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Ultra-Efficient Removal of Mercury from Contaminated Water using Selected Phenylimidazole Derivatives

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Abstract Heterocycles, including imidazole derivatives, are important chemicals due to their structural similarity to active natural molecules. This study explored the utilization of phenylimidazole derivatives for mercury removal from aqueous solutions. Three derivatives were tested, including one harboring a sulfhydryl functional group, thus offering distinct mercury-binding capabilities. Mercury binding experiments revealed modest mercury binding by two derivatives. However, extraordinary efficiency of mercury binding by the sulfhydryl-bearing compound was demonstrated through a reduction in mercury concentrations in pure water by 43-fold in just 15 min and decreasing below the analytical detection limit with prolonged exposure (30 min). Moreover, the ultra-efficiency of mercury binding was demonstrated by a capability to reduce mercury concentrations even in highly contaminated environmental water sample by a factor exceeding 1200-fold in 20 h, using minimal amounts of the compound. This phenylimidazole derivative exhibited comparable efficiency in binding mercury from authentic environmental water samples, hinting at practical applications for real-world remediation efforts. Kinetic investigations showed a pseudo-first order reaction with a rate constant of 0.26 min^{-1} and half-life of 2.69 min that governs mercury binding to this phenylimidazole derivative. These findings underscore the potential of the sulfhydryl-containing phenylimidazole derivative, as swift and effective binding material for aqueous mercury removal. Its rapid action, coupled with binding efficiency, suggests a promising material for addressing mercury contamination in highly contaminated environmental water samples.

Keywords Phenylimidazole derivatives · Mercury polluted water · Mercury binding · Reaction kinetics · Idrijca River

1 Introduction

Heterocycles are one of the most important compounds used in medicinal chemistry. Synthetic nitrogen-containing heterocycles can demonstrate diverse biological activities due to structural similarities with active natural molecules (Gaba & Mohan, 2016). One of the most important heterocycles is imidazole, as imidazole-containing drugs have considerably broad

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scope within clinical medicine (Alghamdi et al., 2021; Kale et al., 2016; Tolomeu & Fraga, 2023).

Imidazole (1,3-diazacyclopenta-2,4-diene; $C_3H_4N_2$) is a five-membered nitrogen-containing heterocyclic aromatic compound (Noriega-Iribe et al., 2020; Zheng et al., 2020). It is an aromatic planar-ring molecule that exists in two equivalent tautomeric forms (1*H*- and 3*H*-imidazole) (Kale et al., 2016; Verma et al., 2013). On the contrary, the third isomer (2*H*-imidazole) is non-aromatic due to the absence of a sextet of π-electrons (Hoz et al., 2005). Imidazole is an amphoteric compound and consequently susceptible to both electrophilic and nucleophilic reactions (Kale et al., 2016; Verma et al., 2013). It is stable regarding thermal decomposition, and acid/base and oxidation/reduction conditions (Kale et al., 2016).

The research of compounds containing imidazole moiety is developing due to their potential applications as, e.g., pharmaceuticals, agrochemicals, corrosion inhibitors, supramolecular ligands, and catalysts (Ben Hmamou et al., 2012; César et al., 2004; Chen, 2016; Gaba & Mohan, 2016; Ngochindo, 1992; Weber & Rios, 2019; Zhang et al., 2014). Imidazole moiety can be readily found in many biologically active molecules and biological building blocks such as histidine, histamine, purines, vitamin B_{12}, and biotin (Gaba & Mohan, 2016; Noriega-Iribe et al., 2020; Verma et al., 2013). Therefore, it is a motif of interest for use in pharmaceutical research and medicine (Alghamdi et al., 2021; Tolomeu & Fraga, 2023).

Heterocycles containing imidazole moiety can play an important role in various biochemical processes (Jin, 2006; Kale et al., 2016). The peculiar structural characteristic of imidazole scaffold (electron-rich ring) allows its derivatives to bind with various biological receptors and enzymes via diverse weak interactions (hydrogen and coordinate bonds, ion–dipole, cation–π and π–π interactions, and hydrophobic and van der Waals forces) (Zhang et al., 2014). The presence of this unique heterocyclic ring in various natural biologically active molecules indicates that its utilization in synthetic compounds might be important for different physiological processes and activities (Narasimhan et al., 2011; Zhang et al., 2014). The presence of the characteristic functional groups on the imidazole ring might be exploited for binding toxic metal ions (such as mercury) and their removal from the contaminated environments.

The presence of imidazole moiety in natural compounds have driven the development of synthetic imidazole-containing therapeutic drugs. Many compounds that contain imidazole scaffold are known pharmacological drugs and been used to treat various diseases (Zhang et al., 2014). According to recent literature, these compounds can possess analgesic, antibacterial, antibiotic, anticancer, anticoagulant, antidepressant, antidiabetic, antiemetic, antifungal, antihistaminic, antihypertensive, anti-inflammatory, antileishmanial, antimalarial, anti-neuropathic, anti-obesity, antioxidant, antiparasitic, antitubercular, antiulcer, antiviral (anti-HIV), and cytotoxic activity/property (Gaba & Mohan, 2016; Kale et al., 2016; Noriega-Iribe et al., 2020; Shalini et al., 2010; Verma et al., 2013; Zhang et al., 2014; Zheng et al., 2020). Also, they can act as an enzyme inhibitor, e.g., inhibitors of β-lactamase, heme oxygenase, p38 MAP kinase, B-Raf kinase, HIV-1 protease, and nitric oxide synthase (Baures, 1999; Lee et al., 1994; Salerno et al., 2012; Shalini et al., 2010; Takle et al., 2006). Aryl imidazoles can be used in photography as photosensitive compounds, but their greatest value is due to their analgesic, anthelmintic, antinociceptive, anti-inflammatory and anticancer properties (Abdel-Rahman et al., 2020; Dutta, 2010; Puratchikody & Doble, 2007; Uçucu et al., 2001). Several phenylimidazole derivatives have been recently characterized for their activities as antiproliferative and antioxidant compounds, and as inhibitors of acetylcholinesterase and xanthine oxidase (Noriega-Iribe et al., 2020).

Mercury (Hg) contamination remains a pressing environmental concern owing to its unique biogeochemical cycling within ecosystems. Mercury, which exists in different forms in the environment, cycles through the atmosphere, water bodies, and soil, and undergoes transformations that contribute to its persistence in the environment. Recent studies emphasize that natural processes like volcanic eruptions and human activities, particularly artisanal and small-scale gold mining, coal combustion, and industrial emissions, contribute significantly to the release of mercury (UN-Environment, 2019). Moreover, advancements in scientific understanding have revealed that even small sources of mercury, such as certain consumer products and waste disposal methods, play a role in its ongoing presence, perpetuating its presence in the environment and posing risks to both wildlife and humans (Beckers & Rinklebe, 2017).

Strategies focusing on comprehensive monitoring, emission reduction, and innovative remediation techniques are being explored to address this persistent global challenge. Mercury contamination in water is an important environmental problem and there has been a growing interest in novel technologies and studies related to the removal of mercury from contaminated water (Abdelaal et al., 2023; Kurniawan et al., 2023; Li et al., 2021; Velarde et al., 2023; Wang et al., 2020; Wang et al., 2023; Zhang et al., 2022). Various technological methods have been created for trapping this harmful metal, encompassing ion exchange, membrane filtration, extraction, and adsorption (Huang et al., 2024; Khan et al., 2020; Long et al., 2021; Štrok et al., 2014). Adsorption stands out as the optimal choice due to its cost-efficiency, effectiveness, and simple preparation with various binding materials being reported in the literature (De Gisi et al., 2016). The utilization of imidazole derivatives for binding mercury ions and their removal from solution has not been widely addressed. Recent literature only describes several techniques that involve utilization of imidazole motif for mercury removal. These include removal of mercury ions from aqueous solutions using imidazolium-based ionic liquid (Mancini et al., 2013) and thiosemicarbazone-modified zeolitic imidazolate framework (Jaafar et al., 2021). Some of the novel materials/chemicals include MoS_2-nanosheet-decorated polyvinylidene fluoride composite membrane (Zhao et al., 2021) and sulfhydryl-functionalized graphene oxide (Bao et al., 2021). Sulfur-containing molecules seem to have special characteristics required for efficient Hg capturing. Therefore, the objective of this work was to test mercury binding potential of selected aryl (phenyl) imidazole derivatives, including one having sulfhydryl functional group, and the possibility of using them as a simple binding material for aqueous mercury in polluted waters.

2 Materials and Methods

2.1 Phenylimidazole Derivatives

Three phenylimidazole derivatives have been selected for this work. Two of them were previously synthesized using a conventional and microwave-assisted methods from benzil, ammonium acetate, and appropriate aldehyde/ketone (Živković, 2011). These phenylimidazole derivatives were 4-(4,5-diphenyl-*1H*-imidazol-2-yl)-2-methoxyphenol **(1)** (98% purity; purchased from BenchChem, Pasadena, CA, USA) and 2,3-diphenyl-1,4-diazaspiro[4.5]deca-1,3-diene **(2)** (98% purity; purchased from Cymit Quimica, Barcelona, Spain). The third derivative, 4,5-diphenyl-1*H*-imidazole-2-thiol **(3)** (97% purity; purchased from Sigma Aldrich, St. Louis, MO, USA), was specifically selected as it contains a $–N=C(SH)-N<$ moiety. This moiety is interesting as it could create a disulfide group (–S–S–), thus acting as a reducing compound, and because it resembles a functional group of dithizone (1,5-diphenylthiocarbazone), a chemical compound that readily forms a stable complex with mercury (Paci et al., 2000). All chemicals were used as received. Chemical structures of investigated phenylimidazole derivatives are presented in Fig. 1.

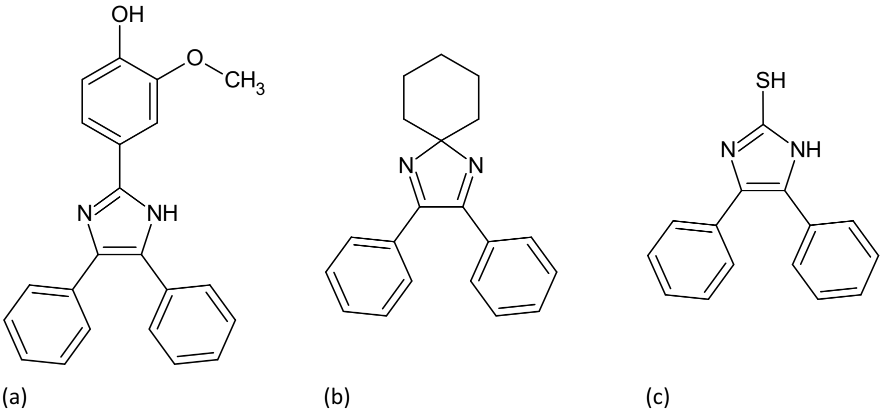

Fig. 1 Chemical structures of investigated phenylimidazole derivatives: (**a**) 4-(4,5-diphenyl-*1H*-imidazol-2-yl)-2-methoxyphenol (1); (**b**) 2,3-diphenyl-1,4-diazaspiro[4.5]deca-1,3-diene (2); (**c**) 4,5-diphenyl-*1H*-imidazole-2-thiol (3)

2.2 Mercury Binding Experiments

To test the mercury binding potential, these phenylimidazole derivatives were suspended in Milli-Q water (Type I water; Merck, Darmstadt, Germany) at the concentrations of 1 mg g^{-1} and 10 mg g^{-1}. These concentrations were chosen to be the same or lower as used in previous work on Hg binding to graphitic carbon nitride material (Chouhan et al., 2019, 2023). Mercury standard solution, obtained by the appropriate dilution of the certified reference material SRM NIST 3133 in Milli-Q water, was then added to a final mercury concentration of 851 ng kg^{-1}. The samples (pH 6.5) were then shaken for 15 min at 350 RPM, filtered through a 0.20-µm PTFE membrane filter, and prepared for the determination of total mercury (THg) in water according to the US EPA method 1631E (US EPA, 2002). In short, aliquots of filtered samples were diluted and treated with bromine monochloride (BrCl) to oxidize all organic compounds in the solution (i.e., phenylimidazoles that might have dissolved and complexed Hg in the aqueous phase) for 24 h. Prior to measurement, samples were treated with hydroxylammonium chloride ($NH_2OH \cdot HCl$) to remove the excess of BrCl. Mercury in the solution was reduced to elemental mercury (Hg^0) using a tin(II) chloride solution, purged out from the solution using nitrogen gas, collected on a gold trap (double amalgamation setup), thermally desorbed, and detected using atomic fluorescence spectrometer (Tekran 2600; Tekran, Toronto, Canada). The analytical system was calibrated using a SRM NIST 3133 solution, while the method's quality control was achieved using a certified reference material BCR 579 (total mercury in coastal seawater).

Based on the preliminary results, the experiments with compound **3** were further modified by extending the shaking period to 30 min and by testing the mercury binding from a seawater matrix (BCR 579) at acidic pH (1.5). Furthermore, mercury binding potential of the compound **3** using a real environmental water sample has been tested. River water originating from the Idrijca River (46.032299°N, 14.026606°E) was collected close to the former Idrija mercury mine in the western Slovenia. This river water is characterized with elevated mercury concentrations and high levels of suspended particulate matter (SPM) during high water flow (Kocman et al., 2011; Žagar et al., 2006). Sample was acidified with hydrochloric acid (Suprapur®) to pH of approximately 1.5 to release most of the particulate bound Hg into water. Therefore, compound **3** was tested for its possible application of quantitative removal of mercury from the real contaminated water sample.

2.3 Determination of Mercury Bound to Phenylimidazole Derivative

Although mercury was presumed to be strongly bound to phenylimidazole derivative, there was a possibility that it was actually reduced by the thiol groups to elemental Hg and subsequently evaporated and lost from the aqueous solution. This was a possibility as thiol groups can be relatively easily oxidized to form disulfide bond. Therefore, the content of mercury bound to the compound **3** had to be determined. Following the binding of mercury in Milli-Q water (851 ng kg^{-1}) to compound **3** (1 mg g^{-1}) after 30-min shaking, as described above, the suspension was filtered through a 0.20-µm PTFE membrane filter and dried at room temperature. Mercury content bound to compound **3** was determined using a mercury analyzer RA-915M with a PYRO-915+ thermal decomposition attachment (Lumex Scientific, St. Petersburg, Russia) that is based on Zeeman atomic absorption spectrometry. The instrument can be applied for determination of Hg content using thermal decomposition in various types of samples (Sholupov et al., 2004). Using a quartz boat, filters were inserted in the thermal decomposition unit that operates at 700 °C. Thermally reduced Hg was released and determined by Zeeman atomic absorption spectrometry. The results were corrected for the overall procedural blank.

3 Results and Discussion

3.1 Mercury Binding by Phenylimidazole Derivatives

Phenylimidazole derivatives **1** and **2** show modest mercury binding (Table 1) as the THg concentrations in water after 15-min shaking are only 2.4–3.9 times lower compared to the starting Hg solution. Higher concentrations of corresponding phenylimidazoles cause larger decrease in aqueous THg concentrations due to more active binding sites. However,

Table 1 Total mercury (THg) concentrations in filtered water samples after the treatment with phenylimidazole derivatives **1**, **2**, and **3**

Compound	c (compound) (mg g^{-1})	Matrix	THg (ng kg^{-1}) t = 0 min	THg (ng kg^{-1}) t = 15 min	THg (ng kg^{-1}) t = 30 min	THg (ng kg^{-1}) t = 20 h
1	10	Milli-Q	851 ± 30.5	219 ± 13.2	226 ± 5.26	n.m
1	1	Milli-Q	851 ± 30.5	355 ± 24.3	331 ± 3.70	n.m
2	10	Milli-Q	851 ± 30.5	223 ± 17.2	231 ± 19.9	n.m
2	1	Milli-Q	851 ± 30.5	276 ± 8.27	287 ± 5.94	n.m
3	10	Milli-Q	851 ± 30.5	< LOD *	< LOD *	n.m
3	1	Milli-Q	851 ± 30.5	19.9 ± 1.01	< LOD *	n.m
3	1	CRM BCR 579	1.80 ± 0.11 ***	n.m	< LOD **	n.m
3	0.33	Idrijca River	4067 ****	n.m	n.m	3.27 ± 0.24

n.m. – not measured.

LOD – limit of detection.

* LOD = 49.3 pg kg.$^{-1}$

** LOD = 77.5 pg kg.$^{-1}$

*** measured value; certified value is 1.90 ± 0.50 ng kg^{-1} (k = 2).

**** maximal measured concentration in the heterogeneous SPM-rich sample.

the overall Hg binding is probably governed by the chemical equilibrium and not by the availability of the binding sites as the tenfold increase in the concentration of these two phenylimidazoles causes only modest decrease in aqueous THg concentrations (Table 1). Furthermore, there are no significant differences in aqueous THg concentrations after 15- and 30-min shaking with either of these two phenylimidazoles. This indicates that Hg binding occurs relatively quickly, but the equilibrium between the solid and liquid phase dictates the amount of dissolved mercury in water.

Contrary to compounds **1** and **2**, 1 mg g^{-1} of compound **3** shows exceptional mercury binding as the THg concentrations in Milli-Q water after 15-min shaking were 43 times lower compared to the starting Hg solution. Higher concentrations of this compound cause a drop of aqueous THg concentration below the limit of detection (LOD). The corresponding LODs (Table 1) were calculated as three standard deviations of the procedural blanks. Longer exposure period (30 min) of this compound (both concentrations) causes a drop of aqueous THg concentration below LOD, which corresponds to more than 17,000 times lower THg value.

To further test the applicability of the compound **3** for mercury capturing in real environmental matrix, it was exposed to an acidified seawater sample (BCR 579). Within 30 min, it was able to reduce THg concentration below LOD, even at 1 mg g^{-1} level (Table 1). The exceptional mercury binding of compound **3** is presumed to be due to the formation of a stable Hg complex, similar to the one with dithizone, in which two ligands form an *N,S*-chelate ring with the metal atom (Paci et al., 2000). The possible application of the compound **3** for mercury remediation in contaminated waters also arises from the very short reaction time and even lower required mass, as compared to graphitic carbon nitride (60 min; 40 mg mL^{-1}), for example (Chouhan et al., 2023).

To test its applicability for mercury remediation, even lower amount of compound **3** (0.33 mg g^{-1} water) was suspended in the Idrijca River water for 20 h. This river sample contained relatively high amount of suspended particulate matter (42.2 ± 4.05 mg kg^{-1}) that is known to have elevated Hg content (Žagar et al., 2006). The total mercury concentration in the heterogeneous river water sample was up to 4067 ng kg^{-1} before exposure to the compound **3**. Shaking with compound **3** reduced THg concentration in this sample by a factor of up to 1200. Given that the amount of compound was at least three times less compared to other tests and that the exposure period was much longer for SPM-rich real environmental water sample, the overall result is satisfactory and presents a possible route for mercury binding from real water systems or a way to quantitatively pre-concentrate Hg for stable isotopic analysis.

3.2 Mercury Binding Kinetics

Time-dependent changes in aqueous THg concentrations in Milli-Q water were determined to assess the binding kinetics of mercury on the compound **3**. The compound **3** (1 mg g^{-1}) was dispersed in Milli-Q water containing 851 ng THg kg^{-1}. Every 5 min, two aliquots of water sample were withdrawn from the sample, filtered through a 0.20-μm PTFE membrane filter, and prepared for the determination of total mercury, as described above.

The time-dependent plot of THg changes is presented in Fig. 2. Linear correlation between ln(THg) and time can be observed, thus indicating that the mercury binding kinetics on compound **3** follows pseudo-first order with a reaction constant rate of 0.26 min^{-1}. The presence of the compound **3** does not seem to influence the reaction order, as it is probably completely undissolved at neutral pH (therefore having the value 1 for the chemical activity). Furthermore, the compound **3** is in great excess with respect to Hg (initial molar ratio of 934,000:1). The experimental THg values at time index of 25 min and 30 min were omitted from the consideration of the reaction order, as they deviate from linearity. This might be because these values were very low with large standard deviation (time index 25 min) or below the LOD (time index 30 min; Table 1). Alternatively, the reaction itself might follow some other reaction order at these very low THg concentrations.

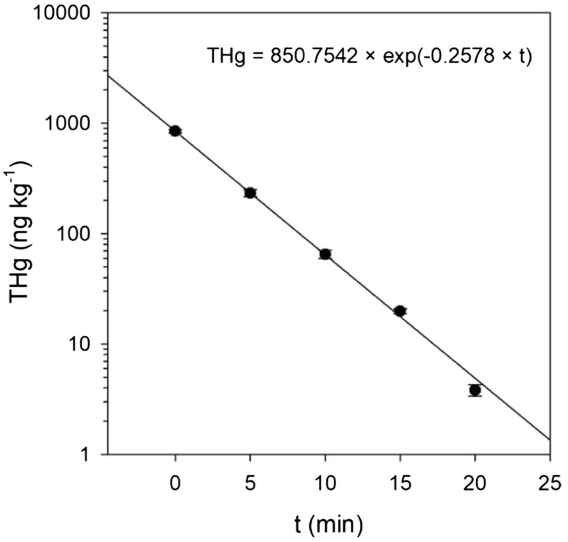

Fig. 2 Time-dependent changes in aqueous THg concentrations exposed to 1 mg g^{-1} of compound 3. Error bars represent one standard deviation of the replicate Hg determinations. The decrease in THg concentration follows the pseudo-first order kinetics with $k = 0.26$ $min.^{-1}$

Using the reaction constant rate of 0.26 min^{-1} for the pseudo-first order chemical reaction, reaction half-life can be calculated using the expression $t_{½} = \ln(2) / k$. Calculated half-life ($t_{½} = 2.69$ min) is fast enough to allow quantitative removal of mercury from aqueous solutions in reasonably short time period for remediation and mitigation purposes.

3.3 Content of Mercury Bound to Phenylimidazole Derivative

To test whether mercury was indeed bound to phenylimidazole derivative **3**, the amount of mercury bound to this compound was directly determined using Zeeman atomic absorption spectrometry. The average recovery of the amount of mercury bound to compound **3** (relative to the mass of Hg that was present in Milli-Q water at the beginning of the experiment) was 98.2 ± 3.35% ($n = 3$), after correction for the procedural blank. Therefore, Hg is indeed quantitatively bound to this compound, thus confirming its utilization for mercury binding applications from water samples. Furthermore, relatively low Hg blank in pure compound **3** (2.34 ng g^{-1}) could allow its utilization in Hg pre-concentration methods for specific analytical purposes (e.g., Hg stable isotope analysis in Hg-contaminated water samples).

3.4 Potential Applications and Limitations

The presence of a sulfhydryl functional group seems to contribute to the extraordinary mercury binding capabilities. A recent paper reviewed sulfhydryl-bearing compounds used for Hg removal from water and wastewater (Pohl, 2020). Mercury binding capabilities of these compounds were based on the theory of hard and soft acids and bases (HSAB), which assumes that soft bases (e.g., sulfhydryl groups) favorably bind to metals that are soft Lewis' acids (e.g., Hg^{2+}) (Pearson, 1963). On the other hand, the presence of hydrophobic phenyl groups in compound **3** lowers its solubility in water. These two effects contribute to effective binding and removing Hg from the water media.

The results presented in this study indicate potential applications of the selected phenylimidazole derivatives in environmental remediation efforts, especially the application of the compound **3**. This phenylimidazole derivative has chemical properties that make it an efficient sorbent for Hg in water, thereby removing it from the environment. Its chelating properties could be exploited in the remediation of metal-contaminated sites by sequestering the metal and reducing its mobility and toxicity to water organisms, and consequently to human consumers. This compound could be also incorporated into environmental functional materials for pollutant removal or pre-concentration and separation processes, such as mesoporous silica materials (Zhang et al., 2012).

However, there are also certain limitations for the use of these compounds, including the compound **3**. The presence of strong oxidizing chemicals in treated water might oxidize the sulfhydryl group to sulfate which would result in consequent slow release of bound mercury back into solution (Blue et al., 2008). The presence of SPM in real environmental water samples might also bias results as strongly bound Hg might not be completely released from the particles upon acidification. Furthermore, determination of low levels of mercury in natural waters always represents a methodological and analytical challenges. Therefore, it might be difficult to calculate how many folds was Hg concentration in waters actually lowered, as levels can get below the analytical limits of detection (Table 1). Furthermore, measurement uncertainty of the analytical method has a strong influence on presented data, especially near the limit of detection, as seen by the necessary removal of two data points during the determination of mercury binding kinetics (Fig. 2).

4 Conclusions

Mercury pollution in water bodies necessitates urgent and effective remediation strategies. Phenylimidazole derivatives emerge as a promising material for mercury binding. Within this study, modest to high efficiency of these derivatives in removing mercury ions from aqueous solutions was demonstrated, highlighting their potential in addressing this environmental challenge.

Based on the results presented in this paper, the research demonstrates the varying efficiency of phenylimidazole derivatives in mercury binding. While compounds **1** and **2** exhibit modest binding capacities, compound **3** is highly effective in reducing mercury concentrations in both synthetic and real environmental matrices. Compound **3** shows exceptional mercury binding ability, considerably reducing THg concentrations in Milli-Q water and acidified seawater below LODs within short exposure periods. Its efficiency was demonstrated even on SPM-rich Idrijca River water, reducing THg levels by a factor of up to 1200.

The kinetics of mercury binding to the compound **3** follow a pseudo-first order reaction with a rate constant of 0.26 min^{-1}. This suggests its potential for rapid mercury remediation. The compound **3** might be a promising candidate for mercury remediation due to its rapid action, high efficiency, and suitability for diverse environmental matrices. Its potential for pre-concentration of mercury in complex water systems might also presents a valuable tool for stable isotopic analysis.

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Author contribution **Igor Živković:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing—original draft preparation, Writing—Review & Editing, Visualization. The author has read and agreed to the published version of the manuscript.

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Data availability The author declares that the data supporting the findings of this study are available within the paper. Should any raw data files be needed in another format they are available from the corresponding author upon reasonable request.

Declarations

Competing Interests The author declares no conflict of interest.

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