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Review

Mercury regulation, fate, transport, transformation, and abatement within cement manufacturing facilities: Review

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ABSTRACT

The USEPA's 2010 mercury rule, which would reduce emissions from non-hazardous waste burning cement manufacturing facilities by an estimated 94%, represents a substantial regulatory challenge for the industry. These regulations, based on the performance of facilities that benefit from low concentrations of mercury in their feedstock and fuel inputs (e.g., limestone concentration was less than 25 ppb at each facility), will require non-compliant facilities to develop innovative controls. Control development is difficult because each facility's emissions must be assessed and simple correlation to mercury concentrations in limestone or an assumption of 'typically observed' mercury concentrations in inputs are unsupported by available data. Furthermore, atmospheric emissions are highly variable due to an internal control mechanism that captures and loops mercury between the high-temperature kiln and low-temperature raw materials mill. Two models have been reported to predict emissions; however, they have not been benchmarked against data from the internal components that capture mercury and do not distinguish between mercury species, which have different sorption and desorption properties. Control strategies include technologies applied from other industries and technologies developed specifically for cement facilities. Reported technologies, listed from highest to lowest anticipated mercury removal, include purge of collected dust or raw meal, changes in feedstocks and fuels, wet scrubbing, cleaning of mercury enriched dust, dry sorbent injection, and dry and semi-dry scrubbing. The effectiveness of these technologies is limited by an inadequate understanding of sorption, desorption, and mercury species involved in internal loop mercury control. To comply with the mercury rule and to improve current mercury control technologies and practices, research is needed to advance fundamental knowledge regarding mercury species sorption and desorption dynamics on materials within cement facilities.

Contents

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1. Introduction

While originally named hydrargyrum, Latinized Greek for "water–silver", the present name for the element mercury originates from Mercury, the Roman god. In similarity to the liquid metal's quick watery movement despite its heavy mass, this god was able to fly swiftly with winged sandals and a winged hat. In an unintended and unrealized likeness to Mercury, the god of thieves, the element also hides throughout the world and steals a human body's health (Swiderski, 2008). Prior to knowledge of mercury's neurotoxicity, the element was widely used in medicine. For instance, the elemental form was ingested so that mercury's heavy mass would clear digestive trouble and women consumed mercury during labor in hopes that the substance mass would help push out the fetus. Even Abraham Lincoln took mercury pills to cope with depression (Swiderski, 2008). In early use, mercury appeared widely useful, but history also presents many examples of the element's toxicity. For instance, the term 'mad as a hatter' originated from mercury poisoning observed among men who treated pelts with mercuric nitrate when making felt hats. In 1971, Iraqi farmers received large quantities of wheat seed treated with an alkylmercury fungicide. The seed was received late in the growing season and drought conditions prevailed, so it was unlikely the seeds would sprout. Instead, the farmers ground the seed into flour and baked it into unleavened bread. In turn, alkylmercury contaminants were baked into the bread hospitalizing 6530 and killing 459 (Swiderski, 2008). In a more recent example, after spilling one to several drops of dimethylmercury on her hand protected by a latex glove, Karen Wetterhahn, a Professor of Chemistry at Dartmouth College, fell into a coma and died 10 months later (OSHA, 1998).

Mercury is now recognized as a chemical of global concern by the United Nations Environment Programme, and is listed as a neurotoxicant by the United States Environmental Protection Agency (USEPA) (UNEP, 2010; USEPA, 2010b). The most common form of mercury exposure for people in the United States (US) is consumption of fish containing methylmercury. A portion of the mercury found in fish can be attributed to atmospheric deposition of mercury emitted from natural and anthropogenic sources (USEPA, 2010b). Current estimates indicate that half of atmospheric mercury can be traced to anthropogenic activities (Pacyna et al., 2006). Under the Clean Air Act (CAA), the USEPA is proactively required to address the annual emissions (i.e., estimated at 100 tons per year (tpy)) of anthropogenic atmospheric mercury released within the US. Utility coal boilers are the primary source of US atmospheric mercury, emitting half of the annual total US emissions (i.e., 50 tpy), while cement manufacturing is also a major source, at 8 tpy (USEPA, 2009a). The USEPA's 2010 final rule on mercury emissions from cement manufacturing facilities is estimated to reduce emissions sixteen-fold down to 0.5 tpy (i.e., a 94% reduction) (USEPA, 2010a). This rule is far more aggressive than the USEPA rule requiring a 70% reduction of mercury emissions from coal-fired power plants which was vacated by the United States Court of Appeals for the District of Columbia Circuit (D.C. Circuit) in 2008 (USEPA, 2010c).

Successful future compliance with USEPA's mercury rule for cement facilities will require aggressive technical innovation. Unfortunately, few researchers are actively working with the industry to address this critical issue.

This article is subsequently intended to summarize the current cement facility mercury issues, as well as highlighting data gaps and research needs by addressing:

1. cement manufacturing,
2. mercury regulations for cement facilities,
3. the use of mercury concentration in inputs to predict average mercury emissions,
4. current knowledge about cement facilities relative to mercury fate and transport,
5. cement-mercury emissions models, and
6. current and proposed cement-mercury control strategies.

2. Overview of cement manufacturing facilities

Cement manufacture has three fundamental stages: preparation of feedstocks, production of clinker, and preparation of cement. The second stage, production of clinker, is the most significant to understanding mercury fate and transport; however, the other two stages do provide contextual information.

Preparation of the feedstocks begins by quarrying raw materials that satisfy the stoichiometric requirements for cement production. The raw feed composition typically used for modern cement production consists of 85% limestone, 13% clay or shale, and less than 1% each of corrective materials such as silica, alumina, and iron ore. Following quarrying, feedstocks are crushed to a size below 20 mm and mixed in a pre-homogenization pile (Alsop et al., 2007). Production of clinker entails the majority of energy expenditure and chemical reactions required to produce cement. Long wet, long dry, preheater, and precalciner processes are practiced and respectively account for approximately 15%, 15%, 20%, and 50% of US cement production capacity (PCA, 2009a). Due to escalating fuel prices and inefficient energy use, long wet and long dry facilities are being phased out. Accordingly, this review focuses on the preheater and precalciner processes.

Fig. 1 provides the various components of a preheater and precalciner cement manufacturing facility. Cement manufacture begins when feedstocks enter the raw mill, creating a fine powder (so-called 'raw meal') in which 85% of material is smaller than 88 μm (Alsop et al., 2007). Prior to entering the kiln, the meal is transferred to homogenizing silos that minimize variation in material (Chatterjee, 2004). Following homogenization, the meal is shuttled to the preheater and precalciner tower, marking the beginning of the chemical transformation of the meal into cement. Cyclone separators (called 'preheaters') in the tower intermix the raw meal with the near 1000 °C exhaust gasses to recover energy, preheat the meal, and begin the chemical reactions producing cement. In preheater facilities, gasses flow directly from the kiln, but in precalciner facilities, fuel and air are supplied both to a combustion vessel within the tower and to the kiln. In fact, typically 60% of fuel is burned in the calciner and greater than 90% calcination is achieved prior to material entry into the rotary kiln (Alsop et al., 2007). Within the kiln, temperatures reach approximately 1400 °C completing the process chemical reactions and producing calcium silicates, called clinker, with a diameter of 10–25 mm (Steuch, 2004). To maximize energy recovery, exhaust gas from the preheater tower is often routed to the raw mill, assisting in drying the incoming feedstocks. Following flow through the raw mill, exhaust gasses are finally released into a dust collector (i.e., a baghouse or electrostatic precipitator (ESP)), which also captures fine particles created when feedstocks are milled. In many cases, this dust is then recycled into the homogenizing silo and serves as a portion of the kiln feed. Feedstocks containing high concentrations of alkali, sulfur, and/or chloride can cause buildups in the kiln and preheaters, inhibiting cement production. Due to their volatility, these materials