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Microplastics are ubiquitous and increasing in soil of a sprawling urban area, Phoenix (Arizona)

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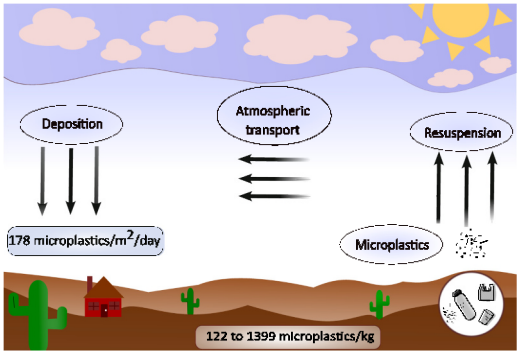
HIGHLIGHTS

- Microplastics occurred in all soil samples collected from a semi-arid region in AZ.
- Spatial variability of microplastics depicted substantial variability.
- A significant increase in soil microplastics was observed from 2005 to 2015.
- The average sizes of soil microplastics is smaller in 2015 compared to those in 2005.
- PE, PS, PVC, PA, PES and PP were the main polymers identified.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics are environmental contaminants that have been extensively studied in marine and aquatic environments; terrestrial ecosystems, where most microplastics originate and have the potential to accumulate, typically receive less attention. This study aims to investigate the spatial and temporal soil concentrations of microplastics in a large desert metropolitan area, the Central Arizona–Phoenix Long-Term Ecological Research (CAP-LTER) area. Soil samples from the Ecological Survey of Central Arizona (ESCA) surveys (2005 and 2015) were leveraged to study spatial distributions and the temporal change of microplastic abundances. The temporal soil microplastics data were supplemented by microplastics deposition fluxes in a central location within the area (Tempe, AZ) for a period of one year (Oct 5th, 2020 to Sept 22nd, 2021). Samples were processed and microplastics were counted under an optical microscope to obtain quantitative information of their distribution in soil. Results for the spatial variation of the microplastic abundances in soil samples in Phoenix and the surrounding areas of the Sonoran Desert from 2015 depict microplastics as ubiquitous and abundant in soils (122 to 1299 microplastics/kg) with no clear trends between different locations. Microplastics deposition fluxes show substantial deposition in the local area (71 to 389 microplastics/m^2/day with an average deposition flux of 178 microplastics/m^2/day) but the role of resuspension and redistribution by dust storms to deposition may contribute to the unclear spatial trends. Comparison between the 2005 and 2015 surveys show a systematic increase in the abundance of microplastics and a decrease in microplastics size. Micro-Raman spectroscopy identified a variety of plastics including PE, PS, PVC, PA, PES and PP. However, a majority of microplastics

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remafined chemficaflfly unfidentfiffiabfle. Poflyethyflene was present fin 75 % of the sampflfing sfites and was the most abundant poflymer on average fin affl sofifl sampfles.

1. Introduction

Mficropflastfics are a growfing concern as pervasfive envfironmentafl poflflutants. They are mostfly secondary fin nature and resuflt from the breakdown of flarger pflastfics over tfime finto mficropflastfics, generaflfly deffined as pflastfic partficfles < 5 mm fin sfize (Arthur et afl., 2009). Prfimary mficropflastfics are purposefuflfly manufactured at thfis smaflfl sfize, tend to have unfiform shapes fincfludfing spherficafl mficrobeads, and orfigfinate from sources such as personafl care and abrasfive cfleanfing products (Reveflfl et afl., 2021). Secondary mficropflastfics generaflfly exhfibfit more dfiverse shapes.

Exfistfing research shows that mficropflastfics are found fin finterconnected envfironments; from the Artfic to Antarctfica, fincfludfing the depths of oceans, freshwater bodfies, atmospherfic deposfitfion and sofifl envfironments (Patfifl et afl., 2022; Lfi et afl., 2023; Zhao et afl., 2023). Whfifle numerous studfies have finvestfigated the occurrence and abundance of mficropflastfics fin marfine and freshwater envfironments (Andrady, 2011; Erfiksen et afl., 2013; Horton et afl., 2017; Bergmann et afl., 2017; Kflefin and Ffischer, 2019; Chandrakanthan et afl., 2023) research fin terrestrfiafl ecosystems remafins fless wefll devefloped despfite the fact that a majorfity of the pflastfic debrfis reachfing the oceans has orfigfinated on fland (Lebreton et afl., 2022).

A revfiew study has estfimated that the annuafl reflease of pflastfic waste to fland fis 4–23 tfimes greater than that refleased to the oceans (Horton et afl., 2017). Afllthough sofifl mficropflastfics were ffirst reported fin 2016 (Dfing et afl., 2022), mficropflastfics research fin terrestrfiafl ecosystems remafins underrepresented and accounts for onfly about 5 % of the research on mficropflastfics (Dfing et afl., 2022). Street runoff, flandffiflfls and atmospherfic deposfitfion are potentfiafl finput pathways of mficropflastfics finto terrestrfiafl ecosystems, makfing sofifls a sfignfifficant sfink for synthetfic mficropartficfles fin terrestrfiafl ecosystems (Moflfler et afl., 2020). Moreover, pflastfic muflch ffiflms and sofifl condfitfioners used fin agrficuflturafl amendments are potentfiafl sources of sofifl mficropflastfics (Ng et afl., 2018). Sofifls can afllso act as a source of atmospherfic mficropflastfics to other envfironmentafl reservofirs through resuspensfion of fugfitfive dust, thereby contrfibutfing to the gflobafl mficropflastfics transport cycfle (Zhang et afl., 2020).

Mficropflastfics fin terrestrfiafl envfironments can cause changes fin sofifl ecosystems affectfing sofifl structure, pflant growth and mficrobfiafl actfivfity (De Souza Machado et afl., 2019). A meta-anaflysfis fin agrficuflturafl sofifls fin Chfina reveafled that mficropflastfics affect sofifl enzymes fincfludfing ffluorescefin dfiacetate hydroflase and urease, finffluence the buflk densfity of sofifl, and finterfere crop growth (Hu et afl., 2022a, 2022b). Furthermore, pflants and crops are flfikefly to uptake mficropflastfics present fin sofifl, subsequentfly becomfing a source of mficropflastfics present fin pflant-derfived food used for human consumptfion (Lfiang et afl., 2023). In sofifl envfironments, mficropflastfics can degrade through mficrobfiafl actfion as mficroorganfisms can utfiflfize the carbon fin pflastfic poflymer chafins for thefir growth (Mohanan et afl., 2020; Huang et afl., 2023). Owfing to the flow organfic matter fin dryfland sofifls (Marusenko et afl., 2011), sofifl mficrobes fin these flocatfions may resort to metaboflfize anthropogenfic carbon sources such as mficropflastfics. Studfies have shown mficropflastfics to have an acute toxfic effect on sofifl organfisms such as nematodes (Kfim et afl., 2020). In addfitfion to the dfirect toxfic effects caused by mficropflastfics, they can afllso act as vectors of other contamfinants such as Poflycycflfic Aromatfic Hydrocarbons (PAHs) and metafls found fin sofifls (Hfifldebrandt et afl., 2021; Hu et afl., 2022a, 2022b).

Thfis study finvestfigates sofifl concentratfions and deposfitfion ffluxes of mficropflastfics fin the Phoenfix, AZ, metropoflfitan area. The study area fis vast (6400 km^2) and fis a rapfidfly growfing urban metropoflfis that has been fintensfivefly studfied for urban ecoflogy for over 20 years (Grfimm and Redman, 2004). Our study utfiflfizes sampfles from the Ecoflogficafl Survey of Centrafl Arfizona (ESCA) performed by the Centrafl Arfizona–Phoenfix Long-Term Ecoflogficafl Research (CAP-LTER), a flarge-scafle ffiefld survey that fis conducted fin the CAP-LTER study area characterfizfing the urbanfized, suburbanfized, and agrficuflturafl areas of metropoflfitan Phoenfix and the surroundfing Sonoran Desert (Grfimm and Redman, 2004). Prevfious studfies have finvestfigated varfious key ecoflogficafl findficators and contamfinants, fincfludfing soot bflack carbon concentratfions and fisotopfic composfitfions, PAHs and flead concentratfions fin sofifls (Marusenko et afl., 2011; Zhuo et afl., 2012; Hamfiflton and Hartnett, 2013). We fleveraged the Ecoflogficafl Survey of Centrafl Arfizona (ESCA) 200-pofint survey (2005 and 2015) to study spatfiafl dfistrfibutfions and the temporafl change of mficropflastfic abundances. We suppflement the temporafl data fin sofifls wfith the coflflectfion of atmospherfic deposfitfion ffluxes of mficropflastfics fin a centrafl flocatfion wfithfin the area (Tempe, AZ) for a perfiod of one year (Oct 5th, 2020 to Sept 22nd, 2021). Affl sofifl and deposfitfion sampfles were processed and characterfized usfing an optficafl mficroscope to obtafin quantfitatfive finformatfion of the dfistrfibutfion of mficropflastfics. Chemficafl characterfizatfion of mficropflastfics was performed by mficro-Raman spectroscopy to understand thefir chemficafl composfitfion.

2. Materials and methods

2.1. Sampfling of micropflastics

2.1.1. Sampfling of micropflastics in soifl

Sofifl sampfles used fin the study were coflflected by CAP-LTER from the ESCA 200-pofint survey (2005 and 2015). CAP-LTER program fis one of twenty-efight LTER sfites funded by the Natfionafl Scfience Foundatfion and fis one of the two LTER sfites that specfifficaflfly study urban ecoflogy (Grfimm and Redman, 2004). CAP-LTER studfies how human actfivfitfies aflter the functfionfing of ecosystems, and subsequentfly urban sustafinabfiflfity, fin Centrafl Arfizona and metropoflfitan Phoenfix where two trfibutarfies of the Coflorado Rfiver, the Saflt and Gfifla Rfivers, merge (Grfimm et afl., 2013). The ESCA 200-pofint survey fis a ffiefld survey that fis conducted every 5 years at approxfimatefly 200 sampfle pflots randomfly flocated fin the urbanfized, suburbanfized, and agrficuflturafl areas of metropoflfitan Phoenfix, and the surroundfing Sonoran Desert (Ffig. S1) (Grfimm and Redman, 2004). The sampflfing pflots (30 m × 30 m) are randomfly flocated usfing a tesseflflatfion-stratfiffied duafl-densfity sampflfing scheme. Forty-efight sampfles from 2015 were seflected to study the spatfiafl varfiatfion aspect, whfifle fourteen sampfles from 2005 and 2015 were anaflyzed for the temporafl varfiatfion of mficropflastfics. Topsofifl sampfles (2 cm depth) were used for thfis study as thfis range coufld be expected to have the hfighest abundance of mficropflastfics accumuflated over tfime by dfirect finput from terrestrfiafl sources and atmospherfic deposfitfion. Top sofifl sampfles from the same study area were sampfled and anaflyzed fin a prevfious study for other envfironmentafl contamfinants, such as PAHs (Marusenko et afl., 2011). The sofifls anaflyzed fin thfis study are desert sofifl sampfles, mafinfly from urban and suburban areas of metropoflfitan Phoenfix and remote areas fin the surroundfing areas of the Sonoran Desert. Accordfing to the Unfited States Department of Agrficuflture – Natfionafl Resource Conservatfion Servfice (USDA-NRCS, http://sofifls.usda.gov/), sofifls fin Centrafl Arfizona are cflassfiffied as arfidfisofls (too dry for the growth of mesophytfic pflant flfife) and entfisofls (flfitfle to no evfidence of deveflopfing pedogenfic horfizons) (Bohn et afl., 2001; Hamfiflton and Hartnett, 2013).

2.1.2. Sampfling of micropflastics in air

Totafl atmospherfic faflflout (dry and wet deposfitfion) sampfles were coflflected by means of gflass funnefls fin 4 L gflass bottfles (Ffig. S2) on the observatfion deck of the Interdfiscfipflfinary Scfience & Technoflogy Bufifldfing

#4 on the Tempe campus of Arfizona State Unfiversfity (33.4179° N, 111.9284° W). Sampfles were coflflected for 14 days each, from Oct 5th, 2020 to Sept 22nd, 2021. The suburban flocatfion fis flocated fin the cfity of Tempe, Arfizona, approxfimatefly efight mfifles east of downtown Phoenfix. Once coflflected, the sampfles were covered wfith aflumfinum fofifl to prevent contamfinatfion.

2.2. Sampfle processing

The sofifl sampfles were wefighed (60 g) and sfieved (8″-FH-SS-SS-US-#3–1/2, Hogentogfler & Co.Inc., MD, USA) to remove materfiafl flarger than 5.6 mm. The foflflowfing approach was based on the methods manuafl from the Natfionafl Oceanfic and Atmospherfic Admfinfistratfion (NOAA) for mficropflastfics anaflyses fin water and beach sedfiment sampfles and adapted as approprfiate to process the sofifl sampfles (Masura et afl., 2015; Chandrakanthan et afl., 2023). Ffirst, sfieved sampfles were subjected to wet peroxfide oxfidatfion to remove naturafl organfic matter. A 10 mL aqueous 0.05 M Fe (II) soflutfion prepared from $FeSO_4 7H_2O$ (Sfigma Afldrfich, MO, USA) and 10 mL of 30 % hydrogen peroxfide (Sfigma Afldrfich, MO, USA) were added to sampfles for 5 mfin at room temperature and subsequentfly heated to 60 °C on a hot pflate (Ffischer Scfientfiffic, NH, USA) to remove naturaflfly occurrfing organfic matter. Sampfles were subsequentfly made to undergo a densfity separatfion step usfing a 1.8 g cm^{-3} NaI soflutfion prepared from NaI (Sfigma Afldrfich, MO, USA). The saturated NaI soflutfion aflflows the separatfion of mficropflastfics, fincfludfing reflatfivefly denser pflastfics such as PVC (≤1.58 g cm^{-3}) from the sampfle (Nueflfle et afl., 2014; Zhang and Lfiu, 2018). After aflflowfing sofifds to settfle, the ffloatfing mficropflastfics were ffifltered through pre-ffired gflass mficroffiber ffifilters (Whatman GF/A, Sfigma Afldrfich, MO, USA). The ffifilters were oven-drfied at 80 °C untfifl compflete dryness fin a Mufffle furnace (Vuflcan 3-1750, CA, USA).

The sampfle coflflectors for totafl atmospherfic faflflout (funnefl and bottfle) were thoroughfly rfinsed thrfice wfith ufltrapure water (>18.4 MΩ cm, Pureflab Fflex, IL, USA) to ensure that afffl mficropflastfics adherfing to the gflass surface were recovered. Sampfles were processed sfimfiflar to an approach adopted fin a recent study reportfing mficropflastfics suspended fin afir fin Tempe, AZ (Chandrakanthan et afl., 2023). The aqueous extracts were obtafined and subjected to wet peroxfide oxfidatfion to eflfimfinate naturafl organfic matter. The soflutfions were vacuum ffifltered through baked gflass ffiber ffifilters (Whatman GF/A, Sfigma Afldrfich, MO, USA). The ffifilters were then oven-drfied at 80 °C to compflete dryness fin a Mufffle furnace (Vuflcan 3–1750, CA, USA).

Preventfive measures to mfinfimfize background contamfinatfion durfing mficropflastfics anaflyses fis fimportant to produce reflfiabfle resuflts (Prata et afl., 2021; Shanmugam et afl., 2022; Hfidaflgo-Ruz et afl., 2012). A proceduraf bflank was run durfing afffl stages of sampfle processfing fincfludfing wet peroxfide oxfidatfion, densfity separatfion and subsequent anaflyses. Gflassware washed wfith a detergent and rfinsed three tfimes wfith ufltrapure water (>18.4 MΩ cm, Pureflab Fflex, IL, USA) were subsequentfly baked and used durfing afffl stages of sampfle processfing and anaflyses. The cflean equfipment was covered to mfinfimfize potentfiafl afirborne mficropflastfics contamfinatfion. Workfing surfaces were cfleaned wfith ufltrapure water (>18.4 MΩ cm, Pureflab Fflex, IL, USA) and fisopropyfl aflcohofl (Ffischer Scfientfiffic, MA, USA) prfior to sampfle anaflyses. The usage of synthetfic textfifles was mfinfimfized, and a ffiber was drawn from the flab coat durfing each day of flaboratory anaflysfis and observed under the optficafl mficroscope to check for ffibers wfith sfimfiflar appearance fin sampfles. The recovery percentage of mficropflastfics from ffifilters was assessed through matrfix spfikes wfith a known number of mficropflastfics (5 μm–5000 μm). A sofifl sampfle (60 g) sourced from the ESCA 200-pofint survey fin 2015 was spfiked wfith a known number of mficropflastfics (n = 60). The mficropflastfics were photographed and sfized usfing a dfigfitafl mficroscope (Lefica DM6B-Z, Germany) equfipped wfith the Lefica DFC7000 T camera and the Lefica Appflficatfion Sufite X (LAS X) software prfior to spfikfing, and were dfistfinctfive of coflor and shape. The artfifficfiafllfly fincorporated mficropflastfics fincfluded Poflyethyflene (PE), Poflypropyflene (PP), Poflyvfinyfl Chflorfide (PVC), Poflystyrene (PS) and Poflyethyflene terephthaflate (PET) rangfing fin sfize from 5 μm - 5000 μm. The spfiked sofifl sampfle was subjected to wet peroxfide oxfidatfion and subsequentfly densfity separatfion sfimfiflar to the approach fin Sectfion 2.2. The spfiked mficropflastfics were observed on a ffiflter under the dfigfitafl mficroscope (Lefica DM6B-Z, Germany) equfipped wfith the Lefica DFC7000 T camera and the Lefica Appflficatfion Sufite X (LAS X) software. The recovery test showed a recovery of 95 % of spfiked mficropflastfics.

2.3. Opticafl microscopy

Ffifilters were examfined under a dfigfitafl mficroscope (Lefica DM6B-Z, Germany) equfipped wfith the Lefica DFC7000 T camera and the Lefica Appflficatfion Sufite X (LAS X) software. The sfize range of finterest was seflected as 5 μm - 5000 μm; both the sfize and the morphoflogy of each mficropflastfic was noted to characterfize each sampfle. The flower sfize flfimfit fis 5 μm due to the smaflflest possfibfle sfize resoflutfion of the dfigfitafl mficroscope. As there fis no standard deffinfitfion for the dfistfinctfion between a ffiber and a fragment fin the mficropflastfics flfiterature, we used an operatfionafl deffinfitfion for the observed shapes of mficropflastfics where a ffiber was recognfized to be cyflfindrficafl fin shape wfith an aspect ratfio (flength/dfiameter) ≥ 3, whfifle a fragment was recognfized to be shard-flfike and fflattened. Mficropflastfics were sfized aflong thefir flargest dfimensfion usfing the software program ImageJ (versfion 1.5, Natfionafl Instfitute of Heaflth, USA, http://fimagej.nfih.gov/fi).

The foflflowfing crfiterfia were used to fidentfify mficropflastfics under the optficafl mficroscope (Hfidaflgo-Ruz et afl., 2012; Chandrakanthan et afl., 2023). Onfly objects that dfid not dfispflay ceflfluflar or organfic structures were fidentfiffied as mficropflastfics. Further, onfly ffibers that were generaflfly equaflfly thfick through thefir entfire flength exhfibfitfing 3-dfimensfionafl bendfing were fidentfiffied as mficropflastfics wfith further accountfing for ffiber spflfittfing whfich can occur fin mficropflastfic ffibers. Ffinaflfly, partficfles that exhfibfit cflear and homogenous coflor throughout thefir entfire structure were fidentfiffied as mficropflastfics wfith the exceptfion of the appearance of bfleachfing, bfiofoufffing and weatherfing whfich coufld cause patterns or strfipes.

2.4. Statisticafl anaflysis of the temporafl variation of soifl micropflastics (2005 and 2015)

A Mann-Whfitney Test ($p \leq 0.05$) was performed usfing OrfigfinPro, Versfion 2023 (Orfigfin Lab Corporatfion, Northampton, MA, USA) to compare dfifferences between the sofifl mficropflastfic abundances fin 2005 and 2015.

2.5. Chemicafl characterization using Micro-Raman spectroscopy

Mficro-Raman spectroscopy was used to provfide chemficafl characterfizatfion of the poflymers fin fidentfiffied mficropflastfics. Afffl mficropflastfics counted under the optficafl mficroscope were anaflyzed under the mficro-Raman spectrometer. Raman data was coflflected from 50 to 3800 cm^{-1} usfing a custom-bufiflt mficroscope and a Mfitutoyo M Pflan Apo 50×/0.42 objectfive. An Acton Research monochromator (SpectraPro-300fi) utfiflfizfing a 600 g/mm gratfing and coupfled to a Roper Scfientfiffic CCD (modefl LN/CCD 1340/100-EB/1) recorded the sfignafl. A Coherent Sapphfire soflfid state cw flaser emfittfing 532 nm served as the excfitatfion source and the data was caflfibrated usfing cycflohexane wfith known peak posfitfions. The spatfiafl and spectrafl resoflutfions of the mficro-Raman spectrometer were 1 μm and 1 cm^{-1} respectfivefly.

3. Results and discussion

3.1. Spatiafl variation of micropflastics

Mficropflastfics were found fin afffl sofifl sampfles, thereby findficatfing thefir ubfiqufitous presence fin sofifl envfironments fin Phoenfix and surroundfing

areas of the Sonoran Desert (Ffig. S3). The mficropflastfic abundance fin sofifl sampfles from 2015 ranged from 122 to 1399 mficropflastfics/kg wfith a heterogeneous spatfiafl dfistrfibutfion depfictfing no cflear spatfiafl trends (Ffig. 1). Three repflficates sampfled were anaflyzed for each sampflfing flocatfion (Tabfle S1). The reflatfive standard devfiatfion of the measurements was on average 6 %. The hfighest mficropflastfic abundance was found at Sfite 14 whfich fis flocated fin a resfidentfiafl area wfithfin the cfity of Phoenfix, AZ. The flowest mficropflastfic abundance was observed at sampflfing Sfite 8; a resfidentfiafl flocatfion wfithfin the cfity of Peorfia, AZ.

Sampflfing sfites K6, I14, L14, U21, AB19 had hfigher mficropflastfic abundances compared to other sampflfing flocatfions and were fin the vficfinfity of roadsfides or pubflfic aflfleyways.

Comparfing the observed mficropflastfic abundances wfith those reported fin flfiterature fis not strafightforward due to the dfiscrepancfies fin sampflfing approaches, sampfle processfing, mficropflastfics fidentfifficatfion, operatfionafl deffinfitfions of mficropflastfics and finstrument detectfion flfimfits. However, fit can be usefufl to put mficropflastfic abundances fin sofifl finto context by comparfing wfith other exfistfing studfies (Tabfle 1). A recent revfiew reported that mficropflastfics fin sofifl gflobaflfly range from 0.36 to 160,000 mficropflastfics/kg, where a majorfity of the reported studfies are from Chfina (Zhang et afl., 2022). Mficropflastfic abundances reported fin Lut and Kavfir Deserts fin Iran, Northeast Tfibetan Pflateau fin Chfina, Swfiss Fflood pflafin sofifls and Badafin Jaran Desert fin Chfina are 1–2 orders of magnfitude flower than that observed fin the current study (Scheurer and Bfigaflke, 2018; Feng et afl., 2020; Abbasfi et afl., 2021; Ifi et afl., 2022). Thfis coufld be attrfibuted to the reflatfivefly hfigher flower sfize flfimfits of finterest fin the aforementfioned studfies (≤ 100 μm, 20 μm, 125 μm and 40 μm) when compared to that of the current study (5 μm) as thfis study observed a greater number of mficropflastfics fin the smaflfler sfize categorfies (Sectfion 3.2). The observed dfifferences fin mficropflastfic abundance coufld afso be

Table 1
Summary of sofifl mficropflastfics reported fin prevfious studfies.

Locatfion	Mficropflastfics/kg	Sfize range/ μm	References
Lut and Kavfir Deserts, Iran	20 (0–83)	≤100–500	(Abbasfi et afl., 2021)
Northeast Tfibetan Pflateau, Chfina	47 (20 110)	20–5000	(Feng et afl., 2020)
Swfiss Fflood pflafin sofifls	≤593	125–5000	(Scheurer and Bfigaflke, 2018)
Northern Germany farmfland sofifls	4 ± 12	1000–5000	(Harms et afl., 2021)
Shanghafi, Chfina farmfland sofifls	78 ± 13	30–5000	(Lfiu et afl., 2018)
Shaanxfi Provfince, Chfina	1430–3410	Domfinant <500	(Dfing et afl., 2020)
Southwestern Chfina	18,760 (7100–42,960)	Domfinant (50–1000)	(Zhang and Lfiu, 2018)
Takflamakan desert, Chfina	119–7292	Domfinant <500	(Lfi et afl., 2022)
Mu Us Desert, Chfina	2696 (1360–4960)	0–5000	(Dfing et afl., 2021)
Lahore, Pakfistan	4483 (1750–12,200)	50–5000	(Raffique et afl., 2020)
Fars provfince, South Centrafl Iran	Pfiefld 1; 380 (40–830) Pfiefld 2; 510 (200 1100)	≤100–5000	(Rezaefi et afl., 2022)
Badafin Jaran Desert, Chfina	6 (1 12)	40–5000	(Wang et afl., 2021)
Phoenfix and the surroundfing areas of the Sonoran Desert	122–1399	5–5000	Current study

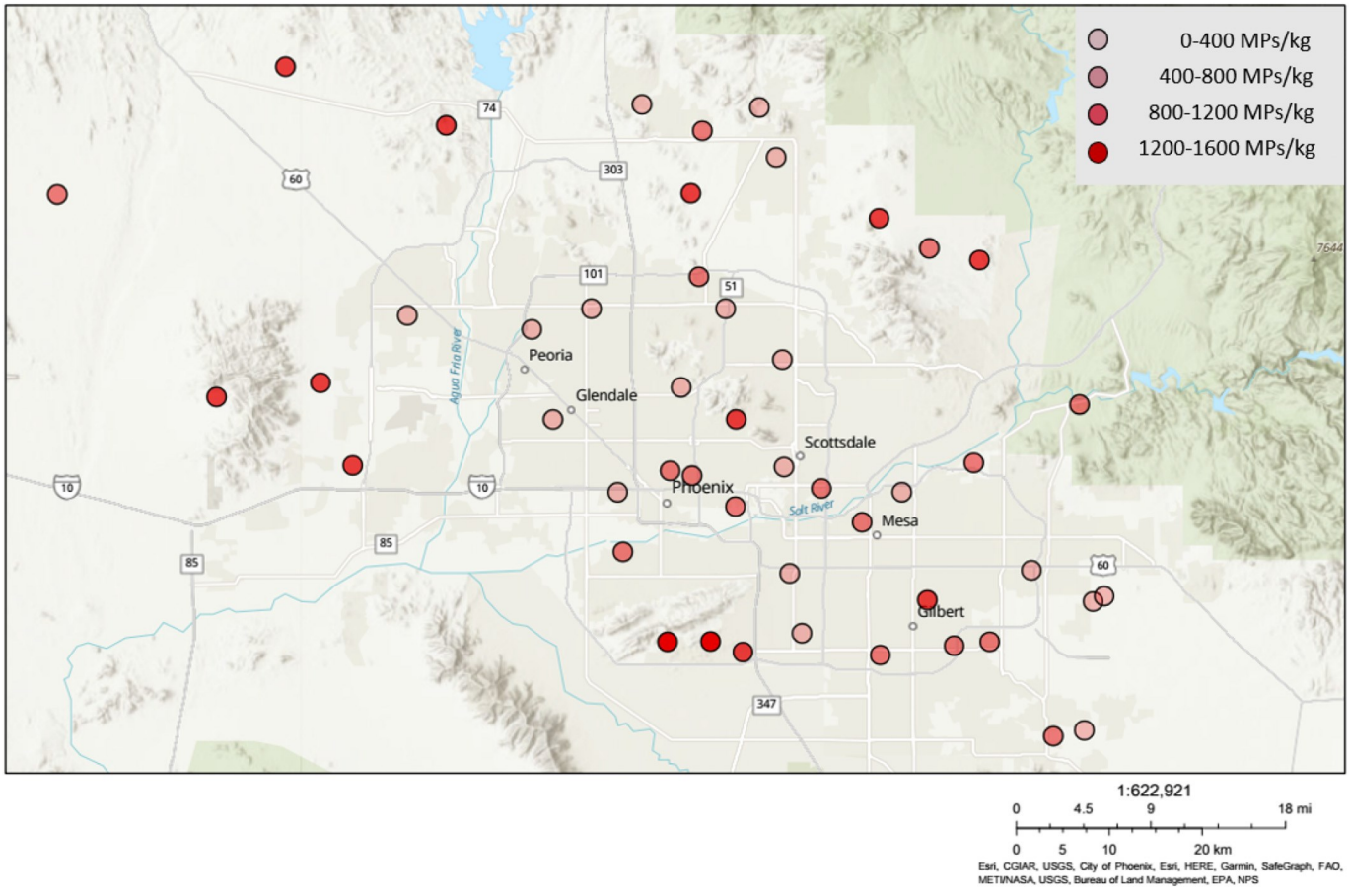

Fig. 1. Spatfiafl dfistrfibutfion of mficropflastfics fin sofifl sampfles from 2015.

attrfibuted to the dfifferences fin mficropflastfics finput vfia dry and wet deposfitfion, resuspensfion and dfiflutfion by mobfifle dust/sand partficfles, and envfironmentafl condfitfions that can weather mficropflastfics to sfizes that evade fidentfifficatfion by the anaflytficafl technfiques used fin each study (e.g. make partficfles smaflfler). The mficropflastfic abundance fin the current study fis an order of magnfitude flower than that reported fin Lahore, Pakfistan (Raffique et afl., 2020); note thfis study specfifficaflfly flfinks the hfigh measured mficropflastfic abundance to hfigh popuflatfion densfity fin the study area.

Substantfiafl varfiabfiflfity was observed spatfiaflfly for mficropflastfic abundance across the study area. A prevfious study finvestfigatfing the densfity of pflastfic trash fin the Sonoran Desert, AZ states the occurrence of passfivefly-dfispersed pflastfic trash fin the desert dfid not appear to dfissfipate wfith dfistance from potentfiafl sources of orfigfin and were findependent of road proxfimfity (Zyflstra, 2013). Prfior work suggests a majorfity of the mficropflastfics fin the envfironment orfigfinate from secondary degradatfion of flarger pflastfics (Kasmurfi et afl., 2022). As such, fit fis not surprfisfing that hfigher mficropflastfic abundances were observed fin reflatfivefly remote areas fin our study as envfironmentafl transport and degradatfion can flead to secondary mficropflastfics. An addfitfionafl source such as atmospherfic deposfitfion of mficropflastfics coufld be a contrfibutfing factor for mficropflastfics fin sofifl.

Three prevfious studfies addressed the spatfiafl varfiatfion of sofifl contamfinants wfithfin the CAP-LTER area (Marusenko et afl., 2011; Zhuo et afl., 2012; Hamfiflton and Hartnett, 2013). PAHs were reported to be present fin surface sofifls aflong major hfighways fin the Phoenfix, AZ metropoflfitan area fin a prevfious study (Marusenko et afl., 2011). A prevfious study reportfing soot bflack carbon concentratfions dfid not observe a strong correflatfion between soot bflack carbon concentratfions and dfistance to the urban core (Hamfiflton and Hartnett, 2013). A study fin 2012 finvestfigated the dfistrfibutfion of trace eflements fin sofifl sampfles as part of the Centrafl Arfizona–Phoenfix Long-Term Ecoflogficafl Research (CAP-LTER) from the Ecoflogficafl Survey of Centrafl Arfizona (ESCA) 200-pofint survey fin 2005. A majorfity of trace eflements depficted hfigher concentratfions above thefir crustafl averages. The author attrfibutes the accumuflatfion of eflements to other addfitfionafl sources such as atmospherfic deposfitfion (Zhuo et afl., 2012). Consfistent wfith these other studfies, the measured mficropflastfics fin the current study aflso show no cflear pattern reflatfive to fland use, anthropogenfic actfivfitfies or proxfimfity to urban core.

A recent study finvestfigatfing the spatfiafl dfistrfibutfion of sofifl mficropflastfics fin Hafinan Isfland, Chfina reports a heterogenous dfistrfibutfion wfith hfigh varfiabfiflfity between sampflfing sfites (Khan et afl., 2023). Aflso, a prevfious study finvestfigatfing the mficropflastfics dfistrfibutfion fin a Centrafl Asfian Desert (an encflosed area wfith aflmost no human actfivfity) attrfibutes the source of mficropflastfics to atmospherfic transport and afirborne mficropflastfics deposfitfion (Wang et afl., 2021). The dfistrfibutfion of sofifl mficropflastfics fin arfid and semfi-arfid envfironments fin Fars provfince, south-centrafl Iran, was reported to be heterogenous between ffiefflds and sampflfing flocatfions wfith no cflear trend (Rezaefi et afl., 2022). Thfis study suggests that the regfion experfiences sfignfifficant dust storms, whfich fis a sfimfiflar occurrence fin the regfion of finterest fin our study (Eagar et afl., 2017). Eagar and coworkers estfimated that haboob dust storms aflone mfight account for 75 % of the dust deposfitfion flocaflfly (Eagar et afl., 2017). Wfind-bflown dust fis an fimportant factor for the transport of anthropogenfic contamfinants such as mficropflastfics fin arfid and semfi-arfid regfions (Ashraffi et afl., 2014). A recent study fin the metropoflfis of Shfiraz, Iran estfimated that a majorfity of mficropflastfics (>90 %) fin deposfited dust were derfived from outsfide of the cfity durfing an fintense dust storm event fin May 2018 (Abbasfi et afl., 2022). Thfis suggests that dust storms coufld affect the flocafl sfite-specfiffic sofifl mficropflastfic abundances through both deposfitfion and resuspensfion, and hence envfironmentafl processes woufld redfistrfibute mficropflastfics throughout the regfion.

3.1.1. Deposition ffluxes of micropflastics

The deposfitfion ffluxes (coflflected as 2-week averages) of mficropflastfics fin Tempe, AZ for a one-year perfiod ranged from 71 to 389 mficropflastfics/

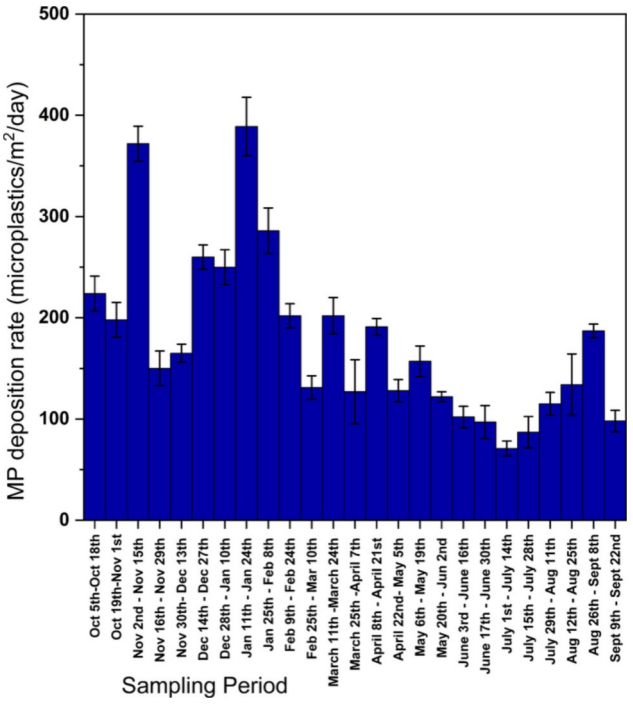

Fig. 2. The deposfitfion fflux of mficropflastfics (MP) fin Tempe, AZ. The error bars represent the standard devfiatfion of the mean obtafined from three repflficate measurements.

m^2/day wfith an average deposfitfion fflux of 178 mficropflastfics/m^2/day (Ffig. 2). Mficropflastfics were fidentfiffied fin afffl deposfitfion sampfles coflflected through the one-year perfiod. Hfigher deposfitfion ffluxes were observed durfing (Nov 2nd- Nov 15th, 2020) and (Jan 11th-Jan 24th, 2021) compared to other bfiweekfly sampflfing perfiods. Rafinfaflfl was not observed and recorded from the weather statfion flocated at the sampflfing sfite (Earth Networks Inc., an AEM company, https://www.earthnetworks.com/) durfing the aforementfioned two bfi-weekfly perfiods. Sfince Tempe, AZ recefived very flfimfited rafinfaflfl durfing the sampflfing perfiods, fit was not possfibfle to perform a correflatfion anaflysfis between mficropflastfics deposfitfion ffluxes and precfipfitatfion events. The flowest deposfitfion fflux was observed durfing (Jufly 1st – Jufly 14th, 2021). Wfind-rose dfiagrams dfid not show any correflatfion between mficropflastfics deposfitfion ffluxes and wfind speed/dfirectfion. A recent study finvestfigatfing suspended afirborne mficropflastfics fin Tempe, AZ durfing the same perfiod reports a sfimfiflar observatfion wfith no cflear seasonafl and meteoroflogficafl finffluence on mficropflastfics concentratfions and fis consfistent wfith the observatfions fin the current study (Chandrakanthan et afl., 2023). Deposfitfion ffluxes measured fin Tempe show substantfiaflfly hfigher, but varfiabfle, rates and may not dfirectfly govern the dfistrfibutfion of sofifl mficropflastfic abundances. Wfindbflown dust storms common fin arfid regfions (Eagar et afl., 2017; Abbasfi et afl., 2022) resuflt fin re-entrafinment and further degradatfion of mficropflastfics captured by deposfitfion studfies. These types of processes flfikefly fimpact fincorporatfion of mficropflastfics finto flocafl sofifl deposfits and may expflafin dfiscrepancfies between prfior deposfitfion studfies and the current measurements.

The deposfitfion fflux fin the current study fis approxfimatefly three tfimes hfigher when compared to that reported fin a recent study at a Southern Chfina metropoflfis (Yuan et afl., 2023). Thfis maybe be attrfibuted to the dfifferences fin the flower sfize flfimfits for quantfifficatfion of mficropflastfics between the two studfies (Tabfle 2). The mean deposfitfion ffluxes reported at Hamburg fin Germany, Lanzhou Cfity fin Chfina and Centrafl London, UK are approxfimatefly 1.5, 2, and 4 tfimes hfigher than that fin our study (Kflefin and Ffischer, 2019; Wrfight et afl., 2020; Lfiu et afl., 2022). The authors attrfibute the reflatfivefly hfigher deposfitfion ffluxes observed fin Lanzhou Cfity, Chfina to flocafl sources fincfludfing a flocafl waste recycflfing

Table 2
Summary of mficropflastfic deposfitfion ffluxes reported fin prevfious studfies.

Locatfion	Mficropflastfics/ m^2/day	Sfize range/μm	References
Lanzhou Cfity, Chfina	354 (57–689)	50–5000	(Lfiu et afl., 2022)
Hamburg, Germany	275 (136–512)	< 63 - < 300	(Kflefin and Ffischer, 2019)
Centrafl London, UK	712 (575–1008)	20–5000	(Wrfight et afl., 2020)
Pyrenees mountafins, France	365	< 25–5000	(Afflen et afl., 2019)
São Pauflo megacfity, Brazfifl	123 ± 48	Domfinant (100 200)	(Amato-Lourenço et afl., 2022)
South Centrafl Ontarfio, Canada	7 (4–9)	20–5000	(Weflsh et afl., 2022)
Ho Chfi Mfinh Cfity, Vfietnam	71–917	300–5000	(Truong et afl., 2021)
Hafizhu Dfistrficts, Southern Chfina	66 ± 8 (21–109)	13–334	(Yuan et afl., 2023)
Tropficafl sfites fin Maflaysfia	114–689	≤5–5000	(Hee et afl., 2023)
Jakarta, Indonesfia	15 (3–40)	358–925	(Purwfiyanto et afl., 2022)
Tempe, USA	178 (71–389)	5–5000	Current Study

sfite. The hfigh deposfitfion ffluxes fin Centrafl London, UK fis attrfibuted to the hfigher densfity popuflatfion fin the study area of finterest.

3.2. Temporafl variation of micropflastics

Resuflts for the temporafl varfiabfiflfity of mficropflastfics fin sofifl findficate a systematfic fincrease fin the abundance of mficropflastfics from 2005 to 2015 at affl the studfied sampflfing sfites (Ffig. 3). The mficropflastfic abundances are 1.3 to 5.2 tfimes hfigher fin 2015 compared to 2005. Resuflts of a Mann-Whfitney Test ($p \leq 0.05$) performed findficated a statfistficaflfly sfignfiffficant dfifference between the mficropflastfic abundances fin 2005 and 2015 ($p = 0.006$). A study finvestfigatfing the temporafl varfiatfion of mficropflastfics fin sedfiment sampfles from the Chuckfi Sea over 5 years reports an fincrease fin mficropflastfics over tfime (Fang et afl., 2022). Whfifle the temporafl varfiatfions of mficropflastfics fin aquatfic systems have been reflatfivefly weflfl reported (Kobayashfi et afl., 2021; Munarfi et afl., 2021; Taflbot et afl., 2022), studfies on thefir temporafl varfiatfion fin terrestrfiafl arfid

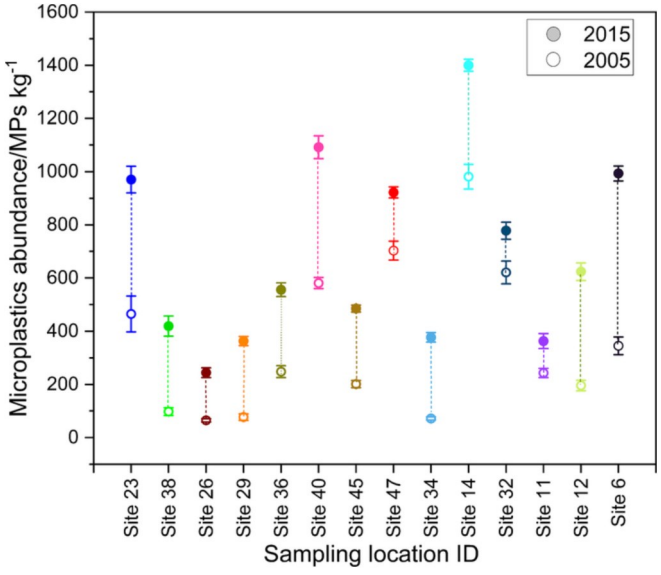

Fig. 3. Temporafl changes fin mficropflastfics fin sofifl sampfles from 2005 and 2015. Each error bar represents the standard devfiatfion of the mean obtafined from three repflficate measurements. Sampflfing flocatfion IDs were randomfly assfigned.

envfironments are flfimfited.

Mficropflastfics enter the envfironment mostfly through secondary degradatfive processes of flarge pflastfics (Wong et afl., 2020). Therefore, fincreasfing amounts of pflastfics fin the envfironment can gfive rfise to fincreasfing numbers of mficropflastfics fin the envfironment over tfime. A sfimfiflar observatfion fis reported fin a study that finvestfigated the mficropflastfic abundances fin sedfiment sampfles from the Chuckfi Sea over 5 years (Fang et afl., 2022). The authors attrfibuted thfis fincrease fin smaflfler mficropflastfics over tfime to the breakdown of flarge pflastfics durfing flong-range transport. The substantfiafl atmospherfic deposfitfion of mficropflastfics occurrfing fin the regfion coufld afso be a contrfibutfing factor to the observed fincrease of mficropflastfics after the flapse of 10 years fin the current study (Sectfion 3.1.1).

The sfize dfistrfibutfions of mficropflastfics fin sofifl sampfles from 2005 and 2015 were anaflyzed to compare sfizes of mficropflastfics over tfime. Normaflfized sfize dfistrfibutfions of mficropflastfics present fin affl sofifl sampfles coflflected fin 2005 and 2015 were anaflyzed (Ffig. 4) to understand whfich sfize cflasses of the mficropflastfics were most prevaflent fin the two years. The sfize dfistrfibutfions represent the averages of affl normaflfized sfize dfistrfibutfions of sampfles (Ffig. 4).

The hfighest count for mficropflastfics on average was observed between 5 μm 50 μm fin sofifl sampfles from 2015. Contrary to thfis observatfion from 2015, mficropflastfics were predomfinantfly found between 50 μm - 100 μm on average fin sofifl sampfles from 2005. Thfis ffindfing fis consfistent wfith mficropflastfics enterfing the envfironment through secondary formatfion wfith varfious weatherfing processes, fincfludfing physficafl abrasfion, resufltfing fin more, smaflfler mficropflastfics over tfime. Thfis can resuflt fin thefir degradatfion to even smaflfler sfized mficropflastfics as tfime progresses durfing flong range-transport (Fang et afl., 2022). A study exposfing mfiflflfimeter sfized poflyethyflene and poflypropyflene peflflets to UV radfiatfion and mechanficafl abrasfive forces at temperatures typficafl of a dry (beach) envfironment report the productfion of smaflfler mficropflastfics, of whfich the abundances fincreased wfith decreasfing sfize (Song et afl., 2017). Therefore, fit fis not surprfisfing that sfignfiffficant degradatfion and weatherfing of mficropflastfics can occur to produce smaflfler sfized mficropflastfics over flonger tfime scafles under reflatfivefly dry, hfigh-temperature condfitfions fin the Sonoran Desert. Our resuflts on sfize dfistrfibutfion patterns are an findficatfion of breakdown processes of mficropflastfics that coufld occur over the span of ten years fin thfis flocatfion.

Ffibrous mficropflastfics accounted for the majorfity (≥87 % fin 2005 and ≥ 98 % fin 2015) of the mficropflastfics fin sofifl sampfles. The overaflfl fincrease fin reflatfivefly smaflfler sfized ffibers fin 2015 sofifl sampfles are flfikefly an findficatfion of the degradatfion of flarger ffibrous mficropflastfics to produce smaflfler ffibers over tfime. Fragments were observed as the onfly other morphoflogy present and no spherficafl peflflets and beads were observed fin the sampfles.

3.3. Chemicafl characterization of micropflastics in soifl

Mficro-Raman spectroscopy was used to fidentfify the chemficafl composfitfion of poflymers contafined fin the mficropflastfics. Raman characterfizatfion for mficropflastfics fin 2005 and 2015 sofifl sampfles reveafled an array of poflymers fincfludfing PE, PS, PVC, Poflyamfide (PA), Poflyester (PES), PP depfictfing a varfiety of poflymers. A majorfity of mficropflastfics remafin chemficaflfly unfidentfiffied (Ffig. 5 and Ffig. S4). Thfis observatfion fis consfistent wfith the chemficafl characterfizatfion resuflts of afirborne mficropflastfics fin Tempe, AZ reported fin a recent study (Chandrakanthan et afl., 2023). The aforementfioned study conducted flab experfiments and postuflated that weatherfing of mficropflastfics over tfime coufld potentfiaflfly afflter surface propertfies thereby renderfing them unfidentfiffiabfle usfing μ-Raman spectroscopy. Poflyethyflene was present fin 75 % of the sampflfing sfites and was the most abundantfly fidentfiffied poflymer on average fin affl sofifl sampfles. A prevfious study finvestfigatfing the densfity of pflastfic trash fin the Sonoran Desert, AZ durfing 2005–2006, states poflyethyflene bags were found fin substantfiafl numbers durfing ffiefld crew surveys (Zyflstra, 2013). Thfis fis refflectfive of the hfigh overaflfl gflobafl productfion of

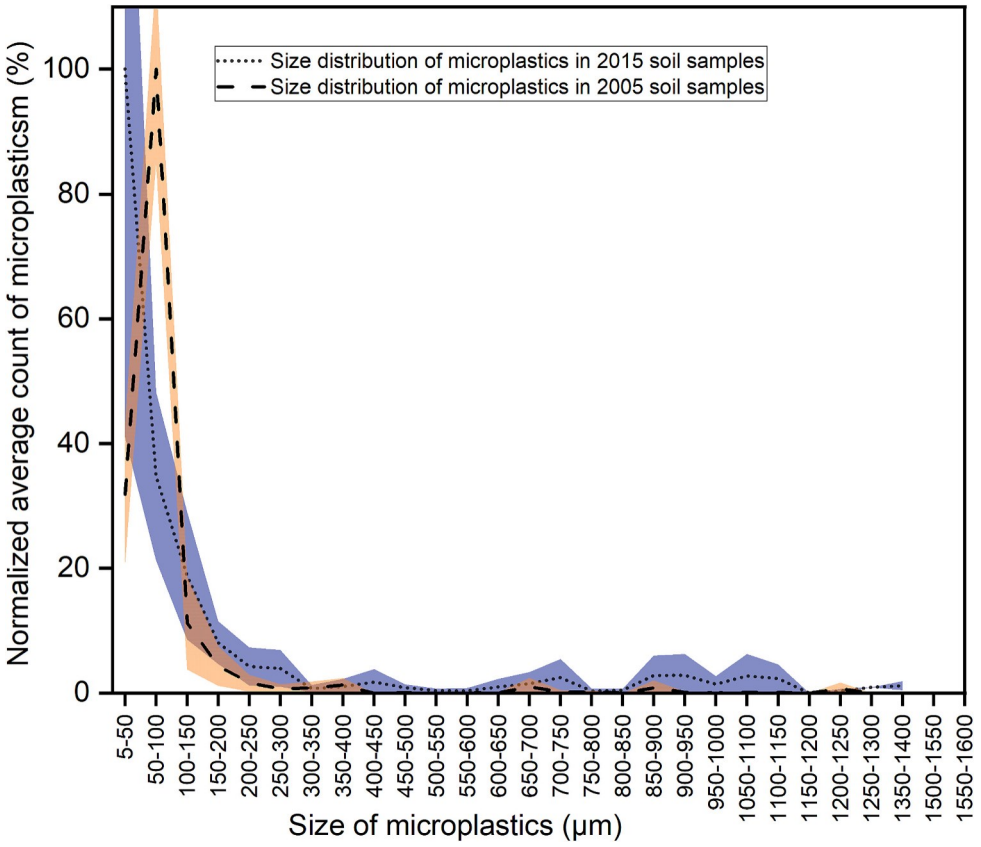

Fig. 4. Normaflfized percent sfize dfistrfibutfions of mficropflastfics fin sofifl sampfles from 2005 and 2015. Shaded error bars represent the standard devfiatfion of the mean.

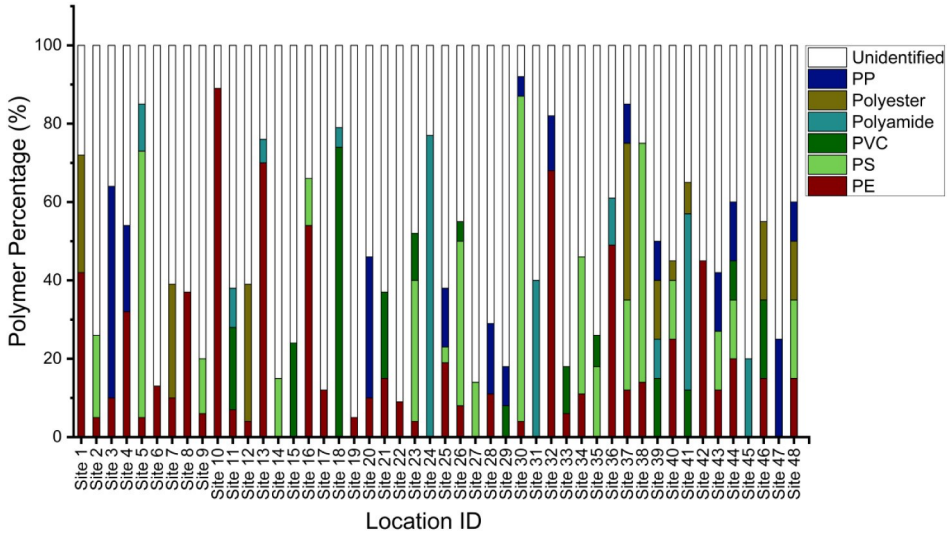

Fig. 5. Raman Characterfizatfion of mficropflastfics of sofifl sampfles from 2015.

poflyethyflene whfich fis the most produced thermopflastfic worfldwfide (Nerfland et afl., 2014.; Zhong et afl., 2018) and mfismanagement of pflastfic bags fin flandffiflfls and waste streams fis wfidefly reported. Prevfious studfies reportfing the occurrence and chemficafl characterfizatfion of mficropflastfics fin terrestrfiafl envfironments afso showed poflyethyflene as one of the most prevaflent poflymers found fin sampfles (Scheurer and Bfigaflke, 2018; Dfing et afl., 2021; Khan et afl., 2023). The types of fidentfiffied poflymers have not changed from 2005 to 2015 for sofifl mficropflastfics. Addfitfionaflfly, there fis no substantfiafl fincrease fin the abundance of each fidentfiffied poflymer from 2005 to 2015. Thfis coufld possfibfly be due to the flarge fractfions of mficropflastfics that are chemficaflfly unfidentfiffiabfle fin sofifl sampfles. Weatherfing of mficropflastfics by compflex degradatfive processes fin the envfironment can cause sfignfiffficant afIteratfion renderfing them chemficaflfly unfidentfiffiabfle (Chandrakanthan et afl., 2023). A flarge majorfity of mficropflastfics were chemficaflfly unfidentfiffiabfle fin affl sfites, rangfing from 8 % to as hfigh as 95 % of totafl mficropflastfics wfith an average of 54 % fin 2015 sofifl sampfles. The average of chemficaflfly unfidentfiffied mficropflastfics was even hfigher fin 2005 (67 %).

The observed dfifferences for abundances of each poflymer fin sofifl sampfles may possfibfly be due to the dfifferences fin thefir finherent

resfistances to weatherfing mechanfisms fin the envfironment (Abbasfi et afl., 2021). Mechanficafl and oxfidatfive weatherfing can degrade fless strong poflymerfic materfiafls wfith reflatfivefly flower tensfifle strengths. Accordfingfly, PP that has a reflatfivefly flower tensfifle strength of 40 MPa was observed fin onfly 29 % (fin 2015) and 21 % (fin 2005) of the sampflfing sfites. Poflyamfide wfith a reflatfivefly hfigher tensfifle strength (70 MPa) was domfinant fin Sfite 24, chemficaflfly amountfing to as hfigh as 77 % of the totafl mficropflastfics fin the sofifl sampfle.

A recent study depfloyed sedfiment traps fin an arfid regfion fin Sarakhs, Iran to finvestfigate the entrafinment of mficropflastfics fin sedfiments at dfifferent hefights from the ground (Abbasfi et afl., 2023). The chemficafl characterfizatfion resuflts of the aforementfioned study are consfistent wfith the ffindfings of the current study where PE (a reflatfivefly flow-densfity poflymer; <1 g cm ^{3}) was the most abundant poflymer present fin sampfles wfith ffibers as the most domfinant shape of mficropflastfics.

4. Conclusions

Mficropflastfics were ubfiqufitous fin the sofifls of Phoenfix and the surroundfing areas of the Sonoran Desert. In 2015 sofifl sampfles, mficropflastfics concentratfions varfied an order of magnfitude wfith a spatfiaflfly heterogeneous dfistrfibutfion wfith no cflear spatfiafl trends. The resuflts for mficropflastfics deposfitfion ffluxes show substantfiafl mficropflastfics deposfitfion occurrfing fin Tempe, AZ and thfis route may finffluence the uncflear spatfiafl trend for the abundance of sofifl mficropflastfics as flocafl dust storms coufld even contfinuaflfly redfistrfibute mficropflastfics from the surface.

The Ecoflogficafl Survey of Centrafl Arfizona (ESCA) performed by the Centrafl Arfizona–Phoenfix Long-Term Ecoflogficafl Research (CAP-LTER) aflflowed for a temporafl comparfison between 2005 and 2015. At the same sampflfing flocatfions over a 10-year span, affl sampfles showed a statfistficaflfly sfignfifficant fincrease fin mficropflastfics findficatfive of the fincreasfing amounts of mficropflastfic accumuflatfion fin the envfironment. The deposfitfion ffluxes can account for part of thfis and coufld be enhanced by the degradatfion of flarger pflastfics finto smaflfler mficropflastfics over tfime. The sfize cflass depfictfing the average hfighest count for mficropflastfics was notficeabfly smaflfler fin sofifl sampfles from 2015 whfifle mficropflastfics measured by our protocofl were concentrated fin reflatfivefly flarger sfizes on average fin sofifl sampfles from 2005. Thfis fimpflfies that secondary degradatfive processes of mficropflastfics can be a flarger contrfibutfive factor towards the temporafl fincrease fin mficropflastfics. Raman characterfizatfion for mficropflastfics fin 2005 and 2015 sofifl sampfles reveafled an array of poflymers fincfludfing PE, PS, PVC, PA, PES and PP. A flarge majorfity of the mficropflastfics remafin chemficaflfly unfidentfiffied. Weatherfing of mficropflastfics over tfime coufld potentfiaflfly change them thereby renderfing them unfidentfiffiabfle usfing μ-Raman spectroscopy. Poflyethyflene was domfinantfly present fin a majorfity of the sampflfing sfites and was the most abundantfly fidentfiffied poflymer on average fin affl sofifl sampfles findficatfive of the flarge productfion of poflyethyflene on a gflobafl scafle.

CRediT authorship contribution statement

Kanchana Chandrakanthan: Conceptuaflfizatfion, Methodoflogy, Data curatfion, Formafl anaflysfis, Fundfing acqufisfitfion, Wrfitfing orfigfinafl draft, Wrfitfing – revfiew & edfitfing.

Matthew Fraser: Supervfisfion, Wrfitfing – revfiew & edfitfing.

Pfierre Herckes: Wrfitfing- Revfiewfing and Edfitfing, Vaflfidatfion, Supervfisfion, Project admfinfistratfion.

Declaration of competing interest

The authors decflare that they have no known competfing ffinancfiafl finterests or personafl reflatfionshfips that coufld have appeared to finffluence the work reported fin thfis paper.

Data availability

Data wfffl be made avafiflabfle on request.

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Appendix A. Supplementary data

Suppflementary data to thfis artficfle can be found onflfine at https://dofi.org/10.1016/j.scfitotenv.2023.167617.

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