Science of the Total Environment 846 (2022) 157390

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Assessing the long-term evolution of mine water quality in abandoned underground mine workings using first-flush based models

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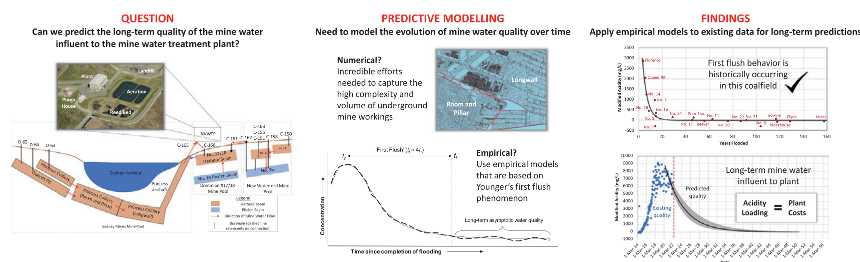
HIGHLIGHTS

- Assessing the historical water quality in mine workings in the Sydney Coalfield
- Verification of first flush phenomenon to describe evolving mine water quality
- Active treatment plant pumps out mine water to prevent environmental discharge.
- First flush models predict long-term quality of mine water influent to the plant.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: José Virgílio Cruz

Keywords:
Acid mine drainage
Mine water treatment
Predictive modeling
Mine water management
Younger's first-flush

ABSTRACT

Coal mining activities can leave an extensive network of abandoned underground workings that gradually flood after operations cease. This rising mine water can eventually lead to uncontrolled releases of harmful acid mine drainage (AMD) to the environment. Treatment plants are used to extract and treat the mine water to maintain its elevation below suspected discharge points. Accurate predictions of long-term water quality, and treatment plant operations, are highly challenging due to the complexity and volume of the underground workings. As numerical models require considerable effort to effectively implement, empirical models that are based on the 'first-flush' phenomenon, where mine water concentrations peak shortly after flooding and then exponentially decline, may provide suitable long-term predictions. The objective of this study was to assess the robustness of first-flush based models for describing mine water behavior at large, complex mine pools in the Sydney Coalfield (Nova Scotia, Canada). Numerous mine pools across the coalfield flooded at various times over 100+ years, with extensive mine water quality data available in various pools of different ages. The historical evolution of mine water quality demonstrated first-flush behavior across key AMD indicator parameters (acidity, sulfate, iron), concentration ranges, and mine pool depths. Two 'newer' mine pools, which only flooded in the past 10–15 years, rely on an active treatment plant to manage mine water levels below environmental discharge points. Influent water quality from each mine pool was sampled bi-weekly between 2011 and 2022, and first-flush models were then applied to predict the future quality of mine water entering the treatment plant over the long-term. Knowledge on long-term influent quality can help to optimize treatment plant requirements and related expenses.

1. Introduction

Environmental contamination by acid mine drainage (AMD) from abandoned underground coal mine workings remains a major and persistent problem worldwide. Upon the cessation of mining activities and closure of the colliery, the de-watering pumps are turned off and groundwater rebound occurs, gradually flooding the open workings (e.g., Gandy and Younger, 2007; Álvarez et al., 2018). The rising mine water and oxygen in the void space interact with the exposed metal sulfides (often pyrite, FeS_2), causing a complex sequence of oxidation-reduction reactions that generate AMD leachate (Johnson and Hallberg, 2005; Nordstrom et al., 2015). Characterized by low pH and high concentrations of acidity, sulfate, heavy metals and other toxic elements (International Network for Acid Prevention (INAP), 2014), AMD emanating from abandoned workings can severely impact environmental receptors such as streams, rivers,

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http://dx.doi.org/10.1016/j.scitotenv.2022.157390
Received 30 May 2022; Received in revised form 6 July 2022; Accepted 11 July 2022
Available online 16 July 2022

and aquifers (e.g., Luís et al., 2011; Galvan et al., 2021; Ojonimi et al., 2019).

AMD is released to the environment when the mine water elevation inside the workings rises to that of possible discharge points (Adams and Younger, 2001). While sealing of the open workings is one of the most desirable approaches for preventing and/or controlling AMD release (e.g., Soroko et al., 2020), it is highly challenging as large numbers of natural and man-made hydraulic connections can exist, sometimes unknowingly, between the workings and the surface, including outcrops, air shafts, and bootleg workings (e.g., MacLeod, 2010; Polak et al., 2016). An alternative approach is to maintain mine water elevations below that of discharge points by continuously pumping mine water out of the workings and into treatment plants constructed over the workings (e.g., Cravotta, 2010; Skousen et al., 2019).

Understanding and modeling the behavior and evolution of mine water quality in flooded workings over time can provide valuable insight to assist the design and long-term operation, cost, and lifespan of a mine water treatment plant. However, accurate understanding and modeling of mine workings is highly challenging. The nature of underground workings, which are known for their large extent and depth, naturally creates a complex and heterogenous network of intersecting mine tunnels, which have the potential to collapse and further complicate the system (Mack et al., 2010; Wolkersdorfer, 2008).

Models for predicting water quality in flooded mine workings can be divided broadly into numerical models and field-based empirical models. Numerical models have been used to simulate various hydraulic and geochemical processes in mine workings (e.g., Vandenberg et al., 2016; Kuchovský et al., 2017; Tomiyama et al., 2020); however, they can be oversimplified and highly time- and cost-intensive. For example, numerical models have represented workings with homogeneous pipe networks or ponds (e.g., Hamm et al., 2008; Vaute et al., 2010), or relied on mine plans that may or may not be available, or no longer represent the true state of the mines due to tunnel collapses or bootleg workings (e.g., González-Quirós and Fernández-Álvarez, 2019). Incredible efforts are usually needed to develop numerical models that can accurately represent the hydrology and geochemistry of these highly complex and intricate systems.

Empirical models have been developed by analyzing decades of historical mine water quality and trends from minefields worldwide. While

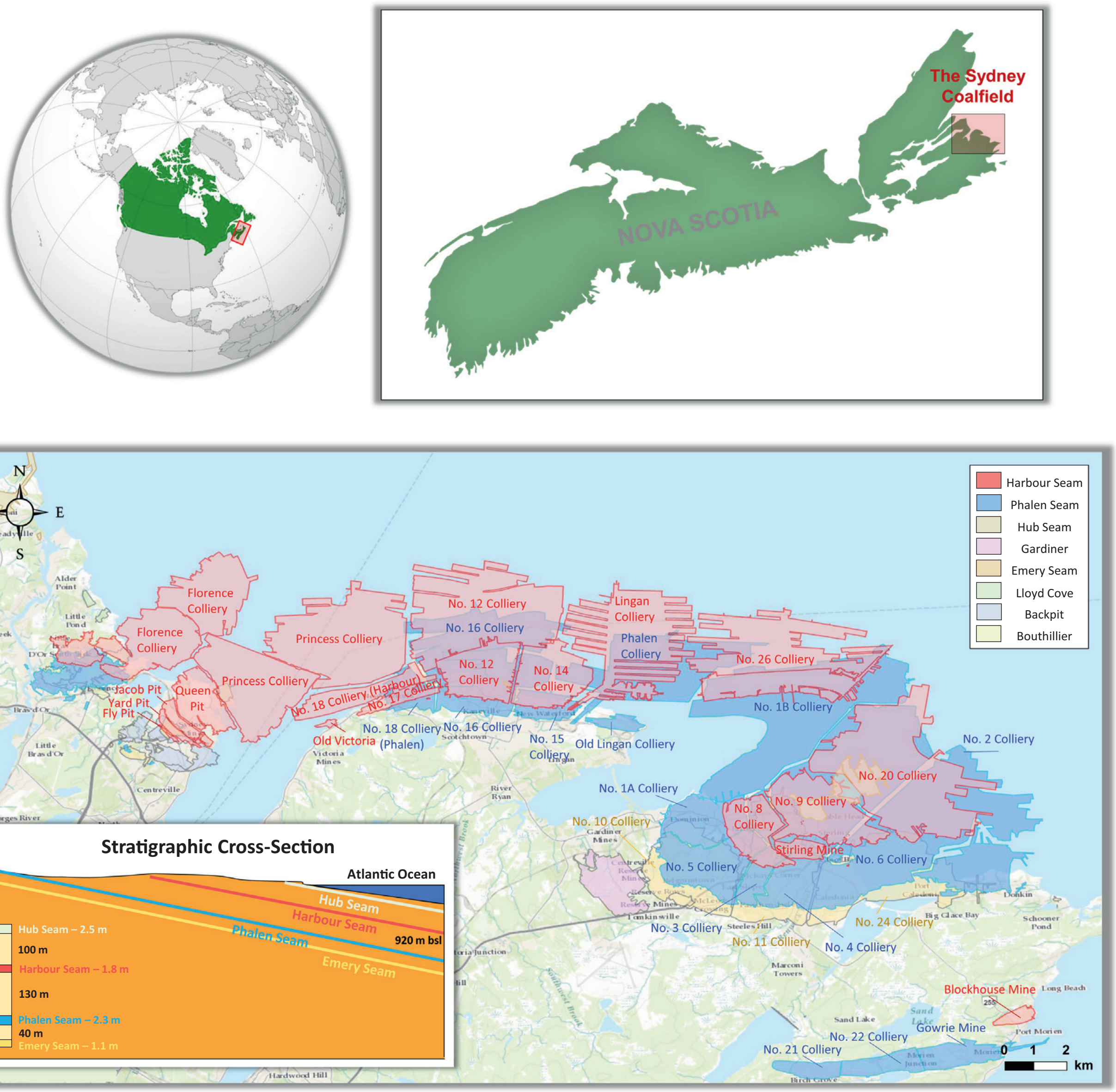

Fig. 1. Site map indicating the location of the Sydney Coalfield in Nova Scotia, Canada, along with outlines of the various mine collieries. A general cross-section of the various coal seams is also shown.

simpler and less intensive, empirical models can be equally, or more, representative of mine water behavior than highly complex numerical models (Younger, 2016). The water quality in mine workings is generally understood to improve over time; as low-quality mine water is extracted from the workings, it is replaced by fresher infiltrating surface water and recharging groundwater (Banks et al., 1997; Wood et al., 1999; Demchak et al., 2004). Younger (2000) examined the long-term behavior of mine water in numerous coal mines in the UK and derived the 'first-flush' phenomenon, where contaminant concentrations peak shortly after flooding and then decrease exponentially, before eventually reaching asymptotic conditions. This phenomenon has been observed in flooded mine workings worldwide, including the USA (Mack et al., 2010), Poland (Gzyl and Banks, 2007), and South Africa (Wolkersdorfer, 2008; Huisamen and Wolkersdorfer, 2016). Gzyl and Banks (2007) and Perry and Rauch (2012) developed and validated empirical first-flush based models to represent mine water behavior at mine pools in Poland and USA, respectively. However, these models are yet to be employed outside these original mine pools to represent the evolution of mine water quality in underground workings. Furthermore, very few studies exist on forecasting mine water quality using any modeling approach (e.g., Tokoro et al., 2020; Iwasaki et al., 2021), with no existing study on predicting mine water influent quality to treatment plants and its impacts on associated operation and maintenance.

The objective of this study is to predict long-term mine water quality and associated operating expenses at an active mine water treatment plant located in the Sydney Coalfield in Nova Scotia, Canada. The suitability and robustness of empirical 'first-flush' models to represent mine water quality evolution in the flooded mine workings in the Sydney Coalfield are first assessed. The most suitable model is then employed to predict the future quality of mine water influent being pumped into an active

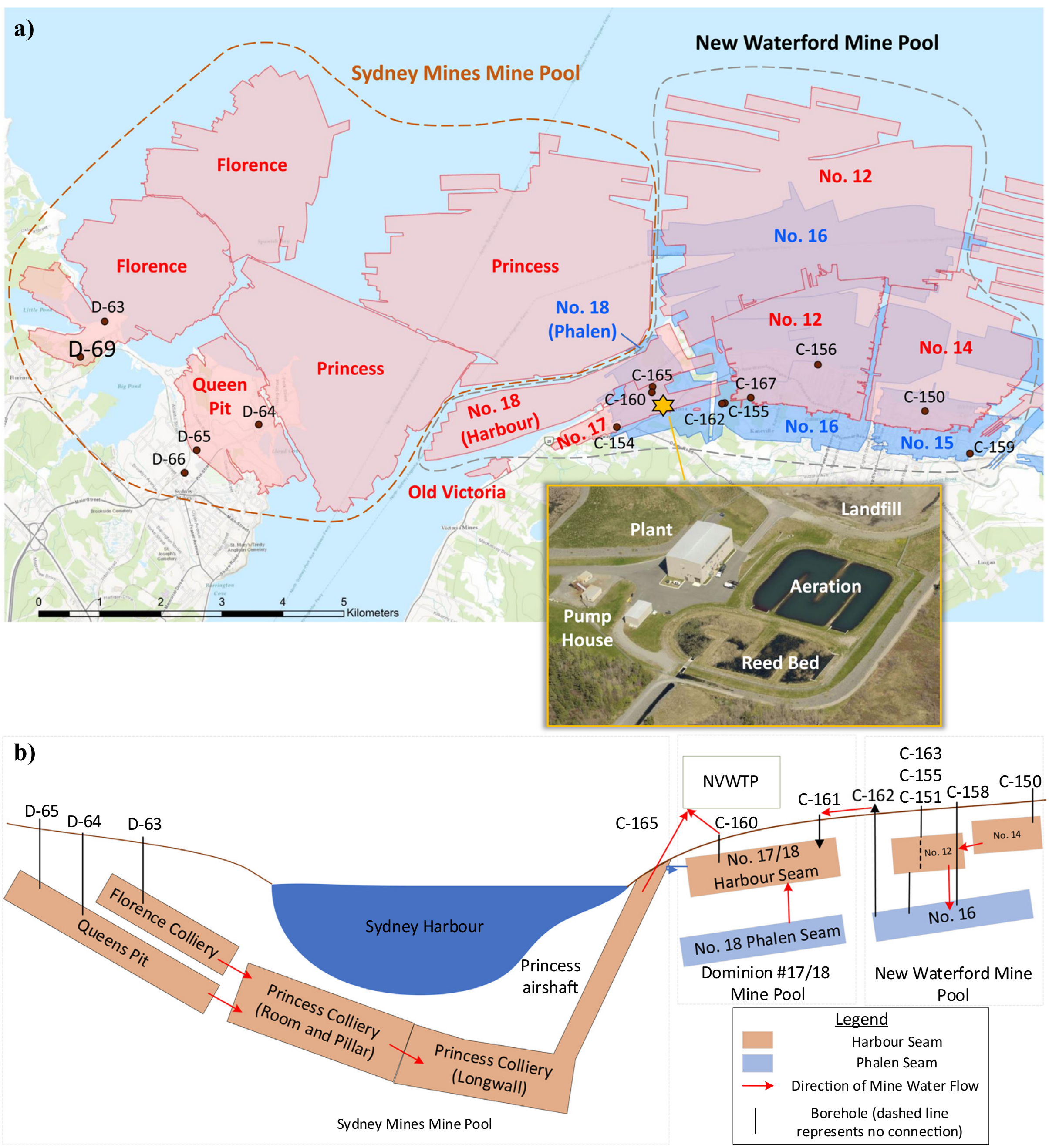

Fig. 2. (a) site map of the Sydney Mines (SM) and New Waterford (NW) mine pools, indicating the location of the New Victoria treatment plant, and (b) conceptual cross-section showing the complex interconnections within the mine pools.

treatment plant, and allow estimates of the associated long-term plant operation costs.

2. Site description

2.1. The Sydney Coalfield

The Sydney Coalfield in Nova Scotia, Canada, is the oldest mined coalfield in North America, with underground mining occurring from the early 1700s to the early 2000s. The coalfield is composed almost entirely of bituminous coal with a total sulfur content between 2.5 and 6 % by weight (wt%) (Hacquebard, 1993; Zodrow, 2005). Mining produced over 2.4 billion tonnes of coal from >50 underground collieries across 11 coal seams. Fig. 1 shows the location of the Sydney Coalfield in Nova Scotia and outlines the key mining collieries.

The long history of mining left behind a complex network of underground workings spanning approximately 2500 km^2. When mining activities ceased, the dewatering pumps were turned off and the workings gradually flooded (i.e., mine water rebound). Many of these workings are hydraulically interconnected, creating several 'mine pools' that consist of multiple connected collieries. The three largest mine pools – Sydney Mines (SM), New Waterford (NW), and 1B – contain workings that produced 450 million tonnes of extracted coal, and left 190 million m^3 of interconnected void space. Flooding of these workings can result in AMD-impacted mine water being released to the environment when its elevation reaches that of discharge points.

In 2001, after mining activities had ceased, a multi-million-dollar mine site closure and reclamation program of the Sydney Coalfield was implemented by Public Services and Procurement Canada (PSPC) (ECBC, 2013). Part of this program was dedicated to the management of mine water and the prevention of AMD release from flooded underground mine workings. Two treatment plants were constructed to manage the three largest mine pools: a passive treatment system at Neville Street for the 1B mine pool (Wolkersdorfer, 2011), and an active treatment system at New Victoria for the SM and NW mine pools.

The Sydney Coalfield is located in a humid continental climate, which is a major climate type of the Köppen classification that exhibits large seasonal temperature contrasts with hot summer and cold winters. The mean annual total precipitation is approximately 1500 mm, while the potential evaporation is 650 mm (Environment Canada, 2022). Precipitation exceeds potential evaporation for most of the year, creating a surplus of water that contributes to overland flow and ground surface infiltration.

2.2. New Victoria treatment plant

Fig. 2a presents a site map of the SM and NW mine pools, associated collieries, and the location of the New Victoria treatment plant. The SM mine pool contains the Princess, Florence, and Queen Pit collieries, all developed within the Harbour Seam, while the NW mine pool contains the Dominion No. 12, No. 14, and No.16 collieries from the Harbour Seam, and Dominion No. 17 and No. 18 from the Phalen Seam. The collieries within the NW mine pool flooded at different times, with Dominion No. 17 and No. 18 flooding in 1972, and No. 16 flooding in 2009. In 2008, water level data from monitoring wells installed within each mine pool indicated that mine water would soon reach discharge elevations and release to the environment. This inspired the design and construction of the New Victoria treatment plant, which has been fully operational since 2011.

Fig. 2b presents a schematic of the various collieries, interconnections, and mine water flow pathways from the mine pools to the treatment plant. The SM mine pool was connected to the plant via an old airshaft tunnel. A well (C-165) was drilled into the tunnel at 127 ft. below sea level (ft. bsl), which then connects to the SM mine pool at a depth of 1318 ft. bsl. To allow mine water to be pumped from the entire NW mine pool to the plant, various boreholes were installed to supplement existing interconnections. Well C-160 was then drilled to 385 ft. bsl into the No.17 colliery. A dedicated pump house was constructed, and wells C-160 and C-165 were instrumented with large mechanical pumps to extract the mine water from both mine pools. The desired elevation ranges for the mine water were determined from expected ground surface discharge elevations and mine pool infilling rates from ground and surface water (there is no evidence that seawater is entering the collieries). Water level data, along with historical data from the mine working plans, determined that the infilling rates for the NW mine pool and SM mine pool were 42 m^3/h (184 US gpm) and 44 m^3/h (193 US gpm), respectively. Pumping operations at the plant were based on maintaining the mine water between the upper and lower elevations established.

The New Victoria treatment plant includes a high-density sludge (HDS) system utilizing aeration, hydrated lime dosing for alkalinity addition, and a settling pond and reed bed for polishing. Following treatment, the treated water is discharged to the ocean, while the generated sludge is placed in large geotextile sludge bags for compression and dewatering, and placed in a dedicated landfill adjacent to the plant. An aerial photograph of the treatment plant is shown in Fig. 2. The treatment plant was designed for a peak combined inflow rate of 114 m^3/h. Monitoring wells in the SM and NW mine pools are instrumented with water level loggers that are powered full-time to continuously inform the treatment plant of mine water elevations. Plant operations then ensure that water elevations remain below potential discharge elevations.

3. Materials and methods

3.1. Mine water geochemical behavior

3.1.1. First-flush phenomenon

Younger (2000) examined the long-term behavior of mine water in numerous coal mines in the UK and derived the 'first-flush' phenomenon,

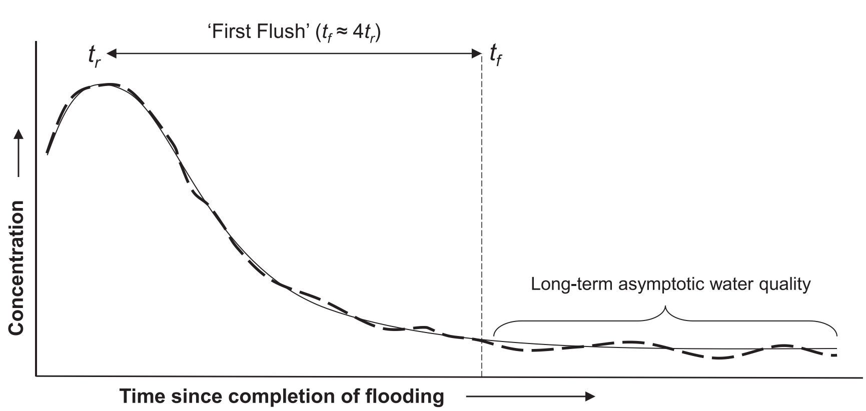

Fig. 3. Conceptual model of the 'first-flush' that shows a peak in contaminant concentration followed by a rapid improvement leading to an eventual steady-state, asymptotic concentration. Note that t_r represents the initial time for the workings to flood, and t_f represents the duration of the first-flush. Modified from Younger (2000).

where contaminant concentrations peak shortly after flooding and then decrease exponentially, before eventually reaching asymptotic conditions. Fig. 3 illustrates the first-flush mine water behavior. This phenomenon has been observed in flooded mine workings worldwide, including the USA (Mack et al., 2010; Mountjoy, 2018), Poland (Gzyl and Banks, 2007), and South Africa (Wolkersdorfer, 2008; Huisamen and Wolkersdorfer, 2016).

3.1.2. First-flush modeling

Empirical models have been developed to represent first-flush mine water behavior and are employed to predict the long-term quality of mine water being pumped from the SM and NW mine pools to the treatment plant. Using long-term sulfate measurements from flooded sections at the Siersza mine in Poland, Gzyl and Banks (2007) developed the following single-phase model, hereafter referred to as the 'G&B model', that is based on the exponential decay of mine water quality to a long-term asymptotic concentration:

$$C = C_v * \exp(m*t) + C_b, \quad (1)$$

where C_v is the peak (or in this case, initial) concentration at $t = 0$, m is the slope of the curve, t is the time, and C_b is the background (asymptotic) concentration, which is estimated to be 90 % of C_v. It should be noted that this equation starts at the peak concentration of the 'first-flush' curve and only models the subsequent decay over time.

Using historical data from five mine pools in West Virginia, USA, Perry and Rauch (2012) developed a similar model to Gzyl and Banks (2007), hereafter referred to as the 'P&R single-phase model', but suggests that mine water quality continues to improve beyond asymptotic concentrations (i.e., to zero concentrations):

$$C = C_v * \exp(m*t) \quad (2)$$

Perry and Rauch (2012) also proposed a 'two-phase' model ('P&R dual-phase model') with different concentration decay slopes at different time periods, as shown in Eqs. (3a) and (3b). The first phase describes a steep, initial decline in concentration, and the second phase describes a long-term, significantly slower decline.

$$C = C_{v,1} * \exp(m_1 * t) \text{ when } t < t_{transition}, \quad (3a)$$

$$C = C_{v,2} * \exp(m_2 * t) \text{ when } t \geq t_{transition}, \quad (3b)$$

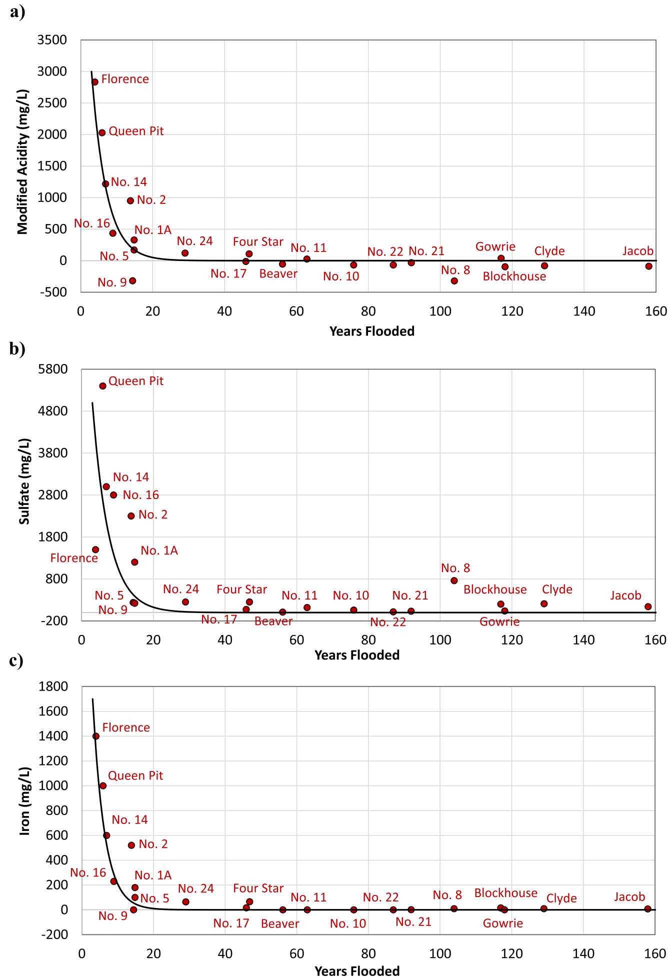

Fig. 4. Mine water quality in each colliery within the Sydney Coalfield versus the number of years that the colliery has been flooded for: (a) modified acidity, (b) sulfate, and (c) dissolved iron concentrations. The black polyline indicates the approximate evolution of water quality over time, which resembles first-flush behavior.

Table 1
Summary of the mine pools, monitoring wells and sampling in the Sydney Coalfield.

Mine pool	Colliery	Year closed	Year flooded	# Wells	# Samples	Earliest sample	Latest sample
NW	No. 12	1973	2011	4	8	4-Sep-08	29-Jul-19
	No. 14	1973	2011	2	9	12-Mar-10	30-Oct-19
	No. 16	1962	2009	4	66	28-Apr-08	19-Aug-19
	No. 17	1921	1972	3	68	21-Feb-05	18-Dec-17
	No. 18	1921	1972	1	4	10-Feb-05	8-Sep-12
	NV plant	–	–	1	204	19-Jan-11	6-Apr-22
SM	Queen Pit	1917	2014	2	12	5-May-09	19-Dec-17
	Florence	1961	2014	3	17	30-Aug-11	15-Mar-19
	NV plant	–	–	1	163	6-Mar-14	6-Apr-22
1B	No.10/11	1949	1955	1	6	22-May-08	1-Dec-17
	No. 1A	Unknown	2003	6	60	24-Nov-03	30-Nov-17
	No. 2	Unknown	2003	1	4	14-Apr-10	21-Nov-16
	No. 24	1953	1981	1	2	23-Sep-08	4-Dec-17
	No. 5	Unknown	2003	7	106	8-Apr-03	28-Nov-17
	No. 9	Unknown	2003	1	4	19-Dec-12	12-Jun-17
	No. 5	Unknown	2003	9	127	21-Feb-03	24-Jan-18
–	No. 8	1914	1917	3	6	12-Mar-07	1-Dec-17
–	Four Star	1969	1970	1	3	26-Jun-09	23-Nov-16
–	Blockhouse	1888	1890	1	10	25-Jun-09	11-Dec-17
–	Gowrie	1897	1901	1	12	25-Jun-09	7-Dec-17
No. 21/22	No. 21	1925	1926	2	23	25-Jun-09	8-Dec-17
	No. 22	1930	1931	3	36	15-May-09	8-Dec-17
–	No. 15	1925	1931	1	6	5-May-09	18-Dec-17

where $C_{v,1}$ is the initial (peak) concentration, m_1 is the initial steep slope, t is time, $C_{v,2}$ is the concentration at the transition between the first and second decay slopes, m_2 is the long-term shallow slope, and $t_{ransition}$ is the time where the model changes from the first to the second decay slope.

3.2. Mine water modeling

3.2.1. Geochemical sampling

Over the past 20 years, numerous monitoring wells have been installed into the various collieries across the Sydney Coalfield. Mine water levels and water samples (for geochemical analysis) have been collected from these wells at various times and frequencies, with a summary provided in Table 1. This sampling data has been used by PSPC to support the management of mine water across the coalfield, including the design and construction of the New Victoria treatment plant.

A general water chemistry analysis (Rapid Chemical Analysis package plus metals scan, RCAp-MS) has been performed on all collected samples to determine a range of parameters such as pH, modified acidity, standard acidity, alkalinity, electrical conductivity (EC), sulfate, chloride, and metals (total and dissolved). The geochemical parameters that best characterize AMD-impacted water include modified acidity, sulfate, iron (Fe), aluminum (Al), manganese (Mn), pH and alkalinity (e.g., Power et al., 2017, 2018). For any samples that have not been directly analyzed for modified acidity, the following well-established and reliable equation to estimate acidity from pH and the sum of the milliequivalents of the dissolved metals, is used (Park et al., 2015):

$$Net\ Acidity = 50*\left(2*\frac{[Fe]}{55.85} + 3*\frac{[Al]}{27} + 2*\frac{[Mn]}{54.94} + 10^{(3-pH)}\right) - alkalinity, \quad (4)$$

where 50 is the equivalent weight of $CaCO_3$, which converts the acidity in milliequivalents per litre into milligrams per litre of $CaCO_3$ equivalent.

In this study, three parameters are used to assess mine water behavior: sulfate, modified acidity and dissolved iron. Therefore, while the first-flush models were generated from one parameter, such as the model by Gzyl and Banks (2007) that was based on sulfate concentrations, this study also verifies the mine water behavior and the robustness of first-flush models for several key AMD indicator parameters.

3.2.2. Long-term historical behavior

The sampling data summarized in Table 1 was assembled and compiled into a comprehensive historical database for detailed analysis and interpretation. As shown in Table 1, mine water samples have been collected from various mine collieries that have flooded at significantly different times. For example, the Gowrie mine colliery flooded in 1901, with the most recent sample collected in 2017, thereby providing mine water quality corresponding with a flooding time of 116 years. In contrast, the SM mine pool, which contains the Queen Pit, Florence, and Princess collieries, flooded in 2014 and provides mine water quality after only eight years of flooding.

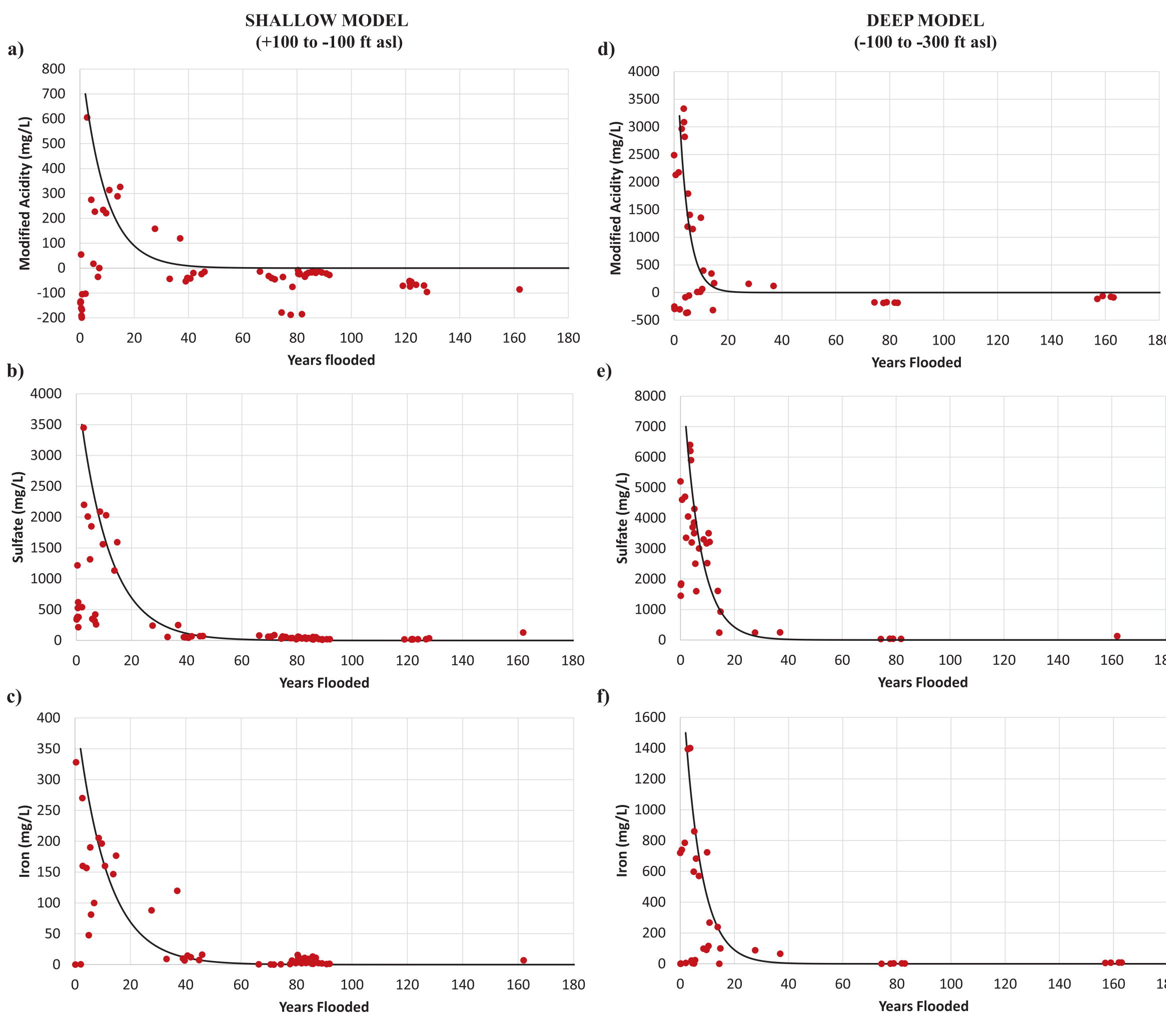

Fig. 5. Mine water quality versus the number of years flooded for modified acidity, sulfate, and iron for the (a-c) shallow model, and the (d-f) deep model. Note that the concentration ranges (y-axes) for each parameter are different for the shallow model and the deep model.

This historical database allows mine water quality and its relationship to flooding time to be examined, and help determine whether mine water quality in the abandoned workings in the Sydney Coalfield is following first-flush behavior. Furthermore, the existence of mine water stratification within the flooded workings is investigated and whether mine of similar water quality that resides in each stratified layer is also following first-flush behavior.

3.2.3. Future treatment plant influent quality

If first-flush behavior is occurring within the coalfied, the empirical models are then employed to predict the long-term quality of mine water influent to the New Victoria treatment plant. First, the samples collected from historical wells installed within the SM and NW mine pools (i.e., not the 'newer' pumping wells for the treatment plant) are used to assess the evolving mine water behavior specific to these mine pools. Plots of mine water quality are generated to assess the associated mine water quality decay curves. Sampling data collected from the two wells pumping mine water to the plant (i.e., C-160 and C-165) are not included in these curves. Each of the empirical models in Eqs. (1) to (3b) are then calibrated to the observed SM and NW curve data to determine the optimal value of the following parameters: (i) peak (initial) concentration, (ii) decay slopes, (iii) initial year (time zero), and (iv) for the two-phase model, the year where the decay slope transitions between the first and second decay slopes. The Sensitivity Toolkit, the Microsoft Excel add-in that can be used for optimizing sensitivity analysis, is employed to adjust each parameter until the global root mean square (RMS) error between observed and modeled data is minimized.

As shown in Table 1, bi-weekly samples of the mine water being pumped to the plant from the SM and NW mine pools have been collected. The SM mine pool flooded in 2014, with 163 samples collected at the pumping well C-165 between 2014 and 2022. In the NW mine pool, No. 17 and No. 18 flooded in 1972, while No. 16 flooded in 2009. Although the mine water from the NW mine pool is being pumped from No. 17 (via

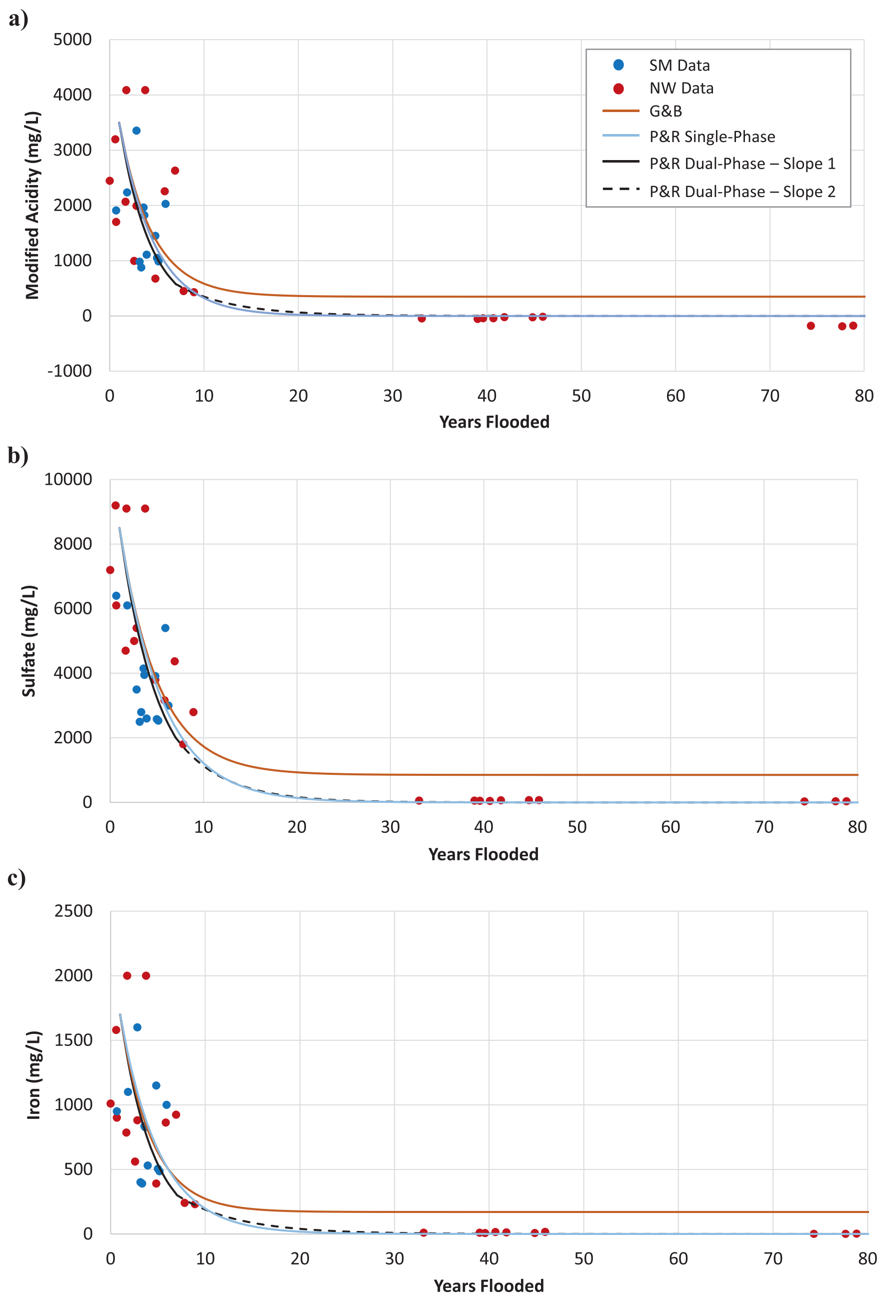

Fig. 6. Historical mine water quality in the SM and NW mine pools for a) modified acidity, b) sulfate and c) iron. Also shown are the calibrated curves for each of the three empirical models.

C-160), this colliery is constantly being recharged with water from No. 16 (see Fig. 2). Therefore, it is difficult to ascertain the exact flooding time of the water from the NW mine pool. Sample collection commenced in 2011 when the plant opened, and a total of 204 samples were collected between 2011 and 2022 from C-160. The optimal first-flush model and associated parameters that are determined from the model calibration discussed earlier are applied to the existing SM and NW water quality to predict future mine water quality being pumped to the plant.

3.3. Treatment cost

Knowledge on future mine water quality can be used to forecast the long-term operational expenses that will be incurred by the treatment plant. In this study, the focus is on expenses associated with lime usage, since this is directly influenced by the quality of mine water influent, and accounts for >50 % of total treatment plant costs each year. Using data that has been collected at the plant since it began operating, a correlation is made between lime consumption and acidity loading to the plant, which is based on the modified acidity of mine water and the pumping rate to the plant. The following equation is used for lime usage costs:

$$LimeCost = \left[\left((Q_{SM} \times C_{SM}) + (Q_{NW} \times C_{NW})\right)\right] \times \frac{M_{lime}}{M_{acidity}} \times Cost_{lime}, \qquad (5)$$

where Q_{SM} and Q_{NW} are the pumping rates [L/s] from the SM and NW mine pools, respectively, C_{SM} and C_{NW} are the modified acidity concentrations [mg/L] in the mine water from SM and NW, respectively, $M_{lime}/M_{acidity}$ is the correlation between the mass of lime required to treat the corresponding mass of acidity [kg/kg], and $Cost_{lime}$ is the estimated cost of lime per kg [CA$/kg]. The flow rates for SM and NW are approximately 44 % and 56 % of the total flow rate going into the plant, respectively, with the average total flow rate throughout plant operations approximately 90 m^3/h. Based on the actual lime costs occurring at the plant, the cost of lime is estimated at CA$500 per 1000 kg (tonne). Eq. (5) is then applied to the predicted mine water acidity concentrations to project costs associated with lime usage over the long term.

4. Results and discussion

4.1. First-flush behavior

4.1.1. The Sydney Coalfield

Mine water samples collected from the various collieries across the Sydney Coalfield in 2017 are analyzed for key AMD parameters. Fig. 4 plots modified acidity, sulfate and iron concentrations for each colliery sample against the number of years that the corresponding colliery has been flooded. A black polyline is superimposed on each plot to illustrate the approximate evolution of mine water quality. It is evident from all three parameters that mine water behavior in the Sydney Coalfield can be reliably described by the 'first-flush' phenomenon.

4.1.2. Shallow and deep mine water

Mine water quality evolution occurring in the Sydney Coalfield is further examined to determine if first-flush phenomenon can also describe mine water behavior at different depth ranges, particularly as mine water within the coalfield has become stratified over time. Historical mine water samples collected across the coalfield over time (i.e., not just single colliery samples in 2017) are categorized by the depth at which they were collected. For example, the depth associated with mine water samples collected from Well A-49 in the Blockhouse colliery is 36 ft. (bsl), while the depth at Well D-63 in the Florence colliery is 268 ft. bsl. Two depth range categories are generated: (i) 100 ft. above sea level (asl) to 100 ft. bsl, hereafter referred to as the 'shallow model', and (ii) 101 ft. bsl to 300 ft. bsl, hereafter referred to as the 'deep model'.

Fig. 5 presents the mine water quality across the coalfield for both the shallow model and deep model. Water quality in both models demonstrate first-flush behavior, even though they exhibit different peak concentrations and decay rates. For instance, the shallow model in Fig. 5a indicates that the peak acidity concentration is ~600 mg/L, while the deep model in Fig. 5d shows the peak acidity concentration at ~3300 mg/L. The peak concentrations for sulfate and iron also display considerable magnitude differences between the shallow and deep models. These distinct contrasts in mine water quality also confirm the occurrence of mine water stratification in the Sydney Coalfield.

The shallow and deep models also demonstrate differences in decay rates, which confirms the suggestion by Perry and Rauch (2012) that higher mine water quality may be slower to improve than lower mine water quality. The shallow model takes approximately 40 years to decline from peak concentrations to zero for all three parameters shown, while the deep model takes approximately 20 years. Therefore, while the water quality in the shallow model is of better quality, the first-flush is slower. This may be due to the upward movement of contaminants from deeper workings through complex advective or convective processes (e.g., Elliot and Younger, 2014; Mugova and Wolkersdorfer, 2022).

4.2. Long-term predictions of plant influent quality

4.2.1. Empirical model calibration

The evolution of mine water quality within the Sydney Coalfield has been shown to follow first-flush behavior over time. Therefore, the first-flush models can be employed for long-term predictions of mine water quality being pumped to the New Victoria treatment plant from the SM and NW mine pools. The models now first need to be calibrated to historical mine water behavior occurring specifically within the SM and NW mine pools to determine, and optimize, the model input parameters (e.g., mine water decay slope). Fig. 6 plots the modified acidity, sulfate and iron

Table 2
Summary of calibration results showing the range of parameter values tested for each model, the final parameter value and the R^2 and d values.

Model	Parameter	Cv range (mg/L)	Slope range (/month)	Final Cv (mg/L)	Final slope (/month)	R^2	d
G&B	Acidity	2000–4000[a]	−0.01 to −0.06[c]	3500	−0.024	0.59	0.77
	Sulfate	5500–10000[b]		8500	−0.020	0.69	0.86
	Iron	1000–2000[a]		1700	−0.025	0.60	0.78
P&R single-phase	Acidity	2000–4000	−0.01 to −0.06	3500	−0.020	0.56	0.82
	Sulfate	5500–10,000		8500	−0.018	0.66	0.90
	Iron	1000–2000		1700	−0.020	0.56	0.84
P&R dual-phase (phase 1)	Acidity	2000–4000	−0.01 to −0.06	3500	−0.025	0.54	0.77
	Sulfate	5500–10,000		8500	−0.020	0.65	0.89
	Iron	1000–2000		1700	−0.024	0.53	0.81
P&R dual-phase (phase 2)	Acidity	2000–4000	−0.01 to −0.06	578	−0.014		
	Sulfate	5500–10,000		2013	−0.016		
	Iron	1000–2000		301	−0.013		

[a] Modified acidity and iron tested at 100 mg/L increments.
[b] Sulfate tested at 500 mg/L increments.
[c] Slopes were tested at 0.001 increments.

concentrations versus flooding time for all samples collected from wells located across the SM and NW mine pools (blue and red circles). Each of the three models – G&B, P&R single-phase, and P&R dual-phase – are then applied to these plots.

The values of model input parameters – peak concentration (C_v), year of peak concentration (t_v), decay slope (m), and year of decay slope transition ($t_{transistion}$) – are altered across a large range of values during calibration. Qualitative and quantitative analysis of each 'tested' model curve is performed until optimal parameter values are obtained. Table 2 presents the range of parameter values that have been tested for each model, along with the final parameter value. The associated R-squared (R^2) and index of agreement (d) values between the final model data (curve) and the field data are also shown.

As shown in Fig. 6, the G&B model matches the field data for the early, rapid decline phase (<10 years), but differs in the later, slow decline phase. Gzyl and Banks (2007) had suggested that mine water quality goes to long-term asymptotic concentrations, but the mine water quality throughout the Sydney Coalfield (Fig. 4) and the SM and NW mine pools (Fig. 6) are shown to decline to zero concentrations, or negative concentrations in the case of modified acidity (i.e., net alkaline). The P&R single-phase model is

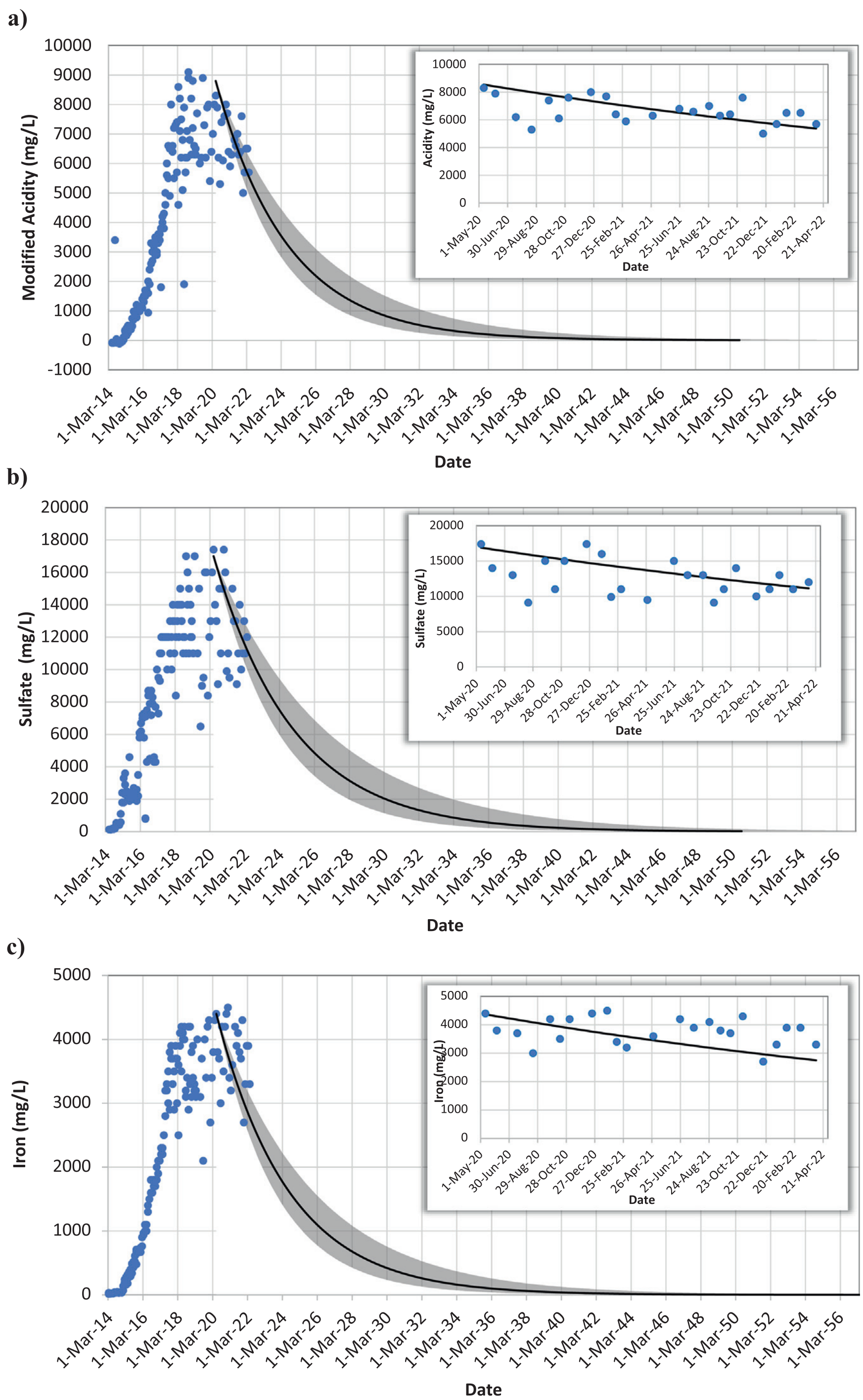

Fig. 7. Long-term predictions of (a) modified acidity, (b) sulfate, and (c) iron for mine water influent from the SM mine pool. The black line is the predicted water quality, while the upper and lower limits are represented by the grey band. Also shown are the correlations between modeled data and any observed data along the first-flush curve (inset).

similar to the G&B model on the early rapid decline, but is significantly better at matching the long-term mine water quality (>10 years) since it assumes the mine water quality declines to zero concentrations over time. As shown in Table 2, the R^2 and *d* values also demonstrate the improved modeling provided by P&R single-phase model. For example, the *d* values for modified acidity, sulfate and iron are 0.77, 0.86 and 0.78 for the G&B model, respectively, and 0.82, 0.90, and 0.84 for the P&R single-phase model.

The P&R dual-phase model provides similar results to the P&R single-phase model. However, it should be noted that this is only possible as it has been calibrated to a complete field data curve (i.e., peak to zero concentration) where its unique input parameters, such as concentration and time at the transition between the first and second slopes of the decay curve, can be obtained. As the P&R single-phase uses a single decay slope from peak to zero concentrations, it can be applied to field data at any stage of the first-flush curve. In contrast, the P&R dual-phase model requires field data after the initial rapid decay slope is complete, so that the transition to the slower decay slope can be determined. The dual-phase model is most suitable for mines that have been flooded for a long time and the initial rapid decay is complete, as is the case for the historical data for the SM and NW mine pools shown in Fig. 6. As this model assigns separate slope values for the initial rapid decline and the later slow decline, the P&R dual-phase model may be more accurate than the one slope permitted in the P&R single-phase model.

As the objective of this study is to predict future mine water quality entering the treatment plant, the first-flush curve associated with the plant influent quality is still being 'created' (e.g., current mine water quality lies along the early, rapid decline portion of the curve). Field data has only been collected since the plant opened in 2011, while the SM mine pool only flooded in 2014. Therefore, based on the analysis of mine water behavior within the SM and NW mine pools, the P&R single-phase model is the most appropriate for long-term predictions of plant influent quality.

4.2.2. Sydney mines mine pool influent

The quality of the mine water being pumped to the plant from the SM mine pool (via Well C-165) has been recorded since the mine pool flooded in early 2014. Fig. 7 plots the concentrations of modified acidity, sulfate and iron between 06 March 2014 and 6 April 2022 (blue circles). All three parameters indicate approximately zero concentrations immediately after flooding, which then gradually increase to peak concentrations by early 2020. It is noted that mine water quality fluctuates around the larger peak concentrations, making it difficult to define the exact date of peak concentration from just one parameter. Therefore, the behavior and trend of all mine water quality parameters are analyzed together to ensure a representative date of peak concentration is selected. This is estimated to occur on 01 May 2020 for all parameters, followed by a gradual decline in concentrations.

The P&R single-phase model is then applied to each parameter in Fig. 7 to predict the future mine water quality from the estimated date of peak concentration. The associated peak concentrations for modified acidity, sulfate and iron are 8600 mg/L, 17000 mg/L and 4400 mg/L, respectively. As determined from the P&R single-phase model calibration (see Table 2), decay slope values for modified acidity, sulfate and iron are −0.020, −0.018 and −0.020, respectively. Table 3 presents the P&R single-phase model input parameters.

The predicted concentrations of modified acidity, sulfate and iron over the long-term are shown in Fig. 7 (black line). To assess how accurate the model predictions are, predicted concentrations are compared to available observed values along the decay curve. This comparison for each AMD parameter is also shown in Fig. 7 (inset), and it is evident that the modeled data matched the general decreasing trend of the observed data.

As shown in Fig. 7a, the modified acidity of the influent mine water from the SM mine pool is predicted to decrease to 1000 mg/L by the year 2029, and then to 100 mg/L by 2039. Similar trends are predicted for sulfate and iron concentrations in Fig. 7b and c, respectively, with sulfate declining to <100 mg/L by 2044, and iron declining to <100 mg/L by 2036.

Fig. 7 also shows the upper concentration limit (UCL) and lower concentration limit (LCL) for each parameter (grey band), which is based on positive and negative changes of 0.005 in the decay slope value. For instance, the acidity decay slope is −0.020, and the UCL and LCL are −0.025 and −0.015, respectively. The earlier calibration process used a range of decay slope values (i.e., −0.01 to 0.06) and this variation of 0.005 was shown to maintain suitable model results for all AMD parameters.

Table 3
Model input parameters for predicting the long-term quality of mine water influent to the plant from the SM and NW mine pools.

AMD parameter	Model input parameter	Sydney Mines	New Waterford
Acidity	C_v	8600	5000
	Date of C_v	01 May 2020	01 Jan 2008
	Decay Slope	−0.020	−0.020
Sulfate	C_v	17,000	13,000
	Date of C_v	01 May 2020	01 Jan 2008
	Decay Slope	−0.018	−0.018
Iron	C_v	4400	2500
	Date of C_v	01 May 2020	01 Jan 2008
	Decay Slope	−0.020	−0.020

4.2.3. New Waterford mine pool influent

Fig. 8 presents the quality of mine water being pumped to the plant from the NW mine pool (via Well C-160) since the plant opened in 2011. It is evident that this mine water quality is further along the first-flush decay curve than the SM mine pool, which is expected as the NW mine pool has been flooded longer. Peak concentrations have already occurred, along with the initial rapid decay stage of the first-flush.

The P&R single-phase model is employed to predict the future mine water quality being pumped from the NW mine pool. The NW mine pool is estimated to have flooded in 2002, and based on observations of the historical data in the coalfield, the peak concentrations are estimated to occur six years later in 2008. As determined from the P&R single-phase model calibration, decay slope values for modified acidity, sulfate and iron are again −0.020, −0.018 and −0.020, respectively. As the peak concentrations are unknown, the modeled decay curve is adjusted to obtain the best match with the observed field data, estimating peak concentrations for modified acidity, sulfate and iron to be 5000 mg/L, 13000 mg/L and 2500 mg/L, respectively. These are lower than the peak concentrations in the SM mine pool, which is expected as the mine water quality is being pumped at a shallower depth, and therefore has higher quality.

The predicted concentrations for each parameter are shown in Fig. 8. To assess how accurate the model predictions are, predicted concentrations are compared to available observed values along the decay curve. This comparison between modeled and observed data is also shown (inset), and indicates that the modeled data reasonably matches the observed data. The modified acidity and sulfate of the influent are predicted to decrease to 100 mg/L by 2024 and 2035, respectively, while iron concentrations had already declined to 100 mg/L in 2021.

It should be noted that only a reasonable match occurs between the modeled and observed data for the NW mine pool. The various collieries in the NW mine pool flooded at different times, with No. 17 flooding in 1972, and No. 16 flooding in 2009. As the lower quality No. 16 mine water is being discharged into the higher quality No. 17 mine water (Fig. 2), which is then being pumped to the treatment plant, it is difficult to ascertain an exact representation of the time and magnitude of peak concentrations. Furthermore, the NW mine water quality is now on the slower, more gradual decline phase of the curve, and the single decay slope in the P&R single-phase model does not provide the optimal match between modeled and observed data. This suggests that the P&R dual-phase model may be more suitable with two differing decay slope values, but again it is difficult to ascertain the time of transistion between the two slopes.

4.3. Costing

Fig. 9a plots the historical lime usage versus the corresponding acidity loading at the New Victoria treatment plant. The acidity loading is based

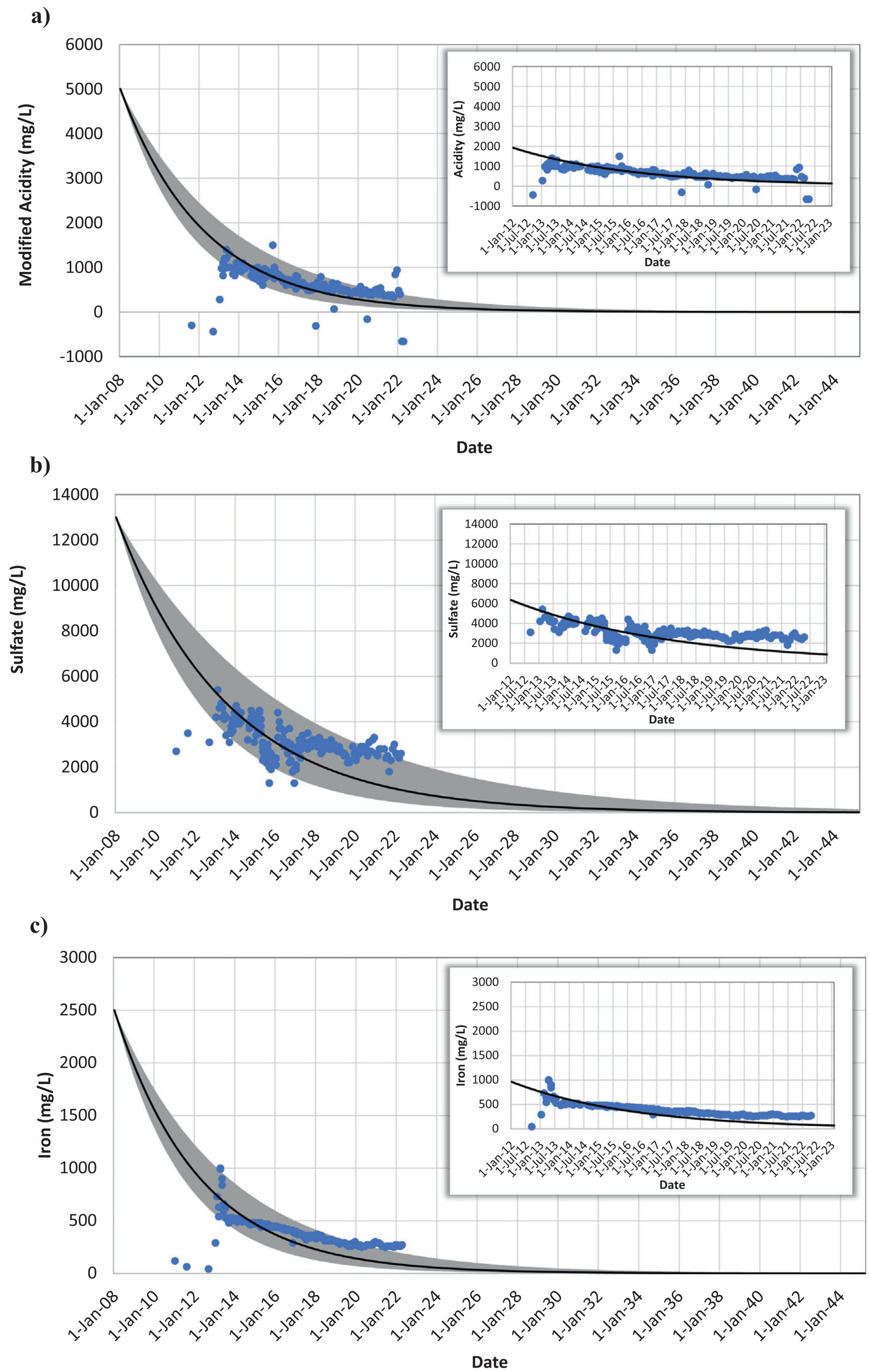

Fig. 8. Long-term predictions of (a) modified acidity, (b) sulfate, and (c) iron for mine water influent from the NW mine pool. The black line is the predicted water quality, while the upper and lower limits are represented by the grey band. Also shown are the correlations between modeled data and any observed data along the first-flush curve (inset).

on the predicted modified acidity of the mine water entering the plant from both mine pools, and the associated pumping rate. As discussed earlier, the average total flow rate is 90 m^3/h, with 56 % coming from the NW mine pool, and 44 % from the SM mine pool. As shown in Fig. 9a, the volume of lime consumed is closely correlated to the acidity of mine water entering the plant. It is estimated that 0.98 kg of lime is required to neutralize 1 kg of acidity load entering the plant.

Using Eq. (5), the cost associated with lime usage over the long-term is determined. This does not include inflation, changes in utility costs, or any other related operational and maintenance expenses. Fig. 9b presents the projected cost for lime usage at the plant over time, along with upper and lower confidence intervals that are based on the UCL and LCL of modified acidity in Figs. 7 and 8. It is predicted that lime expenditure will decrease by approximately 50 % in 2025. By 2037, the lime cost will be less than $35,000 per year, which corresponds to an average daily loading of approximately 190 kg/day, or modified acidity concentration of 100 mg/L (assuming current pumping rates remain the same). A transition to passive treatment could occur at this time (e.g., Trumm, 2010).

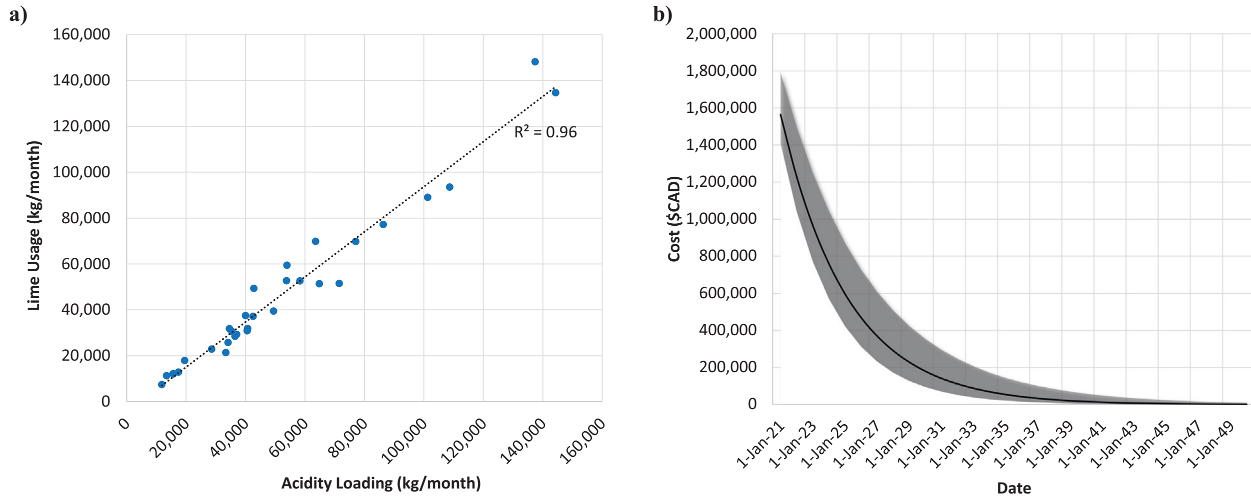

Fig. 9. (a) historical lime usage versus corresponding acidity loading at the treatment plant, with the associated correlation indicating that 0.98 kg of lime is needed to neutralize 1 kg of acidity, and (b) long-term predictions of lime usage costs at the plant.

5. Conclusions

This study assessed the robustness of empirical ‘first-flush’ models for representing mine water behavior in flooded underground workings in the Sydney Coalfield in Nova Scotia, Canada, before being employed, for the first time, to predict the evolution of mine water quality being pumped to an active mine water treatment plant. While numerical models are commonplace across a range of hydrogeological and geoenvironmental investigations, the complexity, heterogeneity, and unknowns in underground mine workings provide challenges for numerical modeling, making them more suitable for when a deep hydrogeologic understanding within the workings is necessary. Instead of trying to model the physical, hydraulic, and geochemical processes that may be causing changes in mine water quality, empirical models focus on the resulting mine water quality and the behavior and trends that have been established from decades of data from coalfields worldwide. Therefore, despite its simplicity and ease of use, empirical models can be more suitable when a low-cost approach is needed.

The historical AMD-impacted mine water quality samples taken from each colliery across the Sydney Coalfield were analyzed and correlated to the year that the specific colliery flooded, which ranged from 5 years to 100+ years. Concentrations of common AMD indicator parameters – modified acidity, sulfate and iron – were plotted against years flooded, and confirmed that mine water quality in the coalfield follows ‘first-flush’ behavior. Furthermore, first-flush behavior was evident in stratified layers of varying mine water quality.

First-flush based models were then employed for predicting the quality of mine water being pumped to the active treatment plant in New Victoria, which is being used to extract and treat the mine water in the Sydney Mines (SM) and New Waterford (NW) mine pools. The empirical models were first calibrated against historical mine water quality in the SM and NW mine pools to determine the long-term decay rates, which were then used as input parameters for the long-term model predictions. The predicted concentrations of modified acidity, sulfate and iron for each mine pool indicated that the concentrations of mine water influent will collectively reach concentrations by the year 2037. This may be acceptable for active treatment to cease, and a transition to either passive treatment or direct marine discharge. The predicted acidity concentrations and loadings were then used to determine the associated long-term lime dosages and costs required for the active treatment process.

This study demonstrates that first-flush phenomena can accurately represent evolving mine water behavior across the historical Sydney Coalfield. While the associated empirical models are easy-to-use, they are highly appropriate for predicting the quality of mine water over the long-term, either naturally within the mine pools, or when the mine water is being pumped to treatment plants. Future predictions of mine water contaminant loadings, either discharging to the environment, or being pumped to treatment plants, are highly beneficial to the various stakeholders of abandoned coalfields, including site owners, regulators, treatment plant designers, and plant operators. Future work will focus on the stability of observed mine water stratification, and the feasibility of pumping and treating mine water from shallower workings.

CRediT authorship contribution statement

Patrick Merritt: wrote original draft, field sample collection, modeling, data analysis and interpretation. **Christopher Power:** funding acquisition, supervision, assisted with data collection, analysis and interpretation, revised drafts.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Christopher Power reports financial support was provided by Mitacs Canada.

Acknowledgements

The authors would like to give a special thanks to Joseph MacPhee and Glenn MacLeod at Public Services and Procurement Canada (PSPC) for their technical expertise and logistical support, along with Beth Mason at the Verschuren Centre for Sustainability in Energy and the Environment, Nova Scotia, Canada. We would also like to acknowledge the various consultants at CBCL Limited and exp. Consultants, including Richard Morykot, Steven Schaller and Arman Polatbekov, for providing some of the historical data presented in this study. This work was partly funded through a Mitacs Accelerate grant (IT14959) in partnership with CBCL Limited.

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