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Identification of Potentially Relevant Radionuclides in the Nuclear Central of Angra dos Reis

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ABSTRACT

A radiologically significant nuclides catalog is paramount to the classification of radioactive waste. In order to produce such a catalog, it is required to know the isotopic composition of the radioactive waste produced in the nuclear power plants, their isotopic inventories and both short and long term toxicity for each relevant nuclide in each exposure scenario. Estimating the waste produced in a power plant is an old problem that still poses a great challenge, even with current technology and methods. This paper describes an attempt at estimating the radionuclide concentration levels of the radioactive waste produced in the Angra dos Reis nuclear power plant. A review on the various methods used around the world to estimate these isotopic compositions was needed in order to achieve such a result. Alongside the review was used a computer simulation with the Origen 6.0 code and calculations to find out the future activities and toxicities for each analyzed radionuclide in each considered exposure scenario. The resulting data is used to build on a radiologically significant nuclides catalog that can be used as guiding tool for the development of radiological containment policies. This data will be helpful for the long term storage of the studied radionuclides.

1. INTRODUCTION

Angra 1 is the first nuclear power plant built on Brazil. It started operation in 1984 and still provides electric power to the present day. Its reactor is a PWR type made by Westinghouse and its main fuel is enriched Uranium. The nominal raw electric power in Angra-1 is around 640 MWe [1].

Starting commercial operation in February of 2001, Angra-2 was the second Brazilian nuclear power plant. It is also a PWR type plant by Westinghouse maker, with nominal raw electric power around 1350 MWe [2]. These two power plants comprise what is called The Angra dos Reis Nuclear Power Central.

The radioactive waste produced in Angra dos Reis is stored on a temporary storage located on the power plants themselves. Currently, there are around 80 thousand barrels containing radioactive waste produced in the power plant, with different kinds of waste, such as spent filters, resins, and solid waste.

A large amount of these wastes should be directed to a waste repository as soon as possible. Since the barrels content is a mix of many kinds of waste and different radionuclides, it is required special attention upon characterization, considering all the effects of each individual nuclide type.

Once the barrels are sealed, it is very costly and troublesome to remove their contents. This is the reason for using theoretical methods of characterization that dismiss the need of reopening the barrels.

1.1. Relative Radiological Relevance

In order to classify the radioactive waste considering all changes regarding different forms of radioactive decay and the spawning of daughter nuclides, it was proposed the concept of Relative Radiological Relevance. The relevance of a radionuclide is considered based on attributes such as its half-life, total amount present on the waste, along with the overall toxicity for different exposure scenarios, and the projection of these attributes on the future.

The first and major factor considered for the radiological relevance is a nuclide half-life. Since the radioactive waste management in nuclear power plants is based on long term safety, only nuclides with half-lives of at least two years were evaluated. It was assumed that nuclides with too short half-lives would mostly likely decay while stored on the temporary storage, being of little to no significance on the figures of the total concentration on the waste after all the years on the temporary storage. Therefore, all nuclides with half-life inferior to two years were promptly discarded from the calculations.

The second factor is the total concentration of the radionuclide on the radioactive waste. The larger the amount of a nuclide during the evaluated period, the more dangerous it is. Nuclides with minimal concentration were promptly discarded from the evaluation.

The third and final factor is the relative radiological risk, which measures how a radionuclide toxicity fares in an exposure scenario compared to other radionuclides. It is often directly proportional to the concentration of the nuclide present on the radioactive waste.

The Relative Radiological Relevance compares the nuclides in different exposure scenarios and different periods of time, classifying each nuclide in comparison with the others and ranking them from the most dangerous to the least dangerous.

Waste that offers few risks on the long term does not need to be stored in a permanent repository, but ratter at an intermediate storage, granting more capacity on repositories for more dangerous radionuclides. Since repository storage capacity is limited, it is paramount to classify all nuclides accordingly in order to ensure that all, or at least most of the more dangerous radionuclides are going to be stored properly.

1.2. Relevant Radiological Waste Catalog

Many countries control their most dangerous radioactive wastes by using a catalog of relevant radionuclides [3]. It is an catalog that contains a list of radionuclides that are more common, have larger half-lives and are toxic enough to be considered more dangerous than the others regarding long term storage on repositories [4].

These radionuclides are classified in order to help the planning of the containment measures, for safety policies and for developing treatment protocols in cases of exposure [5]. Each country has freedom to choose their own relevant radionuclides [6], but most catalogs share a large amount of these nuclides in common [7].

Up to the present moment, Brazil still does not have a radiologically relevant radionuclide catalog. The absence of this catalog, along with other problems, slows down the creation of an inventory classification program and halts the construction of a geological repository, since many of the engineering and safety aspects require previous knowledge of physical and chemical characteristics of the nuclides that are going to be stored and their total amount.

2. OBJECTIVES

The objective was to find out whether there is any way to estimate a quantitative value for the initial concentration of each significant radionuclide in the Angra dos Reis nuclear central waste and classify the radionuclides in a relevant radionuclide catalog.

All objectives should be met using only theoretical methods. Verifying the possibility of attaining this data without the use of the more expensive and time consuming experimental methods was a key objective in order to help to develop a national radioactive waste classification plan.

3. MATERIALS AND METHODS

3.1. Bibliography Review

A bibliography review was needed in order to acknowledge typical values of concentration in similar power plants (PWR) around the world. Obtained data was compiled in a set of tables that will not be discussed in this paper, but rather on a more complete version currently on the IPEN digital library [8].

This data is important because of the known Gaussian correlation between fission products [9] in PWR power plants. There is also a weaker correlation between activation products. These nuclides require that the power plants have similar auxiliary and cooling systems.

Most information found about the subject describes methods and general information about the planning of studies on the concentrations of radionuclides found in power plants waste. There are only a few cases of papers that disclose actual explicit quantitative results.

3.2. Scale 6.0 Software

Alternatively, the software Origen, part of the Scale 6.0 software pack, was used to estimate the production of radionuclides on Angra dos Reis since the plants began operation. Obtained results were then compared to expected values known from the bibliographic review.

Scale 6.0 is a nuclear technology research software pack owned by the Oak Ridge National Laboratory. The software is distributed to institutes by a paid subscription business model and is available to use in some of IPEN laboratories computers [10].

The Origen software is used to simulate the formation of radioactive waste on nuclear power plants based on the initial conditions given by the user. Origen offers several options for calculations and can be used in many different power plant scenarios. The code is written in FORTRAN language and the output is in text format.

Using Origen to estimate the concentration levels of radioactive waste in power plants is a common method in American power plants and is considered reliable by the Electric Power Research Institute (EPRI) [11]. Output fidelity is directly proportional to the total amount of initial conditions information known by the user.

3.3. Estimating the Radiological Relevance

Having the initial concentrations and activities of each radionuclide of the waste, it is possible to estimate the toxicity levels for each nuclide and calculate the radiological risk for all exposure scenarios considered. Further detail about how this is done is found on the aforementioned reference [12].

Dose and time are the main factors to estimate the radiological relevance of a radionuclide. Nuclides that are more toxic for a longer period of time along a given interval are considered to be more relevant in this fixed interval.

Regardless, it is important to note that the radiological relevance is only one of the many possible means of classification. It is subject to modifications or even replacement if further data is researched or different criteria are considered.

4. RESULTS

4.1. Bibliography Review

Many hypotheses were taken from the studied bibliography and used to support the initial conditions on Origen calculations. These hypotheses are responsible for covering some gaps in the known data about the Angra dos Reis nuclear power plant and makes the calculations easier to do.

A very useful and verified hypothesis is that the difference between calculations using the real operation times of a power plant and a calculation using the average operation time is around the same order of magnitude, and therefore realistic operational time is not necessary at all for an estimative [11].

Of remarkable contribution to this study was an paper about the American nuclear power plant located in Surry and Comprised of two PWRs by Westinghouse maker that are very similar to the Angra dos Reis reactors, having roughly the same power output.

The data about the isotopic concentration levels of Surry radioactive waste was obtained using Origen 2, an older version of Origen 6.0. This makes Surry a reasonable enough model for Angra dos Reis. Before proceeding with the calculations for Angra dos Reis, the results for Surry were compared with known data from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and EPRI [13].
In that paper, Surry results were compared to the H.B. Robinson power plant data. Results were consistent between the two power plants.

An inconsistence between the Origen 2 calculations and the experimental data on Surry was noted in the paper when comparing the results of Tecnetium-99 and Iodine-129 [14]. Nevertheless, it is known that the radiochemical method used in the experiment is considered as having a low overall accuracy.

Uncertainties for the Origen 2 method are considered to be around one order of magnitude [15]. This value was estimated comparing the results of computer simulations with radiochemical sampling. However, as of now there is still no known form to estimate an exact global uncertainty to the method, requiring that each evaluated power plant do its own radiochemical sampling in order to find a more reliable uncertainty.

Data acquired through sampling vary more randomly than computer simulation data. It was observed that if there is a large amount of samples in a given sampling analysis the results tend to converge to the same value of the computer simulations. This is actually expected to be the case because of the Large Numbers Law [16].

4.2. Calculating Angra dos Reis Isotopic Concentration

Using all available data about the fuel, shielding and auxiliary systems structure of Angra-1 and Angra-2 as initial conditions on Origen, it was possible to obtain a numeric value for the isotopic concentration of the radioactive waste produced in the power plants.

Both plants had their isotopic concentrations calculated for January of 2022. This specific date was chosen in order to make future comparisons between experimental data and the simulations easier. Total activity for each nuclide in Angra-2 is displayed in **Figure 1.** Graphics for Angra-1 can be found in reference [8]

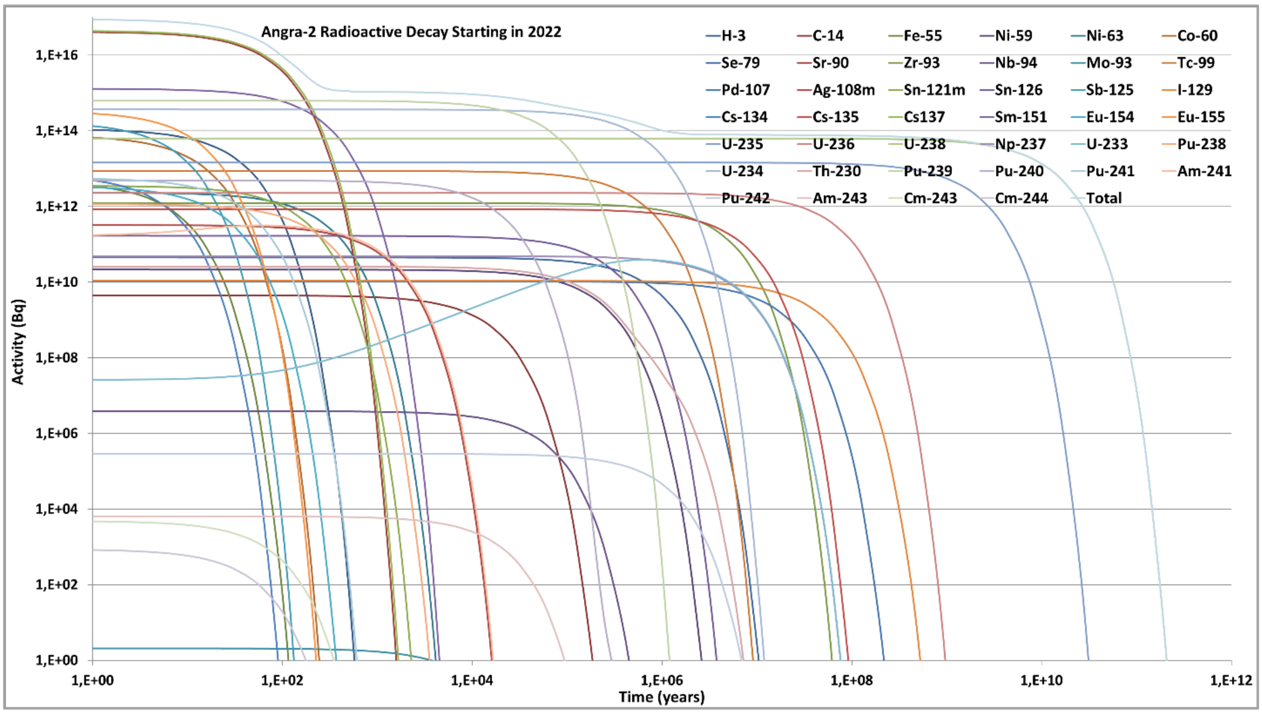

Figure 1: Total activity for each relevant radionuclide in Angra-2.

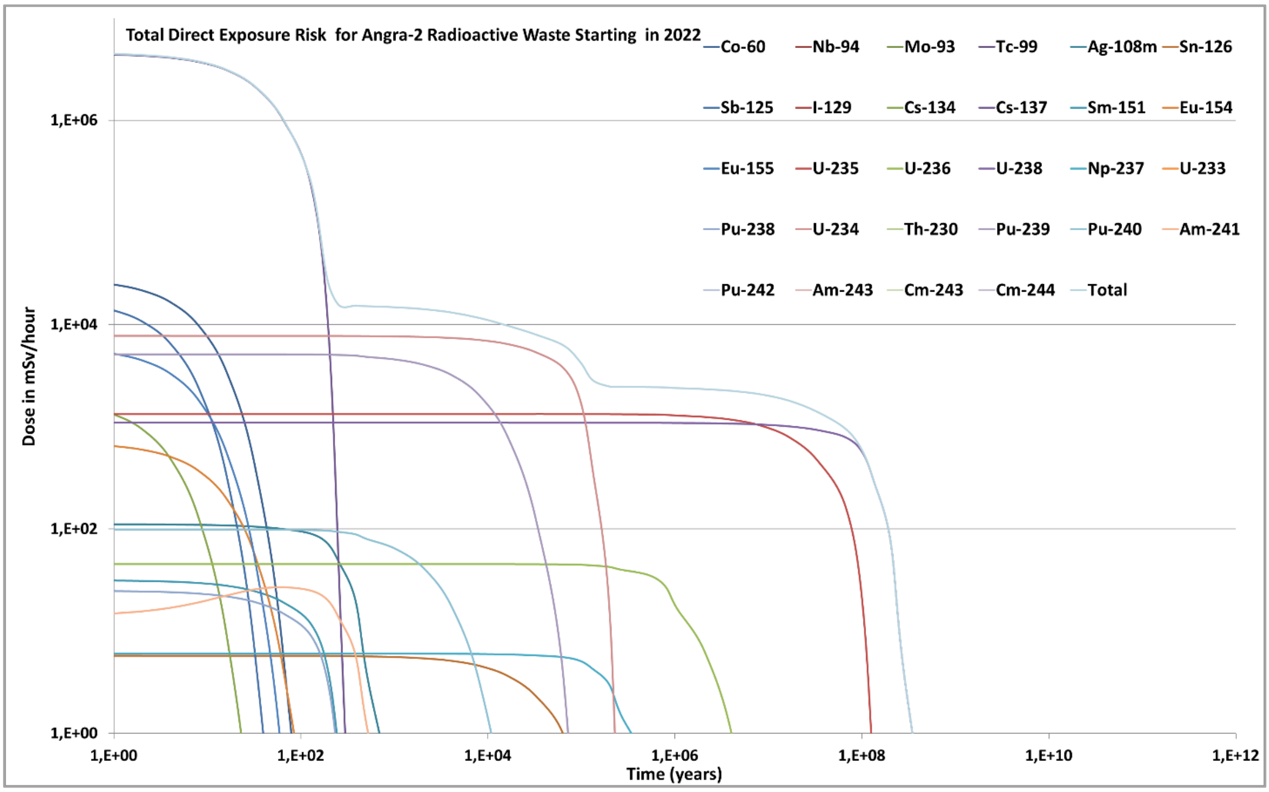

Figure 2: Direct exposure risk for each Angra-2 relevant radionuclide.

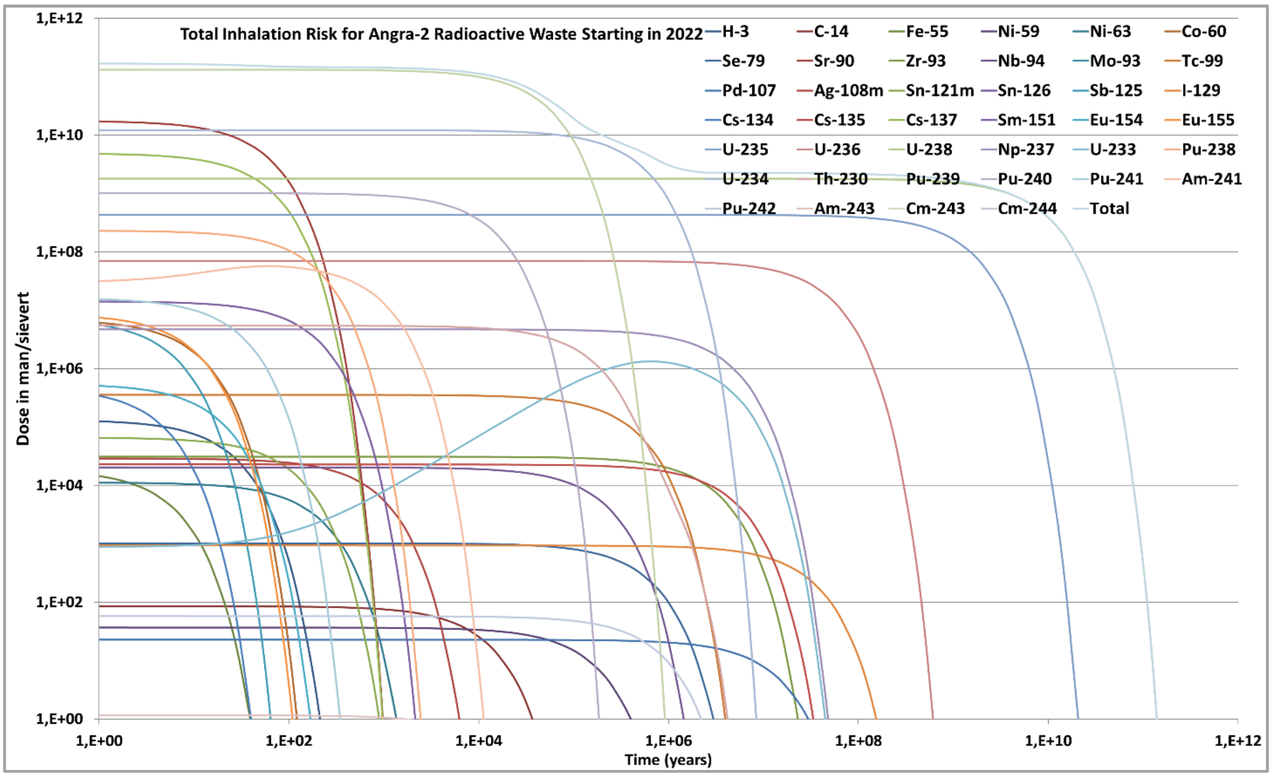

Figure 3: Inhalation risk for each Angra-2 relevant radionuclide.

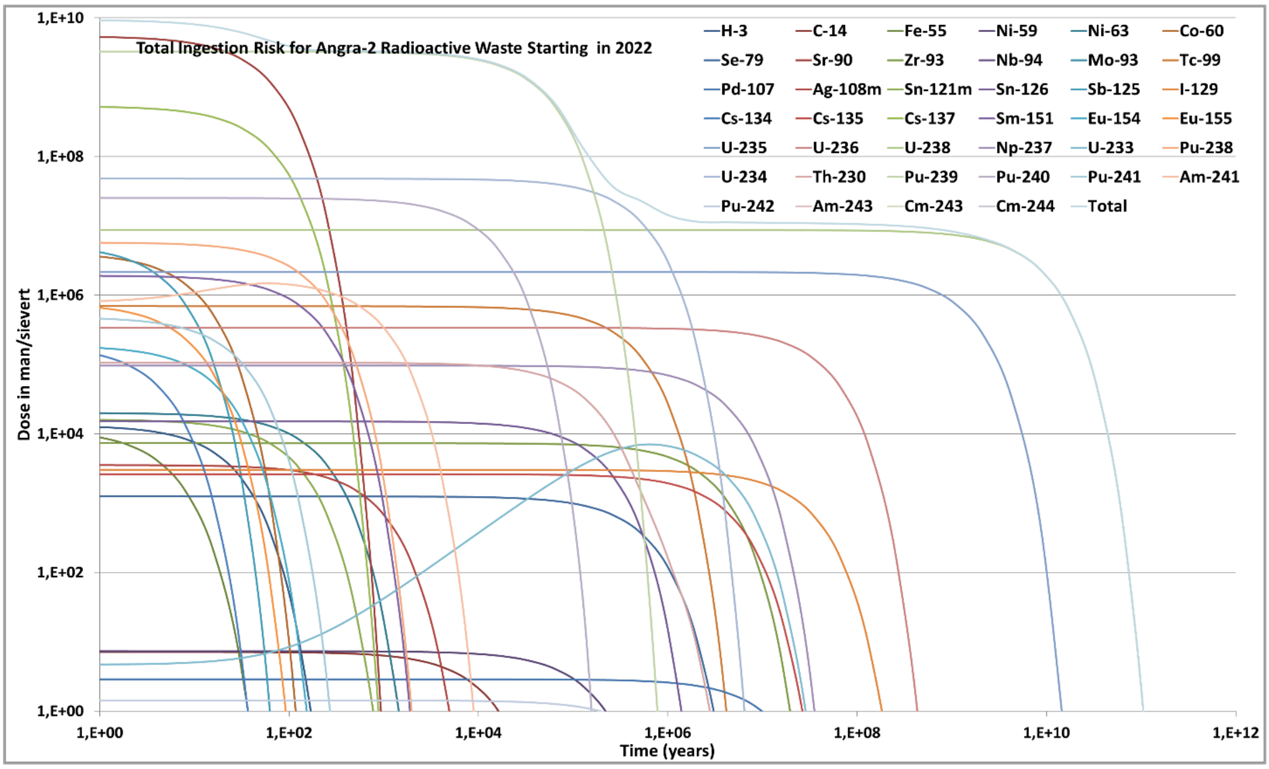

Figure 4: Ingestion risk for each Angra-2 relevant radionuclide.

The results were plotted on many different Excel spreadsheets, with graphics for the radioactive decay of the radioactive waste and toxicity levels for each exposure scenario. **Figure 2, Figure 3** and **Figure 4** show the total risk for each exposure scenario on Agran-2. Several graphics and more detailed information can be found in reference [8]. These graphics resulted in the creation of a radionuclide catalog that ranks the most important nuclides that can be found on the radioactive waste of the Angra dos Reis nuclear central. Angra-2 was chosen as an example for exposing the results, since they are not that different from either Angra-1 or Surry.

All projections for the future consider only nuclides created up until January of 2022. Nuclides generated after this date are omitted from the calculations. This is hardly ideal, since the power plants will continue to generate radioactive waste after the aforementioned date. However, it should also be noted that after the radiochemical sampling analysis these calculations will need to be done again, and on that occasion a more rigorous approach should consider all the radionuclides during the power plants lifetime.

For storage purposes, nuclides generated after January 2022 are of few immediate relevance, since the radioactive waste that is already in intermediate storage does not have any chosen final destination yet.

5. CONCLUSIONS

Bibliographical reviews showed that it is possible to estimate the isotopic concentrations of a PWR power plant waste. Knowledge of this information led to the calculation of such concentrations and the subsequent toxicity associated with each radionuclide for all exposure scenarios considered.

All results were met using only theoretical methods and projections for these results on the future were made, making way for the development of a relevant nuclide catalog.

Lack of detailed information about the composition of the auxiliary systems in the Angra dos Reis power plants led to a semi-quantitative uncertainty that might be over one order of magnitude for the initial isotopic concentrations. It is very likely that the activity values for Cobalt-60 and Iron-55 are greatly underestimated by at least two orders of magnitude, considering the values on the bibliography.

An independent work parallel to this paper was in production [17]. Early data from this other paper showed agreement with many of the obtained results. However, low concentration levels of Uranium-235 and Samarium-151 along with an excess of Plutonium-238 and Plutonium-234 were observed. It is believed that these differences are a result of the different methods and approximations for the shielding and auxiliary systems of each power plant. Further review, comparisons and debate between these papers are expected to happen in the future.

Radiochemical methods might validate this paper findings in the future or point out errors that can be used to refine the calculations. Nevertheless, it is expected that the relative radiological relevance of each radionuclide remains nearly the same.

A relevant radionuclide catalog is paramount for developing containment and storage policies. This catalog is expected to be one of the main tools in the classification of radioactive waste that will undergo permanent disposal in the geological repositories.

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