Producing μBq/m^3 level low ^{226}Ra ultrapure water for the Jiangmen Underground Neutrino Observatory

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Abstract

The Jiangmen Underground Neutrino Observatory (JUNO), a 20~ktons low-background Liquid Scintillator (LS) detector, was primarily designed to determine the neutrino mass ordering. JUNO requires UltraPure Water (UPW) with a ^{226}Ra concentration $\$<50\ \ Bq/m\hat{}3\$$ due to the direct liquid-liquid contact between LS and UPW during detector filling. To meet this stringent requirement, a highly sensitive measurement system capable of detecting 3.9~μBq/m^3 of ^{226}Ra was developed, and the 100~t/h UPW production process was optimized. By integrating selective ion-exchange resin with membrane separation technologies, UPW with a ^{226}Ra concentration of $\$<4\ \ Bq/m\hat{}3\$$ was consistently produced, exceeding JUNO' s specifications and setting a world-leading benchmark. This paper describes the design and implementation of JUNO' s UPW system and the highly sensitive ^{226}Ra measurement system, along with a systematic evaluation of ^{226}Ra removal efficiency across purification stages and final water quality validation.

Full Text

Preamble

Producing µBq/m3 level low 226 Ra ultrapure water for the Jiangmen Underground Neutrino Observatory∗ Xi-Ting Xu,1 Cong Guo,2, 3, 4, † Jin-Chang Liu,2, 3, 4 Yong-Peng Zhang,2, 3, 4 Xiao-Hua Liang,5 Xin-Jian Wen,1, ‡ Chang-Gen Yang,2, 3, 4 Quan Tang,6, § Li-Dan Lv,6 Yuan-Hao Niu,1 and Bin Xiao7

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The Jiangmen Underground Neutrino Observatory (JUNO), a 20 ktons low-background Liquid Scintillator (LS) detector, was primarily designed to determine the neutrino mass ordering. JUNO requires UltraPure Water (UPW) with a 226 Ra concentration <50 µBq/m3 due to the direct liquid-liquid contact between LS and UPW during detector filling. To meet this stringent requirement, a highly sensitive measurement system capable of detecting 3.9 µBq/m3 of 226 Ra was developed, and the 100 t/h UPW production process was optimized. By integrating selective ion-exchange resin with membrane separation technologies, UPW with a 226 Ra concentration of <4 µBq/m3 was consistently produced, exceeding JUNO' s specifications and setting a world-leading benchmark. This paper describes the design and implementation of JUNO' s UPW system and the highly sensitive Ra measurement system, along with a systematic evaluation of 226 Ra removal efficiency across purification stages and final water quality validation.

Keywords

JUNO, Ultra Pure Water, 226 Ra, Revise Osmosis, Resin

INTRODUCTION

Cherenkov Detector (WCD), and a Top Tracker (TT) detector. The CD contains 20 ktons of ultrapure LS housed in a 25 spherical acrylic vessel and is submerged in the WCD, which 26 holds 40 ktons of Ultra Pure Water (UPW) and is instru27 mented with 2400 20-inch MicroChannel Plate Photomulti28 plier Tubes (MCP-PMTs). The WCD provides sufficient wa29 ter shielding to protect the CD from ambient radioactivity 30 and simultaneously serves as a cosmic muon veto. For back31 ground and safety reasons, the CD and WCD must be initially 32 filled simultaneously with UPW. Subsequently, the UPW in 33 the CD is replaced with LS at a production rate of 7 m /h. 34 During this replacement process, UPW and LS come into di35 rect contact. As this stage marks the final step before detector 36 operation, strict control of radionuclides in the water is crit37 ical to prevent contamination of the LS by the UPW. One of 38 the most challenging requirements is to maintain the 39 concentration in the water to below 50 µBq/m [2].

The Jiangmen Underground Neutrino Observatory (JUNO) [1–4] is a multipurpose experiment designed primar4 ily to determine the neutrino mass ordering and to precisely 5 measure neutrino oscillation parameters by detecting reactor

6 antineutrinos.

With 20 ktons of low-background Liquid 7 Scintillator (LS) and an unprecedented energy resolution of 8 3%@1MeV, JUNO is the largest LS-based underground neu9 trino observatory, capable of addressing numerous important 10 topics in astroparticle physics.

JUNO' s extensive physics 11 program encompasses supernova neutrinos, atmospheric 12 neutrinos, solar neutrinos, geoneutrinos, and searches for 13 new physics [1, 2]. Located in an underground laboratory 14 with 700 m overburden, the muon flux at the JUNO site 15 is about 4 mHz/m with a mean energy of 207 GeV [5]. 16 The primary detection channel for reactor antineutrinos in 17 JUNO is the inverse beta decay reaction on protons. JUNO 18 is expected to detect only 60 reactor antineutrino events 19 per day, making stringent control of radioactive backgrounds 20 essential [6].

In the field of low-background water purification and 226 Ra detection, the Sudbury Neutrino Observatory (SNO) [7] and 42 Super-Kamiokande (Super-K) [8] experiments have achieved 43 internationally recognized advances. SNO developed a mea44 surement technique using manganese oxide-coated beads, 45 achieving a Ra detection sensitivity of 10 µBq/m3 . The 46 initial Ra concentration in SNO' s UPW was 50 µBq/m3 , 47 which was later reduced to 10-20 µBq/m through recircu∗ This work is supported by the Strategic Priority Research Program of the 48 lation and purification [9]. Super-K adopted SNO' s methodChinese Academy of Sciences (Grant No. XDA10011200) and the Youth 49 ology and attained a similar sensitivity of 10 µBq/m , alInnovation Promotion Association of the Chinese Academy of Sciences 50 though its UPW exhibited slightly higher Ra levels, with (Grant No. 2023015). † Corresponding author, C. Guo, E-mail address: guocong@ihep.ac.cn 51 initial concentration of 61 ± 13 µBq/m , decreasing to 32 ‡ Corresponding author, X.J. Wen, E-mail address: wenxj@sxu.edu.cn 52 ± 7 µBq/m after recirculation [10]. In terms of production § Corresponding author, Q. Tang, E-mail address:tanquan528@sina.com 53 scale, Super-K' s water system has a capacity of 30 t/h [10],

substantially larger than SNO' s 9 t/h system [7].

UPW SYSTEM

The JUNO experiment employs a 100 t/h UPW system to provide 40 ktons of UPW for the WCD. This system produces In this work, we report a world-leading ultrapure water pu83 water that meets the following specifications: a resistivity 56 rification system that reduces the Ra concentration from 84 >18.2 MΩ*cm, a Rn concentration of <1 × 104 µBq/m3 , 57 4 × 10 µBq/m in the source water to <4 µBq/m , sur∘ 85 and a minimum temperature of 17 C, thereby ensuring high 58 passing both the JUNO requirement and the performance of 86 muon detection efficiency for the WCD, negligible radioac59 comparable experiments. To achieve such ultra-low detec87 tive contribution to the CD, and thermal stability for the 60 tion, we developed a high-sensitivity measurement system 88 CD [1]. In addition, for the initial filling of the acrylic

vessel, 61 based on Mn-fiber adsorption and radon emanation, reaching 89 this system must deliver UPW at 50 t/h with a Ra concen3 62 a sensitivity of 3.9 µBq/m through optimized Mn-fiber syn3 90 tration below 50 µBq/m [2]. 63 thesis. The system operates reliably at 100 t/h, demonstrat91 The JUNO 100 t/h UPW system utilizes tap water as feed64 ing its feasibility for large-scale engineering. Furthermore, 92 water to produce UPW that is compliant with China' s national 65 through systematic optimization and pollution source tracing, 93 electronic grade EW-I standard [11].

As shown in Fig. 2 [Figure 2: see original paper], 66 we identified the softening resin and reverse osmosis mem94 the system comprises aboveground and underground sections. 67 brane as the key removal components (both achieving >99% 95 The aboveground treatment system consists of the following 68 radium removal efficiency), and pinpointed the primary con96 sequential units: 69 tamination sources as sediment in the regeneration sea salt - Bag filters (with a pore size of 5 µm). 70 and air ingress via the tank breather valve, enabling targeted - Multi-media filters (with stratified quartz sand, activated 71 engineering improvements for long term stability. 99 carbon, and anthracite). - Activated carbon filters (using coconut shell-derived car101 bon).

Ra removal This paper is structured as follows. Sec. II describes the 102 - Softening resin columns (with special 3 103 resin from Jiangsu Suqing Water Treatment Engineering 73 UPW system of the WCD. Sec. III details the 3.9 µBq/m 74 highly sensitive Ra in water measurement system. The 104 Group Co., Ltd.). 75 measurement results of the Ra concentrations after each 105 - Cartridge filters (with a pore size of 5 µm). - Reverse Osmosis (RO) membranes. 76 purification stage and in the final product water are presented 77 in Sec. IV. Potential upgrades to the system for enhanced 107 The pretreatment process begins with mechanical filtration Ra removal are discussed in Sec. V, and a summary of 108 using bag filters for bulk particulate removal, followed by 79 the work is provided in Sec. VI. 109 multi-media filtration for the reduction of suspended solids.

Subsequent activated carbon adsorption removes residual 145 diation while oxidatively decomposing organics. Degassing chlorine, organic compounds, ammonia-nitrogen species, ni- 146 membranes, which employ hollow fibers, vacuum pumps, and 112 trites, trace heavy metals, and microbial contaminants. The 147 purging nitrogen, remove dissolved gases via partial pressure 113 ion-exchange softening process achieves Ca /Mg2+ re- 148 differential and are critical for the removal of 222 Rn from wa222 114 moval through cationic substitution. After intermediate stor- 149 ter [14–16]. The microbubble generator enhances the 115 age, water passes through cartridge filters (5 µm) to pro- 150 removal efficiency of the degassing membranes by reloading 116 tect downstream equipment. The RO array provides primary 151 nitrogen into the water [14, 16]. Heat exchangers regulate wa117 desalination via semipermeable membranes, achieving near- 152 ter temperature. UF membranes (with a pore size of <10 nm) 153 remove trace amounts of ions, particles, colloids, and

bacteria 118 complete ionic rejection [12].

The multi-media filters, activated carbon units, and soften- 154 from water. 120 ing resins are regenerable. Multi-media filters and activated The 100 t/h UPW system supplies water for both the WCD 121 carbon filters undergo hydraulic backwashing to restore ca156 and the initial filling of the CD. Owing to their distinct re122 pacity, while softening resins are regenerated with NaCl brine 157 quirements, the two streams undergo separate treatments. The 123 to displace accumulated ions and restore exchange capacity. 158 CD-bound water, which contacts the LS directly, receives 124 This approach ensures sustainable long-term operation. 159 additional UF for enhanced removal of ionic contaminants The product water from the aboveground system is trans238 232 160 (U/Th/226 Ra). The WCD-bound water, by contrast, is 126 ferred underground through a 1,300 m stainless-steel pipeline. 161 routed through a dedicated heat exchanger to remove heat 127 With an elevation differential of 450 m, the water first 162 from the submerged PMT electronics, thereby ensuring sta128 passes through a pressure-reducing valve before entering the 163 ble detector temperatures. 129 storage tank. The underground water system comprises the 130 following components: - Secondary RO membranes (for enhanced desalination). - ElectroDeIonization (EDI) device. - Cartridge filters (with a pore size of 0.1 µm). - Total Organic Carbon (TOC) removal. - Ultraviolet (UV) sterilization. 226 RA CONCENTRATION IN WATER - Degassing membranes.

MEASUREMENT - Microbubble generator. - Heat exchangers. - Ultra-Filtration (UF).

The required 226 Ra concentration of 50 µBq/m3 (2 × The secondary RO membranes provide additional ion re- 167 10−21 g/g) is beyond the detection capability of conventional 141 moval. The EDI unit combines ion-selective membranes and 168 methods like high-purity germanium spectrometry [17] and 142 resin under a DC field for continuous deionization without 169 inductively coupled plasma mass spectrometry [18]. There143 chemical regeneration [13]. Cartridge filters capture residual 170 fore, a custom system was developed that determines the Ra activity by measuring its gaseous daughter, 222 Rn. 144 particles. The TOC/UV systems sterilize through UV irra- 171

of the key adsorbent, the Mn-fiber, from 17 µBq/g to 10.7 $\pm$1.5 µBq/g. This was achieved through three major enhance226 202 ments to the standard synthesis protocol [21, 22]:

The initial step in measuring the Ra concentration 174 in water via Rn emanation is 226 Ra preconcentration. 203 (1) Nitric acid pre-cleaning of the polyurethane fibers. 175 This method leverages the strong adsorption of Ra onto 204 (2) Moving the boiling and rinsing processes to a clean176 MnO2 [9, 19, 20] by using laboratory-synthesized Manganese 205 room. 177 fiber (Mn-fiber) [21, 22]. (3) Substituting distilled water with high-purity water from 207 China Resources C' estbon Beverage (China) Co., Ltd for 208 rinsing—the most impactful change.

Measurement principle

2 KMnO4 +2 H2 SO4 −−→ Mn2 O7 +2 KHSO4 +H2 O (3)

2 Mn2 O7 −−→ 4 MnO2 + 3 O2 ↑

parentheses are the half-lives. The energies shown for α decay are peak energies, while those for β decay are the maximum energies.

Following 226 Ra extraction, the activity of 222 Rn emanated 179 from the Mn-fiber during a sealed period is measured. Fig. 3 [Figure 3: see original paper] 180 illustrates the relevant decay branch of Ra. During sealing, 181 the relationship between the activity of Ra and its 222 Rn 182 progeny activity can be calculated using Eq. 1 [21]:

ARn (t) = ARa ×

× (e−λRa t − e−λRn t) λRn − λRa

where ARn (t) is the 222 Rn activity at time t in mBq, ARa is 185 the initial Ra activity extracted by Mn-fiber in mBq, λRn 186 is the decay constant of Rn, λRa is the decay constant of 212 The Mn-fiber itself is produced by creating a firm MnO2 213 coating on the polyurethane fibers via the reaction of KMnO4 Ra, and t is the Mn-fiber sealing time in days.

Given that the half-life of 226 Ra is significantly longer than 214 with H2 SO4 under heated conditions (Eq. 3, Eq. 4). A pic222 189 that of Rn (i.e., λRa λRn), Eq. 1 can be simplified to 215 ture of the Mn-fiber is shown in Fig. 4 [Figure 4: see original paper]. Gravimetric analysis 216 indicates a 13% mass increase in polyurethane fibers after

190 Eq. 2:

217 being converted into Mn-fiber. ARn (t) ARa = 1 − e−λRn t

Extraction column

Measurement procedures and devices

The 226 Ra extraction column, illustrated in Fig. 5 [Figure 5: see original paper], is conThe 226 Ra concentration measurement comprises four 220 structed from an acrylic tube (20 mm inner diameter and 194 steps: (1) Mn-fiber synthesis, (2) Ra extraction from water, 221 185 mm long) sealed with rubber gaskets and 10 mm quick222 195 (3) Mn-fiber encapsulation, and (4) Rn activity determina- 222 connects. The assembled unit withstands water pressures up 196 tion. 223 to 5 kg/cm . Approximately 5 g of Mn-fiber is packed in the 224 central section, flanked by polyurethane fibers at both ends to 225 filter particulate matter. During operation, water is directed

1. Mn-fiber

226 upward through the column to maximize contact with the Mn227 fiber. A residential water meter (GB/T 778 compliant [23], To meet JUNO's requirement for μBq/m3 -level 226 Ra mea- 228 $±$2% accuracy) at the outlet measures the total volume of wa226 199 surements, we significantly reduced the Ra background 229 ter passed through the Mn-fiber.

(2) Mn-fiber container. Two chamber types are used (Fig. 7 [Figure 7: see original paper]): a standard CF35 tube (150 mm long) for 5 g 263 samples, and a custom chamber (130 mm diameter and 264 300 mm length) for <300 g samples for background mea265 surement. All inner surfaces were electropolished to a rough266 ness of <0.1 μm. After loading, chambers are purged with 267 boil-off nitrogen to remove residual air and then sealed for 268 several days to allow Rn ingrowth. The activity ratio be226 269 tween Ra and Rn can be calculated using Eq. 2. For in222 270 stance, after a 10-day sealing interval, Rn activity reaches 271 83.7% of the initial Ra activity. (3) Dehumification systems. Mn-fibers that have extracted Ra retain substantial water, releasing significant water va274 por during radon transfer that degrades detector efficiency 275 through humidity interference. This system comprises a liq276 uid nitrogen tank, a solenoid valve, a dewar, a PT100 tem277 perature sensor, and a temperature controller. Through the 278 thermostatic regulation of intermittent liquid nitrogen injec279 tion into the dewar, the system maintains a temperature of 280 -60 C inside the dewar, reducing the relative humidity to 3% 281 at a 1 L/min gas flow rate without introducing background. (4) MFC. The MFC is used to regulate the gas flow rate 283 during radon transfer operations, with an adjustment range of 284 0.5–5 L/min. 222 Rn emanation measurement system (5) Vacuum pump. Before measurements, the pump evac286 uates the radon detector and associated pipelines.

Subse231 The 226 Ra activity extracted by Mn-fibers was quantified 287 quently, boil-off nitrogen is used to purge the emanated 232 by measuring its gaseous daughter Rn using a radon ema288 from the Mn-fiber container into the radon detector. 233 nation measurement system (Fig. 6 [Figure 6: see original paper]). The system comprises a (6) Pipelines and valves. All stainless steel gas pipelines 234 radon detector, a Mn-fiber container, a dehumidification sys290 and interconnecting valves are EP-grade components. 235 tem, a mass flow controller (MFC, Model 1179A, MKS), and

a vacuum pump (Model ACP40, Pfeiffer Vacuum). All connections use metal-sealed ConFlat flanges and VCR compo−9 238 nents, ensuring a leak tightness $<1 \times 10$ Pa*m3 /s. Key 291 C. Extraction efficiency calibration 239 components are summarized below; full details were de240 scribed in our previous work [22].

The efficiency of extracting 226 Ra from water with Mn241 (1) Radon detector. The radon detector operates on the 293 fiber was calibrated using a standardized 226 Ra solution with 242 principle of electrostatic collection [24–30], where

the de3 294 a concentration of (9.04 ± 0.53) × 10 µBq/m3 . Our prior 243 tection efficiency for radon progeny correlates with applied 295 work [22] shows consistent extraction efficiency across 244 collection voltage and gas humidity. At 3% relative humid4 296 concentrations (760–6.1 × 10 µBq/m) and water flow rates 245 ity and -700 V collection voltage, the detector achieves 90% 297 (1.6–8 L/min). To cover Ra concentrations from 1 to 1 246 collection efficiency for Rn progeny 214 Po. This corre- 298 × 104 µBq/m3 in JUNO, sample volumes are varied, which 247 sponds to a calibration factor (CF) of 67 ± 6.7 counts per 299 requires dedicated calibration of the extraction efficiency for 248 hour per Bq/m (cph/(Bq/m)). The system' s single-day mea300 different volumes. In contrast to absolute quantification, we 249 surement sensitivity is 0.7 mBq/m for Rn [24]. For each 301 employed a relative scheme with volume-matched control ex250 sample measurement, the radon detector integrates radon ac302 periments to simultaneously mitigate detector systematic un251 tivity over a 24-hour data-taking period. As the measured 303 certainties and background in this work. 252 value represents the average Rn concentration during this Tab. 1 presents the calibrated 226 Ra extraction efficiencies 253 interval, determination of the initial Rn concentration re305 across different water volumes. Throughout the test, an addi254 quires correction for radioactive decay, which can be calcu226 306 tional 150 µL of Ra solution was added per 300 L of water, 255 lated using Eq. 5 [22]: 307 and the water flow rate through the Mn-fiber was maintained 308 at 5 L/min.

ARn−M × λRn × (t2 − t1) ARn = (5) 309 For water volumes below 1 m3 passing through the Mn−λ 310 fiber extraction column, Ra extraction efficiency ap222 257 where ARn is the initial Rn activity after transfer into the 311 proached 100%. Beyond this volume, a progressive decline 258 detector in mBq, ARn−M is the measured Rn activity in 312 in 226 Ra adsorption efficiency was observed, which might be Ra being shed from 259 mBq, t1 is the measurement start time in hours, and t2 is the 313 caused by a small amount of adsorbed 314 the MnO2 coating. 260 measurement end time in hours.

226 Ra extraction efficiencies of Mn-fiber for different water volumes. The uncertainties in UPW volume are derived from the accuracy of the water flow meter. The statistical uncertainties in efficiency are based on Poisson statistics, while the systematic uncertainties are determined by the uncertainty in the activity of the radium solution. For samples No. 1–No. 4, the radon detector datataking time was 24 h, whereas for samples No. 5 and No. 6, it was 15 h.

No. UPW volume (m3)

Ra extraction efficiency (%)

0.3 ± 0.006

99.0 ± 13.8stat ± 5.8sys

0.5 ± 0.01

98.5 ± 10.6stat ± 5.8sys

1.0 ± 0.02

99.5 ± 7.6stat ± 5.8sys

5.0 ± 0.1

94.6 ± 3.3stat ± 5.5sys

10.0 ± 0.2

91.3 ± 2.9stat ± 5.4sys

20.0 ± 0.4

89.2 ± 2.0stat ± 5.2sys

Sensitivity estimation

background event rate The sensitivity of measuring 226 Ra concentration in wa- 326 lation assumes a normally distributed222 327 and radioactive equilibrium between Rn and 226 Ra. 317 ter at a 95% confidence level can be estimated according to The system background was determined by measuring 5 g

318 Eq. 6 [29]:

329 of unused Mn-fiber following the standard sealing and anal330 ysis procedure. Ten replicate measurements yielded a mean $1.645 \times \sigma BG \times Vs$ (6) 331 background of 2.7 CPD (Fig. 8 [Figure 8: see original paper]), reflecting contributions from $24 \times CF \times Vw \times \epsilon \times 10$ 332 both the apparatus and the Mn-fiber.

Using the parameters described above and calculated with 320 where L is the sensitivity in µBq/m , σBG is the statistical 333 321 uncertainty of the system' s background event rate in Counts 334 Eq. 6, a sensitivity of 3.9 µBq/m for Ra concentration in of wa322 Per Day (CPD), Vs = 0.042 m is the total volume of the mea- 335 water was derived. This calculation assumes 20 m 323 surement system, CF = 67 ± 6.7 cph/(Bq/m) is the detector 336 ter passes through 5 g Mn-fiber with a Ra extraction effi3 Rn activity 324 calibration factor, Vw is the sample water volume in m , and ϵ 337 ciency of 89.2%, and the data-taking time for 325 is the Ra extraction efficiency listed in Tab. 1. This calcu- 338 measurement is 24 h.

counter), and resistivity (using an AZ8302 portable meter and a +GF+ Signet 9900 in-line meter) were conducted across the 100 t/h UPW system. Uncertainties are reported as follows: for 226 Ra concentration, the statistical uncertainties are based on Poisson statistics, while the systematic uncertainties include contributions from the calibration factor, water volume, and 226 Ra extraction efficiency; for particulate matter count, uncertainties are statistical only; and for resistivity, uncertainties correspond to the instrument display accuracy. Missing resistivity values are due to meter limitations at high resistivity ranges. The large relative uncertainty at S17 for the particulate matter arises

from counting statistics: each measurement used a 20 mL sample, yielding an average of 2 counts per 20 mL, which converts to 100 ± 70 counts/L based on Poisson uncertainty (2 × 50). For sampling locations, refer to Fig. 2.

Ra concentration

Particulate matter > 0.5 μm

Resistivity

(Counts/L)

(MΩ*cm)

(429 ± 4stat ± 68sys) × 10

601150 ± 5482

0.019 ± 0.001

(186 ± 3stat ± 29sys) × 10

298750 ± 3864

0.019 ± 0.001

(194 ± 3stat ± 31sys) × 10

341850 ± 4134

0.019 ± 0.001

(147 ± 2stat ± 23sys) × 10

17650 ± 939

0.019 ± 0.001

(426 ± 4stat ± 67sys) × 10

22500 ± 1060

0.018 ± 0.001

504 ± 114stat ± 71sys

46600 ± 1526

0.018 ± 0.001

(76 ± 1stat ± 11sys) × 10

341850 ± 4134

0.018 ± 0.001

(55 ± 1stat ± 8sys) × 10

244900 ± 3499

0.052 ± 0.001

79 ± 24stat ± 9sys

2150 ± 328

0.190 ± 0.002

(15.8 ± 0.6stat ± 2.2sys) × 10

10050 ± 709

0.470 ± 0.005

84 ± 25stat ± 17sys

2750 ± 370

96 ± 26stat ± 12sys

2250 ± 335

56 ± 21stat ± 7sys

2150 ± 327

17.74 ± 0.01

49 ± 22stat ± 6sys

900 ± 200

6.3 ± 4.5stat ± 0.7sys

850 ± 206

18.19 ± 0.01

19.4 ± 5.8stat ± 2.2sys

350 ± 132

100 ± 70

18.20 ± 0.01

Counts

(µBq/m)

Location

RESULTS

Entries Std Dev

Ra concentrations across the system

Using the custom-developed device described above, the Ra concentrations in water at different locations of the 343 JUNO 100 t/h UPW system, marked as S1 –S17 in Fig. 2, 344 were measured, and the results are shown in Tab. 2. The water 345 volume used for S1–S5 was 0.5 m , for S6–S8 and S10 was 346 1 m , for S9 and S11–S14 was 5 m , and for S15–S17 was 347 20 m . All measurements were performed after a 10-day Mn0 348 fiber sealing period and a 24-hour Rn counting interval.

Background/CPD 349 Reported uncertainties include both statistical and systematic 350 contributions. During the 45-day filling campaign, a total of ground measurement, the Mn-fiber was sealed for 10 days. The un- 352 measurement results were below the detector' s average backcertainties on the Y-axis are statistical only. 353 ground; therefore, only an upper limit is provided.

In low-salinity solutions, 226 Ra exists predominantly as un2+ 355 complexed Ra [31–33], making it removable via deioniza226 356 tion processes. According to Tab. 2, the system reduces 357 concentration from 4×10 µBq/m to <4 µBq/m , repre341

V. DISCUSSION senting a reduction of over five orders of magnitude. This 412 >99.99% removal efficiency was achieved primarily through 360 the synergistic action of the softening resin and the two-stage The system reduces 226 Ra concentration in water from 4 361 RO membranes, with each exhibiting >99% Ra removal 414 × 105 µBq/m3 to <4 µBq/m3 , corresponding to a total re226 362 efficiency. The Ra removal efficiencies of the main devices 415 duction exceeding five orders of magnitude and an overall 363 used in the UPW system are shown in Tab. 3. The slight vari416 removal efficiency of >99.999%. Even though the RO pro226 364 ation in Ra removal efficiency between the two RO stages 417 vides high removal efficiency for particulate matter and ions, 365 is primarily attributable to differences in influent Ra con- 418 a quantitative breakdown of the contributions from individual 366 centration. This system employs two polishing resins in se419 components (Tab. 3) reveals that the softening resin plays a 367 ries, with tabulated efficiency calculations assuming identical 420 critical and complementary role, accounting for a 99.88 ± Ra removal performance. 421 0.04% reduction alone.

Our resin screening revealed sig369 However, according to the data shown in Tab. 2, mechan- 422 nificant variation in 226 Ra removal efficiency, with cationic 370 ical filtration units (e.g., bag filters, cartridges, multi-media 423 resins demonstrating superior performance over mixed-bed 371 filters) also exhibited incidental Ra removal via adsorp- 424 (anionic/cationic) alternatives. The observed difference is at2+ 372 tion of Ra onto particulates, enabling concurrent removal 425 tributed to the dominant speciation of 226 Ra as Ra2+ in low373 during filtration. Among all devices, only RO integrates both 426 salinity water [31–33], which favors exchange with cationic 374 deionization and particulate removal [34]. 427 functional groups.

This mechanistic understanding guided In addition, the data in Tab. 2 also

indicate that the 226 Ra 428 the selection of a dedicated cationic softening resin over con376 concentration in water increased after passing through each 429 ventional mixed-bed polishing resins. Although the selected Ra rejection (>99%), its pre377 tank, which is primarily attributable to air ingress through 430 softening resin achieves high 378 tank ventilation valves.

Fiberglass-reinforced plastic tanks 431 RO positioning is mandatory and not interchangeable with 379 require pressure-equalizing vent valves to prevent structural 432 post-RO polishing resin due to substantial organic leaching. 380 damage during water level fluctuations. These valves intro- 433 These leachates cause measurable deterioration of water qual381 duce ambient air (unfiltered for aboveground tanks, filtered 434 ity, primarily through resistivity reduction. Quantitative water 382 for underground tanks) containing dust-bound Ra progeny 435 quality analysis showed that placing the polishing resin down383 that dissolve upon water contact. A significant increase in 436 stream of the RO caused resistivity to drop from 18 MΩ*cm Ra concentration after the pretreatment water tank was 437 to below 1 MΩ*cm, confirming that organic leachates from 385 traced to sediment introduced during initial commissioning, 438 the resin degrade water quality if placed after the RO and jus386 when sea salt was used for softening resin regeneration. Al- 439 tifying the current upstream configuration. 387 though sea salt was replaced by clean regeneration salt and 440 In reducing the source of 226 Ra pollution, the following 388 the tank was flushed, residual sediment on the tank walls per- 441 two tasks have been identified: 389 sisted as a leaching source. (1)Thorough cleaning of the pretreatment water tank. The 443 periodic regeneration of softening resin, commonly per444 formed with economical sea salt, was found to introduce sed226 445 iment containing significant Ra contamination. During 390 B.

Relationship between 226 Ra concentration and particulate 446 the initial commissioning phase of the water system, con391 counts and resistivity 447 ventional sea salt was used, resulting in a sharp increase in Ra concentration — from 500 µBq/m3 to 3.0×10^5 Tab. 2 also lists the particulate counts and resistivity values 449 µBq/m3 —after water passed through the pretreatment tank. 393 across the water system. The particulate counts were mea- 450 Visual inspection revealed a substantial layer of sediment at 394 sured using a Bettersize C400 optical particle counter (Dan- 451 the tank bottom, which subsequent analysis confirmed origi395 dong Baxter); the small sample volume (20 mL) contributed 452 nated from the sea salt. The data presented in this paper were 396 to significant data uncertainties. Resistivity was measured us- 453 collected after switching to clean regenerant salt and perform397 ing an AZ8302 portable conductivity tester (Taiwan Hengxin) 454 ing systematic flushing of the tank. The reduction from 3.0 398 upstream of the secondary RO and a +GF+ Signet 9900 in- 455×10 µBq/m to 7.6×10 µBq/m confirms the effective399 line meter downstream. Portable measurements become unre- 456 ness of replacing the regenerant salt and cleaning the tanks, 400 liable when the resistivity exceeds 10 MΩ*cm, and the in-line 457 representing an additional 74.7% reduction in post-tank con401 measurements prevent modification

of the sampling point. 458 centration. 402 These constraints resulted in missing high-resistivity data at 459 However, even after adopting clean salt and flushing, resid403 multiple locations. 460 ual sediment remains adhered to the tank walls. This residual The observed correlations between 226 Ra concentration, 461 contamination accounts for the persistent 226 Ra increase of µBq/m3 observed after the pretreatment water 405 particulate counts, and resistivity are a concurrent outcome 462 7.6×10 406 of the multi-stage purification process. The softening resin 463 tank. To fully eliminate this source of contamination, a thor226 407 achieves >99% Ra removal by ion exchange (Na+ re- 464 ough cleaning of the tank interior is required. As this pro2+ 408 places Ra), which minimally influences resistivity and does 465 cedure entails draining the water tank and a temporary sys409 not filter particles due to its 100–500 µm bead size. The si- 466 tem shutdown, it will be scheduled for a suitable maintenance 410 multaneous decrease in all parameters is attributable to their 467 window in the future. (2)Mitigation of contamination from storage tank breather 411 collective removal through successive treatment stages.

226 Ra removal efficiencies of the devices used in the UPW system. When calculating the 226 Ra removal efficiency of the polishing resin, it is assumed that both resin stages have the same efficiency. 226 Ra removal efficiency (%)

Device

Ra concentration before the device (µBq/m3)

Soften resin

4.3×105

99.88 ± 0.03stat $\pm$ 0.02sys

99.86 ± 0.04stat $\pm$ 0.03sys 1

stage of RO

5.5×104

99.47 ± 0.16stat $\pm$ 0.13sys 2nd stage of RO

1.6×104

42 ± 27stat $\pm$ 10sys

64.1 ± 15.1stat $\pm$ 3.0sys

Polishing resin

valves. Storage tanks introduce measurable radiological con- 494 challenges given the ultra-low concentration target and the tamination primarily through their air-admittance breather 495 large water volume involved. 471 valves.

Comparative measurements at sampling points S9 496 A dedicated 100 t/h UPW production and circulation 472 (upstream of the tank) and S10 (downstream of the tank) show 497 system was developed, integrating multi-stage

purification–473 that the Ra concentration increased from (79 ± 24stat ± 498 including specialized softening resin and RO–with a novel, 474 9sys) µBq/m to (15.8 ± 0.6stat ± 2.2sys) × 103 µBq/m3 499 high-sensitivity 226 Ra measurement system (3.9 µBq/m3) 475 after passing through the tank, an increase of 200 times. This 500 based on Mn-fiber adsorption and radon emanation. System476 sharp rise confirms that air ingress via the breather valve is the 501 atic characterization across the purification stages revealed 477 dominant contamination mechanism. Two mitigation strate- 502 >99% removal efficiency for both the softening resin and the 478 gies have been evaluated: installing particulate-retentive fil- 503 two-stage RO. Quantitative analysis further identified regen479 ters on breather assemblies or using boil-off nitrogen as a 504 erant salt impurities and air ingress via tank breather valves 480 clean breathing gas. While nitrogen blanketing could thor- 505 as the two dominant contamination pathways, accounting for 481 oughly eliminate background contamination, it would require 506 the majority of post-purification recontamination.

The JUNO 100 t/h water system successfully reduced 482 a dedicated gas supply system at high cost. Furthermore, the 507 Ra concentration from 4 × 105 µBq/m3 to <4 µBq/m3 , 483 existing boil-off nitrogen capacity at the JUNO site is insuf- 508 484 ficient to meet the system's operational demand. Therefore, 509 achieving a reduction exceeding five orders of magnitude and 485 installing filtration systems at each tank air inlet has been se- 510 surpassing the JUNO requirement by more than an order of This performance establishes a world-leading 486 lected as the practical solution, which will be implemented 511 magnitude. 512 benchmark in ultra-lowRa water purification. 487 during a suitable maintenance period.

Looking forward, the identified contamination pathways 514 provide a clear basis for further optimization. Planned mea515 sures—including thorough cleaning of the pretreatment tank VI. SUMMARY 516 and installation of high-efficiency particulate filters on tank 517 breather valves—are expected to further reduce the JUNO is a multi-purpose neutrino experiment primarily de- 518 concentration in water. More broadly, the high-sensitivity 490 signed to determine the neutrino mass ordering. To ensure 519 measurement system and the key removal components de491 detector safety and minimize background, the CD must be 520 veloped in this work offer a transferable approach for future 492 filled with UPW containing <50 µBq/m of Ra before LS 521 low-background experiments requiring stringent radiopurity 493 introduction–a requirement that presents substantial technical 522 control of large-scale water systems.

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Note: Figure translations are in progress. See original paper for figures.