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Objet: Projet minier Niocan - miscalculations and other errors in DA21

I have just reviewed exhibit DA21, "Caractérisation des matériaux du site minier St-Lawrence Columbiun", a report dated May 2002 prepared by Roche for Niocan.

I am writing to inform you of some miscalculations and incorrect inferences in the document.

Background

Earlier this year (February 2002) I pointed out in testimony before the TAQ that Roche had seriously miscalculated all the radioactivity coefficients for the samples cited in the Niocan Environmental Assessment document of 2001. These calculations were carried out for samples of slag [scories], tailings [sandy residues], and barren rocks [stériles].

The miscalculations occurred because Roche simply neglected to include in the calculation the majority of the radioactive isotopes present in the samples, in violation of the procedure that is quite clearly described in the Règlement sur les matières dangereuses.

As a result, all of the reported radioactivity coefficients in the Environmental Assessment Report were too low by a factor of ten or more. In the case of one sample of stérile, for example, Roche had calculated a coefficient of 0,4 when the actual coefficient was more than 4,0 . Roche then used this miscalculation to reach the incorrect conclusion that that particular sample of stérile was below the regulatory limit and should therefore not be classified as a dangerous radioactive material (defined as a material for which the radioactivity coefficient exceeds 1).

"Radioactivity Coefficient" Calculations in DA21

In this new report DA21, the Roche team has accepted the validity of my earlier critique and has adopted my spreadsheet format for calculating the radioactivity coefficient, utilizing the complete decay chains of two of the three primordial actinides present in the ore body -- namely uranium-238 and thorium-232.

For some unknown reason, Roche (DA21) still does not include the entire decay chain of uranium-235. Although this may seem a relatively small oversight, it is not in conformity with the procedure as outlined in the regulations.

Example: Sample of Stérile taken in 2002

Table 1 in Annex III of DA21 shows how the Roche team calculates a coefficient of 0,9 for a sample of "stérile" from the SLC site, taken in 2002.

On page 4 of the text, the Roche team states "Ainsi, cet échantillon de stérile montre un coefficient d'activité radioactive totale légèrement inférieur au critère de 1,0 présenté dans le Règlement sur les matières dangereuses, alors que l'échantillon analysé en 1999 montrait un coefficient de 4,4." [The latter sample was in fact originally reported by Roche, in the EA Report, as having a coefficient of 0,4.]

Three Obvious Errors: Improper Procedures

There are three obvious errors in Table 1: (1) the U-235 decay chain has been omitted, (2) the measured activity of Pa-234m has not been used in the calculation, and (3) there is no calculation of the radiation coefficient for Rn-222, Po-218, or K-40.

When these corrections are made, the increments to the radioactivity coefficient are:

- (1) by including the U-235 decay chain, increase = 0,023 (from 0,902 to 0,925);
- (2) by using the measured value for Pa-234m, increase = 0,030 (from 0,925 to 0,955);
- (3) by calculating ratios for Rn-222, Po-218, or K-40, increase = 0,020 (from 0,955 to 0,975).

Correcting these obvious errors leads to a radioactivity coefficient of 0,975, which is getting quite close to the regulatory limit (especially in light of the $\pm 10\%$ accuracy level indicated by Roche!).

For the calculation, see attachment 1 to this letter. Note that I have lowered the estimated activity of polonium-212 to 80 Bq/kg, in accordance with the fact that the activities of polonium-212 and thallium-208 are generally in the ratio of 2 to 1. This lowers the radioactivity coefficient slightly from 0,9751 to 0,9743 -- a drop of 0,0008.

Additional Errors: Incorrect Inferences

The largest contributor to my 0,974 figure comes from Pa-234m, whose activity was measured as 512 becquerels per kilogram ($\pm 10\%$). For some unexplained reason, the actual measurement was disregarded by Roche in favour of the fictitious estimated value of 393 Bq/kg. Any well-trained scientist or mathematician would ask why this should be considered a reasonable thing to do. (Especially since it is not in accordance with the procedure laid down in the Règlement.) I have chosen to use the measured values wherever they exist, because that is the correct procedure.

If the 512 Bq/kg of Pa-234m is correct, or even approximately correct (in the range 460 to 563, using the $\pm 10\%$ specified in the table), then there must also be 460 to 563 Bq/kg EACH of U-238, Th-234, and U-234. Why? Because the stérile is 20 years old, while Th-234 and Pa-234m have half-lives of less than a month. Unless there is 460 to 563 Bq/kg of U-238 to replenish the supply of Th-234 and Pa-234m, these short-lived isotopes could not possibly exist at such activity levels in the sample.

Uranium-238 --> Thorium-234 --> Protactinium-234m --> Uranium-234

Moreover, U-234 is chemically inseparable from U-238 under natural conditions: the ratio between their masses is immutable, and since they belong to the same decay chain, they must maintain the same activity level.

Given the half-lives of the three isotopes U-238, Th-234, Pa-234m, it is clear that even if the radioactive equilibrium were broken at some point in time due to weathering or leaching, it wouldn't matter much -- because that equilibrium would be re-established again within 5 or 6 months. Meanwhile the U-234 activity will always match the U-238 activity for reasons stated in the previous paragraph.

Thus there is every reason to believe that U-238, Th-234, Pa-234m, and U-234 all have equal levels of activity in this sample, and those levels are at least 460 Bq/kg.

Using the lowest value of 460 Bq/kg for all four, we get a radioactivity coefficient of 1,01 [attachment 2]. If we use the measured value of 512 for Pa-234m -- as required by the Règlement -- and use 460 for the others, we get a coefficient of 1,025. [attachment 3] Thus the radioactivity coefficient for this sample of stérile, using the procedure required by the Règlement, is at least $2\frac{1}{2}$ percent higher than the regulatory limit used to define a hazardous radioactive material.

If we use the higher value of 563 Bq/kg for U-238, Th-234, and U-234, and the measured value of 512 for Pa-234m, we get a coefficient of 1,10 -- ten percent over the regulatory limit. [attachment 4]

Thus the authors of the Report have, through miscalculation and incorrect inference, as well as a failure to follow the procedure outlined in the Règlement, arrived at a conclusion which is opposite to the correct conclusion.

Further Confirmation

Further confirmation that 460 Bq/kg is a reasonable figure for the U-238 activity comes from looking at the measured U-235 activity in Table 1, which is 19,7 Bq/kg. It is well known that natural uranium always consists of 99,7 percent U-238 by weight, and 0,7 percent U-235 by weight. This ratio is unalterable by any chemical or physical process that may be encountered in the Oka environment.

The activity of U-235 in any sample of uranium-bearing material from a natural setting, such as the Oka area, must be 4,7 percent of the activity of U-238. [Since U-235 and U-238 belong to different decay chains, their activity levels are not at all equal.] Thus, using the 19,7 Bq/kg measurement for U-235 reported in Table 1 of DA21 leads us to infer the activity of U-238 as $19,7/0,047 = 419$ Bq/kg. Notice that 460 Bq/kg is within 10% of 419 Bq/kg, and is thus within the stated margin of error.

If we use 512 for Pa-234m and only 419 for U-238, Th-234, and U-234, we get a coefficient of 0,992 [attachment 5]. However, it is scientifically impossible to have 512 Bq/kg of Pa-234m, or anything within a 10% range of that value, if there are only 419 Bq/kg of U-238 and Th-234. This logical conundrum is further exacerbated by using 393 Bq/kg as the Roche authors do for all four isotopes.

Either the measured Pa-234m and U-235 values are wrong, and should be re-done, or the estimated activities of U-238, Th-234, and U-234 are way too low. As reported in Table 1, these figures are internally inconsistent.

Implications

There are some important implications to be drawn.

First, it appears that these stériles are more likely than not to fall within the Quebec Government's classification of dangerous radioactive materials, and therefore they should not be used for construction of roads or otherwise be made available for unrestricted use.

Second, the discrepancy between the measured values of Pa-234m [512 Bq/kg] and the measured values of radium-226 [393 Bq/kg], lead-214 [390 Bq/kg], and bismuth-214 [370 Bq/kg], suggests that about 23 percent of the radium-226 that was originally present in the stérile (due to radioactive equilibrium) has escaped into the environment from the stérile.

The possibility that such a release into the environment can occur in only two or three decades underscores the necessity of treating the stériles as potentially hazardous materials and protecting the environment from them.

Third, it appears desirable for the Ministry of Environment to maintain an independent expertise in the field of low-level radioactivity to ensure that regulations are properly followed.

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ATTACHMENT 1: CALCULATION OF RADIOACTIVE COEFFICIENT
SAMPLE OF SLC STERILES 2002 - CORRECTION TO DA21

THORIUM SERIES		C	A	C/A
Th-232		Inferred	Reg. Limit	Ratio
thorium-232		112	4,000	0.028
radium-228		112	40,000	0.0028
actinium-228		110	40,000	0.00275
thorium-228		112	4,000	0.028
radium-224		112	40,000	0.0028
radon-220		112	40,000	0.0028
polonium-216		112	40,000	0.0028
lead-212		125	40,000	0.00313
bismuth-212		110	40,000	0.00275
[2/3] polonium-212		80	40,000	0.002
[1/3] thallium-208		40	40,000	0.001
lead-208		inactive	stable	0
URANIUM SERIES		C	A	C/A
U-238		Inferred	Reg. Limit	Ratio
uranium-238		393	4,000	0.09825
thorium-234		393	4,000	0.09825
protactinium-234		512	4,000	0.128
uranium-234		393	4,000	0.09825
thorium-230		393	4,000	0.09825
radium-226		393	4,000	0.09825
radon-222		380	40,000	0.0095
polonium-218		380	40,000	0.0095
lead-214		390	40,000	0.00975
bismuth-214		370	40,000	0.00925
polonium-214		380	40,000	0.0095
lead-210		380	4,000	0.095
bismuth-210		380	40,000	0.0095
polonium-210		380	4,000	0.095
lead-206		inactive	stable	0
ACTINIDE SERIES		C	A	C/A
U-235		Inferred	Reg. Limit	Ratio
uranium-235		19.7	4,000	0.00493
thorium-231		19.7	4,000	0.00493
protactinium-231		19.7	4,000	0.00493
actinium-227		19.7	4,000	0.00493
thorium-227		19.7	4,000	0.00493
radium-223		19.7	40,000	0.00049
radon-219		19.7	40,000	0.00049
polonium-215		19.7	40,000	0.00049
lead-211		19.7	40,000	0.00049
bismuth-211		19.7	40,000	0.00049
thallium-207		19.7	40,000	0.00049
lead-207		inactive	stable	0
OTHER Primordial		C	A	C/A
K-40		Inferred	Reg. Limit	Ratio
potassium-40		640	400,000	0.0016
SUM OF ALL C/A's		COEFFICIENT =		0.97426

ATTACHMENT 2: CALCULATION OF RADIOACTIVE COEFFICIENT
SAMPLE OF SLC STERILES 2002 - CORRECTION (2) TO DA21

MINIMUM ESTIMATES FOR U-238, Th-234, Pa-234m, U-234
BASED ON MEASURED ACTIVITY OF Pa-234m - LESS 10 percent
(all activity is measured in Bq/kg)

THORIUM SERIES		C	A	C/A
Th-232		Inferred	Reg. Limit	Ratio
thorium-232		112	4,000	0.028
radium-228		112	40,000	0.0028
actinium-228		110	40,000	0.00275
thorium-228		112	4,000	0.028
radium-224		112	40,000	0.0028
radon-220		112	40,000	0.0028
polonium-216		112	40,000	0.0028
lead-212		125	40,000	0.00313
bismuth-212		110	40,000	0.00275
[2/3] polonium-212		80	40,000	0.002
[1/3] thallium-208		40	40,000	0.001
lead-208		inactive	stable	0
URANIUM SERIES		C	A	C/A
U-238		Inferred	Reg. Limit	Ratio
uranium-238		460	4,000	0.115
thorium-234		460	4,000	0.115
protactinium-234		460	4,000	0.115
uranium-234		460	4,000	0.115
thorium-230		393	4,000	0.09825
radium-226		393	4,000	0.09825
radon-222		380	40,000	0.0095
polonium-218		380	40,000	0.0095
lead-214		390	40,000	0.00975
bismuth-214		370	40,000	0.00925
polonium-214		380	40,000	0.0095
lead-210		380	4,000	0.095
bismuth-210		380	40,000	0.0095
polonium-210		380	4,000	0.095
lead-206		inactive	stable	0
ACTINIDE SERIES		C	A	C/A
U-235		Inferred	Reg. Limit	Ratio
uranium-235		19.7	4,000	0.00493
thorium-231		19.7	4,000	0.00493
protactinium-231		19.7	4,000	0.00493
actinium-227		19.7	4,000	0.00493
thorium-227		19.7	4,000	0.00493
radium-223		19.7	40,000	0.00049
radon-219		19.7	40,000	0.00049
polonium-215		19.7	40,000	0.00049
lead-211		19.7	40,000	0.00049
bismuth-211		19.7	40,000	0.00049
thallium-207		19.7	40,000	0.00049
lead-207		inactive	stable	0
OTHER Primordial		C	A	C/A
K-40		Inferred	Reg. Limit	Ratio
potassium-40		640	400,000	0.0016
SUM OF ALL C/A's		COEFFICIENT =		1.01151

ATTACHMENT 3: CALCULATION OF RADIOACTIVE COEFFICIENT
SAMPLE OF SLC STERILES 2002 - CORRECTION (3) TO DA21

MINIMUM ESTIMATES FOR U-238, Th-234, AND U-234
COMBINED WITH MEASURED ACTIVITY OF Pa-234m
(all activity is measured in Bq/kg)

THORIUM SERIES		C	A	C/A
Th-232		Inferred	Reg. Limit	Ratio
thorium-232		112	4,000	0.028
radium-228		112	40,000	0.0028
actinium-228		110	40,000	0.00275
thorium-228		112	4,000	0.028
radium-224		112	40,000	0.0028
radon-220		112	40,000	0.0028
polonium-216		112	40,000	0.0028
lead-212		125	40,000	0.00313
bismuth-212		110	40,000	0.00275
[2/3] polonium-212		80	40,000	0.002
[1/3] thallium-208		40	40,000	0.001
lead-208		inactive	stable	0
URANIUM SERIES		C	A	C/A
U-238		Inferred	Reg. Limit	Ratio
uranium-238		460	4,000	0.115
thorium-234		460	4,000	0.115
protactinium-234		512	4,000	0.128
uranium-234		460	4,000	0.115
thorium-230		393	4,000	0.09825
radium-226		393	4,000	0.09825
radon-222		380	40,000	0.0095
polonium-218		380	40,000	0.0095
lead-214		390	40,000	0.00975
bismuth-214		370	40,000	0.00925
polonium-214		380	40,000	0.0095
lead-210		380	4,000	0.095
bismuth-210		380	40,000	0.0095
polonium-210		380	4,000	0.095
lead-206		inactive	stable	0
ACTINIDE SERIES		C	A	C/A
U-235		Inferred	Reg. Limit	Ratio
uranium-235		19.7	4,000	0.00493
thorium-231		19.7	4,000	0.00493
protactinium-231		19.7	4,000	0.00493
actinium-227		19.7	4,000	0.00493
thorium-227		19.7	4,000	0.00493
radium-223		19.7	40,000	0.00049
radon-219		19.7	40,000	0.00049
polonium-215		19.7	40,000	0.00049
lead-211		19.7	40,000	0.00049
bismuth-211		19.7	40,000	0.00049
thallium-207		19.7	40,000	0.00049
lead-207		inactive	stable	0
OTHER Primordial		C	A	C/A
K-40		Inferred	Reg. Limit	Ratio
potassium-40		640	400,000	0.0016
SUM OF ALL C/A's		COEFFICIENT =		1.02451

ATTACHMENT 4: CALCULATION OF RADIOACTIVE COEFFICIENT
SAMPLE OF SLC STERILES 2002 - CORRECTION (4) TO DA21

MAXIMUM ESTIMATES FOR U-238, Th-234, AND U-234
COMBINED WITH MEASURED ACTIVITY OF Pa-234m
(all activity is measured in Bq/kg)

THORIUM SERIES		C	A	C/A
Th-232		Inferred	Reg. Limit	Ratio
thorium-232		112	4,000	0.028
radium-228		112	40,000	0.0028
actinium-228		110	40,000	0.00275
thorium-228		112	4,000	0.028
radium-224		112	40,000	0.0028
radon-220		112	40,000	0.0028
polonium-216		112	40,000	0.0028
lead-212		125	40,000	0.00313
bismuth-212		110	40,000	0.00275
[2/3] polonium-212		80	40,000	0.002
[1/3] thallium-208		40	40,000	0.001
lead-208		inactive	stable	0
URANIUM SERIES		C	A	C/A
U-238		Inferred	Reg. Limit	Ratio
uranium-238		563	4,000	0.14075
thorium-234		563	4,000	0.14075
protactinium-234		512	4,000	0.128
uranium-234		563	4,000	0.14075
thorium-230		393	4,000	0.09825
radium-226		393	4,000	0.09825
radon-222		380	40,000	0.0095
polonium-218		380	40,000	0.0095
lead-214		390	40,000	0.00975
bismuth-214		370	40,000	0.00925
polonium-214		380	40,000	0.0095
lead-210		380	4,000	0.095
bismuth-210		380	40,000	0.0095
polonium-210		380	4,000	0.095
lead-206		inactive	stable	0
ACTINIDE SERIES		C	A	C/A
U-235		Inferred	Reg. Limit	Ratio
uranium-235		19.7	4,000	0.00493
thorium-231		19.7	4,000	0.00493
protactinium-231		19.7	4,000	0.00493
actinium-227		19.7	4,000	0.00493
thorium-227		19.7	4,000	0.00493
radium-223		19.7	40,000	0.00049
radon-219		19.7	40,000	0.00049
polonium-215		19.7	40,000	0.00049
lead-211		19.7	40,000	0.00049
bismuth-211		19.7	40,000	0.00049
thallium-207		19.7	40,000	0.00049
lead-207		inactive	stable	0
OTHER Primordial		C	A	C/A
K-40		Inferred	Reg. Limit	Ratio
potassium-40		640	400,000	0.0016
SUM OF ALL C/A's		COEFFICIENT =		1.10176

ATTACHMENT 5: CALCULATION OF RADIOACTIVE COEFFICIENT
SAMPLE OF SLC STERILES 2002 - CORRECTION (5) TO DA21

USING U-235 TO ESTIMATE U-238, Th-234, AND U-234
COMBINED WITH MEASURED ACTIVITY OF Pa-234m
(all activity is measured in Bq/kg)

THORIUM SERIES		C	A	C/A
Th-232		Inferred	Reg. Limit	Ratio
thorium-232		112	4,000	0.028
radium-228		112	40,000	0.0028
actinium-228		110	40,000	0.00275
thorium-228		112	4,000	0.028
radium-224		112	40,000	0.0028
radon-220		112	40,000	0.0028
polonium-216		112	40,000	0.0028
lead-212		125	40,000	0.00313
bismuth-212		110	40,000	0.00275
[2/3] polonium-212		80	40,000	0.002
[1/3] thallium-208		40	40,000	0.001
lead-208		inactive	stable	0
URANIUM SERIES		C	A	C/A
U-238		Inferred	Reg. Limit	Ratio
uranium-238		419	4,000	0.10475
thorium-234		419	4,000	0.10475
protactinium-234		512	4,000	0.128
uranium-234		419	4,000	0.10475
thorium-230		393	4,000	0.09825
radium-226		393	4,000	0.09825
radon-222		380	40,000	0.0095
polonium-218		380	40,000	0.0095
lead-214		390	40,000	0.00975
bismuth-214		370	40,000	0.00925
polonium-214		380	40,000	0.0095
lead-210		380	4,000	0.095
bismuth-210		380	40,000	0.0095
polonium-210		380	4,000	0.095
lead-206		inactive	stable	0
ACTINIDE SERIES		C	A	C/A
U-235		Inferred	Reg. Limit	Ratio
uranium-235		19.7	4,000	0.00493
thorium-231		19.7	4,000	0.00493
protactinium-231		19.7	4,000	0.00493
actinium-227		19.7	4,000	0.00493
thorium-227		19.7	4,000	0.00493
radium-223		19.7	40,000	0.00049
radon-219		19.7	40,000	0.00049
polonium-215		19.7	40,000	0.00049
lead-211		19.7	40,000	0.00049
bismuth-211		19.7	40,000	0.00049
thallium-207		19.7	40,000	0.00049
lead-207		inactive	stable	0
OTHER Primordial		C	A	C/A
K-40		Inferred	Reg. Limit	Ratio
potassium-40		640	400,000	0.0016
SUM OF ALL C/A's		COEFFICIENT =		0.99375