<u>Reply to reviewer</u>	
Manuscript Title	: Hyperspectral ultraviolet to shortwave infrared characteristics of marine-
	: harvested washed ashore and virgin plastics
Authors	: Garaba S.P. and Dierssen H. M.
Journal	: Earth System Science Data (ESSD)

#### Elizabeth C Atwood (Referee) Referee comment - 1

#### **General Comments**

The submitted data description paper contains important baseline measurements of both macro- and microplastics, the latter in both weathered, dry and wet, as well as virgin form. This is an important dataset for the field of remote sensing of marine plastics and the authors are highly applauded for making such a dataset public. We all thank you for making this available to everyone.

# Author response - 1

We appreciate your kind words and taking the time to carefully review our manuscript.

## Referee comment - 2

I still have some comments as to how this data should be presented, especially with respect to questions that remain open as to what these data represent.

# **Specific Comments**

Light has different penetration depths dependent on wavelength. This has been somewhat accounted for in the analysis in that wet microplastics were also measured, but a direct discussion of this fact is missing from the entire manuscript (I made note of this being missing at lines 20/21 in the Abstract and at the end of Section 3.2). This point should be addressed in more depth in the manuscript.

#### Author response - 2

Thank you for pointing this out. We have appended text to elucidate more on this (See section  $\underline{4}$  *Discussion* of the revised manuscript).

## Referee comment - 3

It would also help to make the above-mentioned point if in Fig. 4 the 30 ppt saltwater absorption curve over the same wavelengths range would be shown. Furthermore, the maximum penetration depth of light at each particular wavelength could be presented. This would aid in better representing the limitations of the dataset.

## Referee comment - 3

We have now included the absorption coefficient of pure water to further explain the wavebands affected by absorption of water (See *Figure 5* and *4 Discussion* of the revised manuscript).

## **Referee comment - 4**

There is no discussion of what different plastic types are represented in the microplastic samples and in what proportions. I noted this first at line 10 in Section 2.1, where the samples are being separated by size (which never comes up again in the following analyses, so that step is somewhat moot? Consider removing it). This is especially frustrating since you thereafter present the spectral curves for different virgin plastic types.

## Author response - 4

The text has been removed as suggested.

#### Referee comment - 5

The discussion of microplastic spectral curves (first paragraph Section 3.1) focuses only on different colors of the plastic measured, despite the fact that different polymer types play a very important role for spectral curve shape in the NIR-SWIR range. The plastic types of the macroplastics needs to be listed.

# Author response - 5

That is correct, polymer type plays a key factor in the spectral shape. Future sampling is expected to consider recording these additional critical descriptors: object color, shape, polymer composition, date of manufacture, dimensions. We now further acknowledge this limitation in the revised manuscript (See section <u>4 Discussion</u> of the revised manuscript).

# Referee comment - 6

I furthermore find it misleading to use 1 standard deviation in Fig. 3 and 4 to represent the variability in the data. The 1 s.d. curve is only truly representative of the data distribution if the data are normally distributed. In my experience, spectral measurements at any particular wavelength more often tend to be skewed in one direction or the other. Given this issue, I find the upper and lower percentage errors much more informative (one could still use the +/- 34.1% quantile around the mean to remain close to the 1 s.d. idea).

#### Author response - 6

We agree that it can be misleading to provide datasets with 1 standard deviation. We added text to make this caveat clear in the manuscript (See section <u>4 Discussion</u> of the revised manuscript).

## Referee comment - 7

In Fig. 4, it is confusing that the error curves for the different sampling curves overlap and look exactly the same (both exactly the same type of dashed line).

# Author response - 7

We improved the representation of the figure (See *Figure 5* of the revised manuscript).

## **Referee comment - 8**

Section 3.3 brings back the importance of my point above about proportion of different polymer types in the microplastic samples. Given the measured virgin polymer plastic curves, one could perform a spectral unmixing analysis of the dry and wet microplastic samples to determine proportion of different plastic types. One could then validate this separately by analyzing the microplastic samples down to plastic type. Maybe it doesn't make a difference to know what the different proportions of polymer types are in a sample, but I haven't come across a convincing study yet that makes this point.

## Author response - 8

A good point, the spectra from these virgin pellets can be used to expand the spectral reference library by performing unmixing with various combinations and proportions of the polymers. It is an area of interest to be further investigated (See section <u>4 Discussion</u> of the revised manuscript).

## **Technical Corrections**

No technical corrections were found needed.

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# Anonymous Referee #2

Received and published: 17 October 2019

# Referee comment - 1

This paper presents reflectance spectra for various types of plastics in the marine environment

(dry vs wet, macro and micro, washed ashore and marine collected) plus virgin plastic pellets. While spectral reflectance at an arbitrary viewing geometry and illumination environment is not a sufficient input for formal radiative transfer simulations as the authors suggest, these spectra do have value in that they can be used to identify spectral absorption features of potential value for remote sensing.

## Author response - 1

Thank you for taking the time to review and provide constructive feedback on our manuscript. We agree that investigations on the anisotropic distribution of the spectral reflectance of marine-harvested plastics is needed for more accurate simulations and hope future works will consider this (*See section <u>4 Discussion</u> of the revised manuscript*).

## We follow the review guidelines for the ESSD journal, indicated in italics.

Read the manuscript: Are the data and methods presented new? Is there any potential of the data being useful in the future? Are methods and materials described in sufficient detail? Are any references/citations to other data sets or articles missing or inappropriate?

# Referee comment - 2

Overall, the data and methods are new, useful, and presented in sufficient detail. A few terms could use a bit more explanation to make this paper useful as a stand alone product. For example:

1. the Spectral Shape Similarity is described in Garaba and Dierssen 2018, but I think this would be much more powerful if equation 1 from that paper was also included here.

# Author response -2

Thank you for pointing this out. We have appended the methods section by explaining the data processing steps used and appended Equation 1 on spectral shape similarity (*See section <u>2.4 Data processing</u> of the revised manuscript*).

# Referee comment -3

2. I feel that the sphericity and roundness scale from Powers 1953 is outdated and inappropriate for use if the goal is radiative transfer simulation a simple aspect ratio would suffice.

Table 1 has this, great, but was that table in the data files? Perhaps I missed it.

# Author response - 3

Yes, Table 1 was provided in the manuscript and as supplementary material

# Referee comment - 4

3. Derivative analysis needs to be defined. Again, this is in the Garaba and Dierssen 2018 paper, but definition is needed here. I find the description of what exactly constitutes a spectral 'feature' lacking (both here and the 2018 paper), and the listing of these features inconsistent in this paper. For example, the abstract lists four, apparently strong features. Section 3.1 shows eight. Qualitatively, I also question a few of the features – 2046 seems inconsistent, and 2313 seems too close to the edge of the spectral range to be valid. 1417 seems too close to other (water?) absorption features to be useful.

# Author response - 4

-We have included information about derivative analysis and provide a definition of a spectral feature and expand on the methodology used to objectively identify the spectral features reported (*See section* <u>2.5.1</u> <u>Spectra absorption feature</u> of the revised manuscript).

-A new figure to summarize the spectral data analyses has been included (*See <u>Figure 2</u> of the revised manuscript*).

-We have revised the text to clarify the mismatch in the number of absorption features in the abstract and in the results. In the abstract we have added the term 'diagnostic' which means an absorption feature is unique to a particular material in shape and is located around a limited wavelength range (*See Line 27 of the revised manuscript abstract, sections 2.5.1 Spectra absorption feature and 4 Discussion of the revised manuscript*).

-Yes, the 1417 nm feature is affected by atmospheric gases. The other features (centred around 2046, 2313 nm) have also been report in other studies. We have added text to point out the useful wavebands and studies that have already utilized several of the wavebands reported here (*See section <u>4 Discussion</u> of the revised manuscript*).

# Is the article itself appropriate to support the publication of a data set? Referee comment - 5

Yes, with the modifications noted above.

#### Author response -5

Thank you and we have carefully made revisions to improve clarity of the text.

# Check the data quality: Is the data set accessible via the given identifier? Is the data set complete? Are error estimates and sources of errors given (and discussed in the article)? Are the accuracy, calibration, processing, etc. state of the art? Are common standards used for comparison? Referee comment 6

The data do appear available as noted, although the link for the "dry washed ashore macroplastics" appears broken in the abstract (but not the link in section 4).

# <u>Author response -6</u>

The links have been checked and updated.

## Referee comment - 7

As far as I can tell, no uncertainty metrics were provided for the ASD observations, a key missing component. If this exists, it should be included.

## Author response - 7

We agree a comprehensive uncertainty budget is missing and is needed. However, the datasets are provided with the descriptive statistics mean, median and standard deviation. Each measurement was an average of 20 scans.

## Referee comment - 8

Although the paper notes differences in viewing geometry, foreoptic aperture and spectralon plaque reflectance for both the macro and micro plastics, no explanation for these different choices was given. This should be rectified.

## Author response - 8

We assume by doing a white reference with the Spectralon plaque after white referencing we obtained a Lambertian Equivalent Reflectance suggesting minimal effect by geometry. We have added text to clarify this (*See section <u>2.2 Spectral reflectance measurements</u> of the revised manuscript).* 

# Is the data set significant – unique, useful, and complete? <u>Referee comment - 9</u>

Consider article and data set: Are there any inconsistencies within these, implausible assertions or data, or noticeable problems which would suggest the data are erroneous (or worse). If possible, apply tests (e.g. statistics). Unusual formats or other circumstances which impede such tests in your discipline may raise suspicion.

#### Referee comment - 10

The data set, to the best of my ability to confirm, looks good.

**Is the data set itself of high quality?** <u>Referee comment - 11</u> Yes

Check the presentation quality: Is the data set usable in its current format and size? Are the formal metadata appropriate? Check the publication: Is the length of the article appropriate? Is the overall structure of the article well structured and clear? Is the language consistent and precise? Are mathematical formulae, symbols, abbreviations, and units correctly defined and used? Are figures and tables correct and of high quality?

Referee comment - 12

Yes to all of these

Is the data set publication, as submitted, of high quality? <u>Referee comment - 13</u> Yes

# Finally: By reading the article and downloading the data set, would you be able to understand and (re-)use the data set in the future?

# Referee comment -14

Yes, if the above issues are resolved. This could be used to help identify spectral absorption features for qualitative remote sensing algorithms, but not for input to radiative transfer simulations as the authors suggest. The latter requires spectrally resolved complex refractive indicies, measurements of size distribution and sphericity. Hopefully these data will be measured in the future.

#### Author response -14

Thank you and yes further measurements are warranted to support detailed radiative transfer models and suggest future works should consider such investigations *See section <u>4 Discussion</u> of the revised manuscript*).

Yes

Style Definition: Normal: Check spelling and grammar

# Hyperspectral ultraviolet to shortwave infrared characteristics of marine-harvested, washed ashore and virgin plastics

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#### Abstract

Combating the imminent environmental problems associated with plastic litter requires a synergy of monitoring strategies, clean-up efforts, policymaking and interdisciplinary scientific research. Lately, remote sensing technologies have been evolving into a complementary environmental monitoring approach that might have future applications in the operational 15 detection and tracking of plastic litter at repeated intervals covering wide geo-spatial areas. We therefore present a dataset of spectralLambertian-equivalent spectral reflectance measurements from the ultraviolet (UV, 350 nm) to shortwave infrared (SWIR, 2500 nm) of synthetic hydrocarbons (plastics). Spectral reflectance of wet and dry marine-harvested, washed ashore and virgin plastics was measured outdoors with a hyperspectral spectroradiometer. Samples were harvested from the major accumulation zones in the Atlantic and Pacific Ocean suggesting a near representation of plastic litter in global oceans. We 20 determined a representative bulk average spectral reflectance for the dry marine-harvested microplastics-and-identified common absorption features at ~931, 1215, 1417 and 1732 nm, dataset available at https://doi.org/doi:10.21232/jyxq-1m66 (Garaba and Dierssen, 2019a). Similar absorption features were identified in the dry samples of washed ashore plastics, dataset available at https://doi.org/doi:10.21232/ex5j-0z25 (Garaba and Dierssen, 2019b)-... The virgin pellets samples consisted of eleven polymer types typically found in floating aquatic plastic litter, dataset available at https://doi.org/doi:10.21232/C27H34 25 (Garaba and Dierssen, 2017). Magnitude and shape features of the spectral reflectance collected were also evaluated for two scenarios involving dry and wet marine-harvested microplastics, dataset available at https://doi.org/doi:10.21232/r7gg-yv83 (Garaba and Dierssen, 2019c). Reflectance of wet marine-harvested microplastics was noted to be lower in magnitude but had

similar spectral shape to the one of dry marine-harvested microplastics. <u>Diagnostic absorption features common in the marine-harvested microplastics and washed ashore plastics were identified at ~931, 1215, 1417 and 1732 nm.</u> In addition, we include
 metrics for a subset of marine-harvested microplastic particle morphology including sphericity and roundness. <u>These datasets are also expected to improve and expand the scientific evidence-based knowledge on optical characteristics of common plastics found in aquatic litter. Furthermore, tThese open-access-datasets have potential use in will be useful in radiative transfer
</u>

simulationsanalyses exploring the effect of plastics to ocean colour remote sensing and developing algorithms applicable to remote detection of floating plastic litter. The dataset is These datasets are also expected to improve and expand the scientific evidence based knowledge on optical characteristics of common plastics found in aquatic litter.

#### 5 1 Introduction

The amount of plastic litter in the natural environment is growing exponentially, and this challenge has led to a huge demand for integrated and sustainable monitoring strategies (Lebreton et al., 2018;Maximenko et al., 2016;G20, 2017;Werner et al., 2016). One of these widely considered tools that can provide a complementary avenue of wide geo-spatial and spectral information about plastics in natural waters has been remote sensing (Maximenko et al., 2016). Current key requirements

- 10 expected from remote sensing of floating plastics from an interdisciplinary perspective are 'detect, identify, quantify, track' capabilities. Feasibility studies centred on these four requirements have shown promising prospects in remote sensing of floating and submerged litter (Garaba et al., 2018;Aoyama, 2018;Kako et al., 2012;Topouzelis et al., 2019). Although current efforts are promising there is a need to advance remote sensing of plastics and adapt future sensors to generate plastic related end-product. In line with this, a new generation of satellite missions (e.g. PRISMA Italian Space Agency, EnMap German
- 15 Aerospace Centre, PACE National Aeronautics and Space Administration) is anticipated to improve-advance remote sensing of the environment through hyperspectral observations from the ultraviolet (UV, ~350 nm) to shortwave infrared (SWIR, ~2500 nm)(UV) to shortwave infrared spectrum (SWIR). Of course these future missions are not dedicated to plastic litter studies, they are likely to be an essential knowledge-base of high quality, hyperspectral, wide geo-spatial coverage information pertinent to plastics. Going forward most of the Ssatellite missions are going to be supported complemented by observations 20 from unmanned aerial systems, aircrafts and high altitude pseudo-satellites.

A limited number of high quality hyperspectral measurements of plastic types found in marine litter have been reported or are in open-access repositories. We therefore conducted measurements from the UV – SWIR with the goals to contribute towards (i) creating a high quality baseline hyperspectral reflectance dataset of weathered plastics being washed ashore or floating in

25 the oceans (ii) identifying absorption features of naturally weathered plastics, (iii) demonstrating the high reflectivity of plastics compared to other optically active constituents of the oceans, (iv) creating an open-access spectral reference library for improved radiative transfer simulations and (v) proposing algorithms essential to 'detect, identify, quantify, track' plastics. We present the detailed steps that were completed to acquire these measurements of the virgin types and naturally weathered plastics found in marine and land-based litter.

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#### 2 Methods and Materials

#### 2.1 Samples

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We used a set of specimens consisting of macroplastics (diameter > 5 mm) and microplastics (diameter < 5 mm). The macroplastics were collected during clean-up activities along the west coastline of the United States of America (USA), now being used to create awareness under the theme 'Washed Ashore: Art to Save the Sea', a travelling art exhibition in USA. At the time of experiment around midday on 25 March 2015 the exhibit was at the Mystic Aquarium in Connecticut. We believe these objects (buoys, handles, bottle caps, containers, styrofoam, ropes, toys, diving fins and nets) had undergone natural weathering at sea and/or on land based on careful visual inspection with particular interest on shapes, type of original object and colour. The macroplastics had different colour shades ranging from blue, green, yellow, orange, peach, beige, ivory to white.

Marine-harvested microplastic samples were obtained from the west North Atlantic ocean using a Neuston net (mesh size =  $335 \ \mu m$ ) in the top 0.25 m seawater layer (Law et al., 2010). After collection by with the nets, the microplastics were left to dry followed by separation by hand before storage in glass scintillation vials at Sea Education Association archives. In order

- 15 to explore the effect of particle size on the bulk spectral reflectance these microplastics were further grouped after successive filtration through metal sieves with mesh sizes ranging (i) 1.68 2.00 mm, (ii) 2.00 2.38 mm, (iii) 2.38 2.83 mm, (iv) 2.83 3.36 and (v) 3.36 4.00 mm. A very small amount of samples was left after the sieving and was not used for any further analysis. Additional marine-harvested microplastic specimens used in this study were collected from Kamilo Point in Hawaii, USA by Bill Robberson and Anna-Marie Cook (Environmental Protection Agency, USA). The Kamilo Point was not sieved
- 20 as the North Atlantic samples due to the quantity that was available, we therefore classified it as aggregated microplastics. Dry virgin pellets of varying opacity were chosen to represent the polymer source types that have been identified in specimens harvested from sediments and aquatic sampling (GESAMP, 2015;Hidalgo-Ruz et al., 2012). The polymer types considered were polyvinyl chloride (PVC), polyamide or nylon (PA 6.6 and PA 6), low-density polyethylene (LDPE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), fluorinated ethylene propylene teflon (FEP), terpolymer lustran
- 25 752 (ABS), Merlon, polymethyl methacrylate (PMMA).

The variability in <u>apparent</u> colour and shape of the marine-harvested microplastics and washed ashore macroplastics is a plausible representation of the plastic litter that is being found in the aquatic and terrestrial environment but may not necessarily represent all the plastic litter found globally.

#### 30 2.2 Spectral reflectance measurements

The spectral Lambertian-equivalent spectral reflectance (R) measurements of all specimens were conducted outdoors during daylight hours  $\pm 3$  hours around midday using an Analytical Spectral Devices (ASD) FieldSpec® 4 hyperspectral

spectroradiometer (Malvern Panalytical, USA) between 350 and 2500 nm. An 8.5° foreoptic was used during the spectral measurements of the dry macroplastics at a 45° nadir angle 10 cm above target object. A 99 % Spectralon® diffuseLambertian plaque (Labsphere, USA) was used for white referencing and optimizing the integration time of the spectroradiometer. It wais also assumed that by using a Spectralon® Lambertian plaque for white referencing we eliminated the effects of varying setup

- 5 geometry during measurements. Spectra were recorded first over the plaque then from 5 different spots of each dry macroplastic object and then over the plaque. A single spectral measurement was derived as an average of 20 continuous automated scans. Microplastics were aggregated on a black rubber mat to create an optically dense target before each spectral measurement (Figure 1). This black rubber mat was used as background target because it had a negligible spectral reflectance over the spectrum range observed. At a 0° nadir angle, a 1° foreoptic was placed 8 cm above the aggregated microplastics on
- 10 the black mat and reference measurements were made using a Spectralon® diffuseLambertian plaque (Labsphere, USA). Again here, a spectrum was first collected over the plaque followed by 10 measurements over the aggregated dry microplastics and then finally over the plaque. Before each measurement over the dry microplastics, we gently mixed the particles to rearrange the orientation and location of the particles in an effort to get a best representative bulk spectral signal. A similar approach was used to perform furtheradditional spectral measurements of wet microplastics in filtered seawater with a salinity of 30 ppt. Our selected setup was determined to be optimum and minimized instrument and user shading.



Figure 1 Experimental setup with the aggregated (A) dry and (B) wet marine harvested microplastics. A black neoprene rubber was used as a background in a dark spray painted container to mitigate background light during spectral reflectance measurements.

#### 2.5 Data processing

20 Natural outdoor lighting allowed us to measured spectral reflectance with good signal-to-noise from the UV to SWIR, with the exception of certain SWIR regions where the atmosphere is opaque. These regions of the spectrum are shown as gaps in the continuous spectrum, from 1350 nm to 1410 nm and 1800 nm to 1950 nm in the dataset. Average spectra was calculated for each set of repeated measurements. All data processing, statistics and plots were generated in MathWorks MATLAB. Similarity in spectral shapes was determined using the robust spectral contrast angle ( $\Theta$ ). Degree of spectral shape similarity was defined

#### 2.5.1 Spectra absorption features

- 5 In general, the spectral reflectance of an optically active object (e.g. plastic, corals, seawater) has a characteristic shape that explains how it can reflect or absorb light in the UV to SWIR. The spectral shape is a combination of peak (reflection) features and trough (absorption) features that are distinctive optical properties of the objects. An absorption feature would be the waveband at which the object absorb light more strongly or reflects less light compared to the neighbouring wavebands. Here, a-priori knowledge about typical absorption features in hydrocarbons or plastics was combined with visual inspection of
- 10 measured *R* (Figure 2a). Further verification of these identified absorption features was done by applying a moving average filter with a window of 19 nm to derive a smoothed *R*. Second order derivatives of the smoothed spectra were then computed to generate derivative *R* signals with enhanced absorption features (Figure 2b). Using derivatives of spectra has been shown to be a robust analytical tool in remote sensing (Dierssen et al., 2015;Huguenin and Jones, 1986;Russell et al., 2016;Tsai and Philpot, 1998). Continuum removal was applied to the *R* followed by calculating and a relative band depth index to enumerate
- 15 the absorption intensity. Anwas derived using the end and start wavebands was obtained from a MathWorks MATLAB R2016a convhull function. The function objectively locates the convex hull i.e. wavebands immediately before and after the absorption feature waveband (Figure 2c). Band depth indexes are widely used as proxies for detection and quantification of target optically active objects in natural environment (Clark, 1983;Clark, 1999). Absorption features are thus enhanced after being normalized by the continuum removal approach (Figure 2d). Diagnostic absorption features refer to those parts of the spectrum
- 20 unique to a particular object meaning they possess a similar shape and are located at a specific wavelength range they are located (Clark et al., 2003). An inter-comparison to check for diagnostic absorption features was conducted using the macroplastic and microplastic spectra.

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Figure 2. (a) Example spectral reflectance used for visual inspection to identify absorption features highlighted by the vertical lines, (b) second derivative signal validating the location of absorption features, (c) continuum line generated from the convhull function and (d) continuum removed signal.

- 5 The degree of spectral shape likeness among the measured *R* was calculated using a quantitative similarity scoring algorithm (Wan et al., 2002). A spectral contrast angle ranging from ( $0^\circ$  = very strong degree of similarity to  $90^\circ$  = no similarity) wais determined after converting the spectra of two samples into a multi-dimensional vector that is not affected by the inherent intensity of the spectra but only depends on the shape (**Equation 1**). Assuming x and y to be reflectance at a given wavelength of a sampled and reference target the spectral contrast angle ( $\Theta$ ) is derived as,
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$$\Theta = \cos^{-1} \frac{\Sigma x \cdot y}{\sqrt{\Sigma x^2 \Sigma y^2}}$$
(1)

<u>A scale to evaluate spectral shape similarity classified the results of Equation 1</u> as very strong  $(0^\circ \le 9 \le 5^\circ)$ , strong  $(5^\circ < \Theta \le 10^\circ)$ , moderate  $(10^\circ < \Theta \le 15^\circ)$ , weak  $(15^\circ < \Theta \le 20^\circ)$  and very weak  $(20^\circ < \Theta)$  (Garaba and Dierssen, 2018). Absorption features noted by visual inspection were validated by derivative analysis of the measured *R*.



Figure 1 Experimental setup with the aggregated (A) dry and (B) wet marine harvested microplastics. A black neoprene rubber was used as a background in a dark spray painted container to mitigate background light during spectral reflectance measurements.

#### 2.45 Microplastic Particleparticle morphology

5 A Marathon electronic digital calliper was used to measure the size distribution of dry marine-harvested microplastic particles. Additional particle descriptors included sphericity, roundness, and a qualitative description of colour. Particle sphericity and roundness were determined according to a qualitative scale (Powers, 1953).

#### 3 Results

#### 3.1 Macroplastics

- Spectral reflectance of the different dry washed ashore macroplastics (Garaba and Dierssen, 2019b) had significant differences in the visible spectrum related to the intrinsic colour of each object (Figure 23). Blue objects peaked around 450 nm, green objects around 550 nm whilst the white objects had a flatter reflectance in visible wavelengths. Beige and ivory coloured object had rapidly increasing reflectance in the visible, an eightfold reflectance magnitude rise was note from 400 nm to 700 nm. Yellow, peach and orange object also had increasing reflectance in the visible but not as pronounced as in the beige and ivory
- 15 objects, ranging from a three to fourfold increase in reflectance. Overall, the highest reflectance was noted on the beige object R = 0.88 around 850 nm. For all the objects, the reflectance peaked in the NIR followed by a general decrease in the SWIR with several absorption features resulting in localized dips and peaks. Despite the variations in the spectral magnitude and shape, absorption features common to all the macroplastics were observedlocated at wavebands centred close to 931 nm, 1045 nm, 1215 nm, 1417 nm, 1537 nm, 1732 nm, 2046 nm and 2313 nm (Figure 23). The location of the absorption features was
- 20 validated and confirmed by derivative analyses of each respective spectrum.



Figure 23. Spectral reflectance of dry washed ashore macroplastics harvested along the western coast of the USA. Absorption features noted in marine-harvested microplastics are highlighted by the vertical lines.

#### 3.2 Marine-harvested microplastics

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- 5 Spectral reflectance of the dry marine-harvested microplastics (Garaba and Dierssen, 2019a) increased with wavelength reaching highest values in the NIR at 850 nm then decrease towards the SWIR wavebands. All spectra were close to uniform in both spectral shape (mean Θ < 5°) and magnitude (percentage ranges < 40 %) compared to the macroplastics. A non-parametric Kruskal-Wallis one-way analysis of variance was utilized to determine if any differences existed in the measured spectra of the dry marine-harvested microplastics. The statistical test suggested no significant differences (*p* < 0.05) in the spectral reflectance from 350 to 2500 nm. We therefore determined a representative dry marine-harvested microplastic spectral</p>
- endmember (**Figure 34**). Absorption features identified in the dry washed ashore macroplastic specimens (**Figure 23**) were also found in the dry marine-harvested microplastics (**Figure 34**).





Figure <u>34</u>. Endmember spectral reflectance with 1 standard deviation continuous error bars of the dry marine-harvested microplastics. Identified unique absorption features are highlighted by the vertical lines.

5 Wet marine-harvested microplastics (Garaba and Dierssen, 2019c) had identical absorption features found in the dry marine-harvested specimens (**Figure 45**). However, the magnitude of *R* decreased by 12 % in the UV to 90 % in the SWIR due to the presence of water mixed with the samples. The loss of reflectance in the plastics wasis observed to be consistent with the increase in the absorption coefficient of pure water (**Figure 5b**). Average decrease in the measured R was  $56 \pm 23$  %.





Figure 4.5. (a) Mean bulk spectral reflectance of dry and wet marine-harvested microplastics with 1 standard deviation continuous dashed error bars. Absorption features noted in marine-harvested microplastics are highlighted by the vertical lines. (b) Absorption coefficient of pure water (Rottgers et al., 2011).

#### 3.3 Virgin pellets

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Spectral properties of the dry virgin pellets (Garaba and Dierssen, 2017) varied in magnitude and shape (Figure 56). However, two specimens of PA did show very strong similarities (Θ = 2.1°) although the apparent opacity of PA 6 was lower than that of PA 6.6. FEP was generally flat in the NIR to SWIR. Overall, the highest reflectance was observed in the specimens of ABS,
PMMA and PVC whilst the lowest was observed in PET. Only FEP and PVC were noted to have a strong reflectance in the SWIR > 1900 nm with *R* > 0.4. The absorption features that were identified in the marine-harvested and washed were also

found to be consistent with those in virgin pellets (Figure 56).

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#### 5 3.4 Morphology of dry marine-harvested microplastics

Morphometric measurements were completed on a total of 47 microplastic particles from different size classes (**Table 1**). The particles were brittle and could fracture with handling. Sphericity of the observed particles ranged from low to high sphericity whilst the roundness was between sub-angular to very angular (**Table 1**). Dry virgin pellets common ocean plastic litter had varying opacity of the colour white. **Table 1** is available as an excel sheet as supplementary material.

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Table 1. Microplastic particle side distribution and colour.

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Length (mm)	Width (mm)	Height (mm)	Colour	Sphericity	Roundness
7.72	3.25	2.44	green	high	very angular
9.96	5.59	1.94	white	low	very angular
9.34	3.81	2.36	white	low	very angular
3.94	4.56	1.54	white	low	very angular
7.17	4.57	1.50	green	high	very angular
5.65	3.76	2.16	white	low	very angular
3.97	3.43	3.14	white	low	subangularsub-angular
7.56	4.48	0.90	white	low	very angular
6.71	4.11	1.91	white	high	very angular
7.45	4.15	2.22	green	high	very angular
6.50	4.32	1.90	white	low	very angular
8.08	4.51	2.04	white	low	very angular
6.86	4.03	1.56	white	low	very angular
6.64	3.63	0.82	green	low	very angular
7.55	4.62	0.71	white	low	very angular
5.51	4.91	1.59	white	low	very angular
8.50	4.46	1.61	black	low	very angular
5.63	4.49	1.90	white	low	very angular
8.32	4.45	2.00	white	low	very angular
6.28	4.04	1.03	white	low	very angular
4.26	3.94	1.77	white	high	<del>subangualr<u>sub-angular</u></del>
5.55	4.94	1.40	white	high	very angular
5.04	3.79	0.90	white	high	very angular
5.89	4.43	0.91	white	high	very angular
6.47	4.57	1.75	white	low	very angular
13.07	3.82	0.42	white	low	very angular
4.50	4.29	1.35	white	low	sub angular
4.62	4.35	2.09	white	low	sub angular
6.57	4.69	1.37	white	low	very angular
17.21	4.58	0.36	white	low	very angular
7.77	5.32	0.60	white	low	very angular
5.19	4.28	1.26	white	high	very angular
3.85	2.71	2.62	white	high	very angular
6.14	4.41	0.85	white	high	very angular
10.54	3.66	0.35	white	low	very angular
5.59	4.25	1.62	white	high	very angular
8.29	4.66	1.01	white	low	very angular
10.16	4.15	0.89	white	low	very angular

15.08	4.38	0.25	white	low	very angular
7.33	4.34	0.68	white	high	very angular
6.30	4.00	0.61	white	high	very angular
8.52	5.77	0.42	white	high	very angular
7.17	5.64	1.23	white	high	very angular
4.93	4.28	1.09	white	high	<del>subangular<u>sub-angular</u></del>
6.35	5.34	0.77	white	low	very angular
6.68	3.67	0.62	white	low	very angular
7.64	4.46	0.65	white	high	very angular

#### **4 Discussion**

We measured Lambertian-equivalent spectral -reflectances of washed ashore, marine-harvested as well as virgin plastics and identified 8 absorption features (centred around ~931 nm, 1045 nm, 1215 nm, 1417 nm, 1537 nm, 1732 nm, 2046 nm, 2313
5 nm) in most of the weathered specimen. Location of these absorption features agreed well was consistent with prior reports

- (Asadzadeh and de Souza Filho, 2017;Hörig et al., 2001). Of these 8 wavebands, we concluded that ~931 nm, 1215 nm, 1417 nm, 1732 nm were diagnostic absorption features after continuum removal and derivative analyses. Several studies have already shown prospective applications of the ~1215 nm and ~1732 nm wavebands in detection and quantification algorithms for floating and land-based plastic litter (Garaba et al., 2018;Goddijn-Murphy and Dufaur, 2018;Kühn et al., 2004).
   10 Unfortunately, the 931 nm and the 1417 nm absorption features fall outside the atmospheric window. These features pose a challenge for algorithm development as the plastic specific signal will be scrambled in the signal from atmospheric gases
- especially water vapour around ~900 nm and ~1400 nm. We also simulated the potential detection of submerged plastics and observed a decrease in the measured *R* which was consistent with the enhanced absorption of light by pure water in the SWIR (Figure 5). One aspect that was not addressed with the dataset was the depth at which the submerged plastic can be detected,
   it is important to further study the optical properties of plastic litter with varying water depth.

The polymer characterization of the specimen is a key factor in advancing scientific evidence-based knowledge complemented by spectral measurements as it enables researchers to further create essential descriptors for remote sensing applications related to floating-plastic litters. Laboratory techniques are typically used to accurately determine polymer compositions of marine-

- 20 harvested or washed ashore plastics, these include Fourier Transform Infrared (FTIR), Raman spectroscopy and pyrolysis gas chromatography (Thevenon et al., 2014). However, in our case the washed ashore macroplastics were part of an ongoing travelling plastic litter awareness exhibit and no detailed further-analyseis could be conducted to determine polymer composition. The only descriptors obtained from the washed ashore macroplastics were the object color, shape/form and spectral reflectance. Future studiescampaigns are recommendedurged to collect additional high quality descriptors (polymer
- 25 composition, refractive index, date of manufacture, sphericity, individual size distribution) of plastic specimen that will

improve classification and radiative transfer modelling efforts. It is also important to consider the possibilities to expand the spectral reference library through spectral unmixing simulations to create blended polymers from the virgin pellets.

Variability in the spectra was reported within one standard deviation and a median was also determine to be consistent with

5 literature (Dierssen, 2019;Russell et al., 2016;Zibordi et al., 2011). However, future measurements should includeare urged to perform a comprehensive uncertainty budgets to enable advanced error propagation efforts when data is assimilated into radiative transfer models. Implementing the algorithms that use spectral shape and continuum removal approaches reduces the uncertainties related to variations in magnitude. These investigations should also explore the possible anisotropic optical properties of plastic litter especially as it breaks down and weathers in the natural environment.

#### 10 4 Data availability

Quality control was performed according to the guidelines of SeaDataNet. All the datasets are in open-access via the online repository EcoSIS spectral library <u>https://ecosis.org/</u>. The dry marine-harvested microplastics spectral data are available at <u>https://doi.org/doi:10.21232/jyxq-1m66</u> (Garaba and Dierssen, 2019a). Washed ashore plastics spectral data is available at <u>https://doi.org/doi:10.21232/ex5j-0z25</u> (Garaba and Dierssen, 2019b). The virgin pellets spectral data is available at <u>https://doi.org/doi:10.21232/C27H34</u> (Garaba and Dierssen, 2017). Dry and wet marine-harvested microplastics spectral

dataset available at https://doi.org/doi:10.21232/r7gg-yv83 (Garaba and Dierssen, 2019c).

#### 5 Conclusions and outlook

We have established an open-access dataset of hyperspectral reflectances of dry washed ashore macroplastics, dry marineharvested microplastics, artificially wetted marine-harvested microplastics and virgin pellets. The dataset provides some of the first <u>baseline</u> measurements that can be assimilated into radiative transfer modelling to improve scientific knowledge on the contribution of plastic litter to the bulk signal reaching remote sensing sensors. Furthermore, such knowledge about the hyperspectral characteristics of micro and macroplastics litter can be used to evaluate the capabilities and application of current multispectral sensors in remote sensing efforts. Using spectral response functions of current remote sensing tools (airborne,

25 unmanned aerial systems, high altitude pseudo-satellites, satellites) it is also possible to simulate the spectral signature of our dataset, this information would be crucial in the design of future or planned remote sensing tools.

#### Author contribution

SPG and HMD contributed equally to the experiment and manuscript preparation.

#### **Competing interests**

The authors declare that they have no conflict of interest.

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