International
Progress Report

IPR-02-60

Äspö Hard Rock Laboratory

Results of repeated tritium analyses

Preformed during the winter 1998/1999

Äspö and Laxemar

Ann-Chatrin Nilsson

KTH

October 1999

Svensk Kärnbränslehantering AB
Swedish Nuclear Fuel
and Waste Management Co
Box 5864
SE-102 40 Stockholm Sweden
Tel +46 8 459 84 00
Fax +46 8 661 57 19

Äspö Hard Rock Laboratory

Report no.
IPR-02-60
No.
F70K
Author
Ann-Chatrin Nilsson
Date
Oct 1999
Checked by
Ingvar Rhén
Date
2002-04-01
Approved
Christer Svemar
Date
2003-05-22

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Keywords: Tritium, analyses, grounwater prof

Abstract

The Äspö Hard Rock Laboratory (HRL) is located in the Simpevarp area, southeast Sweden, some 35 km north of Oskarshamn. Construction of the underground laboratory commenced in 1990 and was completed in 1995, consisting of a 3.6 km long tunnel excavated in crystalline rock to a depth of approximately 460 m. Prior to, during and subsequent to completion, research concerning the deep geological disposal of nuclear waste in fractured crystalline rock has been carried out. Central to this research has been the characterisation of the groundwater flow system and the chemistry of the groundwater at Äspö prior to excavation (Pre-investigation Phase) and subsequently to monitor changes in these parameters during the evolution of laboratory construction (Construction Phase).

SKB has carried out groundwater sampling at large depths in drilled boreholes during the last decade as part of their hydro chemical investigation programme. The results of the analyses of tritium carried out by IFE (Institutt for Energiteknikk, Kjellner, Norway) have lately turned out to be much higher than the correct values.

In late 1998, 49 selected back-up samples were analysed by the University of Waterloo. This report presents the results of these analyses. In almost all of the cases, the results from the Waterloo analyses are considerably lower than the original results from IFE.

Sammanfattning

Äspölaboratoriet är lokaliserat i Simpevarpsregionen i sydöstra Sverige, ca 35 km norr om Oskarshamn. Byggandet av underjordslaboratoriet, som påbörjades 1990 och färdigställdes 1995, består av en 3.6 km lång tunnel utsprängd och borrad i kristallin berggrund ner till ett djup av ca 460m. Innan, under och efter färdigställandet av laboratoriet har forskning genomförts som rör djupförvar av kärnavfall i kristallin berggrund. Centralt för denna forskning har varit karakteriseringen av grundvattenflödessystemet och grundvattenkemin vid Äspö innan tunnelbygget (Förundersökningsfas) och att därefter monitera dessa parametrar under byggandet av laboratoriet (Konstruktionsfas).

SKB har utfört provtagning av grundvatten på stora djup i berggrunden sedan ett tiotal år tillbaka som ett led i sina hydrokemiska undersökningar. De analysresultat av tritium som ursprungligen redovisades av IFE (Institutt for Energiteknikk, Kjeller, Norge) har dock konstaterats ligga i regel alldeles för högt.

Rapporten redovisar resultaten av tritium från en reservprovserie som analyserats av Universitetet i Waterloo. I nästan samtliga fall är resultaten från Waterloo betydligt lägre än de ursprungliga resultaten från IFE.

Contents

List of Tables

List of Figures

1 Introduction

SKB has carried out groundwater sampling at large depths in drilled boreholes during the last decade as part of their hydrochemical investigation programme. The isotopes tritium, deuterium and oxygen-18 are important parameters for the understanding of groundwater flow and residence times, and sampling and analysis for these parameters have been standard parts of the investigation programme.

Laboratory analyses of tritium has generally been carried out by IFE (Institutt for Energiteknikk, Kjellner, Norway). Lately it has been discovered that the tritium values reported by IFE during the period from autumn 1993 to spring 1998 are erroneous (IDT-99-04). The reported values are in general apparently much higher than the correct values.

During the autumn of 1998, 49 selected back-up samples were sent to the University of Waterloo for repeated tritium analysis. These samples were taken from different boreholes/borehole sections at different times during the period autumn 1993 – spring 1998. The new analyses by University of Waterloo were carried out during the winter 1998/1999.

In this report the results from the new analyses by University of Waterloo are presented and compared with the results from the original analyses by IFE.

A summary of the comparison is presented in Figure 1-1, where tritium values from the new analyses are plotted against their corresponding values from the original analyses.

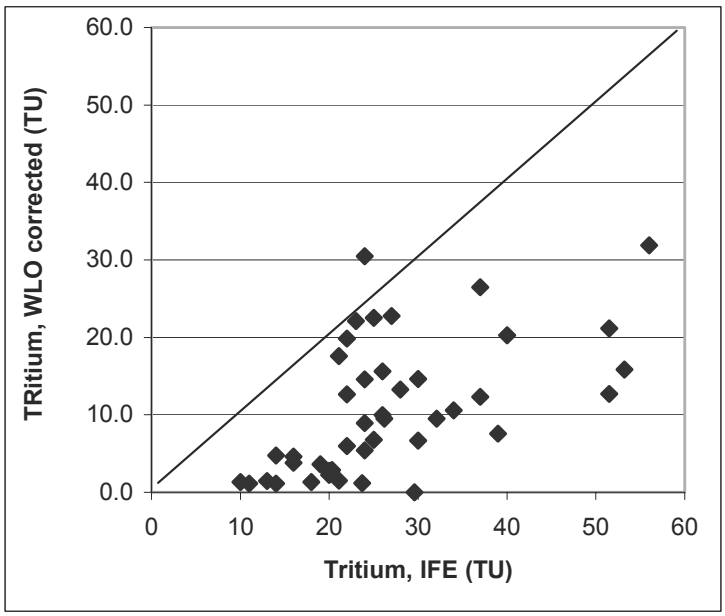


Figure 1-1. *Tritium values from Waterloo plotted against the corresponding original values from analyses by IFE.*

2 Results

The results from the new tritium analyses from University of Waterloo and the original values from IFE are listed in Table 2-3. The values in the column "Tr(WLO)corr" have been corrected for radioactive decay.

In Figures 2-1 to 2-4, the new and the original tritium analyses are compared for different sampling periods. Figures 2-1a to 2-4a show the values from the new analyses (with and without, respectively, correction for radioactive decay) and the values from the original analyses plotted against sample number.

Figures 2-1b to 2-4b show the new (corrected for radioactive decay) analyses plotted against the corresponding values from the original IFE analyses. The different sampling periods considered are divided as follows:

Figure	Time period	Sample numbers	Sample age (years)
2-1	930929-931214	2183-2209	5
2-2	951025-960521	2339-2360	3-2.5
2-3	970925-971001	2416-2451	1
2-4	980302-980312	2476-2491	0.5

In almost all of the cases, the results from the Waterloo analyses are considerably lower than the original results from IFE. In one of the samples the new value is significantly higher than the original, while in three of the samples the new and the original values are approximately equal. The latter samples were collected during the period 930929 – 931213 and are plotted in Figure 2-1. Of the plotted time periods in Figures 2-1 to 2-4, only the latest one (980302 – 980312) shows fairly consistent trends in tritium levels between IFE and Waterloo analyses.

Figures 2-5a to 2-5m compare tritium levels, plotted against sample date, from IFE, CRIEPI and Waterloo for a few different boreholes. Results from the following boreholes are presented:

KR0012B	KA2162B	KA3385A
KR0013B	SA2600A	
KR0015B	KA2862A	
SA0813B	KA3010A	
HA1327B	SA3045A	
SA1420A	KA3110A	

For the boreholes listed above, Figure 2-5 provides reasonably representative overall values for the tritium contents based on the available results. The tritium content of individual samples are, however, more uncertain.

In Figure 2-6, all available analyses for the borehole KLX02 are presented. Tritium values are plotted against length along the borehole to the midpoint of the sampled section. Values below the detection limit have been plotted as zero. Renewed analyses have been carried out during July 1998 as well as during the winter 1998/1999 on samples from the last hose sampling in September 1997. The new analyses were carried out with and without, respectively, prior sample enrichment, which is presented using error brackets in the figure.

The Waterloo results from the last hose sampling in KLX02 show a less marked tritium "jump", at approximately 1100 metres borehole length, than was indicated by earlier analysis results.

Table 2-1. List of laboratories

IFE	Institutt for energiteknikk, Kjeller, Norway
GSF	GSF-Forschungszentrum, Institutt for Hydrologie, Neuherberg, Oberschleissheim, Germany
WLO	Univ. of Waterloo, Dept. of Earth Sciences, Waterloo, Canada
IAEA	IAEA, Vienna
CRI	CRIEPI (Central Research Institute of Electric Power Industry in Japan) or laboratory used by CRIEPI

Table 2-2. Tritium measurement accuracy and detection limits. IFE has reported different values during different time periods.

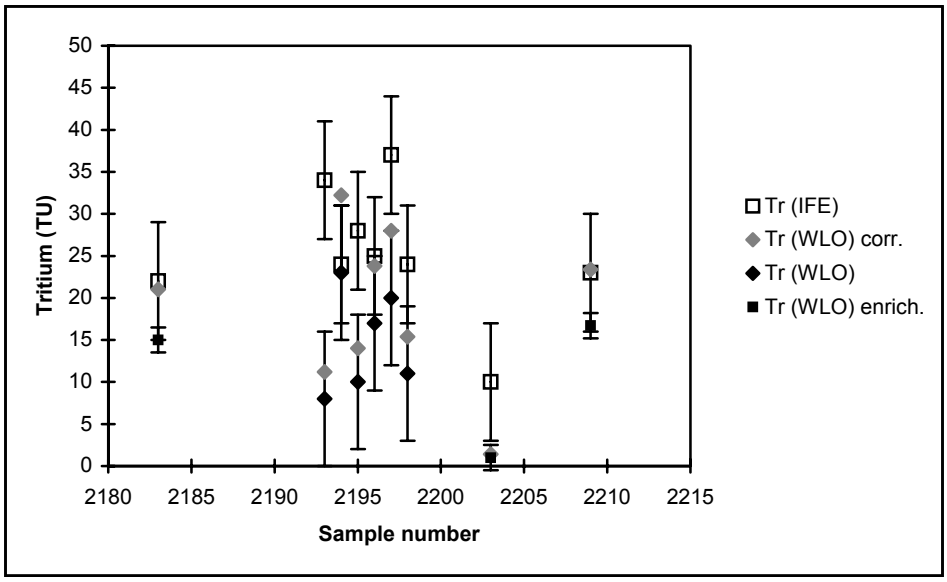
IFE	Detection limit:	4.2 – 8.4 TU	(last 8.4 TU)
	Accuracy:	±4.2 – ± 8.4 TU	(last 6.8 TU, error bars in figures show ± 7 TU)
WLO	Detection limit:	6 TU	
	Accuracy:	± 8 TU	
WLO enriched	Detection limit:	0.8 TU	
	Accuracy:	~ ± 1 TU	

Table 2-3. Comparison between tritium contents from analyses by IFE and the University of Waterloo.

IDCODE	DATE	SECUP	SECLOW	SAMPLE NO	Tr (IFE)	Tr (WLO) corr.	Tr (WLO)	Tr (WLO) enrich.
HBH01	931112	31	50.6	2196	25	23	17	
HBH02	931112	21	32.4	2197	37	26	20	
HBH05	931112	11	22	2198	24	15	11	
KA1755A	971001	88	160	2451	23.7	1.2		1.1
KA2162B	980304			2494	20	2.8		2.7
KA2511A	980305	92	109	2502	30	14.6		14.2
KA2862A	980312	7.37	15.98	2510	13	1.4		1.4
KA3005A	960411	44.78	45.78	2344	30	6.7		5.8
KA3067A	960410	6.55	27.05	2342	14	1.2		1.0
KA3110A	980306	20.05	28.63	2506	26	15.6		15.2
KA3385A	980302	32.05	34.18	2476	16	4.6		4.5
KA3600F	980309			2514	26	10.0		9.7
KI0025F	980305	86	88	2500	19	3.6		3.5
KLX02	970925	150	200	2414		12	11	
KLX02	970925	300	350	2416	29.6	-6	-6	
KLX02	970925	500	550	2424	26.2	10	9	
KLX02	970925	700	750	2427	32.1	10	9	
KLX02	970925	850	900	2429		13.8		13.0
KLX02	970925	950	1000	2430		13.3		12.6
KLX02	970925	1050	1100	2431		13.0		12.3
KLX02	970925	1150	1200	2432		5.1		4.8
KLX02	970925	1250	1300	2433		4.2		4.0
KR0012B	931108	5	10.57	2193	34	11	8	
KR0012B	970930	5	10.57	2443	51.5	21	20	
KR0013B	931108	7.05	16.94	2194	24	30	23	
KR0013B	970930	7.05	16.94	2444	53.2	16	15	
KR0015B	931108	19.82	30.31	2195	28	13	10	
KR0015B	970930	19.82	30.31	2445	51.5	13	12	

KXTT1	960410	15	16	2341	16	3.8		3.3
KXTT2	960411	11.55	13.55	2347	39	7.6		6.6
KXTT2	960412	14.55	15.55	2348	22	6.0		5.2
KXTT3	960410	12.42	14.42	2343	24	5.4		4.7
KXTT3	980304	12.42	14.42	2491	56	32	31	
KXTT4	960409	14.92	23.42	2340	25	6.8		5.9
SA0813B	960521	5.6	19.7	2353	21.1	17.6		15.3
SA1009B	960521	6	19.5	2356	27	22.8		19.8
SA1210A	931214	6	20.5	2209	23	22.1		16.7
SA1229A	980303			2483	37	12	12	
SA1420A	930929	6	50	2183	22	19.9		15.0
SA1420A	960521	6	50	2358	40	20.2		17.6
SA1730A	980303			2486	18	1.3		1.3
SA2074A	960521	6	38.7	2360	22	12.7		11.0
SA2273A	980302			2480	24	8.9		8.7
SA2600A	931202	5.8	19.4	2203	10	1.3		1.0
SA2600A	960521	5.8	19.4	2351	20.3	2.9		2.5
SA2783A	960520	5.8	19.9	2352	21.1	1.5		1.3
SA2880A	980306			2505	14	4.7		4.6
SA3045A	951025			2339	20	2.2		1.9
SA3045A	980306			2508	11	1.1		1.1

a)



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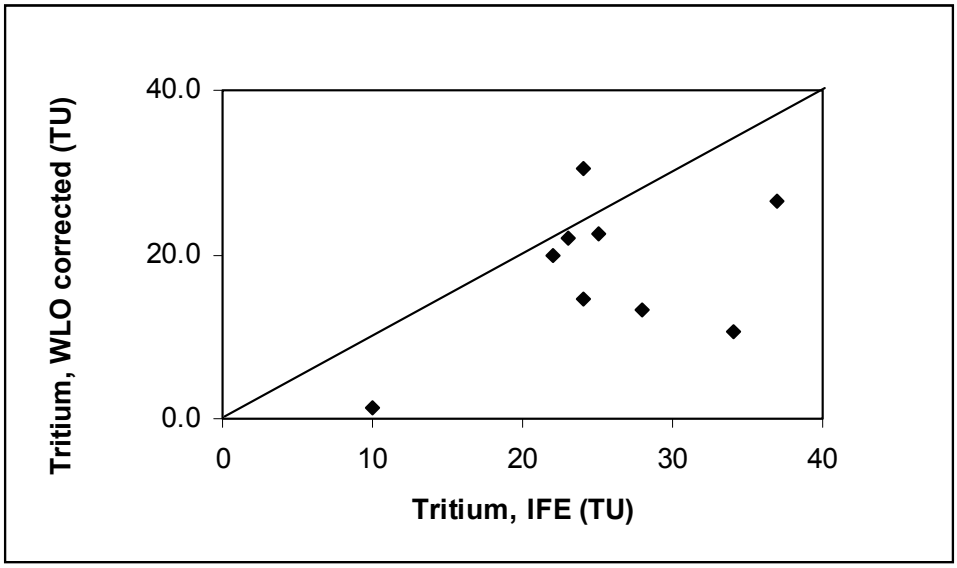
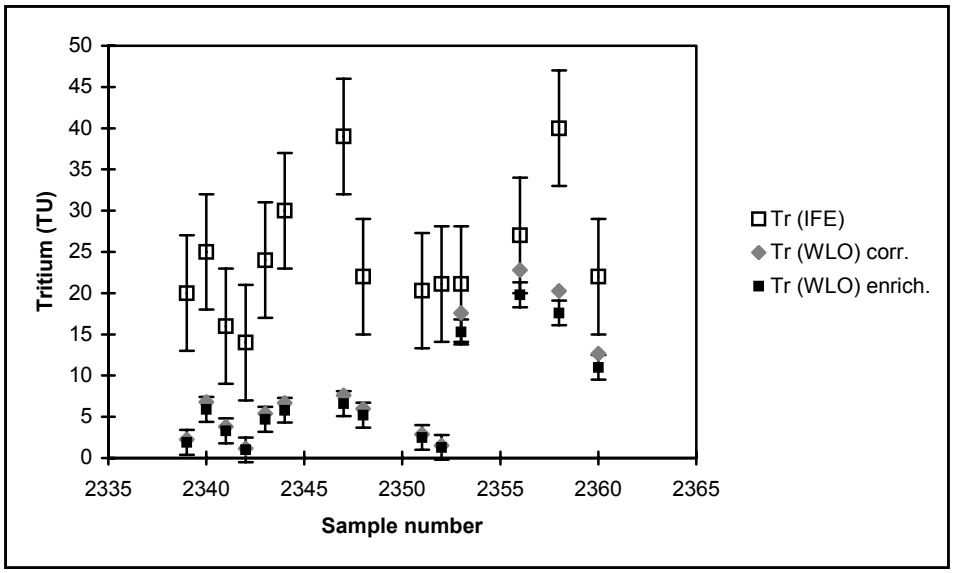


Figure 2-1. *Comparison of original (IFE) and new (WLO) tritium analyses for the sampling period 9309 – 9312.*

a)



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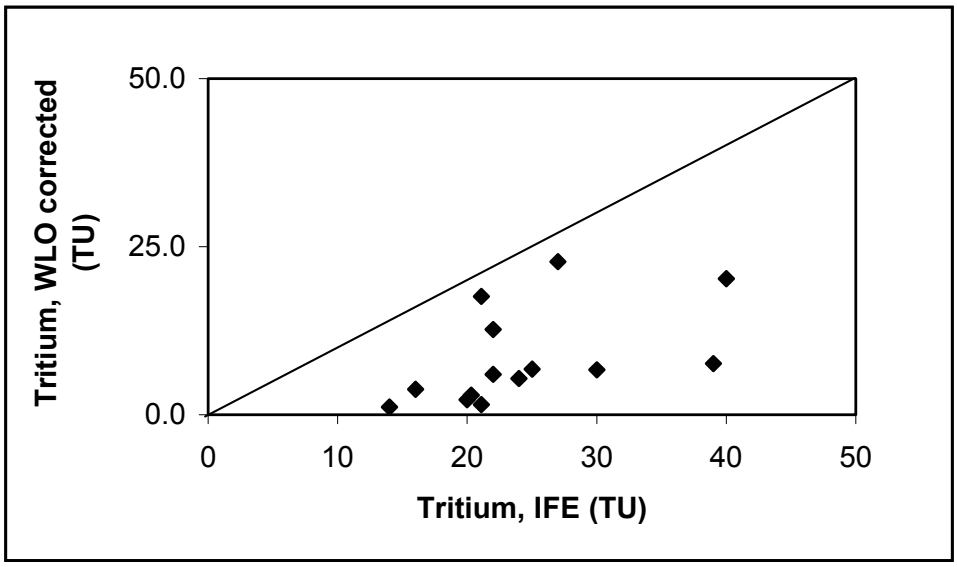
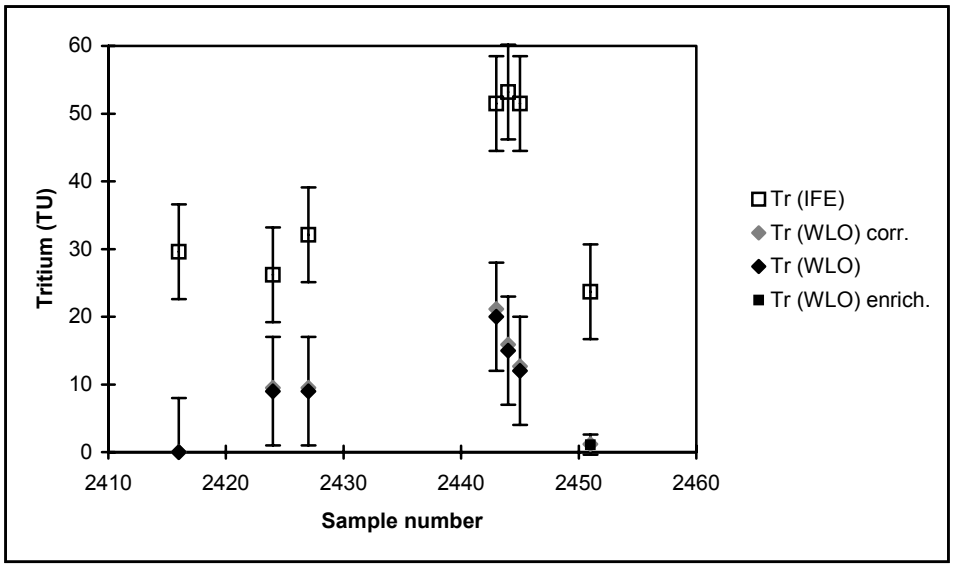


Figure 2-2. *Comparison of original (IFE) and new (WLO) tritium analyses for the sampling period 9604 – 9605.*

a)



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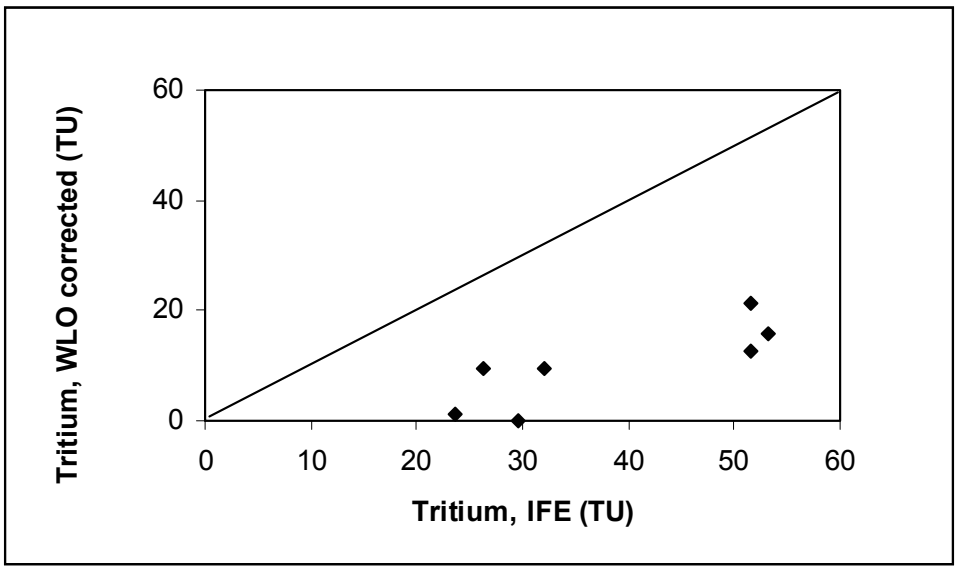
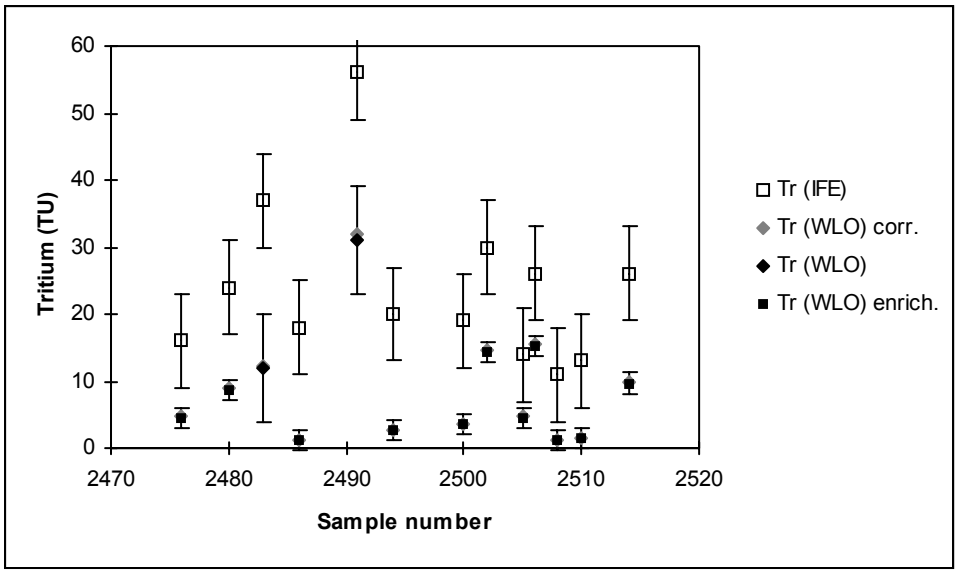


Figure 2-3. *Comparison of original (IFE) and new (WLO) tritium analyses for the sampling period 9709 – 9710.*

a)



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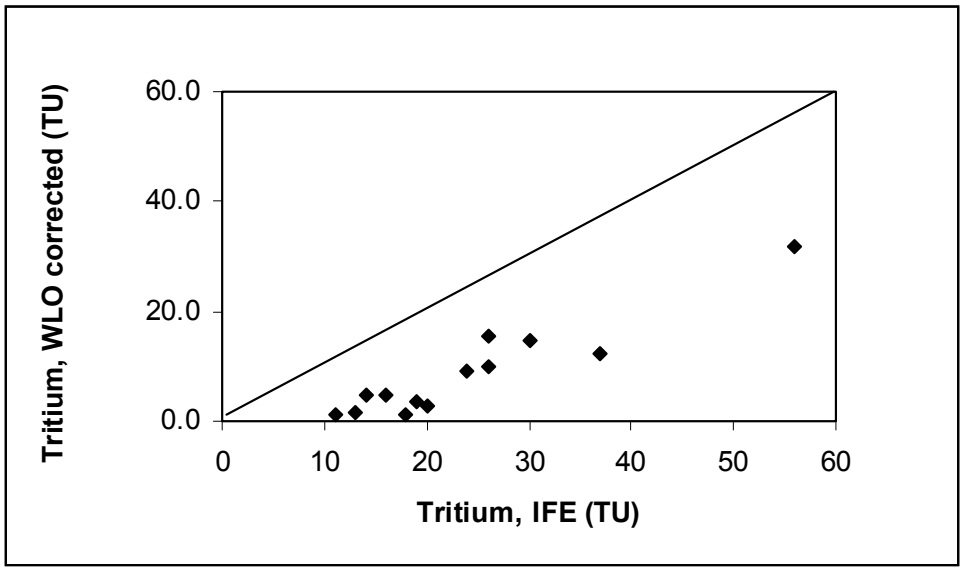
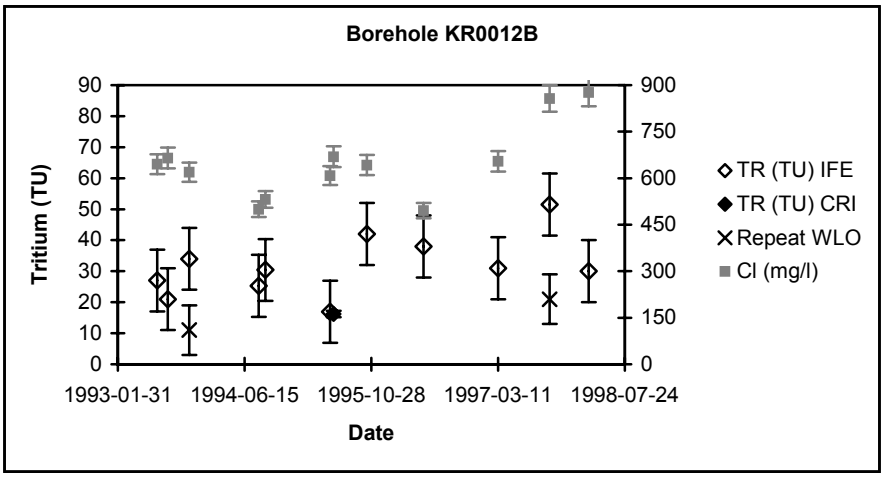


Figure 2-4. *Comparison of original (IFE) and new (WLO) tritium analyses for the sampling period 980302 – 980312.*

a)



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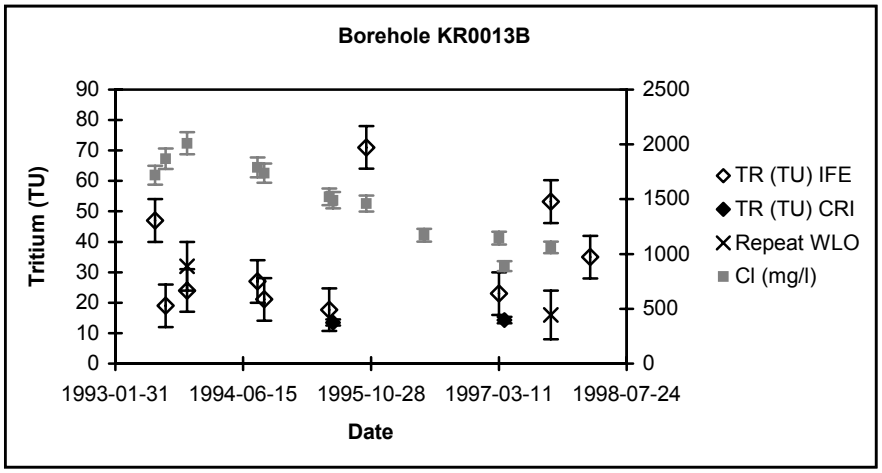


b)

c)

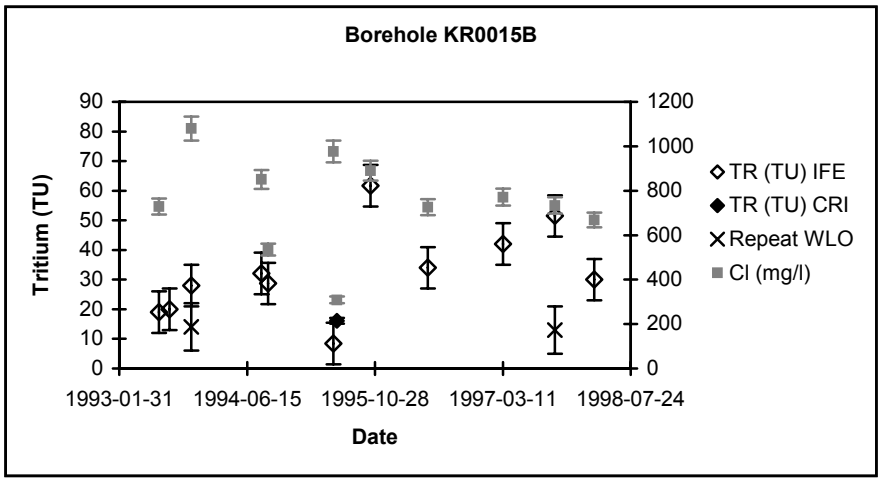
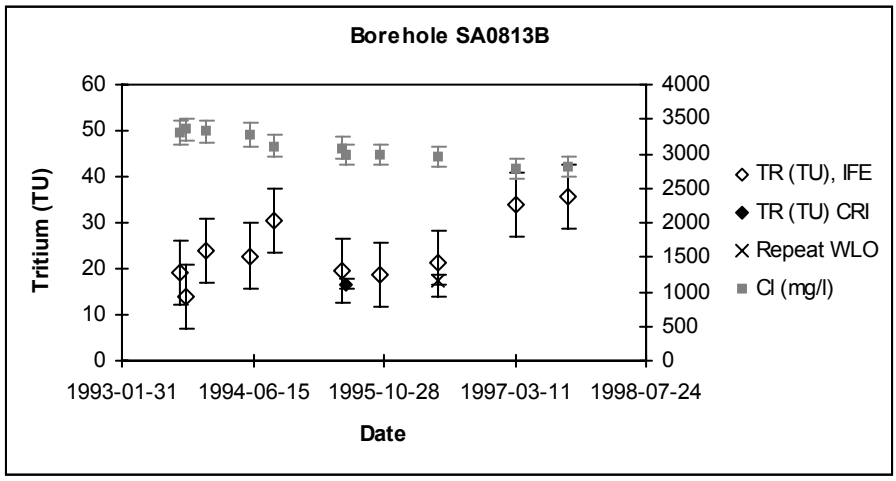


Figure 2-5 a-c. *Tritium and chloride (right axis) contents for selected boreholes.*

d)



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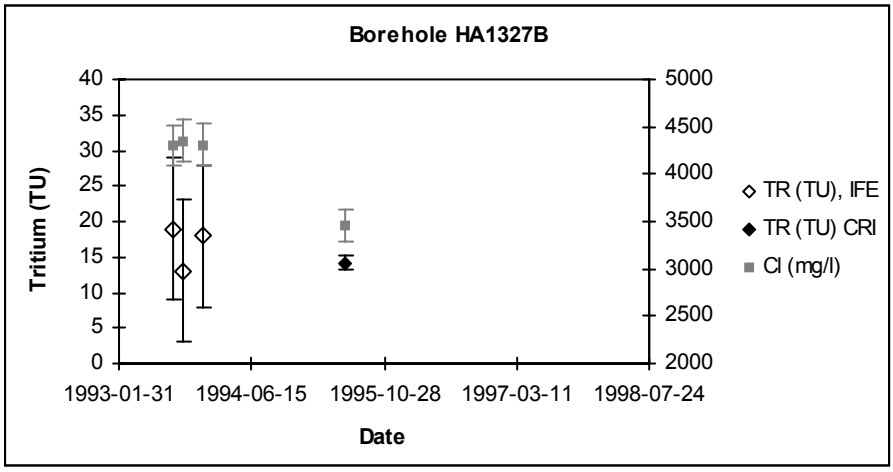

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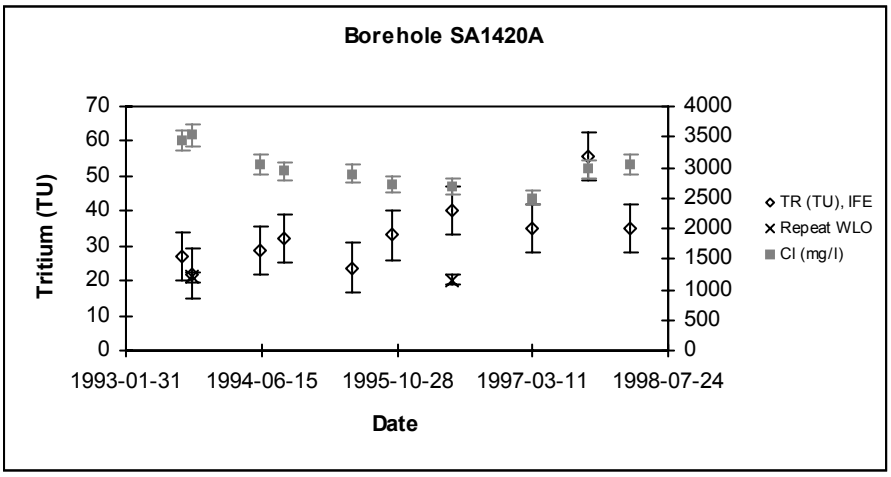
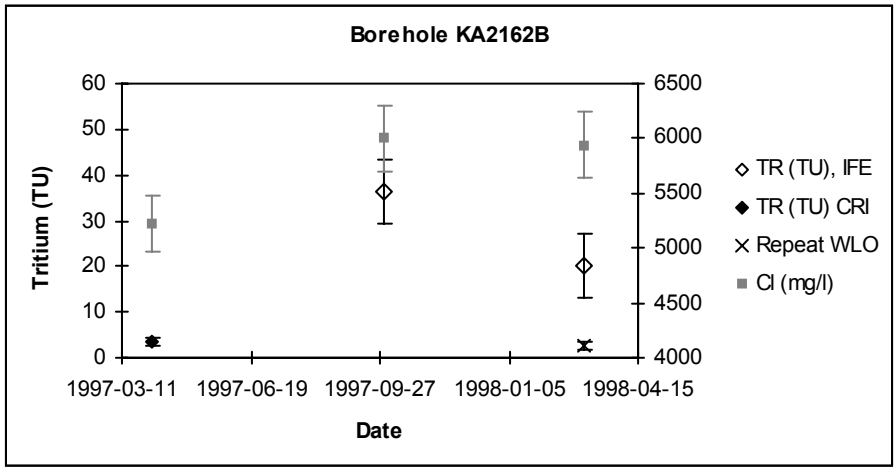
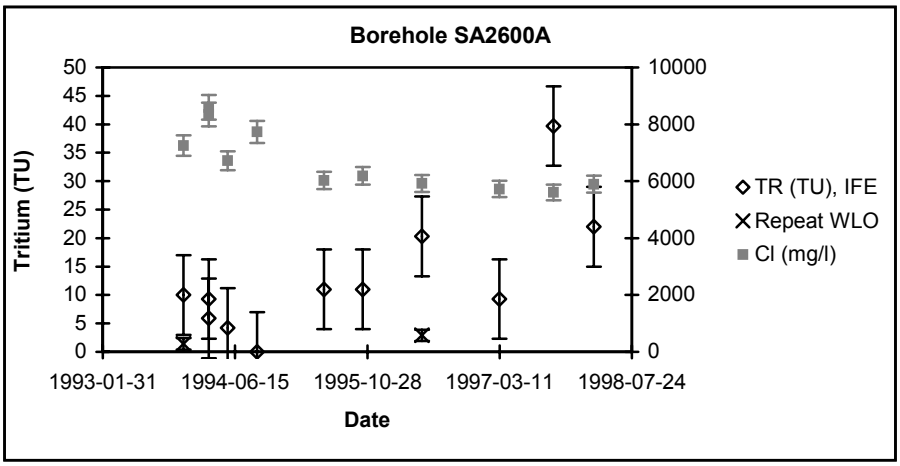


Figure 2-5 d-f. *Tritium and chloride (right axis) contents for selected boreholes.*

g)



h)



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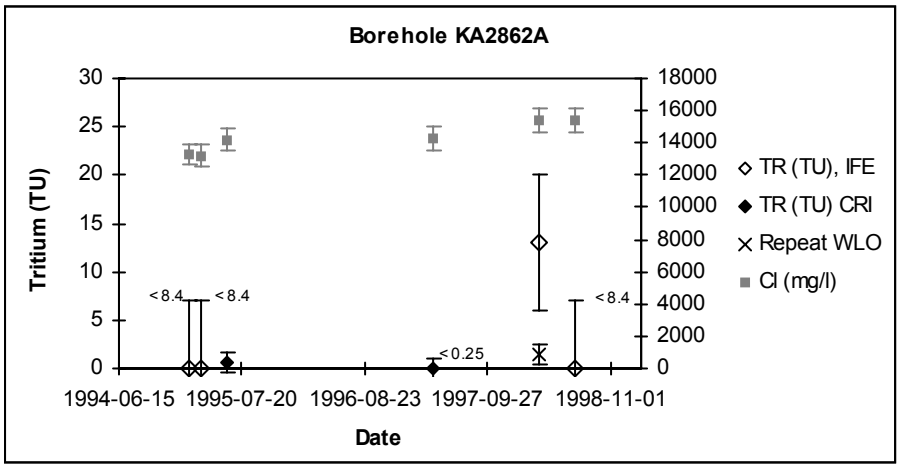
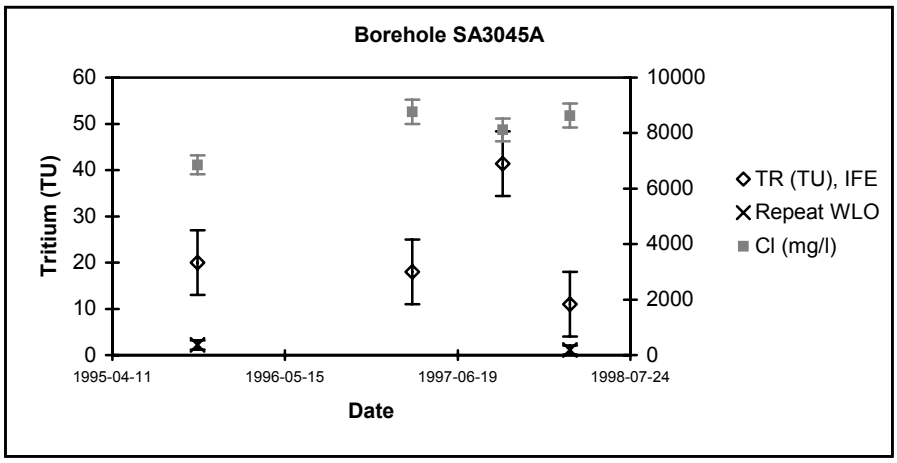


Figure 2-5 g-i. *Tritium and chloride (right axis) contents for selected boreholes.*

j)



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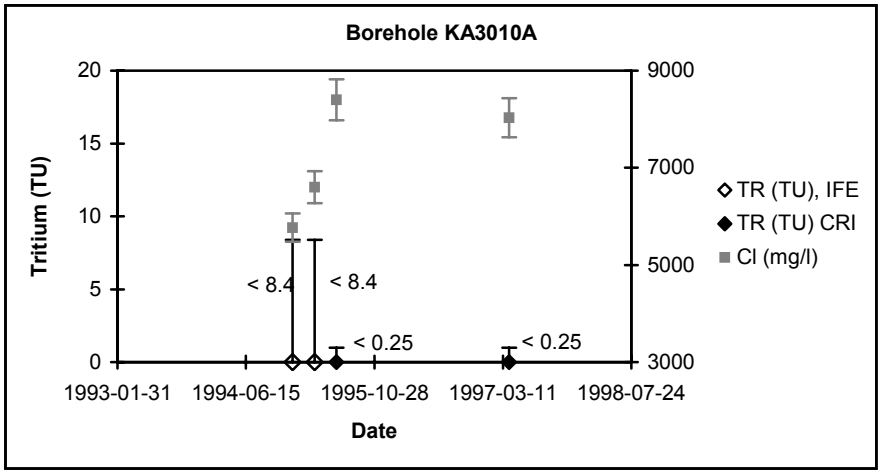

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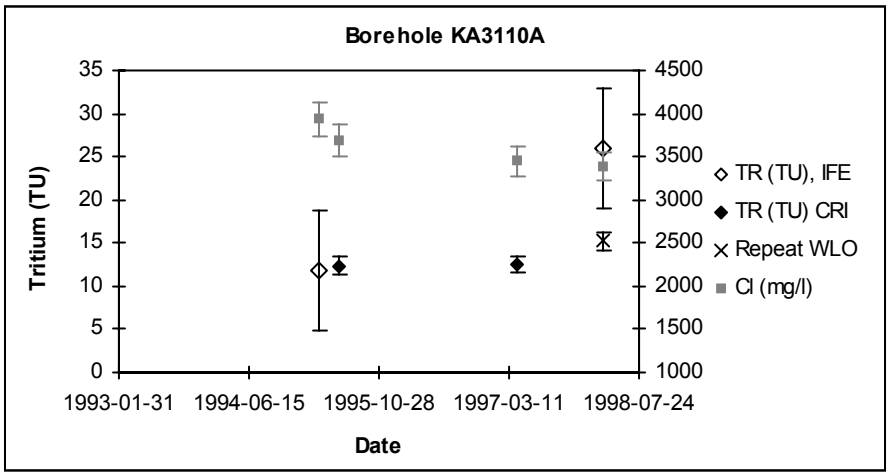

Figure 2-5 j-l. *Tritium and chloride (right axis) contents for selected boreholes.*

m)

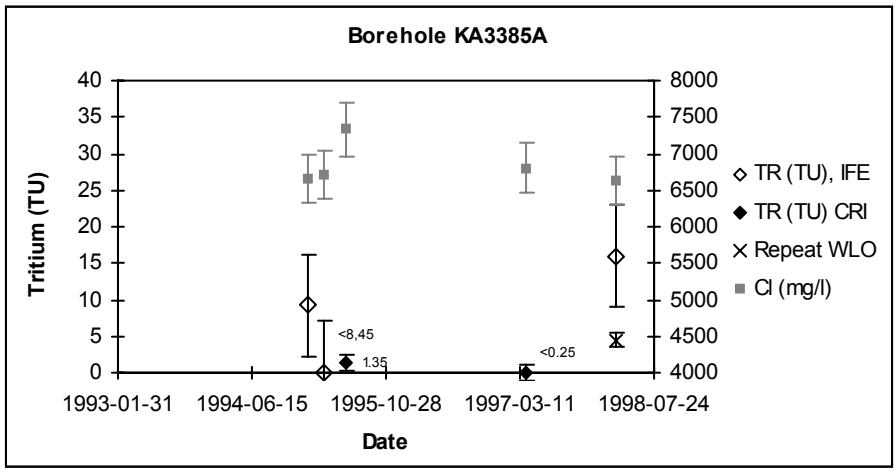

Figure 2-5 m. *Tritium and chloride (right axis) contents for selected boreholes.*

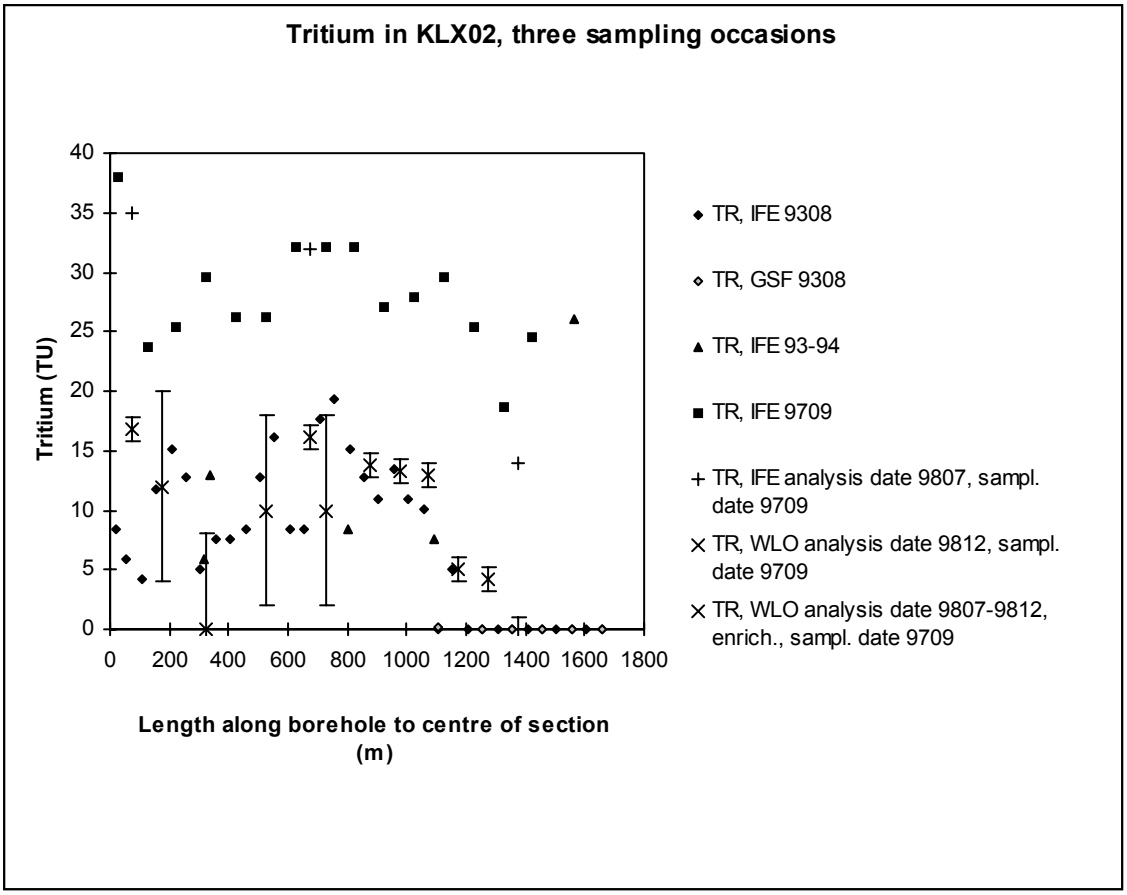

Figure 2-6. *Analyses of tritium from different laboratories from three sampling occasions in borehole KLX02.*

Comments

- In addition to the fact that radioactive decay decreases tritium contents over time, it is likely that some contamination may occur that increases the tritium content because the plastic bottles used for sample storage are not completely diffusion resistant.

- The samples that were sent to the University of Waterloo were originally not intended for tritium analyses and the current routines for tritium sampling were not followed in this case. Therefore, there may be a risk of contamination in connection with sampling.

- The borehole-specific plots in Figure 2-5 show a tendency that reported values from WLO at low levels are somewhat higher than values from CRIEPI and possibly also higher than values from IFE, which may indicate contamination during sampling.

- From Figure 2-5, it appears that tritium levels from WLO analyses vary considerably less from one borehole to another compared with corresponding results from IFE.

- Additional analyses on samples from the "known" boreholes in Figure 2-5 may possibly decrease the uncertainty of existing results but it is probably not possible to identify any trends in the tritium levels.

- Any additional future analyses should primarily be carried out on samples from other boreholes than the ones plotted in Figure 2-5. Samples should be chosen so that at least two new analyses from each borehole/borehole section will be obtained.

- New analyses should be obtained for boreholes included in the monitoring programme.

3 Conclusions

There should be no doubt about that most of the tritium analyses performed by IFE during autumn 1993 to spring 1998 have considerable errors. The errors have apparently no systematic features and do not seem possible to rectify afterwards. These results are, thus, of very limited use.

The problems encountered with tritium analyses show the importance of continuous quality control by using more than one analysis laboratory. This is equally valid for all analysed chemical parameters, SKB's own analyses as well as analyses carried out by other laboratories.