TECHNICAL ARTICLE

Analytical Framework for a Risk-based Estimation of Climate Change Effects on Mine Site Runoff Water Quality

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Abstract A conceptual analytical framework was developed for the risk-based estimation of climate change effects on mine waste runoff water quality. The modeling approach incorporates temporal variability in both precipitation and temperature using trend analyses from historical datasets and the resulting effects on water balances and geochemical weathering rates. A case-study method was used to develop and present the model for regions near the city of Yellowknife, Northwest Territories, Canada. Timeresolved relative precipitation and weathering factors can be calculated using climate data and site-specific estimates regarding mine waste properties. Interpretation of these factors allows an assessment of when the period of highest analyte concentrations are expected to occur under a given risk scenario over a modeling timeframe, whether low precipitation or warming effects are more important in determining water quality issues, and the relative magnitudes of water quality under differing risk scenarios.

Keywords Climate change ·

Geochemical weathering rates \cdot Mine waste \cdot Tailings and waste rock runoff \cdot Water quality

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Introduction

Estimating future water quality from mine wastes such as tailings and waste rock is a significant research and regulatory issue. For proposed and operating mines, closure options are often evaluated on the basis of predicted longterm water quality impacts. The large number of closed and/or abandoned mine sites worldwide also requires reliable water quality estimates to assess carrying costs for industry and governments (Puura and D'Alessandro 2005; Wolkersdorfer and Bowell 2004, 2005a, b) From a more fundamental science perspective, the long-term predictions for mine site water quality challenge our understanding of various climate, hydrology, and geochemical models and their underlying biogeochemical and physical basis (Gammons et al. 2006; Morin and Hutt 1997). In order to estimate mine waste water quality into the future, the typical approach within the regulatory, industrial, and academic communities is to develop advanced probabilistic and/or deterministic water balances for a relatively stationary post-closure site layout and topography that is coupled to steady state geochemical weathering predictions under a variety of risk scenarios (Morin and Hutt 1997). Although some of these water quality models for mine site wastes incorporate forward-looking water balances based on estimated precipitation patterns over the coming century, there is a general neglect of potential climate change impacts on both water balances and geochemical weathering rates.

At the core of these issues are the inherent difficulties in estimating some water balance components, primarily because of an inability to determine possible climate change impacts on their underlying physical drivers. Future temperatures can be estimated using either an extension of historical trends or by downscaling current global climate

models (Frev-Buness et al. 1995: Jones and Moberg 2003: Meehl et al. 2000; Mills 2006; von Storch et al. 1993; Vose et al. 2005). At most sites, greater temporal resolution and ease of modeling is achieved by extending reliable nearby historical climate datasets rather than by employing a more complex downscaling framework. Although water balance components such as evaporation and evapotranspiration depend on factors like temperature, which can be estimated to some extent, the inability to predict changes in the other drivers such as wind speed and relative humidity mean that only precipitation trends (i.e. water inputs) are allowed to vary in most water balances. Thus, it is generally assumed that changes in runoff quantities from mine wastes will vary directly with changes in precipitation. While this is the generally accepted mining approach at present, it is hoped that further advances in climatic prediction abilities will allow us to better estimate future rates of evaporation and evapotranspiration.

With regard to the weathering of mine wastes, there is a complex relationship between temperature and geochemical weathering rates, but prior work has confirmed that the Arrhenius equation is valid (Ahonen and Tuovinen 1992; Elberling 2005). In brief, the Arrhenius equation describes the temperature dependence on the rate of a chemical reaction via the equation $k = A \times e^{-E_a/RT}$, where k is the rate constant, A is the preexponential factor, E_a is the activation energy, R is the ideal gas constant, and T is the absolute temperature. To estimate the change in reaction rate between two temperatures $(k_1 \text{ and } k_2 \text{ at } T_1 \text{ and } T_2,$ respectively), the following modified form of the Arrhenius equation is often used: $\ln(k_1/k_2) = (E_a/R)(1/T_2 - 1/T_1)$. However, some previous studies have shown that temperatures in the actual weathering environment (reaction core) may be different that in the bulk waste mass (MEND 2006). This effect can be due both to the net endo-/exothermicity of any weathering reactions, as well as preferential fluid flow (both air and water) channels in the waste that affect the thermal heterogeneity of the waste mass at small scales. However, this effect is difficult to assess, and to the best of our knowledge, is generally neglected in water quality prediction efforts because of these difficulties and our current quantitative lack of knowledge regarding the magnitude and direction of any such effects. Even at polar sites, where reaction core temperatures can differ substantially from the bulk mine waste mass, good agreement has generally been reported between Arrhenius equation predicted results and field data (Elberling 2005; MEND 2006).

The temperature coefficient, or Q_{10} , is widely used to describe the proportional change in geochemical weathering rate given a 10°C change in temperature. For example, activation energies for pyrite oxidation, a common mine site water quality issue, vary from 50 to 90 kJ/mol (Lowson 1982; McKibben and Barnes 1986; Nicholson et al. 1988; Wiersma and Rimstidt 1984), which translates into Q_{10} values ranging from 2 to 4 at temperatures between 0 and 25°C. At temperatures below -2°C, Q_{10} values as high as 10 for pyrite oxidation have been reported (Elberling 2005), showing an unexpectedly large dependence of weathering rates on temperature during winter months or in permafrost environments. E_a values for pyrrhotite weathering, another key mineral that can cause acidic drainage and subsequent metal leaching problems, range from 52 kJ/mol at pH 6 to 100 kJ/mol at pH 4 (Nicholson and Scharer 1994). This pH dependence illustrates the importance of incorporating temperature, pH, and other solution effects, where possible, on weathering rate constants. Similarly, activation energies for arsenic mineral weathering (which poses a long-term water quality issue at many mine sites) can vary widely depending on pH, other redox sensitive ions, and temperature; examples include arsenopyrite (18–57 kJ/mol), realgar (As₂S₂; 60–65 kJ/ mol), amorphous AsS (124 kJ/mol), and amorphous As₂S₃ (16–17 kJ/mol) oxidation (Lengke and Tempel 2001, 2003; Rimstidt et al. 1994; Yu et al. 2007). This large variation in E_a values leads to Q_{10} factors that can range over an order of magnitude for different arsenic minerals of potential concern. These ranges are similar to the general range of activation energies (ca. 20-120 kJ/mol) reported for various minerals with no resource exploitation significance (e.g. silicates) in natural watersheds (Bird et al. 1986; Brady and Carroll 1994; Dalai et al. 2002; Helgeson et al. 1984; Knauss and Wolery 1988; Rimstidt and Barnes 1980; White and Blum 1995; White et al. 1999).

To estimate short- through long-term weathering rates of mine wastes, samples of the material are generally subjected to static and kinetic laboratory test work such as acid-base accounting, humidity cells, and leach columns (Frostad et al. 2002; Morin and Hutt 1997; Paktunc 1999). The standard laboratory test work commonly only characterizes chemical weathering reactions. Under field conditions, however, the composite weathering of mine wastes is also strongly influenced by biological activities. Thus, field weathering processes may have different effective activation energies than abiotic weathering reactions in the laboratory (Banfield et al. 1999; Moulton and Berner 1998). For example, vegetation and soil microbes promote weathering by modifying the acidity, altering physical properties, and generating chelating ligands, organic acids, and carbon dioxide (Drever 1994). Because of this, some have noted there is often a large discrepancy in mineral weathering rates between laboratory and field scales (Schnoor 1990). However, the wide prevalence of laboratory test work within well-established regulatory and research frameworks, and the lack of adequate fieldwork programs to provide the correlative linkages between laboratory and field weathering rates,

means that mass normalized laboratory weathering rates are typically extrapolated directly to field conditions, perhaps with an additional surface normalization factor (Morin and Hutt 2001). For tailings, such surface area normalization factors may not be necessary, since laboratory test work is performed on material having a particle size distribution close to that expected in the field. By comparison, waste rock dump median particle sizes (d_{50} values) can often be on the order of tens of centimeters (and with less uniformly distributed particle sizes), significantly larger than the typical sub-cm d_{50} values obtained after material is crushed into a relatively uniform distribution for laboratory test work (Morin and Hutt 1997; Stromberg and Banwart 1999). Such differences necessitate surface area normalization factors to avoid gross overestimates of field weathering rates.

Various other differences between laboratory and field conditions can complicate water quality predictions. For example, layers of weathering products are often reported as accumulating in mine tailings profiles. These layers have been shown to limit the availability of oxygen at the surface of oxidizable mineral grains, thereby limiting overall oxygen consumption, oxidation, and weathering rates by the mine waste (Elberling 2005; Nicholson et al. 1988). Although such oxygen transport barriers may confound the application of temperature-based geochemical weathering rate models across an assumed surface water infiltration depth profile, these types of issues need to be evaluated on a site-specific basis with long-term field studies and cannot be predicted in advance. Ground temperatures may also lag behind air temperatures, and the coupling of subsurfaceatmospheric thermal modeling efforts under estimated future climate scenarios will increase confidence in any predictions. However, studies of temperature effects on weathering rates have generally used air temperatures as a soil temperature proxy due to the difficulty, time, and expense of obtaining or estimating reliably integrated ground temperature profiles (Kump et al. 2000; Riebe et al. 2004; White and Blum 1995). As well, the rate of water flow through and across materials affects the rate of weathering (Kump et al. 2000; White and Blum 1995), although this factor is difficult to quantitatively link to monthly precipitation values given how the intensity, duration, and frequency of precipitation at high time resolutions (i.e. minutes) is at least as important as the total monthly amounts available from historical climate records, which is the general temporal limit for future precipitation predictions. For this reason, inclusion of these hydrologic mechanisms that contribute to mine waste weathering remains beyond our practical current predictive abilities, although there is the possibility of coupling both temperature and flow rate influences on weathering rates if sufficiently complex kinetic laboratory and field testing programs are designed. Yet, despite all these limitations and variations,

researchers have reported good modeling correlation for field weathering rates using Q_{10} values between 2 and 4 (Stromberg and Banwart 1999), allowing for some reliable generalizations about the effects of temperature and future climate change on mine waste weathering.

We have developed a conceptual analytical framework for the risk-based estimation of climate change effects on mine waste runoff water quality that incorporates variability in both precipitation and temperature, and the resulting effects on water balances and geochemical weathering rates. We used a case-study approach to develop and present our model for regions near the city of Yellowknife, Northwest Territories, Canada (Fig. 1), the site of two major historical gold-mining operations (Giant Mine and Con Mine) in their closure/post-closure phases, and for which these types of models would be directly applicable in site planning operations.

Materials and Methods

The nearest long-term climate station to Yellowknife is the Yellowknife A climate station: latitude: 62° 27.600'N; longitude: 114° 26.400'W; elevation: 205.70 m MSL; climate ID: 2204100; WMO ID: 71936; TC ID: YZF. Climate data obtained for the Yellowknife A station were from Environment Canada via the Canadian climate normals or averages (CCNA) 1971-2000 database (http://climate. weatheroffice.ec.gc.ca/climate normals/index e.html) and the adjusted historical Canadian climate data (AHCCD) database (http://www.cccma.ec.gc.ca/hccd/). The CCNA datasets do not account for historical errors in measuring temperature, rainfall, or snowfall. The AHCCD database provides adjusted precipitation and homogenized temperature data sets that can be used for climate research. Temperature data can require adjustment due to changes in site locations and/or the effects of physiographic changes near the site (i.e. urbanization, land use alterations). A review of the CCNA and AHCCD datasets between 1971 and 2000 for Yellowknife A indicated no difference in the temperature averages between the two datasets, though the AHCCD represents the efforts of Environment Canada to correct for the common biases in rain and snow undercatch in northern Canada. The CCNA underestimates annual rainfall, snowfall, and total precipitation by 11, 14, and 22%, respectively (Table 1). Thus, the AHCCD dataset was used for all further analyses in the current work in order to obtain the most reliable estimates and trends for site precipitation.

The climate data was analyzed for trends using the nonparametric Mann–Kendall test and the non-parametric Sen's method (Salm et al. 2002). This conceptual approach is established in the regulatory framework for climate analyses, and has been previously used by governmental





Table 1 Comparison of 1971–
2000 climate normals or
averages at Yellowknife A in
the CCNA and AHCCD datasets

Variable	CCNA 1971-2000 avg	AHCCD 1971-2000 avg
Annual rainfall (mm)	164.5	184.4
Annual snowfall (mm)	151.8	176.4
Total annual precipitation (mm)	280.7	360.9
Mean annual minimum temperature (°C)	-9.0	-9.0
Mean annual temperature (°C)	-4.6	-4.6
Mean annual maximum temperature (°C)	-0.2	-0.2

agencies for analyses of adjusted historical climate data (Vincent and Gullett 1999). Frequency analyses were conducted using the DISTRIB 2.20 software (Wanielista et al. 1997) and the unaltered datasets over the period from 1942 to 2006 in the AHCCD database for the Yellowknife A station. The Pearson Type III distribution was used for all datasets, after confirming the best quality of fit relative to other commonly used distributions (e.g. multiparameter log normal, log Pearson, Gumbel, and generalized extreme models). The DISTRIB probability distribution program was chosen as it is a commonly accepted software package used within industry, academia, and government and is available freely on the internet (http://www.cee.ucf.edu/software/) from the University of Central Florida (Orlando, FL, USA).

Results and Discussion

Site Climate Assessment

Key to acquiring inputs towards any predictive assessment of future mine waste runoff water quality is an analysis of historical site climate data for averages and trends. Monthly, seasonal, and annual total precipitation amounts and mean temperatures were available for the Yellowknife A climate station in Northwest Territories, Canada, from the AHCCD dataset. Detailed results of the statistical trend analyses are given in Supplementary Material Figures S1 and S2 and Tables S1 and S2. Statistically significant (P < 0.05) trends in increasing monthly precipitation were only found during January

[P < 0.05; 15.8 (0.3-29.4) mm/century, range is the 95%confidence intervals about the mean], February [P < 0.05; 12.4 (2.6-23.8) mm/century], March [P < 0.05; 12.4 (2.6-23.8) mm/century]0.01; 17.0 (5.3–30.9) mm/century], and November [P <0.01; 32.5 (14.0-50.8) mm/century], although significant increasing seasonal trends were observed for winter [P < 0.01; 43.3 (16.9-69.6) mm/century], spring [P < 0.01; 43.3 (16.9-69.6) mm/century]0.01; 40.0 (10.8–66.5) mm/century], and autumn [P < 0.01; 71.1 (31.5-105.7) mm/century], as well for total annual precipitation [P < 0.001; 185.0 (106.2-262.3) mm/century]. Similarly, significant trends in increasing monthly mean temperatures were only observed for February [P < 0.05; 6.8 (1.4–12.4) °C/century], April [P < 0.05; 5.7 (0.7-11.2) °C/century], June [P < 0.05; 2.5](0.6-4.8) °C/century], and July [P < 0.01; 2.5 (0.8-4.0) °C/ century], although significant increasing seasonal trends were observed for winter $[P < 0.01; 5.9 (1.7-9.6) ^{\circ}C/$ century], spring [P < 0.05; 3.4 (0.2-6.4) °C/century], and summer [P < 0.05; 1.5 (0.0-2.9) °C/century], as well for annual mean temperature [P < 0.01; 2.7 (1.0-4.4) °C/ century]. The temperature and precipitation increases we found are consistent with previous analyses in this region (Rouse et al. 1997; Vincent and Gullett 1999). Thus, the Yellowknife A station appears to be both warming and getting wetter, with the increasing precipitation trends concentrated during the winter months primarily as snow, and the warming occurring predominantly in spring and early summer. Based on this analysis, we would expect Yellowknife to have more intense and earlier spring freshet runoff periods as the increased over-winter snow accumulation melts more rapidly during a warmer spring.

The choice of whether to include non-significant trends in estimated mine site water quality modeling efforts will be determined by consultations with stakeholders and the applicable regulatory frameworks. For the current illustrative example, we have chosen to accept both significant and non-significant monthly climate trends in our analysis. However, we stress that a number of modeling iterations can be readily performed using our analytical approach, whereby non-significant trends are excluded or modified to meet regulatory needs and preferred risk and sensitivity analysis requirements for the public, corporate shareholders, and governmental and financial sector bonding regulations. Based on our choice of using both the significant and non-significant trends in this analytical example, the estimated monthly mean total precipitation amounts and mean temperatures (including upper and lower 95% confidence intervals) in the years 2010, 2025, 2050, and 2100 are given in Supplementary Material Tables S3 and S4, respectively. A summary monthly comparison of the 1971-2000 normals or averages period to these future predictions is also given in Supplementary Material Table S5.

Frequency Analyses

Climate trends estimate the likely future average annual precipitation and temperature patterns at a site, but do not capture any expected year-to-year variation for the projected climate. To meet risk analysis needs in future mine site water quality predictions, inter- and intra-annual variation can be estimated by use of frequency analyses. Frequency distributions cannot be generated for forward extrapolations of linear regressions on time-series trends for the historical Yellowknife A dataset (i.e. frequency analyses to estimate return periods cannot be performed on as yet non-existing future datasets). Rather, the approach we took assumed that the distribution and relative magnitude of extreme events in the future would be similar. despite the different climate. We note that this approach does not account for changes in patterns of extreme weather that may arise from climate change (Easterling et al. 2000; Rosenzweig et al. 2001). To apply the current frequency distributions on future climates at Yellowknife, we first calculated absolute precipitation and temperature frequency values for representative wet/dry and warm/cool return periods (1-in-2, -3, -5, -10, -25, -50, -100, and -200 year), respectively. We then calculated ratios of the magnitude of each return period to the average return period (1-in-2 year) for precipitation, and the differences in the magnitude of each return period from the average return period (1-in-2 year) for temperature. The results of the frequency analyses, and these ratios/differences relative to the average year, are presented in Supplementary Material Tables S6 through S9. These ratios and differences for precipitation and temperature, respectively, can then be applied directly to the estimated average climate parameters in a future year obtained via the regression trends in order to yield a projected frequency distribution at a particular forward time. For example, the estimated monthly and annual total precipitation frequency distributions in the year 2025 are presented in Supplementary Material Table S10, while the estimated monthly and annual mean temperatures in the year 2050 are presented in Supplementary Material Table S11.

Water Balance Processes

Depending on the complexity of the mine site and the regulatory points-of-compliance, the site water balance and water quality models may need to account for surface runoff loss processes such as evaporation from open water bodies and bare soils, evapotranspiration from vegetated surfaces, infiltration losses to local and regional groundwater systems, and over-winter snow sublimation and wind losses. For the hypothetical site against which the current modeling framework is applied, the regulatory

point-of-compliance is where mine waste surface runoff leaves the mine waste footprint, and no vegetation or open water bodies exists on the mine waste. Thus, we have neglected evaporative or evapotranspirative loss pathways in our modeling approach. However, the predictive equations for these two processes are partially based on temperature. Thus, forward-looking site climate temperature predictions can be somewhat extended to evaporation and evapotranspiration, although relative humidity and wind speeds are also primary data inputs when calculating these parameters. Future estimates of both relative humidity and wind speed are not possible to predict based on historical records, and the current downscaled climate models also do not allow reliable predictions. With these limitations in mind, our approach can still be readily adapted to include more complex mine site landforms and water balance/ quality models with evaporative and evapotranspirative loss pathways. The site under consideration is also in a zone of discontinuous permafrost, just 500 km south of the zone of continuous permafrost (Rouse et al. 1997), with 1971-2000 normals or averages period annual temperature of -4.6° C, which is only supposed to rise -1.7° C by the year 2100. Thus, we expect discontinuous permafrost conditions to apply in the study area over the next century, and this type of ground temperature environment (with the exception of sites within deep taliks near larger regional lakes) will generally prevent substantial surface runoff losses to ground water.

For our hypothetical site (an exposed or covered, but unvegetated, tailings or waste rock dump surface), water balances and water quality models often just apply a runoff factor based on available precipitation inputs during each month. Depending on the site specifics, the runoff factor may vary on a month-to-month basis to account for processes such as evaporative losses during surface runoff [which will depend on the slope of the waste (i.e. hydraulic residence time), as well as temperature, relative humidity, and wind speed] and the amount of infiltration losses into the waste (dependent on particle-size distributions, surface runoff residence times, and antecedent moisture/temperature conditions in the waste-including negligible infiltration into frozen soils during the early freshet period). At our site, a uniform runoff coefficient of 1.0 was used throughout the open-water season to simplify the discussions, although we also stress that non-uniform non-unity runoff coefficients can readily be incorporated into the modeling approach.

At Yellowknife, any precipitation falling during a particular month generally appears as runoff during that month between May and October, whereas precipitation between November and April is typically stored on the surface until the May freshet. Thus, the only water balance loss processes for our site are over-winter sublimation and wind losses (wind accumulation may also be important in regions of low topographic relief and high winter wind speeds such as at Yellowknife) that reduce the amount of total over-winter precipitation available for runoff at the spring freshet. At present, there are no reliable means of estimating wind accumulation/losses for snow on the exposed mine wastes, but they may be substantial. The start of an annual snow survey program on mine wastes at a particular site is always recommended, and data from the proposed programs would allow for significant refinement and calibration of both site water balances and water quality models. Sublimation at the site was calculated using Kuzmin's method along with wind speed, vapor pressure, and saturation vapor pressure available from the Environment Canada Yellowknife A climate station 1971-2000 averages or normals (http://climate.weatheroffice. ec.gc.ca/climate_normals/index_e.html). Monthly sublimation losses at Yellowknife were estimated as follows: January, 11.0 mm; February, 11.5 mm; March, 17.3 mm; April, 57.8 mm; November, 12.6 mm; and December, 9.2 mm (total annual sublimation of 119.4 mm). These values are within the expected range of sublimation amounts for this region of northern Canada (Rouse et al. 1997). As noted previously, wind speed and vapor pressure patterns at the site may vary over time due to climate change, but the lack of an easily accessible and available composite historical record for these parameters typically prevents predicting the direction or magnitude of any potential changes. Thus, we generally assume that over-winter sublimation rates remain constant over the modeling periods.

Modeling Framework

Runoff water quality estimates from exposed mine waste materials require the application of advanced geochemical and hydrological analyses coupled with models that account for secondary mineralization and sorption. To best determine representative periods for modeling, the analyses need to account for the following factors: the amount of surficial runoff expected; the degree of weathering occurring over the period of interest via scaling of laboratory geochemical test work data to expected field conditions; the appropriate return periods for risk analysis; and how climate is expected to change on-site over time. The formula for estimating the primary water quality (i.e. before secondary mineralization and/or sorption controls) of surface runoff from exposed mine waste is as follows, $C_a^{1^\circ} =$ $(P_r \times [p_d^f \times d_i \times A_u] \times \Delta t \times [r_w^T/r_w^{20^\circ C}] \times 1 \text{ m}^3 / 1,000 \text{ L})/(I_r \times A_u)$, where $C_a^{1^\circ}$ is the primary concentration (in mg/L) of water quality parameter a, P_r is the late-stage production rate (in mg/kg/d) of parameter a from humidity cell data, ρ_d^i is the field dry density of the mine waste (in kg/m³), d_i is the depth through which the water infiltrates in the mine waste (in m), A_u is unit area (1 m²), I_r is the water depth (in m) infiltrating through mine waste depth d_i over time period Δt (in days), and $[r_w^T/r_w^{20^\circ C}]$ is a kinetic factor to account for different weathering rates under field average temperature T over time period t versus laboratory temperature (20°C) conditions over the same time period.

Chemical reactions generally decrease in rate by at least one-half for each 10°C drop in temperature (i.e. a Q_{10} of 2). Thus, the value of $[r_w^T/r_w^{20^{\circ}\text{C}}]$ can be calculated by the following formula: $[r_w^T/r_w^{20^{\circ}\text{C}}] = 2^{(T_{avg} - T_{20^{\circ}\text{C}})/10^{\circ}\text{C}}$, where T_{avg} is the average field temperature over time period t and $T_{20^{\circ}\text{C}}$ is the average laboratory testing temperature (20°C). Using this approach, on an annual average basis, the predicted increases in temperature between the 1971-2000 climate normals or averages period at Yellowknife is expected to have the following potential increase on the rates of mine waste chemical weathering: 1971-2000, 0%; 2010, 5%; 2025, 9%; 2050, 15%; and 2100, 28%. Thus, mine wastes are expected to chemically weather at a rate about 1.3-fold higher than present by the year 2100. The effects of climate change on physical weathering processes (e.g. freeze-thaw cycling) cannot be reliably predicted. We also stress that Q_{10} values of up to ten have been reported for mineral oxidation processes in sub-arctic regions (see "Introduction" for a review of published values), suggesting that the temperature change influence on geochemical weathering rates, and resulting water quality, could be much higher than we estimate. The actual Q_{10} value to be used in the modeling approach will depend on the results of laboratory and field testing programs, mineralogical analyses, and the risk framework in which the modeling is conducted, and can be calibrated over time in the model as additional field data becomes available. There is much complexity in what controls the rates of chemical weathering and the resulting runoff water quality, but many of the underlying principles and processes remain unknown and site-specific. Thus, the current modeling approach does not represent the ideal, but rather an improvement compared to the overly simplistic steady-state geochemical production models used today in assessing current versus future mine site water quality.

We also stress that d_i may be related to I_r (i.e. $d_i = f[I_r]$), depending on site-specific conditions determined by surface evaporation trends, the intensity-duration-frequency profiles of precipitation and melting events, and the micro- through macro-pore signature of the waste mass (determined by particle size and shape distributions, compaction profiles, and waste placement methods). In the example above, we have assumed that d_i is constant and independent of I_r (where, of course, I_r varies with precipitation and temperature). Where a greater level of modeling precision is desired, the equations we present may readily be modified to include the

more complex (and accurate, but computationally more expensive) view of $d_i = f[I_r]$. Various one-, two-, and three-dimensional variably saturated coupled climate, thermal, hydrologic, and hydrogeologic models with possible flow path preference inclusion are available to potentially develop site-specific relationships between d_i and I_r , which, if desired, can be incorporated via variable substitution into our pre-existing conceptual framework. Precipitation effectiveness correlations, aridity indices, and subsurface mineral weathering thermochemistry modules may also be incorporated to meet varying levels of site-specific calibration and complexity. The reader is referred elsewhere for specific discussions on each of these issues (see e.g. Arora 2002; Faybishenko 2007; Lefebvre et al. 2001a, b; Scanlon et al. 1997; Simunek et al. 2003; Webb et al. 2008).

Furthermore, while substantial effort has been invested in determining the Arrhenius relationship of mine waste weathering at temperatures less than optimum weathering temperatures, little work has been done on the temperaturerate relationship at elevated temperatures that are present in rapidly oxidizing materials (e.g. sulfide oxidation), and that also may become more prevalent from climate related external thermal forcing in mine wastes, even in the absence of internal oxidative heat inputs. For example, the temperature influence on the activity of sulfur oxidizing bacteria at sites from temperate and arctic regions has been considered (MEND 2006). While similar Arrhenius slopes were observed between the two regions at temperatures above and below the optimum temperature, the optimum temperature for biochemical sulfide oxidation was significantly lower (ca. 22°C) at arctic sites compared to their temperate counterparts (ca. 35°C). Consequently, more work needs to be performed on thermal inhibition of biochemical mineral weathering at higher temperatures, including the site-specific dependence of the effects, and the findings included in conceptual models such as ours to better incorporate the actual complex temperature-rate behavior that can occur at elevated temperatures. We cannot also exclude the possibility of an optimum temperature for abiotic mine waste weathering processes due to the interrelationships of various factors such as temperature influences on pH, solubility, formation constants, and other physical and chemical system properties. As work continues in this area and more data become available, the current conceptual mine waste weathering models can be updated.

As discussed above, based on the site climate record at Yellowknife, surficial runoff is expected only during the period between May and October. The temporal resolution of modeling at mine sites in the Canadian north is typically at monthly resolution, since regulatory limits generally specify monthly composite averages (in addition to single

grab-sample maxima), and the limits of climate prediction do not allow more refined considerations that account for reliable daily precipitation events over the course of a year coupled with inclusion of antecedent moisture and temperature conditions. For estimating exposed mine waste surficial runoff water quality during the months of June through October at Yellowknife, only the predicted monthly total precipitation (for runoff volume calculations) and average monthly temperature (for calculating the degree of weathering) are needed, along with fixed assumptions regarding runoff coefficients, infiltration penetration depths, and mine waste densities. During the month of May at Yellowknife, the total runoff will include the cumulative November through April overwinter total precipitation (minus sublimation, and plus/minus wind accumulation/losses) plus the total precipitation received during May, and adjusted for a runoff coefficient. The estimated water quality in the May spring freshet runoff will also need to account for average temperatures between the period of November through May, as this is the extended time period for over-winter accumulated weathering products that are discharged during spring runoff.

Relative Water Quality Indices for Narrowing Relevant Risk Scenarios

To help determine the risk scenarios requiring modeling for mine wastes near Yellowknife, a preliminary framework for calculating relative primary water quality indices was developed. Climate and hydrology inputs to the framework are the total precipitation expected, over-winter sublimation losses, and the mean temperature over the period of interest. The total precipitation available for potential runoff from mine wastes and the mean temperature for geochemical weathering for each time period of interest in water quality modeling between the 1971-2000 climate normals or averages period and the year 2100 is given in Table 2. Note that forward-looking climate predictions were based on a linear extrapolation of the historical dataset, an approach which does not include curvilinear and discontinuous climate responses to anthropogenic forcings. The preferred return period for risk analysis at the project was chosen as a 1-in-200 year event. Although this return period is commonly used in environmental assessments for Canadian mines, the actual values are always the site-specific results of discussions between the mining company, its financiers, government regulators, and other public and private stakeholders. In terms of water quality, the risk analysis would most appropriately consider extreme warm periods (where mine waste weathering rates are higher, leading to greater contaminant loadings per unit of precipitation), extreme dry periods (where less runoff is available per unit of contaminant loadings), and combinations of these two factors. Colder

 Table 2
 Total precipitation and mean temperatures for use in calculating relative water quality indices; note that November–May total precipitation available for runoff includes subtracted sublimation losses of 119.4 mm

	Nov–May	Jun	Jul	Aug	Sep	Oct
Total precipit	ation (mm)					
1971-2000	49.8	29.9	38.0	44.5	36.2	43.0
2010	66.1	25.4	30.0	44.2	41.9	39.4
2025	82.3	28.0	30.3	46.7	45.5	41.5
2050	109.5	32.3	30.7	50.9	51.4	45.0
2100	163.6	40.9	31.6	59.3	63.4	51.9
Mean tempera	ature (°C)					
1971-2000	-15.0	13.4	16.8	14.3	7.1	-1.7
2010	-14.3	13.7	17.3	14.2	7.6	-1.9
2025	-13.7	14.1	17.7	14.1	7.7	-2.2
2050	-12.7	14.7	18.3	14	7.8	-2.8
2100	-10.7	16.0	19.6	13.9	8.1	-4.0

and/or wetter periods offer lower contaminant production rates and more dilution, respectively, which are both favorable in terms of exposed tailings water quality.

With these net runoff and average temperature estimates in Table 2, we applied the frequency analysis factors given in Supplementary Material Tables S7 and S9 to predict runoff and average temperature during the November-May, June, July, August, September, and October periods under the following comparative risk scenarios: average precipitation and temperature; 1-in-10 year dry period coupled with a 1-in-10 year warm period; 1-in-200 year dry period coupled with average temperature; and average precipitation coupled with a 1-in-200 year warm period. These runoff amounts and average temperatures were then combined in the following equations to obtain relative precipitation factors (F_p) and weathering factors (F_w) for the four risk scenarios stated above, respectively, $F_p = \left(P_{avg}^{1971-2000} \times \Delta t\right)/(P_t \times 30 \text{ days})$ and $F_w = \left(2^{\wedge} \left[T_{avg}^t - T_{avg}^{1971-2000}\right]\right) \times (\Delta t/30 \text{ d})$, where $P_{vo}^{1971-2000}$ is the average monthly precipitation (in mm) during the 1971-2000 normals or averages period (40.2 mm), P_t is the expected precipitation (in mm) over the period of interest under a particular risk scenario, T_{avg}^{t} is the average temperature (in °C) over the period of interest under a particular risk scenario, $T_{avg}^{1971-2000}$ is the average annual temperature (in °C) during the 1971-2000 normals or averages period (-4.6°C), and Δt is the length of time for the period of interest (in days). Multiplying the precipitation and weathering factors gives the net hydrogeochemical factor (F_{nh}) , which can be compared between modeling periods for a particular month-risk scenario combination to see relative changes and trends over time, or between modeling periods across different risk scenarios to

interest

ascertain when the timeframe of worst water quality is likely to occur within the modeling domain. A relative global hydrogeochemical factor (F_{rgh}) can be obtained by dividing the F_{nh} by the minimum net hydrogeochemical factor $(F_{nh,min})$ within a particular risk scenario or across various risk scenarios. Note that these factors are not intended to represent absolute physical quantities, but to allow approximate assessments of relative water quality between monthly runoff periods and over time.

The interpretation of these factors allows an assessment of when the month of highest analyte primary concentrations are expected to occur under a given risk scenario, whether low precipitation or warming effects are more important in determining water quality issues, and the relative magnitudes of analyte primary concentrations under differing risk scenarios. Graphical presentations of time-trends between the 1971–2000 normals or averages period and the year 2100 for the F_{D} , F_{W} , and F_{nh} under the four risk scenarios are shown in Fig. 2. Based on the results, the highest analyte primary concentrations (as expressed by the F_{nh} in potential mine waste surficial runoff near Yellowknife during the average year, 1-in-10 year dry + 1-in-10 year warm, and average precipitation + 1-in-200 year warm scenarios is expected to occur in May, when the accumulated



Table 3 Summary of expected trends in monthly water quality frommine waste surface runoff near Yellowknife under the four risk scenarios between the present and the year 2100

Month	Trend in primary analyte concentrations
May	Decrease by ca. 60%
June	Decrease by ca. 13%
July	Increase by ca. 46%
August	Decrease by ca. 27%
September	Decrease by ca. 39%
October	Decrease by ca. 29%

over-winter weathering products are flushed from the tailings. Because of expected increases in over-winter precipitation, net runoff during the May freshet is expected to increase substantially over the coming century, and will more than compensate for increasing weathering rates due to higher temperatures. Thus, analyte primary concentrations in tailings surface runoff during the spring freshet are expected to decrease by about 60% (i.e. a 60% decrease in F_{nh}) between the present and the year 2100 in an average year. A summary of the trends in expected mine waste surface runoff water quality, as represented by temporal trends in F_{nh} values, over the next century under all four scenarios is given in Table 3. The analyte primary concentrations are expected to decline over time during all months except July, since the increasing amounts of expected precipitation will more than compensate for higher weathering rates due to the higher average temperatures throughout the year. A summary of the month-year combination in which the highest analyte primary concentrations are expected, and associated relative water quality indices as represented by comparative F_{rgh} values, under the four risk scenarios is given in Table 4. The relative water quality index shown in this table is an approximate measure of the relative concentrations (i.e. ratio) of water quality parameters in a particular month-year combination under a given scenario compared to that expected in the worst month of an average year. Thus, the July 2100 1-in-200 dry + average temperature scenario presents the risk endmember that needs to be focused upon in more advanced geochemical and water quality modeling, since analyte primary concentrations are expected to be higher under this scenario than in any of the other risk scenarios (e.g. concentrations about sevenfold higher than under the worst month during an average year).

We stress that a key assumption in the modeling approach is that chemical weathering of mine waste is rate limiting as compared to dissolution and water transport rates, so that weathering products are continuously being flushed away and are not reaching solubility limitations that would lead to short-term or longer storage of weathering products prior to subsequent release during future periods with less concurrent weathering and greater water throughflow. Inclusion of such store-and-release mechanisms for weathering products would be possible in a more complex modeling framework. Similarly, the inclusion of secondary and tertiary mineralization and sorption processes along preferential flow paths in the mine waste can also be included by use of appropriate coupled hydrologicgeochemical models. These processes would act to attenuate the primary water quality concentrations presented here. However, one difficulty always encountered when attempting to account for such post-primary mineralization and sorption processes is the potential for previously precipitated/sorbed material to be remobilized and dissolved during future periods with lower inherent geochemical production rates (i.e. a form of cross-contamination and feedback across modeling intervals). These issues are currently impossible to resolve with any certainty given the complexity of mine sites, the heterogeneity of mineralogy and weathering processes, and the unknown nature of surface and subsurface flow paths.

Conclusions

Using a case study approach for the arctic mining region of Yellowknife, Northwest Territories, Canada, we developed a conceptual analytical framework for investigating the risk-based impact of climate change effects on mine waste runoff water quality. Forward-looking trend analyses of historical climate datasets, as well as the application of frequency analyses on the historical data to current and

Table 4 Summary of the month-year combination in which the higher analyte primary concentration is expected, and associated relative water quality indices (F_{rgh} values) under the four risk scenarios considered

Scenario	Month-year combination in which highest primary analyte concentrations are expected	Relative water quality index (F_{rgh})	
Average	May 1971–2000	1.0	
1-in-10 dry + 1-in-10 warm	May 1971–2000	3.2	
1-in-200 dry + average temperature	July 2100	7.4	
Average precipitation + 1-in-200 warm	May 1971–2000	2.0	

future climates, are the primary data inputs to the model. If available, site-specific kinetic data for mine waste weathering, as well as the results of one- or multi-dimensional coupled climate-thermal-hydrologic-hydrogeologic models may also be included in the conceptual approach, depending on the desired level of complexity and available site data. The predicted temporal variability in both precipitation and temperature using trend analyses is used to estimate effects on both site water balances and geochemical weathering rates. As a result, time-resolved relative precipitation and weathering factors can be calculated, and the interpretation of these factors allows an assessment of when the period of highest analyte concentrations would be expected to occur under a given risk scenario over a modeling timeframe. In addition, the approach allows for an assessment of whether low precipitation or warming effects are more important in determining future water quality issues at a mine site, and the relative magnitudes of potential water quality concerns under differing risk scenarios.

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