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Widespread environmental contamination from relic munitions in the southwestern Baltic Sea

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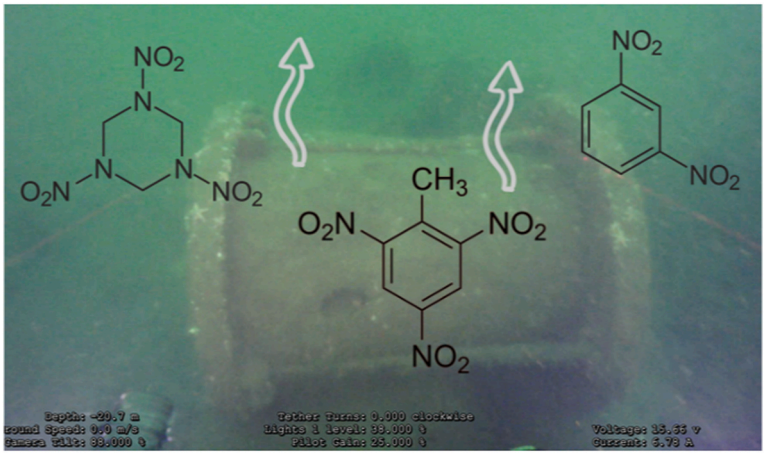
HIGHLIGHTS

- Explosive chemicals were detected at trace (pmol - nmol) levels in water and sediments throughout the German Baltic Sea.
- Explosive chemicals were primarily found in the dissolved phase, not adsorbed on particles or in sediments.
- Contamination hotspots were associated with known WWII-era munition dumpsites.
- An oceanographic model implemented TNT dissolution and degradation and successfully reproduced the observed distribution.
- Results show chemical release and spread from historical sea-dumped waste, which may negatively impact marine ecosystems

GRAPHICAL ABSTRACT



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ABSTRACT

Relic munitions from warfare and intentional dumping contaminate coastal waters worldwide, with an estimated 300,000 tons in the German Baltic Sea alone. These contain toxic conventional explosive chemicals, including 2,4,6-trinitrotoluene (TNT), 1,3,5-trinitro-1,3,5-triazinane (RDX), and 1,3-dinitrobenzene (DNB). Corrosion of metal munition housings in seawater releases these munition chemicals (MCs) to the marine environment. The current study performed detailed environmental sampling throughout German waters of the southwest Baltic Sea in 2017 and 2018, and measured MCs in water, suspended particles, and sediments. At least one MC was detected in nearly every water sample, from sub-pmol/L up to several thousand pmol/L. Most MC were in the dissolved phase, not on suspended particles, and MC content in sediments was patchy and generally low. TNT levels were especially high in Kiel Bay, whereas RDX and DNB concentrations were highest in Lübeck Bay, likely reflecting regional differences in munitions types. A TNT module was developed and implemented in the General Estuarine

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Transport Model (GETM), incorporating TNT input to the water column by dissolution and removal by microbial degradation/transformation. Simulated TNT distributions matched observed environmental patterns well, indicating good parametrization of the primary controls. Dissolved concentrations of the target MCs were generally far below acute or chronic toxicity levels for aquatic organisms, but the highest observed concentrations approached toxic levels, especially for DNB. The inventory of dissolved MC in the study region was approximately 3000 kg, implying that current contamination levels can be sustained continuously for over 800 years by existing munitions on the seafloor.

1. Introduction

Coastal marine waters are contaminated globally with unexploded ordnance and intentionally dumped munitions (Beck et al., 2018), especially as a result of the two World Wars (WWI and WWII). German territorial waters of the North and Baltic Seas are estimated to contain some 1.6 million metric tons of munitions (Böttcher et al., 2011). WWII-era explosives on the seafloor are often in intact and detonable condition (Nawała et al., 2020), highlighting an explosion and security risk. Underwater munition removal is increasingly necessary to allow sediment dredging (Greene et al., 2009) and offshore infrastructure development including aquaculture, wind farms, power and communication cables, and pipelines (Appleyard, 2015). In addition, relic munitions contain toxic and carcinogenic chemicals of conventional explosives and chemical warfare agents (Tornero and Hanke, 2016), and there is growing concern of environmental risks due to chemical release and exposure to marine organisms and humans (Sanderson et al., 2010; Bełdowski et al., 2017; Barbosa et al., 2023).

Underwater munitions include unexploded ordnance from wartime activities, as well as munitions from onshore munition depots that were dumped in designated areas close to coastlines. It was common practice during disposal activities to discard munitions overboard along the way to official dumping areas (Böttcher et al., 2011). Together with low position accuracy in pre-GPS times and dislocation of ordnance by fishing activities and currents, there is substantial uncertainty about the exact location and extent of underwater conventional munitions, even where good records of disposal exist (Benn et al., 2010; Brewer and Nakayama, 2008). It is therefore difficult to predict locations with increased risk of biological exposure to toxic munition-associated chemicals without detailed surveys of the seafloor.

The release of toxic munitions compounds (MCs) into seawater increases as a function of metal shell corrosion and contact with seawater (Wang et al., 2013), and food web exposure to MCs therefore depends on environmental factors affecting corrosion and dissolution (e.g., temperature, salinity, currents). The current state of corrosion of dumped munitions is poorly known in most cases and varies from 'very little' to 'completely degraded away' (Beddington and Kinloch, 2005). As a result, it is not possible to definitively determine if MC are released from underwater munitions without direct chemical measurement.

The primary MCs of concern include 2,4,6-trinitrotoluene (TNT), 1,3,5-Trinitro-1,3,5-triazinane (or "Royal Demolition Explosive", RDX), and 1,3-dinitrobenzene (DNB). Two primary metabolites of TNT, 2-amino-4,6-dinitrotoluene (2-ADNT) and 4-amino-2,6-dinitrotoluene (4-ADNT), are formed during biotic reduction of the respective nitro group (referred to together as ADNT; Juhasz and Naidu, 2007). Further nitro-reduction leads to diamino-mononitro-toluene isomers (DANT), and eventually to triaminotoluene (TAT). A number of studies have reported measurements of these MCs in seawater, sediments, and organisms, although often with limited success (see references in Lotufo et al., 2017; Beck et al., 2018).

Low levels of MCs (sub μg/l) have been occasionally detected directly adjacent to submerged munitions, but most studies have not detected MCs in seawater or sediments. Recent methods relying on sensitive mass spectrometry have shown that dissolved MCs in the Baltic Sea are on the order of 1–10 ng/L (Gledhill et al., 2019; Maser et al., 2023a), although higher TNT concentrations have been observed near exposed explosive solids (Beck et al., 2019; Maser et al., 2023a). Low ng/L levels have also been reported in the North Sea (Maser et al., 2023b) and Scheldt estuary (den Otter et al., 2023). Microbial MC biotransformation products (i.e., ADNTs) have also been detected in the water column, indicating that low measured concentrations of the primary explosives may be controlled by both release and removal processes. Measurable levels of MCs and their degradation products have been observed in sediments as well as in a variety of algae, invertebrates, and fish in the southwestern Baltic Sea (Strehse et al., 2017; Koske et al., 2020; Beck et al., 2022; Kammann et al., 2024).

The paucity of information on the environmental concentrations and spatial distribution of MC contamination in coastal waters makes it difficult to assess the environmental and human risk of this contaminant source. The current work characterizes the spatial distribution of MC in the water column and sediments in a region with abundant munition sources, and assesses their potential fate and impact on the marine environment. The southwest Baltic Sea is an ideal location for such studies because it has numerous, well-constrained munition dumpsites (Böttcher et al., 2011; Kampmeier et al., 2020) in a small area with limited mixing and shallow depth.

1.1. Study site

The current study focuses on German territorial waters of the Baltic Sea (Fig. 1). Regional water transport is controlled by inflow of high-salinity North Sea water through the Kattegat and Danish Belts along the seafloor, and surface outflow of brackish water (Bange et al., 2010). This causes fluctuations in bottom-water salinity between 17 and 24 PSU in Kiel Bay (Lennartz et al., 2014), and drives longitudinal salinity gradients to the east. Vertical mixing is limited during late spring and summer as the sea surface warms and a thermocline develops, which in association with the salinity gradient between inflowing and outflowing water masses forms a strong pycnocline. Vertical mixing to the seafloor occurs in winter due to storms and surface water cooling (Bange et al., 2010). Seasonal hypoxia and anoxia occur in the bottom waters during late summer in Kiel Bay, Lübeck Bay, and the Bay of Mecklenburg (Steinle et al., 2017; BLANO, 2018). Temperatures in the Baltic region are increasing with climate change, and the incidence of heat waves is likely to increase especially in the southwest Baltic (Meier et al., 2021). A heatwave occurred across Europe in summer 2018 (e.g., Gindorf et al., 2022), immediately prior to some of the current field sampling.

2. Methods

2.1. Field sampling

The study region was divided into sub-basins according to oceanographic considerations as well as the distribution of known munition dumpsites or contaminated areas (Fig. 1). This work focuses on an expedition with RV Poseidon during 1–21 October 2018 (cruise number POS530). Data from additional cruises with FK Littorina in September and October 2017 in the western portion of the study region are also included for higher spatial resolution around important munitions sites, and to include shallow water where RV Poseidon could not sample.

2.2. Munition compound analysis in environmental and experimental samples

Method details are described in the SI. Briefly, water column samples were collected with a CTD-mounted Niskin-rosette, and sediment samples were collected with a Van Veen grab (near-surface, to about 10 cm depth), multi-corer (approximately 25 cm depth), or by divers (surface 2–3 cm). Dissolved, particulate, and sediment MCs were processed and analyzed by ultra-high performance liquid chromatography coupled with high-resolution electrospray ionization mass spectrometry (HPLC-ESI-MS; QExactive Orbitrap, Thermo) according to Gledhill et al. (2019). An incubation experiment to evaluate TNT degradation was conducted with TNT-spiked Baltic seawater, and concentrations analyzed by direct-injection HPLC-ESI-MS.

Molar units are primarily used throughout this report in order to allow direct, quantitative comparison of different compounds, especially TNT and its transformation products. The target MC have similar molar masses (227.1, 197.2, 222.1, and 168.1 g/mol for TNT, ADNT, RDX, and DNB, respectively), and mass units in ng can be approximated by multiplying the reported values (in picomoles) by 0.2 ng/pmol.

2.3. GETM model and updated TNT module

Dissolved TNT concentrations were modelled using a 3-dimensional numerical ocean model (General Estuarine Transport Model – GETM; Klingbeil et al., 2018) with a spatial resolution of approximately 600 × 600 m and 42 vertical adaptive terrain-following coordinate levels (Gräwe et al., 2015). The TNT parametrization is based on lab and field experiments previously published (Beck et al., 2019) and described below. The TNT model is coupled to the GETM model via FABM (Framework for Aquatic Biogeochemical Models; Bruggeman and Bolding, 2014). Further details are available in the SI.

3. Results

3.1. Salinity, temperature, and dissolved oxygen

The salinity distribution throughout the study region in 2018 showed water mixing patterns typical of the southwest Baltic Sea (Fig. S1a). High salinity water (>25 PSU) enters the region through the Danish Belts at the far western border (near Flensburg Fjord), and flows eastward along deep channels. Salinity is lower (approximately 7 PSU) in the eastern Baltic (Arkona Basin), and this less dense fresh water flows westward over the intruding seawater. Temperature showed a similar pattern to salinity (Fig. S1b), but was elevated in shallow coastal waters (e.g., Kiel Bay) and restricted basins (e.g., Lübeck Bay). Temperatures during October 2018 ranged between 10 and 15 °C. Dissolved oxygen was high throughout the study region in 2018, except for deep waters in Flensburg Fjord, Kiel Bay, and Lübeck Bay (Fig. S1c). The latter site was particularly oxygen-poor, in some cases reaching anoxic conditions (dissolved oxygen concentrations at or below 1 μM).

3.2. Dissolved munition compounds

One or more of the target MCs were detected in every water sample collected in 2017 (n = 128), and in all but six of the samples from 2018 (98.8%; n = 537). Average concentrations were on the order of 1–100 pmol/L (Table 1). Concentrations of the four target MCs varied widely both within and among sub-regions in the southwest Baltic Sea (Fig. 2). The regions near Kiel (FH, KB, and HB) had highest concentrations of TNT and ADNT, and low levels of RDX and DNB. In contrast, the region near Lübeck (BM, LB) showed relatively lower levels of TNT and ADNT, and much higher concentrations of DNB and RDX. In most sub-regions, RDX and DNB were lower than TNT and ADNT. At the western boundary of the study region, Flensburg Fjord (FF) had intermediate levels of TNT, ADNT, and DNB. At the eastern boundary, the Arkona Basin (AB) showed the lowest MC concentrations. The Fehmarn Belt (FB) is the hydrographic link between the eastern and western halves of the

Table 1
Summary statistics for dissolved, particulate, and sediment MCs. The average ± 1 standard deviation is shown, with the detection frequency in parentheses. Sediment RDX and DNB were only detected in 3 and 1 samples, respectively, so only the range or single value are reported. n.m. = not measured, n.d. = not detected.

	Dissolved 2017 (pmol/L)	Dissolved 2018 (pmol/L)	Particulate 2017 (pmol/L)	Particulate 2018 (pmol/L)	Sediment (pmol/g)
TNT	18 ± 24 (95%)	17 ± 31 (95%)	n.d.	0.26 ± 0.3 (44%)	14 ± 63 (35%)
ADNT	18 ± 14 (96%)	38 ± 34 (98%)	0.51 ± 0.98 (20%)	0.19 ± 0.92 (5%)	10 ± 32 (73%)
DANT	n.m.	n.m.	n.m.	n.m.	2 ± 3 (44%)
DNB	1.5 ± 2.5 (55%)	440 ± 1300 (41%)	0.57 ± 0.05 (3%)	0.30 ± 0.63 (12%)	78 (1%)
RDX	3.4 ± 3.0 (86%)	93 ± 190 (48%)	0.33 ± 0.24 (19%)	0.15 ± 0.91 (8%)	0.9 - 7 (2%)

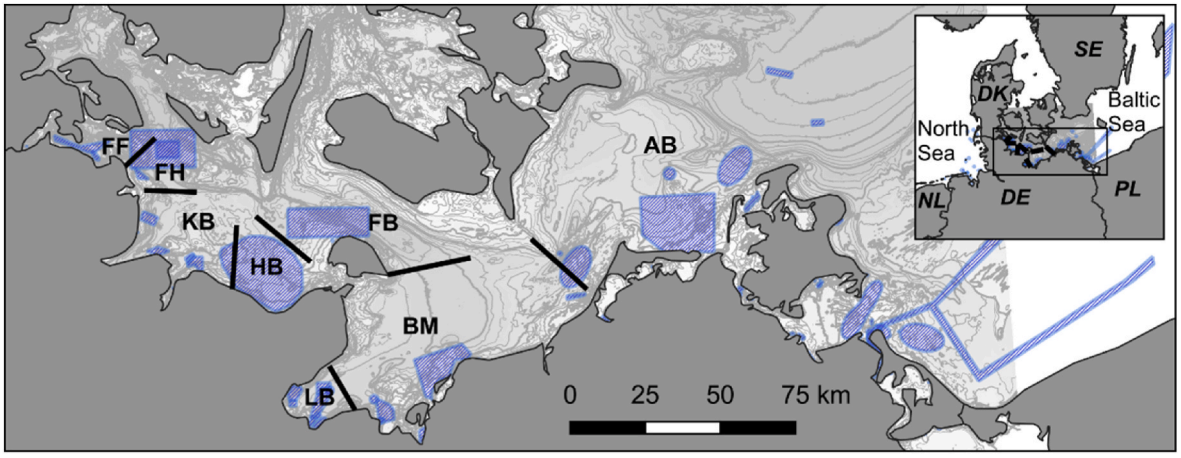

Fig. 1. Study region in the southwest Baltic Sea (FF, Flensburg Fjord; FH, Falshöft; KB, Kiel Bay; HB, Hohwachter Bight; FB, Fehmarn Belt; BM, Bay of Mecklenburg; LB, Lübeck Bay; AB, Arkona Basin). Blue polygons indicate munition dumpsites or areas with known or suspected munition contamination (from Böttcher et al., 2011; redrawn from north.io). Depth contours from GEBCO 2022; darker shading indicates deeper water (maximum depth 40 m).

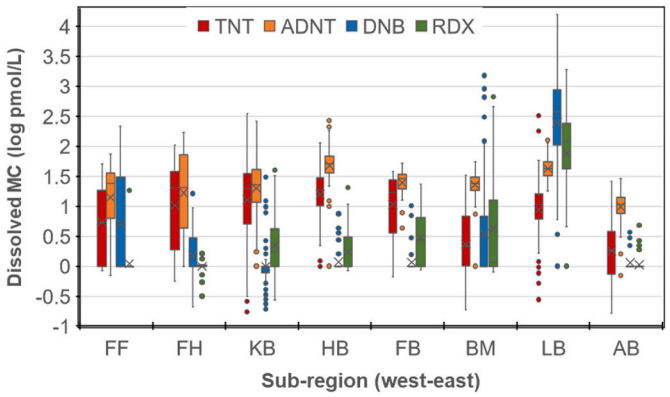

Fig. 2. Dissolved MC concentrations (log scale) in subregions of the southwest Baltic Sea. Boxes represent upper and lower quartiles, whiskers indicate maximum and minimum values, and dots are outliers. Horizontal lines within boxes indicate the median and crosses indicate the mean. Subregion names as in Fig. 1.

southwest Baltic, and MC concentrations were intermediate. Dissolved ADNT concentrations were generally higher than TNT, especially in the eastern part of the study region.

The spatial distribution shows distinct hotspots (west to east) in Falshöft, Kiel Bay, and Lübeck Bay (Fig. 3). These correspond to locations of known munitions dumpsites in Falshöft (BLMP German federal monitoring program ID number BKB10L), Kolberger Heide (in Kiel Bay, BKB04L), and Pelzerhaken (BLB01L) and Haffkrug (BLB02L) in Lübeck Bay. Although the hotspots showed relatively high levels of all MCs, there was a striking spatial difference between TNT/ADNT and RDX/DNB. TNT and ADNT concentrations were highest (above 100 pmol/L) in the western region near Kiel Bay, whereas RDX and DNB levels were relatively low (below 10 pmol/L). The opposite was true in Lübeck Bay, with very high levels of RDX and DNB (up to 1800 and 15,000 pmol/L, respectively), and lower levels of TNT and ADNT (generally lower, but with some samples exceeding 100 pmol/L).

The vertical distribution of dissolved MCs along a west-to-east transect shows these overall differences, and highlights hotspots of MC contamination at the seafloor (Fig. 4). In general, deep water concentrations were higher than those in surface water. This was especially evident for RDX and DNB below 10 m depth in Lübeck Bay, with concentrations exceeding several hundred pmol/L. This trend was less obvious in Kiel Bay, where high concentrations of TNT and ADNT were measured throughout the water column, likely due to the shallower water depth and greater mixing.

3.3. *Munition compounds in suspended particles and sediments*

Particulate TNT was not detected in 2017, but was measurable in 70% of the samples from 2018. Other MCs were detected in the particulate fraction in only 5–20% of the samples from both years, with concentrations as high as 5 pmol/L (i.e., MC associated with suspended particles per unit water volume), but averaging about 0.5 pmol/L or less (Figs. S2a–d). Unitless partitioning coefficients (dMC/pMC, K_d) were generally on the order of 0.1 or less.

A total of 176 sediment samples were collected from the MC hotspots in Flensburg Fjord, Kiel Bay, Hohwachter Bight, and Lübeck Bay (Fig. 5). Many of the target MCs were below detection limits in sediments (Table 1), and most samples with positive detection were from the Kolberger Heide munition dumpsite in Kiel Bay. ADNT was the MC most frequently detected in sediments, followed by DANT and TNT. RDX was

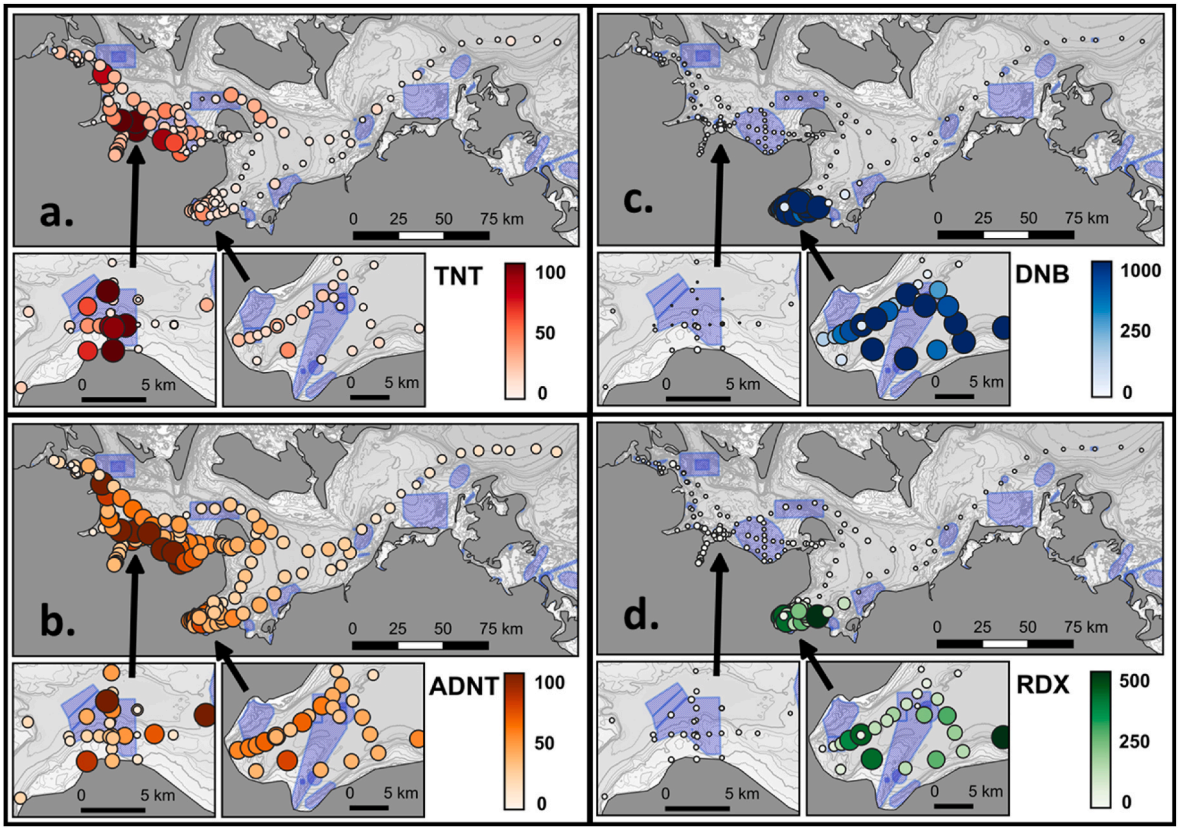

Fig. 3. Spatial distribution of dissolved a) TNT, b) ADNT, c) DNB, and d) RDX in bottom waters throughout the study region. The symbol size and shading indicate concentration (pmol/L). Insets show expanded views of the Kolberger Heide dumpsite (lower lefthand panels), and Lübeck Bay (Haffkrug and Pelzerhaken dumpsites; lower righthand panels). (Figure drawn in QGIS 3.34.0.).

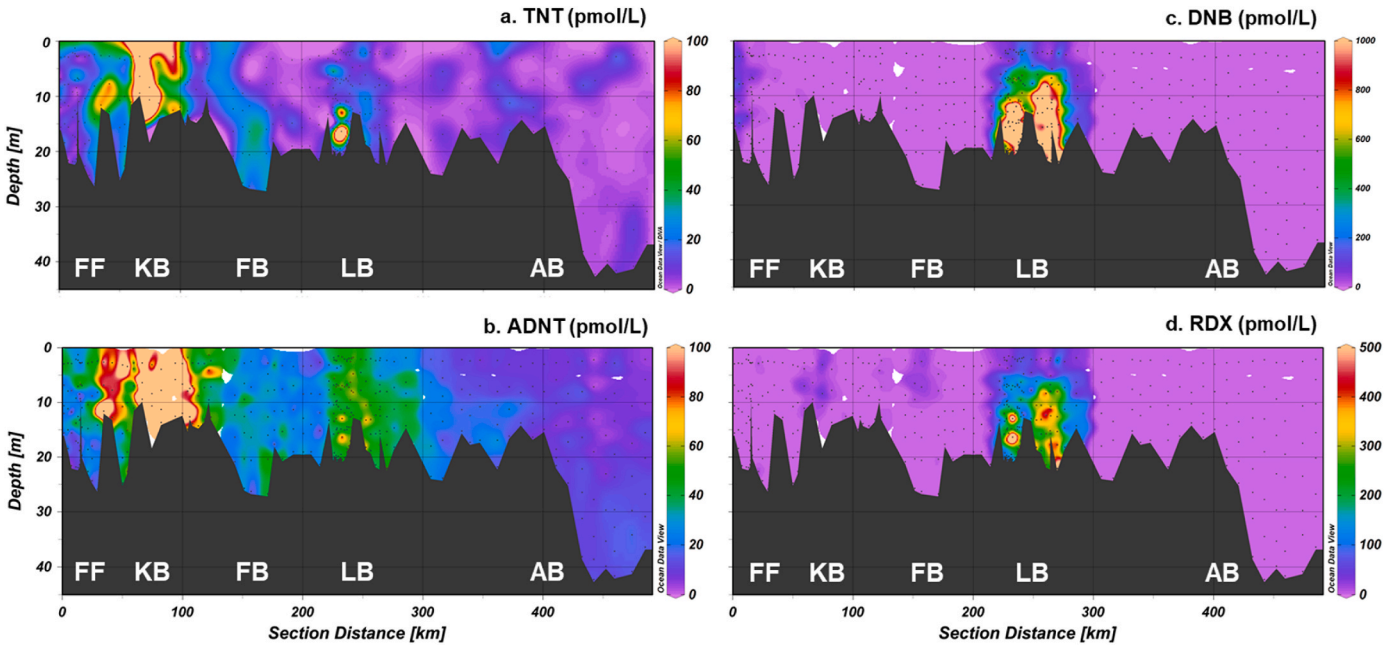

Fig. 4. Vertical distribution of dissolved MCs along a nearshore 2-D section from the western margin of the study region (Flensburg Fjord) to the Arkona Basin in the east (see Fig. S1, sub-region names as in Fig. 1). Note that the scale maxima are lower than the highest measured concentration to highlight spatial differences. Black dots show individual measurements and shading represents weighted-average interpolation. Drawn in Ocean Data View v.5.7.0 (Schlitzer, 2023).

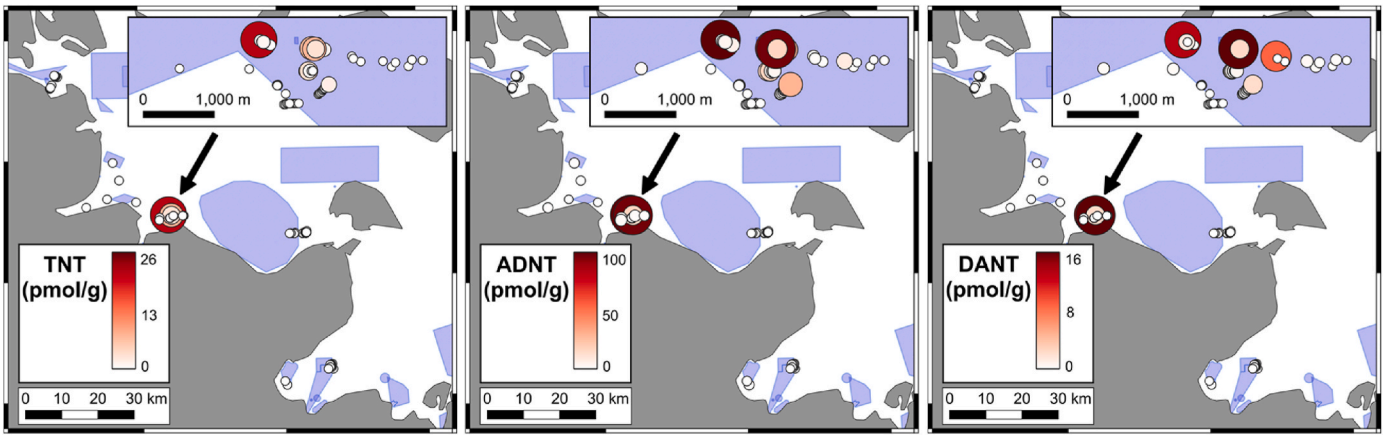

Fig. 5. TNT and transformation products ADNT and DANT in surface sediments. The inset shows an expanded view of samples collected around the Kolberger Heide dumpsite.

detected in only three samples and DNB in only one, all at the Kolberger Heide site.

To determine if heterogeneity in sediment MC content was related to proximity to individual munition objects, divers collected surface sediments radially between 0.5 and 2 m away from four mines in Kolberger Heide. The MC levels in these samples (Fig. S3) varied as widely as observed in the regional samples, with maximum levels between 9 and 465 pmol/g. The highest sediment MC levels (and the sole detection of DNB) were consistently found around one mine (Fig. S3c), suggesting that there is indeed an association between sediment MCs and munition sources. However, there was large heterogeneity in MC levels around individual objects, and no discernible chemical gradients within 2 m of the supposed sources.

Two sediment multi-core (MUC) sites were located about 3 km east and west of the munitions dumpsite at Kolberger Heide. ADNT was detected at both locations, but TNT was detected only at the western site (MUC 1, Fig. 6). Sediment MC content in the cores was very low, less than 1 pmol/g. Replicate cores at each of the two locations showed good agreement of the ADNT profiles, but clear differences among the two locations. In MUC 1, a peak in TNT content was evident between 7 and 17 cm depth, with no TNT detected near the surface. The ADNTs first appear at 19 cm depth, and show peaks at both the surface and 10 cm depth. In MUC 2, the ADNTs first appear at 11 cm depth and increase toward the surface. The isolated high value at 11 cm depth in MUC2-core 2 probably does not reliably indicate a peak.

3.3.1. *TNT degradation experiment*

The degradation experiment results showed strong removal of TNT over the 55-day incubation period (e.g., Fig. S4a), and an increase with temperature (Fig. S4b). Whole water TNT removal rates exceeded abiotic rates (in sterilized samples) by a factor of about four, indicating microbial control. Similar to other microbially-mediated reactions, TNT loss rates were strongly temperature-dependent (Q_{10} = 4.1). The transformation product ADNT increased during the experiment (Fig. S4a), but was never more than 50% of the initial TNT concentration. The subsequent daughter products, DANT and TAT, were not

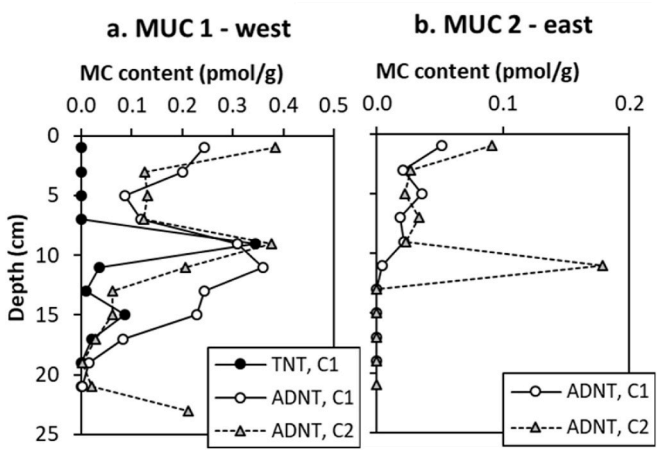

Fig. 6. TNT and ADNT content (pmol/g wet sediment) in replicate sediment cores at two locations a) west and b) east of the Kolberger Heide dumpsite (Kiel Bay).

analyzed in the experiment, so the ultimate fate of the lost TNT is not known (i.e., nitro-group reduction and cleavage vs. ring cleavage and mineralization).

3.3.2. Oceanographic TNT model

The complex vertical structure of the southwest Baltic Sea is evident in the simulated temperature, salinity and TNT concentration fields (Fig. S5). A multi-layer structure is evident along the main mixing channel in the Fehmarn Belt, Kadetrinne and across the Darss Sill. In the TNT field, low concentrations in bottom waters are evident in the Fehmarn Belt, overlain by water masses with a higher TNT concentration. This structure is mirrored by the salinity field. An inflow of dense saline water with low TNT concentration is overlain by less dense fresher water with higher TNT values, likely originating from Lübeck Bay.

A primary unknown in the model is the exposed area of solid explosives subject to dissolution. In order to calibrate the model appropriately, a sensitivity test was first conducted. The size of the exposed TNT surfaces was varied between a factor 10- to 100-fold smaller and larger than the estimated exposed TNT surface (2 cm^2/ha; see SI) to determine the source strength that best matched the measured TNT levels (Fig. S6b). The simulated TNT-concentrations showed a seasonal cycle with values of 2–7 pmol/L in summer (water temperatures 5–20 °C) and about 0.2 pmol/L in winter (water temperatures <5 °C).

The water age, or "ventilation age", measures the time elapsed since last contact with the sea surface, and is a good proxy for stratification. Water ages in winter are <1 d (well mixed water column), but are up to 60 d during summer when vertical exchange is limited by the pycnocline (Fig. S6a). The near-bottom TNT concentration shows a similar pattern of low values in winter and high values in summer. This annual cycle is mainly caused by the temperature dependency of TNT dissolution, and stratification and enrichment below the pycnocline. The model was further used to estimate the total TNT release in the southwest Baltic region, between 13 and 45 mol/d (or, 3 and 10 kg/d; Fig. S5c).

The simulated mean and maximum TNT concentrations (Fig. 7) match well with the field data distribution (Fig. 3). Near the dumpsite hotspots, the model predicts mean concentrations in deep water as high as 7–12 pmol/L. However, some tens of kilometers away, dissolved TNT drops below 1 pmol/L and below 0.2 pmol/L in the Arkona Basin. Maximum values occur in summer when high water temperatures and calm conditions lead to density stratification and enrichment of dissolved TNT in bottom waters near the munition sources.

4. Discussion

4.1. Observed MC levels and context for toxicity to organisms

The vast majority of water and sediment MC levels were very low, at trace to ultratrace levels (pmol to nmol per liter; Fig. S7). A few locations with especially high concentrations were found for all MCs, most obvious for RDX and DNB in Lübeck Bay. These highest levels likely reflect dissolved MC plumes sampled close to source hotspots. The MC levels observed in the current study (10s–100s of pmol per liter) are similar to munitions-contaminated sites in the Scheldt estuary (the Netherlands; den Otter et al., 2023) and Bahia Salina del Sur (Vieques, Puerto Rico; Rosen et al., 2022). Hotspots of release were also observed in the latter study, with concentrations elevated by nearly a factor of 1000 near particular munition objects compared with the rest of the bay. Lotufo and colleagues (2017) compiled a comprehensive dataset of MC toxicity values from the literature for a variety of organisms, and used these to develop the best available thresholds for aquatic toxicity due to acute and chronic exposure to MCs (Table 2). Comparing the highest levels observed in the southwest Baltic Sea with these Water Quality Criteria (WQC) shows that MC concentrations were a factor of 30 below the most sensitive chronic exposure threshold (for DNB), and nearly a factor of 1000 below the chronic WQC for the other MCs. Thus, under

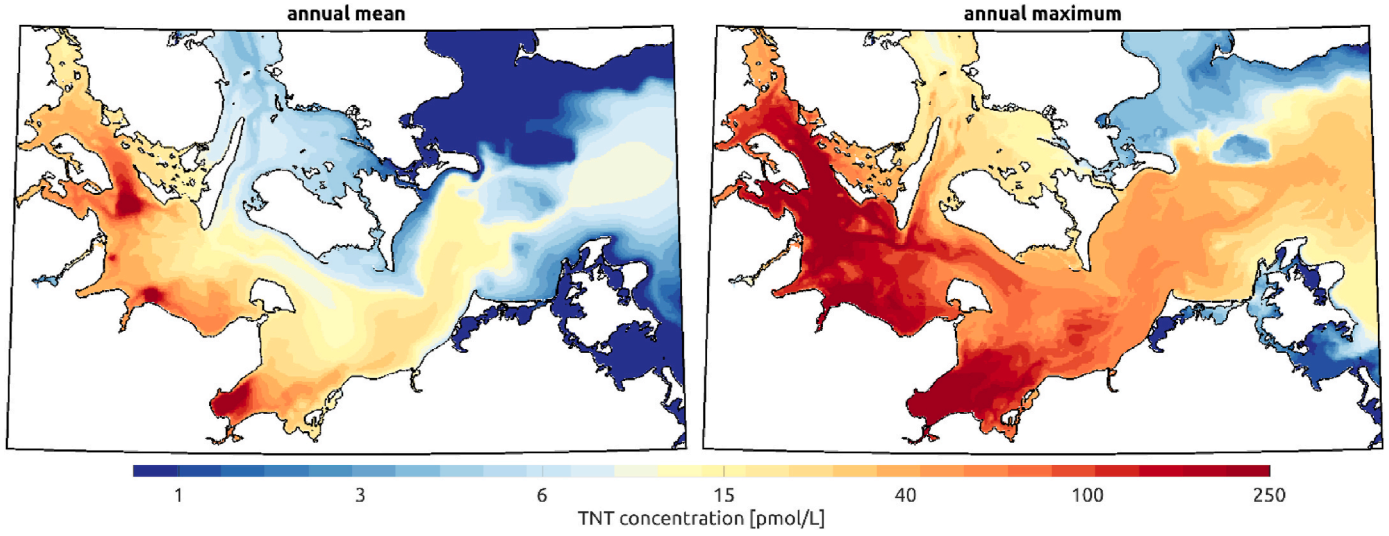

Fig. 7. Simulated TNT concentration in the bottom layer during the period 2006–2018. Left: annual mean concentration, Right: annual maximum concentration. Note the non-linear color scaling.

Table 2
Comparison of acute and chronic Water Quality Criteria (WQC) proposed by Lotufo and colleagues (2017) for the target MCs, and comparison with concentrations observed in the current study.

	Acute WQC (μg/L)	Chronic WQC (μg/L)	Acute WQC (pmol/L)	Chronic WQC (pmol/L)	Max. observed[a] (pmol/L)	Aquatic species[b]
TNT	398	32.6	1.8E+06	1.4E+05	3.5E+02	Marine
ADNT	180	74	9.1E+05	3.8E+05	2.7E+02	Freshwater
RDX	859	853	3.9E+06	3.8E+06	1.9E+03	Marine
DNB	194	76	1.2E+06	4.5E+05	1.6E+04	Freshwater

[a] Maximum dissolved MC concentration observed in the current study.
[b] Toxicity tests on species from freshwater or marine environment used to derive WQC. Values for the WQC in marine species were not available for ADNT or DNB.

current conditions and toxicity knowledge, MCs released from relic munitions do not pose a substantial direct risk to marine life in the southwest Baltic Sea, consistent with results showing no difference in fish health indicators near the most contaminated dumpsites (Kammann et al., 2024). Nonetheless, MCs may contribute to toxicity risk as a component of the environmental chemical contaminant mixture (e.g., Zeliger, 2003), which varies over time.

It is difficult to assess environmental levels of MCs against an appropriate metric for direct risk to humans, as few guidelines exist for human exposure to these chemicals. The German Federal Health Office (Bundesgesundheitsamt; BGA) has set drinking water guidelines for the target MCs measured in this study of 0.1 μg/L (DNB; ~595 pmol/L), 0.2 μg/L (TNT and ADNT; ~880 and 1020 pmol/L, respectively), and 1 μg/L (RDX; 4500 pmol/L) (Wollin and Dieter, 2005). For TNT in tap water, the US EPA has calculated a risk-based screening level of 2.2 μg/L (~9700 pmol/L; EPA, 2014). The US EPA also calculated a residential soil screening level (SSL) for TNT of 19 mg/kg and an industrial SSL of 79 mg/kg (approximately 84,000 and 350,000 pmol/g, respectively). The soil-to-groundwater risk-based SSL is 13 μg/kg (~57 pmol/g).

Human exposure thresholds are several orders of magnitude lower than the WQC for acute and chronic exposure of aquatic organisms (Table 2), although a few of the DNB concentrations in the current study exceed thresholds set for human health risk. Despite the fact that such high values were observed in a limited number of samples at specific hotspot locations, it does highlight the potential for public health concerns if environmental levels increase. At a heavily contaminated military test site in Vieques (Puerto Rico), Sanderson et al. (2017) concluded that exposure to munition related carcinogens, especially TNT via oral exposure, led to elevated cancer risk for the local people. Consumption of seafood may similarly pose a health risk in the Baltic Sea, although studies on that topic have thus far concluded that this risk is negligible (Maser and Strehse, 2020; 2021; Beck et al., 2022; Maser et al., 2024). Good understanding of the environmental behavior and fate of MCs is critical to evaluating current risk to the marine environment, predicting future changes in chemical release and spread, and constraining the role of relic munitions as an ecosystem stressor.

4.2. Sources

The source of dissolved MCs in the study region is primarily from well-known munitions dumpsites containing the vast majority of explosives (i.e., Böttcher et al., 2011). More than 400 explosive mixtures are reported for German explosives alone (Haas and Thieme, 1996), and a variety of mixtures are likely present in the explosive fill at the dumpsites (Kampmeier et al., 2020). Ten samples of solid explosives recovered from Kolberger Heide were identified as Torpex-like, with nearly equal proportions of TNT (41%) and RDX (53%) (Nawała et al., 2020). A dissolution study at the Kolberger Heide dumpsite found different types of explosives, with a "yellow-orange" variety releasing TNT, RDX, and very high levels of DNB (Beck et al., 2019). A "silver-grey" variety released primarily TNT and limited DNB or RDX.

Different explosive mixtures are used in different types of munitions for particular detonation characteristics (Haas and Thieme, 1996). Although not strictly the case, individual marine dumpsites mostly contain specific munition types (e.g., sea mines and torpedo heads, vs. small calibre flak grenades and tank projectiles) due to proximity to wartime storage depots on land or accessibility by ports near rail and road infrastructure (Böttcher et al., 2011; Rapsch and Fischer, 2000). Thus, although the exact sources cannot be definitively identified, it seems clear that release of different explosive chemicals from munitions dumpsites in the southwest Baltic Sea vary among locations, and that this is responsible for the observation of high TNT levels in Kiel Bay compared with high RDX and DNB in Lübeck Bay. The types of munitions present in the locations is known to be different (e.g., Kampmeier et al., 2020; Beck et al., 2024; Greinert et al., 2024).

Measured and simulated TNT concentrations generally matched well, but there was a marked difference in some locations, such as the far west of the study region (compare Figs. 3 and 7). In this location, the model predicts high TNT concentrations near the Little Belt munitions dumpsite (BLMP monitoring areas BKB06L & BKB10L), but field measurements generally showed low TNT levels. This can be explained by either limited amounts of exposed explosive material, or coverage of the munitions with fine-grained sediments (Böttcher et al., 2011) that limits release of MCs into the overlying water column.

Spatial differences in geo- and oceanographic conditions such as water depth and wind exposure may also play a role in the observed regional differences. Water mixing intensity due to currents and waves is likely to substantially control the spatial and temporal distribution of MCs in the Baltic Sea. Laboratory experiments show a strong influence of stirring rate on MC dissolution (e.g., Lynch, 2002). At the Kolberger Heide dumpsite, storm events greatly increase bottom current speeds and can even move munition objects on the seafloor (Kampmeier et al., 2020). The duration and intensity of storms is increasing due to climate change (IPCC, 2014), and this may lead to enhanced MC release in the future.

The sediment MC distribution indicates a connection between MC sources and sediment MC content, although a definitive link is not obvious due to a high heterogeneity. Regional scale patterns show highest sediment MC levels near known major sources (Fig. 5), and meter-scale patterns indicate elevated levels near some munition objects (Fig. S3). It is not clear if (or which) sediment properties may control this heterogeneity, or if sporadically-detected high sediment MCs detected near munitions are due to solid particles abraded from the surface of solid explosives (as observed after in situ low-order detonation, Maser et al., 2023a). Small particles of solid explosives would likely not persist due to their high surface area, but incongruent dissolution of explosive mixtures may hinder complete dissolution (Taylor et al., 2009).

Downcore trends in sediment cores from Kolberger Heide indicate changes in the release of MCs to the water column over time. Sedimentation rates in Kiel Bay are on the order of 2–3 mm/y (e.g., Orani et al., 2020), which puts the date of first MC appearance in the cores at ca. 70–100 years before collection (ca. 1920–1950), and the mid depth peak at ca. 40–60 years ago (1950–1970). The earliest appearance matches well with the timing of the two World Wars (1918–1945), similar to that observed elsewhere for munition-derived Hg (Sun et al., 2016). The mid-depth peak is consistent with MC release during dumping activities post-WWII (Rapsch and Fischer, 2000), which also included dumping of loose explosive material (Kampmeier et al., 2020).

The increase in MC content near the top of the sediment cores indicates that MC release has increased over the past 10–20 years. The current dataset cannot explain why TNT might have been preserved during the initial dumping period, whereas ADNT is apparently preferentially retained in the present era. Sequestration and persistence of TNT and ADNT in sediments is affected by transformation prior to burial (Smith et al., 2015), (differential) partitioning of TNT and transformation products to sediment phases (e.g., Dontsova et al., 2009), and post-depositional sediment diagenesis through both biotic and abiotic processes (Best et al., 1999; Spain, 1995). Thus, both pre- and post-depositional controls may affect the type of MC accumulation over time, and the general trends in the current dataset nonetheless suggest periods of increased release after initial dumping and in the present time.

4.3. Transport

Loss of dissolved TNT in the laboratory experiment gave a half-life of approximately 10 days (Fig. S4b), meaning that distribution patterns reflect release and transport within 50 days. Saline Kattegat water entering the western Baltic as dense bottom currents has a residence time of 14–21 days in the Danish Straits (Gustafsson, 2000), and MCs released in the western regions can be transported eastward through the Fehmarn Belt. The elevated MC levels in the Fehmarn Belt (e.g., TNT) are most likely the result of such transport, and not local sources. Although some areas in Fehmarn Belt are suspected to be contaminated with munitions (Böttcher et al., 2011), geophysical surveys show that munitions objects are not widespread (Kampmeier et al., 2018).

Large-scale circulation between the Fehmarn Belt and Arkona Basin restricts water exchange between Lübeck Bay and the open Baltic (Siegel et al., 2005). This causes the highly elevated MC levels observed in Lübeck Bay but not in the adjacent Bay of Mecklenburg (Fig. 2). Westerly winds transport waters from Lübeck Bay along the coast eastward (Siegel et al., 2005), which is partially visible in the high RDX and DNB concentrations along the southernmost coast of the Bay of Mecklenburg (Fig. 3). Elevated MC levels in surface waters of the Fehmarn Belt probably originated in Lübeck Bay, as confirmed by model predictions (Fig. 4 and S5).

Transport and alteration are evident in the spatial distribution of TNT and ADNT (Figs. 3 and 4). Close to MC sources, TNT was higher than ADNT (e.g., Kiel Bay). This was reversed farther from such sources, with the greatest disparity in the far eastern Arkona Basin (Fig. 2).

4.4. Fate - removal by scavenging and sedimentation

The low detection of the target MCs on particles and in sediments indicates that MCs are not substantially removed from the dissolved phase by scavenging and sedimentation. This is consistent with the low octanol-water partitioning coefficients for these MCs, and their similar low accumulation in biota (e.g., Monteil-Rivera et al., 2009a,b). It is not clear why particulate TNT was detected in 2018, but not in 2017, given that detection limits were unchanged among the two years. This may be linked to the wider spatial scope of sampling in 2018, or to differences in the quality (e.g., inorganic vs. biogenic particles) or quantity of particles at the time of sampling. There was no apparent correlation between particulate and dissolved MC concentrations (Fig. S2), and controls on MC sorption in the study region are not known. Sediment characteristics vary starkly among basins in the southwest Baltic Sea (Leipe et al., 2017), and the sediment sink for MCs may therefore vary spatially and increase during phytoplankton blooms or storms that resuspend sediments.

Regional scale patterns in sediment MCs showed elevated levels near the known TNT hotspot at Kolberger Heide (Fig. 5), but no obvious gradients around individual munition objects (Fig. S3). Thus, sediments are probably a minor repository for MCs, at least in their intact form, or preferential sorption of MCs to particular sediment phases leads to heterogeneous patterns that reflect sediment composition. This is consistent with isotope-labeled TNT in mesocosm experiments that showed only a small amount of TNT transfer from the water column to sediments (Ariyarathna et al., 2017). It is also possible that TNT in sediments is degraded or transformed to unidentifiable derivatives (Smith et al., 2015). RDX was not detected in most of the sediment samples, consistent with its degradation in sediments (Pennington et al., 2011). DNB was only detected in one sediment sample near a mine, and low particulate adsorbed DNB (Fig. S2) suggests that DNB is not scavenged from the aqueous phase or deposited into sediments.

4.5. Fate – removal by degradation or transformation

Dissolved TNT loss in the benchtop experiment gave a half-life of approximately 10 days, primarily due to microbial mediation, consistent with mesocosm experiments (Smith et al., 2013). The similar spatial distributions of RDX and DNB to TNT suggest that they degrade or are otherwise removed on a similar timescale. The degradation or transformation products of DNB are unknown, but RDX is likely to undergo ring cleavage and eventual mineralization (Monteil-Rivera et al., 2009a, b). The very high levels of dissolved DNB observed only in Lübeck Bay, together with low particulate- or sediment-associated DNB, suggest that DNB degrades rapidly in the study region and is probably not transported far from sources.

Temperatures and heat waves in the Baltic region are increasing with climate change (Meier et al., 2021). Due to the strong temperature dependence of both TNT degradation and dissolution (Beck et al., 2018), climate change may have profound future impacts on the spread and fate of MCs, but it is unclear how these competing processes will affect the net release and spread of MCs in seawater (e.g., Scharsack et al., 2021).

4.6. MC inventories

The total inventory of dissolved MCs was calculated for the area around each sampling site (details in SI), and summed for each subregion (Table 3). Estimated MC inventories mirror spatial differences in the water column distribution: the highest inventories of TNT and ADNT were in Kiel Bay and Hohwachter Bight, and the highest inventories of RDX and DNB in Lübeck Bay and the Bay of Mecklenburg. The high inventory of ADNT in the Arkona Basin may reflect older water and TNT degradation, although the large cell size and low sample density gives lower confidence in the inventories in this region.

The total MC inventory is approximately 3000 kg. Assuming that this inventory (I_{MC}) is at steady state ($J_{in} = J_{out}$) and reflects solely the balance of input by dissolution (J_{in}) and removal by degradation ($J_{out} = k_{degrad} \times I_{MC}$, where k_{degrad} is 0.1 d^{-1}), then the MC input/removal flux is on the order of 110 metric tons per year. It is difficult to estimate the exact mass of explosive material in relic munitions, given the wide range of potential munition types and the uncertainty of their identity even at well-studied dumpsites (e.g., Kampmeier et al., 2020). Historical records show that explosive fill constitutes anywhere between 10 and 70% of the total munition mass, although larger munitions typical of many

Table 3
Inventories of dissolved MCs in different subregions of the southwestern Baltic Sea.

Region	TNT (kg)	ADNT (kg)	RDX (kg)	DNB (kg)
Arkona Basin	108	276	8	8
Bay of Mecklenburg	55	281	224	232
Fehmarn Belt	89	119	22	2
Flensburg Fjord	1	18	12	26
Falshöft	0	1	40	54
Hohwachter Bight	68	133	8	1
Kiel Bay	184	250	12	6
Lübeck Bay	20	48	221	714
Total	576	1188	496	982

dumpsites in the German Baltic Sea are generally in the 30–60% range (USDOA, 1943, 1944; USN, 1945, 1946a, 1946b; Royal Armouries, 1945; Fleischer, 2004) Taking the explosive fill as 30% of the total munition mass as a conservative estimate, then the estimated 300,000 tons of munitions in the German Baltic Sea (Böttcher et al., 2011) can sustain current levels of contamination for more than 800 years. This would only be possible under the unlikely scenario of constant corrosion conditions over such a long time. Indeed, modeling suggests that chemical release will greatly accelerate during the early 21st century as munition housings are increasingly breached by corrosion (Glasby, 1997).

There are approximately 1600 tons of explosive material at Kolberger Heide, mostly present in ground mines on the seafloor surface (Kampmeier et al., 2020). The simultaneous seawater exposure of many similar munition objects suggests broadly similar corrosion and breaching. An inventory of 1600 tons of explosive material exposed simultaneously to dissolution at this one dumpsite has the potential to increase MC levels by a factor of 10 or more in the entire German Baltic Sea.

4.7. Prediction of TNT release and spread

To provide a performance measure of the implemented TNT module in GETM, we compare the measured TNT concentration during October 2018 with the simulated values (Fig. 8). Especially for TNT values smaller than 5 pmol/L, the model shows good predictive skill. This good agreement shows that the source and sink control by dissolution and microbial degradation are reasonably well-parameterized. However, because it provides a mean concentration over 600 × 600 m grid cells, the model underestimates the highest measured values.

Not all sources of underwater munitions are included in the model. Even in munition dumpsites, the munition object density is sparse and patchy, and often poorly known. At the Kolberger Heide dumpsite, bathymetric mapping identified 1136 munition objects within the 6.63 km^2 dumpsite (Kampmeier et al., 2020). Even at a conservative coverage of 1 m^2 per object, this represents only about 1.7% of the total area. In addition, the condition of munition objects is known from visual observation to be highly variable. One area with open explosive solids in Kolberger Heide is likely a primary source of MCs to the water column (Kampmeier et al., 2020).

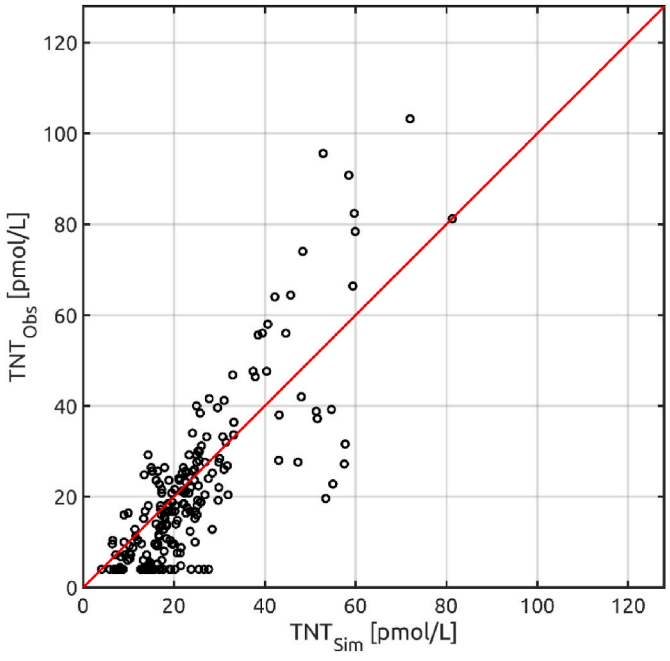

Fig. 8. Comparison of measured and simulated TNT concentrations for October 2018. The red line has a slope of unity and indicates perfect prediction.

Source regions were limited here to those known in German waters, but chemicals such as chemical warfare agents are known to be released from underwater munitions in dumpsites elsewhere in the Baltic region (Missiaen and Henriet, 2002; Bełdowski et al., 2016). Blow-in-place clearance in the Danish Straits has been shown to release substantial MCs (Maser et al., 2023), although most of the region, including a major munitions dumpsite in Aarhus Bay, hasvery low levels of dissolved TNT (<0.3 pM; Sanderson and Fauser, 2023). Improving international communication and cooperation on relic munitions is critical to constrain the extent of the problem in the Baltic region and elsewhere (e.g., JPI Oceans, 2024).

5. Conclusions

Results of the current study clearly show that chemical contamination is released from underwater relic munitions and spreads throughout the Baltic Sea. Concentrations of toxic explosives such as TNT are currently very low, mostly below regulatory drinking water limits or levels shown to have negative toxicological effects on marine organisms. However, future release of MCs from underwater munitions in the southwest Baltic Sea is certain to increase as corrosion advances, and contamination levels will most likely rise if no action is taken to remove munitions from the sea. In the first such action globally, at the end of 2021, the German federal government allocated 100 million Euro for a pilot program to begin test removal and on-land disposal of munitions from underwater dumpsites (Mergener, 2024). In comparison with blow-in-place disposal (e.g., Maser et al., 2023), this method of remediation is likely to have low negative environmental impact, although there is a potential for heightened chemical release due to munition disturbance. The clearance will begin in Lübeck Bay, which has some of the highest levels of environmental MC contamination. This will provide a proof-of-concept for remediating underwater munitions, and allow cost-benefit evaluation in comparison with the "monitored no action" approach proposed in a US military SERDP/ESTCP workshop in 2021 (Lotufo et al., 2021).

This study provides a clear picture of the environmental contamination from only one of many types of chemical waste disposed of in the world oceans prior to the London and Oslo conventions forbidding ocean dumping. There is emerging evidence that similar processes of corrosion and breaching lead to the modern release of toxic historic contaminants, such as DDT, from sea-dumped waste (e.g., Kivenson et al., 2019). Results of the current study suggest that corrosion and release of sea-dumped waste may represent a new hazard, and should perhaps be considered "historical contaminants of emerging concern." In contrast to diffuse environmental pollution, containerized dumped waste is in a concentrated, already-packaged form that can be physically removed from the environment The planned upcoming munitions remediation action in Germany will provide a model for remediating such waste and permanently eliminating at least one environmental stressor from the marine environment.

CRediT authorship contribution statement

Aaron J. Beck: Writing – original draft, Investigation, Formal analysis, Conceptualization. **Martha Gledhill:** Writing – review & editing, Methodology, Investigation. **Ulf Gräwe:** Writing – review & editing, Software, Methodology, Formal analysis. **Mareike Kampmeier:** Investigation. **Anja Eggert:** Writing – review & editing, Software, Investigation, Formal analysis. **Christian Schlosser:** Project administration, Investigation, Funding acquisition. **Beate Stamer:** Methodology, Formal analysis. **Jens Greinert:** Writing – review & editing, Resources, Project administration, Funding acquisition. **Eric P. Achterberg:** Writing – review & editing, Resources, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2025.144115.

Data availability

Data will be made available on request.

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