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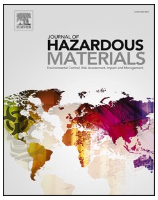


Tracking the invisible threat: Persistent explosive residues in raw and drinking water bodies

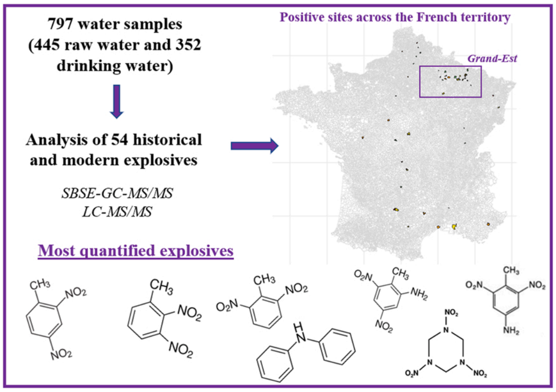
Dani Khoury [*], Christelle Pallez, Sophie Lardy-Fontan, Christophe Rosin

ANSES, Nancy Laboratory for Hydrology, Water Chemistry Department, 40 Rue Lionnois, Nancy 54000, France

HIGHLIGHTS

- First nationwide survey of 54 explosive residues in raw and drinking water
- Sampling represented 20–25 % of the French population
- DNT isomers and RDX were most frequently quantified
- Historical residues were strongly correlated with perchlorate
- Water treatment processes partially remove explosive residues

GRAPHICAL ABSTRACT



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ABSTRACT

This study provides a comprehensive nationwide assessment of explosive residues and their degradation products in French water resources and drinking water, including mainland and overseas territories. A total of 54 target compounds were selected based on historical usage, environmental persistence, and analytical feasibility. Between 2020 and 2022, water samples from nearly 300 sites—representing resources for 20–25 % of the French population—were analyzed using high-performance liquid chromatography–tandem mass spectrometry (HPLC-MS/MS) and gas chromatography–tandem mass spectrometry (GC-MS/MS). Explosive residues were detected in 11 % of the 797 samples, mainly in raw groundwater. Frequently identified compounds included 2,6-dinitrotoluene (2,6-DNT), diphenylamine (DPA), and hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), while others appeared less often but sometimes at high levels. These contaminants were largely linked to historical battlefields and former munitions storage areas, especially in the *Grand-Est* region bordering Germany. Degradation products like aminodinitrotoluenes (ADNTs) were more common in drinking water, possibly due to incomplete removal or transformation during treatment. Significant correlations between DNT, nitrotoluene (NT), and perchlorate suggest a shared origin tied to historical military activity. The findings highlight the need for continued monitoring of explosive residues, not only in France but in other regions affected by historical or current military operations.

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[*] Corresponding author.
E-mail address: dani.khoury@anses.fr (D. Khoury).

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1. Introduction

Explosive residues and their transformation products represent a significant class of environmental contaminants with persistent and hazardous characteristics. These compounds, including 2,4,6-trinitrotoluene (TNT) and its degradation products of aminodinitrotoluenes (ADNTs), hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX), 1,3,5,7-tetranitro-1,3,5,7-tetrazocane (HMX), and various dinitrotoluene (DNT) and nitrotoluene (NT) isomers, have been widely used in military, industrial, and mining applications for over a century. They have been released into the environment through multiple pathways: munitions production, handling, detonation, disposal, and legacy pollution from former battlefields and manufacturing sites [23,59]. These substances pose multiple environmental and human health risks. Many nitroaromatic compounds are toxic, mutagenic, or carcinogenic, while also demonstrating resistance to biodegradation, particularly under aerobic conditions [3,48]. Their presence in groundwater (GW) and surface water (SW) is of increasing concern, especially in regions with a history of military activity or the production of explosives, such as northeastern France—an area heavily impacted by the First and Second World Wars [9]. This issue has been compounded by the persistence of explosive degradation products such as ADNTs, which are often more water-soluble and mobile than their parent compounds [51]. These transformation products can be transported away from their original source, posing a long-term exposure risk. In recent years, several studies have addressed the environmental fate of explosives, primarily in North America and Germany, focusing on localized contamination from military installations or former manufacturing plants [54,8]. However, comprehensive nationwide monitoring campaigns remain limited, particularly in Europe, where historic and modern sources often overlap and where large-scale surveillance of GW and drinking water (DW) has rarely been conducted for explosive residues. France, given its unique combination of historical military contamination and ongoing industrial and defense-related activities, represents a critical case for such an investigation.

Following the widespread detection of perchlorate (ClO_4^-) in SW, GW, and DW catchment areas, concerns were raised about the potential presence of explosive residues like DNTs in water resources [26]. Analyses showed that military sources related to World War I were the primary cause of its contamination. Additionally, ClO_4^- hotspots have already been identified in France through a broad national monitoring program, which can help us to guide the search for related substances in water resources [49]. In fact, ClO_4^- holds particular significance due to its extensive use as an oxidizing agent in both industrial and improvised explosive formulations [38]. Commonly found in fireworks, flares, propellants, and various homemade explosive devices, ClO_4^- is characterized by its high solubility, chemical stability, and persistence in aquatic environments [13–15]. These properties facilitate its migration into water systems, where it may persist for extended periods and serve as an early warning indicator of broader contamination by explosive residues.

The global objective of this work was to assess the occurrence of emerging explosive residues in French water bodies on a national scale. Specific objectives were to select a broad suite of target compounds (54 explosive residues) along with the inclusion of ClO_4^-, design a national sampling strategy of RW and DW sources, characterize the levels and footprint of contamination in water resources and DW, and identify the sources and areas of concern. To our knowledge, this is the first comprehensive nationwide screening of such a broad spectrum of explosive residues in the water supply of either France or Europe.

2. Experimental section

2.1. *Choice of compounds*

The selection of the targeted explosive substances for national screening in France was primarily based on both their historical usage and environmental persistence. The selection of compounds was further based on scientific literature reviews [59,9], analytical feasibility, expert consultations among French environmental institutes and health authorities, notably the *Bureau de Recherches Géologiques et Minières* (BRGM) as well as regulatory standards (e.g., [16], NF EN ISO 22478). The final list includes 54 explosive residues and is found in the supplementary materials. It includes secondary explosive substances (explosive military charges) like TNT, melinite (2,4,6-trinitrophenol or picric acid), RDX (hexahydro-1,3,5-trinitro-1,3,5-triazine or "hexogen"), HMX (cyclotetramethylene-tetranitramine or "octogen"), di- and trinitronaphthalenes, and PETN (pentaerythritol tetranitrate); compounds used in the formulation of primary explosives, such as tetryl (N-methyl-N,2,4,6-tetranitroaniline); impurities and by-products of the environmental transformation of explosive substances with 2,4/2,6-DNT and isomers of ADNT; the family of nitrodiphenylamines used as additives (e.g., as powder stabilizers); and mobile compounds used in the composition of propellant powders, such as nitroglycerin. The physico-chemical properties (molecular weight, melting point, water solubility, octanol/water partition coefficient, Henry's law constant, vapor pressure, and organic carbon/water partition coefficient) of the explosive residues under study are shown in the supplementary materials.

2.2. *Sampling strategy*

The sampling design covered the entire French territory, including both metropolitan and overseas *départements* (a French administrative unit), and was conducted in partnership with the regional health agencies (ARSs) and the French Ministry of Health ([17–19],2020; [20]). Fig. 1 illustrates the spatial distribution of the collected water samples covering the entire French *départements*. A particular focus was placed on the *Grand-Est* region, owing to its historical importance as a center of warfare and intensive ammunition production and storage. This area was therefore investigated in greater detail, with a refined assessment of total concentrations in relation to prominent historical sites and former military-industrial activities.

The current study encompassed nearly 300 sites evenly distributed across the whole territory, for which RW and DW pairs were sampled and analyzed. The collected samples are representative of the water supply for approximately 20–25 % of the French population. For each *département*, three types of sampling sites (RW/DW sample pairs) were used (see Fig. 2): high-flow points (HFs), which include resources providing the highest volume of DW in the *département*; random points (RPs), which refer to resources selected through random sampling within each *département*; and interest points (IPs), which correspond to sites identified by the ARS as having particular significance for targeted substances or because they are considered vulnerable. These include locations with known or potentially high levels of ClO_4^- ions, areas near industrial activities related to weaponry and explosives, and sites close to former weaponry storage or disposal areas. The sampling campaign was held from October 2020 to July 2022. Overall, 797 samples grouped as GW, SW, and DW, were collected during this period. Spot samples were collected in amber glass bottles concurrently from both raw and drinking waters, without consideration of the water's residence time within the treatment facilities. For LC analysis, samples were divided after collection and poured into two 40 mL amber glass vials: one non-acidified, and one stabilized with 0.05 % glacial acetic acid and 2 % acetonitrile in order to extend the analysis window for TNT. The bottles were transported at 4 °C using cold packs in polystyrene containers and delivered to the laboratory within 24–48 h. Only a few samples reached the laboratory one week after collection (particularly in the case of the overseas territories). In most cases, the samples were analyzed within 4–5 days of receipt. Sodium thiosulfate ($Na_2S_2O_3$) was added to DW samples while still in the field to neutralize residual free chlorine and prevent the degradation of molecules after sampling.

2.3. Standards and reagents

Individual standard (comprising isotope-labeled) compounds of a purity higher than 98 % were purchased from Neochema (Bodenheim, Germany), Cluzeau (Sainte Foy la Grande, France), Envilytix GmbH (Borsdorf, Germany), and Techlab (Saint-Julien-lès-Metz, France). Two separate working mixtures containing the targeted compounds were prepared at 1 mg L^{-1} and then stored at 4 °C in the dark. Solvents (water, acetonitrile, isopropanol, and methanol) were liquid chromatography–mass spectrometry (LC–MS) grade (>99.9 % purity) and were purchased from Biosolve (Valkenswaard, the Netherlands). Sodium chloride and acetic acid (CH_3OH) were purchased from Merck KGaA (Darmstadt, Germany). $Na_2S_2O_3$ was purchased from Sigma-Aldrich (France). Formic acid (HCOOH) and ammonium acetate

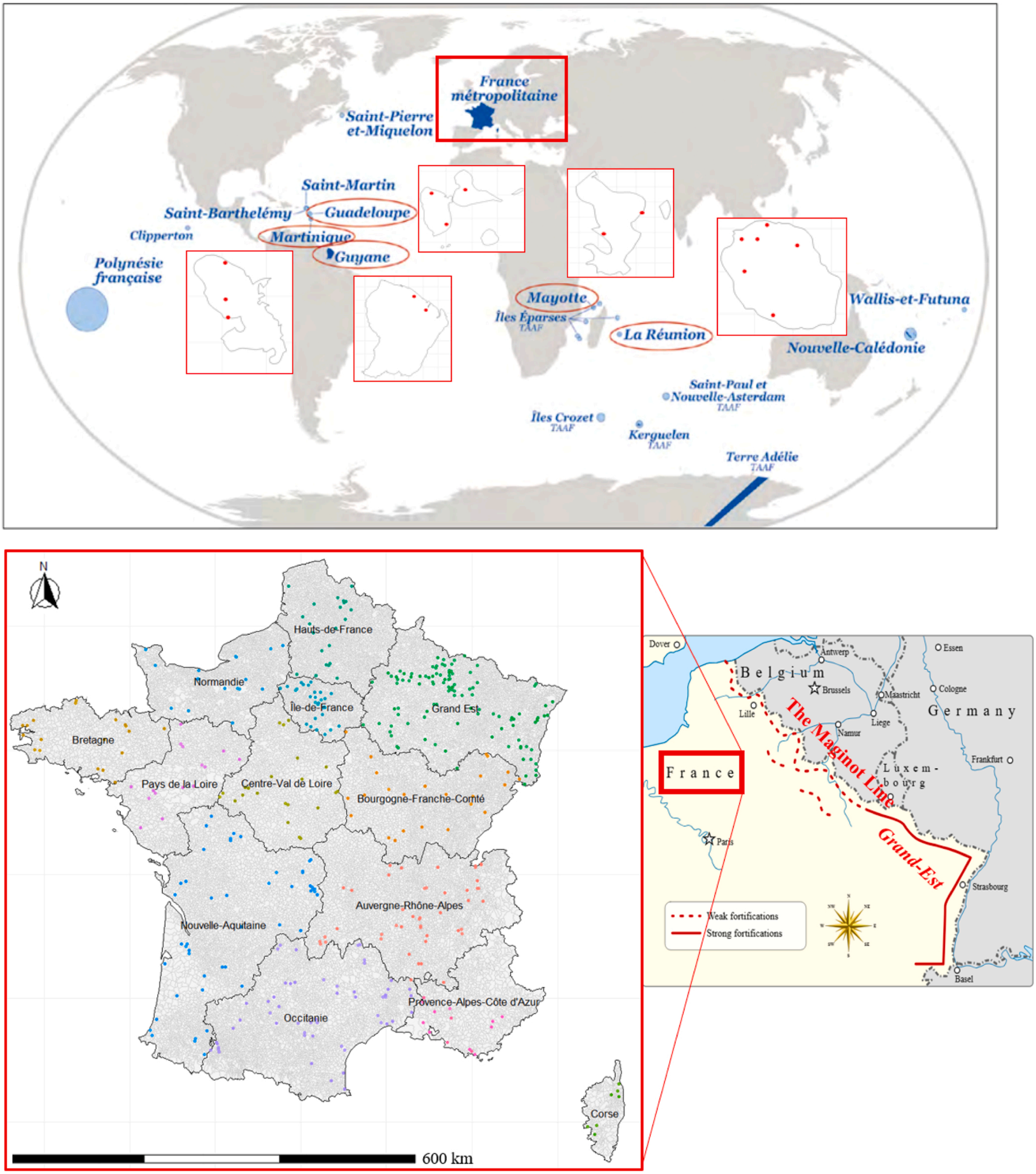

Fig. 1. Sampling locations across the entire French territory, including both metropolitan (shown in the lower figure with the Maginot Line indicated) and overseas territories (shown in the upper figure).

(CH_3COONH_4) were purchased from Biosolve (Valkenswaard, the Netherlands). SBSE was performed using an IKA RO 15 magnetic stirrer manufactured by IKA Werke GmbH & Co. KG (Staufen im Breisgau, Germany).

2.4. Analytical methods

However, given the diversity of explosive substances (in terms of their different physicochemical properties) and the need to analyze a broad range of molecules with the most appropriate quantification limits, two separate analytical methods were implemented. Twenty-three molecules were analyzed via DI-HPLC-MS/MS, and the rest were analyzed using an innovative technique based on SBSE followed by desorption and analysis via GC-MS/MS [21]. The former was adopted for analyzing key compounds such as TNT, HMX, RDX, and nitroglycerin metabolites [11,31], while the latter was particularly suited for differentiating isomers of nitroaromatic compounds. More details on the chromatographic (gradients, mobile phases, columns, etc.) and MS parameters (parent ion, confirmation and quantification ions, collision energy, ionization mode, and declustering potentials) for both analytical methods are shown in the supplementary materials.

2.5. Performance of both methods

These two methods were validated using the statistical approaches outlined in the NF T90–210 standard [1], which is consistent with SANTE guidelines. Validation efforts focused on a representative water matrix, encompassing a diverse set of samples with varying physicochemical characteristics reflecting the environmental and geochemical contexts in which the method would be deployed, including SW, DW, and GW. During the method development phase, a critical assessment of matrix effects was undertaken across these different water types. This process facilitated the appropriate pairing of labeled internal standards with their corresponding target analytes and, where necessary, led to the inclusion of homologous internal standards for compounds inadequately corrected by non-homologous surrogates. This step was crucial to minimizing quantification errors linked to matrix effects [40,44].

For both analytical methods, seven to nine concentration points were used for calibration. The quadratic fit of the calibration curves was systematically verified for each batch of samples, ensuring an R-square value of at least 0.98 and a maximum bias of less than 20 % for each calibration point. Target compounds were quantified using the internal standard method, incorporating deuterated or carbon-13 (^{13}C), or nitrogen-15 (^{15}N) isotopically labeled compounds. These standards were added to each water sample prior to analysis to monitor the overall recovery efficiency of the target compounds throughout the analytical process.

The limit of quantification (LQ) and measurement uncertainty were determined for each compound (see the supplementary materials). The LQ in a matrix for the molecules analyzed via DI-LC-MS/MS ranged from 10 to 50 ng L^{-1}, with an application range between 10 and 1250 ng L^{-1}. For the molecules analyzed by SBSE-GC-MS/MS, the LQ ranged from 1 to 50 ng L^{-1}, with an application range from 1 to 2500 ng L^{-1} depending on the sensitivity of the targeted compounds. The maximum relative uncertainties ($k = 2$) were 50 % at the LQ. Only seven explosive substances had a relative uncertainty higher than 50 % and up to 70 %, caused by some analytical factors (such as the compound's stability). However, they were kept in the set of targeted substances as being informative of the state of contamination of water resources.

The effectiveness of the internal standards in accurately quantifying each analyte was verified through spike recovery measurements, which consistently fell within the 82–112 % range, as recommended by ISO 21253–2:2019 and XP T 90–214 standards. In the absence of the internal standard and considering the stability of the molecules, each sample was further spiked at a mid-range concentration level to account for matrix effects by applying the spike recovery yield of the sample.

Furthermore, a stability study was conducted for various compounds to evaluate the risk of sample degradation prior to analysis. The stability criterion applied corresponds to the measurement uncertainty of each compound. Over a 12-month storage period, no signs of compound instability were observed in the working solutions at 4 °C.

2.6. Quality assurance / quality control

To assess initial system performance and detect any loss of sensitivity during analytical runs, various quality control measures were implemented. Each target compound's identity was verified in accordance with ISO 21253–1:2019 criteria, including retention time within a 2.5 % tolerance, monitoring of two distinct transitions, and an abundance ratio within a 30 % tolerance of calibration samples (International Organization for Standardization, 2019). Standard mixtures were injected approximately every 10 samples, and a solution at the LQ was run at the end of each sequence to check for drift. Analytical results were evaluated using control charts with acceptance criteria set below 30 %.

To evaluate method accuracy and monitor matrix effects, relative recovery was tested in each analytical run by spiking one RW and one DW sample with targeted analytes. Acceptable recoveries ranged between 60 % and 140 %, following SANTE/11813/2017 guidelines (EURL, 2017). Additional quality assurance was provided through external proficiency testing, with participation in inter-laboratory comparisons at least twice annually. Results confirmed the method's reliability with Z scores below 2 ($-2 < z < 2$).

No cross-contamination was observed during method validation. Nonetheless, each analytical batch included negative controls using LC–MS grade water processed throughout the entire workflow, ensuring contamination remained below one-third of the LQ.

Compound stability was assessed prior to field sampling via spiking experiments in both RW and DW over a 3-week period under defined storage conditions, and post-extraction stability was evaluated for up to 3 months at 4 °C in amber vials. Considering the uncertainty range, all

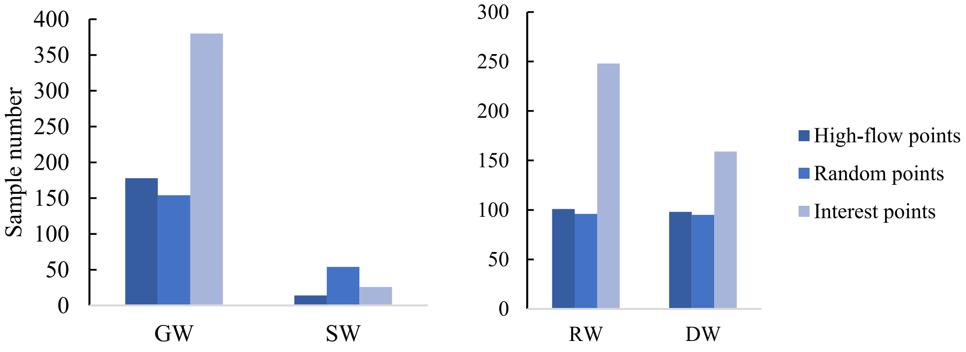

Fig. 2. Repartition of the collected water samples per type and sampling point.

were found to be stable under the defined conditions.

It is worth noting that 39 molecules were processed according to NF EN ISO/IEC 17025 requirements (under Cofrac Accreditation No. 1–2255, scope available at www.cofrac.fr). The remainder (15 molecules) were typically not accredited due to some methodological robustness issues, challenges in obtaining analytical standards, or compound stability limitations. However, they were kept for qualitative rather than quantitative information.

Overall, the validated methods were considered as fit for purpose and were successfully applied in the national monitoring campaign.

2.7. Data analysis

Statistical analyses were conducted using R (Version 4.3.2) correlated with RStudio (Version 2023.12.0 + 369). The Pearson correlation analysis was utilized to evaluate potential correlations among the different compounds and with ClO_4^-. Correlations (r) over 0.5 were considered good and statistical analyses were considered significant for

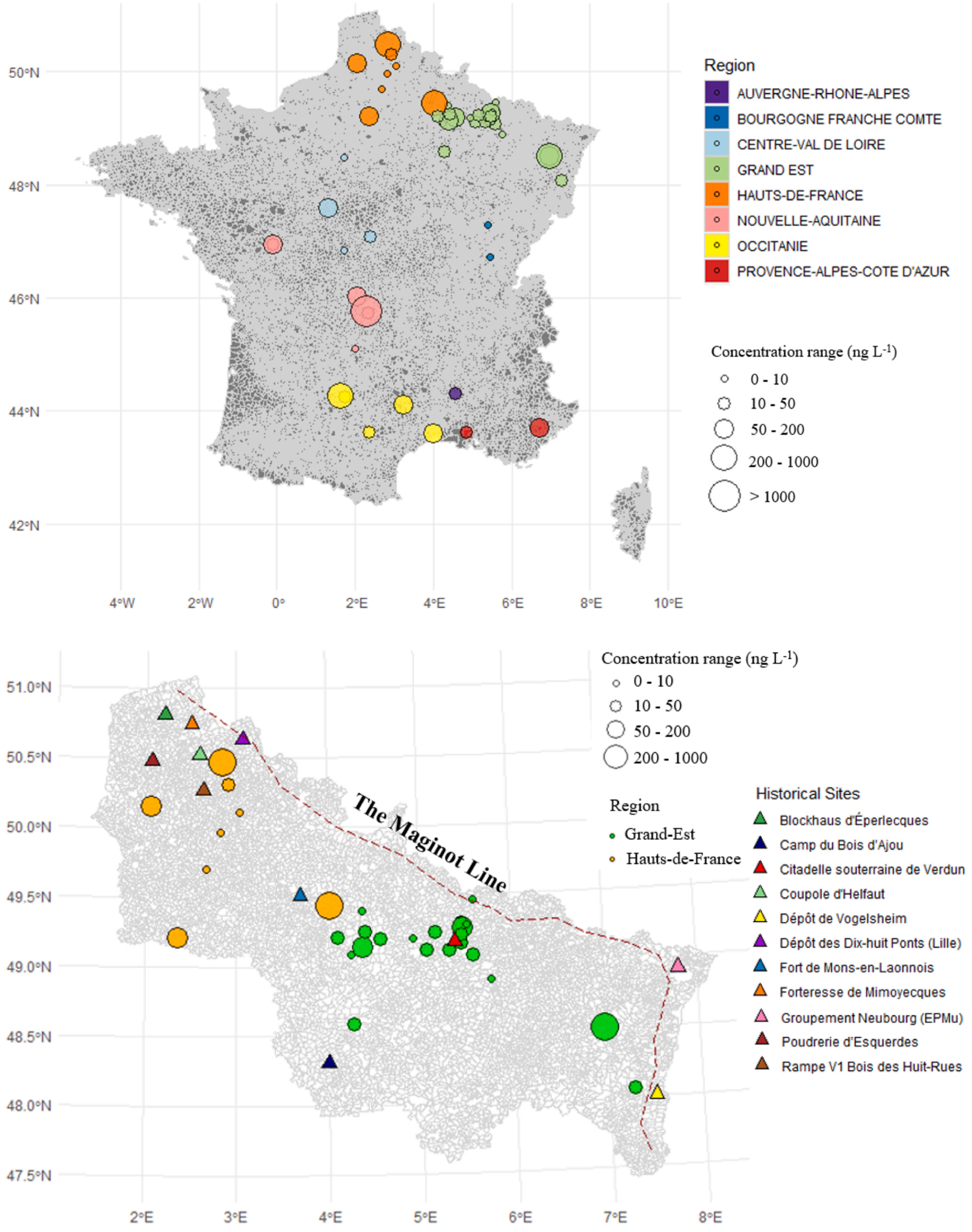

Fig. 3. Spatial distribution of positive samples across France (upper figure), with a zoom on *Hauts-de-France* and *Grand-Est* (lower figure) showing their total concentrations along the most notable historical sites and ammunition production and storage.

p-values below 0.05.

3. Results and discussions

In consideration of the study objectives and the inherent limitations of the sampling framework, data were evaluated at the national scale to capture overarching patterns, rather than disaggregated into regional assessments. Site-specific paired analyses of raw versus drinking water were also not undertaken, and treatment efficacy was therefore not addressed. Such evaluations are methodologically complex, particularly in systems where DW production integrates multiple mixtures of RW, thereby confounding the attribution of treatment performance.

3.1. Explosive residues quantified in RW

3.1.1. Frequency and occurrence

Out of the 54 molecules analyzed, 18 were quantified at least once (33 %). In 13 % of the analyzed samples (58 out of 445), at least one compound was quantified at a concentration higher than the LQ. The majority of positive results in RW were associated with raw GW, representing around 91 % (53 samples) compared with 9 % (five samples) from SW intakes. Some of the analyzed compounds were classified either as persistent, mobile and toxic (PMT) or as very persistent and very mobile (vPvM) [6]. This can explain their frequent and more widespread occurrence in GW compared with SW (very mobile in water). It is also interesting to note that 89 % of positive RW samples came from IPs selected by the ARS, whereas only 7 % were from HFs and 4 % from RPs.

Fig. 3 presents the spatial distribution of positive samples across France (top), with a detailed view of *Hauts-de-France* and *Grand Est* (bottom), highlighting total concentrations (ng L^{-1}) in proximity to significant historical sites as well as former ammunition production and storage facilities. The spatial distribution of explosive compounds across France demonstrates a clear clustering in the northern and eastern regions, especially within *Hauts-de-France* (30 %, 8 out of 27 samples) and *Grand-Est* (23 %, 30 out of 130 samples), which correspond to the main theatres of both World War I and World War II. In both areas, the predominant explosives identified were DNT and NT isomers, as well as ADNT (historical substances), with a very few presence of the modern RDX (3 out of 38 positive samples, 8%). The median values of the cumulative concentrations of quantified explosive residues were also the highest, 51.8 and 17.9 ng L^{-1}, respectively in *Hauts-de-France* and *Grand-Est*. These hotspots coincide with former frontlines, battlefields, and military infrastructures, including artillery storage depots, fortified positions along the Maginot Line, and V-weapon launch bases. Elevated levels quantified in areas such as the *Somme, Aisne, Ardennes, Marne,* and *Meurthe et Moselle* reflect the long-lasting environmental imprint of heavy bombardments and the subsequent burial or abandonment of unexploded ordnance. By contrast, in central and western France the predominant explosive quantified is the modern substance RDX (12 out of 20 positive samples, 60%) whose concentration varied from 12 to 2300 ng L^{-1} (median of 56.5 ng L^{-1}), with a complete absence of historical explosive residues, confirming that contamination is localized and historically driven rather than diffuse. The absence of positive samples in the overseas territories is primarily due to their lack of direct exposure to major wars and limited military activities. This pattern underscores the enduring environmental legacy of armed conflict, with explosive residues persisting as measurable risks to ecosystem integrity, agricultural productivity, and human health more than a century after their deposition.

Table 1 summarizes the quantification frequency (QF, percentage of samples with concentrations $\geq$ LQ) and concentration statistics for each compound in quantified samples only. DNT isomers, RDX, and DPA were the most frequently quantified. Median concentrations varied widely, from low single-digit ng L^{-1} up to tens of ng L^{-1}, with very broad ranges for certain compounds (e.g., RDX ranged from 10 to 2300 ng L^{-1}, 2,6-DNT 1–310 ng L^{-1}). This indicates that while most detections were at relatively low levels, a few hotspot samples contained exceptionally high concentrations. It is important to note that the maximal QF exceeded 7 % (for 2,6-DNT) out of the total number of RW samples (positive and negative; n = 445). Other substances were quantified up to a level of 3 % of the total analyzed RW samples. The most frequently quantified compounds were 2,6-DNT (quantified in 52 % of samples) and other DNT isomers (2,4-DNT in 22 %, 2,3-DNT in 21 %, 3,4-DNT in 17 %). RDX was also common (26 % of samples). By contrast, TNT itself was rarely found (quantified in only one sample, QF of 1 %) even though it was one of the most common explosive substances, widely used since World War I and still used in military and industrial applications (mining, quarries). Several lesser-known nitroaromatic by-products (e.g., DNB isomers and 3-NT) appeared only once or not at all. Median concentrations of the most frequently quantified substances were mostly in the 1–56 ng L^{-1} range. Notably, the only detection of TNT was at 68 ng L^{-1} (in a GW well, likely reflecting a TNT-rich source), while RDX and 2,6-DNT showed median values around 8–56 ng L^{-1}. These high outliers suggest point-source contamination (e.g., near ammunition waste or production sites), whereas most samples had much lower concentrations.

3.1.2. Sources, historical context, and matrix-dependent patterns

As mentioned before, the most frequently quantified explosive residues in water resources were DNT isomers (2,6-DNT, 2,4-DNT, 2,3-DNT, and 3,4-DNT), ADNT, 2-NT, DPA, and RDX (modern). Fig. 4 illustrates the boxplot of these explosive residues measured in RW samples. The boxes represent the interquartile range, with the median indicated by the central line, while whiskers extend to values within 1.5 times the interquartile range. Red points correspond to individual measurements.

Table 1
QF (%) and concentration statistics (ng L^{-1}) for explosive residues in quantified RW samples.

Compound	QF (GW+SW) (n = 58)	QF GW (n = 53)	QF SW (n = 5)	LQ	Median	Range
©2,6-DNT	53	57	–	1	8	1 – 310
©RDX	26	23	60	10	56	10 – 2300
©2,4-DNT	22	25	–	5	11	5 – 26
©2,3-DNT	21	23	–	2	10	3 – 130
©3,4-DNT	17	19	–	5	16	9 – 150
ADNT	16	17	–	10	32	11 – 160
©2-NT	12	13	–	2	4	3 – 15
©DPA	12	13	–	5	12	6 – 18
©ClNB	3	2	20	2	6	3 – 9
HMX	3	4	–	10	49	31 – 66
©2-NP	3	–	40	20	51	23 – 79
©4-NT	3	4	–	5	7	5 – 10
©1-NN	2	2	–	2	2*	–
©1,2-DNB	2	2	–	20	22*	–
©1,5-DNN	2	2	–	1	1*	–
©2-NA	2	2	–	10	13*	–
©TNT	2	2	–	10	68*	–
©3-NT	2	2	–	10	12*	–

ADNT= 2-A-4,6-DNT+ 4-A-4,6-DNT; ClNB= 1-Cl-2-NB+ 1-Cl-4-NB; ©: Accredited by Cofrac; * indicates a single concentration value reported for the compound, as it was detected only once.

DNT exists as six isomers, 2,4- and 2,6-DNT being the two major forms; 2,3-DNT, 2,5-DNT, 3,4-DNT, and 3,5-DNT are minor isomers. Tg-DNT refers to a mixture of DNT isomers (about 76.5 % 2,4-DNT, 18.8 % 2,6-DNT, and 4.7 % minor isomers (2.43 % 3,4-DNT, 1.54 % 2,3-DNT, 0.69 % 2,5- DNT, and 0.04 % 3,5-DNT). DNT is not found naturally in the environment. It is most commonly produced by mixing toluene with sulfuric and nitric acids. The mixture of DNTs is sold as an explosive and is a raw material for the production of 2,4,6-TNT. Tg-DNT is also used as a modifier for smokeless powders in the munitions industry, in automotive airbags, and as a chemical intermediate for the production of toluene diisocyanate (TDI), dyes, and polyurethane foams [3,34,53]. In

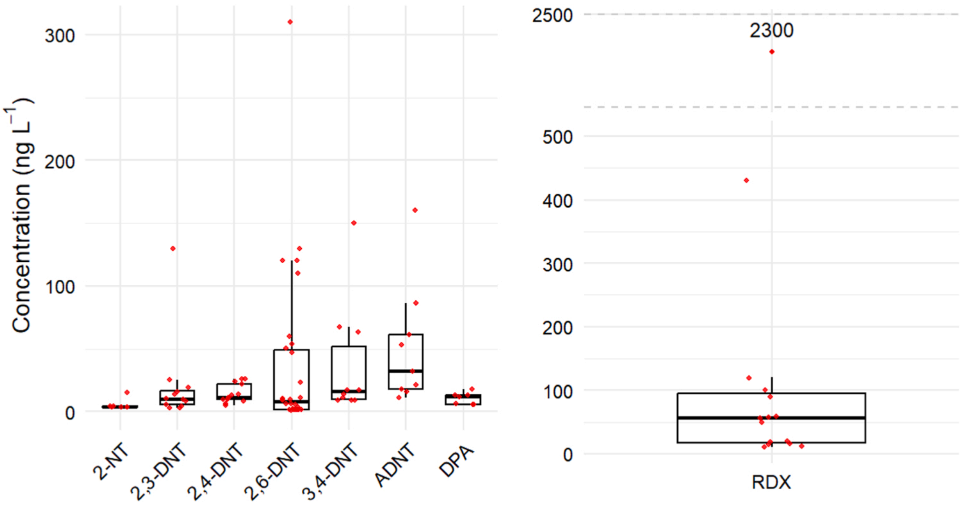

Fig. 4. Concentration boxplots (ng L^{-1}) of the most quantified explosive residues in RW samples.

the current study, 2,6-DNT was by far the most prevalent, quantified in over half of positive samples (53 %). 2,6-DNT is a component of technical DNT (used in polyurethane manufacturing) and a minor impurity in TNT production [36,46]. Its frequent occurrence suggests widespread historical contamination from munitions production or industrial waste. Despite its high QF, concentrations were usually modest (median 8 ng L^{-1}) with one striking exception at 310 ng L^{-1}. The ubiquity of 2,6-DNT indicates that this compound persists and readily migrates. Likely sources include former TNT manufacturing plants and explosive waste dumps where 2,6-DNT leached into water over time. During the 20th century, especially the World Wars, millions of tons of TNT were made (e.g., > 1.6 million tons in Germany during World War II) [64], generating waste streams rich in DNT isomers and other by-products. These waste residuals have had decades to leach into soils and different types of water. 2,6-DNT's moderate water solubility (~180 mg L^{-1} at 20 °C), moderate octanol-water partition (log Kow ~2.1), and low organic carbon partition coefficient (log Koc ~1.7) favor its mobility in aquatic environments [46]. This means that it adsorbs only weakly to soils, sediments or activated sludge. As a result, 2,6-DNT remains mobile in aquatic environments – it does not readily bind to soil and can leach into GW, aligning well with our observations (57 %). In oxygenated surface waters, 2,6-DNT can be broken down by photolysis (sunlight-driven degradation) and other oxidative processes; experiments show nearly complete photodegradation within ~3 days under strong simulated sunlight. However, in dark or low-oxygen environments (e.g., GW), it is persistent–it undergoes little natural attenuation in the absence of light (only ~3 % degraded in 4 days without light). Some microorganisms can biodegrade DNT via reduction or oxidation, but this often yields intermediate products (e.g., ADNT) rather than complete mineralization. Overall, unless actively broken down by sunlight or specialized treatment, 2,6-DNT can remain in water for long periods and migrate with GW flow. 2,4-DNT was also frequently quantified in almost 22 % of RW samples only in GW. 2,4-DNT–the major component of technical DNT (Tg-DNT)–appeared in nearly a quarter of samples (median 11 ng L^{-1}). Its maximum concentration (26 ng L^{-1}) was much lower than that of 2,6-DNT, yet its median level was a little bit higher than 2,6-DNT. Its QF was a bit lower than 2,6-DNT's, but still significant, suggesting 2,4-DNT contamination occurred in many of the same contexts as 2,6-DNT. The lower frequency could imply that either 2,4-DNT is slightly less prevalent in sources or that it is more readily attenuated in the environment than 2,6-DNT. The relatively narrow concentration range may reflect a more diffuse origin, with widespread low-level inputs, as well as other environmental factors (perhaps limited solubility or stronger adsorption) that impose an upper limit on its presence in water resources. It has moderate solubility (~300 mg L^{-1}) and low volatility, so it remains dissolved in water rather than volatilizing. Its adsorption to soil and sediment is low (log Kow ~1.98, log Koc ~1.65), meaning that 2,4-DNT is quite mobile in aquifers and rivers–it can leach through soil into GW and travel downstream in SW [2,34]. In terms of degradation, 2,4-DNT shares pathways similar to those of 2,6-DNT. It can undergo photolysis in sunlight (with reported half-lives of just a few hours in surface waters under strong sun, even faster than 2,6-DNT in some tests. 2,4-DNT is slightly more amenable to natural attenuation than 2,6-DNT, but it is still a mobile and persistent contaminant in many settings. Other DNT isomers, including 2,3-DNT and 3,4-DNT (minor isomers in technical DNT), were slightly less frequent, but still found in 17 % and 21 % of positive RW samples (medians ~10–16 ng L^{-1}). Both showed occasional spikes (up to 130 ng L^{-1} for 2,3-DNT and 150 ng L^{-1} for 3,4-DNT), indicating localized hotspots. 2,3-DNT and 3,4-DNT behaved similarly to 2,4- and 2,6-DNT, with low moderate water solubility (respectively, 220 and 180 mg L^{-1}), low volatility, moderate hydrophobicity (log Kow~2), and moderate adsorption to soil (log Koc ~2.5), so they can leach through soils into GW in much the same way as 2,4-DNT or 2,6-DNT over decades [36]. Thus, they can remain there unless actively treated or degraded.

RDX was also quantified in about 26 % of the RW samples (23 % in GW vs. 60 % in SW), making it the most prevalent modern explosive nitramine in the dataset. This could point to either historical, recent, or ongoing surface discharges into water resources, perhaps from industrial outfalls or contaminated runoff. The median RDX concentration among detections was 56 ng L^{-1}, with a reported range from 10 ng L^{-1} up to 2300 ng L^{-1}. However, the maximum of 2300 ng L^{-1} is a clear outlier, indicating a significant hotspot of RDX in one sample. Such a level points to a severe local contamination (for instance, a water source impacted by historical or modern contamination [10]. It is a highly potent military explosive that was widely manufactured and deployed in warheads (present in > 4000 munitions designs) [27,28,37]. RDX is fairly water-soluble (~60 mg L^{-1} at 25 °C) and only mildly hydrophobic (log Kow~0.9). Consequently, it does not sorb strongly to soils or aquifer materials (log Koc~1.8), and tends to readily migrate with SW and GW. Field experience at military sites confirms that RDX plumes can travel long distances in aquifers, as low soil retention allows leaching into underlying GW (Pitchel, 2012, [4,32]). In general, RDX has been observed to have environmental half-lives in the order of hundreds of days in subsurface environments, making it effectively persistent if not exposed to sunlight or engineered treatment.

ADNTs (2-A-4,6-DNT and 4-A-2,6-DNT) are major TNT degradation products [67]. The absence of quantified TNT in both GW and SW, coupled with the presence of ADNT, indicates that TNT is extensively transformed under environmental conditions (especially in GW under anaerobic conditions). GW is often low in oxygen and high in organic matter, so it favors such reductive transformations, explaining why we see ADNT in several wells but almost no TNT. SW, being oxygenated, might oxidize TNT or flush it out faster; indeed, we did not detect TNT in any surface sample [52]. For instance, whenever TNT is exposed to reducing conditions (e.g., in anoxic GW or during biological metabolism), a substantial fraction is converted to 2-ADNT and 4-ADNT [51,69]. The presence of ADNTs thus appears to be a signature of historical TNT use. Indeed, TNT itself was rarely seen (only once), but ADNT metabolites were more frequently found (17 % in GW). The median level of ADNT was 32 ng L^{-1} (with a maximum of 160 ng L^{-1}). These metabolites have moderate water solubility (~0.006 mg L^{-1}) as well as a moderate soil adsorption coefficient (log Koc ~2.2). This combination means that ADNT can leach or undergo runoff after a rain event into GW and SW [59,67]. Importantly, the presence of dissolved organic matter (DOM) in water could complicate ADNT detection. ADNT may bind to soluble organic matter (SOM) or particulate organic matter (POM), making a fraction of it invisible to conventional analytical methods while still persisting in aquatic systems. This binding behavior may contribute to underestimation of ADNT concentrations in monitoring programs. Upon human exposure, these bound residues may be released, prolonging toxicological effects and raising health concerns. Therefore, water sampling around former manufacturing sites, military ranges, or areas with known explosive residues should prioritize the analysis of TNT in both shallow aquifers and surface runoff, especially after rainfall events or flooding. The presence of ADNT confirms TNT use, with the TNT extensively transformed to ADNT over time. For example, one GW in this study contained 68 ng L^{-1} TNT and 160 ng L^{-1} of ADNT. The finding that ADNTs persist in water resources aligns well with reports that TNT metabolites can be as problematic as TNT itself in the environment [55].

2-NT was the least frequently quantified of the top eight compounds, with a QF of 12 %. This low QF suggests that its contamination is isolated, likely occurring only near specific industrial sites or as a transient breakdown product of other chemicals rather than being a widespread groundwater contaminant. Its median concentration was 4 ng L^{-1}, with a range from 3 ng L^{-1} to 15 ng L^{-1}. Its relatively infrequent detection could be due to several factors. In fact, 2-NT is a starting material/intermediate in manufacturing explosives (and dyes), but not a final product that persists; it may also be more readily lost or transformed in the environment than DNT. Its occurrence further suggested either incomplete TNT/DNT synthesis or environmental breakdown of DNT, since NT can form via partial reduction of DNT. In terms of mobility, 2-NT is moderately soluble (~500 mg L^{-1}) with a log Kow~2.3, and has some affinity for organic carbon in soil (log Koc ~2.2). This indicates that 2-NT is less mobile than other substances but still mobile enough to reach GW, especially through soils with low organic content. Some fraction might sorb, slowing its travel. With respect to persistence, 2-NT exhibits a dual nature: it is not readily biodegradable in standard tests. Laboratory studies have shown that 2-NT tends to resist quick breakdown; one screening assessment found that the biodegradation half-life in water was likely ≥ 182 days). The highest 2-NT concentration was 15 ng L^{-1} in one GW sample. 2-NT is used in organic synthesis and was historically a gelatinizer in some propellants, so possible contributors include industries (dye or chemical plants) or propellant manufacturing sites [37]. In Germany, 2-NT was quantified in SW near former munitions plants at levels up to 22,000 ng L^{-1}. In the USA, it was present in both GW (up to 140,000 μg L^{-1}) and SW (up to 120 ng L^{-1}) at multiple munitions production sites. Additionally, it was quantified in GW at a military training site with a maximum concentration of 25,000 ng L^{-1} [50].

DPA was also least quantified in 12 % of positive RW samples (median 12 ng L^{-1}) with a range from 6 ng L^{-1} up to 18 ng L^{-1}. DPA is not itself an explosive, but is widely used as a stabilizer in propellants and explosives (for example, in older munitions, it was added to nitrocellulose gunpowder to prevent degradation) [29]. Its presence in water therefore signals contamination from propellant residues or munitions manufacturing waste. The moderate detection frequency suggests that DPA contamination is sporadic–likely confined to areas near specific sources (such as old artillery ranges, ammunition depots, or factories where propellants were made or disposed of [45]. DPA has limited water solubility (~25 mg L^{-1} at 20 °C) and is fairly hydrophobic (log Kow ~3.3). Indeed, its mobility in soil is variable: in clay-rich or organic soils it is “somewhat mobile” (higher adsorption), whereas in sandy or weakly organic soils it remains quite mobile. Based on adsorption and desorption characteristics, its mobility varies depending on soil type. It is considered slightly mobile in clay soil, with a log Koc ~2.2. In contrast, it is more mobile in other soils, with log Koc ~1.33 in loamy sand, ~1.13 in loam, ~0.7 in silt loam, and ~1.21 in silty clay loam sediment [30]. These properties reflect its tendency to adsorb to soils and organic matter.

Other explosive residues such as NB and chloronitrobenzene (ClNB) isomers (1-Cl-2-NB, etc.) are intermediates in the dye and pesticide industries [42,43]. The low-frequency detection of ClNB (3 %) and 1-NN (2 %) suggested isolated instances of industrial pollution, possibly near chemical factories or legacy waste dumps. These compounds are less directly tied to explosive fillers, but could co-occur if the production of explosives involved chemical synthesis steps (for example, ClNB can be an impurity in picric acid production) (Hong et al., 2015).

3.1.3. Comparison with previous studies

Many studies investigating contamination by explosive substances have focused on well-known, highly contaminated sites, particularly former military or industrial areas. In contrast, few have aimed to provide a broader, more representative overview of environmental contamination by those emerging contaminants. Current literature has reported significant levels of compounds such as RDX and DNT isomers near such point sources. For instance, DNT isomers have been previously identified in both SW and GW near sources such as former munitions facilities. In a small brook and the Losse River near an old ammunition plant in Germany, for example, concentrations of 2,4-DNT and 2,6-DNT ranged from 500 to 13,000 ng L^{-1} and 100–7600 ng L^{-1}, respectively. In two ponds situated within a decommissioned munitions site in Germany, 2,4-DNT concentrations were between 800 and 1200 ng L^{-1}, while 2,6-DNT levels varied from 70 to 300 ng L^{-1}. Additional monitoring at three contaminated sites along the Elbe River found 2,4-DNT levels between 100 and 1300 ng L^{-1} and 2,6-DNT concentrations ranging from 80 to 500 ng L^{-1} [3]. At former conflict zones in Germany, Sohr et al. [63] reported 2,4-DNT concentrations of 700 ng L^{-1} and 2,6-DNT concentrations of 3100 ng L^{-1}, while sites with less contamination showed 28 ng L^{-1} and 19 ng L^{-1} for 2,4-DNT and 2,6-DNT, respectively [63]. In Dokai Bay, Japan, water samples collected near industrial discharge zones revealed 2,4-DNT levels ranging from non-detectable (n.d.) to 206,000 ng L^{-1} and 2,6-DNT from n.d. to 14,800 ng L^{-1} [47]. 2,3-DNT was also found in these waters, albeit at much lower concentrations, up to 412 ng L^{-1}. Furthermore, multiple DNT isomers, including 2,3-DNT, 2,5-DNT, 3,4-DNT, and 3,5-DNT were found when monitoring wells, including some private wells, near the Badger Army Ammunition Plant in Wisconsin [3,34]. Besides, RDX has been detected in GW samples collected near munitions factories in the USA and Germany, with concentrations reaching up to 14,100 μg L^{-1} in the USA and ranging from 21 to 3800 μg L^{-1} in Germany [12,41,68,8]. In 2011, the US Army reported RDX concentrations in GW ranging from 50 to 18,000 μg L^{-1} and in SW from 80 to 120 μg L^{-1} near the Milan Army Ammunition Plant. Furthermore, near a munition storage depot in Oregon, RDX concentrations were found to reach up to 8160 μg L^{-1} [8]. This compound was also identified at a concentration of 21 μg L^{-1} in a water sample collected from a military training site in Germany [41]. GW samples were collected between May 1994 and August 2004 from monitoring and extraction wells at the Naval Base Kitsap. RDX concentrations in perched zone monitoring wells varied from 0.19 to 1000 μg L^{-1}, while concentrations in shallow aquifer monitoring wells ranged from 0.19 to 550 µg L^{-1}. In extraction wells targeting the shallow aquifer, RDX levels ranged between 0.4 and 660 µg L^{-1}. At another site, which includes a former wastewater lagoon and overflow ditch, RDX concentrations in shallow aquifer GW samples ranged from 0.95 to 3800 µg L^{-1} [70]. Overall, other findings underscore that the most available data reflect contamination near known point sources, often showing high concentrations. In contrast, our results reflect much lower levels, consistent with more diffuse or residual contamination patterns in less impacted areas.

3.2. *Explosive residues quantified in DW*

3.2.1. *Frequency and occurrence*

Out of the 54 molecules analyzed, 17 were quantified at least once (31 %). In 9 % of the analyzed samples (31 out of 352), at least one compound was quantified at a concentration higher than the LQ. The majority of positive results in DW were associated with IPs (65 %), whereas only 20 % came from HFs and 15 % from RPs.

Table 2 summarizes all the compounds that were quantified in DW samples, whereas Fig. 5 illustrates the boxplot of the most quantified explosive residues. For each compound, the QF and concentration statistics (median and range, in ng L^{-1}) were reported. In DW, the most frequently quantified explosive substances were RDX (in 26 % of samples), 2,6-DNT (42 %), 2,4-DNT (23 %), 2,3-DNT (19 %), DPA (19 %), 3,4-DNT (19 %), and 2-NT (13 %). Other measured substances (such as HMX, ADNT, ClNB, DNB, TNT, etc.) were rare, typically quantified only once or twice in positive DW samples. It is important to note that the overall QF exceeded 4 % (for 2,6-DNT) of the total number of samples (positive and negative, $n = 352$). RDX, other DNT isomers, and DPA were quantified up to 2 % in the overall DW samples. Among the most quantified explosive residues, RDX, 2,6-DNT, and 2,4-DNT exhibited the highest median concentrations: RDX ranged from 14 to 850 ng L^{-1} (median 23 ng L^{-1}), 2,6-DNT from 1 to 400 ng L^{-1} (median 12 ng L^{-1}), and 2,4-DNT from 5 to 26 ng L^{-1} (median 13 ng L^{-1}). Other substances (DNT isomers, 2-NT, and DPA) generally had lower medians, with upper ranges usually < 100 ng L^{-1} except for a few outliers (180 and 220 ng L^{-1}, respectively, for 2,3-DNT and 3,4-DNT). These patterns indicate that nitroaromatic compounds and related metabolites were sporadically present in DW, typically at low ng levels. When present, some compounds such as RDX and DNT occasionally reach high values (400 and 800 ng L^{-1}, respectively) pointing to local contamination hotspots. However, the majority of quantifications were low (e.g., highest median of 23 ng L^{-1} for RDX). Other residues mostly had medians varying between 5 and 12 ng L^{-1}. For less frequently quantified substances, some median values were unusually high (80 ng L^{-1} for HMX). However, no conclusion can be drawn about such an occurrence due to their very low QF (once or twice).

Table 2
QF (%) and concentration statistics (ng L^{-1}) for explosive residues in quantified DW samples.

Compound	QF ($n = 31$)	LQ	Median	Range
©2,6-DNT	42	1	12	1 – 400
©RDX	26	20	23	14 – 850
©2,4-DNT	23	5	13	5 – 26
©2,3-DNT	19	2	10	3 – 180
©3,4-DNT	19	5	10	8 – 220
©DPA	19	5	10	5 – 27
©2-NT	13	2	5	3 – 23
HMX	6	10	57	34 – 80
©2-NPh	6	20	34	25 – 42
ADNT	6	10	31	12 – 50
©ClNB	3	2	4*	–
©1-NN	3	2	9*	–
©1,2-DNB	3	20	23*	–
©1,3-DNB	3	10	11*	–
©TNT	3	10	28*	–
©3-NT	3	10	18*	–
©4-NT	3	5	15*	–

ADNT= 2-A-4,6-DNT+ 4-A-4,6-DNT; ClNB= 1-Cl-2-NB+ 1-Cl-4-NB; ©: Accredited by Cofrac; * indicates a single concentration value reported for the compound, as it was detected only once.

3.2.2. *RW vs. DW levels*

To assess removal treatment, we compared DW concentrations with those in source water (RW) from the previous datasets. The results clearly indicated that almost all substances were present in both RW and DW, at a near identical QF. A minor decrease was noted for 3,4-DNT and DPA from 16 ng L^{-1} (QF of 17 %) to 10 ng L^{-1} (QF of 19 %) and from 12 ng L^{-1} (QF of 12 %) to 10 ng L^{-1} (QF of 19 %), respectively. 2,3-DNT and 2-NT had almost the same median levels in both matrices (i.e., 10 and 5 ng L^{-1}, respectively) with almost the same QF (20 % and 12 %). Compounds such as 2,6-DNT and 2,4-DNT exhibited a slight increase (from 8 to 12 ng L^{-1} and 11–13 ng L^{-1}, respectively), indicating partial resistance to treatment or potential desorption from the filter media. A significant reduction was noted for RDX (log Kow~0.9) post-treatment from 56 ng L^{-1} (QF of 26 %) to 23 ng L^{-1} (QF of 26 %) with a lower maximal level (850 vs. 2300 ng L^{-1}). ADNT, a transformation product of TNT, also showed similar levels pre- and post-treatment, but the QF sharply decreased from 16 % to 6 %. Other quantified substances with a QF below or equal to 6 % had almost the same median levels in both matrices, but the conclusions remained unclear. Overall, these data suggest that while low-level presence is common, there are isolated instances of high contamination, possibly near point sources or where conventional DW treatment (likely coagulation/filtration/disinfection) only partially attenuates explosive residues and may even spike some residues. Advanced treatments (e.g., activated carbon or advanced oxidation) are generally required to target such compounds, which are not fully handled by standard processes [22,61]. In fact, some of the quantified substances in DW—such as 2,3-DNT, 2,4-DNT, 3,4-DNT, and 2-NT—are considered PMT, while others—such as 2,6-DNT and RDX—are considered vPvM [5]. PMT/vPvM substances encompass a broad group of chemicals characterized by their presence in the environment, high mobility, and potential risks to human health and ecosystems. Such substances are somewhat challenging to remove during DW treatment processes and thus pose a threat to DW resources [56]. For instance, numerous PMT and vPvM substances are capable of bypassing conventional barriers employed in drinking water treatment plants (DWTPs). This includes advanced technologies such as granular activated carbon (GAC) filtration, ultrafiltration, advanced oxidation processes (e.g., ozonation), and even reverse osmosis [62,71]. Stackelberg et al. [65] observed that despite the use of clarification, chlorination, and GAC filtration, a significant proportion (up to 75 %) of mobile substances remained in the treated effluent. This suggests that where continuous emissions of PMT/vPvM compounds coincide with suboptimal removal in treatment facilities, concentrations may progressively accumulate. Over time, this can lead to persistent circulation of these substances within the water cycle, potentially resulting in long-term and irreversible contamination [39,65,66]. Therefore, the combination of persistence and great ability to disperse in water may explain the high frequency of 2,6-DNT and RDX in finished water, highlighting their PMT- and vPvM-like behavior. The current study's main focus was not on the removal of explosive residues in DWTP. Additional sampling and in-depth investigations—including analysis of treatment processes and consideration of residence time—would therefore have been required to better understand the compounds' behavior.

3.2.3. *Comparison with guidelines*

To date, neither European nor national regulations specify permissible levels for explosive residues in water intended for human consumption. However, various organizations have established guideline values for certain substances. Guideline values in water intended for human consumption are shown below. These were issued by various regulatory and advisory bodies, including the Agency for Toxic

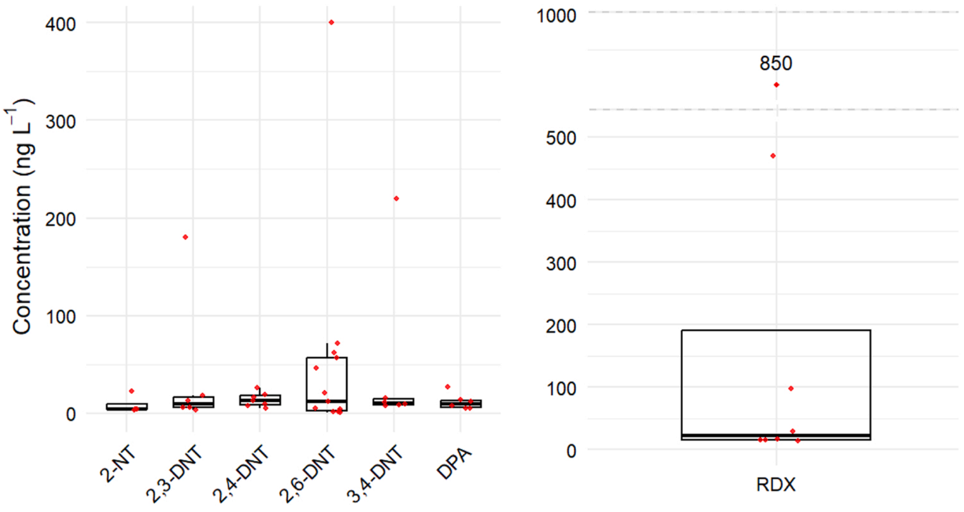

Fig. 5. Concentration boxplots (ng L^{-1}) of the most quantified explosive residues in DW samples.

Substances and Disease Registry [7,60], the Environmental Protection Agency [33], and the Wisconsin Department of Natural Resources [73]. According to EPA [33], the guideline values (drinking water equivalent level, DWEL) for 2,4-DNT and 2,6-DNT are 100,000 and 40,000 ng L^{-1}, respectively. The guideline value for 2-A-4,6-DNT is 1000 ng L^{-1}, whereas those for 2-NT, 3-NT, and 4-NT are 150, 200,000 and 2000 ng L^{-1}, respectively (Wisconsin (2017)). For HMX, the guideline value is 400,000 ng L^{-1} according to the ATSDR [7] and Wisconsin (2017), and 2000,000 ng L^{-1} according to EPA [33]. The guideline or DWEL value for RDX is 100,000 ng L^{-1} by Health Canada (2020) and EPA [33], while a much lower guideline value of 2000 ng L^{-1} is set by Wisconsin (2017). In all cases, these values far exceed the measured concentrations in our samples. For example, the highest level for 2,4-DNT in DW was 26 ng L^{-1}, well below the EPA guideline value of 100,000 ng L^{-1}. Likewise, the 2,6-DNT maximum (400 ng L^{-1}) is several orders of magnitude below 400,000 ng L^{-1}. The one high RDX value (850 ng L^{-1}) demonstrated that in certain cases, RDX can reach concentrations approaching the health advisory guideline value (2000 L^{-1}) (Wisconsin (2017)). The highest level of HMX (80 ng L^{-1}) observed in our data is also far from those set by the regulatory and advisory bodies. For the remaining substances (2,3-DNT, 3,4-DNT, 2-NT, ADNT, and DPA), there are no specific WHO/EU/ANSES DW standards. EPA in the USA does not list guidelines for these; e.g., DPA is not on the EPA DW advisory list [35]. In the absence of formal limits, we note that all the observed levels are extremely low (typically <100 ng L^{-1}, except for a few hotspot values). For reference, a few US states have set very high DW limits for DPA (e.g., 200,000 ng L^{-1} in New Jersey), orders of magnitude above our values [57]. Thus, from a health standpoint, the measured concentrations of nitrotoluene isomers and DPA in DW appear well within any plausible safety threshold. In summary, the observed concentrations fall far below known health advisory levels. This indicates that, despite their persistence, the current levels of explosive residues in DW samples are generally within the bounds of safety set by available guidelines.

3.3. *Potential of ClO_4^- as a marker for contamination by explosive residues*

ClO_4^- ions were analyzed to highlight a possible correlation with nitroaromatic compounds, and to assess whether this chemical can serve as a sentinel indicator for contamination by explosive residues. Overall, it was quantified in 127 samples out of 797 (16 %), and in only 37 samples out of 89 positive samples (40 %). Table 3 summarizes ClO_4^- quantification rates among samples containing historical versus modern explosive residues. The table showed that it co-occurred frequently with historical substances (55 % of those samples), but was almost absent in the majority of positive samples containing modern explosive substances (4 % of those samples).

We selected the historical substances (nitroaromatics) and computed all pairwise Pearson correlations, considering only correlations with $p < 0.05$ as significant. The clustered heatmap, shown in Fig. 6, highlights different patterns. Among historical substances, the strongest positive correlations ($r \sim 0.84$–0.96, $p < 0.001$) occurred between chemically similar NB and NT isomers (e.g., 1,2-DNB vs. 1-NN, or 1-Cl-2,4-DNB vs. 1,3,5-TNB). Nitroaromatic compounds (2-NT, 2,3-DNT, 2,6-DNT, 3-NT, 3,4-DNT, and 4-NT) form a cohesive block ($r > 0.85$, $p < 0.001$), reflecting shared sources such as production residues, formulation practices, or historical disposal of munitions and explosive devices. ClO_4^- appears as a separate cluster but maintains moderate correlations ($r = 0.66$–0.71, $p < 0.001$) with the NT group, consistent with overlapping contamination pathways while retaining distinct origins such as propellant oxidizers. Notably, 2,4-DNT emerges as an outlier, showing only weak-to-moderate correlations ($r = 0.37$–0.53, $p < 0.001$) with both ClO_4^- and other NT isomers. This suggests differences in environmental persistence, degradation, or input sources, reinforcing its unique behavior compared to the broader nitroaromatic cluster. Its isolation in the heatmap emphasizes the importance of considering 2,4-DNT separately from the broader nitroaromatic group. The rest of historical substances analyzed in our study had no significant association with ClO_4^- ($r < 0.1$, $p > 0.05$). Collectively, these results point to two partially overlapping contamination signatures: a strongly interlinked nitroaromatic cluster and a ClO_4^- signal that coincides with, but remains distinct from, the nitroaromatics.

Next, we restricted our analysis to modern explosive substances (RDX, HMX, dinitroanisol (DNAN), etc.). RDX/HMX had many non-detects in this dataset, but we analyzed the available data. Only one pair of modern compounds was significantly correlated: RDX and HMX ($r \sim 0.14$). Crucially, no modern substance showed any significant

Table 3
QF (%) of ClO_4^- (≥ 2 µg L^{-1}) in sample subsets. “Historical” = samples with ≥ 1 historical explosive substance; “Modern” = samples with ≥ 1 modern explosive substance. “n” denotes the number of samples.

Subset	n	ClO_4^- ≥ 2 µg L^{-1}	QF (%)
All samples	797	127	16
Negative samples	686	90	13
Positive samples	89	37	41
Historical substances	66 (out of 89)	36	55
Modern substances	23 (out of 89)	1	4

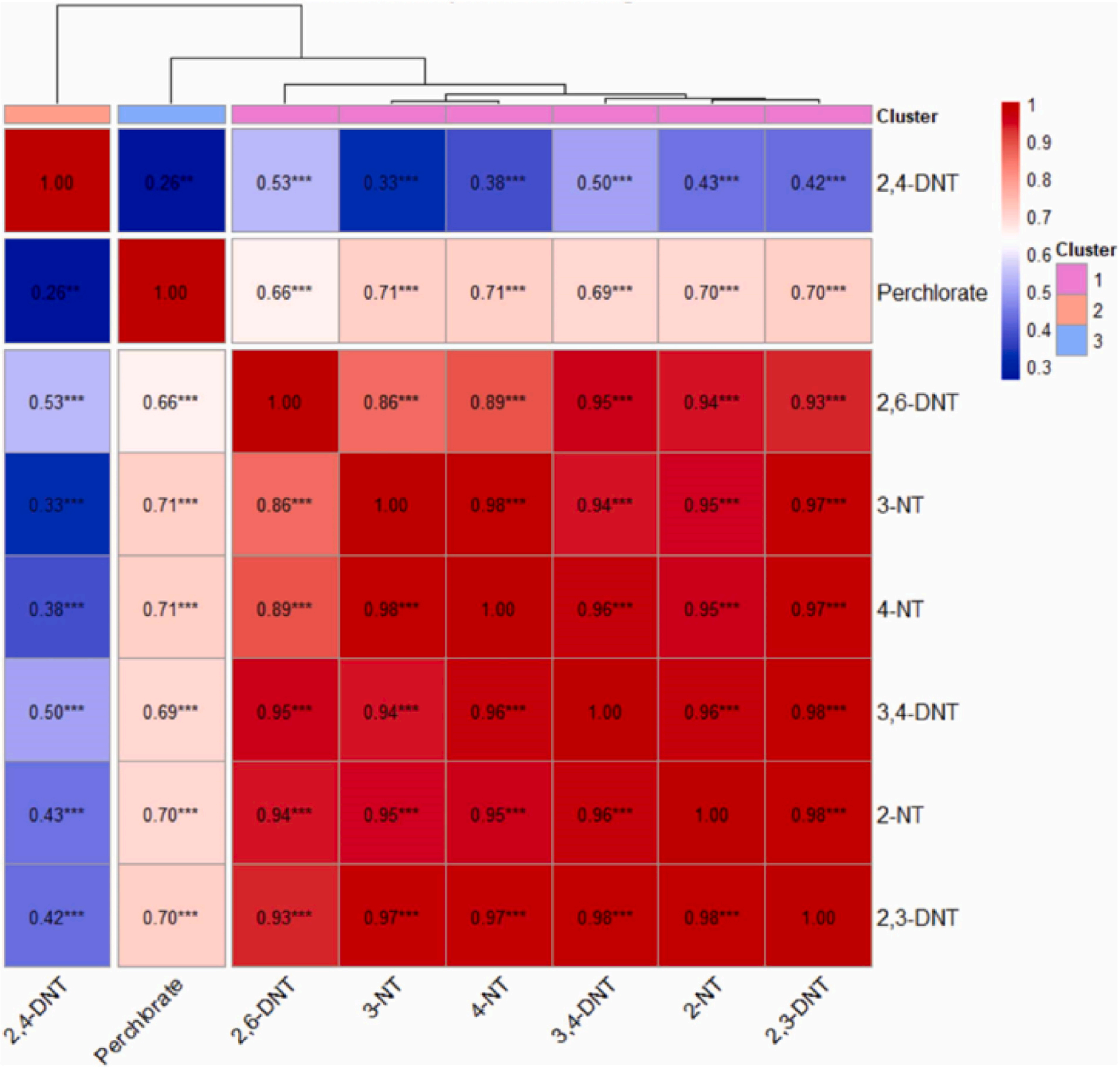

Fig. 6. Clustered heatmap showing correlations between historical explosive residues and ClO_4^- (***$p < 0.001$).

correlation with perchlorate: all pairwise correlations with ClO_4^- were near zero ($r < 0.1$) and statistically non-significant ($p > 0.05$). For example, RDX vs. ClO_4^- gave r ~ –0.02 ($p = 0.63$), DNAN vs. ClO_4^- r ~ 0.06 ($p = 0.09$), and HMX vs. ClO_4^- r ~ –0.02 ($p = 0.64$). The modern correlation heatmap (not shown) is therefore essentially blank in the ClO_4^- row/column. RDX, HMX, DNAN, and other modern markers are inter-correlated to varying degrees (e.g., RDX–HMX), but ClO_4^- stands apart.

Overall, these results showed a marked difference. ClO_4^- co-occurred with historical nitro-explosive compounds but not with modern ones. In the historical subset, it may predict NT contamination (55 % of such samples had ClO_4^-), whereas in the modern subset ClO_4^- was absent. This agrees with prior observations: for example, Walsh et al. [72] found that after detonating PAX-21 (an insensitive munition of RDX + DNAN + ammonium perchlorate), the detonated residue contained a large fraction of ClO_4^- but essentially no RDX/DNAN, with "no correlation between ClO_4^- residues and the RDX and DNAN" [72]. In other words, ClO_4^- contamination from modern propulsion or ignitors leaches independently, while RDX/DNAN residues are nearly wholly consumed. If ClO_4^- is used as a screening indicator for the effects of explosive substances, it will preferentially flag sites with historical ordnance residues (from the World War era) but may miss contamination from modern munitions and energetic materials. This must be considered in environmental and health surveillance: the absence of ClO_4^- does not rule out the presence of modern explosive residues (as shown here), whereas its presence strongly suggests nitroaromatic pollution. However, ClO_4^- can also enter the environment through non-military sources such as fertilizers derived from Chilean nitrate deposits or manufactured perchlorate salts. Indeed, 13 % of our negative samples contained $ClO_4^- \geq 2$ μg L^{-1}, consistent with diffuse agricultural inputs or natural ClO_4^- occurrence rather than explosive residues [24,25,58,74].

4. Conclusion

In summary, our nationwide monitoring indicates that dinitrotoluene (DNT) isomers (particularly 2,6- and 2,4-DNT) and RDX were the most persistent and prevalent explosive residues in raw and drinking water. These compounds were found far more often than other targets. In contrast, trinitrotoluene itself was seldom observed, in agreement with the fact that TNT is rapidly reduced in the environment; instead, its reduction products (aminodinitrotoluenes, or ADNTs) occurred frequently in both matrices. Importantly, standard drinking water treatment (coagulation, filtration, disinfection, etc.) removed only a fraction of these micropollutants given their highly mobile and persistent behavior (similar to PMT or vPvM). Our results were coherent with other global and European field studies. These findings highlight the persistence of legacy contamination in the drinking water system caused by past military and industrial explosives use. Based on our results, monitoring should be prioritized in high-risk region (particularly *Grand-Est*) and near historical contamination sources–for example, former military ranges, munitions factories, ammunition depots, and propellant manufacturing or disposal sites–because they can leach explosive substances into water. Perchlorate may also serve as an early warning sign of contamination by historical compounds, particularly for DNTs. Surveillance programs should target not only parent compounds but also key transformation products (e.g., ADNTs). Furthermore, water utilities should evaluate advanced treatment options (such as granular activated carbon adsorption, advanced oxidation processes, or bioreactor systems) to enhance the removal of explosive residues. This combined approach of targeted monitoring and appropriate treatment upgrades will help mitigate the persistence of explosive residues in drinking water. Future work should also integrate ecological risk assessment approaches, such as species sensitivity distributions, once sufficient ecotoxicity data become available for the most frequently quantified compounds.

Environmental Implication

The occurrence of DNT isomers and RDX, especially in groundwater, stems from historical military and industrial activities. These compounds exhibit high mobility and environmental persistence, classifying some as PMT and/or vPvM substances, and are only partially removed by standard treatment processes. Their presence poses long-term ecological and human health risks, particularly near former battlefields or munitions sites. Perchlorate, frequently co-occurring with nitroaromatic compounds, could serve as an early marker for legacy contamination, underscoring the need for continued monitoring and advanced water treatment solutions.

Ethical Approval

Not applicable

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CRediT authorship contribution statement

Christophe Rosin: Writing – review & editing, Supervision, Project administration, Conceptualization. **Sophie Lardy-Fontan:** Writing – review & editing, Supervision, Conceptualization. **Christelle Pallez:** Writing – review & editing, Validation, Methodology, Formal analysis. **Dani Khoury:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis, Conceptualization.

Consent to participate

Not applicable

Consent to publish

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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. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.139986.

Data availability

Data will be made available on request.

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