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# Metal concentrations and distribution in paint waste generated during bridge rehabilitation in New York State



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## HIGHLIGHTS

• Concentration and distribution of trace metals in the paint waste were addressed.

• The elevated Fe is attributed to the steel grit used as abrasive blasting material.

• Statistical analysis indicated that Pb and Cr areassociated in the paint waste.

• The observation raises concern of leaching from this waste stream.

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## ABSTRACT

Between 1950 and 1980, lead and chromium along with other metals have been used in paint coatings to protect bridges from corrosion. In New York State with 4500 bridges in 11 Regions 2385 of the bridges have been rehabilitated and subsequently repainted after 1989 when commercial use of lead based paint was prohibited. The purpose of this research was to address the concentration and distribution of trace metals in the paint waste generated during bridge rehabilitation. Using hypothesis testing and stratified sampling theory, a representative sample size of 24 bridges from across the state was selected that resulted in 117 paint waste samples. Field portable X-ray fluorescence (FP-XRF) analysis revealed metal concentrations ranged from 5 to 168,090 mg kg<sup>-1</sup> for Pb, 49,367 to 799,210 mg kg<sup>-1</sup> for Fe, and 27 to 425,510 mg kg<sup>-1</sup> for Zn. Eighty percent of the samples exhibited lead concentrations greater than 5000 mg kg<sup>-1</sup>. The elevated iron concentrations may be attributed to the application of steel grit as an abrasive blasting material routinely used by state Departments of Transportation in the paint removal process. Other metals including Ba and Cr were observed in the paint waste as well. As a result of the paint formulation, metals were found to be associated in the paint waste (Pb correlated with Cr (r = 0.85)). The elevated metal concentration impact of leaching from this waste stream.

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#### 1. Introduction

The general practice for protecting steel bridges from corrosion involves applying paint coatings (Boxall and Von Fraunhofer, 1980; Gooch, 1993; Lambourne and Strivens, 1999). Between 1950 and 1980, these paint coatings used a number of metals including lead and chromium for corrosion protection. However, concerns stemming from human health impacts of lead-based paint (LBP) prompted its ban from most applications in the United States in 1978 (Davis et al.,

\* Corresponding author. *E-mail address:* zs34@njit.edu (L. Axe). 1993; Hall, 1972; Jacobs et al., 2002). Lead release from LBP has been associated with health effects including damage to the brain and central nervous system in children, reproductive problems, and high blood pressure (Mielke and Gonzales, 2008; Mielke et al., 2001). In addition, during aging and weathering, paints tend to chalk, chip, flake, and otherwise deteriorate, resulting in an accumulation of pigment material in soils and surface water surrounding painted structures (Hopwood et al., 2003; Kyger et al., 1999). In response to these concerns, the Department of Housing and Urban Development (HUD) and Consumer Product Safety Commission (CPSC) prohibited residential use of LBP since 1978 (CPSC, 1977; National Institute for Occupational Safety and Health (NIOSH), 1992). In New York State, LBP has been prohibited from commercial use since 1989 (New York State Department of Transportation (NYSDOT), 1988). In NYS, as the paint on steel bridges reaches a critical level of deterioration, rehabilitation involves abrasive blasting, which is one of the most effective paint removal approaches (Appleman, 1992). In the blasting operation, abrasive particles are propelled against the surface using a concentrated stream of compressed air. Dust, abrasive, and paint debris are vacuumed simultaneously. Debris is separated for disposal and the abrasive particles are returned for reuse. In NYS, recycled steel grit (i.e., martensite (Society of Automotive Engineers (SAE), 2006)) (comprised by wt. of Fe (>96%), C (<1.2%), Mn (<1.3%), Si (<1.2%), Cr (<0.25%), Cu (<0.25%), and Ni (<0.2%) (Dunkerley et al., 1978)), is applied routinely (NYSDOT, 1988, 2008) as blasting material during the paint removal procedure. Although magnetic separation is used to remove abrasive material particles, a fraction of steel grit remains with paint waste. The paint waste is therefore comprised of paint and the blasting abrasive material steel grit.

Paint waste remains a pervasive problem in U.S. cities (Axe et al., 2009; Caravanos et al., 2006; Ferlauto, 1994; Mielke and Gonzales, 2008; Mielke et al., 2001; Townsend et al., 2004), and an increasing problem in the developing world where LBP is still manufactured and used (Adebamowo et al., 2007; Clark et al., 2006; Nduka et al., 2008). For example, of the 25 exterior paints studied in New Orleans (Mielke and Gonzales, 2008), only one sample revealed lead concentrations less than the HUD (2003) action level of 5000 mg kg<sup>-1</sup>. Caravanos et al. (2006) evaluated lead deposition in ambient dust in New York City boroughs from 2003–2004 and observed concentrations ranging from 138  $\mu$ g m<sup>-2</sup> to 7858  $\mu$ g m<sup>-2</sup>. Given the HUD/EPA dust standard of 431  $\mu g~m^{-2}$  (U.S. EPA/HUD, 2003), bridge rehabilitation and other construction/demolition activities are potential sources for lead and may in part explain lead deposition in this area (Caravanos et al., 2006). In addition to Pb, other metals such as Cr, Ba, and Zn in paint are potential sources of pollution during rehabilitation as well (Mielke et al., 2001; Fjelsted and Christensen, 2007; Ojeda-Benítez et al., 2013). Therefore, evaluating concentration and distribution of metals in the paint waste generated during bridge rehabilitation is necessary.

Studies of paint particles/associated waste have focused on the distribution of Pb (Bernecki et al., 1995; Beckley and Groenier, 2008; Brumis et al., 2001; Daniels et al., 2001; MnDOT, 2004) with occasional measurements of other metals such as As, Cr, Cd, Hg, and Zn (Conroy et al., 1996; Mielke and Gonzales, 2008; Turner and Sogo, 2012). Mielke and Gonzales (2008) found that Pb concentrations were independent of Hg in interior and exterior paint sampled from New Orleans homes. On the other hand, Turner and Sogo (2012) reported correlations between Pb and Cu in exterior paint of urban structures in the UK. These previous studies have demonstrated the presence of elevated Pb in subsurface coatings, however, a systematic analysis of paint waste generated throughout a region or state has not been conducted for bridges under rehabilitation. In addition, previous studies (Bernecki et al., 1995; Martel et al., 1997) have not included analyses to evaluate metal association and dominant forms present in the paint waste.

Therefore, in this study as NYS is working with 4500 bridges that will undergo rehabilitation on some regular basis, the objective of this research is to characterize the paint waste for concentration and distribution of metals. FP-XRF was applied to detect and quantify concentrations in the painted surface and subsurface. Statistical analyses were employed to evaluate the factors that affect metal distribution. Further analysis was conducted using X-ray diffraction (XRD) for mineralogy and field emission scanning electron microscopy (FE-SEM) with energy dispersive X-ray analysis (EDX) to investigate surface coating composition and morphology critical in addressing surface interactions and metal mobility. The results from this study provide fundamental knowledge on the characterization of paint waste needed for subsequent leaching studies (Shu et al., 2014, 2015). The first step in addressing metal mobility is to evaluate metal concentrations and distribution as well as the degree to which metals are present at potentially elevated concentrations.

#### 2. Materials and methods

Laboratory quality assurance and quality control procedures are based on the American Society for Testing and Materials methods (American Society for Testing and Materials (ASTM), 1990). All reagents were of certified analytical grade or trace metal quality. Containers were soaked in a 10% HNO<sub>3</sub> solution for 48 h when using glassware and 24 h for Nalgene® high-density polyethylene (HDPE) containers, then rinsed in Millipore-Q water, dried, and stored in a particle-free environment before use.

#### 2.1. Sample collection and preparation

To obtain a statistically representative number of samples for the study, hypothesis testing and stratified sampling theory was applied for the sample size estimation. With 11 Regions and 2385 bridges rehabilitated and subsequently repainted after 1989, 24 bridges from across the NYS were selected for this study based on the statistical analysis (details are provided in Supporting Information). Between October 2010 and November 2011, 117 samples of paint waste were obtained from 24 bridges under rehabilitation from seven regions (Regions 1, 2, 3, 5, 7, 10, and 11) in NYS (details are provided in Supporting Information Fig. S1). All bridges in the study have been repainted at least once since 1989, when NYS prohibited the commercial use of LBP (NYSDOT, 1988). Duplicate (paint waste) samples were collected from five random locations (directly from bridge surface or collected waste in 50 gal drums) at each bridge site selected. Specifically, two samples were obtained from the same drum location or bridge site with trowels. In total, ten samples were collected for each bridge (four samples for Bridge 3-3) and stored in Nalgene® high-density polyethylene (HDPE) containers at 4 °C (American Society for Testing and Materials (ASTM), 1990).

#### 2.2. Total metal concentrations

Field portable X-ray fluorescence (FP-XRF) is one of the most effective approach to measure Pb and other metal concentrations in paint (Beckley and Groenier, 2008; Brumis et al., 2001; Daniels et al., 2001; Minnesota Department of Transportation (MnDOT), 2004). FP-XRF has proven effective for in situ analysis as demonstrated in field and laboratory results (i.e.,  $R^2 = 0.976-0.992$  for Pb (Markey et al., 2008) and  $R^2 = 0.843-0.996$  for As, Pb, and Zn (Radu and Diamond, 2009)). An advantage of the XRF technique is its ability to probe metal concentrations on the surface as well as subsurface; thus, it has the capability to characterize a wide range of remaining pigments.

To investigate the metal distribution in the paint waste, Ba, Cr, Pb, Fe, and Zn were analyzed with the NITON XL3t-600 series FP-XRF following EPA Method 6200 (U. S. EPA, 1998) using either Soil Mode (metal concentrations < 2% by wt.) or Mining Mode (metal concentrations  $\geq$  2% by wt.). The calibration was verified by analyzing NIST certified reference material (SRM). Additional details including detection limits for the FP-XRF in these two modes are provided in Supporting Information (Tables S1 and S2). Each sample was homogenized by using the four quarter method (Popek, 2003) to represent the sampling interval. The paint samples were loaded into 12 ml sample holders (SC-4331), sealed with transparent membranes, and analyzed for 180 s. The XL3t-600 FP-XRF frame is used to support the analyzer during the detection procedure for continuous analysis. The instrument combines advanced electronics and provide a continuum of X-rays across a broad range of energies with a maximum output of 50 keV. Filters were applied between the X-ray tube and the sample to suppress the continuum radiation while passing the characteristic X-rays from the anode. It is important to note that both Soil Mode and Mining Mode combine fundamental parameters (FP) mode with Compton normalization (for background matrix automatic correction), which provides improved accuracy for samples ranging from less than 2% by wt.

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