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Efficacy of oil and gas produced water as a dust suppressant

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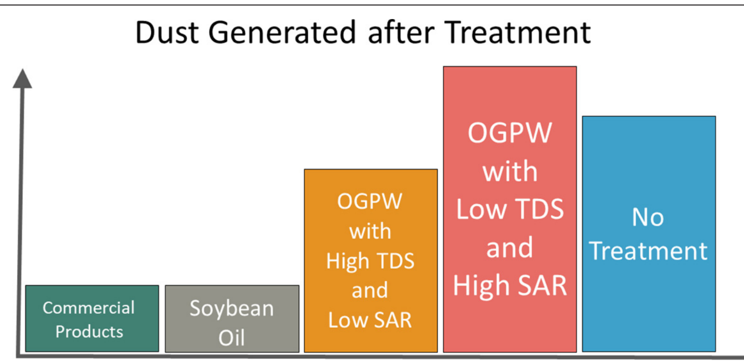
HIGHLIGHTS

- Dust suppression efficacy of all OGPW was less than commercial products.
- Sodium absorption ratio (SAR) and total dissolved solids (TDS) predict efficacy.
- OGPW with low SAR and high TDS performed best as dust suppressants.
- OGPW treated roadways lost dust-suppression efficacy following rain events.

GRAPHICAL ABSTRACT



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ABSTRACT

The effectiveness of oil and gas produced water (OGPW) applied to unpaved roads to reduce particulate matter (PM_{10}) generation has not been well-characterized. Here we quantify the efficacy of OGPW compared to commercial and alternative byproducts as dust suppressants applied to unpaved roads and estimate efficacy of a dust suppressant extrapolated from both lab experiments and published data for OGPW across U.S. states. Both treated and untreated OGPW, simulated brines, and commercial dust suppressants were characterized by major and trace element composition and then applied to road aggregate in the laboratory. PM_{10} generation after treatment was quantified, both before and after simulated rain events to assess the need for multiple applications. We found the dust suppression efficacy of all OGPW to be less than commercial products and alternative byproducts such as waste soybean oil. In addition, OGPW lost efficacy following simulated rain events, which would require repeated applications of OGPW to maintain dust suppression. The dust suppression efficacy of OGPW can be estimated based on two chemical measurements, the sodium absorption ratio (SAR) and the total dissolved solids (TDS). OGPW with the lowest SAR and highest TDS performed best as dust suppressants while high SAR and lower TDS led to greater dust generation.

1. Introduction

There are over one million miles of unpaved roads in the USA (Forman, 2004). In the United States, unpaved roads contribute heavily to particulate matter (PM) pollution, amounting to 47% of fugitive

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emissions of particulate matter less than 10 μm (PM_{10}) (Tasker et al., 2018; U.S.D. of Transportation, 2017). PM_{10} emission potential from unpaved roads is approximately 4-fold greater than paved roads (Kuhns et al., 2005). Inhalable ($<PM_{10}$) and fine PM ($<PM_{2.5}$) have been strongly correlated with impairment of the respiratory system, higher mortality rates, decreased cognitive function, and an estimated 10% of annual deaths worldwide (Dockery et al., 1993; Khan and Strand, 2018; Kim et al., 2015; Zhang et al., 2018).

Suppressing roadway dust can protect human health, improve driver safety, and eliminate unwanted dust deposition in homes or in the environment. Calcium chloride and magnesium chloride are common commercially available dust suppressants; however, the cost of these materials is usually prohibitive for typical road maintenance budgets in rural areas that need these services the most. In an effort to reduce PM_{10} emission from unpaved roads, at least 13 U.S. States allow oil and gas produced water (OGPW) to be spread as a dust suppressant (Tasker et al., 2018). OGPW is economically appealing because it is often available at no cost to the road maintenance operators and a cheap disposal option for oil and gas operators. OGPW, sometimes salty enough to be called brine or oilfield wastewater, often contains high concentrations of calcium and sodium (2530–25,800 mg/L and 23,000–57,300 mg/L, respectively –across the United States) (Lee and Neff, 2011). However, Tasker et al. (2018), showed that some elements of concern, such as lead and radioactive radium (Ra-226 and Ra-228), accumulate with even short-term repetitive treatment of road material with OGPW and have the potential to become airborne; other salts and metals can leach into waterways (Tasker et al., 2018). Additional OGPW components of concern for community members living in the vicinity of dirt roads include trace and heavy metal(loid) pollutants, such as strontium, barium, and arsenic (Pichtel, 2016; Skalak et al., 2014; Tasker et al., 2018). Not all states that allow OGPW spreading regulate or require the quantification of toxic trace elements (e.g., As, Pb, Ra-226, or Ra-228) prior to its use (Goodman, 2017).

There is currently little published research on the effectiveness of OGPW as a dust suppressant, despite its use for this purpose for upwards of 70 years (Payne, 2018). A recent literature review by Payne (2018) suggests that OGPW is not an effective dust suppressant unless spread at a rate well above environmental risk standards (Payne, 2018). High sodium content in OGPW might weaken road aggregate structure and increase dustiness (Payne, 2018; Warrence et al., 2002). Two field studies in North Dakota examined the practice of spreading OGPW. Graber et al. (2017) found that roadways in North Dakota both untreated and treated with OGPW generated similar amounts of PM (Graber et al., 2017). The overall effectiveness of OGPW may be influenced by both the sodium adsorption ratio (SAR) and the total dissolved solids (TDS) of the brine, but the road material clay content may also significantly influence the amount of dispersion of fine materials (Graber et al., 2019). SAR is a water quality indictor (Eq. (1)) often used to describe the potential of irrigation water to damage soils due to high sodium content relative to calcium or magnesium. Stallworth et al. (2020) observed the effects of SAR, TDS, and road aggregate material on synthetic brine dust suppression in a standardized laboratory setting (Stallworth et al., 2020). The authors found greater dust suppression with synthetic brines that contained higher TDS, but lower SAR values. To the authors' knowledge, no studies have systematically analyzed OGPW effectiveness as a dust suppressant, which is increasingly important as the beneficial use of OGPW is increasingly encouraged in research requests for proposals by the U.S. Environmental Protection Agency, U.S. Department of Energy and the U.S. Geological Survey. In addition, no studies have analyzed the longevity of OGPW dust suppressant efficacy following rain events observed in areas where OGPW are typically applied. In this study, rain events were simulated to compare the effectiveness that might be expected in field conditions for three types of treatments: calcium chloride, OGPW, and soybean oil. Finally, if the use of OGPW as a dust suppressant is encouraged, radium removal prior to application on the roadway may be necessary to meet potential regulatory restrictions and to our knowledge no studies have examined the efficacy of treated OGPW as a dust suppressant.

There is a clear need for efficacy studies of OGPW and other dust suppressants as their use may impact human health (Pichtel, 2016). Regulatory agencies, local and state governments, road mangers and oil and gas operators could also use the information to make informed decisions and create regulations regarding beneficial use of OGPW that are protective of human and environmental health. Therefore, the objectives of this research were to 1) quantify the efficacy of OGPW as a dust suppressant compared to commercial and alternative byproducts, and 2) estimate the efficacy of OGPW as a dust suppressant throughout the USA based on state-wide averages of OGPW chemistry. Determining dust suppressant efficacy of treated roadways will help to determine potential human and environmental health effects of using OGPW as a dust suppressant.

2. Methods

2.1. Dust suppressant characterization

Three different types of fluids were investigated for dust suppression efficacy: 1) simulated brines (including $MgCl_2$, $CaCl_2$, and NaCl), 2), OGPW, and 3) alternative products (Table 1). Twenty simulated brines were prepared in the laboratory using metal salts (calcium chloride, magnesium chloride, sodium chloride) and ultrapure water to investigate the mechanistic controls of dust suppression and mimic concentrations of commercial brines (calcium or magnesium chloride). The simulated brine formulations were chosen to reflect a range of observations of SAR and TDS values for OGPW as well as the commercial products such as $CaCl_2$ and $MgCl_2$. Results from a preliminary subset ($n = 9$) of these simulated brines were previously published (Stallworth et al., 2020).

Eight OGPWs, including both untreated conventional and treated unconventional OGPWs were used to benchmark effectiveness relative to simulated brines and alternative products. Three OGPW from conventional wells (vertical wells) that were used for road spreading were collected for this study; one was collected in 2018 from a township in Ohio (identified herein as OHB1) and two were collected from townships in Pennsylvania in 2017, (referred to in this work as PAB1, PAB2). Two conventional OGPWs from Wyoming were used (WYB1, WYB2) (McDevitt et al., 2019, 2020b). Although it is not known if these fluids were ever used for road spreading, Wyoming is a state that allows beneficial reuse of OGPW for agriculture and has a climate that could necessitate dust suppression. Three unconventional OGPW, produced water from wells that were horizontally drilled and hydraulically fractured, were tested. One unconventional OGPW from Colorado was tested (COB1). It was collected from a disposal pond located in Colorado and was reportedly used for dust suppression. Finally, two treated unconventional OGPWs from Pennsylvania were utilized to determine the feasibility of using treated unconventional OGPW as a dust suppressant. One unconventional OGPW was treated minimally, primarily to remove organics (PATO), while the second OGPW was treated to remove radium by mixing with acid mine drainage (PATR) (Ouyang et al., 2019).

Three alternative byproducts were tested including a water treatment plant softening sludge (WTP SS), soybean oil (SOY OIL), and a commercially available product, EnviroKleen®. The water treatment plant softening sludge, from Ohio, was collected after lime softening as was suggested in a survey of dust suppressant options by the South Dakota Department of Transportation (Hua et al., 2016). The pure soybean oil, a byproduct of soybean crushing, is currently used for dust suppression by a township located in northwestern Pennsylvania. The commercially available EnviroKleen® is a synthetic fluid with an added binding agent, which has been approved as a dust suppressant with minimal environmental impacts by the Pennsylvania Center for Dirt and Gravel Roads (Kunz and Little, 2015; Penn State Center for Dirt and Gravel Roads, 2018).

Table 1
Raw data for aerosol generation results of simulated brine-treated, OGPW-treated, and alternative product-treated DSA- road aggregate samples. OGPW include samples from Ohio (OH), Pennsylvania (PA), Wyoming (WY), and Colorado (CO). Alternative products include Water treatment plant softening sludge (WTP-SS), commercial dust suppressant product, Envirokleen, and waste soybean oil (SOY OIL).

Sample ID	SAR	TDS (mg/L)	Log (SAR/TDS)	$AMPM_{10}$	Std dev	Log $AMPM_{10}$	Std error
				mg/m^3	mg/m^3		
No dust suppressant added[a]							
Control - 1	–	–	–	239	16	2.4	0.03
Control - 2	–	–	–	258	15	2.4	0.03
Simulated brines[a]							
$CaCl_2$ - 1	0.12	614,252	−6.7	0.5	0.1	−0.3	0.11
$CaCl_2$ - 2	0.12	614,252	−6.7	0.4	0.4	−0.4	0.48
$CaCl_2$ - 3	0.12	614,252	−6.7	0.4	0.2	−0.4	0.20
$CaCl_2$ - 4	0.12	614,252	−6.7	1.5	0.3	0.2	0.08
$MgCl_2$ - 1	0.13	421,790	−6.5	0.3	0.1	−0.5	0.15
$MgCl_2$ - 2	0.13	421,790	−6.5	0.7	0.2	−0.2	0.15
NaCl - 1	1406	298,701	−2.3	366	33	2.6	0.04
NaCl - 2	1406	298,701	−2.3	267	9	2.4	0.01
High SAR - 1	79	181,232	−3.4	90	8	2.0	0.04
High SAR - 2	79	181,232	−3.4	108	20	2.0	0.08
Mid SAR - 1	14	163,453	−4.1	40	6	1.6	0.06
Mid SAR - 2	14	163,453	−4.1	9	2	0.9	0.11
Low SAR - 1	2	160,141	−4.9	3	1	0.5	0.18
Low SAR - 2	2	160,141	−4.9	6	1	0.7	0.11
Mid SAR/Low TDS - 1	15	24,402	−3.2	296	14	2.5	0.02
Mid SAR/Low TDS - 2	15	24,402	−3.2	349	18	2.5	0.02
Mid SAR/Mid TDS - 1	14	75,080	−3.7	20	3	1.3	0.07
Mid SAR/Mid TDS - 2	14	75,080	−3.7	25	3	1.4	0.05
Mid SAR/High TDS - 1	15	298,911	−4.3	2	0	0.4	0.07
Mid SAR/High TDS - 2	15	298,911	−4.3	3	1	0.4	0.16
Oil and gas produced water samples[b]							
WYB1 - 1	12	5771	−2.7	195	11	2.3	0.03
WYB1 - 2	12	5771	−2.7	196	23	2.3	0.05
WYB2	7	1136	−2.2	171	13	2.2	0.03
OHB1 - 1	59	244,955	−3.6	7	3	0.9	0.16
OHB1 - 2	59	244,955	−3.6	12	3	1.1	0.11
OHB1 - 3	59	244,955	−3.6	15	3	1.2	0.08
OHB1 - 4	59	244,955	−3.6	22	6	1.3	0.12
PAB1 - 1	60	292,000	−3.7	13	4	1.1	0.12
PAB1 - 2	60	292,000	−3.7	8	22	0.9	1.18
PAB2 - 1	106	356,000	−3.5	25	7	1.4	0.11
PAB2 - 2	106	356,000	−3.5	13	3	1.1	0.11
COB1 - 1	42	305,000	−3.9	0.1	0.04	−0.9	0.13
COB1 - 2	42	305,000	−3.9	0.1	0.01	−1.3	0.12
COB1 - 3	42	305,000	−3.9	0.02	0.01	−1.6	0.21
PATR - 1	28	10,079	−2.6	337	20	2.5	0.03
PATR - 2	28	10,079	−2.6	266	26	2.4	0.04
PATO - 1	63	111,073	−3.2	175	11	2.2	0.03
PATO - 2	63	111,073	−3.2	193	13	2.3	0.03
Alternative products							
WTP SS - 1	2	295	−2.2	245	22	2.4	0.04
WTP SS - 2	2	295	−2.2	152	18	2.2	0.05
WTP SS - 3	2	295	−2.2	66	8	1.8	0.05
SOY OIL - 1	–	–	–	0.02	0.01	−1.7	0.23
SOY OIL – 2	–	–	–	0.02	0.01	−1.8	0.24
EnviroKleen® - 1	–	–	–	0.02	0.00	−1.8	0.07
EnviroKleen® - 2	–	–	–	0.02	0.00	−1.7	0.06

[a] Results originally reported in Stallworth et al. (2020).
[b] TDS and values used to calculate SAR originally reported in Tasker et al., 2018.

2.2. Road aggregate characterization

Driving Surface Aggregate (DSA) was obtained from quarry stockpiles in Pennsylvania and had not previously been placed on roadways. DSA meets specifications for particle size distribution and other aggregate testing parameters as described by the Pennsylvania Department of Transportation (PennDOT) and was previously characterized, and consisted almost exclusively of limestone (Stallworth et al., 2020). The DSA used for efficacy testing had a grain size distribution with 81% of grain sizes between 1.18 mm and 9.51 mm. Of the material smaller than 1.18 mm, the grain size distribution for the DSA was 58% sand, 35% silt, and 3% clay.

DSA was used to make representative discs of unpaved road surfaces using the method developed by Stallworth et al. (2020). To create the representative discs, 100 g (dry weight) of homogenized aggregate was moistened to 5% water content, and compacted using a soil Proctor hammer in a cylindrical mold with a diameter of 6.35 cm and a height of 1.5 cm. Samples were compacted with a consistent compaction energy of 13,000 kN-m/m^3 to maintain uniform compression, similar to road beds. Compacted disc samples were then dried for 24 h at 60 °C prior to any dust suppressant application.

2.3. Aerosol generation

Dry road aggregate discs were treated with 5 mL of dust suppressant, using a pipette in 1 mL increments. This application rate corresponds to regulatory agency recommendation rates of 1.6 L/m^2 for $CaCl_2$ and is slightly below the State of Pennsylvania's recommended OGPW spreading rates of 0.5 gal/$yard^2$ (2.3 L/m^2). Treated samples "cured" for 10 min to allow for liquid penetration through the aggregate, and were then dried for 24 h at 60 °C. After 24 h, samples were placed in a small rotary drum (Model B Rotary Tumbler; Thumler's Tumblers) and tumbled for 3 min. During this time, the PM_{10} concentration in the airspace was measured using an aerosol monitor (DustTrak II Aerosol Monitor 8530; TSI). Measured aerosol concentrations from minute 2:00–3:00 were averaged, resulting in an "Average Maximum PM10" (AM PM_{10}) concentration, which was used to compare efficacy between dust suppressant samples. Road aggregate discs with no dust suppressant treatment represented control samples. Detailed methods of aerosol generation and laboratory measurements were previously described in Stallworth et al. (2020). The 2:00–3:00 min interval generally represented a maximum "plateau" in PM_{10} concentrations that were consistent between replicate tests. In addition, these bench-top tests of DSA material correlated with field measurements of PM_{10}.

2.4. Sodium adsorption ratio and total dissolved solids

OGPWs and alternative treatment samples were filtered using a 0.45 μm cellulose acetate filter and then analyzed for cations (Na, Ca, Mg, Sr and K) by inductively coupled plasma optical emission spectrometry (ICP-OES) and anions (Cl, Br, SO_4^{2-}, PO_4^{3-}) by ion chromatography (IC). A single measurement, Log (SAR/TDS) was calculated based on major anion and cation concentrations in order to evaluate the effects of TDS and SAR together. TDS was calculated based on the sum of all dissolved ion concentrations (all in mg/L).

The SAR was calculated based on dissolved metal concentrations as follows:

$$SAR = \frac{Na^{+}\left(\frac{meq}{L}\right)}{\sqrt{0.5 * \left(Ca^{2+}\left(\frac{meq}{L}\right) + Mg^{2+}\left(\frac{meq}{L}\right)\right)}} \quad (1)$$

2.5. Simulated rain events

Synthetic rainwater (pH = 4.2) was created following the EPA synthetic precipitation leaching procedure (SPLP; SW-846 Test Method 1312). The volume of leaching solution applied for each event was based on precipitation records collected by the National Atmospheric Deposition Program for the Kane Experimental Forest, located in the

Allegheny National Forest in Pennsylvania (NTN Site PA29; 41.598, −78.77) that represents an area with historical OGPW application as a dust suppressant. The 80th percentile rain event was calculated from daily precipitation records for the months of June – August in the years 2008–2017. This rainfall depth, 0.43 cm, corresponds to a rain event that would occur once every 5 days. The depth of rain was converted to a volume by multiplying by the surface area of the road disks (~31.6 cm^2) and was approximately 15 mL. For each round of leaching, the sample was placed in a plastic Buchner funnel, and 15 mL of rainwater solution was poured evenly over the disc. Discs were equilibrated for 12 h to allow for sufficient rainwater contact time. A vacuum pump was then used to remove and collect the standing solution. Next, samples were placed in an oven at 60 °C for 24 h, or until the sample remained at constant weight. Finally, samples were transferred into the mechanical rotary drum and tested for suppressant efficacy. This process was completed three times, for a cumulative 45 mL applied leaching solution to represent moderate rain events over roughly a two-week period.

2.6. Radium removal

To reduce environmental and human health risk, radium can be removed from OGPW prior to spreading on roads. OGPWs were characterized for radium activities of two long-lived radium isotopes (^{226}Ra and ^{228}Ra) using a small anode germanium gamma ray spectrometer (Canberra Instruments). Liquid samples (OGPWs and alternative liquids) were preserved to a pH less than 2 with nitric acid. Solid samples (aggregates) were sieved to <1.18 mm. Samples were equilibrated for 21 days and radium determined using the activity of the daughter products, ^{214}Pb at energy levels 295.2 keV and 351.9 keV and ^{214}Bi at 609.3 keV. ^{228}Ra was determined through the activity of decay product ^{228}Ac at 911 keV. A uranium ore tailing standard (UTS-2) from Canadian Certified Reference Material Project was used to calibrate detector efficiencies (http://www.nrcan.gc.ca/mining-materials/). All methods for determination of radium, including activity corrections for high TDS fluids, were previously described in detail (Ajemigbitse et al., 2019; Tasker et al., 2019).

The dust suppression efficacy of an OGPW, PAB1, was measured before and after three types of radium removal via chemical precipitation experiments, 1) sodium sulfate, 2) magnesium sulfate, and 3) barium sulfate. To remove radium, sodium sulfate or magnesium sulfate solution was added to 100 mL of PAB1 to co-precipitate radium with barium and strontium sulfate. The volume of sodium or magnesium sulfate added was held at 25 mL, and the concentration of the solution corresponded to a 1:1, 2:1, or 3:1 sulfate:(strontium + barium) molar ratio, although for each produced water, strontium was present in the greatest amount. Alternatively, solid barium sulfate was added to the brine samples at a solid (g barite) to liquid (kg brine) ratio of 4:2 to 4:4. The mixture was then slowly stirred for 48 h, to allow for the incorporation of radium (Zhang et al., 2014). The supernatant was then decanted, filtered through a 0.45 μm cellulose acetate filter, and used to treat road disc samples. Radioactivity of both the untreated and treated OGPWs was measured using the techniques described above.

2.7. Estimating OGPW efficacy across the USA

OGPW data for states with active conventional oil and gas drilling was accessed from the publicly available United States Geological Survey (USGS) Produced Water Database (Blondes et al., 2018). To ensure representative data quality, only samples within +/- 15% of the major ion charge balance were retained ($n = 74{,}312$ or 48%). Average major element concentrations were then used to calculate average SAR and TDS values for each state. States highlighted include those which currently regulate the spreading of OGPW for dust/deicing or road maintenance (CO, IL, IN, KS, MI, ND, NE, NY, OH, PA, SD, WV, and WY) (Tasker et al., 2018); those which regulate beneficial spreading of OGPW not exclusively for road spreading (AL, NM, TN, and VA) (Tasker et al., 2018); and states which are studying the potential to increase the beneficial use of OGPW, as encouraged by the U.S. Environmental Protection Agency (EPA), U.S. Department Of Energy (DOE), and USGS (e.g., OK, NM, TX) (USEPA, 2019a; USGS, 2020).

3. Results

3.1. Dust suppressant characterization

Three types of dust suppressant fluids (simulated brines, OGPW, and alternative products) were characterized (Table 1, Table S1). As expected, the simulated brines created to represent commercial products, $CaCl_2$, $MgCl_2$, and NaCl had the highest Ca, Mg, and Na concentrations of any samples in the study, respectively. For the conventional OGPWs, TDS ranged from 1100 to 356,000 mg/L, and SAR values ranged from 7 to 106. These OGPWs were highest in sodium (mean = 29,000 mg/L) > calcium (mean = 20,000 mg/L) > magnesium (mean = 3000 mg/L). The Log (SAR/TDS) value varied from −2.2 to −6.7. The Wyoming brines (WYB1, WYB2) had the lowest SAR values (7 and 12) and TDS (1100 and 5800 mg/L) of all the OGPWs tested in this study (McDevitt et al., 2019, 2020b). An untreated unconventional OGPW (COB1, from Colorado) that had been stored in an evaporation pond had a high TDS (305,000 mg/L) and a moderate to high SAR (42). Two treated unconventional OGPWs were also studied (PATO and PATR). PATO had similar TDS (111,000 mg/L) and SAR (63) values compared to the conventional OGPWs, but PATR had a lower TDS (10,100 mg/L) and SAR (28). Of the three alternative dust suppressants, a softening sludge from a water treatment plant had the lowest SAR and TDS (2 and 295 mg/L, respectively). The soybean oil and the commercial product EnviroKleen® were not characterized for TDS or SAR because they are not brines.

3.2. Aerosol generation

All dust concentrations are reported in terms of the Average Maximum PM_{10} (AM PM_{10}) measured over the last minute (t = 2:00–3:00 min:sec) of the dust generation experiments (Stallworth et al., 2020). The DSA road aggregate samples that were not treated with any dust suppressant (control; $n = 2$) generated 239 to 258 mg/m^3 of dust (Fig. 1; Table 1). Soybean oil and EnviroKleen® generated the least amount of dust (0.02 mg/m^3), while $CaCl_2$ (0.4 mg/m^3) and $MgCl_2$ (0.5 mg/m^3) also generated very little dust. WTP SS generated moderately high amounts of dust (AM PM_{10} = 66 to 245 mg/m^3). The road aggregate samples treated with OGPW ($n = 18$) varied by five orders of magnitude from 0.02–337 mg/m^3 for individual samples. The treated unconventional OGPWs generated dust between 184 and 301 mg/m^3. Notably, NaCl generated the largest amount of dust (267 to 366 mg/m^3) of any type of treatment, which was significantly greater than the untreated control ($p = 0.004$).

3.3. Sodium adsorption ratio and total dissolved solids

For brines, dust suppression efficacy is influenced by both the SAR and TDS of the fluid. In a previous study using simulated brines (Stallworth et al., 2020), strong linear relationships suggested that dust suppression efficacy of synthetic brines can be predicted if the SAR or TDS of a simulated brine are known. In this study, the Log (SAR/TDS) was calculated in order to combine the effects of SAR and TDS into one value that could estimate an OGPW's efficacy as a dust suppressant. A linear relationship (Eq. (2)) based on this singular value for the simulated brines supported the applicability of Log (SAR/TDS) to describe these trends (Table S2). The linear regression using the Log (SAR/TDS) of twenty simulated brines and the Log $AMPM_{10}$ generated from DSA discs produced an r^2 of 0.87 and a significant p-value (2×10^{-9}).

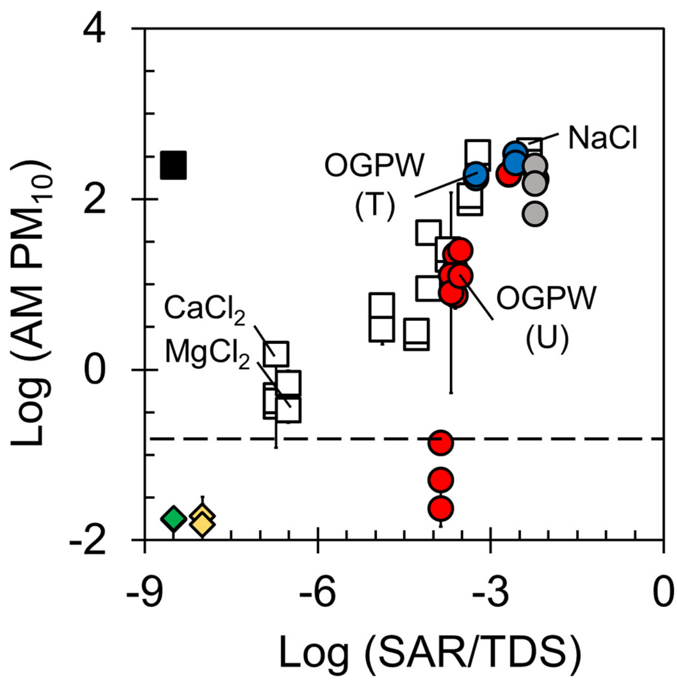

Fig. 1. Lab generated dust concentrations shown on the y-axis as the log of the Average Maximum PM_{10} (AM PM_{10}; mg/m^3) plotted against the log of the quotient of SAR divided by TDS for each dust suppressant. Black squares represent the dust generated by control samples with no treatment; white squares, simulated brines (Stallworth et al., 2020); green diamonds, EnviroKleen®; yellow diamonds, soybean oil; red circles, untreated unconventional OGPW(U); blue circles, treated unconventional OGPW(T); grey circles, water treatment plant sludge (WTP SS). Control, soybean oil, and EnviroKleen® samples were assigned arbitrary Log(SAR/TDS) values to allow for graphing. The dashed horizontal line represents the US EPA 24-h exposure limit for PM_{10}, 0.15 mg/m^3 (Log = −0.92). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

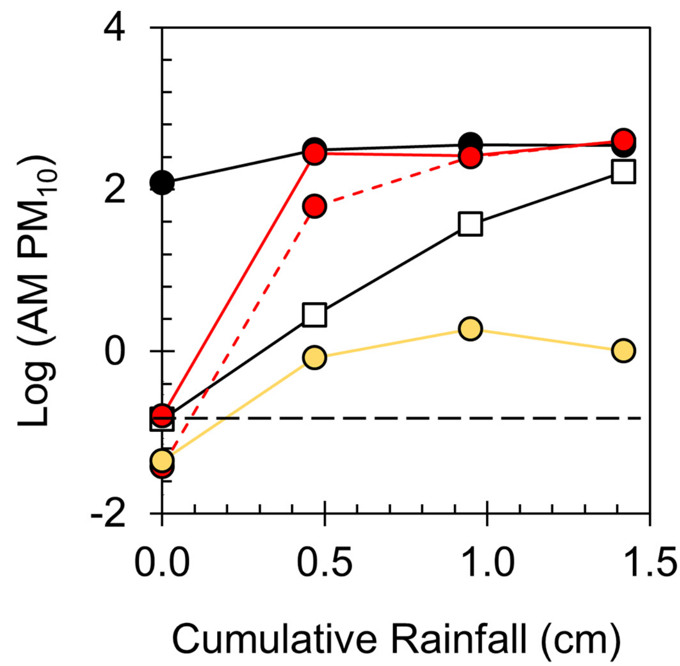

Fig. 2. Efficacy of dust suppressants after moderate summer rain events (0.45 cm) that typically occur every 5–7 days in western PA (i.e., equivalent to the rainfall depth that would fall in 24-h, every 7-days in NW PA). The y-axis shows the dust generated, as Log Average Maximum PM10 (AM PM_{10}), after cumulative increases in applied precipitation (in cm), on the x-axis. Black squares represent the untreated control; red circles connected with a solid line, OHB1; red circles connected with a dashed line, PAB1; white squares, $CaCl_2$; yellow circles, soybean oil. The dashed horizontal line represents the US EPA 24-h exposure limit for PM_{10}, 0.15 mg/m^3. For all conditions, $n = 2$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

$$Log\ AMPM_{10} = 0.66 * Log\left(\frac{SAR}{TDS}\right) + 4 \quad (2)$$

Based on values from the USGS produced water Database, OGPW from Texas has an average TDS of 81,064 and an average SAR of 121. Based on Eq. (2) these values predict an $AMPM_{10}$ of 126.

3.4. Influence of rain events

Four of the suppressants, $CaCl_2$, OHB1, PAB1, and soybean oil, were applied to DSA road materials and then subjected to simulated rain events ($n = 3$). Generally, more dust was generated following each simulated rain event. Soybean oil, however, demonstrated minimal dust generation increase after the first rain event and then little additional dust generation between rain events 2 and 3 (Fig. 2; Table S3).

The samples treated with OGPWs (OHB1, PAB1) lost efficacy by the second rain event and generated as much dust as the untreated-control samples. These samples consistently reached the instrument maximum detection of 400 mg/m^3 (Log = 2.60) following a second rain event. Samples treated with $CaCl_2$ lost efficacy with each rain event more slowly than the OGPW-treated samples and generated nearly as much dust as the untreated-control samples after three rain events.

3.5. Influence of radium removal

In the conventional OGPWs, total radium ($^{226}Ra + ^{228}Ra$) activities were between 20 and 1440 pCi/L, consistent with previous reported results (Dresel and Rose, 2010; Haluszczak et al., 2013; McDevitt et al., 2019; Rowan et al., 2011, 2015; Tasker et al., 2018; Zhang et al., 2014). PATO had a total radium value of 5900 pCi/L, more than double that of the conventional OGPWs used in this study, but similar to results reported for unconventional OGPW (Rowan et al., 2011, 2015). Radium was not detected (ND < 20 pCi/L) in PATR, nor in any of the alternative dust suppressants investigated in this study. The complete chemistry data from the dust suppressant characterizations are presented in Table S1.

One conventional OGPW, PAB1, was treated to remove radium using the method described in Zhang et al. (2014) to co-precipitate radium with barium and strontium sulfate using varying doses of barium, sodium, and magnesium sulfate (Zhang et al., 2014). For all doses of barium, sodium, and magnesium sulfate, the total radium decreased after treatment. For 1× the calculated sodium sulfate dose, the Ra decrease was almost 50% (Fig. 3), but activity still remained well above the EPA industrial discharge limit of 60 pCi/L. In contrast, both barite radium-removal doses reduced radium to near 60 pCi/L. Overall, the dust suppression efficacy of the treated brine decreased slightly (Fig. 3) for magnesium and sodium sulfate treatments, but barite treatments demonstrated similar or better reduction of PM_{10}. Two DSA samples treated with PAB1 generated 10 and 14 mg/m^3 PM_{10}. Following barite addition to remove radium, discs treated with radium-reduced PAB1 generated only 0.2 and 1.1 mg/m^3 dust (Fig. 3).

3.6. Estimating OGPW efficacy by state

The USGS Produced Water Database was utilized to calculate a state average OGPW Log (SAR/TDS) to estimate anticipated dust generation (in Log AM PM_{10}) if applied as dust suppressants (Table S4). Results of predicted dust generation from mean values of SAR/TDS calculated for OGPW from each state are presented in Fig. 4. Results for commercial dust suppressants and simulated brines are included for comparison. States were categorized based on the predicted AM PM_{10} values, which are shown across the continental USA (Fig. 5).

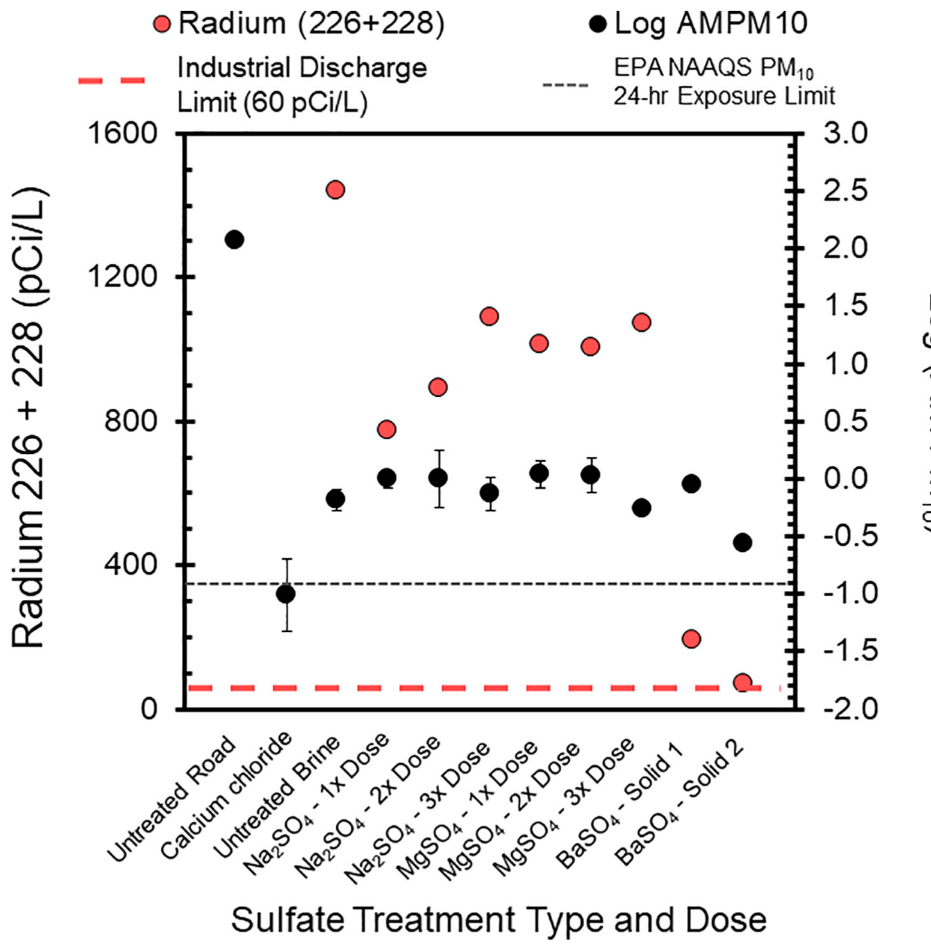

Fig. 3. Total radium (Ra-228 + Ra-226) activities (pCi/L) in treated OGPW (red circles) and resulting effects on dust generation, Average Maximum PM_{10} (AM PM_{10}) (black circles). The untreated road and $CaCl_2$ treatments contained non-detectable activities for total radium. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

4. Discussion

4.1. Dust suppressant efficacy

A total of twenty simulated brines were tested, each with varying SAR and TDS values (Table 1). Similar to previous studies of synthetic brines, there was a strong negative relationship between the SAR and dust suppression efficacy (Graber et al., 2019; Stallworth et al., 2020), and a strong positive relationship between the TDS and dust suppression efficacy (Stallworth et al., 2020). Brines highest in sodium (e.g. NaCl) performed worse than brines with little to no sodium (i.e. $CaCl_2$ and $MgCl_2$).

Eight OGPWs were tested, with ranges of TDS from 1100 to 356,000 mg/L, and SAR from 7 to 106 (Table 1). The results demonstrate the divergent effects on efficacy between TDS and SAR that occurs when OGPW is used for dust suppression. Generally, the untreated conventional OGPW samples (PAB1, PAB2, OHB1) suppressed dust most effectively (7–25 mg/m^3) of the OGPWs. These OGPWs had high SAR values (59–106) and high TDS (245,000–356,000 mg/L). Among these brines, while PAB2 had the highest TDS (an indicator of good dust suppression), it did not out-perform the other two brines. PAB2 also had the highest SAR (an indicator of poor dust suppression), which could have counteracted the benefit of high TDS. The OGPWs which suppressed dust least effectively (PATR, WYB1, WYB2; 171–337 mg/m^3) had low to mid/high SAR values (7–28), and low TDS (1100–10,000 mg/L). PATO performed within the range of the least effective OGPWs; however, it had a much higher TDS and SAR. The contrasts observed with PAB2 and PATO further indicate that TDS and SAR are both factors in OGPW dust suppression and suggest there is a limit to the effectiveness of higher TDS (and higher Ca, Mg dosing) on mitigating the detractive effects of sodium in higher SAR fluids. The implication for road managers and State agencies is that SAR and TDS calculations need to be performed to demonstrate dust suppressant efficacy of OGPW from an OG well.

To capture the combined influence of SAR and TDS in one measurement, the Log (SAR/TDS) value was calculated and plotted versus the log of AM PM_{10} (Fig. 1, Table 1). As the Log (SAR/TDS) becomes smaller (more negative), the expected dust generation decreases. The applicability of this measurement is demonstrated by comparing results from the water treatment plant softening sludge (WTP SS) and the NaCl simulated brine. WTP SS had a very low SAR (2), ideal for dust suppression; however, it also had very low TDS (295 mg/L). As such, it suppressed dust minimally (198 ± 14 mg/m^3) compared to the control (248 ± 11 mg/m^3). The Log (SAR/TDS) of the water treatment plant softening sludge was comparable to the sodium chloride simulated brine. The sodium chloride brine, while high in TDS (299,000 mg/L), also had a very high SAR value (1400). In our experiments, the Log (SAR/TDS) explained a high proportion of total PM_{10} produced (R^2 = 0.87) (Table S2). The utility of Log (SAR/TDS) is a valuable tool for road managers and state regulators seeking to rapidly assess the efficacy of OGPW within their regions. Of note, the COB1 results do not appear to fit the Log (SAR/TDS) trend, as this brine suppressed dust more effectively than predicted. COB1 has a high TDS, and was sourced from an evaporation pond which could lead to better humectant (i.e., moisture retention) properties. Evaporative concentration of the source-brine until the time of COB1 collection could have led to better performance, but

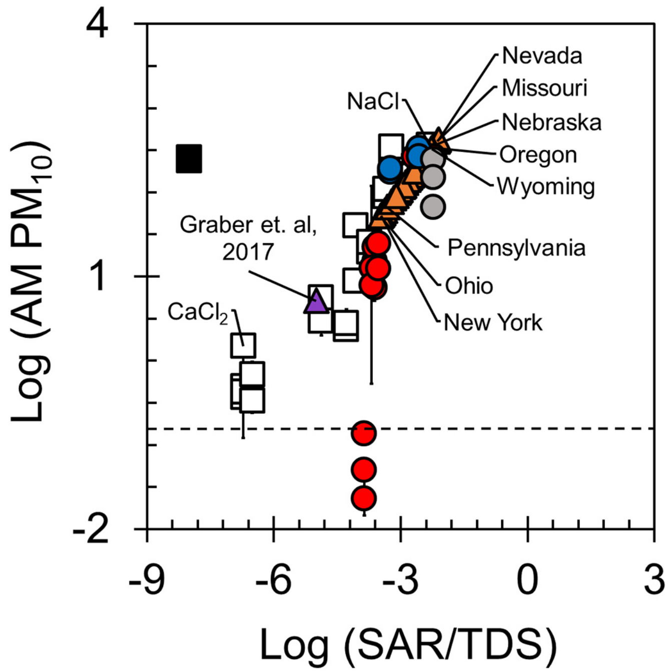

Fig. 4. Dust generated (AM PM_{10}) from laboratory samples during this study and predicted AM PM_{10} of OGPW from selected States based on Log (SAR/TDS). White squares represent simulated brines; red circles, conventional OGPW; blue circles, treated unconventional OGPW; grey circles, water treatment sludge – all laboratory-measured values. Orange triangles represent predicted dust generation for States with data entries in the USGS Produced Water Database. The purple triangle represents the predicted dust generation of the OGPW used in the Graber et al. study (Graber et al., 2017). The dashed horizontal line represents the US EPA 24-h exposure limit for $PM_{10,}$ 0.15 mg/m^3. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

one not predicted by SAR/TDS alone. Alternatively, a significant increase in humidity conditions in the laboratory at the time of testing, as compared to the other samples, could have led to increased suppression efficacy. These results suggest that further tests with the method are needed to explore humidity and other site-specific conditions.

Both the soybean oil (SOY OIL) and EnviroKleen® performed about four orders of magnitude better than WTP SS and three orders of magnitude better than the best performing OGPW (Fig. 1, Table 1). These fluids do not have SAR or TDS values; therefore, they were assigned arbitrary Log (SAR/TDS) values along with the control. Soybean oil is perhaps the most promising alternative product to OGPW because it worked as well as a commercial product and could likely be obtained at a lower price as a byproduct of soybean crushing. Although effective and commercially available, EnviroKleen® would likely be cost prohibitive for many rural road managers with in-place costs ranging from $4000-$10,000 per mile per application (Stallworth et al., 2020). Since each fluid has organic components that likely enhance their efficacies, factors not assessed in this study, they should be investigated before widespread use (i.e., to prevent the potential leaching of organic carbon and/or nutrients into nearby waterways during storm events).

4.2. Total dissolved solids and sodium adsorption ratio

Based on a 'state-average OGPW', New York, Ohio and Pennsylvania would generate the lowest amount of dust if their OGPWs were used as dust suppressants. All three of these states currently or previously spread OGPW. However, even the best performing OGPW from these states did not perform as well as alternative waste products such as soybean oil. The regression predicts that Nevada OGPWs would likely generate the most dust if used as a dust suppressant.

As States seek an inexpensive alternative to commercial dust suppressants and deicing agents, and the U.S. EPA seeks to expand beneficial use of OGPW across the U.S. (USEPA, 2019b), predicting efficacy of OGPW as a dust suppressant in other geographical areas remains imperative to maintain human and ecological health. Using the Log SAR/TDS as a predictor of dust generation, we estimated PM_{10} generation for 37 states (Table S4). We found that the use of OGPW from states such as NV, NE, and WY are expected to perform similarly to NaCl brine and untreated controls. Predicted AM PM_{10} values for CO and WY (Table S4) are near the measured values (COB1, WYB-1, and WYB2) (Table 1). Likewise, OGPW from Appalachian Basin states such as OH, PA, WV and NY are predicted to be more effective. A direct comparison for predicted laboratory PM_{10} dust generation based on the regression calculation (52–67 mg/m^3) for these states was close to the observed

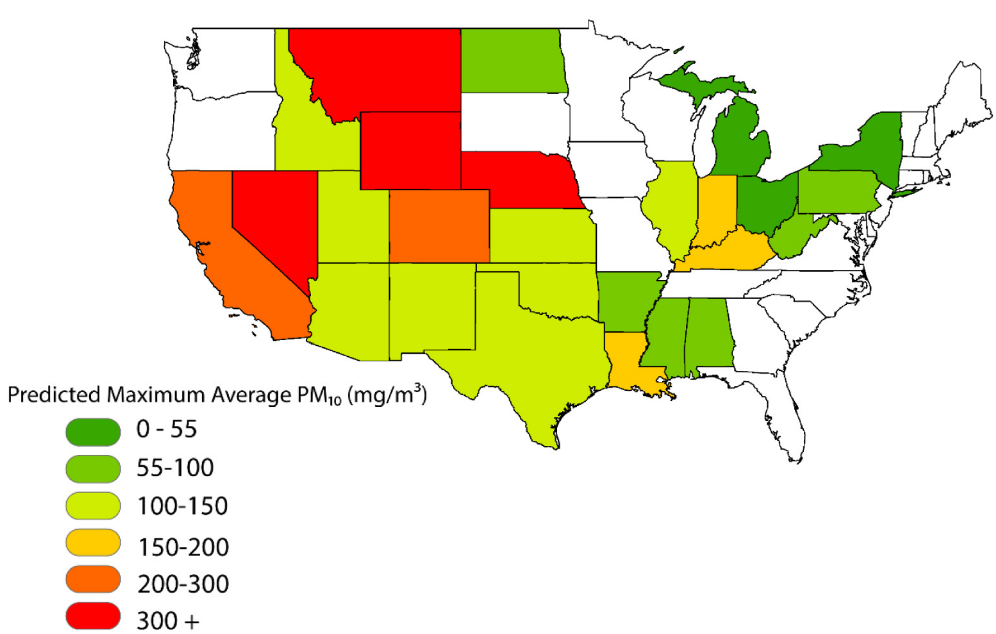

Fig. 5. Predicted dust suppression efficacy across the U.S. based on SAR and TDS values available in the USGS Produced Water Database for each State. States represented in white had less than 50 entries available in the USGS Produced Water Database.

laboratory PM_{10} generation of the tested samples (10–19 mg/m^3) (Table S5). The correlation between concentrations of dust generated in the laboratory and dust generated on roadways is yet to be defined, with additional field scale experiments required.

Due to spatial variability in the TDS and SAR of produced waters across oil and gas plays (Chaudhary et al., 2019; Scanlon et al., 2020), evaluation of OGPW at a finer spatial scale is recommended when considering applicability—especially when considering the high degree of variability within states. States may be able to conduct a similar analysis on a county-level to identify specific counties and/or formations which may be best-suited for OGPW road spreading. For example, a recent field study by Graber et al. (2017) used an ideal OGPW (low SAR, high TDS) from South Dakota in a field test (Graber et al., 2017). While the efficacy results cannot be directly compared, inputting the Log (SAR/TDS) of this OGPW into the regression equation yielded an expected dust generation that was lower than the range observed with the average USGS produced water data for South Dakota. Therefore, analysis at the local level may reveal a wider range of OGPW suitable for use as dust suppressants. However, suppressants used commercially, such as calcium chloride, magnesium chloride, and EnviroKleen®, as well as the alternative soybean oil each outperformed all OGPW tested in this study, which may indicate OGPW application for dust suppression may not be beneficial (Payne, 2018).

4.3. Influence of rain events

While dry conditions motivate the use of dust suppressants, rain events are not uncommon in seasons when suppressants are applied in some regions (e.g., Pennsylvania and Ohio). Therefore, resilience to small rain events that typically occur repeatedly during summer months was investigated for OHB1, PAB1, SOY OIL, and $CaCl_2$. With each rain event, the efficacy of each dust suppressant decreased, excluding SOY OIL, which decreased minimally and then leveled off after the first rain event (Fig. 2, Table S3). The samples treated with OGPW lost efficacy more quickly and to a greater degree than the $CaCl_2$ and SOY OIL samples; by the second rain event, the OHB1 and PAB1 generated as much dust as the untreated-control sample. These samples consistently reached the instrument maximum of 400 mg/m^3. These results further demonstrate the efficacy of an alternative product (soybean oil) relative to the OGPWs, which appear to have limited capacity to suppress dust after typical rain events observed in the summer months.

In a previous study by Tasker et al. (2018), road materials treated with OGPW were leached using the EPA Synthetic Precipitation Leaching Procedure (SPLP) (Tasker et al., 2018). In these simulated rain events on road materials treated with OGPW nearly all sodium, magnesium, and calcium leached out of the road aggregate (Tasker et al., 2018). However, the test was performed on loose, smaller aggregate, with larger volumes of leaching solution and a rotational shaker for equilibration. The method used in the current study attempted to simulate a leaching event closer to what would be observed on a road surface. It is likely that rain events will result in the loss of the constituents responsible for dust suppression over time (Warrence et al., 2002). From this study, roads treated with OGPW may be most susceptible to this loss compared to commercial grade and non-brine alternatives, requiring diligent maintenance (without over-application).

4.4. Influence of radium removal

While several of the regulations that permit the road or land spreading of OGPW specify only conventional OGPW (as opposed to unconventional OGPW generated after hydraulic fracturing (Haluszczak et al., 2013; Tasker et al., 2018)), untreated conventional OGPW still contains levels of radium above regulatory standards, such as the industrial discharge limit of 60 pCi/L (Tasker et al., 2018). The addition of sodium sulfate to OGPW removed about half of the overall radium activity (from PAB1), but also added sodium to the solution, increasing the SAR and limiting its effectiveness as a dust suppressant (Fig. 3). Conversely, the addition of magnesium or calcium sulfate to an OGPW decreased the SAR but did not increase dust suppression efficacy. This decrease in efficacy may be due to dilution – the volume of sulfate solution added had lower TDS and made up 20% of the final volume. Regardless, in all magnesium and sodium sulfate additions, removal of roughly half the radium from OGPW via co-precipitation with barium and/or strontium sulfate led to only a slight decrease in dust suppression efficacy. In contrast, radium removal was optimized with treatments of pre-formed solid barite, which removed around 90% of the radium and maintained SAR and TDS, leading to similar (and in some cases, better) dust suppression efficacy compared to untreated OGPW. Following a single dose with barite, radium values in OGPW (70–140 pCi/L) were near the industrial discharge limit of 60 pCi/L.

4.5. Limitations

Additional contaminants found in OGPW, (i.e. lead, arsenic, radium) may accumulate on roadways and in groundwater (Skalak et al., 2014; Tasker et al., 2018), which raises concerns about the safety of water resources post-application (Chen and Lippmann, 2009; Kim et al., 2015) that were not addressed here. Indeed, brines that are used for road maintenance activities such as deicing and dust suppression raise concerns about increasing the salinity of proximate water resources, because once applied, much of the salt becomes mobile and travels offsite with surface and groundwater (Bair and Digel, 1990; Eckstein, 2011; Piechota et al., 2002) which has negative consequences for agriculture, infrastructure, and aquatic life (Fay and Shi, 2012; Kaushal et al., 2018; Tasker et al., 2018). The removal of heavy metals from OGPW, such as arsenic, has been demonstrated (Akhbarizadeh et al., 2018; Fakhru'l-Razi et al., 2009; McDevitt et al., 2020a). Therefore, removal of metals prior to application on roadways could reduce some of the risks without compromising dust suppression efficacy.

5. Conclusions

Road spreading of OGPW is an established practice that is generating health and efficacy concerns as the practice gains more attention. Compared to commercial counterparts, calcium and magnesium chlorides, the presence of sodium in an OGPW can render an OGPW less effective as a dust suppressant. None of the OGPWs assessed performed as well as the commercial analogs, $CaCl_2$ or $MgCl_2$. Based on average TDS and SAR values for OGPW in each state, the OGPWs tested in this study likely represent the upper limits of efficacy of OGPW as a dust suppressant. If the justification for using OGPW is equivalency with commercial counterparts, evidence points to far less efficacy. However, removal of radium from OGPW to concentrations below regulatory levels that will reduce risk is possible, with minimal impact to efficacy.

CRediT authorship contribution statement

Audrey M. Stallworth: Conceptualization, Methodology, Formal analysis, Validation, Writing – original draft, Writing – review & editing. **Eric H. Chase:** Conceptualization, Methodology, Formal analysis, Validation, Supervision, Writing – review & editing. **Bonnie McDevitt:** Methodology, Formal analysis, Validation, Writing – review & editing. **Katherine Marak:** Methodology, Formal analysis, Validation, Writing – review & editing. **Miriam Arak Freedman:** Methodology, Formal analysis, Validation, Supervision, Writing – review & editing. **Robin Taylor Wilson:** Methodology, Formal analysis, Validation, Supervision, Writing – review & editing. **William D. Burgos:** Conceptualization, Methodology, Formal analysis, Validation, Supervision, Writing – review & editing. **Nathaniel R. Warner:** Conceptualization, Methodology, Formal analysis, Validation, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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