Natural radioactivity concentration in traditional Thai herbal medicine

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Abstract. The aim of this study is to create a database of radioactivity concentration and annual effective dose of the naturally occurring radionuclides: ²²⁶Ra, ²²⁸Ra, ⁴⁰K and ²¹⁰Po in Thai medicinal herb plants. 99 types of popular Thai medicinal herb plants (total of 214 samples) such as Curcuma comosa Roxb., mucuna, white kwao krua, barbed grass, black galingale, lingzhi mushroom, Ginkgo biloba, jiaogulan, plu kaow, turmeric, ginger, safflower, moringa, gotu kola and garlic were randomly collected from drug stores and herbal fair. The activity concentration of ²²⁶Ra, ²²⁸Ra and ⁴⁰K in all herb samples was determined by gamma-ray spectrometry while that of ²¹⁰Po was determined by alpha spectrometry. The activity concentration was found to range from <0.20 to 89.92 Bq kg⁻¹ for ²²⁶Ra, from <0.10 to 39.62 Bq kg⁻¹ for ²²⁸Ra and from 4.83 to 2761.33 Bq kg⁻¹ for ⁴⁰K and from 0.32 to 47.13 Bq kg⁻¹ for ²¹⁰Po. The highest activities concentration of ²²⁶Ra, ²²⁸Ra, ⁴⁰K and ²¹⁰Po were found in the white kwao krua, jiaogulan, plu kaow and ginkgo, respectively. The average total annual effective dose due to ingestion of these herbal plants was assessed to range from 0.0001 to 0.0327 mSv y⁻¹, with highest dose found in jiaogulan.

1. Introduction
The use of medicinal plants for treating diseases is the oldest existing method that humanity has used to cope with illnesses. In Thailand, people began to use plants as medicines predates for more than a thousand years (AD 1238-1377). Thai herbs, known as Sa-mun-phrai in the local language, have been used to prevent and treat diseases and ailments or to support health and healing. The knowledge of traditional medicine based on the usage of Thai medicinal herbs has been gradually developed, systematized, revised, recorded, and passed on from generation to generation throughout the country’s history, as a means of health care for the Thai people. [1] After the World Health Organization began to promote national traditional heritage in 1977, every Thai government has broadly stated in the
country’s public health policy to support research and development of medicinal plants, and over the years the interest and importance of herbal healing has continued to grow. [2]

For therapeutic purposes, specific parts of the herb (aerial parts, root, leaves, fruit, flowers, and seeds) and their extracts are used in medicinal products all over the world. It is estimated that 25% of modern medicines are directly or indirectly derived from local medicinal plants. About 80% of the world population (especially in developing countries) use traditional medicine plants for their primary source of healthcare. [3] Research on plant has increased all over the world and the large body of collected evidence shows the immense potential of medicinal plants used in various traditional systems. [4]

Climatic conditions, different places, as well as environmental factors can affect the medical properties and effectiveness of medicinal plants, and may also have a direct influence on their growth in the place of their occurrence and cultivation. [5] An emerging problem in many developing countries is the industrial waste pollution that threatens the health of the local environment and causes contamination of vegetation by heavy metals, pesticides, and radioactivity. [6] Plants can transfer radioactivity into human nutrient cycle and ecosystem directly by vegetable food products and indirectly by animal food products. Plants used as food commonly have $^{40}$K, $^{232}$Th and $^{238}$U and their progenies such as $^{210}$Po. [7] Therefore, these similarities would be found in plants used for medicinal purposes since plants are the primary pathway of natural radionuclides to enter the human body via the food chain. Thus, the contamination level in the medicinal plants should therefore be monitored and analyzed. Moreover, natural radionuclides such as $^{238}$U, $^{232}$Th and $^{40}$K are found in every human being to certain degree of radiation exposure. [8] According to many studies so far, $^{210}$Po primarily fall on the surface of the plants from the atmosphere, where they can stick for a shorter or a longer period depending on the surface quality of leaves. High values were typically found in tobacco samples due to their longer growing season and special leaf-surface. [9-11] The estimation of risk to humans from medicinal plants through ingestion requires a quantitative understanding of the interrelated pathways by which the radionuclides are eventually ingested by humans. [12] Therefore, it is important to study the uptake and activity distribution of radionuclides and the probable effective radiation dose to humans, by the use of medicinal plants.

The objectives of this study are to determine the specific activity concentrations and the annual effective doses due to ingestion of NORM due to $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po present in selected medicinal plants commonly used in Thailand and to assess the radiological risk associated with the use of these medicinal plants.

2. Materials and Methods

Samples of medical plants were collected from various local markets and drugstores in Thailand. All samples had been certified and licensed by the Thai Food and Drug Administration under the Ministry of Public Health. They included 99 kinds of popular Thai medicinal herb plants (214 samples) used extensively for treating various diseases or complementary medicine. Table 1 displays Classification of medicinal plants on the basis of parts utilized for the present study, as well as their common names, scientific names, and the particular part of the plant used. Each sample had been dried, blended and packaged inside a capsule. All samples have patented license and been certified by the Food and Drug Administration, Ministry of Public Health, Thailand.

2.1 Analytical method

The Thai medicinal herb plant samples, in powder form, were removed from inside of their plastic capsules. They were dried at 80 °C in an oven for 6 hours. Two different analytical techniques were used to determine the natural radionuclides: Gamma spectrometry for $^{226}$Ra, $^{228}$Ra and $^{40}$K and Alpha spectrometry for $^{210}$Po.

2.1.1 Gamma spectrometry. All samples were analysed for activity concentration of $^{226}$Ra, $^{228}$Ra and $^{40}$K using gamma spectroscopy. The dry mass value was used to determine the radionuclide
concentration. Each dried sample, about 100 g, was put in a cylindrical plastic container (2.5 inch in height and 3 inch in diameter). The samples were sealed and kept for at least 30 days in order to ensure the radioactive secular equilibrium before taken for measurement by gamma spectrometric method [13]. Activity concentrations in the samples were measured using one of two high-purity germanium (HPGe) detectors with a relative efficiency of 30 and 60 % in a low background configuration. The $^{226}$Ra and $^{228}$Ra were determined by measuring their gamma emitter daughters, $^{214}$Bi (609 keV), and $^{228}$Ac (911.1 keV), while $^{40}$K was measured via the 1460 keV gamma line. Efficiency calibration was performed using reference samples (RGU, RGTH and RGK) provided by the International Atomic Energy Agency, IAEA. The lower limits of detection for the measured natural radionuclides derived from the background measurements at 48 hours were 0.2 Bq kg$^{-1}$ for $^{226}$Ra.

2.1.2 Alpha spectrometry. The $^{210}$Po were measured using the standard technique (the silver disc technique) [14]. Five grams of dry wt. of samples were spiked with 0.2 Bq of $^{208}$Po (polonium chloride in 0.1 mol L$^{-1}$ HCl provided by Amersham International, UK) as a yield tracer. Each sample was digested using concentrated nitric acid for at least 24 h; hydrogen peroxide was also added to help in oxidizing the organic compounds. When the solution was clear, the sample was then gently evaporated to near dryness. The residue was then dissolved in 100 mL of 0.5 mol L$^{-1}$ hydrochloric acid. The solution was then heated to 80 °C and $^{210}$Po was spontaneously plated onto a rotating silver disc after reduction of iron with ascorbic acid. Alpha counting of $^{208}$Po (5.15 MeV) and $^{210}$Po (5.3 MeV) was done using an alpha spectrometer (ORTEC’s system) with a passive ion implanted silicon detector (active area at 300 mm$^2$, background counts per day at 3.6 and the minimum depletion thickness at 100 mm). The $^{210}$Po activity was corrected for recovery by comparison with measured activity of the $^{208}$Po yield tracer and for radioactive decay since the time of sampling. The lower limit of detection of the method used was 0.4 Bq kg$^{-1}$.

2.2 Quality control procedures
Quality control procedures were applied using reference samples provided by IAEA and duplicate analysis. In addition, all methods and laboratories used in this study are given according to the literature. [15]

2.3. Calculation of radionuclides and annual committed effective dose
Following the spectrum analysis, counting rates for each detected photo peak and activity per mass unit for each of the detected nuclides are calculated. The specific activity (in Bq/kg) is given by the expression (1).

$$A_{sp} = \left( \frac{N}{t} - \frac{N_0}{t_0} \right) \cdot \left( I_{\gamma} \cdot \varepsilon \cdot m \right)^{-1}$$

Where, $N$ is the net counts of a given peak, $t$ = 48 hour is the counting time for the sample, $N_0$ is the background of the given peak, $t_0$ = 48 hours is the counting time for the background, $\varepsilon$ is the detection efficiency, $I_{\gamma}$ is the number of gamma photons per disintegration and $m$ is the mass in kg of the measured sample.

If there is more than one peak in the energy analysis range for a nuclide, an average of the peak activities is made, and the result is then the weighted average nuclide activity. The total uncertainty value is composed of the random and systematic errors in all factors involved in producing the final nuclide concentration result listed in Table 2.

Having obtained the values for the specific activity concentrations of the individual naturally occurring radionuclides in the medicinal plants, the average annual committed effective dose, $E_{ave}$, for ingestion of naturally occurring radioactive materials (NORMs) in the Thai medicinal herb plants were calculated using the expression (2) given by [15].
\[ E_{\text{ave}} = I_p \cdot DCF_{\text{ing}} \cdot A_{\text{sp}} \]  

Where \( DCF_{\text{ing}} \) is the dose conversion factor for ingestion, for each radionuclide (i.e., \( 2.8 \times 10^{-4} \) mSvBq\(^{-1}\), \( 6.7 \times 10^{-4} \) mSvBq\(^{-1}\), \( 6.2 \times 10^{-6} \) mSvBq\(^{-1}\), and \( 1.2 \) µSv Bq\(^{-1}\) for \( ^{226}\text{Ra} \), \( ^{228}\text{Ra} \), \( ^{40}\text{K} \) and \( ^{210}\text{Po} \), respectively, for an adult) [17-19]. \( I_p \) is the consumption rate from intake of NORMs in medicinal plants, \( A_{sp} \) is the activity concentration in the plant sample.

| Parts used | Scientific name | Common name | Number of samples |
|------------|----------------|-------------|-------------------|
| 1. Seeds   | 1. Mucuna pruriens (L.) DC. | 1. Mucuna | 2 |
|            | 2. Piper nigrum | 2. Black pepper | 4 |
|            | 3. Coriandrum sativum | 3. Coriander seed | 1 |
| 2. Flower  | 1. Carthamus tinctorius Linn. | 1. Safflower | 4 |
|            | 2. Hibiscus sabdariffa Linn. | 2. Rosella, Jamaican Sorel, Roselle, Rozelle, Sorrel, Red Sorrel | 2 |
|            | 3. Ganoderma lucidum (Curvis) P. Karst | 3. Lingzhi mushroom | 2 |
|            | 4. Clitoria ternatea L. | 4. Butterfly pea or Blue pea. | 2 |
| 3. Leaves  | 1. Morus alba Linn. | 1. Mulberry tree, White Mulberry | 1 |
|            | 2. Paederia linearis Hook. | 2. Skunk-vine | 1 |
|            | 3. Centothea lappacea (L.) Desv. | 3. Barbed grass | 3 |
|            | 4. Andrographis paniculata (Burm.f.) Wall.ex Nees. | 4. Kariyat, The Creat. | 11 |
|            | 5. Orthosiphon aristatus (Blume) Miq. | 5. Car's Whiskers | 4 |
|            | 6. Moringa oleifera Lam. | 6. Horse radish tree, Drumstick | 4 |
|            | 7. Gynostemma pentaphyllum (Thunb.) Makino. | 7. Jiaoglukan, Gynostemma, Miracle grass | 4 |
|            | 8. Acanthus ebracteatus Vahl | 8. Sea holly, Thistlelike plant | 4 |
|            | 9. Thunbergia laurifolia L. | 9. Laurel clockvine, Blue trumpet vine | 6 |
|            | 10. Gymnema inodorum (L.) Decne. | 10. | 2 |
|            | 11. Biophytum adiantoides Wight ex Edgew. & Hook. | 11. Little tree plant, Sensitive plant. | 1 |
|            | 12. Camellia sinensis var. assamica | 12. Assaam Tea, Thea | 1 |
|            | 13. Rhinacanthus nasutus (L.) Kurz | 13. White crane flower | 1 |
|            | 14. Capparis micrantha DC. | 14. | 1 |
|            | 15. Ginkgo biloba L. | 15. Salisburya adiantifolia, Maidenhair tree, Forty-coin tree | 2 |
|            | 16. 7. Senna alexandrina Mill. | 16. Alexandria Senna | 4 |
|            | 17. Ventilago denticulata Wild. | 17. | 2 |
|            | 18. Stevia rebaudiana Bertoni M. | 18. Stevia | 2 |
|            | 19. Strychnos roborans A.W. Hill., S. lucide R. Br. | 19. Saintignatus Bean. | 1 |
|            | 20. Eclipta prostrata (L.) L. | 20. False daisy, White head, Yerbadetajo herb | 1 |
|            | 21. Senna tora (L.) Roxb. | 21. Acapulo, Candelabra bush, Candle bush, Candlestick senna, Christmas candle, Ringworm bush, Ringworm senna, | 1 |
|            | 22. Litsea glutinosa (Lour.) C.B.Rob. | 22. | 1 |
|            | 23. Lysiphyllum strychnifolium (Craib) A.Schmitz | 23. | 1 |
|            | 24. Schefflera leucantha R.Vig. | 24. Edible-stemmed Vine | 1 |
|            | 25. Piper sarmentosum Roxb. | 25. Wildbetal Leafbush. | 1 |
|            | 26. Gymnanthem u extensum | 26. Bitter Leaf | 1 |
|            | 27. Lasiandus cyanocarpus Jack | 27. | 1 |
| 4. Fruits  | 1. Morinda citrifolia L. | 1. Great morinda, Tahitian noni, Indian mulberry | 4 |
| Parts used | Scientific name | Common name | Number of samples |
|------------|----------------|-------------|-------------------|
| 2. Momordica charantia L. | 2. Bitter gourd | | 3 |
| 3. Perilla frutescens (Linn.) Britton | 3. Perilla | | 1 |
| 4. Phyllanthus emblica L. | 4. Emblic myrrhan, Malaaca tree. | | 2 |
| 5. Syzygium aromaticum (L.) Merr. & Perry. | 5. Clove. | | 1 |
| 6. Terminalia bellirica (Gaertn.) | 6. Beleric Myrobalan | | 2 |
| 7. Aegle marmelo Correa. | 7. Bael | | 1 |
| 8. Illicium verum Hook.f. | 8. Chinese star anise, Star anise, Star aniseed, Badian, Badian, Badian khatai, Bunga lawang, Thakolam | | 1 |
| 9. Piper retroversum Vahl | 9. Long Pepper | | 1 |
| 5. Pod | Oroxylum indicum (L) Kurz | Broken Bone tree, Damocles tree, Indian Trumpet Flower | 1 |
| 6. Bark | 1. Cinnamomum spp. | 1. Cinnamon | 4 |
| 2. Cinnamomum porrectum Kosterm | 2. Safrol laurel | | 1 |
| 7. Trunk | 1. Coscinium fenestratum (Goetgh.) Coebr. | 1. Weniwel, Calumba wood, Kalumbuka | 2 |
| 2. Barleria strigota Wild. | | | 1 |
| 3. Derris scandens(Roxb.) Benth. | 3. Jewel vine, Hog creeper, Malay jewevine | | 3 |
| 4. Roureopsis stenoptetala (Griff.) Schellenb. | 4. | | 1 |
| 5. Cissus quadrangularis L. | 5. Stemed Vine, Ashisanbare | | 3 |
| 6. Piper interrumpent Opiz, Piper ribesoides Wall. | 6. | | 1 |
| 7. Salacia chinensis | 7. | | 1 |
| 8. Tinospora crispa (L.) Miers ex Hook.f. & Thom | 8. Heart-leaved moonseed | | 5 |
| 8. Tuber | 1. Curcuma comosa Roxb. | 1. Java turmeric | 5 |
| 2. Butea superba Roxb. | | 2. | 4 |
| 3. Stephania venosa (Blume) Spreng | | 3. Blylache bush | 1 |
| 4. Puaria candollei Graham ex Benth. var mirifca | | 4. Thai kadzu | 6 |
| 5. Allium sativum L. | 5. Garlic | | 3 |
| 6. Smilax corbularia Kunth | 6. | | 2 |
| 7. Curcuma aromatica Salisb. | 7. Wild Turmeric | | 1 |
| 8. Amorphophallus konjac K.Koch | 8. Konjac | | 1 |
| 9. Cyperus rotundus Linn. | 9. Nut grass | | 2 |
| 9. Rhizome | 1. Curcuma comosa Roxb. | | 1 |
| 2. Sansevieria cylindrica Bojer. | | 2. | 1 |
| 3. Curcuma aeruginosa Roxb. | | 3. | 1 |
| 4. Boesenbergia rotunda (L.) Mansf. | 4. Kaempfer | | 4 |
| 5. Zingiber cassumunar Roxb. | 5. Phlai | | 1 |
| 6. Curcuma longa | 6. Turmeric | | 10 |
| 7. Zingiber officinale Roscoe | 7. Ginger | | 4 |
| 8. Boesenbergia cf. rotunda (L.) Mansf., Boesenbergia Sp | 8. Kaempfer | | 1 |
| 9. Curcuma comosa | 9. | | 1 |
| 10. Zingiber zerumbet (L.) Roscoe ex Sm. | 10. Shampoo ginger, Wild ginger | | 1 |
| 11. Curcuma zedoaria (Christm.) Roscooe | 11. Zedoary | | 1 |
| 12. Curcuma mangga Valeton & Zijp. | 12. Curcuma white | | 1 |
| 13. Curcuma aurantiaca Van Zijp. | 13. | | 1 |
| 14. Euryale ambeinensis (Loud) | 14. | | 1 |
| 15. Ludisia discolor (Ker Gawl.) A.Rich. | 15. | | 1 |
3. Results and Discussions

Table 2 shows the radioactivity concentrations (Bq kg$^{-1}$) of the natural radionuclides $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po in the medicinal plants for this study. All natural radionuclides were determined in 99 kinds of Thai medicinal herb plants in a total of 214 samples. The data showed that all Thai medicinal herb plants had activity concentrations in the range of less than 0.20 to 89.92 Bq kg$^{-1}$ for $^{226}$Ra, less than 0.10 to 39.62 Bq kg$^{-1}$ for $^{228}$Ra, from 4.83 to 2761.33 Bq kg$^{-1}$ for $^{40}$K and from 0.32 to 47.13 Bq kg$^{-1}$ for $^{210}$Po. Based on Table 2, the activity concentration of $^{40}$K was relatively high in all the samples. Tuber (White kwao kruea) had the highest activity concentration of $^{226}$Ra. Leaves (Jiaogulan and Ginkgo) had the highest activity concentration of $^{228}$Ra and $^{210}$Po, respectively. In addition, the highest activity concentration of $^{40}$K was recorded for all parts used (Plu kaow).

The variations in the activity concentrations is most likely due to differences in the geological location of the plants and the radiochemical composition of the soils in which these medicinal plants were grown or cultivated. The levels of activity concentration of natural radionuclides are not normalized across the globe, and each plant has ability to absorb particular elements more than the others [16].

In this paper, the assumption that a unit consumption rate ($I_p$) of 1 kg per annum was used. The number is derived from the medical labels which received the certificate and license from The Thai Food and Drug Administration of the Ministry of Public Health. The average total annual committed effective doses due to ingestion of $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po in the medicinal herb plants were calculated using the corresponding values of activity concentrations for each of the medicinal plants. The results are presented in Table 2. The results varied from 0.0001 to 0.0327 mSv y$^{-1}$ with an average value of 0.0036±0.0006 mSv y$^{-1}$. The highest annual effective dose was found in leaves (Jiaogulan). On the other hand, the highest average was recorded for whole medicinal-plant part, while barks, bud and Pollen showed the lowest average.
Table 2. Radioactivity concentration of $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po (in Bq kg$^{-1}$) and annual committed effective dose (mSv y$^{-1}$) in the Thai medicinal herb plant samples (dry weight).

| Part used of Medicinal plants | Average of Activity concentration in Bq kg$^{-1}$ (Range) | (Range) Average of Activity concentration in Bq kg$^{-1}$ | Average Annual effective dose (mSv y$^{-1}$) |
|------------------------------|----------------------------------------------------------|----------------------------------------------------------|---------------------------------------------|
| 226Ra                        | (<0.20-2.57) 0.86±0.36                                  | (<0.10-5.39) 1.80±0.25                                    | (316.56-520.71) 3.21±0.09                  | (0.0006-0.0042) 0.0034±0.0001 |
| 228Ra                        | (<0.20-2.18) 0.91±0.40                                  | (<0.10-3.67) 0.82±0.18                                    | (92.07-544.64) 0.90±0.12                  | (0.0007-0.0045) 0.0028±0.0002 |
| 40K                          | (<0.20-2.57) 2.55±0.73                                  | (<0.10-39.62) 3.74±0.64                                   | (24.17-1126.69) 6.75±0.18                 | (0.0001-0.0327) 0.0052±0.0008 |
| 210Po                        | (<0.20-2.66) 1.00±0.39                                  | (<0.10-8.31) 1.59±0.80                                    | (43.74-1050.52) 7.03±0.21                 | (0.0007-0.0105) 0.0035±0.0006 |

From the viewpoint of the annual effective dose and the activity concentration of $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po in the medicinal herb plants compared with this study (the conservative comparison), Table 3 showed the results from published work with the annual committed effective dose and the activity concentration of NORMs in the different medicinal herb plants conducted in some countries [16,20-25]. The average activity concentrations of both $^{226}$Ra and $^{228}$Ra in our study appear to be lower than the values obtained in South India, Iraq, Nigeria, Ghana, Serbia, Italy and Brazil. However, our study showed lower average activity concentration of $^{40}$K than the South India, Ghana, Serbia, Italy and Brazil, but higher average concentrations than Iraq and Nigeria. In the case of $^{210}$Po for this study, the average activity concentration of Thailand is lower than that of South India but higher than that of Italy. Also, the average annual committed effective dose due to the ingestion of $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po in Thai medicinal herb plants in this study was slightly higher than the results reported in Nigeria [22] but much lower than that reported in South India [20].
Table 3. Comparison of the activity concentration of $^{226}$Ra, $^{228}$Ra, $^{40}$K and $^{210}$Po (in Bq kg$^{-1}$) in the medicinal plants from this study with those from other countries.

| Country | $^{226}$Ra | $^{228}$Ra | $^{40}$K | $^{210}$Po |
|---------|------------|------------|---------|-----------|
|         | Range      | Average    | Range   | Average   |
| Thailand [This study] | <0.2-89.9 | 1.6 | <0.1-39.6 | 2.6 |
| South India [20] | 2.7–11.2 | 6.7 | 2.4–8.7 | 5.0 |
| Iraq [21] | <1.1-12.6 | 4.7 | <1.3-14.6 | 2.9 |
| Nigeria [22] | 10.8–42.5 | 25.0 | 27.8–41.1 | 35.1 |
| Ghana [16] | 20.4–46.9 | 31.8 | 42.0–70.6 | 56.2 |
| Serbia [23] | 0.6–8.2 | 2.6 | 1.7–15.1 | 7.4 |
| Italy [24] | <0.3-16.6 | 1.9 | - | - |
| Brazil [25] | - | - | <11–43.0 | 21.7 |

Annual effective dose (mSv$^{-1}$)

- Thailand: 0.0036
- South India: 0.0075-0.1067
- Iraq: 0.0034
- Nigeria: 0.0043-0.0069
- Ghana: 0.0054
- Serbia: -
- Italy: -
- Brazil: -

4. Conclusions

This research work provides baseline data for regulations and quality control of medicinal plants used in Thailand. Most of corresponding average annual effective dose determined in this study due to the ingestion of natural radionuclides in Thai medicinal herb plants is far below the average radiation dose of 0.3 mSv received per person worldwide [18]. In this research result presents insignificant annual committed effective dose due to the use of these Thai medicinal herb plants into Plant Medicine. Therefore, the radiological hazard associated with intake of the natural radionuclides in the medicinal plants is insignificant. Hence, the Thai medicinal herb plants samples from this research are considered safe in terms of the radiological hazard.

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