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Gadolinium in the environment

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References

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Abstract

The work focuses on gadolinium, whose presence in the environment and food chain results from natural and anthropogenic activities. The work presents Gd as an emerging contaminant with increasing environmental significance. The theoretical part discusses in detail its characteristics and uses, especially in gadolinium-based contrast agents (GdCA) used in magnetic resonance imaging (MRI). Regulations and recommendations for minimising environmental pollution are also discussed. The work also points out its toxic effects on aquatic organisms and food chain contamination. Emphasis is placed on the relationship between environmental conditions and Gd toxicity. The experimental part includes the optimisation of analytical methods and their application for the determination of Gd, utilising ICP-MS and ICP-OES. A significant part of the experiments was also the monitoring of the mass flow balance of GdCA at MRI facilities. The gadolinium anomaly was calculated to identify the entry of Gd into the environment as a result of human activity. The work results demonstrate the presence of Gd in the food chain and its accumulation in various ecosystems. At the same time, they point to the need for thorough control of waste from MRI facilities, which can contribute to the spread of Gd in the environment. This work brings a new perspective on the issue of Gd as an environmental contaminant and offers procedures for its monitoring and assessment of the effects on ecosystem health.

Abstrakt

Práce se zaměřuje na gadolinium, jehož přítomnost v životním prostředí a potravním řetězci je důsledkem jak přírodních procesů, tak antropogenní činnosti. Práce představuje Gd jako potencionální kontaminant s narůstajícím environmentálním významem. V teoretické části je podrobně rozebrána jeho charakteristika a využití, zejména v kontrastních látkách na bázi gadolinia (GdCA) používaných při magnetické rezonanci (MRI). Diskutovány jsou také regulace a doporučení pro minimalizaci jeho environmentálního znečištění. Práce zároveň poukazuje na jeho toxické účinky na vodní organismy a kontaminaci potravního řetězce. Důraz je kladen na vztah mezi environmentálními podmínkami a toxicitou Gd. Experimentální část zahrnuje optimalizaci analytických metod a jejich aplikaci pro stanovení Gd, kdy byla použita ICP-MS a ICP-OES. Významnou částí experimentů bylo také sledování bilance hmotnostního toku GdCA na MRI pracovištích. Byla vypočetna gadoliniové anomálie, která slouží k identifikaci vstupu Gd do prostředí v důsledku lidské činnosti. Výsledky práce prokazují přítomnost Gd v potravním řetězci a jeho akumulaci v různých ekosystémech. Zároveň ukazují na potřebu důkladné kontroly odpadu z MRI pracovišť, která mohou přispívat k šíření Gd v prostředí. Tato práce přináší nový pohled na problematiku Gd jako environmentálního kontaminantu a nabízí postupy pro jeho monitorování a hodnocení vlivů na zdraví ekosystémů.

Keywords

gadolinium, emerging contaminant, food chain, contrast agents, ecotoxicity

Klíčová slova

gadolinium, potencionální kontaminant, potravní řetězec, kontrastní látky, ekotoxicita

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Introduction

Gadolinium, a rare earth element (REE) with unique physicochemical properties, is widely used in healthcare, as a contrast agent for MRI, as well as in industry, and agriculture.

However, anthropogenic gadolinium, primarily from hospital wastewater containing GdCA, poses significant environmental risks. It accumulates in soil and aquatic ecosystems, leading to the gadolinium anomaly, clear evidence of human-induced contamination. A significant issue is the chemical stability of GdCA, which resist degradation in the environment, increasing their potential for bioaccumulation in food chains. Gadolinium accumulation in organisms may adversely affect not only the organisms themselves but also higher trophic levels, including humans.

To mitigate these risks, solutions such as biodegradable contrast agents, advanced wastewater treatment technologies, and reduced GdCA usage are essential. Preventing gadolinium release and developing recycling strategies for rare earth elements are crucial to minimising its environmental and health impacts.

The dissertation comprehensively focuses on the issue of gadolinium in the environment, particularly its entry into ecosystems, food chains, and subsequent effects on living organisms. The objectives of the study were as follows:

1. A literature review aimed at consolidating available scientific knowledge on the environmental and toxicological effects of gadolinium, providing an overview of methodologies and approaches for mitigating its negative impacts on ecosystems and human health.
2. To collect and evaluate available information and theoretical data from the literature on the presence of rare earth elements in food, with a particular focus on gadolinium, and to quantify the share of anthropogenic gadolinium. Based on these findings, to calculate and assess the existence of a gadolinium anomaly.
3. Analysis of gadolinium in commonly consumed foods (flour, rice, carrots, wine) using inductively coupled plasma mass spectrometry and evaluation of anthropogenic influence through the calculation of the gadolinium anomaly. To determine the extent of anthropogenic gadolinium presence in food.
4. To assess the mass flow balance of GdCA at an MRI facility in the East Bohemia region, including an analysis of consumption, waste production, and residual gadolinium in packaging. To evaluate the quantity of unused waste GdCA over a specified time period.
5. To evaluate the environmental impacts of gadolinium on aquatic ecosystems, focusing on its toxicity to freshwater microalgae. To assess ecological risks associated with various forms of gadolinium, including gadolinium-based contrast agents and inorganic gadolinium.

1. Theoretical part

1.1 Gadolinium

Gadolinium (Gd) is one of the emerging contaminants belonging to REE [1]. One of its key properties is paramagnetism and at room temperature, it exhibits ferromagnetic behaviour, which is relatively rare for lanthanides [2, 3]. In nature, gadolinium occurs in very low concentrations [4] and exclusively in the form of mixed minerals together with other REE [5]. Widely used in medicine, it is the basis of GdCA used in MRI diagnostics [6]. In electronics, it is used in the production of phosphors, optical devices, and permanent magnets. In nuclear power, its isotopes are used as neutron absorbers to increase reactor safety [7]. Gadolinium is also used in agriculture as a component of phosphate fertilisers and as a growth stimulant in animal feed [1].

Gadolinium functions as an inorganic blocker, slowing down physiological processes dependent on Ca^{2+} supply and inhibiting the activity of some enzymes due to the similarity of the ion radii with Gd^{3+}. In the body, it binds to phosphates, hydroxides and carbonates, forming insoluble complexes that are rapidly deposited in the liver, kidneys, and reticuloendothelial system, while accumulation in bones occurs more slowly [8]. The acute toxicity of gadolinium can be significantly reduced by chelation with suitable ligands, which prevent its uptake by cells and increase its safety in use. Chelates of gadolinium are the basis for its application in healthcare [9].

1.1.1 Gadolinium-based contrast agents

GdCA are a fundamental tool in MRI, allowing detailed imaging of soft tissues and organs. The first clinical trial using GdCA was conducted in 1984, and since 1988, these agents have become a common part of clinical practice and biomedical research [3, 10, 11]. Despite its indispensability, free gadolinium (Gd^{3+}) in its hydrated form is highly toxic and must be stabilised in complexes with organic ligands to be safe for use in the human body. However, the stability of these complexes is not absolute and can be compromised by processes such as transmetalation, where ions (e.g., Fe^{3+}, Mg^{2+}, Cu^{2+}, Zn^{2+}, Ca^{2+}) in the body replace gadolinium in the complex, leading to the release of free Gd^{3+} [2, 8, 10].

The chemical structure of GdCA has a significant role in their stability and safety [6, 9, 10]. They are divided into linear and macrocyclic ones according to the type of ligands. Linear chelates are formed by molecules with an open structure, which reduces their stability and increases the probability of releasing free Gd^{3+} into the body [11]. Macrocyclic chelates have a closed structure that significantly increases their chemical stability and minimizes the risk of free gadolinium release [9]. In patients with normal renal function, GdCA are rapidly eliminated (up to 98%). The biological half-life of these agents is approximately 1.5 to 2 hours, with approximately 90–95% of the administered agent being excreted unchanged in the urine within 24 hours of administration [12].

GdCA are a growing environmental concern, mainly due to their persistence and biological activity after elimination from the body. After application, GdCA are excreted unmetabolised, mainly in the urine, and subsequently enter wastewater. Traditional wastewater treatment plants are not able to completely remove these substances, which leads to their presence in aquatic ecosystems. The accumulation of gadolinium

in organisms can have negative impacts not only on the organisms themselves but also on higher trophic levels, including humans. The risk is also increased by the fact that the presence of gadolinium in the environment can be masked by its relatively low concentrations, which, however, can increase at higher levels of the trophic chain [6].

1.2 Gadolinium as an environmental contaminant

Anthropogenic gadolinium (Gd_{ant}) enters the environment from several main sources. The most important is hospital wastewater, which contains residues of GdCA used in MRI [13]. Other sources include the disposal of household electronic waste, such as old televisions or computers, and industrial processes such as petrol production, where it is used as a catalyst [7]. After release into the environment, Gd_{ant} accumulates in soil, wastewater, and surface water sources, resulting in its long-term presence in ecosystems [7, 14]. In the natural environment, the concentration of gadolinium increases compared to natural levels, leading to the so-called gadolinium anomaly [1, 14]. The gadolinium anomaly is direct evidence of the anthropogenic origin of the contamination, as the natural content of gadolinium in nature correlates with the content of other REE. Positive gadolinium anomalies have already been detected in many countries around the world, for example in Japan, Germany, France, the Netherlands, and the Czech Republic [14, 15].

The phenomenon is typical, especially for densely populated areas with intensive use of MRI, where there is a systematic increase in the concentration of Gd in water sources [1, 3]. In the wastewater treatment plant (WWTP) process, iron ions (Fe^{3+}) can replace gadolinium in the chelate, leading to its release as a free ion. This reaction is a result of the higher stability of Fe^{3+}-EDTA complexes compared to Gd^{3+}-EDTA [3, 12]. The fate of Gd_{ant} in the environment depends on its chemical properties and environmental conditions. It can be immobilized on soil particles or sediments, which limits its mobility, or it can remain mobilized as a soluble form that can be transported through waterways. Under specific conditions, it can also bioaccumulate in organisms and gradually increase in concentration in higher levels of the food chain [16, 17].

The presence of Gd_{ant} in natural ecosystems is of considerable concern. Due to its high chemical stability and non-degradability, gadolinium accumulates in aquatic ecosystems, where it can interact with microorganisms and other ecosystem components. Long-term contamination increases the risk of bioaccumulation in the food chain and may indirectly affect human health. Moreover, surface and groundwater contamination is difficult to remove even with advanced treatment methods, contributing to the long-term persistence of this problem [6, 18]. The extent of the environmental impacts highlights the need to improve wastewater treatment and to find alternative, more environmentally friendly ways to use gadolinium in healthcare and industry [6].

1.2.1 Impact on aquatic organisms

Especially its anthropogenic forms, gadolinium poses a potential threat to aquatic ecosystems due to its ability to affect essential biological functions of organisms. One of the main areas of its action is enzymatic activity and energy metabolism, while its presence in the organism often leads to disruption of growth processes [4, 19]. These effects are particularly evident in microorganisms and planktonic species that play

a vital role in the food chains of aquatic ecosystems [19]. Long-term exposure to higher concentrations of gadolinium in ecosystems has been associated with a reduction in biodiversity, which has negative impacts on ecological services such as natural water purification and the stability of fish populations [19, 20]. Disruption of these key processes can result in significant deterioration of the quality of the aquatic environment and the ability of the ecosystem to regenerate. Some species are more tolerant to gadolinium than others, leading to uneven impacts across ecosystems [4, 19, 20].

1.2.2 Food chain contamination

Gadolinium enters the food chain through bioaccumulation processes that start in lower organisms such as microalgae, plankton, and bacteria [21]. Organisms absorb gadolinium from the aquatic environment, where it is found as a result of anthropogenic activities [20]. Accumulation in these organisms forms the basis for the transfer of gadolinium to higher trophic levels. Microalgae and bacteria are the main food components for plankton, molluscs, fish and other aquatic animals [4, 19]. In this way, gadolinium is distributed in the ecosystem, posing a risk to higher trophic levels (see Figure 1) [4]. Although gadolinium concentrations are often low, long-term exposure raises concerns about its cumulative effects on human health [4, 11]. The amount of gadolinium in the human body is unknown, but it is estimated to be at low levels, higher in bones and lower in liver and kidney tissues. [4].

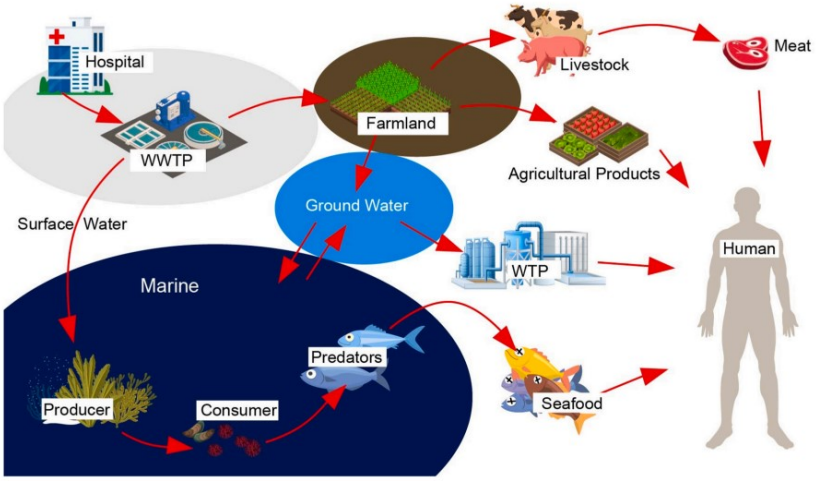

Figure 1 *Indirect entry of GdCA into the human body through the food chain [21]*

Contaminated surface or groundwater can be used to irrigate agricultural crops [22]. During irrigation, gadolinium can interact with soil particles or be directly accessible to plant root systems [16, 17]. Gadolinium tends to bind to mineral and organic substances, which affects its availability to plants [12]. Soluble forms of gadolinium, especially free Gd^{3+} ions or its complexes with organic matter, can be absorbed by plant roots. This process is partly influenced by plant characteristics such as its ability to accumulate heavy metals and lanthanides [23]. Although no confirmed cases of human poisoning through the food chain have been documented [24], concerns persist regarding the long-term effects of low-level REE exposure on human health. These elements accumulate in the blood, brain, and bones after entering the body [25]. To date, only limited information is available on the effects of dietary REE intake on human health [23].

2. Experimental part

2.1 Chemicals

The preparation of calibration standards for ICP-MS (inductively coupled plasma ionization mass spectrometry) and ICP-OES (inductively coupled plasma optical emission spectrometry) analyses of food samples and GdCA was performed using commercially available calibration standards. The multielement calibration standard M 008 with a concentration of 100 mg L^{-1} of elements "A" (Ce, La, Nd, Pr) and 20 mg L^{-1} of elements "B" (Dy, Er, Eu, Gd, Ho, Lu, Sm, Tb, Tm, Yb) and single-element calibration standards Gd and In with a concentration of 1 g L^{-1} were used. For ICP-MS analysis of food samples, multielement calibration solutions containing 10; 5; 1; 0.5; 0.1; 0.05; 0.01 μg L^{-1} of elements "A" and 2; 1; 0.2; 0.1; 0.02; 0.01; 0.002 μg L^{-1} of elements "B" were prepared. An internal indium standard with a concentration of 1 μg L^{-1} was in all working solutions (blanks, calibration solutions and samples). For the analysis of wine samples, a calibration series and a blank were prepared with the addition of 96% ethanol with a final content of 1.4% ethanol in the solution, which corresponds to the estimated ethanol content in ten-fold diluted wine samples for analysis (1.2–1.4% ethanol). Ultrapure water was used to prepare the blanks and standard solutions. For the ICP-OES analysis of the Gd content in waste GdCA, a concentration series of calibration solutions was chosen at 10; 5; 1; 0.1; 0.01 mg L^{-1} Gd. All calibration solutions were supplemented with ultrapure water.

2.2 Test organisms

The DIN medium for Water quality – Fresh water algal growth inhibition test with unicellular green algae (AGI test) was prepared according to EN ISO 8692:2012 [26]. The pH was determined by measurement and adjusted to 8.1 ± 0.2 using HCl (1 mmol L^{-1}) or NaOH (1 mmol L^{-1}) solutions as necessary.
The freshwater green microalgae *Chlorella vulgaris* (CV) and *Raphidocelis subcapitata* (RS) were selected as test organisms for the AGI test for Gd compounds (GdCA and $Gd(NO_3)_3$). Pre-cultivation of microalgae for the AGI test was performed according to the standard two to four days before the test. Cultures were grown in DIN growth medium at 20 °C on a magnetic stirrer. On the day of the test, the cultures were centrifuged, the eluates were removed, and the cell density was measured by the fluorescence method. The culture was then diluted with DIN medium to the desired density not exceeding 10^4 cells mL^{-1}.

2.3 Samples

To monitor anthropogenic Gd in foods, four sample types were analysed: flour (wheat, rye, spelt), rice, carrots, and wine (white, rosé, red). A total of 225 solid samples were collected, including 120 flours, 65 rice, and 40 carrots. Flour originated mostly from the Czech Republic (>70%), Slovakia, Italy, Israel, and Germany. Rice was mainly from Italy, Myanmar, and Pakistan, with some from Vietnam, Thailand, and 28% of unknown origin. Carrots were predominantly Czech (>60%), with some from Slovakia, Italy, Poland, and the Netherlands. Samples were sourced from supermarkets, with a few carrots from private gardens.Rice samples were ground to a powder in a mortar. Carrot samples were washed, cut with a ceramic knife and then dried in a laboratory oven at

105 °C for 48 hours. Dry matter was determined as the ratio of the weight of the dried sample to the weight of the fresh sample. Flour samples were not further treated before decomposition. The samples were stored in plastic bags in a dry and dark place at room temperature. All solid food samples were digested using a a microwave digestion system. In three replicates, each 0.1 g sample was dissolved in 3 mL of 65% sub-boiled HNO_3 and 0.5 mL of 30% H_2O_2, then diluted to 25 mL following digestion.
The beverage food samples included 200 wines: 131 white, 18 rosé and 51 red. The majority came from the Czech Republic (75%), mainly from Moravia (92%). The remaining samples were from Romania, France, Hungary, Slovakia, Austria and 38 without stated origin. The wines were obtained from wine festivals, wine cellars and included 31 varieties and several cuvées. The samples were stored at −18 °C and thawed and diluted tenfold with ultrapure water before ICP-MS analysis.

2.4 Instrumentation

An OptiMass 9500 inductively coupled plasma ionization mass spectrometer was used for ultra-trace analysis of food samples and reference material. The following REE were measured: Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu. Optimised ICP-MS analysis conditions: plasma power 1200 W; plasma, auxiliary, and nebuliser gas flow rates were (all in L min^{-1}) 12, 0.55 and 0.91; multiplier gain 2,700 V; acquisition time 5 s; and three sample replicates. Isotopes were selected to minimise isobaric interferences. The calibration demonstrated a high degree of linearity ($R^2 > 0.999$). The ICP-MS method was validated using certified reference materials (CRM): GBW 10052 Green Tea and GBW 07603 Bush Twigs and Leaves. The CRM were mineralised in triplicate and then prepared before ICP-MS analysis in the same way as the solid food samples. The results were in accordance with the manufacturer's specifications. The recovery ranged from 91–127%, while the long-term repeatability, expressed as relative standard deviation, ranged from 1.33–7.80%. These parameters were monitored using calibration standards (0.1/0.02 and 0.5/0.1 μg L^{-1} of elements "A"/"B"). Instrumental limits of detection (LOD) were between 0.1–0.7 ng L^{-1}.
Analysis of Gd content in GdCA waste container samples was performed using an Integra 6000 inductively coupled plasma optical emission spectrometer, due to the expected higher Gd concentrations in this type of sample. ICP-OES spectrometer setup parameters: plasma power 1,000 W; plasma, auxiliary, and nebuliser gas flow rates were (all in L min^{-1}) 10, 0.4 and 0.52; and three sample replicates. For Gd quantification, the most sensitive spectral lines at wavelengths of 342.247 nm (with background correction +0.0382 nm) and 336.223 nm (with background correction +0.0376 nm) were used, which showed minimal interference with other elements. The LOD of the method was determined to be 0.006 mg L^{-1}. The long-term repeatability and recovery of ICP-OES analyses were verified after each series of 15 samples by performing a recalibration that included a calibration standard with a concentration of 10 mg L^{-1} Gd and a blank. The long-term repeatability of the analysis reached values in the range of 1.86–2.78%, which indicates a high accuracy of the method in repeated measurements. The recovery of the results ranged between 89–103%, which indicates the reliability of the analysis and the ability of the method to accurately quantify the Gd concentrations in the analysed samples.

3. Results and discussion

The dissertation fulfils the set objectives, namely to comprehensively investigate the current knowledge on the presence of Gd of anthropogenic origin in food and its influence on the development of gadolinium anomaly and to conduct a separate experimental investigation. The experimental part focuses on monitoring the presence of Gd of anthropogenic origin in the food chain through elemental analysis of selected REE and subsequent evaluation of its content and gadolinium anomaly in samples of solid foods (flour, rice, carrots) and beverages (wine). The study also analyses the mass flow balance of Gd-based contrast agents at MRI facilities. Last but not least, the ecotoxicity of GdCA and $Gd(NO_3)_3$ on freshwater microalgae, which represent the input link of food chain contamination, is also investigated.

3.1 Literature review of the presence of gadolinium in the food chain

This chapter focuses on the collection and evaluation of available literature data on the presence of REE in foods, with a focus on gadolinium, one of the most studied elements of this group. The aim is to quantify the proportion of anthropogenic gadolinium in foods and, based on these data, to evaluate the existence of a gadolinium anomaly as a parameter indicating its presence.

The data collected include plant products, fungi, and seaweeds, which are significant accumulators of REE and other environmental contaminants. Gd (80–300 μg kg^{-1}) was detected in higher concentrations depending on the location and type of fungus [27], suggesting that fungi can serve as indicators of environmental REE pollution. Significantly different results were recorded in tomatoes, where REE concentrations differed according to the part of the plant. The highest values were in the roots, where Gd reached 150 μg kg^{-1} [28], indicating strong bioaccumulation from the soil. Conversely, fruits contain lower concentrations, probably due to the limited transport of these elements in the plant. A similar trend was observed in rice, where REE concentrations in roots significantly exceeded those in grain. For example, the concentration of Gd in roots reached 4,400 μg kg^{-1}, while in grain it was only 120 μg kg^{-1} [23]. Lower concentrations of REE were detected in orange peels originating from different regions, with Gd values ranging from 6–85 μg kg^{-1} [29]. Pumpkin seeds and their oil show minimal REE concentrations, for example, only 0.064 μg kg^{-1} of Gd was detected [30], which confirms the low affinity of oils for REE. Interesting results are also obtained from analyses of wine and grape must, where REE concentrations were relatively low, with Gd values in the range of 0.005–0.202 μg kg^{-1} for wine and 0.222–0.434 μg kg^{-1} for must [31]. This suggests that grapevine fruits have a limited ability to accumulate these elements from the soil. On the contrary, seaweeds, especially kelp and aonori, contain significant REE concentrations, which is a consequence of their ability to accumulate elements from seawater. 13.6 μg kg^{-1} of Gd was measured for kelp and 19.5 μg kg^{-1} for aonori [32].

The highest concentrations of REE were found in the roots of plants such as tomatoes and rice, confirming the importance of the root system in accumulating elements from the soil. Fruits and oils show lower concentrations, likely due to the limited mobility of these elements in plant tissues. Fungi and seaweeds have a high bioaccumulation capacity, making them suitable indicators of pollution and a potential risk to consumers.

These data help in understanding the distribution of REE in food chains and their anthropogenic origin.
The distribution of REE is commonly determined using the PAAS normalisation standard, which minimises distortion when calculating anomalies and allows for accurate data comparison. PAAS is used to normalise REE concentrations and calculate the gadolinium anomaly, which is the ratio between the Gd concentration normalised to PAAS and the geogenic Gd concentration (Gd_{geo}). The concentrations of Gd_{ant} in foods were calculated by subtracting Gd_{geo} from the total Gd concentration [33].
The gadolinium anomaly values and concentrations of anthropogenic gadolinium in food and crop samples, as calculated from literature data, indicate variations across food types, suggesting that the presence of Gd is influenced by biological and anthropogenic factors. Pumpkin seed oil showed a gadolinium anomaly of 1.2 and only 0.009 μg kg^{-1} for Gd_{ant}, suggesting a predominance of natural Gd sources with minimal anthropogenic contamination. Concentrations in tomatoes varied by plant part, with the highest Gd_{ant} concentration in roots (34.9 μg kg^{-1}). The only sample with a confirmed positive gadolinium anomaly was mushroom Jew's ear, with a value of 1.6, exceeding the threshold of 1.5. Calculations could not be made for rice samples due to undetected Tb concentrations. These findings are important for assessing the impact of anthropogenic activities on REE levels in food and can serve as a basis for further research. Advanced analytical methods, such as ICP-MS with preconcentration techniques, are crucial for detecting gadolinium at very low concentrations and monitoring its transport, accumulation, and potential ecosystem impacts.

3.2 Gadolinium in the food chain

The samples used in this study (wheat flour, spelt flour, rye flour, rice, carrots, and wine – white, rosé, red) were selected as representatives of common European foods. According to the Czech Statistical Office, wheat flour, rye flour, rice, carrots, and wine rank among the most frequently consumed foods in the Czech Republic, emphasising their dietary significance [34]. Contamination of these staple foods could seriously affect public health, as they are key ingredients used in baking, confectionery, and everyday cooking.
Measured Gd concentrations varied across the analysed samples: the highest median concentrations were found in wheat flour (13.2 μg kg^{-1}) and carrots (11.4 μg kg^{-1} dry weight), while the lowest concentrations were detected in wines (0.043–0.16 μg L^{-1}). Overall, the range of Gd concentrations spanned 2.57–154 μg kg^{-1} for flour, 2.08–35.3 μg kg^{-1} for rice, and 5.03–30.3 μg kg^{-1} for carrot dry weight, and 0.013–1.79 μg L^{-1} for wine. Compared to other studies reporting Gd content in rice from 0.003 to 18.620 μg kg^{-1} (for samples from Australia, India, Italy, Pakistan, Sri Lanka, Thailand, Vietnam, and the USA) [35] and 120 ± 80 μg kg^{-1} (for samples from Cuba) [23], the Gd concentrations in rice measured in this study are in a similar range, i.e. comparable. Portuguese studies report average Gd concentrations in wine samples in the range of 0.0046–0.202 μg L^{-1} [31] and 0.016–0.085 μg L^{-1} [36], where our measured values were at a comparable lower limit, but the upper limit was almost nine times higher for white wines than the maximum values found in Portuguese wines. Studies focusing on REE or specifically Gd concentrations in flour or carrot samples were not found, this is an uncharted topic.

There are currently no specific limits set for Gd or total REE content in food in European Union or US legislation [37, 38]. Regulations such as European Regulation (EU) 2023/915 focus on limits for some heavy metals (e.g. Pb, Cd, Hg) and other contaminants, but neither Gd nor REE are included [38]. The lack of specific limits highlights a gap in the legislative frameworks, especially because of the increasing occurrence of REE in the environment caused by anthropogenic activities.

3.2.1 Gadolinium anomaly and anthropogenic gadolinium in food and beverages

A positive gadolinium anomaly was confirmed in several samples across all food types, including wine. Figure 2 shows the gadolinium anomaly in each sample type in the form of boxplots. The highest gadolinium anomaly values were found in wheat flour (0.52–23), while wine exhibited the lowest range (0.36–1.7). Significant anthropogenic contamination was indicated by a sample of wheat flour from the Czech Republic with the highest value of 147 μg kg^{-1} Gd_{ant}. Similar positive values were also observed in spelt and rye flours, rice, and carrots, regardless of their geographical origin. In contrast, positive gadolinium anomaly values in wine were rare, suggesting distinct environmental and production influences.

Figure 2 *Gadolinium anomaly for analysed food and beverage samples*

The data suggest that cereals, particularly flour, show the highest concentrations of Gd_{ant}, warranting further food chain monitoring, especially in regions with intensive GdCA usage. The gadolinium anomaly phenomenon is crucial not only for understanding environmental burdens but also for ensuring food safety due to the toxicity of Gd^{3+} ions. Future research should focus on monitoring the movement of Gd in the food chain and implementing measures to reduce its release into the environment.

3.3 Mass flow balance of GdCA at MRI facilities

The mass flow balance of GdCA at MRI facilities is crucial for monitoring Gd releases into wastewater and healthcare waste, which have potential ecological impacts. This research involved collecting used containers and conducting surveys at selected MRI facilities, i.e. Pardubice Regional Hospital (NPK), Multiscan and Hradec Králové University Hospital (FN HK). The sample set contained 5 different GdCA, namely Gadovist 1 mmol mL^{-1} (Gadovist), Dotarem 0.5 mmol mL^{-1} (Dotarem), ProHance 279.3 mg mL^{-1} (ProHance), Clariscan 0.5 mmol mL^{-1} (Clariscan) as macrocyclic GdCA and Primovist 0.25 mmol mL^{-1} (Primovist) as linear GdCA. The laboratory carried out quantification of collected waste, evaluation of questionnaires, optimisation of the number of GdCA container rinses, and ICP-OES analysis of rinses of all types of waste containers.

3.3.1 ICP-OES analysis of GdCA waste container rinses

To determine the residual Gd content in used GdCA containers, rinsing procedures followed by ICP-OES analysis were conducted. Optimisation of the rinsing process established that five consecutive rinses per container were sufficient for removing residual Gd. Containers were rinsed with 10–15 mL of ultrapure water, depending on the container size, and the collected samples were diluted according to the container type.

Residual Gd concentrations ranged from 2.58–16.4 mg L^{-1} for vials and 0.16–12.7 mg L^{-1} for syringes, corresponding to 0.23–5.2% residual Gd in vials and 0.10–2.4% in syringes. The lowest residual content was found in the 10 mL Primovist syringe, while the highest was observed in the 17 mL ProHance syringe. However, it is not possible to definitively determine whether vials or syringes are more efficient in terms of maximising the use of GdCA content.

3.3.2 Questionnaire survey at MRI facilities

A questionnaire survey was conducted at three selected MRI facilities, focusing on the use, consumption, and disposal of GdCA waste. The survey results provide insights into the types of GdCA used at each facility, their consumption volumes, the average number of MRI scans performed monthly, and the proportion of those involving GdCA, as well as the waste produced and how it is managed. The types of GdCA used at the facilities matched those found in the waste collection. Most facilities purchase GdCA based on need, with NPK being the only one to maintain precise records of regular purchases of supplies.

Multiscan performs the highest number of MRI scans, but only 6% of these involve the use of GdCA, which results in a lower amount of waste. In contrast, FN HK carries out up to half of its MRI scans with the use of GdCA, leading to a higher volume of waste. The usual dose of GdCA is adjusted according to the patient's weight, making it difficult to determine its exact volume. The number of waste GdCA vials ranged from 100 to 165 units. Waste management procedures for GdCA are in line with the regulations for waste disposal in healthcare facilities. The survey revealed clear differences in the amount of GdCA waste, which can be attributed to specific organisational and operational practices. A key factor influencing waste production is the variation in record-keeping and handling of GdCA. NPK, with its detailed tracking

of regular stock purchases and handling, tends to generate higher amounts of waste, while Multiscan likely optimises stock levels and minimises excess, resulting in less waste.

The overall conclusions emphasise the importance of establishing standardised procedures for managing GdCA waste. Recommendations include optimising dosing based on patient weight, improving handling of packaging, and regularly analysing waste production. These measures could help reduce waste and its environmental impact without compromising the quality of diagnostic services.

3.3.3 Balance of waste GdCA at MRI facilities

The analysis of GdCA waste vials at selected MRI facilities provides valuable insights into the consumption and efficiency of these substances. Data from three facilities (NPK, Multiscan, and FN HK) reveals differences in preferred volumes and types of packaging, which influence the amount of residual GdCA in the waste. At NPK, Gadovist 7.5 mL and 10 mL syringes are the most commonly used, with both types being the most efficiently emptied. Other GdCA containers tend to have more residual GdCA. The ratio of empty to residual vials is 1:1. Multiscan also predominantly uses Gadovist 7.5 mL and 10 mL syringes, with a higher proportion of empty vials. However, less of the GdCA is fully utilised, possibly due to the transfer of GdCA to another sterile syringe to avoid contamination. Gadovist 15 mL and Dotarem 20 mL vials are only used sparingly, with the empty-to-residual vial ratio around 2:1. At FN HK, Gadovist syringes (7.5 mL and 10 mL) are the most frequently used, and syringes are more effectively emptied than vials, with a waste ratio of 3:2 (empty: residual).

These findings indicate that all three MRI facilities predominantly use smaller volumes of Gadovist in syringes, which seems to optimise the reduction of GdCA residues in waste. This analysis highlights the importance of selecting appropriate GdCA packaging types to optimise usage and minimise residual waste, thus contributing to more efficient and environmentally-friendly MRI operations.

Figure 3 presents a graph illustrating the balance of Gd quantities at individual MRI facilities, providing an overview of Gd usage and waste.

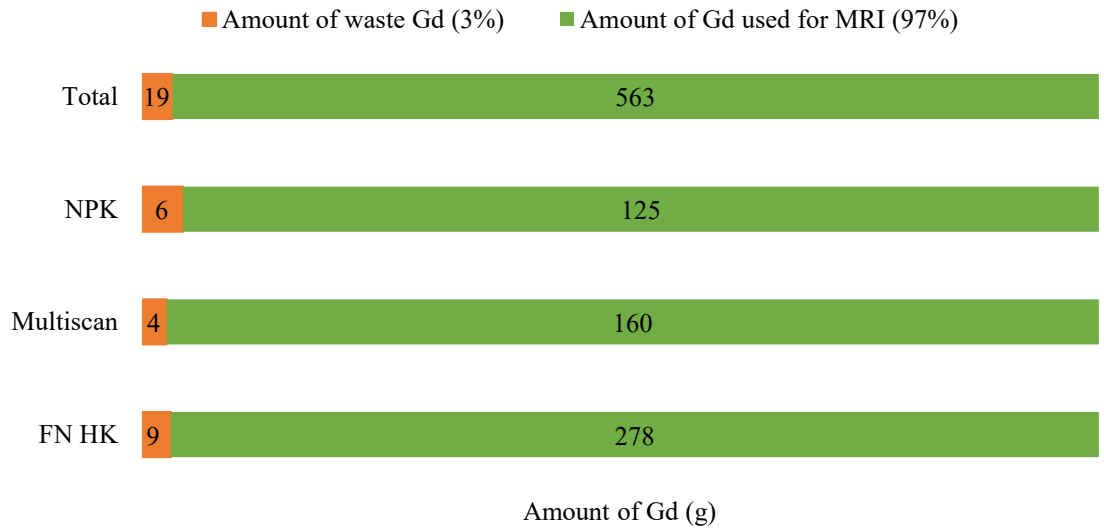

Figure 3 *Balance of Gd quantity at individual MRI facilities*

It displays the total amount of Gd derived from GdCA, broken down into the proportion of Gd effectively utilised for diagnostic purposes and the proportion of Gd waste remaining as residue in unused containers. Over the course of one month, a total of 563 g of Gd (97%) was used across the three selected MRI facilities, while 19 g of Gd (3%) was discarded as residue in waste containers. This demonstrates the high efficiency of Gd utilisation from GdCA containers for imaging purposes. NPK used 125 g of Gd, with 6 g (4.6%) as waste, the highest waste ratio across the facilities, possibly due to specific operational practices. Multiscan used 160 g of Gd, with 4 g (2.4%) as waste, showing the highest efficiency in Gd usage. FN HK used 278 g, with 9 g (3.1%) as waste, consistent with the overall trend of high GdCA utilisation. These results confirm that MRI facilities efficiently use contrast agents containing Gd.

The findings from the GdCA waste balance could significantly influence waste management procedures and regulation. Identifying packaging types that generate the least residual Gd enables healthcare facilities to implement more efficient GdCA handling strategies. Standardising the use of smaller syringes, such as Gadovist 7.5 mL and 10 mL, which produce the least residue, could reduce GdCA waste. Measures such as thorough rinsing of containers (recycling Gd) and more precise GdCA dosing could further minimise residual waste. These insights could also inform regulatory policies, such as mandatory reporting of GdCA waste volumes or standardised disposal procedures. This approach would not only reduce the environmental impact of Gd residues but also optimise the ecological sustainability of healthcare operations [39].

3.4 Study of the environmental impacts of gadolinium on aquatic organisms

With the increasing release of Gd into the environment, especially through wastewater, there is an increasing need to understand its impacts on aquatic organisms and ecosystems. In recent years, more studies have been published on the toxicity of Gd in freshwater environments, identifying risks to important freshwater organisms and proposing strategies to minimise these risks. Our work focuses on the effects of Gd on freshwater microalgae, which are crucial for primary production and ecosystem functioning. The standardized AGI test is a key tool for assessing the toxicity of Gd and its impact on the aquatic environment.

The AGI test (algal growth inhibition test), described in the EN ISO 8692:2012 standard, is a key tool for assessing the toxicity of chemicals in aquatic environments. This test evaluates the impact of chemicals on the growth of microalgae based on changes in biomass, which helps assess ecological risks. Four GdCA were selected for testing: Gadovist (gadobutrol), Dotarem (gadoterate), ProHance (gadoteridol), and MultiHance (gadobenate dimeglumine), all commonly used in clinical practice. For comparison, the inorganic Gd salt, $Gd(NO_3)_3$, was chosen as a model for inorganic forms of Gd. Testing was conducted across a concentration range from 0.01 to 100 mg L^{-1} of Gd, on two types of microalgae: *Chlorella vulgaris* (CV) and *Raphidocelis subcapitata* (RS), to assess the potential toxic effects of these substances on algae growth. Fluorescence measurements during incubation provided data on biomass density, from which EC50 concentrations were calculated, indicating the concentration of the substance causing 50% inhibition of growth.

The results of the test indicate a varying effect of individual GdCA on microalgae. Gadovist showed mild toxicity for RS at concentrations above 10 mg L^{-1}, whereas it had

a negligible effect on CV. Dotarem and ProHance exhibited almost no toxic effect on either microalgae, suggesting their low ecotoxicity. MultiHance showed mild toxicity for RS but had virtually no effect on CV. These findings suggest that the ecotoxicity of GdCA depends on the specific chemical structure and type of the test organism. This could lead to recommendations for favouring less toxic GdCA, such as Dotarem and ProHance, in clinical practice. The AGI test for $Gd(NO_3)_3$ revealed significant toxicity at concentrations above 10 mg L^{-1}, with growth inhibition nearing 100% at 100 mg L^{-1}, indicating a high environmental risk of inorganic Gd for freshwater ecosystems. EC50 values were determined to be 2.4 mg L^{-1} for RS and 3.1 mg L^{-1} for CV. The dose-response curve is shown in Figure 4.

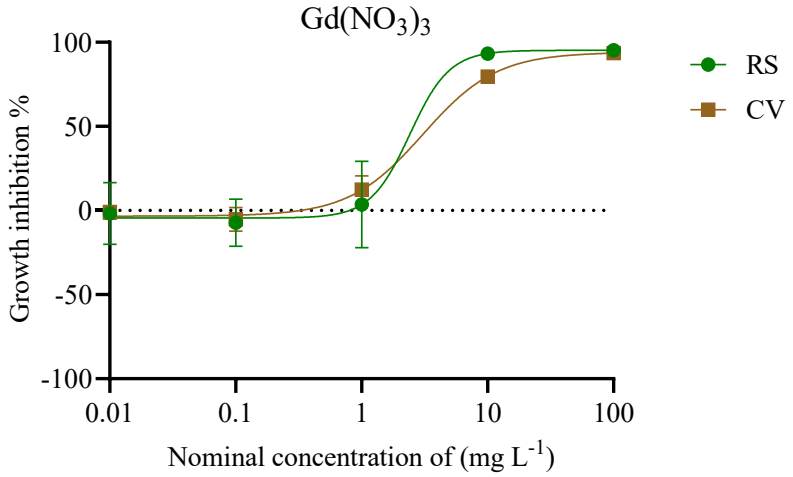

Figure 4 *Dose-response curve for $Gd(NO_3)_3$*

The tests demonstrated that GdCA are less toxic than $Gd(NO_3)_3$, which confirms the importance of chemical composition when evaluating environmental risk. This suggests that the stability of the chelates in GdCA plays a role in preventing the release of free Gd^{3+}, which is biologically active and toxic. Studies focusing on the ecotoxicity of various lanthanides have confirmed that Gd has high toxicity, particularly in its inorganic form, emphasising the risks associated with the release of these substances into aquatic ecosystems [40]. In contrast, chelates like Dotarem and ProHance exhibit lower toxicity, indicating that these substances should be favoured in clinical practice, especially in areas with acidic waters, where the toxicity of inorganic Gd may be elevated.

Conclusion

This work focused on a comprehensive analysis of the presence of gadolinium in the environment and its impact on ecosystems and human health. The main goal was to link the available scientific knowledge about the environmental and toxicological effects of gadolinium, with particular emphasis on its anthropogenic sources, and to offer strategies to mitigate its negative impacts. Part of the work was an analysis of the presence of gadolinium in food, while the proportion of its anthropogenic origin was quantified through the calculation of the gadolinium anomaly.

One of the main findings is the confirmation of the accumulation of gadolinium of anthropogenic origin in ecosystems and food chains. It comes mainly from hospital wastewater containing residues of gadolinium-based contrast agents, electronic waste and industrial processes. Analyses showed that gadolinium concentrations in foods vary according to their type and geographical origin, reflecting variations in exposure levels and contamination distribution.

The experimental part of the work also confirmed that effective management of GdCA usage and waste in MRI facilities can significantly contribute to minimising the environmental burden. Calculations of the mass flow balance of GdCA in the East Bohemian region showed that optimised procedures lead to lower amounts of residual waste and minimise the release of gadolinium into the environment. These findings emphasise the need for consistent control of the management of these substances at both local and global levels.

The results of the ecotoxicological study demonstrated that the composition of gadolinium compounds influences their ecotoxicity. While some GdCA show low or no toxicity, others may pose an ecological risk to aquatic organisms, underlining the importance of finding more environmentally friendly alternatives.

Despite its widespread use, especially in healthcare, it is clear that solutions need to be found to minimise the environmental impacts of gadolinium. Promising measures include the development of biodegradable contrast agents, the introduction of advanced wastewater treatment technologies, and preventing the excessive use of gadolinium in clinical practice. Overall, this work has provided new insights into the presence of gadolinium in food, its anthropogenic origin, and its impact on ecosystems. The results represent an important step towards a better understanding of the environmental impacts of gadolinium and provide a basis for future research aimed at developing more sustainable solutions and protecting the environment and human health.

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