Proper treatment of wastes contaminated by radioactive substances (summary of our technical report)

This report explains our research in ways that are easy to understand.



National Institute for Environmental Studies (NIES) Center for Material Cycles and Waste Management Research (CMW)

Introduction

This paper is a summary of "Proper treatment of wastes contaminated by radioactive substances (Technical report)". The purpose of the summary is to explain the procedures as clearly as possible.

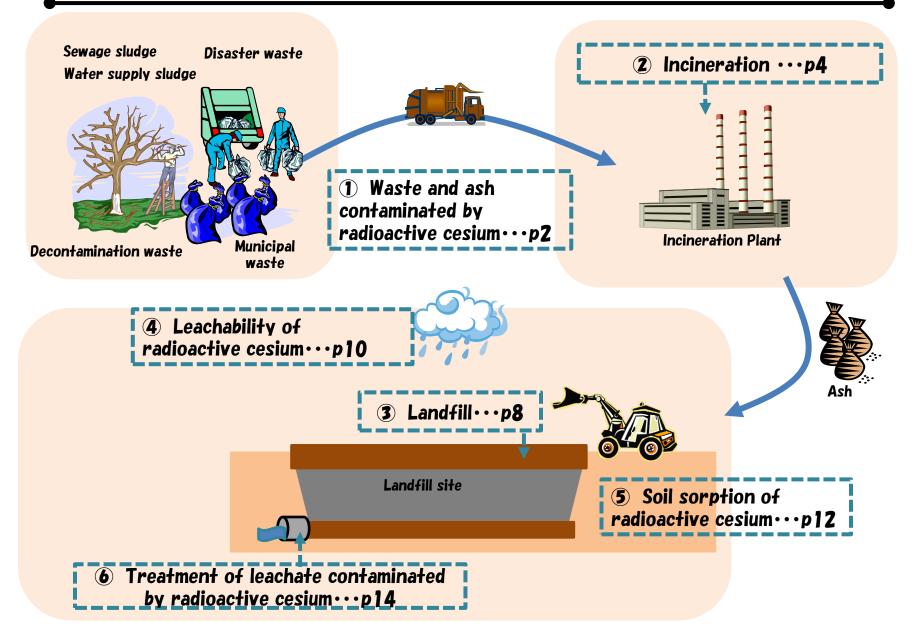
"Proper treatment of wastes contaminated by radioactive substances (Technical report)" can be downloaded from the following sites:

NIES Website Great East Japan Earthquake Information Page

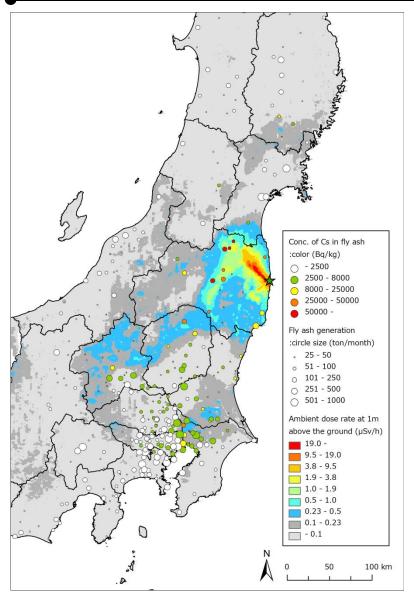
Proper treatment of wastes contaminated by radioactive substances (Technical report) Second edition <Japanese language> http://www.nies.go.jp/shinsai/techrepo_r2_120326s.pdf

Chapter 10 (revision) < Japanese language > http://www.nies.go.jp/shinsai/techrepo_r2_s10+_120416.pdf

Proper treatment of wastes contaminated by radioactive substances (Table of Contents)



Waste and ash contaminated by radioactive cesium(Cs) \$



Generation of fly ash^{*1} contaminated by radioactive Cs

After the accident at The Fukushima Daiichi Nuclear Power Plant, incineration residues in eastern Japan contained radioactive cesium.

This is because vegetation, fallen leaves, and attached soil that would be contaminated by radioactive fallout were collected and burned in incineration plants.

The figure on the left shows fly ash generation from municipal solid waste (MSW) incinerators and the activity of radioactive Cs (July 2011) on the map of air dose rate^{$\times 2$} (November 2011). The green star on the map indicates the Fukushima Daiichi nuclear power plant. The map colors and circle sizes indicate the following:

Color of circle	• • •	Cs activity (Bq/kg) in fly ash
Size of circle	•••	Fly ash generation from MSW incinerator
		(ton/month)
Color of map	•••	Air dose rate(µSv/hour)

The following concerns are apparent from the figure on the left: The Cs activity in fly ash is over 8000 Bq/kg at plants located in areas where the air dose rate is high.

Even in the Kanto area, the Cs activities are more than 8000 Bq/kg, as indicated by yellow and orange circles. Furthermore, Cs activities are several thousand Bq/kg at other sites, as indicated by the large green circles. Appropriate disposal of this ash is mandatory.

The Cs activity in fly ash has been declining with time. However, the activity might increase again if decontamination generates large amounts of vegetative waste.

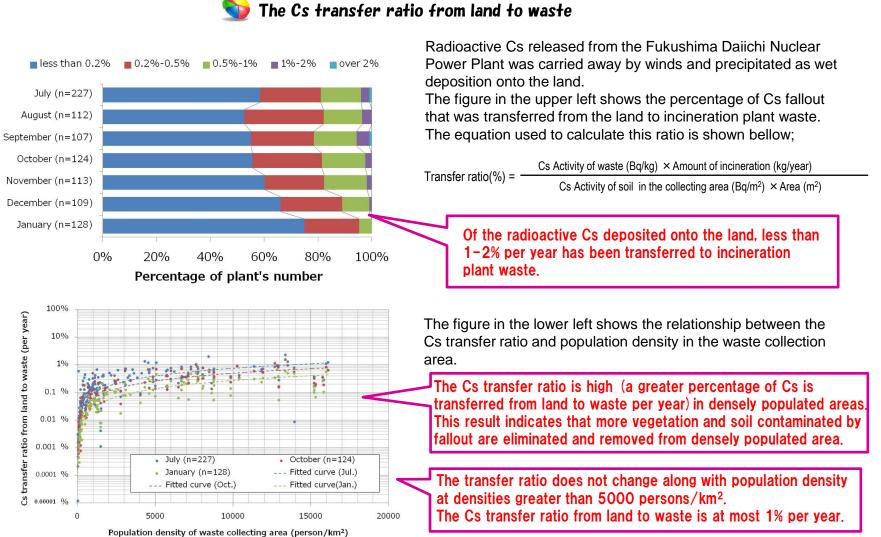
*X*1 There are two types of incineration ash: one is the bottom ash that remains at the bottom of a incinerator; the other is the fly ash, which is the fine ash particles emitted with the stack gases. Radioactive Cs tends to be concentrated in the fly ash. *X*2 Air dose rate is the radiation dosage per unit time of space.

For more information, please read the next page.!

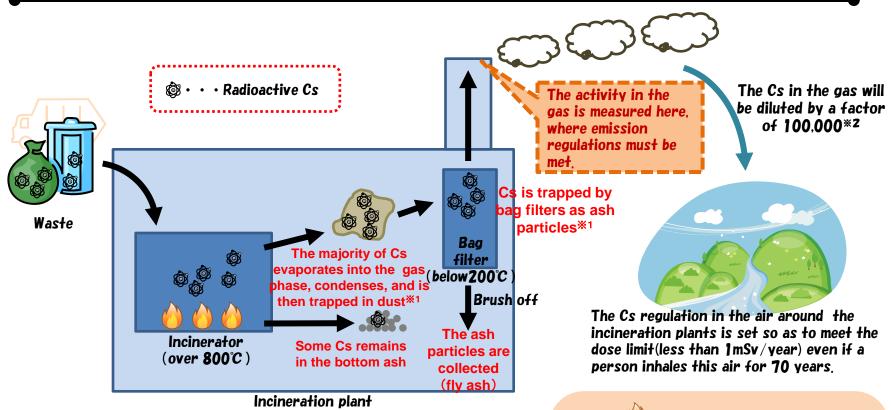
(1) Waste and ash

contaminated radioactive Cs

To obtain more information about the waste and ash contaminated by radioactive Cs, the NIES collected and analyzed data from various incineration plants in 16 prefectures (Iwate, Miyagi, Akita, Yamagata, Fukushima, Ibaragi, Tochigi, Gunma, Saitama, Chiba, Tokyo, Kanagawa, Niigata, Yamanashi, Nagano and Shizuoka). Some of the results are summarized bellow.



2 Incineration Part 1



Behavior of radioactive Cs in the incineration plant

•The waste contaminated by radioactive Cs is burned at a temperature greater than 800°C in the incinerator. The majority of the Cs is volatilized or liquefied and is transported with the stack gases.

•Some Cs remains at the bottom of the incinerator.

•The chemical form of radioactive Cs in the gas will be mainly cesium chloride (CsCl), which is trapped in dust^{%1}.

•Radioactive Cs is trapped in dust^{%1} by a cylindrical filter called a bag filter that is cooled down to a temperature below 200°C.

%1 The dust consists of fine, solid particles in the exhaust gas.

%2 This number is taken from a scenario assessment by the Ministry of the Environment . It is variable and depends on the factors such as weather and stack height.

Cs regulation in gas

•The Cs regulation in the air is set so as to meet the dose limit(under 1mSv/year) even if a person inhales this air for 70 years...

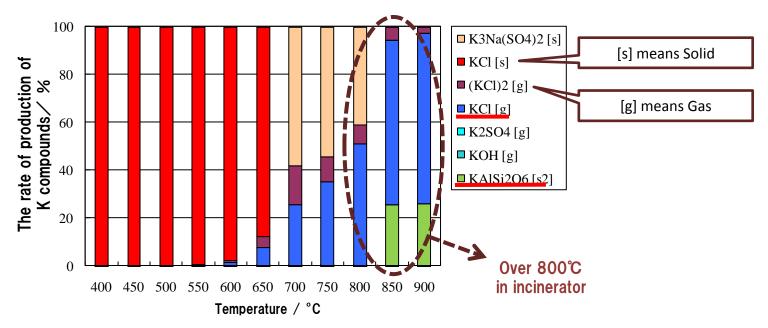
•This Cs regulation must be met at the stack outlet.

•In fact, Cs emitted from the stack is **diluted** by a factor of 100,000^{%2}.

For more information about Cs's behavior, please read the next page.

To elucidate details of Cs behavior in incinerators, the NIES performed a theoretical calculation that assumed a condition of thermodynamic equilibrium. Some of the results are summarized bellow.

In this calculation, cesium (Cs) was replaced by potassium (K), a congener of Cs, because the database for Cs was insufficient to carry out the calculation.



Radioactive Cs in fly ash

Potassium chloride (KCl) gas accounts for the greatest proportion of the potassium at temperatures above 800°C in the above figure.

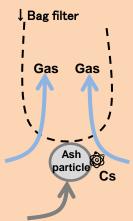
If potassium (K) is replaced by cesium (Cs), KCl would be CsCl (cesium chloride), and CsCl would be the main form of Cs in the fly ash.

Radioactive Cs in bottom ash

the figure shows that KAlSi₂O₆ accounts for the greatest proportion of potassium in the solid phase at temperatures above 800°C. KAlSi₂O₆ is a kind of aluminosilicate mineral. If potassium (K) is replaced by cesium (Cs), KAlSi₂O₆ would be CsAlSi₂O₆, and be CsAlSi₂O₆ would be the main form of Cs in the bottom ash.

2 Incineration Part 2

Function of bag filters



Bag filte

The function of bag filters is to eliminate ash particles from incinerator gas. Bag filters are cylindrical in shape and made of fabric similar to felt. Hundreds of bag filters are used at large incineration plants. As indicated in the figure at left figure, gas can pass through a bag filter. However, most ash particles can not pass through a bag filter because they are larger than the mesh size. Cs is not present in the gas phase at 200°C, which is the temperature in the vicinity of the bag filter.

Cs is trapped as ash particles at $200^{\circ}C_{\circ}$

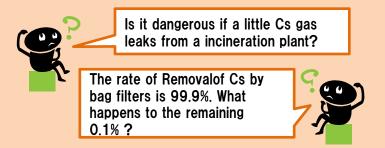
Ash is brushed off bag filters by a "pulse-jet", which eliminates clogs and prevents filters from breaking because of the weight of ash.

Ash on bag filters is brushed off one filter at a time, not every filter at once. In that way there is no interference with filter function.

There are hundreds of bag filters in a large incinerator. Pulse jet treatment is conducted one bag filter at a time.

For more information about Cs's measured result in gas, please read the next page.

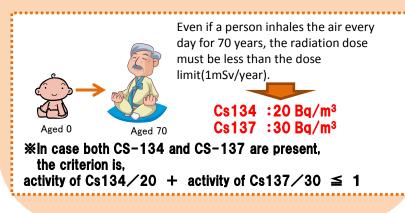
Removal rate of Cs and exposure risk



In previous surveys, radioactive Cs has never been detected under normal measurement conditions at the outlet of stacks from incineration plants that that were using bag filters.

Even if some of the Cs leaks from a stack, it will be diluted by a factor of 100,000 in the air and hence reduced to a level.

Rather than removal of particulate Cs by bag filters, it is important in terms of "exposure risk" that Cs regulations in the gas phase be strictly observed.



X This number is taken from ascenario assessment by the MOE. It is variable and depends on factors such as weather and stack height.

The following table shows the Cs removal rates that were measured for exhaust gas treatment systems of actual incineration plants and waste melting plants where fly ash containing 8000 Bq/kg were generated.

Facility	Facility Thermal process	Inlet conc. (Bq/m ³)		Outlet conc. (Bq/m ³)		Removal rate (%)		Fly ash
Facility		¹³⁴ Cs	¹³⁷ Cs	¹³⁴ Cs	¹³⁷ Cs	¹³⁴ Cs	¹³⁷ Cs	collector
	A Incineration	78	96	< 0.008	< 0.006	>99.99	>99.99	Bag filter
A		98	126	0.008	< 0.007	99.99	>99.99	
		33	42	0.2	0.2	99.39	99.52	Electric
В	Incineration	43	57	0.2	0.2	99.53	99.65	
C	Incineration	58	70	< 0.054	<0.053	>99.91	>99.92	Bag filter
D	Incineration	58	76	< 0.1	<0.1	>99.83	>99.87	Bag filter
D	Electric melting	677	844	<0.1	<0.1	>99.99	>99.99	

Melting process:

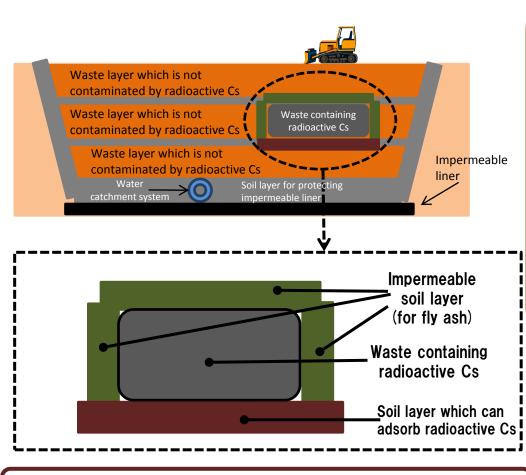
- 1. Incinerated residue and fly ash are heated at a high temperature until they become liquids.
- 2. After heating, the ash cools to become slag and fly ash.

The value of every measurement is far below the Cs regulation explained on the preceding page.



Landfill for waste contaminated by radioactive Cs (less than 8000 Bg/Kg)

Special measures are required to dispose of ash sludge and other wastes contaminated by radioactive. It is acceptable to dispose of the Cs in waste in an existing landfill if the activity of Cs is less than 8000 Bq/kg. However, attention must be paid for to the following points.



For more information about impact assessment after landfill, please read the next page.!

Layer under the waste A 50cm- thick- layer of soil that can absorb radioactive substances must be in place under the landfilled waste

Upper and lateral laver

If the Cs is in a soluble form such as the Cs in fly ash, impermeable soil layers must be in place above and on all sides of the landfilled waste. These layers exclude water and prevent

groundwater from being contaminated by radioactive Cs.

Treatment of leachate

An impermeable liner is enplaced at the lowest part of the landfill. Leachate from waste is collected by drainage from the impermeable liner and must be removed in a proper way before beind discharged. However, radioactive Cs can not be removed by generic treatment systems. Additional processes like absorption and membrane filtration are needed for eliminating Cs

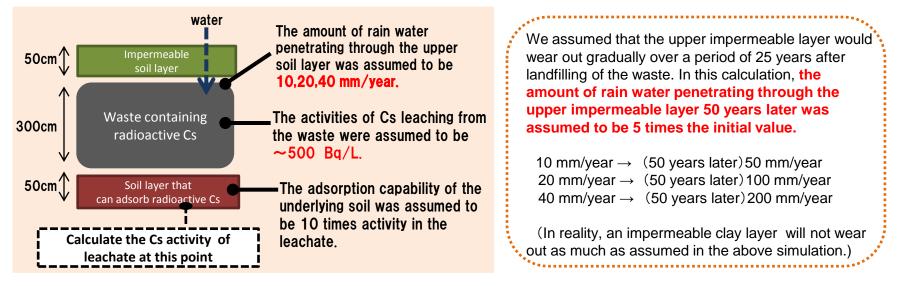
Multiple protective plans for landfill : •Impermeable overlayer for keeping out water •Underlying soil layer for absorbing Cs Additional processes at leachate treatment system

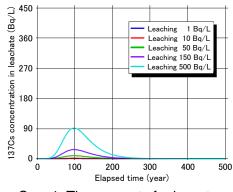
It is also important to check the treated water and groundwater regularly.

3 Landfill



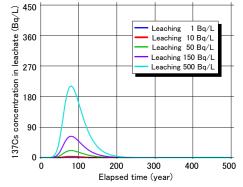
The NIES measures the Cs activity in leachate from landfilled waste containing radioactive Cs in the way explained on the preceding page. The amount of Cs that leaches from landfilled waste depends on the amount of Cs in the waste, the extent of water with the waste, leachability based on the type of waste and so on. The following conditions were used in our assessment.



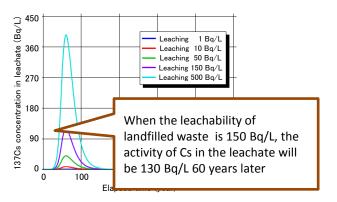


Case1: The amount of rain water penetrating through the upper layer is $10 \text{mm} \rightarrow 50 \text{mm/year}$

Some result of calculation are shown below:



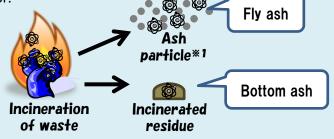
Case2: The amount of rain water penetrating through the upper layer is $20 \text{mm} \rightarrow 100 \text{mm/year}$



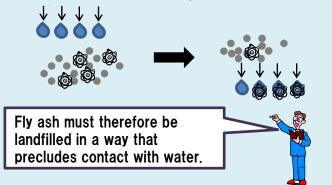
Case3: The amount of rain water penetrating through the upper layer is $40 \text{mm} \rightarrow 200 \text{mm/year}$

Municipal Solid Waste Incinerator Ash (fly ash and bottom ash)

There are two types of ash. One is fly ash which consists of dust and substances volatilized during incineration. Another is bottom ash, which is discharged from the bottom of the incinerator.



■Leachability of radioactive Cs from fly ash Radioactive Cs in fly ash is very soluble in water. In our experiments^{※2}, roughly 64-89% of radioactive Cs leached out from fly ash into water during a contact time of 6 hours.



Bottom ash

Unlike fly ash, the solubility of radioactive Cs in bottom ash is low. In our experiments^{%2}, less than **5.6%** of radioactive Cs in bottom ash leached into water.

Sewage Sludge Ash

When sewage water is treated, solid waste called sewage sludge is generated. The incineration of sewage sludge results in generation of sewage sludge ash. Radioactive Cs is also concentrated in the ash. In our experiments^{%2}, the amount of radioactive Cs leached from sewage sludge ash was **below the detection limit.**

Sludge from Waterworks

Sludge is generated in the process of purifying raw water to be distributed as tap water. The main components of the sludge are sedimented chemicals that are added for water purification, particles of sand, and so on. In our experiments, the amount of radioactive Cs leached from sludge generated by waterworks was **below the limit of detection**.



In our experiments, the amount of radioactive Cs leached from soil was **below the limit of detection.**



Disaster Waste (incombustible waste)

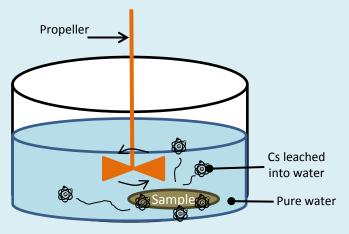
In our experiments, radioactive Cs leached from incombustible disaster waste (demolished concrete, tile, timber, etc.) was very low. In the case of crushed PVC pipe, the leaching ratio was 5.0%

> Except for fly ash, radioactive Cs in waste is not very soluble.

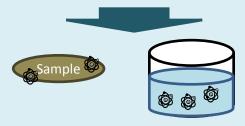
%2 For more information on leaching test, please see the next page.!



The NIES used a leaching test to determine the leachability of radioactive Cs from various types of waste. The applied test method is the Japanese Industrial Standard (JIS) K0058-1, shown schematically below.



First, the sample is placed in a plastic container with an amount of pure water (equal to ten times the volume/weight of the sample). Then, the water is stirred for 6 hours.



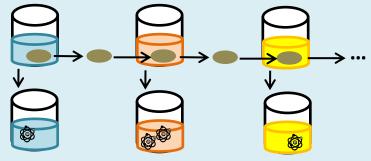
The sample and water are separated by filtration and the amount of radioactive Cs leached into the solution is determine.

XThe results are shown on the previous page

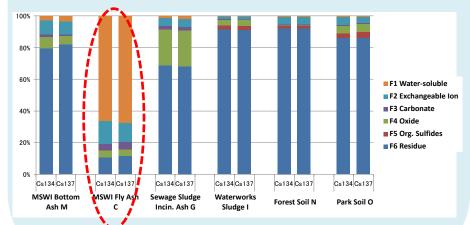
The NIES also used several other leaching tests with different conditions to understand the leaching properties of radioactive Cs. One example is shown below.

"Sequential extraction test"

A sample was sequentially put into several kinds of solvents to partition the radioactive Cs with respect to leachability in those solvents. From the results, we can estimate the chemical form of radioactive Cs in the sample.



The result is shown in the following figure.

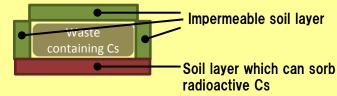


As mentioned on the previous page, only the municipal solid waste incinerator fly ash contains an abundant water-soluble fraction.



📆 The function of soil

When waste containing soluble radioactive Cs is landfilled, an impermeable soil layer must be enplaced around the perimeter and above the waste. In addition, a soil layer that can sorb radioactive Cs must be enplaced under the landfilled waste to prevent Cs from being released to the surrounding environment via leachate.



The survey of the Chernobyl accident revealed that much radioactive Cs was sorbed to soil after the accident. If radioactive Cs can be sequestered by soil, its activity will decrease without contaminating the surrounding environment.

Sorptive property of various soils

The NIES conducted sorption tests^{*} with various types of soil and adsorbents. The results are shown below.



Powdered mordenite (a kind of zeolite)

Granular mordenite (a kind of zeolite)

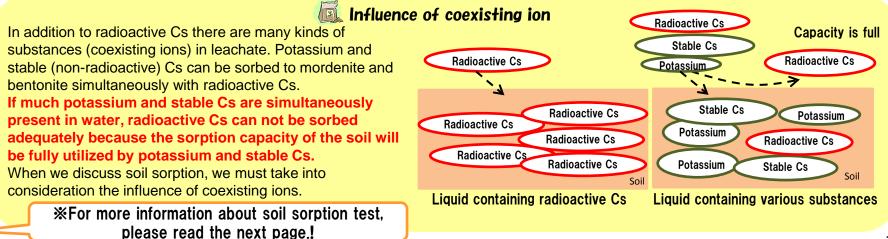
Bentonite (a kind of clay mineral)

4th Cohesive soil collected in Saitama prefecture

5th Decomposed granite soil collected in Ibaragi prefecture

6th Silica sand

A high capacity for sorption of Cs and a certain degree of permeability will be needed to obtain sufficient capacity in the soil layer. If proper soil is unavailable, a suitable material can be designed by mixing several soils and adsorbents.



(5) Soil sorption of radioactive cesium

The NIES conducted soil sorption tests to determine the capacity of soils to adsorb radioactive Cs. The following samples were used in the test.





Silica sand (uniform particle size=0.4~0.6 mm)



Bentonite (low permeable material)



Decomposed granite soil collected in (Cohesive soil collected in Ibaragi prefecture



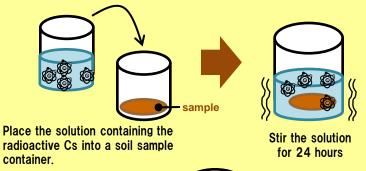
Granular mordenite (particle size $=1.4 \sim 4.0 \text{ mm}$



Landfill cover soil

Saitama prefecture)

(particle size : under 0.2 mm) The protocol for the soil sorption test is shown below.

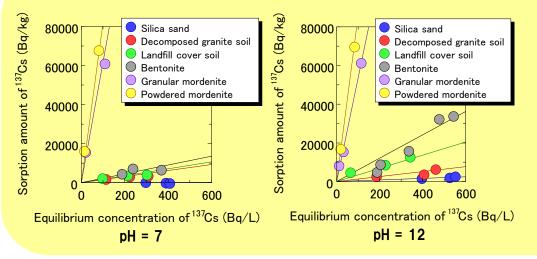






Separate the sample and water by filtration and measure the amount of radioactive Cs in the water to determine how much Cs adsorbed to the soil sample.

Some of the results are shown below.

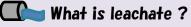


The two figures on the left show the results of a soil sorption test with Cs 137. The solution in the figure on the left is neutral (pH=7), the solution in the figure on the right is alkaline (pH=12).

The horizontal axes of both figures are the mean equilibrium activities of Cs 137, and vertical axes are the mean adsorbed amount of Cs 137. The steeper the slope of the relationship, the higher the sorptive capacity of the sample material.

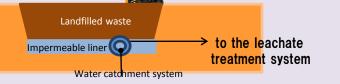
It is apparent that the sorptive properties of powdered and granular mordenite are very high.

6 Treatment of leachate contaminated by radioactive cesium



An impermeable liner is placed immediately below a landfill site to intercept water that would otherwise leak from the landfill into groundwater.

Leachate is the water that penetrates landfill, contacts waste within the landfill, and is collected by drainage conduits on the impermeable liner.





After the leachate is treated, it is discharged to a receiving body of water such as a river, lake or sea. When treated water is discharged, the Cs activity must be less than the regulatory limit for Cs activity in leachate.

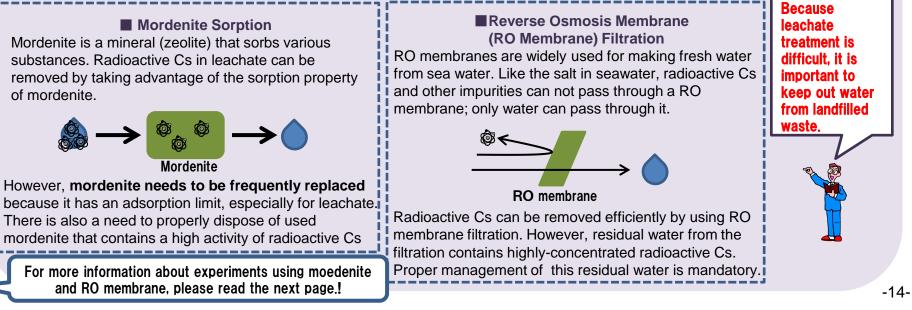


Even if a person drinks the water everyday for 70 years, the radiation dose will be under the dose limit(1mSv/year).

 $\rightarrow Cs 134 : 60 Bq/L$ Cs 137 : 90 Bq/L

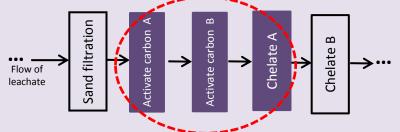
Treatment method for leachate contaminated by radioactive Cs

Various treatment processes are incorporated into leachate treatment systems. However, radioactive Cs can not be removed by generic treatment systems. Additional treatment processes for removing radioactive Cs must be installed. the following two treatment methods are currently available for Cs removal.



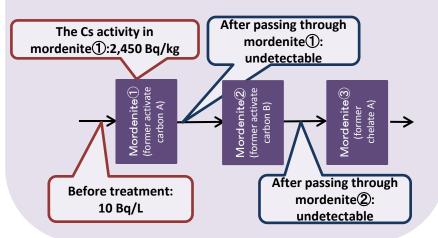
Leachate treatment test

The NIES conducted leachate treatment tests with a real treatment system at a landfill to determine the efficiency of treatment methods using mordenite. The Cs activity in the leachate that was generated at the landfill where we conducted the tests was at most 31 Bq/L, below the regulatory limit for Cs activity in leachate. In our test, some of the filtering materials (activated carbon A, activated carbon B and chelate A) were replaced by mordenite as indicated below.



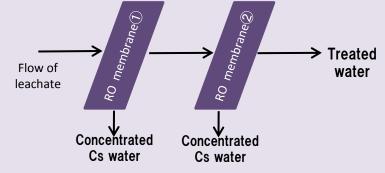
Mordenite was placed into these three filtration unit and leachate was treated for 24 hours

The result is shown below.

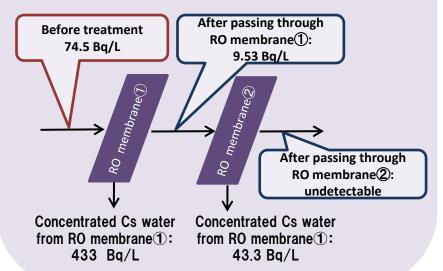


The NIES conducted leachate treatment tests using a RO membrane in a real treatment system at a landfill. The RO membrane used in the tests was the same as the membrane used for desalinating seawater.

In our tests, two RO membrane filtration steps were installed as indicated below.



The result is shown below.

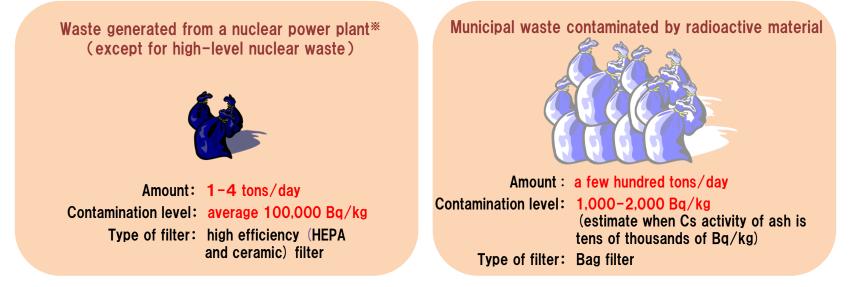


Appendix

Difference of incineration technology between waste from nuclear power plant and municipal waste

The combustible waste generated by a nuclear power plant is incinerated in a special incinerator that is designed for highly- contaminated waste from radioactive materials. The incinerator has high - efficiency filters to remove radioactive materials.

If municipal waste contains radioactive materials, should such filters be installed on municipal waste incinerator, too? In fact, the amount and activity of each kind of waste are quite different.



The contamination level of municipal waste is much lower than that of waste generated by a nuclear power plant. Therefore radioactive materials in the stack gases from an incinerator that burns municipal waste can be removed without a high - efficiency filter.

However, bag filters for municipal waste might not be sufficient if the highly contaminated waste near the Fukushima Daiichi Nuclear Power Plant is going to be incinerated. In such cases, use of high- efficiency filters or double bag filters must be considered.



Landfill for waste contaminated by radioactive Cs (8000-100,000 Bq/kg)

If waste contains 8,000 - 10,000 Bq/kg of radioactivity, strict measures should be taken for landfilled waste.

Basic principles for landfilling of radioactively contaminated waste are the same if the activity is less than 8000 Bq/kg.

- Keep water from landfilled waste
- Emplace soil layer for absorbing Cs under the waste
- Remove Cs in leachate appropriately

Landfill for waste contaminated by radioactive Cs (greater than 100,000 Bq/kg)

If waste contains more than 100,000 Bq/kg of radioactivity, it can be landfilled at an isolated site if the waste is contained within a thick concrete structure.

The "low-level radioactive waste" generated by a nuclear power plant ,with an activity greater than 100,000 Bq/kg, is emplaced in shallow (<10 m) underground concrete pits. The activity must be less than 100 billion Bq/kg for disposal. Although this waste is called "low-level radioactive waste", it includes some waste that is highly radioactive compared to the contaminated waste generated by the Fukushima Daiichi Nuclear Power Plant accident.

Appendix : Basic information about radioactive substances



Types and characteristics

	Types	Characteristics					
	$\alpha(alpha) particle radiation$	An alpha particle can travel only a few centimeters in the air and can be stopped by a piece of paper. Whe it enters the body, it causes localized but intense damage to cells along its path.					
	B (beta) particle radiation	A beta particle can be stopped by aluminum foil or plastic a few centimeters thick.					
	Г(Gamma)ray X ray	These forms of electromagnetic radiation have strong penetrating power and can be stopped by sheet of lead or concrete about 10 centimeters thick.					
	Neutron radiation	A neutron has strong penetrating power. Water or paraffin can slow neutrons.					
Γ	Cesium 137 (half life: 30 years) and strontium 90 (half life: 29 years),						

Cesium 137 (half life: 30 years) and strontium 90 (half life: 29 years) which were released as a result of the Fukushima Daiich Nuclear Power Plant accident, emit beta particles and gamma ray.s



🕅 Units of radiation



Becquerel (Bq) A becquerel is a unit of radioactivity equal to one disintegration per second. Sievert (Sv) A sievert is a measure of the impact of radiation on the human body. The impact is variable and depends on the nature of the radiation and the parts of the body that are exposed.



There are two types of health effects.



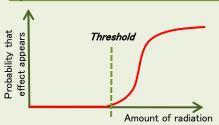
■ Physical effects Direct effects on a person exposed to radiation. **Genetic effects** Effects on future generation.

There are two types of physical effects.

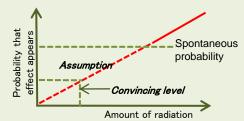
• Acute damage (vomiting, diarrhea, fever, depilation, bleeding) These effects appear when a person is briefly exposed to a large amount of radiation.

•Chronic damage (cancer, shortening of life, cataracts)

These effects appear when a person is exposed to small amounts of radiation for a long time. They are associated with a long latency period.



Deterministic effect Acute and chronic damage, with the exception of cancer and genetic effects, are associated with a threshold above which adverse effects of the radiation on health begin to appear. These effects can be avoided by lowering the level of radiation exposure.



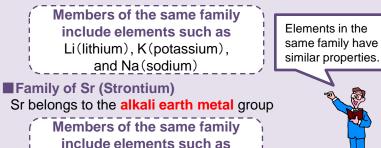
■ Probabilistic effect There is no threshold associated with carcinogenesis and genetic effects. Lowering the degree of radiation exposure reduces the probability that these effects will occur.

Appendix : Characteristics of radioactive substances 🎘 🕹





Family of Cs (Cesium) Cs belongs to the alkali metal group

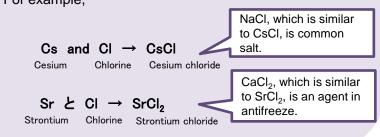


Mg(magnesium)

and Ca(calcium)



In the environment, alkali metal and alkali earth metals bind to other atoms rather than being alone. For example,



% What is Vapor pressure?

Vapor pressure is a measure of the tendency of a substance to change from a liquid or a solid to a gas.

Vapor pressure is low · · · a substance is likely to be a liquid or a solid.

Vapor pressure is high · · · a substance is likely to change from a liquid or a solid to a gas.



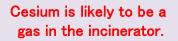
Temperature and other conditions that affect Cs

■ High temperature (inside incinerator: over 800°C)

•Boiling point (temperature associated with liquid to gas transition) of Cs is 690°C.

•Boiling point of cesium chloride is 1300°C.

•Vapor pressure^{*} is very high.



Low temperature

(stack gases from incineration plant : 150-170°C)

The NIES calculated the vapor pressure of cesium chloride at 150-170°C. Cesium chloride is considered to be the main form of cesium in the incinerator.

When temperature is150°C,

vapor pressure is 0.0000000000275 pascal (←unit of pressure) When temperature is 170°C,

vapor pressure is 0.000000000337 pascal **VERY LOW!!**

Cesium is likely to be adsorbed to solid substances like ash rather than being a gas in the stack gases that exit the incineration plant.

In previous surveys, radioactive Cs has never been detected in the stack gases from incineration plants that had bag filters.

