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Assessment of natural uranium and ^{226}Ra concentration in ground water around the uranium mine at Narwapahar, Jharkhand, India and its radiological significance

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Abstract A brief study on dissolved radionuclides in aquatic environment, especially in ground water, constitutes the key aspect for assessment and control of natural exposure. In the present study the distribution of natural uranium and ^{226}Ra concentration were measured in ground water samples collected within a 10 km radius around the Narwapahar uranium mine in the Singhbhum thrust belt of Jharkhand, India in 2007–2008. The natural uranium content in the ground water samples in this region was found to vary from 0.1 to 3.75 μg L^{-1} with an average of 0.87 ± 0.73 μg L^{-1} and ^{226}Ra concentration was found to vary from 5.2 to 38.1 mBq L^{-1} with an average of 13.73 ± 7.34 mBq L^{-1}. The mean annual ingestion dose due to intake of natural uranium and ^{226}Ra through drinking water pathway to male and female adults population was estimated to be 6.55 and 4.78 μSv y^{-1}, respectively, which constitutes merely a small fraction of the reference dose level of 100 μSv y^{-1} as recommended by WHO.

Keywords Natural uranium · ^{226}Ra concentration · Ground water · Ingestion dose

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Introduction

Man has always been exposed to ionising radiation both internally and externally of terrestrial and cosmogenic origin. The quantity of radiation received from natural sources, varies from place to place depending upon the radioactivity content in water, rock, soil, food, building material and air. Uranium is ubiquitous in nature and its level is highest in uranium ore. Because of natural mineralization and for various mining activities, elevated level of uranium and its decay products may be expected in various matrices like water, food, soil, rocks and air in the vicinity of the uranium mine. Hence, assessment of radioactivity around a uranium mine in different matrices draws a special attention in order to quantify the extent of exposure to the members of the public through various routes and to find out the contamination if any due to the mining. Exposure to natural occurring radionuclides can take place through water, food and to certain extent from inhalation of air. According to an estimate drinking water contributes 85% of ingested uranium while food contributes 15% [1]. Uranium has three isotopes present in nature with their isotopic composition, proportions by mass are 99.275% for ^{238}U, 0.72% for ^{235}U and 0.005% for ^{234}U and radioactivity ratios are 0.046 for ^{235}U/^{238}U and 0.05 for ^{235}U/^{234}U. Thus, the radioactivity of ^{235}U in water is theoretically a more negligible level than that of ^{238}U. The health effects of uranium can be divided into carcinogenic and non-carcinogenic effects [2] and these classifications are based on the radiological risk by radiation of uranium isotopes and the chemical risk as a heavy metal. For ingested uranium, the main target organ of toxicity is the kidney [3, 4]. An exposure of about 0.1 mg kg^{-1} of body weight of soluble natural uranium results in transient chemical damages to kidney has been reported [5]. Furthermore, ^{238}U series radionuclides are the major contributors to the radiation dose caused by natural radionuclides ingested with water [6] and ^{226}Ra is an important radionuclide of this series which contributes significant fraction of dose through intake of water. ^{226}Ra is an alpha emitter, having half-life 1622 years, possess only one oxidation state in natural environment i.e., Ra(II), behaves in similar respects, like other alkaline earth metals. Approximately, 20% of the ingested radium is absorbed, is taken into the blood stream and accumulates mainly in the bone [7]. Studies have shown that internal deposition of ^{226}Ra results in the induction of skeletal tumours and paranasal sinus carcinomas (cancer of the sinus cavities) [8, 9]. Due to various health impacts of uranium and radium, there have been increased concerns for radiological quality of ground water principally used for drinking purposes around a uranium mine, in order to assess the probability of radiological hazards due to its intake and to find ground water contamination if any due to the mining activities. The present study attempts to understand the occurrence and distribution of natural uranium and ^{226}Ra in ground water, especially around a uranium mine at Nawapahar of Singhbhum thrust belt Jharkhand, India and to estimate ingestion dose to the member of the public of different age groups due to intake of these radionuclides through water.

Experimental

Site description

Nawapahar uranium mine (22°41′N and 86°16′E) is situated in central region of Singhbhum thrust belt of Jharkhand, India. This is a modern trackless mine in the country with decline to access to the underground and ramp access to the stopes. This mine is about 15 km away from south of Tatanagar and 12 km west of Jaduguda uranium mine. The average ore grade of Narwpahar uranium mine is 0.042% U_3O_8 (or 0.036% U). Twelve locations were selected for estimation of natural uranium and ^{226}Ra content in ground water samples within a 10 km radius around the mine. The environmental map of the area investigated in present study, indicating the sampling locations, is demonstrated in the Fig. 1.

Measurement technique

For estimation of uranium and ^{226}Ra concentration in ground water principally used for drinking purposes, tube well/boreholes samples were periodically collected, within a 10 km radius from the surrounding villages of the mine. Samples were collected after at least 5–10 min of pumping to evacuate more than 3–5 times of tube well/boreholes storage volume. Ten litres of water samples were collected in a previously conditioned polythene container by nitric acid, from the selected location around the mine. The water samples were acidified immediately with nitric acid and subsequently, uranium and ^{226}Ra analyses were carried out in Environmental Survey Laboratory of Environmental Assessment Division, Bhabha Atomic Research Centre, set up at Narwapahar mining site. For estimation of uranium (U) in the environmental samples, whether water or soil, chemically separated U is firstly fused with the fusion mixture comprising of NaF and Na_2CO_3 in the furnace at about 700 °C and then subjected to ultraviolet radiation in the fluorimeter. A standard solution of U is also processed similarly. As the intensity of the fluorescence is directly proportional to U content of the sample, the comparison yields the U content of that to be analysed. The 3650 Å excitation and 5546 Å fluorescence wavelength are unique to uranium [10].

Further, the adequate stirring has the potential to liberate waterborne ^{222}Rn completely from the water sample, which constitutes the working principle of ^{222}Rn and ^{226}Ra estimation using the emanometric technique [11]. For ^{226}Ra estimation by emanometry, water sample is acidified with concentrated HNO_3 and 2 L of it is evaporated to about 70 mL and stored in the ^{222}Rn bubbler. The solution in the bubbler is aerated to remove any ^{222}Rn present. The bubbler is then closed and allowed to stand for a known period usually 7–10 days for adequate build-up of ^{222}Rn. An evacuated scintillation cell having ZnS (Ag) as detector is coupled airtight to the top of the bubbler through a swagelock connector. The stopcocks of bubbler are opened and air is sucked into the cell. A sintered glass disc (porosity: 20–30 μm) in the bottom of the bubbler breaks up the air stream into tinny bubbles which give rise to mechanical agitation for liberating ^{222}Rn from all along the water column. The collected ^{222}Rn sample in scintillation cell is delayed for 3 h to attain equilibrium between ^{222}Rn and its daughters and consequently, it is counted for α-activity to compute dissolved ^{222}Rn and hence the ^{226}Ra content is then quantified. The efficiency of the system is 75% and allowing for maximum build-up period, the minimum measurable activity levels for ^{226}Ra are 4.0 and 0.8 mBq L^{-1} for 10 min and for >100 min counting periods, respectively.

Quality assurance and quality control

The quality of the data is assured by analyzing IAEA matrix reference materials. The precision of the analyses is within ±8% of the certified values, which is given in Table 1. The reliability of the method is further assured by cross-method checks, spike recovery and replicate analysis. All laboratory glassware used for sample processing was

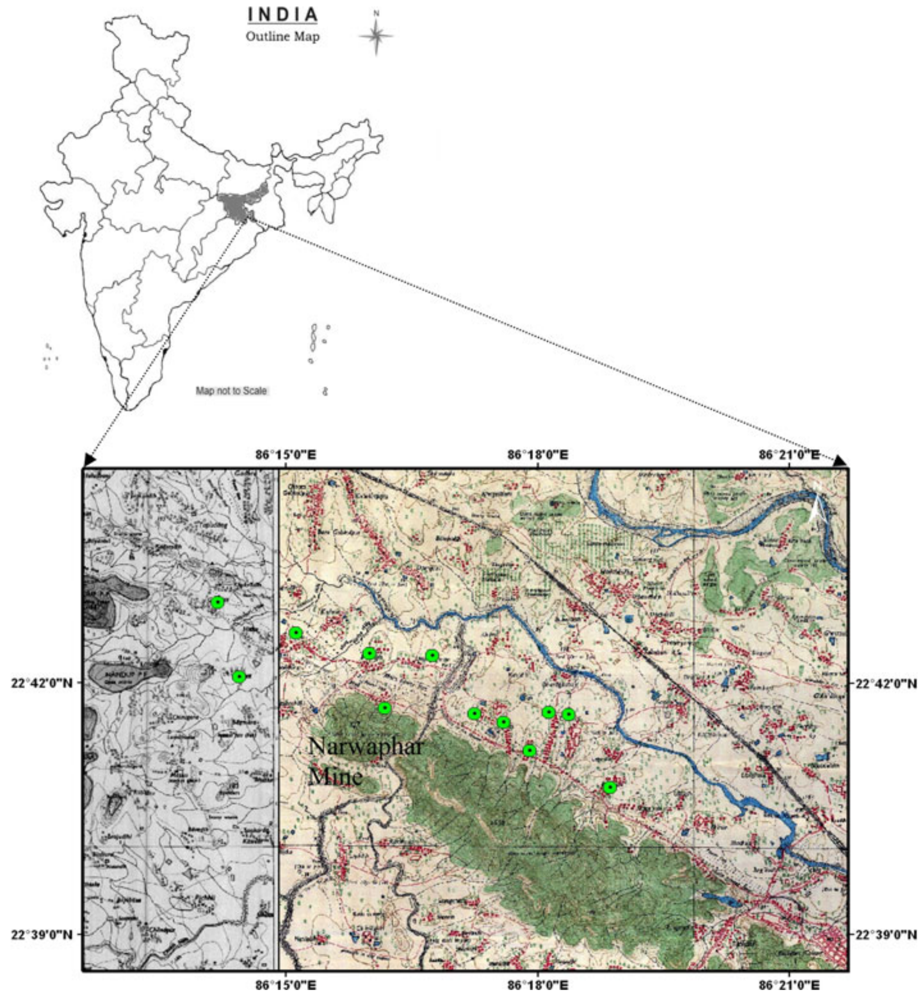

Fig. 1 Map showing the study location around the surrounding villages of the Narwapahar uranium mine

Table 1 Mean concentrations (ranges) of U and ^{226}Ra obtained for IAEA reference materials

Reference material	Radionuclide	Unit	Concentration	Certified value
			Observed value ($n = 7$)	
IAEA RGU-1	U	mg kg^{-1}	399 (392–405)	400 (398–402)
IAEA-326 soil	^{226}Ra	Bq kg^{-1}	32.8 (30.5–33.8)	32.6 (31–34.2)

soaked in 10% nitric acid for 15 days and then rinsed thoroughly with distilled and double distilled water, respectively before use. Reagent blank was taken along with each batch of sample preparation and concentrations observed in the reagent blank were subtracted from the same batch of samples.

Results and discussions

Uranium and ^{226}Ra concentration

Table 2 presents statistical data analyses results of total uranium and ^{226}Ra concentrations in ground water samples around the mine. The natural uranium concentration in the ground water samples was found to vary from 0.1 to 3.7 μg L^{-1} with mean of 0.87 ± 0.73 μg L^{-1}. Various health and environmental protection agencies have recommended the safe limit of uranium in drinking water for human being. WHO [12] has recommended 15 μg L^{-1} in water as safe limit where as USEPA [13] has recommended 30 μg L^{-1} of uranium in water as the safe limit for drinking water. Further, UNSCEAR [6] and ICRP [14] have recommended the safe limits of uranium in drinking water as 9 and 1.9 μg L^{-1}, respectively. All the measurements were found to be well below the recommended limits as per WHO [12], USEPA [13] and UNSCEAR [6]. However, only in 92% water samples, uranium content is

Table 2 Statistical analyses results of U (nat.) and ^{226}Ra concentration in ground water samples around Narwapahar mine

	Uranium concentration (μg L^{-1})	^{226}Ra concentration (mBq L^{-1})
No of data	103	99
Mean	0.87	13.73
Std.dev.	0.73	7.34
Median	0.63	11.50
Range	0.10–3.75	5.2–38.10
Geometric mean	0.65	12.24
Geometric SD	2.4	1.59
25th percentile	0.38	8.80
75th percentile	1.08	17.20

well below the recommended limit as per ICRP [14] and about 8% water samples having uranium content was found to be higher than 1.9 μg L^{-1}. Further, from the results, it may be assumed that there is no movement of radionuclides from the mining site to the ground water in the vicinity and whatever variation in uranium concentration in ground water samples is observed, may be thought of due to the local geological formation of soil and the rocks. The frequency distribution of natural uranium concentration in ground water samples has been studied and is presented Fig. 2. The distribution of the data shows that about 43% of samples contain uranium <1 μg L^{-1} and 36% sample contain <1.5 μg L^{-1}. However, only 4% sample contain uranium >3 μg L^{-1}. The first quartile, median and 3rd quartile of the data was found to be 0.38, 0.63 and 1.08 μg L^{-1}, respectively. Uranium content in ground water samples collected around the Narwpahar uranium mine was comparable with the other parts of India and worldwide values as shown in Table 5, except a few high values such as 0.37–75.3 μg L^{-1} in France, 0.04–12146 μg L^{-1} in Finland, 0.03–48.6 in Germany and 0.008–56.63 μg L^{-1} in China (Table 3).

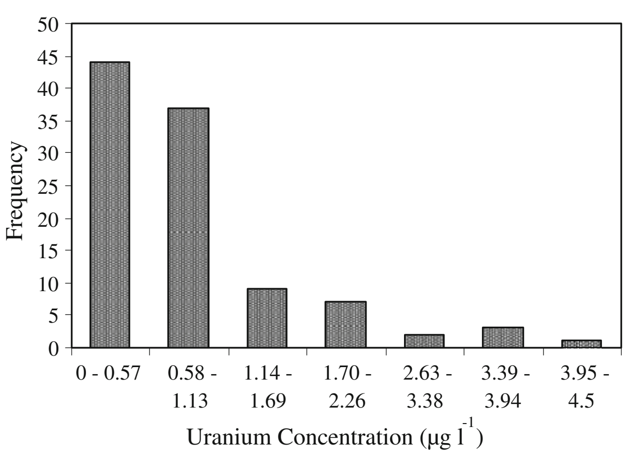

Fig. 2 Frequency distribution of natural uranium concentration in ground water samples

Further, the ^{226}Ra concentration in ground water samples was found to vary from 5.2 to 38.10 mBq L^{-1} with mean of 13.73 ± 7.34 mBq L^{-1}, which is well below the national and international recommended limits. According to the WHO [15] guidelines for drinking water quality, ^{226}Ra concentrations should not exceed 1000 mBq L^{-1} based on daily consumption of 2 L of water and the United States' Environmental Protection Agency has also been proposed the limit of 500 mBq L^{-1} for ^{226}Ra in drinking water [16]. Thus, all investigated ground water samples are found to be acceptable for consumption. Frequency distribution of ^{226}Ra in ground water samples shows (Fig. 3) that about 65% of water samples contain ^{226}Ra of 10–21 mBq L^{-1} and about 12% samples contain 21–38 mBq L^{-1}. However, only 22% of sample contain ^{226}Ra < 10 mBq L^{-1}. The first quartile, median and 3rd quartile of the data was found to be 8.80, 11.50 and 17.20 mBq L^{-1}, respectively. ^{226}Ra content in ground water samples collected around the Narwpahar uranium mine was found to be comparable with other worldwide reported values (Table 5), except a few high values such as 7–700 mBq L^{-1} in France, 0.4–600 mBq L^{-1} in Germany and 10–49000 mBq L^{-1} in Finland. From this study it is observed that the levels of uranium and ^{226}Ra content in ground water samples are well within the respective derived water concentration (DWC) limits [17].

Assessment of ingestion dose

Ingestion dose due to intake of uranium and ^{226}Ra through the drinking water pathway for different age groups was calculated using ICRP dose coefficients [18] and prescribed water intake rates for different age groups.[1] The annual ingestion dose was calculated by the following relation:

$$D\left(\mathrm{Sv\ y^{-1}}\right) = 365 \times \mathrm{DWI} \times \mathrm{DCF} \times C \quad (1)$$

where, D is the ingestion dose (Sv y^{-1}), C is the mean concentration of a particular radionuclide in the water (Bq L^{-1}), DWI is the daily water intake for a specific age group (L day^{-1}) and DCF is the dose conversion factor for a particular radionuclide and for a specific age group (Sv Bq^{-1}).

The water intake rates taken for the infants of 0–6 months and 7–12 months are 0.7 and 0.8 L day^{-1}, respectively, whereas those for the age groups of 1–3, 4–8, 9–13, 14–18 year and adults are 1.3, 1.7, 2.4, 3.3 and 3.7 L day^{-1}, respectively. The water intake rates for female in the age group 9–13, 14–18 year and adults are 2.1, 2.3 and

[1] Dietary reference intakes for water, Food and Nutrition Board, Institute of Medicine, National Academies Press, Washington, DC, USA (www.npa.edu)

Table 3 Comparison of natural uranium and ^{226}Ra concentration of the present study with other parts of India and worldwide values in drinking water samples

S. no	Location	Uranium concentration (μg L^{-1})	^{226}Ra concentration (mBq L^{-1})	References
1	Narwapahar, India	0.10–3.75 (AM = 0.87)	5.2–38.1 (AM = 13.73)	Present study
2	United States	0.02–6.23	0.4–1.18	UNSCEAR [6]
3	France	0.037–75.3	7–700	UNSCEAR [6]
	Germany	0.03–48.6	0.4–600	UNSCEAR [6]
4	Finland	0.04–12146	10–49000	UNSCEAR [6]
5	Romania,	0.03–3.00	0.7–21	UNSCEAR [6]
6	China, Asia	0.008–56.63	0.2–120	UNSCEAR [6]
7	Poland	7.3	1.7–4.5	UNSCEAR [6]
8	Italy	0.02–9.66 (AM = 1.96)	0.5–60.8 (AM = 8.85)	Jia et al. [19].
9	Haryana, Himachal Pradesh and Punjab, India	1.08–20.19	–	Singh et al. [20].
10	Fujian province, China	0.03–13.4 (GM = 0.54)	1.2–941 (GM = 12.7)	Zhuo et al. [21].
11	Argentina	0.04–11.0 (GM = 1.3)	GM = 4.4	Bomben et al. [22]

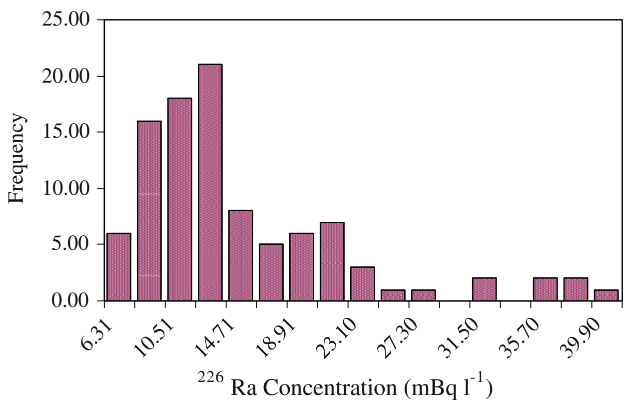

Fig. 3 Frequency distribution of ^{226}Ra concentration in ground water samples

2.7 L day^{-1}, respectively. In the present study, individual concentration of ^{235}U, ^{234}U and ^{238}U was estimated by multiplying the mean concentration of uranium with their individual specific activity and relative abundance in nature, respectively (Table 4). Consequently, ingestion dose was estimated based on individual isotopic concentration by employing Eq. 1 and total dose due to natural uranium through intake of water to the respective age group was estimated by adding the doses calculated from individual isotopes. Similarly, the ingestion dose due to intake of ^{226}Ra also estimated by employing Eq. 1 and using other

Table 4 Natural uranium isotopic abundance and individual specific activity

Isotopes of natural U	Half life (y)	Specific activity (Individual)(Bq/mg)	Mass abundance (%)
^{238}U	4.47×10^9	12.45	99.27
^{235}U	7.04×10^8	80.09	0.72
^{234}U	2.44×10^5	232.1×10^3	0.005

respective parameters like water intake rates for different age groups, ^{226}Ra concentration in water and dose conversion coefficient of ^{226}Ra for different age groups. The annual ingestion dose due to intake of uranium and ^{226}Ra through drinking water pathway to various age groups of male and female is reported in Table 5. It is evident from the results that a maximum ingestion dose of 2.22 μSv y^{-1} due to intake of natural uranium through water was observed to the infant (7–12 months) and a minimum dose of 0.99 μSv y^{-1} was observed to adult female. This is due to the fact that about 10 times the higher dose coefficient for infants (7–12 months) has been observed as compared with adult female, though the water intake is less for infants. Ingestion dose due to intake of uranium to infants (0–6 months) is about 14% less as compared to infants (7–12 months) which can be attributed to the higher intake rate of water for infants (7–12 months). Further, the ingestion dose to the children (1–3 and 4–8 year), female (9–13 and 14–18 year) and male (9–13 year and adults) was found to be comparable. Again for the age group 14–18 year male, the ingestion dose due to intake of uranium is slightly higher than the age group 9–13 year male, this is due to higher water intake, though the dose coefficients are comparable.

Ingestion dose due to intake of ^{226}Ra through drinking water path way was estimated for different age groups. A maximum dose of 24.81 μSv y^{-1} was found to be observed to the age group 14–18 year male and minimum dose of 3.79 μSv y^{-1} was observed to adult female. Similarly the ingestion dose due to intake of ^{226}Ra to the infants (0–6 and 7–12 months) and female in the age group 14–18 year was found to be comparable. In general, the higher values of ingestion dose due to intake of ^{226}Ra were observed in infants and teens.

The total mean ingestion dose due to intake of natural uranium and ^{226}Ra through drinking water to adult male

Table 5 Age dependent mean ingestion dose to the different age groups due to intake U (nat.) and ^{226}Ra through intake of water

Different life stage	Age group	Mean ingestion dose due to U(nat.) (μSv y^{-1})	Mean ingestion dose due to ^{226}Ra (μSv y^{-1})	Total mean ingestion dose (μSv y^{-1})
Infant	0–6 months	1.94 ± 1.62	16.49 ± 8.81	18.43
	7–12 months	2.22 ± 1.85	18.84 ± 10.07	21.06
Children	1–3 years	1.27 ± 1.06	6.32 ± 3.38	7.59
	4–8 years	1.12 ± 0.93	5.28 ± 2.82	6.4
Male	9–13 years	1.33 ± 1.11	9.74 ± 5.21	11.08
	14–18 years	1.82 ± 1.51	24.81 ± 13.26	26.63
	Adult (>18 years)	1.36 ± 1.13	5.19 ± 2.78	6.55
Female	9–13 years	1.17 ± 0.97	8.52 ± 4.56	9.69
	14–18 years	1.27 ± 1.05	17.29 ± 9.24	18.56
	Adult (>18 years)	0.99 ± 0.82	3.79 ± 2.03	4.78

and female was found to be 6.55 and 4.78 μSv y^{-1}, respectively, which is well below the recommended dose limits of 100 μSv y^{-1} as per WHO [12]. Similarly, for other age groups the mean ingestion dose due to intake of natural uranium and ^{226}Ra through drinking water were found to be well below the WHO prescribed reference dose level. The mean annual ingestion dose to the members of the public in study region for all the groups was found to be 12.8 μSv y^{-1}, which is less than 5% of the global average ingestion dose [6].

Conclusions

The preliminary investigation reveals that, the natural uranium concentration in the groundwater samples collected from the surrounding environment of the Narwapahar mine was found to vary from 0.1 to 3.75 μg L^{-1} with mean of 0.87 ± 0.73 μg L^{-1} and ^{226}Ra concentration was found to vary from 5.2 to 38.1 mBq L^{-1} with mean 13.73 ± 7.34 mBq L^{-1}. The levels of uranium and ^{226}Ra in ground water samples around the uranium mine are found to well below national and international regulatory limits. The total mean annual ingestion dose due to natural uranium and ^{226}Ra through drinking water pathway to male and female adults population residing around the mine was estimated to be 6.55 and 4.78 μSv y^{-1}, respectively which constitutes merely a small fraction of the reference dose level of 100 μSv y^{-1} as recommended by WHO. In general Ingestion dose due to intake of natural uranium and ^{226}Ra through water was found to be marginally higher for infants and teens as compared to children and adults. From the radiological risk point of view, the ground water samples collected from the surrounding environment of the mine are very safe and can be used for drinking purpose.

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