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REFILL DRINKING WATER DEPOT RISK ASSESSMENT FOR CHEMICAL HAZARD CONTAMINANT IN 25 CITIES OF EAST JAVA PROVINCE, INDONESIA

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Abstract

Introduction: *Water was a part of the environment that was very susceptible to contamination by various hazardous substances, both physical, chemical, and microbiological. Serious health problems might occur when humans consume contaminated drinking water and can cause infectious and non-infectious diseases. This study aimed to analyze the risk quotient of chemical hazard exposure in drinking water by depot refilling drinking water from 25 cities and regencies in East Java Province.* ***Methods:*** *This study was analytically observational with a cross-sectional research design. The sampling method used was porposive sampling, which took 25 from 38 cities and regencies in East Java province. There were three steps to analyze drinking water health risks: dose-response analysis, exposure path, and risk characteristics analyzed.* ***Results and Discussion:*** *The highest concentrations of each parameter parameter F (1.4132 mg/L),* NO_3 *(56.2780 mg/L),* NO_2 *(22,225 mg/L), Fe (0.3845 mg/L), Mg (4.54 mg/L), and Zn (0.4272 mg/L). Analysis of risk quotient > 1 for parameters* NO_3 *(1.2265),* NO_2 *(7,748), and Mg (1.1308) Indicates that drinking water in the area is not safe for consumption by people weighing 55kg as much as 2L/day for 350 days.* ***Conclusion:*** *Based on the results of studies conducted on those substances (F,* NO_3*,* NO_2*, Fe, Mg, and Zn) there are three substances in the water that have RQ value > 1,* NO_3 *(1.2265),* NO_2 *(7,748), and Mg (1.1308) this means that those substances are risking human health and needed to be addressed controlled.*

INTRODUCTION

Clean water crises such as water scarcity and polluted water are huge environmental and health problems at once since water is one of the most important daily needs, and it is supposed to be hygienic, safe, and healthy for the body (1). Therefore, drinking water sources and their quality must be assured before people consume them. Polluted drinking water may lead to numerous acute or chronic diseases that might be caused by the unwanted substances contained in the drinking water that are consumed by people every single day of their lives, including chemical substances (2). The unwanted chemical substances such as heavy metals (Pb, Cd, Cr, Cu, etc) that people consume for a long time could accumulate in their organs. These prolonged accumulations of unwanted chemical substances

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can increase the risk of getting non-carcinogen or carcinogen health problems (2). Conversely, some drinking water sources may contain chemical substances that could harm the human body from underground water, rainwater, or other water sources (2-3).

WHO (2015) data said that 1.8 billion people drink from contaminated water (4-5). Thus, people consume the unwanted substances in the water and risk getting health problems. One of the studies that had been done said that contaminated water with a high level of nitrate, which is a non-carcinogenic chemical, can lead to baby blue syndrome in infants and may lead to cancer and fecal defects (6). Another study in Northern Lao said that contaminated water is one of the factors that caused intestinal parasite infections (7). Other studies done in Northwest China said that more than 50% of groundwater samples require treatment before being consumed by people, and half the population of that study's area faced non-carcinogenic health risks including the children who are the susceptible group in the population (8).

Numerous studies also had been done to assess chemical substances in the groundwater that many people use this resource as their drinking water as well, and one of them had been done on the southwestern coast of Bangladesh said that the groundwater used by the community has a high level of salinity, and other substances such as EC, Cl^-, HCO_3^-, NO_3^-, PO_4^{3-}. This study considered land use and massive anthropogenic disruption in assessing the water quality (9). Anthropogenic disruption such as urbanization or industrialization, is one of the continual contaminant sources that can affect water quality (1). Another study in India stated that pesticide pollution in groundwater was observed in 90% of water samples groundwater, where groundwater is the main source of drinking water for the human population in India. Pesticides found in groundwater were DDT, lindane, parathion, and tetrachlorvinphos, which are carcinogenic chemicals that may lead to cancer in humans and were categorized into Group 2A, Group 1, Group 2B, respectively (10-11).

Drinking water can become contaminated by heavy metals (loids), which result from both natural processes and human activities. The levels of heavy metal(loids) in natural water depend on factors like the underground water source's geological conditions, hydrogeology, and geochemical properties. These factors include the weathering and erosion of rocks and the presence of ore deposits. On the other hand, human activities like electroplating, metal smelting, mining, manufacturing, and the release of agricultural wastewater can contaminate the water source with heavy metals (12–14). Contaminants inside drinking water need to be eliminated before people consume the water. Carcinogenic contaminants are dangerous for the environment and human body, but non-carcinogenic contaminants also can affect health problems and hospitalizations in society. Since both kinds of contaminants are detrimental to both the environment and humans, risk assessment in drinking water is needed for carcinogenic and non-carcinogenic chemicals.

Data from the Indonesia Central Bureau of Statistics said that 25.81% of families in East Java used refilled water or bottled water for their drinking water sources, 32.30% of families used plumbing water, 36.75% of families used protected spring water, 4.61% of families used underground water, and 0.62% used river water or rainwater or other sources for their drinking water 12. Those families who consume drinking water from refilled water still can't be assured that the water is safe to consume without being processed. Based on the results of BPOM monitoring of refilled water in East Java in 2022, several chemicals were found in refilled water in East Java with contamination levels that exceeded safe limits. These chemicals include Fe (10%), Mn (5%), NO_2 (2%), NO_3 (3%), F (1%), and Zn (0.5%) (15).

The sources of chemical contamination are varied, including groundwater and surface water pollution, using iron and galvanized pipes, and using chlorine as a disinfectant. Only a few studies have analyzed the non-carcinogenic chemical substance dissolved in drinking water in Indonesia. Subsequently, this study is aimed to analyze chemical hazards that may lead to health problems in 25 Regencies of East Java.

METHODS

This study's research type was analytic observational with a cross-sectional research design. The sampling method used purposive sampling, taking 25 regencies and cities from 38 regencies in East Java province. The total number of drinking water samples analyzed was 1,113 from the Refill Drinking Water Depot.

The sampling method for drinking water sampling was conducted regarding SNI 7828:2012. The bottle used for sampling was made from materials that do not affect the sample's characteristics and are easy to wash, and then the water samples were stored in a clean and sterile bottle. The water sample volume was 500 mL or more, with water directly caught from the refill faucet at the Refill Drinking Water Depot in each city/regency. The samplers were the Center for Environmental Health Engineering and Disease Control officers with the acronym BBTKLPP Surabaya. The Surabaya BBTKL Laboratory tested water samples. The chemical parameters tested were Fe (Quality Standard 0.3 mg/L), F (Quality Standard 1.5 mg/L), Mn (Quality Standard 0.4 mg/L), NO_2 (Quality Standard 3 mg/L), NO_3 (Quality Standard 50 mg/L), Zn (Quality Standard 3 mg/L). Those standards were based on the Regulation of Indonesia Health Ministry no. 022023. In identifying the presence of chemical substances contained in water, the following method will be used.

Table 1. The Distribution of Drinking Water Sample

Regency	Amount of Sample
Bangkalan	7
Banyuwangi	11
Bojonegoro	57
Bondowoso	19
Gresik	82
Jombang	70
Mojokerto	36
Kediri	8
Jember	38
Lamongan	4
Lumajang	1
Malang	5
Madiun	1
Nganjuk	5
Pasuruan	4
Pasuruan	173
Probolinggo	2
Pamekasan	3
Sampang	14
Sumenep	27
Sidoarjo	376
Surabaya	122
Situbondo	33
Trenggalek	11
Tuban	4
Total	1,113

Table 2. Method of Identifying the Presence of Chemical Substances Contained in Water

Chemical Substance	The Method	The Superiority of the Method
Fe	Atom Absorption Spectrophotometric (AAS) method (SNI 6989.4:2009).	This method is sensitive to the concentration of Fe around 0,3 mg/L – 10 mg/L, then the AAS tools will be on.
F	Analysis principle of spectrophotometric UV-VIS with the SPADNS reagent.	This method is sensitive to determine F by comparing the reagent volume at 0.5 mL and 1 mL within 5 mL of standard fluoride solution in three different levels of 0.4 mg/L, 1 mg/L, and 1.8 mg/L respectively. Then, the measurements were made with a spectrophotometer UV-VIS at 550 nm.
Mn	Atom Absorption Spectrophotometric (AAS) method (SNI 6989.5:2009).	This method is sensitive to the concentration of Mn around 0,1 mg/L – 10 mg/L, then the Atom Absorption Spectrophotometric (AAS) tools. will be on
NO_2	Atomic Absorption Spectrophotometric (AAS) method (SNI 06-6989.9-2004).	This method is sensitive to the concentration of NO_2 around 0,01 mg/L – 1 mg/L, then the AAS tools will be on.
NO_3	Spectrophotometric UV visible (SNI 6989.79:2011)	This method is sensitive to determine NO_3 concentration from cadmium reduction of nitrate that will results pink color which indicate the NO_3 absorption.
Zn	Atomic Absorption Spectrophotometric (AAS) method (SNI 6989.7:2009)	This method is sensitive to the concentration of Zn around 0,05 mg/L – 2 mg/L, then the AAS tools will be on.

Dose-Response Analysis

To determine the quantity of risk agent toxicity for each chemical species. Toxicity was expressed as a reference dose based on the Integrated Risk Information System US EPA (RfD for ingestion exposure and RfC for inhalation exposure).

Table 3. RfD/RfC Value and the Critical Effects

Risk Agent	RfD (mg/kg/day)	Critical Effects and References
Fe	3×10^{-1}	Causes disruption of oxygen absorption in the blood by symptoms of dizziness, nausea. If consumed in high amounts it can injure the nerves (15)
F	6×10^{-2}	Dental fluorosis and cosmetic effects in epidemiological studies (16)
Mn	1.4×10^{-1}	Hypocholesterolemia, epilepsy, exocrine pancreatic insufficiency, multiple sclerosis, neurotoxic (17)
NO_2	1×10^{-1}	Methemoglobinemia in infants chronically exposed to nitrogen drinking water (18)
NO_3	1.6	Early signs of methemoglobinemia in infants 0-3 months. Epidemiological survey (19)
Zn	3×10^{-1}	Erythrocyte Cu decrease and Zn superoxide dismutase activity in male and female volunteers (20)

Exposure Path

This study exposes risk agents (pollutants) from natural nutrient contents, household waste, and industrial waste contamination to enter the human body by ingestion (swallowed) through drinking water. The people most often exposed are residents around the location (residential). The following formula can calculate the amount agent of risk intake:

$$I = \frac{C \times R \times t_E \times f_E \times D_t}{W_b \times t_{avg}}$$

Notes:

I = Intake, the amount of intake risk agent (mg/kg/hr)

C = Concentration of risk agent, mg/m³ for air medium, mg/L for drinking water, mg/kg for food

R = Intake rate, 20 m³/hr or 0,83 m³/hr (air), 2 L/hr (drinking water)

fE = Annual exposure frequency, 350 days/year,

Dt = Exposure duration, real-time or 30 years projection

Wb = Weight, 70 kg / 55 kg (70 kg based on US-EPA 1990, 55 kg,

t_{avg} = Period average, 30 years x 365 days/year (non-carcinogenic) or 70 years x 365 days/year (carcinogenic).

Risk Characteristics

Health risk characteristics are expressed as Risk Quotient (RQ, risk level) for non-carcinogenic effects and Excess Cancer Risk (ECR) for carcinogenic effects. Fe, F, Mn, NO_2, NO_3, and Zn are classified as non-carcinogenic chemicals. Then the formula used is:

$$RQ = \frac{I_k}{RfD\ atau\ RfC}$$

Notes:

RQ = Risk Quotient

Ink = Intake non-carcinogenic

RfD = Reference Dose (for the ingestion exposure)

RfC = Reference Consentration (for the inhalation exposure)

In this study, the RQ will be calculated based on the maximum concentration results so that we can determine the maximum hazard risk from the maximum concentration. If the RQ value is more than 1 (> 1), the chemical substance has a high risk to human health and needs to be controlled as soon as possible from January to December 2019.

RESULTS

Based on the results of water chemistry laboratory tests of drinking water samples taken from several locations in East Java at 25 cities and regencies depot collection points, it shows 6 chemical substances (Fe, Mn, NO_2, NO_3, F, and Z) that are considered health risks when consumed over a certain period.

Table 4. Maximum Concentration Value of 6 Drinking Water Test Parameters in 25 Cities and Regencies in January-December in East Java

City/ Regency	Fluoride QS : 1.5 mg/L	Nitrate QS : 50 mg/L	Nitrite QS : 3 mg/L	Iron QS : 0,3 mg/L	Manganese QS : 0,4 mg/L	Zinc QS : 3 mg/L
Bangkalan	0.6915	10.207	0.0188	0.1539	0.0491	0.0075
Banyuwangi	1.3000	11.734	0.1857	0.0037	0.0671	0.0075
Bojonegoro	1.3907	33.9950	0.2248	0.0482	0.1083	0.0075
Bondowoso	0.9584	**55.8450**	0.1501	0.0037	0.0491	0.0075
Gresik	1.3876	31.5980	0.7044	0.2151	**2.7334**	0.0075
Jombang	1.1245	14.5570	1.6088	**0.3845**	**1.1508**	0.1192
Mojokerto	1.0031	18.9850	0.0511	**0.3721**	0.1536	0.0075
Kediri	0.5359	0.2030	0.0272	0.0037	0.0965	0.0075
Jember	1.0803	40.5600	2.2361	0.0037	**0.8052**	0.3724
Lamongan	1.0080	7.8380	0.0187	0.0037	0.0491	0.0075
Lumajang	0.4600	6.2170	0.0021	0.0037	0.0491	0.0075
Malang	1.4132	45.6620	0.1084	0.0037	0.0491	0.0075
Madiun	0.3515	0.0019	0.0104	0.0037	0.0491	0.0075
Nganjuk	0.7962	2.4590	0.1606	0.0037	0.0491	0.0075
Pasuruan	0.1766	0.0019	0.0316	0.0037	0.0491	0.0075
Pasuruan	1.3455	39.5880	1.2759	0.1617	**0.4241**	0.0383
Probolinggo	0.4782	13.9750	0.0328	0.0037	0.0491	0.0075
Pamekasan	0.3645	9.1200	0.0021	0.0037	0.0491	0.0075
Sampang	1.1568	15.0400	0.0021	0.0037	0.0491	0.0075
Sumenep	1.0173	47.3170	1.3757	0.0037	**0.6084**	0.0075
Sidoarjo	1.3914	18.6270	**6.9620**	0.1905	**4.5400**	**0.4272**
Surabaya	1.4083	**52.1010**	**22.2225**	0.0962	0.1005	0.0119
Situbondo	1.0918	**56.2780**	0.0281	0.2612	0.0491	0.0827
Trenggalek	1.0161	3.5560	0.0232	0.0037	0.0491	0.0075
Tuban	0.6447	32.1420	2.0440	0.0037	0.0491	0.0075

Notes:

QS = Quality Standard of Drinking Water Based on Permenkes RI No.492/2010

Red Fonts = Concentration more than the quality standard

Based on the RQ (Risk Quotient) table above, it can be seen that cities and regencies that have an RQ value > 1 are Bondowoso, Sumenep, Surabaya and Situbondo for the Nitrate (NO_3) parameter, Sidoarjo Regency and Surabaya City for the Nitrite (NO_2) parameter, and Sidoarjo Regency for Manganese parameter. An RQ value > 1 indicates that people weighing 55 kg are not safe to drink water in the area with an intake rate of 2 L/day for 350 days/year for the next 30 years if the maximum concentration value of water chemical risk agents is not more than table 5.

Besides the chemical concentration dissolved in the water, this study also analyzed the risk quotient (RQ) to determine the hazard risk resulting to the human body. Based on Table 5 from six substances analyzed in this study, three substances (nitrate, nitrite, and manganese) have RQ > 1 with the quality of standard based on the Regulation of Indonesia Health Ministry no. 02 2023. For nitrate, RQ values are around 0.000 – 1.2265, and the comparison of the RQ value > 1 is Situbondo > Bondowoso > Surabaya > Sumenep. Nitrite RQ values are around 0.0007 – 7.7488, and the comparison of the RQ value > 1 is Surabaya > Sidoarjo. The last substance has Mn RQ values of around 0.0122 – 1.1308, with the highest RQ value in Sidoarjo. Those cities and regencies with RQ values > 1 mean that their existence in the water is risking human health and must be controlled as soon as possible.

Table 5. Risk Level of Non-Carcinogenic Impact Due to Exposure to Water Pollutants Expressed in RQ Value

City/ Regency	Fluoride	Nitrate	Nitrite	Iron	Manganese	Zinc
Bangkalan	0.4019	0.2224	0.0065	0.0178	0.0122	0.0009
Banyuwangi	0.7555	0.2557	0.0647	0.0004	0.0167	0.0009
Bojonegoro	0.8082	0.7409	0.0784	0.0056	0.0270	0.0009
Bondowoso	0.5570	**1.2170**	0.0523	0.0004	0.0122	0.0009
Gresik	0.8064	0.6886	0.2456	0.0250	0.6808	0.0009
Jombang	0.6535	0.3172	0.5610	0.0447	0.2866	0.0139
Mojokerto	0.5829	0.4137	0.0178	0.0432	0.0383	0.0009

City/ Regency	Fluoride	Nitrate	Nitrite	Iron	Manganese	Zinc
Kediri	0.3115	0.0044	0.0095	0.0004	0.0240	0.0009
Jember	0.6278	0.8839	0.7797	0.0004	0.2005	0.0433
Lamongan	0.5858	0.1708	0.0065	0.0004	0.0122	0.0009
Lumajang	0.2673	0.1355	0.0007	0.0004	0.0122	0.0009
Malang	0.8213	0.9951	0.0378	0.0004	0.0122	0.0009
Madiun	0.2043	0.0000	0.0036	0.0004	0.0122	0.0009
Nganjuk	0.4627	0.0536	0.0560	0.0004	0.0122	0.0009
Pasuruan	0.1026	0.0000	0.0110	0.0004	0.0122	0.0009
Pasuruan	0.7820	0.8628	0.4449	0.0188	0.1056	0.0045
Probolinggo	0.2779	0.3046	0.0114	0.0004	0.0122	0.0009
Pamekasan	0.2118	0.1988	0.0007	0.0004	0.0122	0.0009
Sampang	0.6723	0.3278	0.0007	0.0004	0.0122	0.0009
Sumenep	0.5912	**1.0312**	0.4797	0.0004	0.1515	0.0009
Sidoarjo	0.8086	0.4059	**2.4276**	0.0221	**1.1308**	0.0497
Surabaya	0.8185	**1.1355**	**7.7488**	0.0112	0.0250	0.0014
Situbondo	0.6345	**1.2265**	0.0098	0.0304	0.0122	0.0096
Trenggalek	0.5905	0.0775	0.0081	0.0004	0.0122	0.0009
Tuban	0.3747	0.7005	0.7127	0.0004	0.0122	0.0009

Table 6. Risk Management Calculation Results to Lower Concentration and Intake Rate

Parameter in City/Regency	Original C mg/L	Safe C mg/L	Original Intake L/hr	Safe Intake
NO_3				
Bondowoso	55.8450	45,8857	2	1,6433
Sumenep	47,3170	45,8857	2	1,9395
Surabaya	52,1010	45,8857	2	1,7614
Situbondo	56,2780	45,8857	2	1,6306
NO_2				
Sidoarjo	6,9620	2,8678	2	0,8238
Surabaya	22,2225	2,8678	2	0,2581
Mn				
Sidoarjo	4,5400	4,015	2	1,7687

DISCUSSION

There are several refilled drinking water resources from the depot in which the people consume the water, and many directly consume it without being processed before. Numerous studies have justified that several unwanted chemical substances are dissolved in the water (1,6,8). Fe, Mn, NO_2, NO_3, F, and Zn are non-carcinogenic chemicals that might cause health problems. Water contaminants could come from industrialization, urbanization, and other human activities that generate waste. The wastes frequently contain hazardous materials such as heavy metals, which are dangerous for the environment's sustainability and human health. Thus, drinking water risk assessment needs to be done in the middle of developing paradigms in society that they tend to ignore their health as long as they can work and walk (16).

Based on Table 4, water samples from two regencies (Jombang and Mojokerto) contain Fe more than the quality standard of drinking water. Also, water samples from six regencies (Gresik, Jombang, Jember, Pasuruan, Sumenep, and Sidoarjo) contain Mn more than the quality standard of drinking water from the regulation of Indonesia Health Ministry. Manganese contamination in refilled water is a problem that needs to be watched out for. This contamination can occur due to various factors, such as contaminated raw water sources, inadequate processing processes, and improper storage and distribution (17-18).

Excess Iron (Fe) and Manganese (Mn) in adults is known as a neurotoxin that can lead to other health problems in pregnant women and newborns because Mn readily crosses the blood-brain barrier in the developing fetus, neonate, and even adults (21-22). Proven by one of the studies that had been done on children who lived close to a ferromanganese plant in Brazil said that the higher the Mn concentration contained in the drinking water, the lower the IQ score in children. It showed that the statistical test for children's Mn in hair (MnH) was negatively related to Full-Scale Intelligence Quotient (IQ) and Verbal −5.78 and −6.72 (23). Another study said that iron (Fe) overexposure from underground water can accumulate in hair and nail tissue (24). Iron (Fe) Iron ingested in large doses in drinking water can damage the intestinal walls and cause death (25). Manganese (Mg) in water When consumed in specific amounts, Manganese can enter the blood and brain (26).

Water samples from two-three regencies and one city (Bondowoso, Surabaya, and Situbondo) based on Table 4 contain more nitrate than the quality standard. Water samples from one regency and one city (Sidoarjo and Surabaya) contain more nitrite than the quality standard of drinking water according to the regulations of the Indonesia Health Ministry. Nitrite and nitrate contamination in refilled water can occur due to various factors, such as contaminated raw water sources, inadequate treatment processes, and improper storage (24). Blood that contains a high level of nitrite known as methemoglobinemia, can inhibit oxygen transport throughout the body. Nitrate-nitrite poisoning found in babies changes their skin into blue and is also known as a blue baby syndrome that usually affects 3–6 weeks old infants (27). In another case, nitrite and ammonia will form nitrosamine that contaminate foods and cause liver damage, lung rash, convulsion, and coma (28). Nitrates (NO_3) and Nitrites (NO_2), as well as long-term exposure to nitrites, can cause skin damage and nitrite poisoning (29).

Based on Table 4, water samples from 1 city (Sidoarjo) contain more Zinc (Zn) than the quality standard of drinking water from the regulation of Indonesia Health Ministry, and Fluoride (F) has a safe concentration in all water samples from all cities and regencies in East Java. They must be maintained to prevent health problems caused by excess Zn and F. Excess Zinc can cause gastrointestinal and immunologic problems. Excess Fluoride can cause dental cavities and urinary tract disease (6,30). Fluoride in drinking water can also be a risk of dental fluorosis, in larger amounts, it can cause bone fluorosis (31). Zinc (Zn) in water causes a rough feeling and can cause vomiting and stool symptoms (25).

Table 6 shows that water treatment efforts are needed to reduce the concentration of Nitrate, Nitrite, and Manganese from the original concentration (original C) to a safe concentration (safe C). The processing that can be done to reduce nitrate and nitrite is an aeration system or reverse osmosis. Meanwhile reducing the concentration of Manganese can be done by aeration method, resin, and brown zeolite. If the processing cannot be carried out, it is necessary to reduce the initial intake rate (original R) to a safe intake rate (safe R). If the intake rate is reduced, it needs a replacement to increase the volume of people's daily drinking water. Replacement drinking water can be obtained from gallons or refills that have better quality (30-31).

This research assessed non-carcinogen chemical concentration and RQ in refill water samples only. Different chemicals could be found if the water samples used are from tap water, groundwater, rainwater, or other water resources. Therefore, further research is needed for other non-carcinogen and carcinogen chemicals from various water resources.

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CONCLUSION

Drinking water consumed by society needs regular assessment for its hygiene, cleanliness, and safety, given that individuals typically consume 2 liters daily. Water samples were collected from refill water depots across 25 cities and regencies in East Java and analyzed for chemical contaminants. The study identified six chemicals with the highest levels of contamination: Fe, Mn, NO_2, NO_3, F, and Zn. While these chemicals are non-carcinogenic to humans, overexposure can lead to health problems such as central nervous system disorders, blood issues, and urinary tract complications.

The study revealed that three substances in the water samples had Risk Quotient (RQ) values exceeding 1, indicating potential risks to human health. Urgent control measures are necessary to mitigate these risks and prevent health problems or outbreaks. One effective control strategy is to reduce these substances' concentration and intake rate.

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