

Discussion: Effect of Changing Water Quality on Galvanic Coupling

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A famous colloquium (Langmuir, 1953) explains how well-intentioned scientists have been tricked into false results by wishful thinking and other factors. That analysis may provide a partial explanation for substantive errors in the March 2012 peer-reviewed Journal article (Effect of Changing Water Quality on Galvanic Coupling) written by Boyd et al. The authors state they have gathered data demonstrating that elevated lead in water arising from galvanic corrosion between direct connections of lead and copper pipe will be “transient” and “short-lived.” Moreover, they assert that bringing lead and copper into direct contact (as sometimes occurs in the field) poses a much smaller risk of galvanic corrosion and lead contamination of water than laboratory simulations in which the metallic pipes are slightly separated with a dielectric spacer and connected externally with a wire (likely to become a more common practice in the field). Their claims have immediate implications for water utility approaches to partial lead service line replacements, which have been linked to a higher incidence of childhood lead poisoning and expenditures exceeding \$100 million at one utility (Brown et al, 2011; Frumkin, 2010; Leonnig, 2008). My analysis

of this article has revealed serious problems with some of the data, analysis, text, and figures.

CLAIM CONTRADICTED BY ELECTROCHEMICAL THEORY AND PRACTICE

According to the authors, when lead and copper pipe are brought into direct contact, “accelerated metal release . . . may be minimal” because of galvanic corrosion. In contrast, if the lead pipe (anode) and copper pipe (cathode) are separated by 1–15 cm and electrical contact is maintained with an external wire, the potential of “the entire lead coupon shifts in an anodic direction,” and “the galvanic coupling has likely accelerated lead release by up to tenfold.” These statements are supported by two figures (Figures 9 and 10) in the March Journal article. This claim is contrary to the well-established “distance effect” as summarized by Bradford (2001):

This ‘distance effect’ offers another way to combat galvanic corrosion: space anode and cathode far enough apart and galvanic corrosion will virtually cease even though the metals are still electrically connected by an external conductor To prevent galvanic corrosion, the plumbers often put insulated connectors between the two kinds of piping. Building codes, however, require

the plumbing to be electrically continuous for grounding purposes so electricians fasten external metal straps across the insulated couplings The insulated spacer between the two pipes separates them enough so that the water’s resistance prevents the exchange of much current.

At no point do Boyd et al acknowledge that their new theory is contradicted by decades of prior peer-reviewed research and practical experience, and a recent paper has verified that the claims in their March Journal AWWA article are incorrect (St. Clair et al, 2012).

KEY FIGURES ARE FLAWED

The results from the Journal AWWA article described in the preceding section were presented at two national AWWA conferences, a graduate engineering ethics seminar, and a US Environmental Protection Agency (USEPA) special Science Advisory Board (SAB) meeting (Reiber, 2011a–c; Boyd et al, 2010a). Indeed, the final USEPA SAB report cited the preliminary presentations of the March Journal article seven times (USEPA, 2011). In their presentations, the authors highlighted their new theories on “so-called galvanic corrosion” of direct connections between lead and copper and used two figures (the same as Figures 9 and 10 that were published in the

March article) to assert that results of other researchers were experimental artifacts that would not occur in practice. As peer-reviewed research of my graduate students was among the studies called into question by these claims (e.g., Triantafyllidou et al, 2011), our group invested more than two person-years of effort trying (without success) to reproduce the data presented in Figures 9 and 10. We eventually came to the conclusion that the results featured in these figures and associated text were not scientifically valid. When the authors did not immediately provide data supporting these figures in response to my requests, I obtained the original PowerPoint® slides used by the authors in their USEPA SAB presentation through a Freedom of Information Act (FOIA) request (USEPA, 2012). I observed that the lines in the graphs floated completely independent of the graphical axis. When magnified, the lines did not have the appearance of scientific data, but looked like lines drawn electronically with Microsoft Draw® or a similar program, as evidenced

by curled ends and other aberrations (Figure 1). Journal editors confirmed that these same graphs were submitted as Figures 9 and 10 in the March Journal article. Agreeing to a proposal made by the authors that they would answer my questions and provide data if the chair of the Journal's Peer Review Editorial Board (PREB) served as intermediary, I again requested the original data behind these figures and a detailed description of the mathematical methods used by the authors to generate the lines.

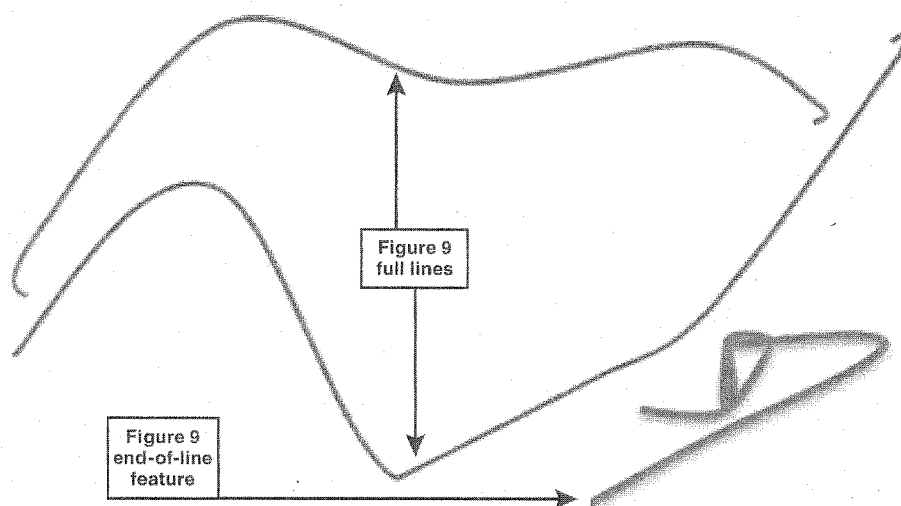
ERRONEOUS DATA IN FIGURES 9 AND 10 ASCRIBED TO GRAPHIC ARTIST

In written responses that were “discussed and agreed upon by all the co-authors of [the Journal] paper,” the authors explained that “[they] couldn’t get the clear and colorful presentation [they] wanted using Excel® graphics routines and asked [an on-staff graphic artist] to prepare the slides from the Excel data.” The graphic artist “was given the instructions to make the images colorful and

large.” According to the authors’ written statements, the artist also:

- made quantitative errors of 156–200% in labeling every x-axis for lead surfaces in Figures 9 and 10;
- created 5 cm of new electrochemical data not collected in experiments and added them onto data for the lead line in Figure 10 of the March Journal article (Figure 2);
- developed erroneous single composite lines from multiple datasets in the Excel spreadsheet given to him or her. For example, the authors stated that the graphic artist somehow combined two sets of data to generate one erroneous composite line in Figure 10 of the Journal article [Figure 2], which had obvious errors exceeding 75 mV compared with the cited spreadsheet data, even after correcting for the flawed x-axis;
- created composite lines for Figure 9 in the Journal article by combining Excel data from four datasets (my analysis shows that the graphic artist composite line is erroneous by more than 50 mV from a simple point-by-point averaging of the four datasets [results not shown but

FIGURE 1 Vertical and horizontal expansion shows “data” from two complete lines appearing in Figure 9 of the Boyd et al March 2012 Journal article and an end-of-line feature from a third line



Source: USEPA, 2012

available upon request; error similar to that illustrated in Figure 2]); and

- created another erroneous composite line in Figure 9 in the Journal article after he or she decided to exclude one of the Excel datasets as an “outlier.”

According to this version of events, none of the authors created Figures 9 or 10, which were used in the Journal article and in their numerous presentations. All errors and extra data added onto lines in the figures were attributed to actions of the unnamed graphic artist, and to the authors’ “lack of oversight” of the unidentified individual’s work. When asked to provide documents

corroborating their claim by the PREB intermediary, the authors did not do so.

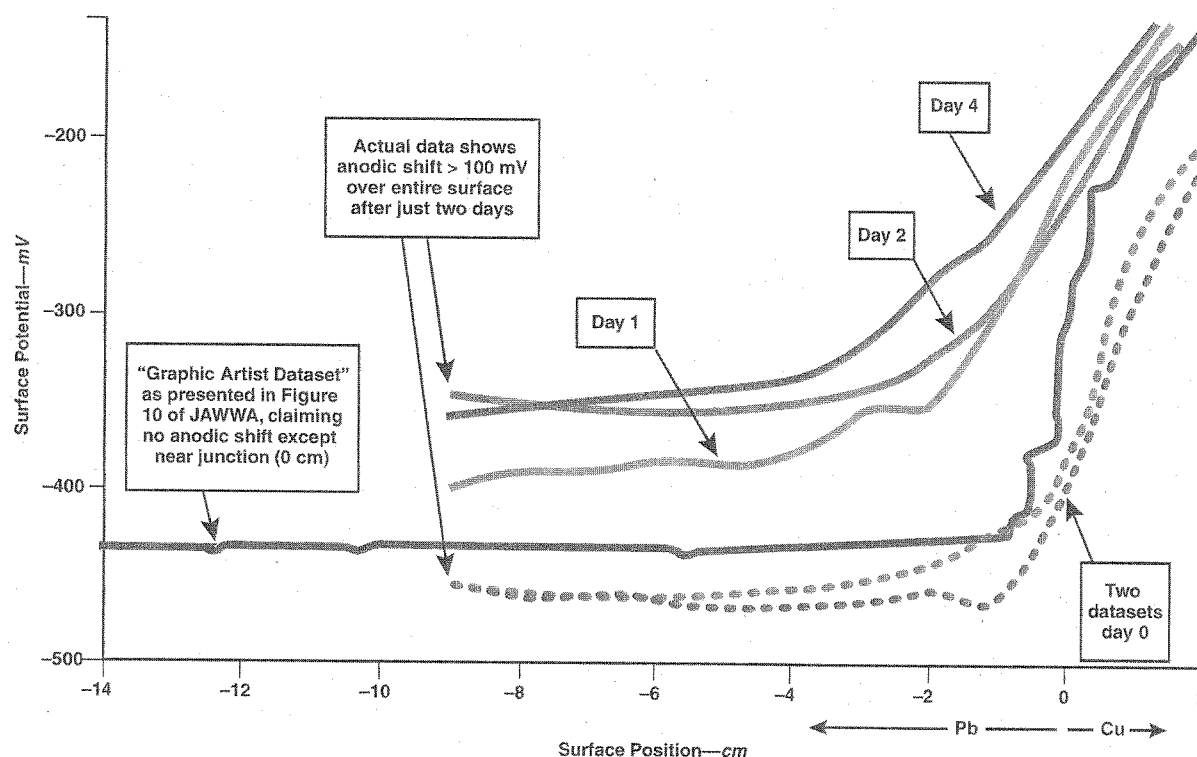
DATA FROM AUTHORS CONTRADICT THOSE IN THE JOURNAL ARTICLE

The data the authors provided to me in spreadsheets do not agree with those presented in the Journal article and actually support conclusions contrary to those stated in the article (Table 1). The magnitude and importance of the discrepancy are illustrated by the following two representative examples.

Example 1. The Journal article states that for a “typical” result,

when lead and copper surfaces were separated by distance and connected by wire externally, the lead surface potential was shifted more than 100 mV, which “likely accelerated lead release by up to tenfold.” Figure 9 and other text in the article describe the anodic shift as “approximately 150 mV” or “about 150 mV.” The authors also assert in the article that this large shift was “stable for periods extending to weeks and likely months.” Expectations for the spreadsheet data based on these assertions are summarized in Figure 3. But according to the actual spreadsheet data identified by the authors as the basis for Figure 9 of

FIGURE 2 Comparison of the line created in Figure 10 of the Boyd et al March 2012 Journal article with data provided in the spreadsheet



Aside from the nonexistent data at a position of 9–14 cm (i.e., –9 to –14 cm above), the composite line also features large errors (> 75 mV at points) compared with day 0 data that the authors stated was the source of the line. More important, the spreadsheet reveals that after day 0, the potential of the entire lead surface shifted anodically, contradicting the authors’ discussion in the Journal article claiming that there was no shift on the midpoint of the lead surface if it was directly connected to copper. Data trends collected after day 0 were not presented or discussed in the Journal article.

Cu—copper, Pb—lead

the Journal article, the lead anodic shift (using their approach of comparing jumpered and unjumpered data) started out at only 58 mV on day 0 and dropped to 6 mV in five days, at which point the experiment was terminated. Thus, the discussion in the Journal article is in error by a factor of greater than 16 times after just five days. According to the data files the authors provided to the PREB and me, the experiments were not run for even a single week.

Example 2. The authors state in the Journal article that when lead and copper were directly connected

and “when the surface potential was measured at midpoint of the copper surface as well as midpoint of the lead surface, . . . the surface potential of each surface appear[ed] unconnected.” The authors further state that “. . . the mapping of the surface potential across the entire bimetallic pipe coupon indicated that the galvanic effect was limited to the immediate vicinity (~ 5 mm) of the lead-copper interface.” When all data from the experiment identified by the authors as the basis for Figure 10 are graphed, a markedly different result is apparent. By day

1 the entire 9-cm lead surface had risen anodically by more than 50 mV versus day 0, and by day 2 the entire 9-cm lead surface had shifted anodically by more than 100 mV versus day 0 (Figure 2). The anodic shift 10 mm from the junction was more than 200 mV after just four days. Such short-term acceleration to galvanic corrosion has been noted and is explained elsewhere (Hu et al, 2012; Francis, 2010; Nguyen et al, 2010).

Overall, the spreadsheet data provided by the authors suggest much greater galvanic acceleration for the

TABLE 1 Illustrative comparisons of Journal article text with galvanic narrative and spreadsheet data

| Journal AWWA text | Galvanic Narrative | Actual Spreadsheet Data |
|---|---|---|
| “In the absence of external electric connection, the coupons exhibited a difference of OCP of approximately 400 mV; in contrast, when the coupons were connected externally, that difference was only 120 mV, most of which was attributed to an anodic shift (polarization) of about 150 mV on the lead surface.” | “Whereas un-jumpered, the coupons gave a potential difference of approximately 400 mV; jumpered, the difference is now only 120 mV, most of which is due to an anodic shift (polarization) of about 150 mV on the lead surface.” | The minimum difference when connected externally was always > 280 mV on day 0. As noted below, the anodic shift was never close to 150 mV. |
| “In this indirect (externally wired) configuration, the shift of the OCPs can be stable for periods extending to weeks and likely months.” | “... in this configuration, it is stable for periods extending to weeks and likely months” | Trial 1 experiment terminated after four days after OCP shift dropped to an average of 5 mV. |
| “The galvanic shift induced by this mode of galvanic coupling can significantly affect lead surface corrosion because an anodic shift of the OCP of more than 100 mV is equivalent (based on relevant Tafel data) to a corrosion current increase approaching an order of magnitude. In other words, when copper and lead are coupled using the indirect mode, the galvanic coupling has likely accelerated lead release by up to tenfold.” | “The effect of the galvanic shift (polarization) on the lead surface corrosion is huge, the anodic shift of more than 100 mV suggests (based on Tafel) a corrosion increase approaching an order of magnitude. . . . In other words, in this jumpered configuration, the galvanic coupling has likely accelerated Pb release by up to tenfold.” | Average anodic shift on day 0 was only 58 mV, on day 1 it was 26 mV, and on day 5 it was 5 mV. For experiment 2, anodic shift was only 29 to -5 mV between zero and five days. |
| “Figure 10 shows that when the surface potential was measured at the midpoint of the copper surface as well as midpoint of the lead surface, the observations were strikingly different from those for the indirectly jumpered coupons (Figure 9). In this abutted (end-to-end) configuration, the potential of each surface appears unconnected except in the area directly adjacent to the physical juncture.” | “... if we measure the surface potential midpoint of the copper surface, as well as midpoint of the lead surface, the observations are strikingly different than the jumpered coupons measured previously. . . . In this configuration, the surface potential of each surface appears unconnected . . . both surfaces (midpoint) retain the electrical potential when they were unconnected.” | The midpoint potential difference for the lead surface between the two configurations was only 20–30 mV on day 0 of both trials. But this is expected for the direct connection, given that the actual midpoint of this lead surface was 29% farther and the lead surface was 29% larger. |
| “In fact, the mapping of the surface potential across the entire bimetallic pipe coupon indicated that the galvanic effect was limited to the immediate vicinity (~ 5 mm) of the lead-copper interface, whereas on the copper surface the effect was limited to a few centimetres of the interface.” | “If we map the surface potential across the entire bi-metallic pipe coupon, we find there is a galvanic effect, but on the lead surface the effect is limited to the immediate vicinity . . . (> 5 mm) of the lead-copper interface; whereas, on the copper, the effect is limited to within a few centimeters of the interface.” | Within four days, the potential of the entire lead surface (9 cm) has risen upwards by 100–200 mV (Figure 2). Experiment was terminated. |

OCP—open circuit potential, Pb—lead

direct connection than for the indirect connection and confirm prior research and theory. The discrepancy is exacerbated by the fact that there was actually a 28% larger surface area in the case of the direct versus the indirect connection, as opposed to the results shown in Figures 9 and 10 and in the text of the Journal article, which falsely made it appear as if the surface areas tested were of equal size.

JAWWA TEXT WRITTEN BEFORE EXPERIMENTS WERE CONDUCTED

A chronology of the authors' e-mails (available on request) reveals that key erroneous statements in the March Journal article text were written before the experiments identified as the basis for figures in the article were conducted or analyzed. For example, the data identified by the authors as the basis for Figure 10 were not collected until Oct. 4, 2010 (according to dates on the data

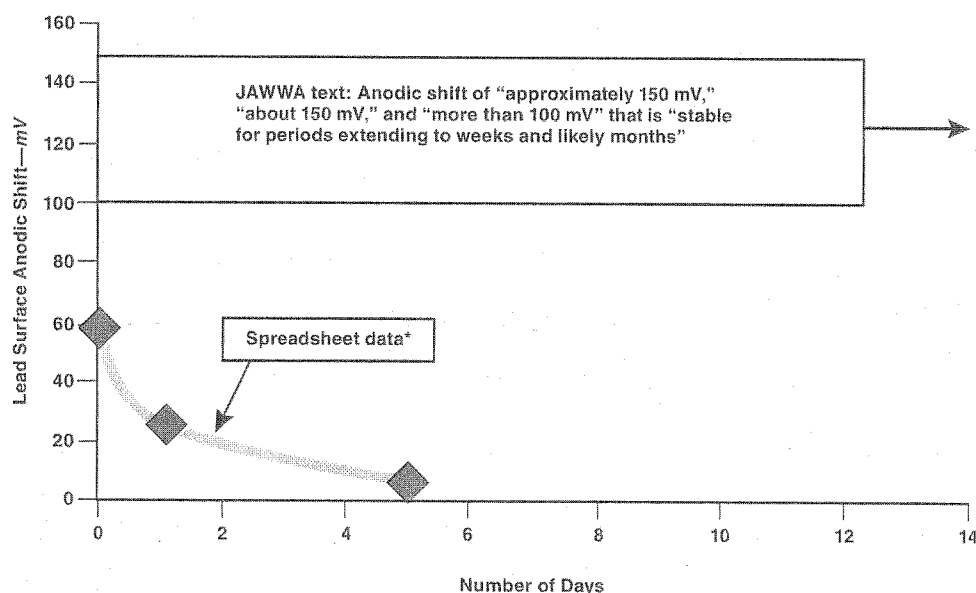
spreadsheet released by the authors and in supporting photos). Yet one week earlier, on Sept. 26, 2010, the first author offered to write a results and discussion section after reviewing self-described "preliminary data" of dubious value and viewing some pictures of the apparatus. He stated his write-up for the paper to be submitted to Journal AWWA could be "based on what I see in the photos and on our previous discussion." On Sept. 30, 2010, in an e-mail with the subject line "Galvanic Narrative," the second author wrote text that became the Journal AWWA article and openly acknowledged that a technician "will provide photos and data as we proceed, assuming [he or she] agrees with the narrative." The authors and technician did not meet to "discuss the data and decide how to present the results" from the experiment until Oct. 13, 2010, more than two weeks after the "Galvanic Narrative" and

Journal AWWA article text were written. As is demonstrated in a point-by-point comparison (Table 1), the authors never substantially updated or altered their "Galvanic Narrative" text for the Journal article based on the actual experimental data. This disconcerting chronology explains the origin of inaccurate discussion in the March 2012 Journal AWWA article. In letters sent to the PREB and me, the authors wrote that the dataset identified as the basis for Figure 10 was dated Aug. 4, 2010. That statement is contradicted by dates in the e-mails, the data spreadsheet itself, and supporting photos.

LARGE ERRORS IN MEASURING TOTAL LEAD NOT DISCLOSED

Serious problems with the work presented in the March Journal article are not limited to the section discussing Figures 9 and 10. In the final report of the project on which Jour-

FIGURE 3 Comparison between the experimental results the authors identified as the source of Figure 9 of the March 2012 Boyd et al Journal article to expectations based on information presented in the article text



*Spreadsheet data were provided by the authors. Results of the actual spreadsheet data were obtained by determining a point-by-point difference between paired jumpered and unjumpered results and taking an overall average for each day. (One out of 192 data points was excluded because it was missing a minus sign.)

nal article Figures 1–6 were based (Boyd et al, 2010b), the authors acknowledge that, “Our setup and operations were not designed to conduct mass balance calculations of all lead and copper, specifically including particulates that accumulated in dead zones” Recent research has consistently demonstrated that this deficiency is problematic relative to tracking accelerated lead release from galvanic corrosion because almost all the extra lead released to the water tends to be particulate (Cartier et al, 2012, 2011; Giammar et al, 2012, 2011; Triantafyllidou, 2011). Giammar and others have demonstrated that unless mass balances such as reservoir acidification are used to recover all of the settled particulate lead, the data can be misleading and can generate false conclusions, even during constant recirculation (Giammar et al, 2012, 2011). Galvanic accelerations to lead release as large as 300% would be completely missed without acidification (Giammar et al, 2011). A third-party review was commissioned by DC Water and the Water Research Foundation, in which the investigators were charged with examining the data that ultimately appeared in Figures 1–6 of the March Journal article (Giammar et al, 2012). The investigators concluded that the work described in the Journal article did not use methods that detected “. . . all of the lead released from the pipe, so these measurements represent lower bounds on the total lead released.” The review also determined that the “. . . underestimation may be mild (a factor of two) or possibly quite significant (a factor of 10 or more).”

It is inappropriate for the authors of the March Journal article to omit knowledge of the large potential errors in their lead measurements. At the very end of the data collection phase described in the article, I contracted with their consulting firm to use the same rigs, pipe samples, waters, and personnel on a followup project. I directed my contacts at the

firm to conduct the first quality assurance/quality control test to quantify the extent of the potential underestimation of lead release. After viewing the results that proved very large error rates—the magnitude of which varied from rig to rig and from experiment to experiment—my contact at the company wrote “. . . youch. We need to get the word out about these reservoirs” (Sandvig, 2008a). Rather than frankly disclosing the large possible errors and their implications, the authors simply state “. . . a small fraction of particulate lead might not have been accounted for because of its potential settling . . .” and further imply the errors would not affect their conclusions.

OTHER DATA SUGGESTING A LARGE ACCELERATION TO LEAD RELEASE WERE IGNORED

At the start of my work with the authors’ rig, I asked for and received a written update on the rig’s status (Sandvig, 2008b). It was stated that all pipe samples had been removed from the rig, stored wet elsewhere, and that the reservoir was full of water and otherwise unaltered from the last experiment described in the March Journal article. I then coordinated with personnel to collect the first (and only) measurement of total lead in the reservoir in accordance with the experiments described in the Journal AWWA article, using the acidification techniques later proven by others to detect accelerated particulate lead release caused by galvanic corrosion (Giammar et al, 2012, 2011). Blind samples were mailed to Virginia Polytechnic Institute and State University. When they were decoded by the consulting firm, the total lead detected in the two reservoirs with lead pipe was 2,639 and 3,243 µg/L. The total lead in the galvanically connected rigs was 9,182 and 9,189 µg/L, showing excellent reproducibility between duplicates. These results are cited in Boyd et al (2010b) and bring the authors’ results into agreement with theory and the

findings of other researchers. That is, they indicate a large contribution to total lead release (> 300%) from galvanic corrosion between directly connected lead and copper pipes.

CITATIONS IN ARTICLE SUPPORTING AUTHORS’ CONCLUSIONS ARE BASED ON DATA THAT ARE ALSO UNAVAILABLE

A report written by the second author (Reiber and Dufresne, 2006) and funded by USEPA Region III is cited in the March Journal article as an example of prior research demonstrating that “lead release effects” due to galvanic coupling are “minimal for aged and passivated surfaces of lead service lines coupled with new copper surfaces.” I submitted an FOIA request to USEPA Region III requesting this lead-in-water data six years ago. These data were never provided to me. Other graphs appearing in this 2006 USEPA report also appear unscientific, as per the prior discussion of Figures 9 and 10 of the March 2012 Journal article. I then requested via the FOIA any data, spreadsheets, or other information that could support the graphs in the 2006 USEPA report and also made another request for the 2006 “lead release effects” data cited in the March 2012 Journal article. On June 1, 2012, USEPA Region III informed me that no one has any data, spreadsheets, or other information to support the authors’ statements in the March 2012 Journal article or to otherwise support graphs appearing in the 2006 USEPA report.

SUMMARY

Many statements and figures in the March 2012 Journal article are without scientific basis, are contrary to established theory and practice, and have been refuted by other investigators who report there are sometimes significant problems with elevated lead from galvanic effects in direct lead–copper connections during partial lead service line replacements. The latter results are based

on situations using new lead pipe (Cartier et al, 2012; Hu et al, 2012; Clark et al, 2011; Triantafyllidou et al, 2011), aged/passivated lead pipe (Cartier et al, 2011; Giammar et al, 2011), examination of field samples (DeSantis et al, 2009), and in experiments using “real” brass connections between lead and copper pipe (Cartier et al, 2011; Clark et al, 2011; DeSantis et al, 2009). Galvanic effects are also sometimes very persistent and depend on a wide range of factors (Cartier et al, 2012; Giammar et al, 2012; Hu et al, 2012; Clark et al, 2011; Giammar et al, 2011; Triantafyllidou et al, 2011; Nguyen et al, 2010; DeSantis et al, 2009). As unfortunate as it would be, I believe that the serious and extensive errors documented in the March 2012 Journal article by Boyd et al justify its retraction from the peer-reviewed literature.

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