Aluminum Sulfate

Livestock

Identification of Petitioned Substance

Chemical Names:
Aluminum Sulfate
Aluminum Sulphate
Sulfuric acid, aluminum salt

Other Name:
Alum

Trade Names:
Al+Clear® Poultry Grade Alum
Liquid Al+Clear®
Al+Clear® A7

CAS Numbers:
Solid Aluminum Sulfate:
10043-01-3 (Aluminum sulfate)

Liquid Aluminum Sulfate:
7784-31-8 (Aluminum sulfate 18-hydrate)

Acidified Liquid Aluminum Sulfate:
7664-93-9 (Sulfuric acid)
7784-31-8 (Aluminum sulfate 18-hydrate)

Other Codes:
EC #233-1350[1]
PubChem 24850[2]
Chemspider 2323[3]

[1] European Community number used for regulatory purposes with the European Union
[2] Main database for National Center for Biotechnology Information
[3] Database of chemicals owned by the Royal Society of Chemistry

Summary of Petitioned Use

Aluminum sulfate is a synthetic substance being petitioned for treatment of carbonaceous poultry litter primarily for the purpose of reducing ammonia volatilization, which is a management challenge in commercial poultry production. The petition states that the intended and current use of aluminum sulfate is as a poultry and livestock bedding amendment, but this report covers its use primarily on poultry bedding. Ammonia is an irritant to the respiratory system of birds, and mitigating ammonia gasses in the poultry house increases animal welfare and improves flock performance (Moore and Watkins 2012). In addition to litter treatment in the poultry house, aluminum sulfate is being petitioned for use in organic crop production as a poultry litter additive. Litter treated with aluminum sulfate differs from non-treated litter, as it contains more total nitrogen and less soluble phosphorous, which increases the nitrogen fertilizer value and reduces phosphorous pollution of surface waters (Moore and Watkins 2012).

Characterization of Petitioned Substance

<u>Composition of the Substance:</u>
Aluminum sulfate is an acid salt consisting of aluminum cations (Al^{+3}) and sulfate anions (SO_4^{2-}). Aluminum sulfate is marketed in three forms as a dry granule, a liquid, and an acidified liquid. The molecular formula of the dry formulation (Al+Clear® Poultry Grade Alum) is $Al_2(SO_4)_3$. In the liquid formulation (Liquid Al+Clear®), it exists as hydrated aluminum sulfate $Al_2(SO_4)_3 \cdot nH_2O$. The acidified formulation (Al+Clear® A7) also contains added sulfuric acid (H_2SO_4).

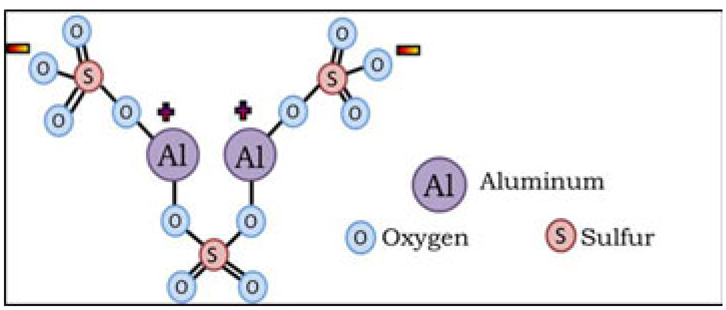

Figure 1: Molecular structure of aluminum sulfate (ChemSpider 2014)

<u>Source or Origin of the Substance:</u>
The manufacturing process for all the forms of aluminum sulfate included in the petition involves reacting liquid sulfuric acid with either bauxite ore containing aluminum hydroxide ($Al(OH)_3$) and hydrated aluminum ($Al_2O_3{\cdot}3H_2O$), or synthetic hydrated aluminum previously refined from bauxite. Bauxite ore is the main source of aluminum for the world and contains various aluminum minerals and two iron minerals (Amethyst Galleries 2014). The process creates hydrated aluminum sulfate per the following reactions:

From bauxite: $3\ H_2SO_4 + 2\ Al(OH)_3\ + 12\ H_2O \rightarrow Al_2(SO_4)_3 \cdot 18\ H_2O$
From hydrated aluminum: $3\ H_2SO_4 + Al_2O_3{\cdot}3H_2O\ + 12\ H_2O \rightarrow Al_2(SO_4)_3 \cdot 18\ H_2O$

The acidified formulation also contains synthetically produced sulfuric acid.

<u>Properties of the Substance:</u>

Table 1. Physical properties of aluminum sulfate formulations.

Description	Liquid Aluminum Sulfate	Dry Ground Aluminum Sulfate	7% Acid Liquid Aluminum Sulfate
Aluminum Metal %	4.2	9.10	3.25
Aluminum as Aluminum Oxide (Al_2O_3) %	8.0-8.3	17.0-17.1	6.1-6.3
Density lb/ft^3 or lb / gal	11.1	41 (powder) – 63-71 (granular)	10.6-10.8
Aluminum sulfate (lb/gal)	5.4	n/a	3.9
Specific Gravity	1.33-1.34	n/a	1.27-1.29
pH	2.4	3.5 in 1% solution	<2.0
Freezing Point	-15ºC/5ºF	n/a	-14ºC/7ºF

Specific Uses of the Substance:

Aluminum sulfate is applied to poultry litter in commercial poultry houses to lower pH of the litter, thus decreasing ammonia volatilization. This improves air quality for the birds and reduces pathogen load, resulting in improved weight gain and flock health (Moore et al. 1996). Furthermore, ammonia creates atmospheric particulate matter causing haze and odor, and reducing ammonia losses has a beneficial impact on the atmosphere (NOAA 2000).

Litter used as bedding in poultry production commonly includes wood shavings, rice hulls and peanut hulls or other carbonaceous organic debris for absorbing manure produced by poultry (Lacy 2002).

Aluminum sulfate allows litter to be used for more poultry flocks before the litter must be removed, thus reducing the input costs and associated environmental resource implications of more frequent litter changes (Sims and Luka-McCafferty 2002).

When the litter is cleaned out from the poultry house, it serves as a source of fertilizer. Litter treated with aluminum sulfate contains higher levels of nitrogen than non-treated litter.. Litter treated with aluminum sulfate also contains less soluble phosphate due to phosphate complexation with aluminum. Excessive phosphorous accumulation is common when organic residues are applied annually for their nitrogen value, and phosphorous is a dangerous pollutant in surface aquatic systems (Moore and Edwards 2005). Runoff from fields where poultry litter is applied contains less phosphorous if the litter is treated with aluminum sulfate, thereby reducing the environmental phosphorous pollution that causes algal blooms in aquatic environments (Moore and Edwards 2005).

Aluminum sulfate is commonly used in conventional poultry production to reduce ammonia volatilization and accumulation in poultry houses, which reduces energy use associated with ventilation and heating. Furthermore, the pathogen load is reduced, and bird weight gain is enhanced due to the lower ammonia and pathogen loads. When the treated litter is spread on land, phosphorus runoff is greatly reduced (Moore and Edwards 2005). Granular aluminum sulfate is applied to litter in the poultry house using a variety of spreader equipment types including de-caking machines, drop spreaders, fertilizer spreaders, or manure spreaders. After spreading it on the surface, label instructions recommend to mechanically work the aluminum sulfate into the litter to avoid bird ingestion, and to enhance the ammonia mitigating efficacy (General Chemical 2010). Liquid aluminum sulfate is typically only applied by certified professional applicators, not by producers themselves, due to transport restrictions on the material (Moore and Watkins 2012). Rates of application range between 5 and 10 percent of the weight of the litter. Low rates typically only control ammonia during the brooder phase of production, when birds are most susceptible to ammonia injury. Low rates also do not prevent phosphorus runoff as well as higher rates (Moore and Watkins 2012). Poultry litter in conventional management systems typically includes wood shavings, rice hulls, and peanut hulls, but a variety of carbonaceous residues could be used. Aluminum sulfate application rates are dictated by the quantity of nitrogen excreted by the poultry, and university recommended guidelines do not consider litter type in application rates (Shah et al. 2006).

Approved Legal Uses of the Substance:

Under FDA regulations, aluminum sulfate is generally recognized as safe (GRAS) as a food additive when used in accordance with good manufacturing or feeding practice (CFR 182.1125(b)). As a food additive, aluminum sulfate is used for pH adjustment in cheese making, pickling and baked goods. It is also used as a styptic for treating wounds. Aluminum sulfate is a precursor in the manufacture of the potassium and sodium-aluminum sulfates used as antiperspirants in personal deodorant products. Aluminum sulfate is not classified by the EPA as an inert of toxicological concern, and is approved as a adjuvant in pesticide formulations, used pre-harvest, and is exempted from the requirement of a tolerance (40 CFR 180.920).

Due to the beneficial impacts on the environment, producers who use aluminum sulfate can apply for cost sharing to implement Conservation Practice Standard 591, Amendments for Treatment of Agricultural Waste under the USDA-NRCS Environmental Quality Incentive Program (EQIP). The practice standard applies to the practice of altering the physical and chemical characteristics of a waste stream as a part of a planned manure or waste management system. The purpose of the practice standard is to facilitate the management and handling of manure and waste, reducing the risk of contamination by pathogens, and to improve or protect air and water quality and animal health. Use of aluminum sulfate is not specified in the conservation practice, but the practice allows for alternative treatment products provided the manufacturer submits peer-reviewed research documenting efficacy from universities or other independent research entities. However, aluminum sulfate and sodium bisulfate are the dominant products used for meeting the practice standard in conventional agriculture (NRCS 2013). The rate of cost share varies between states, and is based on the square footage of the poultry house. The cost share is not available in all states (NRCS 2013).

The cost share under conservation practice standard 591 is only available to livestock producers. However, the implementation of a nutrient management plan, activity code 104, can lead to subsidies for crop producers. To develop a nutrient management plan, producers must consider land-grant university recommended technologies that improve nutrient use efficiency and minimize surface or ground water resource concerns (NRCS 2012). Within the paradigm of this standard, aluminum sulfate litter treatment is a very effective tool to accomplish these goals in an economical manner (Moore and Watkins 2012). However, activity code 104 requires organic operations to manage nutrients in a manner consistent with the NOP.

Action of the Substance:

Aluminum sulfate reacts with water in litter to precipitate aluminum hydroxide and generates acidity through the hydrolysis of water (McBride 1994). The increase in acidity shifts the equilibrium towards generating more sulfuric acid than would originally be present.

$$Al_2(SO_4)_3 \rightleftharpoons 2Al^{3+} + 3SO_4^{2-}$$

$$2Al^{3+} + 4H_2O \rightleftharpoons 2AlOH^{2+} + 2\,H_3O^+ \qquad K = 10^{-4.97}$$

$$2AlOH^{2+} + 4\,H_2O \rightleftharpoons 2Al(OH)_2^+ + 2\,H_3O^+ \qquad K = 10^{-4.93}$$

$$\underline{2Al(OH)_2^+ + 4\,H_2O \rightleftharpoons 2Al(OH)_3 + 2\,H_3O^+ \qquad K = 10^{-5.70}}$$

$$Al_2(SO_4)_3 + 12\,H_2O \rightleftharpoons 2Al(OH)_3 + 6H_3O^+ + 3SO_4^{2-}$$

$$3\,H_3O^+ + 3\,SO_4^{2-} \rightleftharpoons 3\,HSO_4^- + 3\,H_2O \qquad K = 10^{-2.00}$$

$\underline{3\ HSO_4^- + 3\ H_3O^+ \rightleftharpoons 3\ H_2O + 3\ H_2SO_4 \qquad K=10^{-6.38}}$

$6\ H_3O^+ + 3\ SO_4^{2-} \rightleftharpoons 6\ H_2O + 3\ H_2SO_4$

Resulting reaction (McBride 1994): $Al_2(SO_4)_3 + 6\ H_2O \rightleftharpoons 3\ H_2SO_4 + 2\ Al(OH)_3$.

The initial application of aluminum sulfate and subsequent sulfuric acid production lowers the pH of the poultry litter initially to 5.7 but returns to near-neutral (pH 7.0) after 3-4 weeks (Moore et al. 2000). Aluminum hydroxides produced by the reaction with water further react with phosphates (PO_4^{3-}) in the litter to form insoluble aluminum phosphate ($AlPO_4$). The H^+ ion reacts with ammonia gas in litter (NH_3) to form aqueous ammonium (NH_4^+) which is retained in the litter, increasing its nitrogen fertilizer value.

$NH_3 + H_3O^+ \rightleftharpoons NH_4^+ + H_2O$

$2NH_4^+ + SO_4^{-2} \rightleftharpoons (NH_4)_2SO_4 \qquad K'=10^{8.77}$

Ammonia fluxes from alum-treated litter have been shown to be 70% lower than non-treated litter (Moore et al. 2000). The assumed mechanism for the decrease in ammonia volatization is its conversion to ammonium sulfate (Moore et al 1996). The dissociation of ammonium sulfate was measured to be $K=10^{-8.77}$ at 0.7M, making the inverse equilibrium reaction $K'=10^{8.77}$ (Maeda and Iwata 1997). After land application, NH_4^+ is either taken up by plants, volatized to the atmosphere as NH_3 gas if surface applied at high pH, or oxidized to NO_3^{2-} by nitrifying bacteria.

<u>Combinations of the Substance:</u>

Hydrated aluminum sulfate is diluted with water to form liquid aluminum sulfate. Synthetic sulfuric acid is added to liquid aluminum sulfate to produce acidified aluminum sulfate (Al+Clear® A7). Additional adjuvants are not added.

Status

<u>Historic Use:</u>

There is no precedent for use of aluminum sulfate in historical organic production. Aluminum sulfate has been commonly used for several decades in conventional poultry settings for the purposes of reducing ammonia volatilization and improving flock health (Sims and Luka-McCafferty 2002).

Historical use of aluminum sulfate is recorded in Greek and Roman literature as a mordant for enhancing adhesion of fabric dyes. Production methods have evolved over the centuries encompassing various extraction methods from mineral and clay sources. Other uses for aluminum sulfate include printing, tanning hides, fireproofing, paper production, water clarification, sewage treatment, and as a food additive for pH adjustment (Umhau 1936).

Poultry litter has a strong historical precedent as an organic fertilizer, with an average fertilizer grade of 3-3-2 (N-P_2O_5-K_2O). If poultry litter is surface-applied without incorporation, approximately 50% of the total N will be available during the first year, and if the litter is incorporated rapidly after application approximately 60% will be available the first year (Zublena et al. 1997).

<u>Organic Foods Production Act, USDA Final Rule:</u>

Aluminum sulfate is not specifically listed in the Organic Foods Production Act of 1990, although it is a mineral, and therefore appears generically under section 2118(c)(1)(B)(i).

Aluminum sulfate is not listed in the USDA organic regulations.

International

Canada - Canadian General Standards Board Permitted Substances List
http://www.tpsgc-pwgsc.gc.ca/ongc-cgsb/programme-program/normes-standards/internet/bio-org/documents/032-0311-2008-eng.pdf
Aluminum sulfate is not permitted under the Canadian Organic Standards, as it does not appear in CAN/CGSB-32.311-2006, Table 5.3 Livestock Health Care Products and Production Aids.

CODEX Alimentarius Commission, Guidelines for the Production, Processing, Labelling and Marketing of Organically Produced Foods (GL 32-1999)
http://www.fao.org/organicag/doc/glorganicfinal.pdf
Aluminum sulfate is not included in GL 32-1999 Annex 1(B) Livestock and Livestock Products.

European Economic Community (EEC) Council Regulation, EC No. 834/2007
http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32007R0834&from=EN
Aluminum sulfate is not permitted, as it does not appear in 834/2007 Article 14 Livestock Production Rules.

European Economic Community (EEC) Council Regulation, EC No. 889/2008
http://eur-lex.europa.eu/legal-content/EN/TXT/PDF/?uri=CELEX:32008R0889&from=EN
Aluminum sulfate is not permitted, as it does not appear in 889/2008 Chapter 2 Livestock Production.

Japan Agricultural Standard (JAS) for Organic Production
http://www.maff.go.jp/e/jas/specific/pdf/836_2012-2.pdf
Aluminum sulfate is not permitted, as it does not appear in Notification 1608 Japanese Agricultural Standard for Organic Livestock, Partial Revision March 28 2012.

International Federation of Organic Agriculture Movements (IFOAM)
http://www.ifoam.org/sites/default/files/ifoam_norms_version_july_2014.pdf
Aluminum sulfate is not permitted, as it does not appear in IFOAM Norms Appendix 5, Substances for Pest and Disease Control and Disinfection in Livestock Housing and Equipment.

International summary of chemically-related substances:
Several other metal-sulfate substances are permitted in international regulations for various uses in organic production including copper sulfate, iron sulfate, calcium sulfate (mined), magnesium sulfate (mined), potassium sulfate (mined), and zinc sulfate for correcting plant nutrient deficiencies. Copper sulfate is also approved as a fungicide. International organic regulations generally prohibit sulfates of copper, iron, calcium, magnesium, potassium, synthetically produced using sulfuric acid. Aluminum sulfate differs from the allowed materials listed above, as 1) aluminum is neither a plant nutrient nor a fungicide, 2) aluminum sulfate is synthetically produced using sulfuric acid, and 3) none of the other allowed metal sulfates serve as acidifying agents.

Evaluation Questions for Substances to be used in Organic Crop or Livestock Production

Evaluation Question #1: Indicate which category in OFPA that the substance falls under: (A) Does the substance contain an active ingredient in any of the following categories: copper and sulfur compounds, toxins derived from bacteria; pheromones, soaps, horticultural oils, fish emulsions, treated seed, vitamins and minerals; livestock parasiticides and medicines and production aids including netting, tree wraps and seals, insect traps, sticky barriers, row covers, and equipment

cleansers? (B) Is the substance a synthetic inert ingredient that is not classified by the EPA as inerts of toxicological concern (i.e., EPA List 4 inerts) (7 U.S.C. § 6517(c)(1)(B)(ii))? Is the synthetic substance an inert ingredient which is not on EPA List 4, but is exempt from a requirement of a tolerance, per 40 CFR part 180?

A. The substance aluminum sulfate contains sulfur.

B. The substance aluminum sulfate is **not** classified by the EPA as an inert of toxicological concern (it is on EPA List 4 (2004)). The substance is, however, approved as an adjuvant, used pre-harvest, and is exempted from the requirement of a tolerance (40 CFR 180.920).

__Evaluation Question #2:__ **Describe the most prevalent processes used to manufacture or formulate the petitioned substance. Further, describe any chemical change that may occur during manufacture or formulation of the petitioned substance when this substance is extracted from naturally occurring plant, animal, or mineral sources (7 U.S.C. § 6502 (21)).**

Three formulations of aluminum sulfate are listed in the petition, including solid aluminum sulfate, liquid aluminum sulfate, and acidified liquid aluminum sulfate.

The manufacturing process for all formulations involves reacting either aluminum hydroxide ($Al(OH)_3$) or hydrated aluminum ($Al_2O_3{\cdot}3H_2O$) with liquid sulfuric acid. The process creates hydrated aluminum sulfate per the following reactions:

From bauxite: $3\ H_2SO_4 + 2\ Al(OH)_3 + 12\ H_2O \rightarrow Al_2(SO_4)_3 \cdot 18\ H_2O$

From hydrated aluminum: $3\ H_2SO_4 + Al_2O_3{\cdot}3H_2O + 12\ H_2O \rightarrow Al_2(SO_4)_3 \cdot 18\ H_2O$

The reaction is exothermic, and hydrated aluminum sulfate is produced at approximately 110^{o}C. The hydrated aluminum sulfate can be placed in pans and thermally dehydrated to produce solid aluminum sulfate ($Al_2(SO_4)_3$), or water can be added to produce diluted liquid aluminum sulfate (Dogan 2000). Acidified liquid aluminum sulfate is produced by adding synthetic sulfuric acid to liquid aluminum sulfate.

The precursor bauxite is a naturally-occurring, mined aluminum ore containing aluminum hydroxide ($Al(OH)_3$), hydrated aluminum ($Al_2O_3{\cdot}3H_2O$), kaolinite clay, titanium dioxide (TiO_2), hematitie (Fe_2O_3), goethite ($FeO(OH)$), and silicon dioxide (SiO_2) (Ayres et al. 2007). After processing for aluminum sulfate, the kaolinite and non-aluminum oxides remain as inert waste by-products that are used in other industrial products.

Mineral extraction of bauxite is very limited in the United States, but it is extracted in large quantities in Australia, Brazil, China, Guinea, India and Indonesia. As of 2014, Jamaica and Guinea combined supply 70% of the bauxite imported to the United States, with 95% of imported bauxite used for production of aluminum oxide, and 90% of the aluminum oxide produced is smelted into aluminum metal. Most domestic U.S. bauxite extraction is processed into non-metallurgical products (USGS 2014). After mining, bauxite is crushed, washed to remove clays, and dried in a rotary kiln (Ayres et al. 2007).

The precursor hydrated aluminum $Al_2O_3{\cdot}3H_2O$ listed in the petition occurs naturally as a component of bauxite ore, and is also synthetically purified from bauxite ore as an intermediary in the Bayer process for producing aluminum oxide (Al_2O_3) (Ayres et al. 2007; Sonthalia et al. 2013). In this process, sodium hydroxide is added to bauxite ore to increase pH at 150-160^{o}C and 0.5 MPa pressure to solubilize $Al(OH)_4^-$ (aqueous) and is separated from SiO_2, TiO_2, and Fe_2O_3 contaminants which are insoluble at high pH via filtration. The cooled $AlOH_4^-$-containing solution is seeded with fine gibbsite crystals to initiate precipitation of hydrated aluminum, which is hastened by decreasing pH by bubbling CO_2 through the solution. At this stage, excess hydrated aluminum can be filtered and dried. The final phase of the Bayer process is not applied to hydrated aluminum destined for aluminum sulfate production, which would entail high temperature calcination at 1,100 – 1,200°C, producing pure Al_2O_3 (Carter and Norton 2007).

The precursor, sulfuric acid (H_2SO_4), is a synthetic chemical produced by the oxidation of sulfur to SO_2 gas by burning. The SO_2 is further oxidized to SO_3 by either the chamber process, which involves nitrogen compounds with several intermediate reactions, or by the contact process, which relies on metal catalysts to facilitate the oxidation. Various catalysts exist including formulations of ferric oxide, platinum, or vanadium, which serves as the primary catalyst for modern sulfuric acid production. SO_3 is reacted with water to form sulfuric acid (H_2SO_4) (Friedman and Friedman undated). Sulfuric acid was petitioned to the NOP in 2005 and again in 2012 for acidification of poultry litter to below pH 4.5 but not less than 3.5; however it was not recommended by the NOSB for listing at either petition date based on rationale that "Sulfuric acid, when used in livestock manure, is changed to sulfate, which is in this case a synthetically derived plant nutrient" (NOSB 2012). Sulfuric acid is, however, allowed under the NOP for pH adjustment of liquid fish products, so long as the pH is not reduced below 3.5 (7 CFR 205.601(j)(7)).

Evaluation Question #3: Discuss whether the petitioned substance is formulated or manufactured by a chemical process, or created by naturally occurring biological processes (7 U.S.C. § 6502 (21)).

The reaction to produce aluminum sulfate is an acid-base reaction between sulfuric acid and aluminum hydroxide as the base. The hydrated product is thermally dehydrated by evaporation into solid form, or diluted into liquid form.

Although the petitioned aluminum sulfate products are manufactured by chemical processes, aluminum sulfate is also found in nature as mineral deposits formed either through sublimation in volcanic regions, or through weathering of iron sulfates (pyrite) in the presence of aluminous shale. Naturally occurring aluminum sulfate is often contaminated with potassium, sodium, magnesium, manganese, iron, copper, silica, or fluoride, and is not found in sufficient abundance to warrant extraction (Umhau 1936).

Synthetic sulfuric acid is used in the production of aluminum sulfate; however, it is not specifically present in the dry non-hydrated form of aluminum sulfate, but is a rapidly produced degradation product when the substance is applied as petitioned. Synthetic sulfuric acid is still present in liquid aluminum sulfate (CAS 7784-31-8) products. Acidified aluminum sulfate contains enhanced levels of synthetic sulfuric acid.

Evaluation Question #4: Describe the persistence or concentration of the petitioned substance and/or its by-products in the environment (7 U.S.C. § 6518 (m) (2)).

Although total aluminum application to land does increase as a result of aluminum sulfate application, it exits as insoluble aluminum hydroxides or aluminum phosphates. Because aluminum sulfate-treated litter and non-treated litter both tend to increase soil pH, free aluminum in soil is reduced by application of either, as aluminum solubility is controlled by pH as it becomes less soluble with increasing pH. Total aluminum (as aluminum compounds) comprises approximately 7.2% of native soils, which weigh several million pounds per acre depending on depth; land-application of <50 lb total aluminum / acre / year has a miniscule impact on total aluminum content in the soil (Shacklette and Boerngen 1984).

Unlike aluminum, sulfate added to litter from aluminum sulfate application is a crop nutrient. Sulfate is an anion which is not bound on the soil cation exchange capacity, and is subject to leaching with rainfall or irrigation (Havlin et al. 2005).

<u>Evaluation Question #5:</u> Describe the toxicity and mode of action of the substance and of its breakdown products and any contaminants. Describe the persistence and areas of concentration in the environment of the substance and its breakdown products (7 U.S.C. § 6518 (m) (2)).

Toxicity: Aluminum sulfate is considered a dry acid, and is an irritant to the skin and eyes (UN-LIO 2012). However, acidity created by the substance is neutralized by the litter, and litter applied to land generally has a near-neutral pH (Sims and Luka-McCafferty 2002).

Mode of action: Aluminum sulfate reacts with water to create acid, which reduces ammonia losses from litter in confined poultry operations. Furthermore, aluminum causes precipitation of phosphates, reducing phosphorus solubility in the land-applied litter (Moore and Watkins 2012).

Breakdown products:
Breakdown products of aluminum sulfate include Al^{3+}, $Al(OH)^{2+}$, $Al(OH)_2^{+}$, $Al(OH)_3$, SO_4^{2-}, HSO_4^{-}, and H_2SO_4, and H_3O^{+} (McBride 1996). Aluminum phosphate ($Al(PO_4)$) precipitate is also formed via reaction of Al^{3+} with phosphates in the litter (Warren et al. 2008).

Toxicity of breakdown products:
Free Al^{3+} is a toxic species that increases in concentration as pH decreases, and typically reaches phytotoxic levels when pH falls below 5.0 (Havlin et al. 2005). Poultry litter without aluminum sulfate typically ranges in pH from 8.0 to 8.9 (Sims and Luka-McCafferty 2002). Shortly after aluminum sulfate application, pH of the litter decreases to about 5.7, but becomes neutralized (near pH 7.0) after 3-4 weeks due to reaction with NH_3 in the poultry guano (Moore et al. 2000). Thus, although adding aluminum sulfate increases total concentration of aluminum, persistence of the toxic Al^{3+} species is not enhanced. In contrast, application of litter near pH 7.0 to acidic soils decreases solubility of toxic Al^{3+} (Moore and Edwards 2005).

Persistence of the breakdown products:
Aluminum hydroxide and phosphates from aluminum sulfate addition to poultry litter are persistent in the soil after land application due to low solubility (Warren et al. 2008). Sulfates, however, are more soluble, serve as a source of sulfur for crop plants, or are lost to leaching (Havlin et al. 2005).

Contaminants:
The primary contaminants present in the Al_2O_3 precursor to aluminum sulfate include SiO_2, Fe_2O_3, and Na_2O, and could carry though into the final aluminum sulfate product, however do not pose toxicological concerns (Carter and Norton 2007).

<u>Evaluation Question #6:</u> Describe any environmental contamination that could result from the petitioned substance's manufacture, use, misuse, or disposal (7 U.S.C. § 6518 (m) (3)).

Aluminum sulfate is a dry acid, and can create zones of high acidity if accidentally spilled. Acid damage severity from a concentrated spill is dependent on the quantity spilled, and also on the moisture available for reacting. If the spilled material does not come into contact with moisture, the majority of the material could be cleaned up before significant acidification occurs. . But, surfaces of most soils are typically fissured and loose, and sometimes moist, making complete soil cleanup unlikely. Aluminum sulfate is designated as a hazardous substance under the CERCLA (superfund), and discharges exceeding 5,000 lbs (2,270 kg) require notification to the U.S. Environmental Protection Agency (TABLE 302.4 40 CFR).

Localized environmental acidification has a profound impact on chemical equilibrium regulating biological systems. In the soil, acidic conditions cause enhanced solubility of the Al^{3+} species, which is toxic to plant roots. Furthermore, both H^{+} and Al^{3+} are more strongly adsorbed to soil cation exchange sites than calcium, magnesium, and potassium and cause potential soil depletion of these nutrients via leaching. Soil remediation of large aluminum sulfate spills can be accomplished with a liming agent to neutralize the acidity and reduce solubility of Al^{3+} (NIH 2014).

Aluminum sulfate is sometimes deliberately added to water bodies impaired by phosphorus eutrophication, but accidental discharge of large quantities could cause excessive water acidification and subsequent solubilization of Al^{3+} which is toxic to aquatic organisms (UN-ILO 2012).

Personal protective equipment should be used when applying aluminum sulfate in the poultry house, but no specific precautions are needed for handling spent litter treated with aluminum sulfate due to the high level of dilution in the litter. In the poultry house, any aluminum sulfate spills should be incorporated into the litter to prevent ingestion by the birds (Walker and Burns 2000). Applications of liquid ammonium sulfate are typically made by certified applicators due to transport restrictions (Moore and Watkins 2012).

Aluminum sulfate reduces environmental contamination of phosphorus in natural water bodies from surface litter applications, compared to non-treated litter. Moore and Edwards (2005) measured 340% greater cumulative phosphorus load in runoff water from non-treated litter than from treated litter in a paired watershed study.

The process of extracting bauxite ore has a deleterious impact on the environment through habitat degradation and fragmentation by roads, and through carbon emissions (Cooke 1999). After extraction, regulations in some countries require replacement of topsoil and other remediation measures; however quality of land after remediation is unlikely to be equivalent to before-extraction parameters (Cooke 1999). Most of the bauxite extraction worldwide is for the production of aluminum oxide, and less than 5% of bauxite imported into the U.S. is used for other purposes including aluminum sulfate production (USGS 2014).

<u>Evaluation Question #7:</u> Describe any known chemical interactions between the petitioned substance and other substances used in organic crop or livestock production or handling. Describe any environmental or human health effects from these chemical interactions (7 U.S.C. § 6518 (m) (1)).

Aluminum sulfate is being petitioned as an amendment to poultry litter for consideration in organic livestock application. Aluminum sulfate undergoes various chemical interactions with the poultry litter, altering several key chemical characteristics of the litter:

1. The pH of the litter is reduced; however it is unlikely to fall below pH 7.0 in litter collected after the final grow out flock. Initially the treated litter pH does fall to about 5.7 and that pH is maintained for about 3-4 weeks (Moore et al. 2000) (Table 2).

2. Aluminum sulfate reacts with water and naturally-occurring NH_3 in the litter to form NH_4^+, thus stabilizing nitrogen and reducing NH_3 gas volatilization to the atmosphere. In the soil environment, NH_4^+ is transient and is either rapidly taken up by plants, microbially transformed to NO_3^{2-} which can be taken up by plants or lost to leaching, or anaerobically transformed by microorganisms to N_2 and N_2O which are lost to the atmosphere (Halvin et al. 2005). Although nitrogen is more persistent in the litter, there is no effect on cumulative soil nitrogen accumulation compared to non-treated litter, as aluminum sulfate does not alter the organic fraction of the total nitrogen.

Poultry litter is a significant source of NH_3 in the atmosphere, which causes formation of aerosol particles. It is also a source of nitric acid deposition to land or water bodies where it causes land and water acidification and nitrate pollution (NOAA 2000). Aluminum sulfate decreases atmospheric pollution of NH_3 by reducing litter pH, which converts NH_3 to water-soluble NH_4^+ (Shah et al. 2006). Incubation studies estimate approximately 14 g N / kg litter is lost from non-treated litter as NH_3, while ammonia loss from litter treated with aluminum sulfate ranges between 0.7 to 4.07 g N / kg litter between the high and low application rates (Moore et al. 2000. Assuming 40,000 lbs. of litter for a 16,000 square foot poultry house containing 20,000 broilers (Moore and Watkins 2012), this represents a reduction of about 400 lbs. of NH_3-N lost to the atmosphere over a 42-day period with low rates of aluminum sulfate, and about 560 lbs. of NH_3-N at high rates of aluminum sulfate.

3. Litter treated with aluminum sulfate contains less soluble phosphate (PO_4^{3-}) than non-treated litter, as Al^{3+} reacts with PO_4^{3-} to form insoluble $AlPO_4$ (Table 2). Although the total phosphorous concentration in the litter does not change greatly, phosphorous becomes less plant-available, and likelihood of phosphorous transport to surface water is reduced. Aquatic ecosystems tend to be phosphorous-limited, and phosphorous eutrophication of natural water bodies is reduced when land-applied litter is treated with aluminum sulfate. The insoluble aluminum phosphate is not available to plants as nutrients and instead stays in the soil as a mineral (Moore and Edwards 2005).

4. Litter treated with aluminum sulfate contains both higher total aluminum and higher soluble aluminum than non-treated litter (Table 2); however, runoff from fields where aluminum sulfate-treated litter is applied does not contain significantly higher levels of aluminum than fields where non-treated litter is applied (Moore et al. 1998).

5. Litter treated with aluminum sulfate contains higher total sulfur and higher soluble sulfur than non-treated litter (Table 2).

6. Concentration of soluble arsenic is reduced by aluminum sulfate treatment due to arsenic co-precipitation by aluminum (Violante et al. 2006) (Table 2).

Table 2. Average chemical properties of 194 poultry litter batches either treated or not treated with aluminum sulfate (Sims and Luka- McCafferty 2002)

	Non-treated	Alum-treated	Significant Difference ($p<0.001$)
pH	7.8	7.2	Yes
Moisture	30.2	28.9	No
Total Concentrations			
Al (%)	0.1	1.15	Yes
P (%)	2.22	2.01	Yes
C (%)	35.5	32.3	Yes
N (%)	3.97	4.24	Yes
S (%)	0.77	2.49	Yes
As (ppm)	45	44	No
Cu (ppm)	962	877	Yes
Zn (ppm)	644	581	Yes
Soluble Concentrations			
Al (ppm)	4	23	Yes
P (ppm)	1827	596	Yes
NH_4 (%)	0.074	0.29	Yes
S %	0.36	1.52	Yes
As (ppm)	19	7	Yes
Cu (ppm)	272	172	Yes
Zn (ppm)	29	15	Yes

<u>Evaluation Question #8:</u> Describe any effects of the petitioned substance on biological or chemical interactions in the agro-ecosystem, including physiological effects on soil organisms (including the salt index and solubility of the soil), crops, and livestock (7 U.S.C. § 6518 (m) (5)).

Aluminum sulfate is not applied while birds are in the poultry house. The substance is not applied before the first flock grow-out; however, it is systematically applied thereafter before every flock is exposed to the litter. Any spills or concentrations of the product should be dispersed into the litter to avoid consumption by young chicks (Walker and Burns 2000). As stated in the petition, aluminum sulfate is not applied to feed. In the event of accidental ingestion, aluminum sulfate is corrosive and irrigating to the digestive system and kidneys of birds (Dumonceaux and Harrison 2013). In one study, Japanese quail fed aluminum sulfate as >0.10% of their diet reduced body weight accumulation, eggshell strength, plasma inorganic phosphorous, feed consumption, and egg production (Hussein et al. 1988). Physiological effects of aluminum sulfate intake by broiler chickens occurs at higher intake levels than quail, with decreases in weight gain when consumed at >0.93% of the diet. Higher concentrations of aluminum sulfate in the diet cause more severe depressions in weight gain, decreased bone strength, and serum phosphorous. At application rates of 100 g / kg litter, birds would need to ingest 10% of total dietary intake as litter to exceed 0.93% aluminum sulfate in the diet, and the aluminum would need to be in the original non-reacted aluminum sulfate crystalline form which does not persist in the presence of moisture. Typical observed litter ingestion rates are below this threshold, ranging from 2% to 5% of daily dietary intake. Aluminum sulfate is toxic to poultry if directly ingested in large quantities, but not at levels expected from litter consumption (Huff et al. 1996). When aluminum sulfate is used, mortality decreases and poultry weight gain increases, indicating the birds are likely not suffering toxic effects from incidental aluminum sulfate ingestion from the litter (Walker and Burns 2000).

Deleterious effects of aluminum sulfate on the head, skin, feathers, or feet of poultry were not revealed in the literature review, but the material is an irritant (UN-LIO 2012). If aluminum sulfate remains in its original non-reacted dry form, there is potential for foot irritation. Producers can mitigate the potential of bird exposure by rototilling aluminum sulfate into the litter after application, and before birds are placed back in the poultry house. Liquid formulations are less likely to expose birds to concentrations of the chemical due to greater dispersal in the litter compared to dry formulations (Moore and Watkins 2012). Aluminum sulfate tends to dry out the litter, and in turkeys the use of aluminum sulfate decreased the incidence of foot pad dermatitis, which is associated with wet litter (Wu and Hocking 2011).

In addition to the phosphorous-fixing properties of aluminum sulfate, litter treated with aluminum sulfate inhibits microbial phosphorous mineralization from organic matter (Warren et al. 2008). Although the literature review did not reveal problems associated with salinity of litter treated with aluminum sulfate, treated litter contains higher levels of soluble NH_4^+, and sulfur; thus, the salinity is likely higher than non-treated litter. However, salt damage to crops at normal agronomic application rates is likely low due to dilution factors (Sims and Luka-McCafferty 2002). Effects on bird health are positive, as ammonia accumulation causes lung irrigation to poultry (Walker and Burns 2000). Pathogen loads in the broiler house are reduced with aluminum sulfate, which combined with lower ammonia concentration in the air causes increased bird weight gain (Shah et al. 2006).

<u>Evaluation Question #9:</u> Discuss and summarize findings on whether the use of the petitioned substance may be harmful to the environment (7 U.S.C. § 6517 (c) (1) (A) (i) and 7 U.S.C. § 6517 (c) (2) (A) (i)).

Proper use of the substance as petitioned would not be harmful to human health or the environment. The NRCS incentivizes the use of aluminum sulfate for its benefits to the environment including reduced ammonia emissions to the atmosphere, and reduced phosphorous pollution to surface waters (Moore et al. 2000). The improbable misuse of aluminum sulfate was discussed in Question #6.

Although the misuse or spilling of aluminum sulfate could have deleterious effects on the soil (See question #6 for more details), in general, using aluminum sulfate on poultry litter does not pose harm to the environment. As described in Questions #4, 7, and 9, aluminum sulfate reacts with components of the litter to reduce volatile nitrogen and phosphorous runoff, both of which must be managed in a myriad of ways to prevent environmental contamination. Aluminum sulfate treated litter offers one mechanism to achieve these positive environmental effects.

Evaluation Question #10: Describe and summarize any reported effects upon human health from use of the petitioned substance (7 U.S.C. § 6517 (c) (1) (A) (i), 7 U.S.C. § 6517 (c) (2) (A) (i)) and 7 U.S.C. § 6518 (m) (4)).

Aluminum sulfate reacts with water to form sulfuric acid, which is an irritant. Aluminum sulfate is corrosive to the eyes; skin contact causes a rash and burning feeling, and inhalation causes throat and lung irritation (New Jersey Department of Health 2009). The magnitude of the toxic response to aluminum sulfate is completely dose-dependent, and the substance is permitted as a food additive in small quantities. Minor ingestion of dilute solutions causes stomach upset, while substantial ingestion can rarely cause hemorrhagic gastritis, circulatory collapse and multi-organ failure (United Kingdom National Poisons Information Service 1996).

Aluminum is a subject of medical contention with suspected links to Alzheimer's disease. Implications of a link between Alzheimer's disease and aluminum have been made for approximately 40 years. The current large body of research has not concluded specific roles of aluminum in contributing to Alzheimer's disease, but also has not dismissed aluminum as a non-contributor to the disease (Agency for Toxic Substances and Disease Registry 2008; Exeley 2001). Under FDA regulations, aluminum sulfate is generally recognized as safe (GRAS) as a food additive when used in accordance with good manufacturing or feeding practice (CFR 182.1125(b)).

Although aluminum sulfate has chronic toxicity for human exposure, use of the substance as petitioned should not have negative effects on human health. Use of the substance as petitioned decreases ammonia concentration in the atmosphere of poultry houses, which has a positive impact on both health of the birds and health of workers (Moore et al., 2000).

Evaluation Question #11: Describe all natural (non-synthetic) substances or products which may be used in place of a petitioned substance (7 U.S.C. § 6517 (c) (1) (A) (ii)). Provide a list of allowed substances that may be used in place of the petitioned substance (7 U.S.C. § 6518 (m) (6)).

Clinoptilolite is a naturally-occurring aluminosilicate zeolite which can absorb ammonia, reducing volatilization to the atmosphere. The literature contains results of mixed efficacy for this material, with some reports of decreased ammonia in broiler house air, and other reports of increased atmospheric ammonia (Amon et al. 1997; Karamanlis et al. 2008; Shah 2006).

Agricultural lime can be applied to litter between flocks to increase litter pH, chemically inducing volatilization of large quantities of ammonia. The volatized ammonia can then be removed by ventilation before birds are placed back in the poultry house. Removal of ammonia from litter in between flocks reduces ammonia concentration in air for the subsequent grow-out, but does not mitigate ammonia production during the grow-out compared to acidification products. Although lime does not decrease total atmospheric ammonia pollution like aluminum sulfate, phosphorous in the litter is stabilized by complexation with calcium at high pH to reduce eutrophication of natural water bodies after land application of the litter (Shah 2006).

Yucca saponins as a diet supplement can inhibit ammonia production from manure by reducing urease activity; however, reduction of ammonia is not as effective as aluminum sulfate, and this method does not lead to phosphorous stabilization in the litter (Chepete et al. 2012). Powdered root of *Yucca schidigera* is available commercially for this purpose as BioSol-YS-30S Pure Soluble Yucca Powder, and BioSol YS-50LC Pure Yucca Schidigera Liquid Concentrate (Ultra BioLogics Inc. 2004).

Evaluation Question #12: Describe any alternative practices that would make the use of the petitioned substance unnecessary (7 U.S.C. § 6518 (m) (6)).

The petition indicates that the ventilation speed and duration required to remove ammonia from confined spaces chambers excessively chills chicks <10 days old, and therefore aluminum sulfate is a necessary input to combat ammonia accumulation. A common cultural practice of partial-house brooding (installing curtains to create a smaller confined brooder area) reduces the heat input requirements, but also decreases air volume thus concentrating ammonia vapors. Adding capacity to the heating system could allow ventilation without chilling chicks, making aluminum sulfate application unnecessary, but at great equipment and energy expense and with associated greenhouse gas emissions. The use of low-hung infrared brooders can mitigate thermal loss from ventilation if the partial-house brooding strategy is abandoned in favor of diluting ammonia concentrations in the air (Lacy 2000). Additionally, low-hung infrared brooders tend to dry out litter, disfavoring ammonia production (Wu and Hocking 2011; Walker and Burns 2000). Moisture management of litter is also critical for reducing ammonia volatilization. Controlling roof leaks and water line leaks, and using waterers that avoid spillage also mitigates ammonia concentrations (Payne and Zhang 2012).

More frequent changes of fresh litter would reduce the need for aluminum sulfate application but would have no impact on stabilizing phosphorous. Typically aluminum sulfate is not applied to fresh litter, but rather to litter that is being re-used for subsequent flocks of birds. With aluminum sulfate for long-term ammonia management, litter is reportedly reused for up to 35 flocks before it is changed (Sims and Luka-McCafferty 2002). Assuming decreased ventilation is required to ensure warm temperatures for brooding chicks, starting with fresh litter containing essentially zero ammonia, combined with the comparatively low waste production of chicks (and therefore nitrogen) compared to larger birds could mitigate the necessity for aluminum sulfate.

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