Mine Water Environ (2015) 34:3–9
DOI 10.1007/s10230-014-0320-6

REVIEW

# Carbon Dioxide Dynamics and Sequestration in Mine Water and Waste

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Received: 8 August 2014 / Accepted: 10 November 2014 / Published online: 23 December 2014

**Abstract** The role and importance of $CO_2$ in the mining sector has been overlooked until relatively recently. This review presents the complexities of $CO_2$ and mine water evolution. Carbon sequestration using mine waters and solid wastes and recent research on the profound impacts of dissolved $CO_2$ on active and passive treatment were reviewed. The literature indicates great promise for more efficient and fiscally competitive operations, lower environmental impacts, and a decreased carbon footprint for such operations. However, a tremendous amount of research and field testing is necessary to move many of these approaches forward to full scale common application.

**Keywords** Iron oxidation · Acid mine drainage · Passive treatment · Active treatment · Carbon footprint

## Introduction

$CO_2$ is central to many critical global biogeochemical processes that are integral to mine water and waste remediation. The geologic setting of associated minerals and the pH of the resulting water dictates the amount of $CO_2$ dissolved in acid mine drainage (AMD). $CO_2$ also plays a role in the chemistry of AMD production and treatment, for dissolved $CO_2$ can confound water quality measurement and change the requirements for passive or active treatment. More broadly, mine sites and AMD are significant sources of atmospheric $CO_2$ emissions, while remediation efforts can restore natural carbon assimilation capacity and sequester carbon. The following review provides a broad overview of the current literature with regards to $CO_2$ and mining while introducing the relevant articles within this special issue.

## Chemistry of Carbon Dioxide and Mine Water

Concentrations of $CO_2$ in AMD may be elevated due to reaction of acidic water with limestone or via the oxidation of organic carbon (e.g. Stumm and Morgan 1996). This $CO_2$ reduces the pH of AMD through carbonic acid creation (Eqs. 1–3). At high pH (greater than 11), the hydration reaction (Eq. 4) proceeds rapidly to form bicarbonate, while in solutions at pH less than 8, the hydration reaction reaches equilibrium relatively slowly.

$$CO_2 + H_2O \rightarrow H_2CO_3 \qquad (1)$$

$$H_2CO_3 \rightarrow HCO_3^- + H^+ \qquad (2)$$

$$HCO_3^- + H^+ \rightarrow CO_3^{2-} + H^+ \qquad (3)$$

$$CO_2 + OH^- \rightarrow HCO_3^- \qquad (4)$$

In open systems near sea level, the partial pressure of $CO_2$ is approximately $3.9 \times 10^{-4}$ atm; at 25 °C, the equilibrium constant for Eq. 1 is $1.7 \times 10^{-3}$. In a pure $CO_2$–$H_2O$ system at standard pressure and temperature, the pH would be 5.6. As partial pressure increases, pH is further depressed. Increased partial pressure of $CO_2$ results in greater carbonate solubility and occurs in underground mine voids when carbonate minerals are dissolved by acidic waters (Rose and Cravotta III 1998).

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In AMD impacted streams, $CO_2$ degassing, which increases pH, may be driven by partial pressure differences between the stream and the atmosphere, although this process is not always rapid (Sracek et al. 2010). When dissolved metal concentrations are high, metal hydrolysis produces hydrogen ions, which reduces the dissociation of carbonic acid and drives $CO_2$ degassing (Atekwana and Fonyuy 2009). $CO_2$ in circumneutral AMD can lead to an unstable pH that is easily altered by aeration or sample handling (Kirby and Cravotta 2005b). Paktunc (1999) showed the effect of dissolved $CO_2$ on the apparent neutralization potential of water, where higher $CO_2$ content suggests a carbonate-dominated system and a low $CO_2$ content suggests non-carbonate mineral alkalinity.

$CO_2$ can rapidly degas from AMD with partial pressures of $CO_2$ that exceed atmospheric equilibrium. This can occur when AMD discharges from underground workings or from porous and fractured media in underground mine workings that are within or contain carbonate minerals or organic matter. This degassing is a health and safety risk, as it can produce potentially toxic levels of $CO_2$ in confined spaces (Hall et al. 2006).

Dissolved carbonates are often the product of limestone weathering, while weathering of limestone is also governed by the partial pressure of $CO_2$. Solution saturation with respect to limestone is governed in part by the partial pressure of $CO_2$ (e.g. Gbolo and Lopez 2013):

$$K_{\mathrm{calcite}} = \frac{a_{\mathrm{Ca}^{2+}} \times a^2_{\mathrm{HCO}_3^-}}{P_{\mathrm{CO}_2}} \tag{5}$$

where “a” is the activity of calcium or bicarbonate. The activity of bicarbonate is squared in this relationship to represent the stoichiometry of the most common weathering reaction (Stumm and Morgan 1996).

Increased partial pressure of $CO_2$ has been observed in mine waste heaps (Johnson 2003) and underground mine workings (Hall et al. 2006; Nairn et al. 2010). Cravotta et al. (1994) and Sracek et al. (2010) found increased partial pressures in spoil heaps, attributable to dissolution of calcite, dolomite, and siderite. Other work has suggested that oxidation of organic material below the heap may be an additional source of $CO_2$ (Birkham et al. 2003). The contribution of $CO_2$ to depressed pH should not affect measured alkalinity (Kirby and Cravotta 2005b) and Kirby and Cravotta (2005a) suggest that acidity due to $CO_2$ does not need to be accounted for in AMD treatment due to rapid degassing on exposure to the atmosphere. Hot acidity titration (APHA 1998; Kirby and Cravotta 2005a; McAllan et al. 2009) intentionally drives off dissolved $CO_2$ prior to acidity titration to avoid measurement of ephemeral $CO_2$ acidity. The difference between hot and cold acidity may be used in meta-analysis to show differing $CO_2$ contributions to acidity sources across a mining district (e.g. McAllan et al. 2009; Wood 1996). Conversely, Jarvis (2006) suggests that the kinetics of $CO_2$ degassing may be slow without physical or chemical intervention and that elevated $P_{CO_2}$ may inhibit treatment, if it is not accounted for in design.

The ionization reactions shown in Eqs. 2–4 proceed rapidly. However, the hydration (and dehydration) reaction, Eq. 1, proceeds more slowly (Kern 1960; Stumm and Morgan 1996). Hydration and dehydration reactions are catalyzed by various compounds (e.g. $OH^-$) that may be contained in natural waters (Stumm and Morgan 1996). Dissolved $CO_2$ and degassing rates affect iron oxidation kinetics in AMD (Geroni and Sapsford 2011; Kirby et al. 1999, 2009). As $CO_2$ degasses, pH rises, along with increased rates of $Fe^{2+}$ oxidation and $Fe^{3+}$ precipitation. These processes are described in detail by Cravotta and Geroni (2013).

## Carbon Dioxide and Mine Water Treatment

Dissolved $CO_2$ has been shown to effect the chemical requirements, iron oxidation rates, and sludge production in AMD treatment systems (Geroni and Sapsford 2011; Jageman et al. 1988; Kirby et al. 2009). In AMD with elevated $P_{CO_2}$, the $CO_2$ temporarily contributes to acidity until the water reaches equilibrium with the atmosphere (Kirby and Cravotta 2005a, b).

Dissolved $CO_2$ may lead to overestimation of chemical requirements for neutralization before the $CO_2$ offgasses (Kirby and Cravotta 2005a, b) or, in some cases, increase the quantity of chemicals needed for neutralization in systems when the $CO_2$ does not offgas rapidly (Jageman et al. 1988). Aeration is often used prior to active treatment of AMD to reduce dissolved $CO_2$ (Kirby et al. 2009; Younger et al. 2002) for two key reasons; driving off dissolved $CO_2$ raises solution pH, increasing iron oxidation rates and reducing chemical needs (e.g. Cravotta and Geroni 2013; Geroni and Sapsford 2011; Jageman et al. 1988; Kirby et al. 2009; Younger et al. 2002), and, as shown in Eq. 4, dissolved $CO_2$ consumes hydroxyl ions, increasing the amount of neutralizing material that must be used to increase pH (e.g. Means et al. in press; Younger et al. 2002). After neutralization of mineral acidity (see Hedin et al. 1994; Kirby and Cravotta 2005b), Gehm (1944) increased effluent pH from 4.2 to ≈6.0 by rapidly offgassing $CO_2$ using a 1 m weir. Gehm (1944) also showed that a two minute period of aeration could raise the pH from 4.2 to 7.2 through $CO_2$ offgassing. Cravotta (2007) showed a similar rapid pH increase from 5.8 to 7.2. In this special issue, Means et al. (in press) demonstrate that the effectiveness and chemical consumption of hydrated lime systems may be dramatically improved by $CO_2$ degassing.

Since elevated dissolved $CO_2$ decreases iron oxidation rates (Cravotta and Geroni 2013; Geroni and Sapsford 2011; Kirby et al. 2009), the land area needed for passive treatment of circumneutral AMD is heavily dependent on $P_{CO_2}$ (Kirby et al. 2009). While $CO_2$ entering passive AMD treatment systems affects system sizing, it also has the potential to react within alkalinity-producing passive treatment systems. In steel slag leach beds (SSLBs), circumneutral, low metal water is leached through a vertical flow bed of waste steel slag. This acts to add alkalinity to the water before mixing with AMD. $CO_2$ has the potential to react with $Ca^{2+}$ ions within SSLBs at high pH (typically 11–12), creating $CaCO_3$ precipitates (Goetz and Riefler 2014; Kruse et al. 2012) that reduce hydraulic conductivity of the beds, cement the material, and clog the underdrain system. This has led to system failure, although design changes that offgas dissolved $CO_2$ in the source water and exclude air from the underdrain system improve SSLB operation and lifetime (Goetz and Riefler 2014; Kruse et al. 2012).

In systems that are not open to the atmosphere, elevated $P_{CO_2}$ can increase limestone dissolution (Eq. 6). Watten et al. (2004) applied the concept that elevated $P_{CO_2}$ increases limestone dissolution in closed systems by augmenting a pulsed limestone bed reactor with carbonation. The reactor was successful in a lab setting at generating alkalinity and consuming acidity, while the pilot system raised pH high enough for hydrolysis of Fe(III) and Al(III) (Sibrell et al. 2000; Watten et al. 2004). Cravotta (2007) highlights that $CO_2$ within anoxic limestone drains (ALD) enhances dissolution of limestone due to increased $P_{CO_2}$ and decreased pH, following the relationship shown in Eq. 6. This allows for greater treatment efficiency for a given quantity of limestone compared to systems open to the atmosphere such as open limestone channels (Cravotta 2007).

$$CO_2 + H_2O + CaCO_3 \rightarrow 2HCO_3^- + Ca^{2+} \quad (6)$$

Elevated $P_{CO_2}$ has been observed in many AMD discharges worldwide. Either through natural degassing or physical or chemical aeration or oxidation, AMD degassing $CO_2$ to the atmosphere can contribute a significant amount of $CO_2$ to the atmosphere. Similar results have been reported by Raymond and Oh (2009) for the Susquehanna watershed in Pennsylvania. Their findings indicate that over the past century, AMD discharges in the Susquehanna watershed have released approximately 3.1 Tg of carbon to the atmosphere.

Only recently has it been noted that elevated $P_{CO_2}$ can dramatically impact active treatment operations and thus should influence system design. Decarbonation can be an important primary treatment step that allows pH to increase while reducing hydroxylation. In addition, elevated $P_{CO_2}$ can help cause calcite precipitation, which can dramatically decrease dosing efficiency. The relatively simple step of decarbonation can lead to a savings of nearly ¼ of the hydrated lime necessary for overall treatment.

While $CO_2$ is certainly a major consideration in chemical requirements for both passive and active treatment of AMD, treatment systems are also carbon intensive due to chemical production, energy inputs, and construction emissions. Innovative methods to reduce the carbon footprint of treatment systems, including the reuse of $CO_2$ sequestration by-products (Pérez-López et al. 2010), have been effective, although limited in implementation. The analysis presented by Winfrey et al. (in press) in this special issue specifically analyzes the difference between active and passive treatment systems for Mayer Ranch in Oklahoma using emergy and carbon footprint analyses. The carbon intensive nature of AMD treatment is also reflected by Macy et al. (in press) in this issue.

## Carbon Dioxide Sources and Sinks in Mine Water and Waste

Mine water and waste are a result of mineral extraction; extractive industries are carbon intensive and certainly contribute to increased atmospheric $CO_2$. Beyond the extractive phase, legacy mine lands, AMD, and mine waste may be sources or sinks of $CO_2$. While mining has been and remains carbon intensive, there are efforts both to assess and quantify the carbon footprint of AMD treatment and reuse (e.g. Macy et al. in press; Tuazon and Corder 2008; Winfrey et al. in press) and to find innovative methods to sequester carbon using reclaimed mine lands, mine voids, AMD, and mine waste (e.g. Goetz and Riefler in press; Hedin and Hedin in press; Power et al. 2013a, b; Younger and Mayes in press). Mining also indirectly impacts carbon emissions during mining through combustion or processing of the mineral and during treatment of the resulting wastewater.

Mine lands have the potential to be sources and sinks of $CO_2$. Salm et al. (2012) showed significantly higher $CO_2$ emissions in both active and abandoned peat mining areas than in natural peat bogs, while natural and drained peat bogs had higher $CH_4$ emissions. In Swedish peatlands, Sundh et al. (2000) found similar $CH_4$ emissions during the growing season in mined and natural peatlands, while $CO_2$ emissions were dominated by the mined areas. The soil and biomass carbon lost due to surface mining of previously forested lands has been estimated to account for at least 7 % of power plant emissions for a conventional power plant and at least 70 % of power plant emissions for a plant with $CO_2$ capture and sequestration (Fox and Campbell 2010). Glaesser et al. (2005) showed that flooded mine dumps may also act as a $CO_2$ source, which varies with seasonal factors, lake level fluctuation, and $CO_2$ supply variability (Glaesser and Lerche 2005). While the outgassing rate from the study site in Germany was not significantly higher than reference sites, their analysis showed that $CO_2$ development from the waste dump increased $CO_2$ emissions $\approx 30\ \%$, due to combustion (Glaesser et al. 2005). Hall et al. (2006) similarly showed that AMD was a significant source of $CO_2$ in the presence of both remaining coal and carbonate geology.

$CO_2$ flux from reclaimed mine lands varies with soil properties and soil moisture (Jacinthe and Lal 2006). As reviewed extensively by Shrestha and Lal (2006), reclaimed mine soil has been depleted in soil carbon due to perturbations (Fox and Campbell 2010). However, $CO_2$ sequestration potential depends on the rate of soil carbon development. Dere and Stehouwer (2011) showed that $CO_2$ production on reclaimed mine lands amended with manure and paper-mill sludge may result from microbial processes critical for building stable nitrogen stores in reclaimed mine soils. Reclaimed mine lands are estimated to sequester 0.1–3.1 Mg C $ha^{-1}$ $year^{-1}$ in grasslands and 0.7–4 Mg C $ha^{-1}$ $year^{-1}$ in forests, with the potential to offset 16 Tg $CO_2$ annually in the USA alone (Shrestha and Lal 2006). Tripathi et al. (2014) demonstrated an average carbon sequestration potential of 3.6 Mg C $ha^{-1}$ $year^{-1}$ in a survey of differently aged revegetated mine spoils in India.

Little analysis has been done on the carbon balance of active and passive systems for AMD treatment. While these systems are effective at treating AMD, they do have indirect impacts on the environment through chemical needs, production, transportation, system construction, and energy input to the system. In addition, $CO_2$-rich AMD may either naturally or physico-chemically degas, contributing a greenhouse gas to the atmosphere. Conversely, AMD treatment systems improve the quality of the receiving water bodies, which can increase the biological productivity of the receiving aquatic ecosystems, thus increasing carbon assimilation capacity. In one of the few studies done on this topic, Tuazon and Corder (2008) compared the use of lime to bauxite residue (or “red mud”) in the active treatment of AMD. They found much lower $CO_2$ emissions with the use of red mud. In a case study performed for treatment options at a large coal mine in New Zealand, Hengen et al. (2014) found passive treatment approaches to generally have lower CO2 emissions and overall environmental impacts than active treatment approaches. Macy et al. (in press) demonstrate in this special issue that the increased primary productivity is much smaller than emissions due to treatment. The carbon intensity of AMD treatment is certainly a factor to consider in AMD reuse planning; Macy et al. (in press) compares the carbon footprint of using AMD versus freshwater for hydraulic fracturing.

While carbon emissions have not typically been a factor in choosing between active and passive treatment systems, the analysis presented by Winfrey et al. (in press) in this special issue evaluates this difference using emergy and carbon footprint analyses for the Mayer Ranch site in Oklahoma. This may be an effective method to evaluate the secondary impacts of AMD treatment decisions.

Mining is a carbon intensive industry (Mudd and Diesendorf 2008; Norgate and Haque 2010). However, recent developments show potential for carbon sequestration using AMD, mine wastes, and mine voids that can offset a significant percentage of overall carbon emissions. Precipitation of carbonate minerals may sequester $CO_2$ from solution or the atmosphere, particularly magnesium carbonate species (Harrison et al. 2013; Meyer et al. 2014; Power et al. 2013b; Wilson et al. 2014). Silicate minerals, such as serpentite, are also a possibility for carbonation (Power et al. 2013a). Mineral carbonation of mine tailings and mine wastes shows potential for carbon sequestration in platinum group mineral mine tailings as well (Meyer et al. 2014). Atmospheric $CO_2$ uptake through magnesium carbonate mineral precipitation is rate limited by $CO_2$ diffusion into the water (Wilson et al. 2010). To overcome this limitation, Power et al. (2013b) show the potential for carbonic anhydrase to increase carbon sequestration in magnesium-rich solutions through precipitation of magnesium carbonate minerals. Mills et al. (2010) demonstrated the importance of magnesium sulfate minerals, such as konyaite, on $CO_2$ fixation in mine wastes. Decomposition of konyaite releases magnesium that may precipitate as magnesium carbonate minerals in the presence of atmospheric $CO_2$. While magnesium sulfate minerals may drive $CO_2$ fixation, they can also drive further dissolution of carbonate minerals. For example, sulfate minerals like konyaite can weather to form AMD and may dissolve existing carbonate minerals if present (Mills et al. 2010). Goetz and Riefler (in press) explore $CO_2$ sequestration potential within AMD treatment systems via mineral carbonation in this issue.

Romanov et al. (2009) explored the potential for $CO_2$ storage in shallow underground and surface mines, not only through sorption to coal remaining after mining, but also through placement of abundant materials with potential for $CO_2$ sorption, like clay, into the mine voids to store additional carbon. Likewise, there may be potential for $CO_2$ sorption to residues remaining in underground voids after underground coal gasification, potentially at a higher rate than sorption to coal (Kempka et al. 2001). Younger and Mayes (in press), in this issue, suggest a conceptual design for using abandoned mine pits to sequester $CO_2$ by balancing biological $CO_2$ uptake and decomposition rates by controlled pit flooding. Hedin and Hedin (in press), in this issue, discuss the potential to drive carbon storage in iron-limited regions of the oceans using iron derived from passive AMD treatment systems.

Mining has long term impacts on water quality and the global carbon budget (Raymond and Oh 2009). Rivers worldwide have elevated acidity, sulfate, or metal concentrations due to mine drainage from intensive resource extraction (Strosnider et al. 2014; Wolkersdorfer and Bowell 2004a, b, c). The contribution of mine water and wastes to the global carbon and sulfur budgets is significant. Therefore, the importance of both mine water and wastes as carbon sources and sinks should be considered as the mining industry grows (Raymond and Oh 2009).

## Conclusions

This special issue addresses major questions regarding the sustainability of mining but also reveals the pressing technical need for more efficient AMD treatment, lower impact mining, and innovative carbon sequestration approaches. Field-scale studies, likely supported and hosted by industry, are needed to demonstrate the validity of some of the novel carbon sequestration strategies discussed above. Studies focused on the sustainability metrics of different AMD treatment options and land reclamation strategies are lacking in the literature and needed by policymakers. Developing the ideas presented in this special issue in field-ready research has applications within mining and reclamation. Readily applicable findings ought to be disseminated and applied, especially those that influence the bottom line, such as decarbonation for dramatically improved passive and active treatment performance. This special issue presents current research on the interaction of $CO_2$ and AMD and suggests further research on this important and emerging topic.

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