8	3,4	4	6,0	39,6	232	15,8	1	0,01	10
28	Säbysjön	654140	145820	13,2	125,5	4,6	18	7,7	96,6	472	18,3	200	0,5	10000
29	Limmaren	658830	136840	0,1	0,2	4,5	11	5,7	60,0	440	16,6	5	0,02	10
30	Ekoln	650360	137900	18,5	291,8	6,7	16	6,3	59,1	216	15,0	5	0,01	400
31	Kärmen	671930	150450	0,2	7,2	3,2	4	6,0	41,8	232	13,7	2000	0,03	15000
32	Tisaren	696980	147220	0,1	3,5	3,1	5	6,5	115,9	816	12,8	1	7	35000
33	Sandbäckstjärnen	700380	157580	0,0	0,8	2,0	1	6,1	155,6	472	12,2	5	0,01	1500
34	Gapern	725380	166610	0,1	0,3	2,4	32	6,2	11,9	120	20,0	1	0,5	3000
35	Fisklösen	729870	174240	0,3	2,9	4,0	16	6,2	66,6	416	19,3	100	0,02	10
36	Skurutjärnen	729780	173870	0,0	0,2	2,1	8	7,1	58,9	552	20,2	5	0,01	100
37	Tjärnen P. 414	730880	176700	0,0	2,4	2,4	1	6,0	29,1	264	21,7	10	0,2	2000
38	Storsjön	731220	176390	0,0	0,2	2,0	6	6,0	20,2	208	20,9	5	0,02	5000
39	Pengsjön	733960	178480	0,0	0,0	1,7	13	6,4	54,9	464	17,9	1	0,01	1
40	Stensele tjärn	753350	168700	0,1	7,7	3,2	1	8,6	352,0	1088	15,7	200	1	6000
41	Arvidsjaur Lomtjärn	752050	174550	0,1	0,8	2,7	10	4,9	42,7	312	14,7	1	0,01	100
42	Älvsbyn Krokträsk	1742400	7298700	0,3	3,5			Not analysed					0,01	500
43	Älvsbyn Stora Kamträsk	1738700	7297800	0,0	1,0			Not analysed				No/unreliable data		
44	Boden Kallasjön	1767000	7308800	0,0	0,1			Not analysed					0,2	2000
45	Boden Fisktjärn	1763900	7312200	0,0				Not analysed				No/unreliable data		
46	Niemisel Skogstjärn	1784800	7339600	0,0	0,5			Not analysed					0,004	100
47	Ala Lombolo	1687000	7533500	0,3				Not analysed					0,2	1000
48	Sjön Posiojärvi	1745500	7520500	0,1	0,9			Not analysed					0,01	100

TNT (2,4,6 – trinitrotoluene) and its degradation products in the natural environment

The explosives TNT (trinitrotoluene or trotyl) and RDX (hexahydrotrinitrotriazine) are decidedly the most ecologically harmful organic compounds present in the dumped ammunition (Berglind & Liljedahl, 1998). Some fuses can contain RDX but to a small extent. TNT is the main ingredient present in military ammunition dumped in Sweden.

In nature TNT transforms under aerobic and anaerobic conditions. In dry and well aired soils TNT remains constant for a long time. Reports from Weldon Spring, US, suggest that TNT is not broken down (Bradley & Chapelle, 1995) at concentrations of over 100 mmol TNT/kg soil (22 mg/kg) indicating that high concentrations through its toxic impact puts an end to the microbiological activity needed in order for the TNT to transform (mineralise) into carbon dioxide and water (Bradley & Chapelle, 1995). On the other hand, low concentrations of TNT dissolved in water are transformed into other products. TNT and its degradation products are damaging to humans (Yinon, 1990), mutations (Spanggord et al., 1982, Won et al., 1976), poisonous to plants (Plazzo & Legget, 1986), fish (Smock et al., 1976, Degani, 1943) larvae (Won et al., 1976), algae (Won et al., 1976, Smock et al., 1976), fungi (Bumpus et al., 1994) and microorganisms (Klausmejer et al., 1973, Won et al., 1976).

TNT – degradation in an oxygen-rich (aerobic) environment (Sjöström et al., 1999)

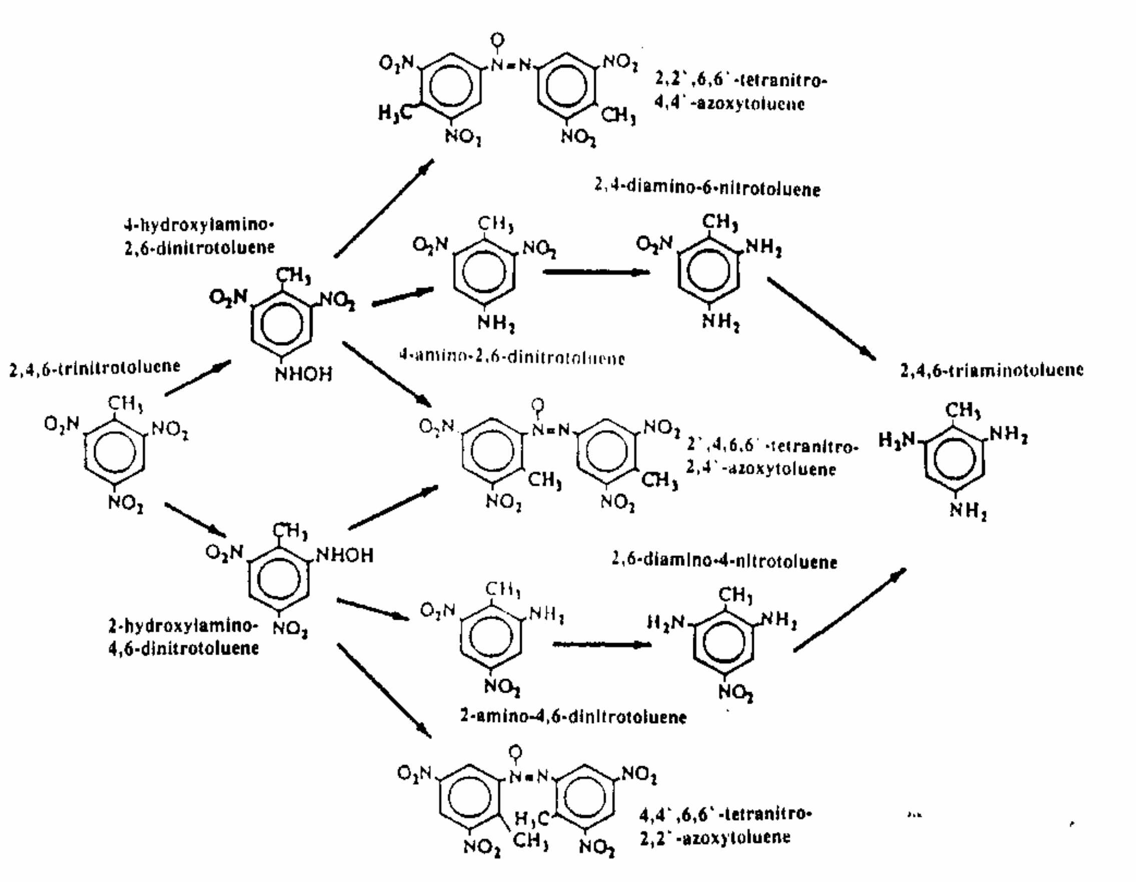
When TNT degrades in presence of oxygen the below degradation products are produced:
- Aminodinitrotoluenes (4-ADNT)
- Diaminonitrotoluenes (2,4-DANT)
- Tetranitroazoxytoluenes (more stable and in some cases more poisonous than TNT)

The degradation products 4-ADNT and 2,4-DANT can react by themselves in presence of oxygen as well as producing various compounds (tetranitroazoxytoluenes). Tetranitroazoxytoluenes are more long-lasting in the natural environment than the original substance TNT, in some cases even more poisonous.

TNT – degradation in oxygen-deficient environments (anaerobic) (Sjöström et al., 1999)

Degradation of TNT in oxygen-deficient environments produces less poisonous degradation products than what is the case in oxygen-rich environments but the former process seldom persists. In normal cases, a mixture of various residual products following a degradation of TNT is therefore probably produced in an oxygen-rich as well as in an oxygen-deficient environment.

In many cases, these degradation products are bound to sediment or soil (Haderlein et al., 1996). The degree of retention varies depending on residual product and type of soil. The binding process in soil as well as in sediment seems to be reinforced, and what is more, to be depending on microbiological activity (Bruns-Nagel et al., 1994, Held et al., 1997).



The figure illustrates the biological degradation pathway of TNT. The reactions forming multi-amino degradation products are favoured in anaerobic environments. Figure in Sjöström et al. (1999).

Environmental risk assessment of ammunition dumped in lakes

By order of the Swedish Armed Forces, and in cooperation with the University of Uppsala, FOI (former FOA), the Division of NBC Defence, has conducted an environmental risk assessment of ammunition dumped in Swedish lakes (Sjöström et al., 1999).

Investigated lakes

With the help of data such as dumped amount, pH value, nutrition status, sensitivity to acidification, dumping depth, sea area and drainage basin, as well as by using statistical data analysis (PCA-analysis), four lakes of different characters were picked out from about seventy in which ammunition has been dumped.

The first object is a well-documented nutritious plains lake, Limmaren, close to the town of Norrtälje, a representative of the group larger lakes, having well buffered water (i.e. having a high pH value and high alkalinity) and as such offering recreation of great value thanks to its location. The lake contains a large quantity of dumped explosives (i.a. 500 kg of explosives) and metals (lead, mercury and copper, especially) as well as being recipient of substantial amounts of industrial pollutants.

The second object is a poorly documented small woodland lake, here called Stensele lake, situated close to the inland town of Storuman, in the County of Västerbotten. It contains substantial amounts of dumped ammunition (i.e. 1000 kg of explosives) and metals (lead, mercury and copper especially). The woodland lake represents small forest lakes with a probable high organic sedimentation, i.e. fallout of dead organic material producing a sediment layer on the lakefloor.

The third object, Pengsjön, representing lakes of medium size, is a well documented, now limed, not too nutritious plains lake situated outside the city of Umeå and containing a relatively substantial amount of dumped ammunition (i.e. 100 kg explosives) and metals (lead, mercury and copper).

The forth and last object, Lomtjärn, situated at Bofors firing range in Karlskoga, is a poorly documented swampy woodland lake containing a substantial amount of dumped ammunition (i.e. 2000 kg explosives) and copper (2000 kg). It is small (about 1 hectare), acid (low pH value) and the water is coloured by humus substances from the bog (i.e. high colour value). Reportedly, the magnitude and character of the dumping (even that resulting from activities at Bofors) have resulted in the woodland lake having “exploded” due to self-ignition.

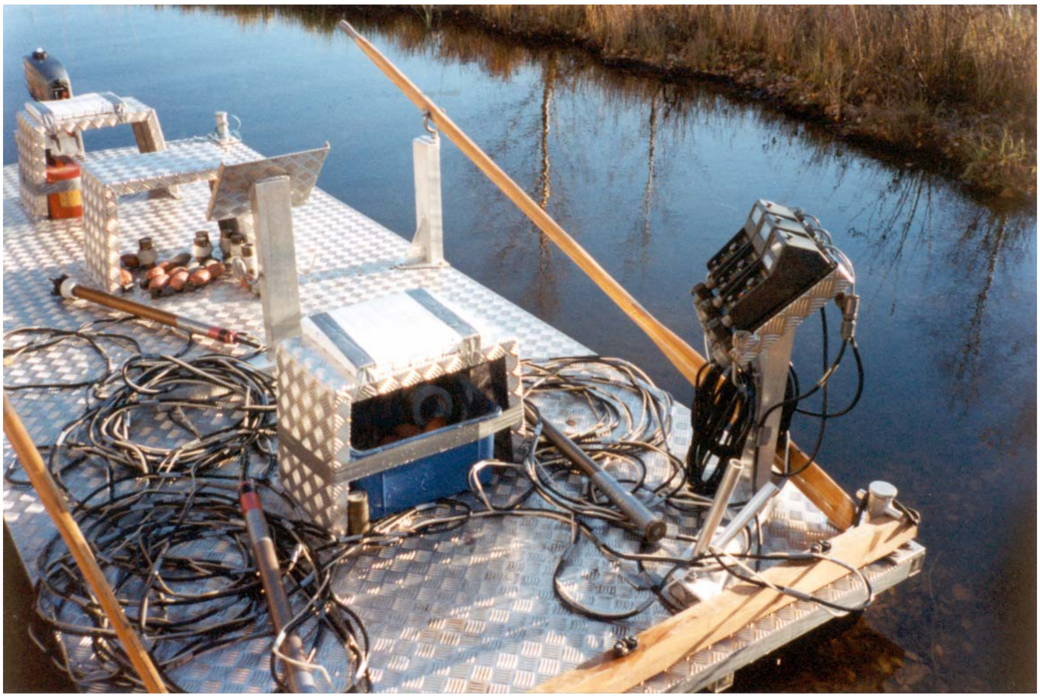


Photo: Jan Sjöström, FOI.

A special magnetic device mounted on an aluminium “raft” was used in the identification and exact positioning of the dumped ammunition. This complicated and time consuming work was performed by FFK, a special branch of the Swedish Defence Materiel Administration (FMV).The information had to be obtained in order to collect relevant sediment samples in the four selected lakes.

Environmental impact of dumped ammunition in lakes

The lakes were investigated with respect to water chemistry, among other things pH value, nutrition status (conductivity), acid sensitivity (alkalinity) and rigidity, in order to establish a database for further analysis (Larsson, 1999). A water sample taken 0.5 m above the lake floor at the dumping grounds was analysed with respect to pH value, conductivity, temperature and eight metals in each lake. Sediment samples down to a depth of one metre were also taken.

In the water and sediment samples taken (three in respective lake), neither TNT nor it's by- and degradation products were found (within the sensitivity range of the analysing instrument). Also, the sediment as well as the water from the lake floors were analysed as regards metals, namely arsenic, cadmium, copper, mercury, nickel, lead and zinc. Regarding the contents of these metals, the results documented by the Swedish Nature Protection Agency (NV, 1990) have been used as guidelines for affirming a high or low content. The scale is divided into five stages, *very low, low, moderately (medium) high, high* and *very high* concentration.

Concentration of metals in water samples

In Pengsjön, the concentration was generally *low* or *very low* (Table 2). In the Stensele woodland lake a *moderately high* concentration of zinc was present at all testing points. The lake Limmaren showed the highest concentration of metals of all investigated lakes, having a *high* to *very high* concentration of copper, arsenic and zinc throughout. As to the other metals analysed, the concentration in Limmaren was *low* to *very low.* In Lomtjärn there were mainly *low* concentrations of metals except for some samples containing a *moderately high* concentration of zinc, lead, cadmium and copper. When it comes to mercury no guide lines for assessments has been produced regarding water (NV, 1990 & 1999). In general the concentration of mercury was found in such a low concentration rendering analysis almost impossible (<0.002mg/l or <2 µg/l). However, in Lomtjärn the concentration of mercury in the water samples was considerably higher (ca one ten-fold).

Table 2. Guideline values for metal contents in water (µg/l) according to NV (1990). Note that some of the values in this table have been slightly revised in NV (1999) published shortly after Sjöström et al. (1999).

Term	Copper	Zink	Cadmium	Lead	Chromium	Nickel	Arsenic
Very low	≤0,3	≤1	≤0,01	≤0,2	≤0,4	≤1	≤0,2
Low	0,3-1,0	1-5	0,01-0,05	0,2-1,0	0,4-2,0	1-5	0,2-1,0
Moderately high	1-2	5-15	0,05-0,1	1-2	2-5	5-10	1-2
High	2-5	15-75	0,1-0,3	2-5	5-20	10-50	2-10
Very high	>5	>75	>0,3	>5	>20	>50	>10

Concentration of metals in sediment samples

In Pengsjön, a moderately *high* concentration of chromium, copper and nickel was found (Table 3). At two testing points deeper down in the sediment a *moderately high* concentration of arsenic was also found. In the Stensele woodland lake a *moderately high* concentration of copper and cadmium was found. The concentration of these substances increased with sediment depth and a *high* concentration of copper was found in two samples. Furthermore, a *moderately high* concentration of arsenic was observed on two deeper levels at one testing point. Moreover, the concentration of metal levels in Stensele lake was *low* to *very low.* Limmaren had a *moderately high* concentration of copper, chromium and nickel and at one testing point also a *moderately high* concentration of lead. No difference in concentration depending on depth was observed. The remaining concentration of heavy metal levels in Limmaren was *low* to *very low.* Lomtjärn had a *low* concentration of metals except for mercury. At one testing point, the concentration of mercury in the surface sediment of Lomtjärn very much exceeded the *very high*-limit. Observe that this testing point was not considered to be near the site where the Armed Forces dumped their ammunitions.

Table 3. Guideline values for metal contents in sediment (mg/kg TS; dry substance) according to NV (1990). Note that some of the values in this table have been slightly revised in NV (1999) published shortly after Sjöström et al. (1999).

Term	Copper	Zink	Cadmium	Lead	Chromium	Nickel	Arsenic
Very low	≤10	≤70	≤0,2	≤5	≤10	≤10	≤5
Low	10-25	70-175	0,2-0,7	5-30	10-25	10-30	5-15
Moderately high	25-50	175-300	0,7-2,0	30-100	25-75	30-75	15-75
High	50-150	300-1000	2-5	100-400	75-300	75-300	75-250
Very high	>150	>1000	>5	>400	>300	>300	>250

Seafloor fauna samples (small organisms living in the lake and on the seafloor)

The seafloor fauna was investigated in samples near the dumping ground and in two samples along a line away from it. Ten samples of surface sediment were taken (using a Swedish sampling device, *Ekmanhämtare*) per sampling site having small organisms larger than 0.5 mm. Using the seafloor fauna samples as basis, the species were determined, in addition to biomass (gram animal/m^2) and abundance (number/m^2).

The lake floor fauna in Pengsjön was fairly species-rich. The number of small organisms and their weight per surface unit was lower than expected when considering the surrounding farm land (usually leaking nutritious substances thus indirectly providing plenty of foodstuffs). The three testing points were very similar, the dumping ground did not differ (not statistically significant) from the other two test sites.

The environment at the Stensele lake was very different from that at Pengsjön. Due to the fact that the woodland lake was small and shallow, the environment was rich in species and individuals as demonstrated by the samples taken. The environmentally threatened (red listed) shell *Valvata macrostoma* (Ehnström et al., 1993) made a notable contribution to all testing points. The number of small organisms (abundance) and their weight per surface unit (biomass) at the testing point outside the dumping ground, was considerably lower (statistically significant) compared to those in the other sites possibly due to a higher content of organic material in the surface sediment.

The lake floor fauna in Limmaren was very species-poor at the particular depth. On the other hand, the number of small organisms (abundance) and their weight per surface unit (biomass) were relatively high. The three testing points were very similar, the dumping ground did not differ (not statistically significant) from the two additional testing sites.

The samples taken in Lomtjärn demonstrated a small amount of species, the shallow testing depth taken into account. The number of small organisms (abundance) per surface unit was very low as was the biomass. The probable dumping ground deviated (statistically significant) as regards the number of species per surface unit when compared to one of the two other testing points.

Simulation tests

The purpose of the simulation tests performed in a controlled laboratory environment was to observe in what way TNT separates between water and sediment and also how hard it binds to the sediment. Sediment and water from each of the four lakes were distributed in glass flasks. A known amount of TNT labelled with a radioactive nuclide (^{14}C) was then dissolved in the lake water, the radioactivity thus facilitating the analyses of TNT. The glass flasks were percolated with nitrogen, air and finally again with nitrogen (20 ml/min) for a period of 53 days at 12°C. The nitrogen provided an oxygen-free environment thus mimic the environment of the natural oxygen-free water in summer and winter (due to stagnation). The percolation of air simulates the oxygen-rich environment produced in the lake floor water in spring and autumn (spring/autumn circulation). The flasks were then left for an additional period of 53 days during which no percolation occurred. When compared to the natural processes the course of events was accelerated, among other things via a relatively high temperature. In the course of the test, the water in the flasks was continuously analysed to examine the concentration of TNT. All output air was analysed to see if TNT was transformed into carbon dioxide and water, i.e. was mineralised. The water phase was also continuously investigated regarding different chemical properties, e.g. pH value, conductivity, as was the biological activity in terms of the number of microorganisms.

In the next phase a leaching test was performed in order to see how tight the produced TNT degradation products (metabolites) were bound to the sediment. In such a sequential extraction gradually stronger solvents were added; from water (the weakest) via acetonitrile (stronger), hydrochloric acid (even stronger) to potassium hydroxide (the strongest). The amount of liberated TNT compounds was analysed following every attempt to release the TNT metabolites from the sediment.

Photo: Ann-Christine Andersson, FOI.

The simulation of TNT degradation and retention into sediment was performed using a system of flasks and bottles in a special climate chamber at FOI, the Division of NBC Defence.

Toxicity test

Part of the lake water from the simulation test, taken out after 53 days, was used. The survival of the water flea (*Daphnia magna*), a small freely swimming fresh water crayfish, having been in this lake water for 24 hours and 48 hours respectively, was examined. Lake water was not applied in the tests using control groups. The water fleas were assumed dead if they did not begin to swim when the water was stirred.

Additionally, tests were also performed to see whether heavy metals affect the toxicity of TNT (synergy effects). Such a test was performed on the water flea (*Ceriodaphnia dubia*), the aim being that of examining what effect water type (in terms of different salts) had on water fleas at the same time being subjected to lead, copper, zinc and TNT.

Results

The simulation tests indicate that the degree of binding of TNT to the sediments gradually increased in the course of the test (exponentially). On the whole, the results came out the same in all samples into which TNT had been added. Different chemical properties such as pH value or conductivity in the lake water did not affect the amount of TNT that bound to the lake sediments under the investigated conditions.

After barely 20 days the concentration of TNT in the water had divided into half and after 50 days 90 % of all TNT had disappeared from the water phase in all samples except for that from Pengsjön where more than 80 % had bound to the sediment. No carbon dioxide containing ^{14}C could be found in the output air of the flasks thus indicating that TNT had not transformed into carbon dioxide and water. A biological activity was present the whole time as the measurements proceeded (up to 51 days). The biological activity was higher in the flasks where TNT had been added compared to those samples into which no addition was made. Those samples also demonstrated more colonies of microorganisms per ml sample (higher biomass), thus pointing to the fact that the bacteria made use of TNT as energy resource having as result an increased growth.

In previous tests it was found that sea sediment had saturated after a period of approximately 168 days, this explaining binding of more TNT (Andersson et al., 1998). The sea sediment had low organic contents of about 2-3 % in contrast to the results from the present study. It shows that after having tested the sea sediment for a period of 160 days a few percent of added TNT remained in the water phase in the different flasks. The organic contents in the sediment were between 12-87 %. The greater ability of the lake sediment to bind TNT presumably depends on its more substantial amount of organic material. There was yet no connection between the binding ability and the amount of dry weight of sediment in the flasks. It did not even matter whether the organic contents was 12 % or 87 % since the accessible contact surface was more than enough for binding of the amount TNT added.

The leaching test made it apparent that TNT and its degradation products had bound tight to the sediments. The largest amount releasing itself, with the help of a single solvent (acetonitrile) was 5 % from the sediment in Pengsjön. At the most, 10 % of the initially added amount TNT was released (all solvent tests put together). This was also the case for the Pengsjö sediment.

The results from the toxicity test showed that the amount of TNT still remaining in the lake water after 53 days of simulation tests had no acute toxic effect on the survival of the water fleas. Repeated toxicity tests have established that the concentration resulting in the death (immobilization) of 50 % of the water fleas (LC50) is 8-10 mg TNT/l (Berglind, oral communication). This is consistent with the above test where the concentration of TNT in the water after 53 days was lower than the LC50 level in water fleas.

The test in which the effect of different factors on survival of water flea was investigated showed that some factors do affect the survival positively while others affect the survival negatively. Factors negatively affecting the water fleas include lead, copper, TNT, salinity (a higher salinity diminished the survival of the fresh water living water flea) and sodium. The survival was positively affected by zinc together with magnesium, potassium and calcium. The interaction between TNT and lead pointed to no negative effect on the water fleas being simultaneously subjected to both substances.

Conclusion and risk assessment

- No evident correlation between the concentration of metals in the water and the sediment has been observed.
- The concentration of heavy metals did not increase near the dumping ground.
- No correlation between the pollution pattern demonstrated by the lakes and the dumped ammunition could be observed, the source thus being another.
- The lake floor fauna demonstrated no unambiguous signs of being affected by the dumped ammunition.
- The investigated lakes were very different in character as confirmed by the fact that their lake floor fauna colonies differed considerably.
- There are no indications of that the dumped ammunition has started leaking yet.
- The proportion of TNT able to be produced in the lake floor water at the event of leakage had no effect on the survival of the water fleas. This is not saying that this holds for all lake floor living small organisms since the sensitivity of species varies strongly.
- The risk of presence of heavy metals from ammunition, having an effect on the toxicity of TNT in the lakes into which ammunition has been dumped is considered to be small.
- The simulation test demonstrates that over 90 % of TNT and its degradation products were bound to the sediment.
- The different chemical properties of the lake water did not affect the amount of TNT bound to the lake sediments under the testing conditions.
- The microorganisms in the lakes transformed TNT by using not coal but nitrogen in TNT as energy source.
- TNT and/or its degradation products are bound tightly to the sediment with the help of microorganisms. Despite having performed leaching tests using chemicals and biological activity, 80-90 % of added TNT was still bound after completed tests.

Lessons learned from additional research

The biological leaching illustrates the bioavailability, i.e. the accessibility of TNT to small living organisms after having bound to the sediment. The question of bioavailability has been treated in research regarding decontamination and soils (Caton et al., 1995, Funk et al., 1995, Sandström & Forsman, 1995, Deborah et al., 1996). In biological remediation TNT is transformed and bound to the soil. A plausible scenario could be that the soil eventually will end up in someone’s lungs through the inhalation of soil particles. In tests where composted, i.e. bioremediated soil was injected into the lungs of mice, it was established that 35 % was secreted via the urine, i.e. about 1/3 of soil-bound TNT was absorbed by their bodies (Palmer et al., 1997). In order to pinpoint the released degradation products and their presumptive health effects more studies are required.

Having been saturated the sediment allows percolation of TNT and its degradation products as demonstrated in the TNT lagoons in Hermiston, Umatilla, Oregon, US, where pollutants have been found in the water below the sediment layer. All TNT will probably bind with the help of the activity of the microorganisms, should the dumping grounds be covered with enough sediment. Accordingly, it is of essence to have some knowledge of both the high and low binding capacity of the surrounding sediment when performing a risk assessment of a dumping ground.

Short-term risks

The investigation of the four lakes has pointed out that there is no measurable TNT pollutant present today in any of the investigated lakes, despite the different prerequisites of, e.g. corrosion of ammunition casings or binding of TNT to sediment.

This finding is consistent with the theoretical calculations showing that it takes more than a thousand years for the ammunition casings to corrode thus allowing the contents to start leaking (Carlsson et al., 1997). Intact ammunition collected from two lakes (Tärnet, the County of Skaraborg, and Rammträsk, the County of Gotland) supports the theory. However, we have to fill the gaps of knowledge regarding the corrosive effect of ammunition in lakes having a low pH value.

Photo: Swedish Armed Forces.

A crate of ammunition that had been lying on the floor of Lake Rammträsk, the County of Gotland, for about 50 years, was brought up and opened. The interior of the crate, as well as the anti aircraft grenades themselves, were intact.

The explanation of the self-ignition of Lomtjärn is probably to be found in the substantial amounts of dumped luminous compounds generated by industrial activity.

Long-term risks

Long-term studies of bound TNT and the releasing of its products from soil or sediment are lacking. The present study describes a three-week test during which one tried to release TNT metabolites. The chemical as well as the bacteriological leaching indicated that the sediment in Pengsjön, a limed medium sized nutrition-deficient plains lake, do not bind TNT as hard as is the fact in the other lakes. Despite that fact, 80 % of the TNT metabolites are still present in the sediment following leaching. The results from this study and lessons learned from earlier experiences seem to indicate that the organic material in the sediment is of importance when it comes to binding of TNT and its degradation products.

Looking at the consequences it is of great importance to what degree the ammunition is covered with sediment when the leaking process starts. In the case where a chemical from the ammunition is leaking straight into the water, for example due to low sediment velocity in the lake, dredging etc. the risk of spreading of metals as well as explosives is imminent.

In the cases where the ammunition is well covered with sediment, the spreading is considered to be negligible. Laboratory results conclude that TNT and its degradation products are tightly and permanently bound to the sediment.

However, in the long run, metals can once again be released from the sediment depending on factors such as changes in pH levels in the water where, for example acidification can cause a sudden leakage of metals. Erosion, diffusion, transportation via small organisms living in the sediment, i.e. those feeding oxygen to and stirring up the sediment (bioturbation); or gas bubbles are other mechanisms worth mentioning in this context.

When performing a risk assessment of dumped ammunition it is vital to have knowledge of the chemical and physical characteristics, sediment velocity, sediment type, hydrogeological conditions such as water-supply, leakage, how often the water is turned over (turnover time) and the future usage of the lake. In order to be able to assess the spreading the turnover time is an important factor; another factor to be taken into account is the human influence through dredging, drainage, expansion, and alteration of land use etc. thus bringing about a monitoring of each and every lake. The latter is of importance since human activity, e.g. dredging, could release bound pollutants.

The effects of sediment polluted explosives on small organisms living in sea sediments were not investigated. The performed toxicity study used water flea as a model organism. No acute effects were found but literature studies, however, show that kidney failure due to intake of polluted particles of explosives can be proved. It is therefore uncertain to what extent the filtrating species living in the sediment would be affected by polluted sediment, particularly when considering the long-term effects.

In the described simulation test where the natural course of events has been accelerated, estimates have been made regarding the degree and speed of the detachment of the bound explosives from the sediment. At FOI, the Division of NBC Defence, research on binding mechanisms in natural soils is on-going thus enabling in-depth analysis and prediction of an eventual future release of sediment bound TNT.

Environmental risk assessment of ammunition dumped in the sea

Dumping of ammunition at sea continued until 1972 when the London Convention, prohibiting combustion and dumping at sea, came into effect. In Sweden, the polluted ammunition was for the most part dumped in the Baltic Sea.

The ammunition dumped at sea can be expected to start leaking earlier than that dumped in lakes since the higher salt content in the sea is presumed to accelerate the rusting process of the ammunition casings. Corrosion is also affected by oxygen access and temperature. The corrosion velocity in a maritime environment, the oxygen access being favourable, is about 0.1 mm/year (OSTC, 1988) having as a result that the explosives will come into contact with the surrounding water within a period of 50-100 years. In oxygen-poor sea and lake floors and sediment the corrosion proceeds more slowly than in open waters. Therefore, hundreds of years might elapse before the explosives start leaking. In order to be able to assess issues such as ecology and safety, FOI has, by order of the Swedish Armed Forces (in cooperation with the Swedish Environmental Protection Agency, NV) conducted a simulation test on TNT leakage at sea (Andersson et al., 2001).

Environmental impact of ammunition dumped in the sea

Samples were taken from three different sea environments, namely Fårö and Möja in the Baltic Sea and also from Stora Pölsan in the North Sea. The seafloors at Möja and Stora Pölsan are oxygen-rich while that at Fårö is oxygen-poor. The character of the sediments was determined by their different chemical and physiological properties. Tests showed that the carbon and nitrogen content of the different sediments was low and that the salt content in the water from Stora Pölsan was about 3-4 % as against the waters in the Baltic Sea, at Möja and Fårö having a salt content of about 0.5-0.6 % (brackish water). The water and sediments were analysed for TNT content as well as for its primary degradation products. No contents of such compounds were found within the sensitivity range of the instrument.

Simulation tests

At a simulation test in a laboratory environment, studies were conducted as to how TNT is distributed between water and sediment and also how hard TNT binds to the sediment. Water and sediment from the three sea environments were transferred into glass flasks. Carbon-14 labelled TNT was later on added thus enabling traceability. The samples from Fårö were percolated by a low flow of nitrogen to maintain an oxygen-free (anaerobic) environment throughout the test. The samples from Möja and Stora Pölsan were however continuously percolated by a low air flow (20 ml/min) for 70 days. In the course of the test and at the end of the same, analyses were performed in order to see in what way TNT and its degradation products separate between the water and the sediment. After additional 15 days (i.e. eighty five days in total), and in accordance with procedures previously followed, attempts were made to extract the compounds which had bound to the sediment (Caton et al., 1995).


Photo: Ann-Christine Andersson, FOI.

In order to secure an oxygen free environment during the simulation test of the Fårö sediment, a hermetically sealed glove box was used.

Toxicity test

Many small organisms live in or near the sea sediment and may therefore come in contact with compounds bound to sediment particles. Research has shown that sediment polluted by, for example heavy metals is poisonous to exposed organisms (Magnusson et al., 1996). It was also evident that extracts from the metal polluted sediment affected the small organisms negatively. For that reason a toxicity test (Berglind & Koch, 2003) was performed on water and sediment extracts from the three sea environments (Andersson et al., 2001). The small seafloor-living crayfish, *N.spinipes* (harpacticod, copepod) found among other places along the Swedish East Coast and tolerating a salt content up to 3.0 % was used as a test organism. The objective of the Berglind & Koch (2003) study was to establish whether the accumulation of TNT and its degradation products in sediment (Andersson et al., 2001) has a damaging effect on organisms living in a sediment environment. In a worst-case-scenario, the TNT pollutants were extracted under both high pressure (10 MPa) and high temperature (150^{o}C). Such an ASE extraction can be considered to mimic an extreme long-term leaching. The small organisms were exposed to the above described water and sediment extract (ASE extract) for 96 hours.

Results

At the end of the simulation test the content of TNT degradation products in all of the sea sediments was low. At the following extraction, the Fårö sediment, having been kept under oxygen-free conditions, demonstrated presence of TNT.

The experiments being completed, degradation products from TNT were found in the flasks containing water from Stora Pölsan. In two of those, into which TNT had been added, unchanged TNT could be traced. No such measurable compound content could be found in all the other samples. When the gas flow was throttled after 70 days, 60 % of the added TNT had disappeared from the water in the flasks having oxygen-free content from Fårö. In the flasks having contents from Möja and Stora Pölsan, with access to oxygen, 95 % of all added TNT had disappeared from the water. The amount of TNT in the water phase was monitored for additional 15 days; the amount of content was continuously dropping.

Nor could any transformation (mineralization) of TNT to carbon dioxide and water be established. No apparent increase in activity of the microorganisms could be observed in the course of the test. The addition of TNT had no effect on the pH value. The variation in the conductivity of the water had no measurable effect on the binding to the sediment.

The different extraction steps taken in order to detach the bound compounds from the sediment corresponded to a long-term leaching of about one month in total. TNT was found to have bound hard to the sediments. With the help of the strongest of the used solvents (potassium hydroxide), the largest quantity of TNT possible to detach from sediment at Fårö (oxygen-free), amounted to 5 % of the original addition.

No acute effect on *Nitocra* (cray fish) was indicated in the toxicity test of sea water and ASE extract in any of the cases. In the ASE extract about 5-12 % of the sediment bound TNT was found. This exchange corresponds to the one from the extraction with potassium hydroxide.

Conclusion and risk assessment

Below the conclusions from the performed tests have been analysed with the help of lessons learned from previous research.

The need for oxygen in the binding process

Under natural conditions TNT is probably accumulated in form of the degradation product 2,4-DANT. The activity of the microorganisms gives a permanent binding of TNT, 2,4-DANT and 4-ADNT. Held et al. (1977) reported results of experiments showing that TNT is built in form of 2,4-DANT. The microorganisms in the soil, having low access to nitrogen are trying to get hold of the nitrogen content in humus. Humus, a rich dark material, is a part of the soil consisting of a large complicated molecule of varying structure. Certain microorganisms make use of certain enzymes (peroxidases) in order to breake down the molecule. The theory is that these enzymes react with 2,4-DANT in that way incorporating it to the humus in the soil. This theory is substantiated by tests in which enzymes (peroxidases) react with 2,4-DANT but not with 4-ADNT or TNT (Held et al., 1997). Additionally, Elovitz

& Weber (2001) have reported a connection between the production of 2,4-DANT (out of TNT), by using microorganisms, and a permanent binding to sediment in aerobic environments. Without presence of oxygen the 2,4-DANT was instead transformed into two unknown substances left in the water phase, this having as result that the binding of the TNT degradations products to the sediment was partly prevented.

The above can explain why 40 % of added TNT remained in the water phase after completed testing in the oxygen-free environment (Fårö), to be compared to the 5 % of TNT remaining in the water in the oxygen-supplied environment (Möja and Stora Pölsan).

Possible leaching

The tests in this study show that reduction products from TNT are bound tightly to the sediment despite the low microbial activity present in the rather non-nutritious sediment. Reports by other scientists within the field concur with these findings. Achtnich et al. (1999a) reported that TNT polluted soil under oxygen-poor/oxygen-rich treatment is bound really hard to the soil sediment. Even after a long-term leaching with water for 21 months, only a few percent of previously bound TNT (Achtnich et al., 1999b) were dissolved and detached during the first few weeks of the 21 months. Achtnich and Lenke (2001) showed that neither fungi producing peroxidases (enzymes) nor increased temperature fluctuation or growth of green algae hardly affect the release of microbially bound TNT degradation products in soil. Only when green algae were observed measurable amounts of TNT products were released, namely 8 %.

These findings point to the fact that long-term leaching of TNT degradation products, bound to the sediment, is negligible. The compounds are built into the soil structure or in the natural sediment (Dryzga et al., 1998, Dryzga et al., 1999).

A strong binding of reaction products produced from TNT can therefore occur under natural conditions on the seafloor but an equilibrium concentration will always be present in the pore water, within the sediment, immediately near a sediment-covered, water-exposed TNT surface. How high a concentration, i.e. how far and in which direction the equilibrium is shifted, will depend on dilution velocity, temperature, leaching velocity etc. The concentration will be higher in oxygen-free sediment floors since the degree of binding is lower here. This in turns means that the pore water of the sediment immediately near a surface exposed to TNT may be poisonous (LC50>8 mg/l, Sjöström et al., 1999). In the study of the toxicity in sediments, having permanently bound TNT degradation products, results showed that microbial transformed TNT bound to sediment in the sea environment poses no acute poisonous threat to small seafloor-living organisms (Berglind & Koch, 2003).

Summary and discussion

Due to several reasons such as aging and malfunction, left ammunition after World War II was discarded in Sweden by dumping in abandoned mine shafts, lakes as well as at sea. By order of the Swedish Armed Forces, the Swedish Defence Research Agency (FOI), the Division of NBC Defence, have conducted several assessments on the environmental risk regarding dumped ammunition. The present report focuses on the results from the investigations on lake and sea dumping. In order to enable such an environmental risk assessment the following steps were involved; literature studies, field work, chemical analyses and laboratory tests (simulation of TNT release), with respect to retention of ammunition components to lake and sea sediment, respectively. Tests have also been carried out in lakes and at sea where the sensitivity to sediment and water polluted with ammunition in small organisms living on the seafloor was analyzed. From an environmental point of view, the risk of leakage of TNT and its degradation products as well as heavy metals from the dumped ammunition is of paramount importance**.**

The results show that the sediment in several lakes is severely polluted; however, no correlation between the pollution pattern and the dumped ammunition could be demonstrated. The investigation thus draws the conclusion that there is still no leakage. Consequently, the dumped ammunition presents no environmental threat and this also holds for investigated sea areas.

The laboratory simulations showed that mostly all added TNT was swiftly broken down through microbiological activity in the sediments to which the degradation products were closely bound (unextractable). This is true for both lakes and the sea but note that a considerably smaller amount of TNT seems to bind to the sediment in an anaerobic environment. Furthermore, toxicity tests showed that the amount TNT still present in the lake water following the simulation test had no acute effect on water flea (i.e. test organism). Similar tests on sea water and extracts of sea sediment showed that TNT did not present any danger to crustaceans living on the seafloor in terms of acute toxicity.

In the long run the risk of environmental impact is determined by to what degree the ammunition is covered with sediment as the leakage process begins. In case of leakage directly to the water, there is a considerable risk of spreading of metals and explosives. When it comes to lakes it might take up to a hundred years before the ammunition casings have rusted, at sea it can be a matter of about fifty to hundreds of years depending on salt content, oxygen feed and temperature. The time-span indicates that the ammunition is well covered before leakage thus creating satisfactory conditions for microbial degradation and retention of the pollutants to sediment. Dredging, small organisms, e.g. crustaceans, shells etc., stirring up the sediments, decomposition, acidification etc. are all examples of mechanical and chemical processes which might increase the risk of pollution.

Considering the long-term environmental risks brought about by the ammunition dumped in lakes as well as at sea, the time-span must be taken into account. In order to answer the question how long it will take for dumped TNT to start leaking, an assessment must be performed regarding how quickly the surrounding metal casings will corrode. As previously mentioned calculations show this will take up to a thousand years (Carlsson et al., 1997). Since the dumped ammunition is mostly packed in crates one must also take into account the time it takes for the explosives to leak. In a previous investigation regarding the dumping made in the lake Vänern by the defence industry, Carlsson et al., (1996) reported leaching times of 2000-20000 years for explosives, the calculations based upon the leaching times for explosives in waste and in process materials deposited in crates on the condition that the crates had not been covered with sediment.

Considering preliminary results from on-going tests using grenades cut in halves as well as theoretical calculations, a leak from dumped ammunition can be demonstrated in two ways. In the first example a hole in the metal casing appears through corrosion causing a flow of TNT to the surrounding water. For instance, in the matter of an anti-aircraft grenade, the solving ability and diffusion of TNT taken into consideration, the leakage will continue for 20000 years before the grenade is emptied, assuming that the hole will widen from 1 to 100 mm, presupposing a free water percolation around the grenade, i.e. the grenade laying outside of the crate. Supposing it is packed in a crate the time-span will increase further due to water percolation in the crate (cf. Holmén et al., 1998).

In the second example the TNT surface is fully exposed (a grenade cut in halves lying on the floor having its section surface up). The leakage will continue for at least 1000 years mainly due to the water percolation around the grenade. In reality, the grenades will successively be embedded in the sediment thus substantially prolonging the leakage. This is mainly due to the fact that TNT effectively binds in the sediments and, too, that the sediment cover will constitute a barrier preventing further leakage. The same leaking periods as those in the example above can therefore be expected.

Finally, from the experiments and tests it is concluded that the deposition areas having a high sedimentation are the most suitable ones when it comes to long-term storage of dumped ammunition.

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