Biopolishing Process for ^{226}Ra Removal

M. Kalin, M.P. Smith, Boojum Research Limited
M. Wittrup, Cameco Corporation

ABSTRACT

^{226}Ra is weathered from waste rock and fluxing from sediments, which have been deposited downstream from the former Rabbit Lake ore deposit in the Link Lake drainage basin in northern Saskatchewan. Lower Link Lake, below the effluent discharge point is populated with *Nitella flexilis*, an algal species of the Characeae with a high affinity for ^{226}Ra. The use algae to contain ^{226}Ra within the drainage basin is evaluated, through the establishment of an algal population in Upper Link Lake. This lake is void of algae and located upstream from the effluent discharge point. As concentrations of ^{226}Ra in the algal biomass were determined as high as 60 Bq/g dry weight, it could be expected that a healthy algal population growing in this location would reduce ^{226}Ra load from the sediments of former Rabbit Lake to the water in Upper Link Lake.

In 1989, 12 tonnes (fresh weight) of *Nitella* were transplanted to Upper Link Lake. The development of the algal population was monitored over five growing seasons. After five years the algal biomass has increased 13 times since transplanting. The growth dynamics of the algal populations suggest, that during one growing season, due to continued growth and decay the population is replaced once. The standing biomass of *Nitella* contained a total of 1.0×10^8 Bq of ^{226}Ra. This is based on estimates of total biomass present in the lake and its concentrations. Load calculations indicate that the algal population retains about 3.9×10^8 Bq of ^{226}Ra in the biomass and in the sediment. This quantity represents about 65% of the annual loading arriving at the discharge point. The algal populations growing over the former Rabbit Lake sediments reduce the ^{226}Ra flux from this area into Upper Link Lake water. Monitoring data at the discharge point, show a reduction in the seasonal fluctuations of ^{226}Ra concentrations after 5 years of growth. These reductions are evidence of the effectiveness of the algae in retaining ^{226}Ra in the drainage basin.

INTRODUCTION

Contaminant removal using natural water-cleansing processes might represent an alternative to perpetual water treatment at the time of decommissioning a mining operation. Such contaminant removal is possible when specific ecosystem characteristics are created within the waste management area and if the contaminant to be removed is geochemically amenable to natural removal. If these two basic conditions are fulfilled, through ecological engineering measures, chemical water treatment requirements can be reduced or replaced.

The Rabbit Lake drainage basin was identified as an ecosystem, where within the waste management area, algal populations are present, which have the ability to remove ^{226}Ra from the water column. These algae grew in the lakes of the drainage basin, prior to commencement of the mining activities. During operation of the mine water quality monitoring data indicated that the concentration of ^{226}Ra decreases upon passage through a bog located between Upper and Lower Link Lake.

Nitella flexilis, a species of attached algae dominates submerged vegetation in this bog. High concentrations of ^{226}Ra in the algal biomass sampled from the bog, confirmed the algae reported affinity for this contaminant. These algae are known for their affinity for ^{226}Ra, as well as for other radionuclides (^{210}Pb, ^{90}Sr, ^{137}Cs, ^{106}Ru), alkaline earth elements and metals (Mudroch & Capobianco, 1978; Dusauskene & Polikarpov, 1978; Harding & Whitton, 1978).

It was proposed to utilize these algae to control contaminant release from Upper Link Lake to the lower parts of the drainage basin. As part of the restoration measures for the decommissioning of this drainage basin, the recolonisation of Upper Link Lake with these algae was evaluated along with a quantification of the capacity of contaminant retention and water cleansing capacity.

The distribution and growth of the algae

Between 1988 and 1989, the distribution and growth of the algae, *Chara vulgaris* and *Nitella flexilis*, both members of the Charophyte family, were studied in the Rabbit Lake drainage basin (Kalin and Smith 1988). Extensive populations of *Nitella* were growing in the bog and in Lower Link Lake, but the algae were entirely absent in Upper Link Lake. The discharge of the Rabbit Lake sediments to Upper Link Lake altered water depth and covered any vegetation with solids. Sediment cores collected during hydrological investigations of Upper Link Lake contained oospores of the algae, suggesting its presence prior to the onset of mining activities.

Growth of the algae was tested in experimental plots in Upper Link Lake through transplanting biomass from Lower Link Lake. Biomass production ranged between 20 g dry weight.m^{-2} to 560 g.m^{-2} per year. It was estimated, based on the concentrations of ^{226}Ra (^{10}Bq.g^{-1}), that a 10 ha underwater algal meadow, with a biomass density of about 250 g *Nitella*.m^{-2} could potentially reduce the total annual ^{226}Ra load by 2.5 x 10^8 Bq per year at the discharge point, referred to as Sedimentation Dam (station W15) at the outflow of Upper Link Lake.

With these contaminant retention estimates at hand, the utilisation of the algae as a natural cleansing process as part of the decommissioning plan was further pursued. In 1988, *Nitella* biomass was transplanted from lower Link Lake to Upper Link Lake. The algae grew very well and a larger scale transplant operation was initiated in 1989.

Prior to 1993, the amount of *Nitella* biomass outside the transplant zones was negligible. However, in 1993 it is estimated that in some transplant zones the amount of biomass is 7 times that transplanted in 1989. It required four growing seasons for the new transplanted *Nitella* population's total biomass to make large net gains compared to the initial biomass transplanted on the submerged parts of the former Rabbit Lake sediments and in other parts of Upper Link Lake.

During the 1989 - 1993 period, the *Nitella* weight gain was approximately 4 tonnes dry weight, or almost 8 times the original biomass transplanted.

During one year of growth it is estimated that 1 tonne of biomass is lost due to fragmentation/transport of parts of the algae or decay, leaving a biomass approximately 6 times that originally transplanted.

In 1994 it appears that the colonization phase (spread and infill) of the *Nitella* population development in two of the transplant zones is almost finished, given the near-complete coverage of the available surface area. Thus in five years extensive colonization had taken place in Upper Link Lake.

Chemical characteristics of the Rabbit Lake drainage basin

From an ecological point of view, the water characteristics of the drainage basin are essentially those of a healthy freshwater ecosystem. The pH of the water is neutral, ranging from pH 7 to 8. The sulphate concentrations, although they decrease in the outflow of Lower Link Lake (W25) from an average of around 50 $mg.L^{-1}$ (W5) to 20 $mg.L^{-1}$, are in the mid range for fresh water in Central Canada (not detectable to 77.3 $mg.L^{-1}$ based on 4696 samples; CCREM, 1987). Chloride concentrations are typical for fresh water, ranging throughout the drainage basin between 1 and 5 $mg.L^{-1}$. The concentration range for Central Canada for chloride is given as <0.1 to 450 $mg.L^{-1}$ (CCREM, 1987). Specific conductivities of 100 to 200 $\mu S.cm^{-1}$ are somewhat higher than in other northern lakes in the region (McArthur River area, 5 to 43 $\mu S.cm^{-1}$; Cigar Lake area, < 6 to 27 $\mu S.cm^{-1}$; M. Wittrup pers.com., 1994).

^{226}Ra with a mean annual average of about 0.3 $Bq.L^{-1}$ is elevated. These concentrations are above natural background concentrations, which are reported as < 0.005 $Bq.L^{-1}$ for Wollaston Lake, <0.005 to 0.06 $Bq.L^{-1}$ in the McArthur River area (M.Wittrup, pers. com., 1994), and to average across Canada from 0.008 $Bq.L^{-1}$ to 0.1 $Bq.L^{-1}$ (Buchnea & van der Vooren, 1985). Average uranium concentrations in the Link Lake drainage basin are elevated above the Link Lakes, with 3 to 4 $mg.L^{-1}$ and drop to a range of 0.2 to 0.4 mg.L^{-1} at Lower Link Lake outflow. Natural background ranges in the McArthur River area from < 0.05 to 2.3 μg.L^{-1} (M. Wittrup, pers. com. 1994) and, in non-mineralized areas across Canada, from 0.04 to 1.4 μg.L^{-1} (Buchnea and van der Vooren 1985). These elevated concentrations of both ^{226}Ra and U are targeted to be removed in Upper Link Lake utilizing algal removal.

Contaminant form and its distribution in the drainage basin

The form in which the contaminants are present in the water is relevant with respect to their removal. Suspended ^{226}Ra, quantified by the difference in concentrations of ^{226}Ra between whole samples and filtered (3 μm) samples, can be removed through sedimentation of the carrier particles. Carrier particles include unicellular algae and other organic debris which adhere to *Nitella* or other periphyton populations.

For ^{226}Ra the dissolved and suspended concentrations are plotted in Figures 1a for water entering Upper Link Lake (Station W5 , Airport Road) and for the water leaving Upper Link Lake (Station W15, Sedimentation Dam) in Figure 1b. Generally, most ^{226}Ra is present in the dissolved form. On the average, the ratio of dissolved to total ^{226}Ra throughout the drainage basin does not change markedly.

It can be noted, that particulary during spring run-off (Figure 1a and 1b) the ^{226}Ra concentrations increase between W5 and W15. The annual averages are 0.14 to 0.36 Bq.L^{-1} compared to 0.28 -0.5 Bq.L^{-1}. Within Upper Link Lake above the Narrows a further distinction can be made, with respect to the source of ^{226} Ra the annual average concentrations range at the Narrows between 0.29 to 0.57 Bq.L^{-1}. This increase in concentration is due to a ^{226}Ra flux from the sediments of the former Rabbit Lake, located above the Narrows monitoring station in Upper Link Lake.

Fig.1a: W5, Airport Road
[Ra226], 1987 -1994

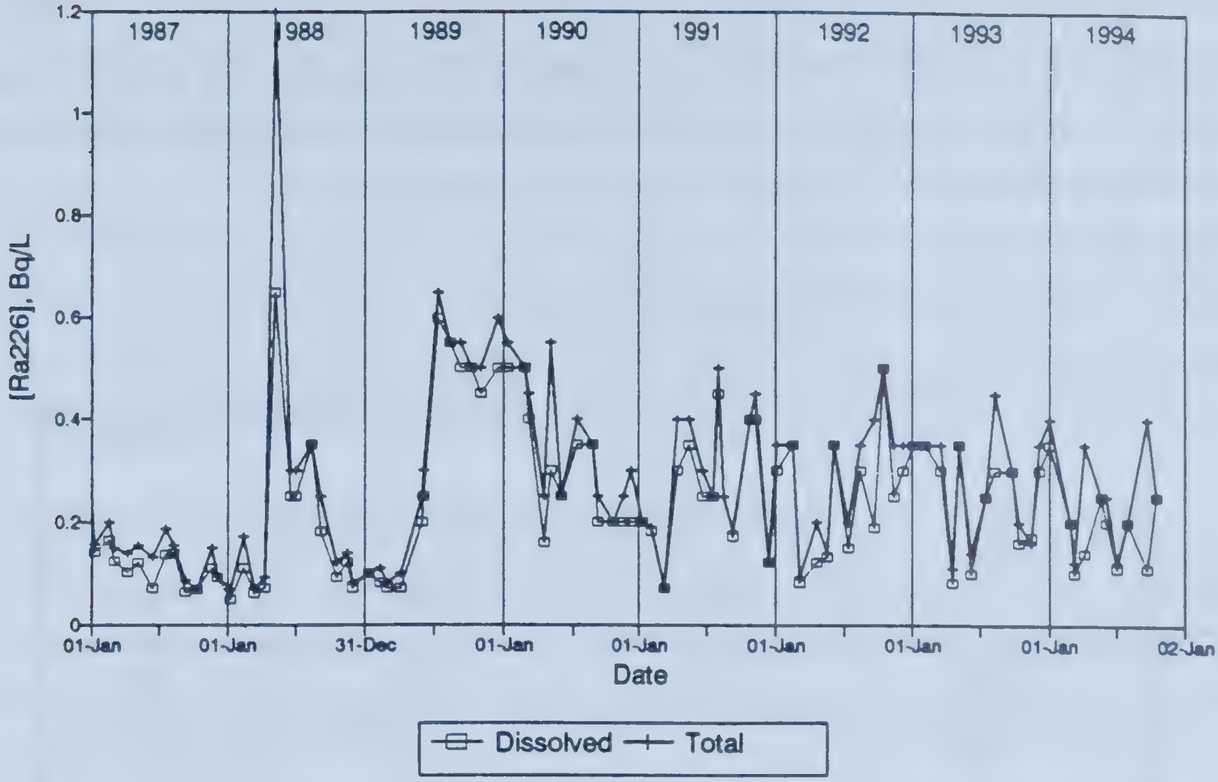
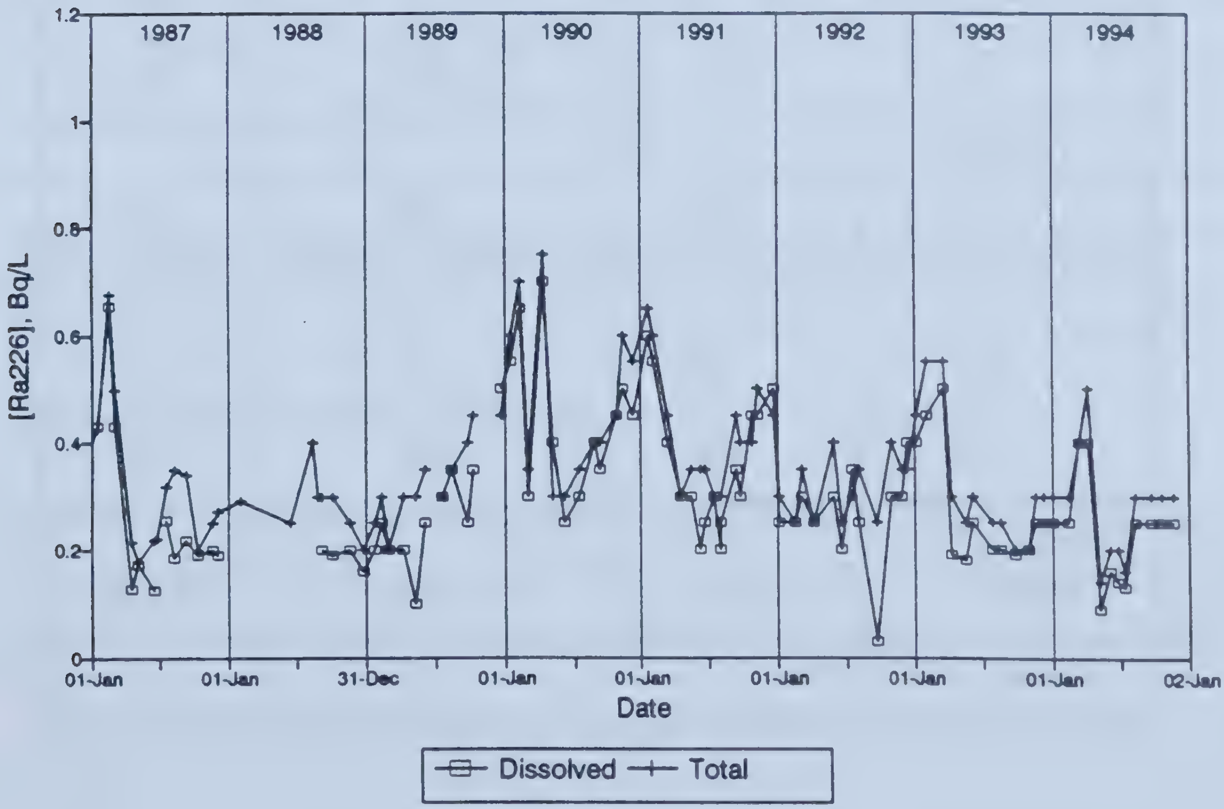


Fig.1b: W15, Sedimentation Dam
[Ra226], 1987-1994



At the outflow of Lower Link Lake a reduction is evident with annual average concentrations ranging from 0.04 to 0.1 $Bq.L^{-1}$.

In Table 1 the annual ^{226}Ra loads are presented for the upper part of the drainage basin. The load is calculated using actual and estimated flows for these stations and the concentrations of ^{226}Ra from the monthly water quality data.

Table 1: Annual ^{226}Ra Load at W5 and W15

Year	W5 Airport Road (MBq)	W15 Sedimentation Dam (MBq)	Difference W15 - W5 (MBq)
1987	129	478	349
1988	492	675	183
1989	272	634	362
1990	179	581	402
1991	236	633	397
Average 1987-91	261	600	339
1992	257	578	321
1993	198	346	148
1994	170	258	88
Average 1992-94	208	394	186
Average, 1987 - 1994			281

The annual loads of ^{226}Ra are summarized for several years for different two locations, water entering Upper Link Lake (W5) and leaving Upper Link Lake at Sedimentation dam. The annual ^{226}Ra loading at W5 between 1987 and 1991 ranged from 129 and 492 MBq of ^{226}Ra, averaging 261 $MBq.year^{-1}$ (Table 1). In the same years 478 and 674 MBq ^{226}Ra arrived at W15 averaging 600 $MBq.year^{-1}$.

The differences in annual load between the stations is calculated by subtracting the annual load at one station from the annual load at another station. These differences indicate whether the load increased from one station to the next (positive difference) or decreased (negative difference).

The average annual ^{226}Ra load increase between W5 and W15 is 339 $MBq.year^{-1}$. This value represents the average annual contribution by the sediments and the drainage basin of the entire Upper Link Lake area. For the period 1992 to 1995, starting 3 years after transplanting of the algae into Upper Link Lake, average annual loads are given in Table 1.

Since that time, the *Nitella* population was starting to grow in such quantities, that its effect on the loading could possibly be evident. Given the three years (1992-1994) average load at Airport Road (W5) with 208 MBq and at Sedimentation Dam (W15) of 394 MBq, a lower load of 394 MBq in these three years compared to the earlier period (1987-1991) of 600 MBq is evident. The differences in load between the two stations over this period is decreasing from year to year, suggesting further that as the algal population increases the flux from the sediments is retained by the algae.

Biological Polishing of Upper Link Lake

The ratio of dissolved and suspended fractions of ^{226}Ra (Figure 1a and 1b) indicate, that essentially removal has to take place of the dissolved fraction of ^{226}Ra. Thus biogeochemical processes including precipitation, coprecipitation or adsorption onto cell walls of zooplankton, phytoplankton, periphyton, and benthic species such as *Nitella* are the likely route of removal for this radionuclide, which is present in trace concentration.

If we assume that the biological material is facilitating these processes take place, it can be expected that the removal for ^{226}Ra displays a seasonal behaviour. Some seasonality is evident in the monitoring data (Fig 1a and Fig 1b). As the biological activity also affects the pH, and pH in turn affects surface adsorption and precipitation processes biological productivity and growth activity in the drainage basin have to be quantified.

Surveys of the drainage basin indicated in 1988 no *Nitella or* any other vascular macrophyte populations were present in Upper Link Lake in contrast to Lower Link Lake. Instead, Upper Link Lake was populated by blue-green (Cyanobacteria) phytoplankton populations in the lake at large, filamentous green (Chlorophyta) algal were growing in the upper part of the drainage basin and in the vegetated parts of the sediment delta. Periodic blooms of algae occurred in Upper Link Lake.

Five major ecological groups of algae populate the Rabbit Lake drainage basin (Table 2). Two types of Cyanobacteria (blue-green algae) cover the sediments (mat like and filamentous forms) in the drainage basin, dominating to different degrees in different sections of the basin. The filamentous forms prevail in ditches and in fast flowing water. Mats cover edges and slower moving portions of ponds and pools. The ^{226}Ra concentrations have been determined for these different groups and are given in Table 2. *Nitella flexilis* contains the highest concentrations followed by the filamentous green and blue green algae.

Table 2: [Ra^{226}] in Algal Biomass in the Rabbit Lake Drainage Basin

Algal Group	Ra^{226}, Bq/g dry weight		
	Range	Avg	n
Filamentous Cyanobacteria	9 - 21	14.0	6
Mat Cyanobacteria	5.5 - 10	8.5	6
Filamentous Chlorophyta	1.4 - 42	14.2	8
Nitella flexilis	5 - 50	20.9	22
(determined between	13 - 43	24.2	10
1988-1992)	1 - 60	13.6	19
	0.6 - 9.5	2.6	16
Cyanobacteria Phytoplankton	0.25 - 0.8	0.5	2

These filamentous and mat species of blue-green algae contain, on average, significant concentrations of ^{226}Ra (14 and 8.5 Bq ^{226}Ra.g dry weight^{-1}, respectively)
The filamentous Chlorophyta (green algae), commonly found during the growing season in the streams passing through the vegetated Delta, on sediments and any available surfaces in Upper Link Lake and Lower Link Lake contain, on average, 14.2 Bq ^{226}Ra.gdw^{-1} .

Nitella biomass transplanted and grown in Upper Link Lake above and below the Narrows contains, on average, relatively high concentrations of ^{226}Ra (20.9 and 24.2 Bq.gdw^{-1}, respectively). Below the Sedimentation dam, in the bog where the ^{226}Ra reductions were originally noted in the monitoring data, *Nitella* biomass typically contains less ^{226}Ra (13.6 Bq.gdw^{-1}), while in Lower Link Lake, biomass contains only 2.6 Bq.gdw^{-1} on average. These concentrations in the biomass suggest, that the sediment / water concentrations of ^{226}Ra are related to those in the biomass.

The final algal group is phytoplankton which, in Upper Link Lake, are dominated by the Cyanobacteria. Species of this group are responsible for the periodic algal blooms observed in Upper Link Lake during the growing season. Algal biomass collected during blooms in Upper Link Lake contained, on average, relatively low concentrations of ^{226}Ra (0.5 $Bq.gdw^{-1}$).

Biological polishing capacity of a contaminant is not only dependant on the total concentration in the biomass, but also on the quantity of biomass produced with in the drainage basin. In biological terms this is referred to as productivity. In Table 3 the growth dynamics of the algal groups, relevant to ^{226}Ra removal are given.

Table 3: Growth Dynamics determined for the Rabbit Lake Drainage Basin

Standing Biomass, g dry weight/m^2	Range
Filamentous Cyanobacteria	379 - 895
Mat Cyanobacteria	95-110
Filamentous Chlorophyta	17 - 68
Nitella flexilis	1 - 1000
Cyanobacteria Phytoplankton	1 - 10 [(2)]
Primary Productivity, g dry weight/m^2 /year [(1)]	**Range**
Filamentous Cyanobacteria	4 - 69
Mat Cyanobacteria	4 - 69
Filamentous Chlorophyta	4 - 69
Nitella flexilis	1.5 - 5
Cyanobacteria Phytoplankton	113

(1) Source: Wetzel, 1983 (2) Estimated; average 2.6 mg dw/L

Depending on growth habit, the specific nutrient availability and growing conditions, each algal group will maintain specific standing biomass densities and growth rates. For instance, depending on the location in the Rabbit Lake drainage basin, *Nitella* standing biomass values can range from 1 to 1000 gdw.m^{-2} (including ash; Table 3).

Typically, higher standing biomass values have been determined in populations in areas of flowing water such the bog and the channel draining Lower Link Lake (up to 1000 gdw.m^{-2}), compared to open lake populations in Lower Link Lake (250 gdw.m^{-2} on average). Since Upper Link Lake is the focus of biological removal of ^{226}Ra, the value, 250 gdw.m^{-2} will be normally employed during projections of standing biomass for mature *Nitella* populations in Upper Link Lake.

The rate of apical growth and basal decay of *Nitella* indicated the productivity of the algae is such that new biomass each year equivalent to the standing biomass at any time. In other words, the *Nitella* biomass growth (productivity) replaced the standing biomass at least once each year. Wetzel (1983) describes this relationship as the Productivity and maximum standing biomass ratio. From his studies of Characean population dynamics, he estimates a higher ratio 3.3 suggesting that the biomass turns over 3.3 times per year, higher then in studies by Kalin et al. 1988.

The standing biomass of populations of filamentous and mat Cyanobacteria and filamentous Chlorophyta was determined for the drainage basin to average for filamentous Cyanobacteria was 250 gdw.m^{-2}, while for the mat populations 100 gdw.m^{-2}, and for the filamentous Chlorophyta, 40 gdw.m^{-2}. Growth rates for these populations can be reliably derived from the literature, as they are not very site specific, and Wetzel's ratio of productivity and maximum standing biomass will be used (Wetzel, 1983) in estimating polishing capacity.

Characteristic to Upper Link Lake is the periodically dense phytoplankton population during the growing season, dominated by Cyanobacteria. Total suspended solids concentrations range from 1 to 10 mg.L^{-1} (1987-1992), of which a large fraction is likely phytoplankton. Assuming an average depth of 1.4 m for Upper Link Lake, then the standing biomass of phytoplankton population averaged 3.6 gdw.m^{-2} (average TSS= 2.56 mg.L^{-1}, 1987 to 1992 Cameco monitoring data). From a summary of Productivity: Maximum Biomass ratios by Wetzel (1983), the ratio for phytoplankton averages 113 g/dry weight/m2/year (Table 3).

Overall, these estimates indicate that while *Nitella* populations can maintain relatively high perennial standing biomass, their productivity is low compared to the other algal groups in the Rabbit Lake drainage basin.

The biological polishing capacity for ^{226}Ra

A primary objective of the application of Ecological Engineering in the Rabbit Lake drainage basin is to reduce the total ^{226}Ra load leaving Upper Link Lake. With the biological growth data at hand, it is now possible to derive estimates of the expected retention of ^{226}Ra in Upper Link Lake.

The results of early studies of ^{226}Ra concentrations in plant materials in 1988 indicated that *Nitella flexilis* biomass accumulated higher concentrations (up to 25 Bq.gdw^{-1}) than any other aquatic plant population and that *Nitella* populations maintain a dense perennial standing biomass (250 gdw.m^{2}) covering the sediment, with the biomass renewing itself at least once per year (Kalin et al., 1988).

Further sampling and analyses of *Nitella* biomass between 1989 and 1992 indicated that even higher ^{226}Ra concentrations than originally determined are present in *Nitella* biomass. Up to 60 Bq.gdw^{-1} was determined in unwashed *Nitella* biomass, analyzed with particulates still trapped in the biomass (bog sampled in April 1990). Meanwhile, concentrations of ^{226}Ra in washed *Nitella* in samples from Upper Link Lake average 21 Bq.gdw^{-1} (above Narrows) and 24 Bq.gdw^{-1} (below Narrows).

Using the August 1993 total standing biomass estimates for *Nitella* in Upper Link Lake (Transplant Zones 1 and 2, 4.9 tonnes dry weight) and the above Narrows average ^{226}Ra content, this biomass currently contains a total of 1.02 x 10^{8} Bq of ^{226}Ra.

The *Nitella* population's total growth, or annual productivity, must exceed the maximum observed standing biomass to compensate for death and decay of older biomass. Wetzel (1983) suggests that the Total Productivity: Maximum Biomass ratio for Characean populations is approximately 3.3, or in other words, the *Nitella* population renews itself 3.3 times each year. If this value is used, a total of 3.9×10^8 Bq ^{226}Ra will be taken up by the *Nitella* population at its current size over the course of a year, a quantity of ^{226}Ra equivalent to the current annual contribution by the former Rabbit Lake sediments on the delta in Upper Link Lake.

Nitella is not the only algal population in Upper Link Lake; blue-green (Cyanobacteria) algal populations are usually dense and can form blooms during the growing season. If an average phytoplankton biomass of 3.6 gdw.m^2 is used, and contains, on average, 0.5 Bq $^{226}Ra.gdw^{-1}$, then the phytoplankton population harbours, on average, only 4.0 x10^5 Bq ^{226}Ra. However, according to Wetzel (1983), phytoplankton have, on average, a Productivity: Maximum Biomass ratio of 113, and over the course of a year, 4.5 x10^7 Bq of ^{226}Ra may be adsorbed onto phytoplankton cells. Most of this biomass will settle to the bottom and join the sediments of Upper Link Lake and the areas downstream.

Filamentous green (Chorophyta) algae have been observed to produce visible biomass during the growing season over the bottom of Upper Link Lake and attach to any available substrate. While the standing biomass of these filamentous green algae are minor, and may amount to only 1 gdw.m^2 during these periods, they contain 14 Bq $^{226}Ra.gdw^{-1}$. The total population could then harbour 3.1×10^6 Bq ^{226}Ra. Using a Productivity: Maximum Biomass ratio of 30, over the course of a year, 9.2×10^7 Bq ^{226}Ra may be adsorbed onto the filamentous green algal population.

CONCLUSION

The projection (Kalin et al. 1988) that the *Nitella* population will spread in Upper Link Lake to cover a total of 12 hectares (Kalin et al., 1988) still appears valid, given the growth and spreading pattern to date. If the approximately 9 ha of lake area with depths between 0.6 and 1.6 m support growth of 250 $gdw.m^{-2}$ *Nitella*, and the 3 ha of lake area with depths between 1.6 to 1.8 m support growth of 100 $g.m^{-2}$ *Nitella*, then the projected total *Nitella* biomass will be 25.5 t of *Nitella* dry weight. Using the average ^{226}Ra concentration in Upper Link Lake *Nitella* (22.6 $Bq.gdw^{-1}$), this *Nitella* population will harbour 5.8×10^8 Bq of ^{226}Ra, an amount nearly equivalent to the 1987-1992 average Upper Link Lake ^{226}Ra loading of 6×10^8 Bq of ^{226}Ra per year. This estimate may be somewhat conservative, as it does not account for possible reductions in net ^{226}Ra flux from Upper Link Lake sediments once the *Nitella* population covers the full 12 ha, and it assumes a *Nitella* biomass turnover of only once per year.

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ENVIRONMENTAL MANAGEMENT FOR MINING

Proceedings of the 19th Annual Meeting of the Canadian Land Reclamation Association/ Association Canadienne de Réhabilitation des Sites Dégradés (CLRA/ACRSD)

October 25-27, 1995
Saskatoon, Saskatchewan