OPEN DUMPING AND BURNING: AN OVERLOOKED SOURCE OF TERRESTRIAL MICROPLASTICS IN UNDERSERVED COMMUNITIES

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Open dumping and burning of solid waste are widely practiced in underserved communities lacking access to solid waste management facilities; however, the generation of microplastics from these sites has been overlooked. We report elevated concentrations of microplastics (MPs) in soil of three solid waste open dump and burn sites: a single-family site in Tuttle, Oklahoma, USA, and two community-wide sites in Crow Agency and Lodge Grass, Montana, USA. We extracted, quantified, and characterized MPs from two soil depths (0-9 cm and 9-18 cm). The abundance of particles found at the three sites (35,000 to 69,200 particles kg^{-1} soil) equals or exceeds reported concentrations from currently understood sources of MPs including biosolids application and other agricultural practices. Attenuated total reflectance Fourier transformed infrared (ATR-FTIR) identified polyethylene as the dominant polymer across all sites (46.2%-84.8%). We also detected rayon (≤11.5%), polystyrene (up to 11.5%), polyethylene terephthalate (≤5.1), polyvinyl chloride (≤4.4%), polyester (≤3.1), and acrylic (≤2.2%). Burned MPs accounted for 76.3 to 96.9% of the MPs found in both community wide dumping sites. These results indicate that solid waste dumping and burning activities are a major source of thermally oxidized MPs for the surrounding terrestrial environment with potential to negatively affect underserved communities.

Environmental Significance

Our work determined the abundance and vertical distribution of microplastics (MPs) in soil surrounding open dumping and burning sites. This work unveils the abundance of MPs in the terrestrial environment around open dumpling and open burning sites near underserved communities. Generation of MPs through open dumping and burning of solid waste is an issue concerning not only our partner communities for this study in Oklahoma and Montana, but also globally, and has profound environmental implications for rural and urban underserved communities globally.

Introduction

An estimated two billion people across the globe rely on open dumping and burning to dispose of solid waste.[1, 2] Though solid waste burning may reduce waste volume, pathogen exposure,[3] and undesirable odor, it lacks protective barriers and results in refuse placed directly above the ground.[4] Open dumping and burning of solid waste are well-known sources of suspended particulate matter, odorous compounds, or leachates among other pollutants;[3, 5] however, they are also an unexplored source of terrestrial microplastics (MPs, plastic pieces with a size between 1 µm and 5 mm[6]).

It is likely that open burning sites generate MPs due to the lack of sufficient or consistent temperatures to incinerate plastic waste, which represents between 6.4 and 13% of the solid waste.[1] Partially-combusted plastics generated from these burning processes have more potential to generate MPs relative to a non-oxidized plastics. Other types of oxidized MPs (e.g., UV oxidized) have shown increased toxicity,[7] sorption capacity,[8, 9] brittleness,[10] or leaching capacity of additives [11-13] among other effects. This highlights the importance of considering not only MP quantity and plastic type, but also the MP chemistry.[14, 15]

Compared to aquatic ecosystems, the current understanding of sources and levels of MP pollution in terrestrial ecosystems is limited.[16-19] Common sources of MPs in soils include improper disposal of domestic and industrial waste, urban and rural runoff, wet deposition, and biosolids.[20, 21] However, even less research has been conducted to understand the abundance and type of MPs in soils due to open dumping and burning of solid waste, which is a common waste disposal practice in underserved rural communities.[1, 3, 22-25] Few studies have examined the occurrence of MPs in waste disposal sites, and even fewer have considered the surrounding soils.[26-28] So far, these few studies agree on microplastic occurrence[26, 27] in a range of 50 to 1110 items kg^{-1}.[28]

https://doi.org/10.26434/chemrxiv-2024-k11sz ORCID: https://orcid.org/0000-0002-4873-0454

About 18-29 million metric tons of plastic waste was burned in 2016.[3, 29] In the US, some rural communities and isolated Native American Tribes lack access to waste collection services and management facilities and, as a result, must burn solid waste in the open. Our work determined the abundance and vertical distribution of MPs in soil surrounding open dumping and burning sites at a single-family open burning site located in Tuttle, Oklahoma (Tuttle burn site), and two Native American community-wide sites in Crow Agency, Montana (Crow Agency burn site and Lodge Grass dump site). We identified the polymer type and functional chemistry of MPs and trends in MPs abundance, polymer size, and surface chemistry in relation to soil physiochemical characteristics, depth, and site history and use.

This work revealed the abundance of MPs in the terrestrial environment around open dumpling and open burning sites near underserved communities, and through this work we developed an FT-IR novel library of thermally-oxidized consumer plastics which improves spectra correlations of oxidized MPs commonly not detected with standard polymer libraries.

Methods

2.1 Quality Assurance and Quality Control

To reduce MP contamination during laboratory procedures, all glassware was sonicated for 30 min in deionized (DI) water and covered with aluminum foil until use. All procedures except for elutriation and fluorescence microscopy were conducted in a designated MP laboratory which had extra air filtration units and regular rigorous cleaning of surfaces. All applicable procedures were conducted in a laminar flow fume hood. Lab coats made of 100% cotton were worn during all laboratory procedures and synthetic clothing was avoided as much as possible during sampling. A triplicate laboratory control was conducted to detect lab-based contamination. To account for base-level environmental MP contamination, background samples were collected and run in triplicate. The background for the Tuttle burn site was a field in nearby Stillwater, OK, and the background for Crow Agency burn site and Lodge Grass dump site was adjacent ranch land. No prior dumping or burning was reported in any of these background sites.

2.2 Site Characterization and Sample Collection

Faculty of Little Big Horn Community College, Crow Agency, MT, identified 'Crow Agency burn site' and 'Lodge Grass dump site', which serve approximately 2,000 and 440 people, respectively. Homeowners of Tuttle, OK, identified the burn site which serves two people. Faculty and the homeowners described current solid waste management practices. The approximate area of each site was determined using Google Earth. The area of the Tuttle burn site was approximately 50 m^2. The areas of the Crow

Agency burn site and Lodge Grass dump site were approximately 32,200 m^2 and 36,000 m^2, respectively (Table S1). Aerial images and sampling coordinates are available in Fig. S1 and Table S3. Partners from Little Big Horn Community College reported that solid waste burning occurred regularly at the Tuttle burn site and Crow Agency burn site, while primarily dumping without burning occurred at Lodge Grass dump site. The Tuttle burn site has been in use for about 30 years, while Crow Agency burn site and Lodge Grass dump site began only about 2 years from the sampling date.

The trash piles were not homogenous; thus, sites were divided into quadrants (based on cardinal directions) to account for variation in soil or MP characteristics. Samples were collected in a randomized design, and samples were taken with two different purposes: 1) we collected a bulk sample to ~18 cm in depth using a shovel to determine texture and organic matter content; and 2) we collected soil from ~0 to 9 and 9 to 18 cm depths using a 2.54 cm diameter soil probe to evaluate MP size and content distribution. Bulk and core samples were taken from the same quadrants. Approximations in sample depth were due to varied compaction levels across the sampling areas, which at times physically limited the depth to which the probe could be driven into the soil. Three sampling locations were selected randomly within each quadrant of the pile. Subsamples were collected from each location and combined to obtain approximately ~1 kg of soil per composite sample, providing four replicate soil samples per site. Samples were collected in 3.78 L new and clean plastic sealable bags to retain moisture during transport and were refrigerated upon arrival at the laboratory. The bulk samples were used for soil texture and organic matter analyses and the 0-9 cm and 9-18 cm probe samples were used for MP analyses.

2.3 Soil Characterization & Preparation

The bulk and probe samples were each passed through a 4.75 mm sieve for homogenization then dried at 50 °C overnight. This drying temperature was chosen to prevent thermal oxidization of MPs in samples. Bulk samples were sent to the Oklahoma State University Soil, Water, Forage Analytical Laboratory for texture analysis using the hydrometer method.[30] The organic matter content of bulk samples was determined with a loss on ignition procedure.[31] Texture and organic matter characterization and area information are provided in the supplementary information file (Table S1).

2.4 Microplastic Extraction

Microplastic extractions from the 0-9 cm and 9-18 cm soil samples of each quadrant were performed in triplicate. For each replicate, 10 g of dry soil was elutriated following the procedure outlined in Forsythe et al[32] to remove dense nonplastic material from the sample. Briefly, 10 g of dry soil was sonicated in DI water to break up aggregates, then elutriated in a column for 15 min using an upflow velocity of 1.3 cm s^{-1}. Particles with a lower settling velocity were captured in a 45 µm effluent collection sieve. Particles in the sieve were rinsed into a glass beaker with water for transport to the designated MPs laboratory. At the MPs laboratory, the water-particle slurry was filtered through a 20 µm stainless steel mesh using a glass filter unit. Particles retained on the mesh were rinsed into a 250 mL glass Erlenmeyer flask with 150 mL of 7.5% w/w NaOCl for digestion. Erlenmeyer flasks were secured on an incubator shaker table at 300 rpm at 50°C for 24 h. After this, digestate was filtered through a 20 µm stainless steel mesh using a glass filter unit. Retained particles were thoroughly rinsed with DI water, then rinsed into 15 mL falcon tubes using 5.1 M $ZnCl_2$ (~693 g/L) for a density separation procedure.[33] Falcon tubes were vortexed then centrifuged at ~12,300 ms^{-2} for 5 min. The supernatant was filtered onto a 20 µm stainless steel mesh, and tubes were refilled with $ZnCl_2$ and vortexed until the pellet was

https://doi.org/10.26434/chemrxiv-2024-k11sz ORCID:

resuspended and well-mixed. This procedure was repeated until each falcon tube had been centrifuged and the supernatant filtered three times. The particles retained from the supernatant were rinsed thoroughly with DI water, then rinsed onto a 13 mm diameter 2 µm pore size Al_2O_3 filter for MPs analyses using a glass filtration unit.

2.5 Particle Quantification and Size Analysis

The particle quantification and size analyses were performed according to Quiambao et al[34] and details of the method are available in the supplementary information. Briefly, extracted particles were imaged with a stereomicroscope (AmScope 7X180X Trinocular Zoom Stereo Microscope) as initial visual identification using Al_2O_3 filters. Each Al_2O_3 filter was placed in a clean glass petri dish and dyed with ~30 µL of 2 µg Nile Red/mL methanol solution for fluorescence microscopy analyses. Particles were left to react with the dye for 10 min then rinsed with 200 µL of ACS grade ethanol on a glass filtration stack to remove excess dye. Filters were placed in a Greiner 6-well plate and imaged with a fluorescence microscope (Cytation 5 Cell Imaging Multi Mode Reader, Agilent Technologies) using an RFP filter cube (excitation 531 nm/emission 593 nm). Gen5® software was used for imaging, which allowed us to obtain a single stitched image of each filter from a series of 4x magnification images (Table S6-S9). The resolution limit of Cytation 5 cell Imaging at this magnification is about 15 µm pixel^{-1}. Each image was preprocessed to reduce background fluorescence. Microplastics were quantified by running the stitched image through the MPVAT 2.0 macros using ImageJ.[35] To avoid overquantification caused by the 15 µm pixel^{-1} limit of the Cytation, particles smaller than 40 µm were excluded from quantification and only the filter flow-through area was considered. Stereomicroscope and fluorescence microscope images are found in Table S6-S9.

2.6 Attenuated Total Reflectance - Fourier Transform Infrared Spectroscopy Analyses of MPs

The functional chemistry of suspected MPs was determined using Attenuated Total Reflectance - Fourier Transform Infrared spectroscopy (ATR-FTIR, Thermo Nicolet iN10 MX) which has a detection limit of 20 µm. One representative filter from each triplicate was selected for ATR-FTIR analysis. Ten percent of the suspected plastic particles identified during fluorescence quantification were analyzed with µ ATR-FTIR; a minimum of five particles were required for all filters regardless of MP count and a maximum cap was set at 15 particles due to lengthy analysis time. In total, 59 particles were examined from Tuttle burn site, 110 from Crow Agency burn site, and 120 from Lodge Grass dump site. Each filter was mounted on a gold mirror slide and a mosaic image of the filter flow-through area was acquired with OMNIC Picta® software to aid in the selection of particles. ATR-FTIR measurements were collected with a cooled detector and Germanium tip, 51 s collection time with 256 scans, spectral range of 4000-675 cm^{-1} and a resolution of 8 cm^{-1}. Aperture size was adapted to fit each examined particle. The resulting spectra were searched against the OMNIC Picta® stock polymer libraries (HR Polymer Additives and Plasticizers, Hummel Polymer Sample Library, Polymer Laminate Films, and Synthetic Fibers by Microscope) and an in-house generated library which included thermally oxidized plastic spectra described below (Table S2). Thermally oxidized and UV aged plastics were added to the library to improve the correlation of the environmentally relevant MPs. Using the procedure by Yang et al [36] a particle match >70% was automatically considered plastic. A match between 60-70% required manual visual comparison against the library polymer spectrum and was interpreted based on similarities in absorption peaks. Any match under 60% was not considered a plastic.

2.7 Pyrolysis Gas Chromatography/Mass Spectrometry

Soil samples were prepared for Pyrolysis gas chromatography/mass spectrometry (py-GC/MS) following the methodology explained in the section 2.3 Soil Characterization and Preparation of this manuscript.[32] Briefly, samples were weighed out between 1 – 2 mg on an ultra-balance (EPE26 Precision Balance, Mettler Toledo). Analysis was conducted with an EGA/PY-3030D pyrolysis unit (Frontier Labs, Koriyama, Japan) attached to an Agilent 6890 GC/5975 MS system (Agilent, Santa Clara, CA). Quantification was performed following the method of described by Forsythe et al[32] and all measured masses were normalized to the original sample mass to give units of mg (plastics) to g (dry soil)$^{-1}$.

Results and Discussion

3.1 Microplastics Occur in Open Dumping and Burning Sites

Particle Abundance: The community-wide open dumping and burning sites contained a higher abundance of particles compared to the single-family site (Fig. 1). The highest particle abundance was detected in the Lodge Grass dump site with an abundance ranging from 7,500 to 35,000 particles kg^{-1} in the 09 cm profile and from 5,000 to 69,200 particles kg^{-1} in the 9-18 cm profile (Fig. 1A). In the Crow Agency site, the particle abundance ranged from 5,400 to 23,600 particles kg^{-1} in the 09 cm profile and 4,400 to 32,900 particles kg^{-1} in the 9-18 cm profile (Fig. 1B). In the Tuttle burn site, the particle abundance ranged from 900 to 12,300 in the 0-9 cm soil profile and from 1,200 to 19,500 particles kg^{-1} in the 9-18 cm soil profile (Fig. 1C).

The abundance of particles at the three sites is equivalent to or far exceeds reported concentrations from currently understood key sources of terrestrial MPs including biosolids application and other agricultural practices.[37, 38] For example, biosolids can contain up to 14,000 items kg^{-1}, and concentrations higher than 5,190 MPs kg^{-1} soil have been found in biosolids-applied agricultural fields.[39, 40] A range from

900 to 40,800 items kg^{-1} soil was found in agricultural soils in Yunnan Province, China – the abundance attributed in part to plastic mulching.[41] The high concentration of MPs detected at the Tuttle burn site, Crow Agency burn site, and Lodge Grass dump site evidenced that open dumping and burning of solid waste are a key source of terrestrial MP pollution, especially in the rural and underserved communities that must utilize the practice.

The quantity of particles detected in each replicate (analytical replicate made from each composite) of all quadrants at Tuttle burn site, Crow Agency burn site, and Lodge Grass dump site is shown in Table S4, respectively. Quantification at background sites and in lab controls is found in Table S5. Stereomicroscopy and fluorescence microscopy for all sites, background, and lab controls are shown in Table S6-S9. Microplastics were detected at all background sites. A range of 1,500 to 4,000 particles kg^{-1} were detected at the Tuttle

burn site background location and 600 to 5,400 particles kg^{-1} at the Crow Agency burn site and Lodge Grass dump site background location (Table S5).

Vertical Distribution of Particle Abundance and Particle Size: The vertical distribution of particle abundance varied from site to site (Fig. 1A-C). At all three sites, the highest MP count was observed in the 9-18 cm soil depth. This may be the result of MPs in the 0-9 cm depth being more susceptible to transport off-site by runoff, or perhaps being transported through the soil and accumulating in a lower depth by infiltration during rain events. Simulated rainfall experiments have shown rainfall intensity contributes to the migration of MPs deeper into the soil profile, with MPs smaller than 1 mm especially prone to deeper migration or accumulation in runoff compared to larger MPs, and fibers and films being more mobile compared to particles.[42]

The abundance and size of detected MPs varied between the sampled depths at each site (Fig. 1A-F). At the Crow Agency burn site, the 0-9 cm core contained 14,733 ± 6,319 particles kg soil^{-1} compared to 17,175 ± 9,085 particles kg soil^{-1} in the 9-18cm core; the highest abundance found in this site was

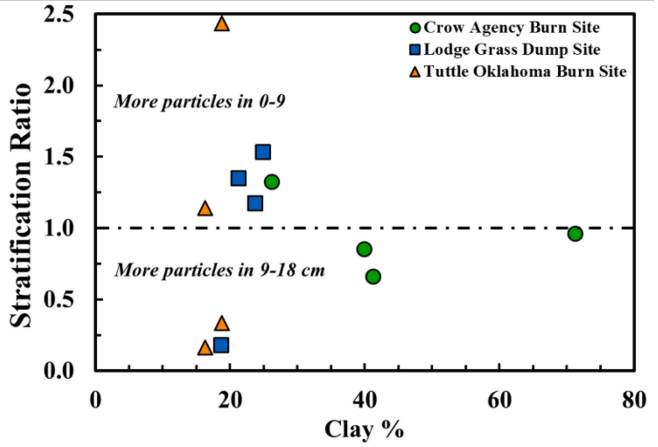


Fig. 3. Stratification ratio of particle abundance (particle concentration at 0-9 cm/particle concentration at 9- 18 cm) plotted against percentage of clay in all quadrants of each site.

136.97 and 226.8 ± 117.30 µm for the 0-9 cm and 9-18 cm cores, respectively (Fig. 1D). At the Lodge Grass site, the average size was 224.3 ± 128.73 and 263.9 ± 187.11 µm for the 0-9 cm and 9-18 cm

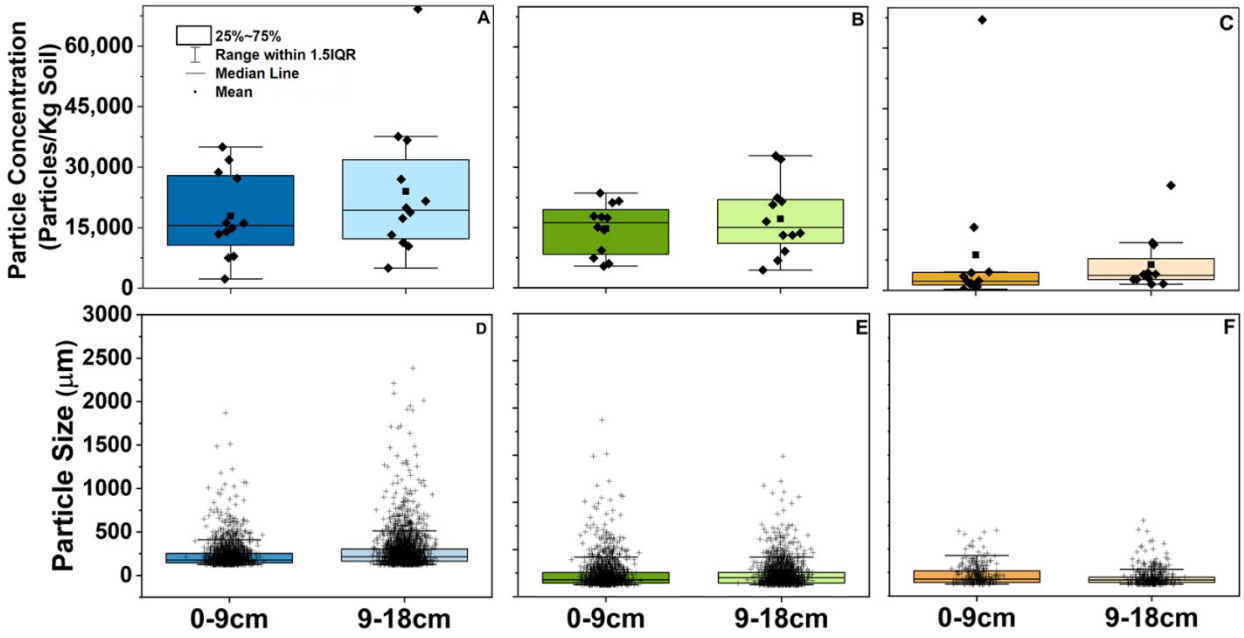


Fig. 1. Abundance of particles (particles kg-1 soil) at each soil depth at Lodge Grass dump site (A), Crow Agency burn site (B), and Tuttle burn site (C) and size distribution of all particles detected with Nile Red at Lodge Grass dump site (D), Crow Agency burn site (E), and Tuttle burn site (F). All analyses were performed in analytical triplicate.

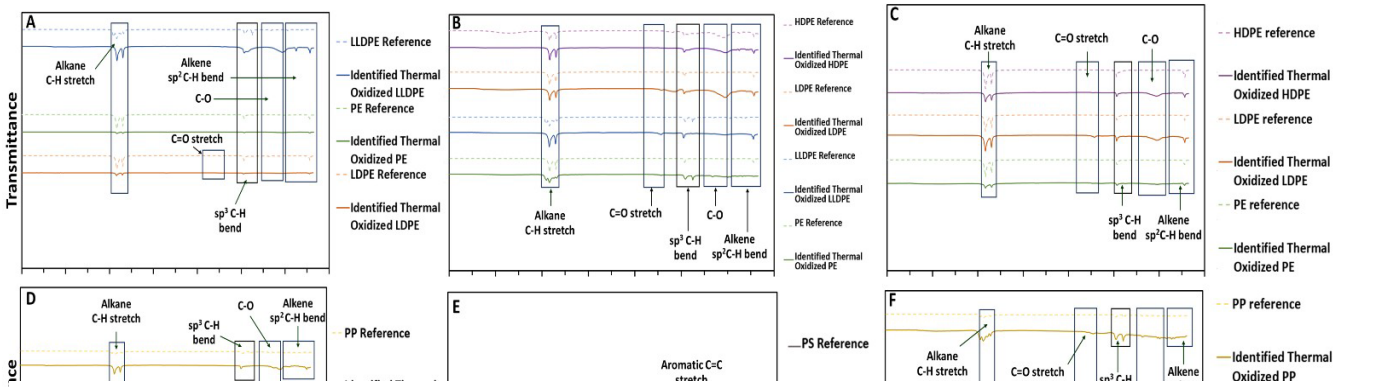
32,000 particles kg soil^{-1} (Fig. 1A). On the other hand, the Lodge Grass site contained 17,917 ± 10,405 particles kg soil^{-1} in the 0-9 cm core compared to 24,000 ±17,364 particles kg soil^{-1} in the 9-18cm core; the highest abundance found in this site was 69,200 particles kg soil^{-1} (Fig. 1B). Finally, in the Tuttle burn site, the average abundance was 2,845 ± 3,224 particles kg soil^{-1} found in the 0-9 cm core compared to 4,283 ± 5,189 particles kg soil^{-1} in the 9-18 cm core; the highest abundance was 19,500 particles kg soil^{-1} (Fig. 1C). The large range of particles quantified across these sites (900 - 69,200 particles kg soil^{-1}) indicate the heterogenicity of the abundance that can be found in open dumping and burning sites. In terms of size, the average size of particles found at Crow Agency site was 228.4 ±

cores, respectively (Fig. 1E). Finally, the Tuttle site contained particles with an average size of 224.5 ± 106.99 and 191.8 ± 84.72 µm (Fig. 1F). Similarly, the wide range of particle size (126.4 – 1872. 8 µm) highlights the heterogenicity of particle sizes found in these sites. We recognize that a smaller size of particles is likely present in these samples; however, the detection limit was restricted by the high throughput of samples we analyzed. We present differences between depths as a stratification ratio (the ratio of particles found in the upper/lower profile) and explore associations with clay content (Fig. 2). Overall, the 0-9 cm depth contained higher particle concentrations in soils with less clay, which may be due to compaction in silty or sandy soils preventing the transport of particles to lower profiles. There are a number of

factors which can contribute to the migration and accumulation of MPs throughout a soil profile.[43] Corn roots have been found to contribute to the upward migration of MPs in soil depth of 6-12 cm, while earthworms have been shown to contribute to the transport of MPs to deeper sediments.[19, 44] Wet and dry cycling can also contribute to the transport of MPs into the deeper soil column, with more cycling corresponding to deeper migration of MPs, and smaller MPs (<21 µm) showing the highest mobility.[45] While these factors are helpful to consider, there are a number of additional variables which may affect the transport of MPs including topography, soil texture, soil compaction, and other soil metrics. Additionally, climatic differences between sites such as annual rainfall, rainfall intensity, and freeze-thaw cycling may contribute to differences in MP transport throughout the soil profile at each site.[45]

In our experiments, the soil texture of the Tuttle burn site was sandy or sandy loam, while the soil sampled at Crow Agency and Lodge Grass contained higher content of clay and may have held more water-stable aggregates. Particularly, the soil sampled in Crow Agency contained 2.5 times more clay than the Tuttle soil. Clay is often positively associated with forming aggregates and controlling pore architecture.[46] In soils with clay content such as Lodge Grass dump site, larger MPs could potentially infiltrate deeper into the soil due to less-compact characteristics and/or greater pore connectivity. Conversely, only smaller particles could infiltrate the highly compact soil at Tuttle burn site due to potentially decreased pore connectivity and reduced water infiltration. Clay particles are highly reactive and may affect the transport of MPs into lower depths.[47] The 9-18 cm profile of Crow Agency burn site contained more MPs in three of the four quadrants; these clayrich soils may promote the accumulation of MPs at this depth due to clay-MP interactions. There was little difference in soil texture across the quadrants of Lodge Grass dump site and the Tuttle burn site (single family). Thus, the variation in stratification ratio should consider other variables alongside texture including bulk density, aggregation, and water infiltration.

Few studies have examined the vertical distribution of MPs through the terrestrial soil environment and have found contrasting results.[39, 48, 49] For example, the MP abundance was slightly higher at 20 cm depth (mean particle concentration = 53.2 items m^{-2}) compared to 5 cm depth (mean particle concentration = 34.6 items m^{-2}), and smaller average particle size was observed in the deeper



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Polymer Type	Relative Abundance (%)					
	Tuttle OK		Crow Agency		Lodge Grass	
	0-9 cm	9-18 cm	0-9 cm	9-18 cm	0-9 cm	9-18 cm
PE	11.5	0.0	2.2	8.5	5.1	10.7
PE - UV Aged	0.0	3.1	0.0	3.4	3.4	0.0
PE - Burned	7.7	28.1	13.3	5.1	16.9	23.2
HDPE - Burned	7.7	3.1	20.0	10.2	20.3	21.4
LDPE - Burned	15.4	34.4	15.6	18.6	22.0	17.9
LLDPE - Burned	3.8	6.3	13.3	18.6	16.9	10.7
PP	0.0	0.0	2.2	1.7	0.0	0.0
PP - Burned	19.2	6.3	6.7	13.6	0.0	0.0
CV - Burned	11.5	9.4	8.9	0.0	5.1	3.6
PS - UV Aged	0.0	0.0	4.4	1.7	0.0	0.0
PS - Burned	11.5	0.0	2.2	5.1	3.4	5.4
PET - Burned	0.0	3.1	2.2	0.0	5.1	0.0
PVC	0.0	0.0	2.2	1.7	0.0	0.0
PVC - Burned	0.0	0.0	2.2	0.0	0.0	0.0
Polyester	0.0	0.0	0.0	0.0	0.0	1.8
Polyester - Burned	0.0	3.1	0.0	0.0	0.0	0.0
Acrylic	0.0	0.0	2.2	1.7	0.0	0.0
Other	11.5	3.1	2.2	10.2	1.7	5.4

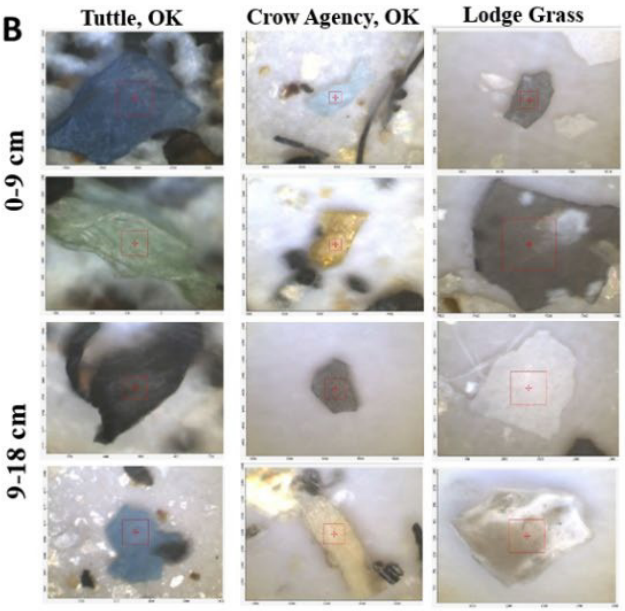

Fig. 4 Microplastics found in the sites sampled. A) Relative abundance. B) Representative microplastics found in the sites of Tuttle OK (top to bottom): Tuttle OK-West-Shallow-1, Tuttle OK-W-Deep-1 Tuttle OKW- Shallow-2, Tuttle OK- W -Deep -7; microplastics found in Crow Agency (top to bottom): CABS-NW Shallow- 2, CABS-NW-Shallow-12 CABS-SE-Deep-4; and microplastics found in Lodge Grass: LG-SW-Deep- 12; LG -SW-Shallow-13; LG-SW-Shallow-7. CABS: Crow Agency burn site; LG: Lodge Grass. N, S, W, E indicate cardinal directions

profiles in agricultural soils from China.[48] A 3-times higher concentration in the 0-10 cm profile compared to the 20-30 cm was quantified in German agricultural fields.[49] Microplastics concentrations did not significantly vary between 0-10 cm and 10-30 cm soil depths of biosolids-applied fields in Spain.[39] Similar to the variation observed between existing studies, we found different trends in MP vertical distribution between sites (Fig. 1). It is important to note that this study does not examine MPs smaller than 20 µm nor nanoplastics (NPs); considering these size fractions in future work will provide a more holistic picture of MP and NP fate and transport. **3.2 Functional Chemistry Analysis of MP**

A total of 58 of the 60 particles examined from Tuttle burn site, 105 of 110 particles examined from Crow Agency burn site, and 118 of 120 particles examined from Lodge Grass dump site were confirmed as MPs by ATR-FTIR. Almost all MPs identified at each site matched with burned or UV aged plastic spectra. Examples of identified MPs from each site are shown in Fig. 3 and all examined particles are shown in Tables S10S16 and Table S21.

Oxidation features or changes in the spectra can be observed in several particles compared to the reference spectra (Fig. 3A-F). The spectra were analyzed according to the correlation charts described in Larkin et al.[50] The functional chemistry of LLDPE MPs indicated changes in the alkane C-H stretching region (3000-2840 cm^{-1}), sp^3 C-H bend (~1410-1325 cm^{-1}), C-O (1125-1000 cm^{-1}), and alkene sp^2 C-H (650-1000 cm^{-1}) compared to the reference spectrum (Fig. 3A). Likewise, the functional chemistry of PE shifted in the alkane C-H stretching region (3000-2840 cm^{-1}), sp3 C-H bend (~1410-1325 cm^{-1}), C-O (1125-1000 cm^{-1}), and alkene sp^2 C-H (1000-650 cm^{-1}) and PP MPs (Fig. 3A, 3B, and 3C). On the other hand, PS MPs show different features in the aromatic C=C- stretching region (1675 – 1475 cm^{-1}) region (Fig 3D-F). Our results indicate that in open burning sites, MPs are exposed to conditions that modify their functional chemistry which likely affects the reactivity and mobility of those MPs in the environment.

Discrepancy in functional chemistry between the reference spectra, weathered environmental, and thermally oxidized microplastics likely leads to the misidentification or underidentification of microplastics by current spectral identification tools. The current challenges of spectral identification highlight the need to generate more environmentally relevant spectral libraries that contain thermally aged polymers. However, more information is needed regarding the occurrence of thermally oxidized MPs in the environment.

High temperatures and UV radiation may be both defined as oxidation processes; however, UV radiation is driven by photochemical reactions (e.g., oxidation, reduction decomposition, and polymerization).[51] Particularly in thicker plastics, these are diffusion dependent and occur in the first 500 to 900 µm layer of the plastic.[52] Thermal oxidation, on the other hand, can affect the surface and bulk within the same process as a function of the temperature regardless the thickness of the plastic. Our work evidenced that open dumping and burning of solid wastes are a source of MPs with a distinct functional chemical signature.

3.3 Thermally Oxidized MPs in Soils Nearby Open Burning Sites

Our results indicated that burned MPs occurred with more frequency in all the sites across the different depths (Fig 4). In the 0-9 cm depth, the prevalence of burned MPs ranged from 71.2% to 89.8% (Fig. 4A), while the prevalence burned MPs of the 9-18 cm depth ranged from 82.1% to 93.8% across the three sites (Fig. 4A). Lodge Grass, MT, was the site with the highest abundance of burned MPs in the 0-9 cm range (89.8%), while Tuttle, OK, was the site with highest relative abundance in the 9-18cm range (93.8%). In all the sites, polyethylenebearing burned MPs (i.e., HDPE, LDPE, LLDPE) showed the highest abundance in both soil depths across all sites (34.6 – 76.3%). Finally, when considering the abundance of all plastics regardless of their state of oxidation, polyethylene-bearing MPs were the most abundant type in all depths and sites (average 69.78% ± 14.61), followed by PP (8.27% ± 7.88), CV (6.41% ± 4.29), and PS (5.62% ± 3.85).

The higher prevalence of PE compared to other polymers was confirmed by py-GC/MS in all the sites sampled for total depths reported (Crow Agency burn site at 5.97 ± 1.75 mg g^{-1}, Lodge Grass dump site at 4.99 ± 1.95 mg g^{-1}, and Tuttle burn site at 6.13 ± 2.59 mg g^{-1}) (Fig. 5.). However, the actual concentration of MPs found in these samples with py-GC/MS may be underestimated it since the reference materials (standards) available for making calibration curves and quantifying MPs also exclude thermally and UV oxidized MPs. Microplastics were found with less frequency at background locations and lab controls than at the open dump and burn sites. Only two MPs were detected from the representative lab control sample: one polyethylene and one polypropylene, likely from laboratory procedures in which plastic could not be avoided. Compared to lab controls and background site particles which were primarily white or clear fibers, a variety of particle morphologies (rectangular, oval-shaped, round, etc.) and colors (blue, pink, grey, etc.) were observed in the open dump and burn site particles (Fig. 4B). Overall, these findings coincide with the polymers most commonly used in household and single-use plastic products that are often discarded in municipal solid waste.[53]

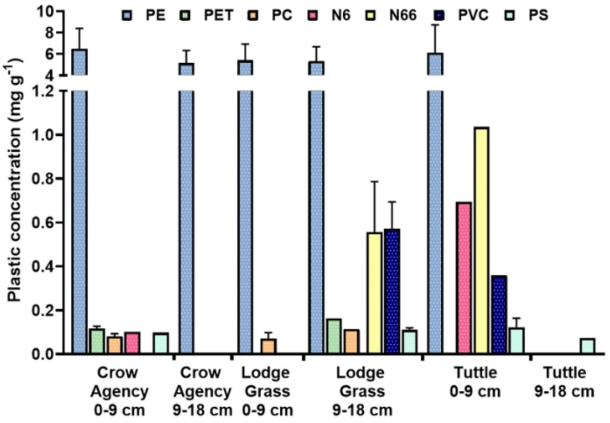

Fig. 5. Soil plastic concentration (mg g^{-1}) measured by pyGC/MS for the Crow Agency burn site, Lodge Grass dump site, and Tuttle, Oklahoma burn site 0-9 and 9-18 cm samples. Plastic types are indicated as polyethylene (PE), polyethylene terephthalate (PET), polycarbonate (PC), nylon 6 (N6), nylon 66 (N66), polyvinyl chloride (PVC), and polystyrene (PS).

Conclusions

The results of this work identify open dumping and burning of solid wastes as a source of elevated concentrations of terrestrial MPs. Burning practices result in the generation of oxidized MPs which

differ in abundance, size, and functional chemistry from parent plastic waste solids. Microplastics with a more oxidized functional chemistry will likely have different effects on soil properties, plants, or biochemistry cycles than the effects of non-thermally oxidized MP that have been described.[44, 54-57] The concentration of MPs found in single family and community-wide sites (up to 69,200 particles kg^{-1} soil) equals or exceeds the concentration of MPs found at other sites with high concentrations of MPs. The high concentration of MPs identified at the single-family open dumping and burning sites highlights the potential of small sites to be as affected as larger sites. Generation of MPs through open dumping and burning of solid waste is an issue concerning not only our partner communities for this study in Oklahoma and Montana, but also has profound global environmental implications for rural and urban underserved communities. Open dumping and burning of solid waste are utilized by approximately a quarter of all humans on earth, and more work is needed to understand the full scope and impact of this practice on the surrounding environment.

Author Contributions

Kendra Z. Hess: methodology, validation, formal analysis, investigation, writing - original draft, and visualization; ***Kyle R. Forsythe:*** methodology, investigation; ***Xuewen Wang:*** methodology, validation, investigation, formal analyses, data visualization; ***Gwen Tipling:*** investigation; ***Jesse Jones***: investigation; ***Melissa Mata:*** investigation; ***Victoria Hughes***: investigation***; Christine Martin:*** resources; ***John Doyle:*** resources; ***Justin Scott:*** methodology, validation, formal analysis; ***Matteo Minghetti:*** methodology, resources, writing – review and editing; ***Andrea Jilling:*** methodology, formal analyses, resources, writing – review and editing, data visualization; ***José M. Cerrato:*** resources, funding acquisition, writing – review and editing, data visualization***; Eliane El Hayek:*** resources, , funding acquisition, writing – review and editing; and ***Jorge Gonzalez-Estrella:*** conceptualization, methodology, writing - review & editing, supervision, resources, project administration, funding acquisition; writing – review and editing, data visualization.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This study was funded by the National Institute on Minority Health and Health Disparities (Award Number P50MD015706), and the U.S. Geological Survey (Grant/Cooperative Agreement G21AP10597). Justin Scott was supported by the National Institute of Environmental Health Sciences (NIEHS)
1R15ES034901-01 The content is solely the responsibility of the authors and does not necessarily represent the official views of the National Institutes of Health nor of the U.S. Geological Survey.

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