Mercury Contamination in Food: Sources, Hazards, Prevention and Detection Techniques for Mercury Speciation Analysis: A Review

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Summary: Mercury is a highly toxic element that threatens both human health and the environment, with its toxicity varying by species, which possesses bioaccumulation and persistence. Common exposure sources include farm produce, aquatic products, and other foods. Therefore, understanding the mercury contamination in food for ensuring food safety, preventing mercury poisoning, and safeguarding human health and the ecological environment. This review summarizes mercury's presence in food, its sources, health hazards, current pollution status, limit standards and preventive and control measures. It also discusses common detection techniques for mercury speciation in food and anticipates the future developments in mercury speciation analysis, aiming to provide technical guidance for preventing and controlling mercury pollution and for screening and detecting mercury content in food.

Keywords: Mercury exposure, Food, Toxicity hazard, Speciation analysis, Detection techniques.

Introduction

Despite decades of effort, fundamental environmental issues remain unresolved, with pollution control still a critical challenge. This ongoing situation poses significant threats to food safety and public health, contributing substantially to the prevalence of human diseases. [1]. Heavy metals, particularly mercury, are significant environmental pollutants. Mercury is highly toxic even at low doses, and can contaminate water and soil, leading to ecological damage. It is absorbed by plants and animals, posing a biological threat, and can enter the human body through the food chain, resulting in mercury poisoning, which poses a serious health risk [2-3].

Recent exposure of mercury to water and soil has resulted in significant food safety concerns, including food mercury contamination and poisoning [4]. Mercury's propensity to accumulate in food, particularly aquatic products, alongside its potent chemical toxicity and biomagnification, poses ~~incalculable~~ irreparable risks to human health upon ingestion [5]. Numerous analytical detection methods have been developed to characterize mercury in food. However, the toxic effects of mercury depend not only the exposure dose but also critically on mercury species [6-7]. Currently, the mechanisms underlying the toxic effects of different mercury species in food require systematic elucidation. Understanding mercury contamination across different media and its pathogenic mechanism in vivo is crucial for advancing practical research on mercury poisoning in food, ultimately enhancing residents' quality of life. This review outlines the presence of mercury in food, its sources, health hazards, current pollution status, limit standards and preventive measures, aiming to inform strategies for mercury poisoning prevention populations. Additionally, it reviews advancements in mercury species analysis and detection technologies in food, offering insights into prevention, control and monitoring efforts.

Heavy metal mercury contamination in food

Heavy metal mercury

Heavy metals are defined as metals or metalloids with a density exceeding 5 g/cm^3, such as mercury (Hg), copper (Cu), cadmium (Cd), chromium (Cr), lead (Pb) and arsenic (As), among others [8-10]. Notably, Hg, Cd, Pb, Cr and As are recognized as the most toxic, collectively referred to as the “five poisons of heavy metals”, due to their harmful effects even at low concentrations. Mercury, unique for being the only metal that is liquid at room temperature and pressure, is prevalent in the environment and is highly toxic, thus it is prioritized as the most hazardous among these five [11]. The forms of mercury (Fig. 1) include elemental, inorganic, and organic compounds. Total mercury is sum of the inorganic and organic mercury contents.

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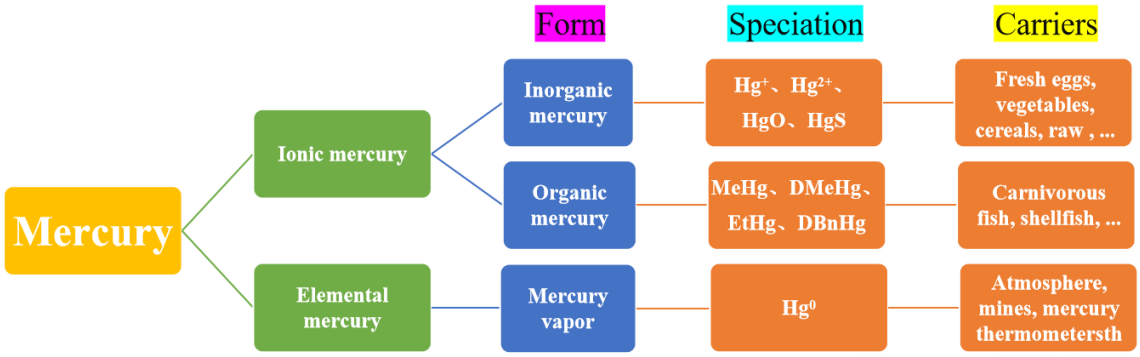

Fig. 1: The main speciation of mercury.

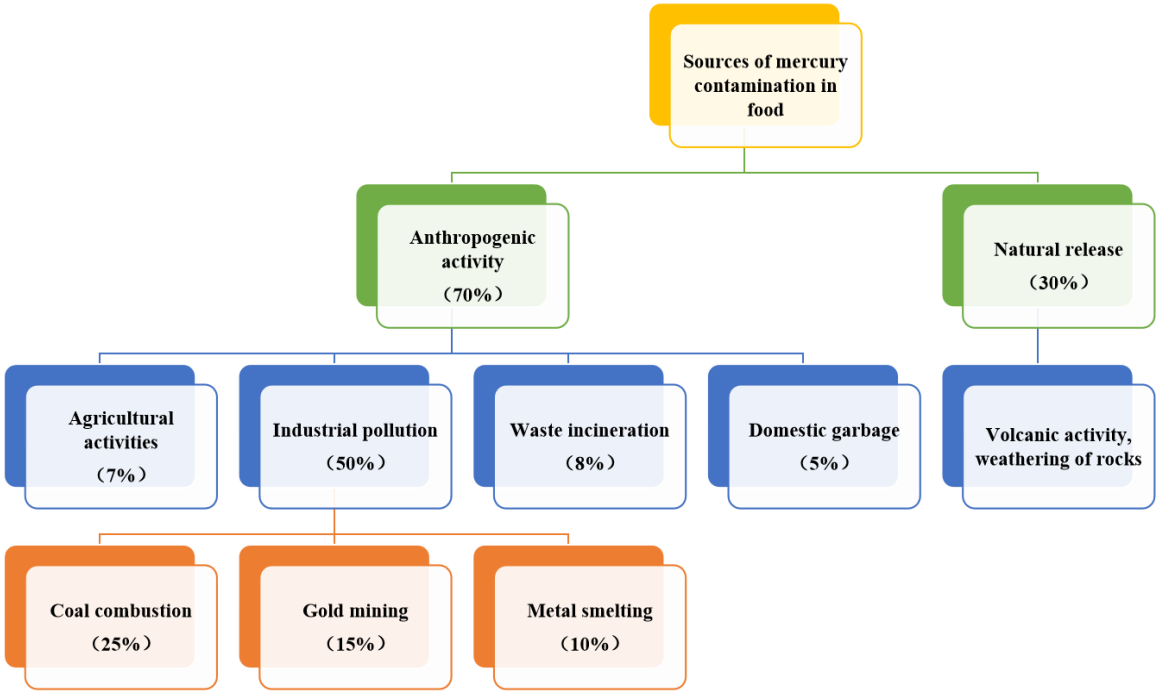

Fig. 2: Sources of heavy metal mercury in food.

Sources of heavy metal mercury production

Heavy metal contaminants in food primarily originate from human activities, with a minor contribution from natural rock erosion and weathering [12]. Mercury, in particular, is largely sourced from the pesticide, battery and paper industries. According to the Global Mercury Assessment 2018 by the United Nations Environment Programme (UNEP), global mercury emissions reached approximately 2,220 tons in 2015, predominantly from Asia (49%), South America (18%), and sub-Saharan Africa (16%). As shown in Fig. 2, mercury contaminates food through various pathways, primarily via human activities such as coal combustion, waste incineration, chlor-alkali production, metal mining, industrial manufacturing, and the release of mercury from domestic waste. A smaller fraction originates from natural sources [13]. Notably, artisanal and small-scale gold mining contributes nearly 40% of total global mercury emissions. However, reducing or eliminating the use of mercury-added products, such as thermometers and batteries, can significantly decrease mercury emissions [14].

Health hazards of the heavy metal mercury

Mercury in the natural environment undergoes rapid and subtle morphological and valence changes. Among heavy metal mercury pollutants, gaseous Hg^0 can be transported over long distances due to its extended atmospheric half-life, eventually oxidizing to Hg^{2+} and depositing in distant water bodies or soils. In aquatic systems, Hg^{2+} is biomethylated to organic mercury (MeHg), leading to bioaccumulation and biomagnification within food chains [15]. In water bodies containing organic matter, bacteria convert Hg^0 into MeHg, or first oxidize Hg^0 to water-soluble Hg^{2+}, which is then absorbed and methylated into lipid-soluble mercury species like MeHg and EtHg (Fig. 3). Organisms at the top of the food chain exhibit higher mercury levels than those at the bottom, such as large carnivorous fish compared to small herbivorous fish and plankton in the marine ecosystems, or songbirds versus spiders, omnivorous insects, herbivorous insects and plants in the forest ecosystems [16-17].

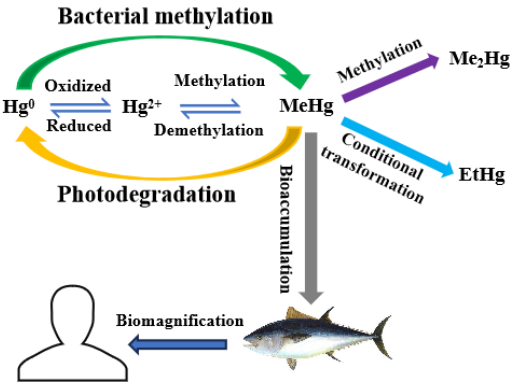

Fig. 3: Mercury cycle and methylation process.

The extent of harm from mercury exposure depends largely on the mercury species ingested **(**Fig. 3). Monomeric mercury is volatile; inhalation of high concentrations of mercury vapor (Hg^0) can lead to poisoning, characterized by chest tightness, shortness of breath, chills and fever, allergic dermatitis, and interstitial pneumonitis. This typically results from short-term exposure to high levels of mercury vapor or dust, either through occupational hazards or accidental release. Conversely, ingestion of Hg^{2+} primarily occurs via the digestive tract, where high concentration can cause acute poisoning, causing gastrointestinal mucosal damage and acute renal failure [18]. Hg^{2+} readily binds to sulfur in the proteins of central nervous system enzymes, forming mercury-thionein, which inactivates enzyme and triggers mental disorders, emotional uncertainty, tremors, and neurological damage [19].

Alkyl mercury compounds, such as fat-soluble MeHg, exhibit significantly higher toxicity compared to water-soluble Hg^{2+}. MeHg demonstrates a strong affinity for biological macromolecules, particularly proteins, and easily permeates cell membranes, crossing the blood-brain barrier to accumulate in tissues, organs, including the brain. This accumulation leads to central nervous system disorders due to the slow metabolism of MeHg in the body. Up to 98% of organic mercury pollutants in the human gastrointestinal tract can be absorbed, facilitating their passage through the placental barrier into the fetal brain, where they accumulate and disrupt normal growth and development [20]. Inorganic mercury poisoning mainly manifests as neurotoxicity and nephrotoxicity, while organic mercury poisoning results in severe neurological damage, tremors and potentially fatal outcomes. Moreover, pregnant women can transmit mercury to the fetus, causing growth retardation, paralysis and mental retardation and adverse effects [21-22].

Heavy metal mercury contamination incidents

Different mercury species exhibit distinct levels of toxicity, with organic mercury presenting the highest toxicity. The historical utilization of mercury by human spans millennia. Notable instances of inorganic mercury poisoning (Table-1) include the administration of “dental powder containing calomel” for toothache treatment in the U.S. in 1921, resulting in “pink disease” among infants characterized by symptoms such as pain in extremities, skin redness and swelling. In Europe during the late 18th century, the addition of mercury nitrate to felt hats for economic gain led to a significant number of workers suffering from “mad hatter's disease”, displaying symptoms like mental derangement, aggression, and physical tremors. Tracing back to China's Han Dynasty, the practice of gilt mirror production involved amalgamating gold with mercury to create “gold-mercury lacquer” which, when applied to copper surfaces, led to fatal mercury poisoning among artisans through inhalation of mercury vapor [23].

Table-1: Historically notable accidents of inorganic mercury poisonings.

Time	Country (or Region)	Poisoning incident
1921	USA	The use of "teething powder containing calomel" to treat toothache resulted in "pink sickness" in infants, i.e., pain in the hands and feet, redness and swelling of the skin, etc.
At the end of the 18th century	Europe	The addition of mercury nitrate to felt hats has resulted in a large number of workers suffering from "mad hatter's disease", with symptoms of insanity, aggressive behavior and body tremors.
Han dynasty	China	The use of the gilt mirror process to mix gold and mercury to make "gold-mercury lacquer" and apply it to the surface of copper objects, resulting in poisoning of the craftsmen by inhaling the mercury vapors.

Table-2: Historically notable accidents of organic mercury poisonings.

Time	Country	Poisoning incident	Cause of poisoning
1953	Japan	the Minamata disease	Long-term consumption of fish and other aquatic products contaminated with methylmercury
1960	China	Songhua River mercury pollution	Long-term consumption of river fish contaminated with benzene (benzene, nitrobenzene, etc.)
1971	Iraq	mercury poisoning from oat grains	Consumption of bread made from tainted wheat treated with MeHg and EtHg fungicides
1997	USA	Karen Wetterhahnmercury poisoning	Two drops of dimethylmercury solution dripped from a pipette onto a glove while performing a dimethylmercury experiment

Inorganic mercury poisoning has historically received limited attention, with concerns heightened only following prominent incidents of organic mercury poisoning (Table-2) that captured global attention in the 20th century. Notable examples include the "Minamata disease incident", the "wheat grain mercury poisoning incident", and the "mercury poisoning incident of American chemist Karen Wetterhahn" [24]. These incidents exemplify the severe consequences of organic mercury exposure. The Minamata disease incident in Japan in 1953 and the wheat mercury poisoning incident in Iraq in 1971 are illustrative instances of food-borne organic mercury poisoning. The former resulted from prolonged consumption of MeHg-contaminated aquatic products, while the latter stemmed from the ingestion of MeHg-contaminated wheat products. The "wheat mercury poisoning" incident resulted in tens of thousands of casualties, while the "Minamata disease" incident affected approximately 3,000 individuals. These cases underscore the substantial toxicity associated with organic mercury exposure.

In northeastern China, the Songhua River experienced a significant mercury pollution event when the Jilin Chemical Industry Company discharged over 150 tons of mercury-laden wastewater into the river (Table 2). This resulted in severe contamination of fish and other aquatic life, with mercury levels in fish far exceeding national safety limits. Prolonged consumption by local fishermen led to mercury content in their bodies tens to hundreds of times above normal, causing symptoms of mercury poisoning such as hand tremors, joint stiffness, muscle weakness, and narrowed visual field [25]. The complex and widespread nature of mercury contamination sources means that food mercury levels occasionally exceed safety standard in markets nationwide. To prevent excessive mercury intake, it is critical to enhance source management, raw material monitoring, process control, finished product sampling, and product substitution. These measures are essential to preemptively address the issue and safeguard public health.

Heavy metal mercury pollution prevention and control measures and safety standards

The severe impact of heavy metal mercury pollution on human health have prompted global initiatives to curb mercury emissions and exposure. The Minamata Convention on Mercury, effective since August 2017, mandates the cessation of production, import and export of mercury-containing batteries and other products from 2020, alongside phasing out of mercury thermometers, sphygmomanometers, and dental amalgams in favor of mercury-free alternatives [26]. In July 2024, the European Union (EU) 2024/1849 prohibited the production, import and export of dental amalgams and certain mercury lamps, with a full ban illegal mercury use and residual mercury in cosmetic starting on July 1, 2026 [27]. Furthermore, in October 2024, China's Ministry of Ecology and Environment announced restrictions on eight types of mercury-added products, including fluorescent lamps and dental amalgam, effective after December 31, 2025 [28].

Table-3: Domestic and international limitation standard of mercury in food （mg/kg）[29-38].

Food category	China	CAC	WHO	EU	FDA	USEPA	Japan	Korea
Canned food supplements for infants and young children	0.02	-	-	-	-	-	-	-
Carnivorous fish and its products	1.0-1.7 (MeHg)	0.8-1.7 (MeHg)	-	1.0	1.0	-	-	1.0 (MeHg)
Aquatic animal and its products	0.5 (MeHg)	-	-	0.3-0.5	-	-	0.3 (MeHg)	0.5
Edible mushroom and its products	0.1 (MeHg)	-	-	-	-	-	-	-
Milk and its products	0.1	-	-	-	-	-	-	-
Cereal and its products	0.02	-	-	-	1.0	-	-	-
Meat & fresh eggs	0.05	-	-	-	-	-	-	-
Food additives	-	-	-	0.1	-	-	-	-
Fresh vegetables	0.01	-	-	-	-	-	-	-
Drinking water	0.001 mg/L	0.001	0.006 mg/L	-	-	0.002 mg/L	-	-
Table salt	0.1	0.1	-	0.1	-	-	-	-

Note: "-" indicates that there is no such criterion.

To prevent mercury poisoning and protect human health, it's crucial to minimize the use of mercury-containing products and establish strict standards for mercury levels in food and permissible intake doses. Table-3 shows the mercury limits in food as set by the Codex Alimentarius Commission (CAC), the World Health Organization (WHO), the Official Commission of the European Union (EU), the U.S. Food and Drug Administration (FDA), the U.S. Environmental Protection Agency (USEPA), and other entities such as Japan and Korea [29-37]. Most regulations focus on drinking water and aquatic products, likely because these are primary source of human mercury exposure. In fish, which occupy a high position in the food chain, mercury concentrations can be up to a million times higher than in their environment, with 75% to 95% of the mercury present as MeHg [35]. In 2022, China updated its National Standard for Food Safety Limits of Contaminants in Foods (GB 2762-2022), specifying limits for total mercury and MeHg [38]. China mandates that the total mercury content in drinking natural mineral water must not exceed 0.001 mg/L. For fresh vegetables, milk and its dairy products, the total mercury content must not exceed 0.01 mg/kg; the total mercury content in grains and their products, canned auxiliary foods for infants and young children must not exceed 0.02 mg/kg. Meat and fresh eggs have a limit of 0.05 mg/kg, while edible salt is similarly regulated. Edible mushrooms and aquatic products are restricted to 0.1 mg/kg, carnivorous fish like tuna to 0.5 mg/kg, and other fish species to 1.0-1.7 mg/kg. China's mercury limits in food are comprehensive, aligning closely with international standards, and in some cases, they are more stringent.

The CAC established the tolerable weekly intake (TWI) for inorganic mercury and MeHg at 4.0 and 0.0016 mg/kg body weight, respectively [29]. On December 20, 2012, the European Food Safety Authority (EFSA) revised the TWI for MeHg to 1.3 mg/kg body weight, below the CAC's 1.6 mg/kg, while maintaining 4.0 mg/kg body weight for inorganic mercury [39]. The USEPA set the reference daily intake of CH_3Hg for adults is 0.0001 mg/kg body weight [34]. In October 2011, the FDA and the EPA classified fish into 3 categories based on mercury content, advising that fish with the lowest mercury levels are the best choice for priority populations such as pregnant women [32-34].

Species analysis of heavy metal mercury in foods

To effectively regulate mercury contamination in food and prevent mercury poisoning, merely establishing mercury limits and safe intake dose of mercury intake is insufficient. Essential technical support for the species analysis and detection of mercury in food is crucial to ensure food safety, protect people's lives and health, contribute to economic development and maintain social stability. In September 2021, China updated and released "GB5009.17-2021 National Standard for Food Safety Determination of Total Mercury and Organic Mercury in Foods". These standard details the pre-treatment techniques and methods for determining total mercury and methylmercury in

food, including atomic fluorescence spectrometry, direct mercury measurement, inductively coupled plasma-mass spectrometry, cold atomic absorption spectrometry for total mercury and liquid chromatography-atomic fluorescence spectrometry, as well as cold atomic absorption spectrometry for methylmercury.

Due to the complex composition of food and the instability of the mercury species, as well as the unmeasured original state, sample pretreatment operations — such as sample digestion, extraction, and enrichment and concentration — are typically conducted based on the specific conditions of the food samples before to mercury analysis [40-43]. These steps aim to isolate the target components, remove interferences, or concentrate the samples to ensure accurate analytical results and accurately reflect the food's intrinsic properties.

Researchers have consistently focused on mercury species analysis. Beyond above standard methods, there remains a critical need for innovative approaches that are simpler, faster, more sensitive, accurate, cost-effective and efficient to meet the growing demand for rapid food safety testing. Leveraging interdisciplinary collaboration and advancements in scientific instrumentation can significantly enhance the development of mercury species. Presently, sensing methods utilizing various functionalized nanomaterials are under extensive investigation (Table-4) [44-45].

Chromatography/spectrometry - mass spectrometry

Mercury detection in food such as atomic fluorescence spectrometry (AFS), cold vapor atomic absorption spectroscopy (CV-AAS), atomic emission spectrometry (AES), atomic absorption spectroscopy (AAS), inductively coupled plasma-mass spectrometry (ICP-MS inductively coupled plasma-optical emission spectrometry (ICP-OES) and inductively coupled plasma-atomic emission spectrometry (ICP-AES). [46-51]. However, single instrument approaches often suffer from limited sensitivity, poor selectivity, complex operation, and an inability to perform species analysis. To address these limitations and enhance the accuracy and sensitivity of mercury determination, researchers continue to develop improved methodologies.

Instrumental coupling technology, which integrates multiple analytical techniques such as separating-separating, separating-identifying, identifying-identifying, has revolutionized analytical methods. By combining these techniques, it overcomes the limitations of single instruments, offering rapid and precise analysis, with high automation and detection efficiency. This approach has gained widespread application. For mercury species analysis, commonly employed methods include liquid chromatography-atomic fluorescence spectrometry (LC-AFS), liquid chromatography-inductively coupled plasma mass spectrometry (LC-ICP-MS), gas chromatography-atomic fluorescence spectrometry (GC-AFS), gas chromatography-inductively coupled plasma-mass spectrometry (GC-ICP-MS), ion chromatography-inductively coupled plasma-mass spectrometry (IC-ICP-MS) and capillary electrophoresis-inductively coupled plasma-mass spectrometry (CE-ICP-MS) [52-57].

Coupled instrumental analytical techniques not only efficiently separate mercury species in food, but also enhance the accuracy and reliability of analytical results, reduce contamination from closed systems, and enable simultaneous separation and quantification for precise analysis. Despite advancements over single-instrument analysis, instrumental coupling techniques have come a long way compared to single instrument analysis, these methods face limitations, including lengthy sample pretreatment, reliance on multiple organic solvents, time-intensive analyses, and complex instrument operation. Notably, 80% of the total analytical time for mercury analysis in food is dedicated to sample pretreatment [45]. Therefore, there is a pressing need to develop simpler, faster, more efficient, portable and environmentally friendly methods for mercury species analysis.

Direct mercury analysis (DMA)

Direct mercury analysis (DMA) determines total mercury in solid, liquid and even gaseous samples using a direct mercury analyzer, eliminating the need for complex sample preparation. In DMA, samples undergo high temperature combustion and catalytic pyrolysis, reducing mercury to monomers. These are then enriched via gold amalgam or directly transported into the detector by a carrier gas [40]. The mercury's atomic absorption is measured at 253.7 nm, or its atomic fluorescence is detected using a mercury lamp [40]. DMA avoids sample digestion and the use of hazardous chemical reagents, offering high sensitivity, rapid analysis, and environmental benefits.

Table-4: Detection techniques for speciation analysis of mercury in food [40-45].

Types	Detection technology	Detected mercury speciation
Instrumental analytical techniques	Atomic fluorescence fpectrometry (AFS)	Total mercury
	Direct mercury analysis (DMA)	Total mercury
	Inductively coupled plasma-mass spectrometry (ICP-MS)	Total mercury
	Cold vapor atomic absorption spectroscopy (CV-AAS)	Total mercury
Instrument coupling Technologies	Liquid chromatography - atomic fluorescence spectrometry（LC-AFS）	MeHg
	Liquid chromatography - inductively coupled plasma - mass spectrometry（LC-ICP-MS）	MeHg
Novel sensing and analyzing techniques based on nanomaterials	Surface enhanced raman spectroscopy (SERS)	Hg^{2+}
	Colorimetric method	Hg^{2+}, MeHg
	Fluorescence method	Hg^{2+}, MeHg
	Electrochemical method	Hg^{2+}, MeHg

Compared to above common instrumental coupling methods, DMA rivals high-end techniques like LC-ICP-MS in detection limits and sensitivity, offering notable benefits for complex food substrates such as fish and rice: no sample preparation, rapid results, and low cost. Chromatography hyphenation excels in mercury species analysis, whereas sensor technology boasts high sensitivity but struggles with interference. Thus, DMA is ideal for routine rapid screening of total mercury in food, while species analysis or extreme trace detection requires combined techniques.

DMA enables rapid detection and analysis within minutes by allowing direct sample introduction, significantly reducing the use of toxic chemicals, potential wastes and contamination risk. This makes DMA a pivotal tool in advancing analytical methods for direct-injection mercury measurement. Windmöller *et al*. [58] quantified total mercury in animal, vegetable, and fish and shellfish tissues, revealing discrepancies with other methods. The findings demonstrated that, compared to another mercury thermal desorption device with using continuous heating, DMA achieves detection and quantification limits as low as 1.04 ng/g and 4.32 ng/g, respectively, by using DMA, underscoring its capability for trace mercury analysis. Tan Wei *et al*. [59] investigated the factors affecting DMA in mercury determination in raw milk, finding optimal performance at a sample volume of 0.10 g, a decomposition temperature of 750℃, and a duration of 2 min, achieving a detection limit of 0.08 μg/kg, with high accuracy and reliability. Yao *et al*. [60] applied DMA to assess the variability in mercury measurement across solid foods of varying particle sizes. However, DMA demands high sample homogeneity, edible mushrooms exhibit greater homogeneity than unsieved rice. Accurate results are attainable when the food particle size is ≤425 μm (or sieve ≥40 mesh).

Surface enhanced raman spectroscopy (SERS)

Emerging mercury analytical techniques offer environmentally friendly, rapid, sensitive, cost-effective, and portable alternatives to traditional methods, which are hindered by cumbersome instruments, high costs, complex procedures, and organic reagent contamination [61]. Nanomaterials, with their unique physicochemical properties, have become pivotal in mercury detection. surface-enhanced Raman spectroscopy (SERS) addresses the limitations of conventional mercury analysis by providing a non-destructive, sensitive and efficient approach. SERS exploits induced excitation on nanomaterials surfaces to create an electromagnetic field, resulting in surface plasmon resonance that significantly enhances molecular scattering. This advancement boosts the sensitivity and specificity of SERS in the quantitative species analysis of mercury in food [62].

Liu *et al*. [63] developed a tapered optical fiber modified with AgNPs to detect Hg^{2+} and used Rhodamine 6G (R6G) as a SERS reporter. They observed that the spectral intensity of the SERS substrate decreases upon reaction with Hg^{2+}, enabling quantitative analysis by measuring the intensity reduction after 5 min. The detection limit of the method is 5.15×10^{-13} M, significantly lower than the USEPA standard of 10^{-8} M, offering trace and rapid, and ultra-sensitive detection. HASSAN *et al*. [64] introduced a SERS method combined with wave number selection chemometrics, utilizing core-shell Au@AgNPs as the SERS substrate and R6G as the signaling probe for Hg^{2+}. In the presence of Hg^{2+}, the citrate ions in Au@AgNPs facilitate complexation and amalgam formation, resulting in R6G desorption and a decrease in SERS signal intensity. The detection limit is 1.0×10^{-3} μg/g, enabling the determination of Hg^2 in fish and drinking water. Although nanomaterial-based SERS sensors offer

high sensitivity, most current SERS methods are limited to detecting Hg^{2+} and a struggle with MeHg detection (Table-5). Hg^2 achieves high selectivity through silver amalgamation or coordination chemistry. In contrast, methylmercury depends on molecular vibration recognition or advanced separation techniques and is prone to interference from coexisting organic molecules. Thus, achieve selectivity for specific targets, particularly in complex matrix samples, remains a significant challenge.

Colorimetric method

The colorimetric method quantifies target concentration by assessing the analyte solution's color change, offering advantages such as visualization, simplicity, speed and efficiency. This technique relies on the target molecules absorbing the UV-Vis light, causing electronic transitions that produces an absorption spectrum, thereby indicating the target's presence and species. Unlike complex instruments, the method is evolving from sensors based on intricate organic materials to those useing simple structure of precious metal nanoparticles (NPs). These NPs are favored in mercury detection in food due to their straightforward synthesis, environmental friendliness, low cost, and rapid response, which alters solution color through NP aggregation.

Early mercury colorimetric sensors relied on thiols or nitrogen-containing ligands reacting with Hg^{2+} to form complexes, but these methods suffered from complex procedures, low sensitivity, and poor selectivity for organic mercury. Researchers have been focused on developing colorimetric sensors with enhanced selectivity and reproducibility for mercury species analysis [73]. The incorporation of nanomaterials, such as gold nanoparticles (Au nanoparticles, AuNPs) and quantum dots (QDs), into the colorimetric sensor design has enabled the selective detection of various mercury species through surface modification. Typically, these sensors are equipped with sensitive elements and transducers that can specifically recognize and interact with mercury.

Among colorimetric sensors, biosensors are particularly well-studied for their ability to form a stable structure of T-Hg^{2+}-T by binding mercury ion to the thymine (T) base in oligonucleotides. These sensors interact specifically with nanomaterials like AuNPs, sulfur-containing groups or ligands through stable Hg-S bond, or with EDTA to form the HgY complex, significantly enhancing Hg^{2+} detection [74-75]. CAI *et al*. [~~71~~76] developed a platform where Hg^{2+} binds to thiol DNA-functionalized AuNPs (T-Hg^{2+}-T) via Au-S bonds. This binding increases spatial hindrance on AuNPs surface, weakening their catalytic activity in the p-nitrophenol and $NaBH_4$ reaction, thereby prolonging the color change time from yellow to colorless. Hg^{2+} concentration was quantitatively measured using the kinetic curve via UV spectrometry, achieving a detection limit as low as 0.20 nM, demonstrating excellent sensitivity. LIU *et al*. [77] introduced a novel colorimetric sensor utilizing on ribavirin-functionalized gold nanoparticles (Rib-AuNPs). Upon adding Hg^{2+}, the Rib-AuNPs solution color swiftly transitions from burgundy to gray-blue, with a naked-eye detection limit of 0.20 μM and a UV-visible spectroscopy detection limit as low as 3.64 nM. This sensor exhibits excellent selectivity and anti-interference, making it suitable for Hg^{2+} analysis in drinking water. LI *et al*. [~~73~~78] proposed a novel colorimetric nanoprobe leveraging the effect of MeHg on the nano selectivity enhancement NA-CDs/AuNPs. This system employs noradrenaline-carbon dots (NA-CDs) as a reducing agent to form gold-mercury amalgams (Au@HgNPs) with MeHg, accelerating electron transfer and free radical generation. This allows for specific and rapid visualization of MeHg within 10 min, achieving a detection limit of 0.06 μg L^{-1}.

Integrating DNA with NPs significantly enhances the development of sensitive and selective MeHg biosensors by enabling precise control over NPs self-assembly, structural arrangement and functional modification [79]. The evolving demands of diverse application scenarios and advancement in instrumentation have spurred the creation of intelligent and portable colorimetric sensors for mercury species analysis. Innovations include microfluidic-based colorimetric sensors, test-paper sensors utilizing AuNPs or functionalized dyes, and smartphone apps for image analysis that capture color changes, facilitating rapid detection and species isolation of trace samples. SHRIVAS *et al*. [~~75~~80] reported a colorimetric sensor using inkjet-printed paper with AgNPs, which transition from yellow to colorless based on mercury concentration. A smartphone app calculates the signal intensity after Hg^{2+} addition, achieving a detection limit of 10 μg·L^{-1}. The method is portable, user-friendly, and suitable for rapid on-site detection of Hg^{2+} in environmental water samples.

Table-5: A comparative table of the sensitivity and selectivity for Hg^{2+} vs. MeHg.

Target	Sensitivity (nM)	Materials	Selectivity	References
Hg^{2+}	5.15×10^{-4}	AgNPs@TOF probe, R6G reporter	Ultra-highly	[63]
	1.0 ng/g	Au@AgNPs substrate, R6G probe	Ultra-highly	[64]
	3.0×10^{-2}	Ag-Hg specific reaction	Ultra-highly	[65]
	5.0×10^{-7}	Au@Ag/COF substrate, Y-shaped DNA double-labeled reporter	Ultra-highly	[66]
	1.3	GA-CS@SeNPs	Ultra-highly	[67]
MeHg	25.13 (& 10.05)	Activity-based ratiometric nanoprobe	Highly	[68]
(&Hg^{2+})	10 (& 0.1)	4-MPBA dual-recognition probe	Highly	[69]

Recent advancements have seen the development of array sensors utilizing metal-organic frameworks (MOFs), covalent organic frameworks (COFs) and multifunctional materials, as well as nano-enzymes that mimic catalytic or sensing units. These innovations have propelled colorimetric sensors for Hg^{2+} analysis to a stage where they can where they can simultaneously detect multiple species with high selectivity. TARASI *et al*. [~~76~~81] designed a sacrificial metal-organic framework (SMOF), specifically $[Zn(oba)(RL3)_{0.5}]_n$-1.5DMF (TMU-59), which exhibits a unique interaction with Hg^{2+}. This interaction leads to the degradation of the MOF structure, causing a color changes from colorless to pink, easily visible to the naked eye for detecting and measuring Hg^{2+} in aqueous and non-aqueous solutions. Future sensor development will likely focus on biomass and other environmentally friendly materials, integrating AI technology to optimize the sensor design and data analysis for intelligent, real-time online monitoring and detection.

Among multiple nanoparticle-based sensors, there are two validated sensors for mercury detection in complex food substrates, such as seafood and plant foods like Pueraria lobata, utilizing nanoparticles for colorimetric analysis [82-83]. The first sensor, based on AgNPs-hydrophobic deep eutectic solvent (AgNPs-HDES) platforms, has a detection limit of 0.23 μM and offers semi-quantitative detection through color change, making it suitable for screening higher mercury concentrations. In contrast, a second sensor employs a dual-mode probe combining colorimetric and ratiometric fluorescence techniques, with a significantly lower detection limit of 27.47 nM (~0.027 μM), enhancing sensitivity. This dual-mode design ensures reliable detection at low concentrations, ideal for trace mercury analysis in plant-based food.

Fluorescence method

Fluorescence methods have addressed several limitations of colorimetric methods, notably their lack of sensitivity. Compared to colorimetric methods, fluorescence methods have better selectivity, reduced false positives/negatives, and applicability in live cell imaging. Since the synthesis of first chemical probe for mercury ions detection in 1992, numerous fluorescent probes have been gradually developed and refined [84]. Only a few organic molecules exhibit fluorescence, with traditional materials including fluorescein, rhodamine, coumarin, naphtholactam, fluoranthene, perylenetetracarboxylic diimide and their derivatives [85]. Due to the cytotoxicity and limited stability of these traditional materials, they have been increasingly supplanted by advanced photoluminescent materials such as inorganic quantum dots and metal nanoclusters (NCs). Nucleic acid aptamers have become prominent in mercury ion detection due to their high specificity and sensitivity. Currently, fluorescent probes for detecting mercury ions encompass conventional organic fluorescent probes, organic small molecule probes, nanomaterial probes, aptamer probes and MOFs [86].

Fluorescent probes detect substrates realize the recognition and detection of substrates through the by interacting recognition groups with the analytes, altering the fluorophores' physicochemical properties. The binding of mercury ions to the fluorophores modifies their structural or electronic properties, thus causing spectral changes like fluorescence enhancement or quenching. The primary mechanisms include chelation-enhanced fluorescence (CHEF), aggregation-induced enhancement (AIE), fluorescence resonance energy transfer (FRET), photoelectron transfer (PET), and excited-state intramolecular proton transfer.

MAHAJAN *et al*. [88] prepared rhodamine fluorescent organic nanoparticles, termed rhodamine Schiff base (FONs-RSB), which exhibit enhanced FONs-RSB fluorescence in the presence of t Hg^{2+} due to the CHEF phenomenon. This method achieves a detection limit as low as 1.68 ng/mL, suitable used

for environmental Hg^{2+} detection and intracellular imaging. AN *et al*. [89] designed a tetraphenylene (TPE)-based peptide fluorescent probe, TPE-Cys-Pro-Gly-His (TPE-CPGH), leveraging the AIE mechanism and peptide benefits. The sulfhydryl group of Cys in the peptide chain and the imidazole nitrogen in His mimics the Hg^{2+} binding site of the metalloprotein, facilitating the spatial coordination of TPE-CPGH with Hg^{2+} and forming ligand complex aggregates, leading to the fluorescence response. This method, with a detection limit of 46.2 nM, offers high specificity, sensitivity, and rapid response, along with low cytotoxicity and high-water solubility, effectively detecting Hg^{2+} in drinking water and reducing Hg^{2+}-induced oxidative stress toxicity in vivo and in vitro. TONSOMBOON *et al*. [~~83~~90] developed a fluorescence electrospinning fiber membrane utilizing the FRET mechanism, featuring the novel mercury-specific organic dye, NF06. This dye exhibited a fluorescence enhancement, achieving a detection limit of 0.309 ppb. The method is applicable for food safety assessment and market sampling because it as it requires only immersing the strips in Hg^{2+}-spiked drinking water. The resulting fluorescence change, indicative of Hg^{2+} concentration, is easily observed without instrumentation. MOF materials are prominent in mercury detection because of their large specific surface area, high stability, and tunable size. LI *et al*. [91] assembled a dual-emission double-emitting material (DEM) combining a zirconium-based organic framework (Zr-MOF) with AgNCs, achieving a very low detection limit of 0.309 ppb. This dual-emission ratiometric fluorescent probe is straightforward, fast, and highly sensitive, with a method detection limit of 1.8 μg L^{-1}. A smartphone-based colorimetric method was also developed to capture the fluorescence color change of this probe at different Hg^{2+} concentrations.

Detecting Hg^{2+} and MeHg in food via fluorescence is particularly challenging. OH *et al*. [~~85~~92] developed a rapid and sensitive fluorescence detection method for Hg^{2+} and MeHg based on a fluorescent probe comprising amino acids and SDS micelles. The method enhances red emission at the wavelength of 575 nm, with detection limits for Hg^{2+}, MeHg were 9.1 nM, 206 nM, respectively. The probe also facilitates real-time intracellular monitoring of Hg^{2+} and MeHg by amplifying red emission and green emission. DENG *et al*. [~~86~~93] developed a method for specific, selective detection of MeHg and Hg^{2+} using an Ag/Hg amalgamation with a MeHg signature DNA template, achieving detection limits at the pM level. The method is suitable for rapid, simple, and selective detection and monitoring of MeHg in environmental and biological samples.

Electrochemical method

Electrochemical methods translate chemical signals into electrical signals — current, potential or impedance — via the specific interactions between electrode surface modification materials and target mercury species. Commonly used methods include voltammetry, potentiometry and electrochemical impedance spectroscopy. Initially, electrochemical methods mainly utilized traditional voltammetry with unmodified electrodes for direct Hg^{2+} detection. These early methods suffered from low sensitivity, serious matrix interference and an inability to differentiate mercury species, although leveraging Hg^{2+} 's redox properties could enhance sensitivity through the enrichment-dissolution process [94].

Nanomaterials like nanoparticles enhance electrode modification by increasing specific surface area and electron transfer rates, while enabling selective adsorption through strong surface functional groups of nanomaterials with mercury ions. ZHANG *et al*. [~~88~~95] fabricated a highly sensitive electrochemical sensor using MnO_2/AuNPs composites and used the square-wave anode stripping voltammetry method to ~~to~~ detect MeHg in food samples. The MnO_2 and AuNPs-modified electrodes exhibited increased active surface area, conductivity, and electrocatalytic activity for MeHg, with good linearity and detection limits as low as 0.051 μg L^{-1} due to the synergistic effects of MnO_2 and AuNPs. LIU *et al*. [96] synthesized AuNPs using potassium borohydride (KBH_4) as a reductant on MOF. They developed an electrochemical sensor for detecting MeHg using a glassy carbon electrode modified with a zeolite imidazoline framework-67 (Au/ZIF67 GCE). This sensor achieved a detection limit of 0.05 μg/L, highlighting the exceptional performance of Au/ZIF67-modified GCE for MeHg detection.

Biomolecules such as aptamers were incorporated into the electrochemical sensor for specific recognition of organic mercury, including MeHg. Aptamers achieve selective binding to mercury species via base pairing or spatial conformation, enabling mercury species analysis with detection limits reaching the ppt level, thereby ensuring accurate detection in complex food matrices [~~90~~97]. While these electrochemical sensors based on aptamers exhibit strong reproducibility and stability in practical applications, their performance is largely

determined by sensor design, material selection, and environmental conditions. WANG *et al*. [98] developed a ratiometric electrochemical aptasensor for the simultaneous detection of trace Pb^{2+} and Hg^{2+} with high resistance to interference, applicable to vegetables containing these metals. This method employs metal-organic frameworks UiO-66-CNTs with stable electrochemical signals to load complementary strand (CS), while guanine- and thymine-rich oligonucleotides, labeled as carbon dots (CDs), serve as aptamers (Apts) that hybridize with the CSs to form M-type DNA complexes. The aptamers specifically capture Pb^{2+} and Pb^{2+}, forming Pb^{2+}-G-quadruplex and T-Hg^{2+}-T complexes, which disrupt the M-type DNA complexes and alter in CDs signals. The detection limits for this method were 2.0 ng mL^{-1} for Pb^{2+} and 0.5 ng mL^{-1} for Hg^{2+}.

The evolution of electrochemical sensors for mercury species in food has advanced a from detecting single inorganic mercury ion to precisely differentiating multiple species, transitioning from large laboratory instruments to rapid field screening [99]. With the integration of nanotechnology, artificial intelligence (AI) and microfluidics, this field is poised to advance towards fully automated miniaturization, intelligence, portability, and standardization, enhancing the efficiency of food safety monitoring.

Conclusions

As a toxic pollutant, mercury is extremely harmful to the human health and environment. This review describes detailly the heavy metal mercury contamination in food and the focus is on discussing several common techniques for analyzing the content of heavy metal mercury in food. Among several methods described above, the most suitable technique for on-site rapid screening without laboratory-level equipment is colorimetric method compared to others requiring specific sample pretreatment. In despite of a large amount of researches being performed over the past few decades, this review indicates that there are still many problems waiting to be resolved, particularly in mechanisms of mercury in the human body and simultaneous detection technologies in species analysis, such as efficient separation and enrichment of trace mercury in complex matrices, breakthroughs in sensitive and low-cost detection techniques, precise control of mercury species stability and dynamic transformations, and the application of multidisciplinary cross-fertilization. Therefore, it is necessary to continue research on mercury to reveal more meaningful information that singles out food safety and human life and health.

This review aims at providing technical inspiration for future studies on thorough mechanism of action and novel analytical methods of mercury. In the future, it is necessary to establish scientific predictive mechanism models that will contribute to security risk assessment and speciation analysis of mercury in food so as to develop more rapid, sensitive, selective, and cost-effective methods. For example, fully automated miniaturized equipment, the combination of Internet of Things and AI technologies and the design of DNA nanomachines are the development trends of heavy metal mercury contamination monitoring and species analysis in food. Additionally, single-particle (cell) analysis can be also used to reveal the microscopic distribution of mercury species in food and their interaction mechanism with biological macromolecules, realizing mercury pollution source tracking and precise prevention and control according to the existing blockchain traceability technology.

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