

Dispersion Concentration of Tritium in Rainwater and from Wet Deposition, released during the Venting of

Tritium Waste Containers with Flange at TA-54 of LANL

Client:Communities for Clean Water (CCW)
New MexicoContractor:Ingenieurbüro Rau
Bottwarbahnstraße 4
D-74081 HeilbronnIn collaboration with:Dr. Winkler
Ingenieurbüro Winkler
Wilhelm-Gülpen-Straße 29
D-52146 Würselen

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1 Task

Los Alamos National Laboratory (LANL) applied for preauthorization to begin construction and operations to vent the Flanged Tritium Waste Containers (FTWCs) at the TA-54 site, building 1028. The FTWC headspace gas contains hydrogen and oxygen, accompanied by radioactive tritium which will be vented along with the headspace gases. The tritium may be in the form of water vapor or as elemental hydrogen gas. There are overall four FTWCs with significant tritium inventory.

The FTWCs at TA-54 contain tritium-contaminated metal parts and molecular sieve media, which is a pebble-like material used to absorb water vapor from exhaust air streams. This molecular sieve media inside the FTWCs is contained in metal canisters, along with some loose media material in bags. Over time, tritiated water vapor that had been adsorbed onto the media can become liberated into the FTWC headspace. Radiolysis can cause separation of the water vapor, possibly resulting in a hazardous hydrogen-oxygen mixture within the FTWC. The Applicants have determined that continued tritium storage in these containers could pose an unsafe condition. To mitigate this hazard, the FTWCs would be vented in-place to remove hazardous gases.

The engineering company M. Rau was contracted to conduct a study to determine the dispersion of tritium, its washing out with rain and the resulting ground contamination during the venting of FTWCs.

2 Methodology

The dispersion calculations will be carried out with the German dispersion model ARTM [1] (Lagrangian dispersion model) used for licensing and regulatory purposes concerning long term releases caused by normal operation of nuclear installations.

The model calculates dispersion, dry and wet deposition of gaseous releases from nuclear installations. The results are two-dimensional fields of concentration and contamination and time series of concentration and contamination for given points.

As input, the model requires representative meteorological data for the location and mean average values for the release period. Results are calculated on the basis of typical release rates by venting the FTWCs, as given by the client.

3 Model Description and Required Data

3.1 Model Description

The numerical calculation of ground contamination by wash out for tritium will be carried out with the model-package ARTM, an adoption of the well-known AUSTAL3.2-model [2] for modelling radionuclide transport, deposition and radionuclide doses. AUSTAL3.2 is in compliance with the German Guidance TI Air. The core of AUSTAL3.2 as well as ARTM is a Lagrangian particle model [1].

The Lagrangian particle model differs fundamentally from the majority of the established numerical modelling techniques which are founded on the computed solution of the advectiondiffusion equation. This model tracks point-like particles representing a trace species on their path through the atmosphere. The particles travel with the mean wind and are additionally subjected to the influence of turbulence. The effect of the turbulence is modelled by adding an additional random velocity to the mean motion for the particle. This random velocity, which is derived from a Markov process, is a function of the turbulence intensity and is different for each particle. The concentration distribution is determined by counting the particles in given sampling volumes and expressed as mean values over the volume and time intervals. The main advantages of the Lagrangian particle model are that the model concept largely reflects the natural phenomena involved in turbulent diffusion. It can be applied to any source geometry desired for any temporal behaviour of a spatially variable source. Required meteorological input information includes the fields of the mean wind components, the wind fluctuations and the diffusion coefficients which can be generated by meteorological pre-processors. For time-dependent calculations, the input parameters must be made available as a time series of fields. Furthermore, emission data will be required. The model output is a time sequence of the spatial distribution of the concentration of the emitted species and its transformation products. The calculated time sequences can be used to determine mean values and instantaneous values for different time periods.

The ARTM-model has additional functions compared to AUSTAL3.2, for example

- Determination of the radiological dose caused by cloud radiation (gamma submersion),
- Calculation of ground contamination caused by wet deposition,
- Considering radioactive decay during dispersion

3.2 Model Options for the Calculations

The following options have been used for the calculations:

3.2.1 Study Area

Of interest in the present case is the Tritium concentration at the nearest inhabited location to the FTWSs. The nearest receptor location is La Vista Church, Nazarene, approximately 2.2 km eastsoutheast of the source (FTCW container). Another place of interest is the Rio Grande River, which is about 5.4 km from the release site. A Site plan is shown in Figure 1.

The calculations were not carried out for the entire area. The activity Concentration of Tritium in air and in rainwater at 2250 m and 5000 m from the source were calculated, whereby constant meteorological conditions (wind direction, wind speed and stability) were selected in each case.



Figure 1: Location of TA-54 Building 1028, La Vista Church and Rio Grande River.

3.2.2 Role of Topography

The area around Los Alamos is a plateau with little topographical structure. Especially between the container station and the nearest receptor point, La Vista Church, the terrain can be considered weakly structured. The Rio Grande river valley about 5 km away has cut into this plateau. For the purposes of estimation, the topography is not taken into account for these estimative calculations.

3.2.3 Building Effects, Plume Rise and Effective Release Height

The tritium is released when the containers are ventilated. There are no large plant buildings in the vicinity of the containers that could significantly influence the dispersion. The release process was set up as a volume source with a base area of 10 m x 10 m and a height of 5 m.

For the following reasons, it is justified to dispense without explicit modeling the wind and turbulence field in the near-source area:

- The locations of particular interest are more than 2000 m away from the source.
- The objective of the study is the contamination of the ground with Tritium, due to its washing out by rain. For the calculation of the washout rate an integration over the entire height of the propagation plume is carried out. Therefore the effective height of the release or building effects are less important.

A possible increase of the effective source height by plume rise is not considered.

3.3 Emission Data

According to the customer, in the event of a release, it must be expected that a mixture of water vapor and oxygen will be released to vent the FTWCs.

It is assumed that the release from a container with 32,000 Curie (Ci) tritium (H3) in the form of water (HTO) takes place over the course of 24 hours.

3.4 Meteorological Data

3.4.1 Data Base

Of interest in the present case is the Tritium concentration at the nearest inhabited location to the FTWSs. The nearest receptor location is La Vista Church, Nazarene, approximately 2.2 km south-southeast of the source (FTWC container).

The quality of dispersion calculations depends on the quality of the dispersion model used and the meteorological input parameters. Therefore, the analysis of meteorological data representative for the site is very important for fixing the model options for the calculations. The required parameters are wind speed, wind direction, atmospheric stability, temperature and precipitation. These parameters should be available at the site or near the site in high temporal resolution (1-h-values) for a desired time period.

Technical Area 54 (TA-54), where the FTWCs are located, also has a meteorological measurement tower. In a first step, the following meteorological data were downloaded for a period of 30 years (1993-2022) from a height of 11.5 m (<u>https://weathermachine.lanl.gov/data-requests/</u> (© Copyright Triad National Security, LLC. All Rights Reserved):

- Wind direction
- Precipitation
- Standard Deviation of Wind direction (SDDIR)
- Standard Deviation of horizontal wind speed (SDSPD1)
- Standard Deviation of vertical wind speed (SDW1)
- Wind Speed (SPD1)

The measurement data do not contain any cloud cover data, which, together with the wind speeds and the time of day, would allow the stability classes required for the dispersion calculation to be determined. Following a suggestion of the EPA [3], the standard deviation of the vertical velocity, standardized with the horizontal velocity and multiplied with a conversion factor, is used to determine the standard deviation of the wind height angle σ_e in degrees. For an initial assignment, a range for σ_e is assigned to the Pasquill-Gifford stability categories A to F. The calculation rule will be as follows:

- SDW1 / SPD1 * 180/pi
- σ_e = SDW1 / SPD1 * 180/pi

In a refinement, a fine adjustment is made using the wind speed at 11.5 m height.

3.4.2 Analysis

The long-term (1992-2023) distribution of wind directions for Los Alamos shows the main maximum for winds from the south-southwest and a secondary maximum for winds from the northwest (Figure 2). A drift of the tritium plume in the direction of La Vista Church is to be expected with winds from the west-northwest.



Figure 2: Wind distribution over the period 1992-2023, based on hourly values.

Figure 3 shows the long-term conditions for the hours in which there was precipitation. The distribution differs significantly from the wind distribution for all hours. Precipitation is to be expected most frequently with winds from the south-southwest. Rain with winds from the west-northwest, the critical wind direction for the nearest location, can only be expected about 12% of the time. Further analysis showed that stability classes D (neutral) and E (stable) in particular are to be expected in the event of rain.



Figure 3: Wind distribution over the period 1992-2023, based on hourly values (only hours with precipitation).

The rain intensity during a rain event is of further interest for the calculation of the wash-out. The release takes place over a maximum of 8 hours. Moving 8-hour averages of the rainfall were calculated over the entire measurement period. Figure 4 shows a histogram of 8h-sums. The 95% value was determined, i.e. the rain event that is only exceeded 5% of the time (10.7 mm/8h resp. 1.3 mm/h. Figure 5 shows a histogram of 1h-sums. The 95% value of the maximum 1 h value was determined in the same way (4 mm/h). Based on this analysis, four scenarios were selected for the calculations, namely no rain, 1 mm/h, 5 mm/h and 10 mm/h.

Conclusion:

Rain events lasting 8 hours or longer are quite rare. This can be concluded from the 98% values of the rain intensity for 1 h and 8 h, which only differ by a factor of 3. Rain events with winds from the west-northwest are significantly rarer than rain events with southwesterly winds. In this respect, the results calculated below (chapter 4) for La Vista Church are possible, but do not have such a high probability of occurrence.



Figure 4: Histogram of precipitation, 8-hour running total over the period 1992-2023.



Figure 5: Histogram of precipitation, 1-hour running total over the period 1992-2023.

4 Results from Dispersion Calculation

4.1 Overview

The following cases were investigated:

Stationary dispersion situation for the two dispersion classes D (neutral) and E (stable) with constant wind speed and wind direction as well as four different, in each case constant precipitation intensities. This results in a total of 8 calculation cases with stationary conditions.

Non-stationary dispersion situation for the two dispersion classes D (neutral) and E (stable) with constant wind speed and wind direction as well as two different episodically occurring precipitation intensities. This results in a total of 4 calculation cases with non-stationary conditions.

Concerning the source, the following data are used:

The horizontal extent is assumed to be 10m x 10m and the vertical extent 5m at ground level The source strength is 32000 Ci in 24 hours, which corresponds to 370,000 μ Ci/s.

All results are proportional to the source strength.

It is assumed that the tritium considered here is released and transported in the form of water (HTO). The washout factor for this is $3.5 e^{-5} 1/m^2$.

4.2 Results for Stationary Cases

The following meteorological data was used:

- Wind speed 3 m/s at 11.5 m above ground level
- Wind direction in the direction of the nearest receptor at 2250 m
- Dispersion class D (neutral-stable) and E (stable)
- No rain, 1 mm/h, 5 mm/h and 10 mm/h

These generic cases serve to illustrate the essential dependencies of the objective quantities on the dispersion class and the precipitation intensity. With regard to the wind speed, it can be assumed that the results are mainly reciprocally proportional to the wind speed, i.e. if the wind speed is doubled, all results are halved.

4.2.1 Activity Concentration in the Air

The Figure 6 (stability Class D) and Figure 7 (stability class E) show the activity concentration with increasing distance from the source for different rainfall intensities.

As expected, the concentration decreases with increasing distance from the source. The value decreases between 1000 m and 2500 m distance by approx. 3/4 (stability class D) or 2/3 (stability class E). At larger distances, the reduction is then significantly smaller.

With increasing rainfall intensity, the total amount of tritium washed out increases and the concentration in the air decreases proportionally. At a precipitation intensity of 1 mm/h, the

decrease in concentration compared to the case without rain is hardly noticeable. The depletion at intensities of 5 mm/h and 10 mm/h is more clearly noticeable.

The washout from the plume is linear with the rain intensity, i.e. at 5 mm/h 5 times more is washed out than at 1 mm/h, at 10 mm/h 2 times more than at 5 mm/h. These ratios can be roughly seen in the graph near the source. With increasing distance, these ratios are reduced, as more has already been washed out at higher rainfall intensities by the time the viewing point is reached.



Figure 6:Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), different rain intensities.
Activity Concentration of Tritium in the air $[\mu Ci/m^3]$ as a function of distance.





It is important to note that the higher the rainfall intensity, the lower the concentrations.

4.2.2 Tritium Activity in Rainwater

The Figure 8 (stability Class D) and Figure 9 (stability class E) show the activity concentration in rainwater with increasing distance from the source for different rainfall intensities.

As expected, the concentration in rainwater decreases with increasing distance from the source. The value decreases between 1000 m and 2500 m distance by approximately 1/2 for both stability class D and E. At greater distances, the decrease is then lower but overall smoother than for the air concentration.

Although the total amount of tritium washed out increases with increasing precipitation intensity, the concentration in the rainwater decreases. On the one hand because the amount of rain is greater and on the other hand because more tritium was washed out until to the location under consideration. However, the values vary only slightly, so that it can be estimated as a good approximation that the activity concentration is not dependent on the intensity of the precipitation.

It should also be noted that in the case of class E, the concentration values in rainwater are only about 30% higher than the values for class D, although the air concentration values are about twice as high. The reason for this is that the washout is effective over the entire plume height, i.e. with regard to the washout, only the different plume width between D and E in the cross direction is relevant.



Figure 8: Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), different rain intensities. Activity Concentration of Tritium in the rainwater $[\mu Ci/L]$ as a function of distance.



Figure 9:Stationary Case: Wind Speed 3 m/s, Stability Class E (stable), different rain intensities.Activity Concentration of Tritium in the rainwater [μ Ci/L] as a function of distance.

4.2.3 Impact of Tritium at 2250 m

The Figure 10, Table 1 (stability class D) and Figure 11, Table 2 (stability class E) show the total impact of tritium on the ground during 24 hours and the activity concentration of Tritium in air and rainwater at 2250 m from the source.

The discussed characteristics regarding air concentration and water concentration for the different rain intensities are noticeable. The air concentration for class E is 2 to 3 times higher than for class D. The dependence on precipitation intensity is very low.

The water concentration for class E, on the other hand, is not even twice as high as for class D. The dependence on precipitation intensity is noticeable but low.

But the input of activity with the rain onto a surface is directly dependent on the rainfall intensity.



Figure 10: Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), different rain intensities. Total impact of Tritium with the rain on the ground, Activity Concentration of Tritium in air and in rainwater at 2250 m from source.

Table 1:Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), different rain intensities
Values corresponding to Figure 10.

Rain Intensity	H3 Air Concentration	H3 Water Concentration	Impact
mm/h	µCi/m³	µCi/L	µCi/m²
0	5.7	-	-
1	5.5	44.7	44.7
5	4.9	40.2	200.9
10	4.2	35.2	351.9



- *Figure 11:* Stationary Case: Wind Speed 3 m/s, Stability Class E (stable), different rain intensities. Total impact of Tritium with the rain on the ground, Activity Concentration of Tritium in air and in rainwater at 2250 m from source.
- **Table 2:**Stationary Case: Wind Speed 3 m/s, Stability Class E (neutral), different rain intensities
Values corresponding to Figure 11.

Rain Intensity	H3 Air Concentration	H3 Water Concentration	Impact
mm/h	μCi/m³	µCi/L	µCi/m²
0	14.0	-	-
1	13.5	70.3	70.3
5	11.8	62.7	313.7
10	10.0	54.5	545.1

4.2.4 Impact of Tritium at 5000 m

The Figure 12, Table 3 (stability class D) and Figure 13, Table 4 (stability class E) show the total impact of tritium on the ground during 24 hours and the activity concentration of Tritium in air and rainwater at 5000 m from the source.

The discussed characteristics regarding air concentration and water concentration for the different rain intensities are noticeable. The air concentration for class E is 2 to 3 times higher than for class D. The dependence on precipitation intensity is very low.

The water concentration for class E, on the other hand, is not even twice as high as for class D. The dependence on precipitation intensity is noticeable but low.

But the input of activity with the rain onto a surface is directly dependent on the rainfall intensity.



Figure 12:Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), different rain intensities.
Total impact of Tritium with the rain on the ground, Activity Concentration of Tritium in air
and in rainwater at 5000 m from source.

Table 3:Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), different rain intensities
Values corresponding to Figure 12.

Rain Intensity	H3 Air Concentration	H3 Water Concentration	Impact
mm/h	µCi/m³	µCi/L	µCi/m²
0	1.7	-	-
1	1.6	22.8	22.8
5	1.3	18.7	93.3
10	1.0	14.5	145.0



- Figure 13:Stationary Case: Wind Speed 3 m/s, Stability Class E (stable), different rain intensities.
Total impact of Tritium with the rain on the ground, Activity Concentration of Tritium in air
and in rainwater at 5000 m from source.
- Table 4:Stationary Case: Wind Speed 3 m/s, Stability Class E (neutral), different rain intensities
Values corresponding to Figure 13.

Rain Intensity	H3 Air Concentration	H3 Water Concentration	Impact
mm/h	μCi/m³	µCi/L	µCi/m²
0	5.7	-	-
1	5.4	41.3	41.3
5	4.2	33.5	167.6
10	3.1	26.0	259.5

4.3 Results for non-stationary Cases

The following meteorological data was used:

- Wind speed 3 m/s at 11.5 m above ground level
- Wind direction in the direction of the nearest receptor at 2250 m
- Dispersion class D (neutral-stable) and E (stable)
- An episode of rain falls with 5 mm/h from 13:00 to 14:00
- An episode of rain falls with 1.5 mm/h from 09:00 to 17:00

These cases are intended to describe the situation for rare situations that occur with a probability of 5%. With regard to the wind speed, it can also be assumed here that the results are essentially reciprocally proportional to the wind speed, i.e. if the wind speed doubles, all results are halved.

4.3.1 Activity Concentration in the Air and in Rainwater

The Figure 14 (stability class D) and Figure 15 (stability class E) show the activity concentration of Tritium in air and rainwater at 2250 m from the source during a time of 24 hours. From 13:00 to 14:00 it rains with an intensity of 5 mm/h.

Likewise, Figure 16 (stability class D) and Figure 17 (stability class E) show the activity concentration of tritium in the air and in rainwater at a distance of 2250 m from the source over a period of 24 hours. In this case, it rains with an intensity of 1.5 mm/h in the period from 09:00 to 17:00.

The minor fluctuation in concentration over time is a characteristic of the ART model, which uses a statistical method. The lower air concentration during the one-hour rain event is significant.

As already shown in the previous chapter, the concentration for class E is about twice as high as for class D. The concentration in water, on the other hand, is only about 3/2 times higher.

As previously shown and discussed, the concentration in the rainwater is almost the same for the two rain events under consideration (1 hour at 5 mm/h and 8 hours at 1.5 mm/h).



Figure 14: Non-Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral), rain intensity 5 mm/h for one hour. Time dependent Activity Concentration of Tritium in air and in rainwater at 2250 m.



Figure 15: Non-Stationary Case: Wind Speed 3 m/s, Stability Class E (stable), rain intensity 5 mm/h for one hour. Time dependent Activity Concentration of Tritium in air and in rainwater at 2250 m.







Figure 17: Non-Stationary Case: Wind Speed 3 m/s, Stability Class E (stable), rain intensity 1.5 mm/h for 8 hours. Time dependent Activity Concentration of Tritium in air and in rainwater at 2250 m.

4.3.2 Impact of Tritium at 2250 m

The Figure 18 and Table 5 show the total impact of tritium on the ground during 24 hours and the activity concentration of Tritium in air and rainwater at 2250 m from the source. The two rainfall events with 5 mm/h for one hour and 1.5 mm/h for 8 hours are shown together.

The air concentration and the water concentration depend only slightly on the type of rain event. However, the total deposited activity varies greatly with the total amount of precipitation, which is more than twice as high for the 8-hour event compared to the 1-hour event.



- Figure 18:Non-Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral) and E (stable),
rain intensity 5 mm/h during 1 hour and 1.5 mm/h during 8 hours.
Total Impact of Tritium with the rain on the ground, Activity Concentration of Tritium in air
and in rainwater at 2250 m from Source.
- Table 5:Non-Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral) and E (stable)Values corresponding to Figure 18.

Case regarding Stability Class / Rain	H3 Air Concentration	H3 Water Concentration	Impact
	µCi/m³	µCi/L	µCi/m²
D, 5 mm/h for 1 hour	5.7	41.0	205
E, 5 mm/h for 1 hour	13.9	64.2	321
D, 1.5 mm/h for 8 hours	5.6	44.5	534
E, 1.5 mm/h for 8 hours	13.8	69.9	839

4.3.3 Impact of Tritium at 5000 m

The Figure 19 and Table 6 show the total impact of tritium on the ground during 24 hours and the activity concentration of Tritium in air and rainwater at 5500 m from the source. The two rainfall events with 5 mm/h for one hour and 1.5 mm/h for 8 hours are shown together.

The air concentration and the water concentration depend only slightly on the type of rain event. However, the total deposited activity varies greatly with the total amount of precipitation, which is more than twice as high for the 8-hour event compared to the 1-hour event.



- Figure 19:Non-Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral) and E (stable),
rain intensity 5 mm/h during 1 hour and 1.5 mm/h during 8 hours.
Total Impact of Tritium with the rain on the ground, Activity Concentration of Tritium in air
and in rainwater at 5000 m from Source.
- Table 6:Non-Stationary Case: Wind Speed 3 m/s, Stability Class D (neutral) and E (stable)Values corresponding to Figure 19.

Case regarding Stability Class / Rain	H3 Air Concentration	H3 Water Concentration	Impact
	µCi/m³	µCi/L	µCi/m²
D, 5 mm/h for 1 hour	1.7	20.1	100
E, 5 mm/h for 1 hour	5.6	36.3	181
D, 1.5 mm/h for 8 hours	1.7	22.6	272
E, 1.5 mm/h for 8 hours	5.5	41.0	492

5 Summary

5.1 Introduction

The report examines the dispersion and environmental impact of tritium released during the venting of Flanged Tritium Waste Containers (FTWCs) at Los Alamos National Laboratory (LANL). The study was conducted by Ingenieurbüro Rau in collaboration with Dr. Winkler and was commissioned by Communities for Clean Water (CCW) in New Mexico.

5.2 Objective

The primary goal of the study is to assess how tritium disperses in the air and how it deposits through precipitation. This evaluation is critical for understanding the potential contamination risks at nearby locations, such as La Vista Church (2.2 km from the release site) and the Rio Grande River (5 km away).

5.3 Methodology

The study utilizes the German dispersion model ARTM, a Lagrangian model used for regulatory assessments of nuclear emissions. The model accounts for:

- **Air dispersion**: Tracking tritium movement in the atmosphere.
- Wet deposition: Analyzing how tritium is washed out by rain.
- Meteorological data: Using 30 years of wind and precipitation records from LANL.

5.4 Key Findings

5.4.1 Stationary Case Analysis

- Air Concentration: Tritium concentration decreases with distance from the source. Stability class E (stable atmospheric conditions) leads to higher concentrations compared to stability class D (neutral conditions).
- **Rainwater Contamination**: Higher rain intensity results in more tritium being washed out of the air but reduces its concentration in rainwater.
- **Ground Deposition**: Tritium deposition on the ground increases with higher rainfall rates, leading to higher contamination levels.

5.4.2 Non-Stationary Case Analysis

- **Short-term rain events** (e.g., 5 mm/h for 1 hour) result in noticeable but temporary reductions in air tritium levels.
- **Extended rain events** (e.g., 1.5 mm/h for 8 hours) lead to higher total deposition on the ground but show similar tritium concentrations in rainwater compared to short-term events.

5.5 Environmental Impact and Conclusion

- The probability of high tritium deposition at the nearest receptor locations is relatively low, as rain events from the critical wind direction (west-northwest) are infrequent.
- However, significant tritium release into the environment occurs during venting, with potential implications for local water bodies and ecosystems.
- The report emphasizes the need for careful monitoring and potential mitigation measures to limit environmental exposure.

This study provides crucial insights into tritium dispersion dynamics and highlights the importance of responsible handling of nuclear waste materials to minimize environmental risks.

6 Reference

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