Enhancement of Municipal Wastewater Biosolids Drying through Interfacial Energy Modifying Amendments to Promote Uniform Agglomeration – Bench Scale Testing

by

Sarah Stine

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Abstract

Biosolids Drying is the process of producing a fertilizer product for beneficial reuse from solids produced during municipal wastewater treatment. The drying of biosolids involves the evaporation of water to stabilize the material and produce a product for beneficial use. Thermal energy needs to be transferred to the biosolids to evaporate the water and heat the solids. Energy can be provided by combustion of fuels, re-use of waste heat or solar radiation (WEF, 2014). The most common technology for biosolids drying in the United States utilizes rotary drum dryers. In these systems, fines and crushed oversized pellets produced during the drying system are mixed with dewatered biosolids upstream of the dryer to create a 55% - 65% dry biosolid in the form of pellets. Reducing the percentage of fines generated during the drying process can potentially reduce the amount of energy required for drying.

In earlier research completed by Zhang (2018) it was shown that energy modifying amendments, specifically cationic polyelectrolytes, can reduce the zeta potential of biosolids in solution and possibly promote aggregation of the fines. One of the tested amendments, polydiallyldimethylammonium chloride (PDADMAC), was also shown to increase the particle size of the biosolids in solution. In this work, a bench scale drying system was designed and developed to apply the polyelectrolyte amendments to biosolids during the mixing phase, and to gauge the impact on the pellet size distribution and the percentage of fines generated after drying. It was shown that PDADMAC, which is a high charge density cationic polyelectrolyte, had a measurable, though inconsistent, impact on pellet size when applied during the mixing phase. This work also highlights the varying characteristics of biosolids and the recycled biosolids produced during the drying process. Both PDADMAC, and polyallyamine, another cationic polyelectrolyte, when applied to biosolids during the mixing phase limited the increase in fines production as the mixing time was increased prior to drying.

Introduction

Wastewater treatment is one of the most important elements of protecting the environment, and this is especially true as population densities increase. The main goal of wastewater treatment is to remove harmful constituents from the water before it is released back into the environment. However an equally important and potentially more challenging aspect of wastewater treatment is solids handling. There are several options for handling solids, including landfilling, incineration and land application. Solids generated during wastewater treatment and treated with the intent of re-use are called biosolids. Land application of biosolids can take several forms based on the treatment of the biosolids prior to application; typically dewatering or drying.

Biosolids Drying is the process of producing fertilizer for beneficial reuse from solids in the municipal wastewater stream. If regulations for pollutant limits and pathogen reduction are met, the dried biosolids can be classified as "Class A" and applied to gardens, agricultural fields and any other place fertilizers are used, without restriction. The biosolids drying process is energy intensive; fundamentally all types of biosolids drying involve the evaporation of water. Thermal energy needs to be transferred to the biosolids both to evaporate the water and heat the solids. Energy can be provided by combustion of fuels, re-use of waste heat or solar radiation. Drying reduces both the weight and volume of biosolids to be processed and disposed. This is particularly important for urban areas where the end product needs to be transported considerable distances to agricultural or open areas.

There are several technologies available for biosolids drying. In the United States, the most prevalent is direct drying using Rotary Drum Dryers. In this drying process, sludge is dewatered to approximately 25% solids, then mixed with already dry recycled product in a mixer called the pugmill. The recycled material fed into the pugmill with the dewatered "cake", is previously dried material considered too large or too fine. The biosolids take the form of pellets in the pugmill and are then transported to the dryer where they are transported through the dryer by hot air. A uniform pellet size is required both to ensure the product does not clump during transportation and in order for the end product fertilizer to be marketable both as an individual fertilizer and for blending with other fertilizers.

Drying biosolids generated during the municipal wastewater treatment process to generate Class A fertilizer has gained traction and become more prominent over the past 20 years. Notably the Great Lakes Water Authority serving the metro Detroit area recently built the largest Biosolids Drying Facility in North America; the facility was put into use in 2015. As more communities around the country update their sludge handling processes, Biosolids Drying is an option that will be considered and often implemented. Any energy requirement reduction in the drying process can have an impact on the overall energy required to handle biosolids in a beneficial and environmentally friendly way. Reducing the amount of dried product recycled to the pugmill could potentially decrease the overall energy required for drying and allow for smaller equipment sizing during design and construction. The goal of this study was to determine an amendment which can be added during the pugmill mixing stage of the drying process to promote uniform agglomeration of biosolids pellets and in turn, reduce the percentage of fines produced when drying biosolids for fertilizer production.

Background

Wastewater Solids

The solids content in sludge produced during municipal wastewater treatment typically contains from 2% to 8% solids. This sludge can consist of primary sludge from primary treatment settling and waste activated sludge from secondary biological treatment. The solids in wastewater sludge can vary significantly depending on the influent to the treatment facility and the types of treatment technologies utilized. The main components making up the solids include microorganisms, organic fibers, inorganic colloids and extracellular polymeric substances (EPS). Biologically activated, or "waste activated", sludge is typically composed of 40 % - 60% EPS (Wei *et al*. 2018 and Christensen *et al*. 2015). The extracellular polymeric substances are made up of proteins, humic substances, polysaccharides, nucleic acids and lipids. EPS components of sludge and specifically sludge flocs are negatively charged; Raynaud *et al.* (2012) found the charge density of the EPS in waste activated sludge to be -1.06 meq/gram. Other studies have found the charge density to be in the range of -0.2 to -1.0 meq/gram (Christensen *et al.*, 2015).

Regulating Land Application of Biosolids

Land application of biosolids in the United States is regulated by the EPA Part 503 Biosolids Rule. The Part 503 rule has differing levels of maximum pollutant concentrations and pathogen reduction requirements based on the intended end use of the biosolids (EPA, 1994). Exceptional Quality Biosolids, or EQ Biosolids, are defined by the EPA are those applied in bulk to agricultural fields and sold as bagged fertilizer to the public. In order to be characterized as Exceptional Quality, biosolids must meet low-pollutant concentration requirements and Class A pathogen reduction (EPA, 1994). Class B biosolids are typically dewatered sludge which can be land applied with greater restrictions, typically to fields not currently producing food for human consumption.

Subpart D of the EPA's part 503 rule addresses pathogen reduction and reduction of vector attraction. Pathogens are defined as organisms, including bacteria, viruses and parasites that can cause disease. Vectors are insects, rodents and other organisms that can spread disease by carrying pathogens. Based on the levels of pathogen and vector attraction reduction a

biosolid will be classified by the EPA as Class A or Class B. Pathogen reduction requirements can be met by either using known/established technologies and meeting baseline pathogen testing requirements, or by using unknown technologies and meeting more stringent pathogen testing requirements. Known technologies which typically achieve the pathogen and vector attraction requirements in order to be classified as Class A include alkaline stabilization, composting and heat drying.

All biosolids, regardless of treatment technology used must meet the following pathogen reduction requirement in order to be classified as Class A (EPA, 1994):

1) The density of fecal coliform in the biosolids must be less than 1,000 most probable numbers per gram total solids; or

2) The density of Salmonella sp. bacteria in the biosolids must be less than 3 most probable numbers per 4 grams of total solids.

The Code of Massachusetts Regulations Title 310 CMR 32 regulates the production, sale or distribution and use of biosolids for land application in the state of Massachusetts. Title 310 is a compilation of regulations from the Department of Environmental Protection. Part 32 of 310 CMR is titled, "Land application of sludge and septage". For land application of biosolids the MassDEP classifies the sludge or biosolids as Type I, II or III. Biosolids classified as Type I may be sold and distributed without any further approval from the Mass DEP; they may be used for growing any type of vegetation and can be "in direct contact with the edible portion of the crop" (MassDEP, 2016).

Any sludge or septage to be land applied in Massachusetts must be stabilized; in order for a sludge or biosolid to be deemed Type I it must be stabilized by one of the methods listed in column 2 of Table 1 and also meet 310 CMR 32 pollutant concentration requirements. Several of the MassDEP pollutant requirements are more stringent than the EPA Part 503 requirements, see Table 2. One of the stabilization methods identified by the MassDEP for Type I generation is heat drying, which is defined by 310 CMR 32 (similarly to the EPA's part 503 rule) as, *"A process in which a dewatered sludge cake is dried by direct or indirect contact with hot gases, and the moisture content is reduced to 10% or lower. Sludge particles shall reach temperatures well in excess of 80°C, or the wet bulb temperature of the gas stream in contact with the sludge at the point where it leaves the dryer shall be in excess of 80°C."* (MassDEP, 2016)

Table 1 - Massachusetts DEP Methods for Pathogen Reduction

Stabilization Methods Types II and III Biosolids	Stabilization Methods Type I Biosolids
Aerobic Digestion	High Temperature Composting
Air Drying	**Heat Drying**
Anaerobic Digestion	Heat Treatment
Low Temperature Composting	Thermophilic Aerobic Digestion
Lime Stabilization	Electron Radiation *
	Gamma Ray Irradiation *
	Pasteurization *

Adapted from 310 CMR 32, Appendix A.
Notes: Type II and III are differentiated by pollutant concentrations.
*Electron Radiation, Gamma Ray Irradiation and Pasteurization also require a treatment process from column 1.

Table 2 - Pollutant Limits and average concentrations in Bay State Fertilizer

Pollutant / Metal	Considered a Plant Nutrient	EPA Part 503 Limit for EQ Biosolids (mg/kg)	Massachusetts DEP 310 CMR 32 (mg/kg)	Bay State Fertilizer Average *June 2008* (mg/kg)
Arsenic	No	41	Not Regulated	Not Detected
Cadmium	No	39	14	2
Chromium	No	1,200	1,000	49
Copper	Yes	1,500	1,000	612
Lead	No	300	300	171
Mercury	No	17	10	2
Molybdenum	Yes	75	40	18
Nickel	Yes	420	200	25
Selenium	No	36	Not Regulated	4
Zinc	Yes	2,800	2,500	1,130

Adapted in part from the EPA's Guide to Part 503 Rule, along with MassDEP 310 CMR 32, MWRA's Bay State Fertilizer Marketing Brochure, and University of Missouri's Soils, Plant Nutrition and Nutrient Management Gardener Manual

The Massachusetts Water Resource Authority (MWRA) processes wastewater from homes and businesses in Boston and 42 other surrounding communities, as far from the city as Clinton and Framingham. The MWRA's wastewater treatment plant is located at Deer Island in Winthop, MA. The WWTP processes wastewater using primary treatment, secondary treatment, sludge digestion and disinfection. Prior to 1991 digested sludge and scum were recombined with chlorinated effluent at the end of the treatment process and discharged into Boston Harbor with the outgoing tide. In 1988, President Bush Sr. stated that Boston harbor was the dirtiest harbor in America (Allen, 2013). It was a political attack of the Democratic Nominee, Michael Dukakis, but regardless of politics, something needed to be done to save Boston Harbor.

Biosolids drying for beneficial reuse had been happening for decades in other parts of the country, most notably Milorganite Fertilizer has been produced from digested sludge by the Milwaukee Metropolitan Sewerage District since 1926 (MMSD, 2007). In 1988, the MWRA made the decision to build a Biosolids Drying Facility (BDF) in Quincy, Massachusetts. New England Fertilizer Company was awarded the contract by the MWRA to design, build and operate the new facility. In 1991, the Quincy BDF was opened and began producing fertilizer. Sludge is transported seven miles from Deer Island to the Biosolids Dryer Facility by a pipeline running under Boston Harbor. The majority of the fertilizer is distributed in bulk, by trucks, to farms and golf courses around the state. A small portion of the fertilizer is bagged and sold as Bay State Fertilizer. Bags can be purchased at the front desk of the BDF or at local garden centers (MWRA, 2018). The fertilizer produced in Quincy meets all EPA and MassDEP requirements for Exceptional Quality and Type I biosolids; see Table 2 with pollutant levels in the Bay State Fertilizer.

It is also worth noting that in 2000 a new outfall was put into use, transporting the disinfected effluent from Deer Island, 9.5 miles into the deep waters of the Massachusetts Bay, rather than Boston Harbor where it was previously discharged. Initial dilution at the new outfall is about 100:1, the Boston Harbor outfall had only been achieving initial dilution of 14:1 (MWRA, 2018).

Processes for Heat Drying Biosolids

There are several heat drying technologies available to convert sludge produced during wastewater treatment into a dry product for beneficial reuse. Drying methods are characterized by their primary method of heat transfer. Methods include convection, conduction and radiation (WEF, 2014). Drying systems are typically called out as "direct drying" (convection) or "indirect drying" (conduction). In direct drying the biosolids comes into direct contact with hot air and gases which cause evaporation (EPA, 2006). Direct dryers include rotary drum dryers, flash dryers, spray dryers and toroidal dryers. Indirect drying systems keep the biosolids separated from the heating medium by a metal plate, common heating mediums include oil or steam. Indirect drying includes steam dryers, hollow flight dryers and tray dryers.

Rotary drum drying is the most prevalent method of biosolids drying in the United States. There are Biosolids Dryer Facilities at Wastewater Treatment Plants around the country using Rotary Drum Drying. These facilities accept a variety of sludge, including primary sludge, waste activated sludge and a combination of primary and waste activated sludge. In some cases the sludge is digested prior to being sent to the Biosolids Dryer Facilities and in some cases it is not. The MWRA's Biosolids Dryer Facility in Quincy, processes only digested sludge. Great Lakes Water Authority (GLWA) recently constructed the largest Biosolids Dryer Facility in North America in Detroit. The BDF in Detroit processes primary and waste activated sludge in varying ratios, but does not process any digested sludge. GLWA noted in a presentation for the Michigan Water Environment Association that they dealt with challenges processing undigested, unscreened primary sludge due to the fiber contents. GLWA also noted significant seasonal variability in the dried biosolids (Khan, 2016). Per presentations by NEFCO, to the same Michigan Water Environment Association, the type of sludge received by a drying facility can have a significant impact on the drying process and quality of the final product. Primary sludge has the highest variability and can contain extraneous materials, waste activated sludge has the best binding properties but can also be the most prone to odors, and digested sludge has the lowest volatility and produces the best quality pellets (Kyzar, 2016). Figure 1 shows a general overview of the wastewater treatment process and the stages at which sludge is generated.
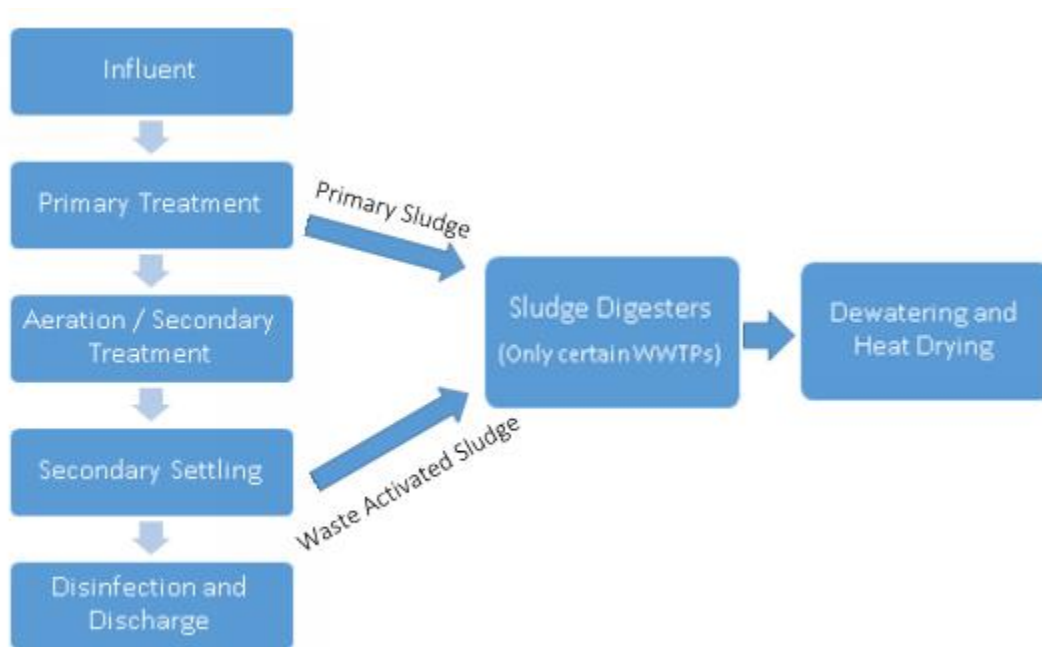
Figure 1 - Wastewater Treatment Process Overview

Biosolids dryer facilities are typically set up with multiple equipment "trains" which allow for flexibility in operations and maintenance. Each train is comprised of a full set of drying equipment and each train can be operated as a standalone system. The MWRA's BDF in Quincy is made up of 6 equipment drying trains and typically processes approximately 160 dry tons per day, 4 – 5 days per week; bringing the yearly average to 106 dry tons per day (MWRA, 2011). GLWA's BDF in Detroit is made up of four dryer trains; the equipment is larger and can process up to 420 dry tons per day (Khan, 2016). The Detroit facility operates 24 hours a day, 365 days a year.

The equipment setup at most Biosolids Dryer Facilities is comparable. In particular the NEFCO operated Quincy and Detroit facilities are very similar; see Figure 2. The process starts with biosolids entering the dryer facilities as sludge. The first step in each train is dewatering, typically with centrifuges. Due to centrifuge sizing there are two centrifuges per dryer train. "Cake", dewatered to 25% - 35% solids (Kyzar, 2016) generated from the centrifuges is mixed with recycled dried fertilizer using a pugmill mixer. Recycled material is made up of dried pellets which were outside of the desired 1-3 mm pellet diameter range (Irujo, 2016). Wet cake

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and dried recycled material are mixed at an approximately 1:1 ratio to generate pellets that are 55% - 65% solids prior to entering the rotary drum dryer (Kyzar, 2016). There are spray headers located within the pugmill which can add water if the percentage of recycled material is too great. The pugmill is where the pellet shape and size distribution of the pellets is determined. The pugmills used in biosolids drying are similar to a twin screw conveyor, however the screw is made up of paddles rather than a continuous smooth screw. This allows the pugmill to transport and mix concurrently.
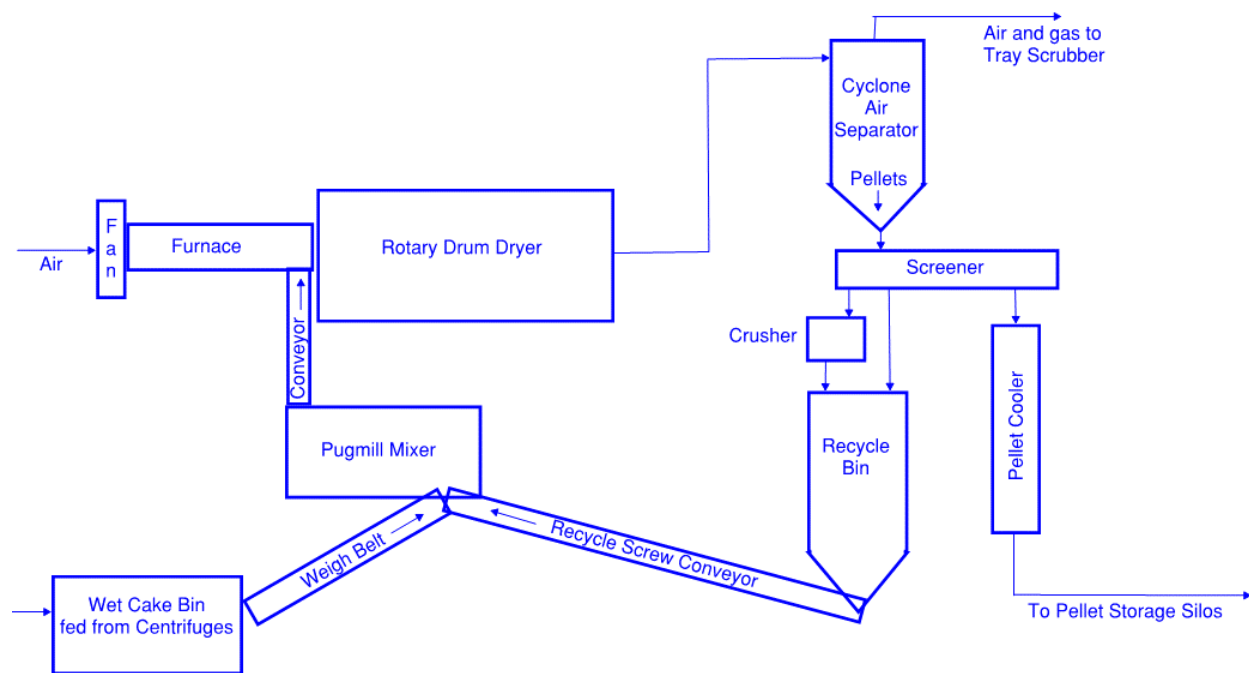
Figure 2 - Biosolids Drying Process Diagram

Note: Based on the drying process at NEFCO's Detroit, MI facility.

While in the rotary drum dryer, evaporation causes pellets created in the pugmill to transition from 55 – 65% solids to 95 – 98% solids. The rotary drum dryers are large rotating drums, with flights to ensure even drying of the biosolids, the dryers in Quincy are single pass, similar to Figure 3. The rotary drums at NEFCO's Detroit facility are a newer triple pass style as shown in Figure 4. Each Dryer in Detroit is 12 feet in diameter and approximately 40 feet long. The pellets formed in the pugmill are transported through the rotating dryer by a large fan. Temperatures used during the drying process are not high enough to cause oxidation of the

organic matter, therefore the organic matter is maintained in the final dry biosolids product (WEF, 2014). After leaving the dryer, pellets are separated from the air and gas in the dryer using a cyclone air separator, the solids are then sorted using a three level pellet screener. Oversized pellets are crushed and then added to the recycle bin, and fines (pellets under 1 mm) are added directly to the recycle bin. Correctly sized pellets are sent through a cooler and on to silos for storage before distribution.

Figure 3 - Single Pass Rotary Drum Dryer

Feeco, https://feeco.com/rotary-dryers/
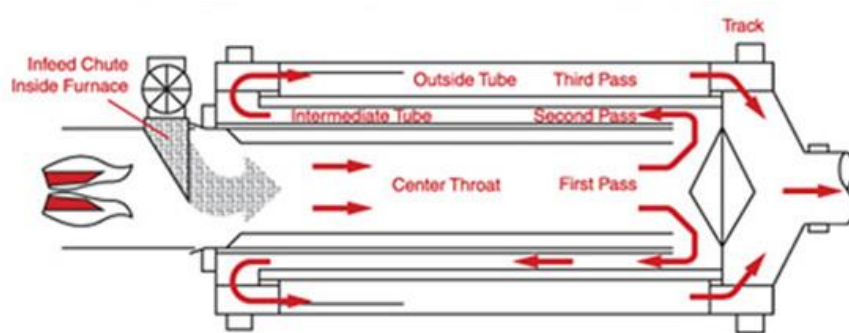

Figure 4 - Newer style Triple Pass Rotary Drum Dryer

KBW Machinery, https://rotarydryer.org/triple-pass-drying

Note: though they are not as clearly shown by the diagram, these dryers do typically have flights to ensure movement.

Drying Energy Requirements

Sludge disposal typically accounts for more than half of the cost and energy associated with the overall treatment of municipal wastewater (Wei *et al*. 2018). Heat drying is more energy intensive than other reuse methods such as composting and Class B land application (EPA, 2006). However, drying typically reduces the volume and weight of biosolids by a ratio of 1:4 compared with dewatered "cake" which could be land applied as Class B biosolids. In major urban areas biosolids often need to be transported significant distances in order to be re-used. The energy required for this transportation is location specific, but can play a major role in a cost/benefit analysis of the energy required for biosolids drying.

At the most basic level all types of biosolids drying bring about the evaporation of water. Thermal energy needs to be transferred to the biosolids both to evaporate the water and heat the solids. In addition to thermal energy required to heat the biosolids, energy is required to operate all of the equipment required to transport, process and handle the biosolids. Thermal energy consumption accounts for the majority of energy use in biosolids drying. Theoretically, evaporation of water requires 970 BTU per pound of water. Drying dewatered cake typically requires 1,400 – 1,700 BTU per pound of water evaporated (EPA, 2006). Energy can be provided by combustion of fossil fuels, re-use of waste heat or solar radiation. Lower temperature dryers, such as belt dryers, are more effectively able to utilize waste heat from existing plant operations than rotary drum dryers (WEF, 2014).

In 2002, New England Fertilizer Company built the second Biosolids Dryer Facility in Massachusetts, at the Greater Lawrence Sanitary District's (GLSD) Wastewater Treatment Plant in North Andover. The facility produces Class A / Type I biosolids fertilizer which is sold to local farms, similar to the MWRA's Quincy facility. A major difference from the plant in Quincy is the fuel source: the GLSD Biosolids Dryer Facility utilizes digester gas produced at the WWTP to fuel the rotary drum dryers. Per GLSD (2018), this saves the district $600,000 per year in operation costs. One of the first biosolids dryer facilities in the country, located at the Milwaukee Metropolitan Sewer District (MMSD), utilizes waste heat from turbine power generation to supplement heat to their rotary drum dryers. The entire treatment plant at MMSD is powered by two turbine generators which were installed in the mid-1970s (MMSD, 2007). Another alternative fuel source is landfill gas; the fuel for the dryers at the Palm Beach County

Solid Waste Authority's Biosolids Dryer Facility, is provided solely by landfill gas, with a natural gas service in place only as a backup (NEFCO, 2018).

Fertilizer Sizing and Blending

One of the ways to increase the value of fertilizer produced in biosolids drying is to blend the fertilizer pellets with other types of commercially produced, typically chemical, fertilizers. Per NEFCO (Irujo, 2016) they have found the ideal pellet size which most fertilizer manufacturers look for, both for fertilizer application and for blending, is 1 – 3 millimeters in diameter. In addition to more uniform blending, uniform pellet size contributes to a more constant fertilizer spreading rate and more consistent absorption by soils (Henderson, 2014).

Until the 1980's, typical fertilizer size ranged between 1 mm and 4 mm in diameter. Since that time, quality expectations for fertilizer have changed to a smaller pellet size range and a somewhat increased average pellet size, typically 2 – 4 mm (Ivell and Nguyen, 2013). The two standard values used in the fertilizer industry to describe size and uniformity are the Size Guide Number (SGN) and the Uniformity Index (UI). These descriptors can be found using the following formulas (Ivell and Nguyen, 2013):

- SGN = d_{50} x 100
    - o d_{50} = medium granular diameter in mm
- UI = d_5 / d_{90} x 100
    - o d_5 = diameter at which 5% of sample by weight is smaller, d_{90} = diameter at which 90% by weight are smaller
    - o A uniformity index of 100 would indicate all pellets are the same size

Trends in the market are moving from typical fertilizers with an SGN of 225 (median diameter of 2.25 mm) to an SGN value closer to 300 (median diameter of 3.0 mm) and a uniformity index as high as 50 or 60. Henderson (2014) stated that a Uniformity Index of 30 would be considered questionable quality, a UI of 40, good quality and a UI of 50 or above excellent quality. A lower UI can result in non-uniform fertilizer spreading and distribution, thus reducing its quality.

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Polyelectrolytes for Coagulation and Dewatering

Polymers are frequently used in water and wastewater treatment for coagulation and flocculation. Polymers have several advantages over tradition coagulants, such as alum. Advantages include lower dose requirements, resulting in less material added to the final volume of sludge and a smaller increase in the ionic load of the treated water. One of the most significant disadvantages of polymers for coagulation is the dosage sensitivity (Bolto and Gregory, 2007). Polymers are commonly used for sludge dewatering; however, depending on the sludge make up, use of polymers can be limited for dewatering due to their sensitivity to pH (Wei *et al.*, 2018). Crittenden (2012) noted that there are often many competing reactions in coagulation systems and reactions sometimes do not proceed as expected.

Polymers can be characterized by their ionic nature, as cationic, anionic or nonionic. Ionic polymers are typically called polyelectrolytes. Polyelectrolytes can be further characterized by their molecular weight (MW) and charge density (CD). Polymers are considered to have a low, medium or high MW as shown in Table 3. The charge density of a polyelectrolyte can be determined by colloid titration and expressed in terms of mole percent of charged groups of milliequivalents / gram. Polyelectrolytes are usually described as having a low, medium or high charge density (Bolto and Gregory, 2007).

Table 3 – Polyelectrolyte Characteristics

Molecular Weight	Charge Density (Mole percentage of ionic groups)
Low $< 10^5$	Low ~ 10%
Medium $10^5 – 10^6$	Medium ~ 25%
High $> 10^6$	High ~ 50% - 100%

(Bolto and Gregory, 2007)

Cationic polyelectrolytes are typically used in sludge dewatering due to the negative charge of the EPS material in the sludge. Polyacrylamide and its derivatives are some of the most commonly used polymers in sludge dewatering (Wei *et al*., 2018). Polyelectrolytes can contribute to multiple mechanisms of flocculation, including particle bridging, charge neutralization, depletion flocculation and electrostatic patch (Dao *et al*., 2016). These

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mechanisms of coagulation are seen in both water treatment coagulation and sludge dewatering operations (Wei *et al.*, 2018 and Bolto and Gregory, 2007).

Charge neutralization and bridging are the most prevalent mechanisms of coagulation in both traditional water treatment and in sludge dewatering, see Figures 5 and 6 (Chen *et al*, 2005). Optimum flocculation occurs when particles have been neutralized, or have a zeta potential of near zero. The zeta potential of a particle describes the electric potential or energy with which a positively or negatively charged particle will move away from a similarly charged particle and toward a particle with an opposite charge. Polyelectrolytes with a high charge density tend to maintain a flat configuration and adsorb to solids in a flat configuration. Due to this tendency, a polyelectrolyte with a high CD will cause coagulation or dewatering almost wholly by charge neutralization and electrostatic patch with very little bridging, see Figures 5 and 7 (Bolto and Gregory, 2007).
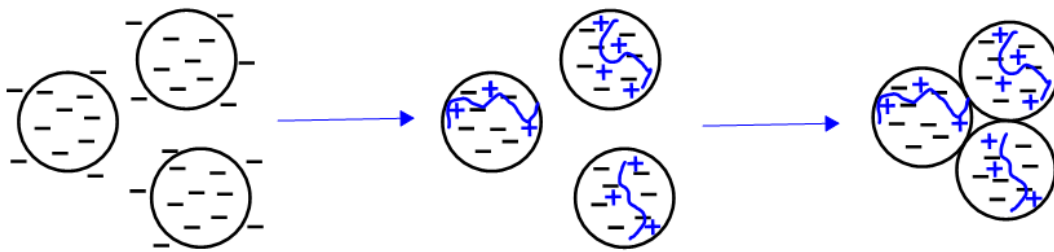

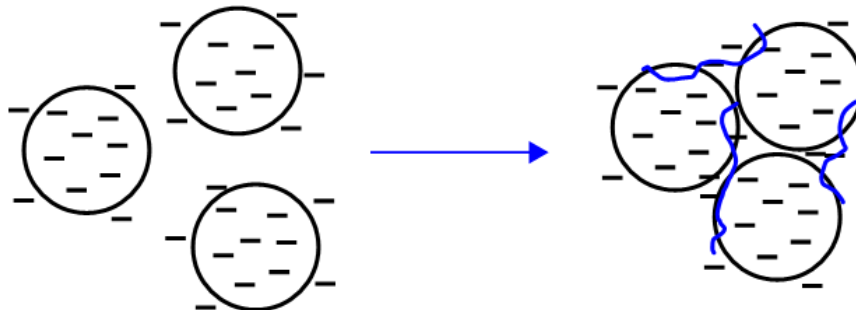**Figure 5 - Charge Neutralization**

Adapted from Dao et al., 2016

Figure 6 - Polymer Bridging
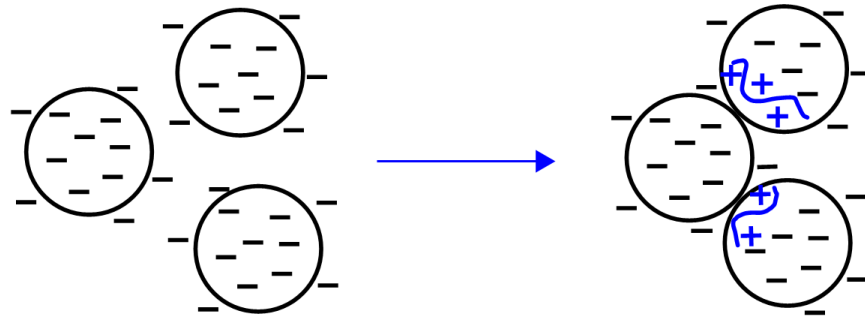
Adapted from Dao et al., 2016

Figure 7 - Electrostatic Patch

Adapted from Dao et al., 2016

Charged particles can be destabilized by oppositely charged ions or polymers. When the amount of polymer that can neutralize the corresponding charge of the particle is adsorbed, the particle's zeta potential will be near zero; meaning the particles will not repel each other (low electrostatic interaction energy) and flocculation or attachment will occur. Particles which are not being repelled by high electrostatic forces will have a higher probability of being acted on by Van der Waals attractive forces and therefore coagulation occurs.

Polymers are typically long-chain molecules. Bridging occurs when one end of the chain adsorbs to a given particle and the other end of the polymer chain adsorbs onto another particle, connecting the two (or more) particles into a floc. Bridging is most common with non-ionic polymers and high molecular weight, low charge density polyelectrolytes (Crittenden, 2012). The electrostatic patch mechanism can occur when a highly charged polyelectrolyte adsorbs onto a weakly charged particle of opposite charge. If the polyelectrolyte cannot be fully neutralized by the one particle it will potentially attract a second charged particle in order to be fully neutralized. The name comes from the fact that a particle would have a small area of positive charge due to the adsorbed polyelectrolyte (Bolto and Gregory, 2007 and Crittenden, 2012).

Previously Completed Research

Helin Zhang (2018) previously completed research at Worcester Polytechnic Institute to gauge the impact of amendments on biosolids in solution. Biosolids at the outlet of the pugmill

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at NEFCO's Quincy facility were collected, dried in an oven, crushed and added to solution. The zeta potential of the biosolids was then evaluated after the addition of five different polyelectrolytes. The polyelectrolytes tested include the following –

- Polydiallyldimethylammonium Chloride (PolyDADMAC or PDADMAC)
    - Cationic; positively charged
    - High charge density; 100 mol%, 6.2 meq/g (Bolto and Gregory, 2007)
    - Medium molecular weight (200,000 – 350,000)
    - Linear formula – $(C_8H_{16}ClN)_n$
    - Density – 1.04 g/mL
    - Solution concentration – 20% by weight
    - Common use – sludge conditioning in water treatment (Crittenden, 2012)
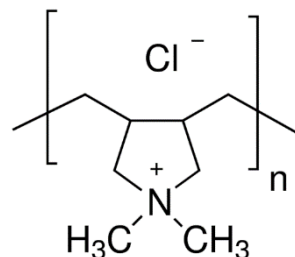
- Polyethylene imine (PEI)
    - Cationic; positively charged
    - Low molecular weight (10,000)
    - Linear formula – $(CH_2CH_2NH)_n$
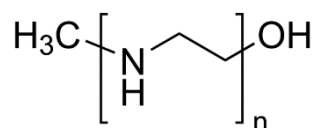
- Polyallyamine (PAM)
    - Cationic; positively charged
    - Medium charge density,
    - Low molecular weight (17,000)
    - Linear formula - $[CH_2CH(CH_2NH_2)]_n$
    - Density – 1.02 g/mL
    - Solution concentration – 20% by weight
    - Common use – primary coagulant in water treatment (Crittenden, 2012)

- Polyethylene oxide
    - Non-ionic
    - Linear formula - $(-CH_2CH_2O-)_n$
    - High molecular weight (1,000,000)
    - Functional group – OH
    - Powder form
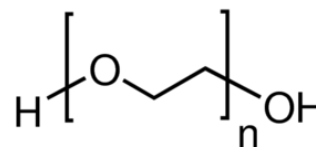
- Polyacrylic acid
    - Anionic
    - Linear formula - $(C_3H_4O_2)_n$

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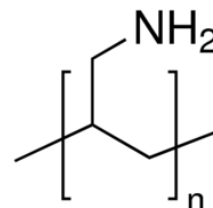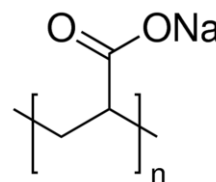The three cationic polyelectrolytes all reduced the zeta potential, while the anionic and non-ionic polyelectrolytes did not have an appreciable impact on the zeta potential. This confirms that the biosolids have a negative surface charge. Zhang (2018) measured the zeta potential of the particles in solution at increasing polyelectrolyte dosages to determine the dose required to bring the zeta potential to zero. The results of her work are shown in Figure 8.
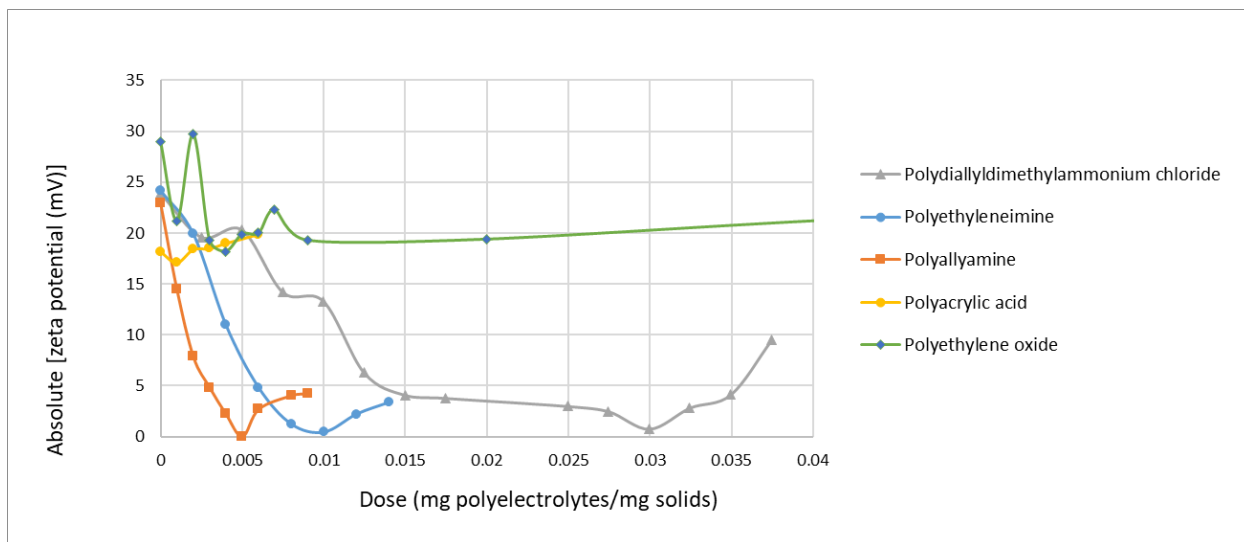
Figure 8 – Impact of Polyelectrolytes on Zeta Potential of Biosolids in Solution
(Zhang, 2018)

In the second phase of research completed by Zhang (2018) the three cationic polyelectrolytes, which successfully lowered the zeta potential, were each added to a solution with biosolids and the resultant particle size was measured. This work showed that of the three cationic polyelectrolytes only PDADMAC had a measurable impact on the particle size; with the addition of PDADMAC the biosolids particle diameter increased by a factor of 4 in 200 minutes. This indicates when PDADMAC was added to a solution with biosolids flocculation was occurring. The results of this work are shown in Figure 9.
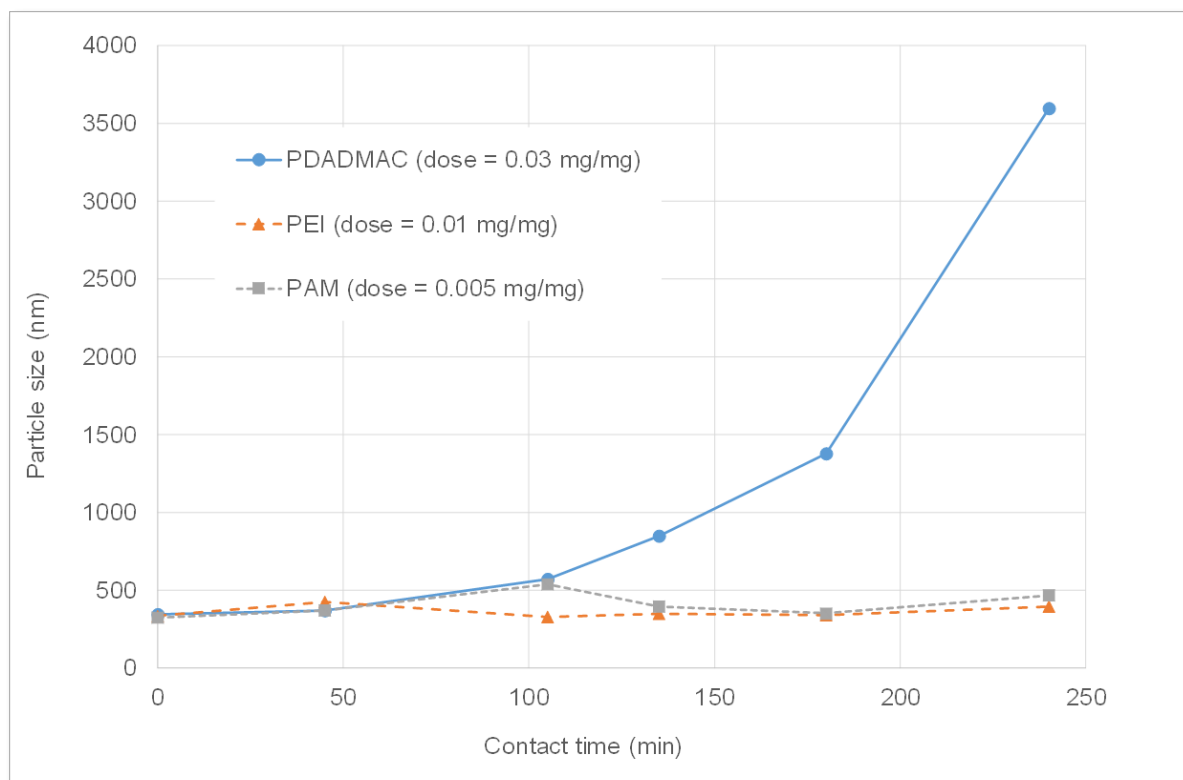
Figure 9 – Impact of Polyelectrolytes on Biosolids Particle Size in Solution *(Zhang, 2018)*

Methods and Materials

The goal of this research was to reduce fines produced in the biosolids drying process, potentially reducing the energy required for biosolids drying. A bench scale system to replicate the main equipment at the "heart" of the Biosolids Dryer Facilities in Quincy, Detroit and many other facilities was developed and utilized for this purpose.

Development of the Bench Scale System

As described previously, biosolids pellets at NEFCO's facilities are created in the pugmill by mixing dewatered biosolids cake and recycled dry biosolids. The pellets are then dried in a rotary drum dryer. Beyond the pugmill and dryer, there are eight to ten other pieces of equipment that make up each dryer train, however with regards to pellet size, dryness and uniformity, the pugmill and dryer are by far the most consequential. To create this "heart" of the biosolids dryer system at a bench scale, a laboratory sized soil mixer was used to replicate the pugmill and a Buchi Rotavapor was used to replicate the dryer.

Samples for the bench scale drying system were collected from the MWRA's Biosolids Drying Facility in Quincy, MA. As previously noted all sludge processed at the Quincy facility has been through the digestion process. Samples were collected in clean plastic containers with screw on lids, with sample sizes ranging from one to three quarts. Samples were collected on four different occasions; between June 2018 and October 2018. All samples were transported back to the WPI laboratory after collection and stored in their sealed containers at $4 \, ^\circ \text{C}$.

Samples collected include the following –

- Dewatered biosolids – "cake"
    - Collected from the weight belt conveyor which transfers the cake from the centrifuge where it is dewatered to the pugmill for mixing with dry biosolids
- Dry biosolids – "recycle"
    - Collected from the recycle bin where product from the screener and crusher are stored before being transferred back to the pugmill
- Mixed biosolids –

- Collected at the end of the pugmill prior to being fed into the dryer

The first phase of laboratory work involved characterizing the samples along with determining mixing parameters and cake to recycle ratios. Small amounts of the cake and mixed biosolids were transferred to small drying tins and the samples were dried in an oven at $100\,^\circ$C for 24 hours. The samples dried were approximately $10 - 20$ grams each. The two samples of dried cake were both within a half percent of 26% solids. Three samples of the mixed material were dried in the oven, the mixed biosolids was determined to be 54% – 56% solids.

The first mixing trials were conducted using a 1:1 volumetric ratio of cake to recycle. 230 mL of recycle material was measured and placed in the mixer, followed by 230 mL of cake. The size of the clumps of cake varied, the cake was broken up slightly by gloved hand if needed to obtain a reasonably accurate volume measurement with limited air space without packing into the beaker for measurement. The standard paddle attachment was used in the mixer (see Figure 10). The following visual observations were noted during mixing: at 30 seconds the material was generally combined, however large pieces of wet material remained; at 2 minutes the color distinction between wet and dry had largely disappeared; and at 3 minutes the material appeared visually to be fully blended. After three minutes of mixing, three small samples were taken from different areas of the mixer, weighed, dried in the oven at $100\,^\circ$C for 24 hours and weighed again to determine both percent solids and mixture uniformity. The samples ranged from 58.3% solids to 60.2% solids. The percent solids found for the samples with the 1:1 ratio was relatively consistent, however it was approximately 4% more dry than the mixed biosolids samples from NEFCO's Quincy Facility.

Figure 10 - Bench Scale Mixer

For the second mixing trial the ratio of cake to recycle was modified to 1.2 cake : 1 recycle. This change was made in order to decrease the percent solids in the mixed product prior to entering the dryer and better replicate the drying process at NEFCO's Quincy Facility. The actual volumes mixed were 275 mL cake and 230 mL recycle. Mixing time was increased to 5 minutes to increase uniformity. Similarly to the first mixing trial three samples were taken from the mixer, weighed and dried for 24 hours in the oven at 100°C. It was found that the samples from this mixing trial ranged from 55.1% solids to 59.5% solids. Based on the information provided from NEFCO that pugmill mix typically ranges from 55% solids to 65% solids and the testing completed on the pugmill sample taken from the Quincy BDF indicating 54% - 56% solids, the volumetric ratio of 1.2 cake : 1 recycle was chosen for bench scale drying moving forward.

The Buchi Rotavapor R-300 consisted of a vacuum pump, cooling coil and tower, hot water bath and solvent/condensate collection container, as shown in Figure 11. The Rotavapor has several adjustable operational parameters including vacuum level, bath temperature and

rotation speed. The Buchi Rotavapor manual recommended several baseline parameters based on the solvent to be evaporated. The "solvent" to be evaporated in this drying work is largely water. The recommended parameters for water are a vacuum of 42 mBar and a bath temperature of 50°C. The vacuum is determined by the pressure required to boil water at a temperature 20°C below the bath temperature. Because the solids being dried have less contact, and therefore less surface area for heat transfer than a liquid, the initial drying trial was conducted at a pressure vacuum of 42 mBar, a bath temperature of 60°C and 20 rotations per minute. A volume of approximately 250 mL was dried in the Rotavapor for each trial. This allowed the material to largely be located in the portion of the vessel in contact with the heating bath (see Figure 11).

Figure 11 - Buchi Rotavapor used as bench-scale dryer.
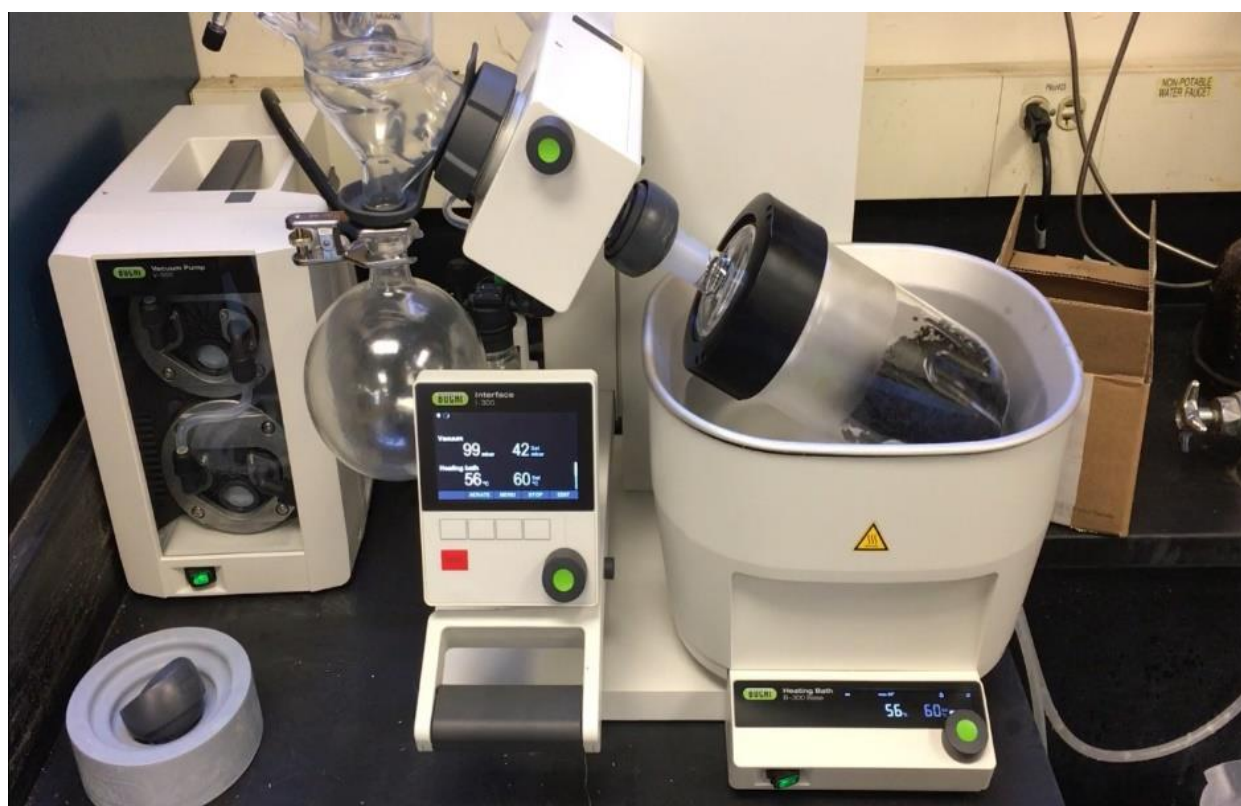
Initial drying trials were run with biosolids without amendment to determine percent solids after increasing times in the dryer. With the initial settings it took 100 minutes in the Rotavapor for the mixed biosolids to reach over 90% solids. The goal for percentage solids after drying is 95% - 98% in order to replicate the pellets dried in a typical Rotary Drum Dryer. For

the second round of drying trials, the bath temperature was increased to 95°C, the mixing was increased to 40 RPM, and the vacuum was unchanged. The mixing speed was increased to create additional agitation to more accurately duplicate a drum dryer. The second drying trials at 95°C were able to achieve over 95% solids content within 60 minutes, (see Figure 13). A temperature of 95°C, a vacuum of 42 mBar, a speed of 40 RPM and a drying time of 60 minutes was used for drying trials moving forward.
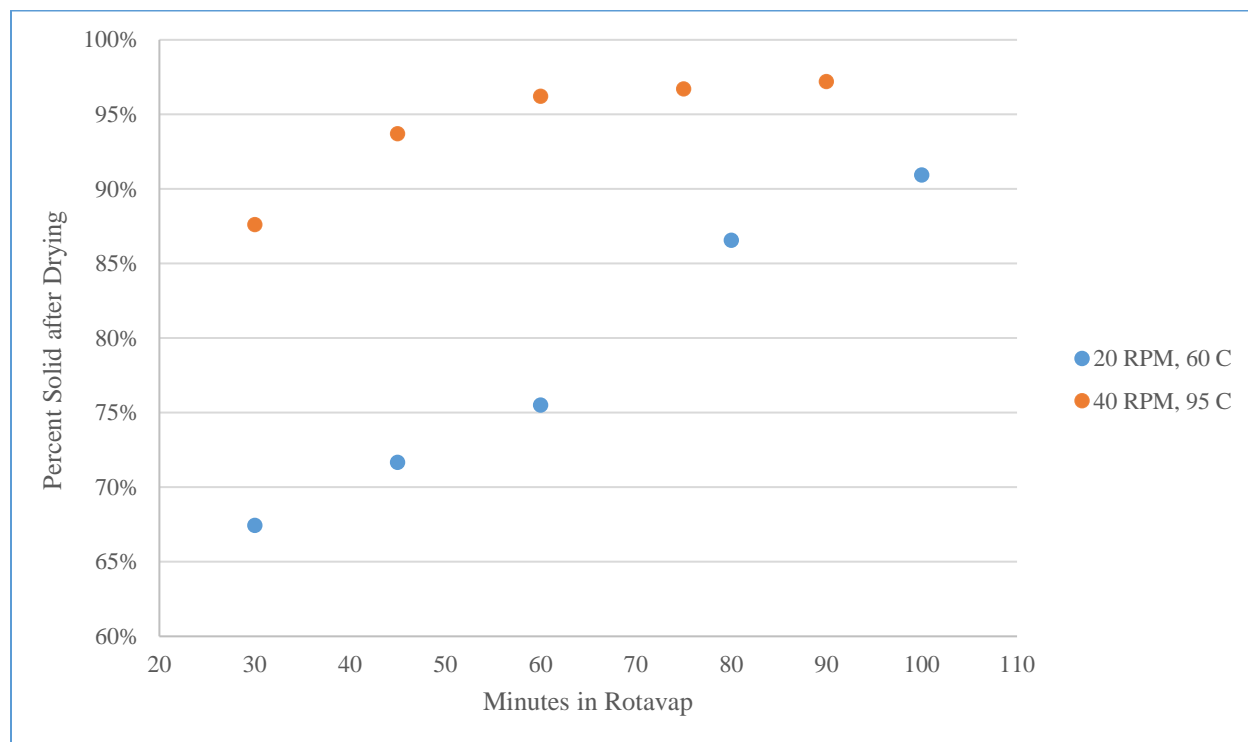
Figure 12 - Drying Time in Rotavapor vs Percent Solids after Drying

After the mixing ratio and rotavapor drying parameters were established, subsequent drying trials began. The variables for this study are amendment, amendment dosage and mixing time. Another unintentional variability is the time of year when the biosolids samples were collected. Amendments and amendment dosage rates were chosen based on the previously discussed work by Zhang, 2018, on this topic. During development of the bench scale a period of 5 minutes was found produce a uniform mixed product. Pellet size distribution was evaluated for a mixing time of 5 minutes and 10 minutes in this study to evaluate the impact of increased mixing time.

Due to equipment sizing, the amount of biosolids initially placed in the rotavapor for drying was half of that created during the mixing process. For the initial drying trials with each amendment, after mixing the cake and recycle for 5 minutes, the amount to be dried was removed, and the remaining mixture was then mixed for another 5 minutes and dried in the rotavapor separately. The impact of the additional mixing time on pellet size distribution and fines generation was evaluated without amendment and for each amendment.

Sieve analysis was completed for the dried samples to provide a pellet size distribution. 8" USA Standard Test Sieves, manufactured by Hogentogler and conforming to ASTM E-11 (standard specifications for test sieves) were used, see Figure 13 and Table 4. All sieves were washed before and after use with warm water and detergent, using a soft bristled brush to clean any material caught in the mesh openings. After drying, the fully dried sample was weighed, the sample was then passed through the set of sieves (shaking manually for 60 seconds). The dried pellets left on each sieve were weighed and recorded. This information was used to create the pellet size distributions and calculate the average pellet diameter. The average pellet diameter was approximated by multiplying the percent by mass solids of each size category by the average diameter for that mesh size, see Table 4.

Figure 13 - Sieves for Pellet Size Analysis

Table 4 - Sieve Size Gradations

Mesh Size	Diameter Range	Average Diameter for Mesh Size
4 mesh	Greater than 4.76 mm	Assumed 5 mm
6 mesh	3.36 - 4.76 mm	4.06 mm
8 mesh	2.38 - 3.36 mm	2.87 mm
16 mesh	1.19 - 2.38 mm	1.78 mm
28 mesh	0.62 - 1.19 mm	0.91 mm
Fines	Less than 0.62 mm	Assumed: 0.30 mm

Drying with Amendments

As discussed in the Background, in previous research Zhang (2018) found that polydiallyldimethylammonium chloride (PDADMAC) reduced the zeta potential and had a significant impact on the particle size of biosolids in solution. This amendment was further evaluated in this research. Another cationic polyelectrolyte amendment, polyallyamine (PAM), was chosen for the second round of drying trials with amendment.

Polyelectrolytes were applied in solution to the biosolids in the mixer, during active mixing. The amount of polyelectrolyte solution applied was determined by the amount required to reduce the zeta potential as found in previous research. The amount of biosolids by mass in the mixer was found by measuring the biosolids cake and recycle before they were placed in the mixer. The mass of cake was reduced by 74% in the polyelectrolyte dosage calculations to account for the water that constitutes the majority of the 26% solids dewatered cake. The mass of the recycle was reduced by 2% based on the assumption that the dry recycle product is approximately 98% solids. PDADMAC was procured as a 20% by weight solution from Sigma-Aldrich (CAS number 26062-79-3) and PAM was procured as a 15% by weight solution from Polysciences, Inc. (CAS number 30551-89-4).

The polyelectrolytes were applied to the mixture using a small spray bottle. The initial dosage application was the dosage found by Zhang, 2018 to be effective at lowering the zeta potential. The amendments were applied starting at time 1:00 of the mixing period and

concluding by time 2:00. This ensured 3 minutes of mixing time after amendment addition with the lowest mixing time of 5 minutes. 3 minutes of mixing time was found during bench scale development to produce a fully mixed product. Spray application was intended to replicate the spray headers found in pugmills at Biosolids Dryer Facilities which are currently used for water application if needed.

After the initial dose of polyelectrolyte (0.03 mg/mg PDADMAC) was applied and the drying trial completed, the dose was determined to have had too significant of an impact (90% of pellets were oversized). The dose was then reduced to evaluate at varying application rates. The dose was cut by 50% to 0.015 mg PDADMAC / mg biosolids and the trial was repeated with a 5 minute mixing time and 10 minute mixing time. Each mixture sample was transferred to the rotavapor vessel and dried for 60 minutes at a temperature of $95\,^\circ C$, a vacuum of 42 mBar and a speed of 40 RPM. A sieve analysis was completed to find the size distribution by weight for each dried sample. After the 0.015 mg/mg trial the PDADMAC dose was reduced further to evaluate impact of the amendment. A full list of trials can be found in appendix #1.

Results and Discussion

The goal of this study was to determine an amendment which can be added during the pugmill mixing stage of the drying process to promote uniform agglomeration of biosolids pellets and reduce the percentage of fines produced when drying biosolids. Testing determined that PDADMAC, a cationic polyelectrolyte, had a measurable however inconsistent impact on the pellet size distribution. Impact of mixing time on biosolids pellet size distribution was evaluated with and without amendments, showing that increased mixing time reduces the average pellet diameter. Another unexpected finding in this work was the variability in the characteristics of the recycle material collected from NEFCO's Dryer Facility in Quincy.

Drying with Amendments

The first amendment to be applied to the biosolids in bench scale drying was PDADMAC. The first step was to determine if the amendment had an impact on pellet size distribution, the second step was to determine an ideal dosage. All of the drying trials with PDADMAC at varying dosages shown in Figure 14 were run with samples from the same sample batch collected from the Quincy BDF in July 2018.

The initial dosage of PDADMAC applied to the biosolids during the mixing stage was 0.03 mg PDADMAC / mg biosolids. This is the dose that was found to lower the absolute zeta potential to near zero which was also the dose used by Zhang (2018) for the particle aggregation (size) tests. In this research, the initial dose of PDADMAC had a major impact on the pellet size. The size increased to approximately 25 mm in diameter, which was significantly larger than the typical 1 mm – 3 mm or 2 mm – 4 mm ranges desired in fertilizer production (see Figures 14 and 15). During mixing it was noted from visual observations that after spray application of PDADMAC the biosolids lost the granular pellet form and took on a form similar to a mass of cookie dough. Trials were then run with PDADMAC doses of 0.015 mg/mg and 0.0087 mg/mg. During these PDADMAC dosage trials, the 0.015 mg/mg dose was found to increase the overall pellet size from an average diameter of 2 mm to an average diameter of 3 mm. In addition, the mass of fines decreased from 7% to 1%, while the oversized pellets increased from 1% to 8%.
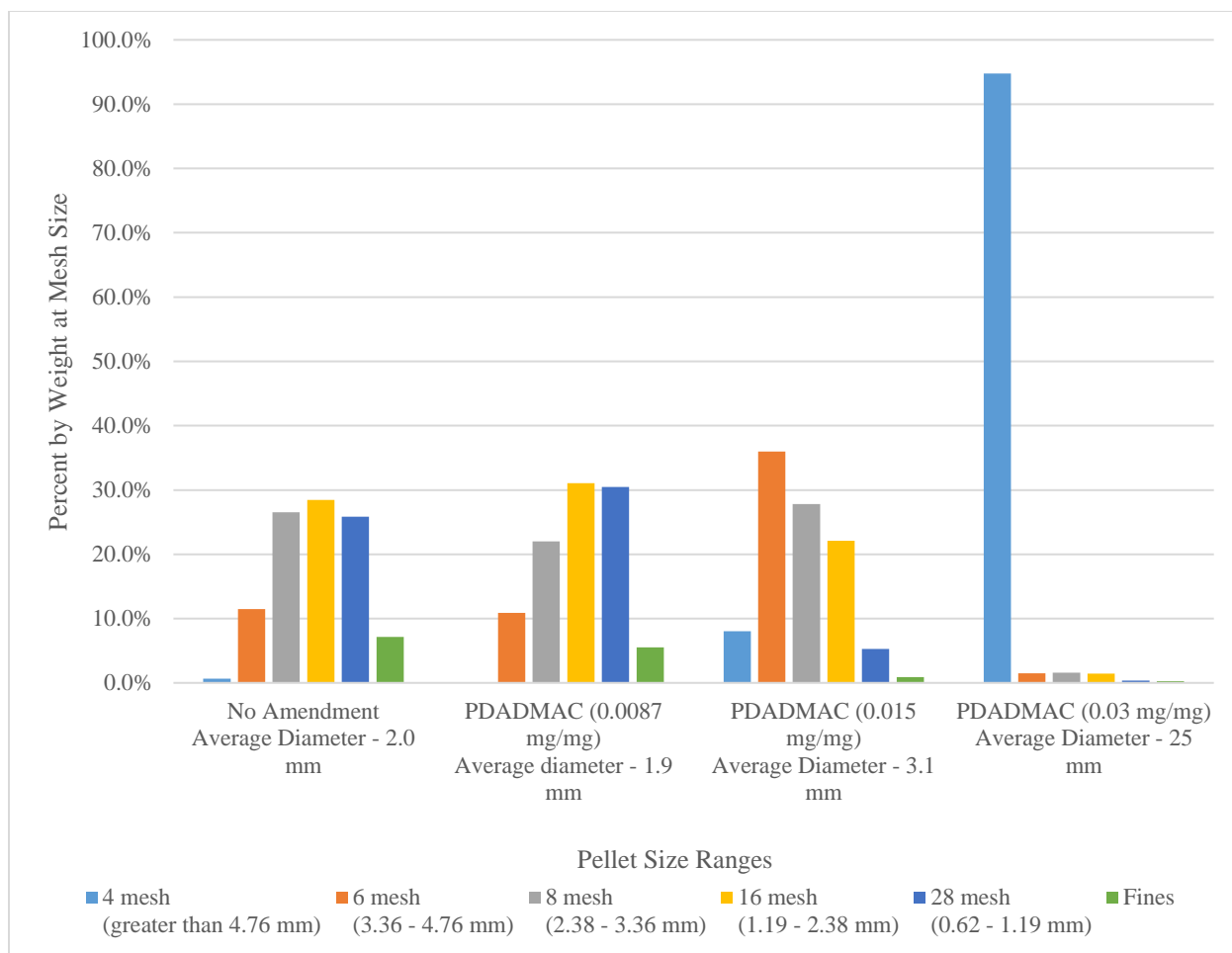
Figure 14 – PDADMAC Amendment Dosage Trials; Pellet Size Distribution

All drying trials displayed in this figure were single trials completed with samples collected from NEFCO's Quincy Facility in July 2018.

Figure 15 - Photos of Pellets Dried with PDADMAC at Varying Concentrations

Based on the trials run with samples collect in July, a dose of 0.015 mg PDADMAC / mg biosolids was chosen as the dose for further testing. Two additional drying trial runs were conducted with a dose of 0.015 mg/mg, with samples collected in both August and October. In drying trials with August 2018 samples, PDADMAC reduced the percentage of fines from 3.7% to 0.4%, however 70% of the pellets created were oversized, greater than 5 mm in diameter. This potentially indicates the dosage of polyelectrolyte was too high. In the final drying trials with samples from October, the 0.015 mg PDADMAC/ mg amendment dose had very little impact on the pellet size distribution. The pellet size distribution very closely matched the size with no amendment from the same samples. This points to variability in the biosolids collected and polyelectrolyte dose sensitivity, see Figure 16. A full list of trials can be found in appendix #1.
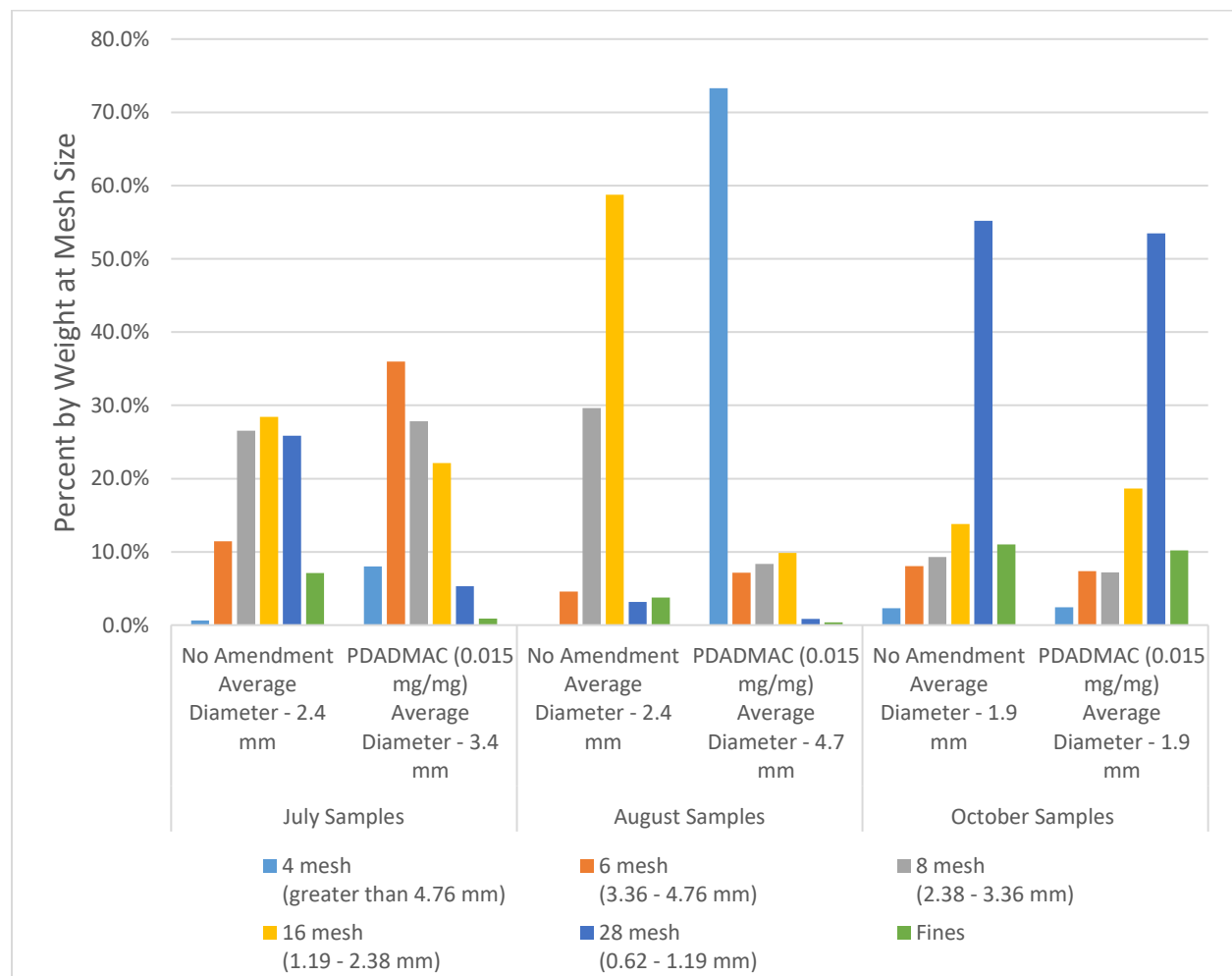
Figure 16 - No Amendment vs PDADMAC 0.015 mg/mg – July, August, October

Polyallyamine (PAM) was another cationic amendment applied to the biosolids during mixing. A dosage of 0.005 mg PAM / mg biosolids was previously found by Zhang (2018) to reduce the zeta potential to near zero. PAM was tested on the biosolids samples collected in August and October. Though the unamended samples from different sample collection dates resulted in different size distributions, the change caused by application of PAM in both trials was minimal, see Figure 19.

Figure 17 - PAM Amendment Trials

Figure 18 - Photos of Pellets Dried with PAM

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Mixing time

To test the impact of mixing time on biosolids, the particle size distributions were measured at two different mixing times for each amendment: 5 minutes and 10 minutes. Compared to the un-amended samples, the amount of fines increased less with PDADMAC and PAM amendments and the oversized particles decreased with increased mixing time. The biosolids samples for the different polyelectrolyte and the no amendment trials were collected on multiple visits to NEF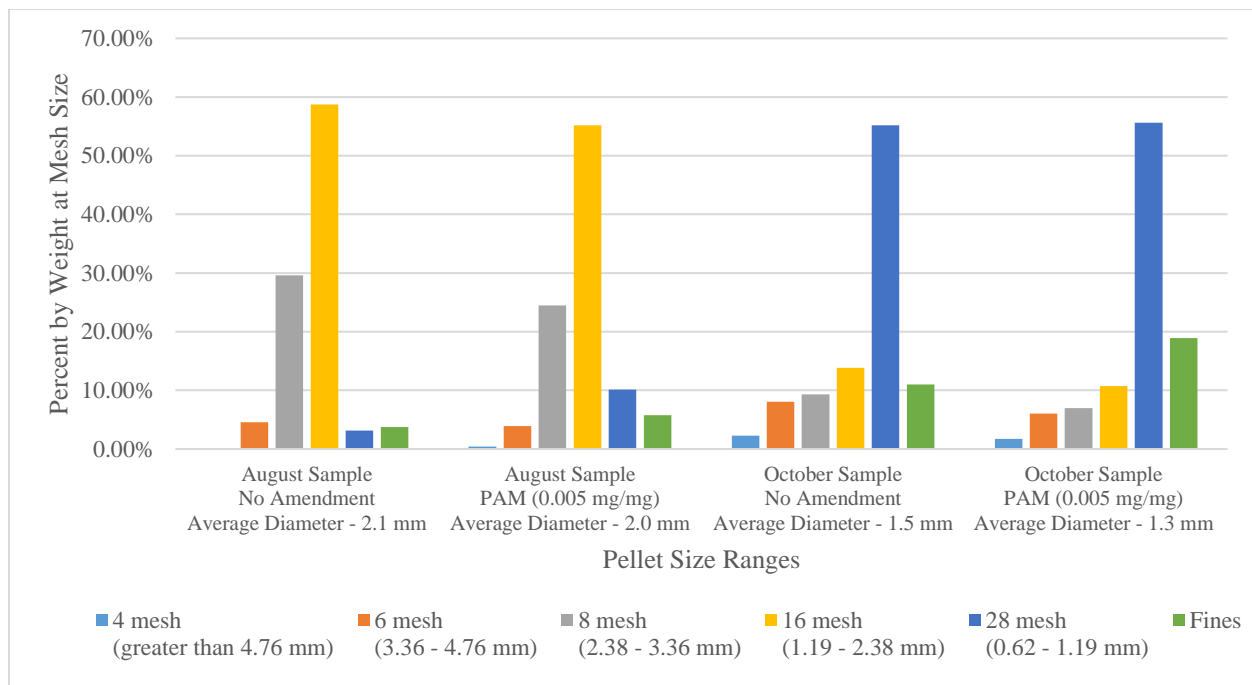CO in Quincy and had varying characteristics; however, each set of 5 minute and 10 minute mix trials inTable 5 were run with material from biosolids samples collected the same day. The percent increase in fines from 5 minutes of mixing time to 10 minutes of mixing time was significantly less for all drying trials in which polyelectrolytes were added during the second minute of mixing.

Table 5 - Percentage of Fines by Mixing Time, amended and unamended.

Drying Trials	5 Minutes Mixing Percent Fines by Weight	10 Minutes Mixing Percent Fines by Weight	Percent Increase in Fines
No Amendment	2.1%	7.1%	238.1%
No Amendment – Second Trial	1.1%	3.8%	241.8%
PDADMAC (0.0087 mg/mg)	3.7%	5.5%	48.6%
PDADMAC (0.015 mg/mg)	0.6%	0.9%	50.0%
PAM (0.005 mg/mg)	3.9%	5.8%	48.7%

Single trial of each row

The Size Guide Number (SGN) and the Uniformity Index (UI) are the fertilizer industry standard descriptors for fertilizer size and uniformity. These values were calculated for the samples with no amendment and for samples collected in July 2018 amended with PDADMAC at a dose of 0.015 mg/mg. The Uniformity Index indicates samples that would be considered poor in the industry, with a UI below 50. However in a biosolids dryer facility the dry product would be screened and sorted with the oversized, and the undersized material sent to the recycle bin. In this case, these numbers simply show the impact of the PDADMAC on the pellet size distribution.

Table 6 - Uniformity Index and Size Guide Number

	No Amendment	PDADMAC (0.015 mg/mg)
UI	22.9	30.0
SGN	207	310

Sample Variability

One of the key findings of this work was the significant variability in the results from samples collected from the Biosolids Dryer Facility in Quincy at different times of the year. Initial samples were collected in June and July 2018. As trials continued, additional samples were collected in August and October 2018. There was major variability in the size distribution of the material collected from the recycle bin at NEFCO, see distribution in Table 7. This analysis was not completed for recycle samples collected in June or July, however based on visual observation and drying results, it is assumed the size distribution of the recycle from the earlier samples fell in between the two distributions.

There are several potential reasons for variability in the recycle biosolids size distribution –

- When a new drying train is put on-line, there is a period when pellets are created before the temperature of the biosolids can be verified to be above 80°C (as required by EPA Part 503 Rule and MassDEP 310 CMR 32). During this period all biosolids exiting the dryer bypass the screener and are sent directly to the recycle bin.

- Seasonal variability in the sludge; seasonal changes have been shown to change the material composition of sludge at some wastewater treatment plants.

- The crusher takes oversized material and crushes it to approximately 1.5 mm in diameter. If at any point the crusher was off for a period of time a greater percentage of fines may be in the recycle bin. (It is not known if this situation occurred prior to any of the biosolids sample collections.)

- Based on the size of the recycle bin, dried recycled material can spend between 2 and 5 hours in the bin while the given train is running; this means it is difficult to judge when an impact to recycle size would be seen at the outlet of the recycle bin.

- When samples were collected in August, the Plant Manager at NEFCO noted that there had been a power outage earlier in the day. This could indicate a new dryer being put back on-line and all pellets being sent to the recycle bin.

Table 7 – Size Distribution of Recycle Sample Material

Diameter Range	Recycle from August	Recycle from October
4 mesh (greater than 4.76 mm)	0.00%	0.88%
6 mesh (3.36 - 4.76 mm)	0.40%	4.06%
8 mesh (2.38 - 3.36 mm)	24.80%	5.12%
16 mesh (1.19 - 2.38 mm)	70.20%	7.07%
28 mesh (0.62 - 1.19 mm)	4.40%	60.78%
Fines	0.20%	21.02%

Conclusions and Future Work

PDADMAC had a noticeable impact on the amount of fines produced and on pellet size distribution during bench scale biosolids drying, although inconsistent from batch-to-batch. PAM, which has a lower charge density and lower molecular weight than PDADMAC, did not have a measurable impact on the pellet size distribution when compared to the drying trials without amendment. This agrees with previous research indicating the particle size of biosolids in solution was increased by PDADMAC and not by PAM. The reasons for inconsistent results from different dosage applications are unknown due to the complexity of the surface chemistry. But the sensitivity of polymer dosing on the outcomes is thought to be due to the inconsistent nature of the biosolids. Both of the cationic polyelectrolytes tested in this work limited the increase in percentage of fines that is typically seen when mixing time is extended.

Additional research would be required to determine if a single dose of polyelectrolyte could be applied consistently to biosolids without the potential of overdosing and creating significantly oversized pellets and likely issues within the pugmill. If an effective dose were to be established an economic feasibility study would need to be completed. The study would need to analyze the energy savings against the cost of the additional polymer blending equipment and the cost of the polyelectrolyte.

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Appendix #1

Trial Variables				Resulting Pellet Size Distribution					
Amendment	Dosage	Mix Time	Samples Collected	4 mesh (greater than 4.76 mm)	6 mesh (3.36 - 4.76 mm)	8 mesh (2.38 - 3.36 mm)	16 mesh (1.19 - 2.38 mm)	28 mesh (0.62 - 1.19 mm)	Fines
None	NA	5 Minutes	July	1.1%	12.0%	31.0%	39.7%	14.4%	2.1%
None	NA	10 Minutes	July	0.7%	11.5%	26.5%	28.4%	25.8%	7.1%
PDADMAC	0.015 mg/mg	5 Minutes	July	46.3%	26.4%	14.7%	9.7%	2.3%	0.6%
PDADMAC	0.015 mg/mg	10 Minutes	July	8.0%	36.0%	27.8%	22.1%	5.3%	0.9%
PDADMAC	0.03 mg/mg	5 Minutes	July	94.8%	1.5%	1.6%	1.4%	0.4%	0.3%
PDADMAC	0.0087 mg/mg	5 Minutes	July	1.8%	11.5%	26.0%	31.6%	25.2%	3.7%
PDADMAC	0.0087 mg/mg	10 Mintues	July	0.0%	10.9%	22.0%	31.1%	30.5%	5.5%
None	NA	5 Minutes	August	14.92%	27.57%	30.13%	26.47%	0.20%	1.10%
None	NA	10 Minutes	August	0.00%	4.58%	29.61%	58.74%	3.16%	3.76%
PAM	0.005 mg/mg	5 Minutes	August	2.30%	7.00%	24.48%	55.48%	7.64%	3.93%
PAM	0.005 mg/mg	10 Minutes	August	0.42%	3.91%	24.48%	55.18%	10.11%	5.79%
PDADMAC	0.015 mg/mg	10 Minutes	August	73.28%	7.14%	8.37%	9.85%	0.86%	0.37%
None	NA	10 Minutes	October	2.30%	8.06%	9.32%	13.82%	55.18%	10.99%
PDADMAC	0.015 mg/mg	10 Minutes	October	2.45%	7.35%	7.21%	18.64%	53.47%	10.20%
PAM	0.005 mg/mg	10 Minutes	October	1.71%	6.04%	6.96%	10.72%	55.64%	18.93%