Study on Clearance Discharge Limits for Medical Radionuclides 99mTc, 131I, 90Y, 223Ra in Wastewater

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Date: 2025-12-16T14:00:15+00:00

Abstract

The radionuclides present in wastewater generated by medical radioactive nuclides have relatively short half-lives, with correspondingly short discharge cycles. The discharge of medical radioactive nuclide wastewater results in relatively low radiation doses to both the public and the environment. Based on the principle of radiation protection optimization and environmental discharge regulatory requirements, clearance discharge management approaches can render the discharge of these radionuclides more reasonable. This study investigates clearance discharge limits for medical radioactive nuclide wastewater. Based on the environmental impact assessment model for radioactive substance discharge recommended by the International Atomic Energy Agency, this study analyzes clearance discharge limits for four medical radioactive nuclides in wastewater: 99mTc, 131I, 90Y, and 223Ra. The clearance limits for these four nuclides are 3.14\$$\times 10^{8} Bq/a, 2.75\times 10^{7} Bq/a, 4.30\times 10^{7} Bq/a, and 1.45\times$\$10^7 Bq/a, respectively. By determining the annual production volume of radioactive wastewater from medical radionuclide application facilities, the annual discharge can be converted to discharge activity concentration values.

Full Text

Research on Clearance Levels for Liquid Discharge of Medical Radioisotopes 99mTc, 131I, 90Y, and 223Ra

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Abstract

Medical radioisotopes typically have short half-lives, and the resulting wastewater discharge periods are correspondingly brief. The radiation doses to the public and the environment from such discharges remain relatively low. Based on the principle of radiation protection optimization and environmental discharge regulatory requirements, a clearance discharge management approach can rationalize the release of these radionuclides. This study investigates clearance levels for wastewater discharge from medical radioisotopes. Using the environmental impact assessment model for radioactive substance discharge recommended by the International Atomic Energy Agency (IAEA), we analyzed clearance levels for liquid discharge of four medical radioisotopes: 99mTc, 131I, 90Y, and 223Ra. The derived clearance levels are 3.14$×10^{8}$ Bq/a, 2.75$×10^{7}$ Bq/a, 4.30$×10^{7}$ Bq/a, and 1.45$×10^{7}$ Bq/a, respectively. By determining the annual volume of radioactive wastewater generated by medical institutions, these annual discharge limits can be converted to activity concentration values.

Keywords: medical radioisotope; wastewater; clearance; limits

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1. Introduction

1.1 Medical Radioisotopes

Medical radioisotopes are characterized by short half-lives, low toxicity, and rapid excretion, making them suitable for diagnostic and therapeutic applications. In nuclear medicine diagnostics, the most common isotope is the gamma-emitting radionuclide 99mTc with a half-life of 6.01 hours, while the most widely used therapeutic radionuclide is the gamma-emitting 131I with a half-life of 8.02 days. With advances in pharmaceutical preparation technology, novel beta- and alpha-emitting radiopharmaceuticals have been approved for clinical use. For example, 90Y microspheres, a beta-emitting radionuclide with a half-life of 2.67 days, are used for liver cancer treatment, while 223Ra, an alpha-emitting radionuclide with a half-life of 11.435 days, is employed in treating castration-resistant prostate cancer.

According to GB18871-2002 (Basic Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources), the discharge of radioactive substances to the environment must not exceed limits approved by the regulatory authority, including both total activity and concentration limits. Wastewater containing radioactive isotopes may be discharged into conventional sewer systems if the activity per discharge does not exceed 1 ALImin (Annual Limit on Intake) and the total monthly discharge does not exceed 10 ALImin, subject to regulatory confirmation. The standard HJ1188-2021 (Radiation Protection and Safety Requirements for Nuclear Medicine) stipulates that nuclear medicine facilities should install decay tanks (either batch or plug-flow type) or dedicated containers, with batch decay tanks required for facilities with 131I therapy wards.

For wastewater containing radionuclides with half-lives shorter than 24 hours, the minimum storage time is 30 days; for radionuclides with longer half-lives, the storage time should be ten times the longest half-life present. Additionally, the total discharge outlet must meet activity concentration limits of 1 Bq/L for total alpha, 10 Bq/L for total beta, and 10 Bq/L for 131I.

Current Chinese standards provide principle-based discharge requirements for radioactive wastewater but lack specific provisions for implementing clearance discharge for various short-half-life radioisotopes used in nuclear medicine. To address this gap, this study derives clearance levels for short-half-life radioisotopes suitable for clearance discharge. Based on IAEA safety standard guidelines on clearance and dose assessment models for liquid effluents, we evaluated clearance discharge limits for four representative medical radionuclides: the short-half-life (<24 h) gamma emitter 99mTc, the volatile radionuclide 131I, the beta emitter 90Y, and the alpha emitter 223Ra. The assessment considered two scenarios (sludge and river water) and used a dose constraint of 10 Sv/a for public exposure from radionuclide discharge.

1.2 Discharge Scenario Considerations

Medical radioactive wastewater in China typically undergoes storage and decay until it reaches clearance levels, followed by treatment at dedicated wastewater treatment stations before discharge to municipal sewage systems. Due to variations in the construction timing of municipal treatment facilities, some stations require manual sludge removal to transport vehicles, while others have direct sludge connections without manual handling. Small-to-medium wastewater treatment stations generate 3–6 tonnes of sludge, which is transported to incineration plants, while the treated water is discharged to nearby rivers or reservoirs.

Based on these discharge pathways, radionuclides may remain either in the sludge from wastewater treatment stations or in the treated water. To conservatively assess radiation impacts, we assume two extreme scenarios: all radionuclides remain in the sludge, or all remain in the treated water. The sludge scenario considers external exposure from sludge at the treatment station and exposure to incineration plant workers. The river water scenario considers radionuclides discharged into surface water bodies, where they may deposit on riverbanks, be ingested by fish, or be used for irrigation, potentially exposing downstream populations through fish consumption or ingestion of irrigated crops.

The IAEA report "Clearance of Materials Resulting from the Use of Radionuclides in Medicine, Industry and Research" derives general clearance levels for radionuclides used in these fields, including example calculations for 131I. The assessment of liquid effluent environmental impacts considers two scenarios: complete retention in sludge and complete retention in water. The sludge scenario accounts for exposure to treatment station workers but not incineration plant workers, while the water scenario considers water ingestion, shoreline deposition, and fish consumption.

Our study selects realistic exposure scenarios and pathways based on the actual fate of nuclear medicine wastewater in China. We conservatively evaluate potential public exposure from radionuclides in medical wastewater, considering manual sludge removal, sludge incineration, and potential water uses, while excluding direct river water consumption.

2. Methodology

2.1 Overview of the Derivation Process

The study follows these steps to derive clearance levels for radionuclide wastewater discharge:

1. Identify exposure scenarios and pathways following wastewater discharge. Medical wastewater is typically treated at dedicated stations before municipal discharge. We consider radionuclide retention in sludge or treated water, with conservative assumptions of complete retention in either medium.
2. Calculate activity concentrations in environmental media for both sludge and river water scenarios based on unit annual discharge.
3. For the sludge scenario, calculate doses from: ground deposition external exposure, resuspended sludge inhalation, incineration inhalation, and incineration immersion external exposure. For the river water scenario, calculate doses from: shoreline deposition external exposure, fish ingestion internal exposure, and crop ingestion internal exposure. Sum the doses from all pathways for each scenario.
4. Derive clearance levels for each scenario by multiplying the unit discharge by the ratio of the selected dose management value (10 Sv/a) to the calculated dose. The final clearance level for each radionuclide is the minimum value among the scenarios.

The derivation process is illustrated in Figure 1.

[Figure 1: see original paper]

2.2 Discharge Models

2.2.1 Sludge Discharge Model This model assesses human exposure when radionuclides from wastewater discharge remain entirely in sludge, considering both wastewater treatment station workers and incineration plant workers.

Assuming a unit discharge of 1 Bq of a radionuclide in annual wastewater, we calculate the potential dose to relevant workers. For conservative estimation, we use a small treatment station’ s sludge production of 3 tonnes (1$×10^{6}$

kg/day). With a sludge density of 1000 kg/m^3 and depth of 1 m, the surface activity concentration of radionuclides in sludge (Cslu,sur) is 1×10^{-3} Bq/m^2.

Sludge from treatment stations is sent to incineration plants. Based on relevant reports, we conservatively assume a municipal solid waste incineration plant with a daily capacity of 50 tonnes, including 3 tonnes of radionuclide-containing sludge. Following the Ministry of Housing and Urban-Rural Development's "Technical Standard for Building Smoke Control and Exhaust Systems" (GB51251-2017), we select a conservative exhaust airflow rate of 5 m^3/s.

The concentration of radionuclides in air during sludge incineration (Ccin) is calculated as:

Ccin = (Pp × Qai) / (Qv × F)

where Qai is the average atmospheric emission rate during incineration of radionuclide i (Bq/s), calculated as:

Qai = (Ai × FRFi) / Tr

where Ai is the activity of radionuclide i in sludge during incineration, FRFi is the release fraction during combustion (1×10^{-2} for 99mTc, 0.5 for 131I, 1×10^{-2} for 90Y, and 1×10^{-3} for 223Ra), and Tr is the combustion duration (1 hour/day for 250 days/year, totaling 250 h/a).

2.2.2 River Water Discharge Model This model assesses exposure when liquid wastewater discharge remains entirely in the water phase and is released into a river. We assume a river width ($\bar{B}$) of 50 m, with residents using the river water for fish farming and crop irrigation located 1000 m downstream on the same side as the discharge point. The irrigation period lasts 120 days over 6 months, with a daily irrigation rate of 9 L/m^2 on peat soil. The assessment uses a unit discharge of 1 curie (3.7×10^{10} Bq) per year.

2.2.2.1 Radionuclide Concentration in River Water

Based on IAEA Report No. 19, for $\bar{B}$ = 50 m, the mean annual river flow rate ($\bar{q}r$) is 30 m^3/s. The 30-year minimum flow rate is qr = $\bar{q}r$/3 = 10 m^3/s. From Table 3 of the report, when qr = 10 m^3/s, the river width B = 28.8 m and depth D = 0.48 m, giving a water velocity of 0.72 m/s. The distance for complete vertical mixing is Lz = 7D. Since the water use location at x = 1000 m exceeds this distance, incomplete lateral mixing must be considered using a partial mixing coefficient Pr = 2.7 (from Table 4 of Report No. 19).

The total activity concentration in river water (Cw,tot) is:

Cw,tot = (Qi/qr) × exp(-λi × x/u)

where λi are the decay constants for 99mTc, 131I, 90Y, and 223Ra (3.20×10^{-5} s^{-1}, 9.98×10^{-7} s^{-1}, 3.00×10^{-6} s^{-1}, and 7.02×10^{-7} s^{-1}, respectively), x is the distance (1000 m), u is the water velocity (0.72 m/s), and Pr is the partial mixing coefficient (2.7).

2.2.2.2 Radionuclide Concentration in Shoreline Sediment

When dissolved radionuclides interact with sediments, the dissolved phase concentration decreases due to sorption onto sediment particles, increasing radionuclide activity in suspended and bottom sediments.

The dissolved radionuclide concentration in water (Cw,dis) is:

Cw,dis = Cw,tot / (1 + 0.001 × Kd × Cs)

where Kd is the distribution coefficient between dissolved and sorbed phases (L/kg) with values of 5, 10, 40, and 500 for 99mTc, 131I, 90Y, and 223Ra, respectively, and Cs is the suspended sediment concentration (5$×10^{-2}$ kg/m^3, the default value).

The concentration in suspended sediment (Cs,wat) is:

Cs,wat = 0.001 × Kd × Cw,dis

The concentration in bottom sediment (Cs,bot) is:

Cs,bot = 0.001 × Kd × Cw,dis × (1 - exp(-λi × Te))

where Te is the effective accumulation time (3.15$×10^{7}$ s, or 1 year).

The surface activity concentration in shoreline sediment (Cs,sho) is:

Cs,sho = 0.1 × 0.001 × Kd × Cw,tot × (1 - exp(-λi × Te))

2.2.2.3 Radionuclide Concentration in Freshwater Fish

The transfer of discharged radionuclides to aquatic food is calculated as:

Cf = Cw,dis × Bp / 1000

where Cf is the radionuclide concentration in freshwater fish (Bq/kg) and Bp is the bioconcentration factor ($\mathrm{Bq \cdot kg^{-1}/Bq \cdot L^{-1}}$) with values of 20, 40, 30, and 50 for 99mTc, 131I, 90Y, and 223Ra, respectively.

2.2.2.4 Radionuclide Concentration in Irrigated Crops

Crops irrigated with river water receive radionuclides from two sources: deposition on crop surfaces and uptake from soil.

The concentration from soil uptake (Cp,soil) is:

Cp,soil = Fv × Cs,bot × (1 - exp(-λEsoil × tb))

where Fv is the concentration factor for crop edible parts (Bq/kg) with values of 5, 0.02, 0.003, and 0.04 for 99mTc, 131I, 90Y, and 223Ra, respectively; λEsoil is the effective decay coefficient for radionuclide activity concentration in soil ($\mathrm{d^{-1}}$), calculated as λi + λs, where λs represents removal processes other than radioactive decay (0.0014 $\mathrm{d^{-1}}$ for 99mTc and 131I, 0 for 90Y and 223Ra);

and tb is the exposure duration (1.10$×10^{4}$ days for 30 years of hospital operation).

The concentration from surface deposition (Cp,sur) is:

Cp,sur = (α × Ddep × (1 - exp(-λEp × te))) / (Yp × λEp)

where Ddep is the deposition rate ($\mathrm{Bq \cdot m^{-2} d^{-1}}$), calculated as Cw,dis × irrigation rate; α is the mass interception coefficient (0.3 $\mathrm{m^2/kg}$); Yp is the crop yield ($\mathrm{kg/m^2}$); λEp is the effective decay coefficient for surface contamination ($\mathrm{d^{-1}}$), calculated as λi + λw, where λw is the removal rate constant for surface deposits (0.05 $\mathrm{d^{-1}}$); and te is the exposure duration during the growing season (120 days).

The total radionuclide concentration in crops (Cp) is:

Cp = Cp,sur + Cp,soil

2.3 Dose Assessment

2.3.1 Sludge Discharge Model 2.3.1.1 Dose to Sewage Treatment Plant Workers

Treatment plant workers are considered to receive both external exposure from sludge ground deposition and internal exposure from inhalation of resuspended sludge. The assessment accounts for 12 hours of radionuclide decay from medical facility discharge to arrival at the treatment plant.

The external dose from contaminated sludge ground deposition (Eex,slu) is:

Eex,slu = Cslu,sur × DFgr × Qf

where DFgr is the effective dose conversion factor for ground deposition ($\mathrm{Sv \cdot s^{-1}/Bq \cdot m^{-2}}$) with values of 7.06$×10^{\{-17\}, 2.44×10}${-16}, 1.46×10$^{\{-16\}, and 7.86×10}${-17}$ for 99mTc, 131I, 90Y, and 223Ra, respectively, and Qf is the occupancy factor (1.8$×10^{6}$ s/year, assuming 2 hours/day for 250 days).

The internal dose from inhalation of resuspended sludge (Ein,slu) is:

Ein,slu = Cslu,sur × Rinh × DFinh × Qf × DL

where Rinh is the annual inhalation rate (8400 $\mathrm{m^3/a}$), DFinh is the effective dose conversion factor for inhalation (Sv/Bq) with values of 1.20$×10^{\{-11\}, 7.40×10}${-9}, 1.70×10$^{\{-9\}, and 6.90×10}${-6}$ for 99mTc, 131I, 90Y, and 223Ra, respectively, Qf is the occupancy factor (0.228 for 2000 working hours/year), and DL is the dust loading from resuspension (0.1 $\mathrm{mg/m^3}$).

2.3.1.2 Dose to Waste Incineration Plant Workers

Incineration plant workers are considered to receive both internal exposure from inhalation of radionuclides released during combustion and external exposure from immersion in contaminated air. The assessment accounts for 24 hours of radionuclide decay from medical facility transfer to the incineration plant.

The internal dose from inhalation (Ein,air) is:

Ein,air = Ccin × Rinh × DFinh × Qf

The external dose from immersion (Eex,air) is:

Eex,air = Ccin × DFim × Qf

where DFim is the immersion dose conversion factor ($\mathrm{Sv} \cdot \mathrm{m}^3 \cdot \mathrm{Bq}^{-1} \cdot \mathrm{s}^{-1}$) with values of 5.14\$$\times 10^{\{\text{-}15\},\, 1.69\times 10}\{\text{-}14\}, 3.18\times 10^{\{\text{-}15\},\, and 5.40\times 10}\{\text{-}15\}$\$ for 99mTc, 131I, 90Y, and 223Ra, respectively.

2.3.2 River Water Discharge Model For radionuclides remaining entirely in water and discharged to rivers, we consider three exposure pathways to the same individual: external exposure from riverbed sediment, internal exposure from fish consumption, and internal exposure from crop consumption.

The external dose from riverbed sediment (Eex,sed) is:

Eex,sed = Cs,sho × DFgr

The internal dose from fish consumption (Ein,f) is:

Ein,f = Cf × Hf × DFing

where Hf is the annual fish consumption (35 kg/a) and DFing is the ingestion dose conversion factor (Sv/Bq) with values of 2.20\$$\times 10^{\{\text{-}11\},\, 2.20\times 10}\{\text{-}8\}, 2.70\times 10^{\{\text{-}9\},\, and 1.00\times 10}\{\text{-}7\}$\$ for 99mTc, 131I, 90Y, and 223Ra, respectively.

The internal dose from crop consumption (Ein,p) is:

Ein,p = Cp × Hp × DFing

where Hp is the annual crop consumption (510 kg/a).

3. Results and Discussion

3.1 Clearance Levels for Sewage Treatment Plant Scenario (Sludge Discharge)

Based on the sludge discharge model in Section 2.2.1, sewage treatment plant workers receive two types of exposure: external dose from sludge ground deposition (Eex,slu) and internal dose from inhalation of resuspended sludge (Ein,slu). Assuming workers receive the sum of both pathways (Etot,slu), clearance levels (Q1) were derived using the 10 Sv/a dose criterion, as shown in Table 1.

3.2 Clearance Levels for Incineration Plant Scenario (Sludge Discharge)

Based on the sludge discharge model, sludge from treatment plants is transferred to incineration facilities, where workers receive internal exposure from inhalation of contaminated air (Ein,air) and external exposure from immersion (Eex,air).

Assuming workers receive the sum of both pathways (Etot,air), clearance levels (Q1’) were derived using the 10 Sv/a criterion, as shown in Table 2.

3.3 Clearance Levels for River Water Discharge Scenario

Based on the river water discharge model in Section 2.2.2, the same individual receives three types of exposure: external dose from shoreline sediment (Eex,sed), internal dose from freshwater fish consumption (Ein,f), and internal dose from crop consumption (Ein,p). The total dose (Etot,riv) is the sum of these pathways. Clearance levels (Q2) were derived using the 10 Sv/a criterion, as shown in Table 3.

3.4 Final Clearance Levels and Comparison

Following the methodology in Section 2, clearance levels were derived for three scenarios: Q1 (sewage treatment plant), Q1’ (incineration plant), and Q2 (river water). The minimum value among these was selected as the regulatory clearance level. The derived clearance levels for 99mTc, 131I, 90Y, and 223Ra are 3.14$×10^{8}$ Bq/a, 2.75$×10^{7}$ Bq/a, 4.30$×10^{7}$ Bq/a, and 1.45$×10^{7}$ Bq/a, respectively. For practical application, these may be rounded to 1.0$×10^{8}$ Bq/a, 1.0$×10^{7}$ Bq/a, 1.0$×10^{7}$ Bq/a, and 1.0$×10^{7}$ Bq/a. For facilities using multiple radionuclides, the sum of the ratios of each radionuclide’ s discharge activity to its clearance level must be less than 1 for clearance discharge to be permissible.

The derivation process shows that clearance levels for 99mTc and 90Y are primarily determined by external exposure from sludge ground deposition, while those for 131I and 223Ra are mainly determined by internal exposure from inhalation at incineration plants. The IAEA technical report “Clearance of Materials Resulting from the Use of Radionuclides in Medicine, Industry and Research” does not derive clearance levels for 223Ra. For 99mTc, 131I, and 90Y, the IAEA-derived clearance levels are 1.0$×10^{9}$ Bq/a, 1.0$×10^{7}$ Bq/a, and 1.0$×10^{10}$ Bq/a, respectively, determined by sludge deposition external exposure (for 99mTc and 131I) and fish ingestion plus direct water consumption (for 90Y). Reference [22] did not consider ground deposition external exposure for 90Y. In our study, the external dose conversion factor for 90Y ground deposition is 4.60$×10^{-9}$ Sv·m^2/Bq·a, taken from the U.S. EPA’ s 2019 report on external exposure to radionuclides. This difference in exposure pathways considered leads to the substantial difference in derived clearance levels for 90Y.

The Environmental Permitting (England and Wales) Regulations specify discharge limits to municipal sewers of 3.7$×10^{7}$ Bq/a for 99mTc, 1.0$×10^{6}$ Bq/a for 131I, 1.0$×10^{7}$ Bq/a for 90Y, and 1.0$×10^{5}$ Bq/a for 223Ra. Except for 223Ra, our derived clearance levels are generally consistent with UK regulations. The higher value for 223Ra in our study results from using a 1‰ release fraction during incineration based on relevant literature.

This study provides a systematic approach and practical guidance for deriving clearance levels for medical radioisotope wastewater in China. By considering the most conservative discharge scenarios and exposure pathways, it offers a theoretical foundation and practical demonstration for establishing clearance levels. Future work should incorporate advances in monitoring technology and practical applications to further refine relevant standards and regulations, thereby supporting the safe development and application of medical radionuclides in China.

Author Contributions

Jiang Wenhua: Manuscript writing and theoretical model calculations
Peng Hui and Dang Lei: Investigation of actual scenarios for model estimation
Xu Ying: Figure and table preparation
Wang Yinning: Data analysis and results compilation

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