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Literature Review Supplement**

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Executive summary

A literature review was undertaken during Highways England's Microplastics Phase 1 project to assess the potential contribution from the strategic road network (SRN) of microplastics pollution in the water environment. Part of the aim of this work was to direct a programme of monitoring. A second phase of research commenced in 2021 and this report sets out the findings of the supplementary literature review undertaken as part of Highways England's Microplastics Phase 2 project. The same methodology as used in the previous literature review was undertaken for Phase 2, with literature searches undertaken on four search engines and a process of sequential title and abstract filtering resulting in the final literature base. Additionally, recommended relevant literature was requested from the interviewees that were consulted in the previous literature review. Only key data or information and significant updates from the new literature gathered have been provided, and this supplement should be read in conjunction with the original literature review.

Although it is well established through research that the dominant source of microplastic pollution from roads is tyre and road wear particles (TRWP), quantifying the sources with standardised analytical techniques is still a challenge. Further analytical techniques have been reported on since the last literature review, including the use of a Laser Direct Infrared (LDIR) analyser (an infrared imaging microscope, which is faster than using the traditional Fourier Transform Infra-Red (FT-IR) spectrometers) and organic tyre constituents as markers in analysis, highlighting this as a leading-edge area of research. A growing body of research is also highlighting the most appropriate methods, and considerations to be taken, for sampling microplastics, as well as more studies which have sampled microplastics from the SRN in locations including drains, road dust and soils adjacent to roads. Some further analytical techniques which help to determine microplastics derived from the SRN, as opposed to other sources, have been reported on, including their higher density and the ability to analyse for paint fragments. These sampling and analytical developments, as well as the understood need for standardisation across research studies, will help to focus future research most appropriately when SRN-derived microplastics are the objective of the study.

The literature shows that information on the fate of microplastics when they enter the environment is limited, with further research into the role of rivers in transport and degradation of TRWPs needed. Additionally, there is a call for further detailed investigation into the various traffic and road factors which can influence microplastic pollution from roads. It is also recognised in the literature that the current techniques used for capturing and retaining sediment (e.g. stormwater ponds / SuDS basins) are suitable for TRWP associated microplastics, and should be retrofitted where possible, with regular maintenance for drainage systems implemented too.

It has been demonstrated that the impacts of microplastics on ecosystems and human health are not clear and further research is needed. Until then, using the principle of precaution it is suggested that mitigation measures should be applied.

1. Introduction

This report is part of Highways England's Microplastics Phase 2 project and follows on from the original literature review undertaken as part of Microplastics Phase 1.

The objective of this report is to identify any new and relevant publications, published since the original literature searches (January 2020) were undertaken, and to provide a supplement to the original literature review. This literature review supplement focuses on any key and significant updates which are relevant to the key findings from the previous literature review.

The research questions, which form the basis of both literature reviews, are shown below and further details on how the questions were identified are provided in the original literature review.

Primary question 1 (PQ1):

- To what extent does the strategic road network (SRN) contribute to microplastics in the water environment?

Secondary questions (PQ1):

- 1a. What are the main sources of microplastics on the SRN?
- 1b. What has changed in terms of the types and occurrence of predominantly pollutants in road runoff since 2010?
- 1c. What are the implications of the changes discussed in question 1b, for Highways England policy (most specifically related to microplastics)?
- 1d. What essential future research activities can be identified to better understand the contribution of road runoff to microplastic pollution and what would be their indicative budget?

Primary question 2 (PQ2):

- What is the most appropriate sampling and analysis method to quantify microplastics of key interest to the SRN?

Secondary questions (PQ2):

- 2a. Are robust and repeatable techniques under development for quantifying microplastics?
- 2b. What are key site characteristics and conditions for sampling microplastics on the SRN?
- 2c. To what extent can the SRN as a source of microplastics be differentiated from other (e.g. airborne) sources?

The following sections are provided within this report:

- Section 2 Approach – this summarises the approach taken and records the number of hits for the literature searches.
- Section 3 Findings – this focuses on any key and significant updates to the findings for both primary question 1 and primary question 2.
- Section 4 Conclusions – this summarises the findings.
- Section 5 References – this provides the references for the articles cited within this literature review.

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- Appendix A Bibliography – this provides the full list of literature which was read for the literature review.
- Appendix B Literature Search Results – provided as separate Excel files, these files show the hits from the literature searches and the filtering process.

2. Approach

2.1. Literature review supplement methodology

The approach to the literature review methodology is the same as that described in the previous report. The search terms were used in the four search engines (on 8th April 2021), with the search restricted for 2020 onwards, in order to capture any literature which has been published since the last search was undertaken (January 2020). The number of hits from these searches is shown in Table 2-1. Additionally, the four people interviewed as part of the original literature review were contacted to request any relevant published research they were aware of in the last year. This resulted in one additional article from Joanna Bradley (SDS Ltd), which is incorporated in Table 2-1 below. An article recommended by Richard Thompson (University of Plymouth) had already been picked up through the search engines.

Table 2-1 Number of hits from each search engine (undertaken 8th April 2021) plus additional recommended literature

	Web of Science	Science Direct	Scopus	Google Scholar	Recommended literature	Full Record (with duplicates removed)
Primary question 1	150	68	192 (capped at 150)	0	1	237
Primary question 2	155 (capped at 150)	65	154 (capped at 150)	0	1	232

2.2. Filtering, extracting and recording information

The same two-stage filtering process, checking and information extraction process, as that described in the previous report was undertaken on the identified literature. The literature in each category of the filtering process is shown in Table 2-2.

Table 2-2 Literature filtering

	Relevant (R)	Somewhat Relevant (SR)	Not Relevant (NR)	Number of articles read*
Primary question 1				
Stage 1: Title Filtering	37	83	117	
Stage 2: Abstract Filtering	40	N/A	80	37

	Relevant (R)	Somewhat Relevant (SR)	Not Relevant (NR)	Number of articles read*
Primary question 2				
Stage 1: Title Filtering	23	49	160	
Stage 2: Abstract Filtering	34	N/A	38	30

* Not all articles were accessible to download and some articles had been read previously as part of the original literature review (which was undertaken January 2020).

3. Findings

As this is a supplement to the original literature review, only key data or information and significant updates from the new literature reviewed are provided. This supplement should be read in conjunction with the original literature review.

3.1. Primary research question 1

- To what extent does the SRN contribute to microplastics in the water environment?

Secondary questions:

- 1a. What are the main sources of microplastics on the SRN?
- 1b. What has changed in terms of the types and occurrence of predominantly pollutants in road runoff since 2010?
- 1c. What are the implications, of the changes discussed in question 1b, for Highways England policy (most specifically related to microplastics)?
- 1d. What essential future research activities can be identified to better understand the contribution of road runoff to microplastic pollution and what would be their indicative budget?

3.1.1. Emerging research on road surfaces using plastic additives

Whilst undertaking this literature review update, from the list of the latest search results, it was clear many of the articles returned were related to the increased use and experimentation of adding plastics to road asphalt/surface mixes. It appears the driving force behind this area of research is twofold; to reduce consumption of 'new materials' (especially those sourced from fossil fuels, e.g. bitumen) in road mixes by repurposing plastic waste; and secondly to improve the performance of the road surface by increasing its longevity of performance (e.g. reduce tendency of rutting, better withstand heavy loads and deformation due to changes in temperature).

This issue of using plastic additives to road surface mixtures was (briefly) mentioned in the 'Construction and maintenance' section (pages 23 and 24) of the previous literature review report. An article was cited referring to the increased performance of road surfaces with plastic additives along with two articles which utilised recycled materials in road construction / resurfacing. It was noted, from one study in Norway (Vogelsang et al., 2019), road surfaces containing polymer modified bitumens (PMBs, which contain a plastic polymer substance called styrene butadiene styrene (SBS)) were speculatively estimated to contribute <1% of total road based microplastic emissions. However, the monitoring of known plastic additives to road surface materials (both markings and the surface itself), is advocated as a potential way of estimating potential degradation rates of these materials (Kitahara & Nakata, 2020). Undertaking such research is likely

to improve the knowledge base of the sources (type and quantification) of microplastics in road dust and stormwater.

At this point in time the authors have not established how much of Highways England's SRN is currently paved with road surfaces which use plastic additives. Therefore, to avoid diversion from the scope related to primary question 1 (To what extent does the SRN contribute to microplastics in the water environment?) it was concluded that papers which relate to the use or experimentation of plastic additives to road surface materials would be rejected. This is due to the subject matter being out with the scope of the original primary question. However, given the breadth and volume of papers related to this area of research, we did feel this topic was worthy of comment and it would be prudent to bring this to the attention of Highways England as a potential area for future research.

Our assumption is the majority of Highways England's current SRN road surfaces are currently composed of 'traditional' asphalt/bitumen mixes with a negligible number of cases where plastic additives are utilised. Subsequently, we would conclude that at this moment the road surfaces of the SRN would not be a significant source of microplastics to the water environment. However, the signposts from the literature review update undertaken, identify that new techniques and technologies are emerging (at an increasing rate), demonstrate the performance benefits of using plastic additives in road mixes. These new techniques could include the use of recycled tyres in road surface technology. Therefore, we would assume in the future, as more road surfaces containing plastics are installed, road surfaces themselves may become a more prevalent source of microplastic pollution sourced from the SRN.

It is estimated the breadth of literature on this topic would benefit from a literature review in its own right. Therefore, although plastic additives is outside the scope of this particular literature review supplement, it is recommended Highways England conduct an independent review of the latest road surface technologies research (perhaps specifically the use of recycled tyres) to fully assess/consider the impacts it may have for related environmental impacts of the SRN in the future.

3.1.2. Confirmation of the main sources of microplastics from roads

It has been noted there are seven major sources of microplastics entering rivers; wastewater treatment plants (WWTPs); combined sewer overflows (CSOs); on-site wastewater treatment systems; transport systems; agriculture; industrial sources; and diffuse litter (Odgen and Everard, 2020). Related to transport systems, road drainage is identified throughout the literature reviewed as being a major source of microplastic pollution. Research of microplastic pollution from terrestrial sources is still in its infancy, however it is evident that the SRN is potentially a major source of microplastic pollution, providing definitive input from TRWP but also road markings and road dust (Jarlskog et al. 2020; Moruzzi et al 2020; Wang et al., 2020; Xu et al., 2020).

There is an acknowledgement that TRWP represent the dominant source of microplastic pollution in road / stormwater runoff (Overdahl et al., 2021) and answers secondary question 1a. However, the variables or factors influencing concentrations and abundance of TRWPs (and other microplastic sources from roads) such as traffic speed, braking, vehicle type etc. is gradually being better understood, although the current knowledge is not as definitive as the sources. This is likely to be due to the fact that there are so many differing variables influencing these sources. Concentrations of TRWPs have been positively correlated to traffic volume (specifically Annual Average Daily Traffic values), traffic density and population density / urbanisation (Bondelind et al 2020; Grossman et al., 2021 Jarlskog et al., 2021; Mengistu et al 2021; Su et al., 2020). Furthermore, high levels of braking and acceleration are also reported as being influential variables producing both larger sizes and a greater number of TRWPs (Knight et al., 2020; Mengistu et al., 2021). As these variables are better understood, it is not surprising studies have reported increasing distance from TRWP sources and a reduction in traffic volume leads to rapid declines in concentrations measured. This indicates TRWPs maybe have a low potential to be transported over long distances (Grossmann et al., 2021). Seasonality effects has also been reported in Gothenburg Sweden, with more TRWPs generated during the summer compared to winter months (Jarlskog et al., 2020; Jarlskog et al., 2021). This effect may be related to the fact TRWPs tend to

accumulate during periods of dry weather and then can be transported in higher concentrations during storm events (Su et al., 2020). Evidence of this process is demonstrated in one study where rainfall intensity was a statistically significant predictor of microplastic concentration in urban stormwater runoff (Smyth et al., 2021). There are limited UK based studies into variables affecting the abundance of microplastics in road runoff.

Table 3: Example of variables influencing rates of microplastic pollution sourced from road networks

Examples of variables influencing rates of microplastic pollution from road networks (the variables listed are not exhaustive or arranged in order of importance)	
Traffic speed	Road type
Traffic volume	Climate
Traffic density / population density / urbanisation	Antecedent rainfall / rainfall intensity
Vehicle type (e.g., passenger vehicles vs. HGVs)	Rates of Littering and degradation rates of litter/ TRWPs containing macro / microplastics
Breaking and acceleration rates (e.g., cornering vs. straight and traffic hotspots vs. free-flowing areas)	Effectiveness of drainage systems (e.g., gully pots, SuDS basins) at retaining microplastic runoff

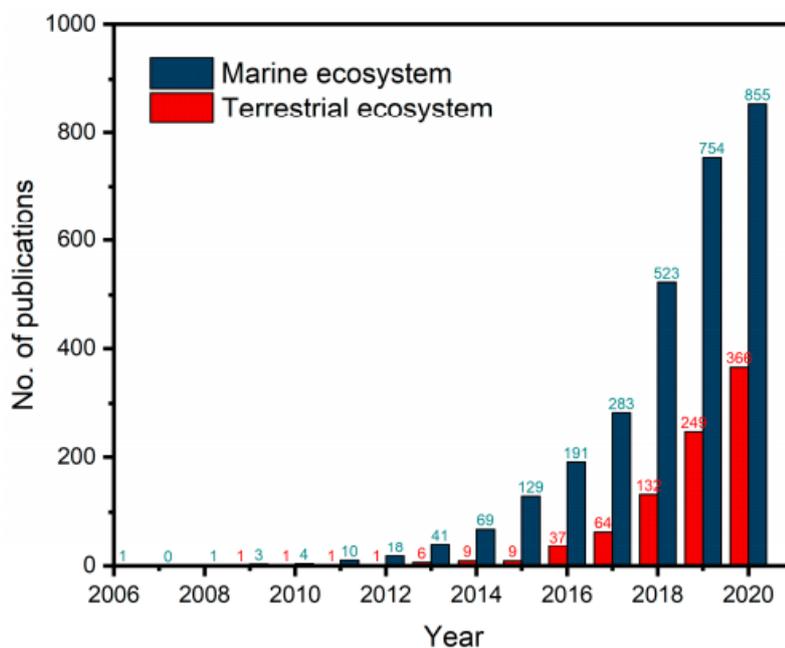
TRWPs generated from roads are subject to change (via degradation, e.g. UV radiation), processes leading to these changes (both susceptibility to breakdown and an estimate of how many times one particle might sub-divide) must be better understood so emissions of TRWPs can be better categorised and assessed (Bondelind et al., 2020; Klockner et al., 2020). It has been noted there are potentially large amounts of TRWPs in fraction of microplastics measuring <20µm. Therefore, developing analytical methods to measure these are necessary to aid understanding of the scale of TRWP sourced microplastic pollution (Jarlskog et al., 2021).

During this latest review, it was evident more articles were available examining the link between road dust and microplastic pollution. Consequently, road dust is being attributed as a major source of microplastics in both urban and rural environments (Narmadha et al., 2020). The microplastic content of road dust predominately consists of microplastics sourced from TRWP, road surface materials and markings (Kitahara & Nakata, 2020; Su et al., 2020; Wang et al., 2020). This implies microplastics sourced from road networks, can either be transported to the water environment directly, i.e., via runoff or indirectly through atmospheric deposition, with road dust being identified as a microplastic source in urban stormwater runoff (Pramanik et al., 2020). Not surprisingly, given its composition, the abundance of microplastics in road dust have been found to be significantly correlated with the same variables linked to TRWP concentrations in stormwater runoff, vehicle traffic density and population (Grossman et al., 2021; Kitahara & Nakata, 2020).

Although diffuse litter is mentioned in the literature as a major source of microplastics (Odgen and Everard, 2020), no literature was found in this latest review which specifically quantified the influence of litter contributing to microplastics fluxes sourced from roads (or even in comparison to input from TRWPs). This is a topic which would likely benefit from increased research so the influence of litter as a source can be better understood.

As an aside, one paper in the latest literature review investigated the contribution of road salt spreading to microplastic pollution from roads. The study investigated road locations in Norway, Sweden and Denmark and concluded microplastics in road salt are a negligible source (average >0.008%) of microplastics from roads compared to other sources, e.g. TRWPs (Rodland et al., 2020).

Figure 1: Figure showing number of papers published associated with microplastic pollution from 2006 to 2020 (taken from He et al., 2020b)



All literature presented above represents the general growth and interest in microplastic research over the last 10 years (since 2010). Figure 1 (taken from He et al., 2020b), shows the growth in published research since 2010 has had an exponential like trend. In relation to secondary question 1b, it has been clearly established microplastics sourced from roads (and within road runoff) is a significant and prevalent form of pollution which is receiving ever growing attention. In turn, the knowledge associated with the extent and quantification of the pollution is also set to increase in the coming years.

3.1.3. Quantitative values associated with microplastic pollution

As much as the sources of microplastic pollution are becoming more established, a recurring theme in the literature is the need for standardised methods of measurement as well as definitive unit of measure so results from different studies sites (mainly TRWPs sourced from roads) can be compared and the influence of specific variables explored (Lee et al., 2020; Miller et al., 2021). From the latest literature review, there is a noticeable shift from research focused on the sources of microplastic pollution to studies more concerned with producing quantitative results to further investigate the controlling variables affecting microplastic pollution. As secondary question 1a has been largely answered, one of the key questions which is arising from 1d would be can we better quantify microplastic pollution from roads and better understand the variables that control the rates of pollution. Numerous methods of measurement currently exist, including density separation and/or filtration, yet there is not a consensus on the most effective. For identifying TRWPs it seems using density separation is the preferred method, but the range of densities used needs better resolution (Miller et al., 2021). Further details on quantification of microplastics, and the methodologies used, are provided in response to Primary Question 2a.

From the latest literature reviewed some studies quantified aspects of microplastic pollution sourced from road environments. Hopefully this research including the variables influencing the microplastics can help contextualise any future results and data Highways England research projects produce related microplastic pollution. In Canada, TRWPs made up 22% of particles in stormwater and microplastic pollution in general was positively correlated to total road length (Grbic et al., 2020). In South Korea, it is estimated that 53,188 tonnes of TRWPs are produced each year with detailed results indicating that areas with larger traffic flows of heavier vehicles (e.g. trucks and buses) tend to produce more TRWPs (Lee et al., 2020).

In Europe, quantitative studies within the latest literature review were found in France, Germany and Sweden. In studies of Paris sub-catchments, plastic debris concentrations in runoff water ranged between 7 and 134 mg/m³ and when extrapolated to the Greater Paris area, the estimated amount of plastic debris discarded into the environment through untreated stormwater (with road drainage forming a major component of this) ranged from 8 to 33 tonnes per year (Treilles et al., 2021). In Germany, the highest concentration of TRWPs in road runoff (water samples) ranged from 5 to 92 mg/g (dry weight) and in corresponding soil samples from 0.2 to about 160 mg/g (dry weight) (Baensch-Baltruschat et al., 2021). The highest concentrations of microplastics in samples, analysed as part of the study, were in tunnel dust (max. 204 mg/g) as well as sediments of a treatment basin for road runoff registering a maximum of 150 mg/g (Baensch-Baltruschat et al., 2021). A study from Sweden utilised hydraulic modelling to indicate that microplastic concentrations were correlated with higher average daily traffic volumes and that microplastics with densities close to 1.0 g/cm³ did not readily settle out in sediment and were more likely to be transported further downstream from where they were produced (Bondelind et al., 2020).

3.1.4. Effectiveness of ponds and basins to treat microplastic pollution

Although standardised quantification of microplastic plastic pollution from road networks remains a challenge, the latest literature review indicated there is a larger volume of research being undertaken to better understand how stormwater ponds and basins help treat microplastics. This area of research (and following contents of this section) should be viewed in relation to secondary question 1c, regarding potential changes needed to Highways England policy to control microplastic pollution from roads.

Stormwater (detention/retention) basins have potential to act as a preliminary barrier to halt spread of microplastics into the wider environment, with the added potential benefit of improving ecological conditions downstream (Grbic et al., 2020; Mengistu et al., 2021; Moruzzi et al., 2020; Smyth et al., 2020). Where microplastics in stormwater ponds have been studied, they have been shown to contain high concentrations of microplastics indicating ponds can act as an important sink for microplastics sourced from road network (Hu et al., 2020).

Bioretention cells / biofiltration systems located near or alongside road networks can also effectively trap and store microplastics (Grbic et al., 2020). Biofiltration systems have been indicated to retain the vast majority of microplastics (mostly around roots systems) (Kuoppamaki et al., 2021). Other studies have quantified the retention of microplastic pollution within biofiltration systems to be ~80-90%, with the most effective retention for larger particles (>100 µm), therefore demonstrating the potential of these systems to be used within road runoff management (Liu et al., 2020; Smyth et al., 2021). Furthermore, finer sediments seem to have a greater potential to retain more microplastics than coarser sediments (He et al., 2020a) indicating systems which contain elements of clay based soils/filter mediums may be more efficient than those will coarser sediments (He et al., 2020a).

It is recognised knowledge gaps exist in the efficiency of different types of stormwater / road runoff basins (e.g., wet/dry ponds, wetlands, bioretention cells) (Shruti et al., 2021). More research is needed to evaluate effects of different media type and depth on efficiency rates as well as confirm optimal conditions for removal (Moruzzi et al 2020; Smyth et al., 2021). Additionally, establishing the most efficient maintenance routines requires extra attention, e.g., how often the systems should be emptied to avoid clogging, so the effectiveness of installed systems remain high rather than potentially causing/creating an acute microplastic pollution (Mengistu et al., 2021; Smyth et al., 2021).

Long term monitoring is needed on a range of spatial and temporal scales (e.g. high frequency storm water sampling to daily scales) to better understand the nature of microplastic transport through these types of systems (Mengistu et al., 2021). Targeted sampling both before and after rainfall events is seen as important, as the role of storm events are thought to be key drivers of the process between basins acting as source or sinks (Hitchcock, 2020; Shruti et al., 2021). Microplastic concentrations increased (as much as 40 times) after storm events, with rainfall providing the best measure to explain variation in microplastic concentrations. This shows that targeted high frequency sampling during storm events can help better understand transport of

microplastics (Hitchcock, 2020). The transport processes affecting microplastics are said to be similar to those of total suspended solids (TSS) therefore, it has been proposed TSS modelling can be used to predict microplastic concentration and retention in bioretention cells. (Smyth et al., 2021).

Another strategy identified to limit the spread of microplastic pollution (as well as metals and polycyclic aromatic hydrocarbon (PAHs) from roads is street/road sweeping (Jarlskog et al., 2021). Street/road sweeping may be an effective way of reducing microplastic build up on roads by collecting and disposing of the material before it enters stormwater systems and potentially transported to other downstream environments (Jarlskog et al., 2020). Generally, the development of knowledge in the source and transportation of microplastics will help better inform the design of treatment features and management strategies. This is particularly relevant in known hotspots of microplastic pollution so the distribution of microplastic pollution (from roads) to the wider environment can be minimised (Koutnik et al., 2021; Shruti et al., 2021). Once again, it has been noted that standardising methods of analysis and harmonising quantification of results are needed so multiple studies can be compared more and common results validated (Shruti et al., 2021).

As the literature demonstrates, the infrastructure needed to control microplastic pollution from road runoff might already largely be in place (in form of basins, ponds and wetlands). Therefore, in relation to 1c, changes needed in policy might be subtle. However, a future research activity (secondary question 1d) might be, can the benefits of (new and already constructed) stormwater basins be better quantified?

3.1.5. Ecotoxicology

As microplastic abundance is more thoroughly investigated and understood, being able to contextualise and develop ecotoxicity levels is gathering more attention. Advancing knowledge on this specific aspect of microplastic pollution it is therefore important to consider the full extent and context of microplastic pollution from the SRN. This area of literature relates to secondary question 1c in terms of the topic of ecotoxicology from road sourced microplastic potentially being more prevalent within Highways England policies and advancing knowledge in this area is a topic of consideration related to 1d.

Within the applicable literature there is widespread agreement, given the apparent abundance of microplastics in the environment more research is needed (Knight et al., 2020; Narmadha et al., 2020; Wang et al., 2020; Xu et al., 2020). However, it is noted research is needed not only into specific ecological risk / prescribed safe limits, but also from the perspective of providing recommendations to improve management and control of plastic pollution to the wider environment (Schell et al., 2020). Only one study in the latest literature review quoted a derived toxicological threshold value, which was a No-Observed Adverse Effect Concentration (NOAEC) of 112 $\mu\text{g}/\text{m}^3$ for respirable TRWP (Baensch-Baltruschat et al., 2020). In the same study, the results of the corresponding risk assessment indicate that the potential risk for human health concerning cardiopulmonary effects, from TRWPs, is low (Baensch-Baltruschat et al., 2020).

3.2. Primary research question 2

- What is the most appropriate sampling and analysis method to quantify microplastics of key interest to the SRN?

Secondary questions:

- 2a. Are robust and repeatable techniques under development for quantifying microplastics?
- 2b. What are key site characteristics and conditions for sampling microplastics on the SRN?
- 2c. To what extent can the SRN as a source of microplastics be differentiated from other (e.g. airborne) sources?

3.2.1. Techniques for quantifying microplastics

Reference was made in the original literature review to the development of a standard relating to microplastics and their sampling and analysis being developed by Defra with the British Standards Institute (BSI) and the International Organisation for Standardisation (ISO). Whilst this is not available yet, Rauert et al., (2021) assessed two technical specifications from ISO (ISO/TS 21396:2017(E) and ISO/TS 20593:2017) which provide methodologies for quantifying TRWP in sediment/soil and air through pyrolysis gas chromatography mass spectroscopy (Pyr-GC-MS). The methodology uses 4-vinylcyclohexene (VCH) (a dimer of the markers styrene butadiene rubber (SBR) and butadiene rubber (BR) (from the pyrolysis of natural rubber (NR) and synthetic rubber (SR)). The methodology assumes: these markers are only formed from the tyre tread; the mass of rubber in tyres is constant; the total rubber content is 50% of the mass of the tyre tread; and, passenger tyres contain 44% SBR+BR whilst truck tyres contain 45% NR. Tyre treads from Australia and Norway were sampled and tested using this methodology with the study indicating: in all tyre samples, the calculated SBR+BR content was below the assumed 44% in passenger tyres (<0.05% to 28%), potentially leading to an under-reporting TRWP concentrations; and, there was variability between, and within, different brands of tyres. This indicates the need to investigate further the tyre compositions of manufacturers and to further develop any standardised methodologies to ensure their robustness.

Density separation

As recorded in the original literature review, different solutions of varying densities are used to separate microplastics from the rest of the sample matrix. Although most common plastics are <1.4 g/cm³, it was noted by Horton et al., (2017) previously that dense composites of road-marking paints, aggregates, paint coating on dense particles or high-density mineral-polymer mixtures did not float during density separation in their study. As cited within Miller et al., (2021) previous reports have shown that the density of TRWP can reach as high as 2.2 g/cm³ (Kayhanian et al., 2012), indicating the need to carefully select the density solution when undertaking analysis of SRN-derived microplastic samples.

Source characterisation

The original literature summarised the main analytical techniques to characterise microplastic particles as: electron microscopy, spectroscopy (including markers), thermoanalytical and melting tests. New updates on these analytical techniques from those previously discussed, as found within the new literature review, are provided below.

A particular use of the melt test, to identify plastic particles was used by Beriot et al., (2020). Soils sampled in Spain for microplastics were processed and then were photographed under a stereo microscope, before and after heating on a hot plate to 130°C for 10 seconds. Microplastic particles were identified based on, among other things, their visual response to heat between the before and after photos.

Scircle et al., (2020), whilst assessing microplastics pollution in the Mississippi Gulf, America, used a Laser Direct Infrared (LDIR) analyser, in which is believed to be the first major use of LDIR in the analysis of microplastics in natural waters. The LDIR is an infrared imaging microscope, which is faster than using the traditional Fourier Transform Infra-Red (FT-IR) spectroscopes. It locates particles within its analysis area, collects information to describe its size, area and shape, obtains a spectra and conducts a library search to provide a match, providing both counts and identification.

Following FT-IR analysis of sediments from gully pots in Norway, Mengistu et al., (2021) used recorded wave numbers and temperatures from pyrolysis in a Parallel Factor Analysis (PARAFAC) model to identify and quantify (in concentrations) rubber materials (i.e. those constituents of tyre particles) within the samples. This was the first study to use PARAFAC to identify tyre wear particles in environmental samples.

Steinmetz et al., (2020) developed a new simple and fast method for quantifying the three most environmentally relevant polymers polyethylene (PE), polypropylene (PP), and polystyrene (PS) in soil. The method used 1,2,4-trichlorobenzene (TCB) for extraction, thereby needing limited preparation, followed by Pyr-GC-MS.

Markers which are indicative of TRWP within samples, in particular Zn, were identified in the previous literature review. Knight et al., (2020), Kloeckner et al., (2020) and Baensch-Baltruschat et al., (2020) all used organic tyre constituents as markers in analysis. Knight et al., (2020) whilst sampling storm drains in the UK, used N-cyclohexyl-2-benzothiazolamine (NCBA) as a marker for tyre wear in GC-MS analysis of sediment samples. Although they did note that the use of chemical markers can result in underestimation of tyre wear particles due to their degradation in the environment, and conversely overestimation can occur if there are additional sources of them within the environment. Kloeckner et al., (2020) concluded that OHBT (2-hydroxybenzothiazole) and ABT (2-aminobenzothiazole) were promising marker compounds for TRWP quantification. Baensch-Baltruschat et al., (2020) in a review of TRWP identified the use of 2-(4-morpholinyl)benzothiazole (24MoBT) and N-cyclohexyl-2-benzothiazolamine (NCBA) as markers and concluded they are less reliable for quantifying TRWP than methods involving thermo-analytics. Grossman et al., (2021) analysed car and truck tyre treads for rubber indicator molecules and concluded that DMVCH (2,4-dimethyl-4-vinylcyclohexene) and SB (cyclohexenylbenzene) were appropriate markers, which could then be used to quantify car and truck tyre wear in environmental samples.

Using their experience of San Francisco Bay, USA in a variety of environmental media, Miller et al., (2021) provided recommendations on best practice for collecting, analysing and reporting microplastics. In terms of analysing and reporting, the key points were:

- Visual identification – generally manual identification is undertaken and is labour intensive and can result in mistakes. Whilst alternative automated methods (FT-IR/Raman mapping or particle tracking with simultaneous Raman spectroscopy) are beginning to be used, the instruments are expensive, and they do not work well on samples which contain high numbers of particles.
- Composition – larger particles (greater than 250 µm) are best analysed with FT-IR spectroscopy, whilst smaller particles are best analysed with Raman spectroscopy. There are limited spectral reference libraries, and additionally weathered plastics have different spectra than virgin plastic, indicating the need for further development of spectral libraries specific to microplastics.
- Reporting – results reporting need to be standardised (for counts as well as mass) and the use of standardised terminology for morphological categories and material categories needs to be adopted, in order to aid useful comparison between studies.

3.2.2. Site characteristics and conditions for sampling microplastics on the SRN

There have been several additional published articles identified during the latest literature review which specifically look at sampling and analysing for microplastics from urban/road runoff, dust and roadside soils.

Knight et al., (2020) undertook a study in Plymouth, UK looking to quantify and understand the distribution of tyre wear particles, as well as identifying the road conditions that influence their generation and subsequent release. Samples were taken from the wet sediment (1 litre) in storm drain sumps (where solids are designed to be captured at the bottom of roadside drains), representing a range of traffic densities (high/low) and levels of braking and accelerating (high/low) (all four combinations of the variables). Additionally, samples from the local environment

comprising sediment from local estuaries (6 samples), soil (4 samples) and road dust (1 sample) were collected, which are adjacent to a major highway in Plymouth. Sampling was undertaken intermittently over a 2-year period, which may have caused seasonal changes to influence their results. They note that it was likely that a proportion of tyre wear particles will have been washed down the drain, and therefore samples from the storm drain sumps will not represent all tyre wear particles. The study concluded that high traffic braking and acceleration was the most influential variable in producing more, and larger, tyre wear particles >50 µm. There was also variability in terms of particle abundance between traffic densities.

Kloeckner et al., (2020) sampled road dust (from road sweepers), sediment from a sedimentation basin and an open settling pond treating highway runoff (bed sediment sampled using a beaker) as 'hotspots' of road runoff. Sediment from a local lake, as an environment considered to have low TRWP concentrations was also sampled for comparison (sampled via dredging), near Leipzig Germany to determine TRWP in these samples. From the differences in TRWP sizes between road dust and the sedimentation basin, they concluded that TRWP of smaller sizes are transported preferentially, and conversely coarser TRWP tend to be retained closer to the road environments. However, once the coarser TRWP disintegrate with age, their transport into the aquatic environment would be facilitated.

Mengistu et al., (2021) sampled the wet sediment (500 ml) from the bottom of gully pots at five sites (four streets and one parking lot, two gully pots at each site) in Norway. The maximum speed limit at each site was 30 km/h and traffic density at the sites was recorded using radar. The characteristics of the sites, in terms of surrounding land use, was also recorded to characterise each of the sites. The results showed that gully pots can act as temporary sinks for TRWP and could be an acute pollution risk for downstream watercourses if the gully pots are not properly maintained. Additionally, the samples with the highest TRWP concentrations were found at the sites with higher traffic density and higher braking/acceleration intensity. However, the full influence of traffic conditions on TRWP in gully pots could only be determined if a study is undertaken whereby all gully pot emptying frequency, TRWP generation and mobilisation factors are controlled.

Jarlskog et al., (2021) collected stormwater, road dust and washwater/sweep sand from a street sweeper for microplastic determination around a reconstruction site in Gothenburg, Sweden. Stormwater was sampled during six events during which street sweeping was carried out weekly, and three events without weekly street sweeping. The samples were collected from the point to which all the stormwater drained from the catchment is diverted before discharging into the river. Sweep sand and washwater were collected shortly after street sweeping, seven times in the autumn when weekly street sweeping was being undertaken. Road dust was collected directly from the road after street sweeping. The study concluded that traffic was an important source of TRWP, with larger amounts of TRWP produced during the summer than the winter. Street sweeping was found to be an effective mechanism to reduce TRWP on the road surface, and thereby preventing them from reaching stormwater.

Su et al., (2020) collected roadside dust from two catchments in Victoria, Australia with differing land use and urbanisation patterns. Sixteen sampling sites across the catchments were sampled in October (Spring) and December (Summer) 2018 to determine any seasonal rainfall effects. Three replicates of 50g of dust were collected in the drainage line receiving runoff from the road. They note that roadside dust is composed of a diverse mixture of material, including atmospheric deposits. Microplastics were found in the roadside dusts sampled, with statistical analysis indicating that urbanisation and rainfall influence microplastic accumulation. The study concluded that monitoring microplastics in roadside dusts can be a valuable tool for the initial screening of pollution in urban areas.

Choi et al., (2020) sampled soil from four different land uses (forest, traffic, residential and agriculture) within a city in South Korea in order to determine if different land uses (including 'traffic') affected microplastics within the soil. Surface soil (0-5 cm depth) within a semi-randomly placed 0.5 x 0.5 m quadrat was sampled in triplicate using an auger. The study concluded that the microplastics found differed between the different anthropogenic activities. High abundances of microplastics were found in the soils surrounding the traffic areas, however urban sources of plastics were surpassed by those which were agriculturally-derived.

Baensch-Baltruschat et al., (2020) in a review of TRWP talk about the paths of TRWP emissions into the environment, which have implications for sampling for microplastics from the SRN.

- **Runoff – TRWP** are washed off during rainfall events. A precipitation rate of at least 2 mm/day is required to mobilise TRWP, according to Unice et al., (2019). Although high rainfall and runoff intensities do not lead to complete wash-off of TRWP, at the higher precipitation rate of 5 mm/day the main share of the TRWPs will be moved from the road surface. Where artificial drainage is not present to capture the runoff, this runoff (incorporating TRWP) will infiltrate directly adjacent to the paved road, within a small zone with a width of 0.75–1.5 m according to Blok (2005) and Kocher et al. (2010).
- **Transport/treatment** – As mentioned above, if there is no artificial drainage present, TRWP is mainly deposited in the soil close to the road. If artificial drainage is present, there is often treatment present, although this can depend upon traffic densities. Within the drainage system, it is expected that the coarser fractions of TRWP are retained in sewer inlets, gully pots and channels while the finer fractions are transported through the entire sewage system. Only a small proportion (or none at all) are expected to reach surface waters through drainage systems, that includes treatment or transport to a wastewater treatment plant.
- **Frequency and representativity of sampling** – Long-term sampling captures natural variability and allows statistical analysis. Within rivers, there is very little information known on sedimentation and degradation in relation to TRWP loads. Lack of sunlight in deeper waters may hamper degradation, and therefore sedimentation may be the major sink of TRWP.

Baensch-Baltruschat et al., (2020) also presented a table (Table 3, not included here) of recent (2000 onwards) studies on the occurrence of TRWP in the environment. This includes the environmental compartments of road dust, surface water, sediments and sludge from road runoff treatment systems.

Miller et al., (2021) provided recommendations on best practice for collecting, analysing and reporting microplastics in different environmental media based on their experience in San Francisco Bay, USA. In terms of collecting samples, although not specifically SRN-related, they are still relevant for consideration when sampling for microplastics, the key points and considerations were:

- **Site selection** – Microplastics analysis is expensive, so it is necessary to consider how to make the best out of a small number of samples. Consider the geography, hydrology and hydrodynamics and bathymetry of the water body, and potential sources (e.g. outfalls). Consider the desired spatial and temporal coverage to be evaluated. Including less urbanised/contaminated or reference sites that can provide context for the degree of contamination.
- **Surface water** – Consider the hydrodynamics and particle transport and where microplastics will be transported. The depth of water from which the sample will be taken and whether this is a grab sample or a net. Seasonality of sampling - wet vs dry season and the runoff.
- **Sediment** – Consider whether you sample in a depositional or erosional area. The depth of sediment sampled affects results, with surface sediments are more representative of recent conditions.
- **Streams/stormwater runoff** - Consider the land use surrounding the watercourse selection and its influence on samples. Influence of seasonality and storm-related transport on loads. Consider setting a storm threshold level for sampling a wet weather event prior to starting a study – this should be set so conditions are sufficiently intense to mobilise small particles from the watershed. Capture the first-flush at the start of a runoff event. Depth-integrated samples

should be collected for accurate loads estimation, these also pick up denser materials which may not be nearer the surface.

Shruti et al., (2021) reviewed the methods for sampling, analysing and identifying microplastics in stormwater. There were a range of methods used for sampling water and sediment, which are summarised in Table 2 in the article (not included here). The article concluded that a standardised approach is required and provides a suggested methodology for: plan objectives; sample collection; sample treatment; and, characterisation (Figure 4 in Shruti et al., 2021). The suggested sample collection issues to be considered for stormwater collection are:

- Rainfall event.
- Capacity and yearly discharge of treatment facility.
- Duration of the stay in stormwater catchment areas.
- Distance from shoreline.
- Depth.

3.2.3. Differentiating the SRN as a source of microplastics

Additional techniques and methods were identified in the latest literature review which can further help identify which microplastics are derived from the SRN.

Kovochich et al., (2021) undertook a lab-based study in order to characterise TRWP for single particle analysis, in order to allow identification of particles which are TRWP-derived. Key findings which help to determine that particles are derived from the SRN in complex media are:

- Particles having a density between 1.0 and 2.2 g/cm³.
- Visually the particles are elongated/round shape with variable amounts of mineral encrustation.
- Compositionally, the surface characteristics include co-localisation of (S + Zn/Na) ± (Si, K, Mg, Ca, and Al), and the co-localisation of organic surface characteristics, such as C₆H₅⁺ and C₇H₇⁺ during the scanning electron microscopy (SEM) coupled with energy dispersive X-ray spectroscopy (EDX) mapping and time-of-flight secondary ion mass spectrometry (ToF-SIMS) analysis.

Klockner et al., (2021) undertook a lab-based study analysing the metal content of various plastic items, including tyre tread rubber, to investigate possible metal fingerprints. Statistical analysis found that only tyre tread was found to have a polymer specific distinct metal composition of Zn, and lower deviations in metal composition compared to other plastic items. The authors did note however that the tyre tread rubber samples used in this study were used passenger car samples and truck tyres may have a different composition. The use of organic tyre constituents as markers, to determine microplastics are SRN-derived has also been shown by Knight et al., (2020), Klockner et al., (2020) and Baensch-Baltruschat et al., (2020).

Choi et al., (2020) when analysing microplastics in soils from different land uses in South Korea identified that visually the microplastics identified in the 'traffic' soils were dominated by black and yellow colours, which they attributed to be primarily due to tyre dust and road paint.

Gaylarde et al., (2021) undertook a review of paint fragments as microplastics within the ocean. Although the focus is mainly on paint fragments from boats, they also highlight road marking paints as an urban source. In terms of methodologies for identification and analysis, they note that it has been suggested that for more accurate identification of coloured microplastics (of which road marking paints are) both FT-IR and Raman spectroscopy should be used. However, Raman spectroscopy is considerably more time intensive than FT-IR, and therefore they suggest that Raman should be used only for particles of 1–50 µm, above which size FT-IR can be relied on as a

rapid method for the identification. A newer analysis, optical photothermal infrared spectroscopy, has recently become available which offers submicron spatial resolution with the possibility to target specific particles with both IR and Raman spectroscopies.

4. Conclusion

A summary of this literature review supplement, in relation to the research questions, is provided below.

Primary question 1 (PQ1):

- To what extent does the strategic road network (SRN) contribute to microplastics in the water environment?

Secondary questions (PQ1):

- 1a. What are the main sources of microplastics on the SRN?
 - TRWPs particles have been established as the dominant type of microplastics sourced from road networks (but it is also acknowledged these have been the particles most researched) (Baensch-Baltruschat et al., 2020).
 - Microplastics from road marking and the road surface itself are also acknowledged as main sources as is diffuse litter.
- 1b. What has changed in terms of the types and occurrence of predominantly pollutants in road runoff since 2010?
 - It has been clearly established microplastics sourced from roads (and within road runoff) is a significant and prevalent form of pollution which is receiving ever growing attention. In turn, the knowledge associated with the extent and quantification of the pollution is also set to increase in the coming years
- 1c. What are the implications of the changes discussed in question 1b, for Highways England policy (most specifically related to microplastics)?
 - Current techniques involved in capturing and retaining sediment (and heavy metals) are suitable for microplastics (associated with TRWP) retention (Baensch-Baltruschat et al., 2021). Where these systems do not exist, efforts should be made for them to be retrofitted. Implementing such systems should slow distribution of microplastic pollution within wider environment. Additionally, implementing regular maintenance programs for road drainage systems is a crucial element of addressing microplastic pollution to ensure collection of microplastics is efficient and the material is removed (causing minimal pollution downstream) and disposed of responsibly (Baensch-Baltruschat et al., 2020).
 - Although outside the scope of this literature it is recommended Highways England investigate the implications of using plastics additives, including recycled tyres, in road surface mixes / technologies to understand the impacts this may have on microplastic pollution sourced from the SRN.
- 1d. What essential future research activities can be identified to better understand the contribution of road runoff to microplastic pollution and what would be their indicative budget?
 - The principal challenge now is to better quantify the inputs from specific sources and this is still hampered by challenges associated with standardisation of analytical techniques (Baensch-Baltruschat et al., 2020).
 - The influence of the following variables, contributing to microplastic pollution from roads still warrant more detailed investigation: road type (high traffic versus low traffic), road surface

material, type of vehicles (HGV versus passenger cars), traffic conditions (high speed travel versus increased intermitted breaking), composition of tyre material (optimisation of reducing wear without comprising safety).

- Specifically related to estimates of traffic derived microplastic concentrations in soil, only two studies (Dierkes et al., 2019 and Chen et al., 2020) are known globally to have been published (Buks & Kaupenjohann, 2020), highlighting the need for more quantifiable research in this area.
- Similarly, current research is limited on the fate of microplastics when they enter the environment, in particular freshwater systems (both natural and man-made, e.g., stormwater systems). More detailed insight is needed regarding the role of rivers in the transport and degradation of TRWPs and the specific environmental conditions which control these processes (Baensch-Baltruschat et al., 2020; Baensch-Baltruschat et al., 2021; Grossman et al., 2021).
- Regarding ecotoxicity, the impacts on ecosystems and human health are not clear and more research is needed to investigate the issue and mitigation measures should be implemented according the principle of precaution (Baensch-Baltruschat et al., 2020).

Primary question 2 (PQ2):

- What is the most appropriate sampling and analysis method to quantify microplastics of key interest to the SRN?

Secondary questions (PQ2):

- 2a. Are robust and repeatable techniques under development for quantifying microplastics?
 - Although standardised techniques are being developed, there is still a need to ensure the appropriateness and repeatability of these (Rauert et al., 2021).
 - Analysis of microplastics within environmental samples is still a leading-edge area of research, which is highlighted by the newly captured methods for undertaking analysis of microplastics in environmental samples which have been published since the last literature review (Section 3.2.1). This has included the first reported use of an LDIR analyser in natural waters (Scircle et al., 2020) and organic tyre constituents as markers in analysis (Baensch-Baltruschat et al, 2020; Kloeckner et al., 2020; Knight et al., 2020).
 - Further research in this area is needed to find standardised analytical techniques which can be undertaken by many researchers, something which was noted by Miller.et al., (2021) from their extensive experience of analysing for microplastics in San Francisco Bay.
 - Consideration of the new techniques identified in this literature review supplement can help to inform the sampling and analysis tasks of this project.
- 2b. What are key site characteristics and conditions for sampling microplastics on the SRN?
 - There is a growing body of research on both the most appropriate methods, and considerations to be undertaken, for sampling microplastics from the SRN, including greater understanding of the TRWP pathways into the environment (Baensch-Baltruschat et al., 2020). This includes consideration of:
 - Site selection – spatial/temporal coverage, reference sites, geography/hydrology of the site.
 - Surface water/sediment sampling – particle transport, depositional/erosional area, depth of water/sediment.
 - Streams/stormwater runoff – land use, seasonality, storm threshold for sampling, first-flush.

More studies have also been undertaken to specifically sample microplastics derived from the SRN, both in roadside drains, road dust, runoff treatment basins and soil adjacent to roads. Consideration of the approaches used in these studies can help to inform the sampling and analysis tasks of this project.

- 2c. To what extent can the SRN as a source of microplastics be differentiated from other (e.g. airborne) sources?
 - Further specific analytical techniques which help to identify those microplastics which have been derived from the SRN have been reported on. These include the density of TRWPs (2.2 g/cm³, Kayhanian et al., 2012), visual identification of yellow and black particles, and a method for paint fragment analysis.
 - These developments, as well as the understood need for standardisation, will help to focus future research most appropriately when SRN-derived microplastics are the objective of the study.
 - Consideration of the analytical techniques identified in this literature review supplement to differentiate the SRN as a source of microplastics can help to inform the sampling and analysis tasks of this project.

Considering the next steps for this (Phase 2) project, it would be recommended that the techniques for sampling and analysing microplastics from the SRN that have been identified within the literature review and literature review supplement are reviewed and considered in the context of the sampling and analysis tasks which are currently being planned. In addition, it is recommended that Highways England conduct an independent review of the latest road surface technologies (specifically those using ground tyre rubber in asphalt) including understanding whether microplastics are considered within the certification process.

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Microplastics Phase 2 –

Literature Review Supplement

Technical Services Framework 2 (SPaTS 2)

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Appendix A. Bibliography

The bibliography provides the full list of literature which was read for the literature review. If information from the literature was used in the review, then they are also provided in the reference list (Section 5).

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Appendix B. Literature Search Results

These are provided as two separate Excel files.

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