

Calibration of a PHREEQC-based geochemical model to predict surface water discharge from an operating uranium mill in the Athabasca Basin

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A PHREEQC based geochemical model has been developed to predict impacts from the McClean Lake Mill discharges through three lakes in the Athabasca Basin, Saskatchewan, Canada. The model is primarily a mixing calculation that uses site specific water balances and water compositions from five sources: 1) two water treatment plants, 2) waters from pit dewatering wells, 3) run-off into the lakes from surface waters, 4) ambient lake compositions, and 5) precipitation (rain and snow) onto the pit lake surface. The model allows for the discharge of these waters into the first lake, which then flows into another nearby lake and finally into a third larger lake. Water losses through evaporation and the impact of subsequent evapoconcentration processes are included in the model.

PHREEQC has numerous mass transfer options including mixing, user specified reactions, equilibration with gas and solid phases, and surface complexation. Thus this program is ideally suited to this application. Preparation of such a complicated model is facilitated by an EXCEL Spreadsheet, which converts the water balance into appropriately formatted mixing proportions and to prepare portions of the PHREEQC input file in a format directly useable by PHREEQC. This allows for a high level of flexibility, while reducing transcription errors. For each scenario, the model path involves mixing of the waters in the first lake, followed by evapoconcentration, equilibration of the resulting solution with gas phases, including carbon dioxide and oxygen and with minerals and surfaces. The resultant composition is mixed in the second lake with more surface water, lake water and precipitation, and then re-equilibrated. This water represents the flow into the final lake; further mixing/dilution is accommodated; chemical equilibration may also occur. Because of the numerous steps and processes that define the pathway, each annual step requires approximately 200 lines of input in PHREEQC.

Models used in the initial calibration used annual water balances and compositions from 2000 to 2011. The calibration indicated that a simple mixing approach could explain the behavior of conservative elements such as sodium, potassium and chloride. For calcium and sulfate, the precipitation of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) may occur in the holding ponds. This lowers the calcium and sulfate discharge from this source term, but also improves the fit in the downstream lakes. Even though most of the major ions are conserved and attenuated through dilution, many of the chemicals of concern such as molybdenum, uranium and arsenic have lower measured concentrations than predicted by the initial mixing models. Attenuation processes beyond simple dilution and typically related to mineral precipitation or surface complexation are responsible for these drops in concentrations. This modeling effort provides insights into the geochemical behavior of trace metals that would not be readily apparent without the model.

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