Aluminium residuals in emergency water treatment: What you wanted to know, but never asked

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Alum is widely used in water treatment because of its many advantages, of which its relative low cost and worldwide availability have ensured it to be the coagulant of choice for emergency water treatment. However, in certain circumstances, its use may leave an undesirable coagulant aluminium residual in the finished water. Such residuals can generate (unwarranted) fear over the use of this useful coagulant (particularly among the misinformed). This article is focused on clarifying topics related to aluminium residuals, such as its occurrence, health risks, significance in emergency water treatment, measurement, and control strategies. These issues are discussed in view of the practical constraints faced in the field during emergencies and where appropriate contrasted with conventional (non-emergency) practice. Emergency water treatment sludge disposal and alternative coagulants to alum are also addressed.

Keywords: alum, coagulation, emergency water treatment, Alzheimer's disease

'ALUM' OR ALUMINIUM SULFATE is the most commonly used coagulant for water treatment because of its wide availability at relatively low cost in almost all parts of the world. In many 'conventional' (i.e. non-emergency situations) water treatment plants alum is used primarily for turbidity reductions and the removal of natural organic matter (NOM). Turbidity is an important aesthetic parameter directly associated with the amount of particulates in the water. It should be reduced to improve the appearance of the finished water and to reduce the effects of the waterborne particulates on downstream processes of filtration and disinfection. The removal of NOM is also to facilitate chlorination by reducing the chlorine demand and because its reaction with chlorine can form unwanted disinfection by-products (DBPs).

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Alum is used in emergency water treatment for the reduction of turbidity

In emergency water treatment alum is also used for the reduction of turbidity. While some removal of other contaminants and substances (e.g. arsenic, pesticides, microorganisms) may also occur, their removal efficiency will only be as good as the measurement of those substances. That is, unless the contaminant/parameter in question can be quantified, its removal efficiency with alum cannot be determined. Typically, the only basic analytical field capacity available in emergencies is test kits such as the 'DelAgua Kit'. Such kits are capable of making determinations of four critical drinking water quality parameters: thermotolerant (faecal) coliforms, turbidity, pH and free/total residual chlorine. Therefore, any additional contaminant removal (other than turbidity and thermotolerant coliforms) that may occur through alum coagulation comes as a secondary benefit. Yet, it should be noted that thermotolerant coliforms are usually used to assess the distributed water quality. Unlike turbidity, which can be quickly measured, thermotolerant coliforms have a relatively long sample processing time (nearly 24 hours after sample collection). It is therefore not practical to use it as a routine operational control parameter for alum coagulation, as any changes to the process need to be quickly measured and with thermotolerant coliforms these could only be assessed the following day. An overview of coagulant-based water treatment practices in emergencies is presented elsewhere (Dorea, 2009).

Some studies have postulated the association of chronic exposure to aluminium residuals in drinking water and neurodegenerative diseases

Alum coagulation can lead to the presence of residual amounts of this metal coagulant in the finished water; these are known as 'aluminium residuals'. Some studies have postulated the association of chronic (i.e. long-term) exposure to aluminium residuals in drinking water and neurodegenerative diseases such as Alzheimer's disease. This has generated (many times an unwarranted) fear of these residuals and alum by association, particularly among uninformed water professionals and the general public they serve. Curiously, concerns over the health effects of using alum in water treatment are not as recent as one may think. According to Baker (1948), one of the reasons that the use of alum in public water supplies did not come sooner was due to the 'influence of ill-informed or prejudiced persons whose word was respected' in the late 19th and early 20th centuries.

The relatively shorter time spans, and the conditions and objectives in emergency relief operations are very different from those in conventional water supplies, requiring an alternative approach in interpreting the information available on aluminium in drinking water. Humanitarian practitioners should therefore be well informed on issues regarding aluminium residuals so as to not compromise benefits alum coagulation has to offer. This article seeks to answer any questions that may be linked to this coagulant by examining issues associated with aluminium residuals (i.e. aluminium occurrence, chemistry, health effects, measurement and control strategies) and discussing them in view of alum coagulation practice and constraints in emergency water treatment.

How do aluminium residuals occur?

Aluminium is one of the most common elements in nature; it is the most abundant metal and the third most plentiful element in the Earth's crust. In drinking water, it can come from the natural content of raw source waters, where it occurs in its dissolved form or as a part of sediments and suspended solids, such as turbidity-causing aluminosilicate clay particulates. However, it is most commonly associated with residuals from alum-based coagulation treatment processes. Problems with high (say > 0.200 mg/l) aluminium residuals can often be linked to systems that are not being run properly (i.e. under- or over-dosing of alum). Usually, well-operated systems should not have a problem with high aluminium residual levels (WHO, 2006). Two other factors that can significantly affect aluminium residual concentrations are the water temperature and pH (Figure 1). Low temperatures (say < 5°C) are known to hinder alum coagulation performance by reducing turbidity reduction efficiencies and causing high aluminium residuals (Morris and Knocke, 1984; van Benschoten et al., 1994). Extremes in pH (i.e. < 5.5 and > 8.0) cause the solubility of aluminium to increase (see following section).

Well-operated systems should not have a problem with high aluminium residual levels

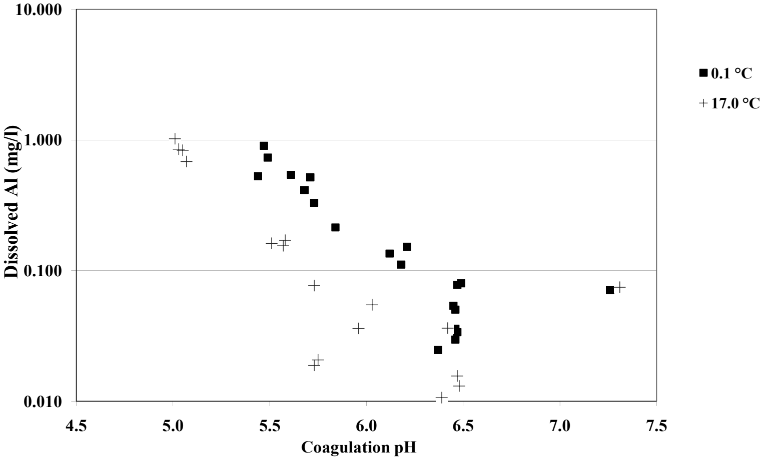

Figure 1. Effect of coagulation pH and temperature on dissolved aluminium (Bérubé and Dorea, 2007)

Aluminium chemistry in a nutshell

> The solubility of aluminium is influenced by the water pH and temperature

In drinking water aluminium can occur in two basic forms: dissolved (soluble) or particulate. The solubility (i.e. how much can be dissolved) of aluminium depends mainly on the water pH (Figure 1) and can be influenced by other factors such as temperature and the chemical composition of the solution. As depicted in Figure 1, the minimum solubility of aluminium will usually lie near the neutral pH range of 6.0 to 7.0 (Bérubé and Dorea, 2007). This will be important to consider during the control of aluminium residuals, as good turbidity reductions with alum can also be achieved within the same pH range (Twort et al., 2000) and the mitigation strategy will depend on the form in which the residual is present (see section 'How can I control it?'). It should be noted that the dosing of alum may change the solution pH, thereby affecting the solubility of the metal. That is, when alum is added to water it undergoes hydrolysis, a reaction in which hydrogen ions (H^+) are released. This increase in hydrogen ion concentration can cause the pH to drop, potentially increasing the aluminium residual (if sufficient alkalinity buffering is not available).

What are the health risks?

One of the reasons conventional water supplies that use aluminium-based coagulants try to keep low coagulant residuals is as a precautionary approach to the possible health effects of drinking-water aluminium exposure. It has been proposed that there is a link between aluminium in drinking water and the onset of chronic neurodegenerative diseases such as Alzheimer's. Yet, by comparing the results from the numerous reviews on the subject it becomes apparent that so far the epidemiological evidence has yielded inconclusive and/or contradictory results between different studies (e.g. Reiber et al., 1995; Flaten, 2001; Krewski et al., 2007 – and references therein). So, to date no association between aluminium residuals in drinking water and Alzheimer's disease has been firmly established. However, owing to the uncertainties in those studies this relationship cannot be totally dismissed and further research is warranted (WHO, 1997; Krewski et al., 2007). The current World Health Organization guideline value for aluminium in drinking water of less than 0.200 mg/l is not health-based (WHO, 2006). This is solely based on aesthetic considerations, as elevated aluminium levels may cause high turbidity or discoloration in finished waters.

> To date no association between aluminium residuals in drinking water and Alzheimer's has been firmly established

The precautionary approach towards residual aluminium in drinking water is because it is a known neurotoxic metal (Reiber et al., 1995) and doesn't have any known biological function; that is, it is a non-essential element. However, drinking water Al toxicity depends

Aluminium must first be absorbed by the body before an effect can be observed

firstly on its (oral) bioavailability. Bioavailability of a substance has been briefly defined by Exley and Birchall (1992) as a measure of its potential to interact within a biological system to produce a concomitant response. In other words, it (i.e. aluminium) must first be absorbed by the human body before an effect (e.g. Alzheimer's disease) can be observed. The simple presence of a substance (e.g. aluminium) does not automatically infer its bioavailability (Exley and Birchall, 1992). If such were the case, according to the data from studies recently cited by Yokel and Florence (2008), a simple cup of tea could be considered a potentially dangerous drink as it can contain up to 80 times more aluminium than drinking water!

The bioavailability of aluminium in drinking water is low (Yokel et al., 2001). This can be partially explained by the chemical changes that the ingested aluminium goes through in the gastrointestinal tract. The simplified solubility model (Reiber et al., 1995) indicates that most (99.9%) of the ingested aluminium from drinking water and other sources, will be excreted in the stool. According to this model, the acidic conditions (pH 1.5 to 2.0) of the stomach should solubilize all the aluminium. On leaving the stomach the pH is raised through the duodenum to about 6.2 and by the time it reaches the small intestine the pH is further increased to 7.3; remembering that in the near-neutral range the solubility of aluminium is at a minimum (Figure 1). As a result, most of the previously solubilized metal in the stomach is converted into aluminium hydroxide precipitates (i.e. solid particulates) that are not absorbed by the intestine walls.

One of the few groups at risk though are those suffering from an impaired or no renal function (i.e. renal dialysis patients), as they are unable to excrete the little aluminium that is absorbed or introduced as a dialysis contaminant and can suffer from aluminium-induced encephalopathy, leading to dementia. It is also worth noting that the major sources of ingested aluminium come from food and medications (50 to 100 times more than from drinking water), but most of the orally ingested aluminium will be excreted as explained above. In view of this, perhaps more concern should be focused on the use of aluminium in vaccines (e.g. Clements and Griffiths, 2002), as its parenteral administration bypasses the body's natural gastrointestinal barrier.

In emergency water treatment, it is recommended to adhere to the applicable local drinking water quality standards. Where these are not available, the WHO *Guidelines for Drinking Water Quality* (WHO, 2006) should be followed. Considering the resource and environmental constraints faced in emergencies, The Sphere Project (2004) recommends key minimum quality indicators in water supply in which aluminium residuals from coagulants are referred to in the last of the key indicators: 'No negative health effect is detected due to short-term use of water contaminated by chemical (including carry-over of treatment chemicals) … and assessment shows no significant probability of such an effect…'

The aforementioned considerations on aluminium and health take in to account chronic (i.e. long-term) exposure to aluminium. There is no mention in the literature of any 'significant probability' of health effects due to short-term exposure to aluminium in drinking water in normal conditions (i.e. excluding extreme alum overdosing accidents). As such, and owing to their temporary nature, emergency responses err on the side of caution and are aligned with the precautionary approach with respect to any possible health risks. Furthermore, emergency water supply has the main objective of ensuring the affected population's survival and prevention of acute waterborne and water-washed diseases which can be the main cause of death in the aftermath of a disaster (Waring and Brown, 2005). The provision of adequate amounts of safe water in emergencies should never be jeopardized by a pragmatic fear of possible health effects due to aluminium residuals in drinking water; particularly with the excellent track record alum has in emergency water supply applications.

The provision of safe water in emergencies should never be jeopardized by a fear of possible health effects due to aluminium residuals

What is its significance in emergency drinking water treatment?

Despite the existing debate on the health-related implications of aluminium in drinking water, high levels are still considered unwanted and should be minimized whenever possible. This is mainly due to the other problems that can occur as a consequence of elevated concentrations of aluminium in treated waters. The main point of concern with high aluminium residuals is the reduction in disinfection efficiency that it may cause. That is, pathogenic microorganisms can become attached to aluminium hydroxide precipitates (i.e. flocs) which can hinder the effectiveness of the disinfection process (Hoff, 1977). The same aluminium hydroxide precipitates can also cause an increased turbidity in the treated water effluent (Costello, 1984) if they cannot be removed during separation processes (i.e. clarification, filtration). This may affect the aesthetic quality of the water, which in some cases can lead to the preference of other, more pleasant, but potentially microbiologically unsafe water sources by the consumers. It has also been shown that particulate aluminium residuals from alum coagulation (i.e. floc) can also affect the performance of downstream treatment processes such as filtration by causing excessive headloss despite low (i.e. < 5 nephelometric turbidity units) recorded turbidities (Dorea and Clarke, 2006). That study was conducted on slow sand filters, but particulate aluminium residuals could also potentially cause a significant shortening of filtration runs in emergency water treatment systems that utilize alum coagulation as a pre-treatment for pressure or membrane filtration systems (e.g. Gabelich et al., 2002). Other (non-health-related) problems associated with elevated concentrations of Al in treated waters have been listed by Driscoll and Letterman (1988) and van Benschoten and Edzwald (1990).

How do I measure it?

Aluminium residuals are not routinely monitored in emergency water treatment

Aluminium residuals are not routinely monitored in emergency water treatment. Only a few studies have reported on its occurrence during emergencies (Clarke et al., 2004; Fredlund, 2005), noting that its measurement was part of research studies and not routine monitoring. Rather high final aluminium levels of just below and over 0.2 mg/l were reported in those studies, which utilized spectrophotometric and other colorimetric methods for the aluminium determinations (not usually available during emergencies). These methods rely on the addition of specific reagents that will react with aluminium resulting in the formation of a coloured solution in proportion to the concentration of the metal. The samples are read by instruments known as spectrophotometers or colorimeters that can detect the intensity of the colour formed and relate it to the colour formed with solutions of known aluminium concentrations (i.e. calibration standards). Colour comparators for aluminium determinations, such as those used for free chlorine and pH measurements, rely on the same principles of spectrophotometry and are a simpler and cheaper alternative suitable for field use such as in emergencies.

The main drawback of these colorimetric methods is that they can suffer from interference from several sources (*Standard Methods,* 1995), such as phosphates, fluoride, iron, manganese and alkalinity, resulting usually in the underestimation of the aluminium concentration. Furthermore, because aluminium is one of the most ubiquitous elements in nature, it too can cause interference due to the contamination of samples with aluminium from external sources (i.e. other than aluminium from the coagulant), leading to overestimates of aluminium levels. As such, all objects in contact with the sample and instruments used for aluminium analysis should be carefully cleaned and tested for possible aluminium interference.

All objects in contact with the sample should be cleaned to avoid aluminium interference

Another important consideration in the measurement of aluminium is the determination of which form the aluminium residual is in: particulate or dissolved. Such information will be useful in designing a control strategy for the aluminium residuals from coagulation (see ‘How can I control it?’ below). The dissolved fraction can be determined simply by filtering the sample through a 0.45 μm ‘pore-size’ membrane, such as those used in the detection of thermotolerant (faecal) coliforms in DelAgua Kits. The particulate aluminium is not measured directly, rather it is calculated as the difference between the total (unfiltered) and dissolved (filtered) aluminium. The pH and turbidity of the original (unfiltered) sample should always be recorded, as these parameters can help to identify the cause of the high residual aluminium (see below).

How can I control it?

Although high aluminium levels may occur because of the overdosing of alum, the simple reduction of alum dose will not necessarily reduce the final residual level. High aluminium residual concentrations can also occur as a result of alum underdosing. The most appropriate control strategy for aluminium residuals will depend on the form in which the aluminium is present. Particulate aluminium can originate from aluminium hydroxide precipitates (i.e. flocs) and usually has a direct correlation with turbidity (Bérubé, 2004). In such cases, improvements in settling conditions can be achieved through coagulation optimization (i.e. jar-testing). Alternatively, simple process upgrades such as ‘floating outlets’ made with jerrycans (Dorea, 2007) can prevent the carryover of flocs from settling tanks, aiding in the reduction of the turbidity-causing particulate aluminium.

Lime may be added to increase the pH to normal, to reduce dissolved aluminium levels

High dissolved aluminium concentration is usually associated with pH below 5.5 as a result of its increased solubility at lower pH values (Figure 1). Increasing the coagulation pH by lime or sodium hydroxide addition to the 6.0 to 7.0 (i.e. minimum solubility) range could aid in reducing the dissolved residual levels by precipitation. This has been shown to be an effective strategy in non-emergency conditions (Bérubé and Dorea, 2008), remembering that this is also the pH range in which good turbidity reductions can be expected (Twort et al., 2000). The lime or sodium hydroxide could be added to the water before the dosing of the alum to result in a (final) coagulation pH between 6.0 and 7.0, noting that there is no recorded experience of this procedure in emergencies. Any dissolved aluminium residual reducing strategy should be tested beforehand by jar-testing and must not jeopardize the turbidity reduction efficiency of the coagulation process, as this may affect the disinfection process and can make the finished water less aesthetically appealing. Details of a simplified procedure for emergency field jar-testing can be found elsewhere (Dorea, 2007, 2009). Although lime can be sourced with relative ease in most places and could be used to try to control dissolved aluminium, it may be the case that this strategy is unfeasible or impractical to implement. In such cases, if possible, an alternative water source should be considered. Otherwise, one may simply need to accept the less efficient treatment process and ensure that there is adequate terminal disinfection to guarantee the microbial safety of the finished water, which is an overriding priority.

What about the sludge?

Sludge is the by-product of the coagulation process, consisting mainly of aluminium hydroxide precipitates and the contaminants removed by alum (e.g. organic and inorganic substances, pathogens, etc.). Typically, most of the residual waste produced in conventional (i.e. non-emergency) water treatment is treated and conditioned prior to disposal in sewers and landfills or application to land. During the 1970s, the most widely practised disposal method in the USA was discharge without treatment into watercourses; this practice now only accounts for 11 per cent of the residual water treatment waste produced (AWWA, 2006).

Owing to its organic content, the reuse of coagulation sludge through its application to land as a fertilizer is also an option in many places. However, land application of alum sludge may be undesirable because it may absorb inorganic phosphorus from the soil, inhibiting phosphorus uptake by plants (Cornwell, 1999). Moreover, alum sludge can also present aluminium phytotoxicity (i.e. toxicity to plants) depending on its pH-dependent solubility. The capacity to monitor and mitigate such effects during an emergency is likely to be non-existent. Therefore, the land application of coagulation sludge is not recommended in order to avoid issues with local farmers and beneficiaries who could lose their crops as a result.

The land application of coagulation sludge is not recommended

In emergency settings it is highly unlikely that there will be sludge disposal facilities and sludge is usually disposed of into watercourses. Depending mainly on the volume and pH of the receiving water (among other water chemistry characteristics), the disposal of the aluminium-laden sludge can have negative impacts on the aquatic ecosystem with consequences ranging from reduced microbial respiration to toxic effects on fish (e.g. Gensemer and Playle, 1999; Dorea and Clarke, 2008). The toxic effects of aluminium on aquatic life are better understood than the possible effects on humans. Interestingly, aluminium emissions into the environment are subject to less regulation than in drinking water (Gardner and Comber, 2003); however, some guideline values are even more stringent: for example, 0.055 mg/l total Al for pH > 6.5 (ANZECC, 2000).

Disposal of coagulation sludge into watercourses may be the only viable option

Although the disposal of coagulation sludge into watercourses may be considered undesirable from an environmental point of view, it may the only viable alternative in emergencies. During such situations there are likely to be limitations on staffing and resources to treat and condition the sludge prior to disposal. Care should be taken that the sludge is disposed of at a point downstream of the treatment plant intake. Alternatively, a drainage ditch can be dug to drain the sludge and subsequently bury it as a temporary solution. It should be borne in mind that safeguarding human health through an adequate supply of water is set at the highest priority during emergencies and can be effectively achieved with alum coagulation, despite the possible environmental impacts of the alum sludge disposal practices.

What about other coagulants?

Alternative coagulants to alum include ferric salts (e.g. ferric chloride and ferric sulfate), polyaluminium chloride, and polyelectrolytes, among others. With the exception of ferric chloride, which is used by Médecins Sans Frontières (van Den Noortgate and Goessens, 2003), none of these coagulants is routinely used in emergencies. For that particular aid agency, the choice of ferric chloride over alum is based on its relative ease of dissolution in water (in comparison to alum) and the wider pH range of 5.0 to 9.0 in which it is effective. The recognized shortcomings of ferric chloride are that it can be difficult to source in the field and, similarly to alum, that it can leave an iron coagulant residual that can result in discoloured finished water with a 'yellowish' appearance; this could raise issues in terms of user acceptability.

Ferric chloride can be difficult to source in the field

Many of the other coagulants are only available in liquid form which may limit the ease with which they can be air freighted, as in such form they may be classified as a 'dangerous good'. Factors such as relatively low price, level of skill and equipment necessary for process control, and local availability for initial stock replenishment do not favour the use of alternative coagulants, particularly when compared with alum. It should be noted that, of the alternatives to alum, special attention should be given to polyelectrolytes, as these water treatment chemicals can leave coagulant residuals such as acrylamide and epichlorohydrin. Health concern over such contaminants has led to the ban or strict control over the use of such coagulants in some countries (Letterman and Pero, 1990). As such, it would be ill-advised to use polyelectrolytes in emergency water treatment, given the likely difficulties in measuring and controlling polyelectrolyte residuals in field conditions.

Other alternatives may leave coagulant residuals that cause concern

So, should I be worried about using alum?

Given its successful track record in emergency water treatment, it is somewhat unfortunate that unwarranted concern is sometimes associated with this coagulant. Alum has many advantages (e.g. local availability, relative ease of transport, adaptability to local conditions and ease of assimilation by unskilled operators) in addition to a widespread application and know-how. As such, its use should not be compromised through fear of postulated health risks.

There are many good reasons to maintain low aluminium levels

Alum is capable of assisting emergency water treatment programmes by providing adequate amounts of good quality water as well as facilitating terminal disinfection. Thus, it plays an important role in the prevention of acute infectious illnesses such as diarrhoeal diseases. Such benefits outweigh any possible chronic health risks associated with long-term exposure to residual aluminium, particularly considering the short time span of emergencies. Nonetheless, there are many good reasons besides possible health risks to maintain low aluminium levels. It is a simple matter of good engineering practice: water purification processes should ideally take substances out and not add more (except for an adequate free chlorine residual)! Furthermore, it has been shown that there are appropriate methods that can be applied to measure and mitigate high residual aluminium levels in emergency field conditions. Problems with the use of alum (as with any water treatment technique) can be avoided with proper training and well-informed professionals.

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