STORAGE OF TRITIATED WATERS IN SALT CAVERNS

Pierre Bérest, Benoît Brouard
Laboratoire de Mécanique des Solides - Ecole Polytechnique

François Beugin, Jonathan Goldberg
Ecole Polytechnique

ABSTRACT

Salt caverns kept at a relatively low pressure by a permanent hydrocarbon blanket offer an appealing solution to the problem of temporary storage of hazardous liquid wastes. Groundwater flow occurs toward the cavern, preventing any wastes leaks. The storage remains safe even in the case of well-head failure and retrievability is possible at any time. Finally, cavern convergence is accelerated by reducing the density of the fluid column in the well but to an extent that remains acceptable in most cases.

Tritiated Water

Tritium is a radioactive isotope of hydrogen with a mass about three times that of ordinary hydrogen. The basic tritium disintegration reaction can be written as follows:

$$^{3}_{1}\mathrm{T} \;\rightarrow\; ^{3}_{2}\mathrm{He}^{+} + \beta^{-} + \nu_e + 18.5 \text{ eV}$$

which means that tritium emits β radiation (electrons). This amount of radiation is not very sharp — it can be stopped by a sheet of paper — but direct contact with the skin or ingestion can lead to severe health problems.

The activity of 1 gram of tritium is 359 TBq or 9700 Ci. The latter figure does not give a clear image of tritium toxicity, as it must be multiplied by a coefficient that takes into account its small radiated energy per gram (327 mW/g) and its short biological period. (Its transient time in the human body is 10 days.) This coefficient is exceptionally small — i.e., $1.7\ 10^{-11}$ in the case of tritium.

From a practical point of view, this means that if 1 cm^2 of skin is kept in contact with tritiated water (whose activity is 100,000 Ci/liter) for 3 minutes, the resultant radioactive dose will be 40 mSieverts. A dose larger than 50 mSieverts causes changes in blood composition, while a dose larger than 500 m Sieverts leads to severe physical damages. The dose of 1 mSievert per year is generally considered to be a reasonable limit for the public. This dose would be reached by a person who drinks tritiated water with a concentration of $2\ 10^{-7}$ Ci/liter for an entire year.

Tritium is a by-product of many nuclear reactions; tritiated waters are a mixture of H_2O, THO and TO_2. Tritium can easily be extracted from waters whose concentration is higher than 2000 Ci/liter. At the present time, only less-concentrated tritiated waters are released in the atmosphere.

Concentration versus Dilution

Management of hazardous (radioactive or chemical) wastes encompasses two main options that can be considered, to some extent, as opposites:

- concentration of the wastes in small volumes of high toxic activity, and confinement of these wastes, whose release to the biosphere must be nil or so small and/or slow that the released amount cannot be harmful; and
- dilution of the wastes in so large a volume of water or air that the resultant activity per unit volume of fluid becomes negligible.

The second option has long been the only one considered for tritiated waters, which, for example, are released in the ocean at locations where waters are stirred by natural streams. Nevertheless, due to increasing concern with environmental protection, there is a strong trend to reduce to zero the amount of radionuclides released in the biosphere. Whether such a trend is sound is not the topic to be discussed in the present paper; we wish to prove that, in some cases, salt caverns do provide a safe confinement solution for hazardous wastes.

Tritium Period

The tritium half-life period is 12.43 years, which means that its activity naturally decreases by 5% per year. In other words, the initial activity of tritium is divided by 250 after one century and by 62,500 after two centuries. A very important characteristic of tritiated waste is that it must be withdrawn from the biosphere during a period of time which, albeit long, is limited. Thus, the main objectives for tritiated-water storage can be summed up as follows.

1. The required volume is several thousands of cubic meters.
2. The tritiated waters released from the storage must be nil.
3. The storage must remain safe during a period of, typically, one to two centuries.
4. The handling of waste during storage operations must be as simple as possible. Risk of pump failure, water leaks and more generally hazardous events must be minimized for all personnel involved.

Basic Storage Principle

These objectives can easily be met by storing tritiated water in salt caverns, provided that the fluid pressure in the cavern is kept significantly smaller than the "halmostatic" pressure — i.e., the pressure of a cavern whose well is filled with saturated brine and opened to the atmosphere at ground level.

As an example, we suppose that 600 m^3/year of tritiated water must be stored for a period of 20 years. We also assume that a cavern whose volume is 16,000 m^3 will be created at a depth of approximately 1000 meters below ground level.

The liquid wastes (tritiated brines) occupy the cavern bottom; the upper part of the cavern is filled with a lighter liquid (for instance, fuel oil). The amount of stored liquid wastes will increase with time; an equivalent volume of fuel oil will be progressively withdrawn from the cavern so that the total volume (brine + hydrocarbon) is kept constant. There is no physical or chemical exchanges between the two liquids (except for a small amount of gas generated at the interface due to radiolytic effects); so, radioactivity is confined in the lower part of the cavern.

The well is equipped with a steel casing whose shoe is anchored above the cavern neck. One (or two) strings are set in the well, delimiting one (or two) annular spaces and a central volume (see Figure 1). The liquid wastes are injected in the cavern through the central tubing, and fuel is removed through the annular space. When the system is at rest, the well's central tubing and annular spaces are filled with fuel oil and the well-head pressure is null or small, resulting in a cavern pressure that is smaller than the halmostatic figure (obtained when the well is filled with brine), and even smaller than the hydrostatic pressure (obtained when the well is filled with soft water).

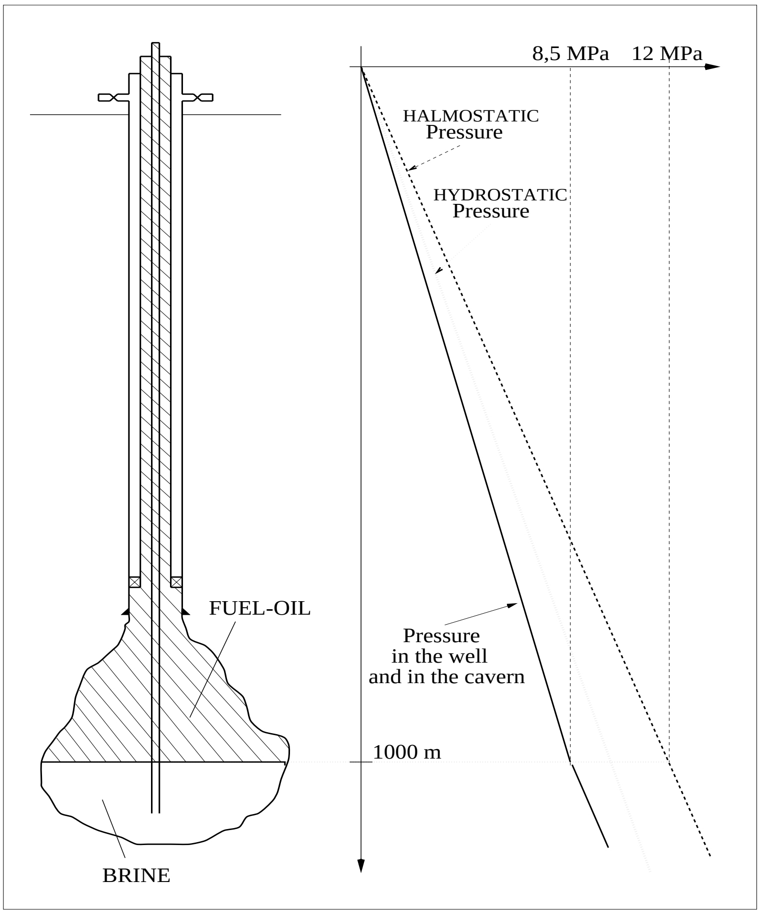

Figure 1: Pressure distributions in the well

Cavern Tightness

Waste confinement in a salt cavern is based on:

- the extremely small permeability of rock salt, and
- the depressurization of the cavern, whose pressure is smaller than the natural pore pressure of the brine in the rock mass.

Rock salt exhibits a very low permeability, because the hydraulic conductivity of its matrix is extremely small, and because no fractures exist in a massive salt formation (except, perhaps, in some disturbed zones encountered at the fringe of salt formations). Several authors think that most of this (small) permeability is induced by the cavern creation — more precisely, by tensile or high deviatoric stresses developed at the cavern wall when cavern pressure is whether very high or very small (see Behrendt et al. 1997). In the fairly pure salt of the WIPP site (New Mexico), precise in-situ measurements in a ventilation shaft gave $K = 10^{-22}$ m^2 (Dale and Hurtado, 1997). In the Etrez site, where anhydrite and clay interbeds are present, a one-year test in a well gave $K = 6.10^{-20}$ m^2. These figures are quite small. If we select, for rough estimation, $K = 10^{-20}$ m^2, a 3- MPa excess of pressure in the cavern (compared to the natural pore pressure in the rock mass) will lead to a 1.6- m^3/year leak in a 16,000-m^3 cavern, or 10^{-4} per year relative loss ov volume.

The natural pore pressure in the rock mass is an important parameter for the above estimation. This pressure is generally considered to be close to the halmostatic pressure. In other words, there is no brine flow to or from a cavern if the cavern well is filled with saturated brine and opened to atmosphere. In some cases (as in the WIPP site above mentioned), the pore pressure can even be significantly higher than halmostatic.

In the proposed concept, due to filling the well with fuel oil, brine pressure in the cavern is always smaller than halmostatic pressure, which means that the brine flow is directed toward the cavern and is small — i.e., 1 to several m^3 per year if we assume the same cavern size and rock properties as before. No flow from the cavern to the rock mass can exist.

Well Integrity

It follows that the crucial issue for storage tightness is the integrity of the well — not salt permeability — because cavern pressure is below hydrostatic. The casing, cementation and casing shoe are, potentially, the weakest points. In the suggested concept, the liquid wastes are confined to the cavern bottom; so, any failure in well integrity will lead to a fuel-oil leak — not a waste leak.

Waste Handling

Tritiated water is highly corrosive. Any failure of the surface equipment (pumps, wellhead, tubes) that can cause water spills can induce harmful consequences for the crew and the environment. The probability of such a failure must be as close as possible to zero. The simplest waste-handling system is the best. A hydrocarbon-filled well allows for purely gravity-driven flow of liquid wastes, which are heavier than hydrocarbons. Thus, pumps are unnecessary.

Retrievability

It is always possible to economically pump out a high percentage of the stored waste from a cavern.

Human Intrusion

The period of time during which wastes must be confined is relatively small (one to two centuries). For such short periods inadvertent human intrusion is not a risk to be considered.

Such events as willful damage, plane crashes or earthquakes can lead to well-head failure. The stored liquid, being heavier than hydrocarbons and soft water, will remain at the cavern bottom, with no risk of upward migration to aquifer layers or ground level.

Cavern Creep

For deep caverns, creep can be a concern. Some case studies (Boucly and Legreneur, 1980; Baar, 1977; Sasse et al., 1973) prove that a few deep caverns have experienced large shrinkage, of typical volume loss rates ranging from 3% to 30% per year. In fact, large volume losses occur in deep natural gas caverns for two reasons:

- the geostatic pressure is high (because the depth is large), and
- the internal pressure is low, especially, of course, when the cavern is almost empty.

In other words, the critical parameter is not cavern depth (i.e., the geostatic pressure), but the gap between the geostatic pressure at cavern depth and the cavern fluid pressure. A natural gas cavity at medium depth can creep as quickly as a brine-filled cavity at greater depth.

In order to clarify this statement, we can estimate that the geostatic pressure, P_∞ (in MPa), due to overburden weight at depth H (in meters) is

$$P_\infty = 0.022\ H$$

which is the equivalent of an average rock density of 2200 kg/m^3.

If the well is filled with a saturated brine, with a density of 1200 kg/m^3, and if no additional well-head pressure is applied to the brine column, then the internal pressure in a cavern located at depth H is

$$P_i = 0.012\ H$$

In this case, the gap between geostatic pressure and internal pressure is simply proportional to depth:

$$P_\infty - P_i = 0.01\ H$$

For example, at a 1000- meter depth, the gap is 10 MPa. This formula indicates that creep and cavern loss rate are **function** of depth. In fact, the cavern loss rate is not proportional to depth, because rock-salt behavior is highly non-linear: a small increase in pressure gap can lead to a drastic increase in convergence rate.

Fuel-Filled Wells

Fuel oil density (850 kg/m^3) is lower than brine density, which is 1200 kg/m^3. When substituting fuel oil for brine in the well, the fluid pressure in the cavern itself is significantly lowered, resulting in faster creep. This effect must be thoroughly evaluated; a too-high convergence rate can be troublesome, because it reduces the available cavern volume.

When a cavern located at depth H is filled with fuel oil, the well-head pressure in the cavern is

$$P_i = 0.0085\ H$$

This means that the gap between the geostatic and internal pressures is

$$P_\infty - P_i = 0.0135\ H$$

The difference for the case of a brine-filled well will be discussed when the salt mechanical behavior is specified.

Salt Constitutive Behavior

Many authors agree the steady-state (or "secondary") salt creep is well described by the Norton-Hoff model, whose uniaxial formulation can be written as:

$$\dot{\varepsilon} = A\ \exp\ (-Q/RT)\ \sigma^n$$

where $\dot{\varepsilon}$ is the axial strain rate of a cylindrical sample ($\dot{\varepsilon} = \dot{h}/h$) submitted to an axial stress, P, and a lateral confinement pressure, q, whose difference is $\sigma = P - q$. In other words, during an uniaxial compression test, σ is simply the applied load ($q = 0$). The (absolute) temperature is T, and the three constants ($A, Q/R$ and n) must be determined through laboratory experiments. The case $n = 1$ is described as being "Newtonian": the flow rate is simply proportional to the applied shear stress. Salt rock is definitely not a Newtonian fluid: the constant n ranges from $n = 3$ to $n = 5$ for most cases.

The uniaxial formulation of material constitutive behavior is very convenient for a description of triaxial test results, but it must be generalized to the three-dimensional case for use in numerical calculations. It is generally assumed that rock salt has no volumetric expansion or contraction. Thus, if d_{ij} holds for the strain rate tensor, σ_{ij} is the stress tensor, s_{ij} is the deviatoric stress tensor, $s_{ij} = \sigma_{ij} - (\mathrm{tr}\sigma)/3$, and J_2 is the second invariant of the deviatoric stress tensor $J_2 = \frac{1}{2} s_{ij} s_{ji}$, then the salt constitutive behavior can be written as:

$$d_{ij} = \frac{\partial \Phi}{\partial s_{ij}} \qquad , \qquad \Phi = A\ \exp\ \left(-\frac{Q}{RT}\right)\ \frac{\left(\sqrt{3J_2}\right)^n}{(n+1)}$$

To what extent such a law captures the main features of the mechanical behavior of salt has been extensively discussed by numerous authors (see, for example, Hardy et al., 1984, 1988, 1996). Primary creep, which is the response to variation of the applied load, is not taken into account; however, in the context of the present paper (caverns submitted to constant pressures), it can probably be neglected. It is our conviction that, in this context, the suggested law provides a satisfying approximation.

Cavern Creep

If P_∞ (geostatic pressure) and P_i (cavern internal pressure) are as in the uniaxial formulation, the steady-state volumetric creep rate of a spherical cavern can be easily computed:

$$\frac{\dot{V}}{V} = -\frac{3}{2}\left[\frac{3}{2n}\ (P_\infty - P_i)\right]^n\ A \exp\left(-\frac{Q}{RT}\right)$$

where $n, A, Q/R$ are as above. A similar formula for a cylindrical cavern has been given by Van Sambeek (1993):

$$\frac{\dot{V}}{V} = -\sqrt{3}\left[\frac{\sqrt{3}}{n}\ (P_\infty - P_i)\right]^n\ A \exp\left(-\frac{Q}{RT}\right)$$

which means that the cylindrical case can be deduced from the spherical case by multiplying the latter formula by $(2\sqrt{3}/3)^{n+1}$. Between all possible shapes, the slowest rate is reached when the cavern is spherical.

Real caverns are neither spherical nor perfectly cylindrical; a flat cavern (i.e., having a diameter much larger than its height) behaves fairly distinctly, but the spherical/cylindrical cases provide a good illustration of the behavior of most caverns of regular shape.

Salt Parameters

Parameters for the salt constitutive equation can be found in the literature. Van Sambeek (1993) [who compiled De Vries (1988) and Senseny et al. (1989) data], Senseny (1984), Wawersik (1984) and Pouya (1991) suggest the following values.

Table 1. Parameters for different salts

Facility	n	Q/R (K)	A (year^{-1}.MPa^{-n})	sphere/cylinder ratio
Avery Island (after D.V.)	3.14	6495	1.30 10^4	1.81
WIPP	5.0	5035	1.04	2.37
Etrez	3.1	4100	0.64	1.80
Avery Island (after S. and al.)	4.0	6565	2081	2.05
Salina	4.1	8715	2.7752 10^5	2.08
Palo Duro - Unit 4	5.6	9760	1.806 10^5	2.58
Palo Duro - Unit 5	5.3	9810	2.52 10^5	2.47
Salado (WIPP7)	5.09	8333	3.67 10^4	2.40
Asse (BGRC1)	6.25	9969	2.51 10^4	2.84
West Hackbr. (WH1)	4.73	6606	452.31	2.28
West Hackbr. (WH2)	4.99	10766	0.94	2.37
Bryan Mound (BM3C)	4.54	7623	1.32 10^3	2.22
Bryan Mound (BM4C)	5.18	8977	1.04 10^5	2.43
Bayou Choctaw (BC1)	4.06	5956	64.03	2.07

It must be clear that these parameters are not **recommended** by the authors, who, in some cases, suggest a more elaborate constitutive equation. The authors do not use the same units, and we have converted the constants to get years and Megapascals, which are more convenient for the problem we are discussing.

Direct comparison of data is difficult: when A is large, $\exp\left(-Q/RT\right)$ is small, resulting in not-so-scattered strain rates, except for Avery Island salt as described by De Vries (quoted by Van Sambeek, 1993). It is convenient to compare these data by computing how a spherical cavern will behave as a function of depth if the two following assumptions are made:

1. the temperature at depth is given by the law $T(K) = 288 + 0.03H$ (In other words, the surface temperature is 15^{o}C or 59^{o}F; at a 1000- meter depth, the rock temperature is 45^{o}C or 113^{o}F.);
2. for the case of a well filled with fuel oil,

$$P_\infty - P_i = 0.0135\ H$$

for the case of a brine-filled well,

$$P_\infty - P_i = 0.01\ H$$

The results are given on Figures 2 to 5. The case of a cylindrical cavern can easily be obtained by multiplying the volume loss rate by the ratio given in Table 1. The volume loss rate is larger in the case of a well filled with fuel oil, as expected, but smaller than 1% per year at a 1000 meters depth.

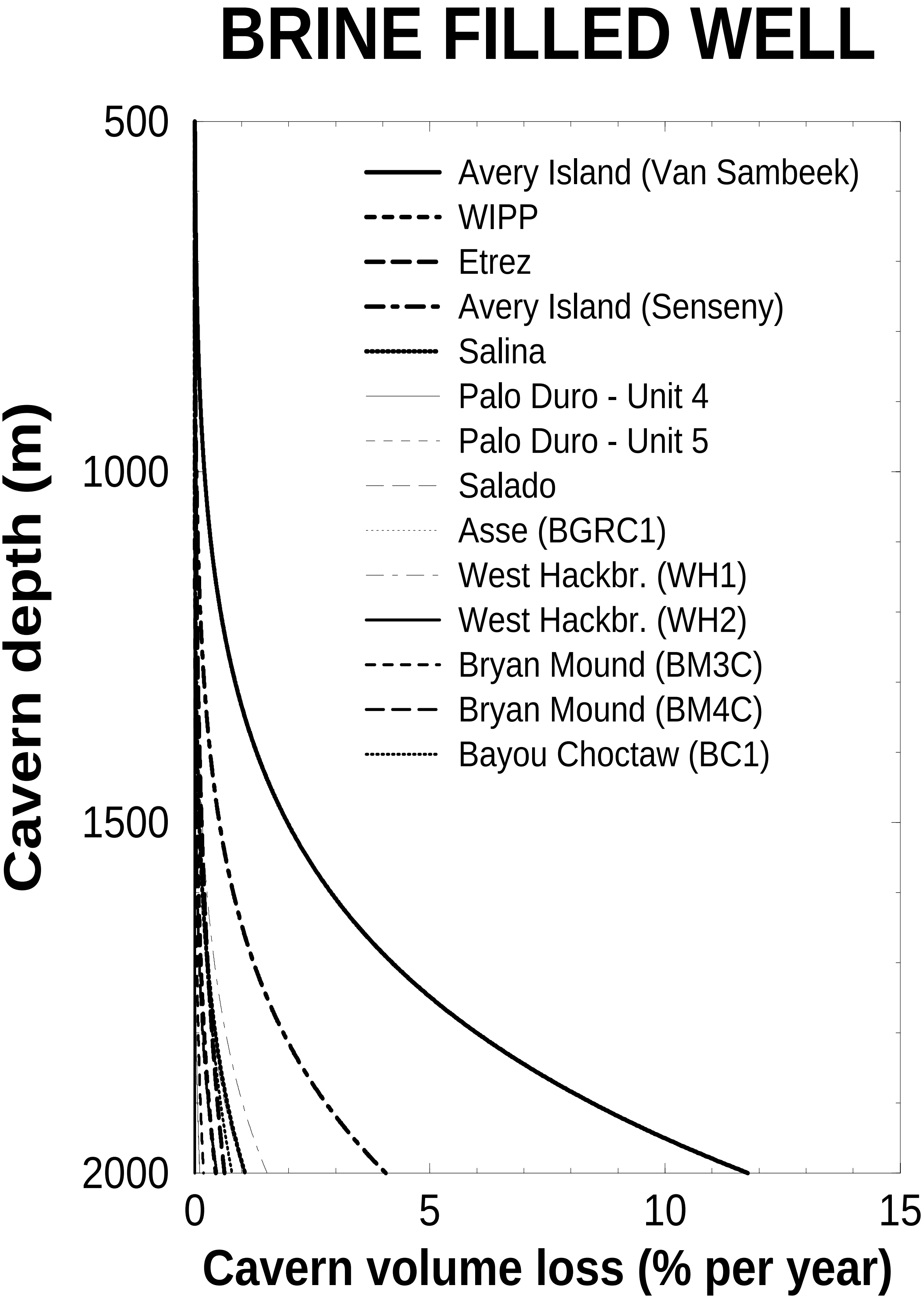

Figure 2: **Data are indicative**

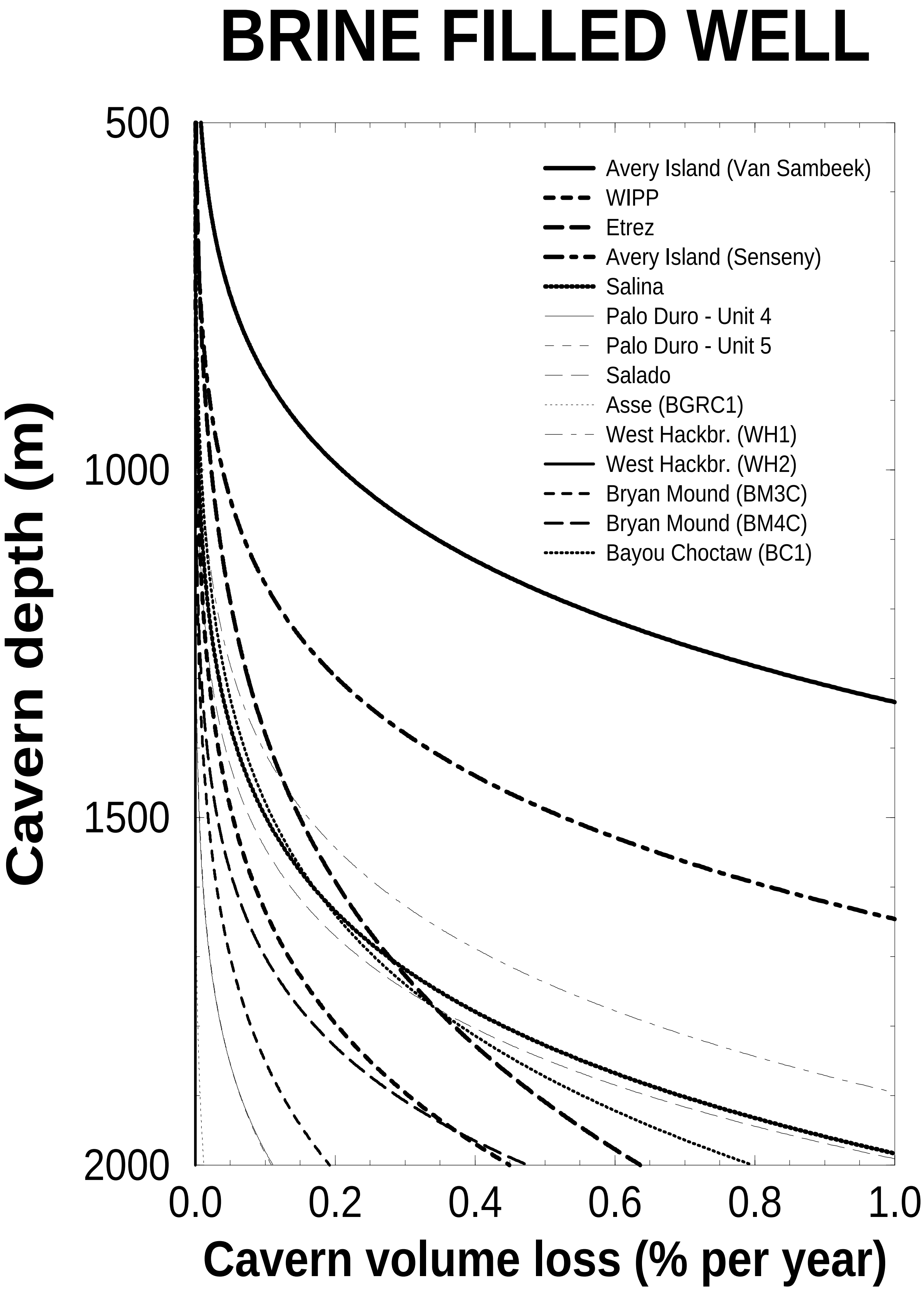

Figure 3: **Data are indicative**

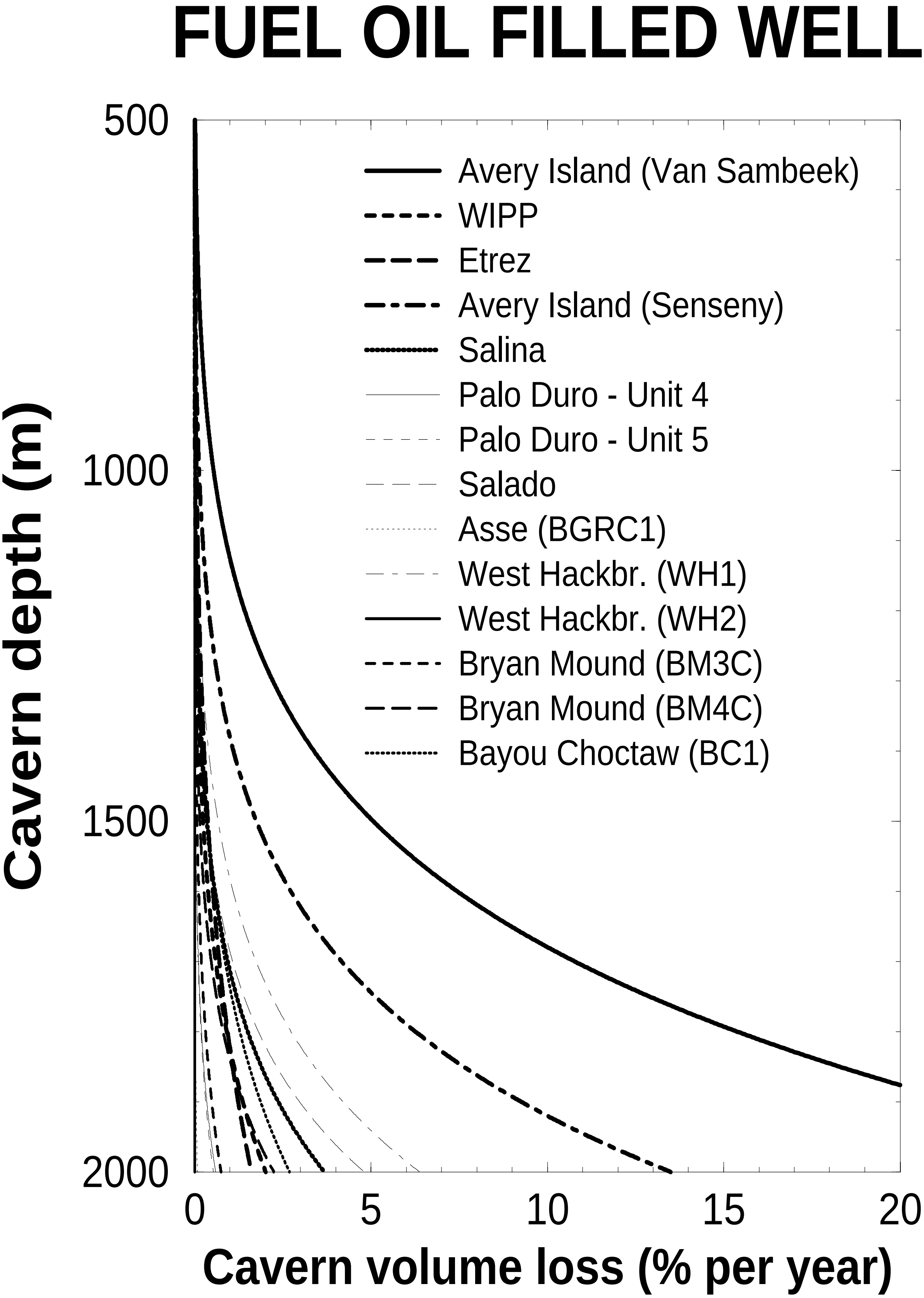

Figure 4: **Data are indicative**

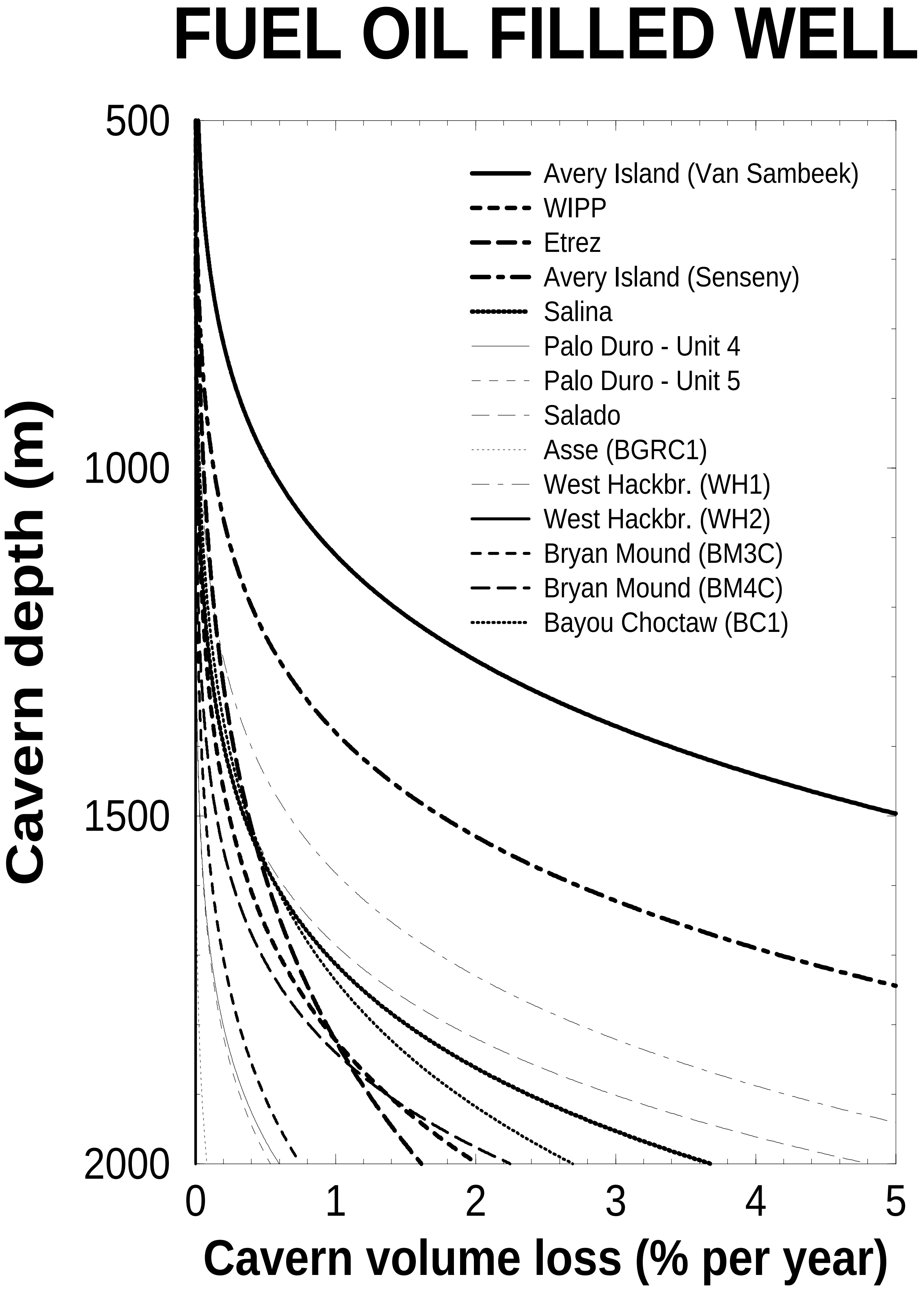

Figure 5: **Data are indicative**

References

[1] Pouya A. *Correlation beween mechanical behaviour and petrological properties of rock salt.* Proc. 32th US Symp. Rock Mech., J.C Roegiers ed., pp. 385-392, 1991.

[2] Behrendt C., Crotogino F.R. Hackney J. *SMRI-Bibliography for Cavern Abandonment.* To be published in 1997, 1997.

[3] Boucly P. Legreneur J. *Hydrocarbon storage in cavities leached out of salt formations.* Proc. Int. Symp. Rockstore 80, Pergamon, Vol. I, pp. 251-258, 1980.

[4] Baar C.A. *Applied Salt-Rock Mechanics, I.* Dlpments in Geot. Eng. 16-A, Elsevier, 1977.

[5] Dale T. Hurtado L.D. *WIPP air-intake shaft disturbed-rock zone study.* Proc. 4th Conf. Mech. Behavior of Salt, Trans Tech Pub., 1997.

[6] Crotogino F.R. *Salt Cavern In-Situ Testing from the constructor's and the operator's viewpoint.* Proc. 1st Conf. Mech. Beh. of Salt - Hardy and Langer ed. - Trans Tech Pub., pp. 613-628, 1984.

[7] Hardy R.H. Langer M. Proc. 1st Conf. Mech. Beh. of Salt, 1984.

[8] Hardy R.H. Langer M. Proc. 2nd Conf. Mech. Beh. of Salt, 1988.

[9] Hardy R.H., Langer M. Berest P. Ghoreychi M. Proc. 3rd Conf. Mech. Beh. of Salt, 1996.

[10] De Vries K.L. *Viscoplastic Laws for Avery Island Salt.* RE/SPEC Inc. Report prepared for Stone and Webster Engineering Corporation (quoted by Van Sambeek, 1993), 1988.

[11] Kuhne G., Rohr W.U. Sasse W. *Kiel gas storage facility, the first city gas cavern in Germany.* Proc. XII World Gas Congress, 1973.

[12] Van Sambeek L.L. *Evaluating Cavern Tests and Subsurface Subsidence Using Simple Numerical Models.* 7th Symp. on Salt, Vol. I, pp. 433-, 1993.

[13] Munson D.E., Fossum A.F. Senseny P.E. *Approach to First Principles model prediction of measured WIPP in situ closure in salt.* Proc. 30th US Symp. Rock Mech., A Wahab Khair ed., pp. 673-680, 1989.

[14] Senseny P.E. *Creep properties of four rock salts.* Proc. 2nd Conf. Mech. Beh. of Salt, Hardy and Langer ed., Trans Tech Pub., pp. 431-444, 1984.

[15] Wawersik W.R. *Alternatives to a power-law creep model for rock salt at temperatures below 160°C.* Proc. 2nd Conf. Mech. Beh. of Salt, Hardy and Langer ed., Trans Tech Pub., pp. 103-128, 1984.