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A study of actinide decay chains on the environmental effect of a geologic disposal of 'rock-like oxide' fuels and uranium-plutonium oxide fuels

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Abstract

Currently, it is planned to dispose rock-like oxide fuel (ROX) spent fuels, like standard light water reactor (LWR) spent fuels and high-level radioactive wastes from reprocessing plants, in a repository located in a deep geologic formation. In order to demonstrate the usefulness of the ROX plutonium-burning system, a comparative study of environmental safety was carried out for the direct disposal of ROX and mixed-oxide fuel (MOX) spent fuels, both of which recycle plutonium. Analyses were performed for the case of ROX and MOX spent fuels disposed into a deep granite rock formation, and the individual dose equivalent, of actinide decay nuclides, due to the ingesting of contaminated drinking water was calculated based on a groundwater migration scenario. The amounts of disposed ROX and MOX spent fuels are equivalent to the generation of electricity by one metric ton of uranium metal (1 MTU). The committed dose equivalents from ROX spent fuels are less than those from MOX spent fuels by ca. 2 orders of magnitude in the conservative solubility case, and by at least one order of magnitude in the realistic solubility case. It is suggested that for actinide decay nuclides the ROX plutonium-burning system will be beneficial to repository safety. In order to confirm the behavior of geologic disposal of ROX spent fuels, it is recommended that the solubility of zirconia should be estimated by experimental studies. © 1999 Elsevier Science B.V. All rights reserved.

1. Introduction

There are more than 220 metric tonnes of highly enriched plutonium (²³⁹Pu) in the world [1,2]. About 100 metric tonnes of this will be recovered from dismantled warheads in the near future. For their management, several options have been proposed and discussed internationally from the viewpoints of security, environmental safety, technological feasibility and economy [3,4]. Of these, the adoption of a fission option seems to be favorable because of the necessary technologies are well established.

Until the commissioning of commercial fast breeder reactor (FBR), it may be reasonable to make use of the well established MOX-LWR recycling. The recycling, however, rapidly degrades the plutonium quality and produces a considerable amount of plutonium with heavier mass numbers, as well as trans-plutonium elements such as Am and Cm. Thus, it is to be expected that the recycling will be limited to only a few times because of licensing and technological difficulties. After a few cycles, therefore, it becomes attractive to adopt the once-through process with a 'rock-like oxide' fuel (ROX), and their through burning plutonium in conventional LWR.

JAERI has been studying a once-through burning system of excess plutonium. In this system, multi-phase rock-like fuels are fabricated based on conventional nuclear fuel technologies. They are irradiated in LWR to generate electricity. The spent fuels obtained will be physically and chemically stable, highly radioactive materials, which could be disposed after 30–50 yrs cooling without further processing. In this paper, a comparative study of environmental safety was carried out for the direct disposal of ROX and MOX spent fuels, which recycle plutonium, in order to demonstrate the usefulness of ROX plutonium-burning system.

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2. Disposal concept and assessment scenario

Disposal concepts of spent fuels have been developed in many countries; USA [5], Canada [6], Sweden [7], Germany [8], etc. The disposal concepts of USA and Germany are based on disposal into unsaturated tuff and bedded salt layers, respectively. Both concepts are not applicable for the Japanese situation taking account of geological conditions in Japan. On the other hand, the concepts developed in Canada and Sweden are based on disposal into a crystalline bedrock which is one possible formation for geologic disposal in Japan. Therefore, we assume here a disposal system similar to the Swedish concept KBS-3. KBS-3 is based on a multi-barrier concept (metallic container–buffer material–geosphere) to protect humans and environment, and is similar to the disposal concepts of several other countries.

The disposal system assumed here consists of the spent fuel rods enclosed into a metallic container (steel canister) with plugging material (glass beads), and the metallic container emplaced within a disposal borehole covered with the buffer material (bentonite). After failure of the container, the shape of buffer material will not be changed significantly, because of a physical effect by the plugging material. The buffer material is expected to self-heal gaps, seal small fissures in the adjoining rock, and buffer the composition of intruding groundwater chemically. Radionuclide transport through the buffer material will be dominated by a diffusion process, because the hydraulic conductivity of compacted bentonite is very low (less than 10^{-11} m s⁻¹ [9]).

The spent fuel is assumed to be disposed in a deep and stable granite rock mass at a depth in the range of 500–1000 m. The repository will be constructed in a stable rock mass not intersected by any major fracture which may be connected with regional fracture zones in order to avoid the occurrence of a short circuit of groundwater from the repository to the biosphere, as shown in Fig. 1(a). Accordingly, radionuclides released from the repository migrate slowly through the stable rock, and are then transported to the biosphere via fracture zones.

The safety of the disposal system depends on the performance of engineered barriers (a waste matrix, a container, a buffer zone, and concrete structures if necessary) to confine radionuclides in the repository, and the ability of natural barriers to retard and dilute radionuclides during the transport in the geosphere and the biosphere. The concept of the disposal system assumed here relies on the following processes:

- The container isolates the waste contained therein from contact with groundwater during the initial period after closure of the repository, before the initiation of radionuclide release.
- After container failure, the rate of radionuclide release is controlled by the dissolution rate of the ma-



Fig. 1. Hydrological profile around the repository system. (a) Location of a potential radioactive waste repository and fractured zones. (b) Conceptual model of repository and geosphere.

trix and also by the solubility of relevant radionuclides.

- The buffer zone (bentonite), through which radionuclides are transported by diffusion and may be absorbed, which retards the release of radionuclides from the repository.
- The geosphere, where most radionuclides released from the repository are effectively retained and delayed by various retention mechanisms, retards the transport of the radionuclides to the biosphere.
- The radionuclides finally entering into the biosphere will be diluted by a large volume of groundwater in adjacent aquifers, and with surface water bodies such as river, lake and/or ocean, before coming into contact with man.

The safety assessment considered here is based on a deterministic groundwater migration scenario, assuming that the performance of the disposal system is not affected by unfavorable events (e.g. earthquake, inadvertent human intrusion, etc.) as is the case of normal evolution scenarios. It is assumed that all the components of the repository are eventually saturated with groundwater.

3. Safety assessment methodology

A safety analysis was conducted by using the computer code system GSRW [10] which was developed for the generic safety assessment of geologic disposal of HLW. The GSRW evaluates the radiological consequences for an individual human or a population due to radionuclide released from geologic radioactive waste repositories based on the normal evolution scenario (groundwater migration scenario).

The GSRW is composed of three interlinked models, a source term, a geosphere and a biosphere model. The first model evaluates the release from the engineered barrier system. The second model analyzes the transport of radionuclides in the geosphere, using analytical or numerical solutions of a mass transport equation involving a one-dimensional advection, a three or a onedimensional dispersion, linear adsorption and coupled decay chains. The third model assesses the transport of radionuclides in the biosphere and the resulting radiological consequences to man, which is based on a dynamic compartment model for the biosphere and dose factors for dose calculations. More detailed information about the source term and geosphere models are described in the following section.

3.1. Source term model

The source term model defines the flux of the radionuclides released from the repository into the surrounding rock formation. The repository may involve components such as waste form encapsulated in the metallic containers, and buffer material. The source term model, therefore, takes into account of following processes:

- The corrosion of the metallic container (steel canister) to determine the time when the dissolution of the matrix begins.
- The flux of a specific radionuclide at the interface between the waste form and the buffer material is determined by the dissolution of the waste matrix, which is controlled by a diffusion in the buffer material and a specific solubility of main component of the waste matrix (UO₂ for the MOX spent fuels, zirconia (ZrO₂) and thoria (ThO₂) for the ROX spent fuels), or the leaching of the radionuclide controlled by a diffusion through the buffer material and a specific solubility of the radionuclide.
- The diffusion-controlled transport of radionuclides through the buffer material, taking account retardation processes, to determine the source term to the geosphere.

The repository after the closure will be gradually refilled with groundwater and reach an equilibrium hydraulic pressure. During this process an inward water flow at a very slow rate will be established and no radioactivity will diffuse out of the repository until the equilibrium pressure has been achieved. The estimation of the time needed for this process requires a complex transient water flow analysis, and the result depends on the repository design and site-specific geohydrologic conditions. The GSRW code does not involve this transient analysis, as it is intended to perform a generic safety assessment of repositories. Also, no radioactivity can be leached out of the waste form which is encapsulated in a container until the container has failed and the form has begun to contact with water. The container failure time depends on the degradation rate of the container, which depends on the electrochemical environment around the container. In this model, the container is taken as a user-defined factor determining a delay time to the onset of radionuclide leaching from the waste.

It is assumed that, upon container failure, the waste form comes in contact with water and radionuclides would be leached out of the form depending either on the dissolution rate of the waste form which is modeled here as a sphere or a spheroid (Model 1), or on their solubility limits (Model 2). In the first model, radionuclides are assumed to be released congruently with the waste form (UO₂, ZrO₂, or ThO₂) in which they are homogeneously distributed on a macroscopic scale. The release of UO₂, ZrO₂ and ThO₂ are modeled by a diffusion mechanism in an infinite medium. The second model does not involve the dissolution process of the waste form, but assumes that the concentration of a specific radionuclide (with lower solubilities than UO₂, ZrO_2 and ThO_2) in water at the boundary between the waste form and the buffer material is controlled by its solubility limit.

The subsequent transport of radionuclides through the buffer zone is modeled by a one-dimensional diffusion through a finite medium in Cartesian coordinates (Model 1), or it is solved including the leaching behavior of radionuclide in spherical coordinates (Model 2). In both models, the concentration of a specific radionuclide is governed by adsorption equilibrium between water and the buffer material.

3.2. Geosphere model

Geologic media assumed here are homogeneous porous media intersected by several dominant fractures connected with groundwater in a sedimentary soil layer or with the ocean, as schematically shown in Fig. 1(b). The groundwater-mediated migration of radionuclides in the homogeneous porous media surrounding the repository is computed by an analytical solution of the mass transport equation involving one-dimensional advection, three-dimensional dispersion, retention and decay chains. The transport of radionuclides through the pipe-like fractures is computed in an analogous way. Although radionuclide retention includes a number of physico-chemical mechanisms such as reversible and irreversible chemical adsorption, physical adsorption, molecular diffusion, filtration, precipitation, aggregation and so on, the identification of all of the mechanisms that will take place is far from being understood. Even if each potential mechanism is successfully identified and could be modeled precisely, the computation with such

models would require a large number of parameters which are not yet available or insufficiently known (e.g., speciation data, kinetic parameters corresponding to each interaction mechanism, geochemical equilibrium data, a complete mineralogical composition of the geologic media, and also the local variation of these data). Therefore, the single retention mechanisms were not specifically distinguished in the model used here, but their total effects on the migration of radionuclides was represented by an empirical distribution coefficient, K_{d} , defined as the ratio of the amount of radionuclide retained on the rock to the amount in the solution. The use of K_d in the model is based on the following assumptions: that K_d is independent of the concentration of the radionuclide, that each radionuclide migrates independently of the others, and that each retention occurs instantaneously and reversibly.

4. Basic assumptions and input data

This assessment of the potential radiological consequences of geologic disposal of MOX and ROX spent nuclear fuels is generic and does not apply to a site specific. However, it might be required in future to define the disposal system to some extent so as to enable the analysis of transport of radionuclides in geologic formations and the biosphere following entry of groundwater into a repository and the release of them to the surrounding strata.

4.1. Radionuclide inventory

The comparative study was carried out based on the radionuclide inventory equivalent to the generation of electricity by 1 MTU in LWR (38 000 MWd). Cell burnup calculations were performed with the SRAC code system [11] based on the JENDL-3 nuclear data

[12]. In the present study, two kinds of ROX fuels; PuO₂stabilized ZrO₂-Al₂O₃ + 20 mol% ThO₂ (Zr/Th: thoria type fuel) and PuO₂-stabilized ZrO₂-Al₂O₃ + 10 mol% UO₂ (Zr/U: zirconia type fuel), were analyzed. The additives ThO₂ and UO₂ were used to avoid small reactivity coefficients of zirconia fuel. Table 1 shows radionuclide inventory of spent fuels 1000 yr after discharge from a PWR with a burnup rate 0.18 MWd/cell and an irradiation time of 1000 days. The radiological consequences of direct disposal of spent nuclear fuels were estimated only for α -nuclides. Because crack data about spent nuclear fuel pellets of MOX and ROX are currently not available, although they are very important information to evaluate the quantities of radionuclides (¹²⁹I, ¹³⁵Cs, ³⁶Cl, etc.) which immediately release to groundwater after failures of the container and fuel cladding material.

4.2. Source term

In the ROX fuels, almost all radionuclides are homogeneously distributed into sparingly soluble fuel matrices (zirconia and thoria), and their leaching is determined by the dissolution rate of the fuel matrices (Model 1). On the other hand, the fuel matrix of MOX is UO_2 , and solubility limits of some elements (Np, Th, etc.) included in the fuel matrix are lower than that of UO_2 . If the atom ratios Np/U and Th/U in the fuel are higher than the corresponding solubility ratios, the leaching of radionuclides are controlled by their solubility limits (Model 2).

This calculation assumes for simplification that the combined lifetime of the container and fuel cladding material is 1000 yr, and the release of radionuclides initiates 1000 yr after the closure of the repository. The thickness of buffer material is assumed to 0.35 m (KBS-3), and the porosity of buffer is 0.3. The diffusion coefficient in the buffer material is assumed to be 10^{-10} m² s⁻¹ for all nuclides.

Table 1

Radionuclide inventories of ROX and MOX spent fuels (atom No. $\times 10^{24}$ per an equivalent of 1 MTU) 1000 yr later discharged from PWR, Zr/U: zirconia type, Zr/Th: thoria type, W: weapons-grade Pu, R: reactor-grade Pu

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Nuclide	Zr/U-W	Zr/U-R	Zr/Th-W	Zr/Th-R	MOX-W	MOX-R	
Pu-240	1.341×10^{1}	2.352×10^{1}	1.156×10^{1}	2.126×10^{1}	1.445×10^{1}	3.059×10^{1}	
U-236	1.549	2.674	1.308	2.387	2.048	3.787	
Th-232	2.417×10^{-5}	4.110×10^{-5}	5.404×10^{2}	4.506×10^{2}	3.727×10^{-5}	6.261×10^{-5}	
Am-241	2.724	4.145	2.485	3.876	2.379	4.705	
Np-237	1.039×10^{1}	1.582×10^{1}	9.460	1.477×10^{1}	9.272	1.815×10^{1}	
U-233	1.473×10^{-8}	8.641×10^{-7}	1.276×10^{1}	1.188×10^{1}	2.127×10^{-8}	$7.945 imes 10^{-7}$	
Pu-242	3.346	1.189×10^{1}	3.236	1.188×10^{1}	2.692	1.118×10^{1}	
U-238	2.161×10^{2}	2.158×10^{2}	6.014×10^{-3}	2.202×10^{-2}	2.061×10^{3}	2.000×10^{3}	
U-234	4.427×10^{-1}	3.104	1.864	4.350	3.436×10^{-1}	3.002	
Am-243	8.312×10^{-1}	3.154	$8.147 imes 10^{-1}$	3.163	5.803×10^{-1}	2.581	
Pu-239	2.363×10^{1}	2.267×10^{1}	1.598×10^{1}	1.422×10^{1}	3.325×10^{1}	5.074×10^{1}	
U-235	8.996×10^{-1}	$8.911 imes 10^{-1}$	$7.710 imes 10^{-1}$	6.922×10^{-1}	2.883	3.802	

4.3. Geochemistry

Groundwaters in deep geologic formations are normally reducing, and are in a state of chemical equilibrium with reactive solids in the geological formation. The chemical equilibria governing the solubility and the adsorption behavior of radionuclides are controlled by the characteristics mentioned above, which are highly site specific in nature. In order to obtain the solubility of a specific element in a specific geological environment, it is necessary to use a geochemical computer code such as EQ3/6 [13]. As mentioned before, taking into account of the lack of site specific scientific basis concerned upon which computations are implemented, we used solubility data reported by NAGRA [14] and conservative distribution coefficient data cited from SKI [15] report, as shown in Table 2. In this table, realistic and conservative values of solubility are estimated by Berner [16], and are used for the investigation of the effect of parameter uncertainty. Adair et al. [17] estimated very low Zr solubilities $(10^{-11}-10^{-9} \text{ mol } 1^{-1})$ at pH 7–9. In the present paper, realistic value of zirconia solubility is assumed to be 10⁻¹⁰ mol 1⁻¹ taking account of the solubility mentioned by Degueldre et al. [18], and conservative value of that is 10^{-9} mol 1^{-1} .

4.4. Hydrology

This report assumes hydraulic conditions of the typical geologic setup in Japan. However, very little is known about the hydrology of deep geologic systems. In view of the lack of information and also the uncertainties about present and future hydrology, the approach used in this analysis is to perform parameter variations to account for the variability of major model hydrological parameters. The hydraulic conductivity and porosity of granite are shown in Table 3 [19]. Although the hydraulic gradient depends on a specific hydrological system, we assume here a value of 0.01 (taking account the value of 0.005 used by SKI [20]), because the repository will be constructed in a region of low hydraulic gradient. The groundwater flow velocity is correspond-

ingly in the range 6×10^{-5} –0.1 m yr⁻¹ in the rock mass, and in the range 0.1–3 m yr⁻¹ in the fractured zones regardless of local and regional fractures. This analysis uses 0.1 and 1 m yr⁻¹ as the conservative flow velocity in the rock mass and the fractured zones, respectively. The migration length of radionuclides from the repository to the biosphere depends on site specific geological situations. This study arbitrarily assumes that the migration length in the homogeneous rock mass and fracture zone is 300 and 1000 m respectively.

4.5. Biosphere

We assume here that the contaminated groundwater migrates through the geosphere and finally enters into an adjacent aquifer which is defined as one of the compartments involved in the biosphere. Water in the aquifer is assumed to be consumed by a local individual as drinking water (0.73 m³ yr⁻¹) with a dilution factor of 10^{-3} , which can be estimated by the ratio of flow rate (Darcy velocity) in the homogeneous rock (3×10^{-4} m yr⁻¹) to that in the aquifer (3 m yr^{-1} [21]). Possible other exposure pathways to the individual are neglected in this analysis, because it was evident through a preliminary analysis that the pathway used here gives the highest radiological consequence among all selected pathways.

5. Results and discussion

Individual dose equivalents due to the geologic disposal of the ROX and MOX spent fuels are shown in Fig. 2 (for the 4n decay chain), Fig. 3 (for the 4n + 1 decay chain), Fig. 4 (for 4n + 2 decay chain), and Fig. 5 (for the 4n + 3 decay chain). In these figures, only radionuclides doses from actinide decay chains are estimated. In the conservative solubility case, the peak dose equivalent due to the MOX spent fuels is 1.1×10^{-9} Sv yr⁻¹ at 1.8×10^{6} yr after disposal, which is contributed from ²³¹Pa (MOX reactor-grade Pu). Whereas that due to the ROX spent fuels is 2.0×10^{-11} Sv yr⁻¹ at 9×10^{6} yr after disposal, which is contributed from ²²⁹Th (ROX

Table 2

Elemental solubility limits for reducing conditions and distribution coefficients (K_d) for buffer material and granite

Element	Solubility limit (mol l ⁻¹)		$K_{\rm d} \ ({\rm m}^3 \ {\rm kg}^{-1}) \ ({\rm conservative})$			
	Realistic value	Conservative value	Buffer material (reducing condition)	Homogeneous rock (reducing condition)	Fractured zone (oxidising condition)	
Pa	_	-	_	0.01	0.01	
Th	5×10^{-9}	1×10^{-7}	0.002	0.01	0.01	
U	1×10^{-7}	7×10^{-5}	0.1	0.01	0.002	
Np	$1 imes 10^{-10}$	1×10^{-8}	0.1	0.1	0.001	
Pu	$1 imes 10^{-8}$	1×10^{-6}	0.1	0.5	1	
Am	1×10^{-5}	1×10^{-5}	0.25	0.5	0.5	
$Zr(ZrO_2)$	1×10^{-10}	1×10^{-9}	-	-	-	

Table 333Hydraulic conductivity and porosity of granite in Japan

Rock type	Hydraulic conductivity (ms ⁻¹)	Porosity (-)	
Fractured zone	$10^{-7} - 10^{-6}$		
Rock mass	$10^{-12} - 10^{-9}$	0.003-0.05	
(homogeneous rock)			

reactor-grade Pu). The peak dose equivalent from ROX spent fuels is less than that from MOX spent fuels by ca. 2 orders of magnitude. The peak value of 10^{-9} Sv yr⁻¹ per an equivalent of 1 MTU is not negligible, taking account of possible amount of disposed spent fuels which is order of 10⁴ MTU. In the realistic solubility case, the peak dose equivalent due to the MOX spent fuels is 5.8×10^{-11} Sv yr⁻¹ at 3×10^7 yr after disposal, which is contributed from the same radionuclide as conservative case. On the other hand, that due to the ROX spent fuels is 1.9×10^{-12} Sv yr⁻¹ at 9×10^{6} yr after disposal, which is also contributed from the same radionuclide as conservative case. The peak dose equivalent from ROX spent fuels is less than that from MOX spent fuels by one or more orders of magnitude. However, the peak dose equivalent of ²²⁶Ra from MOX spent fuels is only higher than that from ROX spent fuels by a factor of 3, reflecting low solubility of uranium. If it is experimentally certified that the solubility of zirconia is very low (less than 10^{-10} mol 1^{-1}) in the waste disposal condition, we can conclude that the ROX plutoniumburning system is superior than the plutonium recycle system using MOX fuels in the environmental safety for α -nuclides.

The dose equivalents from reactor-grade Pu are larger than those from weapons-grade Pu, in any case, owing to the increase of amount of transuranic elements. Comparing the thoria type fuels with zirconia type fuels, the dose equivalent contributed from ²³²Th is increasing in the thoria type fuels, but does not give a dominant radiological effect. On the contrary, the dose equivalents from radionuclides of 4n + 3 decay chain are slightly increasing in the zirconia type fuels due to the increase of ²³⁹Pu inventory in the zirconia type fuels.

6. Concluding remarks

The comparative study of environmental safety was carried out for the direct disposal of ROX and MOX spent fuels assuming the generic geological condition of Japan. The committed effective dose equivalents of actinide decay nuclides due to the exposure pathway ingesting drinking water were calculated using the conservative and realistic values of solubility. The conclusions obtained are as follows:

• The committed dose equivalents from ROX spent fuels are less than those from MOX spent fuels by ca. 2 orders of magnitude in the conservative solubility case, and are less than by one or more orders of magnitude in the realistic solubility case. It is suggested that the ROX plutonium-burning system will be beneficial to repository safety. In order to confirm the



Fig. 2. Individual dose from 4n decay chain of disposed ROX and MOX spent fuels. Zr/U: zirconia type, Zr/Th: thoria type, W: weapons-grade Pu, R: reactor-grade Pu.



Fig. 3. Individual dose from 4n + 1 decay chain of disposed ROX and MOX spent fuels. Zr/U: zirconia type, Zr/Th: thoria type, R: reactor-grade Pu.



Fig. 4. Individual dose from 4n + 2 decay chain of disposed ROX and MOX spent fuels. Zr/U: zirconia type, Zr/Th: thoria type, R: reactor-grade Pu.

safety of geologic disposal of ROX spent fuels, it is recommended that the solubility of zirconia should be estimated by experimental studies.

 The most important nuclide is ²³¹Pa for MOX spent fuels, and the dose equivalents due to ²³¹Pa is 1.1 × 10⁻⁹ Sv yr⁻¹ (MOX reactor-grade Pu, conservative solubility case) per an equivalent of 1 MTU. For ROX spent fuels, the most important nuclide is ²²⁹Th, and the dose equivalents due to ²²⁹Th is 2.0×10^{-11} Sv yr⁻¹ (ROX reactor-grade Pu, conservative solubility case).

- The dose equivalents from reactor-grade Pu are larger than those from weapons-grade Pu owing to the larger amount of transuranic elements.
- Comparing the thoria type fuels with zirconia type fuels, the dose equivalents from the zirconia



Fig. 5. Individual dose from 4n + 3 decay chain of disposed ROX and MOX spent fuels. Zr/U: zirconia type, Zr/Th: thoria type, R: reactor-grade Pu.

type fuels are slightly larger than those from the thoria type fuels due to the increase of 239 Pu inventory.

 When crack data about spent nuclear fuel pellets of ROX are available, the committed dose equivalents due to fission and activated products (¹²⁹I, ¹³⁵Cs, ³⁶Cl, etc.) will be estimated. Sensitivity analyses are also to be done in the future.

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